Effectiveness of Two Different Hydrophobic Topcoats for Increasing of Durability of Exterior Coating Systems on Oak Wood

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Abstract: Top hydrophobic layer can increase durability of exterior coatings on wood. Two hydrophobic topcoats—synthetics and water-based acrylate resin with wax additives were researched as top layer on twenty-four different coating systems applied on oak wood in this experiment. Artificial accelerated weathering lasted six weeks. Changes of color, gloss, surface wetting were evaluated, and microscopic analyses of coated surfaces were done. The results have shown that top hydrophobic layer increases durability of tested coating systems in most cases. However, the effectiveness of the two researched hydrophobic topcoats was different depending on the specific tested coating systems.

Keywords: artificial weathering; durability; exterior coatings; hydrophobic topcoat; oak wood

1. Introduction

Wood is in outdoor applications subjected to degradation caused by abiotic and biotic factors [1]. Even without ground contact (class 3 by EN 350 [2]), the rapid degradation of surface layers caused by the interaction of water and solar radiation takes place [3]. Wood with lower natural durability is more susceptible to biological attack—mainly by wood destroying fungi and insect [1]. Often used option of wood protection against above-mentioned damage is the use of coating systems [4,5].

The other possibility is also the use of hydrophobic coatings or wood hydrophobization itself [6–8]. The hydrophobic compounds generally decrease the amount of absorbed water which leads to decrease of wood swelling and shrinkage [6,9]. This limitation of dimensional changes leads to slower degradation process outdoors [10]. Additionally, hydrophobic coatings prevent wet conditions creating a very favorable environment for the growth of various wood degrading biological organisms (e.g., diverse fungi, bacteria, and insects) [9]. Common surface hydrophobization methods include, but are not limited to, treatments based on silicone [11–13] and surface impregnation with various waxes, oils, or acrylate coatings [9,14–16]. In other studies, wood surfaces were also treated by heat [17] or plasma [18] to improve water resistance. Through the introduction of nanotechnology, the different hydrophobic treatments are developing at a rapid pace [6,19]. Nanoparticles based on titanium dioxide [20], silica nanoparticles [21] or zinc oxide were applied as hydrophobic treatments to wood. Nevertheless, many of these approaches, especially the nanoparticles impact can be discussed [22], have a negative environmental impact and cause damage to the ecosystem because of the possibility for hazardous chemicals to leak from the surface [6].
Hydrophobic coatings themselves are not usually able to ensure sufficient protection of wood surfaces against weathering—they do not protect the underlying wood against solar radiation causing decomposition of lignin and extractives and their leaching connected with visual color changes [7,8,23]. Another use of hydrophobic coatings is their application as topcoat to prevent the synergistic action of solar radiation and water [24,25]. In previous studies [1–28], the efficiency of top hydrophobic coating to prolong the overall service life of coating system applied on Norway spruce and Black locust wood was proved.

It would be preferable to use oak wood (Quercus patreae, L.) in the outdoor applications as an available wood with higher durability against biotic damage (EN 350 [2]). Without surface protection, however, it rapidly changes its color in exterior [29] and leaching of extractives occurs, which can aesthetically damage other parts of construction (balcony, façade, terrace etc.). Due to complex morphological structure of oak wood with opened vessels and specific tannins content [30] is the service life of coating systems on oak wood outdoors insufficient [31]. Hydrophobic layer applied as separate coating was characterized by the lower efficiency than multilayered oil-based coating system based [8]. From these reasons, it is necessary to find a long-lasting coating system on this wood in outdoor applications.

The objective of this study is to evaluate the efficiency of transparent and semi-transparent coating systems applied on oak wood and possible enhancement of their durability by application of hydrophobic layers. The performance of different coating systems is evaluated by measuring color, gloss and surface wettability changes during artificial weathering and by confocal laser scanning microscopy.

