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Research article

Effect of tannic/gallic acid-iron dyeing treatment on surface color and light fastness of bamboo veneer

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ABSTRACT

Currently, the quest for bamboo materials with high color fastness, rich colors and environmental friendliness is rapidly rising due to its potential applications in construction, furniture and decoration. However, finding an easy-to-operate and environmentally friendly dye for bamboo is a necessary task because of the difficulty in treating the dyeing waste liquid of acid dyes and the complexity of the production process of reactive dyes.Five formulations involving metal polyphenol complexes were employed to straightforwardly produce eco-friendly dyed bamboo and the impact of various formulations on the light aging resistance of the dyed veneers was examined. The results indicated that the light resistance of bamboo veneer dyed with the solution containing only $FeSO_4 \cdot 7H_2O$ and tannic acid reached level 4, surpassing the undyed bamboo veneer by three levels. The mechanism of enhanced lightfastness of dyed bamboo veneer was elucidated by XPS analysis. The polyphenol iron complex serves a dual purpose: it absorbs ultraviolet rays and scavenges free radicals within the system. Additionally, it reduced the oxidation of phenolics in the substrate, transforming them into dark-colored quinone structures. This process enhanced the light-aging resistance of the finishing materials. Therefore, this work provides a simple and environmentally friendly method for changing the color of bamboo and provides a new idea for the selection of dyes for bamboo dyeing in actual production.

1. Introduction

Bamboo is an environmentally friendly renewable material that is ideal for replacing wood as a raw material for engineering construction and decoration because of its high strength and toughness, due to the special anatomical features including all longitudinally orientated cellular tissues [1]. The increasing demand for sustainable and environmentally friendly materials in recent years has made bamboo and bamboo products highly valuable in research. Its utilization in various forms as one of the surface decoration materials of wood-based panels makes it widely used in the manufacture of high-grade indoor and outdoor decorative materials and furniture materials, but the monotonous colors and patterns limit its development thus it needs to be discolored. At present, the primary methods for controlling the color of wood materials are physical [2], chemical [3] and other methods of dyeing, as well as the application of light [4], heat [5,6] and other means. The principles of biomimetic structural color [7–9] and bio-dyeing [10,11] have also been utilized to regulate the color of wood research as well. Bamboo discoloration has also been studied by carbonization, alkali

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treatment, and biological treatment [12–14], but the waste liquid from carbonization is difficult to handle and large amounts of heat generated during the heat treatment process pollute the environment [15,16].

The color regulation of bamboo is often based on dyeing treatments, and most of the dyes used for bamboo dyeing are similar to those used for wood. The dyes currently used for bamboo are primarily acid dyes containing sulfonic acid group, carboxylic acid group, and other acidic groups, which can achieve the desired dyeing effect by combining with the amino or acylamino group in the bamboo material [17]. Its dyeing effect is superior, but the wastewater contains not only dyes but also a significant amount of organic matter, resulting in chromaticity, a high chemical oxygen demand, and weak biochemical properties [18]. Therefore, based on the context of the global low-carbon economy, there is an urgent need for a straightforward, eco-friendly, and sustainable way of changing color to improve the visual properties of bamboo, expand its application areas and increase additional-valued bamboo products.

Plant dyeing refers to the process of using plant extracts for dyeing, which has the advantage of being environmentally friendly and is widely used in food colorants as well as the dyeing of fibrous materials such as fabrics, yarns, paper-based packaging materials, etc. However, the exploration of the use of this method in the fields of wood and bamboo dyeing is still in its infancy [19–22]. The property of polyphenols complexing with iron ions is widely used in designing multifunctional materials, which have broad applications in the fields of inks, food, and the environment [23]. Dai et al. developed a green and portable printing technology by using different polyphenol-metal ions to form MPNs of various colors in situ on the surface of the substrate [24]. Limaye achieved reversible color change on the surface of cellulose nanofilm using the pH responsiveness of the polyphenol-iron system [25]. Since polyphenols are distributed in the xylem of most tree species, the iron ion complex method for wood discoloration has attracted the interest of many researchers. Polyphenol-iron discoloration system makes use of phenolic substances extracted from plants and a small amount of iron salts as a medium to produce vegetable dyes, which has the same low-carbon and environmentally friendly benefits as natural dyes, and the finished products have uniform color, high color fastness, and a strong sense of texture hierarchy [26]. To obtain a dark oak flooring with stable color, Qiu et al. used various concentrations of Fe(III) solution to dye the surface of European oak wood [27]. Cao et al. investigated the effect of Fe(II) concentration, immersion time, immersion temperature, and other factors on the coloring of hemp oak veneer by analyzing them experimentally and giving the optimal treatment process [28]. The author's team invented a color-changing wood material by taking advantage of the property that polyphenols are highly susceptible to complexes with iron ions to generate black polyphenol-iron complexes. However, the majority of the polyphenol-iron system discoloration studies that are currently available use iron salt solution brushed on the surface of wood containing polyphenols or wood directly impregnated in an iron salt solution. It is challenging to use this process to dye materials like bamboo, ornamental paper, and other items with poor polyphenols.

