Construction and Building Materials 404 (2023) 132665

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Eco-efficient boards with agro-industrial wastes – Assessment of different adhesives

Eleonora Cintura [a,b,*], Paulina Faria [a], Marta Duarte [b], Lina Nunes [b,c]

[a] CERIS, Department of Civil Engineering, NOVA School of Science and Technology, NOVA University Lisbon, 2829-516 Caparica, Portugal
[b] Structures Department, National Laboratory for Civil Engineering, Avenida do Brasil 101, 1700-066 Lisbon, Portugal
[c] CE3C, Centre for Ecology, Evolution and Environmental Changes & CHANGE, Global Change and Sustainability Institute, University of the Azores, 9700-042 Angra do Heroísmo, Portugal

ARTICLE INFO

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

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.

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^3, they obtained values of thermal conductivity of 0.077 W/(m.K) for both; for a density of 350 kg/m^3, 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 fibreboard 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

* Corresponding author at: CERIS, Department of Civil Engineering, NOVA School of Science and Technology, NOVA University Lisbon, 2829-516 Caparica, Portugal.
E-mail addresses: e.cintura@fct.unl.pt (E. Cintura), paulina.faria@fct.unl.pt (P. Faria), mduarte@lnec.pt (M. Duarte), linanunes@lnec.pt (L. Nunes).

https://doi.org/10.1016/j.conbuildmat.2023.132665
Received 6 February 2023; Received in revised form 25 July 2023; Accepted 28 July 2023
Available online 6 September 2023

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

Board/Panel	Density [kg/m^3]	Thermal conductivity [W/(m.K)]	Ref.
Commercial products			
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 (gypsum plasterboard)	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]
Past studies (literature review)			
Binderless compacted particleboards made up of flax shives (*Linum usitatissimum* L.)	350–500	0.066–0.077	[18]
Binderless compacted particleboards made up of sunflower stems (*Helianthus annuus* L.)	350–500	0.070–0.077	[18]
Internal layer of a sandwich panel (medium density fibreboard filled with a bio-based insulation core) made up of short-cut rapeseed	–	0.048	[19]
Internal layer of a sandwich panel (medium density fibreboard 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 ($Na_2O \bullet_n (SiO_2)$).	152	0.058	[38]
Composites made up of olives fibres (OF) bounded by using a sodium silicate solution ($Na_2O \bullet_n (SiO_2)$).	235	0.062	[38]
Gypsum plasterboard	900	0.25	[39]

Note: *Maximum value; **Minimum value.

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 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.

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 are 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 2, considering only Euro-Mediterranean countries, the selected area for the present work [48].

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];

Table 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 [Tonnes × 10^7]	First producer	Starch supply utilization account [Tonnes × 10^6]
Maize	3.06	France	2.43
Potatoes	1.39	France	0.09
Rice	0.27	Italy	0.03
Wheat	4.76	France	1.31

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

Material	Loose bulk density [kg/m^3] (T = 20 °C, RH = 60 %)	Thermal Conductivity [W/(m.K)] (T = 23 °C, RH = 50 %)
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

- 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.

2. Materials and methods

2.1. Materials

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 reports information about loose bulk density, and thermal conductivity; Fig. 1 shows the particle size distribution of the bio-aggregates, considering past research [61].

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].

The second studied adhesive (used for composites B) was a sodium silicate solution ($Na_2O{\cdot}n\ SiO_2{\cdot}nH_2O$) produced by Labkem, Spain, and analysed by Ingessil S.r.l., Verona, Italy. It was selected considering the

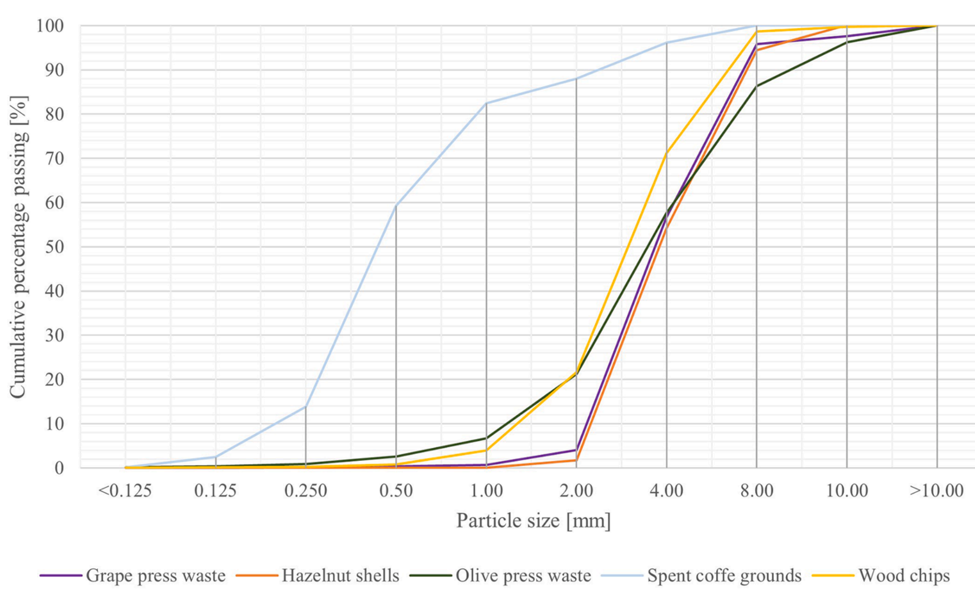

Fig. 1. Particle size distribution of the tested bio-wastes and the wood chips adapted from Cintura et al. [61].

Table 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 (1 h) [%]	0.2	
	Bending strength (dry) [kg/cm^2]	≈ 52	
Sodium silicate solution (composites B)	Mass fraction SiO_2/Na_2O	2.4–2.6	Labkem technical sheet [72]
	$SiO_2 + Na_2O$ [%]	≥ 39	
	Density at 20 °C [g/cm^3]	1.45–1.48	
	Fe_2O_3 [%]	≤ 0.01	
	CaO [%]	≤ 0.1	
	Viscosity (20 °C) [cP]	≥ 100	
	Molar ratio [-]	2.48	Information provided by Ingessil [73]
	Na_2SiO_3 title [% p/p]	41.33	
	SiO_2 [% p/p]	29.17	
	Na_2O [% 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	

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 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 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].

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-

Table 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.

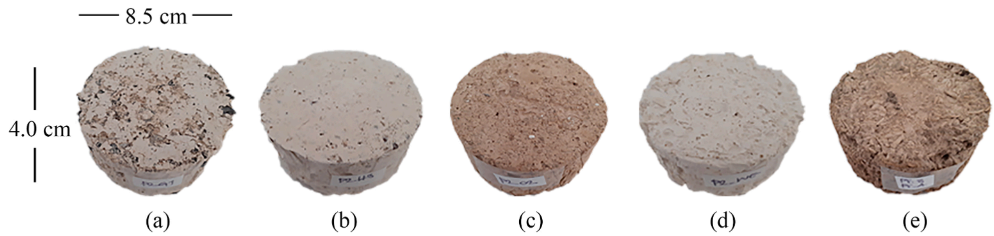

Fig. 2. Samples A – hemi-hydrated gypsum, air lime and citric acid: (a) grape press waste (AG); (b) hazelnut shells (AH); (c) olive press waste (AO); (d) wood chips (AW); (e) wood chips and spent coffee grounds (AWc).

