

WEATHERING PROPERTIES OF WOOD SPECIES TREATED WITH DIFFERENT COATING APPLICATIONS

Ozlem Ozgenc,^{a,*} Salim Hiziroglu,^b and Umit C. Yildiz^c

The objective of this study was to evaluate the discoloration of European beech (*Fagus sylvatica*) and Scots pine (*Pinus sylvestris*) specimens treated with different chemicals and surface coated with different UV absorbers before being subjected to artificial weathering. The results showed that the influence of coatings containing UV absorbers (UV screeners micronized TiO₂ and UVA of hydroxyphenyl-s-triazine types) were similar to each other. The UV screener TiO₂ led to the least discoloration of the coated wood surface, closely followed by the UVA of hydroxyphenyl-s-triazines (HPT). The color stability was determined to be better for pine wood treated with micronized copper preservative coated with UV absorber, in comparison to when it was only coated with UV absorbers and then subjected to weathering. Microscopic observation revealed that the clear-coats penetration behavior was different in wood preservative-treated and in untreated wood of Scots pine, which has various extractives. However, the color stability and coating penetration was nearly the same in beech wood treated with preservatives and in untreated beech wood. We provide an explanation for why these effects occurred and discuss the implications of our findings for the development of weather-resistant wood materials.

Keywords: Accelerated weathering; Wood Preservative; Coating; Color change

Contact information: a: Faculty of Technology, Department of Woodworking Industry Engineering, Karadeniz Technical University 61830 Trabzon –Turkey; b: Department of Natural Resource, Ecology & Management, Oklahoma State University, Stillwater, Oklahoma 74078-6013-USA; c: Faculty of Forestry, Department of Forest Products Engineering, Karadeniz Technical University, 61080 / Trabzon-Turkey; *Corresponding author: ozlem_ozgenc@hotmail.com

INTRODUCTION

Weathering is a complex process that modifies and degrades the overall molecular structure of wood and wood-based products. Generally, surface degradation of wood due to weathering is initiated by solar radiation, as well as other environmental factors. It is a well known fact that the shorter wavelengths of the solar spectrum having higher photon energies are more deeply absorbed into polymeric materials such as wood-based material, and the structure of chemical bonding within the cell wall is adversely influenced. The weathering initially causes the color of the wood surface to change, followed by the occurrence of surface checking and increased roughness of the samples. The discoloration of the surface is a direct indication of the chemical modification in the cell wall due to weathering. Most of the time, weathered wood would have a more pronounced yellow color than unexposed wood as a result of the modification of lignin and hemicelluloses. Such discoloration in the cell wall is influenced by photochemical reactions leading to the degradation of wood constituents, mainly lignin. A major part of solubilized lignin during degradation is washed out by rain. However, fiber-rich cellulose with a higher resistance

against ultraviolet light degradation remains in the cell wall without significant modification and results in the wood acquiring a whitish to gray color (Feist and Hon 1984; Rowell and Barbour 1988).

Wood products used under outdoor environmental conditions are treated or finished using different types of chemicals with the purpose of protection so that the overall service life of the product is extended. Two basic types of finishing or treatment methods are commonly used to enhance resistance of wood surfaces against weathering. The first one is the application of finishes to the surface in the form of thin layers or coatings with limited penetration. Numerous methods including pressurized and non-pressurized applications of chemicals, such as arsenic-based and others are also widely employed to protect wood against weathering. In certain applications, both treatment and finishing can also be used for enhanced weathering resistance of wood (Williams 2005; Jacques 2000).

Several research projects have investigated the application of inorganic chemicals to the surface of treated wood, including copper ethanolamine (Cu-MEA) to enhance its resistance against weathering (Zhang *et al.* 2009). It was determined that a Cu-MEA treatment delays the degradation of lignin due to weathering. It was also found that the discoloration of wood decreased with increasing concentration of copper in the chemical (Williams 2005). According to findings of a past study, the photo-stabilization of pressure-treated wood using chromated copper arsenate (CCA), ammonium copper quat (ACQ1900), and linseed oil also decreased delignification, resulting in a reduction in wood discoloration (Temiz *et al.* 2007). The treatment of solid wood with a melamine-based resin was considered for possible protection of wood products against weathering without changing their natural appearance (Hansmann *et al.* 2006). However, it has some limitation due to its high cost. Another method is the application of clear-coats, which is the easiest and most common method for protecting wood against weathering degradation. Usually coating wood with water-borne transparent acrylic finishes is not very effective against photo-discoloration. However, wood products coated with the semitransparent acrylic was found effective against photo-degradation, which might be due to having a pigment content restricting transmittance of UV light to the wood surface (Schaller and Rogez 2007; Allen *et al.* 2002). Therefore, it is important to reduce the light energy reaching the wood surface with use of a coating to prevent its photo-degradation (Deka and Petrič 2008).

In the last decade, organic and inorganic UV absorbers have received great attention in transparent wood coatings because of their excellent properties as UV blockers. It is known that there is no significant difference between acrylic clear-coats containing the organic and inorganic UV absorbers for stabilizing wood color and protecting the quality of the surface. However, after the exposure of the coated wood to weathering, the color stabilization and quality of both the acrylic clear-coat surfaces showed better quality than that of the control samples (Özgenç *et al.* 2010). The UVA of hydroxyphenyl-s-triazines type (HPT), which combines a high photo-permanence and a high extinction coefficient with very low volatility, provides an outstanding color stabilization potential for high-performance coatings (Schaller *et al.* 2008).

