

The utilisation of turpentine and moss oil as epoxy hardeners for bio-based epoxy nanocomposite coatings

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Abstract:

Despite the growing interest in bio-based epoxy systems, there remains a significant research gap in developing fully bio-derived curing agents that can replace conventional BPA-based epoxy hardeners while maintaining adequate mechanical and physical performance on wood substrates. The objective of this study was to investigate the potential of new bio-based, bisphenol A-free epoxide nanocomposite coatings for wood surfaces, as a replacement for commercially available coating containing bisphenol A. In addition, the surface properties of these coatings were evaluated.

This study involves the use of environmentally friendly, bisphenol A-free, new bio-based epoxy coatings and their nanocomposite derivatives, in which both the resin and hardener are derived from natural sources. The study is original in its use of sustainable natural resources in the coatings industry, as well as in the development of cost effective and readily available systems compared to those derived from petroleum. Furthermore, this research is the first to employ moss oil and turpentine oil as hardeners in epoxy resin curing reactions.

The results obtained with these two oils, which share a similar chemical structure, were compared. The study also investigates the effect of nanoparticles on the physical and mechanical properties of the bio-based coatings. In this study, novel bio-based epoxide nanocomposite coatings for wood surfaces were prepared using a tung oil-based epoxide resin, which was cured with moss and turpentine oil for the first time. Moss oil and turpentine oil were utilised as epoxy hardeners. The wood species selected for this investigation was *Fagus orientalis* (oriental beech). Furthermore, as prospective substitutes for bisphenol A, the characteristics of the new bio-based epoxide coatings, specifically the system obtained using

moss oil and turpentine oil as hardening agent, were evaluated in relation to their nanocomposite derivatives doped with carbon nanoparticles (fullerene, carbon nanotubes, and graphene) for application on wooden substrates. Following the application of various coating materials to the wood surface, evaluations were conducted on the mechanical and physical properties of the wood. This included measurements of water absorption, oven-dry density, and compression strength parallel to the grain of *Fagus orientalis* (oriental beech). The findings revealed that all test specimens showed oven-dry density values higher than those recorded for the control group. After the final absorption period, all coated specimens demonstrated a reduction in water absorption compared to the control. Each coated specimen also exhibited a higher compression strength parallel to the grain than the control group. Consequently, it was established that the implementation of innovative bio-based nanocoatings has the potential to enhance the mechanical and physical properties of *Fagus orientalis* (oriental beech) wood.

Keywords: Bio-based materials, Coatings, Bisphenol A, Epoxy coatings, Essential oils, *Fagus orientalis*, Mechanical properties, Nanocomposites, Surface treatment of wood, Turpentine.

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Introduction

Currently, the wood coating industry is embracing two key areas: the circular economy and bio-based resources. There is a growing body of scientific research focusing on the utilisation of natural additives in coatings (Şen 2001, Mustapha *et al.* 2019). This reflects a broader trend in the industry to identify environmentally-friendly and multipurpose alternatives to traditional synthetic fillers (Sadh *et al.* 2018). These synthetic fillers have often been criticised for their lack of environmental responsibility in production (Sanjay *et al.* 2018, Venkatesan *et al.* 2016).

Kishi and Fujita (2008) demonstrated that an epoxy resin based on liquefied wood and resorcinol exhibited excellent performance as a matrix resin in fibre-reinforced composites.

Epoxy coatings are commonly used on wood surfaces for protection and durability. Epoxy is a type of resin that, when combined with a hardener, creates a strong and durable chemical bond.

It provides a protective layer that can help protect the wood from moisture, chemicals, and

wear. Epoxies are a very adaptable class of polymers with applications in almost every field, from aeronautics to home adhesives (Babahan *et al.*2020, Babahan-Bircan *et al.*2023, Hasan *et al.*2022).

Epoxy nanocomposites for wood are a type of epoxy coating that incorporates nanoscale fillers or additives to enhance the properties and performance of the coating on wood surfaces. These nanomaterials can improve mechanical properties, barrier properties, UV resistance, adhesion, and even provide antimicrobial properties (Hyvönen *et al.* 2006, Kaymakçı 2016). The addition of nanomaterials, such as nanoparticles, nanofibers, or nanoclays, to the epoxy resin can significantly improve the mechanical properties of the coating. This includes increased tensile strength, hardness, and impact resistance, making the wood surface more durable and resistant to wear and tear. Epoxy nanocomposites also offer enhanced barrier properties, providing increased resistance to water, moisture, and other environmental factors. This helps to prevent wood degradation, warping, and rotting, thereby extending the lifespan of the wood surface. Additionally, some nanomaterials used in epoxy nanocomposites, such as silver nanoparticles, possess antimicrobial properties. This means that they can inhibit the growth of bacteria, fungi, and other microorganisms on wood surfaces. This is particularly useful in applications where hygiene and cleanliness are important, such as kitchen countertops or cutting boards.

The addition of vegetable oils, resins, waxes, and natural extractants to wood coating formulations was undertaken with the objective of enhancing their performance when exposed to a range of environmental conditions, including those posed by insects, light, humidity, and temperature (Chang *et al.*1982, Pandey and Pitman 2004, Teaca *et al.*2019, Varganici *et al.* 2021). Plant oils that are not harmful can create a barrier on the surface of wood cells, reducing the amount of water that the wood absorbs. Because of this, these oils show a lot of promise as wood preservatives (Kabasakal *et al.* 2023).

Tung oil has a long historical usage, as evidenced by reports that the Chinese Empire employed it in its ship coatings. Poth asserts that tung oil, linseed oil, and plant oils are the three most significant drying oils (Poth 2001, Du *et al.* 2020). Due to its capacity to repel water, tung oil is employed for the preservation of wooden materials. Humar and Lesar (2013) also highlighted the capacity of tung oil to impede water absorption over varying timeframes, including the short, medium, and long term. Additionally, they underscored its ability to repel water and safeguard against brown and white rot fungi. Nevertheless, the commercial application of pure tung oil is constrained or requires the incorporation of drying agents into its formulation due to its relatively prolonged drying period (exceeding five days). Moreover, it was established that a coating of tung oil was an effective method of preventing the absorption of water and the growth of fungi that cause wood decay, thereby slowing down the deterioration of wood surfaces exposed to the elements. The extensive use of wood in outdoor environments has prompted research and development in both academic and industrial contexts to investigate innovative methods for enhancing wood coatings. In conclusion, tung oil-based epoxy coatings can be considered an optimal solution for enhancing and protecting wood surfaces. They offer a distinctive combination of strength and aesthetic appeal, rendering them suitable for a multitude of applications, including furniture, flooring, and countertops. Tung oil-based epoxy coatings provide exceptional water resistance, chemical resistance, and durability (Altay *et al.* 2024, Kabasakal *et al.* 2024).