Fig. 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).

Fig. 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).

aggregates by using the maximum amount of bio-wastes and the minimum amount of adhesives. Table 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 min, 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, demoulded and then placed at T = 60 °C until the variation in mass was lower than 0.5 %. Figs. 2, 3 and 4 show samples A, B and C, respectively, after casting and curing, reporting their designations.

2.3. Methods

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 (Fig. 5a).

2.3.2. Apparent density and thermal properties

The apparent density was calculated after curing and stabilizing the samples at T = 60 °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 ± 1) °C and RH = (65 ± 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 (Fig. 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.

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

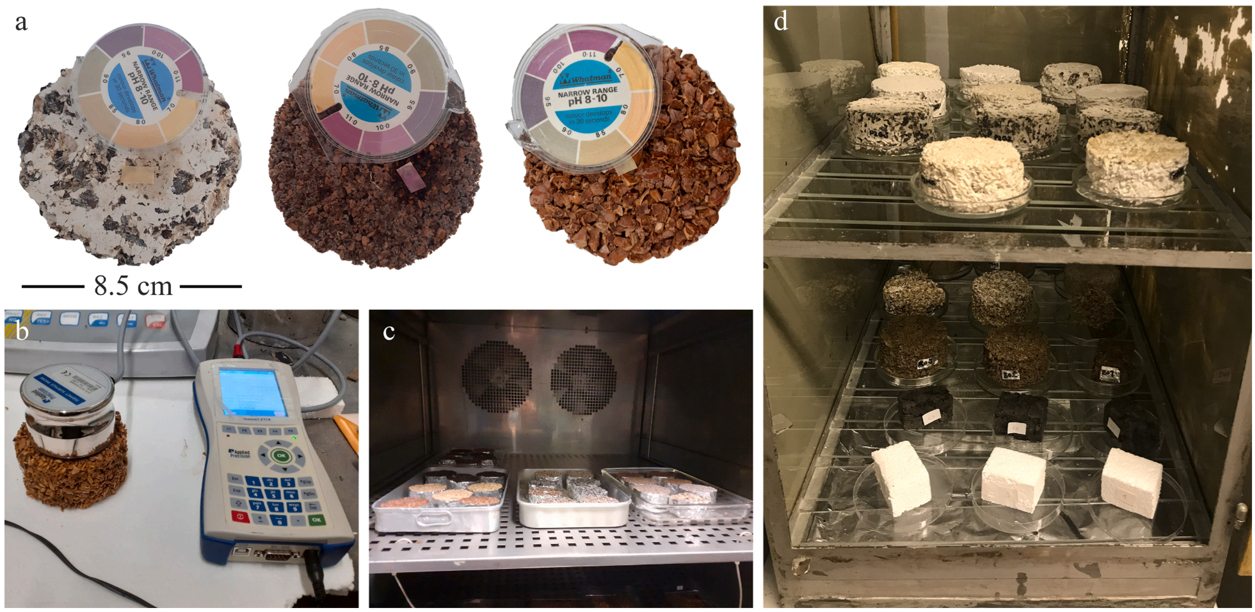

Fig. 5. Main tests of the experimental campaign: (a) analysis of the pH by using the Litmus paper; (b) evaluation of the thermal conductivity by using the ISOMET; (c) samples in the climate chamber to assess the hygroscopicity; (d) samples in the climate chamber to assess the biological susceptibility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

Table 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

= 0.0057 m^2) was exposed (Fig. 5c). The samples were preconditioned in a climatic chamber (Aralab Fitoclima 1000) at T = (23 ± 2) °C and RH = (63 ± 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 Eqs. (1), (2) and (3) to determine the variation of moisture adsorbed/desorbed content over time.

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

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

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

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^2] 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_a) and the desorption phase (MBV_d), calculated as reported in Eqs. (4) and (5), respectively. The last three cycles for each sample and their average values were considered (to evaluate the most stabilized conditions).

$$MBV_a\left(\frac{g}{m^2\,\%\,RH}\right) = \frac{m_{an} - m_{d(n-1)}}{A\ x\ (RH_{high} - RH_{low})} \tag{4}$$

$$MBV_d\left(\frac{g}{m^2\,\%\,RH}\right) = \frac{m_{an} - m_{dn}}{A\ x\ (RH_{high} - RH_{low})} \tag{5}$$

RH_{high} is the highest value and RH_{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].

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 ± 2) °C and RH ≥ 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

Table 7
Ph average values and standard deviation of the composites.

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

ventilation was prevented, and the samples were kept spaced as shown in Fig. 5d. Three samples of an expanded cork agglomerate (ICB) and three of an expanded polystyrene (EPS) panel (5 cm × 3 cm × 3 cm), 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 6 reports the ratings of the top surface of the sample obtained either visually or by using a stereo microscope (Olympus SZX12).

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 (H0 = 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. Results and discussion

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 7 reports the average pH and the standard deviation of the specimens.

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 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 ≥ 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.

3.2. *Apparent density and thermal conductivity*

Table 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 1; the values are in line with the literature ones. Fig. 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 8
Average apparent density and thermal conductivity with standard deviation of the composites.

Composites	Apparent density T = 60 °C [kg/m^3]	Thermal conductivity T = 17 °C RH = 65 % [W/(m.K)]
Composites A with gypsum-based binder		
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
Composites B with sodium silicate solution		
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
Composites C with natural glue		
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) %.

