ISSN 1807-1929
Revista Brasileira de Engenharia Agrícola e Ambiental
v.22, n.10, p.707-712, 2018
Campina Grande, PB, UAEA/UFCG – http://www.agriambi.com.br

DOI: http://dx.doi.org/10.1590/1807-1929/agriambi.v22n10p707-712

Physical performance of particleboards using Castor oil-based adhesive

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Key words:
particleboards
Pinus elliottii
oat hulls
analysis of variance (ANOVA)
wastes

ABSTRACT
Reconstituted wood panels are widely used in various branches of the wood industry, such as in the furniture sectors and various segments of urban and rural construction. In Brazil, agricultural and forestry residues are abundantly generated, making feasible the study, development and application of alternative and sustainable materials, produced from the use of these wastes. The aim of this study was to produce high-density homogenous panels with wood residues of *Pinus elliottii* and oat hulls (*Avena sativa*), pressed with two types of adhesive, castor oil-based polyurethane and melamine formaldehyde, and to evaluate the physical performance of these panels. The physical performance of the panels was evaluated based on the ABNT NBR 14810: 2006, ANSI A208.1: 1999 and BS EN 312: 2003 standards. Analysis of variance (ANOVA) was performed to test the influence of the factors percentage of oat hull, percentage of adhesive and type of adhesive, and the interactions between these factors on the physical properties of the panels. The results indicated physical properties, in some treatments superior to the requirements stipulated by the consulted standards. It was concluded that the panels produced with the castor oil-based polyurethane adhesive showed better physical performance when compared with the panels produced with the melamine formaldehyde adhesive.

Palavras-chave:
painéis aglomerados
Pinus elliottii
casca de aveia
análise de variância (ANOVA)
resíduos

Desempenho físico de painéis de partículas usando adesivo à base de óleo de mamona

RESUMO
Os painéis de madeira reconstituída são muito empregados em vários ramos da indústria da madeira, como nos setores moveleiros e vários segmentos da construção civil urbana e rural. No Brasil são gerados resíduos agrícolas e florestais abundantemente, tornando viável o estudo, desenvolvimento e aplicação de materiais alternativos e sustentáveis, produzidos a partir do aproveitamento destes resíduos. O objetivo deste trabalho foi produzir painéis de partículas homogêneos de alta densidade, com resíduos de madeira de *Pinus elliottii* e casca de aveia (*Avena sativa*), prensados com dois tipos de adesivo, poliuretano à base de óleo de mamona e melamina formaldeído, e avaliar o desempenho físico destes painéis. O desempenho físico dos painéis foi avaliado com base nas normas ABNT NBR 14810:2006, ANSI A208.1:1999 e BS EN 312:2003. Foi realizado uma análise de variância (ANOVA) para testar a influência dos fatores percentual de casca de aveia, percentual de adesivo e tipo de adesivo, e as interações entre estes fatores nas propriedades físicas dos painéis. Os resultados apontaram propriedades físicas, em alguns tratamentos superiores aos requisitos estipulados por normas consultadas. Conclui-se que os painéis produzidos com o adesivo poliuretano à base de óleo de mamona apresentaram melhor desempenho físico, se comparado aos painéis produzidos com o adesivo melamina formaldeído.

Ref. 184953 – Received 04 Sept, 2017 • Accepted 27 Jun, 2018 • Published 26 Aug, 2018

Introduction

The development and characterization of new materials have been continuously experimented with applications of these materials, such as more versatile and efficient applications, reduction of energy costs and waste generation, lower environmental impacts and products with less impact on the planet (Silva et al., 2015).

The use and application of wood products have grown by leaps and bounds in recent years, not only in Brazil but throughout the world. Wood panels are the main raw material in a range of other wood product industries, such as flooring, furniture, packaging, shipbuilding and various construction segments. Brazilian consumption of wood-based panels increased by approximately 15.9% in 2017 compared with 2016, with billing increasing from 246 to 285 million dollars. The expansion and growth of the wood-based panel industries significantly increases the demand for raw materials, forcing the search for other materials (Varanda et al., 2014; Christoforo et al., 2016).

