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NATURAL WEATHERING OF LINSEED OIL PAINTS

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ABSTRACT: Wood deterioration in exterior applications is an important issue not only for the preservation of cultural heritage. Traditional oil paints have been applied to wooden surfaces for centuries and are able to last for 50 to 100 years if maintained regularly. However, there are not many studies that have examined the behaviour and durability of these paints in outdoor conditions. Eight paint systems with linseed oil as a binder and differing in pigment type and number of coats were investigated. Painted specimens were exposed outdoors and monitored for five years during which changes in surface temperature, moisture content, gloss, colour and chemical composition were recorded. The paint colour lightness affected the final surface temperature, which in turn affected moisture intake and release. The initial gloss of the opaque paints decreased significantly over three years to match the level of semi-transparent ones, which had been dull from the beginning. The colour of opaque paints was relatively stable over the period of exposure, except for the white paint, where the contrast with the accumulating dirt and mould was clearly visible. In contrast, semi-transparent paints lost most of the pigmented layer and the resulting colour was significantly influenced by discolouration of exposed wood. The Attenuated total reflectance-Fourier transform infrared (ATR-FTIR) analysis confirmed the gradual degradation of linseed oil in the surface layer of opaque paints, while in the semi-transparent systems, pigment loss was demonstrated and spectra corresponding to wood began to appear.

KEYWORDS: pigments, coating, discolouration, ATR-FTIR spectra, surface temperature

1 – INTRODUCTION

Wooden surfaces deteriorate when kept outdoors due to atmospheric corrosion, in which there is gradual erosion of the outer layer due to the combined effects of various weather conditions. The most significant influences are UV radiation and water, supplemented by temperature fluctuations, mechanical abrasion and air pollution. Wood weathering is manifested by discolouration, the formation of cracks, surface roughening and even relief texture [1]. Various types of coating, such as paints, varnishes and stains, are applied to protect wood against weathering. By reducing wood surface degradation and its loss, with proper maintenance coatings can extend the life of exterior wooden constructions significantly. Linseed oil paints have been in use in interior and exterior applications for a long time; the oldest written reference to outdoor use indicates that linseed oil was already in use as a binder in the 16th century [2]. Pigments – mainly naturally occurring inorganic compounds such as iron oxide, chrome yellow and zinc oxide – were added to the raw linseed oil to create colour. The advantages of linseed oil are its low price, wide availability and reactivity to form a solid protective film, this latter property being due to the large number of double bonds in the fatty acids present in the oil [3, 4]. Traditional oil

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paints allow wood to 'breathe' naturally, facilitating the movement of moisture from the wood to the air. Traditional oil paints are also simpler to maintain and renovate than modern coatings. A characteristic effect associated with paint as it ages is surface oxidation, where a greyish-white powder layer is formed, known as chalking [5]. During exterior exposure, linseed oil as a binder is chemically and physically degraded, primarily due to autoxidation and UV radiation, and pigment particles are released on to the surface [6]. The pigments included in the paint can significantly influence the chemistry of the paint from the point of view of its degradation, oxidation and stabilisation processes [7].

The use of traditional linseed oil paint declined in response to the explosive growth of the petrochemical industry in the second half of the 20th century and due to certain disadvantages like more demanding application, long drying time and related time consumption and labour costs. Oil paints were replaced by solvent-based, polyacrylate dispersion or alkyd paints, which solve some of these issues but do not look natural when used on historical wooden constructions. Šemjakin et al. have stated that traditional, natural-based coatings are now popular in the restoration of old buildings and also in new interiors [2], possibly because linseed oil is perceived as an 'ecological' material offering an environmentally friendly solution [8]. In conserving and restoring historically significant buildings, the general recommendation is to use original materials and processes. Yet oil paints haven't taken off in this area partly due to the lack of interest of profit-oriented companies who cannot see oil paint, with its disadvantages, being profitable, and partly because conservation professionals lack the information about the behaviour of traditional oil paints that would convince them that they are essential in historical building renovation.

