Effect of Accelerated Aging on the Color Changes of Wood Treated with Eco-friendly Formulations Based on Propolis and Silicon Compounds

Barbara Lis,[a] Magdalena Woźniak,[b] Tomasz Krystofiak,[a] and Izabela Ratajczak [b,*]

The aim of this work was to determine the effect of propolis extract and propolis-silane formulations on the color of wood. Additionally, the aim was to assess the influence of accelerated aging (water leaching and UV+IR radiation) on the color change of the treated wood. Scots pine wood was treated with the ethanolic extract of propolis and two propolis-silane formulations. The first formulation contained the propolis extract and silanes, 3-(trimethoxysilyl)propyl methacrylate, and tetraethyl orthosilicate. The second formulation comprised propolis extract, vinyltrimethoxysilane, and tetraethyl orthosilicate. The wood impregnation with the propolis extract and propolis-silane formulations had a significant effect ($p \leq 0.05$) on the color change. This effect involved decreasing the *L** parameter and the displacement of *a** and *b** coordinates in the red and yellow direction, respectively. The propolis extract and the propolis-silane formulations were characterized by low durability relative to the effect of light. Nevertheless, the examined formulations based on natural substances (propolis) and low toxicity chemical compounds (silanes) may be used to impregnate wood elements for indoor applications.

Keywords: Propolis; Silicon compounds; Scots pine; Color; Aging

*Author contact information: a: Department of Wood Based Materials, Faculty of Wood Technology, Poznan University of Life Sciences, Wojska Polskiego 38/42, PL-60637 Poznan, Poland; b: Department of Chemistry, Faculty of Wood Technology, Poznan University of Life Sciences, Wojska Polskiego 75, PL-60625 Poznan, Poland; *Corresponding author: izabela.ratajczak@up.poznan.pl*

INTRODUCTION

Wood as a natural and organic material is susceptible to the destructive action of abiotic and biotic factors including microorganisms, fire, water, or ultraviolet radiation (Rowell 2005; Petric 2013). Natural wood resistance against these degradation factors can be increased by impregnating wood with formulations containing biocides, additives, or other chemical substances, which are often toxic and harmful to the environment. Therefore, there are ongoing investigations aiming to develop biocide-free preservatives for wood protection. These eco-friendly formulations consist of both natural substances and synthetic compounds such as essential oils, caffeine, natural oils, chitosan, silicon compounds, or ionic liquids (Pernak *et al.* 2005; Ghosh *et al.* 2009; Hochmańska *et al.* 2014; Pánek *et al.* 2014; Silva-Castro *et al.* 2018; Kwaśniewska-Sip *et al.* 2019). Among natural materials, propolis has promising application in biocidal-free wood preservatives.

Propolis is a bee product consisting of resin, wax, pollen, bee saliva, and other additives (Castaldo and Capasso 2002). The most widely investigated group of chemical compounds present in propolis from different geographical regions are phenolic compounds, which are mainly responsible for its biological activity (Kalogeropoulos *et al.* 2009; Falcão *et al.* 2013; Woźniak *et al.* 2019). Propolis extracts possess antioxidant,

antibacterial, anticancer, and antifungal properties; therefore, propolis is used in numerous applications including wood protection (Kalogeropoulos *et al.* 2009; Salas *et al.* 2016; Ratajczak *et al.* 2018; Woźniak *et al.* 2019). Budija *et al.* (2008) reported that wood treated with the propolis extract exhibited resistance to *Trametes versicolor* and *Antrodia vaillantii*. Moreover, the propolis extract was combined with other bio-friendly substances and was used as wood preservatives. Silva-Castro *et al.* (2018) indicated that coatings based on chitosan and propolis limited the decay of wood caused by *T. versicolor*, while Ratajczak *et al.* (2018) showed that pine wood treated with a formulation based on propolis extract, caffeine, and silicon compounds exhibited resistance against *Coniophora puteana*. In turn, coatings based on silicone matrix and propolis extract with the additive of TiO_2 possessed anticorrosive and antifouling properties (Peres *et al.* 2018). Moreover, the previous research by the authors of this paper indicated that treatment of pine wood with propolis-silane formulations increased its hydrophobic properties (Woźniak *et al.* 2018).

