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ARTIFICIAL WEATHERING OF MELAMINE-IMPREGNATED SCOTS PINE (*Pinus sylvestris*) AND LARCH (*Larix sibirica*) WOOD

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Abstract:
The utilization of wood material is a sustainable option in many applications but it has some weaknesses, such as susceptibility to degradation caused by weathering. This study investigates the effect of impregnation with an environmentally nontoxic agent on the weathering properties of two wood species, Scots pine (Pinus sylvestris) and larch (Larix sibirica). Test samples were treated with a melamine solution analyzed using scanning electron microscope (SEM) graphs and performance indicators in the color measurement. The results show that weathering caused differences among the tested materials. The melamine-treated larch species was a bit more stable under weathering degradation, especially in the case of the lightness of material. The study shows that the gloss of color has no a significant role in these material combinations and the exact comparison of artificial and actual weathering was not possible. Although the measured results varied, it was assumed that the color alteration of wood material depends on several factors, such as extractives, and may be challenging to explain.

Key words: *impregnation; Scots pine; larch; melamine; weathering.*

INTRODUCTION

The global concern for climate change and greenhouse gases (GHG) has grown in the past decades, and buildings play a remarkable role in these issues. For instance, according to the report of the Intergovernmental Panel on Climate Change (IPCC 2014), buildings as an economic sector account for over 18% of global GHG emissions. Therefore, the selection and life cycle durability of materials require attention from various sectors. Wood material is an alternative that can help to meet the requirements to mitigate climate change. In the future, this may lead to the remarkable growth of the wood-based products, creating also growing economies.

Wood is one of the most important structural materials due to its outstanding properties, such as its high ratio of strength to weight, renewability and ease of processing. In spite of several advantageous features, wood also has some inherent properties that require improvement. The upgrading of materials can be developed by wood modification, which has been accepted as an option for improving the quality of wood during it lifetime. One modification option is impregnation, where pressure and vacuum are utilized to fill the substrate of the wood with an inert material (Hill 2006). Wood is often used as an outdoor material, making it susceptible to environmental degradation.

An example of the inherent properties of wood is vulnerability to weathering, which leads to undesired outcomes like discoloration and reduction in strength. The weathering of wood depends on many factors, including solar radiation, moisture, temperature, oxygen, and air pollutants, but ultraviolet light (UV) is responsible for its primary photochemical process or oxidative degradation (Hon 2001). The surface of wood in outdoor applications is strongly exposed to UV radiation, regardless of whether the material is horizontal or vertical (Lahtela and Kärki 2018). Physical weathering tests are laborious and might include irregularities. Therefore, artificial weathering tests offer faster results in terms of color alteration in controlled repeatable conditions (Wypych 2018). The color of the wood might be a critical criterion in material selection, and color alterations and the rate of discoloration are an indicator of weathering intensity. The color of wood may vary from almost sapwood white to almost black, and characteristics depend on the chemical components of the wood that interact with light. The numerical representation of color can be derived based on trichromatic quality where any color consisting mixing of three other colors, such as color specification system L*a*b*. Changes in color may vary on the same wood surfaces due to different cell volumes and components. ΔE = 3 is the lowest value that can be distinguished by the naked eye, and ΔE = 25 refers to the most intensive wood discoloration (Hon and Minemura 2001).

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Although several studies have dealt with weathering performance, detailed studies on the color alterations of modified wood are lacking. Scots pine (*Pinus sylvestris*) and larch (*Larix sibirica*) are traditional and popular species on the North European market, and often used for outdoor applications. Previously, weathering properties have been measured for the above-mentioned treated species in actual use (Lahtela and Kärki 2018). Therefore, natural and artificial weathering are compared in this study. The effect of different weathering conditions on various species and their treatments is an interested topic for scientists and industries.

OBJECTIVE

The purpose of this study was to investigate the weathering resistance properties of melamine-treated solid wood material. The color variation was caused by the artificial weathering of two species, Scots pine and Larch, during exposure of 700 h. These two species are traditional and popular in outdoor applications. In addition, the results of artificial weathering are compared to the literature data of the same material after real-life conditions.