The number of coats of paint or layers applied affects its performance, with more layers providing better protection [9]. In our study, we applied paint to the surface in either a single layer (semi-transparent) or multiple layers (opaque). A semi-transparent layer was used as short-term surface protection allowing the fresh wood to dry out easily; it also worked as a base layer [10, 11]. An opaque coating acts as long-term UV protection for the wood and also has an aesthetic function. In our project, we aimed to describe changes to linseed oil paint applied to wood outdoors over time and evaluate the influence of different pigments on paint durability.

2 – PROJECT DESCRIPTION

This research was supported by the 'Wooden structures prevention and maintenance for heritage conservation purposes' project NAKI III, reg. No. DH23P03OVV005, provided by the Ministry of Culture of the Czech Republic. We wish to thank the Department of Landscape Management, Faculty of Forestry and Wood Technology of Mendel University in Brno for providing the climate data used in this paper.

3 – EXPERIMENTAL SETUP

3.1 Preparing specimens

We prepared the specimens from one single Norway spruce log (*Picea abies* [L.] Karst.) by cutting boards of 30 mm thickness and drying them naturally to a moisture content of 16%. They were then planed to dimensions 600×140×20 mm. The connecting part of a board (200×140×20 mm) was used as a reference specimen. The measurements noted were taken in areas far from defects and with a radial texture (the growth ring orientation max 45°).

3.2 Preparing the coatings

The linseed oil paints were prepared by manually blending commercially available pigments (Tab. 1) with linseed oil (Leinölfirnis, Oelfabrik Schmidt GmbH; pre-polymerised). Two types of coating were tested:

1. One coat, wiped on with a cloth to leave the wood texture visible (semi-transparent; S)
2. Two coats to achieve full surface coverage (opaque; O); the second coat applied after 24 hours.

All surfaces of each specimen were coated in the same way, paint was applied with a brush. The spreading rate is provided in Tab. 1.

Table 1. Characterization of linseed oil paint systems

System	Colour	Pigment	Mass (g)[a]	Spreading rate (g/m^2)[b]
1	Red	Terra Pozzuoli	243	35/178
2	Green	Chromium oxide green	254	23/161
3	Yellow	Ferric oxide yellow	94	20/110
4	White	Titanium dioxide	250	38/197
5	Natural	None	–	–/–

[a]pigment mass in 100 mL of linseed oil

[b]semi-transparent/opaque

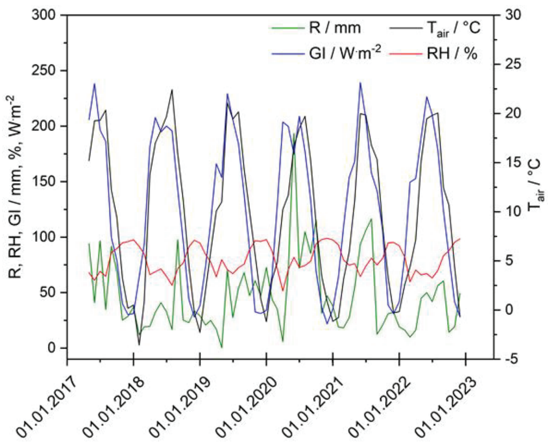

Figure 1. Climate data from exposure location (R – precipitation, T_{air} – air temperature, RH – air relative humidity, GI – global irradiance)

We made four replicates of each coating system and type. The coated panels were stored for several weeks at 20±2°C and 65±5% relative humidity before outdoor exposure.

3.3 Outdoor exposure

The specimens (painted and control unpainted) were exposed in May 2017 (rack exposure angle 45°, south orientation) in Útěchov (465 m a.s.l., 49°17'29.9"N 16°38'08.4"E). Climate data were recorded during the period of exposure and are shown in Fig. 1. The panels wood grain was oriented vertically. The performance of each coating was evaluated every year of exposure for up to six years. The reference specimens were stored in a dark conditioned interior during the whole experiment. Every year, we photographed the specimens, examined the surface and measured the colour. During every examination period, we cut a specimen from two exposed panels for laboratory analysis (only ATR-FTIR analysis results are presented here). The specimens were not cleaned before analysis to avoid any possible influence on the surface.