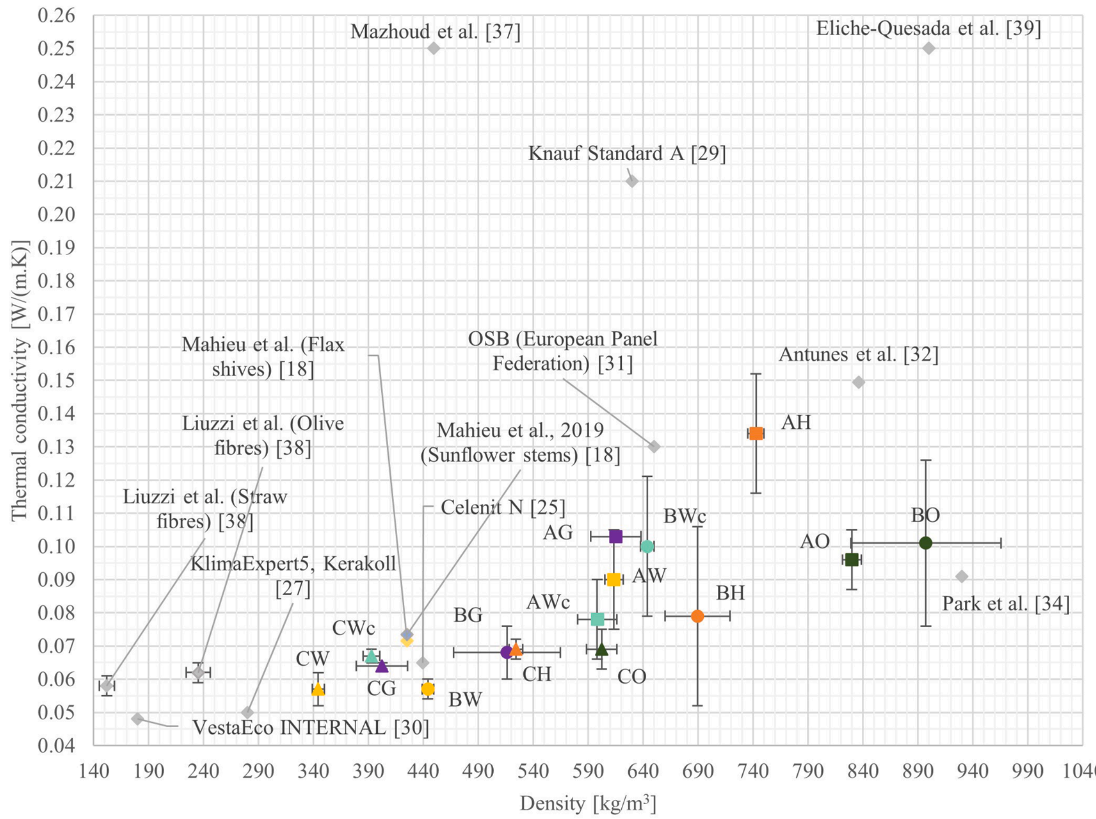

Fig. 6. Correlation between thermal conductivity and density of the studied composites, compared with some literature and commercial values. (See above-mentioned references for further information.)

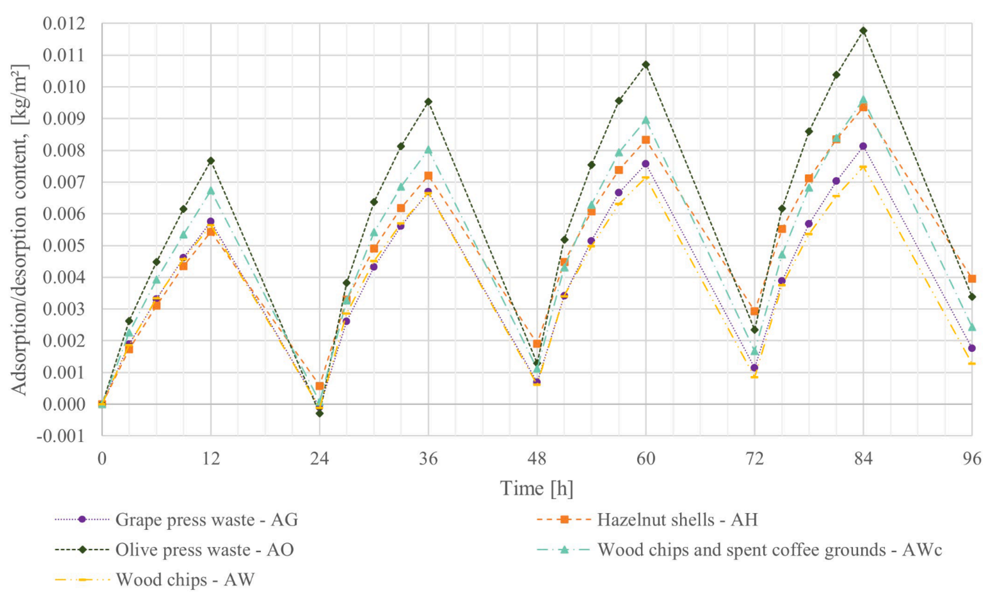

Fig. 7. Moisture variation for four adsorption/desorption cycles of composites A (with gypsum-based binder).

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

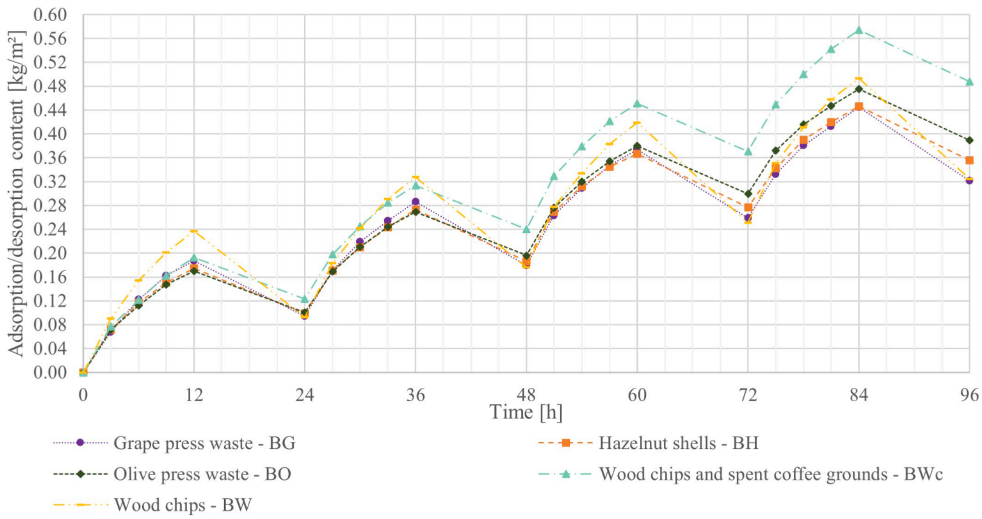

Fig. 8. Moisture variation for four adsorption/desorption cycles of composites B (with sodium silicate solution).

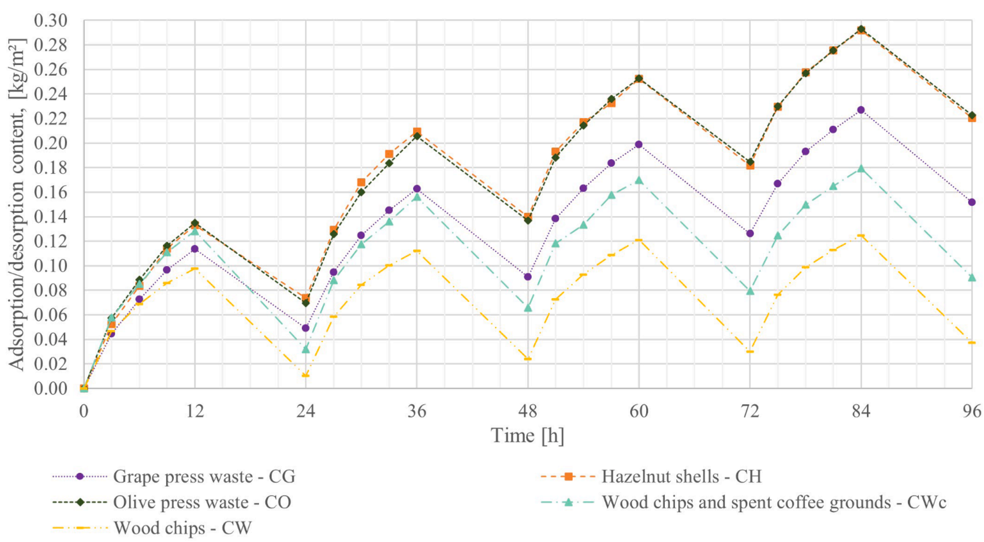

Fig. 9. Moisture variation for four adsorption/desorption cycles of composites C (with starch-based natural glue).

gypsum mortar with palm fibre. They reported values of thermal conductivity between 0.29 and 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 and 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.

