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The influence of thickness and final heat treatment on mechanical properties and dimensional stability of binderless fiberboards from steam exploded *Arundo donax* L

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ABSTRACT

The lack of petroleum-based adhesives in binderless fiberboards (BF) generally lead to low dimensional stability and poor mechanical properties. The steam explosion (STEX) pre-treatment can help to improve both physical and mechanical properties, but in many cases dimensional stability is still a challenge and depend on the characteristics of the raw material and the STEX conditions. This study investigates the effect of thickness and final heat treatment (FHT) on physico-mechanical properties of binderless fiberboards made by Arundo donax L. The manufacturing process involved the STEX pre-treatment at a temperature (Tr) of 190 °C and retention time (tr) of 9.5 min, the hot-pressing (HP) at 7.5 MPa with a three-steps procedure, and the FHT carried out at 180 °C for two different times (tc), 0.5 h and 1 h. Three samples for each condition were made and three different thickness levels were investigated, i.e. 3 mm, 6 mm, and 9 mm. The results obtained were compared with three reference samples made for each thickness for which no FHT was carried out. The results of modulus of elasticity (MOE), modulus of rupture (MOR), internal bond (IB), thickness swelling (TS), and water absorption (WA) showed that FHT had a great influence on dimensional stability but did not lead to any improvement in mechanical performance. On the contrary, the thickness did not have any influence for TS and WA, but a small change in MOE, MOR, and IB was detected. These findings may help to improve the BF manufacturing process and to optimize the results for Arundo donax L.

1. Introduction

Binderless fiberboards (BF) are bio-sourced boards made by taking advantage from the "self-cementing" ability of lignocellulosic raw material, enhanced during HP [1–3]. BF are a more sustainable alternative to conventional fiberboards which normally contain a certain amount of petroleum-derived adhesives, mainly urea-formaldehyde (UF) [4]. UF adhesives are widely used in the wood-based industry because of their water resistance, low curing temperature, short press time, and hardness [4,5]. Although urea is considered a good formaldehyde catcher, UF adhesives and their emissions and volatile organic compounds (VOC) can compromise air quality being toxic for human health, especially when used as construction and indoor decorative and furniture materials [5,6]. This fact, combined with the need to think about the disposal and recyclability of the products, led many researchers to focus on BF. Recently, many studies focusing on the properties of BF have been carried out by using oil palm [7], eucalyptus [8], bagasse [9], wheat straw [10–12], rice straw [13,14], coconut fibers or husks [15,16], giant reed [17], etc.

As to BF, one of the biggest issues concerns dimensional stability, i.e. water absorption (WA) and thickness swelling (TS). BF use to have a high water uptake and low dimensional stability, and this reduces their possible applications [2,18]. The lack of fossil adhesives, that are generally hydrophobic, make BF more vulnerable. Indeed lignocellulosic materials have amphiphilic properties [19] and various researchers associated the low dimensional stability to a high amount of hemicelluloses [2,18,20]. Thus, the hydrolyzation of hemicelluloses is a way to solve this drawback, together

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with the softening of the lignin which then can cover the cellulose fibers creating a more impermeable layer thanks to its hydrophobic properties [21].

This can be achieved by subjecting the raw material to a pre-treatment that dissolves the hemicelluloses without destroying the structure of cellulose, which is the skeleton of BF. There are various techniques that aim to obtain good quality products, such as enzymatic pre-treatment, chemical pre-treatment, and steam-explosion pre-treatment [18]. Among these, excellent results have been obtained with STEX from several point of view, including dimensional stability. For example, Tupiciauskas et al. [22] studied the influence of STEX on binderless boards from hemp shives and wheat straw. They detected a great positive impact of STEX on water uptake when the severity factor ($logR_0$) is above 3.83, which means in their case a Tr of 220 °C and a tr of 2 min. Comparing these results with those obtained by Ferrandez-Villena et al. [23], the positive effect of STEX is clear: in the first research the average TS value obtained is 9%, while in the second study, where no pre-treatment is carried out other than shredding, the minimum TS was about 40%.