Although the weathering of wood has been investigated extensively, there is very little information on the resistance of pressure-treated wood with coated clear acrylic finishes containing an organic UV absorber and UV screener TiO₂. Therefore, the objective of this study was to evaluate the weathering behavior of treated samples of two

widely used species, namely European beech (*Fagus sylvatica*) and Scots pine (*Pinus sylvestris*), finished with an acrylic-based organic UV absorber to get initial data and a better understanding of their behavior.

EXPERIMENTAL

Preparation Samples and Treatment Process

Defect-free samples with dimensions of 105 mm in length by 75 mm wide by 5 mm thick from the sapwood of two species, European beech (*Fagus sylvatica*) and Scotch pine (*Pinus sylvestris*), were conditioned in a climate room having a temperature of 21 °C and relative humidity of 65% until they reached a moisture content of about 12%. Later, the samples were lightly sanded with 120 grit sandpaper.

Two types of water-based wood preservatives were used to treat the samples. **Micronized copper wood preservative** (Celcure MC-850) is a water-based wood preservative that contains a copper-based fungicide and an organic co-biocide (a quaternary ammonium compound). It was supplied from the Osmose Company. **Copper-based wood preservative** (Adolit KD 5) is a water-based wood preservative which contains 20.53% copper (II) hydroxidecarbonate, 10.0% didecylpolyoxethylammonium borate, and 8.0% boric acid. It was supplied from the Remmers Company. Four samples from each species were treated with each type of chemical, employing a full-cell method in a fabrication type tank. The treatment schedule consisted of an initial vacuum of 600 mm Hg for 30 min followed by a pressure of 12 bars for 60 min before samples were removed from the tank. All of the specimens were lightly wiped to remove any excess solution from their surface and weighed at an accuracy of 0.01g to determine the retention value of the chemical in the samples based following equation (AWPA U1-09 2009).

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

In Eq. 1, *G* is the amount of the treating solution absorbed by the sample based on the initial and final weight of each block in grams; *C* is the preservative solution in 100 g of the treating solution; and *V* is the volume of the sample. After the samples were treated with chemicals, they were wrapped in plastic bags for one week at room temperature to make sure that complete fixation of the chemicals was achieved. Table 1 displays retention values of the samples treated with each type of chemical.

Table 1. Retention of Wood Preservatives

Wood Preservatives	Retention (kg/m ³)	
	Beech	Pine
Adolit KD 5 (2%)	6.9 (0.68)	5.9 (0.19)
Celcure MC-850 (3%)	17.6 (0.29)	21.6 (0.49)

*Values in parentheses are standard deviations.

Coating and Artificial Weathering Processes of the Samples

The water-based impregnation agent, having active ingredients of 1.20% propiconazol, and 0.30% iodopropynyl butylcarbamate, was used as a primer for the protection of the samples against biological deterioration, including soft rot and blue stain. The primer was applied to the samples at a spread of 120 g/m² using a brush. Two types of absorbers, the UV screener TiO₂ as an *inorganic UV absorber* and the UVA of hydroxyphenyl-s-triazines class as an *organic absorber*, were used. Commercially produced finishing, having acrylic resin, a copolymer dispersion of methylacrylate/methylmethacrylate/butylacrylate, was used as a topcoat for the specimens. A small amount of defoamer and 2,2,4-trimethyl-1,3-pentandiolemonoisobutyrate, texanol as a coalescing agent was added in the topcoat formulation to reduce the effect of other additives on the photostabilization performance. Three layers of topcoats were also applied to each sample at a spread rate of 100 g/m² by brush. Later, the specimens were sanded with a 240 grit size of sandpaper and kept in room temperature for two days before applying the second layer of topcoat. Table 2 displays the code of wood samples prepared for each variation.

Table 2. Wood Samples Prepared for each Variation

Code	Applied Methods	Amount
Control	Untreated control samples	4
UV1	Applied clear coating containing UV1*	4
UV2	Applied clear coating containing UV2*	4
AQ	Treated with AQ*	4
MQ	Treated with MQ*	4
MUV1	Applied clear coating containing UV1 after treated with MQ*	4
MUV2	Applied clear coating containing UV2 after treated with MQ*	4
AUV1	Applied clear coating containing UV1 after treated with AQ*	4
AUV2	Applied clear coating containing UV2 after treated with AQ*	4
*UV1: Tinuvin 477 DW, *UV2: Hombitec 402 WP, *AQ: Adolit KD 5, *MQ: Celcure MC-850		

Formulation of the Coating Systems

These UV absorbers are synthesized by the Ciba Company (which is now part of BASF) and the Sachtleben Company. Topcoats were formulated from the same commercial acrylic resin, a poly-(methylacrylate/methylmethacrylate/butylacrylate) copolymer dispersion. To exclude effects of other additives on the photostabilization performance, only a small amount of defoamer and 2,2,4-trimethyl-1,3-pentandiolemonoisobutyrate (texanol) as coalescing agent were used in the formulations (see Table 3 and 4).

