Effect of Cellulose Nanofibrils on the Bond Strength of Polyvinyl Acetate and Starch Adhesives for Wood

Wen Jiang,[a] Laura Tomppo,[b] Timo Pakarinen,[c] Juho A. Sirviö,[d] Henrikki Liimatainen,[d] and Antti Haapala [a,]*

Nanocellulose is a competitive reinforcement material for use in biocomposite structures and fibrous products. In this study, adhesive mixtures of dicarboxylic acid cellulose nanofibrils (CNF) were dispersed into commercial polyvinyl acetate (PVAc) and starch adhesives, which were applied to Norway spruce (*Picea abies*) to assess their performance in wood joining. Single-lap joints were prepared and tested with PVAc mixtures with 0 to 0.64 wt% CNF and starch glue mixtures containing 0 to 1.07 wt% CNF. CNF suspensions having three concentrations (0.64, 0.96, and 1.28%) were compared. The results showed that the optimum amount of CNF, 0.48% suspensions, added to PVAc increased the average lap joint strength (EN 205:2003) by 74.5% when compared to control specimens with pure PVAc. Correspondingly, 0.96% CNF suspensions also enhanced the strength of starch adhesive by 34.5%. Lower and higher CNF concentrations showed clearly inferior performance.

Keywords: Adhesion; Nanocellulose; Nanofibril; PVAc; Starch glue; Wood joining

Contact information: a: Wood Materials Science, School of Forestry, University of Eastern Finland, School of Forestry, 80101 Joensuu, Finland; b: Department of Applied Physics, University of Eastern Finland, 70211 Kuopio, Finland; c: Karelia University of Applied Sciences, 80200 Joensuu, Finland; d: Fibre and Particle Engineering, University of Oulu, 90014 Oulu, Finland;
** Corresponding author: antti.haapala@uef.fi*

INTRODUCTION

Adhesives play an important role in the wood industry; they are used to achieve desired strength and stiffness by bonding wood elements. The array of applications is vast and covers many length scales, from fibers and small particles to flakes, veneers, laminates, and lumbers (Stark *et al.* 2010). Adhesives also add value to other residues such as small logs, chips, and less desired tree species used in construction materials.

The properties of wooden products are impacted by many factors, including qualities of wood adhesives and bonding process, *e.g.*, wetting of substrate surfaces, heat, and pressure on the bond line, and drying time (Frihart 2013). Functional fibrous fillers are used in adhesive formulations aiming at higher strength, improving wetting of surfaces, seam rigidity, *etc.* Studies on composite materials show that materials are stronger and stiffer in fibrous form (Chawla 1998). Reinforcing fibers such as glass and carbon fibers, polymer fiber blends, and inorganic nanoparticles have been studied as additives for modifying wood adhesives; their usage introduces some environmental and sustainability issues in otherwise green materials (Singha and Thakur 2008; Wang *et al.* 2011). Cellulosic fibers as renewable materials have gained attention as potential reinforcements for adhesives, but practical applications remain scarce. A number of recent studies have investigated nanocellulose applications in polymers and composites, but there are few related to adhesives for wood bonding.

Recent studies have focused on the reinforcement of urea formaldehyde (UF), melamine formaldehyde (MF), phenol formaldehyde (PF), and polyvinyl acetate (PVAc) with nanocellulose additions. These adhesives have wide applications in producing wood panels (Dunky and Pizzi 2002; Frihart 2013). López-Suevos *et al.* (2010) used cellulose nanofibrils with the addition of acids and sodium hydroxide to reinforce PVAc-latex adhesives, which showed excellent heat resistance properties of the produced panels. Veigel *et al.* (2011) prepared (2,2,6,6-tera-methylpiperidin-1-yl)-oxyl (TEMPO) and sodium chlorite oxidized CNFs as reinforcement materials for UF adhesives, which showed benefits for double cantilever beam specimens even though the nanofibrils were limited to 2 wt% due to increased viscosity of the adhesives. Atta-Obeng *et al.* (2013) investigated PF adhesive reinforcement with microcrystalline cellulose at loading rates up to 10 wt% without addition of any other chemicals, obtaining a significant increase in shear strength and decrease in curing temperature. Liu *et al.* (2014) reinforced PF with three types of nanocelluloses: laboratory-processed nanofibrils, commercial nanofibrils, and nanocrystals. They reported that the cellulose nanomaterials have a positive impact on interlocking formation between woods and they can improve the mechanical properties of adhesives. Kwon *et al.* (2015) announced a similar increase of the tensile shear strength of wood bonded with microfibrillated cellulose and the reinforcing of UF adhesives with an ammonium chloride solution as the hardener. Chaabouni and Boufi (2017) investigated the influence of CNF additions on the properties of waterborne polyvinyl acetate (PVA) adhesive. They used very high CNF addition rates (up to 10 wt%) and observed significant benefits in shear strength and water resistance performance. However, such a high mass fraction of CNFs raises questions on economic feasibility in commercial applications.

