Optimisation of Production Parameters to Develop Innovative Eco-efficient Boards

Eleonora Cintura[1,2(✉)], Paulina Faria[1], Luisa Molari[3], and Lina Nunes[2,4]

[1] CERIS, Department of Civil Engineering, NOVA School of Science and Technology, NOVA University of Lisbon, Caparica, Portugal
ecintura@fct.unl.pt

[2] Structures Department, National Laboratory for Civil Engineering, Lisbon, Portugal

[3] Department of Civil, Chemical, Environmental and Materials Engineering, DICAM, Alma Mater Studiorum, University of Bologna, Bologna, Italy

[4] CE3C, Centre for Ecology, Evolution and Environmental Changes and CHANGE, Global Change and Sustainability Institute, University of the Azores, Angra do Heroísmo, Portugal

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

Keywords: Bio-waste · Hazelnut shells · Sodium silicate

1 Introduction

The use of panels such as particleboards as building materials has several benefits. Besides the easy on-site installation, the economic advantages, and the several applications, there is the possibility of employing different materials as aggregates. Among them, recycled materials and by-products derived from agro-industrial practices [1, 2]. Considering the high environmental impact caused by both the construction sector and

S. Amziane et al. (Eds.): ICBBM 2023, RILEM Bookseries 45, pp. 111–122, 2023.
https://doi.org/10.1007/978-3-031-33465-8_10

agriculture [3], the production of agricultural waste-based materials seems to be an efficient solution that encourages a circular economy system [4]. Several studies addressed the feasibility of producing panels by using agro-industrial wastes, for example, cork, coffee chaff, rice husk and corn cob [5–7]. Many of the developed composites met the requirements of the standards, proving to be a good alternative to conventional materials.

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

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

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

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

2 Materials

2.1 Hazelnut Shells

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

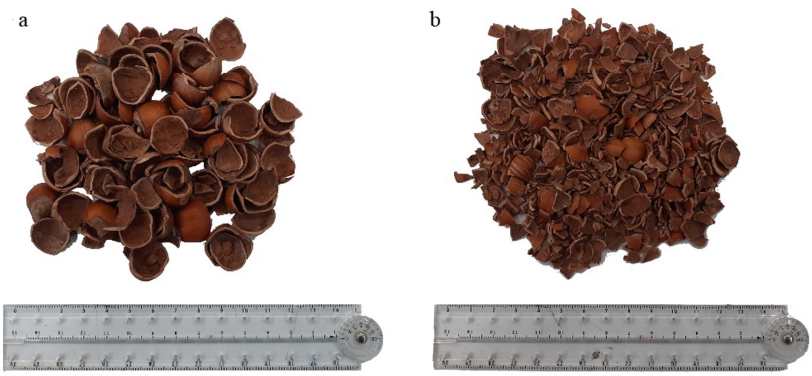

Fig. 1. Hazelnut shells used as aggregates: a) before shredding; b) after shredding.

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

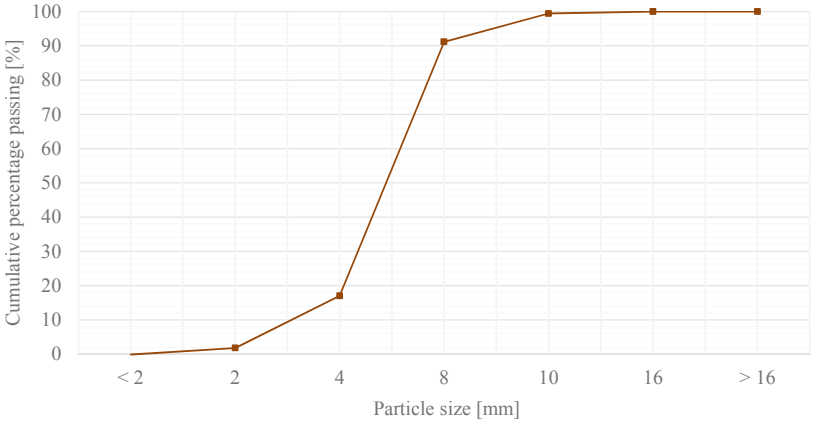

Fig. 2. Particle size distribution of the shredded hazelnut shells.

