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Determination of the Adhesive Content of Medium Density Particleboards Produced with Bio-based Polymer

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ABSTRACT
This work contributes to the use of alternative adhesives in the wood-based industry, where pine wood is commonly used. The investigation identifies the influence and the optimal content (8, 12 and 15wt%) of a bio-based polyurethane adhesive in the production of medium density particleboards (MDP). A compaction pressure of 4 MPa at 100°C for 10 minutes is considered in the manufacture of panels based on pine wood residues and bio-based resin. The bulk density, flexural modulus (MOE) and strength (MOR) properties under static three-point bending are obtained according to the Brazilian standard NBR 14810. The results are compared with NBR 14810 and other standards to verify its performance based on the minimum requirements. Bulk density is not significantly affected by the investigated adhesive levels. MOE and MOR reach average values equivalent to 12wt% and 15wt% of the adhesive, and both meet the minimum requirements established in international normative documents. The adhesive level range responsible for maximizing ρ, MOE and MOR is between 12.42wt% and 15.79wt%.

Keywords:
Particleboard
Pinus wood
Bio-based adhesive
Tukey test
Regression model

1. Introduction
The planting of forests was strongly encouraged in the 1970s, allowing the exploitation and expansion of the reforestation wood stock [¹]. Eucalyptus and Pinus were the genera that most adapted, which motivated the development of several investigations to improve their planting. In this context, high productivity consolidated the commercial use of wood from pine and eucalyptus, although other tree species are planted. In 2014, the area of planted forests in Brazil was over nine million hectares, of which 74.2% were eucalyptus and 21.9% of pine [²]. Therefore, the best use of these two genera as raw material for products derived from wood is fundamental.

Particleboards are extremely interesting from a sustainable...
point of view, since there is a possibility to produce them with waste from processed wood. Data collected by the Annual Industrial Product Survey, carried out by the Brazilian Institute of Geography and Statistics \[2\] reveal a quantity of more than 1.9 million tonnes of wood residues produced annually in the country, an expressive amount that can be reduced with the use in the production of engineered wood products, such as particleboard.

In Brazil, wood is used mainly by the civil construction and furniture sectors, being the main responsible for the technological evolution of particulate sheets \[3\]. The panel production process involves, in addition to the wood residues, the application of a synthetic or natural resin to guarantee the fixation between the particles, being subsequently hot-pressed for a certain time \[4\].

Polymers are of great importance in the production of agglomerated sheets, as they provide different physical-mechanical properties according to their type and concentration \[5,6\]. In the wood products industry, the most used resin is urea-formaldehyde. This is due to its low cost, high cure speed, little colour development and flame resistance \[7\]. However, the emission of formaldehyde, a toxic gas, in the pressing stage \[9\], is one of the factors that encourage the search for new alternatives that are less aggressive to health and the environment.

Currently, there are other adhesives free of harmful and biodegradable gas emissions, such as castor-based polyurethane resin. This biopolymer has as advantages the possibility of manipulation at room temperature, resistance to the action of water and ultraviolet rays and great mechanical resistance, in addition to being produced with a renewable natural resource \[9\]. This resin has been modified by the combination of vegetable oils (castor and others), in order to make its price more competitive.

Due to the high Pinus wood production in Brazil, which implies a considerable volume of sawmills residues, this work describes the manufacture and characterisation, in static bending test, of medium density particleboards made with bio-based resin in different contents (8, 12, 15wt%). A quadratic regression model is applied to determine the ideal adhesive content for each investigated property, as well as the influence of adhesive levels using the Tukey test analysis. This work contributes to a better understanding of the physical and mechanical properties of the agglomerated sheets manufactured with this bio-based adhesive.

2. Material and Methods

Experiments are developed at the Wood and Wood Structures Laboratory (LaMEM), belonging to the Structural Engineering Department (SET), of the São Carlos School of Engineering (EESC), of the University of São Paulo (USP). Figure 1a shows Pinus wood particles ranging in size from 0.8 and 2.8 mm and a moisture content close to 10% \[10\]. The dry mass of a panel is assumed to be 640g with dimensions of $280 \times 280 \times 10$ mm$^3$ and density of 0.7 g/cm$^3$ \[11\]. The mixed polyurethane resin of vegetal oil, composed of a polyol (1.0 g/cm$^3$) and an isocyanate pre-polymer (1.24 g/cm$^3$), is supplied by Kehl® Company (São Carlos-Brazil). The resin components are mixed in a 1:1 ratio \[12\]. A mixer is used to combine the bio-resin and Pinus particles considering the levels of 8, 12 and 15wt% (Figure 1b). A pre-pressing at 0.01 MPa at room temperature, followed by a hot-pressing at 4.0 MPa and 100°C for 10 minutes is used to manufacture the samples according to Varanda et al. \[10\], as shown in Figure 1c.

Figure 1. Pinus sp. particles (a), material mixing (b) and compaction (c)

The Tukey test, at a significance level of 5%, is used to assess the influence of the adhesive content factor [Ad] at 8, 12 and 15wt% levels on bulk density ($\rho$), modulus of elasticity (MOE) and modulus of rupture (MOR) in static bending. Ten panels are produced for each level of adhesive content. From the Tukey test, A denotes the group associated with the highest mean value, B the second highest mean value, and so on. Equal letters imply treatments with statistically equivalent means. The evaluated properties ($\rho$, MOE and MOR) are determined according to the protocols of the Brazilian standard NBR 14810 \[13\]. The feasibility of using the investigated panels as a commercial product is evaluated in accordance with the requirements described in NBR 14810 \[13\] and in international normative documents \[14-16\].