There are several papers on the application of nanocellulose-reinforced adhesives in wood panels. Veigel *et al.* (2012) published another study on UF and melamine-urea-formaldehyde (MUF) adhesives in a matrix of adhesive resins and CNF while using ammonium nitrate in water solutions as a hardener for particleboard and oriented strand board, which showed increased values of fracture energy and toughness (Veigel *et al.* 2012). Cui *et al.* (2015) similarly reported improved strength in particleboard when a tannin cross-linked adhesive system was supported by microfibrillar cellulose in the presence of solid paraformaldehyde as the hardener. Mahrdt *et al.* (2016) published a follow-up paper to Veigel *et al.* (2012), showing the effect of adding microfibrillar cellulose and aqueous ammonium nitrate as a hardener to UF resins for particleboards. The addition improved the board strength due to the more even adhesive distribution and increasing adhesive toughness. Leng *et al.* (2017) investigated the effects of density, CNF addition ratio, pressing method and particle size on the bending properties of wet-formed particleboard.

Although synthetic adhesives are currently widely used in the wood industry, natural adhesives are gaining attention because of the environmental issues associated with harmful emissions from formaldehyde and volatile organic compounds (VOCs) as well as the future scarcity of petrochemicals. However, natural adhesives such as starch, lignin, and protein need modification to achieve comparable properties to commercial adhesives. Fiber reinforcement is a potential and reliable way to improve mechanical properties of natural adhesives and use the amount of adhesive needed in joints. Hence, nanocellulose has many advantages for this application area: it is renewable, biodegradable, has low density, has good mechanical properties, and is non-toxic.

In this study, the influence of dicarboxylic acid cellulose nanofibrils (CNF) on the mechanical properties of PVAc and starch glue of low dosages (g/m^2 in bondline) in dry and wet conditions were investigated. The water dispersions of CNF and adhesives were used in low concentrations and mechanically blended to adhesives with no chemical additives. The main objective of this study was to improve the toughness of different adhesives with various CNF loadings.

EXPERIMENTAL

Materials

Wood material

Industrially dried Norway spruce (*Picea abies*) wood was chosen for this study because it is one of the most commonly used tree species in Finland. Beech is suggested for the tensile strength test because it has a high average strength of 670 to 770 kg/m^3 and is known to give an average shear strength of 10 MPa to the bond layers as a result (Konnerth *et al.* 2016). Spruce has an average density of 470 kg/m^3 with a lower bond line shear strength (7.5 MPa) than that of beech (Konnerth *et al.* 2006, 2016). Prefabricated wood was conditioned in a climate chamber at 20 °C and with a relative humidity of 65% for two weeks before making the specimen. The best wood samples were selected and the design of experiments was controlled with double-blind sampling of preforms between treatments.

Adhesives

Commercial adhesives used for this study were Kiilto B3 modified polyvinyl acetate (PVAc or PVA) glue (Kiilto Ltd, Tampere, Finland) and Emcol UK-H5 modified starch power (Haarla Ltd, Tampere, Finland). Emcol UK-H5 is a pregelatinized and crosslinked sodium salt of a modified potato starch (carboxymethyl ether) readily soluble in cold water. Kiilto B3 is a type of PVAc-based water-borne adhesive with D3 class water resistance (applied in interior conditions with frequent short-term exposure to running or condensed water and/or to heavy exposure to high humidity, exterior applications not exposed to weather, according to EN 204 (2001).

