

## Performance characterization of plywood panels bonded with melamine-urea-formaldehyde resin and cellulose nanofibril/borax as an additive

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### Abstract:

In this study, different loading levels of cellulose nanofibril and borax were added as reinforcement in melamine-urea-formaldehyde adhesive to enhance the performance properties of plywood panels as engineered wood composites. Physical properties (density, thickness swelling, water absorption, and moisture content), mechanical properties (modulus of rupture, modulus of elasticity, and bonding strength), and formaldehyde content were tested using relevant standards. The results showed that cellulose nanofibril and borax had a synergistic effect, resulting in improved physico-mechanical properties. The best results were obtained by combining 3 % cellulose nanofibril and borax. It was determined that the combination of cellulose nanofibril and borax reinforcement resulted in a significant improvement of around 15 % in the thickness swelling, water absorption, and moisture content of plywood panels. The combination of cellulose nanofibril and borax reinforcing resulted in a significant increase of around 26 % in the modulus of rupture and modulus of elasticity of plywood panels, with a bonding strength of around 47 %. The reinforcement technique did result in a 34 % decrease in free formaldehyde content. As a consequence, cellulose nanofibril and borax can be used as effective additives in the production of plywood panels to enhance their performance properties.

**Keywords:** Borax, cellulose nanofibril, melamine-urea-formaldehyde resin, plywood panels.

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## Introduction

In 2020, the production of wood-based panels was 368 million m<sup>3</sup>. Particleboard, medium density fiberboard (MDF), and OSB (oriented strand board) total 250 million m<sup>3</sup>, whereas plywood totals 118 million m<sup>3</sup> (FAO 2020). The need for construction materials, as well as the global growth of residential and commercial structures, has resulted in an increase in demand for plywood (Yildirim *et al.* 2023a). Plywood panels are regarded as the most sophisticated of the many forms of wood-based composites due to their overall quality (Auriga *et al.* 2020). It is mostly used for roof sheathing, siding, floor underlayment, and structural diaphragms.

Engineered wood composites are manufactured with formaldehyde-based adhesives created from petroleum-derived components such as urea, phenol, and melamine. Urea-formaldehyde (UF), melamine-urea-formaldehyde (MUF), and phenol-formaldehyde (PF) resins are the most widely utilized amino plastic adhesives due to their low cost, solubility in water, rapid curing, high reactivity, and simplicity of processing (Ferdosian *et al.* 2017, Candan *et al.* 2022). MUF adhesives in different melamine ratios have high endurance to moisture and climate and are utilized in wet or outdoor applications (Mantanis *et al.* 2018). However, these adhesives have problems with the release of toxic volatile organic compounds (VOCs) and free formaldehyde, which are dangerous to humans and damaging to the environment (Candan 2012).

Environmental concerns and legislative regulations have driven both academia and industry to search for sustainable products. In recent years, there has been significant progress in the development of green, renewable, and sustainable materials (Yildirim *et al.* 2021a). Two ways of manufacturing products with improved characteristics contains the inclusion of fillers or additives as well as the alteration of resin chemistry (Yildirim *et al.* 2021b). The toxic formaldehyde release from wood-based composites could be minimized by introducing

formaldehyde scavengers to adhesives, using sustainable bio-based adhesive systems, or lignocellulosic bionanomaterials (Pizzi 2006, Hemmilä *et al.* 2017, Antov *et al.* 2020).

Recently, nanocellulose has been proven to be a sustainable biomaterial for the production of innovative adhesives and composites (Candan *et al.* 2022). Nanocellulose can increase adhesive mechanical properties and performance while reducing formaldehyde emissions (Candan and Akbulut 2013, Candan and Akbulut 2015, Vineeth *et al.* 2019). The three most common forms of nanocellulose are cellulose nanofibril (CNF), cellulose nanocrystal (CNC), and bacterial nanocellulose (BNC) (Candan *et al.* 2022). In particular, cellulose nanofibrils (CNF) have received a great deal of interest in recent years (Amini *et al.* 2019). CNF provides better mechanical features than CNC because of its longer fibrils, higher aspect ratio, and better interfacial adhesion (Poyraz *et al.* 2018, Tozluoglu *et al.* 2018, Yildirim *et al.* 2023b).