2. Materials and Methods

2.1. Wood Material

The experiment was conducted using oak wood (Quercus patreae, L.) with mean density of $\rho_0 = 705$ kg/m$^3$ harvested in the Czech Republic. The samples in dimensions of $40 \times 20 \times 150$ mm$^3$ ($T \times R \times L$) were sanded with a grain size of 120 in a longitudinal direction and visually sorted. Prepared samples were conditioned in laboratory conditions (20 ± 2 °C and 65% RH) to achieve equilibrium moisture content of 12% before application of coatings and subsequently before each measurement.

2.2. Coating Application

The oak wood samples were treated with oil, acrylate, alkyd and synthetic coatings according to recommendation from producers given in Table 1. The coated samples were subsequently treated with two different transparent top hydrophobic coatings (b, c), which were applied by brush according to a recommendation from a producer, the remaining samples were left without hydrophobic treatment for a comparison (a), see Table 2. The cross ends of samples were sealed using silicon to prevent additional water uptake. The initial properties of prepared coating systems are given in Table 3.

| Sign | Specification | Polymer Base | Transparent x Pigmented | Number of Layers | Spreading Rate (g/m$^2$) |
|------|---------------|--------------|-------------------------|-----------------|--------------------------|
| OL-1 | Linseed oil   | OIL          | T                       | 2               | 100                      |
| OL-2 | Oil water emulsion based on modified linseed oil Based on vegetable oils and fatty acids, with carnauba and candelilla waxes, natural resins, essential oils | OIL | T | 2 | 100 |
| OL-3 | Water-based wood oil with fungicides (BIT and IPBC) | OIL | T | 2 | 80 |
| OL-4 | Natural oils (linseed, sunflower, rapeseed) with metal oxides pigments and titanium dioxide | OIL | T | 2 | 100 |
| Sign | Topcoat Specification | Polymer Base | Transparent × Semitransparent | Number of Layers | Spreading Rate (g/m²) |
|------|-----------------------|--------------|-------------------------------|------------------|----------------------|
| a    | Without hydrophobic treatment | – | – | – | – |
| b    | Synthetic lasur with fungicides (IPBC 0.3%) and UV-stabilizers | SYNTHETIC LASUR | T | 3 | 100 |
| c    | Colorless water-based protection with hydrophobic polymers, 0.1%–0.2% IPBC | ACRYLATE | T | 1 | 100 |

Note: SL-2 was both tested as a top hydrophobic layer (b, see Table 2) and also as coating system.

Table 2. Specification of applied hydrophobic topcoats.

| Coating without Hydrophobic Topcoat (a) | Hydrophobic Synthetic Topcoat (b) | Hydrophobic Water-Solvent Topcoat (c) |
|----------------------------------------|-----------------------------------|---------------------------------------|
| $L_a$ | $a_2$ | $b_2$ | $G_a$ | $CA_a$ | $L_a$ | $a_2$ | $b_2$ | $G_a$ | $CA_a$ | $L_a$ | $a_2$ | $b_2$ | $G_a$ | $CA_a$ |
| OL-1 | 51.7 | 14.6 | 29.6 | 31.1 | 102.3 | 57.6 | 12.2 | 31.1 | 24.3 | 109.9 | 59.0 | 11.0 | 30.8 | 44.8 | 99.3 |
| OL-2 | 49.6 | 11.4 | 26.6 | 2.5 | 99.5 | 58.6 | 9.8 | 28.7 | 5.2 | 111.5 | 60.5 | 8.6 | 25.5 | 4.3 | 97.1 |
| OL-3 | 58.1 | 11.8 | 29.3 | 7.0 | 107.9 | 58.2 | 11.4 | 29.3 | 4.1 | 111.5 | 57.0 | 10.4 | 28.4 | 23.7 | 107.0 |
| OL-4 | 54.7 | 12.6 | 29.1 | 1.8 | 116.2 | 55.6 | 10.7 | 29.0 | 3.6 | 112.8 | 61.5 | 10.2 | 28.5 | 3.5 | 99.4 |
subsequently calculated using Equation (1):

The observation angle of 10°, d/8 geometry and D65 light source, and the SCI method (specular of AW using Spectrophotometer CM-600d (Konica Minolta, Osaka, Japan). The device was set to an Europe, was also used in a study by Van den Bulcke [33] and Pánek [8].