Table 3. Ingredients of Clear Coat Containing UV Screener TiO₂ Prepared

No	Ingredient	p.b.w*	Type	Supplier
1	Neocryl XK 90	73.7	Binder	DSM NeoResins
2	Demi water	20.93		
3	Texanol	0.67	Coalescent	Eastman Chemical
4	Dehydran 1293	1.0	Defoamer	Cognis
5	Coatex BR 100 P	0.6	Thickener	Coatex
6	Rheolate FX 1070	1.3	Thickener	Elementis Specialties
7	Inorganic UV Absorber-TiO ₂	1.8	UV Absorber	Ciba
		100.0		
*Percent by weight				

Table 4. Ingredients of Clear Coat Containing Organic UV Absorber Prepared

No	Ingredient	p.b.w*	Type	Supplier
1	Neocryl XK 90	73.7	Binder	DSM NeoResins
2	Demi water	20.93		
3	Texanol	0.67	Coalescent	Eastman Chemical
4	Dehydran 1293	1.0	Defoamer	Cognis
5	Coatex BR 100 P	0.6	Thickener	Coatex
6	Rheolate FX 1070	1.3	Thickener	Elementis Specialties
7	Organic UV Absorber-HPT	0.9	UV Absorber	Sachtleben
		100.0		

*Percent by weight

Artificial weathering of the samples was performed in a QUV/spray accelerated weathering test unit manufactured by Q-Panel Lab Products, Cleveland, USA. The unit is equipped with UV-A lamps (W/m^2 at 340 nm) maintaining the constant temperature of 60°C in the chamber during the test. The samples were exposed to weathering cycles of UV-light irradiation for 2 hours followed by water spraying for 18 minutes (7 liters per minute) for 28 days (ASTM G53-96 1988).

Color Measurement and Microscopic Analysis of the Samples

Twenty color measurements of each sample were periodically carried out with a Codec 400 Vis spectrophotometer manufactured by Phyma, Gaaden, Austria. The reflection spectrum for each evaluation was developed by measuring a spot of 20 mm in the 400 to 700 nm regions.

Color changes during weathering were constantly monitored at the same location of the specimens based on the method of the Commission International de l'Eclairage (CIE) using color parameters, L^* , lightness, a^* , along the X axis red (+) to green (-), and b^* along the Y axis yellow (+) to blue (-). Color differences ΔE^* of the samples were evaluated using the software Primus 2.0, developed by Phyma, Gaaden, Austria and the equation below,

$$\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \quad (2)$$

Positive values of Δb^* indicated an increment of yellow color, while negative values indicated an increase of blue color. Positive values of Δa^* resulted in a tendency of the wood surface to be reddish, while negative values revealed a tendency to be greenish. The negative ΔL^* values probably occurred due to the fact that the wood surface becomes rougher and darker during UV light irradiation. Measurements were performed at the end of a UV irradiation step to achieve a consistent condition of the samples during the color measurements (ISO 7724-2 1984).

Dry film thicknesses of the coated panel samples were also measured according to ÖNORM B 3803 by employing a SZH Olympus light microscope. Ten measurements were performed on the cross section of the samples to evaluate the film thickness of the coating.

RESULTS AND DISCUSSION

Color Change of the Samples

Table 5 displays the color changes of the European beech and Scots pine samples in the form of CIELAB parameters, namely L^* , a^* , b^* , and ΔE^* after they were exposed to 336 and 672 hours of weathering. It was found that Δa^* and Δb^* were reduced with an increasing exposure time in an accelerated test cycle for the control samples from both species.

The highest average value of ΔL^* was determined for untreated samples. It was also found that ΔL^* was the most sensitive parameter to describe the surface quality of the samples as results of irradiation and water spray. It appears that the chromaticity coordinates, Δa^* and Δb^* were less influenced by the water spraying during the tests (Temiz *et al.* 2005).

The values of ΔE^* were substantially high as a result of 72 hours of exposure for all types of the samples, suggesting that the color change of the samples took place during the initial stage of weathering. Each species showed different color changes. However, in general, softwood specimens had more rapid discoloration within the first 24 hours of irradiation, while that was during the first 12 hours for the hardwood samples (Oltean *et al.* 2008). It is a well known fact that the color change of the wood surface is not only a visual defect but also reflects the chemical modification of the components in wood due to photo-degradation. The color change is attributed to the carbonyl groups of conjugated ketones, aldehydes, and quinines resulting from the modification of lignin and other related compounds (Temiz *et al.* 2005; Pandey 2005). The highest average value of ΔE^* was found for untreated control samples for both species as a result of 336 h and 672 h of weathering time. It appears that micronized copper and copper-based wood preservatives containing copper treatments enhanced the color stabilization of the specimens compared to that of the untreated control samples. The copper modifies the components in the cell wall so that the wood surface can have higher resistance to color change, resulting in an increased roughness of the wood surface. Wood ion complexes are also formed on the wood surfaces, providing a possible higher resistance to the wood surface by blocking the free phenolic groups, which are the reactive sites of photochemical reactions (Grelier *et al.* 2000).

Findings in this work agree with those reported by previous studies stating that copper-based treatment agents such as CCA, ACQ, DDAC, and ACC reduce the photo-degradation rate by retarding the formation of carbonyl groups as a result of weathering (Jin *et al.* 1991; Liu *et al.* 1994).

