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Original Article

Some Physical, Biological, Hardness, and Color Properties of Wood Impregnated with Propolis

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ABSTRACT

Propolis is a bee product collected by honeybees from various tree species in nature. It has antifungal, antibacterial, antioxidant, and anticancerogenic properties. Recently, propolis has been used in wood protection area because it has antifungal properties and is a natural and environmentally friendly material. In this study, some deciduous wood species were treated with methanolic propolis extracts. Propolis solutions were prepared by dissolving propolis in methanol in concentrations of 0.5%, 2.5%, 5%, and 8%. These solutions were forced deep into the Scots pine (*Pinus sylvestris* L.), fir (*Abies nordmanniana*), and spruce (*Picea orientalis* L.) woodblocks under vacuum and pressure. Water uptake and water-repellent efficiencies of the woodblocks were tested at 2-, 4-, and 20-hour immersion periods. Sample blocks were tested against wood-destroying house borer (*Hylotrupes bajulus*) (Coleoptera, Cerambycidae) larvae for 21 weeks. In addition, color changes of woodblocks treated with propolis were determined. According to results obtained in the study, the highest water-repellent efficiency was 61.4% in propolis-impregnated spruce wood at concentration level of 8% during the 2-hour period. These results show that propolis extracts could be used as hydrophobic material for wood. Larvae mortality rates indicated that 8% concentration level was not adequate to overcome *H. bajulus* larvae or propolis is not effective against *H. bajulus* larvae. While L^* value decreases with the increase of impregnation rate, a^* and b^* values increase in all wood types. As the impregnation rate increased, the total color difference also increased.

Keywords: Color change, Hylotrupes bajulus, larvae, propolis, shore D hardness, wood, water absorption

Introduction

When wood material is used without any protection, it generally deteriorates within 3–5 years, depending on wood species and its exposure to soil and environmental conditions in using area. On the other hand, chromated copper arsenate (CCA)-treated wood is 10–20 times stronger than untreated wood (Barton, 2014). Chromated copper arsenate has been used widely to protect wood structures (e.g., decks, porches, landscaping, playground equipment, picnic tables, garden-bed borders, and docks) from rotting due to moisture, insects, and microbial agents until 2002. In 2002, the U.S. Environmental Protection Agency (EPA) announced that the use of CCA should be limited and it should be voluntarily withdrawn in residential areas. Wood material impregnated with CCA posed an environmental risk as it leached from wood with rainwater, and hazardous chemicals such as chromium and arsenic leaked into the environment, putting the health of workers at risk (Barraj et al., 2009). Previous studies showed that since CCA can be leached from CCA-treated wood, groundwaters and soils contacting with CCA are contaminated by arsenic, chromium, and copper (Shibata et al., 2007).

After EPA limited using CCA in residential areas, researchers have developed chemicals to protect wood. However, hazardous chemicals are not preferred due to environmental threats and legal pressures (Temiz et al., 2014). Nowadays, there is an increased need to extend the service life of wood and wood products with green natural wood preservatives. These ecofriendly wood preservatives are based on natural compounds that have low or no toxic effects on humans and environment (Woźniak et al., 2019b). Plant extracts play an important role in wood protection. Several studies have demonstrated that the use of extracts makes the wood very durable as they are potential eco-friendly preservatives (Brocco et al., 2017; Mohammed et al., 2016; Tascioglu et al., 2013).

Propolis is a bee product collected by honeybees from leaves and buds of various trees in nature (Woźniak et al., 2019b). Propolis has antibacterial, antioxidant, and anticarcinogenic properties. It has antifungal properties due

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to the presence of phenolic substance. The studies revealed that propolis is composed of more than 300 components. Although its composition varies depending on the collection region, it contains 50% resin, 30% wax, 5–10% essential compounds, 5% pollen, and numerous other chemicals (Huang et al., 2014). Due to its waxed structure, bees use propolis to cover cracks in the hive and to protect them from temperature and humidity.

Recently, with the growing importance of propolis as an active substance, its wood preservation properties have also been investigated (Woźniak et al., 2019a). Jones et al. (2011) examined the antifungal activity of propolis extract against brown rot fungi (*Coniophora puteana*), Ratajczak et al. (2018) reported that when wood material is treated with propolis extract, caffeine, and silicon compounds, the combination inhibited the growth of *C. puteana* on wood. On the other hand, Budija et al. (2008) have investigated the surface contact angle of wood impregnated with propolis and proved that propolis exhibits hydrophobic properties by forming films on the wood surface.

To date, though some studies have demonstrated the efficiency of propolis against wood-decaying fungi, however, the efficacy of Turkish propolis against wood-destroying insects, color change of wood, shore *D* hardness, water repellent efficiency (WRE), and water absorption (WA) of wood have not been studied. In our previous study, it was reported that propolis can act as a natural wood preservative due to its antifungal properties (Akcay et al., 2020). Similarly, in this study, the efficacy of some wood species impregnated with propolis against *Hylotrupes bajulus* larvae, the WA, WRE of wood, shore D hardness, and color properties were investigated for the first time.

Methods

Wood Samples

Scots pine (*Pinus sylvestris* L.), fir (*Abies nordmanniana*), and spruce (*Picea orientalis* L.) wood specimens were prepared from sapwood in dimensions of 20 × 20 × 10 mm^3 for determination of WA/WRE and 50 × 25 × 15 mm^3 (longitudinal × radial × tangential) for larvae and color tests. The densities of Scots pine, fir, and spruce wood species were 0.46, 0.36, and 0.36 g/cm^3, respectively. The ring widths were an average of 5 mm for each wood species.

Raw Propolis Extraction and Total Phenolic Content

Raw propolis was supplied by Anatolian propolis as the brands of Bee &You (BEE'O®)/SBS Scientific Bio Solutions Inc. (Istanbul, Turkey). After grinding the raw propolis frozen in the freezer, methanolic extract was prepared. About 100 g of propolis was extracted in 95% concentration using 250 mL of methanol on a shaker for 24 hours. After filtering, the solution is accepted as stock solution as 95% and then four different dilutions were prepared with methanol as 0.5%, 2.5%, 5%, and 8%.