2.2 Sodium Silicate Solution and Sodium Bicarbonate

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

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

Property	Value
Weight ratio	2.4
Density [°Bè]	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 °C	1.471
pH (T = 20 °C)	12.40

The sodium bicarbonate, $NaHCO_3$, was produced by Crastan S.p.A., Pontedera (PI), Italy [26]. This is a commonly used sodium bicarbonate. Its use will be justified and detailed in Sect. 3.

3 Production Parameters

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

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

Fig. 3. The considered moulds to produce the samples: metal, wooden and silicone.

The bonding of the samples was easily avoided by using the silicone mould. As for the other moulds, materials that guarantee the remotion of the samples were needed (e.g., resins or anti-glueing materials). The benefit of the wooden and the metal moulds was the possibility of opening them, demoulding the sample without moving it and leaving its sides drying. As a result, the silicone mould and the wooden mould covered by baking paper were selected as the best solutions.

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

Different production parameters were investigated, such as drying at temperatures between T = 60 °C and T = 80 °C and the addition of sodium bicarbonate ($NaHCO_3$) as solid reactive. The mixtures of hazelnut shells and sodium silicate solution (and eventually sodium bicarbonate) were placed in the silicone moulds (8.8 cm × 5 cm ×

2.5 cm high) and 12 different production parameters were considered and compared (A to L). They are reported in Table 2, with the composites' designation. The samples were demoulded after 3 days.

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

Composites' designation	Production parameters	Description
A	Reference	Dring at laboratory conditions without moving the sample
B	Rotation	Dring at laboratory conditions by rotating the sample each 3 h
C	Temperature	Dring at T = 60 °C for 2 h
D	Addition of reactive	Addition of 25% of $NaHCO_3$ (by vol. of sodium silicate solution); drying at laboratory conditions without moving the sample
E	Addition of reactive, rotation	Addition of 25% of $NaHCO_3$ (by vol. of sodium silicate solution); drying at laboratory conditions by rotating the sample each 3 h
F	Addition of reactive, temperature, rotation	Addition of 25% of $NaHCO_3$ (by vol. of sodium silicate solution); drying at T = 60 °C for 1 h by rotating the sample every 30 min
G	Temperature, rotation	Drying at T = 80 °C for 1.5 h, by rotating the sample each 30 min
H	Temperature	Drying at T = 80 °C for 1.5 h, without moving the sample
I	Addition of reactive, temperature	Addition of 10% of $NaHCO_3$ (by vol. of sodium silicate solution), drying at T = 60 °C for 1 h
J	Addition of reactive, temperature	Addition of 2% of $NaHCO_3$ (by vol. of sodium silicate solution), drying dry at T = 60 °C for 1 h
K	Addition of reactive, temperature, rotation	Addition of 2% of $NaHCO_3$ (by vol. of sodium silicate solution), drying at T = 60 °C for 30 min by rotating the sample every 15 min; then, drying at laboratory conditions by rotating the sample each 1 h
L	Addition of reactive, rotation	Addition of 2% of $NaHCO_3$ (by vol. of sodium silicate solution), drying at laboratory conditions by rotating the sample each 1 h

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

4 Results and Discussion

Figure 4 shows the samples of each composite after demoulding.

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

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

Table 3. Results of the composite samples visual analysis.

