

Colorability of Dyed Wood Veneer Using Natural Dye Extracted from *Dalbergia cochinchinensis* with Different Organic Solvents

Tao Zhu,^{a,b} Shunzhi Liu,^{a,c} Kai Ren,^a Jiabao Chen,^a Jinguo Lin,^{a,*} and Jian Li^{a,d,*}

The colorability of wood veneers was evaluated with a natural dye that had been extracted using four different organic liquids. The solvent with the optimal dye effect was selected by measuring their total color difference. In addition, the dyeing mechanism, color fastness of water, and thermal stability of the dyed samples with the optimal dye effect were also investigated. The results showed that the absolute ethanol extraction can obtain dyed samples with a maximum total color difference accompanied with a satisfactory color fastness of water. Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM) analyses suggested that only physical absorption occurred between the natural dyestuff and wood tissues, and the distribution form of natural dyestuffs was akin to the membrane covering the wood vessel wall, wood fiber lumens, ray cells, etc. Moreover, thermogravimetric (TG) results revealed that the residue of the dyed samples was more than that of the untreated samples, which indicated that the thermal stability of wood veneers can be improved after the dyeing process.

Keywords: Colorability; Natural dyes; Organic solvents; Wood veneer

Contact information: a: College of Material Engineering, Fujian Agriculture and Forestry University, Fuzhou, Fujian 350002, China; b: Furniture Products Quality Supervision and Testing Center of Putian, Xianyou, Fujian 351256, China; c: Xiamen Botanical Garden, Xiamen, Fujian 361003, China; d: College of Material Science and Engineering, Northeast Forestry University, Harbin, Heilongjiang 150040, China; * Corresponding authors: fjlinjg@126.com; nefulijian@163.com

INTRODUCTION

High-quality wood resources (HQWR) are favored by most customers for their superior surface characteristics. Recently, the amount of HQWR has decreased, while the demand for it has increased. Thus, current efforts have focused on fast-growing wood utilization. Nevertheless, many inherent drawbacks exist including low density, poor decay resistance (Neil and William 1972), and the inferior surface color appearance (Wang *et al.* 2016). Wood color is a crucial measurement index to determine the appearance quality and market value. Dyeing this type of wood can dramatically improve its surface color, resulting in a more appealing pattern (Liu *et al.* 2015; Canevari *et al.* 2016; Hu *et al.* 2016; Wang *et al.* 2016). In recent years, various colorful dyed woods have been used for industrial production as a raw material for furniture, flooring, indoor decoration, etc. (Arai and Nakajima 2011; Zhao *et al.* 2014; Kielmann and Mai 2016), which have greatly improved the utilization value of fast-growing wood. Currently, common dyestuffs employed in wood dyeing include direct, acid, basic, and reactive dyestuffs, which are all synthetic chemicals (Peng *et al.* 2005). The overuse of synthetic dyes poses a potential threat to human health, and the dye liquor treatment is a large environmental pollution problem (Bangash and Manaf 2005; Tsang *et al.* 2007). As a

consequence, there has been an increasing interest in searching for natural dyes as alternatives due to their eco-friendly nature (Özen *et al.* 2014).

Natural dyes extracted from plants have been more extensively applied in fiber textile dyeing (Park *et al.* 2008; Haji 2010; Gulzar *et al.* 2015; Boutaty *et al.* 2018). For better extraction, various modern techniques have been employed for extraction of natural dyes, including ultrasonic extraction (Adeel *et al.* 2018a), microwave-assisted extraction (Adeel *et al.* 2018b;), plasma treatment (Barani *et al.* 2015), and gamma ray-assisted extraction (Adeel *et al.* 2017). These approaches have improved the color strength remarkably along with obtaining excellent color fastness. In addition, Korean researchers have verified that a nontoxic natural dye could be fabricated from an extract from *Caesalpinia sappan* (sappan wood) and applied to colored paper (Lee *et al.* 2008). However, there are very few studies on the application of natural dye in the wood dyeing process (Jin *et al.* 2015).

Wood color is associated with the extractives content and type (Klumpers *et al.* 1993; Aguilar-Tovar *et al.* 2009; Moya *et al.* 2012), and thus researchers have attempted to employ extractives as the natural dyestuff to dye wood. For example, Özen *et al.* (2014) investigated natural colorants extracted from pomegranate skin and black mulberry with distilled water using the ultrasonic method at different rates on wood dyeing. However, wood extracts contain many organic compounds, *i.e.*, tannins, pigments, resin, and alkaloids, which are mostly found in heartwood (Xu 2006). They are prone to dissolving in organic solvents (OS) compared to water, which may lead to inferior dyeing effects. Dyeing wood with extracts in distilled water may present poor color fastness to water. Thereby, extract from heartwood with organic solvents employed as dyestuff to dye wood veneer may be an alternative to obtain a better dyeing effect. Additionally, the dye liquor treatment will be performed simply by the separation of organic solvents individually from the dye liquor after dyeing with rotary evaporators with a goal of recycling liquid dye and creating no pollution in the environment.