Although much progress has been made in research, dimensional stability is still a challenge in many cases and often depends on the characteristics of the raw material used and the conditions of the pre-treatment. Bearing in mind this framework, the objective of the present study is to verify whether adding a further step to the production process of BF, namely the FHT carried out after STEX and HP, can achieve an improvement in the parameters of dimensional stability, despite using a lower severity in STEX pre-treatment. The aim is to investigate a possible relationship between FHT and physico-mechanical properties and dimensional stability, and, at the same time, to study the influence of thickness on the same properties.

2. Materials and methods

2.1. Raw material preparation

The Arundo donax L. (AD) used in this work was provided by Cañizos Albatera Sl and came from Ribarroja de Turia, Valencia (Spain). The raw material was shredded to obtain chips of about 5 mm in length to be pre-treated by STEX.

STEX was carried out in a 16 L cylindrical reactor where about 750 g of chopped reeds were introduced into, and then immersed in saturated steam. The Tr used in this study was 190 °C, while the tr was 9.5 min, corresponded to a $logR_0$ of 3.62. The R_0, which relates Tr and tr, was calculated as follows:

$$R_0 = \int_{tr(\min)}^{0} \exp\left[\frac{Tr(^\circ C) - 100}{14.75}\right] dt$$

[24] After the tr was passed, the supplied valve of the STEX equipment was opened and the steam pressure released, causing the disruption of fibers. The exploded AD was then rinsed in a washing trolley to remove the hydrolyzed hemicellulose. After being left to dry in ambient conditions for about 3 weeks, the exploded AD was ground to pass 4 mm sieve.

2.2. Manufacturing process of binderless fiberboards

The ground pulp was used to fabricate fiberboards. It was first cold pressed to form a mat of 150x50 mm at three different thicknesses: 3 mm, 6 mm, and 9 mm (named FB, FBD, and FBT, respectively). Nine specimens were fabricated for each thickness, and they were conditioned at 20 °C and 65% of relative humidity (RH) before being hot pressed. The HP process was carried out at 205 °C and involved three steps:

- HP at 1 MPa for 2 min
- Breathing time of 1 min
- HP at 7.5 MPa for 30 s

The obtained samples were then cured by the FHT in an aerated stove at a temperature (Tc) of 180 °C for two different tc of 0.5 h, and 1 h. Three reference samples without the curing step were manufactured for each thickness to study the influence of FHT on BF properties. The Table 1 summarizes the FHT conditions.

2.3. Mechanical and physical tests

The samples were characterized by following the European Standards method, after being conditioned in a climatic chamber at 20 °C and 65% RH until constant mass. MOE and MOR were measured according to EN 310:1994, while IB was calculated following EN 319:1994. Density and TS were measured in accordance with EN 323:1993 and EN 319:1994 respectively. TS and WA were calculated simultaneously by immerging 50 × 50 mm specimens in a deionized water bath. The specimens were weighed and measured before and after a 24 h period of the water bath, and TS and WA were calculated as thickness and weight difference respectively.

2.4. Statistical analysis

Statistical analysis was performed by Rstudio software. Two-way Analysis of Variance (ANOVA) was used as statistical method to evaluate the significance of thickness and tc on BF properties, and their possible interactions. Tc was assumed as constant also for reference samples and thickness and tc were considered as factors of the analysis. A p-value lower than 0.05 was considered as the minimum for the rejection of the null hypothesis.

3. Result and discussion

3.1. Physico-mechanical results

Table 2 shows the results obtained for BF at the three nominal thicknesses of 3 mm, 6 mm, and 9 mm.

Density ranged between 914.78 kg/m^3 and 977.41 kg/m^3, so that all obtained fiberboards can be considered high density fiberboards (HDF). The results obtained did not meet the standards of the European regulation (EN 312:2010), which requires a minimum of 10.5 MPa for MOR and 0.28 MPa for IB when considering the general use in dry conditions, while a maximum of TS 17% is required in wet conditions.

