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# DETERMINATION OF THE COLOR STABILITY OF AN ENVIRONMENTALLY-FRIENDLY WOOD STAIN DERIVED FROM OLEANDER (*NERIUM OLEANDER* L.) LEAVE EXTRACTS UNDER UV EXPOSURE

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## ABSTRACT

This study was conducted to develop an environmentally friendly wood stain derived from oleander (*Nerium oleander* L.) and determine the color stability of this stain to UV light irradiation. Wood stains derived from oleander were prepared from aqueous solution with alum and iron mordant mixtures. Yellow Scots pine (*Pinus sylvestris* L.) and Turkish oriental beech (*Fagus orientalis* Lipsky) wood specimens were used for the study. After treatment with the stain, wood panels were exposed to UV light irradiation for periods of 500, 1000 and 1500 hours. Results showed that oleander extract as a stain for wood provided some stability to color change after UV irradiation.

KEY WORDS: wood stain, photodegradation, color changes, natural dyes, oleander (*Nerium oleander* L.)

## INTRODUCTION

Indoor air quality (IAQ) has become an important community concern due to the increased amount of personal time spent in indoor environment. Nowadays, people generally spend more than 80% of their time in an indoor environment such as home, office, car and shopping centre. Some studies showed that the level of pollutants in indoor environment is

actually higher than that in outdoor environment (Maciejewska et al. 2005, Wang et al. 2007). Indoor air pollutants mainly include nitrogen oxides ($NO_x$), carbon oxides (CO and $CO_2$), volatile organic compounds (VOCs), and particulates. There is a variety of sources of VOCs in indoor air. Many VOCs are known to be toxic and considered to be carcinogenic, mutagenic, or teratogenic. These VOCs have a close relation with the sick building syndrome (SBS), which is one of many terms used by occupants to describe symptoms of reduced comfort or health (Wang et al. 2007). These pollutants are emitted from different sources that some of them are floor coverings, wood-based panel, furniture, solid woods, wood stains and paints are to be importance in the these sources (Tab. 1).

*Tab. 1: Classes of some VOCs and their possible emission sources by wood-based products (Cheng and Brown 2003)*

| **VOC class** | **Environment and sources** |
|---|---|
| Aliphatic and cyclic hydrocarbons | Wood-based panel and furniture, paints, |
| Aromatic hydrocarbons | floor coverings, wood-based panel and furniture, paints |
| Aldehydes | floor coverings, wood-based panel and furniture, solid woods, paints |
| Terpenes | wood-based panel and furniture, solid woods, paints |
| Alcohols | floor coverings, wood-based panel and furniture, solid woods, paints |
| Esters | Wood-based panel and furniture, solid woods, paints |
| Halocarbons | Wood-based panel and furniture |
| Glycols/glycolethers/glycolesters | Wood-based panel and furniture, paints |
| Ketones | floor coverings, wood-based panel and furniture, solid woods, paints |
| Alkene | Wood-based panel and furniture |
| Organic acids | Wood-based panel and furniture, solid woods, paints |
| Ethers | Paints |
| Other VOCs | Wood-based panel and furniture |

In general, three methods are suggested to improve the indoor air quality, namely source control, increased ventilation, and air cleaning (Wang et al. 2007). To reduce the amount of VOC in wood coatings, several technologies are currently used. In recent years, wood preservation industry prefers non-chemical based and vegetable based chemicals for wood treatments (Schulta and Nicholas 2000).

"Nowadays, there is a growing interest in the revival of natural stains in textile staining; arguments based around keywords such as sustainability, green chemistry, improved eco-balances and thereby leading to niche products for special markets" (Bechtold et al. 2007). The introduction of natural stains into wood products for staining and finishing processes is one of the alternative solutions for eliminating environmental pollutants. The natural stains are generally environmentally friendly and have many advantages over synthetic stains with respect to production and application (Luciana et al. 1997).

