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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.

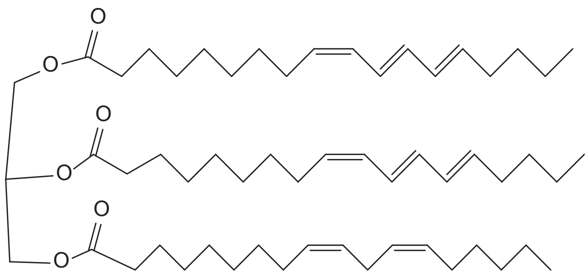
In this study, the feasibility of using new bio-based epoxy coatings obtained from tung oil based-epoxide resin (ETO) with bio-based curing agents was investigated. The curing process was performed using turpentine and moss oil as the curing agents for the tung oil-based epoxide resin (ETO) for the first 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,



Tung oil

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.)

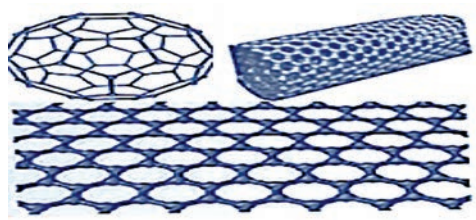


Figure 2 Structure of carbon nanoparticles
Slika 2. Struktura ugljikovih nanocijevi

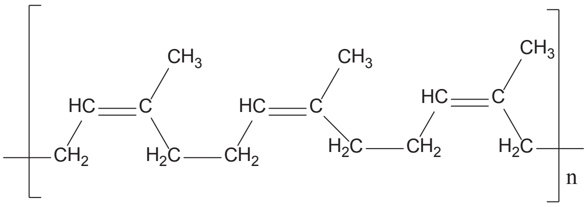


Figure 3 Structure of a sesquiterpenes (cis-polyisoprene)
Slika 3. Struktura seskviterpena (cis-poliizoprena)

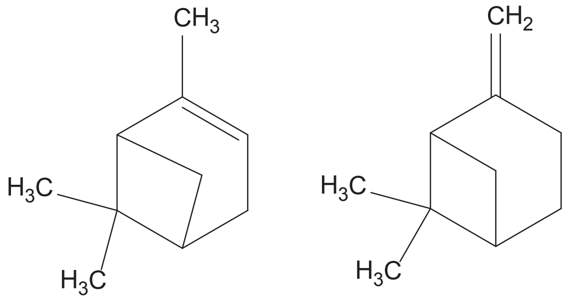


Figure 4 Chemical structure of major components of turpentine oil
Slika 4. Kemijska struktura glavnih komponenata terpentinskog ulja

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 (-**C**H=**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

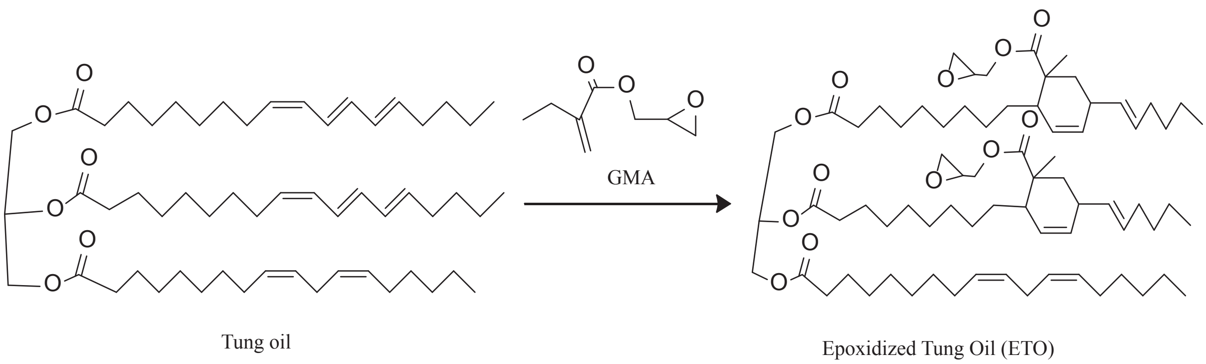


Figure 5 Epoxidation of tung oil reaction
Slika 5. Reakcija epoksidacije tungova ulja

