coatings

Article

Durability of Selected Transparent and Semi-Transparent Coatings on Siberian and European Larch during Artificial Weathering

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Received: 16 November 2018; Accepted: 11 January 2019; Published: 14 January 2019

Abstract: This paper compares the resistance of 20 commercial transparent and semi-transparent coatings applied to European and Siberian larch during artificial weathering in Xenotest. The change in gloss, colour, contact angle of wetting, resistance to *Aspergillus niger* and *Penicillium brevicompactum* moulds was evaluated, and visual changes at the top surface of treated wood species were measured. Overall, the durability of coatings on European larch was higher than that on Siberian larch. The most durable of the tested coatings was a thin-film, i.e., semi-transparent oil-based film containing TiO_2 pigment and propiconazole fungicide. Of the transparent coatings, the most stable was a thick acrylic coating. Conversely, penetrating transparent oil systems had low colour stability and overall lifespan. Artificial weathering of all of the coatings resulted in a marked decrease in their resistance to moulds.

Keywords: artificial weathering; colour; gloss; larch wood; moulds; transparent and semi-transparent coatings; wetting

1. Introduction

Like all natural materials, wood used in building applications is subject to natural weathering—a combination of chemical, mechanical and energy factors acting on its surface. Weathering causes loss of gloss and colour and leads to cracks and change in chemical composition, erosion and roughening of wood surfaces [1,2]. Due to this fact, the appearance of wood changes over the course of several months, with bacteria, moulds and wood-colouring fungi [3,4] contributing to the overall change in colour. Natural weathering can be partially reproduced via artificially accelerated weathering in UV chambers equipped with water spray. However, in such chambers, high-intensity UV radiation combined with rapid fluctuations in moisture kill bacteria and other microorganisms that are involved in natural weathering [5]. UV light causes depolymerisation of lignin and extractives [6,7], which are then washed out by the water, and the wood changes colour simultaneously [8,9]. Degraded wood surfaces are invariably colonized by bacteria and fungi [10]. Paint systems are used to protect wood surfaces against the aforementioned 'complex of abiotic and biotic influences' [11]. The protective function of wood coatings is defined by their thickness [12], water and vapour permeability [13,14], hydrophobicity [15] or adhesion [16]. The aesthetic function is mainly determined by colour consistency [11], gloss [17] and the ability to prevent mould growth [18]. All of the aforementioned characteristics change during exposure outdoors or during artificial accelerated weathering [12,19].

Pigmented coatings cover the natural texture of the wood, however, they simultaneously provide a good protective layer, in particular against the effect of solar radiation [20]. Transparent coatings preserve or enhance the colour and texture of wood, though they also highlight its defects. Transparent coatings containing organic solvents are used, however, these are being increasingly replaced by water-dilutable coatings for ecological reasons [21]. Transparent coatings require more frequent maintenance than pigmented coatings because UV + VIS light penetrates and degrades the film layer and decomposes the wood underneath it [22–25].

On the European market, European and Siberian larch wood is commonly sold for use outdoors. Compared to European larch, Siberian larch contains more extractives and a higher proportion of arabinogalactans [26]. The properties of coatings on larch wood have been examined in several previous studies [27–31]. Coating on larch surfaces with high arabinogalactan and resin contents and/or low wood pH values may interfere and reduce film formation and durability, respectively [30]. On the other hand, a higher extractive content in combination with the narrow sapwood zone enhances the durability of products manufactured from larch in comparison to many other coniferous wood species [32]. However, the natural durability of larch is variable, from very durable to non-durable according to previous studies [33,34].

Moulds degrade coatings and wood, causing changes in colour, gloss, and roughness; some mould species are also harmful to people [35]. Some types of moulds penetrate through paint films and form colonies on and under coatings [36–40]. Growth of mould on wood and wood coatings is most affected by three major factors, i.e., humidity, temperature and nutrients, with moisture being the most important parameter [21]. The time the material is exposed to such factors is also important. Moulds are also an indicator of the possibility of the wood being attacked by decay fungi, in some cases facilitating their entry [41]. Therefore, it is therefore important to establish conditions that prevent the colonization and growth of fungi on paint films. In practice, the addition of fungicides to coatings has proven to be the most effective method of restricting the colonization of paints by moulds, but many of the most effective biocides are no longer available or will be phased out in future [42].

The aim of this work was to compare the quality of selected transparent and semi-transparent coating systems on different polymeric bases applied to Siberian and European larch woods by means of artificial accelerated weathering in Xenotest, and to determine the influence of the larch type on their overall stability.

2. Materials and Methods

2.1. Wood Material

Coatings were applied to samples of European larch (*Larix decidua* Mill; ρ_{12} = 632.5 kg·m^{-3}) and Siberian larch (*Larix sibirica* Ledeb.; ρ_{12} = 652.7 kg·m^{-3}) in dimensions of 60 mm × 50 mm × 20 mm (longitudinal × radial × tangential). They wood was sound and free of biological damage, knots or other growth inhomogeneities. Wood samples were first sanded along the grain with the 120-grit sandpaper. All of samples were conditioned in a laboratory at 20 ± 2 °C and 65% RH) before application of coatings, before artificial weathering, and before coating properties were evaluated.

2.2. Coatings and Their Application

The types and specifications of transparent and semi-transparent UV stabilizing coating systems used to finish wood samples are given in Tables 1 and 2. Coatings were brushed on to samples by brush in a given number of layers and spreading rates (determined by weighing of samples) in accord with technical data supplied by the manufacturers of the coatings (Figure 1).

2.3. Artificial Weathering (AW)

The artificial weathering test was conducted in xenon chamber Q-SUN XE3H (Q-Lab, Cleveland, OH, USA) which simulated the exterior conditions by cycles of irradiation with an intensity of 55 W·m^{-2}

between wavelengths of 300–400 nm and 630 W·m^{-2} between wavelength of 300–800 nm, temperature of 40 °C and 30% relative humidity (2.5 h) and spraying (0.5 h). After every 168 h, of exposure, temperature cycling was performed using a climatic chamber Discovery My DM340 (ACS, Massa Martana, Italy) with temperatures ranging from 80 °C (1 h) to −25 °C (1 h) three times in a row. In total, the samples were cycled for 12 cycles (2016 h) under the aforementioned conditions.

