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RICARDO GABRIEL DE ALMEIDA MESQUITA

**THE USE OF LIGNOCELLULOSIC
BYPRODUCTS AND CELLULOSE
NANOCRYSTALS WITHIN PARTICLEBOARD
PRODUCTION**

LAVRAS – MG

2017

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Tese apresentada a Universidade Federal de Lavras e Universidade de Copenhague como parte das exigências do Programa de Pós-Graduação da Faculdade de Ciências e do Programa de Pós-Graduação em Ciência e Tecnologia da Madeira, área de concentração em Processamento e Utilização da Madeira, para obtenção do título de Doutor (Dupla titulação).

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NANOCRISTAIS DE CELULOSE NA PRODUÇÃO DE PAINÉIS
PARTICULADOS)**

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RESUMO

Neste trabalho, objetivou-se estudar a produção de painéis MDP utilizando-se como matéria prima, partículas de *Eucalyptus*, filamentos de sisal, filamentos de coco e partículas de bagaço de cana. Todos os painéis foram confeccionados em 3 camadas (face/miolo/face) com a utilização de ureia-formaldeído (UF). Os filamentos de coco e sisal foram utilizados em associação com partículas de *Eucalyptus*, somente na camada interior (miolo), em proporções de 0, 10 e 20%. As partículas de bagaço de cana foram separadas em medula e fibras e os painéis foram fabricados, utilizando-se somente fibras e também utilizando-se fibras nas faces e medula no miolo. Os painéis de bagaço de cana também foram fabricados utilizando-se 1% de nanocristais de celulose (NCC) nas faces dos painéis, tanto de fibras como de fibras/medula. A proporção de NCC foi definida confeccionando-se corpos-de-prova de ureia-formaldeído reforçados com diferentes proporções de NCC (0, 0,5, 1, 2, 3 e 5%). Para a confecção dos painéis MDP foi utilizado ciclo de prensagem com as seguintes características: temperatura de 160°C, pressão de 4 MPa e tempo de 8 minutos. Foram avaliadas as seguintes propriedades: absorção de água (WA), inchamento em espessura (TS), densidade, razão de compactação, módulo de elasticidade (MOE); módulo de ruptura (MOR), ligação interna (IB), condutividade térmica (TC). Os painéis reforçados com sisal não apresentaram resultados satisfatórios para a maioria das propriedades avaliadas: MOE, MOR, WA, IB e TS, porém os painéis reforçados com coco, apresentaram propriedades muito similares aos painéis confeccionados somente com partículas de *Eucalyptus*, demonstrando o potencial desse material. Os feixes de sisal formaram tufo de feixes que prejudicaram a dispersão do adesivo no interior dos mesmos, refletindo nas propriedades físico-mecânicas. Os corpos-de-prova de UF reforçados com NCC (1, 2 e 3%) apresentaram resultados superiores (MOE e MOR) aos corpos-de-prova sem reforço, porém o mesmo não foi observado quando os NCC foram aplicados aos painéis de bagaço de cana. Os NCC provavelmente conseguiram se ligar com o adesivo, diminuindo a quantidade de sítios para as ligações entre o adesivo e as partículas de bagaço de cana. Os painéis fabricados de fibras e de fibras/medula apresentaram propriedades estatisticamente iguais para a maioria das propriedades avaliadas: MOE, MOR, WA e TC. Os painéis de bagaço de cana apresentaram potencial para a produção de painéis MDP.

Palavras-chave: Fibras vegetais. Resíduos. *Eucalyptus*. Uréia-formaldeído. Aglomerado. MDP.

ABSTRACT

The objective of this work was to study the particleboard production using *Eucalyptus* particles, sisal filaments, coconut filaments (coir) and sugarcane bagasse particles (SCB). All particleboards were made in three layers (face / core / face) using urea-formaldehyde adhesive (UF). Coir and sisal filaments were used in association with *Eucalyptus* particles, only in the inner layer (core), in proportions of 0, 10 and 20%. The SCB particles were separated into pith and fibers and the particleboards were made using only fibers and using fibers on the faces and pith in the core. SCB particleboards were also manufactured using 1% of cellulose nanocrystals (NCC), by adhesive mass, on the faces of the particleboards. The proportion of NCC was defined using urea-formaldehyde specimens, reinforced with different proportions of NCC (0, 0.5, 1, 2, 3 and 5%). For the particleboards production, a pressing cycle was used with the following characteristics: temperature of 160°C, pressure of 4 MPa and time of 8 minutes. The properties evaluated were: water absorption (WA), thickness swelling (TS), density, compaction ratio, modulus of elasticity (MOE), modulus of rupture (MOR), internal bond (IB) and thermal conductivity (TC). The particleboards reinforced with sisal did not show satisfactory results for most of the properties evaluated (MOE, MOR, WA, IB and TC), but those reinforced with coconut, showed properties very similar to the particleboards made only with *Eucalyptus* particles, demonstrating the potential of this material. The filaments of sisal formed tufts that impaired the dispersion of the adhesive inside the tufts, reflecting in poor physical-mechanical properties. The specimens of UF reinforced with NCC (1, 2 and 3%) presented higher results (MOE and MOR) than non-reinforced specimens did, but the same was not observed when the NCC were applied in the SCB particleboards. NCC were probably able to bind with the adhesive, decreasing the amount of sites for the bonds between the adhesive and SCB particles. Particleboards made with fibers and fibers/pith showed properties statistically equal for most properties (MOE, MOR, WA and TC). SCB showed potential for MDP production.

Keywords: Plant fibers. Residues. *Eucalyptus*. Urea formaldehyde. Chipboards. MPD.

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LISTA DE ABREVIATURAS
(LIST OF ACRONYMS)

MDP	Medium density particleboard
SCB	Sugarcane bagasse
CNF	Cellulose nanofibers; cellulose nanofibrils; nanocellulose
MFC	Microfibrillated cellulose
NCC	Cellulose nanocrystals; nanocrystalline cellulose; whiskers
UF	Urea formaldehyde
MUF	Melamine urea formaldehyde
PF	Phenol formaldehyde
WA	Water absorption
WA2h	Water absorption after two hours of immersion
WA24h	Water absorption after twenty-four hours of immersion
TS	Thickness swelling
TS2h	Thickness swelling after two hours of immersion
TS24h	Thickness swelling after twenty-four hours of immersion
MOE	Modulus of elasticity
MOR	Modulus of rupture
IB	Internal bond
TC	Thermal conductivity
VHC	Volume heat capacity
CR	Compression ratio
TNRR	Thickness non-return ratio
TGA	Thermogravimetric analyzer
DSC	Differential scanning calorimetry
FC	Flexibility coefficient
WF	Wall fraction

LISTA DE SIMBOLOS
(LIST OF SYMBOLS)

EU	Particleboards made with <i>Eucalyptus urophylla</i>
CO-10	Particleboards made with <i>Eucalyptus urophylla</i> + 10% of coir
CO-20	Particleboards made with <i>Eucalyptus urophylla</i> + 20% of coir
SI-10	Particleboards made with <i>Eucalyptus urophylla</i> + 10% of sisal
SI-20	Particleboards made with <i>Eucalyptus urophylla</i> + 20% of sisal
UF-0	Composites made with urea formaldehyde without cellulose nanocrystals
UF-0.5	Composites made with urea formaldehyde + 0.5% of cellulose nanocrystals
UF-1	Composites made with urea formaldehyde + 1% of cellulose nanocrystals
UF-2	Composites made with urea formaldehyde + 2% of cellulose nanocrystals
UF-3	Composites made with urea formaldehyde + 3% of cellulose nanocrystals
UF-5	Composites made with urea formaldehyde + 5% of cellulose nanocrystals
SCB-0	Particleboards of sugarcane bagasse made without cellulose nanocrystals
SCB-0.5	Particleboards of sugarcane bagasse + 0.5% of cellulose nanocrystals
SCB-1	Particleboards of sugarcane bagasse + 1% of cellulose nanocrystals
SCB-2	Particleboards of sugarcane bagasse + 2% of cellulose nanocrystals
SCB-3	Particleboards of sugarcane bagasse + 3% of cellulose nanocrystals
Pith-0	Particleboards made with pith particles from sugarcane bagasse without cellulose nanocrystals

- Pith-1 Particleboards made with pith particles from sugarcane bagasse with 1% of cellulose nanocrystals
- Fiber-0 Particleboards made with fiber particles from sugarcane bagasse without cellulose nanocrystals
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FIRST PART

1 INTRODUCTION

1.1 General introduction

Wood boards are made of smaller elements of the breakdown of wood, such as plates, veneers laths, particles, chips and fibers, which are then bonded with an adhesive under a certain pressure and temperature to produce boards (IWAKIRI, 2005; KELLY, 1977; MALONEY, 1996).

Among the different kinds of particleboards, there are the Medium density particleboard (MDP) and chipboards. The chipboards are composed of wood particles called slivers and they are produced in a single layer (IWAKIRI, 2005; KELLY, 1977; MALONEY, 1996).

The MDPs can be considered an evolution of the chipboards. They are produced in three layers, two of them with smaller particles in the faces and one with bigger particles in the core of the board. They are usually produced with wood particles, and they are mainly used for furniture/internal purposes. The raw material typically used in Brazil is wood from the planted forests of *Pinus* and *Eucalyptus* (BARROS FILHO, 2009). However, any lignocellulosic material can be used for the manufacture of particleboards and it is possible to mix different types of wood with other materials (COLLI et al., 2010; MESQUITA et al., 2015; SORATTO et al., 2013; TRIANOSKI et al., 2013). The use of these lignocellulosic fibers, as reinforcement, has aroused great interest in developing countries, because of their low cost, availability, energy saving, renewable source, non-toxic nature, and also with regard to the environmental issues (MACVICAR; MATUANA; BALATINECZ, 1999; SOYKEABKAEW, 2009; YANG; KIM; KIM, 2003).

With an increase in consumption of wood products, the availability of wood decreases and its cost increases, thereby motivating a search for materials

with similar characteristics, but with higher availability and lower cost. In this context, new lignocellulosic materials have been evaluated for the production of particleboards. Among the possibilities, agricultural residues have emerged as a solution with low cost and high availability (CARVAJAL; VALDÉS; PUIG, 1996; CÉSAR et al., 2014; KWON; AYRILMIS; HAN, 2013; SANTOS et al., 2014).

One of these materials that have a potential are sisal fibers, which are extracted from the leaves of the plant *Agave sisalana* Perr. Sisal has interesting characteristics for reinforcement, both in composites and boards, due to its good mechanical properties (modulus of elasticity and modulus of rupture), low density, and high availability (BRAZILIAN FIBRES, 2015; MARTIN et al., 2009). Brazil is the largest producer of sisal fibers in the world, 120,000 t per year with yields of around 800-1200 kg/ha, and accounts for 58% of the world production (BRAZILIAN FIBRES, 2015).

Another material with potential uses are the fibers from the husk of coconuts, commonly known as coir. The Brazil wastes most of its coconut husk that produces coir fibers, whereas countries like India and Sri Lanka are selling their fibers for multiple uses and generate money for the local economy. It is estimated that the waste amounts to more than US\$ 60 million per year in Brazil (BRAZILIAN FIBRES, 2015).

Studies have shown that residues from sugarcane can be easily formed into particles/fibers, as is done with wood. These researches also contribute to the use and recycling of waste generating environmental gain (BATTISTELLE; MARCILIO; LAHR, 2009; DOOST-HOSEINI; TAGHIYARI; ELYASI, 2014; MENDES et al., 2009; YANG; KIM; KIM, 2003).

Sugarcane bagasse (SCB) is a residue of matted cellulose fiber derived from sugarcane milling process. It has been burning in steam reservoirs to produce energy for industrial use. Sugarcane bagasse has been increasingly

produced in larger quantities due to increased acreage and industrialization of sugarcane, arising mainly from public and private investments in sugarcane production (BOLETIM DE CONJUNTURA ENERGÉTICA, 2013; FIORELLI et al., 2011). However, SCB has some drawbacks such as: high water absorption (WA) due to the high amount of hemicellulose, which means more bonding sites (LI et al., 2011). A high amount of ash and extractives can affect the adhesive cure procedure, as also the adhesion between particles and the adhesive. Furthermore, fibers with less stiffness are damaged due to the mechanical process of milling (GUIMARÃES JÚNIOR et al., 2012).

New technologies have been tested, especially in the area of nanotechnology. Moreover, the reinforcement of composites/boards with nanofibers and nanocrystals (from cellulose) has been shown to be very promising as these materials show a gain in their properties (ATTA-OBENG; VIA; FASINA, 2012; VEIGEL et al., 2012; ZHANG et al., 2011; ZORBA et al., 2008). Improvements in the mechanical properties (internal bond and bending strength) of particleboard reinforced with nanocellulose (CNF) and microfibrillated cellulose (MFC) were found by (MAHRDT et al., 2015; VEIGEL et al., 2012)), and improvements in physical properties (thickness swelling) by Veigel et al. (2012).

New uses of these materials can result in social, economic and environmental development of the country. Sisal cultivation is concentrated in the semiarid region of northeastern Brazil, which is a region that has been exploring just some few economic alternatives. Small farmers produce most of the sisal with a predominance of family agriculture (BRAZILIAN FIBRES, 2015). New uses for coir and sugarcane bagasse can transform the waste into useful products, by contributing to the financial sustainability and diversification of sources of revenue by adding value to waste, and reducing the use of wood and the consequent pressure on forests.

1.2 Objectives and structure of the thesis

The objectives of this study were to produce and characterize MDP using sisal and coconut filaments in combination with *Eucalyptus* particles, and also, to produce and characterize MDP using sugarcane bagasse (pith and fibers) reinforced with nanocrystals of cellulose (NCC), evaluating factors that may interfere with their properties.

In chapter 1, it was investigated the feasibility of coir and sisal inclusion in the core layer of MDPs. The sisal and coir filaments, with 3 cm of length, were included using three proportions (0, 10 and 20%) in association with wood particles from *Eucalyptus urophylla*. In chapter 2, first, it was investigated the effects of NCC inclusion in UF adhesive, using six proportions (0, 0.5, 1, 2, 3 and 5%). Later on, it was investigated the effects of NCC inclusion in UF and applied to SCB particleboards. In chapter 3, it was investigated the NCC inclusion (1%) in UF and applied only in the faces of SCB MDP. In addition, it was investigated the effects of using two materials (pith and fiber) for SCB MPD production. The MDPs were produced using only particles from the rind part of the SCB (called fiber) and also using particles of the SCB inner part (called pith) in the core of the MDPs.

2 LITERATURE REVIEW

2.1 Wood boards

Wood boards (or wood panels) can be defined as compounds of elements of the breakdown products of wood, such as plates, veneers, laths, particles, chips and fibers, which are then bonded with an adhesive under a certain pressure and temperature to produce the boards. These products are also known as reconstituted wood products/engineered wood products and their properties are different from the original wood material (IWAKIRI, 2005; KELLY, 1977; MALONEY, 1996). There are several types of such boards, but they can be separated into three groups according to their constituent material (Figure 1): the laminates (that use wood veneers); the particulate (that use wood/lignocellulosic particles/chips), and the fibrous (that use individual fibers). In each group, there are different kinds of boards and the main boards are showed in the Figure 1. Some of those boards are illustrated in Figure 2.

Figure 1 - Wood boards classification and the main kinds. Laminated veneer lumber – LVL. Medium density particleboard – MDP. Oriented strand board – OSB. Cement bonded particleboard – CBPB. Medium density fiberboard –MDF.

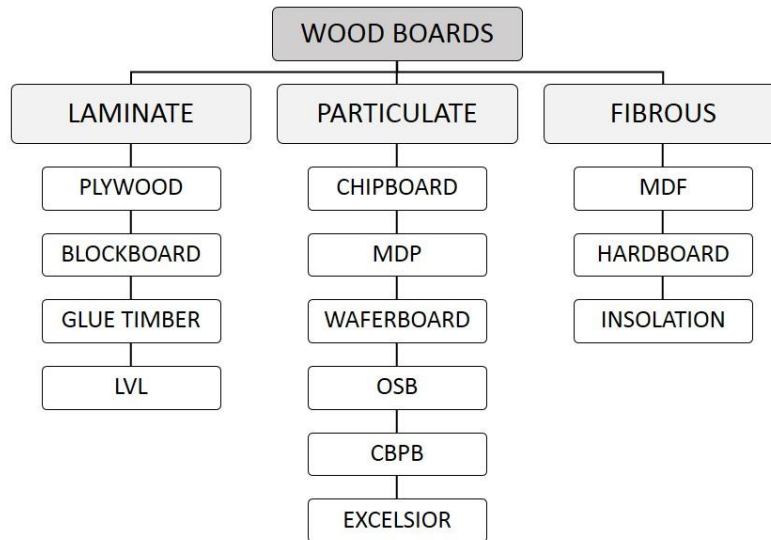


Figure 2 - Illustrative images from wood boards.



Blockboard



Oriented strand board - OSB



Plywood (5 veneers)



Chipboard (one layer)



Medium density fiberboard - MDF



Medium density particleboard – MDP
in three layers with sisal inclusion in
the core

2.2 Coir

From the coconut shell, fiber filaments (Figure 3) are extracted of different lengths, used for the manufacture of a rich variety of items such as clothing, carpets, sacks, pillows, mattresses, and quilts for the automotive industry, brushes, mats, runners, marine ropes, and cork insulation (FERREIRA; WARWICK; SIQUEIRA, 1997).

Figure 3 - Filaments from coco.



The coconut palm (*Cocos nucifera* L.) belongs to the Arecaceae family and has a fasciculate root system, stem stripe type, compound leaves, paniculate inflorescences, and fruit drupe. The fruit is formed by a smooth skin or epicarp, which involves the thick mesocarp fiber, getting to within a very hard layer, the endocarp (FERREIRA; WARWICK; SIQUEIRA, 1997).

The fruit is cut up. From time immemorial, in the Far East and Oceania, "the fiber" (coir) is used in cables, ropes, fishing nets and caulk boats. It is very resistant to the action of salt water, very elastic, as it can stretch to 25% of its own length without breaking. The cells forming the coir are short and only reach a length of 0.7 millimeters. The coir preparation consists of six operations: tannery, pressure, carding, cleaning, sorting and baling (GOMES, 1984).

Brazil wastes most of its coconut husk and coir fibers, while countries like India and Sri Lanka are selling their fibers for multiple uses and generate

money for the local economy. It is estimated that the waste amounts to more than US\$ 60 million per year in Brazil (BRAZILIAN FIBRES, 2015).

Colli et al. (2010) produced particleboard from Paricá wood (*Schyzolobium amazonicum*) with coir (0, 10, 20, and 30%), using UF, 170°C of temperature and 3.1 MPa of pressure. For the bending modulus (MOE), the coir inclusion did not change the mean values, however the mean values for bending strength (MOR) were higher for boards with a high amount of coir (20 and 30%). Colli et al. did not find a difference between the mean values for water absorption (WA) and thickness swelling (TS). Khedari, Charoenvai e Hirunlabh (2003) produced particleboards with only coir and the boards had 53% for TS and 78% for WA (high amount). They found mean values for thermal conductivity of $0.1445 \text{ Wm}^{-1}\text{K}^{-1}$ (0.648 g.cm^{-3} of board density). Lower values ($0.1117 \text{ Wm}^{-1}\text{k}^{-1}$) were found for particleboards (0.611 g.cm^{-3}) made with 90% coir and 10% durian (KHEDARI et al., 2004).

Corradini et al. (2009) studied the chemical composition of five *Cocos nucifera* varieties (green coco fibers). They found that the lignin content varied from 37.2 to 43.9% and cellulose content from 31.5 to 37.5%. Those values are in agreement with the literature (SATYANARAYANA; PILLAI; SUKUMARAN, 1982; SILVA et al., 2000). Due the amount of lignin, the fibers from coco have high durability, however due the lower cellulose content and the high microfibrillar angle the composites made with them have inferior performance when compare with sisal (BISMARCK et al., 2001).