The generation of waste is significant in Brazil. In 2016, about 19.1 million cubic meters of wood-based waste were generated. In the same year, the Brazilian production of particleboards was approximately 3.0 million cubic meters (FAO, 2018).

Among these agroindustrial residues, the hull generated in the processing of the oat cereal has great potential, especially in relation to the amount of raw material that, according to Webster (1986), represents about 30% of the mass of the cereal. Brazilian production of oats was approximately 884,000 t in 2016, that is, approximately 265,000 t of oat hulls were generated (IBGE, 2016).

Adhesive is an important component in the production of panels based on wood and its derivatives, which can represent up to 50% of the total cost of the panel. In relation to the melamine formaldehyde adhesive, it has intermediate physical and mechanical performance between the adhesives urea formaldehyde and phenol formaldehyde (Iwakiri, 2005). Polyurethane adhesives impart high moisture resistance, better physical and mechanical properties, compared with panels bonded with phenolic resins, in addition to not emitting formaldehyde (Ferro et al., 2016). Several studies have been developed with polyurethane adhesive based on castor oil, under alternative lignocellulosic materials for the production of particleboards (Varanda et al., 2013; Gava et al., 2015; Vidil et al., 2016; Cravo et al., 2017; Machado et al., 2017).

In this context, it justifies the study of alternative inputs in the production of particleboard panels. The aim of this study was to produce high-density homogenous particleboards with wood residues of *Pinus elliottii* and oat hulls (*Avena sativa*), subjected to pressure with two types of adhesives, polyurethane based on castor oil and melamine formaldehyde. In addition, it aimed to evaluate the physical performance of the panels (thickness swelling for 2 and 24 h period, water absorption for 2 and 24 h period, density and compaction ratio).

Material and Methods

This study was conducted at the University of São Paulo (USP), Campus of São Carlos, specifically in the Wood and Timber Structures Laboratory (LaMEM), located at the Department of Structural Engineering (SET) of the São Carlos School of Engineering (EESC).

Particleboards were made using wood wastes of *Pinus elliottii* and wastes of oat hulls (*Avena sativa*). The wood of *Pinus elliottii* had an apparent density of 480 kg m^{-3} (12% of humidity) and oat hulls material had an apparent density of 290 kg m^{-3} (12% of humidity). Wood wastes of *Pinus elliottii* were obtained in the city of São Carlos, state of São Paulo. The oat hulls used were obtained from an industry in the sector, located in the city of Porto Alegre, Rio Grande do Sul State.

Two types of adhesives were used: polyurethane based on castor oil (PU) and melamine formaldehyde (MF). The PU adhesive was adopted due to its good performance obtained in previous studies, conducted at the LaMEM/SET, with wood panels (Bertolini et al., 2014; Macedo et al., 2015; Nascimento et al., 2017). This is an adhesive of mostly natural and renewable origin. The PU adhesive used is bicomponent type, at a ratio of 1:1 between prepolymer and polyol, with a solid content of 100%. One of the components (polyol) is derived from vegetable oil, with a density of 1.10 g cm^{-3}, and the other component (prepolymer) is the polyfunctional isocyanate, with density of 1.24 g cm^{-3}.

Melamine formaldehyde adhesive was adopted because it is widely used in wood-based panels industries in Brazil, although its use is controlled and questionable in countries with strict environmental control because it emits formaldehyde, an undesirable substance that is harmful to human health (Samlaic, 1983). The melamine formaldehyde adhesive (MF) used showed the following physicochemical characteristics: pH at 25 °C of 8.48; solid content (3H at 105 °C) of 71.28%; density at 25 °C of 1.26 g cm^{-3}; Brookfield viscosity at 25 °C of 756 centipoises and free formol equal to 0.19%. The melamine formaldehyde adhesive (MF) had an addition of 1.5% ammonium sulfate, relative to the mass of the adhesive. The ammonium sulfate has the catalytic function, i.e., to accelerate the adhesive curing process. It was decided not to use paraffin because the MF adhesive shows good resistance to humidity.

