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International Journal of Adhesion & Adhesives

journal homepage: www.elsevier.com/locate/ijadhadh



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# Tensile shear strength of wood bonded with urea-formaldehyde with different amounts of microfibrillated cellulose

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#### ARTICLE INFO

*Article history:* Accepted 8 April 2015 Available online 18 April 2015

Keywords: Microfibrillated cellulose Wood Urea-formaldehyde Tensile shear strength Lap-joint test

#### ABSTRACT

Urea–formaldehyde (UF) adhesive mixtures with a 5% suspension of microfibrillated cellulose (MFC) at 0.5, 1, 3, and 5 wt% loading levels based on the solid weight (62.4%) of the UF adhesive were prepared. Beech lamellas with dimensions of 5 mm  $\times$  20 mm  $\times$  150 mm were prepared from beech lumbers using a planer saw. The UF adhesive (E0 class) was mixed with the MFC using a magnetic stirrer to achieve a proper distribution of the MFC in the UF adhesive. The tensile shear strength of single lap-joint specimens bonded with UF adhesive containing MFC was determined in accordance with EN 205 (2003). The specimens bonded with UF adhesive containing the MFC showed better tensile shear strengths as compared to the control. As compared to the control specimens, the tensile shear strength of the specimens increased by 5.7% as 3 wt% of the MFC was incorporated into the UF adhesive. However, a further increment in the MFC content up to 5 wt% decreased the tensile shear strength of the specimens). The MFCs were well dispersed in the UF resin and were cross-linked to form a network to reinforce the bond line, improving bonding performance.

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### 1. Introduction

Plant fibers, which are abundant renewable resources, exhibit high specific strength and stiffness, and industrial use of plant fibers as fillers in composites have attracted much interest. Plant fibers are converted into nanofibrous forms by chemical and mechanical treatments. One type of such nanofibers is called microfibrillated cellulose (MFC), which can be obtained by a high pressure homogenizing treatment [1,2].

The MFC was introduced in 1983 [3]. These homogenization passes make strands of nanocellulose fibrils that are used for reinforcement in matrix materials. The properties of MFC have been previously reviewed by Siro and Plackett [4]. The MFC consists of moderately degraded long fibrils that have greatly expanded surface area. Typically, traditional MFC consists of cellulose microfibril aggregates with a diameter ranging from 20 to even 100 nm with a length of several micrometers, rather than single nanoscale microfibrils [5]. Siro and Plackett [4] have reviewed the production mechanisms and properties of MFC and reported the

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http://dx.doi.org/10.1016/j.ijadhadh.2015.04.002 0143-7496/© 2015 Elsevier Ltd. All rights reserved. following routes of treatments for the production of MFC; mechanical: refining and high-pressure homogenization, cryocrushing and grinding. Pretreatments: alkaline pretreatment; oxidative pretreatment; and enzymatic pretreatment.

Micro/nanofibrils have garnered much attention for use in composites, coatings, adhesives, and film because of high specific surface areas, renewability and unique mechanical properties in the past two decades [6]. Microfibrillated cellulose has been shown to be a performance enhancer when added to composite materials such as polylactide foams reinforced with microfibrillated cellulose [7], nanocomposite films of MFC and melamine formaldehyde (MF) adhesive [8], nanocomposites from microfibrillated kraft pulp with a phenol–formaldehyde (PF) adhesive [9], PVA [10], acrylic and two types of epoxy composites reinforced with MFC [11]. All the composites were significantly stiffer and stronger than the unmodified composites. In all cases, it was reported that the cellulose microfibrils form an entangled network of fibers which is shown to be an adhesion mechanism in MFC reinforced composites.

The physical and mechanical properties of wood based composites depend on the adhesive characteristics. Urea–formaldehyde (UF) adhesive is commonly used in the manufacture of wood-based composites such as plywood, laminated veneer lumber, particleboard, and fiberboard. The advantages of UF adhesives are low cost, water solubility, easy use (under a wide variety of curing conditions), relatively low cure temperature, microorganisms resistance,

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low abrasion hardness, excellent thermal properties, and colorless (especially of the cured adhesive) [12]. However, the mechanical performance of adhesive bonds between the UF and wood is limited, in particular under humid conditions. Since the cured UF adhesive's elastic modulus is high, the deformation of the adhesive layer under mechanical loading is usually small. As a result, stress concentrations along the bond line of a wood adhesive joint are generated that reduce the overall strength of the joint [13].

Although UF adhesives are a widely used class of low-priced wood adhesives, it is well known for its pronounced brittleness and tendency to develop microcracks which limits the mechanical performance of UF-bonds [14,15]. The combination of low price and poor mechanical performance makes UF an ideal candidate for studying the effect of added filler [16]. There are limited studies on the properties of wood composites bonded with MFC filled adhesives [13,17]. The objective of this study was to improve the bond strength of a UF adhesive using MFC at different loading levels.

