Journal of Building Engineering 44 (2021) 102625

Contents lists available at ScienceDirect

Journal of Building Engineering

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

Binderless fiberboards for sustainable construction. Materials, production methods and applications

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ARTICLE INFO

Keywords:
Binderless fiberboards
Lignocellulosic materials
Sustainable construction
Pre-treatments
Construction materials

ABSTRACT

Fiberboards are readily available components which can be used in construction for various functions such as furniture, insulation, or soundproofing. Research in the field of fiberboards has developed considerably in trying to match the practical needs of the construction element together with the new environmental challenges, that favour the production of panels in using by-products without adhesives. This review article presents an overview on fiberboard production and may offer a way to establish all the necessary steps to make binderless fiberboards attractive on the market, by considering economic and sustainable issues. Feedstock procurement is analysed, considering the effect of chemical composition of raw material on fiberboard quality. Lignin represents the most important component for bonding ability. However, at the same time, the need to use a by-product, which may result in choosing a material with less lignin and more hemicelluloses, will worsen dimensional stability, and therefore, a pre-treatment of lignocellulosic material may be necessary. Many pre-treatments have been studied and optimised in recent years. This paper analyses mechanical, chemical, hydrothermal and biological ones, and considers the pros and cons of each one of them. The choice of pre-treatment depends on which result is to be achieved. Some applications are considered to conclude the production chain. What emerges is that the application phase is not yet fully developed and scaling up from laboratory to the industrial stage is not yet achieved.

1. Introduction

Fiberboards are elements made of wood chips or other lignocellulosic fibers, usually joined by synthetic binders [1]. They can be classified as low, medium, and high-density fiberboards, and can be used for furniture, indoor and outdoor semi-structural application, insulation, and soundproofing.

Within the past thirty years, research in the field of fiberboards has focused on two main challenges: on the one hand, the need to reduce or even eliminate the amount of synthetic resins [2], and on the other, the attempt to protect forest resources by using by-products [3]. Both topics are part and parcel to the concept of sustainability, which is becoming increasingly important, and thus the main objective of many research applications.

The synthetic adhesives generally used in fiberboard production are mostly formaldehyde-based [4], manufactured from non-renewable resources. The International Agency for Research on Cancer has classified them as carcinogenic to humans, and includes them in the 1B category [1].

Many attempts have been carried out to study the behaviour of fiberboards by replacing synthetic adhesives with the bonding ability of lignin or other bio-based adhesives. This is generally achieved through the selective dissolution of cellulose fiber skins to form a matrix phase that bonds the cellulose fibers cores together [5]. In binderless board production waste lignocellulosic materials are renewable and recyclable, and there are no synthetic binders which make the product expensive [6]. Additionally, without adhesives no curing period is needed [6]. This shortens the production process, thus obtaining both economic and environmental advantages.

The manufacturing technique of fiberboards involves certain interconnected operations, starting from feedstock procurement up to their production and marketing. It is a chain in which initial choices influence results and expected results influence initial choices.

The production process can be divided into five general steps: refining of fibers, drying, blending fibers with resins, forming the resined material into a mat, and hot pressing [7]. Raw material selection should be considered as a first step and upstream of the production chain, as the properties of the final product depend mainly on the fiber chemical composition and the treatment adopted, both in terms of

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https://doi.org/10.1016/j.jobe.2021.102625
Received 5 March 2021; Received in revised form 19 April 2021; Accepted 24 April 2021

Abbreviations

MOE	Modulus of elasticity
MOR	Modulus of rupture
IB	Internal bond
TS	thickness swelling
WA	water absorption
IB	internal bond
Tr	pre-treatment temperature
Tp	pressing temperature
Pp	pressing pressure

pre-treatment and hot pressing. For instance, a low content of hemicellulose has been associated with improved dimensional stability [8,9], i.e., low WA and TS, as well as a high lignin content, lignin being a hydrophobic component [10].

Feedstock procurement must also refer to where fiberboards are to be produced by verifying issues such as abundance, availability, accessibility, easy transport and other costs. Prices for wood chips have increased up to 30% between 2006 and 2011 [11], thus leading to an increase in the use of alternative resources to the present day. Van Dam et al. [12] defined the costs to produce particleboard. They divide the total cost into raw material costs (glue and wood) and production costs (labour and energy). In this study, the former represents 66% of the total cost, divided into 34% for glue and 32% for wood resources. Hence, raw material resources represent more than half of the total cost. Therefore, economic consideration must first be carried out on material selection. Apart from adhesives, raw material has a considerable impact on costs. On account of this, many researchers focused their attention on the use of residual lignocellulosic biomass in the production of binderless fiberboards, thus making the final product both attractive and sustainable on the market [13].

Among lignocellulosic materials, the use of agricultural residues has the added benefit of permitting the disposal of problematic solid waste, which usually does not have any economic alternative [14]. Many fiber-based industries increased their interest in the replacement of wood fibers, due to the low cost of agricultural by-products available worldwide, although total costs depend on the location, quality, and harvesting costs such as chopping, baling or on-farm hauling of crops, among which baling is the most expensive [11,15].

Therefore, using residual materials to produce fiberboards is a cost-effective and environmental-friendly choice which also considers the waste management hierarchy. The Waste Framework Directive 2008/98/CE sets prevention as the foremost priority of waste management, followed by re-use and material recycling. Energy recovery must be considered only if re-use or recycling are not possible, while landfilling or burning should be avoided [16].

Within this framework, this paper represents an overview on binderless fiberboard manufacturing, and focuses the attention on feedstock procurement and fiberboard production techniques. The influence of the chemical composition on board properties are thoroughly analysed, as also the exploration of certain pre-treatments which solve a few drawbacks such as low dimensional stability. Hot pressing is the final step in the manufacturing process, together with a study of the influence of Tp, tp and Pp.

To conclude the production chain, certain applications have been considered, and the need to engage in further work in the field of binderless fiberboards is presented.

2. Feedstock procurement

Feedstock procurement is upstream in the production process and influences each of its stages. This means the resources to make the fiberboard, which include both the lignocellulosic material and the adhesive used.

Lignocellulosic materials make up the most abundant renewable biomaterials on earth, but their use is still not common [17]. Owing to their abundance, underutilization and low cost, they appear as one of the best alternatives that could be converted into value-added materials such as biofuel, chemicals, and bio-based materials like binderless fiberboards [18].

2.1. Effect of chemical composition on fiberboards

Lignocellulosic biomass consists of 35%–55% cellulose, 25%–40% hemicellulose, and 15%–25% lignin [Fig. 1] with a small percentage of extractives, protein, and ash [19].

Chemical composition is an important issue when producing binderless fiberboards, since the lack of binder makes the percentage of each component more effective on fiberboard properties. The chemical composition of raw material varies depending on the type of material and often also on the fiber sources considered.

Research into the field of binderless fiberboards largely explored the behaviour of different fiber sources. Referring to the classification proposed by Dahi H. [10], some of the fiber sources employed are: *fruit fibers* collected from the seed of the plant for which cellulosic content varies greatly, according to the plant type (e.g., acai fruit [20], cotton [21,22]); *leaf fibers* resulting from a cutting process that occur to enable plant growth and cultivation and belong to the agricultural residues category (e.g., banana [13], date palm [3], plantain [23]); *stem fibers*, collected from the surroundings of the plant's stem which are the highest consumed among plant fibers within the industrial application and lead to the production of fiberboards or particleboards of very good quality (e.g. hemp [24,25], kenaf [26–28], totora [29–31]); *stalk fibers* include trees trunk as well as annual plant stems that are left over in fields after harvesting (e.g. cereal straw [32], tree trunks [33]); *fruit shell/husk/hull fibers* collected from the protecting skin of the fruit from which fibers can be extracted and processed (e.g. coconut husk [12,34], rice husk [25], almond shell [35], walnut shell [7]).

