WOOD COLOURING WITH NATURAL DYE EXTRACTS. NEW RESEARCH AND PERSPECTIVES

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Abstract: The aims of the present research were to test new types of natural dyes extracts for wood as a possible alternative to synthetic dyes, and to evaluate the colour changes after a period of one year exposure to natural light, in indoor conditions. Three types of extracts using raw materials: nutshell from Juglans regia L. (E1), onion peels (E2), and dry plant of Chelidonium majus L. (E3) were employed. Spruce wood (Picea abies L. Karst) samples with dimensions of (120x80x5) mm were used. The dye aqueous solutions with a concentration of 5% (E1, E3) and 2.5% (E2) were obtained by solid-liquid extraction at 100°C for three hours. The extracts E1, E2, E3 were modified by the addition of mordant ferrous sulphate Fe₂(SO₄)₃ 3% and coded EM1, EM2, EM3 respectively. The wood was coloured by dipping procedure at 60° C for 30 min. Three replicates were employed for each type of dye solution. After conditioning, two replicates together with the uncoloured samples (M) were further exposed indoors, and one coloured sample remained unexposed, as control. Colour measurements in the CIELab system were performed before and after each period of exposure (30, 60, and 365 days), and the colour differences ΔL, Δa, Δb, and ΔE were calculated. Generally, all the samples dyed with natural extracts had fewer colour differences compared with the uncoloured wood. The lower value for colour change was registered for E2 extract, followed by E3, and E1 extract, respectively. According to the exposure time, the colour differences values ΔE range correspond to a medium colour up to different colour in visual perception. For mordant dye extracts EM1, EM2, EM3 the results indicated a different colour in visual perception. The ΔE values registered were even higher than the uncoloured samples (M). The results were in accordance with similar research in the field. The present study opens new research opportunities in wood finishing and new colouring perspectives with eco-friendly dyes.

Key words: wood colour, dye extract, colour change, mordant ferrous sulphate, ecological.

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1. Introduction

Used since ancient times, the colouring of wood with natural dyes is coming back to the attention of researchers and users. Most natural dyes are from plant sources such as roots, berries, bark, wood, and leaves. While some of these natural dyes are well-known and have been commonly used for centuries, especially for textiles, others have not been studied yet and might be used in the future as new green colouring technologies.

Wood colouring is an important step in finishing technology. It has an essential aesthetic role and a great economic impact and as a result, many types of new dyes have been tested recently by novel investigation methods [1, 4, 11-13, 26]. Colour is always associated with light [16] and the colour fastness of wood to light has been a subject often studied in the last years with potential for further research. The changing in colour of wood is by no means attributed to a change in dye alone or in colouring procedure, but is due mostly to a change in the wood substrate and its chemical components [27, 28].

In recent years, scientists have tested a series of natural dyes on different wood species, cotton, wool or other fabrics [2, 12, 23], and leather [17]. An extended study was done on beetroot dye extract [22, 29, 30] which was focused on the colour changes of wood after UV exposure and the leaching of colour. The procedure of mordanting was used to stabilise the extract colour, but it seems that mordants had a negative colour change performance [29]. However, it was observed that the mordants improved the leaching performance for Scots pine, beech, walnut, and oak [30]. Another natural extract from laurel (Laurus nobilis L.) leaves was tested for colour stability under UV exposure. The results demonstrated that treatments with mordant reduced the total colour change more than the laurel without mordant treatments [10]. The same authors investigated red beet, safflower, and purple cabbage extracts in mixture with water-borne lacquers to finish MDF panels and to test the weathering or mechanical resistance of the surface [8, 9]. Results were encouraging for the dye-lacquer mixtures, but mordants did not seem to be an option for good results in these tests. An experimental study envisaged the testing of the mixture between pigmented coating and natural indigo from Isatis tinctoria (L.) to artificial weathering. Results indicated that the pigment extract protected the coating from colour changes caused by weathering [14].

Another eco-friendly option proposed to be developed for the furniture industry was Quercus ithaburensis (Decne.) acorn dye extract. It was investigated with or without mordant addition for the colouring of Scots pine, Oriental beech, and oak. These types of wood were tested for accelerated weathering. Results showed that the colour change performance of ferrous sulphate mordant application was higher [21].

