Applicational Properties of Reinforced Plywood with Nanomaterials and Kenaf Fiber

Pantea Omrani, Hamideh Abdolzadeh,* Foad Roshan, and Mahdi Ganjkhani

Kenaf fibers were added as a reinforcement between wood veneers of poplar (*Populus deltoides*) bonded with urea–formaldehyde (UF) resin to improve the applicational properties of standard three-layered plywood. Additionally, the influence of two different nanomaterials (nanocellulose and nanosilica)-modified UF resins on the performance of plywood was evaluated. Then, thickness swelling (TS), water absorption (WA), shear strength, and flexural properties were examined. Results indicated that reinforced composites with kenaf fibers improved the modulus of rupture (MOR) and modulus of elasticity (MOE) in both directions. In addition, physical properties, such as TS and WA after 24 h, improved in the reinforced plywood with kenaf and use of nanosilica (KNS) as a filler. The results of the mechanical properties were better than blanks. The treated adhesive, with nanocellulose and nanosilica revealed similar mechanical behaviors. The shear strength of plywood in KNC specimens showed the best result (increased 64.6% compared to blank) and MOR for both the parallel and perpendicular directions to the grain of the surface layers for KNS (105% and 158%, respectively), and MOE for KNS (92.9% and 152%, respectively) compared to the blank.

DOI: 10.15376/biores.18.4.7054-7065

Keywords: Plywood; Nanomaterials; Kenaf fiber reinforced composite; Nanosilica; Nanocellulose

*Contact information: Department of Wood Science & Technology, Shahid Rajaee Teacher Training University, Tehran, Iran; *Corresponding author: h_abdolzadeh@sru.ac.ir*

INTRODUCTION

Wood is a natural, polymeric, cellular fiber composite that is broadly accessible and has been used for all kinds of application purposes throughout the history of mankind (Shi and Walker 2006). Its continued use is because wood and wood-based composites have good properties, such as natural origin and beautiful appearance. Thus, it has been used for construction, furniture, decking, *etc.* Solid wood is used as a construction material. However, for this purpose, it has deficiencies, such as anisotropy, biodegradability, and dimensional limitations. Wood-based products, such as plywood, particleboard, oriented strand board, cross-laminated timber, and fiberboard have been developed to overcome these disadvantages. Among the most used wood-based composites are layered materials, such as plywood or laminated veneer lumber (LVL), which are layered composites with better strength properties than the raw material itself. Despite the many advantages of wood composite products, they also have disadvantages due to the use of adhesive in their construction. The use of synthetic adhesives for manufacturing wood composites, as well as using them *in situ*, brings biological disadvantages and toxicity. For this purpose, researchers have tried to produce wood-based composites with less resin consumption and provide better strength for the products. Construction of wood composites with high strength and durability is one of the aims of designers. Advancements in the production of fiber-reinforcing composites have led researchers to use fibers in the manufacturing of wood composites to produce high resistant and high strength products. In research from 2001, synthetic fibers were used continuously and discontinuously in the polymer matrix for increasing performance (Callister and Rethwisch 2001). If the aspect ratio of fibers (fiber length to fiber diameter, L/d) is higher, the flexibility and quality of fibers will increase (Dresher 1969). In addition to the use of synthetic fibers, researchers have also investigated the possibility of using some natural fibers in the construction of wood composites. Natural fibers have advantages and disadvantages compared to synthetic ones. Wood is a natural composite of cellular fibers and a polymer matrix of lignin (Mahút and Réh 2007). Materials reinforced with natural fibers are considered as a new group of materials and are known as natural fiber composites (NFCs). The advantages of NFCs are low weight and reduction of environmental issues (Baley *et al.* 2019). The NFCs are constructed by plant, animal, and mineral-based fibers (Müssig 2010). In the 1960s, synthetic fiber-based fabrics were used for the first time to increase the mechanical properties of composites (Laufenberg *et al.* 1984). The most recent research to reinforce composites with natural fibers has focused on the use of mineral-based basalt fibers (Kramár *et al.* 2019; Kramár and Král 2019; Tautenhain *et al*. 2019; Jorda *et al.* 2021; Lohmus *et al.* 2021). However, the use of synthetic fibers, such as glass, carbon, or basalt, creates problems during production and later in the stage of using cutting tools. Therefore, to overcome these disadvantages, the use of natural fibers, such as kenaf can be useful. In this regard, several studies have been conducted in the last decade (Zike and Kalnins 2011; Basterra *et al.* 2012; Bal 2014; Jorda *et al.* 2020; Valdes *et al.* 2020; Jorda *et al.* 2021; Ramesh *et al.* 2021).