3.3. Hygroscopicity and moisture buffering value

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

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^2 and 0.010 kg/m^2; $\rho_{A,dc}$ of 0.008 kg/m^2 for all cycles); hazelnut shells achieved the lowest ones ($\rho_{A,ac}$ between 0.005 kg/m^2 and 0.007 kg/m^2; $\rho_{A,dc}$ of 0.005 kg/m^2 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^2 and 0.007 kg/m^2 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^2 and 0.008 kg/m^2; $\rho_{A,dc} = 0.007$ kg/m^2 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^2 and 0.243 kg/m^2; $\rho_{A,dc}$ between 0.144 kg/m^2 and 0.169 kg/m^2) and grape press waste ($\rho_{A,ac}$ between 0.187 kg/m^2 and 0.195 kg/m^2; $\rho_{A,dc}$ between 0.093 kg/m^2 and 0.123

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

Composites	MBV [g/(m^2.%RH)]
Composites A with gypsum-based binder	
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
Composites B with sodium silicate solution	
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
Composites C with natural glue	
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

kg/m^2). Also in this case, composites made up of hazelnut shells reached the lowest values ($\rho_{A,ac}$ between 0.170 kg/m^2 and 0.181 kg/m^2; $\rho_{A,dc}$ between 0.076 kg/m^2 and 0.091 kg/m^2), similar to ones made up of olives press waste ($\rho_{A,ac}$ between 0.169 kg/m^2 and 0.184 kg/m^2; $\rho_{A,dc}$ between 0.069 kg/m^2 and 0.086 kg/m^2) and wood chips with spent coffee grounds ($\rho_{A,ac}$ between 0.190 kg/m^2 and 0.211 kg/m^2; $\rho_{A,dc}$ between 0.069 kg/m^2 and 0.086 kg/m^2). 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^2 and 0.140 kg/m^2) and hazelnut shells ($\rho_{A,ac}$ between 0.106 kg/m^2 and 0.131 kg/m^2); while the lowest ones were the ones of wood chips ($\rho_{A,ac}$ between 0.095 kg/m^2 and 0.102 kg/m^2) and grape press waste ($\rho_{A,ac}$ between 0.101 kg/m^2 and 0.114 kg/m^2). 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^2 and 0.128 kg/m^2. 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^2 and 0.096 kg/m^2 and $\rho_{A,dc}$ between 0.087 kg/m^2 and 0.091 kg/m^2, respectively). The others showed similar performance: $\rho_{A,dc}$ between 0.065 kg/m^2 and 0.075 kg/m^2, for samples made up of grape press waste; $\rho_{A,dc}$ between 0.067 kg/m^2 and 0.073 kg/m^2, for olive press waste; $\rho_{A,dc}$ between 0.058 kg/m^2 and 0.069 kg/m^2, for hazelnut shells.

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 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 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].

Tables 9 and 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 10 since they are not bio-based composites). The researchers achieved values between 0.7 and 0.8 g/(m^2.%RH) for a gypsum board and a gypsum plaster, between 0.4 – 0.5 g/(m^2.%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 Thieblesson et al. [94] reached lower values than the results of the present study. High hygroscopicity

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

Materials/Composites	MBV [g/(m^2.%RH)]	Classification	Ref.
Composites with wood fibre, cellulose wadding, and paper granules with potato starch [a]	1.1–2.5	Good/ Excellent	[94]
Composites with hemp shiv and clay [a]	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) [a]	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 [a]	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 [a]	2.29–2.59	Excellent	[95]
Composites with olive fibre and sodium silicate solution [b]	5.05	Excellent	[38]
Composites with straw fibre and sodium silicate solution [b]	3.29	Excellent	[38]
Clay plaster with hemp shiv and fibres [c]	2.20	Excellent	[96]
Wood-based panel with coniferous wood chips and cement (not specified)[a]	3.25	Excellent	[97]
Chipboard panel[a]	2.25	Excellent	[97]
OSB panel [a]	1.25	Good	[97]
Hemp fibre panel [a]	2.25	Excellent	[97]
Wood fibre panel [a]	3.10	Excellent	[97]

[a] MBV calculated by Nordtest method [82]: 8 h at high RH (75 %) followed by 16 h at low RH (33 %);
[b] MBV ideal calculated according to Nordtest project [82];
[c] MBV calculated from the results of a study that analysed moisture properties by using ISO 24353 [80].

Table 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
Composites A with gypsum-based binder				
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
Composites B with sodium silicate solution				
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
Composites C with natural glue				
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

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.

3.4. Bio-susceptibility to moulds

Table 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 2.3).

Fig. 10 shows the centre square (4 cm^2) of the contaminated composites at the end of the four weeks.

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 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 (Figs. 10f and 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.

Table 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

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

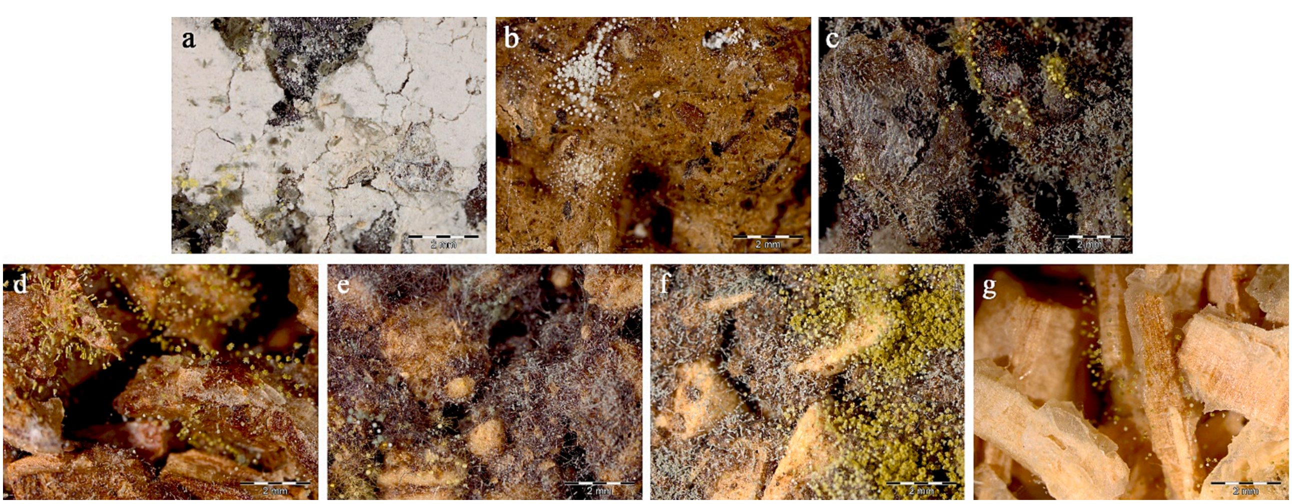

Fig. 10. Mould development at the end of the four weeks: (a) AG; (b) AO; (c) CG; (d) CH; (e) CO; (f) CWc; (g) CW.

Table 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-values less than 0.05.

demonstrated that the composites' final properties depend on both the aggregate and the binder.

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 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 verified (i.e., P-value greater than 0.05, the mean values were the same).

Table 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 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 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 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.

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

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.

CRediT authorship contribution statement

Eleonora Cintura: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft. **Paulina Faria:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing, Project administration. **Marta Duarte:** Investigation. **Lina Nunes:** Conceptualization, Project administration, Methodology, Supervision, Writing – review & editing.