Composite	Visual analysis
A	The sample was not uniform, and it was not dried after 3 days; the sodium silicate solution was mainly on the bottom side; six days were required to demould
B	The sample was quite uniform; in some parts, the sodium silicate solution was not completely uniformly distributed; overall, it was throughout the sample; demoulding was possible after 3 days; the sample was dried after 6 days
C	The sample was quite uniform; in a few parts, the silicate was not completely uniformly distributed (less than for sample B), but overall, it was throughout the sample; 3 days were required to demould; the sample was dried after 6 days
D	The sample seems to be uniform, even if the sodium silicate solution was not perfectly distributed; since after 2 days the sample was dried, the $NaHCO_3$ accelerated the drying process; however, after 6 days, the sample started to release the $NaHCO_3$, probably added in too high quantities; the $NaHCO_3$ was strongly visible on the surface of the sample as a white powder
E	It was broken after 1 day; probably the $NaHCO_3$ absorbed the sodium silicate solution decreasing its bonding performance; the $NaHCO_3$ was visible on the surface of the sample as a white powder
F	After 1 h the sodium silicate seemed to be completely dried but the quantities of the $NaHCO_3$ were too high; the sample did not seem resistant and it was highly crumbly; the $NaHCO_3$ was visible on the surface as a white powder
G	The sample was quite uniform; the sodium silicate solution was not completely uniformly distributed, but overall, it was throughout the sample; demoulding was possible after 3 days; after 6 days the sample was dried; it seemed similar to sample C
H	The sample was not uniform; the sodium silicate solution was deposited on the bottom surface, as happened for sample A; when the drying process finished, it was similar to sample C, even if less uniform
I	The sample did not seem uniform at the end of the drying process and after demoulding; the sodium silicate solution was on the bottom side of the sample; the $NaHCO_3$ was highly visible on the surface of the sample as a white powder
J	The sample was not uniform; the sodium silicate solution was on the bottom part of the sample; it seemed similar to sample I, but more resistant (probably due to the fewer quantities of $NaHCO_3$); due to the less employed quantities, the $NaHCO_3$ was less visible on the surface of the sample
K	The sample was not completely uniform after 3 days; however, the sodium silicate solution was throughout the sample and quite distributed; it seemed similar to sample C; due to the less employed quantities, the $NaHCO_3$ was less visible on the surface of the sample

(*continued*)

Table 3. (*continued*)

Composite	Visual analysis
L	The sample was not uniform; in many parts of the bottom side, there were high quantities of the sodium silicate solution; demoulding was possible after 3 days; the sample seemed similar to C; the $NaHCO_3$ was not highly visible on the surface of the sample

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

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

Thus, the production process was defined. First of all, the bio-aggregates and the sodium silicate solution were mechanically mixed for 10 min, until homogeneity. Then, the mixture was placed without compaction in silicone moulds or wooden moulds covered by baking paper. The mixture was levelled by using a spatula and the moulds were closed by a wooden top. After that, the samples were dried at T = 60 °C for 3 h, being

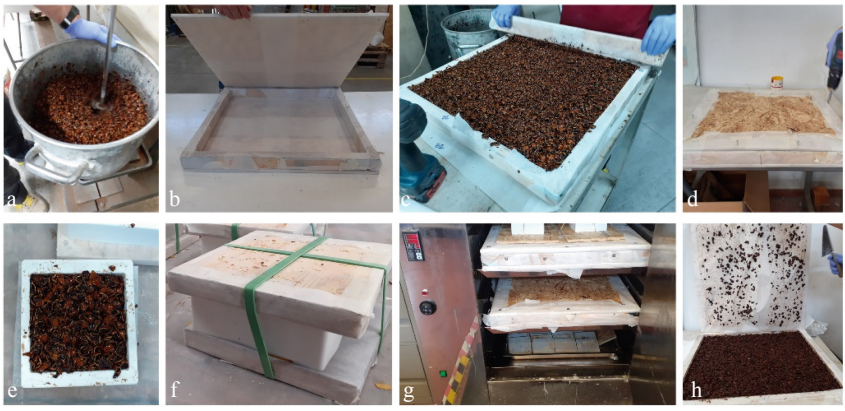

Fig. 5. Production process: a) mechanical mixing; b) wooden mould; c) levelling; d) closure of the wooden mould; e) silicone mould; f) closure of the silicone mould; g) drying at T = 60 °C; h) demoulding.

rotated every 30 min. Finally, they were dried at laboratory conditions, rotated every 30 min and demoulded after 2 days.

