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IMPACT OF UV IRRADIANCE ON SELECTED PARAMETERS OF SCOTS PINE IMPREGNATED WITH SOME COMMONLY USED FIRE- RETARDANTS

ERGUN BAYSAL

Mugla Sitki Kocman University, Faculty of Technology Department of Wood Science and Technology

Mugla, Turkey

CAGLAR ALTAY

Adnan Menderes University, Aydin Vocational School, Department of Furniture and Decoration

Aydin, Turkey

Turkay Turkoglu Mugla Sitki Kocman University, Koycegiz Vocational School Department of Forestry Mugla, Turkey

Hilmi Toker

Mugla Sitki Kocman University, Faculty of Technology, Department of Wood Science and Technology Mugla, Turkey

> Huseyin Peker Artvin Coruh University, Faculty of Forestry Department of Forest Industry Engineering Artvin, Turkey

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ABSTRACT

This study was designed to determine some selected parameters such as gloss, surface roughness, and color changes of Scots pine wood impregnated with commonly used fire-retardant (FR) chemicals after UV irradiance. Sodium acetate (SA), ammonium chlorite (AC), zinc chlorite (ZC), ammoniumsulphate (AS), and di ammonium phosphate (DAP) were used as fire retardants. Wood specimens were prepared from Scots pine (*Pinus sylvestris* L.). Before test, wood specimens were impregnated with 5 % aqueous solution of chemicals according to ASTM D 1413-76 standard.

Results showed that UV irradiance caused gloss loss and increase surface roughness of FR impregnated and un-treated (control) Scots pine specimen. DAP was the most effective chemical in terms of reducing gloss loss and surface roughness of Scots pine after 750 h UV irradiance exposure. UV irradiance caused a dark, reddish, and yellowish color of impregnated and un-treated (control) Scots pine specimen after all UV irradiance periods. Total color changes in color (ΔE^*) exhibited a systematic trend to higher values with increasing UV irradiance time. Total color changes of ZC impregnated Scots pine were the lowest after 750 h UV irradiance exposure.

KEYWORDS: Scots pine, fire-retardants, gloss, surface roughness, color, UV irradiance.

INTRODUCTION

Among the construction materials which are used by people wood holds a special place because of its impressive range of attractive properties, including low thermal extension, low density, and high enough mechanical strength (Bektha and Niemz 2003). It has been popularly and favorably used as a decorative material owing to its aesthetic appearance and characteristics properties (Chang and Chang 2001). However, wood surfaces exposed to outdoors are rapidly degraded because lignin strongly absorbs UV light, which leads to radical-induced depolymerization of lignin and cellulose, the major structural constituents of wood (Evans et al. 2005). The yellowing, browning, and/or graying of wood surfaces indicate the modification of lignin when wood is exposed outdoors (Grelier et al. 2000). The surface properties of wood materials can be enhanced easily by impregnating and finishing with various preservatives to provide different performance characteristics for individual applications, such as high hardness, impact resistance, suitable gloss, and chemical resistance (Chang and Lu 2012, Degirmentepe et al. 2015a). Although, not generally classified as a wood finish, the preservatives protect against weathering (in addition to decay), and a large quantity of preservative-treated wood is exposed to outdoors without any additional finish (Feist 1987). The most effective method of preventing the photodegradation of wood involves a treatment with dilute aqueous solutions of inorganic salts, particularly hexavalent chromium compounds. The application of chromium trioxide to wood surfaces prevents lignin degradation during natural weathering (Evans et al. 1992, Kiguchi and Evans 1998). Pizzi (1980) reported that the beneficial effects of chromium were attributed to the formation of complexes between chromium and guaiacyl units of lignin. Sell et al. (1974) reported that high resistance of the CCB-impregnated wood against weathering has been attributed to the protective effect of Cr-Cu salt solutions of the wood surface. In another study, Williams et al. (1996) found that the chromium oxides in copper-chromate-arsenic (CCA) which bond to the wood after treatment, decrease photodegradation of the wood surface. Chromated copper arsenate (CCA) has provided long-term protection against weathering and erosion (Feist and Ross 1995, Jirouš-Rajković et al. 2004) but it is no longer being produced for use in most residential settings, because it contains chromium and arsenic. Nowadays, several new copper-based wood preservatives such as tanalith-E (TN-E) and adolit-KD5 (AD-KD5) are being used in the forest products industry instead of CCA (Turkoglu et al. 2015a, b). The focus on copper-based preservatives has increased following concerns about environmental effects of chromium and arsenic (Freeman and McIntyre 2008). Copper forms certain complexes with wood components, such as coppercellulose complexes, copper-lignin complexes, and crystalline or amorphous inorganic/organic copper compounds, and reduces the degradation of the wood surface from weathering factors (Temiz et al. 2005, Grelier et al. 2000). Therefore, weathering aspects of treated wood with new