Declaration of Competing Interest

This research was funded by the Portuguese Foundation for Science and Technology (FCT- Fundação para a Ciência e a Tecnologia), with Ph. D. grant PD/BD/150579/2020, as part of the Eco-Construction and Rehabilitation Program (EcoCoRe).

Data availability

Data will be made available on request.

Acknowledgements

This research was funded by the Portuguese Foundation for Science and Technology (FCT- Fundação para a Ciência e a Tecnologia), with Ph. D. grant PD/BD/150579/2020, as part of the Eco-Construction and Rehabilitation Program (EcoCoRe). The authors are grateful for the Portuguese Foundation for Science and Technology support through funding UIDB/04625/2020 of the research unit CERIS. The authors acknowledge Professor Bruno Esteves, Polytechnic Institute of Viseu, and Professor Susana Filipe Barreiros, NOVA School of Science and Technology, NOVA University of Lisbon, for their help in providing some of the materials used in this work. The authors are grateful to Mirko Braga (Ingessil S.r.l.) for the chemical analysis of the sodium silicate solution. The authors acknowledge Professor Ines Flores-Colen and Professor Maria da Glória Gomes, Instituto Superior Técnico, University of Lisbon, for making available the thermal conductivity equipment, and Sónia Duarte, National Laboratory for Civil Engineering, for the help provided for the statistical analysis.

References

[1] M.I. Gomes, P. Faria, T.D. Gonçalves, Earth-based mortars for repair and protection of rammed earth walls. stabilization with mineral binders and fibers, J. Clean. Prod. 172 (2018) 2401–2414, https://doi.org/10.1016/j.jclepro.2017.11.170.

[2] C. Martínez-García, B. González-Fonteboa, D. Carro-López, J.L. Pérez-Ordóñez, Mussel shells: a canning industry by-product converted into a bio-based insulation material, J. Clean. Prod. 269 (2020), 122343, https://doi.org/10.1016/j.jclepro.2020.122343.

[3] M. Posani, M.D.R. Veiga, V. Peixoto de Freitas, K. Kompatscher, H. Schellen, J. Kurnitski, T. Kalamees, Dynamic hygrothermal models for monumental, historic buildings with HVAC systems: complexity shown through a case study, E3S Web of Conferences. 172 (2020) 15007.

[4] S. Greco, L. Molari, Flexural behavior of six species of italian bamboo, Bio-Based Build. Mater. (2022) 723–729, https://doi.org/10.4028/www.scientific.net/CTA.1.723.

[5] T. Dickson, S. Pavía, Energy performance, environmental impact and cost of a range of insulation materials, Renew. Sustain. Energy Rev. 140 (2021), 110752, https://doi.org/10.1016/j.rser.2021.110752.

[6] M. Posani, R. Veiga, V.P. de Freitas, Retrofitting historic walls: feasibility of thermal insulation and suitability of thermal mortars, Heritage 4 (2021) 2009–2022, https://doi.org/10.3390/heritage4030114.

[7] M. La Gennusa, C. Marino, A. Nucara, M.F. Panzera, M. Pietrafesa, Insulating building components made from a mixture of waste and vegetal materials: thermal characterization of nine new products, Sustainability 13 (2021) 13820, https://doi.org/10.3390/su132413820.

[8] G. Donini, S. Greco, L. Molari, A. Zanetti, Structural design of an Italian bamboo house in an Italian regulatory context: revisiting a small building built in Costa Rica with tropical bamboo, Case Studies Constr. Mater. 16 (2022) e00891.

[9] M. Palumbo, A.M. Lacasta, N. Holcroft, A. Shea, P. Walker, Determination of hygrothermal parameters of experimental and commercial bio-based insulation materials, Constr. Build. Mater. 124 (2016) 269–275, https://doi.org/10.1016/j.conbuildmat.2016.07.106.

[10] L. Liu, H. Li, A. Lazzaretto, G. Manente, C. Tong, Q. Liu, N. Li, The development history and prospects of biomass-based insulation materials for buildings, Renew. Sustain. Energy Rev. 69 (2017) 912–932, https://doi.org/10.1016/j.rser.2016.11.140.

[11] L. Barbieri, F. Andreola, I. Lancellotti, R. Taurino, Management of agricultural biomass wastes: preliminary study on characterization and valorisation in clay matrix bricks, Waste Manag. 33 (2013) 2307–2315, https://doi.org/10.1016/j.wasman.2013.03.014.

[12] M.V. Madurwar, R.V. Ralegaonkar, S.A. Mandavgane, Application of agro-waste for sustainable construction materials: a review, Constr. Build. Mater. 38 (2013) 872–878, https://doi.org/10.1016/j.conbuildmat.2012.09.011.

[13] M.P. Ansell, M. Lawrence, Y. Jiang, A. Shea, A. Hussain, J. Calabria-Holley, P. Walker, Natural plant-based aggregates and bio-composite panels with low thermal conductivity and high hygrothermal efficiency for applications in construction, in: Nonconv. Vernac. Constr. Mater., Elsevier, 2020: pp. 217–245. https://doi.org/10.1016/B978-0-08-102704-2.00010-X.

[14] M. Gürü, A.F. Karabulut, M.Y. Aydın, İ. Bilici, Processing of fireproof and high temperature durable particleboard from rice husk, High Temp. Mater. Process. 34 (2015) 599–604, https://doi.org/10.1515/htmp-2014-0092.

[15] G.I. Mantanis, E.T. Athanassiadou, M.C. Barbu, K. Wijnendaele, Adhesive systems used in the European particleboard, MDF and OSB industries, Wood, Mater. Sci. Eng. 13 (2018) 104–116, https://doi.org/10.1080/17480272.2017.1396622.

[16] A.A. Owodunni, J. Lamaming, R. Hashim, O.F.A. Taiwo, M.H. Hussin, M. H. Mohamad Kassim, Y. Bustami, O. Sulaiman, M.H.M. Amini, S. Hiziroglu, Adhesive application on particleboard from natural fibers: a review, Polym. Compos. 41 (2020) 4448–4460, https://doi.org/10.1002/pc.25749.

[17] F.G. Gonçalves, J.B. Paes, Y.M. Lopez, P.G.d.A. Segundinho, R.G.E. de Oliveira, M.V. Fassarella, A.S. Brito, I.L.S. Chaves, R.S.F. Martins, Resistance of particleboards produced with ligno-cellulosic agro-industrial wastes to fungi and termites, Int. Biodeterior. Biodegrad. 157 (2021), 105159, https://doi.org/10.1016/j.ibiod.2020.105159.

[18] A. Mahieu, S. Alix, N. Leblanc, Properties of particleboards made of agricultural by-products with a classical binder or self-bound, Ind. Crops Prod. 130 (2019) 371–379, https://doi.org/10.1016/j.indcrop.2018.12.094.

[19] M. Pavelek, T. Adamová, Bio-waste thermal insulation panel for sustainable building construction in steady and unsteady-state conditions, Mater. (Basel). 12 (2019) 1–17, https://doi.org/10.3390/ma12122004.

[20] A. Ramos, A. Briga-Sá, S. Pereira, M. Correia, J. Pinto, I. Bentes, C.A. Teixeira, Thermal performance and life cycle assessment of corn cob particleboards, J. Build. Eng. 44 (2021), 102998, https://doi.org/10.1016/j.jobe.2021.102998.

[21] C. Pina dos Santos, L. Matias, ITE 50, LNEC, Coeficientes de transmissão térmica de elementos da envolvente dos edifícios, 2006, p. 171.

