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Efficiency of castor oil-based polyurethane in the production of plywood panels

Eficiência do poliuretano à base de óleo de mamona na produção de painéis compensados

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Abstract

Plywood panels are produced with polymeric adhesives derived from petroleum that can be hazardous to health and the environment, since they are made from non-renewable sources and contain organic solvents in their composition, which can cause diseases such as cancer. Polyurethane adhesives derived from castor oil-based are an alternative to those materials, because it is biodegradable and non-hazardous. The aim of this work was to evaluate the mechanical properties of plywood panels made with polyurethane adhesive derived from castor oil-based. Panels were produced with five sheets of *Pinus caribaea bahamensis* wood and three different adhesives: urea-formaldehyde, phenol formaldehyde, and polyurethane with a glue spread line of 180 g/m^2 for phenolic adhesives, and 120, 140, 160, 180 g/m^2 for polyurethane. The panels were evaluated in terms of specific mass, shear strength before and after immersion on boiling and cold water, in cycle and without immersion, and bending strength parallel and perpendicular to the fibers orientation. Results were statistically analyzed. The plywood panels produced with polyurethane adhesive derived from castor oil have superior shear strength and bending strength similar than those produced with the others tested adhesives, and the weight of 120 g/m^2 is the most suitable for bonding.

Keywords: plywood; biodegradable polyurethane; mechanical properties

Resumo

Os painéis compensados são produzidos com adesivos poliméricos derivados do petróleo que podem ser prejudiciais à saúde e ao meio ambiente, pois são provenientes de fontes não renováveis e contêm solventes orgânicos em sua composição, que podem causar doenças como o câncer. Adesivos de poliuretano derivados de óleo de mamona são uma alternativa a esses materiais, pois são biodegradáveis e não prejudiciais. O objetivo deste trabalho foi avaliar as propriedades mecânicas de painéis de compensados feitos com adesivos derivados de poliuretano à base de óleo de mamona. Os painéis foram produzidos com cinco folhas de madeira de *Pinus caribaea bahamensis* e três diferentes adesivos: uréia-formaldeído, fenol-formaldeído e poliuretano com gramatura de linha de cola de 180 g/m2 para adesivos fenólicos, e 120, 140, 160, 180 g/m^2 para poliuretano. Os painéis foram avaliados em termos de peso específico, resistência ao cisalhamento após imersão em água fervente e fria, em ciclo e sem imersão, e resistência à flexão paralela e perpendicular à orientação das fibras. Os resultados foram analisados estatisticamente. Os painéis produzidos com adesivo de poliuretano derivado de óleo de mamona apresentam resistência ao cisalhamento superior e resistência à flexão semelhante aos produzidos com os demais adesivos testados, sendo a gramatura de 120 g/m^2 a mais indicada para a colagem.

Palavras-chave: painel laminado; poliuretano biodegradavel; propriedades mecânicas

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INTRODUCTION

Wood has a set of unique properties when employed as a construction material, such as strength, flexibility, fire resistance, durability, and insulation (IWAKIRI, 2005). Natural wood has an anisotropic and heterogeneous structure, so the main directions of strain are not necessarily those of stress (NGARGUEUDEDJIM; ANNOUAR, 2015)

In order to overcome this limitations, wood panels are produced worldwide, and such as panels that can be plywood, MDF, agglomerated, structural wood, OSB, and laminated wood products (KABOORANI et al., 2012).

Plywood panels are made with wood sheets glued in such a way that the fibers of the individual sheets are arranged perpendicularly to each other. Such arrangement increases the isotropy of the wood, giving the material high dimensional stability and uniform mechanical strength (IWAKIRI, 2005).

The production of plywood panels was boosted by the development of synthetic resins, such as urea-based resins and phenolic resins (IWAKIRI, 2005; SILVA, 2015). These resins are produced with formaldehyde solvent, and it can release this compound for months or years, even though the panels are coated with different materials (BREGINSKI, 2015; SALTHAMMER, 2010).

Inhalation of formaldehyde can cause dizziness, headaches, nausea, vomit, lacrimation, and dryness of the airways (IARC, 2004; HAUPTMANN et al., 2004; IARC, 2015). In July 2004, the International Agency for Research on Cancer – intergovernmental agency which is part of the United Nations Organization -changed the classification of formaldehyde from group 2, probably carcinogenic to humans, to group 1, carcinogenic to humans (IARC, 2004; SOLAL et al., 2008).

