

Research Article

Effect of Iron Oxide on the Protective Photochromism of African Padauk

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African padauk is a tree whose wood is an excellent material for producing decorative items. The wood is susceptible to sunlight exposure and degrades severely, even after short exposures, which leads to discoloration. Ultraviolet irradiation is the main factor that produces discoloration. In this study, chemical changes occurring on the wood surfaces were analyzed by Fourier transform infrared spectroscopy. The discoloration of the African padauk was mainly because of the photodegradation of lignin and extractives by ultraviolet radiation. Additionally, the inhibitory effect of different concentrations of red iron oxide coating was investigated. The results revealed that the chromatism ΔE^* was considerably reduced with the protection of iron oxide. However, the inhibition of the discoloration rate of the specimens covered with a film of red iron oxide increased with values reaching approximately 80%. The red iron oxide coating had a strong inhibitory effect on the discoloration of the African padauk.

1. Introduction

Wood color is an important quality indicator for end users, especially for interior items, such as flooring and furniture. However, the wood color tends to change when exposed to the environment. Usually, wood discoloration is caused by a variety of environmental factors, including sunlight, moisture, and temperature. Among these factors, ultraviolet (UV) light, a component of sunlight, plays the most important role in the discoloration process by inducing photodegradation in wood [1–3]. Because of the decrease in aromatic structures and increase in carbonyl groups, photodegradation of wood eventually results in discoloration on the wood surface [4–6].

African padauk is a hardwood tree species of the genus *Pterocarpus* in the Fabaceae family and is native to tropical areas in Africa. It has a relatively high extractives content and mainly contains homopterocarpin, pterocarpin, and 4-*t*-butyl-2-(4-nitrophenyl) phenol [7]. These compounds give the African padauk its appealing color. In general, this wood is both stable and durable, which makes it a suitable raw material for interior decoration applications. With the decrease in rosewood resources, the value of African padauk is increasing. Previous studies have shown that African

padauk is more prone to degradation under sunlight exposure [8, 9]. Photodegradation causes the color to change initially to red and eventually to brown. Uneven discoloration greatly reduces the reliability and value of African padauk in practical applications. Although many researchers have studied the wood photodiscoloration process [10], studies on the discoloration of African padauk are scarce. A better understanding of the discoloration mechanism of African padauk is needed to determine the appropriate measures for its protection.

The cell wall of wood consists mainly of cellulose, hemicellulose, lignin, and extractive components. Lignin and extractives are more susceptible to light than the other two major components [11, 12]. When lignin is irradiated, the carbonyl, phenolic hydroxyl, and methoxy groups in the lignin with benzene rings form free radicals. The free radicals undergo a photooxidation reaction and generate chromophoric carbonyl and carboxyl groups, which are responsible for wood discoloration. Photooxidative degradation of lignin only occurs in the surface layer of wood. Extractives also play a critical role in the photodegradation of wood. Some researchers believe that extractives are involved in wood discoloration, such as the photodegradation of

polyphenolic substances [13, 14], while others maintain that extractives act as antioxidants to protect the wood against photodegradation [15–17]. Earlier results have indicated that the formation of chromophoric groups, such as C=C and C=O, during the photooxidation process is a major cause of wood discoloration. Variations in the chemical characteristics of the wood surface after irradiation can be detected by means of Fourier transform infrared (FT-IR) spectroscopic analysis [18–20].

Wood photochromism usually occurs on the surface. Different techniques have been developed to address this problem and reduce the photodegradation of wood, including chemical modification, surface coatings, and other methods [21, 22]. Surface coatings are the most convenient and commonly used method, which can form a protective layer by adding the right amount of UV screeners in the coating [23]. Coating systems for protecting the wood from discoloration, including organic UV absorbers and hindered amine light stabilizers, have traditionally not been used for long-term protection. In recent years, inorganic UV absorbers have attracted attention as an alternative treatment for protecting wood surfaces against UV radiation [24, 25]. Adding the nanoparticles TiO₂, ZnO, and SiO₂ to acrylic resin coatings can also have an effect on the shielding of UV radiation [26–28]. However, nanoparticles easily aggregate and weaken the ability to scatter UV light. Additionally, the cost of a nanocoating is higher and the process still remains in the research stage.

Iron oxide pigments as an inorganic UV absorber can prevent light irradiation from reaching wood surfaces and the initiation of free radical reactions, thus providing protection against discoloration. Schauwecker's research shows that particle size of iron oxide and protective effect of wood present the correlation [29]. Moreover, low concentrations of iron oxides have also shown promise as wood surface protection agents, reducing lignin loss and discoloration [30]. Zhang et al. added four kinds of iron oxide pigments into wood-fiber/high-density-polyethylene composites; the experimental results show that the iron oxide red and black performed better than the other two pigments in protection of wood color and mechanical properties [31].

In general, because of similar color of African padauk, the red iron oxide is suitable for surface finishing of African padauk. This study investigated the mechanism of discoloration and the effect of red iron oxide on the discoloration of African padauk. As a tropical hardwood, it is conceivable that the extractives of African padauk may play an important role in discoloration. Fourier transform infrared spectroscopy was used to analyze the variation in the chemical characteristics of the specimen surfaces after irradiation. Different concentrations of red iron oxide were used to examine the color change after irradiation.

