



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 and 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.

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., 2019, 2023b). 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).

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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 Alabdulkareem (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; Vandenbosch 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. Liuzzi 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). Fig. 1 reported the hazelnut world production trend between 2016 and 2021, considering only the first four world producers, according to FAO (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; Vrtošević 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 1 reports some information about the chemical and physical properties of hazelnut shells, according to past studies.

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

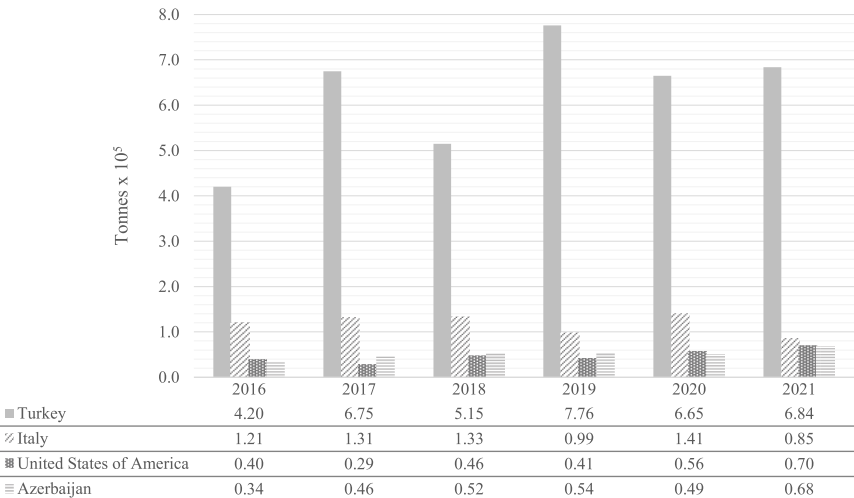


Fig. 1. Hazelnut's world production trend between 2016 and 2021 (FAO, 2021).

Table 1
Chemical and physical properties of hazelnut shells.

Chemical composition [%]				Physical Properties		Reference
Cellulose	Hemicelluloses	Lignin	Ash	Loose bulk density [kg/m ³]	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)

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.

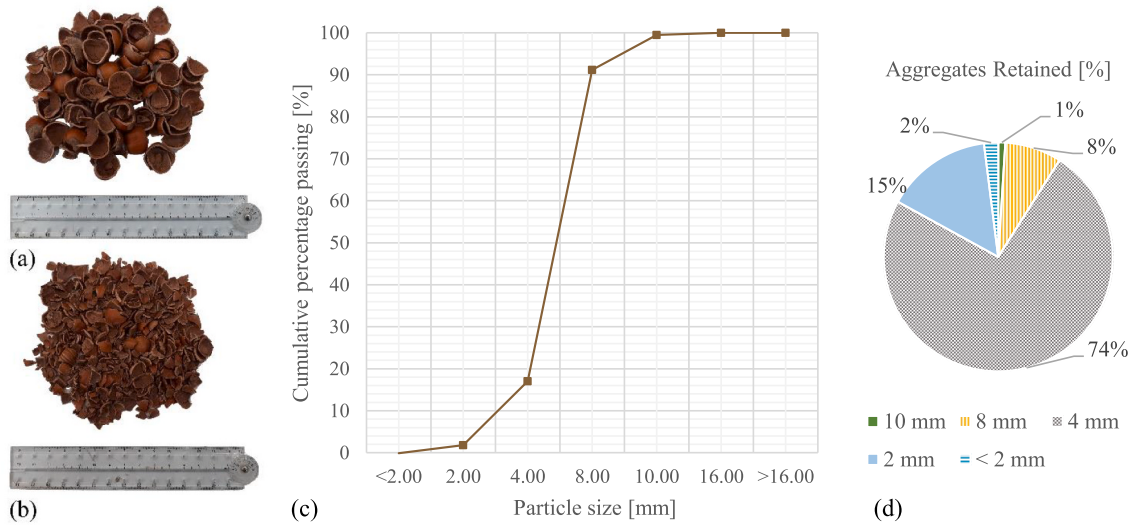


Fig. 2. Samples of hazelnut shells (a) before shredding, (b) after shredding, used to carry out particle analysis, (c) particle size distribution, adapted from (Cintura et al., 2023b), and (d) percentages of aggregates retained in each sieve (considering the average values of three samples).