3.4 Temperature and moisture content

We determined the influence of the pigment type and colour on the surface temperature of the coating during summer using a Flir S65 thermocamera (distance from specimens 2.5 m, surface emissivity used for temperature correction). The temperature was measured on 5th August 2017 at 13:00 (air temperature 30.5 °C and relative humidity 39.5%). Only opaque paints were measured.

The mass of the specimens was measured before exposure and subsequently every month to observe moisture content changes during the year caused by rain and air relative humidity. The mass change was expressed in % according to (1):

$$\mathrm{CM} = \frac{m_w - m_0}{m_0} \cdot 100 \qquad (1)$$

where CM is change in mass, m_0 is specimen mass before exposure and m_w is specimen mass during exposure.

3.5 Measuring colour and gloss

We measured discolouration using a spectrophotometer (Spectro-guide 45/0 gloss, BYK-Gardner GmbH, 400–700 nm, 10° standard observer, D65 standard illuminant) and quantified colour changes using the CIELAB system, in which coordinates include the lightness of the colour (L*) and its location on the red-green axis (a*) and the yellow-blue axis (b*). We took colour readings before exposure and subsequently every year (2017–2022) during exposure at the same five locations along the length of each specimen. The paint was not cleaned in any way during exposure.

The paint gloss was determined using a gloss meter (MG268-F2, KSJ, China, 85° geometry, parallel to the wood grain) at three locations on each specimen before exposure and then every year for three years.

3.6 ATR–FTIR measurement

We obtained ATR-FTIR spectra of the coated specimens using a Nicolet iN10 spectrometer (Thermo Fisher Scientific Inc., USA) with an attached external module iZ10 equipped with a diamond ATR crystal. The spectra were recorded in the range of 4,000-525 cm^{-1} with a spectral resolution of 4 cm^{-1}.

4 – RESULTS

4.1 Surface temperature

The measurement of surface temperature showed significant differences between the paint systems (Tab. 2). Darker oil paint hues absorbed more energy from sunlight and their surfaces were hotter than both unpainted wood and specimens coated with white oil paint, whose surface was almost the same temperature as the air.

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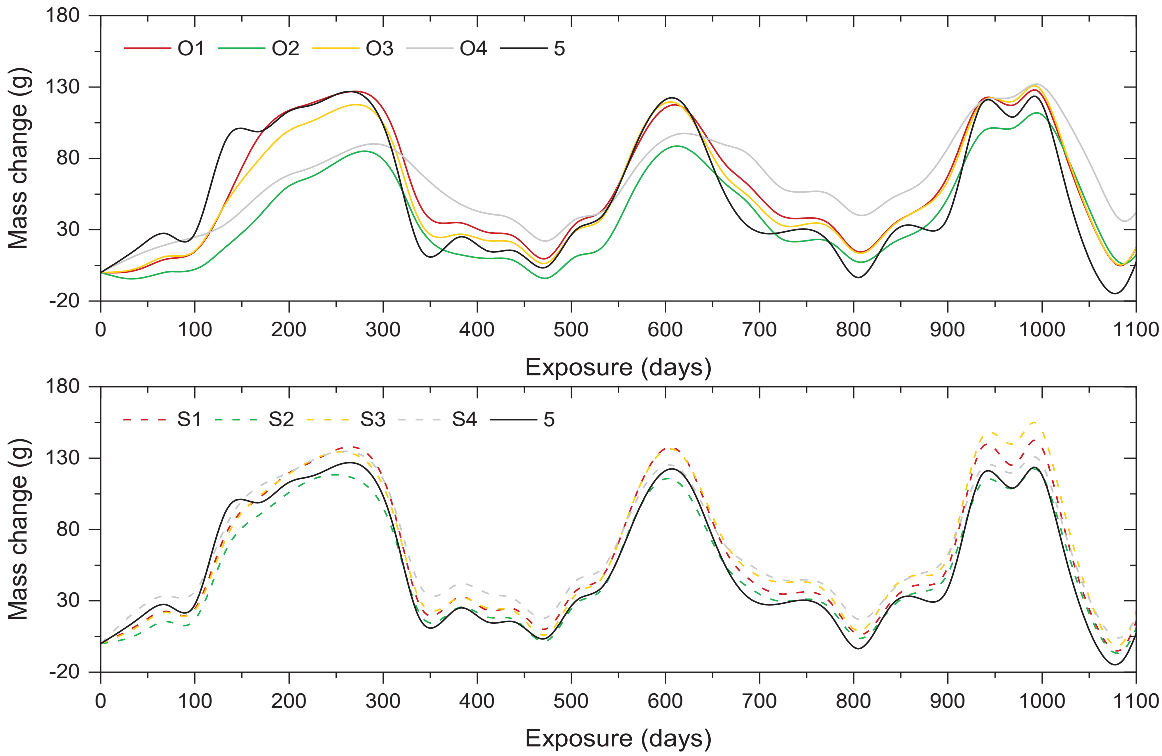