Certain materials are more suitable to produce binderless boards thanks to their chemical composition. Table 1 shows the chemical composition found in the literature of materials used for board production.

Many studies show the influence of feedstock chemical composition in binderless fiberboards, and the impact of each component on the properties of the final product. First, it was found that a smaller amount of hemicellulose leads to a better resistance to water as *hemicelluloses* are hydrophilic and have a high absorption rate [36,37]. They are mainly responsible for moisture sorption and biological degradation [9]. However, the low hemicellulose content may lead to weak bonding strength as it helps in better fibers to fiber binding [2], thus forcing the use of synthetic resins to produce lignocellulosic fiberboards [38].

Lignin is the component that permits adhesion in wood structure, but it is also responsible for ultraviolet degradation [9]. More studies concerning the potential role of lignin in binderless fiberboard production are presented later. A high amount of cellulose and lignin, but low hemicellulose, is generally found in wood materials [Table 1]. Otherwise, non-wood materials contain a higher amount of hemicellulose which leads to poor dimensional stability [38].

Extractives help to solve this drawback but, at the same time, their evaporation during hot pressing may cause delamination [39]. Alvarez-López et al. [23] revealed how extractable components of non-wood materials play a critical role in the development of mechanical strength of self-bonded fiberboards. In this study, organic extractives decreased the reactivity of the surface of the fibers, resulting in less bonding ability and, consequently, less mechanical properties of fiberboards. On the contrary, water extractives serve as catalysts of fiber degradation reaction during thermo-compression, thus leading to the formation of covalent bonds and increasing the mechanical properties.

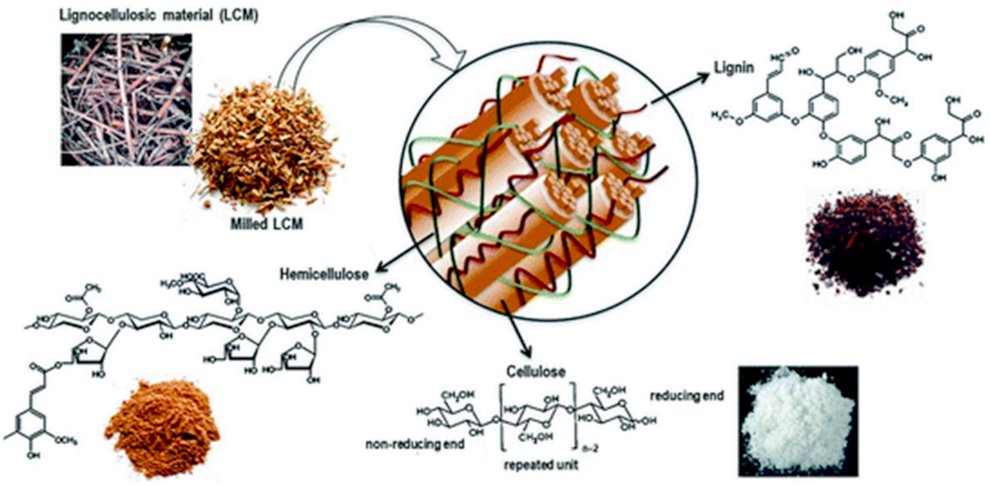

Fig. 1. Structure of lignocellulosic material [145].

Table 1
Summary of chemical composition of some raw materials.

Fibers	Cellulose	Hemicellulose	Lignin	Extractives	Ash	Reference
	%	%	%	%	%	
Softwood	32.1–38.2	16.4–28.5	32.1–38.2	10.7–14.2	0.4–0.9	Anglès et al. [9]
Vitis vinifera	43.6	19.1	24	13.3	3.7	Mancera et al. [64]
Date palm[a]	29.7–50.5	8.1–31.4	11.6	18.2	9.2–12.3	Saadaoui et al. [3]
Oil palm[a]	18.87–52.21	3.17–58.95	20.15–27.35	3.5–20.6	–	Hashim et al. [58]
Eucalyptus	43.4	18.8	28.5	–	–	Xiao et al. [98]
Miscanthus sinensis	42.6	21.1	19.9	4.7	0.4	Velásquez et al. [89]
Cynara cardunculus	49	24	18.3	9.7	5.4	Mancera et al. [70]
Arundo donax	43.1	21.9	22.4	9.3	3.8	Ramos et al. [74]
Totora	24.63–28.3	20.91–23.13	8.9–16.42	1.75–2.13	–	Hidalgo-Cordero et al. [29]
Sugarcane bagasse	37.73–55.2	16.8–27.38	20.03–25.3	17.03	0.59–1.1	Hoareau et al. [72] Ribeiro et al. [146]
Corn stalk	50.57	27.03	16	3.1	3.2	Theng et al. [42]
Rice straw	37.7	27.9	7.2	–	–	Theng et al. [106]
Wheat straw	39.7	30.6	17.7	5.2	7.72	Domínguez-Robles et al. [43]
Flax	70	16	2	–	–	Arévalo et al. [17]
Cotton	39.16–43.9	13.38	22.1–25.74	1.28	4.8–8.16	Fahmy et al. [21] Zhou et al. [22]
Kenaf	37.16	34.31	23.29	3.12	–	Xu et al. [69]
Sunflower	40.9	15.5	21.6	–	–	Klímek et al. [147]
Topinambur	30.9	12.6	16.3	–	–	
Cup plant	38.6	16.3	21.4	–	–	
Tomato stalk	43.11	7.91	12.29	–	–	Taha et al. [148]
Banana bunch	51.05	17.1	14.28	2.76	12.36	Quintana et al. [107]
Coconut husk	23–27	9–24	38.7–41.1	3.2–3.7	2.3–6.80	Araújo Junior et al. [34]
Walnut shell	25.4	21.1	49.1	–	3.6	Pirayesh et al. [80]
Macadamia shell	29.5	30	40.1	–	0.3	Wechsler [149]

[a] bark, leaves, frond, trunk.

To conclude, lignocellulosic materials generally contain a small percentage of ash. *Ash* content is an indicator of non-fibrous proportion and, even in low quantities, it has an abrasive effect on cutting tools and affects the machinability of the wood products [39].