Turpentine's oil primary constituents are volatile terpenic hydrocarbons ($C_{10}H_{16}$), which might vary based on the tree species, growing site, and distillation method. Its boiling point is approximately 150 °C. Turpentine is typically used as a cleaning agent, paint thinner, and varnish thinner. Vitamins and perfumes are two common products that contain derivatives (Cabaret *et al.* 2019). The primary derivative of turpentine is synthetic pine oil, which is utilized in cleaning solutions, disinfectants, and other items with a pine aroma (Zhu *et al.* 2018, Salvador

*et al.*2020, Gallo-Corredor and Sarria-Villa 2014). Turpentine oil is commonly used as a solvent in wood coatings. It is known for its ability to dissolve various resins and natural gums, making it an effective ingredient in many wood finishes. Turpentine oil is often used to thin down thick wood coatings, such as varnishes or oil-based paints. It helps to reduce the viscosity, making the coating easier to apply and ensuring a smoother finish. Turpentine oil can be used as a cleaner for wood surfaces, especially when there are stains or sticky residues. It helps to dissolve and remove the unwanted substances, preparing the wood for further coating or refinishing. Turpentine oil is a volatile solvent, meaning it evaporates relatively quickly. When added to wood coatings, it can help accelerate the drying time, allowing for faster application of multiple coats or finishing processes. Turpentine oil can be added to oil-based wood coatings, such as varnishes or stains, to enhance the glossiness of the finish. It helps to improve the flow and leveling of the coating, resulting in a smoother and shinier surface. But, this study represents the first instance of the utilisation of turpentine oil as a curing agent for epoxy resin.

Moss oil is primarily used in the production of perfumes, candles, and cosmetics. It is not commonly used as a coating for wood or other surfaces. Moss oil is generally not a common ingredient in wood coatings, and there is limited information available on its specific applications or benefits in this field. The limited availability of information on the use of moss oil contributes to the originality of this study. This study marks the first instance of using moss and turpentine oil as a curing agent for epoxy resin. Furthermore, it seeks to elucidate the comparative efficacy of these two oils, which share a similar chemical structure and double bond configuration with turpentine oil.

The incorporation of carbon-based nanomaterials such as carbon black, graphene, carbon nanotubes, and amorphous carbon nanoparticles into coating matrices has been widely studied and shown to be both realistic and effective in enhancing the performance of wood coatings. Carbon nanoparticles (CNPs) contribute to improved UV resistance, mechanical durability,

barrier properties, and in some cases, even antimicrobial effects. Their nano-scale dispersion provides a significant increase in surface area, promoting better interaction with polymer matrices and wood surfaces.

Several recent studies support the practical use of CNPs in wood coating formulations: Jirouš-Rajkovi'c and Mikle'ci'c (2021) successfully incorporated carbon nanoparticles into a waterborne acrylic coating applied on wood, which resulted in enhanced abrasion resistance and UV stability (Jirouš-Rajkovi'c and Mikle'ci'c 2021). Carbon black and related particles are already industrially employed in exterior wood stains and UV-protective coatings, demonstrating their viability on a commercial scale (Wålinder and Johansson 2011). In our study, the carbon nanoparticles were uniformly dispersed in the coating matrix, and no aggregation or sedimentation was observed during application. The enhanced properties observed in coated wood specimens including surface hardness and weather resistance further support the realistic and beneficial use of CNPs in such systems.

In this study, bio-based epoxy resin obtained from tung oil was cured with turpentine and moss oil, and the feasibility of using these oils as curing agents for epoxy resin was investigated. Furthermore, these new bio-based systems were prepared with carbon nanoparticles, and their nanocomposite derivatives were also obtained to investigate how the presence of carbon nanoparticles contributes to mechanical and physical properties. Despite extensive research on bio-based epoxy resins, most commercial systems still rely on petroleum-derived hardeners and bisphenol A-based chemistries, raising environmental and health concerns. Furthermore, the use of natural oil-derived curing agents remains largely unexplored, particularly regarding moss oil and turpentine oil as potential epoxy hardeners. Therefore, there is a clear need to investigate alternative bio-based curing systems capable of achieving comparable performance while reducing environmental impact. The present study addresses this research gap by developing

novel tung oil-based epoxy systems cured with moss and turpentine oils and evaluating their nanocomposite derivatives for wood coating applications.

Materials and methods

Material and chemicals

In this study, oriental beech (*Fagus orientalis* L.) was used as wood material. Wood specimens were prepared in the directions of oven-dry density (20 mm tangential x 20 mm radial x 20 mm longitudinal), water absorption (20 mm tangential x 20 mm radial x 20 mm longitudinal), and CSPG (20 mm tangential x 20 mm radial x 30 mm longitudinal), respectively. The Sigma-Aldrich Chemical Company supplied 2,4,6-tris (dimethyl aminomethyl) phenol, glycidylmethacrylate, phenothiazine, and tung oil (Figure 1). The fullerene C60, 95 %, and graphene nanoplatelet (S. A: 320 m²/g, diameter: 1,5 µm, 99,9 % (multi-walled carbon nanotubes)) were provided by the Turkish business Nanografi (Figure 2).

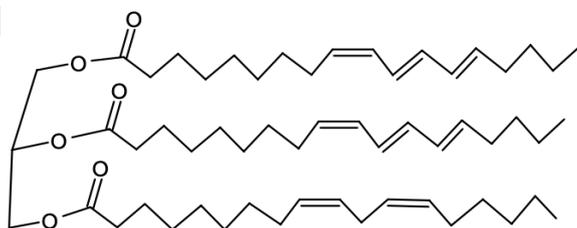


Figure 1: The structure of tung oil (produced by cold pressing the seeds of the tung tree (*Vernicia fordii* (Hemsl.) Airy Shaw)) (Babahan-Bircan *et al.* 2022, Kabasakal *et al.* 2023, Kabasakal *et al.* 2024).

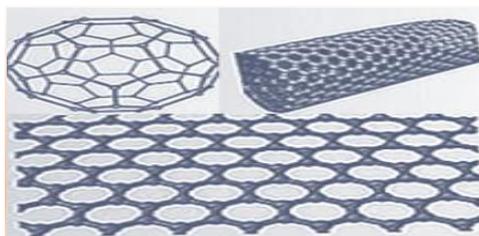


Figure 2: The structure of carbon nanoparticles

The crude moss oil was obtained from the Arifoglu Company, based in the Republic of Turkey. It was extracted from oakmoss (*Evernia prunastri* (L.) Ach.), which grows primarily on the bark of oak woods (*Quercus species*) in forested regions. Nine major groups comprised the chemical components of the moss oil. These included aliphatic hydrocarbons, alcohols, ketones, aldehydes, terpene/terpenoids (monoterpene hydrocarbons, oxygenated monoterpenes, oxygenated sesquiterpenes, and oxygenated sesquiterpenes), and other various substances. According to the chemical profile, the moss oil sample had 47 different chemical ingredients, which together made up 98,6 % of the total amount. The composition of the moss oil was found to be 15 sesquiterpene hydrocarbons (73,6 %), 9 oxygenated sesquiterpenes (19,5 %), 1 oxygenated monoterpene (0,1 %), and 1 monoterpene hydrocarbon (0,1 %).

Sesquiterpenes (Figure 3) represent a class of terpenes that are composed of three isoprene units, and frequently possess the molecular formula $C_{15}H_{24}$ (Çelik 2020, Tosun *et al.*2015).