Dalbergia cochinchinensis belongs to the genera of *Dalbergia*, a family of Fabaceae, which are mainly widely cultivated in Iran, Vietnam, and Southeast Asia region. Due to its remarkable properties such as better dimension stability, high-density, excellent color appearance, *etc.*, it has been listed in the national standard of Hongmu in China, namely GB/T 18107 (2000). In a small town named Xianyou in Fujian province, many *D. cochinchinensis* logs are imported from abroad and employed as the desired raw material in making upscale furniture and artware with high-value (Kielmann and Mai 2016). Nevertheless, plenty of processing remains (PR) are produced annually. Most of the PR is merely used as fuel, resulting in a large amount of waste of rare resources. Because of its abundant extractives contents, the organic extracts from the PR of *D. cochinchinensis* employed as natural dyes to dye wood veneers may provide a new approach to obtain a satisfied dye effect along with increasing the utilization ratio of the PR.

The objective of this work was to develop a novel dyeing method and measure the dye effect with natural dyes extracted from PR of *D. cochinchinensis* in four different organic solvents with different polarities. The optimum organic solvent to extract natural dyes with the best dye effect was selected by measuring the surface total color difference. Moreover, the dyeing mechanism and the color fastness to water of the optimum was explored.

EXPERIMENTAL

Materials

Seven-year-old fast-growing poplar wood was cut in Fujian, in the southeast of China. The sapwood was rotary cut and air dried to make its moisture content less than 8%. The wood veneer with smooth fiber, crack-free, knotless, and color uniformity was selected and prepared with dimensions of 25 x 1.2 x 60 mm (Tangential*Radial*Longitudinal).

Natural Dyes Preparation

The PR of *D. cochinchinensis* residue was cleaned with distilled water, and it was also air-dried to less than 8% moisture content. Afterwards, it was ground with a grinder. The mesh number of wood powder between 40 to 60 was chosen as the raw material to be extracted using a Soxhlet apparatus with four different organic solvents, *i.e.*, absolute ethanol (AE), acetone (AT), dichloromethane (DM), and ethyl acetate (EA). All concentrations of the extraction solution were at the same ratio measured by the wood powder weight in an organic solvent (5 g extracted in 250 ml volume organic solvent), and the extraction temperature was the boiling point of each organic solvent (*i.e.* AE: 79 °C; AT: 57 °C; DM: 40 °C; EA: 77 °C). In addition, the solution was extracted completely when the liquid in the extraction tube was almost colorless. Moreover, the volume loss during the extraction processing was compensated by adding a homologous organic solvent at the end of the extraction period. Finally, the dyeing solution was stored in a light-resistant container in the refrigerator until use.

Wood Veneer Dyeing

A customized dyeing apparatus (CDP) (Fig. 1) was needed due to the volatility of the organic solvent, which was produced according to the experimental requirements by Synthware Co. Ltd., Beijing, China.

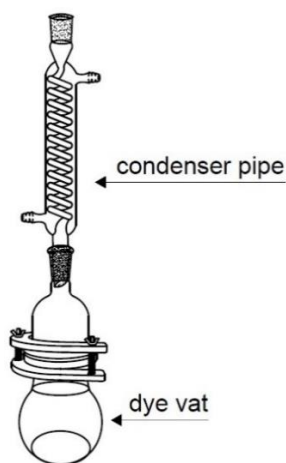


Fig. 1. Diagram of customized dyeing apparatus

The dyeing process was conducted with CDP using a water bath heating under ambient pressure. The dyeing temperature was the same as the extraction temperature, and dyeing time and bath ratio were 6 h and 1:50, respectively. Six wood veneers were vertically inserted into the groove of the plastic dyeing apparatus and appropriate space

was left to ensure evenness during the whole dyeing process. The dye liquor was stirred clockwise with a glass rod every 30 min for even penetration. No dyeing assistant, penetrant, or color fixing agent was added in the dyeing liquor to ensure natural dyed wood veneer was obtained. The chromatic value of wood veneer before and after the dyeing process was tested. The volatile organic solvents in dyed wood veneers can easily release into the atmosphere.

Color Values and K/S Measurement

Surface color change values of wood veneers before and after dyeing were determined with color measurement (Datacolor I7, X-rite, Granville, OH, USA) via the CIE $L^*a^*b^*$ color system employing a D65 standard illuminant and 10° normative standard, which was issued by the International Commission on Illumination. The color values L^* (lightness to darkness), a^* (redness to greenness), and b^* (yellowness to blueness) were measured in three portions along the diagonal of each veneer (two parallel for each organic solvent dye). The average value of each chromaticity parameter was recorded, and the total color difference (ΔE), which denotes the surface color change, was calculated from Eqs. 1 through 4,

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (1)$$

$$\Delta L^* = L_a^* - L_b^* \quad (2)$$

$$\Delta a^* = a_a^* - a_b^* \quad (3)$$

$$\Delta b^* = b_a^* - b_b^* \quad (4)$$

where the ΔL^* , Δa^* , and Δb^* represents the differences values of L^* , a^* , and b^* of the wood veneer before (L_b^* , a_b^* , and b_b^*) and after (L_a^* , a_a^* , and b_a^*) dyeing, respectively.