Formulations based on propolis extracts and silicon compounds may be used for bio-friendly wood protection. The application of finishing in the form of coatings or impregnation formulations may cause color changes in the wood (Vitosyte *et al.* 2018). Therefore, the aim of this paper was to determine the effect of the propolis extract and propolis-silane formulations on the color of wood. Additionally, this paper examined the influence of accelerated aging (water leaching and UV+IR radiation) on the color changes of treated wood.

EXPERIMENTAL

Preparation of Wood Samples

The investigated material was Scots pine (*Pinus sylvestris* L.) sapwood with dimensions of 40 x 40 x 5 mm (± 1 mm) (longitudinal by tangential by radial) without knots and other growth inhomogeneities. The average density of the wood samples was 550 ± 55 kg per m^3 and had a moisture content of 12 ± 1 %.

The wood samples (20 blocks of wood per treatment) were impregnated with the ethanolic extract of propolis (EEP) at a 15% concentration (PROP-MAD, Poznań, Poland) and two propolis-silane formulations. The first formulation (EEP-MPTMOS/TEOS) contained EEP and silanes: 3-(trimethoxysilyl)propyl methacrylate (MPTMOS) and tetraethyl orthosilicate (TEOS) at a 5% concentration. The second formulation (EEP-VTMOS/TEOS) consisted of EEP, vinyltrimethoxysilane (VTMOS), and tetraethyl orthosilicate (TEOS) at a 5% concentration. The silicon compounds were purchased from Merck (Darmstadt, Germany). The wood samples were impregnated using the vacuum method. Samples underwent 15 min of vacuum conditions at 0.8 kPa and 2 h of atmospheric pressure.

The average retention of EEP for wood samples used in this research was 102 kg per m^3. In turn, the wood impregnated with the formulation EEP-MPTMOS/TEOS had a retention of 164 kg per m^3. For wood treated with EEP-VTMOS/TEOS, the average retention was 165 kg per m^3.

Accelerated Aging Procedure

Ten samples of the treated wood were subjected to the leaching procedure. The wood samples were impregnated with deionized water under vacuum. The samples were then soaked, where water was exchanged 10 times for the duration of 2 weeks. The samples of the impregnated wood and wood after the leaching procedure were subjected to an ultraviolet light combined with infrared radiation (UV+IR) aging test. This test was performed using an ultraviolet and infrared radiation quartz lamp (VT 800, FAMED Łódź S.A., Łódź, Poland) with a radiation energy of 740 W. During the measurement, the wood samples were arranged at an angle of 45° and 40 cm from the lamp. The wood samples were irradiated for 1 h at intervals of 15 min.

Color Measurement

The investigation of wood color change was carried out with a Testan DT-145 colorimeter (Anticorr, Gdańsk, Poland) using the CIE $L^*a^*b^*$ measurement system. The test started with a calibration using a white standard. The measuring points were determined on the surface of the samples in order to eliminate the influence of wood drawing on the obtained results. This was also performed to determine any color changes after aging. In these areas, the coordinates of pre- and post-exposure color were determined.

The numerical values of the color changes were calculated by a specialist program included in the equipment of the instrument. The ΔE^* parameter according to the formula was calculated using Eq. 1,

$$\Delta E^* = \sqrt{\left(\Delta L^*\right)^2 + \left(\Delta a^*\right)^2 + \left(\Delta b^*\right)^2} \quad \text{.....(1)}$$

where ΔE^* is the color difference and L^* is the achromatic coordinate of the color (brightness). A value of L^* equal to 100 means that the color is a perfect white and L^* equal to 0 means that the color is a perfect black. The parameters a^* and b^* are the chromatic coordinates of the colors ($+a^*$ indicates red, $-a^*$ indicates green, $+b^*$ indicates yellow, and $-b^*$ indicates blue). For the interpretation of the color difference of the samples (ΔE^*), the ranges given in Table 1 were used.