Total phenolic content of the stock propolis extracts was measured by Folin Ciocalteu method (Singleton et al., 1999). The results were calculated as gallic acid equivalents (GAE) (g/100 mL).

Impregnation of the Samples

Scots pine, fir, and spruce wood samples prepared for larva resistance tests, WA, color, and hardness tests were treated with methanolic propolis extracts at concentration levels of 0.5%, 2.5%, 5%, and 8%. Concentration levels were specifically selected from the same concentrations in which our previous study showed efficacy against wood-decaying fungi. The impregnation process was carried out in a cylindrical vacuum pressure impregnation tank. All samples were dried at 103 ± 2°C for 24 hours, and the dried weights (M_0) were recorded before the impregnation process.

The test specimens were placed in the propolis solution and kept under a vacuum of 0.079 Mpa for 20 minutes and then the pressure was applied at 1.21 Mpa for 15 minutes. After impregnation, the samples were immediately re-weighed (M_1). The retention (R) amount of the samples was calculated using Eq. 1. All samples were conditioned at 20 ± 2°C and 65 ± 2% RH for 4 weeks after treatment:

$$R = \frac{(M_1 - M_0) \times C}{V} \times 10 \text{kg}/\text{m}^3 \quad (1)$$

Here, M_0 (g) is the weight before treatment, M_1 (g) is the weight after treatment, C is the solution concentration, and V is the volume (m^3) of the woodblocks.

Water Absorption and Water Repellent Efficiency

A total of 72 samples from Scots pine, fir, and spruce were used for WA and WRE tests. Control samples having the same annual rings and the samples deep treated with the propolis solution were dried to constant weight at 103 ± 2°C; their dry weights were determined on a balance having a sensitivity of 0.01 g. Test and control samples were immersed in water at 20 ± 1°C, and a heavy weight was put on them to ensure they remained below the water surface. The values of WA of the test and control samples were measured at the end of water-holding periods of 2, 4, and 20 hours. At the end of each period, the samples were removed from the water container and wiped off with a paper towel. The samples were weighed, and the amount of absorbed water (A_1) was recorded. The initial dry weight of the test sample or control sample (A_o) and the A_1 values were used to calculate the WA according to Eq. 2 for each test and control sample separately for each period (Broda, 2018):

$$\text{WA} = \frac{A_1 - A_0}{A_0} \times 100 \quad (2)$$

The WRE value was expressed as a reduction of the WAs of the wood samples impregnated with propolis in different concentrations compared to the control samples. The WRE was calculated using Eq. 3 for each test separately for each period.

$$\text{WRE} = \frac{WA_c - WA_t}{WA_c} \times 100 \quad (3)$$

Here, WA_c is the WA (%) of the control sample at the end of a specified period and WA_t is the WA (%) of the test sample at the end of a specified period.

Larvae Resistance Tests

Larva resistance tests were carried out according to the explanations specified in the second category in EN 47 (2016). This method is used for evaluating the effectiveness of the treatment solution against *H. bajulus* larvae. In the tests, Scots pine, fir, and spruce sapwood samples in dimensions of 50 × 25 × 15 mm were impregnated with propolis (2.5%, 5%, and 8%) and the untreated control samples were used. Ten replicates for each group were tested. Larval tests were performed using 50–60 mg mature larvae obtained by mating adult female and male *H. bajulus* insects under laboratory conditions in the Duzce University Forest Biology and Wood Protection Laboratory.

Holes of 3 mm width and 20 mm depth were drilled in the propolis-impregnated and non-impregnated wood samples. Larvae were introduced into the drilled holes in a head-down position. Test specimens were kept in an incubator test cabin at 27 ± 2°C and 80 ± 2% RH for 48 weeks. First examination was carried out after 24 weeks. At the end of the 48th week, all samples were then broken open, and the dead and live larvae were recorded. Larvae mortality rates (MR) were recorded in the control and propolis-treated samples:

$$MR = \frac{N_f}{N_t} \times 100 \quad (4)$$

Here, MR is larvae mortality (%), N_t is the number of total larvae introduced into the drilled holes of the woods, and N_f is the final number of dead larvae after the test.

Determination of Color and Shore *D* Hardness Properties

The protective chemicals applied to the wood material also cause some physical and chemical changes on the wood surface and therefore color and hardness tests were also carried out. The color parameters of wood species impregnated with propolis and control were measured using CS-10 colorimeter (CHN Spec, Jianggan District, Hangzhou City, China) (Figure 1b) with a measuring area of 8 mm. Color measurements were taken under the conditions of the standard illuminant D65 and 10° observer, as described by the Commission Internationale de L'éclairage (CIE L*a*b* standard) and according to ASTM D 2244-3. A CIELAB system, characterized by the tree axis L^*, a^*, and b^* was used (Ayata, 2019). The three-dimensional CIE L*a*b* color space is given in Figure 1c (Van Nguyen et al., 2019). The corresponding changes ΔL^*, Δa^*, and Δb^* after impregnation were determined by Eqs. 5–7, and the total color difference (ΔE^*) was calculated by Eq. 8:

$$\Delta L^* = L^*_{\text{impregnated}} - L^*_{\text{non-impregnated}} \quad (5)$$

$$\Delta b^* = b^*_{\text{impregnated}} - b^*_{\text{non-impregnated}} \quad (6)$$

$$\Delta a^* = a^*_{\text{impregnated}} - a^*_{\text{non-impregnated}} \quad (7)$$

$$\Delta E^* = \sqrt{\left(\Delta L^*\right)^2 + \left(\Delta a^*\right)^2 + \left(\Delta b^*\right)^2} \quad (8)$$

Shore D hardness was determined with Shore D meter device (stand: model Ld-J Loyka) by loading 5 kg weight according to ASTM D 2240 standard (Figure 1a). Ten tests were done for untreated controls and samples impregnated with different concentration rates (0.5%, 2.5%, 5%, and 8%).

