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Hybrid Sandwich Particleboard Made with Sugarcane, Pínus Taeda Thermally Treated and Malva Fibre from Amazon

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A multilayer particleboard panels, consisted of sugarcane bagasse reinforced composite as core material and hybrid composites made with Pinus taeda particles and Malva fibres as facing materials, were designed and evaluated in this work. Tukey test was used to identify the effect of the facing material, considering different combinations of *Pinus taeda* particles and Malva fibres, on the bulk density, thickness swelling, flexural strength, flexural stiffness and X-ray densitometry. A spray-up process was used to spread castor oil based polyurethane resin upon the dispersive phases followed by a hot compaction at 100°C. The particleboards were classified as medium density panels in accordance to the Brazilian, American and Canadian Standards. All treatments reached the minimum strength except for elastic modulus. Tukey test demonstrated the flexural strength and modulus responses for all treatments were statistically similar. Hybrid particleboard consisted of 75% of Pinus *taeda* wood and 25% of Malva fibres revealed a promising sustainable material for furniture industries, combining strength, low-cost and lower thickness swelling values.

Keywords: *mechanical and physical properties, X-ray densitometry, multilayer particleboards, sugarcane bagasse, Pinus taeda and Malva fibre*

1. Introduction

In recent years, the building and furniture industries have demanded particleboard panels with high durability and sustainable characteristics[1]. The panels have been produced by pressing lignocellulosic materials, leading to high mechanical properties and specific strength/stiffness[2]. The Brazilian particleboard industries have been used a high amount of reforested wood, such as Pinus and Eucalyptus[3,4]. The demand for wood products has been rising considerably year by year, which promotes an increase in reforestation areas with fast-growing species[5]. In general, fifty percent of the particleboard industries use softwood as main raw material, and the others use more than one wood specie in the production lines[6,7]. Commonly, reforested woods do not achieve the structural requirements for particleboard panels design. This situation is considered an economical and environmental concern, in which the scarcity of raw material can promote its valorisation. Instead of increasing the reforestation areas, alternative materials have been considered to replace wood without loss of quality[8].

The generation of lignocellulosic wastes from the Brazilian agricultural industries, such as corncob, rice husk, coffee husk, peanuts husk, coconut husk, castor bean husk, sugarcane bagasse, among others, has been considered promising raw materials and recycling route to meet growing particleboard market demand. In addition, a reduction in the use of reforested wood can also contribute to make the products cheaper[6].

The sugarcane bagasse is a solid waste that remains after grinding process in the ethyl alcohol production. The generation of bagasse depends on the sugarcane species. In general, 30% of bagasse is extracted in a ton of sugarcane processed. The most part of this bagasse is burned in a

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boiler for electrical generation and the other part is used to produce pulp, paper and board. However, the burning of sugarcane bagasse releases pollutants into the atmosphere. The sugarcane waste can be considered underexplored in Brazil[9-11]. The main components of sugarcane bagasse are 32-50% cellulose, 19-25% hemicelluloses, 23-32% lignin, 2% ashes, 46% fibres and 50% humidity[12]. The chemical composition of sugarcane bagasse shows similarity with softwoods. Bagasse is a lignocellulosic waste with a potential use in the production of particleboards[13].

The use of natural fibres has been an alternative as low impact income, strategy of environmental conservation and fast biodegradability. *Malva* and *Juta* are Amazonian fibres commonly used to produce paper, dress room, string, tissue, carpets and sacks for packing of products like coffee, cor and cocoa. In order to use these fibres as reinforcements in composite production, several surface treatments have been proposed in the literature to enhance their interaction with the matrix phase[14-17].

Thermal treatments performed in wood can change its chemical components in the cell wall and extractives. In general, oxygen and hydrogen content are reduced in the wood, since hemicelluloses are more susceptible to degrade in low temperature, due to low molecular weight, branched and amorphous structure with different and substituted monomeric units. Deacetylation reactions lead to the formation of acetic acid, in which it is a catalyst in the depolymerisation, while dehydration leads to the formation of furfural and hidroxymethylfurfural. The crystallinity cellulose increases with amorphous cellulose degradation, therefore, the accessibility in hydroxyls groups to the water molecule decreases. At the same time, a thermal treatment produces dehydration reactions and cellulose oxidation, which promote the lignin condensation[14-17].