Extracts from bark and vegetables or plants were tested for colour stability after accelerated aging on different wood species and the results demonstrated that the wood containing fewer extracts and with the lowest phenol content was also the least resistant to light [5].
Natural dye was extracted from *Dalbergia cochinchinensis* (Pierre ex Laness.) and applied directly on wood veneers and a good dyeing uniformity was obtained [32]. Also, waste and bark extracts from tropical species were used to colour fabrics. Mordants were used to ensure the penetration into the fabric fibres and some of the extracts showed good colouring performance [31, 32]. The heat-treated larch extract dye was used to enrich the colour of whitish wood species or fast-growing species, e.g. poplar, and the enzymatic grafting methods can improve the colour stability of the extract dye in the UV test and outdoor test [3].

Considering these studies and the results obtained, the following objectives of the present research are outlined:

- To find new opportunities for wood natural colouring by testing new types of dyes extracts, as a possible alternative to synthetic dyes;
- To evaluate the colour changes of dyed wood, without finishing protection, in indoor conditions, after a period of one year of exposure.

Also, new opportunities for scientific research are opening up and new perspectives are emerging in terms of industrial interest on wood colouring with economic and eco-friendly dyes.

2. Materials and Methods

Spruce wood (*Picea abies* L. Karst) was chosen for the experiments, as a Romanian native and commercially available species, frequently used in applications from furniture to constructions. Planned wood samples with dimensions of (120 x 80 x 5) mm and radial or semiradial faces were employed. The initial moisture content measured by the gravimetric method was 12.5%. Moisture was determined on separate samples from the same lot. The dye extracts were derived from a fruit residues or dry plants as follows:

1. Nutshell from walnut (*Juglans regia* L.);
2. Onion peels (*Allium cepa* L.);
3. Dry plant of celandine (*Chelidonium majus* L.).

The materials were purchased from local flora and the dye extracts were prepared by boiling in a lab condenser installation for three hours, so that the initial water volume added in the flask remained identical during the extraction process. A concentration of 5 and 2.5% was prepared. The literature in the field indicated a ratio of plant material mass to the volume of liquid of 1:10 or 1:20 (w/v) [9, 10, 29]. Therefore, 20 g of milled nutshells were mixed with 400 ml of distilled water, 10 g of dry onion peels in 400 ml of water, and 20 g of celandine, previously dried at 40°C for 12 hours, in 400 ml of water were employed. After boiling, the dyes were cooled at room temperature and then filtered off. The obtained liquids were visually assessed and the pH, clarity, and colour were determined. After preparation, the dye solutions were separated in two parts, one half was kept unmodified and the other half was mixed with ferrous sulphate (Fe₂(SO₄)₇·H₂O) as mordant, in addition to 3% concentration. The mordant is well-known as a stabiliser of colour extract, to fix the colour on wood and sometimes, to increase the dye retention and also, as an option for colour [8, 27, 29]. This method of adding mordant (named metal-mordanting [27] was chosen to have an
advantage in handling, using a single bath of treatment.

The samples were treated by dipping procedure at 60°C for 30 min. Before and after treatment, the samples were weighted to determine the dye uptake \( \text{g/m}^2 \) – Equation (1).

\[
\text{Csp}[\text{g/m}^2] = \frac{m_2 - m_1}{A}
\]  

(1)

where:
\( m_1 \) is the mass before treatment \( [\text{g}] \);
\( m_2 \) – the mass after treatment \( [\text{g}] \);
\( S \) – the total surface of the sample \( [\text{m}^2] \).

Six series of samples: three of them coded E1-nutshells, E2-onion peels, and E3-celandine and another three with mordant addition, coded EM1, EM2, EM3 respectively, were prepared. Three replicates for each treatment were employed. The specimens were left to dry and conditioned at room temperature and at 55 ± 5% RH until a constant weight.

One coloured sample from each series was kept in the laboratory in dark conditions, as unexposed control, and the other two replicates from each colouring variant, together with the uncoloured samples, were exposed indoors to natural light, filtered through a double glass window, on a special rack, facing south, simulating natural aging, for a period of one year (June 2022 – June 2023). Evaluations of colour change were made after 30, 60, and 365 days of exposure.

