Determination of the color stability of an environmentally friendly wood stain derived from laurel (*Laurus nobilis* L.) leaf extracts under UV exposure

Osman Goktas
M. Emin Duru
Mehmet Yeniocak
Ertan Ozen

Abstract

This study was designed to develop an environmentally friendly wood stain derived from laurel (*Laurus nobilis* L.) and determine the color stability of this stain when exposed to UV light irradiation. Wood stains derived from *L. nobilis* were prepared from aqueous solution with alum and iron mordant mixtures. Scots pine (*Pinus sylvestris* L.) and Turkish oriental beech (*Fagus orientalis* Lipsky) wood specimens were used as staining substrates. After treatment with the stain, the wood panels were exposed to UV light irradiation for periods of 100, 200, and 300 hours. Results showed that wood stain derived from laurel (bay) extract provided some color stability after UV irradiation.

People are exposed to numerous types of pollutants in modern environments. Wood furniture and decorative elements are potential sources for a number of volatile organic compounds (VOCs) indoors. Salthammer et al. (1998) has identified about 150 different VOCs used in the wood finishes and coatings industry. These chemical compounds can be used to impart desirable properties such as dimensional stability, water repellency, fire resistance, color, odor, surface hardness, and mechanical strength. A large number of preservative and stain compounds have been introduced to the market; however, many of them have not gained commercial acceptance due to chemical toxicity, low efficacy, high cost, or corrosiveness (Murphy 1990). Most compounds in preservatives and stains belong to the typical group of solvents that are chemically inert under normal conditions. However, a number of substances used for wood stains and preservatives are known as secondary emission products or reactants (Salthammer et al. 2002). These persistent pollutants and their potential inclusion in waste wood have raised concerns about their health effects on humans and wildlife (Asari et al. 2004).

Many stains from synthetic sources can be allergenic and harmful to humans, thus interest in natural stains has increased considerably during the last few years (Bhattacharya et al. 1998, Deo and Desai 1999). Natural stains are generally environmentally friendly and have many advantages over synthetic stains with respect to production and application (Angelini et al. 1997). "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 solution for eliminating environmental pollutants.

A number of stain crops continue to be grown in the Mediterranean region. These plant species are mostly native to the Mediterranean region. One example is common laurel (*Laurus nobilis* L.), which is used in dyeing wool, silk, linen fibers, and cotton (Ölmez 2004).

Laurel (bay) (*Laurus nobilis* L.) is cultivated in many temperate and warm parts of the world, and its leaves and berries

The authors are, respectively, Associate Professor, Dept. of Wood Sci. and Furniture Design (osmangoktas65@yahoo.com), Assistant Professor, Dept. of Chemistry (eminduru@hotmail.com), Graduate Student, Dept. of Wood Sci. and Furniture Design (mehmetyeniocak@yahoo.com), and Research Assistant, Dept. of Wood Sci. and Furniture Design (eozenertan@hotmail.comr), all at Mugla Univ., Mugla, Turkey. This paper was received for publication in April 2007. Article No. 10344.

Forest Prod. J. 58(1/2):77-80.

are generally widely utilized (Kilic and Altuntas 2006). Laurel leaf traditionally has been used as an herbal medicine, and recent research has revealed that it can be used in treating diabetes and preventing migraines (Fang et al. 2005). Laurel is one of the most widely used culinary spices in all western countries. The essential oil from the leaves (0.8 to 3%) contains mostly 1.8-cineol (50%). The food industry uses the essential oil as seasoning for meat, soup and fish, while other industries use it as a fungus inhibitor and an insect repellent (Baratta et al. 1998). Also found in laurel are eugenol, acetyl eugenol, methyl eugenol, α- and β-pinene, phellandrene, linalool, geraniol and terpineol (Ölmez 2004). Because of these ingredients, the laurel extracts used in this study serve both as a stain and as a preservative.