Since the latter years of the 20th century, the use of boron compounds has increased rapidly. Boric acid (BA) and borax (BX) are common boron preservatives used in the preservation of wood-based composites (Salman *et al.* 2014, Akgul and Camlibel 2021). Borax protects wood and building materials against fire, fungi, and insects (Kartal *et al.* 2019). Boron compounds are predicted to remain among the most promising wood protection compounds in the future due to their multiple advantages, which include low toxicity, low cost, transparency, non-volatility, and non-corrosiveness (Terzi *et al.* 2017).

Many studies have been done to investigate the effects of only nanocellulose and only boron compounds in the manufacture of wood composites. According to Colakoglu and Demirkir (2006), plywood panels modified with a borax-containing resin mixture emit less formaldehyde than unmodified panels. According to Sensogut *et al.* (2009), adding borax to UF resin had no influence on the bending strength and extraction shear strength characteristics of plywood bonded with this resin, but it did reduce the free formaldehyde content. According to Zhang *et al.* (2011), increasing the NCC loading levels up to 1,5 % reduced the formaldehyde emission

values of the plywood panels. It was also found that increasing the NCC content enhanced the bonding strength of plywood panels by up to 1 %. According to Candan and Akbulut (2014), it is feasible to achieve wood composites with enhanced physical and mechanical characteristics by utilizing different nanomaterials. According to Candan and Akbulut (2014), it is feasible to achieve wood composites with enhanced physical and mechanical characteristics by utilizing different nanomaterials. According to Donmez Cavdar *et al.* (2015), composites containing BX have better mechanical properties than composites containing BA. According to Ayrilmis *et al.* (2016), using MFC in UF adhesives can be an ecologically positive move for lowering volatile organic compounds (VOC) in wood-based composites. According to Donmez Cavdar *et al.* (2018), the performance of BX-containing composites was greatly improved. According to Lengowski *et al.* (2021), adding NFC to the PF resin increased viscosity and lowered gel time while maintaining the pH unchanged. According to Yildirim and Candan (2021), particleboard panels with superior physical and mechanical qualities may be produced using CNF and BA. There is a scarcity of scientific data on the combined effect of nanocellulose and boron compounds on wood composites. There is no published research on the combined effects of nanocellulose and borax in MUF adhesive. The research aimed to verify the hypothesis that altering MUF adhesive with the combined effects of CNF and BX can improve physical and mechanical characteristics while decreasing formaldehyde emissions in plywood manufacturing.

## **Material and methods**

## **Materials**

Rotary-cut beech veneers, MUF adhesive, liquid ammonium sulfate, and flour were supplied by Vezirkopru Forest Products and Paper Industry Inc., located in Samsun, Türkiye.

### **Wood materials**

In the laboratory, five-layered plywood (5-ply) samples were produced using rotary-cut beech veneers (400 mm × 400 mm × 1,5 mm). The materials utilized were beech (*Fagus orientalis* L.) veneers.

### **Additive**

CNF and BX were utilized as reinforcing additives in the MUF composition. The CNF was supplied by the AEWCA Advanced Structures and Composites Center at the University of Maine in Orono, Maine, USA. CNF has a white color and is made up of string-like particles that are

10-20 nm wide and 2-3  $\mu\text{m}$  length. The BX, which is referred to as sodium tetraborate decahydrate, was supplied by Eti Maden.

## Adhesive

MUF resin with a solid content of 62 %, liquid ammonium sulfate  $(\text{NH}_4)_2\text{SO}_4$  with a solid concentration of 20 %, and flour. Ammonium sulfate was utilized as a hardener, while flour was utilized as a filler. The MUF resin is liquid, milky white in color, has a pH of 7-8, a viscosity of 60-160 cP, and a specific gravity of 1,190-1,196  $\text{g}/\text{cm}^3$ .