wood preservatives developed a practical importance (Temiz et al. 2007). The most commonly used fire-retardant chemicals in the wood industry are inorganic salts and include ammonium and ammonium phosphate, ammonium chloride, ammonium sulfate, borax, boric acid, phosphoric acid, and zinc chloride (Woo and Schniewind 1987). Fire-retardant chemicals drastically reduce the rate at which flame spread across the wood surface, thereby reducing the capacity of the wood to contribute to a fire (LeVan and Tran 1990, LeVan and Winandy 1990). The UV irradiance performance of commonly used fire-retardant preservatives in wood preservation industry is mostly unknown. Therefore, this study was designed to investigate some selected parameters such as gloss, surface roughness, and color changes of Scots pine impregnated with commonly used fire-retardants. Ammonium sulphate, ammonium chloride, di ammonium phosphate, zinc chlorite, and sodium acetate were selected as fire retardant chemicals for UV irradiance performance of Scots pine.

MATERIAL AND METHODS

Preparation of test specimens and chemicals

Wood specimens measuring 6 x 75 x 150 mm were prepared from air-dried sapwood of Scots pine (*Pinus sylvestris* L.). Aqueous solution of chemicals were dissolved in distilled water to concentration 5 percent. Wood specimens were oven dried at 103 ± 2°C before and after treatment.

Impregnation method

Wood specimens were impregnated with 5% aqueous solution of fire-retardant chemicals according to ASTM D 1413-76 (2007). Retention of chemicals was calculated from the following equation:

$$Retention = \frac{G \times C}{V} \times 10 \qquad (kg \cdot m^{-3})$$
 (1)

where:

G - amount of solution absorbed by wood that is calculated by M_2 - M_1 ,

M₂ - masses of wood after impregnation (g),

M₁- masses of wood before impregnation (g),

C - solution concentration as percentage, and

V - volume of the specimen as cm³.

Gloss test

The gloss values of wood specimens were determined according to ASTM D 523-14 (2014) with a measuring device (Micro-TRI-Gloss). The chosen geometry was an incidence angle of 60°. Results were based on a specular gloss value of 100, which relates to the perfect condition under identical illuminating and viewing conditions of a highly polished, plane, black glass surface. Five replicates were made for each treatment group. Gloss measurements were made in parallel to the fibers.

Surface roughness test

The Mitutoyo Surftest SJ-301 instrument was employed for surface roughness measurements according to DIN 4768 (1990). Surface roughness parameter (Rz) is an arithmetic mean of the 10-point height of irregularities. Five replicates were made for each treatment group. Surface roughness measurements were made in parallel to the fibers.

