

Investigating the surface performance of impregnated and varnished Calabrian pine wood against weathering

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Abstract

The study investigates the gloss and color changes values of Calabrian pine (*Pinus brutia* Ten.) wood impregnated with some copper content impregnation chemicals such as Celcure C4, Korasit KS, and Tanalith E 8000 and then water-based varnish (WBV) and polyurethane varnish (PV) coated after 6 months of weathering. The results of study showed that gloss values of PV coated Calabrian pine wood were higher than that of WBV coated Calabrian pine wood before weathering. The gloss values of all treatment groups decreased after weathering. Lightness values of Calabrian pine wood also decreased for all treatment groups after weathering. Pre-impregnation before PV coating resulted in lower ΔL^* values of Calabrian pine wood. While all treatment groups tended to turn reddish by giving Δa^* positive values, they tended to turn bluish by giving negative Δb^* values. The lowest total color change was obtained with only WBV coated Calabrian pine wood.

Keywords: *impregnation, weathering, varnish, color and gloss test, Calabrian pine wood.*

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1. Introduction

Wood is an environmentally friendly and sustainable natural material used for a wide variety of both structural and non-structural applications^[1,2], especially in building and construction applications^[3]. In addition to many positive properties of wood material, there are some undesirable negative properties. The wood material can be burned, absorb water, exposed to fungal and insect attack, unprotected against outside weathering factors. Environmental conditions such as atmospheric pollutants, oxygen, moisture, sunlight, cold, heat, chemicals, and wind erosion can cause an economic depreciation by reduced service life of wood. Therefore, it is necessary to protect the wood material in certain ways in order to protect it against such weather conditions. The coating is applied to protect wooden surfaces from environmental influences, biological degradation, and corrosion^[4]. Coating adds an aesthetic appearance to wooden surfaces, making it desirable for the users^[5]. However, coating thickness becomes thinner over time. The tissues on the surface undergo deformation^[6]. Wood should be impregnated with suitable material and varnish applied before it is used in structural or furniture

production. After these processes, the wood used outdoors is much more durable in terms of biological decomposition, dimensional changes and photochemical degradation^[7]. Thus, modification techniques with impregnating agents can increase the resistance of wood to weather conditions^[8]. One of the methods applied for the protection of outdoor degradation of wood material is impregnation with water-soluble salts such as chromium and copper. The use of various impregnating agents has become important in place of chromated copper arsenate (CCA) impregnation, which is a water-soluble impregnation agent and has been used in large quantities throughout the world. Most of them do not contain arsenic and can be listed as follows: Acid copper chromium (ACC); alkali copper quat (ACQ); copper azole (CA); copper citrate (CC); copper diethyldithiocarbamate (CDDC); copper HDO^[9]. Since the carcinogenic structure of chromium compounds is well known, the new impregnation agent is generally seen as copper in alternative substances^[10]. Zhang et al.^[11] in their study, found that by impregnating with copper ethanalamine after the accelerated weather conditions test, they greatly prevented the change in the

surface properties of the wood materials. Altay et al.^[12] investigated color changes of Scots pine wood impregnated with copper content chemical before synthetic, cellulosic, polyurethane varnishes (PV) coating after weathering. They found that a good color stability in the Scots pine specimens treated with Wolmanit CX-8 (WCX-8) before PV coated of weathering conditions.

This study was presented at Calabrian pine (*Pinus brutia* Ten.) wood impregnated with a 2.5 and 5 percent aqueous solutions of copper-based chemicals such as Celcure C4, Korasit KS, and Tanalith E 8000. After impregnation, water-based varnish (WBV) and PV were coated to the wood surface. After the preparation process the wood specimens were exposed to the weather condition for 6 months in Mugla Province of Turkey. Therefore, this study aims at investigating the effect of weathering on gloss and color changes of impregnated Calabrian pine wood with copper content impregnation chemicals before water-based and polyurethane varnishes coatings.

2. Materials and Methods

2.1 Preparation of wood specimens

The Calabrian pine (*Pinus brutia* Ten.) wood specimens were used with a dimension of 10 mm x 100 mm x 150 mm (radial x tangent x longitudinal). The test samples were conditioned for two weeks at 20 °C and 65% relative humidity before testing.