2.3 Sisal

Sisal fibers are extracted from the leaves of the plant *Agave sisalana* Perr. (Figure 4). Sisal has interesting characteristics for reinforcement, both in composites and boards, due to their good mechanical properties, low density, and high availability (BRAZILIAN FIBRES, 2015; MARTIN et al., 2009).

Brazil is the largest producer of sisal fibers in the world 120,000 t per year with yields of around 800-1200 kg/ha, and accounts for 58% of the world production (BRAZILIAN FIBRES, 2015). In the industry, sisal is transformed into several varieties of yarns, cords, mats, and blankets, being used for making various products, such as, in reinforcing plastic composites, where researches have shown promising results (BRAZILIAN FIBRES, 2015; CARVALHO; CAVALCANTI, 2006; JOSEPH; MEDEIROS; CARVALHO, 1999; SINGH; GUPTA; VERMA, 1996).

Figure 4 - Sisal filaments.



Sisal belongs to the group of monocots, the Agavaceae family, and the genus *Agave*. The sisal *Agave sisalana* Perr is native from Yucatán (Mexico) and it was introduced in Brazil in 1903. It has a fibrous root system, has no stem, and leaves are free of petioles, reaching two meters long (MEDINA, 1954).

Anatomically sisal fibers belong to the group of structural fibers whose function is to support and give rigidity to the leaves. The extracted material is in the form of filaments, ranging in length from 40 to 160 cm and one-eighth to one-third millimeter diameter, and a thick base, with an angular section or almost cylindrical. The fibrous filaments, in turn, consist of a large number of juxtaposed cells (fibers) and are intimately welded by a pectin substance of nature, which does not separate into its elements during manufacturing operations. These filaments are commonly called fibers and are thick, rough, and hard, and are classified as hard "fibers" (BISMARCK et al., 2001; MEDINA, 1954). Cells (actual fibers) that constitute the sisal filaments are elongated, thick-walled, have a reduced lumen, and a polygonal section measuring 0.8 to 7.5 mm in length with 7 to 47 μm of diameter (MEDINA, 1954).

Different from coir, that has higher lignin amount, sisal has lower lignin content (12%) and higher cellulose content (67%). This higher cellulose amount and low microfibrillar angle (20°) provide high tensile strength (468-640 MPa) and Young's modulus (9.4-22 GPa) to sisal fibers (SATYANARAYANA; PILLAI; SUKUMARAN, 1982).

Mesquita et al. (2015, 2016) studied the use of sisal filaments in association with *Eucalyptus* particles for Medium density particleboard (MDP) production. However, the results were not satisfactory for most properties. The sisal filaments tend to form clumps of fibers harming adhesive dispersion (Figure 5).

Figure 5 - Clumps formation during the adhesive dispersion and the mattress formation



2.4 Sugarcane bagasse

The Portuguese introduced sugarcane (*Saccharum officinarum*) in Brazil in the early sixteenth century, in two states, Pernambuco and São Paulo. There were not many changes until the early 1970s, when was created the Brazilian Alcohol Program (PROÁCOOL), which caused a large increase in the area planted with sugarcane in Brazil (MENDES, 2008).

Brazil is one of the major producers of sugarcane and consequently generates thousands of tons of lignocellulosic residues of sugarcane bagasse. For the season 2016-2017, the culture of sugarcane continues to expand. The forecast is that Brazil has an increase in area of about 318.4 thousand hectares, 3.7% more than in the 2015-2016 harvest. The increase reflects an increase in

area in the Southern Region. The area under sugarcane will be harvested and the destined sugarcane harvest activity in 2016-2017 is estimated to be 8,973.2 thousand hectares. Despite the climatic instability in some producing regions, the outlook is for growth in crop yield. The total forecast of sugarcane to be crushed is 684.77 million tons (COMPANHIA NACIONAL DE ABASTECIMENTO - CONAB, 2016).

Sugarcane is comprised of about 30% juice. The remainder is biomass (bagasse and straw) and inorganic compounds such as silica. Bagasse is the residue generated after grinding and in the process, the fiber is separated from the juice. After the juice extraction, bagasse consists of 46% fiber, 50% water, and 4% dissolved solids. The residue consists of a set of particles with sizes ranging from 1 to 25 mm. Bagasse can be reused as fuel generating energy, such as fertilizer, pulp, food, and more recently has been applied in particleboards and composites (FIORELLI et al., 2011; LOH et al., 2013; MENDES, 2008; MENDES et al., 2010).

The chemical composition of bagasse varies according to several factors, such as, type of cane, soil type, and harvesting techniques. Cellulose content, lignin and hemicellulose present in the pith (not fiber fraction) and the fibers are very close to each other and to those of hardwoods. However, pith and true fiber are different from the morphological point of view regarding their physiological functions and as regards their liquid-absorption capacities. The percentage of the pith varies from around 30 to 35%, while that of the fibers is around 65% (DRIEMEIER et al., 2011; LEE; MARIATTI, 2008; WIDYORINI et al., 2005). The fiber that bagasse has in it comprises of cellulose 27-54%, hemicellulose 14-24%, and lignin 23-30% (MENDES, 2008).

Sugarcane bagasse is used in the sugar and ethanol sector to generate heat, steam, and energy in the production process. The volume of bagasse from the sugarcane harvest was 174 320, 3 x 10³ tons; this value was greater than the

one presented in 2012, which was $140,627.4 \times 10^3$ tons (BOLETIM DE CONJUNTURA ENERGÉTICA, 2013). With this amount of bagasse would be possible to produce 240 million m³ of MDP, this value is two times greater than the production of particleboards in 2014, 110,862,602.00 m³ (FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED STATES - FAO, 2014).

2.4.1 Research using sugarcane bagasse for particleboards production

At present, the most common application of sugarcane bagasse particleboards among countries that industrially market them is for the furniture market. As the use of panels is becoming more frequent it is necessary for the research to meet the functional and environmental economic demands, to promote the proper application of the material's function (CARVALHO, 2012; LEE; SHUPE; HSE, 2006; LOH et al., 2013).

Teixeira, Costa and Santana (1997), assessed the resistance particleboard of sugarcane bagasse (without pith) glued with adhesive based tannin and urea when subjected to degradation by wood-destroying fungi, in an accelerated laboratory test. The plates of sugarcane bagasse cluster when using tannin-based adhesives (tannin-LPF and phenotan-AG) and urea-formaldehyde, and were equivalent to the fungal attack resistance of *P.sanguineus* and *G.trabeum*. The three treatments were classified as "moderately resistant" to the attack by these fungi according ASTM D 2017-81 (AMERICAN SOCIETY FOR TESTING AND MATERIALS - ASTM, 2005).

Mendes (2008) investigated the influence of different levels (6, 9 and 12%) and types (phenol and urea) of adhesives on the physical-mechanical properties of particleboard made with sugarcane bagasse resulting from the production of cachaça. The author noted that for water absorption (WA) and thickness swelling (TS) there was no significant effect of adhesive type. However, there was a statistical difference for different levels of adhesives, and

the contents 9 and 12% were statistically equal, while 6% was shown to be inferior to them. As for the mechanical properties of (bending modulus) MOE, the internal bond and compression, there was no statistical difference for both types as also for adhesive content. It should be noted that the values obtained for the mechanical properties had not reached the minimum value stipulated by the CS 236-66 (COMMERCIAL STANDARD - CS, 1968) standard and the compression ratio values were well above the range recommended in literature.

In addition, Pedreschi (2009) evaluated the production of particleboards of sugarcane bagasse, having as variables the constitution of panels, types, and adhesive levels to compare their performance with the performance of commercial particleboards of sugarcane bagasse from China and the commercial panels of *Eucalyptus* in Brazil. The author noted that the homogeneous panels (one layer) were statistically greater or equal to those produced in three layers. As for the properties of MOE, MOR, and WA there were no statistical differences between the two types of adhesives used (MUF and UF). The author also noted that the best treatments obtained for sugarcane bagasse particleboards produced in the laboratory, as well as the boards produced on an industrial scale in China, were statistically equal to the ones that were industrially produced with *Eucalyptus* wood.

Mendes et al. (2010) investigated the association of eucalyptus particles with bagasse particles in different proportions (25, 50 and 75%), besides different adhesives (phenol and urea) and different adhesive ratios (6, 9, 12%). Only panels with 25% exhibited compression values within the range recommended in the literature. This was due to the low density of sugarcane bagasse. The results showed that there was no significant effect on the percentage of bagasse associated with *Eucalyptus* wood for thickness swelling after 2 and 24 hours, water absorption after 24 hours and modulus of elasticity. All treatments met the minimum value set for thickness swelling as for the

standard CS 236-66 (CS, 1968). For the modulus of elasticity, any treatment reached the minimum value stipulated by the standard, while for MOR and internal bond of the particleboards produced with urea formaldehyde showed values higher than those stipulated by the standard did.

Barros Filho et al. (2011) evaluated the properties of bagasse particleboards with two types of adhesives (urea-formaldehyde and melamine-urea-formaldehyde), with and without paraffin and with and without inclusion of particles of wood (*Eucalyptus* and pine). On an average, the internal bond values of sugarcane bagasse particleboards with wood particles are superior to those produced only with sugarcane bagasse. Moreover, among them the panels including pine showed the best performance. The panels made with only sugarcane bagasse, more paraffin, and adhesive melamine-urea-formaldehyde, showed lower values of WA24h and TS24h (91.2% and 18.13%) and higher values of MOE and MOR (1129 and 5.91 MPa) compared with particleboards made with UF and in association with wood.

Fiorelli et al. (2011) evaluated the potential use of polyurethane resin castor oil based on the production of sugarcane bagasse particleboards. The authors concluded that this resin is shown as an efficient alternative for the production of particleboard of sugarcane bagasse. Also, it is possible to produce bagasse particleboards in the laboratory, with mean values and a variability of mechanical properties equivalent to those manufactured on an industrial scale (the wood particles base), with the density ranging from 0.9 to 1.0 g.cm⁻³. The authors also obtained values of mechanical properties that allowed its use in commercial and industrial structural use.

Freire et al. (2012) compared the physical properties of commercial particleboards from China (from sugarcane bagasse) with commercial particleboards of eucalyptus and pine in Brazil, as well as the influence of these sealing panels with a layer of paraffin. The authors observed that the bagasse

particleboards had higher water absorption values, but smaller values of thickness swelling. For bagasse particleboards, it was observed that the samples with the side susceptible to water ingress showed higher thickness swelling after 24 hours. As for the WA and TS, after 2 hours, lower values were observed for samples with the exposed surfaces and sealed side. For the WA after 24 hours, the samples sealed at the sides and the surfaces were statistically equal.

Oliveira et al. (2014) studied the physical and mechanical properties of commercial particleboards made from Pine, *Eucalyptus*, and SCB. They concluded that particleboards made of SCB showed similar or better performance to those produced with *Eucalyptus* and Pine.

The study of thermal properties of lignocellulosic boards aims to employ them mainly in structures, ceilings, and partition walls, in various environments used by man. Carvalho (2012) investigating the thermal behavior of sugarcane bagasse particleboards ($0,543 \text{ g.cm}^{-3}$ of density), noted that sugarcane bagasse particleboards had low thermal conductivity, providing resistance to heat passage, thus sugarcane bagasse particleboards retained more heat when compared with pine and *Eucalyptus* panels. However, pine and *Eucalyptus* panels showed higher density ($0,645$ and $0,636 \text{ g.cm}^{-3}$ respectively).

2.5 Nanotechnology

Nanoscience and nanotechnology are multidisciplinary, with both approaching, understanding and control of material in the nanoscale. However, they also differ. Nanoscience focuses on understanding the phenomena, while nanotechnology focuses on the effective application of knowledge (DURAN; MATTOSO; MORAIS, 2006; KAMEL, 2007). Nanotechnology involves study at the nanometer scale ($1 \text{ nm} = 1 \times 10^{-9} \text{ m}$), and it has a huge variety of applications: increased resistance and durability of composites, low-cost production of corrosion-resistant steel, production of insulators, production of

films and coatings with self-cleaning effect, production of nanosensors with sensor capacity and self-healing, among others (HUANG; NOTTEN; RASTERS, 2011; KAMEL, 2007; ROCO; BAINBRIDGE, 2005; TORGAL; JALALI, 2010).

Nanotechnology is constantly expanding to more and more areas and so, in recent years, it is also used in the wood industry (CAI et al., 2007a, 2007b; KABOORANI et al., 2012) for wood boards (LEI et al., 2006; LIU; ZHU, 2014; MANTANIS; PAPADOPOULOS, 2010). Use of materials with at least one dimension smaller than 100 nm, which is the most common definition of a nanomaterial, can improve existing properties of materials and introduce completely new characteristics (NIKOLIC; LAWETHER; SANADI, 2015).

Adding hard and stiff nanosized fillers is well known to reinforce both thermoplastic and thermosetting polymers, which are commonly used as binders in wood adhesives. The nano size of these fillers is responsible for their huge surface area, which significantly increases the interface between polymer and filler. With an increase in the ratio of the interface in a composite, interactions that are present on that interface assume much more importance. If these interactions are high, then stress can be efficiently transferred from the polymer to stiffer and stronger nanofillers resulting in a more resilient adhesive with higher adhesive bond strength. Of course, if there are poor interactions with the polymers, voids will be formed between polymer and nanoparticles, stress transfer from the polymer to the nanoparticles would be poor, and improved adhesive response cannot be achieved even possibly causing a premature failure. Unfortunately, with this enormous surface area, the surface energy of these nano fillers is also high (KAMEL, 2007; NIKOLIC; LAWETHER; SANADI, 2015).

Owing to the high surface energy, there is a tendency of nanoparticles to agglomerate, thus reducing the high interfacial area on which improvement of properties is overall based. Agglomeration of nanomaterials is one of the biggest

reasons why nanotechnology has not yet manifested its true potential. Increasing the loading of nano fillers makes it more difficult to achieve good and uniform dispersion, and usually, the biggest improvements have been seen with the addition of low-weight percentage of nanomaterials (ATTA-OBENG et al., 2013; LEI et al., 2010; VEIGEL et al., 2012).

The shape of nanomaterials can be different, and materials that are spherical particles, platelets or fibers with high-aspect ratios can be used. High-aspect ratio is believed to be very influential in reinforcing mechanisms. Among the spherical particles, inorganic oxides like SiO_2 and Al_2O_3 are commonly used because of their high properties of hardness (6 and 9 respectively, on Mohs scale) and high stiffness, can be used in adhesive formulations. Platelets, such as nanoclays, and nanofibers such as cellulose, can also be used (FUFA et al., 2012; MARATHE; KANTAK, 2008; NIKOLIC; LAWATHER; SANADI, 2015). Some improvements in barriers properties have been found when using some layered phyllosilicate, which have a very high aspect ratio (10-1000). The main goal for the successful development of clay-based nanocomposites is to achieve complete exfoliation of the layered silicate in the matrix. The impermeable clay layers produce a tortuous pathway acting as a barrier between gasses and water, forcing them to follow a longer diffusion path. These phyllosilicates usually contain Na^+ or K^+ ions and are only compatible with hydrophilic polymers such as polyvinyl alcohol (PVOH). However, it is possible to modify their surface chemistry through ion exchange reactions using organic and inorganic cations, which can make them compatible with nonpolar polymers. However, this kind of modification is more important and common in plastic composites (CHOUDALAKIS; GOTSIS, 2009; SEE; ZHANG; RICHARDSON, 2009).

2.5.1 Nanocellulose and nanocrystals

The lignocellulosic materials are of the principal sources of cellulose, and they are composed basically of cellulose, hemicellulose, lignin and minor constituents, but the cellulose is the main constituent of the fiber cell wall (ALMEIDA, 1988; MORÁN et al., 2008). The cellulose molecules tend to form hydrogen bonds, intramolecular (between glucose units in the same molecule), and intermolecular (between adjacent molecules of glucose units). The first type of interaction is responsible for a certain rigidity of the chains, and the second unit is responsible for the formation of plant fibers, so the cellulose molecules align to form microfibrils, which form fibrils in turn, and are ordered to form successive cell walls (ALMEIDA, 1988). In general, in plant cell walls, cellulose is in the form of fine fibrils, which have dimensions on a nanometer scale. The diameter of these structures varies from 3 to 5 nm, whereas the length is more than 1 μm . The microfibrils are composed of single crystal regions, most accounting for 60 to 80% linked by amorphous regions that occur in less amounts in plant cell walls, and microfibrils aggregate into larger structures, called macrofibrils. This structure is deconstructed to generate vegetable nanofibers (EICHHORN et al., 2010; IOELOVICH, 2008). The nanostructures from lignocellulosic materials can be obtained by chemical and mechanical processes. The chemical processes result in a purely crystalline structure called whiskers, cellulose nanocrystals or nanocrystalline cellulose (NCC) (FILSON; BENHAMIN; SCHWEGLES, 2009; KABOORANI et al., 2012; TEIXEIRA et al., 2011; ZHANG et al., 2011). The mechanical processes result in crystalline and amorphous regions, called cellulose nanofibers, cellulose nanofibrils, nanocellulose (CNF) or micro/nanofibrils, and microfibrillated cellulose (MFC) (CAMPOS et al., 2013; TONOLI et al., 2012; VEIGEL et al., 2011, 2012). There has also been some research on a microscale, with microcrystalline cellulose (MCC) (ATTA-OBENG; VIA; FASINA, 2012).

2.5.1.1 Studies using nanocellulose and nanocrystals in wood boards

Zhang et al. (2011) investigated the effect of inclusion of modified NCC in the UF adhesive. The original NCC showed a length of 300-400 nm, and a diameter of 30-50 nm. The NCC was modified with 3-aminopropyltriethoxysilane (APTES) and 3-methacryloxypropyltrimethoxysilane (MPS) to improve the compatibility with the UF. The NCC was dispersed into UF at concentrations of 0.5, 1, 1.5 and 2% using an ultrasonic probe. That mixture was later used to produce plywood. The emission of HCHO from UF with modified NCC was reduced by physical absorption and chemisorption without compromising the internal bond strength of plywood. High ratio surface of the modified NCC was the main factor of physical adsorption, which led to the decrease in HCHO. The best result was found for NCC APTES (1.5%), with a reduction from 0.47 mg/L to 0.22 mg/L. For NCC MPS, the best result was for 2% of NCC, and the emission was reduced from 0.47 mg/L to 0.37 mg/L. The bonding strength increased when modified NCC was added, and the highest values were found for 1.0 % of NCC MPS (from 0.72 to 0.77 MPa), and 1.5% of NCC APTES (from 0.72 to 0.89 MPa). The bonding strength decreased for 2% of modified NCC, because of the NCC agglomeration.

Atta-Obeng, Via and Fasina (2012) investigated the effect of the particle size (small and big), specie (sweetgum and pine), and inclusion of MCC (0 and 10%) in particleboards using PF. The inclusion of MCC to particleboard resulted in a decline in board mechanical properties and higher thickness swell, so the MCC effect was negative. According to the authors, inclusion of MCC provides more available hydroxyl groups for moisture sorption, and consequent thickness swelling. The authors did not explain why the mechanical properties decreased with MCC, but probably it is because it used a high amount of MCC, which could agglomerate, react more with the PF, and harm the dispersion that later affected the properties of the particleboards.