2.2. Bonding ability of lignin-based adhesives

The fiberboard production process serves to activate the lignin bonding ability. In turn, lignin contained in the wood or non-wood resources serves to bond fiber itself without any added synthetic or natural adhesive. Nevertheless, in some cases there may be a need to use adhesives to improve mechanical strength and water resistance. If so, lignin can be used as a natural adhesive. The available literature on the subject is extensive. Various researchers studied fiberboard properties made with a percentage of lignin as a natural binder, obtaining considerable results. Anglès et al. [9] studied the effect of the addition of ligno-sulphonate and kraft lignin in fiberboard made by steam exploded softwood. They gained an increasing improvement for both mechanical properties and dimensional stability, especially with kraft lignin, with the increase of lignin amount from 0% to 20%. Velásquez et al. [8] set the maximum amount of added lignin at 20% for pre-treated M*iscanthus sinensis*. They observed a negative effect on physico-mechanical properties with higher quantities. Other researchers obtained equally good results by purifying kraft lignin with sulfuric acid [40] or by using alkaline lignin [41]. Theng et al. [42] also detected an improvement in mechanical properties of fiberboard made from thermomechanically processed corn residues by adding about 20% of purified kraft lignin, thus obtaining excellent results in mechanical strength. Domínguez-Robles et al. [43] used the lignin obtained from the pulping process of the wheat straw employed for fiberboard production itself, achieving flexural strength values always above the ones found in literature or even including commercial fiberboard.

Hence, lignin-based adhesives showed potentials for engineering applications and their abundance makes them even more attractive [44].

2.3. Raw materials

2.3.1. Economic and sustainability considerations

Research into the use of alternative lignocellulosic materials to produce fibreboards is still dynamic and different fiber sources have been studied over the years. Thus, some researchers focused on *forestry residues*, consisting of branches, leaves, bark and other portions of hardwood and softwood [9,45,46]. Some others tried to value *agricultural residues*, including straws or husks mostly left on field after harvests and used for fodder and landfill material or burned in many places [47,48]. Other studies focusing on *industrial by-product*, coming from sawing or milling of primary or secondary wood/non-wood processing, are also available. Fig. 2 shows a schematic representation of the categories defined in this article as alternative lignocellulosic biomass resources.

It has been estimated that the use of forestry and agricultural residues, as well as industrial by-products, is a cost effective and sustainable choice, since they are offshoots of other operations [49]. Especially agricultural and forestry residues are commonly associated with low costs, and low greenhouse gas emissions [50,51]. However, it seems difficult to evaluate their economic potential due to the lack of output and price data [49,52]. Some studies investigating the price of alternative lignocellulosic feedstock are available, although mainly focusing on the use as alternative biofuel sources. As an example, Perlack et al. [53] analysed the cost of corn stover residues for further processing by considering the costs for collecting, handling, and hauling corn stover by baling system, as well as delivering costs to an ethanol conversion facility. They accounted for 90% of total delivered costs for transportation, collection and baling, and farmer payment, and this estimated a significant ethanol cost reduction by using agricultural residues. Many authors indicated the cost of transportation as one of the main restrictions for the potential use of residues [49,50,53,54]. However, harvest costs may significantly lower the total cost, such as in the case of sugarcane bagasse for which the harvesting and delivering costs are associated with the primary sugar crop, and so considered almost zero [52].

As a result, the price of such feedstocks can vary a lot, depending mainly on the different incidence that the various harvest, transport, and hauling factors can have on the total cost, as well as location. Carriquiry et al. [51] indicated a very wide range, varying from 19$ to 84$ per tonne delivered. This is due to the different local conditions and could significantly vary a lot across different studies.

2.3.2. Forestry and agricultural residues

Research paid particular attention to oil and date palm residues proceeding from plantations, which are not used up completely and have a great potential to be converted into value-added products, thanks to their availability [55,56]. Palm trunk is considered as a renewable and sustainable natural resource and it has been utilized as a cellulosic raw material in the production of panel products including particleboard, medium density fiberboard (MDF), cement bonded particleboard, blockboard, plywood, and in the making of binderless board [57]. Hence, oil and date palm have a great potential in the development of binderless fiberboard and many different by-products of this plant can be used for this purpose such as core-parts, mid-parts, fronds, bark, and leaves [3,58], although dimensional stability remains the biggest drawback.

Within agricultural residues, there has been in recent years an increasing interest in cereals as raw material for fiberboard production. Cereal crops are worldwide available and harvesting residues may be an important resource for fiberboard production. Among the cereals, wheat accounted for 34% of total production [59]. Available wheat residues are estimated at 875×10^6 Mg worldwide in 2001 [59]. Research shows that wheat straw is one of the most suitable agricultural residues for producing pulp and paper [60], thus constituting a viable alternative for fiberboards. Such is the example of Domínguez-Robles et al. [1] who developed fully biobased lignocellulosic fiberboards from wheat straw fibers. The wheat straw biomass was refined by enzymatic pre-treatment and mechanical pre-treatment and the results were compared. They obtained fiberboards of good quality for both mechanical and enzymatic pre-treatment, despite obtaining a lower TS than commercial fiberboards.

Analogously to wheat, rice is one of the most important agricultural products in the world, both in terms of food and production volume. Rice straw is estimated at 890×10^6 Mg worldwide in 2001 [59]. Most straw is used for landfilling or is burned, resulting in environmental issues [61]. In binderless board production, it was found that rice straw silica content contributed to water resistance while the wax-like substances negatively affected self-bonding [62].

As an abundant agricultural waste product, *Vitis vinifera* prunings have also been used for fiberboard manufacture. After the pruning season, a large quantity of lignocellulosic material remains in the fields. The average pruning yield per hectare is about five tons [63]. Part of the pruning waste is used as fuel, but large quantities remain unused in the fields, thus increasing the risks of infestations and fire. Moreover, its chemical composition revealed medium cellulose and high lignin percentages that suggested its suitability as an alternative source of wood fibers used in fiberboard production [63–66].

Kenaf has attracted special interest in fiberboard production too, especially for its rapid growth: it can reach a height of 3.6–4.2 m during the growing season [67]. The stalk contains two types of fiber: an outer bast and an inner core. Kenaf fibers have gained popularity mainly for use in automotive application like head restrains, back pads and seat

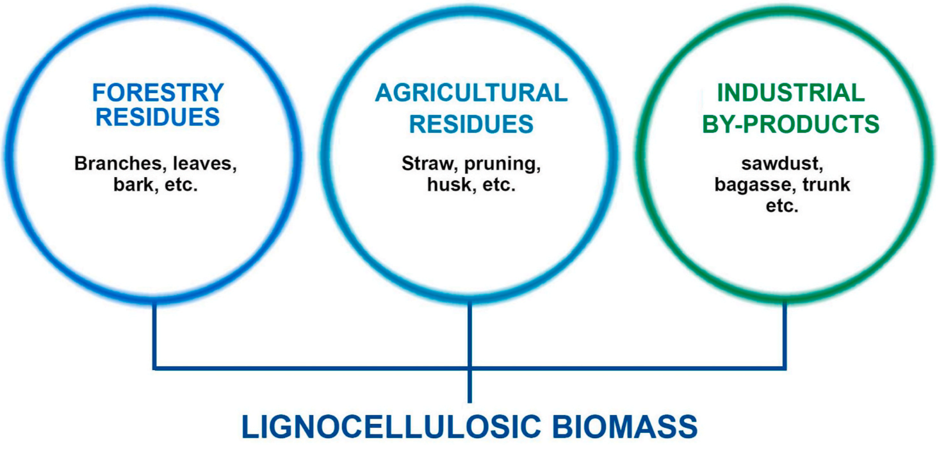

Fig. 2. Schematic summary of lignocellulosic biomass resources for fiberboard manufacturing.

bottoms [68], but the core remains underused and often discarded. Great attention has been devoted to the properties of fiberboards made by this natural fiber also for construction applications, especially for its use as insulation material. For instance, Xu et al. [69] produced low-density fiberboard made from kenaf core, obtaining a thermal conductivity quite similar to the insulation materials generally used in construction like rock wool, and also showing good mechanical properties and dimensional stability.