Color test

The color parameters a^* , b^* , and L^* were determined by the CIEL* a^*b^* method. The L^* axis represents the lightness, whereas a^* and b^* are the chromaticity coordinates. The $+a^*$ and $-a^*$ parameters represent red and green, respectively. The $+b^*$ parameter represents yellow, whereas $-b^*$ represents blue. L^* can vary from 100 (white) to zero (black) (Zhang 2003). The colors of the specimens were measured by a colorimeter (X-Rite SP Series Spectrophotometer) before and after UV irradiance. The measuring spot was adjusted to be equal or not more than one third of the distance from the center of this area to the receptor field stops. The total color changes (ΔE^*) was determined for Scots pine wood as follows ASTM D 1536–58 T (1964). Equations 2-5:

where: Δa^* , Δb^* , and ΔL^* are the changes between the initial and final interval values. Five replicates were made for each treatment group. Color measurements were made in parallel to the fibers.

UV irradiance test

UV irradiance experiment was performed in a QUV weathering tester with eight UVA 340 lamps. The weathering schedule involves a continuous light irradiation of 8 h following with a condensation for 4 h. The average irradiance was 0.89 W·m⁻² at 340 nm wavelengths. The temperature of the light irradiation period and the condensation period was 60°C and 50°C, respectively. Wood specimens were mounted on aluminum panels before placing in the QUV. The changes on wood specimens were monitored every 250 h for a total 750 h.

RESULTS AND DISCUSSION

Gloss changes

Glossiness, the property of reflecting light in a mirror is very important for the aesthetic and decorative appearance of surfaces (Cakicier et al. 2011). Gloss of Scots pine wood specimens was measured at a 60° angle of incidence using a gloss meter. Gloss values of Scots pine wood specimens before and after UV irradiance are given in Tab. 1, along with the retention values of the wood specimens. Retention values were 32.79 kg·m⁻³, 30.69 kg·m⁻³, 35.55 kg·m⁻³, 33.93 kg·m⁻³, and 33.34 kg·m⁻³ for SA, AS, DAP, ZC, and AC impregnated Scots pine specimens, respectively.

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Impregnation chemicals	Retention (kg·m ⁻³)	Before	UV irradiance								
		UV	After 25	0 hours	After 50	00 hours	After 750 hours				
		irradiance	Mean	%	Mean	%	Mean	%			
Control	-	3.88	2.98	-23.20	2.75	-29.18	2.50	-35.62			
Sodium acetate	32.79	3.56	2.90	-18.54	2.64	-25.84	2.22	-37.64			
Ammonium sulphate	30.69	3.50	2.68	-23.43	2.52	-28.00	2.22	-36.57			
Di ammonium phosphate	35.55	3.20	2.90	-9.38	2.86	-10.63	2.74	-14.38			
Zinc chlorite	33.93	2.82	2.66	-5.67	2.52	-10.64	2.34	-17.02			
Ammonium chlorite	33.34	3.16	2.42	-23.42	2.32	-26.58	2.06	-34.81			

Tab. 1: The gloss changes of specimens before and after each UV irradiance periods.

Note: Five replicates were made for each treatment group.

