

# Possibility of Use of NCC-Reinforced Melamine-Urea-Formaldehyde Adhesive in Plywood Manufacturing

## Mogućnost uporabe melamin-urea-formaldehidnog ljepila ojačanog nanocelulozom u proizvodnji furnirskih ploča

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**ABSTRACT** • The possibility of using nanocellulose (NCC) as a filling substance for melamine-urea-formaldehyde (MUF) adhesive was investigated for the process of manufacturing plywood. The adhesive mixtures were prepared with various nanocellulose concentrations. The amount of introduced filler had a significant effect on both resin and plywood characteristics. Fourier transform infrared spectroscopy (FTIR) did not show any major changes between experimental and reference variants. The viscosity of resin increased after the introduction of nanocellulose. The addition of NCC in the amount of 5 g and 10 g per 100 g of solid resin led to an improvement in bonding quality, modulus of elasticity and bending strength. Further increase of NCC concentration caused a deterioration of manufactured plywood properties. In summary, the addition of proper amount of nanocellulose resulted in manufacturing plywood with improved properties.

**Keywords:** plywood; melamine-urea-formaldehyde adhesive; nanocellulose; filler

**SAŽETAK** • U radu je prikazano istraživanje mogućnosti uporabe nanoceluloze (NCC) kao punila za melamin-urea-formaldehidno ljepilo (MUF) koje se upotrebljava u proizvodnji furnirskih ploča. Smjese ljepila pripremljene su dodavanjem različitih koncentracija nanoceluloze. Količina dodanog punila znatno je utjecala na svojstva smole i furnirske ploče. Furierovom infracrvenom spektroskopijom (FTIR) nisu utvrđene veće promjene između eksperimentalnih i referentnih varijanti. Viskoznost smole povećala se nakon dodatka nanoceluloze. Dodatak 5 i 10 g nanoceluloze na 100 g otvrdnute smole rezultirao je poboljšanjem kvalitete vezanja, modula elastičnosti i čvrstoće na savijanje. Daljnje povećanje koncentracije nanoceluloze uzrokovalo je pogoršanje svojstava proizvedenih furnirskih ploča. Ukratko, dodatak odgovarajuće količine nanoceluloze rezultirao je furnirskom pločom poboljšanih svojstava.

**Ključne riječi:** furnirska ploča; melamin-urea-formaldehidno ljepilo; nanoceluloza; punilo

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## 1 INTRODUCTION

### 1. UVOD

Nanotechnology is a relatively new science that has already become the frontier of the 21<sup>st</sup> century. It is defined as the field focused on the control and understanding of the matter at a dimensional range between 1 and 100 nm (Hulla *et al.*, 2015). From the beginning, nanotechnology has been playing the role of a scientific platform joining and linking together various disciplines (Szczesna-Antczak *et al.*, 2012). The objects with the dimensions between 1 and 100 nm have unique mechanical, optical, magnetic and electrical properties (Wegner and Jones, 2005). Thus, the nanoscience also provides many opportunities for wood-based materials manufacturing (Candan and Akbulut, 2013). It seeks to develop advanced materials having significantly improved chemical, physical properties and functions (Wegner and Jones, 2006).

Wood-based materials such as plywood, particleboards, medium- and high density fiberboards are becoming more popular in recent years. The production of plywood, which is a multilayer composite made of glued wood veneers, reached about 157 million m<sup>3</sup> in 2017 and it is still growing (Bekhta *et al.*, 2020; Sydor *et al.*, 2020). Due to its favorable mechanical properties and dimensional stability, it finds an application in many industry branches (Kawalerczyk *et al.*, 2019a). In structural applications, plywood is used in walls, ceilings and roof constructions; in furniture as a basic material for both upholstered and case furniture (Bekhta *et al.*, 2009b; Majewski, 2019). Moreover, it is also appreciated as a packaging material due to its ease of processing, lightness and durability, and in transport as a flooring material or in trailers construction. The properties of plywood and consequently its application depend on the quality of veneers and the type of used adhesive (Kawalerczyk *et al.*, 2020c; Mirski *et al.*, 2011).

Amino resins are synthetic adhesives widely used in wood-based material industry. Their participation in the general range of wood adhesives market is estimated at 85 % (Jóźwiak, 2018; Kamoun *et al.*, 2003). The three main types: UF (urea-formaldehyde), MF (melamine-formaldehyde) and MUF (melamine-urea-formaldehyde) may be distinguished as the most commercially available (Gonçalves *et al.*, 2019; Mirski *et al.*, 2020). The widespread use of MUF adhesives in recent years results from the higher bond quality, water resistance and lower formaldehyde emission in comparison with UF resin (Lei and Frazier, 2015; Tohmura *et al.*, 2001). The adhesives mixtures applied in plywood manufacturing have to contain various kinds of fillers. They are insoluble, non-volatile substances added in order to adjust the viscosity, reduce raw material costs and increase bonding between the wood components (Kawalerczyk *et al.*, 2019b; Ong *et al.*, 2018).