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| Conditionin | Chamber | UV Irradiance | 2.5 h | Water-spray (off), UV Irradiance 1.10 W/m² at 340 nm |
|-------------|---------|----------------|-------|--------------------------------------------------|
| UV Irradiance | UV chamber | 2.5 h | UV Irradiance 1.10 W/m² at 340 nm |
| Water-spray | UV chamber | 0.5 h | Water-spray (on), UV (off) |
| Temperature Cycling | Conditionin | 1 h | Temperature Cycling | Conditionin |
| 3 | 2 | 1 h | T = +80 °C (RH = 25 %) | 1× |

2.3. Artificial Weathering (AW)

Artificial weathering was performed in a UV-chamber QUV (Q-Lab, Cleveland, OH, USA) on the basis of modified EN 927-6 [32] with the test parameters given in Table 4. During the week cycle of irradiation and spraying, the samples were transferred to a conditioning chamber Discovery My DM340 (ACS, Massa Martana, Italy) and exposed to three two-hour cycles of temperature changes from −25 to +80 °C (with 25% RH). The total weathering time consisted of 6 cycles—1008 h of weathering in UV chamber and 36 h of temperature cycling. The alternation of UV radiation, spray, and low temperature cycles, which leads to more accurate simulation of exterior conditions in Europe, was also used in a study by Van den Bulcke [33] and Pánek [8].

Table 4. One cycle of artificial weathering.

| Rank | Phase | Device | Duration | Parameters | Repetition |
|------|-------|--------|----------|------------|------------|
| 1    | Condensation | UV-chamber | 24 h | T = 45 ± 3 °C, Water-spray (off), UV (off) | 1× |
| 2    | UV Irradiance | UV chamber | 2.5 h | T = 65 ± 3 °C, Water-spray (off), UV Irradiance 1.10 W/m² at 340 nm | 48× |
| 3    | Temperature Cycling | Conditionin g chamber | 1 h | T = −25 °C | 3× |
|      | Temperature Cycling | Conditionin g chamber | 1 h | T = +80 °C (RH = 25 %) |

2.4. Color Analysis

The color parameters [34] (CIE 1986) of the test specimens were measured after 1, 3 and 6 weeks of AW using Spectrophotometer CM-600d (Konica Minolta, Osaka, Japan). The device was set to an observation angle of 10°, d/8 geometry and D65 light source, and the SCI method (specular component included) was used. Six measurements per sample were carried out for each weathering time. Color changes evaluations were done in CIE L∗a∗b∗ color space on the basis of L∗, a∗, and b∗ color coordinates, where: L∗ is lightness from 0 (black) to 100 (white); a∗ is chromaticity coordinate + (red) or − (green); b∗ is chromaticity coordinate + (yellow) or − (blue).

The relative changes in color (∆L*, ∆a*, and ∆b*) between the weathered and the initial state were determined. According to the Euclidean distance, the total color difference ∆E* (CIE 1986 [8]) was subsequently calculated using Equation (1):
\[ \Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \] (1)

2.5. Gloss Analysis

Gloss measurements were performed on the basis of EN ISO 2813 [35] using glossmeter MG268-F2 (KSJ, Quanzhou, China). Six measurements at a 60° angle per sample after 1, 3, and 6 weeks of AW were done to evaluate gloss changes.