Panels made with sugarcane bagasse and eucalyptus wood, impregnated with phenol formaldehyde and urea formaldehyde resin at 6, 9 and 12wt% levels, have been evaluated by Mendes et al.[6]. Thickness swelling, water absorption, density, flexural strength and stiffness properties have been assessed. The urea resin has led to similar or superior results than phenol formaldehyde. Mean stiffness values approximately at 1000 MPa did not fit the range from 1750 and 2450 MPa recommended by Canadian Standard CS - 236-66.

Hot-pressed MDF panels made with pine with different percentage of phenol formaldehyde (6, 9 and 12%) have been evaluated via internal bond, flexural strength and stiffness. The panels were thermally treated at 160, 170 and 180 ºC. The findings did not reveal significant changes in mechanical properties when compared to the pristine condition. Moreover, the internal bond results were higher than the pristine condition[18].

Panels made with eucalyptus fibres with different percentage of sugarcane bagasse (25, 50, 75 e 100%) and impregnated with urea formaldehyde have been investigated by Belini et al.[19]. The increase amount of sugarcane bagasse has led to reduced flexural strength and stiffness. At 100% level of sugarcane bagasse the results did not attend the minimum requirements described in the Brazilian Standard ABNT NBR 15316-3[20]. The mean flexural strength and modulus values found for panels made with 100% of sugarcane bagasse were 14.3 and 1736 MPa, respectively. In contrast, the internal bond (0.55 $N.mm^{-2}$) and thickness swelling (12%) findings attended the minimum values recommended by NBR 15316-3[20].

Iwakiri et al.[21] have evaluated the physical and mechanical properties of particleboard panels manufactured with wood particles from Sequoia sempervirens and Pinus taeda combined with urea-formaldehyde resin (UF), using different mixing ratios. The findings achieved for internal bond, modulus of elasticity (MOE) and modulus of rupture (MOR) under static bending have met the minimum requirements of British Standard BS312:2003 in all treatments, revealing that Sequoia sempervirens has great potential for particleboard production[21].

In order to combine the mechanical performance of Pinus taeda and Sugarcane bagasse, hot-pressed hybrid sandwich particleboard panels were investigated in this work. The sugarcane bagasse composite was used as core material, while hybrid composites, made with Pinus taeda thermally treated and Malva fibres, were used as facing materials. The properties such as, bulk density, thickness swelling, flexural strength and stiffness, combined with scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS) and X-ray densitometry analysis were used to characterize the panels.

2. Material and Methods

Pinus taeda wood particles were thermally treated following the methodology reported by Silva[22]. A density of 550 $kg.m^{-3}$ and a wood moisture content of 12% (±2) were initially measured. The thermal treatment was conducted in a metal container built in an electrical furnace. Wood pieces of 560 x 160 x 60 mm^3 in dimensions were placed inside the container. Two iron bars of 10 mm in diameter was placed between the wood timbers to promote the gas circulation. The thermal degradation in wood components occurs faster in oxygen atmosphere[23]. For this reason, the thermal treatment was performed in a nitrogen atmosphere, in which a constant flux was injected to avoid wood oxidation. Seven wood timbers were placed inside the container. The temperature levels were measured by seven thermocouples (type K), in which three of them were placed inside the first wood timber, two inside the sixth wood timber, one inside the electrical furnace and the other inside the container. This setup was used not only to control the furnace temperature but also to verify the heat inside the wood timber. The thermal treatment was carried out at 200ºC (±5) with an initial heating rate at 2.5ºC.min^{-1} until 100ºC, and 0.033ºC. min^{-1} until the final temperature.