SEM Analysis

SEM (Scanning Electron Microscope) Analysis analyses were performed at Duzce University Scientific and Technological Researches Application and Research Center (Duzce, Turkey). SEM images were taken using a Quanta FEG 250 instrument (FEI Europe B.V., Eindhoven, Netherlands). The wood samples were mounted onto aluminum stubs with double-sided carbon tape, and mounted specimens were coated with 10 nm gold film using sputter coater (Desk V-Standard, Denton Vacuum, LLC, NJ, USA) before analyses. Surface morphologies of the samples were investigated with an SEM Quanta FEG 250, which used an electron acceleration voltage of 10 keV. The measurements were taken using cross-section of the woods.

Statistical Analyses

Statistical Package for the Social Sciences (IBM SPSS Corp., Armonk, NY, USA) v.21 software was used to evaluate statistical differences among the means. Duncan's mean separation test was applied at $\alpha \leq .05$ level for the variables determined to have differences based on analysis of variance results.

Results

The total amount of phenolic substance in the stock propolis used in the study was found as 13.56 g GAE/100 mL. Polyphenolic substances are the most effective bioactive compounds of propolis, and they have a very large number of biologically active properties. Studies show that polyphenols have antioxidant, antimicrobial, antitumoral, anti-inflammatory properties, and therefore, propolis is used as a food supplement (Pobiega et al., 2019; Turkut et al., 2019).

Retention Values of the Wood Species Impregnated with Propolis

Retention values of WA and larvae resistance test samples were given in Table 1. When the retention values were examined, the retention value of the wood species increased as expected, as the propolis concentration increased in the WA and larvae specimens in the impregnation process. The highest retention value was found in the Scots pine and larvae

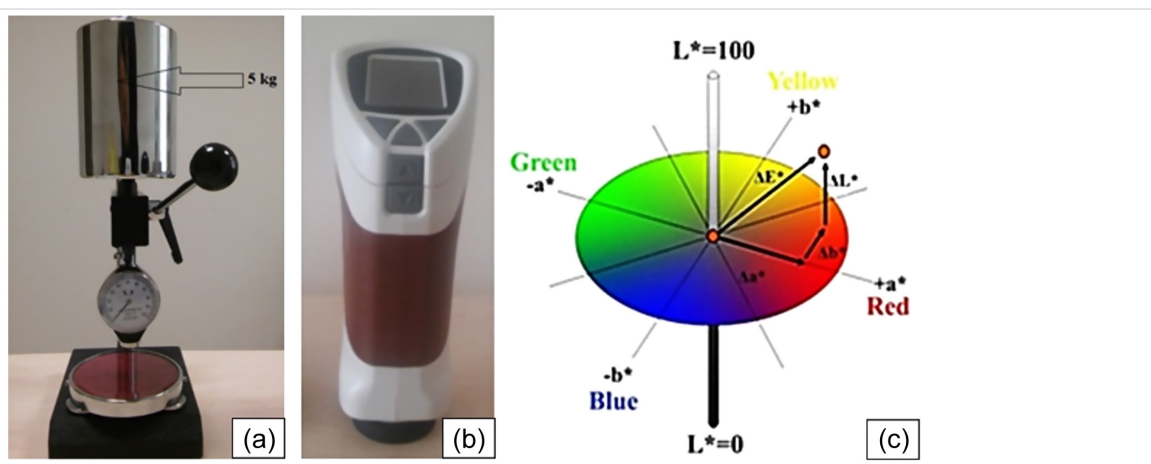

Figure 1.
*Shore D Hardness (a), CS-10 Colorimeter (b) and the Three-Dimensional CIE L*a*b* Color Space (Nguyen et al., 2018) (c).*

Table 1.
Retention Values of Water Absorption and Larvae Resistance Test Samples (kg/m³)

	Water Uptake Specimens Retention			Larva Test Specimens Retention		
Concentration (%)	Scots Pine	Fir	Spruce	Scots Pine	Fir	Spruce
0.5	2.4 (0.06) *a*	2.21 (0.11) *a*	2.27 (0.09) *a*	-	-	-
2.5	10.6 (054) *b*	12.09 (4.01) *b*	8.19 (2.70) *b*	14.44 (5.16) *a*	12.72 (1.11) *a*	2.74 (0.84) *a*
5	24.5 (1.59) *c*	22.15(1.16) *c*	16.77 (5.85) *c*	22.50 (3.95) *b*	26.48 (1.16) *b*	8.84 (4.11) *b*
8	37.0 (2.05) *d*	32.91(1.45) *d*	32.31 (3.16) *d*	39.04 (3.41) *c*	36.76 (5.28) *c*	14.30 (5.17) *c*

There is no statistically significant difference between the same italic letters in the same column ($p < .05$).
The values in parenthesis were standard deviations.

specimens. The retention values of spruce wood for insect specimens were found to be lower than that of pine and fir species. The fact that Spruce wood species contain resins compared to other wood species may have contributed to this result (Gjerdrum & Bernabei, 2007).

Water Absorption and Water Repellent Efficiency

Water absorption and WRE values of Scots pine, fir, and spruce wood impregnated with propolis are shown in Figures 2, 3, and 4, respectively. As it can be seen in Figure 2, untreated scots pine (control samples) absorbed 80.3% water during the 2-hour water immersion period, while the WA rate of the samples impregnated with propolis at concentration level of 8% was measured as 55.8%. Likewise, 89.0% water absorption rate was found in control samples and 66.4% in impregnated samples with 8% propolis in 20-hour period. Water repellent efficiency value was determined as 46.6% in 2-hour period compared to control samples. Even at the end of 20 hours, 36.9% WRE was obtained compared to the control samples. Although 34.9% WRE value was obtained in fir wood species compared to control samples during 2-hour water immersion period, this activity decreased to 8.3% after 20-hour period (Figure 3).

