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Surface properties of wood material impregnated with copper-based chemical (Korasit KS) after natural weathering

Bakır esaslı kimyasal (Korasit KS) ile emprenye edilen ağaç malzemenin doğal yaşlandırma sonrası üst yüzey özellikleri

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Abstract

In this work, the surface characteristics of Oriental beech (Fagus orientalis L.) wood impregnated wit Korasit KS, a water-based and copper-containing impregnation substance, were examined after months of natural weathering. The impregnation material was penetrated into the wood material a 3% and 6% concentration levels according to the ASTM D 1413-07 (ASTM 2007) standard. According t the results, the L* values of all samples decreased after 3 months natural weathering. Impregnate samples provided better color stability than control samples. Moreover, this stability increased as th concentration amount increased in the impregnation process. While the gloss of all samples decrease after natural weathering, this decrease was more in control samples. According to the surfac roughness test results, all samples' Ra, Rz and Rq values increased after natural weathering. Th increase was more in the impregnated samples.

Özet

Bu çalışmada, su bazlı ve bakır içerikli bir emprenye maddesi olan Korasit KS ile emprenye edilen Doğ kayını (Fagus Orientalis L.) odununun 3 aylık doğal yaşlandırma sonrası yüzey özellikleri incelenmişti Emprenye malzemesi ağaç malzemeye ASTM D 1413-07 (ASTM 2007) standardına göre %3 ve % konsantrasyon seviyelerinde nüfuz edilmiştir. Sonuçlara göre tüm numunelerin L* değerleri 3 ayl doğal yaşlandırmadan sonra azalmıştır. Emprenye edilen örnekler, kontrol numunelerinden daha i renk stabilitesi sağlamıştır. Ayrıca emprenye işleminde konsantrasyon miktarı arttıkça bu stabilited artmıştır. Doğal yaşlandırma sonrası tüm numunelerin parlaklığı azalırken, bu azalma kontro numunelerinde daha fazla gerçekleşmiştir. Yüzey pürüzlülüğü test sonuçlarına göre tüm numuneleri Ra, Rz ve Rq değerleri doğal yaşlandırma sonrası artmıştır. Bu artış emprenye edilen örneklerde dah fazla olmuştur.

INTRODUCTION

The need for wood material is increasing day by day due to its thermal properties, good paint and varnishability, sound absorption, easy processing, and more aesthetic than other materials (Percin et al. 2009). Furthermore, wood material has more than 5000 uses (Yazici 2005). Despite all these advantages and usage area, wood material also has some undesirable features such as burning, exposure to mechanical effects, deformation as a result of exposure to weathering (Okcu 2006, Ors et al. 2006). Furthermore, since wood is a renewable organic material, it can be easily damaged by fungi and insects under suitable conditions, it can be biodegraded and this leads to a shortening of the useful life of wood in end use (Sivrikaya et al. 2017). In order to eliminate these disadvantages and increase the durability of the wood material, methods such as impregnation of the wood material with various chemical substances, chemical modification, and surface coating processes are required (Zelinka and Rammer 2005). Impregnation is the process of penetrating the wood material of various protective chemicals in order to prevent the wood material from rotting, burning and deformation (Ors and Keskin 2001, Kartal and Unamura 2004). However, these preservatives used can create a corrosive environment for metallic materials in contact with wood (Rammer et al. 2006, Lin et al. 2009, Zelinka and Stone 2011). Corrosion is the deterioration of alloys and metals as a result of electrochemical and chemical reactions with their surroundings. In addition, according to new findings, it is stated that non-metallic materials are also affected by environmental conditions (Can et al. 2020). Since CCA (Copper, Chrome, Arsenic), which is used as the most effective impregnation agent in the wood protection industry, has been banned due to its harmful effects on the environment, new generation impregnation materials have started to be used (Gezer et al. 2004). Boric acid and CCB (Copper, Chrome, Boron) are some of the new generation impregnation materials used against this negativity in the wood industry (Sivrikaya and Saraçbaşı 2004, Kartal et al. 2006).