## Methods

The experimental design of the CNF/BX-reinforced melamine-urea-formaldehyde adhesive is shown in Table 1.

**Table 1:** The experimental design of the study.

Groups	Cellulose Nanofibril (%)	Borax (%)
Control	0	0
A	1	1
B	1	3
C	1	5
D	3	1
E	3	3
F	3	5

### **The production of plywood panels**

The adhesive reinforcement process was carried out in the Nanotechnology Laboratory at Istanbul University-Cerrahpasa. The MUF adhesive used to manufacture plywood panels was modified with CNF at 1 %, 3 %, and 5 % of the weight of the MUF adhesive liquid solution, and BX at 1 %, 3 %, and 5 % of the weight of the adhesive liquid solution. CNF, BX, ammonia sulfate, and flour were blended into the MUF adhesive using an ultrasonic homogenizer (Hielscher GmbH, Germany). The resin mixes were applied to the surface of the veneers at a rate of 180 g/m<sup>2</sup>, and the panels were pressed in a hydraulic hot press for 8 minutes at a pressure of 2 MPa and a temperature of 120 °C (Cemil Usta Company, Model: SSP 180, Istanbul, Türkiye). After being hot-pressed, plywood panels were conditioned at 20 °C ± 2 °C and a relative humidity of 65 % ± 5 % before being tested for physical, mechanical, and formaldehyde emissions.

### **Physical analysis**

The density of the plywood panels was calculated via TS EN 323 (1999). The thickness swelling (TS) of plywood panels was calculated via TS EN 317 (1999). The water absorption (WA) of the plywood panels was calculated via ASTM D1037-06 (2006). The thickness of all TS and WA samples was measured and weighed. Samples were immersed upright in a water bath heated to  $20\text{ }^{\circ}\text{C} \pm 1\text{ }^{\circ}\text{C}$ . The TS and WA values of the panels were measured after 2 and 24 hours of water immersion, respectively. The moisture content (MC) of the plywood panels was calculated via TS EN 322 (1999). Before weighing, the TS and WA samples were dried in an oven at  $103\text{ }^{\circ}\text{C}$  for 24 hours. The MC values of the samples after 2 hours and 24 hours of water soaking were measured using the oven-dry weight of each sample.

### **Mechanical analysis**

Mechanical performance of plywood samples were tested at the testing machine (IB600, IMAL, Modena, Italy) by Starwood Forest Products Inc., Bursa, Türkiye.

The modulus of rupture and modulus of elasticity of fifteen plywood panels were calculated via TS EN 310 (1999). MOR and MOE samples were cut parallel and perpendicular to the panel's fiber direction. The tests were also carried out separately, with the results calculated. strength measurements have been performed on fifteen plywood panel samples. TS 3969 EN 314-1 (1998) defines bonding strength testing as a way of measuring the bonding quality of plywood panels. The relevant requirements are specified in TS EN 314-2 (1999). Unpretreated samples (dry) and pre-treated samples (wet) were tested for bonding strength. Bonding strength was



measured in dry conditions and in wet conditions after soaking in a water bath with a temperature of  $20\text{ }^{\circ}\text{C} \pm 1\text{ }^{\circ}\text{C}$  for 24 hours.

### **Formaldehyde emission analysis**

Formaldehyde emission tests on plywood panels were done in following the standard TS 4894 EN 120 (1999). A perforator and a UV spectrophotometer (Nova 60, Merck, Darmstadt, Germany) were used to calculate the formaldehyde emission of the CNF-BX-reinforcing plywood panels. This test describes a perforator extraction method for measuring the formaldehyde content of wood-based composites.

## **Results and discussion**

### **Physical properties**

The average density, thickness swelling, water absorption, and moisture content values of CNF/BX-reinforced plywood panels after 2 h and 24 h of water soaking are shown in Table 2. The standard deviation values are shown in parentheses. There were no significant differences in thickness swelling, water uptake, and moisture content between the panels.