2.4. Gloss Measurements (G*)

Gloss was measured according to EN ISO 2813 [43] at an angle of 60° using a MG268-F2 glossmeter (KSJ, Quanzhou, China). Ten gloss measurements were performed for each tested coating system before and after 168, 504, 1008, 1512, 2016 h of AW. Change in gloss (Δ*G**) was calculated as a percentage difference between weathered and unweathered samples.

2.5. Colour Measurements (L*a*b*)

The colour parameters of the tested samples were measured before and after 168, 504, 1008, 1512 and 2016 h of artificial weathering using a spectrophotometer (CM-600d, Konica Minolta, Osaka, Japan). The device was set to an observation angle of 10°, $d/8$ geometry and D65 light source. The SCI setting was used. Ten measurements were carried out for each surface treatment at identical locations on the samples after each weathering cycle (Figure 2). Evaluations were done in CIE-*L*a*b** colour space using *L**, *a** and *b** colour coordinates. The total colour difference of samples Δ*E** (ASTM D2244-16) [44] was subsequently calculated using the following Equation (1):

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

*L** is the lightness from 0 (black) to 100 (white); *a** is the chromaticity coordinate + (red) or − (green); *b** is the chromaticity coordinate + (yellow) or − (blue); Δ*L**, Δ*a**, Δ*b** represent relative changes in colour parameters between the weathered and the initial state.

Table 1. Specification of tested coating systems.

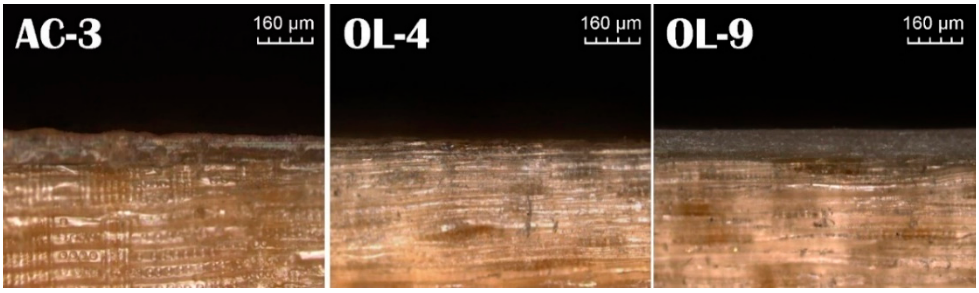
Coating	Type and Specification of Coating *	Coating Base	Transparent/ Semitransp.	Number of Layers	Spreading Rate (g·m^{-2})
REF	Reference without coating	x	x	x	x
AC-1	Acrylate water-based lasur with nanoparticles UV-stabilizers	Acrylate	T	2	100
AC-2	Acrylate water-based lasur with UV-stabilizers and fungicides	Acrylate	T	1 + 2	100
AC-3	Acrylate thick-layer water-based lasur with fungicides (5-chlor-2-methylisothiazol-3(2H)-on)	Acrylate	T	3	100
AL-1	Alkyd water-based lasur with fungicides (IPBC 0.4%) and UV-stabilizers (benzotriazoles < 0.8%)	Alkyd	T	2	100
AL-2	Thixotropic alkyd lasur with UV-stabilizers	Alkyd	T	1 + 1	100
SL-1	Synthetic lasur with fungicides (IPBC 0.3%) and UV-stabilizers	Synthetic lasur	T	2	100
SL-2	Hybrid polyurethane-alkyd synthetic yacht varnish with butanone oxime as additive	Synthetic lasur	T	3	100
SL-3	Synthetic lasur with fungicides (BIT 0.3%) and butanone oxime (0.5%)	Synthetic lasur	T	3	100
OL-1	Oil-based with waxes, natural resins, essential oils	Oil	T	2	80
OL-2	Oil-based with fungicides (BIT and IPBC)	Oil	T	2	80
OL-3	Hemp oils with denaturized white spirit	Oil	T	1 + 2	100
OL-4	Oil water emulsion	Oil	T	2	100
OL-5	Linseed oil	Oil	T	2	100
OL-6	Oil-based with fungicides (propiconazole 0.5%)	Oil	T	1 + 2	100
OL-7	Oil-based with UV-stabilizers (benzotriazoles)	Oil	T	2	100
OL-8	Oil-based with nanoUV-absorbers, plant essential oils	Oil	S	2	80
OL-9	Thin layering oil-based with micronized pigments (TiO_2) and fungicides (propiconazole < 1%)	Oil	S	2	100
OL-10	enetrating oil-based with pigments and terpineol (<2.5%)	Oil	S	2	100
OL-11	Penetrating oil-based with pigments	Oil	S	2	100
OL-12	Thin layering oil-based with micronized pigments (Fe_2O_3) and fungicides (propiconazole < 1%)	Oil	S	2	100

* The specification of coatings is only informative. Some technical data from commercial products was not available. T—Transparent; S—Semi-transparent. The spreading rates are defined for each layer.

Table 2. Initial properties of tested coating systems before artificial weathering.