2. Experimental

2.1. Materials. The logs of African padauk were purchased from Power Dekor (Danyang, China); details are listed in Table 1. The logs were obtained from mature wood above breast height without any cracking, decay, discoloration, or

TABLE 1: Essential parameters of African padauk.

Wood	African padauk
Scientific name	<i>Pterocarpus soyauxii</i>
Density	0.79 g cm ⁻³
Place of origin	Republic of Congo
Water content	8 %
Sample size	75 mm(L) × 50 mm(R) × 4 mm(T)

TABLE 2: Basic information of the red iron oxide.

Property	Transparent Red Iron Oxide
Molecular formula	α -Fe ₂ O ₃
Molecular weight	159.69 g/mol
Appearance	Red powder
Melting point	1565 °C
Boiling point	3414 °C
Iron content	≥ 95% (m/m)
105 °C Volatiles	≤ 1% (m/m)
Hydrotrope	≤ 0.3% (m/m)
Paste content	50%
Particle size	0.4 μm ~ 20 μm

other defects. The red iron oxide was purchased from Tianjin North Coating Co., Ltd. (Tianjin, China), and the basic pieces of information are showed in Table 2.

2.2. Methods

2.2.1. Specimen Coating. The oxide was provided with a special megilp solvent. All of the specimens were polished with sandpaper (400 P) prior to the experiment and stored in a dark environment. To determine the influence on the original color of the African padauk, 3%, 6%, 9%, 12%, and 15% red iron oxide solutions were prepared and tested. The red iron oxide concentration was calculated according to

$$W = \frac{m_1}{m_1 + m_2} \times 100\% \quad (1)$$

where W , m_1 , and m_2 are the concentration of the red iron oxide (%), a mass of the red iron oxide (g), and mass of the megilp (g), respectively.

The specimens were coated using a brushing method and then air-dried. The coating weight was $0.2 \text{ g} \pm 0.01\text{g}$, which was calculated according to

$$\Delta m = m_4 - m_3 \quad (2)$$

where m_3 is the mass of the specimens before coating with red iron oxide (g) and m_4 is the mass after coating with red iron oxide (g).

2.2.2. Color Measurement. There are three samples in each group. The specimens were separately exposed to natural and artificial UV light. Half of each specimen surface was covered with aluminium foil prior to exposure so that the difference between the exposed and original surface was clearly visible.

Natural weathering was achieved in the absence of wind and rain. Artificial weathering was conducted in an accelerated UV radiation chamber (LX-8509, Dongguan Lixin Instruments Co. Ltd., Dongguan, China) equipped with two UVA-340 lamps with a radiation intensity of 30 W/m², which provided a simulation of sunlight radiation in the short wavelength region of 290 nm to 400 nm.

The L^* , a^* , b^* , and ΔE^* parameters were measured by a chromatic aberration meter (CR-300, Minolta Co., Osaka, Japan) at five places on each specimen. The average values were subsequently calculated. Each specimen was tested three times and the average of those values was recorded. The initial color measurement was determined prior to any light irradiation. The intermediary color measurements were performed after 2 d, 4 d, 8 d, 12 d, 20 d, 40 d, 60 d, and 80 d of natural sunlight exposure and 8 h, 16 h, 32 h, 56 h, 80 h, 104 h, and 128 h of UV light exposure.

The overall discoloration of the surface was established by the color deviation ΔE^* , which was calculated using the following formula:

$$\Delta E^* = \sqrt{[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]} \quad (3)$$

where ΔL^* , Δa^* , and Δb^* are the differences between the original and final coordinates (before and after exposure).

2.2.3. FT-IR Spectroscopic Analysis. The specimens were exposed to natural sunlight and artificial UV light until their color stabilized. The powder was obtained using a crusher (BJ-300A, Deqing Baijie Electrical Appliance Co. Ltd., Deqing, China). The discolored powder was obtained by scraping the wood surface with a knife. For extraction of the solid wood, the specimens were soaked in a mixture of ethanol-benzene (1:2 ratio) for 6 h in a Soxhlet apparatus, followed by extraction in pure ethanol for 4 h. The specimens were then oven-dried at 103°C until the weight was constant. Two milligrams of each specimen were blended with 100 mg of KBr powder, ground again, and pressed into tablets. FTIR analyses were carried out at room temperature according to the methodology described by NICOLET 6700, using a Spectrum 2000 FTIR (Thermo Fisher Scientific, Massachusetts, USA) with a universal attenuated total reflectance (UATR) accessory. Potassium bromide (KBr) was used to collect background spectra. Air-dried sample powders were mixed with KBr at a weight ratio of 1:100 before spectrum collection. Ten scans were carried out for each sample in a spectral range from 400 to 4000 cm⁻¹ at a resolution of 4 cm⁻¹.

