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Comparison of the properties of flax shives based particleboards prepared using binders of bio-based lignin and partially bio-based epoxy resin

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Abstract

In order to reduce formaldehyde emissions, an experimental investigation of lightweight flax shives based particleboards, prepared using two different binders: bio-based lignin (lignosulfonate) and partially bio-based epoxy resin (greenpoxy 56), was undertaken. The flax based particleboards (with a target density of 500 Kg/m³), were elaborated by a thermo-compression process using 20 wt% of binder content. The flax shives were directly valorised as they had no pre-treatment, in order to qualify their potential as a raw material. Bending, compression and fire resistance tests showed that the mechanical and flame performances of lignosulfonate-based particleboards were high compared to those of greenpoxy56 based panels. Nevertheless, the latter were found to have an interesting dimensional stability after water immersion and good insulating properties. Moreover, both lignosulfonate and greenpoxy 56 based particleboards met the minimum EN 15197 requirements for non-load bearing flaxboards for use in dry conditions (Type FB2). Such results illustrated the competitiveness of the studied particleboards, as bio-sourced structures, compared to urea-formaldehyde based panels.

Keywords:

Particleboard; Flax shives; Bio-based binders; Mechanical properties; Thermal properties

1 Introduction

Binding agents are widely used in particleboard applications and they create most of the bonds in a particleboard [1]. Phenol-formaldehyde (PF) resin is a widely used binding agent, owing to its strength and moisture resistant properties [2,3]. Other resins, such as urea-formaldehyde (UF), resorcinol-formaldehyde (RF), melamine-formaldehyde (MF) and diisocyanates are used in particleboards [4,5]. However, these are toxic and expensive [2,6]. Thus, the demand for the replacement of such adhesives is not only due to environmental concerns but also out of economic interest [2].

Lignin has a role as a binding agent in the biomass itself. Several studies have reported on the partial replacement of phenol in PF resin by several types of lignin, varying the proportions and incorporation of lignin with PF or other commercial resins in order to fabricate particleboards. The partial replacement of phenol in PF resin by organosolv lignin and modified organosolv lignin (phenolated-lignin) was tested [2]. The chemical modification of lignin is usually performed to increase the reactive sites and thus, their reactivity while undergoing condensation reactions. Many studies have been undertaken on the modification of lignin. Particleboards were produced using resins where chemically modified lignin (such as methylolated lignin, demethylated lignin and methylolated black liquor) partially replaced phenol in PF resins [3]. In certain studies, lignin replaced phenol both partially and completely in phenol-formaldehyde resins in the fabrication of fibreboards [7].

With respect to the incorporation of lignin with conventional resins, modified lignin (such as hydroxymethylated lignin and glyoxalated lignin) were incorporated as a resin

component (diisocyanates and with/without PF resins) in various proportions and used for the fabrication of particleboards [8]. Lei H. et al. [9] prepared particleboards using different resin components, such as glyoxalated lignin, glyoxal, tannin, diisocyanates and PF resins. Stephanou and Pizzi [10] used various proportions of methylolated kraft lignin, diisocyanates and PF resins as binders for the preparation of particleboards. Anglès et al. [11] used pre-treated sawdust material for the fabrication of panels with different types of lignin as the only binder, which varied from 5% to 20% on a dry solids basis. They also prepared binderless panels and compared the effect of adding lignin to the panels. Velasquez et al. [12] used kraft lignin at different proportions, mixed with steam-exploded pulp for the fabrication of particleboards. Privas and Navardused [13] pre-treated flax fibres for fabrication of panels using liginosulfonate as the only binder.

On the other hand, the use of thermosetting matrices as a binder in fibre-reinforced composites is increasingly common because of its mechanical performance and compatibility with different fabricating processes [14,15]. The thermosets have low viscosity compared to thermoplastics, which permits relatively better impregnation among the fibres. Epoxy resin is one of the most widely used thermosetting matrices. It is even used with plant fibres such as flax, hemp, bagasse and sisal [16–20].

In recent decades, the development of an alternative matrix to the conventional oil-based ones has gained interest. The context extends to reducing the CO₂ footprint and dependence on fossil fuels. Formaldehyde-free bio-sourced phenolic resins and bisphenol A-free epoxy resins are the outcomes of such an approach [21].

