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Research Paper

# Evaluation of the incorporation of lignocellulose nanofibrils as sustainable adhesive replacement in medium density fiberboards

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ABSTRACT

Lignocellulose nanofibrils (LCNF) were used as a resin adhesive replacement in fiberboards. The physico-mechanical properties of the panels were affected by the LCNF content and the press temperature. At any given temperature, modulus of rupture (MOR) and internal bond strength (IB) showed a linear relationship with increasing LCNF. A temperature of 180 °C and 20% LCNF content were the optimum processing conditions giving MOR value of 12.1 MPa, close to the minimal recommended for commercial fiberboards. The IB, the thickness swelling, and modulus of elasticity met the standard values. The boards containing thermomechanical pulp (TMP), LCNF and borate minerals showed densities varying between 550 and 610 kg/m$^3$. However, the differential density between the core and surface was reduced as LCNF content increased. The three-dimensional LCNF network with an average particle diameter of 12 ± 3 nm promoted fibers bonding and allowed inter-fiber voids filling, as observed in SEM images. Press-temperatures greater than 180 °C could cause material degradation that lowered the mechanical properties of the fiberboards. LCNF displayed a good bonding ability and showed a promising perspective for adhesive replacement in fiberboards.

## 1. Introduction

Medium density fiberboard (MDF) is an engineered wood product composed of wood fibers generally combined with a synthetic resin and joined together under heat and pressure to form panels (Thoemen et al., 2010). Large-scale production of MDF began in the 1980s in both North America and Europe (André et al., 2008). Compared to high-density fiberboard, MDF has typically a density between 500 and 1000 kg/m$^3$ as specified by the ANSI A208.2-2016 (CPA, 2016). MDF is widely used in housing, furniture and cabinet industries because of its excellent surface and molding characteristics.

The fast growth in housing construction in rapidly developing countries in Asia, South America, Europe, and Russia, in addition to the housing recovery in the USA, affect the global production of MDF. In North America, the production of MDF is projected to reach five million m$^3$ in 2017 (WBPI, 2014). One of the biggest challenges facing this industry is to develop a more advanced knowledge of the relationships between the material used and the final product quality. Bonding of wood fibers is a key issue affecting the quality of fiberboards. Formaldehyde-based resins principally, urea-formaldehyde and phenol-formaldehyde are primary adhesives used in wood composite manufacturing because of their low cost. However, formaldehyde, a suspected human carcinogen, is emitted from fiberboard both during manufacturing and during use. This emission is a serious issue to human health (Doosthoseini et al., 2013; Kouchaki-Penchah et al., 2016). In addition, volatile organic compounds (VOC) concentration in the boards generally cause eye and respiratory irritation, irritability, inability to concentrate, and sleepiness. The climate of environmental awareness and concern that prevail are obstacles to their acceptance by the public. In July 2016, the US Environmental Protection Agency (EPA) finalized a rule to reduce exposure to formaldehyde vapor emission from certain wood materials produced domestically or imported into the United States. This first nationwide standardization, effective on May 2017 included formaldehyde emission standards applicable to hardwood, plywood, and MDF. For MDF, a reduction of the formaldehyde emission to 0.11 ppm, and to 0.13 ppm for thin MDF was

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adopted (EPA, 2016), although the compliance date for the emission standard and labelling has been delayed from December 2017 to March 2018. In the present rule the EPA standard has conserved the formaldehyde emission rate previously required in the phase two of the California Air Resource Board's Airborne Toxic Control Measure (CARB-ATCM) adopted by the Environmental Protection Agency in January 2011 and 2012 for MDF and thin-MDF (≤8 mm), respectively. The phase two CARB-ATCM reduced the standard formaldehyde emission that cannot be exceeded and was formerly fixed at 0.21 ppm for both MDF and thin-MDF in the phase one adopted in July 2009 (California Environmental Protection Agency). Compared to Europe, the European Panel Federation, since 2006 has made obligatory an emission class E1 (≤0.1 ppm) for wood panels. This standard was, however, effective since 1985 in some European countries such as Austria, Sweden, Denmark and Germany, this latter, since 2003 reducing the mandatory emission to 0.03 ppm, which is almost equal to the Japanese emission class F-star 4 (F****) emission standard (Schwab et al., 2012).

Melamine generally combined with urea-formaldehyde has been commonly used to reduce the formaldehyde emission in the panel. However, this thermosetting material reduces the recyclability of the panel and is an additional environmental issue. Kim and Kim (2005) have demonstrated that urea-formaldehyde resins showed a desiccator value of 7.05 ppm and a perforator value of 12.1 mg/100g-panel. The melamine formaldehyde exhibited a desiccator value of 0.6 ppm and a perforation value of 2.88 mg/100g-panel. These values were significantly higher when compared to the EPA recommendations (EPA, 2016).

Developing and using natural adhesives for wood composite bonding have been a topic of considerable research and interest in recent years. Schopper and Kharazipour (2006) proposed the use of a whey protein based adhesive in the production of medium density fiberboards. Pizzi (2006) reviewed different strategies used for eco-friendly wood bonding. The use of tannins, lignin carbohydrates, unsaturated oils as well as wood welding have been described as eco-friendly existing methods used to reduce formaldehyde and VOC.