Figure 2. Moisture content change during three years of weathering (O – opaque, S – semi-transparent)

4.2 Moisture content

One of the fundamental roles of a paint coating is to control moisture movement in response to environmental change [12]. According to our results, the ability of oil paint to exclude moisture depends partly on the pigments it contains. While the red and yellow opaque paint systems provided no better barrier than the unpainted specimen, the white and green systems were able to protect wood against intensive water absorption (Fig. 2). The differences between paints had almost disappeared by the third year of exposure. The colour of the pigment probably also affected the rate of moisture release. While darker pigments lost moisture in summer months relatively quickly, the moisture content of white painted specimens was always the highest and changed slowly, which may have been caused by the temperature to which the specimen is heated by sunlight (Tab. 2). Darker colours absorb more sunlight, which is converted into heat and could accelerate specimen drying. The semi-transparent systems, regardless of pigment, more or less matched the behaviour of unpainted wood and allowed the wood to 'breathe' naturally (Fig. 2).

Table 2. Surface temperature

System	Colour	t_{min} (°C)	t_{max} (°C)	t_{mean} (°C)
1	Red	37.5	53.6	51.9
2	Green	55.6	63.6	61.7
3	Yellow	50.6	55.8	54.5
4	White	32.8	35.8	34.1
5	Natural	34.1	42.7	37.8

Table 3. Gloss change during three years of exposure (O – opaque, S – semi-transparent)

Colour	System	0	1	2	3
Red	S1	2.6	1.3	1.1	1.4
	O1	7.8	1.3	1.0	0.9
Green	S2	15.4	4.8	3.2	2.7
	O2	46.9	27.6	8.8	4.2
Yellow	S3	12.2	3.9	2.7	2.4
	O3	33.0	10.3	2.4	2.2
White	S4	4.8	4.0	2.2	3.0
	O4	45.0	27.8	11.0	3.7

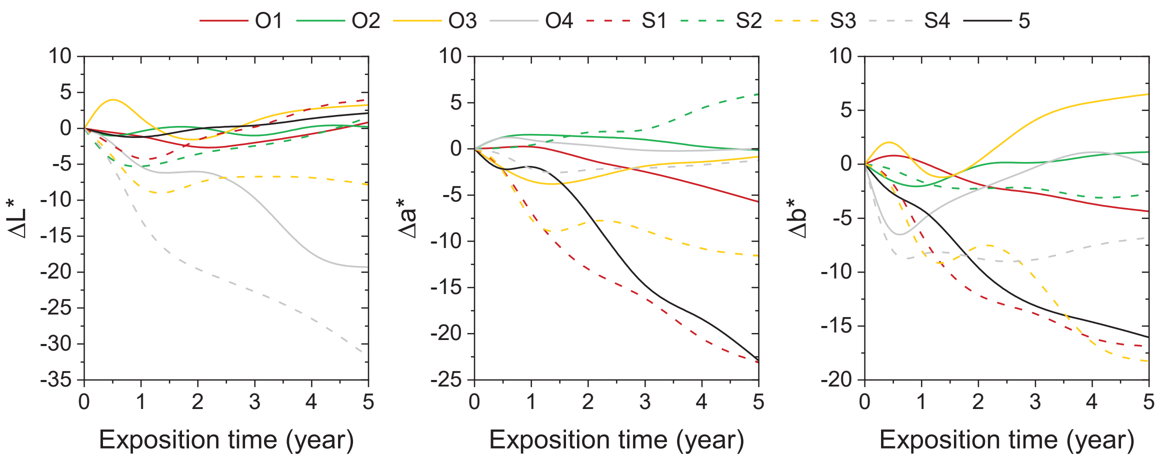

Figure 3. Colour parameters (L, a* and b*) change during weathering (O – opaque, S – semi-transparent, 5 – unpainted)*

