Effects of Wood Roughness, Light Pigments, and Water Repellent on the Color Stability of Painted Spruce Subjected to Natural and Accelerated Weathering

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This study examined the color stability of painted Norway spruce (*Picea abies*) samples subjected to natural and accelerated weathering, using Duncan's tests and correlation analyses. The following effects were studied: (1) the different initial roughness of the wood; (2) use of transparent or lightly-pigmented top-coat layers; and (3) the presence of the final water-repellent layer. Natural weathering at a 45° slope in an industrial zone lasted 104 weeks, whereas accelerated weathering in Xenotest with 0.55 W/m^2 UV irradiation at 340 nm and sprayed water lasted 12 weeks. The color stability of painted spruce, measured in a CIE-$L^*a^*b^*$ system, was not, in the majority of cases, significantly affected by the initial roughness of the wood, the type of top-coat (WoodCare UV or PerlColor) layer, or presence of the final water repellent (AquaStop) layer. The light pine or larch pigments in the top-coat layers had positive color stabilizing effects. In their presence, the darkening ($-\Delta L^*$) and total color differences (ΔE^*) of the painted samples dropped *ca.* 2.5 times during exterior weathering and *ca.* 5 times during Xenotest weathering. Samples painted with transparent coatings turned a reddish shade ($+a^*$) during the Xenotest, while those exposed to the exterior absorbed dirt and became more blue ($-b^*$).

Keywords: Wood roughness; Water repellent; Pigments; Photodegradation; Color changes

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INTRODUCTION

High resistance against weathering is a basic requirement of wood finish coatings in order to maintain a stable appearance and long service life. Coating systems for wooden products exposed to the outdoors are designed to provide protection for the wood from the harsh environment of outdoor weathering, especially the degradation caused by sunlight, oxygen, water, and fungi. Acryl, alkyd, polyurethane, epoxide, and other film-forming coatings designed for exterior use usually contain specific UV-stabilizer molecules (organic UV absorbers – UVA, inorganic screeners used as nano-sized systems as well, *etc.*), micro-particle pigments, hydrophobic additives, and fungicides (Reinprecht *et al.* 2011). Their composition must be balanced and optimized with the aim to achieve their complementary and synergetic effects. For example, Samyn *et al.* (2014) found that poly(styrene-maleimide) nanoparticle coatings without suitable additives (*e.g.*, vegetable oils) do not form a fully protective layer due to their porous structure after drying.

Various transparent water-based paints are modified with UVA, such as hydroxyphenyl-s-triazines (Ozgenc *et al.* 2012; Forsthuber *et al.* 2013), hydroxyphenyl-benzotriazoles (Forsthuber *et al.* 2013), imidized nanoparticles (Samyn *et al.* 2014), polyvalent metal complexes, and other compounds. Mineral screeners – UV blockers such as zinc oxide and titanium dioxide (Cristea *et al.* 2010; Forsthuber *et al.* 2013), HALS – hindered amine light stabilizers (Schaller *et al.* 2009, Šomšák *et al.* 2015), antioxidants such as bark extract (Saha *et al.* 2011), and lignin stabilizers such as succinic anhydride (Teacà *et al.* 2013) are some of the most progressive color stabilizing additives in the transparent or lightly-pigmented coating systems for the finishing of outdoor wood products. It is also important that these coatings have good aesthetic quality and maintain the appearance of the natural, uncoated wood.

The durability of coating systems on wood surfaces is characterized by the state when specific properties become technically dysfunctional or aesthetically unacceptable for the end user or from a technical point of view (Grüll *et al.* 2011). The most important properties of wood coatings are their color stability and functional stability, measured in terms of their hydrophobic properties, liquid water and vapour diffusivity, elasticity, and adhesion strength (Turkulin *et al.* 2006).

The aging processes of coating films and wood involve various irreversible chemical and physical changes in their structures. UV light promotes the breakdown of film-forming components (acryl, alkyd, urethane, epoxide, *etc.*) in the coatings, creating various defects on the painted wood surface. Some of these defects include: damaging the structure of coatings, creating cracks and bubbles; thinning the coatings; and changes in the color of the painted wood surfaces (Masaryková *et al.* 2010). Evaluating the weathering of coating systems present on the wood surface is important for understanding their practical application. Decomposition of coatings can be studied by indicators based on the changes in the Fe^{2+} concentration in the coating films (Grüll *et al.* 2014b), on the changes of the dry film thickness (Mamoňová and Reinprecht 2008; Grüll *et al*. 2014b), on the changes in the micro-structure of the coating films (Masaryková *et al.* 2010), or on the color changes in the coating film-wood substrate composite.

The color stability of wooden products painted with transparent or lightly-pigmented (semi-transparent) coatings is usually influenced by numerous factors. Important effects are attributed to their material characteristics, namely to the wood surface, such as its chemical and anatomical structure, wettability, roughness, sap/heartwood, early/latewood, knots content, *etc*., and also to the coating system, such as its surface energy, polarity, viscosity, and chemical structure of the individual macromolecules, UV absorbers, and other additives. The technologies used to apply the coatings (spraying, dipping, *etc*.) have a milder effect. Finally, outdoor weathering conditions including time and strength of sunlight, rain water, and pollutants have large effects. UV and visible rays of sunlight can penetrate several micrometers into the wood through transparent coatings, and color of the painted wood changes due to the photodegradation of lignin, hemicelluloses, and extractives, in connection with its yellowing and red-browning. When coatings are destroyed, the wooden surfaces begin to gray rapidly due to the leaching of depolymerized and oxidized lignin and some other wood components, and the adsorption of dark pollutants (Evans 2008).

The basic aim of this work is to, under natural and accelerated weathering, evaluate the effects of the initial roughness of wood and the presence of a final water repellent layer on the painted wood on its color stability. Changes in the color of painted wood during weathering, as affected by two light-shade pigments present in two kinds of top-coat layers, is examined, as well.

EXPERIMENTAL

Wood

Norway spruce (*Picea abies* Karst L.) boards were used to prepare weathering samples; the wood samples were without biological damage, cracks, or other non-homogeneities. Samples for natural weathering had dimensions of 375 x 78 x 20 mm (axial x radial x tangential) in accordance with the EN 927-3 (2006) standard. The top surfaces of these samples were divided into two parts, each with length of 187.5 mm: the rough part (which was sanded along the grain with 60-grit sandpaper) and the smooth part (which was sanded with 60-grit sandpaper followed by 120-grit sandpaper). Samples subjected to accelerated weathering in accordance with a partly modified EN 927-6 (2006) test standard, had dimensions of 55 x 38 x 8 mm (axial x radial x tangential), at which their top surface was either rough or smooth. The transverse sections (78 x 20 mm, or 38 x 8 mm) of the samples were treated with silicone.