In order to obtain a new environmentally friendly dyeing method for bamboo discoloration, five polyphenol-iron solutions were compounded to directly dye bamboo veneers lacking polyphenols. In this work, bamboo veneers with cool colors, mainly gray-black and blue-black, were obtained and evaluated for light fastness using a xenon lamp, and the dyeing solution with the most significant improvement in light fastness was picked. The bamboo veneers before and after dyeing and light aging were also examined by Scanning Electron Microscope (SEM) and X-ray Photoelectron Spectroscopy (XPS), which further explains the reasons for the good light fastness of bamboo veneers prepared using the polyphenol-iron discoloration system. On this basis, this study enriches the color of bamboo, investigates a novel low-energy dye for bamboo dyeing, offers a fresh perspective on dye selection for bamboo dyeing in actual production, and broadens the application range of polyphenol-iron discoloration system at the same time.

Table 1
Specific ingredients and contents of the five polyphenol-iron solution formulations.

Formula Number	Reagent	g/mL
①	TA	0.0117
	GA	0.0038
	$FeSO_4 \cdot 7H_2O$	0.015
	10 % Hydrochloric acid	0.0125
	Phenol	0.001
	Methyl Bule	0.0035
	Acacia	0.001
②	TA	0.0117
	GA	0.0038
	$FeCl_2 \cdot 4H_2O$	0.015
	10 % Hydrochloric acid	0.0125
	Phenol	0.001
	Methyl Bule	0.0035
	Acacia	0.001
③	$FeSO_4 \cdot 7H_2O$	0.015
	TA	0.0117
	GA	0.0038
④	$FeSO_4 \cdot 7H_2O$	0.015
	TA	0.0117
⑤	$FeSO_4 \cdot 7H_2O$	0.015
	GA	0.0038

2. Materials and methods

2.1. Materials

Moso bamboo *(Phyllostachys edulis (Carrière) J. Houz.)* sliced veneers were purchased from Yifeng County, Jiangxi Province, China, with dimensions of 350 mm × 130 mm × 0.7 ± 0.1 mm and with a moisture content of about 12 %. Fe (II) sulfate heptahydrate ($FeSO_4 \cdot 7H_2O$, industrial grade, 96 % purity) was obtained from Aladdin Reagent (Shanghai) Co., Ltd. Tannic acid, gallic acid, ferrous chloride tetrahydrate, phenol, methyl blue, and acacia were all AR, 99 % purity, obtained from Macklin Biochemical (Shanghai) Co., Ltd.

Polyphenol-iron dye solutions, formulated using different reagents, were all fixed to 300 ml. The configured dye solution was subjected to a filtration process and stored in a static position protected from light. The types and contents of the components of different formulations of polyphenol-iron dye solution are shown in Table 1.

2.2. Discoloration and aging of bamboo veneer

Firstly, all the bamboo veneers were placed in a constant temperature and humidity box to reach the equilibrium moisture content and then impregnated with different formulations of dyeing solution from Table 1 at atmospheric pressure for 2 h. The test was carried out at room temperature, about 25 °C, and finally, the discolored specimens were placed in a cool, light-protected place to dry for 24 h. The control and specimens after staining with different formulations of dye solutions were named ZC, Z1, Z2, Z3, Z4, and Z5, respectively.

The XE-2-HS xenon lamp climatic test chamber was used to simulate solar illumination, and the instrument parameters were set as follows: radiation intensity 1.10 W/m^2 at 420 nm, temperature 38 °C, relative humidity 50 %. All specimens were fixed in the instrument specimen frame for radiation, respectively, in the xenon lamp before irradiation and irradiation treatment 3, 9, 15, 24, 36, 48, 72, and 96 h after the colorimeter to measure the specimen color.