METHOD

Scots pine (*Pinus sylvestris*) and larch (*Larix sibirica*) wood samples were impregnated using a pressure technology. Before the impregnation, the samples were cut into pieces of 20×50×1000mm. The impregnation was carried out by using a registered pressure apparatus (YIT Teollisuus Oy), at a pressure of 1MPa for 120min. The melamine solution (Prefere 70 0592 L) was obtained from Dynea Chemicals Oy (Hamina, Finland). After impregnation, the samples were dried in an oven at 90°C for a day. The weight percent gain (WPG) of the impregnation was measured according to the following equation (Eq. 1):

$$WPG\,(\%) = \frac{Mm - Mu}{Mu} \times 100 \qquad (1)$$

where: *Mm* is the mass after impregnation and drying;
Mu is the mass (at room temperature) before impregnation.

The performance of the treated samples was evaluated by placing the samples into an accelerated weathering test chamber (Q-Sun Xe-3 Xenon Test Chamber) horizontally perpendicular to the exposure source. The accelerated weathering test included intervals of UV -light and water spray, based on the standard EN ISO 4892-2. The surface color was calculated before the test, after 25, 50, and 100h, and then after every 100h until the end of the test. The color measurements were carried out directly on the specimens' surfaces.

The surface colors of the examined materials were measured using the spectrophotometer Minolta CM-2500d (Konika Minolta Sensing Inc., Japan) that consists of a measuring head of 8mm, a 10° observer, and D65 illumination. The color measurement consisted of nine measurement points per treated species. The CIELAB color system *L*a*b** was used. It consist of a three-dimensional coordinate in the color space, measuring simultaneously including (SCI) and excluding (SCE) the influence of gloss in one measurement. The *L** represents the lightness coordinates, varying from 100 (white) to 0 (black). Correspondingly, *a** indicates the red (+a*) to green (-a*) coordinates, and *b** indicates the yellow (+b*) to blue (-b*) coordinates. The total color alteration (Eq. 2) and the alterations in individual colors were calculated using the following equations:

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

where ΔE^* describes the total color alterations in the L*a*b* color space without the direction
ΔL^* is the difference in lightness between the initial and measured situation
Δa^* is the difference between red and green from start to measured situation
Δb^* is the difference between yellow to blue from start to measured situation

The vertical surface and the cross-section of the treated materials were examined with the scanning electron microscope (SEM) Hitachi SU3500 with the following operation conditions: voltage 15.0kV, vacuum 100Pa, and magnification varying between 20 and 500. For the SEM evaluation, samples dimensions were 9×9×9mm.

RESULTS&DISCUSSION

Treatment

The average WPG values of melamine-impregnated Scots pine and larch were 24.93 and 5.57, respectively. The other potential values for the computation of wood impregnation might be, for instance,

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solution uptake (SU), relative solution uptake (RSU), and impregnation rate (I.R.), but the WPG has been found to be a valid indicator with a melamine solution (Lahtela and Kärki 2017). The standard deviations of WPG values were remarkably high with both species, 14.13 for Scots pine and 5.18 for larch, showing the heterogeneous nature of wood material. In the weathering test, the WPG results might be enough because the penetration of UV light into wood is generally no higher than 75*µ*m (Hon 2001).

Fig. 1 presents the effect of treatment, showing SEM micrographs of the cross-sections of the studied samples. Fig. 1A and Fig. 1D show the samples before the weathering exposure, where the effect of treatment is detectable. The melamine solution has penetrated the wood well, in spite of the growth direction. The surface of the melamine-treated Scots pine is perpendicular to the cross-section face – the so-called radial surface – while the surface of the melamine-treated larch is perpendicular to the core of the wood – so-called tangential surface.