Most of the wood preservative chemicals used against fungi interact either by inhibiting the formation of acetyl coenzyme (COA) or by inhibiting respiratory chain formulations (Zhang et al. 2009). It is thought that copper-containing impregnations provide metal-enzyme interaction with DNA modification and formation of high-activity free radicals produced by copper ions. The interaction between copper-containing preservatives and wood is very important in terms of both the performance of the preservative and its environmental effects (Zhang et al. 2009). Structural differences are observed between wood species because wood consists of complex structures such as cellulose, hemicellulose and lignin and contains different reactive groups (carbonyl, ether, acid, hydroxyl). These differences affect the fixation of chemical substances in wood (Vasishth 1996). In the literature, it was reported that wood impregnated with CCA and ACQ had lower light degradation in the accelerated weathering than that of non-impregnated wood (Jin et al. 1991). Grelier et al. (2000) investigated the photodegradation properties of woods impregnated with copper-amine. Their results showed that the samples impregnated with ACQ and CCA had better photo-oxidation (lignin) protection by accelerated weathering than the control samples. It has been determined that UV rays provide a good surface protection against UV rays for 1600 hours and especially on wood samples treated with ACQ (Grelier et al. 2000). Commercially, wood is treated with waterborne preservatives such as chromated copper arsenic (CCA), copper-azole, ammoniacal copper arsenate (ACZA), and amine/ammoniacal copper quat at a certain retention and penetration to increase the service life of wood (Feist and Hon 1984).

It was aimed to determine the changes in color, gloss, and surface roughness properties of wood material after 3 months of natural weathering by applying Korasit KS impregnation material to samples prepared from Oriental beech (*Fagus Orientalis* L.) in this study. The copper-based Korasit KS material used in the study was preferred because it is water-based, environmentally friendly, and harmless to human health.

MATERIALS AND METHODS

Preparation of Wood Material

The Oriental beech (*Fagus orientalis* L.) wood was prepared in 10 x 100 x 150 mm (radial, tangential, and longitudinal) dimensions for the natural weathering (color, gloss, and surface roughness) test by TSE 2470 (1976) standard.

Impregnation Procedure

According to ASTM D 1413-07, Oriental beech (*Fagus orientalis* L.) was impregnated with Korasit KS (ASTM 2007). The impregnation material used in this study was obtained from the Varkim company in the Izmir province of Turkey (Varkim 2022). After a pre-vacuum for 30 minutes of 760 mm Hg was used throughout the impregnation procedure for 30 minutes, air-dry test samples were allowed to diffuse in the solution at room temperature for the same amount of time. In order to measure the adhesion rate of the impregnation material and avoid being influenced by the moisture of the wood, samples were prepared to be dry both before and after impregnation. At concentrations of 3% and 6%, the impregnation material was applied to the wood material. The following equation was used to calculate the Korasit KS retention:

$$\text{Retention}=\frac{G,C}{V}X100 \text{ (gr/cm}^3\text{)} \quad (1)$$

Where;

G= T2 -T1
T_1 = Sample weight before impregnation (gr)
T_2 = Sample weight after impregnation (gr)
V = Sample volume (cm)
C = Concentration amount (%)

Natural Weathering Test

Wood panels are designed to expose samples to outdoor weather conditions according to ASTM D 358–55 (1970). The samples were placed on panels approximately 50 cm above the soil, facing south and at an angle of 45^0 (Figure 1). Then, the samples placed on the panels were exposed to weathering conditions for 3 months during summer (from June to August 2022) in the Mugla/TURKEY region (Baysal 2008, Kart et al. 2018). A total of 30 wood samples were prepared, 10 of which were wood samples for each sample type. Weathering conditions of Mugla during June-August 2022 were given in Table 1 (Turkish State Meteorological Service Database 2022).

Table 1. Weathering conditions of Mugla from June to August in 2022

Months	June	July	August
Average temperature (^{0}C)	23.1	28.0	26.1
The highest temperature (^{0}C)	33.3	39.5	37.6
The lowest temperature (^{0}C)	13.6	17.2	15.2
The number of the rainy days	11	0	5
Humidity (%)	55.5	33.8	53.3

Figure 1. An image of the experimental setup

Color Changes Test

The *L*, a*,* and *b** color parameters of the samples were calculated using the CIEL*a*b* method for the color test. Here, the a* and b* axes stand in for the chromaticity coordinates and the *L** axis represents lightness. Additionally, the *+a** and *-a** parameters denote red and green colors, respectively. In addition, the *+b** and *-b** parameters stand for yellow and blue, respectively. *L** value ranges from zero (black) to one hundred (white) (Zhang 2003). In this test, the color difference (AE*) was calculated using Equations 2 to 5 according to the ASTM D1536-58T (ASTM 1964) standard.

$$\Delta a^* = af^* - ai^* \quad (2)$$
$$\Delta b^* = bf^* - bi^* \quad (3)$$
$$\Delta L^* = Lf^* - Li^* \quad (4)$$
$$(\Delta E^*) = [(\Delta a^*)^2 + (\Delta b^*)^2 + (\Delta L^*)^2]^{1/2} \quad (5)$$

Where;

10 replicates were performed for each sample group. Also, *Δa**, *Δb**, and *ΔL** represent the changes between the first and last interval values.