Several studies have focused on the use of long flax fibres as a reinforcement in bio-sourced composites but few have been interested in the valorisation of flax shives,

which comprise the woody core generated as a by-product in the flax scutching process. These shives are not valorised as primary materials but for bio-fuel, animal bedding and mulch. These plant particles which are widely available in the north-west of France, have a porous structure and mechanical strength that promises to become a potential replacement for wood particles in light agglomerated panels.

In general, a wide variety of agro-resources such as bagasse [22], hazelnut and almond husk residues [23], [5] and flax shives [24] have been combined with thermosetting based formaldehyde. However, there has been little mention of the use of lignosulfonate or epoxy as being the only binders for the fabrication of plant particleboards [25,26].

In this study, GreenPoxy 56 (a partially bio-sourced thermosetting resin, which has up to 56% of its molecular structure from plant origins) and calcium lignosulfonate are used as a matrix for particleboards. Furthermore, flax shives, an agricultural residue, are directly valorised as they were and with no pre-treatment. Hence, the fabricated particleboard is novel owing to such a combination of the material and the binder.

In the present study, because of environmental concerns and local resource development, a straightforward method for the fabrication of innovative flax shives based particleboards using the bio-based binder lignosulfonate (PLS) and a conventional, partially bio-based binder epoxy resin (PER) has been established and presented. The mechanical and thermal properties of fabricated PLS and PER particleboards are studied, as well as water absorption, thickness swelling and flame tests. An attempt to understand the relationship between fabrication, structure and properties of bio-based particleboards and interactions within them has been undertaken.

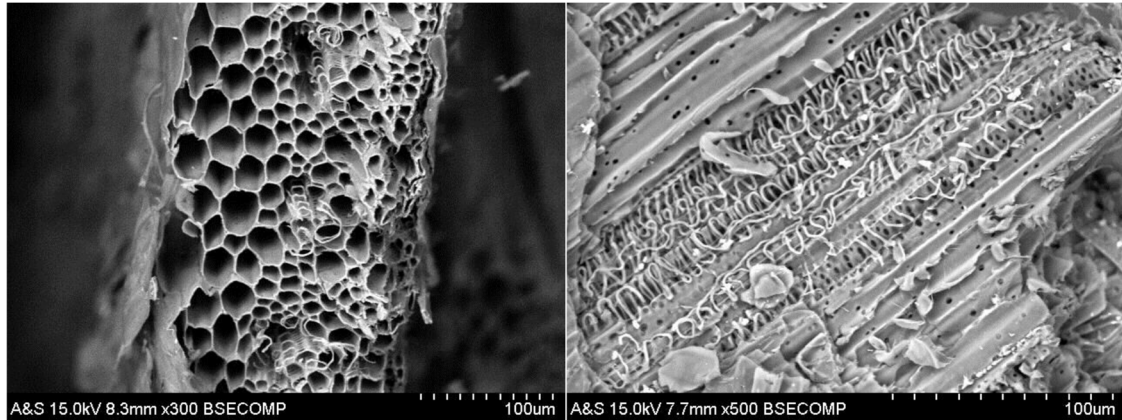
2 Material and methods

Flax shives were purchased from Terre de Lin (cooperative engaged in the cultivation and processing of flax in France). The morphological analysis of the raw flax shives used in this study was performed by using the optical microscope Keyence VHX-700F. It was performed from a sample containing about 100 particles. Flax shives exhibit coarse stick shapes with a 8.3 mm average length, a 1.2 mm average width and a 0.3 mm average thickness. Table 1 summarises the measured geometrical characteristics of raw flax shives.

Table 1. Geometrical characteristics of raw flax shives.

	Length (μm)	Width (μm)	Thickness (μm)
Raw Flax Shives	8371 ± 5018	1248 ± 498	329 ± 108

The moisture content of these particles prior to particleboard production was about 10.2% wet basis (w.b.). The apparent density of flax shives was approximately equal to 0.1 g/cm³. Such a value is due to the alveolar structure of these particles which highlights their potential for insulating panels (Fig. 1). The real density of the flax shives was measured by pycnometer and found to be 0.149 g/cm³.



(a)

(b)

Fig. 1. (a) The alveolar structure of flax shives and (b) presence of holes on its walls.