The poor results obtained were probably due to a lower $logR_0$ that the FHT did not compensate, although an improving effect was clear, especially for dimensional stability.

Table 1
Final Heat Treatment conditions of binderless fiberboards made by steam exploded *Arundo donax* L.

Board Type	Thickness	FHT conditions	
	mm	Tc (°C)	tc (h)
FBR	3	–	–
FB05	3	180	0.5
FB1	3	180	1
FBDR	6	–	–
FBD05	6	180	0.5
FBD1	6	180	1
FBTR	9	–	–
FBT05	9	180	0.5
FBT1	9	180	1

Table 2
Mean values of physico-mechanical results obtained for binderless fiberboard from steam exploded *Arundo donax* L. with 3 mm (FB), 6 mm (FBD), and 9 mm (FBT) thickness, made at different FHT conditions.

Board Type	Density	St. err.	MOE	St. err.	MOR	St. err.	IB	St. err.	TS	St. err.	WA	St. err.
	kg/m^3	–	MPa	–	MPa	–	MPa	–	%	–	%	–
FBR	930.93	11.384	1202	87.275	4.11	0.273	0.507	0.053	77	7.276	133	12.391
FB05	917.36	5.265	895	104.50	3.40	0.665	0.483	0.051	45	7.435	86	3.435
FB1	936.75	7.687	1132	23.355	5.09	0.335	0.395	0.078	36	3.702	58	10.146
FBDR	943.19	7.681	672	2.705	3.87	0.365	0.336	0.042	85	3.111	139	2.873
FBD05	930.28	7.090	559	78.308	4.09	0.305	0.323	0.071	38	1.802	81	3.144
FBD1	914.78	2.037	495	62.287	3.64	0.522	0.333	0.030	25	1.986	49	0.631
FBTR	977.41	5.194	481	23.463	5.21	0.023	0.156	0.023	87	2.618	137	3.390
FBT05	970.90	5.800	533	63.864	5.92	0.029	0.165	0.029	38	1.245	77	0.705
FBT1	968.23	6.431	501	28.446	5.77	0.001	0.144	0.001	24	2.881	47	8.222

Table 3
Two-way ANOVA of the mechanical and physical results of binderless fiberboard from steam exploded *Arundo donax* L.

Property	Factor	D.f.	Sum of squares	Mean squares	F-value	p-value
MOE (MPa)	thickness (mm)	2	1,750,891	875,446	90.99	$8.296 \cdot 10^{-10}$
	tc (h)	2	50,798	25,399	2.640	0.1004
	thickness:tc	4	120,115	30,029	3.121	0.04265
MOR (MPa)	thickness (mm)	2	15.20	7.6019	16.58	$1.013 \cdot 10^{-4}$
	tc (h)	2	0.8748	0.4374	0.9540	0.4049
	thickness:tc	4	3.869	0.9672	2.110	0.1242
IB (MPa)	thickness (mm)	2	0.3975	0.1987	29.54	$2.939 \cdot 10^{-6}$
	tc (h)	2	0.008740	0.004372	0.6498	0.535
	thickness:tc	4	0.01273	0.003182	0.4728	0.755
TS (%)	thickness (mm)	2	95.9	47.90	1.145	0.342
	tc (h)	2	14,857	7428.6	177.41	$4.084 \cdot 10^{-12}$
	thickness:tc	4	466.4	116.6	2.785	0.06021
WA (%)	thickness (mm)	2	156	78.20	0.6176	0.5509
	tc (h)	2	33,503	16751.4	132.38	$4.313 \cdot 10^{-11}$
	thickness:tc	4	292	73.00	0.5771	0.6831

