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Mechanical Performance of Plywood Bonded with Castor Oil-based Polyurethane Resin Enhanced by Nanocellulose

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The study of engineered wood products has expanded due to their production from renewable raw materials. While wood products have traditionally seen limited use in Brazil, they have gained significant traction over the past two decades, fueled by new products and technological advancements. Among emerging materials, nanocellulose has shown promise due to its potential to enhance the adhesive performance and mechanical properties of wood-based panels. This study aimed to incorporate cellulose nanofibers obtained by mechanical defibrillation of pulp from the extraction of the *Eucalyptus grandis* by kraft process in proportions of 0% and 5% into a castor oil-based polyurethane adhesive commercial to produce plywood panels. The mechanical properties of the panels were analyzed, specifically focusing on static bending strength both parallel and perpendicular to the wood fibers, as well as bond quality through shear testing along the glue line. Results indicated that adding nanocellulose to the polyurethane resin significantly enhanced the mechanical performance of the plywood panels. Additionally, it was observed that using eucalyptus wood veneers with polyurethane resin alone already resulted in improved mechanical properties compared to conventional adhesives. Thermal analysis, suggested that nanocellulose incorporation increased the resin's thermal stability, potentially contributing to enhanced durability. Further studies are recommended to explore the viability of nanocellulose at different proportions and its long-term effects on adhesive performance.

Keywords: *Composite, Nanomaterial, Alternative Adhesive.*

1. Introduction

According to Marchesan and Do Rêgo Barros[1], engineered wood products have undergone significant changes, especially in raw material use, properties, and competitiveness, to enhance their application in furniture, flooring, and construction, and establish themselves as a sustainable alternative to non-renewable materials.

One example of these products is wood paneling, which, according to Gonçalves et al.[2], can be manufactured from wood in various forms, such as fibers, particles, veneers, and battens. These components are bonded through processes involving temperature and pressure in multiple stages, using adhesives, thus enabling the creation of viable alternatives to solid wood.

Panels like plywood are widely used and have advantageous properties due to their layered structure. This structure, formed by veneers arranged perpendicularly, enhances dimensional stability and provides better mechanical stress distribution both transversely and longitudinally[3-5].

According to Vieira et al.[6], the most common adhesives used in wood panel production include phenol-formaldehyde, resorcinol-formaldehyde, urea-formaldehyde, and melamine-formaldehyde. Selecting the appropriate adhesive is essential, and the final use of the panels should be considered. For instance, phenol-formaldehyde adhesive is widely used for plywood intended for outdoor use due to its high-water resistance[7].

However, researchers such as Nakano et al.[8], point out that formaldehyde-based resins pose environmental and health concerns, as some formaldehyde does not fully react, remaining in the panel and releasing emissions over time. In this regard, sustainable adhesives, such as those based on castor oil, have emerged as promising alternatives because they are renewable and safe for health[9].

Previous studies have demonstrated that plywood panels manufactured with a castor oil-based polyurethane adhesive exhibit excellent mechanical performance. Specifically, the strength and stiffness values measured in static bending tests exceeded those reported in the literature for plywood made from the same species (*Eucalyptus grandis*) bonded with conventional phenol-formaldehyde adhesive[10].

New materials have been studied to enhance the properties of wood panels, with nanocellulose being a notable example. This material, which has low toxicity, is biodegradable, exhibits high mechanical strength[11], and can be combined with other materials. Vineeth et al.[12] highlight that the incorporation

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of nanocellulose in adhesives for reconstituted wood panels can modify adhesive properties, enhance the mechanical and physical performance of the panels, and even reduce formaldehyde emissions from synthetic adhesives..

Thus, nanocellulose-reinforced polyurethane resin represents a promising innovation, leveraging nanotechnology to improve the performance of forest-based products[13].

According to Pessan et al.[14], cellulose nanofibers (CNFs) can be separated using mechanical methods, often in conjunction with chemical treatments, and exhibit alternating zones of crystalline and amorphous structures, with widths of 5–10 nm and lengths reaching several micrometers.

Despite its promising characteristics, the effects of nanocellulose incorporation into polyurethane adhesives on the mechanical performance of plywood panels remain underexplored. Therefore, this study aimed to evaluate the impact of adding nanocellulose on the static bending strength and shear resistance along the glue line of plywood panels produced with eucalyptus veneers and polyurethane resin, with a specific focus on the adhesive formulation and interaction mechanisms.

