Received: 3 June 2021 | Revised: 24 August 2021 | Accepted: 24 September 2021

DOI: 10.1111/cote.12585

Coloration Technology

ORIGINAL ARTICLE

Colour stability of wood coatings pigmented with natural indigo from *Isatis tinctoria* after accelerated weathering

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Funding information
Academy of Finland

Abstract

An experimental study of artificial weathering of two outdoor coatings pigmented with natural indigo from *Isatis tinctoria* was carried out. The two coatings, natural indigo-pigmented boat varnish and outdoor paint, were exposed to cycles of ultraviolet light, water condensation and water spray for 40 days in a QUV test chamber. Synthetic indigo and commercial ultramarine pigment were employed as reference materials. The effects of artificial weathering on the coatings were evaluated with CIELab parameters, as well as visual inspection. Coating and pigment stability during artificial weathering were studied with reflectance spectrometry, hyperspectral imaging technology and Fourier Transform–infrared spectroscopy. According to the results, coatings with indigo proved to be more resistant to visual and spectral changes than unpigmented or commercially pigmented coatings.

1 | INTRODUCTION

Annually, more than 2 million tons of colourants are produced worldwide, with most of them being derived or synthesised from non-renewable sources.[1] Moreover, the manufacturing processes of colourants usually involve a plethora of environmentally harmful chemicals, causing severe pollution.[2] Therefore, bio-based colourants are currently being investigated as potential alternatives to synthetic colourants.[3] A good example of a colourant with a direct natural alternative is indigo, which was originally produced from plant-based sources, only to be superseded by synthetic indigo production in the late 1800s.[4-6]

As synthetic indigo is produced from aniline, obtained from petrochemical processes, it is imperative to seek alternative ways to produce indigo.[7,8] Thus, there has been a lot of research, and even some lifecycle analysis studies, investigating the possibility of natural indigo as an environmentally friendly alternative to synthetic indigo.[3,9] Many of the indigo plants can only be cultivated in subtropical conditions, but biperennial woad (*Isatis tinctoria*) has become the preferred indigo plant in colder climates such as northern Europe, because of its hardiness and preference for cold winter between growth seasons.[10] Natural indigo produced with woad has the same general characteristics as synthetic indigo, but its properties and behaviour as a colourant may not be entirely equal to its synthetic counterpart, and therefore must be assessed separately.[11]

Among natural colourants, indigo dye generally stands out because of its excellent light fastness properties.[12] Good light fastness is particularly required for outdoor applications, for example, as wood coatings and billboard inks.[13,14] Accelerated weathering tests, which expose samples to ultraviolet (UV) light and water condensation, are a common way to evaluate the colour fastness of coatings

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TABLE 1 Compositions of the coatings

Sample name	Pigment type	Pigment (wt-%)	Thinner (wt-%)	Coating type	Coating (wt-%)
Paint	-	-	-	Linseed oil paint	100
Nat paint	Natural indigo	20	5	Linseed oil paint	75
Synt paint	Synthetic indigo	10	5	Linseed oil paint	85
Ult paint	Ultramarine	5	5	Linseed oil paint	90
Varnish	-	-	-	Boat varnish	100
Nat varnish	Natural indigo	5	5	Boat varnish	90
Synt varnish	Synthetic indigo	5	5	Boat varnish	90
Ult varnish	Ultramarine	5	5	Boat varnish	90

during outdoor use.[15] However, few experiments assessing the light fastness of natural indigo in wood coatings have been conducted. Some other natural colourants have been tested, with promising results: in their 2009 study, Goktas et al showed that the light fastness of wood treated with madder root (*Rubia tinctorium* L.) solution was more resistant to colour change than its non-treated counterparts during exposure to UV light; and in 2015, Yeniocak et al demonstrated that wood stained with red beetroot (*Beta vulgaris*) had better light fastness properties than wood stained with a comparative commercial product.[16,17]