Coating	European Larch				Siberian Larch				
–	*L**	*a**	*b**	*G**	*L**	*a**	*b**	*G**	CA*
REF	**73.85** 0.47	**8.84** 0.23	**23.63** 0.14	**3.56** 0.05	**60.98** 1.38	**10.28** 0.77	**27.15** 1.00	**8.12** 0.65	**76.24** 2.33
AC-1	**68.83** 0.82	**8.51** 0.53	**30.56** 0.34	**28.78** 1.44	**65.73** 2.70	**9.33** 0.51	**32.30** 1.05	**32.22** 1.42	**85.74** 1.05
AC-2	**65.02** 1.37	**11.02** 0.31	**25.92** 0.20	**7.50** 0.68	**64.29** 1.57	**9.23** 0.21	**28.59** 0.21	**7.44** 0.15	**94.33** 1.70
AC-3	**63.96** 1.93	**11.54** 0.38	**25.72** 1.14	**53.80** 0.85	**62.49** 1.53	**9.90** 0.20	**31.04** 0.52	**44.78** 0.92	**86.78** 3.20
AL-1	**67.73** 2.45	**8.98** 1.32	**30.12** 0.44	**8.04** 0.73	**67.82** 1.26	**7.99** 0.19	**30.53** 0.55	**9.70** 0.28	**96.24** 1.81
AL-2	**63.63** 0.32	**12.60** 0.21	**29.37** 0.70	**48.72** 2.80	**58.23** 0.96	**11.43** 0.38	**29.17** 0.63	**20.60** 0.88	**106.68** 1.16
SL-1	**60.26** 1.35	**15.26** 0.60	**33.50** 1.06	**24.80** 0.37	**61.56** 0.85	**10.78** 0.33	**32.98** 0.20	**22.56** 0.36	**111.21** 1.08
SL-2	**66.81** 1.73	**11.93** 1.04	**33.19** 0.73	**41.62** 3.24	**68.37** 0.32	**9.48** 0.17	**32.90** 0.55	**52.30** 3.15	**100.42** 0.93
SL-3	**68.56** 1.99	**11.55** 1.20	**33.15** 0.61	**6.44** 0.13	**62.34** 1.17	**10.81** 0.50	**32.15** 0.62	**8.32** 0.24	**100.67** 2.40
OL-1	**68.50** 0.96	**11.06** 0.55	**32.36** 0.88	**17.54** 0.11	**62.47** 0.85	**10.75** 0.30	**32.05** 0.39	**15.38** 0.40	**105.32** 3.12
OL-2	**63.41** 1.24	**11.86** 0.39	**28.22** 0.68	**7.40** 1.57	**62.72** 1.44	**10.03** 0.22	**29.78** 0.48	**9.96** 0.71	**107.82** 2.81
OL-3	**60.26** 1.26	**13.35** 0.41	**29.15** 0.44	**90.82** 0.55	**61.38** 1.13	**11.64** 0.50	**32.89** 0.55	**86.50** 0.60	**105.44** 1.62
OL-4	**64.88** 1.42	**12.31** 0.46	**31.94** 0.54	**5.70** 0.28	**58.75** 1.12	**11.84** 0.30	**32.03** 0.48	**18.60** 1.39	**61.02** 3.65
OL-5	**66.18** 0.89	**12.76** 0.63	**34.68** 1.35	**48.72** 2.25	**65.40** 1.32	**9.89** 0.53	**34.40** 0.43	**64.58** 3.43	**99.86** 0.94
OL-6	**63.41** 2.31	**11.25** 1.36	**27.49** 2.14	**47.86** 2.44	**59.10** 1.70	**9.54** 0.34	**29.18** 0.70	**50.02** 1.61	**93.16** 1.84
OL-7	**69.30** 0.63	**10.44** 0.29	**26.29** 0.17	**12.84** 0.51	**64.95** 2.52	**9.79** 0.44	**30.34** 0.58	**39.30** 2.10	**78.60** 2.73
OL-8	**72.65** 0.51	**10.09** 0.27	**25.29** 0.73	**27.64** 1.89	**68.50** 0.52	**7.25** 0.25	**21.89** 0.53	**30.48** 0.51	**93.36** 1.21
OL-9	**70.61** 0.74	**7.22** 0.75	**16.98** 1.06	**31.90** 2.08	**68.49** 0.22	**6.44** 0.16	**17.34** 0.36	**27.34** 0.34	**109.15** 1.73
OL-10	**62.50** 1.18	**12.96** 0.38	**31.47** 0.53	**58.42** 2.06	**62.78** 1.00	**10.20** 0.34	**32.72** 0.21	**67.66** 1.84	**104.47** 3.15
OL-11	**55.98** 1.62	**18.32** 0.41	**33.59** 0.94	**10.44** 0.56	**52.11** 0.83	**16.22** 0.20	**31.29** 0.63	**26.32** 2.85	**103.37** 0.54
OL-12	**58.91** 0.74	**14.91** 0.18	**28.48** 0.35	**75.84** 1.61	**55.01** 0.57	**14.38** 0.23	**29.61** 0.50	**69.46** 1.40	**105.22** 1.9

Mean values in bold in black; standard deviations in bold; number of measurements n = 10. *L**, *a**, *b**, *G** and CA* are described in Materials and Methods—parts 2.4–2.6.



(A) (B) (C)

Figure 1. Images from confocal laser scanning microscope showing a typical film of thick acrylate coating ((**A**) AC-3 in this case), penetrating oils ((**B**) OL-4 in this case) and oil-based coatings also creating a surface layer ((**C**) OL-9 in this case) after application. It is possible to see only very poor penetration into the first layer of tracheids in the impermeable larch heartwood.

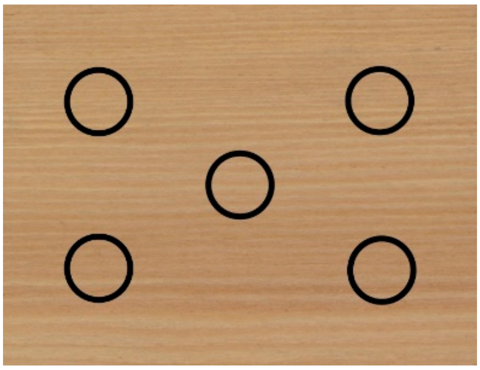


Figure 2. Template used to ensure that locations of colour measurements on samples was the same at each exposure interval—measuring area was given using $d/8$ mm geometry of spectrophotometer.