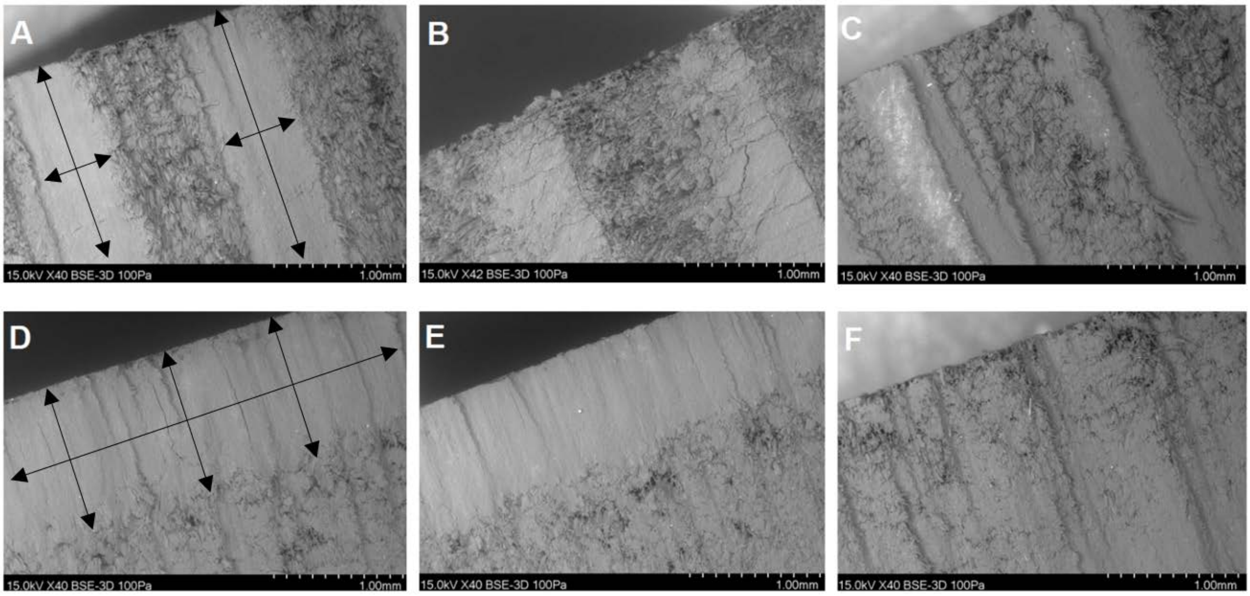

Fig. 1.
SEM figures from the melamine-treated (m-t.) Scots pine (A-C) and larch (D-F) cross sections before (A,D), and after 100h (B, E) and 700h (C, F) exposure of artificial weathering. The clear penetration areas of melamine solution are demonstrated by dark two-way arrows (A,D).

The different WPG results indicated that the wood species had a remarkable effect on the success of treatment. The wood material features, which will affect the impregnation results, are generally known to be the penetration direction and an aspiration, as well as the structural variables of the wood parts, such as specific gravity and heartwood (Stamm 1946). For example, the uptake of solution has been six times higher with pine sapwood compared to pine heartwood (Žlahtič et al. 2017). Depending the wood species, significant penetration occurs longitudinally through tracheids or vessels (Tondi et al. 2013). Radial penetration is again better compared to tangential penetration in pine due to ray tracheids (Scholz et al. 2010). The influencing feature of the impregnation solution is viscosity, which depends on the relative molecular weight and the concentration of the solution. The penetration of the solution increases with decreasing viscosity and increasing molecular weight (Kučerová 2012).

Weathering

The total color alteration (*ΔE*) and the parameters of specific color alterations (*ΔL*, Δa*, Δb**) are presented as an X Y scatter chart in Figs. 2 and 3, to which a trend line has been added between the points using a five-order polynomial. The total color alterations of both species were quite similar. At first, the color alteration increased significantly until 100h, remaining stable for the next hundreds hours. The significant increases in color alteration happened after 400-500h of exposure. In more detail, the color alteration (*ΔE*) of melamine-treated (M-t.) Scots pine had a slightly higher color change compared to the melamine-treated (M-t.) larch during the entire test period, as Fig. 2 shows. In addition, the color alteration of m-t. Larch was somewhat coherent, while the direction of *ΔE* of m-t. Scots pine changed more often.

The 700h artificial weathering test did not reach exactly the same color alteration as after 35 months of actual use (Lahtela and Kärki 2018), but when computational deviation is taken into account, 500h of artificial weathering corresponds to the color alteration of 35 months in real-life weathering degradation. The 700h test seems to be long enough to introduce differences because in the study of Huang et al. (2012A), the most significant color changes took place before 400h, whereas changes were minor during the final 336-1512h. In addition, the similar non-linear alterations within colors were detectable in the same study (Huang et al. 2012A).