In recent years an interesting concept of using cellulose as a modifier for polymers has been increasingly investigated. It is the most important constituent of the plants cell walls and, moreover, it is also synthe-

sized in tunicate sea animals and some bacteria (Henriksson and Berglund, 2007; Janardhnan and Sain, 2011). The attention received by cellulosic particles results from their high surface area, high stiffness and strength. Many studies on using nanocellulose (NCC) as an environmentally friendly modifier of wood adhesives have already been carried out (Ayrilmis *et al.*, 2016a; Gardner *et al.*, 2008; Vineeth *et al.*, 2019).

The effect of nanocellulose addition to amino resins was investigated by Veigel *et al.* (2012). They found that the modification increased the viscosity of liquid adhesive and caused a significant improvement in both particleboard and oriented strand board (OSB) strength properties. Studies conducted by Mahrtdt *et al.* (2016) confirmed that the introduction of cellulosic particles led to the increase in particleboard strength values. Furthermore, the influence of nanocellulose on multilayered wood-based material has also been investigated. Zhang *et al.* (2011) noted a major enhancement in plywood shear strength and the reduction in formaldehyde emission because of the silanized nanocellulose added to UF adhesive. However, studies conducted by Kawalerczyk *et al.* (2020b) did not confirm the effect on the reduction of the harmful formaldehyde emission with the use of non-modified nanocellulose. Ayrilmis *et al.* (2016b) concluded that cellulosic particles added to UF adhesive can reduce the VOCs (volatile organic compounds) emitted from LVL (laminated veneer lumber). The price of nanocellulose may be a limiting factor for some applications but there are many studies on finding the way to obtain it with more cost-effective methods (Babicka *et al.*, 2020; Kawalerczyk *et al.*, 2021).

Despite the fact that MUF adhesives are becoming more popular, especially for applications in kitchens, floors and some structural materials, and although they are increasingly replacing pure UF adhesives, no studies have examined the effect of their nanocellulose-reinforcement on the properties of manufactured plywood panels (Lei and Frazier, 2015). Thus, the aim of the present study was to investigate the effect of adding NCC to MUF adhesive on the plywood performance.

## 2 MATERIALS AND METHODS

### 2. MATERIJALI I METODE

#### 2.1 Materials

##### 2.1. Materijali

Rotary cut birch (*Betula L.*) veneer sheets were purchased from the market with the dimensions of 320 mm × 320 mm × 1.3 mm, moisture content of (5 ± 1) %, without any defects, and they were used for the research purpose. The commercial MUF adhesive with the following characteristics: solid content of 64 - 69 %, gel time at 100 °C of 63 s, viscosity between 1000 and 2500 mPa·s, density of 1.27 g/m<sup>3</sup> and pH of 9.5 - 10.7 was purchased for the experiment. The 20 % aqueous solution of ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) was added as a hardener taking into account the environmental aspects (Aras *et al.*, 2015). In order to adjust

and regulate the viscosity of adhesive mixture, the rye flour was introduced in accordance with the industrial formulations. Nanocellulose, added as a modifier, was purchased from Nanografi Nanotechnology Co. Ltd. (Ankara, Turkey). As declared by the producer, the dimensions of the particles were 300 - 900 nm in length and 10 - 20 nm in width.

## 2.2 Methods

### 2.2. Metode

A 10 % aqueous suspension of nanocellulose was prepared with the use of magnetic stirrer (600 rpm, 10 min), because cellulosic nanoparticles had to be processed in wet state. The experimental adhesive mixtures with the addition of NCC suspension, flour and hardener were homogenized using CAT-500 homogenizer at 1000 rpm for 2 minutes. The control adhesive was prepared in accordance with an industrial formulation. The compositions of both experimental and reference mixtures are summarized in Table 1.

No additional water was added in experimental variants because the nanocellulose was introduced in the state of water suspension. The viscosities of adhesives mixtures and their changes in 6 h were investigated using Brookfield DV-II + Pro viscometer. In order to assess the chemical bonding between the resin and nanocellulose, Fourier transform infrared spectroscopy was carried out. Reference and experimental adhesive mixtures were cured at 140°C and grinded. The obtained powders with a fraction of 0.125 mm were mixed with KBr at 1/200 mg ratio. Spectra were registered using a Nicolet iS5 spectrophotometer (Thermo Fisher Scientific) with Fourier transform at the range of 500 - 4000 cm<sup>-1</sup> at the resolution of 4 cm<sup>-1</sup>, registering 16 scans.

The adhesive mixtures were spread on the surface of external veneers in the amount of 170 g/m<sup>2</sup>. The veneer sets were assembled perpendicularly to each other. Three-layer plywood panels with the dimensions of 320 mm × 320 mm were produced in a hydraulic laboratory press with the following pressing parameters: temperature of 140 °C, unit pressure of 1.3 MPa and pressing time of 4 min. Three replicate panels were produced for all the test groups. Manufactured plywood was tested in order to investigate the following properties:

- formaldehyde (HCHO) emission with the flask method according to EN 717-3 (1996) initially and after 30 days of panels conditioning while left open at (20 ± 3) °C and (65 ± 2) % relative humidity,

- shear strength after 24 h of soaking in water at (20 ± 3) °C and after preparation including boiling in water for 6 h and cooling in water at (20 ± 3) °C for 1 h according to EN 314-1 (2004),
- modulus of elasticity and bending strength parallel and perpendicular to the grains of face layer according to EN 310 (1993).

