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The Effect of Hot-Cold Shock Test on Changes in Gloss and Roughness in Epoxy Resin Coated Wood Material

Utjecaj testa vruće-hladne provjere na sjaj i hrapavost drvnog materijala premazanog epoksidnom smolom

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ABSTRACT • *In this study, epoxy resin was applied as a surface treatment material to wood species of different specific gravity: Scots pine (Pinus sylvestris L.), Turkish beech (Fagus orientalis Lipsky) and sweet chestnut wood (Castanea sativa Mill.). It aimed to determine the changes in roughness values (in accordance with TS 2495 EN ISO 3274 standard and TS 6212 EN ISO 4288 standard) and gloss values (in accordance with TS 4318 EN ISO 2813 standard) in the test specimens pre-post hot and cold shock effect. Surface-treated test specimens were first kept in a fan drying oven at (50 ± 5) °C for an hour and then kept at (-20 ± 2) °C for an hour, in accordance with ASTM D1211-97. All processes were regarded as a cycle, and the experiments continued until 15 cycles were carried out. Then, the gloss and roughness values were determined and analysed with the control specimens in the MSTAT-C statistical program. According to the results, all gloss values of epoxy resin-coated wooden material surfaces decreased after the hot-cold shock experiment, whereas a decrease in roughness was detected. It is thought that this research will contribute to advising the firms that import furniture to nations with various climatic conditions about the physical properties of surface-treated products.*

KEYWORDS: *wood; epoxy resin; gloss; hot-cold shock test; surface roughness*

SAŽETAK • *U ovom je istraživanju za površinsku obradu drva različite gustoće – bora (Pinus sylvestris L.), kavkaske bukve (Fagus orientalis Lipsky) i pitomog kestena (Castanea sativa Mill.), odabrana epoksidna smola. Cilj je bio na ispitnim uzorcima prije i nakon testa vruće-hladne provjere utvrditi promjene hrapavosti (prema standardima TS 2495 EN ISO 3274 i TS 6212 EN ISO 4288) i sjaja (prema standardu TS 4318 EN ISO 2813). Površinski obrađeni ispitni uzorci prethodno su jedan sat sušeni u sušioniku s ventilatorom na 50 ± 5 °C, a zatim su jedan sat hlađeni na –20 ± 2 °C, prema standardu ASTM D1211-97, i taj je postupak ponavljan u 15 ciklusa. Nakon toga u statističkom su programu MSTAT-C određene i analizirane vrijednosti sjaja i hrapavosti svih uzoraka, uključujući i kontrolne. Rezultati su pokazali da su se sjaj i hrapavost drvenih površina premazanih epoksidnom smolom nakon vruće-hladne provjere smanjili. Ovo će istraživanje pridonijeti spoznajama o fizičkim svojstvima površinski obrađenih proizvoda od drva koji se uvoze u brojne zemlje u kojima su klimatski uvjeti različiti.*

KLJUČNE RIJEČI: *drvo; epoksidna smola; sjaj; vruće-hladna provjera; hrapavost površine*

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1 INTRODUCTION
1. UVOD

To protect wood and wood-based products, surface treatments are essential for long-term and efficient use. Especially in wood products, the treatments comprise processes like impregnation, varnishes, and paints (Vardanyan *et al.,* 2015). The varnish, which can form a film layer for the protection of wood, contains basically synthetic, cellulosic and polyurethane varieties; nitrocellulose varnish, alkyd-urethane varnish, acrylic varnish, oil varnish, and water-based varnish are also available as an advanced version of the varnish. The selection of surface treatment and protective layer are essential when wood material is intended to last more extended and be used more appropriately (Ulay and Budakci, 2015). Scientific studies are carried out to examine some physical properties of varnish types after their application to different wood and wood-based material surfaces.

Altiparmak (2017) treated *Terminalia* superba, sweet chestnut (*Castanea sativa* Mill.), and sapele (*Entandrophragma cylindricum*) woods with polyurethane, acrylic, and synthetic varnishes and determined that the gloss increased with the increase in the layer degree (Altiparmak, 2017). After passing natural ageing tests, the Monterey pine (Pinus radiata) wood species test specimens were evaluated using water-based and solvent-based varnished materials. It was determined that the specimens subjected to 500 hours of accelerated aging showed very little gloss and colour variations (Garay *et al.,* 2017). The gloss values of the water-based varnishes were determined to have lower values when compared with the solvent-based varnishes (Yakin, 2001). It was discovered that the wood type was insignificant in terms of the gloss values for varied varnish layers, but the impact of the varnish type was found to be substantial (Budakci, 2003).

The effects of dry heat and artificial heat exposure on the gloss values of specimens of black alder (*Alnus glutinosa* L.) treated with two different types of varnish were examined. In a study comparing the gloss values of specimens coated with UV and water-based varnish, specimens coated with UV varnish had better gloss values than specimens coated with water-based varnish (Salca *et al*., 2021). The study investigated the decorative properties of European beech (*Fagus sylvatica* L.) and Persian walnut (*Juglans regia* L.) specimens. While specimens coated with hydro oils had a visibly higher gloss value in both wood specimens after the oil treatment, control specimens were determined to have lower gloss values (Palija *et al*., 2021). The surface durability of different varnish layers applied in the wood industry was investigated with regard to dry temperature, cigarette fire, wet temperature and gloss. In the study, it was determined that the surfaces with polyurethane (silk mat, matte, filler) and cellulosic filler varnish were resistant to the temperature of 100 °C (dry temperature), while the durability of the synthetic, polyester, cellulosic mat and glossy varnishes was not as good as assumed (Sonmez, 1989). It has been determined that dry heat causes gloss and colour changes on laminate flooring and solid wood. However, it has been determined that the degree of impact on laminate surfaces is lower (Dongel *et al.,* 2008). In their study, Atilgan and Sofuoglu (2023) used varnished wood species of different specific gravity with varnishes frequently used in the industry and exposed them to hot-cold shock tests. They determined a decrease in gloss values and an increase in Rz roughness values of wooden material surfaces after hot-cold shock.

Hot and cold experiments are important indicators for the flexibleness of translucent layers of varnish on the surfaces of wood and their immunity to sudden temperature variations (Sonmez and Kesik, 1999). In their study, Sonmez and Kesik (1999) prepared the test specimens using Turkish beech wood (*Fagus orientalis* Lipsky), Scots pine wood (*Pinus sylvestris* L.) and sessile oak wood (*Quercus petraea* L.) and cellulosic, polyurethane, and acrylic varnish types were put on the specimen surfaces. The test specimens were initially incubated at a temperature of -18 °C for an hour and afterwards at a temperature of 50 °C for another hour. These procedures were regarded as a cycle, and the experiments were carried out up to twenty cycles. The test results determined that, while the gloss values of the oak wood specimens covered with cellulosic varnish have risen, the gloss values of the others have declined (Sonmez *et al.,* 1999). In addition, the cellulosic varnished surface has been stated to show breakage, cracking and colour change when Gubas wood (*Endospermum peltatum* Merr.) is exposed to the hot-cold control experiment through the application of acidic hardening varnish and cellulosic varnish to the wood material surface (Yolanda, 1998). In their study, Budakci *et al*. (2010) applied polyurethane, cellulosic, and acrylic varnishes on the test specimens obtained from the oriental beech, Scots pine and sessile oak, and colour variations were detected following the impacts of accelerated aging and after hot-cold shock methods (Budakci *et al.,* 2010).

Altiparmak (2017) studied the distortion on the surfaces of specimens following the hot-cold shock test with respect to yacht varnish, polyurethane varnish and epoxy varnish types, which were applied on *Terminalia superba*, sweet chestnut (*Castanea sativa* Mill.) and sapele (*Entandrophragma cylindricum*) wood specimens. After 20 cycles of cold check test, no surface cracks or deformations were found on the panel surfaces (Altiparmak, 2017).

Based on the analysis of the hot-cold aging tests conducted on the varnish layers, it is suggested that the wood specimens should be held at the temperatures of -20 and 50 °C for an hour each, and this procedure can be considered as a single period; when the layers do not show any deterioration for ten periods, this should be regarded as satisfactory, and the performance of layers without deterioration in 25 periods should be regarded as an above average performance (Payne, 1965).

In the world, furniture, yachts, etc., made of wood materials are produced in countries and continents using different varnishes. The products are imported or exported. Turkey exports furniture worldwide, from the EU, the USA, and Middle and Far Eastern countries to the Russian Federation.

The sector representatives report a problem: some of the physical properties of the wood material used in furniture, various wood products, and yacht and boat decorations change during the transportation of these products to countries with different climatic conditions. Based on the data obtained from the change in surface roughness and gloss on varnished surfaces, which may be exposed to the effects of sudden climatic changes (hot and cold), it will be possible to select the appropriate type of wood and varnish for the place of use.

The wood species used in the present study were Scots pine (*Pinus sylvestris* L.), sweet chestnut (*Castanea sativa* Mill.) and Turkish beech (*Fagus orientalis* L.), as well as epoxy resin, which is used in the woodworking and furniture industries and the manufacture of various marine vehicles. These tree species and epoxy resin can be deformed in environments where they are exposed to sudden temperature changes (especially furniture, marine vehicles, yachts, etc.). This study aimed to determine the effect of the hot-cold check test on the changes in surface roughness and gloss and to determine the most suitable wood species.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

In this study, Scots pine wood (*Pinus sylvestris* L.), Turkish beech wood (*Fagus orientalis* Lipsky) and sweet chestnut wood (*Castanea sativa* Mill.) were used as the wood materials; transparent pour type epoxy resin (Arc brand 150 ultra-transparent casting type glossy epoxy resin) was used as the surface treatment material. The timbers used in the research were randomly selected from Afyonkarahisar Timber Management. The knot-free parts of the lumber were used as specimens. The lumber was carefully selected to ensure that it was dry, smooth-grained, and free of fungi and insects.

Wood specimens were prepared for gloss and surface roughness tests in dimensions of 100 mm × 100 mm × 10 mm (tangential, longitudinal, radial). Three wood types, one varnish type and five repetitions were prepared for each parameter, i.e. a total of 15 test specimens with regard to ISO 3129 (2019). 75 records were acquired in all tests by measuring 5 different objectives on each test specimen. They were kept at (20±2) °C / (65±2) % conditions to reach air dry (12±2) % humidity until they reached a constant weight in accordance with TS ISO 13061-1, 2021 standards (TS ISO 13061-1, 2021).

Before the test specimens were coated with epoxy resin, their surfaces were sanded with sandpaper No. 80 and 100, respectively. The varnish procedure for the test specimens was conducted by the ASTM-D 3023 principles (ASTM-D 3023, 2017). Epoxy resin was applied with a fine-wire brush in a single layer to be 125 g/m^2, and specimens were left to dry for 24 hours.

This test was carried out on the wood material on which paint and varnish were applied by exposing it to sudden cold and heat. The aim was to identify the performance of varnish layers in sudden temperature variations in natural weather conditions. Within this framework, the test specimens, whose surfaces were treated, were arranged in 100 mm × 100 mm × 10 mm dimensions, and they were placed in the cabinet in an upright position, making sure that they were not touching each other and facing the heating/cooling wall after gloss and roughness measurements were made. First, they were kept in the oven at (50±5) °C for an hour. At the end of this period, the test specimens were transferred to the cooling unit, which was calibrated to (−20 ± 2) °C within 1 minute and kept there for 1 hour. Afterwards, the test specimens were taken from the cabin and left to rest for 15 minutes. All procedures were regarded as a cycle, and the processes continued until there were 15 cycles. The roughness and gloss values of the specimens, which were kept at the normal room temperature for 1 hour, were measured after the test (ASTM D 1211-97, 2001).

The gloss values of the test specimens were measured with a measuring device (Konica Minolta Multi Gloss 268 Plus) with respect to TS 4318 EN ISO 2813 by using the light-reflecting property of the wood material treated with epoxy resin. The selected geometry was measured at an incidence angle of 60° (TSE EN ISO 2813). The device calibration is performed in accordance with ASTM-D-523 (ASTM D523-14). The error rate is reduced by measuring at an angle of 60° on dull and glossy surfaces (Ozen and Sonmez, 1990; Ordu and Sofuoglu, 2016). Five repetitions were conducted for each test group. Five measurements were made on each specimen. Tests were carried out on the specimen surfaces in a parallel direction, and the arithmetic averages of these measurements were recorded using the gloss value. Concerning the gloss measurements conducted at 60°, the surfaces were classified as matte, semi-matt,

1 – Preparation of specimens *1 – Priprema uzoraka*
2 – Test specimens *2 – Iispitni uzorci*
3 – Conditioned cabinet *3 – Komora za kondicioniranje*
4 – Surface treatment application *4 – Površinska obrada*

8 – Results *8 – Rezultati*
7 – Statistical analysis *7 – Statistička analiza*
6 – Surface brightness (gloss) and roughness *6 – Sjaj i hrapavost površine*
5 – Hot-cold shock test *5 – Vruće-hladna provjera*

Figure 1 Schematic representation of methods used during the experiment
Slika 1. Shematski prikaz metoda primijenjenih tijekom eksperimenta

semi-gloss, gloss, and very gloss. The experimental process of the study is given in Figure 1.

Surface roughness tests were carried out using the tactile (needle) scanning method and Time TR220 (Time Group Inc., China). Through the measurements, 0.5 mm/s tracking speed, 2.5 mm pickup length (λc), 5 µm stylus radius and 90° stylus angle operations were performed. The surface roughness values were determined with a precision of ±0.01 µm. The specimen roughness was measured using the mean peak-to-valley height (*Ra*). These measurements were all carried out in accordance with EN ISO 3274, TS 2495, and EN ISO 21920-3 principles. A computerised statistical tool called MSTAT-C was used to examine the test findings. It includes variance analysis and a 95 % confidence level by Duncan's test. Homogeneity groups (HG) were the subject of statistical analyses, with different letters denoting statistical significance.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

The results of multiple variance analysis regarding the effects of wood type and hot-cold shock test on gloss values are given in Table 1.

The hot-cold shock test and wood type have been found to be statistically significant (α=0.05) in terms of gloss value impacts and insignificant in terms of interactions (AB). The Duncan's test results, which were performed to determine between which groups the difference was significant, are submitted in Table 2.

Table 1 Multiple variance analysis regarding the effects of hot/cold shock test and wood type on gloss values
Tablica 1. Analiza višestruke varijance s obzirom na utjecaj vruće-hladne provjere i vrste drva na vrijednosti sjaja

Variance source *Izvor varijance*	**Degree of freedom** *Stupanj slobode*	**Sum of squares** *Zbroj kvadrata*	**Mean square** *Srednji kvadrat*	***F* score**	***P*<0.05**
Wood type (A) / *vrsta drva (A)*	2	1121.136	560.568	92.077	0.0004
Hot/cold shock test (B) *vruće-hladna provjera (B)*	1	1725.921	1725.921	283.494	0.0000
AB	2	61.581	30.791	0.5058	
Error / *pogreška*	54	3287.541	60.880		
Total / *ukupno*	59	6196.180			

Table 2 Results of gloss Duncan's test
Tablica 2. Rezultati Duncanova testa za vrijednosti sjaja

	Process type / *Vrsta procesa*	***X* (Gloss)** *X (sjaj)*	**HG**
*Wood type + **Hot/ cold shock test **Vrsta drva + **vruće-hladna provjera*	Scots pine (control) / *borovina (kontrola)*	94.01	A
	Sweet chestnut (control) / *drvo pitomog kestena (kontrola)*	88.80	AB
	Turkish beech (control) / *kavkaska bukovina (kontrola)*	84.26	BC
	Scots pine (control) / *borovina (nakon ispitivanja)*	82.55	BC
	Sweet chestnut (control) / *drvo pitomog kestena (nakon ispitivanja)*	80.84	C
	Turkish beech (control) / *kavkaska bukovina (nakon ispitivanja)*	71.50	D

*LSD**= 4.937 *LSD***=4.031 *LSD****=6.982

The highest gloss value was obtained in the Scots pine control (94.01), and the lowest was obtained after the Turkish beech + hot/cold shock test (71.50). After applying the hot-cold shock test, the gloss values of the wood type treated with epoxy resin decreased significantly compared to the control groups.

Table 3 presents the findings of a multiple variance analysis concerning the impact of wood type and the hot-cold shock test on the roughness values.

Wood type (A) and its interactions (AB) were found to be statistically insignificant (α=0.05) in terms of its impact on roughness value, while they were found to be significant in terms of hot-cold shock test (B). The Duncan's test results, which were performed to determine between which groups the variations were significant, are given in Table 4.

The highest roughness value was obtained in Turkish beech control (0.0745), and the lowest was obtained after the Scots pine + hot/cold shock test (0.012). In epoxy resin-treated wood types, roughness values decreased after the hot-cold shock test compared to the control groups.

The heat-treated studies in the literature found that the heat treatment changed the gloss values (Ayata *et al.,* 2018; Cavus *et al.,* 2018). Korkut *et al.* (2023) found that glossiness values decreased by heat treatment when perpendicular and parallel glossiness values of heat-treated wild cherry (*Prunus avium*) wood at 212 °C for 1.5 hours and 2.5 hours (ThermoWood method) were compared with the control specimens.

The average roughness (*Ra*) has been determined to decrease in all test specimens following the hot-cold shock test. The highest roughness reduction (78.66 %) has been detected in the Turkish beech wood, while less reduction in roughness was observed in other wood types. It can be agreed that there are significant variations in pre- and post-hot-cold shock tests. Exposing epoxy resin to heat again after curing may have contributed to stretching the set structure (becoming solid, hard and tough) and led to the smooth surface by bringing the molecules closer together. The roughness decrease observed in the study may be due to increased mechanical and chemical bonding between the epoxy resin and wood material. This is in line with the studies

Table 3 Multiple variance analysis regarding the effects of hot/cold shock test and wood type on roughness values
Tablica 3. Analiza višestruke varijance s obzirom na utjecaj vruće-hladne provjere i vrste drva na vrijednosti hrapavosti

Variance source *Izvor varijance*	**Degree of freedom** *Stupanj slobode*	**Sum of squares** *Zbroj kvadrata*	**Mean square** *Srednji kvadrat*	***F* score**	***P*<0.05**
Wood type (A) / *vrsta drva (A)*	2	0.003	0.001	1.7759	0.1791
Hot/cold shock test (B) / *vruće-hladna provjera (B)*	1	0.028	0.028	35.3543	0.0000
AB	2	0.002	0.001	1.1062	0.3382
Error / *pogreška*	54	0.043	0.001		
Total / *ukupno*	59	0.076			

Table 4 Results of Duncan's test for surface roughness (Ra)
Tablica 4. Rezultati Duncanova testa za vrijednosti hrapavosti (*Ra*)

	Process type / *Vrsta procesa*	***X*, *µ*m**	**HG**
*Wood type + **Hot/cold shock test **Vrsta drva + **vruće-hladna provjera*	Scots pine (control) / *borovina (kontrola)*	0.0745	A
	Sweet chestnut (control) / *drvo pitomog kestena (kontrola)*	0.0503	A
	Turkish beech (control) / *kavkaska bukovina (kontrola)*	0.0470	A
	Scots pine (control) / *borovina (nakon ispitivanja)*	0.0159	B
	Sweet chestnut (control) / *drvo pitomog kestena (nakon ispitivanja)*	0.0140	B
	Turkish beech (control) / *kavkaska bukovina (nakon ispitivanja)*	0.0120	B

*LSD**= 0.02001 *LSD***=0.01634

in the literature, which state that the surface roughness in the treated wood materials decreases with the impact of heat treatment.

4 CONCLUSIONS
4. ZAKLJUČAK

The wood industry faces a problem when shipping surface-treated products. This problem is related to the physical properties of the product surfaces changing due to sudden temperature changes.

Determining the elasticity in the varnish layers on the wooden surfaces or the physical deformation caused by sudden temperature changes is considered significant for solving the problem. Therefore, epoxy resin was put on Turkish beech, Scots pine, and sweet chestnut surfaces as a surface treatment material. Exposing the test specimens to -20 and +50 hot-cold shock test conditions for 1 hour was accepted as one cycle. In total, this process was continued for up to 15 cycles. Gloss and roughness measurements were carried out on the treated wooden surfaces to determine the distortions occurring on the surface before and after the experiment.

It may be stated that changes occurring in the surface roughness and gloss values of the varnished wood material after the hot-cold shock test have not affected the surface characteristics and usage functions of test specimens but have just affected aesthetics. In this respect, the producers and consumers should be informed that surface variations like this can occur in the epoxy, which is used in all three types of wood, the furniture, which can be traded to countries with varied climatic factors, and the finishing material, which is used in marine vehicles like yachts/boats.

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Effect of Heat Treatment and Densification Temperature on Roughness, Hardness and Spring-back in Cylindrical Wood Densification

Utjecaj toplinske obrade i temperature ugušćivanja na hrapavost, tvrdoću i elastični povrat pri cilindričnom ugušćivanju drva

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ABSTRACT • *In this research, black pine (Pinus nigra Arnold) was shaped into cylindrical forms through a turning process and then underwent surface densification. The densification process was performed on a lathe using a spinning roller specifically designed and produced for this purpose. Heat-treated (3 hours at 160 °C) and untreated specimens (a large diameter of 33 mm and a small diameter of 21 mm) were used as test material. Two feed rates, low (0.05 mm/rev) and high (0.405 mm/rev), were used in the study. The lathe speed was kept constant at 800 rpm. During densification, the temperature of 600 °C was applied with a temperature blower. This was followed by rapid cooling in an environment of -18 °C. Surface roughness (Ra), hardness, and spring-back measurements were conducted on both densified and undensified areas of the cylindrical solid wood samples. After densification, the Ra value decreased for all specimens and densification conditions, and the radial and tangential hardness increased. The temperature applied during densification (600 °C hot air) did not have much effect on the spring-back in 33 mm diameter specimens, while the application of temperature in 21 mm diameter specimens reduced the spring-back. At low feed rates (0.05 mm/rev) in densification, lower spring-back was obtained.*

KEYWORDS: *surface densification; spinning roller; roughness; hardness; spring-back; heat treatment*

SAŽETAK • *U ovom je istraživanju drvo crnog bora (Pinus nigra Arnold) tokarenjem oblikovano u cilindrične uzorke, a potom podvrgnuto ugušćivanju površine. Postupak je proveden na tokarskom stroju uz pomoć posebno dizajniranoga i proizvedenoga rotirajućeg valjka. Istraživanje je provedeno na toplinski obrađenim (tri sata na 160 °C) i neobrađenim uzorcima (velikog promjera 33 mm i malog promjera 21 mm). U istraživanju su primijenjene dvije posmične brzine: mala (0,05 mm/okr.) i velika (0,405 mm/okr.). Brzina vrtnje tokarskog stroja održavana je konstantnom na 800 okr./min. Tijekom ugušćivanja potrebna je temperatura od 600 °C postignuta uz pomoć*

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grijalice, nakon čega je slijedilo hlađenje pri temperaturi od –18 °C. Mjerenja hrapavosti površine (Ra), tvrdoće i elastičnog povrata provedena su na ugušćenim i neugušćenim cilindričnim uzorcima drva. Nakon ugušćivanja smanjila se vrijednost Ra svih uzoraka i pri svim uvjetima ugušćivanja, a povećala se tvrdoća drva u radijalnome i tangentnom smjeru. Temperatura primijenjena tijekom ugušćivanja (600 °C, vrući zrak) nije znatnije utjecala na elastični povrat u uzoraka promjera 33 mm, dok se u uzoraka promjera 21 mm elastični povrat smanjio. Pri manjoj posmičnoj brzini (0,05 mm/okr.), postignut je manji elastični povrat.

KLJUČNE RIJEČI: *ugušćivanje površine; rotirajući valjak; hrapavost; tvrdoća; elastični povrat; toplinska obrada*

1 INTRODUCTION
1. UVOD

Low-density woods are generally inadequate in terms of hardness, durability and strength. Either high-density or densified materials can be preferred when these properties are required in wood materials (Sandberg *et al.*, 2021). The density of the wood material directly affects the mechanical properties (Blomberg and Persson, 2004). Density can be increased by applying additional treatments to low-density wood. Several environmentally friendly methods have been used to increase density.

There are densification methods where temperature, pressure, steam and one or more of these are used as pretreatment. These are called: thermo-mechanical (TM) process (Salca *et al.*, 2021; Tosun and Sofuoglu, 2021; Sofuoglu, 2022; Sofuoglu *et al.*, 2022), thermo-hygro-mechanical (THM) process (Korkut and Kocaefe, 2009; Navi and Sandberg, 2012; Senol and Budakci, 2016), Viscoelastic-Thermal Compression (VTC) and Thermo-Vibro-Mechanical (TVM) process (Şenol and Budakci, 2016; Bekhta *et al.*, 2017; Senol, 2018). In addition to these methods, there are studies on densification of cylindrical materials (Kaya and Sofuoglu, 2023a; Kaya and Sofuoglu, 2023b; Yesil *et al.*, 2023). In general, mechanical properties such as Young's modulus (*MOE*), modulus of rupture (*MOR*), hardness and surface hardness, *MOR* and Janka hardness were found to increase in densified wood (Laskowska, 2017; Pertuzzatti, 2018; Gao *et al.*, 2019; Senol and Budakci, 2019; Wehsener *et al.*, 2023). Radial and tangential hardness values in densified wood increased with the compression ratio, while scanning electron microscopy analysis revealed that the heat treatment and densification processes led to deformations in the cell walls (Budakci *et al.*, 2016). Studies have also been conducted to identify and reduce the extent of spring-back occurring during the densification process (Kariz *et al.*, 2017; Neyses *et al.*, 2020; Scharf *et al.*, 2023). Wettability analysis revealed that the surfaces densified through the TM process exhibited increased hydrophobicity (Bekhta and Krystofiak, 2016). The effects of densification on surface treatments have also been investigated. Densified samples showed a decrease in surface roughness and an increase in surface brightness (Pelit *et al.*, 2015). Surface brightness and hardness values increased with increasing densification rate (Sofuoglu, 2022). Thermo-mechanical densification significantly affected the color change of beech and oak wood (Laskowska, 2020). The finite element method is also used for densification analysis.

In recent years, heat treatment has become a preferred and widespread modification application to minimize the negative properties and enhance the positive properties of wood materials (Icel and Simsek, 2017). Heat treatment is a widely used modification method that effectively enhances the properties of wood materials (Hill, 2006). Heat treatment was defined by Boonstra (Boonstra, 2008) and Rowell *et al.* (2009) as a physical process that causes permanent changes in the chemical content of cell wall components (cellulose, hemicellulose, lignin, etc.). Heat treatment improves the performance of wood by causing a change in its structure. Heat treatment has many advantages, such as reduced equilibrium moisture content, biological resistance to fungi and insects, and improved dimensional stability due to the reduction of woodworking, increased resistance to external weather conditions, decorative color diversity and extended service life (Wikberg, 2004; Jones and Enjily, 2006). Heat treatment of wood is an effective method of improving dimensional stability and resistance to biodegradation, but at the same time, mechanical properties are reduced (Percin *et al.*, 2024). Densified wood, especially when exposed to water and moisture, tends to return to its previous size. This is due to the spring-back (Pelit, 2014). Various studies are conducted to prevent or minimize the spring-back. Spring-back has been tried to be reduced by heat treatment applied before and after densification (Skyba *et al.*, 2009; Esteves *et al.*, 2017; Fu *et al.*, 2017). Heat-treated wood is becoming increasingly popular for both interior and exterior applications (Jirouš-Rajković and Miklečić, 2019).

In cylindrical specimens, the steam and temperature applied during heat treatment and surface densification led to a reduction in brightness. The steam and temperature used in the surface densification process decreased the *L* and *b* color values while increasing the *a* value (Yeşil *et. al.*, 2023). Under all densification conditions, there was an increase in hardness and brightness values and a decrease in roughness (*Rz*) values (Kaya and Sofuoglu, 2023a). The lowest spring-back ratio was observed in larch wood species, with a value of 0.121

mm per revolution at a feed of 0.121 mm/revolution, a spindle speed of 400 rpm, and a densification depth of 1 mm (Kaya and Sofuoglu, 2023b). Turned wood materials are used in various applications, from furniture parts to tool handles, banisters and wooden toys. The smooth surfaces produced by the turning process also enhance the success of lacquering and painting processes. This enables economical production with lower material and labor costs (Gurleyen, 1998). Surface roughness is one of the valid parameters for making economic evaluations regarding wood materials (Sogutlu, 2005). When the literature is examined, it is seen that the studies on cylindrical densification of wood are limited. There is a lack of data on the improvement of surface properties of wood by cylindrical densification and additional surface modification processes. The densification process reduces the porosity of wood materials and produces smoother surfaces. Especially in low-density wood materials, the overall density is not much affected as only the surfaces are densified. In this case, the material becomes more suitable for surface treatments while retaining its lightness and other advantages. Surface densification can also improve the surface quality of high-density wood materials.

In this study, spring back and surface roughness changes were evaluated in preheated test samples and test samples to which heat treatment was applied during densification. In addition, the success of minimizing the spring back effect and reducing surface roughness in the densification process and heat treatment application of test samples with diameter differences in the same wood species was investigated. The applied densification method is practical and easy to apply since it consists of basic equipment. The method will allow the use of cylindrical wood in places where better surface quality is desired. Considering the effect of heat treatment and temperature on the densification of wood materials by various methods, the effect of radial and tangential hardness, surface roughness and spring-back on the densification of cylindrical wood materials with a spinning roller is important. This study investigated the effect of heat treatment and temperature on the surface densification of large and small-diameter cylindrical specimens of black pine wood species and evaluated the results obtained.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

Black pine (*Pinus nigra* Arnold), which grows naturally in Turkey and has a wide range of uses, was used as the test material in the study. The specimens were obtained by a random sampling method from the timber sales company in the Simav district of Kutahya, from the timber obtained from the logs grown in the Simav region. The dried samples were kept in an air-conditioned cabinet set at appropriate temperature and relative humidity levels in accordance with the ISO 13061 (ISO 13061-1, 2014) (ISO 13061-2, 2014) standard and they reached a moisture content of 12 %.

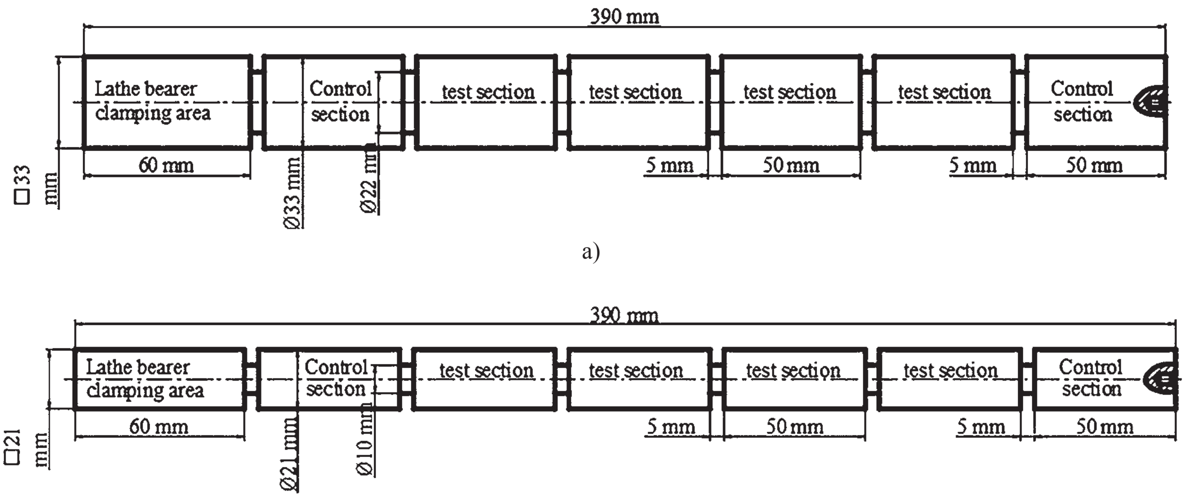
Some of the conditioned and roughly sized black pine samples were subjected to heat treatment by applying high temperatures under atmospheric pressure. The heat treatment conditions were defined as 3 hours at 160 °C, considering literature guidelines, and the application was carried out in a laboratory-type heating chamber (Nüve FN 120) controlled to an accuracy of ±1 °C. The heat-treated specimens were re-conditioned in a climatic chamber (Nüve ID 501) and brought to clear dimensions with the other specimens before turning.

Samples were cut from the lumber with a size of 33 mm × 33 mm × 390 mm and 21 mm × 21 mm × 390 mm. The remaining 60 mm test specimens on the lathe bearer clamping area were left as a square. This square section was fixed to the four-legged lathe chuck. It was fixed between the chuck and the lathe tailstock and processed until reaching the average diameters of 33 mm and 21 mm. After turning, a grooving tool was used to create 5 mm wide channels, dividing the experimental specimens into six 50 mm long cylindrical sections. Four sections were treated with experimental parameters, while the sections at the sample tip and end were retained as controls. A configuration was created on the test specimen, enabling the application of different test parameters in each section. The specimens were burnished using 400-grit sandpaper following the completion of the turning and grooving operations. The experiments were conducted three times for each parameter, distributed across different sections of various test specimens, and their averages were recorded. In total, 24 test specimens were used (Figure 1).

For the densification process, axis of the apparatus and tailstock axis were precisely lined up. Based on experience from previous studies, the surface densification depth was determined to be 1.5 mm for each diameter.

2.1 Densification equipment
2.1. Oprema za ugušćivanje

The location of the heat source, the test specimen, the densification direction and the spinning roller set in the universal lathe are shown schematically in Figure 2. The spinning roller is specially designed and manufactured for surface densification studies. Table 1 summarizes the densification parameters and their levels. Feed, one of the process parameters, is the distance in millimeters that the densification apparatus moves for each revolution of the specimen. In the study, two feed rates, one low (0.05 mm/rev) and one high (0.405 mm/rev), determined by the authors based on their experiences



a)

b)

Figure 1 Technical drawing of test specimens: a) test specimen with large diameter, b) small diameter test specimen
Slika 1. Tehnički crtež ispitnih uzoraka: a) ispitni uzorak velikog promjera, b) ispitni uzorak malog promjera

from their previous studies, were used. The authors have conducted experiments under experimental conditions close to these experimental conditions in their previous studies. Progression difference of approximately 10 times allows the change in experimental results to be clearly seen (Kaya and Sofuoğlu, 2023a; Kaya and Sofuoğlu, 2023b). The lathe speed was kept constant and selected as 800 rpm. 800 rpm is again an average spindle speed used efficiently in previous studies (Yesil *et al.*, 2023). Experiments were carried out with and without heat treatment during densification. The temperature application procedure during surface densification consists of two stages. First, while the test specimen is connected to the lathe, the temperature blower is switched on for 10 minutes to reach the set temperature (600 °C). In the second stage, the blower is fitted to the clamping apparatus prepared for it on the lathe and the experiment is completed. The temperature source was set to remain at the same distance from the test speci-

Table 1 Process parameters used in black pine turning
Tablica 1. Parametri primijenjeni pri tokarenju drva crnog bora

Parameter / *Parametar*	**Coded levels** *Oznaka*	
	Level 1	Level 2
Heat treatment / *toplinska obrada*	Yes	No
Densified diameter, mm *promjer ugušćivanja,* mm	21	33
Heat in densification *zagrijavanje tijekom ugušćivanja*	Yes	No
Feed rate, mm/rev *posmična brzina,* mm/okr.	0.05	0.405

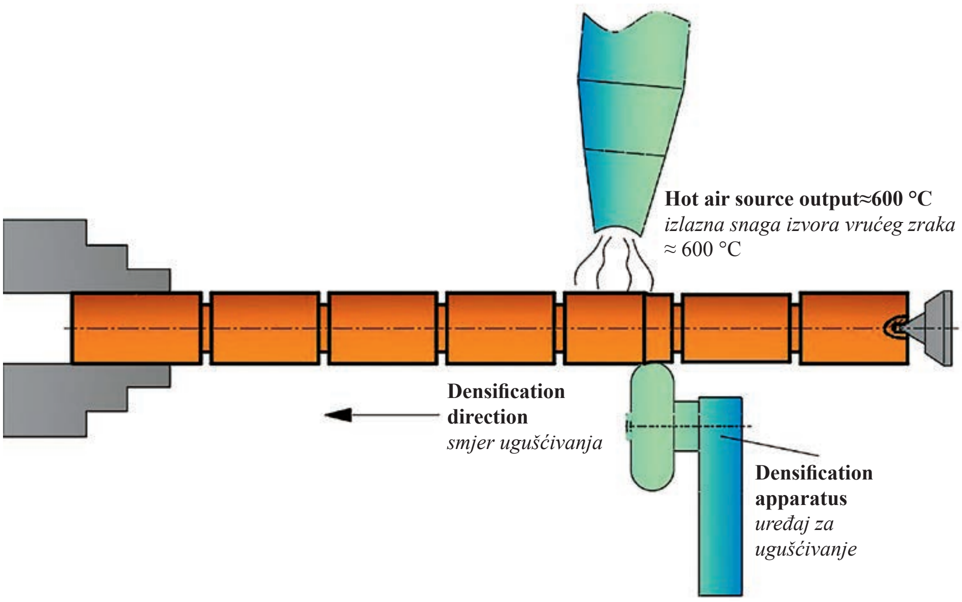


Figure 2 Densification system elements
Slika 2. Elementi sustava za ugušćivanje

a) Air conditianed cabinet
a) Komora za kondicioniranje

b) TOS-GALANTA SUIL 40A Lathe
b) Tokarski stroj TOS-GALANTA SUIL 40A

c) Surface densification
c) Ugušćivanje površine

d) Heat-Treatment
d) Toplinska obrada

e) Spinning roller
e) Rotirajući valjak

f) Cooling
f) Hlađenje

Figure 3 Steps of experimental process
Slika 3. Dijelovi eksperimentalnog procesa

mens in all experiments. Due to the positioning of the clamping apparatus on the lathe sport, the automatic feed mechanism of the lathe carriage was used to move on the part at the speeds set as the experimental parameters. The specimens to which heat was applied during the experiment were removed from the lathe as soon as the experiment was completed and suddenly exposed to a shock cooling at -18 °C. Lignin and hemicellulose, the main components of wood, are sensitive to temperature changes. Sudden cooling causes the structure of these components to solidify, which reduces the spring back. With limited resources available, experiments were only conducted at -18 °C, the temperature of a refrigerator deep freezer. More efficient results will be obtained at lower temperatures. The test samples were also grouped as preheated and unheated. Each test group included large diameter (33 mm) and a small diameter (21 mm). The steps of the test process are also given in Figure 3.

2.2 Determining surface roughness and hardness

2.2. Određivanje hrapavosti i tvrdoće površine drva

Hardness was measured with Tronic PD800 model Shore D tester. Surface roughness was measured with surface roughness tester (Time TR200, Time Group Inc., China). Measurements were made parallel to the grain in three repetitions on each sample in accordance with ISO 24118-1:2023 standard. The measurement parameter (*Ra*) was defined according to ISO 24118-1:2023. The probe speed was set to 10 mm/min and the measuring needle was 4 μm in diameter with a tip angle of 90°. Tronic brand Shore D hardness measuring device was used to measure the surface hardness of the samples. In this method, the depth of the needle inserted into the wood sample with a certain force is measured to determine the relative hardness of the wood. The spring behind the tip is stretched according to the hardness of the material and the hardness of the wood can be determined depending on the tension of the spring (Kaya and Sofuoglu, 2023b). Measurement images taken before and after the experiments are shown in Figure 4.

For hardness and roughness, measurements were taken three times, and their averages were used. The same measurements were taken on control sections of the test samples and compared with the test measurements. In the area where hardness measurements were made, attention was paid to the earlywood and latewood regions. Measurements were made on the same line on both test and control sections.

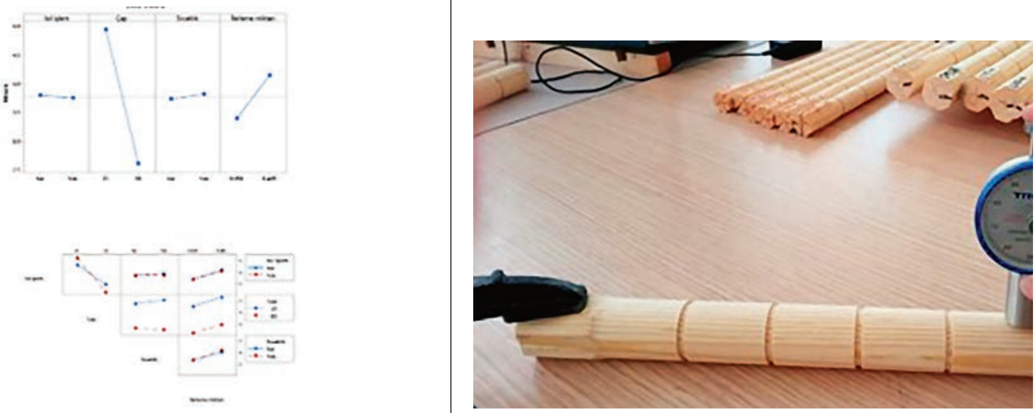
2.3 Calculation of spring-back

2.3. Izračun elastičnog povrata

In the study, instantaneous spring-back effects were evaluated depending on the densification parameters. Each diameter before the test was measured from 4 different points and their averages were taken as D_1. Following the application of the densification parameters, the same measurement procedure was conducted, and the average diameters were recorded after the process was concluded (D_2). A surface densification depth of 1.5 mm was used in the study. To obtain the theoretical diameters (D_3), the densification depth is sub-

a) Spring-back
a) Elastični povrat

b) Surface roughness (*Ra*)
b) Hrapavost površine (Ra)



d) Statistical evaluation
d) Statistička obrada

c) Surface hardness
c) Tvrdoća površine

Figure 4 Measurements as a result of experiments
Slika 4. Mjerenja provedena u istraživanju

tracted from the initial diameter. The spring-back, as shown in equation 1, is calculated by taking the percentage of the difference between D_2 and D_3.

$$\text{Spring}-\text{back}\,(\%)=\frac{(D_2-D_3)}{(D_3)}\times 100 \qquad (1)$$

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

The roughness, hardness and spring back values measured on the densified surfaces and the experimental parameters are shown in Table 2.

Table 2 Hardness, roughness and spring back values for black pine
Tablica 2. Vrijednosti tvrdoće, hrapavosti i elastičnog povrata drva crnog bora

No Br.	**Heat treatment** *Toplinska obrada*	**Diameter** *Promjer*	**Heat** *Zagrijavanje*	**Feed rate, mm/rev** *Posmična brzina,* mm/okr.	**Hardness** / *Tvrdoća*, Shore D				**Roughness** *Hrapavost*		**Spring back** *Elastični povrat*	
					Radial *Radijalni smjer*	**ST. DEV.**	**Tangent** *Tangentni smjer*	**ST. DEV.**	***Ra*, µm**	**ST. DEV.**	**%**	**ST. DEV.**
1	Yes	21	Yes	0.050	56.44	6.09	70.67	5.61	0.96	0.10	4.25	0.59
2	Yes	21	Yes	0.405	52.67	3.94	69.11	8.30	0.92	0.28	4.63	0.48
3	Yes	21	No	0.050	60.89	5.71	76.89	7.44	0.66	0.08	4.22	0.36
4	Yes	21	No	0.405	56.78	5.49	78.33	3.39	1.21	0.47	5.53	0.26
5	Yes	33	Yes	0.050	57.22	5.02	72.89	7.42	0.86	0.29	2.68	0.53
6	Yes	33	Yes	0.405	56.67	3.84	72.33	5.66	0.98	0.17	3.24	0.49
7	Yes	33	No	0.050	57.11	7.93	71.22	6.30	1.16	0.50	2.47	0.52
8	Yes	33	No	0.405	50.00	1.94	70.22	8.36	0.85	0.13	3.40	0.36
9	No	21	Yes	0.050	57.67	3.16	67.89	3.55	1.08	0.14	4.62	0.78
10	No	21	Yes	0,405	52.78	5.80	68.89	4.17	0.94	0.22	5.69	0.94
11	No	21	No	0.050	64.22	8.07	73.89	4.76	0.66	0.10	5.11	0.33
12	No	21	No	0.405	59.22	3.19	72.56	7.09	0.91	0.18	5.52	0.33
13	No	33	Yes	0.050	57.33	6.60	67.78	9.02	0.81	0.03	2.10	0.30
14	No	33	Yes	0.405	57.67	5.92	71.22	9.00	0.94	0.03	2.67	0.28
15	No	33	No	0.050	61.22	5.07	69.33	4.44	0.97	0.13	4.25	0.51
16	No	33	No	0.405	56.44	3.78	67.89	6.03	1.00	0.02	4.63	0.54

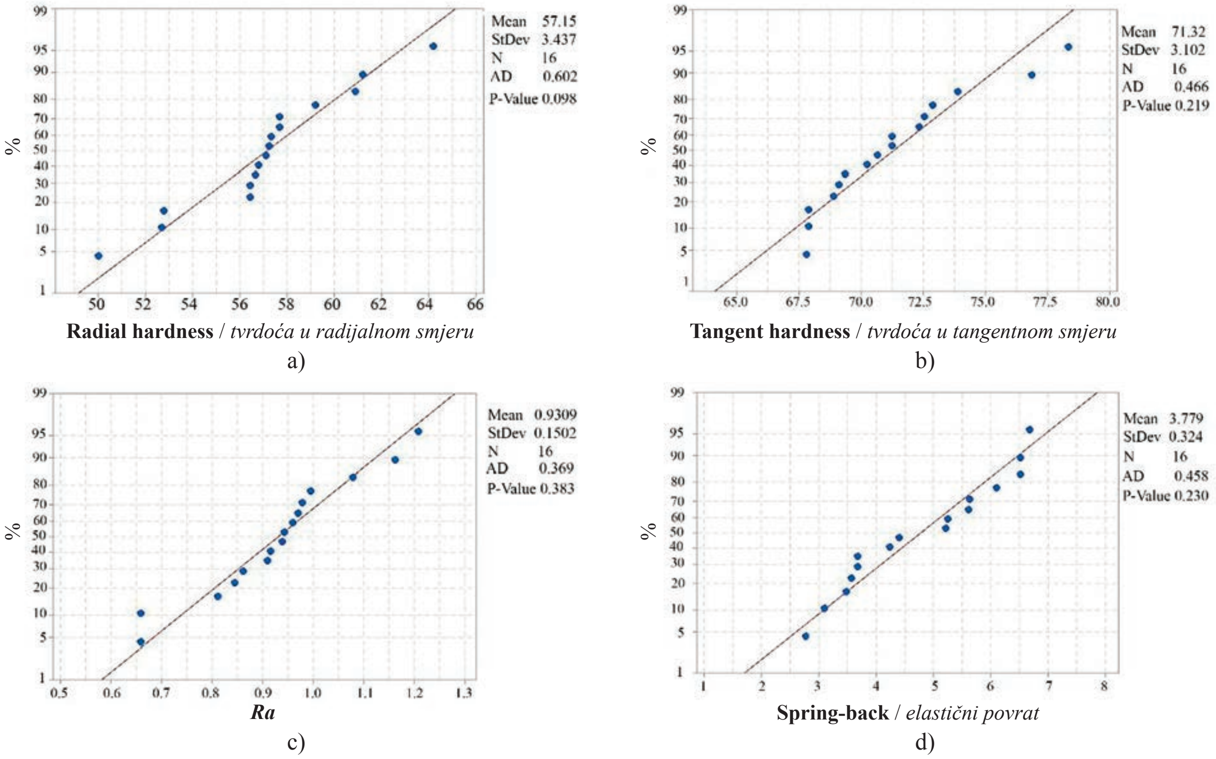


Figure 5 Normality graphs for radial hardness (a), tangent hardness (b), *Ra* (c), and spring-back (d)
Slika 5. Statistički grafovi za tvrdoću u radijalnom smjeru (a), tvrdoću u tangentnom smjeru (b), hrapavost *Ra* (c) i elastični povrat (d)

Since the *P* values is greater than 0.05 (*P* = 0.098 for radial hardness; 0.219 for tangent hardness; 0.383 for *Ra* and 0.230 for spring-back) in Figure 5, the values show a normal distribution at the 95 % confidence level.

After investigation the spring-back effect in densification processes in all conditions, it was established that the spring-back effect was lower at 0.05 mm /rev. The highest spring-back effect occurred at 0.405 mm/rev in small-diameter specimens without heat treatment and in specimens where heat was applied during densification (Figure 6).

When the control specimens are compared with the specimens after densification in various conditions, it is seen that *Ra* value decreased while radial and tangential hardness increased after densification in all conditions and in all specimens (Figure 7). When evaluating the large and small-diameter densified specimens with and without heat treatment considering the densification conditions, it can be seen that heat treatment increases the spring-back percentage and roughness, and decreases the radial and tangential hardness in large-diameter specimens in all densification conditions. In cases where heat was applied during densifi-

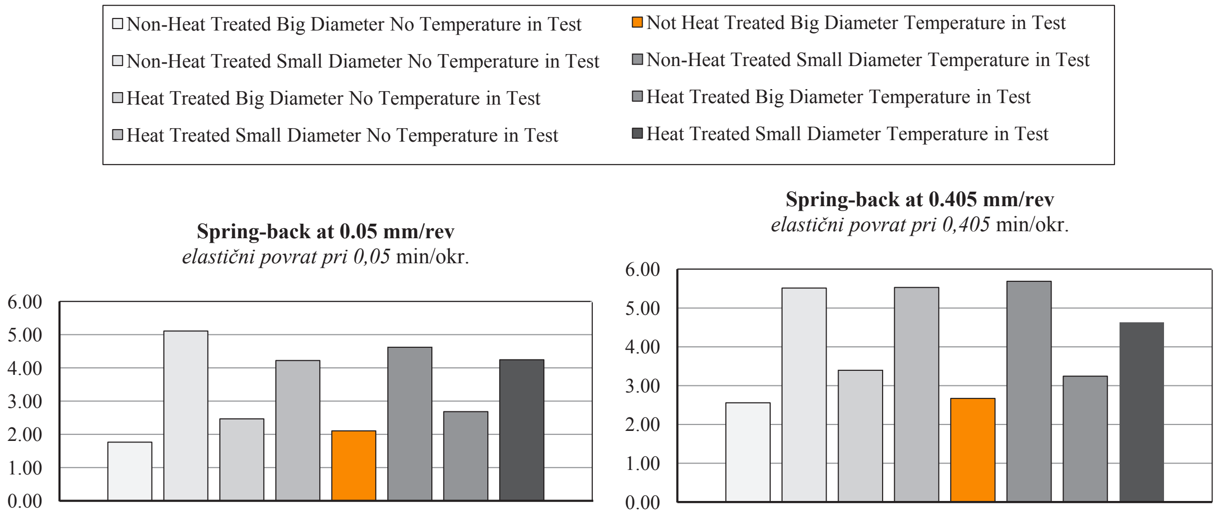


Figure 6 Effect of heat treatment and densification conditions on spring-back
Slika 6. Utjecaj toplinske obrade i ugušćivanja na elastični povrat

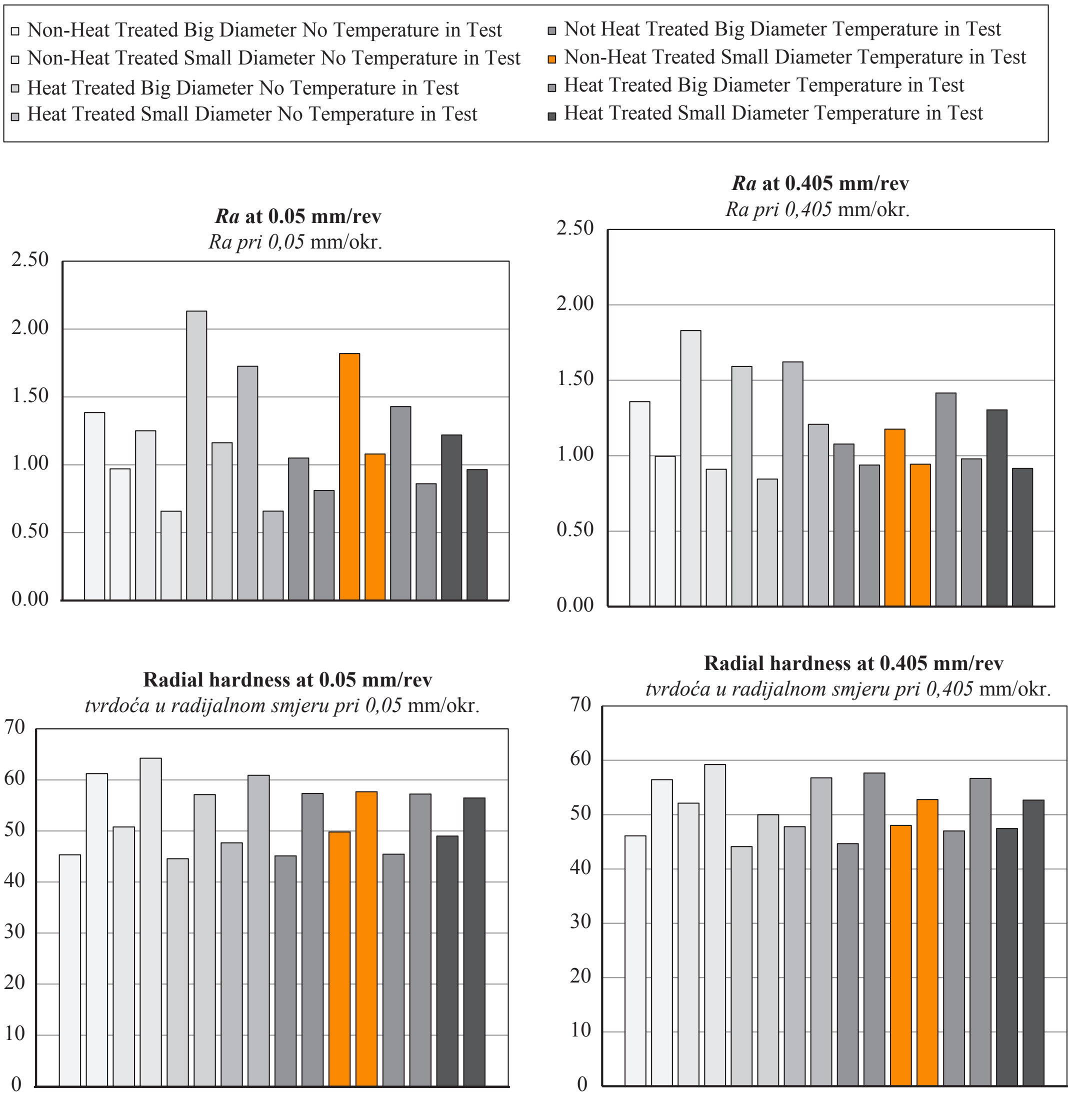


Figure 7 Comparison of differences in before and after test data of test specimens
Slika 7. Usporedba razlika u podatcima prije i nakon ispitivanja uzoraka

cation, close values were obtained in tangential hardness. When the densification process of small-diameter specimens was evaluated, it was observed that the spring-back percentage was lower in heat-treated specimens.

According to the results of variance analysis, at 95 % confidence level, the amount of progress in densification for radial hardness values (0.05 > P = 0.017) and densified diameter for percentage of spring-back (0.05 > P = 0.00) were found statistically significant. In addition, for radial hardness, tangential hardness, average surface roughness (*Ra*) and spring-back, heat treatment, diameter densified, heat application during densification and feed rate were found to be non-significant at 95 % confidence level (Table 3).