It was also reported that ACQ treatment retards the progressive photo-oxidation of lignin compared to the photo-oxidation in untreated control samples (Jin *et al.* 1991). Table 5 shows that during exposure of the acrylic clear-coats containing organic and inorganic UV absorber, photostabilization performance was better than untreated control samples and treated samples. The color changes, ΔE^* , generally decreased with increasing weathering times in accelerated test cycles for control of both species of samples, except for clear-coats containing a UV absorber applied to samples.

The results of various studies showed that the clear-coat containing UV absorber substantially prevented the color changes of the wood surface during weathering (Williams and Brown 1993; Liu *et al.* 1994; Forsthuber and Gröll 2010). It also has been reported that the acrylic resin provides a good performance, demonstrating that a resin

coat that contains a UV absorber is an adequate method for the protection of wood for exterior applications (Decker *et al.* 2004; Custódio and Eusébio 2006). However, water-borne coatings present the problem of the extractive bleeding in the case of pine because of the presence of a resin that has water-soluble phenolic or polyphenolic compounds (Dawson *et al.* 2008). The typical anatomical structure of softwoods and hardwoods shows distinct differences. These structural differences of wood are the most important factors that affect weathering characteristics (Williams 2005). Thus, the results of the performance of a UV absorber on Scots pine sapwood photostabilization were different than that of European beech sapwood samples, which do not have any extractives. In Scots pine, the UV absorbers performed better than pretreatment with preservatives prior to coating; the opposite trend was observed for beech.

The comparison of the efficiency of the difference in the UV screeners TiO₂ and the organic UV absorbers in the prevention of photo-induced discoloration of the coated wooden substrate showed that TiO₂ results in the least discoloration of the coated wood panels (Forsthuber and Grüll 2010), as displayed in Table 5.

The negative lightness stability (ΔL^*) values occur during weathering because the surface becomes darker, which is opposite of the positive lightness stability (ΔL^*) values. In the early stages of weathering, dark woods tend to become light and light woods, dark. Eventually, all woods become gray if fully exposed to UV light and water spray. As can be seen in Table 5, ΔL^* values of some samples were negative after 336 h exposure, but values became positive after 672 h exposure.

The color of wood exposed outdoors is affected very rapidly. Generally, all species exhibit discoloration toward yellow or brown shades due to the chemical exposure, photo-oxidation of lignin, and wood extractives. This yellowing or browning occurs after only several months of exposure in sunny, warm climates. In the absence of micro-organisms, wood can weather to a soft silver gray color as a result of the leaching of the decomposition products of lignin (Dawson *et al.* 2008). The influence of the species on the member's characteristics are related to the content of extractives, and results of UV absorber performances on pine wood photostabilization was found to be different than that from beech samples (Aloui *et al.* 2007). A comparison with the color change results of pine samples showed good correlation between the chromospheres formation and the discoloration of the surface of the samples. It seems that the recent introduction of the lignin stabilizer concept would be one of the alternative ways to overcome this problem. To verify the experience based on the findings that lignin stabilization improved the color stability, the filter experiment was repeated in the presence of a lignin stabilizer pretreatment (Schaller *et al.* 2008).

The color change parameters (ΔL^* , Δa^* , Δb^* , and ΔE^*) of all the samples versus the QUV exposure time are presented for Scots pine sapwood (*Pinus sylvestris*), as illustrated in Fig. 1.

The color change rate of the surface of the samples due to QUV light exposure was initially fast, and then the rate of change diminished to almost zero during the 168 hours of exposure. ΔE^* varied from 0 to almost 10 units. Pine control samples had the highest discoloration as a result of 672 h weathering with distinction from the other types of samples.

Table 5. Color Change Values of Wood Surface after Artificial Weathering Test (672 hours)

