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Alternative Castor Oil-Based Polyurethane Adhesive Used in the Production of Plywood

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Plywood is normally produced with urea-formaldehyde and/or phenol-formaldehyde adhesives. However, the former is considerably toxic and environmentally damaging, while the latter is expensive, thus motivating the search for alternative raw materials in plywood production. The castor oil-based polyurethane adhesive developed at the São Carlos Institute of Chemistry, University of São Paulo, is an environmentally friendly vegetal oil-based polymer that is harmless to humans. The wood species *Eucalyptus grandis* offers favorable properties for plywood the manufacture. The study reported on here involved the use of castor oil-based polyurethane adhesive to produce plywood with *Eucalyptus grandis* layers. The plywood's performance was evaluated based on the results of physical and mechanical tests recommended by the Brazilian code, ABNT. Tests results showed higher values than those reported in the literature and recommended by the ABNT, indicating that the castor oil-based polyurethane adhesive is a promising glue for the manufacture of plywood.

Keywords: *plywood, polyurethane adhesive based on castor oil, properties*

1. Introduction

The advantage of wood-based products is that the wood's properties can be improved through the application of science and technology. These products include wood panels, or plywood, whose consumption around the world and in Brazil has grown steadily.

These panels are produced with adhesives, and the ones available in Brazil, i.e., urea-formaldehyde and/or phenol-formaldehyde adhesives, involve negative factors such as high energy consumption due to the high temperatures required for curing (130 to 160 °C), the high cost of phenol, which is petroleum-based, and their toxicity for humans and the environment.

An alternative adhesive deriving from a natural and renewable source, castor oil-based polyurethane adhesive is classified as impermeable and non-aggressive to the environment and to humans, besides representing a Brazilian technology. This adhesive is cured at ambient temperature, but curing can be accelerated using temperatures of 60 to 90 °C, and it is believed that its large scale availability on the market will bring its price down to a very reasonable level.

The wood panel industry represents an important segment of the country's economy, not only because it generates revenue and jobs but also due to its increasing dynamism, particularly in the furniture and civil construction sectors. However, for this dynamism to continue, technological investments are required to expand and improve the production of panels.

Some of the practical factors the justify the preferential use of wood-based panels are the reliability of the material, stability of the pieces, possibility of producing pieces with dimensions not found in solid wood, use in modern and complex applications with low cost-efficiency ratio, and utilization of most of the log[1].

Wood-based panels are classified as wood-based layers, fiber and particle panels. The layered-based panel include plywood, which consists of thin layers of wood superposed orthogonally and glued together, their fibers laid in alternate directions.

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Today, the glues traditionally available on the market represent the most expensive components of these panels, and their toxicity poses health risks. These two factors *per se* justify the search for alternative adhesives of comparable quality and performance, based on raw materials deriving from natural and renewable sources. In this context, castor oil-based polyurethane adhesive possesses satisfactory properties[2]. This adhesive was developed at the Institute of Chemistry of São Carlos, of the University of São Paulo.

The purpose of this study, therefore, was to evaluate the performance of plywood produced with the wood species *Eucalyptus grandis* based on its physical and mechanical properties and the performance of the castor oil-based polyurethane adhesive, comparing the results with the properties of plywood reported in the literature.

2. Plywood

Plywood, as explained earlier, is produced by superposing and gluing together several layers of wood with their fibers crossed perpendicularly. This procedure, called cross lamination (Fig. 1), confers good rigidity, mechanical strength and dimensional stability on the product. Plywood can be made from either softwoods or hardwoods[3,4].

The layers in plywood are aligned according to a predetermined arrangement so as to obtain equivalence of its properties of stiffness and strength in the main directions of the board. Crossed lamination and gluing of plywood further capitalize on the wood's most desirable physical and mechanical properties[4].

Plywood has well-established applications in several market segments, such as furniture, boatbuilding, special weather-resistant packaging, musical instruments and sports equipment, and in civil construction. In the latter industry, it is applied in external walls, roof closures, flooring, wall panels, internal dividing walls, fronts, composite beams, truss connections, floor panels, concrete molds, arch-shaped roofs, etc.