Veigel et al. (2012) produced cellulose nanofibrils CNF by mechanical fibrillation, and showed average diameter of 35 nm. The CNF was mixed with UF in different reinforcement proportions (0, 1 and 3%), and used to produce particleboards. The viscosity of the adhesives increased with the insertion of CNF, and because of this, it was not possible to use much of CNF as reinforcement. The mechanical performance of the boards (with 1% CNF) was enhanced, whereas, with 3% of CNF it was worse than the control. The author suggested that this deterioration was not directly induced by higher CNF content, but could be explained by the long drying time of the glue wood particles prior to hot pressing. In the same paper, the authors tested the CNF mixed with MUF (at 1% loading level) and made OSB with this adhesive. The viscosity of the adhesives increased with the insertion of CNF, and because of this, it was not possible to use a lot of CNF as reinforcement. The physical mechanical performance of the boards was enhanced, but the TS was above recommended levels.

Mahrtdt et al. (2015) investigated the effects of MFC addition (5%) in UF and its application in particleboards. They observed that the MFC addition increased the viscosity and the cure time of the adhesive UF. Particleboards with MFC inclusion showed higher values (30% more) for IB compared with particleboards without MFC. The authors attributed this better performance to the distribution of the adhesive, since a higher fraction of adhesive + MFC was available for bond-line formation, and a larger part of the wooden particles were covered with the adhesive.

It was observed in this literature review that there are challenges in this field to be investigated and understood, with new possibilities applying to particleboards.

2.6 General considerations

Every day new products are developed, incorporating new properties. Most of the products that do not follow new technologies lose their attractiveness and are replaced with better products. Wood boards/lignocellulosic boards follow the same pattern. Some new boards replace old boards using a new technology or material. Thus, incorporating nanotechnology can help develop boards with better properties and even new properties. Another factor is the worry with regard to the environment; using green materials has become a trend in the world. In this context the use of lignocellulosic wastes/materials (sugarcane bagasse, sisal, and coir) can incorporate value in the particleboard. It can also decrease the pressure on natural forests, as they are an alternative to wood.

3 CONCLUSION

The inclusion of coir filaments (until 20%) proved feasible for MDP production, since most of the properties (MOE, MOR, TS and WA) achieved similar performance of MDPs from *Eucalyptus urophylla*. However, the sisal inclusion, using 3 cm of length and a rotary blender, did not showed feasible, since it resulted deterioration in all properties analyzed. Sisal filaments tend to clump together in the rotary blender and it was difficult for UF to get in between the filaments clumps.

NCC inclusion in UF increased the viscosity of the liquid suspension and a better mechanical performance (MOE and MOR) of solid UF+NCC specimens were found, with 1, 2, and 3% of NCC. In SCB particleboards (one layer) improvement was not observed with NCC inclusion (for MOE, MOR and TS). Probably, because less site links were available for SCB to interact with the adhesive. The WA24H slight decreased with 1 and 2% of NCC inclusion.

The NCC inclusion (1%) in the faces of the particleboards (MDP) did not showed efficient, since for most properties the particleboards with and without NCC were statistically equal. Probably less site links were available for the SCB to interact with the adhesive and because the NCCs increased the adhesive viscosity, this could make adhesive penetration more difficult.

It was found that the separation in pith and fibers does not seem efficient and necessary, since the particleboards made only with fibers and with pith/fibers showed properties statistically equal (MOE, MOR, WA, TC and VHC). However, particleboards made with pith/fibers (without NCC) showed lower TS24h, probably because of the lower pith cellulose content and higher lignin content. The two kinds of material showed very similar anatomical properties. The sugarcane bagasse particles has a potential and can be used to produce particleboards for internal purposes.

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SECOND PART – MANUSCRIPTS

MANUSCRIPT 1 – COIR AND SISAL FIBERS AS FILLERS IN THE PRODUCTION OF EUCALYPTUS MEDIUM DENSITY PARTICLEBOARDS - MDP

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ABSTRACT: The aim of this study was to evaluate the potential use of sisal and coir fibers in combination with *Eucalyptus* particles for the production of particleboard. The particleboards were produced in three layers. The first and third layers (face) were made with small *Eucalyptus* particles. The second layer (core) was made with big *Eucalyptus* particles in combination with coir or sisal fibers. The particleboards were prepared with the substitution on *Eucalyptus* wood for sisal and coir fibers in the particleboards core, in doses of 0, 10, and 20%, relative to the total mass of particles. The particleboards were characterized by mechanical, physical and thermal properties. The results were not satisfactory for particleboards with sisal. However, for coir particleboards the physical-mechanical properties were very similar to those particleboards produced only with *Eucalyptus*. This work demonstrates the potential use of the coir that is commonly disposed in landfills on the Brazilian beaches.

Keywords: Coconut; lignocellulosic fiber; chipboards; thermal properties; panels

1 INTRODUCTION

Wood boards (or wood panels) can be defined as compounds of elements of the breakdown products of wood, such as plates, laths, particles and fibers, which are then surface coated with an adhesive to produce the boards. These new products are also known as reconstituted wood products and their properties are different from the original wood material ¹⁻³.

There are different kinds of wood boards/particleboards, among them the Medium density particleboard (MDP) and chipboards. The chipboards are composed of wood particles called slivers and form a single layer ¹⁻³. The MDPs are an evolution of the chipboards, by having three layers, two layers with

smaller particles in the faces and one layer with bigger particles in the core of the board. They are usually produced with wood particles, and they are mainly used in the furniture industry. The raw material typically used in Brazil is wood from planted forests of *Pinus* and *Eucalyptus*⁴. However, any lignocellulosic material can be used for the manufacture of particleboards, since they have adequate quality⁵⁻¹¹. It is also possible to mix different types of wood to other materials when the lignocellulosic materials do not show adequate quality by themselves, such as: vine pruning residues¹²; poppy husk¹³; coir⁵; coffee stem¹⁴; maize cob¹⁵; coffee husk¹⁶; rice hulls¹⁷ and sugarcane bagasse¹¹. The use of natural fibers as reinforcement has aroused great interest in developing countries because of their low cost, availability, energy saving, renewable source, non-toxic and also with regard to environmental issues¹⁸⁻²⁰.

Among the materials that have potential are sisal fibers, which are extracted from the leaves of the plant, *Agave sisalana* Perr. Sisal has interesting characteristics for reinforcement, both in composites and in boards due to their good mechanical properties, low density and high availability^{21,22}. Brazil is the largest producer of sisal fibers in the world 120,000 t per year with yields of around 800-1200 kg/ha, and accounts for 58% of the world production. In the industry, sisal is transformed into several varieties of yarns, cords, mats and blankets, being used for making various products, such as in reinforcing plastic composites, where researchers have shown promising results^{21, 23-25}. Another material with potential uses are the fibers from the husk of coconuts and these fibers are commonly known as coir, which after processing, produces long and short fibers. The Brazil wastes most of its coconuts husks that coir fibers while countries like India and Sri Lanka are selling their fibers for multiple uses and generate money for the local economy. It is estimated that the waste amounts to more than US\$ 60 million per year in Brazil²¹.

New uses of these materials can result in social, economic and environmental development for the country. Sisal cultivation is concentrated in the semiarid region of northeastern Brazil, which has few economic alternatives. Small farmers produce most of the sisal with a predominance of family agriculture. New uses for coir can transform a waste into useful products²¹. The objective of this study was to evaluate the potential of using sisal and coconut fibers in combination with *Eucalyptus* particles for the production of MDP.

2 MATERIAL AND METHODS

2.1 Raw material

Eucalyptus urophylla particles, sisal fibers (supplied by Embrapa Cotton) and coconut fibers (coir) were used along with urea formaldehyde (UF) as adhesive. The *Eucalyptus urophylla* wood was 7 years old with a basic density of 0.560 g.cm⁻³ and bulk density of 0.586 g.cm⁻³ (11.8% moisture). This wood has grown in Paracatu-MG-Brazil and obtained from Companhia Mineira de Metais.

The sliver particles were generated with a hammer mill and subsequently classified in sieves of 14, 40 and 120 mesh. The particles retained in the sieves of 120 and 40 mesh were used for the faces and core, respectively.

Asasutjarit et al.²⁶ found that the best performance of cement/coir boards were those with coir fiber lengths between 1 to 6 cm. According to those researchers, long fibers decrease the workability and increase the void space. Besides this, the longer the fiber length both water absorption and thickness swelling was higher. Based on this the lignocellulosic fibers were cut manually with the aid of scissors to approximately 3 cm lengths.

The particles and fibers were dried in an oven at 90°C until final

moisture content of 4%.

The adhesive (UF) used had the following characteristics- pH = 9.15, solids content = 53% and viscosity = 0.261 Pa.s.

2.2 MDP production

The adhesive (UF) was spread using a rotary blender in two stages. First, the adhesive was applied to smaller particles in a proportion of 11% (dry basis weight of the particles). The bigger particles (plus fibers) were coated with the adhesive in a proportion of 8% (dry basis weight of the particles). More adhesive was used for the smaller particles because they have higher surface area to volume coated to larger particles and therefore need more adhesive to guarantee good adhesion. Three particleboards were produced for each treatment with a nominal density of 0.750 g.cm^{-3} in three layers, face/core/face, with the proportion of 20/60/20 (basis of particles weight), respectively. Three sisal/coir proportion were used (0, 10 and 20%) and were mixed with the *Eucalyptus* particles in the core (Table 1). As mentioned earlier the only smaller wood particles of 120 mesh were used as surface layers, with no coir or sisal present.

Table 1 Treatments with their composition

Treatments	Face (20% each)	Core (60%)
EU	<i>E. urophylla</i>	100% <i>E. urophylla</i>
CO-10	<i>E. urophylla</i>	90% <i>E. urophylla</i> + 10% of coir
CO-20	<i>E. urophylla</i>	80% <i>E. urophylla</i> + 20% of coir
SI-10	<i>E. urophylla</i>	90% <i>E. urophylla</i> + 10% of sisal
SI20	<i>E. urophylla</i>	80% <i>E. urophylla</i> + 20% of sisal

The particle mattress was subjected to a pre-pressing in a hydraulic press

at a pressure of 0.78 MPa and subsequently transferred to a hot press. The hot press cycle underwent a pressure of 3.92 MPa at 160°C for 8 minutes. After the pressing and subsequent cooling, the edges of the four sides of the boards were cut off, resulting in final dimensions of 47x47x1.5 cm. The boards were then placed in a controlled chamber for ($22\pm 2^\circ\text{C}$ and $65\pm 5\%$ of moisture) for acclimatization until constant weight was achieved prior to physical and mechanical characterization. The production steps are summarized below (Figure 1).



Figure 1 MDP production. (a) Adhesive application to smaller particles. (b) Adhesive application to bigger particles plus fibers. (c) Particle mattress after the pre-pressing. (d) Particle mattress during the hot press

2.3 Physical, mechanical and thermal properties evaluated

The particleboards were characterized by testing their physical, thermal and mechanical tests using standardized tests specified in Table 2.

Table 2 Properties evaluated

Properties	Symbol	Standard/Equipment
Density	D	EN 323 ²⁷
Compression ratio	CR	MALONEY ²⁸
Water absorption after 2h	WA2h	ASTM D1037 ²⁹
Water absorption after 24h	WA24h	ASTM D1037 ²⁹
Thickness swelling after 2h	TS2h	ASTM D1037 ²⁹
Thickness swelling after 24h	TS24h	ASTM D1037 ²⁹
Thickness non-return ratio	TNRR	ASTM D1037 ²⁹
Bending modulus	MOE	DIN 52362 ³⁰
Bending strength	MOR	DIN 52362 ³⁰
Internal bond	IB	ASTM D1037 ²⁹
Thermal conductivity	TC	ISOMET

The compression ratio was calculated by dividing the density of the particleboard by the density of the material. The dimensions of the particleboards and wood, for the calculation of the volume, were measured with a caliper, but the sisal and coconut dimensions have been calculated using a microscope.

The thermal properties were measured with the equipment ISOMET model 2104. The samples with 6x6x1.5 cm were stored in a controlled chamber (20°C and 65% of moisture) until constant weight before being tested. Five samples were measured for each set of samples.

The data were analyzed as a completely randomized design and the properties were compared by the mean values and standard deviation.

3 RESULTS AND DISCUSSION

3.1 Density, moisture and compression ratio

The density and moisture of the boards are graders characteristics that ensure fairness between treatments, thus allowing to proceed with the comparison between the average values of physical and mechanical properties. The density of the MDPs ranged from 0.757 to 0.776 g.cm⁻³ (Table 3). All the boards were classified as medium density, referring to boards with density between 0.590 to 0.800 g.cm⁻³¹. The moisture ranged from 9.43 to 9.80%. Both those properties showed a slight variation of the mean values.

The CR is the relation between the density of the boards by the density of the material. As noted in Table 3, the CR values increased as it included sisal/coir fibers and this is due to the lower density. The densities of the material were measured to be; sisal 0.368 g.cm⁻³ (12.75% of moisture); coir 0.460 g.cm⁻³ (12.15% of moisture); *Eucalyptus* 0.586 g.cm⁻³ (11.8% of moisture). According to Maloney²⁸ the ideal CR values are between 1.3 and 1.6. The recommendation of the author to the mentioned CR range is partly because of CR lower than 1.3 result in lower mechanical strength and high values of CR more than 1.6 creates problems as dimensional instability of the boards. All the MDPs in this study had a CR in the recommended range between 1.3 to 1.6.

Table 3 Values of the particleboards density, humidity and compression ratio

Treatments	Density (g.cm⁻³)	Moisture (%)	Compression ratio
EU	0.757±0.007	9.74±0.07	1.3±0.0
CO-10	0.760±0.026	9.63±0.19	1.4±0.1
CO-20	0.773±0.010	9.80±0.07	1.4±0.0
SI-10	0.776±0.010	9.53±0.05	1.4±0.0
SI-20	0.763±0.007	9.47±0.10	1.4±0.0

Mean values followed by the standard deviation. Density (Bulk density)

3.2 Bending (MOE and MOR)

The mean values of MOE and MOR for the different treatments are shown in Figures 2 and 3, respectively. There was very little difference in the MOE between the samples CO-20, EU and CO-10. This was also true for MOR for EU, CO-10 and CO-20. The coir particleboards values are below those found for Khedari et al.³¹ However, the authors produced particleboards only with coir, which resulted in an MOR of 25 MPa and MOE of 2857 MPa. This probably because using a single material of only coir fibers would probably have better dispersion than in the present case where fibers are mixed with particles. Secondly, longer fibers can result in improved reinforcement instead of a mix of particle and fibers.

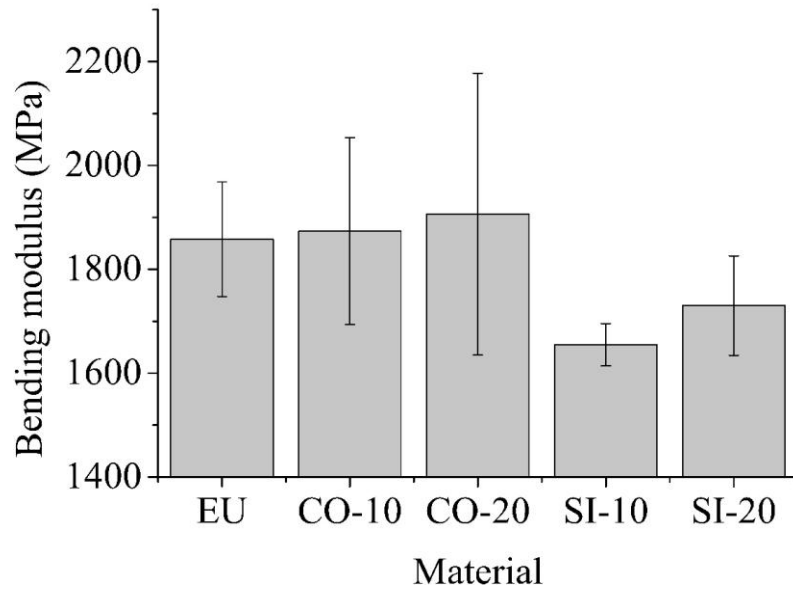


Figure 2 Histogram of the mean values for bending modulus (MOE). Bars indicate standard deviation

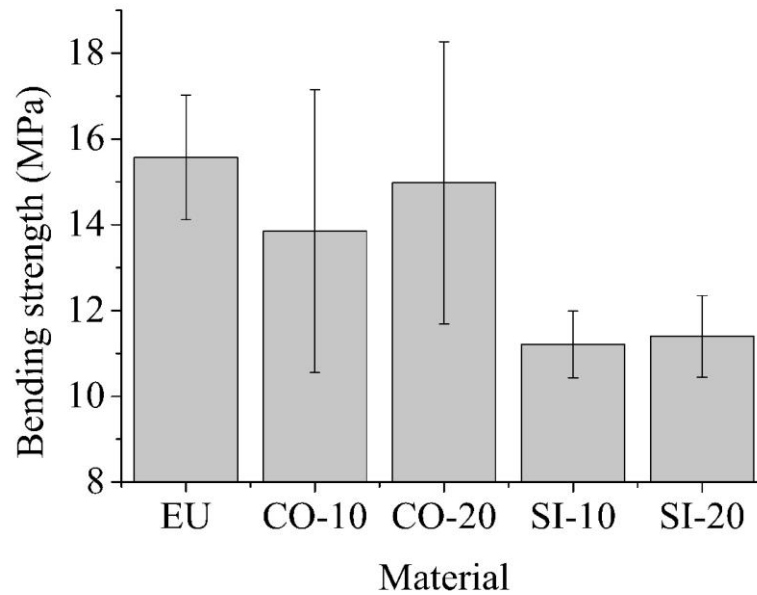


Figure 3 Histogram of the mean values for bending strength (MOR). Bars indicate standard deviation

The MDPs with higher amount of sisal and coir (20%) showed higher standard deviation than the MDPs with 10% of sisal and coir. This may be due to factors related to the processing, since the distribution of the particles and fibers were performed manually, making it more difficult to evenly disperse the two materials when the amount of fibers is increased. During the adhesive application, it was observed that sisal fibers tend to clump together in the rotary blender and it is thus difficult for the UF to get in between the fiber clumps and this is reflecting by lower MOE and MOR of the boards.

Coconut coir has low amounts of hemicellulose and a high amount of lignin 41-45%³²⁻³⁴. Lignin has the ability to act as an adhesive and increases the strength of the fiber bond but this depends on the type of lignin and amount of moisture and temperature of processing³⁵⁻³⁷. Particleboards that have a high

lignin content will be stronger as long as sufficient moisture and temperature is used. High amount of coir improves the cross-links between fibers and decreases space and void ³². Besides that, sisal is a hard fiber and it is more difficult to press than coir, which increase the amount of void, impairing the mechanical properties (MOR, MOE and IB).

Colli et al. ⁵ produced particleboard from Paricá wood (*Schyzolobium amazonicum*) with coir (0, 10, 20 and 30%). For MOE, the coir inclusion did not changed the mean values, however the mean values for MOR were higher for boards with high amount of coir (20 and 30%). It is possible that better dispersion of fibers resulted in the fibers reinforced the boards more efficiently and this effect is greater with higher amounts of fibers.

Melo et al. ¹⁷, investigated the association of rice hulls (0, 20, 40, 60, 80 and 100%) with wood (*Eucalyptus grandis*) in chipboards with 0.650 g.cm^{-3} of nominal density. The authors observed a decrease in the mechanical property values with an increase in the percentage of rice husks- here MOE decreased from 1225 to 196 MPa and MOR from 14.7 to 3.9 MPa. César et al. ¹⁴ and Mendes et al. ¹⁶ observed the same trend in with the use of coffee stem and coffee husk in chipboards. This may be due to waxy surface layers on the husks, which could create a weak boundary layer ^{38,39}, although this aspect was not studied by the authors.

ANSI A208.1 ⁴⁰ stipulates the average minimum values of 1764 MPa for MOE and 11.3 for MOR for particleboards/chipboards made with urea-formaldehyde (class M1). In this study, none of the MDPs made with sisal fibers met the minimum requirements for MOE, however, for MOR all sisal MDPs attained the average minimum value required by the standard. In the case of coir/wood boards both MOE and MOR exceeded the ANSI standards noted above.