The multilayer particleboard panels were constituted of lignocellulosic particles, such as the sugarcane bagasse, Pinus taeda particles and Malva fibres. Firstly, the flakes were ground in a knife mill being pre-classified in a particle size range from 2.8 mm to 5mm in diameter. Subsequently, a sieving process was used to classify the sugarcane in a particle size range between 2.38 mm and 0.50 mm, the Pinus taeda between 1.41 mm and 0.50 mm, and the Malva fibres between 4.77 mm and 3.36 mm.

The panels were designed to have a density of 0.7 g.cm^{-3} in order to achieve a medium density particleboard based on the Brazilian Standard ABNT NBR 14810[24]. The core material was made with sugarcane bagasse considering 6mm in thickness. The facing material is a hybrid composite consisted of treated Pinus taeda particles and Malva fibres with 2 mm in thickness for each side (see Figure 1). Five different fibre configurations (Table 1) were investigated as follows: Treatment A with 100% of Pinus taeda (100P), Treatment B with 75% of Pinus taeda and 25% of Malva fibres (75P/25M), Treatment C with 50% of Pinus taeda and Malva fibres (50P/50M), Treatment D with 25% of Pinus taeda and 75% of Malva fibres (25P/75M), and Treatment E with 100% of Malva fibres (100M).

The response-variables were: flexural strength (MOR_F), flexural modulus (MOE_F), density (ρ) and thickness swelling (TS). Eight samples were fabricated for each experimental condition. The effects of the factors were identified via Analysis of Variance (ANOVA) and Tukey test at 95% confidence interval[11]. The statistical analyses were performed using Minitab v.16.

Castor oil based polyurethane resin was used as matrix phase being impregnated via spray process and mechanically mixed. The resin was supplied by Plural Química Company (São Carlos-Brazil). The spray impregnation allowed to combine castor oil and 4,4′-diphenylmethane di-isocyanate oil (MDI) separately (Figure 2a). Subsequently, the particles were mixed by using a Hobart mixer for 5 minutes (Figure 2b). The material was spread within a mould (Figure 2c), and hot-pressed at 4MPa for 10 min at 100ºC (Figure 2d). This type of resin was chosen to avoid the emission of formaldehyde as reported in the literature[25]. The panels were demoulded after 72h according to the polymer curing time. The physical and mechanical properties were obtained in accordance to the Brazilian Standard ABNT NBR 14810:2[26].

X-ray densitometry (QTRS-01X) manufactured by Quintek Measurement Systems was used to measure the density of multilayer particleboard samples (Figure 2f). The X-ray values across the samples were converted to apparent density by the software QMS. Microstructural analyses of the composites were conducted using a Scanning Electron Microscopy HITACHI (TM-3000) with an electron beam acceleration of 15 kV associated with the Energy Dispersive Spectroscopy (EDS).

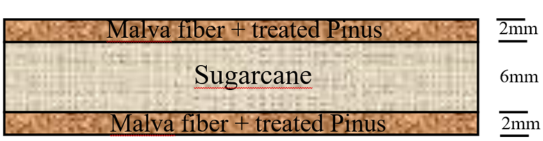

Figure 1. Particleboard manufacturing scheme.

Table 1. Facing material investigation: experimental setup

Treatments	Pinus Taeda (%)	Malva Fibres (%)
A	100	0
B	75	25
C	50	50
D	25	75
E	0	100

3. Results

Table 2 shows the mean values and Tukey test which enables to compare the means by groups, for bulk density (ρ), thickness swelling (TS), flexural strength (MOR_F) and stiffness (MOE_F) properties. Same letters imply treatments with equivalent values.

Based on Tukey test there is no difference in bulk density values, showing a similar letter group (a) for all treatments. The mean bulk density data varied from 0.68 to 0.73 g.cm^{-3} which classify the panels as "medium density" based on the ABNT 14810:1 (0.55 to 0.75 g.cm^{-3})[24] and the Canadian (CS 236-66)[27] and American (ANSI A208:1)[28] (0.6 and 0.8 g.cm^{-3}) standards.