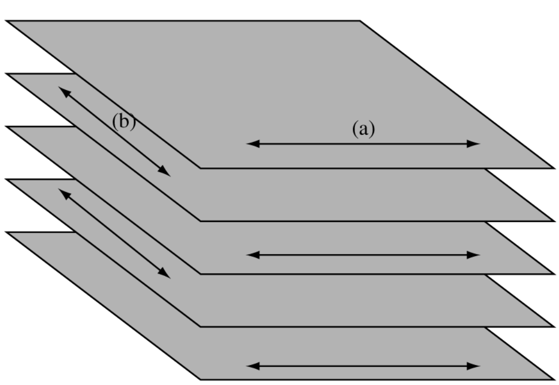

Figure 1. Position of layers in plywood. a) Direction perpendicular to the grain of the external layers; b) Direction parallel to the grain of the external layers. Source: Keinert Jr.[12].

2.1. Plywood specifications

The Brazilian code - ABNT - contains norms specifying the dimensions, tolerances and conditions to be followed for the classification of plywood, as listed below.

ABNT - NBR 9531 (1986)[5] standard: "Plywood - classification" qualifies boards according to the place they are used, based on three basic types: IR (interiors), IM (intermediates) and EX (exteriors).

IR plywood is produced with non-water resistant adhesive, and is applied in places away from water or high relative moisture content. The IM type plywood is destined for internal use, but in environments with high moisture content which may be subject to water splashes. EX plywood is destined for external use or in closed environments, where it is subjected to repeated wet and dry cycles or to the action of water.

ABNT - NBR 9532, (1986)[6] states: "Plywood - specifications" establishes the geometric characteristics, such as assembly, dimensions, shape, thickness and tolerances, as well as other conditions that must be adhered to for each type of board. According to this standard, plywood is divided into the following specifications:

- General use plywood – GER: plywood boards classified generically as IR.
- Mold for reinforced concrete – FOR: plywood boards classified generically as EX. This type of plywood should present a modulus of elasticity of at least 4000 N/mm^2.
- Decorative - DEC: plywood boards classified generically as IM.
- Industrial - IND: plywood boards classified generically as EX.
- Marine applications - NAV: plywood boards classified generically as EX. This type of plywood should present a modulus of elasticity of at least 6000 N/mm^2.
- Slatted - SAR: plywood boards classified generically as IR, whose internal portion consists of wood slats with a maximum width of 40 mm.

2.2. Plywood production

The description given below is based on Zugman[8], who describe the production stages of plywood.

Before they are cut in a laminator, the logs are debarked and placed in tanks containing steam or hot water to soften the wood and increase its plasticity, making it easier to obtain smoother and less breakable layers.

The wood layers are then produced by cutting, sawing or cutting on a rotating lathe. The next stage consists of drying the layers to reduce their moisture to a preestablished content and produce a flat, flexible material. Drying is done very quickly (a few min) and the final moisture content is about 4 to 6%. The dry layers are stored in piles according to their width and class. They should be visually classified by type and size of defects, as well as by the number and characteristics of the grain of the various layers.

In small operations, the adhesive is applied with a brush, but large scale operations use roll spreaders. Another method often used is the curtain method, whereby a machine with a reservoir, having a pair of parallel steel blades with a slit between them mounted underneath the reservoir, spreads a thin curtain, or film, of adhesive over the surface of the layers.

Two important factors are directly correlated with the strength, density and quality of the end product, i.e., the amount of adhesive applied per area and its uniform distribution over the layers[7,8].

Immediately after the adhesive is applied on the layers, they should be assembled to form the plywood board, which is then pressed under a load. It is advisable to exert a slight initial pressure before applying the total load. If a heated press is used, its loading should be performed as quickly as possible to prevent the adhesive from drying out before the board is properly compressed[7,8]. Compression time depends basically on the temperature of the press and the type of adhesive used. For compression temperatures of 100 to 160 °C, compression should be limited to a few min. The gluing pressure by hot pressure may vary from 1.2 to 2.0 MPa.

2.3. Potential of the genus Eucalyptus for the manufacture of plywood

Approximately 70% of the wood used to produce plywood in Brazil comes from native forests and 30% from reforested *Pinus* spp. However, the stock of *Pinus* spp forests has shrunk and the enormous variability of the characteristics of native species makes their use as raw material difficult[10].

The search for new sources of raw materials for the production of plywood and the favorable characteristics of *Eucalyptus* spp wood are strong arguments in favor of research to identify the potential of new species of the genus, its hybrids and clones, the quality of the products derived from them, and the determination of alternative sources of wood.