In this context, the use of adhesives derived from vegetal oils has advantages, such as the reduction in the production of residues that aren't easily degradable, and the elimination of hazards due to inhalation of formaldehyde (AZEVEDO, 2009).

The use of adhesive polyurethane derived from castor oil was considered adequate for adhesion for Edge Glued Panel with *Pinus taeda* and too adhesion for MDF in furniture production (MOLLEKEN et al., 2016; PEREIRA et al., 2016).

The aim of this work was to evaluate the physical and mechanical properties of plywood panels made withpolyurethane adhesives derived from castor oil and to compare the results obtained with other adhesives.

MATERIALS AND METHODS

Sheets of *Pinus caribaea bahamensis* were taken from a 17 years old tree, planted in Itararé-SP, with 33cm of diameteratbreastheight (DBH), with dimensions of 500x500x2 mm and were donated for Valor Florestal Industry. These sheets were oven dried during 24 hours at 100 °C ± 2 °C until the humidity was lowered to 6% to 8% by weight. The employed adhesives were the castor oil-based polyurethane (PU) with solid content of 100%, density of 1,074g/cm^3, pH 5,5; Urea formaldehyde (UF) with solid content of 66%, viscosity of 300 cP, pH = 7,6, densityof 1,28 g/cm^3 and gel time of 60s; Fenol-formaldehyde (PF) with solid content of 51,57%, viscosity of 503cP, pH = 12,66, densityof 1,22 g/cm^3 and gel time of 7 minutes (ROSA, 2015); the PU was donated for Cequil Industria e Comércio de Adesivos de Araraquara, the UF and FF was donated for Hexion.

The process were divided in two stages: Stage 1 – Production and comparison of plywood with UF, PF and PU adhesive with glue grammage line of 180 g/m^2; Stage 2 – Production and comparison of plywood with PU adhesive with glue grammage line of 180, 160, 140 and 120 g/m^2. All the panels were produced with five wood sheets arranged perpendicular to each other. The parameters used to produce the panels are shown in Table 1.

In total, three panels for each condition described in Table 1 were produced. The samples were cut for the shear strength test,three-point bending test and placed in a climatic chamber at 20 ± 3 °C and relative humidity of 65 ± 5 °C.

The specific mass analyses were made according to the NBR 9485 (ABNT, 2011). Shear tests were made after immersion in both cold and hot water, in cycles, and without immersion, according to the EN 314:1996 Standard (EUROPEAN STANDARD, 1996). Three-point bending tests of specimens in parallel and perpendicular directions regarding the fibers alignment were also conducted, according

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Table 1 – Parameters used to produce the plywood panels
Tabela 1 – Parâmetros utilizados na produção dos painéis compensados

Stage	Adhesive	Temperature (ºC)	Weight (g/m²)	Time (min)	Pressure (MPa)
1	Urea-formaldehyde	110	180	8	1
1	Phenol formaldehyde	130	180	10	1
1	Castor oil-basedpolyuretahane	90	180	20	1
2	Castor oil-basedpolyuretahane	90	160	20	2
2	Castor oil-basedpolyuretahane	90	140	20	2
2	Castor oil-basedpolyuretahane	90	120	20	2

to the EN 310:1993 Standard (EUROPEAN STARDAD, 1993). Shear and bending tests were carried out in a universal testing machine EMIC DL 2000.

The results were calculated in statistical program Statgraphics Centurion 16.1.11. The data were submitted to Grubb's test to assess the occurrence of outliers, Kolmogorov'stest to test the adherence of data, Bartlett's test for homogeneity of variance and analysis of variance. When rejected the null hypothesis, was applied Tukey HDS with 95% probability.

RESULTS

Stage 1

Specific mass

The specific mass of the plywood panels with different adhesives are shown in Fig. 1.

The specific mass of all panels were statistically equivalent according to the Tukey's HDS test, ranging from 541 to 565 kg/m³ in Stage 1, Fig. 1. The specific mass of *Pinus caribaea hondurensis* with 12% humidity, as reported by Rezende et al. (2008), varied from 510 kg/m³ to 560 kg/m³, showing that the specific mass of the plywood panels was slightly affected by the adhesives. The process of pressurizing the wooden sheets results in a volume reduction of the material, however, the mass of the panel remains constant, hence increasing the plywood's specific mass.