3.3 Internal bond strength (IB)

The mean IB values of all the boards in this study are shown in Figure 4. The data demonstrate that the IB decreased with an increase of the sisal and coir amount, although the decrease was more pronounced for sisal MDPs. The decrease in IB values can be attributed to the sizing (dispersion of the UF adhesive) process of particles and fibers, because the clumps of fibers may have impaired the adhesive dispersion process and the uniform distribution of the wood particles and fibers. This was especially visible in sisal MDPs, which after sizing, clumps of fibers were observed. As mentioned previously, these fiber clumps prevented the dispersion of the adhesive in the middle of the clumps, thereby impairing the bonding of the materials, since some regions may have had no adhesive. The application of the adhesives is directly related to the quality of gluing the boards and the IB test^{41, 2}. The reduction in the length of the sisal fibers in future work can facilitate and improve the mixing of particles and fibers, which in turn can improve the dispersion of the adhesive and bonding.

This decrease in IB was also observed while using particles of corncob by Scatolino et al.¹⁵ (0, 25 50, 75, 100%) mixed with wood (*Pinus oocarpa*) in chipboards. The mean values decreased from 1.1 to 0.3 MPa with the corn cob inclusion. The authors attributed the results to the low density of corncob, because the increase in the number of particles entailed a reduced availability of adhesive per particle, which reduce the adhesion strength and therefore the IB strength. Materials that have lower density generate a larger amount of particles for a given weight and thus a higher specific surface requires higher amounts of adhesive to obtain good properties⁴¹. The same decrease was observed for Mendes et al.¹⁶ with the inclusion of coffee husk in chipboards made from *Eucalyptus urophylla* and Mesquita et al.⁶ with inclusion of sisal.

The minimum IB required by ANSI A208.1⁴⁰ is 0.40 MPa for UF adhesive (M1 class). Only the MDPs with 20% of sisal incorporation did not meet the minimum requirements.

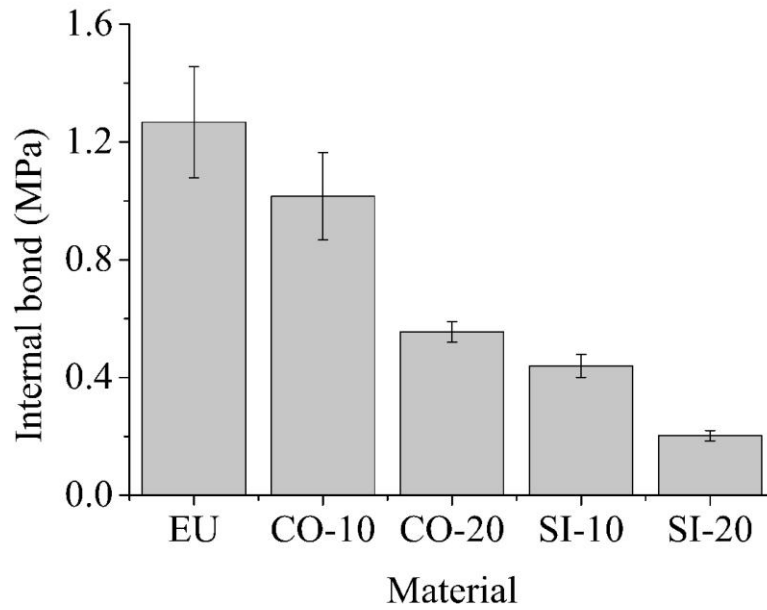


Figure 4 Histogram of the mean values for the internal bond property (IB). Bars indicate standard deviation

3.4 Thickness swelling (TS) and water absorption (WA)

The mean values for TS properties, TNRR and WA for all treatments, are presented below (Figures 5 and 6).

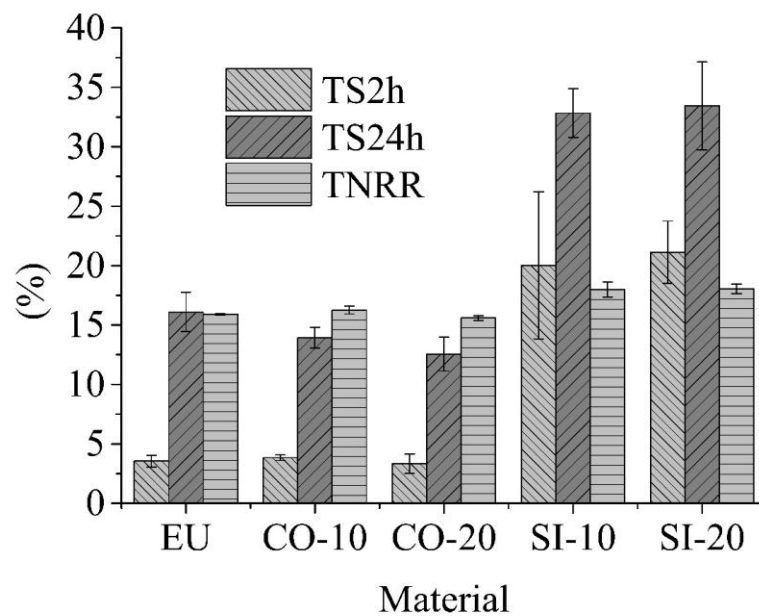


Figure 5 Histogram of the mean values for the thickness swelling after 2 hours (TS2h), thickness swelling after 24 hours (TS24h) and thickness non-return ratio (TNRR). Bars indicate standard deviation

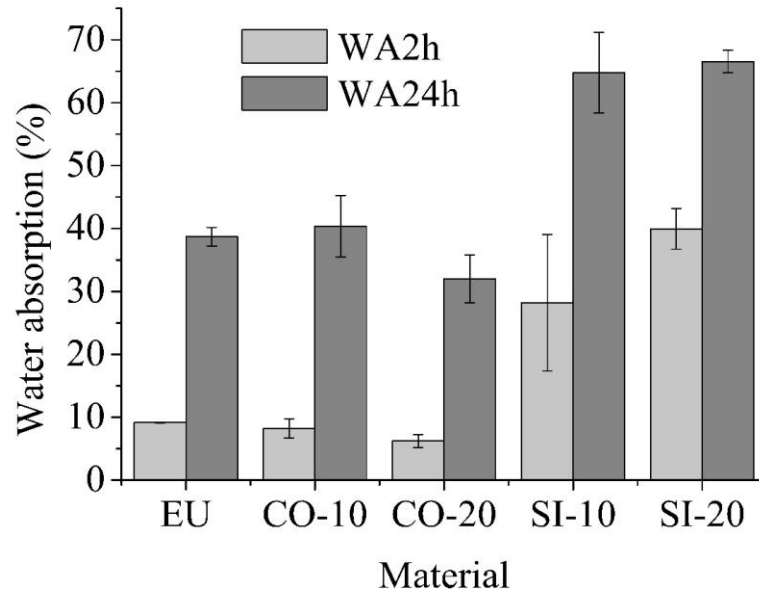


Figure 6 Histogram of the mean values for the water absorption after 2 (WA2h) and water absorption after 24 hours (WA24h). Bars indicate standard deviation

The MDPs reinforced with coir fibers showed satisfactory results for TS and WA after 2 and 24 hours. The mean values for CO-10 and CO-20 were very similar to those made just with particle wood (EU). However, the addition of sisal resulted in a significant decrease of these properties and this is probably due to higher amount of voids because of poor particle and fiber dispersion along with fiber clumping and poor bonding that facilitated the entry of water. MDPs with sisal showed almost the double values of TS and WA after 24 hours, compared to EU, CO-10 and CO-20. Similar results had been observed by César et al.¹⁴ working with lignocellulosic waste and the authors attributed this to bonding problems caused by possible chemical incompatibility or poor distribution between the adhesive and particles. The same trends were observed for Mendes et al.¹⁶ with the inclusion of coffee husk in chipboards. Coconut coir

has low amounts of hemicellulose and a high amount of lignin 41-45%³²⁻³⁴. Lignin has the ability to act as an adhesive and increases the strength of the fiber bond but this depends on the type of lignin and amount of moisture in the material³⁵⁻³⁷. Particleboards that have high lignin content will be rather stronger with a higher water resistance. Higher amount of coir improves the cross-links between fibers and decreases space and void³². Sisal is a hard fiber and it is more difficult to press than coir, which increase the amount of void, increasing the WA.

Keskin et al.¹³ found similar results for TS after 2 and 24 hours using wood pine plus poppy husk up to 25%. The treatment with 25% of poppy husk was statistically equal to those made only with wood pine. Using higher amount of poppy husk (50, 75 and 100%) increased the TS and decreased the IB, MOE and MOR.

The UF adhesive have low water and weather resistance. The aminomethylene linkage is susceptible to hydrolysis, which means that the reaction that forms the UF is reversible when it is attack by water and this is also responsible for the formaldehyde emission. The water causes swelling and therefore movement of the structural components of the MDP contributing to break the bonding between resin and wood surface sites due to mechanical forces and stresses⁴²⁻⁴⁴.

Colli et al.⁵ produced particleboard from Paricá wood (*Schyzolobium amazonicum*) with coir inclusion (0, 10, 20 and 30%) and they did not find difference between the mean values for WA and TS. Khedari et al.³¹ produced particleboards with only coir and the boards had a 53% for TS and 78% for WA. However, those authors produced particleboards with a lower density (0,648 g.cm⁻³), which contribute to increased voids thus increasing the WA and TS values.

Scatolino et al. ¹⁵ observed the same for WA when they increased the amount of corn cob in chipboards, but for TS24h the mean values decreased. The authors attributed this to the increase of compression ratio and the largest amount of extractive corn cob. Apart from WA and TS, extractives such as surface waxes can result in a weak boundary layer ³⁸ and this will play a role in MOR and IB strengths such as the work on wheat straw ³⁹, as discussed earlier.

According Hillig et al. ⁴⁵ and Malloney ²⁸ the physical-mechanical properties of the boards are influenced by the CR and low CR values do not produce good contact between the particles, impairing the bonding and strength of the boards. In addition, the boards have higher spaces/voids, which facilitate the water absorption. However in the current study the CR is within the range recommended (1.3 to 1.6) suggesting that poor adhesive dispersion for sisal fibers and/or possible chemical incompatibility between adhesive and material due to extractives or otherwise.

The ANSI A208.1 ⁴⁰ provides values for only the TS24h that is 8% for the chipboards produced with urea-formaldehyde. Therefore, none of the MDPs produced met the minimum requirement stipulated by the standard.

3.5 Thermal properties

3.5.1 Thermal conductivity

The samples for thermal properties were stored in a controlled chamber with 20°C and 65% of moisture until constant weight. The samples had the density and moisture measured before the test (Table 4).

The thermal conductivity was directly related to the density of the MDPs (Table 4 and Figure 7) and the conductivity decreases with density. This can be

explained by the change in the number and/or volume of voids both between and inside particles owing to their densification. These voids are occupied by air, and since air has a lower thermal conductivity than the wood⁴⁶. The relationships between density and thermal conductivity were also observed by Bekhta and Dobrowolska⁴⁶ when studying the properties of wood-gypsum boards and also by Sampathrajan et al.⁴⁷ measuring properties of boards made from farm residues.

Some researchers try to produce materials with low thermal conductivity, so as to reduce heat transfer and decrease the energy consumption of the building facilities. Thermal conductivity is an indicator of the value of a material as a heat insulator and lower thermal conductivity indicates that the material is better for thermal insulation. This is interesting for energy saving when used for ceiling and wall insulating material/boards^{32, 48}. These same authors found values for particles boards (0.258 g.cm⁻³) about 0.029 W m⁻¹ K⁻¹, and they recommended those boards for thermal insulation.

Khedari et al.³¹ produced coir particleboards just with coir and found mean values of 0.1445 Wm⁻¹K⁻¹ (0.648 g.cm⁻³ of board density). Lower values (0.1117 Wm⁻¹k⁻¹) were found for particleboards (0.611 g.cm⁻³) made with 90% of coir and 10% of durian³².

Yapici et al.⁴⁹ founded that the thermal conductivity were affected by adhesive ratio and pressing time for oriented strand boards (OSB). It was observed that increasing the amount of adhesive and pressing time increases the thermal conductivity. The thermal conductivity values, found by Yapici et al.⁴⁹, range from 0.129 to 0.170 Wm⁻¹k⁻¹. This is obvious since higher the amount of adhesive and higher pressure reduces voids and increases conductivity.

Table 4 Apparent density and moisture of the samples used for the thermal properties

	EU	CO-10	CO-20	EU-10	EU-20
Density ($\text{g}\cdot\text{cm}^{-3}$)	0.774 \pm 0.081	0.727 \pm 0.026	0.713 \pm 0.053	0.702 \pm 0.068	0.774 \pm 0.056
Moisture (%)	9.37 \pm 0.09	9.28 \pm 0.08	9.33 \pm 0.13	9.29 \pm 0.07	9.09 \pm 0.05

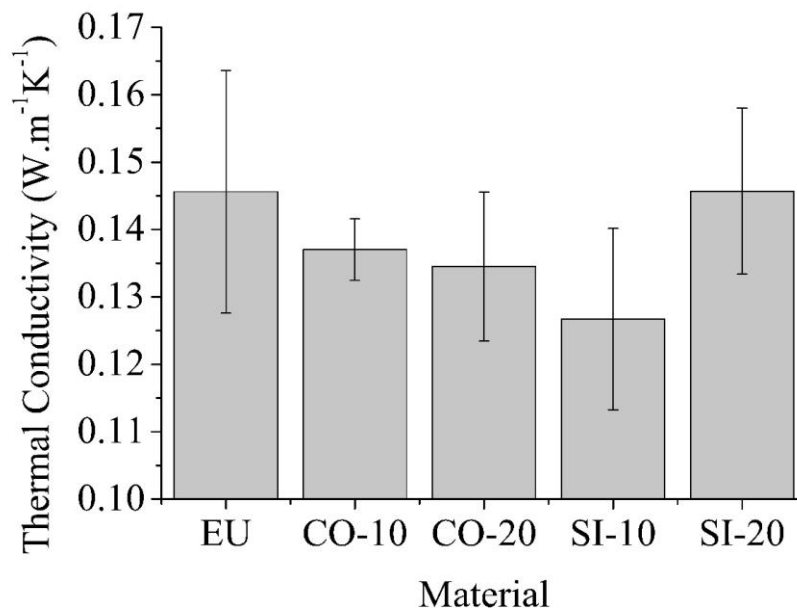


Figure 7 Histogram of the mean values of thermal conductivity. Bars indicate standard deviation

4 CONCLUSIONS

In general, the inclusion of sisal fibers in the MDP production was unfeasible, since it resulted deterioration in all properties analyzed. The inclusion of coconut fibers proved feasible, since for most of the properties the MDPs with coconut achieved the same performance of MDPs from *Eucalyptus*

urophylla. The use of this material contributes to the development of more sustainable materials, since it is a waste, which often is rather simply discarded on beaches.

Further studies need to be done in order to study ways to improve the homogenization of the material and the adhesive application. It should also verify the economic viability of the use of this waste, once the technical feasibility has already been demonstrated in this work.

ACKNOWLEDGMENT

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APPENDIX A -SUPPLEMENTARY MATERIAL TO CHAPTER 1. NOT INCLUDED IN THE MANUSCRIPT.



Figure A 1 F Particleboard production. A – Adhesive application. B – Particle mattress formation. C - Particle mattress after pre pressing. D – Hot pressing. E – Particleboard with 20% of coir. F – Particleboard with 20% of sisal.

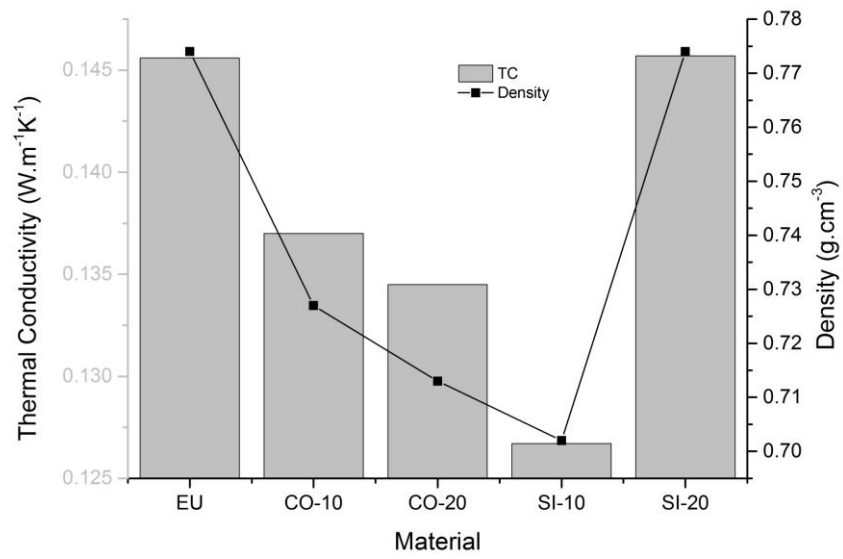


Figure A 2 Histogram of the mean values of thermal conductivity versus density.

Table A 1 Mean values of the properties evaluated.

Properties	EU	CO-10	CO-20	SI-10	SI-20
Density 1	0,76 a	0,76 a	0,76 a	0,78 a	0,76 a
Moisture	9,75 a	9,64 a	9,80 a	9,54 a	9,47 a
CR	1,3 a	1,4 b	1,4 c	1,4 b	1,4 c
MOE	1850 a	1854 a	2030 a	1632 a	1735 a
MOR	15,4 b	15,2 b	15,9 b	11,4 a	11,4 a
IB	1,18 d	0,97 c	0,57 b	0,46 b	0,21 a
TS2h	3,3 a	3,9 a	3,8 a	17,4 b	21,5 b
TS24h	15,4 a	14,2 a	13,4 a	32,2 b	33,2 b
TNRR	15,9 a	16,2 a	15,7 a	17,9 b	18,3 b
WA2h	9,1 a	8,3 a	6,8 a	24,8 b	40,9 b
WA24h	39,2 a	38,7 a	34,2 a	63,7 b	66,7 b
TC	0,15 a	0,14 a	0,13 a	0,13 a	0,15 a
Density 2	0,65 a	0,75 a	0,65 a	0,73 a	0,85 a

Mean values with an average test after ANOVA. Same letters in the line indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

**MANUSCRIPT 2 - UREA FORMALDEHYDE AND CELLULOSE
NANOCRYSTALS ADHESIVE: STUDIES APPLIED TO SUGARCANE
BAGASSE PARTICLEBOARDS**

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ABSTRACT: The aim of this research was to investigate the behavior of urea formaldehyde (UF) adhesive based particleboards using sugarcane bagasse (SCB), *Saccharum officinarum*, with and without the addition of cellulose nanocrystals (NCC). In the first step, the interaction between UF and the NCC was evaluated. The NCC were dispersed in the UF using a sonicator, in different proportions (0, 0.5, 1, 2, 3 and 5%). Subsequent to this, the mixture was dried, milled, and put in molds, to produce flexural specimens. The second step involved evaluation of the interaction among the, NCC, UF, and sugarcane bagasse (SCB). After sonication, the NCC and UF were applied on the SCB particles. They were compressed to produce particleboards. The particleboards were evaluated according to the bending modulus (MOE), bending strength (MOR), water absorption (WA), and thickness swelling (TS). The viscosity of the mixture of NCC and UF increased according to the NCC increase load. At 5% of NCC load, the viscosity increased greatly, which prevented adhesive dispersion in the particleboard. The thermogravimetric analysis of UF showed that better conditions of processing were at 160°C for 8 minutes, thus these parameters were used to produce the specimens. SCB is a promising raw material for particleboard, and it was demonstrated the possibility to produce SCB particleboards with relatively good performance. For the SCB/UF/NCC specimens, the best performance was observed with 1% of NCC. SCB/UF/NCC particleboards did not showed improvement compared with UF/NCC specimens, probably because bond links were not enough, indicating that more studies must be necessary to understand dispersion, viscosity and reactivity of studied system. Addition of NCC in UF improved properties of UF, showing a viscosity increase in UF/NCC liquid suspension, and a better mechanical performance of UF/NCC solid specimens.