When the main effects plot of radial hardness is analyzed, it is found that the hardness is higher in conditions where no heat treatment is applied, lower diameter (21 mm), undertreatment is applied during surface densification, and the feed rate is lower (0.05) in surface densification (Figure 8). When the interaction graph (Figure 9) is evaluated in general, similar trends are generally observed in terms of tangential hardness in binary variables. However, the application of temperature during densification at different diameters (21 and 33 mm) significantly decreased the tangential hardness at low diameters, while it was slightly higher at high diameters. In unheat-treated specimens, the application of temperature decreased the radial hardness significantly, while in heat-treated specimens, although very close to each other, a very small decrease occurred. The increase in hardness is a natural result of the surface densification process. Since the densification process increases the hardness and mechanical properties of wood materials, there are many experiments and research in this field (Blomberg and Persson, 2004; Budakci *et al.*, 2022; Sofuoglu, 2022; Tosun and Sofuoglu, 2021; Sofuoglu *et al.*, 2022). This situation also shows similarities with the studies found in the literature (Rautkari *et al.*, 2009; Budakci *et al.*,

Table 3 Results of analysis of variance (ANOVA)
Tablica 3. Rezultati analize varijance (ANOVA)

Radial hardness *Tvrdoća u radijalnom smjeru*	**DF**	**Adj SS**	**Adj MS**	***F*- Value**	***P* Value**
Heat treatment / *toplinska obrada*	1	22.038	22.038	3.14	0.104
Diameter / *promjer*	1	3.063	3.063	0.44	0.522
Heat / *zagrijavanje*	1	19.019	19.019	2.71	0.128
Feed rate, mm/rev / *posmična brzina,* mm/okr.	1	55.834	55.834	7.96	0.017
Error / *pogreška*	11	77.200	7.018		
Total / *ukupno*	15	177.154			
Tangent hardness *Tvrdoća u tangentnom smjeru*	**DF**	**Adj SS**	**Adj MS**	***F*- Value**	***P* Value**
Heat treatment / *toplinska obrada*	1	30.864	30.8642	4.54	0.057
Diameter / *promjer*	1	14.694	14.6944	2.16	0.170
Heat / *zagrijavanje*	1	23.901	23.9012	3.51	0.088
Feed rate, mm/rev / *posmična brzina,* mm/okr.	1	0.000	0.0000	0.00	1.000
Error / *pogreška*	11	74.833	6.8030		
Total / *ukupno*	15	144.293			
Average surface roughness (*Ra*) *Prosječna hrapavost površine (Ra)*	**DF**	**Adj SS**	**Adj MS**	***F*- Value**	***P* Value**
Heat treatment / *toplinska obrada*	1	0.005232	0.005232	0.19	0.675
Diameter / *promjer*	1	0.003173	0.003173	0.11	0.743
Heat / *zagrijavanje*	1	0.000434	0.000434	0.02	0.903
Feed rate, mm/rev / *posmična brzina,* mm/okr.	1	0.020497	0.020497	0.73	0.412
Error / *pogreška*	11	0.309437	0.028131		
Total / *ukupno*	15	0.338774			
Spring-back / *Elastični povrat*	**DF**	**Adj SS**	**Adj MS**	***F*- Value**	***P* Value**
Heat treatment / *toplinska obrada*	1	0.0090	0.0090	0.05	0.835
Diameter / *promjer*	1	21.8089	21.8089	109.98	0.000
Heat / *zagrijavanje*	1	0.0306	0.0306	0.15	0.702
Feed rate, mm/rev / *posmična brzina,* mm/okr.	1	2.2650	2.2650	11.42	0.006
Error / *pogreška*	11	2.1812	0.1983		
Total / *ukupno*	15	26.2948			

2016; Senol and Budakci, 2016; Laskowska, 2017; Schwarzkopf, 2021; Sofuoglu, 2022; Kaya and Sofuoglu, 2023b).

Based on the analysis of tangential hardness main effects graph (Figure 10), higher tangential hardness was observed when heat treatment was applied, with lower diameter (21 mm) and no heat applied during surface densification. However, approximately the same tangential hardness value occurred at both feed rates in surface densification. When the interaction graph is evaluated in general, similar trends are observed regarding tangential hardness in binary

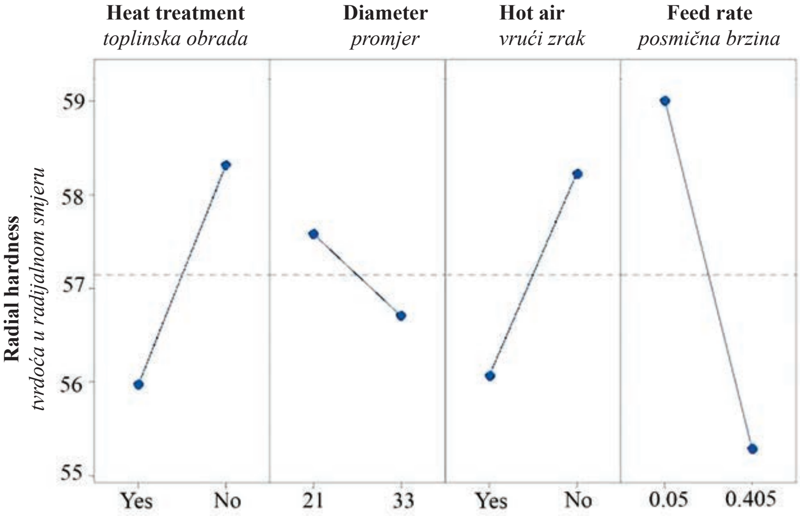


Figure 8 Main effects plot in terms of radial hardness
Slika 8. Dijagram glavnih utjecaja na tvrdoću u radijalnom smjeru

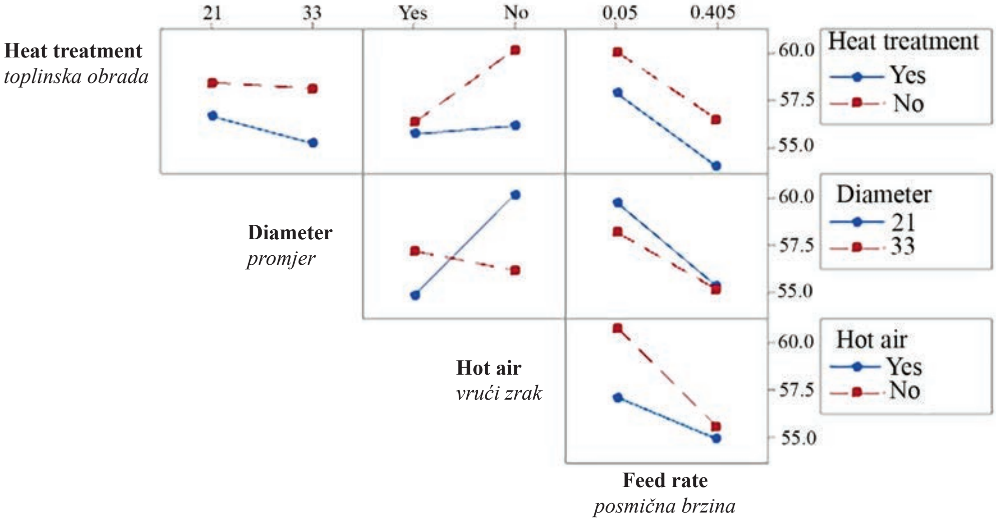


Figure 9 Interactions of heat treatment, diameter, hot air and feed rate of surface densification in terms of radial hardness
Slika 9. Djelovanje toplinske obrade, promjera, vrućeg zraka i posmične brzine na tvrdoću u radijalnom smjeru

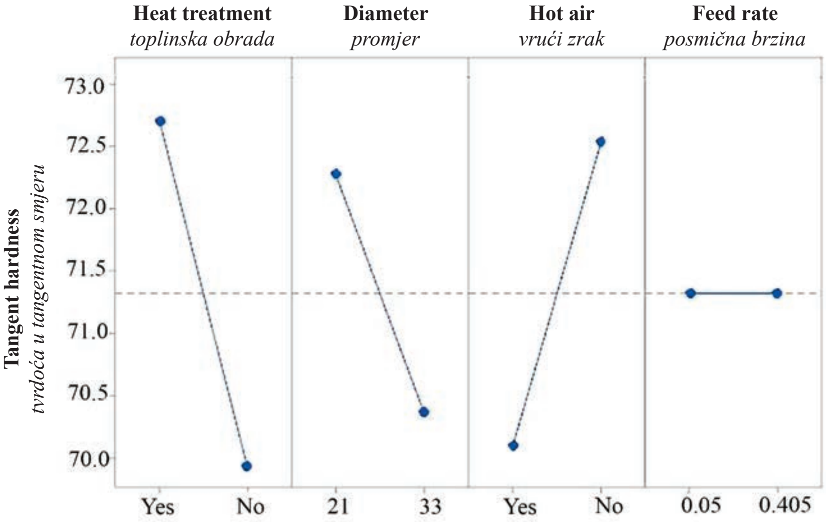


Figure 10 Main effects plot in terms of tangent hardness
Slika 10. Dijagram glavnih utjecaja na tvrdoću u tangentnom smjeru

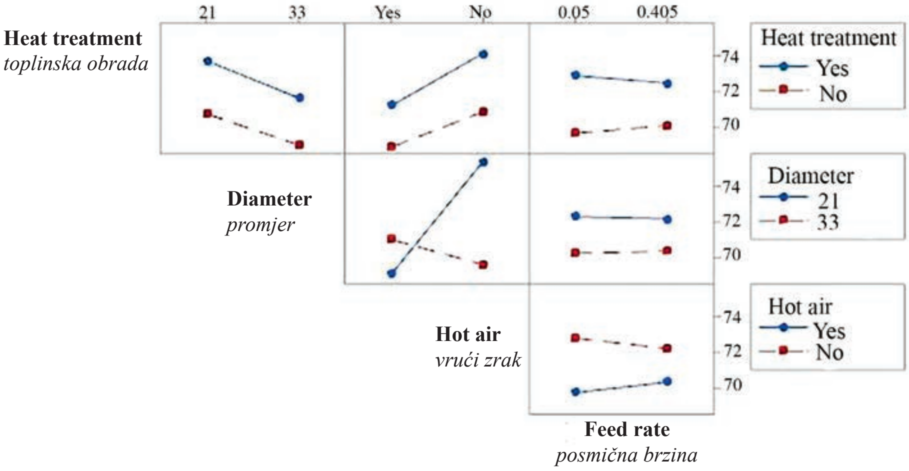


Figure 11 Interactions of heat treatment, diameter, hot air and feed rate of surface densification in terms of tangent hardness
Slika 11. Djelovanje toplinske obrade, promjera, vrućeg zraka i posmične brzine na tvrdoću u tangentnom smjeru

variables. However, applying heat during densification at different diameters (21 and 33 mm) significantly decreased the tangential hardness at low diameter, while it was slightly higher at high diameter (Figure 11). A similar result was observed for radial hardness.

When the main effects graph of roughness was analyzed, the *Ra* value was lower when no heat treatment was applied, the diameter was lower (21 mm), no heat was applied during surface densification, and the feed rate was lower (0.05) (Figure 12). When the interaction graphs for Ra were analyzed, close values were obtained for the specimens with or without heat treatment in densification conditions where heat was applied. However, applying hot air during densification decreased the roughness in the heat-treated samples. Applying hot air increases the roughness at low diameters and decreases the roughness at high diameters. *Ra* increased with the increase in the feed rate in low-diameter specimens, while *Ra* value decreased slightly in high-diameter specimens, although it did not change much. Other interactions between the variables can be seen in the plots (Figure 13).

Literature shows smoother surfaces in solid wood and wood-based materials with increased density (Malkocoglu and Ozdemir, 2006; Zhong *et al.*, 2013; Ayrilmis *et al.*, 2019; Pinkowski *et al.*, 2019; Sofuoglu and Tosun, 2023). Overall, densification is an application that allows the reduction of surface roughness (Bekhta *et al.*, 2014; Pelit and Arısut, 2022; Sofuoglu *et al.*, 2022). The data obtained in this study seem to be consistent with the literature. Uzun *et al.* (2024) reported that, due to the thermal treatment conditions, the average surface roughness (Ra) of the samples increased, and the highest Ra value was recorded in the specimens treated at 200 °C.

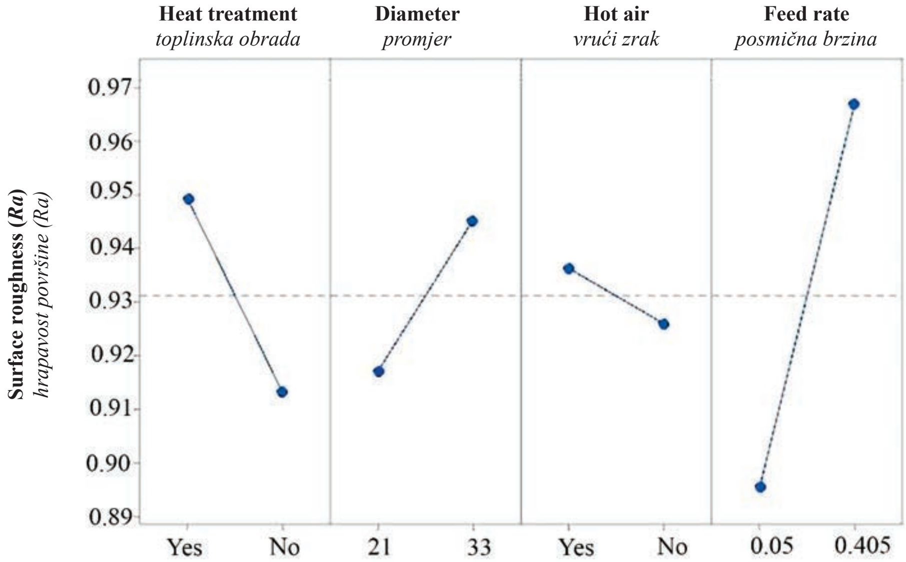


Figure 12 Main effects plot in terms of *Ra*
Slika 12. Dijagram glavnih utjecaja na hrapavost površine *Ra*

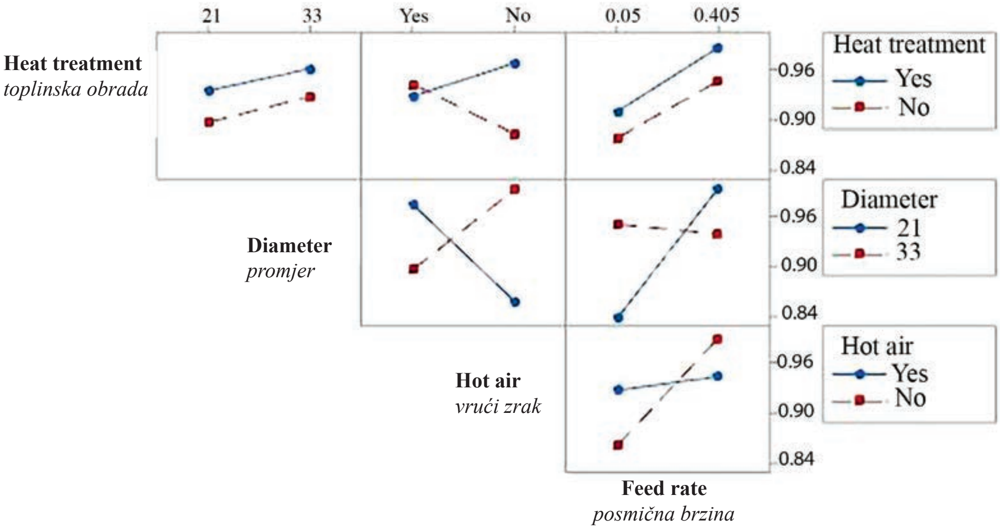


Figure 13 Interactions heat treatment, diameter, hot air and feed rate of surface densification in terms of *Ra*
Slika 13. Djelovanje toplinske obrade, promjera, vrućeg zraka i posmične brzine na hrapavost površine *Ra*

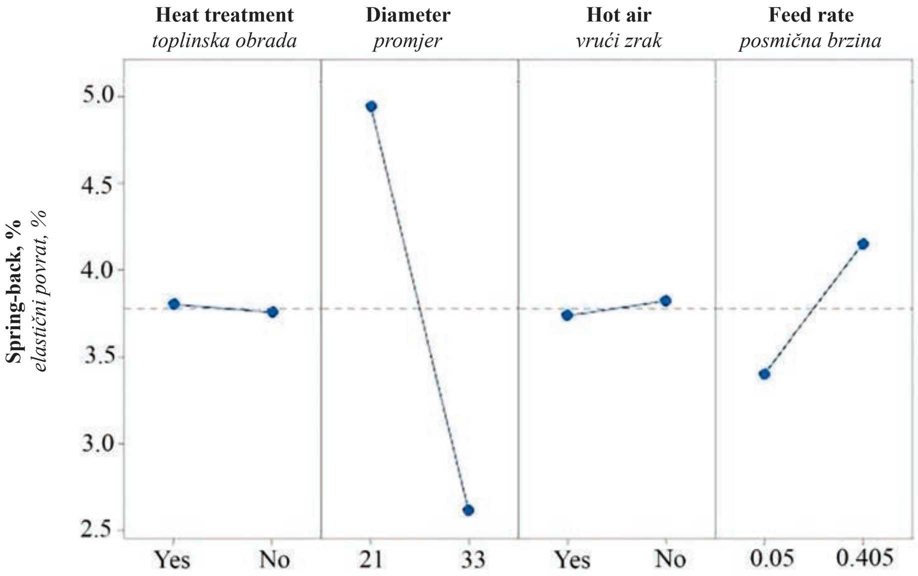


Figure 14 Main effects plot in terms of spring-back
Slika 14. Dijagram glavnih utjecaja na elastični povrat

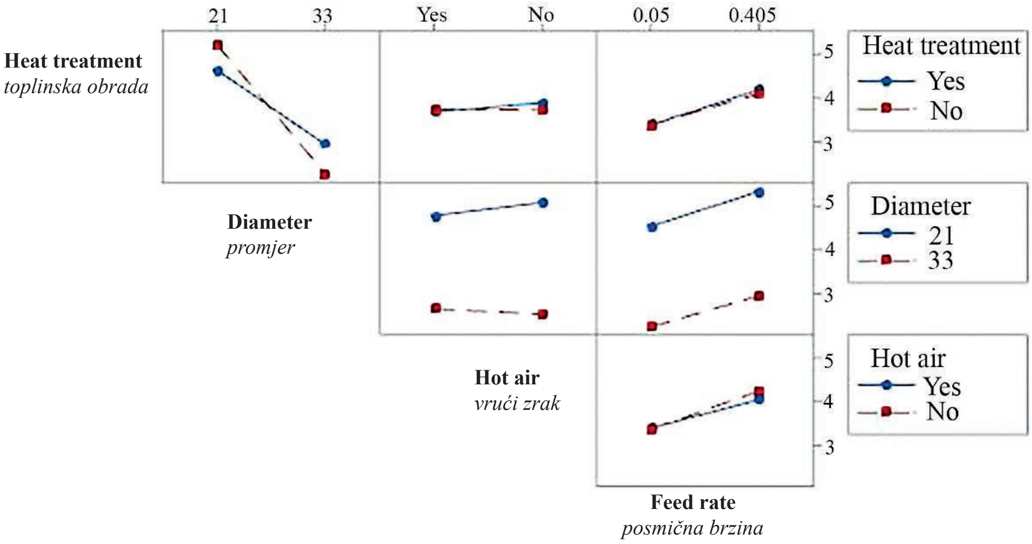


Figure 15 Interactions of heat treatment, diameter, hot air and feed rate of surface densification in terms of spring-back
Slika 15. Djelovanje toplinske obrade, promjera, vrućeg zraka i posmične brzine na elastični povrat

When the main effects graph of spring-back is analyzed (Figure 14), it is seen that the spring-back value is lower in the conditions where no heat treatment is applied, diameter is higher (33 mm), hot air is applied during surface densification, and the feed rate is lower (0.05 mm/rev).

When the interaction graphs related to spring-back are analyzed, it is seen that similar trends to the main effects graph are obtained in each variable group. While the temperature applied during densification did not have much effect on the spring-back in 33 mm diameter specimens, the application of temperature in 21 mm diameter specimens reduced the spring-back (Figure 15).

Temperature and steam are recognized methods for eliminating spring-back (Kunar and Sernek, 2007; Rautkari *et al.*, 2010; Pelit, 2014; Li *et al.*, 2017). The reduction of spring-back recovery is more effective when heat treatment is applied after densification rather than before (Esteves *et al.*, 2017). In addition to reducing spring-back, heat treatment increases the stability and durability of wood (Esteves and Pereira, 2009). The reduction in spring-back is influenced by higher densification and heat treatment temperatures, while the densification time has negligible or no effect (Li *et al.*, 2013). According to experimental studies in the literature, the spring-back effect is a phenomenon observed following densification (Pelit *et al.*, 2014; Kariz *et al.*, 2017; Tenario *et al.*, 2021). According to studies, the density increase resulting from compressing wood material is influenced by the wood species properties, the spring-back effect, and the level of compression applied during densification (Rautkari, 2012; Pelit *et al.*, 2015). Spring-back is significantly influenced by the wood type and densification depth, while feed and spindle speed have no notable effect (Kaya and Sofuoglu, 2023a).

4 CONCLUSIONS
4. ZAKLJUČAK

In this study, black pine *(Pinus nigra* Arnold.) was surface densified in its cylindrical shape. The results obtained from the study are as follows:

When the spring-back effect was examined in densification processes in all conditions, a lower spring-back effect was obtained at low revolutions (0.05 mm /rev).

The highest spring-back effect occurred at 0.405 mm/rev in small diameter samples without heat treatment and in samples where hot air was applied during production.

In both heat-treated and non-heat-treated specimens, a decrease in *Ra* value and increased radial and tangential hardness occurred after densification in all densification conditions.

The application of hot air decreased the roughness in the heat-treated specimens.

Applying hot air in densification increases the roughness at low diameters and decreases the roughness at high diameters.

When the main effects graph of spring-back was analyzed, the spring-back value was lower when no heat treatment was applied, diameter was higher (33 mm), hot air was applied during surface densification, and the feed rate was lower (0.05). The most successful densification was obtained at the lowest feed (0.05 mm/rev).

Spindle speed was not a significant factor in surface densification.

The use of spinning rollers allowed the surface densification of cylindrical wood. At the end of this densification process, it was observed that the properties of the wood material were improved. This method can be demonstrated to be effective for the surface densification of cylindrical wood materials.

The surface densification studies with a spinning roller will enable the application of optimal parameters based on the evaluation of hardness, spring back, and roughness results.

Surface densification process for cylindrical wood materials can be developed with mass production tools.

By making cost-benefit calculations, precision in production for fine workmanship can be provided with surface densification process.

Optimum production approaches will be obtained by trials with different wood species, heating and cooling processes, surface densification depths, feed and spindle speeds.

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Improving Physical and Surface Characteristics of Oriental beech Using PEG-400, Vaseline, and Epoxy Coatings

Poboljšanje fizičkih i površinskih svojstava kavkaske bukovine primjenom PEG-400, vazelina i epoksidnih premaza

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ABSTRACT • *Wood is widely used in construction and furniture industries; however, its physical and surface properties are often affected by weathering. This study evaluates the physical and surface properties of Oriental beech wood treated with some chemicals and protective coatings to improve durability. Specimens were impregnated with a 3 % aqueous solution of polyethylene glycole-400 (PEG-400) and vaseline (V), followed by an epoxy resin (EPR) coating. Physical tests such as oven-dry density, air-dry density and water absorption tests were performed. Also, some surface tests such as color and adhesion strength tests were conducted after three months of natural weathering in Muğla, Turkey. Results demonstrated that the combination of vaseline treatment and epoxy resin coating yielded the highest color stability. Chemical impregnation before epoxy coating caused significant decrease in adhesion strength of wood before weathering. While weathering caused a general decline in adhesion strength, pre-impregnation with chemicals before epoxy coating significantly reduced this decrease. Furthermore, impregnated and coated specimens exhibited higher oven-dry and air-dry densities alongside reduced water absorption compared to untreated control group, highlighting the effectiveness of these treatments.*

KEYWORDS: *impregnation; coating; color; adhesion test; oven-dry and air-dry densities; weathering*

SAŽETAK • *Drvo ima široku primjenu u graditeljstvu i industriji namještaja, no na njegova fizička i površinska svojstva često utječu okolišni uvjeti. U ovoj se studiji ocjenjuju fizička i površinska svojstva kavkaske bukovine obrađene određenim kemikalijama i zaštitnim premazima radi poboljšanja njegove trajnosti. Uzorci su impregnirani 3-postotnom vodenom otopinom polietilen glikola-400 (PEG-400) i vazelinom (V), nakon čega su premazani epoksidnim premazom (EPR). Provedena su ispitivanja fizičkih svojstava kao što su gustoća apsolutno suhog*

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drva, gustoća drva sušenog na zraku i sposobnost upijanja vode. Također, ispitane su boja i adhezivna čvrstoća uzoraka nakon tri mjeseca izlaganja prirodnim vremenskim utjecajima u Muğli, u Turskoj. Rezultati su pokazali da je kombinacijom zaštite vazelinom i epoksidnim premazom postignuta najveća stabilnost boje. Kemijska impregnacija prije nanošenja epoksidnog premaza rezultirala je znatnim smanjenjem adhezivne čvrstoće prije izlaganja vremenskim utjecajima. Izlaganje vremenskim utjecajima prouzročilo je opći pad adhezivne čvrstoće, ali znatno manji u uzoraka s kemijskom impregnacijom. Nadalje, impregnirani i premazani uzorci imali su veću gustoću drva u apsolutno suhom stanju i gustoću drva sušenog na zraku, uz smanjeno upijanje vode u usporedbi s nezaštićenim kontrolnim uzorcima, čime je potvrđena učinkovitost navedenih tretmana.

KLJUČNE RIJEČI: *impregnacija; premaz; boja; ispitivanje adhezije; gustoća drva u apsolutno suhom stanju; gustoća drva sušenog na zraku; izlaganje vremenskim utjecajima*

1 INTRODUCTION
1. UVOD

Wood has been a fundamental raw material throughout human history, valued for its versatility and utility. However, dwindling forest resources have necessitated more efficient processing techniques and strategies to extend its service life. Despite its lower density compared to materials like iron and steel, wood offers several advantages, including ease of transport, resilience under diverse loadings, low energy consumption during processing, and excellent workability (Kurtoğlu, 2000).

Surface treatments such as staining and varnishing can enhance its aesthetic appeal, and wood often becomes more visually striking with age. Epoxy-based coatings have emerged as a viable alternative for enhancing the durability of wood. Unlike biocide-based formulations, epoxy coatings form a protective physical barrier that significantly improves the wood resistance against moisture, UV radiation, and mechanical wear (Gonçalves *et al*., 2022). The hydrophobic nature of epoxy resins reduces water absorption, preventing dimensional instability and fungal decay without the need for toxic chemicals (Wang *et al*., 2021). Moreover, epoxy formulations exhibit strong adhesion properties, which enhance their long-term performance, making them suitable for both interior and exterior applications in harsh environmental conditions (Ferdosian *et al*., 2017)

Recent studies have demonstrated that epoxy coatings not only improve the aesthetic and structural properties of wood but also provide superior performance of coated wood. For instance, epoxy-modified wood has been found to exhibit enhanced mechanical strength and surface hardness, making it more resistant to physical impacts and weathering effects (Sienkiewicz *et al*., 2022). Additionally, researchers have explored hybrid epoxy formulations incorporating nanomaterials, such as silica and graphene, to further improve the thermal stability and barrier properties of epoxy-treated wood (Saba *et al*., 2016). Özgenç *et al.* (2012) highlighted the application of surface treatments such as paints, varnishes, polishes, and water repellents to safeguard wood material surfaces from UV radiation and other environmental factors. Similarly, coatings have been shown to protect fiberboard, particleboard, and solid wood from UV exposure (Burdurlu and Özgenç, 2009). However, top-surface treatments like painting and varnishing often provide only superficial protection. It is worth noting that the protective effect of surface treatments (film layer integrity) largely depends on geographical location and exposure, and in extreme cases this period can be reduced to up to 2 years (Sönmez *et al*., 2002). To ensure long-term durability, it is essential to impregnate wooden materials with water-repellent, biotic, and abiotic chemical substances before varnishing and painting (Örs *et al*., 2002).

Epoxy resins have recently gained prominence as coating materials in the wood preservation industry. These thermosetting plastics, which can transform from liquid to solid through curing agents, are widely used in applications such as insulation, coating, lamination, and marine construction, as well as in space and aviation industries (Saba *et al*., 2016). Epoxy resins offer superior adhesion, mechanical properties, and dimensional stability compared to other thermosets and thermoplastics (Mishra and Biswas, 2013; Jawaid and Abdul Khalil, 2011).

In this study, Oriental beech wood was impregnated with PEG-400, a dimensional stabilizer, and vaseline, a water repellent. The impregnated wood was subsequently coated with epoxy resin to enhance its physical and surface properties. The primary objective of this research is to improve the physical characteristics of Oriental beech wood, such as water absorption, oven dry, air-dry densities, and enhance surface qualities like color stability and strength of film on wood surface exposure to weathering conditions.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

2.1 Preparation of specimens
2.1. Priprema uzoraka

Wood specimens were prepared from the sapwood of a 40 cm diameter Oriental beech tree (*Fagus orientalis*), sourced from 150 cm above ground level.

During the preparation process, wood pieces with smooth fibers, free of knots, cracks, and insect or fungal damage, were carefully selected. The planks were sawn to a thickness of 50 mm. Specimens were air-dried and prepared for the following tests:

Color and adhesion strength tests: Specimens with dimensions of 10 mm × 100 mm × 150 mm (radial × tangential × longitudinal directions).

Oven-dry density and air-dry density tests: Specimens with dimensions of 20 mm × 20 mm × 20 mm (radial× tangential × longitudinal directions).

Water absorption (WA) tests: Specimens with dimensions of 20 mm × 20 mm × 20 mm (radial × tangential × longitudinal directions).

2.2 Impregnation procedure
2.2. Postupak impregnacije

PEG-400 was used as a dimensional stabilizer, while vaseline served as a water repellent chemical. Specimens were impregnated using a 3 % aqueous solution of PEG-400 and vaseline, respectively, following the ASTM D1413-07 (2007) standard. After 30 minutes of vacuum impregnation at 600 mm/Hg, the specimens were submerged in the solutions for 60 minutes at air pressure. The impregnation process was carried out at the oven-dry and air-dry densities for each test specimens.

Since vaseline is solid at room temperature, it was heated to 70 °C before the impregnation process. The retention quantity was calculated using Eq. 1:

$$\text{Retention} = \frac{G \cdot C}{V} \cdot 10^3 \ (\text{kg/m}^3) \tag{1}$$

Where:

$G = T_2 - T_1$

T_2 – Specimen mass following impregnation (g)

T_1 – Specimen oven dry mass before impregnation (g)

V – Specimen volume in cubic centimeters

C – Solution concentration (%)

2.3 Coating procedure
2.3. Postupak premazivanja

Following the impregnation process with PEG-400 and vaseline, the epoxy coating procedure was carried out. The epoxy resin used in this study, Sikafloor®-156, was sourced from Sika, a commercial supplier based in Istanbul. According to the manufacturer's instructions, the epoxy composition was prepared manually by mixing epoxy resin and hardener in a 3:1 ratio (epoxy: hardener by weight).

After coating, the specimens were conditioned under controlled environmental conditions of 20 °C and 65 % relative humidity for two weeks to ensure complete curing. This conditioning process was crucial for stabilizing the specimens before conducting the subsequent tests for color stability, adhesion strength, water absorption, oven-dry density, and air-dry density.

2.4 Color test
2.4. Ispitivanje boje

The CIEL*a*b* color test method was used to determine the specimen colors. In this figure, the a^* and b^* axes represent chromaticity coordinates, and the L^* axis represents lightness. Moreover, red and green are represented by the symbols $+a^*$ and $-a^*$. The constant $+b^*$ represents yellow, and the constant $-b^*$ represents blue. Zhang (2003) states that the L^* value has a range of 0 (black) to 100. In this test, the ASTM D1536-58 T (1964) standard was followed to calculate the color difference (ΔE^*) using Eqs. 2-5.

$$\Delta a^* = a_f^* - a_i^* \tag{2}$$

$$\Delta b^* = b_f^* - b_i^* \tag{3}$$

$$\Delta L^* = L_f^* - L_i^* \tag{4}$$

$$(\Delta E^*) = [(\Delta a^*)^2 + (\Delta b^*)^2 + (\Delta L^*)^2]^{1/2} \tag{5}$$

The variables Δa^*, Δb^*, and ΔL^* reflect the differences between the first and last intervals, respectively.

2.5 Adhesion test
2.5. Ispitivanje adhezije

TS EN ISO 4624 (2016) and ASTM D-4541 (2017) were used to assess the specimens adhesion strengths. Test dollies with a 20 mm diameter were glued on specimens at room temperature (20 °C ± 2 °C) in order to measure the adhesion strength. After that, the specimens were allowed to dry for a full day. The adhesion tester has a feed rate of 5 mm/minute. The adhesion strength (X) was calculated using equation 6.

$$X = \frac{4 \cdot F}{\pi \cdot d^2} \left(\frac{N}{mm^2}\right) \tag{6}$$

Where:

F – Force at break (Newton)

d – Diameter of test cylinder (mm).

2.6 Oven-dry and air-dry densities test
2.6. Ispitivanje gustoće drva u apsolutno suhom stanju i gustoće drva sušenoga na zraku

Following the TS 2472 (1976) standard, the oven-dry density (δ) measurements were carried out. In order to attain a standard mass, wood specimens were dehydrated at 103 °C ± 2 °C. The specimens were cooled before their dimensions, volumes, and masses were measured with an analytical scale that had a sensitivity of 0.01 g, stereometric measurement, and precision calliper measurement with 0.01 mm resolution.

Then, using Eq. 7,

$$\delta == \frac{M_0}{V_0} \ (\text{g/cm}^3) \tag{7}$$

Where:

M_0 – specimen's oven-dry mass (g)

V_0 – specimen's oven-dry volume (cm^3)

The air-dry density (δ) of the test specimens was determined using the TS 2472 (1976) standard. The specimens were housed in a compartment with a temperature of 20 °C ± 2 °C Band a relative humidity of 65 % ± 3 % until their mass stabilized. Following a 0.01 g analytical scale weigh-in, the specimens dimensions, air-dry density, and volumes were calculated using Eq. 8 and the stereometric method.

$$\delta = \frac{M_{12}}{V_{12}} \ (\mathrm{g/cm^3}) \qquad (8)$$

Where:
M_{12} – specimen oven-dry mass (g).
V_{12} – specimen oven-dry volume (cm^3).

2.7 Water absorption test

2.7. Ispitivanje upijanja vode

The specimens were kept for 2, 6, 14, 30, 62, 94, and 126 hours in distilled water in a room environment. Specimens were taken out of the water after each soaking interval, dried on paper, and weighed right away. Consequently, equation (9), which estimates the water absorption (WA) for each specimen, was applied.

$$WA = \frac{M_{\mathrm{f}} - M_{0\mathrm{i}}}{M_{0\mathrm{i}}} \cdot 100 \qquad (9)$$

Where:
$M_{0\mathrm{i}}$ – specimen oven-dry mass following impregnation (g)
WA – water absorption percentage (%)
M_{f} – specimen mass following water absorption (g)

2.8 Weathering test

2.8. Izlaganje vremenskim utjecajima

From January 5, 2024, until April 5, 2024, the wood of Oriental beech was subjected to weathering for three months. In accordance with ASTM D 358-55 (1970), the wood panels were ready for weathering. In the South Aegean Region, close to the Muğla Regional Meteorological Observation Station, a test site has been set up for practical assessments. Table 1 provides the Muğla meteorological statistics.

2.9 Statistical evaluation

2.9. Statistička obrada podataka

The Duncan's test and variance analysis were assessed by the SPSS computer once the test results were obtained, and they were covered with a 95 % confidence level. The homogeneity groups (HG) were the subject of statistical analyses, with different letters indicating statistical significance.

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

3.1 Color test

3.1. Ispitivanje boje

The color and total color change values of specimens impregnated with polyethylene glycole-400 (PEG-400) and vaseline (V) and coated with epoxy resin (EPR) are given in Table 2.

The color of wood is crucial in terms of aesthetic and hedonic notions for consumers. Depending on culture, location, and financial level, wood items may have a higher market value just because of their hue (Korkut *et al.,* 2013). Before weathering, the L^* values of all impregnated and coated specimens were lower than the control group. The specimens treated with PEG-400+EPR exhibited the lowest L^* values. Our findings revealed that all impregnated and coated specimens had higher a* values than the control group. Af-

Table 1 Muğla meteorological statistics
Tablica 1. Meteorološki podatci za Muğlu

Months / *Mjeseci*	**5 January to 5 February** *Od 5. siječnja do 5. veljače*	**6 February to 5 March** *Od 6. veljače do 5. ožujka*	**6 March to 5 April** *Od 6. ožujka do 5. travnja*
Maximum temperature per month, °C *najviša mjesečna temperatura,* °C	16.9	21.0	27.3
Avarage temperature per month, °C *prosječna mjesečna temperatura,* °C	6.9	8.1	10.3
Minumum temperature per month, °C *minimalna mjesečna temperatura,* °C	-2.5	-2.7	-0.9
Humidity per month, % *mjesečna vlažnost,* %	83.7	75.6	69.6
Average wind speed per month, m/s *prosječna mjesečna brzina vjetra,* m/s	1.0	0.8	0.9
Total rainfall per month, mm = kg/m^2 *ukupna mjesečna količina padalina,* mm = kg/m^2	225.2	96.9	78.5
Number of rainy days *broj kišnih dana*	16	5	10
Direct intensity of solar radiation, W/m^2 *izravni intenzitet Sunčeva zračenja,* W/m^2	989	1138	1082

Table 2 Color of impregnated and coated specimens before and after weathering
Tablica 2. Boja impregniranih i premazanih uzoraka prije i nakon izlaganja vremenskim utjecajima

Chemicals *Kemikalije*	**Retention values** *Retencija*	**Color of specimens before weathering** *Vrijednosti boje uzoraka prije izlaganja vremenskim utjecajima*			**Color of specimens after weathering** *Vrijednosti boje uzoraka nakon izlaganja vremenskim utjecajima*			**Color change values after weathering** *Vrijednosti promjene boje nakon izlaganja vremenskim utjecajima*			**Total color changes** *Ukupne promjene boje*	
		L^*	a^*	b^*	L^*	a^*	b^*	ΔL^*	Δa^*	Δb^*	ΔE^*	H.G.
Control *kontrolni uzorak*	-	66.42	10.78	20.82	61.85	7.67	23.70	-4.57	-3.11	2.88	6.23	A
EPR	-	46.33	17.70	28.10	52.59	16.22	29.05	6.26	-1.48	0.95	6.50	A
PEG-400+EPR	14.12	27.95	13.57	17.19	46.48	13.21	25.64	18.52	-0.36	8.45	20.35	B
V+EPR	12.26	47.45	14.75	23.82	52.73	13.25	26.59	5.28	-1.50	2.77	6.14	A

Note: 10 specimens were prepared for each group, EPR – epoxy resin, PEG-400 – polyethylene glycole-400, V – vaseline, H.G. – homogeneity groups.
Napomena: Za svaku je grupu pripremljeno deset uzoraka; EPR – epoksidna smola, PEG-400 – polietilen glikol-400, V – vazelin, H. G. – homogene grupe.

ter weathering, specimens treated with EPR, PEG-400+EPR, and V+EPR showed positive ΔL^* values, while the control group showed negative ΔL^* values.

Negative lightness stability values (ΔL^*) indicate that wood darkens following weathering. Darkening of the control group could be attributed to breakdown of non-cellulosic polysaccharides and lignin (Sönmez *et al.* 2011; Hon and Chang, 1985). According to Temiz *et al.* (2005), ΔL^* is a key indicator of wood surface quality. PEG-400+EPR treated specimens had the highest ΔL^* values following weathering. After weathering, red color content decreased and the yellow color content increased for all specimens.

PEG-400+EPR treated specimens had the highest total color change (ΔE^*) and the most negative results for color stability. V+EPR-treated specimens had the least total color change, making them the most stable group. PEG-400+EPR treatment resulted in significantly higher ΔE^* values compared to other groups.

3.2 Adhesion test
3.2. Ispitivanje adhezije

Table 3 lists the adhesion strength values of specimens both before and after weathering.

The results indicate that specimens treated with PEG-400+EPR and V+EPR exhibited significantly lower adhesion strength values compared to those coated solely with EPR. For instance, while the adhesion strength of the EPR-coated specimens was 4.08 N/mm^2, the values for PEG-400+EPR and V+EPR treated specimens were 1.66 N/mm^2 and 1.61 N/mm^2, respectively. This reduction in adhesion strength is consistent with previous findings. Toker *et al.* (2008) reported that the adhesion strength of Calabrian pine (*Pinus brutia* Ten.) pre-impregnated with borates prior to varnish coating decreased due to the weakening of the wood surface. Similar trends were observed in our study, where pre-impregnation before epoxy coating led to a notable decline in adhesion strength. Additionally, Üstün (2019) examined the adhesion strength of wood treated with copper-based compounds and varnished with water-based, synthetic, and polyurethane varnishes, reporting a consistent decline in adhesion strength across all test groups after natural weathering. Similarly, Baysal *et al.* (2021) found that Calabrian pine specimens treated and coated with copper-based compounds experienced a reduction in adhesion strength during weathering. Our results align with these previous studies, further supporting the impact of weathering on adhesion strength reduction.

The findings also reveal that the PEG-400+EPR and V+EPR treatments significantly mitigated the adhesion strength losses of Oriental beech specimens compared to those coated solely with EPR. Among all

Table 3 Adhesion strength values of impregnated and coated specimens before and after weathering
Tablica 3. Vrijednosti adhezijske čvrstoće impregniranih i premazanih uzoraka prije i nakon izlaganja vremenskim utjecajima

Types *Vrste*	**Retention values** *Retencija*	**Adhesion strength before weathering** *Adhezivna čvrstoća prije izlaganja*	**Adhesion strength after weathering** *Adhezivna čvrstoća nakon izlaganja*	**Adhesion strength decrease after weathering** *Smanjenje adhezivne čvrstoće nakon izlaganja*	
		Mean, N/mm^2	Mean, N/mm^2	%	H. G.
EPR	-	4.08	1.89	-60.53	B
PEG-400+EPR	14.83	1.66	1.23	-25.75	A
V+EPR	11.94	1.61	1.06	-34.16	A

Note: 10 specimens were prepared for each group, EPR – epoxy resin, PEG-400 – polyethylene glycole-400, V – vaseline, H.G. – homogeneity groups.
Napomena: Za svaku je grupu pripremljeno deset uzoraka; EPR – epoksidna smola, PEG-400 – polietilen glikol-400, V – vazelin, H. G. – homogene grupe.

Table 4 Results of oven and air-dry densities test of impregnated and coated specimens
Tablica 4. Rezultati ispitivanja gustoće impregniranih i premazanih uzoraka prije i nakon izlaganja vremenskim utjecajima

Types *Vrste*	**Retention values** *Retencija*	**Oven-dry density, g/cm^3** *Gustoća drva u apsolutno suhom stanju, g/cm^3*	**Increase compared to control, %** *Povećanje u odnosu prema kontrolnim uzorcima, %*	**Std. dev.**	**H.G.**	**Air-dry density, g/cm^3** *Gustoća drva sušenoag na zraku, g/cm^3*	**Increase compared to control, %** *Povećanje u odnosu prema kontrolnim uzorcima, %*	**Std. dev.**	**H.G.**
Control	-	0.62	-	0.04	A	0.67	-	0.04	A
EPR	-	0.70	12.90	0.09	B	0.72	7.46	0.87	B
PEG-400+EPR	14.09	0.78	25.80	0.03	B	0.80	19.40	0.02	B
V+EPR	14.47	0.74	19.35	0.02	B	0.81	20.89	0.22	B

Note: 10 specimens were prepared for each group, EPR – epoxy resin, PEG-400 – polyethylene glycole-400, V – vaseline, Std. dev. – Standard deviation, H.G. – homogeneity groups.
Napomena: Za svaku je grupu pripremljeno deset uzoraka; EPR – epoksidna smola, PEG-400 – polietilen glikol-400, V – vazelin, Std. dev. – standardna devijacija, H. G. – homogene grupe.

treatments, specimens coated only with EPR exhibited the most substantial decrease of 60.53 % in adhesion strength after weathering, whereas the PEG-400+EPR treatment proved to be the most effective, limiting the reduction to only 25.75 %. This suggests that the addition of PEG-400 and vaseline may enhance the durability of epoxy coatings under weathering conditions. However, it is important to note that the pre-impregnation process applied before epoxy coating significantly contributed to the reduction in adhesion strength, highlighting the potential limitations of this approach.

3.3 Oven-dry and air-dry densities test
3.3. Gustoća drva u apsolutno suhom stanju i gustoća drva sušenoga na zraku

Table 4 gives the oven and air-dry densities of specimens impregnated with PEG-400 and vaseline and coated with EPR.

Table 4 illustrates the results of the oven and air-dry density tests performed on EPR-coated specimens impregnated with PEG-400 and V. Density influences the mechanical and physical properties of wood and wood-derived products. In fact, density has a positive relationship with wood mechanical properties, such as hardness, abrasion resistance, and heat value (Kollmann and Cote, 1968). Our findings indicated that the control group had the lowest oven-dry and air-dry density values. The oven-dry and air-dry densities of the control group were 0.62 and 0.67 g/cm^3. According to our findings, the maximum oven-dry density value for PEG-400+EPR treated specimens was 0.78 g/cm^3, while the highest air-dry density for V+EPR treated specimens was 0.81 g/cm^3. The impregnated and coated specimens had greater oven-dry and air-dry density values compared to the control group, ranging from 12.90 % to 25.80 % and 7.46 % to 20.89 %, respectively. Statistically significant differences were seen in the oven-dry and air-dry density values of the EPR, PEG-400+EPR, and V+EPR treatment groups compared to the control group.

3.4 Water absorption test
3.4. Ispitivanje upijanja vode

The WA values of specimens impregnated with polyethylene glycole-400 (PEG-400) and vaseline (V) and coated with epoxy resin (EPR) are given in Table 5.

Wood specimens absorb more water in the early phase than in subsequent phases, according to previous studies (Alma, 1991; Hafızoğlu *et al.,* 1994; Yıldız, 1994). The entry of WA into the wood accessible empty pores at the beginning of the soaking procedure and the gradual shrinkage of those wood spaces could be the cause of these results. As an illustration, in the first two hours the WA rate of the control group was 45.45 %,

Table 5 WA values of impregnated and coated specimens
Tablica 5. WA vrijednosti impregniranih i premazanih uzoraka

Types *Vrste*	**Retention values** *Retencija*	***WA*, %**													
		2 h	H.G.	6 h	H.G.	14 h	H.G.	30 h	H.G.	62 h	H.G.	94 h	H.G.	126 h	H.G.
Control	-	45.45	C	54.27	B	62.13	B	68.72	B	74.13	B	75.52	B	78.33	B
EPR	-	27.60	B	33.39	A	41.46	A	44.22	A	48.61	A	50.40	A	51.71	A
PEG-400+EPR	16.10	18.22	A	23.52	A	35.15	A	40.50	A	45.11	A	47.35	A	49.80	A
V+EPR	12.45	25.26	AB	32.96	A	41.03	A	46.03	A	49.77	A	52.81	A	52.80	A

Note: 10 specimens were prepared for each group, EPR – epoxy resin, PEG-400 – polyethylene glycole-400, V – vaseline, H. G. – homogeneity groups.
Napomena: Za svaku je grupu pripremljeno deset uzoraka; EPR – epoksidna smola, PEG-400 – polietilen glikol-400, V – vazelin, H. G. – homogene grupe.

which is greater than half the rate of 78.33 % in the subsequent 126 hours. In comparison to the impregnated and coated specimens, the control group absorbed more water during each of the water absorption periods.

The water repelling and bulking qualities of the chemicals used contribute significantly to the reduction in WA. Untreated control specimens absorbed 78.33 % of their mass in water, whereas impregnated and coated Oriental beech absorbed 49.80-52.80 %. The hydrophobic properties of impregnated and coated specimens, which cover the wood surface and remain in the cell wall and lumen, cause reduced water penetration into wood (Chao and Lee, 2003). PEG-400+EPR treated specimens absorbed the least amount of water during all water absorption periods.

Reduced water absorption plays a crucial role in improving the resistance of wood against weathering effects. Since weathering involves prolonged exposure to moisture, temperature fluctuations, and UV radiation, limiting water absorption reduces swelling and shrinkage cycles, which can lead to cracks and mechanical deterioration. The hydrophobic properties of EPR, PEG-400+EPR, and V+EPR treatments provide a protective barrier, minimizing water uptake and thereby decreasing the risk of fungal attack, surface erosion, and degradation over time. PEG-400+EPR-treated specimens exhibited the lowest water absorption levels, indicating enhanced protection against moisture-induced deterioration, making them more resistant to environmental weathering.

In treated wood specimens, the amount of water absorbed can be limited or decreased by the movement of polyethylene glycol molecules into the swelling cell wall (Stamm, 1964). In this sense, specimens treated with PEG-400+EPR outperformed other treatments in terms of water absorption. Between the control group and the EPR, PEG-400+EPR, and V+EPR treatment groups, there were statistically significant variations in WA values at all WA intervals. Furthermore, WA values were not substantially different across the EPR-coated, PEG-400+EPR, and V+EPR treated groups.

4 CONCLUSIONS
4. ZAKLJUČAK

This study investigated some physical and surface properties of impregnated and coated Oriental beech wood, focusing on some physical tests such as oven-dry density, air-dry density, and water absorption tests, and some surface tests such as color and adhesion strength tests under weathering conditions.

Specimens impregnated with vaseline and coated with epoxy resin (EPR) exhibited the best color stability, maintaining positive lightness stability throughout the weathering period. Chemical impregnation and weathering resulted in the loss of adhesion strength. However, adhesion strength losses of impregnated and EPR-coated specimens were lower than those of specimens coated only with EPR after weathering.

Chemical impregnation and protective coatings notably increased the oven-dry and air-dry densities of the wood while substantially reducing water absorption across all testing periods. However, the combination of PEG-400 impregnation and EPR coating yielded less favorable results in terms of color stability, oven-dry density, and air-dry density. Despite these limitations, the PEG-400+EPR treatment performed well in reducing WA and preserving adhesion strength, suggesting its potential applicability for specific use cases.

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Effect of Catalysts and Density of Bamboo Wool Cement Board on Physical and Mechanical Properties

Utjecaj katalizatora i gustoće cementne ploče od bambusove vune na njezina fizička i mehanička svojstva

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ABSTRACT • *Bamboo wool cement board (BWCB) is an alternative to produce a new and sustainable material. Different types of catalysts are used for producing wood-cement composites that provide high properties. For this study, the effect of catalysts has been studied by using calcium chloride ($CaCl_2$) and calcium hydroxide ($Ca(OH)_2$). The BWCB samples from Dendrocalamus asper (Schult.F) of different density (0.60, 0.80 and 1.00 g/cm^3) were produced by using portland cement as adhesive. The physical and mechanical properties of BWCB were investigated. It was found that BWCB gained a significant percentage of dimension stability due to the treatment by $CaCl_2$ catalyst. The water absorption was reduced up to 10 %, whereas water content decreased significantly in BWCB with $Ca(OH)_2$. Flexural strength increased in BWCB with $CaCl_2$, and thermogravimetric analysis showed that thermal stability of BWCB increased with $CaCl_2$. Scanning electron micrographs of impact fracture samples are taken to study the mechanism of bamboo/cement interface adhesion due to catalyst. Thus, it can be concluded that the $CaCl_2$ might be a good catalyst for enhancing the properties of BWCB.*

KEYWORDS: *wool cement board; flexural properties; bamboo (Dendrocalamus asper); catalysts; mechanical properties*

SAŽETAK • *Cementna ploča od bambusove vune (BWCB) alternativa je za proizvodnju novoga pametnog i održivog materijala. U tom kompozitu mogu se upotrijebiti različiti katalizatori za postizanje boljih svojstava. U ovom je istraživanju proučavan učinak katalizatora kalcijeva klorida ($CaCl_2$) i kalcijeva hidroksida ($Ca(OH)_2$) na fizička i mehanička svojstva ploča. Uzorci BWCB-a različite gustoće (0,60; 0,80 i 1,00 g/cm^3) proizvedeni su od bambusa (Dendrocalamus asper (Schult. F)) i portland-cementa kao ljepila. Ispitana su fizička i mehanička svojstva uzoraka BWCB-a. Utvrđeno je da su se upotrebom $CaCl_2$ kao katalizatora povećale dimenzijska stabilnost, čvrstoća na savijanje i toplinska stabilnost BWCB-a. Nasuprot tome, primjenom ($Ca(OH)_2$) kao katalizatora upijanje vode smanjilo se do 10 %, a znatno se smanjio i sadržaj vode u BWCB-u. Lomne površine snimljene su*

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pretražnim elektronskim mikroskopom kako bi se proučio mehanizam adhezije bambusa i cementa postignut uz pomoć katalizatora. Moguće je zaključiti da $CaCl_2$ može biti dobar katalizator za poboljšanje svojstava BWCB-a.

KLJUČNE RIJEČI: *vlaknasta cementna ploča; čvrstoća na savijanje; bambus (Dendrocalamus asper); katalizatori; mehanička svojstva*

1 INTRODUCTION
1. UVOD

Wood wool cement board (WWCB) composites are formed when the wood wool is reinforced by matrix, which consists of several materials such as cement, water and chemical as catalyst (Kochova *et al*., 2017; Taiwo *et al*., 2024). The use of WWCB has been rapidly increasing in various manufacturing industries such as building and construction due to its excellent thermal as well as acoustic insulation properties (Koh *et al*., 2024). Advances have been made in the technology-based agricultural biomass in manipulating alternative materials to replace conventional materials, such as non-wood materials (Berger *et al*., 2020). Many attempts have been made to produce wool cement board from non-wood materials such as oil palm fronds (Maynet *et al*., 2021), straw (Lesieck *et al*., 2023), sisal and coconut fiber (Asefa *et al*., 2024; Kochova *et al*., 2020), kenaf core (Wang *et al*., 2021), bamboo fiber (Akinyemi and Osasona, 2017), agricultural residue (Bumanis *et al*., 2024) and other non-wood materials.

Bamboo serves as an ideal alternative material in the construction industry due to its exceptional mechanical and chemical properties. Numerous studies on bamboo as a raw material for bamboo wool cement boards (BWCB) have highlighted its resilience and ability to be harvested much faster than traditional wood. The push for sustainable BWCB production is gaining momentum due to increasing demands for improved safety and environmental sustainability. Utilizing bamboo wool as reinforcement in concrete has proven to be highly beneficial; for example, incorporating 1 % bamboo wool fiber into cement has been shown to significantly enhance the bending strength of mortar (Gelana *et al*., 2019).

However, cement boards from bamboo generally fail to achieve good mechanical properties (Maier *et al*., 2020; Chen *et al*., 2024; Taiwo *et al*., 2024). The main reason for this limitation is the hydroxyl influence and some other polar batches in the fibers (Quintana *et al*., 2024). Bamboo is also known to contain a significant amount of extractive substances, such as resins, waxes, lipids, tannins, silica, hexosan, pentosan, and starch (Wang *et al*., 2016). These extractives are thought to interact with the cement solution, forming complex compounds with the metal ions present. This interaction is believed to contribute to the poor compatibility between portland cement and bamboo wool. As a result, extractives are recognized as inhibitors in the cement hardening process (Delannoy *et al*., 2020; Nazerian *et al*., 2011). Enhancements in the mechanical properties of BWCB have been accomplished through the incorporation of catalysts (Yin *et al*., 2021).

A catalyst is an additive mixed into cement to enhance the bonding strength between the binder and wood particles, ensuring optimal adhesion. It also accelerates water evaporation from the cement board, leading to a quicker hardening process and improved final results. Calcium chloride ($CaCl_2$) is an effective catalyst for hardening cement boards made with sawdust as the primary material (Saeed, 2023). Traditionally, the curing process for cement boards takes at least 28 days to reach full strength. However, the CO_2 injection method significantly reduces the hardening time compared to conventional approaches (Taskirawati *et al*., 2019).