[22] A. Romano, A. Bras, S. Grammatikos, A. Shaw, M. Riley, Dynamic behaviour of bio-based and recycled materials for indoor environmental comfort, Construction and Building Materials. 211 (2019) 730–743, https://doi.org/10.1016/j.conbuildmat.2019.02.126.

[23] H. Binici, O. Aksogan, A. Dıncer, E. Luga, M. Eken, O. Isikaltun, The possibility of vermiculite, sunflower stalk and wheat stalk using for thermal insulation material production, Thermal Sci. Eng Progress. 18 (2020), 100567, https://doi.org/10.1016/j.tsep.2020.100567.

[24] L.D. Hung Anh, Z. Pásztory, An overview of factors influencing thermal conductivity of building insulation materials, J. Build. Eng. 44 (2021), 102604, https://doi.org/10.1016/j.jobe.2021.102604.

[25] Celenit, Celenit N, (2022). https://www.celenit.com/en-UK/celenit-n.php (accessed March 16, 2022).

[26] Celenit, Celenit ABE, (2018). http://www.lusomatec.pt/admin/content/ficha_tecnica_celenit_abe_pdf1568212973.pdf (accessed March 16, 2022).

[27] Kerakoll, KlimaExpert5, (2022). https://maifredicostruzioni.com/wp-content/uploads/2017/12/Scheda-KlimaExpert-5.pdf (accessed March 16, 2022).

[28] Knauf, KNAUF ORGANIC, (2022). https://www.knauf.pt/solucoes/placas/la-madeira/fibralith-organic.html#showtab-tab_documentacion (accessed March 16, 2022).

[29] Knauf, KNAUF STANDARD A, (2022). https://www.knauf.pt/sites/default/files/2020-01/K711B.pt Standard A (2019-11).pdf (accessed March 16, 2022).

[30] Vestaeco, VestaEco INTERNAL, (2022). http://www.vestaeco.com/produkt_VestaEco_INTERNAL,11.html (accessed March 16, 2022).

[31] E.P. Federation, OSB Panels, (2022). https://europanels.org/ (accessed October 6, 2021).

[32] A. Antunes, P. Faria, V. Silva, A. Brás, Rice husk-earth based composites: A novel bio-based panel for buildings refurbishment, Constr. Build. Mater. 221 (2019) 99–108, https://doi.org/10.1016/j.conbuildmat.2019.06.074.

[33] H. Binici, O. Aksogan, C. Demirhan, Mechanical, thermal and acoustical characterizations of an insulation composite made of bio-based materials, Sustain. Cities Soc. 20 (2016) 17–26, https://doi.org/10.1016/j.scs.2015.09.004.

[34] J.H. Park, Y. Kang, J. Lee, S.J. Chang, S. Wi, S. Kim, Development of wood-lime boards as building materials improving thermal and moisture performance based on hygrothermal behavior evaluation, Constr. Build. Mater. 204 (2019) 576–585, https://doi.org/10.1016/j.conbuildmat.2019.01.139.

[35] A.S. Tártaro, T.M. Mata, A.A. Martins, J.C.G. Esteves da Silva, Carbon footprint of the insulation cork board, J. Clean. Prod. 143 (2017) 925–932, https://doi.org/10.1016/j.jclepro.2016.12.028.

[36] H. Fu, Y. Ding, M. Li, H. Li, X. Huang, Z. Wang, Research on thermal performance and hygrothermal behavior of timber-framed walls with different external insulation layer: Insulation Cork Board and anti-corrosion pine plate, J. Build. Eng. 28 (2020), 101069, https://doi.org/10.1016/j.jobe.2019.101069.

[37] B. Mazhoud, F. Collet, S. Prétot, C. Lanos, Effect of hemp content and clay stabilization on hygric and thermal properties of hemp-clay composites, Constr. Build. Mater. 300 (2021), 123878, https://doi.org/10.1016/j.conbuildmat.2021.123878.

[38] S. Liuzzi, C. Rubino, F. Martellotta, P. Stefanizzi, C. Casavola, G. Pappalettera, Characterization of biomass-based materials for building applications: the case of straw and olive tree waste, Ind. Crops Prod. 147 (2020), 112229, https://doi.org/10.1016/j.indcrop.2020.112229.

[39] D. Eliche-Quesada, S. Ruiz-Molina, L. Pérez-Villarejo, E. Castro, P.J. Sánchez-Soto, Dust filter of secondary aluminium industry as raw material of geopolymer foams, J. Build. Eng. 32 (2020) 101656.

[40] H. Younesi-Kordkheili, A. Pizzi, Improving the physical and mechanical properties of particleboards made from urea–glyoxal resin by addition of pMDI, Eur. J. Wood Wood Prod. 76 (2018) 871–876, https://doi.org/10.1007/s00107-017-1242-3.

[41] P. Solt, J. Konnerth, W. Gindl-Altmutter, W. Kantner, J. Moser, R. Mitter, H.W. G. van Herwijnen, Technological performance of formaldehyde-free adhesive alternatives for particleboard industry, Int. J. Adhes. Adhes. 94 (2019) 99–131, https://doi.org/10.1016/j.ijadhadh.2019.04.007.

[42] P. Antov, V. Savov, G.I. Mantanis, N. Neykov, Medium-density fibreboards bonded with phenol-formaldehyde resin and calcium lignosulfonate as an eco-friendly additive, Wood, Mater. Sci. Eng. 16 (1) (2021) 42–48.

[43] S. Cardoso, L. Nunes, P. Faria, Utilização de colas naturais para placas de derivados de madeira - Uma síntese, Cienc. e Tecnol. Dos Mater. 27 (2015) 143–151, https://doi.org/10.1016/j.ctmat.2015.09.001.

[44] S.W. Kariuki, J. Wachira, M. Kawira, G. Murithi, Formaldehyde use and alternative biobased binders for particleboard formulation: a review, J. Chem. 2019 (2019) 1–12, https://doi.org/10.1155/2019/5256897.

[45] L. Muizniece, D. Blumberga, Thermal conductivity of heat insulation material made from coniferous needles with potato starch binder, Energy Procedia. 95 (2016) 324–329, https://doi.org/10.1016/j.egypro.2016.09.014.

[46] D. Nataraj, S. Sakkara, M. Hn, N. Reddy, Properties and applications of citric acid crosslinked banana fibre-wheat gluten films, Ind. Crops Prod. 124 (2018) 265–272.

[47] S. Monteiro, J. Martins, F.D. Magalhães, L. Carvalho, Low density wood particleboards bonded with starch foam—study of production process conditions, Mater. (Basel). 12 (2019) 1975, https://doi.org/10.3390/ma12121975.

[48] E. Cintura, L. Nunes, B. Esteves, P. Faria, Agro-industrial wastes as building insulation materials: a review and challenges for euro-mediterranean countries, Ind. Crops Prod. 171 (2021), 113833, https://doi.org/10.1016/j.indcrop.2021.113833.

[49] FAO, FAO Crops and livestock products, (2020). https://www.fao.org/faostat/en/#data/QCL (accessed August 31, 2022)..

[50] FAO, FAO Supply Utilization Accounts, (2019). https://www.fao.org/faostat/en/#data/SCL (accessed August 31, 2022).