The colour was measured in the CIELab system with an AvaSpec-USB2 spectrometer (Avantes B.V., Apeldoorn, Netherlands). A standard illuminant D65 under 2° standard observer and an integrating AVA sphere were used. Data were processed with AVASOFT—version 7.7. The natural and coloured samples were subjected to colour measurements before colouring with extracts, and afterwards, prior to being exposed to natural aging and after each period of exposure. The measurements were carried out in 5 points/sample, positioning the sphere of 8 mm in diameter to measure each time in the same point, using a special device. The coordinates \( L^* \), \( a^* \), \( b^* \) were registered and the average was calculated. Colour differences \( \Delta E \) were calculated with Equation (2):

\[
\Delta E = \left[ \Delta L^* + \Delta a^* + \Delta b^* \right]^{1/2}
\]  

(2)

where: \( \Delta L^* \), \( \Delta a^* \), and \( \Delta b^* \) are the differences of initial and final values (before and after ageing for different periods of time) of \( L^* \), \( a^* \), and \( b^* \) parameters.

In visual perception, lower differences in colour correspond to lower calculated colour differences. Different ranges of \( \Delta E \) were correlated with the visual changes observed by the human eye as follows: the smallest visible difference by the human eye (0.2 < \( \Delta E < 2.0 \)), small colour difference (2.0 < \( \Delta E < 3.0 \)), medium colour difference (3.0 < \( \Delta E < 6.0 \)), and high colour differences (6.0 < \( \Delta E < 12.0 \)). \( \Delta E \) values above 12 units mean different colours in visual perception.

3. Results and Discussion

3.1. Characteristics of Dye Extracts and Colouring Specifications

Table 1 presents some characteristics of the dye extracts and application rates on the wood samples. The colour of the
aqueous solution varied from brown to yellowish brown, with a transparent appearance and pH = 5-7. The extracts modified with mordant were brown to black in colour, semi opaque, and with a visible tendency of precipitation. Also, the initial colour of the wood samples was visually evaluated.

### Dyeing specifications

<table>
<thead>
<tr>
<th>Extract / Code</th>
<th>Colour / clarity</th>
<th>Application rate, St dev [g/m²]</th>
<th>The visual aspect of the coloured surface</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>Light yellowish brown, clear</td>
<td>183,8 (36,80)</td>
<td>Light brown, positive image</td>
</tr>
<tr>
<td>E2</td>
<td>Reddish brown, clear</td>
<td>154,24 (22,02)</td>
<td>Yellow brown, positive image</td>
</tr>
<tr>
<td>E3</td>
<td>Yellowish green brown, clear</td>
<td>163,52 (14,63)</td>
<td>Yellow, positive image</td>
</tr>
<tr>
<td>EM1</td>
<td>Black, semi opaque</td>
<td>172,17 (24,02)</td>
<td>Yellow grey, positive image</td>
</tr>
<tr>
<td>EM2</td>
<td>Black, semi opaque</td>
<td>169,18 (13,85)</td>
<td>Brown grey, positive image</td>
</tr>
<tr>
<td>EM3</td>
<td>Yellowish black, semi opaque</td>
<td>236,79 (36,37)</td>
<td>Yellowish grey, positive image</td>
</tr>
</tbody>
</table>

#### 3.2. Colour Evaluation

The scanned images from Figure 1 illustrate a colour change in visual perception for the tested samples, meaning a darkening of the surface colour. This evaluation is important from the perspective of the consumer who considers colour as a factor in the quality of a wooden product.

Fig. 1. Uncoloured spruce wood (M) compared with wood coloured with dye extracts (E1, E2, E3) and mordant extracts (EM1, EM2, EM3), before (upper row) and after exposure (bottom row)
The images show both the initial colour of the natural wood (M) compared with the coloured samples (E1-E3, EM1-EM3), as well as the colour changes of the wood after one year exposure under indoor conditions, compared with the unexposed control.

All the wood specimens underwent visible changes resulting in a different colour, due to natural indoor exposure, except for the E2 extract, where the difference between the initial colour and after exposure is slightly visible. After dyeing with mordant, the colour became darker, with grey hues, and after exposure the colours turned intense dark (EM1, EM2, EM3).

A comparison of results after measurements in the CIE Lab system showed a colour change for the tested samples, according to extract type and exposure time. Generally, all the samples dyed with natural extracts (unmodified with mordant) had fewer colour differences compared with the uncoloured wood (Figure 2a).