On the basis of the results obtained in above tests, the proper composition of adhesive mixture was selected. The experimental adhesive labeled “N5” showing the best reinforcing effect was applied in the amount of 170, 160, 150, 140 and 130 (g/m<sup>2</sup>) in order to investigate the possibility of reducing the adhesive consumption in plywood production. The pressing process was conducted at 140 °C with the unit pressure of 1.3 MPa for 4 min. Manufactured plywood panels were tested in terms of shear strength both after 24 h of soaking in water at (20 ± 3) °C and after pretreatment consisting of boiling in water for 6 h and cooling in water at (20 ± 3) °C for 1 h according to EN 314-1 (2004). The obtained results were compared with a reference variant, which was labeled “170 REF” in further part of the paper.

The evaluation of mechanical properties and bonding quality involved 12 samples of each variant. The results were subjected to the multivariate statistical analysis ANOVA. The Tukey test with a significance level of  $\alpha = 0.05$  was applied to distinguish the homogeneous groups with the use of Statistica 13.0 software.

## 3 RESULTS

### 3. REZULTATI

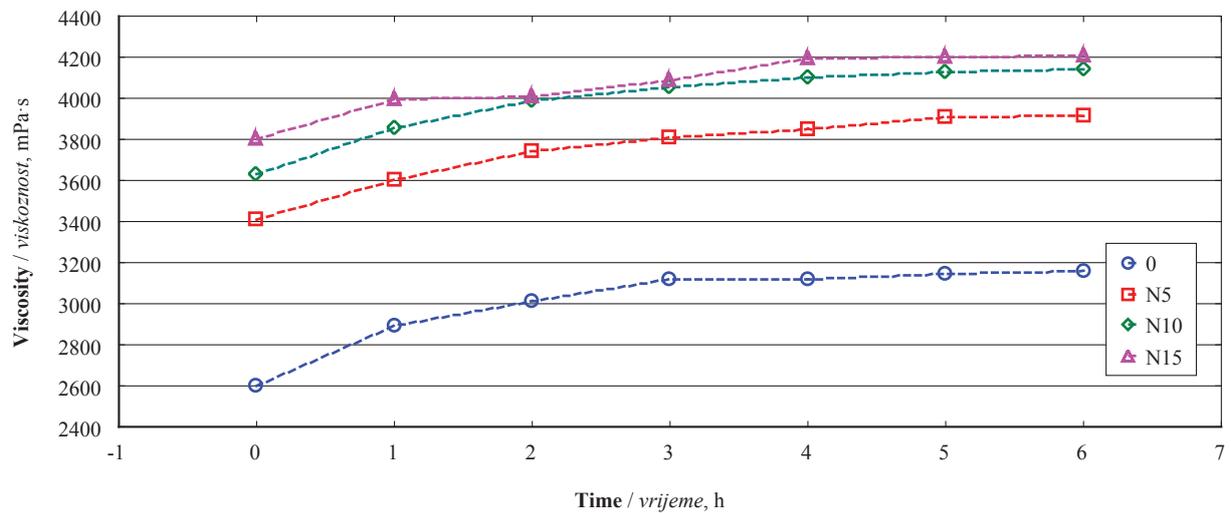
Figure 1 illustrates the time-viscosity dependence. The results indicate that nanocellulose-reinforced MUF resins were characterized by significantly increased viscosity in comparison with reference mixture.

The viscosity of adhesive mixture containing the maximal amount of cellulosic nanoparticles was 46 % and 33 % higher when compared to resin only filled with rye flour initially and after 6 hours of measurements, respectively. The increasing values of all resins viscosity during the test time resulted from the progressive polycondensation reactions and the constant water absorption by hydrophilic fillers. The viscosity of the glue mixture increased with the increasing percentage of nanocellulose addition similarly to investigations reported by Damásio *et al.* (2017). One factor that may

**Table 1** Compositions of adhesive mixtures

**Tablica 1.** Sastav smjesa ljepila

Variant label <i>Oznaka varijante</i>	Quantity (g/100 g of solid MUF resin) <i>Količina (g/100 g otvrdnute MUF smole)</i>			
	NCC suspension <i>NCC suspenzija</i>	Rye flour <i>Raženo brašno</i>	H <sub>2</sub> O	Hardener <i>Otvrdnjivač</i>
0	0	20	10	2.5
N5	5	10	0	2.5
N10	10	10	0	2.5
N15	15	10	0	2.5