Wood Coating Systems

Twelve different wood coating systems (Table 1) were prepared from water-based paints, which were manufactured by Böhme AG Farben & Lacke (Switzerland).

Table 1. Color Components L^*, a^*, and b^* of Reference and Painted Norway Spruce (*Picea abies*) Samples before Natural Weathering

Coating System		**Rough Wood**			**Smooth Wood**		
		*L**	*a**	*b**	*L**	*a**	*b**
	Reference – Native wood	83.7 (0.6)	4.5 (0.3)	19.8 (0.4)	84.2 (0.6)	4.2 (0.3)	19.2 (0.5)
1)	SC-WC/T	76.4 (1.1)	6.8 (0.5)	33.9 (1.0)	77.2 (1.5)	6.4 (0.6)	33.5 (0.9)
2)	SC-WC/T-AS	77.4 (1.4)	6.6 (0.7)	33.9 (0.7)	78.5 (0.8)	5.9 (0.3)	33.7 (0.5)
3)	SC-PCO/T	79.7 (1.2)	5.6 (0.6)	29.3 (0.7)	80.3 (0.6)	5.3 (0.2)	29.2 (0.4)
4)	SC-PCO/T-AS	81.0 (0.7)	5.1 (0.4)	28.3 (0.3)	80.5 (0.9)	5.3 (0.4)	28.3 (0.2)
5)	SC-WC/P	38.2 (0.2)	17.8 (0.3)	16.6 (0.3)	38.8 (0.8)	18.7 (1.1)	17.6 (1.3)
6)	SC-WC/P-AS	39.6 (0.9)	19.7 (1.0)	19.8 (1.4)	40.8 (1.0)	20.9 (1.0)	21.3 (1.7)
7)	SC-PCO/P	43.3 (1.9)	21.5 (1.4)	25.6 (3.2)	43.3 (1.3)	21.8 (1.0)	25.6 (2.1)
8)	SC-PCO/P-AS	47.2 (1.0)	23.7 (0.2)	32.5 (1.6)	48.6 (1.2)	23.9 (0.2)	34.8 (2.0)
9)	SC-WC/L	41.3 (0.4)	22.1 (0.3)	22.5 (0.6)	40.7 (0.6)	21.8 (0.5)	21.4 (1.0)
10)	SC-WC/L-AS	40.7 (0.3)	21.5 (0.4)	21.9 (0.7)	41.5 (0.8)	22.1 (0.6)	22.6 (1.3)
11)	SC-PCO/L	35.3 (0.5)	17.6 (0.8)	13.7 (0.9)	35.7 (1.0)	17.4 (1.8)	13.0 (1.6)
12)	SC-PCO/L-AS	33.7 (0.6)	14.2 (1.1)	10.7 (1.1)	35.0 (1.2)	16.8 (2.3)	12.6 (2.0)

- Abbreviations: primer SunCare 900 (SC), top-coat WoodCare UV (WC), top-coat PerlColor (PCO), water-repellent AquaStop (AS).
- Pigments in the top-coat layers: T = transparent, P = pine, L = larch.
- Mean values are from 18 measurements (see point 2.5).
- Numbers in parentheses are standard deviations.
- Samples before accelerated weathering had very similar *L**, *a**, and *b** color components.

SunCare 900 (SC), a transparent primer based on synthetic resins, organic UV light stabilizers, and 0.1 to 0.2% iodopropynyl butylcarbamate (IPBC) fungicide, was applied as the primer-coat layer. WoodCare UV (WC), a transparent (T), pine (P), or larch (L) pigmented coating based on oil-synthetic polymer resins, a nano-sized polyvalent metal AsS-(arsinoaryltio)-chelate complex for UV protection, hydrophobic additives, and IPBC fungicide, was applied in two layers. PerlColor (PCO), a transparent (T), pine (P), or larch (L) pigmented coating layer based on acrylate resin modified with oils, a nano-sized polyvalent metal AsS-chelate complex for UV protection, hydrophobic additives, and IPBC fungicide, was applied in two layers. Lastly, an AquaStop (AS) transparent, water-repellent and sunblock coating containing AsS-chelate was applied in one layer for one-half of the coating systems (Table 1).

Application of Coating Systems

Coating systems, in accordance with Table 1, were applied to spruce samples conditioned to 12 ± 1% moisture content. All coating systems consisted of one layer of the primer and two layers of the top-coat (WC or PCO) that had different pigmentation (T, P, or L). Six of the coating systems also contained one additional layer of the water repellent (AS). Individual layers were applied at 120 ± 10 g/m^2 with a manual, low-pressure, air spraying technique. Before the following layer was applied, the previously painted samples were sanded with 240-grit sandpaper. The drying time between consecutive paintings lasted 24 h at 20 °C and 65% RH. Reference samples were not painted.

Natural and Accelerated Weathering

Natural weathering of the painted and reference samples was carried out on metal stands at a 45° slope oriented to the South according to EN 927-3 (2006) standard outside the Technical University in Zvolen, Slovakia, Central Europe, at ca. 300 m above sea level. Weathering took place from the May 2, 2010 to April 30, 2014 for periods of 1, 4, 12, 26, 52, 104, 156, and 208 weeks. Samples were weather in a valley with many foggy days, smog, and high temperature differences between summer (35 °C) and winter (-25 °C). The color analyses from the 3rd year (156th week) and 4th year (208th week) are not present in this work, because they were not relevant for samples painted with transparent coating systems due to their severe destruction and loss of protective function.

Table 2. Exposure of Samples in Xenotest[1]: One Period, according to a Partially Modified Standard EN 927-6, Lasting 1 Week

Accelerated Weathering Both steps together lasted 1 week (168 h)			**Conditions**
1st Step		24 h	Temperature (45 ± 3 °C); Water Spray (off); UV (off)
2nd Step	A	2.5 h	Temperature (50 ± 3 °C); Water Spray (off); UV Irradiance (0.55 W/m^2 at 340 nm)
	B	0.5 h	Temperature (20 ± 1 °C); Water Spray (on); UV (off)
	48 sub-cycles (A+B): 48 x 3 h each = 144 h		

[1] In EN 927-6 (2006) standard, the prescribed parameters for test chamber equipped with a UV lamp (2nd step / A) are as follows: temperature = 60 ± 3 °C and UV Irradiance = 0.89 W/m^2 at 340 nm. However, test chamber used in our laboratory had a xenon lamp instead of a UV lamp.

Accelerated weathering of samples was performed in a Xenon Test Chamber Q-SUN Xe-1-S Xenotest (Q-Lab Corporation, USA) equipped with a 1800 W xenon lamp emitting UV and VIS light and spray section of redistilled water according to a partly modified EN 927-6 (2006) standard for 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, or 12 weeks, as shown in Table 2.