Wood Species	Samples	Color Parameters							
		336 hours				672 hours			
		* ΔL	* Δa	* Δb	* ΔE	* ΔL	* Δa	* Δb	* ΔE
<i>Fagus sylvatica</i>	Control	-16.9 (2.6)	-6.1 (0.9)	-9.7 (1.9)	20.4 (1.6)	-22.7 (1.3)	-7.8 (1.9)	-12.2 (2.2)	26.9 (3.3)
	AQ	10.6 (1.2)	4.3 (0.8)	6.1 (1.2)	13.0 (1.0)	52.7 (2.1)	2.6 (0.5)	7.5 (1.5)	12.5 (2.2)
	MQ	16.2 (1.9)	4.8 (1.0)	5.4 (1.6)	17.7 (1.7)	42.6 (2.0)	3.7 (1.0)	10.1 (1.7)	16.6 (2.4)
	UV1	-2.0 (1.1)	-0.8 (1.3)	-0.2 (0.7)	2.0 (0.9)	0.3 (0.9)	-1.9 (0.8)	-0.6 (0.4)	2.0 (2.7)
	UV2	-1.4 (1.4)	-1.3 (0.6)	-1.0 (0.3)	1.3 (0.8)	1.1 (0.7)	-2.8 (0.9)	-1.3 (0.9)	1.9 (1.3)
	MU1	-3.5 (1.7)	3.1 (0.5)	-1.7 (0.6)	5.0 (1.3)	-6.1 (1.2)	3.8 (1.1)	-3.4 (1.1)	8.1 (2.8)
	MU2	-3.8 (1.3)	2.1 (0.7)	-2.5 (0.4)	5.0 (1.2)	-5.0 (1.6)	3.2 (1.0)	-3.3 (1.3)	8.8 (1.9)
	AU1	-4.5 (1.0)	1.7 (1.0)	-3.2 (1.0)	5.8 (1.1)	-6.8 (1.3)	1.9 (0.4)	-5.2 (1.7)	8.8 (1.4)
	AU2	-5.2 (1.2)	2.6 (1.1)	-0.6 (0.2)	5.8 (1.0)	-7.3 (1.1)	3.3(1.2)	-1.9 (0.8)	8.3 (3.0)
<i>Pinus sylvestris</i>	Control	-26.2 (3.9)	-3.0 (0.8)	-8.0 (2.3)	27.6 (2.1)	-22.1 (3.2)	-3.1 (1.9)	-8.8 (2.9)	24.9 (3.2)
	AQ	16.7 (2.8)	7.5 (1.7)	5.5 (2.1)	19.1 (2.7)	57.1 (5.2)	5.1 (3.2)	14.6 (1.4)	22.1 (2.1)
	MQ	22.3 (3.1)	6.3 (1.2)	0.2 (0.8)	23.2 (1.6)	48.5 (4.3)	4.6 (2.6)	13.7 (1.1)	23.9 (3.9)
	UV1	-6.4 (1.7)	5.7 (1.6)	-0.7 (0.9)	8.6 (1.2)	-8.0 (2.1)	7.4 (2.1)	-0.6 (0.4)	10.9 (2.1)
	UV2	-6.2 (1.3)	5.0 (1.1)	-1.5 (1.1)	8.1 (1.6)	-7.0 (2.3)	5.8 (1.8)	-0.8 (1.4)	9.1 (1.1)
	MU1	2.9 (1.1)	-4.0 (0.9)	0.1 (0.9)	5.0 (1.0)	2.8 (1.7)	-5.3 (1.1)	0.9 (1.1)	6.0 (1.6)
	MU2	2.0 (0.8)	-2.2 (0.6)	0.3 (0.7)	3.0 (1.7)	1.9 (1.1)	-3.1 (1.4)	1.6 (1.2)	3.9 (1.3)
	AU1	3.6 (1.5)	-5.2 (1.4)	1.2 (0.9)	6.4 (2.4)	3.6 (0.5)	-6.6 (0.6)	1.1 (0.8)	7.5 (2.1)
	AU2	2.9 (1.0)	-4.1 (1.1)	1.5 (1.1)	3.1 (1.1)	2.8 (1.2)	-5.1 (1.5)	0.1 (0.7)	5.9 (1.6)

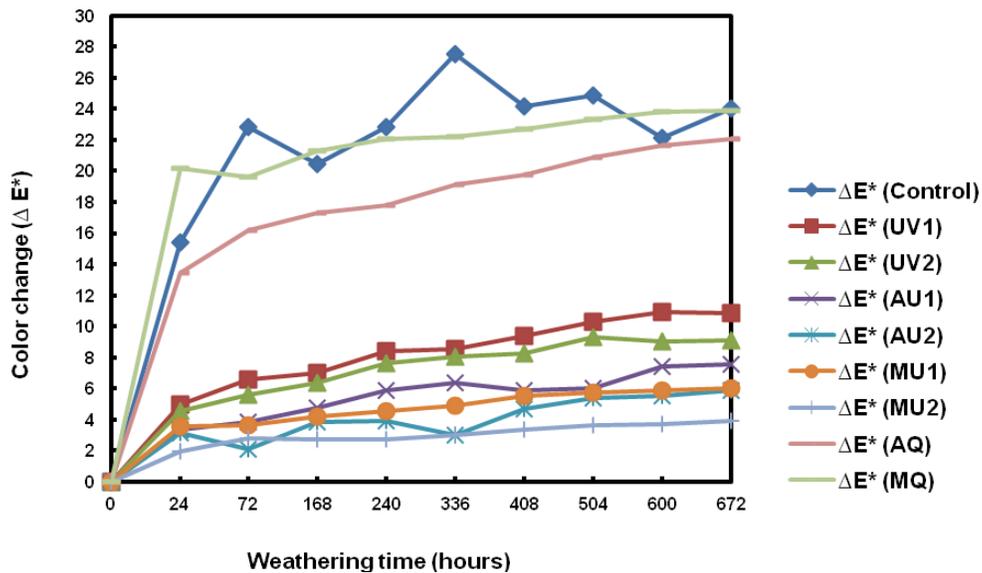


Fig. 1. Color change of weathered Scots pine (*Pinus sylvestris*) samples

The highest color change (ΔE^*) was determined for the samples treated with micronized copper and copper-based preservatives, while the lowest color change (ΔE^*) was found for samples treated with wood-coating containing UV absorbers. When comparing the efficiency of the different UV absorbers and treatments in the prevention of the photo-induced discoloration of the coated wood, clear-coats containing a UV absorber applied after treatment with micronized copper and copper-based wood preservatives resulted in the least discoloration for pine samples, as illustrated in Fig. 1.