Kenaf bast fiber bundles (KBFBs) are commonly used (Holbery and Houston 2006). Kenaf is the name given to a similar fiber obtained from the stems of plants belonging to the genus *Hibiscus*, family Malvaceae, especially the species *H. cannabinus* L. (Kundu 1956; Atkinson 1965). Its single, straight stem consists of an outer fibrous bark and an inner woody core that yield two distinct fibers, bast and core, respectively. Natural fibers, such as kenaf, have been used for improving the applicational properties of NFCs, and in various experimental research, these reinforced products were investigated (Laufenberg *et al.* 1984; Jorda *et al.* 2021). The research results showed that there was a significant difference in the mechanical properties of reinforced plywood according to the type of reinforcement. In addition, the type of resin used as a polymer matrix has also been effective in increasing mechanical resistance. It seems that the use of natural fibers should be given more attention in terms of environmental issues and due to the availability of resources (Jorda *et al.* 2021). Although the use of natural fibers is not new, the use of synthetic fibers in recent decades is also taken into consideration (Baley *et al.* 2021). Speranzini and Tralascia (2010) reinforced wood-based composites with synthetic reinforced polymers, such as glass and carbon (GFRP and CFRP), and some natural fibers. The results showed that the improvement of flexural strength in reinforced composites with natural fibers was less than synthetic fibers, but there were significant improvements compared to the blank. Moezzipour *et al.* (2017) investigated the effect of use of natural fiber reinforcement on the functional properties of wood-based products. They concluded that the use of natural fibers improves the mechanical properties. Valdes *et al.* (2020) reinforced cross-laminated timber (CLT) with natural fibers. They concluded that the load-carrying capacity and stiffness of reinforced products are improved. Jorda *et al.* (2020)

studied the effect of using flax fiber reinforced composites on plywood. Investigations showed that the stiffness and load-carrying capacity has been improved in these products.

Due to the negative environmental impacts of using adhesives such as urea formaldehyde in usual quantities, this research has been conducted to reduce the consumption of these resins as well as to ensure the strength properties of wood-based composites. The purpose of this research is to investigate the effect of using natural fibers to reinforce the adhesive line of plywood and compare their mechanical and physical resistance with conventional plywood. In addition, to determine the effect of reinforcements, the strength properties of the plywood made of nanomaterials (nanocellulose and nanosilica)-modified UF resins were compared to those made of kenaf fibers in the glue line and control samples.

EXPERIMENTAL

Materials

Poplar (*Populus deltoides*) veneers with a thickness of 2 mm and dimensions of 400 mm × 400 mm and an average moisture content of 8% were used. The veneers were conditioned under normal conditions (20 ± 2 °C, 65 ± 5% relative humidity, RH) for at least seven days.

String from mechanically retted KBFBs was obtained and used without any chemical treatments such as delignification or bleaching. Indeed, bundles with 40 cm length were cut from the strings and used as is. Kenaf fibers acted as fiber reinforcement. According to data provided by Schledjewski *et al.* (2006), characteristics of kenaf fibers are shown in Table 1. Strings of kenaf fibers were cut to the length of 40 cm, and after that the fibers were uniformly hand-laid up between wood layers. The amounts of fibers were considered at 160 g/m^2.

Table 1. Characteristics of Kenaf Fibers Used for Reinforcement*

Fibre Type	Fibre Type	Tensile Stiffness (GPa)	Ultimate Stress (MPa)
Kenaf (*Hibiscus cannabinus*)	Fibre bundles	24.6 ± 11.7	418.1 ± 195.3

*Note: Schledjewski *et al.* 2006

The nanosilica (nanoSiO$_2$) and nanocellulose particles were used to modify urea formaldehyde resin (UF). The specifications of nanosilica, silane coupling agent, and cellulose nanofiber (CNF) gel used here are shown in Tables 2 to 4.