Oleander can be used as a natural stain (Grae 1974) and preservative for wood-based products (Goktas et al. 2007). It is one of the most poisonous plants and contains numerous toxic compounds. The most significant of these toxins are oleandrin and neriine, which are cardiac glycosides. They are present in all parts of the plant, but are most concentrated in the sap. Many of Oleander's relatives have similar leaves and also contain toxic compounds. The leaves and the flowers are cardiotonic, diaphoretic, diuretic, emetic, expectorant and sternutatory (Chiej 1984, Duke and Ayensu 1985). A decoction of the leaves has been applied externally in the treatment of scabies (Chiej 1984) and to reduce swellings (Chopra et al. 1986). The plant is used as a rat poison, a parasiticide (Chiej 1984) and an insecticide (Polunin 1969). The pounded leaves and bark are used as an insecticide (Manandhar 2002). A green dye is obtained from the flowers (Grae 1974, Goktas et al. 2007). Oleander is also known to hold its toxicity even after drying (Inchem 2005).

The objectives of this study were to develop an environmentally-friendly natural wood stain derived from oleander plant and to determine its color stability under UV light irradiation. The CIELab system (CIE 1976 L*a*b*) was used to monitor the color change of the specimens after periods of 500, 1000 and 1500 h of UV light exposure.

## MATERIAL AND METHODS

### Wood specimens

Samples of beech (*Fagus orientalis* Lipsky) and Scots pine (*Pinus sylvestris* L.) sapwood were used in this study. Vertical grain 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 to reach equilibrium moisture content.

### Plant material

The oleanders' leaves and flowers used in this study for decay fungi were collected from the region of Mugla-Turkey in September. The collected samples were air dried and kept in the Herbarium of Muğla University-Turkey.

### Extraction of stain

Air-dried powdered leaves of oleander (800 g) were extracted repeatedly with ethyl alcohol (96%) in a Soxhlet apparatus until the last portion of the extract was colorless. Volume loss due to evaporation was compensated by the addition of alcohol at the end of

the extraction to retain the initial volume. After 3 hours, the suspension was passed through a Büchner funnel filter, and a sample of 500 ml of the filtrate taken. The extracts were evaporated under vacuum to leave a residue, which was maintained at 4 °C until use.

## Stain preparation

Ferrous sulfate ($FeSO_4.7H_2O$ technical grade 96% purity, Merck) and alum ($KAl(SO_4)_2.12H_2O$ puriss. p.a. Fluka) were mixed as a concentrated solution with the addition of mordant (3 % and 5 %) to give a final stain concentration of 30 g $L^{-1}$ and 50 g $L^{-1}$ mordant (Guzel and Akgerman 2000).

The prepared stains were separated into three containers for oleander extract + iron mixtures, oleander extract + alum mixtures and control (no mordant), then heated to 60 °C. The wood panels were immersed into the stain containing the oleander extract for 30 min. Any extra solution, left on the specimens, was removed with a clean cloth. Specimens were then left to dry at 20±3 °C in a vertical position.

## Accelerated lightfastness test

A UVA-351 lamp was used as the irradiation system. The test conditions were 50% relative humidity and 20 °C. Specimens were exposed to UV light directly at a distance of 20 cm with angle of 90E (Kamdem and Grelier 2002). Five replicate samples of each treatment and untreated samples were run for 0, 500, 1000 and 1500 h. These irradiation times were selected randomly. The color of the samples was also measured after each irradiation period.

## Color measurement

Color measurements were determined according to ISO 7724, 1984. The CIELab system (The Commission International de i'Eclairage (CIE) is described by three parameters: $L^*$ axis represents the lightness, $a^*$ and $b^*$ are the chromaticity coordinates; $+a^*$ is for the red, $-a^*$ for green, $+b^*$ for yellow, $-b^*$ for blue and $L^*$ varies from 100 (white) to zero (black).