FR impregnation had a negative effect on the gloss of the Scots pine wood before UV irradiance. Because, our results showed that FR treatment before UV irradiance decreased the gloss of Scots pine in some extent. For example, while gloss value of un-treated (control) Scots pine specimen was 3.88 before UV irradiance, gloss values of FR impregnated Scots pine changed from 2.82 to 3.56 before UV irradiance. Simsek and Baysal (2012) investigated gloss values of borate treated some wood species. They found that borate treatments remarkably decreased gloss values of wood species. Ozdemir et al. (2015) reported that water-based wood preservatives increase surface porosity and raised fibers decrease gloss value. Baysal et al. (2016) investigated gloss changes of some copper- based chemicals impregnated Scots pine before weathering. They found that gloss values of copper containing chemicals impregnated Scots pine were highly decreased compared to un-treated (control) Scots pine before weathering. Our results are in good agreement with data Simsek and Baysal (2012), Ozdemir et al. (2015), and Baysal et al. (2016). It can be explained that impregnation process with the solutions might limit the glossiness to point in Scots pine before UV irradiance, probably owing to the absorption and dispersion of the reflected rays by salt crystals prominent in the large lumens of the vessel in the wide early wood sections of the grains. Photoactive ion on the wood surface was assumed to cause same losses in glossiness before UV irradiance. Gloss values of FR impregnated and un-treated (control) Scots pine were decreased after each UV irradiance periods. It may be due to abrasion on the wood surfaces along with the erosion causes gloss loss (Baysal et al. 2016). Baysal et al. (2016) investigated gloss changes of boron and copper-based preservative treated bamboo specimens after 168 to 672 h UV irradiance periods. They found that gloss values were decreased for treated and un-treated bamboo specimens for each UV irradiance periods. Our results are in good agreement with data Baysal et al. (2016). In our study, after 250 h UV irradiance period, while gloss loss was the lowest with ZC impregnated Scots pine (5.67%), it was the highest with AS impregnated Scots pine (23.43%). In this period, gloss loss of SA, DAP, and ZC impregnated Scots pine were lower than un-treated (control) Scots pine specimen. After 500 h UV irradiance period, gloss losses of FR impregnated Scots pine were lower than un-treated (control) Scots pine specimen. While gloss loss was the 29.18% for un-treated (control) Scots pine specimen, it was changed from 10.63 to 28% for FR impregnated Scots pine. The lowest gloss loss was 10.63% for DAP treated Scots pine in this stage. After 750 h UV irradiance period, gloss loss was 35.62% for un-treated (control) Scots pine specimen, it was changed from 14.38 to 37.64% for FR impregnated Scots pine. Our results showed that gloss loss was the lowest DAP impregnated Scots pine (14.38%) followed by ZC impregnated Scots pine (17.02%), respectively after 750 h UV irradiance. Other treatment groups gave nearly same results in terms of gloss loss values with un-treated (control) Scots pine specimen after 750 h UV irradiance exposure.

As a result, FR impregnation decreased gloss values of Scots pine before UV irradiance. UV irradiance caused gloss loss for un-treated (control) and FR impregnated Scots pine during and after UV irradiance exposure. DAP and ZC impregnated Scots pine gave better results than un-treated (control) Scots pine specimen in terms of gloss loss during and after UV irradiance exposure.

Surface roughness changes

Tab. 2 shows surface roughness of FR impregnated Scots pine specimens before and after UV irradiance from 250, 500, and 750 h UV irradiance exposure. FR treatment increased surface roughness of Scots pine wood specimens before UV irradiance.

Tab. 2: The surface roughness of specimens before and after the each UV irradiance periods.

	Before	After UV irradiance									
Impregnation chemicals	UV	After	r 250	A from 50	0 h 011m2	After 750 hours					
	irradiance	ho	hours		After 500 hours		After 750 flours				
	Rz	Rz	%	Rz	%	Rz	%				
Control	16.38	18.62	13.68	19.59	19.61	19.91	21.57				
Sodium acetate	21.59	22.25	3.06	23.15	7.24	26.06	20.70				
Ammonium sulphate	19.66	21.20	7.83	23.79	21.00	24.95	26.90				
Di ammonium phosphate	22.22	23.64	6.39	24.72	11.27	25.70	11.37				
Zinc chlorite	17.91	21.16	18.15	25.84	44.24	29.06	62.20				
Ammonium chlorite	21.10	22.61	7.16	23.33	10.56	24.33	15.27				

Note: Five replicates were made for each treatment group.

