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COMPARISON OF DURABILITY OF WOOD COATINGS CONTAINING DIFFERENT WATERBORNE ACRYLIC RESINS AND UV ABSORBERS IN NATURAL WEATHERING

In this study, pine and beech sapwood samples coated with 12 different waterborne acrylic resin coating systems were exposed to natural weathering in Trabzon, Turkey. The natural weathering test continued for 18 months. In these coating systems, water in different proportions, boric acid, two different acrylic resins and three different UV absorbers supplied by BASF were used. The aim was to compare the durability of 12 different coating systems in natural weathering in terms of colour change, surface roughness and macroscopic evaluation. These test methods were used to evaluate the appearance and physical properties of the coatings after the natural weathering test. The results led to the selection of the best coating formulation for wood durability in natural outdoor conditions. The appearance and physical values after 18 months of the weathering test showed that boric acid increases the durability of the coating for use in outdoor conditions. Additionally, the coating formulation containing especially acrylic resin and Tinuvin 400 DW provided the highest durability against outdoor conditions.

Keywords: colour, weathering, roughness, waterborne acrylic, coating, durability, macroscopic evaluation

Introduction

Wood surface is protected by surface treatments against outdoor conditions such as UV light, high and/or variable humidity. Wood surface protectants also improve aesthetics by providing colour or shine. The durability performance of coatings in outdoor conditions varies based on many factors such as wood types and properties, coating types and properties, application procedures and outdoor conditions. For this reason, depending on the severity of the outdoor factors, the nature and characteristics of coatings, renewal is required every few years. Surface-treated wood can withstand water vapour permeability [Custódio and Eusébio 2006; Teacă et al. 2013; Evans et al. 2015]. The use of coating systems

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containing UV absorbers (acting as filters) minimizes wood degradation due to ultraviolet radiation. New generation coatings may also play a role in resistance to biological hazards if they contain additional wood preservatives, or if wood is pre-treated with a suitable preservative prior to coating [Stirling et al. 2011]. Another requirement for coating is durability over many years. For this reason, a wood protectant coating should cover tackiness, flexibility, durability, permeability and ultimate resistance to fungi. Particularly, if coatings have excellent outdoor durability and provide extraordinary protection against biodegradation factors for wood, it is not an easy task to formulate it as a highly variable material [Forsthuber et al. 2013a; Grüll et al. 2014; Keilmann and Mai 2016].

On the wood surface in outdoor conditions, with the effect of UV rays, the lignin is firstly separated into radicals, which then cause decomposition in other chemical components of the wood. Then, the products of this degradation on the wood surface cause the adhesion between the wood surface and the coating to disappear, resulting in delamination in coating systems [Nkeuwa et al. 2014]. The development of nanotechnology allows wood coatings to develop further, or to achieve new performance characteristics. The use of nano-metal oxides of different sizes increases the scratch and abrasion resistance of UV protection on wood surface coatings. The use of nano-metals with a size of less than 100 nm provides new properties for polymeric surface materials. Transparent iron oxides are generally used as wood preservatives, but they colour the wood structure [Cristea et al. 2012; Fufa et al. 2012]. If one wants to make the colour clear, organic UV absorbents (benzophenone family, benzotriazoles, triazines, malonates) together with HALS (hindered amine light stabilizers), which exhibit synergistic photoprotection, are widely used. Organic UVA may be degraded by visible light from 500 nm to 17 nm, and UVA is ineffective at this wavelength, while harmful UV energy is converted into warming before reaching the bottom layer, whereas HALS cleans away important free radicals such as lignin. At the right doses, a combination of these provides good protection, and their protective properties are reduced during prolonged UV exposure. Inorganic UVA-like nanoparticles (typically 5-50 nm) may provide effective UV protection in the long run because they do not dissociate from the surface matter in outdoor conditions [Schaller and Rogez 2007; Forsthuber and Grüll 2010; Nguyen et al. 2016].