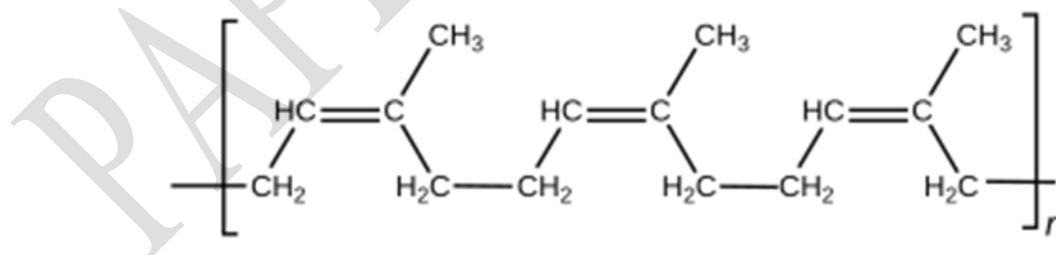


Figure 3: The structure of a sesquiterpene (cis-polyisoprene)

The Arifoglu Company in the Republic of Turkey provided the raw ingredients, which included pure α -pinene, β -pinene, and crude turpentine oil. It was obtained from the calabrian pine (*Pinus brutia* Ten.) species, a widely available pine tree native to the eastern Mediterranean region. The resin extracted from calabrian pine (*Pinus brutia* Ten.) was distilled to produce the turpentine oil used in our formulation. The main ingredient in turpentine, an essential oil made from gum resin, is pinene. Approximately 70-80 % rosin and 20-30 % turpentine make up the gum resin. The gum undergoes a distillation procedure after extraction, and the solid residue at the bottom of the distillation apparatus which is made up of fatty acids and resin acids is referred to as rosin. Turpentine is the volatile, liquid fraction that is mostly composed of α -pinene and β -pinene. There are two structural isomeric forms of pinene, a bicyclic monoterpene (Salvador *et al.* 2020, Afre 2006). The two main constituents of turpentine oil, α -pinene and β -pinene, are seen in Figure 4.

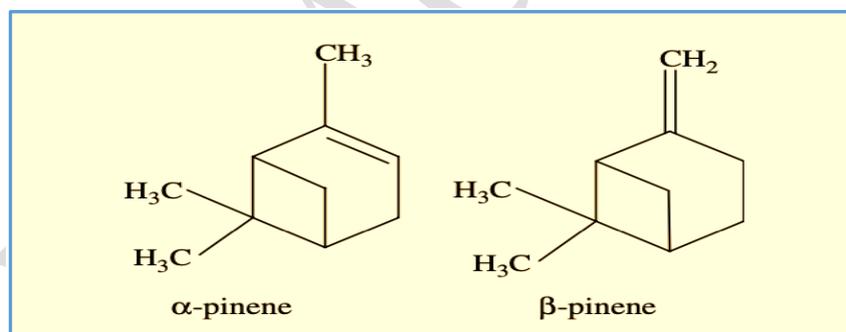


Figure 4: The chemical structure of major components of turpentine oil.

Synthesis of epoxy-functionalized tung oil (ETO)

The epoxidation of tung oil (ETO) was produced using a methodology that was consistent with that of our earlier study (Babahan *et al.*2020), employing the use of tung oil and glycidyl methacrylate via a Diels-Alder reaction. Firstly 25,96 g of glycidyl methacrylate was combined with 80 g of tung oil and 0,8 g of phenonthiazine at 150 °C in the presence of nitrogen gas (Figure 5). The finished product was identified through the use of $^1\text{H-NMR}$ and $^{13}\text{C-NMR}$ spectroscopy (Babahan *et al.*2020).

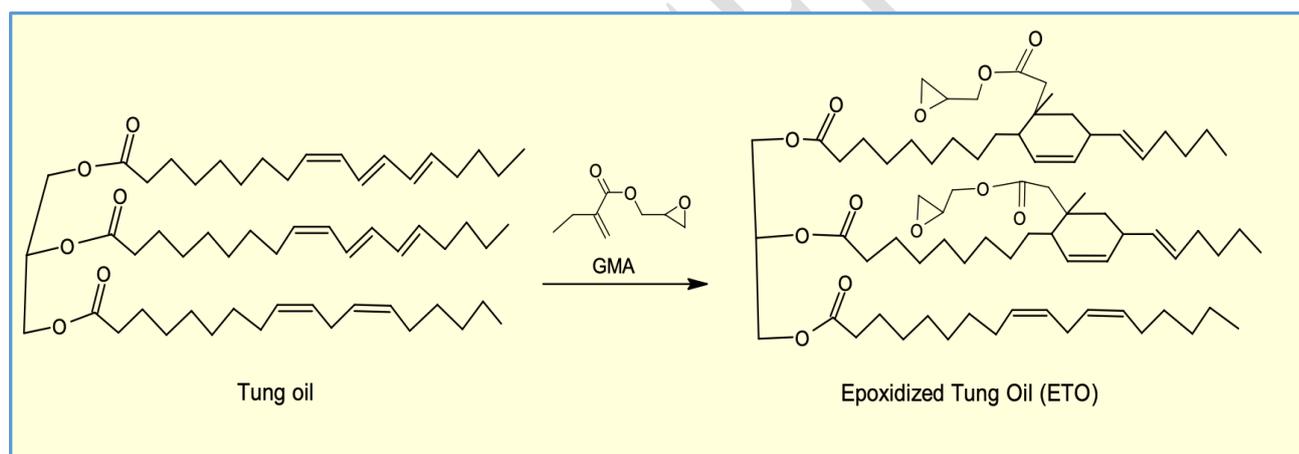


Figure 5: Epoxidation of tung oil reaction

$^1\text{H-NMR}$ (500 MHz, CDCl_3) δ (ppm): 5,64-6,34 (-CH=CH-); 5,22-5,25 (-C(O)O-CH₂-CH-O-C(O)-); 4,25-4,27 (-C(O)O-CH₂-CH-O-C(O)-); 4,12-4,14 (C(O)O-CH₂-CH(CH₂)O); 3,24-3,29 (O(CH₂)CH-CH₂-); 2,71 (-CH(CH=CH)CH₂-); 2,61-2,69 (O(CH₂)CH-CH₂-); 2,28 (-CH₂-C(O)O-); 2,06-2,15 (-CH₂-CH=CH-, -CH=CH-CH(CH=CH₂-); 1,56-1,66 (-CH₂-CH₂-C(O)O-); 1,19-1,45 -(CH₂)₅-CH₂-C(O)O-, -(CH₂)₂-CH₂-CH=CH-); 1,28 (CH₃-C-C(O)O-); 0,83-0,87 (CH₃-CH₂-CH₂). $^{13}\text{C-NMR}$ (500 MHz, CDCl_3) δ (ppm): 172,75 (C(CH₃)-C(O)O-); 134,71 (-CH=CH-); 64,67 (O(CH₂)CH-CH₂-); 48,93 (O(CH₂)CH-CH₂-); 44,17 (O(CH₂)CH-CH₂-).

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

In this study, turpentine oil and moss oil were employed as epoxy hardener agents. In order to facilitate the cross-linking process, 2,4,6-tris (dimethylaminomethyl) phenol was selected as the catalyst.

The epoxide reactant (ETO) was employed in the preparation of formulations at a 1:1 ratio of epoxide to oil, utilising both moss oil and turpentine oil as the epoxide reactant. The system obtained through the utilisation of moss oil as a hardening agent is designated as ME (Figure 6), whereas the system obtained through the utilisation of turpentine oil as a hardening agent is designated as TE (Figure 7). Each of the formulations was supplemented with a few drops of 2 % solution of 2,4,6-tris (dimethylaminomethyl) phenol, which served as the catalyst in this process. The coating formulations were prepared without the use of a solvent. The reactants were combined and subjected to rigorous agitation for a period of two hours following the weighing process. Subsequently, the mixtures were applied to wooden specimens using a brush in order to assess their mechanical and physical performance characteristics. The coated wood specimens were allowed to cure in a controlled environment at 25 ± 2 °C and $50 \pm 5\%$ relative humidity for 48 hours. To ensure consistency across all groups, the dry film thickness of the coatings was measured using a digital thickness gauge and maintained at approximately 120 ± 10 μm .