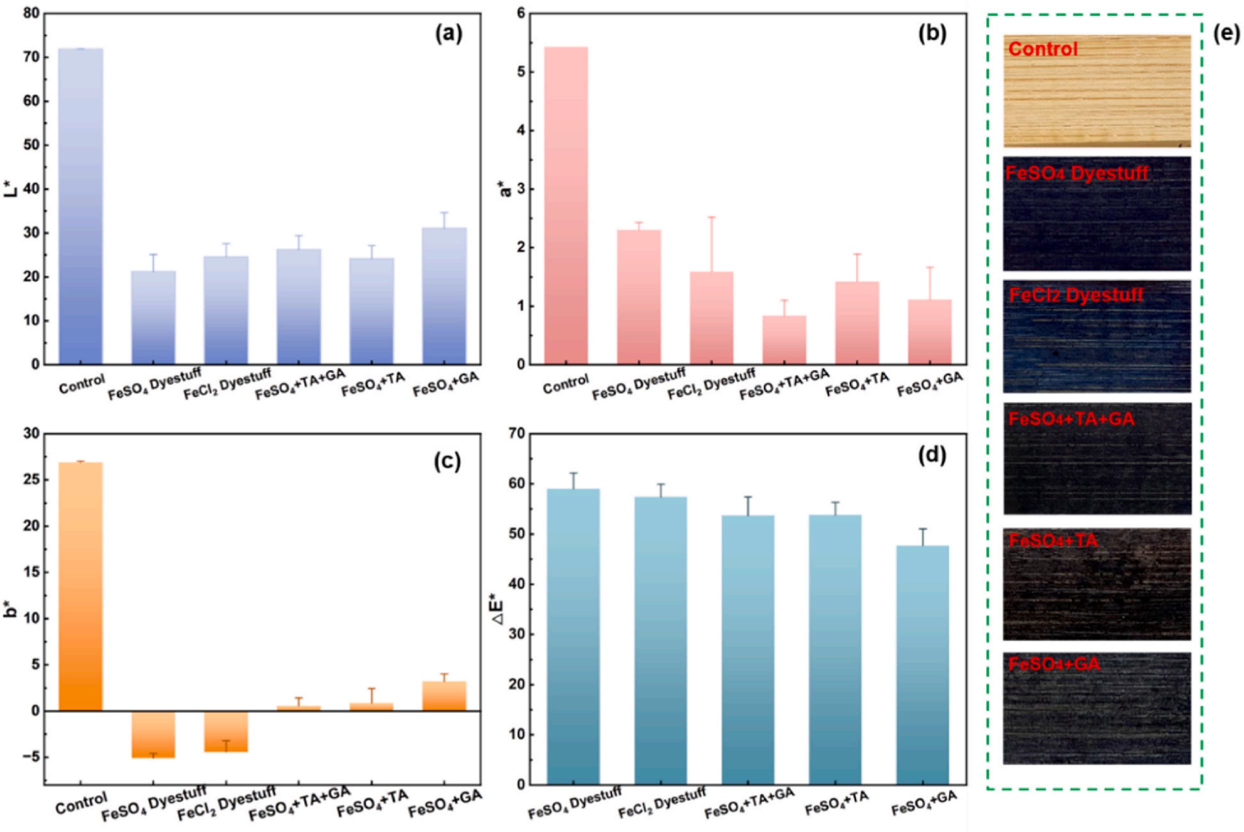

Fig. 1. L*/a*/b* (a–c) Chroma value and Overall color difference ΔE* (d) of control and treated bamboo veneers, and visual expression (e) of bamboo veneer before and after staining.