Based on the Kubelka-Munk theory, the K/S (color strength) value of wood veneer before and after dye can be calculated using Eq. 5, which is determined by the reflectance observed also on the color measurement (Datacolor I7, X-rite). The wavelength range conducted on this work was 360 to 750 nm.

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

where K , S , and R are the absorption coefficient, scattering coefficient, and reflectance, respectively.

Chemical Composition of the Extraction Analyses

The chemical composition of the extraction was analyzed using LECO GC-MS (HRT 4D+, St. Joseph, USA), following the temperature program setting as in the Table 1. Rxi-5MS with specification of 30m×250μm×0.25μm (length×interior diameter×film thickness) was selected as separating chromatographic column. The inlet split ratio, injection temperature, split ratio, and carrier gas were set to 20:1, 280 °C and helium with flow rate of 1.4 mL min⁻¹ respectively. Mass spectrometry conditions comprised mass scan range (35 to 550 aum), ionizer voltage of the MS detector (70 eV), and ion source (LECO EI) temperature (250 °C). LECO's chemical station software and databases (Weily 10.0 and NIST2017) were employed to collect spectra and identify the chemical composition in the extractives.

Table 1. Temperature Program Setting

Temperature (°C)	Rate (°C min ⁻¹)	Duration (min)	Total time(min)
50 (initial)	---	1	1
280 (target)	10	10	34

FTIR, SEM, and Thermostability Analyses

The surface chemical structure of wood veneer before and after dye was characterized by using Fourier transform infrared spectroscopy (FTIR). In addition, the surface chemical structure of extractives of the organic solvent with the best dye effect was also tested. All samples were mixed with KBr at a ratio of 1:100 and grinded into ultrafine powder in the mortar. Then they were placed on the diamond crystal of the FTIR spectrometer (VERTEX 70, Karlsruhe, Germany). Spectra were collected in transmittance mode with 32 scans in the range of 400 to 4000 cm⁻¹ at a resolution of 1 cm⁻¹.

The micromorphology of both the wood veneers before and after dyeing was observed by using a scanning electron microscope (Nova Nano SEM, Portland, OR, USA). The thermostability of the sample powder (40 to 60 mesh) was analyzed with a heat integrated analyser (ATA449F3, Munich, Germany). The initial temperature was set as 30 °C and then raised to 500 °C at a heating rate of 10 °C min⁻¹ under a 50 mL min⁻¹ nitrogen atmosphere flow.

Color Fastness of Water

In order to test the color fastness of water of the samples dyed with the optimum organic solvent extraction, the dyed samples were immersed completely in the hot water (80 °C) for 2 h. Afterwards, the color parameters including L^* , a^* , and b^* of the treated samples were recorded with the colorimeter (Datacolor I7, X-rite). Accordingly, the total color difference (ΔE) obtained with Eq. 1 was employed to measure the color fastness in response to treatment with water.

RESULTS AND DISCUSSION

Dye Effect of Different Organic Solvents Extraction on ΔE and K/S.

The visual assessment was vital to evaluate the dyed wood quality and decorative properties from a common consumers' perspective. All the surface color of wood veneers after dyeing with four different organic solvents extraction underwent apparent changes compared to the untreated samples (Fig. 2). The color parameters of both the treated and untreated samples are shown in Table 2.

The negative ΔL^* values of all the dyed samples indicated the color turning gray, while the values of both Δa^* and Δb^* were positive, suggesting the color turning towards redness and yellowness, respectively. The greater ΔE value invariably exhibits a preferable dye effect when the concentration of dye solution is low in the prior research (Liu *et al.* 2015; Hu *et al.* 2016), so the ΔE value was employed to evaluate the dye effect of four organic solvents used in this work.

As shown in Table 2, AE obtained a maximal ΔE (32.21 ± 2.53), followed by AT (ΔE : 26.10 ± 2.80), then EA (ΔE : 17.32 ± 1.23), and the minimum was DM (ΔE : 16.54 ± 2.02). This finding is coincident with the order of the organic solvents polarity, namely the higher polar solvent will get a better dye effect. This is probably attributable to a greater proportion of the extractives in *D. cochinchinensis* being able to dissolve out in AE

compared with the other three solvents at the same ratio. Hence, AE was selected as the optimal organic solvent to extract natural dyestuff from *D. cochinchinensis* to dye wood veneers in this work.

In addition, a distinct visual assessment of the dyed samples can be observed only when the ΔE variation was over three (Duan 2002). However, all the deviation of ΔE (Table 2) were less than the standard value, which illustrated that all the treated samples obtained a good dyeing uniformity property.