# 2. Materials and methods

### 2.1. Materials

Beech lumbers without defects (knot etc.) were supplied from a commercial lumber company in Chuncheon, South Korea. The lamellas with dimensions of 5 mm  $\times$  20 mm  $\times$  150 mm were prepared from beech lumbers using a planer saw. Prior to bonding, all the lamellas were planed in order to ensure smooth and flat surfaces. These were conditioned in a conditioning room at 20 °C and 65% relative humidity and allowed to reach an equilibrium moisture content of 12%. A commercial liquid UF adhesive (E0 class) with 62.4% solid content was used for the bonding of two types of beech wood. The commercial MFC (BiNFi-s) was supplied from Sugino Machine Ltd. in Japan. The crystallinity index and specific surface area of the MFC were 81.25% and 86 m<sup>2</sup>/g, respectively.

#### 2.2. Preparation of UF adhesive modified with MFC

The UF adhesive mixtures with 5% solution of MFC at 0.5%, 1%, 3%, and 5% loading levels of MFC based on the weight of solid adhesive were prepared. The MFC filled UF adhesive was mixed with a magnetic stirrer for 5 min to achieve a proper distribution of MFC in the UF adhesive. As a hardener 1% of ammonium chloride (NH<sub>4</sub>Cl) solution with 20 wt% solids content based on solids content of the UF adhesive was added to the mixture.

#### 2.3. Bonding of beech lamellas

The preparation of UF adhesive bonded single lap-joint specimens was carried out according to EN 205 [18]. Two beech lamellas were bonded together with the UF adhesive having different amounts of MFC solution and UF adhesive without MFC solution. The adhesive was applied on one surface  $(180 \text{ g/m}^2)$  of the lamella using a rubber roller. Assembling was always performed in parallel grain directions, whereby the lamella without adhesive was always on the bottom, to obtain equal penetration conditions for all specimens. All the specimens were pressed simultaneously in a hydraulic press at 140 °C and 1.6 MPa for 8 min. Prior to testing test specimens were conditioned at  $20 \pm 2$  °C and  $65 \pm 5\%$  relative humidity for one week. A total of 35 specimens, 7 for each type of UF adhesive mixture and control, were produced. The experimental design is presented in Table 1.

#### 2.4. Tensile shear strength test of single lap-joint specimens

Tensile shear strength test of the specimens was performed on an Instron universal testing machine with a speed of 1 mm/min according to EN 205 [18]. The tensile shear strength was calculated as follows:

$$\sigma_{\rm s} = F_{\rm max}/A = F_{\rm max}/a \cdot b(N/mm^2)$$

where  $\sigma_s$  is the tensile shear strength (N/mm<sup>2</sup>),  $F_{max}$  is the maximum load (N) observed, *A* is the bonding surface of the specimen in mm<sup>2</sup>, *a* is the width of bonded face, and *b* is the length of bonded face.

#### 2.5. Morphology

The morphology of the MFC was observed by field emission scanning electron microscope (SEM) (S-4800, Hitachi, Japan). The bond line of the UF adhesive containing the MFC between the lamellas of single lap-joint specimen was coated with carbon using carbon coater (JEE0400, JEOL Ltd., Japan) under vacuum ( $3 \times 10^{-4}$  Pa) for 1 s for the FE-SEM observation.

#### 3. Results and discussion

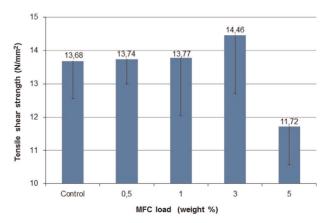
The results of tensile shear strength of lap-joint specimens are shown in Fig. 1. The tensile shear strength of the specimens slightly increased with the incorporation of MFC (0-1 wt%) while it considerably increased as the amount of MFC increased from 1 to 3 wt%. However, further addition of the MFC (5 wt%) into the UF adhesive decreased the tensile shear strength of the specimens. As compared to the control specimens, the tensile shear strength of the specimens increased by 5.7% as 3 wt% MFC (5% suspension

## Table 1

Composition and properties of UF adhesive mixtures.

Code of adhe- sive mixture	Solid content of UF (E0 class) adhesive (%)	Amount of MFC (wt% of solid adhesive)	Amount of hardener (NH <sub>4</sub> Cl) (wt% of solid adhesive)
A (control)	62.4	0	1
В	62.4	0.5	1
С	62.4	1	1
D	62.4	3	1
E	62.4	5	1

MFC: microfibrillated cellulose (5 wt% water suspension).



**Fig. 1.** Tensile shear strength of single lap-joint specimens bonded with UF adhesive containing the MFC (microfibrillated cellulose).

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