2.6. Surface Wetting Measurements—Contact Angle (CA*)

The sessile drop method with static contact angle measurement was performed using the methodology of Bastani et al. [45]. The wettability measurements were conducted using a goniometer (DSA 30E device, Krüss, Hamburg, Germany) on radial surfaces of wood samples before and after 168, 504, 1008, 1512 and 2016 h of artificial weathering. Ten measurements were taken for each coating. Contact angles were determined after 5 s (distilled water with a dosing volume of 5 μL). The change in the contact angle (ΔCA*) was calculated as a percentage difference between weathered and unweathered samples.

2.7. Mould Test

The samples for the mould test (50 mm × 10 mm × 5 mm = longitudinal × radial × tangential) were prepared from coated larch wood samples (60 mm × 50 mm × 20 mm) before and after AW. Their sterilization was performed with a 30 W germicidal lamp (Chirana, Slovakia) from a distance of 1 m at a temperature of 22 ± 2 °C/0.5 h.

Two mould fungi, *Aspergillus niger* and *Penicillium brevicompactum*, were used in a mixture for the mould bioassay of coated larch wood. Samples exposed in Petri dishes on Czapek-Dox agar. The mould bioassay lasted 28 days at 28 ± 1 °C, with relative humidity of 90% ± 3%, in accord with standard STN 49 0604 [46], similar to in the test carried out by Viitanen [47]. The growth activity of moulds (GAM) on the top surfaces of samples was evaluated after 7, 14, 21 and 28 days using the following criteria: 0 = no growth on surfaces; 1 = growth ≤ 10%; 2 = growth ≤ 25%; 3 = growth ≤ 50%; 4 = growth > 50%.

2.8. Visual Evaluation and Microscopic Analyses

To evaluate the visual degradation of coatings, samples were regularly scanned using a desk top scanner at a resolution of 300 DPI resolution (Canon 2520 MFP, Canon, Tokyo, Japan) before and after artificial weathering. Microscopic analyses of coatings and wood surfaces used a confocal laser scanning microscope (Lext Ols 4100, Olympus, Tokyo, Japan) with 216-fold magnification.

2.9. Statistical Analysis

Statistical evaluation of data was done in MS Excel 2013 (Microsoft, Redmond, USA) using mean values and bar graphs, and in Statistica 12 software (Statsoft, Palo Alto, CA, USA) using mean values, standard deviations, linear regression between colour, gloss and CA° changes (their similarities were evaluated on the base of coefficients of determination values R^2), analysis of variance (ANOVA), and Tukey's HSD multiple comparison test at $\alpha = 0.05$ significance level.

3. Results and Discussion

Our results focus on the evaluation of changes in colour (Figures 3–5), gloss (Table 3), visual defects (Figures 6 and 7) and changes in the hydrophobicity of coatings (Figure 8) after artificial accelerated weathering. Additionally, the ability of the coated wood samples to resist mould growth before (Table 4) and after weathering (Table 5) is described.

3.1. Changes in Colour (ΔE^*), Gloss (ΔG^*) and Visual Appearance

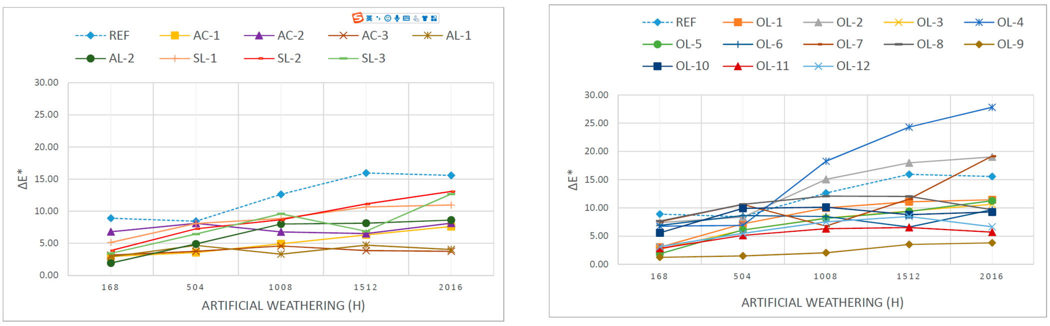
Most of the wood coatings showed pronounced colour change at the beginning of the weathering trial (after 168 h), as others have also observed [48,49]. Considering the finding that that $\Delta E^* < 3$ is a colour difference of wood surfaces that cannot be distinguished by a subjective observer [50], none of the coating systems were able to restrict colour changes during weathering. However, some of the coatings performed better than others.

More pronounced increase in total colour change after 1000 h of accelerated weathering (Figures 3 and 4) was associated with degradation of the protective coating (Figures 6 and 7) and the leaching of photodegraded materials (extractives and lignin) [7] from the underlying wood surface [9]. Despite the assumption that pigmented coating systems are generally more stable in terms of colour than transparent coating systems [11], this assumption is only supported by performance of OL-9 (thin layer oil-based finish containing TiO_2 and fungicide). Due to its TiO_2 pigment content, this coating was more resistant to photodegradation [51]. Semi-transparent coatings such as OL-8; OL-10; OL-11 are eroded from the surface of impermeable wood species such as larch (according to EN 350 [32]), and therefore they rapidly change colour during weathering [52]. The thin OL-12 oil coating showed lower colour stability (OL-12 contained Fe_2O_3 pigments) compared to the OL-9 (contained TiO_2 pigments). Of the tested transparent coatings, those that were more colour stable were acrylate (in particular AC-3), and alkyd (AL-1) finishes. In contrast the oil (OL-1 to OL-7) and synthetic (SL-1 to SL-3) coatings were less stable. Coating colour changes and degradation were greater on Siberian larch, after 2016 h of artificial weathering than those on European larch (Figures 3–7). Such differences were statistically significant (Figure 5). ANOVA analysis yielded a main effect for the wood species, $F(1,21) = 58.6$, $p < 0.001$; coating systems, $F(20,21) = 59.2$, $p < 0.001$; interaction effect between wood species and coating systems $F(20,21) = 23.5$, $p < 0.001$ and was statistically significant in all cases. The precise reason for the significant effect of wood species on coating performance may is not known but it may be related to different chemical composition and higher arabinogalactan [26] content of Siberian larch compared to European larch, which causes faster decomposition of the coating film during exposure [30]. Unlike more complicated colour evaluation models [30,31], colour changes here were only used to compare tested coatings. Using a spectrophotometer with $d/8$ geometry, combined with narrow annual rings of larch (Figures 2, 6 and 7) resulted in colour coordinate values with low standard deviations (see Table 1).