Color Analyses

The color components of the samples were measured with a CR-10 Color Reader (Konika Minolta, Japan) after 48 h of conditioning in the laboratory at 20 °C and 65% RH. Eighteen measurements were recorded for the naturally weathered samples (6 places with 3 replicates) and four measurements were recorded for the accelerated weathered samples (2 places with 2 replicates) – for each type of coating system.

The evaluation of color change was done using the CIE-*L*a*b** color system on the basis of the *L**, *a**, and *b** components, where *L** is the lightness from 100 (white) to 0 (black), *a** is the chromaticity coordinate (+, red; –, green), and *b** is the other chromaticity coordinate (+, yellow; –, blue). The total color difference of the samples between their weathered and initial state, ΔE^* (CIE 1986), was calculated from Eq. 1:

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

Statistical Analyses

Mean values and standard deviations were calculated for all results. The results from natural weathering were statistically analysed using the Duncan test (Statistica 12, StatSoft CR, Czech Republic). Effect of prolonged weathering was studied by correlation analyses (generated in Microsoft® Excel 2013; USA).

RESULTS AND DISCUSSION

Effect of the Initial Roughness of Wood

The effect of the initial roughness of the spruce wood surface on its color stability was not always clear. It was significant for some transparent coating systems (Figs. 1 and 2; Table 3), whereas it was clearly not significant for the pigmented coating systems (Figs. 3 and 4).

For the combination of transparent coating systems and natural weathering, a significant effect of higher roughness of wood on greater total color differences ΔE^* was determined only using PerlColor (PCO) top-coat layer, with or without water-repellent AquaStope (AS) (Table 3 – see Duncan 1, *i.e.*, the Duncan's statistical test No. 1 assessing effect of the initial wood roughness). This result could be explained by a hypothetical opacity in the presence of: (1) the rougher wood; (2) the top-coat layer PCO containing oil modification agents; and (3) the absorption of dark impurities.

Effects of the Top-coat Layer Type and the Presence of Pigments

Due to the presence of transparent coatings, the initial *L** and *a** color components of the native spruce samples changed only slightly, whereas the *b** component increased from *ca.* 19.5 to *ca.* 31 (Table 1), *i.e.*, the spruce samples obtained a more yellowish shade. On the other hand, in the presence of lightly-pigmented top-coat layers (pine or larch) the spruce wood became darker and more reddish shade; the *L** component decreased from 84 to *ca*. 40 and the *a** component increased from 4 to *ca*. 20 (Table 1).

Then, during weathering processes, smaller differences in the chemical composition of the transparent or pigmented top-coat layers of WoodCare UV (WC) versus PerlColor (PCO) were not dominant for the color stability of the painted spruce wood (Figs. 1 through 4; Table 3).

Table 3. Mean Values, Standard Deviations, and Duncan's Test of Significance for Three Effects[1] on the Total Color Differences ΔE^* of Norway Spruce (*Picea abies*) Samples at the End of Natural Weathering

Coating System	**ΔE^* - Total Color Difference at the End (104th Week) of Natural Weathering**							
	Pigmentation of Top-coats & Roughness of Wood							
	T (transparent)				P (pine)		L (larch)	
	Rough wood		Smooth wood		Rough wood		Rough wood	
		Duncan No. 1 2 3		Duncan No. 1 2 3		Duncan No. 2 3		Duncan No. 2 3
Reference – Native wood	40.1 (1.6)	d	41.1 (1.6)	-				
SC[2]-WC	20.2 (2.0)	d - -	20.4 (2.2)	- - -	4.7 (1.4)	- -	9.7 (1.5)	- -
SC-WC-AS	22.0 (1.6)	d - d	21.9 (1.7)	- - d	9.1 (1.9)	- a	7.2 (1.6)	- c
SC-PCO	25.8 (9.6)	b b -	20.3 (5.8)	- d -	6.1 (1.7)	d -	9.6 (2.5)	d -
SC-PCO-AS	28.5 (6.7)	a a c	21.8 (4.5)	- d d	15.4 (3.3)	a a	6.9 (2.0)	d c

[1] Duncan's Tests: (No. 1) Initial Roughness of Wood, *i.e.*, Rough versus Smooth; (No. 2) Type of Top-coat Layer, *i.e.*, PerlColor (PCO) versus WoodCare UV (WC); and (No. 3) Application of Final Water Repellent AquaStop (AS), *i.e.*, its Use versus Non Use

[2] SC (SunCare 900) is a transparent primer layer.

Mean values are from 18 measurements; Numbers in parentheses are the standard deviations; Duncan´s tests: 99.9% significance level (a); 99% significance level (b); 95% significance level (c); and less as 95% significance level at $p \geq 0.05$ (d).

In outdoor exposition a similar continual darkening and increased total color difference usually occurred for samples with transparent WC or PCO top-coat layers (Fig. 1). This result for ΔE^* was confirmed for samples with smooth surfaces by the Duncan test (Table 3 – see Duncan 2, *i.e.*, assessing effect of the type of top-coat layer PCO versus WC). Nevertheless, in the Xenotest, samples painted with transparent WC top-coat layer first became lighter (after the first 3 weeks the ΔL^* was positive – probably due to extraction or destruction of some darker additives – see also smaller initial L^* for samples with WC comparing to PCO in Table 1) and only later acquired a darker shade, while samples painted with transparent PCO top-coat layer were immediately more darkly stained (Fig. 2). A different scenario occurred for samples painted with pine or larch pigmented WoodCare UV (WC) or PerlColor (PCO) top-coat layers. The light pigments significantly improved the color stability of the painted spruce samples (Figs. 3 and 4; Tables 3 and 5).