Almeida et al.(2012) in a study on plywood produced with *Pinus* sp. hybrids with double-glue phenol-formaldehyde adhesive with a glue-line weight of 380 g/m², had a specific mass of 413 kg/m³, a value below that found in this study.

Ayrilmis and Ozbay (2017) produced plywood panels of *Pinus sylvestre is* with fenol-formaldehyde resin, modified with 20% of a bio-oil with a glue-line grammage of 220 g/m²,obtaining a specific mass of 770 kg/m³.

Ozturk et al. (2016) produced plywood panels of *Pinus sylvestris* with a polyethilene adhesive; when utilizing a glue-line grammage of 160 g/m² the resultant density was of 530 kg/m³, which is statistically similar to the one obtained with a glue-line weight of 240 g/m², 528 kg/m³.

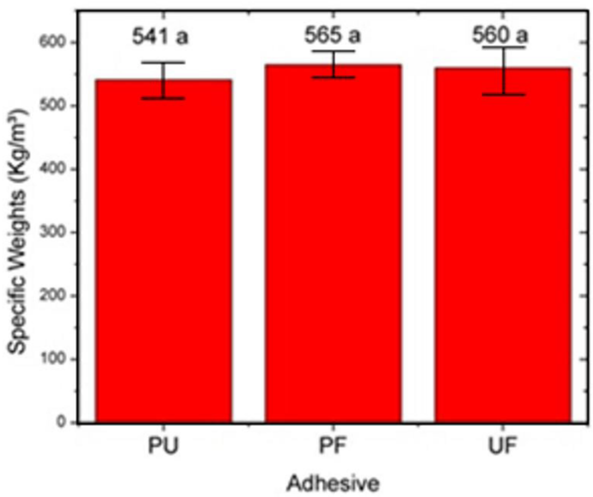

Figure 1 – Specific weight of the plywood panels with humidity 12% at Stage 1
Figura 1 – Massa específica dos painéis compensados com 12% de umidade da Etapa 1

Shear strength

The shear strength of the plywood panels for different conditions are shown in Table 2.

From the data shown in Table 2, it is possible to observe that plywood panels produced with adhesive derived from castor oil-based polyurethane had the highest shear strength for all treatments, and these panels attends the requirements of the EN 314 Standard (EUROPEAN STANDARD, 1996), which is a minimum strength of 1 MPa.

For panels with shear strength between 0.2 and 0.4 MPa, the EN 314 standard establishes that the failure in the wood should be higher than 80%, when the resistance is between 0.4 and 0.6 MPa, the fault must be higher to 60%, in case of resistance between 0.6 and 0.8 MPa the failure must be greater than 40%, and in panels whose shear strength is higher than 1 MPa there is no requirement for failure in the wood.

Failure percentage refers to the amount of wood remaining in the fracture area. A high percentage indicates that the rupture occurred in the wood, not in the contact point between the wood and the adhesive, indicating that the strength of the adhesive is higher than the wood strength.

For dry and cold water conditions, PU plywood panels showed a higher failure percentage than the other ones, as can be seen in Table 2. For boiling and cycle conditions, such percentage was lower than compared to the panels made with phenol formaldehyde adhesive.The chemical bonding between the isoscianide, present in the polyurethane, and free hydroxils of the wood promote an increased adhesion between the composite's components when compared to phenolic adhesives.However, when submitted to cycle and boiling conditions, in which specimens are submerged in water at 100°C, the polyurethane's stiffness and adhesion diminish, since it's glass transition temperature (Tg) is of about 60° C (AZEVEDO, 2009). This also reduces the percentage of specimens that presented failure in the wood when compared to the fenol-formaldehyde adhesive.

Table 2 – Shear strength (RGL – Resistance of the Glue Line) and failure percentage for the wood panels with different adhesives (180 g/m^2)
Tabela 2 – Cisalhamento (RLC – Resistência da Linha de Cola) e porcentagem de falha na madeira para painéis com diferentes adesivos (180 g/m^2)

Treatments		PU	PF	UF
Dry	RGL (MPa)	2.71 a (11.71%)	2.26 b (10.43%)	1.65 c (12.64%)
	Failure (%)	76.33	40.33	21.33
Cold water	RGL (MPa)	1.99 a (16.03%)	0.91 c (24.73%)	1.17 b (20.76%)
	Falha (%)	10.67	6.67	8.33
Cycle	RGL (MPa)	1.11 a (21.28%)	0.79 b (18.52%)	-
	Falha (%)	2.50	5.67	-
Boiling	RGL (MPa)	1.09 a (51.67%)	0.88 a (16.16%)	-
	Falha (%)	5.38	7.33	-

*Values followed by equal letters in the same column are considered the same according to the Tukey's HDS test for 95% confidence. ** Values in parenthesis are the variation coefficients.