The objective of the current study was to examine the behaviour of a natural indigo colourant in wood coatings exposed to artificial weathering, focusing specifically on the changes occurring in the natural indigo pigment. The effects of artificial weathering on two types of coatings using natural indigo, synthetic indigo and a commercial blue pigment were examined with a reflectance spectrophotometer, a portable hyperspectral camera and Fourier Transform–infrared (FTIR) spectrometry. In many fields, reflectance spectrophotometers have been frequently used for colour analysis and for quantifying the effects of surface ageing.[18-22] FTIR spectrometers are also used for the characterisation of surfaces and quality control of materials.[23,24] However, hyperspectral imaging is a relatively new technology, which divides the spectrum of reflected light into multiple channels.[25,26] It has been used in various medical applications, as well as the characterisation of polymer materials, assessment of the environmental state of crops and vegetation, in dyeing recipe predictions, and even in assessments of the condition of paint in civil structures.[25,27,28,29,30]

2 | EXPERIMENTAL

2.1 | Materials

Both natural indigo pigment (Natural Indigo Finland Ltd) and synthetic indigo (Sigma-Aldrich Ltd, MQ200) were used as purchased. Blue ultramarine pigment (630 Ultramarine Blue), along with linseed oil-based varnish (Uula Boat Varnish), thinner (Uula Universal Thinner), white primer (Uula Exterior Primer) and titanium white paint (Uula Linseed Oil Paint), were obtained from Uula Color Ltd. The varnish consists of boiled linseed oil, Uula Universal Thinner, natural resins and additives. The paint consists of boiled linseed oil, titanium dioxide pigment, Uula Universal Thinner and additives. Uula Universal Thinner consists of hydro-treated heavy naphtha with no aromatic compounds. The main component of the coatings, boiled linseed oil, consists of linolenic acid (45%-65%), oleic acid (13%-25%), linoleic acid (10%-20%), palmitic acid (3%-10%), stearic acid (1%-6%) and drying agents.[31-33] Wooden panels cut from planed pine timber were utilised in the coating tests.

2.2 | Sample preparation and testing

Natural and synthetic indigo pigments were ground to fine powder with a planetary ball mill (Fritsch Pulverisette 6 with zirconium oxide grinding balls). Grinding was performed over 10 1-minute periods with a rotational speed of 550 rpm.

Wooden panels with a size of 15 x 70 x 320 mm were hand-sanded with mesh 150 sanding paper according to standard SFS-EN 927–6:2018. After sanding, the samples were ground-coated twice with the white primer paint using a regular paint brush. After each ground-coating, the samples were dried for 48 hours at room temperature.

Coatings for the samples were mixed from pigments, thinner, linseed oil paint and varnish. The coatings were applied to the samples using a regular paint brush. Each sample was coated twice with one of the mixed coatings and dried at room temperature for 48 hours after each coating. The compositions of the coatings are listed in Table 1. The amounts of added pigments were chosen so that the coatings would have a comparable colour

intensity, therefore there is variation between the contents of the two indigo and ultramarine pigments.

All the samples were exposed to accelerated weathering using a UVA test chamber (QUV Accelerated Weathering Tester, Q-Lab Corporation). The accelerated weathering test was conducted according to standard SFS-EN 927–6:2018. The exposure cycle repeated in the test is presented in Table 2. Step 1 of the cycle was performed first, exposing samples to water condensation. Step 1 was followed by step 2, which consisted of 48 repetitions of steps 3 and 4, exposing samples to alternating UV light and water spray. Following that, the whole cycle was started again from step 1. The exposure cycle was continued for 40 days in total, after which the samples did not show substantial changes and the exposure was stopped.

Before, during and after the test, the spectral properties of the samples were monitored by measuring the visible reflectance spectra of the samples (Datacolor 600 TM, Datacolor). For analysing the colour of the samples, the device transferred the spectra to CIELab colour coordinates according to standards ISO 11664–3:2019 and ISO 11664–4:2019. Additionally, both exposed and unexposed samples were photographed with a Specim IQ hyperspectral camera operating at a wavelength range of 400-1000 nm, to determine possible changes in the near-IR reflectance spectra of the samples caused by the weathering test. To investigate the whole IR spectra of the samples, *ca*. 10 x 10 x 1 mm pieces of the top layer of the samples were cut off and FTIR measurements (Alpha Eco-ATR, Bruker) of the pieces were carried out.

3 | RESULTS

The photographs showing the appearance of all the samples coated with linseed oil paint and varnish before and after the 40-day testing period are presented in Figure 1. All the pigments had a much stronger tint in the varnish-coated samples, which were rather dark at the beginning of the experiment. The indigo-pigmented samples were greyish in the paint and blackish in the varnish, whereas the ultramarine pigment gave the varnish a bright blue colour. All the samples, including the unpigmented references, went through visible changes due to accelerated weather exposure. In addition to visual changes, the surfaces of all the samples went from being relatively smooth to relatively rough during exposure.