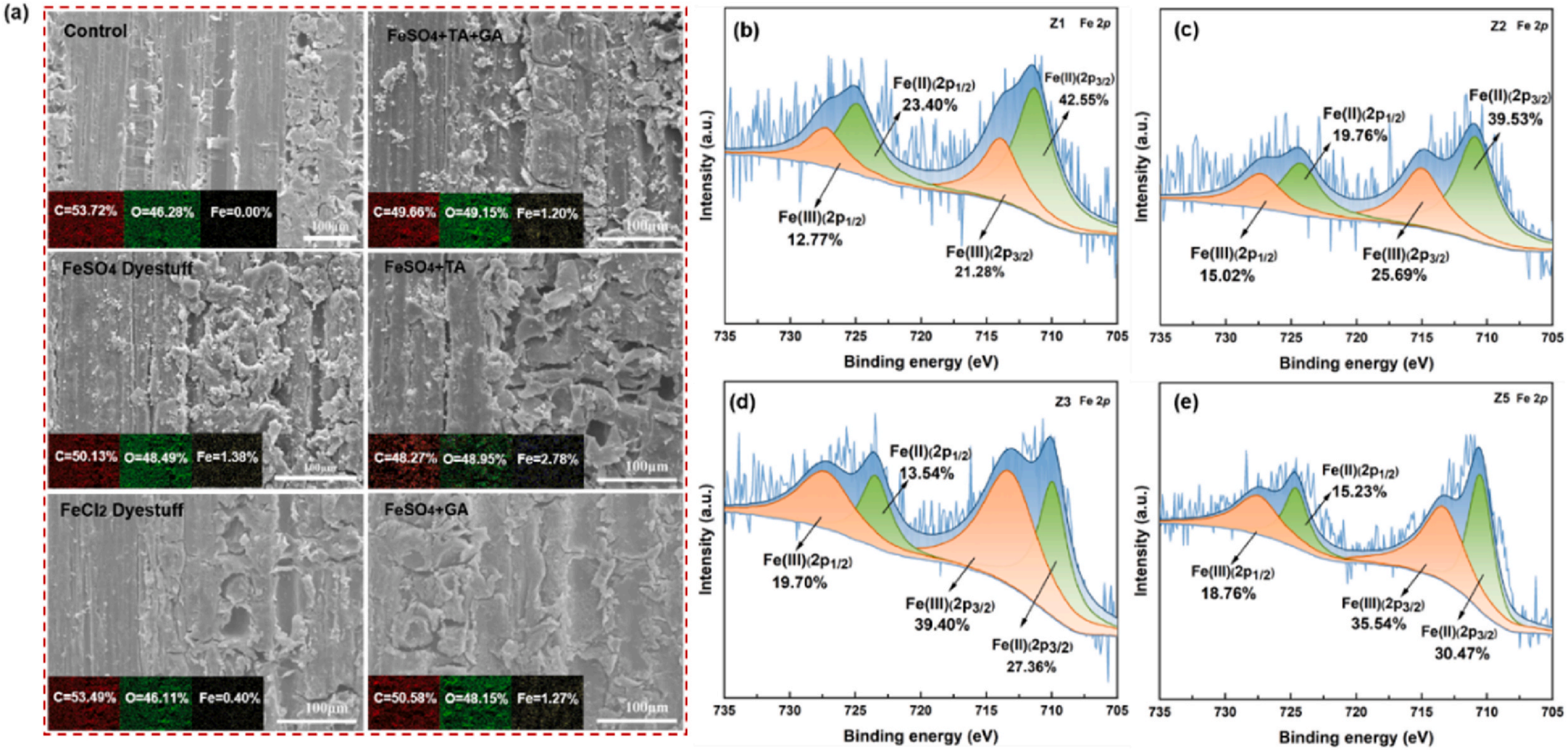

Fig. 2. SEM observations and EDS scanning images (a) of samples, XPS spectra of Fe 2*p* of Z1-Z3 and Z5 veneers (b–e).

2.3. Characterization

The surface morphologies of all the veneer samples were imaged by using an SEM (Hitachi, SU8020, Japan) coupled with an energy-dispersed X-ray (EDS) analyzer (Horiba 7021-H, Japan) under an acceleration voltage of 20 KV. A spectrometer (Thermo Scientific K-Alpha) with a monochromated AI K α X-ray (1486.6 eV) source was used to study the X-ray photoelectronic spectroscopy (XPS) of the veneer samples.

The color was measured before and after the discoloration and during the aging process and characterized by the CIE Lab color system, proposed by the International Lighting Association to assess the color difference [29]. L* indicates the brightness or luminance, i.e., the degree of black to white, a* indicates the degree of red and green, and b* indicates the degree of yellow and blue. L* = 0 is black, L* = 100 is pure white; the positive value of a* is red, and the negative value is green; the positive value of b* is yellow, and the negative value is blue. Three specimens were measured in each group, the chromaticity coordinates L*, a*, b* of the color system of two points were measured for each specimen with a chroma meter (CR-400, K&M Co., Japan) and D65 light source, and the value of L*, a*, and b* were averaged. Calculate the luminance difference ΔL*, the red-green product index difference Δa*, the yellow-blue product index difference Δb*, and the total color difference ΔE* before and after the light exposure of the impregnated specimens, and refer to the LY/T 3138-2019 for the color difference grading standard with the following equations (1)–(4):

$$\Delta L^* = L^* - L_0^* \tag{1}$$

$$\Delta a^* = a^* - a_0^* \tag{2}$$

$$\Delta b^* = b^* - b_0^* \tag{3}$$

$$\Delta E^* = \left[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2\right]^{1/2} \tag{4}$$

where L*, a*, and b* indicate the color index value of the sample after irradiation, L_0*, a_0*, and b_0* indicate the color index value of the sample before irradiation.

3. Results and discussion

Fig. 1 shows the chromaticity values and color visualization expressions of the samples after treatment with different formulations of polyphenol-iron dye solutions. According to Fig. 1a–c, the luminance value L* and the yellow-blue magenta index b* of the samples treated with polyphenol-iron solutions were substantially reduced, and the samples were transformed from the original yellow color of