The objectives of this study were to develop an environmentally friendly natural wood stain derived from laurel plant foliage and to determine its color stability under UV light irradiation. Color measurements were determined according to ISO 7724-2 (1984) after periods of 100 hours, 200 hours, and 300 hours of UV light exposure.

Materials 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 10 mm (radial) by 100 mm (tangential) by 150 mm (longitudinal) were cut and stored in the laboratory at 20±2 °C and 65±5 percent relative humidity to reach equilibrium moisture content (EMC).

Plant material

Laurus nobilis L. (*Lauraceae*) leaves were collected from the Hatay region in Turkey during April and May. The specimens were identified using the Flora of Turkey (Davis 1982). The voucher specimens (No. L-1001) were identified by Prof. R. Mammadov, a taxonomist in the Department of Biology, Faculty of Arts and Sciences, at Pamukkale University in Turkey.

Extraction of stain

Air-dried powdered leaves of *L. nobilis* (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 for 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 500 mL sample of the filtrate was taken. The extracts were evaporated under vacuum conditions, which resulted in a residue that was maintained at 4 °C until use.

Stain preparation

Ferrous sulfate ($FeSO_4.7H_2O$ technical grade 96 percent purity (Merck 1996)) 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: one for the laurel extract + iron mixture; one for the laurel extract + alum mixture; and one for laurel extract with no mordant. Then the solutions were heated to 60 °C and a wood panel of each type, beech and Scots pine, was immersed into each stain solution for 30 minutes (Sonmez and Budakci 2004). 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. Any variation in color difference value (ΔE*) was used to establish a correlation between the accelerated lightfastness testing (Hui and Chang 2001). The average irradiance was about 330 nm at 50 percent relative humidity and 20 °C. Specimens were directly exposed to UV light at a distance of 20 cm and an angle of 90E (Kamdem and Grelier 2002). Five replicate samples treated with each stain solution and untreated controls were run for the randomly selected irradiation times of 0 (no irradiation), 100 hours, 200 hours, and 300 hours The color of the samples was measured after each irradiation period.

Color measurement

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

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 a sample diameter of 10 mm. These values were used to calculate the color differences (ΔE*) as a function of the UV irradiation period according to the following equations:

$$\Delta L^* = L^*_f - L^*_i \quad [1]$$

$$\Delta a^* = a^*_f - a^*_i \quad [2]$$

$$\Delta b^* = b^*_f - b^*_i \quad [3]$$

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

ΔL*, Δa*, and Δb* are the changes between the initial (i) and the final (f) values L*, a*, and b*. These calculated changes from Equations [1] through [3] contribute to the overall color change ΔE* as calculated in Equation [4]. Higher ΔE* values represent greater discoloration.

Results and discussion

Color changes of beech and Scots pine wood samples are shown in **Table 1** and depicted in **Figures 1** and **2**. Positive values of Δb^* indicate increased yellow color and negative values indicate increased blue color. Positive values of Δa^* indicate increased red color while negative values indicate increased green color.

The lowest ΔL^* values were obtained with untreated (without stain) Scots pine and beech wood specimens after 100 hours, 200 hours, and 300 hours of UV exposure. All other treatments caused smaller changes in the lightness (ΔL^*) than the control specimens. The negative lightness stability (ΔL^*) values occur during UV exposure because the surface becomes darker due to photodegradation. Photodegradation occurs when UV light induces changes in chemical composition, particularly in the lignin, causing subsequent color changes

Table 1. — Color changes of wood species exposed to 300 hours UV irradiation.