Keywords: Natural fiber, amino resin, chipboards, thermosetting, nanotechnology

1 INTRODUCTION

Wood boards or composites can be defined as compounds of elements of the breakdown products of wood, such as plates, laths, particles and fibers, which are later combined with adhesive bonding under a certain pressure and temperature (Iwakiri, 2005; Kelly, 1977; Maloney, 1996). There are different kinds of wood boards and particleboards, among them the chipboard. Chipboards are composed of particles known as slivers and form a single layer. They are usually produced with wood particles and are mainly used in the furniture industry. The raw material typically used in Brazil, are *Pinus* and *Eucalyptus* (de Barros Filho et al., 2011). However, any lignocellulosic material can be used for the manufacture of particleboards (Carvajal et al., 1996; da Silva César et al., 2014; Khedari et al., 2004; Kwon et al., 2013; Widyorini et al., 2005).

With an increase in the utilization of wood products, the availability of wood decreases and its cost increases, thereby motivating a search for materials with similar characteristics, but with higher availability and lower cost. In this context, new lignocellulosic materials have been evaluated for the production of particleboards. Among these, many materials have emerged, especially from agricultural waste (Carvajal et al., 1996; da Silva César et al., 2014; dos Santos et al., 2014; Kwon et al., 2013). Studies have shown that residues from sugarcane can be easily formed into particles/fibers, similar to wood. These researches also contribute to the use and recycle of waste, generating environmental gains (Battistelle et al., 2009; Doost-hoseini et al., 2014; Mendes et al., 2009; Yang et al., 2003).

Sugarcane bagasse (SCB) is a residue of matted cellulose fiber derived from sugarcane processing. It is usually burned in steam reservoirs to produce energy for industrial use. SCB has been increasingly produced in larger

quantities due to increased acreage and industrialization of sugarcane, arising mainly from places of public and private investments in sugarcane production (DEA, 2013; Fiorelli et al., 2011). However, SCB has some drawbacks, such as: high water absorption (WA) due to the high amount of hemicellulose, which means more bonding sites for water bonding (Li et al., 2011); high amount of ash and extractives that can affect adhesive cure and adherence between the particles and the adhesive; and fibers with less stiffness and that are damaged due to the mechanical process of milling (Guimarães Júnior et al., 2012).

New technologies have been tested, especially in the area of nanotechnology. In addition, the reinforcement of composites/boards with nanofibers and cellulose nanocrystals (NCC) has shown very promising results (Atta-Obeng et al., 2012; Veigel et al., 2012; Zhang et al., 2011; Zorba et al., 2008). Improvements in the mechanical properties of the particleboard reinforced with nanocellulose (CNF) and microfibrillated cellulose (MFC) were found by (Mahrdt et al., 2015; Veigel et al., 2012)) and improve the physical properties by (Veigel et al., 2012)).

Therefore, the use of this “waste” (SCB) with NCC in the manufacture of particleboards can benefit the producers of sugar, brandy, and ethanol in generating new revenues. This contributes to the financial sustainability and diversification of sources of revenue by adding value to waste and reducing the use of wood and the consequent pressure on forests. However, it is necessary to understand how the NCC behaves with the adhesive used during the production of particleboards. Therefore, the aim of this research was to investigate the behavior of the urea formaldehyde adhesive with NCC and its feasibility in the production of SCB particleboards.

2 MATERIAL AND METHODS

2.1 Raw material and raw material characterization

Urea-formaldehyde adhesive (UF), SCB particles (kindly donated by Edra Ecosistemas Ltda), and freeze-dried NCC (lyophilized) purchased from the University of Maine, were used to produce the test specimens. The sliver particles were generated with a hammer mill and subsequently classified in sieves of 20 and 60 mesh. The particles retained in the sieves of 60 mesh were dried in an oven at 90°C until final moisture content of 4% and later used to produce the test specimens (particleboards).

The adhesive had its solid content measured by drying 1 g of adhesive for four hours in an oven at 105°C. The pH was measured using a pH meter and the viscosity using a Brookfield viscometer. The thermo-oxidative degradation behavior was studied using a thermogravimetric analyzer (TGA) instrument, model Q500. The tests were carried out in a synthetic air atmosphere (20% of O₂ and 80% of N₂) to simulate the real processing conditions, at a heating rate of 10°C.min⁻¹, from room temperature to 850°C. Differential scanning calorimetry (DSC) from TA instrument, model Q100 was used to understand how the UF behaved, from room temperature to 180°C. This DSC analysis was used to set the best cure temperature. An isothermal was carried out, using the temperature established in the first DSC run, to set the ideal time for the UF cure. Scans were performed on 8-15 mg at a scan rate of 10°C.min⁻¹, using UF (dried at 103°C for four hours, without a catalyst). The thermal properties were also studied using TGA and DSC.

2.2 Cellulose nanocrystals dispersion

The NCC were dispersed in 100 g of UF adhesive and sonicated (using a Branson digital sonifier) twice for three minutes each time. NCC of 0, 0.5, 1, 2, 3, and 5% (by adhesive mass, including the water) were used. Samples were run at 60% amplitude, where the beaker was partially immersed in an ice bath to ensure that the samples did not get overheated. The time chosen was through visual observation of the turbidity of resulting suspension. After the dispersion, the Brookfield viscosity was measured at 30°C for all treatments.

2.3 Test specimens production

2.3.1 UF test specimens

After sonification, the adhesive (UF) with NCC was dried in an oven at 105°C for the 4 hours. After this, the material was milled generating pellets. These pellets were put into iron molds and pressed at 1 MPa at 160°C for eight minutes to produce the specimens. Three point bending test were conducted according to ASTM D790-00 (ASTM, 2002), using 1 mm/min of speed. Each specimen was produced with 6.1 x 1.2 x 0.32 cm (L x W x T). Five specimens were tested for each formulation (0, 0.5, 1, 2, 3, and 5%). The UF specimens production are illustrated in Figure 1.

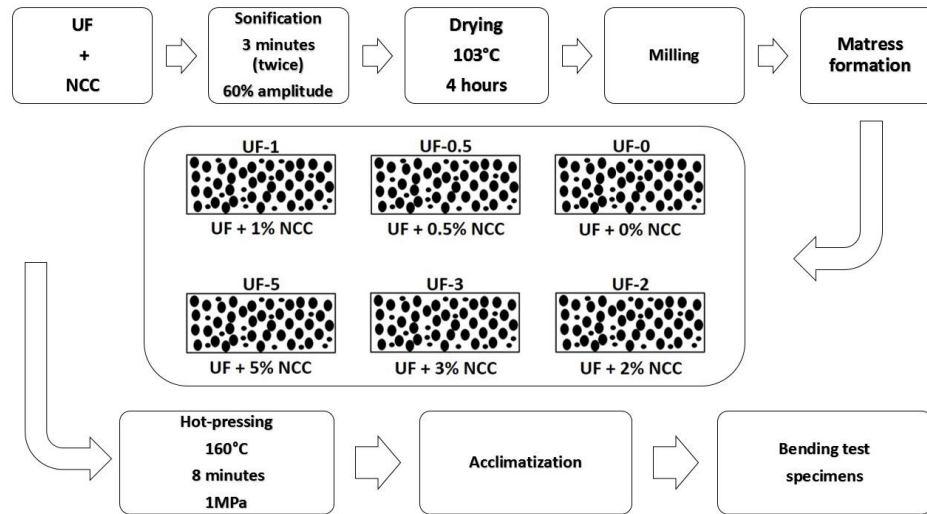


Figure 1 Illustrative scheme of urea formaldehyde (UF) specimens, their production and formulations.

2.3.2 Sugarcane bagasse test specimens

After sonification, the adhesive UF with NCC was applied on the SCB particles. UF and SCB were mixed manually, while taking care to evenly distribute it over the SCB particle. (20% of UF weight by SCB dried mass). The mixture was put into an iron mold (18.6 x 16.3 x 0.32 cm) and pre-pressed at 0.78 MPa in a hydraulic press to decrease the volume. After this, it was hot pressed with 1 MPa at 160°C for eight minutes. The SCB particleboards production is illustrated in Figure 2.

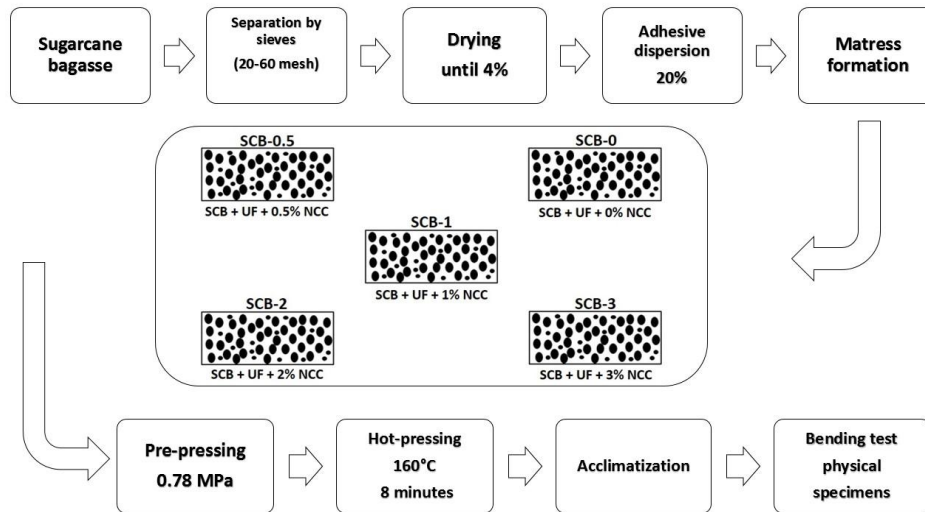


Figure 2 Illustrative scheme of SCB particleboard production and their formulations.

2.4 Thermal, mechanical, and physical properties evaluation of particleboards.

The density was measured using a balance and a caliper following ASTM D 1037 (ASTM, 2006).

The thermal behavior was studied using a TGA for the UF specimens. The samples were heated from room temperature to 600°C in a 40 mL min⁻¹ flow of N₂ and 60 mL.min⁻¹ of air. A heating rate of 10°C min⁻¹ was used. Records of sample temperature, sample mass, its first derivative, and heat flow were taken.

An EMIC DL 3000 universal testing machine was used in the bending test according to ASTM D790 (2000). Bending tests were performed using a three-point bending setup, five specimens, 1 mm/min of speed, and 4 cm of span. The specimens were cut using a three-point bending test according to ASTM D790-00 (ASTM, 2002), using 1 mm/min of speed. Each specimen were

cut with 6.1 x 1.2 x 0.32 cm (L x W x T). Five specimens were tested for each formulation (0, 0.5, 1, 2, and 3%).

Measurement of water absorption after two (WA2h) and twenty-four hours (WA24h) of immersion in water and thickness of swelling after two (TS2h) and twenty-four hours (TS24h) of immersion, were carried out according to EN 317 (EN, 1993).

The data were analyzed as a completely randomized design and the properties were compared by the mean values and standard deviation.

3 RESULTS AND DISCUSSION

3.1 Urea formaldehyde adhesive characteristics

UF in the liquid state showed 54% of solid content, 7.94 pH, and 0.127 Pa.s of viscosity. The TGA and DSC of the UF in the liquid state are presented in Figures 3, 4, and 5.

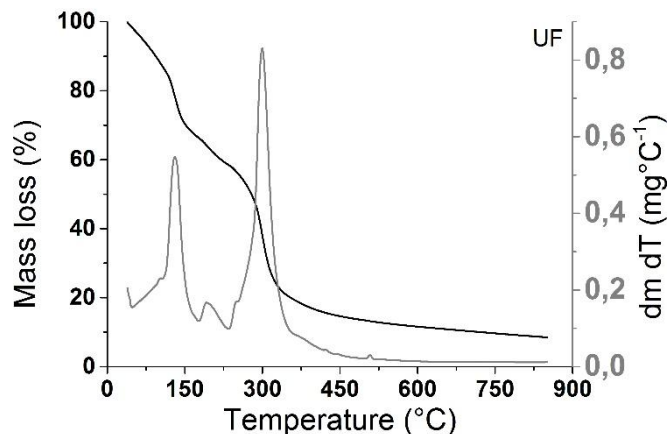


Figure 3 TGA and differential thermogravimetric curves of the urea formaldehyde (UF) adhesive.

In the beginning of the reaction the water used to dilute the adhesive controlled the reaction, but later, the water resulted from the condensation reaction was an important factor. According to (Zorba et al., 2008) methylols, urea, and formaldehyde react to give linear and partly branched molecules, with medium and even higher molar masses. The urea molecules bond with the methylene ether bridges ($-\text{CH}_2\text{-O-CH}_2-$) and methylene bridges ($-\text{CH}_2-$). The first endothermic peak, with a minimum of around 106°C , is related to water evaporation and the second peak around 200°C is attributed to a small mass loss due to the free formaldehyde's slow release (Fig. 3). The peak around 250°C is attributed to the degradation of the methylene-ether bridges in the resin's network (Siimer et al., 2003; Zorba et al., 2008). The last peak with a minimum of around 305°C is related to the decomposition of the most stable unit in the UF adhesive, the methylene diurea (Dunky, 1998; Siimer et al., 2003).

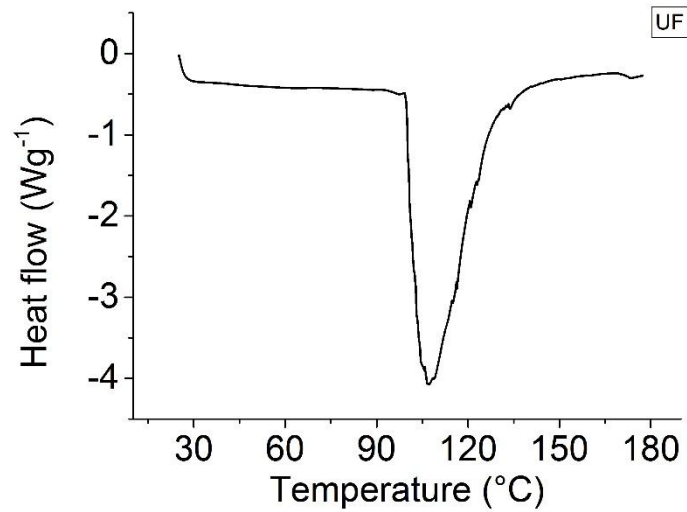


Figure 4 DSC curve for urea formaldehyde (UF).

The reaction is controlled by water loss and observed using the DSC (Fig. 4). The endothermic reaction has prevailed because of the amount of water present in the UF. The water loss peak is at 107°C. The curing of the resin is extended in a wide temperature area, until about 160°C, where it achieves a plateau. Thus, 160°C is the temperature set for specimen production.

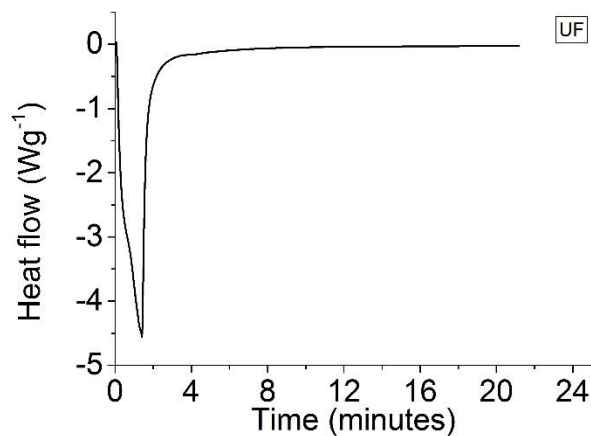


Figure 5 Isothermal at 160°C of urea formaldehyde (UF) set for the cure time.

With the information obtained in the first DSC, an isothermal was made to try to find the best time to process the UF (Figure 5). This isothermal was made with dried UF (105°C for three hours) to try to decrease the influence of water. It was found that at 160°C the time needed for maximum polymerization was eight minutes. Therefore, those parameters were chosen to produce the specimens of UF and UF plus SCB.

3.2 Characteristics of nanocrystals of cellulose

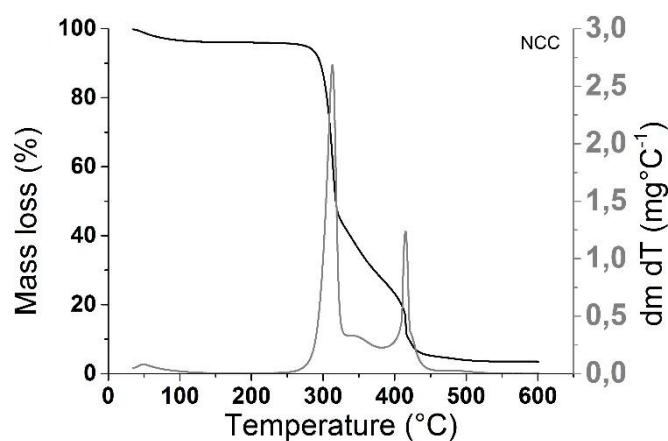


Figure 6 TGA curves of the nanocrystals of cellulose (NCC).

The NCC lost a small mass amount around 100°C due to a small amount of moisture (3.9%) figure 6. However, the NCC started to degrade after 250°C. The NCC is obtained from a fraction with a three-dimensional arrangement, an organized area of the cellulose, and because of this, they are so resistant to thermal degradation. Therefore, it will not degrade at the temperature chosen of hot pressing (160°C). Cellulose degrades between 275°C and 400°C (Yang et al., 2007), but as NCC is a stable structure, it can explain an increase of temperature. Two phases were observed, with a lot of loss weight, the first one from about 300 to 325°C and another from 400 to 425°C. The cellulose is a long polymer of glucose units and its crystalline regions improve the thermal stability (Poletto et al., 2012; Yang et al., 2007; Yang et al., 2006).

3.3 Characteristics of sugarcane bagasse

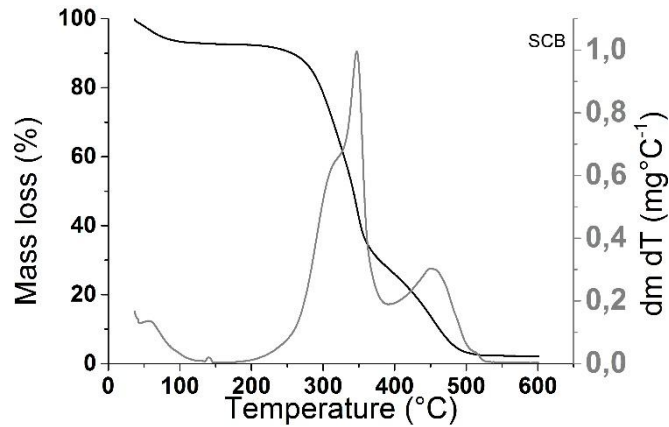


Figure 7 TGA of sugarcane bagasse (SCB).

The TGA of SCB particles is showed in Figure 7. The SCB lost about 8% of mass around 100°C due to the amount of moisture. Cellulose degrades between 275°C and 400°C. Hemicellulose starts its decomposition early, but the highest mass loss occurs from 220° to 315°C. Lignin mass loss occurs at a huge temperature range, with higher mass loss at higher temperatures (Yang et al., 2007). As SCB has more cellulose and hemicellulose, the highest mass loss occurs between 300 and 360°C.