From the preliminary tests, the experimental design was defined. It originated 20 treatments (Tr) as shown in Table 1. For each of the 20 treatments (Tr), five identical particleboards were produced, totaling 100 particleboards. The panels produced had nominal dimensions of 280 × 280 × 10 mm.

The particles of both materials were generated in a Wiley-type mill, using a 2.8 mm aperture sieve. After the generation of the particles, their moisture content was determined in the oven at a temperature of 105 ± 2 °C.

The next step was the preparation and application of the resin. The particles of *Pinus elliottii* and oat hulls were generated in a mill. Then, the particles of both materials and the adhesive were weighed according to the stipulated amounts. Then, the particles and the adhesive were added in the blending equipment until the homogenization of these materials. The adhesive proportions used were 11 and 13% relative to the mass of the particles. The amount of particles used in each panel was defined from the density range (from 850 to 950 kg m^{-3}) and from the volume adopted for the panels (nominal dimensions of 280 × 280 × 10 mm). The particles and the adhesive remained in the glue for at least 5 min.

Table 1. Experimental design adopted

Treatments (Tr)	% OH	% Adhes	TA	Treatments (Tr)	% OH	% Adhes	TA
Tr1	0	11	MF	Tr11	50	13	MF
Tr2	0	11	PU	Tr12	50	13	PU
Tr3	0	13	MF	Tr13	75	11	MF
Tr4	0	13	PU	Tr14	75	11	PU
Tr5	25	11	MF	Tr15	75	13	MF
Tr6	25	11	PU	Tr16	75	13	PU
Tr7	25	13	MF	Tr17	100	11	MF
Tr8	25	13	PU	Tr18	100	11	PU
Tr9	50	11	MF	Tr19	100	13	MF
Tr10	50	11	PU	Tr20	100	13	PU

Tr - Treatment; % OH - Oat hulls percentage; % Adhes - Adhesive percentage; TA - Type of adhesive; MF -Melamine formaldehyde adhesive; PU - Polyurethane adhesive based on castor oil

After complete homogenization of the adhesive with the particles, the mixture was prepressed in order to form the particle "mattress". It received a force of 1 kN, equivalent to a pressure of 0.013 MPa. The prepressing of the panel was carried out using a manual mechanical press, manufactured in-house.

Then, the particle "mattress" already preformed with a thickness of 2 cm (measured by a spacer) was sent to the press. The panels were pressed in a semiautomatic press, with a capacity of 800 kN and a maximum temperature of 200 ºC. The pressing conditions used in this study were: 10 min time, 4 MPa pressure and temperatures of 100 ºC for the polyurethane adhesive based on castor oil, and 160 ºC for the melamine formaldehyde adhesive. After pressing, all the panels produced were conditioned for at least 72 h, aiming at the stabilization and complete cure of the adhesive. After the conditioning period, the panels were subjected to squaring and subsequent sectioning to remove the specimens. Initially about 10 mm were removed from each end of the panel, in a circular saw cutter. The panels had nominal dimensions of 260 × 260 × 10 mm. From the squaring, the panels were sectioned in the dimensions of the specimens. A circular saw was used for the squaring and sectioning of the panels.

All physical characterization of the panels was performed according to the NBR 14810 standard (ABNT, 2006). All the results obtained in the tests were submitted to statistical analysis in order to evaluate if the factors and levels adopted in the experimental design had influence on the physical performance of the panels produced at a significance level of 5%. The statistical analysis adopted was an analysis of variance (ANOVA), using Minitab® software, version 14.

Six samples were removed from each panel, so that it was possible to analyze different physical properties (thickness swelling for 2 and 24 h period, water absorption for 2 and 24 h period, density and compaction ratio), except the moisture content (MC). For this, a specimen was sampled for each of the 10 randomly selected panels, among the 100 panels produced, only to verify the average MC of the evaluated panels. The MC properties of the panels were not subjected to analysis of variance (ANOVA). It was done in factorial arrangement (5 x 2 x 2) was used, corresponding to 5 fractions of particles, 2 percentages of adhesive and 2 types of adhesives. ANOVA had as null hypothesis (H_0) the equivalence of means between treatments, and non-equivalence as alternative hypothesis (H_1). Thus, p-value of the test above the level of significance implies accepting H_0, rejecting it otherwise.