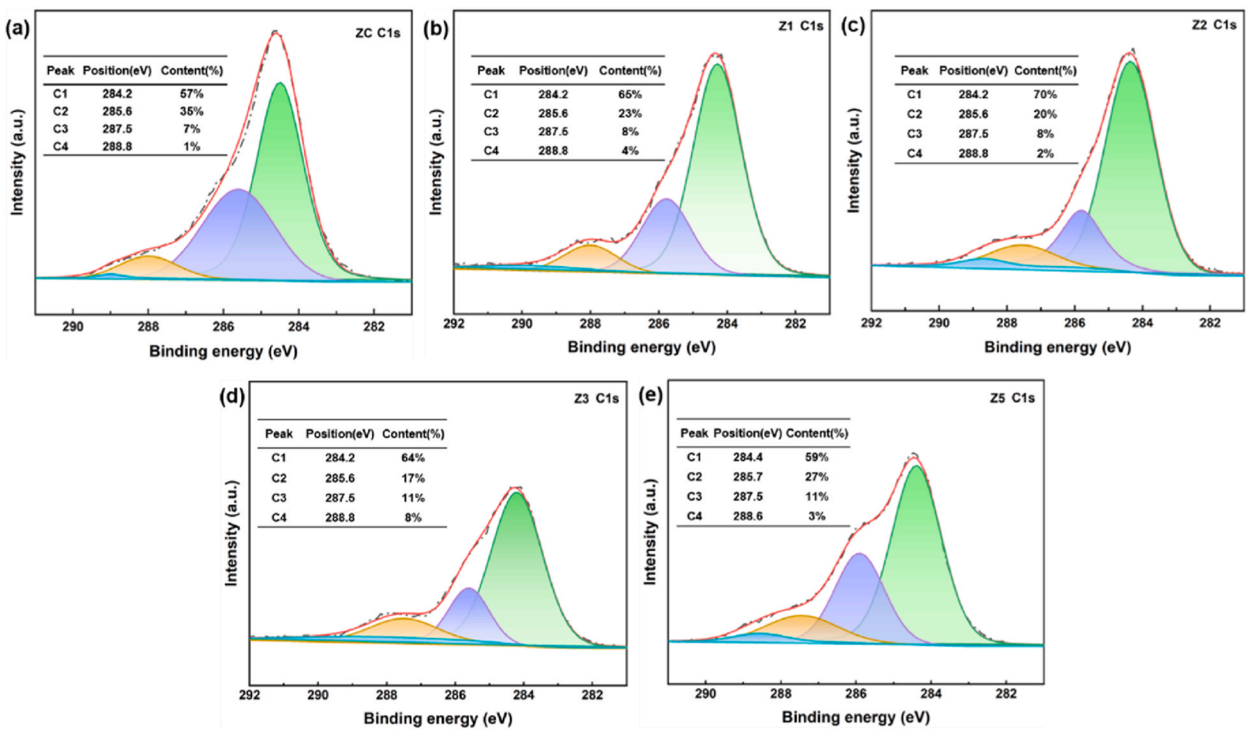

Fig. 3. XPS spectra of C 1*s* of ZC veneers (a), XPS spectrum of C 1*s* of Z1-Z3、Z5 veneers (b–e).

bamboo to blue-black and gray-black. Fig. 1b showed that the a* values of bamboo veneer were most affected by the solution containing only $FeSO_4$, TA and GA (formulation ③). From Fig. 1d and e, it can be intuitively found that different polyphenol iron solutions stained the specimens differently, with the specimens treated with staining solution (formulation①) and $FeCl_2$ staining solution (formulation②) showing an overall bluish-black color with a much larger difference in the coloration, whereas those stained with the addition of only $FeSO_4$ and the different polyphenols were gray-black. Different pH environments resulted in different degrees of complexation of polyphenols with metal ions in the solutions. Moreover, different staining effects were produced by combining pH and Methyl Blue solution, which exhibits a blue color in formulations ① and ②.

The complexation reaction between polyphenols and iron is a complex process. Fe(II) in solution reacts with tannic acid or gallic acid to generate unstable ferrous tannate or ferrous gallate, which is oxidized to a dark-colored trivalent iron complex in air, and polyphenols reduce Fe(III) to Fe(II), which oxidizes itself to quinone, resulting in a darker color [30,31]. Dark-colored polyphenol-iron complexes adhered to the surface of the bamboo veneer, thus resulting in the decreasing lightness value L*. As a result, the bamboo veneers Z3, Z4, and Z5 had gray-black surfaces, which was in line with Huang and Lin's findings [32,33].

Fig. 2 shows the distribution of the colored complexes on the specimen and its XPS spectra of Fe 2*p*. From the EDS energy spectrum of elemental Fe in Fig. 2a, it was found that the polyphenol-iron complexes were uniformly adhered to the surface of the bamboo veneer, which made the bamboo veneer show different colors while preserving the natural texture of the bamboo veneer. According to Fig. 2b–e, Fe(II) and Fe(III) coexisted on the surface of bamboo veneer, and the surface of greyish-black Z3, Z4, and Z5 was dominated by Fe(III), while the surface of blue-black Z1 and Z2 samples was dominated by Fe(II). It was probably because the acidic environment of the ① and ② dye solutions protonated most of the phenolic hydroxyl groups on the polyphenol molecules, which resulted in the formation of competitive chelating between protonated hydrogens and metal ions [34], thus inhibiting the complexation of polyphenols with ferrous ions. Methylene blue, which displays a blue color, was also added to both formulations, and its combination with competitive complexation provided a dark blue color for Z1 and Z2. The polyphenol-iron complexes exhibit a range of forms and colors depending on the pH level. They can form one-coordinate complexes at pH levels below 2, two-coordinate complexes at pH levels between three and six, and three-coordinate complexes in alkaline settings [34]. Similarly, the pH of the dye solution influences the color change system's effectiveness when using polyphenol-iron complex as the dye. Consequently, for the purpose of avoiding influencing the dyeing effect of bamboo, companies must closely regulate the pH environment of the dyeing solution during actual production.