The abundance and availability of the material seem to be the necessary conditions for its feasible use in panel production. In this instance, *Cynara cardunculus* represents a viable alternative. The plant is available in the Mediterranean area and generally cultivated for nutritional purposes. The stalk could be used as a source in fiberboard production, thus helping to halt deforestation. Mancera et al. [70] used *Cynara cardunculus* to produce binderless fiberboard. They obtained boards of good quality which can be used for interior applications, even if their lower lignin and higher ash content led to poorer results compared to other materials.

2.3.3. Industrial by-products

Within industrial by-product, bagasse is probably one of the oldest non-wood resources used for fiberboard production. It is a by-product of sugarcane processing and it is considered one of the most promising non-wood lignocellulosic raw materials, since large quantities of this waste are still left unused or burned [71]. Sugarcane is an annual plant native to Latin America, but its cultivation has been imported into Europe for centuries. Sugarcane has a full cycle in 12 months, whereas trees take years to grow. Its rapid growth cycle makes it a more available resource compared to wood. Besides, the stockpiling of this material may cause spontaneous combustion of the stored bagasse, representing a serious environmental risk [72]. Based on Table 2, the high density fiberboards (HDF) manufactured by steam-injection pressing of sugarcane bagasse showed a high quality in terms of TS, but a worse performance in terms of mechanical strength when compared to panels made from other materials.

2.3.4. Other alternative materials

As shown in Table 2, good results have also been obtained for binderless fiberboard made by *Arundo donax* [73,74], bamboo [75,76] or *Miscanthus sinensis* [77–79] which showed a great adaptability to different climatic conditions, as well as rapid growth, low agronomic input, low production costs. There is an increasing interest in cane-type plants such as *Arundo Donax* and bamboo. They normally have a higher percentage of lignin which is expected to have a positive effect on bonding strength and dimensional stability, as well as having a very high-speed growth when compared to wood resources.

The lignin percentage is even higher in fruit shells. Some interesting results have been held with walnut or almond shell. Thanks to its characteristic of being hydrophobic, lignin could represent a way to overcome one of the major problems arising from the lack of adhesive, though in many cases TS still needs to be improved [35,80]. Shells could play an important role in the manufacture of value-added wood-based panels, and it may be an efficient use of them. In this concern, Hidayat et al. [81] studied the potential of the deoiled seed cake of *Jatropha curcas,* proceeding by the mechanical pressing of the seeds (shell and kernel) followed by deoiling with hexane for the preparation of binderless boards, but better results were obtained with very high Pp, thus making the production very energy consuming. Further studies must be developed to improve fiberboard properties and to make the production less energy intensive.

Table 2 summarises the results achieved in research by presenting mechanical and physical properties of fiberboards made by some of the

Table 2
Summary of mechanical and physical properties of fiberboards found in literature.

	Fiber resource	Pre-treatment	Density	MOR	MOE	IB	TS[a]	WA[a]	Reference
			g/cm^3	MPa	MPa	MPa	%	%	
Forestry/Agricultural residues	Date palm[b]	milling	1.000	6.1–12.9	430–1257	0.02–0.14	230–150	300–100	[3]
	Oil palm[b]	milling	0.800	2–14	–	0.05–0.70	100–20	140–50	[58]
		milling	0.800	1.50–12	–	0.05–0.55	80–20	130–70	[132]
		milling	0.800	5–25	–	0.50–1.10	50–40	110–80	[86]
		milling	0.800	3–6	–	0.20–0.40	65–20	110–65	[115]
		hydrothermal pre-treatment	0.800	3.92–8.76	–	0.17–1.10	111–23	211–72	[57]
	Wheat straw	enzymatic refining	1.100	95	5000	1.5	50	140	[1]
		mechanical refining	1.100	100	6500	1.6	40	140	[1]
		chemical pre-treatment	0.800	7–27	1800–4500	0.20–0.80	190–70	275–100	[97]
	Rice straw	milling	0.800	–	–	0.02–0.17	40–15	100–70	[61]
		extrusion	1.400	27.90–50.30	4200–7700	–	35–10	32–10	[106]
	Vitis Vinifera	steam-explosion	1.400	25–55	4000–7000	0.17–1.20	9–2	13–5	[63,65]
	Cotton	milling	1.200	26–78	–	–	24–10	36–26	[21]
	Cynara cardunculus	steam-explosion/ grinding	1.300	20–55	2527–7123	0.20–1.30	39–4	53–8	[70]
Industrial by-products	Sugarcane bagasse	steam-injection pressing	0.800	1–6	400–800	0.05–0.20	21–7	–	[71]
Other materials	*Arundo donax*	steam-explosion/ grinding/ final heat treatment	1.200	21–42	3254–6249	2.10–3.25	11.13–5.84	23.81–8.99	[74]
	Bamboo	Milling/steam-explosion	0.800	3–16	–	0.20–0.80	50–5	100–20	[75]
	Miscanthus sinensis	steam-explosion/ grinding	1.100	25–45	3090–4630	1.07–2.80	38–8	50–15	[77,89]
	Almond shell	shredding	1.300	7.64–14.01	1362–2295	0.59–1.27	33–18	40–17	[35]
	Jatropha curcas seed cake	oil extraction	–	22.8	5100	–	19	74	[81]
	Commercial HDF		780–893	41.70–42.25	2670–4299	0.39–0.47	13–66	45–80	[43,73,83, 84]

[a] Value at 24h
[b] Bark, leaves, frond, trunk.

aforementioned materials. We compared the results obtained mainly in the field of HDF. Mechanical properties and dimensional stability showed competitiveness compared to commercial HDF especially when the fibers were properly pre-treated, although it seems that they lead to a slightly higher density [43,73,82–84] and this may be a starting point for future research and improvements.

3. Fiberboard production methods

In fiberboard manufacturing, two methods have been developed: wet-forming process, and dry-forming process. In wet-forming process less binders, or no binders at all, are needed [38], due to the hydrogen bonds formation and adhesive behaviour of lignin which occur during heating and drying processes [38]. In dry-forming processes the pre-dried fibers are blended with binder, the mixture is distributed into a mat, and then hot-pressed [1].

As mentioned above, a pre-treatment may improve fiberboard properties in many cases, especially for dimensional stability. It is well known that an increase in the surface area of cellulose fibres led to a higher performance in binderless fiberboards, as well as the hydrolysis of hemicellulose improving the resistance to water [17].

3.1. *Pre-treatments*

Different treatments can be carried out in binderless fiberboard production [Fig. 3]. Raw materials are generally subjected to a prior mechanical treatment (sawing, chipping, shredding, grinding, and milling) before any other treatment, through which particle size is reduced. Many studies showed the importance of particle size, which may have a great effect on board properties. Finer particles improve water resistance of such panels, while bonding strength is enhanced by larger particles [85,86]. Overall, lignocellulosic biomass is always further treated after milling, but in certain cases a good quality binderless fiberboard can be obtained without any other treatment. This paper focuses on mechanical treatments, chemical treatments (acid or alkaline hydrolysis, oxidation agents), hydrothermal treatments (thermomechanical pulping and steam explosion), biological treatment (microbiological and enzymatic) [Fig. 3].