Therefore, it was concluded that the surfaces of FR impregnated Scots pine were rougher after treatment. The wooden materials with rough surface need more sanding process than those with smooth surface. However, sanding process causes a decrease in of material (Follrich et al. 2006). Ayrilmis et al. (2006) investigated the effect of various fire retardants on surface roughness of plywood. They found that samples treated with 6% concentration of boric acid had the highest Rz value. Also, they reported that the Rz values of borax, boric acid, and monoammonium phosphate at higher concentration (6% or 11%) were always rougher than at lower concentrations (3%) except for 11 % concentration of ammonium phosphate. Maldas and Kamdem (1998) reported that surface of CCA treated wood was also rougher than that of un-treated (control) wood. In another study, Baysal et al. (2016) investigated surface roughness of boron and copper containing chemicals treated Scots pine. They found that surface roughness parameter (Rz) of preservative treated Scots pine were higher than un-treated (control) Scots pine specimen. Our results are in good agreement with these researcher's findings. It can be due to the chemical salts on wood surface caused to increase surface roughness of Scots pine wood after impregnation. However, the roughness of wood is a complex phenomenon. Several factors, such as anatomical structure of wood, growing characteristics, machining properties and pre-treatments of wood before machining, should be considered for the evaluation of the surface roughness of wood (Aydin and Colakoglu 2003, Aydın and Colakoglu 2005, Temiz et al. 2005). UV irradiance increased the surface roughness of un-treated and FR impregnated Scots pine. Turkulin et al. (2004) mentioned that light irradiation mostly degraded the middle lamella, which is between two cell walls and holds the cells together. This degradation increases the roughness of the wood surface (Tolvaj et al. 2011). Kerber et al. (2016) also reported that in addition to the leaching of lignin

degraded by natural weathering reactions, the increase in the roughness of the wood is also related to the sudden changes of moisture content (absorption and desorption of the moisture content) causing the presence of superficial cracks. In our study, after 250 h UV irradiance, the increase in Rz, was 13.68% for un-treated (control) Scots pine, it changed from 3.06% to 18.15% for FR impregnated Scots pine. In this period, except for zinc chlorite treatment, other all treatment groups gave smoother surface than un-treated (control) Scots pine specimen. Upon impregnation with chemicals, SA impregnated Scots pine gave the best results, in terms of decreased surface roughness after 250 h UV irradiance. After 500 h UV irradiance exposure, surface roughness increases of AS and ZC treated Scots pine were higher than un-treated (control) Scots pine specimen, in impregnation chemicals, SA gave the best results in terms of decreased surface roughness followed by AC and DAP, respectively. After 750 h UV irradiance exposure, except for AS and ZC treatment, other treatment groups gave better results in terms of surface roughness compared to un-treated (control) Scots pine specimen. While increase of Rz was the highest for ZC treated Scots pine, it was the lowest for DAP treated Scots pine after 750 h UV irradiance. Considering the general behavior of FR preservatives on surface roughness of Scots pine samples after UV irradiance, while DAP, SA, and AC treatment seemed to ensure more smooth surfaces than other preservatives after UV irradiance, surface roughness of the Scots pine was the highest impregnated with ZC after each and all UV irradiance exposure. Ozgenc and Yildiz (2014) investigated the surface roughness of some wood species treated with copper-containing new generation preservatives after artificial weathering. They found that during the weathering time, the surface roughness values of wood treated with copper-based preservatives reduced for all wood species. Compared to the un-treated wood specimen, the treatment with all preservatives, except for didecyldimethylammonium chloride (DDAC), decreased the surface roughness after artificial weathering test. Temiz et al. (2005) determined that the surface roughness values of CCA-treated Scots pine were lower than that of un-treated Scots pine after accelerated weathering. According to our results except for AS and ZC treatment, other treatment groups gave smoother surface than un-treated (control) Scots pine specimen after 750 h UV irradiance exposure.

As a result, FR impregnation increased surface roughness values of Scots pine before UV irradiance. UV irradiance caused rougher wood surface for un-treated (control) and FR impregnated Scots pine during and after UV irradiance exposure. In impregnation chemicals, while DAP, SA, and AC treatment gave the smooth surfaces, AS and ZC treatment gave rougher surface than un-treated (control) Scots pine specimen after 750 h UV irradiance exposure.