Wood	Stain materials	Before exposure			100 hours				200 hours				300 hours			
		L*	a*	b*	ΔL*	Δa*	Δb*	ΔE*	ΔL*	Δa*	Δb*	ΔE*	ΔL*	Δa*	Δb*	ΔE*
Beech	Control (without stain)	68.95	10.97	24.6	−4.34	1.21	−0.17	4.50	−4.95	1.51	−0.85	5.24	−5.27	1.75	−0.81	5.61
	Laurel (without mordant)	68.62	7.76	21.34	0.08	2.9	0.92	3.04	−0.68	3.34	0.90	3.52	−1.24	3.68	1.38	4.12
	Laurel + alum	65.72	4.90	29.74	−1.20	3.56	1.08	3.90	−0.72	3.44	0.68	3.57	−0.88	3.66	0.94	3.87
	Laurel + iron	60.86	8.00	20.38	0.34	2.50	0.78	2.64	−0.42	2.70	0.62	2.80	−0.74	3.36	1.36	3.69
Scots pine	Control (without stain)	79.70	7.51	24.98	−5.95	2.87	−0.7	6.64	−6.58	3.62	−0.88	7.56	−7.16	3.78	0.13	8.09
	Laurel (without mordant)	77.54	5.08	31.52	−2.14	4.3	−1.92	5.17	−3.68	5.06	−3.02	6.94	−4.08	6.24	−1.94	7.70
	Laurel + alum	76.46	4.66	32.08	−1.62	4.76	−3.66	6.21	−2.84	5.6	−3.32	7.10	−2.72	5.14	−2.46	6.31
	Laurel + iron	73.58	4.9	29.74	−3.02	4.26	−2.96	6.00	−3.48	4.24	−2.08	5.86	−4.18	5.78	−1.34	7.25

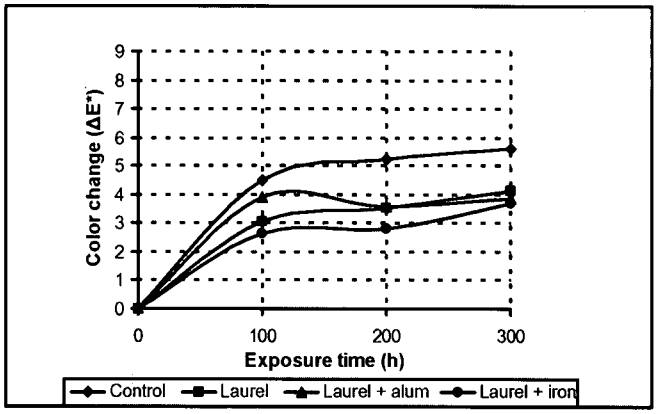

Figure 1. — Color changes of beech after UV exposure.

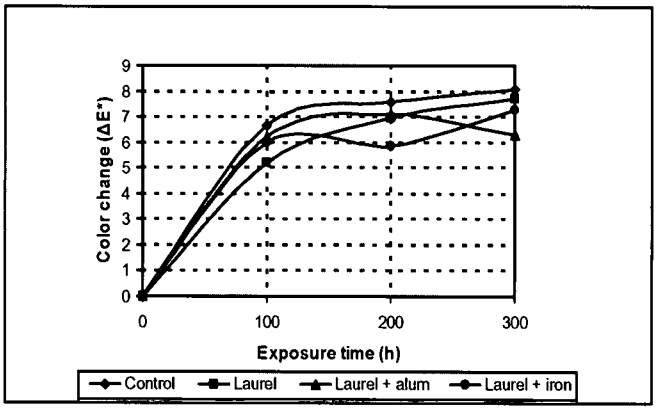

Figure 2. — Color changes of Scots pine after UV exposure.

(Feist and Hon 1984). During this event, light generates free radicals in wood by rapidly interacting with oxygen to produce hydroperoxides, which are easily decomposed to produce chromophoric groups such as carbonyls, carboxyls, quinones, peroxides, hydroperoxides, and conjugated double bonds (Feist and Hon 1984). Thus, the lowest ΔL^* values indicate the highest sensitivity of wood surface quality to UV irradiation (Temiz et al. 2005).

When wood is exposed to the outdoors or artificial UV light for a relatively short period, changes in brightness and color are readily observed (Feist and Hon 1984).The rate of color change after UV light irradiation on the surface of both wood species, regardless of stain type, occurred rapidly during the first 100 hours period then slowed to only small changes during the 100 hours to 200 hours period. These results were similar to a study by Kamdem and Grelier (2002), where almost 80 percent of the final ΔE* value was achieved during the first 100 hours of exposure and only about 20 percent in the following 200 hours.

