Color Adjustment of Black Locust (*Robinia pseudoacacia* L.) Wood by Producing Chromogenic Structure under Heat Treatment

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To improve the appearance of the black locust wood under heat treatment, ammonia (AM), acetic acid (AA), and ammonium acetate (AMA) were adsorbed into the black locust wood flour, with or without aluminum sulfate octadecahydrate (ASO). The color parameters (CIE $L^*a^*b^*$) of the black locust wood flour treated with different conditions were measured. Diffuse reflectance ultraviolet-visible spectroscopy (DRUV-Vis) and the attenuated total reflection/Fourier transform infrared spectroscopy (ATR-FTIR) were conducted to reveal the changes of the chromophore systems. X-ray photoelectron spectroscopy (XPS) was used to demonstrate the extreme valence of the atomic content and the occurrence of a coordination reaction. The color of the wood flour became reddish after the treatment with AMA+ASO. There was an increase in carbonyl structure and carboxyl structure of the wood flour treated with AMA and ASO.

Keywords: Discoloration; Black locust; Chromophore system; Wood modification

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INTRODUCTION

Black locust (*Robinia pseudoacacia* L.) is a deciduous tree native to North America. In Asia and Europe, it is one of the three most successful tree species introduced in the world alongside poplar and eucalyptus trees (Barrett *et al.* 1990). The black locust wood is medium-hard, with a fine texture, fine shrinkage, high strength, and wear and corrosion-resistant properties. It can be used for construction, furniture, pillars, and shipbuilding (Sargent 1905).

The color of wood is an important indicator when evaluating the surface properties and qualities of the wood, and it is the most direct factor affecting consumers' impression of the wood products (Hidayat *et al.* 2017). In traditional markets, reddened hardwoods usually dominate. The color of the heartwood of the black locust is a dark green (Fan *et al.* 2010), which does not meet the needs of the market consumers. This greatly limits its use (Maunu 2002; Sandoval-Torres *et al.* 2010). The color of the black locust wood changes to a darker color under heat treatment (Chen *et al.* 2012). However, the heat-treated wood always suffers from reduced mechanical properties (Esteves and Pereira 2008). It is necessary to find an effective and environmentally friendly way to make the color of wood more favorable during the wood drying process. The color of wood is the combined result of absorption and reflection of visible light (wavelength between 380 to 780 nm) by a chromonic system composed of chromophore groups and auxochrome groups. The wood structure contains various chromophore groups such as the carbonyl group (C=O), vinyl group (C=C), and coniferaldehyde group, which, when conjugated with other double bonds, absorbs part of the visible light and gives the wood a certain color. When the above structures are bonded to an auxochrome group such as OH, -OR, -NH₂, the light absorption moves to a longer wavelength, and the wood color is deepened (Hon *et al.* 2000). Wood components (such as polyphenols) are prone to react with transition metal ions to form coordination compounds, resulting in changes in the wood color (Falkehag 1966). Adjusting the chromophore and the auxochrome through physical or chemical means is a new method to optimize the color of the wood (Mooktzeng and Zulkifli 2018). Through metal coordination, the relevant groups in the ligand are absorbed into the components of the black locust with the metal ion acting as a bridge. The conjugated chromophore/ auxochrome structure is adjusted and modified in the direction of color darkening, which is important to improve the utilization value of the black locust and the efficient usage of forest resources.

Aluminum ion, as a commonly used complexing agent, has been widely used in wood modification because it is colorless and can improve the dimensional stability (Gao *et al.* 2017) as well as flame retardancy (Xiao *et al.* 2016). In the textile industry, alum (an aluminum salt) is a commonly used mordant. Metal ions act as central ions and combine with dyes and ligands on the fibers to form complexes that make fiber dye well. Metal ions are mainly complexed with oxygen-containing ligands in wood, such as carboxyl groups, carboxylates, hydroxyl groups (Tiemann *et al.* 2002; Mohan *et al.* 2007), and lignin phenolic hydroxyl groups. At the same time, ammonium acetate, having both a chromophore group (—C=O) and an auxochrome group (-NH₂), was used as the impregnation solution. In the present article the term "auxochrome" is employed in a broader sense, indicating a substance that shifts color, regardless of the mechanism.

This study aimed to improve red/green value (a^*) through producing chromogenic and auxochrome structures under heat treatment by chemical adsorption. It would make the color of black locust closer to that of rosewood. In addition, the transformation of its chromophore/ auxochrome structure was also analyzed to explain the color changing mechanism.