Color changes

Tab. 3 shows L^* , a^* , and b^* values of un-treated (control) Scots pine, and impregnated Scots pine specimens before and after weathering, and also presents the values of change for all three color parameters (ΔL^* , Δa^* , and Δb^*), as well as the total color changes (ΔE^*) of the Scots pine wood specimens after 250, 500, and 750 h UV irradiance.

Impregnation	Before UV irradiance			After 250 hours			After 500 hours				After 750 hours				
chemicals	Li^*	ai*	bi*	ΔL^*	Δa^*	Δb^*	ΔE^*	ΔL^*	Δa^*	Δb^*	ΔE^*	ΔL^*	Δa^*	Δb^*	ΔE^*
Control	74.63	6.92	24.52	-16.56	+8.4	+9.23	20.73	-19.86	+3.84	+11.47	23.25	-21.72	+3.49	+8.84	23.70
Sodium acetate	68.91	6.29	23.83	-19.36	+6.12	+3.58	20.62	-20.72	+1.62	+4.81	21.33	-22.65	+0.76	+3.23	22.89
Ammonium sulphate	70.08	5.02	24.46	-22.88	+10.4	+1.23	25.14	-25.74	+4.61	+4.05	26.47	-28.97	+4.14	+0.86	29.28
Di ammonium phosphate	72.18	6.88	23.34	-20.71	+7.79	+6.29	23.00	-25.16	+2.91	+7.84	26.51	-26.06	+2.14	+4.60	26.55
Zinc chlorite	71.53	5.89	23.27	-18.52	+7.35	+6.63	20.99	-20.08	+3.06	+8.15	21.89	-21.87	+2.76	+6.10	22.87

Ammonium chlorite 70.88 5.25 22.13 -23.9 +9.53 +4.63 26.14 -28.53 +4.53 +5.66 29.43 -33.44 +3.81 +0.81 33.67

Tab. 3: The color changes of specimens before and after each UV irradiance periods.

Note: Five replicates were made for each treatment group.

Our results showed that that FR treatment darkened Scots pine wood surface. While Li* of un-treated (control) Scots pine specimen was 74.63, before UV irradiance, it changed from 68.91 to 72.18 for FR impregnated Scots pine before UV irradiance. Also, FR treatment caused to decrease in a^* and b^* of Scots pine before UV irradiance. The decrease in the chromaticity coordinates, a^* and b^* for Scots pine indicated that the greenish and bluish due to FR treatment. Baysal (2012) investigated color changes of CCA treated Scots pine before UV irradiance. He found that CCA treatment decreased L^* , a^* , and b^* values of Scots pine compared to un-treated (control) Scots pine. Our results are consistent with data Baysal (2012). Surface of Scots pine had a dark, reddish, and yellowish color after all UV irradiance periods. Reddish and yellowish surfaces of Scots pine after UV irradiance are in consentient with the surfaces of bamboo exposed to natural weathering (Kim et al. 2008), and to photo-irradiation (Wang and Ren 2008). The negative lightness stability (ΔL^*) values occurred during the UV irradiance. Therefore, the wood surface got rougher and darker during the UV irradiance (Grelier et al. 2000). Depolymerization of the lignin on the exposed surface may also render the surface darker (Temiz et al. 2005). The darkening of Scots pine might have been due to the degradation of lignin and other non-cellulosic polysaccharides (Hon and Chang 1985, Grelier et al. 2000, Petric et al. 2004). Dark color as indicated by the negative ΔL^* values with the exposure period were also recorded by Wang and Ren (2008), Qin and Yu (2009), and (Turkoglu et al. 2015a, b, c). Positive values of Δa^* indicate a tendency of wood surface to become reddish. Positive values of Δb^* indicate a tendency of wood surface to become yellowish. The results demonstrated that Δa^* and Δb^* of un-treated (control) and FR impregnated Scots pine had positive values after UV irradiance. The increase in the chromaticity coordinate (Δa^* and Δb^*) may be explained by the changes of some chromophoric groups of lignin (Grelier et al. 2000). After all UV irradiance periods, FR treatment resulted in higher dark surfaces compared to un-treated (control) Scots pine surface. A darker surface in preservative impregnated samples was probably related to preservative impregnation reduced the stabilization of wood color in the visible region by increasing the lignin degradation resulting from UV light. Total color change of Scots pine increased with the increase in UV irradiance period. Wood discoloration is thought to be related with the formation of colored unsaturated carbonyl compounds (quinones) since photochemical reactions of lignin occur (Wang and Ren 2008). ZC and SA treatment showed to be best preservative to stabilize color of Scots pine against UV irradiance factors in this study. Total color changes of AS, DAP, and AC impregnated Scots pine were higher than un-treated Scots pine (control) specimens during and after UV irradiance exposure. Zhang and Kamdem (2000) and Zhang et al. (2009) suggested the effects of formation of metal complexes between wood components and inorganic ions against to weathering control. In our study, wood ion complexes formed at the wood surfaces possibly provide the wood surface with resistance by blocking the free phenolic groups (Grelier et al. 2000). The color stability of the Scots pine samples after 750 h of UV irradiance was in the following order: ZC≥ SA ≥ Control ≥ DAP ≥AS≥AC. Baysal et al. (2016) investigated the color changes of Scots pine (Pinus sylvestris) samples impregnated with boron and copper-based preservatives during UV irradiance from 168 h to 672 h. Results showed that surface Scots pine samples had a dark, reddish and yellowish color after UV irradiance. Moreover, the highest dark color based on the initial stage was seen during the first UV irradiance period (168 h). Our results are in good agreement with data Baysal et al.