Table 2. Nanosilica specifications

Average Particle Size (nm)	Balk Density (g/cm^3)	Density (g/cm^3)	Morphology	Color	Reflects UV Light (%)	Coverage Area (SSA) (m^2/g)
15 to 20	>0.1	2.4	Porous	White	85	~ 640

Table 3. Silane coupling agent specifications

Molar mass (g/mol)	Ignition Temperature (°C)	Density (g/cm^3)	Boiling point (°C)	Assay (%)
222.36	300	1.02	261 to 263	≥ 97

Table 4. Specifications of the cellulose nanofiber gel

Formula	Material State	Color	Production Method	Average nanofiber diameter (nm)	Purity (%)
$(C_5H_{10}O_5)n$	Gel (2.5%)	White	Mechanical synthesis	35	≥ 99

The nanosilica and cellulose nanofiber gel used for this study was prepared from nano Sadra Company, Iran. Silane coupling agent, γ amino propyl tri ethoxy silane ($H_2NC_3H_6Si\ (OC_2H_5)_3$) was used for treatment of nanosilica. It was purchased from Merck Company. The characteristics of UF resin are shown in Table 5. Ammonium chloride was applied as a hardener, and wheat flour was used as filler in blank. The mass composition of the adhesive was as follows 100:1:50, for resin, hardener, and filler, respectively.

Table 5. Specifications of UF resin

Solid Content (%)	Viscosity (cp)	Gel time (s)	pH	Density (g/cm^3)
55	320	70	7	1.3

Methods

UF resin modification with nanomaterials

To treat the nanosilica, silane coupling agent and the nanosilica were dispersed in 280 mL acetone (95 %) and then were mixed by means of magnetic stirrer for 8 h. The obtained transparent mixture was placed in an oven at the temperature of 60 °C. Finally, the rest of the sediment was ground and used in the composition of resin as filler. Due to the necessity of processing cellulosic particles in wet states, 10% aqueous suspension was mixed with a magnetic stirrer (700 rpm, 10 min). The adhesive filled with cellulosic suspension was mixed with CAT-500 homogenizer at 1000 rpm for 2 min to achieve the high level of particles dispersion.

Fabrication of Plywood

The manufactured plywood panels consisted of three veneers with fiber orientation 0-90-0 and two layers of kenaf fibers between them (Fig. 1). The configuration of the different elaborated composite panels is shown in Table 6. The pressing for manufacturing of plywood was carried out under pressure of 1.5 MPa at temperature of 120 °C for 7 min.

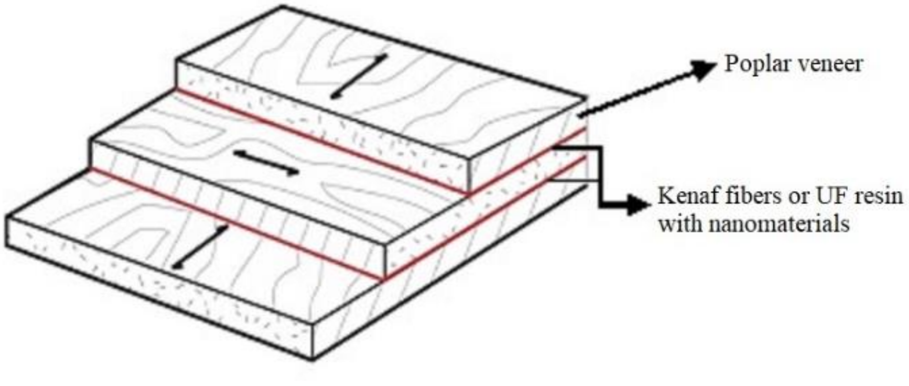

Fig. 1. Layers arrangement of the plywood panels (arrows show the grain direction)

Table 6. Composition of Panel Sets

Panel Set	Description	Construction [K: Kenaf, V: Veneer, (-): UF resin, NC: Nanocellulose and NS: Nanosilica]
Blank	Reference plywood (without any reinforcement)	V-V-V
NC	Plywood by reinforcing UF with nanocellulose	V NC V NC V
NS	Plywood by reinforcing UF with nanosilica	V NS V NS V
K	Plywood reinforced by kenaf fibers	V K V K V
KNC	Plywood reinforced by kenaf fibers and nanocellulose	V K NC V K NC V
KNS	Plywood reinforced by kenaf fibers and nanosilica	V K NS V K NS V

The static bending strength (MOR) and modulus of elasticity (MOE) were determined based on the EN 310 (2005) standard for both the parallel (∥) and perpendicular (⊥) directions to the grain of the surface layers. Bonding quality was determined through the tensile test perpendicular to the surface of the board, according to EN 314 (2005) standard (Fig. 2). All tests were conducted with a Zwick Roell universal testing machine.