4.3 Colour and gloss

The opaque versions of the paint systems were always glossier than the semi-transparent coatings (Tab. 4). One-layer coatings are characterised by a low paint spread rate, which covers the wood texture only partly and does not form a continuous film. Opaque red paint had significantly less gloss, even though the ratio between pigment and linseed oil was comparable to the green and white systems. Surface gloss is affected by many factors like oil absorption of the pigment, the binder ratio, pigment particle size and the pigment's own properties of light absorption and reflection. Surface gloss gradually decreased during the period of exposure and after three years was comparable for all systems on test. The matting of the surface is related to paint chalking caused by binder deterioration leaving loose, powdery pigment on the film surface, which can also cause colour fading. Surface chalking due to the decomposition of the linseed oil is typical for oil paints. The gradual accumulation of dirt on the paint surface in outdoor conditions can also affect the gloss value [13].

The unpainted spruce wood showed characteristic discolouration during weathering – it became darker and less yellow and took on a grey colouration [14]. Generally, the colour lightness of the paint was relatively stable in almost all cases (Fig. 3, Tab. 4). The most significant changes were observed on light surfaces, which were visibly darkened by accumulated dirt (Fig. 5). The chromatic parameters a* and b* were most stable in green paint (chromium oxide pigment). All used pigments are highly light stable, but many paints, especially brighter ones, fade and turn duller with time. Fading is aggravated by chalking since the chalk produced is generally white or very light and masks the paint colour [13]. Exterior coatings normally gather dirt and become increasingly soiled from smoky air, pollen and others factors and subsequent mildew growth, which influence all colour parameters. Controlled chalking can be an asset, especially in white or light paints, since it is a self-cleaning process and helps to keep the surface clean and white by dirt washing off with the chalk during rain [15]. The colour as measured can thus be influenced by the time of the reading since the last intensive rain.Semi-transparent paint discolouration is mainly related to the loss of pigment during exposure (Fig. 5) and is influenced by colour changes in the wood substrate (Fig. 3). The colour of S2 appeared to be relatively stable despite significant loss of pigment from the wood surface, which was due to the relatively close absolute value of the colour parameters of freshly painted wood and aged unpainted wood (45.2 vs. 51.3 for L*, -10.4 vs. 1.4 for a* and 12.1 vs. 5.2 for b*) compared to other pigmented paint.

Table 4. Colour parameters before and after five years' exposure for individual systems (O – opaque, S – semi-transparent)

	L*		a*		b*	
	before	**after**	**before**	**after**	**before**	**after**
O1	39.0	39.9	28.1	22.3	21.0	16.6
O2	41.6	41.8	-13.2	-13.4	11.2	12.4
O3	54.0	57.2	17.7	16.8	42.0	48.5
O4	92.0	72.7	-0.4	-0.4	9.6	9.6
S1	42.9	46.9	32.6	9.5	26.3	9.4
S2	45.2	46.7	-10.4	-4.5	12.1	9.4
S3	57.7	49.9	20.2	8.6	41.8	23.5
S4	88.1	56.3	2.4	1.2	12.6	5.8