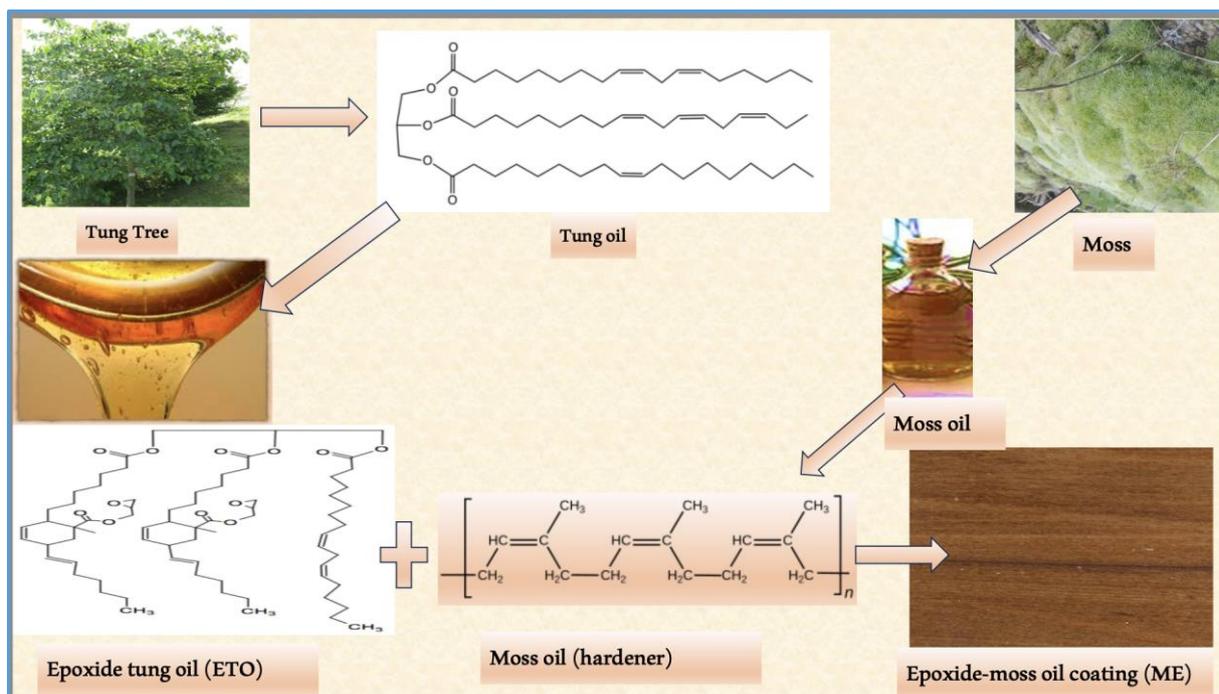


Figure 6: The preparation of epoxide-moss oil coatings (ME).

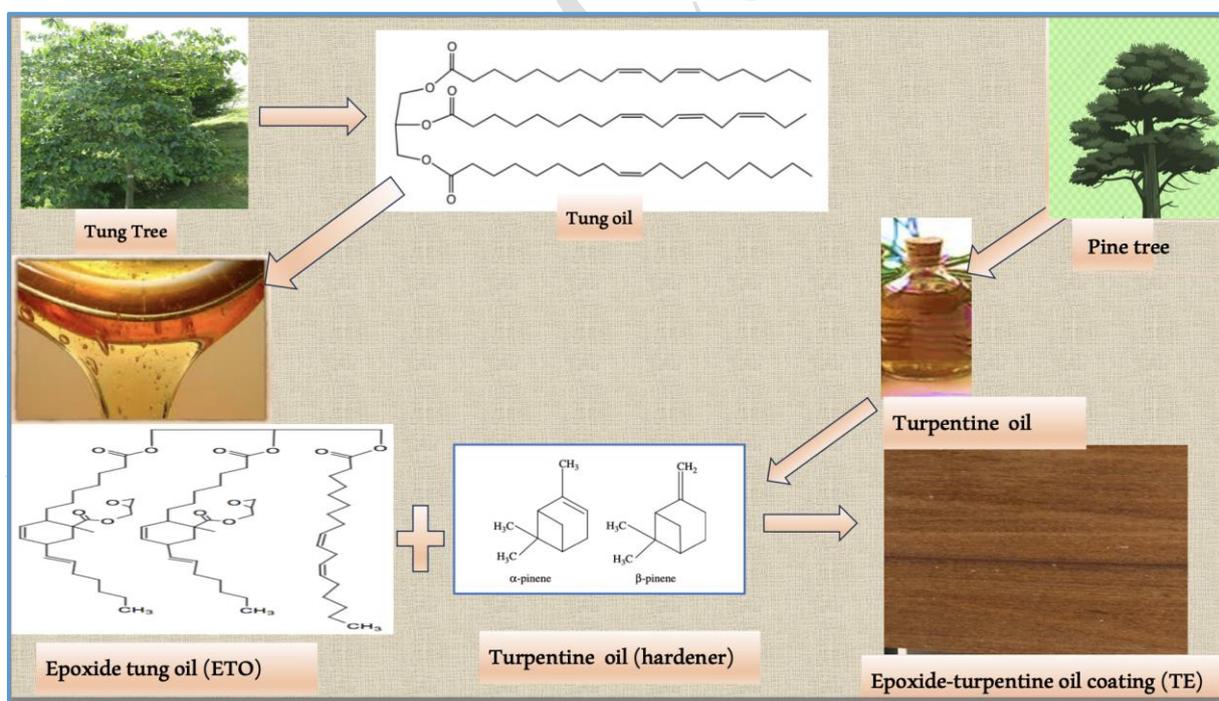


Figure 7: The preparation of epoxide-turpentine oil coatings (TE).

Preparation of bio-based epoxide-nanocomposite coatings

Figure 8 provides a comprehensive account of the tools and experimental configurations employed, accompanied by a schematic illustration of the procedures involved in the synthesis of epoxide-moss oil and epoxide-turpentine oil nanocoatings. These were developed in accordance with the methodology outlined in our previous research (Babahan-Bircan *et al.*2023). Nanocomposites were constructed using a variety of carbon nanoparticles, including graphene, carbon nanotubes, and fullerenes. The carbon-based nanoparticles were initially combined with acetone for a period of two hours at a temperature of 25 °C within a sonication bath. This process was conducted with the objective of preventing the deposition of nanoparticles and facilitating their dispersion within the epoxy matrix. Subsequently, the nanoparticles were mixed once more for a period of two hours in the aforementioned solvent, utilising a mechanical mixer, at a mass concentration of 0,10 %. Subsequently, 5 g of ETO epoxide resin was applied following the combination of acetone with the nanoparticles (5 g: 0,10 % by mass of epoxide resin). The reaction was administered at a controlled temperature for a period of two hours at rt. To extract the mixtures of nanoparticles and epoxide resin (ETO) from the solvent, the specimens were left at room temperature for a period of 24 hours. Subsequently, a 1:1 mass ratio of 5 g of bio-based hardeners, namely turpentine or moss oil, was added to the epoxy/nanoparticle mixture, and the mixture was vigorously agitated for approximately two hours. The formulation process did not employ the use of solvents. A 2 % addition of 2,4,6-tris (dimethylaminomethyl) phenol was made to all formulations.

