COLOR STABILITY OF WOOD TREATED WITH MADDER ROOT (*RUBIA TINCTORIUM* L.) EXTRACT AFTER LIGHTFASTNESS TEST

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ABSTRACT

The aim of this study was to develop environmentally-friendly wood dyestuff derived from madder root (*Rubia tinctorium* L.) and to determine the color stability of this colorant under UV light irradiation. The dyestuff was mixed with alum and iron for mordants. Wood specimens were prepared from Scots pine (*Pinus sylvestris* L.) and beech (*Fagus orientalis* Lipsky). Wood dyestuff was applied on all surfaces of wood specimens by dipping method. Afterwards, wood panels were exposed to lightfastness test for periods of 500, 1000, and 1500 hours. Results show that almost all solutions of madder root, compared with control, have less color changing value. Meanwhile the highest color stabilities were obtained on the wood samples treated with madder root solutions without mordant after lightfastness test.

KEY WORDS: wood dye, color changes, madder root, lightfastness test, mordant

INTRODUCTION

In modern environments, people are exposed to numerous types of pollutants. Among others, wooden furniture and decoration elements are potential sources for great number of volatile organic
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compounds (VOC) for indoors. Many chemical components are used in wood finishes and coating (color, colorant, stainer, paint) industry. Salthammer et al. (1998), identified about 150 different VOC. Moreover, these chemical components can be used to impart other desirable properties such as dimensional stability, water repellency, fire resistance, color, smell, surface hardness, and mechanical strength. A large number of preservative and colorant compounds were introduced to the market; however, many of them did not gain much acceptance, because of chemical toxicity, low efficacy, high cost, or corrosiveness (Murphy 1990) of the compounds. Most compounds belong to the group of typical solvents and are chemically inert under living conditions. However, a variety of substances such as is known as secondary emission products or reactants (Salthammer et al. 2002). These wood colorant and preservatives are potentially included in waste wood and are one of the highly prioritized persistent pollutants that have raised concerns about effects on humans health (Asari et al. 2004).

Many dyes from synthetic sources can be harmful and cause allergies in humans; therefore, interest in natural dyes has been increased considerably during the last few years. Natural dyes are generally environmentally-friendly and have many advantages over synthetic dyes with respect to the production and application (Singh et al. 2005, Luciana et al. 1997, Tsatsaroni et al. 1998). People had been utilizing natural substances, most of which were derived from plants sources, as dyes. Madder, for instance, was a popular natural dye obtained from a perennial herb, Rubia tinctorium L. that could be used to produce shades of red, pink, purple, and brown (Angelini et al. 1997, Vankar 2000). Also, madder root (Rubia tinctorium L.) produces anthraquinone pigments in its roots, one of them being alizarin (1,2-dihydroxy anthraquinone) which have been used for dyeing in textiles since 2000 B.C. (Angelini et al. 1997).

Turkey has a rich flora because of its geographical position and climate. It is well known that plant based dyes were widely used by the Turks in both central Asia and Anatolia through the history. Rubia tinctorium L. plant is a multi-year climbing plant of 50-80 cm., which belongs to Rubia species of Rubiaceae family (Davis et al. 1965). Dyestuff is found in the roots of the plant and dyestuff amount in the roots changes between 1-4% depending on the conditions of the region. Old roots contain more dyestuff compared to the young ones. Dyestuff is used in dyeing wool, silk, linen fibers, and also used for coloring cottons. The color obtained while coloring the cotton red through a special method is known as Turkish red or Edirne red throughout the world. Rubia tinctorium L. is known dyestuffs that originate from root material of field grown madder included anthraquinone derivates and the most important one is alizarin (Merck 1996). Other anthraquinone aglycanes in madder are purpurine, lucidine and quinazarine (Angelini et al. 1997, Bechtold at al. 2003). It is known that anthraquinone derivatives have been used as anti-inflammatory, anti-microbial, antibacterial and anti-diuretic drugs (Golcu et al.2002, Swain 1966).

The objectives of this study were to develop an environmental-friendly natural wood dye derived from madder root (Rubia tinctorium L.) and to determine its color stability under UV light irradiation. To achieve this goal CIELAB system was used to monitor the color change of the samples during 1500 h of UV light exposure.