The reason for the high WRE values of wood species impregnated with propolis compared to the control samples is attributed to the presence of waxy and resinous substances with hydrophobic properties in the propolis structure (Huang et al., 2014). Propolis, on the other hand, forms a film layer on the wood surface after impregnation thereby forming a barrier against WA. It also increases the contact angle of water with the wood surface. Hence, water repellent activity value increases by hydrophobic properties gained by wood material (Ratajczak et al., 2018). Budija et al. (2008) applied propolis extract on wood surfaces with a brush which formed a thin film leading to gain of hydrophobic properties to wood. In recent years, environmental concerns have increased interest in protective chemicals such as non-biocidal, renewable, and water repellents (Humar & Lesar, 2013).

According to the results obtained from the current study, it has been revealed that the environmentally friendly propolis can be used effectively against the WA of the wood in service. Furthermore, it was reported that propolis was not leached from treated wood due to its waxed nature as in our previous study (Akcay et al., 2020). It is an important issue for water repellents that preservatives are not leached from wood. Wax emulsions are the most important water repellents used in the wood preservation (Lesar & Humar, 2011).

SEM Analysis Results

The pictures obtained as a result of the SEM analysis are shown in Figure 5. When SEM analysis results are examined, it is seen that cell lumens in cross-sections of all tree species are coated with propolis wax. SEM images confirmed the hypothesis that propolis helps in gaining water repellency to the wood by forming a coating on the wood surface when compared to the untreated control samples. It has also been

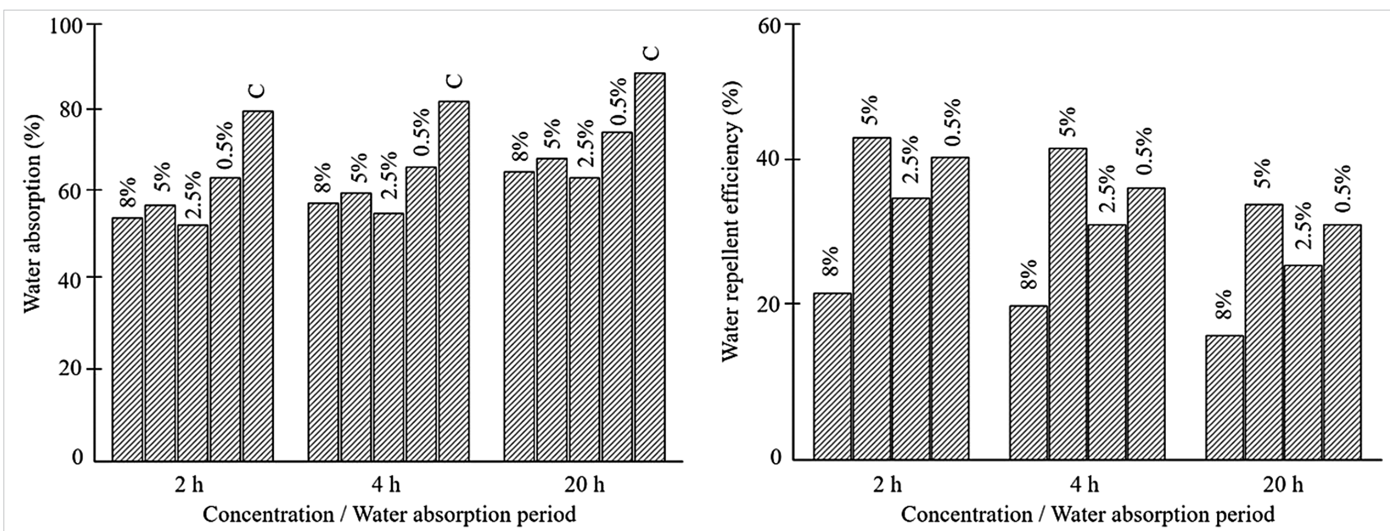

Figure 2.
WA and WRE of Scots Pine (Pinus sylvestris L.) Treated with Propolis at Different Water Absorption Periods.
WA = water absorption; WRE = water repellent efficiency.

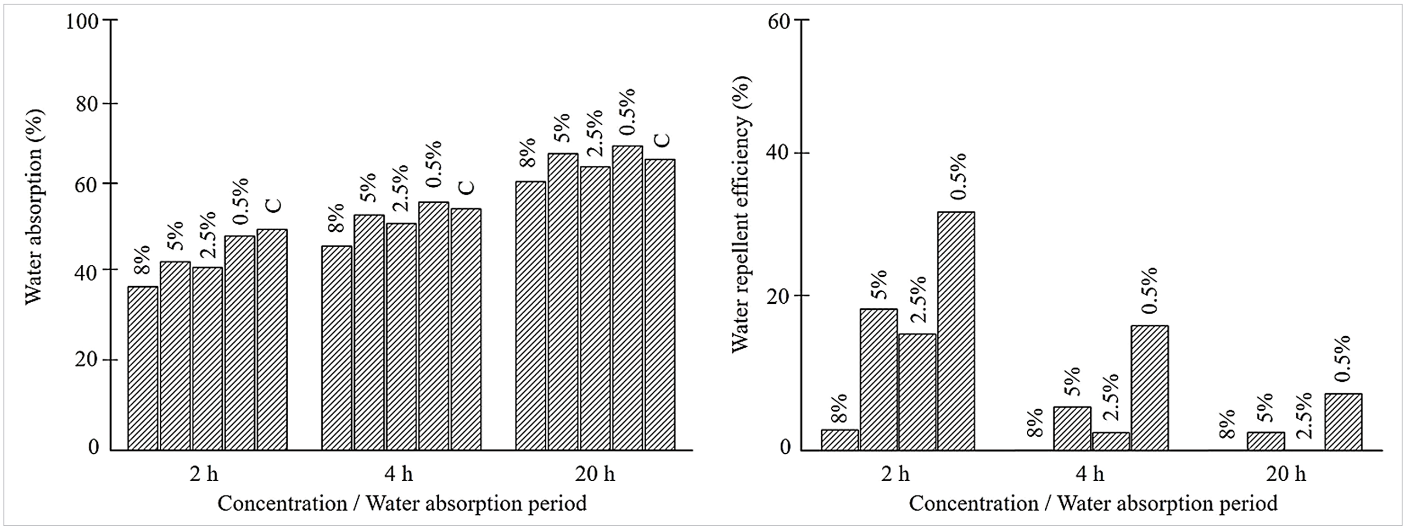

Figure 3.
WA and WRE of Fir (Abies nordmanniana) Wood Treated with Propolis at Different Water Absorption Periods.
WA = water absorption; WRE = water repellent efficiency.