2. Materials and methods

2.1. Materials

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 (Fig. 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 (Fig. 2b), as reported in a previous study (Cintura et al., 2023b). Fig. 2c and d shows 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 Fig. 2c and d, 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 $(0.099 \pm 0.006)\text{ W/(m.K)}$.

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 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 1).

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

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 min. The fresh composite was placed in quadrangular silicon ($10\text{ cm} \times 10\text{ cm} \times 4\text{ cm}$), cylindrical silicon (diameter = 10 cm,

high = 4 cm), and quadrangular wooden home-made ($50\text{ cm} \times 50\text{ cm} \times 4\text{ cm}$) 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 min. After that, they were dried at laboratory conditions, rotated every 30 min (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). Fig. 3 shows the samples produced by using silicon moulds.

2.3. Test methods

Table 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.

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.

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 $10\text{ cm} \times 10\text{ cm} \times 4\text{ cm}$ 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 (Fig. 4a).

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,

Table 2

Characteristic of the sodium silicate solution provided by Ingessil Srl (Ingessil, 2022).

Property	Value
Weight ratio [-]	2.40
Density [$^{\circ}\text{Bé}$]	46.45
Molar ratio [-]	2.48
Sodium silicate concentration [% p/p]	41.33
SiO_2 [% p/p]	29.17
Na_2O [% p/p]	12.16
Density [g/ml] at $T = 20\text{ }^{\circ}\text{C}$	1.471
pH ($T = 20\text{ }^{\circ}\text{C}$)	12.40

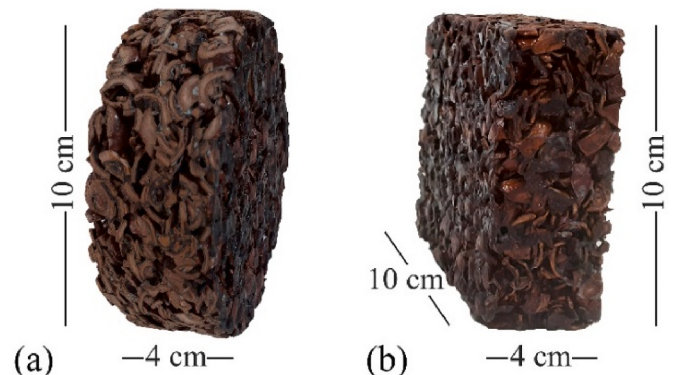


Fig. 3. Samples after casting and curing, to evaluate: (a) acoustic properties (diameter = 10 cm; high = 4 cm); (b) hygroscopicity ($10\text{ cm} \times 10\text{ cm} \times 4\text{ cm}$).

Table 3

Laboratory tests, designation, number and dimensions of composite samples, and references.

Properties	Samples			References
	Designation ^a	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 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)

^a 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.

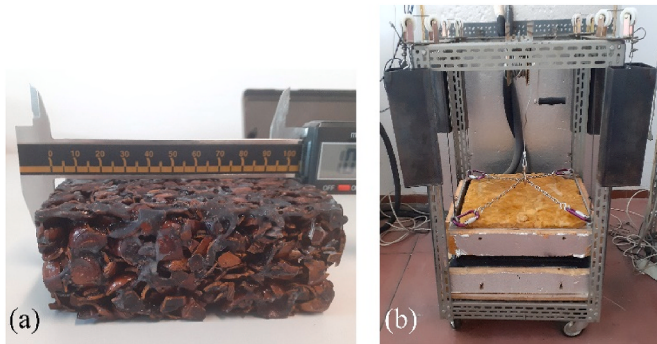


Fig. 4. (a) Sample 10 cm × 10 cm × 4 cm and the Fervi Digital Caliber used to determine the dimensions for apparent density; (b) equipment used to evaluate the thermal conductivity by considering the heat flow method.