MATERIAL AND METHODS

Wood specimens

Specimens of beech (Fagus orientalis Lipsky) and Scots pine (Pinus sylvestris L.) sapwood were used in this study. Specimens measuring 10mm (radial) x 100mm (tangential) x 150 (longitudinal) mm were cut and stored in the laboratory at 20°C ± 2°C and % 65 ± %5 relative humidity in order to reach equilibrium moisture content.
Plant material
The *Rubia tinctorum* L. used in this study was collected according to conventional method (Davis et al. 1965) from the region of Manisa-Turkey. Dried and powdered 3-year-old root material of *Rubia tinctorum* L. (100 g) was refluxed with 4000 ml water. After 3 hours, the suspension was passed over a Buchner filter, and a sample of 500 ml of the filtrate was taken. The remainder of the filtrate was put into freeze dryer in order to dry under reduced pressure (Derksen et al. 2002).

Dyeing procedure
In the experiments, FeSO$_4$7H$_2$O (technical grade 96% purity, Merck) and alum KAl(SO$_4$)$_2$.12H$_2$O (puriss. p.a. Fluka), were mixed as concentrated solution with the addition of mordant (3 % and 5 %) to give a final dyebath concentration of 30 g L$^{-1}$ and 50 g L$^{-1}$ mordant (Trotman 1984, Guzel and Akgerman 2000). Prepared dyestuff have been separated into three containers (for madder + iron, madder + alum, and without mordant), and then heated to 60 °C. The wood panels were immersed into the dyebath containing the dyestuff extract for 30 min (Sonmez and Budakci 2004). Extra solution which was left on the specimens was swept with a clean cloth. Afterwards, specimens were left to dry under the temperature of 20±3 °C on a vertical position.

Lightfastness test
For this study, an irradiation system that is equipped with UVA-351 lamp was utilized. Among the light-induced properties, the variation in color difference value ($\Delta$E$^*$) was the best for establishing the correlation between the accelerated lightfastness testing (Hui and Chang 2001). The test conditions were 50% relative humidity and 20 °C temperature. The samples were exposed to UV light directly at a distance of 5 cm. The total exposure times ranged from 0 to 1500 h. (3 periods x 500 h.). The color of the samples were also measured after each irradiation period. Five replicates were made for both wood species.

Color measurement
Color measurements were determined according to EN ISO 105-JO3 (1999). The color differences of wood surfaces were determined using a portable color reader (Konica Minolta-Color Reader CR-10). The L’, a’, and b’ color space (according to the Commission International de l’Eclaireage-CIE) was used for color evaluation in which L’ axis represents the lightness, a’ and b’ are the chromaticity coordinates. In the CIELAB coordinates, +a’ represents red, -a’ green, +b’ yellow, -b’ blue, and L’ varies from 100 (white) to zero (black). L’, a’, and b’ color coordinates of each samples before and after exposure to UV light irradiation were calculated based on a D65 light surface as established by the CIE 1976 (Billmeyer and Saltzman 1981). These values were used to calculate the color differences ($\Delta$E$^*$) using the following expressions:

\[
\begin{align*}
\Delta L^* &= L_f^* - L_i^* \tag{1} \\
\Delta a^* &= a_f^* - a_i^* \tag{2} \\
\Delta b^* &= b_f^* - b_i^* \tag{3} \\
\Delta E^* &= \sqrt{\Delta L^*^2 + \Delta a^*^2 + \Delta b^*^2} \tag{4}
\end{align*}
\]

Where;
- $\Delta L^*$, $\Delta a^*$, and $\Delta b^*$ are the changes between the initial (i) and the final (f) values.
- $\Delta E^*$ is the standard CIE color difference method, which is simply the distance between the two colors, calculated in three-dimensional L', a', and b' color space. The higher the value of $\Delta E^*$ is, the more considerable the discoloration.
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RESULTS AND DISCUSSION

The color changes of beech and Scots pine wood samples are shown in Tab. 1, Tab. 2, and figured in Fig. 1 and Fig. 2 respectively. Positive values of $\Delta b^*$ indicate an increment of yellow color and negative values an increment of blue color. Positive values of $\Delta a^*$ indicate a tendency of wood surface to reddish while negative values mean a tendency to greenish. Results show that $\Delta b^*$ is generally reduce with increased exposure times in accelerated test cycle for both wood samples treated with madder root extracts.