The flax shives used for this study belong to the same batch that was already studied in the laboratory using the Van Soest approach [27]. Table 2 summarises the measured biochemical composition of the biomass.

Table 2. Flaxshivesbiochemical composition [27].

Organic matter	Composition
	(%)
Cellulose	43 ±1.3
Hemicelluloses	19.5 ±0.7
Lignin	32.3 ±1.7
Other solubles	5.2 ±0.2

This study uses flax shives in their raw state, as they are marketed, without any heating or the addition of external wax or water-repellent chemicals.

Commercial partially bio-based epoxy resin GreenPoxy56 (GP 56) and calcium lignosulfonate (LS) were used separately as a binder in the manufacturing of

particleboards. GP 56 resin was purchased from Sicomin and crosslinked by SD8822 hardener according to the weight ratio of 100/31, as suggested by Sicomin. The combination of these two components ensures good fluidity in the mixture compared to the other tested hardeners, whose dynamic viscosity at 20 ° C is 507 mPa.s. The LS was provided by Carl Roth GmbH + Co. KG.

2.1 Fabrication of particleboards

The flax shives were sieved through meshes of 4 mm and 1 mm diameter. The residue in the 1 mm sieve was used for the fabrication of particleboards. A quantity of LS, corresponding to 20% relative to the total weight of the panel, was dissolved in water and then mixed with the flax shives (80% relative to the total mass). The mass of added water was not included in the total weight due to its evaporation at the time of manufacture. The resulting mixture was thermo-compressed in a Scamex Presse 15T at 190°C for 30 minutes. Epoxy resin (20% w/w) was mixed with the flax shives (80% w/w) and thermo-compressed at 90°C for 10 minutes. For all panels, the pressure applied corresponded to the pressure required to close the piston mould, which guaranteed a constant volume of the panels 300*300*10 mm³. For this, steel shims were placed between the mould and the piston to ensure a similar thickness in all boards. During the manufacturing cycle, the press was controlled by the gap between the piston and shims.

For all PLS and PER panels, the target density was equal to 500 kg/m³. Nevertheless, it was proved that, after manufacturing under these conditions, PER panels had a mean density of approximately 470 kg/m³ lower than that of the PLS ones (500 kg/m³). This amounts to an increase in the thickness of PER of about 10.8 mm due to a spring-back

after the removal of the upper plate of the press. In fact, during the hot pressing a large amount of water vapour migrates towards the centre of the panel, which requires a longer pressing cycle to allow the evacuation of moisture. The release of pressure when the moisture content in the core of the panel is higher may provoke spring-back or non-reversible dimensional changes [28]. Such a density difference (of 30 kg/m³) will have little influence on the mechanical and physical properties of particleboards. However, thermal properties can be affected. Therefore, for a better comparison between PER and PLS insulation properties, the thermal results of PER panels were adjusted to 500 kg/m³. Moreover, observations by X-ray micro-tomography were carried out on samples taken from the PLS and PER panels, in order to determine the porosity rate between the flax shives. The processing of the images obtained by the Image J software gave a porosity rate equal to $8.8 \pm 2.9\%$ for the PLS panels and $10.1 \pm 5.3\%$ for the PER ones. This small difference in the porosity rate will allow comparison between the mechanical and thermal properties of the PLS and PER panels. The processed particleboards were preserved at room temperature ($\sim 20^{\circ}\text{C}$) and relative humidity ($\sim 55\%$) until used in the experiments.

2.2 Mechanical tests

Three-point bending, compressive and internal bond tests were performed on the particleboards using a type 5867 INSTRON universal testing machine. For each type of test, at least five specimens were taken from different panels and tested to ensure the reproducibility of the results. All the mechanical tests were conducted at least one week after the particleboards had been manufactured under room temperature and humidity.

Three-point bending tests were performed to study the flexural behaviour of the PLS and PER particleboards. The modulus of elasticity (MOE) and modulus of rupture (MOR) were extracted. The tests were conducted on longitudinal specimens according to Standard NF EN 310.

Compression tests were carried out on specimens of dimensions 50*50*10 mm³ according to the ASTM C365 standard.