**Table 2:** Average physical properties values of CNF/BX-reinforced plywood panels.

Groups	Density (kg/m <sup>3</sup> )	Thickness Swelling 2 h (%)	Thickness Swelling 24 h (%)	Water Absorption 2 h (%)	Water Absorption 24 h (%)	Moisture Content 2 h (%)	Moisture Content 24 h (%)
Control	670	3,30 (0,45)	7,41 (0,89)	22,79 (3,50)	49,09 (4,42)	34,13 (4,41)	62,97 (5,68)
A	740	3,14 (0,36)	7,17 (0,57)	16,08 (1,87)	44,68 (2,98)	28,53 (2,55)	59,20 (4,30)
B	700	3,22 (0,42)	7,20 (0,78)	18,36 (2,83)	44,75 (3,02)	30,46 (2,23)	59,76 (4,42)
C	710	3,50 (0,47)	8,09 (1,02)	20,40 (3,26)	53,76 (5,37)	33,21 (3,67)	68,33 (7,14)
D	730	2,69 (0,29)	6,67 (0,52)	17,72 (2,51)	44,62 (2,76)	29,11 (2,78)	56,99 (3,77)
E	710	3,11 (0,34)	6,79 (0,55)	18,61 (2,87)	45,32 (3,32)	28,25 (2,41)	56,85 (3,59)
F	720	3,16 (0,38)	8,82 (1,37)	23,32 (4,84)	57,71 (7,92)	29,54 (2,99)	64,33 (6,15)

### Density

The average density of the CNF/BX-reinforced plywood panels ranged from 700 to 740 kg/m<sup>3</sup>.

### Thickness swelling

As shown in Table 2, the thicknesses of plywood panels after 24 hours were F > C > Control > B > A > E > D. Except for Groups C and F, the findings show clearly that the TS values of the

plywood panels were much lower than those of the unreinforced panels for both the 2 h and 24 h water soaking periods. After 2 and 24 hours of soaking, Group D had approximately 5 % and 9 % reduced TS. When the CNF % 1 and BX loading levels were increased to 5 % during a 24-hour water soaking period, the TS values of the reinforced panels increased by 9,17 % compared to the unreinforced panels. As the CNF 3 % and BX content were increased to 5 % during a 24-hour water soaking period, the TS values of the reinforced samples increased by 19,02 % compared to the unreinforced panels.

The TS values of the panels decreased with increasing CNF content. Because CNF has a larger surface area, it improves interfacial adhesion and reduces composite swelling. However, when BX content increased, so did the TS values. Because BX causes higher TS in wood-based composites, it should not be used at a loading level of more than 1 %.

Similar findings have been reported by other researchers. Veigel *et al.* (2012) the thickness swelling values of the UF-bonded particleboard panels modified with 1 % NC were lower. Ayrilmis (2013), as the boron compound content increases, so does the dimensional stability of the composite.

### **Water absorption**

Plywood panels followed the same trend as thickness swelling after 24 h, with  $F > C > \text{Control} > B > A > E > D$ . Except for Groups C and F, the findings clearly reveal that the WA values of the plywood panels were much lower than those of the unreinforced panels over the 24 h water soaking period. When compared to the unreinforced panels, Group D showed the greatest decline in 24 h WA values of 10,01 %. The WA values of the panels after 2 h and 24 h water

immersion increased as the CNF and BX loading levels increased. As the BX loading levels were increased to 5 % during a 24h water soaking period, the WA values of the reinforced panels increased by 9,51 % compared to the unreinforced panels. As the CNF content was increased to 3 % and the BX content was increased to 5 % for the 24 h water soaking period, the WA values of the reinforced panels increased by 17,55 % compared to the unreinforced panels.

This was expected, and it can be linked to the presence of CNF, which helped with water absorption. CNF is particularly hydrophilic due to the presence of hydroxyl (OH) groups on its surface.

Similar results were found by other authors. According to Kawalerczyk *et al.* (2020) nanocellulose could be used as a resin modifier in the production of water-resistant plywood.