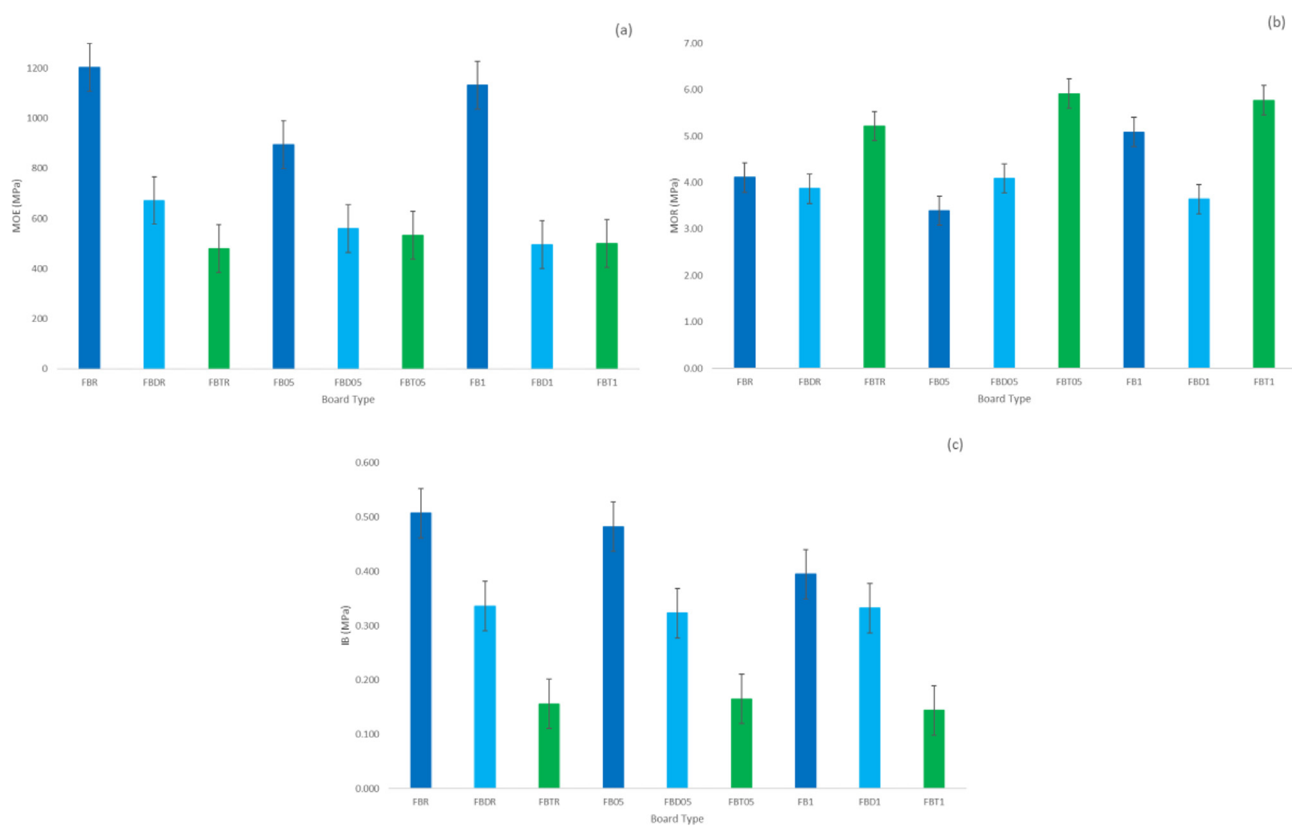

Fig. 1. Mean values of MOE (a), MOR (b), and IB (c) of binderless fiberboards from steam exploded Arundo donax L.

Table 4
Multiple comparison of ANOVA for mechanical results of binderless fiberboards from steam exploded *Arundo donax* L.

Property	Factor	Multiple comparison	p-value
MOE (MPa)	thickness (mm)	6–3	0.0000000
		9–3	0.0000000
		9–6	0.3038649
MOR (MPa)	thickness (mm)	6–3	0.4093386
		9–3	0.0022067
		9–6	0.0001019
IB (MPa)	thickness (mm)	6–3	0.0132490
		9–3	0.0000020
		9–6	0.0008001

3.2. Statistical analysis

Two-way ANOVA was carried out to investigate the extent to which the properties are dependent on the two factors considered, i.e. the thickness and the tc, and to corroborate the earlier observations and then observe results on a statistical basis. Table 3 reported the results obtained by the analysis in which the significant p-values are highlighted.