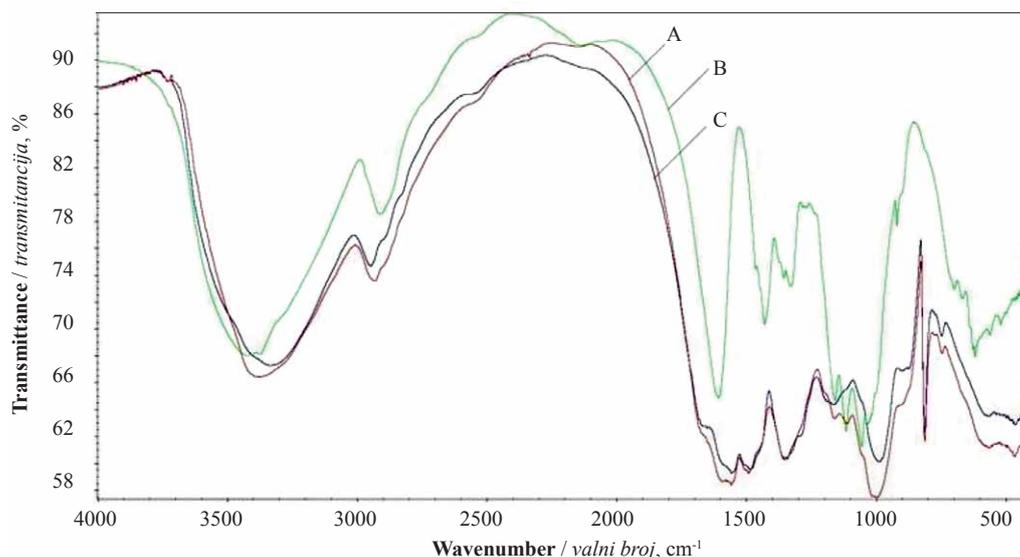
**Figure 1** Viscosity of adhesives mixtures  
**Slika 1.** Viskoznost smjesa ljepila

have contributed to this major increase in viscosity is the highly hydrophilic nature of cellulose. It tends to absorb water due to its chemical structure – the surfaces of cellulose are covered in hydroxyl groups on the equatorial positions of glucopyranose ring (Yamane *et al.*, 2006). Another factor is the increased chemical reactivity of nanoparticles in general (Shahbazi *et al.*, 2013). The introduction of nanocellulose probably caused a substantial interfibrillar interaction. The number hydroxyl groups located on the surfaces of individual fibrils can cause a considerable interaction between them, which can lead to the formation of temporary bonds (Iotti *et al.*, 2011).

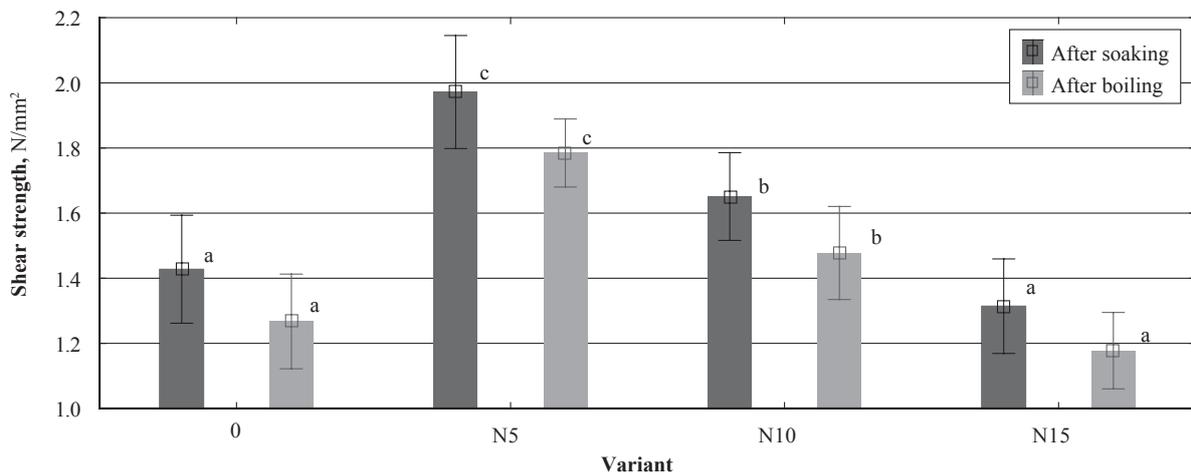
A similar effect was also observed in case of the NCC-reinforcement of UF and PF adhesives (Kawalerczyk *et al.*, 2020b, a; Mahrtdt *et al.*, 2016). Hong and Park (2017) distinguished the viscosity of adhesive as one of the most important factors affecting the strength properties of cured bonds. Studies conducted by Derkyi *et al.* (2008) also confirmed that the rheological properties of UF resin had a significant effect on plywood

bonding quality. The adhesive characterized by too low viscosity penetrates extensively into porous surface of the veneer during application and pressing. Consequently, the layer remaining on the veneer surface is no longer sufficient to ensure good quality of the bond (Sellers, 1985). The viscosity may also be a factor limiting the amount of nanocellulose added to the adhesive. The lack of water in the mixture can affect the crosslinking of adhesive and moreover, the addition of highly hydrophilic filler can prevent water from evaporating during the pressing process (Mahrtdt *et al.*, 2016; Réh *et al.*, 2019). Furthermore, it is also hard to evenly spread the adhesive characterized by too high viscosity and adjust it to the applying equipment. In case of this research, both reference and experimental mixtures obtained values that allowed the application without any difficulties.