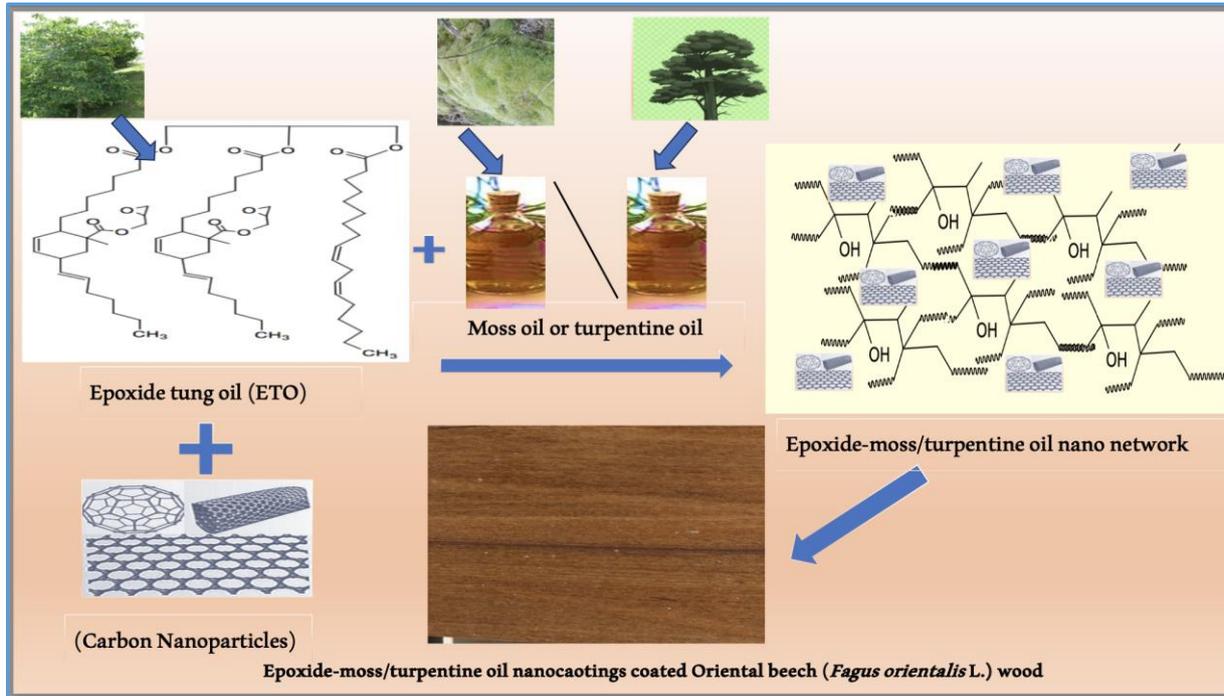


Figure 8: The preparation of bio-based epoxide nanocomposites utilising moss and turpentine oil in conjunction with carbon nanoparticles.

Oven-dry density test

Oven-dry density measurements were taken in accordance with TS ISO 13061-2 (2021). Furthermore, In total, 90 wood specimens measuring 20 mm × 20 mm x 20 mm were made, with 10 from each specimen group. The specimens were dried in an oven at a temperature of 103±2 °C until they reached the stated weight. Subsequently, the specimens' diameters were gauged with a precision calliper featuring a resolution of 0,01 mm, their volumes were calculated with the stereometric method, and their weights were recorded on an analytical scale with a sensitivity of 0,01 g.

$$\delta_0 = \frac{M_0}{V_0}$$

Calculated according to TS ISO 13061-2 (2021) standard.

In here;

M_0 = The specimen's oven-dry weight (g).

V_0 = The specimen's oven dry volume (cm³).

Water absorption test

In total, 90 wood specimens measuring 20 mm × 20 mm x 20 mm were made, with 10 from each specimen group. Specimens were kept in room temperature for 2, 6, 14, 30, 62, 94, and 126 h using distilled water. Following each soaking time, specimens were removed from the water, patted dry with paper, and then immediately weighed. Therefore, Equation 1 was applied to ascertain each specimen's WA.

$$WA = \frac{M_f - M_{oi}}{M_{oi}} \times 100 \quad (1)$$

In here;

WA = Water absorption (%),

M_f = Specimen's weight after water absorption period (g),

M_{oi} = Oven dry weight of specimen before water absorption period (g).

Compression strength parallel to grain (CSPG)

In accordance with ISO 13061-17 (2017), a universal test apparatus with a 39,2266 N capacity and a 6 mm/min loading period was used to perform the compression strength parallel to grain test (Liu *et al.* 2019). A total of 90 wood specimens, 10 from each specimen group, with dimensions of 20 mm × 20 mm x 30 mm, were used. Prior to testing, wood specimens were conditioned for two weeks at 20 °C and 60 % relative humidity.

$$\sigma_B = \frac{P}{a \cdot b}$$

Calculated according to ISO 13061-17 (2017) standard.

In here;

σ_B – CSPG (N/mm²),

P – load at break (N),

a, b – specimen cross-section dimensions (mm).

Statistical evaluation

The Duncan test and variance analysis were looked at using the SPSS software once the test results were acquired with a 95 % confidence level ($p < 0.05$). Homogeneity groups (HG) were the subject of statistical investigations; distinct letters denote statistical significance.

Results and discussion

This study encompasses the utilisation of environmentally benign, human-friendly and BPA-free healthy epoxy coatings and their nanocomposite derivatives, with both the resin and hardener derived from natural sources.

The study is original in terms of the utilisation of sustainable natural resources in the coating industry, as well as the development of cost-effective and readily accessible systems in comparison to those derived from petroleum. Furthermore, this research is original in that it employs the use of moss oil and turpentine oil as hardeners in epoxy resin curing reactions for the first time. The results obtained with these two oils, which have a similar chemical structure, were compared. Furthermore, the impact of nanoparticles on the physical and mechanical properties of bio-based coatings is also examined.