Self-bonded and all-lignocellulosic fiberboards also gained attention in the literature (Arévalo and Peijs, 2016). In order to obtain acceptable bonding, pretreatments are generally applied on the fiber surface. Authors such as Zhang et al. (2015) and Li et al. (2015) proposed a chemical pretreatment using Fenton's reagent to activate the fiber surface and/or enzymatic pretreatment using laccase, which is widely found in plants and fungi.

Lignin-containing CNF, commonly known as lignocellulose nanofibrils (LCNF) isolated directly from unbleached wood without pretreatments is a promising material (Morales et al., 2014) and is starting to gain more attention in bioresource engineering. Spence et al. (2011) had estimated the total energy cost during the isolation of cellulose nanofibrils (CNF) from bleached hardwood using 9 passes micrograinding to be $95/ton, providing a final cost of the material to be about $445/ton. The saving of the pulp bleaching step, the easy processing (Osong et al., 2013) and lower energy consumption make LCNF a low-cost material with the potential to be used on industrial scale. Similar to cellulose nanofibrils (CNF) obtained from bleached pulp, LCNF are branched and random filaments with diameters varying between 10 and 50 nm and length of several micrometers. Its structure and dimension confer it has a high aspect ratio and large surface area. In addition to their high strength and modulus, LCNF fibrils are biocompatible and can be functionalized. Previous studies of Kojima (Kojima et al., 2013) have evaluated the bonding effects of lignocellulose nanofiber (LCNF) when mixed with wood flour. They found that physical and mechanical properties of the wood flour board were significantly improved with an addition of LCNF. Similar results were obtained by the same authors when using LCNF as binder for fiberboards made from hardwood and softwood pulps (Kojima et al., 2016).

In addition to its potential natural bonding ability and its lower cost as compared to most common cellulose nanofibers generally isolated from bleached pulp, LCNF was considered as an adhesive replacement in our study. The optimal condition for preparing LCNF filled fiberboards having physico-mechanical properties close to commercial panels was investigated. The effect of LCNF used as binder and press temperature was evaluated. The effect of boric acid- borax mixture used to impart fire retardancy on the board's properties and as antifungal agents will not be assessed in the present work. ASTM 1037–1999 and ANSI A208.2-2016 (CPA, 2016) were used as reference for the characterization and evaluation of the board's properties. A particular focus will be given to panels obtained from the potentially best processing conditions.

## 2. Materials and methods

### 2.1. Lignocellulose nanofibrils (LCNF) production and characterization

Unbleached never-dried thermomechanical pulp (TMP) was prepared through a three-stage atmospheric refining process of uncooked mixed softwood at 35 wt.% consistency as detailed in our previous work (Diop et al., 2017) using a two-disc Sprout Bauer pulp refiner (Andritz Sprout-Bauer, Inc., PA, U.S.A.). The average dimensions of the TMP fiber used for both the production of fiberboards and LCNF were measured using TechPap MorFi fiber analyzer (Saint-Martin-d'Hères, France) and were 552 μm in length and 37.1 μm in diameter with 78% fines (fines%). The percent of fines was determined from MorFi analysis and was defined as the percent of particles with diameter less than 0.2 mm present in the pulp.

A Super masscolloider (model MKZB15-50J Masuko Sangyo Co. Ltd, Japan) was used for cellulose fibers micrograinding at 2200 rpm. The clearance between the stationary and rotary grinding discs was set to −20 μm once zero gap was determined. Raw material slurry at 3% solids consistency was recirculated through the system allowing multiple refining phases. Fractions of the slurry were collected every 30 min within 100 min and analyzed using a TechPap MorFi fiber analyzer (Saint-Martin-d'Hères, France) to determine the fines amount (fines%). 100% fines were obtained after 1 h in the LCNF suspension that was cold stored at 4 °C.

The microstructure of LCNF was analyzed using a tabletop scanning electron microscope (SEM) (Hitachi TM3000, Japan) at different magnifications at an accelerating voltage of 15 kV. The diameter of the nanofibrils was determined from a minimum of 100 measurements from the transmission electron microscope (TEM) (CM10 TEM, Philips, Amsterdam, Netherlands) images obtained from a drop of 0.1 wt.% LCNF suspension negatively stained using 1% uranyl acetate solution. Digital image analysis software (ImageJ) was used to measure fiber diameters.

### 2.2. Fiberboard production and characterization

A wet forming process was used to produce adhesive-free fiberboards using never-dried TMP with targeted densities fixed between 550 and 650 kg/m$^3$. TMP was homogeneously mixed with LCNF and a designed amount of borate minerals using a 4.73 L mixer (Kitchenaid, Benton Harbor, MI). A 5% (w: v, considering the total liquid volume in the unpressed mat) mixture of borax and boric acid (1:1, w: w) was used as antifungals and concomitantly imparted a fire retardant property to the final boards. The formulations containing between 970–1000% water with respectively 15, 20, and 25 wt.% (dry basis) of LCNF were manually placed in a rectangular forming box (12 × 12 × 6 cm) placed on a screen wire mesh that retained the materials and cold-pressed at 490 kPa at room temperature. Cold pressing consolidates the particles and dewaters the mixture, allowing a removal of about 88% of the initial free water. The clear filtrate obtained, indicated that most of the particles were trapped in the mat and the amount of potential LCNF lost during the pressing was considered negligible. The pressed mat (with moisture content varying between