



# DEVELOPMENT OF BIO-BASED PARTICLEBOARDS FOR BUILDINGS' INDOOR USE

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## **Development of bio-based particleboards for buildings' indoor use**

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## ABSTRACT

The environmental problems are more and more serious, and solutions to moderate climate change are urgently required. The construction sector is widely responsible for energy, water, and raw materials consumption and for pollution; thus, the use of low embodied energy and sustainable building products are efficient possibilities to overcome environmental challenges.

Nowadays the opportunity of enhancing and re-proposing natural materials for building practices is increasingly catching on. Wastes and by-products seem to be an efficient solution: bio-waste-based products with good quality and ease of use are more and more analysed and proposed, moderating the problem of solid waste disposal as well. Furthermore, they can contribute to passively control indoor conditions, guaranteeing both healthy places and user well-being, and lowering the energy demands of buildings.

Starting from this knowledge, this thesis aimed to produce sustainable and eco-efficient indoor particleboard reusing agro-industrial bio-wastes and bio-resources as aggregates with a significant contribution to improving indoor conditions. The bio-aggregates were bounded by non-toxic adhesives that can secure low environmental impact in the production process, being able to moderate bio-based materials' drawbacks.

The outcome was the development and assessment of bio-based particleboards for buildings' indoor applications with hazelnut shell, *Arundo donax* and sodium silicate. A horizontal analysis was carried out; hence several properties were considered; among others, bio-susceptibility, and performance in case of fire were investigated, as they are two of the main drawbacks of using bio-based building materials. Finally, the sustainability of the proposed boards was evaluated qualitatively by considering the Life Cycle Assessment.

**Keywords:** *Arundo donax*; Acoustic comfort; Bio-waste; Bio-resource; Bio-susceptibility; Fire safety; Hazelnut shell; Hygroscopicity; Hygrothermal comfort; Sodium silicate.



## RESUMO

Os problemas ambientais são cada vez mais graves e soluções para moderar as alterações climáticas são urgentemente necessárias. O sector da construção é responsável por grande consumo de energia, água e recursos naturais, e por poluição; por isso, a utilização de materiais de baixa energia incorporada e a produção de produtos de construção sustentáveis é uma possibilidade eficaz no contributo para resolver os desafios ambientais.

A possibilidade de utilizar materiais naturais na construção está a ganhar cada vez mais importância. A utilização de resíduos e subprodutos parece ser uma solução eficiente, uma vez que permite moderar o problema da sua gestão. Produtos eficientes à base de bio-resíduos estão a ser cada vez mais estudados e propostos. Além disso, podem contribuir para assegurar o controlo passivo das condições interiores, o que pode garantir locais mais saudáveis e o conforto dos utilizadores, e reduzir o consumo energético dos edifícios.

Tendo em conta estas considerações, a tese teve como objetivo produzir uma placa compósita eco-eficiente reutilizando resíduos agro-industriais e bio-recursos como bio-agregados, para melhorar as condições interiores dos edifícios. Os bio-agregados foram aglutinados por adesivos não tóxicos que pudessem garantir um baixo impacto ambiental no processo de produção e atenuar as vulnerabilidades dos bio-compósitos.

O resultado foi o desenvolvimento e caracterização de placas bio-compósitas com base em casca de avelã e *Arundo donax* com silicato de sódio para aplicações em interiores. Foi realizada uma análise horizontal e consideradas várias propriedades, entre elas a bio-suscetibilidade e a reação ao fogo, uma vez que são duas das recorrentes desvantagens da utilização de produtos de construção de base biológica. Finalmente, a sustentabilidade dos painéis propostos foi avaliada qualitativamente, considerando a Avaliação do Ciclo de Vida.

**Palavras chave:** *Arundo donax*, Bio-resíduo; Bio-recurso; Bio-suscetibilidade; Cascas de Avelã; Conforto higrotérmico; Higroscopicidade; Conforto acústico; Segurança ao fogo; Silicato de sódio.





## RIEPILOGO

I problemi ambientali sono ogni giorno più gravi e sono necessarie soluzioni efficaci per contrastare il cambiamento climatico. Il settore edile è uno dei maggiori responsabili a causa del grande fabbisogno di energia, dell'ampio uso di risorse naturali e della produzione di prodotti inquinanti. Per questo, la produzione e l'impiego di materiali da costruzione sostenibili rappresenta un'opzione efficace per fronteggiare le sfide ambientali.

I materiali naturali e i sottoprodotti delle attività umane sono sempre più presi in considerazione, poiché evitano l'uso e la produzione di sostanze nocive e dannose per l'ambiente e permettono di ridurre il problema dello smaltimento dei rifiuti. Inoltre, i prodotti da costruzione a base di rifiuti organici possono garantire il controllo passivo delle condizioni interne, la salubrità degli ambienti, il benessere degli utenti e la riduzione della domanda energetica.

Partendo da tali considerazioni, il progetto di dottorato aveva l'obiettivo di produrre un pannello truciolare sostenibile ed eco-efficiente capace di migliorare le condizioni di comfort interno. Scarti agroindustriali e bio-risorse sono stati considerati come aggregati; come leganti sono stati analizzati adesivi non tossici, capaci di moderare gli svantaggi derivanti dall'impiego di materiali a base bio e garantire un processo produttivo a basso impatto ambientale.

A fronte di una dettagliata ricerca bibliografica, sono state realizzate diverse campagne sperimentali, combinando teoria e pratica. Il risultato è stata la produzione di pannelli a base bio composti da gusci di nocciola, *Arundo donax* e silicato di sodio per applicazioni interne di cui sono state analizzate diverse proprietà (analisi orizzontale), tra cui la bio-suscettibilità e le prestazioni in caso di incendio, due dei maggiori inconvenienti derivanti dall'uso di questi materiali. La sostenibilità dei pannelli è stata, infine, analizzata qualitativamente per mezzo della valutazione del ciclo di vita.

**Parole Chiave:** *Arundo donax*, Bio-residuo; Bio-risorse; Bio-suscettibilità; Comfort acustico; Comfort igrometrico; Gusci di nocciole; Igroscopicità; Sicurezza in caso di incendio; Silicato di sodio.



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# INTRODUCTION

## 1.1 Context

The construction sector is one of the most polluting human activities, producing harmful residues and requiring a huge percentage of global water and resources (Dickson and Pavía, 2021; Martínez-García et al., 2020). It has been identified as the largest single energy consumer in Europe (Posani et al., 2021), using 40% of the world's energy and requiring a significant amount of resources (Asdrubali et al., 2015; European Commission, 2019). Considering the proposals of the Agenda 2030 (UNO, 2015) and the European Green Deal (European Union Council, 2019), which want to respond to the urgent need for more sustainable building practices, one of the most important challenges is guaranteeing climate-neutral activities.

Building materials are widely responsible: their production, transport, use, and end-of-life cause high environmental impact. Furthermore, they can influence the working principles of building, by affecting both energy demand and comfort, ensuring a passive control of indoor conditions and securing users' well-being and health (Boquera et al., 2021; Nguyen et al., 2017). The low performance of the buildings' envelopes significantly contributes to the environmental impact caused by the construction sector as well. The lack of insulation in the existing built stock (deriving from the traditional adaptation of local construction to the climate, the vernacular knowledge loss, and climate change) is responsible for higher energy demands. The use of low hygroscopic materials can preclude a passive control of indoor conditions. Low acoustic properties of the envelope can determine an inefficient use of the designed building spaces (e.g., the closing of the windows, hindering the natural ventilation).

The production of more performant buildings can certainly moderate the energy requirements, hence the energy consumption (Posani et al., 2022). Furthermore, more comfortable, and healthy indoor environments can encourage people to use the designed spaces. The enhancement of indoor comfort can secure both human health and the reduction of operational costs (Fratoni et al., 2019;

Posani et al., 2021), ensuring the well-being of the environment and occupants. Two types of indoor comfort are considered: hygrothermal and acoustic. Hygrothermal comfort depends on the operative temperature, a combination of air temperature and radiant temperature in the room, and it is quite difficult to predict (Ranesi et al., 2022). Consequently, a simplified assessment typically involves several parameters, such as predicted mean vote (PMV), predicted percentage of dissatisfaction (PPD), internal temperature, and relative humidity. Acoustic comfort considers protection against noise and relies heavily on effective sound insulation (Sakthivel et al., 2021), a crucial factor that enhances the comfort and well-being of the users (De Salvio et al., 2023). The control of the acoustic performance could be guaranteed by the layout of the employed materials and their distribution (Fratoni et al., 2023), as well as the indoor relative humidity, and temperature (Cingolani et al., 2022).

In this scenario, the research is more and more interested in looking for eco-friendly solutions to moderate and prevent environmental problems, and to improve the quality of the buildings and indoor conditions. Among these, the employment of bio-based and bio-wastes-based building materials seems to be an efficient possibility (Nunes et al., 2017). It can lower production costs, encourage circular economy practices, and guarantee the disposal of solid wastes (Heniegal et al., 2020; Mahieu et al., 2019; Priyadarshini et al., 2021). Bio-wastes are constantly produced and a significant part of them tends to be landfilled, sometimes causing environmental problems and negative effects (Barbieri et al., 2013; Madurwar et al., 2013). To encourage their use, solutions that secure similar or better performance than conventional products should be proposed.

Past studies already investigated different types of composites by using bio-wastes and bio-resources, such as plasters (Lachheb et al., 2019; Liuzzi et al., 2018), adhesives (binders and glues) (Kariuki et al., 2019; Monteiro et al., 2019; Muizniece and Blumberga, 2016), boards and panels (Eschenhagen et al., 2019; Mahieu et al., 2019; Syamani et al., 2020) and bricks (Raut et al., 2011; Taurino et al., 2019). The results demonstrated that these types of composites can be competitive with conventional ones, with the advantages of sustainability and economic cost. Depending on the considered bio-wastes and bio-resources, the final composites demonstrated certain potentiality: thermal insulation performance (Platt et al., 2023), acoustic absorption (Martellotta et al., 2018), hygroscopicity and moisture buffering capacity (Liuzzi et al., 2018), or mechanical resistance (Molari et al., 2017).

Certainly, all the bio-wastes have several common benefits: they are renewable, recyclable, and biodegradable (Liuzzi et al., 2018), many are widely available all year long, they secure a low environmental impact and have low costs (Berardi and Iannace, 2015), they are not toxic and can guarantee healthy indoor conditions (Nguyen et al., 2017; Palumbo et al., 2017). Nevertheless, there are also recognized drawbacks, such as low fire resistance (Palumbo et al., 2017), high water and moisture absorption (Nguyen et al., 2020), low dimensional stability, and low durability. Furthermore, due to the high hygroscopicity, organic composition, and nutrient content, bio-wastes are highly susceptible

to biological deterioration, namely to fungi, mould, and insects' attacks (Gradeci et al., 2017; Palumbo et al., 2017; Tobon et al., 2020). The high bio susceptibility of bio-based building materials can modify their properties, reduce durability, and lead to health risks for building users (Ginestet et al., 2020). All this information must be known and considered for proper production and employment of bio-based building products.

Among building products, boards are widely used due to their easy on-site installation, their multiple possibilities of applications, and their economic advantages (Çöpür et al., 2008; Ganne-Chédeville and Diederichs, 2015). Particleboards (boards made of particles bounded by adhesives and used for internal and external applications (Gürü et al., 2015)) can be produced by using several techniques, sometimes cumulative (e.g. thermal treatment, pressure process, bonding by adhesives, air-drying) and materials, such as wood chips, fibres, and by-products like bio-wastes, proposing a possibility of waste disposal (Gürü et al., 2015; Mantanis et al., 2018). Past studies have already assessed different types of bio-waste-based boards and panels: thermal insulation boards (Pavelek and Adamová, 2019), acoustic absorption panels (Liuzzi et al., 2020b), coating panels with high hygroscopicity, hence the ability to control internal conditions (Antunes et al., 2019), and particleboards with mechanical resistance (Ferrandez-Villena et al., 2020), demonstrating their several potentialities.

## 1.2 Objective and methodology

Starting from this background, this PhD project investigates indoor bio-based boards that could passively control the internal conditions of buildings. The aim is to produce a sustainable and eco-efficient indoor particleboard to reuse agro-industrial wastes and bio-resources, able to improve indoor comfort (hygrothermal and/or acoustic), ensuring safety in case of fire.

The bibliographic research and laboratory tests supported the selection of the materials, considering benefits, drawbacks, and the final composite's production and application. After identifying the materials, experimental campaigns were carried out to evaluate the composites' properties, mainly focusing on the ones related to indoor comfort and safety. Finally, an introductory life cycle assessment provided information to qualitatively assess the sustainability of the proposed solutions.

The outcomes were the proposal of eco-efficient bio-waste-based building composites, their horizontal characterization, and the description of test methods to analyse these still uncommon building products. Hence, a possible solution to overcome the problem of bio-waste disposal, to reduce the energy consumption by buildings while contributing to passively improve indoor conditions, is provided. In addition to overcoming environmental challenges, this secures users' well-being and contributes to energy efficiency. Finally, besides the main findings and the contribution to this research field, the described work could be useful for developing further research on bio-based building composites' production and assessment.

The research is divided into several phases:

1. Initial bibliographical research on bio-based composite, bio-wastes, and bio-resources already used in construction (mainly considering boards), advantages and possible strengths of bio-based composites, drawbacks, and properties to improve (focusing on bio-susceptibility), bio-aggregates, adhesives (natural and synthetic binders and glues), additives, production process, laboratory tests, and possible gaps in the literature.
2. Selection of some bio-aggregates (favouring the ones not already analysed in past research) and experimental campaign to characterize their relevant properties to predict the ones of the final products. Materials' promising insulation properties, their production, and availability were considered for the selection. Four bio-wastes were examined at first: grape press waste, hazelnut shells, olive press waste, and spent coffee grounds. As for the performed laboratory tests, conventional and unconventional ones were considered (e.g., physical properties and biological susceptibility, one of the main problems of using bio-wastes for building practices).
3. Selection of possible adhesives and additives, considering the previous bibliographic research, to produce boards made of the selected bio-aggregates. Three different adhesives were considered and investigated: a hemihydrated gypsum-based binder, a sodium silicate solution, and a starch-based natural glue. Performance of several practical tests to assess the feasibility of producing boards with the selected materials, define the production process, and evaluate the best compositions.
4. Analysis of boards' properties by performing both already known and standardized laboratory tests and not commonly considered ones. Among them, biological susceptibility to mould without inoculation was considered. Conventional insulation boards, already known and widely studied (considering both commercial products and past research literature), were considered as controls.
5. Selection of one bio-aggregate (the hazelnut shells) and one adhesive (the sodium silicate solution) to produce a composite for further analysis and defining a possible optimization. As for this latter, the addition of another bio-resource already studied and known was considered, namely *A. donax*, known as giant reed or giant cane. The aim was to secure an improvement of the composite's properties, always considering a bio-based matrix.
6. Definition of the most promising aggregate-binder mixture combinations, considering the feasibility of producing boards. Analysis of the best grain size of the bio-aggregate; analysis of the possible production process, definition of the experimental campaign (essential equipment for the production and tests, casting and curing time, the definition of the standardised and no-standardised methods, required replications of samples and their shape and size, testing conditions, etc.). Practical tests to define the production process.
7. Production of bio-based boards, considering both the selected bio-aggregate (the hazelnut shells) and adhesive (the sodium silicate solution), and the selected bio-resource for the optimisation (the *A. donax*). Production of optimized composites by using a mixture of the two bio-aggregates.



8. Performance of laboratory tests to provide a complete characterization of the proposed boards (horizontal analysis): hygrothermal, acoustic, and mechanical properties, bio-susceptibility to mould and insects, performance in case of fire, analysis of materials and boards' composition, analysis of the internal structure, and porosity.
9. Analysis of the results: strengths and weaknesses of the proposed boards (optimized and not), analysis of the most promising performance, and considerations for further studies.
10. Introductory and qualitative Life Cycle Assessment of the proposed boards to define their sustainability and carbon footprint. Identification of possible future studies and applications.

## 1.3 Thesis layout

The present document is divided into six chapters that describe the developed work, main achievements and findings, and conclusions. The introduction to the main topic of the Ph.D. is provided in this first one.

Chapters 2 to 5 describe the different phases of the Ph.D. project, and they are presented by the same structure: a preamble, which introduces the addressed topics; the articles, which describe the developed work in the considered phase, and concluding remarks. Nine articles related to the Ph.D. project, published or submitted to indexed journals (and waiting for feedback), are reported, with the same structure considered for the publication. Hence, at the end of each one, the references were reported in the reference style of the journal. Complementary unpublished sections are also included in the thesis, namely in Chapters 2 and 5.

The chapters' contents are:

- Chapter 2: Initial literature review, which includes, both published and unpublished information.
- Chapter 3: Analysis of the selected bio-aggregates, as raw materials and, together with the selected adhesive and additives, of bio-composites.
- Chapter 4: Further analysis of a selected composite, namely with one bio-aggregate and one adhesive among the previously analysed ones, and the selected bio-resource for the composite's optimization.
- Chapter 5: Experimental campaign to assess the properties of the optimized boards and launched activities for further analysis.
- Chapter 6: General conclusion of the research work is presented, as well as the perspectives, and information about further studies.
- References: The references of the thesis document are reported, without including the references of the articles (which are reported immediately after each of them in the journal's style).
- Appendix: Supplementary materials, namely some detailed results of the initial literature review (Chapter 2).



## MATERIALS AND TESTS FOR BIO-WASTE-BASED BOARDS

### 2.1 Preamble

The development of sustainable architecture practices is an efficient solution to reduce energy consumption, moderate the environmental impact of the construction sector, and improve indoor comfort and users' well-being. It is widely known that buildings consume 40% of the total energy and require a significant amount of resources (Asdrubali et al., 2015; European Commission, 2019; Posani et al., 2021). For this reason, the research is more and more interested in investigating innovative sustainable solutions. The use of natural and environmentally friendly building materials is a widely analysed possibility. It can guarantee passive control of internal conditions, securing better indoor air quality and reducing energy waste, without spending additional resources (Dittenber and GangaRao, 2012; Nguyen et al., 2018). Furthermore, the improvement of the thermal and acoustic insulation performance of the building stock lowers the energy demand, moderating the pollution and costs derived from the building sector (Binici and Aksogan, 2016) and avoiding inefficient use of the designed building spaces.

Past research addressed the production of different types of bio-based composites and evaluated their properties. Considering the promising thermal and acoustic insulation performance of some bio-waste-based composites, the economic advantages derived from their use (Asdrubali et al., 2015), the possibility of moderating the problem of waste disposal and promoting circular economy practices (Binici and Aksogan, 2016; Gonçalves et al., 2021; Liuzzi et al., 2017), bio-wastes seem to be promising as building materials. The use of bio-wastes as building materials has simultaneously advantages and disadvantages, whose assessment allows more conscious employment, enhancing the strengths and moderating the weaknesses.

As for the benefits, their high porosity ensures the production of composites with thermal insulation and acoustic absorption properties (Berardi and Iannace, 2015; Dittenber and GangaRao,

2012). Their good moisture buffering capacity guarantees the control of relative humidity, hence the indoor conditions (Liuzzi et al., 2020a; Nguyen et al., 2018). Indeed, bio-waste-based composites can passively improve both acoustic and hygrothermal comfort. Some bio-waste-based composites showed good mechanical properties, achieving the mechanical strength values required by standards to be employed as building products (Cabral et al., 2018; Sanjay et al., 2018). Furthermore, bio-wastes are recyclable and biodegradable, they have a low environmental impact and guarantee the development of passive construction techniques. Finally, they secure the production of healthy building materials, avoiding the use of harmful substances for human health (Ali et al., 2020).

On the other hand, they showed low resistance to water and fire; they could have low dimensional stability and low durability (Dittenber and GangaRao, 2012). Being bio-wastes organic substances, they are highly prone to biological attacks, and then degradation (Berardi and Iannace, 2015; Palumbo et al., 2015). Some of bio-wastes' properties could be both advantages and disadvantages, depending on the final use. For example, the hydrophilic nature could both guarantee the passive control of indoor conditions and entail low dimensional stability, low water resistance, and lower mechanical properties at high relative humidity.

Starting from these considerations, an initial literature review was carried out to individuate the bio-wastes already analysed to produce composites, focusing on panels, boards, and the passive control of indoor comfort (Section 2.2). A classification of the already employed bio-wastes was provided. After considering several past studies, only the agro-industrial wastes (bio-wastes derived from agricultural practices and agro-industrial processes) were detailed. Information about their production, their chemical composition, and their properties was collected, focusing the analysis on the Euro-Mediterranean Countries. Furthermore, the benefits and drawbacks of bio-wastes as building materials were pointed out, mainly focusing on biological susceptibility. As outcomes, four bio-wastes produced in the considered area, not already widely analysed, were selected: grapes press waste, hazelnut shells, olives press waste, and spent coffee grounds.

Then, the literature review was carried out to choose the adhesives, the additives, and the production process (Sections 2.4 and 2.5). Materials that could guarantee low environmental impact and the feasibility of producing boards, improving bio-wastes' advantages, and moderating their drawbacks, were considered. Furthermore, production processes that avoided high temperatures were preferred, to prevent bio-aggregates' degradation and ensure low environmental impact. Materials that were not harmful to human health and did not emit harmful substances were selected. Again, particular attention was paid to the biological susceptibility, which influenced the choice of both the adhesives and the additives (Section 2.3).

Finally, a review of laboratory tests already performed to analyse building materials was carried out (Section 2.6). Hygrothermal performance and biological susceptibility of agro-industrial wastes, both individually and in composites, were considered. This was due to some of the main aims, namely

the improvement of indoor comfort (in this case considered as the hygrothermal one), and the moderation of the bio-based materials' drawbacks. The results of this review demonstrated that several past studies considered the evaluation of bio-wastes as building materials by performing different laboratory tests, usually for composites. Furthermore, the data collection confirmed that bio-susceptibility was not so commonly assessed. However, many laboratory tests can be carried out to assess it, due to the different bio-agents and critical conditions. The outcome was a summarised list of methods which has been a simplified starting point to study agro-industrial wastes and agro-industrial waste-based composites for building practices.

## 2.2 Article A - Agro-industrial wastes as building insulation materials: A review and challenges for Euro-Mediterranean Countries

The information reported in this Section was published in *Industrial Crops and Products* 171 (2021) 113833 - <https://doi.org/10.1016/j.indcrop.2021.113833>

### Abstract

This study investigates the possibility of using agro-industrial wastes for building products, mainly focusing on their insulation properties. A classification of bio-wastes is provided, namely of the lignocellulosic ones and their features and properties are described. Information about three main topics is collected: world production and consumption of some crops already used as building materials, their chemical composition and their most studied properties. Since the considered materials are lignocellulosic and they have many common features, a comparison is made. The aim is to have comparable information to support future research related to the production of eco-efficient indoor insulation boards. The result of this research is the choice of four different agro-industrial wastes produced in the Euro-Mediterranean Countries. This area was chosen as buildings typically have little or no insulation due to the regional mild climate; however, particularly with climate change, indoor hygrothermal comfort is poor. The collection of information allows some conclusions to be reached about the different bio-wastes already studied and identify gaps in the literature.

**Keywords:** Bio-wastes; Boards; Fibres; Insulation; Natural materials; Thermal properties.

### 2.2.1 Introduction

The increasing interest in environmental impact and safeguarding of natural resources leads research to find out solutions to reduce the negative effects of human practices. The building sector has been identified as having a significant impact in terms of its ecological consequences. Often it requires high energy inputs, produces high volumes of environmentally harmful wastes, and consumes a huge percentage of global water and resources (Chikhi et al., 2013; Asdrubali et al., 2015; Ratiarisoa et al., 2016; Viel et al., 2019). Building materials are one of the major factors influencing the overall environmental impact of the construction sector due to their production, transport, use and end of life. To reduce this problem, some strategies have been found out, whilst others are still being implemented: the use of environment-friendly materials, the employment of local materials, the encouragement of circular economy practices, passive building techniques and the development of new techniques such as off-site construction (Hosseini et al., 2018; Jin et al., 2020). Building materials can

also influence the working principles of building, both negatively and positively. They can ensure a passive control of indoor hygrothermal conditions influencing both energy demand and comfort, as well as occupants' well-being and health.

Nowadays, research trends focus on the possibility of using natural materials such as fibres in construction products with clear advantages: low environmental impact, less energy demand, low cost, low density, large-scale availability, biodegradability and good insulation and mechanical properties (Mohanty et al., 2005; Dittenber and GangaRao, 2012; Faruk et al., 2012; Berardi and Iannace, 2015; Nguyen et al., 2018a; Sanjay et al., 2018). Several studies evaluated the use of bio-wastes for building products, considering various possible applications, and analysing their different properties. For example, Asdrubali et al. (2015) collected information about natural and recycled materials used for thermal and acoustic insulation. Nunes et al. (2017) reviewed the use of agricultural products in the building sector, reporting their physical and mechanical properties. They also gave information related to bio-based materials used for particleboards' production. Popescu (2017) reported that research interest in using renewable fibres and bio-wastes for panels' production is growing. Posani et al. (2019) addressed the use of some natural materials for insulating historic constructions. Each material exhibits different properties due to its chemical composition, structure and characteristics (e.g., moisture buffering capacity, thermal insulation performance, mechanical behaviour, etc.). Knowing those properties allows suitable choices to be made, to guarantee the required performance of the composites that include them.

As previously mentioned, the production and transport of materials are among the aspects that influence the main environmental impacts of a building. Employing local materials ready-made in their natural form, such as earth, bamboo, reeds, without using synthetic compounds, offers a sustainable solution. Another efficient possibility is the use of bio-wastes. They are continuously produced, and a significant part of those wastes tends to be landfilled. Sometimes they cause environmental problems and negative effects, but they can also be useful resources when employed in construction (Karade, 2010; Raut et al., 2011; Madurwar et al., 2013; Viel et al., 2018; Kazmi et al., 2018). This guarantees an improvement in the disposal of residues, an incitement to recycle and to eco-friendly construction practices. To encourage their use, it is extremely important to propose solutions that secure similar or better performance compared to conventional building materials.

The present article focuses on lignocellulosic materials, considering both fibres and raw materials as they are. Natural fibres are filamentous substances found in animals, plants and minerals. Vegetal ones are composed mainly of cellulose, whereas animal ones are rich in protein (John and Thomas, 2008). Past studies provided different classifications (Mohanty et al., 2005; Jawaid and Abdul Khalil, 2011; Faruk et al., 2012; Nguong et al., 2013; Berardi and Iannace, 2015; Sanjay et al., 2018). Figure 2.2-1 shows an adaptation of that proposed by Jawaid and Abdul Khalil (2011), considering all types of fibres. As concerns the lignocellulosic fibres, their features depend on several factors,

such as location and climate conditions of crop production, plant species and harvesting (Dittenber and GangaRao, 2012).

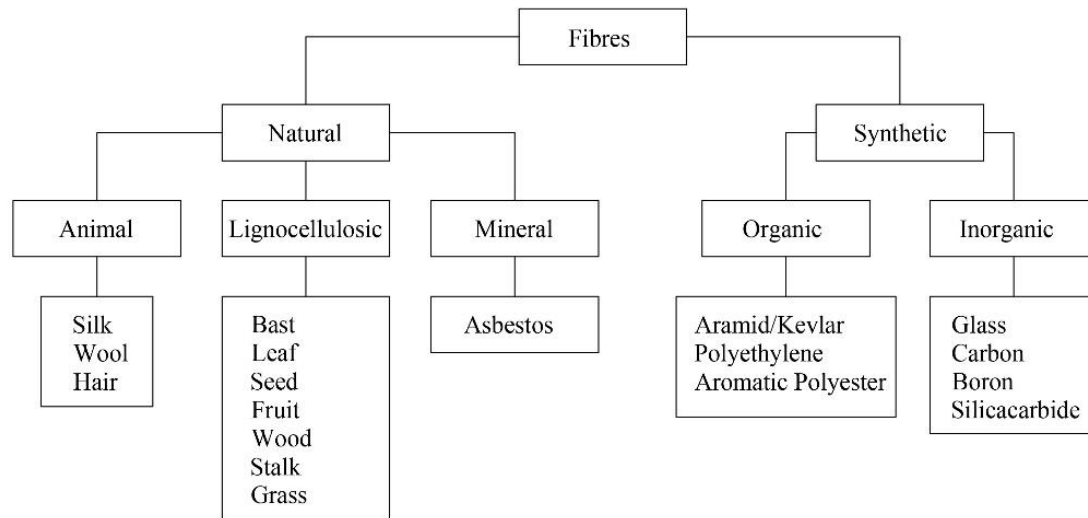


Figure 2.2-1. Classification of natural and synthetic fibres (adapted from Jawaid and Abdul Khalil, 2011).

The present article carries out a general analysis concerning the use of bio-wastes, studied to create several construction products, such as bricks, plasters, adhesives, and particleboards, and it gives a classification of lignocellulosic bio-wastes. It subsequently deals only with agro-industrial bio-wastes already studied as building materials and their properties. It collects information about worldwide production, chemical composition, and properties (focusing mainly on thermal performances). As a result of this data collection, production and consumption in Euro-Mediterranean Countries were taken into account to select some bio-wastes to be studied in more detail. As concerns chemical composition and properties, the article focuses on board production to collect useful information for future research.

The results allow an overview of the state of the art, identifying the gaps in the literature and possible new solutions to produce insulation boards for interior use. The outcome is the selection of four bio-wastes, largely produced in Euro-Mediterranean Countries, to consider and study as aggregates for innovative eco-efficient building composites.

## 2.2.2 Lignocellulosic bio-wastes as building materials and products

Among the natural materials that can be used as building materials, bio-wastes can be an effective possibility. As previously anticipated, they are continuously produced, available in large quantities and can be used to produce different composites. Considering the ones already studied for construction products, it is possible to differentiate some types of bio-wastes and make a classification: those derived from forest management, floriculture and use of flowers and shrubs; those derived



from agricultural and manufacturing practices and produced by the use of the agro-products and those from the food industry. Their world production and consumption are substantial and increasing over the years (Papadopoulou et al., 2015; Viel et al., 2018). For this reason, they can be a good source for natural composite production. Tables 2.2-1, 2.2-2 and 2.2-3 report some examples of bio-wastes already examined in past studies related to building materials, considering this classification (forest management and floriculture, agricultural practices and agro-industrial processes respectively). Due to the large quantity of studies, only the research works published in the last decade (2010-2020) were considered.

### 2.2.3 Bio-wastes derived from forest management and floriculture

Several past research analysed the use of bio-wastes derived from forest management and floriculture. Cárdenas et al. (2018) studied the possibility of using stems of Hortensia (*Hydrangea macrophylla*) to produce thermal insulation materials. The researchers investigated the physical and chemical properties of this plant and studied a composite material adding commercial polyurethane. The results demonstrated that it has good insulation properties and better performances if compared with commercial expanded polystyrene. Asdrubali et al. (2015) considered many past studies that demonstrated the possibility of using residues from forest pruning to produce thermal insulation boards. They reported that particleboards made up of cattail fibre (*Typha*), a plant characterized by very fast growth and that produces negative effects on the other crops, have good performances due to the low thermal conductivity (between 0.044 W/(m.K) and 0.061 W/(m.K)). Ratiarisoa et al. (2016) studied the possibility of using lavender (*Lavandula* sp.) straw, derived from a distillation treatment used in essential oil extraction, to produce a composite. They investigated the physical properties and thermal, mechanical and hygroscopic behaviour of a composite with a pozzolanic binder, made up of Metakaolin and slaked lime. Results showed promising hygrothermal characteristics (thermal conductivity, moisture buffering capacity and water vapour permeability) if compared with other bio aggregates already studied. Viel et al. (2018) considered different bio-based aggregates, including rape straw, derived from Brassica. They investigated its chemical and physical characterization and demonstrated that it has good hygrothermal properties, low thermal conductivity, and high moisture buffer value and water absorption. Water absorption results indicated a high porosity, an important feature for thermal insulation performances. Indeed, there is a relationship of inverse proportionality between thermal conductivity and open porosity. Moisture buffer value showed the potential of a material to passively control indoor conditions. The researchers concluded that bio-based aggregates could be used to produce insulation products such as indoor boards. Other past studies (Jawaid and Abdul Khalil, 2011; Faruk et al., 2012; Sanjay et al., 2018) have reported that sisal fibres, extracted from *Agave*, are another efficient possibility to produce composites for building.

Table 2.2-1. Example of materials derived from forest management and floriculture already studied for building products considering research works published in 2010-2020.

| Lignocellulosic wastes                                        | bio- | Designed product  | Part of waste      | Description*                                                                                                                                                                         | References                              |
|---------------------------------------------------------------|------|-------------------|--------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------|
| Hortensia ( <i>Hydrangea macrophylla</i> (Thumb.) Ser)        |      | Polymers          | Stem               | Dried stems of Hortensia to produce a bio-polymer for thermal insulation materials. Study of samples made up of hortensia and commercial polyurethane.                               | Cárdenas et al., 2018                   |
|                                                               |      | Polymers, binders | Stem               | Feasibility of the production of a bio-polymer for thermal insulation materials and a binder for insulation panels production.                                                       | Cárdenas, 2020                          |
| Cattail ( <i>Typha latifolia</i> L.)                          |      | Boards            | Fibre              | A past studies that analysed thermal insulation boards composed of Cattail fibres and Methylene Diphenyl Diisocyanate binder.                                                        | Asdrubali et al., 2015                  |
| Rape straw ( <i>Brassica napus</i> L.)                        |      | Concretes         | Straw              | Rape straw derived from the woody heart of the stems. Analysis of hygric properties of concrete made of rape straw and lime.                                                         | Nguyen et al., 2017; Rahim et al., 2016 |
|                                                               |      | Aggregates        | Straw              | Characterization of rape straw to evaluate its possible use as a raw building material for different compounds.                                                                      | Viel et al., 2018                       |
| Agave ( <i>Agave</i> sp.)                                     |      | Boards            | Fibre              | Thermal insulation material made of agave fibres/wheat straw and corn starch as a binder.                                                                                            | Ali et al., 2020a                       |
| Apple of Sodom ( <i>Calotropis procera</i> (Aiton) W.T.Aiton) |      | Boards            | Fibre              | Boards made up of Apple of Sodom fibres, Phenol-Formaldehyde resin and cornstarch as binders.                                                                                        | Ali and Zeitoun, 2012                   |
|                                                               |      | Composites        | Fibre              | Hybrid insulating composites made up of Apple of Sodom fibre, date palm tree surface fibre and Agave fibre and cornstarch, white cement and glue (wood adhesive 78-1040) as binders. | Alabdulkarem et al., 2018               |
| Lavender ( <i>Lavandula angustifolia</i> Mill.)               |      | Aggregates        | Straw              | Analysis of lavender straw as possible bio-aggregate in a pozzolanic matrix composed of metakaolin and slaked lime.                                                                  | Ratiarisoa et al., 2016                 |
| Miscanthus ( <i>Miscanthus</i> spp.)                          |      | Boards            | Fibre              | Boards made up of Miscanthus fibres, sunflower stalk and natural binders (starch-based binder, wood glue made of casein, bone glue).                                                 | Eschenhagen et al., 2019                |
|                                                               |      | Boards            | Fibre              | Thermal insulation boards made up of Miscanthus fibres, cement-based binder and hydrated lime.                                                                                       | Savic et al., 2020                      |
| Reed ( <i>Phragmites australis</i> (Cav.) Trin ex. Steud.)    |      | Aggregates        | Wooden parts, bark | Evaluation of the acoustic properties of the raw material to produce sound-absorbing boards.                                                                                         | Berardi and Iannace, 2015               |
| Cork ( <i>Quercus suber</i> L.)                               |      | Boards            | Granules           | Sandwich panels with thermal insulation properties having cork as core and commercial flax fibres and epoxy resin as external layers.                                                | Liu et al., 2017; La Rosa et al., 2014  |

| Lignocellulosic wastes                      | bio- | Designed product | Part of waste | Description*                                                                                                         | References                |
|---------------------------------------------|------|------------------|---------------|----------------------------------------------------------------------------------------------------------------------|---------------------------|
|                                             |      | Aggregates       | Granules      | Past studies that examined cork for mortar and concrete production (to replace stone).                               | Chikhi et al., 2013       |
|                                             |      | Aggregates       | Granules      | Acoustic properties of the raw material are evaluated to produce sound-absorbing boards.                             | Berardi and Iannace, 2015 |
|                                             |      | Concretes        | Granules      | Cement-bonded composites made of cork granules to produce light-weight concrete.                                     | Karade, 2010              |
| Eucalyptus fibres ( <i>Eucalyptus</i> spp.) |      | Composites       | Fibre         | A past study that investigated the feasibility of using Eucalyptus fibres as reinforcement in cement-based matrices. | Chikhi et al., 2013       |

\* Note: The description section reports "past study/past studies" when the cited articles refer to other past research work.

Table 2.2-2. Example of materials derived from agricultural practices already studied for building products considering research works published in 2010-2020.

| Lignocellulosic wastes     | bio- | Designed product     | Part of waste | Description*                                                                                                                                                                                                                                                              | References                    |
|----------------------------|------|----------------------|---------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------|
| Banana ( <i>Musa</i> spp.) |      | Composites           | Fibre         | Past studies that analysed banana fibres and several materials: cement, polyurethane, aliphatic polyester resin, PP, urea-formaldehyde, PE, polyester, polyvinyl alcohol.                                                                                                 | Faruk et al., 2012            |
|                            |      | Composites           | Fibre         | Past studies that considered several banana composites such as banana/kenaf and unsaturated polyester, sisal/banana and polyester, banana/glass and phenol-formaldehyde, banana/glass and polypropylene (PP), banana/glass hybrid fibre reinforced polystyrene composite. | Jawaid and Abdul Khalil, 2011 |
|                            |      | Composites           | Fibre         | Past studies that considered banana reinforced composites.                                                                                                                                                                                                                | Sanjay et al., 2018           |
|                            |      | Mortars              | Fibre         | Feasibility of using banana fibres for polymer-modified cement mortars (banana fibres could partially replace cement).                                                                                                                                                    | Akinyemi and Dai, 2020        |
|                            |      | Composites           | Fibre         | A past study that analysed epoxy composites reinforced by banana fibre.                                                                                                                                                                                                   | Nguong et al., 2013           |
|                            |      | Boards               | Fibre         | Feasibility of using banana fibres partially replacing maritime pine ( <i>Pinus pinaster</i> Ait.) particles for cement-bonded particleboards                                                                                                                             | Nunes et al., 2021            |
| Cereal (several species)   |      | Plasters             | Straw         | A past study that evaluated the hygrothermal properties of clay plasters with barley ( <i>Hordeum vulgare</i> ) straw and corn pith.                                                                                                                                      | Liuzzi et al., 2018           |
|                            |      | Boards               | Straw         | A past study that considered fibreboards with barley straw.                                                                                                                                                                                                               | Nguyen et al., 2017           |
|                            |      | Insulation materials | Straw         | Past studies that analysed insulation materials with cereal straws.                                                                                                                                                                                                       | Liu et al., 2017              |

| Lignocellulosic wastes     | bio-               | Designed product     | Part of waste | Description*                                                                                                                                                                 | References                    |
|----------------------------|--------------------|----------------------|---------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------|
|                            |                    | Composites           | Straw         | Past studies that considered cereals straw in different applications.                                                                                                        | Cárdenas et al., 2018         |
|                            |                    | Binders              | Grains        | Cereals grains (corn and wheat) to produce starch used as a binder.                                                                                                          | Monteiro et al., 2016         |
|                            |                    | Boards               | Straw         | Thermal insulation material made up of agave fibres/wheat straw and corn starch as a binder.                                                                                 | Ali et al., 2020a             |
|                            |                    | Boards               | Straw         | Straw used for bio-insulation materials (fibreboards).                                                                                                                       | Nguyen et al., 2020           |
|                            |                    | Boards               | Straw         | Past studies that considered cement-bonded particleboards.                                                                                                                   | Karade, 2010                  |
| Corn ( <i>Zea mays</i> L.) |                    | Plasters             | Pith          | A past study that evaluated the hygrothermal properties of clay plasters with barely straw and corn pith.                                                                    | Liuzzi et al., 2018           |
|                            |                    | Insulation materials | Straw, stalk  | Past studies that analysed insulation materials with corn.                                                                                                                   | Liu et al., 2017              |
|                            |                    | Natural glues        | Starch        | A past study that considered corn to produce wood adhesives for indoor applications.                                                                                         | Cardoso et al., 2015          |
|                            |                    | Boards               | Cob           | Past studies that considered particleboards made of ground corn cobs and wood glue.                                                                                          | Asdrubali et al., 2015        |
|                            |                    | Insulation materials | Cob           | A past study that considered corn to produce thermal insulation material.                                                                                                    | Chikhi et al., 2013           |
|                            |                    | Aggregates           | Cob           | Hemp shiv, corn cob and natural binders to produce thermal insulation materials.                                                                                             | Viel et al., 2019             |
|                            |                    | Insulation blocks    | Husk          | Insulation block made of wheat straw and corn husk.                                                                                                                          | Rojas et al., 2019            |
|                            |                    | Boards, cement       | Husk, cob     | Past studies that considered particleboards composed of waste tissue paper manufacturing and corn peel, and cement produced by corn cob ash.                                 | Madurwar et al., 2013         |
|                            |                    | Insulation materials | Cob           | A past study that considered corn cob for bio-insulation materials.                                                                                                          | Nguyen et al., 2020           |
|                            |                    | Boards               | Starch        | Thermal insulation material composited of agave fibres/wheat straw and corn starch as a binder.                                                                              | Ali et al., 2020a             |
| Cotton spp.)               | ( <i>Gossypium</i> | Boards               | Stalk         | A past study that considered thermal insulation fibreboard made by a hot-pressing method.                                                                                    | Liu et al., 2017              |
|                            |                    | Boards               | Stalk         | A past study that considered particleboards made of cotton stalk manufactured without using chemical binders.                                                                | Asdrubali et al., 2015        |
|                            |                    | Composites           | Fibre         | Past studies that considered cotton fibres for different applications.                                                                                                       | Cárdenas et al., 2018         |
|                            |                    | Composites           | Fibre         | Past studies that considered several cotton fibres composites such as jute/cotton and novolac phenolic or polyester, cotton/ramie and polyester, sisal/cotton and polyester. | Jawaid and Abdul Khalil, 2011 |

| Lignocellulosic wastes                            | bio- | Designed product     | Part of waste | Description*                                                                                                                                       | References                    |
|---------------------------------------------------|------|----------------------|---------------|----------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------|
|                                                   |      | Composites           | Fibre         | Past studies that analysed cotton-polyester composites.                                                                                            | Shalwan and Yousif, 2013      |
|                                                   |      | Boards, bricks       | Stalk, waste  | Past studies that considered cotton stalk for fibreboard production without chemical additives and cotton waste for brick production.              | Madurwar et al., 2013         |
|                                                   |      | Bricks               | Waste         | A past study that considered cotton waste and lime powder waste to produce bricks.                                                                 | Raut et al., 2011             |
| Date palm fibres ( <i>Phoenix dactylifera</i> L.) |      | Mortars              | Fibre         | A past study that evaluated mortars made up of sand, cement, and date palm fibres.                                                                 | Liuzzi et al., 2018           |
|                                                   |      | Insulation materials | Fibre         | A past study that evaluated the properties of date palm and gypsum-based materials.                                                                | Liu et al., 2017              |
|                                                   |      | Insulation materials | Fibre         | A past study that evaluated the properties of date palm and gypsum-based materials.                                                                | Asdrubali et al., 2015        |
|                                                   |      | Insulation materials | Fibre         | Properties of date palm and gypsum-based materials.                                                                                                | Chikhi et al., 2013           |
|                                                   |      | Insulation materials | Fibre         | Evaluation of thermal insulation and mechanical properties of a composite made up of date palm surface fibres and cornstarch solution as a binder. | Ali and Alabdulkarem, 2017    |
| Flax ( <i>Linum usitatissimum</i> L.)             |      | Composites           | Fibre         | A past study that considered lime-flax shiv composite.                                                                                             | Nguyen et al., 2017           |
|                                                   |      | Insulation materials | Fibre         | Past studies that considered flax fibres for insulation materials.                                                                                 | Liu et al., 2017              |
|                                                   |      | Composites           | Fibre         | Past studies that analysed flax fibre composites.                                                                                                  | Faruk et al., 2012            |
|                                                   |      | Composites           | Fibre         | Past studies that considered several flax fibres composites such as flax/glass and polypropylene, cotton/flax and polyethylene.                    | Jawaid and Abdul Khalil, 2011 |
|                                                   |      | Composites           | Fibre         | Past studies that considered flax fibre reinforced composites.                                                                                     | Sanjay et al., 2018           |
|                                                   |      | Boards               | Shiv          | A past study that evaluated the properties of fibreboards made up of flax-shiv and casein binder.                                                  | Nguyen et al., 2018a          |
|                                                   |      | Aggregates           | Shiv          | Characterization of rape straw to evaluate the feasibility of using it as a raw building material for different composites.                        | Viel et al., 2018             |
|                                                   |      | Boards               | Shiv          | Past studies that considered fibreboards composed of flax fibres without binders, with tannin binder and protein binder.                           | Nguyen et al., 2020           |
|                                                   |      | Boards               | Shiv          | Properties of particleboards made up of flax shiv, sunflower stems and urea-formaldehyde as a binder.                                              | Mahieu et al., 2019           |
| Hemp ( <i>Cannabis sativa</i> L.)                 |      | Concrete, Boards     | Shiv, Fibre   | Past studies that considered hemp concrete properties and fibreboards made up of hemp fibres.                                                      | Nguyen et al., 2017           |

| Lignocellulosic wastes                             | bio- | Designed product     | Part of waste  | Description*                                                                                                                                                                          | References                    |
|----------------------------------------------------|------|----------------------|----------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------|
|                                                    |      | Insulation materials | Straw, fibre   | A past study that evaluated the properties of hemp fibres for insulation materials.                                                                                                   | Liu et al., 2017              |
|                                                    |      | Boards               | Fibre          | Past studies that considered thermal and acoustic insulation properties of boards made of hemp fibre.                                                                                 | Nguyen et al., 2018b          |
|                                                    |      | Aggregates           | Shiv           | Characterization of hemp shiv to evaluate the feasibility of using it as a raw building material for different composites.                                                            | Viel et al., 2019             |
|                                                    |      | Boards               | Fibre          | Considered acoustic insulation properties of boards produced by treating hemp fibres with materials to improve the fire resistance (soda or boron salts).                             | Berardi and Iannace, 2015     |
|                                                    |      | Composites           | Fibre          | Past studies that considered hemp fibres in composites.                                                                                                                               | Faruk et al., 2012            |
|                                                    |      | Composites           | Fibre          | Past studies that considered hemp fibre composites such as hemp/glass and polypropylene.                                                                                              | Jawaid and Abdul Khalil, 2011 |
|                                                    |      | Aggregates           | Shiv           | Characterization of hemp shiv to evaluate the feasibility of using it as a raw building material for different composites.                                                            | Viel et al., 2018             |
|                                                    |      | Concretes            | Fibre          | Past studied that considered bio-concrete.                                                                                                                                            | Nguyen et al., 2020           |
| Kenaf ( <i>Hibiscus canna-</i><br><i>binus</i> L.) |      | Boards               | Fibre          | Acoustic insulation properties (sound absorption) of pure kenaf samples.                                                                                                              | Berardi and Iannace, 2015     |
|                                                    |      | Composites           | Fibre          | Past studies that considered kenaf fibres in composites.                                                                                                                              | Faruk et al., 2012            |
|                                                    |      | Composites           | Fibre          | Past studies that considered several kenaf fibres composites such as kenaf/glass and natural rubber and banana/kenaf hybrid composites.                                               | Jawaid and Abdul Khalil, 2011 |
|                                                    |      | Composites           | Fibre          | Past studies that considered fibre-reinforced composites such as kenaf/PP, kenaf/corn starch, kenaf/epoxy.                                                                            | Shalwan and Yousif, 2013      |
| Rice ( <i>Oryza</i> spp.)                          |      | Boards               | Husk           | Boards made up of rice husk, earth, gypsum and air lime.                                                                                                                              | Antunes et al., 2019          |
|                                                    |      | Boards               | Husk, straw    | A past study that evaluated particleboards composed of rice husk and acoustic properties of rice straw measured in a reverberation room.                                              | Asdrubali et al., 2015        |
|                                                    |      | Composites           | Husk, straw    | Past studies that analysed rice husk and rice straw composites.                                                                                                                       | Faruk et al., 2012            |
|                                                    |      | Bricks               | Husk, husk ash | Past studies that considered rice husk for brick production.                                                                                                                          | Ozturk et al., 2019           |
|                                                    |      | Bricks, cement       | Husk, straw    | Past studies that considered brick made of different compounds: sludge from dried water treatment and rice husk, rice husk ash and expanded polystyrene, clay sand and rice husk ash. | Madurwar et al., 2013         |
|                                                    |      | Bricks               | Husk           | A past study that evaluated rice husk as cement replacement material.<br>A past study that evaluated clay-sand brick varying percentages of rice husk ash and firing times.           | Raut et al., 2011             |

| Lignocellulosic wastes | bio- | Designed product | Part of waste | Description*                                                                                                                       | References            |
|------------------------|------|------------------|---------------|------------------------------------------------------------------------------------------------------------------------------------|-----------------------|
|                        |      | Glues            | Bran          | A past study that considered the adhesive properties of rice bran (composed of starch and protein) to fabricate plywood specimens. | Patel et al., 2013    |
|                        |      | Composites       | Husk          | Properties and manufacturing process of bio-composites made of rice husk.                                                          | Muthuraj et al., 2019 |

\* Note: The description section reports "past study/past studies" when the cited articles refer to other past research work.

Table 2.2-3. Example of materials derived from agro-industrial processes already studied for building products considering research works published in 2010-2020.

| Lignocellulosic bio-wastes                           | Designed product      | Part of waste | Description*                                                                                                                                 | References                 |
|------------------------------------------------------|-----------------------|---------------|----------------------------------------------------------------------------------------------------------------------------------------------|----------------------------|
| Bagasse/Sugarcane ( <i>Saccharum officinarum</i> L.) | Boards                | Fibre         | Past studies that considered thermal conductivity and acoustic properties of bagasse boards.                                                 | Asdrubali et al., 2015     |
|                                                      | Cement                | Fibre, ash    | Past studies that examined bagasse as cement reinforced and bagasse ash as cement replacement material.                                      | Madurwar et al., 2013      |
|                                                      | Insulation materials  | Fibre         | A past study that evaluated bagasse fibres properties for thermal insulation materials.                                                      | Nguyen et al., 2020        |
|                                                      | Boards                | Fibre         | Past studies that considered particleboards made of bagasse and manufacture process.                                                         | Karade, 2010               |
|                                                      | Boards                | Waste         | Analysis of indoor insulation boards without chemical binding additives.                                                                     | Panyakaew and Fotios, 2011 |
|                                                      | Boards                | Stalk         | Particleboards made of bagasse stalk (cut and dried) and citric acid.                                                                        | Syamani et al., 2020       |
| Cassava ( <i>Manihot esculenta</i> Crantz)           | Binders               | Starch        | Sour cassava starch, distilled water, chitosan solution, <i>Populus</i> fibres and glycerol as binders for particleboards production.        | Monteiro et al., 2016      |
|                                                      | Binders               | Starch        | Sour cassava starch, distilled water, chitosan solution, <i>Pinus pinaster</i> fibres and glycerol as binders for particleboards production. | Monteiro et al., 2019      |
| Citrus ( <i>Citrus</i> spp.)                         | Binders and additives | Citric acid   | Review that reported properties of citric acid as a binder and additive to modify wood properties.                                           | Lee et al., 2020           |
|                                                      | Binders               | Citric acid   | Citric acid dissolved in water and used as adhesive for particleboards production.                                                           | Widyorini et al., 2016     |
|                                                      | Boards                | Citric acid   | Particleboards made of bagasse stalk (cut and dried) and citric acid (dissolved in water).                                                   | Syamani et al., 2020       |

| Lignocellulosic bio-wastes             | Designed product | Part of waste         | Description*                                                                                         | References                      |
|----------------------------------------|------------------|-----------------------|------------------------------------------------------------------------------------------------------|---------------------------------|
| Coconut ( <i>Cocos nucifera</i> L.)    | Composites       | Fibre                 | Past studies related to the possibility of replacing synthetic fibres in composites.                 | Liuzzi et al., 2018             |
|                                        | Boards           | Fibre                 | A past study that demonstrated the feasibility of using coconut fibres for board production.         | Ashour et al., 2010             |
|                                        | Boards           | Fibre                 | Acoustic insulation properties of coconut brown fibres.                                              | Berardi and Iannace, 2015       |
|                                        | Composites       | Fibre                 | Past studies that considered fibre-reinforced composites such as coir/PP, coir/polyester.            | Shalwan and Yousif, 2013        |
|                                        | Boards           | Fibre, pith           | A past study that analysed particleboards made of durian peel and coconut coir                       | Madurwar et al., 2013           |
|                                        | Boards           | Fibre                 | A past study that provided some recommendation to produce cement-bonded boards and their properties. | Karade, 2010                    |
|                                        | Boards           | Husk (fibre and pith) | Analysis of indoor insulation boards without chemical binding additives.                             | Panyakaew and Fotios, 2011      |
| Coffee ( <i>Coffea</i> spp.)           | Boards           | Chaff                 | Acoustic properties of samples made of coffee chaff and polyurethane glue.                           | Ricciardi et al., 2017          |
|                                        | Geo-polymers     | Ground                | New polymer composed of coffee grounds, blast furnace slag and fly ash.                              | Kua et al., 2016                |
|                                        | Bricks           | Ground                | Brick made of clay and coffee ground, compressed, dried and fired.                                   | Eliche-Quesada et al., 2011     |
|                                        | Plasters         | Ground                | Analysis of a plaster made of semi hydrate gypsum and different percentages of coffee grounds.       | Lachheb et al., 2019            |
|                                        | Bricks           | Ground                | Fired clay bricks with coffee grounds.                                                               | Muñoz Velasco et al., 2016      |
| Durian ( <i>Durio</i> spp.)            | Boards           | Peel                  | A past study that evaluated particleboards composed of durian peel and different type of binders.    | Asdrubali et al., 2015          |
|                                        | Boards           | Peel                  | A past study that analysed particleboards made of durian peel and coconut coir.                      | Madurwar et al., 2013           |
| Hazelnut ( <i>Corylus avellana</i> L.) | Cement           | Shell                 | A past study that evaluated the properties of cementitious composite with hazelnut shells.           | Karade, 2010                    |
|                                        | Polymers         | Shell                 | Modified polymer (HDPE) using hazelnut shells.                                                       | Salasinska and Ryszkowska, 2012 |



| Lignocellulosic bio-wastes                | Designed product            | Part of waste   | Description*                                                                                                                                                | References                                         |
|-------------------------------------------|-----------------------------|-----------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------|
|                                           | Boards                      | Shell           | Feasibility of replacing wood particles with hazelnut shells for particleboard production using resin and hot pressing method.                              | Lopes et al., 2012                                 |
| Jute ( <i>Corchorus</i> spp.)             | Composites                  | Fibre           | Past studies that considered jute fibres composites such as jute/glass and polypropylene.                                                                   | Jawaid and Abdul Khalil, 2011                      |
|                                           | Composites                  | Fibre           | Past studies that considered fibre-reinforced composites such as jute/polylactic acid, jute/polyester.                                                      | Shalwan and Yousif, 2013                           |
|                                           | Insulation materials        | Fibre           | A past study that evaluated jute fibres properties for thermal insulation materials.                                                                        | Nguyen et al., 2020                                |
| Olive ( <i>Olea europaea</i> L.)          | Plasters                    | Fibre           | Analysis of clay plasters with olive fibres.                                                                                                                | Liuzzi et al., 2018                                |
|                                           | Mortars                     | Stone           | Viability of replacing clay with olive stones for lightweight expanded clay mortars.                                                                        | del Río Merino et al., 2017                        |
|                                           | Bricks                      | Pomace          | A past study that evaluated the effect of adding olive pomace in porous bricks.                                                                             | Ozturk et al., 2019                                |
|                                           | Bricks                      | Husk            | A past study that evaluated thermo-mechanical properties of clay bricks with unburnt olive husk.                                                            | Madurwar et al., 2013                              |
|                                           | Boards<br>Ceramic materials | Fibre<br>Pomace | Boards made of sodium silicate, olive waste and barely straw.<br>A past study that evaluated the thermal properties of ceramic materials with olives waste. | Liuzzi et al., 2020<br>Eliche-Quesada et al., 2011 |
| Grape ( <i>Vitis</i> spp.)                | Glues                       | Pomace          | Feasibility of extracting tannins from wine production waste to produce a natural glue.                                                                     | Cardoso et al., 2015                               |
|                                           | Bricks                      | Pomace          | Fried clay bricks with different percentages of dried wine waste.                                                                                           | Taurino et al., 2019                               |
|                                           | Boards                      | Waste           | Particleboards made up of softwood chip, grapevine chip and melamine modified urea-formaldehyde (MUF).                                                      | Wong et al., 2020                                  |
| Soy ( <i>Glycine max</i> (L.) Merr.)      | Glues                       | Seed            | Past studies that considered soy flour as natural glue for wood.                                                                                            | Cardoso et al., 2015                               |
|                                           | Glues                       | Seed            | Past studies that used soy protein resin.                                                                                                                   | Dittenber and GangaRao, 2012                       |
|                                           | Binders                     | Protein         | Past studies that demonstrated soy protein adhesive properties.                                                                                             | Monteiro et al., 2016                              |
| Sunflowers ( <i>Helianthus annuus</i> L.) | Glues                       | Seed            | Past studies that considered fibreboards composed of natural fibres and bio-glues (kraft lignin and soybean protein).                                       | Nguyen et al., 2017                                |
|                                           | Boards                      | Piths           | A past study that evaluated properties of particleboards made of sunflowers pith with different density and grain size.                                     | Asdrubali et al., 2015                             |
|                                           | Boards                      | Plant           | Particleboards realized by a cake produced by drying sunflowers and thermo-pressing.                                                                        | Evon et al., 2012                                  |

| Lignocellulosic bio-wastes                | Designed product | Part of waste | Description*                                                                                                                         | References                  |
|-------------------------------------------|------------------|---------------|--------------------------------------------------------------------------------------------------------------------------------------|-----------------------------|
|                                           | Boards           | Stalk         | Boards made up of miscanthus fibres, sunflower stalk and natural binders (starch-based binder, wood glue made of casein, bone glue). | Eschenhagen et al., 2019    |
|                                           | Boards           | Stem          | Properties of particleboards made of flax shiv, sunflower stems and urea-formaldehyde as a binder.                                   | Mahieu et al., 2019         |
| Tea ( <i>Camellia sinensis</i> L. Kuntze) | Bricks           | Waste         | A past study that evaluated clay brick with tea waste.                                                                               | Eliche-Quesada et al., 2011 |
|                                           | Bricks           | Waste         | Past studies that considered the feasibility of adding tea waste in clay-based bricks.                                               | Ozturk et al., 2019         |
|                                           | Plasters         | Plant         | Plaster made of hemihydrate gypsums and black tea powder.                                                                            | Huang et al., 2019          |
|                                           | Bricks           | Waste         | Properties of bricks with 5% of waste tea (by weight), dried and fired.                                                              | Raut et al., 2011           |

\*Note: The description section reports "past study/past studies" when the cited articles refer to other past research work.

## 2.2.4 Bio-wastes derived from agricultural practices and agro-industrial processes

Taking into consideration the classification provided in Section 2.2.2 (Tables 2.2-1, 2.2-2 and 2.2-3), this study considers in detail only the bio-wastes derived from agriculture practices and agro-industrial processes used to produce building products. It does not carry out a detailed study of bio-wastes deriving from forest cleaning and floriculture. Past research and results are reported below.

### 2.2.4.1 Bio-wastes derived from agricultural practices

Agriculture generates a large number of bio-wastes to ensure crop growth or harvesting, which are often unused, but they can be an important resource for construction. Past research demonstrated the possibility of using these bio-wastes for several applications, considering both raw materials and composites. There are several types of building products made up of residues of agriculture practices. As examples of the application of the raw materials, D'Alessandro et al. (2017) investigated the thermal and acoustic behaviour of a straw bale wall, carrying out laboratory tests and *in situ* analysis. Experimental results demonstrated good thermal performances: thermal conductivity values of the samples with a density of  $80 \text{ kg/m}^3$  were between  $0.050 \text{ W/(m.K)}$  and  $0.060 \text{ W/(m.K)}$ , thus lower than the maximum recommended for thermal insulators, i.e.  $0.065 \text{ W/(m.K)}$  (AFNOR, 1983). *In situ* measurements demonstrated that straw walls work well in winter conditions but not in summer, due to the high diffusivity and low thermal inertia. In terms of acoustic behaviour, this bio-waste wall did not show great performance, due to lack of mass. Madurwar et al. (2013), Asdrubali et al. (2015) and Liu et al. (2017) reported studies that evaluated thermal behaviour of boards comprising cotton stalk as the fibre component without adding any chemical binders. They had good thermal performance, resulting from a low thermal conductivity. Regarding the use of bio-wastes to produce cement mortars, Akinyemi and Dai (2020) considered treated banana fibres, wood ash and styrene-butadiene polymer. The researchers evaluated mechanical and thermal properties demonstrating that the composite could be used to produce cement elements. Collet and Pretot (2014) analysed the thermal properties of hemp concrete mixing hemp shiv and hydraulic lime. They concluded that thermal performances varied due to the formulation, density and water content. Nunes et al. (2021) studied the properties of cement-bonded particleboards produced by replacing maritime pine particles with banana pseudostem. The researchers observed that increasing the banana fibres (from 0 to 75%) led to higher bulk density (from  $1754 \text{ kg/m}^3$  to  $1995 \text{ kg/m}^3$ ) and thermal conductivity (from  $0.233 \text{ W/(m.K)}$  to  $0.279 \text{ W/(m.K)}$ ).

#### 2.2.4.2 Bio-wastes derived from agro-industrial processes

Another source of bio-waste is the use of agricultural products in industrial processes and their consumption. Several studies investigated their properties, encouraging their reuse. For example, Huang et al. (2019) investigated the performances of a composite of tea waste powder and hemihydrate gypsum to produce plastering paste. The tea waste addition determined higher density and longer setting times. Density increased from  $2.14 \text{ g/cm}^3$  (for the control sample, with no tea waste) to  $2.69 \text{ g/cm}^3$  by adding 2.5% in mass of tea waste. Initial and final setting times increased from 15 min and 22 min (control sample) to 840 min and 3500 min (sample with 2.5% in mass of tea waste), respectively. Furthermore, Huang et al. (2019) reported that tea waste acted as a water-reducing agent: to obtain the normal consistency of gypsum paste, the water requirement decreased by increasing tea waste content. Different results were obtained by Ozturk et al. (2019), in their evaluation of the properties of tea wastes for the production of fired clay brick. The increase of waste content by 12.5% by weight determined high porosity (up to 56.5%) and water absorption (from 15.9% to 35.3%) of the brick samples. Furthermore, the addition of 12.5% of tea waste reduced bulk density (from  $1.79 \text{ g/cm}^3$  to  $1.38 \text{ g/cm}^3$  at  $950^\circ\text{C}$  firing temperature and from  $1.88 \text{ g/cm}^3$  to  $1.41 \text{ g/cm}^3$  at  $1050^\circ\text{C}$ ) and thermal conductivity (up to 42%) and guaranteed sufficient compressive strength, considering that required for bricks, i.e. 7 MPa (CEN, 2011). Liuzzi et al. (2018) assessed clay-based plaster with different percentages of olive pruning waste fibres (from 4% to 12% by the total weight). They verified that increasing fibre content resulted in a lower density (from  $1669 \text{ kg/m}^3$  to  $1409 \text{ kg/m}^3$ ) and thermal conductivity (from  $0.593 \text{ W/(m.K)}$  to  $0.428 \text{ W/(m.K)}$ ) and higher porosity. Hence, adding this natural waste improved thermal insulation performance. Past research also considered the use of coffee grounds to produce different building materials. Muñoz Velasco et al. (2016) used this bio-waste as aggregates to produce fired clay brick. They demonstrated that it improved thermal performances since thermal conductivity reduced from  $0.73 \text{ W/(m.K)}$  (for bricks without coffee grounds) to  $0.38 \text{ W/(m.K)}$ .

#### 2.2.4.3 World production and consumption

Production and transport of building materials entail energy wastes and environmental impacts. Choosing local products reduces this problem, enhancing the characteristic of the considered area. This is the primary aim of using bio-wastes for building products. World production, major producers and major consumers are useful information to make sustainable choices. Depending on the country or region, a specific bio-waste can be selected and used.

Table 2.2-4 shows the ranking of the production in 2019 of agricultural products already studied for building products in decreasing order of world production volume and five major producing countries according to FAOSTAT (FAO, 2020a). Table 2.2-4 also reports data provided by Atlas Big

(2020) as an additional source of information. The production from Euro-Mediterranean Countries is highlighted in grey shade.

Table 2.2-4. World production of agricultural products in 2019 (adapted from FAO (2020a) and Atlas Big (2020).

| Material                                  | FAOSTAT (2020)                                 |                                                                   |                                        | Atlas Big (2020)                                        |
|-------------------------------------------|------------------------------------------------|-------------------------------------------------------------------|----------------------------------------|---------------------------------------------------------|
|                                           | World production<br>[x 10 <sup>5</sup> tonnes] | Five major producers and<br>production [x 10 <sup>5</sup> tonnes] | Land used - Coun-<br>try area [1000ha] | World production per<br>year [x 10 <sup>5</sup> tonnes] |
| Sugar<br>cane                             | 19498.85                                       | Brazil                                                            | 7528.95                                | 18892.69                                                |
|                                           |                                                | India                                                             | 4054.16                                |                                                         |
|                                           |                                                | Thailand                                                          | 1310.02                                |                                                         |
|                                           |                                                | China                                                             | 1099.63                                |                                                         |
|                                           |                                                | Pakistan                                                          | 668.80                                 |                                                         |
| Maize                                     | 11571.05                                       | United States<br>of America                                       | 3470.48                                | 10602.48                                                |
|                                           |                                                | China                                                             | 2609.58                                |                                                         |
|                                           |                                                | Brazil                                                            | 1011.39                                |                                                         |
|                                           |                                                | Argentina                                                         | 568.61                                 |                                                         |
|                                           |                                                | Ukraine*                                                          | 358.80                                 |                                                         |
| Wheat<br>( <i>Triti-<br/>cum</i><br>spp.) | 7657.74                                        | China                                                             | 1336.01                                | 7494.68                                                 |
|                                           |                                                | India                                                             | 1035.96                                |                                                         |
|                                           |                                                | Russian Fed-<br>eration                                           | 744.53                                 |                                                         |
|                                           |                                                | United States<br>of America                                       | 522.58                                 |                                                         |
|                                           |                                                | France                                                            | 406.05                                 |                                                         |
| Rice                                      | 5050.96                                        | China                                                             | 1410.07                                | 7425.42                                                 |
|                                           |                                                | India                                                             | 1184.89                                |                                                         |
|                                           |                                                | Indonesia                                                         | 364.21                                 |                                                         |
|                                           |                                                | Bangladesh                                                        | 364.09                                 |                                                         |
|                                           |                                                | Viet Nam                                                          | 289.80                                 |                                                         |
| Palm oil<br>( <i>Elaeis</i><br>spp).      | 4106.97                                        | Indonesia                                                         | 2456.33                                | 588.00                                                  |
|                                           |                                                | Malaysia                                                          | 990.65                                 |                                                         |
|                                           |                                                | Thailand                                                          | 167.72                                 |                                                         |
|                                           |                                                | Nigeria                                                           | 100.25                                 |                                                         |
|                                           |                                                | Colombia                                                          | 83.90                                  |                                                         |
| Banana                                    | 1171.23                                        | India                                                             | 304.60                                 | 1132.12                                                 |
|                                           |                                                | China                                                             | 119.98                                 |                                                         |
|                                           |                                                | Indonesia                                                         | 72.81                                  |                                                         |
|                                           |                                                | Brazil                                                            | 68.13                                  |                                                         |
|                                           |                                                | Ecuador                                                           | 65.83                                  |                                                         |
| Seed<br>cotton                            | 825.89                                         | China                                                             | 235.05                                 | 261.73                                                  |
|                                           |                                                | India                                                             | 185.50                                 |                                                         |
|                                           |                                                | United States<br>of America                                       | 129.56                                 |                                                         |
|                                           |                                                | Brazil                                                            | 68.93                                  |                                                         |
|                                           |                                                | Pakistan                                                          | 44.95                                  |                                                         |
| Grapes                                    | 770.62                                         | China                                                             | 143.72                                 | 775.18                                                  |
|                                           |                                                | Italy                                                             | 79.00                                  |                                                         |
|                                           |                                                | United States<br>of America                                       | 62.33                                  |                                                         |

| Material                                                         | FAOSTAT (2020)                                 |                                                                   |        | Atlas Big (2020)                       |                                                         |
|------------------------------------------------------------------|------------------------------------------------|-------------------------------------------------------------------|--------|----------------------------------------|---------------------------------------------------------|
|                                                                  | World production<br>[x 10 <sup>5</sup> tonnes] | Five major producers and<br>production [x 10 <sup>5</sup> tonnes] |        | Land used - Coun-<br>try area [1000ha] | World production per<br>year [x 10 <sup>5</sup> tonnes] |
| Coconut                                                          | 624.74                                         | Spain                                                             | 57.45  | 50595.73                               | 583.53                                                  |
|                                                                  |                                                | France                                                            | 54.90  | 54908.70                               |                                                         |
|                                                                  |                                                | Indonesia                                                         | 171.29 | 191686.22                              |                                                         |
|                                                                  |                                                | Philippines                                                       | 147.65 | 30000.00                               |                                                         |
|                                                                  |                                                | India                                                             | 146.82 | 328725.90                              |                                                         |
|                                                                  |                                                | Sri Lanka                                                         | 24.69  | 6561.00                                |                                                         |
| Sun-<br>flower<br>seed                                           | 560.73                                         | Brazil                                                            | 23.31  | 851577.00                              | 473.47                                                  |
|                                                                  |                                                | Russian Fed-<br>eration                                           | 153.79 | 1709825.00                             |                                                         |
|                                                                  |                                                | Ukraine                                                           | 152.54 | 60355.00                               |                                                         |
|                                                                  |                                                | Argentina                                                         | 38.26  | 278040.00                              |                                                         |
|                                                                  |                                                | Romania                                                           | 35.69  | 23840.00                               |                                                         |
|                                                                  |                                                | China                                                             | 24.20  | 960001.29                              |                                                         |
| Pineap-<br>ples<br><i>Ananas<br/>como-<br/>sus</i> (L.)<br>Merr. | 286.10                                         | Costa Rica                                                        | 33.28  | 5110.00                                | 262.90                                                  |
|                                                                  |                                                | Philippines                                                       | 27.48  | 30000.00                               |                                                         |
|                                                                  |                                                | Brazil                                                            | 24.27  | 851577.00                              |                                                         |
|                                                                  |                                                | Indonesia                                                         | 21.96  | 191686.22                              |                                                         |
|                                                                  |                                                | China                                                             | 21.59  | 960001.29                              |                                                         |
|                                                                  |                                                |                                                                   |        |                                        |                                                         |
| Olives                                                           | 194.67                                         | Spain                                                             | 59.65  | 50595.73                               | 192.70                                                  |
|                                                                  |                                                | Italy                                                             | 21.94  | 30207.00                               |                                                         |
|                                                                  |                                                | Morocco                                                           | 19.12  | 44655.00                               |                                                         |
|                                                                  |                                                | Turkey                                                            | 15.25  | 78535.00                               |                                                         |
|                                                                  |                                                | Greece                                                            | 12.28  | 13196.00                               |                                                         |
|                                                                  |                                                |                                                                   |        |                                        |                                                         |
| Coffee                                                           | 100.36                                         | Brazil                                                            | 30.09  | 851577.00                              | 92.18                                                   |
|                                                                  |                                                | Viet Nam                                                          | 16.84  | 33123.00                               |                                                         |
|                                                                  |                                                | Colombia                                                          | 8.85   | 114175.00                              |                                                         |
|                                                                  |                                                | Indonesia                                                         | 7.61   | 191686.22                              |                                                         |
|                                                                  |                                                | Ethiopia                                                          | 4.83   | 113625.94                              |                                                         |
|                                                                  |                                                |                                                                   |        |                                        |                                                         |
| Tea                                                              | 65.12                                          | China                                                             | 27.92  | 960001.29                              | 59.66                                                   |
|                                                                  |                                                | India                                                             | 13.90  | 328725.90                              |                                                         |
|                                                                  |                                                | Kenya                                                             | 4.59   | 58037.00                               |                                                         |
|                                                                  |                                                | Sri Lanka                                                         | 3.00   | 6561.00                                |                                                         |
|                                                                  |                                                | Viet Nam                                                          | 2.69   | 33123.00                               |                                                         |
|                                                                  |                                                |                                                                   |        |                                        |                                                         |
| Jute<br><i>Corcho-<br/>rus</i> spp.)                             | 33.76                                          | India                                                             | 17.09  | 328725.90                              | 33.92                                                   |
|                                                                  |                                                | Bangladesh                                                        | 16.00  | 14757.00                               |                                                         |
|                                                                  |                                                | China                                                             | 0.30   | 960001.29                              |                                                         |
|                                                                  |                                                | Uzbekistan                                                        | 0.15   | 44892.40                               |                                                         |
|                                                                  |                                                | Nepal                                                             | 0.11   | 14718.00                               |                                                         |
|                                                                  |                                                |                                                                   |        |                                        |                                                         |
| Hazel-<br>nuts                                                   | 11.25                                          | Turkey                                                            | 7.76   | 78535.00                               | 7.43                                                    |
|                                                                  |                                                | Italy                                                             | 0.99   | 30207.00                               |                                                         |
|                                                                  |                                                | Azerbaijan                                                        | 0.54   | 8660.00                                |                                                         |
|                                                                  |                                                | United States<br>of America                                       | 0.40   | 983151.00                              |                                                         |
|                                                                  |                                                | Chile                                                             | 0.35   | 75670.00                               |                                                         |
|                                                                  |                                                |                                                                   |        |                                        |                                                         |
| Flax                                                             | 10.86                                          | France                                                            | 8.50   | 54908.70                               | 8.73                                                    |
|                                                                  |                                                | Belgium                                                           | 0.94   | 3053.00                                |                                                         |
|                                                                  |                                                | Belarus                                                           | 0.46   | 20760.00                               |                                                         |

| Material                                                                 | FAOSTAT (2020)                                 |                                                                   |                                        | Atlas Big (2020)                                        |
|--------------------------------------------------------------------------|------------------------------------------------|-------------------------------------------------------------------|----------------------------------------|---------------------------------------------------------|
|                                                                          | World production<br>[x 10 <sup>5</sup> tonnes] | Five major producers and<br>production [x 10 <sup>5</sup> tonnes] | Land used - Coun-<br>try area [1000ha] | World production per<br>year [x 10 <sup>5</sup> tonnes] |
| Sisal<br>( <i>Agave<br/>sisalana</i><br>Perrine<br>ex<br>Engel-<br>mann) | 2.07                                           | Russian Fed-<br>eration                                           | 0.38                                   | 1709825.00                                              |
|                                                                          |                                                | China                                                             | 0.18                                   | 960001.29                                               |
|                                                                          |                                                | Brazil                                                            | 0.87                                   | 2.81                                                    |
|                                                                          |                                                | United Re-<br>public of<br>Tanzania                               | 0.33                                   |                                                         |
|                                                                          |                                                | Kenya                                                             | 0.21                                   |                                                         |
|                                                                          |                                                | Madagascar                                                        | 0.18                                   |                                                         |
|                                                                          |                                                | Haiti                                                             | 0.14                                   |                                                         |
| Hemp                                                                     | 1.74                                           | France                                                            | 0.78                                   | -                                                       |
|                                                                          |                                                | Democratic<br>People's Re-<br>public of Ko-<br>rea                | 0.15                                   |                                                         |
|                                                                          |                                                | China                                                             | 0.15                                   |                                                         |
|                                                                          |                                                | Netherlands                                                       | 0.14                                   |                                                         |
|                                                                          |                                                | Poland                                                            | 0.14                                   |                                                         |
|                                                                          |                                                |                                                                   |                                        |                                                         |

Note: No data is presented when it is not provided by the considered reference.

The three major agricultural products are sugar cane, maize and wheat. Considering only the Euro-Mediterranean Countries, Italy is one of the largest producers of grapes, hazelnuts and olives. Spain is the world's largest producer of olives and the fifth largest producer of grapes. Greece is among the fifth largest producers of olives. France is a major producer of flax, hemp and among the main producers of wheat and grapes. These productions and countries are marked in grey in Table 2.2-4.

For the region considered in this article, consumption and use are hereby assessed in more detail. According to FAOSTAT (FAO, 2020b), Spain, Italy and Greece are the three major producers of olive oil and olives, hence great producers of olives residues; Portugal is among the top ten world producers. Italy, Spain and France are the three major producers of wine and therefore the major grape users; Portugal and Greece are among the first fifteen countries. According to INC Statistical Yearbook (INC, 2020), Italy is not only one of the largest producers but also the top consumer of hazelnut in the world. In addition, world consumption is increasing every year. The International Coffee Organization (ICO, 2020) gives information about coffee consumption by importing countries in 2013, the last year for which there is a large amount of comparable information. France, Italy and Spain are some of the major consumers. Therefore, coffee bio-wastes derived from its use are considered rather than its industrial production (coffee grounds).

These results allow choosing for further research four agricultural products as important sources of bio-waste in Euro-Mediterranean Countries: grapes, olives, and hazelnuts due to their production and coffee because of its consumption. Using them guarantees a sustainable practice ensuring a reduction in costs and consumption derived from production and transport.

#### 2.2.4.4 Chemical composition

Agro-industrial wastes considered in this work are vegetal products. Even if they have different origins, they have similar characteristics such as chemical composition. Cellulose, hemicelluloses and lignin are the main structural components of vegetal fibres and agro-products (Summerscales et al., 2010; Faruk et al., 2012; Pedras et al., 2020), with other natural substances, such as extractives, pectin, waxy substances like suberin or cutin and ash. Suberin is present in most wood barks and mainly in cork-rich barks such as *Quercus suber* L., *Q. cerris* L. or *Pseudotsuga menziesii* (Mirb.) Franco (Graça and Pereira, 1999; Lopes et al., 2000; Esteves et al., 2017; Sen et al., 2020). Cutin is the major component of the cuticle, a hydrophobic layer that covers the aerial epidermis of all land plants, providing protection against desiccation and external environmental stresses (Yeats and Rose, 2013). Each of the components plays a specific role. They vary depending on the plant and are different in the distinct parts of the same plant. The components depend on several factors: the plant and its age, the location and climatic context, the soil conditions, harvesting, the extraction and the degradation process (Jawaid and Abdul Khalil, 2011).

Cellulose is a linear homopolymer that can be represented by  $[C_6(H_2O)_5]_n$ , where  $n$  represents the degree of polymerization that can reach 10 000 or higher (Pan, 2011). It has a partially crystalline structure that guarantees hydrophobic behaviour (Mohanty et al., 2005; John and Thomas, 2008; Jawaid and Abdul Khalil, 2011; Summerscales et al., 2010). It is an insoluble polysaccharide composed of fermentable sugar and is formed by repeating units of  $\beta$ -D-glucopyranose linked, by glycosidic bonds. For this reason, glucose is the product of its degradation (Singh Nee Nigam and Pandey, 2009; Nguyen et al., 2017; Echeverria and Nuti, 2017). Cellulose is present in large quantities on earth and it is the main component of the vegetable cell walls, providing structural support to them. Pre-treatments to prepare cellulose are normally costly and harmful (John and Thomas, 2008; Summerscales et al., 2010; Mikkonen and Tenkanen, 2012; Madurwar et al., 2013; Lei and Feng, 2020).

Hemicellulose is the second most widespread group of polysaccharides on earth (Ramos et al., 2017). Hemicelluloses are made up of different types of fermentable sugars: copolymers of glucose, glucuronic acid, mannose, arabinose, galactose and xylose, cross-linked with cellulose with hydrogen bonds (Yang et al., 2007; Summerscales et al., 2010; Ramage et al., 2017). Hemicelluloses have an amorphous linear structure with a lower degree of polymerization than cellulose. They are major constituents in plants and are extremely important in their structure, acting as a filler for the cellular network connecting cellulose and lignin (Singh Nee Nigam and Pandey, 2009; Mikkonen and Tenkanen, 2012; Ramos et al., 2017).

Lignin is a three-dimensional, aromatic polymer consisting of phenylpropane units, joined together, by carbon-carbon and carbon-oxygen-carbon bridging bonds (Mahmood et al., 2016). Its formula can be represented by  $[C_{10}H_{12}O_3]_n$  (Pan, 2011). It is amorphous and it has a complex structure composed of three different monomers derived from p-coumaryl, coniferyl, and sinapyl alcohols



(Lourenço et al., 2016). It is not soluble in several solvents, and it cannot be divided into monomeric units. Comparing with cellulose and hemicelluloses, it is the most complex compound to decompose. There is still not an exact method to isolate lignin in its native state from the fibre (John and Thomas, 2008; Nguyen et al., 2017). It provides rigidity to the cell walls of plants and works as structural support, even if cellulose plays a more important role in this task (John and Thomas, 2008; Jawaid and Abdul Khalil, 2011; Dittenber and GangaRao, 2012). It provides covalent linking into the cell wall, ensuring mechanical strength to the cell wall and therefore to the whole plant. It has good resistance to water and microorganism attack (Singh Nee Nigam and Pandey, 2009).

Table 2.2-5 provides information about the percentages of the main macromolecular compounds in lignocellulosic materials; cellulose, hemicelluloses, lignin and waxes but also ash content for agricultural products analysed in past research and considered earlier (Table 2.2-4). Since each of those components determines different properties in building elements, knowing the chemical composition allows important considerations to be made. A significant amount of research has been published in this field, but this review has considered and reported only past studies published between 2000-2020. Poletto et al. (2014) analysed natural fibres and reported that chemical composition influences their physical and mechanical features. Larger quantities of hemicelluloses cause higher moisture absorption and desorption, granting a higher moisture buffering capacity (Cascione et al 2019) and degradation at lower temperatures. Fibres with high percentages of crystalline cellulose have higher thermal stability and tensile properties. This also influences water absorption, as indicated by Cabral et al. (2018). Panyakaew and Fotios (2011) reported that lignin can be used as a natural resin to produce particleboards and that cellulose fibres can give higher stability when used as a filler in composites. Zhou et al. (2010) indicated that lignin and cellulose are the two major compounds used to manufacture binderless fibreboards, thus avoiding the use of synthetic binders such as formaldehyde-based resins that are considered as a potentially harmful solution due to volatile organic compounds released in service. Hot pressing treatment secures this result, but it also produces boards with high densities, not optimal for thermal insulation, although with higher mechanical properties. Cardoso et al. (2015) considered lignin as an attractive material to produce natural glues for wood; however, they say that it is difficult to reach efficient results if compared with conventional binders. Hemmilä et al. (2017) described the viability of using lignin to produce adhesive materials, reporting several examples. Younesi-Kordkheili and Pizzi (2020) used lignin as an additive to obtain a natural wood adhesive, demonstrating the feasibility of using it for panel production. Owodunni et al. (2020a) studied the possibility of producing coconut particleboards using natural binders and concluded that high content of lignin enhanced the bonding process. Improving the understanding of the relationship between properties and cellulose, hemicelluloses and lignin content is an interesting topic for future research. A more detailed version of Table 2.2-5 is given as supplementary material.

Table 2.2-5. Chemical composition of several bio-wastes considering research works published in the last twenty years (2000-2020).

| Material                    | Chemical composition [%] |                |             |       |           | References                                                                                                                                                                                                                           |
|-----------------------------|--------------------------|----------------|-------------|-------|-----------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
|                             | Cellulose                | Hemicelluloses | Lignin      | Waxes | Ash       |                                                                                                                                                                                                                                      |
| Banana (range)              | 60.00-65.00              | 10.00-19.00    | 5.00-10.00  | -     | -         | (Idicula et al., 2006; Jawaid and Abdul Khalil, 2011; Dittenber and GangaRao, 2012; Nguong et al., 2013)                                                                                                                             |
| Coconut fibres/coir (range) | 21.00-64.00              | 0.15-67.63     | 19.40-47.00 | -     | 0.74      | (Khedari et al., 2004; Phan et al., 2006; Jawaid and Abdul Khalil, 2011; Panyakaew and Fotios, 2011; Faruk et al., 2012;; Dittenber and GangaRao, 2012; Nascimento et al., 2016; García et al., 2016; Laborel-Préneron et al., 2016) |
| Coffee chaff (range)        | 13.00-24.00              | 11.00-19.30    | 17.80-32.00 | -     | 5.94-6.90 | (Zarrinbakhsh et al., 2016; Buratti et al., 2018; Quosai et al., 2018)                                                                                                                                                               |
| Coffee grounds (range)      | 8.60-52.42               | 6.80-44.50     | 18.30-24.52 | -     | 0.98-2.52 | (Muñoz Velasco et al., 2016; Zarrinbakhsh et al., 2016; Buratti et al., 2018; Ribeiro et al., 2018; Lachheb et al., 2019; Pedras et al., 2019)                                                                                       |
| Corn cobs/ Maize (range)    | 33.70-48.10              | 31.90-44.40    | 3.30-30.30  | -     | 0.46-2.88 | (García et al., 2016; Viel et al., 2018; Viel et al., 2019; Cárdenas, 2020)                                                                                                                                                          |
| Corn cob/ Maize residues    | 27.10                    | 13.87          | 0.64        | -     | 0.43      | (Viel et al., 2019)                                                                                                                                                                                                                  |
| Corn/Maize stalks           | 35.00-39.60              | 16.80-35.00    | 7.00-18.40  | -     | -         | (Cárdenas, 2020)                                                                                                                                                                                                                     |
| Corn stover                 | 47.40                    | 30.30          | 22.30       | -     | -         | (García et al., 2016)                                                                                                                                                                                                                |
| Cotton (range)              | 58.48-90.00              | 5.70-42.50     | 2.00-21.45  | 0.60  | 5.54      | (Jawaid and Abdul Khalil, 2011; Pirayesh and Khazaeian, 2012; Dittenber and GangaRao, 2012; Nguong et al., 2013; García et al., 2016; Cárdenas, 2020)                                                                                |
| Cotton stalk (range)        | 32.00-66.20              | 18.40-28.00    | 15.40-26.00 | -     | 8.16      | (García et al., 2016; Nguyen et al., 2017)                                                                                                                                                                                           |

| Material                    | Chemical composition [%] |                |             |           |           | References                                                                                                                                                                                                                                  |
|-----------------------------|--------------------------|----------------|-------------|-----------|-----------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
|                             | Cellulose                | Hemicelluloses | Lignin      | Waxes     | Ash       |                                                                                                                                                                                                                                             |
| Flax (range)                | 28.51-81.00              | 13.00-27.00    | 2.00-31.20  | 1.50-1.70 | 1.48-4.20 | (Jawaid and Abdul Khalil, 2011; Faruk et al., 2012; Dittenber and GangaRao, 2012; Nguong et al., 2013; Papadopoulou et al., 2015; García et al., 2016; Laborel-Préneron et al., 2016; Viel et al., 2018; Viel et al., 2019; Cárdenas, 2020) |
| Flax fine                   | 28.51                    | 15.80          | 18.14       | -         | 4.20      | (Viel et al., 2018)                                                                                                                                                                                                                         |
| Flax fine residues          | 16.21                    | 5.83           | 7.74        | -         | 2.92      | (Viel et al., 2019)                                                                                                                                                                                                                         |
| Grapes waste pomace (range) | 15.00-50.16              | 6.62-11.30     | 30.30       | -         | 3.70-7.00 | (Barbieri et al., 2013; Muñoz et al., 2014; Pedras et al., 2020)                                                                                                                                                                            |
| Hazelnuts (range)           | 55.10                    | 34.50-34.60    | 35.10-41.40 | -         | 8.22-8.23 | (Çöpür et al., 2007; Pirayesh and Khazaeian, 2012)                                                                                                                                                                                          |
| Hazelnut shells (range)     | 22.90-34.60              | 11.30-23.50    | 30.20-41.40 | -         | 1.40-8.22 | (Çöpür et al., 2007; Guler et al., 2008; Lopes et al., 2012; Salasinska and Ryszkowska, 2012; Licursi et al., 2017)                                                                                                                         |
| Hemp (range)                | 44.00-78.30              | 5.50-22.40     | 2.90-28.00  | 0.80      | 0.67-8.80 | (Summerscales et al., 2010; Jawaid and Abdul Khalil, 2011; Dittenber and GangaRao, 2012; Shalwan and Yousif, 2013; Nguong et al., 2013; Papadopoulou et al., 2015; Laborel-Préneron et al., 2016; Viel et al., 2018)                        |
| Jute (range)                | 33.40-72.00              | 12.00-22.70    | 11.80-28.00 | 0.50      | 0.62      | (Phan et al., 2006; Jawaid and Abdul Khalil, 2011; Faruk et al., 2012; Dittenber and GangaRao, 2012; Madurwar et al., 2013; Nguong et al., 2013; Laborel-Préneron et al., 2016)                                                             |
| Kenaf (range)               | 31.00-72.00              | 3.00-33.90     | 8.00-21.20  | -         | 4.00      | (Jawaid and Abdul Khalil, 2011; Faruk et al., 2012; Dittenber and GangaRao, 2012; Nguong et al., 2013; Papadopoulou et al., 2015; Laborel-Préneron et al., 2016)                                                                            |
| Palm oil (range)            | 49.00-65.00              | 21.00          | 11.00-29.00 | -         | 2.00      | (Jawaid and Abdul Khalil, 2011; Dittenber and GangaRao, 2012; Laborel-Préneron et al., 2016)                                                                                                                                                |

| Material                    | Chemical composition [%] |                |             |             |           | References                                                                                                                                                                         |
|-----------------------------|--------------------------|----------------|-------------|-------------|-----------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
|                             | Cellulose                | Hemicelluloses | Lignin      | Waxes       | Ash       |                                                                                                                                                                                    |
| Palm oil frond              | 56.03                    | 27.51          | 20.48       | -           | 2.40      | (Jawaid and Abdul Khalil, 2011)                                                                                                                                                    |
| Olive husks                 | 25.00                    | 24.60          | 50.40       | -           | 14.21     | (García et al., 2016; Christoforou et al., 2017)                                                                                                                                   |
| Olive whole stones          | 31.90                    | 21.90          | 26.50       | -           | -         | (Fernández-Bolaños et al., 1999)                                                                                                                                                   |
| Olive seed husks            | 36.40                    | 26.80          | 26.00       | -           | -         | (Fernández-Bolaños et al., 1999)                                                                                                                                                   |
| Olive pomace                | -                        | -              | -           | -           | 3.20-8.63 | (Christoforou et al., 2017)                                                                                                                                                        |
| Pineapples                  | 81.00                    | -              | 12.70       | -           | -         | (Idicula et al., 2006)                                                                                                                                                             |
| Ramie (range)               | 68.60-85.00              | 13.00-16.70    | 0.50-0.70   | 0.30        | -         | (Jawaid and Abdul Khalil, 2011; Faruk et al., 2012; Dittenber and GangaRao, 2012)                                                                                                  |
| Rice husk (range)           | 30.00-45.00              | 19.00-25.00    | 20.00-33.00 | 14.00-17.00 | 14.00     | (Faruk et al., 2012; Brás et al., 2019)                                                                                                                                            |
| Rice straw (range)          | 36.20-57.00              | 19.00-33.00    | 8.00-24.00  | 8.00-38.00  | -         | (Faruk et al., 2012; García et al., 2016; Cárdenas, 2020)                                                                                                                          |
| Sisal (range)               | 38.20-78.00              | 10.00-26.00    | 8.00-26.00  | 2.00        | -         | (Idicula et al., 2006; Jawaid and Abdul Khalil, 2011; Dittenber and GangaRao, 2012; Faruk et al., 2012; Madurwar et al., 2013; Nguong et al., 2013; Laborel-Préneron et al., 2016) |
| Sugar cane/baggasse (range) | 32.00-55.20              | 16.80-37.50    | 10.00-25.30 | -           | -         | (Faruk et al., 2012; Dittenber and GangaRao, 2012; García et al., 2016; Cárdenas, 2020)                                                                                            |
| Sunflower seed              | 24.10                    | 28.60          | 29.40       | -           | -         | (Cárdenas, 2020)                                                                                                                                                                   |
| Sunflower stalks            | 42.10                    | 29.66          | 13.44       | -           | -         | (Cárdenas, 2020)                                                                                                                                                                   |
| Sunflower whole plant       | 23.93                    | 7.83           | 9.13        | -           | -         | (Evon et al., 2012)                                                                                                                                                                |

| Material               | Chemical composition [%] |                |            |       |           | References                                                            |
|------------------------|--------------------------|----------------|------------|-------|-----------|-----------------------------------------------------------------------|
|                        | Cellulose                | Hemicelluloses | Lignin     | Waxes | Ash       |                                                                       |
| Sunflower shells       | 66.20                    | 18.40          | 15.40      | -     | -         | (García et al., 2016)                                                 |
| Tea residue            | 33.30                    | 23.20          | 43.50      | -     | -         | (García et al., 2016)                                                 |
| Vine shoots            | 41.14                    | 26.00          | 20.27      | -     | -         | (Cárdenas, 2020)                                                      |
| Wheat straw<br>(range) | 32.90-50.00              | 15.00-35.50    | 5.24-20.00 | -     | 0.82-5.90 | (Karade, 2010; Faruk et al., 2012; Viel et al., 2018; Cárdenas, 2020) |

Note: range means the maximum and minimum value reported in the considered studies.

The results reported in Table 2.2-5 show that the highest values of cellulose content are reached by cotton, flax, hemp, ramie and sisal (the average value is 44.16%). Coconut, coffee chaff, corn, cotton and sugarcane reach greater percentages of hemicellulose (average value=21.83%); hence, they might demonstrate higher moisture buffering capacity (Cascione et al., 2019). Coconut fibres, hazelnut shells and tea residues show the highest values of lignin (the average value is 20.09%). Therefore, good bonding capacities could be expected for these materials and also better mechanical strength and better resistance to water and microorganisms' attack.

#### **2.2.4.5 Properties**

The properties of bio-wastes vary depending on their chemical composition, plant species, their production and location, climatic characteristics and harvesting (Dittenber and GangaRao, 2012). By knowing those properties and the effect of specific treatments, it is possible to produce building materials with different performances. Using natural wastes and fibres as building materials have simultaneously several advantages and disadvantages. Past studies demonstrate that they frequently improve thermal insulation and acoustic absorption thanks to their high porosity. Furthermore, they can improve indoor hygrothermal conditions favouring indoor comfort. Since bio-wastes usually have good moisture buffering capacity, they can control relative humidity (Mohanty et al., 2005; Dittenber and GangaRao, 2012; Berardi and Iannace, 2015; Sanjay et al., 2018; Nguyen et al., 2018a; Nguyen et al., 2018b; Liuzzi et al., 2020). In addition, many of them have good mechanical properties, they can improve stiffness and strength and they have a high resistance to fracture (Mohanty et al., 2005; Berardi and Iannace, 2015; Sanjay et al., 2018). Natural wastes and natural fibres are recyclable and biodegradable, and they guarantee better outcomes for the environment and human health in comparison to synthetic ones. Using bio-wastes is a passive technique that ensures healthier architecture and favours a lower energy impact (Ali, 2016; Ali et al., 2020b). As Nguyen et al. (2017) reported, natural wastes can reduce energy waste for heating and cooling and improve indoor air quality by influencing moisture buffering within a building. Rojas et al. (2019) evaluated the thermal performance of insulation materials made up of natural fibres (wheat straw and corn husk). Researchers demonstrated the feasibility of using these natural fibres. Liuzzi et al. (2018) considered a clay-sand plaster with natural fibres showing that by increasing fibres content, both thermal properties and moisture absorption/desorption improved. Better insulation and control of indoor air ensure lower energy consumption to control the indoor conditions. On the other hand, they have a low resistance to water, microorganism attack and fire. Due to their high porosity, natural wastes can have low dimensional stability and low durability. Even if some appropriate treatments can limit these drawbacks, it is important to consider them for the production of building materials. Table 2.2-6 shows a list of some of these advantages and disadvantages.

Table 2.2-6. Advantages and disadvantages of using bio-wastes and natural fibres for the production of eco-efficient construction materials (based on Mohanty et al., 2005; John and Thomas, 2008; Dittenber and GangaRao, 2012; Faruk et al., 2012; Berardi and Iannace, 2015; Nguyen et al., 2017; Liuzzi et al., 2018).

| Advantages                                        | Disadvantages                                                               |
|---------------------------------------------------|-----------------------------------------------------------------------------|
| Renewability, recyclability, and biodegradability | Hydrophilic nature                                                          |
| Sustainable life cycle, low environmental impact  | Degradation at high temperature (problems for pressing at high temperature) |
| Economical production, low cost                   | High moisture and water absorption                                          |
| Non-abrasive                                      | Swelling, dimensional variation                                             |
| Healthy indoor conditions                         | Low durability                                                              |
| Insulation properties                             | Biological susceptibility to moulds                                         |
| Passive indoor environmental control              | Biological susceptibility to insects                                        |
| Low density                                       | Low resistance to fire                                                      |

Knowing the properties of natural wastes and fibres allows making considerations on their performance as building materials. The article considers the most studied properties for thermal insulation composites, according to the bibliographic research: density, thermal conductivity, moisture content, and tensile strength. Information related to water absorption and porosity was also investigated, but these values are not reported due to the identified gap in the literature.

Tables 2.2-7 and 2.2-8 show the results of the analysed studies (published between 2000-2020), making a distinction between raw materials and natural fibres considered alone, and the composite boards. The values change due to the characterization of the fibres, treatment, measurement process and conditions. Table 2.2-7 reports values of thermal conductivity, demonstrating the lack of information. Most of the considered studies did not evaluate the thermal conductivity of the raw materials but only of the resulting composites. This is an important outcome justifying further studies related to raw materials. Table 2.2-7 gives also information about physical characterization describing the length and diameter of natural fibres. These are important to make preliminary consideration of materials performance. Fibre length influences flexural strength, as indicated by Sanjay et al. (2018), as well as their content in the composites, which also affects thermal and mechanical properties. For example, Antunes et al. (2019) evaluated board samples made up of rice husk, earth, gypsum and air lime. They demonstrated that higher percentages of fibres improved the insulation properties, whereas mechanical properties (compressive and flexural strength) decreased by changing the rice husk percentage from 15% to 30%. Liuzzi et al. (2016) studied the thermal properties of blends made up of different percentages of clay, sand, straw and cement that can be used for boards or brick production. The results demonstrated that increasing the amount of straw guarantees better thermal performances.