Table 1. Range of the ΔE^* Indicator

Variability range	Color difference
$0 < \Delta E^* < 1$	Not a perceptible difference
$1 < \Delta E^* \leq 2$	Very small, recognizable only by an experienced observer
$2 < \Delta E^* \leq 3.5$	Average, also recognizable by an inexperienced observer
$3.5 < \Delta E^* \leq 5$	Clearly
$\Delta E^* > 5$	Distinctly

Statistical Analysis

The statistical analysis of date obtained in this paper was performed using STATISTICA 10 (StatSoft Polska, Kraków, Poland).

RESULTS AND DISCUSSION

The results of the color measurements are presented in Table 2. Impregnation of wood with the propolis extract and propolis-silane formulations caused a change in the individual coordinates of the color, leading to a clear difference in its reception. In relation to untreated samples, a remarkable decrease in the *L** parameter was observed, indicating their darkening and displacement of the *a** and *b** coordinates in the direction of red and yellow, respectively. These changes may be related to the chemical composition and color of the products used for wood protection and/or chemical modification of wood constituents as a result of impregnation (Turkoglu *et al.* 2015). The obtained differences in the value of the *L** index in relation to the unprotected wood amounted for particular formulations, respectively: EEP (23.9), EEP-MPTMOS/TEOS (16.9), and EEP-VTMOS/TEOS (16.9). This data indicated a high susceptibility to the darkening of the propolis extract and propolis-silane formulations. The dominant color in the tested systems was yellow (*b**), ranging from 32.2 to 37.8. Considering the *a** parameter, the positive values from the red color range were noted. For all the tested samples, they were twice as high as the initial samples and oscillated at the level of about 15 units. When assessing the development of color attributes for the impregnated samples exposed to UV+IR radiation, it was found that the changes in color coordinates in relation to the wood surface before aging deepened.

Table 2. Effect of Radiation Time on the Color Coordinates of the Wood Treated with the Propolis Extract and Propolis-silane Formulations

Color coordinates	Exposure time [min]	Wood Samples						
		Untreated samples	EEP		EEP-VTMOS/TEOS		EEP-MPTMOS/TEOS	
			UL	L	UL	L	UL	L
*L**	Control	85.458	64.753	65.187	68.600	67.599	68.549	66.719
	15	84.262	62.269	63.115	66.619	65.955	66.531	64.597
	30	83.520	61.835	62.682	66.260	65.758	66.253	64.175
	45	83.303	61.764	62.437	65.941	65.539	65.796	63.868
	60	82.840	61.520	62.291	66.024	65.319	65.551	63.554
*a**	Control	7.403	15.587	16.649	14.002	14.000	14.261	15.554
	15	7.323	16.477	17.691	14.743	14.983	15.231	16.517
	30	7.352	16.677	17.913	14.856	15.140	15.494	16.810
	45	7.343	16.710	17.964	14.882	15.128	15.649	16.883
	60	7.322	16.761	18.004	14.922	15.204	15.692	16.857
*b**	Control	21.601	33.433	37.839	32.603	35.065	35.158	35.664
	15	21.599	30.747	35.468	30.855	33.497	32.735	33.676
	30	21.865	30.446	35.245	30.572	33.313	32.581	33.381
	45	22.155	30.292	34.903	30.910	33.136	32.423	33.199
	60	22.593	30.190	34.825	30.508	33.097	32.218	33.228

*Note: UL represents the unleached wood samples and L represents the wood samples leached with water.

As a function of exposure time, a systematic tendency to deepen the degree of darkening, increase the chromatic coordinate *a**, and decrease the *b** parameter was observed. This is most likely a consequence of photolysis in the course of reactions and free radical processes. These processes lead to chemical changes in the wood raw material caused mainly by the regrouping of the aromatic structure of lignin (Donath *et al.* 2007; Lesar *et al.* 2011; Genco *et al.* 2011; Zheng *et al.* 2015; Oberhofnerová *et al.* 2017).