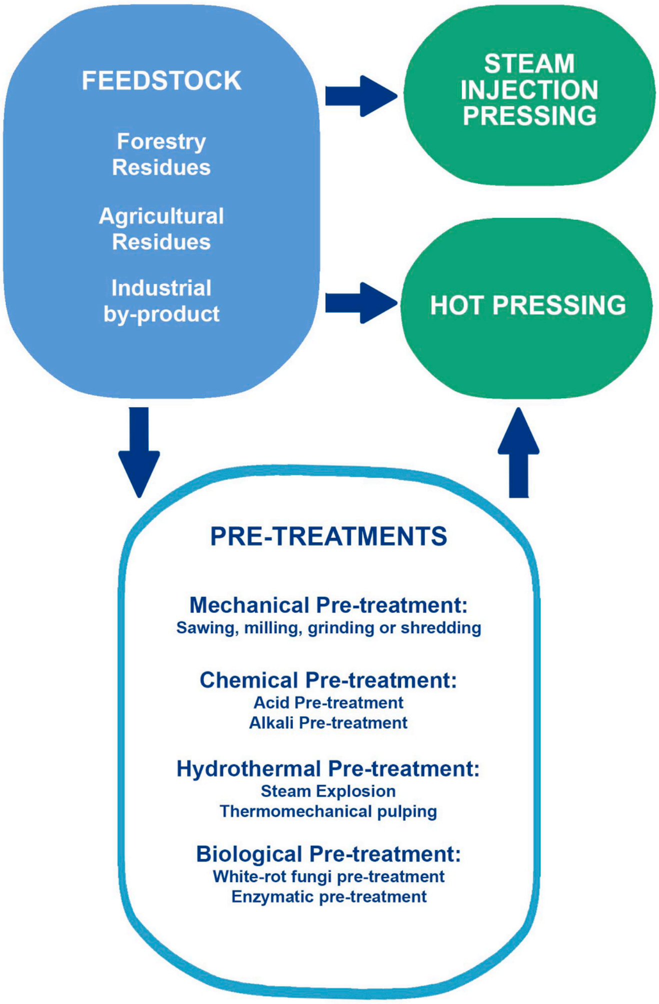

Fig. 3. Schematic summary of different pre-treatments of lignocellulosic biomass for fiberboard manufacturing.

3.1.1. *Mechanical pre-treatment*

Mechanical pre-treatment increases the total accessible surface area, thus improving the accessibility of constituents and leading to better bonding strength [87]. Milling reduces the crystallinity of cellulose, the substrate particle size, and the degree of polymerization [88]. Most researchers combine mechanical pre-treatment with other methods, since it does not modify the chemical composition or the cell-wall structure of the raw material. When mechanical pre-treatment is the only method carried out, fiberboard properties are deeply dependent on chemical composition and it is more difficult to overcome the main drawbacks such as high hydrophilicity. Hidalgo-Cordero et al. [29] clearly showed the dependency between chemical composition and the properties of the panels. They observed very different results for fiberboards made from shredded fibers of totora stem, pith and rind, as they have different amounts of the main chemical components, i.e. cellulose, hemicellulose, and lignin.

As a result, the general trend is to use mechanical treatments either before or after other treatments, thus rendering mechanical treatment more advantageous. For instance, grinding proved to be particularly effective in increasing IB, if carried out on steam-exploded pulp [89]. However, fine grinding requires a large amount of energy. Therefore, further research should be devoted to maintaining a balance between efficiency improvement and cost.

3.1.2. *Chemical pre-treatments*

Chemical pre-treatments can be at low, neutral or high pH [90]. Typically, low pH pre-treatments (acid pre-treatment) remove most of the hemicellulose and a small portion of biomass lignin. Neutral pH pre-treatments (controlled pH) remove much of the hemicellulose but leave most of the cellulose and lignin intact [90]. High pH pre-treatments (alkali pre-treatments) remove a large fraction of lignin and some hemicelluloses [90].

In *acid pre-treatment* method, acids are used as catalysts to hydrolyse lignocellulosic components [56]. Common chemicals used are sulfuric acid, hydrochloric acid, or acetic acid [91]. Acid pre-treatment results in the disruption of van der Waals forces, hydrogen bonds and covalent bonds that hold together the biomass components, which consequently cause the solubilization of hemicellulose [92]. Acid pre-treatments can be carried out with both concentrated and diluted acids. The use of concentrated acid may cause corrosion of the equipment, high consumption of the acid, toxicity to the environment, and energy demand for acid recovery [92]. Dilute acid hydrolysis presents the advantage of lower acid consumption but in return, a higher temperature is required [56]. This can be performed in combination with other treatments to reach a further hydrolysis of hemicellulose. It has been proven that the addition of sulfuric acid during steam explosion treatment improves the solubilization effect as well as a wider redistribution of lignin, thus leading to a better resistance to water [9,88]. However, fiber length decreases when acid catalyst is used, thus leading to poorer mechanical strength [88].

Alkaline pre-treatment of lignocellulosics originates from soda pulping patented in 1854 [93]. The main alkaline reagents are sodium hydroxide, potassium hydroxide, aqueous ammonia, calcium hydroxide and oxidation reagents. This pre-treatment consists in adding a base to biomass and leads to an increase of internal surface area by a swelling of the cell wall, a decrease of polymerization degree and crystallinity, a destruction of links between lignin and other polymers, and lignin breakdown [94]. Compared to other chemical pre-treatment technologies, alkaline hydrolysis can be conducted at a lower temperature and pressure, causing less sugar degradation than acid pre-treatment, but the reaction times take several hours, days, or even weeks for softwood. Actually, the yield of alkaline pre-treatment depends on the lignin amount: the lower the lignin content, the better the treatment effect. In fact, it turns out that the alkaline pre-treatment is more effective on hardwoods and agricultural residues than on softwoods, as softwoods normally have a lignin content higher than hardwoods, at around 26% [94].

Sodium hydroxide proved to be effective to improve digestibility [95]. Ahmad et al. [96] implemented an alkaline treatment with sodium hydroxide on rattan furniture waste to produce binderless particleboards. The treatment hugely increased MOR and IB values and proved to facilitate hydrolysing hemicellulose, improving dimensional stability too. However, sodium discharge in the process effluent is difficult to be recycled and limits its application on the pilot scale [96].

Moreover, potassium hydroxide could be used but it shows high chemical loading, a high cost when applied on a large scale and causes environmental pollution [95].

Calcium hydroxide might be better as it is low cost, safer, more environmental-friendly, and can be easily recovered. Nevertheless, as a weak alkali, it may not improve biomass digestion significantly [95].

Lastly, an oxidation agent, such as Fenton's reagent, is currently being studied [38]. Fenton's reagent is composed of ferrous chloride and hydrogen peroxide which could activate the fiber surface and facilitate the adhesive bonding between fibers. Halvarsson et al. [97] produced binderless fiberboard from wheat straw by using oxidative activation of wheat straw fibers, performed by adding hydrogen peroxide during defibration. They obtained an improvement in fiberboard properties by increasing the added hydrogen peroxide from 2.5% to 4%. However, it is believed to have a harmful effect because of the instability of board quality and insufficient mechanical strength obtained [97]. Further studies and improvements are needed.