Table 2.2-7. Properties of natural raw materials considering studies published between 2000-2020. Where details are not provided in the table, they were not reported in the relevant study.

| Materials             | Diameter of fibres [μm] | Length [mm] | Moisture content [%] | Density [kg/m <sup>3</sup> ]                             | Thermal conductivity [W/(m.K)] | Tensile strength [MPa] | References                         |
|-----------------------|-------------------------|-------------|----------------------|----------------------------------------------------------|--------------------------------|------------------------|------------------------------------|
| Banana tree fibres    | 120                     | -           | -                    | 1350                                                     | -                              | 550                    | (Idicula et al., 2006)             |
|                       | 12-30                   | 300-900     | 8.7-12               | 1350                                                     | -                              | 500                    | (Dittenber and GangaRao, 2012)     |
|                       | 80-250                  | -           | -                    | 1350                                                     | -                              | 529-759                | (Sanjay et al., 2018)              |
| Coffee chaff          | -                       | -           | -                    | 70                                                       | -                              | -                      | (Ricciardi et al., 2017)           |
| Coffee grounds        | -                       | -           | -                    | 262-390 <sup>b</sup>                                     | -                              | -                      | (Massaro Sousa and Ferreira, 2019) |
|                       | -                       | -           | -                    | -                                                        | 0.2                            | -                      | (Lachheb et al., 2019)             |
| Coffee (dry berry)    | -                       | -           | 12                   | 422-440 <sup>b</sup>                                     | -                              | -                      | (Echeverria and Nuti, 2017)        |
| Coffee (fresh berry)  | -                       | -           | 65                   | 616-645 <sup>b</sup> ; 616 <sup>b</sup> (wet processing) | -                              | -                      | (Echeverria and Nuti, 2017)        |
| Coffee (green coffee) | -                       | -           | 12                   | 650-750 <sup>b</sup> ; 250 <sup>b</sup> (wet processing) | -                              | -                      | (Echeverria and Nuti, 2017)        |
| Coir/Coconut          | -                       | -           | -                    | 1200                                                     | -                              | 175                    | (Faruk et al., 2012)               |
|                       | 20-150                  | 20-150      | -                    | 1150-1460                                                | -                              | 95-230                 | (Dittenber and GangaRao, 2012)     |
|                       | 100-460                 | -           | -                    | 1150                                                     | -                              | 108-252                | (Sanjay et al., 2018)              |
|                       | 270-2380                | 35-50       | 0.8-3.2              | 1177 <sup>a</sup>                                        | -                              | 73-505                 | (Laborel-Préneron et al., 2016)    |
|                       | -                       | -           | -                    | 1250                                                     | -                              | 220                    | (Jawaid and Abdul Khalil, 2011)    |
|                       | -                       | -           | -                    | 1200                                                     | -                              | 175                    | (Eichhorn et al., 2001)            |
| Cotton                | 10-45                   | 10-60       | 7.85-8.5             | 1500-1600                                                | -                              | 287-800                | (Dittenber and GangaRao, 2012)     |
|                       | -                       | -           | -                    | 1600                                                     | -                              | 287-597                | (Sanjay et al., 2018)              |
|                       | 12-35                   | 15-56       | -                    | 1510                                                     | -                              | 400                    | (Jawaid and Abdul Khalil, 2011)    |
|                       | -                       | -           | -                    | 1500-1600                                                | -                              | 287-597                | (Eichhorn et al., 2001)            |
| Flax                  | -                       | -           | -                    | 1500                                                     | -                              | 345-1035               | (Faruk et al., 2012)               |
|                       | 12-600                  | 5-900       | 8-12                 | 1400-1500                                                | -                              | 343-2000               | (Dittenber and GangaRao, 2012)     |
|                       | -                       | -           | -                    | 1500                                                     | -                              | 345-1500               | (Sanjay et al., 2018)              |
|                       | -                       | -           | 7                    | 1530                                                     | -                              | 1339                   | (Summerscales et al., 2010)        |
|                       | -                       | 33          | -                    | 1541                                                     | -                              | -                      | (Baley, 2002)                      |
|                       | 5-38                    | 10-65       | -                    | 1400                                                     | -                              | 800-1500               | (Jawaid and Abdul Khalil, 2011)    |
|                       | -                       | -           | -                    | 1500                                                     | -                              | 345-1035               | (Eichhorn et al., 2001)            |



| Materials           | Diameter of fibres [μm] | Length [mm] | Moisture content [%] | Density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] | Tensile strength [MPa] | References                      |
|---------------------|-------------------------|-------------|----------------------|------------------------------|--------------------------------|------------------------|---------------------------------|
|                     | 13                      | -           | -                    | -                            | -                              | -                      | (Laborel-Préneron et al., 2016) |
|                     | -                       | -           | -                    | -                            | 0.075                          | -                      | (Benfratello et al., 2013)      |
| Hazelnut husk       | -                       | -           | -                    | 230 *                        | -                              | -                      | (Çöpür et al., 2007)            |
| Hemp                | -                       | -           | -                    | 1480                         | -                              | 690                    | (Faruk et al., 2012)            |
|                     | 25-500                  | 5-55        | 6.2-12               | 1400-1500                    | -                              | 270-900                | (Dittenber and GangaRao, 2012)  |
|                     | -                       | -           | -                    | 1480                         | -                              | 550-900                | (Sanjay et al., 2018)           |
|                     | 35                      | 8.5-17      | -                    | 1500 <sup>a</sup>            | -                              | 900-1077               | (Laborel-Préneron et al., 2016) |
|                     | -                       | -           | -                    | -                            | -                              | 690                    | (Eichhorn et al., 2001)         |
|                     | 10-51                   | 5-55        | -                    | 1480                         | -                              | 550-900                | (Jawaid and Abdul Khalil, 2011) |
|                     | -                       | -           | -                    | -                            | 0.048                          | -                      | (Page et al., 2017)             |
|                     | -                       | -           | -                    | -                            | 0.04                           | -                      | (Romano et al., 2019)           |
|                     | -                       | -           | -                    | -                            | 0.051                          | -                      | (Laborel-Préneron et al., 2018) |
| Jute                | 20-200                  | 1.5-120     | 12.5-13.7            | 1300-1490                    | -                              | 320-800                | (Dittenber and GangaRao, 2012)  |
|                     | -                       | -           | -                    | 1460                         | -                              | 393-800                | (Sanjay et al., 2018)           |
|                     | 1000                    | 20-40       | -                    | 1460-1700 <sup>a</sup>       | -                              | -                      | (Laborel-Préneron et al., 2016) |
|                     | -                       | -           | -                    | 1300                         | -                              | 393-773                | (Faruk et al., 2012)            |
|                     | -                       | -           | -                    | 1300                         | -                              | 393-773                | (Eichhorn et al., 2001)         |
| Kenaf               | 18-37                   | 1.4-11      | -                    | 1200                         | -                              | 295                    | (Jawaid and Abdul Khalil, 2011) |
|                     | -                       | -           | -                    | 1400                         | -                              | 223-930                | (Dittenber and GangaRao, 2012)  |
|                     | 81                      | -           | -                    | 1400                         | -                              | 250                    | (Sanjay et al., 2018)           |
|                     | 130                     | 30          | -                    | 1040 <sup>a</sup>            | -                              | 1000                   | (Laborel-Préneron et al., 2016) |
| Palm oil            | -                       | -           | -                    | 700-1550                     | -                              | 248                    | (Faruk et al., 2012)            |
|                     | 150-500                 | -           | -                    | 700-1550                     | -                              | 80-248                 | (Dittenber and GangaRao, 2012)  |
| Olive pruning waste | -                       | -           | -                    | 1251 <sup>c</sup>            | -                              | -                      | (Liuzzi et al., 2018)           |
|                     | -                       | -           | -                    | -                            | 0.062                          | -                      | (Liuzzi et al., 2020)           |
| Pineapple leaves    | 20-80                   | -           | -                    | 1440                         | -                              | 413-1627               | (Sanjay et al., 2018)           |
|                     | 50                      | -           | -                    | 1526                         | -                              | 413                    | (Idicula et al., 2006)          |
|                     | 20-80                   | 3-9         | -                    | -                            | -                              | -                      | (Jawaid and Abdul Khalil, 2011) |
| Pineapple           | -                       | -           | -                    | 800-1600                     | -                              | 400-627                | (Faruk et al., 2012)            |
|                     | -                       | -           | -                    | 1500                         | -                              | 170-1627               | (Jawaid and Abdul Khalil, 2011) |
| Ramie               | -                       | -           | -                    | 1500                         | -                              | 560                    | (Faruk et al., 2012)            |
|                     | 20-80                   | 900-1200    | 7.5-17               | 1000-1550                    | -                              | 400-1000               | (Dittenber and GangaRao, 2012)  |
|                     | -                       | -           | -                    | 1500                         | -                              | 220-938                | (Sanjay et al., 2018)           |

| Materials            | Diameter of fibres [μm] | Length [mm] | Moisture content [%] | Density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] | Tensile strength [MPa] | References                      |
|----------------------|-------------------------|-------------|----------------------|------------------------------|--------------------------------|------------------------|---------------------------------|
|                      | -                       | -           | -                    | -                            | -                              | 400-938                | (Eichhorn et al., 2001)         |
|                      | 18-80                   | 40-250      | -                    | 1500                         | -                              | 500                    | (Jawaid and Abdul Khalil, 2011) |
| Sisal                | 205                     |             |                      | 1450                         | -                              | 350                    | (Idicula et al., 2006)          |
|                      |                         |             |                      | 1500                         | -                              | 511-635                | (Faruk et al., 2012)            |
|                      | 8-200                   | 900         | 10-22                | 1330-1500                    | -                              | 363-700                | (Dittenber and GangaRao, 2012)  |
|                      | 50-300                  | -           | -                    | 1450                         | -                              | 227-400                | (Sanjay et al., 2018)           |
|                      | 150-300                 | 50-72       | -                    | 1370 <sup>a</sup>            | -                              | 580                    | (Laborel-Préneron et al., 2016) |
|                      | -                       | -           | -                    | 1500                         | -                              | 511-635                | (Eichhorn et al., 2001)         |
| Sugar cane (bagasse) | 10-34                   | 10-300      | -                    | 1250                         | -                              | 222-290                | (Dittenber and GangaRao, 2012)  |
|                      | -                       | -           | -                    | 250-350 <sup>b</sup>         | 0.049-0.055                    | -                      | (Liuzzi et al., 2020)           |
|                      | -                       | -           | -                    | 1250                         | -                              | 290                    | (Faruk et al., 2012)            |
| Wheat straw          | 500-3000                | 15-50       | 280-350              | 868 <sup>a</sup>             | 0.0414-0.0486                  | -                      | (Laborel-Préneron et al., 2016) |

Note: <sup>a</sup> Absolute density; <sup>b</sup> Loose bulk density; <sup>c</sup> Real density; <sup>\*</sup> calculate by water displacement method.

Table 2.2-8. Properties of boards with bio-wastes considering studies published between 2000-2020.

| Material       | Summary of sample preparation                                                                                                                                                                                                                                            | Density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] | References                                     |
|----------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------|--------------------------------|------------------------------------------------|
| Banana bunch   | The raw material was pressed by a roll mill, dried at 40°C and cut in chips (<2mm) exposed to a steam pre-treatment, filtered and rinsed. Fibreboards of 3mm thickness and 1000 kg/m <sup>3</sup> density were produced by pressing treatment without synthetic binders. | 1000                         | -                              | (Quintana et al., 2009; Madurwar et al., 2013) |
| Coconut fibres | Tested specimens were 200mmx400mmx60mm. Thermal conductivity is determined by hotbox apparatus for constant temperatures (T=38°C).                                                                                                                                       | 85                           | 0.058                          | (Ashour et al., 2010)                          |
| Coffee chaff   | Cylindrical samples (diameter=29 mm and 100mm, thickness from 10 mm to 40 mm) bonded by a cold-water-based polyurethane glue (density=1000 kg/m <sup>3</sup> ). Percentage of 5.5% of the total weight.                                                                  | 346.4                        | -                              | (Ricciardi et al., 2017)                       |
|                | Cylindrical samples (diameter=29 mm and 100mm, thickness from 10 mm to 40 mm) exposed to a hot pressure treatment (maximum pressure of 15,000 kg; T= 140 °C).                                                                                                            | 960.45                       | -                              | (Ricciardi et al., 2017)                       |
|                | Cylindrical samples (diameter=29 mm and 100mm, thickness from 10 mm to 40 mm) exposed to a cold pressure treatment (maximum pressure of 15,000 kg).                                                                                                                      | 1047.5                       | -                              | (Ricciardi et al., 2017)                       |

| Material         | Summary of sample preparation                                                                                                                                                                                                                                                                                                                                                                                                                                    | Density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] | References                                                                                                    |
|------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------|--------------------------------|---------------------------------------------------------------------------------------------------------------|
| Corn (Maize) cob | The raw material is granulated and particleboards (250mm x 250 mm x 50 mm) are made with wood glue.                                                                                                                                                                                                                                                                                                                                                              | -                            | 0.139                          | (Pinto et al., 2012)                                                                                          |
|                  | Thermal conductivity is determined by using two heat flux meters and by collecting results for seven days. The temperature was continuously measured (T indoor=23°C). Tested boards are low density and are made using hot pressure treatment and urea-formaldehyde resin.                                                                                                                                                                                       | -                            | 0.096                          | (Panyakaew and Fotios, 2008)                                                                                  |
|                  | Samples (250 mm x 250 mm) with different thickness (30 mm, 50 mm, 60 mm and 80 mm) were considered. Wood glue was used as a binder. The method to produce the samples was: mixing the materials, moulding, natural curing and unmoulding. The thermal conductivity value was evaluated using the heat flux method.<br>*Average values (considering different thicknesses).<br>** Estimated value.                                                                | 334*                         | 0.101**                        | (Paiva et al., 2012)                                                                                          |
| Cotton stalk     | The fibres were immersed in water, exposed to a steam treatment (pressure=1.2 bar), and dried at 100°C. Specimens were produced by pressure treatment using a CGM-30 high-frequency press. The binderless fibreboards were then stored at 25°C and 65±5% RH for one week to have a moisture content of 8%. The thermal conductivity of the samples (300mm x 300mm x 25mm) was evaluated at room temperature.                                                     | 150-450                      | 0.059-0.082                    | (Zhou et al., 2010; Madurwar et al., 2013; Asdrubali et al., 2015; Nguyen et al., 2017; Liuzzi et al., 2020;) |
| Date Palm        | The thermal conductivity of raw material cut out from the petiole or bunch of the plant wood (samples: 44mm x 44mm and thicknesses between 4.2mm-5mm) is determined by using a periodic method at room temperature.                                                                                                                                                                                                                                              | 187-389                      | 0.083                          | (Agoudjil et al., 2011)                                                                                       |
| Hazelnut husk    | Particles between 0.8 mm-1.5 mm were dried to 3% moisture content at 100–110°C and bonded by urea-formaldehyde, phenol-formaldehyde and melamine-urea formaldehyde. Ammonium chloride was used as a hardener. The boards were pressed at 25 kg/cm <sup>2</sup> and T=150°C for 6 min and then conditioned at 20±2 °C and 65±5% RH to reach a moisture content of 12%. Particleboards are produced at a pressure of 25 kg/cm <sup>2</sup> at 150°C for 6 minutes. | 600-700                      | -                              | (Çöpür et al., 2007)                                                                                          |
| Hemp             | Hemp fibres were chopped, mixed with water, hydraulic lime and hydrated lime and put in a mould. They were removed after 4 days and after one week they were put in a climatic chamber (T=50°C) for 6 days. After reaching a constant mass, thermal conductivity is calculated by a heat flow meter with the plates at 5°C and 25°C.                                                                                                                             | 369-475                      | 0.0899-0.1079                  | (Benfratello et al., 2013; Nguyen et al., 2018b)                                                              |
| Kenaf            | The raw material was cut in chips (thickness of 0.5 mm) and the particles were air-dried to 12% moisture content. A sealed steam injection press allowed the mat                                                                                                                                                                                                                                                                                                 | 150-200                      | 0.051-0.058                    | (Xu et al., 2004; Madurwar et al., 2013)                                                                      |

| Material         | Summary of sample preparation                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         | Density<br>[kg/m <sup>3</sup> ] | Thermal conduc-<br>tivity [W/(m.K)] | References                                |
|------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|-------------------------------------|-------------------------------------------|
|                  | pressing (pressure=1.10 MPa at 183°C for 7-10 minutes). Finally, the binderless boards were conditioned at room temperature and reach a moisture content of 5%–7%. Thermal conductivity is evaluated according to the American Society for Testing Materials (ASTM C518-17a).                                                                                                                                                                                                                                                                                                                         |                                 |                                     |                                           |
| Pineapple leaves | The board was made of dried shredded pineapple leaves and natural rubber latex as a binder. The leaves were manually cut, soften in NaOH solution for 30 minutes, dried at T=80°C for 12h, mechanically cut and dried again at T=80°C for 12h. Then natural rubber was spread in different ratios to produce specimens of 200mmx200mmx15mm.                                                                                                                                                                                                                                                           | 178-232                         | 0.035-0.043                         | (Tangjuank, 2011; Asdrubali et al., 2015) |
| Rice husk        | Boards were made of rice husk (30% by volume of earth), earth, hemihydrate gypsum (20% by volume of earth) and air lime (10% by volume of earth). Earth, gypsum, lime and water were mixed for 90 s. then rice husk was added and mixed mechanically for 1 min. The samples were moulded, dried at T=23 °C 50% RH for 2 weeks, demoulded and tested after two more weeks. Thermal conductivity is evaluated by using an ISOMET 2104 Heat Transfer Analyser equipment at T=23 °C and RH=50%.<br>*Only the composition with 30% rice husk is reported as show the best thermal insulation performances. | 650.8                           | 0.102                               | (Antunes et al., 2019)                    |

Note: No data is presented when it is not provided by the considered reference.

Density and thermal conductivity are extremely important for insulation properties, board production and application on buildings. They are strictly connected, and they widely affect porosity (Chikhi et al., 2013; Palumbo et al., 2016; Nguyen et al., 2017; Nguyen et al., 2018b; Nguyen et al., 2020). The lower the density and thermal conductivity, the better is the thermal insulation performance, as Panyakaew and Fotios (2011) reported. In terms of raw material density, there are many different values because of the different characteristics of the fibres and crops. Board density is lower due to its constituents and the production method. Considering thermal conductivity, it is possible to note that several materials have promising values, both for raw material and when used in boards. Normally for a thermal insulation element, thermal conductivity is lower than 0.065 W/(m.K) (AFNOR, 1983).

Figures 2.2-2 and 2.2-3 show the comparison between considered particleboards and an insulation expanded corkboard (ICB), already commercially widespread and used. Considering Tártaro et al. (2017) and Fu et al. (2020), ICB has a density of 100-114 kg/m<sup>3</sup> and thermal conductivity of 0.042 W/(m.K). The results demonstrate good insulating properties of bio-wastes and the feasibility of using them for boards' production. Figure 2.2-4 shows the correlation between thermal conductivity and density considering Table 2.2-8.

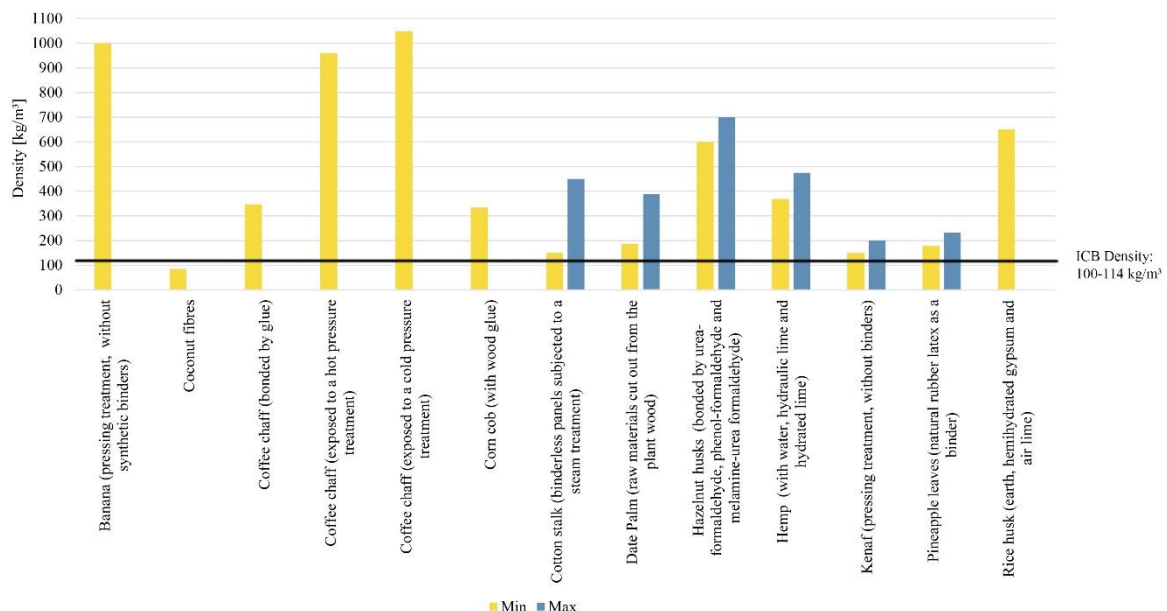


Figure 2.2-2. Density of particleboards with different bio-wastes (based on Table 2.2-8) compared with density of insulation expanded corkboard (ICB).

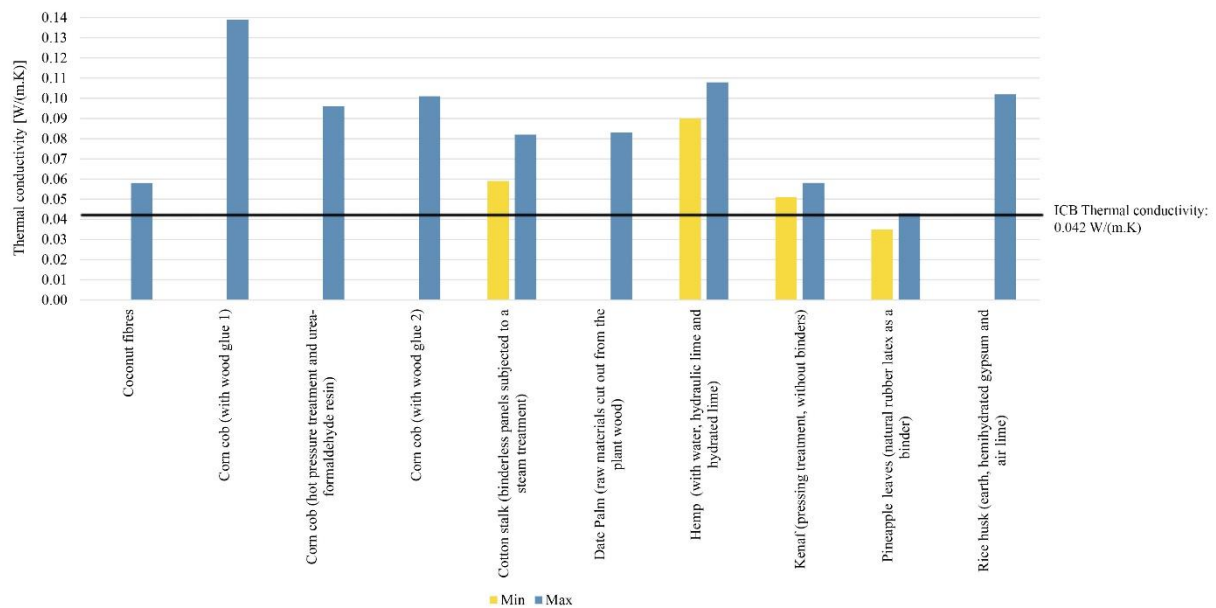


Figure 2.2-3. Thermal conductivity of particleboards with different bio-wastes (based on Table 2.2-8) compared with thermal conductivity of insulation expanded corkboard (ICB).

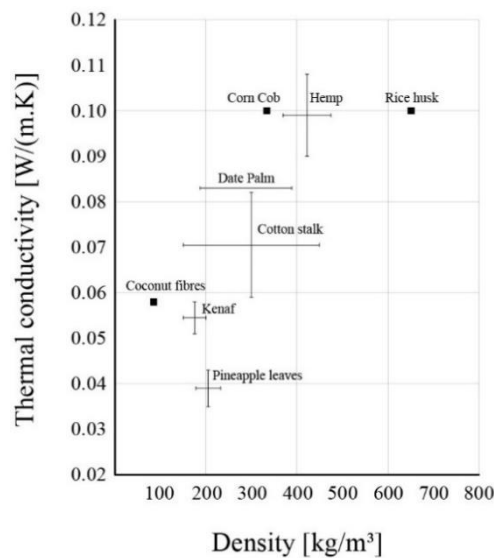


Figure 2.2-4. Correlation between thermal conductivity and density of boards with bio-wastes (based on Table 2.2-8).

Tensile strength is another widely studied property, especially for reinforced composites with natural fibres. Past research reported that cellulose contents and length of the fibres influence their tensile strength (John and Thomas, 2008; Faruk et al., 2012; Nguyen et al., 2018a), a longer fibre length results in lower values. Considering the tensile properties of the raw materials, shown in Table 2.2-7,

it is difficult to define the relationship between the percentage of cellulose, fibre length and tensile strength. The values are extremely different from each other, and the range is too large.

## **2.2.5 Bio-wastes from the Euro-Mediterranean area suitable for building products and associated challenges**

As previously described, this article focuses on Euro-Mediterranean Countries (Table 2.2-4) where thermal retrofitting is needed in many traditional buildings. That retrofitting must be compatible with the existent walls and should be eco-efficient.

### **2.2.5.1 Selected bio-wastes from the Euro-Mediterranean area**

Several past studies have considered the four wastes chosen (Section 2.2.4.3) as possible building materials and evaluated their thermal performances. Liuzzi et al. (2020) investigated olive pruning waste and barley straw to produce an insulating board, and a mixture of clay sand and olive fibres to produce a plaster (Liuzzi et al., 2018). Both studies demonstrated that higher contents of olive fibres improved thermal performances.

Eliche-Quesada et al. (2011) described the feasibility of adding coffee grounds to clay for brick production. They demonstrated that this bio-waste reduced density and increased insulation performances and porosity. Kua et al. (2016) investigated the optimum composition to produce a geopolymeric material using coffee grounds, fly ash and blast furnace slag. Lachheb et al. (2019) demonstrated that adding coffee grounds improved the thermal properties of an industrial gypsum plaster with a density of 1150 kg/m<sup>3</sup>.

For waste pomace from wine production, Muñoz et al. (2014) and Taurino et al. (2019) tested clay bricks with the addition of this bio-waste. They both obtained building elements with lower bulk density, higher porosity and better insulation properties in comparison to a reference sample. Wine wastes also reduced linear drying shrinkage. Pedras et al. (2020) studied the chemical characterization of red wine grape pomace and said that lignin content is rather high. This information can suggest possible adhesive properties of the bio-waste.

Studies with hazelnut shells (Çöpür et al. 2007) showed the possibility of producing particleboards using urea-formaldehyde, phenol-formaldehyde and melamine-formaldehyde as binders. They demonstrated that this is a feasible solution, and this bio-waste can be used for board production. Nevertheless, these have lower dimensional stability (higher thickness swelling and water absorption) than wood-particleboards. This may be solved using a water repellent.

The sustainability of using these bio-wastes for building products is supported also by information related to their collection. For example, Dam and Harmsen, (2010) reported that spent coffee grounds are not suitable for burning and are therefore disposed of in landfills together with domestic and commercial solid waste. Past studies (Dam and Harmsen, 2010; Echeverria and Nuti, 2017)

addressed different practices to reuse them such as animal feed, composting and fermentation. Pedras et al. (2020) reported that agro-wastes are often used for animal feed, citing olive pomace, grape pomace and coffee grounds. Hazelnut shells are normally used for combustion and heating (Lopes et al., 2012; Demirer et al., 2018).

Table 2.2-9 collects information related to the chemical composition and properties of these four selected materials. For cellulose, hemicelluloses and lignin contents, past studies reported much information. On the contrary, extractives, waxes and ashes are not reported in Table 2.2-9 due to a lack of data in the analysed articles.

The selected materials have percentages of cellulose lower than the average value of Table 2.2-5 (44.16 %), contents of hemicellulose similar to the average value (21.83 %) and higher content of lignin (average value= 20.09%). Hazelnut shells, coffee grounds and grape waste pomace have the highest values of cellulose and lignin, hence they could show adhesive properties. Coffee grounds and hazelnut shells have the highest values of hemicelluloses. Considering the information provided in Section 2.2.4.4, they could not guarantee good resistance to water, but they may provide good moisture buffering capacity.

Considering the properties previously analysed (density, thermal conductivity, moisture content and tensile strength), there is a gap in the literature. It will be interesting to define the properties of each bio-waste as raw material and then deepen the information related to boards, made from these bio-wastes and eventually a binding agent. Since the selected materials seem to have promising thermal behaviour, based on previous past studies, future research can deepen this knowledge to propose innovative sustainable building products. These considerations should be verified by experimental tests that evaluate the properties of the selected bio-wastes relating them with their chemical composition.

#### **2.2.5.2 General problems of using bio-wastes for building materials**

Although the use of bio-wastes particles and fibres for building products has several advantages, it also involves some drawbacks, as Table 2.2-6 previously described. It is important, taking into consideration also this aspect, to ensure proper use of these resources. Knowing the disadvantages of bio-wastes and fibres in building products allows the prevention of problems deriving from their use. Considering past studies, several examples of problems of using bio-wastes are reported, as the hydrophilic nature, high moisture and water absorption, low dimensional stability, durability and resistance to fire, and increased susceptibility to moulds and insects (Mohanty et al., 2005; John and Thomas, 2008; Dittenber and GangaRao, 2012; Faruk et al., 2012; Berardi and Iannace, 2015; Nguyen et al., 2017; Liuzzi et al., 2018). Some of these are detailed in this section.



Table 2.2-9. Chemical composition and properties of four selected bio-wastes from spent coffee, wine production, hazelnut shells and olive oil production.

| Material              | Chemical composition [%] |                | Properties - raw materials |                              |                                | References                                                                                                                                                                       |
|-----------------------|--------------------------|----------------|----------------------------|------------------------------|--------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
|                       | Cellulose                | Hemicelluloses | Lignin                     | Density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] |                                                                                                                                                                                  |
| Coffee grounds        | 8.60-52.42               | 6.80-44.50     | 18.30-24.52                | 380 <sup>b</sup>             | 0.20                           | (Muñoz Velasco et al., 2016; Zarrinbakhsh et al., 2016; Buratti et al., 2018; Ribeiro et al., 2018; Lachheb et al., 2019; Massaro Sousa and Ferreira, 2019; Pedras et al., 2019) |
| Grapes (waste pomace) | 15.00-50.16              | 6.62-11.30     | 30.30                      | -                            | -                              | (Barbieri et al., 2013; Muñoz et al., 2014; Pedras et al., 2020)                                                                                                                 |
| Vine shoots           | 41.14                    | 26.00          | 20.27                      | 105 <sup>b</sup>             | -                              | (Cárdenas, 2020; Wong et al., 2020)                                                                                                                                              |
| Hazelnut shells       | 22.90-34.60              | 11.30-23.50    | 30.20-41.40                | 230*                         | 0.10                           | (Çuhadaroglu, 2005; Çöpür et al., 2007; Guler et al., 2008; Salasinska and Lopes et al., 2012; Ryszkowska, 2012; Liscursi et al., 2017)                                          |
| Olive husks           | 25.00                    | 24.60          | 50.40                      | -                            | -                              | (García et al., 2016)                                                                                                                                                            |
| Olive whole stones    | 31.90                    | 21.90          | 26.50                      | -                            | -                              | (Fernández-Bolaños et al., 1999)                                                                                                                                                 |
| Olive seed husks      | 36.40                    | 26.80          | 26.00                      | -                            | -                              | (Fernández-Bolaños et al., 1999)                                                                                                                                                 |
| Olive pruning waste   | -                        | -              | -                          | 1251 <sup>a</sup>            | 0.062                          | (Liuzzi et al., 2018; Liuzzi et al., 2020)                                                                                                                                       |

Note: <sup>a</sup> Real density; <sup>b</sup> Loose bulk density; \*calculate by water displacement method.

One of the most investigated disadvantages of bio-wastes is their hydrophilic nature (Dittenber and GangaRao, 2012; Jawaid and Abdul Khalil, 2011; Nguyen et al., 2020). This property can determine also positive behaviours, depending on the type of construction element. It results in high moisture adsorption capacity whilst also allowing a passive control of the indoor condition. At the same time, it entails low dimensional stability, high swelling of the fibres if in contact with water and lower mechanical properties (Nguong et al., 2013; Faruk et al., 2012; John and Thomas, 2008). As reported in Table 2.2-6, these are recognized drawbacks of using natural fibre products and bio-aggregates for building practices. To mitigate these problems, it could be necessary using additives to control hygroscopic behaviour and mechanical properties.

Another disadvantage of lignocellulosic materials is their high flammability. As Aladejana et al. (2020) said, fire resistance is one of the most important properties for building materials. Nowadays some bio-materials made of natural wastes and fibres do not guarantee the required properties

without using solutions that correct this shortcoming (Liu et al., 2017). The high flammability of lignocellulosic materials limits their application without specific treatments and fire retardants (Belayachi et al., 2017; Prakash Chaudhari and Bhole, 2020). This can lead to higher costs and higher production times, and sometimes to lower sustainability. In addition, both flame exposure and flame retardants can adversely affect the mechanical properties of natural fibres, as Kim et al. (2018) reported.

Several studies (John and Thomas, 2008; Berardi and Iannace, 2015; Liuzzi et al., 2017; Antunes et al., 2019; Owodunni et al., 2020b) pointed out another significant drawback: being organic elements, they are more susceptible to fungi, mould and insects attack. As Palumbo et al. (2017) reported, to evaluate the feasibility of using agricultural wastes for building products, fire resistance and fungal susceptibility are the two most important issues to consider. Depending on the material and the construction element, the possible biological attacks will be different. It varies according to environmental conditions (temperature and relative humidity) and materials properties such as nutrient content, chemical composition and hygroscopicity (Palumbo et al., 2017; Viel et al., 2019b). Specific treatments can mitigate this problem, but research into suitable treatments for bio-based materials and natural fibres is limited and warrants further research (Liuzzi et al., 2017). More information related to biological susceptibility is reported later.

Finally, since the article studies agro-industrial waste, it is important to consider the harmful effects that agricultural practices could have on the environment. Some crops, intensive cultivations and agricultural trade could degrade natural resources. They could cause deforestation, soil erosion, overuse of water, pollution and negative effects on the biodiversity of animal and vegetal species (Hosonuma et al., 2012; Dudley and Alexander, 2017; Balogh and Jámbor, 2020). For example, Foster et al. (2011) and Savilaakso et al. (2014) pointed out the harmful effects of the cultivation of palm oil, highlighting the impact on biodiversity and high carbon emission. Beyer et al. (2020) reported these effects for the cultivation of soybean, rapeseed, sunflower and groundnut oil, albeit in lower amounts. Several studies reported that rice cultivations are responsible for the emission of greenhouse gases: carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) (Cai et al., 2003; Anand et al., 2005; Bhattacharyya et al., 2012). Furthermore, many crops require a high amount of water. Water demand is increasing and causes several environmental problems (Boretti and Rosa, 2019; Guerrero-Baena and Gómez-Limón, 2019). Hence, it is important to take into consideration that the use of agro-industrial bio-wastes to ensure a more sustainable architecture must not lead to higher environmental impacts caused by agricultural practices.

#### **2.2.5.3 Biological susceptibility**

To carry out future research related to bio-insulation boards, increased biodeterioration risk is one of the most important drawbacks to consider. Several past studies evaluated this problem

considering boards made up of bio-wastes and fibres. For example, Antunes et al. (2019) considered the biological susceptibility of a rice husk, earth, gypsum and air lime board concluding that, as expected, it increases with a higher content of rice husk. Monteiro et al. (2020) studied the biological susceptibility of particleboards made up of maritime pine, cardoon (*Cynara cardunculus* L.) and a starch-based bio-binder. They considered subterranean termites and wood decay fungi (*Gloeophyllum trabeum* (Pers.) Murrill) demonstrating a high susceptibility of cardoon and starch. Farag et al. (2020) analysed particleboards made up of olive stone and a polyester liquid resin as a binder, concluding that this binding material ensures good protection from fungal and bacteria attack. Palumbo et al. (2017) evaluated mould growth of an insulation board composed of corn pith, sodium alginate and some additives that mitigate fire and biological susceptibility (boric acid, aluminium hydroxide, ammonium polyphosphate, montan wax, acetic acid and lactic acid).

Curling (2017) highlighted the importance of temperature and moisture content for biological growth and described different methods to evaluate the susceptibility of the materials to biodeterioration agents such as moulds, decay fungi or insects, making a distinction between laboratory, field and service life tests and monitoring. Stefanowski et al. (2017) evaluated mould growth of specimens made up of corn pith and additives by a rapid screening method. The researchers highlighted the influence of hygric properties in biological attack and they gave a list of mould species that are normally found in indoor environments. Parracha et al. (2021) monitored the biological susceptibility to moulds of External Thermal Insulation Composite System (ETICS) adapting a method from two American standards (ASTM D5590-17 and ASTM C1338-19). A mixed spore suspension of two common moulds was applied on the surface of the specimens and the biological growth analysed during a four weeks exposure period. Visual grading was used to evaluate the biological growth that was very restricted due to the presence of biocides or the high pH of the surfaces.

To prevent biodeterioration Liu et al. (2017) suggested the use of a multilayer structure including the application of biocides and keeping the materials in dry condition. An alternative is the use of some natural materials that can prevent this problem such as air lime. Santos et al. (2017) used air lime to prevent fungal colonization in an earth-based mortar. Bumanis et al. (2020) reported a past study that demonstrated an improvement in the biological resistance of gypsum-based mortar by adding lime additives. Citric acid has widely been investigated as a fungal growth deterrent (Essoua Essoua et al. 2016). Furthermore, Lee et al. (2020) reported that it improves the biological resistance of wood and particleboards if used as a bio-binder.

## 2.2.6 Conclusions

It has been widely demonstrated that using natural fibres and other bio-wastes for building products is a feasible and efficient possibility. To guarantee more sustainable solutions, bio-wastes could be considered to produce building products, such as particleboards, insulation boards, masonry units

and mortars. Since they are constantly and widely produced, namely but not exclusively for feeding mankind and animals, they are available in large quantities. Sometimes they cause negative effects by increasing environmental problems and their disposal can be a problem. Employing them in buildings both improves recycling practices and may guarantee eco-effective solutions.

This article collects information considering past studies related to the use of natural fibres and other bio-materials for building products, focusing on bio-wastes. First, it investigates bio-materials already studied for construction practices. Then it gives a classification of bio-wastes and details the ones derived from agriculture practices and agro-industrial processes, aiming at selecting some of them to carry out future studies. It collects information about worldwide production, chemical composition, physical and mechanical properties. Knowing world production and the ranking of the major producers of crops allows taking into consideration the origins of bio-wastes, so helping to minimise environmental impact. Since transport and production of building materials are responsible for high energy consumption, this information for potential local supplies offers solutions to ecological problems. Bio-wastes derived from either olive oil and wine production or hazelnut and coffee consumption were chosen for further work in order to achieve innovative sustainable solutions.

The analysis of chemical composition gave a preview of the possibilities derived from the use of a specific natural material, but further studies are needed. Since the agro-industrial wastes are lignocellulosic materials, they have similar components: cellulose, hemicelluloses and lignin with other natural substances, such as extractives, pectin, waxy substances and ash. Each one affects the performances differently, hence knowing their percentages is useful information to preview the possible properties of the composite materials produced with them. Information related to these aspects was collected for all the agro-industrial wastes considered and then was further investigated for the four selected bio-wastes.

For physical and mechanical properties, this study made a distinction between bio-wastes and boards produced using them. This gave useful information for future research related to board production. Several studies demonstrated that using natural wastes in building products, such as plasters, brick and particleboards, often determines lower values of density and thermal conductivity and higher porosity.

For properties of raw materials, the collected information shows there is a lack of information concerning thermal conductivity and water absorption. Future research should further characterize the agro-industrial bio-wastes considered in this article, evaluating them as raw materials which can be extremely useful to develop a decision tool for the choice of materials for composites. Among the considered past studies related to board production, boards made of dried shredded pineapple leaves have the lowest conductivity (0.035-0.043 W/(m.K)), while the ones with coconut fibres and palm oil boards reach the lowest values of density (85 kg/m<sup>3</sup>). Several studies lack important information concerning the specimens' preparation and curing to allow comparison of results, such as

the initial drying of the materials, the pre-processing and some information on the production method of the specimens (aggregate-glue mixing method, information related to moulding and demoulding), conditioning of specimens and measurement method.

This future research, based on the data collection presented in this work will contribute to the use of bio-wastes in building products, reduce the gaps of knowledge found in literature and propose innovative, efficient, and more sustainable materials.

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### Supplementary material

Table SM 1 in the Appendix reports the detailed results related to the chemical composition of agro-industrial wastes used for building materials considered in Table 2.2-5 in the article.

## 2.3 Discussion on bio-susceptibility

As previously introduced, one of the biggest problems of using bio-wastes as building materials is biological susceptibility, even if this property is not so commonly assessed. Together with the organic composition, and the presence of nutrients and microorganisms in the raw materials, this drawback is caused by their hygroscopic nature (Ginestet et al., 2020; Gradeci et al., 2017; Tobon et al., 2020). Apart from the obvious aesthetic impact, the biological attack can lower the durability of the composites, affect materials' properties, and restrict their performance. The organisms' attack can modify the chemical composition of the materials (such as lignin, hemicellulose, and cellulose content) and compromise their natural structure (Bumanis et al., 2020; Gonçalves et al., 2021; Jones, 1999; Martins et al., 2011). Furthermore, biological degradation can affect the human health and well-being of the users, causing diseases and lowering indoor air quality (Brambilla and Sangiorgio, 2020; Ginestet et al., 2020; Stefanowski et al., 2017).

On the other hand, nowadays, analyses are being carried out to take advantage of biological development. Mycelium-based composites, whose name derived from mycelium, the filamentous part of a fungus (Alaneme et al., 2023) are being more and more investigated. They are developed to be applied for several purposes, such as packaging, in the automotive industry, and for the manufacturing of household items, furniture, and building materials (Aiduang et al., 2022; Cerimi et al., 2019). As for this latter, different composites have been proposed. Schmitt et al. (2021) demonstrated the feasibility of recycling beech sawdust and a spent mushroom substrate into mycelium-based composites with thermal insulation properties. Jiang et al. (2017) investigated the manufacturing process to produce a bio-composite by using a mycelium-based binder. Dias et al. (2021) produced and studied a Mycelium-Miscanthus bio-composite as a possible building product for indoor applications.

Bio-based composites' bio-susceptibility was always considered throughout the development of the Ph.D. project, and it influenced some of the main choices. Part of the initial literature review was therefore dedicated to the collection of information about this drawback. Furthermore, laboratory tests to assess the resistance to biological attack of the proposed composites were performed. This section describes some of the bio-agents that can be responsible for biological degradation, such as bacteria, fungi, insects, and marine borers.

Bacteria, mainly large groups of Gram negative bacteria and mycobacteria, are all microorganisms that exist inside buildings. *Streptomyces* is the most prevalent genus in building composites (Pietarinen et al., 2008). They can change materials' structure and increase the susceptibility to fungi. However, they cause relatively little damage to building materials, hence laboratory tests to evaluate bacteria susceptibility are not as common as for other biological attacks (Curling, 2017).

Fungi are divided into several groups. Among them, the most known contaminants for building materials are *Ascomycetes*, called mould and blue-stain fungi, and *Basidiomycetes*, called decay fungi (Burge and Otten, 1990). The first ones easily grow on building materials' surfaces. The most common indoor moulds are *Penicillium* spp. (e.g. *Penicillium chrysogenum*), *Aspergillus* spp. (such as *Aspergillus niger*, *Aspergillus versicolor*) *Chaetomium* spp. and *Cladosporium* spp. strains (e.g. *Cladosporium sphaerospermum*), *Alternaria alternata*, and *Stachybotrys chartarum* (Brambilla and Sangiorgio, 2020; Bumanis et al., 2020; Burge and Otten, 1990; Verdier et al., 2014). As for *Basidiomycetes* (decay fungi), they easily contaminate wooden constructions and wooden-based building materials, both damaging the performance and causing serious economic problems (Gabriel and Švec, 2017; Stienen et al., 2014). Depending on the species, they are responsible for brown-rot (widely spread in Europe and North America), white-rot (less common in temperate zones), and soft-rot (Schmidt, 2007; Šimůnková et al., 2021; Thybring, 2017). Brown-rot fungi deteriorate cellulose and hemicellulose; they affect the strength properties and cause a significant weight loss of the contaminated materials. White-rot fungi deteriorate predominantly lignin (Gusse et al., 2006; Kumar et al., 2020; Thybring, 2017; Vanerek et al., 2014). Soft-rot fungi normally attack the material surface and do not penetrate more than 2 cm. They appear as a soft mass in case of high humidity, as brown coloration in dry conditions, similar to brown-rot attacks (Maria Silva Brito et al., 2020). The most common indoor wood-decay are *Serpula lacrymans* (the most destructive and uncontrollable fungus in Europe) and *Coniophora puteana* (Gabriel and Švec, 2017; Schmidt, 2007). Furthermore, *Rhodonía placenta*, *Gloeophyllum trabeum*, or *Trametes versicolor* are widely known, and they have been already analysed on building composites (Shalbafan et al., 2016; Šimůnková et al., 2021). The fungi attack depends on several factors: moisture content, environmental conditions (climate, temperature, relative humidity), exposure conditions, fungal species, and spore levels (Hoang et al., 2010; Schmidt, 2007). Furthermore, fungal growth is caused by some materials' characteristics such as surface texture, nutrient content, chemical composition, and pH (Echeverria et al., 2021; Gabriel and Švec, 2017; Gonçalves et al., 2021). For this latter, past studies reported that alkaline materials, with a pH greater or equal to 10, can better resist chemical decomposition and mould growth (Parracha et al., 2021; Verdier et al., 2014).

Finally, considering the insect attack, subterranean termites are one of the highest hazards in Mediterranean countries (Duarte et al., 2021; Ewart et al., 2016). Globally, there are more than 3000 species; the areas that present the high diversity are the tropical areas of Africa, South America, and Asia (Duarte et al., 2017). Six species are widely spread in Europe, all belonging to the genus *Reticulitermes* (Blattodea, Isoptera, Rhinotermitidae): five native (*R. grassei*, *R. banyulensis*, *R. urbis*, *R. lucifugus*, and *R. balkanensis*) and one introduced (*R. flavipes*). This latter, typical of the eastern part of the United States, has spread to various regions of the world such as Canada, the Bahamas, South America, and Europe (Gascón-Garrido et al., 2013; Treu et al., 2020). Termites could easily deteriorate

building materials and worsen their performance. They can degrade materials either by using them as a source of food or by creating tunnels. They are also responsible for transporting moisture inside buildings, also securing favourable conditions for mould growth (Kutnik et al., 2010).

Table 2.3-1 reports some examples of past studies that evaluated the biological attack of bio-waste-based composites, considering decay fungi, mould, and subterranean termites. As it shows, for the same type of biological attacks, there are different bio-agents. As regards white and soft-rot attacks, past research mainly considered wood and wood-based composites rather than bio-waste-based ones.

Table 2.3-1. Examples of already analysed biological attacks of bio-waste-based composites caused by brown rot fungi, mould, and termites.

| Bio agents                                                                                                                                | Composites                                                                                                                  | Ref.                     |
|-------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------|--------------------------|
| Brown rot fungi                                                                                                                           |                                                                                                                             |                          |
| <i>Gloeophyllum trabeum</i> (Pers.)<br>Murrill                                                                                            | Particleboards with cardoon particles and a cassava starch-based binder                                                     | (Monteiro et al., 2020)  |
| <i>Brunneoporus malicola</i> (Pers.)<br>Murr. ( $\approx$ <i>Gloeophyllum trabeum</i> ) and<br><i>Trametes versicolor</i> (L., Fr.) Quél. | Particleboards with papaya stalk, macadamia nut car-pel, coffee husks, eucalypt sawdust, and urea-formal-dehyde as a binder | (Gonçalves et al., 2021) |
| <i>Gloeophyllum trabeum</i>                                                                                                               | Corn stalk-based composites with high-density poly-ethylene as the matrix and some preservatives for bi-ological attack     | (Xuan et al., 2017)      |
| White rot fungi                                                                                                                           |                                                                                                                             |                          |
| <i>Pycnoporus sanguineus</i>                                                                                                              | Corn stalk-based composites with high-density poly-ethylene as the matrix and some preservatives for bi-ological attack     | (Xuan et al., 2017)      |
| Soft rot fungi                                                                                                                            |                                                                                                                             |                          |
| <i>Chaetomium globosum</i>                                                                                                                | Corn stalk-based composites with high-density poly-ethylene as the matrix and some preservatives for bi-ological attack     | (Xuan et al., 2017)      |
| Moulds                                                                                                                                    |                                                                                                                             |                          |
| <i>Aspergillus niger</i>                                                                                                                  | Earth-based mortars with siliceous sand, oat fibres and hydrated air lime                                                   | (Santos et al., 2017)    |
| <i>Aspergillus niger</i> and <i>Penicillium funiculosum</i>                                                                               | ETICS with expanded cork thermal insulation and lime-based coating                                                          | (Parracha et al., 2021)  |
| -                                                                                                                                         | Panels with hemp shiv and rape straw, with and with-out starch derivate binders, additives, and solvents                    | (Viel et al., 2019b)     |
| Termites                                                                                                                                  |                                                                                                                             |                          |
| <i>Cryptotermes brevis</i> Walker, Isop-tera: Kalotermitidae                                                                              | Particleboards with papaya stalk, macadamia nut car-pel, coffee husks, eucalypt sawdust and urea-formal-dehyde as a binder  | (Gonçalves et al., 2021) |
| <i>Coptotermes formosanus</i>                                                                                                             | Boards with pineapple leaves and a mix of citric acid and sucrose as a binder                                               | (Indrayani et al., 2015) |
| <i>Reticulitermes grassei</i> Clément                                                                                                     | Particleboards with cardoon particles and a cassava starch-based binder                                                     | (Monteiro et al., 2020)  |

Biological susceptibility can be evaluated by many different laboratory tests, as detailed in Section 2.6. It can be avoided or moderated by using several strategies, such as specific treatments, the addition of additives, or the employment of preservative techniques. Past studies reported that heat treatment improves resistance to fungi (Esteves et al., 2021; Maria Silva Brito et al., 2020). Also, chemical treatments have been proven to be effective (Essoua Essoua et al., 2016; Paes et al., 2015). Another possibility is combining the bio-wastes with materials that increase the biological resistance, such as air lime (Krejsová and Doleželová, 2019; Santos et al., 2017), sodium benzoate (de Carvalho et al., 2020), boric acid (Palumbo et al., 2017), and citric acid (Indrayani et al., 2015; Treu et al., 2020). Finally, specific techniques to produce the composites could guarantee better resistance to fungi and insects' attacks. Brambilla and Sangiorgio (2020) described as possible strategies the reduction of thermal bridges, the increase of air changes or ventilation rates, and the control of low indoor relative humidity.

As better described in the following chapters, for the Ph.D. project *Aspergillus niger* and *Penicillium funicullosum* were considered to evaluate mould growth; as for the insect attack, subterranean termites *Reticulitermes grassei* are selected as the model organisms.

## 2.4 Adhesives: binder and glues

After analysing the bio-aggregates and discussing bio-susceptibility, the research considered the adhesives (binders and glues). A literature review was carried out considering both natural and synthetic products. Some of the collected information is reported in Table 2.4-1. Three different possibilities were selected to be investigated.

First, hemihydrate gypsum was considered for its several benefits. It is widely used for building products due to its low price, low environmental impact (when transportation is not long), and high availability (Bumanis et al., 2020; Lushnikova and Dvorkin, 2016). It is easily workable, and the literature presents it has provided good thermal and acoustic insulation properties (Boquera et al., 2021; Fatma et al., 2019). Gypsum is considered a natural flame retarder (Bumanis et al., 2020; Gürü et al., 2015) and has a very fast drying (Antunes et al., 2019), which can avoid the use of high temperatures to produce gypsum-based composites. Despite the benefits, gypsum also presents some drawbacks, such as low resistance to water (Binici et al., 2016; Lushnikova and Dvorkin, 2016), cracking, and high relative humidity (Bumanis et al., 2020; Fatma et al., 2019).

Other outcomes of the data collection (Table 2.4-1) were a sodium silicate solution and a starch-based glue. The sodium silicate solution, also known as water glass, was selected for its several benefits: bonding capacity, no toxicity, resistance to bio-deterioration and fire (Lee and Thole, 2018; Liuzzi et al., 2020a), and safety to human health. Nevertheless, it presents some drawbacks, such as low moisture resistance (Song et al., 2021). Past research has already considered the production of

low-density wood-based boards by using sodium silicate, but only a few studies detailed the production process (Lee and Thole, 2018).

As for the starch-based natural glue, more investigations were carried out to select the type of starch and, finally, maize was chosen for its high production in the considered area, as better reported in Section 3.3.

## 2.5 Additives

Due to the possible drawbacks derived from the use of agro-industrial wastes, the possibility of using additives was considered. The analysis was focused on biological susceptibility, as one of the main problems (Section 2.3).

According to past research, several materials can be employed. Santos et al. (2017) demonstrated that the addition of air lime to earth-based plasters increased the pH and improved the biological resistance. Antunes et al. (2019) added air lime to rice husk-earth-gypsum-based panels both to delay the hardening process of hemihydrate gypsum and to improve the resistance to biological attack. Krejsová and Doleželová (2019) used white hydrated lime to improve the mould resistance of calcined grey gypsum-based composites, as well as the moisture resistance. Palumbo et al. (2017) demonstrated that boric acid improved the biological resistance of insulation boards made up of corn pith and sodium alginate. Unfortunately, as reported in this past study, its toxicity is under evaluation, hence, its employment has been limited. De Carvalho et al. (2020) reported that sodium benzoate can be an efficient solution to prevent biological attacks. Treu et al. (2020) considered sorbitol and citric acid. The researchers analysed esterified wood and investigated the resistance against marine wood borers, and subterranean termites. Indrayani et al. (2015) considered citric acid as a biological repellent: they tested a medium-density fibreboard made of pineapple leaf fibre and citric acid as an adhesive and demonstrated that this latter improved the resistance against termites.

As the outcome of this part of the literature review, air lime and citric acid were selected as additives. Air lime was used as an additive to the gypsum-based binder. Since the biological susceptibility remained low, also citric acid was considered, as better described in Section 3.3. Citric acid was added also into the starch-based glue. As past studies report, starch has high bio-susceptibility and it could be better used modified (Bumanis et al., 2020). As for the sodium silicate solution, no additives to improve bio-susceptibility were considered. According to past research, sodium silicate demonstrated good resistance to bio-deterioration (Bakatovich et al., 2022; Li et al., 2019; Liuzzi et al., 2020a).

Table 2.5-1. Data collection of past studies about adhesives for boards and panels production.

| Binder/Glues                       | Synthetic description of past research and properties of the considered binder/glue                                                                                                                                                                                                                               | Reference                  |
|------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------|
| Acrylic resins                     | Analysis of several adhesives including both synthetic and renewable-based ones: acrylic resins (synthetic thermosetting or thermoplastic resins), used as paints, coating, adhesives, and sealants.                                                                                                              | (Solt et al., 2019)        |
| Casein                             | Production of medium density insulating panels made up of <i>Miscanthus</i> ( <i>Miscanthus giganteus</i> ) and Sunflower ( <i>Helianthus annuus</i> L.) stalk as aggregates, wood glue made of casein as a binder.                                                                                               | (Eschenhagen et al., 2019) |
| Chitosan                           | Analysis of several adhesives: chitosan is a polycarbohydrate soluble in acidic aqueous media, used in agriculture, water treatment, medicine, cosmetics, and fibres.                                                                                                                                             | (Solt et al., 2019)        |
|                                    | Analysis of different production parameters to produce wood-based particleboards; a mixture of cassava starch, chitosan, glycerol, and propionic acid was employed as the adhesive.                                                                                                                               | (Monteiro et al., 2019)    |
| Citric Acid*                       | Several properties of citric acid: can be used as a binder for lignocellulosic materials, wood modification, and particleboards' production; can reduce water absorption, improve resistance against termites and fungi, and increase the modulus of elasticity, compression strength, and dimensional stability. | (Lee et al., 2020)         |
|                                    | Analysis of the feasibility of using citric acid and sucrose as adhesives to produce particleboards; properties of the produced particleboards, demonstrating the promising performance of the innovative adhesive system.                                                                                        | (Umemura et al., 2013)     |
|                                    | Assessment of citric acid as a natural adhesive for bamboo particleboard; it improved the dimensional stability and mechanical properties of the boards.                                                                                                                                                          | (Widyorini et al., 2016)   |
|                                    | Production of particleboards made of sugarcane bagasse stalks as aggregates, and citric acid as the adhesive which could be used as a bonding agent to produce high-quality particleboards.                                                                                                                       | (Syamani et al., 2020)     |
|                                    | Particleboards made of coconut fibres and a potato starch-based adhesive, modified by adding citric acid, which improves the properties of the particleboards as well as the binding capacity of the adhesive.                                                                                                    | (Owodunni et al., 2020)    |
|                                    | Analysis of the feasibility of producing films by using banana fibre and wheat gluten. The composite was cross-linked with citric acid which improved the mechanical properties and water resistance.                                                                                                             | (Nataraj et al., 2018)     |
| Bone glue                          | Performance of bio-insulation materials based on bamboo fibres and bone glue made by dissolving glue in water.                                                                                                                                                                                                    | (Nguyen et al., 2017)      |
|                                    | Description of the production of medium-density insulating panels made of <i>Miscanthus</i> ( <i>Miscanthus giganteus</i> ) and Sunflower ( <i>Helianthus annuus</i> L.) stalk as aggregates, bone glue made of gelatine as a binder; this protein-based binder is usually used in crafts.                        | (Eschenhagen et al., 2019) |
| Emulsion polymer isocyanates (EPI) | Description of synthetic and renewable-based adhesives, such as the Emulsion polymer isocyanates, which consist of two components: pMDI and an emulsion latex containing hydroxyl groups; used for panel production, to bond plastics to wood surfaces, and for bonding OSB.                                      | (Solt et al., 2019)        |



| Binder/Glues                                      | Synthetic description of past research and properties of the considered binder/glue                                                                                                                                                                                              | Reference                            |
|---------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------|
| Epoxy resin*                                      | Analysis of the influence of epoxy resin on urea-lignin-glyoxal resins for plywood panels' production. The epoxy resin improved solids content, density, and viscosity, accelerated its gel time, and increased the shear strength, water resistance, and dimensional stability. | (Younesi-Kordkheili and Pizzi, 2020) |
| Furfural                                          | Overview of the use of alternative binders: furfural is reported as a substitute for formaldehyde in particleboard formulation.                                                                                                                                                  | (Kariuki et al., 2019)               |
|                                                   | Description of different types of adhesives and glues: furfural, produced by the acid treatment of carbohydrates from waste plant material is less dangerous than urea formaldehyde and is currently considered to be a purely synthetic glue.                                   | (Cardoso et al., 2015)               |
| Glyoxal                                           | Description of synthetic and renewable-based adhesives: glyoxal is the simplest and most important dialdehyde, liquid at ambient temperature. Its oxidation in an aqueous solution produces glyoxylic acid.                                                                      | (Solt et al., 2019)                  |
|                                                   | Description of several bio-based adhesive systems for wood panels: lignin in combination with glyoxal, which is a non-toxic aldehyde less reactive than formaldehyde.                                                                                                            | (Hemmilä et al., 2017)               |
| Lignin                                            | Description of synthetic and renewable-based adhesives: lignin, a complex polymer present in all plants, can be isolated and used as an adhesive.                                                                                                                                | (Solt et al., 2019)                  |
|                                                   | Review about different types of adhesives and glues: lignin could be a suitable material, although it has a complex structure, and it can be difficult to be used alone to produce glues.                                                                                        | (Cardoso et al., 2015)               |
|                                                   | Analysis of the influence of epoxy resin on urea-lignin-glyoxal resins for plywood panels' production; information about lignin, that is produced by bagasse soda black liquor.                                                                                                  | (Younesi-Kordkheili and Pizzi, 2020) |
|                                                   | Description of several bio-based adhesive systems for wood panels: lignin is described and presented as a new bio-based alternative adhesive.                                                                                                                                    | (Hemmilä et al., 2017)               |
| Oil                                               | Review of different types of adhesives and glues: oils derived from coconut, corn, cottonseed, palm, olives, peanuts, flaxseed, safflower, soybean, sunflower seed, and rapeseed.                                                                                                | (Cardoso et al., 2015)               |
| Polyamidoamine-epichlorohydrin (PAE)              | Analysis of synthetic and renewable-based adhesives: Polyamidoamine-epichlorohydrin (PAE) resins are reported for making tissue paper, paper towels, and base paper for liquid packaging.                                                                                        | (Solt et al., 2019)                  |
| Polyethylene terephthalate (PET)                  | Description of several types of adhesives: polyethylene terephthalate (PET) is produced from monomers derived from crude oil; it is mainly used for textile fibres and plastic bottles.                                                                                          | (Solt et al., 2019)                  |
| Polylactic acid                                   | Polylactic acid is a renewable and biodegradable polyester; lactic acid derives from the fermentation of glucose; used for bottles, cups, packages, and films.                                                                                                                   | (Solt et al., 2019)                  |
| Polymeric methylene diphenyl diisocyanate (pMDI)* | Description of synthetic and non-synthetic adhesives: pMDI is used in the production of polyurethanes.                                                                                                                                                                           | (Solt et al., 2019)                  |
|                                                   | Assessment of the influence of the addition of pMDI in Urea-glyoxal resin on particleboards' properties; pMDI increases viscosity, solid content, dimensional stability, and improves bending properties.                                                                        | (Younesi-Kordkheili and Pizzi, 2018) |

| Binder/Glues                        | Synthetic description of past research and properties of the considered binder/glue                                                                                                                                                                         | Reference                   |
|-------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------|
| Poly (vinyl acetate) adhesive (PVA) | Description of synthetic and non-synthetic adhesives: PVA is used as paint, as a binder in paper, and for wood applications such as furniture construction. Formaldehyde and other materials are usually added as a preservative.                           | (Solt et al., 2019)         |
|                                     | Analysis of the properties of innovative composites made up of almond skin waste as aggregates and polyvinyl acetate glue as the adhesive; the composites have good physical, hygrothermal, and acoustic properties.                                        | (Liuzzi et al., 2020b)      |
| Polystyrene (PS)                    | Description of different adhesives: polystyrene, used in technical and household products, such as EPS as packaging or insulation material.                                                                                                                 | (Solt et al., 2019)         |
| Sodium silicate solution            | Analysis of composites made of straw and olive fibres as aggregates, and a sodium silicate solution as an adhesive; promising thermal insulation properties and sound absorption capacity.                                                                  | (Liuzzi et al., 2020a)      |
|                                     | Assessment of the properties of particleboard made of sugarcane bagasse and sodium silicate as a binder; the distribution of the sodium silicate has a great impact on the final properties.                                                                | (Ng et al., 2018)           |
| Soy                                 | Description of different types of renewable-based adhesives: soy proteins as a good solution to achieve good bending strength.                                                                                                                              | (Solt et al., 2019)         |
|                                     | Information on adhesives and glues: several types of soy-based glues; one of the most used consists mainly of soy flour and a small amount of a curing agent, namely polyamide amine epichlorohydrin (PAE).                                                 | (Cardoso et al., 2015)      |
|                                     | Investigation on possible natural components of wood adhesive resins: soy flour mixed with different materials, such as formaldehyde, lignin, and glyoxal.                                                                                                  | (Amaral-Labat et al., 2008) |
|                                     | Description of several bio-based adhesive systems for wood panels: soy protein is inexpensive and easy to handle, it requires low pressing temperatures, and can bond wood with relatively high moisture content.                                           | (Hemmilä et al., 2017)      |
| Starch and Starch Derivatives       | Starch and starch derivatives as a good possibility to produce adhesives; starch is produced by several plants and used both for food and in non-food technical applications (e.g., as an adhesive, for the paper industry, and as a textile sizing agent). | (Solt et al., 2019)         |
|                                     | Evaluation of the influence of different production parameters on the properties of low-density particleboards made of sour cassava starch formulation as an adhesive and recycled wood particles as aggregates.                                            | (Monteiro et al., 2019)     |
|                                     | Assessment of a starch-based adhesive for low-density particleboards; it has low water resistance; the addition of chitosan improved its properties.                                                                                                        | (Monteiro, 2020)            |
|                                     | Description of the production of medium-density insulating panels made up of <i>Miscanthus</i> ( <i>Miscanthus giganteus</i> ) and Sunflower ( <i>Helianthus annuus</i> L.) stalks as aggregates and a starch-based binder.                                 | (Eschenhagen et al., 2019)  |
|                                     | Analysis of particleboards made of coconut fibres and a potato starch-based adhesive; assessment of the starch-based adhesive modified by adding citric acid, which improves the binding capacity and particleboards' properties.                           | (Owodunni et al., 2020)     |
|                                     | Description of several bio-based adhesive systems for wood panels: starch adhesives consist of joined glucose units; due to their high viscosity, it is difficult to use them for industrial applications.                                                  | (Hemmilä et al., 2017)      |

| Binder/Glues                | Synthetic description of past research and properties of the considered binder/glue                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           | Reference                                                                                                                                                     |
|-----------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------|
|                             | Description of starch as a natural glue: it is a widely available, inexpensive, renewable, and biodegradable polymer; it has low moisture resistance that can be moderated (e.g., by adding citric acid, borax, and sodium hypochlorite).<br>Analysis of composites made up of coniferous needles and a starch-based binder.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | (V. Gadhave et al., 2017)<br>(Muizniece and Blumberga, 2016)                                                                                                  |
| Tannin                      | Description of tannins and their several applications: they could be employed as adhesives for wood-based products; good resistance to fungi and moisture.<br>Description of different types of adhesives and glues: tannins are present in wood, bark, and wine; the extraction of tannins is a very simple process.<br>Description of several bio-based adhesive systems for wood panels: tannins have good resistance to water and moisture and very low formaldehyde emission; they can be extracted from several plants, but they are not available in large quantities to compete with synthetic adhesives in most parts of the world.                                                                                                                                                                                                                                                                                                                                  | (Solt et al., 2019)<br>(Cardoso et al., 2015)<br>(Hemmilä et al., 2017)                                                                                       |
| Thick spent sulphite liquor | Assessment of spent sulphite liquor, a by-product from the acidic sulphite wood pulping process, combined with wheat flour and used as an adhesive; the binder is sprayed onto wood chips; the composite meets the standards' requirements for particleboards.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                | (Ferreira et al., 2018)                                                                                                                                       |
| Urea-formaldehyde           | Overview of the use of binders: urea formaldehyde is widely used for particleboards production; it guarantees good performance; by adding chemical additives formaldehyde emission can be reduced.<br>Description of synthetic and no synthetic adhesives: urea-formaldehyde resins, usually modified with more expensive materials.<br>Analysis of boards made of banana ( <i>Musa acuminata</i> ) chips and urea-formaldehyde; the boards have good mechanical properties.<br>Evaluation of composite made up of the by-product derived from the production of sugar from sugar beet and urea-formaldehyde as the adhesive; the produced boards meet the requirements for particleboards.<br>Analysis of the feasibility of producing particleboards by using almond shells and urea-formaldehyde, adding ammonium chloride as a hardener.<br>Assessment of the feasibility of using walnut shells and urea-formaldehyde resin to produce boards, possibly mixed with wood. | (Kariuki et al., 2019)<br>(Solt et al., 2019)<br>(Papadopoulos, 2018)<br>(Borysiuk et al., 2019)<br>(Pirayesh and Khazaeian, 2012)<br>(Pirayesh et al., 2012) |
| Urea-glyoxal (UG)           | Evaluation of the influence of the addition of pMDI in urea-glyoxal (UG) resin on particleboards' properties; UG is a non-toxic and eco-friendly material that can be used as an adhesive.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    | (Younesi-Kordkheili and Pizzi, 2018)                                                                                                                          |

\* Reported past studies that considered the material both as an adhesive and an additive.

## 2.6 Article B - A review of laboratory tests to evaluate agro-industrial wastes properties as building materials

The information reported in this Section was presented in the conference TEST&E 2022, NOVA FCT, Campus of Caparica, Almada, Portugal, June 21-23, 2022, based on an extended abstract. A chapter was afterwards written and published in Chastre et al. Testing and Experimentation in Civil Engineering. TEST&E 2022. RILEM Bookseries, vol 41. Springer, Cham (2023) 55-66 - [https://doi.org/10.1007/978-3-031-29191-3\\_5](https://doi.org/10.1007/978-3-031-29191-3_5)

### Abstract

The current research trend is finding out feasible solutions to guarantee sustainable building practices. Among the studied possibilities, the production of eco-friendly materials is widely investigated. To propose efficient building products the proper evaluation of their properties is extremely important. The present work considers some laboratory tests to analyse building materials, focusing on agro-industrial wastes (bio-wastes of floriculture and forest management, agricultural practices and agro-industrial processes). It considers hygrothermal performance and biological susceptibility of agro-industrial wastes both individually and in composites. Several past studies addressed this topic by using different laboratory tests and mainly considering composites. Although bio-susceptibility is one of the main drawbacks of bio-based building materials, it is not so commonly assessed. However, many laboratory tests can be carried out, due to the different bio agents and critical conditions. The present work aims at collecting this information to give a summarised list of methods that could be a simplified starting point to study agro-industrial wastes and agro-industrial waste-based composites for building practices.

**Keywords:** Bio-based, Bio-susceptibility, Bio-waste, Composites, Hygroscopicity, Thermal properties, Raw material.

### 2.6.1 Introduction

It is widely known that the building sector is one of the human activities most responsible for environmental impact, requiring about 40% of the total energy use [1,2]. Eco-efficient solutions to moderate this problem have been studied. Among them, the possibility of using agro-

industrial wastes (from forest management and floriculture, agricultural practices and agro-industrial processes) to produce building composites with thermal insulation properties. These composites can improve the energy performance of the building stock, moderate the environmental impact, and reduce the volumes of wastes disposal [3-5].

Several past studies investigated the hygrothermal properties of composites made up of agro-industrial wastes considering different methods. Brás et al. [6] analysed the thermal performance of cement-based mortars with expanded cork granulate, a by-product derived from cutting and trimming operations, by using the transient method. They demonstrated that cork improves mortar's thermal insulation capacity. La Gennusa et al. [1] combined several agro-industrial wastes with hydraulic lime and evaluated their thermal performance by heat flow meter. They demonstrated that the most uniform samples made up of the lightest aggregates had better insulation properties, although all the considered materials showed promising performance. Furthermore, they concluded that thermal conductivity was widely influenced by samples' composition, density and temperature during the tests. Czajkowski et al. [7] analysed the thermal properties of commercial insulating panels made up of cereal straw by considering the transient method and the inverse method. The researchers demonstrated that both methods were effective and that considering the temperature's influence guaranteed higher accuracy of the thermal conductivity measurements.

Even though most of the past studies focused on composites, also the properties of the raw materials are extremely important, as they are closely linked [8]. Past research assessed this topic mainly considering the recommendations given by Amziane et al. [9]. This document reported the outcome of a RILEM technical committee, proposing how to evaluate some properties of bio-aggregates. However, many other different methods could be considered, too. Knowing the several possibilities to evaluate hygrothermal properties of agro-industrial wastes allows selecting the most suitable ones for each type of application.

Even if the employment of agro-industrial wastes for building practices guarantees many benefits, there are also several drawbacks, such as lowering fire resistance, lowering durability and increasing bio-susceptibility [10,11], while increasing hygroscopicity. Laboratory tests have to be focused on the analysis of these problems too, to guarantee a complete evaluation. Knowing the drawbacks helps to moderate and solve them, making agro-industrial waste-based composites more efficient. Biological susceptibility is one of the biggest drawbacks; it affects the durability of the materials, human health, and internal comfort. It depends on different factors, such as chemical composition (e.g. cellulose, hemicellulose, lignin contents), environmental conditions (temperature and relative humidity), surface texture, or pH [12-15]. For

this reason, many laboratory tests could be carried out. This allows finding out solutions to moderate biological attacks, e.g. by specific treatments of the agro-industrial wastes, combining different materials, or using specific techniques to produce composites. Santos et al. [16] used air lime to improve resistance to mould growth of earth-based plasters. De Carvalho et al. [17] produced panels made up of sunflower stalks and gypsum, adding potassium benzoate to inhibit fungal proliferation. Brambilla and Sangiorgio [12] reported strategies to lower bio-susceptibility in buildings: the reduction of thermal bridges and the increase of air changes or ventilation rates to control indoor relative humidity.

The feasibility of using agro-industrial wastes for building practices could be evaluated by considering several laboratory tests. The present work aims at summarizing some already used, focusing on hygrothermal performance and biological susceptibility. It considers both agro-industrial wastes and agro-industrial wastes-based composites. The outcome is a summarised list of information that could be a starting point to improve the already known laboratory tests and develop new ones.

## 2.6.2 Testing agro-industrial wastes and composites

### 2.6.2.1 Agro-industrial wastes

Since raw materials' properties are linked to the ones of the final products [8], the analysis of agro-industrial wastes individually secures a more conscious use of them. Several recent past studies [18-21] reported that there are no unified standards to evaluate the properties of agro-industrial wastes. Therefore, the same standards of conventional materials may be considered. However, many past studies referred to RILEM Technical Committee 236-BBM "Bio-aggregate-based building materials" recommendations [9]. The document described guidelines for evaluating loose bulk density, particle size, thermal conductivity, water content, water absorption and hygroscopicity of bio-aggregates, reporting hemp shiv as an example.

Laborel-Préneron et al. [18] referenced recommendations of RILEM TC 236-BBM [9] to study the properties of barley straw, corn cob and hemp shiv. They evaluated particle size distribution (by image analysis method), loose bulk density, thermal conductivity (by hot plate method) and water absorption. For sorption and desorption behaviour they considered Dynamic Vapour Sorption (DVS) method. Cintura et al. [22] analysed loose bulk density and particle size distribution (by sieving method) of grape and olive press wastes, hazelnut shells, spent coffee grounds and Maritime pine (*Pinus pinaster* Ait.) chips by using the same recommendations [9]. Loose bulk density was evaluated also according to EN 1097-3 [23], while thermal

conductivity was assessed by the transient method, by using the Modified Transient Plane Source sensor (ISOMET).

Different methods from the recommendations of RILEM TC 236-BBM could be considered to evaluate bio-aggregates' properties. For example, Brouard et al. [24] analysed rape straw and sunflower stem and described a different method to evaluate water absorption. The researchers immersed the aggregates in water for 1 hour at different temperatures and considered their mass at specific times. They also evaluated microstructure, to investigate aggregates' pore size, by using Scanning Electron Microscopy (SEM). Ansell et al. [20] referred to RILEM TC 236-BBM [9] to characterize hemp, flax, rape and wheat straw, and corn cob only for loose bulk density. Microstructural morphological properties were evaluated by SEM, porosity by mercury intrusion porosimeter (MIP), thermal conductivity by heat flow meter, and moisture buffering value (MBV) by the NORDTEST method [25]. Liuzzi et al. [26] considered olive leaves and olive branches. They analysed porosity and density by using helium gas pycnometer and microstructural morphological properties by SEM.

As these past studies demonstrated, there is not one specific and unified method to characterize agro-industrial wastes. This allows selecting the most suitable laboratory tests, according to the considered materials. On the other side, only using the same methods could guarantee an effective comparison between different materials.

#### **2.6.2.2 Composites**

Building composites have been more studied than raw materials [11]. Depending on the type (i.e., particleboards, plasters, mortars, etc.), there are suitable standardized and non-standardized assessment methods used.

Binici et al. [27] produced composites of corn stalks and epoxy resin (Araldite-LY-554), gypsum and Portland cement (CEM I 42.5) as binders. The researchers evaluated water absorption according to ASTM 67 [28] and thermal conductivity according to ASTM C1113 [29]. The results demonstrated that the composites could be considered thermal insulation materials.

Lima and Faria [30] used the German standard DIN 18947 [31] to evaluate density, thermal conductivity and hygroscopicity of plastering mortars made up of clayish earth and siliceous sand and different percentages of oat straw and typha fibre-wool. As the researchers reported, DIN standards are known as an important reference for earth-based materials. The results demonstrated that natural fibres improved the thermal insulation properties though depending on the fibres and content.

Liuzzi et al. [26] considered European standards to study the hygrothermal properties of a clay plaster with olive fibres (leaves and branches). Water vapour permeability was evaluated according to EN 1015-19 [32], and hygroscopicity properties by using EN 12571 [33]. Porosity was evaluated by helium gas pycnometer. As for thermal properties, the researchers used the transient method (ISOMET); for MBV, the NORDTEST protocol [25] was used. They demonstrated that olive fibres improved the insulation performance of the composite. NORDTEST [25] is a widely used method to determine the MBV [34]. It can be applied to different composites. For example, it was considered for panels made up of rice husk and earth, stabilised with hemihydrated gypsum and air lime [35]; for panels made up of hemp shiv and a bio-based binder of starch derivative and a crosslinking agent [20]; for hemp-lime (hemp shiv, hydraulic lime and a rapid setting additive) [36].

Monteiro et al. [37], Farag et al. [38] and Nunes et al. [39] considered European standards for particleboards. They analysed particleboards composited of cardoon particles and a cassava starch-based binder, panels made up of olive stones and unsaturated polyester liquid resin (Jet sealer), and cement-bonded particleboards with banana pseudostem, respectively. Bulk density was evaluated according to EN 323 [40]; moisture content according to EN 322 [41].

Palumbo et al. [42] considered EN 12664 [43] to study the thermal conductivity of samples of different bio-based insulation materials: hemp-lime, hemp fibre, wood wool, wood fibre, barley straw-starch and corn pith-alginate. As for the hygroscopicity properties, ISO 24353 [44] was referenced.

As this data collection shows, there are many different methods already known and used to evaluate bio-waste-based composites and a great number of past studies that addressed this topic.

### **2.6.2.3 Bio-susceptibility**

Several laboratory tests are already used to evaluate biological susceptibility, but this property is not so commonly considered, neither for agro-industrial wastes individually nor for composites. As previously anticipated, this is a known drawback of agro-industrial wastes, hence it should be further assessed.

Table 2.6-1 collects examples of information from past studies that evaluated biological susceptibility to some of the relevant biodeterioration agents.



Table 2.6-1. Examples of past studies that evaluated bio-susceptibility of agro-industrial waste, individually and in composites.

| Biological attack | Bio agents                                                                                                                          | Standard/ Method                                       | Composites/ Materials*                                                                                                   | Ref. |
|-------------------|-------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------|------|
| Brown rot fungi   | <i>Gloeophyllum trabeum</i> (Pers.) Murrill                                                                                         | "Mini-block method" [45]                               | Particleboards with cardoon particles and a cassava starch-based binder                                                  | [37] |
|                   | <i>Brunneoporus malicola</i> (Pers.) Murr. ( $\approx$ <i>Gloeophyllum trabeum</i> ) and <i>Trametes versicolor</i> (L., Fr.) Quél. | Adapted from AWP A E-10 [46] and AWP A E-30 [47]       | Particleboards with papaya stalk, macadamia nut carpel, coffee husks, eucalypt sawdust and urea-formaldehyde as a binder | [13] |
| Moulds            | <i>Aspergillus niger</i>                                                                                                            | Adapted from ASTM D5590-17 [48]                        | Earth-based mortars with siliceous sand, oat fibres and hydrated air lime                                                | [16] |
|                   | <i>Aspergillus niger</i> and <i>Penicillium funiculosum</i>                                                                         | Adapted from ASTM D5590-17 [48] and ASTM C1338-19 [49] | ETICS with expanded cork thermal insulation and lime-based coating                                                       | [14] |
|                   | <i>Aspergillus niger</i> and <i>Penicillium funiculosum</i>                                                                         | Adapted from ASTM D5590-17 [48] and ASTM C1338-19 [49] | Cement-bonded particleboards with banana fibres, maritime pine ( <i>Pinus pinaster</i> ) chips and Portland cement       | [39] |
|                   | <i>Aspergillus niger</i> and <i>Penicillium funiculosum</i>                                                                         | Adapted from ASTM D5590-17 [48]                        | Grape press waste, olive press waste, hazelnut shells, spent coffee grounds and wood chips as raw materials              | [8]  |
|                   | No inoculation                                                                                                                      | Johansson et al. [50]                                  | Insulation composites with corn pith bonded with sodium alginate and calcium sulphate dihydrate                          | [10] |
|                   | -                                                                                                                                   | Viel et al. [51]; assessment according to ISO 846 [52] | Panels with hemp shiv and rape straw, with and without starch derivate binders additives and solvents                    | [51] |
| Termites          | <i>Cryptotermes brevis</i> Walker, Isop-tera: Kalotermitidae                                                                        | Maistrello [53]                                        | Particleboards with papaya stalk, macadamia nut carpel, coffee husks, eucalypt sawdust and urea-formaldehyde as a binder | [13] |
|                   | <i>Coptotermes formosanus</i>                                                                                                       | Mass loss and survival rates of termite                | Boards with pineapple leaves and a mix of citric acid and sucrose as a binder                                            | [54] |
|                   | <i>Reticulitermes grassei</i>                                                                                                       | Cintura et al. [8]                                     | Grape press waste, olive press waste, hazelnut shells, spent coffee grounds and wood chips as raw materials              | [8]  |
|                   | <i>Reticulitermes grassei</i> Clément                                                                                               | Adaptation of EN 117 [55]                              | Particleboards with cardoon particles and a cassava starch-based binder                                                  | [37] |

\*Note: only bio-based composites are reported.

In addition to the assessment of biological susceptibility, some past studies investigated the possibilities to moderate and solve it. Considering the ones in Table 2.6-1, Indrayani et al. [54] reported that citric acid avoided degradation by termites; Palumbo et al. [10] demonstrated that boric acid could improve resistance to mould growth.

### 2.6.3 Discussion

This collection of information demonstrates that there are many different methods to evaluate hygrothermal properties and bio susceptibility of agro-industrial wastes, both as raw materials and included in composites.

Agro-industrial wastes' properties as raw materials are commonly evaluated according to the recommendations of RILEM Technical Committee 236-BBM [9]. This is a useful and important guideline, also considering the relevance that bio-aggregates are taking in the last years. It is in step with the times and answers to the current research's requirements. Nevertheless, this document does not assess some important properties that could improve the evaluation of the performance of bio-aggregates. Some information could be further detailed or deepened [8]. For example, as Brouard et al. [24] described, water absorption could be evaluated considering the influence of temperature. As for the particle size analysis by the sieving method, the sieving time could be specified according to the type of bio-aggregates [8]. The thermal conductivity could be related to hygroscopic capacity. Other important information could be added, namely the evaluation of biological susceptibility by considering different bio-deterioration agents. However, all these are considerations to improve a cutting-edge document and an important reference to study bio-aggregates by a unified method. Finally, it is important to underline that the lack of a specific standard to evaluate agro-industrial wastes' properties could be both an advantage (flexibility in the choice of the methods) and a disadvantage (difficult comparison between the results and different studies).

As for composites, several standardized methods could be considered. Many of them are not specific for agro-industrial wastes, as they were drawn up for conventional materials. Considering the research trend of using bio-based materials, these methods could be updated. Specific standards can be proposed too, as, for example, the German standard for earth plasters, considered by Lima and Faria [30]. Nevertheless, the presence of many different methods already used has some benefits. First, it is possible to choose the most suitable one for the considered composites. Then, the same property could be evaluated by different laboratory tests that could enrich the discussions of the results and allow reaching important conclusions. Finally, since many past studies used the same methods, there are also many results. This

makes easier the comparison between different materials and the discussions and validation of the results.

As regards the resistance to biological attack, many different methods and standards guarantee the evaluation of bio-susceptibility after inoculation [51], depending on the bio-deterioration agents, the exposure method and the considered conditions. Table 2.6-1 presents, as an example, only a small set of past studies that addressed the topic of bio susceptibility of agro-industrial wastes. The bibliographic research carried out for this work demonstrated that, even if the biological attack is one of the main drawbacks of agro-industrial wastes, is not so commonly addressed [16]. It has to be further evaluated since it can cause radical changes to materials' properties, compromise durability and affect human health [10,51,56]. The analysis of biological susceptibility could guarantee better use of agro-industrial wastes, as biological vulnerability could be prevented or moderated. Research should further investigate this topic, and studies related to the use of agro-industrial waste should always consider it. The already known methods to evaluate biological susceptibility could be improved and new laboratory tests proposed. Some of the already known and used could be collected to define a guideline to investigate this property. A greater number of studies that assess this property would also allow easier comparisons between different materials therefore an easier understanding of the results.

## 2.6.4 Conclusions

The present work reports laboratory tests to evaluate hygrothermal properties and biological susceptibility of agro-industrial wastes, considered as raw materials and in composites. It aims at providing a first review to be used as support for future research works. This work allows reaching the following conclusions:

- There are no specific standards to analyse agro-industrial wastes as raw materials. Past studies evaluated hygrothermal properties and bio susceptibility according to different methods.
- Past research usually analysed composites made up of agro-industrial wastes according to the standards used for conventional materials. This facilitates the comparison between them but can be complemented.
- Biological susceptibility is not so commonly assessed. Being one of the main drawbacks of using agro-industrial wastes, it should be further investigated.

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## 2.7 Concluding remarks

Thanks to the data collection, analysis, and conclusions of the past section, a motivated selection of bio-aggregates, adhesives, and additives was made. Grape press waste, hazelnut shells, olive press waste, and spent coffee grounds were chosen as bio-aggregates due to their local production and availability. For adhesives, a hemihydrate gypsum-base binder, a sodium silicate solution, and a starch-based glue were selected. To mitigate the drawbacks of bio-based composites, air lime, and citric acid were added as additives.

Considering many past studies that produced bio-based boards and panels, it was possible to get in touch with several suitable production processes, pre-treatments, and treatments of the final products. This information allowed the selection of the most suitable pre-treatment of the bio-aggregates and the boards' production process. However, this theoretical information needed an implementation by practical experiences, described in the following sections.

The review carried out on laboratory tests demonstrated that there are different methods to analyse bio-wastes' properties as raw materials and in composites. However, past research usually considered the standards used for conventional materials. Hence, although this could be useful to carry out comparisons, for some properties more details should be provided. This could ensure the characterization of bio-based raw materials and composites by unified and standardized methods, more suitable for this type of materials. However, it seems that gradually, even at the level of standards, changes are taking place by including the analysis of unconventional materials.

Some of the already performed laboratory test methods were investigated. Thus, a global vision of the experimental campaigns that could be performed was provided. Hygrothermal properties were mainly considered, due to the focus on indoor comfort. However, as both the hygrothermal and acoustic properties strictly affect indoor conditions, the acoustic absorption capacity was considered in the next studies and practical tests. Furthermore, biological susceptibility was addressed due to its importance for these types of building products and for indoor air quality and inhabitants' health.

Finally, in addition to the important information for the development of the Ph.D., these first steps of the project have been extremely useful in learning how to analyse scientific documents and how to summarize the literature review for scientific purposes, providing a first experience of scientific review dissemination. In conclusion, the initial literature review made an important contribution both in justifying the choices to carry out the project and in the development of a scientific research method.



## ANALYSIS OF SOME SELECTED MATERIALS ON THE BIO-BASED BOARDS' EFFICIENCY

### 3.1 Preamble

After the literature review, laboratory tests to evaluate the properties of the selected materials and the feasibility of producing boards were performed. The analysis carried out on the bio-aggregates and the different adhesives is reported and detailed in this section.

As a lack of information concerning the properties of the selected agro-industrial wastes was detected (Section 2.2), the first experimental campaign was developed to characterize them as raw materials. Hence, some properties of grapes and olives press waste, hazelnut shells, and spent coffee grounds were analysed; maritime pine wood chips were selected as the control material, representing wood waste. This choice was made to ensure an easier comparison and validation of the results, as, in the literature, there is more information on wood chips' properties rather than on the considered agro-industrial wastes. Furthermore, wood chips were considered as reference material for bio-susceptibility tests to validate the analysis, as better described in Section 3.2.2.

The previous literature review, described in Section 2.6, ensured the identification of the different laboratory tests to characterize the bio-wastes. Furthermore, the data collection allowed knowing the reference past studies to develop the comparisons and validate the results, to define the requirements of the standards, and to decide the grain size that could guarantee the feasibility of producing boards.

After preparing the bio-wastes, grain size, loose bulk density, thermal conductivity at different RH, water absorption, hygroscopic, and bio-susceptibility to mould and termites were analysed. The physical properties were assessed by considering and adapting different methods. Grain size analysis was carried out according to the recommendation of the RILEM

Technical Committee 236-BBM "Bio-aggregate-based Building Materials" (Amziane et al., 2017) and to EN 933-2 (EN 933-2, 2020). The particle size distribution curve was determined as the average values of three samples for each bio-waste. Loose bulk density was determined both according to Amziane et al. (2017) and Laborel-Préneron et al. (2017), and considering EN 1097-3 (1998) as the ratio between the mass and the volume of the raw materials. The thermal conductivity was determined by considering the transient method and testing the samples at two different RH levels; the results confirmed that, as expected, the thermal conductivity increased with RH (Liuzzi et al., 2017). Finally, the water absorption, hygroscopic properties, and moisture buffering value were evaluated by adaptations of the recommendation of RILEM TC (Amziane et al., 2017) and of ISO 24353 (2008), and the studies of Laborel-Préneron et al. (2017) and Rode et al. (2005), respectively. As for the bio-susceptibility tests, specific methods were performed, considering standardized ones and past studies. Mould growth was analysed according to ASTM D5590-17 (2017) and Parracha et al. (2021); termites' attack was assessed considering the information described by Nunes and Duarte (2017).

The selected adhesives play an important role in the boards' final performance, affect the feasibility of the production, and influence their embodied energy. Thus, an experimental campaign was dedicated to assessing their properties. Once analysing the bio-aggregates, the addition of the different adhesives and the composites' properties were considered. As previously anticipated (Section 2.4), three different adhesives were considered: a hemihydrated gypsum-based binder, a sodium silicate solution, and a starch-based natural glue. All of them met the considered goals:

- avoid the use of harmful products;
- guarantee low energy consumption in samples' production;
- allow drying and curing at low temperatures;
- produce samples by using the same adhesive for the different aggregates;
- avoid bio-aggregates degradation;
- moderate the biological susceptibility.

Several practical tests were performed to define the most suitable production process and the percentages of aggregates-adhesive. The aim was to maximize the quantities of bio-aggregates and minimize the amount of adhesive, guaranteeing the feasibility of producing boards by using all the considered bio-wastes. The experimental campaign provided a general characterization by assessing different properties: visual observation and pH, apparent density, thermal conductivity, hygroscopicity, and resistance to mould without inoculation. Literature values are considered to validate and discuss the results. This part was detailed in Section 3.3.

## 3.2 Article C - Bio-wastes as aggregates for eco-efficient boards and panels: Screening tests of physical properties and bio-susceptibility

The information reported in this Section was presented in the conference CEES 2021, International Conference on Construction, Energy, Environment and Sustainability, Coimbra, Portugal, 2021, and published in the conference proceeding (Cintura, E., Nunes, L., Faria, P., 2021. Characterization of agro-wastes to be used as aggregates for eco-efficient insulation boards, in: CEES 2021 - International Conference on Construction, Energy, Environment and Sustainability. ISBN: 978-989-54499-1-0, Coimbra, Portugal). Then, an extended version was published in Infrastructures 7 (2022) 26 - <https://doi.org/10.3390/infrastructures7030026>

### Abstract

Screening tests were developed or adapted from RILEM recommendations, standards and past studies, and carried out to characterize some agro-industrial wastes and to assess their feasibility as aggregates for eco-efficient building composites. Spent coffee grounds, grape and olive press waste and hazelnut shells were used, as well as maritime pine chips as control material. Particle size distribution, loose bulk density, thermal conductivity and hygroscopicity properties were analysed. The selected bio-wastes did not show good thermal insulation properties if compared with some bio-wastes already studied and used for thermal insulation composites. Values of loose bulk density and thermal conductivity were between 325.6–550.5 kg/m<sup>3</sup> and 0.078–0.107 W/(m. K); moisture buffering values higher than 2.0 g/(m<sup>2</sup>.%RH). Biological susceptibility to mould and termites were also tested, using not yet standardized methods. The low resistance to biological attack confirms one of the greatest drawbacks of using bio-wastes for building products. However, final products properties may be changed by adding other materials, pre-treatments of the wastes and the production process.

**Keywords:** agro-industrial waste; bio susceptibility; board; coffee ground; grape press waste; hazelnut shell; insulation; olive pomace; panel; thermal properties.

### 3.2.1 Introduction

The global climate is changing as a consequence of human activity and the problem has become so serious that the European Commission is calling for a climate-neutral Europe by 2050 [1]. To reach such an ambitious goal, the strategic importance of the construction sector is

clear. Indeed, the building industry is recognized to have a strong environmental impact for several reasons. Among them are energy consumption, production of solid waste and harmful gases, and the lack of insulation of the existing building stock, responsible for high energy losses [2–4]. The urgent need for more sustainable building practices is leading to increased research focused on the production, transport and use of building materials that can be shown to have a lower impact on the environment [5,6]. In this context, eco-friendly insulation materials and composites emerge as an interesting solution. They can reduce energy consumption, mitigate the production of harmful wastes, reduce the waste volumes to manage and passively control hygrothermal conditions, improving indoor air quality and comfort [7,8].

Nowadays, a significant amount of research focuses on the feasibility of using bio-wastes to produce several building materials, such as binders, mortars and concrete, plasters, masonry blocks, insulation boards, and coating panels. That is due to the large production of bio-wastes and their good potential in construction practices [9,10]. The use of bio-wastes can both lower production costs and encourage circular economy practices, as well as being an innovative method for waste disposal [5,11–13].

Several recent studies attempted to evaluate the feasibility of producing composite boards using bio-wastes. For example, Eschenhagen et al. [14] analysed boards made up of *Miscanthus* (*Miscanthus* spp.) fibres, sunflower (*Helianthus annuus* L.) stalks and natural binders (starch-based binder, wood glue made of casein and bone glue made of gelatine). Composites were obtained with a density of 190 kg/m<sup>3</sup> (average value) and thermal conductivity between 0.057 W/(m. K) and 0.068 W/(m.K) for *Miscanthus* fibres, 280 kg/m<sup>3</sup> and 0.065–0.077 W/(m.K) for sunflower stalks. Considering the reference value of 0.065 W/(m.K) as the maximum thermal conductivity of a good thermal insulator material [15], these bio-wastes showed potential for insulation boards production. Ali et al. [16] evaluated the possibility of making boards using wheat and agave straw bonded with corn starch. The resulting composites also showed good potential as insulating boards with thermal conductivity values of about 0.052 W/(m.K). Nunes et al. [17] produced cement-bonded particleboards replacing maritime pine (*Pinus pinaster* Ait.) chips with banana tree (*Musa* sp.) pseudostem waste. The researchers demonstrated that the increase in banana fibre content from 0% to 75% (by weight) reduced thickness swelling from 0.38% to 0.11%, improving dimensional stability.

Among the large number of past studies that considered bio-wastes for boards' production, only a few detailed the properties of the bio-aggregates individually. Table 3.2-1 presents a summary of the particle size of bio-aggregates already used for boards' production.

Table 3.2-1. Particle size of bio-aggregates already studied for boards' production.

| References               | Description                                                                                                                                                                                                                                                                                                                                          | Particle Size (mm) |
|--------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------|
| Wong et al. [18]         | Particleboard of grapevine and pine chips (0.25–1.00 mm particles used for the surface layer; 1.00–6.00 mm for the core) and mixed with melamine modified urea-formaldehyde (MUF). Three layers were formed, and hot-pressed ( $T = 170^{\circ}\text{C}$ , pressure = 3.6 MPa, time = 5 min).                                                        | 0.25–6.00          |
| Binici et al. [19]       | Insulation composite of sunflower stalk fibre, sunflower stalk sponge and wheat stalks shredded to reach dimensions of 5–10 mm, vermiculite and gypsum as a binder. Materials were dry mixed, the water was added, and the fresh mortars were placed in moulds in three layers and compacted.                                                        | 5–10               |
| Liuzzi et al. [20]       | Insulation composite of straw fibres (size = 30 mm) and olive fibres (size = 20 mm) mixed with a sodium silicate solution $\text{Na}_2\text{O}_n(\text{SiO}_2)$ without pressing. The materials were cured at environmental conditions for 28 days and finally dried at $50^{\circ}\text{C}$ until constant mass.                                    | 20–30              |
| Eschenhagen et al. [14]  | Insulation composite of Miscanthus and sunflower stalk fibre grounded with a 1.5 cm screen and sieved to obtain fibres equal or bigger than 10 mm, starch-based binder, wood glue made of casein and bone glue made of gelatine.                                                                                                                     | $\geq 10$          |
| Pavelek and Adamová [21] | Insulation boards consisted of a premanufactured panel filled with shredded rapeseed ( <i>Brassica napus</i> ) and woodchips from coniferous trees (sandwich panel).                                                                                                                                                                                 | <0.25–8            |
| Mati-Baouche et al. [22] | Insulation composite of sunflower stalks (shredded and sieved to obtain particle size between 1 and 6.3 mm) and chitosan from shrimp shell. The materials were mixed, pressed at $20^{\circ}\text{C}$ for 1 min (pressure between $1 \times 10^{-3}$ - $32 \times 10^{-3}$ MPa) and then dried in an oven ( $T = 50^{\circ}\text{C}$ , time = 50 h). | 1–6.3              |
| Pásztor et al. [23]      | Insulation composite of chipped black locust ( <i>Robinia pseudoacacia</i> ) tree bark and urea-formaldehyde (UF) based resin, by pressing at $T = 120^{\circ}\text{C}$ for 6 sec per final mm thickness.                                                                                                                                            | <1–45              |
| Binici et al. [24]       | Insulation composite of corn stalks (particle size between 0.5–4 mm), epoxy resin, gypsum and Portland cement (CEM I 42.5). Materials were mixed for 5 min, compacted ( $T = 20^{\circ}\text{C}$ , time = 1 min, pressure = 0.07–0.27 MPa) and dried at $50^{\circ}\text{C}$ for 50 h.                                                               | 0.5–4              |
| Wang et al. [25]         | Cement-bonded particleboard made up shredded grapevine stalk, cement and 3% calcium chloride ( $\text{CaCl}_2$ ) by weight of cement. The boards were cold pressed (pressure = 1.25 MPa, time = 8 h) and then conditioned at $T = (20 \pm 2)^{\circ}\text{C}$ , RH = $(65 \pm 5)\%$ for 28 days.                                                     | 9–80               |
| Kusumah et al. [26]      | Particleboard of sweet sorghum ( <i>Sorghum bicolor</i> L. Monech) bagasse and citric acid dissolved in water as adhesive. Citric acid was sprayed onto the dried particles and the boards are hot-pressed ( $T = 200^{\circ}\text{C}$ , time = 10 min, maximum pressure = 6.5 MPa).                                                                 | 0.9–5.9            |
| Buratti et al. [27]      | Panels of rice husk (length 9 mm, width of 1 mm) and cold-water-based polyurethane glue (density = $1000 \text{ kg/m}^3$ ). For comparison samples of cork (size 0.8 mm–1 mm) and glue were fabricated (they were cured at $T = 100^{\circ}\text{C}$ , time between 90–120 min).                                                                     | 9<br>0.8–1         |

Particle size is one of the most studied properties of aggregates but remains unknown for many bio-wastes. The reported methods and experimental tests tend to focus on the composites [10]. The analysis of the bio-wastes should be further investigated since their characteristics will be certainly linked to the performance of the final products [28]. Knowing bio-wastes properties allows more conscious choices to be made to achieve the final requirements for the produced composites [29]. Along with the European Standards for aggregates in general, the Recommendations of RILEM Technical Committee 236-BBM "Bio-aggregate-based building materials" [30] have been widely used to study properties of bio-aggregates. For example, Page et al. [31] used them to characterize hemp shiv and flax fibres; Laborel-Préneron et al. [32] to analyse barley straw, hemp shiv and corn cob; Antunes et al. [33] to evaluate rice husk properties; Barbieri et al. [34] to study wheat husk. Using the same methods allows conforming the procedures and guarantees an easier comparison between different materials. However, complementary characterization may be important and further test procedures may be needed. As bio-based products are frequently vulnerable to biological deterioration, bio-susceptibility tests are a good example of this need.

Due to their organic composition, and factors like the presence of nutrients and micro-organisms in the raw materials, as well as typical high hygroscopicity, bio-based wastes are often prone to biological deterioration [35,36]. Bio susceptibility is indeed recognised as one of the main drawbacks of bio-based building materials [37,38]. It can lead to modified properties and eventually to reduced durability. In particular, besides the obvious aesthetic impact, mould growth on interior applications can also lead to health risks for building users, namely respiratory diseases, such as asthma and allergic rhinitis [39]. Insect attack can also be a major cause of deterioration of building materials, thus worsening their performance. In the case of bio-based components and structures, subterranean termites represent the highest hazard in Mediterranean countries [40,41] and can be used as model organisms to evaluate susceptibility. The biological resistance of raw materials provides useful information and can help to forecast their impact on the final performance of composite materials. Thus, it is an important property that has to be considered to optimize the design of bio-based products.

Taking into consideration the previous information, this article reports the characterization of four bio-wastes analysed individually to determine their properties as raw materials: spent coffee grounds, grape press waste, olive press waste and hazelnut shells. Typically, spent coffee grounds are usually disposed of in landfills, used as composting and fermentation or as animal feed, such as olive and grape pomace [42,43]. Hazelnut shells are used for combustion and heating [44].



Maritime pine (*Pinus pinaster* Ait.) chips were considered as control material, as its characteristics are better known and recycling is frequent, namely from construction and demolition waste. These bio-wastes were chosen considering the referenced feasibility of using them as building materials and their world production, focusing on Euro-Mediterranean [10]. The use of local materials decreases the environmental impact derived from the transport and consumption phases and secures high availability. The results were compared with other bio-based materials already studied and used as aggregates (cork waste particles, corn cob particles, hemp shiv, and rice husk), namely for producing boards and panels. The potential of the analysed bio-wastes as aggregates for this type of composite production and, how to optimize their use, was discussed. This work also aims at both describing and discussing the methods to evaluate properties of bio-aggregates, some not so common, as the cases of bio-susceptibility tests, and reporting the obtained properties, comparing with literature whenever possible. The study reduces the gaps of knowledge found in literature and evaluates the feasibility of using complementary bio-wastes to produce composite boards and panels.

## 3.2.2 Materials and methods

### 3.2.2.1 Materials

Spent coffee grounds were provided by the central bar of the National Laboratory of Civil Engineering (LNEC), Lisbon, Portugal. They are usually thrown away for waste disposal. They were air-dried at room temperature for 6 weeks over an absorbent paper, regularly stirred to improve the dry rate. Grapes and olive press wastes were provided by Esporão company located in Reguengos de Monsaraz, Portugal. They were spread over a plastic tarp for 7 and 10 weeks, respectively, to air dry and also regularly mixed to allow better ventilation. Olive press waste needed a longer drying period due to the initial high amount of liquid content that, according to the producer, depends on the olives' quality of each season. Hazelnut shells were provided by Borges Agricultural & Industrial Nuts SA company, located in Reus, Spain. This bio-waste was used as received. Details on the cellulose, hemicellulose and lignin content of these bio-wastes have been described previously by Cintura et al. [10], such as their disposal and use.

To simulate recycled pine wood a maritime pine board, obtained from a local wood shop (Lisbon, Portugal), was cross cut and then shredded in a laminar mill five times to reach a chip dimension of less than 10 mm. Figure 3.2-1 shows the analysed materials and their colour scale according to the PANTONE Uncoated RGB scale [45].