$L^*$, $a^*$, and $b^*$ color coordinates of each sample were determined before and after exposure to UV light irradiation. The color was measured on a color reader (Konica Minolta-Color Reader CR-10) using a D65 light source and 10 mm for sample diameter. These values were used to calculate the color differences ($\Delta E^*$) as a function of the UV irradiation period according to the following equation:

$$\Delta L^* = L^*_f - L^*_i \quad (1)$$

$$\Delta a^* = a^*_f - a^*_i \quad (2)$$

$$\Delta b^* = b^*_f - b^*_i \quad (3)$$

$$\Delta E^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \quad (4)$$

Where $\Delta L^*$, $\Delta a^*$ and $\Delta b^*$ are the changes between the initial (i) and the final (f) values. $L^*$, $a^*$ and $b^*$ contribute to the color change $\Delta E^*$. The higher the value of $\Delta E^*$ is, the greater the discoloration.

## RESULTS AND DISCUSSION

Color changes of beech and Scots pine wood samples were shown in Tab. 2. and depicted in Figs. 1, and 2. Positive values of $\Delta b^*$ indicate an increment of yellow color and negative values an increase of blue color. Positive values of $\Delta a^*$ indicate a tendency of wood surface to reddish while negative values mean a tendency toward green.

The highest negative value of lightness stability ($\Delta L^*$=7,46) was obtained on the species of Scots pine untreated samples. The negative lightness stability ($\Delta L^*$) values occur during the UV exposure because the surface becomes darker. The reason is that, photodegradation by UV light induces changes in chemical composition, particularly in the lignin, and subsequent color changes (Feist and Hon 1984). This event, occurred by free radicals that generated in wood by light rapidly interact with oxygen to produce hydroperoxides which in turn are easily decomposed to produce chromophoric groups such as carbonyls, carboxyls, quinones, peroxides, hydroperoxides, and conjugated double bonds (Feist and Hon 1984). So, the lowest values of $\Delta L^*$ that is the most sensitive parameter of the wood surface quality on the irradiation (Temiz et al. 2005).

The rate of color change on surface of both wood species that treated/untreated after UV light irradiation occurred rapidly for during the first 500 h period then slowed to only small changes at 1000 to 1500 h. When wood is exposed to the outdoors or in artificial UV light for a relatively short period, changes in brightness and color are readily observed (Feist and Hon 1984). These results were similar to a study of Kamdem and Grelier (2002), and Temiz et al. (2005).

The lowest color changes ($\Delta E^*$ = 2,59) were determined for the beech wood control specimens (without stain) after 1500 h UV exposure. Meanwhile the highest color changes ($\Delta E^*$ = 8,42) were determined on the Scots pine control specimens (without stain) after 1500 h UV exposure. Generally, the color changes values of Scots pine (treated/untreated) were higher than the changes of the beech wood (treated/untreated). In study of Şahin (2002) too, in UV experiments, soft woods showed greater color change than hard woods. The reason for the differences between two wood groups may be due to the differences of chemical compositions of the two different woods (hardwood and softwood) (Temiz et al. 2005, Söğütlü and Sönmez 2006), and interaction of Oleander extract compounds with wood components, resulting in the different photo-degradation effects of UV irradiation. 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). Lignin has aromatic, phenolic and carboxylic groups that absorb rays of different energy levels. Although cellulose is not sensitive to UV light of wavelengths longer than 340 nm (Feist and Hon 1984). Therefore, the color change of Scots pine (soft wood) would have to be higher than the beech wood (hard wood) because of the lignin degradation. As a consequence, most of the components in wood are obviously capable of absorbing enough visible and UV light to undergo photochemical reactions leading ultimately to discoloration and degradation ( Feist and Hon 1984). However, the roughness of color stabilized wood is a complex phenomenon because wood is an anisotropic and heterogeneous material. Several factors, such as anatomical differences, growing characteristics, machining properties, pre-treatments (e.g. steaming, drying, etc.), can effect the color stability (Temiz et al. 2005).