2005; Baldinelli et al., 2019; La Gennusa et al., 2021; Platt et al., 2023). Fig. 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 × 50 cm × 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).

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 × 10 cm × 4 cm (H1, H2, H3). A moisture barrier was provided by using an aluminium foil (Fig. 5a) that allowed the exposure of only one adsorption/desorption face (area = 0.1 m²), as reported in Fig. 5b. The samples were preconditioned in a climatic chamber (Climatest ARGO-Lab CH 250), shown in Fig. 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).

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 (1)–(3)

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

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

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

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 [mm²] is the exposed surface.

MBV was calculated and rated considering an adaptation of Rode et al. (2005). It was considered as the average value between the MBV of the sorption (MBV_a) and the desorption phase (MBV_d), calculated as reported in Equations (4) and (5), respectively.

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

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

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

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, α , 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 (Fig. 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 (Fig. 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

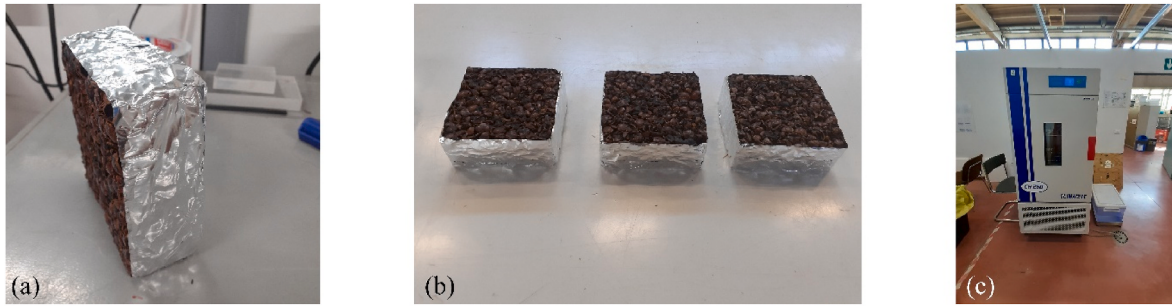


Fig. 5. (a) Detail of the aluminium foil used to provide a moisture barrier and expose only one surface of the sample to relative humidity variations; (b) three samples (10 cm × 10 cm × 4 cm) H1, H2, and H3, covered by the aluminium foil and considered to evaluate the hygroscopic properties; (c) climatic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

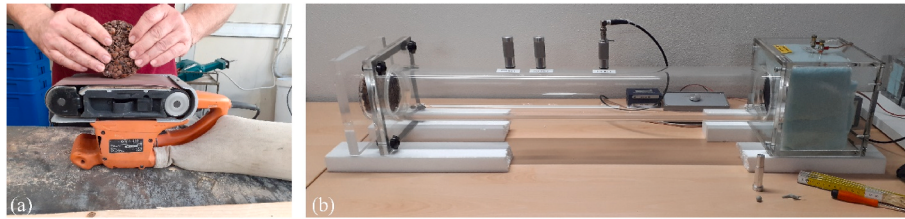


Fig. 6. (a) Smoothing process of a cylindrical sample to fit into the end of the impedance tube; (b) impedance tube of plexiglass (diameter = 10 cm) with the loudspeaker and the sample at the extremities.

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).

2.3.5. Flexural strength

The two samples 50 cm × 50 cm × 4 cm employed for the thermal analysis (H8, and H9) were cut in strips each 10 cm, and the eight obtained samples 50 cm × 10 cm × 4 cm were conditioned at $T = (20 \pm 1) ^\circ\text{C}$ and $\text{RH} = (60 \pm 5) \%$. The considered designation, reported in Table 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, 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 Fig. 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.