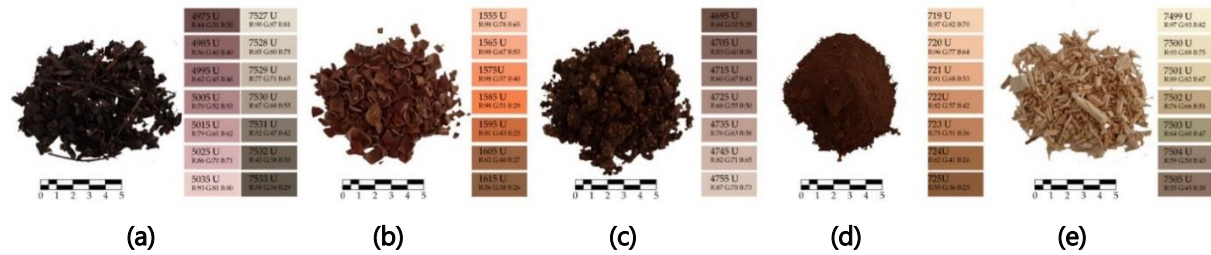


Figure 3.2-1. Bio-wastes size and colour scale after air drying: (a) grape press waste; (b) hazelnut shells; (c) olive press waste; (d) spent coffee grounds; (e) maritime pine chips.

After air drying, the materials were sampled as described by Amziane et al. [30], they were placed on a flat surface into a pile that was divided into quarters. Two parts were selected and further divided into quarters. This procedure was repeated until having enough material to carry out the laboratory tests. Sampling guarantees a better mixture of particles, avoiding the segregation between the coarser and finer ones.

### 3.2.2.2 Methods

Experimental tests were performed after drying the bio-wastes at 60 °C until a constant mass was reached (change in mass of less than 0.1% over 24 h), except for bio-susceptibility tests for which the bio-wastes were dried also at 103 °C for 24 h. Then, they were stabilised in a conditioning room ( $T = (20 \pm 2) ^\circ\text{C}$ ,  $(60 \pm 5) \%$  relative humidity (RH)), adapting the method proposed by Amziane et al. [30] to equilibrate at known laboratory conditions. All the laboratory tests were performed without pressing the bio-aggregates. Table 3.2-2 summarizes the tests performed, the samples and relevant references. Further details are presented in the following sections.

Table 3.2-2. Summary of laboratory tested properties, samples, considered/adapted references and standards.

| Properties          |                            | Samples                                                                                                               | References and Contribution                                                                                                                                               | Description                                                             | Strengths                                                                                               | Limitations                                                                                                                                        |
|---------------------|----------------------------|-----------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------|
| Physical properties | Particle size distribution | 3 samples of 100 g of the materials                                                                                   | Amziane et al. [30] described the method and reported EN 933-2 [46]                                                                                                       | Sieving methods by using 200 mm diameter sieves                         | Widely used method: easier comparison between different aggregates considering the same laboratory test | Sieving time is not specified; for the considered agro-industrial wastes, a sample of $45 \pm 10$ g in case of 200 mm diameter sieves is too small |
|                     | Loose bulk density         | Cylindrical glass (diameter = 6 cm height = 11 cm) filled with 3 samples of the material                              | Amziane et al. [30] introduced the method; Laborel-Préneron et al. [32] provided a detailed description too                                                               | By considering aggregates' weight and the corresponding volume of water | Applicability to different bio-aggregates                                                               | It could be less rapid and immediate than EN 1097-3 [47] (replacement of the volume of aggregate with a volume of water)                           |
|                     |                            | Cylindrical plastic container (100 mL) filled with 3 samples of the material                                          | EN 1097-3 [47] described the laboratory test                                                                                                                              | The ratio between mass and volume                                       | Fast laboratory test                                                                                    | Not specific for bio-aggregates                                                                                                                    |
|                     | Thermal conductivity       | 5 different points of the surface of an open container (diameter = 18.50 cm, high = 5.80 cm) filled with the material | Antunes et al. [33] evaluated the thermal conductivity of bio-aggregates by using the transient method; Liuzzi et al. [20] described its advantages and reasons to use it | Transient method                                                        | Possibility of analysing small samples (faster stabilization at different RH) and loose aggregates      | Fast evaluation; the results could be as not precise as other methods (e.g., heat flow)                                                            |
|                     | Water absorption           | 3 samples of 25 g of the materials                                                                                    | Amziane et al. [30] introduced the method; use and                                                                                                                        | Difference between dry and wet mass                                     | Possibility of testing loose aggregates                                                                 | No specification of opening mesh (permeable bag could be                                                                                           |

| Properties         |                                | Samples                                                                                                               | References and Contribution                                                                                                              | Description                        | Strengths                                                                                                                                | Limitations                                                                                                                                          |
|--------------------|--------------------------------|-----------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------|
|                    |                                |                                                                                                                       | details are described by Laborel-Préneron et al. [32]                                                                                    |                                    |                                                                                                                                          | better described) and of water's temperature; possible compaction of the aggregates; rotation speed should be automatically controlled               |
|                    | Sorption/desorption properties | 3 plastic box (11.0 cm × 14.8 cm, high = 2 cm) with an open-top surface filled with the materials                     | ISO 24353 [48] described the laboratory test; Rode et al. [49] provided the MBV classification                                           | Samples at different RH            | Same sorption/desorption time (12 h); not developed considering indoor conditions in a specific area (differently from Rode et al. [49]) | The lack of a numeric classification in ISO 24353 [48] makes difficult the comparison between different aggregates and the discussion of the results |
| Bio susceptibility | Mould                          | 5 + 5 Petri dishes (diameter = 9 cm) filled with similar volumes of materials. Two exposure methods                   | ASTM D5590-17 [50] provided information about the test and the evaluation; Parracha et al. [51] detailed the adaptations to the material | Evaluation of contaminated surface | Different exposure methods could be considered; the control secures the validation of the tests                                          | Subjective/visual evaluation                                                                                                                         |
|                    | Termites                       | 3 Petri dishes (diameter = 9 cm), moistened calibrated sand on one side and similar volumes of materials on the other | Nunes and Duarte [52] provided information to develop the test                                                                           | Termites' survival                 | Innovative laboratory test to analyse loose aggregates                                                                                   | Control of moulds development for high sugar content materials                                                                                       |

### 3.2.2.2.1 Particle size analysis, loose bulk density and thermal conductivity

Grain size analysis was carried out using the mechanical sieves method, considering Amziane et al. [30], as mentioned in Table 3.2-2. Due to the different grain sizes of the studied materials, the apertures of the successive openings of the sieves covered a wide range (from 10 mm to 0.125 mm) in accordance with EN 933-2 [46]. The sieving time, not specified in the considered references, was 3.5 min. The particle size distribution curve was determined by considering the average values of the three samples.

Bio-wastes loose bulk density was calculated considering two different methods. The first one considered the method presented by Amziane et al. [30] and Laborel-Préneron et al. [32]. An empty cylindrical glass was weighed and then filled with the material until half the volume. The container was sealed and upend ten times and shaken to obtain a horizontal surface. The level of the material was marked. The material was removed, and the container was filled with water and weighed. The loose bulk density was determined by Equation (3.2-1). The average value of the three tested samples was considered significant when the coefficient of variation was less than 5%.

$$\rho_{\text{Aggregate}} \left( \frac{\text{kg}}{\text{m}^3} \right) = \frac{m_{\text{Aggregate}}}{m_{\text{H}_2\text{O}}} \rho_{\text{H}_2\text{O}} \quad (3.2-1)$$

The second method, based on EN 1097-3 [47], consisted of calculating the ratio between the mass and the volume of the materials. A cylindrical container with a known volume ( $V$ ) was weighted ( $m_1$ ), filled by dropping the material from a height of 40 cm, shaken to obtain a horizontal surface without compacting the material, and weighted again ( $m_2$ ). The loose bulk density was calculated by Equation (3.2-2).

$$\rho_{\text{Aggregate}} \left( \frac{\text{kg}}{\text{m}^3} \right) = \frac{m_2 - m_1}{V} \quad (3.2-2)$$

Measurements of thermal conductivity in building materials may be performed through steady-state methods or transient methods. In this work, a transient method was adopted, considering the method reported by Antunes et al. [33] and Liuzzi et al. [20], as reported in Table 3.2-2. The transient method offers several benefits [53,54], in particular the smaller dimension of the testing area. This allowed performing the measurement in different parts of the surface of each sample, indicated in Figure 3.2-2a. Bio-wastes' thermal conductivity was measured by using an ISOMET 2104 Heat Transfer Analyser with a 60 mm diameter contact probe API 210412, ranging values between 0.04 and 0.30 W/(m.K). To evaluate the correlation between the thermal conductivity and RH, the samples were stabilised in a climatic chamber (Fitoclima 300 EDTU) at  $T = 23^\circ\text{C}$ ,  $\text{RH} = 50\%$ , then  $T = 23^\circ\text{C}$ ,  $\text{RH} = 75\%$ . When constant mass was reached (change in mass of less than 0.1% over 24 h), thermal conductivity was calculated, protecting the samples from the airflow during the measurement by a cover box (Figure 3.2-2b). To validate this method, mass was controlled before and after the test to

guarantee a variation of less than 0.1%. The average value of the five measurements was considered significant if the coefficient of variation was less than 5%.

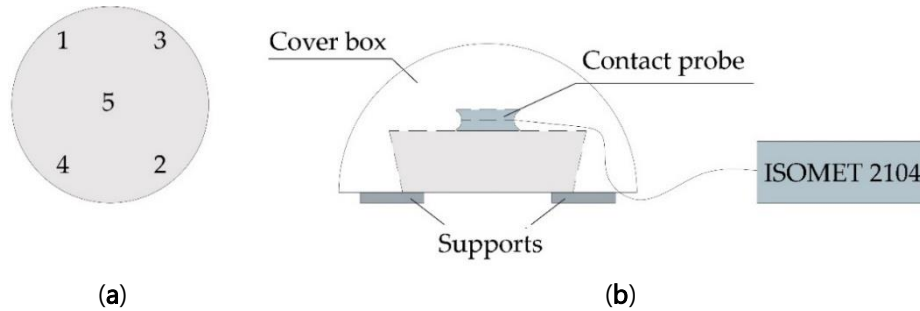


Figure 3.2-2. (a) Points where thermal conductivity was calculated; (b) Method to calculate thermal conductivity (adapted from Cintura et al. [29]).

#### 3.2.2.2.2 Water absorption and sorption/desorption properties

For the water absorption test, based on Amziane et al. [30] and Laborel-Préneron et al. [32], an empty permeable bag was put in water until reached complete wetting. Then it was put in a salad spinner, turned 100 times (approximatively two rotations/second) and tared. It was filled with 25 g of each of the bio-wastes ( $m_0$ ) and immersed in water. After one minute, the permeable bag with the waste was removed from the water, put in the salad spinner, turned 100 times again, and weighed. The permeable bag was put in the water again and this procedure was repeated after 1 min, 15 min, 4 h and then every 24 h until a constant mass was reached (change in mass less than 0.1 % over 24 h). Water absorption was determined considering Equation (3.2-3), where  $m_A$  is the wet mass.

$$W(\%) = \frac{m_A - m_0}{m_0} \times 100 \quad (3.2-3)$$

The average value of three tested samples of each material was considered significant when the coefficient of variation was less than 5%. By knowing the bulk density of the bio-wastes, it was possible to determine the ratio between the volume of the absorbed water and the volume of the considered bio-wastes. The water absorption of the bio-wastes allows making considerations about the influence they may have on the workability of future composites.

The hygroscopicity of the bio-wastes was determined by considering the moisture adsorption/desorption capacity in response to humidity variation through cyclic tests, as indicated in ISO 24353 [48]. The plastic boxes filled with the materials (open-top surface =  $A$ ), were preconditioned at  $T = (23 \pm 2)^\circ\text{C}$  and  $\text{RH} = (63 \pm 2)\%$  in a climatic chamber (Fitoclima 1000) until constant mass was reached (change in mass of less than 0.1% over 24 h) and weighed ( $m_0 = m_{d(n-1)}$ ). Then they were conditioned at  $T = (23 \pm 2)^\circ\text{C}$ ,  $\text{RH} = (75 \pm 2)\%$  for 12 h and weighed every 3 h (moisture sorption process,  $m_n$ ), and at  $T = (23 \pm 2)^\circ\text{C}$ ,  $\text{RH} = (50 \pm 2)\%$  for 12 h (moisture desorption process,  $m_{dn}$ ).

The cyclic value of the moisture adsorption content,  $Q_{A,ac}$  and the moisture desorption content,  $Q_{A,dc}$  were calculated according to Equations (3.2-4) and (3.2-5) to determine the variation of moisture adsorbed/desorbed content over time.

$$\rho_{A,ac} \left( \frac{\text{kg}}{\text{m}^2} \right) = \frac{m_{an} - m_{d(n-1)}}{A} \quad (3.2-4)$$

$$\rho_{A,dc} \left( \frac{\text{kg}}{\text{m}^2} \right) = \frac{m_{an} - m_{dn}}{A} \quad (3.2-5)$$

The moisture content difference between adsorption and desorption is calculated considering Equation (3.2-6).

$$\rho_{A,sc} \left( \frac{\text{kg}}{\text{m}^2} \right) = \rho_{A,ac} - \rho_{A,dc} \quad (3.2-6)$$

For easier comparison of the results with literature values, the moisture buffering value (MBV) was also calculated, adapting the method defined by Rode et al. [49]. MBV is the average value between MBV for the sorption phase ( $MBV_a$ ) calculated as reported in Equation (3.2-7), and MBV for the desorption phase ( $MBV_d$ ) calculated as reported in Equation (3.2-8). The last three cycles and the average values of each bio-waste were considered.

$$MBV_a \left( \frac{\text{g}}{\text{m}^2 \% \text{ RH}} \right) = \frac{m_{an} - m_{d(n-1)}}{A \times (RH_{high} - RH_{low})} \quad (3.2-7)$$

$$MBV_d \left( \frac{\text{g}}{\text{m}^2 \% \text{ RH}} \right) = \frac{m_{an} - m_{dn}}{A \times (RH_{high} - RH_{low})} \quad (3.2-8)$$

In these equations,  $m_{an}$  (g) is the value of the mass at the end of the sorption phase,  $m_{d(n-1)}$  is the value of the mass at the end of the desorption phase of the previous cycle,  $m_{dn}$  is the value of the mass at the end of the desorption phase,  $A$  ( $\text{m}^2$ ) is the exposed surface,  $RH_{high}$  is the highest value of RH (75%) and  $RH_{low}$  is the lowest one (50%, based on ISO 24353 [48], differently from Rode et al. [49]). The MBV of the analysed materials, calculated as described, can be compared with MBVs of literature even when calculated under different conditions. Indeed, the reference to RH variation included in the value makes the comparison easier [55].

#### 3.2.2.2.3 Bio susceptibility to mould and termites

To evaluate the bio-susceptibility to moulds, two different exposure methods were applied. Ten Petri dishes (diameter = 9 cm) were used for each bio-waste under test with five dishes containing 20 mL of culture media (4 % malt, 2 % agar). A similar amount of previously steam sterilized bio-wastes (approx. 13 mL) was added to each Petri dish and all plates were inoculated with 1 mL of a mixed spore suspension of *Aspergillus niger* and *Penicillium funicullosum*. The fungal strains used came from LNEC' fungal collection.

The Petri dishes with culture media were left for four weeks at  $(22 \pm 1) ^\circ\text{C}$  and  $(70 \pm 5) \% \text{ RH}$ . Five Whatman n° 1 filter papers (diameter = 45 mm) were used as controls for this exposure method.

The inoculated dishes without culture media were conditioned for the same time and at the same temperature but at 100% RH. For these, maritime pine chips were considered as reference material.

The samples were visually graded each week and mould growth was estimated considering the classification provided in Table 3.2-3. At the end of the four weeks, the samples were also visualized under a stereo microscope (Olympus SZX12) to confirm the grading results. Carrying out the bio susceptibility test with and without culture media allowed evaluating the materials both in favourable conditions for mould growth and as they are (raw materials).

Table 3.2-3. Rate of mould growth adapted from ASTM D5590-17 [50].

| Rating | Description      | Contaminated Surface (%) |
|--------|------------------|--------------------------|
| 0      | None             | 0                        |
| 1      | Traces of growth | <10                      |
| 2      | Light growth     | 10 to 30                 |
| 3      | Moderate growth  | 30 to 60                 |
| 4      | Heavy growth     | >60                      |

For testing against subterranean termites, a protocol was developed following previous work [52]. Three Petri dishes (diameter = 9 cm) for each bio-waste were half filled with approx. 7 mL of the materials and on the other half similar amounts of moistened (25% w/w) Fontainebleau™ sand were placed. Groups of 50 termites (*R. grassei*) were then introduced on each Petri dish. The termites were field collected (Sesimbra, Portugal) and maintained at optimal conditions until required for testing but never more than two months.

All tested samples were left for four weeks at  $(24 \pm 1) ^\circ\text{C}$  and  $(80 \pm 5) \% \text{ RH}$ , constantly monitored to control both termites' survival and mould growth. At the end of this period, the percentage of survival of the termites was evaluated and mould growth was qualitatively estimated. Because of the well-known durability of maritime pine, its chips were considered as controls to validate the test. A survival level lower than 50% would not be accepted [41]. Mould growth can have a limiting impact on termite survival and therefore was also closely followed.

### 3.2.3 Results and discussion

#### 3.2.3.1 Particle size analysis

Figure 3.2-3 shows the particle size distribution of the considered bio-wastes.



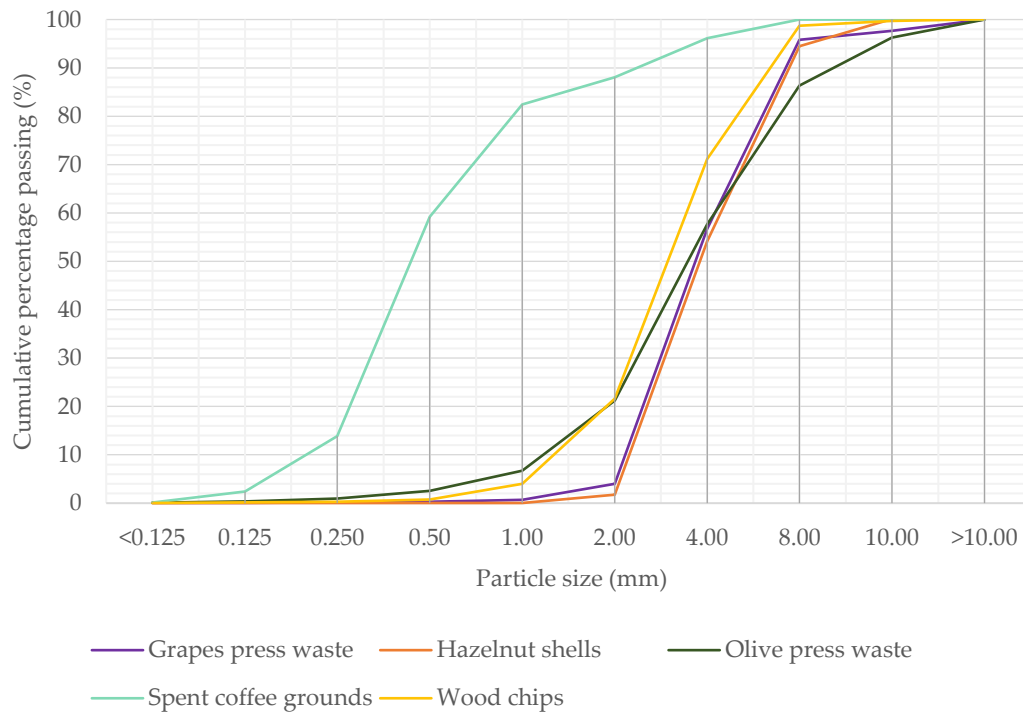


Figure 3.2-3. Particle size distribution of the tested bio-wastes and the wood chips.

Spent coffee grounds have grain sizes much lower than the other selected bio-wastes and the control; thus, for insulation boards, they might play a better role as fine aggregates. The other bio-wastes present more similar characteristics (particle size of about 1 mm–8 mm).

The comparison between the results and literature values reported in Table 3.2-1 allows evaluation if shredding the bio-wastes will be required in case of using them for boards' production. The grain size of the analysed bio-wastes is in the range of the considered past studies, in which it varies between 0.25 mm and 80 mm. Therefore, shredding the bio-wastes to produce boards does not seem to be necessary. This guarantees a more sustainable practice since the bio-wastes can be used directly as they are as by-products. The shredding may be necessary in case of compressed panels' production for which a finer grain size might be more adequate [17].

Collet [56] reported some past studies about the influence of aggregates' grain size on the thermal properties of the composites. The results demonstrate that the finer the particles, the higher the thermal conductivity. Laborel-Préneron et al. [32] demonstrated that, in earth-straw composites, the shorter the pieces of straw, the higher the compressive strength. Future studies could deepen the influence of grain size on the properties of boards since it can affect physical and mechanical properties [17,18]. The influence of sieving time on the results of the grain size analysis of bio-aggregates could be further investigated, too. The test could be carried out considering several sieving times, evaluating the differences in the results, and defining which guarantees the smallest deviation in the grain size distribution, as Martínez-García et al. [57] reported.

### 3.2.3.2 Loose bulk density and thermal conductivity

Table 3.2-4 reports the results of loose bulk density of the bio-wastes analysed by the two methods, comparing them with literature values.

Table 3.2-4. Measured loose bulk density of the analysed bio-wastes and comparison with literature values.

| Material             | Loose Bulk Density (kg/m <sup>3</sup> ) |                             | Literature Values (kg/m <sup>3</sup> ) | Reference                       |
|----------------------|-----------------------------------------|-----------------------------|----------------------------------------|---------------------------------|
|                      | According to Amzi-ane et al. [30]       | According to EN 1097-3 [47] |                                        |                                 |
| Grapes press waste   | 343.88 ± 12.00                          | 325.60 ± 5.82               | 105 ± 5 <sup>a</sup>                   | Wong et al. [18]                |
|                      |                                         |                             | 1449 ± 2 <sup>b</sup>                  | David et al. [59]               |
|                      |                                         |                             | 1420 ± 3 <sup>c</sup>                  | David et al. [59]               |
| Hazelnut shells      | 550.50 ± 19.53                          | 549.66 ± 11.92              | 230                                    | Çöpür et al. [60]               |
| Olive press waste    | 449.43 ± 8.60                           | 427.32 ± 9.62               | 1251 <sup>d</sup>                      | Liuzzi et al. [61]              |
|                      |                                         |                             | 616 <sup>e</sup>                       | del Río Merino et al. [62]      |
| Spent coffee grounds | 478.81 ± 5.40                           | 447.73 ± 5.13               | 380 ± 20 <sup>f</sup>                  | Massaro Sousa and Ferreira [63] |
| Wood chips           | 256.98 ± 8.10                           | 250.20 ± 4.59               | 130 ± 2 <sup>g</sup>                   | Wong et al. [18]                |

<sup>a</sup> Particles of grapevine milled to a thickness of <1 mm particles; <sup>b</sup> Wine pomace; <sup>c</sup> Vine shoots;

<sup>d</sup> Olive pruning waste; <sup>e</sup> Olive stone; <sup>f</sup> Grain sizes between 600 and 500 µm; <sup>g</sup> Commercial pine.

Values of the considered bio-wastes have the same order of magnitude of literature ones. The main differences derive from different types and particle sizes that widely affects the values of loose bulk density [58]. Wood chips show the lowest values of loose bulk density, as expected. Grape press waste could be the best aggregate to produce thermal insulation boards since it has the lowest loose bulk density value between the bio-wastes.

Considering the materials chosen for comparison, granular cork waste derived from cork panels, bottles cap manufacturing, cork industry (not expanded cork) showed values of about 150–160 kg/m<sup>3</sup> [64]. For expanded cork granulate, Brás et al. [65] reported values of 112 kg/m<sup>3</sup> and Nóvoa et al. [66] values of 220 kg/m<sup>3</sup>. For corn cob, Ansell et al. [58] obtained values of about 344–406 kg/m<sup>3</sup>. Laborel-Préneron et al. [32] evaluated corn cob and hemp shiv, achieving results of 497 kg/m<sup>3</sup> and 153 kg/m<sup>3</sup>, respectively. For hemp shiv, Page et al. [31] reported values of 110 kg/m<sup>3</sup>. Antunes et al. [33] studied rice husk, reporting a loose bulk density of 85 kg/m<sup>3</sup>. The selected bio-wastes have a higher loose bulk density, except for corn cob evaluated by Laborel-Préneron et al. [32]. In this case, only hazelnut shells show higher values. Grape press waste has a loose bulk density closer to the one of corn cob evaluated by Ansell et al. [58].

Table 3.2-5 shows the values of thermal conductivity of the analysed bio-wastes. The thermal conductivity values are higher than the required to consider a material as thermal insulation, namely not lower than 0.065 W/(m.K) [15]. None of the tested bio-wastes can be classified as such. To produce boards with good thermal resistance using these bio-aggregates, it will be necessary to combine them with other materials with good thermal insulation properties or produce them in a way

that air is entrapped within the composite in small volumes. Grape press waste has the lowest thermal conductivity, similar to one of the wood chips: it seems the most promising bio-aggregates to produce composites with good thermal insulation performance. Taking into account the correlation with loose bulk density, thermal conductivity values are in line with the expected since there is a direct proportionality [67]. Hazelnut shells have the worst thermal insulation behaviour (the highest thermal conductivity), having the highest loose bulk density; grape press wastes and maritime pine chips show the lowest values of thermal conductivity, having the lowest values of loose bulk density.

Table 3.2-5. Thermal conductivity of the analysed bio-wastes and comparison with literature values.

| Material             | Thermal Conductivity (W/(m.K)) |                     | Literature Values (W/(m.K)) | References           |
|----------------------|--------------------------------|---------------------|-----------------------------|----------------------|
|                      | T = 23 °C, RH = 50%            | T = 23 °C, RH = 70% |                             |                      |
| Grape press waste    | 0.078 ± 0.002                  | 0.081 ± 0.002       | -                           |                      |
| Hazelnut shells      | 0.107 ± 0.003                  | 0.115 ± 0.003       | 0.1                         | Çuhadaroglu [68]     |
| Olive press waste    | 0.089 ± 0.004                  | 0.097 ± 0.003       | -                           |                      |
| Spent coffee grounds | 0.092 ± 0.002                  | 0.099 ± 0.005       | 0.2                         | Lachheb et al. [69]  |
| Wood chips           | 0.077 ± 0.001                  | 0.082 ± 0.002       | 0.0568-0.0629 <sup>a</sup>  | Cetiner and Shea [7] |

<sup>a</sup> Materials particle size between 1 mm–4 mm, conditioned at RH = 50%.

Considering the materials chosen for comparison, all the bio-wastes have higher values of thermal conductivity, except for the one of corn cob. However, the test conditions of temperature and RH are frequently not defined.

Figures 3.2-4 and 3.2-5 show the correlations between thermal conductivity and RH, and between thermal conductivity and loose bulk density. As expected, the thermal conductivity increases proportionally to RH and the loose bulk density, for all tested bio-wastes. Figure 3.2-5 also reports the values of the materials chosen for comparison, being the lower thermal conductivity justified by lower loose bulk density.

Gomes et al. [70] and Cherki et al. [64] reported values of thermal conductivity of expanded granular cork of about 0.035–0.070 W/(m.K) and 0.049–0.050 W/(m.K), respectively. For hemp shiv, Laborel-Préneron et al. [32] reported values of 0.051 W/(m.K) while Page et al. [31] of 0.048 W/(m.K). Antunes et al. [33] evaluating rice husk achieved results of about 0.047 W/(m.K). Comparing with the results of Laborel-Préneron et al. [32], among the tested bio-wastes only hazelnut shells have a higher thermal conductivity than corn cob's one (0.096 W/(m.K)). Considering the results of Ansell et al. [58], the thermal conductivity of corn cob (0.085 W/(m.K)) is higher than grape press waste and wood chips. Despite these results, the analysed bio-wastes may be used as bio-aggregates for insulation composite boards and panels' production, if the composition and methods are able to improve the insulation performance.

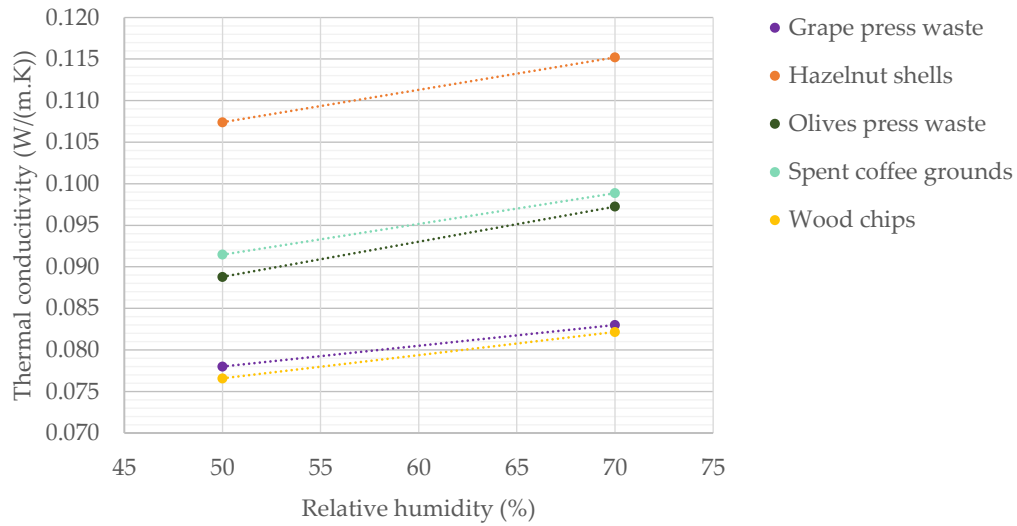


Figure 3.2-4. Correlation between thermal conductivity and RH of the tested bio-wastes.

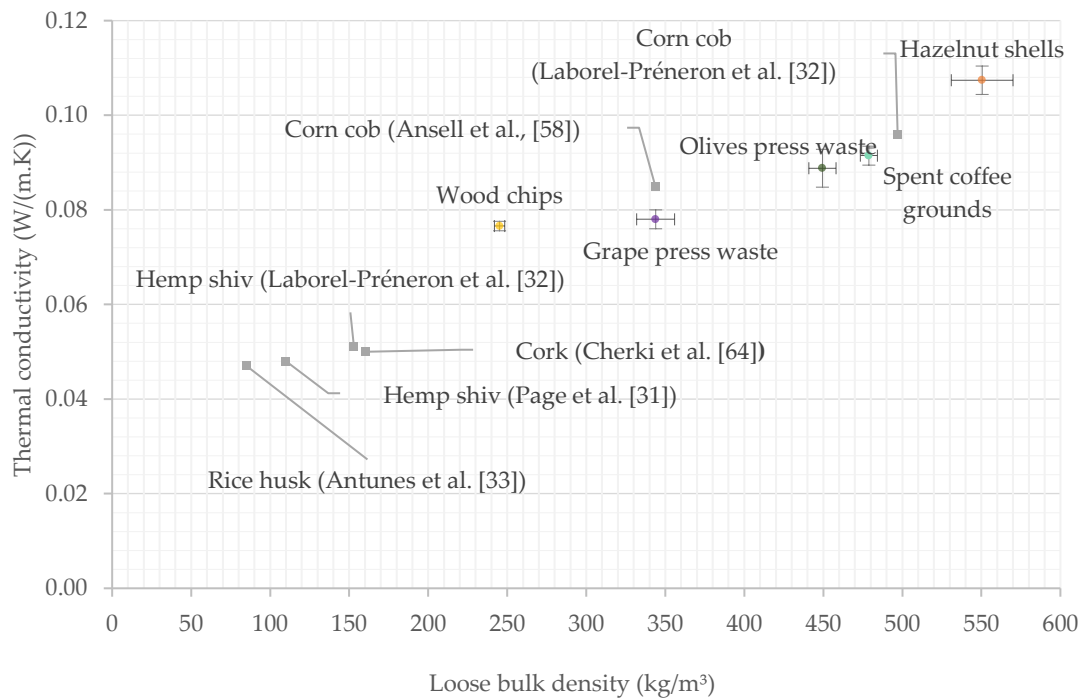


Figure 3.2-5. Correlation between thermal conductivity and loose bulk density of bio-wastes.

### 3.2.3.3 Water absorption and sorption/desorption properties

Figure 3.2-6 reports the results of water absorption of the five studied bio-wastes considering the method proposed by Amziane et al. [30], and Table 3.2-6 presents the water absorption after 48 h.

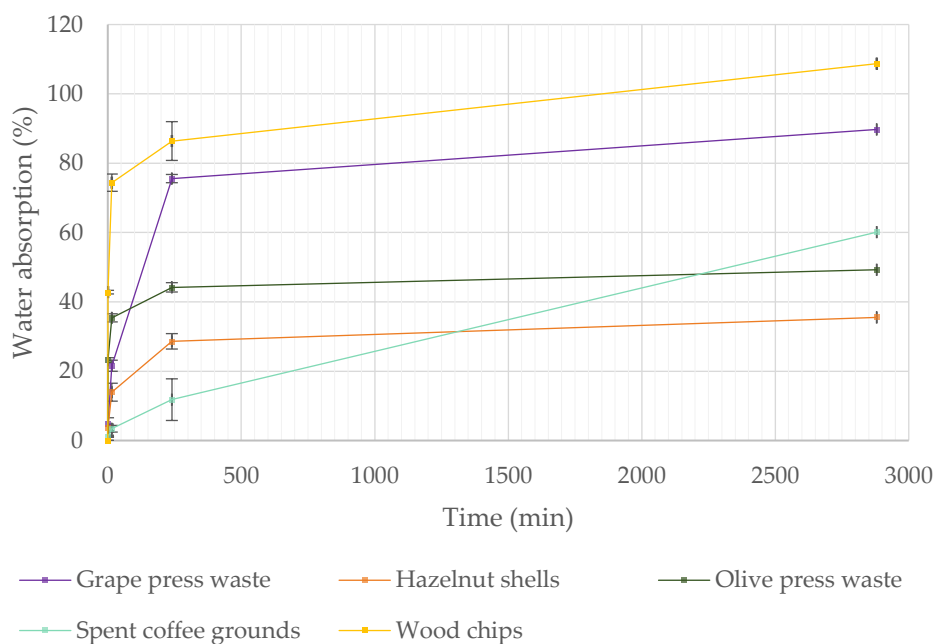


Figure 3.2-6. Water absorption as a function of time, tested according to Amziane et al. [30].

Table 3.2-6. Water absorption of the bio-wastes after 48 h of immersion.

| Bio-Waste            | Absorption after 48 h (%) |
|----------------------|---------------------------|
| Grape press waste    | 89.7 ± 1.2                |
| Hazelnut shells      | 35.5 ± 2.2                |
| Olive press waste    | 49.3 ± 2.1                |
| Spent coffee grounds | 60.1 ± 6.0                |
| Wood chips           | 108.7 ± 5.6               |

Except for the spent coffee grounds, the analysed bio-wastes show a similar behaviour along time (Figure 3.2-6), starting to saturate after approx. 4 h of immersion, although with different levels of water absorption. Comparing with past studies [32,33], the correlation between water absorption and time is similar.

Regarding the absorption after 48 h (2880 min, Table 3.2-6), the different percentages can be observed. The results were compared with some literature ones determined by the same method [30]. Laborel-Préneron et al. [32] reported that water absorption after 48 h for corn cob is 123 % and 380 % for hemp shiv. Page et al. [31] obtained values of about 450 % for hemp shiv. Antunes et al. [33] of about 300 % for rice husk.

Values of water absorption of the analysed bio-waste are lower than in the literature. The discrepancy is probably caused by the considered method that could lead to differences in the results: the used permeable bags could be different, an eventual compaction of the materials during the permeable bag's closing or inside the bags, as well as the compaction of the material during the absorbing phase and the manual rotation speed. To ensure a direct comparison between different

materials, the method should be complemented. For example, the opening mesh of the permeable bag should be defined according to the grain size of the bio-aggregates, or the rotation time could be controlled automatically to avoid manual operation mistakes. Another important factor that could influence the water absorption capacity is the temperature of the water, as demonstrated by Bouasker et al. [71].

Most materials absorb a greater quantity of water during the first 24 h. Hazelnut shells show the lowest water saturation content. They achieved the lowest value of water absorption after 24 h ( $32.9 \pm 2.6$  %), followed by olive press waste ( $48.9 \pm 1.6$  %). Wood chips and grape press waste show the highest values of water absorption after the first 24 h ( $101.9 \pm 1.6$  % and  $(88.0 \pm 0.7)$  %, respectively).

Information about water absorption is extremely useful for future boards' production in case of using a hydraulic matrix, as Laborel-Préneron et al. [32] reported. To better investigate this property, the ratio between the volume of absorbed water and the volume of the materials was calculated and its evolution with time is reported in Figure 3.2-7.

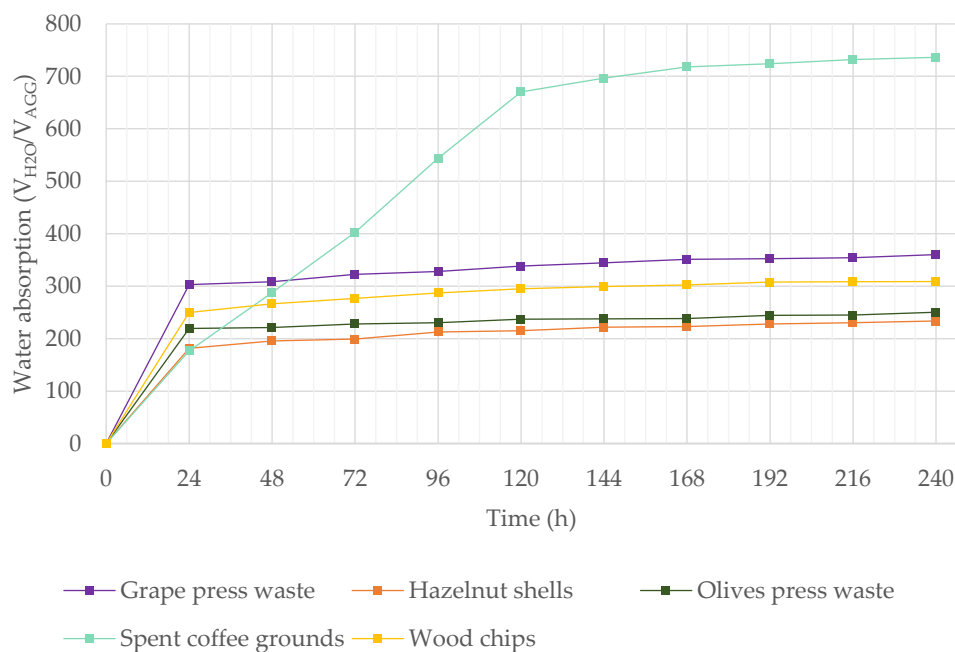


Figure 3.2-7. Variation of water absorption during time considering the ratio of the volume of absorbed water and the volume of the material.

Supposing to produce boards or panels by wet methods and by defining the percentages of the components in volumes, with the same quantities of bio-wastes, composites with hazelnut shells should absorb less water. Composites with grape press waste will probably need more water for production than the others to obtain the same workability because more water will be absorbed by the bio-wastes. Figure 3.2-7 also shows that, differently from the evaluation of water absorption in terms of mass, wood chips show lower water absorption than grapes press waste. Again, the

performance of spent coffee grounds differs greatly from the other bio-wastes. This one shows a slower water absorption: after 120 h it is still absorbing water and the constant mass is reached only after 10 days. The difference between spent coffee grounds and the other bio-wastes may depend on the different grain sizes but other characteristics of the coffee grounds may also have influence.

Figure 3.2-8 shows the variation in moisture adsorbed/desorbed content over time for the considered bio-wastes.

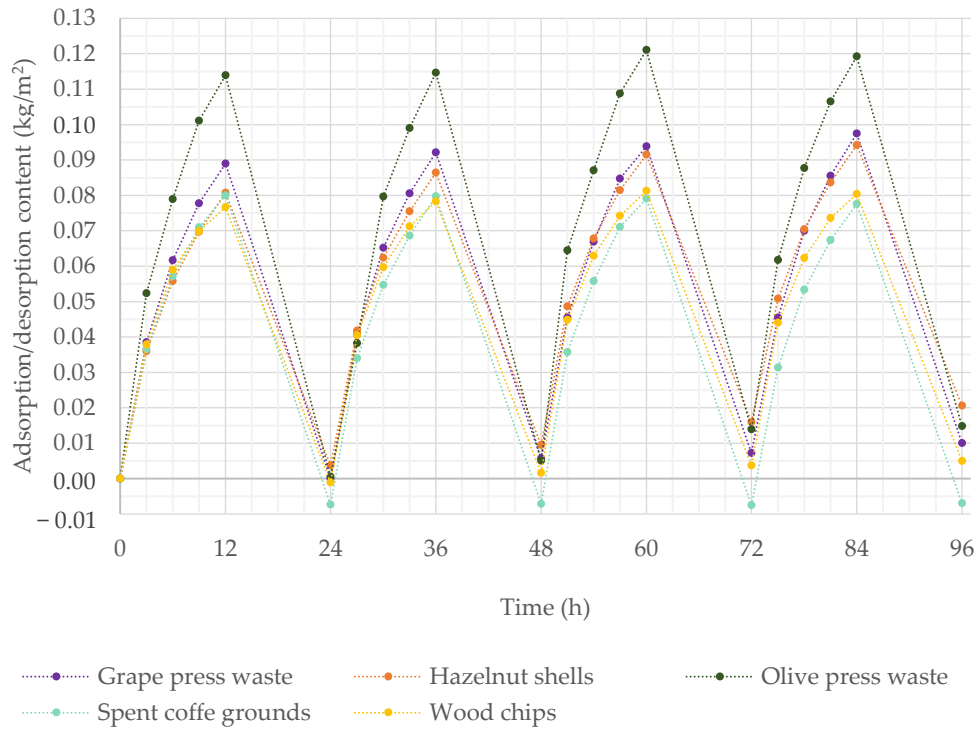


Figure 3.2-8. Moisture adsorption/desorption content for four cycles between 50 % and 75 % RH.

As Collet [56] and Romano et al. [72] reported, hygroscopicity properties widely affect the thermal behaviour of the composites and the control of the dynamic moisture changes. MBV values show the potential of bio-aggregates as hygric regulators [58]. For these reasons, the obtained results could anticipate some of the future properties of the boards.

Olive press waste achieved the highest values of moisture adsorption ( $Q_{A,ac}$  between 0.114 kg/m<sup>2</sup> and 0.116 kg/m<sup>2</sup>) and desorption content ( $Q_{A,ad}$  between 0.104 kg/m<sup>2</sup> and 0.113 kg/m<sup>2</sup>) during the four cycles. Wood chips and hazelnut shells reached the lowest values both in the sorption and desorption phases. Values of moisture sorption are between 0.076 kg/m<sup>2</sup> and 0.080 kg/m<sup>2</sup> for wood chips, between 0.078 kg/m<sup>2</sup> and 0.083 kg/m<sup>2</sup> for hazelnut shells. As for desorption, the values are between 0.074 kg/m<sup>2</sup> and 0.078 kg/m<sup>2</sup> and between 0.074 kg/m<sup>2</sup> and 0.077 kg/m<sup>2</sup>, respectively.

Considering the moisture content difference between sorption and desorption,  $Q_{A,SCr}$  values of grape press waste, olives press waste and wood chips are close to zero during the first cycle (between -0.001 kg/m<sup>2</sup> and 0.001 kg/m<sup>2</sup>). Hence, these aggregates have similar sorption and desorption

capacity. The first cycle shows not already stabilized bio-wastes moisture capacity. Except for wood chips and spent coffee grounds, sorption capacity is greater than desorption. During the second cycle, the bio-wastes sorption capacity improved. For all the bio-wastes, moisture adsorption content,  $Q_{A,ac}$ , is greater than moisture desorption content,  $Q_{A,ad}$ . After the first cycle, also for spent coffee grounds the moisture content difference is near to zero (maximum value of  $0.001 \text{ kg/m}^2$ ). Hazelnut shells showed the greatest moisture content difference for all the cycles (values between  $0.004 \text{ kg/m}^2$  and  $0.006 \text{ kg/m}^2$ ), with higher moisture sorption capacity than desorption.

However, the moisture content differences are very small for all the bio-wastes and during all cycles, less than  $0.01 \text{ kg/m}^2$ . Furthermore, the measurement method (opening and closing of the climatic chamber) might have determined an error that has to be taken into consideration. As Romano et al. [72] reported, the difference between the bio-wastes might depend on a micro-capillary network formation that is created during water molecules sorption/desorption.

Table 3.2-7 reported MBV values [ $\text{g}/(\text{m}^2 \cdot \% \text{ RH})$ ] and the classification provided by Rode et al. [49]: Negligible : MBV =  $0.0\text{--}0.2$ ; Limited: MBV =  $0.2\text{--}0.5$ ; Moderate: MBV =  $0.5\text{--}1.0$ ; Good: MBV =  $1.0\text{--}2.0$ ; Excellent: MBV  $\geq 2.0$ . MBV. Values were calculated to allow a comparison between the bio-wastes and other already studied materials. Indeed, a gap in literature did not allow a comparison to be made between the variation in sorption/desorption content during time of the analysed materials and the ones considered for comparison evaluated by the same test.

Table 3.2-7. MBV of analysed bio-wastes classified according to Rode et al. [49].

| Materials            | MBV<br>( $\text{g}/(\text{m}^2 \cdot \% \text{ RH})$ ) | Classification according to<br>Rode et al. [49] |
|----------------------|--------------------------------------------------------|-------------------------------------------------|
| Grape press waste    | $3.54 \pm 0.09$                                        | Excellent                                       |
| Hazelnut shells      | $3.13 \pm 0.16$                                        | Excellent                                       |
| Olives press waste   | $4.38 \pm 0.13$                                        | Excellent                                       |
| Spent coffee grounds | $3.44 \pm 0.01$                                        | Excellent                                       |
| Wood chips           | $3.10 \pm 0.06$                                        | Excellent                                       |

Olive press waste showed the highest MBV, the wood chips and the hazelnut shells had the lowest ones. In composites' production, olives press waste might guarantee a greater sorption/desorption capacity. They could therefore be the worst aggregates to produce thermal insulation boards, but the best ones in the case of indoor coating panels. Indeed, a high moisture buffering capacity can contribute to the passive control of the internal conditions and indoor air quality. However, these considerations should be verified: the performance of the composites widely varies depending on the complementary components (binders, additives) and the production method. Future studies should be devoted to determining the properties of these bio-wastes when combined with other materials to produce building composites. Overall, even in case of performances worse than the ones of typical composites, the use of bio-wastes replacing raw materials may offer a competitive and



more sustainable solution than other common ones. Furthermore, it would be interesting to investigate the relationship between the bio-aggregates and composites' properties.

According to Ansell et al. [58], a correlation of linear proportionality was expected between loose bulk density and MBV values for natural fibres. They showed that MBV increased with bulk density for hemp shiv, flax shiv, wheat straw and rape shiv. Holcroft and Shea [73] obtained different results considering hemp-lime: the lower the density, the higher the MBV. In the present study, the comparison between the materials does not allow defining a correlation between loose bulk density and MBV. MBV varies depending on other parameters too (e.g., grain size, chemical composition). Cintura et al. [10] reported chemical compositions of the analysed bio-wastes by considering past studies. The researchers accounted it as an important parameter that influences materials' physical and mechanical features. They supposed that the selected bio-wastes may have good moisture buffering capacity, confirmed by the results of the present study.

Considering literature values of some of the materials chosen for comparison, corn cob showed values of 3.24 g/(m<sup>2</sup>.% RH) [58], higher only than hazelnut shells' ones. Hemp shiv had values of about 2.09–2.53 g/(m<sup>2</sup>.% RH) [58], lower than the ones of all analysed bio-wastes. This demonstrates that the selected bio-wastes have good hygroscopicity behaviour. It might be a drawback for insulation boards production, even if, as anticipated, the performances will depend on the final compositions.

#### 3.2.3.4 Bio susceptibility to mould and termites

Figure 3.2-9 gives an idea of the mould growth over the five materials and the control when inoculated with culture media and after four weeks.

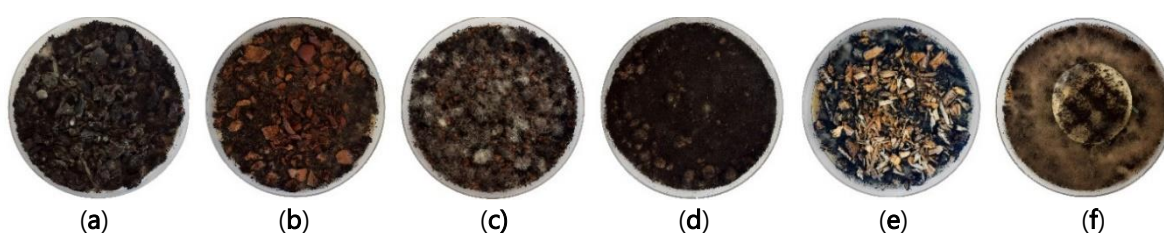


Figure 3.2-9. Mould growth (test with culture media) after four weeks: (a) grape press waste; (b) hazelnut shells; (c) olive press waste; (d) spent coffee grounds (e) wood chips; (f) filter paper control.

All the analysed bio-wastes with culture media are highly susceptible to mould attack, as expected. They reached the maximum value (4-contaminated surface more than 60%) during the second week of testing. The culture media contributes to the biological attack by increasing the sugar content available to the fungi. The controls (Figure 3.2-9 f) demonstrated the validity of the test; all replicates reached grade 4 also on the second week of exposure.

Table 3.2-8 reports the results of weekly mould growth for the samples without culture media and Figure 3.2-10 shows the materials after four weeks of exposure.

Table 3.2-8. Average rate ( $\pm$  standard deviation) of mould growth for the samples without culture media.

| Material             | Mould Development   |                     |                     |                     |
|----------------------|---------------------|---------------------|---------------------|---------------------|
|                      | Week 1              | Week 2              | Week 3              | Week 4              |
| Spent coffee grounds | 2.20 ( $\pm 0.45$ ) | 3.00                | 4.00                | 4.00                |
| Grape press waste    | 3.00                | 4.00                | 4.00                | 4.00                |
| Olive press waste    | 3.00                | 4.00                | 4.00                | 4.00                |
| Hazelnut shells      | 0.40 ( $\pm 0.55$ ) | 2.30 ( $\pm 0.45$ ) | 3.00                | 4.00                |
| Wood chips           | 0.40 ( $\pm 0.55$ ) | 1.40 ( $\pm 0.55$ ) | 1.80 ( $\pm 0.45$ ) | 2.70 ( $\pm 0.67$ ) |

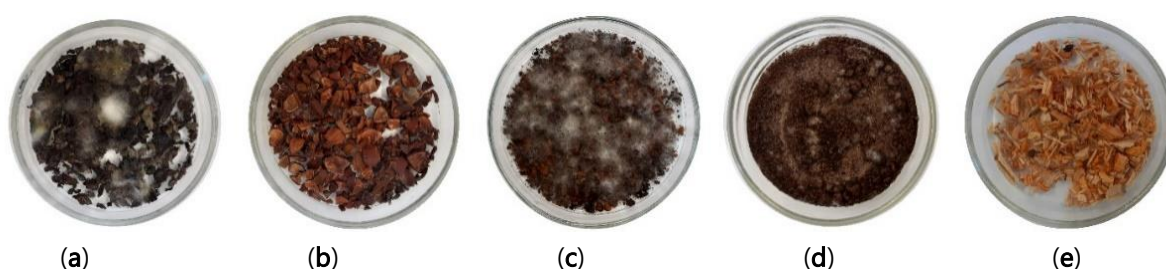


Figure 3.2-10. Mould growth (test without culture media) after four weeks of exposure: (a) grape press waste; (b) hazelnut shells; (c) olive press waste; (d) spent coffee grounds; (e) wood chips.

Even without surrounding the culture media, all the analysed bio-wastes have a high susceptibility to mould. Differently from the previous results, the samples reached the maximum value during the third week of testing, confirming the higher susceptibility caused by culture media (Figure 3.2-9). The biological attack of the controls (Figure 3.2-10 e) demonstrated the validity of the test: being wood chips, mould growth was expected. They were rated as 2.70 (contaminated surface between 30% and 60%) after four weeks, hence all the bio-wastes showed a lower biological resistance.

Hazelnut shells showed similar behaviour to control during the first week, being both rated as 0.4. Nonetheless, this bio-waste reached the maximum values at the end of testing as the others. Grape and olive press waste showed the lowest resistance to mould, being rated at 4 during the second week. Spent coffee grounds reached the maximum value during the third week. As previously anticipated, high hygroscopicity improves mould growth. Considering the results of water adsorption, they could be considered consistent with the bio susceptibility ones: olives and grapes press waste showed the highest moisture buffering capacity, hazelnut shells and wood chips the lowest one. However, considering only the correlation between hygroscopicity and biological attack is an oversimplification. Bio susceptibility depends also on many other factors such as materials' composition, pH, surface properties, roughness, chemical and physical capabilities of mould species [51,74]. Mould growth could be further investigated since these laboratory tests aim at showing an initial

assessment. The results confirm that, as bio-based materials, all the tested bio-wastes have a high susceptibility to mould.

Past research reported that caffeine improves the biological resistance of wood, lowering fungal growth. Kwaśniewska-Sip et al. [75] diluted caffeine into a water solution to treat Scots pine (*Pinus sylvestris* L.) samples; Šimůnková et al. [76] considered Norway spruce (*Picea abies* L. Karst.) samples. In both cases, caffeine solution improved wood's biological resistance. Nevertheless, even if spent coffee grounds could be a source of chemicals that can moderate mould growth, they cannot be used to avoid the biological attack, as Barbero-López et al. [77] reported. Indeed, the raw material is known as a good material to allow mould growth [78,79], as confirmed also in the present study.

Table 3.2-9 reports the results related to the susceptibility to subterranean termites, considering the average value for each bio-waste. The survival rate of the maritime pine (higher than 50 %) confirms the validity of the test. Figure 3.2-11 shows the samples after the four weeks.

Table 3.2-9. Average rate ( $\pm$  standard deviation) of survival of *R. grassei* colonies for the bio-wastes.

| Material             | Survival Rate (%)     | Mould Development * |
|----------------------|-----------------------|---------------------|
| Spent coffee grounds | 0.00                  | Extensive           |
| Grape press waste    | 63.33 ( $\pm 27.30$ ) | Limited             |
| Olives press waste   | 0.00                  | Extensive           |
| Hazelnut shells      | 10.67 ( $\pm 9.45$ )  | Limited             |
| Wood chips           | 81.33 ( $\pm 4.62$ )  | Limited             |

\* Visually and qualitatively evaluated after four weeks.

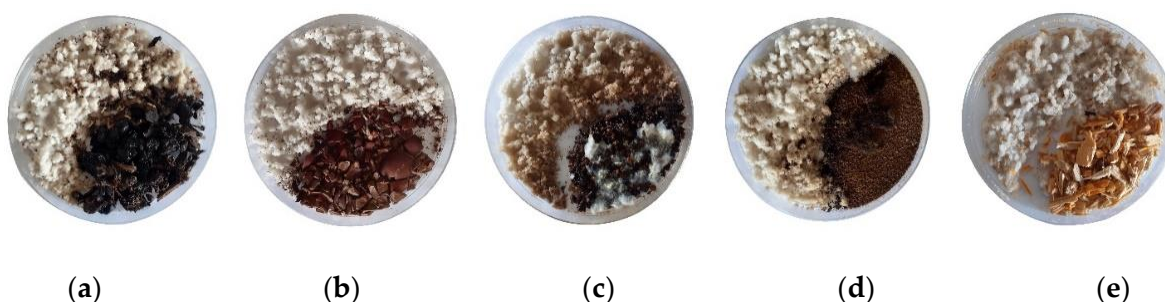


Figure 3.2-11. Termites' survival and mould growth after four weeks exposure: (a) grape press waste; (b) hazelnut shells; (c) olive press waste; (d) spent coffee grounds; (e) wood chips.

The termites were able to consume all bio-wastes, though their ability to survive was highly influenced by both the available cellulose they could use and the fast development of moulds that they were not able to limit. The bio-waste with the highest amount of cellulose (grape press waste with small parts of grapevine) [10] was not surprisingly the most consumed by termites and the one that kept the highest survival rate of the termites during the test period. Hazelnut shells have better resistance to termites than wood chips, but they are not completely unaffected: 10.67 % of the termites

survived deteriorating a part of the material. Cintura et al. [10] reported that hazelnut shells have a cellulose content of about 22.90–34.60%. According to past studies, wood chips have a cellulose content of about 32.09–50.00% [80–82]. The results are in line with this information: termites probably fed on hazelnut shells, but the cellulose content, i.e., the necessary nutrients to survive, was not enough, hence they slowly died.

Termites' survival for spent coffee grounds and olives press waste was rated at 0%, but these results cannot be considered as a demonstration of their best resistance to termites. Indeed, these materials also showed a high mould growth (Table 3.2-9 and Figure 3.2-11 a,d) that could be the cause of termites' mortality. Šimůnková et al. [76] investigated caffeine's resistance from termites, applying a caffeine and water solution on wood and exposing the samples to *Reticulitermes flavipes* (Kollar). The results of their study demonstrated that caffeine improves the biological resistance of wood. Nevertheless, as previously anticipated, this cannot demonstrate good resistance to termites of spent coffee grounds. As Cintura et al. [10] reported, collecting information from several past studies, cellulose content in this bio-waste is between 8.60–52.42%. The range of variation is high, but spent coffee grounds have the lowest values when compared with the other considered bio-wastes. This information and the results of the present work cannot confirm a high termite resistance with spent coffee grounds, also due to the mould formation during the four weeks. Maybe this bio-waste could be used as a source of caffeine to apply on future eco-efficient boards/panels.

### 3.2.3.5 Comparison between bio-wastes and wood chips

As previously anticipated, maritime pine chips were considered as control material. Table 3.2-10 reports a comparison between the properties of the bio-wastes and wood chips, considering their use as thermal insulation materials. It shows which properties could be considered better (in green) or worse (in orange) than maritime pine ones for thermal insulation boards. The maximum/minimum values are highlighted with stronger shades of green/orange. When they are not highlighted, there is no difference between the considered bio-waste and wood chips. When they are highlighted in grey, more considerations have to be made. It is important to remind that this work presents some screening tests and properties of bio-aggregates. The performance of the final composites will widely depend on their composition and production method.

The analysed bio-wastes have higher values of loose bulk density and thermal conductivity than maritime pine chips. This may result in composites with worse thermal insulating performances than wood-based ones. A higher loose bulk density of the aggregates could be a drawback for thermal insulation boards' production, a benefit in case of coating panels'. The values of absorption capacity after 48 h are lower for the selected bio-wastes; hence, they may secure a higher water resistance when used in composites. For MBV, the values are higher than wood chips' ones, except for hazelnut shells.

Table 3.2-10. Comparison between values of considered bio-wastes and maritime pine chips for thermal insulation boards.

| Properties                                         | Materials  |                   |                 |                    |                      |
|----------------------------------------------------|------------|-------------------|-----------------|--------------------|----------------------|
|                                                    | Wood Chips | Grape Press Waste | Hazelnut Shells | Olives Press Waste | Spent Coffee Grounds |
| Loose bulk density (kg/m <sup>3</sup> )            | 245.14     | 343.88            | 550.5           | 449.43             | 478.81               |
| Thermal conductivity T = 23 °C, RH = 50% (W/(m.K)) | 0.077      | 0.078             | 0.107           | 0.089              | 0.092                |
| Thermal conductivity T = 23 °C, RH = 75% (W/(m.K)) | 0.082      | 0.081             | 0.115           | 0.097              | 0.099                |
| Absorption capacity after 48 h (%)                 | 108.7      | 89.7              | 35.5            | 49.3               | 60.1                 |
| MBV (g/(m <sup>2</sup> .% RH)) *                   | 3.02       | 3.5               | 2.94            | 4.18               | 3.38                 |
| Bio-susceptibility to mould with culture media     | 4          | 4                 | 4               | 4                  | 4                    |
| Bio-susceptibility to mould without culture media  | 2.7        | 4                 | 4               | 4                  | 4                    |
| Bio-susceptibility to termites—Survival rate (%)   | 81.33      | 63.33             | 10.67           | n.a. *             | n.a. *               |

\* Note: MBV—see detailed evaluation below; n.a. not available—the results need confirmation.

It seems that composites produced with the bio-wastes could have a higher contribution as moisture regulators than wood-based ones, guaranteeing better passive equilibrium of indoor humidity levels. This could be a benefit in case of internal coating panels' production, and a drawback in case of external panels or thermal insulation boards. A high MBV could affect the durability of the building composites. In terms of bio-susceptibility, the analysed bio-wastes show low resistance to mould, as well as wood chips. For termites, the outcomes of spent coffee grounds and olive press waste cannot be considered as a demonstration of high resistance: they could result from mould growth. Hazelnut shells show better resistance to termites' attacks than wood chips, but they are not completely unaffected. For eco-efficient building products, this drawback should be further investigated and solved.

The comparison between bio-wastes allows concluding that grape press waste is the most similar to maritime pine chips. It may be a good replacement of wood particles in future boards' production, even if many other factors play an important role (e.g., binders, production phase, treatments).

The final performance of panels and boards produced with the studied bio-wastes will strongly depend on their composition. The selected binders could both moderate or increase the benefits or the drawbacks of using these bio-wastes as aggregates. For example, for a hydraulic matrix, the use of water could cause the degradation of the bio-wastes and increase mould growth. Differently, a binder with a pH higher than 10 could guarantee less vulnerability to biological attack [51]. The

thermal insulation properties could be improved by using a binder that secures the bonding of the aggregates by leaving a porous network between them.

Furthermore, the use of additives and the mixture with other material may widely affect the properties of the final product. For example, citric acid can moderate biological susceptibility and water absorption [83–85]. Past studies reported that boric acid, lime and potassium benzoate moderate mould growth [37,86,87]. On the other hand, they might increase the values of bulk density and thermal conductivity.

Pre-treatments of the bio-wastes may influence their properties, as Antunes et al. [33] demonstrated. As for the production methods, the curing phases [88] may be important, depending on the binder matrix. A fast and ventilated drying environment for the curing of the final product could avoid mould growth. Finally, the production process may widely affect the final properties. For example, pressing the composites increases bulk density and mechanical resistance, while just moulding could guarantee boards with lower bulk density and better thermal insulation properties.

Future studies could further investigate how all these parameters can influence eco-efficient boards and panels' performance.

### 3.2.4 Conclusions

This study investigated the properties of some bio-wastes (grape and olive press waste, hazelnut shells and spent coffee grounds), available in Euro-Mediterranean Countries, to evaluate the feasibility of using them as aggregates for composite boards and panels. Maritime pine chips were considered as control, simulating recycled timber. Particle size distribution, loose bulk density, thermal conductivity, water absorption and hygroscopic properties were assessed, but particularly bio susceptibility to mould and termites was also analysed. From the results obtained, the following conclusions can be drawn:

- Grain size analysis demonstrated that shredding these bio-wastes to produce insulation boards does not seem necessary. Spent coffee grounds differ from the other bio-wastes and, for composites' production, they might be used as fine aggregates.
- The selected bio-wastes do not show by themselves good thermal insulation characteristics. Combining them with other materials, pre-treatments or production processes could improve the thermal insulation performance of the final products.
- The analysed bio-wastes have good hygroscopic behaviour, with high MBV, an advantage in case of the production of indoor coating boards to secure a passive control of indoor relative humidity and comfort.
- For all bio-wastes, the results confirmed the high biological susceptibility to the tested organisms. This drawback must be further investigated and mitigated for composites' production, namely by combining the bio-wastes with materials resistant to biological attack or by specific pre-treatments.

- Using the recommendations of RILEM TC 236-BBM “Bio-aggregate-based building materials” to evaluate properties of bio-wastes, considered as raw materials, allows them to be analysed and guarantees a simple comparison between different ones. However, some additional details for testing should be included, as well as complementary insect and fungi bio-susceptibility tests.

The present study both describes laboratory tests to analyse bio-wastes properties and provides a preliminary assessment of the considered ones, filling gaps in the literature. Using the selected bio-wastes as aggregates may be a competitive and more sustainable solution than the use of the traditional aggregates. Future studies will deepen the feasibility of producing eco-efficient composites and evaluate the properties of boards or panels.

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### 3.3 Article D - Eco-efficient boards with agro-industrial wastes – Assessment of different adhesives

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#### Abstract

Laboratory tests were carried out to evaluate the feasibility of producing boards or coating panels with good hygrothermal performance by using different agro-industrial wastes (grape and olive press wastes, hazelnut shells, spent coffee and pine) and adhesives (gypsum-based binder, sodium silicate solution and starch-based glue). Hygrothermal properties and resistance to mould growth, including the effect of citric acid, were evaluated. The results highlighted the benefits and drawbacks of each adhesive: the binder seems more adequate to produce bio-based plastering mortars than boards; the other adhesives seem efficient solutions to produce boards with good hygrothermal properties; for the natural glue the bio-susceptibility needs improvement.

**Keywords:** Air lime; Biological susceptibility; Bio-waste; Citric acid; Hemihydrate gypsum; Hygroscopicity; Natural glue; Sodium Silicate; Starch; Thermal property.

#### 3.3.1 Introduction

The construction sector is one of the most polluting ones, as it produces harmful residues and requires a large amount of global energy and resources, as it has been stated by several researchers [1–4]. It has been identified as responsible for the highest emissions and as the largest energy-consumer activity in Europe [5,6]. Both the management of buildings, the construction process, and the operational phase during the lifetime of buildings are widely responsible for the environmental impact [7], hence building materials too. For these reasons, research is currently focusing on the possibility of using eco-efficient materials and environmentally friendly solutions [8]. Among them, the use of agro-industrial wastes for construction practices has been widely investigated. They are continuously produced locally in large volumes, they can be used for composites having similar properties to conventional ones, and they are biodegradable and non-toxic [9,10]. Furthermore, the use of local agro-industrial wastes to produce building materials reduces carbon emissions and secures the disposal of solid waste, another cause of environmental problems [11–13].

Panels and particleboards for building practices are widely used due to their easy on-site installation, their multiple possibilities of applications and their economic advantages. They can be produced by using several techniques, such as compression, thermal treatment, use of an adhesive,

and materials, such as wood chips, fibres and agro-industrial wastes, an efficient solution for waste disposal [14–17]. Many past studies focused on the evaluation of thermal insulation properties of agro-industrial wastes-based boards and panels. For example, Mahieu et al. [18] analysed binderless compacted particleboards made up of flax shives (*Linum usitatissimum* L.) and sunflower stems (*Helianthus annuus* L.). For a density of 500 kg/m<sup>3</sup>, they obtained values of thermal conductivity of 0.077 W/(m.K) for both; for a density of 350 kg/m<sup>3</sup>, they achieved values of 0.066 W/(m.K) for flax shives and 0.070 W/(m.K) for sunflower stems (calculated under steady-state conditions). Pavelek and Adamová [19] evaluated the thermal insulation properties of a sandwich panel: a medium density fibre-board filled with a bio-based insulation core. For this latter, they compared short-cut rapeseed and short-cut wood chips. They obtained thermal conductivity values of 0.048 W/(m.K) for rapeseed and 0.065 W/(m.K) for wood chips (calculated under steady-state conditions). Ramos et al. [20] studied the properties of particleboards made up of corn cob and polyvinyl acetate (PVA) and Fabricol AG222 (FAG222) as adhesives. They obtained thermal conductivity values of 0.052 W/(m.K) for PVA and 0.087 W/(m.K) for FAG222 (calculated by the heat flux meter system). Considering that 0.065 W/(m.K) is considered the maximum value to classify materials as thermal insulations [21,22], and past research considered a value of 0.1 W/(m.K) [2,23,24], the previous studies demonstrated the feasibility of using agro-industrial wastes to produce thermal insulation boards that meet the standards' requirements. Table 3.3-1 reports the density and the thermal conductivity of commercial panels with different functionality (not only thermal insulation panels) and the values reported in past studies that analysed the thermal performance of bio-based panels and boards.

Table 3.3-1. Density and thermal conductivity of different types of panels and boards (considering commercial products and literature values).

| Board/Panel                                                                                                                                    | Density<br>[kg/m <sup>3</sup> ] | Thermal conductivity<br>[W/(m.K)] | Reference |
|------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------|-----------------------------------|-----------|
| <b>Commercial products</b>                                                                                                                     |                                 |                                   |           |
| Celenit N panel (thermal and acoustic insulation board)                                                                                        | 346-533                         | 0.065                             | [25]      |
| Celenit ABE panel (thermal and acoustic insulation board)                                                                                      | -                               | 0.075                             | [26]      |
| KlimaExpert5 (thermal insulator layer: wood fibre)                                                                                             | 280*                            | 0.040-0.060                       | [27]      |
| Knauf Organic (sound-absorbing panel made up of coniferous wood wool mineralized, cement mortar and lime)                                      | -                               | 0.08                              | [28]      |
| Knauf Standard A                                                                                                                               | 630**                           | 0.21                              | [29]      |
| VestaEco Internal (internal thermal insulation panel made with lignocellulosic fibres and polymeric methylenediphenyl isocyanate, PMDI, resin) | 180                             | 0.048                             | [30]      |
| OSB panel according to the European Panel Federation                                                                                           | 650                             | 0.13                              | [31]      |
| <b>Past studies (literature review)</b>                                                                                                        |                                 |                                   |           |
| Binderless compacted particleboards made up of flax shives ( <i>Linum usitatissimum</i> L.)                                                    | 350-500                         | 0.066-0.077                       | [18]      |



|                                                                                                                                         |               |               |      |
|-----------------------------------------------------------------------------------------------------------------------------------------|---------------|---------------|------|
| Binderless compacted particleboards made up of sunflower stems ( <i>Helianthus annuus</i> L.)                                           | 350-500       | 0.070-0.077   | [18] |
| Internal layer of a sandwich panel (medium density fibre-board filled with a bio-based insulation core) made up of short-cut rapeseed   | -             | 0.048         | [19] |
| Internal layer of a sandwich panel (medium density fibre-board filled with a bio-based insulation core) made up of short-cut wood chips | -             | 0.065         | [19] |
| Particleboards made up of corn cob and polyvinyl acetate (PVA) as adhesives                                                             | -             | 0.052         | [20] |
| Particleboards made up of corn cob Fabricol AG222 (FAG222) as adhesives                                                                 | -             | 0.087         | [20] |
| Expanded Polystyrene (EPS)                                                                                                              | -             | 0.045         | [20] |
| Extruded Polystyrene (XPS)                                                                                                              | -             | 0.037         | [20] |
| Panels casted with a mix of rice husk, earth, air lime, hemi-hydrated gypsum, and water.                                                | 650.8-1021.6  | 0.102-0.197   | [32] |
| Boards made up of cement, gypsum, NaOH, aluminium dust, water, and corn stalk                                                           | -             | 0.100-0.200   | [33] |
| Panels of wood waste and Portland cement, lime, and water                                                                               | 875.62-984.31 | 0.0742-0.1078 | [34] |
| Insulation corkboard (ICB)                                                                                                              | 115           | 0.045         | [35] |
| Insulation corkboard (ICB)                                                                                                              | -             | 0.042         | [36] |
| Composites of hemp shiv and clay                                                                                                        | 361.3-537.2   | -             | [37] |
| Composites made up of straw fibres (SF) bounded by using a sodium silicate solution ( $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ ).    | 152           | 0.058         | [38] |
| Composites made up of olives fibres (OF) bounded by using a sodium silicate solution ( $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ ).   | 235           | 0.062         | [38] |
| Gypsum plasterboard                                                                                                                     | 900           | 0.25          | [39] |

Note: \*Maximum value; \*\*Minimum value.

In addition to bio-aggregates, the selected binders, and other adhesives play an important role in the insulation boards' production. They can widely influence the final performance, as well as the feasibility of producing composites and their eco-friendly characteristics [13,40,41]. The choice of binders and other adhesives is as important as the bio-aggregates' one. Combining eco-efficient aggregates with binders and adhesives that cause high environmental impact or that are unhealthy and not sustainable solutions. For example, urea-formaldehyde resins are widely used due to their many advantages, but their toxicity is already known and created much concern [16,40,42]. Cardoso et al. [43] collected information about bio-based adhesives for wood-based panels and reported tannins, lignin, proteins, carbohydrates, and vegetal oils as natural possibilities. Solt et al. [41] presented a detailed review of formaldehyde-free adhesive systems for particulate wood, considering the possible future restrictions for formaldehyde-based ones. They assessed both synthetic and renewable-based adhesives. Kariuki et al. [44] reported several past studies that considered the use of starch and starch's derivatives as an alternative to formaldehyde-based resins. Starch has been already used for board production. For example, Muizniece and Blumberga [45] used potato starch binder and dried needles of spruce and pine to produce thermal insulation particleboards. Nataraj

et al. [46] referred to wheat gluten as a promising natural glue. Monteiro et al. [47] investigated sour cassava starch for an adhesive system for wood-based particleboards. Starch could be a feasible solution to produce environmentally friendly binders, also because of its supply utilization and production quantity. They are reported in Table 3.3-2, considering only Euro-Mediterranean Countries, the selected area for the present work [48].

Table 3.3-2. Information about starch production quantity (during 2020) and supply utilization accounts (during 2019) in Euro-Mediterranean Countries according to FAO [49,50].

|          | Production quantity<br>[Tonnes x 10 <sup>7</sup> ] | First producer | Starch supply utilization<br>account [Tonnes x 10 <sup>6</sup> ] |
|----------|----------------------------------------------------|----------------|------------------------------------------------------------------|
| Maize    | 3.06                                               | France         | 2.43                                                             |
| Potatoes | 1.39                                               | France         | 0.09                                                             |
| Rice     | 0.27                                               | Italy          | 0.03                                                             |
| Wheat    | 4.76                                               | France         | 1.31                                                             |

Another important point for the production of agro-industrial waste-based composites is their bio-susceptibility. It can affect and modify materials' properties and durability [51,52], as well as, depending on the application, can compromise building inhabitants' health. Biological susceptibility depends on several factors: chemical compositions (i.e. cellulose, hemicellulose, lignin content), temperature, relative humidity (RH), moisture content, the surface of the materials, and the pH [17,53–55]. Both the aggregates and the adhesives play an important role in the biological resistance of the final product [56]. For example, a bio-based binder without any biocide could be easily attacked by microorganisms. Hence, using additives, adhesive, or binder with high biological resistance (e.g., boric acid [51], sodium benzoate [57], lime [58,59] and citric acid [56,60]) could be certainly more suitable for agro-industrial-based composites.

The present work shows the preliminary studies carried out to produce eco-efficient boards by using agro-industrial wastes as aggregates. It focuses on the evaluation of different adhesives. The selected aggregates are grape and olive press wastes, hazelnut shells and spent coffee grounds. Maritime pine (*Pinus pinaster* Ait.) chips were used as control material, according to previous studies [48,61]. Three types of adhesives are evaluated: one based on hemi-hydrated gypsum and air lime, a sodium silicate solution and one natural glue made of starch. They were selected considering the following criteria:

- Avoid the use of harmful products, such as formaldehyde-based adhesives which can be sources of unhealthy substances [62];
- Guarantee low energy consumption in samples' production by using materials with low embodied energy, avoiding high pressure, not shredding the bio-aggregates, and drying and curing at low temperature;

- Produce samples by using the same binders for the different considered aggregates, hence considering compatibility aggregates-binders for all;
- Prevent bio-aggregates degradation by avoiding production processes that require high-temperature and moderate biological attacks by using binder, adhesive or additives that can moderate the biological susceptibility.

The aim is to study the feasibility of producing eco-efficient insulation thermal boards or coating panels with insulation thermal properties by using the selected materials and to evaluate their hygrothermal properties and bio-susceptibility.

### 3.3.2 Materials and methods

#### 3.3.2.1 Materials

##### 3.3.2.1.1 Bio-aggregates

Grape press waste (G), hazelnut shells (H), olive press waste (O) and spent coffee grounds (c) were selected as bio-aggregates [48], as they are very common in Euro-Mediterranean Countries. Maritime pine (*Pinus pinaster* Ait.) chips (W) were used as control material. The aggregate wastes G, H and O were collected from industries and spent coffee grounds from a coffee shop and prepared as reported by Cintura et al. [61]. G and O were air-dried for 7 and 10 weeks, respectively; H was used as received. Their physical properties and bio-susceptibility as raw materials were evaluated considering the possible development of composites. Table 3.3-3 reports information about loose bulk density, and thermal conductivity; Figure 3.3-1 shows the particle size distribution of the bio-aggregates, considering past research [61].

Table 3.3-3. Loose bulk density and thermal conductivity of the considered bio-wastes adapted from Cintura et al. [61].

| Material                 | Loose bulk density<br>(T = 20 °C, RH = 60%)<br>[kg/m <sup>3</sup> ] | Thermal Conductivity<br>(T = 23 °C, RH = 50%)<br>[W/(m.K)] |
|--------------------------|---------------------------------------------------------------------|------------------------------------------------------------|
| Grape press waste – G    | 343.88 ± 12.00                                                      | 0.078 ± 0.002                                              |
| Hazelnut shells – H      | 550.50 ± 19.53                                                      | 0.107 ± 0.003                                              |
| Olive press waste – O    | 449.43 ± 8.60                                                       | 0.089 ± 0.004                                              |
| Spent coffee grounds – c | 478.81 ± 5.40                                                       | 0.092 ± 0.002                                              |
| Wood chips – W           | 256.98 ± 8.10                                                       | 0.077 ± 0.001                                              |

##### 3.3.2.1.2 Adhesives

One adhesive (composites A) was made up of hemi-hydrated gypsum and air lime. Hemi-hydrated gypsum, produced at low temperatures (120-180 °C), was provided by Sival Company, Portugal. Hydrated air lime CL80-S, according to EN 459-1 [63], was provided by Calcidrata S.A., Portugal. It was applied with the aim of both delaying the hardening process of gypsum and improving biological resistance [32,58,59]. After several mix designs, since resistance to mould was low, citric acid,

produced by La Droguerie Éco, France, was added with the aim of moderating biological susceptibility [56,64,65].

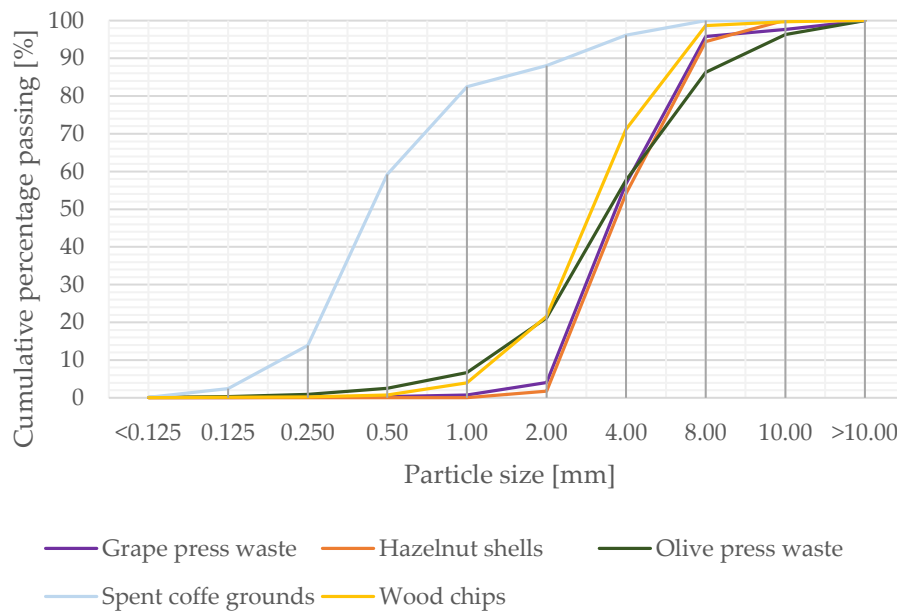


Figure 3.3-1. Particle size distribution of the tested bio-wastes and the wood chips adapted from Cintura et al. [61].

The second studied adhesive (used for composites B) was a sodium silicate solution ( $\text{Na}_2\text{O} \cdot n \text{SiO}_2 \cdot n\text{H}_2\text{O}$ ) produced by Labkem, Spain, and analysed by Ingessil S.r.l., Verona, Italy. It was selected considering the study of Liuzzi et al. [38] that showed promising insulation properties for composites made up of sodium silicate solution mixed with bio-aggregates. The sodium silicate solution allowed avoiding pressing the samples and curing them at environmental temperatures. Furthermore, sodium silicate is resistant to fire and may contribute to avoid chemical decomposition and mould growth [38,66]; hence it could moderate some of the drawbacks of using bio-waste as building materials [48].

A natural glue made up of maize starch was selected as the third adhesive (composites C), considering the high production of maize in Euro-Mediterranean Countries during the last years (Table 3.3-2). The starch-based glue was prepared by mixing starch, boiled water and vinegar which has an antimicrobial effect on fruit and vegetables [67]. With the aim to improve mechanical properties, resistance to water and biological attack, citric acid was added [46].

Table 3.3-4 shows the information on the selected materials. The percentages of citric acid were maintained low to avoid a high delay in the setting time, to secure good workability and to moderate the inhibitory effect on the setting process of gypsum [68–70].

Table 3.3-4. Information on the main materials used for the three adhesives.

| Material                                | Physical and chemical properties/compositions     |           | References                             |
|-----------------------------------------|---------------------------------------------------|-----------|----------------------------------------|
| Hemi-hydrated gypsum (composites A)     | Water/gypsum ratio [kg/l]                         | 1450      | Sival technical sheet [71]             |
|                                         | Hardening time [min]                              | 12 ± 3    |                                        |
|                                         | Setting time [min]                                | 28 ± 5    |                                        |
|                                         | Linear expansion (1h) [%]                         | 0.2       |                                        |
|                                         | Bending strength (dry) [kg/cm <sup>2</sup> ]      | ≈ 52      |                                        |
| Sodium silicate solution (composites B) | Mass fraction SiO <sub>2</sub> /Na <sub>2</sub> O | 2.4-2.6   | Labkem technical sheet [72]            |
|                                         | SiO <sub>2</sub> +Na <sub>2</sub> O [%]           | ≥ 39      |                                        |
|                                         | Density at 20 °C [g/cm <sup>3</sup> ]             | 1.45-1.48 |                                        |
|                                         | Fe <sub>2</sub> O <sub>3</sub> [%]                | ≤ 0.01    |                                        |
|                                         | CaO [%]                                           | ≤ 0.1     |                                        |
|                                         | Viscosity (20 °C) [cP]                            | ≥ 100     | Information provided by In-gessil [73] |
|                                         | Molar ratio [-]                                   | 2.48      |                                        |
|                                         | Na <sub>2</sub> SiO <sub>3</sub> title [% p/p]    | 41.33     |                                        |
|                                         | SiO <sub>2</sub> [% p/p]                          | 29.17     |                                        |
|                                         | Na <sub>2</sub> O [% p/p]                         | 12.16     |                                        |
|                                         | pH at T = 20 °C [-]                               | 12.40     |                                        |
| Maize starch-based glue (composites C)  | Water/starch ratio [ml/g]                         | 5         | -                                      |
|                                         | Vinegar/starch ratio [ml/g]                       | 0.28      |                                        |
|                                         | Citric Acid/starch ratio [g/g]                    | 0.04      |                                        |

### 3.3.2.2 Composites and samples production

The mix designs were selected both by considering past studies and by practical tests. The choosing criteria were guaranteeing the workability and feasibility of producing samples with all the different bio-aggregates by using the maximum amount of bio-wastes and the minimum amount of adhesives. Table 3.3-5 shows the selected formulations in volume and the mix percentages by volume. Furthermore, the composites' designations were reported: the first letter identified the adhesive (A – gypsum-base binder, B – sodium silicate solution, C – natural glue); the following letter identified the aggregates (G – grape press waste, H – hazelnut shells, O – olive press waste, W – wood chips, c – spent coffee grounds). Hence, spent coffee grounds, a waste with particle size smaller than the others, was employed as fine aggregate and combined with wood chips for some composites.

The different aggregates and the adhesive were mechanically mixed for 3 minutes, until homogeneity. The mixes were placed in cylindrical PVC moulds (diameter = 8.5 cm, high = 4 cm) without compaction. Three samples were produced for each composite. Samples A were dried and cured at environmental conditions, guaranteeing ventilation. The curing time was 28 days [74]. Samples B were cured for 28 days at environmental conditions and then put at T = 50 °C until the variation in mass after 24 h was less than 0.5 % to secure a complete drying [38,75]. Samples C were placed for 3 days at T = 50 °C to secure a fast drying, de-moulded and then placed at T = 60 °C until the

variation in mass was lower than 0.5 %. Figures 3.3-2, 3.3-3 and 3.3-4 show samples A, B and C, respectively, after casting and curing, reporting their designations.

Table 3.3-5. Formulation of composites A based on gypsum, B with sodium silicate and C with natural glue with the different bio-aggregates.

| Composites   | Designations   | Material             | Quantities [ml] | Mix percentages by volume* [%] |
|--------------|----------------|----------------------|-----------------|--------------------------------|
| Composites A | AG, AH, AO, AW | Aggregate            | 200             | 45.8                           |
|              |                | Hemi-hydrated gypsum | 100             | 22.9                           |
|              |                | Air Lime             | 35              | 8.0                            |
|              |                | Citric Acid          | 2               | 0.5                            |
|              |                | Water                | 100             | 22.9                           |
|              |                |                      |                 |                                |
|              | AWc            | Aggregate            | 200             | 37.2                           |
|              |                | Spent coffee grounds | 100             | 18.6                           |
|              |                | Hemi-hydrated gypsum | 100             | 18.6                           |
|              |                | Air Lime             | 35              | 6.5                            |
|              |                | Citric Acid          | 2               | 0.4                            |
|              |                | Water                | 100             | 18.6                           |
| Composites B | BG, BH, BO, BW | Aggregate            | 250             | 75.8                           |
|              |                | Sodium silicate      | 80              | 24.2                           |
|              | BWc            | Aggregate            | 200             | 50.0                           |
|              |                | Spent coffee grounds | 100             | 25.0                           |
|              |                | Sodium silicate      | 100             | 25.0                           |
| Composites C | CG, CH, CO, CW | Aggregate            | 200             | 55.6                           |
|              |                | Natural glue         | 160             | 44.4                           |
|              | CWc            | Aggregate            | 200             | 36.4                           |
|              |                | Spent coffee grounds | 100             | 18.2                           |
|              |                | Natural glue         | 250             | 45.5                           |

\* Calculated considering the total volume.

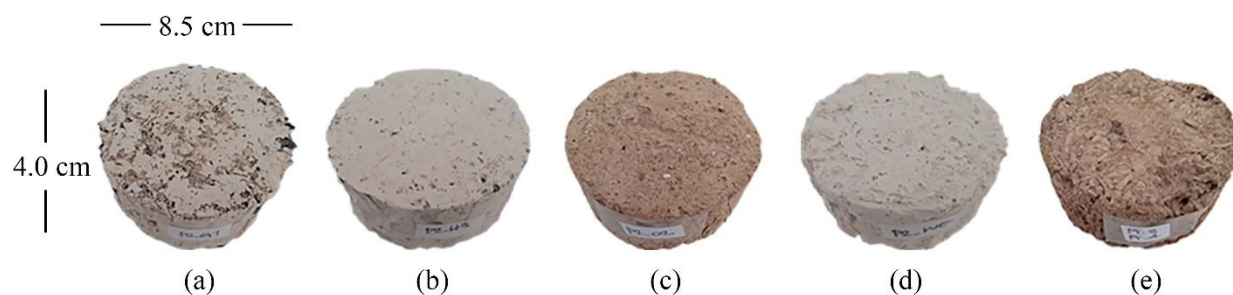


Figure 3.3-2. Samples A – hemi-hydrated gypsum, air lime and citric acid: (a) grape press waste (AG); (b) hazelnut shells (AH); (c) olive press waste (AO); (d) wood chips (AW); (e) wood chips and spent coffee grounds (AWc).



Figure 3.3-3. Samples B – sodium silicate solution: (a) grape press waste (BG); (b) hazelnut shells (BH); (c) olive press waste (BO); (d) wood chips (BW); (e) wood chips and spent coffee grounds (BWc).

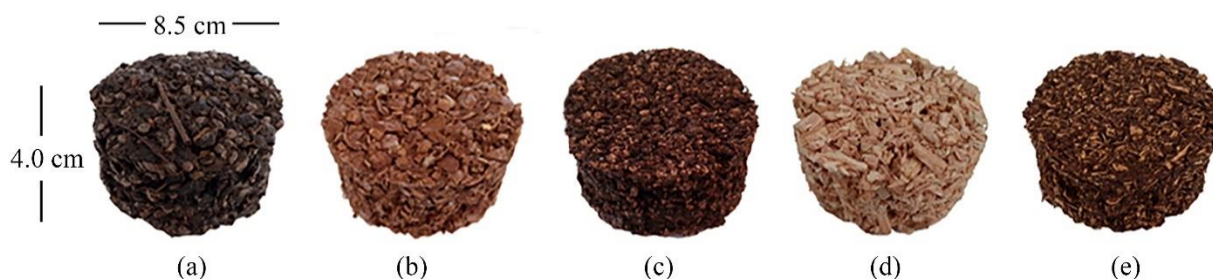


Figure 3.3-4. Samples C – starch-based natural glue: a) grape press waste (CG); b) hazelnut shells (CH); c) olive press waste (CO); d) wood chips (CW); e) wood chips and spent coffee grounds (CWc).

### 3.3.2.3 Methods

#### 3.3.2.3.1 Visual observation and pH

The samples were constantly monitored during the casting and curing process, evaluating the presence of cracks, the distribution of the adhesive on the surface, and the eventual mould growth. At the end of the curing time, the pH of the composites was evaluated by using Litmus paper. A drop of demineralized water was applied on the samples' surface by using a pipette and the Litmus papers were placed on it. The pH evaluation was obtained considering the final colour of the Litmus paper (Figure 3.3-5a).

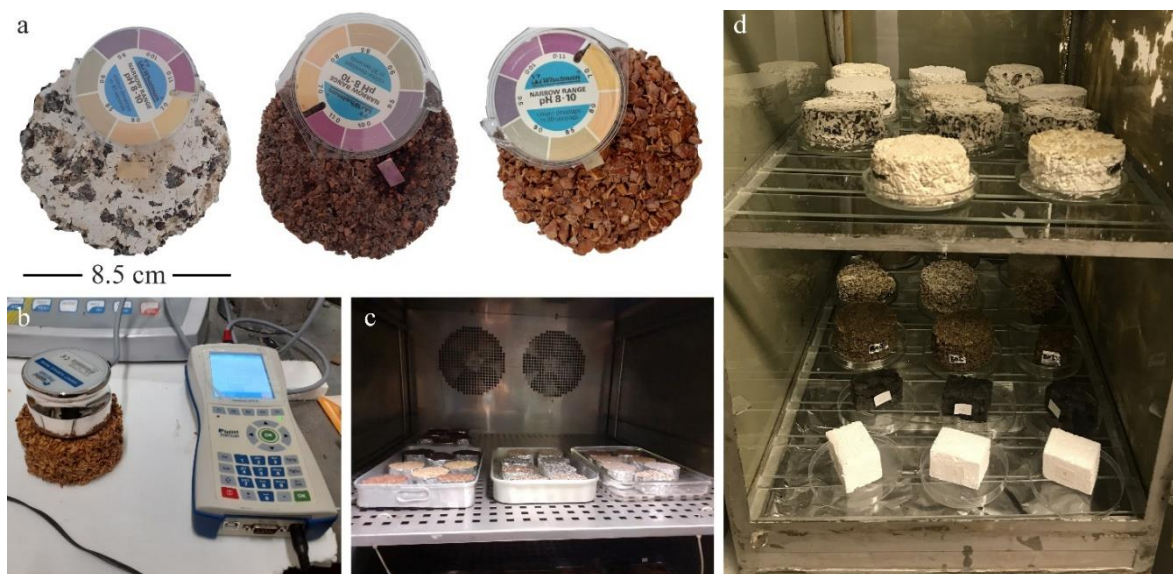


Figure 3.3-5. Main tests of the experimental campaign: (a) analysis of the pH by using the Litmus paper; (b) evaluation of the thermal conductivity by using the ISOMET; (c) samples in the climate chamber to assess the hygroscopicity; (d) samples in the climate chamber to assess the biological susceptibility to moulds, together with ICB and EPS panels (samples 5 cm x 3 cm x 3 cm).

### 3.3.2.3.2 Apparent density and thermal properties

The apparent density was calculated after curing and stabilizing the samples at  $T = 60\text{ }^{\circ}\text{C}$ , considered as having a change in mass less than 0.5 % after 24 h. It was determined by the average ratio between mass and volume of the three samples, adapted from the EN 323 [76]. The mass was assessed with a Precise Analytical Balance and the volume was calculated using a Helios Digital Caliper. The thermal properties were calculated after stabilising the samples at  $T = (17 \pm 1)\text{ }^{\circ}\text{C}$  and  $\text{RH} = (65 \pm 5)\%$ . Several methods could be considered to determine the thermal conductivity, namely the effectiveness in conducting heat: steady-state hot-plate, transient hot-bridge, hot-disk, photo-thermal methods [19]. The transient method was chosen according to past studies [32,77,78] and considering its several benefits [79]. Therefore, the thermal conductivity of the samples was evaluated by using an ISOMET 2114 Heat Transfer Analyser (Figure 3.3 5b) with a 60 mm diameter contact probe IPS 1105, ranging values between 0.04 and 0.30 W/(m.K). The average values of the three samples for each composite were considered.

### 3.3.2.3.3 Hygroscopicity and Moisture Buffering Value (MBV)

Sorption and desorption properties were calculated based on the ISO 24353 [80] but using the produced samples. The test allowed to define the hygroscopicity, namely the capacity of adsorbing/releasing moisture when relative humidity (RH) rises/decreases, respectively [81]. Three samples of each composite were covered by using a moisture barrier consisting of aluminium foil. Only the adsorption/desorption face (area =  $0.0057\text{ m}^2$ ) was exposed (Figure 3.3-5c). The samples were pre-conditioned in a climatic chamber (Aralab Fitoclima 1000) at  $T = (23 \pm 2)\text{ }^{\circ}\text{C}$  and  $\text{RH} = (63 \pm 2)\%$  until reaching a constant mass (change in mass of less than or equal to 0.1 % over 24 h). Four cycles



of 12 h were carried out, by conditioning at RH = 75 % (sorption phase) and RH = 50 % (desorption phase). After two cycles of sorption-desorption (aiming at reaching more stabilized conditions) the samples were weighed every 3 h to evaluate the moisture change during the time. Values of the moisture adsorption content,  $\rho_{A,ac}$ , the moisture desorption content,  $\rho_{A,dc}$ , and the moisture content difference,  $\rho_{A,sc}$  were calculated according to Equations 3.3-1, 3.3-2 and 3.3-3 to determine the variation of moisture adsorbed/desorbed content over time.

$$\rho_{A,ac} \left( \frac{\text{kg}}{\text{m}^2} \right) = \frac{m_{an} - m_{d(n-1)}}{A} \quad (3.3-1)$$

$$\rho_{A,dc} \left( \frac{\text{kg}}{\text{m}^2} \right) = \frac{m_{an} - m_{dn}}{A} \quad (3.3-2)$$

$$\rho_{A,sc} \left( \frac{\text{kg}}{\text{m}^2} \right) = \rho_{A,ac} - \rho_{A,dc} \quad (3.3-3)$$

$m_{an}$  [kg] is the value of the mass at the end of the sorption phase,  $m_{d(n-1)}$  [kg] is the value of the mass at the end of the desorption phase of the previous cycle,  $m_{dn}$  [kg] is the value of the mass at the end of the desorption phase,  $A$  [m<sup>2</sup>] is the exposed surface.

The moisture buffering value (MBV), which provides a numerical value of the absorbed/released moisture considering cycles with two levels of relative humidity (RH) [13], was calculated and rated according to an adaptation of Rode et al. [82]. RH and cycle times described by the ISO 24353 [80] were considered. The MBV was determined as the average value between the MBV of the sorption (MBV<sub>a</sub>) and the desorption phase (MBV<sub>d</sub>), calculated as reported in Equations 3.3-4 and 3.3-5, respectively. The last three cycles for each sample and their average values were considered (to evaluate the most stabilized conditions).

$$\text{MBV}_a \left( \frac{\text{g}}{\text{m}^2 \% \text{ RH}} \right) = \frac{m_{an} - m_{d(n-1)}}{A \times (\text{RH}_{\text{high}} - \text{RH}_{\text{low}})} \quad (3.3-4)$$

$$\text{MBV}_d \left( \frac{\text{g}}{\text{m}^2 \% \text{ RH}} \right) = \frac{m_{an} - m_{dn}}{A \times (\text{RH}_{\text{high}} - \text{RH}_{\text{low}})} \quad (3.3-5)$$

$\text{RH}_{\text{high}}$  is the highest value and  $\text{RH}_{\text{low}}$  is the lowest one, 75 % and 50 % respectively, according to ISO 24353 [80]. Ahead, MBV results will be compared with some literature values, considering also some calculated under different conditions of RH, which is an important information to highlight, since considering different methods (different cycles and RH), the MBV may change [83].

#### 3.3.2.3.4 Bio-susceptibility

Biological susceptibility was evaluated by exposing the samples at high RH, as this favours mould growth [84–86]. The samples were placed for four weeks in a conditioned room at  $T = (23 \pm 2) ^\circ\text{C}$  and  $\text{RH} \geq 85 \%$ , conditions that allow mould growth [87], without being sterilised or inoculated. Temperature and RH were constantly monitored by using a data logger (Hobo U12-011). It was placed close to the samples to record as much as possible the exposure conditions. Natural

ventilation was prevented, and the samples were kept spaced as shown in Figure 3.3-5d. Three samples (5 cm x 3 cm x 3 cm) of an expanded cork agglomerate (ICB) and three of an expanded polystyrene (EPS) panel, materials that demonstrated good resistance to mould growth [87,88], were used as controls. Fungal development was evaluated each week according to an adaptation of ISO 846 [89], considering method A. Table 3.3-6 reports the ratings of the top surface of the sample obtained either visually or by using a stereo microscope (Olympus SZX12).

Table 3.3-6. Rate of mould adapted from ISO 846 [89].

| Intensity of growth | Evaluation                                                                                                            |
|---------------------|-----------------------------------------------------------------------------------------------------------------------|
| 0                   | No growth apparent under the microscope                                                                               |
| 1                   | No growth visible to the naked eye, but clearly visible under the microscope, covering up to 25 % of the test surface |
| 2                   | Growth visible to the naked eye, covering up to 25 % of the test surface                                              |
| 3                   | Growth visible to the naked eye, covering up to 50 % of the test surface                                              |
| 4                   | Considerable growth, covering more than 50 % of the test surface                                                      |
| 5                   | Heavy growth, covering the entire test surface                                                                        |

#### 3.3.2.3.5 Statistical analysis and correlation between raw materials and composites' properties

An analysis of variance (ANOVA) was performed to evaluate significant differences among the different types of composites by using MATLAB R2023a. Two variables were evaluated: the adhesives and the bio-aggregates. A significance level of 5 % was considered and the P-values were analysed to define if the null hypothesis ( $H_0$  = there are no differences among the groups analysed: adhesives and bio-aggregates) was verified. Post-hoc tests were performed to distinguish the significantly different adhesives or bio-aggregates, by considering a multiple comparison test.

Finally, the results were compared to the ones of the bio-aggregates considered as raw materials, analysed in a previous study [61].

These analyses could help to define the contribution of each material in the final performance, pointing out when the properties were mainly influenced by the aggregates or the adhesives. In addition to providing important information for the discussion, it could be extremely useful for future studies, since they could support the choice of the materials for composites' production.

### 3.3.3 Results and discussion

#### 3.3.3.1 Visual observation and pH

The visual and qualitative observation allowed noticing that samples A seemed to be the most fragile ones after curing. They seemed weakly bonded and tended to crumble during their manipulation. Samples B and C appeared more resistant; the aggregates were well bounded.

Table 3.3-7 reports the average pH and the standard deviation of the specimens.

Table 3.3-7. pH average values and standard deviation of the composites.

| Composites                                        | pH $\pm$ standard deviation |
|---------------------------------------------------|-----------------------------|
| <b>Composites A with gypsum-based binder</b>      |                             |
| Grape press waste – AG                            | 8.4 $\pm$ 0.1               |
| Hazelnut shells – AH                              | 8.1 $\pm$ 0.1               |
| Olive press waste – AO                            | 8.9 $\pm$ 0.3               |
| Wood chips and spent coffee grounds – AWc         | 8.9 $\pm$ 0.1               |
| Wood chips – AW                                   | 8.2 $\pm$ 0.3               |
| <b>Composites B with sodium silicate solution</b> |                             |
| Grape press waste – BG                            | 11.0 $\pm$ 0.1              |
| Hazelnut shells – BH                              | 10.9 $\pm$ 0.3              |
| Olive press waste – BO                            | 10.9 $\pm$ 0.3              |
| Wood chips and spent coffee grounds – BWc         | 10.8 $\pm$ 0.1              |
| Wood chips – BW                                   | 11.0 $\pm$ 0.1              |
| <b>Composites C with natural glue</b>             |                             |
| Grape press waste – CG                            | 8.8 $\pm$ 0.3               |
| Hazelnut shells – CH                              | 8.5 $\pm$ 0.1               |
| Olive press waste – CO                            | 8.5 $\pm$ 0.1               |
| Wood chips and spent coffee grounds – CWc         | 8.8 $\pm$ 0.3               |
| Wood chips – CW                                   | 8.0 $\pm$ 0.1               |

The results confirmed the adhesives' influence on the properties of the final products. Composites B, bounded by the sodium silicate solution (pH = 12.40, Table 3.3-4), had values of pH of about 11. Composites A and C showed pH values of about 8.0-8.9 for all aggregates. The pH could be a predictor of bio-susceptibility [53,90], as previous research reported that alkaline materials (pH  $\geq$  10) can prevent chemical decomposition and moderate mould growth [85,91]. Therefore, good resistance to mould growth was expected from the B composites, but it is important to highlight both that this is a rapid evaluation, and that biological susceptibility depends on many factors.