CIELab lightness (L^*) and CIELab colour parameters (a^* and b^*) during exposure testing are shown in Figure 2. The lightness of the painted samples (solid lines) decreased substantially during exposure, whereas the lightness of the varnished samples (dashed lines) stayed at an approximately constant level for all the samples, except for the varnished sample with no colourant. The white paint without blue pigment appeared to darken more than the pigmented paint samples. Also, the varnish without pigment underwent a greater change in lightness compared with the pigmented samples, although it stayed at much a higher level compared with them.

Considering the changes of the colour coordinates from green to red (a^*), and from blue to yellow (b^*), the colour of the coatings pigmented with synthetic indigo changed less than the other samples (Figure 2B). All the samples displayed some yellowing, and, except for the ultramarine-pigmented samples, also some reddening. The shade of the pigmented, painted samples ended up close to the original, indicating a greyish colour, whereas the varnished samples displayed more variation and change in their colours. For the pigmented, varnished samples, it was characteristic that they merely changed in colour but not in lightness (except for the sample with synthetic indigo, which showed a slight decrease in lightness).

To further illustrate and compare the scale of visual changes in the samples during testing, the total colour change (ΔE^*) was calculated from the lightness and both colour values using the following equation:

$$\Delta E^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}},$$

where ΔL^*, Δa^* and Δb^* are changes in the CIELab parameters over the exposure time.

The total colour changes over time are shown in Figure 3. As can be seen from the figure, the samples with synthetic indigo represented the least total colour change

TABLE 2 Exposure cycle of the test. The cycle was repeated six times

Step	Function	Temperature (°C)	Duration (h)	Conditions
1	Condensation	45 ± 3	24	-
2	Subcycle step 3 + 4	-	144 h consisting of 48 cycles of 3 h consisting of steps 3 and 4	-
3	UV light	60 ± 3	2.5	Irradiance set point 0.89 W/(m^2*nm) at 340 nm
4	Water spray		0.5	6 L/min, UV off

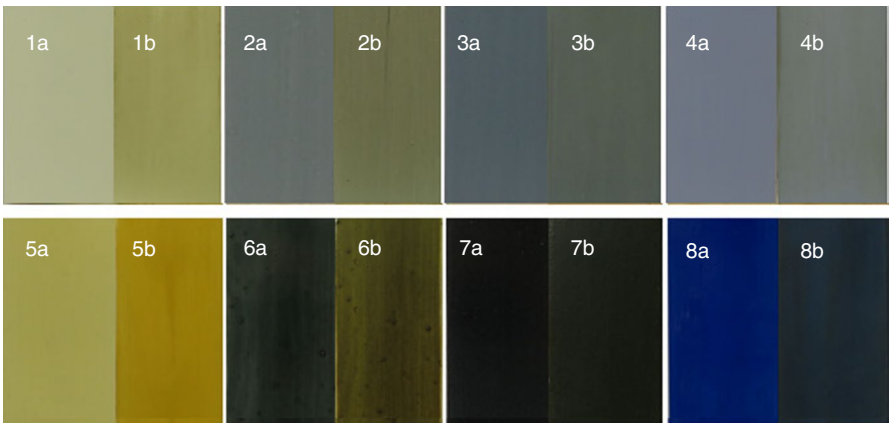


FIGURE 1 The painted (1–4) and varnished (5–8) samples before and after the 40 days of exposure testing. Pigments of sample pairs from left to right: unpigmented (1 and 5), natural indigo (2 and 6), synthetic indigo (3 and 7) and ultramarine pigment (4 and 8). The unexposed samples are labelled with "a" and the exposed samples are labelled with "b"