One the most commonly used species of eucalyptus used in southern Brazil is the *E. grandis* grown in the state of São Paulo[10]. The genus *Eucalyptus* can be considered one of the most promising for producing alternative products to replace native species. Its wood can be used for various purposes, among them the production of panels.

Jankowsky[11] evaluated *Eucalyptus grandis* wood for the production of layers and plywood panels, concluding that this wood is technologically viable for use in the manufacture of plywood.

3. Adhesives

Adhesives are one of the main components in the production of wood-based panels.

Plywood is produced according to two main specifications: a) for indoor use, glued with urea-formaldehyde resin, which is employed chiefly in the furniture industry, and b) for outdoor use, glued with phenol-formaldehyde resin, employed in civil construction[9-12].

Urea-formaldehyde adhesives have a serious drawback, which is the emanation of formaldehyde fumes and its use is limited in countries with strict environmental control. Phenol-formaldehyde adhesive is produced from synthetic phenolic resins. One of its negative factors is the cost of phenol, whose raw material is petroleum. This adhesive required high curing temperatures that vary from 130 to 160 °C [13,14].

3.1. Alternative adhesive: castor oil-based polyurethane

In the early 1980's, the Department of Chemistry and Molecular Physics of São Carlos made an important national contribution through its studies on polyurethane resin-based adhesives. The researchers of the Analytical Chemistry and Polymer Technology Group developed a castor oil-based polyurethane resin that offers several advantages, such as handling at ambient temperature, strong resistance to water and UV rays, strong mechanical strength and the fact that it derives from a natural and renewable source easily found in most of the country[15].

The castor-oil plant *(Rícinos communis)* is a plant of the Euphorbiaceae family from which castor oil is extracted. This plant is known internationally as Castor-Oil Plant and in Brazil as Caturra[16].

Polyols and prepolymers with different characteristics can be synthesized from this natural and renewable source, which, when mixed, result in a polyurethane. Its competitiveness in relation to other polymers is basically tied to three factors: it derives from a natural and renewable raw material, its mechanical properties are superior to those of petroleum-derived polymers, and the cost of the diisocyanates in Brazil is reasonable[15].

Claro Neto[15] characterized the physical and mechanical properties of a castor oil-based polyurethane polymer. The study of the chemical properties aimed to characterize the functional organic groups present in the polyol/prepolymer components that react during preparation of the polyurethane. The castor oil-based polyurethane was developed for use in medical applications as a bone implant material.

To expand the application of this product, Jesus[2] and Azambuja[17] studied of the behavior of castor oil-based polyurethane adhesive for use in glued laminated timber (GLT) with reforested species of the genera *Eucalyptus* and *Pinus*. The adhesive's efficiency was evaluated based on mechanical tests of shearing strength parallel to the grain, tensile strength perpendicular and tensile strength parallel to the grain. The results reported by these authors demonstrated that castor oil-based polyurethane adhesive is a good alternative for the technological use of GLT.

4. Materials and Methods

4.1. Production of plywood boards

An experimental model of plywood was fabricated in the Wood and Timber Structures Laboratory of the Department of Structural Engineering, School of Engineering of São Carlos, University of São Paulo.

The adhesive, made of the Institute of Chemistry of São Carlos of the University of São Carlos, which was applied with a brush, was castor oil-based polyurethane resin of the bicomponent type, B1640 polyol and A249 prepolymer, in a 2:1 ratio. The adhesive was applied in simple lines, with a layer of glue applied to the layers on only one of the surfaces. The gluing parameters are given in Table 1.

The plywood panels were manufactured with seven layers of the species Eucalytus grandis having a nominal thickness of 0.2 cm. Four panels were produced with a nominal thickness and area of 50 × 50 cm, from which the test specimens (14 for each test) were taken for characterization of their physical and mechanical properties.

The wood layers were supplied by the Wood based Panels and Peeling Laboratory, Department of Forest science, of the Superior School of Agriculture “Luiz de Queiroz”, of the university of São Paulo. These layers were classified according to the Brazilian Code NBR 9531 (1986)[5], in the classes: N, B, C and D, from the best to the worst quality.

Plywood panels were manufactured with class C layers.

4.2. Experiments

The tests involved in the characterization of the plywood were: moisture content, specific gravity, modulus of elasticity (MOE) and modulus of rupture (MOR) obtained from the static bending test and glue-shear strength.