As a result, FR impregnation decreased lightness values of Scots pine before UV irradiance. UV irradiance caused dark, reddish, and yellow color for FR impregnated and un-treated (control) Scots pine during and after UV irradiance exposure. Total color changes were lower ZC and SA impregnated Scots pine than un-treated (control) Scots pine specimen during and after UV irradiance exposure.

CONCLUSIONS

Surface roughness, gloss, and color changes of FR impregnated Scots pine after UV irradiance were investigated in this study.

FR treatment caused gloss loss and increased surface roughness of Scots pine before UV irradiance. UV irradiance increased surface roughness and decreased gloss values of Scots pine wood specimens. While in gloss values, DAP and ZC impregnated Scots pine gave better results than un-treated (control) Scots pine specimen, surface roughness values, DAP, AC, and SA impregnated Scots pine gave better results than un-treated (control) Scots pine specimen after UV irradiance. UV irradianced un-treated (control) and FR impregnated Scots pine wood specimens became darker, yellowish, and reddish. Total colors change of un-treated (control) Scots pine wood was lower than AS, DAP, and AC impregnated Scots pine after UV irradiance. In FR chemicals, ZC impregnated Scots pine gave the best results in terms of best color stabilization after 750 h UV irradiance exposure.

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*Ergun Baysal
Mugla Sitki Kocman University
Faculty of Technology
Department of Wood Science and Technology
Mugla
Turkey
*Corresponding author: ergun69@yahoo.com

PHONE: +90-252-211 1708

CAGLAR ALTAY
ADNAN MENDERES UNIVERSITY
AYDIN VOCATIONAL SCHOOL
DEPARTMENT OF FURNITURE AND DECORATION
AYDIN
TURKEY

Turkay Turkoglu
Mugla Sitki Kocman University
Koycegiz Vocational School Department of Forestry
Mugla
Turkey

HILMI TOKER

MUGLA SITKI KOCMAN UNIVERSITY

FACULTY OF TECHNOLOGY

DEPARTMENT OF WOOD SCIENCE AND TECHNOLOGY

MUGLA

TURKEY

Huseyin Peker Artvin Coruh University, Faculty of Forestry Department of Forest Industry Engineering Artvin Turkey