2.6. Surface Wettability Analysis

The performance of different hydrophobic layers on oak wood surface wettability was investigated. The water contact angle on oak wood tangential surfaces was measured using a goniometer Krüss DSA 30E (Krüss, Hamburg, Germany). The sessile drop method was used to compare the wettability of different surface treatments during artificial weathering. Ten measurements per sample were performed after 1, 3, and 6 weeks of weathering with distilled water drops with a dosing volume of 5 μL. The value of contact angle was recorded in 5 s after drop deposition on the surface as in other studies [36–38]. The phenomena of spreading and absorption of water drops on the oak wood surface was investigated via variations of the weathering time, coating system and applied hydrophobic layer.

2.7. Macroscopic and Microscopic Analysis

Surfaces were regularly scanned using Canon 2520 MFP scanner with 300 DPI resolution (Canon, Tokyo, Japan) to evaluate degradation of tested coating systems. Microscopic structural changes of coatings and wood surfaces were studied using confocal laser scanning microscope Lext Ols 4100 (Olympus, Tokyo, Japan) with 108-fold magnification.

2.8. Statistical Evaluation

Statistical analyses were performed in MS Excel (Microsoft, Redmond, WA, USA) and Statistica (StatSoft, Palo Alto, CA, USA) using mean values, scatter plots with mean values and ANOVA (analysis of variance) with significance level \( \alpha = 0.05 \).

3. Results and Discussion

The change of surface properties of tested coating systems during AW exposure was evaluated with respect to the type of polymer base (oil, acrylate, alkyd, synthetic—Table 1) and hydrophobic topcoat (acrylate and synthetic—Table 2), see Figure 1.

![Figure 1. Efficiency of hydrophobic layer on the color (a) and gloss change (b) after 6 weeks of AW.](image)
3.1. Color and Gloss Evaluation

In statistical analysis the effect of polymer base type on color and gloss parameters was evaluated as statistically significant ($p = 0.00$). In contrary, the effect of hydrophobic topcoat type on color changes was evaluated as statistically insignificant ($p = 0.07$) and significant for gloss changes ($p = 0.03$). The following graphs show the values of gloss and color changes of individual coating systems with oil and acrylate polymer base (Figure 2) and alkyd and synthetic base (Figure 3). Based on the results, the lowest color and gloss changes were generally recorded for oil and synthetic coating systems (Figure 1). The color change $\Delta E^* < 3$, which cannot be recognized by human eye [39], was after AW test observed only in the case of oil coating systems—OL-8c, OL-6c, OL-8a and OL-10b.

The decrease of gloss during AW exposure occurred almost in all tested variants of coating systems, except OL-3b, OL-4a, OL-7a, AC-2b and AC-2c.

**Figure 2.** Gloss and color changes of oil and acrylate coating systems after 6 weeks of AW.

**Figure 3.** Gloss and color changes of alkyd and synthetic coating systems after 6 weeks of AW.
3.2. Surface Wettability Evaluation

The wetting contact angle provides an overview of the functionality of hydrophobic layers during weathering (Figures 4–6). The change of contact angle of tested coating systems during AW exposure was statistically evaluated with respect to the type of polymer base (oil, acrylate, alkyd, synthetic) and the use of the hydrophobic layer (without hydrophobic treatment, acrylate and synthetic treatment), see Figure 4. Based on the results, there is a significant loss of contact angle due to AW, mainly when hydrophobic layer was not applied (Figure 4). In the case of coating systems without hydrophobic layer (a), the acrylate and synthetic coatings systems have reached the lowest contact angle values. The efficiency of hydrophobic topcoats on the surface wettability was observed for both types, but more pronounced positive effect was observed for the hydrophobic acrylate water-based topcoat (c) in comparison with synthetic one (b).

![Figure 4](image)

**Figure 4.** Efficiency of hydrophobic layer on the contact angle change after 6 weeks of AW.

In statistical analysis the effects of polymer base type and hydrophobic topcoat on contact angle values were evaluated as statistically significant ($p = 0.00$). The following graphs show the values of contact angle changes of oil and acrylate coating systems (Figure 5) and alkyd and synthetic coating systems (Figure 6). At the same time, the factor of total color change is also observed.