Changes in gloss were more pronounced for oil coatings and slightly higher for coatings on Siberian larch wood (Table 3). The change in the gloss of the coatings was more pronounced than the change in colour after 168 h of artificial weathering, in accord with previous findings [17,25]. Some acrylate (AC-1 only for European larch), alkyd (AL-2 only for European larch) and synthetic finishes (SL-1 for both wood species) were better at retaining their initial gloss during exposure. Overall, these characteristics point to the rapid degradation of the surface layers of the coating system [17], however, the gloss change results did not generally correspond with the overall durability of the coatings that was evaluated visually (Figures 6 and 7).

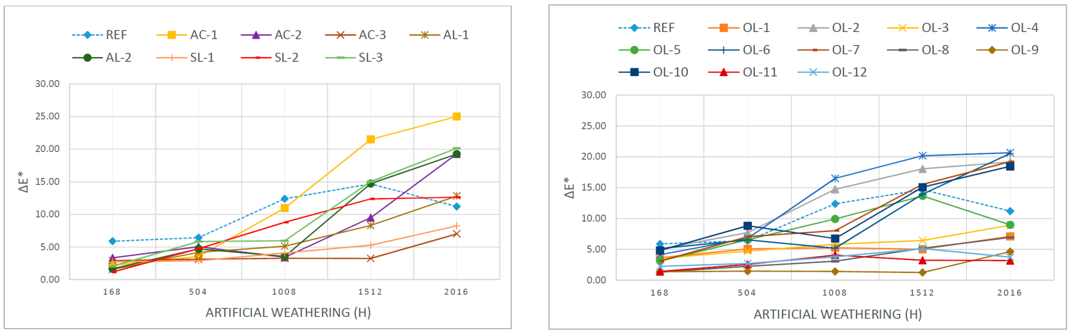
Table 3. Gloss changes of tested coatings on the European and Siberian larch during artificial weathering.

Coating Systems	European Larch					Siberian Larch				
	ΔG_{168}	ΔG_{504}	ΔG_{1008}	ΔG_{1512}	ΔG_{2016}	ΔG_{168}	ΔG_{504}	ΔG_{1008}	ΔG_{1512}	ΔG_{2016}
REF	3.86	61.60	93.63	64.44	34.76	−2.93	−15.13	−3.00	−29.22	−37.75
AC-1	0.28	−11.01	−5.53	−9.29	−5.45	−3.16	−21.48	−30.13	−52.44	−50.69
AC-2	20.24	38.79	47.24	40.62	44.54	13.16	15.89	19.41	20.71	26.97
AC-3	−8.14	−6.24	−13.72	−20.09	−29.81	−2.02	−12.31	−19.81	−24.56	−29.35
AL-1	−19.37	−26.88	−25.17	−27.22	−25.12	−13.78	−19.20	−23.29	−19.44	−12.47
AL-2	−2.38	−5.04	−14.72	6.02	12.41	−4.62	−5.46	−4.62	−18.55	−42.99
SL-1	−12.66	−15.48	−18.06	1.87	20.71	−6.29	−13.66	−9.84	8.92	12.69
SL-2	4.68	14.40	16.92	16.53	14.91	1.00	−8.26	−2.57	−18.24	−31.99
SL-3	−31.35	−41.91	−40.67	−9.59	0.98	−46.87	−55.74	−43.18	−37.92	−39.87
OL-1	11.40	−9.03	−38.10	−68.53	−82.44	−19.98	−4.15	−40.62	−73.32	−84.38
OL-2	−32.12	−32.21	−14.00	−25.55	−33.16	−43.01	−49.15	−43.95	−49.37	−52.03
OL-3	−7.82	−17.91	−20.89	−23.89	−21.60	−16.57	−37.44	−44.86	−42.74	−42.32
OL-4	−17.70	7.76	32.73	2.00	−19.73	−38.55	−57.97	−64.70	−74.11	−78.31
OL-5	−21.72	−51.77	−69.50	−60.82	−73.08	−18.38	−44.94	−65.70	−70.54	−86.97
OL-6	−3.13	−32.01	−43.20	−58.14	−68.03	−13.42	−37.52	−52.36	−62.53	−68.06
OL-7	−2.76	−8.54	−26.01	−35.60	−37.20	−14.57	−31.50	−63.59	−73.75	−76.32
OL-8	65.89	86.50	82.04	29.59	−52.05	42.75	40.40	47.26	−31.74	−66.75
OL-9	−43.85	−60.27	−60.32	−63.94	−70.94	−49.02	−61.22	−64.65	−64.95	−73.01
OL-10	−40.73	−82.93	−89.28	−87.75	−80.54	−66.09	−86.18	−92.40	−91.87	−92.01
OL-11	−50.04	−68.00	−71.39	−60.53	−71.34	−56.11	−75.75	−78.02	−80.74	−91.31
OL-12	−28.37	−35.54	−41.14	−49.49	−57.52	−24.41	−39.54	−51.23	−56.54	−73.04



(**A**) (**B**)

Figure 3. The total colour difference of acrylate, alkyd, synthetic (**A**) and oil-based (**B**) coating systems applied to European larch during artificial weathering lasting 168 h, 504 h, 1512 h and 2016 h.



(**A**) (**B**)

Figure 4. The total colour difference of acrylate, alkyd, synthetic (**A**) and oil-based (**B**) coating systems applied to Siberian larch during artificial weathering lasting 168, 504, 1512 and 2016 h.