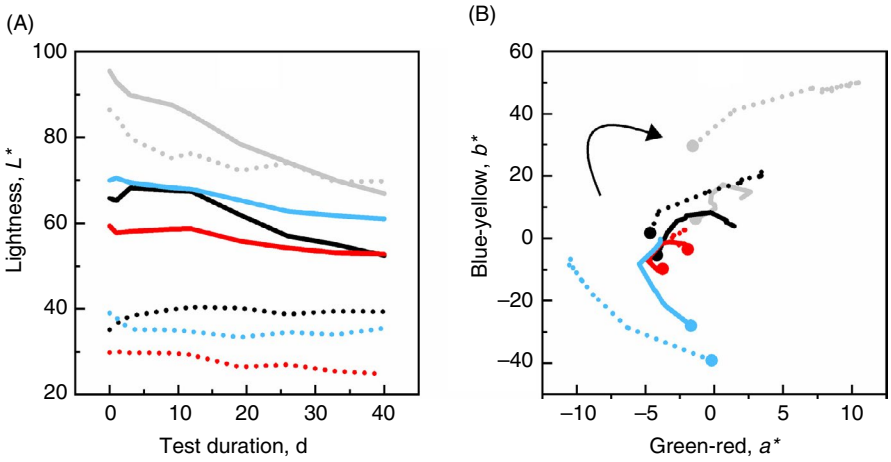


FIGURE 2 A, CIELab lightness L^* as a function of the exposure time, and B, CIELab colour parameter b^* as a function of a^*. In B, the first time point is indicated by a larger marker and the arrow shows the direction of a^* and b^* development in the samples over time. The solid lines represent painted samples and the dotted lines represent varnished samples. Colours of the samples: black = natural indigo, red = synthetic indigo, blue = ultramarine pigment and grey = uncoloured coating

among all the samples. Those samples coated with no colourant, and the samples with commercial pigment, underwent significant colour changes in the experiment.

Reflectance spectra of the aged samples after 0, 1, 3, 9 and 40 days of exposure at the wavelength range of 400-700 nm are presented in Figure 4. All the paint samples had a steep increase of reflectance near 400 nm, which is typical for titanium dioxide.[34] In general, for all the samples, the shape of the reflectance spectrum tended to flatten over exposure time. Flattening of the spectra could be seen, especially in the pigmented samples, and in the natural and synthetic indigo samples, as the peak of the spectrum at around 430-470 nm moved towards higher wavelengths as the samples were exposed. A peak shift could also be observed for the ultramarine-containing varnished sample, even although the peak only shifted slightly. The spectral changes of the paint samples after 40 days of exposure showed practically a full loss of features related to blue pigments, and the spectra finally had a similar shape to the sample that contained only titanium dioxide as the pigment.[35,36] The final shape in the varnished samples containing synthetic indigo and ultramarine had features of the blue pigment in the visible wavelength region.

Based on the location of the main peak in the reflectance spectra of indigo samples, the reflectance of all samples at 430 nm was studied during exposure testing. From the results, at 430 nm a relative loss of reflectance over time was calculated for each sample (Figure 4). Interestingly, the reflectance losses of indigo samples behaved in a very similar manner. Also, the reflectance of both ultramarine and unpigmented samples behaved very similarly over time. The reflectance loss at 430 nm was smallest for the varnished and painted indigo samples and largest for the varnished unpigmented and ultramarine samples.

Reflectance spectra of the samples were measured from 400 to 1000 nm to determine how the colourants affected changes in coatings during exposure. For all samples, the hyperspectral reflectance values before and after exposure are presented in Figure 5. The changes in the spectral region beyond the visible wavelength region were quite dramatic for the paint samples containing blue pigments,

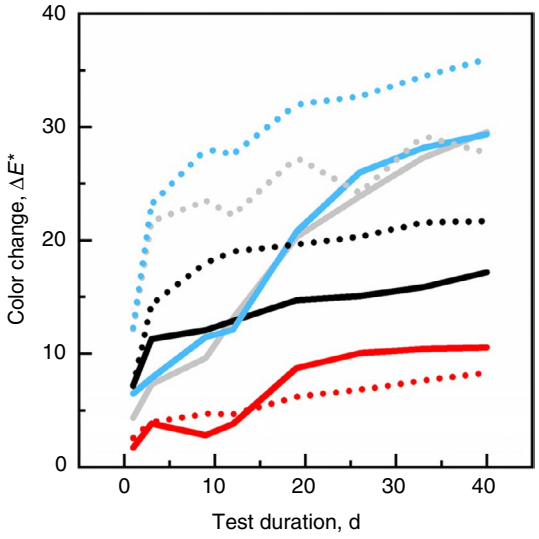


FIGURE 3 Development of CIELab total colour change during the exposure period. The solid lines represent painted samples and the dotted lines represent varnished samples. Colours of the samples: black = natural indigo, red = synthetic indigo, blue = ultramarine pigment and grey = uncoloured coating

as the reflectance decreased over 750-1000 nm. The varnished samples did not display such substantial changes.