Results and Discussion

The results obtained for the physical properties of the panels were compared with the requirements established by the standards NBR 14810 (ABNT, 2006), A208.1 (ANSI, 1999) and EN 312 (BS, 2003).

The average moisture content of the panels evaluated was 9.2%. The particleboards produced by Weber & Iwakiri (2015) had a moisture content in the range of 7.3 to 9.0%, that is, similar to the mean moisture content of the panels of this study. According to Iwakiri (2005), the ideal moisture content of the panels is below 10%.

Table 2 shows the mean values (x) and coefficients of variation (cv) of the physical properties of the panels.

In Table 2, 10 treatments (2, 4, 6, 8, 10, 12, 14, 16, 18, 20, treatments produced with PU adhesive) showed a thickness swelling inferior to 8%, meeting the requirement of the standard NBR 14810 (ABNT, 2006). For the thickness swelling property for 24 h period, nine treatments (Tr 1 to 4, 6 to 8, 12 and 20) had a swelling of less than 16%, according to EN 312 (BS, 2003).

The water absorption properties (for 2 and 24 h periods) do not have requirements in any of the four standards consulted. For the water absorption property for 2 h period, the mean result obtained by Gava et al. (2015) is similar to the range of results obtained in this study. Particleboards produced by Fiorelli et al. (2015) exhibited water absorption of up to 69.8%, i.e., higher than the results obtained here (water absorption in the range of 2.9 to 40.5%). For the water absorption property for 24 h period, the values obtained in this study (water absorption in the range of 12.8 to 43.5%) are similar to the mean value of 38.8% presented by Gava et al. (2015). This large variation of the physical properties is associated to the different materials used in the production of the panels, due to the incompatibility of these materials with the adhesives used and due to the pressing variables such as temperature, pressure and time (Iwakiri, 2005).

The density of the panels exhibited great variation among the evaluated treatments, with results in the range of 882 to 1066 kg m^{-3}. All treatments showed high density (above 800 kg m^{-3}). This variation was also observed in other studies, such as Bertolini et al. (2014), Fiorelli et al. (2015) and Gava et al. (2015). According to Iwakiri (2005), differences in the final density of the panel are associated with the material used in the respective process of manufacture, variables such as the moisture content of the particles and the differences in density between the particles used in the manufacture of such

Table 2. Results of the physical properties of the panels

Tr.	Stat.	TS 2 h	TS 24 h	WA 2 h	WA 24 h	D	CR
		(%)				(kg m^{-3})	
1	x̄	12.2	14.5	30.3	37.4	882	1.84
	cv (%)	9.0	8.1	18.5	12.2	2.5	2.50
2	x̄	7.6	12.7	12.5	29.2	892	1.86
	cv (%)	15.1	12.2	17.2	17.4	2.4	2.40
3	x̄	9.6	12.0	22.3	29.8	921	1.92
	cv (%)	11.0	7.0	14.6	12.0	1.9	1.90
4	x̄	5.6	11.3	5.1	18.4	931	1.94
	cv (%)	17.5	15.9	12.4	15.6	4.9	4.90
5	x̄	15.9	18.5	33.9	41.1	907	2.10
	cv (%)	10.0	4.4	9.2	6.5	3.3	3.30
6	x̄	7.6	13.9	6.7	26.7	972	2.25
	cv (%)	16.8	18.2	15.2	14.6	4.9	4.90
7	x̄	14.6	15.7	28.0	37.2	926	2.14
	cv (%)	10.4	9.6	18.6	15.5	4.1	4.10
8	x̄	5.5	12.9	5.7	21.2	980	2.27
	cv (%)	17.8	9.6	19.3	16.9	5.1	5.10
9	x̄	21.3	22.8	40.5	43.5	966	2.51
	cv (%)	10.7	13.9	18.2	14.4	1.9	1.90
10	x̄	7.8	16.8	6.9	26.9	941	2.44
	cv (%)	18.9	10.5	10.0	16.1	9.1	9.10
11	x̄	16.7	17.9	29.5	35.9	968	2.51
	cv (%)	8.3	5.5	9.8	10.7	2.6	2.60
12	x̄	5.8	11.8	5.0	16.5	990	2.57
	cv (%)	12.2	16.8	14.7	9.6	7.1	7.10
13	x̄	27.7	28.8	39.1	42.5	977	2.90
	cv (%)	6.8	6.4	12.8	10.6	2.7	2.70
14	x̄	7.5	22.7	7.1	30.8	995	2.95
	cv (%)	19.7	9.2	12.3	15.2	2.6	2.60
15	x̄	18.9	20.3	30.1	34.1	995	2.95
	cv (%)	9.0	6.1	8.2	6.6	1.8	1.80
16	x̄	6.8	17.1	7.0	29.6	931	2.76
	cv (%)	13.6	10.8	9.3	17.2	7.6	7.60
17	x̄	27.5	28.7	36.8	40.4	997	3.44
	cv (%)	9.5	6.8	18.7	12.9	4.3	4.30
18	x̄	6.0	19.2	4.0	20.8	1022	3.52
	cv (%)	17.3	13.4	14.5	18.4	6.6	6.60
19	x̄	21.6	25.1	29.1	32.9	1066	3.68
	cv (%)	9.6	2.6	16.3	11.6	3.0	3.00
20	x̄	2.6	12.0	2.9	12.8	1015	3.50
	cv (%)	13.6	17.0	18.1	16.0	7.2	7.20