The use of thermally treated wood was motivated by improving the dimensional stability of multilayer particleboard panels. Hemicelluloses are considered the most hydrophilic component of wood and the most thermally sensitive. This is the first component to degrade being revealed by the consumption of hydroxyl groups from the water molecules. For this reason, reductions in equilibrium moisture content and thickness swelling have been reported in the literature[15,29,30]. In order to avoid any environment effect mainly on the core material, the peripherical sides of

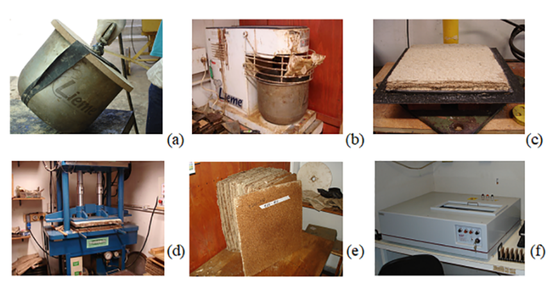

Figure 2. Spray impregnation (a), mechanical mixing (b), particleboard mould (c), uniaxial hot compaction (d), panels after processing (e) and X-ray densitometry tree scanner (f).

Table 2. Physical and mechanical properties of particleboards

	ρ (g.cm^{-3})		TS (%) – 2h		TS (%) – 24h		MOR_F(MPa)		MOE_F(MPa)	
Treatments	Mean ± SD	Tukey	Mean ± SD	Tukey	Mean ± SD	Tukey	Mean ± SD	Tukey	Mean ± SD	Tukey
A (100P)	0.71 ± 0.07	a	1.6 ± 0.2	a	6.4 ± 1.2	a	11 ± 3	a	1188 ± 404	a
B (75P/25M)	0.70 ± 0.09	a	2.4 ± 0.7	ab	7.6 ± 0.7	a	11 ± 4	a	1089 ± 472	a
C (50P/50M)	0.68 ± 0.08	a	3.8 ± 0.8	bc	11.6 ± 1.6	b	11 ± 4	a	1106 ± 462	a
D (25P/75M)	0.73 ± 0.11	a	4.9 ± 1.7	cd	12.6 ± 1.7	b	11 ± 4	a	1165 ± 452	a
E (100M)	0.72 ± 0.08	a	5.8 ± 1.8	d	16.8 ± 2.8	c	12 ± 4	a	1285 ± 396	a

Values are mean ± standard deviation (SD). Means followed by the same letter are not significant (Tukey α = 5%).

the samples were painted with epoxy polymer. Treatment A, made with 100% of treated Pinus taeda in outer layer, achieved the lowest thickness swelling response (group a). In contrast, a higher thickness swelling was reached when 100% of Malva fibres was considered (group d). Malva fibres absorb more water, for this reason the thickness swelling (TS) was higher than those treatments with a large amount of Pinus. The Brazilian Standard ABNT NBR 14810:2[26] recommends no more than 8% for thickness swelling with 2 h; and no more than 14% at 24 h for internal use in dry conditions. Although the statistical analyses show different means for thickness swelling at 2h, all treatments attend the standard limit, except for Treatment E which reaches 16.8 (group c). Fiorelli et al.[31] have investigated the thickness swelling of sugarcane particleboards containing 5 and 8 mm fibre lengths impregnated with castor oil resin. The lowest swelling of 20% has been achieved for 8mm fibres. This behaviour has been attributed to the particle packing effect which affects the composite microstructure and pore sizes. Filho[8] has investigated a sugarcane particleboard reinforced with Pinus or Eucalyptus particles considering a weight fraction of 50%. These panels were produced with 9% of formaldehyde urea and formaldehyde melamine. The thickness swelling values ranged from 7 to 26% for 2h, and from 26 to 36% for 24h. The thermal treatment of Pinus taeda particle was considered the main responsible for the improved thickness swelling. Murata, Watanabe and Nakano[32] have studied the fibre saturation point (FSP) for Spruce wood thermally treated. The FSP has started to decrease above 150ºC.