### 3.3.3.2 Apparent density and thermal conductivity

Table 3.3-8 shows the values of bulk density and thermal conductivity of the analysed composites.

The results can be compared with some commercial products and data from the literature, namely the ones mentioned in Table 3.3-1; the values are in line with the literature ones. Figure 3.3-6 shows the correlation between the thermal conductivity and the apparent density of the studied composites, and the materials considered for comparison (reporting only the ones for which both density and thermal conductivity are detailed). When a range was reported, the average value was considered.

Table 3.3-8. Average apparent density and thermal conductivity with standard deviation of the composites.

| Composites                                        | Apparent density<br>T = 60 °C<br>[kg/m <sup>3</sup> ] | Thermal conductivity<br>T = 17 °C RH = 65 %<br>[W/(m·K)] |
|---------------------------------------------------|-------------------------------------------------------|----------------------------------------------------------|
| <b>Composites A with gypsum-based binder</b>      |                                                       |                                                          |
| Grape press waste – AG                            | 615.1 ± 22.7                                          | 0.103 ± 0.001                                            |
| Hazelnut shells – AH                              | 742.5 ± 7.5                                           | 0.134 ± 0.018                                            |
| Olive press waste – AO                            | 829.7 ± 8.5                                           | 0.096 ± 0.009                                            |
| Wood chips and spent coffee grounds – AWc         | 598.2 ± 17.9                                          | 0.078 ± 0.012                                            |
| Wood chips – AW                                   | 613.4 ± 8.4                                           | 0.090 ± 0.015                                            |
| <b>Composites B with sodium silicate solution</b> |                                                       |                                                          |
| Grape press waste – BG                            | 516.0 ± 48.5                                          | 0.068 ± 0.008                                            |
| Hazelnut shells – BH                              | 689.3 ± 29.4                                          | 0.079 ± 0.027                                            |
| Olive press waste – BO                            | 897.2 ± 68.4                                          | 0.101 ± 0.025                                            |
| Wood chips and spent coffee grounds – BWc         | 643.7 ± 6.2                                           | 0.100 ± 0.021                                            |
| Wood chips – BW                                   | 444.0 ± 5.2                                           | 0.057 ± 0.003                                            |
| <b>Composites C with natural glue</b>             |                                                       |                                                          |
| Grape press waste – CG                            | 402.4 ± 23.2                                          | 0.064 ± 0.001                                            |
| Hazelnut shells – CH                              | 524.2 ± 6.00                                          | 0.069 ± 0.003                                            |
| Olive press waste – CO                            | 602.3 ± 13.8                                          | 0.069 ± 0.006                                            |
| Wood chips and spent coffee grounds – CWc         | 392.7 ± 7.5                                           | 0.067 ± 0.002                                            |
| Wood chips – CW                                   | 344.4 ± 5.4                                           | 0.057 ± 0.005                                            |

\*Apparent density after curing and stabilized at T = 60 °C; thermal conductivity at T = (17 ± 5) °C, RH = (65 ± 5) %.

The analysed composites had higher values of apparent density and thermal conductivity than the considered commercial insulation boards. They showed better insulation thermal performance than an OSB panel [31] and plasterboard Knauf Standard A [29]. Only BW, CG and CW had better thermal performance than Celenit N [25]. Considering past studies [21,22], for which the maximum thermal conductivity value is 0.065 W/(m·K), neither of them could be considered as thermal insulation boards. According to past studies which reported a maximum value of 0.1 W/(m·K) [2,23,24], the composites respected the requirement, except for AH, AG, BWc and BO.

Composites A showed the lowest thermal insulation performance. Between them, grape press waste had the best insulation properties. The considered matrix could probably be better used to produce thermal mortars than insulation panels since the required thermal conductivity has to be lower than 0.2 W/(m·K) [6], although other requirements are also needed. For example, Fatma et al. [92] evaluated the properties of a gypsum mortar with palm fibre. They reported values of thermal conductivity between 0.29-0.51 W/(m·K) depending on the fibre's percentage and treatment. Adamopoulos et al. [93] analysed gypsum-based composites with different percentages of waste wood and reported thermal conductivity values between 0.209-0.312 W/(m·K). Posani et al. [78] considered mortars made up of hydraulic lime and cork aggregates and reported values of 0.098 W/(m·K) and 0.127 W/(m·K). Composites B showed more promising thermal insulation performance. The ones made up of grape press waste and wood chips had properties similar to Celenit N [25]. The others

had a high value of standard deviation. This is probably caused by the different aggregates' grain size in each sample and the sodium silicate distribution. The better thermal insulation performance might be determined by the presence of spaces between the aggregates, due to the production process (without pressing). Composites C showed the best thermal insulation performance, having the lowest values of apparent density and thermal conductivity. The good insulation performances of the composites B and C might be caused both by the presence of many voids and by the good thermal insulation properties of the adhesives. Further studies should be carried out.

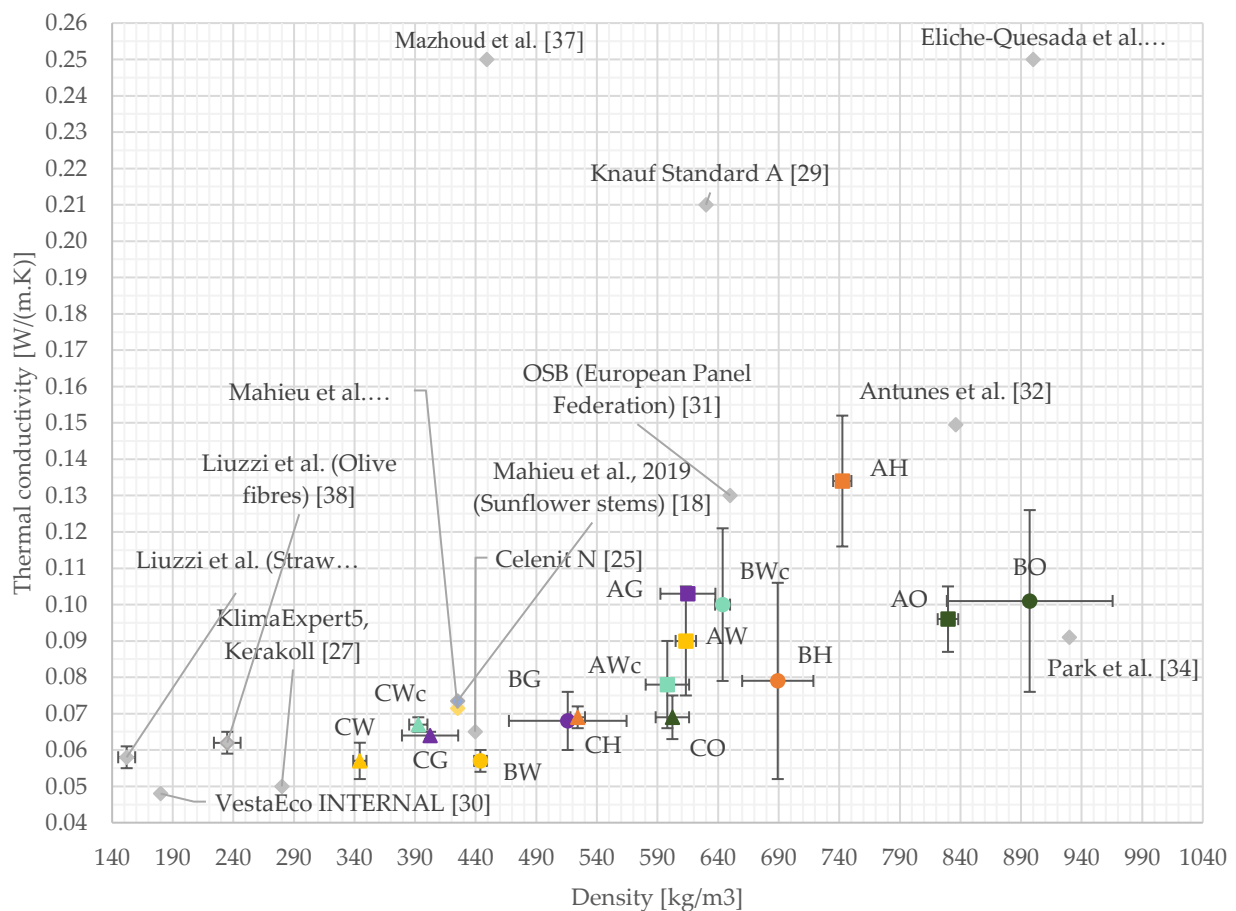


Figure 3.3-6. Correlation between thermal conductivity and density of the studied composites, compared with some literature and commercial values.

### 3.3.3.3 Hygroscopicity and Moisture Buffering Value

Figures 3.3-7, 3.3-8, and 3.3-9 report the four cycles of sorption/desorption for composites A, B, and C respectively.

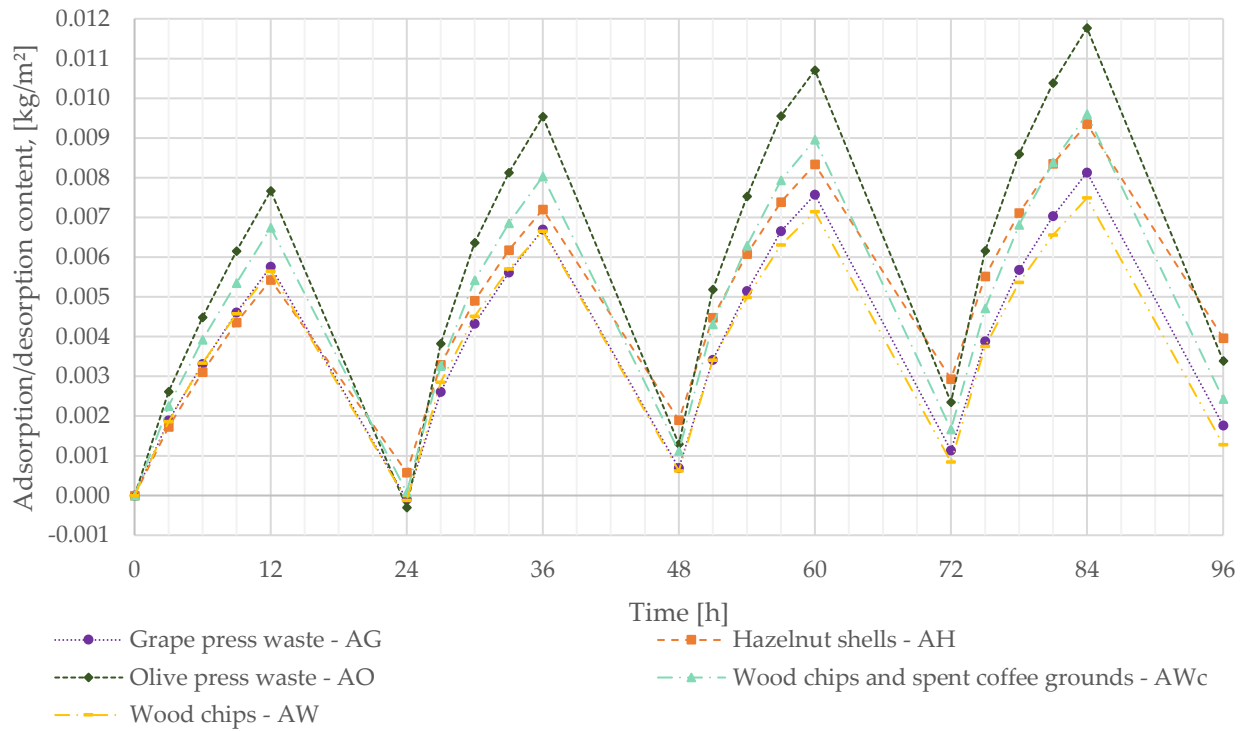


Figure 3.3-7. Moisture variation for four adsorption/desorption cycles of composites A (with gypsum-based binder).

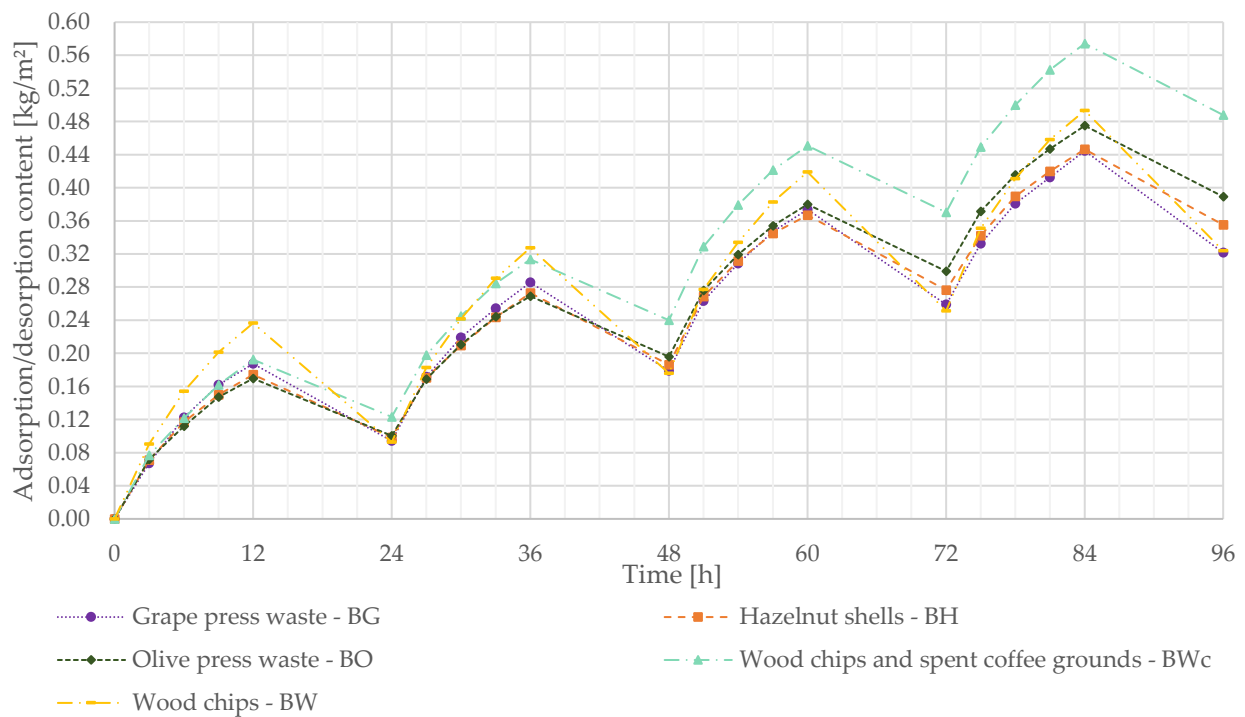


Figure 3.3-8. Moisture variation for four adsorption/desorption cycles of composites B (with sodium silicate solution).

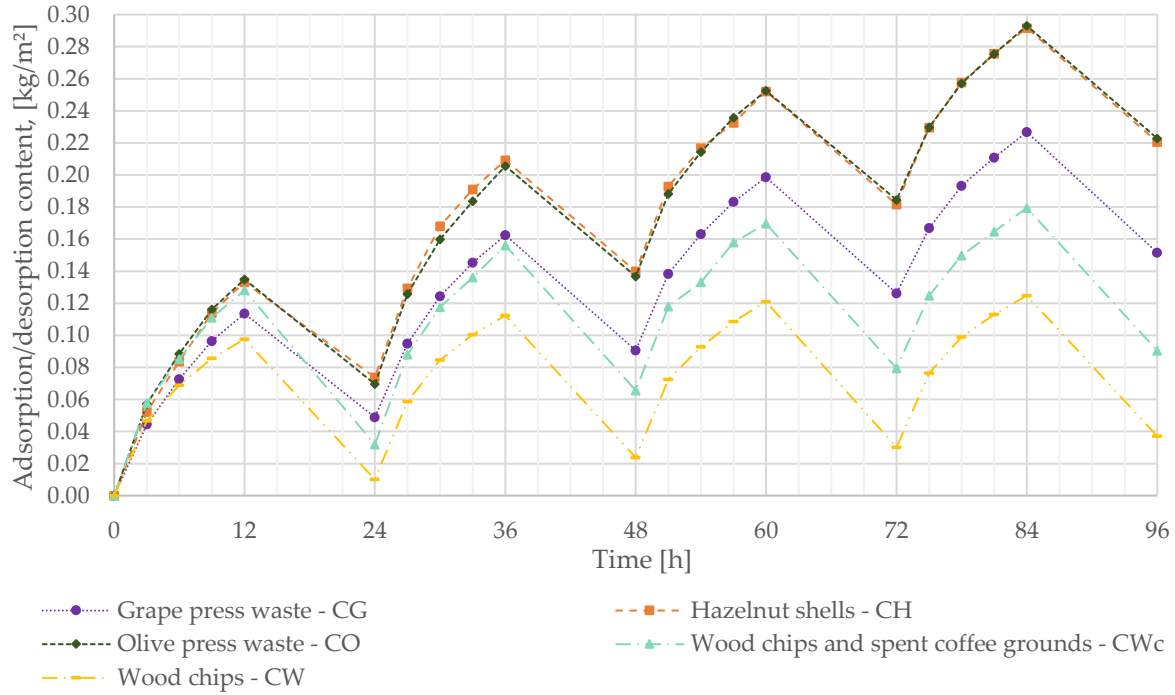


Figure 3.3-9. Moisture variation for four adsorption/desorption cycles of composites C (with starch-based natural glue).

For composites A, the ones made up of olive press wastes reached the highest values of moisture variation during the four cycles ( $\rho_{A,ac}$  between 0.008 kg/m<sup>2</sup> and 0.010 kg/m<sup>2</sup>;  $\rho_{A,dc}$  of 0.008 kg/m<sup>2</sup> for all cycles); hazelnut shells achieved the lowest ones ( $\rho_{A,ac}$  between 0.005 kg/m<sup>2</sup> and 0.007 kg/m<sup>2</sup>;  $\rho_{A,dc}$  of 0.005 kg/m<sup>2</sup> for all cycles). Composites made up of grape press waste and wood chips had similar performance. Values of moisture sorption and desorption were between 0.006 kg/m<sup>2</sup> and 0.007 kg/m<sup>2</sup> for both. The addition of spent coffee grounds in the composites made up of wood chips increased the moisture content during the sorption and desorption phases ( $\rho_{A,ac}$  between 0.007 kg/m<sup>2</sup> and 0.008 kg/m<sup>2</sup>;  $\rho_{A,dc}$  = 0.007 kg/m<sup>2</sup> for all cycles). For all composites, sorption capacity was higher than desorption, keeping remaining moisture after desorption. This demonstrated the high hygroscopicity of the bio-wastes.

Composites B showed the highest values of moisture content for wood chips ( $\rho_{A,ac}$  between 0.235 kg/m<sup>2</sup> and 0.243 kg/m<sup>2</sup>;  $\rho_{A,dc}$  between 0.144 kg/m<sup>2</sup> and 0.169 kg/m<sup>2</sup>) and grape press waste ( $\rho_{A,ac}$  between 0.187 kg/m<sup>2</sup> and 0.195 kg/m<sup>2</sup>;  $\rho_{A,dc}$  between 0.093 kg/m<sup>2</sup> and 0.123 kg/m<sup>2</sup>). Also in this case, composites made up of hazelnut shells reached the lowest values ( $\rho_{A,ac}$  between 0.170 kg/m<sup>2</sup> and 0.181 kg/m<sup>2</sup>;  $\rho_{A,dc}$  between 0.076 kg/m<sup>2</sup> and 0.091 kg/m<sup>2</sup>), similar to ones made up of olives press waste ( $\rho_{A,ac}$  between 0.169 kg/m<sup>2</sup> and 0.184 kg/m<sup>2</sup>;  $\rho_{A,dc}$  between 0.069 kg/m<sup>2</sup> and 0.086 kg/m<sup>2</sup>) and wood chips with spent coffee grounds ( $\rho_{A,ac}$  between 0.190 kg/m<sup>2</sup> and 0.211 kg/m<sup>2</sup>;  $\rho_{A,dc}$  between 0.069 kg/m<sup>2</sup> and 0.086 kg/m<sup>2</sup>). The moisture content difference of the composites B was higher than for composites A but similar between the different agro-industrial wastes. Probably the sodium

silicate solution had a higher influence than the aggregates on the hygroscopicity. In this case, the addition of spent coffee grounds in the wood chips-based composite lowered the moisture content.

Composites C showed a lower hygroscopicity capacity than composites B, and higher than composites A. This is in line with the expectation, as the adhesive is a natural glue, hence should have a high hygroscopicity. In this case, the highest values of moisture sorption were achieved by the composite made up of olive press waste ( $\rho_{A,ac}$  between 0.112 kg/m<sup>2</sup> and 0.140 kg/m<sup>2</sup>) and hazelnut shells ( $\rho_{A,ac}$  between 0.106 kg/m<sup>2</sup> and 0.131 kg/m<sup>2</sup>); while the lowest ones were the ones of wood chips ( $\rho_{A,ac}$  between 0.095 kg/m<sup>2</sup> and 0.102 kg/m<sup>2</sup>) and grape press waste ( $\rho_{A,ac}$  between 0.101 kg/m<sup>2</sup> and 0.114 kg/m<sup>2</sup>). The addition of spent coffee grounds in the sample made up of wood chips improved the moisture sorption content:  $\rho_{A,ac}$  between 0.100 kg/m<sup>2</sup> and 0.128 kg/m<sup>2</sup>. As for the desorption phases, the wood chips-based samples, with and without spent coffee grounds, achieved the highest values ( $\rho_{A,dc}$  between 0.089 kg/m<sup>2</sup> and 0.096 kg/m<sup>2</sup> and  $\rho_{A,dc}$  between 0.087 kg/m<sup>2</sup> and 0.091 kg/m<sup>2</sup>, respectively). The others showed similar performance:  $\rho_{A,dc}$  between 0.065 kg/m<sup>2</sup> and 0.075 kg/m<sup>2</sup>, for samples made up of grape press waste;  $\rho_{A,dc}$  between 0.067 kg/m<sup>2</sup> and 0.073 kg/m<sup>2</sup>, for olive press waste;  $\rho_{A,dc}$  between 0.058 kg/m<sup>2</sup> and 0.069 kg/m<sup>2</sup>, for hazelnut shells.

Table 3.3-9. MBV of the composites with gypsum-based binder A, sodium silicate solution B and natural glue C.

| Composites                                        | MBV [g/(m <sup>2</sup> .%RH)] |
|---------------------------------------------------|-------------------------------|
| <b>Composites A with gypsum-based binder</b>      |                               |
| Grape press waste – AG                            | 2.70 ± 0.18                   |
| Hazelnut shells – AH                              | 2.43 ± 0.33                   |
| Olive press waste – AO                            | 3.66 ± 0.36                   |
| Wood chips and spent coffee grounds – AWc         | 3.08 ± 0.23                   |
| Wood chips – AW                                   | 2.63 ± 0.14                   |
| <b>Composites B with sodium silicate solution</b> |                               |
| Grape press waste – BG                            | 6.11 ± 2.14                   |
| Hazelnut shells – BH                              | 5.12 ± 2.72                   |
| Olive press waste – BO                            | 5.29 ± 2.42                   |
| Wood chips and spent coffee grounds – BWc         | 5.63 ± 3.44                   |
| Wood chips – BW                                   | 8.06 ± 2.18                   |
| <b>Composites C with natural glue</b>             |                               |
| Grape press waste – CG                            | 3.61 ± 0.97                   |
| Hazelnut shells – CH                              | 3.78 ± 1.44                   |
| Olive press waste – CO                            | 3.79 ± 1.38                   |
| Wood chips and spent coffee grounds – CWc         | 3.99 ± 0.55                   |
| Wood chips – CW                                   | 3.74 ± 0.25                   |



Comparing the different binders, the sorption capacity was greater than desorption for all composites. The one made with sodium silicate solution (composites B) showed the greatest hygroscopicity capacity, followed by the ones with natural glue (composites C).

To better analyse the hygroscopicity properties, MBV was calculated for each sample as previously described. Table 3.3-9 reports the MBV.

Considering the classification proposed by Rode et al. [82], all the samples were classified as “excellent”. The results were compared with some literature values, reported in Table 3.3-10. They were calculated by different methods: by Nordtest [82], considering 8 h at high RH (75 %) followed by 16 h at low RH (33 %); considering MBV ideal as described in Nordtest [82]; MBV calculated from the results of a study that analysed moisture properties by using ISO 24353 [80]. The reference to RH variation allowed comparing MBV results even if calculated considering different conditions, but this comparison was carried out to give an idea of the moisture buffering capacity. Indeed, as previously anticipated, considering different methods gives different MBV results [83].

Table 3.3-10. MBV of different bio-based composites, reported in literature.

| Materials/Composites                                                                                                                                              | MBV [g/(m <sup>2</sup> .%RH)] | Classification  | Ref. |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------|-----------------|------|
| Composites with wood fibre, cellulose wadding, and paper granules with potato starch <sup>a</sup>                                                                 | 1.1-2.5                       | Good/ Excellent | [94] |
| Composites with hemp shiv and clay <sup>a</sup>                                                                                                                   | 2.07-2.28                     | Excellent       | [37] |
| Composites with hemp shiv and clay stabilized with 5% of air lime-based binder (Thermo®) from BCB and 5% of Portland cement (CEM I 52.5 N CE CP2 NF) <sup>a</sup> | 2.24-2.33                     | Excellent       | [37] |
| Hemp concrete: hemp and binder paste with around 75% of air lime, 15% of hydraulic binder and 10% of pozzolan <sup>a</sup>                                        | 1.84-2.02                     | Excellent       | [95] |
| Rape straw concrete: rape straw and binder paste with around 75% air lime, 15% hydraulic binder and 10% pozzolan <sup>a</sup>                                     | 2.29-2.59                     | Excellent       | [95] |
| Composites with olive fibre and sodium silicate solution <sup>b</sup>                                                                                             | 5.05                          | Excellent       | [38] |
| Composites with straw fibre and sodium silicate solution <sup>b</sup>                                                                                             | 3.29                          | Excellent       | [38] |
| Clay plaster with hemp shiv and fibres <sup>c</sup>                                                                                                               | 2.20                          | Excellent       | [96] |
| Wood-based panel with coniferous wood chips and cement (not specified) <sup>a</sup>                                                                               | 3.25                          | Excellent       | [97] |
| Chipboard panel <sup>a</sup>                                                                                                                                      | 2.25                          | Excellent       | [97] |
| OSB panel <sup>a</sup>                                                                                                                                            | 1.25                          | Good            | [97] |
| Hemp fibre panel <sup>a</sup>                                                                                                                                     | 2.25                          | Excellent       | [97] |
| Wood fibre panel <sup>a</sup>                                                                                                                                     | 3.10                          | Excellent       | [97] |

<sup>a</sup> MBV calculated by Nordtest method [82]: 8 h at high RH (75%) followed by 16 h at low RH (33%);

<sup>b</sup> MBV ideal calculated according to Nordtest project [82];

<sup>c</sup> MBV calculated from the results of a study that analysed moisture properties by using ISO 24353 [80].

Tables 3.3-9 and 3.3-10 show the high hygroscopicity capacity of bio-based composites, as expected [9,32,98]. All of them were classified as good or excellent. Furthermore, the comparison demonstrated that the results were in line with literature values. Composites A could be better compared with gypsum boards or gypsum and lime plasters. Ramos et al. [99] calculated the MBV of a gypsum

board, gypsum plaster and gypsum-air lime (50 % - 50 %) plaster (not reported in Table 3.3-10 since they are not bio-based composites). The researchers achieved values between 0.7 -0.8 g/(m<sup>2</sup>.%RH) for a gypsum board and a gypsum plaster, between 0.4 - 0.5 g/(m<sup>2</sup>.%RH) for a gypsum-air lime plaster, which are not high in comparison to bio-based products. This demonstrates that the addition of agro-industrial wastes can improve the hygroscopicity capacity of gypsum-based gypsum products. As for the composites made up of agro-industrial waste and sodium silicate solution, also past studies reported high values of MBV [38]. As for starch-based composites, Mnasri et al. [97] reported that a starch-based binder improves the MBV, even if Thiebleson et al. [94] reached lower values than the results of the present study. High hygroscopicity capacity could be good for coating panels since the composites could contribute to a passive equilibrium of moisture indoor conditions and, therefore, to comfort and health. On the other side, this could be a drawback for the biological attack: moisture storage inside the composites could encourage mould growth.

### 3.3.3.4 Bio-susceptibility to moulds

Table 3.3-11 shows the results of bio-susceptibility to moulds, considering the average values of the evaluation (rating from 0 to 5 as described in Section 3.3.2.3).

Table 3.3-11. Mould development on the composites and control commercial ICB and EPS evaluated according to ISO 846 grading scale [89].

| Composites                                        | Week1 | Week2 | Week3 | Week4 |
|---------------------------------------------------|-------|-------|-------|-------|
| <b>Composites A with gypsum-based binder</b>      |       |       |       |       |
| Grape press waste – AG                            | 0.67  | 0.67  | 2.00  | 2.67  |
| Hazelnut shells – AH                              | 0.00  | 0.00  | 0.00  | 0.00  |
| Olive press waste – AO                            | 0.00  | 0.67  | 2.67  | 4.00  |
| Wood chips and spent coffee grounds – AWc         | 0.00  | 0.00  | 0.00  | 0.00  |
| Wood chips – AW                                   | 0.00  | 0.00  | 0.00  | 0.00  |
| <b>Composites B with sodium silicate solution</b> |       |       |       |       |
| Grape press waste – BG                            | 0.00  | 0.00  | 0.00  | 0.00  |
| Hazelnut shells – BH                              | 0.00  | 0.00  | 0.00  | 0.00  |
| Olive press waste – BO                            | 0.00  | 0.00  | 0.00  | 0.00  |
| Wood chips and spent coffee grounds – BWc         | 0.00  | 0.00  | 0.00  | 0.00  |
| Wood chips – BW                                   | 0.00  | 0.00  | 0.00  | 0.00  |
| <b>Composites C with natural glue</b>             |       |       |       |       |
| Grape press waste – CG                            | 0.00  | 5.00  | 5.00  | 5.00  |
| Hazelnut shells – CH                              | 0.33  | 1.00  | 4.00  | 4.00  |
| Olive press waste – CO                            | 1.00  | 4.33  | 5.00  | 5.00  |
| Wood chips and spent coffee grounds – CWc         | 1.00  | 2.00  | 5.00  | 5.00  |
| Wood chips – CW                                   | 0.33  | 1.00  | 1.00  | 3.00  |
| Expanded cork agglomerate – ICB                   | 0.00  | 0.00  | 0.00  | 0.00  |
| Expanded polystyrene – EPS                        | 0.00  | 0.00  | 0.00  | 0.00  |

Figure 3.3-10 shows the centre square (4 cm<sup>2</sup>) of the contaminated composites at the end of the four weeks.

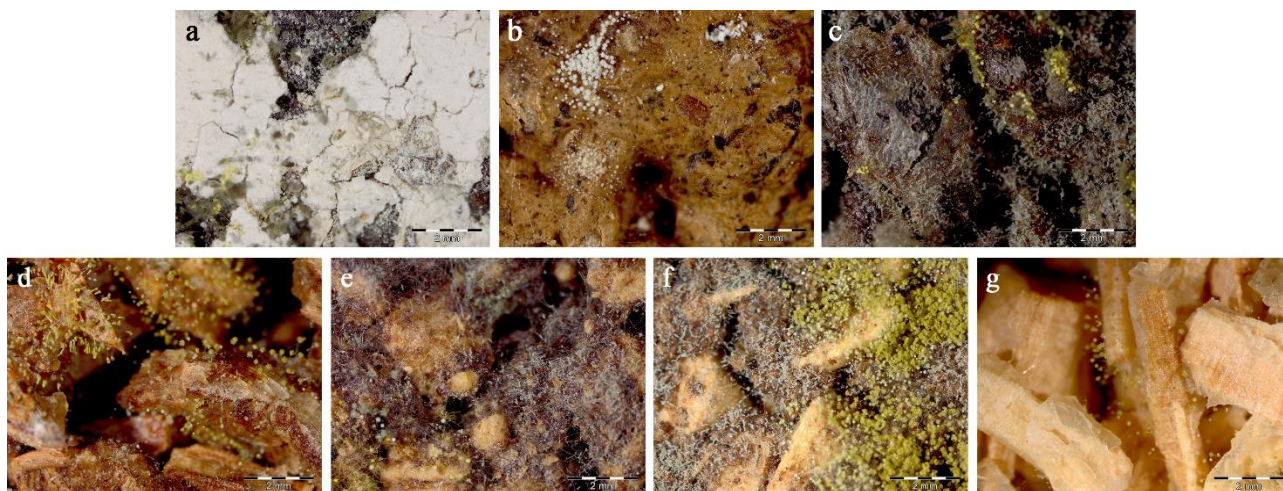


Figure 3.3-10. Mould development at the end of the four weeks: (a) AG; (b) AO; (c) CG; (d) CH; (e) CO; (f) CWc; (g) CW.

The results demonstrated the influence of the adhesive on the biological resistance of the tested composites. Composites B showed the highest resistance to mould, as expected. According to the literature, a pH higher than 10 can prevent mould growth [85,90,91]. Considering Table 3.3-7, the results were in line with the expectations. Nevertheless, the evaluation of the biological resistance cannot be related only to the pH values since other factors play an important role, as previously reported. Composites A, with pH values between 8.1 and 8.9, demonstrated better biological resistance than expected, especially for hazelnut shells, wood chips and wood chips with spent coffee grounds that did not present mould growth. The others (made with grape and olive press waste) never reached the highest values. The starch-based glue (composites C) had the lowest resistance to mould. After the first week, all the samples were contaminated. Samples made with grape press waste reached the maximum values during the second week; for the ones made up of olive press waste and spent coffee grounds with wood chips it was during the third week. Furthermore, it seems that the addition of spent coffee grounds decreased the resistance to mould growth of wood-based samples (Figures 3.3-10f and 3.3-10g). These results were consistent with past research: starch-based composites had low resistance to biological attack and were affected by humidity [100–102]. This property could probably be improved by adding higher quantities of citric acid or other biocidal agents.

The controls were rated as 0.00 during the four weeks and these results are consistent with the literature. The critical moisture level for EPS is 90-95 % [87], hence the samples were supposed not to be affected by mould growth at the considered RH. ICB was elsewhere rated as 1.00 but considering higher values of RH [88]. This demonstrated both the validity of the test and the low biological resistance of composites A and C. As for composites B, they were rated as the controls, as for the considered conditions they showed the same resistance to moulds.

Comparing the agro-industrial wastes, grape and olive press waste had the lowest biological resistance, while wood chips and hazelnut shells had the highest one. This was in line with the results of past work that considered the hygroscopicity properties of the aggregates [61]. The higher moisture buffering capacity created more favourable conditions for mould growth, even if considering only this information is an oversimplification, as Cintura et al. [61] reported. Certainly, the results demonstrated that the composites' final properties depend on both the aggregate and the binder.

### 3.3.3.5 Statistical analysis and correlation between raw materials and composites' properties

The statistical analysis showed that considering the adhesives as the variable, for all the properties, the null hypothesis was not confirmed (P-value less than 0.05), hence at least one of the mean values was different. As for the bio-aggregates, for all the properties the alternative hypothesis was not verified (i.e., P-value greater than 0.05, the mean values were the same).

Table 3.3-12 summarizes the results of the ANOVA (reporting F and P-values) for the significantly different results, namely when the adhesives were considered as the variable.

Table 3.3-12. Results of ANOVA (F and P values) for the results which were significant (adhesives as the variable).

| Property                                        | F      | P-value |
|-------------------------------------------------|--------|---------|
| pH                                              | 112.13 | <0.001  |
| Apparent density                                | 4.17   | 0.042   |
| Thermal conductivity                            | 5.46   | 0.021   |
| MBV                                             | 23.59  | <0.001  |
| Bio-susceptibility (mould growth after 4 weeks) | 17.52  | <0.001  |

Table 3.3-13 reports the p-values of the post-hoc tests for the results, which were significant (adhesives as the variable). The first two columns show which groups were compared with each other (A – gypsum-base binder, B – sodium silicate solution, C – natural glue, as defined in Section 3.3.2.2.). The p-values allowed identifying which and how the groups were significantly different from each other.

These statistical analyses showed a strong influence of the adhesives on the analysed properties. Therefore, as previously reported, the gypsum-based binder, the sodium silicate solution and the starch-based glue predominated over the aggregates in the final performance of the composites. However, further, and detailed analysis should be performed to confirm this conclusion.

The results were compared with the ones obtained for raw materials reported in a previous study [61] to evaluate eventual correlations. Values of spent coffee grounds were not reported since this agro-industrial waste was only used as fine aggregate and evaluated together with wood chips. Table 3.3-14 shows the values in different font-style: the highest ones are highlighted in bold; the second

ones (in descending order) are in *italics*, the third ones (in descending order) are not edited (normal), and the smallest ones are underlined.

Table 3.3-13. p-values of the post-hoc tests for the results which were significant (adhesives as the variable).

| Group1               | Group2 | p-value* |
|----------------------|--------|----------|
| pH                   |        |          |
| A                    | B      | 0.0000   |
| A                    | C      | 0.9936   |
| B                    | C      | 0.0000   |
| Apparent density     |        |          |
| A                    | B      | 0.8729   |
| A                    | C      | 0.0462   |
| B                    | C      | 0.1093   |
| Thermal conductivity |        |          |
| A                    | B      | 0.2079   |
| A                    | C      | 0.0162   |
| B                    | C      | 0.3300   |
| MBV                  |        |          |
| A                    | B      | 0.0001   |
| A                    | C      | 0.1899   |
| B                    | C      | 0.0012   |
| Bio-susceptibility   |        |          |
| A                    | B      | 0.2276   |
| A                    | C      | 0.0045   |
| B                    | C      | 0.0002   |

\*Note: the numbers in bold represent  $p < 0.05$ .

For loose bulk density and apparent density, it seemed to be a correlation between raw materials and composites (considering also standard deviation), except for composite BG, made up of grape press waste. The lower apparent density of these samples could be caused by the production process: the use of sodium silicate solution without pressing might produce internal voids.

As for thermal conductivity and MBV, there was no linear correspondence (e.g., a lower value of thermal conductivity for the aggregates corresponds to a higher value for composites A). However, the results of thermal conductivity were in line with past studies since the waste aggregates considered individually did not show good thermal insulation properties [103]. Considering the sorption/desorption properties of the bio-wastes as raw materials, olive press waste achieved the highest values of moisture adsorption and desorption content, while hazelnut shells had the lowest ones. Hence, for composites A there was a correlation between the raw materials and composites properties. For the other composites (B and C), there was not a certain correlation between raw material and composites. As for biological susceptibility, the comparison was not reported because different types of biological attacks were considered.

Table 3.3-14. Comparison between properties of raw materials [61] and composites considering the three analysed adhesives.

| Aggregates        | Loose bulk density    |                      | Apparent density     |                      |
|-------------------|-----------------------|----------------------|----------------------|----------------------|
|                   | Raw materials         | Composites A         | Composites B         | Composites C         |
| Grape press waste | 343.88 ± 12.00        | 615.1 ± 22.7         | <u>516.0 ± 48.5</u>  | 402.4 ± 23.2         |
| Hazelnut shells   | <b>550.50 ± 19.53</b> | <i>742.5 ± 7.5</i>   | <i>689.3 ± 29.4</i>  | <i>524.2 ± 6.00</i>  |
| Olive press waste | <i>449.43 ± 8.60</i>  | <b>829.7 ± 8.5</b>   | <b>897.2 ± 68.4</b>  | <b>602.3 ± 13.8</b>  |
| Wood chips        | <u>256.98 ± 8.10</u>  | <u>613.4 ± 8.4</u>   | 643.7 ± 6.2          | <u>344.4 ± 5.4</u>   |
|                   | Thermal conductivity  |                      |                      |                      |
|                   | Raw materials         | Composites A         | Composites B         | Composites C         |
| Grape press waste | <u>0.081 ± 0.002</u>  | <i>0.103 ± 0.001</i> | 0.068 ± 0.008        | 0.064 ± 0.001        |
| Hazelnut shells   | <b>0.115 ± 0.003</b>  | <b>0.134 ± 0.018</b> | <i>0.079 ± 0.027</i> | <i>0.069 ± 0.003</i> |
| Olive press waste | <i>0.097 ± 0.003</i>  | 0.096 ± 0.009        | <b>0.101 ± 0.025</b> | <b>0.069 ± 0.006</b> |
| Wood chips        | 0.082 ± 0.002         | <u>0.090 ± 0.015</u> | <u>0.057 ± 0.003</u> | <u>0.057 ± 0.005</u> |
|                   | MBV                   |                      |                      |                      |
|                   | Raw materials         | Composites A         | Composites B         | Composites C         |
| Grape press waste | <i>3.54 ± 0.09</i>    | <i>2.70 ± 0.18</i>   | <i>6.11 ± 2.14</i>   | <u>3.61 ± 0.97</u>   |
| Hazelnut shells   | 3.13 ± 0.16           | <u>2.43 ± 0.33</u>   | <u>5.12 ± 2.72</u>   | <i>3.78 ± 1.44</i>   |
| Olive press waste | <b>4.38 ± 0.13</b>    | <b>3.66 ± 0.36</b>   | 5.29 ± 2.42          | <b>3.79 ± 1.38</b>   |
| Wood chips        | <u>3.10 ± 0.06</u>    | 2.63 ± 0.14          | <b>8.06 ± 2.18</b>   | 3.74 ± 0.25          |

Note: The values are classified considering the descending order by using different font styles: bold = highest values; italic = second; normal = third; underlined = smallest values.

These results demonstrated the high impact of the binders on the properties of the final products, as expected [13,102]. It is important to specify that these considerations are more qualitative than quantitative, and they could be considered a rapid and initial assessment. To achieve more detailed considerations, further studies should be carried out.

### 3.3.4 Conclusions

This study investigated the feasibility of producing agro-industrial waste-based insulation boards or coating panels with hygrothermal performance. The outcomes are preliminary results, useful to carry out future studies about the production of eco-efficient composites.

Three different adhesives were evaluated: a gypsum-based binder, a sodium silicate solution, and a starch-based natural glue. The present study provides a first evaluation of three adhesives.

As for the aggregates, the considered agro-industrial wastes were grape press waste, olive press waste, hazelnut shells and spent coffee grounds. Maritime pine chips were considered as control. The influence of the coffee waste fine aggregates was only assessed together with the control aggregate. Apparent density, thermal conductivity, hygroscopicity and resistance to mould growth at high relative humidity were assessed.

The following conclusions can be achieved:

- Mixing the considered agro-industrial wastes with a gypsum-based binder, with air lime and citric acid, may be an efficient solution to produce thermal plastering mortars, better than for thermal insulation boards or coating panels with good thermal insulation performance. These composites could be further analysed to evaluate their properties as such. The addition of citric acid may be interesting to improve the resistance to biological attack of bio-waste and gypsum-based composites.
- Using sodium silicate solution as an adhesive could be an efficient solution to produce thermal insulation boards or coating panels. It provides high hygroscopicity, advantageous behaviour for coating panels to equilibrate the indoor RH levels, and good resistance to mould growth. This solution needs to be further detailed and optimized, considering more information (e.g., the mechanical properties, different types of biological attacks, and different compositions of binder-aggregates).
- A starch-based natural glue could be a good possibility to produce thermal insulation boards, considering the results of apparent density and thermal conductivity. Nevertheless, this binder demonstrated low resistance to mould, even when citric acid was added. This solution could be further investigated, trying to improve the biological resistance (by adding more citric acid or other biocidal products).
- Comparing the different agro-industrial wastes, grape press waste and hazelnut shells show the best properties in terms of thermal insulation performance and biological susceptibility. However, it is difficult to find out a certain correlation between the raw materials and composites. Even when knowing the properties of the agro-industrial wastes it seems mandatory to assess the properties of the composites, as the selected adhesive widely affects the final performance.

Grape press waste and hazelnut shells mixed with the sodium silicate solution could be a feasible solution to produce bio-based thermal insulation boards and coating panels. Starch-based natural glue could be another interesting possibility if resistance to mould growth is improved. An LCA analysis will be performed to assess the global performance of the composites.

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### 3.4 Concluding remarks

The results allowed achieving important conclusions that supported the next steps of the project. Furthermore, through the implementation of the theoretical knowledge acquired by the literature review, the practical skills for particleboards' production and testing improved.

The characterization of the bio-aggregates (Section 3.2) pointed out the main difference between the selected agro-industrial wastes. Grapes press waste, hazelnut shells, and olives press waste have similar grain size distributions and they could be used as medium-size aggregates; the spent coffee grounds could be used as fine aggregates. The considered bio-wastes could be not shredded and employed as they were provided. This could lower the energy waste and environmental impact derived from the production process. As for the evaluated properties, the selected materials did not show good thermal insulation performance, which might be improved by considering strategies such as pre-treatments, changing the grain size, or the addition of other materials in the final composites, such as more performant bio-resources or some additives. They are highly hygroscopic and showed high MBV, an advantage for passive control of indoor relative humidity and internal comfort. The resistance to biological attack was low, as expected, and should be improved by specific strategies (e.g., by adding materials that can moderate it, or pre-treatments). All these results and considerations are important for future studies.

The experimental campaign carried out considering the three different adhesives (Section 3.3), demonstrated, the feasibility of producing agro-industrial waste-based insulation boards by using the selected materials. The results allowed making important considerations, useful for carrying out the production of indoor particleboards. Indeed, this part of the work ensured the selection of the materials that will be considered in the next steps of the project: hazelnut shells as aggregates, and the sodium silicate solution as the adhesive. The choice of the aggregates was made both considering the results of the experimental campaign and the literature review. The availability throughout the year in large quantities has been of fundamental importance, as better detailed below. As for the sodium silicate solution, it was selected because it could improve some of the main drawbacks of bio-based composites, such as the resistance to biological attack and fire, even if these properties must be verified in the produced composites. It is important to clarify that the evaluation of MBV (Section 3.3) was carried out to provide an idea of the hygroscopic behaviour. The calculation of the MBV was used as a tool of comparison between the different composites tested. Indeed, the indications provided by the considered standard were followed, but the determination of the MBV was a simplification. The same disclosure must be applied in the other part of the project that considered MBV calculations (Chapters 4 and 5). More detailed information on the materials' choice is provided in Chapter 5.



However, also the unchosen materials could be considered for further studies since all of them showed interesting properties. The other evaluated bio-aggregates could be used to produce panels or other building composites, as well as the other evaluated adhesives. Among these, starch-based natural glue seemed to be much more promising for thermal insulation boards, but it has not been chosen because it is a food product. The idea is to evaluate the possibility of reusing by-products (bio-waste), rather than bio-products. The gypsum-based binder with the bio-aggregates could be an interesting possibility to develop eco-efficient plaster with thermal mortars.

Finally, an important contribution of these first experimental campaigns was the application of the previously mentioned methods of analysis (Section 2.6). The information collected during that part of the literature review provided guidance for the production and study of samples. Then, practical tests allowed knowing the considered materials, not only from a theoretical point of view but also by laboratory analysis, an important step for the development of a new composite.



## BIO-BASED BOARDS MADE OF BIO-AGGREGATES AND SODIUM SILICATE

### 4.1 Preamble

After selecting the materials (i.e., hazelnut shells as bio-aggregates and a sodium silicate solution as the adhesive) further analysis was carried out to produce the indoor particleboard samples and test their properties. Furthermore, as a possible optimization, the addition of another bio-resource (*Arundo donax*) was considered. It was not assessed from the start as it is not classified as an agro-industrial waste. The choice of this bio-resource was made because it is largely available in many European countries, such as Portugal and Italy, and more alternatives for its valorisation are important to reduce the high volumes to manage (Carneiro et al., 2016). Further information about this choice is provided below. Both the aggregates were selected considering their availability over the year as well, moderating the problem of seasonality and, hopefully, the ones derived from climate change.

To assess this possible optimization, first, a composite made of the bio-resource and the sodium silicate solution was produced and tested. Then, the composite made of the mixture of the two bio-aggregates was considered, as detailed in Chapter 5. These parts of the project were carried out in collaboration with Alma Mater Studiorum - University of Bologna.

The hazelnut shells and the sodium silicate solution were provided by local producers, which also helped to better describe their production process and properties. Since the considered hazelnut shells (from Italy) could be different from the ones used in the previous experimental campaign (from Portugal), their characterization was provided, performing the same test methods described in Chapter 3. As for the adhesive, a sodium silicate solution having the same properties as the already tested one was employed.

To improve the properties of the previously produced composite samples, studies to define the production process were carried out (Section 4.2). Several practical tests were performed on:

- hazelnut shells' grain size;
- percentages of aggregates-adhesives (to maximize the first, minimize the sodium silicate, and provide enough mechanical resistance);
- type of mould (wooden, silicon, metal);
- production process (e.g., time of mixing, moulding, demoulding);
- casting and curing phase;
- drying process.

At the same time, the new experimental campaign was planned to define the laboratory tests, the required equipment, and the time analysis, and to produce composite samples with different sizes, shapes, and repetitions according to the considered references and standards. The initial literature review (Section 2.6) supported the choices. Several samples were produced, and laboratory tests were performed to evaluate the physical, hygrothermal, acoustic, and mechanical properties (Section 4.3).

As for the optimization, namely the improvement of the low mechanical performance of the hazelnut shells-based composite, the addition of *Arundo donax* was considered. *A. donax*, known as giant reed or giant cane, is a non-food invasive crop native to Asia, now present worldwide, particularly in southern Europe, due to its ability to tolerate different climates and soil conditions. This herbaceous, perennial plant was selected for its availability, its promising mechanical performance, and its thermal insulation capacity (Barreca, 2012; Malheiro et al., 2021). Hence, it could guarantee better mechanical performance to the final composite, contributing to indoor comfort as well.

*A. donax* was harvested and prepared considering the same process employed for hazelnut shells. A similar grain size was considered to lower the number of different parameters, for an easier comparison between the materials and the composites. So, samples of *A. donax* and sodium silicate were produced and tested (Section 4.4). The same laboratory tests considered for the hazelnut shell-based composites were performed, together with the assessment of air flow resistivity. The aim was to evaluate the properties of composites made of the two aggregates separately, before evaluating the mixture of the two, namely the optimized boards (Chapter 5).

For both the experimental campaigns a comparison with past studies and commercial products was carried out to discuss and validate the results. Past research was mainly considered as this ensured easier comparisons and more detailed considerations. Indeed, past studies provide detailed descriptions of samples' compositions, laboratory test methods, and discussion of the results. The commercial products are already optimized and then, they could easily show better performance.

## 4.2 Article E - Optimisation of production parameters to develop innovative eco-efficient board

The information reported in this Section was presented in the 5<sup>th</sup> International Conference on Bio-Based Building Materials - ICBBM 2023, 21-23 June, Vienna, Austria, and published in: Amziane, S., Merta, I., Page, J. (eds) Bio-Based Building Materials. ICBBM 2023. RILEM Bookseries, vol 45. Springer, Cham, 111-122 - [https://doi.org/10.1007/978-3-031-33465-8\\_10](https://doi.org/10.1007/978-3-031-33465-8_10)

### Abstract

Laboratory tests were carried out to define production parameters of innovative eco-efficient composites made up of hazelnut shells as aggregate and a sodium silicate solution as adhesive. The aim was to maximize the content of bio-aggregates and minimize the amount of adhesive, guaranteeing the feasibility of producing samples. Therefore, after preliminary testing, the percentages of hazelnut shells and the sodium silicate solution were kept constant: 70% and 30% of the total volume, respectively. However, the characteristics of the considered composites did not allow the production of uniform samples. The sodium silicate solution was not rapidly absorbed by the bio-aggregates; during the drying process, it was deposited on the bottom side of the samples. The uniformity of the samples is required to guarantee a correct evaluation of their performance and future homogeneous panels. Hence, different production parameters were investigated, such as drying at  $T = 60\text{ }^{\circ}\text{C}$  or  $T = 80\text{ }^{\circ}\text{C}$  during different periods of time, and the addition of different percentages of sodium bicarbonate was also considered. The visual analysis during drying, the final uniformity, thus the distribution of the sodium silicate solution, the resistance and the crumbliness of the samples allowed defining the best production process. The most uniform sample was selected, and its production parameters can thus be applied to produce innovative composites.

**Keywords:** Bio-waste, Hazelnut shells, Sodium silicate.

### 4.2.1 Introduction

The use of panels such as particleboards as building materials has several benefits. Besides the easy on-site installation, the economic advantages, and the several applications, there is the possibility of employing different materials as aggregates. Among them, recycled materials and by-products derived from agro-industrial practices [1,2]. Considering the high environmental impact caused by both the construction sector and agriculture [3], the production of agricultural waste-based materials seems to be an efficient solution that encourages a circular economy system [4]. Several studies addressed the feasibility of producing panels by using agro-industrial wastes, for example, cork,

coffee chaff, rice husk and corn cob [5-7]. Many of the developed composites met the requirements of the standards, proving to be a good alternative to conventional materials.

In the development of these innovative composites, the origin of the considered agro-industrial wastes is extremely important, too. Using local by-products lowers, even more, the environmental impact, moderating the transport variable. Considering previous work [8] hazelnut shells were selected as agro-industrial waste. Indeed, according to FAO [9], the first three producers of hazelnut in 2020 were Turkey, Italy, and the United States. They produced 665,000 tonnes, 140,560 tonnes, and 64,410 tonnes, respectively.

Besides the production, availability was considered. The seasonality of agricultural products could be an obstacle to their supply. Nevertheless, hazelnuts are available all over the year, as reported by the industry. Indeed, they are harvested in August and September, and they are left to dry naturally or mechanically [10], a process that can require some months. The drying process guarantees several benefits, such as the improvement of the chemical and physical stability of the food and a greater resistance to mould growth [10,11]. After drying, the hazelnuts are normally stored. The maximum storage period varies depending on the storage conditions and it could be even 48 months [10,12]. Hence, there is a large quantity of hazelnut shells available, throughout the year, and not only during the harvesting season.

Another important parameter to produce eco-efficient composites, such as boards and panels, is the selected adhesive [13,14]. A sodium silicate solution, also known as water glass, was used for its several benefits, such as harmlessness for human health (formaldehyde-free adhesive), high resistance to mould, and the prevention of chemical decomposition [15-17]. However, it has also some drawbacks, such as high hygroscopicity, and hence low moisture resistance [18,19]. Furthermore, the sodium silicate solutions may be highly fluid and require much time for the hardening process. To improve the bonding and the setting time some strategies can be considered, such as heat or chemical treatments [20]. Several additives may accelerate the hardening process of the composites (e.g., sodium carbonate, sodium bicarbonate, aluminium sulphate [21,22]), as well as drying at high temperatures.

Starting from this knowledge, the feasibility of producing boards made up of hazelnut shells as aggregates and sodium silicate solution as the adhesive was investigated. Practical tests were carried out to define the best production process. Different parameters were analysed, such as drying the samples at different temperatures for different durations, and the addition of sodium bicarbonate intended to accelerate the hardening process. The problem was the slow drying process of the sodium silicate solution and the slow absorption by the hazelnut shells. During the hardening phase, the adhesive was deposited on the bottom side of the samples making them non-uniform. Therefore, an experimental campaign was performed with the aim to produce uniform samples required to secure a correct evaluation of the composites' properties, and future production of panels.

## 4.2.2 Materials

### 4.2.2.1 Hazelnut shells

The hazelnut shells were provided by Raccolti di Cin, Baldissero d'Alba (CN), Italy (Figure 4.2-1a). Practical tests (not detailed for the sake of brevity) were carried out to evaluate the most suitable grain size to produce samples. The use of the hazelnut shells as they were provided, did not allow cohesion between the aggregate and the binder. Hence, they were shredded by using a mechanical mill to have grain sizes mainly between 4 mm and 8 mm (Figure 4.2-1b), similar to a previous study [23].

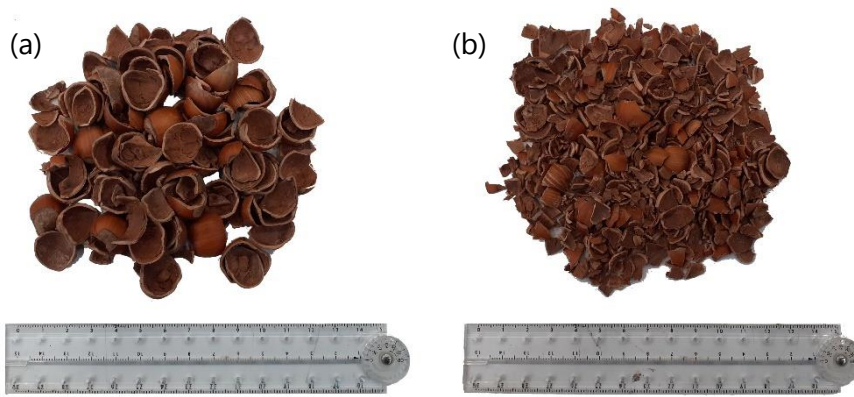


Figure 4.2-1. Hazelnut shells used as aggregates: a) before shredding; b) after shredding.

Hazelnut shells were dried at  $T = 60\text{ }^{\circ}\text{C}$  until constant mass (change in mass after 24 hours less than 0.1%) and characterized according to the recommendation of RILEM Technical Committee 236-BBM “Bio-aggregate-based Building Materials” [24] and past work [23]. The initial water content was  $(6.5 \pm 0.2)\%$ ; the loose bulk density was  $(469.3 \pm 5.8)\text{ kg/m}^3$ ; the particle size distribution is reported in Figure 4.2-2.

### 4.2.2.2 Sodium silicate solution and sodium bicarbonate

The sodium silicate solution was provided by Ingessil, Montorio (VR), Italy, which also carried out the chemical analysis. Table 4.2-1 reports the characteristics of the sodium silicate solution.

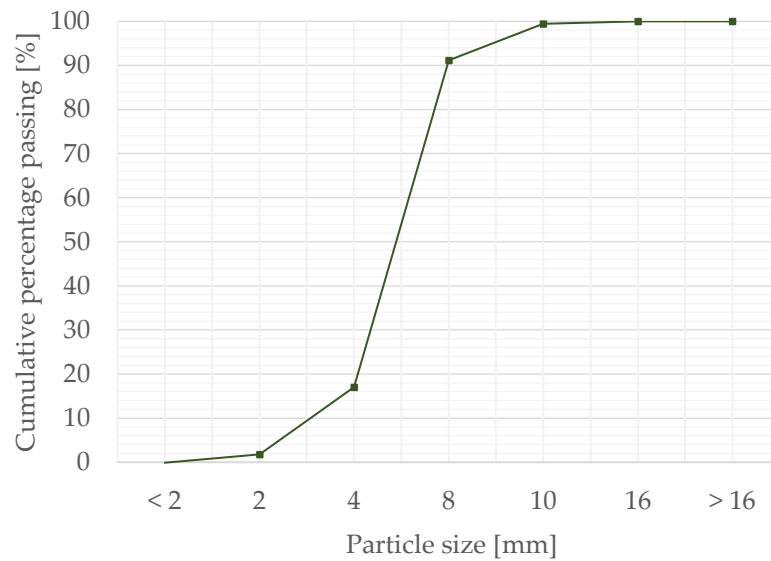


Figure 4.2-2. Particle size distribution of the shredded hazelnut shells.

Table 4.2-1. Characteristic of the sodium silicate solution provided by Ingessil [25].

| Property                              | Value |
|---------------------------------------|-------|
| Weight ratio                          | 2.4   |
| Density [°Bé]                         | 46.45 |
| Molar ratio                           | 2.48  |
| Sodium silicate concentration [% p/p] | 41.33 |
| SiO <sub>2</sub> [% p/p]              | 29.17 |
| Na <sub>2</sub> O [% p/p]             | 12.16 |
| Density [g/ml] at T=20°C              | 1.471 |
| pH (T = 20 °C)                        | 12.40 |

The sodium bicarbonate, NaHCO<sub>3</sub>, was produced by Crastan S.p.A., Pontedera (PI), Italy [26]. This is a commonly used sodium bicarbonate. Its use will be justified and detailed in Section 4.2.3.

### 4.2.3 Production parameters

As previously described, the mix design was selected trying to maximize the content of hazelnut shells and minimize the sodium silicate solution. The selected ratio was 70% of aggregates and 30% of adhesive (by total volume). Lower quantities of adhesive did not guarantee mechanical resistance.

The first samples were produced by mechanically mixing the hazelnut shells and the sodium silicate solution. Then, the mixture was put into moulds and left air-drying. As Figure 4.2-3 reports, different moulds were tested: standaradized prismatic metallic (4 cm x 4 cm x 16 cm), quadrangular wooden home-made, and silicone ones (both with 10 cm x 10 cm x 4 cm high). The aim was to avoid the sample's bonding to the moulds.





Figure 4.2-3. The considered moulds to produce the samples: metal, wooden and silicone.

The bonding of the samples was easily avoided by using the silicone mould. As for the other moulds, materials that guarantee the remotion of the samples were needed (e.g., resins or anti-glueing materials). The benefit of the wooden and the metal moulds was the possibility of opening them, demoulding the sample with-out moving it and leaving its sides drying. As a result, the silicone mould and the wooden mould covered by baking paper were selected as the best solutions.

Besides the type of mould, the uniformity of the samples was investigated. It is required for a correct evaluation of the samples' performance. Since the sodium silicate solution was not rapidly absorbed by the hazelnut shells, depositing on the bottom side of the samples, the hardening process had to be accelerated.

Different production parameters were investigated, such as drying at temperatures between  $T = 60\text{ }^{\circ}\text{C}$  and  $T = 80\text{ }^{\circ}\text{C}$  and the addition of sodium bicarbonate ( $\text{NaHCO}_3$ ) as solid reactive. The mixtures of hazelnut shells and sodium silicate solution (and eventually sodium bicarbonate) were placed in the silicone moulds (8.8 cm x 5 cm x 2.5 cm high) and 12 different production parameters were considered and compared (A to L). They are reported in Table 4.2-2, with the composites' designation. The samples were demoulded after 3 days.

The visual analysis during drying allowed defining the most efficient production parameters. The final uniformity, hence, the distribution of the sodium silicate solution, the resistance and the crumbliness of the composites were evaluated. The production parameters that guarantee to have the most uniform samples were selected for future composites' production.

Table 4.2-2. Sample's designation and description of the production parameters.

| Composites' designation | Production parameters                       | Description                                                                                                                                                                                                                                |
|-------------------------|---------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| A                       | Reference                                   | Drying at laboratory conditions without moving the sample                                                                                                                                                                                  |
| B                       | Rotation                                    | Drying at laboratory conditions by rotating the sample each 3 h                                                                                                                                                                            |
| C                       | Temperature                                 | Drying at $T = 60\text{ }^{\circ}\text{C}$ for 2 h                                                                                                                                                                                         |
| D                       | Addition of reactive                        | Addition of 25% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution); drying at laboratory conditions without moving the sample                                                                                                       |
| E                       | Addition of reactive, rotation              | Addition of 25% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution); drying at laboratory conditions by rotating the sample each 3 h                                                                                                 |
| F                       | Addition of reactive, temperature, rotation | Addition of 25% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution); drying at $T = 60\text{ }^{\circ}\text{C}$ for 1 h by rotating the sample every 30 min                                                                          |
| G                       | Temperature, rotation                       | Drying at $T = 80\text{ }^{\circ}\text{C}$ for 1.5 h, by rotating the sample each 30 min                                                                                                                                                   |
| H                       | Temperature                                 | Drying at $T = 80\text{ }^{\circ}\text{C}$ for 1.5 h, without moving the sample                                                                                                                                                            |
| I                       | Addition of reactive, temperature           | Addition of 10% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution), drying at $T = 60\text{ }^{\circ}\text{C}$ for 1 h                                                                                                              |
| J                       | Addition of reactive, temperature           | Addition of 2% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution), drying dry at $T = 60\text{ }^{\circ}\text{C}$ for 1 h                                                                                                           |
| K                       | Addition of reactive, temperature, rotation | Addition of 2% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution), drying at $T = 60\text{ }^{\circ}\text{C}$ for 30 min by rotating the sample every 15 min; then, drying at laboratory conditions by rotating the sample each 1 h |
| L                       | Addition of reactive, rotation              | Addition of 2% of $\text{NaHCO}_3$ (by vol. of sodium silicate solution), drying at laboratory conditions by rotating the sample each 1 h                                                                                                  |

## 4.2.4 Results and discussion

Figure 4.2-4 shows the samples of each composite after demoulding.

Table 4.2-3 described the results of the visual analysis and the considerations during the drying process, after three days from the production and after demoulding.



Figure 4.2-4. Representative composite samples produced by using different production parameters after demoulding, from A to L.

As shown in Figure 4.2-4 and considering the descriptions reported in Table 4.2-3, the more relevant production parameters seemed to be the temperature and the rotation. The addition of sodium bicarbonate did not improve the hardening process. Furthermore, the sodium bicarbonate could determine a lower bonding capacity (e.g., sample E). This was in line with the results achieved by Lee and Thole [18]. These researchers analysed the properties of the sodium silicate used as a binder for particleboard production, modified by several additives, including sodium bicarbonate. The researchers concluded that the addition of sodium bicarbonate decreased the bonding performance.

The most uniform samples were C and G, hence a combination of the two production parameters was selected. To secure a lower environmental impact, the lower temperature,  $T = 60\text{ }^{\circ}\text{C}$ , was chosen. The rotation of the sample was considered every 30 min for 3 h instead of only 2 h. These production parameters accelerated the hardening process of the sodium silicate, guaranteeing its distribution throughout the sample.

Thus, the production process was defined. First of all, the bio-aggregates and the sodium silicate solution were mechanically mixed for 10 min, until homogeneity. Then, the mixture was placed without compaction in silicone moulds or wooden moulds covered by baking paper. The mixture was levelled by using a spatula and the moulds were closed by a wooden top. After that, the samples were dried at  $T = 60\text{ }^{\circ}\text{C}$  for 3 h, being rotated every 30 min. Finally, they were dried at laboratory conditions, rotated every 30 min and demoulded after 2 days.

Figure 4.2-5 shows the production of two samples. Both wooden and silicone moulds are reported.

Table 4.2-3. Results of the composite samples visual analysis.

| Composite | Visual analysis                                                                                                                                                                                                                                                                                                                                                                                               |
|-----------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| A         | The sample was not uniform, and it was not dried after 3 days; the sodium silicate solution was mainly on the bottom side; six days were required to demould                                                                                                                                                                                                                                                  |
| B         | The sample was quite uniform; in some parts, the sodium silicate solution was not completely uniformly distributed; overall, it was throughout the sample; demoulding was possible after 3 days; the sample was dried after 6 days                                                                                                                                                                            |
| C         | The sample was quite uniform; in a few parts, the silicate was not completely uniformly distributed (less than for sample B), but overall, it was throughout the sample; 3 days were required to demould; the sample was dried after 6 days                                                                                                                                                                   |
| D         | The sample seems to be uniform, even if the sodium silicate solution was not perfectly distributed; since after 2 days the sample was dried, the $\text{NaHCO}_3$ accelerated the drying process; however, after 6 days, the sample started to release the $\text{NaHCO}_3$ , probably added in too high quantities; the $\text{NaHCO}_3$ was strongly visible on the surface of the sample as a white powder |
| E         | It was broken after 1 day; probably the $\text{NaHCO}_3$ absorbed the sodium silicate solution decreasing its bonding performance; the $\text{NaHCO}_3$ was visible on the surface of the sample as a white powder                                                                                                                                                                                            |
| F         | After 1 h the sodium silicate seemed to be completely dried but the quantities of the $\text{NaHCO}_3$ were too high; the sample did not seem resistant and it was highly crumbly; the $\text{NaHCO}_3$ was visible on the surface as a white powder                                                                                                                                                          |
| G         | The sample was quite uniform; the sodium silicate solution was not completely uniformly distributed, but overall, it was throughout the sample; demoulding was possible after 3 days; after 6 days the sample was dried; it seemed similar to sample C                                                                                                                                                        |
| H         | The sample was not uniform; the sodium silicate solution was deposited on the bottom surface, as happened for sample A; when the drying process finished, it was similar to sample C, even if less uniform                                                                                                                                                                                                    |
| I         | The sample did not seem uniform at the end of the drying process and after demoulding; the sodium silicate solution was on the bottom side of the sample; the $\text{NaHCO}_3$ was highly visible on the surface of the sample as a white powder                                                                                                                                                              |
| J         | The sample was not uniform; the sodium silicate solution was on the bottom part of the sample; it seemed similar to sample I, but more resistant (probably due to the fewer quantities of $\text{NaHCO}_3$ ); due to the less employed quantities, the $\text{NaHCO}_3$ was less visible on the surface of the sample                                                                                         |
| K         | The sample was not completely uniform after 3 days; however, the sodium silicate solution was throughout the sample and quite distributed; it seemed similar to sample C; due to the less employed quantities, the $\text{NaHCO}_3$ was less visible on the surface of the sample                                                                                                                             |
| L         | The sample was not uniform; in many parts of the bottom side, there were high quantities of the sodium silicate solution; demoulding was possible after 3 days; the sample seemed similar to C; the $\text{NaHCO}_3$ was not highly visible on the surface of the sample                                                                                                                                      |

Finally, after 28 days of curing, the samples were put at  $T = 50\text{ }^{\circ}\text{C}$  until reaching a constant mass (variation in mass after 24 hours less than 0.5%) to secure a complete drying. This final procedure is defined considering past studies [4,17].

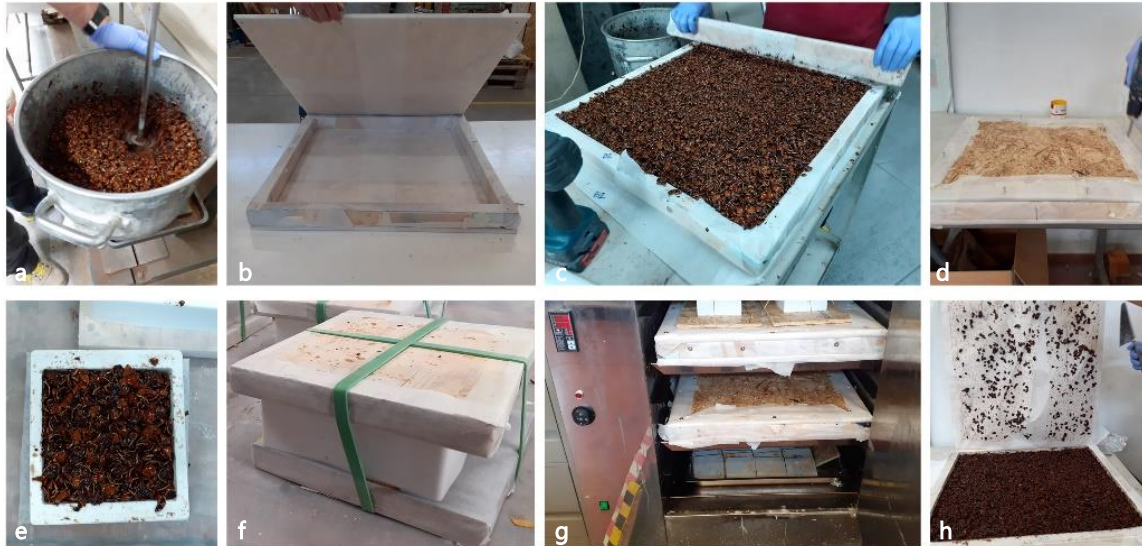


Figure 4.2-5. Production process: a) mechanical mixing; b) wooden mould; c) levelling; d) closure of the wooden mould; e) silicone mould; f) closure of the silicone mould; g) drying at  $T=60^{\circ}\text{C}$ ; h) demoulding.

## 4.2.5 Conclusions

Production parameters to produce boards made up of hazelnut shells as aggregates and sodium silicate solution as adhesive were tested. The drying at different temperatures for different periods, the rotation of the samples and the addition of sodium bicarbonate as an additive were assessed. The work aimed at producing uniform samples, required for a correct evaluation of their properties and future production.

The following conclusions were achieved:

- Both drying with thermal treatment ( $T = 60\text{ }^{\circ}\text{C}$  and  $T = 80\text{ }^{\circ}\text{C}$ ) and the addition of sodium bicarbonate accelerated the hardening process of the sodium silicate solution.
- High quantities of sodium bicarbonate worsened the bonding properties of the sodium silicate solution; the produced samples were more crumbly and less resistant.
- Rotating the samples before the sodium silicate solution was completely dried allowed its distribution throughout the sample and increased the final uniformity.

None of the samples achieved perfect uniformity. This is probably due to the selected materials and the manually controlled production process (the impossibility to ensure a constant rotation of the samples during drying). Nevertheless, the combination of temperature and rotation seemed to be the best production parameters.

As a result, the production process to have uniform samples was defined and tested. It can be used to produce composites for future analysis of these innovative building products.

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## 4.3 Article F - A feasible re-use of an agro-industrial by-product: Hazelnut shells as high-mass bio-aggregate in boards for indoor applications

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### Abstract

The present work investigates the feasibility of producing boards, with unconventional materials, namely hazelnut shells as a high-mass bio-aggregate and a sodium silicate solution as a no-toxic adhesive, and discusses possible applications based on an extensive characterization. The aim is to define a feasible reuse of a largely produced agro-industrial by-product to reduce the high environmental impact caused by both the construction and the agriculture sectors, by proposing a building composite that improves indoor comfort. The presented combination of aggregate-adhesive generated a product with characteristics interesting to explore. The thermal conductivity is moderated, and the composite achieved values of  $\sigma_{\max} = 0.39 \text{ N/mm}^2$  for flexural strength and  $\sigma_{\max} = 2.1 \text{ N/mm}^2$  for compressive strength, but it showed high sorption capacity with a moisture buffering value of about  $3.45 \text{ g/(m}^2 \text{ \%RH)}$ , and a peak of sound absorption between 700 - 900 Hz. Therefore, the boards' most promising performance parameters seem to be their high hygroscopicity and acoustic absorption behaviour, namely in the frequency range of the human voice. Hence, the proposed composite could improve indoor comfort if applied as an internal coating board.

**Keywords** Acoustic absorption; Granular material; Hygroscopicity; Mechanical property; Sodium silicate; Thermal property.

### 4.3.1 Introduction

Environmental problems and climate changes caused by human activities have become so serious that a radical transformation is urgently needed (Trobiani Di Canto et al., 2023; Posani et al., 2022). The construction sector is widely responsible: building materials production, use, and end-of-life cause emissions of harmful substances and energy consumption (Lisienkova et al., 2022; Hung Anh and Pásztor, 2021). Furthermore, building materials and products can guarantee a passive control of indoor conditions, lowering energy demand (Martínez-García et al., 2020) and improving internal comfort (Barreca et al., 2019; Posani et al., 2023b, 2019). Thus, current research is more and more focused on finding strategies that can improve the sustainability of the construction sector, such as

the encouragement of circular economy practices, the use of more advanced fabrication methodologies, and the development of passive building techniques (Nistratov et al., 2022; Sambucci and Valente, 2021). Moreover, employing local materials to produce eco-friendly building composites derived from natural products is a widely investigated possibility (Donini et al., 2022).

Together with the construction sector, agriculture, paramount to feeding populations, may have a negative impact on the environment (Duque-Acevedo et al., 2022). Indeed, it is responsible for excess land use, deforestation, energy and water consumption, and the continuous production of large amounts of waste that have to be disposed of (Barbieri et al., 2013; Gaspar et al., 2020). In this scenario, the use of agro-industrial wastes as building materials seems to be an attractive possibility to explore. It may contribute to reducing energy consumption, costs, and carbon emissions derived from the production process (Binici et al., 2020; Brás et al., 2019; La Gennusa et al., 2021), as well as encouraging a circular economy (Liuzzi et al., 2020b) and moderating the problem of the disposal of agricultural wastes (de Azevedo et al., 2022).

Several studies have already addressed the feasibility of using agro-industrial wastes as building materials, mainly considering the use of fibres. For example, Sam-Brew and Smith (2015) considered hemp and flax fibres as reinforcement for particleboard production; Ali and Alabdulkarem (2017) evaluated the thermal and mechanical properties of a composite made up of date palm fibres; Maraldi et al. (2018, 2016) analysed the mechanical behaviour of straw bales; Akinyemi and Dai (2020) and Nunes et al. (2021) employed banana fibres in particleboards. Some studies considered also other types of agro-industrial by-products, such as corn cob (Ramos et al., 2021; Viel et al., 2019), cork granules (La Rosa et al., 2014; Liu et al., 2017), olive stones (del Río Merino et al., 2017) and pruning waste (Martellotta et al., 2018). All these residues have many advantages, being recyclable, biodegradable, cheap, and with low environmental impact (Liuzzi et al., 2018; Vandenbossche et al., 2012). In addition, they provide promising properties when used to produce building composites, such as good thermal insulation behaviour, high sound-absorption performance, no harmful effects on human health, and large availability (Bardage, 2017; Mati-Baouche et al., 2016). Finally, bio-wastes-based composites can passively control indoor conditions (Cascione et al., 2022; La Gennusa et al., 2021; Laborel-Préneron et al., 2017), both improving indoor comfort and lowering energy demands (Posani et al., 2023a; Viel et al., 2017).

On the other side, agro-industrial-based composites also present some drawbacks, namely low resistance to water and fire, and high biological susceptibility (Palumbo et al., 2017). This latter is caused by several factors, such as the organic composition, the presence of nutrients, and the hygroscopic nature of bio-wastes (Ginestet et al., 2020; Tobon et al., 2020). The low resistance to biological attack is a great drawback of bio-based materials because it can modify their properties, restrict their performance, worsen indoor air quality, and cause human diseases (Brambilla and Sangiorgio, 2020; Stefanowski et al., 2017). It can be avoided and moderated by specific treatments

(e.g., heat or chemical treatments), by using materials that increase the biological resistance (e.g., lime (Krejsová and Doleželová, 2019), sodium benzoate (de Carvalho et al., 2020), boric acid (Palumbo et al., 2017) and citric acid (Treu et al., 2020)), by reducing of thermal bridges, increasing air changes or ventilation rates, and controlling relative humidity (Brambilla and Sangiorgio, 2020). Even if many studies about bio-based composites did not evaluate this property (Santos et al., 2017), its assessment is important for their production and durability.

Both the origin of the agro-industrial wastes and their availability are important (Cintura et al., 2021). The employment of local materials lowers the environmental impact caused by materials' transportation. As for the availability, the seasonality of agro-industrial wastes could be an obstacle to large-scale production. Cintura et al. (2021) collected many past studies that employed agro-industrial wastes as building materials, pointing out some literature gaps, namely some by-products not already analysed that could be promising possibilities. Based on this knowledge, the present work assessed the feasibility of using not already widely investigated bio-wastes to produce a building composite.

Among the building products, particleboards (boards made of particles bounded by adhesives and used for internal and external applications (Gürü et al., 2015)) present several benefits. They can be produced by using wood chips, fibres, and agro-industrial wastes (Monteiro et al., 2020). They are economically advantageous and easily assembled and employed for different uses (Mantanis et al., 2018).

Past research already addressed the feasibility of producing bio-waste-based boards and panels, analysing different properties (e.g., hygrothermal, acoustic, and mechanical properties). For example, Ricciardi et al. (2021) evaluated the thermal performance and the Life Cycle Assessment (LCA) of cork, rice husk, and coffee chaff-based insulation panels. The aggregates were mixed and bounded by a cold-water-based polyurethane glue and high quantities of cork improved the thermal performance of the composites, while rice husk and coffee chaff secured low environmental impact. They determined the best compositions of aggregates to achieve a good compromise on thermal insulation and environmental impact. Luizzi et al. (2020b) considered the hygrothermal and acoustic properties of almond skin waste-based panels, using polyvinyl acetate aqueous solution (PVA) and a biodegradable gum Arabic aqueous solution as binders. They reported thermal conductivity of less than 0.1 W/(m·K) and an acoustic absorption that outperformed typical thick, porous materials. Auriga et al. (2022) produced particleboards by replacing wood particles with hemp shivs in the face and core layers. They demonstrated that the replacement in the core layer improved mechanical performance, moisture resistance, and dimensional stability.

As past research showed, several possibilities can be investigated to produce boards made up of agro-industrial wastes: by using fibres or other parts of the by-products (e.g., straw, grains, piths, husk, shiv, stones, skins), by changing the considered adhesives (binders, resins, glues) and the

production process (using heat treatments, hot-pressing, spraying the adhesives), by using additives to improve properties and/or moderate the drawbacks, as well as by assessing different final applications (Borysiuk et al., 2019; Monteiro et al., 2019).

The present work considers this literature background and presents experimental studies to produce boards for indoor applications by employing unconventional materials: hazelnut shells as bio-aggregate and a sodium silicate solution as the adhesive.

Hazelnut (*Corylus avellana* L.) shell was considered due to their large availability in the selected area for the present work (Cintura et al., 2021). Figure 4.3-1 reported the hazelnut world production trend between 2016 and 2021, considering only the first four world producers, according to FAO (FAO, 2021).

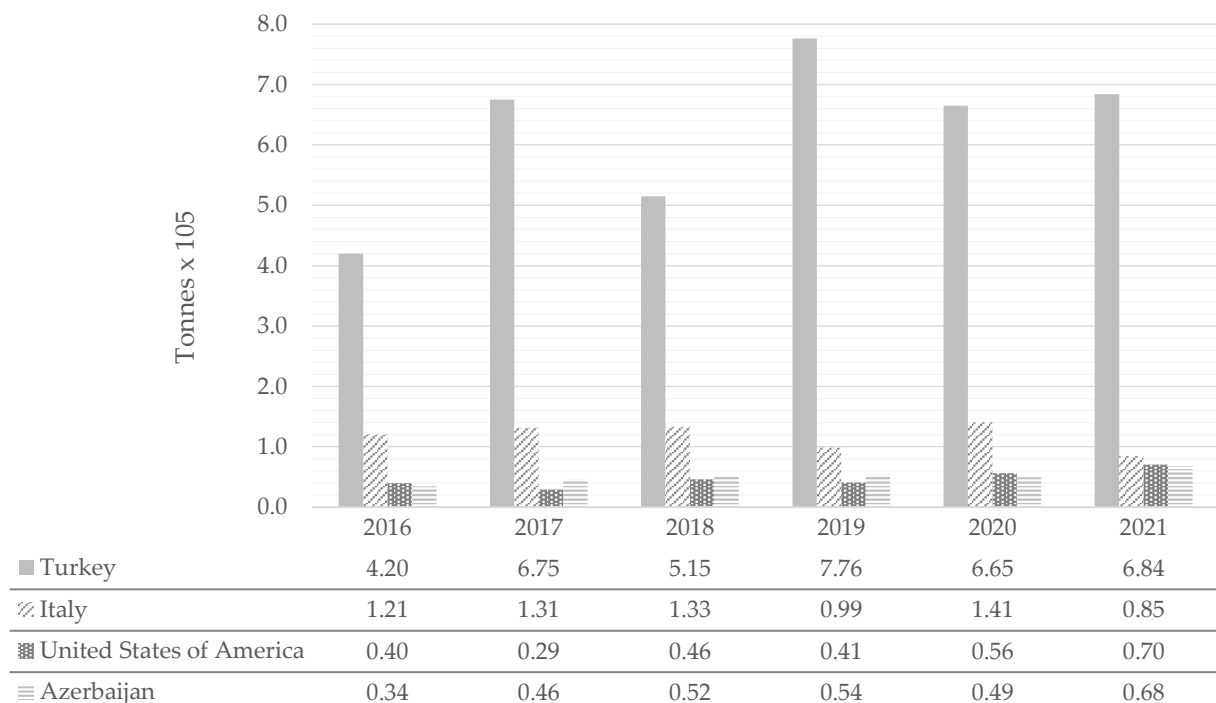


Figure 4.3-1. Hazelnut's world production trend between 2016 and 2021 (FAO, 2021).

Turkey has been the first producer during the last six years, with much higher production, followed by Italy as the second and the United States of America as the third, except for 2017, 2018, and 2019 where Azerbaijan produced greater quantities. Hazelnuts are normally harvested in August and September and left to dry for some months. The drying improves their chemical and physical stability and the resistance to mould growth (Özilgen and Özdemir, 2001; Vrtodušić et al., 2022). After drying, the hazelnuts are stored for up to 48 months. Finally, they are provided to the industry usually without shells (Markuszewski et al., 2022; Özilgen and Özdemir, 2001). Since this process requires more than two years after harvesting, there is a huge quantity of hazelnut shells available all over the year. This information, collected by the literature review and oral information of a producer, suggested

selecting hazelnut shells as aggregate as a widely available agro-industrial waste. Table 4.3-1 reports some information about the chemical and physical properties of hazelnut shells, according to past studies.

Table 4.3-1. Chemical and physical properties of hazelnut shells.

| Chemical composition [%] |                |        |      | Physical Properties                     |                                | Reference                         |
|--------------------------|----------------|--------|------|-----------------------------------------|--------------------------------|-----------------------------------|
| Cellulose                | Hemicelluloses | Lignin | Ash  | Loose bulk density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] |                                   |
| 22.90                    | 23.50          | -      | 1.40 | -                                       | -                              | (Licursi et al., 2017)            |
| 28.90                    | 11.30          | 30.20  | 4.00 | -                                       | -                              | (Lopes et al., 2012)              |
| -                        | -              | -      | -    | -                                       | 0.1                            | (Çuhadaroglu, 2005)               |
| -                        | -              | -      | -    | 550.50 ± 19.53                          | 0.107 ± 0.003                  | (Cintura et al., 2022)            |
| 34                       | 20             | 35-41  | -    | -                                       | -                              | (Salasinska and Ryszkowska, 2012) |

The possibility of using hazelnut shells for particleboards has been already addressed in some past studies. Lopes et al. (2012) considered the replacement of wood particles with hazelnut shells in the core and face layers of particleboards produced by using a urea-formaldehyde resin. The researchers reported that no significant differences were detected in internal bonding, density, and thickness swelling. Hence, this by-product might be used instead of wood chips, guaranteeing more sustainable building products and circular economy practices. Çöpür et al. (2008) demonstrated the feasibility of using hazelnut shells and husks in medium density fibreboards as a mixture with wood fibres. Although the addition of hazelnut husk and shell reduced the internal bond strength of the boards, this possibility could reduce the use of raw material. Barbu et al. (2020) considered hazelnut shells with melamine-urea formaldehyde to produce hot-pressed boards, demonstrating that the dimensional stability and hardness were higher than spruce-based particleboards. However, although past research considered the possibility of using hazelnut shells in boards' production, they have not already been widely investigated as such.

As for the adhesive, a sodium silicate solution was chosen because of its bonding capacity, no toxicity, and high resistance to biological attack and to chemical decomposition (Lee and Thole, 2018; Liuzzi et al., 2020a). Despite the several benefits, sodium silicate is not completely non-environmentally impactful, it has low moisture resistance, and it shows low bonding strength in wood-based composites (Lee and Thole, 2018; Song et al., 2021). Several past studies addressed the production of low-density wood-based boards by using sodium silicate as an adhesive; however, only a few of them detailed the production process (Lee and Thole, 2018). In the present study, this latter is defined

according to Cintura et al. (2023b). The idea was to avoid harmful products, high temperatures, and pressing in the production phase, and to cast and cure the composites at environmental conditions.

Thermal capacity, hygroscopicity, acoustic, and mechanical properties of the hazelnut shells-based composite were evaluated and compared with literature values and commercial products. As an outcome, a general characterization of the produced boards was provided to define possible suitable applications. The aim is to demonstrate the feasibility of using the selected materials to produce indoor particleboards that can improve indoor comfort, lowering the environmental impact derived from both the construction and agriculture sectors.

## 4.3.2 Materials and methods

### 4.3.2.1 Materials

#### 4.3.2.1.1 Bio-aggregate

Hazelnut (*Corylus avellana* L.) shells, provided by Raccolti di Cin, Baldissero d'Alba (CN), Italy, were used as bio-aggregate since they are widely available in the area where analyses were conducted (Cintura et al., 2021). Several practical tests were carried out to determine the most suitable grain size to produce samples. The results demonstrated that hazelnut shells cannot be used as they were obtained directly from the industry (Figure 4.3-2a), as their grain size and shape did not allow cohesion between the aggregate and the adhesive (Cintura et al., 2023b). Hence, they were shredded to grain sizes mainly between 4 mm and 8 mm (Figure 4.3-2b), as reported in a previous study (Cintura et al., 2023b). Figures 4.3-2c and 4.3-2d show the particle size distribution and the percentages of aggregates retained in each sieve, respectively, considering the average values of three samples.

The hazelnut shells were dried at  $T = 60\text{ }^{\circ}\text{C}$  and characterized according to the recommendation of the RILEM Technical Committee 236-BBM 'Bio-aggregate-based building Materials' (Amziane et al., 2017). All the tests were repeated for three different samples and the average values were considered. As reported in a past study (Cintura et al., 2023b) the initial water content was  $(6.5 \pm 0.2)\%$ ; the loose bulk density was  $(469.3 \pm 5.8)\text{ kg/m}^3$ . With the particle size distribution of Figures 4.3-2c and 4.3-2d, the thermal conductivity was evaluated by the heat flow method (better described below) by placing the dried material in a wooden mould  $50\text{ cm} \times 50\text{ cm} \times 4\text{ cm}$ . The thermal conductivity value was  $(0.099 \pm 0.006)\text{ W/(m.K)}$ .

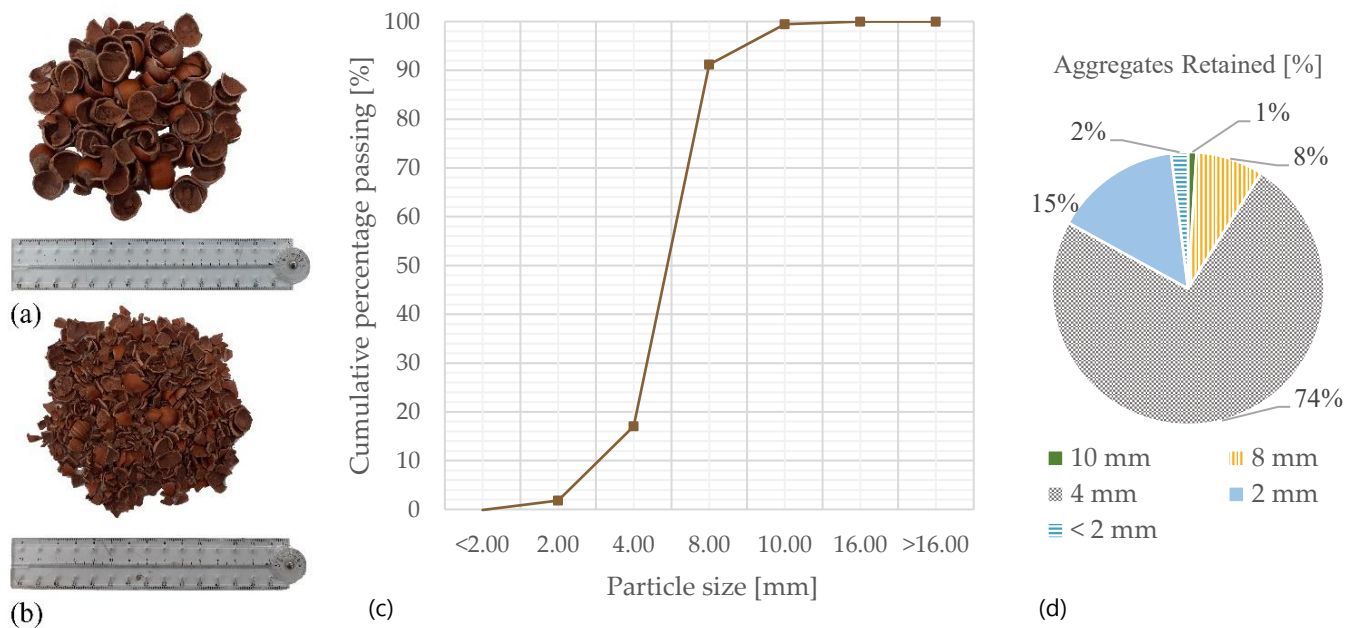


Figure 4.3-2. Samples of hazelnut shells (a) before shredding, (b) after shredding, used to carry out particle analysis, (c) particle size distribution, adapted from (Cintura et al., 2023b), and (d) percentages of aggregates retained in each sieve (considering the average values of three samples).

#### 4.3.2.1.2 Adhesive

A sodium silicate solution,  $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ , provided by Ingessil S.r.l., Montorio (VR), Italy, and having the characteristic reported in Table 4.3-2, was employed as adhesive, considering the promising results of past studies (Cintura et al., 2023a; Liuzzi et al., 2020a) and its several benefits, such as its bonding capacity, no toxicity, and high resistance to biological attack and to chemical decomposition (as reported in Section 4.3.1).

The sodium silicate solution was used as it was obtained directly from the industry, hence with the properties reported in Table 4.3-2, without adding any chemicals.

Table 4.3-2. Characteristic of the sodium silicate solution provided by Ingessil Srl (Ingessil, 2022).

| Property                              | Value |
|---------------------------------------|-------|
| Weight ratio [-]                      | 2.40  |
| Density [°Bè]                         | 46.45 |
| Molar ratio [-]                       | 2.48  |
| Sodium silicate concentration [% p/p] | 41.33 |
| $\text{SiO}_2$ [% p/p]                | 29.17 |
| $\text{Na}_2\text{O}$ [% p/p]         | 12.16 |
| Density [g/ml] at T = 20 °C           | 1.471 |
| pH (T = 20 °C)                        | 12.40 |

#### 4.3.2.2 Samples production

The mix design was selected after several tests, not detailed for the sake of brevity. To maximize the content of aggregates, minimize the amount of adhesive, and secure sufficient mechanical resistance, the selected ratio was 70% hazelnut shells and 30% sodium silicate solution (by total volume). The studies carried out to define the best production process were detailed in a previous study (Cintura et al., 2023b). The hazelnut shells and the sodium silicate solution were mechanically mixed for 10 minutes. The fresh composite was placed in quadrangular silicon (10 cm x 10 cm x 4 cm), cylindrical silicon (diameter = 10 cm, high = 4 cm), and quadrangular wooden home-made (50 cm x 50 cm x 4 cm) moulds, and levelled by using a spatula, to obtain a horizontal surface, and the moulds were closed by a wooden top. The samples were dried at  $T = 60\text{ }^{\circ}\text{C}$  for 3 h, being rotated every 30 minutes. After that, they were dried at laboratory conditions, rotated every 30 minutes (for half the day) to secure the homogeneity, and demoulded after 2 days. After 28 days of curing, they were put at  $T = 50\text{ }^{\circ}\text{C}$  until reaching a constant mass (variation in mass after 24 h less than 0.5%) and a complete drying (Liuzzi et al., 2020a, 2020b). Figure 4.3-3 shows the samples produced by using silicon moulds.

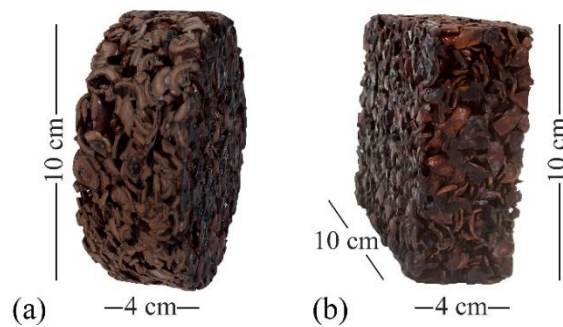


Figure 4.3-3. Samples after casting and curing, to evaluate: (a) acoustic properties (diameter = 10 cm; high = 4 cm); (b) hygroscopicity (10 cm x 10 cm x 4 cm).

#### 4.3.2.3 Test methods

Table 4.3-3 reports the analysed properties, the designation, size, and number of samples, and the considered standards and references to carry out the laboratory tests, better described in the following text.

##### 4.3.2.3.1 Visual observation

The samples were constantly monitored during the drying and curing times. As reported by Cintura et al. (2023b) the sodium silicate solution was not rapidly absorbed by the aggregate, and it could be deposited on the bottom part of the samples, resulting in non-uniform samples, required for a correct evaluation of the composites' properties. The visual analysis provided a rapid check of the correct manufacture of the samples.



Table 4.3-3. Laboratory tests, designation, number and dimensions of composite samples, and references.

| Properties           | Samples                                                                        |        |                           | References                                |
|----------------------|--------------------------------------------------------------------------------|--------|---------------------------|-------------------------------------------|
|                      | Designation*                                                                   | Number | Size [cm]                 |                                           |
| Visual observation   | -                                                                              | All    | Varied                    | (Cintura et al., 2023b)                   |
| Apparent density     | H1, H2, H3                                                                     | 3      | 10 x 10 x 4               | (EN 323, 1993)                            |
| Thermal conductivity | H7, H8, H9                                                                     | 3      | 50 x 50 x 4               | (Baldinelli et al., 2019; EN 12667, 2001) |
| Hygroscopicity       | H1, H2, H3                                                                     | 3      | 10 x 10 x 4               | (ISO 24353, 2008)                         |
| Sound absorption     | H4, H5, H6                                                                     | 3      | diameter = 10<br>high = 4 | (ISO 10534-2, 2001)                       |
| Flexural strength    | H8_01, H8_02, H8_03, H8_04, H9_01, H9_02, H9_03, H9_04                         | 8      | 50 x 10 x 4               | (ASTM D7264-21, 2021; UNI-11842, 2021)    |
| Compressive strength | H8_01A, H8_01B, H8_02A, H8_02B, H8_03A, H9_01A, H9_01B, H9_02A, H9_02B, H9_03A | 10     | 10 x 10 x 4               | (EN-826, 2013)                            |

\* Note: Samples H8 and H9, for thermal conductivity, were cut in strips to evaluate the flexural strength. Then, the non-damaged parts of the samples employed for the bending test were considered to evaluate compressive strength. This information was highlighted by the designations, which referred to the names of the original samples.

#### 4.3.2.3.2 Apparent density and thermal properties

The apparent density, which considers the volume including both the solid and the intra-particle voids (Ratsimbazafy et al., 2021), was calculated after the curing process and drying at  $T = 50\text{ }^{\circ}\text{C}$ , as the average ratio between mass and volume, considering an adaptation of EN 323 (EN 323, 1993). Three quadrangular 10 cm x 10 cm x 4 cm samples (H1, H2, H3) were weighted by using an electronic balance Kern EW 2200-2NM. The dimensions were verified by using a Fervi Digital Caliber (Figure 4.3-4a).

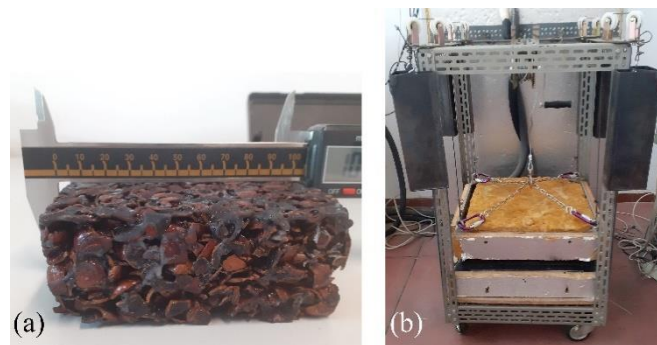


Figure 4.3-4. (a) Sample 10 cm x 10 cm x 4 cm and the Fervi Digital Caliber used to determine the dimensions for apparent density; (b) equipment used to evaluate the thermal conductivity by considering the heat flow method.

The thermal properties can be determined by the steady-state methods (e.g., guarded hot plate, heat flow meter, and guarded hot boxes) and the transient methods (e.g., hot wire and line source) (Collet, 2017; Posani et al., 2022). The first one provides a more accurate measurement and considers the thermal performance of materials in equilibrium conditions (constant temperature throughout the

samples), but requires more expensive equipment, larger samples, and more time (Yüksel, 2016). This method, described in EN 12667 (EN 12667, 2001), consists of registering the heat flow throughout the sample, placing it between two different plates at known constant temperatures (heated and cooled), and determining the thermal conductivity, namely the materials' effectiveness in conducting heat, by considering one-dimensional integral form of the Fourier equation (Al-Homoud, 2005; Baldinelli et al., 2019; La Gennusa et al., 2021; Platt et al., 2023). Figure 4.3-4b reports the employed equipment, described, and validated in a past study (Baldinelli et al., 2019). The average value and the standard deviation of three samples (H7, H8, H9) 50 cm x 50 cm x 4 cm, stabilized at  $T = (20 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (60 \pm 5) \%$ , were considered. They were weighted before and after the test to control the possible change in mass caused by the variation of the relative humidity, as this is an important parameter to be considered in thermal conductivity measurements (Collet, 2017; Nguyen et al., 2020).