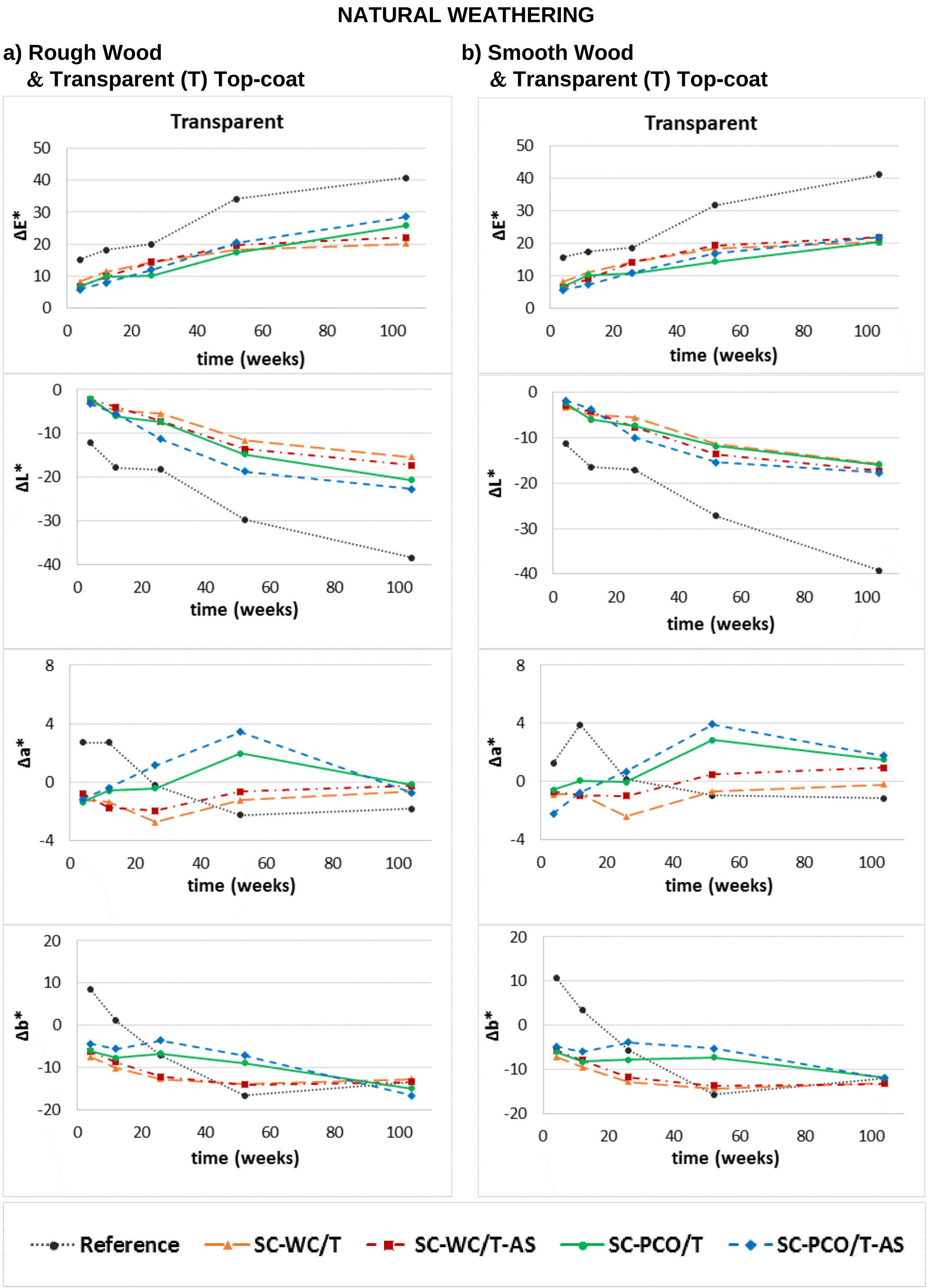


Fig. 1. Color changes – a) of Rough; b) of Smooth Norway spruce (*Picea abies*) samples painted with coating systems containing the transparent (T) top-coat layer – during natural weathering

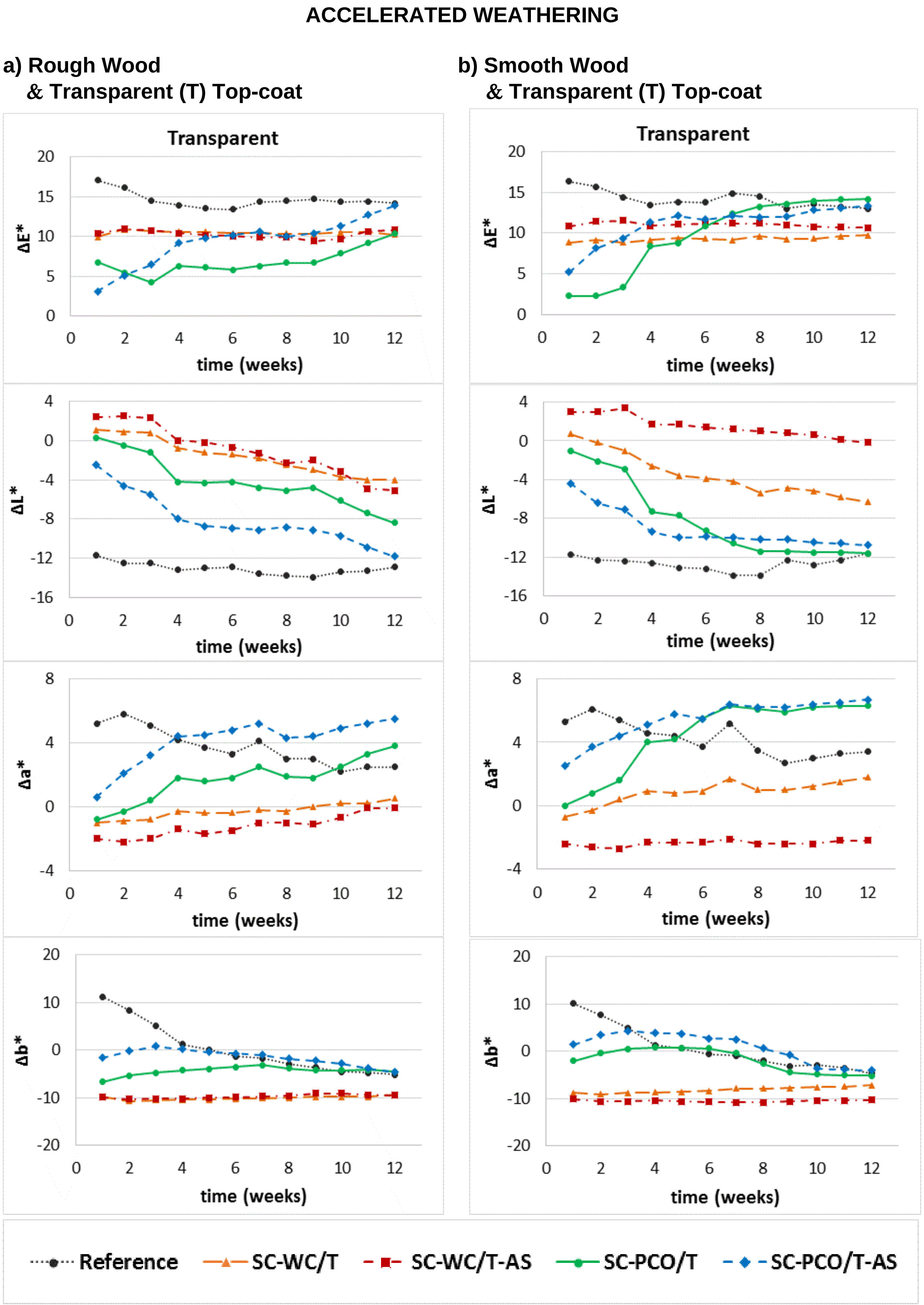


Fig. 2. Color changes – a) of Rough; b) of Smooth Norway spruce (*Picea abies*) samples painted with coating systems containing the transparent (T) top-coat layer – during accelerated weathering