The interaction between the two factors of thickness and tc was always non-significant, except for MOE. It turned out that for the MOE there was an interaction between the two factors, although the p-value is slightly below the value of 0.05. Besides, the analysis showed that the tc of FHT was significant only for dimensional stability, i.e. TS and WA. No dependence on thickness was detected for TS and WA, while a strict dependence on this of MOE, MOR, and IB can be clearly observed.

3.3. Influence of thickness on mechanical properties

As observed in statistical analysis, the thickness only influenced MOE, MOR, and IB. Fig. 1 (a), (b), and (c) show the mean values for MOE, MOR, and IB with respect to thickness. The MOE results were higher for the lower thickness. The 3 mm specimens presented a MOE that was almost double that of the other thicknesses, while there seems to be no difference between the thicknesses of 6 and 9 mm. This result was confirmed by multiple comparison showed in Table 4.

The results for MOR (Fig. 1 (b)) were less linear, and the multiple comparison in Table 4 shows a significant difference between the 9 mm thickness with the other ones, while between 6 mm and 3 mm there was no significant difference. The higher values obtained for the 9 mm thickness were probably due to the greater inertia.

Instead, the effect of thickness on IB (Fig. 1 (c)) was evident. The difference in IB was more than double between the smaller and the larger thickness. Besides, the multiple comparison in Table 3