Nanofibrillated cellulose

Cellulose nanofibrils used in this study were produced from birch kraft pulp (*Betula verrucosa* and *pendula*) using a two-step oxidation method based on sequential periodate-chlorite oxidation followed by mechanical homogenization, according to the previously reported method (Liimatainen *et al.* 2012). First, 9 g of hardwood cellulose pulp was oxidized with 7.38 g of sodium periodate (Sigma Aldrich, Munich, Germany) at 55 °C for 3 h in the absence of light and further reacted with 4.53 g of sodium chlorite (Sigma Aldrich) in a 1 M aqueous solution of acetic acid (Sigma Aldrich) for 48 h at room temperature. The obtained dicarboxyl acid cellulose (anionic charge content 1.75 mmol/g) was further nanofibrillated using a microfluidizer (Microfluidics M-110EH-30, Newton, MA, USA). The detailed characteristics of cellulose pulp and CNF are given by Liimatainen *et al.* (2012). The obtained CNF suspension with a concentration of 0.64%

was stored in sealed containers to avoid moisture evaporation and absorption from the environment.

Preparation of PVAc and Starch with CNF Addition

Three CNF suspensions (CNF-S) with a solid content of 0.64%, 0.96%, and 1.28% were mixed with adhesives at room temperature by a high-performance homogenizer (T25 ULTRA-TURRAX®, IKA-Werke, Staufen, Germany). Micronized CNF at 0.64% was further prepared to 0.96% (+50% dry matter) and 1.28% (+100% dry matter) CNF suspensions were attained by slow evaporation of water at 60°C. PVAc mixtures were obtained by maintaining a constant ratio of commercial PVAc products to CNF-S of 1:1 (w/w) (CNF concentration of each modified adhesive is shown in Table 1). Pure PVAc, both with and without deionized water, were used as references (PVA_0 and PVA_w, respectively). Starch glue mixtures were prepared by mixing dry starch flakes with water or with the three CNF suspensions at a ratio of 1:11 (w/w), forming suspensions with CNF contents of 0.53%, 0.80%, and 1.07%, as shown in Table 1. Starch powder mixed with deionized water (without CNF) was used as a reference ($Starch_0$).

Table 1. References and Adhesives Modified with Different CNF Suspensions

Sample	Adhesive System Composition	CNF Content, wt%	CNF Loading Level
PVA_0 (control)	Kiilto B3	0	0
PVA_w	Kiilto B3 + water	0	0
PVA032	Kiilto B3 + 0.64% CNF-S	0.32	1
PVA048	Kiilto B3 + 0.96% CNF-S	0.48	2
PVA064	Kiilto B3 + 1.28% CNF-S	0.64	3
$Starch_0$ (control)	Solid starch + water	0	0
Starch053	Solid starch + 0.64% CNF-S	0.53	1
Starch080	Solid starch + 0.96% CNF-S	0.80	2
Starch107	Solid starch + 1.28% CNF-S	1.07	3

* *Note*: PVAc and starch adhesives with the same CNF loading level mean that the same CNF suspensions were used.

Mechanical Test

Spruce sapwood was cut to 160 × 96 × 5.5 mm for use in the single-lap joint test. All wood surfaces were planed before bonding to obtain even roughness and minimize the effect of surface unevenness to adhesion strength, as proposed by Söğütlü (2017). The wood pieces were glued and pressed under a pressure of 0.5 MPa at ambient temperature for 20 mins for PVAc and for 1 h for starch glue. Glued samples were cut to the final sizes (150 × 20 × 11 mm), as shown in Fig. 1, which was suitable for the mechanical test. Finally, 50 g/m^2 of unmodified PVAc (PVA_0) and 100 g/m^2 of PVAc mixtures (PVA_w, PVA032, PVA048, and PVA064) were applied to the sample surfaces; for starch glues, 78 g/m^2 was applied directly. In this study, PVAc was used less than the required amounts from Kiilto Ltd (preferably 120 to 150 g/m^2) to better assess the influence of CNFs. The cellulose nanofibrils were included in both suspensions directly and the specimen was conditioned at 20 °C and at a relative humidity of 55% for 7 days before testing.

Bond strength was measured with the tensile shear strength of lap joints (EN 205 2003). Lap joint specimens were prepared and tested on a universal testing machine (Instron 3367, Glenview, IL, USA).