The most commonly used inorganic nanoparticles as UVA are oxides such as $TiO₂$, ZnO and CeO₂. These nanocrystalline oxides behave like semiconductors. Moreover, $TiO₂$ and ZnO may also exhibit photocatalytic behaviour by formation of reactive free radicals in a reaction mechanism [Schaller and Rogez 2007; Saha et al. 2013]. For this reason, photocatalytic behaviour is used in the production of self-cleaning surface materials, but in order to be effective, it is also necessary to protect the inorganic or organic inert substances of UVA nanoparticles with surface treatments. Self-cleaning surfaces are of great interest

in numerous applications. Recently, while maintaining a high level of transparency, inert formation studies based on $SiO₂$ or silanes suppressing photocatalytic activity have intensified. In the case of $CeO₂$, the electron generated after stimulation does not move away from the surface at ambient temperature [Cristea et al. 2012; Nikolic et al. 2015; Nguyen et al. 2016, 2017]. $TiO₂$ in anatase modification surface materials is a well-known photocatalyst used in the industry because of its efficiency in decomposition of contaminating organic compounds after exposure to UV radiation. TiO₂ as a photocatalyst is remarkable because it is not toxic, it is chemically inert, and it is not light and cheap [Forsthuber et al. 2013b; Chen et al. 2014; Grüll et al. 2014; Kotlίk et al. 2014].

The durability of Scots pine and Oriental beech wood surfaces coated with 12 different coating systems was evaluated in natural outdoor conditions. These coating systems consisted of 2 different acrylic resins, 3 different UV absorbers, distilled water, boric acid and some additives. The change in the coating systems exposed to outdoor conditions was determined by colour and roughness analysis. Furthermore, the change in the sample surface after the natural weathering test was determined by macroscopic evaluation.

Materials and methods

Wood materials

As the raw materials, Scots pine (*Pinus sylvestris* L., 0.49 g/cm³ dry density) and Oriental beech (*Fagus orientalis* L., 0.61 g/cm³ dry density) were selected and cut into pieces with dimensions of 350 mm in length by 70 mm wide by 20 mm thick from sapwood according to the TS EN 927-3 standard. The wood samples were conditioned in a climate room at 23 ± 2 °C and 65 ± 5 relative humidity until constant weight, and 12% moisture content was attained. One control and three test samples were cut for each variation. The cross sections of the wood samples were coated with epoxy paint to increase resistance to weathering conditions.

Coating systems

A commercial water-based impregnation product, with active ingredients of 1.20% propiconazole and 0.30% iodopropynyl butylcarbamate, was used as a primer for protection of the samples against biological deterioration, including soft rot and blue stain. The primer was applied to the samples at a spread rate of 120 g/m² using a brush. Tinuvin 400 DW was used as a UV absorber in this study. Commercially produced finishing, including acrylic resin, a copolymer dispersion of methyl acrylate/methyl methacrylate/ butyl acrylate, was used as a topcoat for the specimens. A small amount of defoamer and 2,2,4-trimethyl- -1,3-pentandiolemonoisobutyrate, a coalescent agent, was added in the topcoat

formulation to reduce the effect of the other additives on the photostabilisation performance. These formulation products were supplied by the BASF Company for the wood coatings (Table 3). Three layers of topcoats were also applied to each sample at a spread rate of 100 g/m^2 using a brush. Later, the specimens were sanded with a 240-grit size sandpaper and kept at room temperature for two days before applying the second layer of the topcoat. The characteristic features of the wood coating materials in the study are given in Tables 1 and 2.

Acrylic resin code	Detailed information				
Acrylic Resin 1	Superior weathering resistance, excellent blushing resistance, tack-free films, also for coloured aggregates				
Acrylic Resin 2	Exceptional outdoor durability and film elasticity together with outstanding water barrier properties, blocking resistance and wet adhesion				

Table 1. Acrylic resin types for wood coating systems

Table 2. UV absorbers for wood coating systems

Table 3. Formulations of wood coating systems

Natural weathering test

For the purposes of the natural weathering test, $350 \times 70 \times 20$ mm test and control wooden pieces were prepared by removing moisture to prevent decay and painting them with 2-Epoxy white paint in sections as shown by the TS EN 927-3 standard. Then, the wood samples were stored for approximately two weeks in an environment with a temperature of 20°C and relative humidity of $65 \pm 5\%$ prior to the natural weathering test.

Natural weathering test assemblies were installed on the coast at various altitudes in the province of Trabzon in the Black Sea Region of Turkey. The control and test wood samples were placed, as indicated in the TS EN 927-3 standard, in the assemblies installed in Trabzon (Turkey). The natural weathering test continued for 12 months.