The magnitudes of the color change on all treated specimens, Scots pine, were less when compared to that of untreated samples. However, the color changes on all treated specimens of beech wood were higher when compared to that of untreated samples. Therefore, oleander extract as a stain for Scots pine contributes to the color stability. Although the reason of this color stability on Scots pine cannot be fully explained with current data, this can be attributed to the formation of complexes between oleander extract and wood components. Metal mordants

have been pointed out as the reason for color stability. Because metal ions, promotes free radical formation (Feist and Hon 1984) of wood components even when they are exposed to light. The stabilization of lignin by iron was reported to occur through the formation of complex (Kamdem and Grelier 2002). Ölmez reported that (2004) iron and alum mordants contributed to color stability of wool yarn that was stained by oleander extract. However, in this study, color changes of oleander + alum and iron mordants cannot support the literatures given above.

There can be a concern about poison of oleander that will be a risk for applications on interior wood-based products. It is thought that Oleander may contain many other unknown or un-researched compounds that may have dangerous effects (Inchem 2005). However, all of furniture or wood products have to be coated by coatings. Moreover, the amounts of the poisonous substances that are used in the study were very little. Even so, we know that oleander is used as a drug for miscellaneous pharmaceutical product and other therapeutic preparation (Inchem 2005). Besides, evaluated stain will be a natural wood preservative against to fungi and insects, thanks to poisonous substance of oleander. Nevertheless, there is an absolute necessity that preventive measures have to be executed during application.