3.4 Viscosity of a mixture of urea formaldehyde and cellulose nanocrystals.

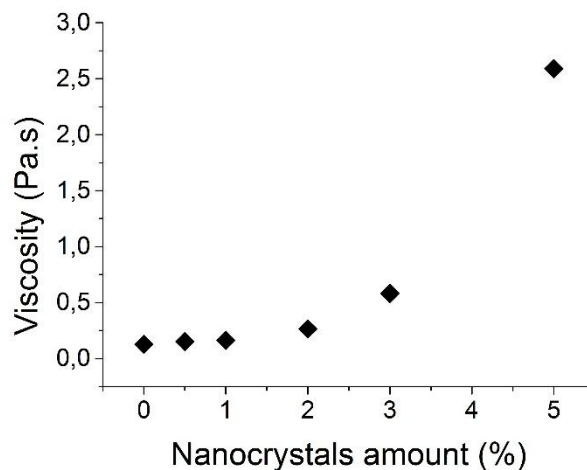
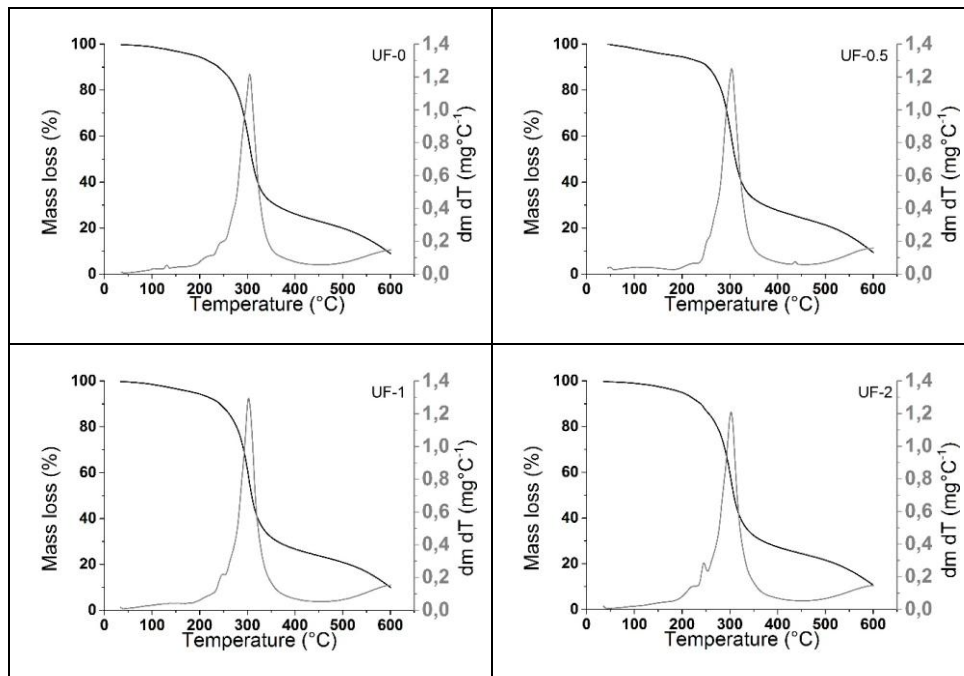


Figure 8 Viscosity of a mixture of urea formaldehyde and nanocrystals of cellulose

The change in viscosity with the addition of NCC is shown in Figure 8. The viscosity increased directly with the amount of NCC. This effect occurs because the high surface area NCC increases the hydrogen bonds with the compounds in the system, increasing the viscosity of NCC/UF. On account of the high viscosity of UF of 5% this treatment was not used to produce particleboards. This high viscosity could prevent the dispersion and penetration of the adhesive, as indicated by the researchers. High viscosity can decrease the flowing ability, wetting behavior, adhesive distribution, and adhesive penetration (Dunky, 2004).

3.5 Thermal properties of the formulations

The TGA of the UF specimens did not differ with NCC reinforcement, because both materials, UF and NCC, had a similar peak of degradation and the mixture low amount of NCC reinforcement (Figure 9). However, it seems that there is a tendency for the peak to increase from 225°C to 255°C, and this is attributed to the degradation of methylene-ether bridges, and accordingly the NCC increases.



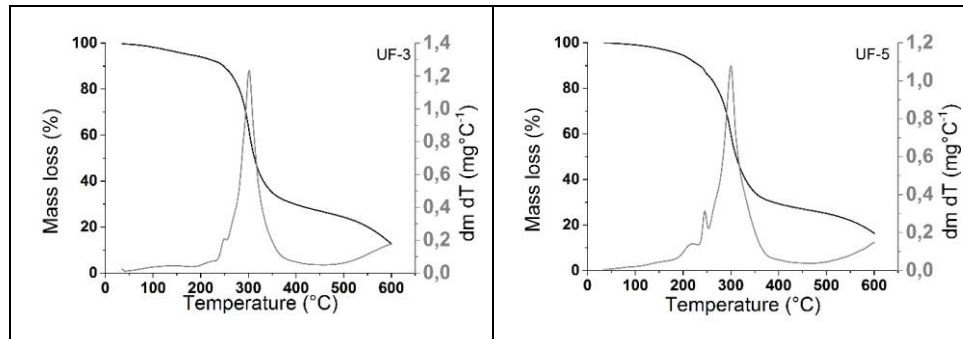


Figure 9 TGA of urea formaldehyde UF specimens. (UF-0) specimens without NCC; (UF-0.5) specimens with 0.5% NCC; (UF-1) specimens with 1% NCC; (UF-2) specimens with 2% NCC; (UF-3) specimens with 3% NCC; and (UF-5) specimens with 5% NCC.

3.6 Physical and Mechanical properties

The physical and mechanical properties of UF and SCB specimens are shown in Table 1.

Table 1 Physical and mechanical properties of UF and SCB specimens. Density; bending modulus (MOE); bending strength (MOR); water absorption after two hours (WA2h) and twenty-four hours (WA24h); thickness swelling after two hours (TS2h) and twenty-four hours (TS24h). Mean values followed by standard deviation.

Treatments	Properties						
	Density (g.cm ⁻³)	MOE (MPa)	MOR (MPa)	WA2h (%)	WA24h (%)	TS2h (%)	TS24h (%)
UF-0	0.999±0.079	2277±125	14±2	-	-	-	-
UF-0.5	1.035±0.060	2163±147	15±2	-	-	-	-
UF-1	1.041±0.046	2576±63	16±3	-	-	-	-
UF-2	1.065±0.017	2724±343	17±3	-	-	-	-

UF-3	1.069±0.030	2588±211	15±2	-	-	-	-
UF-5	1.101±0.024	2956±237	14±1	-	-	-	-
SCB-0	0.557±0.027	1185±95	16±1	52±3	95±5	8±1	13±1
SCB-0.5	0.559±0.054	886±85	11±2	81±13	104±16	16±1	20±1
SCB-1	0.563±0.027	1126±88	15±2	54±11	86±10	9±1	15±1
SCB-2	0.556±0.046	994±99	14±2	54±3	91±9	10±1	17±3
SCB-3	0.545±0.027	1032±38	14±1	81±9	112±11	12±1	16±1

The density of the specimen is a characteristic that grades and ensures a fair evaluation between treatments, thus allowing to proceed with the comparison between the average values of the mechanical properties. The density of the UF specimens ranged from 0.999 to 1.100 g.cm⁻³ and the density of the SCB specimens ranged from 0.557 to 0.565 (Table 1). For the UF specimens, the density showed a slight increase according to the amount of NCC that had increased, but the standard deviations suggest that the densities were about the same. The same occurred with the SCB specimens. According to (Iwakiri, 2005) these particleboards could be classified as low density particleboards.

The UF bending modulus (MOE) first decreased with 0.5% of NCC and later increased with 1 and 2% of reinforcement, decreasing again with 3%. However, only the reinforcement with 0.5% NCC was lower than the control treatment. The bending strength (MOR) showed a behavior very similar to the bending modulus, however, all treatments with reinforcement had higher MOR's than the control treatment. Veigel et al. (2011) added NCC in the UF adhesive and proposed that the positive effect observed was caused by the role that NCC played in the process of crack formation. Furthermore, the more stress taken up by the NCC increases the MOR. Adhesion is thus one key factor in improving strengths apart from proper dispersion. Reaction between UF and NCC should explicate this behavior, because NCC is rigid and after reaction it can be result a

more rigid composite.

The MOE and MOR of SCB also showed a decrease with 0.5% and later an increase with 1% of reinforcement. However, differing from the UF specimens, all treatments with NCC showed lower values than the control treatment. According to the standard CS 236-66 (CS, 1968), specimens are classified as type 1 (particleboards for internal use with UF adhesive). As for the densities (lower than 0.60 g.cm^{-3}), the minimum required for MOE and MOR are 1030 MPa and 5.5 MPa, respectively. All treatments exceeded the minimum for MOR, but for MOE the control treatment and treatments with 1 and 3% of NCC were the only ones that achieved the minimum values of the standard.

According to (Veigel et al., 2011)) increase in the viscosity can reduce the adhesive penetration into the cellular wood cavities. This may explain why the properties increased only in the UF when the NCC are added only to the UF, but do not increase when put in SCB particleboards. Penetration is an important step in particleboard production since it creates “mechanical locking” because the adhesive can create hooks that can improve the performance of the mechanical properties. Another factor that can affect mechanical properties in SCB/UF/NCC is the reduction of the number of reactive groups in the UF to react with SCB, because they can react preferentially with NCC, with less reaction with SCB. NCC has high surface area and reactivity comparing with SCB, and increase the rigidity (MOE) of the interface UF/NCC with SCB.

The mean values for MOE and MOR are very close to those found for (Battistelle et al., 2009) by using sugarcane bagasse to produce a particleboard with 0.56 g.cm^{-3} of density. (Doost-hoseini et al., 2014) evaluated SCB particleboards with a density of 0.5 g.cm^{-3} and found lower mean values (12 MPa) for MOR. However, those authors used lower amount of adhesive UF (12%) and also the lower density (0.5 g.cm^{-3}) which could explain the lower mean values. (Carvalho et al., 2015) evaluated commercial SCB particleboards

from China and found mean values for MOR of 17.2 MPa, similar to those in the present study, however, there was no information about the production process of the particleboards from China.

(Atta-Obeng et al., 2012) investigated the effect of the species (sweetgum and pine) and inclusion of microcrystalline cellulose (MCC) (0 and 10%) in particleboards using phenol formaldehyde (PF). The inclusion of MCC in particleboards resulted in a decline in the boards mechanical properties (MOE and MOR). The mechanical properties decreased because a high amount of MCC was used, which could be because of higher viscosities and the MCC can agglomerate and harm the dispersion that later affected the properties of the particleboards. Similar with this study, MCC has surface area higher than particles, and it can react preferentially with adhesive (PF).

(Mahrtdt et al., 2015) studied the effects of MFC addition (5%) in UF and its application in particleboards. They observed that MFC addition increased the viscosity and the cure time of the adhesive UF, as it was observed in this work. However, those authors did not evaluate the properties of MOE, MOR, water absorption (WA), and thickness swelling (TS), but only of the internal bond (IB). Particleboards with MFC inclusion showed higher values (30% more) for IB compared to particleboards without MFC. The authors attributed this better performance to the distribution of the adhesive, as a higher fraction of adhesive + MFC was available for bond-line formation, and a larger part of the wooden particles were covered with the adhesive. This is an important aspect that was not achieved in this study, and it needs to be improved and better understood.

(Veigel et al., 2012) mixed CNF with UF in different reinforcement proportions (0, 1, and 3%) and used it to produce particleboards. The viscosity of the adhesives increased with the insertion of CNF and because of this, it was not possible to use a high amount of CNF as reinforcement. The mechanical

performance (internal bond, bending strength, fracture energy, and fracture toughness) of the boards (with 1% CNF) was enhanced, whereas, with 3% of CNF it was worse than the control. The internal bond increased by 10% from 0.62 to 0.68 MPa and the bending strength was enhanced from 18.4 to 19.5 MPa. The author suggested that this deterioration was not directly induced by a higher CNF content, but could be explained by the long drying time of the glue on the wood particles prior to hot pressing. This fact showed that the reactivity time is another key fact that can change the properties in particleboards with CNF.

The WA increased significantly with 0.5% and 3% of NCC load in the SCB specimens as compared to the control. For 1% and 2% of the NCC load, the WA_{2h} was almost the same, with a slight decrease was observed for WA_{24h}. For all NCC loads the TS was higher than for particleboards without NCC. However, the increase in TS was not linear.

(Doost-hoseini et al., 2014) evaluated SCB particleboards with 0.5 g.cm^{-3} of density and found higher mean values for WA_{2h} (110%), WA_{24h} (140%), TS_{2h} (16%), and TS_{24h} (19%). However, those authors used a lower amount of adhesive UF (12%) and this could explain the large difference between the mean values in this present research that used 20% UF. Increasing the amount of adhesive, decreases the WA and TS (Mendes et al., 2009). (Carvalho et al., 2015) evaluated the commercial SCB particleboards from China and found mean values for WA_{2h} of 17.3%, WA_{24h} of 53.3%, TS_{2h} of 5.3%, and TS_{24h} 11.5%. Those mean values are much lower than those found in the present study are. However, they did not have the parameters of the China particleboards production.

(Atta-Obeng et al., 2012) investigated the effect of the species (sweetgum and pine) and inclusion of MCC (0 and 10%) in particleboards using PF. The inclusion of MCC in the particleboard resulted in a higher thickness

swell, both after two and twenty-four hours, thus the MCC effect was negative. According to the authors, inclusion of MCC provides more available hydroxyl groups for moisture sorption and consequent thickness swelling.

(Veigel et al., 2012) mixed CNF with UF in different reinforcement proportions (0, 1, and 3%), and used it to produce particleboards. The TS performance of the boards (with 1% CNF) decreased, whereas, with 3% of CNF it increased more than that of the control.

According to the standard CS 236-66 (CS, 1968) specimens are classified as low density (lower than 0.60 g.cm^{-3}) and the maximum required for TS24h is 30%. All treatments have shown mean values lower than 30%. The standard does not provide maximum values for WA.

For most properties, the specimens with NCC have shown higher variability than the control. The same was observed by (Veigel et al., 2011) and the author attributed that to the sub-optimal dispersion of NCC. The adhesive dispersion (by hand) may have entailed an incomplete and heterogeneous dispersion, which means it is possible that some particles did not receive adhesive or received less adhesive, reducing the bonding and consequently the particleboards properties.

4 CONCLUSIONS

NCC improved UF properties, showing a viscosity increase in liquid suspension UF/NCC, and a better mechanical performance of solid specimens UF/NCC.

In the SCB/UF/NCC particleboards, improvement was not observed in those specimens because less site links were available for the SCB to interact with the adhesive and higher viscosity of UF/NCC could decrease adhesive

penetration. These factors could affect the mechanical behavior of SCB/UF/NCC particleboards.

New studies should be carried out to understand better how the dispersion, reactivity and viscosity of the system UF/NCC with SCB could improve the properties of particleboards.

SCB is a promising material for particleboard production, and it was showed the possibility to get SCB particleboards with appropriate performance, using an agroindustrial waste and collaborating with the environment.

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APPENDIX A - SUPPLEMENTARY MATERIAL TO CHAPTER 2. NOT INCLUDED IN THE MANUSCRIPT



Figure B 1 Production of UF composites. A – NCC dispersion. B – Adhesive with 0% of NCC and with 5% of NCC. C – Adhesive being dried. D – Adhesive dried. E – Adhesive milled. F – UF pellets in the iron mold. G – Hot pressing. H – UF composite. I Bending test.

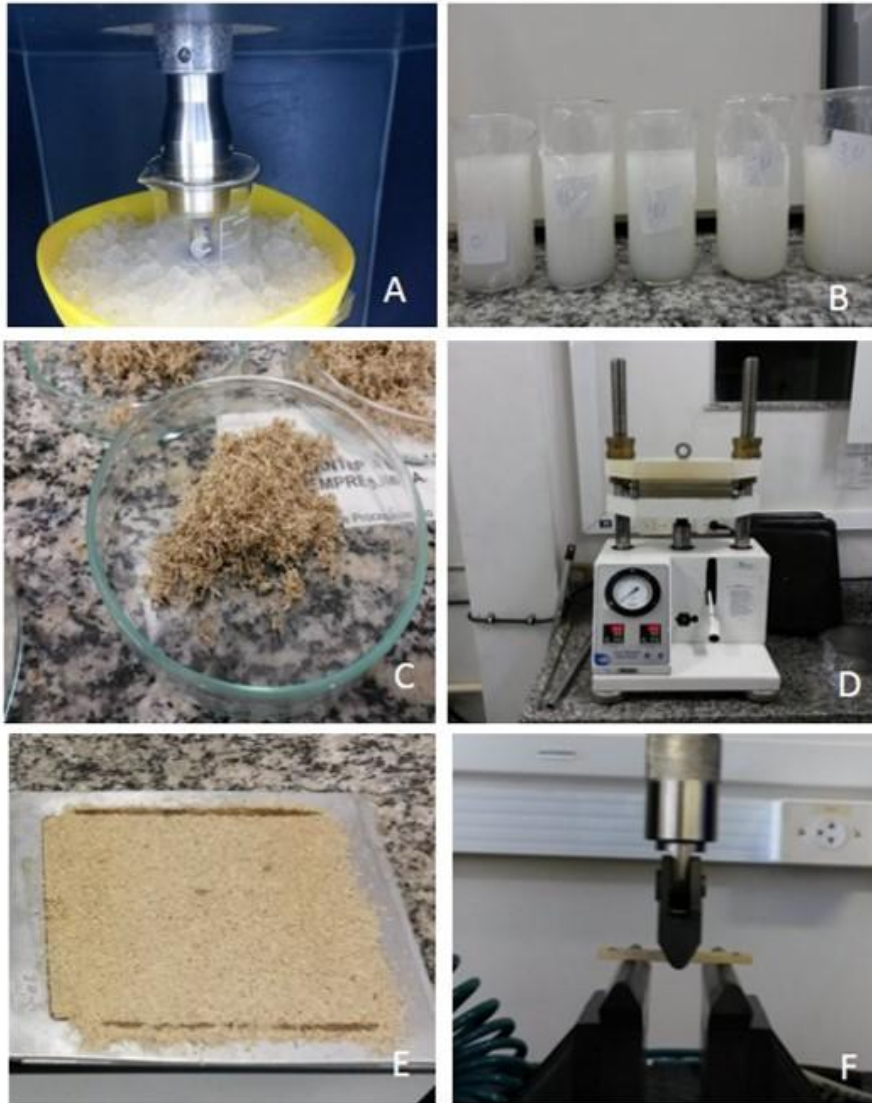


Figure B 2 Production of SCB particleboard. A – NCC dispersion. B – Adhesive with NCC. C – SCB particles. D - Hot press. E – SCB particleboard. F – Bending test.