shown in previous literature studies that propolis fills the cell lumen, decreasing the moisture rate in the cell and increasing the resistance against fungi and water repellency (Akcay et al., 2020; Ratajczak et al., 2018; Woźniak et al., 2019b).

Larvae Mortality Rates

H. bajulus beetle (old house borer) is one of the major wood-destroying insects. It generally feeds on softwoods. For this reason, larva experiments were carried out in three softwood species. At the end of the experiment, all control and impregnated samples were broken, and the MRs were calculated and presented in Table 2. While 100% live larvae was determined in Scots pine and spruce control woods, 80% was determined in fir wood. According to EN 47 standard test method, if more than 70% of larvae in control samples are alive, the larvae resistance tests are acceptable and valid. When the propolis concentration level reached 8%, larvae MR was determined as 20% in Scots pine wood. Larva mortality rates indicated that propolis was not effective against *H. bajulus* even at the highest concentration level (8%) in the present study.

In recent years, especially after the 2000s, the use of CCA-impregnated wood has been accepted as an environmental risk and its use in residential areas has been limited. Instead of environmentally hazardous chemicals, more environmentally friendly plant extracts, tannins, resins, and waxes have been used. The use of plant extracts has gained importance especially in the protection of interior wooden materials. The effectiveness of plant extracts against wood-rotting fungi and insects has been demonstrated in many studies (Şen et al., 2017; Tascioglu et al., 2013). Propolis is an environmentally friendly material, and recent studies accepted its wood preservative potential (Wozniak et al., 2020).

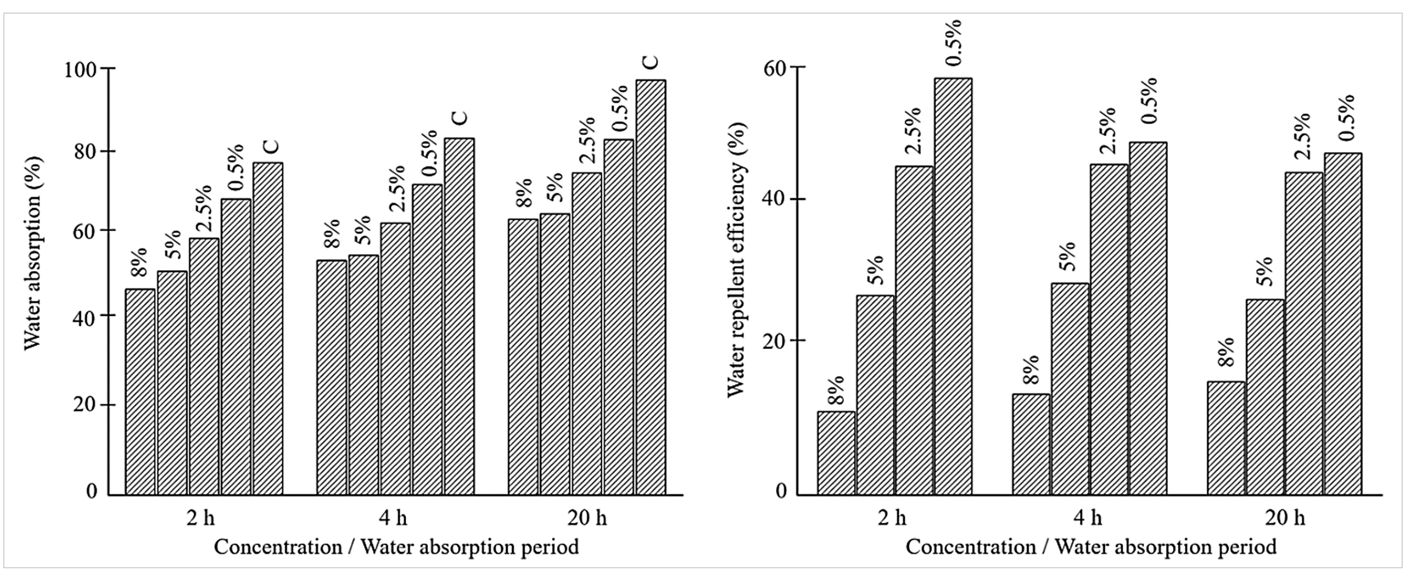

Figure 4.
WA and WRE of spruce (Picea orientalis L.) Wood Treated with Propolis at Different Water Absorption Periods.
WA = water absorption; WRE = water repellent efficiency.