Only coating systems, that were not noted for decrease of the contact angle, but for slightly increase after 6 weeks of AW, were coating systems AC-2c, AD-3c, OL-8c, OL-9c, OL-10c, SL-1c a SL-2c. That confirmed the results stated in Figure 4—that acrylate hydrophobic topcoat had the most positive effect on the wettability of tested surfaces. The rest of tested coating systems was noted for slightly or more pronounced decrease of contact angle values after 6 weeks of AW. Full surface wettability (100% decrease of initial value) was determined for some coating systems without hydrophobic topcoat and alkyd coating system with synthetic hydrophobic layer AL-3b.
3.3. Visual and Microscopic Evaluation

Based on the results presented in Table 5 and Figure 7, it can be seen that the results of color and gloss changes (Figures 2 and 3) or changes in the contact angle (Figures 5 and 6) did not always correspond to visually observed degradation of coating systems. For example, coating systems OL-10b and OL-10c were characterized by a relatively low color change (Figure 2) and minimal decrease of contact angle (Figure 5), but their overall visual degradation was quite pronounced (Table 5 and Figure 7). If the top layer of coating is degraded, but the penetrated layer remains, the significant visual changes occur, but the contact angle can be unaffected [8,40]. At the same time, it can be stated that coatings with better results of visual evaluation according to Table 5 (e.g. OL-8, OL-9, SL-3, SL-6b) were characterized by a lower decrease of contact angle values $\Delta CA^*$ and total color change $\Delta E^*$ (except AL-3). Some selected coating systems were for a comparison analyzed by confocal laser scanning microscopy before and after AW test (Figure 8).
Table 5. Degradation of coating systems (based on methodology done by De Windt [41]).

| Sign | Without Hydrophobic Topcoat (a) | With Synthetic Hydrophobic Topcoat (b) | With Acrylate Hydrophobic Topcoat (c) |
|------|----------------------------------|----------------------------------------|--------------------------------------|
| OL-1 | 10                               | 8                                      | 6                                    |
| OL-2 | 10                               | 10                                     | 10                                   |
| OL-3 | 10                               | 10                                     | 10                                   |
| OL-4 | 10                               | 10                                     | 10                                   |
| OL-5 | 8                                | 6                                      | 4                                    |
| OL-6 | 8                                | 4                                      | 8                                    |
| OL-7 | 10                               | 10                                     | 10                                   |
| OL-8 | 4                                | 6                                      | 4                                    |
| OL-9 | 4                                | 4                                      | 2                                    |
| OL-10| 10                               | 8                                      | 8                                    |
| AC-1 | 10                               | 10                                     | 10                                   |
| AC-2 | 10                               | 8                                      | 6                                    |
| AC-3 | 8                                | 8                                      | 6                                    |
| AL-1 | 10                               | 8                                      | 10                                   |
| AL-2 | 10                               | 8                                      | 8                                    |
| AL-3 | 2                                | 4                                      | 4                                    |
| AL-4 | 6                                | 6                                      | 6                                    |
| AL-5 | 8                                | 8                                      | 6                                    |
| SL-1 | 8                                | 6                                      | 6                                    |
| SL-2 | 10                               | 10                                     | 8                                    |
| SL-3 | 4                                | 4                                      | 4                                    |
| SL-4 | 8                                | 6                                      | 6                                    |
| SL-5 | 6                                | 6                                      | 2                                    |
| SL-6 | 6                                | 2                                      | 8                                    |

Note: The evaluation was based on the level of degradation: i.e., 0 = none; 2 = small aesthetical changes; 4 = mild (easy to retreat); 6 = moderate (maintainable); 8 = striking (maintenance is difficult); 10 = advanced (maintenance coat cannot restore the defects). De Windt [41].