Since the reinforcing effect of nanocellulose is associated with chemical bonding, the Fourier transform infrared spectroscopy (FTIR) was carried out in order to investigate the chemical interactions. The



**Figure 2** FTIR spectra of: A – modified resin labeled N5; B – nanocellulose; C – unmodified MUF resin  
**Slika 2.** FTIR spektri: A – modificirana smola označena kao N5; B – nanoceluloza; C – nedomificirana MUF smola



**Figure 3** Shear strength of plywood (a, b, c letters indicate homogeneous groups)

**Slika 3.** Čvrstoća na smik furnirske ploče (a, b i c slova označuju homogene grupe)

transmittance spectra of cured modified and non-modified resins are presented in Figure 2. Spectra of all modified resins regardless of the variant had the same course.

The broad band at  $3400\text{ cm}^{-1}$  was assigned to the O-H stretching of hydroxyl groups in each sample. The C-H stretching of methylene groups was recorded in the range from  $2920\text{ cm}^{-1}$  to  $2890\text{ cm}^{-1}$  (Luo *et al.*, 2015). The slight difference in intensity of C-H band was probably induced by the hydrogen bonding in MUF and NCC sample. For MUF and MUF+NCC samples, the peak at  $1690\text{ cm}^{-1}$  corresponded to C=O groups in amide (Müller *et al.*, 2009; Pandey and Pitman, 2003). In the case of NCC sample, the peak at  $1620\text{ cm}^{-1}$  was assigned to O-H vibration of absorbed water (Wulandari *et al.*, 2016). The peak at  $1320\text{ cm}^{-1}$  corresponded to C-H and C-O vibrations contained in the polysaccharide rings and it was observed at spectra of NCC and MUF+NCC. Spectra of MUF and MUF+NCC were characterized by a peak around  $1560\text{ cm}^{-1}$ , which was assigned to the C-N stretching of secondary amines. Moreover, the peak at  $1360\text{ cm}^{-1}$  was assigned to the C-N stretching of  $\text{CH}_2\text{-N}$  with confirmed presence of amine groups (Luo *et al.*, 2015). The peak at  $1060\text{ cm}^{-1}$  corresponded to vibration from the pyranose ring (spectra of NCC) (Wulandari *et al.*, 2016). In the case of triazine rings, the peak at  $1190\text{ cm}^{-1}$  was observed. It was assigned to C-N stretching vibrations (Yuan *et al.*, 2016). Spectra of MUF + NCC was characterized by occurrence of a peaks at  $1060\text{ cm}^{-1}$  and  $1120\text{ cm}^{-1}$ , which corresponded to pyranose rings and aliphatic ring, respectively (Luo *et al.*, 2015). These peaks were not observed at MUF spectra. Presumably, it resulted from the fact that interactions occurred between nanocellulose and MUF resin chemical. The peak at  $810\text{ cm}^{-1}$  was assigned to the typical stretching of the triazine ring of melamine and it was observed at spectra of MUF resin and MUF+NCC (Gao *et al.*, 2012; Kandelbauer *et al.*, 2007; Reimschuessel and McDevitt, 1960; Sun *et al.*, 2011).

The transmittance spectra of reference and reinforced adhesive revealed mostly the characteristic

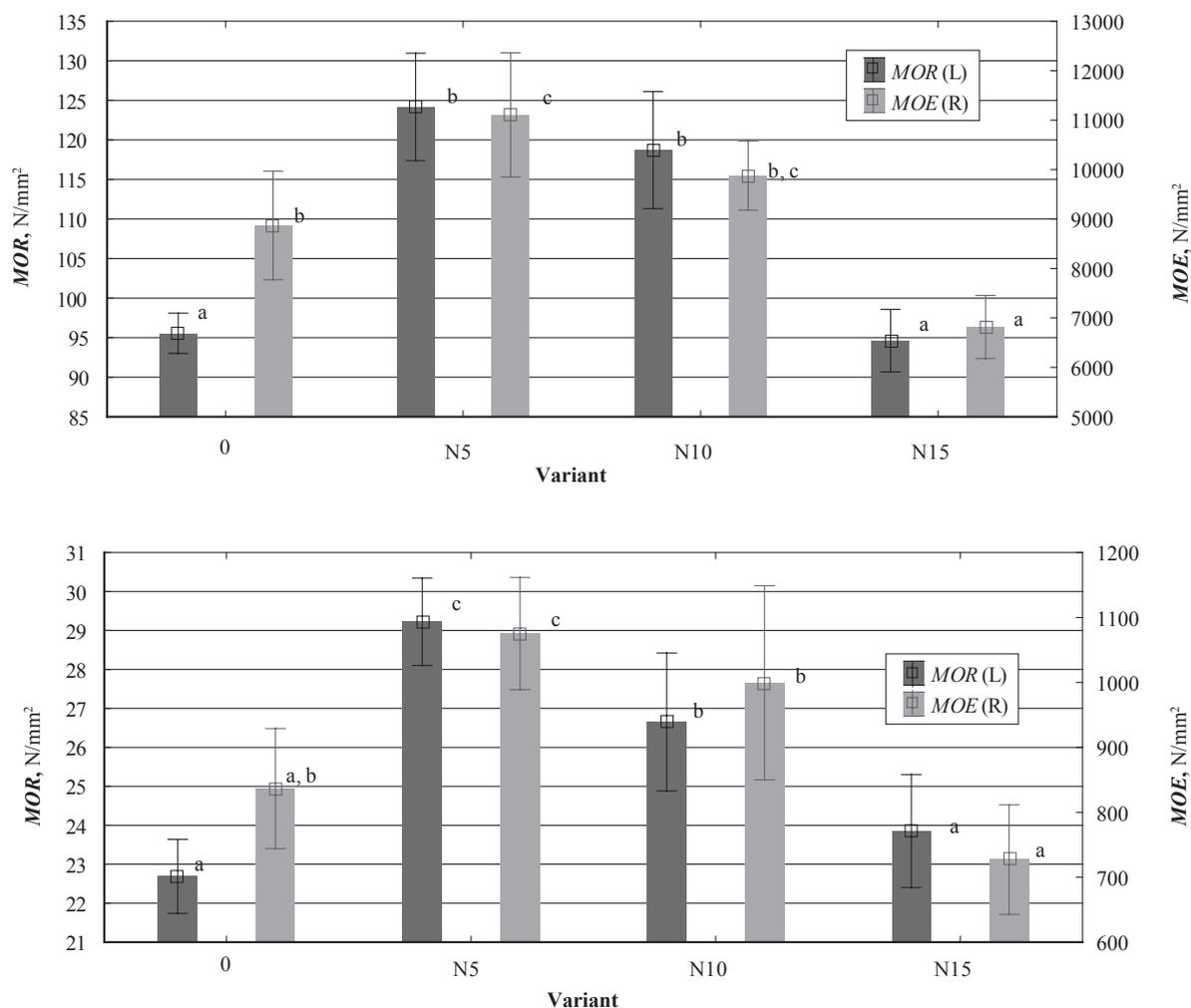
functional groups of MUF resins. According to literature, the interfacial bonding of nanocellulose with resins can be attributed to the reaction between the methylol groups of resin and hydroxyl groups of cellulose (Singha and Thakur, 2008).