Fig. 2. Visual assessment of wood veneers before and after dye with different organic solvent extraction

Table 2. Color Parameters of Wood Veneers Before and After Dyeing with Different Organic Solvent Extraction

OS	Before dye			After dye			Color differences values			
	L_b^*	a_b^*	b_b^*	L_a^*	a_a^*	b_a^*	ΔL^*	Δa^*	Δb^*	ΔE
AE	89.00 (0.68) ^a	1.11 (0.39)	16.51 (0.78)	62.50 (1.88)	17.77 (0.93)	24.04 (0.87)	-26.49 (2.11)	16.66 (1.12)	7.53 (1.73)	32.21 (2.53)
AT	86.04 (0.85)	2.28 (0.40)	16.23 (0.62)	64.03 (2.46)	15.50 (1.10)	20.73 (1.33)	-22.02 (2.44)	13.22 (1.29)	4.50 (1.25)	26.10 (2.80)
EA	88.34 (0.45)	1.50 (0.25)	15.93 (0.59)	74.51 (0.77)	9.90 (0.72)	21.75 (1.21)	-13.83 (0.88)	8.40 (0.80)	5.82 (1.17)	17.32 (1.23)
DM	87.98 (0.73)	1.79 (0.27)	15.86 (0.66)	78.14 (1.66)	5.71 (0.88)	28.41 (1.44)	-9.84 (1.45)	3.92 (0.86)	12.56 (1.41)	16.54 (2.02)
Numbers (a) in the parentheses stand for the standard deviation of 36 replicates.										

Moreover, linear-regression analysis was conducted aiming at exploring the contribution of each color parameters (ΔL^* , Δa^* , and Δb^*) on ΔE of dyed samples. As shown in Fig. 3, there was a good linear relation ($R^2 \geq 0.90$) in both the ΔL^* and Δa^* with ΔE of AE and AT with the exception of DM and EA.

For the significance of the effect of AE and AT, the change of ΔE was significantly influenced by ΔL^* , followed by Δa^* , and the influence of Δb^* represented was relatively minor. Consequently, the natural dyestuffs extracted from *D. cochinchinensis* with AE played a prominent effect on the variation of L^* and a^* values, resulting in the total surface color change of wood veneers.