The effect of the exposure time on the Δ*L** value of wood treated with the propolis extract and the propolis-silane formulations is presented in Fig. 1.

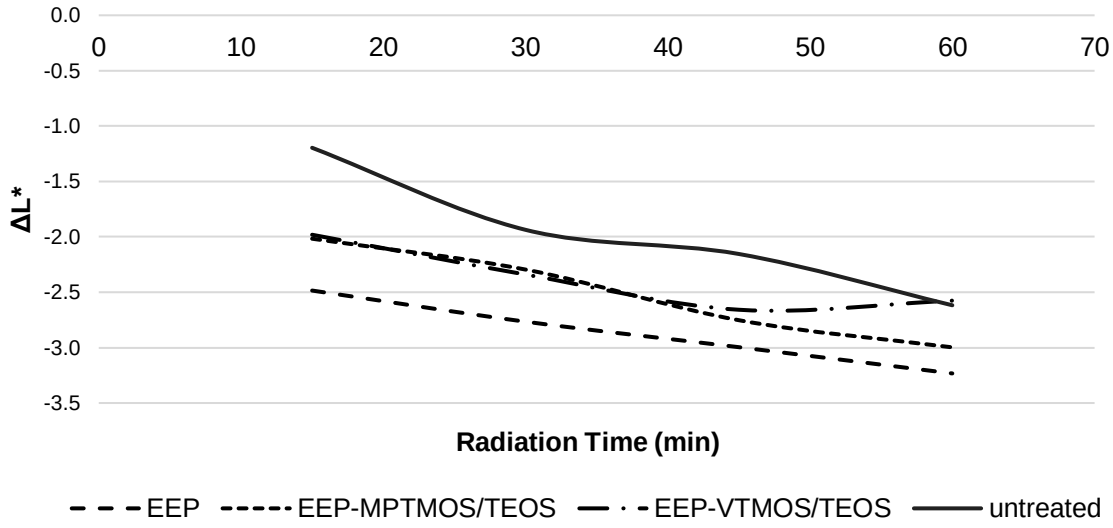

Fig. 1. The course of the Δ*L** coordinate in function of the radiation time

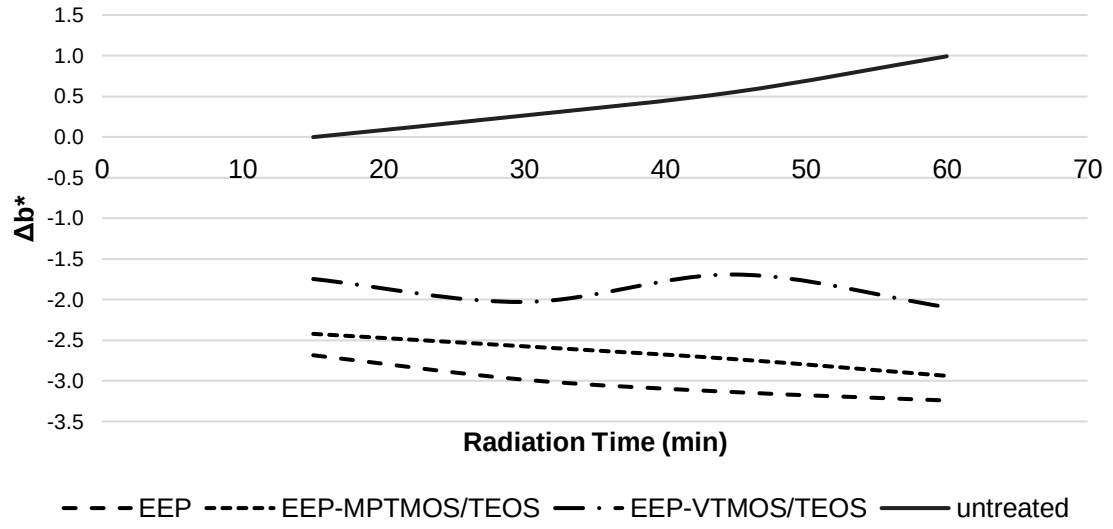

Fig. 2. The course of the Δ*b** coordinate as a function of the radiation time

The smallest decrease in brightness in relation to untreated wood after 1 h of exposure to radiation (amounted to 2.58) was recorded for the wood treated with the formulation EEP-VTMOS/TEOS. In the case of this impregnating system, however, an ambiguous shaping of this parameter was observed. In the last phase of radiation adopted,