Tr - Treatment; Stat. - Statistics of average values (x) and coefficient of variation (cv); TS 2 h - Thickness swelling for 2 h period; TS 24 h - Thickness swelling for 24 h period; WA 2 h - Water absorption for 2 h period; WA 24 h - Water absorption for 24 h period; D – Density; CR - Compaction ratio

panels. High-density particleboards must have a density of 800 kg m^{-3} or higher, according to A208.1 (ANSI, 1999) and Iwakiri (2005).

As for the compaction ratio of the panels (Table 2), the mean values obtained were in the range of 1.84 to 3.68, very similar to those found by Mendes et al. (2010), which obtained values in the range of 1.39 to 3.12, for panels produced with sugarcane bagasse and *Eucalyptus* wood, and with adhesives phenol formaldehyde and urea formaldehyde. For both studies, the wide range of compaction ratio values is associated with the different densities of the materials used in the panels manufactured.

Normality and homogeneity tests of variances for the ANOVA residuals referring to the physical properties of panels were evaluated by Anderson-Darling, Bartlett and Levene methods (Christoforo et al., 2016). The ANOVA residuals, per response variable investigated, showed normal distribution. The variances of residuals between treatments were equivalent, and the residuals per property were independent. Table 3 presents the ANOVA results for the investigated physical properties.

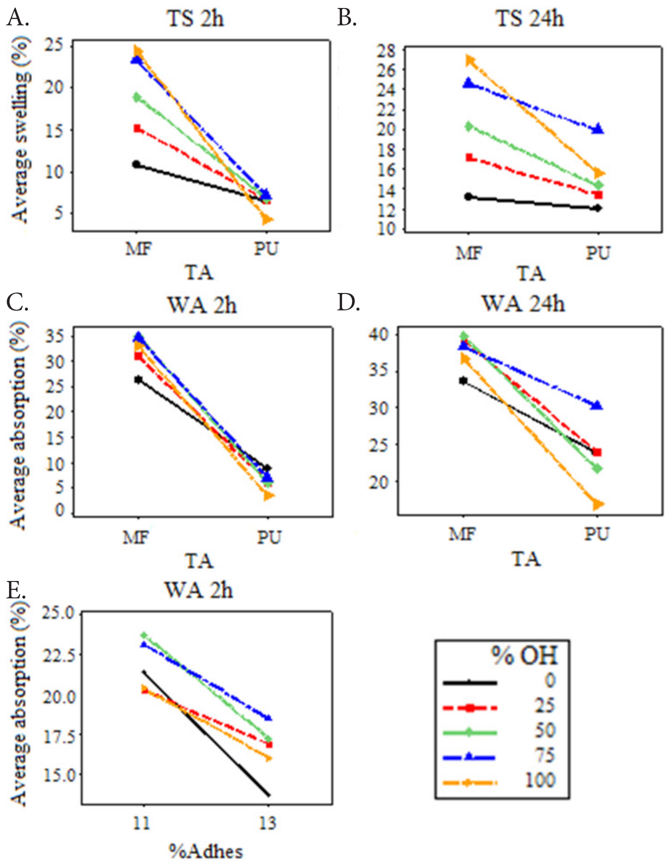
P-values lower than or equal to 0.05 are underlined and considered significant at a significance level of 5% (95% confidence) over the evaluated property. When the interaction between the factors (2:2, 3:3) was considered as significant in any of the properties evaluated, main effects graphs were generated to support the interpretation of interaction effects.