#### 4.3.2.3.3 Hygroscopicity and moisture buffering value

The hygroscopicity, namely the capacity to adsorb and release moisture when relative humidity increases and decreases (Ranesi et al., 2022), and the moisture buffering value (MBV) are usually evaluated by considering the variation in mass over time due to exposure to high relative humidity. In the present work, the standardized method ISO 24353 (ISO 24353, 2008) was considered, employing three samples 10 cm x 10 cm x 4 cm (H1, H2, H3). A moisture barrier was provided by using an aluminium foil (Figure 4.3-5a) that allowed the exposure of only one adsorption/desorption face (area =  $0.1 \text{ m}^2$ ), as reported in Figure 4.3-5b. The samples were preconditioned in a climatic chamber (Climatest ARGOLab CH 250), shown in Figure 4.3-5c, at  $T = (23 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (63 \pm 5) \%$  until reaching a constant mass (change in mass of less than or equal to 0.5% over 24 h).

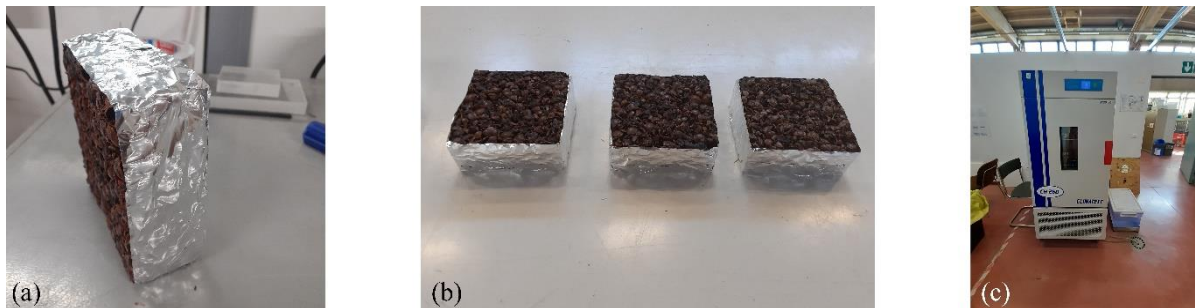


Figure 4.3-5. (a) Detail of the aluminium foil used to provide a moisture barrier and expose only one surface of the sample to relative humidity variations; (b) three samples (10 cm x 10 cm x 4 cm) H1, H2, and H3, covered by the aluminium foil and considered to evaluate the hygroscopic properties; (c) climatic chamber (Climatest ARGOLab CH 250) employed for the hygroscopicity test.

Four cycles of 24 h were considered: a 12 h-sorption phase at  $\text{RH} = 75 \%$  and a 12 h-desorption phase at  $\text{RH} = 50 \%$ . To determine the moisture content as a function of time, the samples were weighed every 3 h; hence, considering the variation of mass and knowing the exposed surface, the sorption/desorption capacity was determined. Moisture adsorption content,  $\rho_{A,ac}$ , moisture

desorption content,  $\rho_{A,dc}$ , and moisture content difference,  $\rho_{A,sc}$  were calculated according to Equations 4.3-1, 4.3-2 and 4.3-3

$$\rho_{A,ac} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \frac{m_{an} - m_{d(n-1)}}{A} \quad (4.3-1)$$

$$\rho_{A,dc} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \frac{m_{an} - m_{dn}}{A} \quad (4.3-2)$$

$$\rho_{A,sc} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \rho_{A,ac} - \rho_{A,dc} \quad (4.3-3)$$

where  $m_{an}$  [g] is the value of the mass at the end of the sorption phase,  $m_{d(n-1)}$  [g] is the mass at the end of the previous desorption cycle,  $m_{dn}$  [g] is the value of the mass at the end of the desorption phase, and  $A$  [mm<sup>2</sup>] is the exposed surface.

MBV was calculated and rated considering an adaptation of Rode et al. (Rode et al., 2005). It was considered as the average value between the MBV of the sorption (MBV<sub>a</sub>) and the desorption phase (MBV<sub>d</sub>), calculated as reported in Equations 4.3-4 and 4.3-5, respectively.

$$\text{MBV}_a \left[ \frac{\text{g}}{\text{m}^2 \% \text{RH}} \right] = \frac{m_{an} - m_{d(n-1)}}{A \times (\text{RH}_{\text{high}} - \text{RH}_{\text{low}})} \quad (4.3-4)$$

$$\text{MBV}_d \left[ \frac{\text{g}}{\text{m}^2 \% \text{RH}} \right] = \frac{m_{an} - m_{dn}}{A \times (\text{RH}_{\text{high}} - \text{RH}_{\text{low}})} \quad (4.3-5)$$

$\text{RH}_{\text{high}}$  and  $\text{RH}_{\text{low}}$  were the highest and the lowest values, respectively, namely 75 % and 50 %, according to ISO 24353 (ISO 24353, 2008). The values of the last three cycles were considered to use the most stabilized conditions.

#### 4.3.2.3.4 Acoustic properties

Acoustic properties were evaluated by using the impedance tube method (direct measurement) according to ISO 10534-2 (ISO 10534-2, 2001). The method provides a fast evaluation of the acoustic impedance and absorption properties of small-size samples, easily placed, and displaced into the equipment. When placing the samples at the extremity of the impedance tube (or Kundt's tube), an acoustic excitation is generated, whose pressure is measured by microphones. Knowing the ratio between the sound power that enters the surface of the tested sample and the incident sound power for a plane wave at normal incidence, the sound absorption coefficient at normal incidence,  $\alpha$ , and the normal incidence could be determined by some corrective formulas (Othmani et al., 2017; ISO 10534-2, 2001).

The cylindrical samples H4, H5, and H6 (diameter = 10 cm, high = 4 cm) were conditioned at  $T = (20 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (60 \pm 5) \%$ , smooth (Figure 4.3-6a) to be placed into a Plexiglas impedance tube with an interior diameter of 10 cm and an operating frequency range between 100 Hz and 2000 Hz (Figure 4.3-6b), and sealed to reduce the possible gaps between the tube and the samples. A loudspeaker and a signal amplifier (Samson 120A with SNR = 96 dB) were used to generate an exponential sine sweep (Corredor-Bedoya et al., 2021), converted to an analogue signal by the Digital

to Analog Converters (DAC) of the soundcard (RME 802). A single microphone 1/2" PCB recorded the signal and the Analog-to-Digital Converter (with a sample rate of 44.1 kHz at 24-bit depth) converted it into digital audio, which was processed by ITA-Toolbox (Berzborn et al., 2017) through Transfer-Function Method (Chung and Blaser, 1980a, 1980b). Since the production process with the selected materials could result in non-perfectly homogeneous samples, the acoustic test was undertaken for both the front and back sides of each sample (distinguished by different designations, namely the letters A and B).

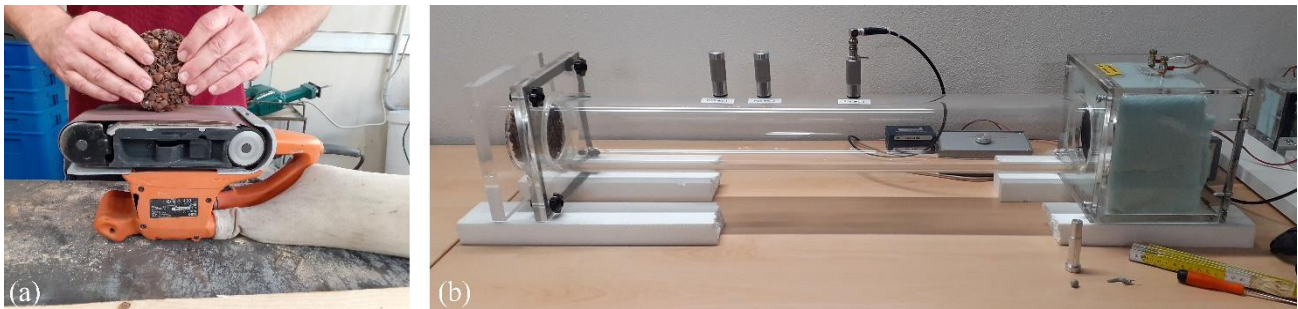


Figure 4.3-6. (a) Smoothing process of a cylindrical sample to fit into the end of the impedance tube; (b) impedance tube of plexiglass (diameter = 10 cm) with the loudspeaker and the sample at the extremities.

#### 4.3.2.3.5 Flexural strength

The two samples 50 cm x 50 cm x 4 cm employed for the thermal analysis (H8, and H9) were cut in strips each 10 cm, and the eight obtained samples 50 cm x 10 cm x 4 cm were conditioned at  $T = (20 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (60 \pm 5) \%$ . The considered designation, reported in Table 4.3-3 (namely H8\_01, H8\_02, H8\_03, H8\_04, H9\_01, H9\_02, H9\_03, H9\_04), wanted to mark the original samples employed.

Each strip was subjected to a four-point bending test to determine the flexural properties, adapting UNI 11842 (UNI-11842, 2021) and ASTM D7264 (ASTM D7264-21, 2021). The test was performed at laboratory conditions by using a Galdabini universal machine with a maximum capacity of 100 kN in displacement control with a velocity of 0.5 mm/min. The distance between supports was 39 cm; hence the forces were applied each 13 cm, as shown in Figure 4.3-7a.

As reported in ASTM D7264 (ASTM D7264-21, 2021), flexural properties could vary depending on which surface of the sample is compressed. Hence, the results depend on how the sample is placed (i.e., varying the considered lower/upper surface for the test). For this reason, both sides of the samples were tested, due to the possible non-homogeneous distribution of the sodium silicate solution. Half of the tested samples were placed with the upper side as the rougher one (designated as Samples\_UP); the others in the opposite way, namely the upper side as the flatter one and the lower side as the rougher one (designated as Samples\_LOW). This allowed evaluating of mechanical performance for both faces and assessing how a different sodium silicate distribution may affect the bending performance.



Figure 4.3-7. Galdabini universal machine employed to evaluate the mechanical properties: (a) sample 10 cm x 50 cm x 4 cm used to analyse the flexural strength; (b) sample 10 cm x 10 cm x 4 cm used to evaluate the compressive strength.

The force and displacement were recorded during the test, carried out until the breaking load, by a linear displacement transducer, LVDT, HBM Model WA20mm. The stress-strain curve was determined by considering Equations 4.3-6 and 4.3-7:

$$\sigma_{(intrados)} \left[ \frac{N}{mm^2} \right] = \frac{Fl}{bh^2} \quad (4.3-6)$$

$$\epsilon_{(intrados)} [-] = \frac{108}{23} \frac{h\delta}{l^2} \quad (4.3-7)$$

where  $F$  [N] is the applied force,  $l$  [mm] is the span of the sample,  $b$  [mm] and  $h$  [mm] are the width and thickness of the sample, respectively, and  $\delta$  [mm] is the displacement. The flexural strength  $\sigma_{max}$  [N/mm<sup>2</sup>] of each sample was calculated as the one corresponding to the maximum applied force,  $F_{max}$  [N], as described in Equation 4.3-8; then the average value and standard deviation were considered.

$$\sigma_{(max)} \left[ \frac{N}{mm^2} \right] = \frac{F_{max}l}{bh^2} \quad (4.3-8)$$

The modulus of elasticity was determined considering Equation 4.3-9 as reported in ASTM D7264 (ASTM D7264-21, 2021), considering 20% and 60% of  $\sigma_{max}$  ( $\sigma_{60\%}$  and  $\sigma_{20\%}$ ) and the corresponding  $\epsilon$  ( $\epsilon_{60\%}$  and  $\epsilon_{20\%}$ ) (UNI-11842, 2021).

$$E \left[ \frac{N}{mm^2} \right] = \frac{\Delta\sigma}{\Delta\epsilon} = \frac{\sigma_{60\%} - \sigma_{20\%}}{\epsilon_{60\%} - \epsilon_{20\%}} \quad (4.3-9)$$

Again, the measurement was carried out for each sample and then the average value and standard deviation were considered and reported.

#### 4.3.2.3.6 Compressive strength

Compressive strength was evaluated according to an adaptation of EN 826 (EN-826, 2013). Samples 10 cm x 10 cm x 4 cm were cut from the extremities of the samples used for flexural strength (not damaged by the bending test). In this case, for the samples' designation (reported in Table 4.3-3), H8 and H9, indicated which were the original samples (employed for thermal analysis); the numbers 01, 02, and 03 indicated the considered strips (employed for bending test); A and B distinguished the

two parts of the samples (employed for bending test) considered for the compression. A total of ten samples were tested: H8\_01A, H8\_01B, H8\_02A, H8\_02B, H8\_03A, H9\_01A, H9\_01B, H9\_02A, H9\_02B, H9\_03A.

The samples were preloaded with a force of 114.25 N, then placed between the two plates in the Galdabini machine with a maximum capacity of 100 kN (Figure 4.3-7b) and compressed until the breaking load was achieved, with a constant velocity of 2 mm/min. This latter was selected after several tests as it allowed the samples' breaking in more time, guaranteeing a better data acquisition. Compressive strength and strain were calculated by considering Equations 4.3-10 and 4.3-11.

$$\sigma \left[ \frac{N}{mm^2} \right] = \frac{F}{A_0} \quad (4.3-10)$$

$$\varepsilon [-] = \frac{\delta}{d_0} \quad (4.3-11)$$

where F [N] is the applied force (recorded by the equipment),  $A_0$  [mm<sup>2</sup>] is the initial cross-sectional area of the samples,  $\delta$  [mm] is the displacement during the test (recorded by the equipment), and  $d_0$  [mm] is the initial thickness of the samples. Compressive strength  $\sigma_{max}$  [N/mm<sup>2</sup>] was calculated for each sample by considering the breaking load and then the average value and standard deviation were determined.

The modulus of elasticity E was calculated considering Equation 4.3-9, namely  $\sigma$  and  $\varepsilon$  at 20% and 60% of the compressive test breaking load, and then the average value and the standard deviation were determined.

### 4.3.3 Results

#### 4.3.3.1 Visual observation

The constant visual observation guaranteed the production of uniform samples. The evaluation was purely qualitative and was carried out to check the correctness of the production process. At the end of the curing time, all the samples showed a sufficiently uniform distribution of the sodium silicate solution, although for some samples more content was deposited on the bottom side. For this reason, both sides of the same samples were tested when necessary.

Furthermore, no biological colonization was visually observed even after tests with moisture, such as for hygroscopic performance.

#### 4.3.3.2 Apparent density and thermal properties

Table 4.3-4 reports the results of the apparent density at T = 50 °C, the thermal conductivity at T = 20 °C, and RH = 60 % for each sample, the average value, and the standard deviation (S.D.).

Table 4.3-4. Results of apparent density at T = 50 °C, thermal conductivity at T = 20 °C, RH = 60% for each tested sample, average value, and standard deviation (SD).

| Composite          | Apparent density [kg/m <sup>3</sup> ] | Thermal conductivity [W/(m.K)] |
|--------------------|---------------------------------------|--------------------------------|
| H1                 | 729                                   | -                              |
| H2                 | 698                                   | -                              |
| H3                 | 680                                   | -                              |
| H7                 | -                                     | 0.16                           |
| H8                 | -                                     | 0.15                           |
| H9                 | -                                     | 0.17                           |
| Average value ± SD | 702 ± 25                              | 0.16 ± 0.01                    |

The use of volume instead of mass to define the quantities of aggregate-binder and the uncontrolled distribution of the grain size could determine a variation in mass between the samples. Furthermore, the homogeneous distribution of the sodium silicate between the aggregates was only visually checked and the internal disposition of the adhesive and the aggregates was not controlled. Consequently, some samples might contain more fine aggregates, exhibit different internal porosity, and have fewer internal voids, or smaller ones. However, all these considerations were reported by the standard deviation which was low if compared with the average value. Hence, a good homogeneity between samples was guaranteed.

The results were compared with some commercial products: Celenit N, a thermal and acoustic insulation board made of wood fibre and Portland cement, had a density of 346 - 533 kg/m<sup>3</sup> (Celenit, 2022); an OSB panel, a density of 650 kg/m<sup>3</sup> (Federation E.P. OSB Panels, 2022). The apparent density of hazelnut shells-based boards was higher probably due to the bio-aggregate. Indeed, the hazelnut shells had a bulk density higher than wood particles (Cintura et al., 2022). Antunes et al. (2019) considered panels made up of rice husk, earth, air lime, and hemi-hydrated gypsum and reached values of density between 651 kg/m<sup>3</sup> and 1022 kg/m<sup>3</sup>, more similar to the ones of the present study. This similarity was probably due to the considered binder, as rice husk particles had a bulk density lower than the one of the hazelnut shells, namely 85 kg/m<sup>3</sup> (Antunes et al., 2019).

As for the thermal properties and the comparison with commercial products: an Expanded Polystyrene (EPS) board had a thermal conductivity of 0.045 W/(m.K) (Ramos et al., 2021); an Extruded Polystyrene (XPS) board of 0.037 W/(m.K) (Ramos et al., 2021); an insulation expanded cork board (ICB) of about 0.042 - 0.045 W/(m.K) (Fu et al., 2020; Tártaro et al., 2017); a Celenit N board had a declared thermal conductivity of 0.065 W/(mK) (Celenit, 2022).

The maximum value to consider a material as a thermal insulator is 0.065 W/(m.K) (Aurélien Laborel-Préneron et al., 2017; Pina dos Santos and Matias, 2006; Romano et al., 2019), even if several past studies considered 0.1 W/(m.K) (Binici et al., 2020; Martínez-García et al., 2020; Mati-Baouche et al., 2014). Considering that the value of thermal conductivity was higher - (0.16 ± 0.01) W/(m.K) -,



the hazelnut shells-based composite cannot be considered a good thermal insulator and is not competitive with the commercial ones. This was expected since the materials considered for comparison had also a lower density. However, the results were in line with literature values. For example, Park et al. (Park et al., 2019) reported thermal conductivity values between 0.074 W/(m.K) and 0.108 W/(m.K) for wood-lime boards; Mathews et al. (Mathews et al., 2023) achieved values between 0.066 W/(m.K) and 0.128 W/(m.K) for cardboard-based panels; Antunes et al. (Antunes et al., 2019) achieved values of 0.102-0.197 W/(m.K) for the previously mentioned rice husk-earth based panels. For this latter, considering the higher value of the reported range, namely 0.197 W/(m.K), the hazelnut-based composite showed better thermal insulation performance.

Thermal properties might be improved by an optimization of the percentual proportion of aggregate - adhesive, and the production process, namely by lowering the amount of sodium silicate. Cintura et al. (2023a) evaluated the thermal conductivity at  $T = 17\text{ }^{\circ}\text{C}$  and  $\text{RH} = 65\%$  of samples made up of 25 % of sodium silicate solution and 75 % hazelnut shells (percentages by volume) and achieved values of  $(0.08 \pm 0.03)\text{ W/(m.K)}$ . However, the increase of the thermal insulation performance by using less amount of sodium silicate solution may cause a worsening of the mechanical properties. The use of 25 % of adhesive (by volume) did not secure enough mechanical resistance. Furthermore, not only the percentages aggregate-adhesive but also the test method, the considered conditions and the type of hazelnuts were different, and these aspects have to be considered to carry out the comparisons. Finally, as Mathews et al. (2023) reported, homogeneity and morphology play an important role in thermal insulation performance.

#### 4.3.3.3 Hygroscopicity and moisture buffering value

Figure 4.3-8 shows the moisture adsorption/desorption curves for four adsorption/desorption cycles considering three composite samples H1, H2, and H3.

The results demonstrated that the composite samples were highly hygroscopic. The moisture sorption capacity was greater than the moisture desorption capacity for all cycles. As for the moisture content difference, it remained almost the same for all cycles, with values between  $0.057\text{ kg/m}^2$  and  $0.066\text{ kg/m}^2$ . This meant that the composite had a great moisture storage capacity: when the relative humidity increased it absorbed moisture; when it decreased, it did not release it totally. According to the moisture content difference values, this behaviour remained constant during the four cycles.

To have a numerical value, rate the samples according to Rode et al. (Rode et al., 2005), and carry out an easier comparison with past research, the MBV was calculated (as described in Section 4.3.2.3) for the last three cycles. It is important to underline that the MBV (named practical MBV) was defined by the Nordtest Project (Rode et al., 2005), considering 8 h-sorption phase at  $\text{RH} = 75\%$  and 16 h-desorption phase at  $\text{RH} = 33\%$ . In the present work, different conditions were considered (defined by ISO 24353 (ISO 24353, 2008)), and this could determine some differences in the results.



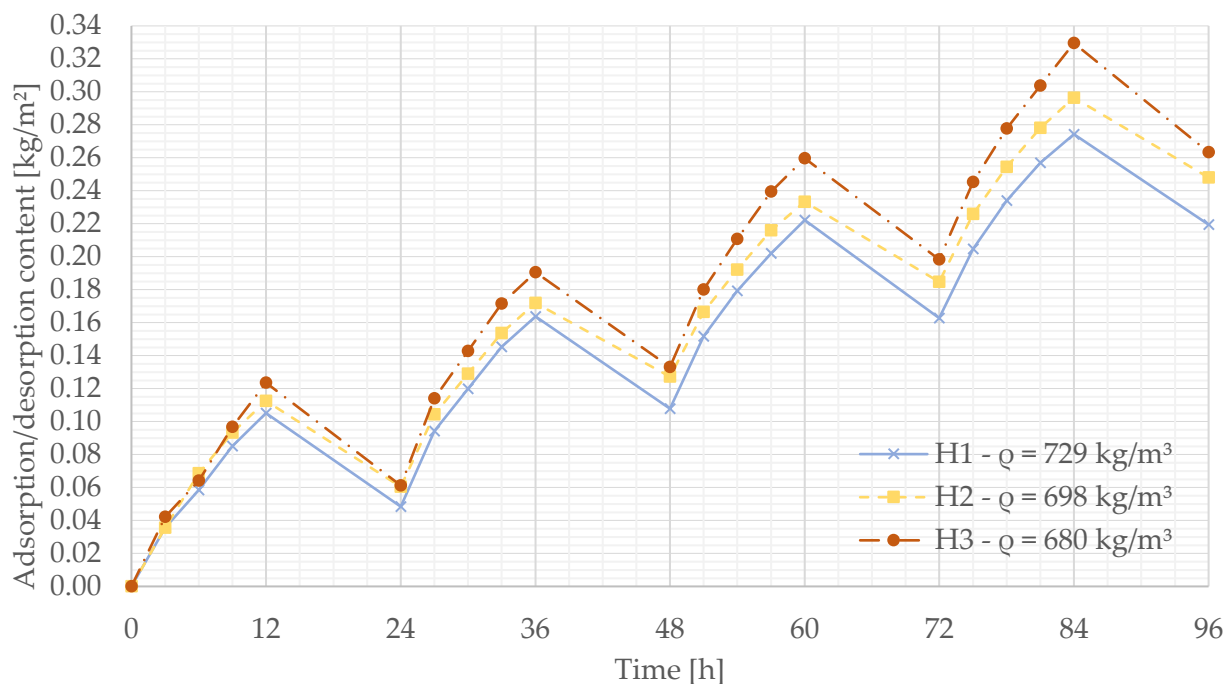


Figure 4.3-8. Moisture adsorption/desorption for four cycles of the three tested composite samples.

The MBV (considering the average value of the three tested samples and the standard deviation) was  $(3.45 \pm 1.76) \text{ g/(m}^2 \text{ \%RH)}$ . Liuzzi et al. (Liuzzi et al., 2020a) calculated the ideal MBV of composites made up of straw fibres and olive fibres bounded by sodium silicate solution. Practical and ideal MBV are similar for homogeneous composites and if the penetration depth is equal to or lower than the thickness of the samples (Rode et al., 2005). The researchers reported values of  $5.05 \text{ g/(m}^2 \text{ \%RH)}$  for straw-based composites and  $3.29 \text{ g/(m}^2 \text{ \%RH)}$ , for olive fibre-based ones. Considering the classification provided by Rode et al. (Rode et al., 2005), the samples made up of bio-aggregates and sodium silicate were rated as "Excellent", showing high moisture buffering capacity. This confirmed the expectation since the sodium silicate solution is highly hygroscopic (low moisture resistance is one of its main drawbacks (Lee and Thole, 2018; Song et al., 2021)). Furthermore, hazelnut shells as raw material showed high hygroscopicity and moisture absorption capacity greater than desorption (Cintura et al., 2022), coherently with the properties of the composite and other bio-based materials (Bardage, 2017; Lei and Feng, 2020). Hence the great absorption capacity of the composites was probably caused both by the bio-waste used as aggregate and the adhesive.

Cintura et al. (2023a) evaluated the moisture sorption/desorption capacity of samples made up of hazelnut shells and sodium silicate solution (75 % and 25 % by total volume, respectively, although not exactly the same). The researchers reported the values of the moisture content variation, considering the last four of six cycles of sorption-desorption. Figure 4.3-9 reports the comparison of the

results, considering the same cycles: the last two cycles for the samples evaluated in the present work; and the first two cycles of the reference work.

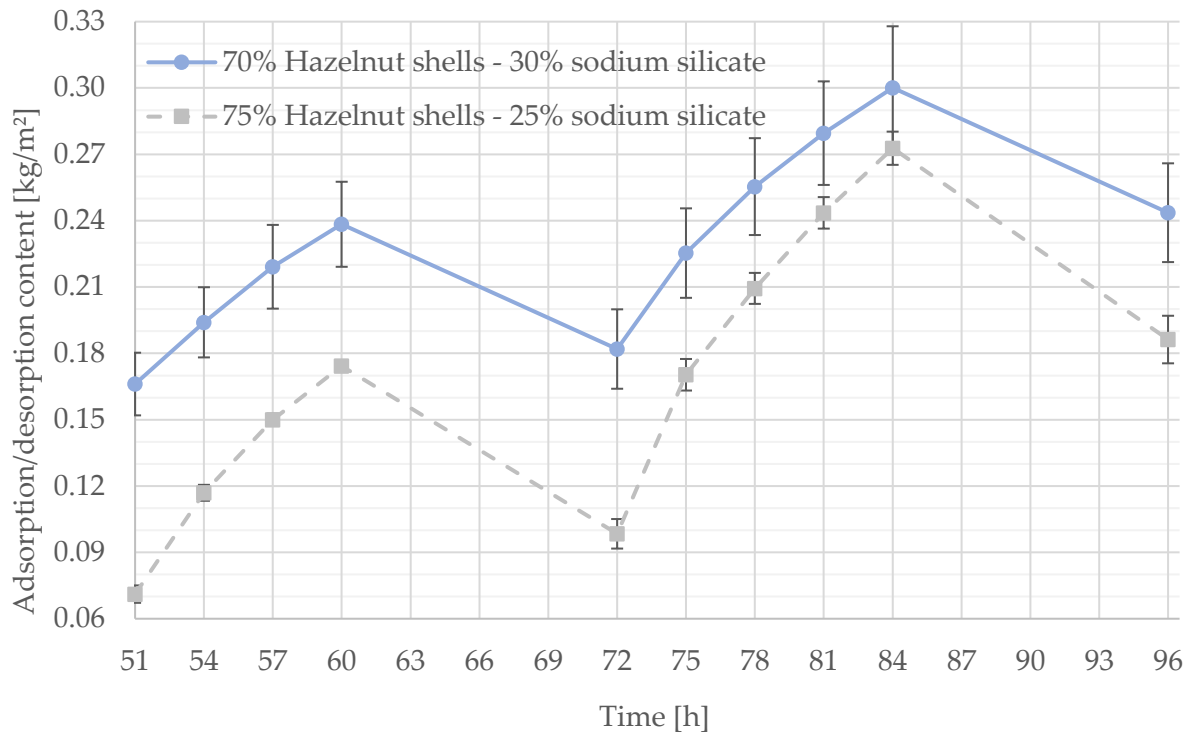


Figure 4.3-9. Moisture adsorption/desorption content for two adsorption/desorption cycles: comparison of the results (average value of three samples) for the composite evaluated in the present study and a composite made up of 75% of hazelnut shells and 25% of sodium silicate solution by the total volume (Cintura et al., 2023a).

Figure 4.3-9 shows that the values of the present study were in line with the past research. The trend of the curve was similar; the differences in the values could be determined by several aspects, e.g., the use of a different type of hazelnut shells, differences in the samples' production, and the employment of different equipment. Carrying out a simplified comparison, due to the several differences in the test's conditions, the highest values of moisture content were achieved for the highest percentages of the sodium silicate solution. This could demonstrate that the great absorption capacity was derived from the sodium silicate, more than from the hazelnut shells.

#### 4.3.3.4 Acoustic properties

Figure 4.3-10 reports the results of the acoustic absorption coefficient for each sample tested by both sides: the exposed surface rougher (dotted lines, sample designation "A", namely H4A, H5A, H6A), and the mould base surface, flatter (continuous lines, samples designation "B", namely H4B, H5B, H6B). Frequencies lower than 100 Hz were not reported due to the operating frequency range of the employed impedance tube (Section 4.3.2.3).

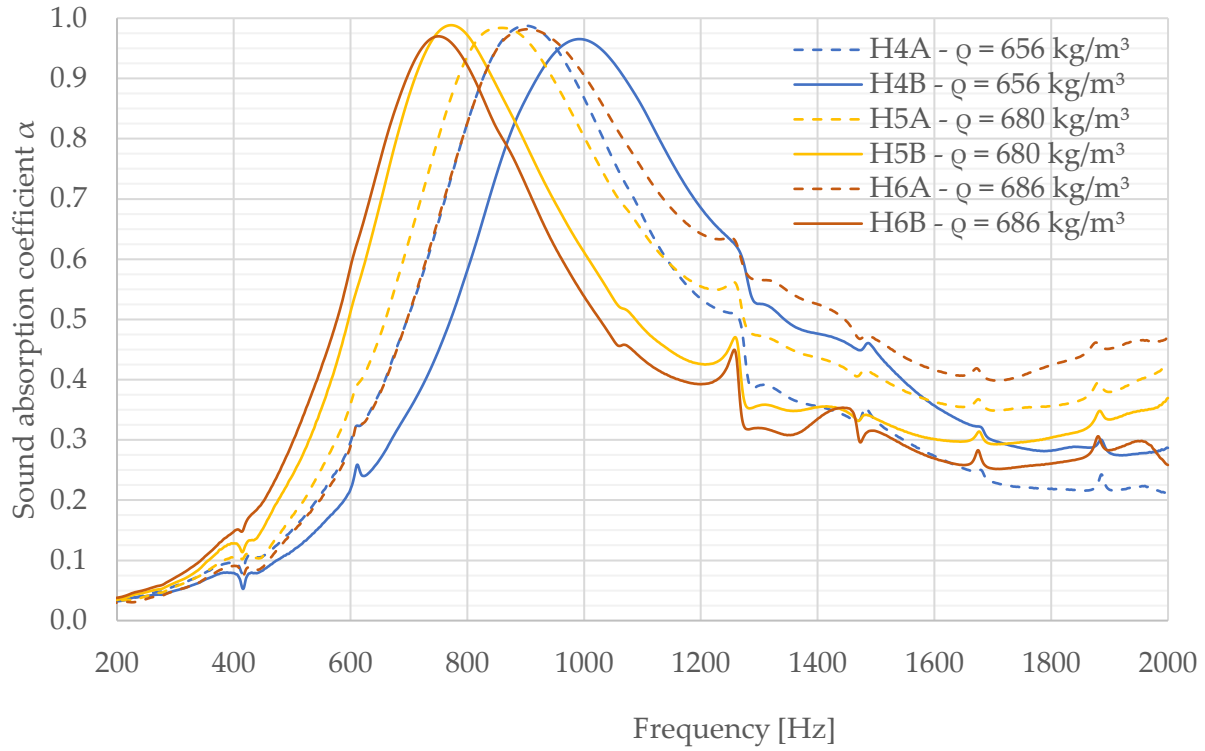


Figure 4.3-10. Sound absorption coefficients of both sides of the three tested samples. Continuous lines represent the results for the flatter samples' exposed surfaces (lower part of the mould), namely the samples designated by letter B (H4B, H5B, H6B); the dotted lines for the ones rougher (upper part of the mould), sample designated by letter A (H4A, H5A, H6A).

The results for the two sides of the samples were similar and this confirmed these samples seemed to be homogeneous in terms of sodium silicate content. The differences among the curves could be caused by the random disposition of the bio-aggregates (de Carvalho et al., 2020) and, therefore the different internal voids of each sample. The acoustic absorption behaviour suggests that the samples could be considered granular material (Horoshenkov and Swift, 2001).

Granular materials may be viewed as a special case of porous materials, and, as Figure 4.3-10 shows, they show a first peak of normal incidence sound absorption,  $f_0$ , described by Equation 4.3-12:

$$f_0 = \frac{1}{2\pi} \sqrt{\frac{k}{M_{eff}}} \quad (4.3-12)$$

where  $M_{eff}$  is the effective mass of granular material and  $k$  is the apparent dynamic longitudinal elastic modulus, a parameter that depends on several properties (e.g., bulk density, thickness, sound pressure on the material's surface), as better detailed in past studies (Horoshenkov and Swift, 2001; Voronina and Horoshenkov, 2004).

The results were compared and validated with some literature values. Liuzzi et al. (2020b), analysing the acoustic behaviour of almond skin-based composites, proved it is strongly dependent on

the thickness, tortuous morphology, and pore structure. Martellotta et al. (2018) considered composites made of wastes deriving from olive pruning (average leaf length of 40 mm) and chitosan dissolved in acetic acid and water as binder, produced without pressing. The researchers obtained a sound absorption curve that showed a less pronounced peak at higher frequencies. In any case, most of the materials from the literature review show poor acoustic properties at mid frequencies and good acoustic properties at high frequencies only, also because the bio-wastes are mainly used and analysed as fibres or as light aggregates, rather than as high-mass granular aggregates. Instead, the material under study showed good absorption properties around 800 Hz and is suitable in environments where absorption is often needed by human noise sources, such as restaurants, offices, or museums (D'Orazio et al., 2020).

#### 4.3.3.5 Flexural strength

Figure 4.3-11 shows the stress-strain curve for the eight tested samples. The dotted lines represent the samples tested with the upper side as the flatter one and the lower side as the rougher one (Samples\_LOW). The continuous ones represent the samples tested in an opposite position, with the upper side as the one rougher, and the lower side as the flatter one (Samples\_UP).

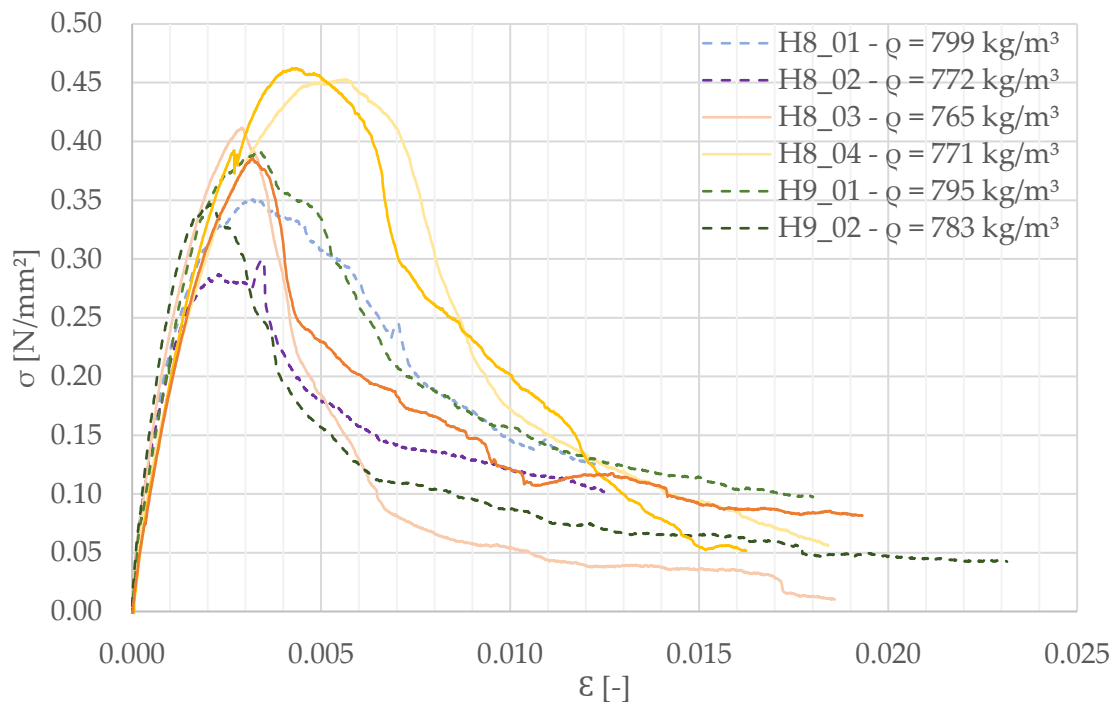


Figure 4.3-11. Stress-strain curves of the eight tested samples. The dotted lines represent the samples with the upper side as the flatter one, and the lower side as the rougher (Samples\_LOW); the continuous lines represent the samples with the upper side as the rougher one, and the lower side as the flatter one (Samples\_UP).

Table 4.3-5 reports the maximum recorded force and displacement (considering the average values and standard deviation), the flexural strength, and the modulus of elasticity, calculated as reported in Equation 4.3-9.

Table 4.3-5. Average values and standard deviation of maximum force, displacement, flexural strength, and modulus of elasticity for the bending test.

|             | $F_{\max}$<br>[N] | $\delta_{\max}$<br>[mm] | $\sigma_{\max}$<br>[N/mm <sup>2</sup> ] | $E_{(20\%-60\%)}$<br>[N/mm <sup>2</sup> ] |
|-------------|-------------------|-------------------------|-----------------------------------------|-------------------------------------------|
| Samples_LOW | 156.2 ± 25.4      | 2.4 ± 0.6               | 0.35 ± 0.04                             | 204.4 ± 34.7                              |
| Samples_UP  | 203.6 ± 21.5      | 3.0 ± 1.0               | 0.43 ± 0.04                             | 167.4 ± 25.1                              |
| All         | 179.9 ± 33.4      | 2.7 ± 0.8               | 0.39 ± 0.06                             | 185.9 ± 34.3                              |

All the samples showed the same flexural behaviour, with a first elastic behaviour followed by a post-peak path. The comparison between the differently stressed samples (continuous and dotted lines in the graph) demonstrated that when the lower side was the flatter one, hence the flatter side was stressed by flexural stress, the maximum applied force was 30% higher and the flexural strength increased by 23%. This can be caused both by a greater percentage of sodium silicate that increased the internal bond of the aggregates, as reported by Ng et al. (Ng et al., 2018), and by the presence of a more linear surface as the stressed side, guaranteed by the mould surface during the curing time. The type of aggregate (slightly rounded and small, as reported in Figure 4.3-1) and the production process determined a rougher-surface composite, that implied low flexural strength. These results demonstrated that the resistance was mainly provided by the adhesive.

Binici and Aksogan (2016) evaluated the mechanical properties of boards made up of olive seeds, wood chips, ground PVC, dehydrated gypsum, epoxy, and water. For the samples made of dehydrated gypsum, olive seeds, and different percentages of water, the researchers achieved values of flexural strength between 0.061 N/mm<sup>2</sup> and 0.074 N/mm<sup>2</sup>. Antunes et al. (2019) for rice husk-earth-lime-gypsum panels reported values of flexural strength between 0.08 - 0.12 N/mm<sup>2</sup>. The results of the hazelnut shell-based composite were in line with literature values, and showed greater flexural strength, even if their mechanical performance remained low.

In the considered past studies, the bending test was always performed by considering a three-point bending test. As previously reported, by this method the possible effect of shear stress cannot be avoided, but, on the other hand, as these composites had usually a non-perfectly plain surface, the three-point bending test could be helpful for the implementation of the test.

#### 4.3.3.6 Compressive strength

Figures 4.3-12 and 4.3-13 show the stress-strain curve for the compressive test and one sample after the test, respectively. As for the sample designation, letters A and B distinguished the two extremities

of the original strips already employed for the bending test (i.e., the samples employed for flexural resistance analysis, cut after the test).

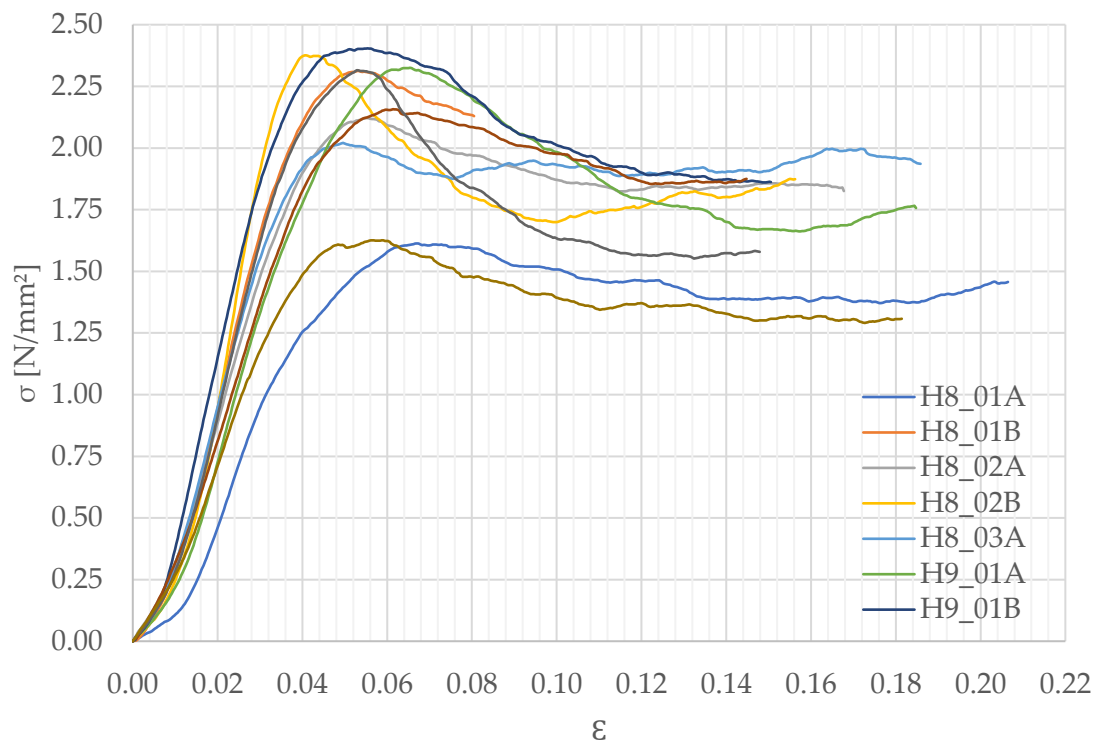


Figure 4.3-12. The stress-strain compressive curve of the ten tested samples 10 cm x 10 cm x 4 cm.



Figure 4.3-13. Rectangular sample of hazelnut shells and sodium silicate solution after the compressive test and the achievement of the breaking load.

Table 4.3-6 presents the average values and standard deviation of the maximum recorded force and displacement, the compressive strength, and the modulus of elasticity, calculated as reported in Equation 4.3-9.

Table 4.3-6. Compressive test: average values and standard deviation of maximum force, displacement, compressive strength, and modulus of elasticity.

| $F_{\max}$<br>[N] | $\delta_{\max}$<br>[mm] | $\sigma_{\max}$<br>[N/mm <sup>2</sup> ] | $E_{(20\%-60\%)}$<br>[N/mm <sup>2</sup> ] |
|-------------------|-------------------------|-----------------------------------------|-------------------------------------------|
| 21737.8 ± 2992.3  | 2.4 ± 0.4               | 2.1 ± 0.3                               | 64.9 ± 13.8                               |

All the samples achieved a peak and then collapsed. All the samples showed similar behaviour before the peak. Then, they showed a flat post-peak behaviour (Figure 4.3-12). The compressive behaviour of the samples was consistent with expectations: the semi-round shape aggregate and the production process resulted in a low cohesion between the aggregates and the adhesive, leading to the rupture.

Antunes et al. (2019), for the composite made essentially of rice husk and earth, achieved values between 0.13 N/mm<sup>2</sup> and 0.40 N/mm<sup>2</sup> for compressive strength. Mathews et al. (2023) considered thermal insulation panels made up of different percentages of recycled cardboard as aggregates, boric acid, and three different binders. The values of compressive strength were between 7.80 - 15.00 N/mm<sup>2</sup> for the corn starch-based binder, 6.60 - 7.00 N/mm<sup>2</sup> for the lime, and 6.20 - 8.50 N/mm<sup>2</sup> for the clay. The Celenit N board (Celenit, 2022), previously considered to compare the other properties, showed an extremely different mechanical behaviour, being classified as ductile material. Indeed, for the compressive strength, the maximum strength at 10% deformation was reported, not achieving the breaking, as indicated in the reference standards (EN-826, 2013):  $\sigma \geq 0.2$  N/mm<sup>2</sup> (for a thickness of 15-40 mm) and  $\sigma \geq 0.15$  N/mm<sup>2</sup> (for a thickness of 50-75 mm). The same behaviour was shown by expanded cork boards (ICB), analysed by Simões et al. (Simões et al., 2019). They considered both standard and medium density, namely between 90 - 110 kg/m<sup>3</sup> and 140-160 kg/m<sup>3</sup>, respectively. The reported compressive strength for a thickness of 50 mm was 0.154 N/mm<sup>2</sup> (standard density) and 0.205 N/mm<sup>2</sup> (medium density), but these values were considered at 10% strain value since the ICB did not reach the breaking point, demonstrating the great difference in performance between the two composites, previously compared.

Therefore, the results of the tested composite were in line with the literature values; considering the composites that demonstrated the same behaviour. The differences could be derived from the different test methods as well as the different shapes and sizes of the samples. Probably, a smaller grain size could increase the compressive strength, since it could reduce the size of the internal voids, although also changing other properties. Certainly, the analysed composites cannot be used as structural products, as expected, but as coatings or intermediate layers.

## 4.3.4 Discussion

### 4.3.4.1 Main findings of the present study

The main findings and contributions of the present study can be summarized in three points.

First, this research demonstrated the feasibility of producing a composite by considering the selected materials and the described production process. The combination of the hazelnut shells and the sodium silicate solution resulted in the production of high-mass boards, less investigated than fibres in the field of bio-waste-based composites (Faruk et al., 2012; Sanjay et al., 2018; Savio et al., 2022).

The described wide-range experimental campaign provided a methodology for the characterization of an unconventional composite, that could be useful for future studies.

Finally, the most interesting finding was the high hygroscopicity together with an uncommon acoustic absorption behaviour of the hazelnut shells-based composite. As better detailed in the following sections, an MBV of about  $(3.45 \pm 1.76) \text{ g}/(\text{m}^2 \% \text{RH})$ , and a peak of sound absorption between 700 - 900 Hz suggested that the proposed composite could improve indoor comfort if applied as an internal coating board.

### 4.3.4.2 Comparison with other studies about high-mass panels and boards

Table 4.3-7 provides a summary of the comparison between the hazelnut shells-based composites and some significant literature values of past studies considered in the previous sections (Section 4.3.3). Only high-mass bio-aggregate-based composites more similar to hazelnut shells-based ones were reported.

Considering the thermal conductivity and density, the hazelnut shells-based composite could be easily compared to the high-density panels proposed by Antunes et al. (2019). It showed better mechanical properties and, considering the panels with 15% dried rice husk, also better thermal insulation. Both properties mainly depended on the bio-based aggregate and the configuration provided by the mixture aggregate-adhesive. The mechanical performance (both compressive and flexural strength) appeared low, but the comparison demonstrated it was in line with past research.

Composites made using the same adhesive (Liuzzi et al., 2020a) presented more discrepancy in terms of physical properties, but this was because the reference past study, which achieved better thermal insulator performance and lower density, considered lighter and fibrous aggregates.

Concerning the hygroscopic properties, all the bio-composites presented high MBV, coherently with the expectation. The employed bio-aggregate, the sodium silicate solution, and the binder, consisting mainly of clayish earth, were known to be highly hygroscopic (Lima et al., 2020).



Table 4.3-7. Summary of the comparison between the hazelnut shells-based composites and literature values.

| Composites                                                                        | Properties                      |                                        |                                 |                                                    |                                              |                                                      | Reference                      |
|-----------------------------------------------------------------------------------|---------------------------------|----------------------------------------|---------------------------------|----------------------------------------------------|----------------------------------------------|------------------------------------------------------|--------------------------------|
|                                                                                   | Density<br>[kg/m <sup>3</sup> ] | Thermal con-<br>ductivity<br>[W/(m.K)] | MBV<br>[g/(m <sup>2</sup> %RH)] | Sound absorption coeffi-<br>cient *                | Flexural<br>strength<br>[N/mm <sup>2</sup> ] | Compres-<br>sive<br>strength<br>[N/mm <sup>2</sup> ] |                                |
| Hazelnut shells and sodium silicate                                               | 702 ± 25 <sup>a</sup>           | 0.16 ± 0.01                            | 3.45 ± 1.76 <sup>c</sup>        | α≈0.8 at 700-900 Hz                                | 0.39 ± 0.06                                  | 2.1 ± 0.3                                            | Present work                   |
| Olive pruning waste and so-<br>dium silicate                                      | 235 ± 11 <sup>b</sup>           | 0.062 ± 0.003                          | 3.29 <sup>d</sup>               | α≈0.8 at 3150 Hz                                   | -                                            | -                                                    | (Liuzzi et al.,<br>2020a)      |
| Straw fibres and sodium sili-<br>cate                                             | 152 ± 7 <sup>b</sup>            | 0.058 ± 0.003                          | 5.05 <sup>d</sup>               | 25 mm: α≈0.8 at 2000 Hz<br>50 mm: α≈0.8 at 1250 Hz | -                                            | -                                                    | (Liuzzi et al.,<br>2020a)      |
| Earth, hemihydrated gypsum,<br>air lime, and 15% dried rice<br>husk <sup>1</sup>  | 1021.6 ±<br>16.8                | 0.197 ± 0.004                          | 2.97                            | -                                                  | 0.12 ± 0.03 <sup>e</sup>                     | 0.37 ± 0.05                                          | (Antunes et al.,<br>2019)      |
| Earth, hemihydrated gypsum,<br>air lime, and 30% dried rice<br>husk <sup>1</sup>  | 650.8 ±<br>14.6                 | 0.102 ± 0.004                          | 4.11                            | -                                                  | 0.08 ± 0.01 <sup>e</sup>                     | 0.13 ±<br>0.001                                      | (Antunes et al.,<br>2019)      |
| Earth, hemihydrated gypsum,<br>air lime, and 30% boiled rice<br>husk <sup>1</sup> | 886.2 ±<br>65.3                 | 0.121 ± 0.004                          | 4.94                            | -                                                  | 0.08 ± 0.01 <sup>e</sup>                     | 0.40 ± 0.25                                          | (Antunes et al.,<br>2019)      |
| Shredded stems of sunflower<br>and chitosan                                       | 186.42 ±<br>0.49                | 0.077                                  | -                               | α≈0.65 at 1000 Hz<br>(except one sample)           | -                                            | -                                                    | (Mati-Baouche<br>et al., 2016) |
| Olive pruning waste and chi-<br>tosan <sup>2</sup>                                | 79.5                            | -                                      | -                               | α≈0.5 at 1250 Hz                                   | -                                            | -                                                    | (Martellotta et<br>al., 2018)  |

Note:

\* Only similar absorption curves were considered, namely curves that presented a peak. If the curve presented two peaks, the α value for the first one is reported.

<sup>1</sup> Percentages in volume of earth; <sup>2</sup> Only one type of sample of the considered study was reported (the most significant for the comparison with the present study);<sup>a</sup> Apparent density; <sup>b</sup> True density; <sup>c</sup> Calculated by considering ISO 24353 (ISO 24353, 2008); <sup>d</sup> MBV<sub>ideal</sub>; <sup>e</sup> Calculated by considering three-point bending test.

The sound absorption results were simplified by reporting the absorption coefficient value, but it would be useful to also analyse the absorption curve trends for all the considered studies. The comparison pointed out that the hazelnut shells-based composite better performed at frequencies lower than the reported composites. Glé et al. (2011) and Kinnane et al. (2016) evaluated the acoustic properties of hemp shiv as raw material and provided a curve more similar to the one presented in this study, pointing out the influence of the compaction and the grain size of the aggregates in the results. However, the sound absorption curves of the hemp concrete were different from the hazelnut shells-based ones. Hence, these results were not reported in Table 4.3-7. The results of sound absorption for the hazelnut shells-based composite were very similar to those obtained by Martellotta et al. (2018), although the researchers achieved a less pronounced peak. All in all, all the reported composites seemed to have internal voids that guaranteed the dissipation of the sound wave combining porosity, tortuosity, and stiffness.

#### **4.3.4.3 Implication and explanation of findings**

The results and the comparison with the literature values demonstrated that the hazelnut shells-based composite had promising sound absorption performance and interesting behaviour at low and medium frequencies. As previously described (Section 4.3.3.4), it showed acoustic absorption performance typical of granular materials, extremely rare in bio-based materials. Furthermore, the composite showed high moisture storage capacity, guaranteed both by the hazelnut shells and the sodium silicate solution (Cintura et al., 2022; Song et al., 2021).

The thermal conductivity value was too high to consider the composite as a good thermal insulator. It might be more comparable to thermal mortars, as 0.2 W/(m.K) is the maximum value required to consider a composite as such (EN 998-2, 2016).

As for the mechanical properties, they were consistent with expectations, as detailed in Sections 4.3.3.5 and 4.3.3.6, in line with other examples of unconventional, non-structural boards (Table 4.3-7). The rupture was caused by the interface aggregate-adhesive. Despite both the hazelnut shells and the dried sodium silicate solution being brittle materials, a post-peak branch is present in the stress-strain diagram of the composite.

In any case, the research did not aim at proposing a composite with high flexural and compressive strength, although enough mechanical performance is required to guarantee the applicability of the building products, such as transportation, installation, and use.

#### 4.3.4.4 Strengths and limitations

The hazelnut shells-based composite presented both strengths and weaknesses.

First, it could be considered a sustainable building solution, mainly due to the employed aggregate: being the hazelnut shells widely produced by-products, the environmental impact caused by its production process was minimized (Rubino et al., 2023). Moreover, hazelnut shells are largely available all over the year, which overcomes one of the main drawbacks of using agro-industrial waste, namely the seasonality. Finally, the hazelnut shells used in this study were locally produced, reducing transportation energy waste when used locally.

Although the sodium silicate solution has a higher environmental impact, it has several advantages (Section 4.3.1). Among these, the non-toxicity (Chai et al., 2021; Lee and Thole, 2018) is an extremely important aspect, since a bio-waste-based composite could have many benefits, but applications are reduced if it can be harmful to human health. For instance, a formaldehyde-based bio-board could be more sustainable than others but potentially dangerous and could cause several diseases (Owodunni et al., 2020; Solt et al., 2019). Additionally, the sodium silicate solution can potentially increase the resistance to biological attack (Lee and Thole, 2018; Liuzzi et al., 2020a), as no biological colonisation was visually observed even after hygroscopic tests. It allowed low-temperature casting and curing, and avoided hot-pressing during the production process, further reducing energy use (Cintura et al., 2023a).

A qualitative evaluation pointed out that several aspects made the choice of the aggregate, the adhesive, the production process, and the casting and curing phases environmentally friendly. Certainly, further studies are needed to demonstrate the sustainability of the proposed solution, such as a life cycle assessment.

The results of the acoustic analysis were certainly one of the main strengths of the proposed composite since it could guarantee the absorption of frequencies in the range of human voice, which is between 500 - 1000 Hz (EN IEC 60268-16, 2020; ISO 3382-3, 2012). The high moisture storage capacity could be a benefit as the hazelnut shells-based composite could adsorb moisture, securing a passive control of indoor conditions.

However, the high hygroscopicity could also cause several problems such as materials' degradation, thickness swelling, variation in the final performance, the mould growth at a critical moisture level (Johansson et al., 2020; Nunes et al., 2017). Nevertheless, and as previously mentioned, no visual observation of biological colonization is a good preliminary indicator for a good performance.

#### 4.3.4.5 Recommendations and future studies

The proposed boards might be suitable as indoor coatings with thermal performance, but mainly interesting sound absorption and hygroscopic capacity that gets even more interesting, as they could provide a passive control of the indoor conditions, too (Nguyen et al., 2020; Ratiarisoa et al., 2016). They could be employed in indoor rooms where the speech and the involuntary listening of the other occupants can cause acoustic discomfort, such as lecture halls, offices, restaurants, and libraries (Cingolani et al., 2021; D'Orazio et al., 2020; De Salvio et al., 2021), apart from contributing to indoor hygrothermal comfort too.

The low mechanical performance should be taken into consideration for application. It seems an adhesive application should be better suited than an alternative with a space layer behind it. The mechanical performance might be improved by varying the considered parameters or by adding additional materials.

Further studies could be carried out. For example, it could be interesting to define the contribution of temperature and relative humidity to the test results (Fusaro 2023; Cingolani et al., 2022; Liuzzi et al., 2017), how the high hygroscopicity could modify the final performance, and how the properties could change by considering different parameters (e.g., by changing the samples' thickness, aggregates' grain size, and production process). The reaction to fire must be assessed, considering not only the flammability but also the possible production of toxic gases by combustion and smouldering (Laborel-Préneron et al., 2017; Palumbo et al., 2017). Finally, the degradation and the biodeterioration could be evaluated, by analysing aspects such as the biological attack by fungi and insects, and the critical moisture levels, as these aspects could deeply change the final performance (Johansson et al., 2013; Martins et al., 2011).

#### 4.3.5 Conclusions

In this study, a composite board made up of hazelnut shells as aggregate and a sodium silicate solution as adhesive was investigated. The described work provided an important contribution to this research field.

First, it demonstrated the feasibility of re-using a widely available by-product for building practices. This possibility can lower the environmental impact derived from the construction sector and the disposal of agro-industrial wastes.

The proposed composite was made of natural, non-harmful materials. As an additional scientific value, the considered agro-industrial by-product was employed as a high-mass bio-

aggregate. This type of composite is not so commonly investigated in the field of bio-based building products.

The description of the considered methods and the laboratory experience provided useful information for future research. Indeed, hygrothermal, acoustic, and mechanical properties were assessed and compared with the literature to provide a broad characterization of the produced boards. This horizontal analysis allowed getting in touch with different areas of knowledge in the field of civil engineering.

As for the evaluated properties, the following conclusions were achieved:

- The hazelnut shells-based boards did not show very good thermal insulation performance. Lower values of both apparent density and thermal conductivity may be achieved by using several strategies, such as lowering the amount of the sodium silicate solution (but this could worsen the mechanical properties), using alternative adhesives (instead of the sodium silicate solution), or a different production process.
- The composite showed high hygroscopicity, probably due to the sodium silicate solution, with a greater capacity of sorption rather than desorption. This property could be an advantage for application as internal coating boards to ensure passive control of building indoor conditions. However, although no biological colonization was visually observed after the hygroscopic test, the high moisture storage capacity could cause composites' degradation and/or modify their properties.
- As for sound absorption, the hazelnut shells-based board performed as a granular material, showing the highest absorption capacity for frequencies around 700 - 900 Hz, which are in the frequency range of the human voice. The results of the acoustic analysis are extremely interesting also because the composite showed an uncommon behaviour, considering the bio-waste-based composites already analysed. A possible optimization could improve the absorption performance, expanding the high acoustic absorption capacity for a larger frequency range.
- As expected, the hazelnut shells-based board did not show high mechanical properties. A higher content of sodium silicate solution could probably improve the flexural strength, decreasing the internal voids and increasing the cohesion between the hazelnut shells. However, minimizing the use of adhesive and using higher quantities of by-products would be better for a more sustainable building composite.

The achieved conclusions demonstrated that a possible application of the composite could be as indoor coatings with good acoustic absorption performance, absorbing the medium frequencies (e.g., in lecture halls, offices, classrooms) and acting as a moisture buffer. The evaluation of the resistance to fire and the biological susceptibility should be addressed.

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## 4.4 Article G - Characterization of an *Arundo donax*-based composite: A solution to improve indoor comfort.

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### Abstract

*Arundo donax* (giant reed or giant cane) is a widely available, perennial, invasive, non-food crop, present worldwide and employed for several uses, including building practices. Considering the increasing demand for sustainable building materials, *A. donax* can be an efficient solution. This study investigated its properties as a bio-aggregate mixed with a sodium silicate solution as an adhesive. A horizontal analysis that provided a general characterization of the composite was carried out. The results showed that the *A. donax*-based composite had an apparent density of 517 kg/m<sup>3</sup>, thermal conductivity of 0.128 W/(m.K), and high hygroscopicity, with a moisture buffering value of 4.33 g/(m<sup>2</sup> %RH), property that could be both an advantage for indoor comfort and a drawback. The uncommon sound absorption behaviour can be comparable to granular materials, with the highest sound absorption coefficient values,  $\alpha$ , between 600 Hz and 700 Hz. Due to the range and the shape of the acoustic absorption property, this material may be helpful in acoustic treatments for speech noise. The mechanical tests defined flexural and compressive strength, respectively, 0.35 N/mm<sup>2</sup> and 0.9 N/mm<sup>2</sup>, ensuring applicability. Above all, these tests opened new possible solutions for *A. donax*-based composite production either alone or in combination with other agro-industrial wastes and justified further tests, such as fire resistance and bio-susceptibility.

**Keywords:** Acoustic absorption; Bio-waste; Giant reed; Granular Material; Hygroscopicity; Mechanical properties.

| Nomenclature, Symbols and Acronyms |                                           |            |                       |
|------------------------------------|-------------------------------------------|------------|-----------------------|
| A                                  | Samples area [m <sup>2</sup> ]            | $\epsilon$ | Strain [-]            |
| $\rho$                             | Apparent density [kg/m <sup>3</sup> ]     | F          | Applied force [N]     |
| $\alpha$                           | Sound absorption coefficient              | l          | Sample span [mm]      |
| R                                  | Airflow resistance [Pa.s/m <sup>3</sup> ] | b          | Sample width [mm]     |
| $\Delta p$                         | Air pressure difference [Pa]              | h          | Sample thickness [mm] |
| qv                                 | Air flow rate [m <sup>3</sup> /s]         | $\delta$   | Displacement [mm]     |

|                    |                                                             |                      |                                                                             |
|--------------------|-------------------------------------------------------------|----------------------|-----------------------------------------------------------------------------|
| $r$                | Airflow resistivity [kPa.s/m <sup>2</sup> ]                 | $\delta_{\max}$      | Maximum displacement [mm]                                                   |
| $d$                | Thickness [m] (in air flow resistivity eq.)                 | $E$                  | Modulus of elasticity [N/mm <sup>2</sup> ]                                  |
| $m_0$              | Initial mass [kg]                                           | $\sigma_{60\%}$      | $\sigma$ at 60% of $F_{\max}$ [N/mm <sup>2</sup> ]                          |
| $m_{d(n-1)}$       | Mass of previous desorption cycle [kg]                      | $\sigma_{20\%}$      | $\sigma$ at 20% of $F_{\max}$ [N/mm <sup>2</sup> ]                          |
| $m_{an}$           | Mass of considered absorption cycle [kg]                    | $\varepsilon_{60\%}$ | $\varepsilon$ at 60% of $F_{\max}$ [-]                                      |
| $m_{dn}$           | Mass of considered desorption cycle [kg]                    | $\varepsilon_{20\%}$ | $\varepsilon$ at 20% of $F_{\max}$ [-]                                      |
| $\rho_{A,ac}$      | Moisture sorption [kg/m <sup>2</sup> ]                      | $\Delta\sigma$       | Difference between $\sigma_{60\%}$ and $\sigma_{20\%}$ [N/mm <sup>2</sup> ] |
| $\rho_{A,dc}$      | Moisture desorption [kg/m <sup>2</sup> ]                    | $\Delta\varepsilon$  | Difference between $\varepsilon_{60\%}$ and $\varepsilon_{20\%}$ [-]        |
| $\rho_{A,sc}$      | Moisture content difference [kg/m <sup>2</sup> ]            | $A_0$                | Sample initial cross-sectional area [mm <sup>2</sup> ]                      |
| MBV                | Moisture buffering value [g/(m <sup>2</sup> .% RH)]         | $d_0$                | Sample initial thickness [mm]                                               |
| MBV <sub>a</sub>   | MBV during the sorption phases [g/(m <sup>2</sup> .% RH)]   | $f_0$                | Normal incidence sound absorption                                           |
| MBV <sub>d</sub>   | MBV during the desorption phases [g/(m <sup>2</sup> .% RH)] | $k$                  | Apparent dynamic longitudinal elastic modulus                               |
| RH                 | Relative humidity [%]                                       | $M_{\text{eff}}$     | Effective mass [kg]                                                         |
| RH <sub>high</sub> | Highest value of RH (considered 75%) [%]                    | $F_{10}$             | Force at 10% strain [N]                                                     |
| RH <sub>low</sub>  | Lowest value of RH (considered 50%) [%]                     | $\delta_{10}$        | Displacement at 10% strain [mm]                                             |
| $\sigma_{\max}$    | Flexural strength [N/mm <sup>2</sup> ]                      | $\sigma_{10}$        | Compressive strength at 10% strain [N/mm <sup>2</sup> ]                     |
| $F_{\max}$         | Maximum applied force [N]                                   | $E_{10}$             | Modulus of elasticity [N/mm <sup>2</sup> ]                                  |
| $\sigma$           | Stress [N/mm <sup>2</sup> ]                                 |                      |                                                                             |

#### 4.4.1 Introduction

Research is increasingly interested in the feasibility of using bio-based products for building practices, such as bio-wastes and bio-resources derived from agriculture. This solution both lowers the environmental impact caused by the construction and agriculture sectors (Ansell et al., 2020; Viel et al., 2019) and reduces the problem of solid waste disposal (Gaspar et al., 2020; Liuzzi et al., 2020b). Furthermore, it moderates the energy consumption derived from the production and use of building materials (Ntimugura et al., 2020).

Past research already addressed this topic. Cintura et al. (Cintura et al., 2021) reported many studies that considered bio-wastes and bio-resources for construction practices. Three main groups of bio-wastes were identified according to their origin: those derived from forest management and floriculture, those derived from agricultural and manufacturing techniques, and the ones produced by the food industry. Their physical and chemical properties, benefits, and drawbacks were described, as well as the studies that examined their employment as

building products. As this collection of studies demonstrated, many possibilities have already been investigated, considering several materials and types of composites. For example, Azevedo et al. (de Azevedo et al., 2022) evaluated different agro-industrial solid wastes (pine-apple, sugar cane, açai, coconut, and rice) to produce cementitious materials, such as mortars and concretes; Solt et al. (Solt et al., 2019) analysed formaldehyde-free adhesives for particleboards' production and reported cashew nut shell liquid; Rault et al. (Raut et al., 2011) and Muñoz Velasco et al. (Muñoz Velasco et al., 2016) considered bricks' production by using cotton waste and coffee grounds, respectively.

Among the several types of building composites, particleboards were widely investigated. They seemed to be promising building products for their versatility (e.g., employed in floors, ceilings, divider walls, and furniture), their affordability, and their sustainability (Gürü et al., 2015; Mahieu et al., 2019; Rubino et al., 2023). Furthermore, they ensured easy use of different types of bio-wastes, such as banana tree fibres (Nunes et al., 2017), hemp (Auriga et al., 2022; Pennacchio et al., 2017), corn cob (Ramos et al., 2021), flax fibres (Sam-Brew and Smith, 2015) and tree bark (Tudor et al., 2021).

In addition to these, some of the main addressed topics in modern research are the use of materials not harmful to human health and the improvement of indoor air quality and comfort (Fratoni et al., 2019), which can lower operational costs too (Posani et al., 2022; 2021). Therefore, one of the biggest challenges is the proposal of eco-friendly materials, able to passively ensure good indoor conditions, guaranteeing the well-being of both the environment and the users, and lowering energy requirements.

Two types of indoor comfort could be considered: hygrothermal and acoustic. The hygrothermal comfort is related to the operative temperature, a combination of air temperature and mean radiant temperature in the room (Posani et al., 2023). It depends on several factors, sometimes difficult to forecast (Ranesi et al., 2022). Therefore, a simplified evaluation was usually considered, involving information such as the predicted mean vote (PMV), predicted percentage of dissatisfaction (PPD), internal temperature, and relative humidity. The acoustic comfort is related to the protection against noise and is mainly guaranteed by adequate sound insulation (Sakthivel et al., 2021). Moreover, the control of the indoor acoustic environment is a crucial factor that enhances the comfort and well-being of the users (D'Orazio et al., 2020; De Salvio et al., 2023). This purpose makes a proper distribution of the materials necessary, as their acoustic performance may depend on their layout (Fratoni et al., 2023), relative humidity, and temperature (Cingolani et al., 2022).

Bio-based building materials seemed to be an efficient possibility for both types of comfort. Hence, apart from the low environmental impact and eco-friendly aspects, bio-wastes showed several benefits when incorporated into building products, namely sound absorption capacity (Martellotta et al., 2018; Othmani et al., 2017), thermal insulation performance (Muizniece and Blumberga, 2016; Platt et al., 2023), passive control of the hygrothermal indoor conditions and the air quality (Liuzzi et al., 2018), as well as mechanical resistance (Molari et al., 2017; Nadhari et al., 2019) to enable applicability. Past research already demonstrated that bio-wastes could have promising thermal insulation and acoustic absorption performance. Hence, they could improve hygrothermal and/or acoustic indoor comfort. For example, Trobiani Di Canto et al. (Trobiani Di Canto et al., 2023) employed wheat bran and the waste material banana peel to produce a thermal insulation board. Ricciardi et al. (Ricciardi et al., 2021) considered cork scraps, rice husk, and coffee chaff as bio-wastes and reported their promising thermal insulation performance. Martellotta et al. (Martellotta et al., 2018) demonstrated that wastes deriving from olive pruning could produce a composite with interesting acoustic behaviour. Tang et al. (Tang et al., 2018) evaluated the acoustic properties of corn husk and concluded that it had good acoustic absorption performance, appropriate for noise reduction.

However, bio-waste-based building composites also have some drawbacks, such as high hygroscopicity and low resistance to water and fire (Bakatovich et al., 2022), which can cause their degradation. Moreover, the presence of nutrients and the organic composition may increase the susceptibility to biological attack, a severe hazard both for materials and users (Ginestet et al., 2020) since it can lower composites' performance, degrade indoor air quality, and cause human diseases (Echeverria et al., 2021; Stefanowski et al., 2017). This drawback can be moderated or prevented by adding additives, physical or chemical treatments, the employment of specific strategies, and constant monitoring (Chastre et al., 2023; de Carvalho et al., 2020; Dennis et al., 2021).

Starting from this knowledge, the present work investigates the feasibility of producing a bio-based composite (i.e., bio-based board), for internal use that could improve indoor comfort (hygrothermal and/or acoustic). Unconventional materials not harmful to human health were selected, considering both the benefits and drawbacks of bio-based composites (Cintura et al. 2023a). The bio-resource guaranteed the bio-based aspect. Among the many benefits, the selected adhesive moderated some of the bio-based products' weaknesses.

*Arundo donax* L. (commonly known as giant reed or giant cane) is an herbaceous, perennial, and non-food crop, native to Asia but invasive in many regions of the world. It consists of stems 4 – 6 m high, diameters between 2 – 3 cm, and 2 – 3 mm thick. Due to its high

tolerance to different climates and soil conditions, it can be found worldwide, mainly in southern Europe (Caponetto et al., 2023; Jámbor and Török, 2019; Molari et al., 2021). The annual crop yield depends on several factors, such as the soil, climate, use of fertilisation, and irrigation. Hence, past studies that quantified this information reported different results. Ceotto et al. (Ceotto et al., 2021) carried out a three-year experiment and achieved an annual dry matter yield of up to 51.4 Mg DM ha<sup>-1</sup> (higher in the case of a harvesting treatment). Furthermore, the researchers reported a medium-term (6 years) and a long-term experiment (12 years) that achieved a dry matter yield of about 39.6 Mg ha<sup>-1</sup> and 37.7 Mg ha<sup>-1</sup>, respectively. Alexopoulou et al. (Alexopoulou et al., 2015) evaluated *A. donax*'s long-term productivity (11 to 22 years) in the Mediterranean South area. The researchers achieved a mean annual biomass production of 15.7 Mg DM ha<sup>-1</sup> of *A. donax*. Considering long-term production, a value of 13.6 Mg DM ha<sup>-1</sup> year<sup>-1</sup>. Danelli et al. (Danelli et al., 2021) reported that past research achieved yields of up to 20 Mg DM ha<sup>-1</sup>.

Its wide availability and properties made *A. donax* employed for several purposes, such as producing musical instruments for the paper industry, as a source of biomass, biofuel, and building practices (Malheiro et al., 2021). As for this latter, it was already employed for panels, reinforcement for concrete and plasters, support for roofs and coverings, or in ceilings. It contributed to thermal insulation and promising mechanical performance (Barreca, 2012; Malheiro et al., 2021). Molari et al. (Molari et al., 2021) provided a mechanical characterization of *A. donax*. They demonstrated that it could be employed as a structural material, although it has not commonly been used as such. Carneiro et al. (Carneiro et al., 2016) developed and characterized a new construction technology, based on the cob earth technology by turning it lightweight (known as *ReedCob*) by using mainly *A. donax* and earth. The researchers demonstrated the feasibility of using this construction system. They reported its several benefits: low density and high thermal insulation performance, high mechanical flexural strength, and easy and fast production. Fiore et al. (Fiore et al., 2014) analysed the properties of *A. donax* and concluded that it could be used as reinforcement in polymer composites. García-Ortuño et al. (García-Ortuño et al., 2011) demonstrated the feasibility of producing particleboards by using shredded *A. donax* particles as aggregates and urea formaldehyde resin as adhesive.

As the knowledge of the physical and chemical properties of raw materials could help to understand the performance of the final composites better (Cintura et al., 2021; Viel et al., 2018), some properties of *A. donax* available from previous studies (Fiore et al., 2014; Molari et al., 2021) are reported in Table 4.4-1. The differences between the values are expected to be

derived from the employed test methods and conditions (e.g., regions of growing, test setup, sizes of samples, laboratory conditions, and samples' moisture content).

Table 4.4-1. Physical and chemical properties of *A. donax* reported in past studies (Fiore et al., 2014; García-Ortuño et al., 2011; Malheiro et al., 2021; Molari et al., 2021).

| Property                                        | Values                     | References                   |
|-------------------------------------------------|----------------------------|------------------------------|
| Real density [kg/m <sup>3</sup> ]               | 1168 ± 3                   | (Fiore et al., 2014)         |
| Bulk density [kg/m <sup>3</sup> ]               | 893                        | (Fiore et al., 2014)         |
| Apparent density (C-shape) [kg/m <sup>3</sup> ] | 577                        | (Molari et al., 2021)        |
|                                                 | 476-524                    | (Malheiro et al., 2021)      |
|                                                 | 583                        | (Malheiro et al., 2021)      |
| Cellulose content [%]                           | 43.59                      | (Fiore et al., 2014)         |
| Hemicellulose content [%]                       | 20.5                       | (Fiore et al., 2014)         |
|                                                 | 28.48 - 32.03 <sup>a</sup> | (García-Ortuño et al., 2011) |
| Lignin content [%]                              | 17.2                       | (Fiore et al., 2014)         |
|                                                 | 17.70 - 21.31 <sup>a</sup> | (García-Ortuño et al., 2011) |
| Ash [%]                                         | 1.9                        | (Fiore et al., 2014)         |
|                                                 | 3.00 - 6.14 <sup>a</sup>   | (García-Ortuño et al., 2011) |
| Thermal conductivity [W/(m.K)]                  | 0.06                       | (Bakatovich et al., 2022)    |
| Tensile strength [N/mm <sup>2</sup> ]           | 248                        | (Fiore et al., 2014)         |
|                                                 | 103.7 - 127.4 <sup>b</sup> | (Molari et al., 2021)        |
| Young's modulus                                 | - 15.3                     | (Molari et al., 2021)        |
| Tension [N/mm <sup>2</sup> ]                    |                            |                              |
| Compressive strength [N/mm <sup>2</sup> ]       | 57                         | (Molari et al., 2021)        |
| Young's modulus                                 | - 13.4                     | (Molari et al., 2021)        |
| Compression [N/mm <sup>2</sup> ]                |                            |                              |

Note: <sup>a</sup> The lowest and highest values are reported (without differentiating node and internode); <sup>b</sup> Values for samples with node and internode, respectively.

Considering the benefits of using giant reed and its promising performance, in the present work, it was employed as the aggregate (by shredding the stems in particles), though in the past it has been studied mainly as fibres. Indeed, according to past research related to bio-based composite, the employment of bio-wastes and bio-resources as particles/high-mass aggregates is not so commonly investigated (Faruk et al., 2012; Sanjay et al., 2018; Savio et al., 2022).

As for the adhesive, a sodium silicate solution was considered due to its several benefits, such as its bonding capacity and its non-toxic nature (Lee and Thole, 2018; Liuzzi et al., 2020a). Indeed, in addition to the sustainability and eco-efficient aspects, the non-harmfulness of the final composites must be guaranteed. This could be provided by avoiding dangerous products, such as formaldehyde-based adhesives which can be hazardous to human health (Lee et al.,

2020; Owodunni et al., 2020). Furthermore, the sodium silicate solution may improve the resistance to biological attack and chemical decomposition (Lee and Thole, 2018; Liuzzi et al., 2020a), which can shorten the service life of building materials (Jones et al., 2022). Finally, as reported before, the sodium silicate solution was selected to avoid high pressure in the production process and high temperatures for drying and curing to secure lower environmental impact (Cintura et al., 2023a). Nevertheless, some drawbacks were reported: the sodium silicate solution employed as an adhesive demonstrated high hygroscopicity and low bonding strength in wood-based composites (Lee and Thole, 2018; Song et al., 2021).

The production process and the experimental campaign were described in a previous study (Cintura et al., 2023c) characterizing a hazelnut shell-sodium silicate solution composite. This enabled the comparison between different bio-wastes/bio-resources employed as aggregates with the sodium silicate solution as the adhesive. As Bakatovich et al. (Bakatovich et al., 2022) investigated a similar giant reed-based composite but with some differences in the binder and different aggregate grain sizes, this work was considered to compare the results.

Hygrothermal, acoustic, and mechanical properties were evaluated to provide a general characterization of the produced boards, and this contributed to defining the most promising applications. The work aimed at demonstrating the feasibility of producing a bio-based board for indoor application to both lowering the environmental impact and improve indoor comfort. Furthermore, it intended to describe a methodology for a wide-range analysis of unconventional building composites, which could be useful for future research.

## **4.4.2 Materials and methods**

### **4.4.2.1 Materials**

#### **4.4.2.1.1 Giant reed**

Giant reed was collected from a cultivation of the Department of Agricultural and Food Sciences, of the University of Bologna, in Cadriano, Bologna, Italy. It was harvested and air-dried in the Laboratory at room temperature for one year. The reeds were cut to have grain sizes mainly between 4 mm and 8 mm to secure the feasibility of producing boards (Cintura et al., 2023b), and using aggregates with similar particle size distribution as the reference study (Cintura et al., 2023c), even if differing for the elongated shape. Giant reed was then dried at  $T = 60\text{ }^{\circ}\text{C}$  until constant mass (variation in mass less than 0.1% after 24 h) and characterized according to the recommendation of the RILEM Technical Committee 236-BBM “Bio-aggregate-based building materials” (Amziane et al., 2017). The initial water content was  $(6.43 \pm$



0.08) %, and the loose bulk density was  $(181.3 \pm 6.3) \text{ kg/m}^3$ . Figure 4.4-1 shows the particle size distribution and the employed giant reed. Other properties of *A. donax* have been previously reported in the literature (Table 4.4-1).

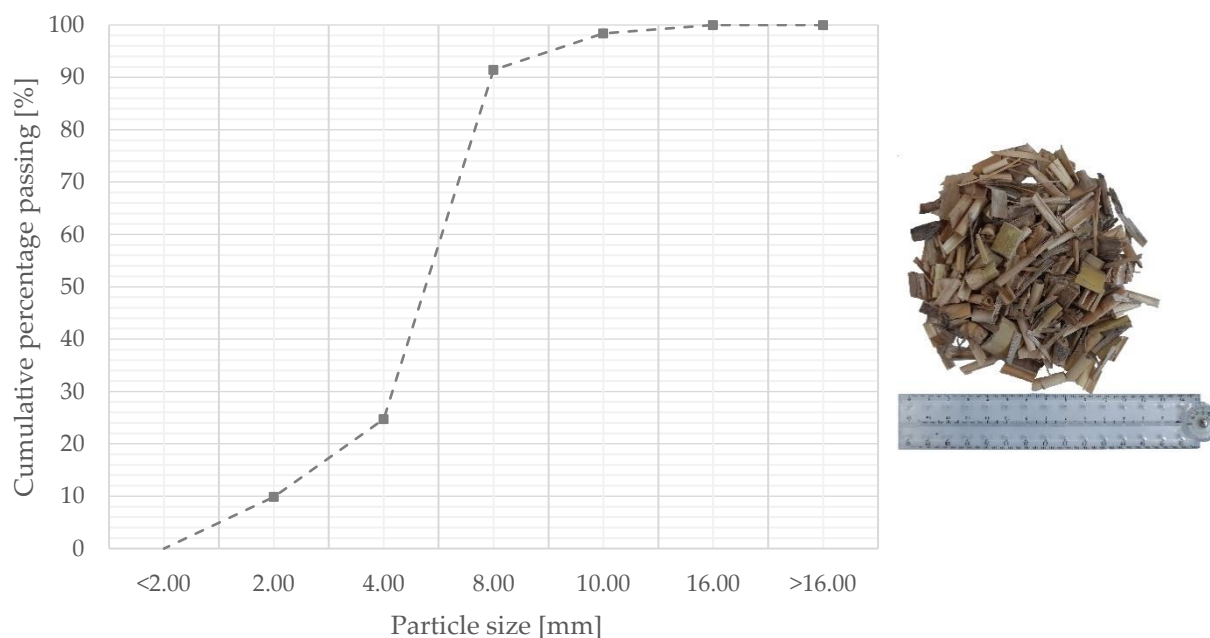


Figure 4.4-1. Particle size distribution and the employed giant reed.

#### 4.4.2.1.2 Sodium silicate solution

As for the adhesive, a sodium silicate solution,  $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ , provided by Ingessil Srl, Montorio (VR), Italy, was employed coherently with the previously described aims, hence considering the reference study (Cintura et al., 2023c). Table 4.4-2 summarizes its properties.

Table 4.4-2. Properties of the employed sodium silicate solution provided by Ingessil Srl (Ingessil).

| Property                                           | Value |
|----------------------------------------------------|-------|
| Weight ratio [-]                                   | 2.40  |
| Density [ $^{\circ}\text{Bé}$ ]                    | 46.45 |
| Molar ratio [-]                                    | 2.48  |
| Sodium silicate concentration [% p/p]              | 41.33 |
| $\text{SiO}_2$ [% p/p]                             | 29.17 |
| $\text{Na}_2\text{O}$ [% p/p]                      | 12.16 |
| Density [g/ml] at $T = 20\text{ }^{\circ}\text{C}$ | 1.471 |
| pH [-] at $T = 20\text{ }^{\circ}\text{C}$         | 12.40 |

#### 4.4.2.2 Samples preparation

The mix design and the production process were selected considering the reference past studies (Cintura et al., 2023b, 2023c), that evaluated the feasibility of producing composite boards made of hazelnut shells as high-mass bio-aggregates and sodium silicate solution as the adhesive. The mix design and the production process were defined after several practical tests to obtain the most sustainable building composite, maximizing the use of hazelnut shells, minimizing the amount of sodium silicate solution, and guaranteeing samples' production. Indeed, fewer quantities of adhesive did not allow sufficient mechanical resistance. Both the production process and the materials' quantities were kept the same for easier comparison between the results, as previously reported, ensuring that the type of bio-aggregates was the only different parameter. Hence, the percentages (by volume) of giant reed and sodium silicate solution were 70%–30%, respectively.

The aggregates and the adhesive were mechanically mixed for 10 min until homogeneity (Figure 4.4-2a) and then placed into silicon or wooden moulds (thickness = 4 cm), without being compacted, and levelled by using a spatula (Figures 4.4-2b and 4.4-2c) to have a horizontal surface. The 4 cm thickness was selected as it was suitable for both thermal and acoustic boards. Furthermore, it was a good compromise for the feasibility of production and the implementation of the tests, and its representativeness, considering the size of the bio-aggregates used. Lower thickness could also determine problems related to samples' resistance, while a higher one could determine composites too heavy to be easily analysed. The moulds were closed by fixing a wooden top (Figure 4.4-2d) and put at  $T = 60\text{ }^{\circ}\text{C}$  for 3 h, then air-dried, constantly rotated every 30 min (for half a day) to secure a homogeneous distribution of the sodium silicate solution. After 2 days, the samples were demoulded, cured at laboratory conditions for 28 days, and dried at  $T = 50\text{ }^{\circ}\text{C}$  until reaching a constant mass (considered as a variation in mass in 24 h less than 0.5%) to secure a complete drying (Liuzzi et al., 2020a, 2020b).

Figure 4.4-2e shows some rectangular and cylindrical composite samples (10 cm x 10 cm x 4 cm, and diameter = 10 cm and high = 4 cm, respectively).

#### 4.4.2.3 Test methods

All tests were performed after drying the samples at  $T = 50\text{ }^{\circ}\text{C}$  until reaching a constant mass (variation in mass in 24 h less than 0.5%), as previously described (Section 2.2). The evaluated properties, the number, designation, and sizes of the produced composite samples, and the relevant references are summarized in Table 4.4-3. Further details are described in the following sections.



Figure 4.4-2. Some steps of the production process of composite samples made of giant reed and the sodium silicate solution: a) mechanical mixing of the aggregates and the adhesive; b) moulding (wooden mould 50 cm x 50 cm x 4 cm) by using a spatula; c) cylindrical mould (diameter = 10 cm, high = 4 cm) with the mixture of giant reed and sodium silicate solution; d) closed moulds (10 cm x 10 cm x 4 cm); e) rectangular and cylindrical composite samples after demoulding and curing phase.

Table 4.4-3. Tests performed, number, designation, and dimensions of composite samples, and considered references.

| Properties           | Composite samples |                                                                                                                |                            | References                                      |
|----------------------|-------------------|----------------------------------------------------------------------------------------------------------------|----------------------------|-------------------------------------------------|
|                      | Number            | Designation                                                                                                    | Sizes [cm]<br>(high = 4cm) |                                                 |
| Visual observation   | All               | -                                                                                                              | Varied                     | (Cintura et al., 2023b)                         |
| Apparent density     | 6                 | A1, A2, A3, A4, A5, A6                                                                                         | 10 x 10                    | (EN 323, 1993)                                  |
| Thermal conductivity | 2                 | A10, A11                                                                                                       | 50 x 50                    | (EN 12667, 2001)                                |
| Hygroscopicity       | 6                 | A1, A2, A3, A4, A5, A6                                                                                         | 10 x 10                    | (ISO 24353, 2008)                               |
| Sound absorption     | 3                 | A7, A8, A9                                                                                                     | Diameter = 10              | (ISO 10534-2, 2001)                             |
| Air flow resistivity |                   | A7, A8, A9                                                                                                     | Diameter = 10              | (ISO 9053-2, 2020)                              |
| Flexural strength    | 6                 | A10_01, A10_02, A10_03,<br>A11_01, A11_02, A11_03                                                              | 50 x 10 (cut)              | (UNI-11842, 2021)<br>(ISO 14125 1998 +A1, 2011) |
| Compressive strength | 10                | A10_01_A, A10_01_B,<br>A10_02_A, A10_02_B,<br>A10_03_A, A11_01_A,<br>A11_01_B, A11_02_A,<br>A11_02_B, A11_03_A | 10 x 10 (cut)              | (EN-826, 2013)                                  |

#### 4.4.2.3.1 Visual observation and qualitative evaluation of the production process

The slow absorption of the sodium silicate solution by the aggregates could result in non-perfectly uniform samples, required for a correct evaluation of the properties of the composite. The samples were constantly monitored during drying and curing time to avoid this drawback. The visual analysis provided a qualitative evaluation of the correctness of the production process. Furthermore, qualitative considerations by visual checks after each test were made.

#### 4.4.2.3.2 Apparent density and thermal conductivity

The apparent density was measured by adapting EN 323 (EN 323:1993), hence by weighting three samples (stabilized at  $T = 50\text{ }^{\circ}\text{C}$ ) by an electronic balance Kern EW 2200–2NM, by determining their dimensions by a Fervi Digital Caliber, and finally by calculating the ratio between mass and volume (Figure 4.4-3a) and considering the average value and standard deviation as the representative value.



Figure 4.4-3. Main tests of the experimental campaign: a) evaluation of the apparent density of a giant reed-based composite sample; b) equipment to evaluate the thermal conductivity by the heat flow method; c) composite samples into the climate chamber during the hygroscopicity test; d) a cylindrical sample of giant reed into the impedance tube to evaluate the sound absorption capacity; e) giant reed-based sample (10 cm x 50 cm x 4 cm) in the Galdabini machine at the beginning of the bending test; f) giant reed-based sample (10 cm x 10 cm x 4 cm) before the performance of the compressive test.

The thermal performance can be evaluated through different methods, classified as steady state and transient methods (Posani et al., 2022). The steady-state methods analyse the thermal performance in equilibrium conditions, hence when temperature throughout the sample remains constant. Although the steady-state technique requires larger samples, more expensive

equipment, and more time than the transient method, it was selected to provide the most accurate results (Yüksel, 2016). Hence, thermal conductivity was determined by the heat flow method, according to EN 12667 (EN 12667:2001).

The samples 50 cm x 50 cm x 4 cm were stabilized at  $T = (20 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (60 \pm 5) \%$  and placed between two different plates (heated and cooled). Thermal conductivity was assessed by measuring the heat flow through the samples, namely the capacity of conducting heat, using the equipment described and validated by Baldinelli et al. (Baldinelli et al., 2019), and reported in Figure 4.4-3b. The samples' mass was evaluated before and after the test to ensure that moisture content remained constant, as this parameter could affect the final results (Liuzzi et al., 2017; Palumbo et al., 2016). The average values and the standard deviation of the thermal conductivity were considered.

#### 4.4.2.3.3 Hygroscopicity properties

The hygroscopicity of the composite was evaluated according to ISO 24353 (ISO 24353:2008). The laboratory test consists of subjecting the samples to different relative humidity levels, by measuring the variation in mass over time and determining the moisture sorption/desorption capacity. A moisture barrier, consisting of an adhesive aluminium foil that enabled the sealing, was applied all over the samples, except for a single surface corresponding to the top of the samples ( $A = 0.1 \text{ m}^2$ ). First, they were placed in a climate chamber (Climatest ARGOLab CH 250), preconditioned at  $T = (23 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (63 \pm 5) \%$  until reaching a constant mass (change in mass of less than or equal to 0.5% over 24 h) and weighed ( $m_0 = m_{d(n-1)}$ ). Then, the samples were conditioned at  $T = (23 \pm 2) ^\circ\text{C}$ ,  $\text{RH} = (75 \pm 2) \%$  for 12 h (sorption phase) and weighed every 3 h ( $m_{an}$ ), and at  $T = (23 \pm 2) ^\circ\text{C}$ ,  $\text{RH} = (50 \pm 2) \%$  for 12 h (desorption phase) and weighed ( $m_{dn}$ ) (Figure 4.4-3c), as required by ISO 24353 (ISO 24353:2008). This procedure was repeated four times (four cycles). Moisture sorption ( $\rho_{A,ac}$ ) and desorption ( $\rho_{A,dc}$ ) content, and moisture content difference ( $\rho_{A,sc}$ ) were determined according to Equations 4.4-1, 4.4-2, and 4.4-3, respectively.

$$\rho_{A,ac} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \frac{m_{an} - m_{d(n-1)}}{A} \quad (4.4-1)$$

$$\rho_{A,dc} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \frac{m_{an} - m_{dn}}{A} \quad (4.4-2)$$

$$\rho_{A,sc} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \rho_{A,ac} - \rho_{A,dc} \quad (4.4-3)$$

The moisture buffering value (MBV), which quantifies the amount of absorbed and released moisture considering cycling between two levels of RH (Ansell et al., 2020), was evaluated by

adapting and simplifying the method defined by Rode et al. (Rode et al., 2005), as the average value between MBV during the sorption phase ( $MBV_a$ ), described by Equation 4.4-4, and for the desorption ( $MBV_d$ ), calculated as reported in Equation 4.4-5. The values of the last three cycles were considered.

$$MBV_a \left[ \frac{g}{m^2 \% RH} \right] = \frac{m_{an} - m_{d(n-1)}}{A \times (RH_{high} - RH_{low})} \quad (4.4-4)$$

$$MBV_d \left[ \frac{g}{m^2 \% RH} \right] = \frac{m_{an} - m_{dn}}{A \times (RH_{high} - RH_{low})} \quad (4.4-5)$$

where  $RH_{high}$  is the highest value of RH (75%) and  $RH_{low}$  is the lowest one (50%). Although the conditions were different from the reference (Rode et al., 2005), namely different values of RH and different durations of the sorption/desorption cycles, the result provided a quantitative value to describe the moisture buffering capacity of the analysed composites and enabled a comparison with past studies.

#### 4.4.2.3.4 Acoustic properties

The sound absorption capacity can be evaluated by several techniques, divided into empirical models and inverse methods – i.e., considering physical properties strictly related to acoustic absorption, such as air flow resistivity, flexural and compressive strength – and direct measurements using an impedance tube (Othmani et al., 2017; Rey et al., 2012). This latter method is standardized and described by ISO 10534-2 (ISO 10534-2: 2001): a sample is placed at the end of a tube (Kundt's tube), where an acoustic excitation is generated, and acoustics pressure is registered by microphones. The value of the incidence absorption coefficient is determined by measuring the sound wave variations and using some corrective formulas. Although the direct measurement method provides only the normal incidence sound absorption and could be subjected to errors due to the placement of the samples in the tube (Martellotta et al., 2018), it was selected for its several advantages, such as the rapid evaluation, the possibility of using small samples and the easy test implementation (Rubino et al., 2023).

Three conditioned samples (diameter = 10 cm, high = 4 cm) at  $T = (20 \pm 1) ^\circ C$  and  $RH = (60 \pm 5) \%$ , were smoothed, placed at the end of a Plexiglas Kundt's tube, and sealed with plasticine to reduce the possible gaps between them and the tube (Figure 4.4-3d). An exponential sine sweep (Corredor-Bedoya et al., 2021) was generated by a loudspeaker and a signal amplifier (Samson 120 A with SNR = 96 dB) and converted to an analogue signal by the Digital to Analog Converters of the soundcard (RME 802). The signal was recorded by a single microphone 1/2" PCB and converted by the Analog-to-Digital Converter (sample rate of 44.1 kHz at 24-bit depth), and processed by ITA-Toolbox (Berzborn et al., 2017) through the Transfer-

Function Method (Chung and Blaser, 1980a, 1980b). The test was performed for each sample's front and back sides to avoid possible measurement errors caused by producing non-perfectly uniform samples.

#### 4.4.2.3.5 Air flow resistivity

The measurement of airflow resistance of porous and fibrous acoustical materials was standardised in ISO 9053-2 (ISO 9053-2:2020) and ASTM C-522-03 (ASTM C522-03:2015), which described the measurement equipment and the test procedure. The airflow resistance of a material is defined by Equation 4.4-6 as:

$$R \left[ \frac{\text{Pa} \times \text{s}}{\text{m}^3} \right] = \frac{\Delta p}{q_v} \quad (4.4-6)$$

where  $\Delta p$  [Pa] is the air pressure difference across the sample with respect to the atmosphere and  $q_v$  [ $\text{m}^3/\text{s}$ ] the air flow rate through the sample. One derived quantity of great interest in applied acoustic is the airflow resistivity,  $r$ , defined by Equation 4.4-7 as:

$$r \left[ \frac{\text{Pa} \times \text{s}}{\text{m}^2} \right] = \frac{\Delta p}{q_v} \times \frac{A}{d} \quad (4.4-7)$$

where  $A$  [ $\text{m}^2$ ] is the section of the tested sample and  $d$  [m] is the thickness of the tested sample (considered homogeneous) in the flow direction. In the latest version of ISO 9053-2 (ISO 9053-2:2020),  $\sigma$  was used to identify airflow resistivity. Still, in this article,  $\sigma$  denotes "Stress"; so the old notation  $r$  used in ISO 9053:1991 was considered to avoid misunderstanding.

The measurements of airflow resistivity were carried out on each sample three times by using the equipment described by Schiavi et al. (Schiavi et al., 2011). The same samples employed to evaluate the sound absorption capacity (diameter = 10 cm, high = 4 cm) were considered. Each sample was placed with the rougher side facing the environment and the flatter side facing the inside of the measurement tube.

#### 4.4.2.3.6 Flexural strength

The flexural properties were evaluated using a four-point bending test, adapting UNI 11842 (UNI-11842:2021) and ISO 14125 (ISO 14125:1998 +A1:2011). The samples used in the thermal analysis (A10 and A11) were cut into strips 10 cm x 50 cm x 4 cm. The designations, namely A10\_01, A10\_02, A10\_03, A11\_01, A11\_02, A11\_03, highlighted the considered original sample.

The six samples were conditioned at  $T = (20 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (60 \pm 5) \%$ , and tested at laboratory conditions in displacement control by using a Galdabini universal machine (maximum capacity of 100 kN) with a velocity of 0.5 mm/min. The supports' distance was 39 cm. Hence, the force was applied each 13 cm, as shown in Figure 4.4-3e. Some samples were placed with the rougher side as the lower one (samples ROU\_LOW) and the others in the opposite

way, namely with the rougher side as the upper one (samples ROU\_UP) to evaluate the bending performance for both faces. A linear displacement transducer, LVDT, HBM Model WA20mm, recorded the force and displacement during the test to determine the stress-strain curve defined by Equations 4.4-8 and 4.4-9, and the flexural strength  $\sigma_{\max}$  [N/mm<sup>2</sup>] of each sample calculated as the one corresponding to the maximum applied force,  $F_{\max}$  [N] (Equation 4.4-10).

$$\sigma_{(\text{intrados})} \left[ \frac{\text{N}}{\text{mm}^2} \right] = \frac{Fl}{bh^2} \quad (4.4-8)$$

$$\varepsilon_{(\text{intrados})} [-] = \frac{108}{23} \frac{h\delta}{l^2} \quad (4.4-9)$$

$$\sigma_{(\max)} \left[ \frac{\text{N}}{\text{mm}^2} \right] = \frac{F_{\max}l}{bh^2} \quad (4.4-10)$$

where  $F$  [N] is the applied force,  $l$  [mm] is the span of the sample,  $b$  [mm] and  $h$  [mm] are the width and thickness of the sample, respectively, and  $\delta$  [mm] is the displacement. The test was carried out until reaching the breaking load, or the maximum displacement that the LVDT could register. The measurements were carried out for each sample; therefore, the average values and standard deviation were considered. The modulus of elasticity,  $E$ , was calculated as described in Equation 4.4-11, by considering the values of  $\sigma$  and  $\varepsilon$  at 20% and 60% of the maximum applied load,  $F_{\max}$ , (UNI-11842, 2021):

$$E \left[ \frac{\text{N}}{\text{m}^2} \right] = \frac{\Delta\sigma}{\Delta\varepsilon} = \frac{\sigma_{60\%} - \sigma_{20\%}}{\varepsilon_{60\%} - \varepsilon_{20\%}} \quad (4.4-11)$$

#### 4.4.2.3.7 Compressive strength

Compressive behaviour was determined by adapting EN 826 (EN-826:2013). The undamaged extremities of the strips used for the bending test were cut to produce samples 10 cm x 10 cm x 4 cm, as reported in Table 4.4-3. The designation highlighted the original sample, which was employed first for thermal analysis, and then for bending test. The rectangular samples were placed between the two plates in the Galdabini machine (maximum capacity of 100 kN), with a joint of 11.65 kg, and compressed at a velocity of 2 mm/min (Figure 4.4-3f).

Equations 4.4-12 and 4.4-13 describe the relation to determine the stress-strain curve, considering  $F$  [N] as the applied force and  $\delta$  [mm] the displacement during the test, both recorded by the LVDT (HBM Model WA20mm),  $A_0$  [mm<sup>2</sup>] the initial cross-sectional area of the samples, and  $d_0$  [mm] the initial thickness of the samples.

$$\sigma \left[ \frac{\text{N}}{\text{m}^2} \right] = \frac{F}{A_0} \quad (4.4-12)$$



$$\varepsilon [-] = \frac{\delta}{d_0} \quad (4.4-13)$$

Compressive strength was determined by considering the average and standard deviation of the compressive stress corresponding to the breaking load or 10 % strain (thus,  $\varepsilon = 0.1$  if the samples did not achieve rupture), as defined by EN 826 (EN-826:2013). The modulus of elasticity was determined by Equation 4.4-11, considering the  $\sigma$  and  $\varepsilon$  at 20% and 60% of the compressive strength.

### 4.4.3 Results

#### 4.4.3.1 Visual observation and qualitative evaluation of the production process

All the samples seemed sufficiently uniform at the end of the curing time. However, some seemed to have a higher amount of sodium silicate on one side (the base of the samples). As this possible problem had been foreseen, more than the minimum recommended number of composite samples were produced and tested (e.g., for the apparent density and the hygroscopicity test six samples were considered) and laboratory tests were performed for both sides of the samples whenever necessary, as described in Section 4.4-2.3.