To identify the moisture content, NBR 9584 (1986)[18], each test specimen was weighed and then oven dried at 103 ± 2 °C to a constant mass. The moisture content was obtained through Eq. 1.

$$TU = \frac{Mu \quad Ms}{Ms} \cdot 100 \qquad (1)$$

where: TU = moisture content in %; M_u and M_s = the test specimen’s moist and dry mass in g.

Equation 2 was used to obtain the specific gravity, NBR 9485 (1986)[19]. The nominal dimensions of these specimens were (100 × 50 × 14) mm.

$$\rho = \frac{m}{v} \qquad (2)$$

where: Mea = specific gravity in g/cm^3; M = the specimen’s mass in g; C, l, e = length, width and thickness of the specimen in cm.

For the static bending test, according to the NBR 9533 (1986)[20], the specimen is supported at both ends and a load is applied at mid-span (Fig. 2), using a chopper continuously at a constant velocity. The nominal dimensions of these specimens were: length of 400 mm; width of 70 mm and thickness of 14 mm.

The loading velocity was calculated using Eq. 3.

$$V = \frac{K.L^2}{6.e} \qquad (3)$$

where: K = 0.00005, fiber deformation rate; and V = Loading velocity in mm/s.

The loads and the corresponding vertical displacements were recorded (minimum of 12 readings) in order to determine the modulus of elasticity (MOE) based on Eq. 4, and the modulus of rupture (MOR) based on Eq. 5.

$$E_b = \frac{L.^3.(F_2 \quad F_1)}{4.l.e^3 (S_2 \quad S_1)} \qquad (4)$$

$$T_r = \frac{3.F_{máx.}.L}{2.l.e^2} \qquad (5)$$

where: E_b = modulus of elasticity, in N/mm^2; L= distance between the centers of the supports, in mm; l.e = width and thickness of the specimen, in mm; $F_2 – F_1$ = load increment in the straight portion of the load-strain curve, in N; S_2 - S_1 = vertical displacement increment at mid-span, corresponding to $F_2 – F_1$, in mm; T_r = modulus of rupture, in N/mm^2 and F_{max} = rupture load, in N.

The procedure for the glue-shear strength test is described in the NBR 9534 (1986)[21] code. This standard establishes that at least five test specimens should be taken from each plywood board.

This test served as the basis to evaluate the quality of the glue line and the plywood was classified according to the place where it would be utilized. The plywood boards

Table 1. Gluing parameters for plywood manufactured with castor-oil polyurethane adhesive.

Gluing parameters	
Moisture content of sheets	4 a 6%
Viscosity of adhesive	6 min
Gluing pressure	1.2 MPa
Time of pressure	10 min
Time curing total	4 days
Temperature of pressure	60 °C
Amount of adhesive used per area of sheet (weight)	250 g/m^2

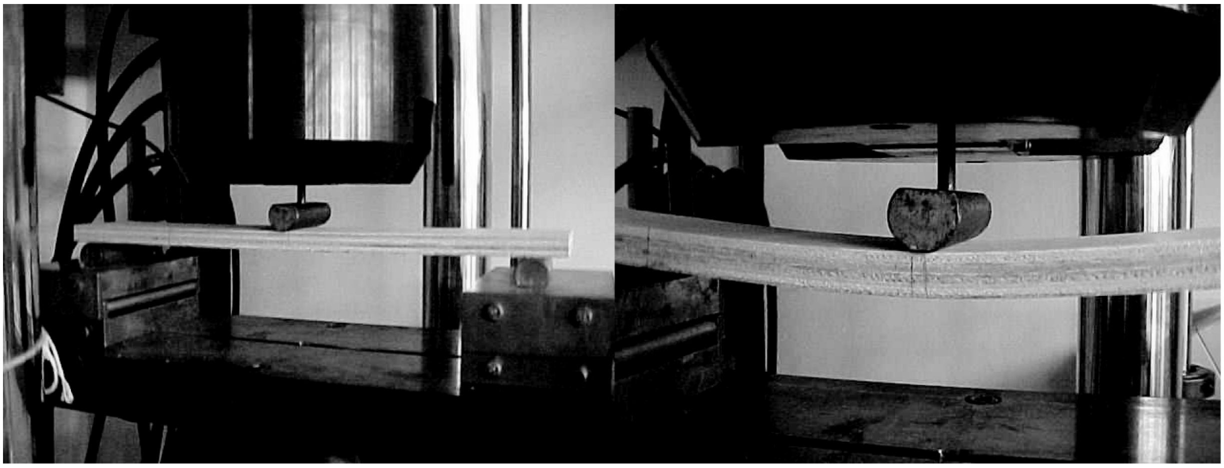

Figure 2. Mechanical test for determination of static bending properties.