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

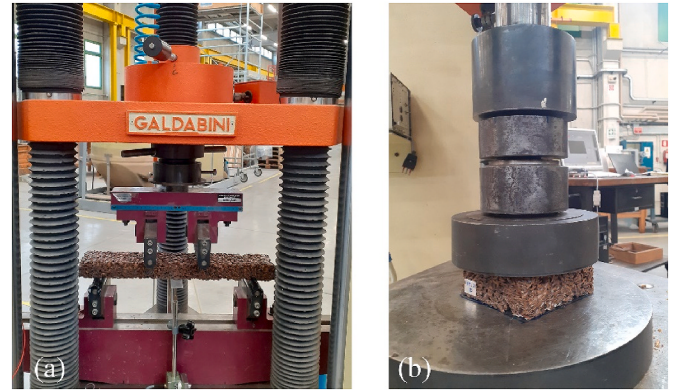


Fig. 7. Galdabini universal machine employed to evaluate the mechanical properties: (a) sample 10 cm × 50 cm × 4 cm used to analyse the flexural strength; (b) sample 10 cm × 10 cm × 4 cm used to evaluate the compressive strength.

considering Equations (6) and (7):

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

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

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 δ [mm] is the displacement. The flexural strength σ_{max} [N/mm²] of each sample was calculated as the one corresponding to the maximum applied force, F_{max} [N], as described in Equation (8); then the average value and standard deviation were considered.

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

The modulus of elasticity was determined considering Equation (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 ϵ ($\epsilon_{60\%}$ and $\epsilon_{20\%}$) (UNI 11842:2021, 2021).

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

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

2.3.6. Compressive strength

Compressive strength was evaluated according to an adaptation of EN 826 (EN 826, 2013). Samples 10 cm × 10 cm × 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 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 (Fig. 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 (10) and (11).

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

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

where F [N] is the applied force (recorded by the equipment), A_0 [mm²] is the initial cross-sectional area of the samples, δ [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/mm²] 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 (9), namely σ and ϵ at 20% and 60% of the compressive test breaking load, and then the average value and the standard deviation were determined.

3. Results

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.

3.2. Apparent density and thermal properties

Table 4 reports the results of the apparent density at $T = 50^\circ\text{C}$, the thermal conductivity at $T = 20^\circ\text{C}$, and $\text{RH} = 60\%$ for each sample, the average value, and the standard deviation (S.D.).

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,

Table 4

Results of apparent density at $T = 50^\circ\text{C}$, thermal conductivity at $T = 20^\circ\text{C}$, $\text{RH} = 60\%$ for each tested sample, average value, and standard deviation (SD).

Composite	Apparent density [kg/m ³]	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 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/m³ (Celenit,); an OSB panel, a density of 650 kg/m³ (Federation, 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/m³ and 1022 kg/m³, 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/m³ (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/(m.K) (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. (2019) reported thermal conductivity values between 0.074 W/(m.K) and 0.108 W/(m.K) for wood-lime boards; 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. (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 $T = 17^\circ\text{C}$ 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 ±

0.03) 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.

3.3. Hygroscopicity and moisture buffering value

Fig. 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 kg/m² and 0.066 kg/m². 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. (2005), and carry out an easier comparison with past research, the MBV was calculated (as described in Section 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 RH = 75% and 16 h-desorption phase at 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.

The MBV (considering the average value of the three tested samples and the standard deviation) was (3.45 ± 1.76) g/(m² %RH). 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 g/(m² %RH) for straw-based composites and 3.29 g/(m² %RH) for olive fibre-based ones. Considering the classification provided by 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 probability 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. Fig. 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.

Fig. 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.

3.4. Acoustic properties

Fig. 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 2.3).

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).