Gloss Test

The gloss values of the test samples were calculated according to the ASTM D 523-14 (ASTM 2018) standard and with the (Micro-TRI-Gloss) device. The geometry chosen here represents an angle of incidence of 85. In addition, 10 repetitions were applied for each sample group in this test.

Surface Roughness Test

With a Mitutoyo Surftest SJ-301 instrument, roughness measurements were taken under DIN 4768 (1990) standard. In this work, the average peak-valley height (Rz), a measure of surface roughness, was measured. The stylus with a diamond tip had a 5 m radius and a 90° conical angle, and it was used to examine the surface roughness profile. The stylus was fed at a rate of 0.5 mm/s1 over an 8 mm sampling length (Zhong et al. 2013). Every treatment group received 10 replications. Parallel to the fibers, measurements of surface roughness was taken. A total of 3 measurements were taken from each sample.

Statistical Analysis

The data obtained from the experiments were transferred to the SPSS 16.0 statistical program, and the Duncan test applied at 95% confidence level was calculated. In this program, in order to determine homogeneity groups (HG), alphabetic letters are used to indicate whether there is statistical significance between sample types.

RESULTS AND DISCUSSION

Retention Amounts

Retention amounts of Oriental beech wood samples impregnated with Korasit KS are given in Table 2.

Table 2. Retention amounts of Oriental beech wood samples impregnated with Korasit KS

Chemicals	Retention (gr/cm³)
Control	-
Korasit KS (%3)	18.91
Korasit KS (%)	22.30

In this study, the retention amount was found to be 18.91 gr/cm^3 in samples impregnated with Korasit KS at the rate of 3%, while the retention amount was found as 22.30 gr/cm^3 in 6% samples.

Color Changes Test

Color changes test values of Oriental beech wood samples impregnated with Korasit KS are given in Table 3.

After natural weathering, the *L** values of all samples decreased. The reduction in the control samples was greater than in the impregnated samples. Furthermore, all samples showed a tendency to be reddish and yellowish, giving positive *a** and positive *b** values, respectively. According to the results of the total color change values (*ΔE**), the color stability of the impregnated samples was more preserved than the control samples. In addition, better color stability was achieved as the amount of concentration increased in the impregnation process. The highest color change was detected in the control samples. In addition, the lowest total color change (*ΔE**) was determined in the samples impregnated at 6% concentration. After natural weathering, the samples that were impregnated at 6% concentration were the ones that preserved the color stability the most and showed the most positive properties. According to Zhang et al. (2009), copper-amine complexes preserve the color of treated wood and increase the UV resistance of the surface by forming a complex with constituents of wood. As a result, copper-treated wood had enhanced color performance. It is supported by the information in the literature that the impregnation process provides color stability in wood material. Özgenç and Yıldız (2014) impregnated wood with micronized copper quat (MCQ), ammonia copper quat (Celcure AC 500), copper (II) sulfate pentahydrate (Cu(II)SO4.5H2O), and Didecyldimethylammonium chloride (DDAC). They then determined the color changes of these samples after accelerated weathering. According to their results, impregnation materials with Celcure AC 500 and MCQ provided less color change than DDAC and Cu(II)SO4.5H2O materials. In addition, Celcure AC 500 provided the lowest color change in the applied samples. Temiz et al. (2007) reported that complexing between chromium and guaiacyl units of lignin delayed the total color change in wood treated with CCA (chromated copper arsenate) and linseed oil. The application of chrome-copper-boron (CCB) treatment to wood material was advised by Yalinkilic et al. (1999) to stabilize the surface color of varnish-coated wood panels. According to Humar et al. (2011), copper-ethanolamine-based wood preservatives affect some properties of water-borne acrylic coatings used on wooden substrates. In our study, copper-containing impregnation material gave positive values after natural weathering conditions. Therefore, our study was compatible with the literature. The graphical representation of the color changes test values after weathering is shown in Figure 2.