Internal bonding strength corresponds to the force required to break the sample by a z-directional tensile test. It gives an estimation of the shive-to-shive bond. Specimens of dimensions 50*50*10 mm³ were glued onto stainless steel supports with SR7100/SD7105, provided by Sicomin. The thickness of the adhesive layer was approximately 0.5 mm. Then, samples were tested according to the NF EN 319 standard.

2.3 Thermal conductivity

A Netzsch HFM 436 Lambda meter was used to measure the thermal conductivity λ (W/m.K) of the particleboards. Two panels of dimensions 300*300*10 mm³ were perfectly superimposed to correctly establish the balance between the upper plate (hot) and the lower plate (cold) set at different temperatures, where the heat flows from the upper towards the lower plate. The temperature gradient (ΔT), which is the difference between the upper and lower plates, was 20°C.

2.4 Water absorption and thickness swelling

The water absorbed (WA) and the thickness swelling (TS) of the processed particleboards were measured by immersing them in water at room temperature for 24

hours. Sampling was carried out by cutting the processed particleboards into specimens of 50*50 mm². The weight of the specimen and thickness at the centre were measured prior to their immersion in water and after removal.

2.5 Flame test

“The particleboards’ fire resistance was tested according to the NF EN ISO 11925-2 Standard. The measurements were undertaken in a dedicated chamber where samples of dimensions 290*90*10 mm³ were exposed to a propane flame, inclined by 45° on the lower edge of the sample, for a duration of 15 s. Experiments were carried out in triplicate on each board. After exposure to the flame, the absence of ignition and of flaming droplets was controlled and the height of the damaged zone was measured [27]”.

3 Results and discussion

3.1 Mechanical properties

According to standard ANSI A208.1 (1999), particleboards have been classified into three types of nominal density: low (<640 kg/m³), medium (640-800 kg/m³) and high (>800 kg/m³). In this study, the density of PLS and PER particleboards was about 500 kg/m³. The choice of this low density was made so that our panels could be compared with other less dense materials, such as balsa and cork.

Table 3 summarises the mean values and standard deviations of the mechanical properties for both PLS and PER particleboards under bending, compression and internal bond investigation, compared to values given by standard EN 15197 (2007) for non-load bearing flaxboard in dry conditions (Type FB2).

Table 3. Mechanical properties of PLS and PER thermo-compressed particleboards and requirements from NF EN 15197 standard of FB2 type panels.

	PLS	PER	FB2
Density (kg/m ³)	497.6±4	469.9±18	-
MOE (MPa)	2467±202	1517±225	1500
MOR (MPa)	9.9±1.2	8.4±1.2	9
E_{comp} (MPa)	79.3±2.6	58.3±4.3	-
σ₁₀ (MPa)	5.4±0.16	3.6±0.27	-
IB (MPa)	0.17±0.01	0.41±0.03	0.23

The results show that the MOE (2467.0 MPa) and MOR (9.9 MPa) values of PLS particleboards are higher than those of the PER ones (1517.0 MPa and 8.4 MPa, respectively). Moreover, the standard deviation is less in the PLS particleboards. Essentially, this reveals a better wetting of flax shives by the lignosulfonate solution, leading to improved bonding. In fact, the optimum amount of solution used for the manufacture of PLS boards, results in a homogenous mixing of the flax shives and the

lignosulfonate. The amount of time spent in the thermo-compression press allows the evaporation of the water and plasticisation of the lignosulfonate under temperature and pressure. This could lead to a consistent mechanical strength for the PLS particleboards.