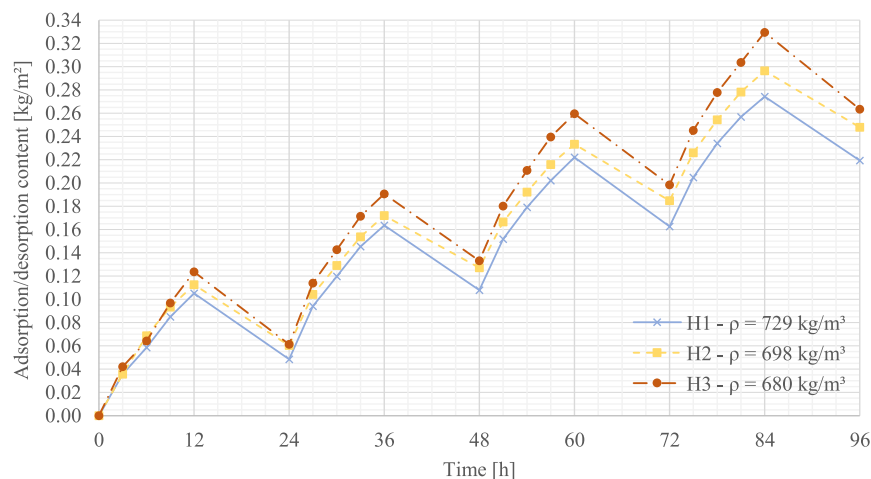


Fig. 8. Moisture adsorption/desorption for four cycles of the three tested composite samples.

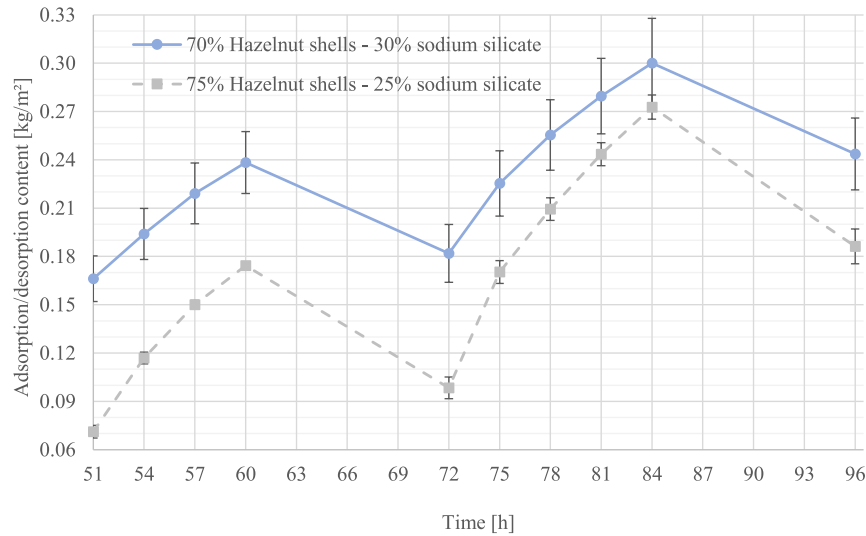


Fig. 9. Moisture adsorption/desorption content for two adsorption/desorption cycles: comparison of the results (average value of three samples) for the composite evaluated in the present study and a composite made up of 75% of hazelnut shells and 25% of sodium silicate solution by the total volume (Cintura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