2.2 Impregnation process

The Calabrian pine wood specimens were impregnated with 2.5% and 5% aqueous solutions of impregnation chemicals according to ASTM D1413-07e1^[13]. The retention values of the test samples at the end of the impregnation were calculated by the following formula 1:

$$Retention = \frac{G.C}{V} \times 10^3 \left(Kg / m^3 \right) \quad (1)$$

$$G = T_2 - T_1$$

T₁: Weight of specimen before impregnation (g)

T₂: Weight of specimen after impregnation (g)

V: Volume of specimen volume (cm³)

C: Concentration (%)

2.3 Application of varnish

The Calabrian pine wood specimens were varnished with polyurethane and water-based varnishes after the impregnation process in this study. Polyurethane varnish application; all surfaces and edges of specimens were applied polyurethane varnish with a spray gun according to ASTM D3023-98 standard^[14]. Before the application of polyurethane varnish, the surfaces were cleaned from dust by the suitable sanding process. After the polyurethane varnish was mixed thoroughly, it was thinned by the addition of thinner, and two cross-layers were applied on normal porous surfaces. Then, the wood specimens were sanded with sand number 220 and the topcoat application started. In water-based varnish application, after sanding

the wooden surfaces with 220 grit sandpaper, the varnish was taken to an empty container and coated to all surfaces and edges of the wood with a brush. During the application, the undiluted varnish was applied in 2 layers. Polyurethane and water-based varnish application values are as follows:

Polyurethane varnish: Filling: 100 g/m² – Topcoat: 100 g/m²

Water-based varnish: Filling: 100 g/m² – Topcoat: 100 g/m²

2.4 Gloss test

The gloss values Calabrian pine wood specimens were determined using a gloss meter (BYK Gardner, MicroTRI-Gloss) according to ASTM D523-14^[15]. The measurement geometry was chosen a 60° incidence angle. Ten replicates were made for each treatment group. Gloss measurements were made parallel to the fibers.

2.5 Color test

The color parameters *a**, *b** and *L** were determined by the CIEL*a*b* method (Figure 1).

The *L** axis shows brightness, while *a** and *b** are color coordinates. The parameters *+a** and *-a** show red and green, respectively. The parameter *+b** shows yellow, while *-b** shows blue. *L** can range from 100 (white) to 0 (black). The total color changes (ΔE^*) for Calabrian pine wood specimens were determined by ASTM D 1536–58 T^[16]. Equations 2-5:

$$\Delta a^* = af^* - ai^* \quad (2)$$

$$\Delta b^* = bf^* - bi^* \quad (3)$$

$$\Delta L^* = Lf^* - Li^* \quad (4)$$

$$(\Delta E^*) = ((\Delta a^*)^2 + (\Delta b^*)^2 + (\Delta L^*)^2) \quad (5)$$

Where: Δa^* , Δb^* , and ΔL^* are the changes between the first and last range values. Ten replicates were made for each treatment group. Color measurements were made parallel to the fibers.

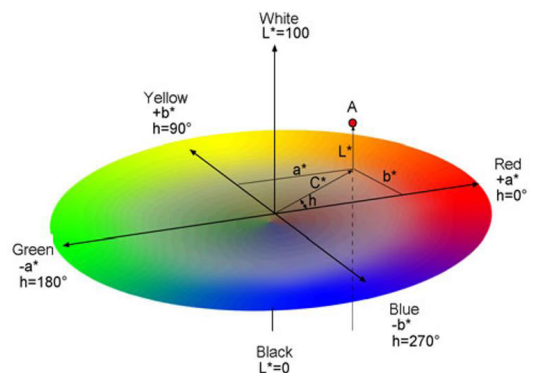


Figure 1. The CIEL*a*b* colour space^[6].

2.6 Weathering test

The Calabrian pine wood was exposed to weathering for 6 months from May to November in 2019. The wood panels were prepared for weathering according to ASTM D 358-55^[17]. A test site has been established for practical evaluations near Muğla Regional Meteorological Observation Station in the South Aegean Region. The meteorological data of Muğla is provided in Table 1.

2.7 Statistical evaluations

IBM SPSS® program was used to evaluate the test results. Variance Analysis and Duncan tests were performed on the results. The Variance Analysis and Duncan test applied at 95% confidence level in these test result. Statistical evaluations were made on homogeneity groups (HG) where different letters reflect statistical significance according to test results. Ten replicates were made for each treatment group and total 140 data were analysed.