No biological colonization was visually observed, neither after the hygroscopicity tests, namely after placing the samples at RH = 75% for 12 h, four times.

#### 4.4.3.2 Apparent density and thermal conductivity

Table 4.4-4 reports the apparent density values at  $T = 50\text{ }^{\circ}\text{C}$ , thermal conductivity at  $T = 20\text{ }^{\circ}\text{C}$ , and RH = 60 %, the average values for both, and the standard deviation (S.D.) for the apparent density.

The variations among the samples could have occurred due to the production process, namely due to the uncontrolled distribution of different grain sizes aggregates, and the sodium silicate solution. The samples could have some differences in aggregate sizes, internal porosity, and voids' volume. All these possibilities were reported by the standard deviation, which was low if related to the average value. Hence, a good uniformity between the tested samples was guaranteed.

The mass variation of the samples employed to evaluate the thermal performance (weighed before and after the test) was less than 0.5%. The moisture content was considered constant.

Table 4.4-4. Results of apparent density at  $T = 50\text{ }^{\circ}\text{C}$ , thermal conductivity at  $T = 20\text{ }^{\circ}\text{C}$ , RH = 60% for each tested composite sample, average value, and standard deviation (S.D.).

| Composite samples  | Apparent density<br>[kg/m <sup>3</sup> ] | Thermal conductivity<br>[W/(m.K)] |
|--------------------|------------------------------------------|-----------------------------------|
| A1                 | 434                                      | -                                 |
| A2                 | 518                                      | -                                 |
| A3                 | 642                                      | -                                 |
| A4                 | 493                                      | -                                 |
| A5                 | 518                                      | -                                 |
| A6                 | 499                                      | -                                 |
| A10                | -                                        | 0.123                             |
| A11                | -                                        | 0.132                             |
| Average $\pm$ S.D. | 517 $\pm$ 68                             | 0.128                             |

#### 4.4.3.3 Hygroscopicity and moisture buffering value

Figure 4.4-4 shows the moisture content along the time for the four adsorption/desorption cycles.

The composite demonstrated high hygroscopicity, with moisture sorption content greater than moisture desorption for all cycles, being  $p_{(A,ac)} = 0.13\text{ kg/m}^2$  and  $p_{(A,dc)} = 0.08\text{ kg/m}^2$  at the beginning of the test, and  $p_{(A,ac)} = 0.14\text{ kg/m}^2$  and  $p_{(A,dc)} = 0.08\text{ kg/m}^2$  after the four cycles (average values of the composite samples). The moisture content difference was almost the same for all cycles, between  $0.04\text{ kg/m}^2$  and  $0.07\text{ kg/m}^2$  (average values). The differences between the samples might derive from a different distribution of the sodium silicate solution in the exposed surface, which could determine different moisture absorption capacities. The moisture storage capacity was more significant than the release one during the test; the composite samples were so hygroscopic that they did not stabilise after each of the four cycles.

As described in Section 4.4.2.3, the MBV was calculated and rated considering the Nordtest methodology (Rode et al., 2005), even if this defined the practical MBV by considering different conditions: 8 h-sorption phase and 16 h-desorption phase at RH = 75 % and RH = 33 %, respectively. In the present work, the MBV was calculated according to the sorption/desorption phases' conditions described in ISO 24353 (ISO 24353:2008). This could determine some differences in the results. Hence, this information should be considered for comparisons and discussions. The MBV was  $(4.33 \pm 1.84)\text{ g/(m}^2\text{ \%RH)}$  (average value of the six tested samples) and the giant reed-based composite can be rated as "Excellent" (MBV > 2).

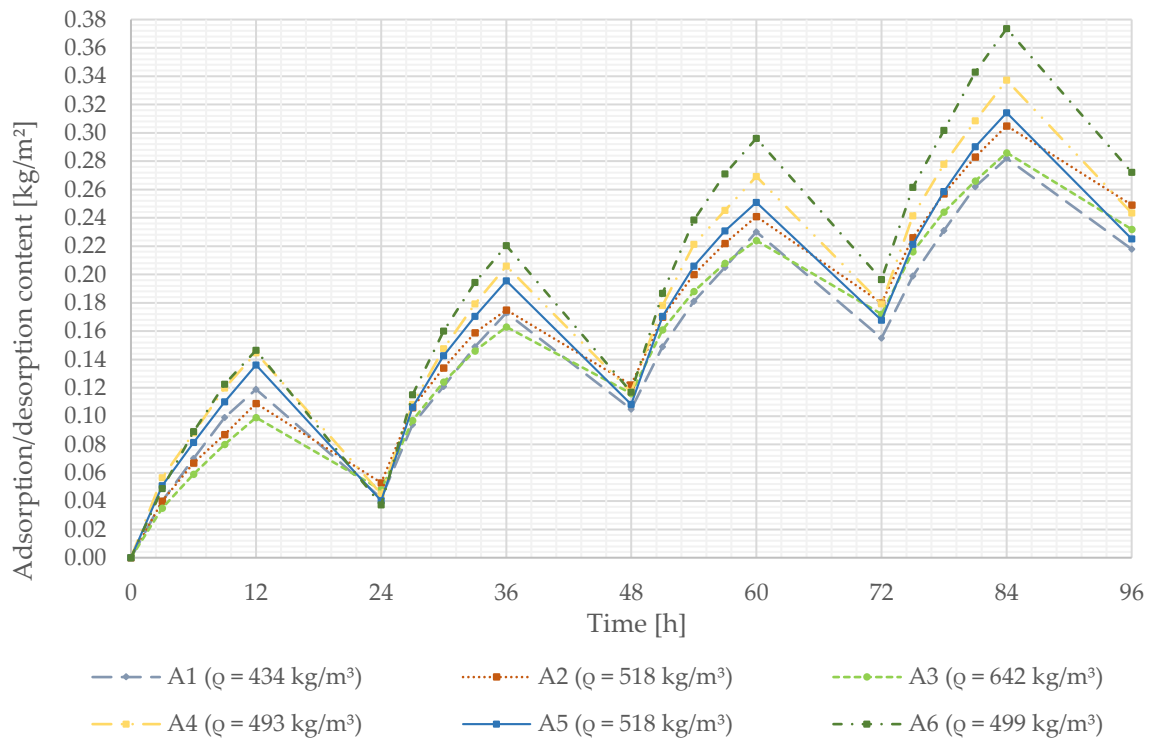


Figure 4.4-4. Moisture adsorption/desorption for four cycles of the tested composite samples.

#### 4.4.3.4 Acoustic properties

Figure 4.4-5 reports the results of the acoustic absorption coefficient evaluated for both sides of the samples: the dotted lines represent the results for the rougher ones (samples A). In contrast, the continuous lines represent the results for the flatter sides (samples B).

The acoustic absorption behaviour was the same for all samples with differences in the absorbed frequencies, especially for two of them, which had the exposed sides as the flatter ones and absorbed higher frequencies (almost 1100 Hz). The variations could be caused by the differences in the disposition of the aggregates, hence the distributions of internal voids. Indeed, as de Carvalho et al. (de Carvalho et al., 2020) reported, aggregates' particle sizes and distribution could widely influence the results. At frequencies between 600 Hz and 700 Hz, the sound absorption coefficient,  $\alpha$ , was almost 1; the curve reached a peak, decreased, and then slightly increased again.

The most common acoustic absorption behaviours are due to porous materials, membrane absorbers, and resonators. Porous materials are widely used for their great acoustic performance (Koizumi et al., 2002), provided by the porosity: when a sound wave strikes the material's surface, it causes the vibration of the internal air molecules transforming the sound into energy (viscous and dissipation phenomena). Porous absorbers usually work at high frequencies and their performance varies depending on the thickness (Selvaraj et al., 2019). Due to the

resonance phenomenon, the membrane absorbers and resonant absorbers can absorb lower frequencies. At specific frequencies, the absorbers can freely vibrate (membrane absorbers) or the interaction between the sound wave and the air molecules inside the voids acts as a spring-mass system (resonant absorbers). In this latter case, the vibration enabled the dissipation of the acoustic wave, even in more complex configurations (Cingolani et al., 2021).

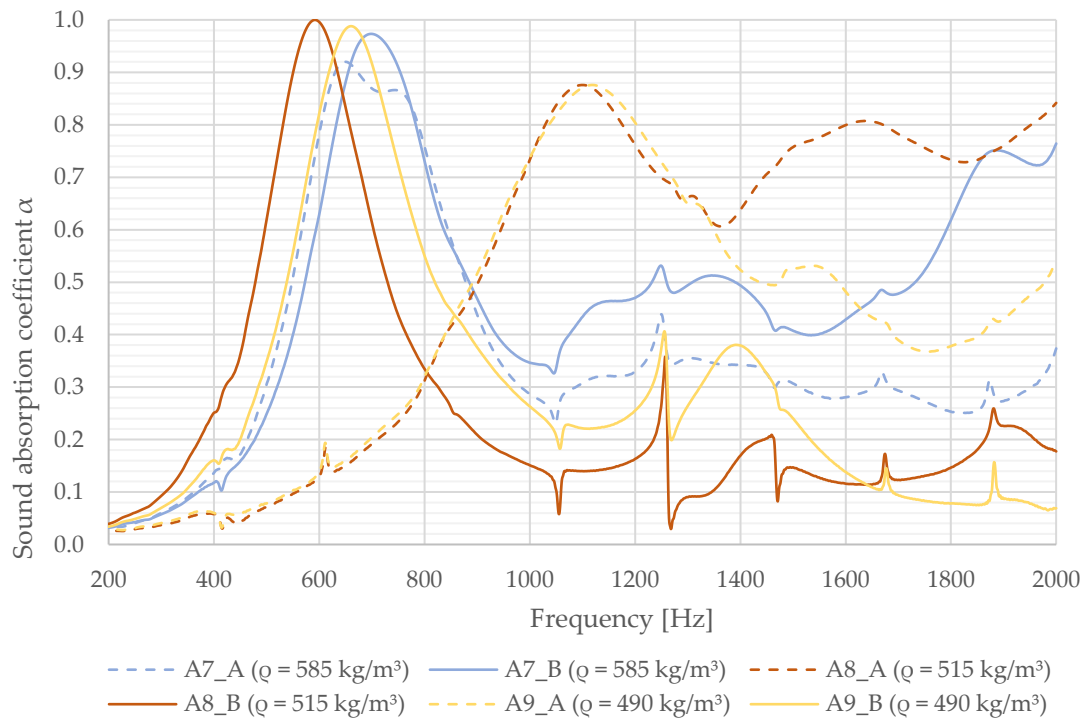


Figure 4.4-5. Sound absorption coefficients of both sides of the three tested samples. Continuous lines represent the flatter exposed surfaces' results and the rougher ones' dotted lines.

The giant reed-based composite showed a less common acoustic-absorption behaviour, and it could be comparable to granular material (Horoshenkov and Swift, 2001). Granular materials may be considered a particular case of porous materials, and their first peak of normal incidence sound absorption,  $f_0$ , can be described by Equation 4.4-14:

$$f_0 = \frac{1}{2\pi} \sqrt{\frac{k}{M_{\text{eff}}}} \quad (4.4-14)$$

where  $M_{\text{eff}}$  is the effective mass of granular material and  $k$  is the apparent dynamic longitudinal elastic modulus, detailed in past studies (Horoshenkov and Swift, 2001; Voronina and Horoshenkov, 2004). After the first peak, the composite behaviour was closer to the typical porous materials, with a slight increase in the sound absorption curve.

#### 4.4.3.5 Air flow resistivity

Table 4.4-5 reports the results of air flow resistivity and the extended uncertainty for the three tested composite samples. As previously described, the test was carried out three times for each sample, by placing the rougher side facing the environment and the flatter one facing the inside of the measurement tube.

Table 4.4-5. Results of air flow resistivity and the extended uncertainty for the three tested composite samples.

| Composite samples | Air flow resistivity<br>[kPa s/m <sup>2</sup> ] | Extended un-<br>certainty |
|-------------------|-------------------------------------------------|---------------------------|
| A7                | 23.69                                           | ± 0.57                    |
| A8                | 7.46                                            | ± 0.18                    |
| A9                | 18.33                                           | ± 0.55                    |

The extended uncertainty was good, namely under 3%, but the reproducibility was relatively low. Further analysis could better explain these results. For example, the internal distribution of the voids, namely the tortuosity and porosity. Nevertheless, this information was considered and provided to better understand the acoustic properties.

#### 4.4.3.6 Flexural strength

Figure 4.4-6 shows the stress-strain curve. The dotted lines represent the samples placed in the Galdabini machine with the rougher side as the lower one (samples ROU\_LOW); the continuous lines represent the samples with the rougher side as the upper one (samples ROU\_UP). Table 4.4-6 reports the maximum force and displacement, the flexural strength, and the modulus of elasticity (considering the average values and standard deviation).

The bending behaviour was reported for all samples until achieving the maximum load (100 kN), as previously described (Section 4.4.3.5). Indeed, none achieved the rupture, and the test was performed up to the maximum capacity set in the machine. Figure 4.4-7 shows four samples after the bending test.

The samples with the rougher side as the upper one (samples ROU\_UP), having as the stressed side by the flexural stress the one with more amount of sodium silicate, demonstrated greater bending behaviour and higher flexural strength (Table 4.4-6). For them, both the maximum force and the flexural strength increased by 67%, while the modulus of elasticity, increased by 44%. The elongated shape of the giant reed with higher quantities of sodium silicate solution guaranteed high cohesion, thus good mechanical resistance and increased flexural

strength. This could be an advantage for future applications as indoor coating insulation boards, although the applied load remained low if compared with structural composites.

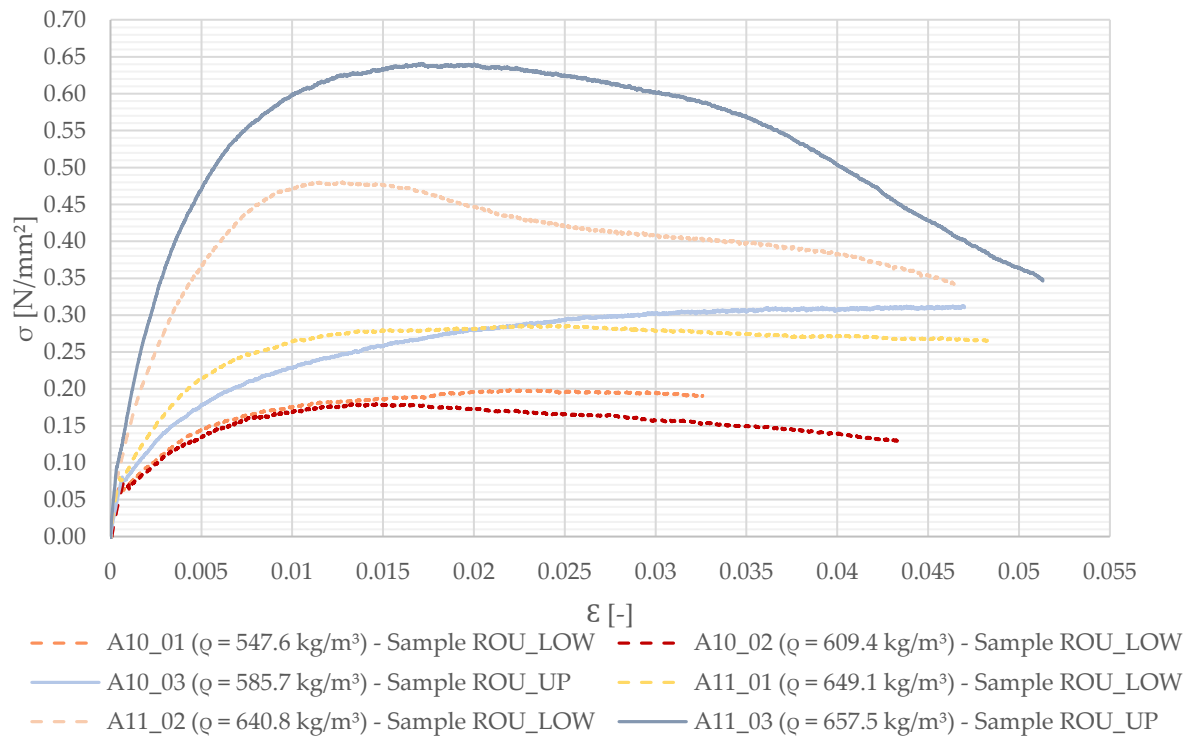


Figure 4.4-6. Bending behaviour of the tested samples. The dotted lines represent the samples placed in the Galdabini machine with the rougher side as the lower one (samples ROU\_LOW); the continuous lines represent the samples with the rougher side as the upper one (samples ROU\_UP).

Table 4.4-6. Average values and standard deviation of maximum force, displacement, flexural strength, and modulus of elasticity for the bending test.

|                        | $F_{\max}$<br>[N] | $\delta_{\max}$<br>[mm] | $\sigma_{\max}$<br>[N/mm <sup>2</sup> ] | $E_{(20\%-60\%)}$<br>[N/mm <sup>2</sup> ] |
|------------------------|-------------------|-------------------------|-----------------------------------------|-------------------------------------------|
| <b>Samples ROU_LOW</b> | 98.0 ± 43.6       | 16.3 ± 5.0              | 0.29 ± 0.14                             | 41.8 ± 21.8                               |
| <b>Samples ROU_UP</b>  | 148.7 ± 63.3      | 29.2 ± 18.4             | 0.48 ± 0.23                             | 60.4*                                     |
| <b>All</b>             | 114.9 ± 51.2      | 20.6 ± 11.3             | 0.35 ± 0.18                             | 48.0 ± 30.1                               |

Note: \*The S.D. was not calculated for Samples ROU\_UP because only two samples were considered and reported.



Figure 4.4-7. Four samples (10 cm x 50 cm x 4 cm) of giant reed and sodium silicate solution after the bending test.

#### 4.4.3.7 Compressive strength

Figure 4.4-8 reports the stress-strain graph, showing the compressive behaviour of the samples until 10% strain.

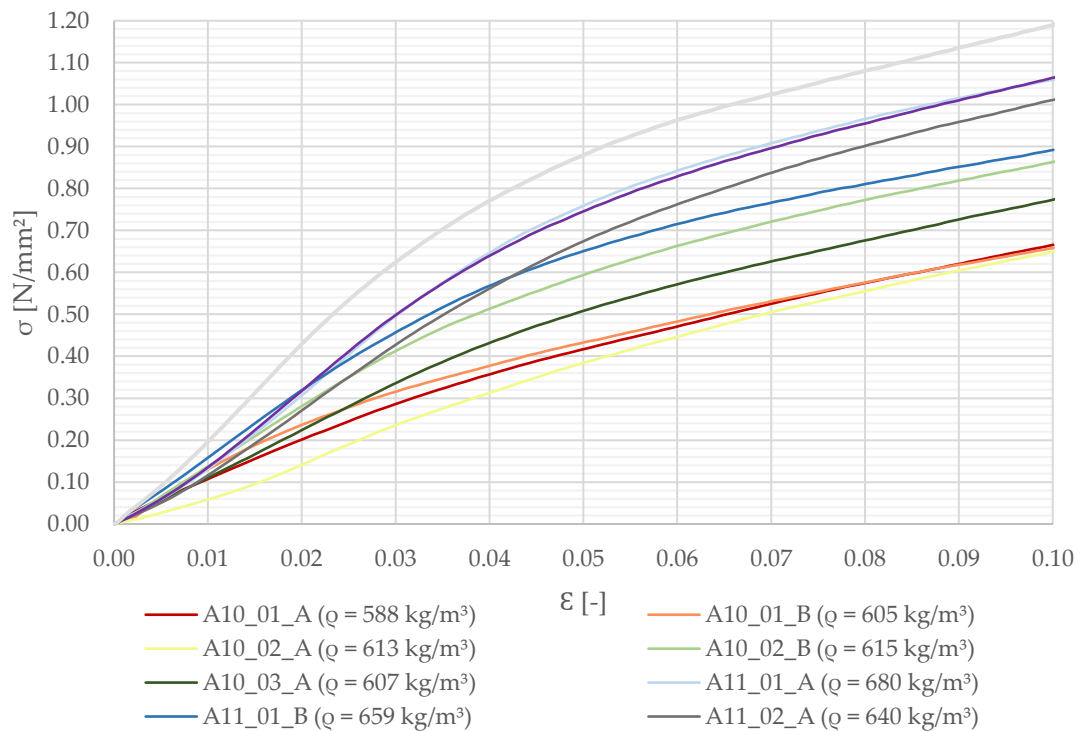


Figure 4.4-8. Compressive behaviour of the tested composite samples: stress-strain graphic until 10% strain.

Table 4.4-7 reports the average values and standard deviation of the force at 10% strain ( $F_{10}$ ) and the corresponding displacement ( $\delta_{10}$ ), the compressive strength ( $\sigma_{10}$ ), and the modulus of elasticity ( $E_{10}$ ), calculated as reported in Equation 4.4-9.

Table 4.4-7. Average values and standard deviation of the tested samples: maximum force and displacement, compressive strength, and modulus of elasticity.

| $F_{10}$<br>[N] | $\delta_{10}$<br>[mm] | $\sigma_{10}$<br>[N/mm <sup>2</sup> ] | $E_{(20\%-60\%)}$<br>[N/mm <sup>2</sup> ] |
|-----------------|-----------------------|---------------------------------------|-------------------------------------------|
| 8932 ± 1929     | 3.7 ± 0.1             | 0.9 ± 0.2                             | 13.0 ± 4.3                                |

The compressive behaviour did not show a peak and then a collapse: the samples did not achieve the yield. The samples were pressed and flattened, and the test stopped at the set maximum load (100 kN). For this reason, according to ISO 826 (EN-826:2013), compressive strength and the corresponding force were determined by considering 10% strain ( $\epsilon = 0.1$ ), as described in Section 4.4.2.3.

Figure 4.4-9 shows the stress-strain curve, considering all the compressive test performance times.

The compressive test results allowed making important considerations, both on materials' behaviour (giant reed, sodium silicate solution, and their mixture) and on feasible production processes, namely by pressing the composites, as detailed in the following section. The composite did not achieve the yield, but it flattened. Figure 4.4-10 reports an example of two tested samples, before and after the test.

## 4.4.4 Discussion

### 4.4.4.1 Comparison with past studies

The results of each property were compared with literature values and commercial products to validate and discuss them. It is essential to underline that the values considered for the comparison could be determined in different laboratory conditions, for different shape and size samples, and by other test methods. Although the present study evaluated the giant reed as a particle aggregate, bio-wastes and bio-resources employed as fibres and particles were considered. This enriched the considerations, showing how bio-based composites could result in building products with different properties.

Figure 4.4-11 shows the correlation between thermal conductivity and apparent density of the giant reed-based composite and some literature values, better detailed in Cintura et al. (Cintura et al., 2023a).



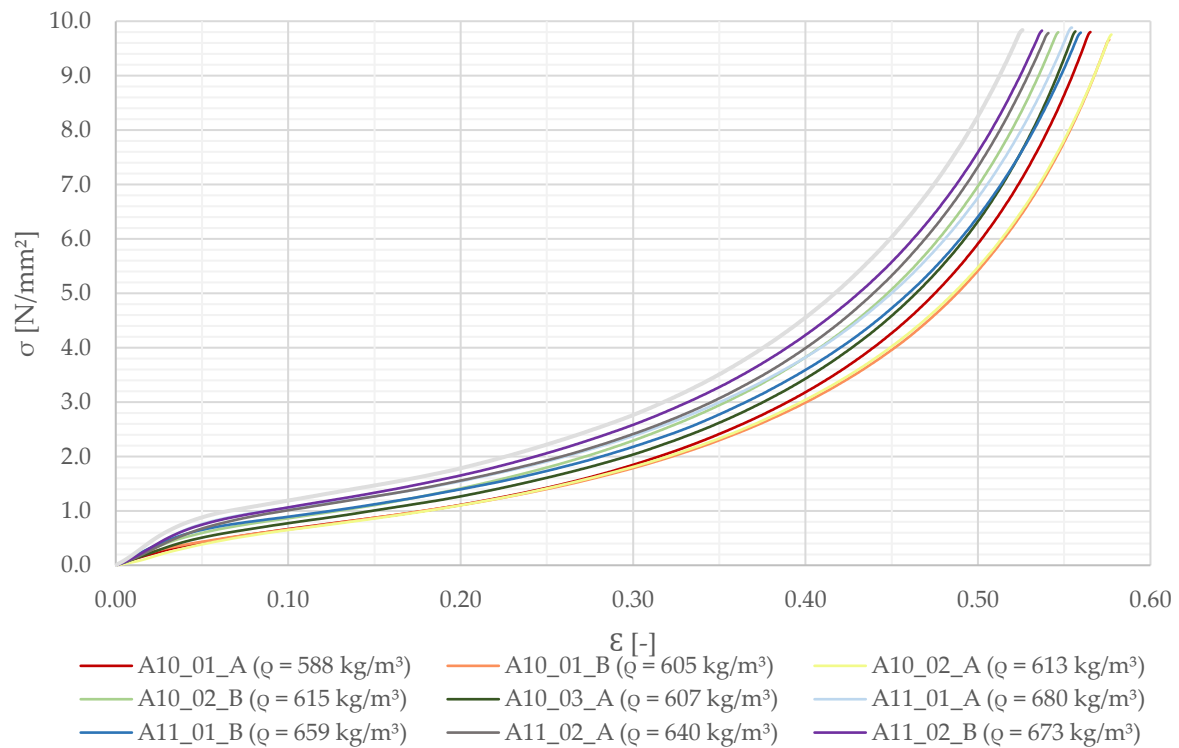


Figure 4.4-9. Compressive behaviour of the tested composite samples: stress-strain graphic until reaching the maximum capacity.



Figure 4.4-10. Example of two giant reed-based composite samples employed for the compressive test before (left) and after the compressive test (right).

The composite of giant reed and the sodium silicate solution showed values of apparent density higher than all the ones considered for comparison, except for the boards made of rice husk, earth, air lime, and hemi-hydrated gypsum (Antunes et al., 2019), boards made of wood waste, Portland cement, and lime (Park et al., 2019), and a composite of hazelnut shells and sodium silicate (produced with the same production process and percentages of aggregate-adhesive (Cintura et al., 2023c)). As for the thermal properties, the giant reed-based composite had values of thermal conductivity similar to the hazelnut shells-based one (Cintura et al.,

2023c), lower than rice husk-based boards (Antunes et al., 2019), and higher than the others considered composites.

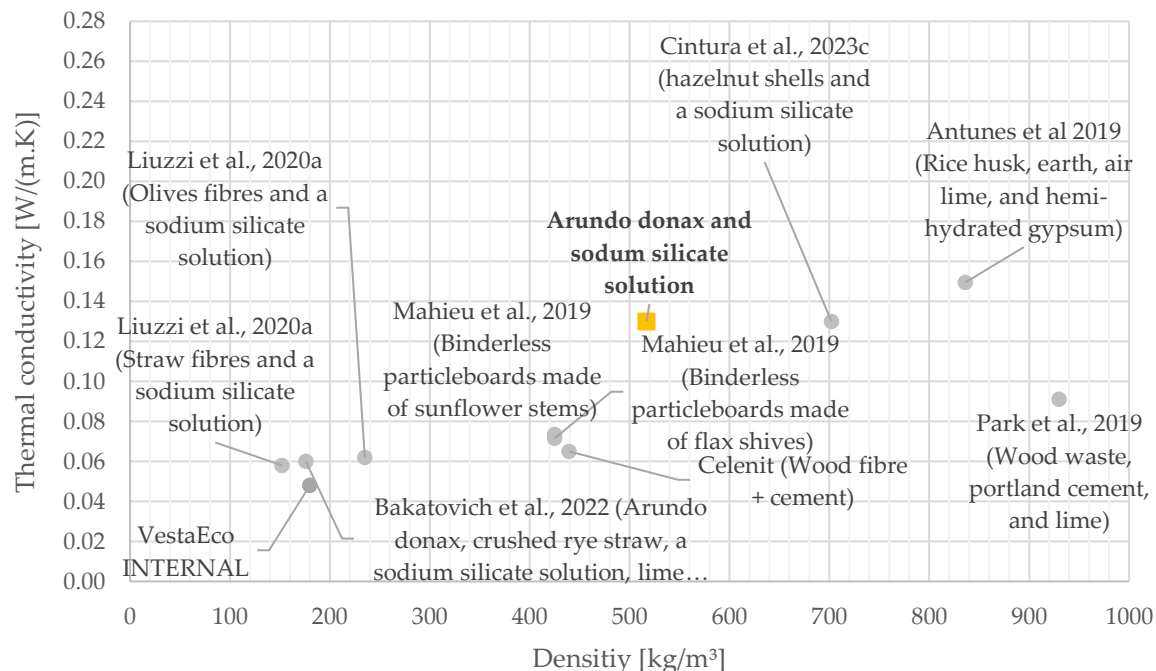


Figure 4.4-11. Correlation between thermal conductivity and apparent density of the studied composite compared with literature values and commercial products reported in (Cintura et al., 2023a).

As previously mentioned, it is essential to underline that, even if all the compared products were bio-based, some of them were produced by using fibres instead of particles. This resulted in lower density and better thermal insulation performance (Cintura et al., 2021; Panyakaew and Fotios, 2011).

The result of the comparison was in line with the expectation. The commercial products, namely thermal and acoustic insulation Celenit N boards made of mineralized wood wool and Portland cement (Celenit, 2022), and internal boards composed of lignocellulosic fibres and PMDI resin VestaEco (Vestaeco, 2022), were optimized to meet the requirements to be used as insulators. Mahieu et al. (Mahieu et al., 2019) did not use any binder. Hence, the final properties depended on the production process and the fibres (light materials with good thermal performance). Park et al. (Park et al., 2019) considered wood waste (light aggregate) and an optimized mix design. Antunes et al. (Antunes et al., 2019) achieved higher density and thermal conductivity values, probably due to the clay-based binding matrix. As for the composites with sodium silicate solution, Liuzzi et al. (Liuzzi et al., 2020a) considered both lighter aggregates and lower amounts of sodium silicate solution. Bakatovich et al. (Bakatovich et al., 2022) achieved lower

thermal conductivity values for *A. donax*-based composites bounded by sodium silicate with lime and gypsum thanks to the addition of ray straw. Indeed, by replacing the *A. donax* with straw, the thermal insulation capacity of the mixture of the two aggregates (without any binders) improved. Cintura et al. (Cintura et al., 2023c), achieved higher values of both density and thermal conductivity for the hazelnut shells-based composites, due to the selected aggregates, with higher loose bulk density –  $(469.3 \pm 5.8) \text{ kg/m}^3$  –.

Considering the reference values to define a material as a thermal insulator, namely  $0.065 \text{ W/(m.K)}$  (Pina dos Santos and Matias, 2006; Romano et al., 2019), even if some part studies reported  $0.1 \text{ W/(m.K)}$  (Martínez-García et al., 2020; Mati-Baouche et al., 2014), the composite cannot be considered as such, even if good thermal insulation properties were expected from giant reed (Barreca, 2012; Carneiro et al., 2016; Malheiro et al., 2021).

Figure 4.4-12 shows the comparison between the results of the hygroscopicity analysis for the giant reed-based composite and the hazelnut shells-based one (average values and standard deviation of the analysed samples), evaluated by Cintura et al. (Cintura et al., 2023c).

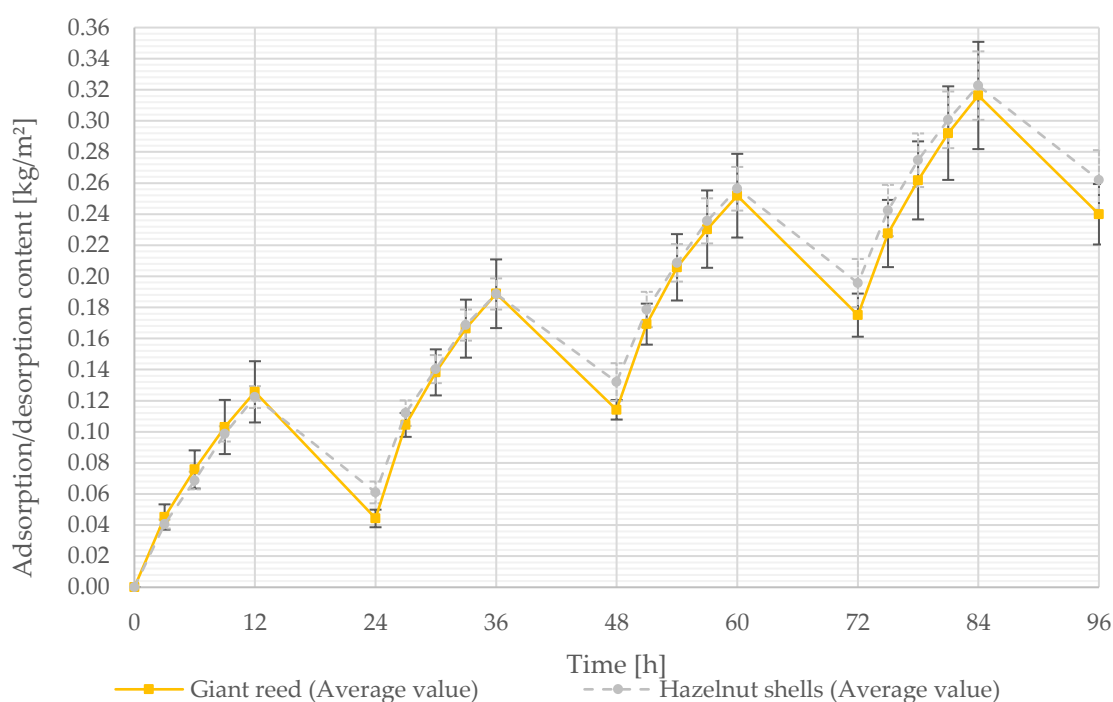


Figure 4.4-12. Moisture adsorption/desorption content for four cycles: comparison between the average value of the tested giant reed-based composite samples and hazelnut shell-based ones, evaluated by Cintura et al. (Cintura et al., 2023c)

The production process, aggregates-adhesive percentages, and the tested method of the two composites were the same. Hence, the differences derived from the bio-aggregates (physical

and chemical properties), and the different distribution of the sodium silicate solution in the adsorbing/desorbing surface. Figure 4.4-12 demonstrates that the giant reed-based composite had greater moisture storage capacity than the hazelnut shell-based one (considering the standard deviations). This could be caused by an excess sodium silicate solution on the giant reed-based samples' surface, determined by the employed method. However, the hygroscopicity behaviour was almost the same, as well as the moisture content. The maximum values of moisture content were  $\rho_{(A,ac)} = 0.14 \text{ kg/m}^2$  and  $\rho_{(A,dc)} = 0.08 \text{ kg/m}^2$  for the giant reed-based composite,  $\rho_{(A,ac)} = 0.12 \text{ kg/m}^2$  and  $\rho_{(A,dc)} = 0.06 \text{ kg/m}^2$  for the hazelnut shells-based. Both showed high hygroscopicity compared with a past study (Cintura et al., 2023a). This result also confirmed the sodium silicate solution's hygroscopic properties (Lee and Thole, 2018; Song et al., 2021).

The comparison of the MBV –  $(4.33 \pm 1.84) \text{ g/(m}^2 \text{ \%RH)}$  – with already cited past studies that employed the sodium silicate solution as the adhesive demonstrated that the results were coherent with the literature. Liuzzi et al. (Liuzzi et al., 2020a) reported values of the ideal MBV of  $5.05 \text{ g/(m}^2 \text{ \%RH)}$  for composites made up of straw fibres and sodium silicate solution, and  $3.29 \text{ g/(m}^2 \text{ \%RH)}$ , for olive fibres and the sodium silicate solution. Practical MBV and ideal MBV are similar in the case of homogeneous composites and if the penetration depth is less or equal to the samples' thickness (Rode et al., 2005). Cintura et al. (Cintura et al., 2023c) reported MBV of  $(3.45 \pm 1.76) \text{ g/(m}^2 \text{ \%RH)}$  for hazelnut shells and sodium silicate solution (calculated considering the same conditions as the present study).

Composites as similar as possible to giant reed-based ones were considered to compare and discuss the acoustic results, namely made of bio-wastes employed as particles rather than fibres. A composite made of olive pruning waste and chitosan dissolved in acetic acid and water as a binder had a sound absorption curve similar to the one of giant reed-based with a peak at higher frequencies (Martellotta et al., 2018). The same results were achieved for a composite made of sunflower stalks and chitosan (Mati-Baouche et al., 2016). Hazelnut shells-based boards with sodium silicate solution (Cintura et al., 2023c) showed the same acoustic absorption behaviour. The same adhesive and production process resulted in stiff samples with internal cavities. The main difference was the second slight increase of the curve after the first peak (Figure 4.4-5), in the case of the giant reed-based composite, more similar to porous materials. For the hazelnut shell-based composite the sound absorption coefficient achieved the higher values at frequencies between 700 Hz and 900 Hz, while for the giant reed-based one was between 600 Hz and 700 Hz. In both cases, this frequency range was in the one of the human voice, between 500 Hz and 1000 Hz (EN IEC 60268-16:2020; ISO 3382-3:2012). Further

analyses should be carried out since a combination of the two bio-aggregates may determine an increment of the amplitude of the peak, hence of the frequency range. The mixture of round-shaped aggregates (from hazelnut) with elongated ones and the sodium silicate solution could result in a sound absorber with even more advantageous behaviour.

Glé et al. (Glé et al., 2011) reported the acoustic absorption of hemp shiv analysed as raw material. They marked the influence of the particle size, concluding that the smaller ones, the better sound absorption, especially for low frequencies. The acoustic absorption curve was similar to the one of the giant reed-based composite. Indeed, the first peak in the absorption curve occurred around 600 Hz; then, a second peak at 1700 Hz. For the giant reed-based composite, the second peak was not so evident; in this case, the sound absorption curve increased. Kinnane et al. (Kinnane et al., 2016), reporting the sound absorption curve of the hemp shiv as raw materials, showed similar behaviour. The researchers highlighted the importance of the particles' compaction.

Even if these examples referred to the sound absorption performance of the raw materials, they provided helpful information for further analysis. Moreover, they demonstrated that the sound absorption performance of giant reed-based composite was probably due to aggregate shapes, compaction, and dispositions, which produced voids in the samples' surface and internal layer.

Finally, the comparison was carried out for the results of mechanical tests. Ng et al. (Ng et al., 2018) considered boards made up of shredded sugarcane bagasse and a sodium silicate solution in different percentages. The researchers reported higher flexural strength values, between 15.40 N/mm<sup>2</sup> and 21.19 N/mm<sup>2</sup>, probably due to the production process, which consisted of hot pressing and determined higher density. They concluded that a higher amount of sodium silicate improved the flexural strength. Savic et al. (Savic et al., 2020) investigated the properties of composites made of *Miscanthus x giganteus* fibres, mineral binders (based on cement and hydrated lime), and pozzolanic materials. They tested prismatic samples and reported flexural strength values between 0.11 N/mm<sup>2</sup> and 0.63 N/mm<sup>2</sup> (more similar to the ones achieved for giant reed), varying depending on the considered binder. Mathews et al. (Mathews et al., 2023) tested insulation panels made up of recycled cardboard as aggregates and different binders. They achieved values of flexural strength between 0.11 N/mm<sup>2</sup> and 0.36 N/mm<sup>2</sup> for lime-based binder and boric acid, and between 0.34 N/mm<sup>2</sup> and 0.91 N/mm<sup>2</sup> for clay-based one (again, more comparable to giant reed's ones).

The comparison with the reference past study using hazelnut shells as aggregates showed interesting results. The composite evaluated by Cintura et al. (Cintura et al., 2023c) had

a different behaviour: the strain-stress curve represented an initial elastic behaviour, then the achievement of a maximum load (peak). As all the other parameters were kept identical, the differences were caused by the physical and chemical characteristics of the aggregates and the physical properties derived from the mixture aggregates-adhesive. In addition to its greater mechanical properties (Carneiro et al., 2016; Molari et al., 2021), the elongated and flattened shape of the giant reed guaranteed more cohesion between the particles and the sodium silicate solution, which widely influenced the final performance (Ng et al., 2018). As for the numerical values, the results were similar, since for the hazelnut shells-based composite were  $\sigma_{\max} = (0.43 \pm 0.04) \text{ N/mm}^2$ , when the rougher side was the upper one,  $\sigma_{\max} = (0.35 \pm 0.04) \text{ N/mm}^2$  when it was the lower one, and  $\sigma_{\max} = (0.39 \pm 0.06) \text{ N/mm}^2$  as the average value of all the tested samples. For the giant reed-based composite, the sodium silicate solution's distribution influenced the flexural strength more than the hazelnut shells-based one.

As for the compressive behaviour, the composite made up of giant reeds showed a stress-strain curve similar to straw bales (Maraldi et al., 2017, 2016; Molari et al., 2017). This result was considered to select the compared reference studies. Barbieri et al. (Barbieri et al., 2020) regarded composites of hemp and wheat husk as aggregate and a mixture of lime putty and natural hydraulic lime as a binder (called lime hemp and lime wheat husk concrete, respectively). Lime wheat husk concrete showed a compressive behaviour similar to the giant reed-based composite and a compressive strength of  $(0.16 \pm 0.02) \text{ N/mm}^2$ . As for the lime hemp concrete, a value of  $(0.24 \pm 0.03) \text{ N/mm}^2$  was determined. Savic et al. (Savic et al., 2020) for prismatic samples of *Miscanthus x giganteus* fibres, mineral binders (based on cement and hydrated lime), and pozzolanic materials reported values between  $0.139 \text{ N/mm}^2$  and  $1.349 \text{ N/mm}^2$ , depending on the binder. Celenit N (Celenit, 2022) at 10% deformation demonstrated compressive stress higher or equal to  $0.2 \text{ N/mm}^2$ , for thickness between 15 mm and 40 mm. These comparisons revealed that the results were consistent with the literature. Moreover, although the giant reed-based composite did not show enough mechanical resistance to be employed as a structural material, it demonstrated promising performance, especially for compressive strength.

The compression test showed another feasible, maybe more efficient, production process: pressing the mixture of giant reed and sodium silicate solution. As a first characterization, the apparent density (average value of all the tested samples) was calculated before and after the test. Consistently with the expectation, it increased, by 19%. Further analysis should be carried out to determine how a pressing process could change the composite's final properties.

#### 4.4.4.2 Implications and explanations of the results

The analysis of the results and the comparison with past studies ensured essential considerations. First, the thermal analysis demonstrated the significant influence of the selected adhesive/binder on the final composite's performance (Cintura et al., 2023a). Although the promising thermal properties of the considered bio-aggregate, the composite did not result in a low thermal insulation product (assuming the standard requirement for these insulators). The influence of the adhesive was clear also in hygroscopicity, even if it is less influencing. In this case, the results were determined by both the giant reed and the sodium silicate. To further investigate the influence of each material, laboratory tests could be performed by considering them separately (i.e., the aggregates, the sodium silicate, and the composite).

The acoustic behaviour seemed to be determined mainly by the final configurations of the composite (stiffness with voids). Indeed, the results of the giant reed-based composite and the hazelnut shells-based one were highly similar. Past studies that evaluated raw materials' acoustic behaviour (Kinnane et al., 2016; Glé et al., 2011) suggested that the sound absorption curve of the aggregates (as raw materials, without any binders) might be similar. Thus, bonding them by keeping them in a similar configuration to the one they had as raw materials, might be the solution to have this acoustic performance. The present study ensured this by the sodium silicate solution, mixed without pressing. However, again, an analysis of the materials considered separately (i.e., aggregates, adhesive, and composite) would be helpful to support this consideration. Finally, the result of the air flow resistivity suggested that further analysis of the internal distribution of voids (porosity and tortuosity) might be performed. This analysis could be a starting point for future studies, focused on the deep investigation of acoustic properties.

The mechanical performance was provided by both the aggregate and the mixture aggregates-adhesive. In addition to the giant reed's excellent mechanical properties (Molari et al., 2021), the aggregates' elongated shape and the sodium silicate solution with the production process, ensured good cohesion. This aspect avoided the samples' yield and improved the mechanical resistance. Although the composite cannot be used for structural purposes, the results were extremely interesting, especially for the compressive strength. They outlined a production process that might produce a stronger and more mechanically performing composite.

#### 4.4.4.3 Benefits, drawbacks, and possible applications

The study demonstrated the several benefits of the analysed composite. First, the sustainable aspects, that were guaranteed by the selected materials. As past studies reported (Mahieu et al., 2019; Rubino et al., 2023), being *A. donax* an abundant, renewable, and perennial by-product, the energy consumption derived from the production process could be considered negligible. The sodium silicate solution may have a higher environmental impact. Still, it enabled the use of low temperatures during the casting and curing phases, lowering the energy consumption during the production process. Furthermore, confirming the information provided by past research (Lee and Thole, 2018; Liuzzi et al., 2020a), it moderated some of the main drawbacks of using bio-wastes, namely the resistance against biological attacks, avoiding any biological colonization (even after the hygroscopic tests). Finally, the sodium silicate solution was considered a non-toxic adhesive, not harmful to human health. These qualitative considerations indicate that various aspects made the composite eco-efficient, but a quantitative evaluation (e.g., life cycle assessment) should be carried out.

Considering the evaluated properties, the main advantages of the proposed composite are derived from acoustic absorption (absorbing the frequencies of human noise sources) and hygroscopic moisture buffering capacity. It is expected that giant reed-based boards can be suitable in environments where the speech and the involuntary listening of the users can cause acoustic discomfort (e.g., restaurants, offices, lecture halls, or museums (Cingolani et al., 2021; D’Orazio et al., 2020; De Salvio et al., 2021)). The high hygroscopicity and the high moisture buffering capacity can passively contribute to indoor conditions control. Hence, the results suggested a suitable composite’s employment as internal coating boards to improve hygro-thermal and acoustic comfort. However, the resistance to fire should be assessed to provide further support to this consideration.

Further analysis of the acoustic properties could be performed, both to better understand the giant reed-based composite behaviour and to propose an optimization. Since the acoustic absorption capacity is strongly related to the thickness, tortuous morphology, and pore structure (Asdrubali et al., 2016; Liuzzi et al., 2020b), the internal structure could be analysed. Regarding the possible optimization, as previously mentioned, the amplitude of the curve generated by the absorption peak should be increased, guaranteeing an absorption coefficient  $\alpha \approx 1.00$  for a broader range of frequencies. Finally, an interesting possibility to explore could be the combination of the considered giant reed-based boards with Mass-Loaded Vinyl (MLV) panels or barriers. They consist of effective sound-blocking products due to their resonant capacity, employed in walls, ceilings, and floors (Commercial-Acoustics, 2023; Gupta, 2019).



Combining these two products (e.g., applying an MLV barrier in one face of the giant reed-based boards) might result in an optimized, more performant acoustic absorber. As past research demonstrated (O’Flaherty and Alam, 2021), MLV barriers could improve boards’ sound absorption performance. Furthermore, the MLV could moderate one of the possible drawbacks of bio-based composites, namely the easy degradation and the low durability (Cintura et al., 2021).

The high hygroscopicity could be both an advantage and a disadvantage, securing a passive control of indoor conditions and causing composite’s degradation. High moisture content could modify the materials' properties and make them more susceptible to biological attack (Esteves et al., 2021; Krejsová and Doleželová, 2019; Verdier et al., 2014). Nevertheless, considering, in general, the low resistance to biological attack of agro-industrial wastes, the results of the visual observation after the hygroscopicity test were promising, confirming one of the benefits of using the sodium silicate solution. In addition to the contribution of the giant reed and the sodium silicate solution, separately (Section 4.4.4.2), it could be interesting to evaluate the influence of moisture content on the other properties, perform the hygroscopicity test for more than four cycles, and determine the possible critical moisture levels (Johansson et al., 2020).

Even if the thermal analysis results demonstrated that the giant reed-based composite cannot be considered a thermal insulator, this property could probably be improved by changing some parameters (e.g., the grain size, the percentages of aggregates-adhesive, and the production process). However, the composite could be employed as coating boards with some thermal insulation capacity. The influence of RH on thermal conductivity could be further analysed, as they are strictly related (Cintura et al., 2022; Liuzzi et al., 2017). This study was not carried out in the present work, as a detailed thermal performance evaluation was not one of the main aims. Furthermore, the low-promising performance did not encourage further analysis of this property. Lower thermal conductivity values could be achieved at lower values of RH, but much lower levels would not represent the indoor conditions.

Finally, as the compressive test demonstrated, pressing the composite was a feasible production process. Maybe, even if this would determine higher energy demand, it could improve giant-reed-based boards’ properties.

#### 4.4.5 Conclusions

This study analysed the properties of a composite of bio-waste-aggregates, namely particles of giant reed (*Arundo donax* L.) and a sodium silicate solution. Apparent density, thermal

conductivity, sound absorption, and flexural and compressive strength were evaluated to provide a wide-range characterization of the unconventional composite. The outcomes helped define the giant reed-based composite's most promising properties and support the decision to carry out future studies on producing eco-efficient boards. A comparison with the literature was carried out to understand better, discuss, and validate the results.

The achieved conclusions are reported below.

- The production process, as well as the mix design (percentage aggregates-adhesive), could be optimized, for example, by using less amount of sodium silicate solution or pressing the mixture of materials. Furthermore, future studies could evaluate the influence of the aggregates' particle size on the composite's final properties.
- The tested composite had higher values of apparent density and, coherently, higher thermal conductivity values than the composites considered for comparison. The composite did not meet the standard requirements to be classified as a thermal insulator. Still, the thermal performance may be improved by modifying some production parameters, as good thermal insulation capacity was expected from *A. donax*. Furthermore, the correlation between thermal conductivity and RH could be analysed.
- The composite showed high hygroscopicity, probably mainly determined by the sodium silicate solution, but the giant reed contributed to the moisture storage capacity. The moisture buffering value is in line with literature values and the composite showed high moisture buffering capacity, which could be both a benefit and a drawback. Even after the hygroscopicity test, no mould growth was detected. Hence, the composite did not show increased susceptibility to biological attack.
- The sound absorption behaviour was not so typical, considering bio-waste-based boards already studied, and it was typical of granular materials. The highest sound absorption coefficient values were achieved for frequencies between 600 Hz and 700 Hz, which was in the frequency range of the human voice. Hence, possible employment of the giant reed-based composite could be as indoor coating boards with acoustic absorption capacity. Further investigation could better explain the composite's behaviour and provide optimization, by enlarging the frequency range with a high acoustic absorption coefficient.
- The mechanical performance was inadequate for using the giant reed-based composite as structural boards or masonry units. The composite did not reach a rupture load, both for the bending and compressive test, and the amount and the distribution of the sodium silicate solution influenced the flexural strength values. The compressive performance was higher than other bio-based composites considered for comparison. Furthermore, the performed analysis demonstrated the feasibility of

producing a giant reed composite by pressing the materials and suggested ideas for future studies.

The laboratory tests to analyse the properties of boards made up of giant reed and sodium silicate solution demonstrated their promising use as sustainable internal coating boards, that would be not toxic for human health, and able to control acoustic and hygrothermal indoor comfort.

Further studies could be carried out on the already analysed properties and new ones. Indeed, the present work wanted to provide a general characterization of the composite as a starting point for further studies. The most promising aspects, namely the hygroscopicity and the acoustic performance, should be further evaluated by a vertical analysis. Hence, these properties could be optimized. Furthermore, a life cycle analysis is needed to quantify the eco-efficiency of this bio-waste-based building solution, as well as the evaluation of composite's degradation (e.g., resistance to biological attack and reaction to fire).

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## 4.5 Concluding remarks

The results of the developed laboratory experiences provided important information on the properties of the hazelnut shell-based boards, the sodium silicate solution used as an adhesive, and the properties of *Arundo donax*, both as raw material and when used in composites. Useful considerations for the development of the Ph.D. project were made. The concluding remarks of this part of the project could be summarized as follows.

The production method of the composites and the percentages aggregates-adhesive were defined and optimized, especially for the hazelnut shells-based composite samples. For these, the sodium silicate solution appeared uniformly distributed. Some steps of the process should be improved for potential future industrial production. However, this part ensured the acquisition of manual skills for producing and drawing up a detailed process. In the Ph.D. project implementation, the importance of carrying out practical tests, and transforming theoretical knowledge into practice, took increasing importance.

The laboratory experience allowed a better understanding of the studied composites: benefits and problems of the selected materials, the time required for the production, the feasibility of modifying the samples (e.g., cutting, filing), and their response to the different conditions, namely the variation of temperature and relative humidity. In addition to knowing the composites' properties, the experimental campaign ensured learning the use of the equipment and a better understanding of the proposed methods. For example, the analysis of bending behaviour allowed making important considerations on the most suitable laboratory test for the considered composites, useful for future experimental campaigns. Moreover, thanks to the collaboration of different departments of the University, hence, the collaboration of different experts, the acquired wealth of knowledge was increased.

The results of the performed analyses on hazelnut shell-based boards demonstrated that the composite cannot be employed as an indoor thermal insulator board as the thermal conductivity is not low enough. On the other hand, the hygroscopicity and the acoustic properties seemed promising, and they should be further analysed. The hazelnut shell-based composite could passively control indoor hygrometric and acoustic conditions. For these properties, the results suggested the rougher surface the better, but this consideration should be verified by further analysis. The mechanical tests demonstrated that the hazelnut shell-based board cannot be used as a structural element, in line with the expectations.

As for the *A. donax*-based boards, the results showed that, probably, by adding this bio-resource to the hazelnut shell-based composite, in addition to the mechanical properties, the

thermal insulation ones may be improved. Indeed, both the thermal conductivity and the apparent density of the *A. donax*-based composite were lower than the ones of the hazelnut shells-based one. However, also for the *A. donax*-based composite, the thermal conductivity is not low enough to be considered a thermal insulator, while the hygroscopicity and the acoustic properties were promising. Again, for acoustic and hygroscopicity performance, the results suggested the rougher surface the better. The comparison of the mechanical properties showed the deep differences between the composites, hence the influence of the selection of the raw materials in the final performance. Indeed, differently from the other properties, the composites showed a completely different behaviour both for flexural and compressive strength. Probably, this was determined also by the sodium silicate solution, namely the combinations of the aggregates and adhesive. Furthermore, different from indoor comfort, for mechanical performance, in particular for flexural strength, a flatter surface determined an improvement of the performance for both composites.

Finally, the *A. donax*-based composite samples obtained from the compressive strength test should be investigated after the test. The samples flattened out without breaking, and this suggested another suitable production process for *A. donax*-based boards (i.e., pressing at low temperatures). Overall, the results opened new possible developments of the project and provided new ideas for future studies about these bio-waste-based building products.



## OPTIMIZATION OF COMPOSITES: THE MIXTURE OF TWO BIO-AGGREGATES

### 5.1 Preamble

After the previous experimental campaigns, that analysed composite board samples made of hazelnut shells and sodium silicate solution, and *A. donax* and sodium silicate solution (separately), the mixture of the two aggregates was considered. Since composites based on hazelnut shells showed poor thermal insulation and mechanical performance, the addition of *A. donax* aimed to improve these properties. However, the thermal properties, following the results and considerations reported in Chapter 4, were no longer considered.

Some of the already employed composite samples, described in Sections 4.3, and 4.4 were tested again, as well as the hazelnut shells, provided by Raccolti di Cin, Baldissero d'Alba, Cuneo, Italy. As for the *A. donax*, two different cultivations were considered. The first one, employed to produce the samples described in Section 4.4, was collected from cultivation in Cadrano, Bologna, Italy. To produce new samples and carry out some of the tests described in this Chapter, *A. donax* was re-harvested from cultivation in Ozzano dell'Emilia, Bologna, Italy. Both the *A. donax* and the hazelnut shells were shredded and dried at  $T = 60\text{ }^{\circ}\text{C}$  and new samples were produced by considering the bio-wastes both separately and mixed.

Again, the composition was 70% aggregates and 30% adhesive (by volume) for all. Hence, the compositions were:

- For *A. donax*-based composite: 70% of *A. donax* and 30% of sodium silicate solution
- For hazelnut shell-based composite: 70% of hazelnut shells and 30% of sodium silicate solution

- For the mixture-based composite: 35% of *A. donax*, 35% of hazelnut shells, and 30% of sodium silicate solution.

The composite samples' designation is reported in the following sections.

Except for the thermal conductivity, not promising for both the bio-composites, the same analyses described in Chapter 4 were performed. Hence, the produced composite samples were smaller than 40 cm x 40 cm due to the standards' requirements and the employed types of equipment. This information could be useful for future research, as the assessment of larger samples could provide additional information on composites' behaviour.

Moreover, some new properties were assessed: the internal structure and the porosity, by carrying out an X-ray computed tomography (Section 5.2); the thermal stability, the water content, and the inorganic fraction composition of the bio-aggregates, by thermogravimetric analysis, TGA, and the composites' compositions by treating them in a muffle furnace (Section 5.3); the performance in case of fire (Section 5.3); the resistance to biological attack (Section 5.5). The X-ray computed tomography was performed to better understand both the acoustic and hygroscopic properties, making consideration on the different types of porosity. As for the other tests, they ensured a more complete characterization of the proposed boards, also by assessing some properties not so commonly considered (i.e., the performance in case of fire and the biological susceptibility).

Furthermore, the influence of each material on the final performance was investigated. Hence, hygroscopicity and behaviour in case of fire were evaluated also by considering the raw materials separately (the aggregates and the air-dried sodium silicate solution). The aim was to define the influence of materials' formulation, providing a more complete analysis of the composites' properties and a more detailed discussion of the results. It would be interesting to perform the same analysis for acoustic absorption.

As for the biological susceptibility, a detailed experimental campaign was planned and developed (Section 5.5). However, the results demonstrated that the considered conditions to test the biological attack cannot, therefore, occur as the composite samples could not resist the high relative humidity levels for a long period. Further analysis should be performed to validate the results and assess the composites' biological susceptibility.

The last performed analysis was the evaluation of the eco-efficiency of the composite samples by a life cycle assessment (LCA), described in Section 5.6. Some qualitative considerations were made and the information was set to perform further studies and address this topic quantitatively, by using a specific software (SimaPro).

## 5.2 Article H: Hygroscopicity and morphology of bio-based boards – Influence of the formulation

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### Abstract

The internal structures and the hygroscopicity of bio-based boards consisting of giant reed (*Arundo donax* L.) and hazelnut shells as bio-aggregates, and a sodium silicate solution as the adhesive, were investigated. The aim was to evaluate the influence of each material (the bio-aggregates and adhesive) and their distributions in the boards on the final performance. By carrying out X-ray computed tomography, the internal structures and the porosities of the boards were determined, allowing important considerations of their hygroscopicity. The voids' percentages were between 26% and 36% of the total volume of the composites. Both the materials and the composites demonstrated high hygroscopicity. In particular, the mixtures of the bio-aggregates and the sodium silicate allowed reaching a moisture buffering value of 7.44 g/(m<sup>2</sup>%RH) for the *A. donax*-based composite, 3.86 g/(m<sup>2</sup>%RH) for the hazelnut-shell-based composite, and 4.65 g/(m<sup>2</sup>%RH) for the mixture-based composite. Besides the identification of the contributions of the materials, a detailed discussion of the assessed properties was carried out to use these bio-based boards in vernacular historic construction. The results show how the aggregate type and the adhesive content affected the final behavior, demonstrating the importance of a conscious material choice. Furthermore, helpful information for the future development of these types of bio-based boards and their possible optimization was provided.

**Keywords:** agro-industrial waste; *Arundo donax*; bio-aggregate; bio-resource; giant reed; hazelnut shell; hygroscopicity; moisture buffering; sodium silicate; tomography.

### 5.2.1 Introduction

Research is nowadays increasingly focused on studying and proposing innovative and sustainable solutions practices. The environmental impact of the construction sector (for new construction, refurbishment, use, and deconstruction) is widely recognized [1,2], as well as the worldwide interest in reducing the production of toxic and environmentally damaging substances [3]. Both the Agenda 2023 [4] and the European Green Deal [1], among several

proposals, have reported the need for a reduction in energy demand and the encouragement of a circular economic system. Thus, among the investigated innovative possibilities, the use of bio-wastes and bio-resources seems to be a particularly attractive solution. It reduces energy consumption [5], ensures a circular economy, and moderates the problem of waste disposal [6,7]. Furthermore, the use of these types of materials, which do not act as water vapor barriers, can guarantee cultural heritage preservation, namely, vernacular architecture.

Past research has already considered bio-based building composites and assessed their properties. Both bio-wastes (such as agro-industrial wastes) and bio-resources have been investigated (such as straw [8], corn pith [9], rice husk [10], banana fibers [11], hemp shives [12], and reed [13]). Depending on the biomaterials used, the bio-based composites demonstrated different promising properties, namely, good thermal insulation performance, sound absorption capacity, high hygroscopicity, or adequate mechanical resistance. Several types of bio-based composites and products have been produced and studied, such as plastering mortars [14], glues [15], bricks [16], and boards [17,18], which are the ones considered in this study.

Bio-based boards are usually made of bio-aggregates (bio-wastes or bio-resources) and an adhesive (glue or binder). In addition to the bio-wastes/bio-resources, the employed adhesive also significantly influences the board's final properties, as well as its sustainability and toxicity [19,20]. Considering the same bio-aggregates and different adhesives, the composites' behaviors deeply change [21]. Using adhesives that could be harmful to human health, such as formaldehyde-based ones [22,23], reduces the non-toxicity and the eco-efficiency derived from the use of bio-based materials. The selected adhesive could also moderate some of the main drawbacks, such as low resistance to water, fire, and biological attack [24–26], improving the final composite's performance. Thus, knowing the properties of both the aggregates and the adhesive, individually and in composites, is crucial in building composites' production [27,28].

The present work addresses the topic of materials' influence on the final performance of composite boards. In particular, this research study considered bio-based boards made of bio-aggregates, namely, hazelnut shells and giant reed (*Arundo donax* L.), bounded by a sodium silicate solution [29,30] (these two reference studies provided characterizations of the proposed composites, evaluating their physical and mechanical properties).

Hazelnut shells are a widely produced bio-waste [29], available throughout the year due to hazelnut treatment before their use [31]. Some past studies have already considered the possibility of using hazelnut shells for building practices [32,33]. Nevertheless, they have not been largely investigated as such [34]. *A. donax* L., known as giant reed or giant cane, is an

herbaceous, perennial, non-food, invasive, and largely available bio- resource [35]. It can be found worldwide due to its tolerance to different climates and soil conditions [5]. It has already been employed for construction practices—i.e., in panels, concrete, plasters, and new construction technologies—showing promising mechanical and thermal properties [13]. Being natural products, both bio-aggregates could be employed in innovative building products compatible with vernacular architectural elements [36,37]. Indeed, they would be compatible with the traditional walls and roofs and could ensure their preservation.

Sodium silicate, also known as “water glass”, is a widely produced material, which has demonstrated several benefits. It is water-soluble, fire-resistant, non-toxic, and, hence, not dangerous to human health, environment-friendly, and economically convenient [38–40]. It has been employed in several sectors, such as to produce detergents and cleaning compounds [38], for material treatments [40], in ceramics [41], and in the production of geopolymers [42] and concretes [43]. Among its several uses, the employment of sodium silicate as a binder to produce bio-based building composites could be a promising possibility [44]. Indeed, in addition to guaranteeing the feasibility of their production, it can reduce some of the main weaknesses derived from their organic matrices, improving their resistance to fire [45] and preventing chemical decomposition [46,47]. Some past studies have investigated the employment of sodium silicate as an adhesive for bio-based building boards. For example, Bakatovich et al. [39] evaluated the properties of reed and straw fiber-based composites bonded with sodium silicate and rosin, and Liuzzi et al. [46] produced composites made of olive fibers and straw and a sodium silicate solution as the adhesive. Both studies reported promising thermal insulation performance of the composites. Cintura et al. [21] considered some bio-based boards bounded with different adhesives and evaluated their biological susceptibility without inoculation. The composites with sodium silicate demonstrated the highest resistance, avoiding mold growth at high relative humidity levels. Besides its several benefits, it has also shown some drawbacks, such as high hygroscopicity and low moisture resistance [45,48], which could cause the degradation of the materials [49]. Furthermore, it could be less eco-efficient than other adhesives (i.e., bio-based ones or natural glues). Indeed, higher temperatures could be required for its production [38], even if lower than those required for other widely used adhesives (such as cement-based binders).

As hazelnut-shell-based and *A. donax*-based boards have demonstrated interesting hygroscopic behavior [29,30], which can be useful to passively contribute to indoor comfort [50], reducing energy consumption [51], the present study mainly considered this property. The contribution of each material was investigated by testing them individually and in composites.

Furthermore, the materials' dispositions in the proposed boards were analyzed, assessing the correlation between the hygroscopicity and internal structure. X-ray computed tomography, which is a non-invasive 3D-imaging technique able to supply crucial information on the studied products [52], ensured the evaluation of the internal configurations of the boards. This technique has been employed in several fields, such as medical, industrial, and cultural heritage [53], as well as to investigate bio-based composites [54]. It provides information such as the pore size distribution, porosity, and tortuosity of the porous network [55]. In this work, X-ray computed tomography ensured the identification of the distributions of the aggregates and adhesive and the porosities of the bio-based boards. Finally, statistical analysis (ANOVA) was carried out for further consideration of the results.

The present work aimed to analyze the composites' properties and define the influence of each material on the final performance, both in terms of the materials' properties and the aggregate/adhesive's internal distribution.

## 5.2.2 Materials and specimens

### 5.2.2.1 Bio-aggregates and adhesive

Giant reed (*Arundo donax* L.) and hazelnut shells were selected as bio-aggregates, characterized, and prepared as described in past studies [29–31]. *A. donax* was harvested from the cultivation of the Department of Agricultural and Food Sciences, the University of Bologna, in Ozzano dell'Emilia, Bologna, Italy. Hazelnut shells were provided by Raccolti di Cin, Baldissero d'Alba, Cuneo, Italy. Both were shredded to have grain sizes mainly between 4 mm and 8 mm and dried at  $T = 60\text{ }^{\circ}\text{C}$  until reaching a constant mass (with a variation in mass of  $\leq 0.1\%$  after 24 h) [29,30].

Figure 5.2-1, adapted from the reference studies in [29–31], shows the particle size distributions of both bio-aggregates.

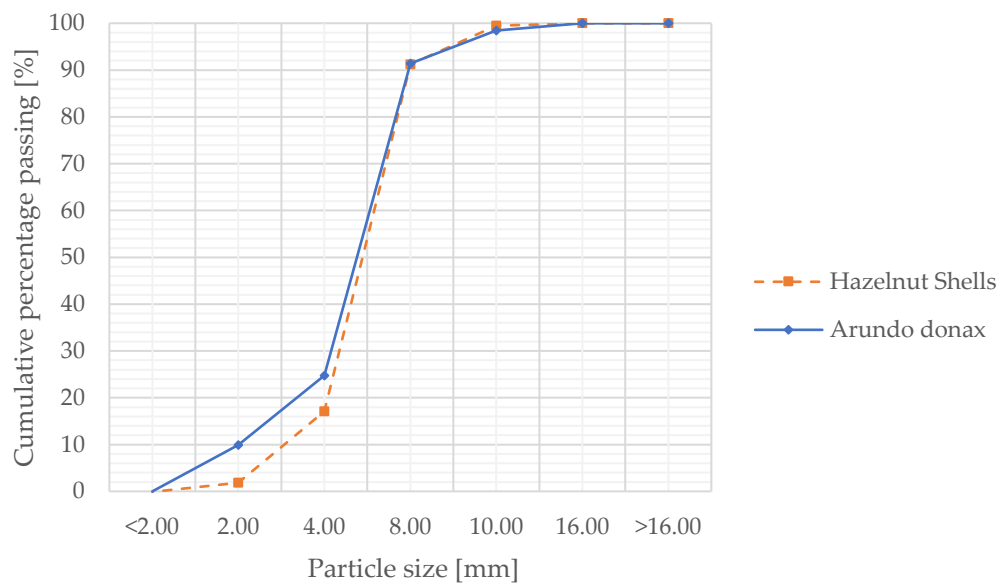


Figure 5.2-1. Particle size distribution of *Arundo donax* and hazelnut shells.

A sodium silicate solution,  $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ , provided by Ingessil Srl, Montorio, Verona, Italy, with a density of 1.471 g/mL, a sodium silicate concentration of 41.33% *p/p*, and pH (T = 20 °C) of 12.40, was employed as the adhesive, coherent with the reference studies [29,30].

#### 5.2.2.2 Composites samples preparation

The samples were produced as described by Cintura et al. [29,30]. The same percentages of the bio-aggregates/adhesive and the same production process were considered. They were selected after several tests, maximizing the contents of the aggregates, minimizing the amount of the adhesive, and securing sufficient mechanical resistance. The selected percentages were 70% aggregates and 30% adhesive (by total volume).

Table 5.2-1 shows the mix designs and the composites' designations. As for the latter, the first letters of the materials were considered (**A** = *A. donax*, **H** = hazelnut shells, and **AH** = *A. donax* + hazelnut shells).

Table 5.2-1. Mix designs (quantities of aggregates and adhesives in percentages by the total volume) and the composites' designations considered for the experimental analysis..

| Composites' designation | Materials quantities (% by total volume) |                 |                          |
|-------------------------|------------------------------------------|-----------------|--------------------------|
|                         | <i>A. donax</i>                          | Hazelnut shells | Sodium silicate solution |
| A                       | 70                                       | -               | 30                       |
| H                       | -                                        | 70              | 30                       |
| AH                      | 35                                       | 35              | 30                       |

The bio-aggregates and the sodium silicate solution were mechanically mixed for 10 min, placed into silicon molds without compaction, and leveled using a spatula to obtain a horizontal surface. The molds were closed and maintained at  $T = 60\text{ }^{\circ}\text{C}$  for 3 h and then under laboratory conditions (air-dried), always being rotated to secure a homogeneous distribution of the sodium silicate solution [31]. After 2 days, the samples were de-molded and cured under laboratory conditions for 28 days. To secure complete drying, they were finally dried at  $T = 50\text{ }^{\circ}\text{C}$  until reaching a constant mass (a variation in mass of less than 0.5% after 24 h) [46,56]. The composite samples were analyzed after being dried under this condition.

Cylindrical (diameter = 10 cm; thickness = 4 cm) and rectangular (10 cm  $\times$  10 cm; thickness = 4 cm) samples were produced to be employed for different analyses (as described in the following section). Figure 5.2-2 shows the produced samples. Three replications for each composite were considered.



Figure 5.2-2 Composite samples made of bio-aggregates and sodium silicate solution. Rectangular and cylindrical samples: (a,d) *A. donax*-based composite—A; (b,e) hazelnut-shell-based composite—H; and (c,f) *A. donax* + hazelnut-shell-based composite—AH.

As better detailed in the following section, the materials (bio-aggregates and adhesive, individually) were tested too. In this case, the bio-aggregates were employed after the preparation described in Section 5.2.2.1 (i.e., after shredding and drying at  $T = 60\text{ }^{\circ}\text{C}$  until reaching a



constant mass). For the designations, the repetition of the letters was considered: *A. donax* = **AA**; hazelnut shells = **HH**; and *A. donax* + hazelnut shells = **AAHH**. Again, three samples for each bio-aggregate were tested. As for the adhesive, the sodium silicate solution was placed in a rectangular silicon mold (10 cm × 10 cm) and left to air-dry under laboratory conditions for 28 days. The employed designation was **S** (= sodium silicate solution), and four replications were considered.

### 5.2.3 Test methods

Table 5.2-2 summarizes the tests performed, the numbers, sizes, and designations of the samples used, and the considered references.

Table 5.2-2. Tests performed, number, size, and designation of the samples, and references.

| Property                  | Material        | N. | Size [cm]      | Samples                 | Ref.                         |
|---------------------------|-----------------|----|----------------|-------------------------|------------------------------|
|                           |                 |    |                | Designation             |                              |
| Apparent density          | <i>A. donax</i> | 3  | 10 x 10, h = 4 | A1, A2, and A3          | Adaptation of EN 323 [57]    |
|                           | Hazelnut shells | 3  | 10 x 10, h = 4 | H1, H2, and H3          |                              |
|                           | Mixture         | 3  | 10 x 10, h = 4 | AH1, AH2, and AH3       |                              |
| Tomography                | <i>A. donax</i> | 3  | d = 10, h = 4  | A4, A5, and A6          | -                            |
|                           | Hazelnut shells | 3  | d = 10, h = 4  | H4, H5, and H6          |                              |
|                           | Mixture         | 3  | d = 10, h = 4  | AH4, AH5, and AH6       |                              |
| Hygroscopicity Materials  | <i>A. donax</i> | 3  | 15 x 11, h = 2 | AA1, AA2, and AA3       | Adaptation of ISO 24353 [58] |
|                           | Hazelnut shells | 3  | 15 x 11, h = 2 | HH1, HH2, and HH3       |                              |
|                           | Mixture*        | 3  | 15 x 11, h = 2 | AAHH1, AAHH2, and AAHH3 |                              |
|                           | Sodium silicate | 4  | 10 x 10 x h*   | S1, S2, S3, and S4      |                              |
| Hygroscopicity Composites | <i>A. donax</i> | 3  | 10 x 10 x 4    | A1, A2, and A3          | ISO 24353 [58]               |
|                           | Hazelnut shells | 3  | 10 x 10 x 4    | H1, H2, and H3          |                              |
|                           | Mixture         | 3  | 10 x 10 x 4    | AH1, AH2, and AH3       |                              |

\* Note: Mixture = 50% *A. donax*–50% hazelnut shells of the total volume of the filled container (without adhesive); sodium silicate = samples of the air-dried (lab. conditions) sodium silicate solution with a thickness (h) varying between 2 and 4 mm.

#### 5.2.3.1 Apparent density

An adaptation of EN 323 [57] was considered to determine the apparent density of the composites. Three prismatic samples of each composition (A1, A2, and A3; H1, H2, and H3; and AH1, AH2, and AH3) of 10 cm × 10 cm × 4 cm (Figure 5.2-2a–c) were stabilized at T = 50 °C (with a variation in mass of less than 0.5% after 24 h), weighed with a Kern EW 2200-2NM electronic balance, and measured with a Fervi digital caliper. The ratios between the mass and volume were calculated, and the average values and the standard deviations were determined.

### 5.2.3.2 Internal structure and porosity

Three cylindrical samples of each composition (A4, A5, A6, H4, H5, H6, AH4, AH5, and AH6), with diameter = 10 cm and  $h = 4$  cm (Figure 5.2-2d–f), were used to analyze their internal structures and porosities using X-ray computed tomography. For this purpose, a CT system was used. The apparatus (Figure 5.2-3a) was assembled in-house (by Physics and Astronomy Department of the University of Bologna) as a multipurpose CT system and consisted of a Bosello XRG120.IT conventional X-ray tube, a Varian PaxScan 2520D flat-panel X-ray detector, and a Physik Instrument manipulation axis set with an M-038 micrometric sample rotation stage and M413.3PD for detector translation. The samples were placed with one face over the rotation stage in a horizontal position.



Figure 5.2-3. X-ray computed tomography: (a) employed apparatus; (b) three samples separated by expanded polystyrene boards placed on the rotation platform.

Two or three samples, separated with expanded polystyrene boards (which are transparent to X-rays), were placed vertically on the rotation platform, one on top of the other (Figure 5.2-3b), to optimize the scanning time. A standard scanning protocol for the system was used: 900 projections over a 360-degree angle; a 4-frame average for each projection; and low magnification ( $M = 1.115$ ). The voltage (70 kV), current (6.5 mA), and detector frame rate (3 fps) were chosen according to the sample size and material after an initial evaluation of the X-ray absorption and signal outcome in the detector. No filtration was used. The overall scanning time was 1 h.

After scanning each group of samples, CT slices were obtained via raw data elaborations with the in-house software PARREC, EXE\_2023\_01\_26\_ PARREC\_BHC\_AV, (implementing a standard version of the filtered back-projections cone-beam Feldkamp algorithm [59]). One of

the great strengths of the performed X-ray computed tomography was the large number of reconstructed slices, which ensured the reproduction of the entire internal structures of the samples at a high resolution.

Slices produced with the reconstruction software were imported into the ImageJ/Fiji 1.53c freeware image processing software for a more convenient data visualization and further elaboration via a segmentation and pore classification process (Section 5.2.4.2). The three-dimensional rendering of the reconstructed X-ray-scanned objects and the first simple overall porosity measurements were performed by importing CT slices with the VGStudioMax 2.1 commercial software (Volume Graphics).

It is important to underline that although the samples employed for the tomography were not the same as those employed for the hygroscopic analysis (described in the following Section 5.2.3.3), the results were considered significant to carry out a comparison between the internal structures and hygroscopicity as all the samples were produced at the same time with the same mixtures of the aggregate–adhesive, production process, and conditions

### 5.2.3.3 Hygroscopic properties

The hygroscopicity (i.e., the ability to adsorb/release moisture under relative humidity (RH) variations [60]) can be determined via several tests, divided into those performed using the step response method and those performed in a flux chamber [61]. In the present study, the step response method was considered, according to ISO 24353 [58]. The variation in mass over time was evaluated for both the materials (the bio-aggregates and adhesive, considered separately: AA, HH, AAHH, and S) and the composites (A, H, and AH).

The bio-aggregates were placed in plastic boxes with open-top surfaces and filled up to a height of 2 cm (Figure 5.2-4a), as described by Cintura et al. [28]. This ensured the exposure of only one sample face (15 cm × 11 cm). Three replicates of each bio-aggregate were tested (AA1, AA2, and AA3; HH1, HH2, and HH3) as well as the mixture of 50% *A. donax* and 50% hazelnut shells (AAHH1, AAHH2, and AAHH3).

As for the dried sodium silicate solution and the composites, a moisture barrier was provided using aluminum foil (Figure 5.2-4b,c), according to ISO 24353 [58] and the method described by Cintura et al. [29,30]. Thus, only the top surface (area = 0.1 m<sup>2</sup>) was exposed. Four replicates of the dried sodium silicate solution of 10 cm × 10 cm and a thickness between 2 and 4 mm (S1, S2, S3, and S4) were tested (Figure 5.2-4b). As for the composites, three samples of 10 cm × 10 cm and a thickness of 4 cm for each composite—the same employed to evaluate the apparent density—were considered (Figure 5.2-4c).



Figure 5.2-4. Samples for the hygroscopicity test: (a) plastic boxes (15 cm × 11 cm) filled up to 2 cm with the different aggregates (samples AA, HH, and AAHH); (b) air-dried sodium silicate solution covered with aluminum foil, except for the exposure surface (area = 0.1 m<sup>2</sup>); (c) composite samples (aggregates + sodium silicate solution) A, H, and AH (10 cm × 10 cm × 4 cm) covered with aluminum foil, except for the exposure surface (area = 0.1 m<sup>2</sup>); and (d) samples in the climatic chamber.

The samples were exposed to four cycles of 24 h (a 12 h sorption phase at 75% RH and a 12 h desorption phase at 50% RH) using a climatic chamber (Climatest ARGOLab CH 250) after being pre-conditioned at T = 23 °C and RH = 63% (Figure 5.2-4d) until reaching a constant mass (with a change in mass not higher than 0.5% over 24 h). The samples were weighed every 3 h to determine the moisture adsorption ( $\rho_{A,ac}$ ) and desorption content ( $\rho_{A,dc}$ ), moisture content difference ( $\rho_{A,sc}$ ), and, thus, the sorption/desorption capacity, as indicated in ISO 24353 [58] and described in Equations (5.2-1)–(5.2-3).

$$\rho_{A,ac} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \frac{m_{an} - m_{d(n-1)}}{\text{Area}} \quad (5.2-1)$$

$$\rho_{A,dc} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \frac{m_{an} - m_{dn}}{\text{Area}} \quad (5.2-2)$$

$$\rho_{A,sc} \left[ \frac{\text{kg}}{\text{m}^2} \right] = \rho_{A,ac} - \rho_{A,dc} \quad (5.2-3)$$

where  $m_{an}$  (kg) is the mass at the end of the sorption phase of the considered cycle,  $m_{d(n-1)}$  (kg) is the mass at the end of the previous desorption cycle,  $m_{dn}$  (kg) is the mass at the end of the desorption phase of the considered cycle, and Area (m<sup>2</sup>) is the exposed surface.

The moisture buffering value (MBV) quantifies the content of adsorbed and released moisture considering cycling between two levels of RH [62,63]. It was calculated considering the average value between the MBV of the sorption, MBV<sub>a</sub> (Equation (5.2-4)), and the desorption phase, MBV<sub>d</sub> (Equation (5.2-5)), of the last three cycles [29,30].

$$\text{MBV}_a \left[ \frac{\text{g}}{\text{m}^2 \text{ \% RH}} \right] = \frac{m_{an} - m_{d(n-1)}}{\text{Area} \times (\text{RH}_{\text{high}} - \text{RH}_{\text{low}})} \quad (5.2-4)$$

$$\text{MBV}_d \left[ \frac{\text{g}}{\text{m}^2 \text{ \% RH}} \right] = \frac{m_{an} - m_{dn}}{\text{Area} \times (\text{RH}_{\text{high}} - \text{RH}_{\text{low}})} \quad (5.2-5)$$

where the  $RH_{high}$  and  $RH_{low}$  are 75% and 50%, respectively [58]. The samples were classified by adapting the method described by Rode et al. [64], and the results were compared with values in past studies and literature. It is important to underline that the classification was provided considering the study by Rode et al. [64], although the evaluation was carried out considering the ISO 24353 [58], as the considered method could determine differences in the results [61,65].

#### 5.2.3.4 Statistical analysis

An analysis of variance (ANOVA) was performed using MATLAB R2023a to define if the materials (considered as the variable) identified statistically distinguishable groups, both individually and in composites. Although three samples for each group were tested, this screening test provided an idea of the influences of the employed bio-aggregates in the final performance.

Analysis of variance (ANOVA) tests if the null hypothesis ( $H_0$ )—which considers the analyzed groups as equal—is verified. Hence, if the  $H_0$  is true, the groups are not distinguishable; otherwise, the alternative hypothesis is confirmed, and at least one of the means is different. Once the groups are defined, the experimental data are analyzed with descriptive statistical values, namely, the p-value and F-value [66,67]. If the F-value is higher than 1 and the p-value is higher than 0.05, the null hypothesis ( $H_0$ ) is confirmed (there are no distinguishable groups). If the F-value is less than 1 and the p-value is less than 0.05, the  $H_0$  is, hence, false, and the alternative hypothesis is confirmed.

The considered groups were the three composites (the *A. donax*-based one (A), hazelnut shell-based one (H), and mixture-based one (AH)) regarding their apparent densities and porosities. For the hygroscopicity, the three bio-aggregates (*A. donax* (AA), hazelnut shells (HH), and the mixture of *A. donax* and hazelnut shells (AAHH)) were considered, too. The selected significance level was 5%, and the p-values were analyzed to determine if the null hypothesis ( $H_0$  = no differences between the groups) was verified. Post hoc tests were performed using a multiple-comparison test.

### 5.2.4 Results and discussion

#### 5.2.4.1 Apparent density

Table 5.2-3 reports the results of the apparent densities of the three samples for each composite, the average values, and standard deviations (SD).

Table 5.2-3. Results of apparent densities at T = 50 °C and porosities: individual and average values and standard deviation (SD).

| Composite samples                 | Apparent density [kg/m <sup>3</sup> ] | Voids [%] * | Solids [%] * |
|-----------------------------------|---------------------------------------|-------------|--------------|
| <i>A. donax</i>                   |                                       |             |              |
| A1                                | 416                                   | -           | -            |
| A2                                | 438                                   | -           | -            |
| A3                                | 555                                   | -           | -            |
| A4                                | -                                     | 26          | 74           |
| A5                                | -                                     | 35          | 65           |
| A6                                | -                                     | 33          | 67           |
| Average Value ± S.D.              | 470 ± 75                              | 31 ± 5      | 69 ± 5       |
| Hazelnut shells                   |                                       |             |              |
| H1                                | 628                                   | -           | -            |
| H2                                | 723                                   | -           | -            |
| H3                                | 680                                   | -           | -            |
| H4                                | -                                     | 35          | 65           |
| H5                                | -                                     | 38          | 62           |
| H6                                | -                                     | 34          | 66           |
| Average Value ± S.D.              | 677 ± 47                              | 36 ± 2      | 64 ± 2       |
| <i>A. donax</i> + Hazelnut shells |                                       |             |              |
| AH1                               | 667                                   | -           | -            |
| AH2                               | 576                                   | -           | -            |
| AH3                               | 579                                   | -           | -            |
| AH4                               | -                                     | 21          | 79           |
| AH5                               | -                                     | 27          | 73           |
| AH6                               | -                                     | 29          | 71           |
| Average Value ± S.D.              | 607 ± 52                              | 26 ± 4      | 74 ± 4       |

\*Note: Percentages by total volume.

The values of the apparent density were in line with the ones in past studies that considered *A. donax*-based and hazelnut-shell-based composites, namely, 517 kg/m<sup>3</sup> and 702 kg/m<sup>3</sup>, respectively [29,30].

The distribution of the aggregates and the sodium silicate was not controlled during the production process. Hence, the samples could have had some differences in aggregate sizes, internal porosity, and void volume. The variations between the samples of the same composite may have determined some differences in the results of other properties (e.g., the hygroscopicity, as discussed in the following section). However, as the standard deviation related to the average value was low, good uniformity between the tested samples was guaranteed. Moreover, it seemed that better homogeneity was ensured for the hazelnut shell-based composite.

The *A. donax*-based composite showed the lowest values of apparent density, followed by the mixture of the two aggregates. The hazelnut-shell-based composite had the highest values. Thus, *A. donax* lowered the apparent density in the mixture-based composite. These results are coherent with the expectation, and there was some correspondence with the values of the loose bulk density of the two aggregates. For the hazelnut shells, the loose bulk density was higher than that for *A. donax*—469 kg/m<sup>3</sup> and 181.3 kg/m<sup>3</sup>, respectively [29,30].

#### 5.2.4.2 Internal structure and porosity

Figure 5.2-5 reports one sample for each composite and its internal structures, showing both transverse and longitudinal sections, acquired from ImageJ/Fiji software. For a more precise comparison, the same sections were considered for each sample. Among all the frames, the middle one (i.e., 1/2 of the total number of frames) and the quarter ones (1/4 and 3/4 of the total number of frames) were reported. This allowed showing the successive sections of the internal structure, simplifying it.

These results provide a visual evaluation of the homogeneity of the samples and the internal configurations of the aggregate–adhesive combinations. Indeed, Figure 5.2-5 shows the sizes and shapes of the voids, the possible connections between the internal voids, the orientations of the aggregates, and the distributions of the sodium silicate solution. In both the transverse and longitudinal sections, the white parts represent the sodium silicate; the grey parts represent the aggregates; and the black parts (inside the samples) represent the voids.

The differences between the internal structures of the composites were more pronounced along their thickness ( $h = 4$  cm). The hazelnut-shell-based samples seemed more homogeneous than the *A. donax*-based ones. For these, the sodium silicate (the white parts in Figure 5.2-5) was mainly on the bottom side (the one in contact with the mold during the production process). This is in line with the expectation, as the optimization of the production process was defined by considering hazelnut shells [31]. Furthermore, the differences between samples were coherent with the values of the standard deviation of the apparent density (Section 5.2.4.1).

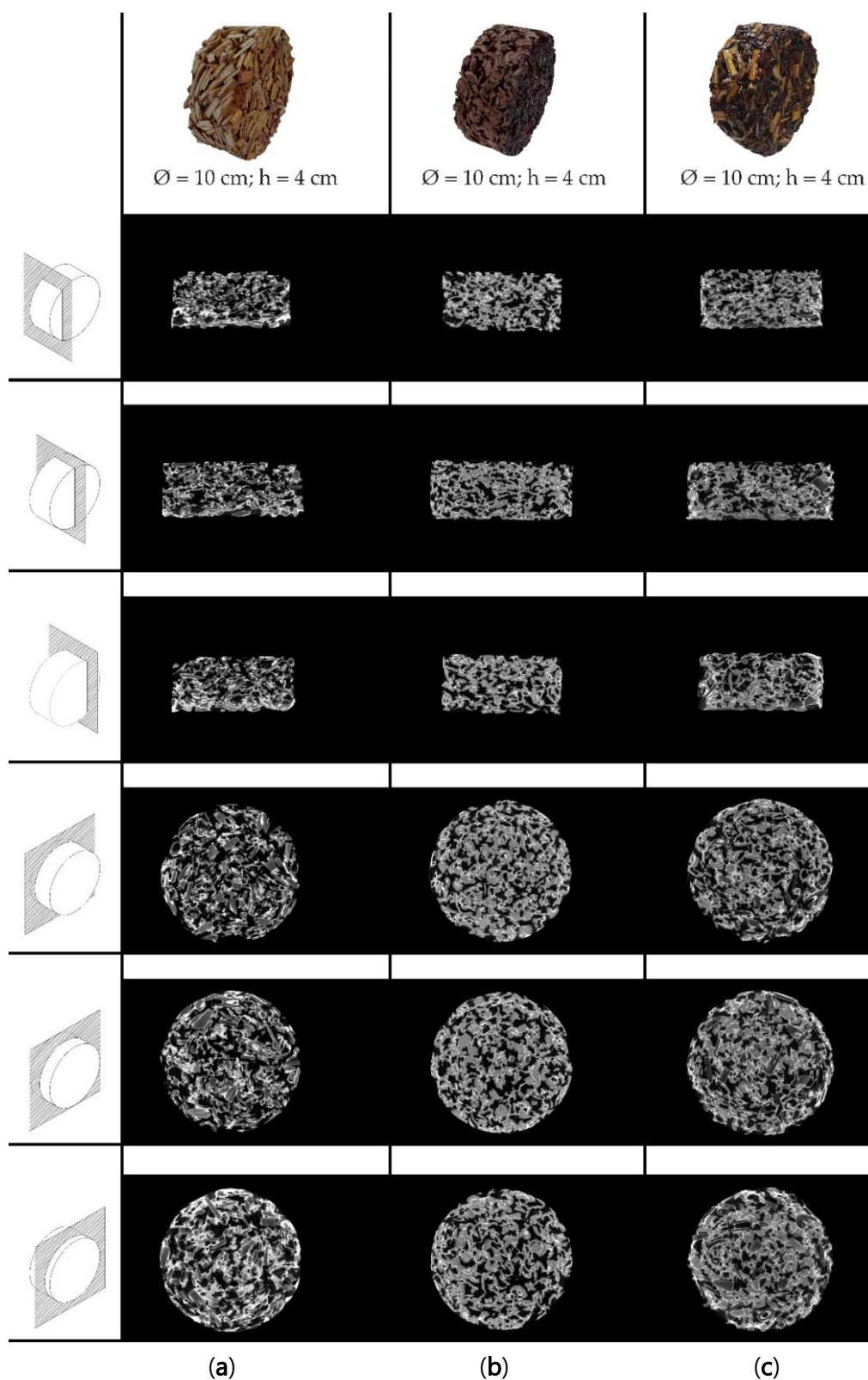


Figure 5.2-5. Cylindrical samples of each composite and their corresponding internal structures (transverse and longitudinal sections): (a) *A. donax*-based composite; (b) hazelnut-shell-based composite; and (c) mixture-based composite.



Evident differences were found between the shapes of voids and aggregates, which determined different internal connections. The contribution of the sodium silicate solution's distribution significantly affected the type of porosity. As represented in Mati-Baouche et al.'s study [68], the combination of the adhesive–aggregate produced solid, closed pores, open pores, dead-end pores, dead-end clusters, and, hence, kinematic porosity, and deadend porosity (properties mainly considered in the acoustic field).

Table 5.2-3 reports the results of the internal porosity of each composite sample, the average values, and the standard deviation (SD).

The composites showed similar percentages of solids/voids, with a variation of only 5%. Hence, the porosity seemed to be determined by the mixture of the bio-aggregate– adhesive rather than by the type of bio-aggregate. The latter seemed to influence mainly the shape and size of voids, their connections, and the tortuosity. The standard deviation related to the average value was low (with a maximum of 5%). Hence, the reproducibility was good: similar results were achieved for the three replications.

The evaluated porosity was considered “macroporosity” as the percentage of macropores, determined by the arrangement of the shredded bio-aggregates [62,68], was provided. The large sizes of pores were determined by the aggregate grain size, between 4 and 8 mm (Section 5.2.2.1). Further analysis could be performed to define the other types of porosity, as discussed in Section 5.2.4.5.

The quantification of the percentages of solids/voids was a starting point for the analysis of further characteristics of the internal structures of the samples. Certainly, in addition to the number of voids, their shapes, their connections, and the tortuosity influenced the final performance of the composites, as discussed in the following sections.

### 5.2.4.3 Hygroscopic properties

Figure 5.2-6 shows the moisture adsorption/desorption content curves of the materials for four adsorption/desorption cycles. Three samples for each aggregate (i.e., three samples of *A. donax* (AA), three of hazelnut shells (HH), and three of the mixture of *A. donax* and hazelnut shells (AAHH)), and four samples of sodium silicate (S) were considered and reported. To provide a clearer representation, the same color and style of lines were considered for the same tested material.

The differences between the moisture content values for each material become more marked along the cycles. Indeed, at the beginning of the test (12 h and 24 h), the curves achieved values that varied within  $\pm 0.03$  kg/m<sup>2</sup>; at the end (84 h and 96 h), the range increased

( $\pm 0.06 \text{ kg/m}^2$ ). As for the curves, all the materials showed similar behavior. *A. donax* (both alone and mixed with the hazelnut shells) stabilized earlier than the others, having similar values during the four testing days. Meanwhile, for both the hazelnut shells and the dried sodium silicate solution, the curve increased during the test, demonstrating a higher moisture storage capacity, more markedly for the sodium silicate. Indeed, the sodium silicate solution achieved the highest values of moisture adsorption and desorption content for all cycles, followed by the hazelnut shells, and then by *A. donax* and the mixture of the two aggregates. These results are in line with the expectation, as sodium silicate is known to be highly hygroscopic [44,48].

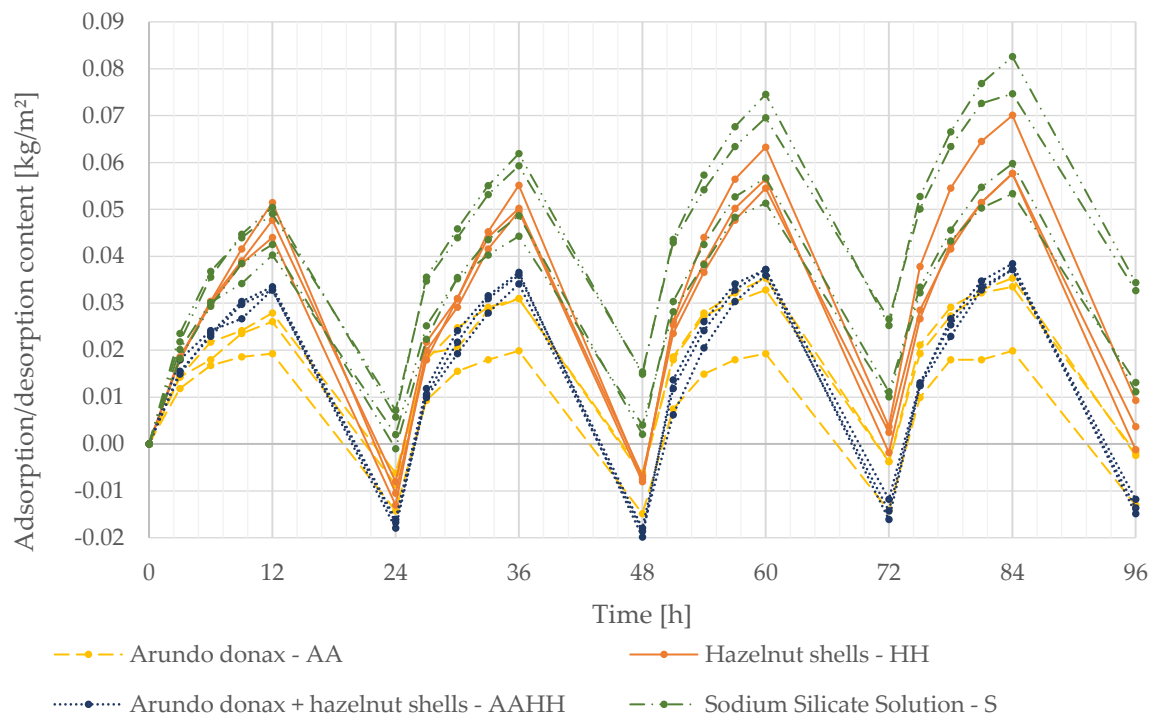


Figure 5.2-6. Moisture adsorption/desorption contents for four cycles of three samples of each bio-aggregate and four samples of the sodium silicate solution, considered separately.

Considering the values of moisture adsorption ( $\rho_{A,ac}$ ) for each cycle (average values), the hazelnut shells achieved the highest values, with a  $\rho_{A,ac}$  between  $0.048 \text{ kg/m}^2$  and  $0.066 \text{ kg/m}^2$ . *A. donax* achieved the lowest values, with a  $\rho_{A,ac}$  between  $0.024 \text{ kg/m}^2$  and  $0.038 \text{ kg/m}^2$ . The same was found for the moisture desorption content ( $\rho_{A,dc}$ ): the hazelnut shells showed the highest values, with a  $\rho_{A,dc}$  between  $0.057 \text{ kg/m}^2$  and  $0.059 \text{ kg/m}^2$ , and *A. donax* showed the lowest ones, with a  $\rho_{A,dc}$  between  $0.034 \text{ kg/m}^2$  and  $0.037 \text{ kg/m}^2$ . The moisture content difference,  $\rho_{A,sc}$ , for the bio-aggregates decreased after the first cycle, differently from the sodium silicate solution, for includeh the  $\rho_{A,sc}$  increased during the test:  $0.003 \text{ kg/m}^2$  after the first cycle,  $0.006$

kg/m<sup>2</sup> after the second, 0.009 kg/m<sup>2</sup> after the third, and, finally, 0.005 kg/m<sup>2</sup>. This shows that the sodium silicate stored the moisture, and its sorption capacity increased.

Cintura et al. [28] evaluated the hygroscopicity of some agro-industrial wastes, including hazelnut shells, using the same test method. Although the researchers achieved higher values for both the sorption and desorption moisture content, the trend of the curve was similar, and the hazelnut shells showed a moisture storage capacity. The differences between the values could be determined by the different types of hazelnut shells (their physical and chemical properties) as well as the differences in the employed equipment.

Hygroscopicity is strictly related to the chemical and physical properties of the material, including the porosity [54,62,69], as previously reported (Section 5.2.4.2). The results indicate a greater porosity for the hazelnut shells than for *A. donax*, which is different from the expectation. Indeed, the results of the loose bulk density (469 kg/m<sup>3</sup> for hazelnut shells [29] and 181.3 kg/m<sup>3</sup> [30] for *A. donax*) would lead to hypothesize a greater porosity for *A. donax*, as the two properties are correlated [70]. The same for the thermal insulation performance, as *A. donax* demonstrated a promising thermal insulation performance [35,39], and the two properties are strictly connected [60,71]. However, considering only the porosity is too simplifying, as many other parameters affect both the hygroscopicity and the other properties.

As for the dried sodium silicate solution, its high hygroscopicity is derived from its chemical composition. It ensures an easy reaction with water and makes the sodium silicate able to absorb moisture from the environment, as detailed in past research [38,48].

Figure 5.2-7 shows the moisture adsorption/desorption content curves during the four cycles for the composite samples (i.e., three samples of the *A. donax*-based composite (A), three of the hazelnut-shell-based one (H), and three of the mixture-based one (AH)). Again, the same color and style of lines were considered for the samples of the same composite.

The hazelnut-shell-based composite achieved the lowest values of moisture content in the four cycles. The composite made of *A. donax* (both alone and combined) reached the highest values. The hygroscopicity depended on the aggregates, the adhesive, and their coupling [72], as well as the conformation of the composite, namely, the adhesive's distribution, the porosity, and the grain size of the aggregate [73]. For this reason, one of the samples of the *A. donax*-based composite showed different values: the exposed surface could have had a different distribution of the sodium silicate solution or different aggregate grain sizes. However, considering the average curves, the composites showed similar behavior (similar trends in the moisture adsorption/desorption curves).