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

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

5 Conclusions

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

The following conclusions were achieved:

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

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

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

Acknowledgements. This research was funded by FCT, the Portuguese Foundation for Science and Technology, with Ph.D. grant PD/BD/150579/2020, as part of the Eco-Construction and Rehabilitation Program (EcoCoRe). The authors are grateful for the FCT support through funding UIDB/04625/2020 of the research unit CERIS and for the support of the project BIO-FIBRE funded by the Erasmus+ Programme of the European Union.

The authors acknowledge Lorenzo Coraglia (Raccolti di CIN) for giving information about the production and harvesting of hazelnut and for donating the hazelnut shells to carry out the research project. The authors acknowledge Dr. Mirko Braga (Ingessil) for his help and suggestion to correctly use the sodium silicate solution as adhesive and Prof. Stefania Liuzzi (Poliba) to provide suggestions to carry out the research project.

References

1. Gürü, M., Karabulut, A.F., Aydın, M.Y., Bilici, İ: Processing of fireproof and high temperature durable particleboard from rice husk. High Temp. Mater. Process. **34**(6), 599–604 (2015). https://doi.org/10.1515/htmp-2014-0092

2. Owodunni, A.A., et al.: Adhesive application on particleboard from natural fibers: a review. Polym. Compos. **41**(11), 4448–4460 (2020). https://doi.org/10.1002/pc.25749
3. Duque-Acevedo, M., Lancellotti, I., Andreola, F., Barbieri, L., Belmonte-Ureña, L.J., Camacho-Ferre, F.: Management of agricultural waste biomass as raw material for the construction sector: an analysis of sustainable and circular alternatives. Environ. Sci. Eur. **34**(1), 70 (2022). https://doi.org/10.1186/s12302-022-00655-7
4. Liuzzi, S., Rubino, C., Stefanizzi, P., Martellotta, F.: Performance characterization of broad band sustainable sound absorbers made of almond skins. Materials **13**(23), 5474 (2020). https://doi.org/10.3390/ma13235474
5. Ricciardi, P., Belloni, E., Merli, F., Buratti, C.: Sustainable panels made with industrial and agricultural waste: thermal and environmental critical analysis of the experimental results. Appl. Sci. **11**(2), 494 (2021). https://doi.org/10.3390/app11020494
6. Ramos, A., et al.: Thermal performance and life cycle assessment of corn cob particleboards. J. Build. Eng. **44**, 102998 (2021). https://doi.org/10.1016/j.jobe.2021.102998
7. António, J., Tadeu, A., Marques, B., Almeida, J.A.S., Pinto, V.: Application of rice husk in the development of new composite boards. Constr. Build. Mater. **176**, 432–439 (2018). https://doi.org/10.1016/j.conbuildmat.2018.05.028
8. Cintura, E., Nunes, L., Esteves, B., Faria, P.: Agro-industrial wastes as building insulation materials: a review and challenges for Euro-Mediterranean countries. Ind. Crops Prod. **171**, 113833 (2021). https://doi.org/10.1016/j.indcrop.2021.113833
9. FAO: FAOSTAT Crops. https://www.fao.org/faostat/en/#data/QCL. Accessed 30 Nov 2022
10. Ansell, M.P., et al.: Natural Plant-Based Aggregates and Bio-Composite Panels with Low Thermal Conductivity and High Hygrothermal Efficiency for Applications in Construction, Non-conventional and Vernacular Construction Materials, 2nd (edn.) Elsevier (2020). ISBN: 9780081027042. https://doi.org/10.1016/B978-0-08-102704-2.00010-X
11. Vrtodušić, R., Ivić, D., Jemrić, T., Vuković, M.: Hazelnut postharvest technology: a review. J. Cent. Eur. Agric. **23**(2), 423–454 (2022). https://doi.org/10.5513/JCEA01/23.2.3493
12. Markuszewski, B., Bieniek, A.A., Wachowska, U., Bieniek, A., Krzymińska, I.: Effect of biological treatment used before harvesting and storage methods on the quality, health and microbial characteristics of unripe hazelnut in the husk (*Corylus avellana* L.). PeerJ **10**, e12760 (2022). https://doi.org/10.7717/peerj.12760
13. Younesi-Kordkheili, H., Pizzi, A.: Improving the physical and mechanical properties of particleboards made from urea–glyoxal resin by addition of pMDI. Eur. J. Wood Wood Prod. **76**(3), 871–876 (2017). https://doi.org/10.1007/s00107-017-1242-3
14. Ansell, M.P., et al.: Natural Plant-Based Aggregates and Bio-Composite Panels with Low Thermal Conductivity and High Hygrothermal Efficiency for Applications in Construction, Non-conventional and Vernacular Construction Materials, 2nd edn., Elsevier (2020). ISBN 9780081027042
15. Nordström, J., et al.: Concentration- and pH-dependence of highly alkaline sodium silicate solutions. J. Colloid Interface Sci. **356**(1), 37–45 (2011). https://doi.org/10.1016/j.jcis.2010.12.085
16. Li, L., Tang, X., Ouyang, Q., Tao, N.: Combination of sodium dehydroacetate and sodium silicate reduces sour rot of citrus fruit. Postharvest Biol. Technol. **151**, 19–25 (2019). https://doi.org/10.1016/j.postharvbio.2019.01.006
17. Liuzzi, S., Rubino, C., Martellotta, F., Stefanizzi, P., Casavola, C., Pappalettera, G.: Characterization of biomass-based materials for building applications: the case of straw and olive tree waste. Ind. Crops Prod. **147**, 112229 (2020). https://doi.org/10.1016/j.indcrop.2020.112229
18. Lee, S.J., Thole, V.: Investigation of modified water glass as adhesive for wood and particleboard: mechanical, thermal and flame retardant properties. Eur. J. Wood Wood Prod. **76**(5), 1427–1434 (2018). https://doi.org/10.1007/s00107-018-1324-x