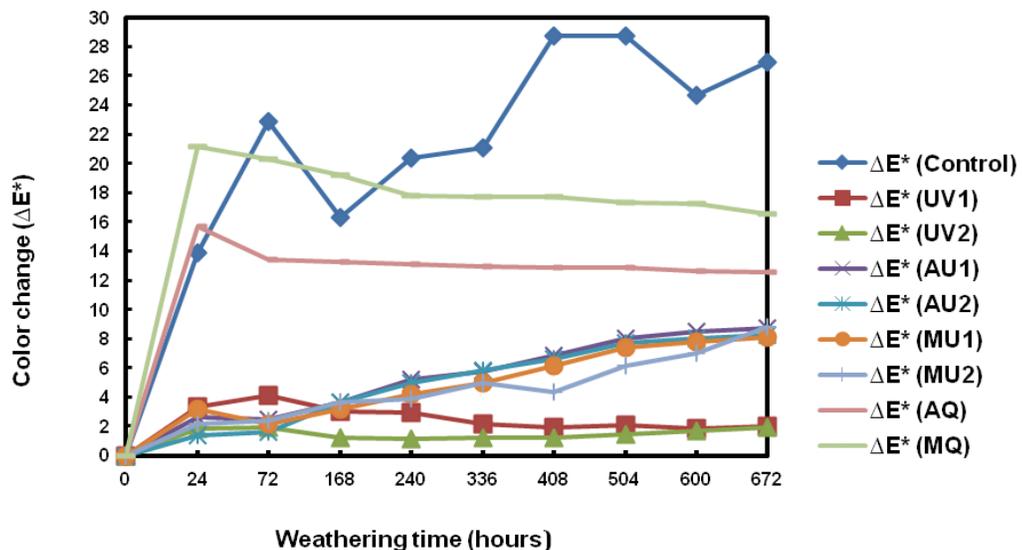


Fig. 2. Color change of weathered European beech (*Fagus sylvatica*) samples

The color change parameters (ΔL^* , Δa^* , Δb^* , and ΔE^*) of European beech (*Fagus sylvatica*) sapwood samples versus the QUV exposure are illustrated in Fig. 2. The specimens treated with micronized copper and copper-based wood preservatives for 672 h of weathering somehow had a similar color to those of the control samples. However, the ΔE of the treated samples was lower than that of the control samples. The lowest color change (ΔE^*) was determined for the untreated wood-coated having UV absorbers. It was observed that during the 672 h UV exposure, color changes showed a progressive increase when powerful UV absorbers, such as an organic UV absorber and UV screener TiO_2 , were used. On the other hand, for the control samples and those finished without using the UV absorber, the samples showed an increased color change during 336 h exposure time.

Coating Penetration of the Samples

Generally penetration of preservatives in Scots pine (softwood) is expected to be higher than beech (a hardwood having vessel elements). But the depth of the coating penetration was greater in beech than pine according to the results of this study. The extractives in pine wood and swelling of the cell wall due to solvent ingress have an important influence on penetration (Bulcke *et al.* 2008). Based on the microscopic evaluation, it was observed that the treated pine samples had a higher coating penetration than those of untreated samples. However, this was exactly opposite in the case of beech samples. Moreover, treatment with wood preservatives did not significantly affect the drying rates of the finishes on the wood when compared to the untreated wood samples. The color change of the untreated pine wood was higher than that of the treated pine samples. This could be due to lower penetration of the coating chemical in the untreated pine specimens. The coating did not penetrate deeply into the pine samples because penetration was confined to the exposed lumens of the tracheids in the outermost cell layer, as shown in Fig. 3. However, the ray cells exhibited deep penetration. In comparison, in both beech samples coated with UV screeners TiO_2 and organic UV absorbers, the ray cells had deeper penetration, as illustrated in Figs. 3 and 4.

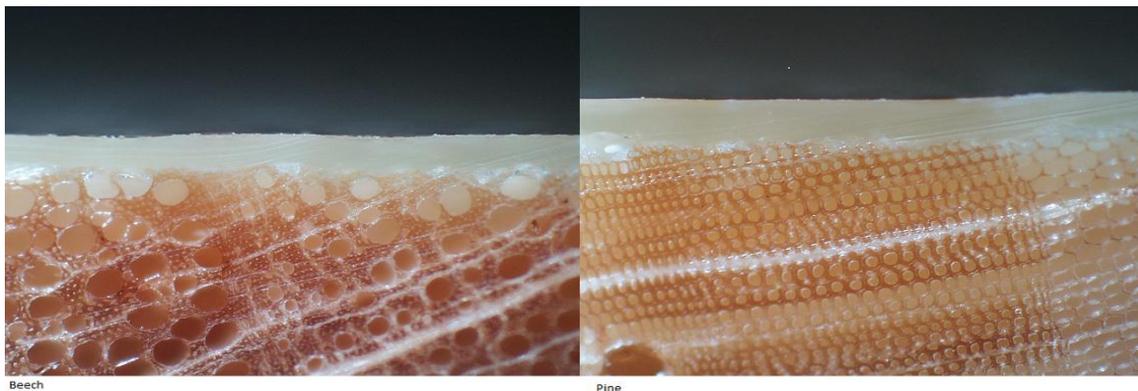


Fig. 3. Light micrograph of transverse section through beech and pine wood applied clear-coats containing UV screeners TiO_2