As can be seen from Figure 5, most of the differences between the samples in the visible range were in two areas, 400-480 and 500-750 nm. In the 400-480 nm area for painted and varnished samples, synthetic indigo and natural indigo samples displayed less spectral change than samples with commercial pigment or unpigmented samples. In the 500-750 nm area for painted samples, natural indigo showed the least spectral change, closely followed by synthetic indigo and commercial pigment, whereas the unpigmented sample displayed far more spectral change. For varnished samples in the 500-750 nm area, there was least change in the unpigmented and synthetic indigo samples. In the 500-750 nm area, commercial pigment and natural indigo demonstrated the most spectral change of the varnished samples.

FTIR spectra of the samples over the wavelength range 400-3200 cm^{-1} were measured to investigate the possible chemical changes of the coatings during the artificial weathering test. The FTIR spectra of the unpigmented and indigo coloured samples are presented in Figure 6.

The spectra of unexposed samples were virtually identical, with the exception of painted samples in the 1550-1650 cm^{-1} area, whereas the spectra of exposed samples displayed some differences in certain wavelength areas. According to the FTIR database of the Institute of Chemistry in the University of Tartu, the largest absorption peaks of linseed oil are located at 2923, 2853, 1742 and 1160 cm^{-1}, with smaller peaks at 1461 and 721 cm^{-1}.[37] Knowing that most of the varnished coating consists of linseed oil, it is unsurprising that these peaks could also be seen in the FTIR spectra of the varnished samples. According to Baran et al,[38] the largest peaks, indicating synthetic indigo, are found at around 1623 and 1022 cm^{-1}. Their study also showed that natural indigo has additional wide peaks at around 1410 and 1022 cm^{-1}. For varnished samples, the peak which was most apparent was the one at 1615 cm^{-1}. In painted samples, absorption peaks caused by linseed oil at 2923, 2853, 1742, 1461 and 1160 cm^{-1} were also the most dominant ones. In addition to linseed oil, titanium dioxide is a major component in white base paint. According to a study conducted by Al-Amin et al,[39] titanium dioxide has a small absorption peak at 1632 cm^{-1} and a large wide area at 500-700 cm^{-1}, peaking at 690 cm^{-1}. These peaks could also be observed in the spectra of the paint-coated samples. Indigo pigments added to the paint could be observed at the 1615 cm^{-1} peak area, which were smallest for the sample with no indigo and largest for the sample with synthetic indigo.

FTIR spectra of all samples became substantially flatter during the exposure period. Furthermore, the strong peaks at 2860 and 2930 cm^{-1} disappeared almost completely and the peak at 1740 cm^{-1} became substantially smaller in all samples. The unpigmented samples and samples with synthetic indigo preserved more of their original features compared with samples with natural indigo. However, the peak typical of indigo pigment at 1615 cm^{-1} was preserved in both synthetic and natural indigo samples.

4 | DISCUSSION

4.1 | Colour changes and visual appearance

Based on L^*, a^*, b^* and ΔE^* values, synthetic indigo-pigmented samples displayed the best colour fastness performance, followed by natural indigo-pigmented samples. The good stability of indigo-containing samples indicate that it is possible that the indigo pigment may have protected the sample surface from degradation caused by exposure to UV light, surface condensation and water spray. The UV absorbance of indigo is very high and, among colourants found in nature, it has exceptional UV stability.[12,40]

4.2 | Pigment stability

The paint system was designed to create full coverage on the wood surface to avoid complications in the colour