However, the problems in the manufacture of a cement-bonded composite requires 2-3 weeks for curing of cement. A catalyst can be used to accelerate the hardening reaction of cement with reinforcing materials. The chemical catalyst, such as calcium chloride ($CaCl_2$), magnesium chloride ($MgCl_2$) or calcium hydroxide ($Ca(OH)_2$), was reported to be successful, with the curing time shortened to a few minutes (Andales *et al*., 2019). The same authors also observed that additives have reduced the inhibitory effects of wood on the setting of portland cement. Furthermore, Ashori *et al*., (2012) investigated the properties of cement-bonded composite boards with the addition of $CaCl_2$ using portland cement as adhesive. They showed that all properties of the boards were improved when the $CaCl_2$ content was increased from 3 to 7 %.

This study aims to evaluate the impact of different types of catalyst ($CaCl_2$ and $Ca(OH)_2$) and bamboo wool board densities on the material's properties. The treatment seeks to develop lightweight yet durable bamboo-cement wool boards, offering an innovative solution to address home and building security challenges in Indonesia. The research is anticipated to produce partition wall products with a combination of lightness, strength, and compliance with standard requirements.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

2.1 Materials
2.1. Materijali

This research is focused on betung bamboo (*Dendrocalamus asper*) obtained from Sumedang, West Java, Indonesia. The study utilized about 150 cm of the

Table 1 Properties of portland cement composite (PCC)
Tablica 1. Svojstva kompozita portland-cementa (PCC)

Characteristics / *Svojstva*	**Description** / *Opis*
Air content of mortar, volume, % *sadržaj zraka u mortu, volumen, %*	4.0 – 8.0
Fineness, specific surface, m^2/kg *finoća, specifična površina*, m^2/kg	400 – 480
Autoclave / *autoklav*	
- Expansion / *širenje, %*	0.05 - 0.35
- Shrinkage / *utezanje, %*	0.01 - 0.20
Compressive strength, kg/cm^2 *Čvrstoća na tlak,* kg/cm^2	
- 3 days / *3 dana*	160 - 180
- 7 days / *7 dana*	220 - 245
- 28 days / *28 dana*	290 - 340
Setting time, vicat *Vrijeme vezanja*, vicat	
- Initial / *početno,* min	130 - 165
- Final / *završno,* min	230 - 280
False set / *lažno vezanje*	70 - 77
SO_3, %	1.5 - 2.0
Chloride / *klorid,* %	0.001 - 0.01

basal section of each bamboo stem, known for its short and unevenly spaced internodes. This segment was cut into 40 cm lengths. A 50 kg bag of Portland cement CEMI 42.5N (PCC), obtained from the local market, was used as a binding material. The standard characteristics of the PCC are presented in Table 1. Calcium chloride ($CaCl_2$) and calcium hydroxide ($Ca(OH)_2$) were supplied by Bratachem Company, Indonesia, and were used as catalysts. The catalyst was added into the PCC system in order to improve the curing process and compatibility in the bamboo wool cement board system. Physical data of catalysts are shown in Table 2.

2.2 Preparation of bamboo wool
2.2. Priprema bambusove vune

Bamboo stalks, each 40 cm in length, are divided into two sections and processed into bamboo wool using a wool-making machine (Takekawa Iron Work, China). The produced bamboo wool is initially air-dried until its moisture content reaches approximately 15 %. It is then soaked in cold water for 48 hours before being dried again to reduce the moisture content back to 15 % (Figure 1).

2.3 Hydration test
2.3. Ispitivanje hidratacije

The hydration temperature was monitored inside an insulated box. The cement mixtures had a water-cement mass ratio of 0.5, while the cement/bamboo wool mixture followed a ratio of 6.9:1.0. A thermocouple wire was placed approximately at the center of each sample and connected to a Graphtec midi LOGGER GL220 for data recording. All experiments were conducted at room temperature. The variation in the time taken to reach the hydration temperature between the cement/water mixture, and the bamboo wool with cement/water mixture indicates the effectiveness of the cement hardening inhibitor. The following equation from Hofstrand *et al*., (1984) was used:

$$I = 100 \cdot \frac{T - T_s}{T_s} \qquad (1)$$

Where: I – inhibitory hardening; T – T time of hydration of bamboo wool-cement mixture; T_s – time of cement hydration (cement-water mixture).

2.4 Preparation of bamboo wool cement board
2.4. Priprema cementne ploče od bambusove vune

Bamboo wool cement boards (BWCB) were produced with target densities of 0.60 g/cm^3, 0.80 g/cm^3, and 1.00 g/cm^3. The catalysts, $CaCl_2$ and $Ca(OH)_2$, were used at a concentration of 2 %. After being soaked in cold water for 48 hours, the bamboo wool was redried to a moisture content of around 15

Table 2 Catalyst used for the bamboo wool cement board system
Tablica 2. Katalizatori upotrijebljeni za cementnu ploču od bambusove vune

Type of catalyst *Vrsta katalizatora*	**Description** / *Opis*
Calcium chloride ($CaCl_2$) *kalcijev klorid* ($CaCl_2$)	Physical properties / *Fizička svojstva* - Appearance: White, odorless, powder in the form of granules or flakes / *izgled: bijeli prah bez mirisa, u obliku granula ili pahuljica* - Density / *gustoća*: 2.15 g/cm^3 - Melting point / *talište*: 772 °C - Boiling point / *vrelište*: 1,935 °C Chemical properties / *Kemijska svojstva* - Water-soluble: Dissolves in water to give both ions / *topljivost u vodi: topljiv u vodi uz stvaranje oba iona*
Calcium hydroxide ($Ca(OH)_2$) *kalcijev hidroksid* ($Ca(OH)_2$)	Physical properties / *Fizička svojstva* - Appearance: white-and-beige powder, odorless, crystalline in the form of hexagonal crystal structure / *izgled: bijelobež prah bez mirisa, heksagonalne kristalne strukture* - Density / *gustoća*: 2.24 g/cm^3 - Melting point / *talište*: 580 °C Chemical properties / *Kemijska svojstva*: - Water-soluble: Dissolves not very soluble in water / *topljivost u vodi: slabo topljiv u vodi*

Figure 1 Manufacturing process of bamboo wool: a) bamboo stalks, b) bamboo strips, c) wool-making machine Takekawa Iron Work, d) bamboo wool, e) bamboo wool soaked in cold water, f) bamboo wool after being soaked
Slika 1. Proces proizvodnje bambusove vune: a) stabljike bambusa; b) trake bambusa; c) stroj za izradu vune Takekawa Iron Work; d) bambusova vuna; e) bambusova vuna namočena u hladnoj vodi; f) bambusova vuna nakon namakanja

% and then weighed to meet the specified target densities. The bamboo wool was then treated with a catalyst solution, where the catalyst accounted for 2 % of the total water used per panel sheet. To evaluate the effect of the catalyst on BWCB properties, a catalyst-to-bamboo wool ratio of 4 % of the bamboo wool weight was used. The catalyst-treated wool was mixed with cement, and the specific proportions of

Table 3 Proportions of cement, bamboo wool, water and catalyst based on the target density of BWCB
Tablica 3. Udjeli cementa, bambusove vune, vode i katalizatora na temelju ciljane gustoće BWCB-a

Density, g/cm^3 *Gustoća,* g/cm^3	**Bamboo wool, g** *Bambusova vuna,* g	**Cement, g** *Cement,* g	**Water, ml** *Voda,* ml	**Catalyst, g** *Katalizator,* g
0.60	300	525	600	12
0.80	400	700	800	16
1.00	500	875	1000	20

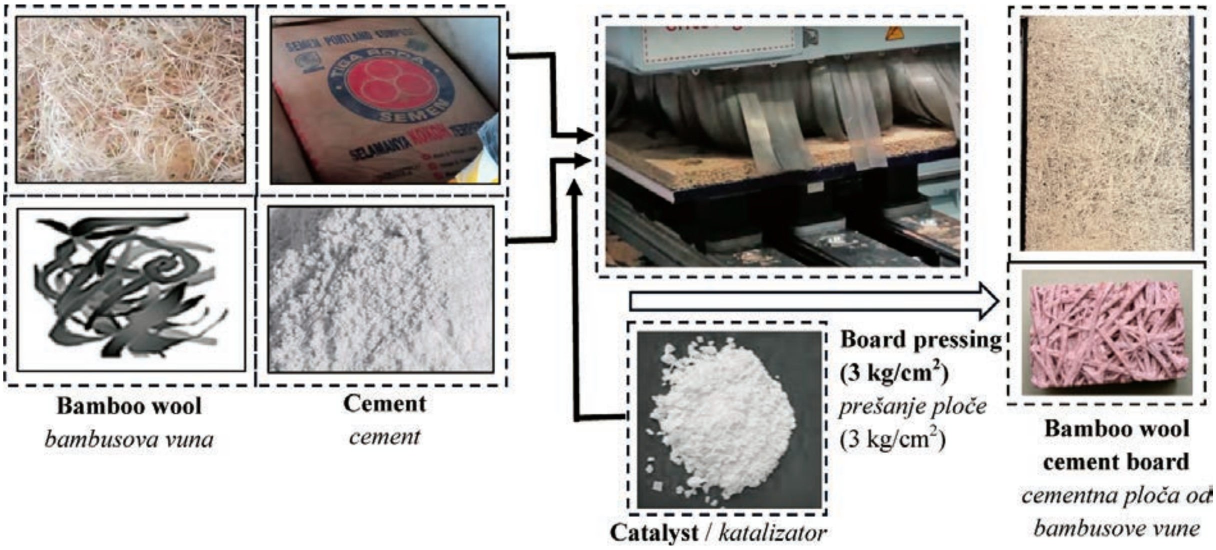

Figure 2 Manufacturing process of bamboo wool cement board (BWCB)
Slika 2. Proces proizvodnje cementne ploče od bambusove vune (BWCB)

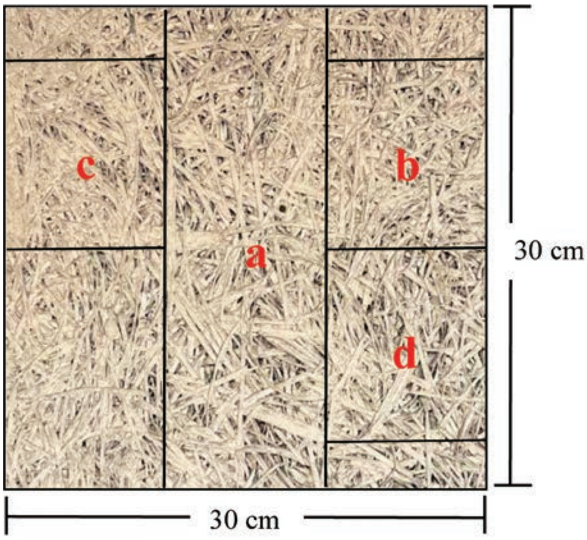

Figure 3 Process for dividing test samples: a) flexural strength test (30 cm × 10 cm × 1.5 cm); b) thickness swelling test (10 cm × 10 cm × 1.5 cm); c) water absorption and thickness shrinkage test (10 cm × 10 cm × 1.5 cm); d) density and water content test (10 cm × 15 cm × 1.5 cm)
Slika 3. Postupak izrade ispitnih uzoraka: a) ispitivanje čvrstoće na savijanje (30 cm × 10 cm × 1,5 cm); b) ispitivanje debljinskog bubrenja (10 cm × 10 cm × 1.5 cm); c) ispitivanje upijanja vode i utezanja (10 cm × 10 cm × 1.5 cm); d) ispitivanje gustoće i sadržaja vode (10 cm × 15 cm × 1.5 cm)

cement, bamboo wool and water for each target density are outlined in Table 3.

The mixture of cement, bamboo wool, water, and catalyst was formed into sheets measuring 30 cm × 30 cm × 1.5 cm, and specific pressure applied in the pressing machine was 3 kg/cm^2 and maintained for 24 h. The molded bamboo wool boards were then hydrated in a well-ventilated area (25 °C, 45-65 % RH) for 21 days to ensure better curing of the cements (Figure 2). Six bamboo wool cement boards were produced for each individual recipe.

Each BWCB sheet is trimmed into sections corresponding to the dimensions needed for the specific property tests. The preparation of the BWCB test samples adheres to the ASTM D 1037-78 standard, as shown in Figure 3.

2.5 Properties of BWCB
2.5. Svojstva BWCB-a

2.5.1 Physical properties
2.5.1. Fizička svojstva

The evaluation of BWCB physical properties is conducted in accordance with ASTM D 1037-78 standards. Thickness swelling is determined by measuring the test sample thickness at four points prior to soaking, as illustrated in Figure 4.

After measuring the thickness, the test sample was submerged in cold water for 24 hours, followed by a second thickness measurement. The thickness swelling was then calculated using the following formula:

$$\text{Thickness swelling} = [(T_2 - T_1)/T_1] \cdot 100\ \% \qquad (2)$$

Where: T_1 – initial thickness (cm) and T_2 – thickness after 24-hour soaking (cm)

The water absorption test is a continuation of the thickness swelling test, based on the weight gain after being soaked for 24 hours, which is then compared to the initial weight. The amount of water absorption is calculated using the formula:

$$\text{Water absorption} = [(B_2 - B_1)/B_1] \cdot 100\ \% \qquad (3)$$

Where: B_1 – initial weight (g) and B_2 – weight after soaking (g).

2.5.2 Mechanical properties
2.5.2. Mehanička svojstva

Mechanical properties were evaluated using a 3382 Instron UTM with a 100 kN capacity (Instron, Norwood, MA, USA). The samples, measuring 30 cm × 10 cm × 1.5 cm, underwent flexural tests at a crosshead speed of 1.15 mm/min and a span of 40 mm, in accordance with ASTM D790-02.

2.5.3 Morphological properties
2.5.3. Morfološka svojstva

The samples were examined using a SEM (Leo Supra 50 VP, Carl Zeiss, SMT, Germany) to study the microscopic structure and fracture surfaces caused by the flexural tests. Prior to analysis, small samples, 1–2 mm thick, were coated with a 10 nm gold layer by sputtering and vacuumed for 20 minutes. SEM images were taken at an accelerating voltage of 15 kV.

2.6 Data analysis
2.6. Analiza podataka

A one-way analysis of variance (ANOVA) and Duncan's multiple range test were used to analyze the

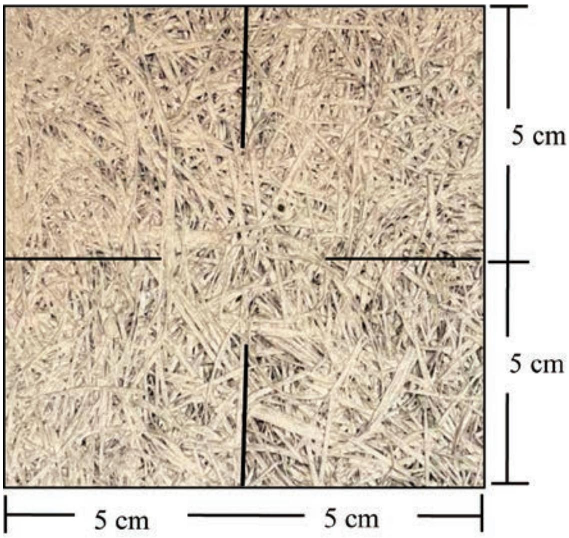

Figure 4 Procedure for measuring thickness swelling of the test sample
Slika 4. Postupak mjerenja debljinskog bubrenja

data. All statistical tests were performed using SAS at a 95 % confidence level.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

3.1 Hydration temperature
3.1. Temperatura hidratacije

The highest hydration temperature was observed at 8 h for the cement-water mixture, reaching 66 °C, and at 10 h for the cement-bamboo wool mixture, reaching 45.5 °C. According to the studies by Yin and Li (2024), the maximum hydration temperature of the cement and bamboo powder mixture is considered favorable, as it exceeds 41 °C. This can occur because bamboo betung (*Dendrocalamus asper*) has a higher hemicellulose content (27 %) (Fatriasari *et al*., 2014). High hemicellulose content can inhibit the bond between bamboo-forming material and cement (Snoeck *et al*., 2015).

Based on the percentage of the inhibitory effect of 25 % for cement hardening from the research results, it can be concluded that the treatment of soaking bamboo wool in cold water for 48 hours is quite effective in dissolving substances that inhibit cement hardening with bamboo wool. The components of bamboo extractives cause the inhibitory effect or retarding degree of cement hydration (Andreola *et al*., 2024). On the other hand, da Gloria *et al*. (2021) suggested the sugars and starches present in bamboo have been identified as the most critical compounds causing incompatibility between bamboo and cement.

3.2 Physical properties analysis
3.2. Analiza fizičkih svojstava

3.2.1 Thickness swelling properties
3.2.1. Debljinsko bubrenje

Figure 5 shows that the percentage of thickness swelling in BWCB decreases as the density increases after being soaked in cold water for 24 hours. The highest average thickness swelling, 3.81 %, occurs at a density of 0.60 g/cm³, while the lowest, 1.65 %, is observed at a density of 1.00 g/cm³. Furthermore, $Ca(OH)_2$ is a more effective catalyst than $CaCl_2$, as the boards treated with $Ca(OH)_2$ demonstrate a lower thickness swelling value (2.48 %) compared to those treated with $CaCl_2$ (2.70 %).

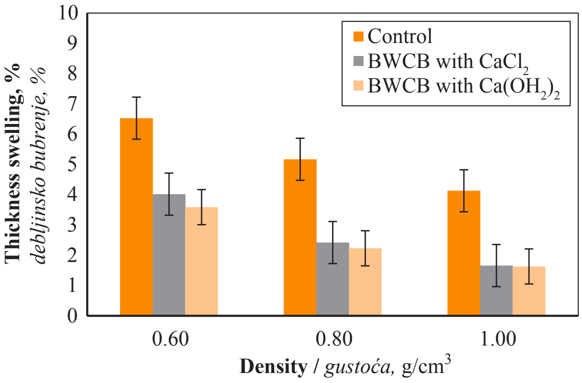

Figure 5 Thickness swelling of BWCB samples as a function of density
Slika 5. Debljinsko bubrenje uzoraka BWCB-a s obzirom na gustoću

Statistical analysis indicates that the use of Calcium Chloride ($CaCl_2$) and Calcium Hydroxide ($Ca(OH)_2$) catalysts at the same concentration (2 %) has no significant effect on the thickness expansion of BWCB. However, the board density has a highly significant impact on thickness swelling. Higher densities are associated with reduced thickness swelling, likely due to the increased cement content, which improves the boards dimensional stability. These findings are consistent with Adelusi *et al*. (2021), who reported that a higher matrix content in panel production leads to a lower percentage of thickness swelling.

Duncan's two-mean difference test revealed significant differences among treatments B1 (low density), B2 (medium density), and B3 (high density) at the 95 % confidence level. This indicates that density variations affect the thickness swelling of the panels after 24 hours of soaking in cold water. Low-density BWCBs contain more void spaces compared to high-density ones, allowing greater water absorption. Increasing the board density reduces these voids, thereby improving its thickness swelling and water absorption properties. On the other hand, higher-density BWCBs display lower thickness swelling percentages, likely due to their denser, more compact structure, which hinders water penetration during soaking (Sewar *et al*., 2024).

Furthermore, moisture exposure in natural fiber-reinforced cement board samples can lead to the leaching of water-soluble compounds and the degradation of fibers or particles into low molecular weight byproducts, which may weaken the bond between the fibers and the cement in the composite material (Choi, 2022). The absorption of water by materials is closely linked to the durability of composites, as it facilitates the introduction of harmful substances into the composite. This is considered one of the primary causes of reinforcement bar corrosion, cracking, and structural expansion. Zhao *et al*. (2019) examined the impact of incorporating pineapple leaf fibers and ramie fibers on the capillary water absorption of cement mortar. They found that as the natural fiber content increased, the water absorption of the mortar also rose, primarily because the natural fibers created pathways for water infiltration.

3.2.2 Water absorption properties
3.2.2. Upijanje vode

The data in Figure 6 indicates that the water absorption of BWCB, after 24 hours of soaking in cold

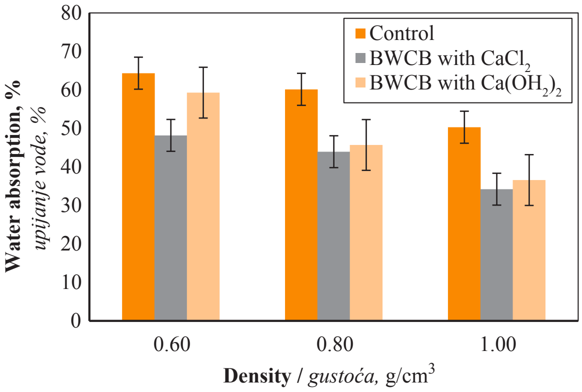

Figure 6 Water absorption of BWCB samples as a function of density
Slika 6. Upijanje vode BWCB uzoraka s obzirom na gustoću

water, decreases as panel density increases, regardless of the catalyst type used. Panels with a density of 0.60 g/cm³ exhibit the highest average water absorption at 51.25 %, while those with a density of 1.00 g/cm³ show the lowest at 35.40 %. The density of bamboo cement boards has a direct effect on their water absorption properties (de Araújo *et al.*, 2011). Lower-density boards have more interconnected pores or air pockets due to lower compaction during production. These voids act as capillaries, allowing water to rapidly enter and spread within the board (Awoyera *et al.*, 2023). Similarly, Zhao *et al.*, (2019) studied cement-based composites reinforced with natural plant fibers, specifically pineapple leaf fiber and ramie fiber. Their findings revealed that the tested composites exhibited significantly higher capillary absorption and chloride diffusion coefficients compared to plain composites, with the differences becoming more pronounced as the fiber volume fraction increased.

Furthermore, BWCB produced with a $CaCl_2$ catalyst have a lower water absorption percentage compared to those made with a $Ca(OH)_2$ catalyst. $CaCl_2$ catalyst speeded up the hydration reaction of cement. Faster hydration can lead to denser microstructure in the board, meaning fewer pores and capillaries are left open (Yin and Li, 2022). As a result, water absorption is reduced because denser structures are less permeable. The catalyst can also enhance bonding between the wool bamboo and the cement matrix, thus creating a tighter interface and reducing voids to prevent water penetration (Figure 7).

Statistical analysis reveals that using $CaCl_2$ and $Ca(OH)_2$ catalysts, both at a concentration of 2 %, has a highly significant impact on the water absorption capacity of BWCB. $CaCl_2$ proves to be more effective, resulting in a lower water absorption rate of 42.11 % compared to 45.53 % for boards using $Ca(OH)_2$. This difference is likely due to the faster hardening reaction facilitated by $CaCl_2$ in the cement-bamboo mixture, which reduces water absorption since hardened cement does not retain water (Selvan, 2021). Conversely, boards using $Ca(OH)_2$ may harden more slowly, retaining more water during the same period.

The results of the statistical analysis showed that treatment A1 ($CaCl_2$) was significantly different from A2 ($Ca(OH)_2$) at the 5 % test level. In accordance with the results of studies by Taskirawati *et al.* (2019), $CaC1_2$ as a catalyst gave better results compared to $Ca(OH)_2$ although the difference was not that great. The density of BWCB significantly influences its water absorption capacity, with higher density boards exhibiting lower water absorption. This is likely because high-density boards have greater wool coverage by cement, resulting in fewer pores. Consequently, less water is absorbed by the wool, as hardened cement does not retain water. Water absorption in panels is affected by the surface area not covered by adhesive and the volume of voids between particles (Mayer *et al*., 2022).

Duncan's two-mean difference test indicated significant differences among treatments B1 (low density), B2 (medium density), and B3 (high density) at the 5 % significance level. As panel density increases, the percentage of water absorption decreases, aligning with the observed trend in panel thickness expansion after 24 hours of immersion in cold water.

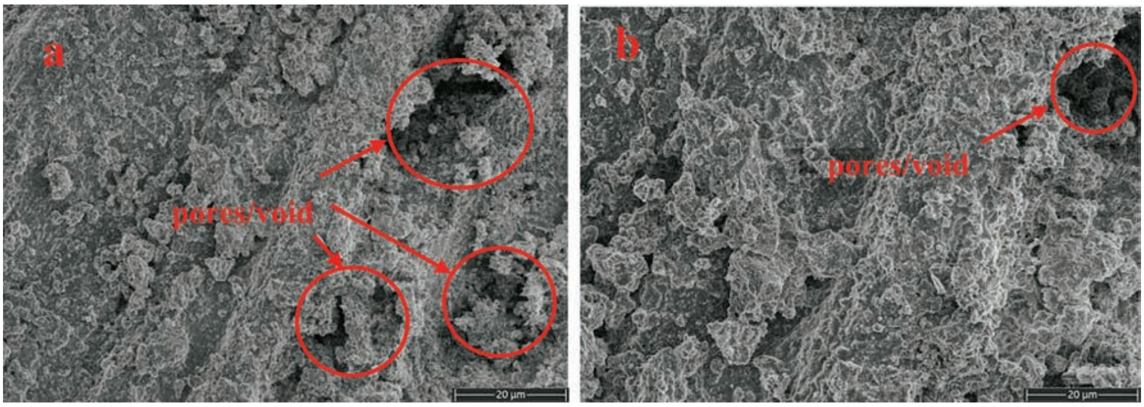

Figure 7 SEM microscopy of BWCB: a) with $Ca(OH_2)_2$; b) with $CaCl_2$
Slika 7. SEM mikrografija: a) BWCB-a s $Ca(OH_2)_2$; b) BWCB-a s $CaCl_2$

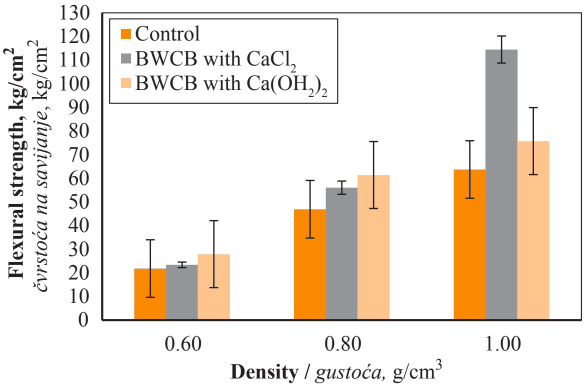

Figure 8 Flexural strength of BWCB samples as a function of density
Slika 8. Čvrstoća na savijanje BWCB uzoraka s obzirom na gustoću

3.3 Analysis of mechanical properties
3.3. Analiza mehaničkih svojstava

Figure 8 presents the flexural strength of BWCB across two catalyst types and three target density levels, demonstrating that increased density leads to higher flexural strength. Panels with a density of 1.00 g/cm³ achieve the highest average flexural strength of 95.11 kg/cm². Moreover, the CaCl$_2$ catalyst proves to be more effective in enhancing flexural strength compared to the Ca(OH)$_2$ catalyst.

In general, the internal catalyst effect of wool bamboo increases the compactness of the cement matrix, which improves the flexural strength of the samples. The results confirmed that the catalyst with cement is very effective for stabilizing the bamboo wool in matrix. The studies by Suresh and Murugaiyan (2021) suggested using calcium chloride to improve the natural fiber stabilization of matrix cement. Meanwhile, the bridging effect of wool bamboo cannot be ignored, as it can hinder the propagation of cracks in the matrix and increase the fracture energy.

Statistical analysis indicates that the use of CaCl$_2$ and Ca(OH)$_2$ catalysts significantly impacts the flexural strength of BWCB, as F-count exceeds F-table. The average flexural strength of boards made with the CaCl$_2$ catalyst is 64.62 kg/cm², compared to 55.02 kg/cm² for those using the Ca(OH)$_2$ catalyst.

The results of Duncan's Two-Mean Difference Test showed that treatment A$_1$, (CaC1$_2$) was significantly different from treatment A$_2$ (Ca(OH)$_2$) at the 95 % test level. This means that the use of different catalysts affects the value of the BWCB flexural strength, where the flexural strength of the bamboo wool board using the Ca(OH)$_2$ catalyst is lower than that of the CaCl$_2$ catalyst. From the viewpoint of the average density value of the BWCB produced as a result of this type of catalyst treatment, the smallest value was obtained with the Ca(OH)$_2$ catalyst, namely 0.77 g/cm³, while the value obtained with the CaC1$_2$ catalyst was 0.79 g/cm³. Accordingly, it can be assumed that a better flexural strength value is found in BWCBs that have a higher density.

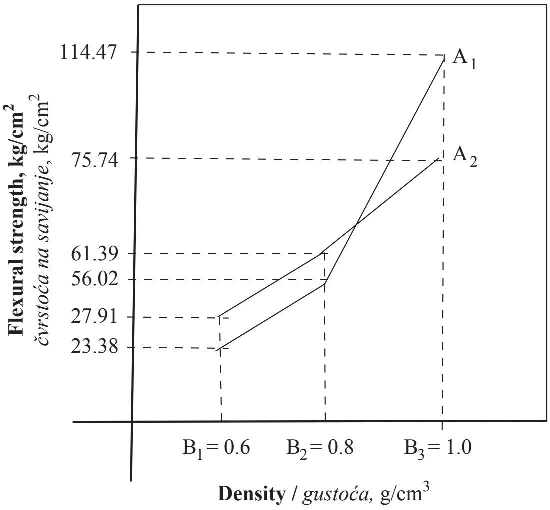

Figure 9 Interaction graph between factors A and B
Slika 9. Graf interakcije između faktora A i B

Meanwhile, treatments B$_1$ (low density), B$_2$ (medium density), and B$_3$ (high density) show significant differences at the 5 % test level, indicating that variations in density influence the flexural strength of bamboo wool boards. Higher density boards exhibit greater flexural strength. This aligns with Šipušić *et al.* (2022), who noted that increasing density reduces the cavities within the wool board. These cavities serve as weak points in the material under load, as they prevent effective load transfer between particles. The low (B1), medium (B2), and high (B3) densities, when combined with CaCl$_2$ (A1) and Ca(OH)$_2$ (A2) catalysts, significantly impact the flexural strength of BWCBs. This indicates that variations in density affect the flexural strength of the boards using both CaCl$_2$ and Ca(OH)$_2$ catalysts. However, only the CaCl$_2$ catalyst at a density of 1.00 g/cm³ significantly improves the flexural strength of the BWCB, while densities of 0.60 g/cm³ and 0.80 g/cm³ show no significant effect. This is likely because higher density reduces the number of cavities in the panel, leading to an increase in flexural strength. Additionally, both types of catalysts exhibit a similar effect on flexural strength at each density level, except at a density of 1.00 g/cm³. The effect of the interaction between the two types of catalysts and varying densities on flexural strength can be better understood through the graph presented in Figure 9.

Figure 9 depicts the effect of the interaction between factor A (catalyst type) and factor B (three density levels) on the flexural strength of bamboo wool board. This interaction aligns with the findings by Hirschmüller *et al.* (2021), who noted that interactions are represented by non-parallel lines in an interaction graph.

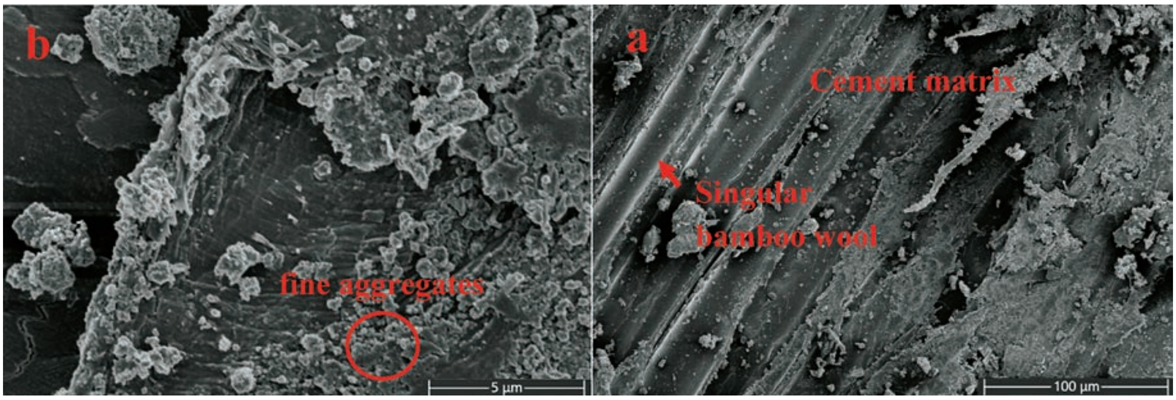

Figure 10 SEM microscopy: a) of BWCB control; b) BWCB with catalyst
Slika 10. SEM mikrografija: a) kontrolnog BWCB-a; b) BWCB-a s katalizatorom

3.4 Analysis of morphological properties
3.4. Analiza morfoloških svojstava

The SEM microscopy results for the BWCB control and BWCB with catalyst are shown in Figure 10. The SEM image in Figure 10a displays a compact and uniform bond between the cementitious matrix and bamboo wool fiber. The fiber coating of hydration products is also clearly visible, indicating strong fiber-matrix adhesion.

Figure 10b shows the characteristic interface between the cement and the bamboo wool with catalysts, which acts as an accelerator in hardening the board, so that the bond between the cement will be better if the hardening is good (Chen *et al*., 2023). Electron microscopy of bamboo wool cement containing catalyst particles reveals a uniform distribution within the cement binder. However, clumping is still present, albeit with a lower volume of agglomerates. Moreover, in the analyzed BWCB with catalyst, no significant porosity was observed in the interfacial region, only indicating the presence of catalyst near the fibers. This analysis indicates that both catalyst types exhibit excellent chemical compatibility with cement, with the $CaCl_2$ catalyst showing exceptional performance in improving BWCB properties. This conclusion is supported by SEM testing, which was used to analyze the morphology and microstructure of bamboo wool-reinforced cement composites. The SEM images revealed a homogeneous and dense cement matrix. However, the SEM images of the BWCB displayed a coarser surface, more uniform bamboo wool strands, and a smoother overall texture.

The findings suggest that both types of catalysts exhibit strong chemical compatibility with cement. Additionally, the $CaCl_2$ catalyst demonstrates outstanding performance in enhancing BWCB. The morphology and microstructure of bamboo wool-reinforced composites cement were examined and analyzed, supported by SEM test. SEM image detected that the cement matrix products were homogenous and dense. However, the SEM images of the BWCB showed the coarser surface and more equal strands of bamboo wool and a smoother surface. The results of BWCB with catalyst indicated that the capillary porosity decreased due to the cement matrix curing and increased the compatibility between the cement and bamboo wool. The longevity and weathering resistance were also enhanced (Soroushian *et al*., 2012).

These studies revealed that the mechanical properties of BWCB containing $CaCl_2$ surpass those of BWCB with $Ca(OH)_2$. Gradual water addition during stirring helps improve the dispersion of catalyst particles. He *et al.* (2019) developed cement-based photocatalyst composites by mixing the photocatalyst with cement slurry through stirring. The findings suggest that both types of catalysts exhibit strong chemical compatibility with cement. Additionally, the $CaCl_2$ catalyst demonstrates outstanding performance in enhancing BWCB.

4 CONCLUSIONS
4. ZAKLJUČAK

Density is a key factor influencing the thickness swelling of BWCB, with higher densities resulting in reduced thickness expansion. The choice of catalyst also plays a significant role in determining the properties of BWCB. Calcium hydroxide, when incorporated into the bamboo wool and cement mixture, delivers better mechanical properties compared to calcium chloride. Additionally, the interaction between BWCB density and catalyst type has a notable impact on its physical and mechanical characteristics. Catalysts exhibit strong compatibility with the cement matrix used in BWCB production. SEM morphological analysis further highlights the significant effect of catalysts on the cement structure.

The use of catalysts with cement can make crinkled cement composites flatter, resulting in the decrease of pore intensity. The results presented herein indicated a significant improvement in physical and mechanical properties.

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The EU-27 Import of Selected Wood Fuels and Current Changes in Supplying

Uvoz odabranih drvnih goriva u zemlje EU-27 i promjene u opskrbi

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ABSTRACT • *The paper presents the results of the analysis of the supply of wood pellets, wood chips, wood briquettes, sawdust and wood waste and scrap on the EU market from non-EU countries in the period 2010-2023. The aim of the study was to identify the most important non-EU countries for the supply of selected wood fuels to the EU market and the volume of imports. In addition, the value of imports of selected fuels from non-EU countries and from individual non-EU countries that are the largest suppliers to the EU market were analyzed. The study pays particular attention to the period 2022-2023, during which the EU initially reduced and then suspended imports of wood fuels from Russia and Belarus. The EU met part of its energy needs by importing wood fuels from non-EU countries during the period under review. The analyses carried out provided an insight into the EU's energy dependence on the import of selected wood fuels from non-EU countries. Regression models were used in this paper to determine the impact of EU imports of wood pellets from non-EU countries on the consumption of wood pellets in the EU.*

KEYWORDS: *wood pellets; wood chips; wood briquettes; sawdust; wood waste and scrap; energy*

SAŽETAK • *U radu su prikazani rezultati analize opskrbe tržišta EU-a drvnim peletima, drvnom sječkom, drvnim briketima, piljevinom i drvnim ostatcima iz zemalja izvan EU-a u razdoblju 2010. – 2023. Cilj studije bio je identificirati najvažnije zemlje izvan EU-a za opskrbu tržišta Unije odabranim drvnim gorivima i odrediti količinu uvoza. Osim toga, analizirana je vrijednost uvoza odabranih goriva iz zemalja nečlanica Unije te pojedinih zemalja izvan EU-a koje su najveći opskrbljivači njezina tržišta. Posebna pozornost u ovom istraživanju pridana je razdoblju 2022. – 2023., tijekom kojega je Unija isprva smanjila, a zatim obustavila uvoz drvnih goriva iz Rusije i Bjelorusije. Tijekom promatranog razdoblja Unija je dio svojih energetskih potreba zadovoljila uvozom drvnih goriva iz zemalja koje nisu njezine članice. Provedene analize dale su uvid u energetsku ovisnost Unije o uvozu odabranih drvnih goriva iz zemalja koje nisu njezine članice. U radu su primijenjeni regresijski modeli kako bi se odredio utjecaj uvoza drvnih peleta iz zemalja nečlanica EU-a na potrošnju drvnih peleta u Uniji.*

KLJUČNE RIJEČI: *drvni peleti; drvna sječka; drvni briketi; piljevina; drvni otpad i ostatak, energija*

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1 INTRODUCTION
1. UVOD

The European Renewable Energy Directive (RED I) defines the targets, rules, principles and support schemes for the use of renewable energy up to 2020 (European Commission, 2009). The target originally set in RED I that the share of renewable energy in gross final energy consumption in the EU should be 20 % by 2020 was achieved within the planned timeframe (European Commission, 2009). In RED II, a share of at least 32 % of renewable energy in gross final energy consumption in the EU by 2030 was set (European Commission, 2018).

EU targets for renewable energy consumption are always closely linked to climate challenges. In this aim, in 2020 it was proposed that the share of renewable energy in gross final energy consumption in the EU should reach 40 % by 2030 in order to achieve the planned reduction in greenhouse gas emissions (European Commission, 2020). In 2021, in the Proposal of RED II it was suggested that the share of renewable energy in gross final energy consumption in the EU should be increased to 38 % - 40 % by 2030 (European Commission, 2021). The proposed increase is justified by the achievement of the EU's climate-neutrality targets by 2050 (European Commission, 2021; Parajuli *et al*., 2024). The REPowerEU plan was adopted in 2022, which further promotes the use of renewable energy in the EU. Due to the disruption in the European energy market caused by the war between Russia and Ukraine, the REPowerEU plan was adopted with the aim of rapidly reducing the EU's dependence on Russian fossil fuels by accelerating the green transition (European Commission, 2022). In 2023, RED III prescribes a share of 42.5 % of renewable energy in gross final energy consumption in the EU by 2030, with the members of the Union achieving a share of 45 % (European Commission, 2023). Wood fuels play an important role in achieving the planned EU target for renewable energy consumption. In 2016, "more than 60 % of the biomass supplied for energy purposes in the EU was wood-based" (Camia *et al*., 2021). With 45 % of the EU covered by forests and other forest land, there is a good raw material base for the production of wood fuels (European Court of Auditors, 2021). In 2018, almost half of the wood biomass used in the EU was used for energy production (European Court of Auditors, 2021). "The use of wood for energy offers a flexible way to provide renewable energy, with additional benefits for the global climate and forests" (Brack *et al*., 2018).

Of the selected fuel categories, wood pellets and wood chips are the most traded on the European market (U.S. Department of Agriculture, 2024a). The EU has been the largest producer and consumer of wood pellets in the world for years, producing 44 % and consuming 50 % of global production (Petrović, 2014; Bioenergy Europe, 2024; U.S. Department of Agriculture, 2024a). In 2021, production reached 19.6 million tons, in 2022 it increased to 20.3 million tons, and in 2023 to 20.8 million tons (Voegele, 2022; U.S. Department of Agriculture, 2024a). Consumption of wood pellets in the EU reached 22.9 million tons in 2021 and 24.8 million tons in 2022 (U.S. Department of Agriculture, 2024a). In 2023, wood pellet consumption in the EU decreased by 1.2 % to 24.5 million tons, which was the first decline in consumption since 2015 (U.S. Department of Agriculture, 2024a). The total amount of wood chips available on the European market is not only used for energy production, but also for the production of wood-based panels as well as pulp and paper (Lamers *et al*., 2012; Jiang *et al*., 2017). Wood chips used for energy production generally have shorter transportation distances in terms of moisture content and heat output (Lamers *et al*., 2012). Wood chips and sawdust, which are by-products of wood processing, are often used for energy production in wood industry factories (Sahoo *et al*., 2022).

The study, whose results are presented in this paper, was carried out with the aim of determining the volume of imports of wood pellets, wood chips, wood briquettes, sawdust and wood waste and scrap from non-EU countries into the EU. The study also aimed to identify the most important non-EU countries for supplying the EU market with selected wood fuels. One more aim of the study was to determine the extent to which the EU meets its energy needs by importing selected fuels from non-EU countries. The study paid particular attention to the period 2022-2023, during which the EU suspended imports of selected wood fuels from Russia and Belarus. Accordingly, the resulting changes in the supply of the EU market were analyzed.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

The study, whose results are presented in this paper, was carried out according to the methodology adopted, which is based on the following. The determination of the energy demand met by the EU-27 in the period 2013-2022 through the import of wood pellets, wood briquettes and wood chips from non-EU countries is based on the values of the net calorific value of the analyzed fuels, as defined in the European standard EN ISO 17225 (2-4): "Solid biofuels - Fuel specifications and classes". In accordance with the requirements defined in the second part of the standard, the lower calorific value of this fuel of 16.5 MJ/kg was used to determine the energy obtained from the combustion of wood pellets (Petrović, 2014). The calculation also includes sawdust imported by the EU from non-EU countries during the period under consideration. It was assumed that the entire quantity of imported sawdust from non-EU was used for the production of wood pellets in the EU and based on the experience of manufacturers in Serbia, that 1.2 tons of sawdust can be used to produce one ton of wood pellets. The calculation of the energy obtained from the combustion of wood briquettes is based on the net calorific value of this fuel of 15.5 MJ/kg (EN ISO 17225, 2021).

International trade in wood briquettes and similar fuels used to be recorded under the same code in the Eurostat database until 2021, which is why it is not possible to determine the amount of energy obtained from the combustion of each type of fuel. When calculating the energy obtained from combustion, the net calorific value of the wood briquettes was used, as it is not known from which type of wood similar fuels were produced and what their moisture content was.

For the calculation of the energy obtained by burning wood chips, a moisture content of 35 % was assumed, whereby the net calorific value for coniferous wood chips at the specified moisture content is 11.63 MJ/kg and the net calorific value for hardwood at the same moisture content is 11.5 MJ/kg (EN ISO 17225, 2021). The adopted moisture content of wood chips is explained by the hygroscopic properties of wood, which dries at higher air temperatures. Trade in wood chips with a high moisture content is avoided (or limited to short distances) in order to save transportation costs.

The calculation of the energy demand met in the EU by the import of wood chips from non-EU countries was therefore carried out specifically for wood chips from softwood and hardwood, but the data is presented in a summarized form. To determine the maximum energy potential that could be achieved in the EU by burning wood chips imported from non-EU countries during the analyzed period, it was assumed that the total quantity of imported wood chips was used to generate energy.

The analysis of the EU-27 energy balances for the period 2013-2022 showed that wood pellets, wood chips and wood briquettes were used in the household, commercial and public services, industry and agriculture sectors. Energy balances from the Eurostat database were used to determine the energy demand that could be met in four sectors in the EU by importing selected wood fuels from non-EU countries (Eurostat, 2024a). The energy balance for 2023 was not yet available in the Eurostat database at the time of publication. The substitution of energy that could be obtained in the EU by burning wood fuels imported from non-EU countries, with the energy from natural gas was calculating using a net calorific value of this fossil fuel of 40 MJ/m^3 (Norwegian Petroleum, 2024).

An analysis of international trade flows of selected wood fuels was carried out on the basis of data from the Eurostat database (Eurostat, 2024b). In the Eurostat database, the data for selected wood fuels are listed in Chapter 44: Wood and articles of wood. The Eurostat database uses eight-digit codes, which are identical to customs tariffs, to record international trade in selected wood fuels (Eurostat, 2024b). For the purposes of the study, the codes for wood pellets, wood chips, wood briquettes, sawdust and wood waste and scrap were used as in Table 1 (Eurostat, 2024b).

In the period 2010-2012, trade in non-agglomerated sawdust and sawdust agglomerated into briquettes and similar forms of fuel was recorded under a com-

Table 1 Eight-digit codes for wood fuels used in Eurostat database (Eurostat, 2024b)
Tablica 1. Osmeroznamenkasti kodovi za drvna goriva korišteni u bazi podataka Eurostata (Eurostat, 2024b)

Type of wood fuel *Vrsta drvnoga goriva*	**Eight-digit codes and periods in which they were used** *Osmeroznamenkasti kodovi i razdoblja u kojima su korišteni*
Wood chips: coniferous *drvna sječka: četinjače*	44012100: „coniferous wood in chips or particles“ (2010 –)
Wood chips: broadleaves *drvna sječka: listače*	44012200: „wood in chips or particles“ (2010 – 2020) 44012210: „wood in chips or particles, of eucalyptus“ (2021 –) 44012290: „wood in chips or particles“ (2021 –)
Wood pellets *drvni peleti*	44013020: „sawdust and wood waste and scrap, agg. in pellets“ (2010 – 2011) 44013100: „wood pellets“ (2012 –)
Sawdust *piljevina*	44013930: „sawdust of wood, not agglomerated“ (2013 – 2016) 44014010: „sawdust not agglomerated“ (2017 – 2021) 44014100: „sawdust not agglomerated“ (2022 –)
Wood waste and scrap *drvni otpad i ostatak*	44013980: „wood waste and scrap, not agg. excl sawdust“ (2013 – 2016) 44014090: „wood waste and scrap, not agg, excl sawdust“ (2017 – 2021) 44014900: „wood waste and scrap, not agg, excl sawdust“ (2022 –)
Wood briquettes *drvni briketi*	44013920 „sawdust and wood waste and scrap, agg. in logs, briquetts or similar forms, excl. pellets“ (2013 – 2016) 44013900: „sawdust and wood waste and scrap agg. in logs, briquetts or similar forms, excl. pellets“ (2017 –) 44013200: „wood briquettes“ (2022 –)

mon code. Therefore, the analysis of EU imports of these fuels was performed as of 2013, when two codes were introduced to record the above-mentioned fuels. The same analysis procedure was used for wood waste and scrap. The term total imports is used in this paper to refer to the sum of EU wood fuel imports from non-EU countries and wood fuel imports between EU Member States.

Regression models were used in the paper to determine the functional dependency between EU imports of wood pellets from non-EU countries and the consumption of this fuel in the EU. Wood pellets were selected for the analysis because it is assumed that they are used entirely for energy production. The regression models were created based on data from the following sources. Data on the import of wood pellets into the EU from non-EU countries was taken from the Eurostat database, while data on the consumption of wood pellets in the EU for the period 2014-2023 was taken from the report of the U.S. Department of Agriculture (Eurostat 2024b; U.S. Department of Agriculture 2022, 2024a). To determine the impact of wood pellet imports from non-EU countries on wood pellet consumption in selected sectors in the EU, linear, power and exponential regression models were created.

The selection of the regression model that best describes the analyzed dependencies was based on the highest value of the coefficient of determination and the smallest standard error (Glavonjić *et al*., 2009). The power regression model is converted into a linear form and presented in Section 3.4. In determining the impact of imports from non-EU countries, which are the largest suppliers of wood pellets to the EU market, on the consumption of this fuel in the EU, regression models were created to examine the impact of imports from the USA, followed by Ukraine and Brazil. The parameters of three series of models (linear, power and exponential for each country) showed that the largest impact on the consumption of wood pellets in the EU comes from imports from Ukraine. The influence of imports from Ukraine on the consumption of wood pellets in the EU is best represented by a power regression model (largest coefficient of determination and smallest standard error). The power model is converted into a linear form and presented in Section 3.4. The regression models were created using the “Statistics 7.0” software package that combines correlation and regression in a single methodological tool (StatSoft Europe GmbH, Germany).

3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

This chapter presents the results of the analysis of the supply of the EU market with selected wood fuels from non-EU countries. The main non-EU countries supplying the EU market with selected fuels were identified, as well as the value of EU imports of these fuels in the period 2010-2023. Particular attention in the analysis was paid to the period 2022-2023, during which the supply on the EU market changed due to the reduction and then suspension of imports from Russia and Belarus. The percentage share of energy demand in four sectors in the EU met by imports of selected wood fuels from non-EU countries in the period 2013-2022 was determined. Regression models demonstrated the existence of a functional dependency between EU imports of wood pellets from non-EU countries and the consumption of this fuel in the EU.

3.1 Supply of selected wood fuels to the EU market

3.1. Opskrba tržišta EU-a odabranim drvnim gorivima

In the period from 2010 to 2023, non-EU countries were the primary suppliers of wood chips and wood pellets to the EU market (Figure 1). A peak in EU imports of wood chips from non-EU countries was observed in 2019, reaching 7.6 million tons, followed by a steady decline in imports, which fell to 2.6 million tons by 2023 (Eurostat, 2024). Although the import of wood fuel from Russia and Belarus was significantly reduced and ultimately halted during 2022-2023, this situation did not impede the supply of wood pellets to the EU market. In response to these changes, the EU had record imports of wood pellets from non-EU countries, totaling 5.9 million tons in 2022, with a decrease to 4.9 million tons in 2023 (Eurostat, 2024). The import of wood briquettes, sawdust, and wood waste and scrap from non-EU countries remained considerably lower than that of wood chips. A record import of wood briquettes reached 1.0 million tons in 2018, followed by sawdust imports of 0.5 million tons in 2021, and wood waste and scrap imports of 1.5 million tons in 2013 (Eurostat, 2024). Following these peak values, the EU decreased the import of all three wood fuel types from non-EU countries until 2023, with wood briquette import dropping to 0.4 million tons, sawdust to 0.13 million tons, and wood waste and scrap to 1.2 million tons (Eurostat, 2024).

3.1.1 Major non-EU countries for the supply of wood chips to the EU market

3.1.1. Glavne zemlje izvan EU-a za opskrbu tržišta Unije drvnom sječkom

During the period under review, Belarus and Russia were the largest suppliers of wood chips to the EU market, while Uruguay, Brazil and Norway were significantly less important (Figure 2). In 2019, record imports from these countries amounted to 7.3 million tons, accounting for 96.7 % of imports from non-EU

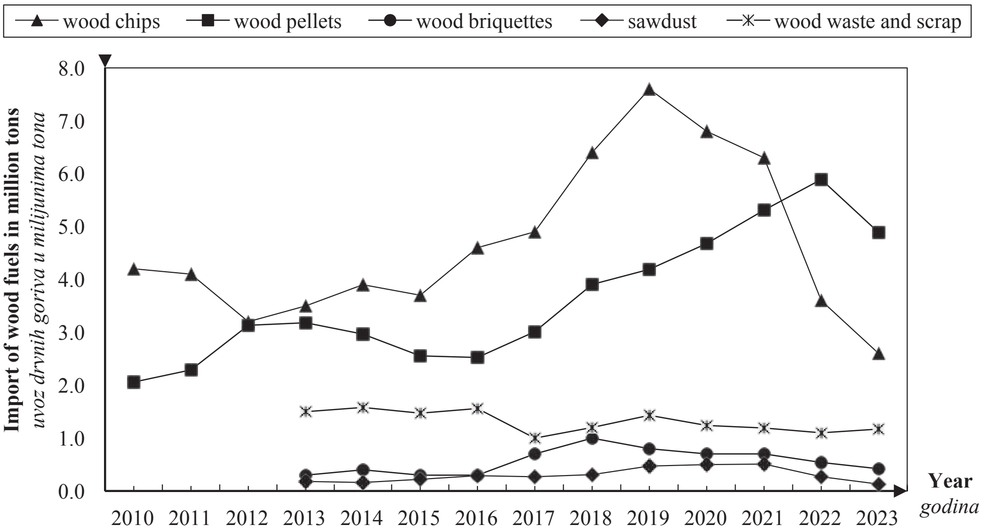

Figure 1 Market supply in the EU-27 with wood chips, wood pellets, wood briquettes, sawdust and wood waste and scrap from non-EU countries in the period 2010-2023 (Eurostat, 2024)
Slika 1. Tržišna opskrba u EU-27 drvnom sječkom, drvnim peletima, drvnim briketima, piljevinom te drvnim otpadom i ostatcima iz zemalja izvan EU-a u razdoblju 2010. – 2023. (Eurostat, 2024.)

countries (Belarus 43.9 %, Russia 28.7 %, Uruguay 12.7 %, Norway 7.3 %, Brazil 4.1 %) (Eurostat, 2024). After the EU achieved record imports of wood chips from Belarus of 3.3 million tons in 2019 and from Russia of 2.5 million tons in 2020, imports from these two countries were reduced in 2022 and stopped in 2023 (Eurostat, 2024). As a result, EU imports of wood chips from non-EU countries were 42.0 % lower in 2022 than in 2021 and 28.3 % lower in 2023 than in 2022 (Eurostat, 2024).

With the exception of the period 2010-2011, imports from Uruguay did not exceed 1.0 million tons in the other years of the period under review, nor did imports from Norway and Brazil (Eurostat, 2024). Russia mainly supplied wood chips to Finland, Belarus to Poland, Lithuania and Latvia, Brazil and Uruguay to Portugal and Norway to Sweden. In 2023, Russia was replaced by Latvia and Estonia on the Finnish market, Belarus was replaced by Lithuania on the Polish market, while Latvia became the largest supplier to Lithuania and Estonia to Latvia (Eurostat, 2024).

3.1.2 Major non-EU countries for the supply of wood pellets to the EU market

3.1.2. Glavne zemlje izvan EU-a za opskrbu tržišta Unije drvnim peletima

With the exception of 2010, when Canada was the largest supplier of wood pellets to the EU market, it was the USA and Russia in the other years of the period under review. Ukraine, Brazil, Belarus and Canada were less important for supplying the EU market in the other years of the period under review (Figure 3). In 2021, the EU imported 5.1 million tons from the coun-

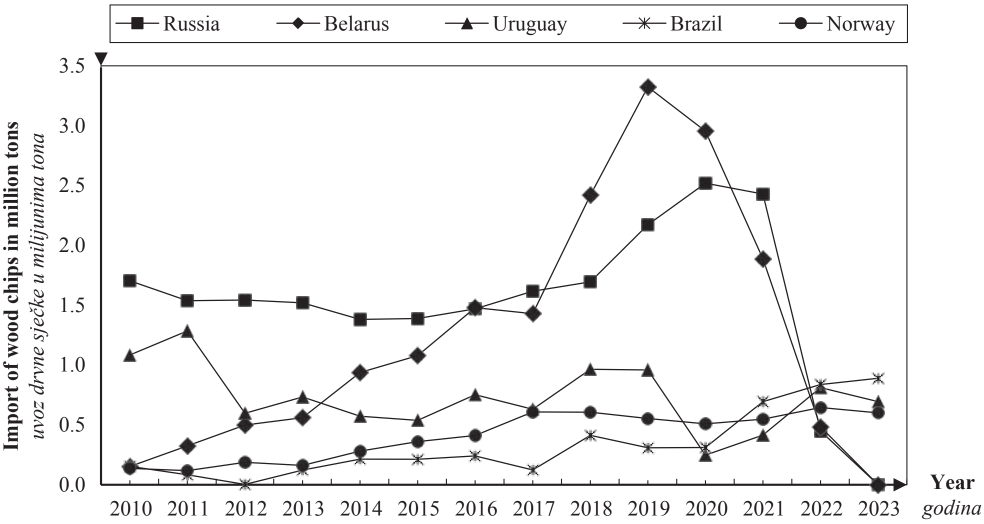

Figure 2 Major non-EU countries for the supply of wood chips to the EU market in the period 2010-2023 (Eurostat, 2024)
Slika 2. Glavne zemlje izvan EU-a za opskrbu tržišta Unije drvnom sječkom u razdoblju 2010.– 2023. (Eurostat, 2024.)