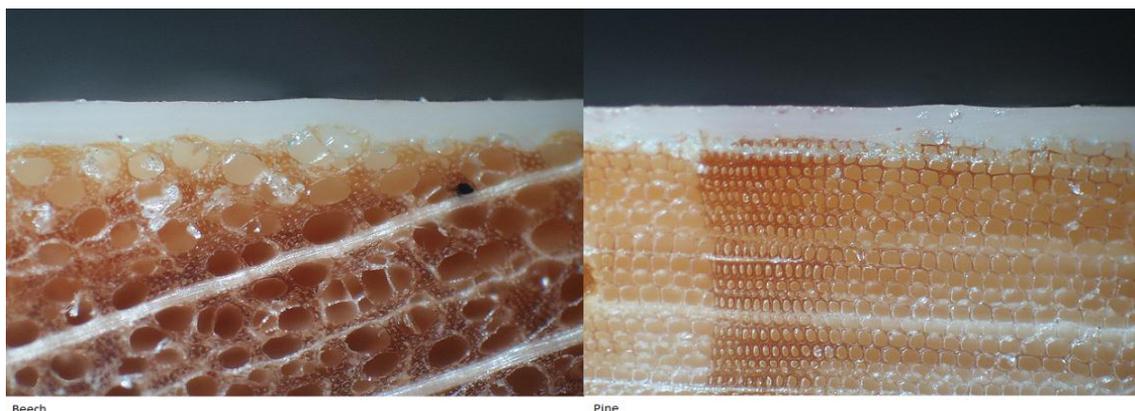


Fig. 4. Light micrograph of transverse section through beech and pine wood applied clear-coats containing organic UV absorber

The depth of the coating penetration was similar for both beech samples to which clear-coats containing UV screeners TiO_2 and organic UV absorbers were applied. The depth of the penetration in beech woods was more uniform and confined to 2 to 3 outer cell layers. The rays penetrated well in all samples, including both pine samples having clear-coats, UV screeners TiO_2 , and organic UV absorbers. It is apparent from the micrographs that the depth of the coating penetration was greater in the beech samples as compared to that of the pine specimens. The reason for the differences of the depth of the penetration in pine compared to beech could be due to the anatomical structure of the species.

The coating thickness was reduced when the pine wood was pretreated with increasing amounts of micronized copper and copper-based preservatives. However, during sectioning, the coating tended to detach in places from the surface of the pretreated beech samples, suggesting that despite increased porosity, the coating did not interlock into the surface sufficiently for the wood-coating interface to overcome the mechanical stresses generated during sectioning.

The tracheids in the surface layers of the pretreated beech woods tended to separate in the region of the middle lamella, and the cells of this region would be one distinct point of weakness if the coating did not penetrate sufficiently and uniformly enough to stabilize this region (Dawson *et al.* 2008).

The wood surfaces exposed to UV irradiation and water spray had several checks, splits, and cracks. Visual evaluation was carried out to evaluate the cracks caused by weathering. The system used to quantify the size of the cracks is described in ISO 4628. The coated sample showed no cracks, but had basic changes due to weathering. However, surface cracks were found on both of the control samples and treated samples. Pine control samples had the highest numbers of cracks identified as class 5, while beech control samples were within class 3. The treated samples had no big cracks and substantial modification on their surface was observed as class 2.

CONCLUSIONS

1. The most effective protection for stabilizing wood color involved pretreated wood coated with a UV absorber in the case of pine, and untreated wood coated with a UV absorber in the case of beech.
2. The results showed that the influence of the coating containing UV screeners TiO₂ and organic UV absorber (UVA of hydroxyphenyl-s-triazines type) is similar with small differences.
3. The coating penetration increased with higher coating absorbency for pretreated surfaces for pine. The organic UV absorber represents an outstanding stabilization potential for high-performance coatings.
4. Other methods provided better protection against color changes relative to untreated control samples. As some cracks were observed on the surface of the control samples and treated wood, UV absorbers prevented the cracking of the clear-coat surfaces during weathering.

ACKNOWLEDGMENTS

We would like to thank Holzforschung, Austria for having provided us with research material, and University of Natural Resources and Applied Life Sciences (BOKU), Institute of Wood Science and Technology for laboratory support.

REFERENCES CITED

- Allen, N. S., Edge, M., Ortega, A., Liauw, C. M., Stratton, J., and McIntyre, R. B. (2002). "Behaviour of nanoparticle (ultrafine) titanium dioxide pigments and stabilizers on the photooxidative stability of water based acrylic and isocyanate based acrylic coatings," *Polym. Degr. Stabil.* 78, 467-478.
- Aloui, F., Ahajji, A., Irmouli, Y., George, B., Charrier, B., and Merlin, A. (2007). "Inorganic UV absorbers for the photostabilisation of wood-clearcoating systems: Comparison with organic UV absorbers," *Appl. Surf. Sci.* 253(8), 3737-3745.
- AWPA (2009). "Use category system: User specification for treated wood," *American Wood Protection Association Standard*, AWPA U1-09.
- ASTM (1996). "Standard practice for operating light--- and water- exposure apparatus (Fluorescent UV-Condensation Type) for exposure of non-metallic materials," ASTM G 53-96, *Annual Book of Standards of AWPA standards*, West Conshohocken, PA.
- Bulcke, J. V., Acker, J. V., and Stevens, M. (2008). "Experimental and theoretical behavior of exterior wood coatings subjected to artificial weathering," *J. Coat. Technol. Res.* 5(2), 221-231.
- Custódio, J. E. P., and Eusébio, M. I. (2006). "Waterborne acrylic varnishes durability on wood surfaces for exterior exposure," *Progr. in Org. Coat.* 56, 59-67.