destined for indoor use were evaluated through a dry test, intermediate plywood was evaluated in a damp test, and the exterior plywood was evaluated in a post-boiling test, based on their tensile strength values. These tests also yielded the wood's failure percentage, evaluated subjectively, whose value complements the results of the shear tests.

The dry test involves applying the load until complete rupture of the test specimen is achieved, with moisture content in equilibrium with the environment (approximately 12%). In the humid test, the specimen is completely submerged in water (15 ± 5) °C for 24 h and tested without prior drying. For the boiling state test, the specimen is submerged in boiling water for 4 h and dried for 20 h at a temperature of 62 ± 3 °C. The test specimen is then again submerged in boiling water for 4 h, cooled on cold water and tested while still damp. The tensile strength is calculated using Eq. 6.

$$t_r = \frac{F_{máx}}{a.b} \quad (6)$$

where: t_r = tensile strength, in N/mm^2; $F_{máx}$ = rupture load, in N; a = distance between grooves, in; b = width of the specimen, in mm.

5. Results and Discussion

Tables 2 and 3 list the results obtained for moisture content, specific gravity, modulus of rupture and modulus of elasticity for plywood manufactured with castor oil-based polyurethane adhesive, considering the parallel and perpendicular direction of the grain of the external layers.

Rocco Lahr[22] and Sales[23] demonstrated that the values of the variation coefficients for the properties of wood are in the order of 18%, for wood with a moisture content of 12%. Tables 2 and 3 indicate that lower variation coefficients were obtained than those listed for wood. The Brazilian standards do not stipulate values for the variation coefficients of plywood. We have therefore adopted the standards listed for solid wood as a reference.

Table 2. Results in the tests for direction parallel to the grain of the external layers.

Specimens	Specific of gravity (g/cm^3)	Modulus of rupture (N/mm^2)	Modulus of elasticity (N/mm^2)
C01	0.80	76	10080
C02	0.81	94	11577
C03	0.84	92	11631
C04	0.82	80	8565
C05	0.79	88	9835
C06	0.83	98	12106
C07	0.78	77	10758
C08	0.78	97	11928
C09	0.81	83	11700
C10	0.80	81	11318
C11	0.80	83	11710
C12	0.82	83	11418
C13	0.81	101	11965
C14	0.80	87	10877
Average	0.81	87	11105
Variation coefficient (%)	2.2	9.3	9.0
Moisture content (%)		11.4	

Table 4 lists the results of the wood's tensile strength and failure tests obtained in the glue-shear strength test carried out under dry, wet and boiling conditions.

The Brazilian NBR 9534 (1986)[24] code does not specify a reference value for tensile strength in the shear test and requires a minimum wood failure percentage of 60, 60 and 80%, respectively, to classify plywood for indoor, intermediate and outdoor use. This standard is currently under review, and there is a tendency for adopting the requirements of the European Standard - EN 314-2 (1993)[24] presented in Table 5.

The European standard correlates the average tensile strength value with the average value of the failure percentage of wood. The requirements in terms of wood failure decrease as the tensile strength increases, and for values equal to or higher than 1.0 MPa there is no longer any failure percentage requirement.

The average tensile strength values listed in Table 4 exceeded 1.0 MPa under all the conditions tested here (dry, wet and boiling). Based on the European standard's requirements, it can be stated that plywood manufactured with castor oil-based polyurethane adhesive is suitable for indoor, intermediate and outdoor applications.

The low variation coefficient values indicate homogeneity in the manufacturing process and good distribution of the glue over all the layers.

5.1. Comparison of the mechanical properties of plywood found in the literature with those of plywood manufactured with castor oil-based polyurethane adhesive

Table 6 lists the values of some physical and mechanical properties reported in the literature for plywood made of Eucalyptus grandis and phenol-formaldehyde glue. This plywood was produced with seven layers and pressed for 13 min at 130 °C.

