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COMPARATIVE STUDY OF WOOD COLOR STABILITY USING ACCELERATED WEATHERING PROCESS AND INFRARED SPECTROSCOPY

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ABSTRACT

The objective of this study was to investigate effects of artificial weathering on color stability of six tropical wood species: *Apuleia leiocarpa* (Vog.) Macbride, *Bagassa guianensis* Aubl., *Dipteryx odorata* (Aubl.) Wild., *Hymenea courbaril*/Linn., *Manilkara bidentata* A. Chev., *Tabebuia* sp. Chemical composition of weathered wood was also studied by FT-IR spectra. With the progressive artificial weathering color of wood changed gradually. *Hymenea courbaril* Linn. wood occurred the greatest range of the total color change. The largest changes in intensity of the wood color took place at the beginning of artificial weathering process. FT-IR analysis indicated the occurrence of lignin and hemicellulose oxidative changes resulting in formation of carbonyl and carboxyl compounds during weathering. Oxidation products are the main cause of wood surface discoloration. Additionally, changes suggesting depolymerisation of cellulose were identified as well.

KEYWORDS: Color stability, accelerated weathering, CIE LCh, FTIR, tropical wood, infrared spectroscopy.

INTRODUCTION

Variability of weather conditions such as solar radiation, changes in temperature or humidity as well as prolonged exposure to the above mentioned environmental factors causes process called weathering also referred to as "aging"(Hon et al. 1986). Since color changes under exploitation of wood material are one of the most crucial and important factors in determining its usage, creating appropriate color information databases is highly useful. Behavior of wood under aging processes has already been described in detail in literature (Hon et al. 1986, Colom et al. 2003, Oltean 2009). However, knowledge in this field of expertise is still incomplete, mainly due to the fact that a number of new wood species have been introduced into the European market in recent years. Most known studies either concentrates on examining wood types that can be found in moderate climate zone or investigate surface discoloration of just a small number of tropical wood species native to South America and Asia (Sudiyani et al. 2003, Baar and Gryc 2012). Difference in wood structure and its chemical composition between tropical and moderate zone wood species points that impact of weather condition has different effect.

The main aim of this research was to determine and compare the influence of artificial weathering on color change of six species of tropical wood (assuming the absence of biotic interactions) from South America were undertaken. The artificial weathering method simulates natural environmental conditions (the typical temperate climate). The research includes wood commercially available in Europe. As the process of artificial weathering was progressing, changes in color of tested wood species were determined. Process of changes was monitored with FT-IR spectrometer with attenuated total reflectance unit.

MATERIAL AND METHODS

Six species of tropical wood from South America were used in this study: garapa (*Apuleia leiocarpa* (Vog.) Macbride), tatajuba (*Bagassa guianensis* Aubl.), cumaru (*Dipteryxodorata* /Aubl.) Wild.), courbaril (*Hymenea courbaril* Linn.), massaranduba (*Manilkara bidentata* A. Chev.), ipe (*Tabebuia* sp.). Samples of each type of wood were collected from one board to obtain "identical sample". Thanks to that samples were similar and a structure were kept in order to appearing changes in the weathering process were a main factor deciding on the examined properties. Dimensions of samples were 20 x 20 x 30 mm (R x T x L). Prior to the determination of properties, each group was conditioned in air at a temperature close to 20°C (±2°C) and relative moisture content (RMC) around 60% (±5%).

The artificial weathering cycle was based on literature (Kaminski et al. 1982, Follrich 2011). The same procedure was used in previous research (Jankowska and Kozakiewicz 2014). One artificial weathering cycle took 30 hours and was separated into three steps. The first step was soaking specimens in water at 20°C (16 h). The conditions of second step (8 h) were 70°C and 5-10% moisture content and the third step was performed at 30°C and 20-25 RMC (6 h) with irradiation with UV rays. Four fluorescent lamps100R's Lightech of 100 W each, and the spectrum 300 - 400 nm (90% of the radiation spectrum is a wavelength of 340 -360 nm) were used for irradiating.

The analyses of color wood changes were made according to ISO 7724-3 (1984). The spherical SP60 Spectrophotometer was used in this research. To determine differences in color three parameters L*, C*, h (L* - coordinate of lightness, C* - chroma coordinate, h - hue coordinate) were used. The total color difference ΔE between the two colors was calculated using the Eq:

$$\Delta E^* = [(\Delta L^*)^2 + (\Delta C^*)^2 + (\Delta h)^2]^{1/2},$$

where ΔL^*, ΔC^*, Δh - represent the differences values between the original and the final coordinates, before and after artificial weathering.

To keep the natural color, wood samples had been isolated from direct sunlight until the first test was obtained. The surface color of samples was measured before the start of artificial weathering and during this process. Six measurements were carried out on each sample (measurements were carried out on longitudinal sections).