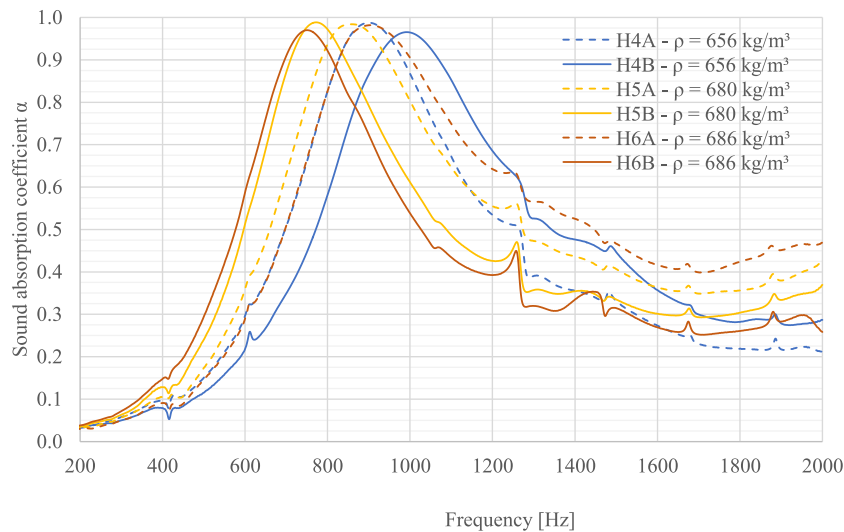


Fig. 10. Sound absorption coefficients of both sides of the three tested samples. Continuous lines represent the results for the flatter samples' exposed surfaces (lower part of the mould), namely the samples designated by letter B (H4B, H5B, H6B); the dotted lines for the ones rougher (upper part of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

Granular materials may be viewed as a special case of porous materials, and, as Fig. 10 shows, they show a first peak of normal incidence sound absorption, f_0 , described by Equation (12):

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

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).

3.5. Flexural strength

Fig. 11 shows the stress-strain curve for the eight tested samples. The

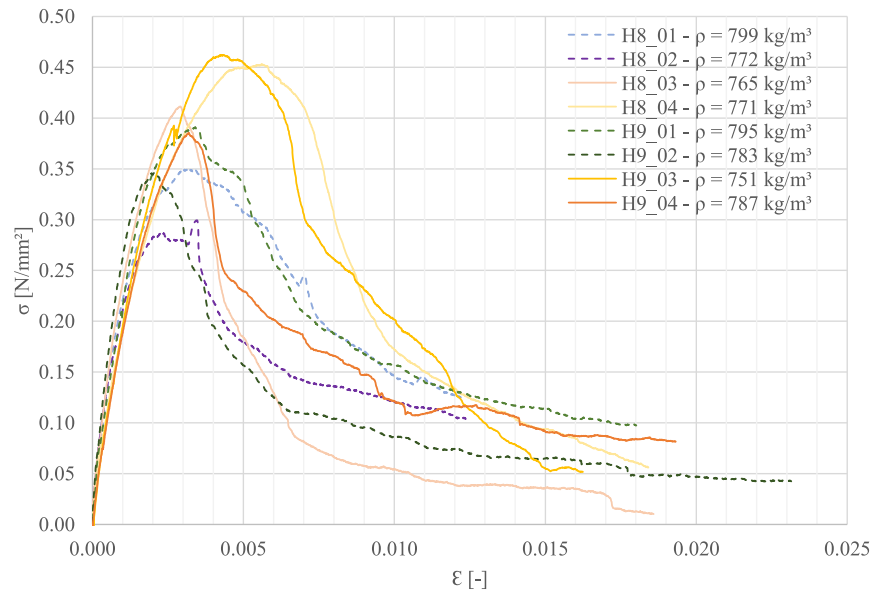


Fig. 11. Stress-strain curves of the eight tested samples. The dotted lines represent the samples with the upper side as the flatter one, and the lower side as the rougher (Samples_LOW); the continuous lines represent the samples with the upper side as the rougher one, and the lower side as the flatter one (Samples_UP).

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).

Table 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 (9).

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. (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 Fig. 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/mm² and 0.074 N/mm².

Table 5

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

	$F_{\max}$ [N]	$\delta_{\max}$ [mm]	$\sigma_{\max}$ [N/mm ²]	$E_{(20\%-60\%)}$ [N/mm ²]
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

Antunes et al. (2019) for rice husk-earth-lime-gypsum panels reported values of flexural strength between 0.08 and 0.12 N/mm². 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.