EXPERIMENTAL

Wood Samples

Black locust wood were collected from Guangxi Province, southwest China. The wood logs were cut and ground. The 40- to 60-mesh (250 μ m to 425 μ m) wood flour was collected and air-dried to a moisture content of 7% to 12% before further experimentation. *Pterocarpus macrocarpus* Kurz (PMK) heart wood collected from Guangxi Province, southwest China was employed as the target color.

Chemical Adsorption Treatment

Ammonia (AM), acetic acid (AA), ammonium acetate (AMA), and aluminum sulfate octadecahydrate (ASO) (analytical grade reagents) were dissolved in distilled water, respectively, and stirred at 20 °C. The wood flour was impregnated in each solvent with a concentration of 0.1 M under a vacuum (12 h at 0.8 MPa). Five replicates were prepared for each treatment method. The adsorption treated samples were heated in an oven for 12 h at 90 °C. Figure 1 illustrates the scheme of the adsorption experiment of ASO and AMA on wood components. It was worth noting that the pH of all treatment solutions is less than 3, and the dominant soluble species in these solutions is Al ³⁺ (Duan and Gregory 2003).



Fig. 1. Scheme of the adsorption experiment of ASO and AMA on wood components

Color Measurement

A Konica Minolta CM-2300d Chroma Meter (Tokyo, Japan) was used to record the color parameter according to the CIE $L^*a^*b^*$ system (Forsthuber and Grüll 2018; Mononen *et al.* 2002) using a standard illuminant D65 and 10° standard observer. Wood flour before and after treatment were laid on a flat surface evenly when testing. Five measures were read per sample. The CIE $L^*a^*b^*$ color system is a three-dimensional color space that defines the lightness (L^*) of the sample, as well as the color coordinates (a^* and b^*). The lightness L^* (ranging from 0 for black to 100 for white), and color coordinates a^* (ranging from negative values for green to positive values for red on the green–red axis) and b^* (varying from negative values for blue to positive values for yellow on the blue– yellow axis) were measured. Color differences of the samples before and after the treatment were calculated based on the following equations,

$$\Delta L^* = L^{*}_1 - L^{*}_0 \tag{1}$$

$$\Delta a^* = a^*{}_1 - a^*{}_0 \tag{2}$$

$$\Delta b^* = b^*{}_1 - b^*{}_0 \tag{3}$$

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

where L_{1}^{*} , a_{1}^{*} , b_{1}^{*} represents each of the samples after treatment; L_{0}^{*} , a_{0}^{*} , and b_{0}^{*} represents each of the samples before treatment. Color measurements were performed on three points for each sample and an average value was calculated.

Chromophore / Auxochrome Structures Analysis

Diffuse reflectance ultraviolet-visible spectroscopy (DRUV-Vis) before and after treatment were recorded over the wavelength range of 200 nm to 700 nm at room temperature on a UV-vis spectrophotometer (Shimadzu UV-2550, Kyoto, Japan) equipped with an integrated sphere. As a white (R_{∞}) optical standard, the reflectance spectra were recorded against BaSO₄. Wood flour were made into thin slices (30 mm diameter) by EVA (ethylene-vinyl acetate copolymer) hot glue so that the loose powder could be detected in the spectrophotometer. According to the Kubelka-Munk theory, the scanning results of the reflection spectrum were converted into K/S spectrum as follows,

$$K/S = \frac{(1-R)^2}{2R}$$
 (5)

where R is the reflectivity (on a scale from 0 to 1); K is the absorption coefficient; and S is the scattering coefficient.

ATR-FTIR (the attenuated total reflection/Fourier transform infrared) spectra were recorded in absorbance mode using a Perkin Elmer Spectrum Gx instrument (Perkin Elmer, Shanghai, China). The spectra were acquired by accumulating 64 scans at a resolution of 4 cm⁻¹ in the absorbance mode from 4000 cm⁻¹ to 400 cm⁻¹. The relative absorbance intensity (H') of the carbonyl groups was calculated based on the Eq. 6,

 $H_t = H/H_{(1506)}$ (6)

where $H_{(1506)}$ represents the intensity of the aromatic skeleton at 1506 cm⁻¹; H and H_t represent the intensity and relative intensity, respectively, of the carbonyl groups at 1506 cm⁻¹.