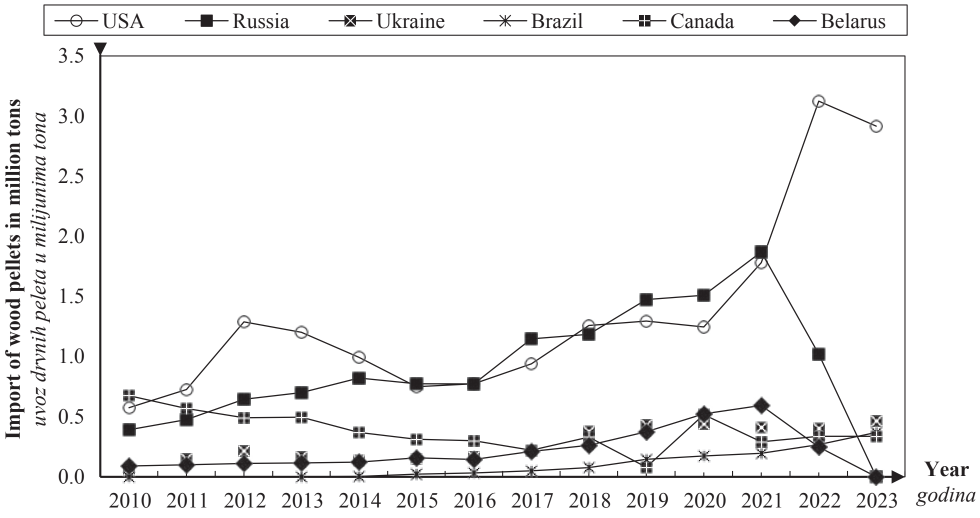

Figure 3 Major non-EU countries for the supply of wood pellets to the EU market in the period 2010-2023 (Eurostat, 2024)
Slika 3. Glavne zemlje izvan EU-a za opskrbu tržišta Unije drvnim peletima u razdoblju 2010. – 2023. (Eurostat, 2024.)

tries mentioned above, which corresponded to 96.9 % of imports from non-EU countries (Russia 35.2 %, the USA 33.5 %, Belarus 11.2 %, Ukraine 7.7 %, Canada 5.5 %, and Brazil 3.8 %). Due to the decline in imports from Russia in 2022, the USA again became the largest supplier of wood pellets to the EU market. In the same year, the EU increased its wood pellet imports from the USA by 75.4 % compared to 2021. In 2022, EU wood pellet imports from the USA, Ukraine, Brazil and Canada accounted for 70.1 % of imports from non-EU countries (USA 53.0 %, Ukraine 6.8 %, Canada 5.7 %, and Brazil 4.6 %). Although imports from the USA fell by 6.7 % in 2023 compared to 2022, the share of the four largest suppliers in EU imports from non-EU countries rose to 83.2 % (USA 59.4 %, Ukraine 9.5 %, Brazil 7.5 %, and Canada 6.8 %). By 2021, Russia and the USA supplied most wood pellets to the Netherlands, Belgium and Denmark, Brazil to Italy, Canada to the Netherlands, Belarus to Latvia and Lithuania and Ukraine to Poland and Slovenia. In 2023, the USA, Brazil and Ukraine retained their markets, and Canada exported to Italy, Denmark and France. The new situation forced some members of the Union to work with new suppliers, with the result that Lithuania imported most of its pellets from Vietnam in 2023.

3.1.3 Major non-EU countries for the supply of wood briquettes to the EU market

3.1.3. Glavne zemlje izvan EU-a za opskrbu tržišta Unije drvnim briketima

During the analyzed period, Ukraine and Russia were the largest suppliers of wood briquettes to the EU market, while the importance of Bosnia and Herzegovina, Belarus and Norway were significantly lower (Figure 4). In 2018, the EU imported a record 792,000

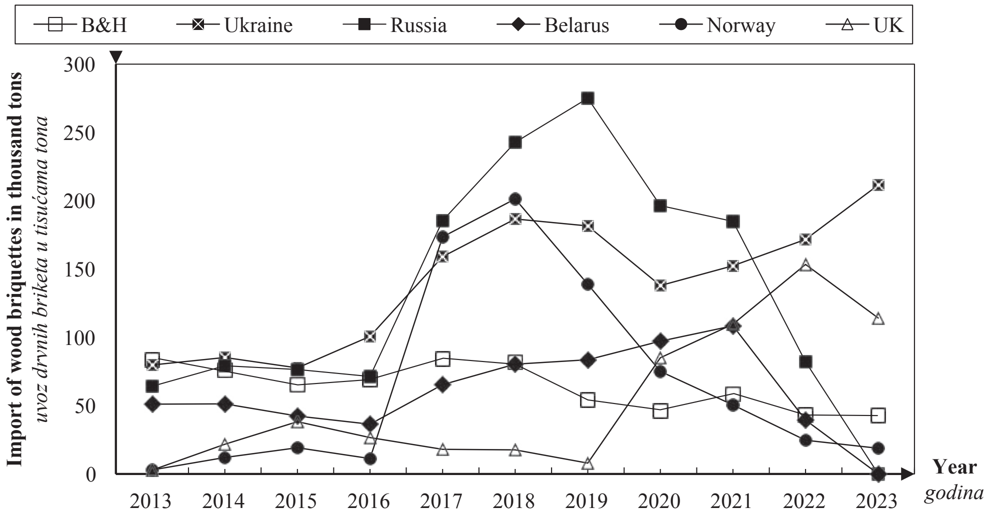

Figure 4 Major non-EU countries for the supply of wood briquettes to the EU-27 market in the period 2013-2023 (Eurostat, 2024)
Slika 4. Glavne zemlje izvan EU-a za opskrbu tržišta EU-27 drvnim briketima u razdoblju 2013. – 2023. (Eurostat, 2024.)

tons of wood briquettes from these countries, which accounted for 81.8 % of imports from non-EU countries (Russia 25.1 %, Norway 20.7 %, Ukraine 19.3 %, Bosnia and Herzegovina 8.4 %, and Belarus 8.3 %). In terms of supplying the EU market with wood briquettes, Russia and Belarus were replaced by Ukraine and the UK. A record EU import of wood briquettes from the UK was achieved in 2022 and from Ukraine in 2023. Imports from Bosnia and Herzegovina and Norway were reduced in the period 2022-2023. In the structure of wood briquette imports from non-EU countries to the EU in 2023, the share of the countries mentioned amounted to 91.6 % (Ukraine 50.0 %, the UK 27.0 %, Bosnia and Herzegovina 10.2 %, and Norway 4.4 %). In 2021, Russia supplied the most to Lithuania, Denmark and Germany, Belarus to Germany and Lithuania, Bosnia and Herzegovina to Slovenia, Austria, Italy and Germany, the United Kingdom to Sweden and Ireland, Norway to Sweden, Ukraine to Germany, Slovakia and Poland. In 2023, Bosnia and Herzegovina retained its markets, the United Kingdom and Norway also retained theirs, while Ukraine supplied Poland the most.

3.1.4 Major non-EU countries for the supply of sawdust and wood waste and scrap to the EU market

3.1.4. Glavne zemlje izvan EU-a za opskrbu tržišta Unije piljevinom te drvnim otpadom i ostatcima

During the analyzed period, Russia, Belarus and Norway were the largest suppliers of sawdust to the EU market. In 2021, the EU achieved a record import of 511,800 tons of sawdust from the mentioned countries, which corresponded to 99.5 % of imports from non-EU countries (Russia 38.9 %, Belarus 36.7 %, and Norway 23.9 %). In 2020, record imports from Russia of 219,300 tons were achieved, and from Belarus of 188,600 tons in 2021. Due to the reduction and subsequent suspension of imports from Russia and Belarus, the EU imported 46.9 % less sawdust from non-EU in 2022 than in 2021 and 53.4 % less in 2023 than in 2022. Russia and Belarus were replaced on the European market by the UK, from which the EU imported 7.8 – fold higher more sawdust in 2022 than in 2021 and 4.7 – fold higher more in 2023 than in 2022. Imports from Norway increased slightly in 2022 compared to 2021, while they decreased in 2023. Russia exported the most sawdust to Finland, and Belarus and Norway to Latvia. In 2023, Russia was replaced by Sweden on the Finnish market, while Latvia mainly imported sawdust from Lithuania, Denmark and Norway.

Norway, the UK, Switzerland, Russia and Ukraine were the largest suppliers of wood waste and scrap to the EU market in the period 2013-2021. The largest import from Russia of 221,700 tons was registered in 2016 and it did not exceed 130,000 tons until 2021 (Eurostat, 2024). In 2021, the EU imported 0.9 million tons of wood waste and scrap from the five largest suppliers, accounting for 77.1 % of imports from non-EU countries, and the share of the four countries rose to 78.6 % in 2022 and 82.2 % in 2023 (Eurostat, 2024). The suspension of imports from Russia mainly benefited the UK and Ukraine. Imports of wood waste and scrap from the UK, which were less than 1,000 tons in 2020, increased to 22,899 tons in 2022 and 102,269 tons in 2023 (Eurostat, 2024). In 2021, the EU imported 56,368 tons of wood waste and scrap from Ukraine, in 2022 the value increased to 102,015 tons, and in 2023 the import reached the level of 2021 again (Eurostat, 2024). In 2021, Russia supplied the most wood waste and scrap to Finland, Norway to Sweden, Switzerland to Germany, Austria, Italy and France, and the USA and Canada to France. In 2023, Russia was replaced on the Finnish market by Sweden and the UK, while the other countries retained their markets (Eurostat, 2024).

3.2 Value of EU wood fuel imports from non-EU countries

3.2. Vrijednost uvoza drvnoga goriva u EU iz zemalja nečlanica

Although imports from Russia and Belarus were reduced, the EU achieved a record level of imports of analyzed wood fuels from non-EU countries in 2022. The largest share of the record value of EU imports of all analyzed types of wood fuels of € 1.82 billion were wood pellets with a record import value of € 1.27 billion (Eurostat, 2024). In the value structure of EU imports from non-EU countries, wood pellets accounted for 69.5 % in 2022 (2021:61.4 %), while the share of wood chips was 20.3 % (2021:28.0 %), briquettes 5.5 %, wood waste and scrap 4.0 % and sawdust 0.7 %. In 2023, the value of EU imports of wood pellets decreased by 14.9 % compared to 2022, and the value of EU imports of selected wood fuels from non-EU countries decreased to € 1.59 billion (Eurostat, 2024). The trend towards growth in the value of EU imports from non-EU countries in the period 2022-2023 was also observed for wood waste and scrap, which increased from € 73.2 million (2022) to € 84.4 million (2023) (Eurostat, 2024). In contrast, the value of the import of wood chips has steadily decreased since 2019, when it reached a record value of € 400.5 million, to € 309.4 million in 2023. The same trend was recorded for sawdust, the import value of which fell continuously from a record value of € 14.4 million in 2019 to a level of € 9.9 million by 2023 (Eurostat, 2024). The record value of wood briquette EU imports of € 106.5 million was achieved in 2018 and decreased to € 99.5 million in 2022, i.e. to € 70.1 million in 2023 (Eurostat, 2024).

3.2.1 Value of EU imports of analyzed wood fuels from certain non-EU countries

3.2.1. Vrijednost uvoza analiziranih drvnih goriva u EU iz određenih zemalja nečlanica

In the period 2010-2021, Russia, the USA, Belarus, Ukraine, Brazil and Canada were the EU's largest foreign trade partners for the wood fuels analyzed. In 2021, the EU imported € 1.09 billion worth of wood fuels from these countries, which corresponded to 87.7 % of the value of imports from non-EU countries (Russia 33.4 %, the USA 25.3 %, Belarus 10.4 %, Brazil 7.5 %, Ukraine 7.0 %, Canada 4.1 %) (Eurostat, 2024). Although wood fuel imports from Russia and Belarus decreased in 2022, the EU recorded a record import value of € 1.45 billion from the six countries analyzed in the same year. However, the share of these countries in the value of EU imports from non-EU countries fell to 79.6 % in the same year (USA 37.4 %, Russia 12.3 %, Brazil 10.2 %, Ukraine 10.2 %, Canada 5.5 %, and Belarus 4.0 %) (Eurostat, 2024). In 2023, the value of EU imports from the fourth countries analyzed fell to € 1.11 billion, and their share in the value of imports from non-EU countries fell to 71.6 % (USA 41.9 %, Brazil 13.1 %, Ukraine 10.4 %, and Canada 6.2 %) (Eurostat, 2024).

During the period under review, Russia supplied the EU market with all types of wood fuels. In the value structure of EU imports from Russia in 2021, wood pellets had a share of 61.7 %, wood chips 31.4 %, wood briquettes 5.0 %, sawdust 1.2 % and wood waste and scrap 0.7 %. After the EU achieved a record import value of € 416.5 million from Russia in 2021, this value dropped to € 223.7 million in 2022 (Eurostat, 2024). Apart from wood waste and scrap, Belarus supplied the EU market with all other wood fuels analyzed. Wood chips and wood pellets accounted for the largest share in the value structure of EU imports from Belarus. The record value of EU imports of wood fuels from Belarus of € 156.4 million was reached in 2019 and fell to € 73 million by 2022 (Eurostat, 2024).

After the imports from Russia and Belarus were reduced and then suspended, the EU increased the import of wood fuels from other non-EU countries, which were the largest suppliers to this market. In 2022, a record level of wood fuel imports from the USA of € 681.3 million (2021: € 314.9 million) was reached, and that amount fell to € 650.7 million in 2023 (Eurostat, 2024). In 2022, wood pellets accounted for 95.8 of the value structure of EU imports from the USA (2021: 92.9 %) and wood waste and scrap for 4.2 %, while the share of pellets rose to 97.7 % in 2023 (Eurostat, 2024). During the analyzed period, Ukraine supplied the EU market with all types of wood fuels. The value of EU imports from Ukraine increased to € 186.1 million in 2022 (2021: € 87.5 million), falling to € 160.8 million in 2023 (Eurostat, 2024). In 2022, compared to 2021, the value of EU imports of wood pellets from Ukraine increased by € 62.2 million, wood briquettes by € 26.9 million, wood waste and scrap by € 7.3 million, wood chips by almost € 2.3 million, and only the value of sawdust imports decreased by € 90,334 (Eurostat, 2024). In 2023, only the value of the import of wood chips increased, while the value of the import of other fuels decreased.

Brazil supplied the EU market with wood chips and wood pellets. The value of EU imports of these fuels from Brazil amounted to € 185.5 million in 2022 (2021: € 93.8 million) and reached a record value of € 203 million in 2023 (Eurostat, 2024). In 2022, wood pellets accounted for 46.0 % of the value structure of EU imports from Brazil (2021: 35.2 %), and in 2023 it fell to 45.3 %. In 2022, the EU imported € 99.6 million worth of wood pellets and wood waste and scrap from Canada (2021: € 50.8 million), and the import value fell to € 95.9 million in 2023 (Eurostat, 2024). In 2022, the share of wood pellets in the value structure of EU imports from this country was 82.5 % (2021: 87.6 %), and in 2023 it fell to 73.3 %.

3.3 Meeting the EU's energy needs by importing wood fuels from non-EU countries

3.3. Zadovoljavanje energetskih potreba EU-a uvozom drvnih goriva iz zemalja nečlanica

In the period 2013-2022, the EU could meet less than 1.0 % of its energy needs in the household, commercial and public services, industry and agriculture

Table 2 Meeting energy demand (%) by imports of selected wood fuels from non-EU countries in the commercial and public services, households, industry and agriculture sectors in the EU-27
Tablica 2. Zadovoljavanje potražnje energije (%) uvozom odabranih drvnih goriva iz zemalja izvan EU-a u sektorima komercijalnih i javnih usluga, kućanstava, industrije i poljoprivrede u EU-27

Year / *Godina*[*]	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022
Share / *Udio*, %	0.36	0.39	0.35	0.38	0.45	0.58	0.65	0.66	0.64	0.58

[*] The calculation is based on the assumption that the total amount of sawdust imported from non-EU countries was used for the production of wood pellets in the EU-27 countries and on the assumption that the total amount of wood pellets, wood chips and wood briquettes imported from non-EU countries was used for energy production.

[*] *Izračun se temelji na pretpostavci da je ukupna količina piljevine uvezene iz zemalja izvan EU-a iskorištena za proizvodnju drvnih peleta u zemljama EU-27 te na pretpostavci da je ukupna količina drvnih peleta, drvne sječke i drvnih briketa uvezenih iz zemalja izvan EU-a korištena za proizvodnju energije.*

Table 3 Meeting energy demand (%) by imports of selected wood fuels from non-EU countries in the commercial and public services, households, industry and agriculture sectors in the EU-27
Tablica 3. Zadovoljavanje potražnje energije (%) uvozom odabranih drvnih goriva iz zemalja izvan EU-a u sektorima komercijalnih i javnih usluga, kućanstava, industrije i poljoprivrede u EU-27

Year / *Godina*	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022
Billions of m^3 *Milijarde* m^3	2.5	2.5	2.3	2.6	3.1	3.9	4.4	4.3	4.4	3.8

sectors by importing wood chips, wood pellets and wood briquettes from non-EU countries (Table 2). Due to a lack of knowledge about the structure, type of wood, moisture content and the type of wood fuel for whose production it could have been used, wood waste and scrap were not included in the calculation, which is why the data in Table 2 is lower than the actual values.

If the energy produced in the EU by importing wood fuels from non-EU countries in the period 2013-2022 were to be replaced by energy from natural gas, the required quantities of natural gas would be as shown in Table 3.

3.4 Regression models
3.4. Regresijski modeli

By developing regression models, it was found that EU imports of wood pellets from non-EU countries have a strong influence ($R^2 = 0.91$) on the consumption of wood pellets in the EU. The dependence studied is best described by a power model with an Eq. 1 of the form:

$$y = 8.639606 \cdot x_1^{0.611892} \tag{1}$$

Where:

y – Consumption of wood pellets in the EU (million tons);

x_1 – EU import of wood pellets from non-EU countries (million tons).

The power regression model exhibited a higher coefficient of determination and a lower standard error compared to the linear and exponential models. The parameters of the equation show that if EU imports of pellets from non-EU countries increase by 1.0 %, the consumption of wood pellets in the EU is expected to increase by 0.61 %. From the values of the t-statistics and the F-test, it can be concluded that parameters a and b are statistically significant, as is the correlation coefficient (Tables 4 and 5). The model is not loaded with autocorrelation (DW test).

Table 4 Results of power regression of wood pellet consumption in the EU (y) as a function of EU imports of wood pellets from non-EU countries (x_1)
Tablica 4. Rezultati potencijske regresije potrošnje drvnih peleta u EU-u (y) kao funkcije uvoza drvnih peleta u Uniju iz zemalja nečlanica (x_1)

Model	**Constant – *a*, millions of tons**	**Constant – *b***
	8.639606	0.611892
St. error	0.091512	0.066601
t-test (model)	23.56368	9.18749
t-test (table)	2.262	2.262
|*t*-test|	$\|t_a\| > t_{0.05}$	$\|t_a\| > t_{0.05}$

Designations in the table have the following meaning: y – consumption of wood pellets in the EU-27, x_1 – EU import of wood pellets from non-EU countries, St. error – standard error, *t*-test (model) – *t*-test value obtained in the power regression model, *t*-test (table) – *t*-test value from Student`s Table.
Značenje oznaka u tablici: y – potrošnja drvnih peleta u EU-27, x_1 – EU uvoz drvnih peleta iz zemalja izvan EU-a, St. error – standardna pogreška, t-test (model) – vrijednost t-testa dobivena u modelu potencijske regresije, t-test (tablica) – vrijednost t-testa iz Studentove tablice.

Table 5 Statistical characteristics of the regression model
Tablica 5. Statistički pokazatelji regresijskog modela

Statistical characteristic *Statistički pokazatelji*	**Values** / *Vrijednosti*
R	0.95573494
R^2	0.91342927
R^2_{cor}	0.90260793
$F_{(1.8)}$	84.410 ($F_{test\ (0.05)}$: +)
SE	0.06182
DW	1.792520 ($DW_{test\ (0.05)}$: +)

Designations in the table have the following meaning: R – coefficient of correlation, R^2 – coefficient of determination, R^2_{cor} – corrected coefficient of correlation, F – F-statistic, SE – standard error, DW – the Durbin-Watson statistic, 0.05 – significance level.
Značenje oznaka u tablici: R – koeficijent korelacije, R^2 – koeficijent determinacije, R^2_{cor} – korigirani koeficijent korelacije, F – F-statistika, SE – standardna pogreška, DW – Durbin-Watsonova statistika, 0,05 – razina značajnosti.

Regression models were created to determine the impact of EU wood pellet imports from Ukraine on wood pellet consumption in the EU. It was found that the dependence analyzed is best described by the power regression model, whose Eq. 2 has the following form:

$$y = 7.1207 \cdot x_2^{0.270515} \tag{2}$$

Where:

y – Consumption of wood pellets in the EU (million tons);

x_2 – EU value of wood pellet imports from Ukraine (€ million).

The power regression model exhibited a higher coefficient of determination and a lower standard error compared to the linear and exponential models. The high value of the correlation coefficient and its statistical significance (F-test) show that there is a strong correlation between the phenomena analyzed. The value of the coefficient of determination shows that 94 % of the changes in the consumption of wood pellets in the EU can be explained by changes in the

Table 6 Results of power regression of wood pellet consumption in the EU (y) as a function of wood pellet imports from Ukraine (x_2)
Tablica 6. Rezultati potencijske regresije potrošnje drvnih peleta u EU-u (y) kao funkcije uvoza drvnih peleta u Uniju iz Ukrajine (x_2)

Model	Constant – *a*, 10^6 t	Constant – *b*
	7.1206997	0.270515
St. error	0.094521	0.024803
t-test (model)	20.76805	10.90645
t-test (table)	2.262	2.262
|*t*-test|	$\|t_a\| > t_{0.05}$	$\|t_a\| > t_{0.05}$

Designations in the table have the following meaning: y – consumption of wood pellets in the EU-27, x_2 – EU import of wood pellets from Ukraine, St. error – standard error, *t*-test (model) – *t*-test value obtained in the power regression model, *t*-test (table) - *t*-test value from Student`s Table.
Značenje oznaka u tablici: y – potrošnja drvnih peleta u EU-27, x_2 – EU uvoz drvnih peleta iz Ukrajine, St. error – standardna pogreška, t-test (model) – vrijednost t-testa dobivena u modelu potencijske regresije, t-test (tablica) – vrijednost t-testa iz Studentove tablice.

Table 7 Statistical characteristics of the regression model
Tablica 7. Statistički pokazatelji regresijskog modela

Statistical characteristic *Statistički pokazatelji*	**Values** / *Vrijednosti*
R	0.96797905
R^2	0.93698344
R^2_{cor}	0.92910637
$F_{(1.8)}$	118.95 ($F_{test\ (0.05)}$: +)
SE	0.05274
DW	2.28266 ($DW_{test\ (0.05)}$: +)

Designations in the table have the following meaning: *R* – coefficient of correlation, R^2 – coefficient of determination, R^2_{cor} – corrected coefficient of correlation, *F* – *F*-statistic, *SE* – standard error, *DW* – the Durbin-Watson statistic, 0.05 – significance level.
Značenje oznaka u tablici: R – koeficijent korelacije, R^2 – koeficijent determinacije, R^2_{cor} – korigirani koeficijent korelacije, F – F-statistika, SE – standardna pogreška, DW – Durbin-Watsonova statistika, 0,05 – razina značajnosti.

value of pellet imports from Ukraine (Tables 6 and 7). Parameters a and b are statistically significant, and the model is not loaded with autocorrelation (DW test). The parameters of the equation show that if EU import of wood pellets from Ukraine increases by 1.0 %, the consumption of wood pellets in the EU is expected to increase by 0.27 %.

3.5 Interpretation of analysis results
3.5. Interpretacija rezultata analize

Until 2021, non-EU countries were the major suppliers of the EU market with wood chips and from 2022 with wood pellets. The importance of non-EU countries for supplying the EU market with wood briquettes, sawdust, wood waste and scrap is significantly lower compared to wood chips and wood pellets. EU imports of wood chips from non-EU countries began to decline in 2020 and were further reduced by the cessation of imports from Russia and Belarus. In 2022-2023, the EU increased its imports of wood chips from other major non-EU supplier countries such as Brazil, Uruguay and Norway. However, imports of wood chips in 2023 were the lowest imports from non-EU countries in the period 2010-2023.

In contrast to wood chips, EU imports of wood pellets from non-EU countries reached a record level in 2022, only to fall slightly in 2023. It is assumed that the decline in imports is due to a mild winter in the 2023/2024 heating period and that there are still stocks on the market from the 2022 imports. In 2022-2023, the EU was able to maintain the stability of the wood briquette market thanks to the increase in imports from its members, as imports from non-EU countries decreased. The EU market would have been even more destabilized by the interruption of imports of wood briquettes from Russia and Belarus if imports from the Ukraine had not increased in the period 2022-2023. The suspension of imports of sawdust from Russia and Belarus has seriously affected the supply of the EU market, as these two countries, together with Norway, were the three largest suppliers to this market. Imports of wood waste and scrap into the EU decreased in 2022 compared to 2021 and increased again in 2023. Russia was less important for the supply of the EU market, which is why it did not destabilize it. The supply of wood chips, wood briquettes, and sawdust to the EU market was destabilized due to disruptions in imports from Russia and Belarus, in contrast to wood pellets and wood waste and scrap. In the period 2022-2023, the EU did not find new non-EU trading partners that could replace Russia and Belarus in supplying its market with woodchips, briquettes and sawdust. EU imports of the aforementioned fuels from non-EU countries therefore show a negative trend in the period 2022-2023. In contrast, Russia and Belarus were replaced by the USA in the supply of pellets to the EU market. As a result, the EU reached a record level of pellet imports from non-EU countries in 2022.

Until 2021, the EU paid the most for imports of analyzed wood fuels to Russia, the USA, Belarus, Ukraine, Brazil and Canada. Due to the decline and subsequent suspension of imports from Russia and Belarus, the value of EU wood fuel imports from all other major non-EU supplier countries increased. In 2022, the value of EU imports of wood fuels from the USA, Brazil, Ukraine, Canada and the United Kingdom doubled, while in 2023 the value of imports from Brazil and the United Kingdom increased and the value of imports from the USA, Canada and Ukraine decreased (Eurostat, 2024).

Given the results obtained in meeting energy needs in the EU sectors of households, commercial and public services, industry and agriculture by importing analyzed wood fuels, it is clear that the suspension of imports from Russia and Belarus will not cause important changes in the supply of energy to the EU market.

The impact of Ukraine on the EU wood pellet market should be highlighted in particular, given the values of the parameters obtained in the regression model. Given the current situation in Europe, it is difficult to predict Ukraine's role in supplying wood pellet to the EU market in the coming years.

4 CONCLUSIONS
4. ZAKLJUČAK

The study whose results are presented in this paper, was carried out using official statistical data taken from two sections of the Eurostat database, one relating to international trade in goods and the other to energy trade. In the period 2010-2023, the EU imported mainly wood chips and wood pellets from non-EU countries and significantly less wood briquettes, sawdust and wood waste and scrap. In all years of the analyzed period, EU members were the largest suppliers of selected wood fuels to their own market, except for the period 2018-2021, when non-EU countries dominated the supply of wood chips to the EU market. The EU achieved record imports of wood briquettes from non-EU countries in 2018, wood chips in 2019, sawdust in 2021 and wood pellets and wood waste and scrap in 2022.

The suspension of imports from Russia and Belarus in 2022-2023 has hit the EU wood chip market the hardest. The decline in EU imports of wood chips in that period continued the negative trend in imports of this fuel from non-EU countries that began in 2020. The EU market of wood pellets was stable in the period 2022-2023, mainly thanks to the USA. Imports of other analyzed wood fuels from non-EU countries are significantly lower compared to wood chips and wood pellets, which is why they have a smaller impact on the functioning of the EU market. Depending on the type of wood fuel, Russia and Belarus have been replaced in the supply of the EU market by other non-EU countries, most frequently by the USA, Ukraine and the United Kingdom. In the period 2022-2023, an increase in wood fuel imports was also recorded from other major non-EU countries.

Until 2021, Russia was the major supplier of wood pellets to the EU market and Belarus of wood chips and wood pellets. In 2021, the EU recorded a record value of wood fuel imports from Russia of € 416.5 million and from Belarus of € 126.9 million, which in total represented 30.0 % of the value of wood fuel imports from non-EU countries. In the period 2013-2022, by importing analyzed wood fuels from non-EU countries, the EU could meet 0.35 % (2015) and 0.66 % (2020) of energy needs in the commercial and public services sector, households, industry and agriculture. According to the values determined, the suspension of wood fuel imports from Russia and Belarus is not expected to result in any significant changes in final energy consumption in the selected sectors on the EU market. Regression models have shown that EU imports of wood pellets from non-EU countries have impact on the consumption of this fuel in the EU. Imports of wood pellets from Ukraine were also found to have influence on the consumption of this fuel in the EU. Further research in this area should focus on the factors of price and non-price competitiveness of wood pellets supplied to the EU market from non-EU countries. The suspension of imports from Russia and Belarus could be an opportunity for non-EU countries in south-eastern Europe to strengthen their role in supplying the EU market with wood pellets.

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The Effect of Different Relative Humidity Conditions on Mechanical Properties of Historical Fir Wood Under the Influence of Natural Aging

Utjecaj relativne vlažnosti zraka na mehanička svojstva ostarjelog drva jele izloženoga prirodnom starenju

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ABSTRACT • *Wood undergoes degradation and aging due to various physical and biological factors throughout its lifespan. Among these factors, environmental conditions such as relative humidity, temperature, rainfall, and UV radiation play a crucial role in the physical, chemical, and biological deterioration of wood. In this study, the effects of relative humidity on the density, bending strength, and compressive strength of wood were investigated. Fir beam elements from a traditional wooden structure, estimated to be approximately 150 years old, and freshly cut fir wood specimens were tested under relative humidity conditions of (30 ± 5) %, (65 ± 5) %, (85 ± 5) %, and (95 ± 5) % at a temperature of (20 ± 2) °C. A one-way ANOVA analysis was conducted to evaluate the effects and interactions of different relative humidity conditions on density, bending strength, and compressive strength for both naturally aged fir and freshly cut fir specimens. The results indicate that naturally aged fir wood exhibits greater stability in terms of density; however, strength loss is more pronounced under high relative humidity conditions. These findings suggest that natural aging has a significant impact on the preservation of wooden structures with historical and cultural value and contribute to a better understanding of such structures.*

KEYWORDS: *fir; natural aging; relative humidity; historical wood*

SAŽETAK • *Drvo je tijekom svoga životnog vijeka zbog raznih fizičkih i bioloških čimbenika podložno razgradnji i starenju. Među tim čimbenicima odlučujući utjecaj na fizičko, kemijsko i biološko propadanje drva imaju relativna vlažnost zraka, temperatura, padaline i UV zračenje. U ovom su istraživanju ispitivani učinci relativne vlažnosti zraka na gustoću drva, čvrstoću na savijanje i čvrstoću na tlak. Elementi jelovih greda iz tradicionalne drvene konstrukcije, približne procijenjene starosti od 150 godina, i uzorci svježe posječene jelovine ispitani su pri relativnoj vlažnosti zraka 30 ± 5 %; 65 ± 5 %; 85 ± 5 % i 95 ± 5 % te pri temperaturi 20 ± 2 °C. Provedena je jednosmjerna*

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ANOVA analiza kako bi se procijenili učinci i interakcije različitih uvjeta relativne vlažnosti zraka na gustoću, čvrstoću na savijanje i čvrstoću na tlak prirodno ostarjelih i svježe posječenih uzoraka jelovine. Rezultati su pokazali da prirodno ostarjela jelovina pokazuje veću stabilnost u smislu gustoće, međutim, njezin gubitak čvrstoće u uvjetima visoke relativne vlažnosti zraka veći je od gubitka čvrstoće uzoraka svježe posječenog drva. Ti rezultati sugeriraju da prirodno starenje znatno utječe na očuvanje drvenih konstrukcija povijesne i kulturne vrijednosti te pridonose boljem razumijevanju tih konstrukcija.

KLJUČNE RIJEČI: *jelovina; prirodno starenje; relativna vlažnost; ostarjelo drvo*

1 INTRODUCTION
1. UVOD

Wood is a natural polymer primarily composed of cellulose, hemicellulose, and lignin. These components provide structural support to the living tree and confer a certain degree of resistance against microorganisms. Due to the partially crystalline nature of cellulose in wood, it exhibits some resistance to microbial attack. Additionally, lignin, which consists of phenylpropane units, functions as a heterogeneous polymer and offers considerable resistance to certain decay fungi (Scheffer and Morrell, 1998).

Wood is a natural material and, like many other natural materials found in nature, it is hygroscopic, meaning it absorbs moisture from its surroundings. The moisture interaction between wood and the surrounding atmosphere varies depending on the relative humidity and temperature of the air, as well as the existing moisture content in the wood (Glass and Zelinka, 2021). From a materials science perspective, the relative humidity range between 0 % and approximately 95 - 98 % is generally referred to as the *"hygroscopic moisture range",* whereas humidity levels exceeding this threshold are termed the *"over-hygroscopic moisture range".* The dominant mechanisms of moisture absorption in wood behave differently within these two ranges. In the hygroscopic range, water primarily binds to the hydrogen bonds in the cell walls. In the over-hygroscopic range, water uptake predominantly occurs through capillary condensation within macro voids, such as cell lumina and pit chambers, external to the cell walls. This over-hygroscopic range is also the moisture level at which fungal decay in wood occurs (Fredriksson, 2019).

The moisture content in wood has a significant impact on its performance, influencing important material properties such as resistance to degradation, mechanical properties, and dimensional stability (Thybring and Fredriksson, 2021). In the short term, moisture in wood often leads to surface deterioration, which can be mitigated by protective coatings such as varnish, lacquer, or paint (Glass and Zelinka, 2021). However, the long-term effects of moisture fluctuations become more pronounced, particularly in humid and warm regions where atmospheric moisture levels are high. For instance, prolonged exposure to varying relative humidity reduces the long-term strength of wood elements under load. Additionally, it exacerbates creep effects, leading to excessive deformation, cracking, or premature failure (Wang *et al*., 2021).

Many physical environmental conditions, such as relative humidity, not only play a role in the degradation of wood but also influence its aging mechanisms. The aging process in wood begins when a tree is felled. In the absence of microbial influence, changes in the chemical components of wood occur very slowly and are dependent on environmental conditions (Fengel, 1991). On the other hand, Aydın and Aydın (2020) noted that the degradation period is not only dependent on the type and conditions of the environment, such as the wood contact with soil, but also on the region location, the material position, and the wood species. Various studies in the literature have examined the material properties, general behavior, and condition of wood under the influence of different physical factors (such as sunlight, temperature, and humidity) during natural aging. One such study was conducted by Matsuo *et al*. (2011). In this study, the color properties of wood samples exposed to natural aging effects and those subjected to temperatures between 90 °C and 180 °C were compared. The study concluded that natural aging causes a slow and slight oxidation of wood color, while thermal treatment accelerates the color changes occurring during aging. Popescu *et al*. (2009) used X-ray photoelectron spectroscopy (XPS) to analyze wood samples from different-aged and stored linden trees. The study analyzed the chemical changes on the surfaces of six-year-old, undamaged wood and degraded wood samples approximately 150, 180, and 250 years old using the XPS method. The study found that the surface of the 150-year-old wood samples (early aging stage) exhibited significant chemical changes, with the highest ratio of carbon atoms and the lowest ratio of oxygen atoms during this aging stage. Zhang *et al*. (2024) studied and compared the moisture content (MC), density, compressive strength parallel to grain (CSPG), conventional static bending strength (BS), modulus of elasticity in static bending (MOE-BS), shear strength parallel to grain (SSPG), compressive strength perpendicular to the grain (CSEG), chemical composition, and microstructure of both new and old wood samples from five different tree species in restored buildings. The study concluded that significant changes were observed in the physical and mechanical properties of wood subjected to natural aging. However, the extent of these changes was found to vary depending on the wood species, initial properties, storage conditions, and environmental factors. In a study conducted by Topaloglu (2023), changes in the properties of wood from two façade elements of a traditional building approximately 100 years old were examined during the natural aging process. FTIR analysis in the study revealed that the cellulose and lignin on the surface of aged wood had degraded over time. It was found that the moisture content and density values of the aged wood were lower than those of the new wood samples, and the water absorption rate of the aged wood increased with the natural aging process. Liu *et al*. (2019) investigated the effects of sunlight and artificial light sources on wood subjected to outdoor conditions by exposing three different wood species to natural sunlight for 733 days and artificial xenon light for 180 hours. The study found that wood exposed to artificial xenon light showed more severe aging compared to wood exposed to natural sunlight. Additionally, the aging process under artificial xenon light was approximately 30 times faster than natural aging. Cavalli *et al*. (2016) reviewed the differences in the mechanical properties of wood elements of different ages through literature analysis and examined the research findings. Additionally, based on their analysis, they provided recommendations for future studies. While studies in the literature have focused on changes in material properties of wood elements under the influence of factors such as sunlight and temperature, as previously discussed, there are also studies on the material properties of naturally aged wood under different relative humidity conditions. One such study, conducted by Gereke *et al*. (2011), investigated the behavioral differences between new and naturally aged wood. The study concluded that similarities in moisture behavior were observed between the old and new wood samples. Han *et al*. (2023) explored the effects of natural aging on the moisture sorption behavior of structural components of a historic wooden building in China. In this study, three experimental groups were formed: naturally aged, decayed wood samples; aged, sound wood samples; and reference wood samples. The results showed that the naturally aged decayed wood samples were more hygroscopic than the aged sound wood samples, and both of these samples were more hygroscopic than the reference wood samples. Additionally, the hemicellulose and lignin contents of the samples were found to change with the aging of the wood. Xin *et al*. (2024) investigated the effects of environmental factors on natural aging and the aging mechanism of larch (*Larix principis-rupprechtii Mayr*) timber. Experimental studies were conducted on 130 samples of larch timber processed from the outer wood (sapwood) region to examine the effects of ultraviolet (UV) irradiation time and drying-wetting (D-W) cycles on the color, physical, and mechanical properties of timber samples. The study concluded that the timber color was significantly affected by UV irradiation and D-W cycle aging. During the D-W cycle, the density and bending properties of the timber decreased significantly, while the effect of UV irradiation was found to be weak. Žlahtič-Zupanc *et al.* (2018) prepared samples from European oak (*Quercus robur / Q. petraea*), sweet chestnut (*Castanea sativa*), European larch (*Larix decidua*), Scots pine (*Pinus sylvestris*) heartwood and sapwood, Norway spruce (*Picea abies*), and beech (*Fagus sylvatica*) trees and investigated the moisture behavior of wood under outdoor conditions. Two different sample groups were prepared for the study. Small samples were exposed to natural outdoor conditions for 9, 18, and 27 months and then analyzed in the laboratory using various methods (contact angle, short- and long-term water absorption, and water vapor absorption). Large samples were also placed outdoors in a single layer and equipped with moisture monitoring sensors for 18 months. The results showed that exposure to natural outdoor conditions could change the wood performance with respect to water, and this change was more pronounced in thermally modified wood, where a decrease in moisture performance was observed.

Recent studies in the literature have focused on the physical and mechanical properties of wood materials under the influence of natural aging. However, there are few studies in the literature that compare the changes in the physical and mechanical properties of wood used in traditional structures and naturally aged wood with freshly cut wood under different relative humidity conditions. The aim of this study is to determine the effect of the natural aging process on the physical and mechanical properties of fir wood. In this context, the changes and differences in the mechanical properties of wood obtained from fir trees used as structural materials in historical buildings, which have undergone natural aging, and freshly cut fir wood were examined under specific standards and different environmental relative humidity conditions. By comparing naturally aged fir and freshly cut fir samples, the changes in fundamental properties such as density, bending strength, and compressive strength of the material under different relative humidity conditions were analyzed. ANOVA analysis was used in the study to determine statistically significant differences. The findings identified the behavior and performance changes of the fir wood from a historical building, approximately 150 years old, under different environmental relative humidity conditions. The results of this study provide important insights, especially in terms of the long-term durability and safety of historical wooden structures and objects. Furthermore, by analyzing the material behavior of fir wood under different environmental relative humidity conditions, it is believed that this study will fill an important gap in the literature regarding conservation and restoration works.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

The wood material used in this study was obtained from the load-bearing wooden beam of a traditional wooden structure, estimated to be approximately 150 years old, located in Ankara, Türkiye, and currently undergoing restoration. The building consists of a basement, ground floor, first floor, and second floor, and it was constructed using the traditional wooden frame technique with adobe infill between the wooden frames (Survey Report, 2021). Over time, the wooden elements in the building have deteriorated due to environmental effects, leading to color changes, dimensional changes, decay, and cross-sectional losses in the wood. Specifically, these effects have been more pronounced in the load-bearing wooden posts and beams in the basement, which are more exposed to moisture, and in the wooden frame wall structure located on the facade of the building (Figure 1). Due to the decision to proceed with reconstruction of the building, a load-bearing wooden beam, approximately 70 mm × 100 mm × 1200 mm in size, was taken from the damaged, deteriorated, and unusable first-floor ceiling beams and examined in the experimental study. Upon examining the cross-section of the historical wooden sample, it was observed that it mostly consisted of juvenile wood and exhibited numerous nail joint holes, cracks, and biologically induced degradation.

For comparison in the study, a piece of freshly cut solid timber of the same species, with a high juvenile wood ratio, was selected and supplied from the sawmill.

2.1 Wood identification and determination of anatomical properties

2.1. Identifikacija drva i određivanje njegovih anatomskih svojstava

To initially identify the wood species of the sample obtained from the historic structure, samples of dimensions 10 mm × 10 mm × 10 mm were taken. The samples were boiled in pure water until they sank to the bottom to soften the wood tissue and allow the air within the tissue to escape, preparing them for sectioning. The samples were then stored in a mixture of alcohol, glycerin, and pure water in a 1/1/1 ratio until they were ready to be sectioned. To prevent fungal contamination, a small amount of phenolic acid was added to the mixture (Gerçek, 2011; Merev, 1998). Then, sections were obtained in three different orientations – transverse, radial, and tangential – using a Reichert sliding microtome at a thickness of 15-20 microns. The obtained sections were bleached in sodium hypochlorite for 5-10 minutes and then washed with pure water. Prior to staining, 1-2 drops of acetic acid were added to the environment for pH balancing, and after waiting for 1-2 minutes, the samples were washed again with pure water. After these processes, the sections were stained in 50 % safranin 0 for 5 minutes. Once the staining was complete, the sections were transferred to a 50 % alcohol-water mixture. Standard preparation techniques were applied to the sections, and permanent preparations were made in glycerin gela-

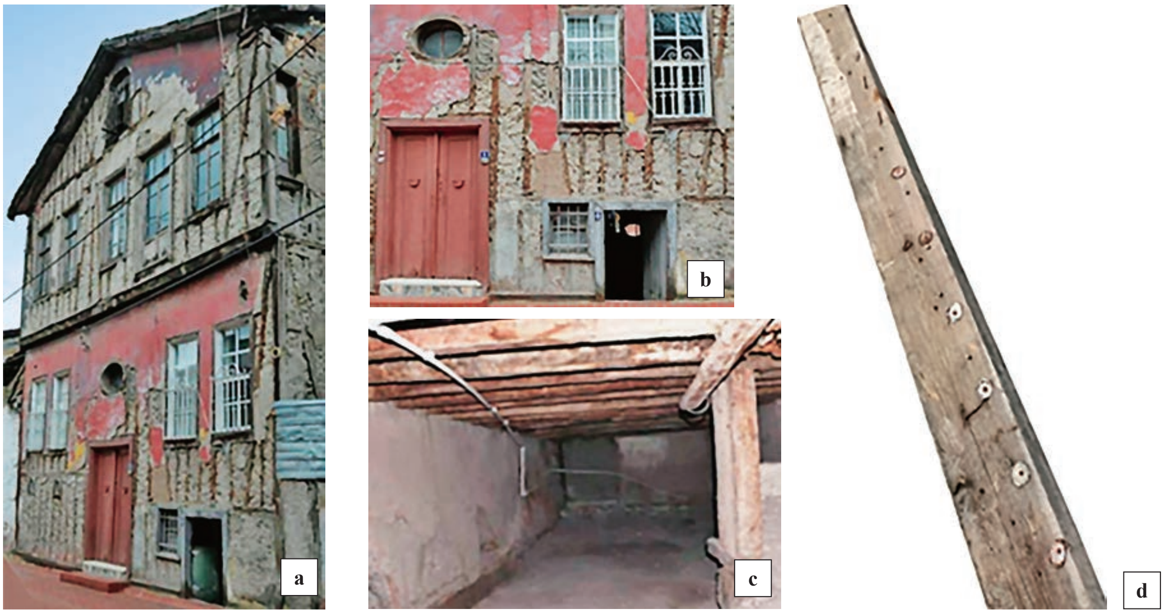

Figure 1 Traditional wooden structure selected for the experimental study (Survey Report, 2021): a) adobe infill between the wooden frame system, b) color and dimensional changes in wooden elements of the ground floor, c) color changes and decay in wooden elements of the basement, d) wooden beam sample taken for the experimental study

Slika 1. Tradicionalna drvena građevina odabrana za eksperimentalno istraživanje (izvješće o istraživanju, 2021.): a) ispuna od ćerpiča između sustava drvenih okvira, b) promjene boje i dimenzija drvenih elemenata prizemlja, c) promjene boje i propadanje drvenih elemenata podruma, d) uzorak drvene grede uzet za eksperimentalno istraživanje tin (Ives, 2001). The microphotographs of the permanent preparations of the wood samples were taken with the help of a digital camera attached to an Olympus BX50 research microscope and analyzed using BAB Bs200ProPlus Image Processing and Analysis Software (Bab, 2000). The obtained photographs were compared with wood atlases and reference preparations. As a result of these comparisons, the original wood species and genus of the samples taken from the historic structure were determined.

2.2 Density, bending, and compression tests
2.2. Ispitivanje gustoće, čvrstoće na savijanje i čvrstoće na tlak

After the wood samples obtained from the historical structure and freshly cut wood were prepared to appropriate test dimensions, they were conditioned in a climate chamber (Figure 2) at (20 ± 2) °C. Initially, the samples were kept at (30 ± 5) % relative humidity (RH) until they reached equilibrium moisture content, followed by conditioning at relative humidity values of (65 ± 5) %, (85 ± 5) %, and (95 ± 5) %, respectively, until equilibrium moisture content was reached (Figure 2). Once the samples achieved the desired equilibrium moisture content at the specified conditions, density, bending, and compression tests were performed separately for each moisture group (Table 1). Due to the limited number of test samples obtained from the historical structure, additional samples of the same quantity and dimensions were prepared from freshly cut wood for comparison.

2.2.1 Determination of densities at different relative humidity values
2.2.1. Određivanje gustoće uzoraka drva pri različitim vrijednostima relativne vlažnosti zraka

The density test was conducted in accordance with ISO 13061-2 standard. Samples measuring 20 mm × 20 mm × 30 mm were cut from the wood material obtained from both the historical building and the freshly cut wood. For each test group, 10 prepared samples were initially conditioned in a climate chamber at (20 ± 2) °C and a relative humidity of (30 ± 5) % until they reached equilibrium moisture content. Once the samples reached equilibrium moisture content, their weights were quickly measured using a precise scale, and the cross-sectional dimensions were recorded. This procedure allowed the calculation of the sample density at (20 ± 2) °C and (30 ± 5) % relative humidity. The same test sample was subsequently used for relative humidity values of (65 ± 5) %, (85 ± 5) %, and (95±5) %, and the samples were conditioned in the

Figure 2 Conditioning of the prepared test samples in the climate chamber
Slika 2. Kondicioniranje pripremljenih ispitnih uzoraka u klimatizacijskoj komori

Table 1 Experimental work plan
Tablica 1. Plan istraživanja

Experimental group *Vrsta uzoraka*		**Experimental tests** *Ispitivani parametri*	**Specimen dimensions, mm** *Dimenzije uzoraka, mm*	**Number of specimens** *Broj uzoraka*				**Total number of specimens** *Ukupan broj uzoraka*
				30 % RH	**65 % RH**	**85 % RH**	**95 % RH**	
1	Historical wood *ostarjelo drvo*	Density / *gustoća*	20×20×30	10	10	10	10	10*
		Bending *čvrstoća na savijanje*	20×20×330	5	5	5	5	20
		Compression *čvrstoća na tlak*	20×20×30	8	8	8	8	32
2	New wood (Control group) *svježe posječeno drvo (kontrolni uzorci)*	Density / *gustoća*	20×20×30	10	10	10	10	10*
		Bending *čvrstoća na savijanje*	20×20×330	5	5	5	5	20
		Compression *čvrstoća na tlak*	20×20×30	8	8	8	8	32

* In the density test, the same sample was used for the relevant relative humidity values
* *U ispitivanju gustoće upotrijebljen je isti uzorak za ispitivanje pri različitim relativnim vlažnostima zraka.*

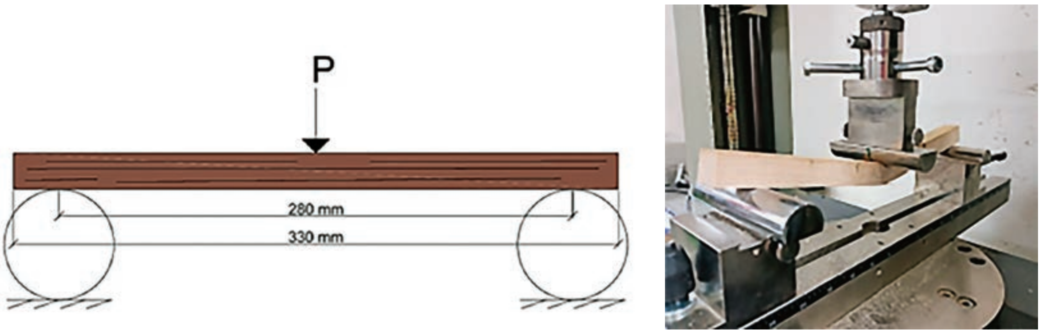

Figure 3 Bending test: left – experimental setup, right – application of bending test
Slika 3. Ispitivanje čvrstoće na savijanje: lijevo – eksperimentalni postav, desno – prikaz ispitivanja

climate chamber. After the sample reached equilibrium moisture content in the climate chamber at (20 ± 2) °C and relative humidity values of (65 ± 5) %, (85 ± 5) %, and (95 ± 5) %, the density was recalculated for each of the relevant humidity values.

2.2.2 Determination of bending resistance at different relative humidity values

2.2.2. Određivanje čvrstoće na savijanje pri različitim vrijednostima relativne vlažnosti zraka

The bending strength determination test was conducted in accordance with ISO 13061-3 standard (Figure 3). Five samples of 20 mm × 20 mm × 330 mm were cut and prepared from both the historical structure and newly cut wood. The prepared samples were conditioned at (20 ± 2) °C and (30 ± 5) % relative humidity until they reached constant mass in a climate chamber. Once these conditions were met, the samples were placed between two parallel cylindrical supports. A force was applied to the test samples at a constant speed from a single point on the top during the experiment. Upon completion of this process, an additional set of five specimens was prepared, and the same procedures were systematically conducted under relative humidity conditions of (65 ± 5) %, (85 ± 5) %, and (95 ± 5) %, with the bending strengths recorded accordingly.

2.2.3 Determination of compressive strength at different relative humidity values

2.2.3. Određivanje čvrstoće na tlak pri različitim vrijednostima relativne vlažnosti zraka

The compressive strength test was conducted in accordance with ISO 13061-17 standard (Figure 4). Samples measuring 20 mm × 20 mm × 30 mm were prepared from the wood material obtained from both the historical building and freshly cut wood, with eight samples of each. The prepared samples were first conditioned in a climate chamber at a temperature of (20 ± 2) °C and a relative humidity of (30 ± 5) % until they reached equilibrium moisture content. Afterward, the samples were removed from the climate chamber and subjected to a compressive strength test parallel to the fibers. Upon completion of this process, an additional set of eight specimens was prepared, and the same procedures were systematically conducted under relative humidity conditions of (65 ± 5) %, (85 ± 5) %, and (9 ± 5) %, with the compression strengths recorded accordingly.

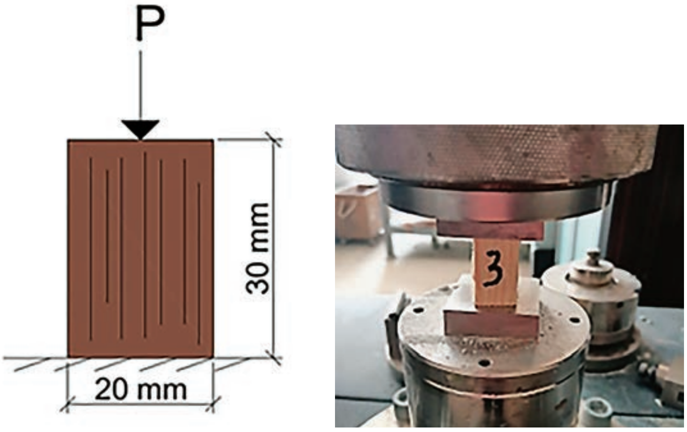

Figure 4 Compression test: left – experimental setup, right – application of compression test
Slika 4. Ispitivanje čvrstoće na tlak: lijevo – eksperimentalni postav, desno – prikaz ispitivanja

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

As a result of the species identification test conducted on the original wood samples, it was determined that the original wood samples belong to the *Abies* sp. (fir) genus. For comparison purposes, a control group consisting of newly cut, solid, and juvenile wood-rich (juvenile wood) *Abies* sp. (Fir) wood was selected for both original and solid naturally aged *Abies* sp. wood. Anatomical cross-sectional images of the original historical *Abies* sp. (fir) sample are shown in Figure 5.

Figure 6 shows the density distributions of naturally aged solid fir wood and freshly cut solid fir wood under different relative humidity conditions, presented as whisker plot graphs. In addition, one-way ANOVA was applied to determine the statistically significant differences in the density, bending strength, and compressive strength values of naturally aged fir wood and new solid fir samples under different relative humidity conditions. The results are shown in Table 2. The ANOVA results indicated significant differences between the groups in both the new ($F = 7.908$, $p < 0.001$) and naturally aged ($F = 7.336$, $p = 0.001$) fir samples. According to the Duncan test results, the samples at the 65 % relative humidity level were significantly different from the other groups. These results suggest that the interaction between relative humidity and the aging process of fir wood may vary, and that naturally aged fir wood may exhibit a more stable structure in terms of density at lower relative humidity levels. Upon examining Figure 6, the median density of naturally aged fir samples was generally higher than that of new fir wood. This was only different at the 65 % relative humidity level, where the median density of the new fir samples was considerably higher than all other samples. On the other hand, the generally higher density of naturally aged fir wood compared to new solid fir wood indicates that the aging process has a positive effect on the density of fir wood and leads to an increase in density. This result was also observed in the work of Zhang *et al*. (2024) on camphor tree (*Cinnamomum camphora*) and catalpa (*Catalpa ovata*) species, where wood density increased with aging. However, in the same study, the opposite result was found for Chinese arborvitae (*Platycladus orientalis*), red maple (*Acer palmatum*), and Scots pine (*Pinus sylvestris*), where density decreased with increased aging. Similarly, Unver *et al*.