Spectra of unprocessed and artificially weathered wood were registered in mid infrared range (700 – 400 cm^{-1}), using Thermo Fisher Scientific Inc. Nicolet 6700 FTIR spectrometer with ATR (attenuated total reflectance) unit. Each spectrum was subjected to data pre-processing techniques (baseline and offset corrections, Savitzky-Golay smoothing) with the use of GRAMS AI software. Integral intensities of spectral bands (peak areas), selected during visual analysis, were calculated in GRAMS/AI software. Relative intensity with respect to band at 1033 cm^{-1}, was determined according to methodology proposed by Feist et al. (1986).

RESULTS AND DISCUSSION

The artificial weathering resulted in changes of the appearance of wood samples. Different wood species showed a different behavior of color changes. The first cycles of artificial weathering caused no obvious change in color, but after each stage of the process differences were observed - samples became darker, which was probably the result of dissolved dye substances in deeper layers, and then depositing them on the surface of wood during drying. Next cycles of ageing caused a gradual leaching of dye substances, causing brightening the color of wood. Irradiation with ultraviolet light also had an impact on changing the color of weathered wood. The weathering process with soaking in water of the small specimens resulted in the extraction of polar wood extractives, and that this procedure overlaps with the UV-irradiation. According to the literature (Hon et al. 1986), UV radiation causes decomposition of lignin especially in the surface layer, what causes the yellowing or browning of wood. With the increasing number of aging cycles, contained in the wood dyes washed out and finally the amount of dyes in top layers of wood has become very limited and wood color became a little paler (decrease parameter C^*). Moreover, wood yellowing was observed (the increase in the value of a parameter h to values describing the color tones of yellow - 90°). The biggest scope of changes color hue occurred in tatajuba, garapa and courbaril wood (Fig. 1). Resultant of change in lightness/brightness, chroma and color hue is total color change (ΔE). Nature of change was the same for all studied wood species. Only the scope of changes was different. In the first fifty cycles of artificial aging increase in the parameter ΔE was observed. After an initial increase total color change (to 30-40 ageing cycles), decrease in ΔE was observed. The highest intensity of the color changes was observed during the initial phases of weathering process. The greatest scope of total color change (ΔE) was found in case of courbaril (the maximum value of ΔE was 61.6), in case of garapa the maximum value of ΔE was 29.7 and tatajuba wood 28.6. The smallest scope of color changes was found in case of ipe wood (the maximum value of ΔE was 7.2). The findings generally support former studies dealing with European wood species (Colom et al. 2003, Oltean 2009, Filson et al. 2009).

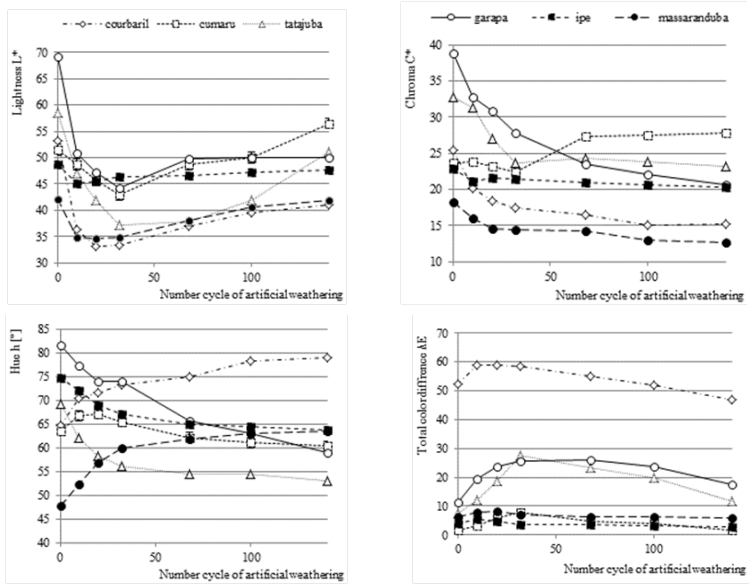

Fig. 1: Changes of color parameters and the total color difference during artificial weathering.