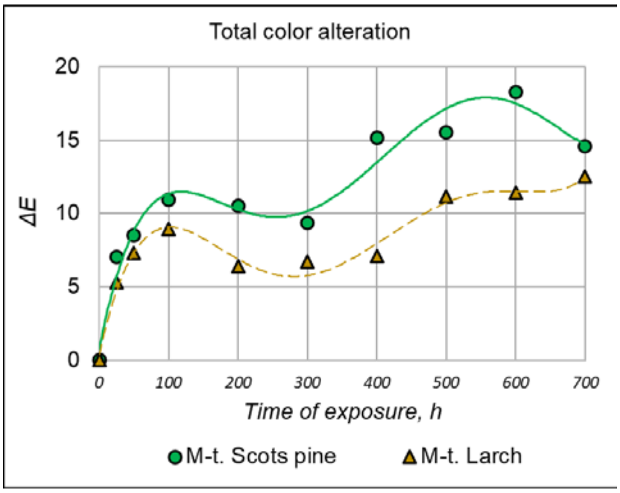

Fig. 2.
Total color alteration (ΔE) of studied materials, melamine-treated (m-t.) Scots pine and larch, during the test period.

Fig. 3 shows that lightness (*ΔL*) is the most remarkable parameter in color measurement for both wood species. The same observation was also made by Huang et al. (2012B). The color change between red and green (*Δa*) was nearly identical and minor for both melamine-treated (m-t.) species but the color change from blue (-*b*) to yellow (+*b*) was stronger in m-t. Scots pine than in m-t. Larch after 200h of exposure. However, the transformation was not significant over the entire 700h period.

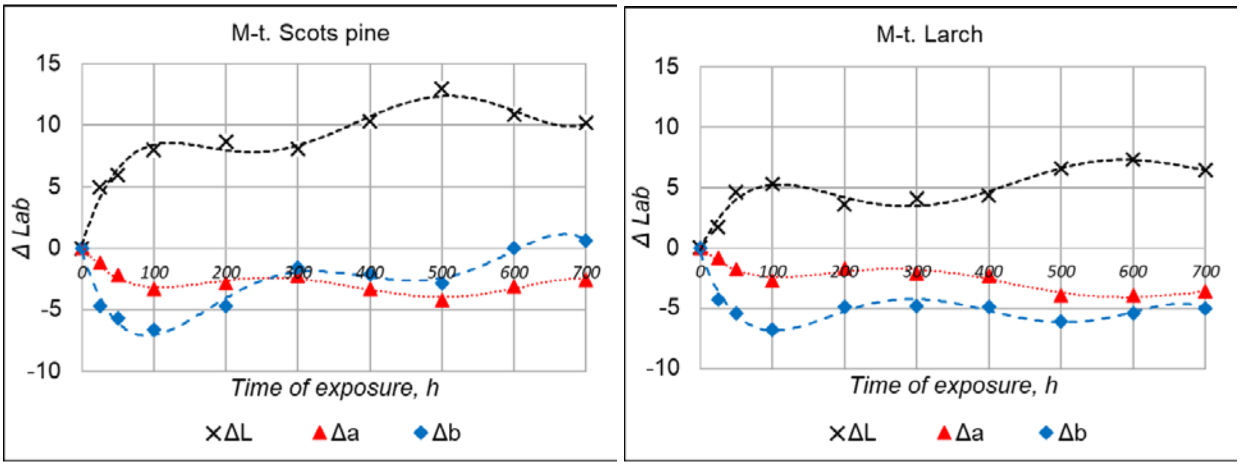

Fig. 3.
Color alteration of melamine-treated (m-t.) Scots pine and larch, according to all parameters (ΔL, Δa, Δb) during the test period.

Table 1 shows that gloss has no significant role in wood color measurement because most often the differences between included (SCI) and excluded (SCE) gloss were less than ±2.00 percentage. The highest numerical percentage difference (-971.05) was achieved on the yellow-blue axis (*Δb*) of Scots pine after 600h, but the measured values of *Δb* with SCI and SCE were 0.04 and -0.37, respectively. This is insignificant, as nothing below *ΔE* = 3 is perceptible to the naked eye (Hon & Minemura 2001). Another high numerical percentage difference (-134.92) occurred in the lightness (*ΔL*) of larch after 25h but also then, the difference between *ΔL* with SCI and SCE was minor: 1.70 and 3.99, respectively. The results showed that the gloss value has no significant role in m-t. Scots pine and m-t. Larch, and it is not a necessary measurement parameter.