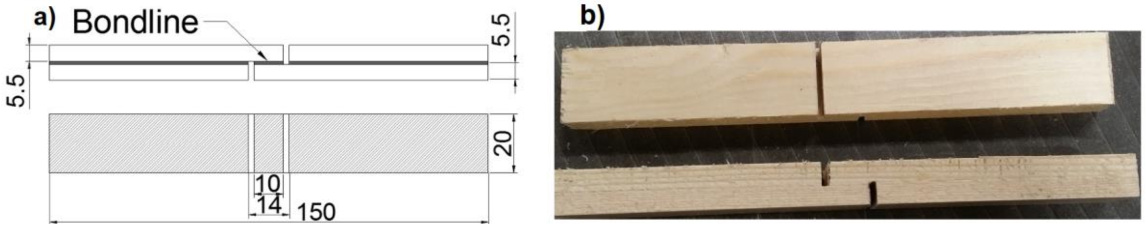

Fig. 1. (a) Diagram of single lap joint shear strength samples with adhesive bondline (dimensions in mm); (b) the lap joint specimen.

Ten specimens for PVAc and starch adhesives were selected based on external assessment, *e.g.* glue seam evenness and absence of knots. Lap-joint shear strength calculated from Eq. 1,

$$\tau = \frac{F_{max}}{A} = \frac{F_{max}}{a \times b} \qquad (1)$$

where τ is shear strength (N/mm^2), A is overlapped bonded surface area (mm^2), a is the length of the bonded surface (mm), and b is the width of the bonded surface (mm).

Statistical Analysis

All data from mechanical tests was analyzed by using a one-way ANOVA test and independent T-tests in SPSS software (IBM, Chicago, IL, USA). ANOVA was used for determining the significances of nanocellulose concentrations on the shear strength between and within the groups. There was a statistically significant difference in the mean strength of different adhesive groups when the significance (Sig.) value was smaller than 0.001. T-tests were additionally used to compare if every two adhesive groups were different from each other.

RESULTS AND DISCUSSION

Figure 2 shows the average shear strength of the two types of adhesive groups with 95% confidence interval values on the bars. As shown in Fig. 2a, the highest average lap joint shear strength of the PVAc groups was obtained from samples glued by PVA048 with a value of 10.47 N/mm^2, compared with the average strength of 6.00 N/mm^2 achieved from PVA$_0$ group. Compared with the control group PVA$_0$, PVAc mixed with water or CNF suspensions showed higher bond strength, with increases of 38.5% from PVA$_w$, 30.8% from PVA032, 74.5% from PVA048, and 6.5% from PVA064. Pure PVAc dried very quickly due to the small amount of glue used. However, the addition of both water and CNF suspensions to PVAc improved the ability to wet the wood surface with comparable bond strength (8.31 to 10.47 N/mm^2 from PVA$_w$, PVA032, and PVA048) to the average bond line shear strength of 7.5 MPa for spruce studied by Konnerth *et al.* (2006a,b). In addition, the amount of PVAc mixed with water and CNF was significantly lower than the manufacturer's instructions. This result suggests that it is possible to apply smaller doses of commercial PVAc products by mixing water or CNF suspensions used in this study.