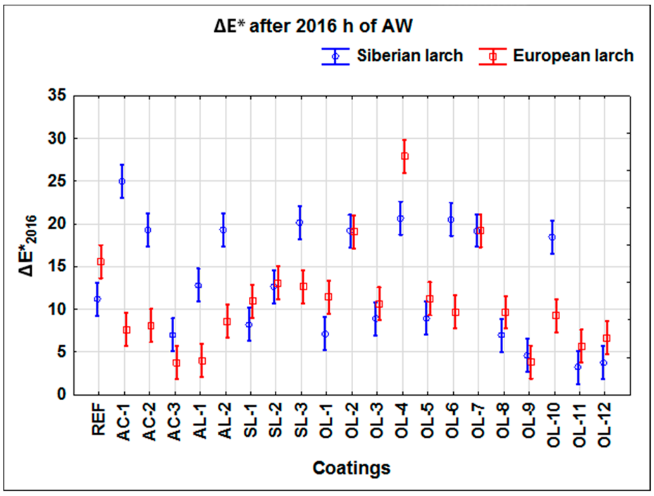


Figure 5. Total colour difference of coating systems applied to European and Siberian larch after 2016 h of artificial weathering evaluated as 95% two-side confidence intervals. The Tukey HSD test shows that the differences in the analysed values were statistically significant (p-value < 0.05) for coating systems AC-1, AC-2, AL-1, AL-2, SL-3, OL-4, OL-6 and OL-10.

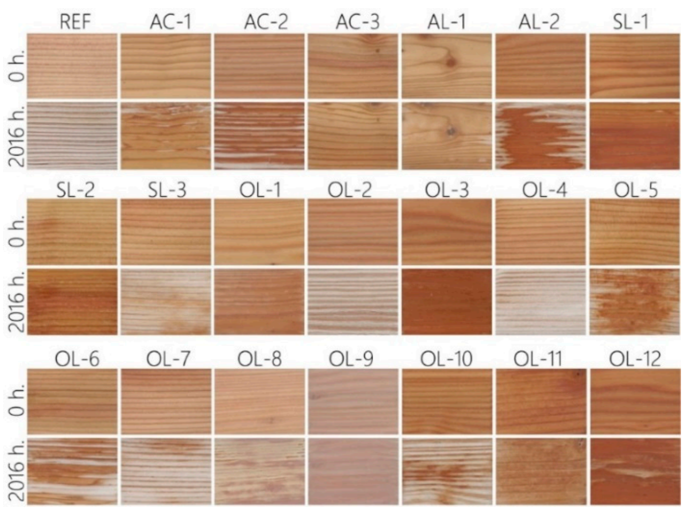


Figure 6. Visual evaluation of coating systems applied to European larch before (0 h) and after (2016 h) AW.

Figure 7. Visual evaluation of coating systems applied to Siberian larch before (0 h) and after (2016 h) AW.

Visual evaluation of coatings after artificial weathering confirms the more pronounced degradation of coatings on Siberian larch compared to European larch. (Figure 6 versus Figure 7). Flaking of acrylate and alkyd thick-layer finishes and decomposition and defoliation of oil-based coatings was observed. In the work of Grüll et al. [30] some waterborne coatings for the finishing of larch wood with higher arabinogalactans content were mentioned as being inadequate, however, our results indicate that synthetic and oil-based coatings have problems on Siberian larch.

3.2. Changes in Wetting

Surface wetting measurements (Figure 8) indicate the overall impairment of the protective function of the coating systems against water [25,29]. In all cases where there was a significant decrease in CA* (above 50%) of coatings, there was always complete degradation and loss of adhesion of the coating systems (Figures 6 and 7). Despite their high colour changes (OL-1; OL-6 and OL-7), some of the transparent oil coatings, and also the pigmented coating OL-9, were able to maintain the hydrophobicity of wood surfaces. The positive effect of oil finishes on the hydrophobicity of larch during weathering was noted by Žlahtič and Humar [29]. Amongst the other coatings, AC-2 and SL-1 (and partially also AC-3) showed good hydrophobicity after 2016 h of weathering on both European and Siberian larch. Overall, for more of the durable tested coatings, lower CA* changes corresponded with lower colour change ΔE^*, even taking into account the different performance of the coatings on the two larch species (Figures 3–5, Figure 8). ANOVA analysis yielded a main effect for the wood species, $F(1,21) = 17.9$, $p < 0.001$; coating systems, $F(20,21) = 30.7$, $p < 0.001$; and interaction effect between wood species and coating systems $F(20,21) = 3.1$, $p < 0.001$ and was statistically significant in all cases. Tukey HSD test, indicated that significant differences (at a 95% significance level) resulted from the performance of two of the tested coating systems (Figure 8).

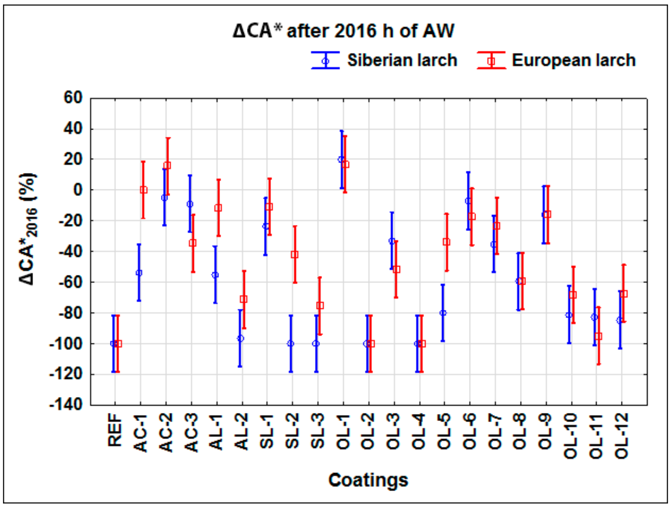


Figure 8. Changes in the water contact angle on the tested coatings in percentages after 2016 h of artificial weathering evaluated as 95% two-side confidence intervals. The Tukey HSD test shows that the differences in the analysed values were statistically significant (p-value < 0.05) for coating systems AC-1 and SL-2.