Colour measurement

The reflection spectrum from a Konica Minolta CM-600d instrument was acquired from an area measuring 8 mm in diameter with 100 in the 400-700 nm wavelength range. Five measurements were recorded for each sample. The CIE (Commission Internationale de l'Eclairage) colour parameters *L** (lightness), *a** (along the X axis red $(+)$ to green $(-)$) and b^* (along the Y axis yellow $(+)$ to blue (–)) were calculated using the Konica Minolta Colour Data Software CM-S100w Spectra MagicTM NX Lite (ISO 7724-2), from which the colour differences *ΔE** were calculated according to the formula given below:

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

Surface roughness measurement

A TR100 Surface Roughness Tester was employed for measurement of surface roughness. The Ra and Rz roughness parameters were measured to evaluate the surface roughness of the un-weathered and weathered (treated and untreated) samples' surfaces according to DIN 4768 (DIN 1990). Ra is an arithmetic mean of absolute values for profile departures within the reference length, and Rz is the arithmetic mean of the 5-point height of irregularities [DIN 4768 (DIN 1990)]. The cut-off length was 2.5 mm, the sampling length was 12.5 mm, and the detecting tip radius was 5 mm for the surface roughness measurements.

Macroscopic evaluation

After the weathering test, macroscopic changes (erosion, checks and cracks) on the sample surfaces were evaluated according to the principles of the ASTM D 660, ASTM D 661 and ASTM D 662 standards. The samples were visually rated on a scale of 0-10 with 0 indicating a surface with a high level of degradation and 10 indicating a flawless surface with no degradation. Pictures of the samples were also taken with a NIKON D7500 + AF-S DX NIKKOR 18-105 VR Digital SLR camera.

Results and discussion

Colour change

The colour changes of the wood samples with coating for different exposure times are presented in Table 4 according to the CIELAB parameters, *∆L**, *∆a**, *∆b** and *∆E*. The colour changes of the wood samples were determined before the weathering test and at the end of the 1st, 4th, 6th, 9th and 12th months of the weathering test.

The colour change rates (*∆E**) were fast for formulations A and F in the beech samples until the end of the 4th month of the weathering test, as seen in Figure 1. However, the colour change rates diminished slightly for the formulations between the 4th and 9th months of the weathering test, while they decreased significantly for all formulations on the Scots pine surfaces during the weathering time (Fig. 2). The largest colour change occurred in formulation K for beech and L for pine, while the smallest colour change was found in formulations E and P for the beech and pine samples (Figs. 1 and 2).

Fig. 1. Colour changes (*E****) of beech wood surfaces treated with acrylic-based coating systems**

A greater colour change was found in the pine wood coated with all formulations than the beech wood, while the colour change parameters were also different based on the wood species. The extent of colour changes varied based on the wood species. Discoloration of the wood surface is influenced by lignin,

carbohydrate and extractives, which can be oxidized and degraded under UV irradiation [Deka and Petric 2008]. The changes in the content of wood components could explain the differentiation between wood species.

Fig. 2. Colour changes (*E****) of pine wood surfaces applied with acrylic-based coating systems**

Table 4 demonstrates the colour changes of all formulations on the Scots pine and beech samples exposed to the natural weathering test for 12 months. As seen, the colour changes (*∆E**) were quite different for all formulations.

When the *∆a** values of the beech samples were examined, it was observed that the colours tended to turn greenish for all formulations, while they turned reddish for all formulations on the pine surface during the natural weathering test. Meanwhile, the *∆b** values of the colours became bluish for all formulations that were coated on the pine and beech samples for 12 months. It is known that *∆L** is an important parameter for the determination of surface quality and overall colour changes [Ozgenc et al. 2012]. The light stability (*∆L**) of the beech samples for all formulations was negative, which pointed out that the wood surface became darker when the exposure time increased. The changes in the light stability *∆L** values tended to be negative for all formulations on the beech and pine surfaces. The negative values of *∆L** showed that the wood surface changes were darker. However, lignin polymerization and degradation of other cellulosic polysaccharides under UV radiation result in a darkened surface, which changes the *∆L** values toward negative [Deka and Petric 2008, Korkut et al. 2012].