An XPS (X-ray photoelectron spectroscopy) analysis was performed with a ThermoVG Scientific Sigma Probe (Thermo Fisher ESCALAB 250Xi, Waltham, MA, USA) using a microfocusing monochromatic AlKa X-ray source at an operating pressure between 10⁻⁹ mbar and 10^{-8} mbar.

RESULTS AND DISCUSSION

Modification Effects on the Color Parameters of Wood Samples

The color parameters (ΔL^* , Δa^* , Δb^* and calculated ΔE^*) of the samples treated with different chemical solvents (0.1 M) are presented in Fig. 2. It can be seen that the lightness (L^*) of all the wood flour after treatment decreased, while both a^* and b^* increased significantly, which indicated that the wood flour became darkened and reddish after the treatment. The total color changes (ΔE^*) of the samples after treatment with ASO was much higher than that of the sample treated without ASO. The chemical adsorption of wood flour using AMA with ASO as a bridge showed the largest total color change (ΔE^*), and also the a^* value increased significantly. This finding demonstrated that the synergistic effect of ASO and AMA contributed to the increase in the red chromatic index. ASO played an important role in the color changes of the wood flour during the chemical adsorption process. This may be due to the coordination of metal ions with lignin that forms a stable metal ion-lignin complex, resulting in an increase in the absorption coefficient in the visible region (Schmalzl *et al.* 1995; Guillon *et al.* 2001; Peart *et al.* 2001).

To investigate the effect of the concentration of ASO and AMA on color parameters of the wood flour, 0.5 g wood flour was impregnated with different concentrations (0.1 to 0.5 M) of ASO under vacuum at 0.8 MPa for 12 h followed by the same treatment conditions of ligand solution (AMA). The color parameters (ΔL^* , Δa^* , Δb^* and calculated ΔE^*) of the wood flour treated with ASO followed by the AMA with different concentrations are presented in Fig. 3.



Fig. 2. The changes in color parameters after treatment with different agents at 90 $^\circ C$ for $12 \ h$



Fig. 3. The color parameters of the wood flour treated with different concentration (0.1 to 0.5M) of ASO/AMA at 90 $^\circ$ C for 12 h

The ΔL^* decreased gradually with the increase in the concentration of ASO/AMA. The Δa^* increased slightly with the increase in the concentration of the solvents. The Δb^* decreased with the increase in the concentration from 0.1 M to 0.2 M. However, little change was observed for Δb^* when the concentration of the solutions was higher than 0.2 M. The total color difference (ΔE^*) was most significant when the concentration increased to 0.5 M. The maximum a^* value was 16.98 (±1.16), which was obtained when treated with an ASO/AMA concentration of 0.5 M. The a^* was close to that of some rosewood, such as *Dalbergia bariensis* (17.14), *Dalbergia frutescens* var. *tomentosa* (15.54), and *Dalbergia oliveri* (17.61). This suggested that the concentration of the solvents played an important role on the a^* values.

DRUV-vis Spectral Analysis

The light absorption coefficient spectra was used to identify the changes in the chromophoric and auxochrome structures, which can help to understand the reasons for the color changes better (Johansson 2000). The DRUV–vis spectra of wood flour after treatment with 0.5 M of ASO, AMA, and ASO/AMA respectively are shown in Fig. 4.



Fig. 4. The DRUV-Vis spectrum of wood flour before and after adsorption treatment with ASO, AMA, ASO/AMA, respectively

After the adsorption treatment with ASO and ASO+AMA, the K/S value in the spectral region between 200 nm to 700 nm increased significantly, which indicated an increase in the conjugated double-bond structures, such as quinoid structures (370 nm to 550 nm) (Wei *et al.* 2017). This led to an increase in a* parameter. The shoulder peak at 391 nm moved to 416 nm with the ASO or ASO+AMA treatment. The increased absorption intensity resulted in a yellowish color in the samples. The absorption intensity of the peak at 400 nm - 600 nm increased significantly after the wood flour was treated with ASO + AMA. This finding was consistent with the increase in the chromaticity values of a^* and

 b^* , which conforms to the principle of color reduction (Farnsworth 1943) in that the color of the samples become dark brown after treatment. Compared with the PMK, the sample treated with ASO + AMA showed stronger absorption at 350 nm to 499 nm. The *K/S* ratio of the sample treated with ASO + AMA was slightly lower than that of the PMK at 499 nm - 600 nm, which was the absorption area of the yellow light. It indicated that the color of the wood flour treated with ASO + AMA was similar to that of PMK.