Fig. 3 shows the XPS high-resolution spectra of the C 1*s* region of samples before and after discoloration, where the C 1*s* peak can be

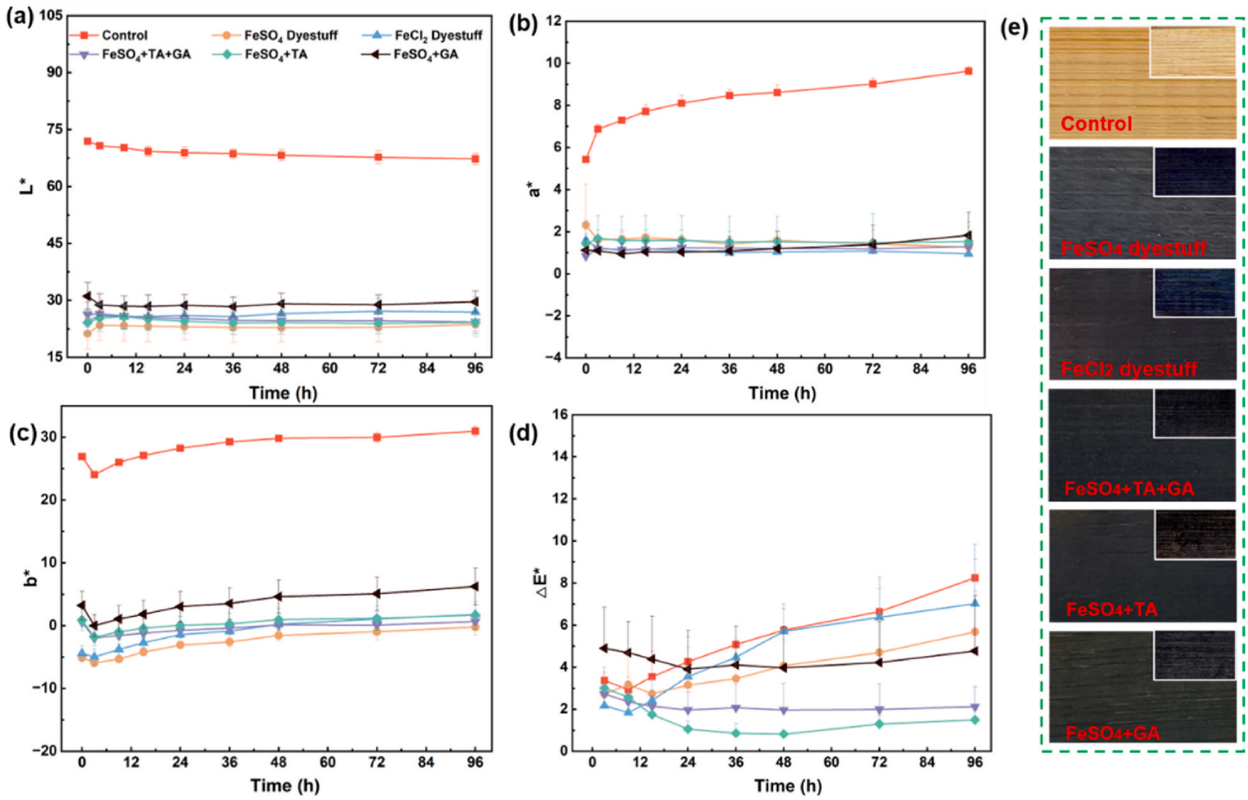

Fig. 4. Curves of L*/a*b* Chroma value (a–c) and color differences ΔE* (d) of specimens treated with different formulations of polyphenol-iron dyes with light aging times. Digital images (e) of the sample before and after aging, the white box in the upper right corner of the image represents the sample before aging.

subdivided into four components corresponding to C1 (C═C or C–C), C2 (–C–O), C3 (–O–C–O– or –C═O), and C4 (O═C–O–) [35]. Compared the control bamboo of Fig. 3a and the stained bamboo veneers of Fig. 3b–e, it was obvious that the proportion of C1, C3, and C4 increased after staining, and the content of C2 decreased to different degrees, which was due to the surface of bamboo veneer adhering to the dye solution and phenolics in the dye solution consuming phenolic hydroxyl groups by complexing with ferrous ions and oxidizing into quinone by themselves, resulting in a decrease in the proportion of C2 and an increase in the proportions of C1, C3, and C4. The largest decrease in the content of C2 of the bamboo veneer of Z3 ($FeSO_4$ +TA + GA), was possibly due to the polyphenols in the dye solutions with different degrees of complexation with ferrous ions.

Fig. 4 shows the effect of polyphenol-iron solution treatment on the light discoloration of bamboo veneer, and the change in chromaticity value and the total color difference of bamboo veneer after 96 h of xenon lamp irradiation are shown in Table 2 and Fig. 4d. It could be found intuitively that the color of bamboo veneer before and after light aging became darker and yellower from Fig. 4a, e; the increase of the reddish-green color a* (Fig. 4b) and the yellowish-blue color b* (Fig. 4c) could also prove this point. The ZC sample showed the biggest change because the photodegradation of the bamboo veneer in the UV light caused photochemical reactions like lignin degradation to produce a red o-quinone structure and a yellow p-quinone structure. This led to the discoloration in the direction of the red and yellow development [36].