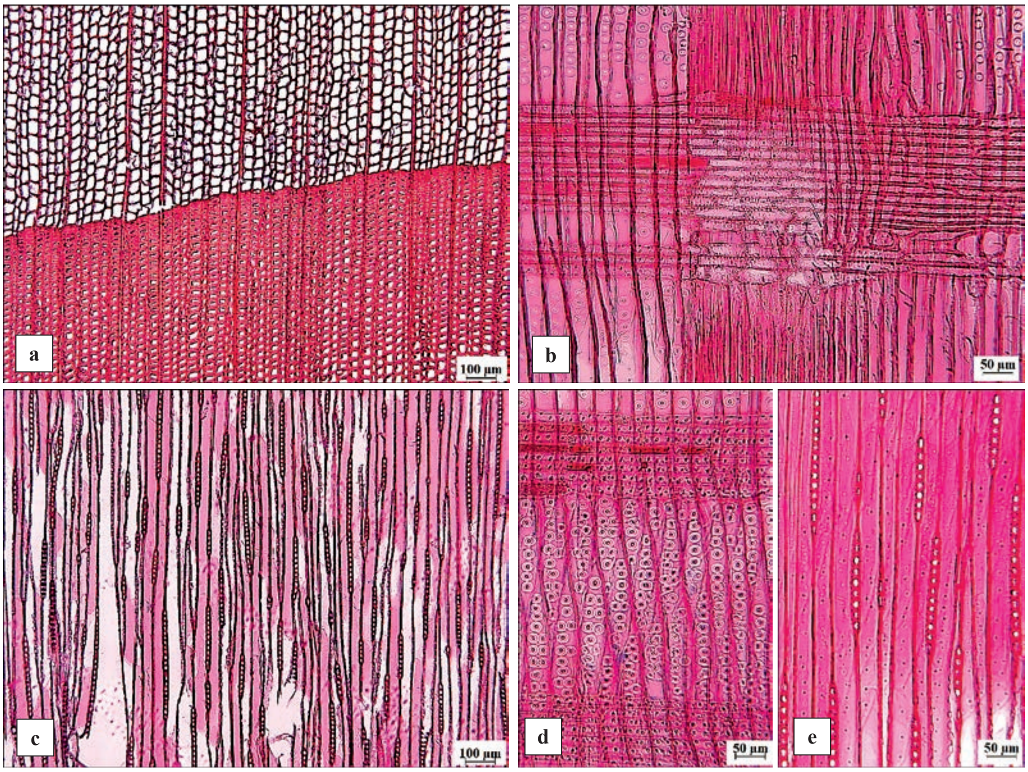

Figure 5 *Abies* sp. (Fir) wood: a) Transverse section showing distinct growth rings, and gradual transition from earlywood to latewood; b) Radial section showing homogeneous rays, and crystals present in the marginal and submarginal ray cells; c) Tangential section showing exclusively uniseriate rays; d) Radial section showing uniseriate and biseriate pits on tracheid radial wall; e) Tangential section showing small pits on tracheid tangential walls

Slika 5. Drvo jele (*Abies* sp.): a) poprečni presjek s jasnim godovima i postupnim prijelazom iz ranoga u kasno drvo; b) radijalni presjek koji prikazuje homogene trakove i kristale u marginalnim i submarginalnim stanicama trakova; c) tangentni presjek koji prikazuje isključivo jednostruke trakove; d) radijalni presjek koji prikazuje jednostruke i dvostruke jažice na radijalnoj stijenci traheide; e) tangentni presjek koji prikazuje male jažice na tangentnim stijenkama traheida

Table 2 ANOVA analysis and Duncan test groups for density, bending strength, and compressive strength values under different relative humidity conditions
Tablica 2. ANOVA analiza i grupe Duncanova testa za vrijednosti gustoće, čvrstoće na savijanje i *čvrstoće na tlak* pri različitim uvjetima relativne vlažnosti zraka

Tests *Ispitivanje*	**Relative humidity** *Relativna vlažnost zraka*	**New sample Mean ± SD** *Svježe posječeno drvo; srednja vrijednost + standardna devijacija*	**Duncan (new)** *Duncanov test (novi)*	**Old sample Mean ± SD** *Ostarjelo drvo; srednja vrijednost + standardna devijacija*	**Duncan (old)** *Duncanov test (stari)*	***F* value (new)** *F-vrijednost (novi test)*	***p* value (new)** *p-vrijednost (novi test)*	***F* value (old)** *F-vrijednost (stari test)*	***p* value (old)** *p-vrijednost (stari test)*
Density, g/cm^3 *gustoća,* g/cm^3	30 %	0.4430 ± 0.0247	a*	0.4524 ± 0.0144	a	7 908	0.000	7 336	0.001
	65 %	0.4903 ± 0.0235	b	0.4575 ± 0.0127	ab				
	85 %	0.4530 ± 0.0221	a	0.4696 ± 0.0143	bc				
	95 %	0.4614 ± 0.0211	a	0.4785 ± 0.0137	c				
Compression strength, N/mm^2 *čvrstoća na tlak,* N/mm^2	30 %	50.64 ± 4.09	d	52.33 ± 1.27	d	143 939	0.000	263 433	0.000
	65 %	44.39 ± 3.74	c	36.66 ± 2.07	c				
	85 %	32.04 ± 2.87	b	31.98 ± 1.69	b				
	95 %	20.49 ± 0.92	a	25.45 ± 2.67	a				
Bending strength, N/mm^2 *čvrstoća na savijanje,* N/mm^2	30 %	86.10 ± 8.20	b	90.10 ± 8.55	d	23 153	0.000	29 445	0.000
	65 %	82.18 ± 6.35	b	80.24 ± 7.04	c				
	85 %	57.44 ± 7.34	a	64.86 ± 7.37	b				
	95 %	55.98 ± 7.60	a	49.50 ± 6.16	a				

*Groups with the same letter do not show a statistically significant difference between them ($p < 0.05$). SD – Standard deviation. / *Među grupama označenim istim slovom nema statistički značajne razlike (p < 0,05). SD – standardna devijacija.*

(2024) noted a tendency for density reduction in naturally aged Scots pine (*Pinus sylvestris L.*) and unbarked oak (*Quercus petraea L.*), but an increase in density was observed when these woods were treated with tannin. When examining the graph in Figure 6, the interquartile range (IQR) of the naturally aged fir samples was narrower, and their density distribution was more homogeneous. In contrast, the density distribution of new solid fir samples was wider. This suggests that with aging, the density values of fir wood in different samples tend to converge. Furthermore, when looking at the graph as a whole, it is evident that the density of the naturally aged fir samples significantly increased with the rise in relative humidity. This result was more inconsistent for the new solid fir samples.

Figure 7 displays the bending strength behavior of naturally aged and freshly cut fir samples under different relative humidity conditions on a whisker plot graph. With the increase in relative humidity, both the naturally aged fir samples and the freshly cut fir samples exhibited a decrease in bending strength. This can be explained by the increase in the water content of the wood up to the fiber saturation point, leading to a reduction in bending resistance (Küch, 1943; cited in Berkel, 1970). The trend of decreasing bending strength median values with increasing relative humidity for both groups indicates that the mechanical properties of fir samples are adversely affected by increased relative humidity. Similarly, ANOVA analysis revealed significant differences in bending strength for both the new fir ($F = 23.153$, $p < 0.001$) and naturally aged fir samples ($F = 29.445$, $p < 0.001$). For new fir samples, bending strength was measured as 86.10 N/mm^2 at 30 % relative humidity, dropping to 55.98 N/mm^2 at 95 % relative humidity. Similarly, for the naturally aged fir samples, the bending strength at 30 % relative humidity was 90.10 N/mm^2, which decreased to 49.50 N/mm^2 at 95 % relative humidity. The naturally aged fir wood generally exhibited higher bending strength compared to newly cut fir wood, but it was found to be more sensitive to increases in ambient relative humidity (Table 2). Furthermore, as seen in Figure 7, the median bending strength for the naturally aged fir samples was generally higher than that of the newly cut fir samples under different relative humidity conditions. The only exception was at 65 % relative humidity, where the bending strength of both the naturally aged fir and newly cut fir samples was similar, and at 95 % relative humidity, where the naturally aged fir sample had lower strength. The higher bending strength of the naturally aged fir samples under varying relative humidity conditions indicates that aging strengthens the bending strength of fir wood. However, Kranitz *et al.* (2016) reported the opposite result, stating that aging weakens the flexibility and strength properties of wood, especially reducing the plastic deformation capacity. In a study by Sonderegger *et al.* (2015), the bending strength of naturally aged spruce trees (120-150 years old) was higher than that of new spruce trees. However, the bending strength of fir samples aged 120-150 years was lower than that of newly cut fir. This contradictory result in the historical fir wood was emphasized by Sonderegger *et al.* (2015), who argued that the key factor influencing the bending strength was the wood density, rather than the tree age.

Another notable aspect in the graph is that, although the trend of bending strength reduction due to the increase in ambient relative humidity is similar for both groups, the loss of bending strength at each relative humidity increase stage is more pronounced in the naturally aged fir wood samples. In particular, at 95 % relative humidity, the reduction in bending strength loss is notably higher for the naturally aged fir wood samples. This result can be explained by the deterioration and aging of wood due to various environmental (weathering) or biological factors (fungi, bacteria, insects, marine borers) (Unger *et al.*, 2001). Specifically, the mechanical strength loss in deteriorated and damaged wood is particularly attributed to the degradation of hemicellulose and the loss of sugars in hemicellulose (Nilsson and Rowell, 2012).

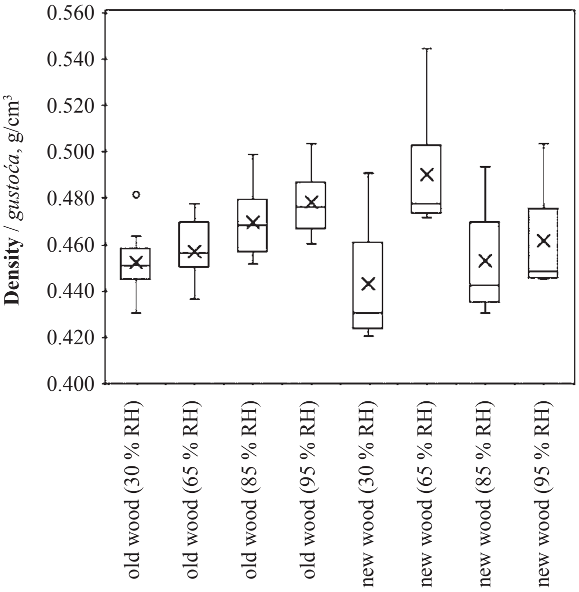

Figure 6 Whisker box plot graph of density values of naturally aged fir wood and new fir wood at different relative humidity levels
Slika 6. Kutijasti dijagram vrijednosti gustoće uzoraka prirodno ostarjele i svježe posječene jelovine pri različitim relativnim vlažnostima zraka

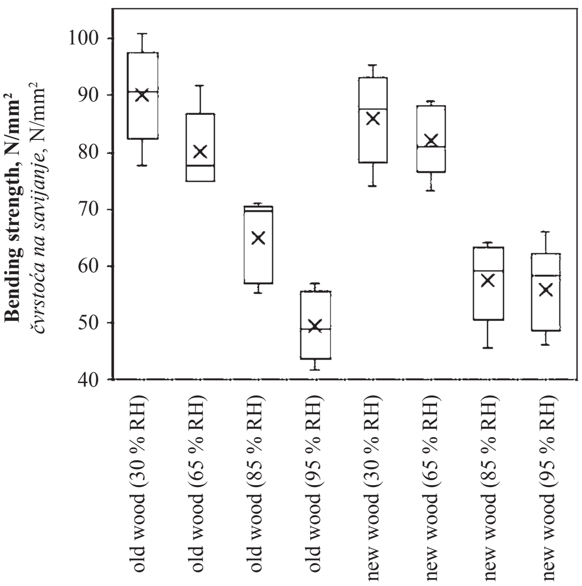

Figure 7 Whisker box plot graph of bending strength values of naturally aged fir wood and new fir wood at different relative humidity levels
Slika 7. Kutijasti dijagram vrijednosti čvrstoće na savijanje uzoraka prirodno ostarjele i svježe posječene jelovine pri različitim relativnim vlažnostima zraka

Figure 8 shows the whisker plot graph of the compression strength of naturally aged and new fir wood samples under different environmental relative humidity conditions. In general, as the relative humidity increased, both the naturally aged fir samples and the new solid fir samples exhibited a decrease in compression strength. High environmental relative humidity negatively affected the compression strength of fir samples in both groups, leading to a decline. Similarly, ANOVA analysis revealed statistically significant differences in compression strength between both new ($F = 143.939$, $p < 0.001$) and aged ($F = 263.433$, $p < 0.001$) samples, depending on the relative humidity level (Table 2). In both groups, as the relative humidity increased, a noticeable decrease in compression strength was observed. For new solid fir samples, the compression strength at 30 % relative humidity was 50.64 N/mm², whereas at 95 % relative humidity, it decreased to 20.49 N/mm². Similarly, for the naturally aged fir samples, the compression strength was 52.32 N/mm² at 30 % relative humidity, dropping to 25.45 N/mm² at 95 % relative humidity. This indicates a reduction in mechanical strength due to the water uptake of

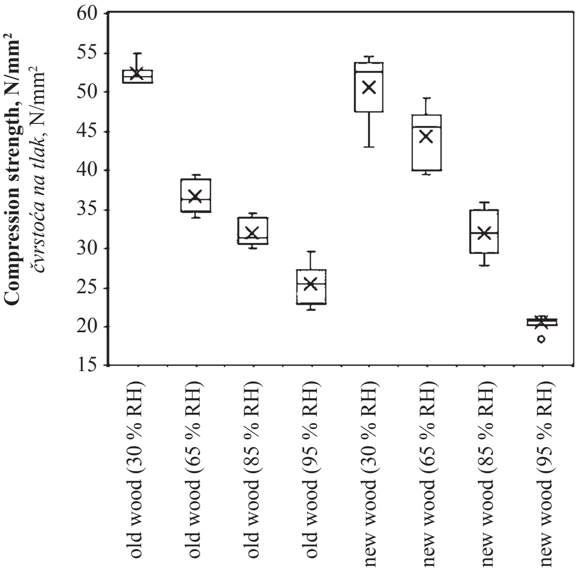

Figure 8 Whisker box plot graph of compression strength values of naturally aged fir wood and new fir wood at different relative humidity levels
Slika 8. Kutijasti dijagram vrijednosti **čvrstoće na** tlak uzoraka prirodno ostarjele i svježe posječene jelovine pri različitim relativnim vlažnostima zraka

the wood. However, when examining the graph in Figure 8, it can be observed that the interquartile range (IQR) for the naturally aged fir samples is narrower. This suggests that the compression strength values in the longitudinal direction of naturally aged fir samples are more consistent and closer to one another. Another point is that the compression strength median for naturally aged fir samples is generally lower than that of the new solid fir samples. A similar result has been reported by Xin *et al*. (2022), where aging negatively affected mechanical properties. On the other hand, Attar-Hassan (1976) observed the opposite result, indicating a 7 % increase in crushing strength and a decrease in the compression proportional limit as a result of aging. This indicates that, while the load-bearing capacity of wood increases with aging, the elastic limit decreases. In another study, Erdhardt *et al*. (1996) mentioned that aging caused only minimal changes in mechanical properties depending on the age of the tree and varying environmental relative humidity conditions. Another important point shown in the graph in Figure 8 is that for the 30 % relative humidity condition, the compression strength values for both groups are similar. However, for the 95 % relative humidity condition, the compression strength of the new fir samples was noticeably lower than that of the naturally aged fir samples. The significant decrease in the compression strength of new solid fir samples under high relative humidity conditions suggests that naturally aged wood is less affected by high relative humidity in terms of compression strength.

4 CONCLUSIONS
4. ZAKLJUČAK

The results obtained in this study can be summarized as follows:

With the increase in relative humidity, the density of the naturally aged fir samples showed a significant increase. This result was more inconsistent for the new solid fir samples.

As the relative humidity increased, both the naturally aged fir wood samples and the new solid fir wood samples exhibited a tendency for a decrease in bending strength. However, the highest bending strength was obtained for both experimental groups at the relative humidity of 30 %.

With the increase in relative humidity, both the naturally aged fir samples and the new fir samples showed a tendency for a decrease in compression strength. At high relative humidity (95 % RH), the compression strength of both groups was negatively affected, resulting in the lowest compression strength values.

Statistically significant differences in terms of density, bending, and compression strengths were found based on different relative humidity levels through ANOVA analysis. It was determined that the naturally aged fir wood was more stable in terms of density, while the loss of strength became more pronounced under high relative humidity conditions.

In conclusion, when evaluated under different relative humidity conditions, the fir wood subjected to natural aging generally demonstrated superior performance in terms of density and bending strength compared to the new fir wood. However, in terms of compression strength, it was determined that the naturally aged fir wood exhibited lower strength than the new fir wood across different relative humidity levels.

In general, the natural aging process causes significant and noticeable changes in the physical and mechanical properties of wood materials. To more comprehensively and thoroughly examine these changes, it is considered essential to take into account environmental and mechanical factors, such as temperature fluctuations, humidity conditions, and load history, as highlighted by Froıdevaux *et al*. (2012). In this context, understanding the natural aging process of wood, commonly used in traditional historical buildings, is of great importance for the preservation and maintenance of wooden cultural heritage. In this study, evaluations were conducted solely on fir trees (*Abies* sp.) grown in Ankara, Turkey; however, conducting similar studies on various tree species from different regions is believed to broaden the scope of the findings.

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Analysis of Heat-Treated Taurus Cedar *(Cedrus libani)* Wood Surface Properties with Fourier Transform Infrared (FTIR) Spectroscopy and Contact Angle Measurement

Analiza svojstava površine toplinski obrađenog drva libanonskog cedra (*Cedrus libani*) uz pomoć Fourierove infracrvene spektroskopije (FTIR) i mjerenja kontaktnog kuta

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ABSTRACT • *This study examined the changes in the chemical properties of heat-treated cedar (Cedrus libani) wood as a function of four different treatment temperatures (120, 150, 180 and 210 °C) and three different treatment durations (2, 5 and 8 hours). Particular emphasis was placed on the changes observed in the 1740 cm^{-1} carbonyl absorption band in the FTIR spectra of the esterification reaction products, which occurred at 120, 180 and 210 °C for 2, 5 and 8 hours. Surface energies of the samples were also calculated using Young's equation and contact angle measurements with three different liquids: water, glycerol, and hexane. Subsequently, the surface free energies were analyzed using the Lifshitz-van der Waals/acid-base (LW-AB) approach. FTIR analysis indicated that esterification reactions occurred more intensely at temperatures between 180 °C and 210 °C. Contact angle measurements revealed a decrease in polarity and a corresponding increase in non-polarity with increasing heat treatment temperature and duration. Free surface energy analysis demonstrated an increase in the LW component in cedar wood samples treated at 120, 180 and 210 °C for all treatment durations (2, 5 and 8 hours), indicating that surface modification had occurred. These results suggest an increase in nonpolar effects within the surface energy components of the samples, resulting in increased hydrophobicity.*

KEYWORDS: *heat treatment; FTIR; contact angle measurement; cedrus libani; surface energies*

SAŽETAK • *U ovoj studiji predstavljeno je istraživanje promjena kemijskih svojstava toplinski obrađenog drva libanonskog cedra (Cedrus libani) pri četiri različite temperature (120, 150, 180 i 210 °C) i tri različita trajanja obrade (dva, pet i osam sati). Poseban je naglasak stavljen na promjene karbonilne apsorpcijske vrpce na 1740*

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cm^{-1} u FTIR spektrima produkata reakcije esterifikacije pri temperaturi obrade 120, 180 i 210 °C tijekom dva, pet i osam sati. Također, izračunane su površinske energije uzoraka uz pomoć Youngove jednadžbe i mjerenja kontaktnog kuta trima različitim tekućinama: vodom, glicerolom i heksanom. Nakon toga, slobodne su površinske energije analizirane primjenom Lifshitz-van der Waals/acid-base (LW-AB) pristupa. FTIR analiza pokazala je da su reakcije esterifikacije intenzivnije pri temperaturama između 180 i 210 °C. Mjerenjima kontaktnog kuta otkriveno je da se s povećanjem temperature i trajanjem toplinske obrade smanjuje polarnost i povećava nepolarnost uzoraka. Analiza slobodne površinske energije pokazala je povećanje LW komponente na uzorcima cedrovine obrađenima na 120, 180 i 210 °C za sva tri trajanja obrade (dva, pet i osam sati), što upućuje na to da je nastala modifikacija površine. Ti rezultati potvrđuju povećanje nepolarnih učinaka unutar komponenata površinske energije uzoraka, što rezultira povećanom hidrofobnošću toplinski obrađenog drva.

KLJUČNE RIJEČI: *toplinska obrada; FTIR; mjerenje kontaktnog kuta; libanonski cedar; površinska energija*

1 INTRODUCTION
1. UVOD

Wood, a sustainable material, has been an important raw material for centuries and has been used in many different fields. It is widely used in construction and decoration, especially due to its high strength properties compared to its weight. However, some disadvantageous properties of wood have limited its use in certain applications. Researchers have developed various methods to eliminate or minimize these limitations. Among these methods, chemical treatment, heat treatment and impregnation are the most important ones (Kocaefe *et al*., 2007; Hill, 2011; Bal, 2013; Sivrikaya *et al*., 2015a; Svenson *et al*., 2023).

The processes carried out to change or improve the disadvantages of wood material such as its sensitivity to moisture, low levels of dimensional stability, low hardness, limited resistance to abrasion, fungi, termites and UV radiation are generally referred to as “wood modification”. The modification process causes permanent changes in the chemical structure of wood (Altınok *et al*., 2010; Sandberg *et al*., 2017; He *et al*., 2023). The fact that heat treatment, which is one of the modification methods, is commercially applicable and accepted as environmentally friendly makes it stand out. With heat treatment, permanent changes are created especially in the chemical structure of the polymeric structure that forms the wood, making the material more resistant to the mentioned disadvantages (Korkut *et al*., 2008; Esteves and Pereira, 2008; Korkut and Kocaefe, 2009; He *et al*., 2023).

The main objective of heat treatment is to alter the chemical structure so as to reduce hygroscopicity and improve wood properties. During this process, chemical modifications occur, including the degradation of hemicelluloses and the esterification of hydroxyl groups, which decrease the number of accessible OH groups, thereby reducing wood swelling and shrinkage (Gerardin *et al*., 2007; Kocaefe *et al*., 2008; Hill *et al*., 2021; Čabalová *et al*., 2022). As a result of heat treatment, increases in lignin content, decay resistance, and dimensional stability have been observed, while mechanical properties and water absorption capacity tend to decrease. Additionally, improvements in resistance to weathering have been reported (Bal, 2013; Tufan *et al*., 2022).

Although most of the free and bound water is lost from wood up to 150 °C, some of the water bound to the cell wall continues to remain up to higher temperatures (Gezici-Koç *et al*., 2017; Penvern *et al*., 2020; Aydın, 2021; Hill *et al*., 2021). At temperatures higher than 150 °C, heat treatment chemical processes speed up (Boonstra, 2008; Piernik *et al*., 2022). The range between 160 and 240 °C is typical for heat treatment (Militz, 2002; Sikora *et al*., 2018). According to Kamdem *et al*. (2002) and Hill (2007), the process temperature and duration vary from 180 to 280 °C and 15 to 24 hours, respectively, depending on the type of wood, sample size, moisture content, desired mechanical performance, resistance to biological factors, and desired properties like stability.

FTIR is an easy and nondestructive analytical˝tool for determining changes in the chemistry of wood structure due to modification (Chien *et al*., 2018; Peng *et al*., 2022). Because of these characteristics, its importance and areas of use have grown. FTIR analysis gives important information about the changes in the O-H, C-H, N-H, and C-O groups of cellulose structures and intra- and inter-chain hydrogen bonds. Thus, it provides more detailed information about the interactions of the polymer components of wood material at the molecular level (Cherdkeattikul *et al*., 2020; Kaya *et al*., 2021).

In the literature, heat treatment has been applied to many different types of wood. The changes that occur as a result of these processes have been investigated in detail. Most of the studies have focused on these types, especially due to the durable and aesthetic structures of cedar wood species. For example, as a result of 1 and 2 hours of heat treatment at 220 °C on western red cedar samples, it was determined that pits formed in the tracheid walls, ray parenchyma and pit aspiration of the wood material (Awoyemi and Jones, 2011). In the study conducted on eastern red cedar wood samples, a decrease in the shear and hardness values of the samples occurred as a result of the heat treatment applied at 120 °C. It was concluded that the sample surface roughness improved when the temperature increased and time was kept constant. Also, a decrease was reported in the water absorption values, swelling ratio and diffusion coefficient of samples depending on these parameters (Dilik and Hiziroglu, 2012; Kasemsiri *et al*., 2012).

Thermally treating the wood leads to substantial alterations in color, chemical composition, dimensional stability and biological resistance of the materials, depending on the treatment temperature and duration. Jimenez *et al*. (2011) found that coloration of Malapapaya (*Polyscias nodosa*) wood changed from yellowish to dark brown when processed at various temperatures. Similarly, Barcik *et al*. (2015) showed that temperature significantly facilitated the darkening of oak and Scots pine, while the darkening effect was slightly more pronounced in birch. When studying changes in chemical structure, Srinivas and Pandey (2012) reported a decrease in the content of hydroxyl groups with heat treatment of rubbberwood and silver oak which affected the hygroscopicity of the material. Likewise, Xu *et al*. (2019) also reported that the amount of hydroxyl groups decreased, while the carbonyl groups increased, with increasing temperature in white oak (*Quercus alba*).

In terms of dimensional stability, Builes *et al*. (2021) found that the dimensional stability of Pinus oocarpa (*ocote pine*) wood was increased by heat treatment as a result of the removal of hydroxyl groups. Similarly, Wang *et al*. (2018) also found a relationship between the decrease in the effective free hydroxyl group content and the decrease in the immobilized free water content, which led to a decrease in the equilibrium moisture content and thus increased the dimensional stability in Masson pine (*Pinus massoniana*). Bytner *et al*. (2021) also noted that the equilibrium moisture content and water uptake rate of black poplar (*Populus nigra*) wood were reduced by heat treatment. Similarly, Durmaz *et al*. (2019) showed that, although the compressive and flexural strengths of Scots pine (*Pinus sylvestris*) were reduced by heat treatment, the dimensional stability was significantly increased. In terms of biological durability, Altgen *et al*. (2020) reported that thermal degradation of hemicelluloses in Scots pine increased the resistance to brown rot fungi. Additionally, Vidholdova *et al*. (2022) showed that higher temperature heat treatment greatly improved the decay resistance of Norway spruce (*Picea abies*) wood against brown rot and white rot fungi.

According to some studies, the increased surface roughness formed during heat treatment in the range of 200-220 °C is the result of the combined effects of thermal stresses, hemicellulose degradation, lignin rearrangement and structural irregularities (Nabil *et al*., 2018; Hill *et al*., 2021; Wu *et al*., 2021; Mastouri *et al*., 2023). It was found that cracks occurred in the samples and the presence of these cracks decreased the compressive, flexural and tensile strengths of the heat-treated samples of Deodar cedar (*Cedrus deodara*) wood (Oliveira *et al*., 2022). According to the obtained data, its resistance against decaying of wood material was increased by applying it on Japanese cedar (*Cryptomeria japonica*). Another study showed that, when Japanese cedar (*Cryptomeria japonica*) samples were subjected to heat treatment at 20, 40, 60, 80, and 100 °C, the highest compression strength, yield stress, and Young's modulus were obtained at 20 °C, while these values decreased at 100 °C (Chang *et al*., 2019; Toba *et al*., 2024). However, the majority of these studies have focused on different cedar species, and research conducted on Lebanon Cedar/Toros Cedar (*Cedrus libani)* is quite limited.

In recent years, the production and use of heat-treated wood have increased significantly in the wood industry. This process, which is widely used in certain tree species, has been applied to cedar wood, which is widely distributed in the Mediterranean region and used for various purposes, but detailed information about the chemical changes that occur is still limited. Considering this deficiency, the chemical changes of Taurus cedar (*Cedrus libani*) wood after heat treatment were investigated. While previous studies have mainly focused on other coniferous species, especially Cedrus atlantica and Cedrus deodara, this study aims to fill this gap by investigating a coniferous species of significant regional importance. The study provides information not only on the general changes of functional groups, but also on the quantitative changes in specific bonds, such as carbonyl groups around 1740 cm^{-1}, with respect to temperature and time, as shown by FTIR measurements. Furthermore, by using surface contact angle measurements to relate these chemical changes, a comprehensive explanation of the effect of heat treatment on the hydrophilicity and hydrophobicity surface properties was provided.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

Cedar log samples were selected at 1-meter and 3-meter intervals from a cutting area under the jurisdiction of the Burdur-Bucak Forest Management Directorate. These samples were cut into 50 cm lengths and prepared from the sapwood portions. The cedar samples were air-dried and then processed to dimensions of (22 × 70 × 350) mm^3. Heat treatment was conducted at four different temperatures (120 °C, 150 °C, 180 °C, and 210 °C) and for three different durations (2 hours, 5 hours, and 8 hours). The heat treatment conditions were established taking into consideration the range of temperature and time available in the literature. The hemicellulose starts to degrade at around 140 °C and then the cellulose structure changes (Esteves *et al*., 2007; Rowell *et al*., 2009). It is known that temperatures of 240 °C and beyond can result in the deterioration of mechanical properties of wood (Hill *et al*., 2021; Adhikari *et al*., 2025). Thus, in the current study, the lowest treatment temperature was selected at 120 °C to study possible structural differences at a temperature lower than 140 °C and the highest temperature was 210 °C to prevent substantial mechanical damage.

To evaluate the effect of temperature and time relationship on the material, 2, 5 and 8 h treatment times were selected (Hill *et al*., 2007; Esteves *et al*., 2007; Esteves and Pereira, 2009; Hill *et al*., 2021; Adhikari *et al*., 2025). A 2 h time period was selected to obtain rapid improvement at high temperatures, while a 5 h time period was determined for the best improvement at medium temperatures. On the other hand, an 8 h time period was selected to monitor the early stage of chemical changes at low temperatures.

The treatment was performed using a laboratory furnace (FN 500, Nüve Co., Ankara, Turkey) capable of maintaining temperatures within ± 1 °C in an ambient air environment (Figure 1). Control samples were also prepared for each temperature group and evaluated under the same conditions. Following heat treatment, all test samples were conditioned at 65 % relative humidity and 20 °C.

2.1 FTIR analysis
2.1. FTIR analiza

FTIR spectroscopy was used to determine the chemical changes occurring in the heat-treated cedar samples compared to the control samples. With the help of this analysis, changes in the structural composition of cedar wood samples as a result of heat treatment were investigated. It was determined that the changes occurred in parallel with the disintegration of amorphous cellulose (decomposition temperature 200-240 °C) and irregular structures of hemicelluloses in the structure of the samples, resulting in an increase in the crystallinity of cellulose (Yang *et al*., 2016; Chang *et al*., 2019). In this change, since the amount of cellulose remained unchanged, a proportional increase in the crystallinity of cellulose occurred. These increases can only be explained by the decrease in amorphous regions. The effects of this change on the OH groups and cellulose crystal structures in the cedar samples were investigated in detail by FTIR spectroscopy.

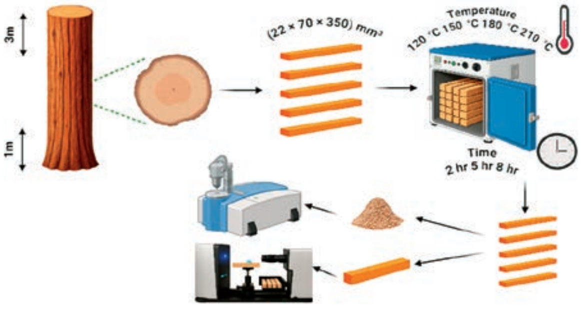


Figure 1 Heat treatment application process and performance tests
Slika 1. Postupak toplinske obrade i ispitivanja svojstava cedrovine

Functional group analysis was performed using IR Prestige-21 model FTIR spectrometer (Shimadzu Corp, Japan). For this analysis, samples were prepared as solid powder mixed with KBr and pressed into pellets. Specifically, the samples were ground with KBr in an agate mortar and subsequently pressed into tablets at 10 bar pressure. Since KBr is outside the visible region, the obtained spectra are attributed solely to the sample. Spectra were acquired by averaging 10 scans over the range of 4000-400 cm^{-1}. These spectra were then normalized at 2950 cm^{-1}.

2.2 Contact angle measurement
2.2. Mjerenje kontaktnog kuta

Contact angle measurements typically employ droplet volumes ranging from 0.5 to 10 µL to minimize gravitational deformation. Variations in droplet volume can influence measurement results due to the combined effects of gravity and droplet size. While 2-3 µL droplets are often ideal for contact angle measurements, the low water retention of superhydrophobic surfaces can hinder the deposition of droplets smaller than 4 µL. For accurate measurements, it is crucial to perform readings immediately after droplet deposition to prevent evaporation. Simultaneously, allowing a brief period for droplet spreading and interaction with the surface is also essential (Zhang *et al*., 2004; Ferrari *et al*., 2006; Tianyi and Jiang, 2018). Accordingly, contact angle measurements of the cedar samples were performed by capturing images every 5 seconds for 30 seconds after a 5 µL water droplet was placed on the surface.

In this study, mechanical cutting was applied to equalize the surface roughness of the samples. Surface free energy is theoretically defined for ideal and smooth surfaces. However, real surfaces can deviate from the ideal due to factors such as roughness, contamination, and heterogeneity. Such surface characteristics can affect the measured contact angles, leading to uncertainties reflected in surface free energy calculations (Gindl *et al*., 2001; Qin *et al*., 2015). In this context, the surface free energy values obtained in this study should be evaluated not as absolute values, but only as a tool for relative comparison between measurements performed under the same experimental conditions.

Samples were prepared using the sessile drop method on an Attention instrument in accordance with the TAPPI T558 om-97 (Canada) standard (TAPPI). Cedar samples with dimensions of (22 × 70 × 350) mm³ (tangential × radial × longitudinal) were used to measure contact angles. The contact angles of both control and heat-treated samples were measured using water, hexane, and glycerol (hexane and glycerol were supplied by Labor Teknik, Isparta, Turkey). Initially, ten drops of water were placed on each sample to determine the water contact angle. This procedure was then repeated using hexane and glycerol. Subsequently, comparisons were made between the control and heat-treated samples, and changes in surface properties and surface free energy were analyzed.

2.2.1 Young's equation

2.2.1. Youngova jednadžba

According to Young's equation (1), the relationship between contact angles and surface free energy of cedar wood samples was calculated (Gerardin *et al.*, 2007; Zhao and Jiang, 2018).

$$\cos\theta = \frac{\gamma_{\mathrm{gs}} - \gamma_{\mathrm{ls}}}{\gamma_{\mathrm{gl}}} \tag{1}$$

In the equation, cos θ represents the contact angle, γ_{gs} represents the gas-solid surface free energy, γ_{ls} represents the liquid-solid surface free energy and γ_{gl} represents the gas-liquid surface free energy.

2.2.2 Lifshitz-van der Waals and acid-base approach

2.2.2. Lifshitz-van der Waals i kiselo-bazni pristup

Lifshitz-van der Waals/acid-base (LW-AB) approach was applied using the contact angle values and surface free energy data of water, hexane and glycerol. Thanks to this approach, it was determined whether the surface free energies of cedar wood samples show polar or apolar properties. It also allowed a detailed examination of the changes in the acidic-basic components. All these analyses were carried out using Eq. 2, Eq. 3 and Eq. 4. (Gerardin *et al*., 2007; Chen *et al*., 2020).

$$\gamma_{\mathrm{sl}}^{\mathrm{LW}} = \left(\sqrt{\gamma_{\mathrm{lv}}^{\mathrm{LW}}} - \sqrt{\gamma_{\mathrm{sv}}^{\mathrm{LW}}}\right)^2 \tag{2}$$

$$\gamma_{\mathrm{sl}}^{\mathrm{AB}} = 2\left(\sqrt{\gamma_{\mathrm{sv}}^{+}\cdot\gamma_{\mathrm{sv}}^{-}} + \sqrt{\gamma_{\mathrm{lv}}^{+}\cdot\gamma_{\mathrm{lv}}^{-}} - \sqrt{\gamma_{\mathrm{sv}}^{+}\cdot\gamma_{\mathrm{lv}}^{-}} - \sqrt{\gamma_{\mathrm{sv}}^{-}\cdot\gamma_{\mathrm{lv}}^{-}}\right) \tag{3}$$

$$(1+\cos\theta)\cdot\gamma_{\mathrm{lv}} = 2\left(\sqrt{\gamma_{\mathrm{sv}}^{\mathrm{LW}}\cdot\gamma_{\mathrm{lv}}^{\mathrm{LW}}} + \sqrt{\gamma_{\mathrm{sv}}^{+}\cdot\gamma_{\mathrm{lv}}^{-}} + \sqrt{\gamma_{\mathrm{sv}}^{-}\cdot\gamma_{\mathrm{sv}}^{+}}\right) \tag{4}$$

Here, cosθ represents the cosine of the contact angle; γ^{LW} represents the Lifshitz-van der Waals surface free energy; γ^{AB} represents the acid-base surface free energy; γ_{SV} represents the solid-gas surface free energy; γ_{lv} represents the liquid-gas surface free energy; γ^{+} represents the acidic (electron acceptor) component of the surface free energy; and γ^{-} represents the basic (electron donor) component of the surface free energy.

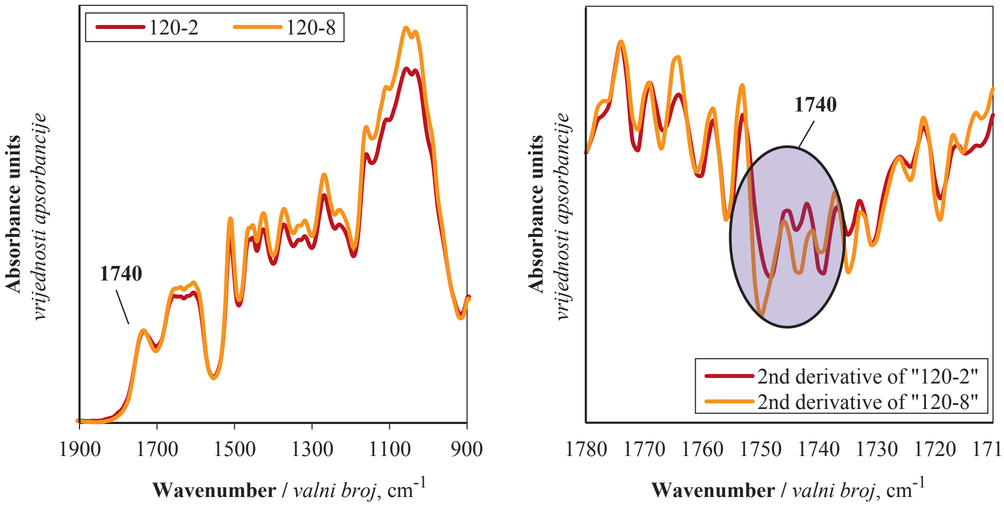
3 RESULTS AND DISCUSSION

3. REZULTATI I RASPRAVA

3.1 FTIR analysis

3.1. FTIR analiza

The wavenumber in the 1800-800 cm⁻¹ range represents the stretching and bending vibrations between molecules. FTIR examines the changes in absorption bands related to cellulose, lignin and hemicelluloses in this range (Kubovsky *et al*., 2020). In this study, FTIR analyses of cedar wood samples subjected to heat treatment at different temperatures (120 °C, 180 °C and 210 °C) and durations (2, 4, 6 and 8 hours) were per-



a) b)

Figure 2 Shows (a) FTIR spectra and (b) second derivative spectra of the 1740 cm⁻¹ absorption band for cedar samples heat-treated at 120 °C for 2 and 8 hours

Slika 2. Prikaz (a) FTIR spektara i (b) spektara druge derivacije apsorpcijske vrpce 1740 cm⁻¹ uzoraka toplinski obrađene cedrovine na 120 °C tijekom dva i osam sati formed. Depending on temperature and duration, one mole of water is removed from alcohol and ether molecules during heat treatment, leading to esterification (Tjeerdsma and Militz, 2005). To explore in detail, the changes in the ester structure that occur because of the OH groups that are eliminated from the environment as a result of the heat treatment, the second derivative of the 1740 cm^{-1} carbonyl absorption band was obtained. Additionally, the time-dependent variations in absorbance values at the wavelengths of 1600, 1500, 1460, 1420, 1373, 1318, 1267, 1160, and 1032 cm^{-1} are presented in Table 1.

As noted by Esteves *et al*. (2013) and Gaff *et al*. (2023), the FTIR is also an important technique to reveal the variations of characteristic functional groups (in particular, in the 1750-1700 cm^{-1} range, carbonyl and ester groups, ketones, aldehydes and carboxylic acids) in the composition of lignin and hemicelluloses. Additionally, the 1740 cm^{-1} absorption band sensitive to structural variation changes that occur in several modifications of different functional groups e.g., esterification, was also detected in this work (Figure 2a; Hou and Wu, 2019). In the research, the differences that resulted from heat-treated samples at 120 °C during a period of time were analyzed. This was done based on the absorbance values of 0.856 (2 hours) and 0.872 (8 hours), which were the numbers resulting from this study. These data were not found to be statistically significant. Also, these findings were verified by previous research. Tjeerdsma and Militz (2005) proved that thermal treatment of the temperatures higher than 185 °C changes the rates of the esterification processes significantly. In the study conducted by Esteves *et al.* (2013), it was stated that the 1740 cm^{-1} absorption band was minimally affected by heat treatment at 170 °C.

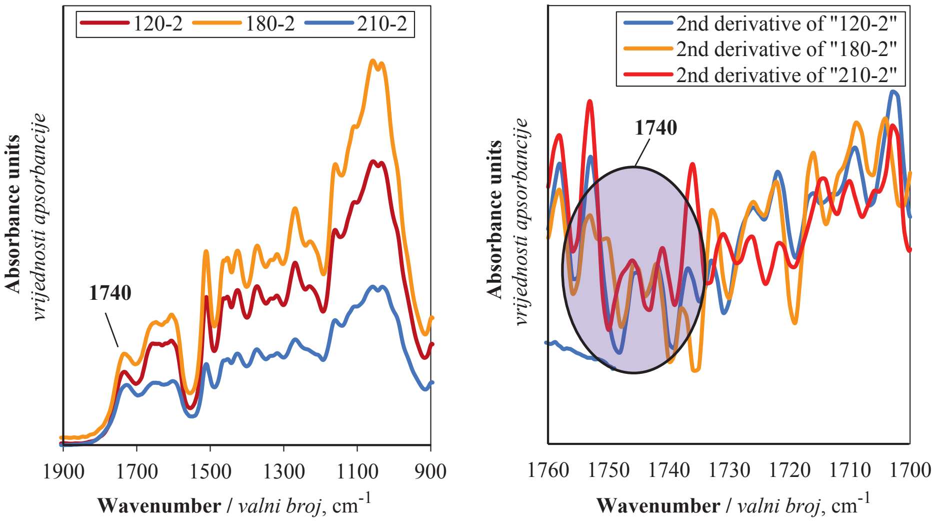
The C = C stretching vibrations in the aromatic rings in the lignin structure were attributed to the absorption band at 1600 cm^{-1}. Therefore, the indicated absorption band indicates the presence of ring structures and unsaturated bonds in that region (Gaff *et al*., 2023; Qian *et al*., 2024). The analysis revealed the intensity of the absorption band is slightly rising as the duration of treatment of constant temperature increases. Factors of this effect include the increase in the production of conjugated carbonyl groups, condensed lignin processes, and carboxylation of polysaccharides (González-Peña *et al*., 2009, Kubovsky *et al*., 2020, Li *et al*., 2024).

The absorption band at 1500 cm^{-1} corresponds to the stretching vibrations of the benzene ring and the C=C vibrations of the aromatic skeleton characteristic of the lignin structure (Kubovsky *et al*., 2020). At constant temperature, an increase in absorption values occurred due to the increase in heat treatment time. The increases are due to improvements in the chemical structure of the wood material. The increase in lignin content occurred due to the degradation of amorphous carbohydrates in hemicellulose structures and structural changes in the guaiacyl and syringyl units of lignin in wood structures (Li *et al*., 2015). Also, in another study, C=C aromatic stretching vibrations in the lignin structure were reported to be responsible for the 1505 cm^{-1} region. Furthermore, the study authors speculated that the growth occurred here due to the loss of extractive compounds and carbohydrates from the structure more than because of this wavelength (Moosavinejad *et al*., 2019).

Table 1 Variation of FTIR spectroscopy absorbance values with wavenumber (cm^{-1}) under different temperature and time conditions

Tablica 1. Varijacije apsorbancije FTIR spektroskopije s valnim brojem (cm^{-1}) pri različitim temperaturnim i vremenskim uvjetima

Temperature *Temperatura*	**Wavenumber** / V*alni broj,* cm^{-1}								
	1600	1500	1460	1420	1373	1318	1267	1160	1032
120 °C	2 hour 1.225	2 hour 1.379	2 hour 1.752	2 hour 1.809	2 hour 1.897	2 hour 1.788	2 hour 2.175	2 hour 2.574	2 hour 3.360
	8 hour 1.317	8 hour 1.495	8 hour 1.865	8 hour 1.956	8 hour 2.048	8 hour 1.938	8 hour 2.374	8 hour 2.836	8 hour 3.728
120 °C	2 hour 1.225	2 hour 1.379	2 hour 1.752	2 hour 1.809	2 hour 1.897	2 hour 1.788	2 hour 2.175	2 hour 2.574	2 hour 3.360
180 °C	2 hour 1.521	2 hour 1.786	2 hour 2.203	2 hour 2.265	2 hour 2.382	2 hour 2.283	2 hour 2.823	2 hour 3.359	2 hour 3.397
210 °C	2 hour 0.764	2 hour 0.803	2 hour 1.013	2 hour 1.073	2 hour 1.141	2 hour 1.099	2 hour 1.262	2 hour 1.493	2 hour 1.503
120 °C	8 hour 1.317	8 hour 1.495	8 hour 1.865	8 hour 1.956	8 hour 2.048	8 hour 1.938	8 hour 2.374	8 hour 2.836	8 hour 3.728
180 °C	8 hour 1.521	8 hour 1.646	8 hour 2.049	8 hour 2.120	8 hour 2.270	8 hour 2.222	8 hour 2.697	8 hour 3.266	8 hour 4.412
210 °C	8 hour 0.887	8 hour 0.829	8 hour 1.085	8 hour 1.135	8 hour 1.246	8 hour 1.231	8 hour 1.378	8 hour 1.654	8 hour 2.137



a) b)

Figure 3 Shows (a) FTIR spectra of cedar wood samples heat-treated at 120, 180, and 210 °C for 2 hours, and (b) the second derivatives of the 1740 cm^{-1} absorption band of the same samples

Slika 3. Prikaz (a) FTIR spektara toplinski obrađenih uzoraka cedrovine na 120, 180 i 210 °C tijekom dva sata i (b) drugih derivacija apsorpcijske vrpce 1740 cm^{-1} istih uzoraka

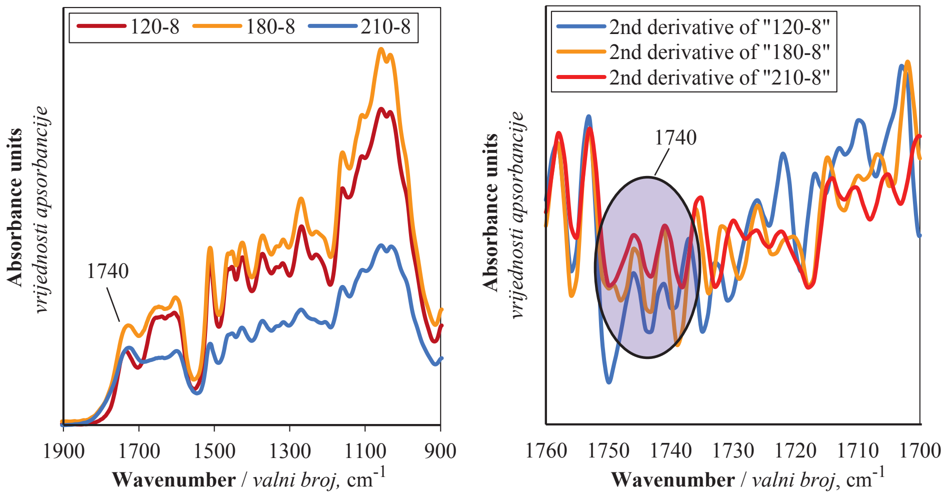
The 1460 cm^{-1} absorption band corresponds to the asymmetric C-H deformation vibrations in the lignin matrix (Piernik *et al*., 2022; Godinho *et al*., 2024). Each vibrational interaction leads to the formation of the 1420 cm^{-1} absorption band, for which the increase in absorbance intensity after exposure to heat treatment is observed. The increase in intensity of the 1420 cm^{-1} absorption band can be interpreted to be a result of the enhancement of C-H deformation vibrations in lignin and carbohydrate constituents, while the contribution of an aromatic skeletal vibrations system in lignin structure may lessen due to decomposition. The intensity of this effect has been linked to the duration of thermal treatment (Kacik *et al*., 2016; Jaruwat *et al*., 2018). Moreover, some of the observed spectral changes have been ascribed to chemical processes, including the deactivation of the methoxyl structure and the degradation of the lignin structure (Kubovsky *et al*., 2020).

The CH_2 bending vibrations of cellulose and hemicellulose structures have been linked to the absorption band observed at 1371 cm^{-1} (Caputo *et al*., 2021; Liao *et al*., 2024). In comparing the two cedar wood samples used in the study, it was observed that the intensity of absorption of the absorption band increased with increasing duration of heat treatment. In the 1318 cm^{-1} absorption band, similar increases were seen with extended processing time. Based on these results, higher density structures were formed due to an increase in the material's density (Kubovsky *et al*., 2020).

The C−O stretching vibrations in the guaiacyl ring have been attributed to the absorption band at 1267 cm^{-1}. It has been reported in the literature that for heat-treated wood samples, an increase in absorbed energy values occurs due to an increase in the treatment time at constant temperature. This increase indicates that, unlike for syringyl structures, degradation can take place at lower process temperatures and that the time of heat treatment can also affect this degradation (Kubovsky *et al*., 2020; Varga *et al*., 2020).

The absorption band at 1161 cm^{-1} was attributed to the C-O-C stretching vibrations of cellulose and hemicellulose structures. (Caputo *et al*., 2021). This absorption band has been observed to change markedly with increasing time, particularly for different durations of heat treatment. The increase in absorbance values indicates the occurrence of dehydration processes and the slow degradation of carbohydrate components. Intermolecular or cross-links are made that facilitate the formation of covalent bonds between molecules by these processes to form ether bonds. According to Kubovský and Kačík (2014) and Kačík *et al*. (2016), the absorption band at 1030 cm^{-1} can be attributed to C-O-C stretching vibrations for the primary alcohol functional groups found in the structure of cellulose and hemicellulose, as well as C-O deformation vibrations for the primary alcohol part of cellulose. A gradual change has occurred in the structure of cellulose, one of the most important components of wood material, according to the observed increases in this absorption band (Kubovsky *et al*., 2020; Dassanayake *et al*., 2023).

Analysis of the 1740 cm^{-1} absorption band absorbance values presented in Figure 3a (120 °C-2 hours → 0.856, 180 °C-2 hours → 1.063, 210 °C-2 hours → 0.677) demonstrates a significant variation observed after 2 hours of treatment at 210 °C. This observed change could be attributed to the conjugation of carbonyl groups with other double bonds, such as those found in aromatic or alkene structures, as well as the



a) b)

Figure 4 Shows FTIR spectra (a) of cedar samples heat-treated at 120, 180, and 210 °C for 8 hours, and (b) the second derivative FTIR spectra of the same samples at the 1740 cm^{-1} absorption band
Slika 4. Prikaz (a) FTIR spektara toplinski obrađenih uzoraka cedrovine na 120, 180 i 210 °C tijekom osam sati i (b) drugih derivacija apsorpcijske vrpce 1740 cm^{-1} istih uzoraka

formation of additional carbonyl or carboxyl groups through oxidation reactions that occur during thermal decomposition processes (Kubovský *et al*., 2020). Furthermore, the existence of ester bonds in aliphatic and aromatic compounds, as well as the modifications that take place in these bonds, might be linked to the alterations observed in the absorption band at 1740 cm^{-1} (Gordobil, 2018).

When the heat treatment time is kept constant, the absorption band at 1600 cm^{-1} (high peak) had the most significant change at 210 °C / 2-hour with the increase in temperature. It was also determined that the 1500 cm^{-1} absorption band underwent similar changes. The 1460 cm^{-1} and 1420 cm^{-1} absorption bands exhibited the same effect after a 2-hour heat treatment at 210 °C. These changes were attributed to the gradual decomposition of lignin components within the material's structure, as well as to the impact of high temperatures on the functional capacity of methoxyl groups (Kubovsky *et al*., 2020).

An important change in absorbance values was noticed within the 1371 cm^{-1} absorption band after the heat treatment at 210 °C for 2 hours while the intensity of the 1318 cm^{-1} absorption band increased with rising temperatures, but a sharp decrease was observed after treatment at 210 °C for 2 hours. The largest change in the 1267 cm^{-1} absorption band occurred at 210 °C / 2-hour heat treatment conditions. These observed changes collectively demonstrate the influence of elevated temperatures on structural density and degradation phenomena.

At the 1160 cm^{-1} absorption band, a substantial modification was evident following the 2-hour treatment at 210 °C. Concurrently, a marked alteration, particularly during the aforementioned 2-hour period at 210 °C, was observed within the 1030 cm^{-1} absorption band. These changes indicate gradual transformations within the cellulose matrix (Kubovsky *et al*., 2020).

The absorbance values for the 1740 cm^{-1} absorption band, as shown in Figure 4a (120 °C-8 hours → 0.872, 180 °C-8 hours → 1.139, 210 °C-8 hours → 0.872), exhibit a significant change in the second derivative spectra (Figure 4b) between 180 °C and 210 °C, over the 8-hour treatment period. While this change is attributed to the increased rate of thermal decomposition at temperatures above 180 °C, the origin of variations in the 1740 cm^{-1} absorption band is debated in the literature. One study suggests that these changes are due to hemicellulose degradation caused by heat treatment (Wang *et al*., 2024), while another attributes the increase in this absorption band to the presence of carbonyl groups (Tuncer *et al*., 2024).

Within the 1600 cm^{-1} absorption band, a marginal increase in absorbance values was evident with increasing temperature over the 8-hour duration. However, the most substantial alteration was observed at 210 °C following an 8-hour treatment. Concurrently, the most significant modification at the 1500 cm^{-1} absorption band was also found to occur at 210 °C after an 8-hour treatment period.

Consistent with the trends observed at the 1460 cm^{-1} and 1420 cm^{-1} absorption bands, the most substantial changes in absorbance within the 1371 cm^{-1}, 1318 cm^{-1}, 1267 cm^{-1}, 1160 cm^{-1}, and 1030 cm^{-1} absorption bands were likewise evident at 210 °C following an 8-hour treatment. Notably, within the 1318 cm^{-1} absorption band, a concomitant increase in intensity was also observed with increasing temperature.