Glue line quality is one of the most important properties of plywood affecting both its physical and mechanical characteristics (Bekhta *et al.*, 2009a). Thus, the shear strength test was carried out since it is the fundamental indicator of the adhesive performance in plywood panels (Bekhta *et al.*, 2016; Rohumaa *et al.*, 2013). The results of bonding quality are summarized in Figure 3.

In order to fully evaluate how changes in adhesive formulation influenced the mechanical properties of plywood, such as bending strength (*MOR*) and modulus of elasticity (*MOE*), investigations were made both parallel and perpendicular to the grains of the face layer. The results are presented in Figure 4.

On the basis of the research conducted, it was confirmed that the key to obtain optimum reinforcement effect was the amount of added nanoparticles (Ferreira 2017). The major improvements in shear strength values were obtained in variants where the percentages of added NCC were 5 and 10 %. The best results were observed in case of variant labeled N5 and the increase was 38 % and 41 % in comparison with reference panels after soaking and after boiling, respectively. The introduction of 10 g of cellulosic nanoparticles also led to the increase of bonding quality compared to the mixture filled only with flour. As expected based on the previous research, as the amount of nanocellulose increased, the shear strength value constantly decreased (Kawalerczyk *et al.*, 2020b, a; Veigel *et al.*, 2012). Bonding quality of panels labeled N15 was decreased by about 8 % both after soaking and boiling in water. However, all plywood samples met the requirements of EN 314-2 (1993) and their shear strength exceeded  $1\text{ N/mm}^2$ .

Mechanical properties, such as modulus of elasticity and bending strength (also called modulus of rupture) of the NCC-modified plywood show significantly



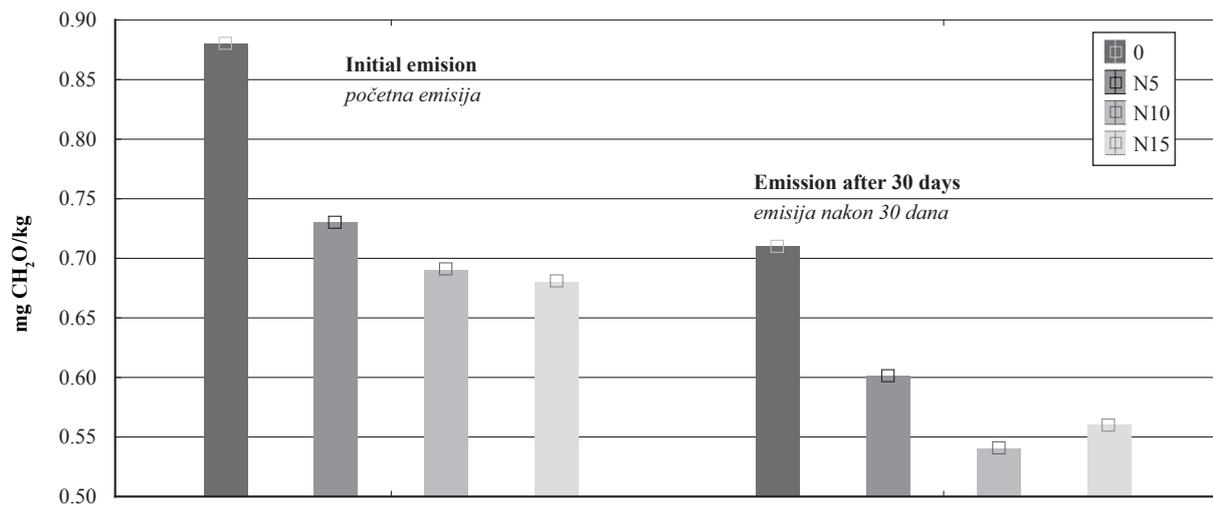
**Figure 4** Bending strength (*MOR*) and modulus of elasticity (*MOE*) of plywood tested: (a) parallel and (b) perpendicular to grains of face layer (a,b,c letters indicate homogeneous groups)