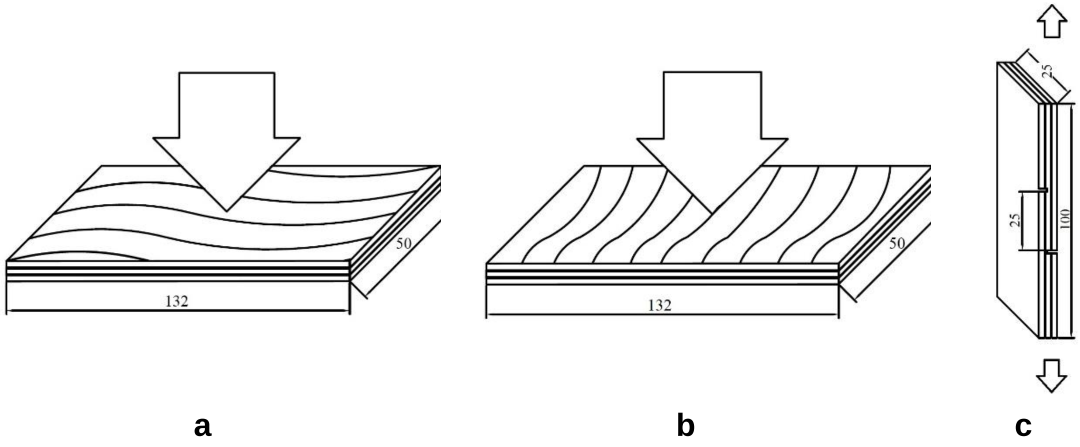

Fig. 2. Dimensions of mechanical test specimen (the static bending specimen (a: ∥ and b: ⊥) and shear test specimen (c))

The MOR and MOE were calculated following Eqs. 1 and 2, respectively,

$$MOR = \frac{3F_{\max} \times L}{2bh^2} \tag{1}$$

where $F_{\max}$ is the maximum force at the time of rupture (N), L is the span between supports (mm), b is the width of the samples (mm), and h is the height of the samples (mm).

$$MOE = \frac{\Delta F \times L^3}{\Delta a \times 4bh^3} \tag{2}$$

In Eq. 2, ΔF is the load increment, L is the span between supports (mm), Δa is the deflection (mm), b is the width of the samples (mm), and h is the height of the samples (mm).

Water absorption (WA) and thickness swelling (TS) after 24 h of soaking in water were measured according to EN 317 (1993). Samples were soaked in distilled water at room temperature for 2 h and 24 h. The results of the determination of the applicational properties of specimens were statistically analyzed by SPSS statistics software program (IBM Corp., Armonk, NY, USA). To determine significant differences between treatments, analysis of variance (ANOVA) at 95% confidence level was performed. A comparison of the means was performed by Duncan test, with 0.05 significance level.

RESULTS AND DISCUSSION

Mechanical Properties

Figures 3 and 4 show the effect of the type of nanomaterials and kenaf fiber on the MOR and MOE of specimens. It was found that variables significantly affected the MOR and MOE ($p < 0.05$) for both directions. The ANOVA analysis showed that use of kenaf had the greatest effect on the MOR and MOE of specimens, while the use of only nanomaterials affected the MOR and MOE to a lesser extent (Table 7).