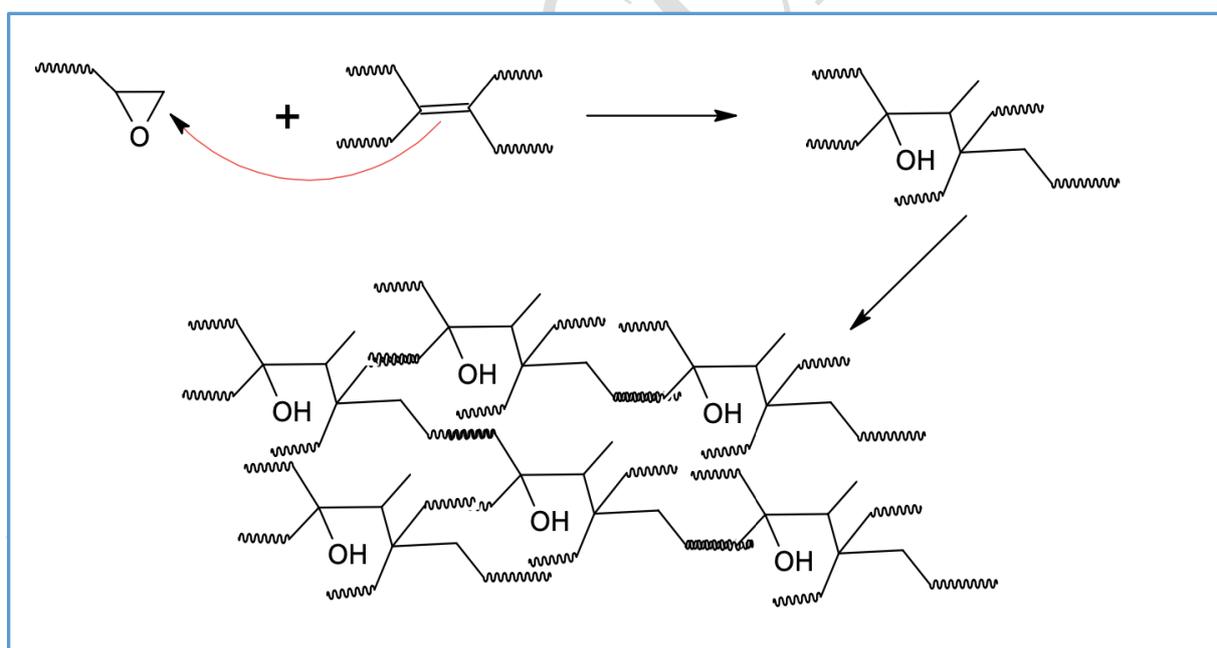


Figure 9: The chemical reaction of epoxy ring opening and cross-linking.

New bio-based epoxy nano-coating systems have been developed to replace BPA-based epoxy coatings in the wood industry. The wood species selected for this research is oriental beech (*Fagus orientalis* L.). To achieve this aim, tung oil-based epoxide resin (ETO) was used to

create a new generation of bio-based epoxide nano-coatings. Furthermore, hardeners such as moss oil and turpentine oil were also used for the first time in this study for epoxy curing. The first stage of the process involved the combination of tung oil with glycidyl methacrylate, which forms the basis of the bio-based epoxide resin (ETO). The epoxide resin was then cured with hardeners (ME and TE systems were obtained). To obtain nanocomposite derivatives of these coatings, the epoxide resin (ETO) was premixed with carbon nanoparticles (including graphene, carbon nanotubes (CNTs) and fullerenes) before curing with moss oil and turpentine oil. The physical and mechanical properties of oriental beech (*Fagus orientalis* L.) wood, including water absorption (WA) levels, oven-dry density and compressive strength parallel to the grain (CSPG) were also investigated. Comparative analysis of epoxy coatings obtained by curing bio-based epoxy resin prepared from tung oil with moss/turpentine oil and its nanoparticle-containing derivatives revealed interesting findings. The oven dry density, water absorption and compressive strength parallel to fibre (CSPG) results are presented in Table 1, Table 2, Table 3, Table 4.

In the first step, bio-based epoxide resin (ETO) was obtained by epoxidisation of tung oil by Diels-Alder reaction. This bio-based epoxide resin (ETO) was cured with bio-based hardeners. An examination of the chemical structures of the active substances in turpentine and moss oil, which are used as hardeners, reveals that both contain double bonds in a similar manner. 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, resulting in a ring opening reaction. At this juncture, crosslinking ensues. The reaction between oxirane groups and double bonds results in an increase in cross-linking density. It is yet to be documented in the existing scientific literature that a epoxidized tung oil between bearing double bonds compounds, used for the preparation of corresponding coatings, can be carried out successfully. The process is depicted in Figure 9.

In contrast, commercial epoxy coatings used for wood substrates typically rely on a diglycidyl ether of bisphenol A (DGEBA) epoxy resin and an amine-based curing agent (e.g., polyamines, aliphatic or cycloaliphatic amines). In these systems, curing proceeds via a well-established mechanism where amine hydrogen atoms act as nucleophiles and attack the electrophilic carbon in the oxirane ring. This results in a highly efficient and fast ring-opening polymerization, producing a densely crosslinked, thermoset network with excellent mechanical and chemical resistance. This mechanism has been widely reported and optimized for various applications including protective wood coatings (May 1988, Kirk 1991).

The key distinction lies in the source and nature of the crosslinking agent: in our system, crosslinking is achieved through reactive sites inherently present in the epoxidized-tung oil backbone with double bonds, whereas in commercial systems, crosslinking relies on external hardeners such as multifunctional amines. While commercial systems typically offer higher crosslink density and faster cure kinetics, bio-based systems provide environmental advantages and tunable flexibility, albeit often requiring longer cure times and elevated temperatures to achieve full network formation (Yang *et al.* 2008, Sharma and Kundu 2006).

Oven-dry density

Table 1 lists the oven-dry density values for the coated oriental beech (*Fagus orientalis* L.) specimens and the control group.

Table 1: Oven-dry density values of oriental beech (*Fagus orientalis* L.) specimens

Chemicals	Oven-dry density (kg/m ³)	Standart deviation	Homogeneity groups	Increases compared to control group (%)
Control	0,641	0,36	A	
ME	0,686	0,21	C	(+7,02)
MEC	0,651	0,28	AB	(+1,56)
MEF	0,682	0,33	C	(+6,39)
MEG	0,663	0,39	BC	(+3,43)
TE	0,681	0,19	C	(+6,24)
TEC	0,648	0,20	A	(+1,09)
TEF	0,655	0,34	AB	(+2,18)
TEG	0,649	0,15	A	(+1,24)

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.

In our study, oven-dry density values were higher in all coated test groups than in the control group. The specimens coated with ME had the maximum oven-dry density value (0,686 kg/m³) in the experiment, while the control group had the lowest oven-dry density value (0,641 kg/m³). The homogeneity group (C) in our investigation included ME, MEF, and TE, and there was no statistically significant difference between them. In the AB group, the MEC and TEF samples did not significantly alter the results.

Additionally, compared to specimens coated with their derived nanocomposite materials, MEC, MEF, MEG, TEC, TEF, and TEG, specimens coated with ME and TE showed higher oven dry density values. The oven-dry density values of specimens coated with ME and its derivatives, MEC, MEF, and MEG, were found to be higher than those of specimens coated with TE and its derivatives, TEC, MEF, and TEG. After covering specimens of oriental beech (*Fagus orientalis* L.) with epoxy resin, Altay measured the oven-dry densities of the specimens (Sikafloor 156) (Altay 2022a). The coated specimens did better than the group under control. The oven-dry density of wood coated with biobased epoxide amine nanocoatings was examined by Kabasakal *et al.* (2023). They discovered that the oven-dry density of the wood was raised

by the plant oil-based nanocomposite resin coating method. As our work involved coated specimens with an oven-dry density increase, the results aligned (Altay 2022a, Kabasakal *et al.*2023)

Considering the effects that wood material is exposed to during its use, the physical properties of wood are of great importance. The density of wood material is closely related to many properties of wood such as thermal, acoustic, gluing, resistance, drying, impregnation and processability properties (Türkyılmaz and Vurdu 2005). The density of the wood mostly determines its many functions. For example, compared to low-density wood, high-density wood has more strength, flexibility, and surface hardness. It provides stronger protection against corrosive impacts. Softwood might sometimes be advantageous since it facilitates processing and causes less shrinkage and swelling. This is possible when the wood is light (Örs and Keskin 2008). Where a wood product can be used depends on its density. Wood with a high density offers superior resistance. It resists abrasions rather well. Better processing properties are another benefit of low-density materials (Kollmann and Cote 1968, Bektaş and Güler 2001). As the density of the coated specimens in our experiment rose, it may be expected that the resistance properties and resistance to abrasions would also rise.