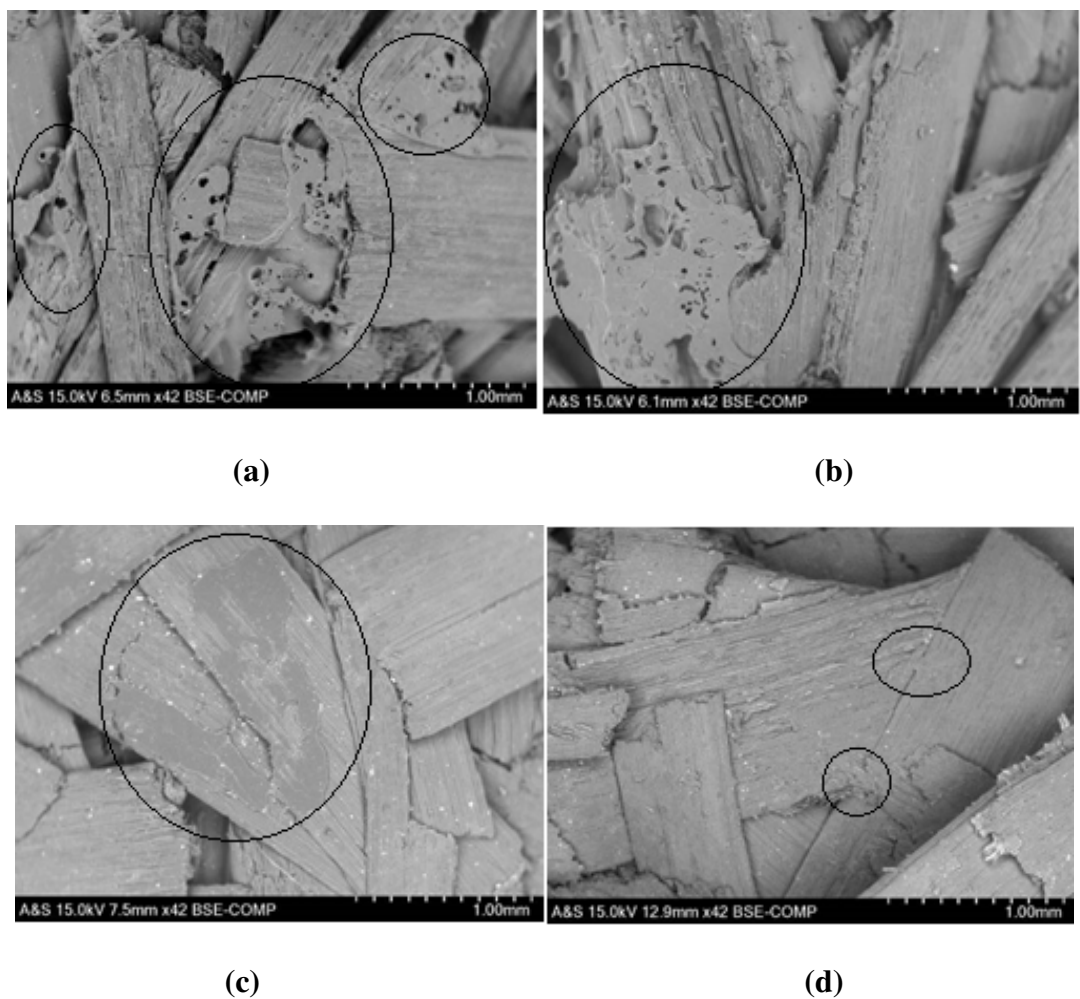


Fig. 2. SEM images of the thermo-compressed PLS (a, b) and PER (c, d) with highlight over the areas with matrix.

However, the relatively viscous and lower volume epoxy resin used does not result in such a level of homogeneity in the mix. Thus, the matrix distribution is more heterogeneous. This can be seen on the SEM images. Fig. 2a and Fig. 2b show several

binder bridges that connect the flax shives. In contrast, Fig. 2c and Fig. 2d show less abundant and continuous resin bridges.

Considering the literature, lignosulfonate binder constitutes a veritable internal adhesive agent that provides better cohesion inside the particleboards. Nguyen et al. [25] showed that replacing a part of the nerve or bone glue with 9% (w/w) of lignosulfonate considerably increases both the modulus and bending strengths of bamboo particleboards. Anglès et al. [11] showed that the addition of lignin (up to 20%) significantly increases the mechanical properties of softwood panels compared to binderless ones. Evon et al. [29] showed that because of the highest link between cellulosic chains and lignins inside raw shives, the mobilisation of the ligneous binder during the thermo-pressing process seemed to be really difficult. Thus, the particle wetting and panel cohesion are insufficient. Adding 25% Biolignin improved the wettability of the surface area of the particles. As a result, the adhesive properties of this natural binder activates cross-linking formation between the cellulosic chains in plant particles and bending properties are improved.

Furthermore, it can be observed that flax shives have interesting mechanical properties compared to other particles used in binderless particleboards, such as sun flower [27], banana trunk waste [30] and bark and leaves of oil palm biomass [31]. This can be attributed to the high percentage of cellulose chains (43%) and lignin (32.3%) in flax shives, which are the main structural components providing strength and stability.