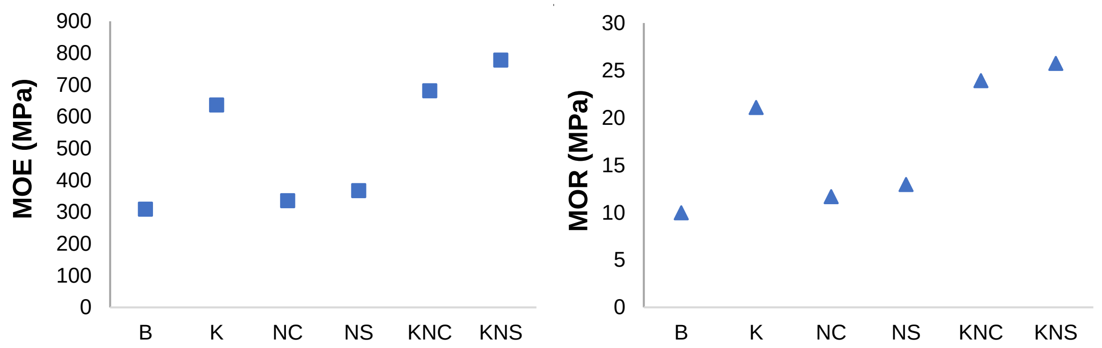

Fig. 3. MOE$_\perp$ and MOR$_\perp$ of boards bonded by reinforced adhesives

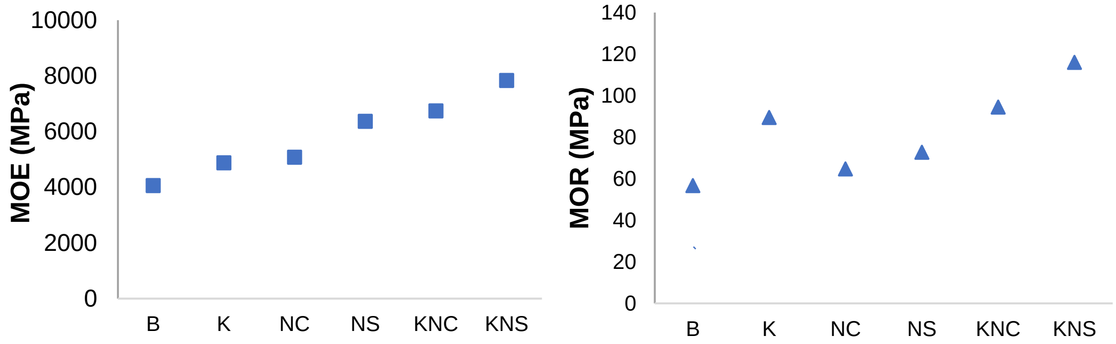

Fig. 4. MOE$_\parallel$ and MOR$_\parallel$ of boards bonded by reinforced adhesives

The highest MOR$_\parallel$ and MOE$_\parallel$ values 116 and 7840 MPa, respectively, corresponded to KNS. The lowest values of MOR$_\parallel$ and MOE$_\parallel$ were 56.7 and 4060 MPa, respectively, which belonged to the blank. It was reported that the use of reinforced adhesives reduces the use of adhesives and, as a result, formaldehyde emissions can be reduced (Chen *et al.* 2018).

Additionally, the bending properties of plywood were increased when using nanomaterials in combination with kenaf fibers. The kenaf cells act as reinforcements for hemicellulose and lignin matrices. Therefore, the cell wall is a composite structure of lignocellulosic material reinforced by helical microfibrillar bands of cellulose (Li *et al.* 2000). Lignin is of the main components of kenaf fibers with the greatest likelihood for self-adhesion (Stevens 2010). The total cellulose, hemicellulose, and lignin contents of kenaf fibres are about 36 to 72, 20 to 21, and 9 to 19%, respectively. Both lignin and hemicellulose soften under high moisture and temperature conditions. Hemicellulose more readily forms hydrogen bonds to bond the adjoining fibers, while lignin more readily forms chemical bonds (Frihart 2005). Lignin contribution in bonding has also been demonstrated by researchers on vibrational welding of wood joints (Omrani *et al.* 2012). Hence, it is expected that lignin and hemicelluloses of kenaf may have contributed to bonding the layers of plywood in this research too.

Studies on thermoplastic polymers have shown that the penetration of polymer into the wood's pores reinforced the adhesion. Further, studies have confirmed that the mechanism of bonding thermoplastic polymers in wood as a porous material is a mechanical locking type (Goto *et al.* 1982; Smith *et al.* 2002; Kajaks *et al.* 2012; Fang *et al.* 2014; Song *et al.* 2016; Chang *et al.* 2018).

The benefit of using smaller particles, such as nano-size, is reinforcing adhesion in composites. One of the theories involves reinforcement effects on the degree of crystallinity of matrix. Some documents have reported evidence for increased crystallinity in the presence of nano-size materials such as cellulose-based reinforcements (Hubbe and Grigsby 2020). The increasing surface area of reinforcements could affect crystallinity. Inducing additional crystallinity within the plastic phase generally provides a higher elastic modulus, leading to greater stiffness (Krishnaiah *et al.* 2017).