The changes in the chromaticity values of the polyphenol-iron solution-impregnated specimens were mainly concentrated in the early stages of xenon lamp irradiation. After the luminance value of the dyed-treated bamboo veneer tended to stabilize, the L* of ZC still tended to decrease, indicating that the polyphenol-iron solution impregnation-treated specimen had better chromatic stability compared with the untreated specimen (Fig. 4a). After 96 h of irradiation, the a* values of the dyed bamboo slices changed very little, and the five polyphenol-iron solution dyeing treatments could reduce the change of red-green index a* in the process of photochromism; however, only solutions ③ and ④ could reduce the change of b* values, and thus the color fastness ratings of Z3 and Z4 were higher. Compared with ZC, the total color difference ΔE* of light aging of bamboo slices dyed with five polyphenol-iron solution treatments was reduced to different degrees, among which Z3 and Z4 samples were more effective in resisting light discoloration, with the total color difference reduced by 74.3 % and 81.9 % (Table 2), respectively, and their light fastness level reaching 4, which was attributed to the polyphenol-iron complex on the surface absorbing the ultraviolet light and isolating the role of oxygen. The color stability of Z1 and Z2 samples was not significantly improved compared with ZC, which may be related to the chelation degree of polyphenols with Fe(II) and the acidity and alkalinity of the dye solution.

The results of the XPS test on the oxygen-containing functional groups on the surface of dyed bamboo supported the conclusion that the dye solution improved the lightfastness of bamboo to a certain extent. Fig. 5 shows the XPS C 1*s* and Fe 2*p* spectra before and after the aging of the Z4 specimen. The lightfastness of the Z4 specimen had the best effect. According to Fig. 5a_1-a_2, the C1 content on the surface decreased, and the C2, C3, and C4 contents all increased after UV aging, which was the same as the experimental results of wang [37]. When UV light irradiated the dyed veneer, part of it was reflected, part of it was absorbed, and the rest of it reached the surface of the bamboo veneer through the coating. At this time, the polyphenol-iron complexes attached to the substrate surface acted as the organic UV absorber, replacing the bamboo veneer to absorb part of the UV energy to occur photochemical reactions, including phenolics photodegradation, conjugated double bond reduction, resulting in a decrease in the content of C1. And phenoxy radicals photodegradation in the presence of oxygen and water induced the oxidation of phenolics in polyphenol-iron complexes to quinone, leading to an increase in C3 content [38]. It corresponded to the increase in a* and b* of bamboo veneer after UV aging. However, polyphenol-iron complexes did not completely block or absorb ultraviolet rays, so there was still a part of the bamboo veneer lignin decomposition, methoxy content increased, and the relative content of C2 increased. Fe(II) was further oxidized to Fe(III), which consumed the free radicals in the system and reduced the photodegradation of lignin in the substrate, thus enhancing the photostability of the finishing material and effectively preventing the photochromic discoloration of the substrate (Fig. 5b_1-b_2).

4. Conclusion

In this paper, we prepared five polyphenol-iron solutions for eco-friendly bamboo veneer dyeing. Using a xenon lamp aging apparatus, we accessed their light fastness level, resulting in cold-toned bamboo veneer with predominant blue-black and gray-black shades. We identified solutions that notably enhanced the resistance to light-induced discoloration of bamboo and elucidated the underlying mechanisms. Among the five polyphenol-iron solutions, solution ③ ($FeSO_4·7H_2O$ + TA + GA) and solution ④ ($FeSO_4·7H_2O$ + TA) showed the most significant improvement in bamboo light fastness, achieving a light fastness grade of 4 for the dyed bamboo veneer, a notable improvement compared to the undyed veneer by three grades. This enhancement can be attributed to the ability of polyphenol-iron complex to absorb ultraviolet light and effectively shield the substrate from photo-induced discoloration.

Table 2
Total color difference and grade of specimens before and after aging.

	ΔL*	Δa*	Δb*	ΔE	Color Difference Grade
ZC	−4.68	4.19	4.10	8.24	1–2
Z1	2.40	−1.04	4.87	5.68	2–3
Z2	2.30	−0.64	6.18	7.02	2
Z3	−1.91	0.46	0.07	2.12	3–4
Z4	−0.01	0.11	0.81	1.49	4
Z5	−1.47	0.71	3.01	4.76	2–3