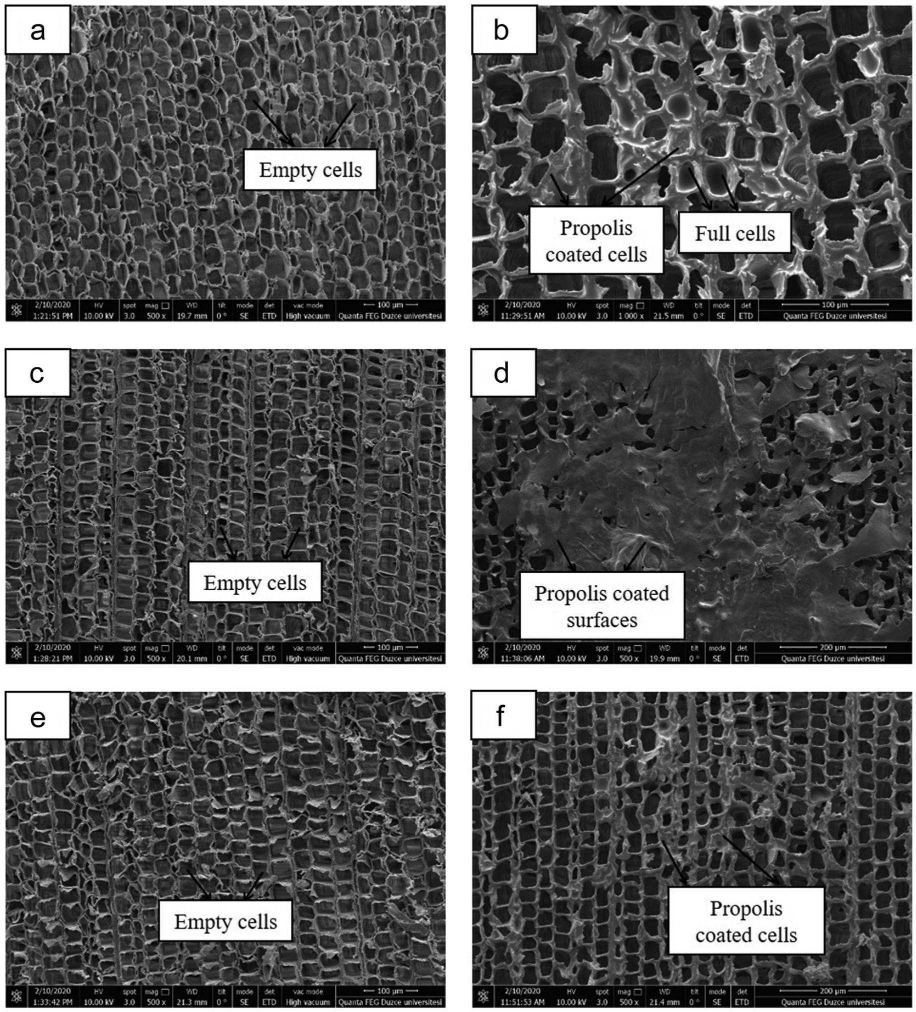

Figure 5.
SEM Images of Propolis-Treated Woods on Cross-Section (a: Scots Pine Control, b: Scots Pine Wood Treated with 8% Propolis, c: Fir Wood Control, d: Fir Treated with 8% Propolis, e: Spruce Wood Control, f: Spruce Wood treated with 8% Propolis.

To date, the effectiveness of propolis against insects damaging wood material has not been investigated. This study is the first to demonstrate the efficacy of wood material impregnated with propolis against old house borer insects (*H. bajulus*) that cause severe damage to wood. However, the current study showed that the same concentration level of propolis was not sufficient for *H. bajulus* larvae death or propolis has no toxicity against these insect larvae. That is why higher concentration level of propolis is needed to overcome *H. bajulus* larvae. The absence of larval deaths in spruce wood treated with propolis is attributed to the more difficult impregnation of this wood species compared to Scots pine (Yalinkiliç et al., 1996).

Table 2.
Larvae Mortality Rates in Woods Treated with Propolis

Weeks	Concentration (%)	Scots Pine (%)	Fir (%)	Spruce (%)
24	Control	0	20	0
	2.5	10	0	0
	5.0	0	0	0
	8.0	20	0	0
48	Control	0	20	0
	2.5	20	0	0
	5.0	0	20	0
	8.0	20	0	0

At the end of the larval experiments, it was shown that the larvae differed in destroying the control samples. Particularly, scots pine and spruce wood control specimens were more damaged than fir control wood. Sivrikaya et al. (2015) and Yalçin et al. (2018) also reported that the development of *H. bajulus* larvae in the Scots pine was better than fir wood.

Color Change and Shore D Hardness of Wood Treated with Propolis

Variance analysis results for color parameters and shore *D* hardness are shown in Table 3. According to the determined results, lightness (*L**), red color (*a**) tone, and yellow color (*b**) tone values were found to be significant. When wood type (A) and Shore *D* hardness impregnated (B) were found to be significant, interaction (AB) was not obtained significant (Table 3).

Wood type, impregnation concentration rate, and single comparison are given in Table 4. The highest lightness and yellow color value for the wood type was obtained in Scots pine, while the highest red value was obtained

Table 3.
Variance Analysis Results for Color Parameters (L, a*, and b*)*

Test	Source	Sum of Squares	*df*	Mean Square	*F*	Sig.
*L**	Wood type (A)	212.604	2	106.302	433.935	.000*
	Impregnated (B)	1448.113	4	362.028	1477.834	.000*
	Interaction (AB)	144.152	8	18.019	73.555	.000*
	Error	33.071	135	0.245		
	Total	885 344.032	150			
*a**	Wood type (A)	20.970	2	10.485	70.852	.000*
	Impregnated (B)	202.311	4	50.578	341.779	.000*
	Interaction (AB)	29.671	8	3.709	25.063	.000*
	Error	19.978	135	0.148		
	Total	5464.736	150			
*b**	Wood type (A)	280.065	2	140.032	461.589	.000*
	Impregnated (B)	4268.919	4	1067.230	3517.911	.000*
	Interaction (AB)	254.647	8	31.831	104.924	.000*
	Error	40.955	135	0.303		
	Total	104 075.947	150			
Shore *D* hardness	Wood type (A)	278.973	2	139.487	7.447	.001*
	Impregnated (B)	801.427	4	200.357	10.697	.000*
	Interaction (AB)	224.093	8	28.012	1.496	.164**
	Error	2528.600	135	18.730		
	Total	259 604.000	150			

*Significant (according to $\alpha \leq .05$).
**Insignificant.

in fir wood. For the impregnation rate, the highest lightness value was obtained in control samples, whereas highest red and yellow tones were obtained in impregnated samples with 8% concentration (Table 4).

Average values of CIE *L*a*b** parameters on Scots pine, spruce, and fir propolis-impregnated and non-impregnated woods are given in Table 5.

