

The Use of Nanocellulose in the Production of Medium Density Particleboard Panels and the Modification of Its Physical Properties

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Wood-based panel applications recently have expanded and become increasingly competitive, especially within the furniture and civil construction industries. To remain competitive, such products must present physical properties that meet consumer needs. In this context, the incorporation of nanomaterials is gaining momentum, mainly as a means to improve the physical characteristics of panels, thereby expanding their applications. The aim of this study was to evaluate the physical properties of medium density particleboard (MDP) panels after adding various proportions of nanocellulose in place of water to the urea-formaldehyde (UF) adhesive in MDP panel production. The results showed that the addition of nanocellulose resulted in no significant statistical difference in the density and moisture content of the panels. When tested for thickness swelling, only the panel with 100% nanocellulose solution exhibited a significantly higher value. The panels were subjected to scanning electron microscopy (SEM) analysis, which showed that the addition of nanocellulose led to a more polished, less irregular surface. Such physical effects of nanocellulose can potentially make panels more suitable for coating applications. The feasibility of coatings on nanocellulose MDP panels can be verified through future tests to determine the surface roughness of the panels.

Keywords: Particleboard; Physical characteristics; Nanoparticle

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INTRODUCTION

In recent years, the use of wood products has grown due to their wide range of applications and environmentally friendly attributes. Among several wood products one can find industrially produced wood panels, which have many applications, particularly in furniture manufacturing and civil construction. Plywood sheets, medium density particleboard (MDP), oriented strand board (OSB), high density fiberboard (HDF), and medium density fiberboard (MDF) are some of the most common types of wood panels.

Among the various types of wood panels, MDP panels are considered to be the most advanced due to their manufacturing process and overall quality. Within MDP panels, the

particles are positioned differently, with the larger particles arranged in the core layer of the panel and the thinner particles in the two outer layers (Maciel 2001).

According to Trianoski (2012), there has recently been a persistent effort by the forest industry regarding the consolidation of the MDP manufacturing process. The goal of this consolidation has been to improve the entire production process and profitability through cost reduction measures and efficiency improvements. In the particleboard industry, several companies have invested in items such as machinery in order to improve manufacturing assembly lines (Barbosa *et al.* 2014). As a result of these investments, the quality of particleboard products is continually improving.

Nanoscience involves the handling of physical systems at a nanometer scale, with typical lengths not exceeding 100 nm. In this regard, nanocellulose is a material that shows promising performance results. The depletion of petroleum-based resources and the possible environmental problems of these energy sources has stimulated international interest in the development of environmentally sustainable materials. Plant and wood-based materials, primarily composed of cellulose, hemicellulose, and lignin, are a prime source for the development of sustainable materials (Miao *et al.* 2014).

Cellulose is an abundant natural material derived from sustainable and renewable resources, acting as the structure that reinforces plant assemblies. Due to its low cost, biodegradability, low density, and remarkable physical and mechanical properties, cellulose has been subjected to intense research and development (Klemm *et al.* 2005).

Cellulose has been used as a raw material in several industrial processes, such as in the manufacturing of pulp and paper and synthetic textile fibers used as key components in coatings, optical films, pharmaceuticals, and cosmetics, among others. In addition, the functionality, durability, and uniformity of cellulose makes it a commonly used material in other fields. In recent year, cellulose nanoparticles (CN) have become a popular point of research for the generation of new biomaterials (Habibi *et al.* 2010), several composites that had a type of nanocellulose in its constitution have shown improved performance characteristics or properties which shows potential using nanocellulose (Leng *et al.* 2017).

The unique molecular architecture of natural cellulose consists of fibrils and crystallites that, at the nanoscale, allow for the extraction of nano constituents by mechanical and chemical methods. Cellulose nanofibers (CNF) are long, thin, and flexible formations composed of alternating crystalline and amorphous domains. By contrast, nanocellulose crystals (NCCs) are rod-like, stiff crystalline structures; such particles are released after the amorphous domain division, whereby these crystalline structures are freed from other amorphous structures during the process. Other types of nanocellulose, such as amorphous nanocellulose (ANC) and nanoyarn cellulose (NYC), have also been reported in literature (Dufresne 2012).

According to Rebouillat and Pla (2013) acid hydrolysis is a process that is currently used in research. This process involves subjecting purified cellulosic material into some sort of strong acid, such as phosphoric, maleic, hydrochloric or sulfuric being the last two acids most used nowadays. Other variables in the process should also be accounted for as the acid concentration, the ratio of the cellulose fibers to acid solution, temperature, mixing rate, and reaction time (Rebouillat and Pla 2013)

Several research efforts have focused on different nanocellulose applications, and variations have been developed due to its availability, light weight, nanoscale, unique morphology, and outstanding physical and chemical properties (Pranger and Tannenbaum 2008).