Spectral analysis indicated that major chemical changes occurred during weathering treatment (Tab. 1). In all studied wood species the peak position of the band at 1735 cm^{-1} shifted to higher wavelengths, changes in the intensity of the band were also observed. The courbaril wood showed the greatest scope of changes. In case of cumaru wood the relative intensity of the band decreased after weathering treatment whereas in all other studied wood species the relative intensity increased. In spectral range 1740–1720 cm^{-1} various overlapping stretching vibrations of bond C=O in carbonyl and carboxyl compounds can occur, the absorption near 1735 cm^{-1} can be attributed to vibrations of C=O in xylan units of hemicellulose, as well as vibrations of bond C=O in lignin oxidation products (for example carboxylic acids). The occurrence of oxidative changes is confirmed by the increase in intensity of band at 1630 cm^{-1} in samples of all studied species. This absorption is due to C=O carbonyl streching in conjugated systems (aromatic compounds) which would suggest the formation of carbonyl compounds (Sudiyani et al. 1999). According to previous studies oxidation products are the main factor in discoloration of wood during weathering (Feist et al. 1986). After weathering treatment the relative intensity of band at 1505 cm^{-1} decreased, the absorption at this wavelength is due to skeletal stretching vibration of C=C in the aromatic ring of lignin. Similar results were reported by Sudiyani et al. (1999), Sudiyani et al. (2003). In the case of all studied wood species, the decrease in relative intensity of band at 1455 cm^{-1} was observed, also the peak of band slightly shifted toward lower wave lengths. Except for wood courbail, the band at 1422 cm^{-1} decreased in relative intensity and shifted toward higher wavelengths. In wood courbail small increase of absorption and shift toward lower wavelengths was observed at 1422 cm^{-1}. The bands at 1455 cm^{-1} and 1422 cm^{-1} are due to bending vibrations of C-H bond in aliphatic chain.

Tab. 1: The values of relative intensity of bands in studied spectral range.

Wood species	Testingtime	Wavenumbers (cm^{-1})										
		1725	**1630**	**1590**	**1505**	**1455**	**1422**	**1259**	**1230**	**1202**	**897**	**830**
garapa	before weathering	1.49	1.36	2.07	1.04	1.00	0.83	0.85	1.14	0.57	0.85	1.42
	after weathering*	1.67	1.69	2.36	0.89	0.86	0.81	0.83	0.83	0.41	0.83	1.24
tatajuba	before weathering	1.40	1.30	2.09	1.04	1.07	0.95	1.23	0.82	0.41	0.82	1.64
	after weathering*	1.62	1.97	2.97	0.99	0.93	0.93	0.75	0.75	0.75	1.28	1.71
cumaru	before weathering	1.42	1.40	2.31	1.18	1.23	1.05	1.49	1.49	0.75	0.75	1.49
	after weathering*	1.02	1.78	2.14	0.12	0.59	0.86	0.00	0.00	0.00	0.00	0.00
courbaril	before weathering	1.51	1.32	2.00	0.97	0.95	0.84	0.84	1.17	1.17	1.17	1.56
	after weathering*	1.94	2.34	3.56	0.89	0.87	0.91	0.91	0.72	0.72	0.72	1.45
massaranduba	before weathering	1.54	1.41	2.13	1.05	1.05	0.93	0.93	0.85	1.28	1.28	1.71
	after weathering*	1.88	1.77	2.29	0.77	0.80	0.79	0.79	0.68	0.68	0.68	1.37
ipe	before weathering	1.54	1.43	2.07	1.06	1.02	0.90	0.85	1.28	0.43	1.28	1.28
	after weathering*	1.93	1.81	2.16	0.74	0.78	0.78	0.66	0.66	0.66	0.66	1.32

* 140 cycles of artificial weathering were conducted.

Shifts of bands in the range 1300-1347 cm^{-1}, attributed to wagging vibrations of CH_2 group in cellulose, were identified. For wood courbaril the decrease in relative intensity of band at 1335 cm^{-1} (in-plane bending vibration of O-H in cellulose) was also observed. The relative intensity of bands at 1259 cm^{-1} and 1230 cm^{-1}, notably decreased after weathering. Absorption of infrared radiation in the range 1290 – 1200 cm^{-1} is due to streching C-O vibrations in lignin (guaiacyl units) and hemicellulose (acetyl and carboxyl groups in xylans). Similar results were reported by Sudiyani et al. (1999) and Sudiyani et al. (2003). Bands in spectral range 1130-930 cm^{-1}, atributed to C-O streching vibrations in cellulose and hemicellulose, evidently shifted during weathering but no significant change was noted in relative intensity. After weathering treatment the decrease in integral intensity of band at 897 cm^{-1}, attributed to out of plane C-H ring deformation in cellulose was observed.

CONCLUSIONS

Purpose of the study was to investigate effects of artificial weathering on color stability in six tropical wood species from South America and by using spectrophotometer we were able to obtain general view of tropical wood behavior under artificial weathering processes. Furthermore we were able to observe that wood species the longer exposed to artificial weathering processes the further the change of color would proceed, initially turning dark and then lighter. These processes differed in case of tested wood species and took place with varying intensity. For instance *Hymenea courbaril* Linn. (courbaril wood) presented the greatest range of the total color change. Another finding was that in the first stages of weathering process wood surface discolored the most rapidly. FT-IR analysis, which was carried out indicated the occurrence of lignin and hemicelluloses oxidative changes resulting in formation of carbonyl and carboxyl compounds during weathering process. Additionally changes suggesting depolymerisation of cellulose were identified as well. Moreover spectral analysis results were helpful in determining spectral range suitable for monitoring chemical changes of studied wood during weathering processes.

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