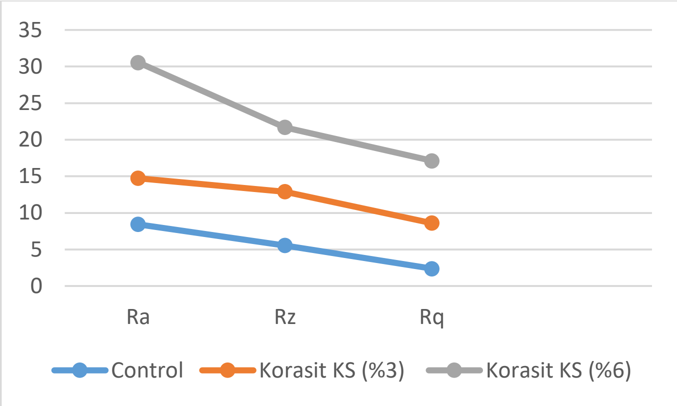

Figure 2. Color changes test values after weathering

Gloss test

Gloss test values of Oriental beech wood samples impregnated with Korasit KS are given in Table 4.

Before weathering, it was determined that the impregnated samples showed lower gloss values compared to the control samples. Accordingly, before weathering, the impregnation treatment had a negative effect on the gloss values of the wood samples.

After natural weathering, the gloss of all samples decreased. While less decrease was observed in the samples impregnated compared to the control samples, a statistical difference was detected between the control and impregnated samples. Chromophoric structures in lignin, which are released as a result of the photodegradation of wood, absorb light. During the gloss measurement, the light coming to the sample surface is absorbed by the lignin, causing the value measured by the device to be low (Can 2018). Baysal et al. (2014) determined that Scotch pine wood treated with copper-based impregnation materials decreased in brightness after 500 hours of accelerated weathering. Baysal et al. (2016) investigated the gloss values of bamboo (*Phyllostachys basmasoides*) wood impregnated with Tanalith-E, ACQ and Wolmanite-CB after accelerated wethering. Results showed that following an accelerated weathering period, the gloss of bamboo samples that had been impregnated diminished. Turkoglu et al. (2015) studied at the gloss values of Scots pine wood that had been pre-impregnated with a few copper-based

Table 3. Color changes test values of Oriental beech wood samples impregnated with Korasit KS

Chemicals	Concentration (%)	Color values after natural weathering						Total Color Changes	
		Δ_L*	Std. dev.	Δ_a*	Std. dev.	Δ_b*	Std. dev.	Δ_E*	Homogeneity group
Control	-	-7.51	1.12	3.15	0.47	2.58	0.38	8.54	A
Korasit KS	3	-6.90	1.03	1.69	0.25	1.04	0.15	7.17	A
Korasit KS	6	-3.05	0.45	0.11	0.02	1.62	0.24	3.45	B

Note: *Std. dev.: Standard deviation.*

Table 4. Gloss test values of Oriental beech wood samples impregnated with Korasit KS

Chemicals	Concentration (%)	Gloss values before natural weathering 85⁰ Mean	Gloss values after natural weathering 85⁰ Mean	Gloss changes values after natural weathering (%)	
				85⁰	Homogeneity group
Control	-	2.70 (0.45)	1.30 (0.38)	-51.85	A
Korasit KS	3	1.73 (0.13)	0.95 (0.21)	-45.08	AB
Korasit KS	6	1.88 (0.78)	1.21 (0.14)	-35.63	B

Note: *Values in parentheses represent the standard deviation.*

compounds. They discovered that pre-impregnation with chemicals resulted in a loss of gloss in samples of Scots pine wood. Ozdemir et al. (2015) found that water-based wood preservatives decreased the gloss values of the wood and increased the porosity of the surface. Our study's findings in this area were consistent with those found in the literature. The graphical representation of the gloss test values after weathering is shown in Figure 3.

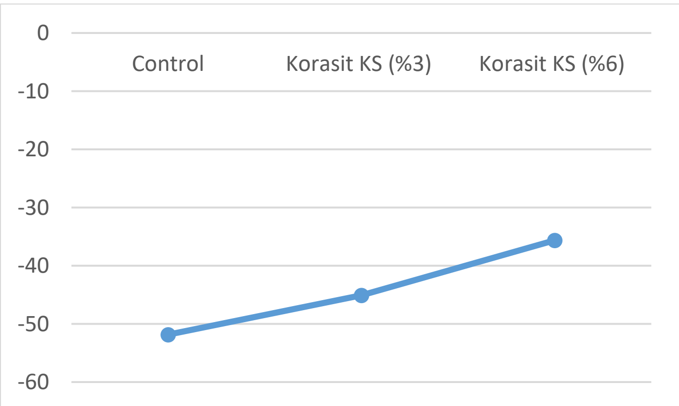

Figure 3. Gloss test values after weathering (%)

Surface Roughness Test

Surface roughness values of Oriental beech wood samples impregnated with Korasit KS are given in Table 5.