According to standard EN 15197, the bending properties of PLS particleboards under investigation satisfy the requirements for non-load bearing flaxboard for use in dry conditions (Type FB2). However, the MOR of PER boards (8.4 ± 1.2 MPa) is slightly below the standard value (9.0 MPa).

Beyond the bending response, the PLS panels showed an even performance in compression with a modulus and compressive stress at 10% relative deformation, respectively (equal to 79.3 MPa and 5.4MPa vs 58.3 MPa and 3.6 MPa for PER particles). These low compressive properties of PER panels were attributed to its high porosity rate, which reduces its compactness. Such results reaffirm interest in the use of PER particleboards as an insulator material in construction.

However, the internal bonding strength of PLS particleboards (0.17 MPa) seemed very low compared to PER ones (0.41 MPa). This reveals that the mechanical properties of the bio-based epoxy resin, which are very high compared to bio-based lignin, strengthen and increase the internal bonds of PER particleboards.

3.2 Thermal conductivity

As explained above, for a better comparison between PER and PLS insulation properties, thermal results were adjusted to 500 kg/m³. The results reveal that PER particleboards are more thermally insulating than PLS ones, presenting a mean value of thermal conductivity equal to $0.074 \pm 0.002 \text{ W/m K}$ as opposed to $0.081 \pm 0.001 \text{ W/mK}$ for PLS (see Table 4).

Several parameters directly affect thermal conductivity, such as the nominal density, the manufacturing conditions (pressure and temperature) and the raw material utilised in the production of the particleboard [32].

Table 4. Raw particles and particleboards thermal conductivity. Density of particleboards is 500 kg/m³.

*The real density of Cement-flax shives particleboard FSC was 520 kg/m³. But, it was adjusted to 500 kg/m³.

Materials	Thermal conductivity (W/m.K)	References
FS bark	0.042±0.001	
SF bark	0.051±0.002	
FS binderless	0.077±0.001	[27]
SF binderless	0.077±0.002	
FUF	0.074±0.002	
SUF	0.075±0.001	
FSC*	0.118±0.005	[33]
PER	0.074±0.002	Present study
PLS	0.081±0.001	

In this study, the dimensions and density of the panels and the nature and quantity of the flax shives used are identical. Thus, the difference in thermal conductivity hints at the binder and the porosity of the particleboard. The porosity lies within the individual flax shives due to its alveolar structure (Fig. 1) and between shives.

The addition of a binder consumes such cavities. However, as already explained in the section on mechanical properties, the distribution of the bio-based epoxy is more heterogeneous. So, porosity distribution in PER should explain the difference in thermal conductivity compared to PLS.

Cravo et al. [34] explained the effect of the high porosity in particleboards on the reduction of thermal conductivity. In fact, few solid contacts result in many voids being

filled with air. The heat transfer in the panels occurs through the solid particles and the air. The latter has lower thermal conductivity than the particles. As a result, the insulation of the particleboards increases.