Second-degree polynomials (Equation 1) are used as alternative way to estimate the adhesive content responsible for the extreme values of the evaluated properties.

$$Y = a_0 + a_1\text{Ad} + a_2\text{Ad}^2$$ \hspace{1cm} (1)

In Equation 1, Y denotes the dependent response-variable ($\rho$, MOE, MOR); Ad is an independent factor (adhesive content) and $a_i$ consists of coefficients obtained...
by polynomial interpolation.

3. Results and Discussion

Figure 2 shows the mean values, coefficients of variation (CV - wt%), mean confidence intervals (reliability level 95%) and Tukey test (5% significance) of the properties evaluated for each adhesive content. The Tukey test shows that the variation in adhesive contents does not significantly affect the mean bulk density values (Figure 2a). Regarding MOE (Figure 2b), the adhesive levels of 12 and 15wt% are considered statistically equivalent showing group of letter A (Figure 2b). Similarly, the lowest flexural strength (MOR) is achieved using 8wt% adhesive, and the highest values are achieved when 12 or 15wt% adhesive levels are considered, since their means are statistically equivalent (Group A, Figure 2c).

Table 1 presents the requirements for different standards [13-16] and castor oil-based adhesive literature [17-18] with respect to the bulk density, modulus of elasticity and flexural strength of particleboards.

| Type      | Document | ρ (g/cm³) | MOE (MPa) | MOR (MPa) |
|-----------|----------|-----------|-----------|-----------|
| Standards | NBR 14810 [13] | ---- | 3350 | 22 |
|           | ANSI A208.1 [14] | > 0.8 | 2400 | 16.5 |
|           | CS 236-66 [15] | > 0.8 | 2450 | 16.8 |
|           | EN 312 [16] | ---- | 2300 | 16 |
| Literature | Bertolini et al. [17] | 0.73 | 2911 | 29 |
|           | Paes et al. [18] | 0.93 | 1636.38 | 14.42 |

The results of bulk density of the panels manufactured with a bio-based are all greater than 0.8 g/cm³, which shows that they are in accordance with the requirements of international standards [14-15] and also close to the results found by Bertolini et al. [17].

In relation to MOE, only panels manufactured with 12wt% (2543 MPa) and 15wt% (2675 MPa) of adhesive meet the minimum requirements of international standards [14-16], but not those established by the Brazilian standard [13]. It is noteworthy that the MOE results for all adhesive levels are higher than the mean value found by Paes et al. [18], which involves Pinus wood and castor oil-based adhesive. In order to achieve better results of MOE, it is necessary to study new manufacturing parameters, such as pressure, pressing time and temperature, and their interaction effects, which will be the scope of future research.

Panels made with 8wt% of adhesive result in an mean flexural strength (19.11 MPa) higher than the minimum requirements stipulated by international standards [14-16], while the other levels of adhesive (10wt% and 12wt%) result in higher mean values than the Brazilian standard [13], both values being higher than the MOR found by Paes et al. [18].

Table 2 presents the interpolation results obtained in Equation 1, revealing the levels of adhesive content (Ad) that maximize each response-variable. The model obtained for bulk density indicates that the highest value occurs when the adhesive content level is 12.42wt%. However, it is worth noting that this factor is not considered significant by the Tukey test. The quadratic model indicates that panels with 15.79wt% of adhesive content achieve the highest flexural modulus (2655 MPa), only 0.75% less
than those obtained for panels produced with 15wt% of adhesive (2675 MPa). The statistical model indicates that the use of 13.86wt% of adhesive content leads to a flexural strength of 24.44MPa, only 0.77% less than 24.63 MPa obtained for panels produced with 15wt% of adhesive.

Table 2. Interpolation between properties and adhesive levels

| Models | Optimal Adhesive Content (%) |
|--------|-----------------------------|
| $\rho$ (g/cm$^3$) = 0.591 + 3.845 $\times 10^{-2}\cdot Ad - 1.548 \times 10^{-3}\cdot Ad^2$ | $Ad = 12.42$wt% $\rightarrow \rho = 0.83$ g/cm$^3$ |
| MOE (MPa) = 857.43 + 227.79 $\cdot Ad - 7.21\cdot Ad^2$ | $Ad = 15.79$wt% $\rightarrow$ MOE = 2655 MPa |
| MOR (MPa) = $-2.504 + 3.889 \cdot Ad - 0.140\cdot Ad^2$ | $Ad = 13.86$wt% $\rightarrow$ MOR = 24.44 MPa |

4. Conclusion

The main conclusions of this work are described below:

(1) The adhesive content does not significantly affect the bulk density ($\rho$) of the manufactured panels.

(2) The adhesive levels of 12wt% and 15wt% lead to equivalent values of flexural modulus (MOE) and strength (MOR), being significantly higher than the MOE and MOR of those panels made with 8wt% adhesive. Due to the equivalence between 12wt% and 15wt% in both properties, it is recommended to manufacture panels with 12wt% of bio-based adhesive.

(3) The bulk density of the panels for both levels of adhesive meets the requirements of Brazilian and international regulatory standards.

(4) In terms of MOE, 12wt% or 15wt% of adhesive levels meets international normative requirements, but not the minimum requirement established by the Brazilian standard.

(5) In terms of MOR, 8wt% of adhesive leads to panels with mean values higher than international normative requirements, while 12wt% and 15wt% are exceed the minimum requirements established by the Brazilian standard.

(6) The adhesive contents responsible for maximizing the values of bulk density, flexural modulus and strength are equal to 12.42wt%, 15.79wt% and 13.86wt%, respectively.

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