For plywood panels of *Pinus caribaea* wood produced with urea-formaldehyde adhesive in weight 350 g/m^2, Iwakiri et al. (2001) reported values of shear strength of 1.28 MPa and 0.75 MPa for dry and cold water treatments, respectively. Shear strenght values for phenol formaldehyde adhesives reported in the same work were 2.34 MPa and 1.45 MPa for dry and boiling treatments, respectively. Such values are very close to those obtained it the present work.

Mendes et al (2013) produced panels with *Pinus taeda* slides and phenol-formaldehyde adhesive in grammage 420 g/m^2 distributed in double line and obtained resistance of the glue line in dry treatment of 2.9 MPa, and after boiling 1.63 MPa.

Three-point bending test

The results obtained in the three-point bending test are shown in Table 3.

The Modulus of Rupture (MOR) and the Modulus of Elasticity (MOE) in bending strength of the panels, with samples cut in parallel orientation to the fibers of the wood sheet upper layer are statistically equal for all tested adhesives. For thesamples cut in perpendicular orientation, the results for phenol formaldehyde and polyurethane are similar, but panels made with urea-formaldehyde adhesives had lower strength.

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Table 3 - Flexural strength (MOR), and elastic modulus (MOE) for the wood panels with different adhesives (180 g/m²)
Tabela 3 - Resistência à flexão (MOR) e módulo elástico (MOE) para painéis com diferentes adesivos (180 g/m²)

Orientation	Property	Adhesive used in the production of the panels		
		PU	PF	UF
Parallel	MOR (MPa)	63.32 (16.80%) a	61.19 (20.84%) a	61.98 (12.98%) a
	MOE (GPa)	6.61 (17.35%) a	7.19 (27.87%) a	7.15 (21.22%) a
Perpendicular	MOR (MPa)	34.78 (14.22%) a	32.18 (18.39%) a	25.22 (14.98%) b
	MOE (GPa)	2.13 (13.13%) ab	2.25 (19.75%) a	1.92 (13.18%) b

* Values followed by equal letters in the same line are considered the same according to the Tukey's HDS test for 95% confidence. ** Values in parenthesis are the variation coefficients.

Plywood panels with higher values of density tend to present higher modulus of rupture and modulus of elasticity (TRIANOSKI; IWAKIRI, 2018). In addition to the direct relationship between the density of the plywood and the density of the wood used in its composition, other factors such as the chemical and anatomical properties of the wood of different species may influence the mechanical properties of the panels.

In a study of *Eucalyptus grandis* with adhesive derived from castor oil-based polyurethane with adhesive grammage of 250 g / m², Dias and Lahr (2004) obtained in parallel orientation the fibers 87MPa of resistance to rupture and 11.11 GPa of modulus of elasticity, in analysis perpendicular to the fibers the resistance to rupture was of 61 MPa, and modulus of elasticity of 6.24 GPa.

Trianoski and Iwakiri (2018) studied plywood panels produced with sheets of *Pinus taeda* with 18 years and adhesive Urea Formaldehyde with adhesive grammage of 180 g / m² and obtained, in parallel orientation to the fibers, rupture strength of 47.89 MPa and modulus of elasticity of 3.99 GPa; when the fibers were perpendicularly analyzed, the tensile strength was 34.11 MPa and the modulus of elasticity was 1.87 Gpa.

Campos et al. (2009) analyzed plywood of *Pinus* sp. with polyurethane adhesive and adhesive grammage of 400 g / m² and double glue line, obtained in parallel orientation the fibers resistance to rupture of 44 MPa and modulus of elasticity of 13 GPa

The variations may occur depending on the kind of the wood, its grammage, type of adhesive, as well as the environment conditions during the production of the plywood panels.

Stage 2

Specific mass

The specific mass of the plywood panels with different glue grammage line are shown in Fig. 2

In figure 2 it can be seen that there was no statistical difference with respect to the apparent density of the panels at humidity of 12%, that is, the slides chosen to compose the panels have the same characteristic, eliminating the influence of the specific mass in the quality of the panels and the interference of this factor in the static flexural strength of the composites.