A comparison of the values listed in Tables 2 and 3 against those of Table 6 indicates that the plywood produced with the alternative castor oil-based polyurethane adhesive displays properties superior to those of commercial plywood. These values were higher in the direction perpendicular to the grain of the outside layers.

The Brazilian code NBR 9532 (1986)[6] determines minimum MOE values for plywood for outdoor and structural applications. A comparison of those values and those obtained for the plywood produced with castor oil-based poly

The MOE values are higher than those required by the

Table 3. Results in the tests for direction perpendicular to the grain of the external layers.

Specimens	Specific of gravity (g/cm³)	Modulus of rupture (N/mm²)	Modulus of elasticity (N/mm²)
C15	0.80	61	5700
C16	0.76	61	5887
C17	0.77	67	6338
C18	0.79	60	5783
C19	0.79	53	6159
C20	0.80	60	5830
C21	0.82	73	7407
C22	0.83	55	5384
C23	0.82	64	7199
C24	0.80	62	5888
C25	0.81	57	6319
C26	0.79	54	6110
C27	0.80	64	6427
C28	0.81	63	6975
Average	0.80	61	6243
Variation coefficient (%)	2.4	8.8	9.5
Moisture content (%)		11.4	

Table 4. Results of the plywood's glue-shear strength.

Condition of test	Average values of Glue-shear strength		
	Tensile strength (t_r)		Wood's failure (%)
	t_r N/mm² (Mpa)	Variation coefficient (%)	
Dry (Moisture content: 11,4%)	3.1	13	78
Wet	2.7	12	32
Boiling	1.9	6	16
Number of specimens		14	

Table 5. Requirements of the European Standard for the plywood's glue-shear strength.

Glue-shear strength	
Tensile strength - t_r (MPa)	Wood's failure (%)
0.2 ≤ TR < 0.4	≥ 80
0.4 ≤ TR < 0.6	≥ 60
0.6 ≤ TR < 1.0	≥ 40
1.0 ≤ TR	no exigency

Source: EN 314-2 (1993)[24].

Table 6. Mean values of physical and mechanical properties for plywood made of Eucalyptus grandis and phenol-formaldehyde glue.

Static bending		
Direction	Parallel to the grain	Perpendicular to the grain
Modulus of rupture (N/mm²)	10830	4902
Modulus of elasticity (N/mm²)	80	49
Moisture content (%)	10.5	
Specific gravity (g/cm³)	0.72	

Table 7. Comparison between values of the alternative plywood properties with specified by ABNT.

Plywood specification	Módulus of elasticity (N/mm²)
ABNT NBR 9532 (1986)[6]	4000 - Mold for reinforced concrete 6000 - marine applications plywood
Manufactured with castor-oil-based polyurethane adhesive	11105 – parallel direction 6243 – perpendicular direction

NBR 9532 (1986)[6] code for plywood with satisfactory characteristics for outdoor application in molds for reinforced concrete and for marine applications. The values stipulated by the code correspond to the direction parallel to the fibers of the outside layers. According to Olin [5], the MOR and MOE values in static flexure in the parallel direction are always superior to those obtained in the direction along the fibers of the outside layers.

6. Conclusions

The plywood panels produced with castor oil-based polyurethane adhesive showed highly satisfactory results. The values of the properties of strength and stiffness obtained in the static bending test were higher than those reported in the literature for plywood made of the same species (Eucalytus grandis), using traditional glue (phenol-formaldehyde).

The MOEs exceeded those required by the NBR 9532[6] standard for plywood, showing characteristics suitable for outdoor applications. It should be noted that the MOE of plywood manufactured with castor oil-based polyurethane glue in the parallel direction to the grain exceeded the standard value listed in the code for the direction parallel to the external layers.

The results obtained in the glue-shear strength tests indicate the suitability of using the plywood studied here in indoor, intermediate and outdoor applications. The tests showed little variability, indicating the good quality of the glue line.

Low variation coefficients were obtained for all the physical and mechanical properties of the panels, which is a good indicator of the product's homogeneity and of the good performance in the production and tests of the plywood.

It is important to note that, in the manufacture of the plywood with castor oil-based polyurethane adhesive, the temperature of compression was 60 °C. This represents a significant reduction in energy consumption, considering that the curing of phenol-formaldehyde resin is processed at temperatures of 130 to 160 °C. Thus, the castor oil-based polyurethane adhesive is a promising alternative for use in plywood production.

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