an upward trend was observed and the specimen finally reached a level like that of untreated wood. The change of ΔL^* values for EEP and EEP-MPTMOS/TEOS was equal to 3.23 and 3.00, respectively. The Δb^* coordinate (Fig. 2) for treated wood was reduced when compared to untreated samples. This indicated a slight reduction of the yellow attribute and a gradual tendency to increase the blue shades on the surface. Short UV+IR aging within the range of 1 h causes that the coatings become "more blue" with exposure. For the chromatic coordinate value Δa^* (Fig. 3), a slight increase in the redness effect was observed during the increase in exposure time.

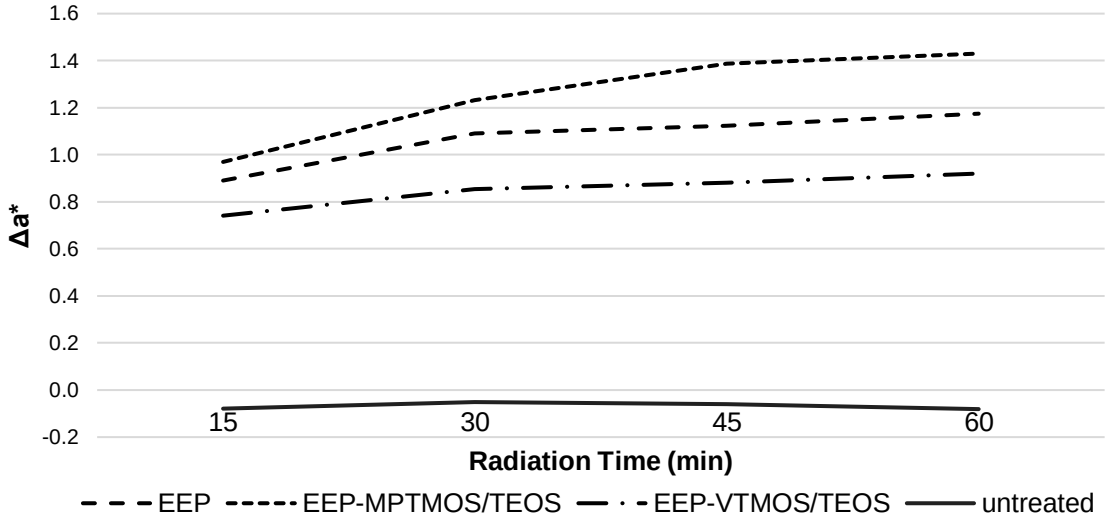

Fig. 3. The course of the Δa^* coordinate as a function of the radiation time

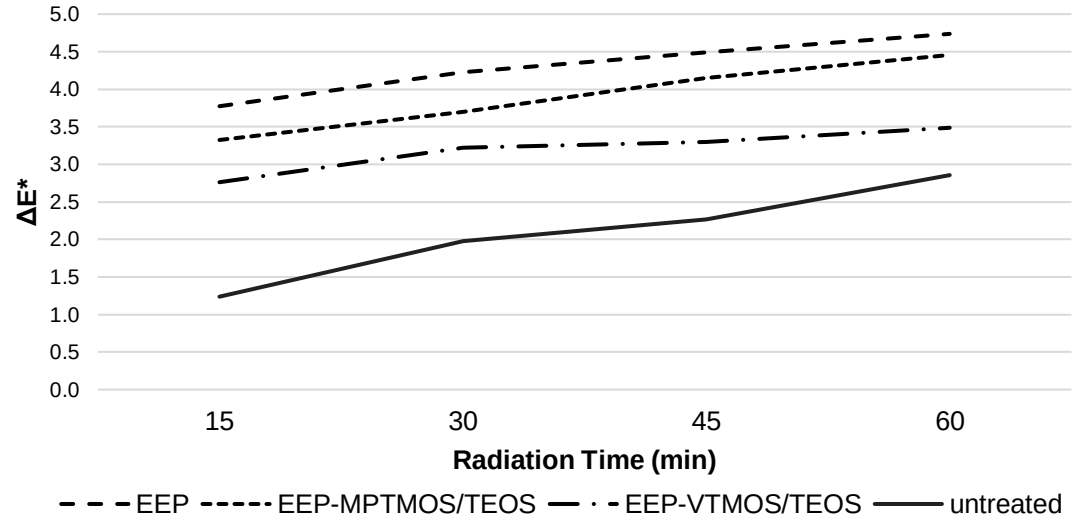

Fig. 4. The course of the ΔE^* factor as a function of the exposure radiation time

Similar results were obtained by Lesar *et al.* (2011), who conducted research on the influence of waxes on spruce wood protection. The research went further to focus on the aspect of surface photodegradation under accelerated aging conditions (Lesar *et al.* 2011). The relationship concerning the Δb^* parameter for untreated wood was different in comparison to wood impregnated with EEP and the propolis-silane formulations. This

parameter for unprotected samples increased, causing stronger yellowing and darkening accompanied by a weaker reddening effect in the control wood. Figure 4 shows the values of the ΔE^* parameter as a function of the exposure time, which expresses the total change in color of the tested formulations between the initial and given aging stage.