A comparative analysis of the data presented in Figure 5 indicates that the heat treatment protocol main-

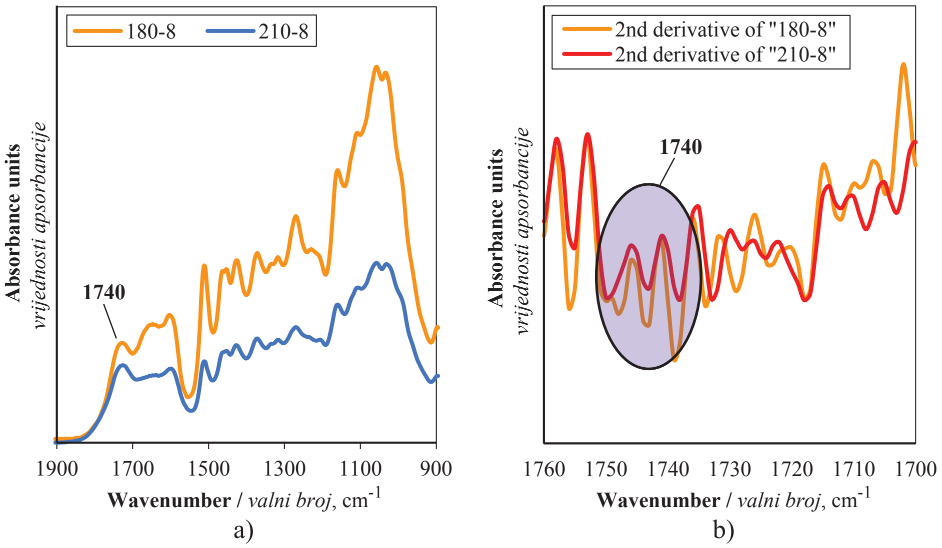


Figure 5 Shows FTIR spectra (a) of cedar samples heat-treated at 180 and 210 °C for 8 hours, and (b) the second derivative FTIR spectra of the same samples at the 1740 cm^{-1} absorption band
Slika 5. Prikaz (a) FTIR spektara toplinski obrađenih uzoraka cedrovine na 180 i 210 °C tijekom osam sati i (b) drugih derivacija apsorpcijske vrpce 1740 cm^{-1} istih uzoraka

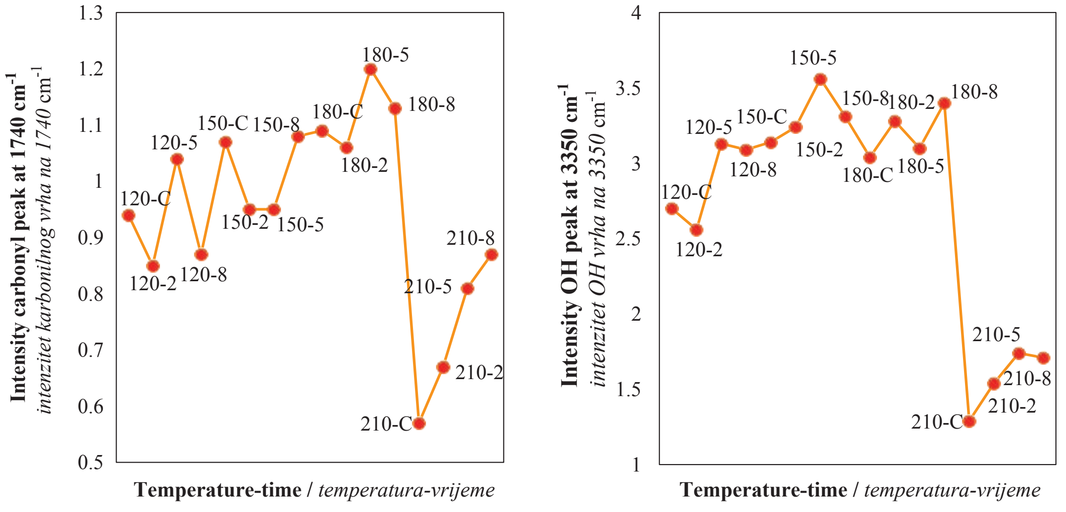
tained a constant duration while investigating the influence of increasing temperature on the spectral characteristics of various functional groups. Specifically, the most substantial modifications in both the 1740 cm^{-1} absorption band and its associated second derivative were evident during the 8-hour treatment period spanning 180 to 210 °C. Analogous variations were also observed in other functional groups (Figure 4).

According to Table 2, the heat treatment of cedar wood samples at 120 °C, 180 °C, and 210 °C resulted in various chemical changes, leading to increases and decreases in specific FTIR spectral absorption bands. Table 2 further details the underlying reasons for these observed changes at different temperatures.

As the temperature increased, the intensity of the hydroxyl groups in the 3350 cm^{-1} absorption band decreased due to esterification (Figure 6b). Particularly at 210 °C and short heating durations (2 hours), a significant decrease in the intensity of this absorption band is observed. This reduction can primarily be attributed to

Table 2 Factors influencing increases and decreases in FTIR absorption bands at specific temperatures as a result of heat treatment
Tablica 2. Čimbenici koji utječu na povećanje i smanjenje FTIR apsorpcijskih veza na određenim temperaturama kao rezultat toplinske obrade

Increases in temperatures of 120 °C and 180 °C *Povećanje na temperaturama 120 i 180* °C (Zhu *et al*., 2016; Zhang *et al*., 2017; Li *et al*., 2017; Sikora *et al*., 2018; Aytin *et al*., 2022; Zhang *et al*., 2023; Lyu *et al*., 2024)	**Reductions in temperatures of 180 °C and 210 °C** *Smanjenje na temperaturama 180 i 210* °C (Esteves and Pereira, 2009; Wang *et al*., 2016; Candelier *et al*., 2016; Výbohová *et al*., 2018; Kubovský *et al*., 2020; Piernik *et al*., 2022; De Ligne *et al*., 2024)
As the material becomes more crystalline, certain vibrational modes may become more regular, leading to stronger absorption. *Kako materijal postaje kristalniji, određeni vibracijski modovi mogu postati pravilniji, što dovodi do jače apsorpcije.*	The degradation of lignin, hemicellulose, and cellulose can lead to material loss, resulting in a decrease in the intensity of the functional groups responsible for the observed peaks. *Razgradnja lignina, hemiceluloze i celuloze može dovesti do gubitka materijala, što rezultira smanjenjem intenziteta funkcionalnih skupina odgovornih za uočene vrhove.*
Oxidation reactions or other chemical changes can result in the formation of new light-absorbing functional groups in the analyzed region. *Oksidacijske reakcije ili druge kemijske promjene mogu rezultirati stvaranjem novih funkcionalnih skupina koje apsorbiraju svjetlost u analiziranom području.*	Some degradation products may be volatile and migrate away from the sample, contributing to further attenuation of the signal. *Neki produkti razgradnje mogu biti hlapljivi i udaljiti se od uzorka, pridonoseći daljnjem slabljenju signala.*
The removal of water molecules as the material dries may enable other functional groups to produce more prominent signals. *Uklanjanje molekula vode dok se materijal suši može omogućiti drugim funkcionalnim skupinama da proizvode jače signale.*	At high temperatures, the material may undergo charring, resulting in major spectral changes. *Na visokim temperaturama materijal se može pougljeniti, što rezultira velikim spektralnim promjenama.*



a) b)

Figure 6 a) Carbonyl group intensity of the 1740 cm^{-1} absorption band of cedar samples subjected to heat treatment at 120 150-180-210 °C for 2-5-8 hours b) Hydroxyl group intensity of the 3350 cm^{-1} absorption band of cedar samples subjected to heat treatment at 120-150-180-210 °C for 2-5-8 hour

Slika 6. a) Intenzitet karbonilne skupine apsorpcijske veze na 1740 cm^{-1} toplinski obrađenih uzoraka cedrovine na 120-150-180-210 °C tijekom 2-5-8 sati, b) intenzitet hidroksilne skupine apsorpcijske veze na 3350 cm^{-1} toplinski obrađenih uzoraka cedrovine na 120-150-180-210 °C tijekom 2-5-8 sati

esterification reactions. After esterification, the intensity of the carbonyl peak (Figure 6 a) increased. The highest increase was observed at 210 °C. Consequently, it was determined that the esterification reaction occurred more in the same samples of cedar wood at low temperatures due to the number of OH groups. The decrease in the intensity of accessible hydroxyl groups (Figure 6b) can be attributed to the formation of pre-existing cross-linking or the recrystallization of cellulose, which were determined to occur during heat treatment (Tjeerdsma and Militz, 2005). Esterification causes a decrease in the intensity of hydroxyl groups in the wood, resulting in a reduction in the intensity of the hydroxyl peak at 3350 cm^{-1} (Herrera *et al*., 2014). In another study, the observed decrease in peak intensity, indicative of hydroxyl (O-H) group stretching, was attributed to hemicellulose degradation (Haseli *et al*., 2024).

However, as the heating duration increases, this effect reverses, and either the decrease in hydroxyl absorption band intensity ceases or an increase is observed. This phenomenon can be explained by the depletion of reactants necessary for esterification and, more importantly, the formation of new hydroxyl groups due to the degradation of wood components (lignin, hemicellulose, and cellulose) (Esteves and Pereira, 2009; Brebu and Vasile, 2010; Výbohová *et al*., 2018; Taghiyari *et al*., 2020). While lignin degradation releases phenolic hydroxyl groups, carbohydrate degradation can also generate new hydroxyl groups. Additionally, changes in hydrogen bonding may influence the absorption band intensity. Under prolonged heating or at excessively high temperatures, charring may occur, leading to significant spectral changes (Boonstra and Tjeerdsma, 2006; Kim *et al*., 2014).

3.2 Contact angle measurement
3.2. Mjerenje kontaktnog kuta

In this research, some of the cedar wood samples were heat-treated, while others were not. Subsequently, contact angle measurements were conducted, and the properties of cedar wood samples with and without heat treatment were compared. These comparison results are shown in Figure 7.

When the contact angle θ is greater than or equal to 90°, the liquid tends to "ball up" on the surface, indicating poor wetting or low wettability and water-repellent properties. Conversely, when θ is less than 90°, the wettability of the surface increases (Friedrich, 2021; Krainer and Hirn, 2021). To investigate the effects of modification on cedar wood samples, contact angle measurements were performed using polar (water and glycerol) and non-polar (hexane) liquids. As shown in Figure 7, heat treatment decreased wettability with polar liquids but increased it with the non-polar liquid.

FTIR analyses provided a crucial foundation for understanding the chemical changes occurring in wood, as well as their effects on surface properties. Chemical transformations in lignin and hemicelluloses at high temperatures during heat treatment reduced the surface wettability of cedar wood, imparting an important property that can increase the material's water resistance. These changes are believed to result from processes such as the growth of cross-links between lignin and polysaccharides and the reduction of polar groups in the

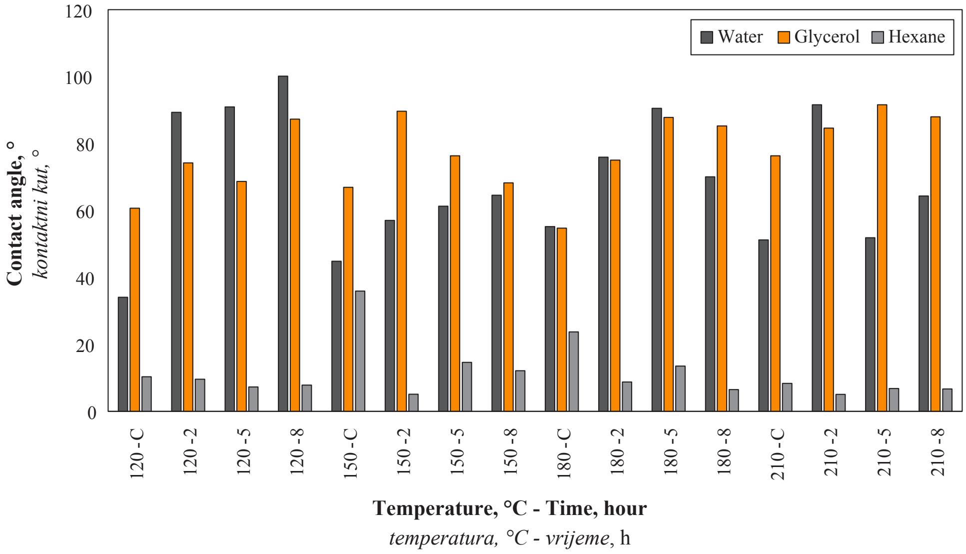


Figure 7 Contact angles of cedar wood samples after heat treatment depending on temperature and time
Slika 7. Kontaktni kutovi uzoraka cedrovine nakon toplinske obrade ovisno o temperaturi i vremenu

structure due to esterification reactions on the material's surface. The surface structures of wood materials differ from each other before and after heat treatment. This makes the differentiation of thermally treated/untreated samples very critical. In general, wood material is less wettable in polar solvents and more wettable in non-polar solvents after a heat treatment. Consequently, FTIR analyses have allowed for a general perspective for assessing the performance characteristics of cedar wood samples following the heat treatment, thus helping us to understand the polarity modifications emerging by the heat process (Gerardin *et al*., 2007; Budhe *et al*., 2020; Lovaglio *et al*., 2022).

Table 3 Free energy components on the surfaces of heat-treated cedar wood samples at different temperatures and times
Tablica 3. Komponente slobodne površinske energije toplinski obrađenih uzoraka cedrovine pri različitim temperaturama i vremenima

Cedar wood samples *Uzorci cedrovine*	γ^{LW}	γ^{AB}	γ^{+}	γ^{-}	γ_{tot}
120 °C-control	0.22	(-)4.28	(-)1.08	4.25	4.06
120 °C 2 hour	(-)1.94	9.84	7.32	3.31	7.90
120 °C 5 hour	(-)2.34	9.60	8.44	2.73	7,26
120 °C 8 hour	(-)2.64	11.60	8.32	4.04	8,96
150 °C-control	3.83	(-)8.60	(-)3.88	4.77	(-)4.77
150 °C 2 hour	(-)1.51	5.25	1.18	5.86	3.74
150 °C 5 hour	(-)0.13	5.32	1.52	4.66	5.19
150 °C 8 hour	(-)0.62	6.95	3.19	3.78	6.33
180 °C-control	1.57	3.32	0.89	3.11	4,89
180 °C 2 hour	(-)1.59	8.89	5.09	3.89	7.3
180 °C 5 hour	(-)1.39	10.18	5.71	4.54	8.79
180 °C 8 hour	(-)1.75	8.42	3.56	4.99	6.67
210 °C-control	(-)0.75	4.00	0.81	4.95	3.25
210 °C 2 hour	(-)2.76	11.09	7.53	4.09	8.33
210 °C 5 hour	(-)1.05	1.94	(-)0.15	6.26	0.89
210 °C 8 hour	(-)1.52	7.10	2.31	5.46	5.58

These parameters were obtained from the contact angles of glycerol, hexane and water on cedar wood surfaces after heat treatment. The free surface energy values of the samples of polar-nonpolar and acidic-basic wood components were determined using these data according to the Lifshitz-van der Waals/acid-base (LW-AB) method (Mohammed-Ziegler *et al*., 2004; Gerardin *et al*., 2007; Qin *et al*., 2014).

According to other researchers, negative surface free energy values accompanied by high contact angles can occur due to the surface roughness of heat-treated wood samples, their chemically heterogeneous structure, and the limited penetration of the liquids used (Mohammed-Ziegler *et al*., 2004). Microscopic surface roughness can affect the determination of the contact angle due to the uncertainty of the real contact line, and the asymmetric arrangement cross asymmetrical arrangement of the multiple functional groups on the surface may lead to directional variations in the contact angle (Oss *et al*., 1988; Tadmor, 2004). Kung *et al*. (2019) have reported that topographical and chemical heterogeneities can exist at the micro and nanoscale that can induce non-ideal wetting and variability which, when not considered carefully, can be mistaken for true physical responses. Similarly, Alnoush *et al*. (2021) revealed that the wettability of calcite is distinctly affected by increasing surface roughness, which is not due to changes in the intrinsic surface energy, but to measurement artefacts. These effects are most prominent as the contact angles approach the detection limits, since small misfits in assumed droplet shape or imaging inaccuracies can lead to substantial errors in energy calculations.

Another study observed that very weak repulsive interactions led to negative values on the interface (Hwang *et al*., 2011). Furthermore, as heat treatment alters the C-O-C stretching absorption band in the cellulose structure, other alterations in the material's surface characteristics might transpire. The adhesion process may be adversely affected by the reduction or alteration of this absorption band, which is typically absorbed in the 1050-1150 nm range (Garside *et al*., 2003).

One of the factors that significantly affects the contact angle is the presence of a chemically heterogeneous structure. As the volume of liquid drops on the substrate increases and the liquid drops move on the surface, water drops can be fixed on the surface due to the change in the chemicals on the surface of the material and, as a result, contact angles can be affected. At the same time, water drops can be fixed due to energy differences in the functional groups on the surface. Due to these reasons, liquid drops dropped on the surface are prevented from overcoming energy barriers and cause inaccurate measurement of contact angles (Cansoy *et al*., 2011; Yu *et al*., 2022).

According to the values in Table 3, significant increases occurred at 150 °C and 180 °C compared to 120 °C. This change was found to be consistent with the increases in the LW component after heat treatment in the study by Gérardin *et al*. (2007). However, LW values were negative at 210 °C and other temperatures due to the prolongation of the heat treatment time. This might be caused by unexpected structural variations on the materials' surface, such as modifications to the cellulose, lignin, and hemicellulose structures. Furthermore, surface free energy may rise while surface energy and polarity fall with longer heat treatment times. The substance may become more hydrophobic as a result of strong surface binding of molecules caused by this rise in surface free energy. At the same time, as it reduces the liquid surface area, it may suggest a strong molecular connection. According to Gerardin *et al*. (2007), Israelachvili (2011), and Vagli *et al*. (2024), all of these modifications may restrict the material's ability to interact with other chemicals.

The decrease in hydroxyl groups in cedar wood samples resulted in a drop in the 3350 cm^{-1} absorption band. This decline was particularly noticeable in cedar wood samples that were heated to 210 °C for two, five, and eight hours. This temperature shift indicates that the cellulose structure has begun to break down. Furthermore, depending on the temperature, the formation of water-loving groups like carbonyl groups may result in an additional drop in the density of hydroxyl groups.

There was a discernible change in the electron acceptor ($^{+}\gamma$) component when cedar wood samples were heated to 180 °C and 210 °C (Table 3). Depending on the heat treatment used, a considerable rise in the electron acceptor (γ^{+}) component was seen in a research by Gerardin *et al*. (2007). Wood material's capacity to absorb moisture is diminished as a result of decrease of hydrophilic OH groups caused by the modification process. Since heat treatment is generally effective at temperatures above 150 °C, no significant change was detected in the samples at 120 °C. However, it was determined that changes occurred in the samples as the temperature was kept constant and the heat treatment time increased. In this case, it can be argued that the beginning of modification of the samples was observed in the heat treatments of 2, 5 and 8 hours at 180 °C, as a result of the decomposition of hemicellulose structures, and reduction of the amorphous structures (Gerardin *et al*., 2007). Also, the period of modification started to slow down at 210 °C, as the cellulose structure began to disintegrate in the cedar wood samples.

During thermal treatment, the degradation of hemicellulose and the loss of hydrophilic functional groups (e.g., hydroxyl groups) decrease the surface water affinity and lead to a decrease in the electron donor component ($^{-}\gamma$) (Gao *et al*., 2022). At the same time, lignin molecules migrate towards the surface and the structure of lignin is rearranged, creating a more polar surface. This decrease in the electron acceptor component (γ^{+}) also contributes to a decrease in the overall surface energy (Rodriguez-Fabia *et al*., 2022). The remarkably low γtot value (0.89) of the sample after 5 h of heat treatment at 210 °C clearly demonstrates the effect of these mechanisms. Due to the large-scale destruction of hemicellulose and the condensation of lignin and cellulose, a polar and inert surface was formed, which reduced the energetic interactions of the surfaces and made it more hydrophobic (Gan *et al*., 2023). It was also noted that a significant decrease in acid-base properties was observed due to the presence of functional groups associated with the remaining lignin (Li *et al*., 2014).

The electron donor (γ^{-}) component increased at 120 °C for 2 h, 5 h and 8 h, at 150 °C for 5 h and 8 h and at 210 °C for 2 h. This can be explained by the chemical changes that occur as a result of heat treatment, which change the surface free energy of the material with different components. Such an increase in the surface free energy also causes a decrease in the contact angle. Moreover, amorphous hemicellulose molecules, such as xylose, mannose and arabinose, are degraded due to heat treatment. As a result of this degradation, there is an increase in crystallinity and lignin content (Gerardin *et al*., 2007).

4 CONCLUSIONS
4. ZAKLJUČAK

Upon analysis of heat-treated Taurus cedar samples, the decomposition of acetyl groups began at 180 °C and progressed notably when the temperature reached 210 °C, leading to a decreased content of acetyl groups that was more pronounced at the latter temperature.

Surface contact angle research revealed that heat treatment increased the wettability of non-polar molecules while decreasing that of polar ones. This decrease in wettability results from the removal of hydroxyl groups during the heat treatment process. The observed increase in electron acceptor component and van der Waals forces served as additional substantiation of the material alteration. Changes in the electron donor component under certain processing conditions revealed that hydrophilic groups had been eliminated.

The thermal modified Taurus cedar (*Cedrus libani*) wood (120-150 °C for 2 and 5 hours) has acquired improved properties suitable for the internal applications (as furniture and decorative coatings), the surface hydrophobicity and the structural stability. More intensive treatment (180-210 °C for 5 and 8 hours) further enhances water repellence, thus making the material compatible with outdoor applications (cladding, garden furniture). Nevertheless, the mechanical strength can decrease with the increase of the severity of the treatment, and therefore such materials are recommended for use in non-load-bearing applications. Optimizing heat treatment parameters plays an important role in improving the properties and service life of wood products. Moreover, research involving micro- and nano-scale modification, which aims to create superhydrophobic surfaces, is important for future increasing the durability and functionality of woody materials.

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Improving Surface Properties of Wood Material Against Weathering by Using New Bio-based Epoxy Nanocomposites

Poboljšanje svojstava površine drvnog materijala izloženoga vremenskim utjecajima primjenom novih epoksidnih nanokompozitnih biopremaza

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ABSTRACT • *The present study investigates the potential of new bio-based epoxide nanocomposite coatings for wood surfaces to replace BPA (bisphenol A)-based commercial wood coatings in terms of their surface properties. This study focuses on the use of environmentally friendly and BPA-free new bio-based epoxy coatings and their nanocomposite derivatives for the wood surface. Both the resin and the curing agents are derived from natural sources. The study is original as it uses sustainable natural resources in the coatings industry and develops cost-effective and readily available systems compared to those derived from petroleum. The research is also original as it is the first to use moss oil and turpentine oil as curing agents in epoxy resin curing reactions. These two oils have similar chemical structures, and the results are comparable. In addition, the effect of nanoparticles on the surface properties of bio-based coatings was investigated. In this study, Oriental beech was used as wood. New bio-based epoxide nanocoatings were produced using tung oil-based epoxide resin (ETO) as the coating material. Additionally, hardeners such as moss oil and turpentine oil were also used. The first step in making ETO was to combine tung oil with glycidyl methacrylate. After premixing the epoxide resin with carbon nanoparticles (graphene, CNTs, and fullerenes), moss oil and turpentine oil were used as bio-based hardeners to cure the resin. The produced materials were then applied to the wooden surface as a coating, and after weathering for 3 months, the colour, gloss and surface roughness of the wood were examined. The results showed that after weathering, the ΔL* value of all test samples decreased. According to the results of the total colour change (ΔE*), the samples of epoxidized tung oil with carbon nanotubes (TEC) added gave the lowest value and were the most stable against colour change.*

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According to the gloss test results, all samples lost gloss after weathering. In addition, TEC samples showed the lowest gloss loss. After weathering, roughness parameters of all samples increased. At all three roughness (Ra, Rz and Rq) values, the samples of epoxidized tung oil (TE) showed the least roughness increase and were the most stable against surface roughness.

KEYWORDS: *bio-based coating; epoxy coating; moss oil; turpentine oil; nanocomposites; surface properties; Oriental beech wood; BPA (Bisphenol-A)*

SAŽETAK • *U studiji se istražuje potencijal novih epoksidnih nanokompozitnih biopremaza za drvne površine kao zamjena za komercijalne premaze na bazi BPA (bisfenola A), i to u smislu njihovih površinskih svojstava. Istraživanje je usmjereno na upotrebu ekološki prihvatljivih novih epoksidnih biopremaza bez BFA i njihovih nanokompozitnih derivata na površini drva. Smola i otvrdnjivač dobiveni su iz prirodnih izvora. Istraživanje je originalno u smislu uporabe održivih prirodnih resursa u industriji premaza i razvoja isplativih i lako dostupnih sustava u usporedbi s onima dobivenim iz nafte. Istraživanje je također originalno po tome što je prvo u kojemu je upotrijebljeno ulje mahovine i terpentinsko ulje kao otvrdnjivač za epoksidne smole. Ta dva ulja imaju slične kemijske strukture i njihovi su rezultati usporedivi. Osim toga, istražen je utjecaj nanočestica na površinska svojstva biopremaza. U istraživanju je kao drvni materijal rabljena kavkaska bukovina, a kao premazno sredstvo ispitani su novi epoksidni bionanopremazi proizvedeni od epoksidne smole (ETO) na bazi tungova ulja. Prvi korak u pripremi ETO-a bilo je kombiniranje tungova ulja s glicidil metakrilatom. Nakon prethodnog miješanja epoksidne smole s ugljikovim nanočesticama (grafenom, ugljikovim nanocijevima i fulerenima), ulje mahovine i terpentinsko ulje korišteni su kao biootvrdnjivači za otvrdnjivanje smole. Tako proizvedeni materijali zatim su naneseni na drvnu površinu kao premaz, a nakon izlaganja vremenskim utjecajima tijekom tri mjeseca ispitani su boja, sjaj i hrapavost površine drva. Rezultati su pokazali da se nakon izlaganja vremenskim utjecajima vrijednost ΔL* svih ispitnih uzoraka smanjila. Prema rezultatima ukupne promjene boje (ΔE*), uzorci epoksida na bazi tungova ulja s dodanim ugljikovim nanocijevima (TEC) najmanje su promijenili boju uzoraka. Prema rezultatima ispitivanja sjaja, svi su uzorci nakon izlaganja vremenskim utjecajima izgubili sjaj, a TEC uzorci pokazali su najmanji gubitak sjaja. Nakon izlaganja vremenskim utjecajima, svi su se parametri hrapavosti (Ra, Rz and Rq) svih uzoraka povećali, a uzorci s epoksidnim tungovim uljem (TE) pokazali su najmanje povećanje hrapavosti.*

KLJUČNE RIJEČI: *biopremaz; epoksidni premaz; ulje mahovine; terpentinsko ulje; nanokompoziti; svojstva površine; kavkaska bukovina; BPA (bisfenol A)*

1 INTRODUCTION
1. UVOD

Wood is obviously a carbohydrate polymer. It is an excellent construction material. In comparison to other building materials, including steel and other metals, it has superior tensile strength (modulus) per unit density. Wood is an organic material that is found in abundance and can be obtained at a relatively low cost. The quality of the end product is contingent upon the quality of the raw materials used in its production. The use of high-quality timber is predicated on the expectation of yielding end products of commensurate quality (Ali *et al.*, 1997). Despite the numerous positive attributes of wood, its composition is susceptible to adverse environmental factors that can compromise its integrity. One such disadvantage is the propensity of wood to succumb to external weathering over time. The process by which the natural elements of sunlight, water, and wind cause degradation to wooden surfaces is known as weathering. The definition of weathering has been subject to variation among scholars (Williams and Feist, 1999; Evans, 2009). Weathering is defined as the process of surface damage to wood that occurs when it is exposed to the elements (Williams and Feist, 1999). It has been established that the initial indication of weathering is a change in the colour of the wood, which is subsequently followed by the fibres in the wood becoming looser and the surface of the wood gradually eroding (Williams and Feist, 1999).

In addition to chromatic and chemical alterations, physical changes also occur in the wooden material. In the context of outdoor exposure, the surface of the wood material undergoes a transformation, becoming rough and exhibiting initial signs of cracking. This process leads to a weakening of the texture, accompanied by the occurrence of bending, distortion and sprains (Feist, 1983). It has been demonstrated that the application of wood finishing and/or treatment procedures can offer a protective barrier against the detrimental effects of external factors on wood products. The use of wood preservatives is the preferred option. As Temiz *et al.* (2007, 2014) and Stirling and Temiz (2014) observe, pentachlorophenol, creosote, chromated copper arsenate and other copper-based wood preservatives, including amine copper azole and alkaline copper quaternary, are the main wood preservatives used in the treatment process. The use of coatings represents a secondary option. The surface treatment of wood can be accomplished through a variety of methods, including coating and finishing. Two categories of finishes can be distinguished: natural finishes, including oils, water repellents and semi-transparent penetrating stains; and opaque coatings, such as paints and solid-colour stains (Temiz *et al*., 2007; Temiz *et al*., 2014; Feist and Ross, 1995). In the coatings sector, there has been a recent increase in the use of natural vegetable oils. Vegetable oil is the optimal choice due to its ready availability, affordability, environmental friendliness, and sustainability. The literature has identified a number of additional renewable components that have attracted attention, including cellulose, protein, natural oil, lignin, starch, and sugar (Ahmad *et al*., 2012; Zhang and Kessler, 2015). Chemical modification of vegetable oils is the most common method for the production of polyacids, polyalcohols and epoxy derivatives, with the latter being the most widely used.

Vegetable oils that have undergone epoxidation can be functionalized using amines, anhydrides, allylic alcohols, thiols, and unsaturated acids (α, β) (Lehnen *et al.,* 2014; Mustata *et al.,* 2016; Huang *et al.,* 2014; Manthey *et al.,* 2014; Mashouf Roudsari *et al*., 2014; Pin *et al.,* 2015; Rosu *et al.,* 2015).

Research has been conducted on the use of epoxidized vegetable oils in the coating of wood, with the aim of preserving the material from external weather conditions. The findings have been documented in the extant literature. The photostabilizing impact of 2-hydroxy-4 (2,3 epoxypropoxy)-benzophenone (HEPBP), a reactive UV absorber, as a primer for pine wood was the subject of a study by Olsson *et al* (2012). The present study examines the impact of using HEPBP in conjunction with epoxy-functionalized vegetable oil as a priming system. The colour measurements demonstrated that pre-treating wood with epoxidized soybean oil and HEPBP resulted in a reduction of colour change after 400 hours. In their study, Kabasakal *et al*. (2024) investigated the surface attributes of naturally aged Oriental beech (*Fagus orientalis* L.) covered with derivatives of nanocomposite and epoxide-amine (ETO + D230) derived from vegetable oil. The findings of the study indicated that all coated samples exhibited a reduction in surface roughness and gloss loss in comparison to the control group following weathering. Rosu *et al*. (2016) investigated the effects of consecutive treatments with succinic anhydride and epoxidized soybean oil, respectively, on colour changes while exposed to ultraviolet radiation.

In this study, the feasibility of using new bio-based epoxy coatings obtained from tung oil based-epoxide resin (ETO) with bio-based curing agents was investigated. The curing process was performed using turpentine and moss oil as the curing agents for the tung oil-based epoxide resin (ETO) for the first time. Furthermore, these new bio-based systems were prepared with nanoparticles, and their nanocomposite derivatives were also obtained. The effect of the presence of nanoparticles on the surface properties was investigated. The surface properties of Oriental beech wood, including colour, gloss, and surface roughness, were also investigated. In the present study, an attempt was made to introduce a more natural, environmentally harmless herbal coating method to the industry, as opposed to industrial epoxy coatings.

2 MATERIALS AND METHODS

2. MATERIJALI I METODE

2.1 Material and chemicals

2.1. Materijal i kemikalije

The Sigma-Aldrich Chemical Company provided tung oil (Figure 1), as well as phenothiazine, glycidylmethacrylate, and 2,4,6-tris (dimethyl aminomethyl) phenol. The Nanografi company, based in Turkey, supplied the graphene nanoplatelet (S. A: 320 m²/g, diameter: 1.5 µm, 99.9 % (multi-walled carbon nanotubes), and fullerene C60, 95 % (Figure 2).

Arifoglu, a company based in the Republic of Türkiye, supplied the crude moss oil. Nine major groups of chemical constituents were identified in the moss oil. These included: terpene/terpenoids (monoterpenic hydrocarbons, oxygenated monoterpenes, sesquiterpenic hydrocarbons, oxygenated sesquiterpenes), aliphatic hydrocarbons, aldehydes, ketones, alcohols and other miscellaneous compounds. The chemical profile revealed that the moss oil sample contained 47 different chemical constituents,

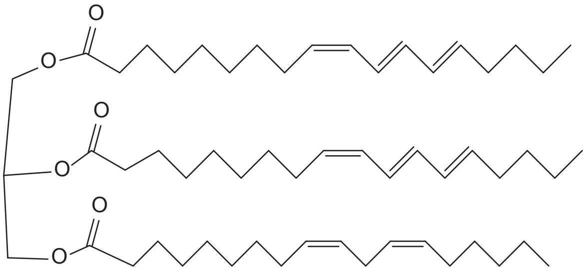

Figure 1 Structure of tung oil (Babahan-Bircan *et al.,* 2022; Kabasakal *et al*., 2023; Kabasakal *et al.,* 2024)
Slika 1. Struktura tungova ulja (Babahan-Bircan *et al.,* 2022.; Kabasakal *et al*., 2023.; Kabasakal *et al.,* 2024.)

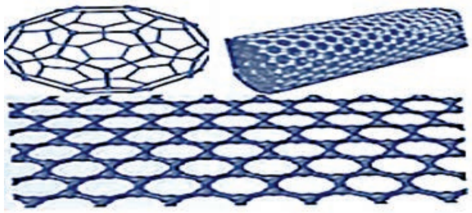

Figure 2 Structure of carbon nanoparticles
Slika 2. Struktura ugljikovih nanocijevi

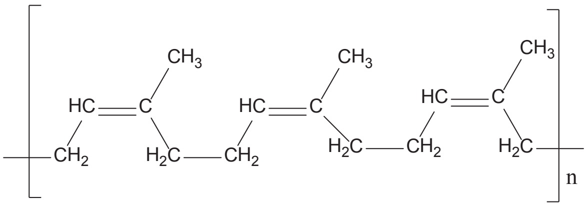

Figure 3 Structure of a sesquiterpenes (cis-polyisoprene)
Slika 3. Struktura seskviterpena (cis-poliizoprena)

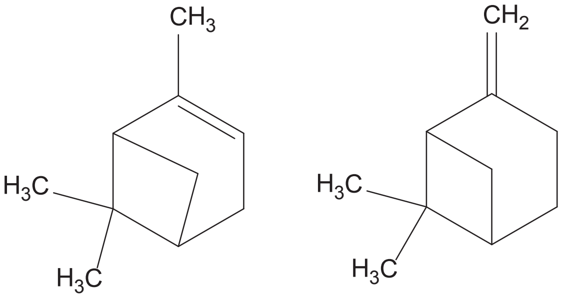

Figure 4 Chemical structure of major components of turpentine oil
Slika 4. Kemijska struktura glavnih komponenata terpentinskog ulja

which together represented 98.6 % of the total. Moss oil was identified as 15 sesquiterpene hydrocarbons (73.6 %), nine oxygenated sesquiterpenes (19.5 %), one oxygenated monoterpene (0.1 %) and one monoterpene hydrocarbon (0.1 %). Sesquiterpenes (Figure 3), often with the molecular formula $C_{15}H_{24}$ (Çelik, 2020; Tosun *et al.,* 2015), are a class of terpenes composed of three isoprene units.

The raw materials, namely crude turpentine oil, pure α-pinene and β-pinene, were sourced from the Arifoglu Company in the Republic of Türkiye. Turpentine, an essential oil derived from gum resin, contains pinene as its main constituent. The resin consists of about 70-80 % rosin and 20-30 % turpentine. After extraction, the gum resin is subjected to a distillation process in which the solid residue at the bottom of the distillation apparatus is known as the rosin, which consists of resin acids and fatty acids. The volatile liquid fraction, consisting mainly of α-pinene and β-pinene, is known as turpentine. Pinene is a bicyclic monoterpene that exists in two structural isomeric forms (Salvador *et al.,* 2020; Afre *et al.,* 2006). The main components of turpentine, α-pinene and β-pinene, are shown in Figure 4.

2.2 Synthesis of epoxy-functionalized tung oil (ETO)

2.2. Sinteza epoksidom funkcionaliziranoga tungova ulja (ETO)

Tung oil epoxidation (ETO) was prepared using a methodology that is consistent with our previous study (Feist and Ross, 1995), using tung oil and glycidyl methacrylate via a Diels-Alder reaction. Initially, 25.96 g of glycidyl methacrylate was combined with 80 g of tung oil and 0.8 g of phenothiazine at 150 °C in the presence of nitrogen gas (Figure 5). The identification of the final product was carried out using H NMR [H-NMR (500 MHz, CDCl$_3$) δ(ppm): 5.64-6.34 (-C**H**=C**H**-), 5.22-5.25 (-C(O)O-CH$_2$-C**H**-O-C(O)-), 4.25-4.27 (-C(O)O-C**H**$_2$-CH-O-C(O)-), 4.12-4.14 (C(O)O-C**H**$_2$-CH(CH$_2$)O), 3.24-3.29 (O(CH$_2$)C**H**-CH$_2$-), 2.71 (-C**H**(CH=CH)CH$_2$-), 2.61-2.69 (O(C**H**$_2$)CH-CH$_2$-), 2.28 (-C**H**$_2$-C(O)O-), 2.06-2.15 (-C**H**$_2$-CH=CH-, -CH=CH-CH(CH=C**H**$_2$-), 1.56-1.66 (-C**H**$_2$-CH$_2$-C(O)O-), 1.19-1.45 -(C**H**$_2$)$_5$-CH$_2$-C(O)O-, -(C**H**$_2$)$_2$-CH$_2$-CH=CH-), 1.28 (C**H**$_3$-C- C(O)O-), 0.83-0.87 (C**H**$_3$-CH$_2$-CH$_2$)] and C NMR [C-NMR (500 MHz, CDCl$_3$) δ(ppm): 172.75 (**C**(CH$_3$)-C(O)O-), 134.71 (-CH=**C**H-), 64.67 (O(CH$_2$) **C**H-CH$_2$-), 48.93 (O(CH$_2$) **C**H-CH$_2$-), 44.17 (O(**C**H$_2$) CH-CH$_2$-)]spectroscopy (Babahan *et al.,* 2020).

2.3 Preparation of bio-based epoxide coatings (ME and TE)

2.3. Priprema epoksidnog biopremaza (ME i TE)

In this study, turpentine and moss oil were used as epoxy curing agents. 2,4,6-tris (diethylaminomethyl) phenol was selected as catalyst to facilitate the cross-linking process. Both moss oil and turpentine oil were

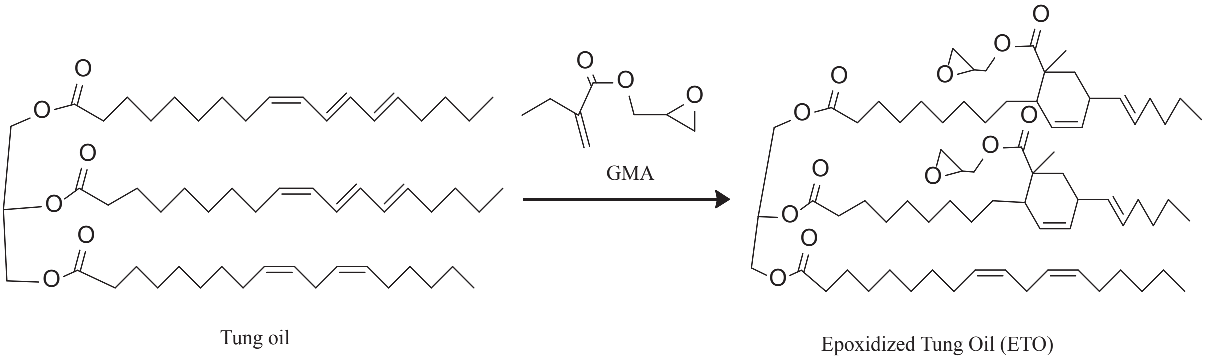

Figure 5 Epoxidation of tung oil reaction
Slika 5. Reakcija epoksidacije tungova ulja

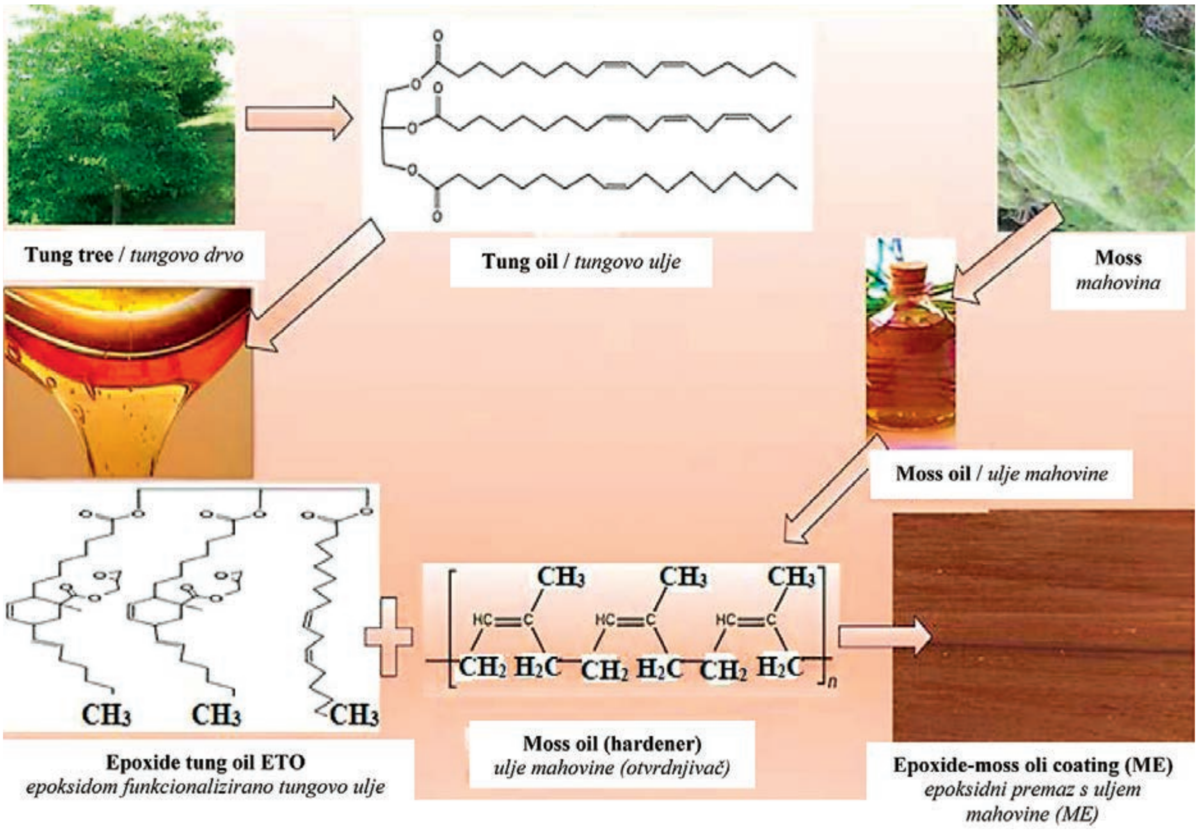

Figure 6 Preparation of epoxidized moss oil coatings (ME)
Slika 6. Priprema epoksidnog premaza s uljem mahovine (ME)

used as epoxide reactants in the preparation of formulations with a 1:1 ratio of epoxide to oil. The system obtained using moss oil as the curing agent is referred to as ME (Figure 6), while that obtained using turpentine oil as the curing agent is referred to as TE (Figure 7). A few drops of a 2 % solution of 2,4,6-tris (diethylaminomethyl) phenol was added to each formulation. This acted as a catalyst in the process. In order to prepare the coating formulations, no solvent was used. After the reactions were weighed, they were mixed and forcefully agitated for two hours. A brush was then used to apply the mixes to wood samples. Evaluations were conducted on the surface performance characteristics. All formulations were subjected to a curing process at a temperature of 25 °C for a duration of 24 hours, with the use of air circulation and a thickness of 200 µm.

2.4 Preparation of bio-based epoxide-nanocomposite coatings
2.4. Priprema epoksidnih nanokompozitnih biopremaza

A comprehensive description of the tools and experimental configurations used is given in Figure 8. This is accompanied by a schematic illustration of the procedures involved in the synthesis of epoxidized moss oil and epoxide-turpentine oil nanocoatings. These have been developed in accordance with the methodology outlined in our previous research work (Williams, 1995). A variety of carbon nanoparticles including graphene, carbon nanotubes and fullerenes were used to construct the nanocomposites. The carbon nanoparticles were initially combined with acetone in a sonication bath at a temperature of 25 °C for a period of two hours. This process was carried out with the aim of preventing the deposition of nanoparticles and facilitating their dispersion in the epoxy matrix. The nanoparticles were then remixed in the above solvent for a period of two hours using a mechanical mixer at 0.10 % mass concentration. After combining acetone with the nanoparticles (5 g: 0.10 % by mass of epoxy resin), 5 g of ETO epoxy resin was applied. The samples were left at room temperature for 24 hours to extract the mixture of nanoparticles and epoxide resin (ETO) from the solvent. Then, 5 g of bio-based hardeners, namely turpentine or moss oil, were added to the epoxy/nanoparticle mixture in a 1:1 mass ratio. The mixture was stirred vigorously for about two hours. The formulation process did not involve the use of any solvents at all. 2,4,6-dimethylaminomethyl phenol was added to all formulations.

2.5 Preparation of wood
2.5. Priprema drva

Oriental beech (*Fagus orientalis* L.) specimens in the radial, tangential, and longitudinal dimensions of 10 mm × 100 mm × 150 mm were created for the

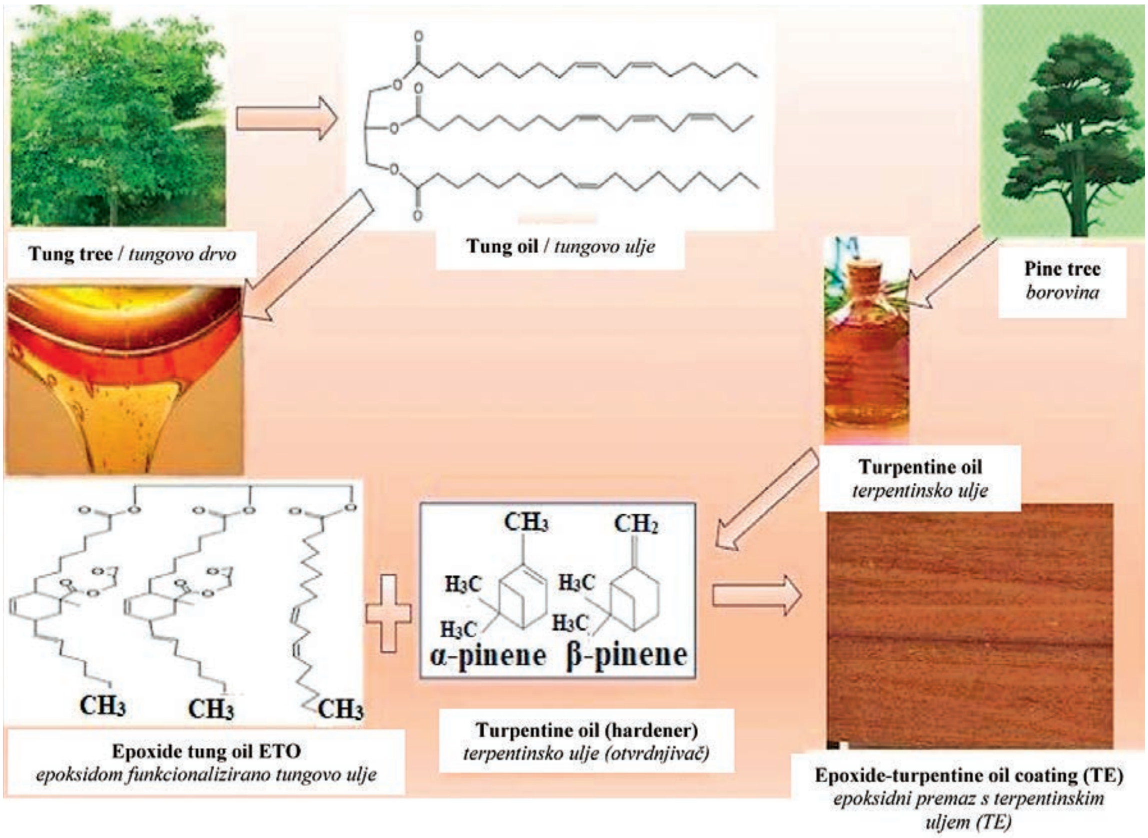

Figure 7 Preparation of epoxide-turpentine oil coatings (TE)
Slika 7. Priprema eposidnog premaza s tungovim uljem (TE)

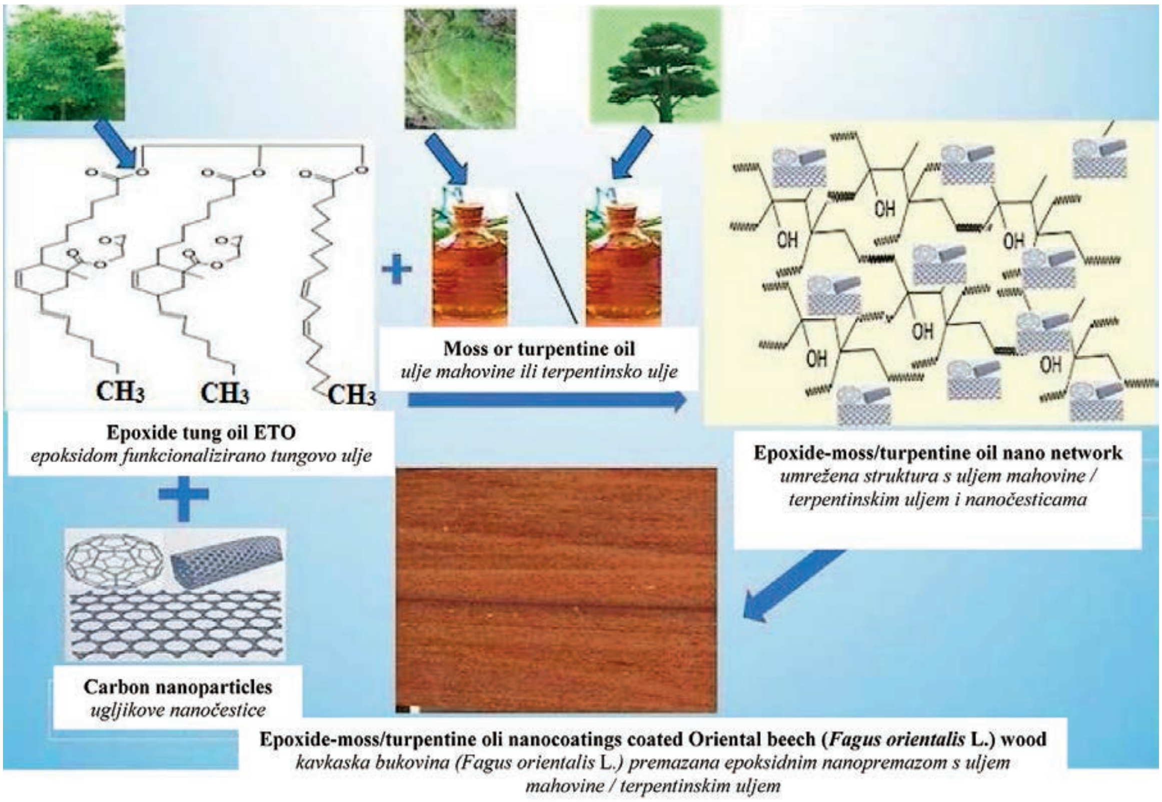

Figure 8 Use of moss and turpentine oil in combination with carbon nanoparticles to produce bio-based epoxy nanocomposite coatings
Slika 8. Upotreba ulja mahovine i terpentinskog ulja u kombinaciji s ugljikovim nanočesticama u pripremi epoksidnih nanokompozitnih biopremaza

weathering test. Appropriate sanding was used to remove any dust from the wood surfaces prior to applying the epoxy finish. Subsequently, the wooden samples underwent 220 grit sanding before the final coat was applied. One layer of the epoxy coating method was used in the investigation. On wooden surfaces, 100 g of epoxy coating were applied per square meter. A total of 90 wood specimens were prepared, 10 specimens for each control and coated groups.

2.6 Colour test
2.6. Ispitivanje boje

The *CIEL*a*b** method for the colour test was used to determine the samples' *L*, a*,* and *b** colour characteristics. The chromaticity coordinates are represented by the *a** and *b** axes in this diagram, while lightness is represented by the *L** axis. Additionally, red is represented by the symbol *+a*,* while green is represented by the symbol *-a*. +b** and *-b** are variables that alternately indicate blue and yellow. The *L** value, according to Zhang *et al*. (2003), goes from 0 (black) to 100 (white). To calculate the colour difference *(ΔE*)* in this test, Eqs. 2-5 were applied in compliance with ASTM D1536–58 T (1964) standard.

$$\Delta a^*=a_f^*-a_i^* \quad (2)$$

$$\Delta b^*=b_f^*-b_i^* \quad (3)$$

$$\Delta L^*=L_f^*-L_i^* \quad (4)$$

$$\Delta E^*=[(\Delta a^*)^2+(\Delta b^*)^2+(\Delta L^*)^2]^{1/2} \quad (5)$$

Where the differences between the values of the first and last intervals are represented, respectively, by Δa^*, Δb^*, and ΔL^*.

2.7 Gloss test
2.7. Ispitivanje sjaja

Oriental beech gloss levels were determined using the Micro-TRI-Gloss equipment and the ASTM D 523-14 (2018) standard. The geometry used here represents an incidence angle of 85°.

2.8 Surface roughness test
2.8. Ispitivanje hrapavosti površine

In accordance with DIN 4768 (1990), roughness was tested with a Mitutoyo Surftest SJ-301 instrument. Surface roughness parameters such as *Ra, Rz,* and *Rq* were determined.

2.9 Weathering test
2.9. Izlaganje vremenskim utjecajima

Wood panels are designed to expose specimens to weather conditions, as per ASTM D 358-55 (1970). The specimens were subsequently exposed to external weathering for ninety days in the Mugla region in 2024 (March, April, and May) while they remained on the panels. The weather statistics for Muğla are shown in Table 1.

2.10 Statistical evaluation
2.10. Statistička obrada

The experimental data were transferred to SPSS 16.0 statistical program and then Duncan test was calculated to determine non-parametric differences between groups with 95 % confidence level. Homogeneity groups (HG) were created using alphabetical letters to test whether there was statistical significance between various sample types.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

The present study encompasses the use of eco-friendly, human-friendly, and bisphenol A-free epoxy coatings, along with their nanocomposite derivatives. The resin and the curing agent are both derived from natural sources.

The present study is original in terms of its focus on the utilization of sustainable natural resources within the coatings industry, and the development of cost-effective and readily available systems in comparison to oil-based systems. The research is also original in that it represents the first use of moss and turpentine oils as curing agents in epoxy curing reactions. A comparison has been made between the results obtained with these two oils, which have a similar chemical structure. Furthermore, the impact of the nanoparticles on the surface characteristics of the bio-based coatings will be investigated.

New bio-based epoxy nanocoating systems have been developed to replace BPA-based epoxy coatings

Table 1 Muğla meteorological statistics (Muğla meteorological data, 2024)
Tablica 1. Meteorološka statistika za Muğlu (Meteorološki podatci za Muğlu, 2024.)