The smallest color changes ($\Delta E^* = 3.69$) were found in the beech specimens treated by laurel + iron stain after 300 hours of UV exposure. The largest color changes ($\Delta E^* = 8.09$) were found in the untreated Scots pine specimens. The magnitude of the color change on all treated specimens of both woods was less than that of the untreated specimens. Therefore, laurel extract as a stain for wood contributes to color stability. Although the reason for this color stability cannot be fully explained with current data, it may be attributable to the formation of complexes between laurel extract and wood components. Another reason for this color stability may be the presence of mordants, which are metal ions that promote free radical formation of wood components even when exposed to light (Feist and Hon 1984). The stabilization of lignin by iron has been found to occur through the formation of a complex (Kamdem and Grelier 2002). It has also been found that iron and alum mordants contribute to the color stability of wool yarn stained by laurel extract (Ölmez 2004).

Generally, the color changes values of Scots pine (treated or untreated) were higher than the color changes of beech wood (treated or untreated). UV experiments by Şahin (2002) also showed greater color changes in soft woods than in hard woods. This may be due to the different chemical compositions of the two different wood types (hardwood and softwood) (Temiz et al. 2005, Söğütlü and Sönmez 2006), and their interactions with laurel extract compounds, resulting in different photodegradation effects from UV irradiation. Soft woods generally have 2 to 10 percent more lignin than hard woods. Of the major wood constituents, lignin contributes 80 to 95 percent of the UV absorption coefficient of wood (Tereza et al. 2004). Lignin has aromatic, phenolic and carboxylic groups that absorb rays of different energy levels. In contrast, 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 be higher than that of beech wood (hard wood) because of the higher level of lignin degradation. Most wood components are capable of absorbing enough visible and UV light to undergo photochemical reactions ultimately leading to discoloration and degradation (Feist and Hon 1984). However, the color stability of wood is a complex phenomenon because wood is an anisotropic and heterogeneous material. Several factors such as anatomical differences, growing characteristics, machining properties,

and pretreatments such as steaming and drying can affect color stability (Temiz et al. 2005).

Conclusion

This study concerned color stability after UV exposure of wood stain derived from laurel extract. The stain was applied in three different forms: natural laurel (without mordant); laurel + iron, and laurel + alum. After 300 hours of UV irradiation, all staining methods showed smaller color changes than the untreated specimens of beech and Scots pine. It is clear that laurel extract as a wood stain contributes to color stability. After 300 hours of exposure, beech specimens treated with the laurel extract + iron mixture provided the smallest color changes. The results demonstrated that treatments with either mordant reduced the total color change more than the laurel without mordant treatments. This is probably caused by laurel + mordant mixtures forming complexes with wood components. The results also showed that color changes of treated Scots pine specimens were greater than in beech wood specimens. This difference is likely caused by the different chemical compositions between hardwood and softwood and the resultant effect on the rate of absorption of different UV rays.

Literature cited

Angelini, L., L. Pistelli, P. Belloni, A. Bertoli, and S. Panconesi. 1997. Rubia tinctorium a source of natural dyes: Agronomic evaluation, of alizarin and industrial assays. Industrial Crops and Products 6:303-311.

Asari, M., H. Takatsuki, M. Yamazaki, T. Azuma, H. Takigami, and S. Sakai. 2004. Waste wood recycling as animal bedding and development of biomonitoring tool using the CALUX assay. Environment Inter. 30(5):639-649.

Baratta, M.T., H.J.D. Dorman, S.G. Deans, D.M. Biondi, and G. Ruberto. 1998. Chemical composition, antimicrobial and antioxidative activity of laurel, sage, rosemary, oregano and coriander essential oils. J. of Essential Oil Res. 10:618-627.

Bechtold, T., A.M. Ali, and R. Mussak. 2007. Natural dyes for textile dyeing: A comparison of methods to assess the quality of Canadian golden rod plant material. Dyes and Pigments 75(2):287-293.