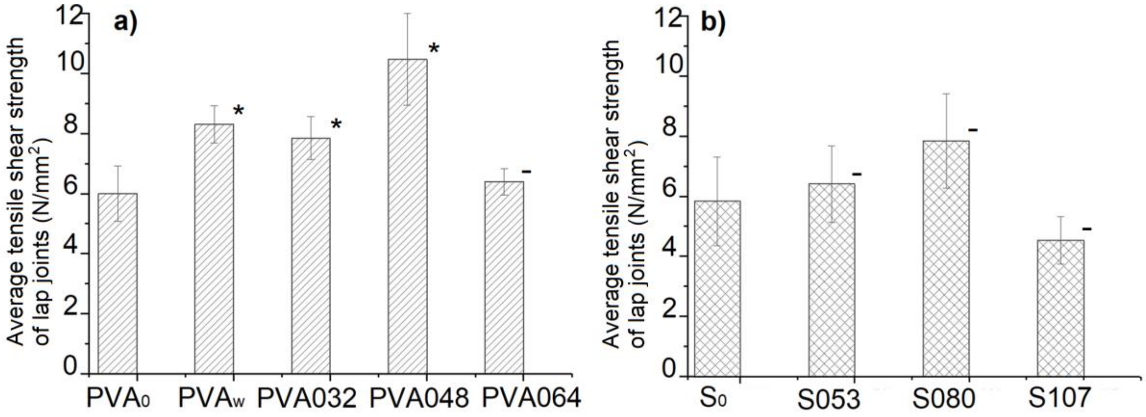

Fig. 2. The average tensile shear strength of single-lap joint specimens: (a) PVAc adhesives, (b) starch adhesives. The error bars indicate the standard deviation within the measurements in each group (N = 10 pcs). Data are presented as mean ± stdev. and analysed using a t-test indicated as * for $p < 0.01$ and – with no statistical significance in relation to references without CNFs.

Figure 2b demonstrates the relationship between increasing nanocellulose loadings and the bond strength of starch. The average strength values of the starch adhesives in single-lap joint tests indicated that the addition of 0.64% and 0.96% CNF suspensions increased the average bond strength by 9.9% and 34.5%, respectively, over starch glue alone. It was observed during the experiments that adhesives with higher CNF content were more viscous. The presence of 1.07 wt% CNF (1.28% CNF suspension used) in the starch mixtures led to a clearly lower bond strength resulting from the increased viscosity, which was also observed from PVA064 with a decreased amount of shear strength. Also, the highest shear strength occurred in the PVAc and starch groups with the same CNF loading levels (3) (Table 1). However, a severe problem appearing in starch adhesives was the high variation between the samples in the test group, which indicated that starch adhesives are not stable and the strength of specimens glued by starch adhesives may differ significantly.

The ANOVA results and significance (Sig.) values for the PVAc adhesives and starch adhesives in lap joint tests are shown in Table 2. The significance values for both were smaller than 0.05, indicating that there were statistically significant differences in the average strength of lap joint samples from both groups. The t-test by SPSS shows the significant influence of the addition of water and nanocellulose to PVAc adhesives on the adhesion strength.

Table 2. ANOVA and t-Test Results and Significance Values within Test Groups

ANOVA Test #	**F**	**Sig.**
Lap-joint test for PVAc adhesives	15.762	0.000
Lap-joint test for starch adhesives	4.328	0.015
t-Test Comparing CNF Treated to Reference Lap Joint Shear Strength	**t**	**Sig. (2-tailed)**
PVA_0, PVA_w	-4.165	0.001
PVA_0, PVA032	3.072	0.008
PVA_0, PVA048	-5.250	0.000
PVA_0, PVA064	-0.782	0.445
$Starch_0$, Starch053	-0.577	0.576
$Starch_0$, Starch080	-1.839	0.091
$Starch_0$, Starch107	1.513	0.156

Petković *et al*. (2017) showed recently how increasing the amount of water in PVAc adhesive increase also its hydrophilic properties, which in turn lowers the contact angle between PVAc and the surface (better wetting) and lowers the surface free energy. The relation of contact angle, wetting and surface energy has been reviewed *e.g.* by Yuan and Randall (2013). However, the t-test indicated no significant differences between samples of the starch adhesives because of the high variance of results, likely due to more challenging curing of starch in the presence of high moisture content and absence of heat treatment. The trend is in fair agreement with Qiao *et al*. (2014), who found similar behaviour in CMC addition to starch in plywood and the optimal benefit from only 0.375% CMC dose, while higher concentrations produced weaker bond strength.

In the previously reported studies, the shear strength in single-lap joint tests ranged from 4.5 to 7.5 MPa (PF adhesives added with microcrystalline cellulose by Atta-Obeng *et al.* 2013), from 6.7 to 8.5 MPa (*Hibiscus sabdariffa* fibre addition to UF adhesives by Singha and Thakur 2008), and from 11.7 to 14.5 MPa (UF with microfibrillated cellulose by Kwon *et al.* 2015). In this study, shear strength of modified PVAc adhesives, with the addition of water, 0.64% and 0.96% CNF suspensions, showed results from 7.85 to 10.47 MPa. The shear strength of starch was between 4.42 and 7.64 MPa, which was lower than that of PVAc from this study and synthetic adhesives from other studies. The addition of 0.96% CNF to starch also showed a good result of 7.64 MPa, which is comparable to the value of 7.5 MPa by Knonnerth *et al.* (2006) and the study on spruce bonded by a series of adhesives. Considering the unstable performance upon exposure to humidity and the long curing time, modified starch glue still must overcome technical problems before it is suitable as a wood adhesive.