Water absorption

Table 2 displays the water absorption (WA) values of the coated oriental beech (*Fagus orientalis* L.) specimens and the control group. Table 3 shows the reduction in the coated species' WA levels as compared to the control group.

Table 2: WA values of oriental beech (*Fagus orientalis* L.) specimens.

Chemicals	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	42,05	D	48,42	B	56,38	D	62,26	C	75,79	C	80,92	E	83,52	D
ME	34,91	ABCD	46,05	B	54,88	BC	62,05	A	69,56	B	78,61	CD	79,45	BC
MEC	29,72	ABC	39,50	AB	49,40	AB	59,33	B	67,13	AB	75,16	BC	75,72	A
MEF	27,04	AB	39,39	AB	51,50	BC	61,22	A	71,51	B	78,50	CD	79,26	BC
MEG	25,49	A	32,35	A	44,77	A	55,04	A	67,13	AB	75,02	BC	77,46	AB
TE	31,96	ABC	41,16	AB	48,95	B	56,40	A	66,45	A	76,23	C	78,56	B
TEC	32,33	CD	48,34	B	56,00	D	60,46	BC	70,00	B	76,02	C	77,13	AB
TEF	38,66	D	47,98	B	55,59	C	60,34	BC	67,46	AB	71,12	A	76,18	A
TEG	36,04	BCD	47,14	B	55,45	C	61,75	C	67,52	AB	73,61	AB	78,39	B

H.G.: Homogeneity groups, 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: Epoxidized tung oil, TEC: Epoxidized tung oil with added carbon nanotubes, TEF: Epoxidized tung oil with added fullerene, TEG: Epoxidized tung oil with added graphene.

The control group in our study absorbed the most water overall during all water absorption times. The water absorption of spruce treated with tung oil was compared to that of control specimens by Humar and Lesar (2013). After soaking the wood for seven days, they discovered that the control specimens absorbed more water than the wood treated with tung oil. The epoxidization procedure using tung oil achieved decreased water absorption compared to the control group during all water absorption periods, which is consistent with the findings of Humar and Lesar (2013). Following the 62 h and subsequent WA periods, there was a statistically significant difference in the WA values between the coated all test groups and the control group. After 126 h of water absorption, the results showed that the ME, TE, and their nanocomposite derivatives absorbed less water than the control group. The results of the study showed that MEC-coated specimens were the most effective at inhibiting the absorption of water. Research has produced waterborne polyurethanes (PUDs) from a variety of monomers derived from vegetable oils, including internal emulsifiers and polyols (Liang *et al.*2018, Liu *et al.*2019, Liang *et al.*2019). Altay *et al.* (2024) examined the water absorption (WA) of oriental beech (*Fagus orientalis* L.) wood coated with a novel bio-based epoxide-amine (EP) and the nano-composite coating derivatives that resulted from the reactions of epoxy-functionalized tung, such as fullerenes, graphene, and carbon nanotubes. Their results showed

that, in comparison to control specimens, coated specimens' water absorption decreased after 200 hours of water absorption. The water absorption levels of oriental beech (*Fagus orientalis* L.) coated with bio-based epoxide amine nanocoatings were investigated by Kabasakal *et al.* 2023. Epoxy-coated samples performed better than control samples in terms of resistance to water absorption during all water absorption periods, according to the results of the water absorption (WA) test. The results of our investigation are in agreement with those of Kabasakal *et al.* (2023) and Altay *et al.* (2024).

Table 3: Decreases of water absorption values of wood specimens compared to the control group

Chemicals	Decrease in water absorption values compared to the control (%)						
	2h	6h	14h	30h	62h	94h	126h
Control							
ME	(-16,97)	(-4,89)	(-2,66)	(-0,33)	(-8,22)	(-2,85)	(-4,87)
MEC	(-29,32)	(-18,42)	(-12,38)	(-4,70)	(-11,42)	(-7,11)	(-9,33)
MEF	(-35,69)	(-18,64)	(-8,65)	(-1,67)	(-5,64)	(-2,99)	(-5,10)
MEG	(-39,38)	(-33,18)	(-20,59)	(-11,59)	(-11,42)	(-7,29)	(-7,25)
TE	(-23,99)	(-14,99)	(-13,17)	(-9,41)	(-12,32)	(-5,79)	(-5,93)
TEC	(-23,11)	(-0,16)	(-0,67)	(-2,89)	(-7,63)	(-6,05)	(-7,65)
TEF	(-8,06)	(-0,90)	(-1,40)	(-3,08)	(-10,99)	(-12,11)	(-8,78)
TEG	(-14,29)	(-2,64)	(-1,64)	(-0,81)	(-10,91)	(-9,03)	(-6,14)

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: Epoxidized tung oil, TEC: Epoxidized tung oil with added carbon nanotubes, TEF: Epoxidized tung oil with added fullerene, TEG: Epoxidized tung oil with added graphene.

These PUDs are widely utilized in sealants (Kang *et al.*2018), sound-absorbing foams (Meng *et al.*2021), adhesives (Dodangeh *et al.*2020), flexible electronic devices (Ren *et al.* 2020, Jia *et al.*2021), coatings (Cheng *et al.*2016, Cui *et al.*2020), and adhesives (Meng *et al.*2021). Rather than using organic solvents, they employ water. Remarkably, novel triglyceride structures and long, flexible fatty acid chains added to the polyurethane backbone result in incredibly flexible and water-resistant polymer films (Zhang *et al.*2013, Alberto *et al.*2021). A hybrid epoxy composite with a 33 % weight reinforcement concentration was developed by Jain and Gupta (2018) and is partially reinforced with teak and sal wood flooring. The findings indicate that teak and sal wood absorb less water as a result of hybridization. Our findings,

which show that applying epoxidation using plant-based chemicals reduces water absorption compared to the control group, are in line with those of Jain and Gupta (2018). The control group's water absorption was 83,52 % after the 126 h water absorption period. Additionally, the rate of water absorption ranged between 76,18 % and 78,56 % in the specimens epoxidized by adding tung oil and its derivatives, whereas it varied between 75,72 % and 79,45 % in the specimens epoxidized by adding moss and its derivatives. Bisphenol C is used in the synthesis of polyetheramide polyols from fatty amides of mahua oil in another study by Raychura *et al.* (2018). Low surface wettability was found in the coating performance investigation, which may lengthen the shelf life of the wood substrate. Plant oils are typically employed for their ability to resist water; when applied for wood preservation, this lowers the amount of free water in the wood while having little effect on the diffusion of bound water into the cellwalls. As a result, there may be a minor increase in the dimensional stability of the wood (Paajanen and Ritschkoff 2002). Hyvönen *et al.* 2006 state that natural oils like linseed oil and tall oil can prevent wood from absorbing water (Hyvönen *et al.* 2006). Extended oil treatments reduce the water-absorbing capacity of sapwood. Prolonged oil application processes are fundamentally water-repellent, even if they retain a significant amount less oil than pure long oil. Tomak and Yıldız (2012) state that vegetable oils can create a barrier on the surface of wood cells to stop water absorption. Consequently, these oils have a lot of potential in the wood protection industry. Since no chemical association between oil and wood has been found, linseed oil impregnation could only reduce the water adsorption rate and not the final moisture content (Tomak and Yıldız 2012). Kaymakçı (2016) looked at how well wood polymer nanocomposites based on carbon nanotubes and polypropylene absorbed water. Their research led them to the conclusion that wood polymer nanocomposites absorb less water when carbon nanotubes are added. In terms of water repellency and anti-swelling effectiveness, epoxidized linseed oil-treated wood has considerably surpassed linseed oil-treated wood (Kaymakçı 2016).