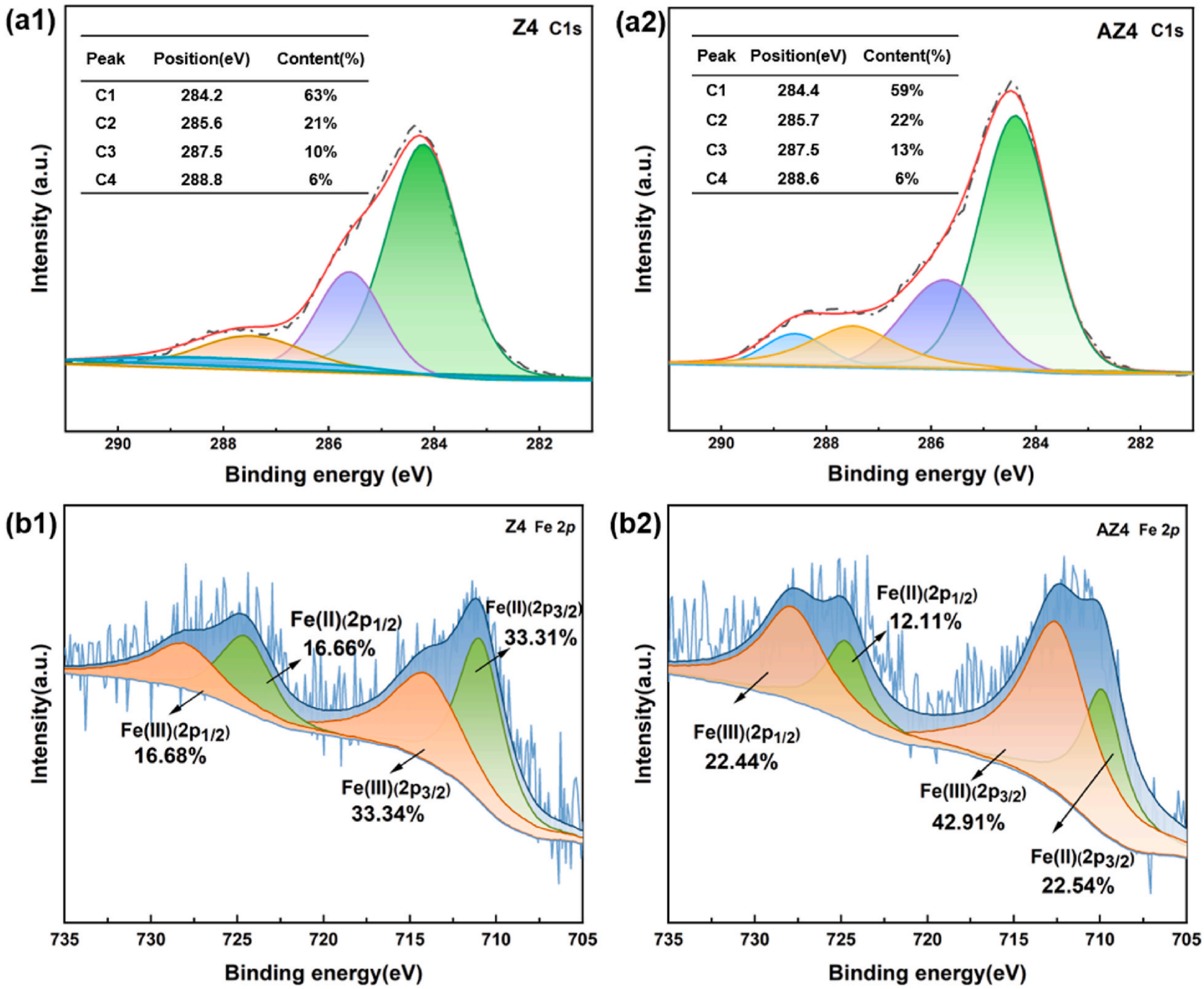

Fig. 5. XPS C 1*s* and Fe 2*p* spectra before (a_1, b_1) and after aging (a_2, b_2) of Z4 specimen.

Simultaneously, in the presence of an oxygen-rich environment, the conversion between Fe(II) and Fe(III) occurs within the polyphenol-iron complex. This process consumes free radicals within the system and mitigates the oxidation of phenolic compounds in the substrate, transforming them into darker quinones. Consequently, this mechanism bolsters the finish material's resistance to light-induced aging.

Bamboo dyed with minimal energy consumption can be used to make furniture, wall panels, flooring, handicrafts, and packaging materials. In this study, we focused only on the color modulation of bamboo, using two of the most common polyphenols, tannic acid and gallic acid, to change the color by complexing with iron ions. Future studies can create a variety of colors of dyed bamboo by varying the metal ions and polyphenol kinds, develop more eco-friendly bamboo dyes, improve the dyeing process, and offer a theoretical basis for actual production. The polyphenol-iron discoloration system can also be employed on diverse substrates to expand its applicability.

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Data availability statement

Data will be made available on request.

Additional information

No additional information is available for this paper.

Ethics declarations

Review and approval by an ethics committee was not needed for this study because it did not involve biomedical research.

CRediT authorship contribution statement

Mengjia Zhu: Writing – original draft, Investigation, Formal analysis, Data curation. **Shiqin Liu:** Methodology, Investigation, Data curation. **Huijuan Bai:** Resources, Formal analysis. **Yuxiang Huang:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Yanglun Yu:** Writing – review & editing, Resources, Methodology, Conceptualization. **Wenji Yu:** Supervision, Resources, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e24082.

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