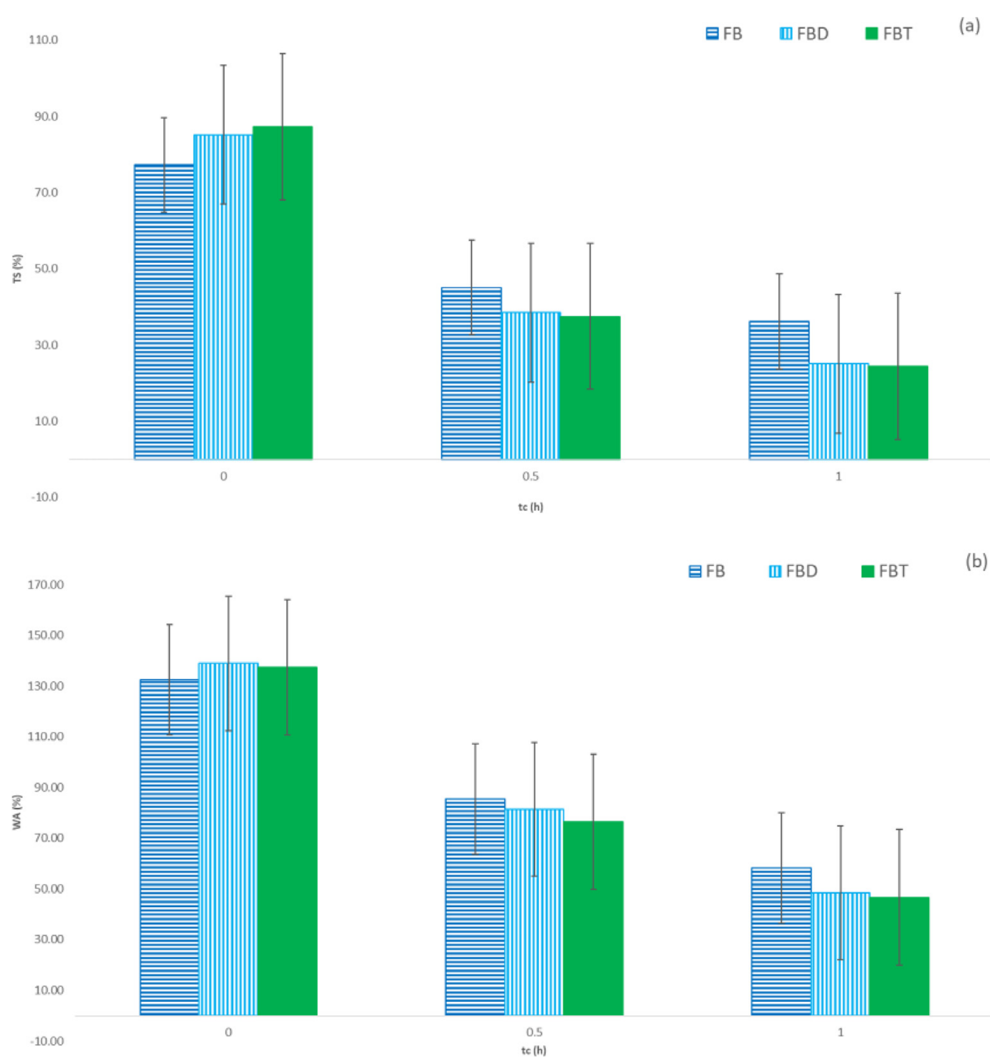

Fig. 2. Mean values of TS (a) and WA (b) of binderless fiberboards from steam exploded *Arundo donax* L.

Table 5
Multiple comparison of ANOVA for Thickness swelling and water absorption of binderless fiberboards from steam exploded *Arundo donax* L.

Property	Factor	Multiple comparison	p-value
TS (%)	tc (h)	0.5–0	0.0000000
		1–0	0.0000000
		1–0.5	0.0050073
WA (%)	tc (h)	0.5–0	0.0000000
		1–0	0.0000000
		1–0.5	0.0001187

showed that IB is much more susceptible to thickness change than the other parameters. The difference among all thicknesses were significant and the higher values were obtained for the smaller thickness. Indeed, the IB measures the internal cohesion of the fibers, and a greater thickness does not allow the heat to penetrate in depth during the HP and, therefore, the bond between fibers is weaker.

3.4. Influence of final heat treatment on dimensional stability

Fig. 2 (a) and (b) report the mean values of TS and WA respectively, with respect to tc. TS and WA performed similarly, and the statistical analysis showed the great influence of tc on TS and WA. This may be due to two phenomena: on the one hand the hydrolyzation of hemicellulose which, considering the reduced $logR_0$, did not reach the completion during the pre-treatment but improved thanks to FHT, and on the other hand the process of covering of the fibers with the softened lignin. Moreover, the multiple comparison (Table 5) showed that there is a significant difference between all group considered, i.e. 0 h, 0.5 h, and 1 h. This result permits to draw a plan to obtain further improvements in the dimensional stability of the BF and fulfil the regulations.

4. Conclusions

The results presented in this study showed that the FHT produce a good improvement in dimensional stability by reducing both TS and WA by more than double with 1 h of FHT at 180 °C. Boards produced without the FHT showed the lowest dimensional stability. The thickness did not show any influence in dimensional stability but only for the values of MOR and IB that are strictly related to dimensional parameters. Best results for MOR were obtained for 9 mm thick BF, ranging between 5.21 and 5.92 MPa with no difference due to the tc modification. The tc modification did not have any influence in IB too, which performed similarly to MOR, but best results were obtained for the lower thickness. Further research is needed to study different conditions of FHT, varying both the Tc and the tc, and to investigate the relationship there may be between the effects of STEX pre-treatment $logR_0$ and those of FHT on BF properties.

CRediT authorship contribution statement

Federica Vitrone: Conceptualization, Methodology, Formal analysis, Validation, Investigation, Data curation, Visualization. **Diego Ramos:** Conceptualization, Methodology, Investigation, Resources, Data curation, Writing – review & editing. **Francesc Ferrando:** Conceptualization, Methodology, Investigation, Resources, Writing – review & editing, Project administration, Funding acquisition. **Joan Salvadó:** Conceptualization, Methodology, Writing – review & editing, Supervision, Funding acquisition.

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.

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