The lower ΔE value for colour change was registered for the E2 extract, followed by the E3 and E1 extract. Much higher ΔE values, above those of uncoloured M, were recorded for mordant extracts EM1 and EM3 and below the uncoloured for EM2, respectively (Figure 2b). During the exposure period the colour differences ΔE evolved in time between 3.34-10.89 for extract E1, 7.64-14.91 for extract E2, 11.16-18.74 for E3, compared with uncoloured M (13.5-21.9). Therefore, all three extracts used for wood colouring contribute to colour stability. They could be a good option to reduce the colour change of spruce wood, which was tested in other research in a simulated indoor sunlight exposure for 600 h, equivalent with one year of natural exposure, and obtaining a great value of ΔE =18-20 [19] or ΔE = 34.1 [18]. Moreover, it is well-known that softwoods contain more lignin than hardwoods with 2-10% and were sensitive to photodegradation [6, 10, 27]. The dye extracts mixed with ferrous sulphate registered significant colour change expressed by the ΔE values between 17.15-31.30 for EM1, 9.0-20.91 for EM2 and 18.86-33.28 for EM3 (Figure 1b). All these results correspond to a medium colour change up to different colour in visual perception (ΔE > 12).
Mordant was added as a colour stabiliser, but the high values obtained for the colour changes of mordant dyes were somewhat contrary to this assumption. Some researchers have found lower colour changes after exposure in different tests for mordant extracts [6, 10, 21] and they explained in cited studies that a stabilisation of lignin was obtained, even exposed to the light, as a result of the interaction of ferrous sulphate ions and wood components. High values for colour change ($\Delta E > 30$) similar with the present study have been achieved by other researchers who tested natural extracts mordanted with ferrous sulphate on wood [29] or textiles [22, 31], and exposed the coloured materials in conditions of UV weathering, artificial light or other tests.

The colour dynamics indicated a more rapid change ($\Delta E$) at the beginning of the exposure and the process slowed down until the end of the test (Figure 3).

The photochemical degradation of wood due to sunlight occurs fairly rapidly on the exposed wood surface [6]. The behaviour of the tested samples is somewhat in accordance with many studies in the field that reported a rapid degradation of the colour at the beginning of the exposure, even natural wood or finished wood was exposed to simulated indoor sunlight, UV tests or accelerated weathering [15, 19, 20, 25]. Therefore, the wood substrate has a great influence on colour change, but the type of colorant of other type of coating or treatment must be considered and tested in the future.

Generally, all results showed negative $\Delta L$ (lightness) values associated with the darkening of the samples (Figure 4). This quantitative evaluation corresponds to the visual appearance of the samples (Figure 1) which indicate that the colour turned to blackish. Rather high values were obtained for mordanted dyes during the entire period of testing for all the specimens (Figure 4b), with values of $\Delta L$ that exceeded 18 or 30 at the end of the test. The darkening of the wood colour is attributed to the chemical change of the lignin as a result of photodegradation including visible and UV light [21, 29], but also to the type of dye extract and mordanting procedure. The $\Delta a$ redness values remained in a positive interval for natural samples M.
and coloured with extracts E1 and E3. The extract E2 had a negative $\Delta a$ value after 30 days of exposure, meaning a greenish tendency, but the colour gradually returned to red by the end of the evaluation (Figure 5a). The mordanted extracts recorded the smallest $\Delta a$ redness, ranging between negative and positive values in interval (-2 and +2) (Figure 5b).

**Fig. 4. Variation of lightness after each time of exposure for natural and coloured wood samples: a. dye extracts; b. dye extracts with mordants**

**Fig. 5. The redness variation: a. dye extracts; b. dye extracts with mordants**

**Fig. 6. The yellowness variation: a. dye extracts; b. dye extracts with mordants**
The mechanisms of wood photodegradation have been investigated and it appears that lignin is a key structure because this component is able to absorb in the UV/visible region due to its chromophore groups [6, 7, 18, 27]. As previous research has shown, the yellowing with positive $\Delta b$ is also attributed to the lignin degradation of the tested samples, but the contribution of the dye extract should also be considered. The E3 extract registered a $\Delta b$ value with a great variability of data (Figure 6a) and the dyes mixed with ferrous sulphate EM1-EM3 indicated negative results of $\Delta b$ associated with a blue colour (Figure 6b).

4. Conclusions

The present research is welcome in the context of the growing interest in natural resources and environmental protection. The obtained results provide new information about wood colouring with natural extracts from plant and fruits, exposed indoors to natural sunlight for one year. These eco-friendly dye extracts could be of scientific or industrial interest for wood colouring in the future.

In fact, of all the colouring variants tested, E2 (onion peels extract) recorded the lowest colour change values. It could be a promising variant for further testing, but more physical and chemical investigations and tests must be considered. Moreover, if ferrous sulphate as mordant is added, intense colours are obtained and they could have the potential to substitute exotic wood species requested by customers, but which are more sensitive to colour change.

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