Lower color differences ΔL^*, Δa^*, Δb^*, and ΔE^* established during weathering were clearly observed for samples painted with pigmented coating systems in comparison to transparent ones (Figs. 1 and 3). For example, after 104 weeks of natural weathering (Figs. 1 and 3), the ΔL^* and ΔE^* values of reference – native samples ("Rough" and "Smooth" on average: $\Delta L^* = -38.8$; $\Delta E^* = 40.6$) due to transparent coating systems decreased only two times ($\Delta L^* = -17.9$; $\Delta E^* = 22.6$), but several times more due to pigmented coating systems ($\Delta L^* = -2.4$; $\Delta E^* = 8.0$).

NATURAL WEATHERING

a) Rough Wood
& Pine (P) or Larch (L) Top-coat

b) Smooth Wood
& Pine (P) or Larch (L) Top-coat

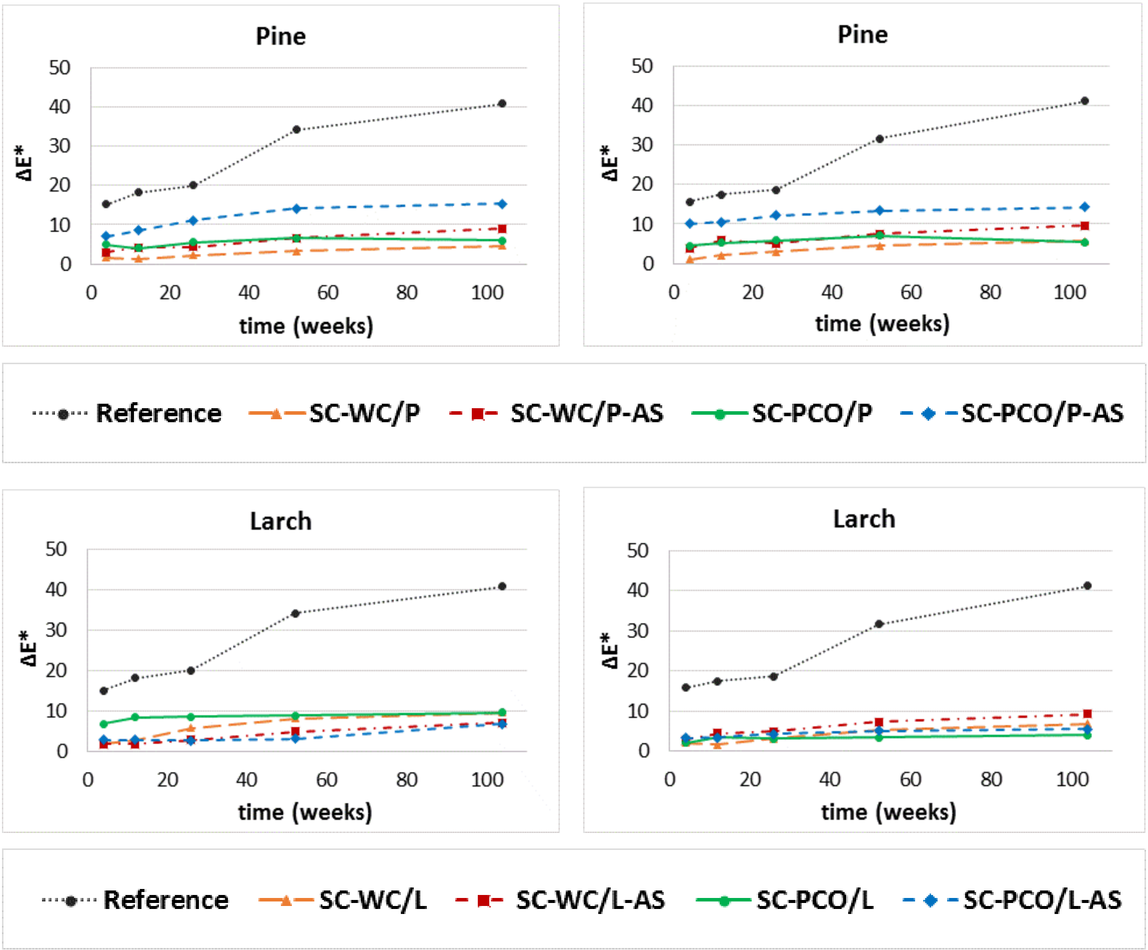
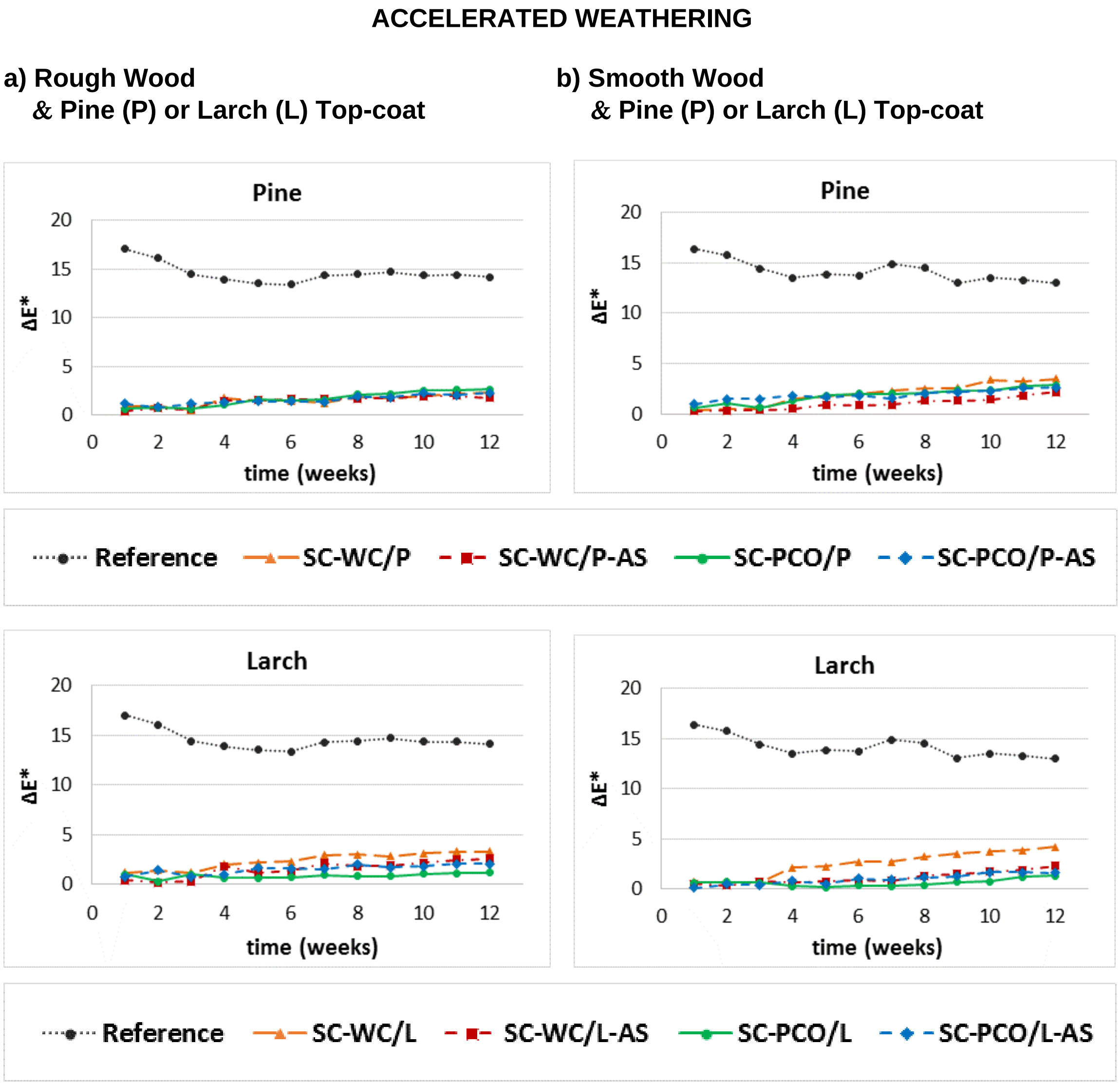