2. Methodology

The panels were produced from *Eucalyptus grandis* donated by the Miraluz Indústria e Comércio de Madeiras Ltda veneers cut to 420 x 420 mm, following the process used by Galdino et al.[9]. The polyurethane adhesive used in this study was derived from castor oil and consisted of two components: a pre-polymer and a polyol. The pre-polymer was synthesized from castor oil and an isocyanate compound, while the polyol component contained hydroxyl groups that facilitated cross-linking during curing. The adhesive is 100% solids and solvent-free. – Commercial adhesive ImpervegAGT 1315 Initially, the veneers were dried in a laboratory oven at 60 ± 3°C for approximately 12 hours. Then, the polyurethane adhesive mixture, in a 1:1 ratio between pre-polymer and polyol, combined with nanocellulose, was manually applied in a double glue line using a plastic spatula[15]. Given the veneers' dimensions of 420 x 420 x 2 mm, an adhesive weight of 80g[15] was used per glue line, with 5% of this amount replaced by nanocellulose. It is worth noting that cellulose nanofibrils were produced through mechanical defibrillation of pulp from the extraction of the *Eucalyptus grandis* by kraft process, using the method proposed by Govea et al.[16].

With each adhesive application, a veneer was laid perpendicularly over the previous one, resulting in a stack of five veneers for each panel. The adhesive open time was carefully controlled to ensure proper bonding before pressing. After assembly, cold pre-pressing was carried out using a manual press with 5 kgf/cm^2 pressure for 10 minutes to compact the panel and facilitate heat transfer during the subsequent hot pressing[15].

The panel was then hot-pressed at approximately 65 bar and 100 ± 5°C for 10 minutes, divided into three pressing cycles of 3 minutes each, interspersed with 30 seconds of pressure relief, in a pneumatic hydraulic press. These pressing cycles were chosen based on preliminary tests to optimize resin curing and panel consolidation. After pressing, the panel was cooled at room temperature and squared off to remove excess material from the edges and ensure a cleaner finish. After curing, the panels were conditioned in a controlled environment at 20°C and 65% relative humidity for 72 hours before testing to ensure moisture equilibrium. Only then were the test specimens extracted for mechanical testing of static bending and shear strength along the glue line, conducted according to ABNT NBR 17002:2021[17] and ABNT NBR ISO 12466-1:2022[18] standards, respectively.

The panel production process is illustrated in Figure 1.

After testing, the data were statistically analyzed using the R software, version 3.4.2 (2017). A normality test (Shapiro-Wilk) and homogeneity of variance test (Levene's test) were performed before applying statistical analyses. A t-test was performed to check for statistically significant differences at a 5% significance level. The results obtained were compared with the literature and with commercial panels.

To characterize the adhesive, its viscosity, gelation time, thermogravimetric behavior (TGA), and differential scanning calorimetry (DSC) properties were analyzed.

The viscosity assessment followed the ASTM D1200-10:2023[19] standard, utilizing a Ford #4 cup. The flow duration was determined using a stopwatch. For the gel time evaluation, 5 g of adhesive was placed in a test tube submerged in glycerin heated to 130°C, with continuous agitation. The time required for gel formation was recorded.

A Perkin Elmer TGA 7 thermogravimetric analyzer was used to carry out the TGA and DSC analyses to