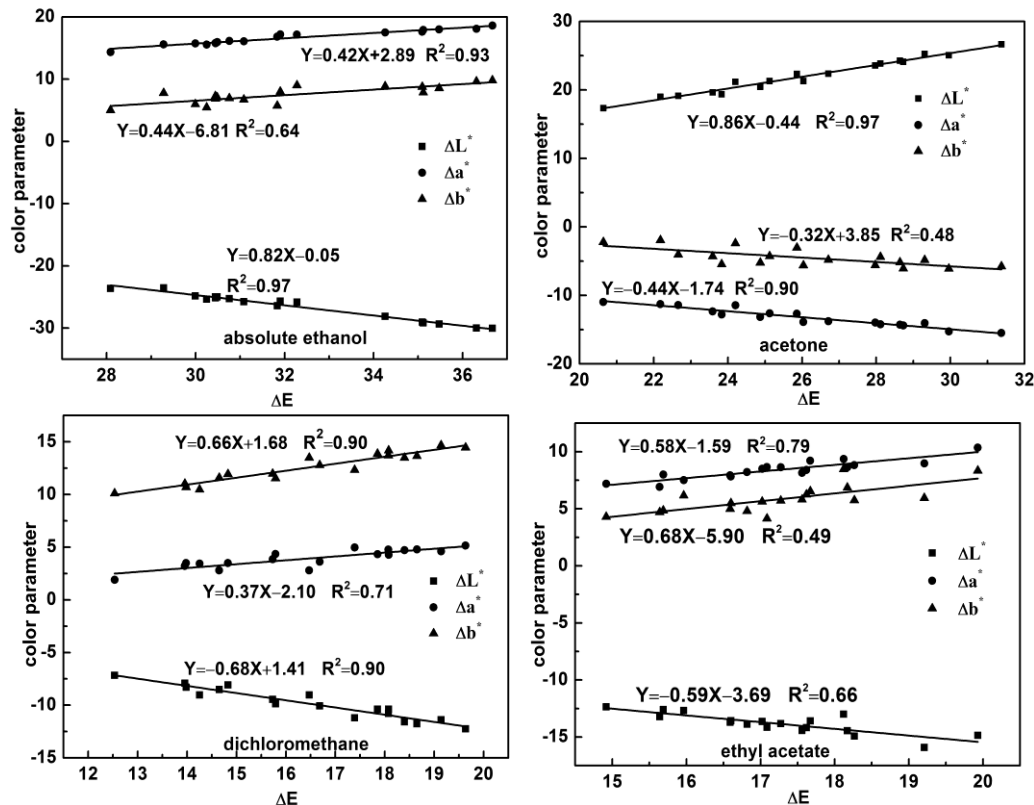


Fig. 3. Linear regression analysis of ΔL^* , Δa^* , and Δb^* of dyed samples

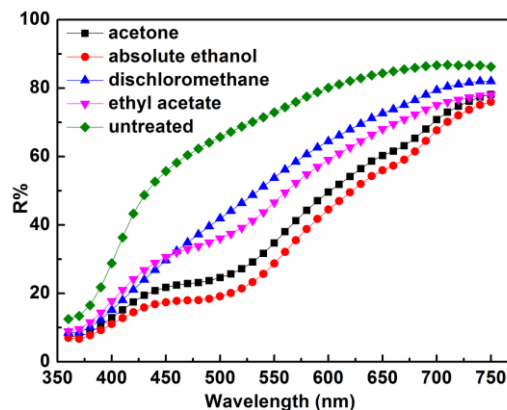


Fig. 4. Spectral reflectance curves of samples before and after dyeing

The lightness of the wood surface was determined by the spectral reflectance, and the lightness is positively correlated to the R value (Cheng 2010). The reflectance spectrum of samples before and after dyeing in the visible region is shown in Fig. 4. The R value of the untreated sample overall was higher at each wavelength compared with all the dyed samples, and that of the wood veneers dyed with AE extraction was the smallest, which agreed with the obvious decline of L^* . The maximum ΔL^* value was -26.49 ± 2.11 (AE), suggesting that the surface color of that appeared to have greatest change of all the dyed samples in terms of darkness.

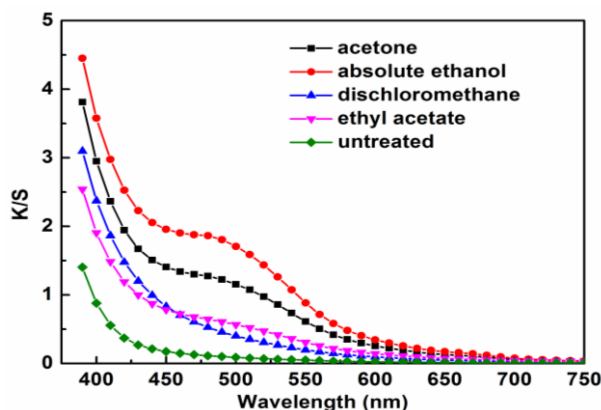


Fig. 5. K/S absorption over scattering spectra of samples before and after dyeing

The K/S ratio of absorption over scattering can exhibit the level of surface color change attributed to by the dye effect with different solvents. It can be seen that the K/S value of samples dyed with AE extraction was maximal in the whole visual region compared to the untreated samples (Fig. 5), which revealed the AE extraction can obtain the best dyeing effect. This finding is coincident with the ΔE variation result in Table 2.

Main Chemical Composition of the AE Extraction Analysis

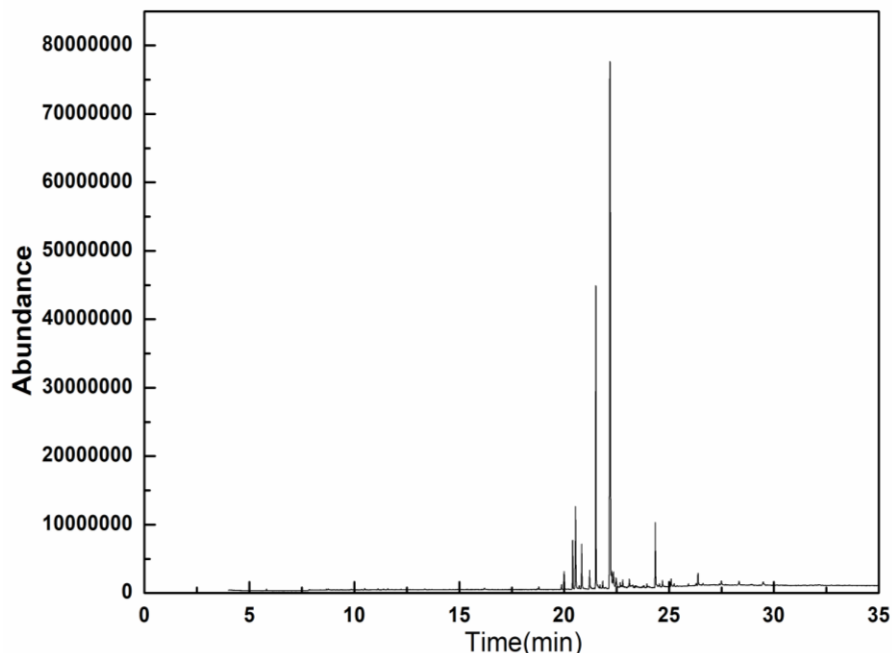
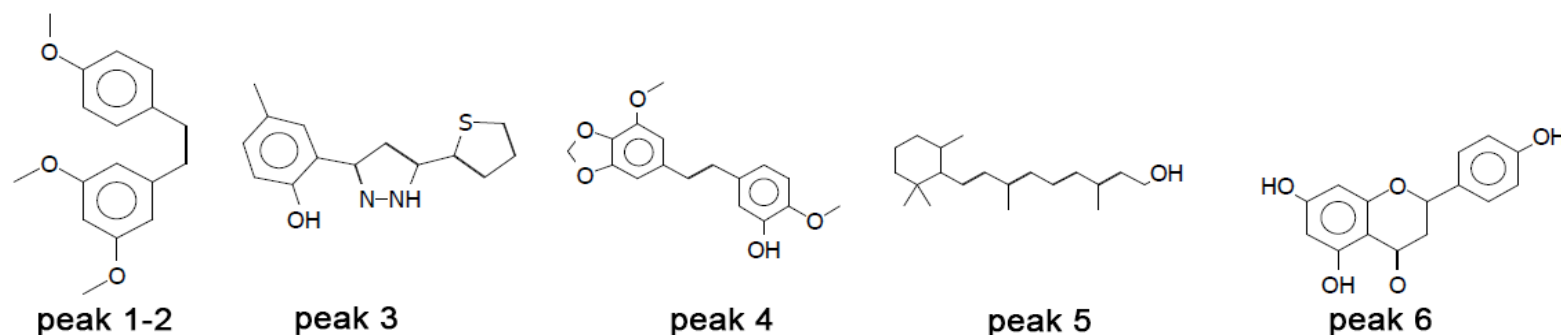