[51] M. Palumbo, A.M. Lacasta, A. Navarro, M.P. Giraldo, B. Lesar, Improvement of fire reaction and mould growth resistance of a new bio-based thermal insulation material, Constr. Build. Mater. 139 (2017) 531–539, https://doi.org/10.1016/j.conbuildmat.2016.11.020.

[52] B.K. Stefanowski, S.F. Curling, G.A. Ormondroyd, A rapid screening method to determine the susceptibility of bio-based construction and insulation products to mould growth, Int. Biodeterior. Biodegradation. 116 (2017) 124–132, https://doi.org/10.1016/j.ibiod.2016.10.025.

[53] M. Viel, F. Collet, Y. Lecieux, M.L.M. François, V. Colson, C. Lanos, A. Hussain, M. Lawrence, Resistance to mold development assessment of bio-based building materials, Compos. Part B Eng. 158 (2019) 406–418, https://doi.org/10.1016/j.compositesb.2018.09.063.

[54] A. Brambilla, A. Sangiorgio, Mould growth in energy efficient buildings: causes, health implications and strategies to mitigate the risk, Renew. Sustain. Energy Rev. 132 (2020), 110093, https://doi.org/10.1016/j.rser.2020.110093.

[55] C.A. Echeverria, J. Ozkan, F. Pahlevani, M. Willcox, V. Sahajwalla, Effect of hydrothermal hot-compression method on the antimicrobial performance of green building materials from heterogeneous cellulose wastes, J. Clean. Prod. 280 (2021), 124377, https://doi.org/10.1016/j.jclepro.2020.124377.

[56] Y. Indrayani, D. Setyawati, S.S. Munawar, K. Umemura, T. Yoshimura, Evaluation of termite resistance of medium density fiberboard (MDF) manufacture from agricultural fiber bonded with citric acid, Procedia Environ. Sci. 28 (2015) 778–782, https://doi.org/10.1016/j.proenv.2015.07.091.

[57] P.S. de Carvalho, M.D. Nora, L.C. da Rosa, Development of an acoustic absorbing material based on sunflower residue following the cleaner production techniques, J. Clean. Prod. 270 (2020), 122478, https://doi.org/10.1016/j.jclepro.2020.122478.

[58] T. Santos, L. Nunes, P. Faria, Production of eco-efficient earth-based plasters: Influence of composition on physical performance and bio-susceptibility, J. Clean. Prod. 167 (2017) 55–67, https://doi.org/10.1016/j.jclepro.2017.08.131.

[59] J. Krejsová, M. Doleželová, Resistance of mortars with gypsum, lime and composite binders against molds, Acta Polytech. CTU Proc. 21 (2019) 16–20, https://doi.org/10.14311/APP.2019.21.0016.

[60] A. Treu, L. Nunes, E. Larnøy, Macrobiological degradation of esterified wood with sorbitol and citric acid, Forests. 11 (2020) 776, https://doi.org/10.3390/f11070776.

[61] E. Cintura, P. Faria, M. Duarte, L. Nunes, Bio-wastes as aggregates for eco-efficient boards and panels: screening tests of physical properties and bio-susceptibility, Infrastructures. 7 (2022) 26, https://doi.org/10.3390/infrastructures7030026.

[62] H. Younesi-Kordkheili, A. Pizzi, Improving the properties of urea-lignin-glyoxal resin as a wood adhesive by small addition of epoxy, Int. J. Adhes. Adhes. 102 (2020), 102681, https://doi.org/10.1016/j.ijadhadh.2020.102681.

[63] European Committee for Standardisation (CEN), EN 459-1:2015: Building lime Part 1: Definitions, specifications and conformity criteria. Brussels, 2015.

[64] S.H. Lee, P. Md Tahir, W.C. Lum, L.P. Tan, P. Bawon, B.-D. Park, S.S. Osman Al Edrus, U.H. Abdullah, A review on citric acid as green modifying agent and binder for wood, Polym. (Basel) 12 (8) (2020) 1692.

[65] Z. Huaxu, L.S. Hua, P.M. Tahir, Z. Ashaari, S.S.O. Al-Edrus, N.A. Ibrahim, L. C. Abdullah, S.F. Mohamad, Physico-mechanical and biological durability of citric acid-bonded rubberwood particleboard, Polymers (Basel). 13 (2021) 1–12, https://doi.org/10.3390/polym13010098.

[66] S.J. Lee, V. Thole, Investigation of modified water glass as adhesive for wood and particleboard: mechanical, thermal and flame retardant properties, Eur. J. Wood Wood Prod. 76 (2018) 1427–1434, https://doi.org/10.1007/s00107-018-1324-x.

[67] N.H. Budak, E. Aykin, A.C. Seydim, A.K. Greene, Z.B. Guzel-Seydim, Functional properties of vinegar, J. Food Sci. 79 (2014) R757–R764, https://doi.org/10.1111/1750-3841.12434.

[68] M. Lanzón, P.A. García-Ruiz, Effect of citric acid on setting inhibition and mechanical properties of gypsum building plasters, Constr. Build. Mater. 28 (2012) 506–511, https://doi.org/10.1016/j.conbuildmat.2011.06.072.

[69] G. Vasconcelos, P.B. Lourenço, P. Mendonça, A. Camões, R. Mateus, L. Bragança, A.G. Brito, E. Poletti, Proposal of an innovative solution for partition walls: mechanical, thermal and acoustic validation, Constr. Build. Mater. 48 (2013) 961–979, https://doi.org/10.1016/j.conbuildmat.2013.07.079.

[70] F. Matalkah, H. Bharadwaj, A. Balachandra, P. Soroushian, Development and characterization of gypsum-based binder, Eur. J. Adv. Eng. Technol. 4 (2017) 153–157.

[71] Sival, Technical sheet Gypsum – fine plaster. https://sival.pt/img/cms/pdfs/fichas_tecnicas_pt/gesso estuque fino_1.pdf (accessed October 27, 2021).

[72] Labkem, Technical sheet Sodium Silicate Solution. https://esp.labbox.com/wp-content/uploads/FT/PDS-10511.pdf (accessed October 27, 2021).

[73] Ingessil, https://www.ingessil.com/it (accessed June 28, 2022).

[74] D. Dai, M. Fan, Preparation of bio-composite from wood sawdust and gypsum, Ind. Crops Prod. 74 (2015) 417–424, https://doi.org/10.1016/j.indcrop.2015.05.036.

[75] S. Liuzzi, C. Rubino, P. Stefanizzi, F. Martellotta, Performance characterization of broad band sustainable sound absorbers made of almond skins, Mater. (Basel). 13 (2020) 5474, https://doi.org/10.3390/ma13235474.

[76] European Committee for Standardisation (CEN), EN 323:1993: Wood-based panels - Determination of density. Brussels, 1993.

[77] S. Liuzzi, C. Rubino, P. Stefanizzi, A. Petrella, A. Boghetich, C. Casavola, G. Pappalettera, Hygrothermal properties of clayey plasters with olive fibers, Constr. Build. Mater. 158 (2018) 24–32, https://doi.org/10.1016/j.conbuildmat.2017.10.013.

[78] M. Posani, R. Veiga, V.P. de Freitas, Thermal mortar-based insulation solutions for historic walls: an extensive hygrothermal characterization of materials and systems, Constr. Build. Mater. 315 (2022), 125640, https://doi.org/10.1016/j.conbuildmat.2021.125640.