Results of single lap joint tests with pure PVAc and starch both indicated that the addition of nanocellulose increased the shear strength, but the highest amount of added CNF decreased the strength. There are potentially two reinforcement mechanisms of nanocellulose for resins. The first mechanism is that the stiffness of CNF itself improves the toughness of the bond line when dispersing in the adhesives. On the other hand, the amounts of CNF addition were limited, as it caused an increasing viscosity of the adhesives that exhibited the wood penetrating ability. Veigel *et al.* (2011) also claimed that cellulose addition increased the viscosity of adhesives, which restricted the amount of nanofibrils that could be added. In this study, the highest amount of CNF in suspensions (1.28%) led to over-viscous systems that were less applicable to the wood surface as an even film and may have required a longer curing time, although the potential benefits of CNF dose and press time were not investigated nor optimized here.

Another potential reinforcement mechanism is the cross-linked network between the nanofibers and adhesive formulations, although we were unable to support this with experimental data. The strength of the bond layer was likely increased because of the crosslinking network from the methylene groups of PVAc molecules and the methylol groups of starch reacting with the hydroxyl groups from CNF. As noted earlier by Kwon *et al.* (2015), a cross-linked network between the methylol groups of the UF molecules and the hydroxyl groups of the cellulose was explained. Additionally, high amounts of water in both commercial PVAc adhesives and CNF suspensions led to a water-based environment for the adhesive mixtures with good surface wetting property along with a good bond strength hold by nanocellulose fibres. This was attributed by the hydrophilic property of the cellulose fibres. One can also speculate whether the yield stress distribution in lap joint was more evenly distributed along the bond line in the presence of CNFs, but this cannot be shown with the data.

Pictures of fracture samples and typical fracture modes of single lap joints are displayed in Fig. 3. All the samples used were selected and controlled with no knots or visible problems. The fracture of the samples observed in this study was in most cases as in mode (v) with cohesive failure and wood surface fracture at the same time (Fig. 3a). Different fracture circumstances may occur, as shown in Fig. 3b: (i) material under force; (ii) cohesive failure indicating that the adhesive may need strengthening; (iii) the possibility of interfacial failure due to poor interaction between materials and adhesives; and (iv) material failure occurring before the adhesives fracture (River *et al.* 1991; Comyn 1997). This was due to the adhesive layer being very thin, and close attachment caused by deep adhesive penetration and planed wood surfaces. Fracture modes (iii) and (iv) were never observed in this study, which means the bonding was sufficient and, with this regard, spruce was considered a suitable platform material for this test.

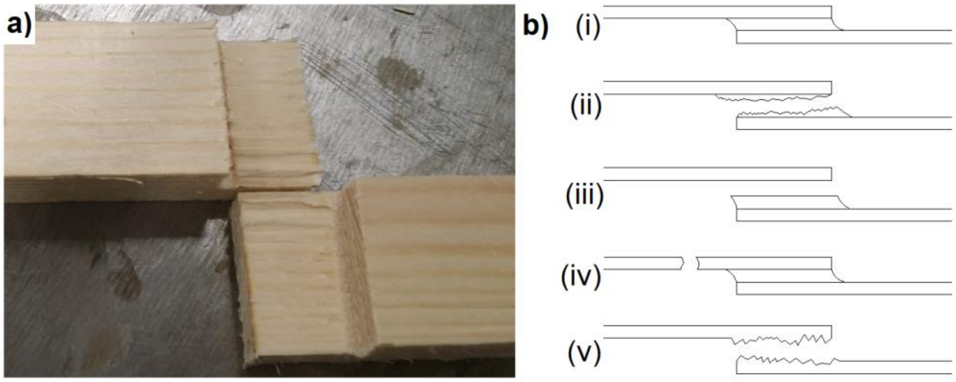

Fig. 3. (a) Fracture sample and (b) fracture modes of lap joints (modified from Comyn 1997).