This indicator allows for an objective evaluation of color changes occurring on the surface of the studied materials under the influence of aging (Pandey 2005). The general evaluation of the obtained data carried out in relation to the criteria presented in Table 2 demonstrates that there is an unfavorable tendency of changes in this parameter. In the time intervals of exposure to UV radiation in the samples, a noticeable increase of this attribute was observed after 15 min. The resulting color differences were recognizable by an inexperienced observer. The recorded values were only at the level in the unprotected wood surface. The recorded values in this case were only differentiated by an experienced person according to the range presented in Table 1.

The fast change of color is undoubtedly caused by the absorption of UV radiation by the wood, which can reach a depth of up to approximately 75 µm. This causes the initiation of photochemical reactions in wood constituents, mainly in lignin. The result is the formation of radicals, which react with oxygen and transform into chromophoric carbonyl and carboxyl groups responsible for photodegradation. The resulting discoloration is the first sign of chemical changes in wood during aging. Their intensity depends on wavelength, radiation time, temperature, type of light source, and wood species. It should also be noted that UV radiation in combination with IR can lead to a synergistic effect. This effect contributes to the intensification of processes occurring on the surface, which catalyzes the color change (Tolvaj and Faix 1995; Ayadi *et al.* 2003; Jirous-Rajkovic *et al.* 2004; Pandey 2005; Kataoka *et al.* 2007; Nowaczyk-Organista 2008; Deka *et al.* 2008; Oberhofnerová *et al.* 2017; Cogulet *et al.* 2018).