3. Results and Discussions

3.1 Gloss test results

The gloss and gloss losses values of impregnated and varnished Calabrian pine wood before and after weathering are demonstrated in Table 2. A clear coating application is the easiest and most common method to protect wood against weather conditions and increase its distinctive appearance.

Varnished wood samples reflect the natural appearance of wood surfaces^[18].

In this study, while the highest gloss value (99.52) was obtained with PV coated Calabrian pine wood, the lowest gloss value (44.56) was obtained with 5% Korasit KS+WBV treated Calabrian pine wood before weathering. Baysal et al.^[18] found that the glossiness of Scots pine impregnated with some copper content chemical before polyurethane varnish coating ranged from 96 to 103 depending on the treatment. Another study, Toker et al.^[19] reported that the gloss values of Calabrian pine pre-treated with borates before polyurethane varnish coating ranged from 89.1 to 97. Our results were similar to Baysal et al.^[18] and Toker et al.^[19]. Except for the Korasit KS+WBV and Tanalith E 8000 + WBV treatment groups at all concentrations, the glosses of the Calabrian pine wood increased with rising concentration levels of chemicals. Pre-impregnation with chemicals before the PV coating decreased the gloss values of Calabrian pine wood. The chemicals can limit gloss to a certain degree in test samples before weathering. The reason for the experimental results reached may be due to the dispersion and absorption of rays reflected from salt crystals. There are photoactive ions on the wood surface. Varnished wood due to photoactive ions may loss of gloss before decomposition^[7]. These results are similar to the gloss values of Scots pine and Oriental beech wood impregnated before varnishing^[20,21]. Gündüz et al.^[22] investigated gloss values of Scots pine pre-impregnated with some copper-based chemicals such as Wolmanit CX-8 and Celcure AC 500 before water-based varnish coating. They found that pre-impregnation with chemicals before

Table 1. Meteorological data of Muğla.

Months	May	June	July	August	September	October
Average temperature per month (°C)	18.4	23.4	26.1	27.8	22.6	18.0
Humidity per month (%)	54.4	55.4	42.7	40.0	53.3	65.1
Average wind speed per month (m/sn)	1.3	1.4	1.8	1.7	1.4	0.9
Total rainfall per month (mm=kg/m ²)	9.0	76.5	43.2	16.4	51.4	78.6
Number of rainy days	5	12	1	1	5	5

Table 2. Gloss change values of Calabrian pine wood specimens before and after natural weathering.

Impregnation + Varnish	Concentration (%)	Retention (Kg/m ³)	Before natural weathering		After 6 months of natural weathering		Gloss losses (%)	
			gloss values		gloss values		Mean	HG
			Mean	SD	Mean	SD		
PV	-	-	99.52	9.72	55.32	5.95	-44.41	C
WBV	-	-	46.26	6.50	24.15	3.25	-47.79	DE
Celcure C4+PV	2.5	13.48	97.60	11.19	57.24	6.10	-41.35	B
Celcure C4+PV	5.0	30.92	98.36	10.06	43.30	9.58	-55.97	G
Korasit KS+ PV	2.5	14.88	97.93	9.76	56.25	7.03	-42.56	B
Korasit KS+ PV	5.0	32.11	98.66	8.87	50.90	8.55	-48.40	E
Tanalith E 8000 +PV	2.5	15.05	93.12	12.49	54.57	6.45	-41.39	B
Tanalith E 8000 +PV	5.0	31.47	96.36	13.11	51.66	7.48	-46.38	D
Celcure C4+ WBV	2.5	13.48	50.43	6.89	19.79	5.69	-60.75	H
Celcure C4+ WBV	5.0	30.92	54.68	8.05	33.94	5.04	-37.92	A
Korasit KS+ WBV	2.5	14.88	52.90	12.61	24.62	5.84	-53.45	F
Korasit KS+ WBV	5.0	32.11	44.56	6.39	22.90	5.69	-48.60	E
Tanalith E 8000 + WBV	2.5	15.05	52.62	7.90	22.46	4.26	-57.31	G
Tanalith E 8000 + WBV	5.0	31.47	52.62	5.72	32.15	5.89	-38.91	A

Note: Results reflect the average of 10 Calabrian pine wood specimens. PV: Polyurethane varnish; WBV: Water-based varnish; SD: Standard deviation; HG: Homogeneity groups; A-G letters: Statistical differences.