While the lightness (*L**) value decreases with the increase in impregnation rate in all wood species, it was concluded that the red color (*a**) tone, yellow color (*b**) tone values, and shore *D* hardness increased (Table 5).

In a study conducted by Soylamis (2007), protim WR-230 and imersol aqua chemicals were compared with impregnated samples and

Table 4.
Wood type, Impregnation Concentration Rate, Single Comparison for Color Parameters, and Shore D Hardness

		Lightness (*L**)		Red Color (*a**) Tone		Yellow Color (*b**) Tone		Shore *D* Hardness	
Wood Type	*N*	Mean	*HG*	Mean	*HG*	Mean	*HG*	Mean	*HG*
Scots pine	50	77.99	A*	5.48	C	27.20	A*	43.16	A*
Spruce	50	77.11	B	5.78	B	23.91	C	40.78	B
Fir	50	75.14	C	6.38	A*	26.05	B	39.94	B
		Lightness (*L**)		Red Color (*a**) Tone		Yellow Color (*b**) Tone		Shore *D* Hardness	
Impregnation ratio (%)	*N*	Mean	*HG*	Mean	*HG*	Mean	*HG*	Mean	*HG*
Control	30	80.61	A*	4.33	E	17.96	E	37.73	C
0.5	30	79.57	B	4.93	D	21.82	D	40.37	B
2.5	30	76.98	C	5.95	C	25.91	C	41.33	B
5.0	30	74.12	D	6.61	B	30.70	B	42.23	B
8.0	30	72.46	E	7.59	A*	33.21	A*	44.80	A*

N = number of measurement; *HG* = homogeneity group.
*Highest value.

Table 5.
*Average Values of CIE L*a*b* Parameters on Materials of Scots Pine, Spruce and Fir Propolis-Impregnated and Non-impregnated Woods*

Test	Wood Type	Impregnation Ratio (%)	*N*	Mean	Std. Deviation	*HG*	Minimum	Maximum
Lightness (*L**) Value	Scots Pine	Control	10	82.35	0.14	A*	82.10	82.54
		0.5	10	81.34	0.14	B	81.15	81.54
		2.5	10	77.49	0.21	F	77.17	77.80
		5.0	10	75.33	0.25	H	74.92	75.65
		8.0	10	73.45	1.13	J	72.03	74.82
	Spruce	Control	10	79.27	0.12	D	79.03	79.50
		0.5	10	78.79	0.10	E	78.60	78.92
		2.5	10	77.54	0.30	F	77.05	77.90
		5.0	10	75.31	0.35	H	74.83	75.93
		8.0	10	74.61	0.13	I	74.36	74.78
	Fir	Control	10	80.22	0.15	C	79.94	80.37
		0.5	10	78.58	0.41	E	77.50	78.95
		2.5	10	75.90	0.20	G	75.68	76.14
		5.0	10	71.71	1.04	K	70.09	72.84
		8.0	10	69.31	0.83	L	67.78	70.24
Red Color (*a**) Tone	Scots Pine	Control	10	3.93	0.07	H	3.82	4.00
		0.5	10	4.11	0.07	H	4.04	4.25
		2.5	10	5.66	0.19	E	5.41	5.95
		5.0	10	6.35	1.06	D	4.87	7.69
		8.0	10	7.37	0.41	B	6.81	8.30
	Spruce	Control	10	4.92	0.08	G	4.83	5.05
		0.5	10	5.51	0.06	EF	5.38	5.59
		2.5	10	5.76	0.25	E	5.48	6.15
		5.0	10	5.84	0.24	E	5.64	6.46
		8.0	10	6.89	0.25	C	6.54	7.33
	Fir	Control	10	4.16	0.08	H	4.06	4.29
		0.5	10	5.19	0.44	FG	4.15	5.59
		2.5	10	6.43	0.11	D	6.29	6.59
		5.0	10	7.63	0.61	B	7.04	8.67
		8.0	10	8.51	0.33	A*	8.00	8.92
Yellow Color (*b**) Tone	Scots Pine	Control	10	19.83	0.20	I	19.40	20.10
		0.5	10	21.14	0.18	H	20.93	21.41
		2.5	10	29.36	0.21	E	28.93	29.62
		5.0	10	32.60	0.26	C	32.33	33.26
		8.0	10	33.09	1.41	B	31.80	36.69
	Spruce	Control	10	17.42	0.27	J	16.92	17.81
		0.5	10	21.25	0.35	H	20.55	21.80
		2.5	10	21.61	0.52	H	20.99	22.53
		5.0	10	29.44	0.37	E	28.74	29.84
		8.0	10	29.81	0.40	DE	29.06	30.31
	Fir	Control	10	16.64	0.14	K	16.55	16.99
		0.5	10	23.07	0.80	G	21.26	23.77
		2.5	10	26.77	0.21	F	26.44	27.16
		5.0	10	30.05	0.84	D	29.01	31.69
		8.0	10	33.73	0.44	A*	33.24	34.51

N = number of measurement; *HG* = homogeneity group.
*Highest value.

Table 6.
Total Color Difference Results

Propolis Concentration (%)	Scots Pine				Spruce				Fir			
	Δ*L**	Δ*a**	Δ*b**	Δ*E**	Δ*L**	Δ*a**	Δ*b**	Δ*E**	Δ*L**	Δ*a**	Δ*b**	Δ*E**
0.5	−1.01	0.18	1.31	1.66	−0.48	0.59	3.83	3.90	−1.64	1.03	6.43	6.72
2.5	−4.86	1.73	9.53	10.84	−1.73	0.84	4.19	4.61	−4.32	2.27	10.13	11.24
5.0	−7.02	2.42	12.77	14.77	−3.96	0.92	12.02	12.69	−8.51	3.47	13.41	16.26
8.0	−8.90	3.44	13.26	16.34	−4.66	1.97	12.39	13.38	−10.91	4.35	17.09	20.74

control samples in Uludag fir (*Abies bornmulleriana* Mattf.) and chestnut (*Castanea sativa* Mill.) woods. According to the results, red and yellow tone values were reported to increase after impregnation.