3.1.3. Hydrothermal pre-treatments

Hydrothermal pre-treatments are wet treatments, generally developed at high temperature. Several papers have been published on hydrothermal treatments. They generally lead to a more condensed lignin that has a high average molecular weight and contribute to the self-bonding formation of lignocellulosic materials [98].

Steam treatments and thermomechanical pulping are analysed below.

The first can be carried out before forming a mattress (steam-explosion pre-treatment), during hot pressing (steam injection pressing), and after hot pressing (post-treatment steaming) [99]. Among these, the *steam-explosion pre-treatment* is considered to be particularly effective in improving all fiberboard properties. In steam-explosion, biomass is subjected to high pressure steam followed by a sudden decompression in a batch reactor [Fig. 4].

When the pressure is released, the steam expands within the lignocellulosic matrix, causing the separation of individual fibers and the disruption of cell wall structure [100].

This method is industrially practiced in the Masonite process [41]. The lignocellulosic biomass results in a pulp in which most of hemicellulose is hydrolysed and which can be hot pressed to produce fiberboard without using synthetic binders, thanks to the creation of a good bonding strength between fibers [55,89].

Temperature and pressure are two important parameters of steam explosion pre-treatment. The Tr generally ranges between 160 and 260 °C with the corresponding saturated steam pressure of 5–50 bar [100].

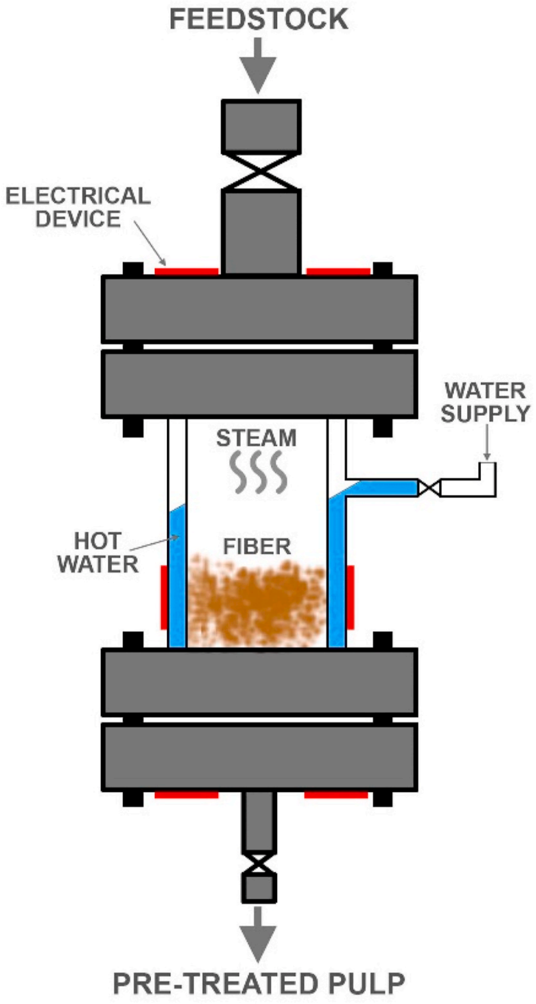

Fig. 4. Schematic representation of steam-explosion reactor (Adapted from Ref. [107]).

Tr and tr are combined in the severity factor, which is a commonly used parameter in steam pre-treatment and defined as follows:

$$R_0 = \int_0^{tr} \exp\left[\frac{Tr - 100}{14.75}\right] dt$$

[101] As a combination of Tr and tr, severity factor has a great influence on fiberboard properties. Some studies demonstrated that boards made from fibers exploded under too high and severe conditions (high Tr or long tr) becoming brittle and showing low MOR and MOE [9,77]. An increase in the pre-treatment severity can improve the physical properties of the panels but there is an optimum value with no further improvement.

The advantages of a steam-explosion pre-treatment are reported to be as follows: recovery of all constitutive wood components without the destructive degradation components, lower environmental impact, lower capital investment, higher potential for energy efficiency, and less hazardous process chemicals and conditions [102].

However, it has been observed that steam explosion has a high effectiveness for pre-treatment of agricultural residues and hardwood, but it is less effective for softwood. In such cases, using an acid catalyst during steam-explosion process becomes significant [94].

It is appropriate to also mention the *thermomechanical process*. In a thermomechanical treatment the pulp is made by heating the chips at Tr above 100 °C and by mechanically separating the fibers in a pressurized refiner. Thermomechanical pulping process has become one of the major

Table 3
Effect of pre-treatments on fiberboards properties, pros, and cons.

Pre-treatments		Effect on fiber and board properties	Pre-treatment pros and cons
Mechanical pre-treatment	Sawing, milling, grinding, shredding	This increases the total accessible surface area, improving the accessibility of constituent [87], increasing the bulk density and porosity [88], and IB [89]. It is generally combined with other treatments.	This may require a large amount of energy [47].
Chemical pre-treatment	Acid pre-treatment	This removes most of the hemicellulose and a small portion of lignin [90,92]. It has been combined with steam explosion pretreatment [9], leading to improved WA and TS.	Concentrated acid may cause corrosion of the equipment, high consumption of the acid, toxicity to the environment, and energy demand for acid recovery [92]. Diluted acid requires a higher temperature [56].
	Alkaline pre-treatment	This removes a large fraction of lignin and some hemicelluloses [90]. It led to an improvement in all fiberboard properties, especially in TS [96].	This can be conducted at a lower temperature and pressure, causing less sugar degradation [94]. In turn, reaction times take several hours or days [91].
	Oxidation agents	This facilitates the adhesive bonding between fibers [38], thus improving mechanical properties, WA and TS [97].	This is believed to have a harmful effect because of the instability of board quality and insufficient mechanical strength obtained [38].
Hydrothermal pre-treatment	Steam explosion	This is known for being particularly effective for improving all fiberboard properties [99], especially dimensional stability, if carried out in a certain severity factor range [77].	This permits the recovery of all constitutive wood components without the destructive degradation components, lower environmental impact, lower capital investment, more potential for energy efficiency, less hazardous process chemicals and conditions [102]. In turn, Energy consumption must be considered.
Biological pre-treatment	Microbiological pre-treatment	This can selectively metabolize low molecular weight lignin and hemicellulose while leaving cellulose unaffected [109]. It has shown a good effect on mechanical properties of fiberboards [47,109].	This needs a long residence time (10–14 days), extremely precise growth conditions and the need for a large space [109].
	Enzymatic pre-treatment with laccase	This removes the lignin molecule from cellulosic material and helps the polymerization of lignin [111]. It can improve fiberboard properties, both mechanical and physical [38,112,113]	This involves a mild reaction condition, less by-products and being environmentally friendly [38]. In turn, the high cost of laccase makes it less feasible [114]

processes for the manufacturing of ultra-high yield pulps, accounting for 20% of the world pulp production together with other mechanical pulps [103]. Refining pressure and time are the two main manufacturing parameters which consequently influence almost all properties of the panels [104]. Gao et al. [105] compared thermomechanical treatment and grinding process. They showed that the thermomechanical treatment of bark via refining lowers the required Tp and shortens the tp needed for manufacturing binderless bark fiberboard.

Extrusion is a promising thermomechanical pre-treatment for use in biomass conversion because it is cheap, the monitoring of temperature and screw speed is good, and it has high shear and excellent processing capacities [106]. Twin-screw extruder used for the pre-treatment of fibers from agricultural waste for composite production is cheaper than the other fiber-removing technologies, and it does not change the chemical and thermal properties of the raw biomass [106].