Research has demonstrated the effectiveness of wax and vegetable oil emulsions as water repellents (Temiz *et al.* 2013). Because the vegetable oil-based epoxidation coating procedure lowers the specimens' water absorption at the conclusion of the 126-hour WA period in our inquiry, our results are comparable to those of the previously mentioned studies.

Compression strength parallel to the fiber (CSPG)

Table 4 displays the CSPG values for the coated oriental beech (*Fagus orientalis* L.) specimens and the control group.

Table 4: The CSPG values of oriental beech (*Fagus orientalis* L.) specimens.

Chemicals	CSPG (kg/cm ²)	Standard deviation	Homogeneity groups	Increases compared to control group (%)
Control	57,73	3,91	A	
ME	59,75	4,13	AB	(+2,24)
MEC	58,42	2,75	A	(+1,19)
MEF	58,53	4,09	A	(+0,15)
MEG	60,46	4,37	AB	(+3,45)
TE	60,16	6,55	AB	(+2,94)
TEC	59,34	2,15	AB	(+1,54)
TEF	60,40	2,77	AB	(+3,35)
TEG	60,12	4,09	AB	(+2,87)

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: Epoxidized tung oil, TEC: Epoxidized tung oil with added carbon nanotubes, TEF: Epoxidized tung oil with added fullerene, TEG: Epoxidized tung oil with added graphene.

The control group's CSPG values were 57,73 kg/cm², while the MEG coated specimens had the highest values in the study, measuring 60,46 kg/cm². Every coated test specimen had CSPG values that were higher than those of the control group. However, the CSPG values of the coated test groups and the control group did not differ statistically significantly. Using carbon

nanoparticles, Jeffamine D2000, and tung oil-based epoxide resin (Babahan-Bircan *et al.* 2023) effectively cured graphene, CNT, and fullerene for the first time. All of the cured coatings exhibited robust mechanical properties, such as impact resistance, adhesion to crosshatch, and hardness. Compared to epoxide-amine nanocoatings, epoxide-amine coatings frequently displayed inferior mechanical properties.

It was shown in a different study that *Calotropis gigantea* enhanced the mechanical properties of epoxy composites reinforced with jute fibers, including their ultimate flexural, tensile, and compressive strengths (Garai *et al.* 2005).

The CSPG characteristics of wood coated with biobased epoxide amine nanocoatings on oriental beech (*Fagus orientalis* L.) were examined by Kabasakal *et al.* 2023. The vegetable oil-based nanocomposite resin coating technology shown favorable qualities against mechanical effects on wooden materials, as per the CSPG findings. The mechanical properties of wood-epoxy polymer composites are influenced by the micromechanical properties of both epoxy polymers and wood cell walls. The molecular makeup of the functional groups may have an impact on this characteristic in addition to crosslinking (Vinod *et al.* 2018). Vinyl and epoxy resins have similar mechanical characteristics. They are frequently mixed with other materials, including carbon fiber, to produce high-performance composites (Dang *et al.* 1997, Wang *et al.* 2010). When the commercial epoxy agent Sika floor 156 was applied to oriental beech (*Fagus orientalis* L.) wood, specimens coated with epoxy showed higher CSPG values than the control group (Altay *et al.* 2022b). Our study's results concur with those of Kabasakal *et al.* (2023) and Zhou *et al.* (2015).

The present study demonstrates that bio-based epoxy resins can serve as promising alternatives to petroleum-derived systems in wood coating applications. The use of natural epoxides not only aligns with environmental sustainability goals but also provides comparable or improved performance in terms of mechanical and physical properties of treated wood. Notably, the

application of bio-based epoxy coatings significantly reduced water absorption (WA) of the wood specimens, indicating enhanced barrier properties. This finding aligns with previous reports which emphasize that the crosslinked network formed by epoxies limits moisture diffusion and improves dimensional stability (Mallakpour *et al.* 2015). Furthermore, coated specimens exhibited increased oven-dry density and improved parallel-to-grain compressive strength (CSPG) compared to uncoated controls. Overall, the findings confirm that bio-based epoxy coatings not only offer a sustainable solution for wood protection but also contribute to improved mechanical resilience and long-term durability.

Conclusions

Bio-based epoxy resin (ETO), derived from epoxidising tung oil, was cured with bio-based moss oil and turpentine oil 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 surface. In this study, the use of moss oil and turpentine oil as hardeners for epoxy resin represents a novel approach. The addition of carbon nanoparticles to bio-based epoxy-oil and epoxy-turpentine oil systems has resulted in the formation of nanocomposite derivatives. The new bio-based coatings and their nanocomposite derivatives were examined and evaluated for their mechanical and physical characteristics.

The findings indicated that the oven-dry density values of oriental beech (*Fagus orientalis* L.) exhibited an increase when nanocomposite derivatives of ME and TE (derived from seaweed oil/turpentine oil) were employed. Epoxidized plant oil-coated specimens consistently outperformed the control group in terms of water repellency. All coated test specimens had

CSPG values that were higher than the control group's. The CSPG levels of the coated test group and the control group did not, however, differ statistically significantly.

Consequently, it has been demonstrated that the application of innovative bio-based nano-coatings enhances the mechanical and physical properties of oriental beech (*Fagus orientalis* L.) wood. In comparison to the control group, the test specimens exhibited reduced water absorption values and elevated oven-dry density and compressive strength values.

In conclusion, the application of innovative biobased nano-coatings has been demonstrated to enhance the mechanical and physical properties of oriental beech (*Fagus orientalis* L.) wood. In comparison to the control group, the test specimens exhibited reduced water absorption values and greater oven-dry density and compression strength values. Furthermore, moss oil and turpentine oil can be used as an epoxy hardener, thereby reducing the environmental impact. The utilisation of bio-based materials allows for the conservation of energy and petrochemical raw materials. Overall, this study demonstrates that natural oil-derived curing agents such as moss oil and turpentine oil represent promising alternatives to conventional petroleum-based hardeners in epoxy coating systems. The developed bio-based nanocomposite coatings not only improve mechanical and physical performance of wood materials but also contribute to the advancement of sustainable coating technologies. These findings provide a foundation for future research focused on optimizing bio-based curing mechanisms and scaling environmentally friendly epoxy systems for industrial applications.

Authorship contributions

I.B.B.: Supervision, experimental, analyzing data, writing-original draft preparation. Ç.A.: Experimental, interpretation of the analyzed data, writing and reviewing of the manuscript. B.K.: Experimental, writing and reviewing of the manuscript. S.E.: Experimental, writing and

reviewing of the manuscript. H.T.: Interpretation of the analyzed data. M.Ç.: Experimental E.
K.: Experimental

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Declaration of interest

The authors declare no conflict of interest.

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