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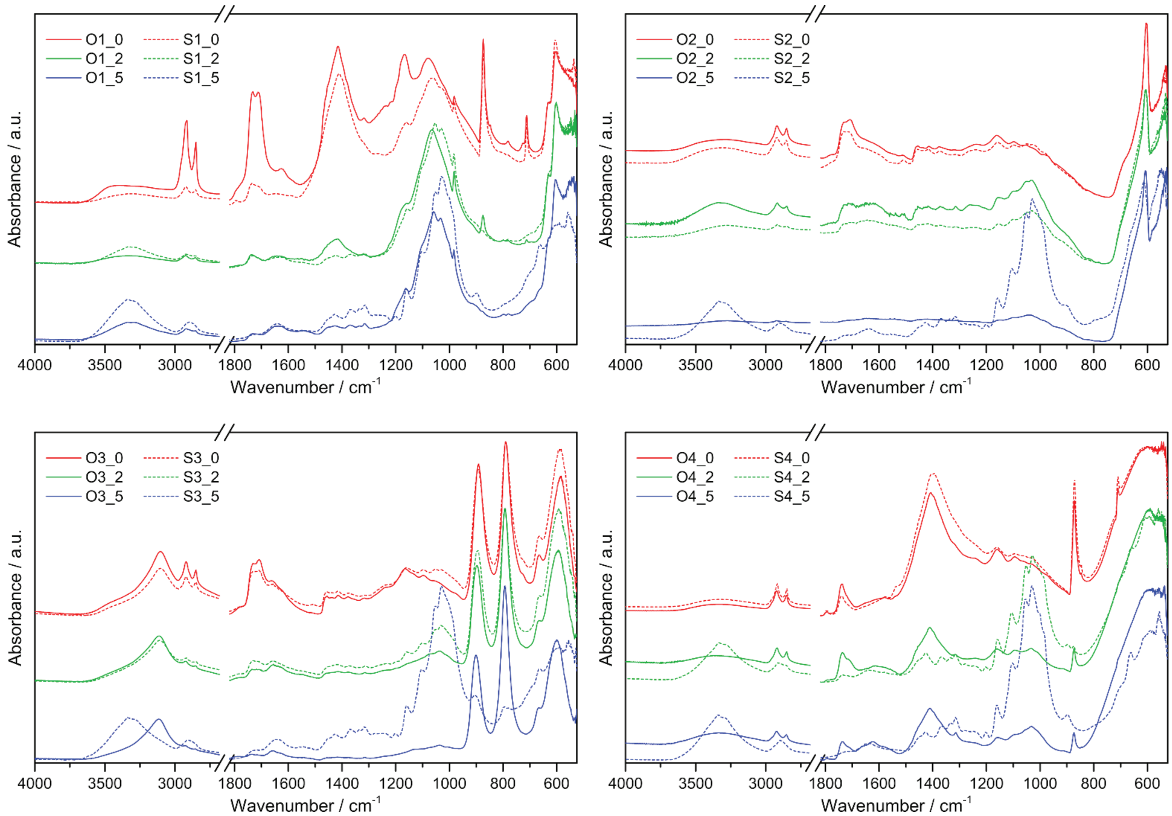

Figure 4. FTIR spectra of paint systems before (0) and during exposure (2 and 5 years); O – opaque, S – semi-transparent)

4.4 ATR-FTIR measurement

Fig. 4 shows the changes in the ATR-FTIR spectra of each system during weathering. Prominent peaks in the spectra correspond to linseed oil and the various pigments. The main peaks of linseed oil are at 2,920 cm^{-1} and 2,850 cm^{-1}, which correspond to the stretching vibration of the methylene groups in fatty acid chains, and also the peak at 1,740 cm^{-1} corresponding to the

C=O stretching vibration of the ester carbonyl groups in triglyceride molecules [16]. All these peaks are visible in spectra of paints before exposure (Fig. 4). The individual pigments have characteristic spectra, for example, chrome oxide is represented by peaks at 610 cm^{-1} and 550 cm^{-1} and ferric oxide by peaks at 790 cm^{-1} and 890 cm^{-1}. The spectra of the pigments themselves become more pronounced during weathering, especially in opaque paints, when linseed oil in the surface layer of paint degrades, leaving a layer of pure pigment on the surface. It is evident that during the second year of exposure, there was a significant loss of linseed oil. In the semi-transparent systems, most of the paint was eroded away by weathering, leaving the wood exposed. The ATR-FTIR spectra of weathered specimen taken after five years, given in Fig. 4, show the same basic structure, a strong broad O-H stretching at 3,300–3,600 cm^{-1} and the fingerprint region which is assigned to stretching vibrations ascribed to different groups of wood components at 1,800–800 cm^{-1}. Most of these bands have contributions from both carbohydrates (cellulose and hemicellulose) and lignin [17]. At the same time the pigment peak intensity decreased. In the specimen with opaque paint, the pigment still covered the surface of the specimen after five years, although signals from linseed oil were no longer detectable.