In addition, the homogeneity in the panel production was also evaluated by the adjusted coefficient of determination (R^2 adj) for each property investigated. All physical properties evaluated showed sufficient homogeneity, with the exception of density, for which adjusted R^2 reached 43.94%.

Figure 1 shows the graphs of interaction between factors for the investigated physical properties, considered significant by ANOVA, according to Table 3.

It can be observed, in Figures 1A to D, that the panels produced with the polyurethane adhesive based on castor oil (PU) had lower mean values of thickness swelling for 2 and 24 h periods, and water absorption for 2 and 24 h periods, when compared with panels produced with melamine formaldehyde adhesive (MF). The panels produced with the PU adhesive showed better physical performance.

In Figure 1E, panels produced with a higher percentage of adhesive (13%) had lower mean values of water absorption for 2 h period compared with panels produced with 11% of adhesive. Panels produced with 13% adhesive showed better physical performance.



% OH - Oat hulls percentage; TA - Type of adhesive; TS 2 h - Thickness swelling for 2 h period; TS 24 h - Thickness swelling for 24 h period; WA 2 h - Water absorption for 2 h period; WA 24 h - Water absorption for 24 h period; % Adhes - Adhesive percentage

Figure 1. Interaction between factors for physical properties. Interaction % OH x TA: for TS 2 h (A); for TS 24 h (B), for WA 2 h (C), for WA 24 h (D) and interaction % OH x % Adhes for WA 2 h (E)

Table 3. ANOVA results regarding the physical properties

Factors and interactions	TS 2 h	TS 24 h	WA 2 h	WA 24 h	D	CR
% OH	0.000	0.000	0.116	0.073	0.000	0.000
% Adhes	0.000	0.000	0.000	0.000	0.070	0.107
TA	0.000	0.000	0.000	0.000	0.500	0.764
% OH × % Adhes	0.261	0.093	0.019	0.238	0.255	0.272
% OH × TA	0.000	0.000	0.000	0.000	0.060	0.145
% Adhes × TA	0.072	0.576	0.143	0.927	0.198	0.124
% OH × % Adhes × TA	0.106	0.051	0.236	0.256	0.150	0.107
R^2 adj (%)	96.25	89.87	93.50	81.08	43.94	94.97

% OH - Oat hulls percentage; % Adhes - Adhesive percentage; TA - Type of adhesive; R^2 adj - adjusted coefficient of determination of ANOVA; TS 2 h - Thickness swelling for 2 h period; TS 24 h - Thickness swelling for 24 h period; WA 2 h - Water absorption for 2 h period; WA 24 h - Water absorption for 24 h period; D – Density; CR - Compaction ratio

Conclusions

1. The panels were classified as high-density (above 800 kg m^{-3}).

2. Most of the treatments evaluated met at least one of the requirements of the standards consulted.

3. Panels produced with polyurethane adhesive based on castor oil showed better physical performance than panels produced with melamine formaldehyde adhesive.

4. The panels with 75 and 100% oat hull particles had higher densities as well as lower physical performances than panels with 0, 25 and 50% of oat hull particles.

Acknowledgments

The authors are grateful to the Coordination for the Improvement of Higher Education Personnel (CAPES) for the support to the study.

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