3.6. Compressive strength

Figs. 12 and 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).

Table 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 (9).

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 (Fig. 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/mm² and 0.40 N/mm² 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 and 15.00 N/mm² for the corn starch-based binder, 6.60–7.00 N/mm² for the lime, and 6.20–8.50 N/mm² 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

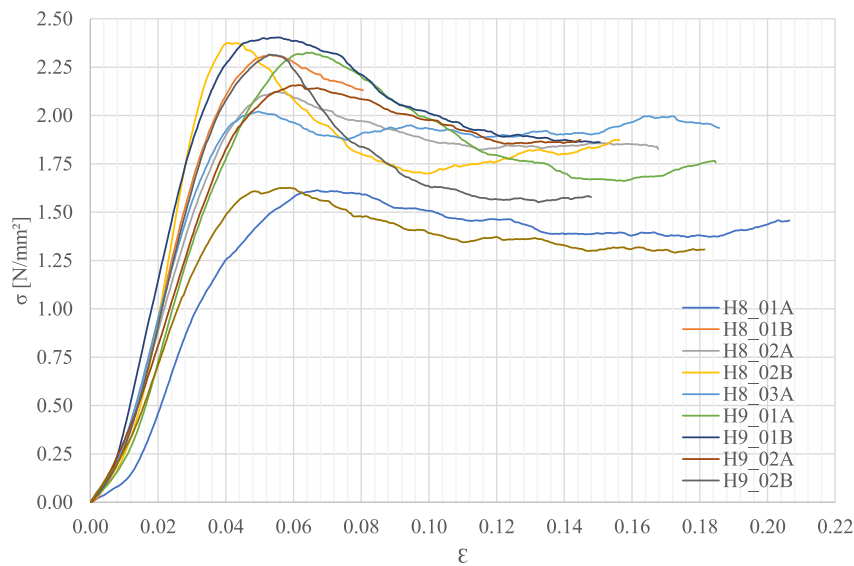


Fig. 12. The stress-strain compressive curve of the ten tested samples 10 cm × 10 cm × 4 cm.



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

Table 6

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

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

the reference standards (EN 826, 2013): $\sigma \geq 0.2$ N/mm² (for a thickness of 15–40 mm) and $\sigma \geq 0.15$ N/mm² (for a thickness of 50–75 mm). The same behaviour was shown by expanded cork boards (ICB), analysed by Simões et al. (2019). They considered both standard and medium density, namely between 90 and 110 kg/m³ and 140–160 kg/m³, respectively. The reported compressive strength for a thickness of 50 mm was 0.154 N/mm² (standard density) and 0.205 N/mm² (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. Discussion

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 ± 1.76) g/(m² %RH), and a peak of sound absorption between 700 and 900 Hz suggested that the proposed composite could improve indoor comfort if applied as an internal coating board.

4.2. Comparison with other studies about high-mass panels and boards

Table 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 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

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

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

Note: ^a 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. ¹ Percentages in volume of earth; ² Only one type of sample of the considered study was reported (the most significant for the comparison with the present study); ^a Apparent density; ^b True density; ^c Calculated by considering ISO 24353 (ISO 24353, 2008); ^d MBV_(deh); ^e Calculated by considering three-point bending test.

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).

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 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. 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 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 3.5 and 3.6, in line with other examples of unconventional, non-structural boards (Table 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.

4.4. 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 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 colonization 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 and 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.

4.5. 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 et al., 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).

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.

CRediT authorship contribution statement

Eleonora Cintura: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft. **Paulina Faria:** Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review & editing. **Luisa Molari:** Conceptualization, Investigation, Writing – review & editing. **Luca Barbaresi:** Conceptualization, Investigation, Writing – review & editing. **Dario D’Orazio:** Conceptualization, Investigation, Writing – review & editing. **Lina Nunes:** Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Eleonora Cintura reports financial support was provided by Foundation for Science and Technology.

Data availability

Data will be made available on request.

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