Table 4. Colour coordinates of wood surfaces applied with acrylic-based coating systems **Table 4. Colour coordinates of wood surfaces applied with acrylic-based coating systems**

Surface roughness change

The changes in the surface roughness values of the waterborne acrylic coating systems are presented in Figure 3. As seen here, there were significant differences after the weathering test for the 12 formulations. However, the rate of change in the Ra and Rz values of formulations G for beech wood and L for pine wood was found to be quite low after the natural weathering process.

Fig. 3. Rate of changes in surface roughness values of wood surfaces treated with acrylic-based coating systems

The wood coating systems prevented wood surface degradation and provided effective preservation against intensive weathering conditions. The smoothest surface was obtained with formulations H, P and K for both pine and beech samples before the weathering test. When the performance of coatings is considered, formulations K and P provided the smoothest surfaces for both wood species after the weathering test. The highest changes in the surface roughness values were recorded from formulations D and L for both pine and beech samples after the weathering test.

The wood species affects the rate of change in the roughness values of coating systems after outdoor testing. It was determined that the change in the coating systems applied to the surface of the pine wood was rather high in comparison to the beech wood. Wood is anisotropic and composed of heterogeneous materials. As a result of this, tree species, density, moisture content and the wood's anatomical properties (diameter of vessels and tracheids, and the proportion of early and latewood) affect wood surface roughness [Kiliç et al. 2006; Csanády et al. 2015]. When wood is exposed to outdoor conditions,

UV rays, humidity, temperature and oxygen degrade wood surfaces and affect surface roughness. Surface wettability and contact angle, which are sensitive against roughness, have a significant effect on coating as well as the evaluation of coating performance [Csanády et al. 2015].

The surface roughness values of the pine and beech wood coated with 12 different formulations before and after the natural weathering test are shown in Table 5.

Beech Sample	Before Weathering		After Weathering		Pine Sample	Before Weathering		After Weathering	
Code	Ra	Rz	Ra	Rz	Code	Ra	Rz	Ra	Rz
\mathbf{A}	0.79 (0.14)	16.6 (3.87)	1.00 (0.25)	13.3 (2.79)	\mathbf{A}	1.10 (0.20)	18.9 (4.73)	1.09 (0.49)	13.0 (6.87)
B	0.52 (0.07)	18.9 (2.66)	0.40 (0.05)	6.17 (0.88)	B	0.72 (0.12)	20.9 (3.01)	0.34 (0.06)	4.89 (1.48)
\mathcal{C}	0.71 (0.08)	27.4 (3.97)	0.40 (0.05)	6.76 (0.72)	$\mathbf C$	0.92 (0.33)	23.5 (3.78)	0.58 (0.06)	7.49 (2.85)
D	0.99 (0.18)	18.8 (4.59)	1.54 (0.46)	16.4 (3.52)	$\mathbf D$	1.05 (0.22)	13.8 (4.67)	1.32 (0.28)	15.2 (4.92)
E	0.48 (0.07)	12.1 (3.02)	0.48 (0.09)	6.05 (1.81)	E	0.62 (0.16)	13.9 (3.92)	0.46 (0.46)	5.82 (0.92)
$\boldsymbol{\mathrm{F}}$	0.63 (0.32)	13.1 (4.40)	0.52 (0.05)	7.58 (0.89)	${\bf F}$	0.58 (0.08)	9.23 (2.03)	0.78 (0.89)	9.15 (6.56)
G	1.23 (0.31)	19.2 (5.90)	1.26 (0.35)	16.2 (5.89)	G	0.92 (0.35)	15.0 (4.22)	1.23 (0.48)	17.3 (6.50)
H	0.56 (0.11)	15.7 (3.76)	0.38 (0.07)	6.90 (6.90)	H	0.39 (0.04)	11.8 (3.16)	0.34 (0.10)	5.87 (1.43)
K	0.59 (0.06)	21.8 (2.75)	0.41 (0.07)	7.25 (0.84)	K	0.51 (0.09)	15.4 (15.4)	0.50 (0.50)	7.42 (1.86)
L	1.08 (0.18)	15.3 (3.16)	1.52 (0.35)	17.5 (7.12)	L	1.50 (0.62)	18.1 (4.42)	1.53 (0.16)	15.2 (4.37)
\mathbf{P}	0.67 (0.21)	14.6 (7.48)	0.50 (0.10)	6.72 (1.23)	\mathbf{P}	0.52 (0.07)	12.4 (3.97)	0.46 (0.08)	5.23 (0.78)
T	0.58 (0.13)	12.3 (2.25)	0.56 (0.07)	7.57 (1.05)	T	0.66 (0.20)	12.2 (2.95)	0.52 (0.08)	6.52 (1.22)