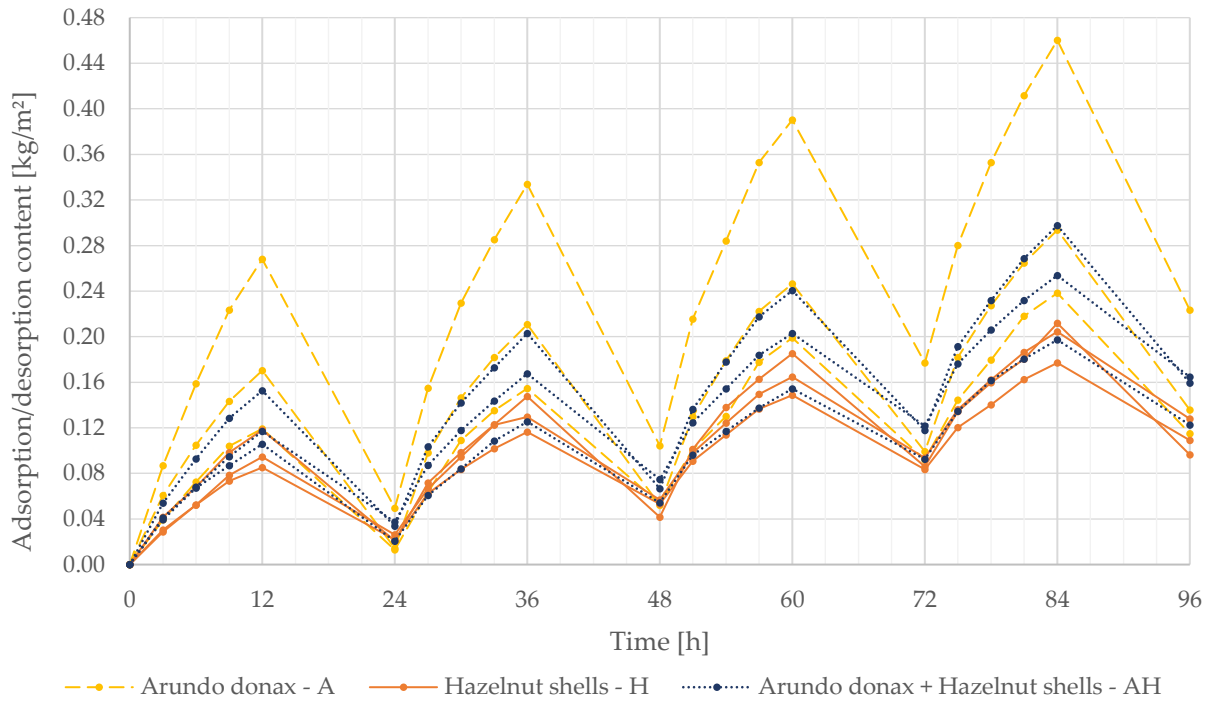


Figure 5.2-7. Moisture adsorption/desorption contents for four cycles of three samples for each composite.

None of them reached stability after the four cycles, as the adsorption and desorption moisture contents continued to increase. Indeed, considering the average values, the moisture absorption content,  $\rho_{A,ac}$ , increased from 0.099 kg/m<sup>2</sup> to 0.110 kg/m<sup>2</sup> for the hazelnut-shell-based composite (increasing by 11%); from 0.186 kg/m<sup>2</sup> to 0.208 kg/m<sup>2</sup> for the *A. donax*-based one (increasing by 12%); and from 0.125 kg/m<sup>2</sup> to 0.139 kg/m<sup>2</sup> for the mixture-based one (increasing by 11%). The moisture desorption content,  $\rho_{A,dc}$ , increased from 0.076 kg/m<sup>2</sup> to 0.087 kg/m<sup>2</sup> for the hazelnut-shell-based composite (increasing by 13%); from 0.160 kg/m<sup>2</sup> to 0.173 kg/m<sup>2</sup> for the *A. donax*-based one (increasing by 8%); and from 0.094 kg/m<sup>2</sup> to 0.101 kg/m<sup>2</sup> for the mixture-based one (increasing by 7%). The moisture content difference,  $\rho_{A,sc}$ , was between 0.023 kg/m<sup>2</sup> and 0.037 kg/m<sup>2</sup> for the hazelnut-shell-based composite; between 0.026 kg/m<sup>2</sup> and 0.053 kg/m<sup>2</sup> for the *A. donax*-based one; and between 0.031 kg/m<sup>2</sup> and 0.045 kg/m<sup>2</sup> for the mixture-based one. The highest values of moisture content difference were achieved during the third cycle for all composites. After this cycle, the samples started to stabilize.

To investigate in more detail the correlation between the materials and the composites, the average values for all were considered. They are reported in Figure 5.2-8.

These results show the strong influence of the sodium silicate on the final performance. Indeed, by adding the sodium silicate solution, the hygroscopicity and the moisture storage capacity of the bio-aggregates strongly increased. For example, as Figure 5.2-8 shows,

considering the last cycle, from values of moisture content between 0.03 kg/m<sup>2</sup> and 0.06 kg/m<sup>2</sup> after 84 h for the bio-aggregates, the sodium silicate allowed achieving values in the range of 0.20–0.33 kg/m<sup>2</sup> after 84 h. The moisture content difference,  $\rho_{A,SC}$ , increased, too. This is in line with past studies that demonstrated that the adhesives (binder/glue) strongly affected the final properties of the composites [19,21,60]. The advantage here, in comparison with other more common binders, is that the sodium silicate solution has a significative positive effect on the hygroscopic capacity.

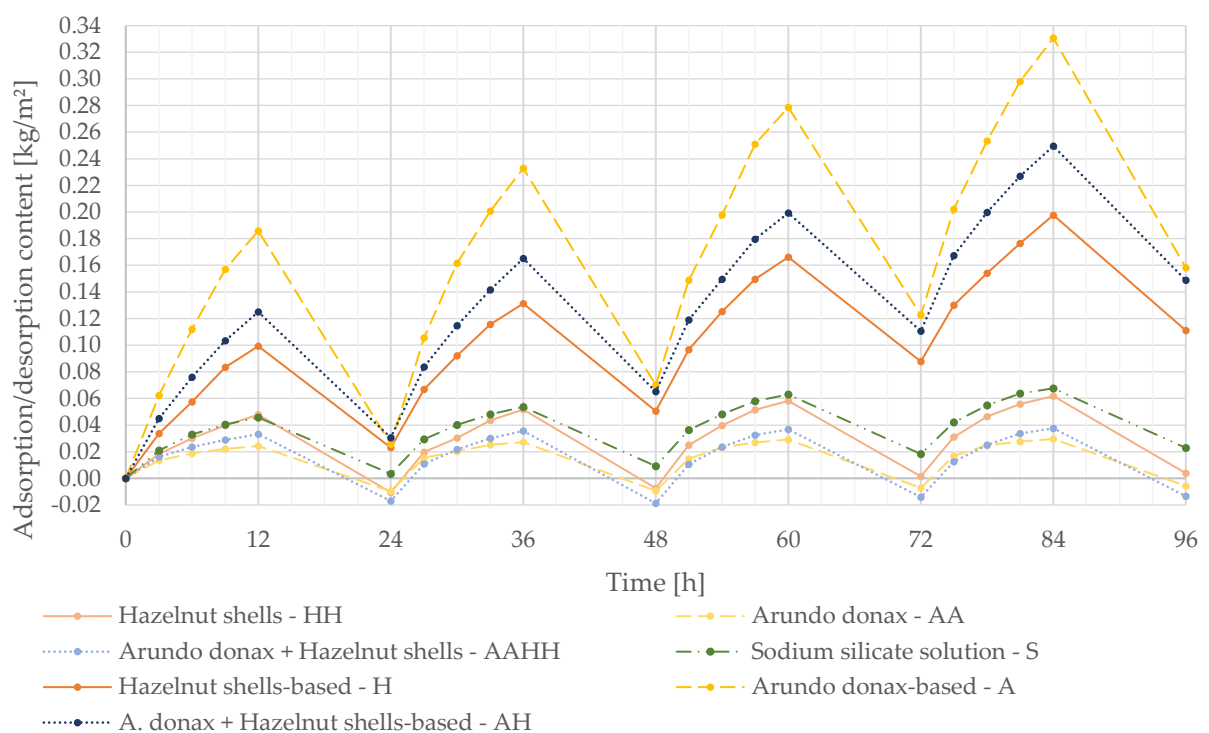


Figure 5.2-8. Moisture adsorption/desorption contents for four cycles of the tested samples: comparison between materials and composites (average values).

Comparing the hygroscopicity of the raw materials and the composites, there was no correspondence. Indeed, the most hygroscopic raw materials (hazelnut shells) did not produce the most hygroscopic composite (which was the *A. donax*-based one). Hence, a correlation between the aggregates and composites cannot be defined. This was caused by the combination of the sodium silicate–aggregate, in particular, by the different sodium silicate distributions on the exposed surfaces. Indeed, according to Figure 5.2-5, the *A. donax*-based samples had a greater amount of sodium silicate on one side, and this could have significantly affected their hygroscopic behavior. The results and the practical experience suggest that the greater the surface area covered by sodium silicate, the greater the capacity to absorb moisture.

Furthermore, the mismatch between the materials and composites demonstrates, again, the strong influence of the adhesive on the performance of the final composites. It is important to underline that the sodium silicate distribution in the composites was assessed only qualitatively, and further analysis should be carried out. However, the results already indicate this correlation between the two properties.

Table 5.2-4 reports the moisture buffering values and the classification, following Rode et al. [64], for both the materials—considering the aggregates and adhesive individually—and the composites (with the average values of the last three cycles and standard deviation), calculated as described in Equations (5.2-4) and (5.2-5) (Section 5.2.3.3).

Table 5.2-4. Moisture buffering values (MBVs) of the materials and composite samples, considering the average values and standard deviations (SDs) of the three samples for each material/composite.

| Samples              | MBV [g/(m <sup>2</sup> % RH)] | Classification (Rode et al. [64]) |
|----------------------|-------------------------------|-----------------------------------|
| Materials            |                               |                                   |
| <i>A. donax</i> - AA | 1.47 ± 0.04                   | Good                              |
| Hazelnut shells - HH | 2.41 ± 0.14                   | Excellent                         |
| Mixture - AAHH       | 2.11 ± 0.03                   | Excellent                         |
| Sodium silicate - S  | 1.92 ± 0.18                   | Good                              |
| Composites           |                               |                                   |
| <i>A. donax</i> - A  | 7.44 ± 1.25                   | Excellent                         |
| Hazelnut shells - H  | 3.86 ± 0.83                   | Excellent                         |
| Mixture - AH         | 4.65 ± 1.12                   | Excellent                         |

Considering the materials, both the *A. donax* and the dried sodium silicate solution were rated as good. Hence, they had lower MBVs than the hazelnut shells. Coherently, the addition of *A. donax* to the hazelnut shells resulted in a lower MBV. As for the sodium silicate result, a higher value of MBV was expected. However, this result was evaluated by considering the average values between the MBV<sub>a</sub> and MBV<sub>d</sub> of the last three cycles. Considering these separately, the MBV of the sodium silicate solution was MBV<sub>a</sub> = 2.05 ± 0.10 g/(m<sup>2</sup>%RH) and MBV<sub>d</sub> = 1.79 ± 0.01 g/(m<sup>2</sup>%RH). Hence, as expected, the MBV of the sorption phase was higher than that of the desorption phase (Table 5.2-4 reports its average value).

The MBVs of the composites are coherent with the previous results (Figure 5.2-7). Again, the values of *A. donax* were influenced by the distribution of the sodium silicate, which determined higher values. As for the mixture, the results are in line with the expectation: the

combination of the two materials determined a lowering of the moisture buffering capacity, and then of the MBV.

#### 5.2.4.4 Statistical analysis

Table 5.2-5 summarizes the results of the ANOVA (reporting F and  $p$ -values).

Table 5.2-5. Results of ANOVA (F and  $p$ -value) considering raw materials as the variable.

| Property         | F    | $p$ -value |
|------------------|------|------------|
| Apparent density | 9.53 | 0.013739   |
| MBV*             | 9.33 | 0.008070   |
| Porosity         | 5.15 | 0.049987   |

\*Note: For MBV, both the materials (individually) and the composites were considered as the variable

The statistical analysis showed that considering the materials as the variable, the alternative hypothesis was confirmed ( $F < 1$ ; the  $p$ -value was less than 0.05; and the  $H_0$  was false, as described in Section 5.2.3.4) for all. Hence, at least one of the groups was different.

Table 5.2-6 reports the  $p$ -values of the post hoc tests, defining where the differences were. Indeed, the  $p$ -values allowed identifying which and how the groups were significantly different from each other. The first two columns show which groups were compared with each other, considering the three composites (A, H, and AH) for all the properties, and also the materials individually (AA, HH, and AAHH) for the hygroscopicity. For easy reading of the results, when the materials (individually) are compared with each other, the results are underlined; when they are compared with the composites, they are underlined. *A. donax* is not reported in italics to avoid any confusion.

$p$ -values of less than 0.05 (highlighted in bold in Table 5.2-6) indicated which groups were significantly different. Both for the apparent density and MBV, the results for the *A. donax*-based and hazelnut-shell-based composites defined two distinguishable groups. As for the porosity, a significant difference was found between the hazelnut-shell-based composite and the mixture-based one. This confirms the observations reported in the previous section (Section 5.2.4.2): the porosity was determined more by the combination of the sodium silicate–aggregate than by the type of aggregate.

Table 5.2-6. p-values of the post hoc tests for the results that were significant (considering adhesives as the variable).

| Group A                                    | Group B                                    | p-value *    |
|--------------------------------------------|--------------------------------------------|--------------|
| Apparent Density                           |                                            |              |
| <b>A. donax-based composite - A</b>        | <b>Hazelnut shell-based composite - H</b>  | <b>0.012</b> |
| A. donax-based composite - A               | Mixture-based composite - AH               | 0.066        |
| Hazelnut shell-based composite - H         | Mixture-based composite - AH               | 0.380        |
| MBV                                        |                                            |              |
| <b>A. donax-based composite - A</b>        | <b>Hazelnut shell-based composite - H</b>  | <b>0.038</b> |
| A. donax-based composite - A               | Mixture-based composite - AH               | 0.136        |
| Hazelnut shell-based composite - H         | Mixture-based composite - AH               | 0.968        |
| <u>A. donax (raw material) - AA</u>        | <u>Hazelnut shells (raw material) - HH</u> | <u>0.932</u> |
| <u>A. donax (raw material) - AA</u>        | <u>Mixture (raw material) - AAHH</u>       | <u>0.987</u> |
| <u>Hazelnut shells (raw material) - HH</u> | <u>Mixture (raw material) - AAHH</u>       | <u>1.000</u> |
| <i>Hazelnut shells (raw material) - HH</i> | <i>A. donax-based composite - A</i>        | <i>0.004</i> |
| <i>Hazelnut shells (raw material) - HH</i> | <i>Hazelnut shell-based composite - H</i>  | <i>0.709</i> |
| <i>Hazelnut shells (raw material) - HH</i> | <i>Mixture-based composite - AH</i>        | <i>0.305</i> |
| <i>Mixture (raw material) - AAHH</i>       | <i>A. donax-based composite - A</i>        | <i>0.002</i> |
| <i>Mixture (raw material) - AAHH</i>       | <i>Hazelnut shell-based composite - H</i>  | <i>0.539</i> |
| <i>Mixture (raw material) - AAHH</i>       | <i>Mixture-based composite - AH</i>        | <i>0.198</i> |
| <i>A. donax (raw material) - AA</i>        | <i>A. donax-based composite - A</i>        | <i>0.001</i> |
| <i>A. donax (raw material) - AA</i>        | <i>Hazelnut shell-based composite - H</i>  | <i>0.244</i> |
| <i>A. donax (raw material) - AA</i>        | <i>Mixture-based composite - AH</i>        | <i>0.074</i> |
| Porosity                                   |                                            |              |
| A. donax-based composite - A               | Hazelnut shell-based composite - H         | 0.405        |
| A. donax-based composite - A               | Mixture-based composite - AH               | 0.244        |
| <b>Hazelnut shell-based composite - H</b>  | <b>Mixture-based composite - AH</b>        | <b>0.043</b> |

\*Note: The numbers in bold represent p-values of less than 0.05; underlined results represent materials (individually) compared with each other; and italicized results represent materials (individually) compared with the composites.

Considering the comparisons between the raw materials for the MBV (the underlined values in Table 5.2-6), they did not determine distinguishable groups (the p-values were always higher than 0.05). As for the comparison between the materials and composites (values reported in italics in Table 5.2-6), significant differences were always found between the materials (AA, HH, and AAHH) and the *A. donax*-based composite (A). This suggests that for the *A. donax*-based composite, the sodium silicate solution strongly influenced the final performance. This is in line with the previous results, both for hygroscopicity and internal structure (Figure 5.2-5).

#### 5.2.4.5 Additional discussion

In addition to the considerations and comparisons carried out in the previous sections, further discussions were conducted. These ensured a better understanding of the composites'



properties and materials' contributions. Moreover, the benefits and drawbacks of the studied composites were pointed out, as well as their possible applications.

Considering the apparent density, as the addition of *A. donax* lowered the results of the hazelnut-shell-based composite, this could probably improve the insulation thermal properties, too. Density and thermal conductivity are strictly correlated [74,75]. However, this consideration should be verified.

Hygroscopicity, porosity, and internal structure are strictly connected. The porosity includes a large range of pore sizes: macropores derived from the bio-aggregates' arrangement, mesopores within the aggregates and the adhesive, and the micropores of the aggregates and the adhesive. Even if the present study considered only the macroporosity, all these porosities affected the moisture storage capacity, together with the ratio of the aggregate/adhesive and the grain size [62,70]. The moisture sorption/desorption capacity also depends on the surface area available for vapor exchange and other hygroscopic properties [73].

The porosity and the information about the internal structure could provide a better understanding of many other properties, too. They are strictly connected to the final performance of the composite. In addition to the hygroscopicity [62], they could influence the sound absorption and thermal insulation [76,77].

Considering all the results, it was confirmed that the hygroscopicity depended on both the aggregates and the sodium silicate. The latter more significantly affected the moisture storage capacity. As the percentages of the aggregate–adhesive were the same for all samples, the differences in the results were mainly caused by the sodium silicate distribution. This changed due to the types of aggregates, namely, their physical and chemical properties and their reactions with the adhesive. The statistical analysis confirmed these considerations. For the MBV, the distinguishable groups (Table 5.2-6) were the composites with *A. donax* and the sodium silicate solution. According to Figure 5.2-5, these composites showed less homogeneity and a higher amount of sodium silicate on one side of the samples (Section 5.2.4.2).

Bio-aggregates are known to be highly porous and, hence, highly hygroscopic. Knowing their internal structures, too, is useful to understand their properties even better [54]. However, other types of analysis are more suitable for this purpose, such as scanning electron microscopy (SEM), mercury intrusion porosimetry (MIP), and sorption techniques. Future studies could consider these laboratory tests to investigate the correlation between these properties on a micro-scale.

High hygroscopicity could be both an advantage and a drawback. Indeed, hygroscopic building materials and products can passively contribute to the control of indoor conditions

[61] and can secure indoor hygrothermal comfort and better indoor air quality [78,79], which are extremely important for human health and users' well-being [80]. On the other hand, a high moisture storage capacity and moisture content could determine the material's degradation [24]. Furthermore, bio-based composites can be easily attacked by microorganisms due to their chemical composition, their organic nature, and their pH [81,82]. Apart from the aesthetic impact, a biological attack can lower the durability of composites, affect the materials' properties, and restrict their performance, modify their chemical compositions, and compromise their natural structures [83]. Furthermore, biological degradation can affect human health, causing diseases and lowering indoor air quality [84,85].

However, sodium silicate has demonstrated good resistance to biological attack [21] and is known to avoid mold growth and material decomposition [45–47]. The present study confirmed this property, as no biological colonization was detected after the hygroscopicity test. Moreover, as moisture storage can be crucial, determining the critical moisture level [86,87] could be useful to better define this possible drawback and moderate it. Furthermore, some strategies could improve low moisture resistance, such as the addition of additives, such as phosphorus, boron, or silica fume [48].

The results suggest that the hazelnut-shell-based and *A. donax*-based composites could be employed as indoor coating boards, passively regulating internal conditions, hygrothermal comfort, and energy demand [73,88,89]. These types of building composites could also be useful for the preservation of architectural heritage, namely, vernacular buildings. Being bio-based, the employed materials would be compatible with vernacular architecture's typical materials. Their high hygroscopicity and moisture buffering capacity would avoid the creation of water vapor barriers, which is crucial to avoid wall and roof degradation. Moreover, by knowing these types of composites' behaviors, other possibilities could be proposed, which could be suitable for historic architectural heritage, too. A possible effective solution for the bio-based composites could be a superficial application of the sodium silicate solution to both protect the bio-based materials and secure the passive control of indoor hygroscopic conditions. However, this is an idea for future studies, suggested by the analysis of the present study, which should be investigated and confirmed.

## 5.2.5 Conclusions

The present study analyzed *A. donax*-based and hazelnut-shell-based boards produced using a sodium silicate solution as the adhesive. The performed laboratory tests considered their apparent density, internal structure, porosity, and hygroscopicity. The connection between the

composites and the materials' properties and the influence of the distributions of the aggregates and the adhesive were considered, too. Indeed, this research work wanted to investigate the correlation between the final performance of the boards and the contribution of each composing material.

The results allowed the achievement of the following conclusions:

- The hazelnut-shell-based composite had a higher apparent density than the *A. donax*-based one. Therefore, the addition of *A. donax* to the mixture of hazelnut shells and the sodium silicate solution lowered the apparent density. Hence, it could probably improve the thermal performance, too. The results of the replications and the values of standard deviation confirm the good homogeneity between the samples of each composite.
- The tomography enabled knowing the internal structures of the composites. A qualitative evaluation suggested a better homogeneity in the hazelnut-shell-based composite. In the *A. donax*-based composite, the sodium silicate solution was deposited more on one side of the sample (the bottom face). This could affect the final performance, such as the moisture storage capacity.
- The tomography allowed defining the porosity, too. The results demonstrate that this property was mainly determined by the contents of the selected materials (the bio-aggregates and adhesive) rather than by the type of bio-aggregate. The latter affected the type of voids (size and shape) and the sodium silicate distribution.
- Both the materials and the composites were highly hygroscopic, as expected. The addition of the sodium silicate solution increased the hygroscopicity and the moisture storage capacity. Furthermore, the sodium silicate's distribution in the samples significantly influenced their final performance, and this caused a mismatching between the results of the materials and composites. These results were confirmed by the statistical analysis (ANOVA).

The correlations between the properties of the materials (individually) and the composites were defined. Knowing which and how these parameters could influence the final performance is extremely helpful for their optimization and future research development. The strong influence of the adhesive content on the final properties was confirmed, as well as the high hygroscopicity of the considered composites. The results suggest that in the case of the employment of the composites as internal coating boards, they could passively control indoor hygrothermal comfort. This could improve both indoor conditions and internal comfort and lower energy demands. Furthermore, when considered as bio-based boards, they could be compatible with the elements in vernacular architecture. Hence, starting from this research

study, useful solutions for the preservation of cultural architectural heritage could be developed.

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### 5.3 Article I: Bio-based boards made of hazelnut shell and *A. donax* for indoor applications - A solution with good performance in case of fire

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#### Abstract

The present study investigated the reaction to fire of bio-based boards for indoor applications made of *A. donax* and hazelnut shells as aggregates. A sodium silicate solution was employed as the adhesive due to its several advantages. Among others, the possibility of moderating some of the main drawbacks of bio-based building composites, such as the resistance to fire. The considered materials were analysed both individually, to test their inherent properties, and when integrated into the composites, ensuring considerations about materials' influence on the final products' properties. Two different test methods, using a cone calorimeter, were considered and performed. The results showed that the sodium silicate solution avoided flaming and smoking, in case of a constant heat application with and without an igniter (spark), demonstrating the benefit of its use in this type of bio-based composites. Overall, the particleboards demonstrated their ability to comply with fire behaviour consistent with the Class A1 requirements, while the bio-components on themselves were characterized by an intermediate fire risk propensity. Thus, the present study provided an effective solution to avoid one of the main drawbacks of bio-based composites. It demonstrated the feasibility of employing the proposed bio-based boards as indoor coating, with no risk to human life in case of fire.

**Keywords:** *Arundo donax*; Fire behaviour; Giant reed; Hazelnut shell; Sodium silicate.

| Nomenclature, Symbols and Acronyms |                                   |       |                              |
|------------------------------------|-----------------------------------|-------|------------------------------|
| A                                  | <i>A. donax</i>                   | pHRR  | Peak Heat Release Rate       |
| H                                  | Hazelnut shells                   | THR   | Total Heat Release           |
| AH                                 | <i>A. donax</i> + Hazelnut shells | TGA   | Thermogravimetric analysis   |
| S                                  | Sodium silicate solution          | SPR   | Smoke Production Rate        |
| AS                                 | <i>A. donax</i> -based composite  | TSP   | Total Smoke Production       |
| HS                                 | Hazelnut shells-based composite   | TTI   | Time To Ignition             |
| AHS                                | Mixture-based composite           | FIGRA | Fire Growth Rate             |
| FR                                 | Flame Retardant                   | EHC   | Effective Heat of Combustion |
| HRR                                | Heat Release Rate                 | SBI   | Single Burning Item          |

### 5.3.1 Introduction

Due to the increasingly serious environmental problems, mainly caused by the construction sector, nowadays research is focused on proposing and evaluating the performance of sustainable solutions [1]. Several possibilities are being investigated, such as new building products, innovative construction practices, and environmentally friendly materials.

Particleboards present many advantages: easy on-site installation, flexibility of applications, and low costs [2,3]; they have a large diversity of applications, and a significant volume of production is required worldwide. Furthermore, they can be produced by using both wood chips and alternative materials, such as bio-fibres and wastes derived from agro-industrial practices and forest cleaning [4,5]. As these materials are environmentally friendly [6], they have many benefits: they lower energy consumption [7] and the composites embodied energy, guarantee circular economy practices [8], and secure the disposal of bio-wastes as well, which could cause environmental problems [9–11]. Due to their several promising properties, studies on bio-based particleboards are more and more carried out [12,13].

Depending on the employed bio-based materials, different performances could be achieved. For example, Mati-Baouche et al. [14] considered a composite made of sunflower stalks bounded by chitosan that demonstrated promising acoustic absorption, guaranteed by porosity. Paiva et al. [15] evaluated the thermal insulation performance of corn cob particleboards and achieved advantageous values thanks to the raw material properties, as demonstrated also by Bovo et al. [16]. Nadhari et al. [17] assessed the mechanical properties of banana trunk waste binderless particleboards and reported encouraging results.

Bio-based particleboards, and bio-based composites in general, in indoor applications, could improve hygrothermal and acoustic comfort [18] passively contributing to indoor

conditions and lowering operational costs [19]. The hygrothermal comfort is related to several factors, such as temperature and relative humidity [20]. The acoustic one is related to the protection against noise and is mainly guaranteed by adequate sound insulation [21], and proper distribution of the materials, as it may depend on both their layout and sound absorption capacity [22]. Both comforts are important to guarantee safe and healthy places for the users.

On the other side, bio-based particleboards can present some drawbacks due to their intrinsic organic (typically cellulosic) matrix [23]. They showed, indeed, low water resistance [24,25], they were prone to biological colonisation [26–28], such as mould growth, fungi development, and insect attack [29], and they displayed poor behaviour in case of fire [30,31]. The latter, in particular, posed a serious threat to the indoor applicability of bio-based particleboards, since cellulosic waste residues typically underwent decomposition at moderate temperatures, often below 300–400°C [32,33]. Thermal degradation resulted in the generation of heat, emission of smoke and soot, and the release of volatile substances able to ignite [34]. In response, Flame Retardant (FR) systems were frequently employed to mitigate or hamper the thermal decomposition and combustion processes inherent in combustible materials like polymer composites [35]. These systems were designed to intervene in various stages of polymer combustion [35], either physically or chemically, affecting the heating, pyrolysis, ignition, and propagation stages of thermal degradation processes.

The enhancement of the behaviour in case of fire remains a critical challenge in contemporary industrial settings, prompting the application of several different approaches. Among these, the application of coatings with FR materials [36,37], the incorporation of FR materials into composites [38], or the utilization of inherently flame-resistant matrices can be chosen depending on the specific requirements. The application of traditionally formulated FR additives or coatings (based mainly on halogens), employing principles ranging from gas phase intervention to interrupt radicals chain reaction propagation, has recently faced scrutiny due to environmental concerns [39]. Hence, the exploration of alternative FR materials and systems has been required. Organometallic complexes, particularly ferrocene-based derivatives, emerged as a promising avenue, demonstrating catalytic properties in char formation that effectively suppress smoke [40,41]. Nanofillers were also positively utilized in FR systems, primarily to promote the formation of a protective intumescent layer in the condensed phase, hindering oxygen penetration, reducing the transfer of volatile gases to the flame, and, ultimately, extinguishing it.

However, for particleboards, these options have limited applicability due to the intrinsic heterogeneous nature of the products, which would imply the use of a dispersed matrix phase to be modified with these approaches. When dealing with heterogeneous aggregates for particleboards, the selection of inherently FR matrices represents a viable route to produce FR composites. Table 5.3-1 reports some past studies that assessed this possibility.

Table 5.3-1. Past studies on fire retardants for boards and particleboards.

| Type of board                                                                                                                                                                                                            | Fire retardant                                                                            | Ref. |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------|------|
| Particleboards made of wood chips and urea-formaldehyde, produced by pressing at 160 bar and 130°C for 8 minutes                                                                                                         | Wollastonite nanofibers                                                                   | [42] |
| Particleboards made of wood chips, expandable polystyrene, a mixture of Polymethyl acrylic, expandable graphite, and melamine polyphosphate, produced at 130°C for 55 minutes                                            | A mixture of polymethyl acrylic emulsion, expandable graphite, and melamine polyphosphate | [43] |
| Particleboards made of walnut shells and urea-formaldehyde and phenol-formaldehyde binder, produced by a hydraulic press at 90°C with 100 atm. for 19 minutes                                                            | Urea-formaldehyde blend                                                                   | [44] |
| Particleboards made of hemp fibres, rice husk, ammonium dihydrogen phosphate and a mixture of water and starch, produced by pressing at 10Mpa, 140°C for 6 minutes                                                       | Ammonium dihydrogen phosphate                                                             | [45] |
| Particleboards made of poplar wood powder treated with ammonium tris isocyanurate triphosphate, and urea-formaldehyde resin, produced in a hydraulic press at 165°C and 2-3 MPa for 8 minutes                            | Ammonium tris isocyanurate triphosphate                                                   | [46] |
| Plywood boards made of a thin layer of wood, melamine formaldehyde resin and phosphate ester acid, produced at room temperature; the solution containing the flame retardant was directly applied on the boards          | Montmorillonite polyphosphate                                                             | [47] |
| Plywood boards made of a thin layer of wood, melamine formaldehyde resin and phosphate ester acid, produced at room temperature by dispersing the solution containing the flame retardant directly on the plywood panels | Cyclic acid phosphate ester loaded with nano-silica                                       | [48] |

According to complementary past studies [49,50], sodium silicate showed interesting properties as a fire protector. It is known as "*water glass*", and it is a non-toxic and environment-friendly material [51,52]. It was employed in several sectors due to its benefits and economic convenience [53]: for detergents and cleaning compounds [53], for materials treatments [52] for ceramics and geopolymers [54,55], as a remediation tool for water [56], as binders in bio-based building boards [50,51]. As for this latter, the sodium silicate guaranteed both the

feasibility of boards' production [49] and the reduction of some of the main bio-based materials' weaknesses, such as biological attack [57]. Hence, silicon-based composites could be an efficient possibility to explore. With this approach, the issue of flammability could be conveniently overcome, ensuring the production of sustainable particleboards made of agro-wastes and bio-resources. Differently from traditional FR, sodium silicate is a safe material that, when released into the environment, does not bring along specific concerns [51,52]. Thus, even in the aftermath of a potential fire, the discarded waste residue based on sodium silicate aggregate should not pose additional environmental safety concerns.

Considering this background, the present study investigated bio-based particleboards for indoor applications by using a sodium silicate solution as the binding matrix, for its several benefits. Among others, its good resistance to fire [49,50]. As for the aggregates, giant reed (*Arundo donax* L.) and hazelnut shells were considered according to past studies [18,58]. Besides the advantages of bio-based materials, both are widely available all over the year in the Euro-Mediterranean area [23], avoiding the problem of the seasonality of bio-wastes/bio-resources, which could discourage their use for building composites' production. Both hazelnut shells-based and giant reed-based composites demonstrated promising properties to improve indoor comfort, such as high hygroscopicity and interesting sound absorption capacity. Hence, they could be suitable and effective as indoor coating boards [18,58]. However, to guarantee their application as such, their performance in case of fire must be verified.

Although fire behaviour has not been widely investigated yet [59], some past studies considered this property by assessing ignition, incombustibility, and Heat Release Rate [60], as well as the smoke and toxic gas production, one of the most common causes of fire deaths [61]. Different test methods could be considered to evaluate the fire reaction of bio-based materials. For example, Palumbo et al. [62] described a small-scale flammability test, performed on a fire testing technology pyrolysis combustion flow calorimeter. The researchers considered a bio-based board made of corn pith and a natural gum (sodium alginate) and determined the Heat Release Rate (HRR) and the Total Heat Release (THR). They considered boric acid as a possible effective solution to improve the performance in case of fire. Rocha et al. [60] tested the fire reaction of bamboo, wood, and rice husk wastes by using a cone calorimeter. They analysed also their physical, chemical, and thermal properties, by performing the thermogravimetric analysis (TGA). The researchers determined the HRR and the THR and considered a possible treatment to improve the performance in case of fire of these bio-aggregates (e.g., alkali treatment, which could reduce heat release parameters for wood and bamboo). Antunes et al. [19] performed a non-standardised test to assess the performance of a rice husk-earth-



gypsum-air lime composite in case of fire. They exposed the considered composite to a flame near a border with a resource to a torch. In all cases, the results of the fire-reaction tests allowed rating the materials according to their performance, verifying if they meet the standard requirements, and suggesting possible improvements if necessary (e.g., application of fire retardants). Nevertheless, further investigation on bio-based composites' performance in case of fire should be performed to improve this property [63].

Starting from these considerations and considering past studies, this research work addresses the fire reaction of the *A. donax*-based and hazelnut shells-based boards with sodium silicate binding matrix by using a cone calorimeter. Some of the previously cited studies described in detail the research project that investigated and analysed the proposed bio-boards. First, bibliographic research was carried out to select some bio-aggregates [23], which were tested to provide their characterization as raw materials [64]. After that, different adhesives were considered and assessed [57], and the sodium silicate was selected. Then, the bio-based boards made of bio-aggregates and sodium silicate were produced and tested. The most suitable production process was investigated [65], and a horizontal analysis was performed, by assessing different properties, such as the hygrothermal, acoustic, and mechanical ones [18,58]. Finally, the influence of materials formulation was investigated by considering the hygroscopic properties [66]. The research project, described in the reported references, suggested that the hazelnut shell-based and the *A. donax*-based boards could be employed as indoor coatings. As it can be an important drawback for this type of application, the present study wants to complement their characterization, evaluating their performance in case of fire.

The aim is to evaluate the reaction to fire of the materials, considered individually (bio-aggregates and the dried sodium silicate solution), and the composites produced with them. Hence, in addition to the reaction to fire, considerations on the influence of each material and the correlation between materials and composites will be made, supported by TGA characterization. Furthermore, this study wants to assess, by laboratory experience, the sodium silicate's benefit, also when it is employed as an adhesive in bio-based boards.

## 5.3.2 Materials and samples production

### 5.3.2.1 Bio-aggregates and adhesive

*Arundo donax* L., known also as giant reed and/or giant cane, was provided by cultivations in Cadriano and Ozzano dell'Emilia, Bologna. Hazelnut shells were provided by Raccolti di Cin, Baldissero d'Alba, Cuneo, Italy. Both the aggregates were shredded, using a mechanical mill,

to have grain sizes between 4 – 8 mm (Figure 5.3-1), dried in an oven at  $T = 60^{\circ}\text{C}$  until constant mass (variation in mass  $\leq 0.1\%$  after 24 hours), and characterized as described by Cintura et al. [18,58].

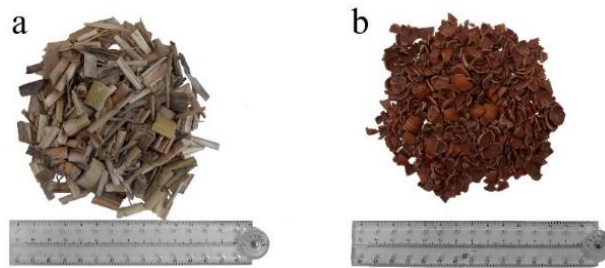


Figure 5.3-1. Bio-aggregates after shredding: (a) *A. donax*, (b) hazelnut shells (adapted from [18,58]).

A sodium silicate solution,  $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ , with the properties reported in Table 5.3-2, was provided by Ingessil Srl, Montorio, Verona, Italy [67].

Table 5.3-2. Characteristic of the sodium silicate solution provided by Ingessil Srl.

| Property                                   | Value |
|--------------------------------------------|-------|
| Weight ratio [-]                           | 2.40  |
| Density [ $^{\circ}\text{B}^{\circ}$ ]     | 46.45 |
| Molar ratio [-]                            | 2.48  |
| Sodium silicate concentration [% p/p]      | 41.33 |
| $\text{SiO}_2$ [% p/p]                     | 29.17 |
| $\text{Na}_2\text{O}$ [% p/p]              | 12.16 |
| Density [g/ml] at $T = 20^{\circ}\text{C}$ | 1.471 |
| pH ( $T = 20^{\circ}\text{C}$ )            | 12.40 |

The bio-aggregates were placed in plastic boxes without any binder; they were tested individually (100% of the considered volume of *A. donax* or hazelnut shells) and as a mixture (50% of the considered volume of *A. donax* and 50% of hazelnut shells). The sodium silicate solution was placed into three silicon moulds 10 cm x 10 cm x 4 cm, filled at a height varying between 2 – 4 mm and it was dried at laboratory conditions. The materials (both the aggregates and the sodium silicate) were stabilized at  $T = 20^{\circ}\text{C}$  and  $\text{RH} = 60\%$  in a conditioned room, and then tested. For the samples' designation, the first letter of the material was considered: **A** = *A. donax*; **H** = Hazelnut shells; **AH** = *A. donax* + Hazelnut shells; **S** = Sodium silicate solution.

### 5.3.2.2 Composite samples

The composite samples were produced as described by Cintura et al. [18,58], considering the same quantities of the materials, namely, 70% aggregates - 30% sodium silicate binder

(percentages by volume). This ratio was selected after several practical tests, not reported for the sake of brevity, and detailed in previous studies [57,65] where other properties of the composites were assessed. The aim was to maximize the content of bio-wastes/bio-resources, minimize the sodium silicate solution, and guarantee enough mechanical resistance, avoiding the breaking of the samples.

Three composites were produced and consisted of:

- 70% of *A. donax* and 30% of sodium silicate solution
- 70% of hazelnut shells and 30% of sodium silicate solution
- 35% of *A. donax*, 35% of hazelnut shells, and 30% of sodium silicate solution.

The bio-aggregates and the binder were mechanically mixed for 10 minutes, then placed into moulds, and levelled by a spatula without compaction. The production process was described in more detail in the reference studies [18,58]. The moulds were closed, placed at  $T = 60^{\circ}\text{C}$  for 3 hours, then at laboratory conditions for 2 days, always rotated to secure a homogeneous distribution of the sodium silicate solution [65]. Finally, the samples were demoulded and cured at laboratory conditions for 28 days and dried at  $T = 50^{\circ}\text{C}$  until constant mass (variation in mass less than 0.5% after 24 hours) [50,68]. Three samples 10 cm x 10 cm x 4 cm and three cubic samples with 3.5 cm sides for each composite were produced. Then, they were stabilized at  $T = 20^{\circ}\text{C}$  and  $\text{RH} = 60\%$  in a conditioned room and tested.

Again, for the composite samples' designation, the first letters were considered: **AS** = *A. donax* + Sodium silicate solution; **HS** = Hazelnut shells + Sodium silicate solution; **AHS** = *A. donax* + Hazelnut shells + Sodium silicate solution.

Table 5.3-3 reports the apparent density of the composite samples, evaluated in a past study [66].

Table 5.3-3. Characteristics of the obtained bio-composites: the apparent density of the composite samples adapted from Cintura et al. [66] and the inorganic residue evaluated via muffle oven treatment.

| Composite                             | Apparent density [ $\text{kg/m}^3$ ] | Inorganic residue [%] |
|---------------------------------------|--------------------------------------|-----------------------|
| <i>A. donax</i> -based composite – AS | $470 \pm 75$                         | 35                    |
| Hazelnut shells-based composite – HS  | $677 \pm 47$                         | 23                    |
| Mixture-based composite – AHS         | $607 \pm 52$                         | 34                    |

### 5.3.3 Methods

The performed laboratory tests, and the number and size of the samples, were reported in the following sections.

#### 5.3.3.1 Bio-aggregates and composites characterization

The thermal stability, water content, and the inorganic fraction composition of the considered bio-aggregates (i.e., *A. donax*, A, and hazelnut shells, H), were assessed by thermogravimetric analysis, TGA, using a TA Instruments TGA Q550 Discovery Series under a nitrogen atmosphere (flow rate 60 mL/min) at a 10°C/min heating rate, from 25°C to 800°C using a Pt pan.

As for the composite samples (AS, HS, AHS), the evaluation of their composition was carried out by thermal treatment. Three cubic samples for each composite, with 3.5 cm (Figures 5.3-2a, 5.3-2b, and 5.3-2c) were weighed ( $m_0$ ), placed in a muffle furnace at  $T = 900^\circ\text{C}$  (Figure 5.3-2d) until the complete degradation, and then weighed again ( $m_f$ ).

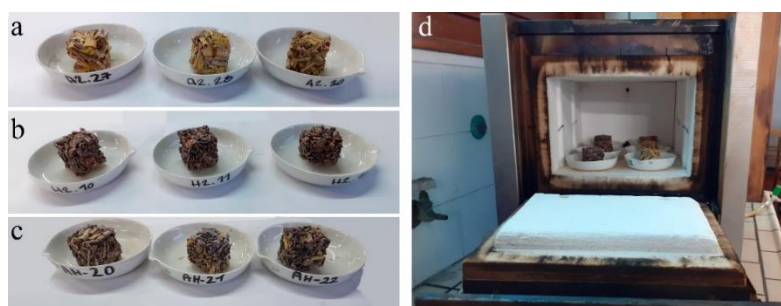


Figure 5.3-2. Evaluation of the inorganic fraction of bio-aggregate composite cubic samples (3.5 cm): a) *A. donax*-based; b) hazelnut shells-based; c) mixture AH-based; d) samples into the muffle furnace.

#### 5.3.3.2 Reaction to fire

Reaction to fire was evaluated by using an oxygen consumption cone calorimeter (Fire Testing Technology Limiter FFT Cone Calorimeter) (Figure 5.3-3a), according to ISO 5660-1 [69]. This method considers the correlation between the net heat of combustion and the required oxygen and consists of burning the samples in ambient conditions. The cone calorimeter is one of the most effective bench-scale methods for studying the flammability properties of materials [70].

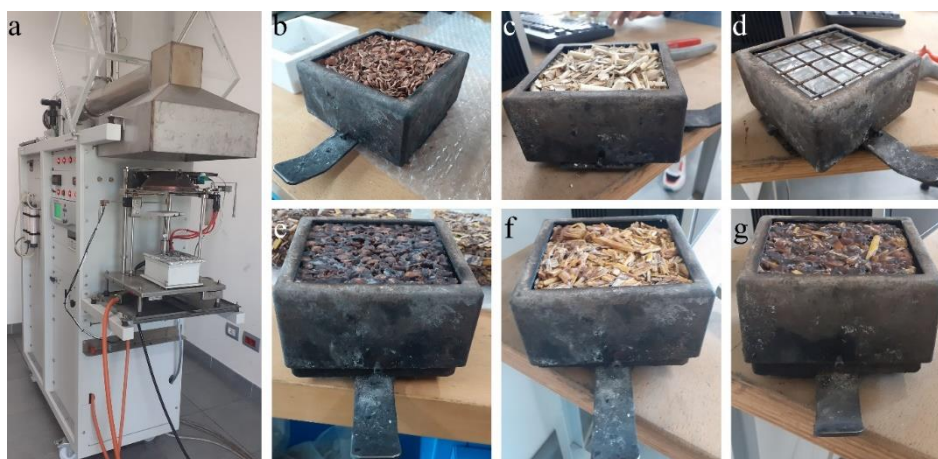


Figure 5.3-3. Test to evaluate the fire reaction by exposing the materials to a constant heat source. a) Cone calorimeter; samples into the metal mould (10 cm x 10 cm x 4 cm) before the test: b) hazelnut shells H, c) *A. donax* A; d) air-dried sodium silicate solution S; e) hazelnut shells-based composite HS; f) *A. donax*-based composite AS; g) mixture-based composites AHS.

The tests were carried out with a heat flow of  $35 \text{ kW/m}^2$  (about  $650^\circ\text{C}$ ) which can be roughly compared to a small-scale fire. The instrument could work with a heat flow ranging between 5 and  $100 \text{ kW/m}^2$ . The higher irradiation levels led to better reproducibility, more clearly defined ignition time, and shorter measurement times. Furthermore, they also corresponded to more fully developed fire conditions. Hence, often a smaller irradiation level better fits the fire protection goals. Indeed, it accounts for the material behaviour in a condition close to the initial stage of a fire, showing its ability – or even better, inability – to ignite and propagate, and finally, to contribute to the fire expansion. This condition seems the most interesting for a preliminary evaluation of particleboards employed as building components which, as such, had an important role in fire safety conditions.

Among the several properties provided by the tests, the Heat Release Rate (HRR), the pHRR (Peak Heat Release Rate), the Total Heat Release (THR), the Smoke Production Rate (SPR) and Total Smoke Production (TSP) were considered. HRR is the heat released per unit area for samples under a constantly imposed heat flux; pHRR is the maximum value of the heat release rate profile. THR is the potential for thermal energy available for the combustion of the material, which increases the probability of fire [60]. As for the smoke production, described by SPR and TSP, it was analysed due to its dangerousness in case of fire [61,71,72].

Two different tests were performed and, in the present study, were identified as “Test Method A” (non-combustibility test) and “Test Method B” (Single Burning Item test, SBI). Both would require different equipment than the cone calorimeter. However, the results presented

hereafter were intended for a preliminary evaluation of the potential of the proposed composites in terms of performance in case of fire.

#### 5.3.3.2.1 Test method A (simulating non-combustibility test)

Test Method A requires that the samples are subjected to a heat flow for 30 minutes, in the absence of ignition, simulating as much as possible what is specified in EN ISO 13501 [73] and EN ISO 1182 [74]. The lack of an external sparkle ignition is applied to highlight the intrinsic risk of fire of a material or composite, trying to assess its potentially non-combustible behaviour. The latter, indeed, accounts for the safest class of products to be used in building and construction.

One sample for each material (A\_01, H\_01, AH\_01, S\_01) was placed into a metal standardized mould 10 cm x 10 cm x 4 cm. For the bio-aggregates, the mould was filled until the top (Figures 5.3-3b and c). For the dried sodium silicate solution, the sample was placed up to non-flammable support and then into the mould. This allowed it to reach the top since the upper surface of the sample should be at a standardized distance from the cone. A metallic net was used to retain the samples during the test (Figure 5.3-3d) avoiding expansion which would diminish the distance from the heat source in the cone. As for the composites, one sample for each one (AS\_01, HS\_01, AHS\_01), 10 cm x 10 cm x 4 cm, was placed into the metal mould (Figures 5.3-3e, f, and g). The mould was, then, placed under the cone heater at a constant distance of 25 mm. A constant heat flux of 35 kW/m<sup>2</sup> (corresponding to a temperature between 650°C and 670°C) was applied for 30 minutes (without an igniter). In addition to the previously reported properties, by Test Method A, the flashover index, evaluated as pHRR/TTI (where TTI represents the Time to Ignition), was determined.

#### 5.3.3.2.2 Test method B (simulating Single Burning Item)

Test Method B is a complementary testing condition to the previously presented Test Method A. It was designed to simulate the flame exposure that would be experienced by coatings on the walls of a room when a "wastepaper basket" ignites adjacent to the coatings itself. Then, to simulate these conditions, a sparkle ignition is presented, representing the threat of an adjacent fire which would surely set the volatiles on fire.

In Test Method B, samples were subjected to heat flow for 10 minutes in the presence of ignition, simulating as much as possible what is specified in EN ISO 13501 [73]. This should ensure the simulation and the achievement of data correlated to an analysis carried out according to EN ISO 13823 [75], namely the Single Burning Item (SBI) test. The remaining two replicas of each material (A\_02 - 03, H\_02 - 03, AH\_02 - 03, S\_02 - 03) and composite (AS\_02 - 03, HS\_02 - 03, AHS\_02 - 03) were placed into the metal mould, as previously described. The

composite samples, originally with a thickness of 4 cm, were sanded by a mechanical roller with sandpaper to reach a thickness of 2 cm. This ensured enough distance between the sample's surface and the source of the spark. The mould was placed under the cone shape (constant distance = 25 mm) and as previously anticipated, in addition to the constant heat flux of 35 kW/m<sup>2</sup>, a spark (igniter) was generated. Besides the previously mentioned properties, by Test Method B, the Fire Growth Rate, FIGRA<sub>0,2MJ</sub> index, was determined.

## 5.3.4 Results

### 5.3.4.1 Bio-aggregates and composites characterization

The curves obtained by the TGA test on the bio-aggregates are reported in Figures 5.3-4a and b.

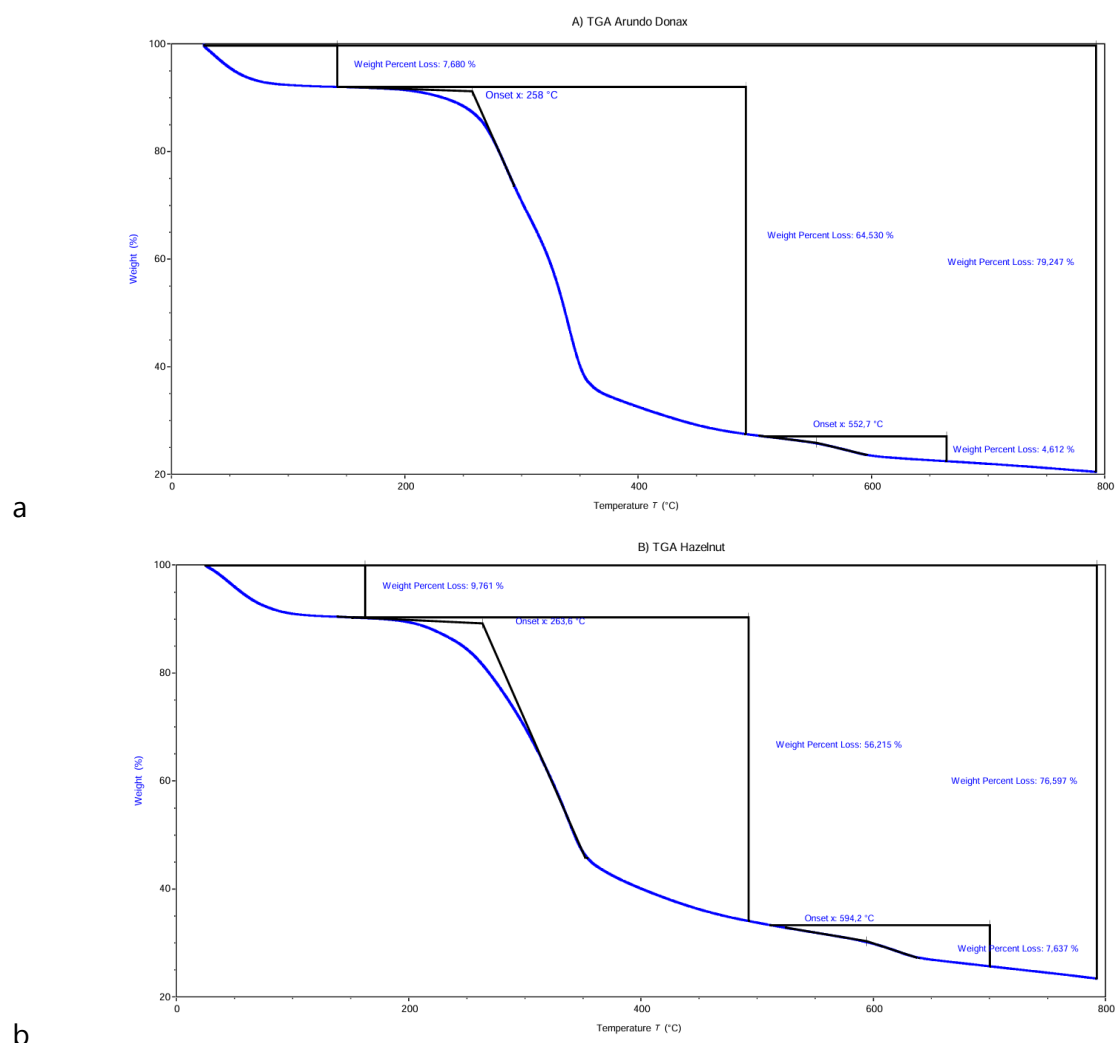


Figure 5.3-4. TGA thermograms of the bio-aggregate samples: a) *A. donax*, b) hazelnut shells.

The results showed that, despite their different nature, size, and shape, the bio-aggregates were quite similar in terms of thermal stability, with an onset weight loss located around 260°C. Although *A. donax*, A, appeared more in a fibrous arrangement than hazelnut shells, H (Figure 5.3-1), the water content was not significantly different. Furthermore, while working in a nitrogen atmosphere, the ability to leave a charred residue was slightly higher for H, with about 8% wt char content at 800°C vs. 5% wt in the case of A. The charring ability is usually an indication of the fire-resisting ability of a material [76,77]. Comparing the results of the two bio-aggregates, a similar fire behaviour would be expected.

As for the composite samples, Table 5.3-3 shows the inorganic residue evaluated via muffle oven treatment. Owing to the macroscopic size of the bio-aggregates, it was not possible to run representative TGA tests of the composites to provide a reliable indication of their composition. The results showed that, overall, the binder fraction was slightly higher in the case of the two composites made of *A. donax*, AS and AHS, while the hazelnut-based composite samples, HS, had a lower inorganic residue fraction. This result could be determined by a higher capacity of *A. donax*, more fibrous, to retain the sodium silicate solution than the hazelnut shells, with a different, less fibrous surface in the exterior part of the shell. The significant difference in the inorganic fraction content might affect the overall flame response of the composites.

### 5.3.4.2 Reaction to fire

#### 5.3.4.2.1 Test method A (simulating non-combustibility test)

The results of the test Method A, (application of constant heat, without generating any sparks, and analysing the fire reaction for 30 minutes), are reported in Table 5.3-4.

Table 5.3-4. Reaction to fire tested by Test Method A: results of bio-aggregates and composite samples.

| Sam-<br>ples | Time<br>to Ig-<br>nition<br>[s] | Peak Heat Re-<br>lease Rate<br>(pHRR)<br>[kW/m <sup>2</sup> ] | Heat Release<br>Rate (HRR)<br>[kW/m <sup>2</sup> ] | Total Heat<br>Release<br>(THR)<br>[MJ/m <sup>2</sup> ] | Total Smoke<br>Production<br>(TSP)<br>[m <sup>2</sup> ] | Flashover<br>Index<br>[kW/s.m <sup>2</sup> ] |
|--------------|---------------------------------|---------------------------------------------------------------|----------------------------------------------------|--------------------------------------------------------|---------------------------------------------------------|----------------------------------------------|
| A_01         | 33                              | 200.8                                                         | 10.5                                               | 64.4                                                   | 1.5                                                     | 6.1                                          |
| H_01         | 108                             | 268.0                                                         | 81.5                                               | 145.6                                                  | 4.9                                                     | 2.5                                          |
| AH_01        | 40                              | 217.3                                                         | 10.8                                               | 107.4                                                  | 1.4                                                     | 5.4                                          |
| S_01         | n.d.*                           | 12.4                                                          | n.d.*                                              | 0.2                                                    | 0.9                                                     | n.d.*                                        |
| AS_01        | n.d.*                           | 11.7                                                          | 5.8                                                | 10.4                                                   | 0.5                                                     | n.d.*                                        |
| HS_01        | n.d.*                           | 22.6                                                          | 5.3                                                | 9.5                                                    | 1.3                                                     | n.d.*                                        |
| AHS_01       | n.d.*                           | 12.7                                                          | 5.5                                                | 10.0                                                   | 0.5                                                     | n.d.*                                        |

\*n.d. = not detectable.



Figure 5.3-5 shows the Heat Release Rate (HRR) for Test Method A of materials and composites (the type of the tested materials/composites instead of the samples' designation was reported).

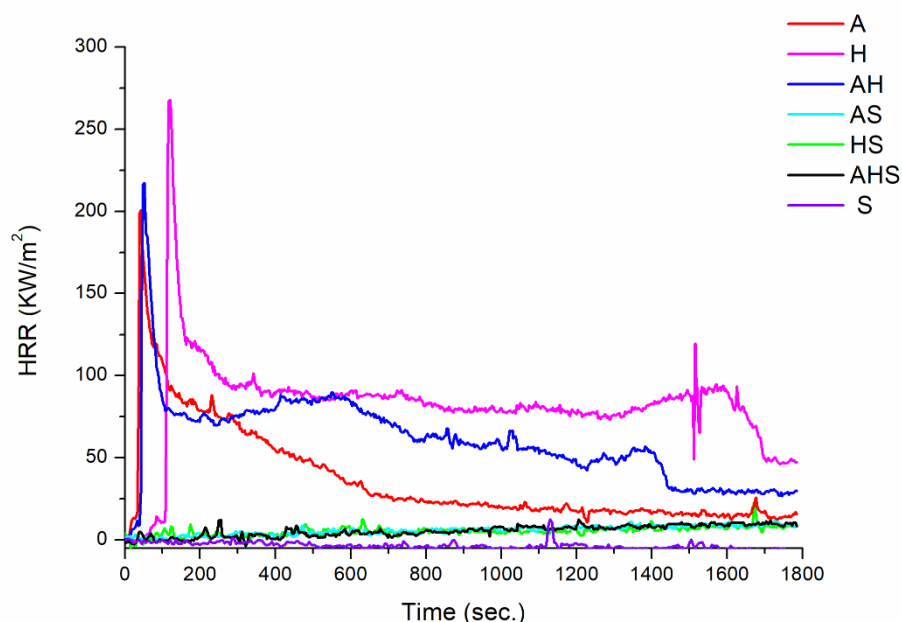


Figure 5.3-5. Heat Release Rate (HRR) of the materials and the composite samples determined by the cone calorimeter test: Test Method A simulating non-combustibility test (reporting the type of the tested materials/composites instead of the samples' designation).

The bio-aggregates curves presented an initial delay step, which represented the time required to reach the degradation temperature and start releasing ignitable volatiles. Then, both A and H curves increased sharply until a peak, reached between the first and the second minute of the test [60]. Both bio-aggregates were extremely fast in reaching the pHRR, with values of  $\text{pHRR} = 268.0 \text{ kW/m}^2$  (at 120 s) for hazelnut shells,  $\text{pHRR} = 217.3 \text{ kW/m}^2$  (at 51 s) for the mixture of the two aggregates, and  $\text{pHRR} = 200.8 \text{ kW/m}^2$  (at 42 s) for *A. donax* (Table 5.3-4).

These results were in line with the results of TGA (Section 5.3.4.1), with a slightly higher charring ability of H, which indeed, delays the peak in the HRR. Even when mixing A and H in a 50/50 volume ratio, the prevailing behaviour was determined by *A. donax* which first ignited and then sparked an anticipated combustion of the hazelnut shells fraction (TTI = 42 s for A, TTI = 52 s for AH). This observation, together with the testing conditions, accounted for the strong and inherent behaviour of the two different bio-aggregates: they were set on fire depending on their tendency to release volatiles upon thermal degradation.

Petrella [78] defined that the ratio of the peak HRR to time to ignition, namely  $\text{pHRR}/\text{TTI}$ , is an indication of the materials' propensity to flashover. Higher values of  $\text{pHRR}/\text{TTI}$  suggested

a greater hazard, and a low tendency to flashover when below 0.1. In this study, all the bio-aggregates entered an intermediate risk region (pHRR/TTI in the 1-10 range). *A. donax* seemed to impart a higher hazard, even mixed with the hazelnut shells, having both A and AH similar flashover tendency, which doubled the one of H. However, the full hazard risk cannot be determined by considering only this parameter.

The Total Heat Release (THR) provides a full picture of the potential fire hazard behaviour since it represents the inherent energy in a material independent of environmental factors. As THR could be calculated by integrating the curve of HRR over time [30], its trend shows how fast the materials achieved the highest values of HRR. The results are represented in Figure 5.3-6.

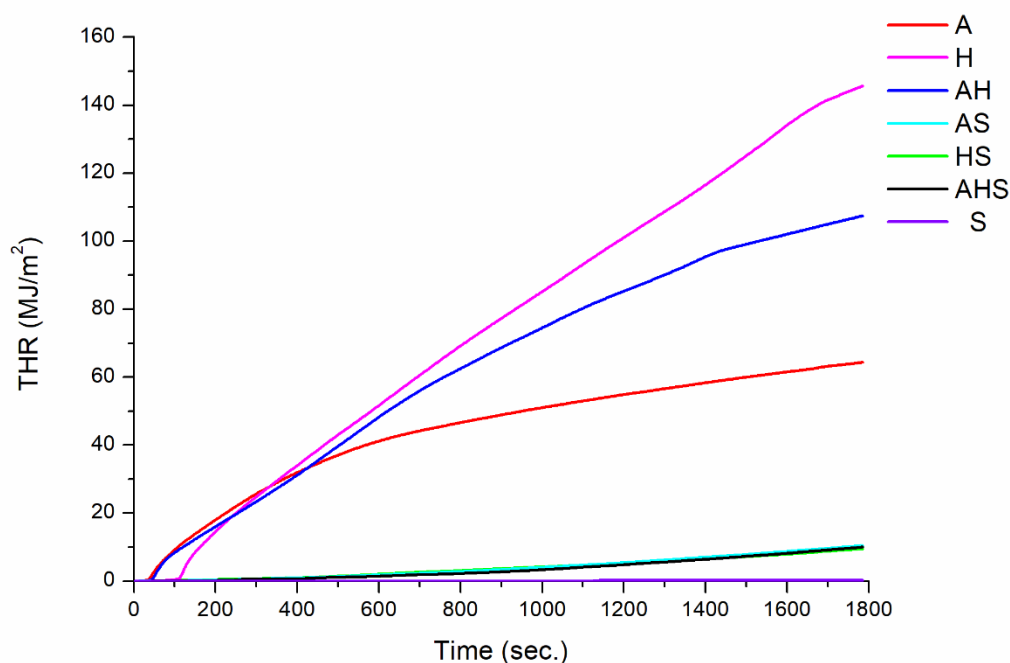


Figure 5.3-6. Total Heat Release (THR) of the materials and the composites determined by the cone calorimeter test: Test Method A simulating non-combustibility test (reporting the type of the tested materials/composites instead of the samples' designation).

The hazelnut shells reached the highest values of HRR the fastest, followed by the mixture of the two aggregates and the *A. donax*. While A set to lower values of Heat Release after the first burst, H instead kept on releasing a higher amount of heat all over the test. Indeed, after the peak, the achieved minimum values are HRR = 46.1 kW/m² (at 1740 s) for hazelnut shells, HRR = 26.6 kW/m² (at 1746 s) for the mixture of the two aggregates, and HRR = 10.7 kW/m² (at 1227 s) for *A. donax*.

When the bio-aggregates were included in a composite structure with sodium silicate as a binder, the fire response was drastically modified. The trend observed for all the composites, regardless of their main component, was almost flat all over the test and the curves did not show any marked peak (Figure 5.3-5).

In the absence of an external ignition source, as in the case of Test Method A, some random sparkle can occur during the 30-minute test span. However, none of them could be considered as sustained flame which represented "*the existence of flame on or over the surface of the specimen for periods of over 10 s*" [69]. Hence, they cannot be used to define the flash-over potential of the sample, which, consequently, was 0, and accounted for intrinsically safe composites in all the cases. This result was outstanding, since the application of such a convenient binder, which already demonstrated potential benefits in hygrothermal absorption [66] and also as a potential barrier for fungi and moulding attacks [57], was able to fully hamper fire development. The performance in case of fire of the bio-aggregates was widely improved by the addition of the sodium silicate solution when producing the composites. Nonetheless, there was a correspondence between the materials and the composites. While not reporting any hazard, the highest values of HRR were achieved by hazelnut shells (both individually and in composites), possibly owing both to the intrinsic ability of H to release more heat but also to the lower sodium silicate content in the composite (both pointing at the same result). The addition of *A. donax* to hazelnut shells lowered the maximum values of the HRR both in the bio-aggregates and in the composite. Since the presence of sodium silicate binder hampered completely the peak heat evolution, this mixed approach was beneficial in terms of fire protection, even in the case of fire development.

Another important parameter in fire protection is the prevention of smoke evolution. Indeed, smoke represents a significant threat to people exposed to fires, since hampers visibility slowing down escape, while also producing a significant amount of toxic and potentially lethal products. Figure 5.3-7 reports the Smoke Production Rate (SPR) of the materials and the composite samples.

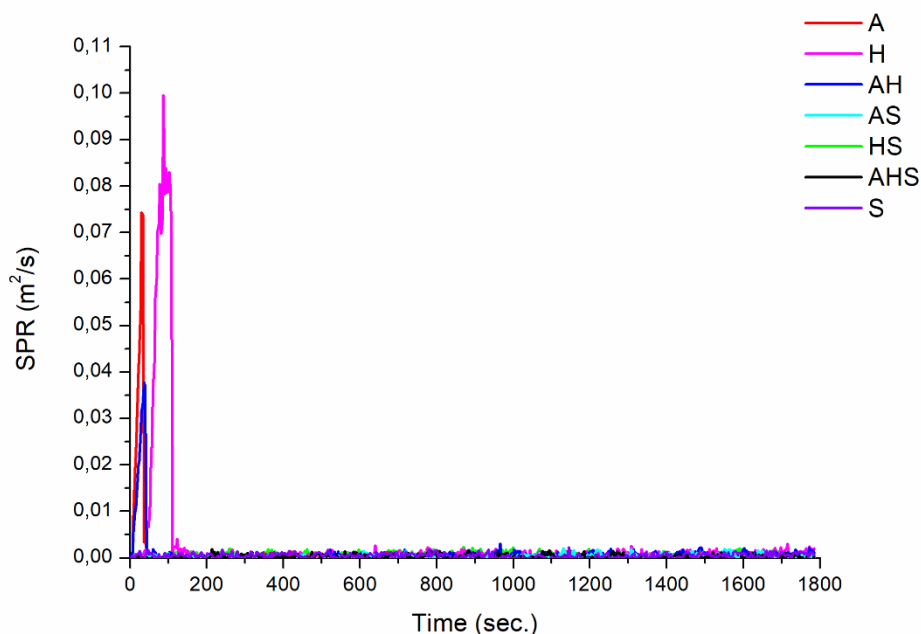


Figure 5.3-7. Smoke Production Rate (SPR) of the materials and the composite samples determined by the cone calorimeter test: Test Method A simulating non-combustibility test (reporting the type of the tested materials/composites instead of the samples' designation).

The principle of the smoke measurement is based on the observation that a volume of combustion products exponentially decreases the intensity of transmitted light as a function of distance from the observation point; hence in the present case, SPR is a measurement of the smoke obscuration ability.

The curves of the SPR were in line with the ones of the HRR for the aggregates, reaching a peak before the first two minutes – maximum values of  $\text{SPR} = 0.100 \text{ m}^2/\text{s}$  (at 87 s) for hazelnut shells,  $\text{SPR} = 0.038 \text{ m}^2/\text{s}$  (at 36 s) for the mixture of the two aggregates,  $\text{SPR} = 0.074 \text{ m}^2/\text{s}$  (at 30 s) for *A. donax*. It is interesting to observe that the smoke production peak significantly anticipated the heat release peak, supporting the hazard of this parameter that, even in the very initial stages of the fire development, represented an insidious threat to human life. The smoke production of the composites was practically insignificant throughout the test for *A. donax* containing composites and this behaviour was, in contrast, a very outstanding result in terms of fire safety and protection of the proposed particleboards. When the hazelnut shells composite was analysed, a higher amount of smoke was released. However, it is worth pointing out that, differently from the aggregates, in the composite the smoke evolved all along the 30-minute test, avoiding an intense fast smoke release, namely a lower threatening condition.

Hence, in all cases, the great impact of the sodium silicate binder presence was evident, avoiding or strongly suppressing and diluting the production of smoke (Figure 5.3-7).

Figure 5.3-8 reports the Total Smoke Production (TSP) for Test Method A of materials and composites.

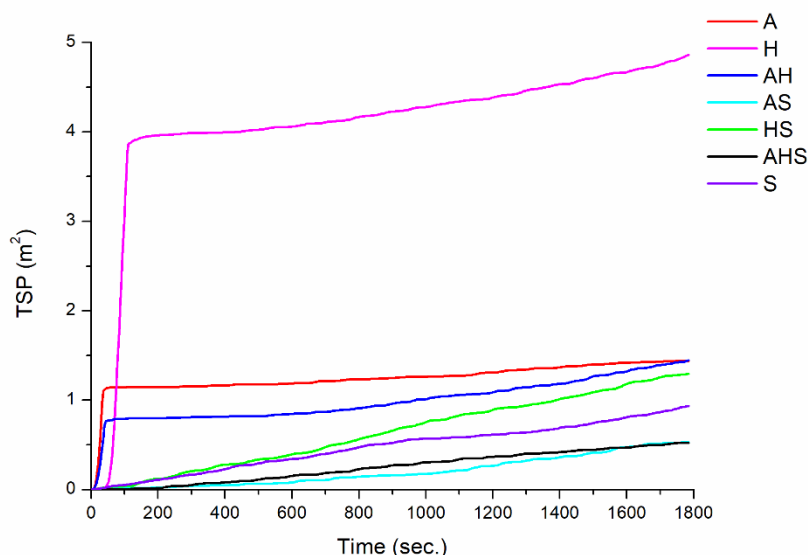


Figure 5.3-8. Total Smoke Production (TSP) of the materials and the composites determined by the cone calorimeter test: Test Method A simulating non-combustibility test (reporting the type of the tested materials/composites instead of the samples' designation).

Analogously to THR and HRR, the TSP showed how fast the materials achieved the highest values of SPR, hence the fastness of the hazard evolution, demonstrating that the small quantities of smoke were released after minutes. The sodium silicate both protected the bio-based materials from burning and improved the smouldering effects, as these properties were closely related [30,60].

#### 5.3.4.2.2 Test method B (simulating of Single Burning Item)

The results of the test Method B, (application of constant heat, generating a spark, and analysing the fire reaction for 10 minutes), are reported in Table 5.3-5.

Figure 5.3-9 reports the Heat Release Rate (HRR) for Test Method B of the materials and the composite samples (as for the designations, the type of the tested composites were reported instead of the samples).

Table 5.3-5. Reaction to fire tested by Test Method B: results of bio-aggregates and composite samples, reporting both tested samples.

| Sam-<br>ples | Time<br>to Ig-<br>nition<br>[s] | Peak Heat<br>Release<br>Rate<br>(pHRR)<br>[kW/m <sup>2</sup> ] | Heat Re-<br>lease Rate<br>(HRR)<br>[kW/m <sup>2</sup> ] | Total<br>Heat Re-<br>lease<br>(THR)<br>[MJ/m <sup>2</sup> ] | Effective<br>heat of<br>combustion<br>(EHC)<br>[MJ/kg] | Total<br>Smoke<br>Produc-<br>tion (TSP)<br>[m <sup>2</sup> ] | Fire Growth<br>Rate<br>(FIGRA <sub>0,2M</sub> )<br>[W/s] |
|--------------|---------------------------------|----------------------------------------------------------------|---------------------------------------------------------|-------------------------------------------------------------|--------------------------------------------------------|--------------------------------------------------------------|----------------------------------------------------------|
| A_02         | 7                               | 171.0                                                          | 48.0                                                    | 28.2                                                        | 14.2                                                   | 0.4                                                          | 7855                                                     |
| A_03         | 8                               | 164.3                                                          | 33.3                                                    | 19.6                                                        | 13.5                                                   | 0.2                                                          | 6242                                                     |
| H_02         | 50                              | 412.5                                                          | 99.1                                                    | 58.2                                                        | 11.1                                                   | 0.3                                                          | 3947                                                     |
| H_03         | 51                              | 400.1                                                          | 88.9                                                    | 52.2                                                        | 11.8                                                   | 0.3                                                          | 3896                                                     |
| AH_02        | 26                              | 242.6                                                          | 83.5                                                    | 50.0                                                        | 7.3                                                    | 0.1                                                          | 4284                                                     |
| AH_03        | 27                              | 241.5                                                          | 82.5                                                    | 52.2                                                        | 7.4                                                    | 0.2                                                          | 4209                                                     |
| S_02         | n.d.*                           | 10.0                                                           | 0.3                                                     | 0.5                                                         | n.d.*                                                  | 0.3                                                          | 14                                                       |
| S_03         | n.d.*                           | 13.1                                                           | 1.7                                                     | 1.1                                                         | 0.8                                                    | 0.6                                                          | 231                                                      |
| AS_02        | n.d.*                           | n.d.*                                                          | n.d.*                                                   | 1.4                                                         | 1.4                                                    | 0.4                                                          | n.d.*                                                    |
| AS_03        | n.d.*                           | n.d.*                                                          | n.d.*                                                   | 0.2                                                         | n.d.*                                                  | 0.1                                                          | n.d.*                                                    |
| HS_02        | n.d.*                           | n.d.*                                                          | n.d.*                                                   | 0.3                                                         | n.d.*                                                  | 0.2                                                          | n.d.*                                                    |
| HS_03        | n.d.*                           | n.d.*                                                          | n.d.*                                                   | 0.3                                                         | n.d.*                                                  | 0.3                                                          | n.d.*                                                    |
| AHS_02       | n.d.*                           | n.d.*                                                          | n.d.*                                                   | 1.3                                                         | 1.1                                                    | 0.1                                                          | n.d.*                                                    |
| AHS_03       | n.d.*                           | n.d.*                                                          | n.d.*                                                   | 1.2                                                         | 0.9                                                    | 0.1                                                          | n.d.*                                                    |

\*n.d. = not detectable.

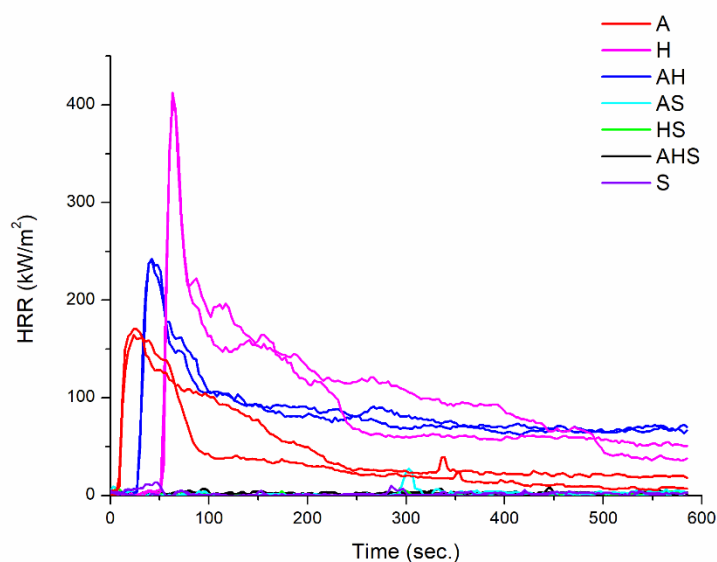


Figure 5.3-9. Heat Release Rate (HRR) of the materials and the composite samples determined by the cone calorimeter test: Test Method B simulating Single Burning Item (reporting the type of the tested materials/composites instead of the samples' designation).

The trends of the curves confirmed the results achieved by performing Test Method A, but the differences between the materials were more evident. Indeed, exposing the samples to a spark

resulted in a quicker ignition, occurring when they reached the minimum flammability range due to the ignitable volatiles produced during thermal degradation. In Test Method B, *A. donax*, A, and the composite, AH, exhibited more distinct values compared to Test Method A. The presence of ignition indeed preceded the flame event, potentially aligning with the previously discussed higher charring ability of hazelnut shells under such conditions, thus delaying the contribution of the H component to flame generation.

Again, for HRR, hazelnut shells achieved the highest peak ( $\text{pHRR} = 412.5 \text{ kW/m}^2$ , at 63 s), followed by the mixture of the two aggregates ( $\text{pHRR} = 242.6 \text{ kW/m}^2$ , at 42 s), and *A. donax* ( $\text{pHRR} = 171.0 \text{ kW/m}^2$ , at 24 s), considering the highest value between the two samples for each composite. In the presence of a spark, all the aggregates reached a peak before the first minute, faster than for Test Method A, due to the more critical conditions that aim at highlighting the very first flammability condition reached. Then, the curve decreased and achieved minimum values of  $\text{HRR} = 35.4 \text{ kW/m}^2$  (at 579 s) for the hazelnut shells,  $\text{HRR} = 61.7 \text{ kW/m}^2$  (at 417 s) for the mixture of the two aggregates and  $\text{HRR} = 5.4 \text{ kW/m}^2$  (at 552 s) for *A. donax*, considering the minimum value between the two samples for each composite. As for the Total Heat Release, THR, reported in Figure 5.3-10, hazelnut shells reached an overall higher amount, followed by the mixture of the two aggregates and the *A. donax*, analogously to Test Method A.

The addition of the sodium silicate, once again lowered the value of the HRR and THR (Figures 5.3-9 and 5.3-10, respectively), in line with the results of Test Method A. Hence, the sodium silicate widely influenced the final performance and improved the fire reaction of the bio-aggregates. For the composites, the curves of HRR (Figure 5.3-9) did not show a peak.

As reported in Table 5.3-5, by Test Method B, it was possible to evaluate  $\text{FIGRA}_{0,2\text{MJ}}$  index, namely the Fire Growth Rate index, which represents the maximum HRR throughout time. Then, it measures the heat contribution to developing fire from specific materials, allowing predicting the tendency for fire growth. Similarly, to the previous case in evaluating the flashover potential,  $\text{FIGRA}_{0,2\text{MJ}}$  showed that the bio-aggregates represented hazardous materials. The addition of sodium silicate widely changed the scenario, with the condition in which FIGRA cannot be evaluated, practically accounting for the negligible risk of fire growth.

Figure 5.3-11 shows the Smoke Production Rate (SPR) of the materials and the composites.

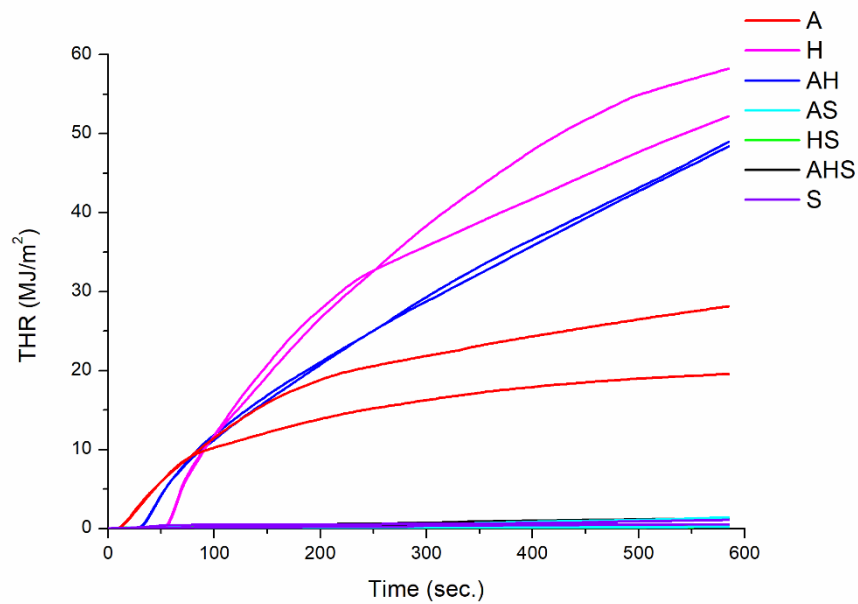


Figure 5.3-10. Total Heat Release (THR) of the materials and the composites determined by the cone calorimeter test: Test Method B simulating Single Burning Item (reporting the type of the tested materials/composites instead of the samples' designation).

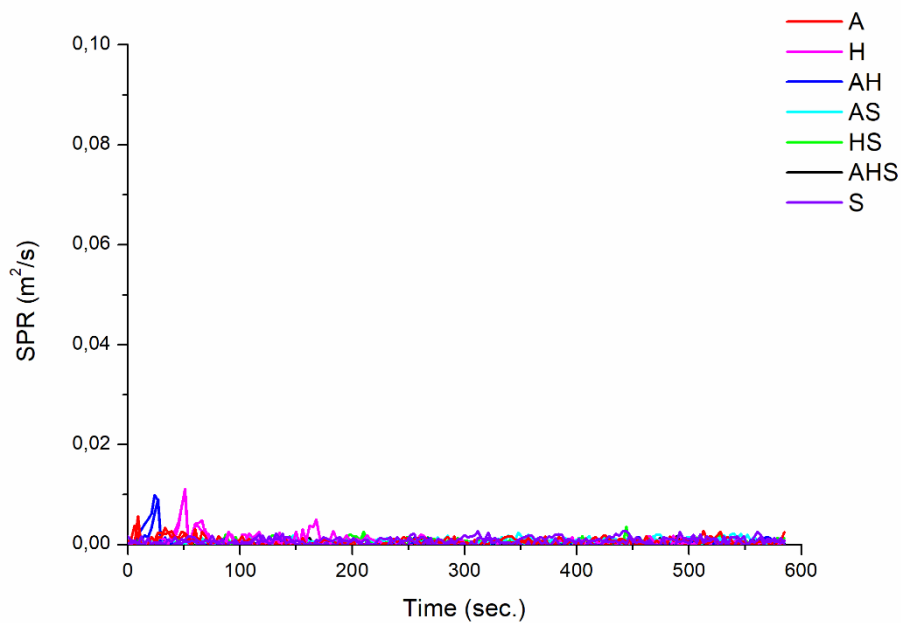


Figure 5.3-11. Smoke Production Rate (SPR) of the materials and the composite samples determined by the cone calorimeter test: Test Method B simulating Single Burning Item (reporting the type of the tested materials/composites instead of the samples' designation).



Considering the aggregates, for Test Method B, the SPR was lower than for Test Method A. By generating a spark, the maximum values of SPR were  $0.011 \text{ m}^2/\text{s}$  for hazelnut shells,  $0.010 \text{ m}^2/\text{s}$  for the mixture and  $0.006 \text{ m}^2/\text{s}$  for *A. donax*. Differently from the previous result, the *A. donax* showed the lowest value of the peak. In any case, in the presence of ignition, the smoke production was significantly lower, possibly owing to the flame production in the presence of a lower concentration of ignitable volatiles. As for the composites, again, the sodium silicate solution avoided smoke production and improved the fire reaction behaviour. The results of TSP are reported in Figure 5.3-12.

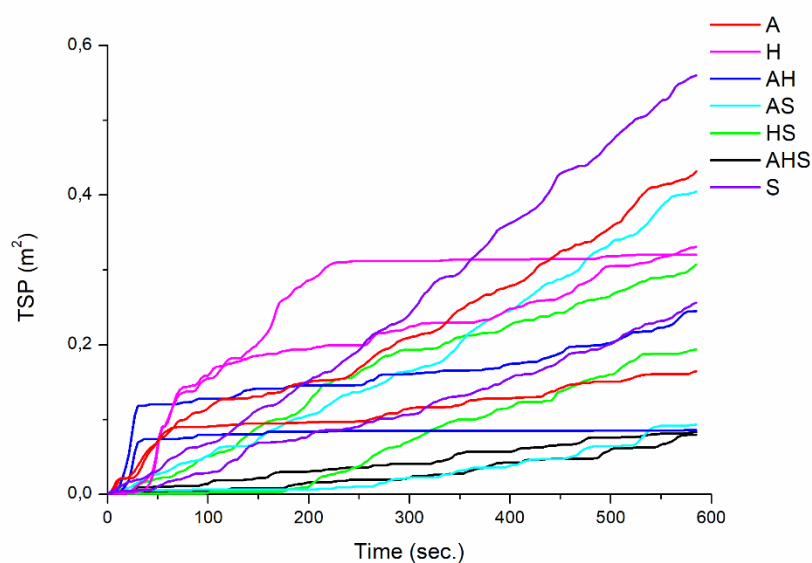


Figure 5.3-12. Total Smoke Production (TSP) of the materials and the composites determined by the cone calorimeter test: Test Method B simulating Single Burning Item (reporting the type of the tested materials/composites instead of the samples' designation).

Figure 5.3-13 shows the samples after the test. The bio-aggregates burnt (Figure 5.3-13a) due to the low resistance to fire typical of bio-based materials [30,31]; the sodium silicate swelled, alone (Figure 5.3-13b) and in the composites (Figure 5.3-13c). This reaction avoided composites' burning and smoke production, hence the sodium silicate created a sort of barrier which protected the bio-aggregates, as better discussed in the following section.

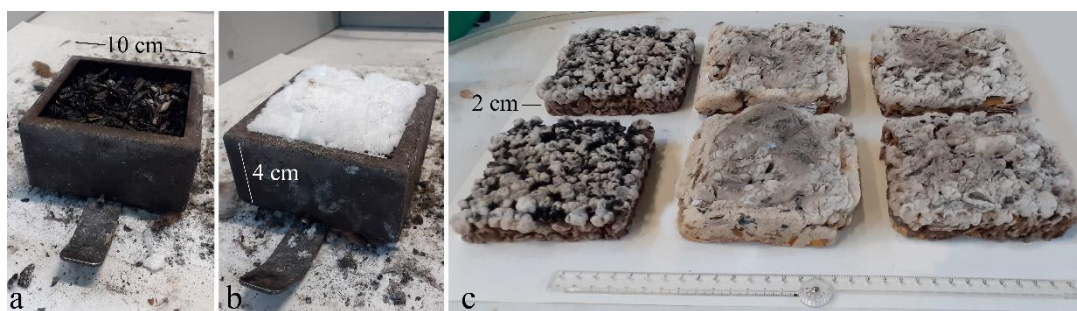


Figure 5.3-13. Samples after the assessment of fire reaction (Test Method B): a) mixture of *A. donax* and hazelnut shells AH; b) dried sodium silicate solution S; c) composite samples: hazelnut shells-based ones HS (left); *A. donax*-based ones AS (centre); mixture-based ones AHS (right).

## 5.3.5 Discussion

### 5.3.5.1 Main findings of the research

The results of the reaction to fire of the bio-based composites were extremely promising and confirmed the sodium silicate's great influence [71]. Indeed it improved the performance in case of fire, lowering the values of the Heat Release Rate (HRR) and the Total Heat Release (THR) and significantly avoiding smoke production, one of the main problems in case of fire [71,72]. The sodium silicate created a silica-rich hybrid layer, which protected the bio-aggregates with an intumescent barrier, as explained by Li et al. [79]. Due to its inorganic characteristics, it shielded the organic bio-aggregates from burning [71].

The results of the fire reaction tests (Tables 5.3-4 and 5.3-5) suggested that the analysed composites could be considered as non-combustible products. Hence, they could be employed as indoor coating boards ensuring the minimization of the hazards due to smoke and toxic gases.

Comparing the two performed tests, some differences were evident. For the HRR of the bio-aggregates, higher values were achieved in Test Method B (Figure 5.3-9), while the HRR curve decreased more evidently in Test Method A (Figure 5.3-5). This was in line with the expectation, as for Test Method A, the samples were exposed to constant heat during the test, without any other source of ignition, differently from Test Method B, where a spark generation (igniter) was introduced. For this latter, the peak of the curve was achieved faster, as also represented by THR trends (Figures 5.3-6 and 5.3-10). These two curves also showed some differences between the composites: in Test Method B their values were almost zero throughout the test, while in Test Method A they were a bit higher. As for the smoke production for bio-aggregates (as no significant smoke was produced by the composites bonded by sodium silicate), in Test Method B the Total Smoke Production (TSP) was lower than in Test Method A,

coherently with the results of THR, as the two properties are related [30,60]. This result was caused by the different heat applications: more uniform heat radiation burnt more quantities of materials, thereby generating more smoke and heat [30].

The considerations on the two performed tests demonstrated as Test Method A and Test Method B, simulating two different conditions, produced different heat and smoke responses of the building materials. Thus, to assess the reaction to fire of building materials, both were significant.

The comparison with past studies that assessed the fire reaction of bio-aggregates, and the sodium silicate validated the achieved results.

Rocha et al. [60] considered rice husk, bamboo and wood shaving and carried out the same tests performed in the present study for the materials: TGA and reaction to fire by using a cone calorimeter and applying, however, a higher constant heat flux of 50 kW/m<sup>2</sup>, representing an already well-developed fire. For TGA the researchers achieved similar results, highlighting a preliminary water loss and then a sequence of weight drops accounting for cellulosic base products' decomposition. A final higher weight loss was possibly referred to the decomposition of the aromatic lignin component which is the most stable fraction and possibly also responsible for the charring ability of the bio-based products. As for the fire reaction, the bio-aggregates showed similar HRR curves to the ones of the present study, namely an initial delay period, then a peak and a decrease. The achieved values of HRR were lower than the ones of the present study, especially for the hazelnut shells. This was caused potentially by a different material distribution that might alter the fire reaction, as well as by different heat flux applied. Moreover, while the actual values are not precisely stated by Rocha et al. [60], it seemed that the analysed materials displayed a higher water content than the ones of the present study. Thus, they produced not only a lower mass to burn but also a higher water fraction that helped smooth the fire evolution itself. However, the fire reaction behaviour (trend of HRR curves) was positively comparable.

Li et al. [71] assessed the reaction to fire of wood modified with a sodium silicate solution and provided the HRR, THR, SPR, and TSP. The results demonstrated that, by impregnating the wood with the sodium silicate, all the above cited properties were improved, as the values of all the curves lowered. This confirmed the promising performance in case of fire of the sodium silicate, which acted as a protection barrier for bio-based materials [79], in agreement with the present results.

While it is a difficult comparison to be carried out, since the manufacturing of the different products can be widely variable, it seems that a key point in boosting the fire resistance of

the boards is a complete covering of the biomass with the intumescent sodium silicate binder. This feature, achieved in the present work, cannot be controlled, or compared with other literature data, but it should be a key point in developing a potential scale up of such particleboards.

#### 5.3.5.2 Fire classification

The results of the present study indicated the composites' fire classification. Indeed, building materials are divided into six classes, from class A to F, plus related subclasses, based on their reaction to fire. Class A1, for example, includes those materials that do not contribute in any phase of the fire, including the fully developed fire phase, and can automatically fulfil all the requirements of the lower classes. Class F, the worst, labels all products for which their reaction to fire cannot be determined or cannot be classified in one of the previous classes. EN ISO 1182 [74], EN ISO 1716 [80] and EN 13823 [75] are useful standards to refer to for obtaining the A classification, according to ISO 13501 [73], and provide the use of different instrumentation and preparation of samples with a certain geometry.

Several different approaches were issued over the years to set fire classification. The cone calorimeter was initially discarded owing to the small sample size that can accommodate. However, in the present case, the cone calorimeter test carried out on particleboards was used as a means of comparison with a more complex (and costly) technique that would imply the preparation of significantly bigger samples (such as a single burning item). Law et al. [81] analysed the evolution of such standards and reported that to comply with the A1 class – the non-combustible item class – in the Single Burning Item (SBI) condition, samples should show an Effective Heat of Combustion (EHC) below 2.0 MJ/kg (according to the German requirements [81]) and no flame whatsoever.

All the analysed bio-based composites were compliant with this requirement, and they also well complied with the A2 class requirement. This aspect, together with practically null Fire Growth Rate (FIGRA) and flashover index accounted all to the potential for the discussed formulations to undergo certification in the A2, as the minimum, but possibly also A1. It was not clear what should be the applied heat flux. In their discussion, Law et al. [81] reported that a Heat Flow of 20-40 kW/m<sup>2</sup> was acceptable for defining the SBI, with some hint at choosing 40kW/m<sup>2</sup>. While the present data refer to a heat flow of 35 kW/m<sup>2</sup>, corresponding at nearly 650°C, it seemed an acceptable situation to work for assigning class A parameters, being them A1 or A2. Consequently, the data on flame behaviour presented herein cannot be effectively utilized to ascertain the material's adherence to the standard. However, an inference may be

drawn from the observed performance that, if tests were performed following the stipulated standard parameters, compliance with the specified constraints would likely be achieved.

### 5.3.5.3 Additional considerations and future studies

The analysed bio-based composites presented several benefits. First, being made of agro-industrial wastes and being bio-resources, they could be a suitable solution to reuse these wastes. Both the bio-aggregates were widely produced and available all over the year, thus overcoming the problem of seasonality. Furthermore, the materials employed in this study were locally produced, hence the energy requirement for both the production and the transport was minimized, boosting their sustainability.

Even if the selected adhesive was not bio-based, the sodium silicate has many advantages, as described in Section 5.3.1, as also confirmed further in the present study. Indeed, the performed laboratory tests showed that the sodium silicate solution eliminated one of the main drawbacks of bio-based products, namely the low fire resistance. This is an extremely promising result for this research field. Furthermore, the achieved results demonstrated that the proposed boards, which could be used as indoor coatings improving indoor comfort [18,58], possibly meet the A1 fire class.

On the other hand, according to past studies [18,58,66], the proposed boards showed some weaknesses that should be moderated or solved. For example, the mechanical performance appeared low, both for compressive and flexural strength. Cintura et al. [18,58] reported values of compressive strength of about  $\sigma = 2.1 \text{ N/mm}^2$  and flexural strength of about  $\sigma = 0.39 \text{ N/mm}^2$  for hazelnut shell-based composite. The *A. donax*-based boards did not achieve the breaking load, and they seemed more similar to straw bales, with value of compressive strength of about  $\sigma = 0.9 \text{ N/mm}^2$  and flexural strength of about  $\sigma = 0.35 \text{ N/mm}^2$ . Hence, both the hazelnut shell-based and *A. donax*-based boards seemed inadequate as structural boards or masonry units [18], although the results were in line with past research [58].

The high hygroscopicity of the composites could be both a benefit and a drawback [66]. Indeed, despite it could be useful to control indoor conditions, it could also cause the degradation of the materials [62]. Some strategies could improve low moisture resistance, such as the addition of certain additives (i.e., phosphorus, boron, or silica fume [82]). However, this possibility should be demonstrated and better detailed to define if the benefits might be more than the drawbacks.

Finally, a great weakness could be the low resistance to biological colonization of the bio-aggregates [64]. Nevertheless, according to past research [49,50] the sodium silicate

solution could also avoid chemical decomposition and biological colonisation, but these properties should be further investigated.

Thus, the present study demonstrated that the use of sodium silicate as a binder is an efficient solution to produce boards made of eco-friendly materials, safe for human health from different points of view. Finally, the combination of these materials results in a building composite that could improve indoor conditions.

To provide further support to these considerations a qualitative cost-effectiveness analysis of the proposed bio-based boards was considered. As the employed bio-aggregates were by-products, their cost was derived only from the transport of the materials. Hence, the final cost of the boards will be mainly caused by the sodium silicate and the manufacturing process. Although sodium silicate production required some costs, it could be produced by innovative processes, which made it cheaper [83,84]. Furthermore, as previously reported, the quantities of the adhesive were minimized (only 30%), and the production process avoided high temperatures, pressing and the use of specific treatments. Hence, a low production cost was expected. However, a more detailed analysis and a life cycle assessment (LCA) would better support these considerations, as well as quantify the sustainability of the proposed boards.

### 5.3.6 Conclusions

This study considered bio-based boards for indoor applications. The employed bio-aggregates were giant reed (*A. donax* L.) and hazelnut shells. A sodium silicate solution was employed as the adhesive. The thermal stability, water content, and the inorganic fraction composition of the raw materials and the composites and their reaction to fire were evaluated.

The following conclusions were achieved:

- The particleboards' composition strongly resented the quality of the employed agro-wastes and bio-resources under fire. Hazelnut shells determined significantly lower inorganic waste residue than *A. donax*.
- The two performed test methods were useful for carrying out considerations about the fire reaction of materials and composites. By a cone calorimeter method, it was possible to apply constant heat, with and without generating a spark (igniter), and to have different results for heat release and smoke production of the samples. These results allowed making different considerations, crucial to assess the possible hazards of the proposed composites.
- Considering the standard requirements for building products and the results of the fire reaction test, the considered bio-based composites could be classified as class A2 and potentially A1.

- The sodium silicate widely improved the fire reaction of the bio-aggregates, avoiding their burning and smoke production. Hence, the considered bio-based boards show a good performance in case of fire and could be employed as indoor panels without risks to humans.

An effective solution to improve the bio-based boards' behaviour in case of fire was, hence, described and validated. Future studies could further analyse boards' properties by changing some parameters (e.g., the grain size of the bio-aggregates, the percentages of the sodium silicate solution, and the additions of some additives) and investigate the possibility of using a sodium silicate solution as a superficial treatment in bio-based building products. Furthermore, the performance in case of fire of the considered boards could be further detailed by considering more replications, different conditions, and hence different test methods.

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### **Data availability**

Data will be made available on request.

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## 5.4 Acoustic and mechanical properties

### 5.4.1 Context

The mechanical and acoustic characterization of the composites was carried out, further detailing the analysis described in Chapter 4. The two bio-aggregates were considered both separately and mixed to produce the composite samples, as described in Sections 5.1, 5.2, and 5.3. The aim was to evaluate, first, the properties of the one-aggregate-based boards, and then the optimization ones, namely the mixture-based composite. This would ensure a better understanding of the variation, and, possibly, the improvement, in the composites' performances.

As reported in Section 5.1, new samples were produced; the hazelnut shells were provided by Raccolti di Cin, Baldissero d'Alba, Cuneo, Italy, and the *A. donax* was harvested in Ozzano dell'Emilia, Bologna, Italy. Both the aggregates were shredded and dried at  $T = 60\text{ }^{\circ}\text{C}$  until constant mass. The same sodium silicate solution,  $\text{Na}_2\text{O}\cdot n(\text{SiO}_2)$ , provided by Ingessil Srl, Montorio, Verona, Italy was employed as the adhesive.

The performed tests were the ones already considered in the previous experimental campaigns, described in Chapter 4. This allowed an easier comparison of the results and understanding of the possible variations in the composite samples' behaviour. As for the mechanical properties, only compressive strength was evaluated, but further analysis should consider the flexural strength as well. The test methods and the results of acoustic and mechanical properties are described in detail in the following sections.

### 5.4.2 Samples production, and designation

The composite board samples were produced by the already employed production process, described in detail in Section 4.2. Both cylindrical (diameter = 10 cm, thickness = 4 cm) and rectangular (10 cm x 10 cm x 4 cm) samples were produced. The cylindrical ones were employed to evaluate the acoustic absorption behaviour, while the rectangular ones were for the compressive strength. The 10 cm diameter was selected due to the employed equipment to evaluate acoustic absorption capacity, namely the already described impedance tube (Section 4.3.2.3.4 and 4.4.2.3.4). The 4 cm thickness was selected as it could be suitable for both thermal and acoustic boards. Although the thermal insulation properties were no longer considered due to the results of the previous experimental campaign, the same thickness was considered for the composite samples. Furthermore, it ensured samples' production and test



implementation: thinner dimensions might determine problems of samples' production; thicker ones could result in samples too weighty, as reported in Section 4.4.2.1.

Table 5.4-1 summarizes all the information about the composite samples employed for the acoustic and the mechanical analysis: the composites' designation, the mix design, the samples' size and designation, and apparent density, including average and standard deviation, determined as previously described (Sections 4.3.2.3.2, and 4.4.2.3.2). For the designation, the first letter of the material was considered for the composites based on one bio-aggregate or the mixture of two: **A** = *A. donax*; **H** = Hazelnut shells; **AH** = *A. donax* + Hazelnut shells. To distinguish the two types of *A. donax*-based composites, the one produced by *A. donax* collected from a cultivation in Cadriano was designated as A1, the other one as A2. This latter was used to produce both *A. donax*-based samples and the ones made of the mixture of the two aggregates. To distinguish the hazelnut shell-based composite, the already tested and described in the previous section were reported as H, and the new ones were reported as H2.

As composite samples A1 and H were the same analysed in the reference past studies (Section 4.3 and 4.4) the same numeration was considered. For the other samples (A2, and AH) the numeration did not start from 1 because they were produced together with other samples, (used to assess the performance in case of fire, described in Section 5.3) and numbered considering these. The values of apparent density showed that, despite minor variations, the uniformity among the samples was guaranteed, as shown by the low values of standard deviation. Particularly, the hazelnut shell-based and the mixed-based composites exhibited notably enhanced homogeneity. *A. donax*-based composites (for both types of *A. donax*) had the lowest values of apparent density.

## 5.4.3 Methods

### 5.4.3.1 Acoustic absorption

To assess the acoustic absorption the laboratory test already described in Sections 4.3.2.3.4 and 4.4.2.3.4 were performed. The three samples for each composite, A1, A2, H, and AH, were stabilized at  $T = (20 \pm 1) ^\circ\text{C}$  and  $\text{RH} = (60 \pm 5) \%$ , smoothed, placed at the end of an impedance tube, and sealed with plasticine to reduce the possible gaps between them and the tube. A loudspeaker, by a signal amplifier (Samson 120A with SNR = 96 dB), generated an exponential sine sweep (Corredor-Bedoya et al., 2021), converted to an analog signal by the Digital to Analog Converters of the soundcard (RME 802). The signal was recorded by a single microphone 1/2" PCB and converted by the Analog-to-Digital Converter (sample rate of 44.1 kHz at 24-bit

depth), and processed by ITA-Toolbox (Berzborn et al., 2017) through the Transfer-Function Method (Chung and Blaser, 1980a, 1980b). The test was performed for each sample's sides (designated as Flatter - base of the mould - and Rougher - its top).