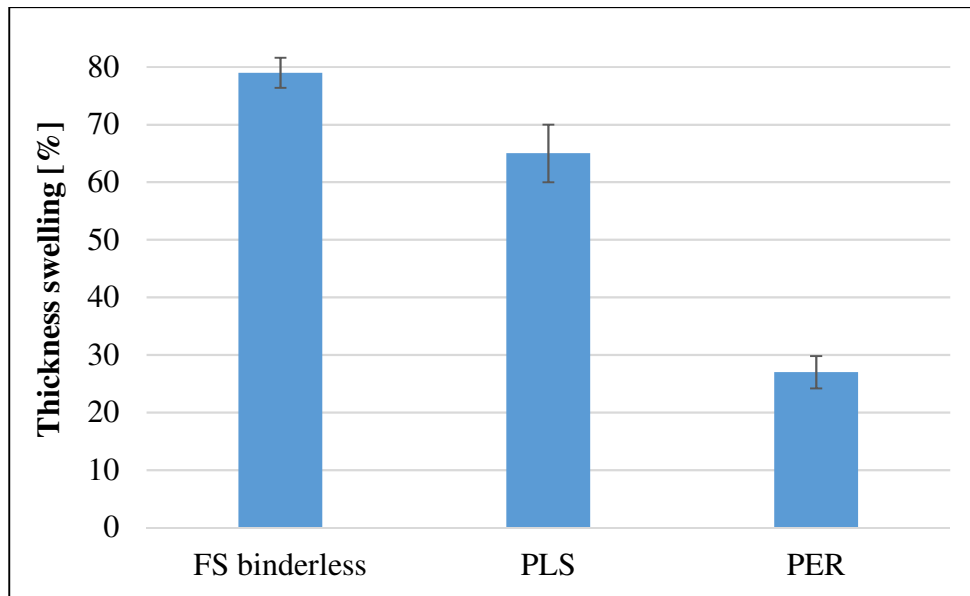
Regardless of the porosity rate, comparing the thermal conductivity of the binderless flax shives panels (0.077 W/m.K) with those of PER and FUF (0.074 W/m.K), PLS (0.081 W/m.K) and FSC (0.118 W/m.K), seem to show that the thermoset resin (GreenPoxy 56 and urea formaldehyde) have improved the thermal insulation of the panels contrarily to the bio binder and cement. Looking at the literature, it seems that epoxy cross-linked polymers are good thermal insulating materials [35], [36].

According to Wang [37], materials with thermal conductivities lower than 0.25 W/m.K are considered to be thermal insulators. Therefore, despite the scattering of the thermal conductivity results in this study and in the literature, plant particleboards have been shown to have insulating properties, which are required in building applications thanks to their alveolar structure and important internal porosity.

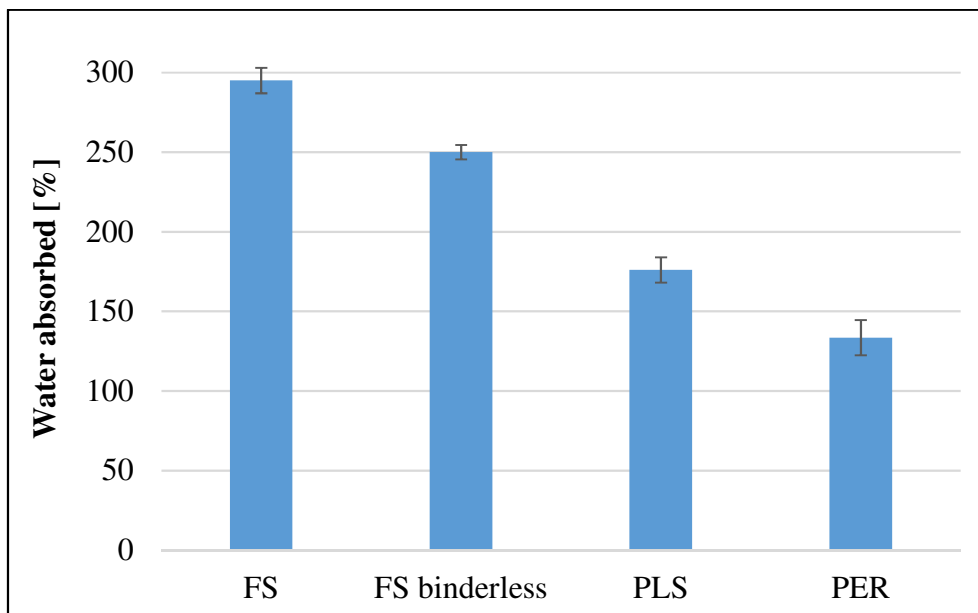
3.3 Dimensional properties

Fig. 3 and Fig. 4 present the results of thickness swelling (TS) and water absorption (WA) of PLS and PER particleboards after immersion in water for 24h, respectively as well as previously measured results for bulk flax shives and flax shives (FS) binderless boards [27]. It has been shown that TS results for FS binderless board ($79\pm2.6\%$) are higher than those of PLS panels ($65\pm5\%$), which, in turn, are more than two times higher than PER ones ($27\pm2.8\%$). Likewise, the WA of FS binderless panels ($250\pm4.5\%$), which is directly related to TS, is higher compared to those of PLS

329 (176±8%) and PER (133.46±11%) boards. Bulk flax shives scored the highest WA
330 value (295±8%).