**Slika 4.** Čvrstoća na savijanje (*MOR*) i modul elastičnosti (*MOE*) ispitivane furnirske ploče: (a) u smjeru žice površinskog sloja i (b) okomito na smjer žice površinskog sloja (a, b i c slova označuju homogene grupe)

higher average values than reference plywood. The most satisfactory results were obtained in case of plywood modified with the smallest addition of nanocellulose. Modulus of elasticity and bending strength increased by approx. 27 % in comparison with control samples. Further addition of modifier at the level of 10 g also caused an improvement in plywood performance but, as expected from shear strength values, the maximum concentration of nanocellulose caused a notable decrease of panel strength properties.

The addition of nanocellulose to MUF resin shows similar results to the effect on UF resin modification (Kawalerczyk *et al.*, 2020b). Zhang *et al.* (2011) introduced silane-modified NCC in plywood manufacturing and observed that bonding quality increased by about 24 %. Furthermore, investigations of Damásio *et al.* (2017) showed that the shear strength of a glue line increased by 56 % after the addition of 8 % CNF (cellulose nanofibrils). Ayırlımış *et al.* (2016a) stated that the enhancement in UF-glue joints strength resulted from their increased ductility. Veigel *et al.* (2012) also confirmed the reinforcing nature of nanocellulose. Properties of manufactured particleboards and OSB (oriented strand boards) were improved when the CNF

was added to UF resin. The reason for that major reinforcing effect is, as reported by Vineeth *et al.* (2019), the improvement in fracture energy and fracture toughness. Veigel *et al.* (2011) showed that the addition of nanocellulose in the amount of 2 % resulted in the increase of the toughening effect up to 45 %. The enhancement can be also attributed to the chemical bonding between the methylol groups of resin and the free hydroxyl groups contained in cellulosic chain (Fornué *et al.*, 2011). Moreover, Hu *et al.*, (2014) confirmed that the presence of cellulose nanocrystals increase the wood-adhesive bonding and interactions. Another aspect leading to the reinforcing effect of the introduction of nanocellulose is the changes in resin morphology. Major fragility of amino resins results from their tendency to develop microcracks (Thomas *et al.*, 2019). They deteriorate the mechanical performance of manufactured materials, and limiting their occurrence has a significant effect on the strength properties of glue lines (Kawalerczyk *et al.*, 2019b). Kawalerczyk *et al.* (2020a) investigated the effect of nanocellulose addition on phenol-formaldehyde resin morphology. Studies have shown that the modification enhanced the structure of cured resin and made it significantly less



**Figure 5** Formaldehyde emission from plywood  
**Slika 5.** Emisija formaldehida iz furnirske ploče

porous, more solid and compact. The deterioration observed in bonding quality, MOR and MOE resulted from too high concentration of nanoparticles, which consequently led to the formation of agglomerates. It has particularly negative effect since the nanofillers act like carriers of stress along the glue line and the occurrence of agglomerates cause the accumulation of load at certain points of the bond (Singha and Thakur, 2008).

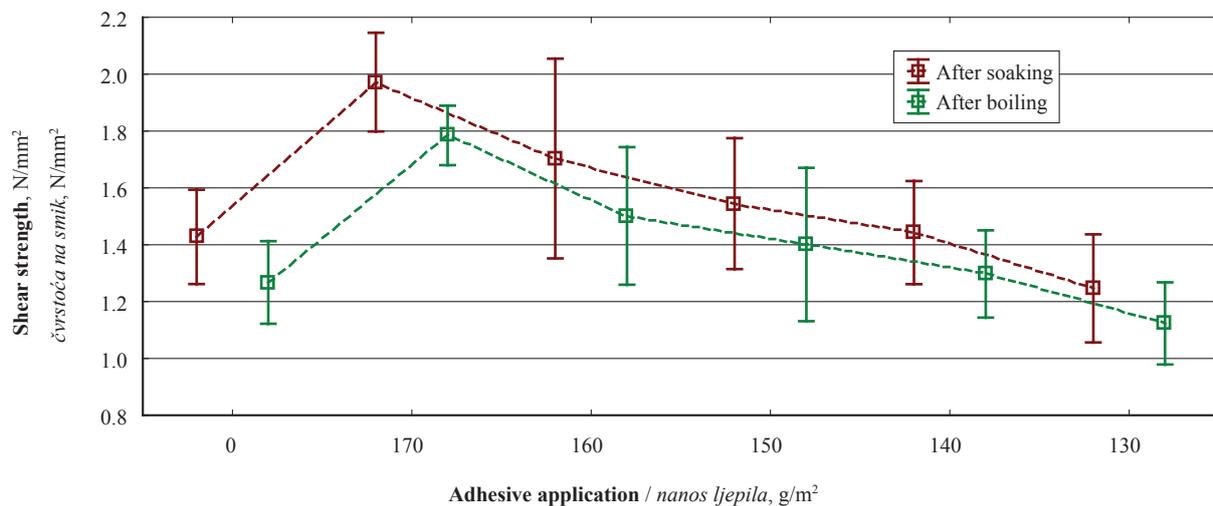
A major disadvantage of amino resins is a harmful formaldehyde emission (Dziurka and Mirski, 2014). According to literature, there are three main sources of emitting formaldehyde from adhesives: hydrolytic degradation of cured resin, residual formaldehyde contained in the resin and condensation reaction between hydroxymethyl groups and other aromatic carbon or two hydroxymethyl groups (Tohmura *et al.*, 2001). Since building materials including wood-based materials are considered to be one of the most common sources of formaldehyde emission, it is important to reduce it. The results of HCHO emissions are presented in Figure 5.