Table 1

Percentage differences of measured colors between included (SCI) and excluded (SCE) gloss

Parameter	25 h	50 h	100 h	200 h	300 h	400 h	500 h	600 h	700 h
ΔE Scots pine	-3.23	-1.61	-1.79	-1.70	-0.84	-0.69	-0.86	-0.45	-0.01
ΔE Larch	-13.05	-1.50	-1.72	-2.06	-2.01	-2.57	-1.09	-0.67	-0.58
ΔL Scots pine	-0.65	1.01	-0.08	-0.42	-0.39	-0.76	-0.38	0.40	-0.01
ΔL Larch	- 134.92	-1.20	-1.87	-2.25	-2.28	-4.12	-2.41	-2.19	-1.78
Δa Scots pine	-0.65	1.01	-0.08	-0.42	-0.39	-0.76	-0.38	0.40	-0.01
Δa Larch	-2.32	-0.06	-0.70	-0.58	-0.69	-1.11	-0.82	-0.70	-0.55
Δb Scots pine	-6.60	-4.76	-4.89	-7.15	-20.88	-18.84	-13.62	- 971.05	58.59
Δb Larch	-2.94	-1.83	-1.79	-2.08	-2.30	-2.46	-2.26	-2.72	-2.67

Figs. 1 and 4 also present the weathering effects of Scots pine and larch showing SEM micrographs from the cross-sections and surfaces of the samples. The effects of weathering can be seen in the samples after exposure. The SEM micrographs of the cross-sections show cracks on the surface of both samples after 700h (Figs. 1C and 1F) of exposure. However, after 100h of exposure, the cracks are detectable only in m-t. Scots pine (Fig. 1B), whereas m-t. Larch (Fig. 1E) seems similar to the non-weathered m-t. Larch sample (Fig. 1D). Cracking is also visible on the SEM surface sample after 700h (Figs. 4C and 4F) compared to the SEM samples before the test (Figs. 4A and 4D).

The previously described effect of different surface sections of Scots pine (radial) and larch (tangential) might have an influence on anatomical structure changes. Bordered pits could be observed in the radial surfaces but could not easily be found on the tangential surfaces. After 500h of UV exposure, half-bordered pits were damaged on the radial surfaces (Hon 2001).

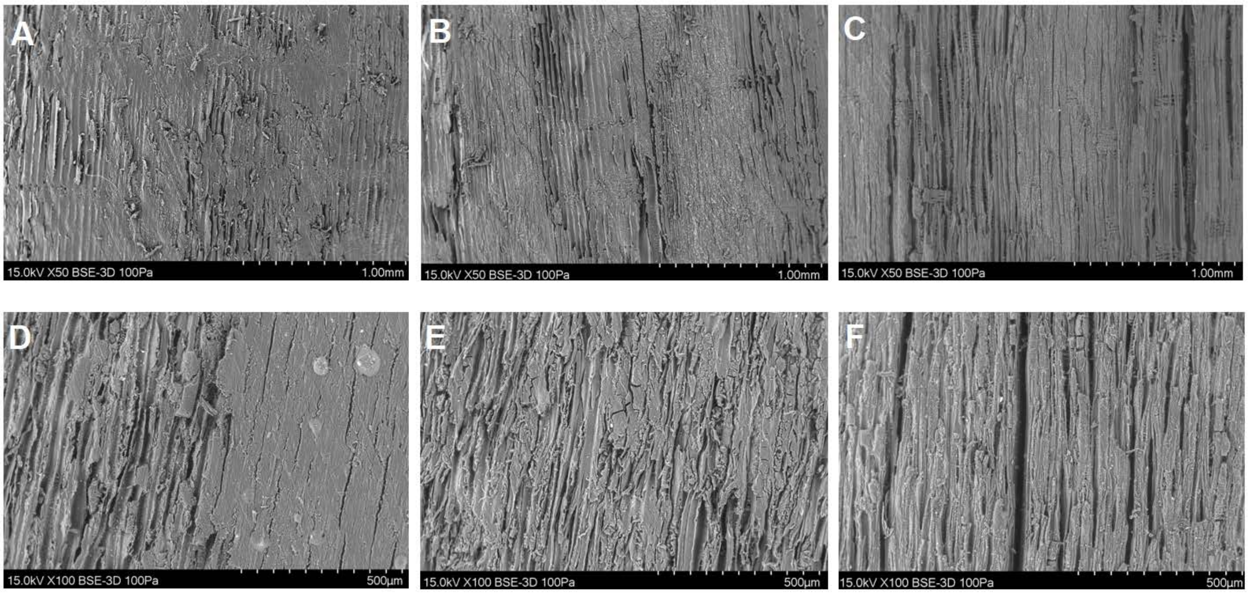

Fig. 4.
SEM figures from melamine-treated (m-t.) Scots pine (A-C) and Larch (D-F) surfaces before (A,D), and after 100h (B, E) and 700h (C, F) exposure of artificial weathering.