[79] M.G. Gomes, I. Flores-Colen, F. da Silva, M. Pedroso, Thermal conductivity measurement of thermal insulating mortars with EPS and silica aerogel by steady-state and transient methods, Constr. Build. Mater. 172 (2018) 696–705, https://doi.org/10.1016/j.conbuildmat.2018.03.162.

[80] International Organization for Standardization (ISO), ISO 24353:2008: Hygrothermal performance of building materials and products - Determination of moisture adsorption/desorption properties in response to humidity variation. Switzerland, 2008.

[81] A. Ranesi, M. Posani, R. Veiga, P. Faria, A Discussion on winter indoor hygrothermal conditions and hygroscopic behaviour of plasters in southern Europe, Infrastructures. 7 (2022) 38, https://doi.org/10.3390/infrastructures7030038.

[82] C. Rode, R.H. Peuhkuri, L.H. Mortensen, K.K. Hansen, B. Time, A. Gustavsen, T. Ojanen, J. Ahonen, K. Svennberg, C. Arfvidsson, R.H. Jesper TotalRode, L. H. Peuhkuri, K.K. Mortensen, B. Hansen, A. Time, T. Gustavsen, J. Ojanen, K. Ahonen, J.T.A. Svennberg, Moisture Buffering of Building Materials, Department of Civil Engineering, Technical University of Denmark, 2005. BYG Report No. R-127)ISBN 8778771951.

[83] A. Ranesi, P. Faria, R. Veiga, Moisture Buffering Value of Plasters: The Influence of Two Different Test Methods, in: TEST&E 2022, Lisbon, 2023: pp. 97–105. https://doi.org/10.1007/978-3-031-29191-3_8.

[84] P. Johansson, G. Bok, A. Ekstrand-Tobin, The effect of cyclic moisture and temperature on mould growth on wood compared to steady state conditions, Build. Environ. 65 (2013) 178–184, https://doi.org/10.1016/j.buildenv.2013.04.004.

[85] T. Verdier, M. Coutand, A. Bertron, C. Roques, A review of indoor microbial growth across building materials and sampling and analysis methods, Build. Environ. 80 (2014) 136–149, https://doi.org/10.1016/j.buildenv.2014.05.030.

[86] S. Ginestet, C. Aschan-Leygonie, T. Bayeux, M. Keirsbulck, Mould in indoor environments: the role of heating, ventilation and fuel poverty. a French perspective, Build. Environ. 169 (2020), 106577, https://doi.org/10.1016/j.buildenv.2019.106577.

[87] P. Johansson, I. Samuelson, A. Ekstrand-tobin, K. Mjörnell, P.I. Sandberg, E. Sikander, Microbiological growth on building materials – critical moisture levels, State of the art, Extr. from Rep. SP Rapp. 200511 from 2005(32) (2005) 11.

[88] European Technical Assessment, ETA 17/0389: 2017 – Granulated expanded cork. ITeCons, 2017.

[89] International Organization for Standardization (ISO), ISO 846:2019 - Plastics - Evaluation of the action of microorganisms. Brussels, 2019.

[90] C. Udawattha, H. Galkanda, I.S. Ariyarathne, G.Y. Jayasinghe, R. Halwatura, Mold growth and moss growth on tropical walls, Build. Environ. 137 (2018) 268–279, https://doi.org/10.1016/j.buildenv.2018.04.018.

[91] J.L. Parracha, G. Borsoi, I. Flores-Colen, R. Veiga, L. Nunes, A. Dionísio, M. G. Gomes, P. Faria, Performance parameters of ETICS: Correlating water resistance, bio-susceptibility and surface properties, Constr. Build. Mater. 272 (2021), 121956, https://doi.org/10.1016/j.conbuildmat.2020.121956.

[92] N. Fatma, L. Allègue, M. Salem, R. Zitoune, M. Zidi, The effect of doum palm fibers on the mechanical and thermal properties of gypsum mortar, J. Compos. Mater. 53 (2019) 2641–2659, https://doi.org/10.1177/0021998319838319.

[93] S. Adamopoulos, D. Foti, E. Voulgaridis, C. Passialis, Manufacturing and properties of gypsum-based products with recovered wood and rubber materials, BioResources. 10 (2015) 5573–5585, https://doi.org/10.15376/biores.10.3.5563-5572.

[94] L.M. Thieblesson, F. Collet, S. Prétot, C. Lanos, H. Kouakou, O. Boffoue, Hygric and thermal characterization of composites coupling recycled or raw bio-based materials and starch, 35 (2017) 409–415. https://doi.org/https://doi.org/10.26168/icbbm2017.62.

[95] M. Rahim, O. Douzane, A.D. Tran Le, G. Promis, T. Langlet, Characterization and comparison of hygric properties of rape straw concrete and hemp concrete, Constr. Build. Mater. 102 (2016) 679–687, https://doi.org/10.1016/j.conbuildmat.2015.11.021.

[96] A. Thomson, D. Maskell, P. Walker, M. Lemke, A. Shea, R. Lawrence, Improving hygrothermal properties of clay, in: 15th Int. Conf. Non-Conventional Mater. Technol. (NOCMAT 2015), 2015: pp. 897–898.

[97] F. Mnasri, S. Bahria, M.-E.-A. Slimani, O. Lahoucine, M. El Ganaoui, Building incorporated bio-based materials: Experimental and numerical study, J. Build. Eng. 28 (2020), 101088, https://doi.org/10.1016/j.jobe.2019.101088.

[98] D.M. Nguyen, A.-C. Grillet, T.M.H. Diep, C.N. Ha Thuc, M. Woloszyn, Hygrothermal properties of bio-insulation building materials based on bamboo fibers and bio-glues, Constr. Build. Mater. 155 (2017) 852–866, https://doi.org/10.1016/j.conbuildmat.2017.08.075.

[99] N.M.M. Ramos, J.M.P.Q. Delgado, V.P. De Freitas, Influence of finishing coatings on hygroscopic moisture buffering in building elements, Constr. Build. Mater. 24 (2010) 2590–2597, https://doi.org/10.1016/j.conbuildmat.2010.05.017.

[100] S. D'Amico, M. Hrabalova, U. Müller, E. Berghofer, Influence of ageing on mechanical properties of wood to wood bonding with wheat flour glueEinfluss der Alterung auf die mechanischen Eigenschaften von Weizenmehlleim für die Verklebung von Holz, Eur. J. Wood Wood Prod. 70 (5) (2012) 679–688.

[101] S. Monteiro, L. Nunes, J. Martins, F.D. Magalhães, L. Carvalho, Low-density cardoon (Cynara cardunculus L.) particleboards bound with potato starch-based adhesive, Polym. (Basel) 12 (2020) 1–16, https://doi.org/10.3390/polym12081799.

[102] G. Bumanis, L. Vitola, I. Pundiene, M. Sinka, D. Bajare, Gypsum, geopolymers, and starch—alternative binders for bio-based building materials: a review and life-cycle assessment, Sustainability 12 (2020) 5666, https://doi.org/10.3390/su12145666.

[103] E. Cintura, L. Nunes, P. Faria, Characterization of agro-wastes to be used as aggregates for eco-efficient insulation boards, in: CEES 2021 - Int. Conf. Constr. Energy, Environ. Sustain., ISBN: 978-989-54499-1-0, Coimbra, Portugal, 2021.