3.1.4. Biological pre-treatments

Biological pre-treatment is an eco-friendly process that requires low energy input, low disposal costs, and milder operating conditions against other pre-treatment strategies [108]. This usually refers to the deconstruction of lignin structures in the cell wall using microbes or enzymes as catalysts.

Within biological pre-treatments, fungi have become popular as sources of commercial plant cell wall degrading enzymes [94]. *White-rot fungi* can selectively metabolize low molecular weight lignin and hemicellulose, while leaving cellulose relatively unaffected [109]. This results in increasing the mechanical properties of binderless fiberboards since it increases the number of hydroxyl groups, crystallinity, polysaccharide, and laccase content [47,110]. However, the rate of biological pre-treatment is very slow for industrial purposes. Fungi treatment needs long residence time (10–14 days) and precise growing conditions to be effective [109]. For these reasons there is no suitability for industrial application at present.

Enzymatic treatment can also be included into biological pre-treatments. This often involves a mild reaction condition, less by-products, and a low environmental impact. Laccase, as an oxido-reductase agent, removes the lignin molecule from the cellulosic material and helps the polymerization of lignin through free radical reactions [111]. During the fiber treatments, laccase barely penetrated into fibers and mainly oxidized the lignin on the surface [108]. As a result, free radicals are generated on the fiber surface, acting as potential reactive sites for further cross-linking reactions in the production of fiberboards [38].

Laccase treatment can improve bonding properties, thanks to the surface modification, which occurs with the precipitation of lignin extractives on the surface [112]. Typically, laccase treatment led to an improvement in both mechanical and physical properties, although the concentration of laccase is less effective on MOR, MOE and IB [23,38]. Otherwise, the different concentrations largely affect thermal properties [113]. Fibers treated with a higher concentration showed higher thermal stability, probably due to the grafting reaction of low molecular weight components on the fiber, with the lignin [113]. However, the high cost of laccase made it difficult to be applied on a large scale [114].

Table 3 summarises and compares the effect of pre-treatments on fiber and board properties, as well as outlining the advantages and disadvantages of each pre-treatment, by considering results obtained in the field of binderless fiberboards.

3.2. Effect of pressure temperature (tp), time (tp) and pressure (pp)

Hot pressing is the last step for fiberboard manufacturing. A final heat treatment carried out after pressing to further improve binderless board properties, and especially TS and WA, has also been considered, but is still under study [74].

Fig. 5 shows a schematic representation of the hot press generally used when producing binderless fiberboards.

The main parameters that have to be taken into account when performing hot pressing are: Tp, tp, and Pp. If heated at high Tp, the lignin of the pre-treated biomass surrounds and bonds the fibers [2,9]. Moreover, the water loss produced during the hot pressing may contribute to the formation of covalent bonds between chemical components of lignocellulosic biomass [1]. Tp has a strong influence on binderless fiberboard properties, even more so when no binder is used, as well as tp and Pp. As the Tp and tp increase, MOR and IB are observed to increase

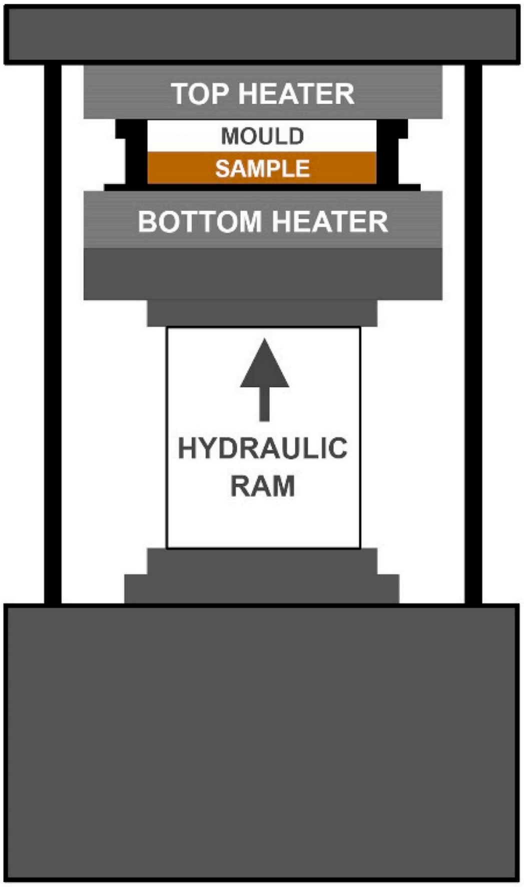

Fig. 5. Schematic representation of hot press generally used (Adapted from Ref. [81]).

and WA and TS of the fiberboards also improve, although not to the same extent as the mechanical properties [29,38,115].

Okuda et al. [26] studied chemical changes of kenaf core binderless fiberboard after hot pressing and its influence on fiberboard properties. Part of the lignin and hemicellulose decomposes during the hot-pressing process and this leads to a condensation reaction in lignin [26]. Boon et al. [116] indicated that the increasing in Tp, tp, and Pp, intensified the stability against moisture, mechanical strength, and the resistance of microorganism decay of the specimens. Nonaka et al. [117] investigated the effect of high Tp on binderless particleboards made by sugarcane bagasse and recycled wood chips. They found an improvement in mechanical properties and dimensional stability by using high Tp for both bagasse and wood chips.

It was found that normally Tp ranges from 110 °C to 230 °C. On the other hand, tp values varying among 10 and 50 min were found in literature. Most authors [9,70,83,89] divided the hot pressing into three steps, the first one of hot pressing, a breathing time, and a last shorter hot-pressing step. The three-steps in hot pressing help to prevent the formation of bubbles inside the specimens, which may compromise the mechanical properties of fiberboard [118].

Recently, steam injection has gained attention as a manufacturing process for fiberboards [119]. As mentioned, this can be considered as a combination of pre-treatment and hot pressing, as it consists of injecting steam directly into the material while it is being pressed in the mold. Thus, all fractions are formed during treatment and remain in the fibers [23], while in steam-explosion the degraded components are solubilized in water and removed after processing. Currently, there are some researchers studying steam injection process to produce low density fiberboards, especially with kenaf core [27,120]. They obtained binderless boards of relatively good quality, as they showed an improved IB and TS when compared with the hot pressing method [120].

4. Applications in construction and further work

4.1. Current applications

The construction industry makes extensive use of wood-based panels, which cause environmental problems both in terms of meeting the high demand for wood and emissions from the use of synthetic adhesives [121]. Binderless fiberboards represent a possible solution to both problems. On the one hand, the use of alternative fibres supports the concept of forest protection and on the other hand, the absence of formaldehyde emissions makes these panels particularly suitable for indoor use applications such as wall partitions, false ceilings, and furniture [2]. Besides, many studies investigated the feasibility of binderless boards for thermal insulation. Natural fibers generally have good thermal properties [122] and this has motivated many researchers to investigate their behaviour within this field. Some examples available in the literature reviewed the use of renewable materials such as sunflower [123], coconut husks and bagasse [124], cotton stalk fibers [22] eucalyptus leaves and wheat straw fibers [125], and hemp and corn residues [121]. A recent review [126] investigated the use of agro-waste for sustainable construction, also in considering binderless boards. In Table 4 we reported the thermal conductivity of fiberboards made by alternative natural materials, compared with commercial ones. Generally, a material can be considered as an insulator when its thermal conductivity does not exceed 0.07 W/mK [127]. Most alternative fiber sources showed good thermal properties, although common insulation material used in construction still exhibits better features. However, non-renewable resources used to manufacture insulating materials, like EPS or XPS, have more impact in terms of equivalent carbon emission than naturally derived ones [128].