Figure 7. Visual changes of selected tested coating systems on oak wood. OL-2a: the same degradation was observed using hydrophobic treatments b and c; OL-9: positive effect of hydrophobic treatment (c) on color stability of AW samples; OL-10: partly positive effect of hydrophobic topcoat application b and c; AL-1: very similar degradation was observed using hydrophobic treatments b and c; AL-4: positive effect of pigment content increasing durability of alkyd coating (in comparison with transparent AL-1a); SL-1: positive effect of application of topcoat b; SL-6b: positive effect of application of topcoat b.
3.4. Final Discussion of Results

The results of the experiment in this work confirmed that application of the top hydrophobic layer can extend the overall durability of coating systems on oak wood as well as on other wood species [24,1,28]. However, it is necessary to apply a suitable hydrophobic layer to a specific coating system, because the resulting effect differs (Figures 5 and 6). The effect of a specific coating system (polymer base and additive content) on its overall service life was confirmed [41–43]. The choice of a suitable oak wood exterior coating system is crucial (Table 4). The application of the top hydrophobic layer did not significantly improve the properties of less suitable coating systems during exposure (Figures 2–8, Table 5). In the case of oil thin layer systems, the positive effect of pigments on the overall service life and color stability (OL-8 and OL-9, see Figures 2 and 5) was confirmed, which is consistent with other studies [26,40,41]. However, in the case of penetration pigmented oil without the top hydrophobic layer OL-10, rapid leaching of coating and loss of functionality occurred. Top hydrophobic layer on alkyd coating systems basically had no significant positive effect on the color stability and only low effect on the increase of the hydrophobicity of tested variants (AL-1 and AL-5). The performance of synthetic coatings depended on a specific combination of layers. Only coating system SL-5 with acrylate hydrophobic layer (c) was characterized by improved functionality, in particular by preserving hydrophobicity after AW (Figure 6). Overall, the topcoat with hydrophobic additives in the acrylate water base (c) appear to be more appropriate for application on oak wood in comparison with synthetic topcoat (b). From all the tested coating systems, OL-8 and OL-9 oil-based coatings with the use of top hydrophobic layer, showed to be suitable for protection of oak wood in terms of color stability and hydrophobic effect as well as overall durability. However, it is necessary to expect a more pronounced reduction of gloss parameter [8] and in the case of OL-9 also color change (Figure 2). The performance of SL-3, SL-5 with hydrophobic topcoat (c) and SL-6 with
hydrophobic topcoat (b) can be positively evaluated from the other tested coatings. On the contrary, transparent coating systems have generally not been effective, their higher vulnerability to degradation in comparison with pigmented ones has been fully manifested [5]. In some cases, their overall service life can be increased by application of top hydrophobic layer (coatings OL-1, OL-5 and 6, SL-1), which is in the line with previous studies on spruce and black locust [1,28]. For the objective determination of coating systems quality after weathering, it is helpful to use the evaluation based on the combination of properties: the overall color and gloss change (Figures 2 and 3) and surface wettability (Figures 5 and 6) followed by a visual evaluation (Table 5, Figure 7).

4. Conclusions

Coated oak wood is relatively quickly subjected to degradation in exterior applications causing changes in color and gloss and reduced resistance to rainwater. The results of the experiments in this work confirmed that a particular selection of the coating system can both positively and negatively affect the overall service life of oak wood in exterior. The semi-transparent oil coatings were generally more stable in color and retained hydrophobicity in comparison with other variants. However, they were more prone to decrease of gloss. Subsequent application of the top hydrophobic layer can in the most cases increase the overall durability of the coating system. The topcoat with hydrophobic additives in the acrylate water base appeared to be more appropriate for application on oak wood in comparison with synthetic hydrophobic topcoat. Based on the variants tested, it can be generally recommended to apply the top hydrophobic layer on oil and partially acrylate coatings. For alkyd systems, the positive effect of additional hydrophobic layer was minimal. In the case of synthetic, the hydrophobic layer only rarely improved the evaluated properties. Although the experiment brings some positive results, oak wood exterior coating systems require further research with the aim to increase their overall durability, especially for transparent and less pigmented semi-transparent variants preserving natural color and texture of oak wood.

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