Fig. 6. Total ion chromatogram of AE extraction

Total ion chromatogram spectra of AE extraction were presented at Fig. 6. It can be clearly seen that main peaks were found at retention time between 20 min and 25 min. For further clarification, relative peak areas were used to exhibit the relative content of main compositions of AE extraction, which can be obtained by calculating ratio of each peak area to sum of all the peaks area.

Table 3. Result of GC-MS Chemical Composition of AE Extraction

Peak	Name	Retention time (min)	Relative area(%)	Similarities(%)	Chemical formula
1	Cis-trismethoxyresveratrol	20.41	1.36	70	C ₁₇ H ₁₈ O ₃
2	Cis-trismethoxyresveratrol	20.55	1.47	68	C ₁₇ H ₁₈ O ₃
3	Phenol,4-methyl-2-[5-(2-thienyl)pyrazol-3-yl]-	20.84	1.01	70	C ₁₄ H ₁₂ N ₂ OS
4	2-Methoxy-5-[2-(7-methoxy-2H-1,3-benzodioxol-5-yl)ethenyl]phenol	21.51	15.65	66	C ₁₇ H ₁₆ O ₅
5	Retinol	22.19	68.00	60	C ₂₀ H ₃₀ O
6	Naringenin	24.34	3.64	71	C ₁₅ H ₁₂ O ₅

**Fig. 7.** Structural formula of main chemical composition of AE extraction

Although the temperature procedure had been optimized before the test started, there were still several peaks merged together due to little difference in retention time of the adjacent peaks. Hence, the adjacent peaks can be treated as two individual peaks only when the difference retention time between them is more than 0.1 min (Xu *et al.* 2014). Meanwhile the peak with relative area more than 1% can be considered as a single and main characteristic peak vector (Xu *et al.* 2013). Its chemical compositions were identified by retrieval of NIST2017 mass spectral and wiley (version 10.0) libraries.

As shown in Table 3, there were six main chemical components, *cis*-trismethoxyresveratrol, phenol,4-methyl-2-[5-(2-thienyl)pyrazol-3-yl]-, 2-methoxy-5-[2-(7-methoxy-2H-1,3-benzodioxol-5-yl)ethenyl]phenol, retinol, and naringenin. The results presented in Fig. 7 indicated that both chromophoric (unsaturated double bonds) and auxochrome groups (hydroxyl, carboxyl, and ether group, *etc.*) (Li 2013; Cheng 2010) were present in the main composition of the AE extract, which probably led to the surface color change of wood veneers after dyeing. However, further study is still needed regarding relationship between the chemical groups and the specific chromaticity index (L , a^* and b^*) variation.

FTIR Analysis of the Untreated and Samples Dyed with AE Extraction

The FTIR spectra of the samples before and after dyeing with AE extraction are shown in Fig. 8. The main absorption peaks were marked and corresponded to the functional group of hydroxyl (3402 cm^{-1}) and C-H stretching vibration (2920 cm^{-1}). Also, the cellulose, hemicellulose, and lignin represent absorption peaks at 874 cm^{-1} , 1739 cm^{-1} , and 1512 cm^{-1} , respectively (Pandey 1999; Liu *et al.* 2015). It can be seen that no obvious differences existed between these because no absorption peaks were generated during the entire dyeing process, which displayed that only physical adsorption occurred between the natural dyestuff and wood tissues.