### **Moisture content**

As shown in Table 2, the MC of plywood panels after 24 hours were  $C > F > \text{Control} > B > A > D > E$ . Except for Groups C and F, the results show that plywood panels have dramatically lower MC values than unmodified panels during a 24 h water immersion duration. The MC values of the panels increased as the boron compound content increased. When compared to unreinforced panels, plywood panels reinforced with Group E showed the highest reductions in 2 h and 24 h MC values of 28,20 % and 15 %, respectively.

Similar results have been reported by other researchers. According to Hansted *et al.* (2019), adding nanocellulose to medium density particleboard panels resulted in no statistically significant variation in moisture content.

## Mechanical properties

The average modulus of rupture, modulus of elasticity, and bonding strength values of CNF/BX-reinforced plywood panels are shown in Table 3. The standard deviation values are shown in parentheses. There were no significant differences in MOR, MOE, and BS between the panels.

**Table 3:** Average mechanical properties values of CNF/BX-reinforced plywood panels.

Groups	Modulus of Rupture (MPa) Parallel	Modulus of Rupture (MPa) Perpendicular	Modulus of Elasticity (MPa) Parallel	Modulus of Elasticity (MPa) Perpendicular	Bonding Strength (MPa) Dry	Bonding Strength (MPa) Wet
Control	74,13 (9,23)	37,20 (3,62)	8,097 (804,66)	2,506 (266,75)	1,77 (0,26)	1,45 (0,29)
A	88,91 (12,98)	45,41 (10,27)	8,793 (944,01)	2,884 (612,05)	2,78 (0,94)	2,04 (0,76)
B	70,67 (7,33)	38,55 (6,62)	7,776 (787,02)	2,551 (338,22)	2,12 (0,37)	1,45 (0,31)
C	55,08 (6,68)	46,98 (11,67)	5,764 (418,66)	3,180 (951,56)	2,38 (0,82)	1,05 (0,27)
D	74,67 (9,68)	41,43 (8,70)	7,554 (710,20)	2,673 (562,03)	2,17 (0,56)	1,47 (0,57)
E	78,67 (10,25)	39,91 (7,98)	8,111 (810,94)	2,671 (549,93)	2,50 (0,85)	1,33 (0,41)
F	72,10 (8,58)	38,16 (5,19)	7,547 (636,14)	2,547 (435,51)	2,08 (0,31)	1,45 (0,54)

## Modulus of rupture

As the BX loading levels increased from 1 % to 5 %, the MOR (parallel to grain) values of the plywood panels were reduced. Group 2 performed best in modulus of rupture parallel to the grain, with a 19,93 % improvement. When it came to the modulus of rupture perpendicular to the grain, the plywood panels outperformed the control panels. The MOR of the panels improved when the BX content was lowered from 5 % to 1 %. The modulus of rupture values in parallel samples were larger than those in perpendicular samples. This happened because the load apparatus's load direction was perpendicular to the grain direction of the surface layers of the parallel samples but parallel to the grain direction of the perpendicular samples.

Similar findings have been reported by other authors. Leng *et al.* (2017) observed that the CNF addition ratio had the greatest impact on the MOR. Claramunt *et al.* (2019) found that adding either type of nanocellulose increased the MOR results. The MOR of nanocellulose-based plywood was remarkably increased, according to Kawalerczyk *et al.* (2021).

### **Modulus of elasticity**

The MOE values of the parallel samples were greater than those of the perpendicular samples. This was due to the load apparatus's load direction being perpendicular to the grain direction in the surface layers of the parallel samples, whereas it was parallel to the grain direction in the surface layers of the perpendicular samples. In terms of perpendicular to grain findings,

reinforced plywood panels outperformed the control panels. The maximum improvement in the perpendicular sample reinforced with 1 % CNF/5 % BX was determined to be 26,89 %.

Other writers have reported similar results. According to Efhamisisi *et al.* (2016), the inclusion of boron enhanced the MOE of the adhesive. Sun *et al.* (2019) reported that the CNF boosted the MOE, with a 2,5 % increase being the best.

### **Bonding strength**

Bonding strength testing is an important indicator of adhesive behavior in plywood panels. The bonding strength values of untreated (dry) and pre-treated (wet) CNF/BX-reinforced plywood panels or unreinforced plywood panels are shown in Table 3.