Fig. 3. Total color differences $\Delta E^*$of a) Rough and b) Smooth Norway spruce (*Picea abies*) samples painted with coating systems containing the pine (P) or larch (L) pigmented top-coat layer during natural weathering

A similar good color stabilization effect of pigments was seen for accelerated weathering conditions, *i.e.*, after 12 weeks of Xenotest, when the ΔL^* and ΔE^* values of reference – native samples ($\Delta L^* = -12.3$; $\Delta E^* = 13.6$) decreased slightly for samples painted with transparent coatings ($\Delta L^* = -7.3$; $\Delta E^* = 11.6$), but markedly for samples painted with pigmented ones ($\Delta L^* = -0.5$; $\Delta E^* = 2.4$). Results related to the color stabilization effect of pigments were confirmed as well as by the Duncan test, always with $p \geq 0.05$ between the total color differences of samples painted with transparent and pigmented coatings.


ACCELERATED WEATHERING

a) Rough Wood & Pine (P) or Larch (L) Top-coat

b) Smooth Wood & Pine (P) or Larch (L) Top-coat

Fig. 4. Total color differences ΔE^* of a) Rough and b) Smooth Norway spruce (*Picea abies*) samples painted with coating systems containing the pine (P) or larch (L) pigmented top-coat layer during accelerated weathering

Generally, in accordance with other works (*e.g.*, Evans and Chowdhury 2010; Reinprecht and Pánek 2013), it was confirmed that the pigments in coatings significantly increased the color stability of painted wood surfaces.

Effect of the Final Water Repellent

Application of the final water repellent AquaStop (AS) with one-half of the coating systems (Table 1) did not significantly affect the color stability of painted spruce samples (Figs. 1 through 4). This observation was, in most cases, confirmed by the Duncan´s test for the total color differences ΔE^* of naturally weathered samples having or not having the final AquaStop (Table 3 – see Duncan 3, *i.e.*, the Duncan's test No. 3 assessing effect of the final AquaStop layer use versus non use).

For samples painted with transparent top-coat layers, the AquaStop layer sometimes caused surface darkening (Figs. 1 and 2; see ΔL^* values), and also partly, though not significantly, increased their total color differences ΔE^* (Table 3).

One interesting result was that the AquaStop layer applied on samples with the pine pigmented top-coat WC or PCO layer provoked significantly greater color changes at beginning of natural weathering; this was probably due to specific photochemical or other reactions in the coating system "top-coat layer with pine pigment & water repellent" (Fig. 3), at which these color differences remained to the end of test (Table 3).

Effect of Prolonged Weathering

With prolonged weathering, the total color differences ΔE^* of the painted spruce samples became more positive (Figs. 1 through 4). Samples with transparent coatings typically had the greatest color changes at the beginning of weathering (Figs. 1 and 2). Samples with pigmented coatings had as well as an intensive color changes at the beginning of outdoor exposure carried out in an industrial zone (Fig. 3), however, in Xenotest their color stability during the first weeks of weathering was evidently better (Fig. 4).

In the case of natural weathering, samples treated with transparent coating systems underwent, in the first four weeks, a rapid change of Δb^* to negative values in the range from -4 to -8, and a moderate change of ΔL^* and Δa^* also to negative values about ca. -2.5 and -1.5 (Fig. 1). The ΔE^* values were also relatively high in the first-weeks of natural weathering for samples with transparent and pigmented coating systems, respectively (Figs. 1 and 3; Table 5). A quick color changes (darkening and graying) of all painted samples in outdoor exposure can be explained by a quick absorption of dirt particles on their surfaces, which caused their dark blue-green shade.

For the accelerated weathering test, rapid changes of ΔL^*, Δa^*, Δb^*, and ΔE^* occurred within the first week of exposure for samples painted with transparent coatings (Fig. 2; Tables 4 and 5). On the other hand, the color stability of samples painted with pigmented coating systems (pine or larch pigment in the top-coat layer) was evidently higher, and ΔE^* usually after the first week did not exceed a value of 1, or after the 12th week a value of 3 (Fig. 4; Tables 3 and 5). A higher color stability of samples painted with pigmented coatings determined in the Xenotest comparing to the exterior (Table 5 – see evidently smaller parameters $/A + B/$ at the first week of accelerated weathering) can be explained by absence of pollutants having darker shades.