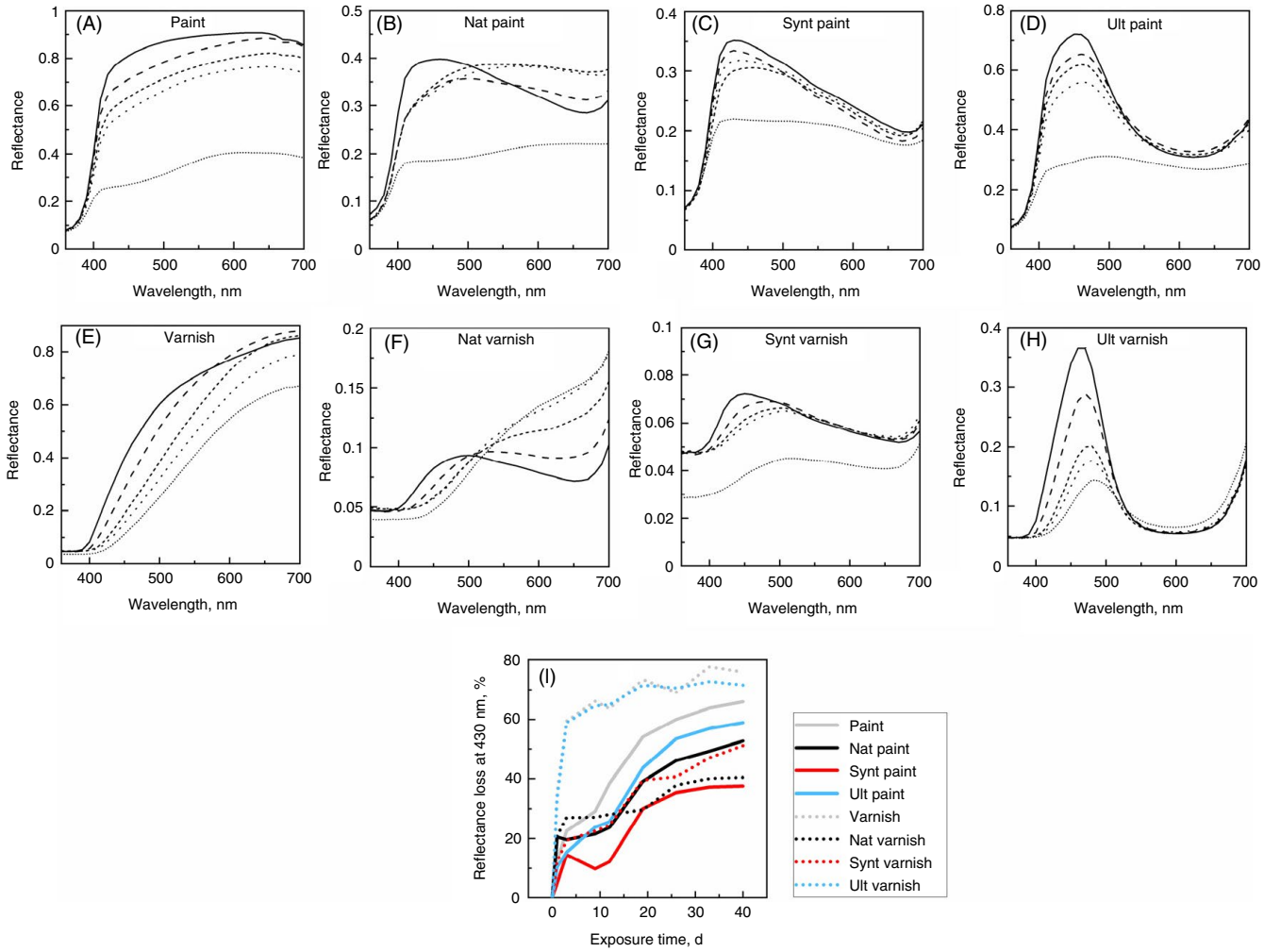


FIGURE 4 Development of visible reflectance spectra of the samples during the exposure period, A–H, and reflectance and reflectance loss at 430 nm over exposure time, I. In A–H, spectra were measured before exposure (solid lines) and after 1 day (long dashed lines), 3 days (dashed lines), 9 days (dotted lines) and 40 days (small dotted lines) of exposure

analysis due to changes occurring in the colour of the wood surface. Including titanium dioxide created a fully covered coating, and the visual and measured colour originated fully from the coating system. However, the presence of the titanium dioxide pigment may have facilitated the degradation of the indigo pigment through photocatalytic reactions, which would be a plausible explanation for the flattening of the spectral features measured for the painted samples (Figure 4).[41] Part of the colour changes and flattening of the reflectance spectra could be attributable to increased light scattering of the aged samples, showing up as increased reflectance at the visible light region of 400-700 nm. This could indicate degradation of the sample surface caused by a combination of water condensation and UV light exposure, which is an ageing mechanism of the coating. However, the shift of the spectral peak on indigo samples might suggest chemical changes occurring at the sample surface, possibly due to oxidative degradation of indigo by UV radiation, which is additionally enhanced by the titanium dioxide particles. There are some reports in the literature concerning using titanium dioxide for dye remediation through photocatalytic reactions, which may also take place in our coating system. The irradiation in the accelerated weather test was applied at UVA range, which overlaps with the absorbance bands of both indigo and titanium dioxide. In the paint systems, this may have led to enhanced ageing due to the excitation of titanium dioxide, in contrast to varnish, where there was no titanium dioxide. Potential reaction products for indigo oxidation are isatin and isatoic anhydride.[40,42] However, no sign of their presence was found in the FTIR analysis (Figure 4).