As a new nanoscale biopolymer category, nanocellulose is a promising biobased-

material for several industrial applications, such as chemical, personal care, bio-composites, and pharmaceuticals. The high reinforcing strength and stiffness of nanocellulose make it a material with great potential (Yahya *et al.* 2015).

EXPERIMENTAL

Eucalyptus grandis wood particles were used to produce the MDP panels. The *E. grandis* wood particles were donated by a timber company located on the countryside of São Paulo, Brazil. Urea-formaldehyde (UF) was used as an adhesive, and ammonium sulfate was used as a catalyst. A paraffin emulsion was used as a curing additive. A nanocellulose suspension in water was obtained from the acid-hydrolysis method. The solids content of the resin, the paraffin, and the catalyst were 66%, 57.2%, and 13.1%, respectively.

The experiment was carried out with four different treatments of various nanocellulose concentrations, as seen in Table 1.

Table 1. Characteristics of the Treatments

Treatments	Cellulose Nanoparticles (%)
1	0
2	0.25
3	0.5
4	1

Nanocellulose Production

The nanocellulose was prepared from pre-hydrolyzed *Eucalyptus* kraft pulp. A purification step was performed to eliminate hemicellulose. The nanocellulose was obtained by acid-hydrolysis, process that is known as one of the most effective (Rebouillat and Pla 2013). This was done with sulfuric acid (65%), centrifugation, sonication, and dialysis, a procedure that was adapted from Silva and D'Almeida (2009). The wood was chopped and sieved before undergoing cooking and bleaching. A conventional kraft cooking process was carried out to produce the pulp. All cooking was done in a rotary laboratory digester with electric heating. The digester contained four individual reactors with a capacity of one and a half liters each. The cooking conditions are shown in Table 2.

Table 2. Conditions of the Kraft Process

Temperature (°C)	165
Time to Temperature (min)	60
Time at Temperature (min)	90
Liquor to Wood Ratio	4:1
Sulfidity (%)	30
Active Alkali as Na ₂ O (%)	19

After cooking, the chips were removed from the reactor capsules and washed with water at room temperature using a 0.06 mm (150 mesh) stainless steel screen. After washing, the fibers were placed in laboratory blender at a low consistency. The fibers were then centrifuged to a consistency of approximately 30% and were placed in polyethylene plastic bags for storage.

Bleaching with chlorine dioxide

The concentration of chlorine dioxide (ClO₂) was calculated according to Eq. 1,

$$\left[\frac{g}{L}\right] = \frac{V \times N \times Eq}{V_{ClO_2}} \quad (1)$$

where ClO₂ is the concentration of ClO₂ in g/L, *V* is the volume of sodium thiosulphate (Na₂S₂O₃) used in the titration, *N* is the normality of Na₂S₂O₃, and *Eq* is the amount of ClO₂ that reacts per gram of chloride ion (Cl⁻).

The appropriate chemical quantities were calculated and mixed manually with the pulp in polyethylene bags. The bags were heated in a microwave oven to the desired temperature and transferred to a temperature-controlled bath.

In the oxidative extraction stage, the bleaching liquor containing H₂O, NaOH, and H₂O₂ was added to the pulp at room temperature. After being manually mixed and microwave heated, the material was transferred to a temperature-controlled steam bath, where it was kept for 3 h. The same procedure was followed for the addition of hydrogen peroxide. The bleaching process conditions are shown in Table 3.

Table 3. Bleaching Conditions

Pulp Consistency (%)	10
Temperature (°C)	70
Time (min)	180
H ₂ SO ₄ (mL)	0.65
ClO ₂ (mL)	1508
H ₂ SO ₄ (kg/t)	0.5
ClO ₂ (kg/t)	5

The bleached pulp was then subjected to a production process to obtain nanocellulose. Three grams of pulp were placed in a beaker, and for each gram of pulp, 8 mL of H₂SO₄ was added. The solution was mixed for 35 min with a magnetic stirrer. Upon completion of the reaction, the beaker containing the mixture was placed in a water bath at a temperature of 12 °C. The cooled solution was then diluted with 170 mL of deionized water and centrifuged at a speed of 3,000 rpm for four cycles of 5 min each.

The remaining solution was diluted in 180 mL of deionized water and divided into 9 different samples. Each of these 20 mL samples underwent different filtration and ultrasound times according to Table 4.

Table 4. Ultrasound Conditions

Sample	Ultrasound Time (min)
1A	20
1B	30
1C	40
1D	50
2A	10
3A	20
3B	30
3C	40
3D	50

Samples 1A, 1B, 1C, and 1D were filtered on a 0.45 µm filter before being sonicated. Samples 2A, 3A, 3B, 3C, and 3D were filtered on a 0.45 µm filter after being

sonicated. After obtaining the nanocellulose, the particles were analyzed in a Malvern Zetasizer (Malvern, UK). One mL of each sample was collected with an automatic pipettor and placed in the apparatus for analysis.