After natural weathering, an increase in *Ra*, *Rz,* and *Rq* values of all samples were detected. This increase in the impregnated samples was higher than in the control samples. In addition, as the concentration amount increased in the impregnation process, the surface roughness value of the samples increased. In this study, the impregnation process caused the wood material to have a rougher surface. A statistical difference was found in the control and impregnated samples. According to investigations on surface roughness, lignin loss, color measurements, and microscopic examinations, copper azole demonstrated resistance to weathering effect and reduced the degradation caused by stain-fungi (Temiz et al. 2005). Due to the heterogeneity and anisotropy of wood, surface roughness is a complicated phenomenon. The evaluation of the surface roughness of wood should take into account several factors, including anatomical variations, growing traits, machining properties, pre-treatments (such as steaming, drying, etc.) applied to the wood before machining, and copper-wood interactions after impregnation (Aydin and Colakoglu 2003). Copper is still the primary biocide component used today in the protection of wood in ground contact or exposed to weather, although copper-based preservatives have been widely utilized for more than a century (Lebow et al. 2004). Numerous researchers have observed that wood treated with alkylammonium compounds (AACs), copper ethanolamine (Cu-EA), ammoniacal copper quat, copper-azole, tanalith-E 3491, and wolmanit CX-8 is more resistant to weathering effects (Jin et al. 1991, Cornfield et al. 1994, Temiz et al. 2005, Nejad and Cooper 2011). In plywood treated with various fire retardants, rougher surfaces developed as the concentration increased, according to Ayrilmis et al. (2006). Baysal et al. (2021)

Table 5. Surface roughness values of Oriental beech wood samples impregnated with Korasit KS

Chemicals	Concentration (%)	Surface roughness values after natural weathering (%)					
		R_a	H.G	R_z	H.G	R_q	H.G
Control	-	8.43 (1.26)	A	5.55 (0.83)	A	2.39 (0.35)	A
Korasit KS	3	14.73 (2.20)	B	12.91 (1.93)	B	8.62 (1.29)	B
Korasit KS	6	30.54 (4.58)	C	21.70 (3.25)	C	17.12 (2.56)	C

Note: *Values in parentheses represent the standard deviation. H.G.: Homogeneity group.*

impregnated Calabrian pine (*Pinus brutia*) wood with Celcure C4, Korasit KS, and Tanalith E 8000. Then they investigated the roughness properties of wood after weather conditions. According to the results, higher surface roughness values were determined in the impregnated wood samples. Results from our study by Ayrilmis et al. (2006) and Baysal et al. (2021) is consistent with the results. The graphical representation of the surface roughness test values after weathering is shown in Figure 4.

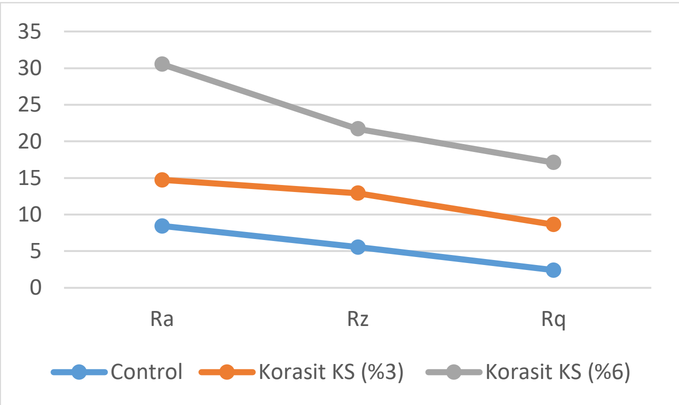

Figure 4. Surface roughness test values after weathering (%)

CONCLUSIONS

The results showed that *ΔL** values of control and impregnated wood samples decreased and this decrease was more in control samples. It also showed that the control and impregnated samples tended to be reddish and yellowish, respectively, giving positive *a** and *b** values after natural weathering. The total color changes showed that the impregnated samples gave lower values and protected the color stability better than the control sample. The gloss of control and impregnated samples increased before weathering while it decreased after weathering. The least decrease was observed in the samples impregnated at 6% concentration after weathering. The surface roughness of all samples increased after weathering. This increase after weathering was less in the control sample compared to the impregnated samples.

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