4.3 | Coating stability and ageing

Based on hyperspectral data, both painted and varnished samples containing indigo showed more

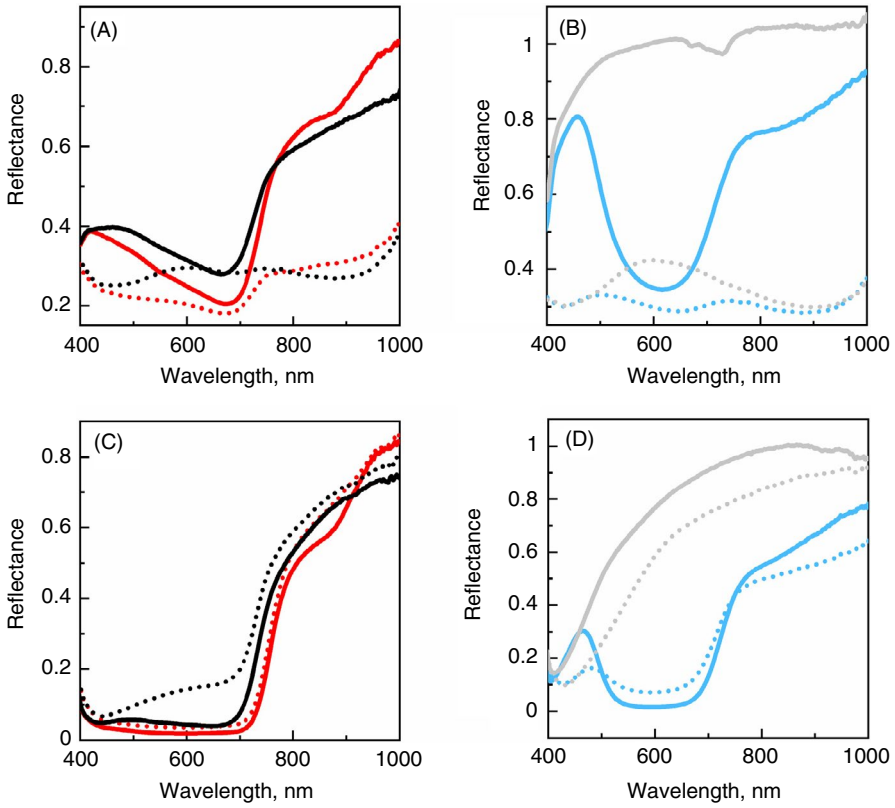


FIGURE 5 Reflectance spectra of the samples before and after the exposure period as measured with the portable hyperspectral camera. Painted samples are presented in A and B, and the varnished samples are presented in C and D. Unexposed samples are presented with solid lines and exposed samples with dotted lines. Colours of the samples: black = natural indigo, red = synthetic indigo, blue = commercial pigment and grey = uncoloured coating