- Dawson, B. S. W., Singh, A. P., Kroese, H. W., Schwitzer, M. A., Gallagher, S., Riddiough, S. J., and Wu S. (2008). "Enhancing exterior performance of clear coatings through photostabilization of wood. Part 2: Coating and weathering performance," *J. Coat. Technol. Res.* 5(2), 207-219.
- Decker, C., Masson, F., and Schwalm, R. (2004). "Weathering resistance of waterbased UV-cured polyurethane-acrylate coatings," *Polym. Degr. Stabil.* 83, 309-320.
- Deka, M., and Petrič, M. (2008). "Photo-degradation of water borne acrylic coated modified and non-modified wood during artificial light exposure," *BioResources* 3(2), 346-362.
- Feist, W. C., and Hon, D. N.-S. (1984). "Chemistry of weathering and protection," In: Rowell, R. M. (ed.) *The Chemistry of Solid Wood*, Advances in Chemistry Series No. 207, American Chemical Society, Washington, DC, USA, Chapter 11, pp. 401-451.
- Forsthuber, B., and Grüll, G. (2010). "The effects of HALS in the prevention of photo-degradation of acrylic clear topcoats and wooden surfaces," *Polym. Degr. Stabil.* 95(5), 746-755.
- Grelier, S., Castellan, A., and Kamdem, D. P. (2000). "Photoprotection of copper-amine-treated pine," *Wood and Fiber Science* 32(2), 196-202.
- Hansmann, C., Deka, M., Wimmer, R., and Gindl, W. (2006). "Artificial weathering of wood surfaces modified by melamine formaldehyde resins," *Holz Roh Werkst.* 64(3), 168-203.
- ISO 4628-4 (1982). "Paints and varnishes-evaluation of degradation of paint coatings-designation of intensity, quantity and size of common types of defect. Part 4: Designation of degree of cracking," ISO Standard.
- ISO 7724-2 (1984). "Paints and varnishes - Colorimetry. Part 2: Color measurement," ISO Standard.
- Jacques, L. F. E. (2000). "Accelerated and outdoor/natural exposure testing of coatings," *Prog. Polym. Sci.* 25(9), 1337-1362.
- Jin, L., Archer, K., and Preston, A. (1991). "Surface characteristics of wood treated with various AAC, ACQ and CCA formulations after weathering," International Research Group on Wood Protection Documents, 22. Annual meeting, Kyoto-Japan, IRG/WP/2369.
- Liu, R., Ruddick, J. N. R., and Jin, L. (1994). "The influence of copper (II) chemicals on the weathering of treated wood, Part I. ACQ treatment of wood on weathering," International Research Group on Wood Protection Documents, 23. Annual meeting, Nusa Dua-Bali, IRG/WP/94-30040.
- Özgenç, Ö., Forsthuber, B., Teischinger, A., and Hansmann, C. (2010). "Comparison of colour change in wood clear-coating systems including inorganic and organic UV absorbers for exterior use," International Research Group on Wood Protection Documents, 41. Annual meeting, Biarritz-France, IRG/WP/10-40499.
- Oltean, L., Teischinger, A., and Hansmann, C. (2008). "Wood surface discolouration due to simulated indoor sunlight exposure," *Holz Roh Werkst.* 66(1), 51-56.
- ÖNORM B 3803. (2002). "Holzschutz im Hochbau – Beschichtungen auf maßhaltigen Außenbauteilen aus Holz durch holzverarbeitende Betriebe."

- Pandey, K. K. (2005). "Study of the effect of photo-irradiation on the surface chemistry of wood," *Polym. Degr. Stabil.* 90(1), 9-20.
- Rowell, M., and Barbour, R. J. (1988). "Archaeological wood: Properties, chemistry and preservation," In: Proceedings of 196th meeting of the American Chemical Society, Washington, Chapter 11.
- Schaller, C., and Rogez, D. (2007). "New approaches in wood coating stabilization," *J. Coat. Technol. Res.* 4(4), 401-409.
- Schaller, C., Rogez, D., and Braig A. (2008). "Hydroxyphenyl-s-triazines: Advanced multipurpose UV-absorbers for coatings," *J. Coat. Technol. Res.* 5(1), 25-31.
- Temiz, A., Yildiz, U. C., Aydın, I., Eikenes, M., Alfredsen, G., and Çolakoğlu, G. (2005). "Surface roughness and color characteristics of wood treated with preservatives after accelerated weathering test," *Appl. Surf. Sci.* 250(1-4), 35-42.
- Temiz, A., Terziev, N., Eikenes, M., and Hafren, J. (2007). "Effect of accelerated weathering on surface chemistry of modified wood," *Appl. Surf. Sci.* 253(12), 5355-5362.
- Williams, G. R., and Brown, J. (1993). "Natural exposure weathering test: Their role in the assessment of wood preservative efficacy," International Research Group on Wood Protection Documents, 24. Annual meeting, Orlando-Florida, IRG/WP/93-20006.
- Williams, R.S. (2005). "Handbook of wood chemistry and wood composites," In: *Weathering of Wood*, Forest Service, Forest Products Laboratory, USDA, Chapter 7.
- Zhang, J., Kandem, D. P., and Temiz, A. (2009). "Weathering of copper-amine treated wood," *Appl. Surf. Sci.* 256(3), 842-846.

Article submitted: June 12, 2012; Peer review completed: July 15, 2012; Revised version received and accepted: August 9, 2012; Published: August 16, 2012.