5 – CONCLUSION

Linseed oil paints made in the traditional way with different pigments were applied to exterior wooden surfaces and monitored for five years. Opaque paint systems performed well; green paint in particular protected the wood for the whole exposure period. We applied only two coats to save time; usual practice is to

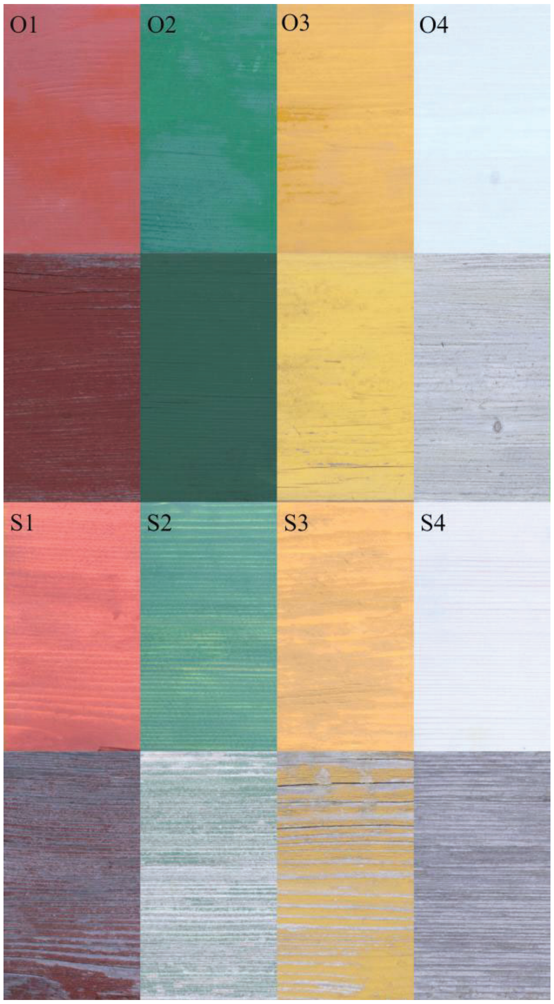

Figure 5. The surface appearance change of painted specimens before (1st and 3rd rows) and after 5 years of exposure (O – opaque, S – semi-transparent)

apply at least three coats, which would significantly extend their lifespan. Our experimental results revealed that semi-transparent paint (one coat) degraded significantly within three years. Such a coating would be used to protect wood only temporarily and should be re-painted in one or two years if exposed to direct sunshine and rain.

The type of pigment affected the colour darkness of the surface and this in turn affected its temperature during sunny days. The surface temperature can influence the rate of moisture exchange, as heating accelerates the drying of the wood.

The initial gloss of opaque paint became dull over time and, after three years, a matt surfaces comparable to semi-transparent paint remained. The main reason for colour fading of oil-based paints is chalking, which together with accumulated dirt and mildew also affects the colour parameters. This effect was most pronounced in light colours, which contrast more with the dark colour of the dirt. Semi-transparent paints lost its pigment layer after two years of exposure and the measured discolouration was therefore dominated by the change in the colour of the wood itself.

The ATR-FTIR spectra of opaque paints showed that peaks corresponding to linseed oil decreased in the surface layer and a layer of almost pure pigment was created. Semi-transparent paints were characterised by a partial decrease in the pigment peaks and the appearance of peaks typical for wood due to loss of more continuous protective pigment layer. Oil paints exhibit specific behaviour during weathering, which affects their appearance and maintenance options. Modern paint systems look different when they age and therefore can have an inappropriate effect on historical monuments if compared to original painting of wooden parts.

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