Bhattacharya, N., B.A. Doshi, and A.S. Sahasrabudhe. 1998. Dyeing jute with natural dyes. Am. Dystuff Reporter. 87(4):26-29.

Davis, P.H. 1982. Flora of Turkey and the east Aegean. 7:534-535.

Deo, H.T. and B.K. Desai. 1999. Dyeing cotton and jute with tea as a natural dye. Coloratiopon Technology 115:224-227.

Fang, F., S. Shengmin, Y.C. Kuang, G. Alexander, H. Chi-Tang, and T.R. Robert. 2005. Isolation and identification of cytotoxic compounds from Bay leaf (*Laurus nobilis*). Food Chem. 93(3):497-501.

Feist, W.C. and D.N.-S. Hon. 1984. Chemistry of weathering and protection. *In*: The Chemistry of Solid Wood. R.M. Rowell, ed. Advances in Chemistry Series, No. 207. American Chemical Soc., Washington, D.C. Chapter 11, pp. 401-451.

Guzel, B. and A. Akgerman. 2000. Mordant dyeing of wool by supercritical processing. J. of Supercritical Fluids 18(3):247-252.

Hui, T. and C.T. Chang. 2001. Correlation between softwood discoloration induced by accelerated lightfastness testing and by indoor exposure. Polymer Degrad. and Stabil. 72:361-365.

ISO 7724-2. 1984. Paints and Varnishes—Colorimetry. Part 2. Color Measurement. ISO Standard 7724-2. ISO Standard, Geneva, Switzerland.

Kamdem, P.D, and S. Grelier. 2002. Surface roughness and color change of copper amine and UV absorber-treated red maple (Acer rubrum) exposed to artificial ultraviolet light. Holzforschung 56(5):473-478.

Kilic, A. and E. Altuntas. 2006. Wood and bark volatile compounds of *Laurus nobilis* L. Holz als Roh- und Werkstoff 64(4):317-320.

Merck. 1996. The Merck Index, 11th Ed. Merck Co. Inc., Rahway, New Jersey. pp. 46.

Murphy, R.J. 1990. Historical perspective in Europa. *In*: Proc. of First Inter. Conf. on Wood Protection with Diffusible Preservatives, Margaret Hamel, ed. Nov. 28-30, 1990, Nashville, Tennessee. pp. 9-13.

Ölmez, F.N. 2004. Colors and fastness values obtained from laurel (Laurus nobilis L.) in different boiling times. J. of Agr. Sci. (Univ. of Yuzuncu Yil.) 14(1):35-40.

Salthammer, T., M. Bednarek, F. Fuhrmann, R. Funaki, and S.I. Tanabe. 2002. Formation of organic indoor air pollutants by UV-curing chemistry. J. of Photochemistry and Photobiology 152(1-3):1-9.

_____, A. Schwarz, and F. Fuhrmann. 1998. Emission of reactive compounds and secondary products from wood-based furniture coatings. Atmos. Environ. 33:75-84.

Sönmez, A. and M. Budakci. 2004. Process of woodworking Surfaces II. Sevgi Ofset. Ankara.

Söğütlü, C. and A. Sönmez. 2006. The effect of UV lights on color changes on some local wood processed with differential preservatives. J. of Faculty of Engineering and Architecture of Gazi Univ. 21(1):151-159.

Şahin, T.H. 2002. The studies of color changes in wood and cellulose. J. of Forest Faculty (Suleyman Demirel Univ.) 2. pp. 57-70.

Temiz, A., U.C. Yildiz, A. Ismail, E. Morten, A. Gry, and G. Colakoglu. 2005. Surface roughness and color characteristics of wood treated with preservatives after accelerated weathering test. Appl. Surf. Sci. 250(1-4):35-42.

Tereza, C.M.P., K.O. Santos, and J.C. Rubim. 2004. A spectrocolorimetric study on the effect of ultraviolet irradiation of four tropical hardwoods. Bioresource Tech. 93:37-42.