CONCLUSIONS

1. The addition of water and CNF to PVAc adhesives increased the lap-joint strength by 6.5% to 74.5% compared with the pure PVAc used to bond similar specimens.
2. The addition of 0.96% CNF suspensions provided PVAc and starch adhesives with the highest bond strength, mainly contributed to the cross-linked network by the reaction between methylene groups of PVAc molecules and the methylol groups of starch with the hydroxyl groups from CNF.
3. The maximum concentrations of CNFs in adhesive mixes were limited due to an increased viscosity of the adhesives that exhibited the penetrating ability to the wood surfaces.
4. In addition, this study suggests that a good bonding of wood can be obtained with a significantly lower dose of commercial PVAc by adding water and CNF suspensions.

ACKNOWLEDGMENTS

The authors thank Haarla Ltd. for contributing materials for this study. Mr. Mikko Matveinen, Mr. Riku Tiira, and Mr. Hannu Tolvanen of Karelia University Applied

Sciences are gratefully acknowledged for their support in the sample preparation and testing. Teollisuusneuvos Heikki Väänänen's Fund is acknowledged for financial support.

REFERENCES CITED

Atta-Obeng, E., Via, B. K., Fasina, O., Auad, M. L., and Jiang, W. (2013). "Cellulose reinforcement of phenol formaldehyde: Characterization and chemometric elucidation," *Int. J. Composite Mater.* 3(3), 61-68. DOI: 10.5923/j.cmaterials.20130303.04

Chaabouni, O., and Boufi, S. (2017). "Cellulose nanofibrils/polyvinyl acetate nanocomposite adhesives with improved mechanical properties," *Carbohyd. Polym.* 156, 64-70. DOI: 10.1016/j.carbpol.2016.09.016

Chawla, K. K. (1998). "Reinforcements," in: *Composite Materials—Science and Engineering* (3rd Ed.), Springer, New York, pp. 7-72.

Comyn, J. (1997). *Adhesion Science*, the Royal Society of Chemistry, Cambridge, UK.

Cui, J., Lu, X., Zhou, X., Chrusciel, L., Deng, Y., Zhou, H., Zhu, S., and Brosse, N. (2015). "Enhancement of mechanical strength of particleboard using environmentally friendly pine (*Pinus pinaster* L.) tannin adhesives with cellulose nanofibres," *Ann. For. Sci.* 72, 27-32. DOI: 10.1007/s13595-014-0392-2

Dunky, M., and Pizzi, A. (2002). "Wood adhesives," in: *Adhesion Science and Engineering: Surfaces, Chemistry and Applications*, D. A. Dillard and A. V. Pocius (eds.), Elsevier Science B.V., Amsterdam, Netherlands, pp. 1039-1103.

EN 204 (2001). "Classification of thermoplastic wood adhesives for non-structural applications," European Committee for Standardization, Brussels, Belgium.

EN 205 (2003). "Adhesive—Wood adhesives for non-structural applications—determination of tensile shear strength of lap joints," European Committee for Standardization, Brussels, Belgium.

Frihart, C. R. (2013). "Wood adhesion and adhesives," in: *Handbook of Wood Chemistry and Wood Composites* (2nd Ed.), R. M. Rowell (ed.), CRC Press, Boca Raton, FL, USA, pp. 255-313., DOI: 10.1016/j.jclepro.2015.07.070

Konnerth, J., Gindl, W., Harm, M., and Müller, U. (2006). "Comparing dry bond strength of spruce and beech wood glued with different adhesives by means of scarf- and lap joint testing method," *Holz als Roh- und Werkstoff* 64(4), 269-271.

Konnerth, J., Kluge, M., Schweizer, G., Miljković, M., and Gindl-Altmutter, W. (2016). "Survey of selected adhesive bonding properties of nine European softwood and hardwood species," *Holz als Roh- und Werkstoff* 74(6), 809-819.