Parameter / *Izmjerena veličina*	**March** *Ožujak*	**April** *Travanj*	**May** *Svibanj*
Maximum monthly temperature, °C / *Najviša mjesečna temperatura,* °C	27.30	29.10	32.90
Average temperature per month, °C / *Prosječna mjesečna temperatura,* °C	10.30	16.90	18.70
Minimum temperature per month, °C / *Najniža mjesečna temperatura,* °C	-0.90	4.60	6.40
Humidity per month, % / *Mjesečna vlažnost,* %	69.60	56.60	53.90
Monthly average wind speed, m/s / *Prosječna mjesečna brzina vjetra*, m/s	0.90	1.00	1.00
Monthly average wind speed, mm = kg/m^2 / *Prosječna mjesečna brzina vjetra*, mm = kg/m^2	78.50	51.60	10.00
Total number of rainy days / *Ukupan broj kišnih dana*	10.00	3.00	4.00

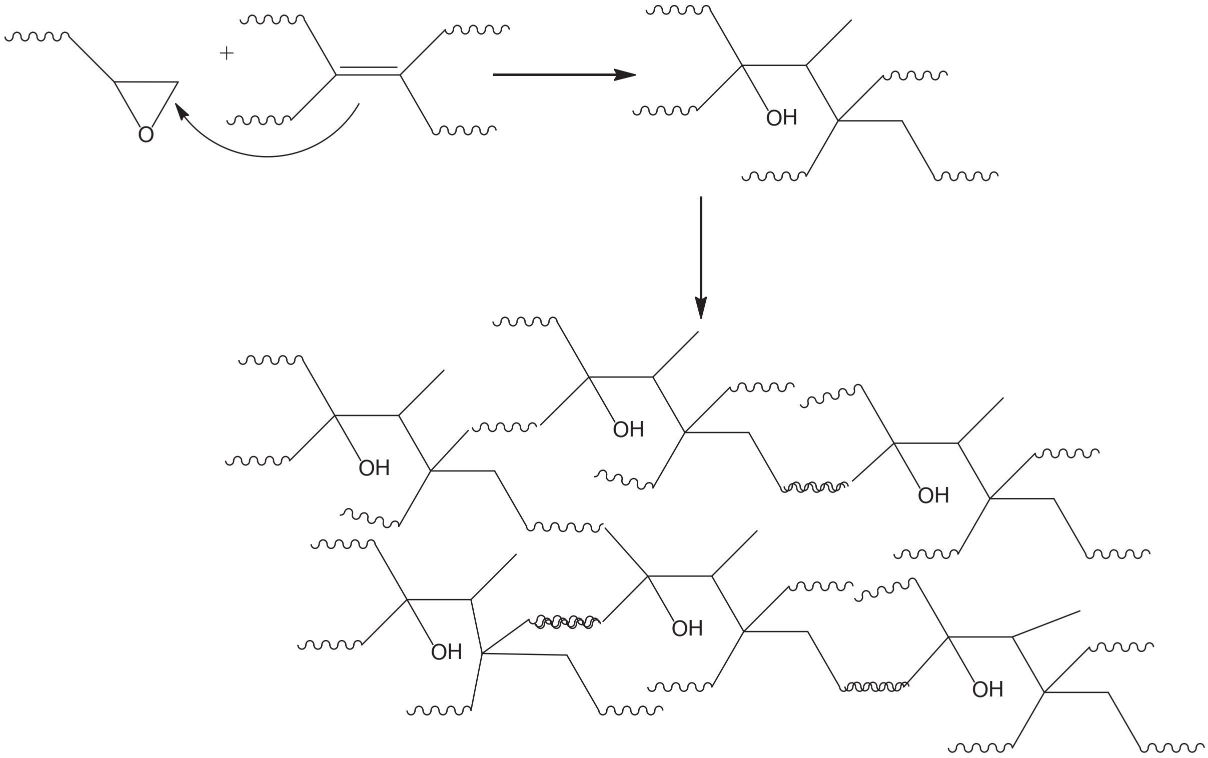

Figure 9 Chemical reaction of epoxy ring opening and cross-linking
Slika 9. Kemijska reakcija otvaranja i umrežavanja epoksidnog prstena

in the wood industry. For this research, Oriental beech (*Fagus orientalis* L.) was the wood species of choice. To achieve this, an epoxy resin based on tung oil was used in a Diels–Alder reaction (ETO) to create a new generation of bio-based epoxy nanocoatings. Additionally, this study was the first to use hardeners such as moss oil and turpentine oil to cure the epoxy resin. The first step in the process involved combining tung oil with glycidyl methacrylate. This forms the basis of the bio-based epoxy resin (ETO). The epoxy resin was then cured using curing agents to obtain ME and TE systems. The epoxy resin (ETO) was premixed with carbon nanoparticles (including graphene, carbon nanotubes (CNTs) and fullerenes) before being cured with moss oil and turpentine oil in order to obtain nanocomposite derivatives of these coatings. The surface properties of beech wood, including colour, gloss and surface roughness, were also investigated. A comparative analysis of the epoxy coatings obtained by curing the bio-based tung oil epoxy resin with moss/turpentine oil and its nanoparticle derivatives produced some interesting results. The results for colour, gloss and surface roughness are presented in Tables 2–4.

In the initial phase of the study, a bio-based epoxy resin (ETO) was synthesized through the epoxidation of tung oil using the Diels-Alder reaction. The curing of this bio-based epoxy resin (ETO) was achieved through the use of bio-based hardeners. A thorough examination of the chemical structures of the active ingredients in turpentine and moss oil, which are used as hardeners, reveals the presence of analogous double bonds in both substances. These double bonds enable them to act as nucleophiles (Hanif *et al*., 2024; Parker and Isaacs, 1959). In the presence of an epoxy ring and a nucleophile, the latter attacks the former, thereby initiating the ring-opening reaction. At this point, crosslinking occurs (Hanif *et al*., 2024; Parker and Isaacs, 1959). An increase in crosslinking density is achieved through the reaction between oxirane groups and double bonds. To the best of the present author's knowledge, the use of epoxidized tung oil in this manner has not been documented in the extant scientific literature. The process is illustrated in Figure 9.

3.1 Colour
3.1. Boja

As illustrated in Table 2, the values for the colour changes, both before and after weathering, are presented for the coated Oriental beech samples and the control group.

In the course of the present investigation, it was established that the L^* values of the control samples prior to weathering were 71.47. In addition, a decline in L^* values was observed for samples treated with epoxidized plant-based compounds in comparison to the control samples. Surface darkening has been demonstrated to be associated with a decline in initial L^* values, while lighter hues are observed in samples when L^* values increase (Bonifazi *et al*., 2017; Salas *et al*., 2016). A decline in the ΔL^* values of all samples

Table 2 Values of colour changes before and after weathering
Tablica 2. Vrijednosti promjene boje prije i nakon izlaganja uzoraka vremenskim utjecajima

Chemicals *Kemikalije*	**Before weathering** *Prije izlaganja vremenskim utjecajima*			**After weathering for 3 months** *Nakon tri mjeseca izlaganja vremenskim utjecajima*			**After weathering for 3 months** *Nakon tri mjeseca izlaganja vremenskim utjecajima*			**Total colour changes** *Ukupne promjene boja*			
	L^*	a^*	b^*	L^*	a^*	b^*	ΔL^*	Δa^*	Δb^*	ΔE^*	*Std. Dev.*	*COV*	*H.G.*
Control	71.47	6.16	19.43	65.88	10.80	20.98	-5.59	4.64	1.55	7.42	2.12	28.57	BC
ME	65.40	10.41	30.32	55.90	15.13	26.76	-9.5	4.72	-3.56	11.18	5.45	48.74	D
MEC	58.27	11.49	31.09	48.85	16.41	25.74	-9.42	4.92	-5.35	11.89	4.12	34.65	D
MEF	61.50	11.91	31.42	54.30	15.65	27.00	-7.2	3.74	-4.42	9.23	2.78	30.11	BCD
MEG	58.87	11.95	30.71	52.15	16.19	18.68	-6.72	4.24	-12.03	14.41	1.13	7.84	E
TE	61.18	11.49	31.03	53.61	15.13	26.71	-7.57	3.64	-4.32	9.44	3.56	37.71	CD
TEC	58.01	7.78	24.11	54.91	11.11	21.15	-3.1	3.33	-2.96	5.42	2.45	45.20	A
TEF	60.75	11.00	30.24	54.55	15.69	27.14	-6.2	4.69	-3.1	8.36	3.15	37.67	BC
TEG	55.93	9.10	25.96	51.38	12.36	22.15	-4.55	3.26	-3.81	6,77	4,14	61,15	AB

Note: ME – Epoxidized moss oil, MEC – Epoxidized moss oil with added carbon nanotubes, MEF – Epoxidized moss oil with added fullerene, MEG – Epoxidized moss oil with added graphene, TE – Epoxied tung oil, TEC – Epoxidized tung oil with added carbon nanotubes, TEF – Epoxidized tung oil with added fullerene, TEG –Epoxidized tung oil with added graphene, Std. Dev. – Standard deviation, COV – Coefficient of Variation, H.G. – Homogeneity group
Napomena: ME – epoksidirano ulje mahovine, MEC – epoksidirano ulje mahovine s dodatkom ugljikovih nanocijevi, MEF – epoksidirano ulje mahovine s dodatkom fulerena, MEG – epoksidirano ulje mahovine s dodatkom grafena, TE – epoksidirano tungovo ulje, TEC – epoksidirano tungovo ulje s dodatkom ugljikovih nanocijevi, TEF – epoksidirano tungovo ulje s dodatkom fulerena, TEG – epoksidirano tungovo ulje s dodatkom grafena, Std. Dev. – standardna devijacija, COV – koeficijent varijacije, H.G. – homogene grupe

was observed during the process of weathering. The parameter that best captures the evolution of a wood surface colour is designated as ΔL^* (De Lorean, 1999). Consequently, the surface of the wood underwent a process of darkening with the passage of time. The darkening that is associated with weathering on Oriental beech trees is caused by photodegradation and leaching of lignin and other non-cellulosic polysaccharides (Sönmez *et al*., 2011; Hon and Chang, 1985). Following three months of weathering, a propensity to turn reddish was observed in all samples, yielding a value of $+\Delta a^*$. Furthermore, the analysis revealed that, except for the control samples, all coated samples exhibited a propensity to undergo a blue colouration, resulting in a $-\Delta b^*$ value. The MEG samples demonstrated the most significant colour change, while the TEC samples exhibited the least colour change, as indicated by the total colour change (ΔE^*) values. The enhanced colour stability of this species, as evidenced by its reduced overall colour change, is indicative of its superior resistance to weathering. Consequently, the group demonstrating the most efficacy in counteracting colour change was the TEC samples. The degree of wood degradation behind the layers of coatings has been shown to directly correlate with colour change (Saha, 2011). In comparison with the control samples, all of the coated samples in the present investigation, with the exception of the TEC and TEG samples, failed to demonstrate adequate colour stability. Mahlberg *et al.* (2006) conducted a study on the subject of the ageing of wood surfaces that had been treated with multifunctional alkoxysilanes, which had been formed by means of sol-gel deposition. A minimal colour change was observed on the sol-gel-coated wood surfaces following a rapid weathering process. As the TEC samples in our study demonstrated the lowest colour change value, the findings of Mahlberg *et al*. (2006) and our own results are consistent. Moreover, a lesser degree of colour change was exhibited by TE and its derivative-coated samples in comparison with ME and its derivative-coated samples.

The objective of Altay *et al.* (2024) was to modify the colour characteristics of Oriental beech wood following weathering. The findings of the research indicate a predilection for novel bio-based epoxide-amine (EP) coatings in the field of coating materials. The creation of fullerene, graphene, and carbon nanotube-containing nano-composite coating derivatives was achieved through the interaction between epoxy-functionalized tungsten and carbon nanotubes. Epoxide tung oil (ETO) doped with nanoparticles and isophorone diamine, a diamine hardener, were cured. The findings demonstrated that, subsequent to weathering, the epoxy nanocomposite-covered samples exhibited a more stable colour shift in comparison to the control group. The effects of consecutive treatments with succinic anhydride and epoxidized soybean oil, respectively, on colour changes while exposed to ultraviolet radiation were investigated by Rosu *et al*. (2016). The findings indicated that the total colour change values exhibited a diminished increase with irradiation time and dose when compared to untreated wood. The findings of this study demonstrate a congruence with those of Rosu *et al*. (2016) and Altay *et al*. (2024), as evidenced by the observation that the specimens coated with TEC exhibited a diminished overall increase in colour change in comparison to the control group.

3.2 Gloss
3.2. Sjaj

As illustrated in Table 3, the gloss alterations that occurred prior to and following the weathering process are documented for the coated Oriental beech samples and the control group.

Glossiness, defined as the result of incident light reflected from different directions on any given surface, is a useful metric for evaluating the quality of a finished product (Ged *et al*., 2010; Vardi *et al*., 2010). Despite the prevalence of high gloss surfaces in the furniture industry, matte gloss continues to hold significance in several solid wood furniture markets (Ettwein *et al*., 2017). The analysis demonstrated that the gloss value of the control samples was 9.23 prior to weathering. With the exception of the MEF and TEF samples, a decline in gloss values was observed for the remaining coated samples in comparison to the control samples. The gloss of a surface is influenced by a multitude of factors beyond the chemical composition of the coating itself. As Lee *et al*. (2003) demonstrate, the coating can be rendered highly glossy by means of additional applications and polishing processes.

In the present study, it was observed that the gloss of the coated samples increased when fullerene was added prior to weathering. It was evident that all samples, both coated and uncoated (control), exhibited a decline in gloss values subsequent to three months of weathering. The ME samples demonstrated the most significant decline, while the TEC samples exhibited the least amount of decrease. Prior to the application of a commercial hydrophobic topcoat, Panek *et al*. (2017) modified the oak wood surfaces by employing UV stabilisers, ZnO and TiO_2 nanoparticles, and hindered amine light stabilizer (HALS). The control coating system was an oil-based transparent coating. Throughout the six-week artificial weathering test, changes in gloss were systematically evaluated. Following a period of three weeks, the luster of the oil-based coating diminished. In view of the finding of a decline in gloss in all coated samples following weathering, the results of the present study are consistent with those reported by Panek *et al*. (2017). A statistically significant discrepancy was identified between the TEC samples and the other samples. Moreover, at the 95 % confidence level, no statistically significant difference was identified between the MEC and TEG samples. Kabasakal *et al*. (2024) investigated the alterations in the gloss of Oriental beech. In the study conducted by Kabasakal *et al*. (2024), the alterations in gloss observed in Oriental beech following weathering were investigated. These alterations were attributed to the application of vegetable oil-based epoxide-amine (ETO + D230) and its nanocomposite derivatives, which were impregnated with carbon nanoparticles. The findings of the study demonstrated that, whilst the gloss values of all samples decreased, the decline observed in the coated samples was less pronounced than that of the control. The findings of the present study demonstrated comparable values. The results of the study demonstrated that samples treated with tung oil exhibited a lower level of gloss compared to samples treated with moss oil. In a separate study, Rosu *et al*. (2021) used epoxidized oils of grapeseed, soybean, and linseed, in conjunction with diglycidyl ether of bisphenol A that was cross-linked employing a resin acid/maleic anhydride adduct, to formulate eco-friendly polymer coatings for the purpose of shielding pine wood surfaces from ultraviolet (UV) radiation. The fabrication of semi-transparent film-covered surfaces was achieved by subjecting wood to synthetic formulations and subsequently curing them at elevated temperatures. The findings demonstrated that, in comparison to uncoated wood, the

Table 3 Gloss changes before and after weathering
Tablica 3. Promjene sjaja prije i nakon izlaganja uzoraka vremenskim utjecajima

Chemicals *Kemikalije*	**Before weathering** *Prije izlaganja vremenskim utjecajima*	**After weathering for 3 months** *Nakon tri mjeseca izlaganja vremenskim utjecajima*	**After weathering for 3 months, %** *Nakon tri mjeseca izlaganja vremenskim utjecajima, %*			
	85°	*85°*	*85°*	*Std. Dev.*	*COV*	*H.G.*
Control	9.23	2.00	-78.33	6.15	-7.85	C
ME	9.06	1.4	-84.54	4.78	-5.65	CD
MEC	4.62	1.8	-61.03	5.26	-8.61	B
MEF	9.42	2.37	-74.84	7.45	-9.95	C
MEG	7.70	1.62	-78.96	4.13	-5.23	C
TE	9.13	2.00	-78.09	3.78	-4.84	C
TEC	2.92	1.55	-46.91	6.65	-14.17	A
TEF	11.20	2.07	-81.51	4.48	-5.49	CD
TEG	6.76	2.2	-67.45	2.78	-4.12	B

gloss rate of coated wood samples was more consistent when subjected to UV irradiation.

The findings of the present study are consistent with those of Rosu *et al*. (2021), as indicated by the results obtained.

3.3 Surface roughness
3.3. Hrapavost površine

As illustrated in Table 4, the surface roughness of the coated Oriental beech samples and the control group was subjected to a series of weathering tests, which revealed significant alterations in surface texture before and after the weathering process.

The anatomical structure and characteristics of the wood, the machining tools, and the cutting conditions all exert a significant influence on the resultant surface roughness of wood products. The wood quality in terms of density, moisture content, texture, and anatomical structure have been identified as the factors influencing roughness (Aguilera and Muñoz, 2011; Gurau *et al*., 2006). A key desirable quality of wood used in industry is its low surface roughness. In the present study, the investigation focused on the weathering behaviour of coated samples over a period of three months. The results obtained revealed an increase in the roughness parameters (*Ra, Rz, and Rq*) of the coated samples in comparison to the control samples. However, this increase was not observed in the TEF samples. The *Ra, Rz*, and *Rq* values of the control samples were found to be 2.08, 13.94, and 2.70, respectively. The investigation revealed a decline in the roughness values of the coated samples at all levels of roughness following three months of weathering, in comparison to the control sample.

The application of vegetable-based epoxy to wood had a mitigating effect on the samples' surface roughness in our investigation. Following exposure to the elements, the untreated control samples exhibited a substantial density of cracks on their surfaces. The combination of these fissures with the degradation of the earlywood cells (Xie *et al*., 2008) resulted in a significant increase in surface roughness. The conclusions of the present study are corroborated by those of Xie *et al.* (2008), who observed an increase in surface roughness values in the uncoated samples in comparison to the coated ones. The effects of water spray and UV light irradiation on the surface roughness of untreated and pretreated Scots pine sapwood samples were examined by Özgenç *et al*. (2013). A variety of seed oils were applied to the specimens, including canola, sesame, nigella, pomegranate, parsley, and soybean. A comparison of the surface roughness values of the control samples with those of the pine wood samples treated with vegetable oil revealed a general decrease in surface roughness during the irradiation process. The findings of this study are in alignment with those of Özgenç *et al*. (2013).

The anisotropic and heterogeneous nature of wood renders it a complex material, and a number of factors, including anatomical variations, growth traits, and machining properties, must be considered when assessing the surface roughness of wood. The investigation revealed generally a statistically variation in roughness values between the coated and control (untreated) samples. Furthermore, the samples treated with epoxy and tung oil (TE) exhibited the lowest values of surface roughness in all surface roughness assessments (*Ra, Rz, and Rq*). In addition, these samples rendered the wood material the most stable against roughness. In a recent study, Wang *et al*. (2024) investigated the combination of stearoyl chloride grafted cellulose nanocrystals (SCNCs) and cellulose nanocrystals (CNCs) to create a novel coating material. This material demonstrated exceptional compatibility with wood wax oil (WWO) coatings. The findings in-

Table 4 Values of surface roughness changes before and after weathering
Tablica 4. Vrijednosti promjene hrapavosti površine prije i nakon izlaganja uzoraka vremenskim utjecajima

Chemicals *Kemikalije*	**Before weathering** *Prije izlaganja vremenskim utjecajima*			**After weathering for 3 months** *Nakon tri mjeseca izlaganja vremenskim utjecajima*			**After weathering for 3 months, %** *Nakon tri mjeseca izlaganja vremenskim utjecajima, %*											
	*Ra**	*Rz**	*Rq**	*Ra**	*Rz**	*Rq**	*Ra**	*Std. Dev.*	*COV*	*H.G.*	*Rz**	*Std. Dev.*	*COV*	*H.G.*	*Rq**	*Std. Dev.*	*COV*	*H.G.*
Control	2.08	13.94	2.7	4.15	24.48	5.1	99.51	8.75	8.79	D	75.60	9.75	12.89	D	88.88	4.15	4.66	C
ME	3.00	17.59	3.81	5.21	30.25	6.78	73.66	6.12	8.30	C	71.97	7.15	9.93	D	77.95	7.85	10.07	BC
MEC	2.51	14.44	3.36	4.13	23.43	5.47	64.54	7.48	11.58	BC	62.25	4.56	7.32	BCD	62.79	6.25	9.95	B
MEF	2.94	19.83	3.5	4.59	30.15	6.11	56.12	4.75	8.46	B	52.04	5.12	9.83	BC	74.57	4.89	6.55	BC
MEG	3.18	18.49	3.98	4.29	28.1	5.58	34.90	3.78	10.83	A	51.97	6.75	12.98	BC	40.20	3.24	8.05	A
TE	3.68	20.31	4.64	4.39	25.42	5.68	19.29	2.45	12.70	A	25.16	3.12	12.40	A	22.41	4.89	21.82	A
TEC	2.64	14.84	3.29	4.02	26.18	5.78	52.27	6.16	11.78	B	76.41	8.75	11.45	D	75.68	3.55	4.69	BC
TEF	1.81	10.68	2.26	3.05	17.89	4.22	68.50	7.14	10.42	BC	67.50	3.25	4.81	CD	86.72	2.15	2.47	C
TEG	2.29	14.42	2.94	3.47	21.45	5.12	51.52	5.56	10.79	B	48.75	3.45	7.07	B	74.14	6.35	8.56	BC

dicated minimal variation in the arithmetic mean height and a predominantly smooth texture on surfaces coated with WWO. Furthermore, a steady decrease in roughness was observed as the amount of SCNC in the coatings increased. In their study, Kabasakal *et al*. (2024) investigated the changes in the roughness quality of naturally aged Oriental beech that had been covered with derivatives of nanocomposite materials and vegetable oil-based epoxide-amine (ETO+D230). The findings of the study indicated that all coated samples exhibited smoother surfaces in comparison to the control group following weathering. The outcomes of the present study are corroborated by extant literature, as evidenced by the decrease in surface roughness of the coated samples following weathering in comparison to the control samples.

4 CONCLUSIONS
4. ZAKLJUČAK

Bio-based epoxy resin (ETO), derived from epoxidising tung oil, has been cured with bio-based moss oil and turpentine to create new coating systems for wood surfaces that are both human and environmentally friendly. These coating systems offer an alternative to BPA-based epoxy coatings for wood surfaces. In this study, the use of moss oil and turpentine oil as hardeners for epoxy resin constitutes a novel approach. The incorporation of carbon nanoparticles into epoxy oil and epoxy-turpentine oil systems has been shown to yield nanocomposite derivatives. An investigation was conducted into the surface properties of novel bio-based coatings and their nanocomposite derivatives, with a view to drawing comparisons between them.

The present study investigates the enhancement of properties such as colour, gloss and surface roughness of epoxidized tung oil containing amine-cured carbon nanoparticles coated on wood material following weathering over a period of three months. The findings of this study demonstrate that, while the coated samples exhibited minimal positive outcomes in terms of total colour change (ΔE^*) in comparison to the control samples in weathering-induced colour changes, the TEC samples demonstrated enhanced colour stability in comparison to the control and other coated groups. Subsequent to weathering, a decline in gloss value was observed for all samples. Furthermore, TEC samples exhibited the least loss of gloss. As demonstrated by the surface roughness results after weathering, whilst the roughness of all samples increased, this increase was less pronounced in the coated groups in comparison to the control group.

In summary, the newly developed bio-based nanocoatings have been shown to enhance the gloss and roughness properties of Oriental beech. This outcome indicates that the use of vegetable-based epoxy is advisable in scenarios where the preservation of wood material's gloss against external weather conditions and the creation of a less coarse surface are desired.

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Material Recycling of Particleboard and Fibreboard – a Literature Review

Recikliranje materijala od iverica i ploča vlaknatica – pregled literature

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ABSTRACT • *The demand for sustainable products and production methods is driving renewed interest in wood as a renewable and environmentally friendly raw material. The recycling of wood-based materials, such as particleboard and fibreboard, is crucial in meeting this demand while conserving natural resources. This article reviews advances in the technology of wood-based material recycling, analysing the literature from the turn of the millennium to the present day, with a focus on pulping processes and their effects on physical and mechanical properties. The review identifies several established and emerging processes, including mechanical, thermohydrolytic and innovative ohmic heating processes, and notes that apart from the established processes, no other processes have been successfully scaled up to industrial application. A particular challenge remains in the processing of materials bonded with urea-formaldehyde resins, where recycling has a significant impact on particle geometry and adhesive residues, affecting product quality. Promising improvements in mechanical properties have been observed with resistive heating, suggesting potential for this technique in future recycling applications. Overall, this review highlights the need for scalable, efficient recycling solutions to meet industry sustainability goals.*

KEYWORDS: *fibreboard; literature review; particleboard; recycling*

SAŽETAK • *Zahtjev za održivim proizvodima i procesima potiče ponovno zanimanje za drvo kao obnovljivu i ekološki prihvatljivu sirovinu. Recikliranje materijala na bazi drva, poput iverice i ploča vlaknatica, imperativ je za zadovoljavanje potražnje drva uz očuvanje prirodnih resursa. Članak donosi pregled napretka u tehnologiji recikliranja materijala na bazi drva analiziranjem literature od prijelaza tisućljeća do danas, s naglaskom na procesima proizvodnje pulpe i njihovu utjecaju na fizička i mehanička svojstva drvnih čestica. U radu je identificirano nekoliko ustaljenih i novih procesa, uključujući mehaničke, termohidrolitičke i inovativne omske procese zagrijavanja, te se naglašava da osim ustaljenih procesa nijedan drugi proces nema industrijsku primjenu. Poseban je izazov obrada materijala lijepljenih urea-formaldehidnim smolama, pri čemu recikliranje ima važan utjecaj na geometriju čestica i ostatke ljepila, što utječe na kvalitetu proizvoda. Obećavajuća poboljšanja mehaničkih svojstava uočena su pri omskom zagrijavanju, što pokazuje da ta tehnika ima potencijala u budućim metodama recikliranja. Općenito, u ovom se radu ističe potreba za učinkovitim rješenjima recikliranja kako bi se postigli industrijski ciljevi održivosti.*

KLJUČNE RIJEČI: *ploča vlaknatica; pregled literature; iverica; recikliranje*

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1 INTRODUCTION
1. UVOD

The accumulation of medium-density fibreboard (MDF) and chipboard waste poses significant environmental and economic challenges. These wood-based materials, widely used in furniture and construction, contain adhesives and resins that complicate recycling efforts and can lead to harmful emissions when improperly disposed of. The growing volume of this waste, coupled with its resistance to conventi onal recycling methods, has created an urgent need for innovative solutions.

The amount of waste wood in Germany in 2020 was about 16.2 million m³, of which only about 15 % was recycled (Mantau, 2023). The remaining 85 % of waste wood will be used for energy production. This current low recycling rates represent a significant loss of wood chips and fibres that could be reused in new products. A key element of the Circular Economy Act, which came into force in 2015, is to increase the recycling of waste. While the German law does not specify a recycling rate, the European Union's circular economy package includes a target to recycle at least 65 % of municipal waste by 2035 and at least 70 % of construction and demolition waste and packaging waste by 2030. In Germany, a total of 7.8 million tons of waste wood from these three sectors was generated in 2016, which is around 70 % of the total volume (Flamme *et al.,* 2020). This means that there is still a large gap to the planned recycling rates. Wood-based materials in which the wood fibres have been glued pose a particular challenge for the recycling of waste wood. Particleboard and fibreboard are widely used furniture materials whose increasing demand and relatively short life cycles lead to increased waste generation and disposal problems (Zimmer *et al.,* 2023). This article provides an overview of recycling technologies for these wood-based materials, identifies challenges and discusses possible solutions.

Therefore, this article reviews scientific publications on particleboard and fibreboard recycling that have been published since the papers "On the Recycling of Chipboard and MDF Panels" Parts 1 & 2 by Franke and Roffael were published (1998a and 1998b). The first part of Franke and Roffael's work (Franke and Roffael, 1998a) investigated the hydrolysis resistance of cured urea formaldehyde (UF) resins in particleboard and medium density fibreboard (MDF) under the influence of steam hydrolysis, while the second part (Franke and Roffael, 1998b) compared formaldehyde emissions from UF-bonded particleboard and MDF panels with fresh pine flakes during and after various hydrolysis processes.

Until around the turn of the millennium, the processes for recycling wood composites were divided into mechanical, thermohydrolytic and chemical processes. Apart from mechanical processes for crushing materials into chips and reusing them in chipboard, only a few other processes have become established industrially. Mechanically crushed chips have a number of disadvantages. Typically, the structure of the wood particles is severely damaged; in addition, the chips have a lower water retention value and the wettability with binding agents is poorer than with fresh chips or chips obtained by thermo-hydrolytic digestion (Franke, 1999). The Pfleiderer process, the WKI process and the Formaplan process are among the few industrially realisable processes for the thermo-hydrolytic digestion of particleboard.

2 MATERIALS AND METHODS
2. MATERIJALI I METODE

Based on the work of Franke and Roffael (1998a and 1998b), the scientific literature on particleboard and medium density fibreboard (MDF) was searched, analysed and compiled into a literature review using three research methods in literature databases.

Method 1: Analysis of literature referencing the work of Franke and Roffael (1998a and 1998b) by examining sources citing these two articles in literature databases.

Method 2: Evaluation of an existing literature review on the production of wood composites from recycled wood (Nguyen *et al*., 2023). The literature on particleboard and MDF was extracted from the literature review, clustered, analysed and used as a reference for further literature searches. The literature was summarised in the results chapter.

Method 3: Keyword search in scientific databases:

- Google Scholar
- ResearchGate
- Science Direct
- Web of Science

The following keywords were used:

- Fibre recycling
- Furniture waste
- Waste MDF
- Wood waste sorting technologies
- Wood materials sorting technologies

The literature was first categorised by subject into particleboard recycling processes and MDF recycling processes. The literature was then subdivided into categories such as the type of wood used, the delignification process and the glue that may be broken down or opened up by the process. Tables at the beginning of each chapter provide an overview for ease of reference. The text describes the respective processes and studies, as well as the physical and mechanical properties investigated. A table in the results section provides an overview of the physical and mechanical property results.

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

3.1 Recycling processes for particleboards and their physical and mechanical properties
3.1. Procesi recikliranja ploča iverica i njihova fizička i mehanička svojstva

3.1.1 Tabular overview
3.1.1. Tablični pregled

The methods for recycling chipboard can be summarised as follows (Table 1). The various digestion processes mentioned here are explained in more detail.

3.1.2 Thermal hydrolysis
3.1.2. Toplinska hidroliza

With the Conti-Recycling process according to Kirchner and Kharazipour (2002), the residence time could be reduced by 5-7 minutes as an alternative to the established Pfleiderer process with a residence time of about 10 minutes. Particleboard waste with UF resin binder is pulped at a steam pressure of 2-6 bar and temperatures between 120-180 °C and continuously conveyed by screw conveyors through five successive spirals to an outlet which ends in a vibrating channel for material sorting and screening. The process is also suitable for recycling MDF (Kirchner and Kharazipour, 2002). In investigations on the shift of the molar ratio of formaldehyde to urea in the resin of hydrolysis residues by Roffael and Kraft (2004), chipboard waste was thermohydrolytically pulped in an autoclave at 100 °C, 110 °C, 120 °C and 130 °C; a liquor ratio of particleboard:water of 1:5 and a pulping time of 60 minutes. It was found that the molar ratio of formaldehyde:urea (F : U) in the hydrolysis residues increased significantly with higher pulping temperatures, indicating more extensive adhesive degradation.

Lykidis and Grigoriou (2008) tested four different thermohydrolytic (steam) treatments under different pressure-temperature-duration-conditions. Furthermore, a second recycling cycle was tested. It was observed that the recovered particles showed higher amounts of k < 1 mm compared to the original reference material, which likely indicates that the recycling process involves a slight reduction in wood particle size. Except for the modulus of elasticity in static bending, all mechanical properties of the tested recycled panels decreased compared to the reference panels. A reduction in particle size was also noted, with the suspicion that this could negatively affect bending strength. Regarding hygroscopic properties, water absorption and thickness swelling increased. An increase in bulk density was also observed. The quality was further deteriorated by the second recycling process. Using higher temperatures leads to thermal decomposition of the wood components, resulting in a significant deterioration of the mechanical properties of the recycled panels. The degradation in quality can be significantly reduced by using milder hydrolysis temperatures. It is recommended to use blends of fresh and recycled wood particles to avoid significant degradation in the quality of the produced panels while still meeting applicable specifications. In further investiga-

Table 1 Overview of the literature focusing on the recycling of waste wood into chipboards. The literature was categorised based on the origin of the waste wood, the pulping process, and the adhesive (UF – urea formaldehyde; PF – phenol formaldehyde) suitable for the digestion process

Tablica 1. Pregled literature o recikliranju otpadnog drva u ivericu. Literatura je kategorizirana na temelju podrijetla otpadnog drva, procesa proizvodnje pulpe i ljepila (UF – urea formaldehid; PF – fenol formaldehid) prikladnoga za proces digestije.

Wood-composite *Kompozitno drvo*	**References** *Literatura*	**Waste wood origin** *Podrijetlo drvnog otpada*	**Digestion process** *Proces digestije*	**Adhesive** *Ljepilo*
Particleboard *iverica*	Franke and Roffael, 1998a	Particleboard / *iverica*	Thermal hydrolysis *toplinska hidroliza*	UF
	Kirchner and Kharazipour, 2002	Particleboard / *iverica*	Thermal hydrolysis *toplinska hidroliza*	UF
	Lykidis and Grigoriou, 2008	Particleboard / *iverica*	Thermal hydrolysis *toplinska hidroliza*	UF
	Zamarian *et al.*, 2017	MDF, Particleboard, Plywood, Construction and demolition wood *MDF, iverica, furnirska ploča, konstrukcijsko drvo*	Mechanical processes *mehanički procesi*	UF
	Laskowska and Mamiński, 2018	Plywood / *furnirska ploča*	Mechanical processes *mehanički procesi*	UF, PF
	Iždinský *et al.*, 2020	MDF, Particleboard, Paletts, HDF *MDF, iverica, palete, HDF*	Mechanical processes *mehanički procesi*	Unknown *nepoznato*
	Lubke *et al.*, 2020	Plywood / *furnirska ploča*	Thermohydrolysis *termohidroliza*	UF

tions by Lykidis and Grigoriou (2011) using thermohydrolysis, partial agglomeration of the recovered chip material was addressed, which is attributed to incomplete hydrolysis of the UF resin under the parameters used. Using new hydrolysis parameters (150 °C/10 min), the issue could largely be resolved. Under the aforementioned parameters, better results were achieved in terms of shear strength, surface strength, bending modulus of elasticity, and hygroscopic properties compared to experiments by Lykidis and Grigoriou (2008).

3.1.3 Mechanical processes
3.1.3. Mehanički procesi

Zamarian *et al.* (2017) and Azambuja (2018) applied a purely mechanical shredding method in laboratory tests to produce particleboard from a mixture of furniture waste and construction and demolition wood (MDF, particleboard, plywood and timber). The material was pre-crushed in a crusher and then treated in two passes in a special shredder. The addition of recycled chip material resulted in a reduction in water absorption and thickness swelling after 2 and 24 hours. Residual paraffin emulsions and adhesive residues from the manufacture of the original panels are thought to affect resistance. There is also an increase in bulk density due to the adhesive residues (Zamarian *et al,* 2017; Azambuja, 2018). Azambuja (2018) found that the aspect ratio of particles from particleboard and plywood panels had statistically lower average values compared to fresh reference particles and particles from wood. Regarding shear strength, the panels of all treatments made with particles from furniture waste showed equal or better values compared to the reference panels. For flexural strength, only the 10 % furniture waste treatment met the minimum requirements of the standard (Zamarian *et al.,* 2017). Since significantly better results were obtained for shear strength than for bending strength up to a certain proportion of recycled chips, it is recommended to mix the particles up to 50 % in the core layer of the particleboard, as less bending stress occurs in the core layer and the stress here affects the shear strength more (Azambuja, 2018).

Laskowska and Mamiński (2018) investigated the properties of particleboards made from post-industrial UF and PF bonded plywood. A purely mechanical method using a wood shredder was used. Initially, as observed in the studies by Zamarian *et al.* (2017) and Azambuja (2018), an increase in bulk density was observed, which, in the case of plywood, is influenced by the adhering residual adhesive and the compaction effect during veneer pressing. As a result of the increase in bulk density, it was observed that pressing times could be reduced by 3 to 6 % when using recycled material. A review of the literature by Humphrey and Bolton (1989) confirms this phenomenon and describes how the core layer becomes less compact at high bulk densities, leading to an acceleration of heat transfer. Similar to the studies by Zamarian *et al.* (2017) and Azambuja (2018), a decrease in flexural strength and a reduction in thickness swelling were also observed. Furthermore, the type of original binder (UF or PF) was found to have no effect on the comminution process and the properties of the particles produced.

Iždinský *et al.* (2020) also used mechanical crushing in their investigations to produce UF resin-bonded laboratory particleboards for property determination from a mixture of material waste, fir wood pallets, old furniture and defective particleboards. All materials were processed with a ring flaker and then ground into particles in a mill. Reduced thickness swelling was observed after 2 and 24 hours of immersion in water, particularly for UF-bonded particleboard. The *MOR*, *MOE* and internal bond IB of the particleboards were negatively affected by increasing amounts of recycled material. The results are in agreement with those of Zamarian *et al.* (2017), Azambuja (2018) and Laskowska and Mamiński (2018).

3.1.4 Thermohydrolysis
3.1.4. Termohidroliza

Lubke *et al.* (2020) soaked pieces of chipboard for 30 minutes to absorb moisture and allow the samples to swell. A Pallmann drum chipper with longitudinal slots was used for shredding. The bending strength of the recycled particleboard decreased by 13 % and the shear strength by 34 %, while the modulus of elasticity in static bending showed a slight increase of 1.3 %. Comparing the results with those of other studies, it should be noted that Lykidis and Grigoriou (2008; 2011) obtained similar results under hydrothermal conditions. They also found that after the second hydrothermal treatment under pressure (167 °C, 20 min), the Young's modulus increased by 20.7 %. The researchers suggest that the thermohydrolytic treatment increases the elasticity of the chips, which can lead to a reduction in the long-term deflection of the panels. Multiple recycling of the panels further degrades the aforementioned properties, as also described by Lykidis and Grigoriou (2011).

3.2 Recycling processes for MDF and their physical and mechanical properties
3.2. Procesi recikliranja MDF ploča i njihova fizička i mehanička svojstva

3.2.1 Tabular overview
3.2.1. Tablični pregled

The methods for recycling MDF can be summarised as follows (Table 2). The various digestion processes mentioned here are explained in more detail.

3.2.2 Thermomechanical and thermohydrolytic pulping processes

3.2.2. Termomehanički i termohidrolitički procesi proizvodnje pulpe

Krzysik *et al.* (1997) used a thermomechanical pulping process for the production of MDF, specifically processing chips from construction and demolition wood (species: spruce, pine, fir) and paper. The chips were fibrillated on a Sprout-Bauer refiner at a steam temperature of 77 °C and a disc gap of 0.3 mm for ½ minute. The bending strength of the test boards decreased with increasing board thickness, with only 6 mm thick boards meeting the standard. However, the bending modulus largely complied with or exceeded the standard. The authors specifically referenced the ANSI A208.2 standard for MDF, which was the prevalent guideline for MDF properties in North America at the time of their research. Similarly, the internal bond strength decreased with increasing board thickness, with none meeting the standard. Thickness swelling did not meet the requirements for any thickness. However, it was observed that the fibres could be easily wetted with all adhesives, indicating the potential for the production of MDF from these waste materials. With regard to the influence of parameters on hydrolysis, it should be noted that the degradation of cured UF resins depends on their chemical structure and degree of cross-linking (Elbert, 1995; Fleischer and Marutzky, 2000). This includes the fact that UF resins with a high F:U molar ratio have a higher resistance to hydrolysis than those with a low F:U molar ratio (Lubis *et al.,* 2018a). As adhering residual resin remains a core issue in recycling, Lubis *et al.* (2018b) focused on resin removal from recycled fibres and found that hydrolysis time has less influence on cured UF resins than temperature or molar ratio. Under hydrolysis (neutral and alkaline pulping), residual resins are not completely removed as they remain in the cell walls of the fibres (approximately 23-59 %). Acid or chemical pulping, on the other hand, can remove resins much better (more than 74 %).

Buschalsky and Mai (2021) used MDF with a relatively low F:U molar ratio (1.11) for recycling. UF-bonded MDF was thermohydrolytically pulped in an autoclave with water at 95 °C for 20-30 min and

Table 2 Overview of literature focusing on recycling of waste wood into MDF. The literature has been categorised according to the origin of the waste wood, the pulping process and the adhesives suitable for the pulping process.
Tablica 2. Pregled literature o recikliranju otpadnog drva u MDF ploču. Literatura je kategorizirana na temelju podrijetla otpadnog drva, procesa proizvodnje pulpe i ljepila (UF – urea formaldehid; PF – fenol formaldehid) prikladnoga za proces proizvodnje pulpe.

Wood-composite *Kompozitno drvo*	**References** *Literatura*	**Waste wood origin** *Podrijetlo drvnog otpada*	**Digestion process** *Proces digestije*	**Adhesive** *Ljepilo*
Medium density fibreboard *srednje gusta vlaknatica*	Krzysik *et al.,* 1997	Construction demolition wood / *konstrukcijsko drvo od rušenja*	Thermo-mechanical *termomehanički*	varying / *promjenjivo*
	Franke and Roffael, 1998a	MDF	Thermohydrolysis *termohidroliza*	UF
	Kirchner and Kharazipour, 2002	MDF	Thermohydrolysis *termohidroliza*	UF
	Buschalsky and Mai, 2021	MDF	Thermohydrolysis *termohidroliza*	UF
	Savov *et al.,* 2023	MDF	Thermohydrolysis *termohidroliza*	UF
	Mantanis *et al.,* 2004	MDF	Thermo-hydrolytic-mechanical *termohidrolitičko-mehanički*	UF
	Ihnát *et al.,* 2018	MDF, Chipboard, OSB	Thermo-hydrolytic-mechanical *termohidrolitičko-mehanički*	UF, MUF
	Lubke *et al.,* 2020	Chipboard	Thermo-hydrolytic-mechanical *termohidrolitičko-mehanički*	UF
	Wan *et al.,* 2014	MDF, Chipboard, OSB	Steam explosion *parna eksplozija*	UF, PF
	Dix *et al.,* 2001	MDF	Chemical decomposition *kemijska dekompozicija*	UF, MUF, PF, pMDI
	Ateş *et al.,* 2023	MDF	Chemical decomposition *kemijska dekompozicija*	UF
	Moezzipour *et al.,* 2018	MDF	Ohmic heating *omsko zagrijavanje*	UF
	Elias and Bartlett, 2018	MDF	Micro-release-process *process mikrootpuštanja*	UF
	Ateş *et al.,* 2023	MDF	Microwave-process *mikrovalni proces*	UF

processed into test panels. The disintegration process required significantly lower temperatures and was significantly shorter than in previous studies. Nitrogen content analysis showed that the recovered fibres contained approximately 30 % of the original UF. The density of the test boards was higher than the target density and the density of the reference boards. Thickness swelling was reduced by 50 % after the first recycling, by 25 % after the second recycling for 10 mm boards and by 40 % for 16 mm boards compared to the reference. Internal bond strength was identical to the reference after the first recycling and increased by 64.5 % for 10 mm MDF after the second recycling, which the scientists attributed to a slight reduction in fibre length that can occur during recycling, resulting in increased surface area and better adhesion (Back, 1987). However, there is a reduction in flexural strength with fibre shortening (Myers, 1983). In addition, a high proportion of fines contributes to a reduction in the modulus of elasticity (Dix *et al.,* 2001).

Ateş *et al.* (2023) used the WKI method and found that some fibres were extremely thick. In addition, the investigations showed a decrease in flexural strength and an increase in thickness swelling. The main reason for this is the presence of glue and paraffin residues on the recycled fibres. These microscopic residues can remain on the surface of the recycled fibres and even reduce the bonding ability of the fibres by binding to the -OH groups in the fibres, which could also affect the decrease in internal bond strength described in previous works (Zeng *et al.,* 2018). A significant increase in recycled fine fibres was also observed. There was also some decrease in fibre fractions with lengths greater than 1.24 mm, which can be classified as coarse fibres.

3.2.3 Thermohydrolytic-thermomechanical pulping processes

3.2.3. Termohidrolitičko-termomehanički procesi proizvodnje pulpe

Mantanis *et al*. (2004) attempted to industrialise a thermohydrolytic-thermomechanical pulping process for MDF with the aim of increasing the proportion of recycled fibre in production to over 20 %. MDF was pre-shredded into chips using a hammer mill and mixed with fresh wood chips at a ratio of 3:1. Impurities were removed by metal detection, screening and washing. The refining time in the refiner was 3-3 1/2 minutes at a pressure of 8.2 mbar with a fibre throughput of 5.0 t/h. It was found that the process could be used in production lines with single stage presses for E2 class MDF products. At 25 % waste, an internal bond strength of 0.60 N/mm² was achieved, which was more than half that of the reference boards. The addition of an unknown additive at 10 % resulted in an almost complete improvement. Similar behaviour was observed for flexural strength. Thickness swelling could be reduced by using waste boards. It is reported that even at a substitution level of 25 %, and without changing the press cycle and productivity, the new process resulted in "recycled" MDF boards with properties far exceeding European standards.

In studies by Roffael *et al.* (2010), MDF waste was pre-crushed and subjected to thermo-hydrolytic treatment at 170 °C for 5 minutes, followed by fibrillation in the refiner. Changes in the morphology of TMP (thermomechanically pulped fibres) from used particleboard and fibreboard to TMP from virgin wood were observed. Recycled TMP had a shorter fibre length and a higher fine fraction within the recovered fibres. TMP from wood can be replaced up to 30 % by TMP from used chipboard and fibreboard in the production of UF resin-bonded MDF without significant impairment of the mechanical-technological properties of the boards produced.

Ihnat *et al.* (2018) recycled a mixture of particleboard, MDF, and OSB into fibre. In addition to the usual UF bonded particleboard and MDF, Melamine-Urea-Formaldehyde Resin (MUF) bonded particleboard, MDF and OSB were also used. The MUF bonded samples were first treated in boiling water at 180 °C to induce swelling. Adhesive bridges were then broken by pressing and the samples were re-comminuted using a Palmann drum chipper. Finally, the material was fibrillated on a Sprout-Waldron refiner and refined on a Valley-Hollander. UF bonded particleboard was boiled for 30 minutes to a moisture content of 40 % and MDF for 3 minutes to a moisture content of 45 %. The same process technology was then used as for the MUF bonded wood materials, except for the pressing step. The fibres obtained were then characterised and not processed into laboratory boards. Chipboard waste with UF bonding has a high proportion of long fibres, while that from MUF-bonded chipboard and OSB has a high proportion of fine fibres, due to the greater brittleness of the MUF-wetted chips. Fibres obtained from MUF-based MDF waste are suitable for MDF production immediately after processing.

Lubis *et al.* (2018b) produced recycled fibres from MDF waste using a thermohydrolytic-mechanical treatment (steam treatment: 30 min at 180 °C and 600 kPa) for laboratory boards with different recycled fibre content. After steam treatment, the fibres were comminuted in two batches using a hammer mill (1500 rpm) and a refiner (atmospheric pressure; 1000 rpm; plate gap 1 mm). The shorter fibre lengths and higher fine fraction within the recycled fibres compared to virgin fibres observed by Roffael *et al.* (2010) were confirmed. The highest internal bond strength occurred at 10 % recycled fibre content, regardless of pulping method and wood species, after which the internal bond strength decreased. Mechanical properties, including flexural strength and modulus, showed a similar trend with increasing recycled fibre content. Thickness swelling and water absorption decreased with increasing recycled fibre content. It is recommended that 10 % recycled fibre can be substituted for fresh fibre without compromising properties.

Lubke *et al.* (2020) used a thermohydrolytic mechanical pulping process to produce fibres for MDF from particleboard waste. The waste was first shredded and then boiled in water for 30 minutes to achieve a moisture content of at least 40 %. A Sprout-Waldron disc refiner was then used to process the chips into fibres in a single pass through the refiner's grinding heads. Several recycling processes were carried out and thermomechanical pulping, targeting a 13°SR-24°SR range, was found to be suitable for the production of fibreboard from recycled chips. The °SR, or Schopper-Riegler number, is a measure of the drainage rate of pulp suspension and indicates the degree of pulp refining. It directly relates to the fibre properties and their suitability for MDF production. A higher °SR value indicates a slower drainage rate and typically corresponds to more refined fibres with better bonding potential, which is crucial for achieving the desired strength properties in the final MDF product. The final stages of cascade utilisation in this material stream demonstrated that the properties met the lowest levels required by the applicable standards.

Following Savov *et al.* (2023), thermal hydrolysis was carried out in an autoclave using saturated steam as the heat transfer medium. In contrast to previous studies, MDF was used in samples with dimensions similar to those of wood chips. The influence of the hydrolysis process parameters, namely process time and temperature, on the properties of the recycled MDF wood fibre was investigated. Hydrolysis temperatures of 121 °C and 134 °C were used, with three durations for each temperature - 30, 45 and 60 minutes. After hydrolysis, the resulting fibre fraction was refined using a hammer mill. The fractional and elemental composition of the recycled fibres obtained was evaluated. Consistent with the findings of Roffael *et al.* (2010) and Lubis *et al.* (2018b), the recycled MDF fibres had a similar fibre morphology and fractional composition to the fresh fibres but were shorter in length.

3.2.4 Steam explosion and chemical digestion processes

3.2.4. Parna eksplozija i procesi kemijske digestije

Wan *et al.* (2014) used a steam explosion process to produce and evaluate fibres for the production of MDF from waste particleboard, MDF (UF-bonded) and OSB (PF-bonded). The recycled material was first impregnated with 0.5 % butane tetracarboxylic acid in the expectation of complete dissolution of the chips and fibres. According to the literature, water vapour at 235 °C acts on a vessel. The pressure difference between the pressure vessel and the atmosphere creates an explosion effect at the outlet of the pressure vessel, which breaks the material back into fibres. It has been found that recycled fibres from the steam explosion process are 30 % shorter than virgin industrial fibres. When particleboard is used as the raw material for recycled fibres, the fibre length increases compared to MDF fibres, but remains shorter than industrial fibres. The length of fibres from OSB, on the other hand, exceeds that of industrial fibres. According to the report, particleboard and OSB have a high potential for fibre production by steam explosion.

According to Dix *et al.* (2001), urea resin bonded MDF coated with paper was digested in a sodium hydroxide solution (10 % NaOH based on atro MDF) at 200 °C for 2 hours. The digestion process resulted in an increase in bending and transverse tensile strength when 30 % recycled fibre content was used. At 50 % recycled fibre content, the changes in properties are small. Thickness swelling is significantly reduced when the chemically recovered fibres are used, so that the test panels meet the requirements of EN 622-5 even without the use of hydrophobing agents. In a study by Ateş *et al.* (2023), acid hydrolysis was used to recycle MDF using formic acid and phosphoric acid. The MDF samples to be recycled were heated and mixed in a 25 % acid solution. The acid hydrolysis treatment resulted in deformation of the boards during hot pressing and damage to the fibres. Acid hydrolysis severely damages the fibres and should not be used for the recycling of wood-based materials.

In addition, chemical treatment of MDF waste with sodium sulphite and sodium hydroxide reactivated the cured resin, eventually leading to low resin content in recycled MDF (Nakos *et al.,* 2001).

3.2.5 Ohmic heating and micro-releasing processes

3.2.5. Omsko zagrijavanje i procesi mikrootpuštanja

A new ohmic heating process was developed specifically for MDF recycling by Moezzipour *et al.* (2018). For a typical thermo-hydrolytic-mechanical reference recycling process, shredded MDF waste from industry was steamed in a laboratory autoclave at 105 °C for 150 min at 4 bar pressure. The steamed waste was then defibred in a refiner. To recycle MDF waste using an electrical process, shredded MDF waste was soaked in warm water for 30 minutes and then poured into a chamber containing salt water (electrolyte). Finally, an electric current was passed through the mixture, releasing the fibres from the MDF waste.

In terms of the previously described change in fibre geometry due to thermohydrolytic digestion, the fibres from the ohmic heating process were longer and similar to the standard fibres from Iranian MDF mills, as less fibre degradation occurs in the electrical heating process due to the removal of pressure and the long heating time in the autoclave without a refining stage in the electrical heating process. In addition, the lignin and hemicellulose content was found to be lower than that of the reference fibres, especially in thermohydrolysis pulps. No degradation of cellulose was observed. The thickness swelling of the recycled thermohydrolysis treated MDF was higher than that of the reference. Recycled MDF from the electric heating process had lower swelling than that from thermohydrolysis.

The *MOR* of the panels produced using the electrical heating method exceeded the EN standard, while the panels produced using the thermohydrolytic method did not meet the requirements. It is very clear that the modulus of elasticity of the recycled fibre panels produced by the electrical heating method was higher than that of the panels produced by the thermohydrolytic method. Even the *MOE* values of the MDF boards produced exceeded the requirements of the relevant standard specifications (EN310). The internal bonding (IB) of the recycled boards from the thermohydrolytic treatment was lower than that of the reference.

Elias and Bartlett (2018) report on the micro-release process, which works in a similar way to the ohmic heating process: The MDF waste is shredded and separated, immersed in 98 °C water for 5 minutes, which triggers microwaves that cause the fibres to swell. Finally, the recycled fibres can be used as raw material to replace part of the virgin fibres and produce recycled MDF. The samples separated for recycling by the micro-separation method were kept in boiling distilled water for 5 minutes before swelling for 5 minutes at medium wave frequency in an industrial microwave oven. Complete defibration was performed manually. The microwave process had no effect on fibre shortening. These trials in an industrial plant successfully demonstrated that the addition of up to 20 % recycled material can be achieved without adverse effects.

3.3 Changes in physical and mechanical properties of recycled particle and fibre boards

3.3. Promjene fizičkih i mehaničkih svojstava recikliranih ploča iverica i vlaknatica

The recycling of particleboard and fibreboard usually results in a deterioration of the physical and mechanical properties of the new wood-based materials produced from it compared to the original material. Some processes also report improvements in these properties for individual characteristics. Various sources provide information on whether and how the physical and mechanical properties of recycled boards change. Tables 3 and 4 give an overview of the changed physical and mechanical properties of particle- and fibreboard according to the studies of the authors mentioned.