WBV coating caused increase in the gloss of Scots pine. In addition, Gündüz et al.^[23] studied gloss values of Scots pine pre-impregnated with some copper-containing chemicals before polyurethane varnish (PV) coating. They reported that pre-impregnation with chemicals before PV coating caused decrease in the gloss of Scots pine. In our study, after the weathering process, the glosses of all Calabrian pine wood decreased. Because erosion and abrasion on the wood surfaces cause gloss loss in the varnish layers after weathering^[7,24]. Gloss losses values after 6 months of weathering range from -37.92% to -60.75%. Türkoglu et al.^[20] found that gloss loss of some copper-based chemicals treated and polyurethane varnish coated Scots pine ranged from 59.91% to 69.90% after 6 months of weathering. Surprisingly, our results showed that lower concentration levels (2.5%) of chemicals for PV coated Calabrian pine wood caused decrease in gloss losses of Calabrian pine wood samples after weathering. For WBV coated Calabrian pine wood, higher concentration levels of chemicals resulted in lower gloss losses of Calabrian pine wood after weathering.

3.2 Color test results

The color parameters, color changes, and total color change values of the Calabrian pine wood impregnated with copper content chemicals and varnished before and after weathering are given in Table 3. According to Table 3, while the highest L^* value (70.89) was obtained with WBV coated Calabrian pine wood, the lowest L^* value (43.13) was measured with 5% Tanalith E 8000+PV treated Calabrian pine wood before weathering. Our results showed that pre-impregnation with copper-based chemicals before varnishing caused decrease L^* values of Calabrian pine wood to some extent.

Baysal^[25] investigated the color characteristics of Scots pine wood impregnated with some copper content chemicals before polyurethane varnish coating after accelerated weathering. They reported that pre-impregnation with chemicals before varnish coating reduced L^* values of Scots pine wood before accelerated weathering. The results of our study are in good agreement with data Baysal^[25]. Gündüz et al.^[22] studied color characteristics of Wolmanit CX-8, Adolit KD-5 and Celcure AC 500 impregnated Scots pine wood before water-based varnish (WBV) coating after accelerated weathering. They reported that L^* values of Scots pine wood decreased impregnated with chemicals before WBV coating before weathering. The results are in good agreement with Gündüz et al.^[22]. Concentration levels of chemicals had no significant effect on the L^* values of Calabrian pine wood before weathering. While a^* values range from 0.87 to 6.32, b^* values vary between 30.34 to 42.29 before weathering. After 6 months of weathering, the ΔL^* values of Calabrian pine wood decreased for all treatment groups. Since wood surfaces are delicate to UV light, the ΔL^* values were negative in all treatments. Chemical changes ensue in wood constituent such as lignin on surfaces of wood during photo degradation^[26]. Depolymerisation of lignin may cause the darkening of the wood surface^[18,27,28]. Pre-impregnation with chemicals before PV coating gave lower ΔL^* values than only PV coated Calabrian pine wood. Because ΔL^* value was -7.75 for only PV coated Calabrian pine wood, it changed from -0.29 to -6.65 for impregnated and PV coated Calabrian pine wood. ΔL^* values of Calabrian pine wood increased with increasing concentration levels of chemicals. All treatment groups tended to turn reddish and bluish, giving a positive Δa^* and negative Δb^* values, respectively after natural weathering. Ghosh et al.^[28] studied color characteristics of a copper-containing chemical such

Table 3. Color change values of the Calabrian pine wood specimens before and after natural weathering.