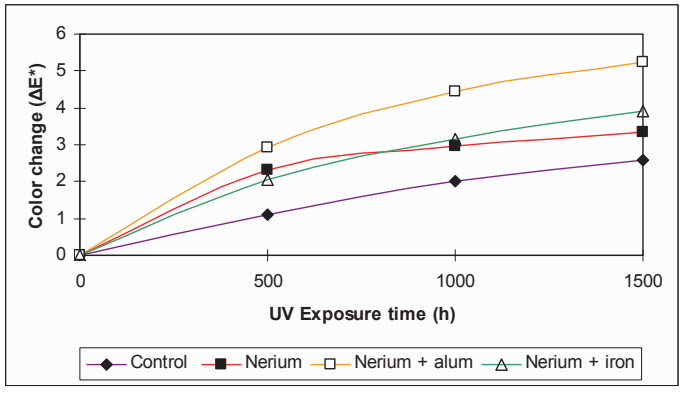


*Fig. 1: Color changes of beech wood exposed to 1500 h UV irradiation*

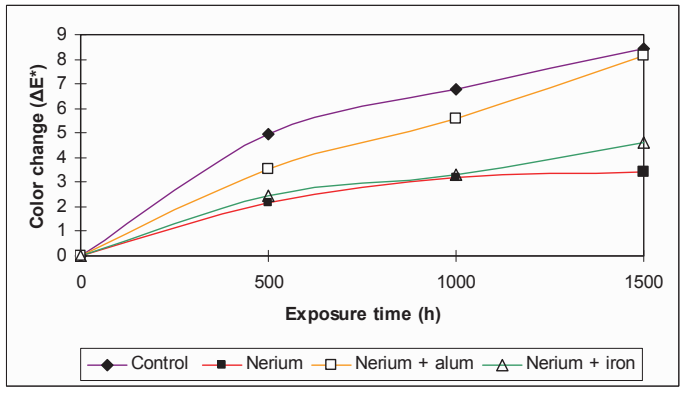


*Fig. 2: Color changes of Scots pine exposed to 1500 h UV irradiation 694-705*

*Tab. 2: Color changes of wood species exposed to 1500 h UV irradiation*

| | | 500 h | | | | 1000 h | | | | 1500 h | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Wood** | **Stain materials** | $\Delta L^*$ | $\Delta a^*$ | $\Delta b^*$ | $\Delta E^*$ | $\Delta L^*$ | $\Delta a^*$ | $\Delta b^*$ | $\Delta E^*$ | $\Delta L^*$ | $\Delta a^*$ | $\Delta b^*$ | $\Delta E^*$ |
| **Beech** | Control (without stain) | -0,86 (2,03) | 0,07 (0,12) | -0,7 (1,20) | 1,11 (0,14) | -1,58 (0,14) | 0,31 (0,28) | -1,19 (1,55) | 2,00 (0,73) | -2,15 (0,42) | 0,46 (0,35) | -1,36 (1,48) | 2,59 (0,26) |
| | Oleander (without mordant) | -1,78 (0,24) | 0,26 (0,38) | -1,48 (0,64) | 2,33 (0,49) | -2,18 (0,53) | 0,48 (0,37) | -0,57 (1,05) | 2,97 (0,93) | -2,52 (0,70) | -0,60 (0,46) | -0,71 (1,28) | 3,33 (1,13) |
| | Oleander + Alum | -2,05 (0,07) | 0,85 (0,63) | -1,90 (2,51) | 2,92 (0,23) | -3,10 (0,14) | 1,25 (0,77) | -2,95 (0,07) | 4,46 (0,26) | -3,55 (0,07) | -1,60 (0,70) | -3,50 (1,14) | 5,24 (0,35) |
| | Oleander + Iron | -1,78 (0,07) | 0,75 (0,14) | -0,71 (0,42) | 2,06 (0,20) | -2,73 (0,14) | 1,30 (0,70) | -0,91 (0,28) | 3,16 (0,23) | -3,33 (0,14) | -1,70 (0,14) | -1,11 (0,07) | 3,90 (0,13) |
| **Scots pine** | Control (without stain) | -4,3 (0,07) | 2,35 (0,63) | 0,66 (0,49) | 4,94 (0,06) | -5,89 (0,56) | 3,33 (0,42) | -0,09 (0,91) | 6,76 (0,60) | -7,46 (0,63) | 3,89 (0,12) | 0,14 (1,27) | 8,42 (0,90) |
| | Oleander (without mordant) | -1,97 (0,76) | 0,75 (0,89) | 0,50 (0,88) | 2,17 (1,07) | -2,88 (0,79) | 1,35 (1,01) | -0,25 (1,03) | 3,19 (1,11) | -3,10 (0,73) | -1,48 (0,96) | 0,07 (1,24) | 3,43 (1,01) |
| | Oleander + Alum | -2,70 (0,14) | 1,90 (0,14) | -1,20 (0,00) | 3,51 (0,18) | -4,10 (0,56) | 3,05 (0,35) | -2,30 (0,84) | 5,60 (0,95) | -5,65 (0,07) | -4,50 (0,00) | -3,80 (0,42) | 8,16 (0,24) |
| | Oleander + Iron | -1,85 (0,49) | 0,55 (0,07) | -1,50 (0,42) | 2,44 (0,61) | -2,45 (0,63) | 0,90 (0,14) | -2,00 (0,56) | 3,29 (0,77) | -3,50 (0,70) | -1,20 (0,14) | -2,75 (0,49) | 4,61 (0,79) |

Values in parentheses are standard deviations.

## CONCLUSIONS

This study deals with UV color stability characteristic of wood stain derived from oleander extract. The stain has been applied in three different forms, which were natural (without mordant), oleander + iron and oleander + alum mixtures. All stain methods showed lower color changes compared to the untreated specimens of Scots pine after 1500 h exposure in UV irradiation test cycle. It is clear that, oleander extracts as a wood stain for Scots pine contributes to stability of color. However, all stain methods showed higher color changes compared to untreated specimens of beech wood. After 1500 h of exposure, beech specimens treated with oleander extract + iron mixture provided the lowest color changes after untreated samples. The results demonstrated that treatments with iron mordant retarded the total color change more compared to the without mordant treatments and alum mordant. Probably this is caused by oleander + iron mordant mixture by forming complexes between units' wood components. Color changes of treated Scots pine specimens were higher than the specimens of beech wood. We deduced that this difference caused by difference of chemical compositions between hardwood and softwood and reception of different UV rays rate

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