When compared to the control group, all reinforced panel groups resulted in higher bonding strength values than untreated (dry). The highest improvement was 57,06 % with Group A. CNF enhances the bonding strength of plywood panels. Because of its large specific surface area, high specific stiffness, and aspect ratio, CNF forms strong ties. Mechanical performance requirements have improved as a result of the addition of CNF.

Except for the panels reinforced with Group C and E, the pre-treated sample (wet) values of the plywood panels had higher bonding strengths than the unreinforced panels. With Group A, the biggest improvement was 40,68 %. Wetting pre-treatment Group A panels demonstrated sufficient water resistance, as wet bonding strength was equivalent to dry bonding strength.

Other writers have reported similar results. Kasmani and Samariha (2019) reported that adding 8 % NFC enhanced bonding strength by roughly 10,9 % compared to 0 % NFC. Kawalerczyk *et al.* (2021) increased the bonding strength of UF resin by adding NCC.

## Formaldehyde content

Table 4 shows the formaldehyde emission values of CNF/BX-reinforced plywood panels or unreinforced plywood panels.

**Table 4:** Average formaldehyde content values of CNF/BX-reinforced plywood panels.

Groups	Formaldehyde Content (mg/100 g oven dry board)
Control	0,30
A	0,20
B	0,22
C	0,40
D	0,21
E	0,25
F	0,48

Except for Group C and Group F, the results show that the formaldehyde content of the plywood panels was much lower than that of the unmodified panels. The formaldehyde content of Control Group was 0,30 mg/100 g. When adding 1 % CNF and 1 % BX, the formaldehyde content of plywood bonded by the developed Group A decreased by 33,33 % to 0,20 mg/100 g. When adding 3 % CNF and 1 % BX, the formaldehyde content of plywood bonded by the developed Group D decreased by 30 % to 0,21 mg/100 g.

As the BX additions increased, the formaldehyde content of the plywood began to rise slightly. The chemical nature of BX may have increased the release of free formaldehyde, which was a significant component of plywood formaldehyde emissions. As a result, when the BX addition



exceeded 1 %, the formaldehyde content increased. CNF, on the other hand, can absorb free formaldehyde due to its large specific surface area, lowering the resin's free formaldehyde content and the formaldehyde emission of the produced plywood. The CNF could absorb free formaldehyde from MUF resin as a shielding effect, and its barrier properties decreased panel formaldehyde emissions.

Similar findings have been reported by other researchers. Colak and Colakoglu (2004) determined that the formaldehyde emission values of plywood panels were affected differently by various boron compounds. According to Ayrimis *et al.* (2016), the application of MFC with UF resin is an environmentally favorable approach for lowering formaldehyde and VOC emissions from wood-based panels.

## **Conclusions**

Physical and mechanical properties are important performance features for plywood panels used as building and structural composites in outdoor applications. Furthermore, exposure to formaldehyde emissions in the home can be extremely hazardous to one's health. As a result, reducing formaldehyde emissions is crucial.

The combined impacts of CNF/BX on the performance properties of plywood panels were investigated in this study. The results of this study showed that the synergistic effects of CNF and BX reinforcement had a major influence on the physico-mechanical properties and formaldehyde emission of plywood panels. The panels produced with a 3 % CNF/BX had the

lowest TS, WA, and MC values while achieving the highest MOR, MOE, and BS values. The panels manufactured with a 1 % CNF/BX combination had the lowest formaldehyde content. The use of CNF and BX combined has the potential to produce high-performance green plywood panels for construction, furniture, interior, and exterior materials. Future studies should focus on the synergistic effect of different sustainable lignocellulosic bionanomaterials and different boron compounds on high-performance and environmentally friendly wood composites.

#### **Authorship contributions**

M. Y.: Conceptualization, data curation, formal analysis, investigation, methodology, writing – original draft. Z. C.: Supervision, writing – review & editing. T. A.: Supervision. D. J. G.: Supervision. S. M. S.: Supervision

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