Tab. 1: Color changes of beech wood exposed to 1500 h UV irradiation

<table>
<thead>
<tr>
<th>Dyestuff</th>
<th>Before UV test</th>
<th>After 500 h</th>
<th>After 1000 h</th>
<th>After 1500 h</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control (without colorant)</td>
<td>70.45</td>
<td>11.18</td>
<td>26.08</td>
<td>-0.32</td>
</tr>
<tr>
<td>Madder (without mordant)</td>
<td>76.63</td>
<td>11.18</td>
<td>26.05</td>
<td>-0.32</td>
</tr>
<tr>
<td>Madder + Alum</td>
<td>74.84</td>
<td>10.46</td>
<td>25.28</td>
<td>-0.28</td>
</tr>
<tr>
<td>Madder + Iron</td>
<td>74.84</td>
<td>10.46</td>
<td>25.28</td>
<td>-0.28</td>
</tr>
</tbody>
</table>

Tab. 2: Color changes of Scots pine exposed to 1500 h UV irradiation

<table>
<thead>
<tr>
<th>Dyestuff</th>
<th>Before UV test</th>
<th>After 500 h</th>
<th>After 1000 h</th>
<th>After 1500 h</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control (without colorant)</td>
<td>70.63</td>
<td>11.31</td>
<td>26.05</td>
<td>-0.32</td>
</tr>
<tr>
<td>Madder (without mordant)</td>
<td>77.44</td>
<td>12.65</td>
<td>28.05</td>
<td>-0.18</td>
</tr>
<tr>
<td>Madder + Alum</td>
<td>77.44</td>
<td>12.65</td>
<td>28.05</td>
<td>-0.18</td>
</tr>
<tr>
<td>Madder + Iron</td>
<td>77.44</td>
<td>12.65</td>
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</tr>
</tbody>
</table>

The color change on surface of both wood (treated/untreated) after UV light irradiation was fast for during the first 0 to 1000 h and thereafter the rate of change diminished to almost plateau at 1000 to 1500 h. Like study of Kamdem (2002), almost 80 % of the final value of $\Delta E^*$ is achieved after 1000 h exposure and only 20 % in the following 1500 h. During the UV test cycle of control samples (without colorants), wood surfaces become darker. The lowest values of $\Delta L'$ were obtained on both untreated wood samples for beech and Scots pine samples. The negative value of $\Delta L'$ value probably occurs because the wood surface became rougher and darker after the UV light irradiation UV irradiation is known to depolymerize lignin and other wood components (Kamdem 2002). Depolymerization of the lignin on the exposed surface may also render the surface darker. $\Delta L'$ was the most sensitive parameter for describing the wood surface quality on the irradiation (Temiz et al. 2005). While, the total color difference of untreated beech wood was slowly increased for all exposure periods, but it was sharply increased during all exposure periods for untreated Scots pine. This color difference was attributed to the carbonyl groups of conjugated ketones, aldehydes, and quinines resulting from the modification of lignin and related compounds.

It was observed that color differences of Scots pine were remarkably higher than beech after 1500 h lightfastness exposure. In study of Şahin (2002) too, in UV experiments, gymnosperm woods showed higher color change than angiosperm woods. Some of the reasons for this difference are anatomical differences, growing characteristics, machining properties of wood, and pre-treatments (e.g. steaming, drying, etc.). Interaction of madder root extracts compound with wood components, also results in differences against photo-degradation effect of UV irradiation (Temiz et al. 2005, Soğutlu and Sönmez 2006). Besides, generally, soft woods have
2-10 % more lignin than hard woods. From the major wood constituents, lignin contributes with 80-95 % to the UV absorption coefficient of wood (Tereza et al. 2004).

The highest color stability values were obtained for beech wood ($\Delta E^* = 1.66$), and for Scots pine ($\Delta E^* = 3.16$) by madder root without mordant. It can be seen from these results; the mordants used here supports the color changing. Between two mordant, iron is more effective than alum on discoloration.

Fig. 1: Color changes of beech wood exposed to 1500 h UV irradiation

Fig. 2: Color changes of Scots pine exposed to 1500 h UV irradiation
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CONCLUSION

Color stability of Scots pine and beech specimens treated with madder root and added with alum and iron mordants was studied after lightfastness test.

Results showed that wood samples treated with madder solution without mordant resulted in the best total color stability after lightfastness test. Madder treatment may reduce the wood color change by reducing the lignin degradation resulting from UV light irradiation (Kamdem and Grelier 2002). But iron and alum mordants that are used here support the color changing. Between two mordant iron is more effective than alum on discoloration. It was observed that total color differences of untreated Scots pine wood were remarkably higher than beech wood after all exposure periods. While total color difference of untreated beech wood was slowly increased during the all exposure periods, it was sharply increased during all exposure periods for untreated Scots pine.

From the point of sustainability, all compared conventional techniques based on synthetic dyes produced from non-regenerable sources, while in case of natural dyeing; the dyestuffs are extracted from regenerable sources. The possibility of generating the dyeing matter from renewable natural sources makes natural dyes an interesting class of colorants (Bechtold et al. 2003). The results of this study may give an opportunity to other tests to evaluate leachability of these natural colorants under different environmental conditions.

REFERENCES


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