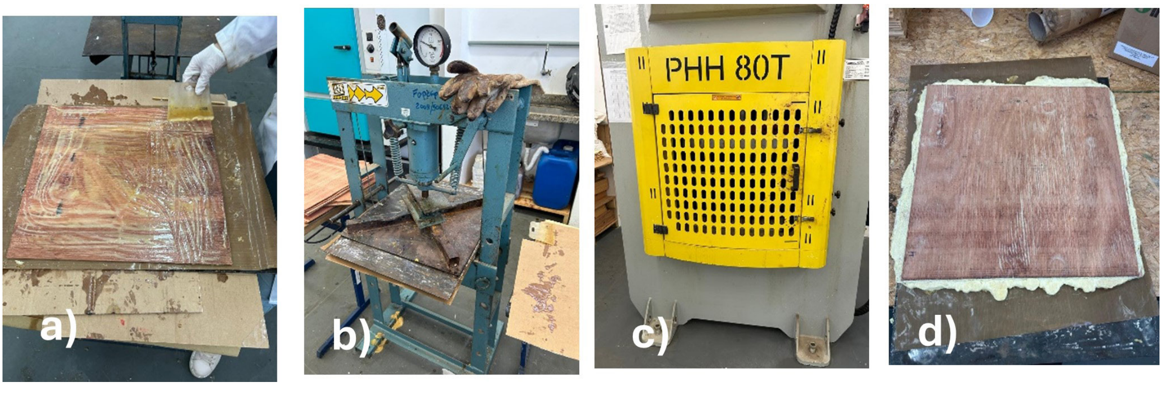

Figure 1. Plywood production - a) Adhesive stage; b) Pre-pressing; c) Hot pressing; d) Finished plywood panel.

determine the suitability of polyurethane resin for plywood manufacturing, particularly in combination with nanocellulose reinforcement. The thermal analyses were performed under a nitrogen atmosphere to prevent oxidation and ensure accurate results. The measurements were conducted across a temperature range of 30°C to 900°C, applying a heating rate of 10°C/min in an inert atmosphere, in line with the protocol described by Galdino et al.[15].

3. Results

Table 1 presents the adhesive's viscosity and gel time properties, both with and without the partial replacement of nanocellulose.

The viscosity remained consistent between treatments 0% and 5%. However, the gelation time decreased with the addition of nanocellulose, which may be attributed to its interaction with the adhesive matrix, altering polymerization dynamics.

Figure 2 presents the TGA and DSC graphs for 0% and 5% nanocellulose.

Analyzing Figure 2 for the polyurethane adhesive, it is possible to identify three degradation stages that occur at approximate temperatures for both samples: 0 ºC to 250 ºC; 250 ºC to 500 ºC, and 500 ºC to 900 ºC. The first stage is related to the initial loss of mass associated with humidity and some volatile compounds. The mass loss of the second stage corresponds to the degradation of the prepolymer and polyol, and in the last stage, which occurs above 500 ºC and extends to 900 ºC, carbonization of the remaining structures occurs. When comparing graphs (a) and (b), greater thermal stability is noticeable when analyzing the peaks, associated with the ability of nanocellulose to act as structural reinforcement in the resin (reference). The initial stage also showed a slight increase in mass loss associated with humidity. This data suggests that the incorporation of nanocellulose improved the thermal resistance of the resin.

Table 2 presents the average results for glue line shear, the modulus of rupture (MOR) and modulus of elasticity (MOE) in the direction parallel (//) and perpendicular (|_) to the fibers. The results obtained differed statistically at a 5% significance level.

Analyzing the values in Table 1, it is evident that using nanocellulose in the panels resulted in superior outcomes compared to ABIMCI[20] standards for boards with 5 layers and a thickness of 15 mm.

The values obtained in the static bending test showed that the replacement of 5% nanocellulose affects positively the MOR and MOE in both directions.

In the research by Kawalerczyk et al.[21], using urea-formaldehyde adhesive in plywood panels, the MOE values confirm the positive influence of the introduction of nanocellulose, resulting in an 8% improvement in samples tested in parallel and 17% improvement in samples tested perpendicularly. The author states that the reason for the improvement may be a decrease in stress concentrations along the bond lines containing nanocellulose. As for MOR, the authors found an improvement of 20% in samples tested parallel to the grains and 32% perpendicularly.

Table 1. Adhesive characterization.

Nanocellulose (%)	Viscosity (cP)	Gel time (s)
0%	664,58	316
5%	664,58	264

Table 2. Results of the glue line shear and bending test (MPa).

Nano (%)	fv	MOR //	MOE //	MOR |_	MOE |_
0	2,26 A (0,88)*	39,11 A (14,41)	6802,80 A (2796,03)	25,75 A (11,43)	2600,14 A (1100,52)
5	2,63 A (0,81)	55,06 B (23,03)	9326,17 B (3097,69)	37,79 B (18,94)	3407,75 B (1608,89)
ABIMCI	2,2	32,90	6933,10	22,70	2633,40

* Values in parentheses refer to standard deviations.