3.3. Effect of Moulds

The individual coatings showed different resistance to mould growth (growth of moulds, GAM) (Table 4). GAM was not related to the polymer base of the coating system, but rather to the effects of fungicides, which accords with other research [22,27,47]. In particular, there was a positive effect of fungicides in coatings AL-1, SL-1, OL-2, OL-9, and OL-12 (IPBC fungicides and propiconazole) on the resistance of coatings to mould growth, in accord with previous findings [27,53]. The underlying larch species also influence mould growth on coatings, as mould growth was less pronounced on Siberian larch (1/2 to 1 degree better) than on European larch.

The resistance of coatings to moulds after accelerated weathering of samples in Xenotest for 2016 h is interesting (Table 5) and confirmed the results Gobakken and Westin who employed natural weathering in their research [27]. In the latter work [27], the growth of *Aureobasidium pullulans* on naturally weathered surfaces was observed. *Aureobasidium pullulans* required preconditioning by other microorganisms in order to grow on paint film [54]. In this work, taking this fact into account, a mould test of samples after accelerated weathering in a UV-chamber under sterile conditions was carried out with *Aspergillus niger* and *Penicillium brevicompactum* (see Materials and Methods). In the final 28 days of the mould test, only the OL-9 coating performed well, which, due to its pigment content, was more colour stable (Figure 5) and continued to maintain its hydrophobicity (Figure 8). Small visually monitored degradation of this coating, in particular for European larch (Figures 6 and 7), also affected its ability to maintain its effectiveness against mould growth. In addition to its fungicide (propiconazole) content (the same as in OL-12 containing Fe_2O_3 pigments—see Table 1), a biocidal effect of TiO_2 in this coating (OL-9) is also evident, as has been reported for anatase form of TiO_2 [55]

Table 4. Growth activity of moulds (GAM) on surfaces of larch samples treated with commercial coatings before artificial weathering (B-AW).

Coating	European Larch (B-AW)					Siberian Larch (B-AW)				
	GAM (from 1 to 4)					GAM (from 1 to 4)				
	4 Day	7 Day	14 Day	21 Day	28 Day	4 Day	7 Day	14 Day	21 Day	28 Day
REF	2–3	3–4	3–4	4	4	3–4	4	4	4	4
AC-1	0–1	1	1-2	2	2–3	0	0-1	1	1	1–2
AC-2	0–1	0–1	0-1	0–1	0–1	0	0	0	0	0
AC-3	2–3	3	3–4	3–4	3–4	1–2	2–3	2–3	2–3	2–4
AL-1	0	0	0	0	0	0	0	0	0	0
AL-2	2	3–4	3–4	3–4	3–4	2	3–4	4	4	4
SL-1	0	0	0	0	0-1	0	0	0	0	0
SL-2	0–1	0–1	1	1	1–2	0	0–1	0–1	0–1	1
SL-3	2–3	3–4	3–4	3–4	3–4	1–2	2	2	2	2
OL-1	1	1	2	2–3	3	1	1	1–2	2	2–3
OL-2	0	0–1	0-1	0-1	0–2	0	0	0	0	0
OL-3	0–1	1–2	2–3	3	3	0	0	1	2	2–3
OL-4	2–3	4	4	4	4	3–4	4	4	4	4
OL-5	1–2	2–3	4	4	4	2	3	4	4	4
OL-6	2–3	4	4	4	4	1–2	3–4	4	4	4
OL-7	2	4	4	4	4	1–2	3-4	4	4	4
OL-8	0	0	0–1	0–1	1–2	0	0	0	0	0–1
OL-9	0	0	0	0–1	1	0	0	0	0	0
OL-10	0–1	2	2–3	2–4	3–4	1	2	2–3	3	3–4
OL-11	1–3	2–4	3–4	4	4	1–2	2	2–3	2–3	2–3
OL-12	0–1	1	1	1	1	0	0	0	0	0–1

Table 5. Growth activity of moulds (GAM) on surfaces of larch samples treated with commercial coatings after artificial weathering lasted 2016 h (A-AW).

Coating	European Larch (A-AW)					Siberian Larch (A-AW)				
	GAM (from 1 to 4)					GAM (from 1 to 4)				
	4 Day	7 Day	14 Day	21 Day	28 Day	4 Day	7 Day	14 Day	21 Day	28 Day
REF	4	4	4	4	4	4	4	4	4	4
AC-1	2–3	3–4	4	4	4	4	4	4	4	4
AC-2	1–2	3–4	3–4	4	4	1–3	2–3	2–4	3–4	3–4
AC-3	2–3	3	3	3–4	3–4	3	3–4	3–4	3–4	3–4
AL-1	2–3	4	4	4	4	2–3	3	3–4	3–4	4
AL-2	2–3	3	3–4	3–4	3–4	4	4	4	4	4
SL-1	1–3	3	3–4	3–4	3–4	3–4	4	4	4	4
SL-2	1–2	1–2	2–3	2–3	3	1–3	2–3	3–4	3–4	3–4
SL-3	4	4	4	4	4	4	4	4	4	4
OL-1	1–2	2–3	3–4	3–4	4	1–2	2–3	2–3	3–4	3–4
OL-2	4	4	4	4	4	3	4	4	4	4
OL-3	1–2	2–3	3–4	3–4	3–4	1	1–2	1–2	2–3	3
OL-4	4	4	4	4	4	4	4	4	4	4
OL-5	2–3	3	3–4	3–4	3–4	3–4	3–4	4	4	4
OL-6	2–3	3–4	4	4	4	4	4	4	4	4
OL-7	4	4	4	4	4	4	4	4	4	4
OL-8	3	4	4	4	4	4	4	4	4	4
OL-9	0–1	1	1–2	1–2	1–2	1–3	2–3	2–3	2–4	2–4
OL-10	3–4	4	4	4	4	4	4	4	4	4
OL-11	4	4	4	4	4	4	4	4	4	4
OL-12	2–4	3–4	3–4	3–4	4	4	4	4	4	4

After accelerated weathering in Xenotest, the mould resistance of many of the coatings decreased (Table 4 versus Table 5). This phenomenon was most evident after weathered coated samples were exposed to mould for longer during the bioassay, i.e., typically after 28 days compared to 4 days (e.g., coatings AC-2, SL-2, OL-3). This observation has two possible explanations: 1) Fungicides in coatings, mainly IPBC, are susceptible to leaching [56], and 2) in coatings where there was a more significant degradation of the film due to previous artificial weathering (Figures 6 and 7), the decrease in mould resistance was very pronounced after 4 days of exposure (e.g., AL-1; SL-1; OL-2; OL-8; OL-12).