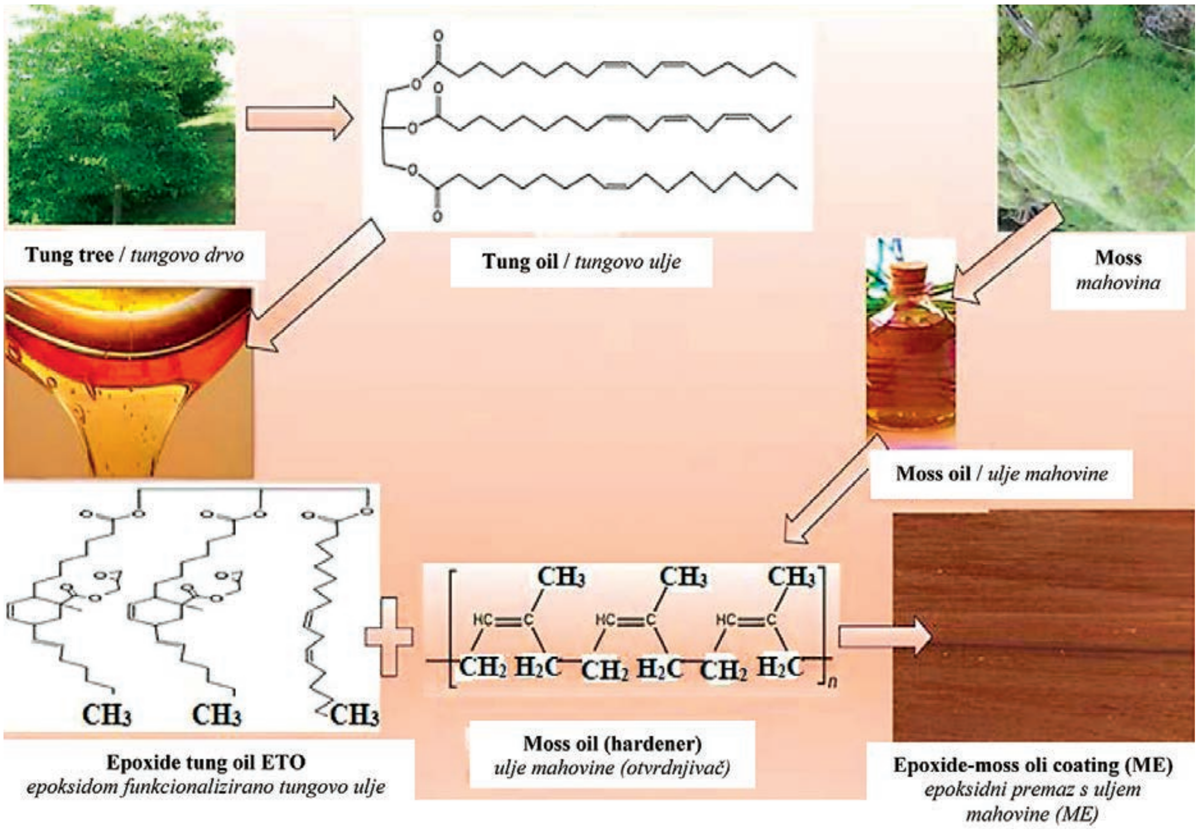


Figure 6 Preparation of epoxidized moss oil coatings (ME)
Slika 6. Priprema epoksidnog premaza s uljem mahovine (ME)