In the next stages of the aging process of all tested samples, the level of observed values ΔE^* increased. However, the rate of color changes was already lower. The lowest dynamics of the color changes was observed in the case of untreated wood. After the end of the exposure cycle (1 h), the value of 2.86 was noted. The protected systems with light fastness was lower and in the range of 3.49 to 4.73. Among the tested formulations, the smallest changes after irradiation were observed for wood samples treated with EEP-VTMOS/TEOS. A 21% decrease in resistance was observed in comparison to untreated wood, while for the remaining variants it was higher. The remaining variance amounted to 56% for EEP-MPTMOS/TEOS and 66% for EEP. Literature data suggests that the obtained results may be influenced by the hydrophobic character of the wood surface. Hochmańska *et al.* (2014) conducted research on the resistance to aging factors of impregnation and protection systems based on alkyd resin with silicon compounds. Their research concluded that the decrease in hydrophilic properties of treated wood results in an increase in the surface resistance to photodegradation (Hochmańska *et al.* 2014). The above observations also confirm the results obtained by the authors in the wettability studies of the preparations in question (Woźniak *et al.* 2018). The highest contact angle value was recorded for the wood surface treated with EEP-VTMOS/TEOS, which also showed the lowest values of ΔE^*. However, this does not explain the high color stability for untreated wood, where a high affinity to water was observed. Therefore, the authors think that this may be related to the initial color of the wood, which may affect the rate of discoloration. According to literature data, light colored surfaces are assumed to be less susceptible to changes than dark ones. The photooxidation process is more intensive, therefore reducing their durability over time (Forsthuber and Grüll 2018). According to Genco *et al.* (2011), it can be assumed

that both the wood and the preservative can change color. It should be noted that the color of propolis varies (from yellow, red, green, light, to dark brown) depending on the source and the age of the plant from which it was harvested (Castaldo and Capasso 2002; Falcão *et al.* 2013). This makes its chemical composition highly variable. So far, more than 300 substances have been identified in propolis samples from different geographical regions (Castaldo and Capasso 2002; Kalogeropoulos *et al.* 2009; Wagh 2013; Berretta *et al.* 2017). Among them, polyphenols could be responsible for the color, and they undergo degradation over time. Their stability decreases with an increasing temperature, which would explain the greater color change as a result of the interaction of UV+IR radiation in relation to the untreated wood.

When considering the color stability of the tested formulations after the leaching process, similar trends like the treated wood before the leaching procedure were observed. However, compared to the samples before leaching the values of the indicators in question were higher by approximately 2 to 3 units. As the aging process progressed, a trend towards less brightness was observed. All the variants of formulations were characterized by a more yellow color, whose share in the function of exposure time decreased. Changes were also observed in the red color for which an increase in intensity was registered. It is most likely that a lower variability in color results from a decrease in the amount of extraction compounds (Genco *et al.* 2011).

The results showed that the impregnation of wood with the developed formulations caused a change in the surface color of pine wood, which darkened during the exposure to the UV+IR radiation. The application of the propolis extract and propolis-silane formulations did not protect the wood against a color change. The ΔE^* value for treated wood was higher as a function of the exposure time than for an unprotected surface. The obtained results to some extent correspond to the results of Jankowska and Szczęsna's (2011) experiments. It was demonstrated that accelerated aging considered on the selected wood species from South-East Asia (teak, merbau, campus) with the products applied (coating, wax, polishes), leads to a change in the color of wood. The size of which depends on both the species and the using product (Jankowska and Szczesna 2011). The use of preparations on the surface of wood does not contribute to the improvement of lightfastness of the substrate, but only to the equalization of its color. Similar observations were confirmed by Sógutlu and Sónmez (2006), who evaluated the effect of UV radiation on the color change of various wood species enriched with teak oil, liquid paraffin, and shellac. The authors stated that the obtained finishing's did not protect the wood against a color change (Sógutlu and Sónmez 2006).

CONCLUSIONS

1. Wood impregnation with the propolis extract and propolis-silane formulations had a significant effect ($p \leq 0.05$) on the color change. The change in coloration involved a lowering of the L^* parameter and shifting the a^* and b^* coordinates in the red and yellow direction, respectively.
2. The propolis extract and formulations consisting of propolis extract and silanes did not influence the improvement of wood surface durability on the effect of light.
3. The tested formulations for wood impregnation in terms of lightfastness can be ranked as follows (highest to lowest): EEP-VTMOS/TEOS, EEP-MPTMOS/TEOS, and EEP.

4. The EEP and propolis-silane formulations may be recommended for indoor use in situations where there is no direct exposure to radiation.
5. The tested, bio-friendly formulations based on natural (propolis) and low toxicity (silanes) constituents may be used to impregnate wood elements for indoor application (*e.g.* archaeological wooden monuments in museums).

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