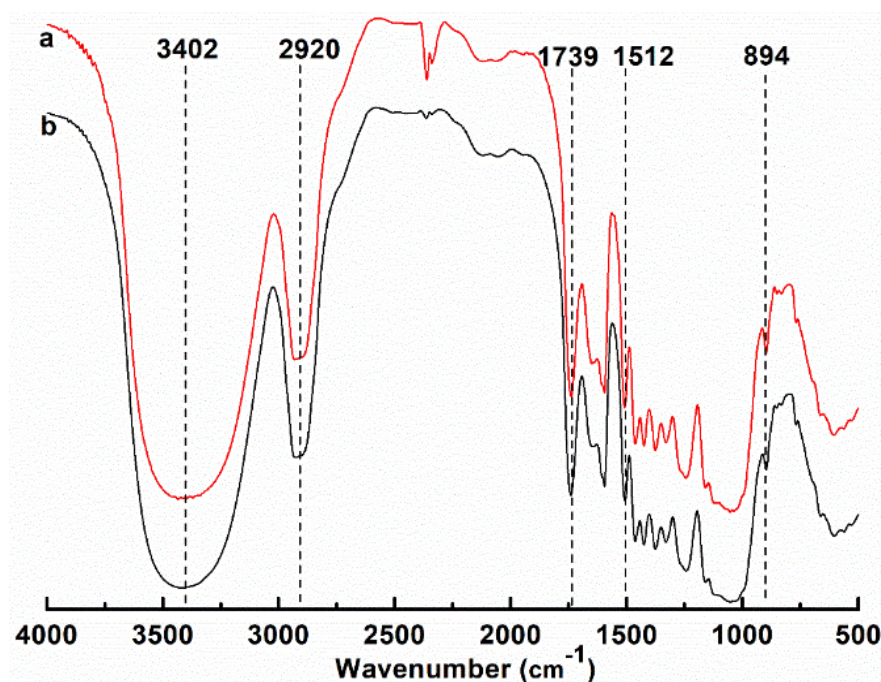


Fig. 8. FTIR spectra of undyed (b) and dyed (a) wood veneers with AE extraction

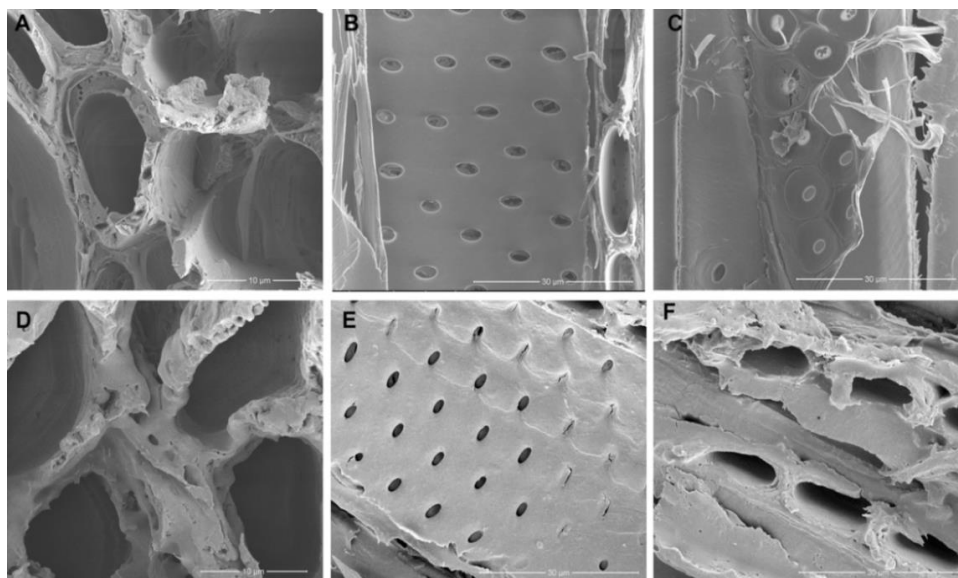


Fig. 9. SEM micrographs of wood veneers before (A, B, and C) and after (D, E, and F) dyeing with AE extraction

SEM Analysis

A comparison analysis of SEM micrographs between the untreated and dyed samples was conducted to observe the form of natural dyestuff distribution in the wood tissue. The surface of wood fiber (Fig. 9C), vessel wall (Fig. 9B) of the untreated sample was smooth, and that pits on the vessel wall (Fig. 9B) and wood fiber (Fig. 9C) can be seen clearly. In the tangential section of the dyed sample, both the wood ray (Fig. 9F) and the vessel wall (Fig. 9E) were covered by the dyestuff. Moreover, some pits were even completely covered (Fig. 9E). In addition, as shown in Fig. 9D, the wood fibers were also covered by the dyes accompanied with numerous dyes aggregated on the wood fibers in the transverse section compared with the untreated samples (Fig. 9A). Consequently, the results demonstrated that the dyes extracted from *D. cochinchinensis* with AE may have generated a membrane covering the wood tissues. This finding is completely different from the previous studies on the distribution form of the synthetic dyestuff in the wood tissues (Hu *et al.* 2016; Wang *et al.* 2016, 2018).