MDP Panels Production

The methodology for the production of MDP panels followed the research done by Silva *et al.* (2016). The particles were graded and placed in a greenhouse until a humidity of 3% was reached at 103 ± 2 °C.

The adhesive dosage was fixed based on the dry weight of particles using an 8% dose of adhesive for the inner layer and a 10% dose of adhesive for the outer layers. The formation of the MDP mattress used a total of 2000 g of particles, the distribution of which was 20%, 60%, and 20%, on the outer, inner, and outer layers, respectively. Afterwards, the adhesives for the inner and outer layers were made up. The inner layer adhesive was comprised of 193.1 g of resin, 7.1 g of catalyst, 21.34 g of paraffin, of 19.73 g of water, or nanocellulose solution.

The percentage of water added (19.73g) was calculated in equations proposed by Eleotério (2000). Treatment 1 had no addition of nanocellulose; therefore, 19.73 g of water were added. For treatments 2, 3 and 4 the percentages added are presented in Table 1, resulting in the addition of nanocellulose solution in the proportions of 4.93 g, 9.87 g, and 19.73 g, respectively. For treatments 2 and 3 the mass complementation was performed with water addition.

The preparation of the additives (adhesive, catalyst, paraffin emulsion, water and nanocellulose solution) was mixed in a hand mixer until the constituents were homogenized for further spraying.

The outer layer adhesive was comprised of 128.94 g of resin, 4.69 g of catalyst, and 14.23 g of paraffin. The adhesive materials were mixed in a spinner before being sprayed onto the wood particles.

After sizing, the material was deposited in a forming box with a dimension of 42 cm × 42 cm × 30 cm. The particle mattress was formed manually in a three-layer forming box. The two outer layers were comprised of smaller particles, while the inner layer of the panel was comprised of larger particles. This method created a close replicate of commercially manufactured MDP panels in a 20:60:20 ratio.

After forming, the panels were hot pressed for a total pressing cycle of 600 seconds. Two pressure relief steps lasting 30 seconds at intermediate times were performed. The specific pressure was constant at approximately 40 kgf/cm² and the pressing temperatures were 150 °C and 180 °C.

Tests and Results Analysis

Physical tests were performed to determine the density, moisture content, and thickness swelling after a 24-h water immersion. The sample preparation and the procedures for each test were done in accordance with the ABNT NBR 14810 (2013) standard.

Determination of apparent density

Density was determined using 10 specimens with dimensions of 50 mm × 50 mm. Density calculation were performed according to Eq. 2,

$$D = \frac{m}{w \times l \times t} \times 1,000,000 \quad (2)$$

where D is the apparent density (kg/m^3), m is the mass of the specimen (g), w is the width (mm), l is the length (mm), and t is the thickness (mm). Width and length were measured with a caliper. Thickness was measured at five different points using a micrometer with 0.001 mm of precision. Finally, mass was determined using a precision scale.

Determination of moisture content

Moisture content was determined using 10 specimens with dimensions of 50 mm \times 50 mm. Initial mass was measured using a precision scale. Moisture content calculations were performed according to Eq. 3,

$$U = \frac{m_i - m_d}{m_i} \times 100 \quad (3)$$

where U is the moisture content (%), m_i is the initial mass of the specimen (g), and m_d is the dried mass of the specimen (g). Samples were dried in a laboratory oven at 103 ± 2 °C until reaching a constant mass. Fluctuation between mass measurements were less than 0.1%.

Determination of thickness swelling

To determine the thickness swelling, 10 specimens with dimensions of 50 mm \times 50 mm were submerged in water. Thickness swelling calculations were performed according to Eq. 4,

$$TS = \frac{t_s - t_i}{t_i} \times 100 \quad (4)$$

where TS is the thickness swelling (%), t_s is the specimen thickness after soaking (mm), and t_i is the initial thickness of the specimen (mm). Specimens were soaked for 24 h before being tested for thickness swelling. Measurements were done by a micrometer with 0.001 mm of precision.

The results from the physical tests were analyzed using analysis of variance and Tukey tests. A significance level of 5% was selected and *R* software version 3.3.1 was used for the data analysis. Each treatment was split into six variations.

RESULTS AND DISCUSSION

The average values for the density, moisture content, and thickness swelling tests are listed in Table 5. All treatments presented density values in accordance with ABNT NBR 14810 (2013), which indicates a standard density range of 0.55 g/cm^3 to 0.75 g/cm^3 .