The minimum values for flexural strength (MOR) and modulus (MOE) considered by the Brazilian[26], Canadian[27] and American[28] standards are: 11.0 and 1800 MPa, 11.2 and 2450 MPa, and 11.0 and 1725 MPa, respectively. All treatments are in accordance to the standard requirements with respect to MOR values. The highest MOR (~12MPa) was achieved by Treatment E consisted of 100% of Malva fibres. In contrast, no treatment could attend the MOE limits required by these standards. It is emphasized the use of a different hardener can enhance the stiffness of the castor oil based polyurethane matrix phase, consequently, being able to attend the structural requirements in future investigations. Mendes et al.[6] have studied sugarcane particleboards combined with eucalyptus wood. The MOR results (~12-15MPa) have reached the minimum requirements found in the normative documents. Particleboards made only with eucalyptus wood achieved higher MOE values (up to 47%) than the Brazilian standard requirements (~1.7-2.7 GPa)[30]. Belini et al.[19] have investigated particleboard panels reinforced with sugarcane bagasse and eucalyptus fibres impregnated with formaldehyde urea. The increase in sugarcane bagasse fraction (at 75%) has led to reduced MOR (~22.9MPa) and MOE (~2.6GPa) properties. In addition, particleboards produced only with sugarcane bagasse did not reach the minimum values recommended by the Brazilian Standard[20]. In the present work, the statistical analyses (see Table 2) showed there is no difference among treatments regarding to flexural strength and stiffness properties.

Figure 3 shows the Scanning Electron Microscopy (SEM) images obtained in backscattering mode for Treatments A-E. All images were obtained at ×50 of magnification, except for Treatment D which was analysed in a higher magnification (×200) to observe the impregnation process. Figure 3a and Figure 3e show the microstructure of particleboards consisted of treated Pinus (Treatment A) and Malva fibres (Treatment E), respectively. Figures 3b and 3d show Treatments B and D, respectively, revealing the increasing amount of Malva fibres from Treatment B to D. Figure 3f shows Treatment D at ×200 of magnification revealing a uniform matrix phase distribution along the dispersive phases. Figure 3f shows Pinus particles in the right side and Malva fibres in the left side of the image. Bertolini[33] and Nascimento[34] have reported that the mechanical properties of particleboards depend mainly on the interaction of the phases, in which the amount and the distribution of the matrix phase in the system is extremely significant, providing higher durability and low water absorption.

The white spots presented in Figure 3f were identified via Energy Dispersive Spectroscopy (EDS). Figure 4 shows the EDS micrographs of sugarcane bagasse (a) and Malva fibre (b) which reveals the presence of silicon elements, being attributed to silicon dioxide (SiO_2). Similar results

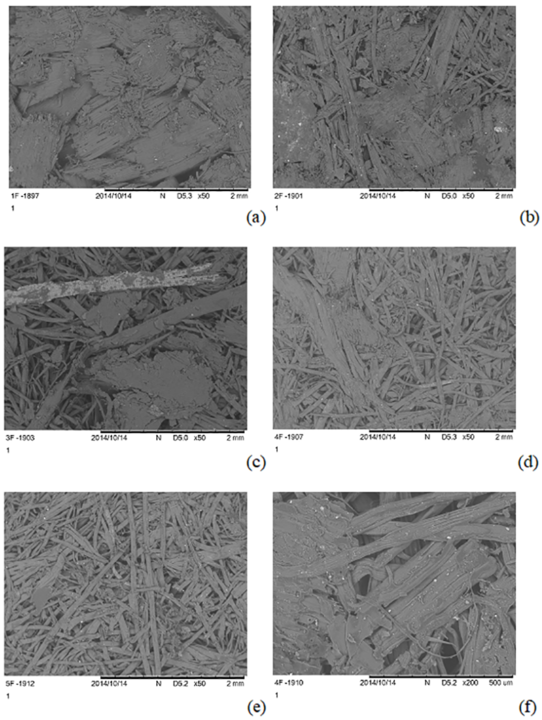

Figure 3. SEM images: (a) treatment A, (b) treatment B, (c) treatment C, (d) treatment D, (e) treatment E at ×50 and (f) treatment D at ×200 of magnification.