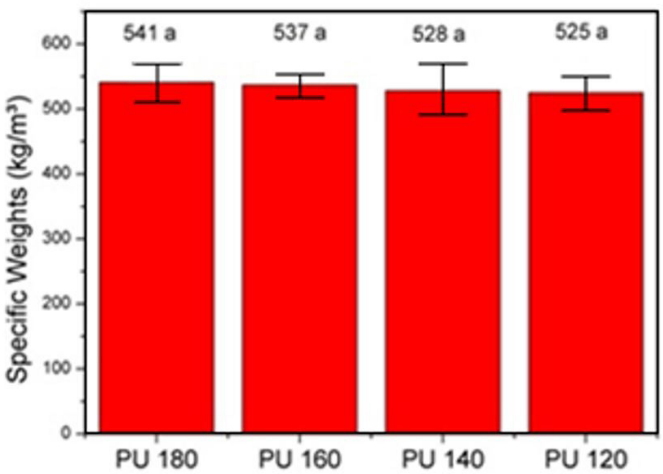

Figure 2 – Specific weight of the plywood panels with humidity 12% at Stage 2
Figura 2 – Massa específica dos painéis compensados com 12% de umidade da Etapa 2

A higher density of the wood may influence the formation of the adhesive bond between the blades, because of the greater difficulty of penetration of the adhesive, in addition to generating greater internal pressure of steam during the hot pressing. This contributes to the reduction of the polymerization rate of the resin (IWAKIRI et. al., 2007).

In an ABIMCI (2007) report on the analysis of panels of *Pinus sp*. and phenolic adhesive for trade, intended for external use in structural application, has a specific average mass of panels of 552 kg/m³, which is very close to that obtained in this work.

Shear strength

The shear strength of the plywood panels for different conditions are shown in Table 4.

Table 4 – Shear strength (RGL – Resistance of the Glue Line) and failure percentage for the wood panels with PU with glue spread line of 180, 160, 140 and 120 g/m²

Tabela 4 – Cisalhamento (RLC – Resistência da Linha de Cola) e porcentagem de falha na madeira para painéis com PU e gramatura de linha de cola de 180, 160, 140 e 120 g/m²

Treatments		PU 180	PU 160	PU 140	PU 120
Dry	RGL (MPa)	2.71 a (11.71%)	2.70 a (10.16%)	2.38 b (14.32%)	2.62 a (7.52%)
	Failure (%)	76.33	86.00	94.33	84.67
Cold water	RGL (MPa)	1.99 a (16.03%)	1.97 a (10.52%)	1.75 a (10.12%)	1.81a (14.64%)
	Falha (%)	10.67	8.67	23.33	11.79
Cycle	RGL (MPa)	1.11 a (21.28%)	1.44 a (18.04%)	1.42 ab (10.99%)	1.09 b (52.36%)
	Falha (%)	2.50	7.33	26.0	10,71
Boiling	RGL (MPa)	1.09 a (51.67%)	1.44 a (11.62%)	1.32 a (11.68%)	1.14 a (45.73%)
	Falha (%)	5.38	6.53	11.33	10.67

* Values followed by equal letters in the same line are considered the same according to the Tukey's HDS test for 95% confidence. ** Values in parenthesis are the variation coefficients.

Statistical differences were observed in the dry and boiling cycle treatments. All weights presented mean values of shear stresses above the minimum value of 1.0 MPa, in accordance with EN 314-2: 1996.

The plywood panels of the *Eucalyptus grandis* with adhesive castor oil-based polyurethane with adhesive grammage of 250 g/m² studied by Dias and Lahr (2004) show presented a shear stress of 3.1 MPa and 78% of failure in the wood in dry condition, 2.7 MPa and 32% of failure in the wood when wet and 1.9 MPa and 16% of failure in the wood after boiling.

Bianche et al (2016) analyzed panels produced with *Pinus* sheets and adhesive polyurethane derived from castor oil with adhesive grammage of 150 and 200 g / m². Considering glue line 150 g / m², in dry condition, the shear strength was 4.67 MPa and the wood failure was 36%; already in line of glue grammage of 200 g / m² the resistance to the shear was of 4.24 MPa and the failure in the wood of 48%. When submitted to the cold water condition they obtained for panels with 150 g / m² of adhesive shear strength of 2.76 MPa and 10.9% of failure in the wood; in panels with a glue line grammage of 200 g / m² the shear strength was 3.12 MPa and the wood failure was 20.3%. In panels whose glue line was thicker, 250g / m², the authors noticed a reduction in shear strength and failure of the wood in both conditions (BIANCHE et al, 2016)

The results obtained by Bianche et al (2016) and Dias and Lahr (2004) suggest that the higher weight of glue line, to an optimum point, allied to the characteristics of the wood used influence the shear strength, as well as the percentage of wood failure.