Impregnation + Varnish	Conc. (%)	Color values before natural weathering			Color change values after natural weathering			ΔL^*	Δa^*	Δb^*	Total color change values after natural weathering				
		L_i^*	a_i^*	b_i^*	L_f^*	a_f^*	b_f^*				ΔE^*				
											Mean	HG	Mean	HG	
PV	-	67.92	6.32	41.51	60.17	14.18	37.10	-7.75	G	7.86	ABC	-4.41	A	11.88	DE
WBV	-	70.89	4.79	33.03	66.00	11.01	30.84	-4.89	DE	6.22	DE	-2.19	A	8.20	G
Celcure C4+PV	2.5	49.78	4.15	42.29	44.36	12.15	23.51	-5.42	E	8.00	ABC	-18.78	EF	21.12	AB
Celcure C4+PV	5.0	44.47	1.24	30.34	40.79	8.45	25.83	-3.68	C	7.21	BCD	-4.51	A	9.26	G
Korasit KS+ PV	2.5	50.36	3.82	40.50	43.71	12.32	23.17	-6.65	F	8.50	AB	-17.33	E	20.41	B
Korasit KS+ PV	5.0	46.90	2.09	33.53	43.36	8.55	23.59	-3.54	C	6.46	DE	-9.94	C	12.37	DE
Tanalith E 8000 +PV	2.5	50.46	3.04	39.34	50.17	11.95	29.22	-0.29	A	8.91	A	-10.12	C	13.48	D
Tanalith E 8000 +PV	5.0	43.13	3.07	37.93	42.67	9.49	16.51	-0.46	A	6.42	DE	-21.42	G	22.36	A
Celcure C4+ WBV	2.5	49.57	2.07	33.74	49.32	10.24	26.27	-0.25	A	8.17	ABC	-7.47	B	11.07	EF
Celcure C4+ WBV	5.0	44.01	2.00	31.34	39.64	9.04	17.17	-4.37	CD	7.04	CD	-14.17	D	16.41	C
Korasit KS+ WBV	2.5	56.12	3.59	39.60	46.75	9.46	19.80	-9.37	H	5.87	DE	-19.80	FG	22.67	A
Korasit KS+ WBV	5.0	49.99	2.18	33.07	48.40	9.44	24.41	-1.59	B	7.26	BCD	-8.66	BC	11.41	EF
Tanalith E 8000+ WBV	2.5	51.01	3.30	38.80	44.21	8.86	17.86	-6.80	F	5.56	E	-20.94	FG	22.70	A
Tanalith E 8000+ WBV	5.0	52.35	0.87	30.56	48.53	8.97	26.63	-3.82	C	8.10	ABC	-3.93	A	9.77	FG

Note: Results reflect the average of 10 Calabrian pine wood specimens. PV: Polyurethane varnish; WBV: Water-based varnish; Conc: Concentration (%); HG: Homogeneity groups; A-G letters: Statistical differences

as Wolmanit CX-8 impregnated and PV coated Oriental beech after 500 h of Scots pine specimens after accelerated weathering. They revealed that Scots pine specimens showed reddish, giving a positive Δa^* values and gave negative Δb^* values tended to turn bluish. Türkoğlu et al.^[20] studied color changes of a copper-based chemical such as Adolit KD 5 impregnated and PV coated Oriental beech wood samples. They explained that Oriental beech wood gave negative Δa^* and Δb^* values, respectively after 6 months of weathering. In this study, while the highest total color change (ΔE^*) was obtained in 2.5% Tanalith E 8000+WBV treated Calabrian pine wood, the lowest total color change was determined in the only WBV coated wood specimens. The total color changes of Calabrian pine wood vary between 8.20 to 22.70 after weathering. In this study, as the concentration levels of chemicals increased, total color change of Calabrian pine wood decreased. In other words, the increase in concentration levels of chemicals has produced positive results in terms of total color changes of Calabrian pine wood. Yalinkilic et al.^[7] investigated the weathering performance of Scots pine and chestnut wood impregnated with chromium-copper-boron (CCB) before polyurethane varnish coating. They reported that CCB impregnation reduced total color changes of Scots pine and chestnut wood. Türkoğlu et al.^[20] reported that impregnation of wood some copper content chemicals before polyurethane varnish coating reduced total color changes of wood after weathering. Gündüz et al.^[22] found that pre-impregnation with copper-based chemicals such as Adolit KD 5, Wolmanit CX-8, and CAC 500 before water-based varnish coating reduced total color changes of Scots pine after 1000 h of accelerated weathering. Our results are similar to Yalinkilic et al.^[7], Türkoğlu et al.^[20], and Gündüz et al.^[22].

4. Conclusions

The gloss losses were observed for all treatment groups after weathering. The gloss values of PV coated Calabrian pine wood were higher than that of SBW coated Calabrian pine wood before weathering. While higher concentration levels for WBV coated Calabrian pine wood resulted in lower gloss losses, higher concentration levels of PV coated Calabrian pine wood resulted in higher gloss losses after weathering. ΔL^* values of impregnated and coated Calabrian pine wood decreased. All treatment groups giving positive Δa^* and negative Δb^* values tended to turn reddish and bluish, respectively after weathering. Total color change (ΔE^*) was the lowest for only WBV coated Calabrian pine wood. Additionally, higher concentration levels of chemicals resulted in lower total color changes of Calabrian pine wood after weathering.

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