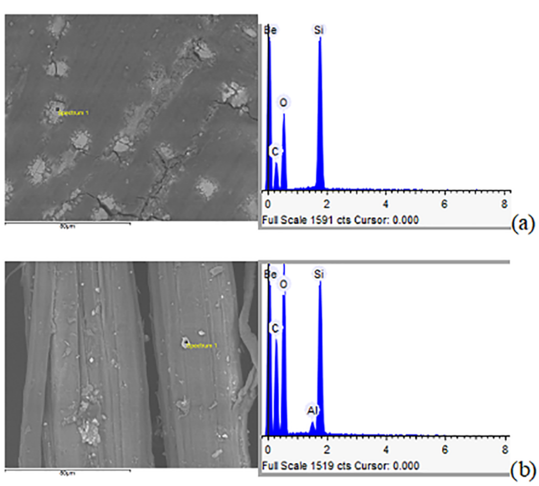

Figure 4. EDS obtained for (a) sugarcane bagasse and (b) Malva fibre

X-ray densitometry was carried out based on a set of ten samples for each Treatment. Table 3 shows the mean density for all treatments.

have been found by Belini et al.[19], in panels produced with different percentages of eucalyptus wood and sugarcane bagasse. Some plants species can absorb silicon from the soil solution in the form of H_4SiO_4, which is commonly found at concentrations that range from 0.1 to 0.6 mM at the pH levels found in most agricultural soils[35].

Figure 5 shows the X-ray densitometry for Treatment A (a) and Treatment B (b). Filho et al.[36] have investigated MDF panels via X-ray densitometry, obtaining a type M profile, which indicates the presence of higher density at the outer layers. This profile is common for X-ray densitometry analyses in MDF panels produced with eucalyptus wood and another forestry species. In the present work, the profile found is inverse to the typical one for MDF, showing higher density for the core region. A slight increase in density was identified for treatment B, in which 25% of Malva fibres are incorporated.

Table 3. Mean density obtained via X-ray densitometry.

Treatments	Mean density (Kg.m^{-3})
A (100P)	584 ± 106
B (75P/25M)	596 ± 99
C (50P/50M)	550 ± 69
D (25P/75M)	582 ± 62
E (100M)	603 ± 76

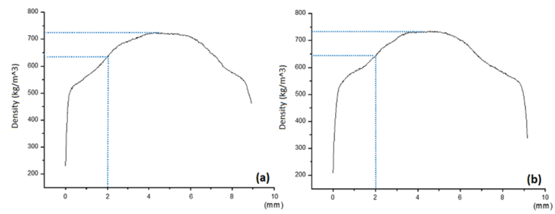

Figure 5. Profile densitometry by X-ray densitometry: (a) Treatment A and (b) Treatment B

4. Conclusions

All particleboards were classified as medium density panels based on the Brazilian, Canadian and American standards. The thermal treatment performed in Pinus wood makes the panel more hydrophobic. Treatment A, consisted of 100% of Pinus wood, achieved lower levels of thickness swelling at 2h and 24h. In contrast, treatment E, made with 100% of Malva fibres, achieved higher thickness swelling values, however those at 24h did not achieve the standard requirements. All treatments reached the minimum flexural strength level, in contrast, the elastic moduli of the panels did not achieve the standard requirements. The presence of silica content in sugarcane and Malva fibres was identified by SEM and EDS, being attributed to the plant absorption in soil. X-ray densitometry revealed higher density in the core material. The incorporation of Malva fibres also contributes to increase the density of the sandwich panels. Tukey test demonstrated that both flexural strength and modulus responses for all treatments was statistically similar. For this reason, Treatment B, consisted of 75% of Pinus taeda wood and 25% of Malva fibres, demonstrated to be a promising hybrid particleboard, combining strength, low-cost and lower thickness swelling values.

5. References

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