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

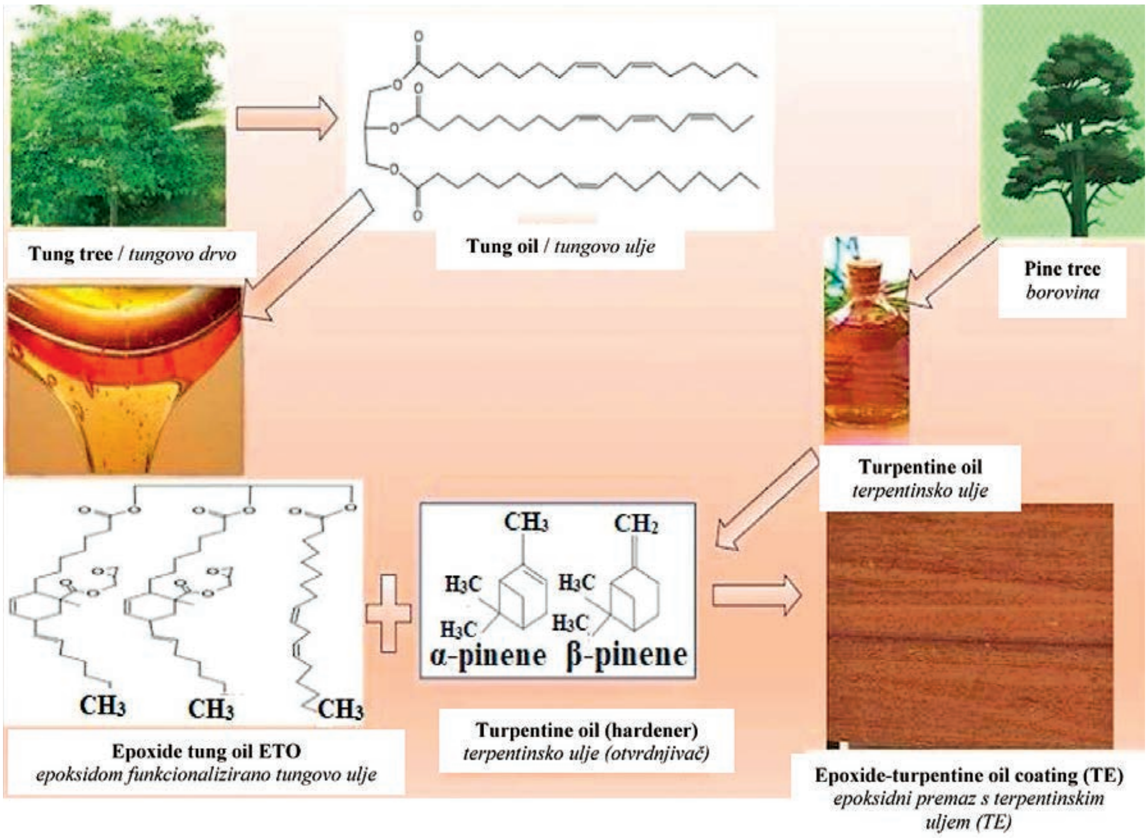


Figure 7 Preparation of epoxide-turpentine oil coatings (TE)
Slika 7. Priprema eposidnog premaza s tungovim uljem (TE)