4.2. Further work

Despite their potential, binderless fiberboards are struggling to break into the market of building materials. Literature results offer few application examples, and there are still many gaps, which make it difficult to have a clear idea of the competitiveness of these products on the market for construction materials. To begin with, WA and TS are a major concern and need further study and improvement [2]. In addition, there is almost a complete lack of economic information.

As a result, further studies could address the following points:

- Improving the physical characteristics of the binderless fiberboards.
- Finding new and viable ways for the real use of these sustainable boards.
- Studying the economic competitiveness on the construction materials market, considering the influence of the raw material and the manufacturing process on the product.
- Studying the environmental impact and benefits compared to current fiberboards.

Although further research is needed to fill this lack of information, the following studies may be important in reaching the goal:

4.2.1. Economic considerations

From a sustainability perspective, environmental and economic considerations should be the basis of decisions in the manufacturing of any product.

The literature focusing on binderless fiberboards largely refers to the cost-effectiveness when using alternative lignocellulosic resources to wood [20,31,32,43,49,73,129,130], and avoids the use of synthetic adhesives [34,57,119,131] as they contribute from 30% to 60% to the overall cost of the final product [12,132]. However, not many studies

Table 4

Summary of thermal conductivity of some fiberboards made by natural fiber sources compared with conventional insulation materials.

	Binderless fiberboard source	Density	Thermal conductivity	Reference
		kg/m^3	W/(mK)	
Forestry residues	Wood (pine, lauan)	450–630	0,151	[69]
	Eucalyptus leaves	211	0,045–0055	[125]
	Date palm	176–260	0,0475–0,0697	[150]
Agricultural residues	Corn cob[a]	513,0–455,7[b]	0,1569–0,1435[b]	[121]
	Rice husk	150–175	0,044–0056	[151]
	Wheat straw	150–250	0,0481–0,0521	[22]
	Cotton stalk	150–450	0,058–0082	[22,124]
	Hemp fiber	369–475	0,090–0108	[152]
	Kenaf	150–200	0,051–0058	[69]
	Sunflower fiber	358–687	0,082	[123]
	Coconut coir	300–611	0,047–0,1117	[126, 153]
	Sugarcane fiber	100–125	0,046–0049	[122]
Industrial by-products	Coconut husk and bagasse	250–350	0,046–0068	[124]
	Bagasse	90–140	0,047–0050	[154]
	hemp shiv[a]	176,4–186,1[b]	0,0785–0,0778[b]	[121]
Common insulation materials	Mineral wool (fiberglass and rockwool)	20–200	0,033–0045	[155]
	Expanded Polystyrene (EPS)	15–35	0,031–0038	[155]
	Extruded Polystyrene (XPS)	25–45	0,030–0040	[155]
	Sheep wool	10–25	0,038–0054	[155]
	Plaster/Wood shavings (25%)	1605	0,28	[156]

[a] with natural binder.

[b] These values are obtained from specimens at 23 °C and 50% relative humidity. They change in dry conditions.

have been found that rigorously analyse this issue in the production of binderless fiberboards.

As an example, Uitterhaegen et al. [133] presented some economic considerations concerning binderless coriander fiberboards made by using twin-screw extrusion process. In this case, the low cost of extrusion process, combined with the low cost of coriander straw, resulted in a low total cost which varied between 0.44 and 0.46 €/kg. This suggests a possible competitiveness of such products when considering that the price of commercial fibers (hardwood and softwood) is normally around 0,50 €/kg [133]. Nevertheless, further work is needed on this point. There is a need for an in-depth study of the economic advantage that can be derived from the aforementioned panels, which also considers the influence of the different treatments, as well as their environmental impact and sustainability.

4.2.2. *Sustainability considerations*

Nowadays, the environmental impact of the products is a major issue. The forest industry, associated with the production of current panels, is responsible for high energy consumption and environmental pollution [134], through the emission of formaldehyde and volatile organic compounds during the production, use, and later disposal of the panel product. Moreover, building sector plays an essential role in the global energy scenario, and its environmental impact could be reduced by introducing more sustainable materials [135,136]. For instance, in a recent review, Maraveas [157] deeply analysed the use of agro-waste in construction materials sector, showing how the use of these resources help to meet sustainability challenge.

Life cycle assessment (LCA) is a methodology that has proved to be the most appropriate for assessing the environmental impact of products [137], since it considers the entire life cycle, i.e. the sourcing of raw material, its processing, the manufacturing of the product and its marketing, up to the end of its life. Several studies, focusing on environmental assessment of wood-based panels, have pointed out that the high environmental impact is mainly due to the use and processing of wood resources, and the formaldehyde emissions related to the use of synthetic adhesives [138–142].

Nevertheless, to the best of our knowledge, very few studies concerning green binderless fiberboards are available at the present. As an example, González-Garcia et al. [143] carried out an environmental assessment of green hardboards made by a two-component adhesive based on a laccase activated system, and demonstrated their industrial viability. Also, Freire et al. [144] evaluated the environmental impact of green MDF and HDF and compared their results with commercial urea-formaldehyde bonded boards. They demonstrated the superiority of these boards in most of the impact categories (climate change, acidification, land use, particulate matter, water depletion, and fresh water eutrophication).

Aforementioned research provides useful information on the potential role of binderless fiberboards in the building sector but the literature is still too limited to draw appropriate conclusions. For this reason, further studies considering the environmental impact and the principles of circular economy are needed.

5. Conclusions

To sum up, this review may offer a useful approach to determine all the necessary steps to make binderless fiberboard attractive for the market, as regards economy and sustainability related issues.

Circular economy principles have been actively introduced into the European market, promoting product recyclability, and saving the use of raw materials. Within this framework, the field of binderless fiberboards made by unconventional raw materials may represent a viable alternative to the current wood market.

The reviewed literature offers many positive perspectives and the results of this study can be summarised as follow:

- Most of the results obtained for binderless fiberboards showed a great competitiveness with commercial ones in terms of mechanical, physical, and thermal properties.
- Raw material selection has a great impact from several points of view, including economics and sustainability, as well as having an influence on the board's properties due to the chemical composition.
- Agricultural residues and industrial by-products represent a viable alternative for binderless fiberboards production.
- Adequate pre-treatment and hot pressing decrease the influence of the chemical composition of the raw material in the production of binderless fiberboards and ensure good fiber bonding without using adhesives.

On the other hand, despite the progress in the production of binderless fiberboard, its commercialization is still a challenge. The difficulty seems to lie in scaling up production from the laboratory to the field of industry. Many drawbacks need to be addressed:

- Water resistance and dimensional stability are still a challenge in many cases and depend mainly on fiber pre-treatment
- There is no systematic analysis of the cost of these boards with reference to the entire production process and the various alternatives in respect of pre-treatment
- Only a few applications are available and further studies are needed.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We would like to acknowledge the “Universitat Rovira i Virgili” (URV) for providing the research infrastructure.

Federica Vitrone acknowledges support from URV through a PhD scholarship.

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