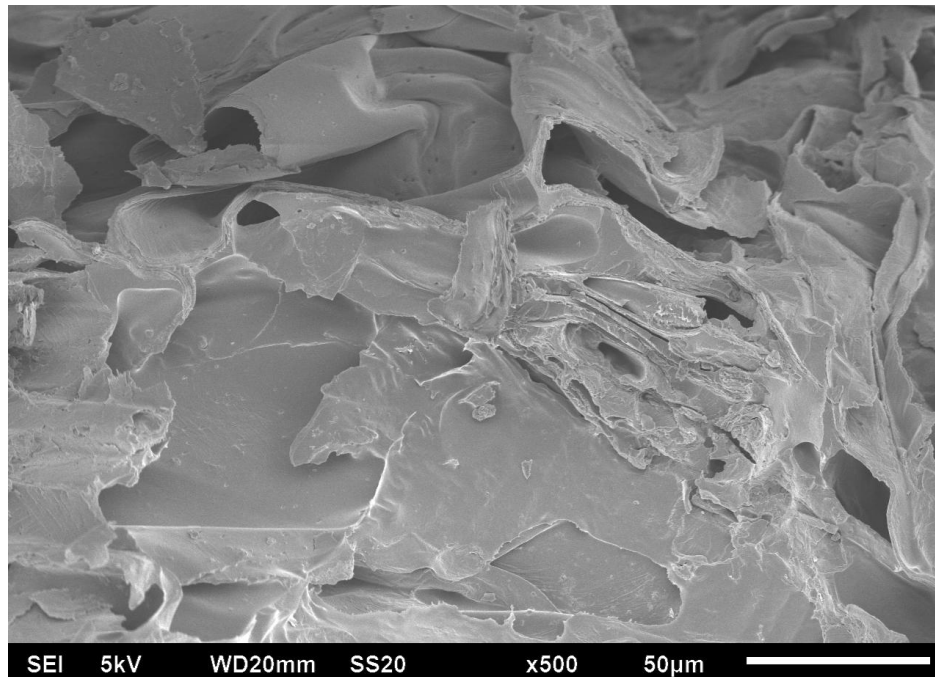


Figure B 3 SEM micrographs of the fracture surface from SCB specimens without NCC (particleboard). View of surface morphology (500 x magnification).

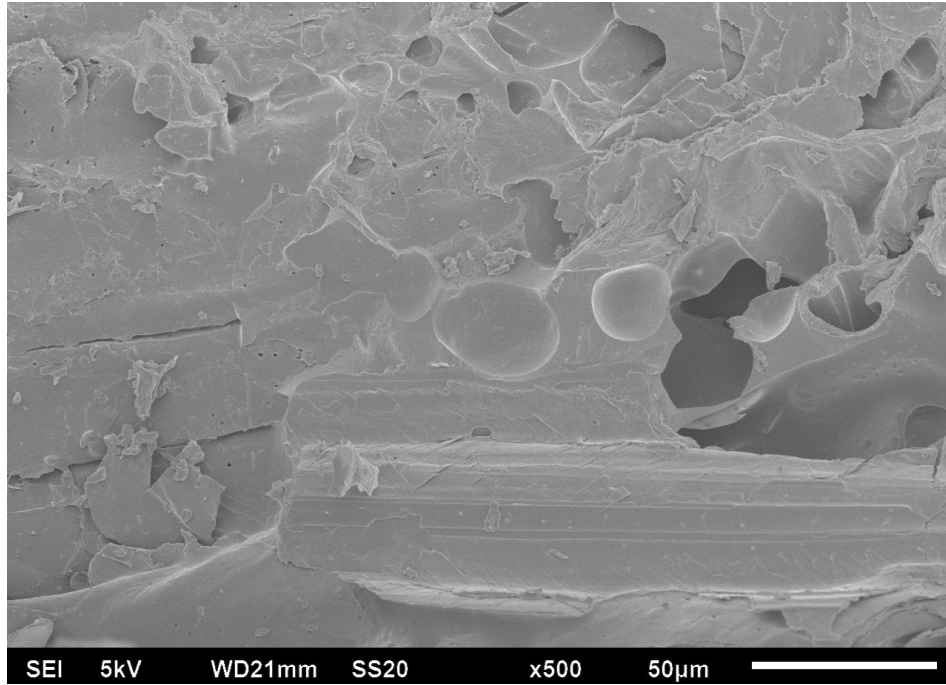


Figure B 4 SEM micrographs of the fracture surface from SCB specimens with 3% of NCC (particleboard). View of the surface morphology (500 x magnification).

Table B 1 Physical and mechanical properties of UF and SCB specimens. Density; bending modulus (MOE); bending strength (MOR); water absorption after two hours (WA2h) and twenty-four hours (WA24h); thickness swelling after two hours (TS2h) and twenty-four hours (TS24h).

Treatments	Properties						
	Density (g.cm ⁻³)	MOE (MPa)	MOR (MPa)	WA2h (%)	WA24h (%)	TS2h (%)	TS24h (%)
UF-0	0.999 a	2277 a	14 a	-	-	-	-
UF-0.5	1.035 a	2163 a	15 a	-	-	-	-
UF-1	1.041 a	2576 b	16 a	-	-	-	-
UF-2	1.065 a	2724 b	17 a	-	-	-	-
UF-3	1.069 a	2588 b	15 a	-	-	-	-
UF-5	1.101 a	2956 c	14 a	-	-	-	-
SCB-0	0.557 A	1185 C	16 B	52 A	95 A	8 A	13 A
SCB-0.5	0.559 A	886 A	11 A	81 B	104 B	16 D	20 B
SCB-1	0.563 A	1126 C	15 B	54 A	86 A	9 A	15 A
SCB-2	0.556 A	994 B	14 B	54 A	91 A	10 B	17 B
SCB-3	0.545 A	1032 B	14 B	81 B	112 B	12 C	16 A

Mean values with an average test after ANOVA. Same letters in the column indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

**MANUSCRIPT 3 - THE EFFECT OF CELLULOSE NANOCRYSTALS
IN SUGARCANE BAGASSE PARTICLEBOARDS OF PITH AND
FIBERS**

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ABSTRACT: The aim of this research was to investigate sugarcane bagasse (SCB) particleboards, using two kinds of particles, from pith and fibers, reinforced with cellulose nanocrystals (NCC). The NCCs were dispersed in the adhesive urea formaldehyde (UF) using a sonicator in two proportions (0 and 1%). The NCCs and UF were applied in the SCB particles using a rotary blender. The particleboards were produced in three layers using two kinds of particles: pith and fibers. After dispersion of the adhesive, the mixture was compressed to produce the particleboards, which were characterized by physical, mechanical and thermal properties. The anatomical and chemical features relating to pith and fiber were very similar. The particleboards made with and without NCCs did not differ statistically for most properties: modulus of elasticity (MOE), modulus of rupture (MOR), water absorption (WA) thermal conductivity (TC), and the volume heat capacity (VHC). Particleboards made only with fibers and without NCCs showed higher internal bond (IB). In the other hand particleboards made only with fibers in the face and pith in the core (without NCCs) showed lower thickness swelling after 24 hours (TS24h). Probably, the NCCs did not improve the particleboards performance, because less links were available for SCB to interact with the adhesive. For the others properties the particleboards made with pith and fibers did not showed statistical difference (MOE, MOR, TC, WA, TC and VHC), demonstrating that separation of pith and fiber are not significant and does not affect the final properties of particleboards. The SCB particles can be used to produce particleboards for internal purposes, as an alternative way to get new materials from agriculture residues.

Keywords: Natural fiber, medium density particleboard, chipboards, urea formaldehyde, nanotechnology

1 INTRODUCTION

Sugarcane bagasse (SCB) is a residue of lignocellulosic fiber derived from sugarcane processing and juice extraction. It is used in the sugar and ethanol industry to generate heat, steam and energy. Sugarcane acreage and industrialization has been increasingly in the last years, therefore also the SCB amount (DEA, 2013; Driemeier et al., 2011; Fiorelli et al., 2011; Rasul et al., 1999; UNICA, 2017). The chemical composition of SCB varies according to several factors, such as type of cane, soil type and harvesting techniques. Cellulose content, lignin and polyoses in the pith (not fiber fraction) and the fibers closely resemble each other as well as hardwoods (Hemmasi et al., 2011). However, pith and true fibers are different in morphological functions and concerning their liquid absorption capacities (Driemeier et al., 2011; Lee and Mariatti, 2008; Martinez et al., 1997; Widyorini et al., 2005). The volume of bagasse from the sugarcane harvest in Brazil was $174320 * 10^3$ tons (DEA, 2013). The magnitude of such a volume and its availability contributes to increasing interest in this waste by product.

Researchers have been studying new applications for SCB, and trying to bring about changes in a residue by product to create new products with more added value, such as composites (Loh et al., 2013; Oliveira et al., 2016b), and one example of these composites is the particleboard (Lee et al., 2006; Widyorini et al., 2005). Particleboards are normally produced with elements of the breakdown of wood (particles/chips), which are later combined with adhesive bonding under pressure and high temperature (Iwakiri, 2005; Kelly, 1977; Maloney, 1996). They can be produced in single layers (chipboards) or in three layers (medium density particleboards - MDP). Regarding the raw material wood is the most used, although, any lignocellulosic material can be used for the particleboard production, such as SCB (Carvajal et al., 1996; Guler et al., 2016;

Han et al., 2005; Hazrati-Behnagh et al., 2016; Jonoobi et al., 2016; Khazaeian et al., 2015; Lee et al., 2006; Oliveira et al., 2016a; Rangavar et al., 2016). One advantage of SCB is that, it can be easily converted into sliver particles, similarly as with wood (Driemeier et al., 2011; Garzón-Barrero et al., 2016; Hemmasi et al., 2011; Lee et al., 2006; Wu, 2001).

Due the chemical composition and the mechanical processing, SCB has some drawbacks such as: high water absorption (WA) (Lee and Mariatti, 2008; Xu et al., 2009); high amounts of ash and extractives that can affect the adhesive cure and the adherence between particles and adhesive (Widyorini et al., 2005; Xu et al., 2009); and, fibers (cells) with less stiffness and more damaged because of the sugarcane milling process (Guimarães Júnior et al., 2012; Mesquita et al., 2016a). New studies have been done trying to overcome those problems.

Nanotechnology has been applied in adhesives, particleboards and composites to overcome the lignocellulosic drawbacks and to improve their properties (Candan and Akbulut, 2013; Kumar et al., 2013; Liu and Zhu, 2014; Marzbani et al., 2015; Salari et al., 2012, 2013). The incorporation of nanofibers and nanocrystals from cellulose has shown to be very promising since the particleboards have shown better performance (Atta-Obeng et al., 2012; Veigel et al., 2012; Zhang et al., 2011; Zorba et al., 2008). Better mechanical properties performance (internal bond and modulus of rupture) of particleboards reinforced with nanocellulose (CNF) and microfibrillated cellulose (MFC) were found by (Mahrdt et al., 2015; Veigel et al., 2012), and also better performance in physical properties (thickness swelling) by (Veigel et al., 2012).

Thus, the aim of this research was to produce and investigate the effects of cellulose nanocrystals (NCC) inclusion in particleboards made with pith and fiber particles.

2 MATERIAL AND METHODS

2.1 Raw material and raw material characterization

Urea formaldehyde adhesive (UF), NCC freeze dried, and SCB particles were used to produce the particleboards. Sugarcane bagasse particles were separated in two materials: pith and fibers.

The adhesive solid content was measured by drying 1 g of adhesive during 4 hours in an oven at 105°C. The pH was measured using a pH meter and the viscosity was measured using a Brookfield viscometer.

The sugar cane bagasse (SCB) was from São Paulo State, Brazil, kindly donated by Edra Ecosistemas Ltda. The length, width, and lumen diameter of the individual cells from fibers and pith were performed with the aid of Olympus BX41 microscope, using Wincel Regent PRO software. Each of the morphological features was measured in at least 30 fibers. To separate the particles in individual fibers the samples passed through maceration process. Anatomical terms describing the fibers were used as recommended by the International Association of Wood Anatomists -IAWA (Wheeler et al., 1989). The flexibility coefficient (FC) and wall fraction (WF) parameters were calculated for individual fibers (De Paula and da Silva Junior, 1994; de Paula, 1993; Fonseca et al., 2013). The FC was calculated by the ratio between the fiber lumen diameter and the fiber cell diameter, expressed in percentage. The WF was calculated by the ratio between cell wall thickness and fiber cell diameter, expressed as a percentage. The pith and fiber particles had the chemical analysis measured according to the TAPPI standard (TAPPI, 2011).

The NCC freeze-dried were purchased from University of Maine – EUA. Transmission electron microscope was used to take TEM images from NCC.

2.2 Cellulose nanocrystals dispersion

The NCCs were dispersed directly in the adhesive (UF). It used 0 and 1% of NCC (by adhesive mass, including the water). The NCCs were placed in 100 g of adhesive and sonicated twice during 3 minutes each time. The samples were run at 60% amplitude and the beaker was partially immersed in an ice bath to avoid that the samples get overheated (Figure 1). The time was chosen through visual observation of the turbidity of the resulting suspension. After the dispersion, the adhesive had a Brookfield viscosity measured at 30°C.



Figure 1 Beaker immerse in ice for NCC dispersion using a Branson digital sonifier.

2.3 Particleboards production

The SCB was separated in two materials: pith and fiber. The separation was made by density difference using vibrating sieves. Later on, the SCB was

passed through the hammer mill to generate the sliver particles and, subsequently, they were classified in sieves of 14, 20 and 120 mesh. The particles retained in the sieves of 120 and 20 mesh were used for the faces and cores, respectively (particleboards in 3 layers). The particles were dried in an oven at 90°C until they achieved a final moisture content of 4%.

The adhesive was spread using a rotary blender in two steps. Initially, the adhesive was applied to smaller particles (for the two faces) in a proportion of 12% (dry basis mass of the particles). Later on, the bigger particles (for the core) were coated with the adhesive in a proportion of 10% (dry basis mass of the particles). Due to the higher surface area to volume, more adhesive was used for the smaller particles to guarantee good adhesion. For each treatment, three particleboards were produced with a nominal density of 0.650 g.cm⁻³. The proportion used in the three layers (face/core/face) was 20/60/20 (basis of mass particles), respectively. The adhesive reinforced with NCC was used only in the faces, and all the faces were produced only with fibers (Table 1). The particleboards produced were compared with commercial SCB particleboards from China.

Table 1 Treatments

Treatments	Face (20%) - small particles (basis of mass particles)	Core (60%) - big particles (basis of mass particles)
Commercial	-	-
Pith-0	Fibers	Pith
Pith-1	Fibers + 1% NCC	Pith
Fiber-0	Fibers	Fibers
Fiber-1	Fibers + 1% NCC	Fibers

To decrease the mattress thickness, the particle mattress was pre-

pressing in a hydraulic press at 0.78 MPa of pressure. Later on, it was transferred to a hot press machine (3.92 MPa of pressure, at 160°C, during 8 minutes) (Figure 2).



Figure 2 Particleboard production. Adhesive dispersion, particle mattress formation, mattress after pre pressing and hot pressing.

After the hot pressing and subsequent cooling, at room temperature, the edges of the boards were trimmed, resulting in final dimensions of 30 * 30 * 1.5 cm. The boards followed to controlled chamber ($22\pm 2^{\circ}\text{C}$ and $65\pm 5\%$ of moisture) for acclimatization until constant mass was achieved before of properties characterization (Figure 3).

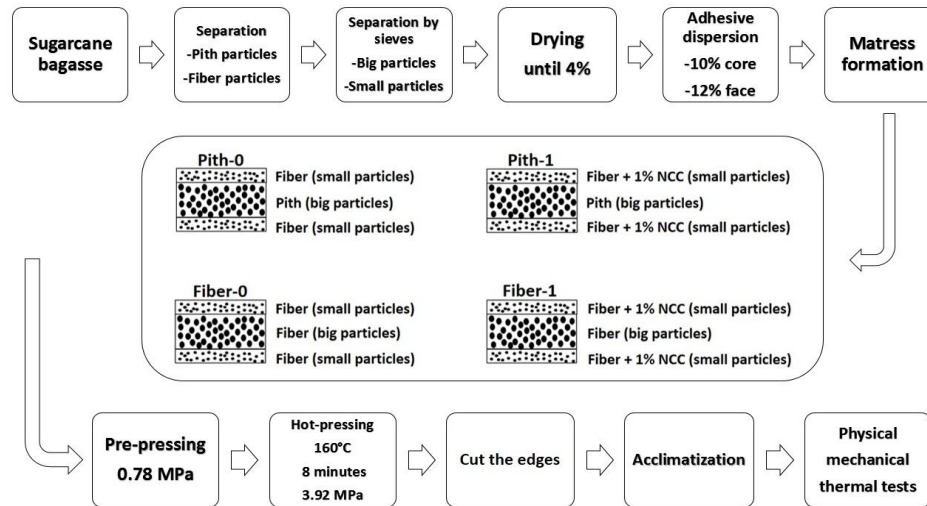


Figure 3 Illustrative scheme of particleboard production

2.4 Particleboards characterization

Particleboards were evaluated by measuring their physical, mechanical and thermal properties by using standardized tests: the density was evaluated according to EN 323 standard (EN-323, 1993), using 5 samples per treatment; the WA after 2 hours (WA2h), WA after 24 hours (WA24h), thickness swelling after 2 hours (TS2h), and thickness swelling after 24 hours (TS24h) of immersion in water according the EN 317 (EN-317, 1993), using 12 samples per treatment; the modulus of elasticity (MOE), modulus of rupture (MOR), and internal bond (IB) according the ASTM D1037 (ASTM, 2006) using an EMIC DL 3000 universal testing machine. 6 samples were used for MOE and MOR per treatment and 15 samples for IB.

Thermal properties were measured in an equipment ISOMET model 2104. The samples with 6 * 6 * 1.5 cm were placed in a controlled chamber (20°C and 65% of moisture) until constant mass before being tested. Five

samples were measured for each treatment.

The data were analyzed as a completely randomized design. It was made an analysis of variance (ANOVA) and when significant the Scott-Knott ($p < 0.05\%$) test average. The results were processed with the aid of SISVAR software.

3 RESULTS AND DISCUSSION

3.1 Urea formaldehyde adhesive characteristics

UF in the liquid state showed 54% of solids contents, 7.94 of pH and 0.127 Pa.s. of viscosity (at 30°C). The viscosity of the adhesive increased with the NCC inclusion, from 0.127 to 0.162 Pa.s. More details about the UF adhesive characteristics can be found in a previous work (Mesquita et al. 2017 in submission).

3.2 Cellulose nanocrystals characteristics

The figure 4 shows the TEM images of the cellulose nanocrystals. More details about the NCC characteristics can be found in a previous work (Mesquita et al. 2017 in submission).

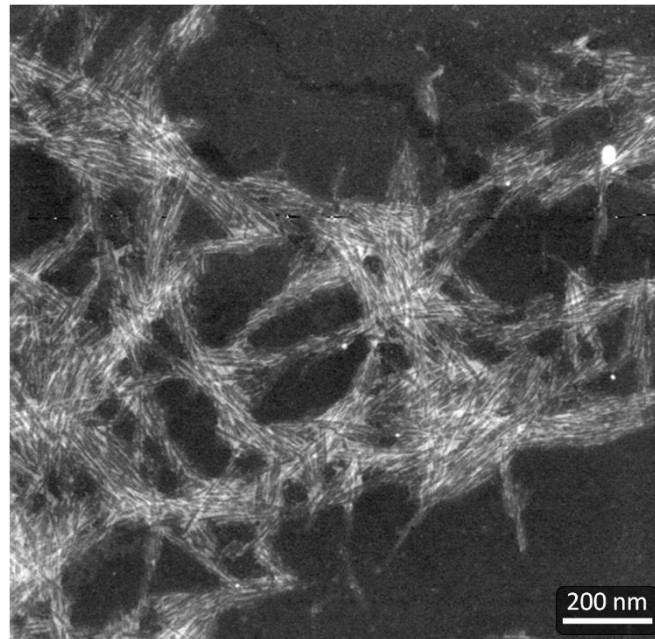


Figure 4 TEM micrograph of NCC.

3.3 Pith and fiber characteristics

The anatomical characteristics did not differ for all characteristics (Table 2). The values of the fiber cell length and diameter are in agreement with the literature, and the SCB has intermediate values between *Eucalyptus* and Pine wood (Hemmasi et al., 2011; Mesquita et al., 2016a). Hemmasi et al. (2011) measured just fibers and found similar mean values: 1590 μm (length), 20.96 μm (cell diameter), 5,64 μm (wall thickness) and 9.72 μm (lumen width). The cell wall fraction is related to the rigidity of the cell and values above 60% are normally related to stiffer fibers (Junior et al., 2010). However, the values of fibers and pith were very similar, close to 40%.