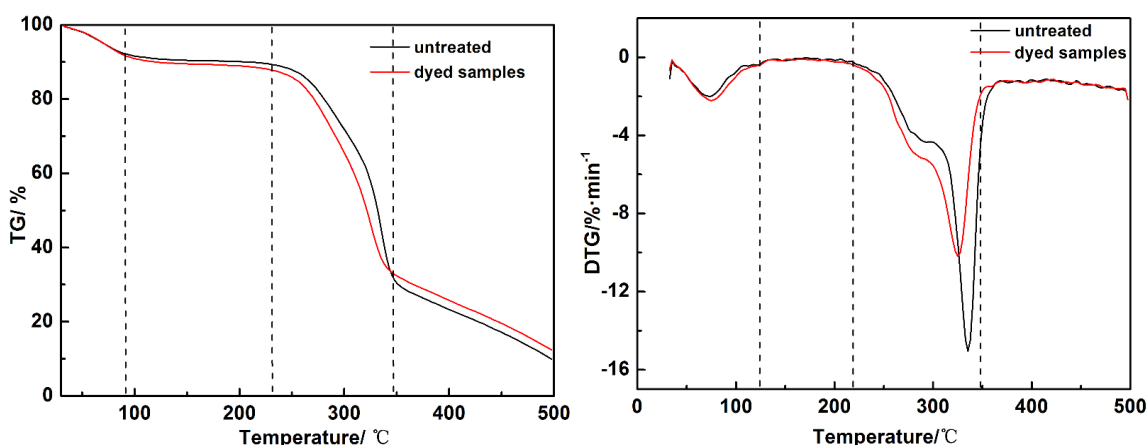
TG Analysis

TG and DTG curves of untreated and samples dyed with AE extraction were combined to analyze the thermal stability variation. As revealed in Fig. 10, the whole pyrolysis process of wood can be divided into four stages, and most of the degradation occurred in the third stage. At this stage shown in the Table 4, the temperature of the untreated samples at the maximum degradation rate was 335.3 °C, which was 10.3 °C higher than that of the dyed samples, suggesting the temperature at the maximum degradation occurred beforehand. Nevertheless, the residue of the dyed samples at the fourth stage increased from 10.4% to 12.4% compared with the untreated samples, which indicated that the thermal stability of samples after the dyeing process performed better. These findings suggest that some extractives in the wood veneers with poor thermal stability may be more prone to dissolving out in the AE solution during the dyeing process, resulting in an increased proportion of chemical components with a satisfactory thermal stability.

Table 4. Thermogravimetric Values of Thermal Degradation between Untreated and Dyed Samples

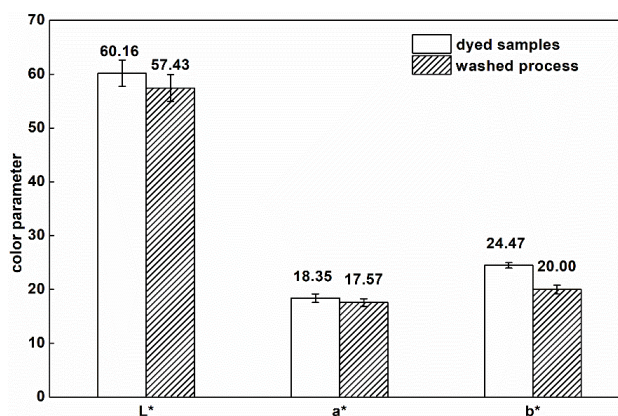
Sample	T_{\max}^b (°C)	Residue c (%)
Untreated samples	335.33	10.35
Dyed samples	324.99	12.43

b : Temperature at maximum degradation rate
 c : Values determined at 500 °C with heating rate of 10 °C min⁻¹

**Fig. 10.** TG and differential TG curves of untreated and dyed samples

Color Fastness of Water Analysis

Figure 11 shows that all color parameters of the dyed samples decreased after being soaked in the hot water, which had a relatively noticeable impact on the b^* value followed by L^* , along with a minor impact on a^* , suggesting that, to some extent, the natural dyestuff related to b^* and L^* can be removed by heating them in the hot water but with the exception of a^* . Additionally, the ΔE value (5.48 ± 0.52) can be calculated with Eq. 1, which exhibited the dyed samples with AE extraction having the satisfied color fastness of water property compared with the previous study (Deng *et al.* 2009). This may be attributed to the dyestuffs distribution form in the wood tissues leading to larger potential physical binding force generated between the dyestuff and wood tissues, as indicated by the preceding SEM analysis. However, further studies to account for this phenomenon are still needed.

**Fig. 11.** Color parameters variation of dyed samples before and after being soaked in hot water

CONCLUSIONS

1. Extracts from *D. cochinchinensis* process remains with four different organic solvents can be used as natural dyestuffs to dye wood veneers directly. The absolute ethanol (AE) extract obtained the most favorable dye effect according to the measured ΔE and K/S values. The change of ΔE was significantly influenced by the variation of ΔL^* , followed by Δa^* , and the influence of Δb^* represented was relatively minor. In addition, the performance of the color fastness of water of the dyed veneers with AE extraction was better compared with the previous findings.
2. Only physical absorption occurred between the dyestuff and wood tissues according to the Fourier transfer infrared (FTIR) analysis of the untreated and the samples dyed with AE extraction. The thermogravimetric (TG) analysis showed the residue of dyed samples was more than that of the untreated samples, which demonstrated the thermal stability of samples increased after dyeing with AE extraction.
3. Scanning electron microscopy (SEM) analysis indicated that the distribution form of natural dyestuffs was akin to a membrane covering the wood tissues, including vessel walls, wood rays, and wood fibers, which was completely different from the dyestuffs distribution form of synthetic dyestuff in the previous study.

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