Fig. 5 displays the degree of weathering in snapshots taken of the surface of samples during the weathering test. The snapshots were not taken in a standard mode, but alterations can be detected from the images: especially the Scots pine sample was clearly lighter after 700h compared to previous ones.

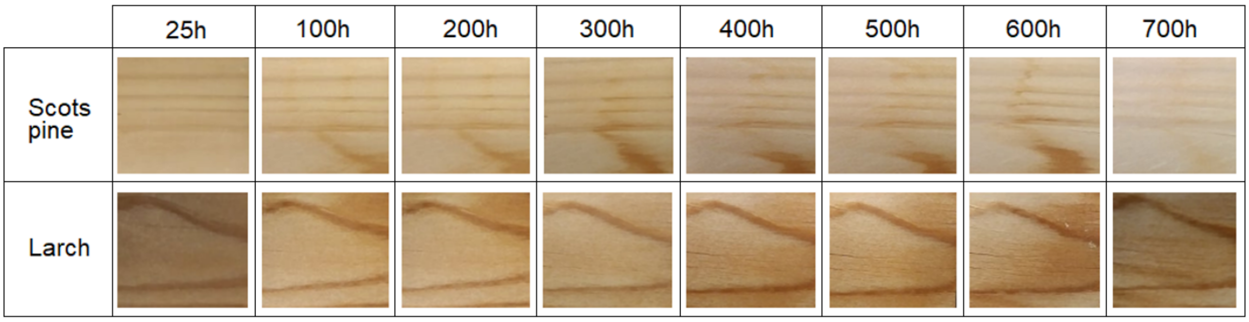

Fig. 5.
Images of surfaces of melamine-treated (m-t.) Scots pine and larch during various time periods in artificial weathering exposure.

Different wood components, such as cellulose, lignin, and extractives, react to weathering differently. Lignin is quite sensitive to UV radiation and may start to degrade within a short time (Williams 2010). A melamine solution could improve resistance to color changes because melamine resin could act as a diffusion-hindering matrix and delay the formation of lignin decay (Müller and Steiner 2010). Huang et al. (2012B) have stated that the discoloration of wood is caused by the combination of the photodegradation of lignin and extractives. Pine and larch have almost the same content of cellulose and lignin (Rowell et al. 2013), and thus, extractives have the most remarkable role in the color change, as Chang et al. (2010) have stated, which might explain the differences between species. The amount of extractives is low in both wood species compared to other wood components, but extractives characterize each species chemically and are the predominant contributor to wood color (Umezawa 2001). Especially water-soluble extractives give species their attractive color and can be carried onto surfaces through wetting-drying cycles (Williams 2010). Larch characteristically has an abundance of extractives (Kretschmann 2010). The effect of extractives might decreased over time due to the evaporation of volatile components because of aging (Beyer et al. 2018). Extractives have proven antioxidant properties, which can inhibit the discoloration of wood (Ayadi et al. 2003).

The weathering of wood depends on many environmental factors, such as the combination of solar radiation, moisture, temperature, oxygen and air pollutants, while in artificial weathering, the factors are well controlled (Hon 2001). The comparison of artificial weathering with natural exposure tends to be slightly ambiguous due to differences between northern and tropical climates, such as the equator. The degrading effect of xenon lamps is greater compared to sunlight (Tolvaj and Mitsui 2010), but xenon arc filters demonstrably create the best correlation with natural weathering (Wypych 2018). One difference between artificial and natural conditions are external factors in natural exposure, such as rain, which together with impurities might decrease strength retention (Hon 2001).

CONCLUSIONS

In this study, the weathering performance of melamine-treated Scots pine (*Pinus sylvestris*) and larch (*Larix sibirica*) was studied by performing artificial weathering tests. It can be stated that the treatment and weathering performance of the treated wood are dependent on the wood components and their heterogeneity. Extractives in wood might hamper the penetration of a solution but also restrain color changes due to the weathering exposure.

This study demonstrated that lightness is the most significant parameter in color changes during artificial weathering exposure. Substantial alterations in colors were measured during the first 100h of exposure, when approximately three quarters of the measured color changes occurred, whereas color changes after 100h until the end of the test were not as remarkable. In spite of the changes in color, artificial weathering and actual exposure could not be compared exactly and is subject of further studies.

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