In addition, some studies determined that lightness (*L**) values decreased for jabon (*Anthocephalus cadamba*) wood after treatment with polymerized merbau extractives (Malik et al., 2018), for scots pine (*P. sylvestris* L.) and alder (*Alnus glutinosa* subsp. barbata) wood samples after treatment with tanalith-E and celcure AC 500 chemicals (Kilinc, 2019), for Scots pine (*P. sylvestris* L.) and alder (*Alnus glutinosa* subsp. barbata) wood samples after treatment with CCA, ACQ-1900, ACQ-2200, Tanalith-E 3491, and wolmanit CX-8 chemicals (Temiz, 2005), and for Scots pine (*P. sylvestris* L.) wood samples after treatment with 2% aqueous solution of some copper-based chemicals such as celcure AC-500, wolmanit CX-8, and adolit KD-5 (Ustun, 2019). The findings obtained in the study showed compatibility with the literature.

The results of total color difference in scots pine, spruce, and fir wood impregnated with different concentration rates are shown in Table 6. According to Table 6, it was concluded that as the impregnation concentration rate increases, the total color difference increases in all tree species. For specimens impregnated at 8% concentration, the highest total color difference value was obtained in fir wood, followed by scots pine and spruce wood species. While the lightness value gave negative results, the red color (*a**) tone and yellow color (*b**) values gave positive results for all concentration ratios (Table 6).

Surface chemicals (varnishes, paint, etc.) applied to wood material have negative properties in terms of human health. It is always important for the environment and human health to use "natural dyes and chemicals with resistant properties" to avoid unnecessary use of volatile organic compounds. Propolis is an environmentally friendly chemical that is resistant to fungi. In addition to these properties, it will be important to use it instead of harmful chemicals (varnishes, paint, etc.) of its color tone.

Reasons for applying bleaching process in wood materials were to obtain lighter shades of the same color by preserving the fiber structure of the wood material, to increase the aesthetic value of the wooden product by keeping the colors under control on the wood material surfaces, to use various types of wood together in harmony when necessary and to provide economy, to ensure continuity of color and to change color in some wood species and to reduce the possibility of fading, to remove the discolorations and chemical stains caused by mold fungi and the discoloration

Table 7.
Average Values of and Shore D Hardness on Materials of Scots Pine, Spruce and Fir Propolis-Impregnated and Non-impregnated Woods

Wood Type	Impregnation Ratio (%)	*N*	Mean	Std. Deviation	*HG*	Minimum	Maximum
Scots pine	Control	10	38.10	4.43	CDE	33.00	43.00
	0.5	10	41.50	4.62	BCDE	35.00	49.00
	2.5	10	42.20	7.55	BC	33.00	54.00
	5.0	10	44.10	5.40	B	35.00	51.00
	8.0	10	49.90	6.54	A*	41.00	59.00
Spruce	Control	10	37.50	5.72	E**	30.00	46.00
	0.5	10	40.30	3.53	BCDE	33.00	44.00
	2.5	10	41.70	5.60	BCDE	36.00	47.00
	5.0	10	42.00	4.97	BCDE	36.00	51.00
	8.0	10	42.40	1.71	BC	40.00	44.00
Fir	Control	10	37.60	1.71	DE	36.00	40.00
	0.5	10	39.30	0.67	CDE	39.00	41.00
	2.5	10	40.10	0.99	BCDE	39.00	41.00
	5.0	10	40.60	0.52	BCDE	40.00	41.00
	8.0	10	42.10	1.45	BCD	40.00	44.00

N = number of measurement; *HG* = homogeneity group.
*Highest value.
** Lowest value

on the parts of the wood material in contact with the metals, to make the surface properties of the wood material more prominent and to obtain clearer, brighter, and cleaner surface treatments (Edwin & Carter, 1983).

Scots pine presented a significantly higher shore *D* hardness than the other species, followed by spruce and fir (Table 7). The hardness results of spruce and fir were obtained very close to each other. The shore *D* hardness of poplar wood samples impregnated with phenol formaldehyde resins (64.61) was also generally higher than control samples (42.35) (Li et al., 2018). Devi and Maji (2012) reported that shore *D* hardness was improved in simul wood that was chemically modified by treatment with styrene-acrylonitrile copolymer, glycidyl methacrylate, and organically modified nanoclay. Dong et al. (2016) reported that nano-SiO_2 ratio increased shore *D* hardness for poplar (*Populus* spp.) wood samples from 43.52 to 59.66. As the propolis concentration and the hardness increase, it can form an important feature for furniture and parquet industry areas where hardness is desired. Wood hardness is an important feature, especially in the flooring and furniture industries (Hansson & Antti, 2006).

Conclusion and Recommendations

The efficacy of Turkish propolis against wood-destroying insect *H. bajulus*, color change, shore *D* hardness value, water repellent, and WA of wood have been studied for the first time in this study. The highest WRE was obtained as 61.4% in propolis-impregnated spruce wood at concentration level of 8% during the 2-hour period in the wood species. After 20-hour immersion period, WRE value was 50.4% at 8% concentration level compared to the control samples. These results showed that propolis extracts could be used as hydrophobic material for wood. Larvae MRs indicated that 8% concentration level was not adequate to overcome *H. bajulus* larvae or propolis has no toxicity on *H. bajulus*, although same concentration level inhibited wood-decaying fungi in literature data. For wood types impregnated at 8% concentration, the highest total color difference value was obtained in fir wood, followed by scots pine and spruce wood species. When L^* value decreases with the increase of impregnation rate in all wood types, it was concluded that a^* and b^* values increased. It was observed that the hardness value increased with increase in impregnation rate. As conclusion, the results indicated that propolis could be used as an environmentally compatible, natural water repellent with useful surface properties such as hardness and color properties in wood production industry.

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