Plywood panels manufactured with the use of NCC-modified resin were characterized by significantly lower formaldehyde emissions. As the concentration of cellulosic nanoparticles increased, the amount of emitted HCHO was substantially reduced. Thus, the best results were observed in case of variant N15, where the decrease was 23 %. Studies conducted by Zhang *et al.* (2011) have shown very similar effects. Authors stated that the addition of modified NCC led to reduced HCHO emission due to physical adsorption and chemisorption. Moreover, Li *et al.* (2015) hypothesized that the decreased formaldehyde emissions can be attributed to increased viscosity of an adhesive. According to the author's theory, the glue mixture characterized by low viscosity penetrates into the wood pores, which consequently increases the emission of plywood. It was also reported that the monomeric formaldehyde and polyoxymethylene glycols contained in the resin can easily interact with OH groups of nanoparticles (Candan and Akbulut, 2013; Dudkin *et al.*, 2006).

Furthermore, cellulose itself is able to irreversibly bound formaldehyde in small quantities (Bekhta *et al.*, 2019; Kamath *et al.*, 1985). Another reason for this reduction can be the unique characteristics of nanoparticles such as chemical activity, tremendous surface area or physical properties. Liu and Zhu (2014) explained that the decrease in HCHO emissions results from the ability of nanoparticles to absorb free formaldehyde from adhesives.

The resin costs are more than 60 % of the total wood-based materials manufacturing costs (Cao *et al.*, 2018). Both the modification of veneer surface and the modification of adhesives are investigated factors that can possibly lead to the reduction of the binding agent consumption. The possibility to reduce the amount of applied resin was determined on the basis of shear strength test and the results are presented in Figure 6.

As the amount of applied glue mixture decreased, the bonding quality also decreased. The shear strength values were reduced since the quantity of applied resin was insufficient to fully and evenly cover the veneer surface (Bekhta and Marutzky, 2007). Introducing nanocellulose in the amount of 5 g per 100 g of solid MUF resin had a positive effect on its properties, durability and morphology, and consequently it allowed to reduce adhesive spread rate by 30 %. The experimental plywood glued with reference resin mixture in the amount of 170 g/m<sup>2</sup> was characterized by equally good shear strength as panels manufactured with the use of NCC-reinforced adhesive in the amount of 140 g/m<sup>2</sup>. The differences between these two variants were not statistically significant and the p-value was 0.999020 and 0.999987 after soaking and after boiling, respectively. However, further decrease of adhesive spread rate led to a notable deterioration in bonding quality, but the results still met the requirements of EN 314-2 (1993). The modification of resin seems to be more effective than surface modification, e.g. veneer compression (Bekhta and Marutzky, 2007). Corresponding results were obtained by Dukarska and Czarniecki (2016) in studies concerning the nano-SiO<sub>2</sub> addition to MUPF (melamine-urea-phenol-formaldehyde) adhesive. The



**Figure 6** Reduction of adhesive application in manufactured plywood  
**Slika 6.** Smanjenje nanosa ljepila u proizvodnji furnirske ploče

introduction of fumed nanosilica allowed to decrease the resin spread rate in plywood production by 30 %. In future, it would be interesting to investigate how the reduction of MUF adhesive consumption influences the emission of free formaldehyde.

## 4 CONCLUSIONS

### 4. ZAKLJUČAK

The time-viscosity dependence showed a significant increase after the addition of nanocellulose. As the amount of added nanomodifier increased, the viscosity values also increased.

The Fourier transform infrared spectroscopy (FTIR) did not show any major changes between experimental and reference samples, thus it did not explain the chemical interaction between nanocellulose and melamine-urea-formaldehyde resin.

The addition of small amounts of nanocellulose (5 g and 10 g) led to the improvement in bonding quality and mechanical properties such as modulus of elasticity and bending strength. The best results were obtained in case where the concentration of NCC was up to 5 g per 100 g of solid resin.

The MUF adhesive modification with nanocellulose caused a decrease in the amount of emitting formaldehyde.

The addition of nanocellulose allowed the reduction in adhesive consumption by 30 %.

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