As expected, based on the hydrophilic properties of CNFs, viscosity may significantly increase with the addition of the nanomaterials. This may lead to restricting over-penetration of reinforced adhesive into the wood layer on which the glue line starving is avoided. The relatively high viscosity of CNF modified UF resin is caused by considerable interfibrillar interaction. Surfaces of cellulosic fibrils are covered with hydroxyl groups that can lead to the formation of temporary bonds between the adjacent fibrils (Iotti *et al.* 2011; Kawalerczyk *et al.* 2020).

Table 7. Average Values of Physical and Mechanical Properties of the Plywood

Applicational Properties	Blank	K	NC	NS	KNC	KNS
∗∗MOE ⊥ (MPa)	308.74	636.46	335.02	367.09	681.01	777.48
changes to the blank (%)		106.14	8.51	18.90	120.58	**151.82**
∗∗MOR ⊥ (MPa)	9.97	21.08	11.68	12.96	23.92	25.73
changes to the blank (%)		111.38	17.12	29.92	139.85	**158**
∗∗MOE ǁ (MPa)	4062.68	4878.85	5078.11	6370.94	6741.7	7837.41
changes to the blank (%)		20.09	24.99	56.82	65.94	**92.91**
∗∗MOR ǁ (MPa)	56.74	89.52	64.72	72.75	94.52	116.08
changes to the blank (%)		57.77	14.06	28.21	66.58	**104.58**
∗∗Shear Strength (MPa)	1.82	2.60	2.20	1.96	2.99	2.76
changes to the blank (%)		42.66	21.16	7.97	**64.65**	51.73
∗∗WA after 24h (%)	84.59	52.92	74.37	68.32	40.48	41.41
changes to the blank (%)		-37.44	-12.08	-19.23	**-52.13**	-51.04
∗∗TS after 24h (%)	9.68	6.21	7.63	9.45	6.07	3.85
changes to the blank (%)		-35.77	-21.11	-2.32	-37.28	**-60.24**

∗∗ Significant at 1% level

The mark (ǁ) for Parallel and (⊥) for perpendicular directions to the grain of the surface layers

Shear strength is one of the most effective features in plywood. Figure 5 examines the effect of the reinforced adhesive with kenaf fiber and nanomaterials on the shear strength of specimens. The results showed that they differed significantly based on Duncan's test ($p > 0.05$). The lowest and highest strength of the specimens were 1.81 and 2.76 MPa, respectively, for the blank and KNS samples. As mentioned earlier, the resistance of wood composites depends on the penetration of the adhesive into the wood's pores. The appropriate penetration of the adhesive inside the pores of the wood provides the possibility of making stronger bonding. The use of nanoparticles makes it possible for the adhesive to show better performance (Chang *et al.* 2017).

With the formation of new bonding between nanosilica and hydroxyamide of UF resin, the viscosity of reinforced resin is increased compared to pure UF, and a stronger adhesive line is provided due to less penetration of resin into pores and cracks (Caffery 1970; Yang *et al.* 2004; Moezzipour *et al.* 2013). In the UF-silica hybrid systems, one feasible mode of interaction is the formation of H-bonding between the carbonyl and NH groups of the carbonyl and NH groups of the UF domain with the siloxane (Moezzipour *et al.* 2013).

The better performance of boards reinforced with kenaf fibers for shear stresses is due to the chemical composition of kenaf. Kenaf fibers have higher amounts of hemicellulose in their structure, which causes self-bonding between these fibers and wood layers and increases the shear resistance of the reinforced boards (Moezzipour *et al.* 2017).