Table 5. Surface roughness values of wood surfaces treated with acrylic-based coating systems

*Mean values from 4 samples; Values in parentheses are standard deviations.

Macroscopic evaluation

Due to the fact that such factors as the region's altitude, climate conditions and air pollution differ in terms of deformations on wood surfaces coated with acrylic systems exposed to natural weathering tests, the visual evaluation points of the samples were also found to be different. After the 12-month weathering test, the visual evaluation performance of the beech and pine surfaces coated with 12 formulations was evaluated as to the ASTM D662-93 standard (Table 6). In comparison to the pine samples, macroscopic evaluation of the beech surfaces coated with 12 different acrylic formulations received quite high scores (Table 6).

Sample Code	Beech	Pine
А	h	6
B		
\cap		
E		
F		
G		
Н		
K		
D		
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Table 6. Macroscopic evaluation of coated wood samples after artificial weathering test

It may be seen in Table 6 that especially the wood samples coated with formulations P and K for both beech and pine received quite high visual evaluation scores. It was also seen that formulation H for pine and formulation T for beech provided very high protection after natural weathering testing. According to the results obtained from similar studies, coating treatment with water-repellent transparent acrylic resin does not provide effective protection against photo-degradation under natural weather conditions. However, a high durability performance of wood surfaces subjected to organic or inorganic UV absorbent acrylic resin coating was determined [Custόdio and Eusébio 2006; Schaller et al. 2008; Evans et al. 2008; Forsthuber et al. 2013a].

The pine and beech surfaces coated with formulations A, D, G and L were darkened. Additionally, the pine and beech surfaces coated with acrylic coating due to the air pollution outside were also dirtied. The dirty layer forming on the wood surface coated with waterborne acrylic resin containing UV absorber may be cleaned with a slightly damp cloth [Xie et al. 2008; Nejad 2011; Nejad and Cooper 2011].

The beech surfaces coated with formulations B, E and H were found to have deformations on their sides after the natural weathering test, as seen in Figure 4.

Fig. 4. Wood surfaces treated with acrylic-based coating systems

Conclusions

Wood surfaces coated with waterborne acrylic containing UV absorber or HALS provide quite good protection against degradation caused by natural weathering conditions. In this study, we evaluated the performance of 12 different

waterborne acrylic wood coating systems against natural weathering conditions. Despite the intensive weathering conditions, the wood coating systems presented a very good performance. Especially formulation P for both wood species improved colour stability and significantly prevented macroscopic deformation and changes in surface roughness. However, for both types of wood, formulation E provided very high colour stability, but it did not provide effective protection for surface roughness and macroscopic deformation. According to the macroscopic evaluation, formulations P and K completely eliminated such erosion as the formation of cracks, tears and fibre stand-up on the wood surfaces exposed to weathering conditions over 12 months. It is thought that the

durability performance of the formulation P and K acrylic systems shall be investigated on heat-treated or pre-treated wood surfaces in outdoor conditions.

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List of standards

- **ASTM D660-93:2011** Standard Test Method for Evaluating Degree of Checking of Exterior Paints, ASTM International, West Conshohocken, PA
- **ASTM D661-93:2011** Standard Test Method for Evaluating Degree of Cracking of Exterior Paints, ASTM International, West Conshohocken, PA
- **ASTM D662-93:2019** Standard Test Method for Evaluating Degree of Erosion of Exterior Paints, ASTM International, West Conshohocken, PA
- **DIN 4768:1990** Determination of values surface roughness parameters Ra, Rb, Rmax using electrical contact (stylus) institute
- **EN 927-3:2013** Paints and varnishes Coating materials and coating systems for exterior Part 3: Natural weathering test
- **ISO 7724:1984** Paints and varnishes Colorimetry

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