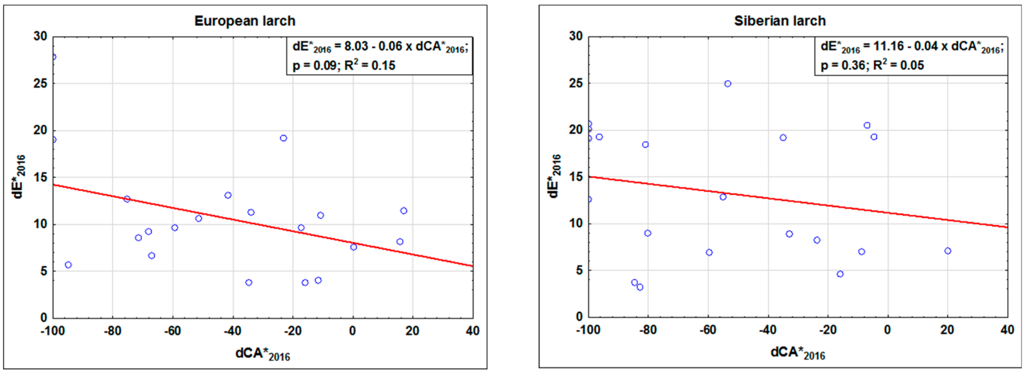
The specified results point to a strong effect of mould on the overall aesthetic and functional deterioration of coatings and colour changes in coated wood during long-term outdoor exposure [11,36,42,57]. They also suggest that the required testing standards for coating resistance to mould growth [58–60] should take into account changes in mould resistance of coating films due to weathering.

The addition of mould testing to artificial weathering would bring the laboratory testing of coatings [58,59,61] closer to real conditions during exterior exposure [62–64]. Such a change would facilitate better choices of coating systems for wood used outdoors, i.e., coatings which are better at retaining their antifungal efficiency after long-term exposure to UV and VIS radiation, water and temperature fluctuations.

This work has also confirmed the influence of the wood species [37] on coating performance (Figures 5–8). The longevity of coatings on European and Siberian larch appeared to influenced by their properties even though they contain similar extractives [32]. A positive effect of pigments and fungicides (mainly in coating OL-9 with TiO_2 pigments and 1% of propiconazole) on coating longevity was observed, but not in all cases. Gobakken and Westin [27] also observed a positive effect of fungicides and pigments on mould resistance of coatings exposed to natural weathering.

Siberian larch is a wood that has adverse effects on coating durability. Further research is needed to solve this problem. One possible approach is chemical treatment of the surfaces to reduce the impact of extractives on coating performance [30], or the use of UV-stabilizers or nanoparticles for surface pre-treatment of wood [11]. Another approach is using of plasma treatments to increase the hydrophobicity and penetration or adhesion of coatings on wood surfaces [65–67], or the use of UV-short-pulse laser incisions [68] to increasing surface penetrability and adhesion of the protective films.

The colour stability (Figures 3–5), surface wetting (Figure 6), and gloss changes (Table 3) were poorly correlated with coating durability (Figure 9). The measured data was variable and often did not exactly correspond with degradation of the tested coatings. Visual evaluation (Figures 6 and 7) was a better evaluation criterion for the performance of the coatings.



(**A**) (**B**)

Figure 9. Linear correlations between total colour changes (Δ*E**) and changes in surface wetting (ΔCA*) after 2016 h of accelerated weathering of coatings on European (**A**) and Siberian larch wood (**B**). Very poor correlations were also found between Δ*E** and gloss changes (Δ*G**): R^2 = 0.02 for European and R^2 = 0.02 for coatings on Siberian larch; between Δ*G** and ΔCA*: R^2 = 0.02 for European and R^2 = 0.06 for coatings on Siberian larch wood.

4. Conclusions

Our results show an influence of wood species (Siberian larch v. European larch) on the overall durability of coatings on wood samples exposed to artificial weathering. Generally, the durability of coatings applied to European larch was better than on Siberian larch. This generalization is relevant to synthetics and oil-based coatings, as well as waterborne coatings. A positive effect of pigment content on the performance of oil-based coating (OL-9) was observed. Only penetrating pigmented coatings had poor long-term durability and colour consistency. Of the tested transparent coatings, the acrylate coating (AC-3) and, to a lesser extent the alkyd coating (AL-1) had the best colour consistency and overall durability. Transparent oil-based coatings showed high colour change during accelerated weathering. Overall, it is clear from our results that additives have a greater impact on the quality and durability of coatings compared to the polymer base.

Coatings were more susceptible to mould growth after artificial accelerated weathering. This finding suggests that weathering pre-treatments should be used prior to mould bioassays during laboratory testing of exterior coatings. A direct relationship between overall coating durability during artificial weathering and the ability to subsequently resist mould growth was only observed for the most durable coating that was tested (OL-9, oil-based coating containing pigment and fungicide).

Author Contributions: Writing-Original Draft Preparation, K.Š.; Methodology, Data Curation, Project Administration and Visualization, E.O.; Writing-Review & Editing and Investigation, L.R.; Conceptualization, Writing-Review & Editing, M.P. (Miloš Pánek); Investigation, M.P. (Milan Podlena); Investigation, I.Š.

Funding: This research was funded by the project "Expand and support of the transfer of technologies and knowledge at CULS by activities of "proof-of-concept", reg. No. CZ.07.1.02/0.0/0.0/16_023/0000111-Activity KZ04-Development of facade wood cladding with increased durability and colour stability".

Conflicts of Interest: The authors declare no conflict of interest.

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