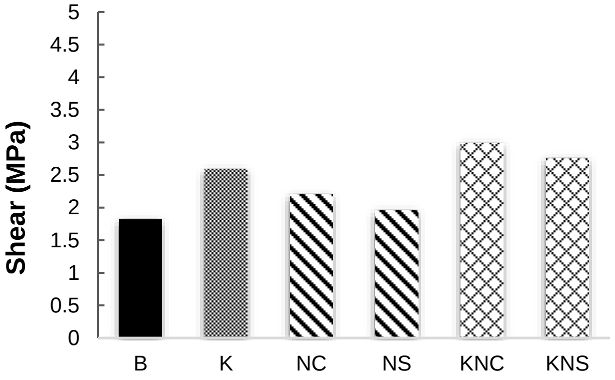

Fig. 5. Shear strength of board bonded by reinforced adhesives

Physical Properties

The influence of the nanomaterials and use of kenaf fiber as reinforced adhesive on TS and WA after soaking for 2 h and 24 h are presented in Fig. 6. As shown, thicknesses of specimens after 2 h soaking were B > NC > NS > K > KNC > KNS. The treatment's trend after 24 h was B > NC > NS > K > KNS > KNC. Statistical analysis showed that variables significantly ($p < 0.05$) affected WA and TS. The lowest value of WA (24 h) was recorded in KNC samples (40.49%), and the highest was recorded in blank samples made with UF resin (84.59%). The lowest TS (24 h) values corresponded to the KNS samples (3.85%), while the highest TS (24 h) corresponded to the blank samples made UF resin. It can be seen from Fig. 6 that the WA of plywood samples decreases with use of nanomaterials and kenaf fiber. As mentioned before, due to the creation of a network of new crosslinking by hydrogenation bonds between nanomaterials and UF and reinforced the glue line, the physical characteristics of the boards have been improved. As shown in Fig. 6, the use of kenaf fibers with higher amounts of hemicellulose also increases chemical bonds between fibers and wood layers. Therefore, the possibility of water entering the structure of reinforced boards is reduced.

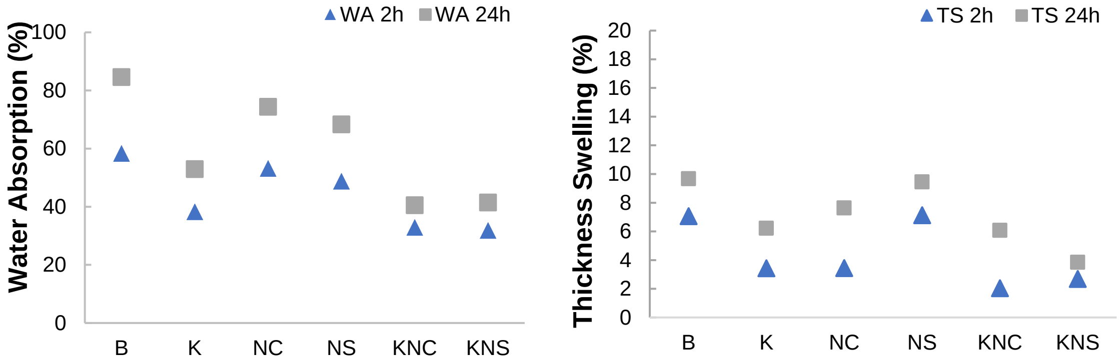

Fig. 6. WA and TS of specimens after 2 h and 24 h

CONCLUSIONS

1. Kenaf fiber with urea-formaldehyde (UF) resin and two different nanomaterials, *i.e.*, cellulose nanofibers (CNF) and nanosilica as fillers was used for preparing poplar plywood.
2. The usage of nanomaterials and kenaf fiber significantly affects the applicational properties of specimens.
3. Mechanical properties of plywood reinforced by kenaf fibers and nanofibers (KNC) and plywood reinforced by kenaf fibers and nanosilica (KNS) specimens increased compared to blank boards. The highest increase corresponded to KNS specimens at the perpendicular direction to the grain of the surface layer. It can be concluded that addition of kenaf fibers and use of nanomaterials increases the flexure strength of the planes. KNC plywood samples showed bonding strengths identical to samples. This is due to not only the effect of kenaf fiber, but also the use of nanomaterial and restricted penetration of resin into the wood layers.
4. Both nanosilica and CNFs are available nanomaterials which in combination with kenaf fibers exhibited considerable results in modifying UF resin and improved its performance in the plywood. The use of these materials in manufacturing wood-based composites reduces the consumption of UF petroleum-based resin. On the other hand, by using less resin and environment friendly materials, better mechanical strength and physical resistance can be obtained, and the release of formaldehyde can also be reduced.
5. In contrast, reinforced UF resin by nanomaterials and kenaf fibers caused a decrease in thickness swelling and water absorption after 2 and 24 h soaking in water.

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Article submitted: June 16, 2023; Peer review completed: July 8, 2023; Revised version received and accepted: August 15, 2023; Published: August 21, 2023.
DOI: 10.15376/biores.18.4.7054-7065