ATR-FTIR Spectra Analysis

The ATR-FTIR spectra of wood flour treated with different solutions of 0.5 M concentration was normalized at 1506 cm⁻¹ (Fig. 5). When the metal ions of Al(III) were added to the wood flour, the peak originally at 2917 cm⁻¹ shifted slightly to 2915 cm⁻¹, indicating that an interaction occurred between the wood components and Al(III) (Zhang *et al.* 2016). The characteristic absorption peak of the C-O single bond stretching vibration at 1031 cm⁻¹ weakened due to coordination with Al(III) to transfer the benzene ring π electrons to the ring after treatment with ASO (Ó'Coinceanainn *et al.* 2003). Thus, many C-O-R-O-C-group structures formed through coordination reactions between the wood components (cellulose, hemicellulose, and lignin) and Al(III) (Perkins 1996).



Fig. 5. The ATR-FTIR spectra of wood flour before and after treatment with ASO, AMA, ASO/AMA, respectively

The bands at 1737 cm⁻¹, 1236 cm⁻¹, and 894 cm⁻¹ are the characteristic absorption peaks of acid compounds, which are attributed to C=O and C-O stretching, and C-H deforming, respectively (Halpern *et al.* 2014; Franklin and Guhanathan 2015). The intensity of the peaks at 1737 cm⁻¹ and 894 cm⁻¹ increased after treatment with ASO, ASO+AMA, or AMA. It indicated that there has been an increase in C=O groups during adsorption treatment. The control group showed that the conjugated C=O groups at 1656 cm⁻¹ and the aromatic skeletal of lignin at 1594 cm⁻¹ decreased after the adsorption

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treatment. It can be inferred that Al(III) is bonded with C=O group in the wood flour and the ligand and metal are mainly absorbed into the lignin unit in the wood (Zhang *et al.* 2018).

XPS Spectral Analysis

To investigate the chemical structure of the coordination compounds upon the adsorption process, an XPS analysis was conducted, and the spectra are shown in Fig. 6.



Fig. 6. The XPS of C1S and O1S of samples before and after adsorption treatment. (a) Curve fitting of the C1s of the control; (b) Curve fitting of the C1s of the samples treated with ASO (0.5M); (c) Curve fitting of the C1s of the samples treated with ASO+AMA (0.5M); (d) Curve fitting of the O1s of the control; (e) Curve fitting of the O1s of the samples treated with ASO (0.5M); (f) Curve fitting of the O1s of the samples treated with ASO+AMA (0.5M); (d) Curve fitting of the O1s of the Samples treated with ASO+AMA (0.5M); (f) Curve fitting of the O1s of the Samples treated with ASO+AMA (0.5M); (f) Curve fitting of the O1s of the Samples treated with ASO+AMA (0.5M); (f) Curve fitting of the O1s of the Samples treated with ASO+AMA (0.5M); (f) Curve fitting of the O1s of the Samples treated with ASO+AMA (0.5M); (f) Curve fitting of the O1s of the Samples treated with ASO+AMA (0.5M)

The carbon atoms of the wood component can be divided into four combinations according to the ratio of oxygen and hydrogen atoms (Dorris and Gray 1978). The C1, with an electron binding energy of 284.7 eV, is the C combined with the other carbon atoms or hydrogen atoms, which are mainly from the lignin phenylpropane and fatty acids, fats, and waxes, etc. The C2 is the C combined with a non-carbonyl oxygen. The electron binding energy of C2 is 286.3 eV. The C3 is the C combined with two non-carbonyl oxygens or with one carbonyl oxygen, which is mainly a ketone group and an aldehyde group in a lignin molecule with a binding energy of 287.5 eV. The structural feature reflects the oxidation of the chemical components on the surface of the wood flour. The C4 is the C combined with a carbonyl oxygen or a non-carbonyl oxygen mainly from the carboxylate groups. The electron binding energy of C4 is 288.7 eV. Figure 6 shows the XPS of C1S and O1S of the control and adsorption treated wood flour. The percentage of the electron binding energy of the C1, C2, C3, and C4 and the ratio of C3/C2 are shown in Table 2. The O1 represents the C=O structure in lignin, while O2 represents the C-O in the carbohydrate (Tshabalala et al. 2005). The electronic binding energy of O1 and O2 and the ratio of O1/O2 are shown in Table 3. Tables 2 and 3 show that after the treatment with ASO+AMA, the ratio of C4, C3/C2, and O1/O2 increased. These findings indicated that the carbonyl groups and carboxyl groups increased after the treatment. The increase in the carbonyl and carboxyl groups resulted in a decrease in lightness and changes in the color parameters (Rowell and Rowell 2005). The results of the XPS analysis were consistent with the ATR-FTIR and DRUV-VIS spectral analysis, as well as the changes in the chromaticity values.