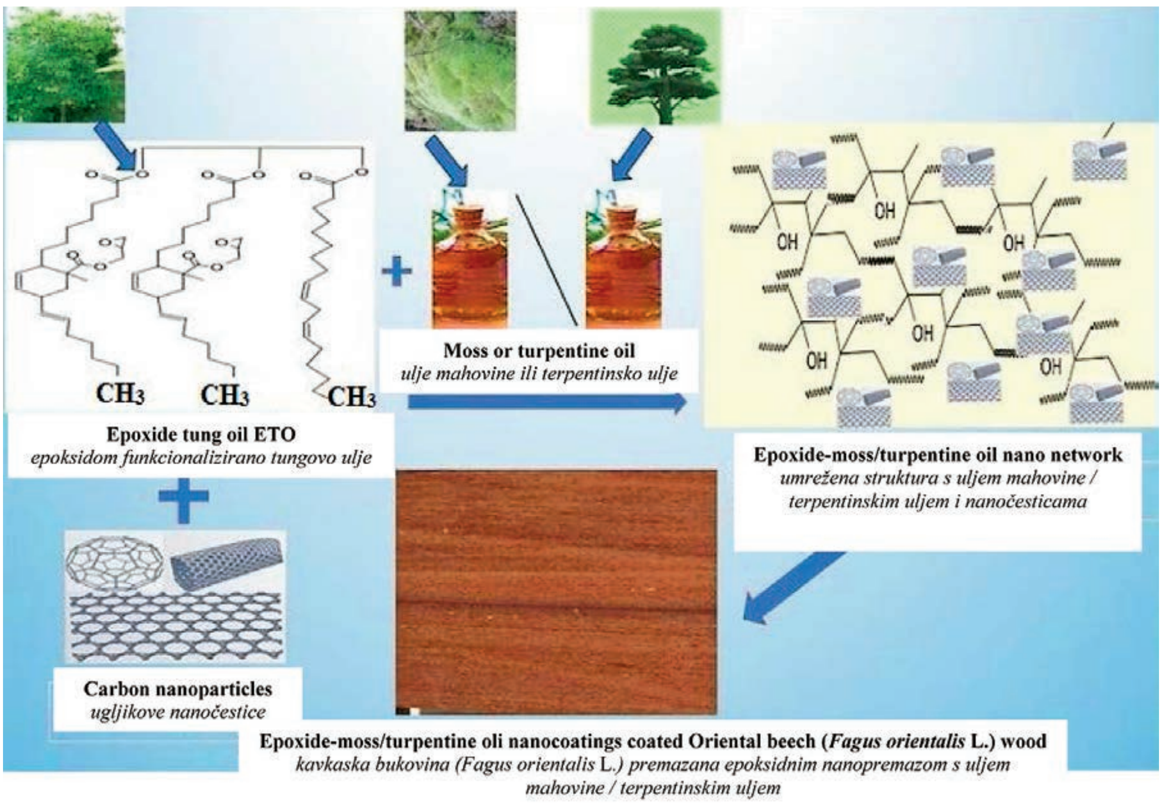


Figure 8 Use of moss and turpentine oil in combination with carbon nanoparticles to produce bio-based epoxy nanocomposite coatings
Slika 8. Upotreba ulja mahovine i terpentinskog ulja u kombinaciji s ugljikovim nanočesticama u pripremi epoksidnih nanokompozitnih biopremaza

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/m² / *Prosječna mjesečna brzina vjetra*, mm = kg/m²	78.50	51.60	10.00
Total number of rainy days / *Ukupan broj kišnih dana*	10.00	3.00	4.00

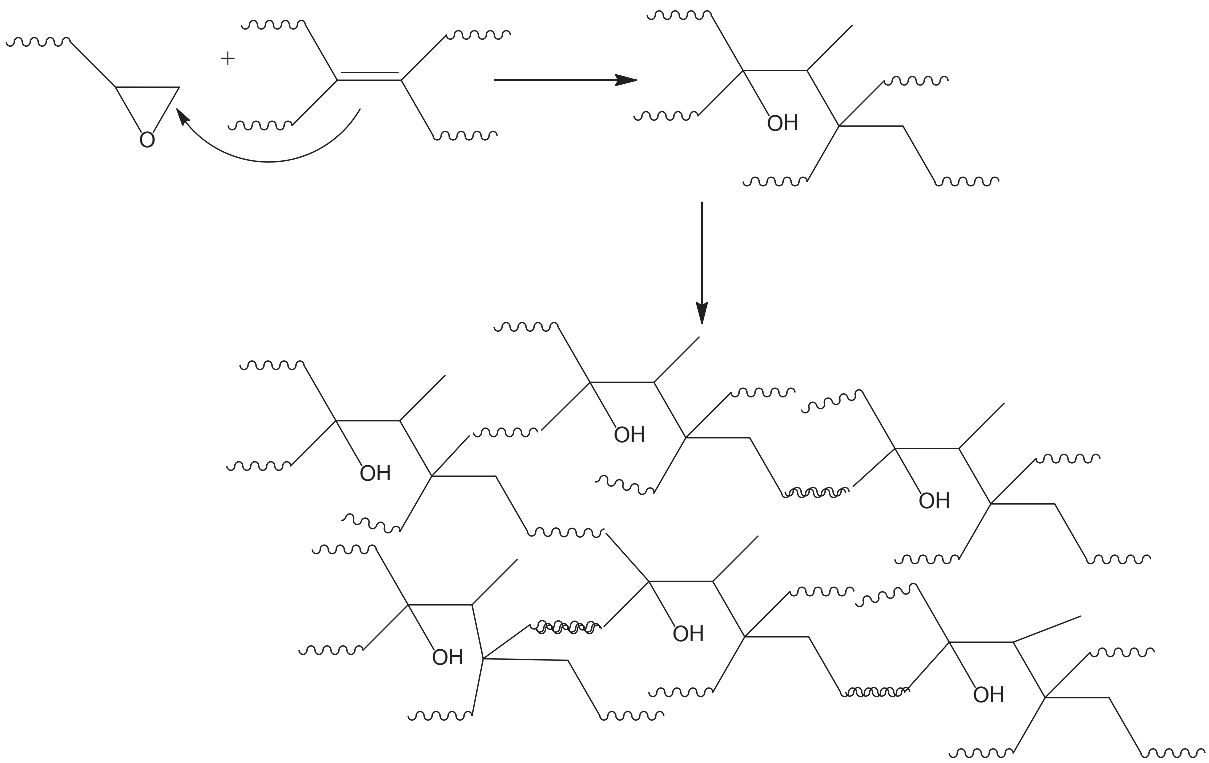


Figure 9 Chemical reaction of epoxy ring opening and cross-linking
Slika 9. Kemijska reakcija otvaranja i umrežavanja epoksidnog prstena

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, cross-linking 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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