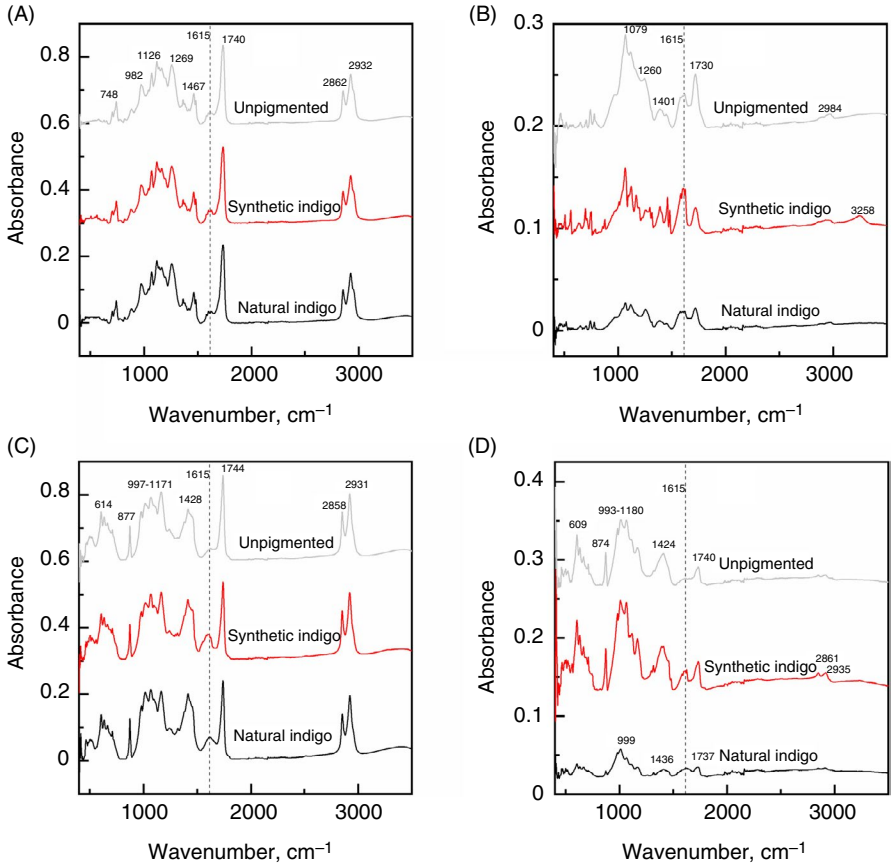


FIGURE 6 Fourier-transform infrared absorption spectra of the samples before and after the artificial weathering period. A, Varnished unexposed samples; B, Varnished and exposed samples; C, Painted and unexposed samples; and D, Painted and exposed samples

resistance towards reflectance loss than samples with no indigo (Figure 5). This indicates that compounds in varnish mixtures reflecting wavelengths of 700-1000 nm were not particularly sensitive to artificial weathering, but that the compounds in the paint mixtures were. Furthermore, it seems that indigo in the coating mixtures slowed down reflectance changes occurring in the coatings when exposed to artificial weathering (Figure 4I).

Flattening of the peaks of all FTIR spectra in Figure 6 is a sign of degradation in the coating materials during exposure. This indicates that the coating with natural indigo degraded faster than the other coatings, even although natural indigo appeared to slow down reflectance changes in the coatings during exposure testing. In addition, the peaks typical of indigo at 1615 cm^{-1} were still present in natural and synthetic indigo coatings after exposure, and had shrunk substantially less than other peaks in the spectra. This indicates that indigo in the coating mixtures was not as sensitive to artificial weathering as other parts of the coating.

5 | CONCLUSIONS

The obtained results showed that indigo pigment protected a linen oil-based coating applied to wood against colour changes caused by artificial weathering. Although the performance of natural indigo coatings was not quite as good as that of synthetic indigo coatings, they still yielded substantially better protection for wood than those coatings with commercial blue pigment, or those with no coloured pigment. Furthermore, the characteristics of synthetic indigo coatings were similar to their synthetic counterparts. The choice of other pigments and fillers in the coatings may affect the stability of both indigo and the matrix quite substantially, which was observed as accelerated ageing of the paint formulations due to titanium dioxide. Therefore, careful evaluation of the stability properties of the coatings under relevant conditions, and understanding of the potential interactions of the raw materials and components, are essential when designing a sustainable high-performance outdoor coating system.

ACKNOWLEDGEMENTS

Dr. Timo Pääkkönen from Aalto University is thanked for assistance in measuring FTIR spectra. Joni Hyttinen from the University of Eastern Finland is thanked for guidance in hyperspectral imaging. The Academy of Finland is acknowledged for supporting the work by funding the project “Bio Based Dyes and Pigments for Colour Palette (BioColour)” (Grant 327209).

CONFLICT OF INTERESTS

There is no conflict of interest related to the manuscript.

INFORMED CONSENT

The manuscript does not contain work requiring ethics approval or patient consent. No material from other sources are included in the manuscript. The manuscript contained no work related to clinical trials.

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How to cite this article: Jordan J, Helander R, Laaksonen P. Colour stability of wood coatings pigmented with natural indigo from *Isatis tinctoria* after accelerated weathering. *Coloration Technol*. 2022;138:210–218. https://doi.org/10.1111/cote.12585