Samples	C1 (284.7 eV)	C2 (286.3 eV)	C3 (287.5 eV)	C4 (288.7 eV)	C3/C2
Control	59.55	29.56	7.11	3.77	0.24
ASO	57.05	31.05	7.84	3.62	0.25
ASO+AMA	63.42	21.98	7.75	6.84	0.35

Table 1. Subpeak Area Fractions of C1S and Ratio of C3/C2

Table 2. Subp	beak Area	Fractions	of C1S	and Ratio o	of C3/C2
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Samples	O1(C=O) (531.7 eV)	O2(C-O) (532.8 eV)	01/02
Control	10.52	89.48	0.12
ASO	13.90	86.10	0.16
ASO+AMA	27.68	72.32	0.38

As shown in Fig. 7, the N1s peak was deconvoluted into two components at binding energies of 399.6 eV and 400.2 eV, which was ascribed to the nitrogen-containing groups of $-NH^-$ and $-NH_2$ in the wood flour (Li *et al.* 2019). After the treatment, the N1s spectra of the ASO and ASO+AMA treated wood flour comprised three peaks with binding energies of 399.6 eV, 400.2 eV, and 401.5 eV. The appearance of the peak with a binding energy of 401.5 eV confirmed the formation of a N•••AL complex. The N atom in the amine groups share a lone pair of electrons with the electron-withdrawing Al (III) ion, causing the coordinate interaction between the N atom and Al (III) ion. The content of N in the wood flour after adsorption and treatment with ASO+AMA was higher than that of the control, which also indicated that NH⁴⁺ was successfully absorbed into the wood components.



Fig. 7. The XPS of N1S before and after ASO and ASO/AMA (0.5M) adsorption treatment. (a) Curve fitting of the N1s of the control; (b) Curve fitting of the N1s of the samples treated with ASO; (c) Curve fitting of the N1s of the samples treated with ASO+AMA

	Control	ASO	ASO+AMA
C1S	77.76	76.69	77.47
N1S	2.20	2.54	2.74
O1S	19.83	20.59	18.97
AI2P	0.21	0.18	0.81

Table J. Liemental Composition of Lach Cample Measured by A	Table 3.	Elemental	Composition	of Each Sam	ple Measured	by XPS
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The XPS spectra of Al2p (Fig. 8) shows that the electron binding energy of the main characteristic absorption peak of AL2p in the wood flour after ASO+AMA(0.5M) impregnation treatment was 74.52 eV, which was 0.38 eV lower than the electron binding energy (74.90 eV) in Al₂(SO₄)₃ (Li *et al.* 2019). This indicated that Al(III) participated in the coordination reaction (Ciabatti *et al.* 2010; Roselin and Selvin 2011).



Fig. 8. The XPS of AL2p after ASO/AMA (0.5M) treatment

CONCLUSIONS

- 1. The black locust wood treated with aluminum sulfate octadecahydrate (ASO) showed positive higher *a** value compared to those treated with ammonia (AM), acetic acid (AA), or ammonium acetate (AMA) alone. The treatment using ASO+AMA exhibited the best efficiency on the red color improvement.
- 2. A significant increase in *a** value could be obtained when the black locust wood was treated with 0.5 M of ammonium acetate (AMA) combined with aluminum sulfate octadecahydrate (ASO) for 12 h at 90 °C. The resulting *a** was close to that of *Pterocarpus macrocarpus* Kurz (PMK) heartwood.
- 3. The Al(III) ion coordinated with lignin that formed a stable metal ion-lignin complex, resulting in an increase in the absorption coefficient in the visible region. The ligand with carbonyl group or amino group were absorbed into the lignin unit with Al(III) acting as a bridge. The adjusted of the chromophore /auxochrome structures made the black locust reddish.

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