331
332 **Fig. 3.** Thickness swelling of specimens cut out from flax shives binderless
333 particleboards (FS), PLS and PER.



335
336 **Fig. 4.** Water absorbed of bulk flax shives (FS) and specimens cut out from FS
337 binderless particleboards, PLS and PER.

Compared to other plant particles, such as wood, sunflower, tomato stalks and bagasse [38], [27], flax shives have the highest TS and WA capacity. This can be explained by its high alveolar system, thin pore walls, low bulk density and chemical composition. The low water resistance of both FS binderless and PLS particleboards (compared to PER) can be explained by the higher susceptibility of the bio-glue to hydrolysis in water than cured GP56 resin networks. During immersion of the bio-based epoxy resin, water interacts with the polar groups of the resin, conducting the rupture of the hydrogen bonded interchains and intrachains [39]. However, the diffusing water creates hydrogen bonds with the flax shives. Therefore, immersion in water reduces but does not eliminate the interactions between shives and resin and causes the thickness swelling. Therefore, many authors have proposed some treatments and studied their effects on the water intake of lignocellulosic aggregates. Coating plant particles with hydrophobic substances (such as linseed oil and paraffin wax) prior to their mixing with resin, is a commonly used treatment because of its low consumption of energy and friendliness to the environment [40], [41]. Surface treatment is a further solution to improve the water resistance. Sain et al. [42] suggest a chemical modification of flax shives in the refiner in situ by introducing a reaction product of polyols and organic anhydrides. These different treatments enhance bonding between the hydrophilic particles and hydrophobic binder and, thus, reduce the affinity to water absorption.

3.4 Resistance of particleboards to fire

The appearance of the PLS and PER particleboards after fire testing is shown in Fig. 5.

The conformity of the panels to the standards is controlled through the measurement of the height damaged by the flame.



(a) (b) (c)

Fig. 5. (a) Aspect after test fire of the whole PLS particleboards, (b) the first PER panel and (c) the second PER board.

As soon as a small flame of 2 cm was applied for 15 seconds on the lower edge of the first PER sample, a smouldering fire developed, which continued even after the burner was stopped. In parallel, smoke was released. The fire spread inside the panel and the probes fell down. After 1 minute, the fire was extinguished in the first PER panel. The damaged height was approximately 13 cm.

For the second PER sample, the flames quickly rose up and even exceeded the height of the panel. The extinction of the flame was not natural. The panel was blackened on all sides. However, PLS specimens retained their integrity during and after the fire tests on all four of the samples tested. There was no production of flaming droplets during exposure to the flames. Moreover, PLS panels showed self-extinguishing behaviour

after the burner stopped. The measured height of the damaged zone was equal to 5.6 ± 0.2 cm.

According to the NF EN ISO 11925-2 requirements on the height limit of the damaged zone which is acceptable in the building sector (which must not exceed 15 cm) and from the scattered findings on PER specimens, only PLS panels comply with this standard, especially with class E.

The poor resistance to fire of PER particleboards is essentially caused by the resin, which is easily flammable. Therefore, several approaches have been conducted in order to improve the fire resistance of biocomposites, such as chemical modification of existing adhesives [43], direct addition of flame retardant materials [44], incorporation of mineral particles into the board composition [45] and surface treatment of lignocellulosic particles [46].

4 Conclusions

On the one hand, this study set out to valorise flax shives as an abundant natural resource in France and, on the other, to investigate the use of alternative binders to the usual commercial ones based on formaldehyde, in order to produce lightweight particleboards. Two different binders were separately used: a commercial, partially bio-based epoxy resin (GP 56) and calcium lignosulfonate (LS).

A comparison between the bending and compressive behaviours of LS and GP56 based particleboards shows that totally bio-sourced panels have better properties than partially bio-sourced ones due to a better wetting of flax shives by the LS binder. However, this type of binder influences the dimensional instability of flax based particleboards by

increasing their rate of water absorption and thickness swelling. This can be explained by the higher susceptibility of bio-glue to hydrolysis in water than cured bio-based epoxy resin networks. Moreover, thermal tests have justified interest in the use of the bio-based epoxy based particleboards as insulator materials in the building sector. Nevertheless, the use of these materials in such a field remains critical because of its poor fire resistance compared to PLS particleboards, which comply with class E.

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