All the articles dealing with the morphology of the particles state that the particle size changes as a result of recycling, which has a particular effect on the bending strength. The change in the degree of thinness, particularly in the case of chips, and the shortening of the fibre length should be mentioned here. The cause of this significant reduction in bending strength values can be explained by the effect of hydrothermal heating on the thermal degradation of wood components, in particular the reduction of lignin (Moezzipour *et al.,* 2018). Yilgor *et al.* (2001) also found that the decrease in mechanical properties was mainly due to the thermal decomposition of wood during hydrothermal treatments, which is confirmed by the work of Franke and Roffael (1998a) and Lykidis and Grigoriou (2008) on thermohydrolysis. The shortening of chips and fibres

Table 3 Overview of change in physical and mechanical properties for particleboard recycling (*IB* – internal bonding, *MOR* – modulus of rupture, *MOE* – modulus of elasticity, *TS* – thickness swelling, / – not mentioned)
Tablica 3. Pregled promjena fizičkih i mehaničkih svojstava iverice (*IB* – *čvrstoća* na raslojavanje, *MOR* – modul loma, *MOE* – modul elastičnosti, *TS* – debljinsko bubrenje, / – nije navedeno)

References *Literatura*	**Particle size** *Veličina iverja*	***IB***	***MOR***	***MOE***	***TS***
Lykidis and Grigoriou, 2008	reduction *smanjenje*	reduction *smanjen*	reduction *smanjen*	improvement *poboljšan*	increase *povećan*
Lykidis and Grigoriou, 2011	/	reduction *smanjen*	reduction *smanjen*	improvement *poboljšan*	increase *povećan*
Zamarian *et al.,* 2017	reduction *smanjenje*	improvement *poboljšan*	reduction *smanjen*	comparable *usporediv*	decrease *smanjen*
Azambuja, 2018	reduction *smanjenje*	improvement *poboljšan*	reduction *smanjen*	comparable *usporediv*	decrease *smanjen*
Iždinský *et al.,* 2020	/	reduction *smanjen*	reduction *smanjen*	reduction *smanjen*	decrease *smanjen*
Lubke *et al.,* 2020	/	reduction *smanjen*	reduction *smanjen*	improvement *poboljšan*	/

Table 4 Overview of change in physical and mechanical properties for MDF recycling (IB = internal bonding, MOR – modulus of rupture, MOE – modulus of elasticity, TS – thickness swelling, / – not mentioned)
Tablica 4. Pregled promjena fizičkih i mehaničkih svojstava ploče vlaknatice (IB – *čvrstoća* na raslojavanje, MOR – modul loma, MOE – modul elastičnosti, TS – debljinsko bubrenje, / – nije navedeno)

References *Literatura*	**Particle size** *Veličina vlakana*	**IB**	**MOR**	**MOE**	**TS**
Krzysik *et al.*, 1997	/	reduction *smanjen*	reduction *smanjen*	improvement *poboljšan*	increase *povećan*
Buschalsky and Mai, 2021	/	improvement *poboljšan*	not significant *bez značajne promjene*	not significant *bez značajne promjene*	decrease *smanjen*
Ateş *et al.*, 2023	reduction and high proportion of fines *smanjena, uz visok udio finih vlakana*	reduction *smanjen*	/	improvement *poboljšan*	decrease *smanjen*
Mantanis *et al.*, 2004	/	comparable with additive used *usporediv s upotrijebljenim aditivom*	comparable with additive used *usporediv s upotrijebljenim aditivom*	/	decrease *smanjen*
Roffael *et al.*, 2010	reduction and high proportion of fines *smanjena, uz visok udio finih vlakana*	reduction over 30 % recycled fibre content *smanjen za više od 30 % recikliranih vlakana*	reduction over 30 % recycled fibre content *smanjen za više od 30 % recikliranih vlakana*	reduction over 30 % recycled fibre content *smanjen za više od 30 % recikliranih vlakana*	reduction over 30 % recycled fibre content *smanjen za više od 30 % recikliranih vlakana*
Ihnat *et al.*, 2018	reduction depending on the raw material *smanjena ovisno o vrsti sirovine*	/	/	/	/
Lubis *et al.*, 2018b	reduction and high proportion of fines *smanjena, uz visok udio finih vlakana*	reduction *smanjen*	reduction *smanjen*	reduction *smanjen*	decrease *smanjen*
Lubke and Ihnát, 2020	/	slight reduction *blago smanjen*	slight reduction *blago smanjen*	slight reduction *blago smanjen*	slight increase blago povećan
Savov *et al.*, 2023	reduction and high proportion of fines *smanjena, uz visok udio finih vlakana*	/	/	/	/
Wan *et al*., 2014	increase *povećana*	/	/	/	/
Dix *et al.*, 2001	/	reduction over 50 % recycled fibre content *smanjen za više od 50 % recikliranih vlakana*	reduction over 50 % recycled fibre content *smanjen za više od 50 % recikliranih vlakana*	/	decrease *smanjen*
Moezzipour *et al.*, 2018	comparable *usporediva*	improvement *poboljšan*	improvement *poboljšan*	improvement *poboljšan*	decrease *smanjen*

can be beneficial in terms of transverse tensile strength, which has been shown in some studies to be improved up to a certain level of recycled material, which the scientists attribute to an increase in surface area and better adhesion.

However, there is controversy regarding the transverse tensile strength, e.g. Zeng *et al.* (2018) describe that adhering paraffin and glue residues reduce the transverse tensile strength and also the flexural strength, which may explain the problems with internal bonding (IB) above a certain proportion of recycled particles. Hydrothermal degradation of hemicelluloses may also play a role. Hemicelluloses are responsible for forming hydrogen bonds with the resin. The degradation of hemicelluloses means fewer hydroxyl groups, which reduces the potential for bonding between wood and resin (Moezzipour *et al.,* 2018).

The reason for the high thickness swelling of MDF recycled by the hydrothermal method could be the removal of lignin from the fibre surface due to the effect of heat and the increased fibre swelling. In fact, the intense heat of the hydrothermal process leads to an increase in fibre hygroscopicity (Lykidis and Grigoriou 2008; Michanickl and Boehme 2003). Fibre length plays an important role in the dimensional stability of MDF boards. Therefore, a greater reduction in fibre length will result in increased thickness swelling of MDF boards. As mentioned above, fibres from electrical heating are significantly longer than fibres from hydrothermal heating. The reason for the significant reduction in the bending test parameters (MOR and MOE) in the boards produced with fibres from the hydrothermal process can be explained by the influence of hydrothermal heating on the thermal degradation of the wood components, in particular the lignin degradation (Table 4). Lignin plays a very important role in the self-bonding mechanism, and lignin condensation reactions that occur within and between fibres during hot pressing can improve the properties of MDF boards (Okuda *et al.,* 2006). Zhang *et al.* (2013) reported that the reduction in lignin content leads to a reduction in the modulus of elasticity of individual fibres, which may affect the quality of the boards produced. Yilgor *et al.* (2001) found that the decrease in mechanical properties is mainly due to thermal decomposition of the wood during hydrothermal treatment, confirming the work of Franke and Roffael (1998a) and Lykidis and Grigoriou (2008) on thermohydrolysis. It is assumed that the UF resin is hydrolysed again by the residual acid during hot pressing, as described by Lubis *et al.* (2018a).

Indeed, the lower levels of hemicelluloses in the hydrothermally recycled fibres compared to the electrical heating method (Table 4) lead to lower IB values in the boards made from them. Hemicelluloses are responsible for the formation of hydrogen bonds with the resin. The reduction of hemicelluloses means fewer hydroxyl groups, which reduces the potential for the formation of bonds between wood and resin (Popović *et al.,* 2015). The IB strength of MDF can be influenced by natural bonds formed by hydroxyl groups of hemicelluloses.

3.4 Literature-based approaches to increasing the proportion of recycled content in particle and fibreboard production

3.4. Pristupi povećanju udjela recikliranog materijala u proizvodnji ploče iverice i vlaknatice utemeljeni na literaturi

Waste wood is currently mainly used in the production of particleboard. As the proportion of recycled material is usually only between 20 and 30 % due to the process, the problems identified need to be addressed. By evaluating and supplementing the results of the literature studies, the project “ReSpan - Recycling of particleboard materials”, on which this article is based, has derived the following approaches to potentially increase the proportion of recycled fibre in the production of wood-based materials:

- Improving sorting technology: The use of advanced sorting technologies can improve the efficiency of waste wood processing and lead to more homogeneous and higher quality recycled material. This step could be further supported by more efficient disposal and collection systems.
- Use of alternative adhesives: The use of alternative, more environmentally friendly adhesives can reduce formaldehyde emissions from recycled wood-based materials. In the future, it would certainly make the most sense to use binders that are easier to degrade or reactivate. This could revolutionise future recycling processes and significantly improve the sustainability of the wood processing industry. In this context, it would also be interesting to know whether the bond strength can be increased by using bioadhesives. The use of pMDI could also be useful for the adhesive problem, as the binder is known to form strong bonds and could compensate for the problem of residual adhesive adhesion.
- Optimisation of process technology: It would also be conceivable to adapt the production processes for recycled fibreboards to include a combination of recycled fibres in the core layer and high-quality materials in the outer layers. One step would be to adapt the process technology to produce particles with a suitable geometry. In addition, the development of alternative processes would be a sensible step. For example, solvolytic processes that keep the used adhesive reactive in a similar way to existing chemical processes would make sense.
- Use of a mixture of recycled OSB, particleboard and plywood for the production of MDF: This could initially expand the range of applications within the waste wood classes. As described in the article, chipboard and MDF particles are well suited for recycling. The materials could be pre-shredded, thermohydrolytically treated and then thermomechanically defibrated, possibly destroying the glue bridges from MUF and PF bonding. The fibres could then be used to make MDF.

4 CONCLUSIONS

4. ZAKLJUČAK

The literature review has provided a comprehensive overview of the state of the art in particleboard and MDF recycling technology, particularly with regard to pulping processes and the resulting physical and mechanical properties. Despite significant progress, no large-scale industrial recycling processes have been fully established, mainly due to challenges such as degradation of physical and mechanical properties and inefficiencies of current recycling methods. Most research and practical applications still focus on the degradation of UF resins, while emerging techniques such as ohmic heating, as investigated by Moezzipour *et al.* (2018), show potential for improving mechanical properties.

Future advances in this area could benefit from the development of adhesive systems designed for easier degradation and reactivation, supporting circular economy goals. In addition, optimising process technology - possibly by combining different recycled wood types such as OSB, particleboard and plywood in MDF production - could create more versatile applications. Improved sorting technologies and a more efficient collection system for waste wood materials could also lead to more homogeneous recycled material quality, overcoming current limitations in physical and mechanical properties.

While the post-millennium literature provides valuable insights and advances, a shift towards industry-ready recycling processes is still needed to effectively meet sustainability and recycling goals. Collaborative efforts between research and industry can help bridge this gap and provide a robust foundation for future large-scale recycling systems that are better aligned with environmental goals and regulatory standards.

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Synthetic Adhesives in Production of Engineered Wood Products: A Review

Sintetička ljepila u proizvodnji kompozitnog drva: pregled literature

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ABSTRACT • *Adhesives used in the production of wood composite materials, including engineered wood, are a significant component that characterizes the performance of the final products. Also, the adhesive type, quantity and properties significantly impact the cost of the resulting materials. The aim of this review was to present some of the basic characteristics of bonding agents for engineered wood. Based on the information provided, a suitable adhesive can be selected to form final products that meet the needs of consumers. Therefore, it is of essential importance to know the advantages and disadvantages of adhesives, since global trends require the creation of environmentally friendly products. The latter must contain a minimum amount of free formaldehyde. The object of this review was to deliver an analysis of the most commonly used synthetic resins in the production of engineered wood products (EWPs), namely amino based, phenolic based, isocyanate, polyurethane and polyvinyl acetate.*

KEYWORDS: *bonding agents, construction industry, composite materials, properties*

SAŽETAK • *Ljepila koja se rabe u proizvodnji kompozitnog drva važna su komponenta koja utječe na svojstva gotovih proizvoda. Osim toga, vrsta, količina i svojstva ljepila uvelike utječu na cijenu proizvedenih materijala. Cilj ovog rada bio je predstaviti neka osnovna svojstva ljepila za kompozitne materijale na bazi drva. Na temelju iznesenih informacija može se odabrati prikladno ljepilo za izradu proizvoda koji zadovoljavaju potrebe potrošača. Stoga je iznimno važno znati prednosti i nedostatke ljepila jer globalni trendovi zahtijevaju izradu ekološki prihvatljivih proizvoda. Oni moraju sadržavati minimalnu količinu slobodnog formaldehida. Cilj ovog rada bio je provesti analizu najčešće upotrebljavanih sintetičkih smola u proizvodnji kompozitnog drva (EWP) na bazi aminokiselina i fenola, izocijanata, poliuretana i polivinil acetata.*

KLJUČNE RIJEČI: *vezivo; građevna industrija; kompozitni materijali; svojstva*

1 INTRODUCTION
1. UVOD

All areas of our lives are developing, and this leads to a growing share of the construction industry (FAO, 2022). The main components in the creation of the structural elements of these facilities are bricks, concrete, steel, but in recent years wood is also increasingly used (Švajlenka and Pošiváková, 2023).

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Wood as a structural element has always been one of the most popular, practical and economical building materials (Senft, 2002). Even today, wood constructions are preferred not only because of the lower financial investment, but also because of their exceptional resistance to extreme climatic changes. The choice of wood used in the construction industry depends on several factors, such as price, distribution according to the region, durability, environmental impact, etc. In addition, it is crucial to consider the specific structural elements and the aesthetic characteristics of the housing or facilities being built. The most widely used species in the construction industry are coniferous species, of which Scots pine (*Pinus sylvestris* L.) accounts for a significant share. The wood of this species is mainly used to produce glued-laminated timber (GLT), cross-laminated timber (CLT), partition walls, doors, windows, flooring, etc. The reason for this is that Scots pine is distinguished by a beautiful texture, high resistance to mechanical loads, relatively low density; furthermore, it is easy to process and economically advantageous (Mederski *et al.,* 2015; Mclean, 2019; Burawska-Kupniewska *et al.,* 2020). Wood from the *Quercus* genus or other hard deciduous or tropical species is preferred in constructing heavy-duty structures where greater durability is required.

On the other hand, the aesthetic appearance of wood as a natural material leads to a feeling of high comfort and tranquillity for the inhabitants. Along with all the advantages of wood, there are also numerous disadvantages, primarily related to its anatomical structure, which leads to significant anisotropy. When using wood in load-bearing constructions, these disadvantages have a significant negative impact on their mechanical performance. Modern technologies allow many wood disadvantages to be overcome by removing areas of concentrated influence and subsequent bonding using synthetic adhesives (Akiner *et al.,* 2022; Bayat, 2023).

2 TYPES OF SYNTHETIC ADHESIVES IN WOODWORKING, FURNITURE AND CONSTRUCTION INDUSTRY

2. VRSTE SINTETIČKIH LJEPILA U DRVOPRERAĐIVAČKOJ I GRAĐEVNOJ INDUSTRIJI TE INDUSTRIJI NAMJEŠTAJA

The woodworking industry is the largest consumer of adhesives worldwide, and 70 % of the total volume of adhesives produced is applied in this sphere (Sandberg, 2016).

Adhesives used for producing engineered wood must be sufficiently stable in water, resistant to fungal attack and have high shear strength. Synthetic-based adhesives that meet the above requirements are primarily used in manufacturing this type of product. These resins are toxic to one degree or another due to the release of formaldehyde emissions into the atmosphere. For this reason, depending on the application of the glued wood materials, the use of the appropriate type of adhesive is recommended. The reduction of the free formaldehyde contained in the bonded products is an essential necessity, as it largely determines the environmental friendliness of the final product. According to European standard EN 717-1 (CEN 2004) for emission class E1, this indicator should be ≤ 0.124 mg/m^3 air (Sung *et al.,* 2012; Gonçalves *et al.,* 2021; Chrobak *et al.,* 2022).

The correct choice of the type and composition of the adhesive determines not only the strength and durability of the adhesive joint, but also the price of the products manufactured. Therefore, the choice of adhesive in a specific case must be consistent not only with the factors to which the structure will be exposed but also ensure low cost and high operational strength (Pizzi, 2003).

In this case, the adhesives join the wood elements in a bond that resembles or is close to the bond between the wood fibres in their natural state. Wood materials obtained in this way should not be inferior in terms of physical and mechanical parameters to natural wood of the same dimensions (Ülker, 2016).

The adhesive used to obtain EWPs in the constructions must ensure the structure durability for a long period. In most cases, the operation of these structures takes place outdoors under the influence of humidity and/or dryness of the air, as a result of which the wood swells or dries up. These constructions are exposed to significant changes in temperature fluctuations and the adhesives used for their production must withstand these changes (Senft, 2002). The so-called predicted assessment of the resistance of these joints also plays an essential role in solving problems of engineered wood durability.

This assessment provides information about the lifespan of the structures built. It further shows that the obtained values for the durability of the bonded products meet the requirements, and the possibilities for improving the connections and adhesives used (Hänsel *et al.,* 2021).

In the given case, the durability of the glued wood means its ability to store the adhesion and cohesion bonds over a long period until the minimum permissible values are reached or until it is destroyed.

2.1 Classification of synthetic wood adhesives

2.1. Klasifikacija sintetičkih ljepila za drvo

Synthetic adhesives used in the woodworking, furniture and construction industry can be classified based on several criteria (Dunky and Pizzi, 2002; Ormondroyd, 2015). Depending on the production method, adhesives are divided into polymerization and polycondensation types. In polymerization adhesives, the molecules of the monomer are linked to form macromolecules, resulting in a polymer while maintaining the chemical composition of the compound. The release of a low-molecular product does not accompany the polymerization process. Polymerization adhesives include polyvinyl acetate, polyvinyl chloride, polyacrylic, etc.

In the second case, polycondensation involves bonding two or more monomers into a higher molecular weight product, during which low molecular weight by-products, such as water, ammonia, hydrogen chloride, etc., are released. Polycondensation adhesives include phenol-formaldehyde, urea-formaldehyde, melamine-formaldehyde, etc.

Depending on the water resistance of the adhesive joints, adhesives are classified as follows: non-water-resistant (polyvinyl alcohol and polyvinyl acetate adhesives), medium water resistant (urea-formaldehyde adhesives), and high-water resistant (phenol-formaldehyde, melamine-formaldehyde, and resorcinol-formaldehyde adhesives).

According to their intended use, adhesives are divided into two groups:

- Structural: They find application for products and structures subjected to high mechanical stress, such as multi-layer glued load-bearing beams, columns, arches, solid wood panels, etc. Adhesives for the manufacturing of structural glued wood products must comply with the relevant European standards: EN 301 (CEN 2023), EN 15425 (CEN 2023) and EN 16254 (CEN 2023), as well as the harmonized standards defined by the European Union's Construction Products Regulation (CPR) to assess the performance of construction products: EN 14080 (CEN 2013), EN 14374 (CEN 2004) and EN 1995-1-1 (CEN 2004).
- Non-structural: In these cases, the adhesive joint is not designed to withstand significant loads. Therefore, these adhesives are mainly used in the furniture industry, such as furniture, decorative elements, packaging, toys, kitchen utensils, etc. The main standards that non-structural adhesives must comply with are ISO 19209 (ISO 2017), EN 12765 (CEN 2016) and EN 17619 (CEN 2021).

Synthetic adhesives can be categorized into liquid, paste-like, powdery, and sheet forms based on their appearance before use.

Depending on the curing mechanism, adhesives can be divided into thermoplastic and thermosetting. Thermoplastics are polymers with a linear structure that can be melted when heated in a solid state and can cure upon cooling. This process is reversible. In contrast, thermosetting polymers have a two-or three-dimensional cross-linked structure that becomes solid, infusible, and insoluble upon heating. In this case, the process is irreversible.

The most widely used adhesives in the woodworking industry are thermosetting polymeric condensation synthetic resins, which form adhesive joints with very good physical and mechanical properties.

2.2 Most commonly used adhesives for the production of EWPs

2.2. Najčešće upotrebljavana ljepila za proizvodnju kompozitnog drva

The most commonly applied thermosetting resins for the manufacturing of EWPs are amino resins, phenolic resins, isocyanates, and polyurethane adhesives. The conditions for their synthesis and curing are of particular importance. Upon heating, this group of polymers transforms into cross-linked solid materials that cannot be further processed. Cross-linking occurs through the formation of a significant number of covalent bonds, creating infusible and insoluble products.

A deep understanding of these processes will enable the optimization of the curing process by adding various substances. This, in turn, will create conditions to reduce the impact of harmful emissions, and enhance the properties of adhesive joints (Mo *et al.,* 2015; Liu *et al.,* 2018).

Common thermoplastic wood adhesives applied for the production of EWPs are polyvinyl acetate emulsions.

Based on this knowledge, opportunities for the production of environmentally friendly, reliable, and durable products will be created (Dorieh *et al.,* 2022a)

2.2.1 Phenol-formaldehyde (PF) adhesives

2.2.1. Fenol-formaldehidna (PF) ljepila

Phenol-formaldehyde oligomers are obtained by interacting with synthetic phenol or its derivatives (cresol, xylenol, and resorcinol) with formaldehyde (Berdnikova *et al.,* 2021). Phenol is condensed with formaldehyde in an alkaline medium with an excess of formaldehyde to obtain resole (thermosetting) oligomers. The ratio of phenol to formaldehyde used in resoles is between 1:1.5 and 1:2.2. Hydroxides of alkali (potassium, sodium) and alkaline earth metals (barium, calcium) as catalysts in the synthesis of resoles are applied. The reaction for the production of resoles primarily depends on changes in the reagent molar ratio, the reaction temperature and duration, as well as the type and amount of catalyst.

Resole PF resins are a mixture of linear and branched oligomers containing a large number of hydroxymethyl groups (–CH_2OH), which are capable of further transformations. Their curing is carried out by heating (130-150 °C) or in the presence of acid catalysts (benzenesulfonic acid, phosphoric acid). Upon heating, resole transforms into resitol and resite. These stages are accompanied by an increase in the molecular weight of the polymer and the formation of a three-dimensional cross-linked structure.

Novolac (thermoplastic) oligomers are synthesized in the presence of acid catalysts (such as hydrochloric acid, sulfuric acid, or oxalic acid) with excess phenol. As a result, novolac PF oligomers with a predominantly linear structure are formed and phenolic cores are connected by methylene bridges. Novolac resins are solid and soluble. Their curing occurs using urotropine, and less frequently, paraformaldehyde at temperatures between 150-180 °C. The production of PF resins is schematically shown in Figure 1 (Atta-Obeng, 2011).

These adhesives are characterized by very good adhesion to various materials, and high cohesive strength. Other significant advantages of these adhesives include the availability of raw materials and ease of synthesis (Berdnikova *et al*., 2021). The formed adhesive joint is distinguished by high bonding strength, heat resistance (up to 300-400 °C), water resistance (both in cold and hot water), and biological resistance.

One of the disadvantages of PF adhesives is their red-brown colour, which leads to staining of the adhesive joint. Fragile adhesive joints with high hardness are formed, making machining difficult and leading to the wear of the cutting tools. The significant disadvantage of this type of adhesive is the toxicity of phenol and formaldehyde, as well as the formaldehyde emissions. For this reason, bonding products that are used indoors are not recommended. The toxicity problem can be solved in various ways, one of which is the replacement of PFR with UFR or MFR resins.

Several authors have conducted studies on using various compounds known as formaldehyde scavengers that capture the released formaldehyde (Kristak *et al.,* 2022). Ramdugwar *et al.* (2022) investigated the addition of sodium sulphite and sodium metabisulfite

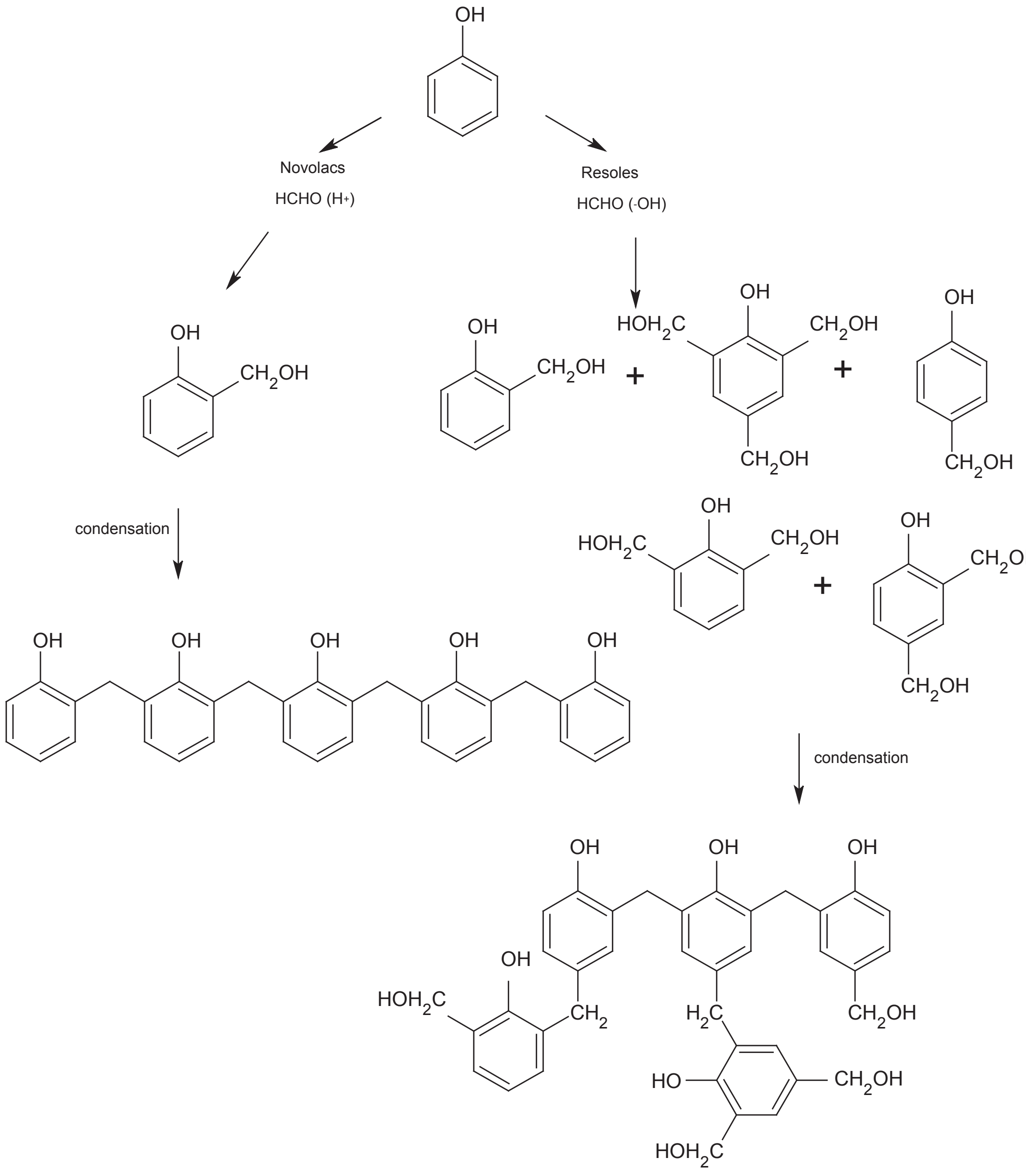


Figure 1 Synthesis of novolac and resole PF resin (Atta-Obeng, 2011)
Slika 1. Sinteza novolaka i rezol PF smole (Atta-Obeng, 2011.)

to phenolic resin to reduce formaldehyde emission. The authors found that these two substances significantly decrease formaldehyde content without affecting adhesive performance. Other authors considered the incorporation of tannin from the bark of *Acacia mangium* into the formulation of low molecular weight PF resin to absorb free formaldehyde. The resulting modified adhesive meets the standard requirements, and tannin can be used as a formaldehyde scavenger without compromising the strength of the adhesive joints (Hoong *et al.,* 2010).

2.2.2 Phenol-resorcinol-formaldehyde (PRF) and resorcinol-formaldehyde (RF) adhesives

2.2.2. Fenol-rezorcinol-formaldehidna (PRF) i rezorcinol-formaldehidna (RF) ljepila

Phenol-resorcinol-formaldehyde (PRF) adhesives are obtained by introducing resorcinol during the synthesis of PF resin. Substituting phenol with resorcinol leads to resorcinol-formaldehyde (RF) resin formation. These two types of adhesives are characterized by a significant reduction in curing time compared to PF resin. The chemical reactions associated with producing and curing of PRF and RF resins are similar to those for PF resin (Dunky and Pizzi, 2002; Durairaj, 2005; Wibowo and Park, 2024). Resorcinol is responsible for the accelerated curing at room temperature, both in the composition of RF and PRF resins. Pure RF oligomers are typically produced at a molar ratio of formaldehyde to resorcinol of less than one, in a neutral or slightly alkaline medium.

PRF adhesives are commonly prepared by reacting phenol with formaldehyde, resulting in the formation of a PF resole polymer with a predominantly linear structure. In a subsequent reaction, resorcinol in excess that reacts with the –CH2OH groups is added to produce the PRF polymers.

RF adhesives can be used for hot (150-180 °C) and cold gluing. Curing at room temperature occurs very rapidly, which leads to the development of stresses in the adhesive joint and to its faster failure. Therefore, it is preferred to operate at lower temperatures. In contrast to PF resins, these adhesives cure at room temperature in an acidic, neutral, or slightly alkaline medium.

RF adhesives form adhesive joints characterized by excellent adhesion, high water, heat, atmospheric and chemical resistance, and n on-toxicity. A key disadvantage of these adhesives is the high cost of resorcinol. Therefore, they are used in combination with PF resins. Another approach to reducing resorcinol content is to modify PF resins by grafting resorcinol onto the active hydroxymethyl groups of low condensation resoles obtained from the reaction of phenol with formaldehyde (Dunky and Pizzi, 2002).

2.2.3 Urea-formaldehyde (UF) adhesives

2.2.3. Urea-formaldehidna (UF) ljepila

The synthesis of urea-formaldehyde (UF) resin occurs in two stages (Kloeser *et al.,* 2007). The first stage involves the interaction between urea and formaldehyde in a weakly basic or neutral medium, primarily resulting in the formation of mono- and dihydroxymethylurea (Figure 2). In the second stage, condensation of the obtained products takes place, leading to the formation of low-molecular-weight polymers. At acidic pH, the molecular weight of the condensation products increases due to the formation of ether and methylene links.

UF adhesives are two-component adhesives and cure either upon heating or at room temperature. For cold curing, diluted solutions of inorganic acids (phosphoric, sulfuric) or organic acids (oxalic, phthalic) are used as hardeners. The quantitative proportion of the hardener is up to 5 % relative to the weight of dry resin.

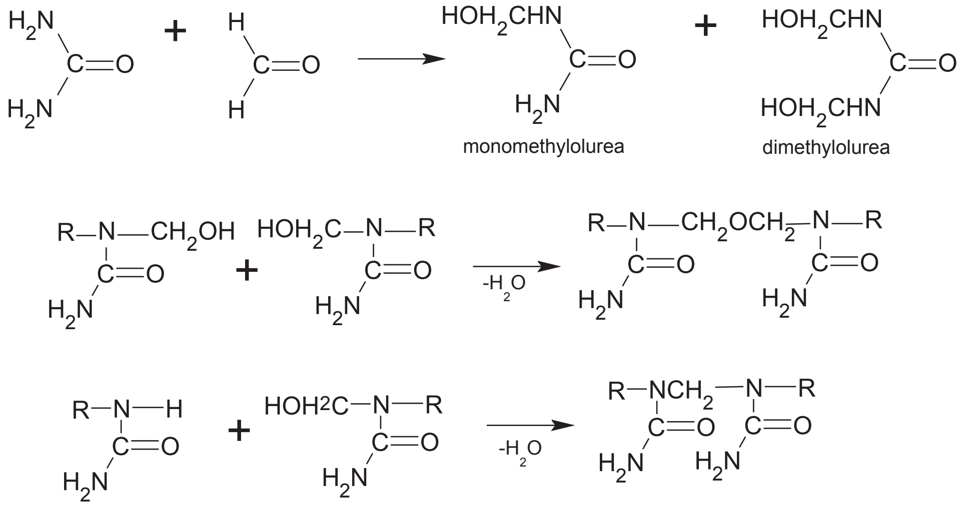


Figure 2 Condensation reactions for obtaining urea-formaldehyde resins
Slika 2. Reakcije kondenzacije za dobivanje urea-formaldehidnih smola

Salts of strong acids (ammonium chloride, ammonium sulphate, ammonium nitrate) in the form of 20 % solutions or as powder are employed for hot bonding. Their quantity is typically around 1 % relative to the weight of dry resin.

UF adhesives are inexpensive and readily available. They are characterized by good adhesion to wood, light resistance, and colourlessness. The adhesive joints have moderate atmospheric resistance and are resistant to microorganisms. These adhesives are easy to use and cure quickly, especially at high temperatures. UF adhesives exhibit good strength characteristics (Ülker, 2016; Antov *et al.,* 2021).

Despite the listed advantages, these adhesives also have some disadvantages. They are very sensitive to temperature and pH of the environment. They exhibit moderate water resistance and have a short shelf life. The adhesive joints are rigid and brittle, which can generate internal stress and lead to crack formation during curing. In addition, a serious drawback is the emission of formaldehyde. This can be controlled by an appropriate F/U molar ratio or by adding substances to the adhesive mixture that can capture the free formaldehyde (Pizzi, 2003; Maslosh *et al.,* 2005; Bilgin and Colakoglu, 2021).

2.2.4 Melamine-formaldehyde (MF) and melamine-urea-formaldehyde (MUF) adhesives

2.2.4. Melamin-formaldehidna (MF) i melamin-urea-formaldehidna (MUF) ljepila

Melamine-formaldehyde (MF) oligomers are obtained through the polycondensation of melamine (1,3,5-triamino-2,4,6-triazine) and formaldehyde in two stages, as illustrated in Figure 3 (Merline *et al.,* 2013; Dorieh *et al.,* 2022b). In the first stage, the aldehyde reacts with the amino groups, leading to the generation of hydroxymethyl compounds (hydroxymethylation step). A neutral or slightly alkaline medium (pH 7-8.5) favours this reaction. The addition of the first three molecules of formaldehyde occurs rapidly, forming trihydroxymethylmelamine (Figure 3). In the condensation stage, the hydroxymethyl compounds can react with each other, leading to the formation of methylene and methylene ether linkages. The former predominates at pH between 7 and 8, while the latter - at pH above 9. The curing of MF resins occurs in the presence of an acidic catalyst at elevated temperatures (130-150 °C), but it can also proceed in a neutral or slightly alkaline medium.

The structure of the obtained MF adhesives depends on the synthesis conditions, namely: mole ratios of the raw materials, temperature, and pH.

Due to the high cost of MF adhesives, the production and use of so-called melamine-urea-formaldehyde (MUF) resins are becoming more prevalent. These thermosetting resins are obtained by the interaction of urea, melamine and formaldehyde. Relative mass proportions of melamine to urea typically range from 50:50 to 30:70 (Pizzi and Ibeh, 2014). The properties of these MUF resins are influenced by the synthesis method, the order of component addition, molar ratios, and melamine content (Tohmura *et al.,* 2001). The latter significantly affects the solid content, gelation time, and viscosity, as well as the chemical properties of the resin.

MUF resins outperform UF resins because they offer several advantages, such as better water resistance, colourlessness, lower price, and fewer formaldehyde emissions (Kim and Park, 2021).

One of the most valuable properties of MF adhesives is their high water resistance, not only in cold but also in boiling water. In this regard, they do not fall short of PF resins. Other significant advantages of these adhesives include their non-hazardous nature and excellent bonding strength. The adhesive joints are colourless and more moisture and heat resistant than UF adhesives (Dorieh *et al.,* 2022b).

A disadvantage of MF adhesives is their high cost, which limits their application. Therefore, MUF adhesives combine the good adhesion properties of melamine adhesives with the lower cost of urea adhesives (Li *et al.*, 2023). Another drawback of MF adhesives is limited storage stability due to the presence of many imino and hydroxymethyl groups (Lan *et al.,* 2019).

2.2.5 Polyurethane and polyisocyanate adhesives

2.2.5. Poliuretanska i poliizocijanatna ljepila

Polyurethane-based adhesives have extensive applications in the woodworking and furniture industry (Dimitrov *et al.,* 2017). The starting components for the synthesis of polyurethanes (PU) are polyisocyanates-methylene diphenyl diisocyanate (MDI) or toluene diisocyanate (TDI) and polyols (Akindoyo *et al.,* 2016; Kemona and Piotrowska, 2020). The most crucial unit in the composition of PU is the urethane linkage (–NH–COO–), which is formed by the reaction of isocyanate group with hydroxyl group from the polyols (Figure 4).

Isocyanates used in the production of wood composites vary depending on the type of wood composite and the required physical and mechanical properties. Organic isocyanates, typically polymeric methylene diphenyl diisocyanate (pMDI), have been used as adhesives since the early 1970s. The synthesis of pMDI starts with the condensation of aniline with formaldehyde in an acidic solution (Conner, 2001). As a result, a complex mixture of isomeric diamines and oligomeric polyamines is formed. The pMDI is produced by the phosgenation of aniline-formaldehyde condensates.

The adhesive properties of pMDI are based on the reactivity of isocyanate groups, which react with com-

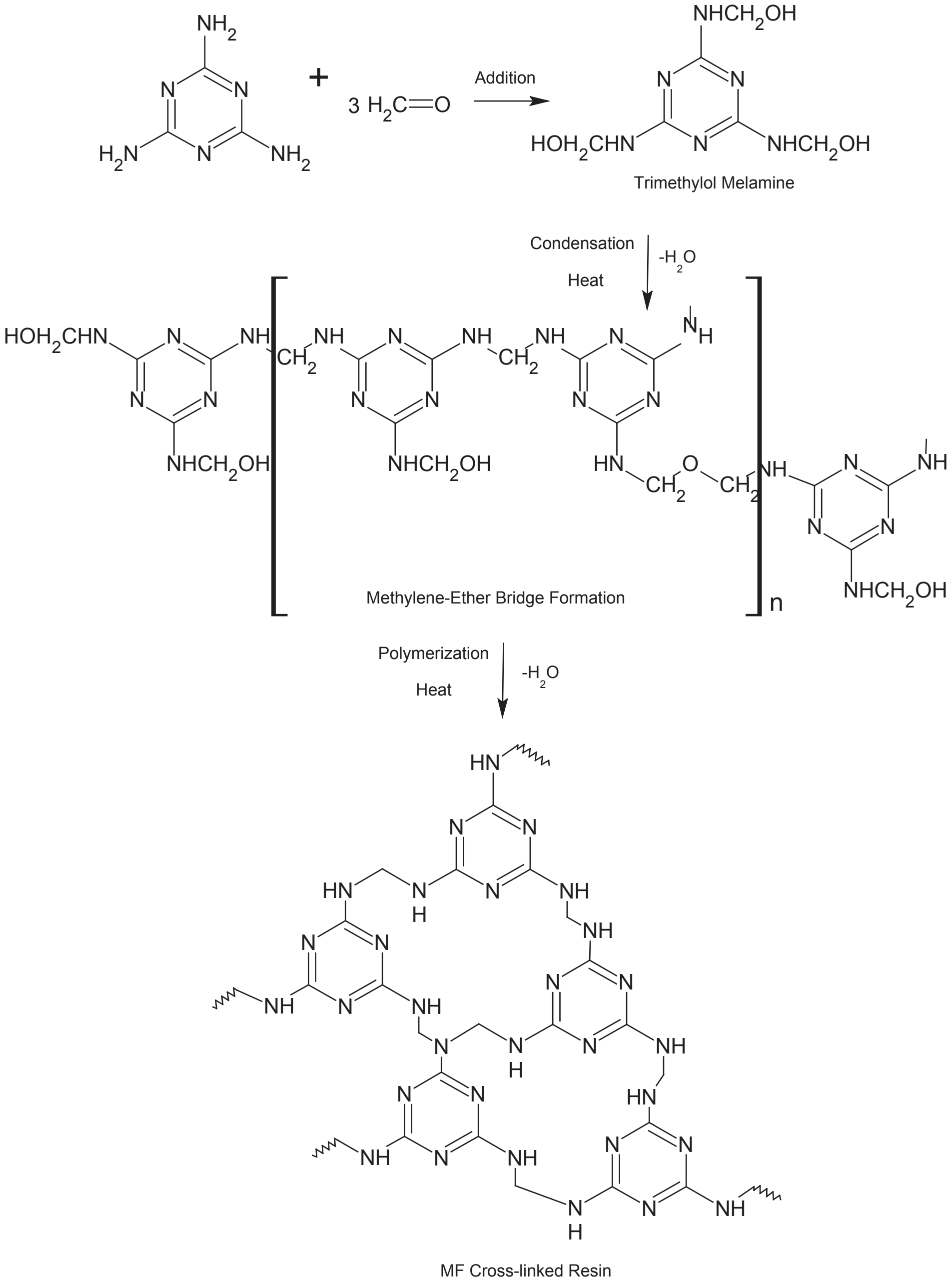


Figure 3 General reactions to form MF resin
Slika 3. Opće reakcije dobivanja MF smole

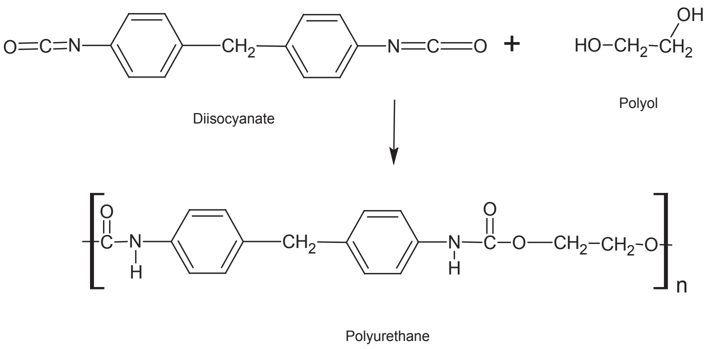


Figure 4 Reaction example for polyurethane synthesis
Slika 4. Primjer reakcije sinteze poliuretana

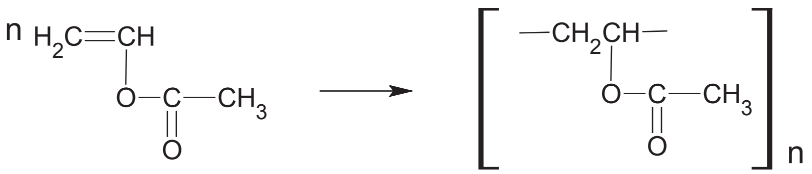


Figure 5 Polymerization reaction to produce polyvinyl acetate
Slika 5. Reakcija polimerizacije za dobivanje polivinil-acetata

pounds containing active hydrogen, such as water, alcohols, and amines. The reaction rate increases with heat. pMDI is an effective bonding agent for wood at increased moisture content, as it reacts with water to produce polyurea (Kloeser *et al.,* 2007). Covalent bonds may theoretically be formed between hydroxyl groups in the wood and the isocyanate, facilitating the anchoring of polyurea and filling the gaps between wood pieces.

PU adhesives are noted for their high adhesion to various materials, including wood, due to the presence of polar –N=C=O groups. They have several advantages such as universality; and can cure at room temperature, under heat for a short period, and at low pressure. Durability, atmospheric resistance, as well as very good strength indicators characterize the adhesive joints (Shirmohammadli *et al.,* 2023).

The primary disadvantage of PU adhesives is their toxicity due to the presence of isocyanates. It can be reduced by using blocked isocyanates (Wirts *et al.,* 2003).

Polyisocyanate adhesives, primarily based on pMDI, have several advantages. They exhibit excellent adhesion and cohesion, high reactivity, and effective bonding, forming connections even in high moisture content. The resulting adhesive joint is characterized by excellent hardness and plasticity, along with biological and chemical resistance (Kawalerczyk *et al.,* 2023). There are no formaldehyde emissions, which negatively affect the production process and are associated with additional costs.

Along with these advantages, pMDIs also have some disadvantages, namely their high cost and the tendency to bond with other materials, such as press tops, which limits their use for gluing in the surface layers of wood composites. Another disadvantage is the toxicity of isocyanates; therefore, measures to prevent the risk of diseases when using these substances are needed (Kloeser *et al.,* 2007; Frihart, 2012).

2.2.6 Polyvinyl acetate (PVAc) adhesives
2.2.6. Polivinilacetatno (PVAC) ljepilo

Polyvinyl acetate (PVAc) adhesives are thermoplastic, polymerization adhesives based on polyvinyl acetate, which is obtained by polymerization of vinyl acetate according to the following scheme (Figure 5), (Frihart, 2012):

The raw materials for the production of vinyl acetate are acetylene and acetic acid, which interact in the following reaction (Figure 6):

Depending on the method of polymerization, polyvinyl acetate can be obtained either as a solution in an organic solvent and a water emulsion or as a solid that melts upon heating. The PVAc emulsion finds the greatest application in wood bonding, specifically at 65 °C (Gadhave and Dhawale, 2022). Hydrogen peroxide is the initiator of the reaction, polyvinyl alcohol is an emulsifier, and formic acid acts as the pH regulator.

Plasticized and non-plasticized PVAc emulsions are white liquids with varying viscosities. Its pH is acidic due to the presence of acetic acid. Plasticisers, particularly dibutyl phthalate, can be added to the polyvinyl acetate emulsion. Plasticized and non-plasticized PVAc emulsions can bond wood quickly, either upon heating or without heating.

Wood bonding occurs because of a physical process in which the solvent (water) penetrates the depths of the wood. The polymer film remains leaving it in the adhesive layer and forming an elastic adhesive joint (Bomba *et al.,* 2014).

Using these adhesives in the furniture and construction industry guarantees technological efficiency. Therefore, depending on the duration of a specific operation, their polymerization and polycondensation can be accelerated by adding a hardener or rising the temperature. This is accompanied by an increase in the bond number between molecules and the formation of a spatial structure, enhancing the adhesive properties of specific resins. Based on the presented interactions between the molecules during the curing process, a particular type of binder can be selected to ensure the desired performance characteristics of the final products.

Unfortunately, the curing of these adhesives that guarantee high atmospheric resistance and performance under severe weather conditions, is associated with releasing large amounts of formaldehyde emissions. For

$$\mathrm{HC{\equiv}CH} + \mathrm{H_3C{-}COOH} \longrightarrow \mathrm{H_2C{=}CHOCOCH_3}$$

Figure 6 Reaction to produce vinyl acetate
Slika 6. Reakcija dobivanja vinil-acetata

this reason, the addition of formaldehyde scavengers aims to limit this negative factor as much as possible.

PVAc has long been among the cheapest adhesives used in the woodworking industry. They are characterized by good adhesion to various materials, absence of internal stress in the adhesive joint, complete safety, light resistance, storage stability, elasticity of the adhesive joint, and simplicity in bonding (Gadhave and Dhawale, 2022; Aydemir, 2014).

The disadvantages of PVAc adhesives are expressed in their thermoplasticity, low heat and water resistance. These bonding compounds undergo modification to improve their physical and mechanical properties (Kaboorani and Riedl, 2011; Morsi *et al.,* 2024).

3 TYPES OF EWPs INTENDED FOR CONSTRUCTION INDUSTRY
3. VRSTE KOMPOZITNOG DRVA U GRAĐEVNOJ INDUSTRIJI

In recent decades, it has been observed that wood consumption has been increasing not only in the furniture industry, but also progressively, in the construction industry (FAO, 2014; FAO, 2020). There has been heightened demand for and manufacturing of a wide variety of products, including glued laminated timber, composite panel materials, solid wood materials, and glued solid wood products. These items are mainly used in the construction of housing, buildings and facilities, as well as in the automotive, railcar and aircraft industry.

Composite laminated materials, commonly called EWPs, cover a wide range of assortments. Depending on the intended application of the final products, these items are formed by bonding solid wood components without defects, or by gluing veneer sheets, veneer pieces, large wood particles (wood chips), etc. (Bayat, 2023; Yadav and Kumar, 2021).

Rapid development of the construction industry necessitates an increased use of wood-based structural elements to build a significant portion of homes, buildings, and facilities. A substantial part of this includes multi-layered glue-laminated solid wood beams (GLT), single-layer glue-laminated wood beams (KVH structural timber), glued laminated panels with mutually perpendicular fibre orientation (CLT), and double T-beams. There is also a growing utilization of structural materials based on veneer sheets and pieces (LVL, PSL, SCL, and OSB).

Table 1 presents some of the most commonly used EWPs in the construction industry.

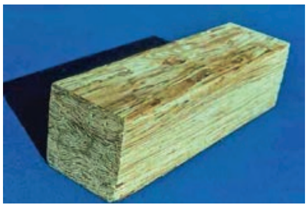
4 APPLICATION OF EWPs
4. PRIMJENA KOMPOZITNOG DRVA

The fields of application for the materials described above are quite diverse. These products are pri-

Table 1 Classification of composite materials (APA Wood)
Tablica 1. Podjela kompozitnih materijala (APA Wood)

Types of engineered wood (EWPs) *Vrste kompozitnog drva (EWP)*	**Definition** / *Definicija*	**Image** / *Slika**
GLT/Glued-laminated timber (glulam) *lijepljeno lamelirano drvo (glulam)*	Structural materials (beams) produced by layers of dimensional lumber bonded together with structural adhesives *konstrukcijski materijal (grede) proizveden od slojeva dimenzioniranog drva spojenih konstrukcijskim ljepilima*	
KVH structural timber *KVH konstrukcijsko drvo*	Structural materials (single-layer beams) produced by longitudinal finger-jointed splicing corresponding to strength class C24 *konstrukcijski materijal (jednoslojne grede) proizveden uzdužnim spajanjem zupcima koji odgovaraju klasi čvrstoće C24*	
CLT/Cross- laminated timber *CLT / unakrsno lamelirano drvo*	Panels of glued lamellae with mutually perpendicular orientation of the fibre direction *ploče od lijepljenih lamela s međusobno okomitom orijentacijom smjera vlakana*	
I-joists, I-beams *I-grede*	Double T- beams of OSB (or plywood) and solid wood *dvostruke T-grede od OSB-a (ili furnirske ploče) i masivnog drva*	

Table 1 Continue
Tablica 1. Nastavak

Types of engineered wood (EWPs) *Vrste kompozitnog drva (EWP)*	**Definition** / *Definicija*	**Image** / *Slika**
LVL/Laminated veneer lumber *LVL / lamelirano furnirsko drvo*	Materials (beams) obtained by pressing several layers of veneer with configuration in the longitudinal direction *materijal (grede) dobiven prešanjem nekoliko slojeva furnira s konfiguracijom u uzdužnom smjeru*	
SCL/Structural composite lumber *SCL / konstrukcijsko kompozitno drvo*	Materials obtained by bonding veneer pieces and wood particles under high pressure *materijal dobiven lijepljenjem komada furnira i drvnih čestica pod visokim tlakom*	
PSL/Parallel strand lumber *PSL / građevno drvo od traka furnira*	Materials produced from veneers clipped into long strands laid in parallel formation and bonded together with an adhesive *materijal proizveden od furnira izrezanih u duge trake položene paralelno i spojene ljepilom*	
OSL/Oriented strand lumber *OSL / građa s orijentiranim iverjem*	Materials obtained by gluing wood strand elements with wood fibres primarily oriented along the length of the product under high pressure *materijal dobiven lijepljenjem drvnog iverja pod visokim tlakom s vlaknima pretežito orijentiranima duž duljine proizvoda*	
OSB /Oriented strand board, waferboard *OSB / ploča s orijentiranim iverjem*	Boards of oriented large-sized wood particles *ploče od orijentiranih drvnih čestica velikih dimenzija*	

*The sources from which the images were taken are indicated in the references (83-91).
**Izvori iz kojih su slike preuzete navedeni su u popisu literature (83. – 91.).*

marily used in residential and public buildings, sports facilities, bridges, awnings, etc. Therefore, in the construction industry, EWPs are mainly categorized into two groups: load-bearing and enclosing structures.

Enclosing structures most commonly include products such as plywood, OSB, and in recent years, there has been a rapid increase in the consumption of CLT. The choice of binder depends on the environment in which it will be used. The most frequently used adhesives for manufacturing these products are PF (softwood plywood and OSB), pMDI (for OSB), UF (hardwood plywood), and PVA (Aydin *et al.,* 2006; Ferreira *et al.*, 2022; Sulastiningsih *et al.,* 2024; Ibrahim and Febrianto, 2013; Chanda *et al.,* 2022). The most suitable adhesives used for CLT production are melamine, PRF, and resorcinol-based resins, as well as one-component PUR adhesives (Mohd Yusof *et al.,* 2019; Miyazaki *et al.,* 2024).

Load-bearing structures include beams, arches, frames, trusses, domes, spatial structures, and others. One of the most commonly used materials in constructing these facilities is glued laminated timber (GLT).

Glued laminated beams of the GLT type have a low volumetric mass, high strength properties, and significant durability under various conditions, which is generally measured in decades. Depending on the operating environment, special attention is paid both to the type of wood used to form the structural package, and to the adhesive used for their preparation. Overall, GLT products provide four general-use categories, which are specified in ANSI A190.1-2012 (2013).

Framing Elements: This framing type is a common choice for residential construction. This GLT class is recommended only for hidden locations.

Industrial: These industrial-grade GLT are used in the construction of structures where aesthetic appearance is not essential. Their application is primarily seen in elements that are not visible to the public, as these materials may exhibit certain imperfections in the wood, such as joints, cavities, holes, etc.

Architectural: When GLT is to be used as facade material or external element of buildings and structures, the architectural class offers high product quality, free of voids and defects in the wood, and with a very good aesthetic appearance.

Premium (very high class, made to order): This GLT class is produced only to special orders and is typically used in areas where high concentrations of people are expected. The Premium class offers wood with smooth surfaces and fine finishes, resulting in a high-quality final product.

The most commonly used adhesives for GLT production are PRF type, MUF, one-component PUR and in some cases, PVAc adhesives (Fiorelli and Dias, 2006; Derkowski *et al.,* 2022; Ayanleye *et al.,* 2023; Kytka *et al.,* 2024). The latter are used only when the structural elements are not exposed to severe atmospheric conditions, such as drastic temperature changes and high humidity.

Another type of EWP for constructing load-bearing structures is a material created by bonding veneer sheets or pieces. These typically include products such as LVL, SCL, PSL, OSL, etc. Since the defects in the wood used to construct these products and the bonding of veneer pieces are clearly visible, these products are primarily applied in the construction of hidden structures.

The most commonly used adhesives for manufacturing LVL, SCL, PSL, and OSL are PF, MF, MUF, and pMDI (Ferraz *et al.,* 2009; Kurt and Çavuş, 2011; Kurt and Cil, 2012; Sun *et al.,* 2021; Malanit *et al.,* 2011).

4 CONCLUSIONS
4. ZAKLJUČAK

This review focuses on the most common synthetic adhesives applied in the manufacturing of some EWPs. The type of bonding agents not only guarantees high resistance of the final products to adverse weather conditions but may also be associated with formaldehyde emissions. Therefore, the choice of adhesive must be consistent with the operating conditions.

The adhesives based on melamine, phenol and resorcinol considered in this study are particularly preferred due to their high moisture resistance, temperature fluctuations and high durability under structural loads. It should be noted, however, that their significantly high price makes the final product more expensive. UF adhesives are particularly preferred in industry due to their high cost-efficiency and good adhesion properties, but they have low water resistance. Other types of adhesives such as polyurethane and isocyanate are known for their excellent mechanical properties and water resistance. Their use guarantees very strong adhesive joints, as well as great durability of the elements. It should be noted that some of the adhesives described so far release free formaldehyde, the amount of which depends on their synthesis.

High water resistance does not characterize PVAc adhesives, but they have several other advantages such as elasticity, ease of processing, and environmental friendliness. As a result, they are increasingly used in both the furniture and construction industries.

The future of adhesives for EWPs is focused on sustainability, performance, and innovative applications. Market growth is projected, driven by increasing urbanization, demand for sustainable building materials, and advancements in adhesive technology. All these factors contribute to a growing interest in developing high-performance adhesives for use in demanding environments and create opportunities for optimizing manufacturing processes.

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HRVATSKA KOMORA
INŽENJERA ŠUMARSTVA
I DRVNE TEHNOLOGIJE

HRVATSKA KOMORA INŽENJERA ŠUMARSTVA I DRVNE TEHNOLOGIJE

Osnovana je na temelju Zakona o Hrvatskoj komori inženjera šumarstva i drvne tehnologije.

Komora je samostalna i neovisna strukovna organizacija koja obavlja povjerene joj javne ovlasti, čuva ugled, čast i prava svojih članova, skrbi da ovlašteni inženjeri obavljaju svoje poslove savjesno i u skladu sa zakonom, promiče, zastupa i usklađuje njihove interese pred državnim i drugim tijelima u zemlji i inozemstvu.

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- **poticanje donošenja propisa kojima se utvrđuju javne ovlasti Komore,**
- **reagiranje struke na pripremu propisa iz područja šumarstva, lovstva i drvne tehnologije,**
- **suradnja s nadležnim institucijama i zastupanje struke u odnosu prema njima,**
- **organizacija stručnoga usavršavanja,**
- **zastupanje interesa svojih članova,**
- **izdavanje pečata i iskaznice ovlaštenim inženjerima,**
- **briga i nadzor poštivanja kodeksa strukovne etike,**
- **osiguravanje članova Komore za štetu koja bi mogla nastati investitorima i trećim osobama i sl.**

Članovima Komore izdaje se rješenje, pečat i iskaznica ovlaštenoga inženjera.
Za uspješno obavljanje zadataka te za postizanje ciljeva ravnopravnoga i jednakovrijednoga zastupanja struka udruženih u Komoru, članovi Komore organizirani su u razrede:

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