The addition of nanocellulose increased the moisture content of the panels, which is explained by the fact that nanocellulose is used in aqueous solution, which increased the equilibrium moisture. Although the moisture content of the panels increased with increasing nanocellulose content, statistical analysis showed no significant difference. The Brazilian Standard ABNT NBR 14810 (2013) does not indicate ideal reference values for this parameter.

A Tukey test at a 5% significance level revealed that greater amounts of nanocellulose in the samples resulted in a significant difference in the swelling. Similar results were seen in previous work done by Cardoso *et al.* (2016), also as was studied by Amini *et al.* (2017), the panels presented an increase in the thickness swelling, mostly due to its increase in density. According to ABNT NBR 14810 (2013), the reference values

for thickness swelling of comparable samples after a 24-hour immersion in water should not exceed 15% (class Type P4). Treatment 4 was the only condition that did not meet this value. Class P4 is made up of non-structural panels for use in dry conditions.

Table 5. Average Values Obtained for the Physical Tests of Density, Moisture Content, and 24 h Thickness Swelling

Treatment	Nanocellulose Solution	Density (g/cm ³)	Moisture Content (%)	24-hThickness Swelling (%)
1	0%	0.70 a	16.88 a	7.67 b
2	25%	0.66 a	19.49 a	10.17 b
3	50%	0.64 a	22.73 a	10.50 b
4	100%	0.69 a	23.83 a	16.83 a

Means followed by equal letters present no statistical difference (Tukey, $\alpha = 0.05$)

Scanning electron microscopy (SEM) with an aperture size of 100 μm was used to analyze the physical effects of nanocellulose. The SEM images for all four treatments are shown in Fig. 1. The nanocellulose fills empty space in the material, making the surface sleeker, which may interfere with mechanical testing.

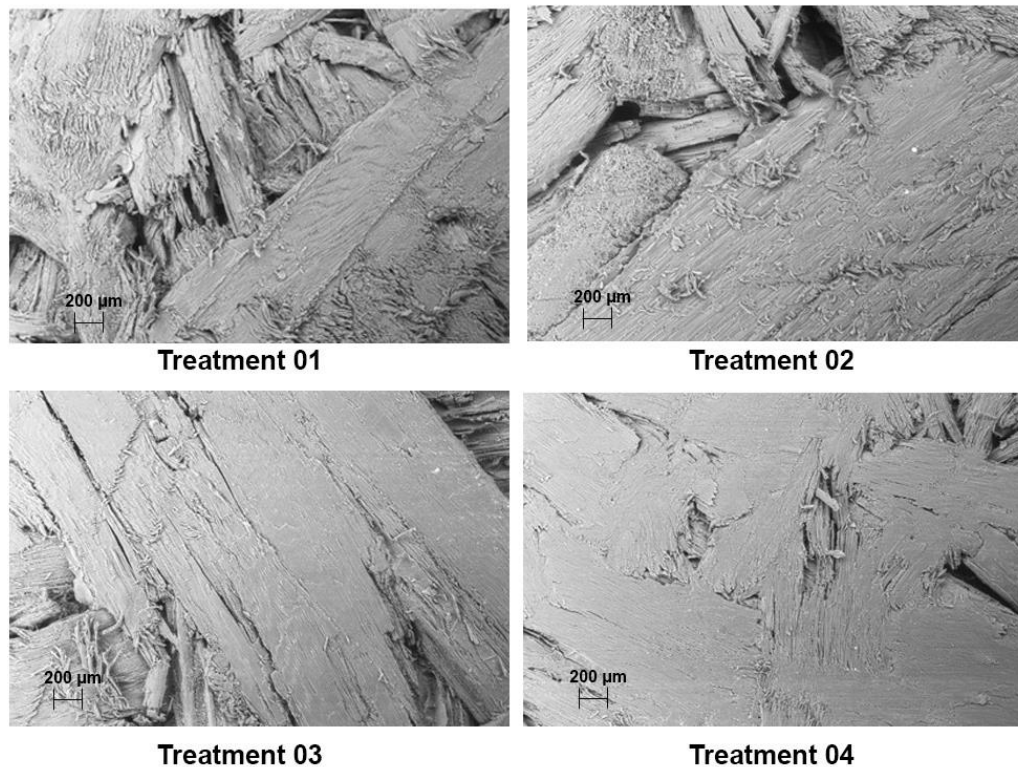


Fig. 1. Visual analysis done by SEM imagery for all treatments.

CONCLUSIONS

1. The laboratory production of MDP panels with nanocellulose presented promising results from physical tests, indicating the possibility of laboratory scale production.
2. Microscopic analysis through SEM imagery showed that a higher percentage of added nanocellulose made the surface of the panel glossier and smoother. Panels with added nanocellulose also exhibited fewer irregularities, indicating a better surface quality, which provides ideal conditions for paint jobs, varnishes, and coatings. Such assertions can be demonstrated with future roughness testing.

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