In the above mentioned context, when colors of samples exhibited large changes usually within the first days of weathering, it is clear that changes of ΔL^*, Δa^*, Δb^*, and ΔE^* with weathering prolongation (t) cannot be, mainly at using transparent coatings, expressed in an appropriate manner with the simplest linear correlation of the type "Δcolor-component = B x t". Nonlinear correlations, *e.g.*, exponential expressions, proved to be better based on the coefficients of determination (R^2); however, for a practice, they were too complicated. Therefore, to evaluate the experimental data, the following linear correlation (Eq. 2) was designed with a proviso that parameter B will be reflected only from the first week of weathering ($t = 1$) to defined weeks of weathering ($t = 104$ natural; $t = 12$ accelerated),

$$\Delta\text{color-component} = A + B \text{ x } t \tag{2}$$

where Δcolor-component is either ΔL^*or ΔE^* (changes of Δa^* and Δb^* were not always clear, so in this work, they were not tested by correlation analyses); t is time of weathering in weeks (in this work a maximum time $t = 104$ for natural weathering, and $t = 12$ for accelerated weathering); A is the initial parameter of color change ($t = 0$), which in theory of weathering should be zero ($A = 0$), but in this work, it is a defined value (see Tables 4 and 5); and $A + B$ (if $t = 1$) is a parameter for ΔL^* or ΔE^* in the first week of weathering.

Table 4. Linear Correlations "$\Delta L^* = A + B$ x t" with Coefficients of Determination (R^2) for Modelling a Weathering of Reference and Painted Norway Spruce Wood Samples having Different Initial Roughness

Coating System	**Natural Weathering**						**Accelerated Weathering**					
	(t = 1 to 104 weeks)						(t = 1 to 12 weeks)					
	Rough wood			Smooth wood			Rough wood			Smooth wood		
Reference	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2
Native wood	-13.1	-0.257	0.96	-11.5	-0.274	0.98	-12.3	-0.116	0.45	-12.6	-0.012	0.05
	ΔL^* = -12.30 – 0.266 x t						ΔL^* = -12.45 – 0.064 x t					
Transparent	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2
SC-WC/T	-3.0	-0.129	0.95	-3.2	-0.127	0.96	1.7	-0.514	0.97	0.4	-0.608	0.92
SC-WC/T-AS	-2.8	-0.153	0.92	-3.3	-0.148	0.92	3.6	-0.715	0.96	3.5	-0.307	0.92
SC-PCO/T	-3.0	-0.181	0.96	-3.7	-0.126	0.93	0.3	-0.693	0.90	-2.5	-0.902	0.76
SC-PCO/T-AS	-4.6	-0.196	0.89	-3.5	-0.158	0.83	-3.7	-0.680	0.85	-6.0	-0.478	0.72
	ΔL^* = -3.39 – 0.152 x t						ΔL^* = -0.35 – 0.612 x t					

"$A + B$" approximates the ΔL^* in the first week of weathering ($t = 1$).

Linear growths of ΔL^* (to negative values) and ΔE^* with the passage of time of natural weathering (according to Eq. 2, and hypothetically starting from its first week) were confirmed by high coefficients of determination R^2, which were usually *ca.* 0.9 (Tables 4 and 5). For the linear growth of $\Delta E^* = A + B$ x t, a less significant trend was observed only in the case of using coating systems with pigmented top-coat layer PerlColor (SC-PCO/P; and SC-PCO/L), when the values of R^2 were lower (0.44 or 0.09; and 0.65 or 0.57). This may be due to a smaller effect of the natural weathering prolongation on the total color difference ΔE^* (computed from ΔL^*, Δa^*, and Δb^*) of spruce painted with pigmented PerlColor (*i.e.*, parameter B was only 0.017 or 0.007; and 0.020 or 0.014) – (see Table 5).

Similar trends of linear growths of ΔL^* (to negative values) and ΔE^* were found for a prolonged time of accelerated weathering of spruce samples painted with transparent coatings, *i.e.*, for ΔL^* with R^2 from 0.72 to 0.97, and for ΔE^* usually above 0.6 (Tables 4 and 5). However again, at determining ΔE^*, there in the case of small values of the "B" parameter (even negative numbers of B for SC-WC/T; and SC-WC/T-AS, *i.e.*, -0.009; and -0.042 or -0.043, which are valid only for the time-span of performed experiment), the values of R^2 were small as well (0.02; and 0.10 or 0.31).

Generally with prolonged weathering, the most rapid color changes of painted spruce samples could be observed at using the transparent coating systems. This is evident from the "*A*" and "*B*" parameters of the individual linear correlations, and also from the summary linear correlations expressed in the form of "$\Delta L^* = A + B$ x t" or "$\Delta E^* = A + B$ x t", separately for transparent, pine pigmented, and larch pigmented coating systems (Tables 4 and 5). For spruce samples painted with transparent coating systems could be seen a typical dark-graying at their natural weathering in an industrial zone (Fig. 1, Table 4), while at their accelerated weathering even a lightening during the first weeks of exposure (Fig. 2, Table 4).

Table 5. Linear Correlations "$\Delta E^* = A + B$ x t" with Coefficients of Determination (R^2) for Modelling a Weathering of Reference and Painted Norway Spruce Wood Samples having Different Initial Roughness

Coating System	**Natural Weathering**						**Accelerated Weathering**					
	(t = 1 to 104 weeks)						(t = 1 to 12 weeks)					
	Rough wood			Smooth wood			Rough wood			Smooth wood		
Reference	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2
Native wood	15.0	0.267	0.93	14.2	0.269	0.96	15.5	-0.141	0.24	15.6	-0.227	0.58
	$\Delta E^* = 14.60 + 0.268$ x t						$\Delta E^* = 15.55 – 0.184$ x t					
Transparent	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2
SC-WC/T	10.0	0.112	0.84	9.8	0.116	0.85	10.5	-0.009	0.02	8.9	0.064	0.68
SC-WC/T-AS	8.7	0.147	0.84	8.3	0.150	0.85	10.5	-0.042	0.10	11.3	-0.043	0.31
SC-PCO/T	6.5	0.189	0.99	7.4	0.127	0.97	4.5	0.358	0.62	6.1	0.582	0.54
SC-PCO/T-AS	5.8	0.230	0.97	6.0	0.163	0.95	4.2	0.797	0.87	7.5	0.560	0.73
	$\Delta E^* = 7.81 + 0.154$ x t						$\Delta E^* = 7.94 + 0.283$ x t					
Pine pigment	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2
SC-WC/P	1.3	0.033	0.95	1.5	0.045	0.92	0.7	0.135	0.76	0.1	0.302	0.96
SC-WC/P-AS	3.0	0.060	0.98	4.3	0.052	0.91	0.6	0.131	0.75	0.0	0.164	0.94
SC-PCO/P	4.8	0.017	0.44	5.3	0.007	0.09	0.3	0.205	0.96	0.5	0.199	0.92
SC-PCO/P-AS	8.1	0.081	0.84	10.4	0.042	0.87	0.8	0.118	0.87	1.1	0.125	0.87
	$\Delta E^* = 4.84 + 0.042$ x t						$\Delta E^* = 0.50 + 0.172$ x t					
Larch pigment	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2	*A*	*B*	R^2
SC-WC/L	2.6	0.077	0.86	1.7	0.053	0.92	1.0	0.214	0.91	0.2	0.346	0.94
SC-WC/L-AS	1.5	0.057	0.98	3.3	0.060	0.94	0.2	0.214	0.83	0.0	0.161	0.89
SC-PCO/L	7.7	0.020	0.65	2.6	0.014	0.57	0.6	0.039	0.33	0.2	0.057	0.32
SC-PCO/L-AS	2.1	0.041	0.84	3.4	0.022	0.83	0.8	0.110	0.74	0.0	0.140	0.93
	$\Delta E^* = 3.11 + 0.043$ x t						$\Delta E^* = 0.40 + 0.160$ x t					

"$A + B$" approximates the ΔE^* in the first week of weathering (t = 1).