The average values obtained for panels glued with phenolic adhesive presented 1.25 MPa of resistance to the glue line for the treatment cycle; and 1.07 MPa for boiling treatment (ABIMCI, 2007).

Based on the requirements of the European standard,produced with the polyurethane adhesive derived from castor oil in the grammage of 180 g/m², 160 g/m², 140 g/m², 120 g/m² under the conditions used for this work, may be indicated for internal and external use.

Three-point bending test

The mean static bending results for weights of 180 g/m², 160 g/m², 140 g/m² and 120 g/m² are shown in Table 5.

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Table 5 - Flexural strength (MOR) and elastic modulus (MOE) for the wood panels with polyurethane adhesive and glue spread line of 180, 160, 140 and 120 g/m²

Tabela 5 – Resistência à flexão (MOR) e módulo elástico (MOE) para painéis com adesivo poliuretano e gramatura de linha de cola de 180, 160, 140 e 120 g/m²

Orientation	Property	Glue Line Weight for PU Adhesive			
		PU 180	PU 160	PU 140	PU 120
Parallel	MOR (MPa)	63.32 a (16.80%)	65.08 a (18.38%)	63.81 a (11.49%)	56.88 a (23.07%)
	MOE (GPa)	6.61 ab (17.35%)	7.25 a (20.88%)	6.54 ab (13,31)	5.53 b (21.08%)
Perpendicular	MOR (MPa)	34.78 a (14.22%)	27.29 b (7.76%)	27.00 b (15.19%)	31.07 ab (9.59%)
	MOE (GPa)	2.13 ab (13.12%)	1.98 a (13.03%)	2.09 a (24.04%)	2.12 a (11.87%)

* Values followed by equal letters in the same line are considered the same according to the Tukey's HDS test for 95% confidence. ** Values in parenthesis are the variation coefficients.

Through the statistical analysis it can be seen that there were no variations of resistance parallel to the fibers between the different weights. This means that a plywood panel produced with the 180 g/m² polyurethane adhesive may have similar strength to the same panel produced at 120 g/m². From an economic point of view, this finding is of fundamental importance, since reducing the amount of adhesive in the panel making did not reduce the flexural strength values.

Campos et. al. (2009) obtained, for plywood panels of *Pinus* sp. with bicomponent polyurethane based on castor oil using a weight of 400 g/m³, pressing at 1.5 MPa at 60°C for 15 minutes, MOR ranging from 39 to 56 MPa and MOE of 10 to 15.3 GPa for static bending.

According to ABIMCI (2007) report, in relation to static bending, mean values for parallel and perpendicular to the grain rupture modules (MOR) were 45.36 MPa and 32.05 MPa, respectively. The mean values for the parallel and perpendicular modulus of elasticity (MOE) were 5.1 GPa and 2.6 GPa, respectively.

It is possible to state that the polyurethane adhesive derived from castor oil can replace the phenol-formaldehyde based adhesive, which is a structural adhesive indicated for external use, when used in compensated panels.

The variations occurred in the analyzed results may be related to the scattering methodology employed and the quality of the wood used. In addition, for the weights of 180 g/m² and 160 g/m² the adhesive was run through the edges of the panel, which means that not all the adhesive applied was actually used in the panel gluing process, influencing in the results obtained in this work.

CONCLUSIONS

- The specific weight of the plywood panels was not affected by the type of adhesive employed in its production;
- The shear strength of plywood panels produced with adhesive made with castor oil-based polyurethane is higher than those produced with formaldehyde adhesives;
- The variation of the weight of the glue line did not affect the static flexural strength of the panels;
- When compared to plywood panels made with formaldehyde adhesives, there was no reduction in flexural strength of the plywood panels produced with the adhesive made with castor oil-based polyurethane;
- The production of plywood panels with *Pinus caribaea bahamensis* and adhesive made of castor oil-based polyurethane is technically viable;
- The most viable glue line weight for the production of these panels with polyurethane adhesive derived from castor oil is 120 g/m².

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