19. Song, L., Liu, W., Xin, F., Li, Y.: Study of adhesion properties and mechanism of sodium silicate binder reinforced with silicate fume. Int. J. Adhes. Adhes. **106**, 102820 (2021). https://doi.org/10.1016/j.ijadhadh.2021.102820
20. Fawer, M., Concannon, M., Rieber, W.: Life cycle inventories for the production of sodium silicates. Int. J. Life Cycle Assess. **4**(4), 207 (1999). https://doi.org/10.1007/BF02979498
21. Wang, Y., He, F., Wang, J., Hu, Q.: Comparison of effects of sodium bicarbonate and sodium carbonate on the hydration and properties of portland cement paste. Materials **12**(7), 1033 (2019). https://doi.org/10.3390/ma12071033
22. Liu, S., Shen, Y., Wang, Y., He, H., Luo, S., Huang, C.: Synergistic use of sodium bicarbonate and aluminum sulfate to enhance the hydration and hardening properties of portland cement paste. Constr. Build. Mater. **299**, 124248 (2021). https://doi.org/10.1016/j.conbuildmat.2021.124248
23. Cintura, E., Faria, P., Duarte, M., Nunes, L.: Bio-wastes as aggregates for eco-efficient boards and panels: screening tests of physical properties and bio-susceptibility. Infrastructures **7**(3), 1–26 (2022). https://doi.org/10.3390/infrastructures7030026
24. Amziane, S., Collet, F., Lawrence, M., Magniont, C., Picandet, V., Sonebi, M.: Recommendation of the RILEM TC 236-BBM: characterisation testing of hemp shiv to determine the initial water content, water absorption, dry density, particle size distribution and thermal conductivity. Mater. Struct. **50**(3), 1–11 (2017). https://doi.org/10.1617/s11527-017-1029-3
25. Ingessil: https://www.ingessil.com/it. Accessed 30 Nov 2022
26. Crastan S.p.A: https://www.crastan.it/. Accessed 30 Dec 2022