Table 2 Anatomical and chemical analysis

Anatomical characteristics	Fiber	Pith
Length (μm)	1512.33 a	1474.31 a
Cell diameter (μm)	20.38 a	19.41 a
Lumen diameter (μm)	12.34 a	11.06 a
Cell wall thickness (μm)	4.02 a	4.17 a
Flexibility coefficient (%)	58.13 a	54.62 a
Cell wall fraction (%)	41.86 a	45.37 a

Same letters in the line indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$).

Driemeier et al. (2011) classified particles (from pith and rind) in four sieves (4, 0.85, 0.6, and 0.15 mm). They classified as rind those particles that were retained in the three first sieves, and the last one as a mix of fibers and pith. Those particles that passed through the 0.15 mm sieve were classified as pith. However, they observed the presence of fibrous particles in all powders, including the pith fraction. They explain that it is related to the limitation of sieves to hold fibrous particles, which may pass sieves longitudinally. In this present work, it was also possible to find true fiber in the pith particles.

Table 3 Chemical composition of pith and fiber

Chemical composition	Fiber (%)	Pith (%)
Cellulose	57.33 a	53.9 b
Hemicellulose	17.10 a	17.2 a
Lignin soluble	0.76 a	0.39 b
Lignin insoluble	24.63 b	26,4 a
Extractives	1.24 a	1.11 b
Ash	1.18 a	0.82 b

Same letters in the line indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$).

The pith showed a higher amount of lignin, and the fibers showed a higher amount of cellulose, extractives and ash (Table 3). The pith and fibers did not show high amounts of extractives, and this can be explained by the method of sugarcane processing that can extract some part of the extractives. These results are in agreement to those found in the literature (Hemmasi et al., 2011).

3.4 Physical and Mechanical Properties

3.4.1 Density and Internal Bond

Most of the properties are directly affected by density and moisture, because of this, they are important properties that ensure fairness among treatments, thus allowing comparing the average values of physical and mechanical properties. According to Iwakiri, (2005) all the particleboards were classified as medium density (density between 0.59 and 0.80 g.cm⁻³). The moisture ranged from 8.5 to 9.0%. Both those properties showed a slight variation of the mean values, except the commercial boards from China, which showed higher density (Figure 5).

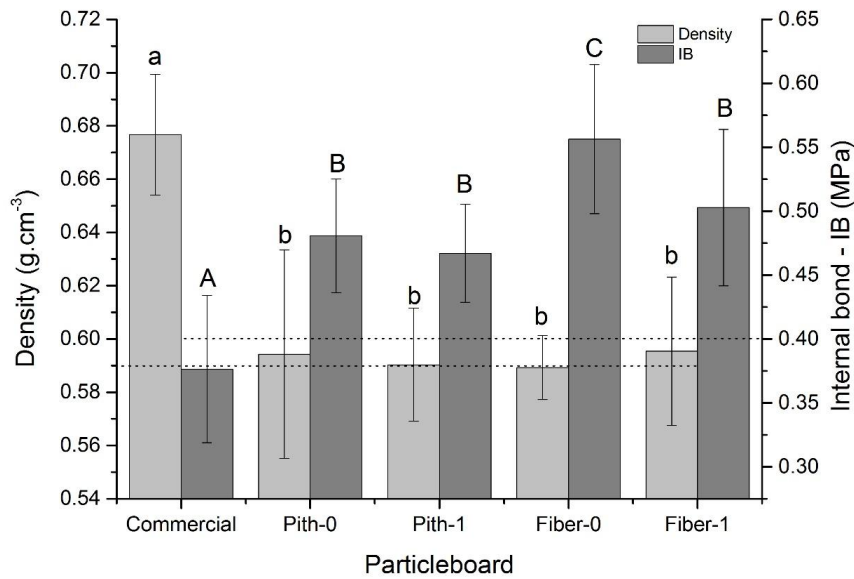


Figure 5 Histogram of the mean values for density, and internal bond (IB). Bars indicate standard deviation. Lines indicate minimum values for medium density particleboards and minimum standard values for IB. Same letters indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

All treatments showed best performances for IB, than the commercial SCB particleboards from China. The particleboards made only with fibers showed high mean values than those made with pith. The inclusion of NCC decreased the mean values for boards made only with fibers. Hydroxyls NCC groups probably react faster with UF leaving less available reacting groups for the SCB particles, which could explain the worst performance of Fiber-1. The NCCs inclusion increase the viscosity and this can hinder the adhesive penetration in the cell wall or voids, leading to less hooks formation. Considering that all the effects of distribution, dispersion and viscosity of the adhesive are very low, the reactivity between NCC and UF could dominate the

process. One suggestion is to increase the relative concentration of functional groups in the formulation of the adhesive, to compensate this reaction effect with the NCC.

The fibers and pith of both materials were classified according to sieve sizes however, after passing through the sieves it was possible to see that the particles from pith were smaller than from fibers. This means that they have more surface area for the adhesive cover, which could explain the lower mean values for particleboards made with pith in the core. According to Driemeier et al. (2011) for the non-spherical particles, the smaller projections are of the dimension effectively controlled by sieves.

Mahrtdt et al. (2015) investigated the effects of MFC addition in particleboards. The MFC was mixed with UF in two proportions (0 and 5%), and they found different results. The MFC inclusion increased the IB mean values of the particleboards (30% more). The better performance for IB was attributed to the distribution of the adhesive, because a higher fraction of adhesive + MFC was available for bond-line formation, and a larger part of the wooden particles could be covered with the adhesive.

Widyorini et al. (2005) studied binderless particleboards made of pith and fibers. They found that bagasse pith particles provided better IB than rind particles. According to them, bagasse pith particles were more easily deformed than bagasse rind particles, enlarging the bonding contact area.

The ANSI A208.1 (ANS, 1993) provides minimum IB values of 0.40 MPa for UF adhesive (M1 class). All particleboards meet the minimum requirements, except the commercial boards from China.

3.4.2 Modulus of elasticity (MOE) and modulus of rupture (MOR)

The best performances for MOE and MOR were found for commercial

boards (Figure 6). The NCC inclusion did not affected the MOE and MOR, since the particleboards with NCC inclusion were statistically equal to those without NCC. No statistical difference were observed among the particleboards made with pith and fiber for MOR and MOE.

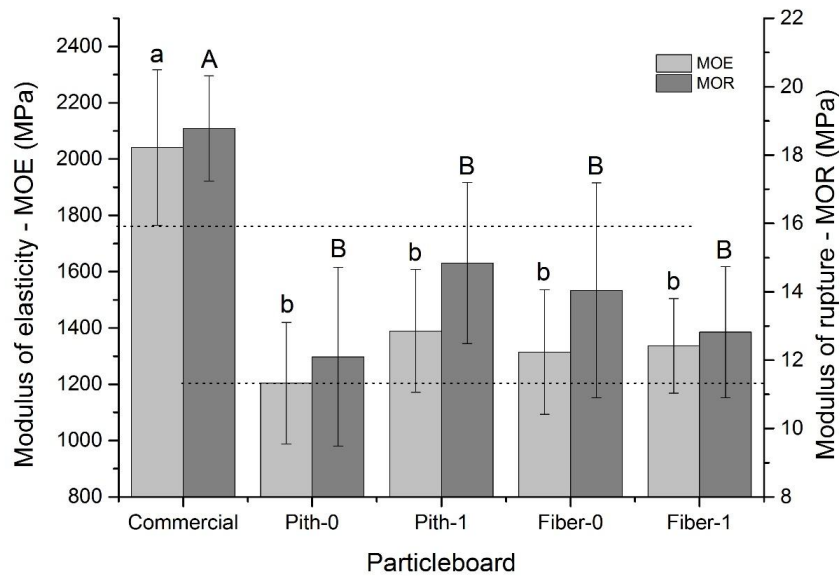


Figure 6 Histogram of the mean values for MOE, and MOR. Bars indicate standard deviation. Lines indicate minimum standard values for MOE and MOR. Same letters indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

It was possible to see that commercial particleboards used longer particles, which can explain the best performance for the mechanical properties. The sliver particles were not measured, although, it was clearly visible that the length of the particles from commercial particleboards were longer than the particles used in the present work (Figure 7). Increasing the length, it is possible

to increase the slenderness ratio (length/thickness). Higher slenderness ratio increases the bending properties (Iwakiri, 2005; Lee et al., 2006). Associate to this, the commercial particleboards showed higher density, which is directly related with the mechanical properties (Wu, 2001).



Figure 7 Commercial SCB particleboards from China with longer particles (in the right) and SCB particleboards made in the present work (in the left)

Widyorini et al. (2005) studied binderless particleboards made of pith and fibers. They found that bagasse pith particles provided better board properties (MOE and MOR) than rind particles. According to them, bagasse pith particles were more easily deformed than bagasse rind particles, enlarging the bonding contact area. They also found no difference for TS.

Lee and Mariatti, (2008) studied the addition of particles from pith and fibers in polyester composites. The composites using particle fibers had shown better mechanical (MOE and MOR) and physical properties (WA) than those made with pith particles.

ANSI A208.1 (ANS, 1993) stipulates the average minimum values of 1764 MPa for MOE and 11.3 for MOR, for particleboards/chipboards made with urea formaldehyde (class M1). Just the commercial boards meet the minimum

requirements for MOE, although all particleboards meet the minimum requirements for MOR.

Atta-Obeng et al. (2012) studied the effect of MCC inclusion in particleboards made with two species: sweetgum and pine. The MCC was applied in PF adhesive in two proportions: 0 and 10%. The MCC inclusion had a negative effect in the particleboards mechanical properties (MOE and MOR), decreasing the mean values. Probably the mechanical properties decreased because it used a high amount of MCC, which could agglomerate and harm the dispersion that affecting the properties of the particleboards. The surface area could also be related to this effect. Similar with this study, the MCC can react preferentially with the PF due the higher surface area, decreasing the sites for particles bond.

Veigel et al. (2012) investigated the effect of cellulose nanofibrils (CNF) inclusion in particleboards. They applied the CNF in the UF using different proportions (0, 1 and 3%). Better mechanical performance (MOR and IB) was found using 1% of CNF however, using 3% of CNF showed lower mean values than the control. The author suggested that this lower mean values using 3%, was not directly induced by higher CNF content, but could be explained by the long cold drying time (12 hours) of the glue and wood particles prior to hot pressing.

3.4.3 Water absorption (WA) and thickness swelling (TS)

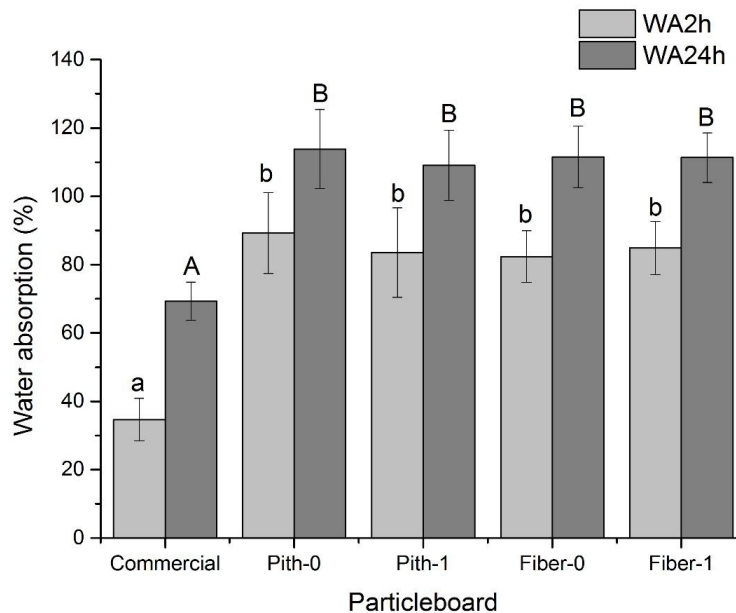


Figure 8 Histogram of the mean values for the water absorption after 2 hours (WA2h) and water absorption after 24 hours (WA24h). Bars indicate standard deviation. Same letters indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

The commercial boards from China showed the lowest mean values for WA2h and WA24h (Figure 8). As mentioned before, the particles of China particleboards were higher, which decrease the superficial area and offer less site for hydrogen bonds between water and fibers. The adhesive type and the adhesive proportion have a huge influence in these properties however, there is no information about the adhesive (kind and proportion) used in those SCB particleboards from China.

The NCC inclusion did not affect these properties (WA2h and WA24h), neither the materials (pith and fiber).

The aminomethylene linkage present in the UF adhesives is susceptible to hydrolysis, which means that the UF has low water and weather resistance. This explain the higher WA of particleboards made with UF. The water can cause swelling, which can induce a movement of the structural components of the MDP, breaking the bonding among resin and wood surface sites due to mechanical forces and stresses (Dunky, 1998; Dunky, 2004; Liu et al., 2008), creating more voids for water entrance. Nevertheless, UF is still used for particleboard production because it is indicate for internal purposes. Furthermore, it is easy to manipulate and it is cheaper when compared with others adhesives, as PF.

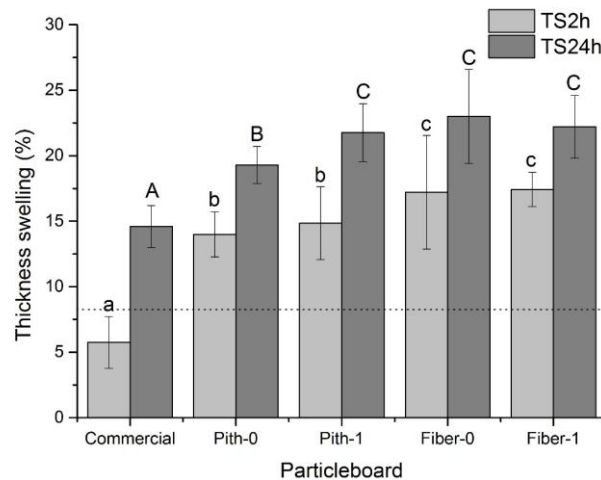


Figure 9 Histogram of the mean values for the thickness swelling after 2 hours (TS2h), thickness swelling after 24 hours (TS24h). Bars indicate standard deviation. Line indicates maximum standard values for TS24h. Same letters indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

The commercial boards from China showed the lowest mean values for TS2h and TS24h (Figure 9). The NCC inclusion increased the TS24h for pith-1. No effect was found with NCC inclusion for TS2h for both materials (pith and fiber). Pith and fiber showed similar amounts of hemicellulose, which could offer more hydroxyls groups for water sorption, but cellulose content also plays as a major role (Mohanty et al., 2000). Thus, materials with more cellulose tends to show higher dimension instability, because the water can enter between the cellulose chains causing their distancing and swelling. The particleboards made with pith showed higher dimension stability because the pith particles showed less cellulose and higher amount of lignin that it is hydrophobic.

Doost-hoseini et al. (2014) studied SCB particleboards with 0.5 g.cm^{-3} of density and found higher mean values for WA2h (110%), WA24h (140%), and similar results for TS2h (16%) and TS24h (19%). Since they used a similar adhesive proportion (12%) probably the WA was higher due the lower density, which means more voids, where the water could enter. Atta-Obeng et al. (2012) studied the effect of MCC inclusion in particleboards made with two species: sweetgum and pine. The MCC was applied in PF adhesive in two proportions: 0 and 10%. The MCC inclusion had a negative effect in the particleboards properties (TS2h and TS24h), increasing the mean values. The authors, attributed this to more available hydroxyl groups for moisture sorption, and consequent thickness swelling, caused by MCC inclusion.

Veigel et al. (2012) investigated the effect of cellulose nanofibrils (CNF) inclusion in particleboards. They applied the CNF in the UF using different proportions (0, 1 and 3%). The TS mean value, decreased with 1% CNF inclusion however, with 3% of CNF increased and the mean value was higher than the control.

The ANSI A208.1 (ANS, 1993) establishes maximum values for only TS24h, that is 8% for chipboards produced with urea formaldehyde. Therefore, none of the MDPs produced showed lower values stipulated by the standard.

3.5 Thermal properties

The thermal conductivity (TC) and volume heat capacity (VCH) showed higher values for SCB particleboards from China (Figure 10). This can be explained by the density of those boards that were higher than the density of the particleboards produced in the present work (Bekhta and Dobrowolska, 2006).

The inclusion of NCC did not affected the TC and the VHC, neither the kind of material (pith and fiber). The particleboards produced in this work were statistically equal.

The relationships between TC and density were also observed by others authors: Bekhta and Dobrowolska, (2006) when evaluating the properties of wood-gypsum boards; Sampathrajan et al. (1992), studying properties of boards made from farm residues and Mesquita et al. (2016b) studying wood particleboards with coir and sisal inclusion.

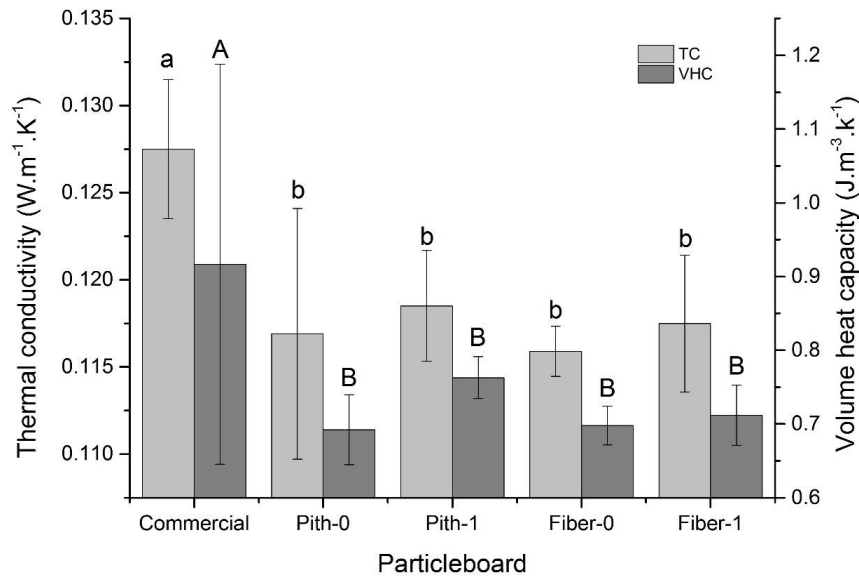


Figure 10 Histogram of the mean values for the thermal conductivity (TC) and volume heat capacity (VHC). Bars indicate standard deviation. Same letters indicate that there is no statistical difference by Scott Knott test ($\alpha=0.05$)

The TC is important property, especially when the particleboards will be used for insulator purpose. TC is an indicator of the value of a material as a heat insulator and lower TC is indicate for thermal insulation (Khedari et al., 2004).

4 CONCLUSIONS

The sugarcane bagasse particles can be used to produce particleboards for internal purposes. It was found that the separation in pith and fibers does not seem efficient, since the MDP properties of both were very similar. In addition, the two kinds of material showed very similar anatomical and chemical properties.

The particleboards produced in this study showed worst performance for mechanical (MOE and MOR) and physical (TS and WA) properties when compared with commercial SCB particleboards from China. Probably due the higher density of commercial particleboards.

The NCC inclusion in the faces of the particleboards did not showed efficient, since for most properties the particleboards with and without NCC were statistically equal. Probably less site links were available for the SCB to interact with the adhesive and because the NCCs increased the adhesive viscosity, this could make adhesive penetration more difficult.

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**APPENDIX A - SUPPLEMENTARY MATERIAL TO CHAPTER 3. NOT
INCLUDED IN THE MANUSCRIPT**



Figure C 1 Particles called fibers (in the left), longer than particles from the pith (in the right).