Kwon, J. H., Lee, S., Ayrilmis, N., and Han, T. H. (2015). "Tensile shear strength of wood bonded with urea-formaldehyde with different amounts of microfibrillated cellulose," *Int. J. Adhes. Adhes.* 60, 88-91. DOI: 10.1016/j.ijadhadh.2015.04.002

Leng, W., Hunt, J., and Tajvidi, M. (2017). "Effects of density, cellulose nanofibrils addition ratio, pressing method, and particle size on the bending properties of wet-formed particleboard," *BioResources* 12(3), 4986-5000. DOI: 10.15376/biores.12.324986-5000

Liimatainen, H., Visanko, M., Sirviö, J. A., Hormi, O. E. O., and Niinimäki, J. (2012). "Enhancement of the nanofibrillation of wood cellulose through sequential periodate-chlorite oxidation," *Biomacromolecules* 13, 1592-1597. DOI: 10.1021/bm300319m

Liu, C., Zhang, Y., Wang, S., Meng, Y., and Hosseinaei, O. (2014). "Micromechanical properties of the interphase in cellulose nanofibre-reinforced phenol formaldehyde bondlines," *BioResources* 9(3), 5529-5541. DOI: 10.15376/biores.9.3.5529-5541

López-Suevos, F., Eyholzer, C., Bordeanu, N., and Richter, K. (2010). "DMA analysis and wood bonding of PVAc latex reinforced with cellulose nanofibrils," *Cellulose* 17(2), 387-398. DOI: 10.1007/s10570-010-9396-8

Mahrdt, E., Pinkl, S., Schmidberger, C., van Herwijnen, H. W. G., Veigel, S., and Gindl-Altmutter, W. (2016). "Effect of addition of microfibrillated cellulose to urea-formaldehyde on selected characteristics and distribution in particle board," *Cellulose* 23, 571-580. DOI: 10.1007/s10570-015-0818-5

Petković, G., Rožić, M., Vukoje, M., and Pasanec Preprotić, S. (2017). "Interactions in polyvinyil acetate - paper adhesive joint and influence on its adhesion parameters," *Annals of Faculty Engineering Hunedoara - International Journal of Engineering*, XV, 1; 35-42.

Qiao, Z., Gu, J., Zuo, Y., Tan, H., and Zhang, Y. (2014). "The effect of carboxymethyl cellulose addition on the properties of starch-based wood adhesive," *BioResources* 9(4), 6117-6129.

River, B. H., Vick, C. B., and Gillespie, R. H. (1991). "Wood as an adherent," in: *Treatise on Adhesion and Adhesives,* J. D. Minford (ed.), Marcel Dekker, NYC, USA, 7, 1-230.

Singha, A. S., and Thakur, V. K. (2008). "Effect of fibre loading on urea-formaldehyde matrix based green composites," *Iran. Polym. J.* 17(11), 861-873.

Söğütlü, C. (2017). "Determination of the effect of surface roughness on the bonding strength of wooden materials," *BioResources* 12(1), 1417-1429.

Stark, N. M., Cai, Z., and Carll, C. (2010). "Wood-based composite materials – Panel products, glued-laminated timber, structural composite lumber, and wood-nonwood composite materials: Chapter 11," in: *Wood Handbook: Wood as an Engineering Material*, Centennial Ed., General technical report FPL (GTR-190), Madison, WI, U.S. Dept. of Agriculture, Forest Service, Forest Products Laboratory, pp. 11.1-11.28. DOI: 10.2737/FPL-GTR-190

Veigel, S., Müller, U., Keckes, J., Obersriebnig, M., and Gindl-Altmutter, W. (2011). "Cellulose nanofibrils as filler for adhesives: Effect on specific fracture energy of solid wood-adhesive bonds," *Cellulose* 18, 1227-1237. DOI: 10.1007/s10570-011-9576-1

Veigel, S., Rathke, J., Weigl, M., and Gindl-Altmutter, W. (2012). "Particle board and oriented strand board prepared with nanocellulose-reinforced adhesive," *J. Nanomater.* 2012 (15), 1-8. DOI: 10.1155/2012/158503

Wang, Z., Gu, Z., Hong, Y., Cheng, L., and Li, Z. (2011). "Bonding strength and water resistance of starch-based wood adhesive improved by silica nanoparticles," *Carbohyd. Polym.* 86(1), 72-76. DOI: 10.1016/j.carbpol.2011.04.003

Yuan, Y., and Randall L. T. (2013). "Contact angle and wetting properties: Chapter 1.," in: *Surface Science Techniques*, G. Bracco and B. Holst (eds.), Springer Verlag, Berlin, Heidelberg, pp. 3-34.

Article submitted: October 26, 2017; Peer review completed: December 4, 2017; Revised version received and accepted: December 16, 2017; Published: February 5, 2018.
DOI: 10.15376/biores.13.2.2283-2292