Table 5.4-1. Information about the samples employed to test acoustic and mechanical properties: type of composite, size, number, and designation of the samples, apparent density, and standard deviation.

| Composite                                                                                                         | Size                               | Samples        | Apparent density [kg/m <sup>3</sup> ] |
|-------------------------------------------------------------------------------------------------------------------|------------------------------------|----------------|---------------------------------------|
| A1 - <i>A. donax</i> -based composite produced by <i>A. donax</i> collected in Cadriano, Bologna, Italy           | Diameter = 10 cm, thickness = 4 cm | A1_07          | 585                                   |
|                                                                                                                   |                                    | A1_08          | 515                                   |
|                                                                                                                   |                                    | A1_09          | 490                                   |
|                                                                                                                   |                                    | Average ± S.D. | 530 ± 49                              |
| A2 - <i>A. donax</i> -based composite produced by <i>A. donax</i> collected in Ozzano dell'Emilia, Bologna, Italy | 10 cm x 10 cm, thickness = 4 cm    | A2_04          | 447                                   |
|                                                                                                                   |                                    | A2_05          | 473                                   |
|                                                                                                                   |                                    | A2_06          | 462                                   |
|                                                                                                                   |                                    | A2_07          | 495                                   |
|                                                                                                                   |                                    | A2_08          | 567                                   |
|                                                                                                                   |                                    | Average ± S.D. | 489 ± 47                              |
|                                                                                                                   | Diameter = 10 cm, thickness = 4 cm | A2_09          | 580                                   |
|                                                                                                                   |                                    | A2_10          | 490                                   |
|                                                                                                                   |                                    | A2_11          | 427                                   |
|                                                                                                                   |                                    | Average ± S.D. | 499 ± 77                              |
| H - Hazelnut shell-based composite already tested in the previous experimental campaign                           | Diameter = 10 cm, thickness = 4 cm | H_04           | 656                                   |
|                                                                                                                   |                                    | H_05           | 680                                   |
|                                                                                                                   |                                    | H_06           | 686                                   |
|                                                                                                                   |                                    | Average ± S.D. | 674 ± 16                              |
| H2 - Hazelnut shell-based composite produced for this experimental campaign                                       | 10 cm x 10 cm, thickness = 4 cm    | H2_01          | 712                                   |
|                                                                                                                   |                                    | H2_02          | 823                                   |
|                                                                                                                   |                                    | H2_03          | 768                                   |
|                                                                                                                   |                                    | H2_04          | 708                                   |
|                                                                                                                   |                                    | H2_05          | 688                                   |
|                                                                                                                   |                                    | Average ± S.D. | 740 ± 55                              |
| AH - Mixture-based composite made of <i>A. donax</i> from Cadriano, Bologna, Italy, and hazelnut shells           | 10 cm x 10 cm, thickness = 4 cm    | AH_04          | 671                                   |
|                                                                                                                   |                                    | AH_05          | 684                                   |
|                                                                                                                   |                                    | AH_06          | 715                                   |
|                                                                                                                   |                                    | AH_07          | 643                                   |
|                                                                                                                   |                                    | AH_08          | 621                                   |
|                                                                                                                   |                                    | Average ± S.D. | 667 ± 36                              |
|                                                                                                                   | Diameter = 10 cm, thickness = 4 cm | AH_09          | 719                                   |
|                                                                                                                   |                                    | AH_10          | 756                                   |
|                                                                                                                   |                                    | AH_11          | 745                                   |
|                                                                                                                   |                                    | Average ± S.D. | 740 ± 19                              |

\*Percentages by volume.

#### 5.4.3.2 Internal structure and porosity

The internal structure and the macroporosity of the samples were assessed and described in Section 5.2.3.2 by performing an XR computed tomography by an in-house assembled apparatus. It consisted of a Bosello XRG120.IT conventional X-ray tube, a Varian PaxScan 2520D flat-panel X-ray detector, and a Physik Instrument manipulation axis set with an M-038 micrometric sample rotation stage and M413.3PD for detector translation. The three cylindrical samples of each composite, separated by expanded polystyrene boards (transparent to X-rays), were placed vertically on a rotation platform. The scanning parameters and the detailed description of the test are reported in Section 5.2.3.2.

#### 5.4.3.3 Compressive behaviour

Compressive behaviour was determined by adapting EN 826 (2013), as described in Sections 4.3.2.3.6 and 4.4.2.3.7. The rectangular samples were placed between the two plates in the Galdabini machine (maximum capacity of 100 kN), with a joint of 11.65 kg, and compressed in displacement control at the velocity of 2 mm/min. The force and displacement were recorded during the test by a linear displacement transducer, LVDT, HBM Model WA20mm. The test was carried out until the breaking load or until the maximum capacity of the LVDT, namely up to its maximum compression.

The stress-strain curve was obtained having  $\sigma$  [N/mm<sup>2</sup>], obtained by the quotient of  $F$  [N] and the initial cross-sectional area of the samples  $A_0$  [mm<sup>2</sup>] in ordinate and  $\epsilon$  [-] obtained by Equation 5.4-1 in abscissae, with  $\delta$  [mm] the displacement during the test recorded by the LVDT (HBM Model WA20mm), and  $d_0$  [mm] the initial thickness of the samples.

$$\epsilon [-] = \frac{\delta}{d_0} \quad (5.4-1)$$

Compressive strength  $\sigma_{\max}$  [N/mm<sup>2</sup>] was calculated for each sample by considering the compressive stress corresponding to the breaking load or 10 % strain (thus,  $\epsilon = 0.1$  if the samples did not achieve rupture), as defined by EN 826 (2013). The average value and standard deviation were considered.

The modulus of elasticity was determined by Equation 5.4-2 for each sample, considering 20% and 60% of  $\sigma_{\max}$  ( $\sigma_{60\%}$  and  $\sigma_{20\%}$ ) and the corresponding  $\epsilon$  ( $\epsilon_{60\%}$  and  $\epsilon_{20\%}$ ) (UNI-11842, 2021); then the average value and standard deviation were considered.

$$E \left[ \frac{N}{mm^2} \right] = \frac{\Delta\sigma}{\Delta\epsilon} = \frac{\sigma_{60\%} - \sigma_{20\%}}{\epsilon_{60\%} - \epsilon_{20\%}} \quad (5.4-2)$$

## 5.4.4 Results and discussion

Some of the results already reported in Sections 4.3.3, and 4.4.3 were represented here for easier comparison and discussion.

### 5.4.4.1 Acoustic absorption

Figure 5.4-1 (adapted from Section 4.4.3.4) and Figure 5.4-2 show the results of the acoustic absorption coefficient evaluated for both sides of the samples made of *A. donax* and sodium silicate (A1 and A2). The dotted lines represent the results for the rougher ones; in contrast, the continuous lines represent the results for the flatter sides.

In Figure 5.4-1, the acoustic absorption behaviour was similar for all samples. Two samples absorbed higher frequencies, peaking at almost 1100 Hz. Discrepancies in the values of sound absorption coefficient might be caused by the differences in aggregates' disposition, which affected the internal voids' distribution (de Carvalho et al., 2020). In the range between 600-700 Hz, the sound absorption coefficient  $\alpha$  was almost 1. The curve showed a peak, followed by a slight decrease; then it increased again. The *A. donax*-based composite showed an acoustic absorption behaviour similar to granular material (Horoshenkov and Swift, 2001), a particular case of porous materials. After the first peak, the composite behaviour was closer to the typical porous materials, with a slight increase in the sound absorption curve. These results were discussed in more detail in Section 4.4.3.4.

Regarding Figure 5.4-2, the samples did not show results as homogeneous as for the first type of *A. donax* (Figure 5.4-1). Furthermore, especially for the samples with the exposed surface as the flatter one (continuous lines), the curves achieved lower values of the sound absorption coefficient.

The sample A2\_10\_Flatter did not show a single marked peak, but some small peaks, which were determined by the acoustic measurements and the equipment. The sound absorption coefficient did not exceed  $\alpha = 0.25$ . For this, the results were very different from the others, probably due to a different sodium silicate distribution (more sodium silicate, less surface porosity). In the case of A2\_09\_Flatter and A2\_11\_Flatter, however, higher values of sound absorption coefficient were achieved. The sound absorption curve for A2\_09\_Flatter increased at frequencies of 600-700 Hz, then decreased. As for the A2\_11\_Flatter, the curve showed a marked peak  $\alpha \approx 0.75$  at 1000 Hz, a decrease, and then another peak at 1700 Hz.

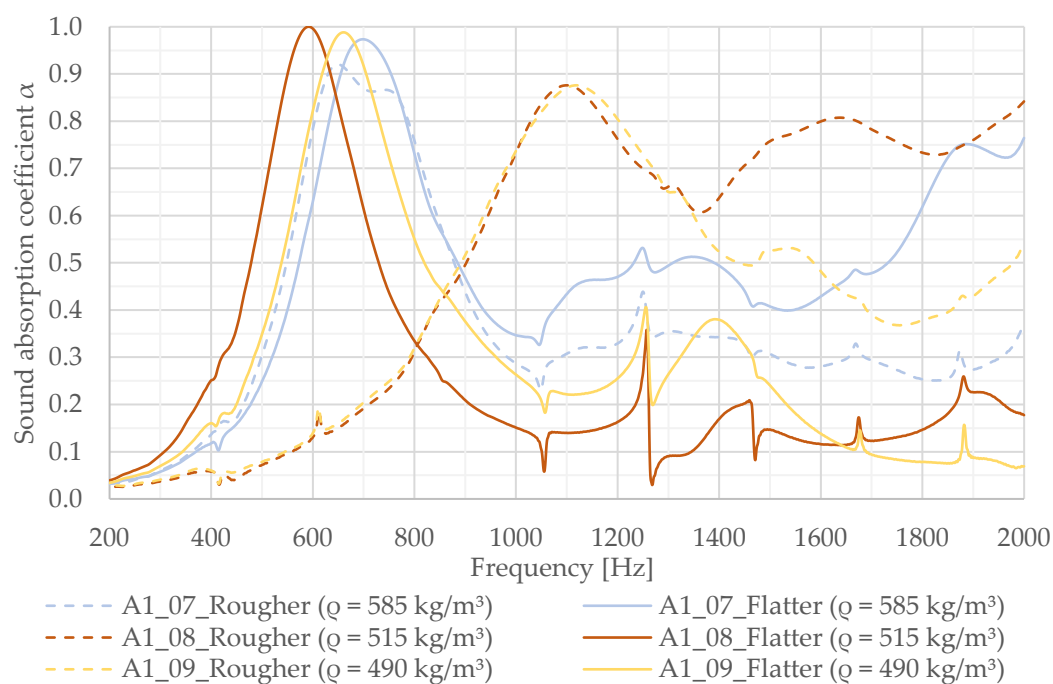


Figure 5.4-1. Sound absorption coefficient of both sides of the three tested composite samples made of *A. donax* and sodium silicate solution (A1). Continuous lines represent the flatter exposed surfaces' results and the rougher ones' dotted lines.

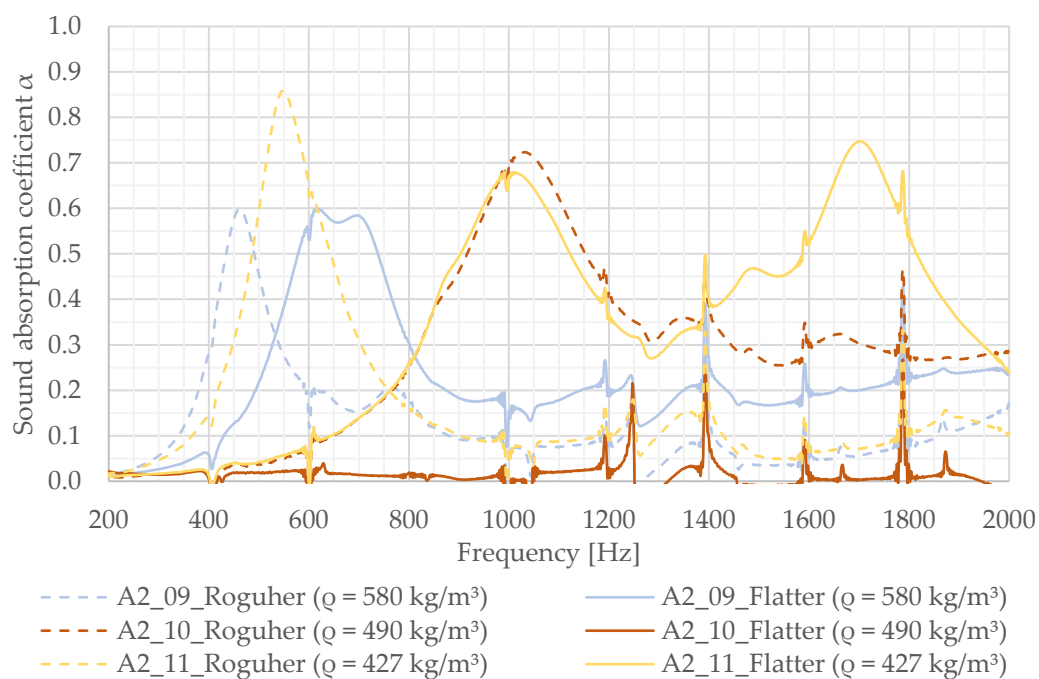


Figure 5.4-2. Sound absorption coefficient of both sides of the three tested composite samples made of *A. donax* and sodium silicate solution (A2). Continuous lines represent the flatter exposed surfaces' results and the rougher ones' dotted lines.

When the exposure surface was the rougher one (dotted lines), the curves' trends were more similar to the ones represented in Figure 5.4-1: an initial marked peak, a decrease, and then another increase. In this case, the three peaks were reached at different frequencies (between 400 – 500 Hz, between 500 – 600 Hz, and between 1000 – 1100 Hz). Furthermore, the achieved values of the sound absorption coefficient,  $\alpha$ , were lower than in Figure 5.4-1. However, the frequency ranges were always in the range of human voice (EN IEC 60268-16, 2020; ISO 3382-3, 2012).

Although the values of the sound absorption coefficient were different, the trend of the curves tended to be similar. Further analysis is required to better explain the samples' behaviours. It seemed to be strictly dependent on the production process and it was widely influenced by the sodium silicate distribution, which was probably different in *A. donax*-based composite A2. This demonstrated that the sound absorption behaviour mainly depended on the composition of aggregates-adhesive, instead of the type of aggregates.

Figure 5.4-3 (adapted from Section 4.3.3.4) shows the results of the acoustic absorption coefficient evaluated for both sides of the samples made of hazelnut shells and sodium silicate (H). Again, the dotted lines represent the results for the rougher ones; in contrast, the continuous lines represent the results for the flatter sides.

The curves of the three replications were similar. Again, the differences among the curves could be caused by the random disposition of the bio-aggregates (de Carvalho et al., 2020), hence by the different internal voids. Again, the acoustic absorption behaviour suggests that the samples could be considered granular material, a special case of porous materials. This was determined by the type of employed aggregate, namely high-mass aggregate, mixed with the selected adhesive (the sodium silicate solution). Indeed, comparing Figure 5.4-3 with Figures 5.4-1 and 5.4-2, the second increase of the curve was less marked. Thus, the composite's behaviour was less similar to porous materials. These results were discussed in detail in Section 4.3.3.4.

Figure 5.4-4 shows the results of the acoustic absorption coefficient evaluated for both sides of the samples made of both *A. donax* and hazelnut shells, with sodium silicate (AH). Again, the dotted lines represent the results for the rougher ones, while the continuous lines represent the results for the flatter sides.

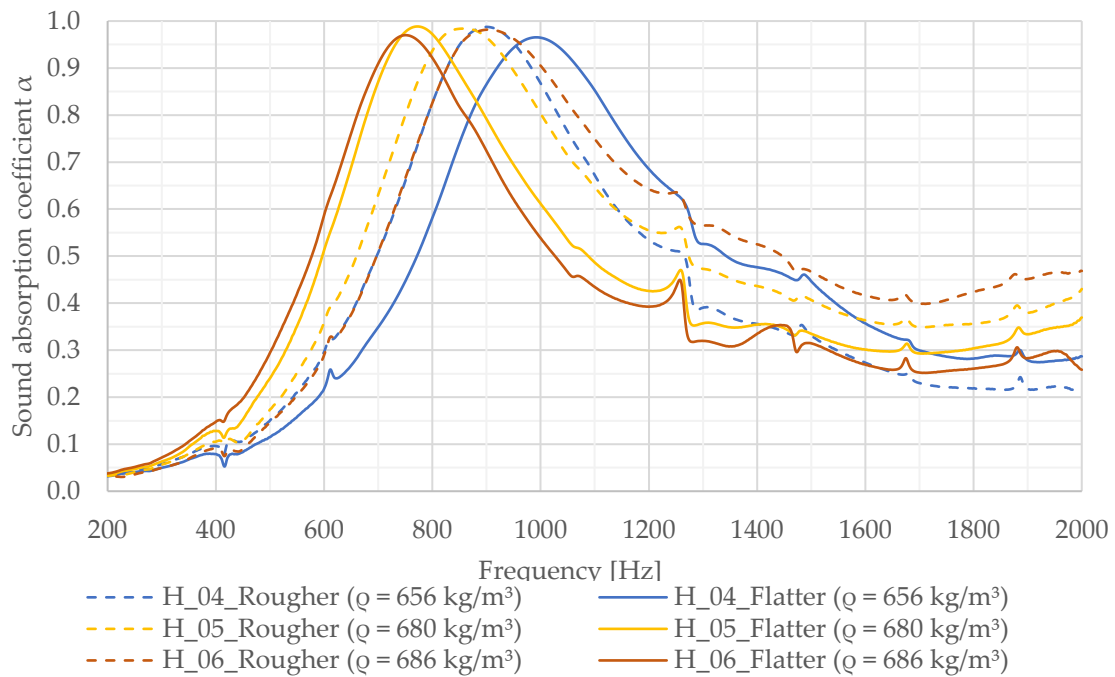


Figure 5.4-3. Sound absorption coefficient of both sides of the three tested composite samples made of hazelnut shells and sodium silicate solution (H). Continuous lines represent the flatter exposed surfaces' results and the rougher ones' dotted lines.

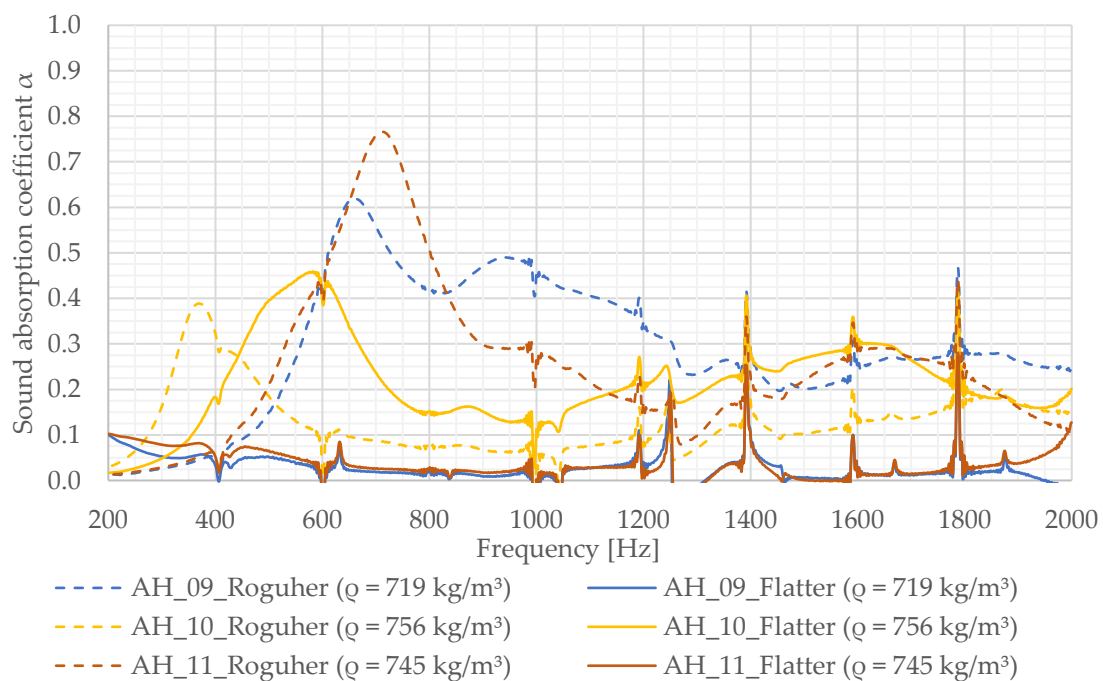


Figure 5.4-4. Sound absorption coefficient of both sides of the three tested composite samples made of *A. donax* and hazelnut shells with sodium silicate solution (AH). Continuous lines represent the flatter exposed surfaces' results and the rougher ones' dotted lines.

The composite samples made of the mixture of the two aggregates did not show a great uniformity in the sound absorption curve, as well as the *A. donax*-based ones, considering A2 (Figure 5.4-2).

Two of them, AH\_09\_Flatte and AH\_11\_Flatte did not show the marked peak, expected according to previous results (Figures 5.4-1 and 5.4-3). For these, the sound absorption curve showed the small peaks determined by the measurement and the equipment. Probably, for these samples, the sodium silicate solution produced a surface closer to what is known as closed pores (Mati-Baouche et al., 2016), avoiding the sound wave from entering the porous structure of the samples.

The other samples' curves (AH\_10\_Flatte, AH\_09\_Rougher, AH\_10\_Rougher, and AH\_11\_Rougher) showed a peak in the frequency range between 350 Hz and 750 Hz. The values of the sound absorption coefficient were lower than the ones of *A. donax*-based and hazelnut shells-based composites (with only one bio-aggregate). The achieved maximum values of  $\alpha$  were almost 0.75. However, the trend of the curve was the same as the previous one (a peak, a decrease, and another increase).

The mixture of the two aggregates resulted in lower values of sound absorption behaviour, but this was probably determined by the production process, rather than by the combination of the two aggregates. Again, the results suggested that the sound absorption capacity was widely affected by the sodium silicate's distribution on the samples' surface.

Although further studies are needed, these achieved results were promising, as the samples showed an uncommon sound absorption behaviour: a curve with a marked peak, instead of an increasing curve, typical of the bio-based materials, namely of the porous absorbers. It could be interesting and useful to optimise this acoustic absorption behaviour, increasing the amplitude of the peak. This would guarantee great sound absorption capacity for a larger frequency range, increasing the possible suitable employment of these bio-based boards.

#### **5.4.4.2 Internal structure and porosity**

To make further considerations on the acoustic absorption capacity, the results of the XR computed tomography, reported in Section 5.2.4.2 were considered. Figure 5.2-5 reports the same sections for each sample: the middle one, namely the frame number  $\frac{1}{2}$  of the total number of frames, and the quarter ones, namely  $\frac{1}{4}$  and  $\frac{3}{4}$  of the total number of frames. It ensured a visual evaluation of the samples' homogeneity and the internal configuration of solids and voids, showing voids' shape and size, connections, and materials distribution.



The differences in internal structures among the composites were more marked across their thickness. The hazelnut shells-based composite showed the greatest homogeneity. This was in line with the expectation, as the production process was optimized for this composite (Section 4.2). Furthermore, the great homogeneity of the hazelnut shells-based composite samples was shown by the acoustic absorption curve (Figure 5.4-3). In the *A. donax*-based composites with and without the hazelnut shells (Figures 5.2-5a and 5.2-5c), the sodium silicate appeared on the bottom side (i.e., the one in contact with the mould during the production process), more markedly than in hazelnut shells-based one. This difference among the composites was expected and was consistent with the standard deviation values of the apparent density (Table 5.2-4) and the curve of absorption coefficient (Figures 5.4-1, 5.4-2, and 5.4-4).

Figure 5.2-5 pointed out also the differences between the shape of voids and aggregates. The distribution of the sodium silicate solution significantly influenced the type of porosity. As Mati-Baouche et al. (2016) described, the combination of adhesive-aggregates produced solid, closed pores, open pores, dead-end pores, dead-end clusters, hence kinematic porosity and dead-end porosity, properties predominantly considered in the acoustic field. Table 5.4-2 reports the results of the porosity of each composite, the average values, and the standard deviation.

Table 5.4-2. Porosity of each sample, and average values with the standard deviation.

| Composite samples                        | Voids [%]* | Solids [%]* |
|------------------------------------------|------------|-------------|
| <b><i>A. donax</i></b>                   |            |             |
| A1_07                                    | 26         | 74          |
| A1_08                                    | 35         | 65          |
| A1_09                                    | 33         | 67          |
| Average Value $\pm$ S.D.                 | 31 $\pm$ 5 | 69 $\pm$ 5  |
| <b>Hazelnut shells</b>                   |            |             |
| H_04                                     | 35         | 65          |
| H_05                                     | 38         | 62          |
| H_06                                     | 34         | 66          |
| Average Value $\pm$ S.D.                 | 36 $\pm$ 2 | 64 $\pm$ 2  |
| <b><i>A. donax</i> + Hazelnut shells</b> |            |             |
| AH_09                                    | 21         | 79          |
| AH_10                                    | 27         | 73          |
| AH_11                                    | 29         | 71          |
| Average Value $\pm$ S.D.                 | 26 $\pm$ 4 | 74 $\pm$ 4  |

\*Note: Percentages by total volume.

As reported in Section 5.2, the composites showed similar percentages of solids/voids, with a variation of only 5%. The porosity seemed to be determined by the mixture of bio-aggregates-adhesive rather than by the type of bio-aggregate. The type of aggregate mainly influenced the shape and size of voids, their connection, and the tortuosity. The standard deviation related to the average value was low (maximum of 5%). Hence, the reproducibility was good: similar results were achieved for the three replicates.

The quantification of the percentages of solids/voids wanted to be a starting point for the analysis of further characteristics of the internal structure of the samples. Certainly, in addition to the number of voids, their shapes, their connection, and the tortuosity, influenced the final performance. Knowing these parameters would be useful to better understand and explain the acoustic absorption behaviour. Indeed, the behaviour of the acoustic curve, resulting from acoustic wave's striking the sample, could be investigated by specific software, namely MATLAB. A predictive model of the samples' acoustic behaviour, not available in the literature, could be developed.

#### 5.4.4.3 Mechanical properties

Figures 5.4-5, 5.4-6, and 5.4-7 show the stress-strain curve for the compressive test for each composite, reporting the results for all the tested samples.

As shown by Figures 5.4-5, 5.4-6, and 5.4-7, the three composites had different behaviours. Comparing the samples of the same composite, the curves were similar.

All the samples of *A. donax*-based composite (Figure 5.4-5) did not achieve the yield. They showed an increasing stiffness increasing the stress. They had a compressive behaviour similar to straw bales (Maraldi et al., 2017, 2016; Molari et al., 2017). The elongated and flattened shape of the shredded *A. donax* improved the cohesion between the particles and the sodium silicate solution, influencing the final performance. The test was stopped at the maximum capacity of the employed LDVT (namely at  $\epsilon \approx 0.5$ ). According to EN 826 (2013), compressive strength and the corresponding force were determined by considering 10% strain ( $\epsilon = 0.1$ ), as reported below.

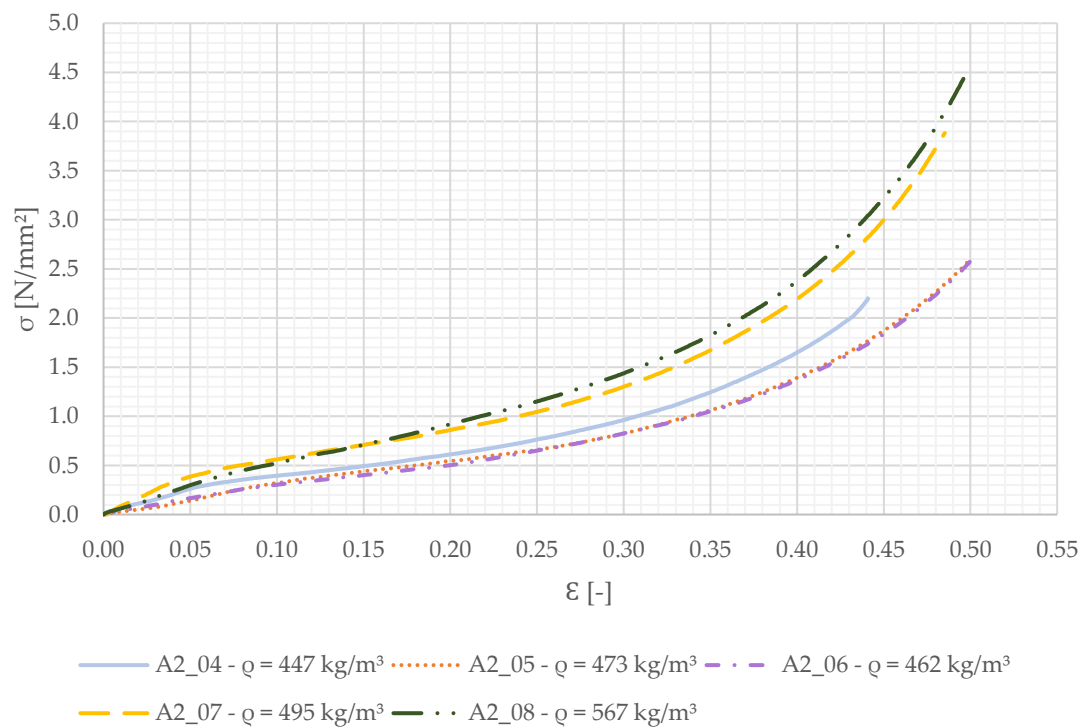


Figure 5.4-5. Stress-strain compressive curve of the *A. donax*-based composite samples, A2.

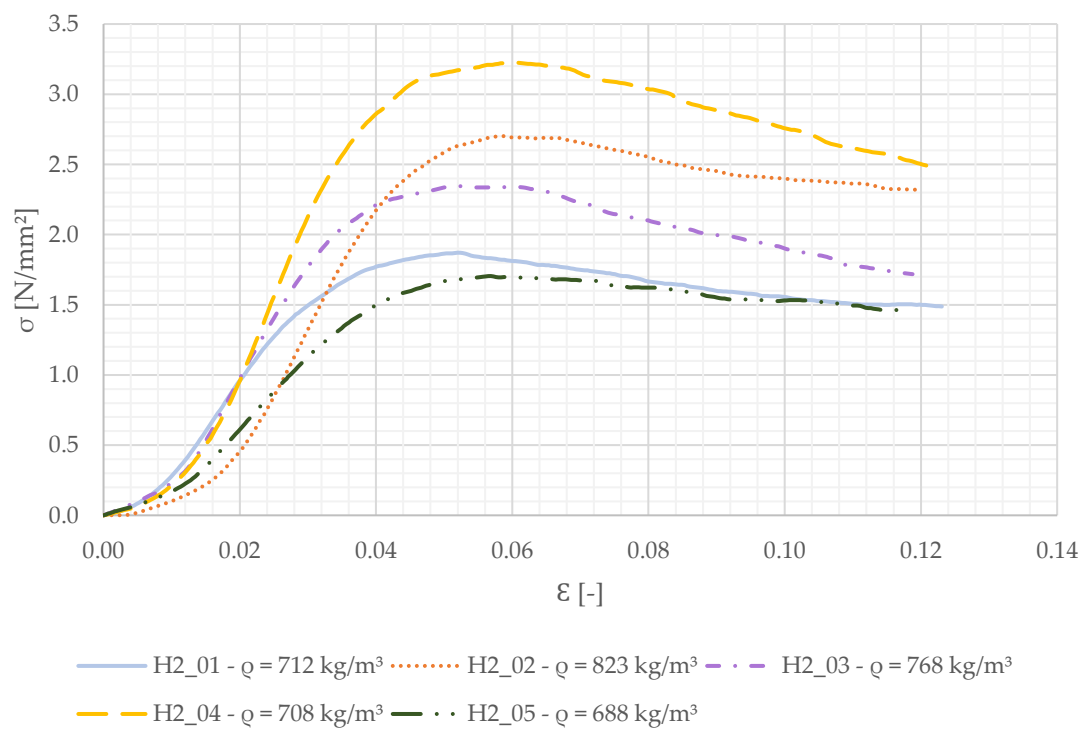


Figure 5.4-6. Stress-strain compressive curve of the hazelnut shell-based composite samples, H2.

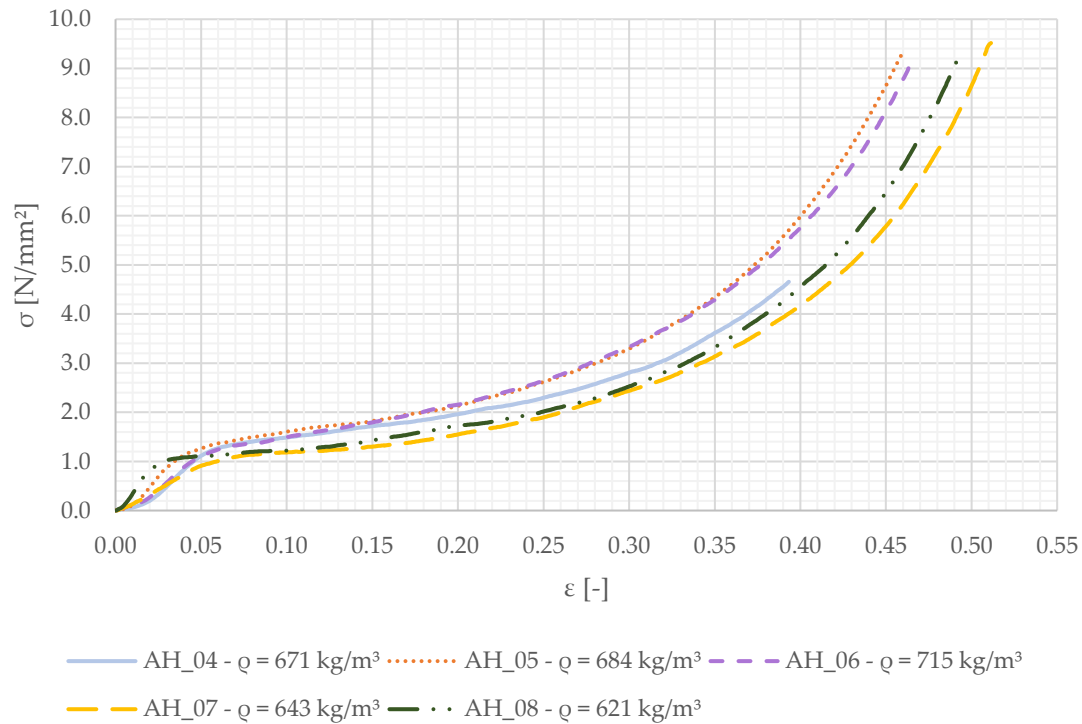


Figure 5.4-7. Stress-strain compressive curve of the mixture-based composite samples, AH.

In the case of the hazelnut shell-based composite (Figure 5.4-6), all the samples achieved the yield. The curves rose, achieved a peak, and then showed a flat post-peak behaviour. As reported in Section 4.3.3.6, the semi-round shape of the hazelnut shells resulted in a low cohesion between the aggregates and the adhesive, leading to the rupture. Hence, the hazelnut shell-based board achieved the breaking due to the interface aggregate-adhesive, which could be considered as brittle materials.

For both the composites the results were in line with the expectation and coherent with the previous result (Chapter 4).

The curves of the composite samples made of the mixture of *A. donax*, and hazelnut shells (Figure 5.4-7) showed an initial increasing curve, then a small flat path, and then another zone with increasing stiffness. The samples did not achieve the yield, and, except for the initial part, they showed a behaviour similar to the *A. donax*-based samples, hence to straw bales. Comparing the behaviour of purely *A. donax* composites (Figure 5.4-5) with the mixture-based one (Figure 5.4-7), for this latter the maximum achieved values of the compressive strength were higher and this curve increased faster.

Table 5.4-3 presents the average values and standard deviation of the maximum force and the corresponding displacement, compressive strength, and modulus of elasticity, calculated as reported in Equation 5.4-3. As anticipated, when the samples did not achieve the yield,

namely for the *A. donax*-based composites with and without the hazelnut shells, A2, and AH, the values were determined considering the 10% strain (thus,  $\varepsilon = 0.1$ ) according to EN 826 (2013).

Table 5.4-3. Average values and standard deviation of the maximum force and the corresponding displacement, compressive strength, and modulus of elasticity.

| Composite | $F_{\max}^*$<br>[N] | $\delta_{\max}^*$<br>[mm] | $\sigma_{\max}^*$<br>[N/mm <sup>2</sup> ] | $E_{(20\%-60\%)}$<br>[N/mm <sup>2</sup> ] |
|-----------|---------------------|---------------------------|-------------------------------------------|-------------------------------------------|
| A2        | 4435 ± 1241         | 4.000 ± 0.100             | 0.4 ± 0.1                                 | 5.12 ± 1.94                               |
| H2        | 25722 ± 7054        | 2.300 ± 0.200             | 2.4 ± 0.6                                 | 82.6 ± 22.9                               |
| AH        | 1693 ± 1223         | 0.420 ± 0.004             | 0.2 ± 0.1                                 | 16.6 ± 13.3                               |

Note: \* For the *A. donax*-based composite, A2, and the mixture-based one, AH, the values were determined considering the 10 % strain as the samples did not achieve the yield (EN-826, 2013).

As expected, the proposed bio-based boards could not be used as structural building products, being the values of compressive strength still low. Comparing them with literature values, they are in line with past studies for non-structural composites (Chapter 4). For example, Antunes et al. (2019) for rice husk-based boards reported values of compressive strength between 0.13 - 0.40 N/mm<sup>2</sup> and a compressive behaviour similar to the one of the hazelnut shell-based composite. Simões et al. (2019) considering expanded cork boards (ICB) achieved a compressive behaviour similar to the one of the *A. donax*-based composite and reported values of compressive strength between 0.154 N/mm<sup>2</sup> (standard density boards) and 0.205 N/mm<sup>2</sup> (medium density boards).

The values of the hazelnut shell-based panels were in line with the ones achieved in the previous experimental campaign (Section 4.3), as the compressive strength evaluated was  $\sigma_{\max} = 2.1$  N/mm<sup>2</sup>. The comparison between the values demonstrated a good reproducibility of the hazelnut shell-boards, despite the heterogeneity of the employed aggregates (i.e., different grain size and uncontrolled distribution within the composite).

For the *A. donax*-based boards, the obtained behaviour was the same as the previous study, but the values were different (Section 4.4). Indeed, the compressive strength and the modulus of elasticity were higher, almost double ( $\sigma_{\max} = \sigma_{10} = 0.9$  N/mm<sup>2</sup>). This was caused by the non-uniform distribution of the *A. donax* and the sodium silicate solution. Indeed, the production process, the casting and curing time, the stabilizing conditions, and the performed test were the same. Further study should be performed to better analyse this property and the differences in the results of the compressive strength. However, the mechanical behaviour under compressive stress was confirmed.

Finally, the mixture-based boards showed a smaller  $\sigma_{\max}$  than the *A. donax*-based ones, A2. However, the addition of *A. donax* to the hazelnut shell-based composite aimed at improving the mechanical performance, and it managed to do it by changing its compressive behaviour. The results showed the prevalence of *A. donax* behaviour on the final mechanical performance even if, as previously reported, the curve showed a different initial path, caused by the presence of the hazelnut shells. This influenced the maximum displacement value,  $\delta_{\max} = \delta_{10}$  which was lower for the mixed-based composite AH. As for the modulus of elasticity,  $E$ , it was more similar to the one of the *A. donax*-based composite evaluated in the previous experimental campaign (Section 4.4). However, even for this optimized composite, the mechanical performance remains low. The results were more useful and interesting for making considerations on the contribution of the employed materials.

## 5.5 Biological susceptibility

### 5.5.1 Context

An experimental campaign was planned and performed to investigate the bio-susceptibility of the bio-based boards. All three composites were considered: the *A. donax*-based, the hazelnut shell-based, and the mixture-based ones. In this paragraph, the considered test methods were described in detail.

As previously anticipated, the samples cannot resist the high relative humidity levels required to perform the tests and the results were considered invalid. Nevertheless, this part of the Ph.D. project was reported as it could contribute to future research. Indeed, methods to assess the bio-susceptibility that can be applied to evaluate other types of bio-based composites were provided. The resistance to decay fungi, initially considered and planned, was not performed due to these results. Hence, the description of this test method has not been reported in this document. Only the resistance to moulds and termites' attack was considered.

### 5.5.2 Materials

The materials used to produce the composites were the hazelnut shells provided by Raccolti di Cin, Baldissero d'Alba, Cuneo, Italy, the *A. donax* harvested in Ozzano dell'Emilia, Bologna, Italy, and the sodium silicate solution,  $\text{Na}_2\text{O} \cdot n(\text{SiO}_2)$ , provided by Ingessil Srl, Montorio, Verona, Italy. The considered production process was the same employed for the other samples (described in detail in Chapter 4).

Three boards for each composition were produced to be cut as the EN 118 (2013), the equipment's size and the test's procedure required small samples. This information influenced the sizes of the produced boards (Figure 5.5-1 a, b, and c), namely 4 cm x 3 cm x length, to be cut each 1 cm.

Due to the grain size of the aggregates and the production process, cutting the boards each 1 cm was not possible. The internal voids caused the breaking of the samples in the cutting phase. Thus, after several practical tests, the experimental campaign was re-defined and adapted to the feasibility of producing samples. The produced boards were cut each 4 cm, and the produced samples were 40 mm x 40 mm x 30 mm (Figure 5.5-1 d, e, and f).

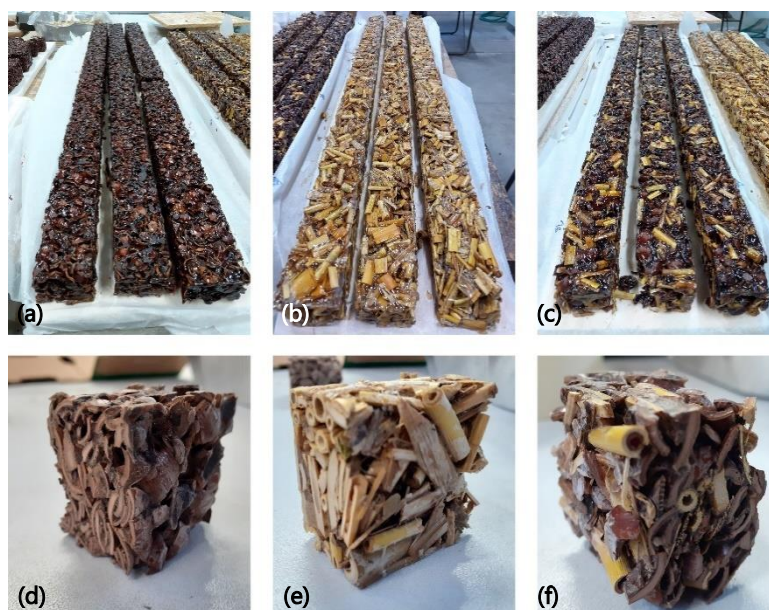


Figure 5.5-1. Samples for bio-susceptibility tests before and after cutting: (a, d) hazelnut shells-based; (b, e) *Arundo donax*-based; (c, f) *Arundo donax* and hazelnut shells-based.

As reported in Section 5.4, the considered designations were A2 (*A. donax*-based composite samples), H2 (hazelnut shell-based composite samples), and AH (mixture-based one), considering the first letter of the employed bio-aggregates. The number 2 was added to distinguish these samples from the previously tested ones (described in Chapter 4).

### 5.5.3 Methods

Four tests were performed, three to assess the resistance to mould growth, and one to evaluate the resistance to termites' attack. For each one, nine replicates (40 mm x 40 mm x 30 mm) of each composite were considered. This is the minimum required number to perform the tests according to the considered references. The test methods were better described in the following sections.

#### 5.5.3.1 Resistance to moulds

Three different tests were carried out, considering past research (Parracha et al., 2021; Santos et al., 2017) and adapting ASTM D5590-17 (2017). They were reported as "Test A", "Test B" and "Test C" and could be summarized as:

1. Test A: Inoculation and culture media
2. Test B: Inoculation without culture media
3. Test C: No inoculation, no culture media.



For Test A, nine flasks for each composition (A2, H2, AH) containing 60 ml of culture media (4% malt, 2% agar) were used to evaluate biological susceptibility to mould. The samples were steam sterilized for 20 min in an autoclave. Then, they were placed into the flasks and inoculated with 1 mL of a mixed spore suspension of *Aspergillus niger* and *Penicillium funicul-losum*. They were left for four weeks at T = 22 °C and RH = 70%. Four Whatman n°1 filter papers (diameter = 45 mm), three samples of Expanded Polystyrene Boards, EPS, and three samples of Insulation Cork Boards, ICB, were used as controls.

For test B, the nine samples for each composition were steam sterilized for 20 min in an autoclave and placed into nine flasks and inoculated with 1 mL of a mixed spore suspension of *Aspergillus niger* and *Penicillium funicullosum*. They were left for four weeks at T = 20 °C and RH > 90%. Three samples of an Expanded Polystyrene Boards, EPS, and three samples of an Insulation Cork Boards, ICB, were used as controls.

Finally, For Test C, the nine samples for each composition were placed into plastic containers without being sterilised or inoculated, following the same test described in Section 3.3.2.3.4. The plastic containers were kept open and spaced and placed in a climatic chamber at T = 20 °C, RH > 90%. Natural ventilation was prevented by placing all the samples into a box. Three samples of Insulation Cork Boards, ICB, and three samples of Expanded Polystyrene Boards, EPS, 40 mm x 40 mm x 30 mm, were used as controls.

All the samples were visually analysed each week both by the naked eye and by using a stereo microscope, Olympus SZX12. For Test C only one of the nine samples was monitored by using the stereo microscope.

The mould growth for all the testes was estimated considering the classification provided in Table 5.5-1, adapted from ASTM D5590-17 (2017).

Table 5.5-1. Rate of mould growth adapted from ASTM D5590-17 (2017).

| Rating | Description      | Contaminated surface [%] |
|--------|------------------|--------------------------|
| 0      | None             | 0                        |
| 1      | Traces of growth | < 10                     |
| 2      | Light growth     | 10 to 30                 |
| 3      | Moderate growth  | 30 to 60                 |
| 4      | Heavy growth     | > 60                     |

### 5.5.3.2 Resistance to termites

Nine samples (40 mm x 40 mm x 30 mm) were cut from three different boards (considering the same percentages of aggregates-adhesives) and they were evaluated considering the EN 118

(2013), without any treatment. The samples were conditioned at  $T = (20 \pm 2) ^\circ\text{C}$  and  $\text{RH} = (65 \pm 5) \%$  for two weeks. A ground glass end of tubes (interior diameter = 25 mm; length = 110 mm) was bonded with resin at the centre of the surface of each sample. To ensure this procedure, a thin wooden board was glued to the top of the samples and the glass tube was glued to this part. Sand and water (1:4 v/v water-sand ratio) were placed into the tube until two-thirds of the volume of the entire tube was filled. 250 worker termites were placed into the tube, and it was closed with a piece of aluminium foil to avoid both the evaporation of the water and the escaping of the termites (Figure 5.5-2).



Figure 5.5-2. Samples to evaluate the resistance to termites according to an adaptation of EN 118 (EN 118, 2013) with the glass tube glued to the wooden boards.

The samples were placed at  $T = 24 ^\circ\text{C}$  and  $\text{RH} = 70\%$ , each one in a glass box, and monitored for 8 weeks. At the end of the tests, the grade of attack was rated as described in Table 5.5-2 and the percentual moisture content and termites' survival were determined. Maritime pine (*Pinus pinaster* Ait.) untreated solid wood samples were used as control (survival rate of the termites for the control must be more than 50%).

Table 5.5-2 – Rate of termites' attack, adapted from EN 118 (2013)

| Rating | Evaluation                                                                                                                                                                                                                                                                   |
|--------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 0      | No attack                                                                                                                                                                                                                                                                    |
| 1      | Superficial erosion of insufficient depth to be measured on an unlimited area of the test sample, or attack to a depth of 0.5 mm provided that this is restricted to an area or areas not more than 30 mm <sup>2</sup> in total                                              |
| 2      | Erosion of 1 mm in depth limited to not more than 1/10 of the surface area of the test sample; or single tunnelling to a depth of up to 3 mm                                                                                                                                 |
| 3      | Erosion of < 1 mm in depth over more than 1/10 of the surface area of the test sample, or erosion of > 1 mm to < 3 mm in depth limited to not more than 1/10 of the surface area of the test sample, or isolated tunnelling of a depth > 3 mm not enlarging to form cavities |
| 4      | Erosion of > 1 mm to < 3 mm in depth of more than 1/10 of the surface area of the test sample, or tunnelling penetrating to a depth > 3 mm and enlarging to form a cavity in the body of the test sample                                                                     |

## 5.5.4 Results and discussion

### 5.5.4.1 Resistance to moulds

As previously reported, all the samples were assessed each week, by necked eye and by using the stereo microscope (Olympus SZX12) to confirm the grading results.

For Test A, namely the assessment of mould growth with inoculation and culture media, all the samples were rated as 0 (according to the classification provided by (ASTM D5590-17, 2017), reported in Table 5.5-2). However, the test was considered invalid as the sodium silicate solution passed into the culture medium and the samples "melted". In some of the *A. donax*-based samples, there was a small mould growth in the culture media. In some of the mixture-based samples some moulds grew on one side of the samples, but just a few spots, probably due to the melted sodium silicate solution. There was no growth on the top of the samples.

As for the controls, in the filter paper mould growth was normal and pronounced. In one of the EPS samples, mould grew in one of the corners. In ICB samples, a not accentuated mould growth was detected. Figure 5.5-3 represents the samples and the controls at the end of the test (after 4 weeks).

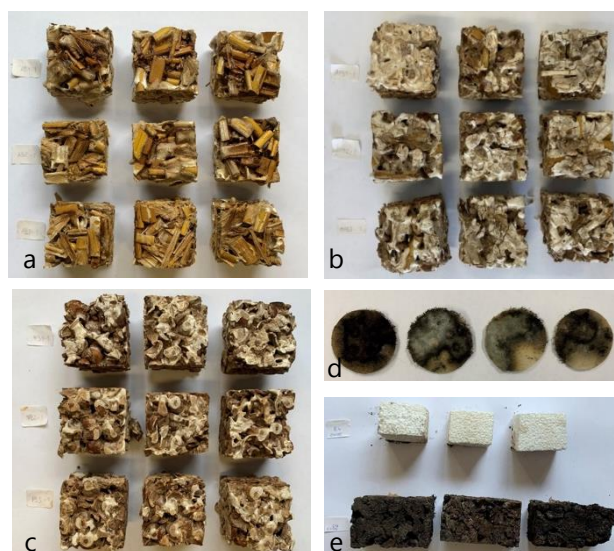


Figure 5.5-3. Samples after 4 weeks of exposure to culture medium and spore solution (test A): (a) *A. donax*-based samples; (b) hazelnut shell-based samples; (c) mixture-based samples; (d) filter paper; (e) expanded polystyrene samples (EPS) and expanded cork boards (ICB).

For Test B, namely the assessment of mould growth with inoculation and without culture media, all the samples were rated as 0 according to the classification provided by (ASTM D5590-17, 2017) (Table 5.5-2). One sample made of hazelnut shells broke due to the melting of the sodium silicate solution. Hence, it was not evaluated. Some other samples showed some sodium silicate solution at the bottom, but they did not break, and they were evaluated. However, it is important to underline that, due to these conditions, the results could not be considered valid, and the test should be performed again, changing the considered conditions or the employed sodium silicate solution to secure a better resistance to high RH. However, in this latter case, a new composite would produce, and then it should be re-characterized, because other properties could be changed. Nevertheless, this provided important information about the low resistance of the sodium silicate at high RH (in this case higher than 90%, constantly for 4 weeks). As for the controls, there was no mould growth on the EPS samples, while there was a small spot of mould growth on the expanded cork samples.

For Test C, namely the assessment of mould growth without inoculation, no mould growth on any of the samples or controls was detected (Figure 5.5-4).

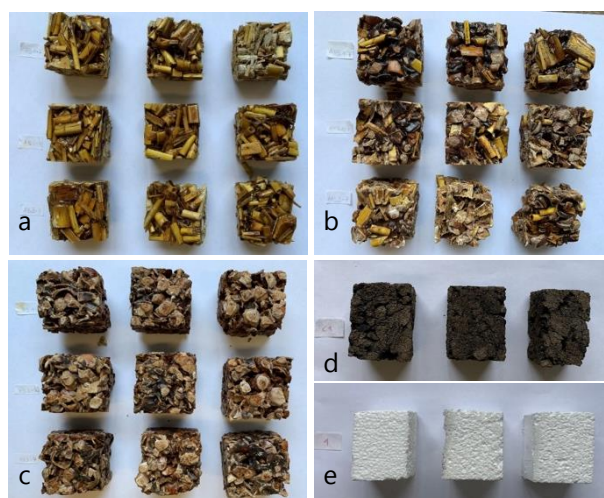


Figure 5.5-4. Samples after 4 weeks of exposure at high RH, without inoculation: (a) *A. donax*-based samples; (b) hazelnut shell-based samples; (c) mixture-based samples; (d) expanded polystyrene samples (EPS); (e) expanded cork boards (ICB).

Good resistance to mould growth was expected due to the sodium silicate solution. Indeed, according to past research (Lee and Thole, 2018; Liuzzi et al., 2020a) and a previous experimental campaign (Section 3.3), the sodium silicate demonstrated high resistance to biological attack and chemical decomposition. This was precisely one of the reasons for choosing this binder. However, the results of bio-susceptibility to mould cannot be considered valid due to the uncertainties about the execution of the test, mainly caused by the "melting" of the sodium silicate solution.

Although the results highlighted an important drawback of the proposed boards, namely the low resistance to very high RH, it's important to pointed out that the conditions considered (RH almost 100%, constantly, for a long time) are not usually reached in indoor environments.

Some possibilities to overcome this weakness should be the addition of some additives, such as lime ( $\text{Ca}(\text{OH})_2$ ), calcium chloride ( $\text{CaCl}_2$ ), sodium chloride ( $\text{NaCl}$ ), or cement ( $(\text{CaO})_3\cdot\text{SiO}_2$ ), investigating and verifying also which type. This information was provided by the sodium silicate producer and should be tested.

Another possibility could be considering lower maximum values of RH, although this could compromise the validity of the tests. Indeed, the considered conditions were selected according to the standards and because they are favourable to mould growth. Hence, in this case, a new test method should be defined, implemented, and validated.

#### 5.5.4.2 Resistance to termites

During the third week of monitoring, dead termites appeared. At the end of the test, the wood glued to the samples was soaked and the samples were stuck to the bottom of the glass box. Hence the sodium silicate seemed to be melted a bit.

Although the termites tried to settle in and cross the test samples, there was 0% termite survival, while in the control tests survival was over 50% (confirming the validity of the test's set-up). However, the test was not considered valid, and it must be re-performed.

The same considerations made for the test to assess the resistance to mould could be made for the termites' attack. Although the "melting" of the samples determined the invalidity of the test, a good resistance was expected also to termites. Indeed, as reported in the technical sheet provided by Ingessil (1998), the ingestion toxicity of the sodium silicate is extremely low but could cause diseases and the possibility of death if ingested in very high quantities. These vary according to the concentration percentual weight of the solution and the weight ratio of the sodium silicate. In this case, if related to termites' weight and considering that the boards should be the only insects feeding for eight weeks, it could be lethal. Nonetheless, this information should be verified and confirmed by laboratory tests.

## 5.6 Life cycle assessment

### 5.6.1 Context

The final step of the PhD project addressed the sustainability aspects of the analysed bio-based boards and aimed at describing their eco-efficiency. The information to determine the emissions of the composites was set. The LCA and the analysis by using a specific software have not been carried out. However, the initial studies and considerations were presented in this section together with some qualitative considerations.

The Life Cycle Assessment (LCA) is one of the developed techniques to define the possible impacts associated with manufactured and consumed products (ISO 14040, 2006; ISO 14044, 2006). This tool has been employed to define both the impacts and the advantages related to different policy choices (Cecere et al., 2024). It has been applied in several fields, such as organic waste management (Bottausci et al., 2023), urban infrastructures (Corominas et al., 2020), site remediation services (Morais and Delerue-Matos, 2010), but also to evaluate the socio-economic impacts of processes, products, and services (Huertas-Valdivia et al., 2020).

LCA is widely employed in the construction sector (Santos et al., 2021), as it allows identifying the impact of the construction materials during their entire life (Barbhuiya and Das, 2023), providing important support for decision-making (Almeida et al., 2021).

Although LCA is usually performed during the early-stage design decisions (Cascione et al., 2022; Roberts et al., 2020), here it was considered at the final stage. A detailed analysis could not be carried out in an early-stage design due to the unconventional aspects of the composites and their ongoing definition, which determined too much unknown information. One of the weaknesses of the LCA is indeed the requirement for a large number of accurate and reliable data (Barbhuiya and Das, 2023; Santos et al., 2021). Hence, LCA was used as an assessment complementary tool to the study performed during the Ph.D.

According to the reference standards (ISO 14040, 2006; ISO 14044, 2006), four iterative phases have to be considered to carry out an LCA analysis:

1. The goal and scope definition;
2. The life cycle inventory analysis (LCI);
3. The life cycle impact assessment (LCIA);
4. The interpretation phase.

After defining the goal of the study, the scope of the LCA study was defined.

Within the scope of the study, the functional unit, the reference flow, and the system boundaries are outlined.

The functional unit refers to the quantified performance of a product system for use as a reference unit. The reference flow is the quantification of the outputs provided by the considered product system, required to define the function described by the functional unit. As for the system boundary, it describes which unit processes are part of the system under study (ISO 14040, 2006; ISO 14044, 2006). For this latter, four different possibilities can be considered:

- Cradle to Grave, which considers the entire process, from the raw material extraction to the product's end-of-life treatment;
- Cradle to Gate, which considers the processes from the raw material extraction until the end of the production phase;
- Gate to Grave, which considers the processes from the use until the end-of-life
- Gate to Gate, which considers the processes from the production phase only.

The system boundaries are reported in Figure 5.6-1, adapted from Bossagh and Hassanzadeh (2012).

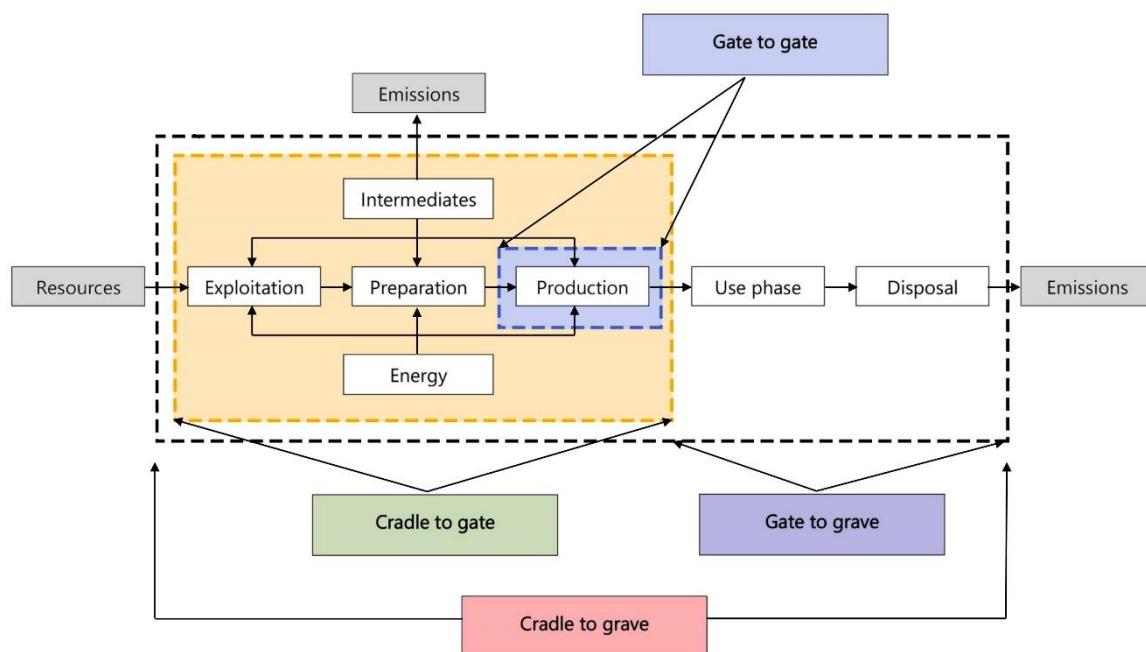


Figure 5.6-1. System boundaries (ISO 14044, 2006) adapted from Bossagh and Hassanzadeh (2012).

After defining the first aspects, the second phase, namely the LCI, could be carried out. The LCI reports all relevant data describing the considered processes (Morais and Delerue-Matos, 2010). The more minutely the data collection, the more precise the LCA (Barbhuiya and Das, 2023). The source of data could be different, for example primary data, provided directly from the producer, secondary data, estimate or determined by investigating and collecting information, software database (etc.).



Furthermore, collected the inventory data, the LCIA can be performed. At this stage, a specific software can support the analysis. The selected functional unit must be always considered to describe the outcomes. Interpretation and normalization ensure the understanding and the comparison between the results (Barbhuiya and Das, 2023; Owens, 1997).

This information makes clear the complexity of performing a detailed LCA study, independently from the type of product/process. The initial choices (i.e., the functional unit, the reference flow and boundary system) widely influence the type of analysis and the output. .

## 5.6.2 Performed analysis and discussion

Based on the information reported in Section 5.6.1, the life cycle assessment of the *A. donax*-based, and hazelnut shells-based boards was set up. As anticipated in Section 5.6.1, the LCA was not carried out quantitatively; only some qualitative considerations were made (e.g., the definition of goals, scope, and boundary system and the set-up of LCI).

Figure 5.6-2 reports the conceptual scheme drawn in the initial stage. This ensured the definition of the goal and scope of the LCA as well as the boundary system (Cradle to Gate).

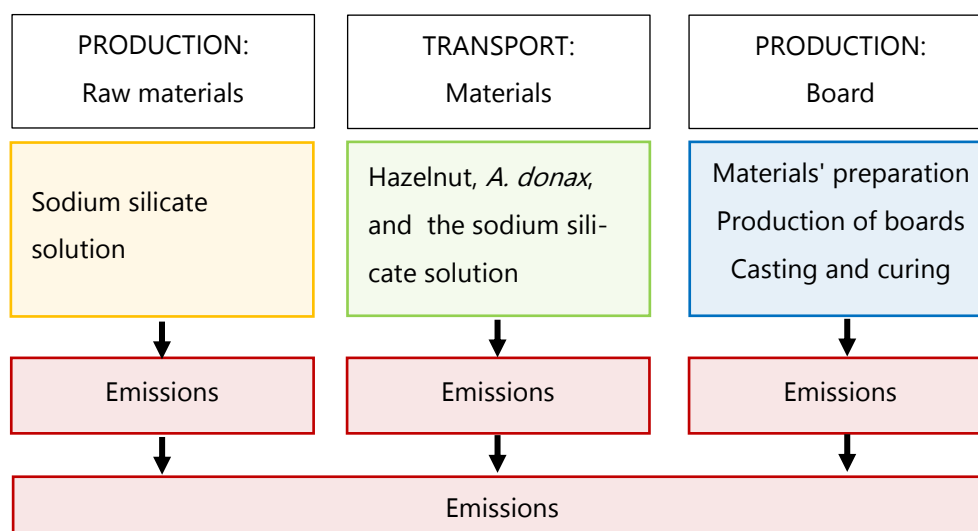


Figure 5.6-2. LCA starting chart to synthesize the considered phases and parameters for the considered bio-based boards.

A boundary system Cradle to Gate was chosen to evaluate the environmental impact of the process defined and described in this Ph.D. project. The evaluation of a different boundary system would not have been significant at this stage. As for the emissions, different ones can be considered, such as in air, in water, waste generation (etc.). The carbon footprint of the

boards could be assessed by considering the CO<sub>2</sub> production only, and this would provide initial information about their sustainability.

One board was considered as the functional unit. As for the collected data, primary and secondary data were considered in this first stage. The primary data would be defined according to the Ph.D. project, namely all the information reported in the previous Chapters; the secondary ones would be provided by the producer or by carrying out investigations (e.g., technical data sheet of the sodium silicate solution, literature, etc.).

The LCI was set up and a summarized list of the required information was drawn (Table 5.6-1). The reported data should be collected to perform the LCA of the proposed bio-based boards.

Table 5.6-1. Draft of the Life Cycle Inventory analysis to perform the LCA of the considered bio-based boards.

| Phase                                      | Data source    | Note                                                                       |
|--------------------------------------------|----------------|----------------------------------------------------------------------------|
| <b>Production process of raw materials</b> | Secondary data |                                                                            |
| Hazelnut shells                            |                | Considered by-products                                                     |
| <i>A. donax</i>                            |                | Considered by-products                                                     |
| Sodium silicate                            |                |                                                                            |
| Drying (1400°C)                            |                | There are two different processes to produce the sodium silicate solution. |
| Cooling                                    |                |                                                                            |
| Hydrated, dissolved in water 140-160°C     |                | The one reported was described by (Ingessil, 1998)                         |
| Purified by decantation and/or filtration  |                |                                                                            |
| Storage tank                               |                |                                                                            |
| <b>Transport (from producer to Lab)</b>    | Secondary data |                                                                            |
| Hazelnut shells                            |                | Distance = 320 km                                                          |
| <i>A. donax</i>                            |                | Distance = 20 km                                                           |
| Sodium silicate                            |                | Distance = 160 km                                                          |
| <b>Production process - 1 board</b>        | Primary data   |                                                                            |
| Drying bio-aggregates T = 60°C             |                | The defined production process                                             |
| Mixer                                      |                |                                                                            |
| Drying at T = 60 °C                        |                |                                                                            |

The data collection has not been carried out, but some qualitative considerations were made. First, hazelnut shells and *A. donax* would not be considered in the production phase as they are by-products. In this case, they do not provide any apport on the carbon footprint/emissions produced in the raw materials production phase. A positive impact could be considered by the CO<sub>2</sub> capture along the growing of the *A. donax* and hazelnut trees. Their main negative impact would derive from their transport. As both were largely available and produced in the considered area (Chapter 2) and they were provided by local producers (Chapter 4), CO<sub>2</sub> production and emissions in general were expected to be relatively low. This aspect was considered at the

stage of choosing the materials to secure less environmental impact derived from the transport.

As for the production process, a low energy requirement, CO<sub>2</sub> production, emissions, and environmental impact were expected. Indeed, high temperatures, mechanical pressing, thermal or chemical treatment of the materials and casting and curing phases were defined aiming at securing low energy requirements. The maximum employed temperature in the production process was  $T = 60\text{ }^{\circ}\text{C}$ ; the materials were mixed, then placed in the moulds, and left dried, and the casting and curing phases were carried out at laboratory conditions.

Considering a qualitative evaluation, a higher energy requirement should be determined by the sodium silicate production. However, as previously anticipated, these considerations should be confirmed and verified by the data collection and by specific software.

## 5.7 Concluding remarks

A complete characterization of the proposed bio-waste-based composites was provided by carrying out previously considered characterization and complementary ones. In addition to the evaluation of bio-based boards' properties, the contribution of the employed materials (aggregates and adhesives) and their formulation on the final performance of the composite boards was investigated. The studies described in this Chapter, namely the analysis of the internal structure, the performance in case of fire, and the assessment of biological susceptibility are not so commonly performed. Thanks to the collaboration between the University of Bologna, the National Laboratory of Civil Engineering and the NOVA University Lisbon, there was the possibility of working with different departments and laboratories to perform these new studies. The results of this experimental campaign allowed achieving many interesting concluding remarks.

The addition of *A. donax* completely changed the hazelnut shells-based composite's compressive behaviour. The prevalence of *A. donax* on the final mechanical performance was evident. However, the compressive behaviour of the mixture-based composite was different from the *A. donax*-based one. This, again, highlighted the importance of knowing the materials' properties to properly use them in composites, as they could deeply change the outcome namely when they are mixed.

As for the acoustic absorption, although differences in the bio-aggregates' behaviour were detected, it seemed that it was more influenced by the combination aggregates-adhesive rather than the bio-aggregates. Indeed, the results suggested that the sound absorption capacity was widely affected by the sodium silicate's distribution on the samples' surface. However, for all three composites (with each of the two bio-aggregate alone and the mixture of them), the acoustic absorption behaviour remained extremely interesting and promising, as well as not conventional for these types of composites. The highest values of the sound absorption coefficient were indeed in the frequencies of the human voice. Further studies could better explain these aspects, such as the assessment of the materials' acoustic absorption capacity individually, the evaluation of this property by considering different thicknesses, and the analysis of the correlation with the results of the internal structure.

The high hygroscopicity of the materials and the composites was proved. The analysis demonstrated that this property was widely increased by the sodium silicate solution, and this seemed to be confirmed by the statistical analysis too. The results suggested that a convenient application of bio-based boards would be as indoor coatings. Indeed, the possibility of

passively controlling both the acoustics and the hygrothermal conditions indoor, passively improving comfort, was confirmed.

The bio-based boards' performance in case of fire was extremely promising. All three composites were compliant with the A2 class requirement for fire reaction classification. The addition of the sodium silicate solution avoided the burning of the bio-aggregates and the smoke production. This both demonstrated the possibility of using these boards as indoor coatings without any risks in case of fire and suggested a solution to overcome one of the main drawbacks of bio-based composites. Indeed, a sodium silicate solution-based superficial treatment applied on these and other bio-based composites' surfaces, might be effective for further improving the performance in case of fire. The analysis of the materials' formulation was carried out for the hygroscopicity and performance in case of fire, by considering both the raw materials and the composites. Although it ensured considerations of materials' behaviour, chemical analysis should be performed. This would contribute to further consideration of materials' influence on bio-based composites' final properties. However, the achieved results demonstrated the high influence of the adhesive, in the final performance.

The tests carried out to assess the bio-susceptibility have to be repeated. However, the results pointed out both a weakness (low resistance to very high relative humidity) and a possible strength (resistance to biological attack) of the proposed composites due, in particular, to the sodium silicate solution. In case of the results' confirmation and validation, this finding could be beneficial for bio-based composites as the sodium silicate could be a solution to avoid this drawback. Hence, a sodium silicate solution-based superficial treatment applied on these and other bio-based composites' surfaces, in addition to the performance in case of fire, might improve the resistance to biological attack. Nevertheless, this information has to be confirmed by further analysis.

Although the life cycle assessment was not performed, some initial considerations suggested that the boards could be considered ecologically efficient. Indeed, the highest energy requirement, CO<sub>2</sub> production and emissions would be caused by the sodium silicate solution, while the contribution of the other bio-materials and the production process would be low. However, these considerations are only qualitative. The information reported in this chapter provided some preliminary information for performing an LCA study in the future.

Finally, this part of the PhD project, in addition to the characterization of the composites and the analysis of materials' properties, provided important guidelines for a horizontal analysis, namely extended to different fields. This information could be useful for future studies on unconventional bio-waste-based composites.



## CONCLUSIONS

### 6.1 Final remarks

This Ph.D. project is a contribution to the development of bio-waste-based building composites (in particular, particleboards for indoor use) by sustainable building practices. Several possibilities for producing boards for indoor use were investigated by considering different materials. First, it focused on some bio-aggregates available in Euro-Mediterranean Countries: grapes and olives press wastes, hazelnut shells, and spent coffee grounds. Then, it considered different adhesives (a hemihydrate gypsum-based binder, a sodium silicate solution, and a starch-based glue), and additives (air lime and citric acid). Finally, the addition of another bio-resource, *Arundo donax*, (also known as giant reed or giant cane) to optimize the considered hazelnut shells-based composite.

The bibliographic research supported the selection of the materials and allowed knowing different laboratory tests and methods to evaluate raw materials and composites' properties. The results of this first step of the project demonstrated that many different standardized methods are used, but they are especially for conventional composites rather than for raw materials. Furthermore, the literature's gaps were pointed out.

The experimental campaigns and the practical experiences allowed getting in touch with the possible practical problems derived from the boards' production process, the use of the selected materials, and the definition and implementation of the laboratory tests. As an outcome, a simple low-energy production process for the considered composites was defined. Furthermore, the importance of developing practical tests by using the selected materials, and experimental activities in this type of research, as a support to the theoretical information, was demonstrated.

Both standardized and unstandardized test methods were considered, proposing new ones as well, since the studied materials and products are yet considered unconventional for civil engineering. Furthermore, properties not so commonly assessed were considered, such as bio-susceptibility, performance in case of fire, and analysis of the internal structure by X-ray computed tomography.

The experimental campaigns were implemented thanks to several collaborations that involved people from different Departments and Universities, qualified in different research fields. In addition to the Department of Civil Engineering, NOVA School of Science and Technology, NOVA University Lisbon, and the Structure Department, National Laboratory of Civil Engineering (LNEC), Portugal, part of the samples' production and the experimental campaign was carried out at the Department of Civil, Chemical, Environmental, and Materials Engineering, the Department of Industrial Engineering, the Department of Industrial Chemistry, and the Department of Physics and Astronomy, Alma Mater Studiorum, University of Bologna, Italy. Furthermore, professors of the Department of Civil Engineering Sciences and Architecture, Politecnico University of Bari, Italy, the staff of Ingessil S.r.l., and the hazelnut shells producer provided important support in samples' production and the analysis of their properties. All these collaborations allowed expanding the investigation fields, enriching the research project, and achieving important conclusions.

Some tests were not initially planned, but their development was of great interest. Some others, initially considered, were modified, or not performed (e.g., bio-susceptibility). The results of the experimental campaigns demonstrated that the selected materials and production process did not allow the development of thermal insulator bio-particleboards. Indeed, the composites did not meet the standard requirements for insulation boards, although they demonstrated thermal insulation capacity. Nevertheless, other properties seemed promising, and they were further detailed during the final stage of the project, together with an initial Life Cycle Assessment.

The achieved conclusions can be summarized as follows:

- All the bio-aggregates, adhesives, and additives considered in the bibliographic research are suitable to produce bio-based building composites that can perform well for several requirements. Several properties can be investigated to provide a general characterization, and the results can be an important starting point for developing future scientific studies aiming at proposing new eco-efficient building products. There is a need to standardize experimental tests, so that results can be comparable and validated.



- Considering the described production process, the selected agro-industrial wastes (grapes and olives press wastes, hazelnut shells, and spent coffee grounds) and the wood chips with starch-based natural glue may be suitable to produce thermal insulation boards; with the gypsum-based binder, thermal plastering mortars; with the sodium silicate solution, coating boards that can control indoor conditions.
- The analysed bio-aggregates, namely, hazelnut shells and *A. donax*, combined with the sodium silicate solution, can be used to produce composites with interesting and promising acoustic properties, to be employed as indoor coatings to improve acoustic comfort. In addition to being able to absorb frequencies of the human voice, they showed an uncommon acoustic absorption behaviour, not typical for this type of material.
- Both the considered bio-wastes and the sodium silicate solution demonstrate a high moisture response and this propriety can ensure a passive control of the indoor conditions. However, it can be a hazard of composites' degradation. The analysis carried out to better understand this performance, namely the influence of materials' formulation by an X-ray computed tomography, demonstrates that the moisture buffering capacity is widely influenced by the sodium silicate solution.
- The results of the fire reaction are extremely promising, as the composites are classified as A2 according to fire classification. Hence, the sodium silicate solution overcomes one of the main drawbacks of bio-based composites, namely the low resistance to fire. The burning and the smoke production are avoided, indeed, demonstrating the possibility of using these boards as indoor coatings without any risks in case of fire.
- Although the tests to assess the bio-susceptibility of the hazelnut shell-based, *A. donax*-based and mixture-based boards must be repeated, the composites made by using the sodium silicate solution as the adhesive show promising performance. No biological colonization is detected after placing the samples at high relative humidity for a long time, differently from the composites produced by using starch-based natural glue and gypsum-based binder. Hence, the sodium silicate seems able to also overcome this bio-based composite drawback. However, further studies should be carried out, to assess the other types of biological attack as well.
- Although a qualitative Life Cycle Assessment (LCA) was not performed, some considerations suggested promising results in terms of the sustainability of the bio-composites. The energy requirement and CO<sub>2</sub> production would be mainly determined by the sodium silicate solution. However, these considerations are only qualitative and must be confirmed.

In addition to the conclusions achieved from the results of the studies, it is important to underline another aspect of this PhD project: the described experimental campaigns, as well as

the data collection, provided a methodology to characterize innovative building materials by a horizontal analysis, namely considering different properties and research fields.

## 6.2 Future studies

The Ph.D. project contribution to the research for the future development of bio-based building composites includes ideas for the future, reported in this Section.

First, starting from the provided information, the LCA should be performed quantitatively by specific software (e.g., SimaPro) to quantify the emissions of the boards, hence their sustainability. Different boundary systems and data sources could be considered, comparing the outcomes. Furthermore, the results could be compared to commercial or already studied boards as well. Then, a cost analysis could be carried out.

Although the boards would be employed for indoor applications, the resistance to water and water absorption could be interesting information to be evaluated. Moreover, also the dimensional stability could be determined by immersing in water, or by considering different temperatures and relative humidity levels. Some screening tests were carried out qualitatively to assess these properties; however, standardized scientific methods should be implemented and performed to better define them. Tests to assess the durability of the proposed boards could be interesting to perform, in addition to the ones that partially consider this aspect (namely the bio-susceptibility and reaction to fire). Furthermore, the possibility of covering the boards with a finishing or a plaster could be investigated, as well as how this coating would modify the final performance.

The acoustic properties of the proposed boards could be further investigated by performing additional tests. For example, the evaluation of the acoustic absorption capacity of the materials separately (the bio-aggregates and the dried sodium silicate solution), and the composite samples' acoustic properties at different thicknesses and by modifying the external surfaces (e.g., by sanding the sodium silicate layer if the adhesive was not homogeneously distributed). Higher frequencies could be considered, and the air flow resistivity could be determined for all the composites. This latter, together with the internal distribution of voids (i.e., the results of the tomography) could be used to better explain the acoustic properties. The results could be employed to carry out simulations with specific software (e.g. MATLAB) and to define an acoustic predictive model, not already presented in the literature.

The hygroscopicity could be further analysed, as well. By additional tests, it could be determined when stability could be achieved when the composites are exposed to high relative

humidity. Studies on the desorption phase could be carried out, as the moisture release behaviour would widely influence the control of indoor hygrothermal conditions. The correlation between hygroscopicity, and acoustic and mechanical properties could be investigated, and the definition of a critical moisture level could provide further information on biological susceptibility. As for this latter, the resistance to moulds, fungi, and termites should be assessed by implementing and performing test methods, suitable for the considered samples.

The test to evaluate the compressive behaviour of *A. donax*-based samples suggested pressing at low temperatures as another possible production process. This would probably ensure the feasibility of producing prototype samples, that could be tested and compared to the ones analysed in this Ph.D. project.

The samples after the test to assess the performance in case of fire could be analysed as well. This would define how the application of constant heat could change materials' properties. Furthermore, the results of this test suggest that a sodium silicate-based superficial treatment could be a solution to improve bio-based composites' performance in case of fire. An interesting future research would be the assessment of this possibility, to define if and how the sodium silicate could overcome this drawback also in other bio-based building products.

Finally, all the considered materials, both the aggregates and the adhesive, could be employed to produce other innovative eco-efficient composites that could control hygrothermal comfort (i.e., thermal plastering mortars and/or thermal insulation boards). However, as these possibilities were considered in the first step of the Ph.D. and not analysed in detail, other applications or implementations could be found. Another application of some results from these studies could be the replacement (totally or partially) of wood chips in wood-based boards by the considered bio-aggregates. Analysis of this possibility and the variation of the final performance due to the percentages of the materials could be carried out. Moreover, the proposed composites could be optimized by modifying some parameters, such as grain size, aggregate-binder percentages, the production method (as already mentioned previously), and the casting and curing phases. Other tests and analyses could be performed according to the final application of the proposed boards, such as the evaluation of VOC emissions, and the possibility of improving indoor air quality by absorbing pollutants. The possible installation of the boards should be investigated, as well as possible covering and finishing.

The described project could be repeated and applied considering another region in the world, then other bio-wastes and bio-resources, and hence, other bio-composites.



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## APPENDIX

Supplementary material related to Article A (Chapter 2) and Article I (Chapter 5) are reported in this Section.

Table SM 1 reports the detailed results related to the chemical composition of agro-industrial wastes used for building materials considered in Table 2.2-5 in Chapter 2. It can be found, in the online version, at doi: <https://doi.org/10.1016/j.indcrop.2021.113833>.

Table SM 1. Chemical composition of several bio-wastes considering research works published in the last twenty years (2000-2020).

| Material             | Chemical composition [%] |                |             |       |      | References                      |
|----------------------|--------------------------|----------------|-------------|-------|------|---------------------------------|
|                      | Cellulose                | Hemicelluloses | Lignin      | Waxes | Ash  |                                 |
| Banana               | 60.00-65.00              | 19.00          | 5.00-10.00  | -     | -    | (Jawaid and Abdul Khalil, 2011) |
|                      | 63.00-67.60              | 10.00-19.00    | 5.00        | -     | -    | (Dittenber and GangaRao, 2012)  |
|                      | 63.00-64.00              | 19.00          | 5.00        | -     | -    | (Idicula et al., 2006)          |
|                      | 64.00                    | 10.00          | 5.00        | -     | -    | (Nguong et al., 2013)           |
|                      | Range 60.00-65.00        | 10.00-19.00    | 5.00-10.00  | -     | -    |                                 |
| Coco-nut fibres/coir | 64.00                    | -              | -           | -     | 0.74 | (Phan et al., 2006)             |
|                      | 51.12                    | 67.63          | 36.73       | -     | -    | (Panyakaew and Fotios, 2011)    |
|                      | 31.60                    | 25.50          | 35.10       | -     | -    | (Nascimento et al., 2016)       |
|                      | 52.20                    | 28.40          | 19.40       | -     | -    | (García et al., 2016)           |
|                      | 32.00-43.80              | 0.15-20.00     | 40.00-45.00 | -     | -    | (Dittenber and GangaRao, 2012)  |
|                      | 32.00-43.00              | 0.15-0.25      | 40.00-45.00 |       |      | (Faruk et al., 2012)            |
|                      | -                        | -              | 45.84       | -     | -    | (Khedari et al., 2004)          |

| Material | Chemical composition [%] |                |             |       |           | References                                    |
|----------|--------------------------|----------------|-------------|-------|-----------|-----------------------------------------------|
|          | Cellulose                | Hemicelluloses | Lignin      | Waxes | Ash       |                                               |
| Range    | 32.00-43.00              | 0.15-0.25      | 40.00-45.00 | -     | -         | (Jawaid and Abdul Khalil, 2011)               |
|          | 21.00-43.00              | 0.25-22.00     | 27.00-47.00 | -     | -         | (Zhou et al., 2010)                           |
|          | 21.00-64.00              | 0.15-67.63     | 19.40-47.00 | -     | 0.74      |                                               |
| Coffee   | 22.83                    | 19.30          | 28.59       | -     | 5.94      | (Buratti et al., 2018)                        |
| chaff    | 13.00-24.00              | 11.00-12.00    | 21.00-32.00 | -     | -         | (Quosai et al., 2018)                         |
|          | 23.60                    | 12.10          | 17.80       | -     | 6.90      | (Zarrinbakhsh et al., 2016)                   |
| Range    | 13.00-24.00              | 11.00-19.30    | 17.80-32.00 | -     | 5.94-6.90 |                                               |
| Coffee   | 8.80                     | 44.50          | 13.10       | -     | -         | (Pedras et al., 2019)                         |
| ground   | 10.00                    | 40.00          | 20.00       | -     | -         | (Ribeiro et al., 2018)                        |
| s        | 8.60                     | 36.70          | -           | -     | 1.60      | (Lachheb et al., 2019)                        |
|          | 49.65                    | 6.80           | -           | -     | 0.98      | (Muñoz Velasco et al., 2016)                  |
|          | 52.42                    | 7.20           | -           | -     | 1.33      | (Muñoz Velasco et al., 2016)                  |
|          | 12.31                    | 33.44          | 24.52       | -     | 2.52      | (Buratti et al., 2018)                        |
|          | 23.00                    | 19.40          | 18.30       | -     | 2.20      | (Zarrinbakhsh et al., 2016)                   |
| Range    | 8.60-52.42               | 6.80-44.50     | 18.30-24.52 | -     | 0.98-2.52 |                                               |
| Corn     | 33.70-41.20              | 31.9-36.6      | 6.10-15.90  | -     | -         | (Cárdenas, 2020)                              |
| (Maize)  | 36.78                    | 38.81          | 3.30        | -     | 0.46      | (Viel et al., 2018)                           |
| cobs     | 38.80                    | 44.40          | 11.90       | -     | 2.88      | (Viel et al., 2018)                           |
|          | 36.78                    | 38.81          | 30.30       | -     | 0.46      | (Viel et al., 2019a) (after alkali treatment) |
|          | 48.10                    | 37.20          | 14.70       | -     | -         | (García et al., 2016)                         |
| Range    | 33.70-48.10              | 31.90-44.40    | 3.30-30.30  | -     | 0.46-2.88 |                                               |
| Corn     | 27.10                    | 13.87          | 0.64        | -     | 0.43      | (Viel et al., 2019a) (after alkali treatment) |
| cob      |                          |                |             |       |           |                                               |
| resi-    |                          |                |             |       |           |                                               |
| dues     |                          |                |             |       |           |                                               |
| Corn     | 35.00-39.60              | 16.80-35.00    | 7.00-18.40  | -     | -         | (Cárdenas, 2020)                              |
| (Maize)  |                          |                |             |       |           |                                               |
| stalks   |                          |                |             |       |           |                                               |
| Corn     | 47.40                    | 30.30          | 22.30       | -     | -         | (García et al., 2016)                         |
| stover   |                          |                |             |       |           |                                               |
| Cotton   | 71.60                    | 42.50          | 20.50       | -     | 5.54      | (Pirayesh and Khazaeian, 2012)                |
|          | 58.48                    | 14.38          | 21.45       | -     | -         | (Cárdenas, 2020)                              |
|          | 82.70-90.00              | 5.70           | <2          | 0.60  | -         | (Dittenber and GangaRao, 2012)                |
|          | 82.70                    | 5.70           | -           | -     | -         | (Jawaid and Abdul Khalil, 2011)               |
|          | 88.00                    | 5.70           | -           | 0.60  | -         | (Nguong et al., 2013)                         |
|          | 87.50                    | 17.10          | -           | -     | -         | (García et al., 2016)                         |



| Material | Chemical composition [%] |                |             |           |           | References                        |
|----------|--------------------------|----------------|-------------|-----------|-----------|-----------------------------------|
|          | Cellulose                | Hemicelluloses | Lignin      | Waxes     | Ash       |                                   |
| Range    | 58.48-90.00              | 5.70-42.50     | 2.00-21.45  | 0.60      | 5.54      |                                   |
| Cotton   | 66.20                    | 18.40          | 15.40       | -         | -         | (García et al., 2016)             |
| stalk    | 32.00-46.00              | 20.00-28.00    | 26.00       | -         | 8.16      | (Nguyen et al., 2017)             |
| Range    | 32.00-66.20              | 18.40-28.00    | 15.40-26.00 | -         | 8.16      |                                   |
| Flax     | 71.00                    | 18.60-20.60    | 2.20        | 1.50      | -         | (Faruk et al., 2012)              |
|          | 34.90                    | 23.60          | 22.30       | -         | -         | (Cárdenas, 2020)                  |
|          | 28.51                    | 15.80          | 18.14       | -         | 4.20      | (Viel et al., 2019a)              |
|          | 62.00-72.00              | 18.60-20.60    | 2.00-5.00   | 1.50-1.70 | -         | (Dittenber and GangaRao, 2012)    |
|          | 61.00                    | 27.00          | 8.00        | -         | -         | (Laborel-Préneron et al., 2016)   |
|          | 81.00                    | 14.00          | 3.00        | -         | -         | (Laborel-Préneron et al., 2016)   |
|          | 60.00                    | 16.00          | 3.00        | -         | -         | (Laborel-Préneron et al., 2016)   |
|          | 64.10                    | 16.70          | 2.00        | -         | -         | (Jawaid and Abdul Khalil, 2011)   |
|          | 71.00                    | 19.60          | 2.20        | 1.70      | -         | (Nguong et al., 2013)             |
|          | 75.90                    | 20.70          | 3.40        | -         | -         | (García et al., 2016)             |
|          | 62.00-72.00              | 18.60-20.60    | 2.00-5.00   | 1.50-1.70 | -         | (Papadopoulou et al., 2015)       |
| Flax     | 44.63                    | 24.41          | 20.98       | -         | 1.48      | (Viel et al., 2018)               |
| shiv     | 38.06                    | 25.03          | 31.20       | -         | 1.71      | (Viel et al., 2018)               |
|          | 53.00                    | 13.00          | 24.00       | -         | >2        | (Viel et al., 2018)               |
|          | 39.90                    | 26.80          | 24.20       | -         | -         | (García et al., 2016)             |
| Range    | 28.51-81.00              | 13.00-27.00    | 2.00-31.20  | 1.50-1.70 | 1.48-4.20 |                                   |
| Flax     | 28.51                    | 15.80          | 18.14       | -         | 4.20      | (Viel et al., 2018)               |
| fine     |                          |                |             |           |           |                                   |
| Flax     | 16.21                    | 5.83           | 7.74        | -         | 2.92      | (Viel et al., 2019a)              |
| fine     |                          |                |             |           |           |                                   |
| resi-    |                          |                |             |           |           |                                   |
| dues     |                          |                |             |           |           |                                   |
| Grapes   | 15.00                    | 11.30          | 30.30       | -         | 7.00      | (Pedras et al., 2020)             |
| (waste   | -                        | -              | -           | -         | 3.70      | (Muñoz et al., 2014)              |
| pom-     | 50.16                    | 6.62           | -           | -         | -         | (Barbieri et al., 2013)           |
| ace)     |                          |                |             |           |           |                                   |
| Range    | 15.00-50.16              | 6.62-11.30     | 30.30       | -         | 3.70-7.00 |                                   |
| Hazel-   | 55.10                    | 34.60          | 41.40       | -         | 8.23      | (Pirayesh and Khazaeian, 2012)    |
| nuts     | 55.10                    | 34.50          | 35.10       | -         | 8.22      | (Çöpür et al., 2007)              |
| Range    | 55.10                    | 34.50-34.60    | 35.10-41.40 | -         | 8.22-8.23 |                                   |
| Hazel-   | 34.00                    | 20.00          | 35.00-41.00 | -         | -         | (Salasinska and Ryszkowska, 2012) |
| nut      |                          |                |             |           |           |                                   |
| shells   | 34.50                    | -              | 35.10       | -         | 8.22      | (Çöpür et al., 2007)              |
|          | 22.90                    | 23.50          | -           | -         | 1.40      | (Licursi et al., 2017)            |

| Material   | Chemical composition [%] |                |             |       |           | References                      |
|------------|--------------------------|----------------|-------------|-------|-----------|---------------------------------|
|            | Cellulose                | Hemicelluloses | Lignin      | Waxes | Ash       |                                 |
| Range      | 34.60                    | -              | 41.40       | -     | -         | (Guler et al., 2008)            |
|            | 28.90                    | 11.30          | 30.20       | -     | 4.00      | (Lopes et al., 2012)            |
|            | 22.90-34.60              | 11.30-23.50    | 30.20-41.40 | -     | 1.40-8.22 |                                 |
| Hemp       | 74.40                    | 17.90          | 3.70        | -     | -         | (Jawaid and Abdul Khalil, 2011) |
|            | 68.00-74.40              | 15.00-22.40    | 3.70-10.00  | 0.80  | -         | (Dittenber and GangaRao, 2012)  |
|            | 67.00-78.30              | 5.50-16.10     | 2.90-3.30   | -     | -         | (Summerscales et al., 2010)     |
|            | 64.00                    | 16.00          | 4.00        | -     | -         | (Laborel-Préneron et al., 2016) |
|            | 67.00                    | 5.50           | 2.90        | -     | -         | (Shalwan and Yousif, 2013)      |
|            | 72.00                    | 20.10          | 4.70        | 0.80  | -         | (Nguong et al., 2013)           |
|            | 68.00-74.40              | 15.00-22.40    | 3.70-10.00  | 0.80  | -         | (Papadopoulou et al., 2015)     |
| Hemp shiv  | 49.97                    | 21.42          | 9.52        | -     | 0.67      | (Viel et al., 2018)             |
|            | 51.60                    | 21.50          | 12.90       | -     | 6.60      | (Viel et al., 2018)             |
|            | 47.50                    | 6.40           | 8.00        | -     | 8.80      | (Viel et al., 2018)             |
|            | 44.00                    | 18.00          | 28.00       | -     | 2.00      | (Viel et al., 2018)             |
| Hemp fines | 42.66                    | 18.87          | 11.52       | -     | 1.62      | (Viel et al., 2018)             |
| Range      | 44.00-78.30              | 5.50-22.40     | 2.90-28.00  | 0.80  | 0.67-8.80 |                                 |
| Jute       | 61.00-71.00              | 14.00-20.00    | 12.00-13.00 | 0.50  | -         | (Faruk et al., 2012)            |
|            | 64.40                    | 12.00          | 11.80       | -     | -         | (Jawaid and Abdul Khalil, 2011) |
|            | 59.00-71.50              | 13.60-20.40    | 11.80-13.00 | 0.50  | -         | (Dittenber and GangaRao, 2012)  |
|            | 72.00                    | 13.00          | 13.00       | -     | -         | (Laborel-Préneron et al., 2016) |
|            | 33.40                    | 22.70          | 28.00       | -     | -         | (Madurwar et al., 2013)         |
|            | 58.00-63.00              | 20.00-22.00    | -           | -     | 0.62      | (Phan et al., 2006)             |
|            | 66.00                    | 17.00          | 12.50       | 0.50  | -         | (Nguong et al., 2013)           |
| Range      | 33.40-72.00              | 12.00-22.70    | 11.80-28.00 | 0.50  | 0.62      |                                 |
| Kenaf      | 72.00                    | 20.30          | 9.00        | -     | -         | (Faruk et al., 2012)            |
|            | 31.00-72.00              | 20.30-21.50    | 8.00-19.00  | -     | -         | (Dittenber and GangaRao, 2012)  |
|            | 70.00                    | 3.00           | 19.00       | -     | -         | (Laborel-Préneron et al., 2016) |
|            | 53.40                    | 33.90          | 21.20       | -     | 4.00      | (Jawaid and Abdul Khalil, 2011) |
|            | 51.00                    | 21.50          | 10.50       | -     | -         | (Nguong et al., 2013)           |
|            | 31.00-72.00              | 20.30-21.50    | 8.00-19.00  | -     | -         | (Papadopoulou et al., 2015)     |
| Range      | 31.00-72.00              | 3.00-33.90     | 8.00-21.20  | -     | 4.00      |                                 |

| Material           | Chemical composition [%] |                |             |             |           | References                       |
|--------------------|--------------------------|----------------|-------------|-------------|-----------|----------------------------------|
|                    | Cellulose                | Hemicelluloses | Lignin      | Waxes       | Ash       |                                  |
| Palm oil           | 60.00-65.00              | -              | 11.00-29.00 | -           | -         | (Dittenber and GangaRao, 2012)   |
|                    | 49.00                    | 21.00          | 23.00       | -           | -         | (Laborel-Préneron et al., 2016)  |
|                    | 65.00                    | -              | 19.00       | -           | 2.00      | (Jawaid and Abdul Khalil, 2011)  |
| Range              | 49.00-65.00              | 21.00          | 11.00-29.00 | -           | 2.00      |                                  |
| Palm oil frond     | 56.03                    | 27.51          | 20.48       | -           | 2.40      | (Jawaid and Abdul Khalil, 2011)  |
| Olive husks        | 25.00                    | 24.60          | 50.40       | -           | -         | (García et al., 2016)            |
|                    | -                        | -              | -           | -           | 14.21     | (Christoforou et al., 2017)      |
| Olive whole stones | 31.90                    | 21.90          | 26.50       | -           | -         | (Fernández-Bolaños et al., 1999) |
| Olive seed husks   | 36.40                    | 26.80          | 26.00       | -           | -         | (Fernández-Bolaños et al., 1999) |
| Olive pomace       | -                        | -              | -           | -           | 3.20-8.63 | Christoforou et al., 2017)       |
| Pine-apples        | 81.00                    | -              | 12.70       | -           | -         | (Idicula et al., 2006)           |
| Ramie              | 68.60-76.20              | 13.00-16.00    | 0.60-0.70   | 0.30        | -         | (Faruk et al., 2012)             |
|                    | 68.60-85.00              | 13.00-16.70    | 0.50-0.70   | 0.30        | -         | (Dittenber and GangaRao, 2012)   |
|                    | 68.60                    | 13.10          | 0.60        | -           | -         | (Jawaid and Abdul Khalil, 2011)  |
| Range              | 68.60-85.00              | 13.00-16.70    | 0.50-0.70   | 0.30        | -         |                                  |
| Rice husk          | 35.00-45.00              | 19.00-25.00    | 20.00       | 14.00-17.00 | -         | (Faruk et al., 2012)             |
|                    | 30.00                    | 20.00          | 33.00       | -           | 14.00     | (Brás et al., 2019)              |
| Range              | 30.00-45.00              | 19.00-25.00    | 20.00-33.00 | 14.00-17.00 | 14.00     |                                  |
| Rice straw         | 41.00-57.00              | 33.00          | 8.00-19.00  | 8.00-38.00  | -         | (Faruk et al., 2012)             |
|                    | 36.20-47.00              | 19.00-24.50    | 9.90-24.00  | -           | -         | (Cárdenas, 2020)                 |
|                    | 52.30                    | 32.80          | 14.90       | -           | -         | (García et al., 2016)            |
| Range              | 36.20-57.00              | 19.00-33.00    | 8.00-24.00  | 8.00-38.00  | -         |                                  |
| Sisal              | 65.00                    | 12.00          | 9.90        | 2.00        | -         | (Faruk et al., 2012)             |
|                    | 65.80                    | 12.00          | 9.90        | -           | -         | (Jawaid and Abdul Khalil, 2011)  |
|                    | 65.00                    | 12.00          | 9.90        | -           | -         | (Idicula et al., 2006)           |
|                    | 60.00-78.00              | 10.00-14.20    | 8.00-14.00  | 2.00        | -         | (Dittenber and GangaRao, 2012)   |

| Material | Chemical composition [%] |                |             |       |           | References                      |
|----------|--------------------------|----------------|-------------|-------|-----------|---------------------------------|
|          | Cellulose                | Hemicelluloses | Lignin      | Waxes | Ash       |                                 |
|          | 65.80                    | 12.00          | 9.90        | -     | -         | (Laborel-Préneron et al., 2016) |
|          | 73.00                    | 13.00          | 11.00       | -     | -         | (Laborel-Préneron et al., 2016) |
|          | 38.20                    | 26.00          | 26.00       | -     | -         | (Madurwar et al., 2013)         |
|          | 73.00                    | 12.00          | 12.00       | 2.00  | -         | (Nguong et al., 2013)           |
| Range    | 38.20-78.00              | 10.00-26.00    | 8.00-26.00  | 2.00  | -         |                                 |
| Sugar    | 55.20                    | 16.80          | 25.30       | -     | -         | (Faruk et al., 2012)            |
| cane     | 32.00-55.20              | 16.80          | 19.00-25.30 | -     | -         | (Dittenber and GangaRao, 2012)  |
| (ba-     | 40.00-41.30              | 27.00-37.50    | 10.00-20.00 | -     | -         | (Cárdenas, 2020)                |
| gasse)   | 47.40                    | 29.10          | 23.50       | -     | -         | (García et al., 2016)           |
| Range    | 32.00-55.20              | 16.80-37.50    | 10.00-25.30 | -     | -         |                                 |
| Sun-     | 24.10                    | 28.60          | 29.40       | -     | -         | (Cárdenas, 2020)                |
| flower   |                          |                |             |       |           |                                 |
| seed     |                          |                |             |       |           |                                 |
| Sun-     | 42.10                    | 29.66          | 13.44       | -     | -         | (Cárdenas, 2020)                |
| flower   |                          |                |             |       |           |                                 |
| stalks   |                          |                |             |       |           |                                 |
| Sun-     | 23.93                    | 7.83           | 9.13        | -     | -         | (Evon et al., 2012)             |
| flower   |                          |                |             |       |           |                                 |
| whole    |                          |                |             |       |           |                                 |
| plant    |                          |                |             |       |           |                                 |
| Sun-     | 66.20                    | 18.40          | 15.40       | -     | -         | (García et al., 2016)           |
| flower   |                          |                |             |       |           |                                 |
| shells   |                          |                |             |       |           |                                 |
| Tea      | 33.30                    | 23.20          | 43.50       | -     | -         | (García et al., 2016)           |
| residue  |                          |                |             |       |           |                                 |
| Vine     | 41.14                    | 26.00          | 20.27       | -     | -         | (Cárdenas, 2020)                |
| shoots   |                          |                |             |       |           |                                 |
| Wheat    | 38.00-45.00              | 15.00-31.00    | 12.00-20.00 | -     | -         | (Faruk et al., 2012)            |
| straw    | 32.90-50.00              | 24.00-35.50    | 8.90-17.30  | -     | -         | (Cárdenas, 2020)                |
|          | 43.04                    | 29.66          | 5.24        | -     | 0.82      | (Viel et al., 2018)             |
|          | 38.60                    | 32.60          | 14.10       | -     | 5.90      | (Viel et al., 2018)             |
|          | 45.00                    | 28.00          | 18.00       | -     |           | (Karade, 2010)                  |
| Range    | 32.90-50.00              | 15.00-35.50    | 5.24-20.00  | -     | 0.82-5.90 |                                 |





DEVELOPMENT OF BIO-BASED PARTICLEBOARDS FOR  
BUILDINGS' INDOOR USE