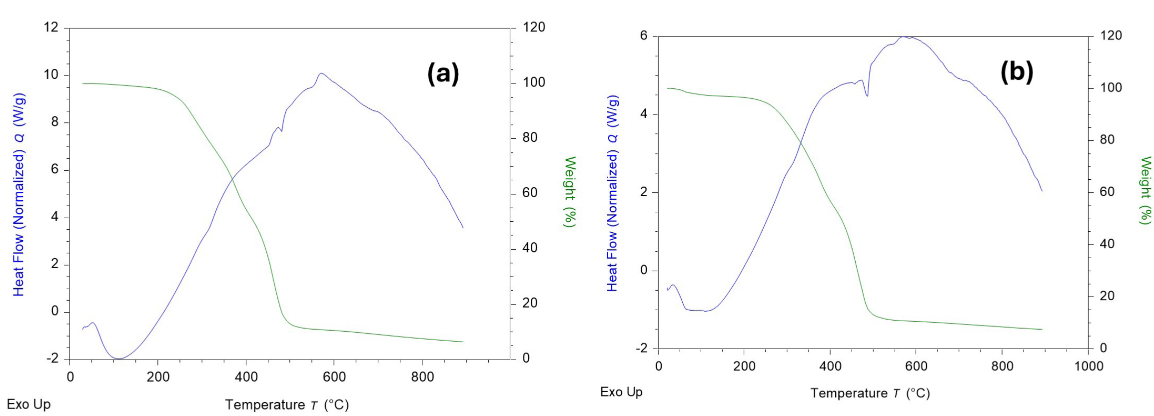

Figure 2. TGA and DSC graphs for polyurethane with 0% (a) and 5% (b) of nanocellulose.

Analyzing the values of glue line shear, it can be stated that the addition of nanocellulose resulted in a slight increase in shear strength, although the difference was not statistically significant at the 5% level.

According to ABNT NBR 12466-2:2022[18], shear strength values above 1MPa are considered acceptable, regardless of whether the rupture occurred in the wood or the adhesive.

Comparing these findings with literature data, Galdino et al.[15] obtained MOR values between 35 and 50 MPa, MOE values between 5000 and 7000 MPa, and shear values between 1.5 and 2.5 MPa in plywood panels produced with polyurethane resin. Our results demonstrate a significant performance improvement, particularly in MOR and MOE values.

Superior performance in parallel bending was achieved by Bortoletto and Moreschi[22], who produced plywood panels with phenol-formaldehyde resin, obtaining values between 85.5 and 132.4 MPa for MOR, and between 12578.9 and 19711.4 MPa for MOE. Despite the lower performance in comparison to phenol-formaldehyde panels, the use of castor oil-based polyurethane combined with nanocellulose represents a sustainable alternative with promising mechanical properties. Further studies are necessary to explore additional improvements in adhesive formulations and nanocellulose integration.

4. Conclusion

This study demonstrated that replacing 5% of the castor oil-based polyurethane adhesive with nanocellulose yielded promising results, particularly in static bending performance in both the parallel and perpendicular directions to the fibers. The incorporation of nanocellulose significantly enhanced the modulus of rupture (MOR) and modulus of elasticity (MOE), surpassing values obtained in the literature. Indicating that the interaction between the adhesive and the wood sheets was greater, justified by the greater stability of the adhesive confirmed by the thermogravimetric analyses performed.

Furthermore, the thermogravimetric analysis (TGA) indicated that the presence of nanocellulose improved the thermal stability of the adhesive, with a delayed onset of mass loss. Although the shear strength along the glue line showed a slight increase, this difference was not statistically significant at the 5% level. Nevertheless, all shear strength values exceeded the values obtained in the literature.

These findings highlight the potential of nanocellulose as a reinforcing agent in polyurethane-based adhesives, contributing to the development of more sustainable and high-performance plywood panels. However, additional research is necessary to optimize the formulation, explore different nanocellulose concentrations, and assess its long-term durability in various environmental conditions. Future studies should also investigate the economic feasibility and scalability of using nanocellulose-modified polyurethane adhesives in commercial plywood manufacturing. Furthermore, studies that evaluate the replacement of other nanocellulose contents in the polyurethane adhesive.

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Data Availability

The complete dataset supporting the findings of this study is available upon request from the corresponding author [Cristiane Inácio de Campos – E-mail: cristiane.campos@unesp.br]. The dataset is not publicly available due to the pending publication of the doctoral thesis following its defense.