Connections between the Natural and Accelerated Weathering

The basic aim of accelerated weathering of wood or other materials is to achieve similar changes in their esthetical and technical properties in a shorter time – color, strength, hardness, wettability, *etc*. – than through natural weathering (Creemers *et al.* 2002, Sandak *et al.* 2013, Grüll *et al.* 2014a, Jankowska and Kazakiewicz 2014). A climatic index – composed of global irradiation, total precipitation, number of days with more than 0.1 mm precipitation, *etc*. – could be a helpful tool for looking at the relationships between different atmospheric conditions. However, at this time more round-robin tests and practical experience point to the fact that results from laboratory weathering can hardly be compared to results from outdoor weathering (Creemers *et al.* 2002; Rüther and Jelle 2013). Similarly, results achieved in this work give only a limited view of required accelerated weathering times by which similar color changes as seen with natural weathering could be induced.

In this work connections between accelerated and natural weathering, coming from the CIE-$L^*a^*b^*$ color system, were analyzed separately for two time intervals: (1) from the zero to first week – using the "$A + B$" parameter in the Eq. 2, *i.e.*, for $t = 1$; (2) from the first week to the twelfth week in Xenotest, or to the 104th week in exterior – using the initial "$A + B$" parameter and also the directive "$B \text{ x } t$" parameter in Eq. 2.

Using transparent coating systems, the darkening $-\Delta L^*$and the total color differences ΔE^* of painted spruce increased, especially within the first week ($t = 1$) of weathering (Tables 4 and 5 – see values of the "$A + B$" parameter; Figs. 1 and 2). The summary ΔE^* values were very high after the first week of aging in Xenotest, *i.e.*, $\Delta E^* = A + B \text{ x } t = 8.223$, and also after the first week of natural weathering, *i.e.*, $\Delta E^* = A + B \text{ x } t = 7.964$ (Table 5). This is in accordance with the work of Sharratt *et al.* (2009) who found the majority of color changes within the first 24 hours of accelerated aging in surfaces of the Scots pine. Generally, the high values of $\Delta E^* = ca.\ 8.0$ determined for spruce wood painted with transparent coatings already within the first week of natural or accelerated weathering were caused: (1) at natural weathering due to absorption of dark dirt with a blue-greening effect during the first days of exposure under a slope of 45° in an industrial zone (Fig. 1; Table 4), (2) at accelerated weathering due to significant transfer of UV-light through transparent coatings to lignin and hemicelluloses of spruce, which caused apparent changes in the chromaticity coordinates a^* and b^* (Fig. 2), while the effect of their darkening $\Delta L^* = A + B = -0.962$ was yet small (Table 4).

A different course of color changes was observed for spruce samples with pigmented coating systems. Within the first week of accelerated weathering in Xenotest, the samples painted with pigmented top-coat layers had better color stability, *i.e.*, the ΔE^* ($A + B$) for pine = 0.672, or larch = 0.56. On the other hand, with natural weathering, the greatest growth of the ΔE^* occurred due to absorbed dirt within the first week of outdoor exposure, when the ΔE^* ($A + B$) for pine = 4.882, or larch = 3.153 (Table 5).

Further, the first week was chosen as a hypothetical new beginning time for weathering, where for spruce wood with transparent coatings the ΔE^* changed only about two-times faster in Xenotest ($B = 0.283$) than outdoor ($B = 0.154$). However, spruce samples with pigmented coatings behaved otherwise, because the ΔE^* value changed four-times faster during the accelerated weathering ($B = 0.172$ for pine pigmentation, or 0.160 for larch pigmentation) when compared to natural weathering ($B = 0.042$ for pine pigmentation, or 0.043 for larch pigmentation) – as seen from the summary of linear correlations (Table 5). These results, related to different behaviors of transparent and pigmented coatings in "a hypothetical new beginning time of weathering" – starting from its first week – could be explained by processes ongoing already from time zero to the first week of weathering, *i.e.*, (1) outdoor in an industrial environment a rapid absorption of dirt on wood surfaces painted with transparent or pigmented coatings, (2) in Xenotest a rapid color change of wood surfaces only at using transparent coating systems.

CONCLUSIONS

1. Different initial roughness of spruce wood before painting had little or no impact on its color stability when subjected to weathering.

2. Minor differences in chemical compositions of the top-coat layers (WoodCare UV versus PerlColor) and use or non-use of a final water repellent (AquaStop) had in most cases no significant effect on the color stability of painted spruce wood.
3. There was a clear, positive color stabilization effect of the pine or larch light pigments present in the top-coat layer of coating systems.
4. When subjecting samples to natural weathering in an industrial zone at a 45° slope, the greatest color changes occurred within the first weeks with both transparent and pigmented coating systems; this was mainly due to the absorption of dark impurities and the rapid drop of the L^* chroma value.
5. When subjecting samples to accelerated weathering in Xenotest, the greatest color changes with transparent wood coatings already occurred within the first week, with significant changes in the chromaticity coordinates a^* and b^*. On the other hand, colors of the samples painted with pigmented coatings changed continually from the zero to the twelfth week.
6. With accelerated weathering can be modeled intensive natural weathering of light-wood species (*e.g.*, spruce) painted with transparent coatings. However, in light-wood species painted with light-pigmented coatings the accelerated weathering cause gradual color changes, while at natural weathering in an industrial zone the surfaces obtain quickly blue-grey shades.
7. In practice, the darkening and the total color differences with prolonged weathering (t) usually change exponentially. However, in a simplistic view, they in this work were evaluated by linear correlations using a hypothetical new beginning time of weathering – a time point when some of color changes apparently slowed down. Description of the color changes can then be performed: (1) from the zero time to a hypothetical new beginning time of weathering, – in this work the first week was chosen; and (2) by using linear correlation "Δcolor-component = $A + B \times t$", – in this work from the first week ($t = 1$) to the end of weathering.

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