3. Results and Discussion

3.1. FT-IR Analysis. The FT-IR spectra of the African padauk before and after irradiation are presented in Figure 1. The spectra showed that the intensity of the absorption peak at 1731 cm⁻¹, assigned to carbonyl group stretching [32, 33], increased after irradiation, whereas the intensities of the lignin absorption peaks at 1510 cm⁻¹, 1462 cm⁻¹, 1267 cm⁻¹, and 1230 cm⁻¹ decreased after irradiation [34]. These results indicated the generation of chromophoric carbonyl groups and degradation of lignin on the wood surface

after irradiation. As was demonstrated by previous research results, carbonyl and phenoxyl free radicals in the lignin underwent demethoxylation by absorbing the UV light and formed chromophoric groups. As a result, the intensity of the absorbance by lignin at 1510 cm⁻¹ decreased, while the carbonyl group intensity at 1731 cm⁻¹ increased because of lignin photodegradation.

To gain a better insight into the wood photodegradation process after irradiation, the relative peak height ratios were compared between the extracted and nonextracted African padauk specimens. The results are shown in Figure 2. The peaks at 1370 cm⁻¹, 1158 cm⁻¹, and 898 cm⁻¹ were mainly from cellulose and had no remarkable effect on the photodegradation of wood. The 1510 cm⁻¹/1370 cm⁻¹ and 1731 cm⁻¹/1370 cm⁻¹ ratios were calculated for all of the extracted and nonextracted wood specimens.

The 1510 cm⁻¹/1370 cm⁻¹ ratios (absorption by lignin) for the nonextracted and extracted African padauk specimens before irradiation also exhibited some differences. The ratio of the extracted African padauk specimen was 0.98, which was lower than that of the nonextracted specimen (1.23). The reason for this was that the main components of the extractives were phenolic compounds, and thus the ratio decreased when the extractives were removed.

Regardless of whether it was a nonextracted or extracted African padauk specimen, the 1510 cm⁻¹/1370 cm⁻¹ ratio (absorption by lignin) was reduced after irradiation, which indicated that lignin underwent photodegradation during irradiation. Also, the 1510 cm⁻¹/1370 cm⁻¹ ratio of the nonextracted African padauk specimens was higher than that of the extracted specimens. Thus, the presence of extractives accelerated the photodegradation process of lignin when exposed to sunlight.

The 1731 cm⁻¹/1370 cm⁻¹ ratio (carbonyl groups) of the nonextracted and extracted African padauk specimens increased after irradiation. This indicated that the photodegradation of lignin produced carbonyl groups. Furthermore, the carbonyl groups formation rate in the 1731 cm⁻¹/1370 cm⁻¹ ratio of the nonextracted African padauk was higher than that of the extracted specimens. Accordingly, the presence of extractives might have led to the formation of more carbonyl groups after irradiation. This phenomenon was explained by the presence of phenolic extractives. Extractives can absorb light and undergo a photochemical change. Photodegradation of the phenolic compounds in the extractives may also have generated carbonyl groups because of the degradation of lignin.

3.2. Color Change during Exposure. The overall color difference (ΔE^*) of the specimens after exposure to natural sunlight is shown in Figure 3. The specimens S1-SU, S2-SU, and S3-SU were three duplicate African padauk that were exposed to sunlight. The results in Figure 3 revealed that the change trend of the overall ΔE^* was consistent as the irradiation time increased. Rapid discoloration was observed at the beginning of sunlight irradiation and then discoloration slowed down. During the first 20 d of sunlight irradiation, the ΔE^* value of the African padauk exhibited a

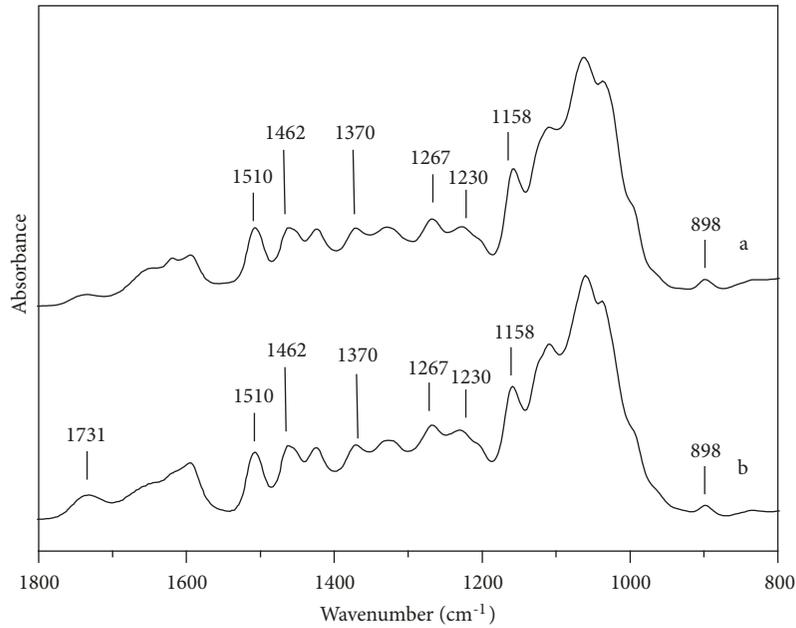


FIGURE 1: FT-IR spectra of the African padauk (a) before and (b) after irradiation.

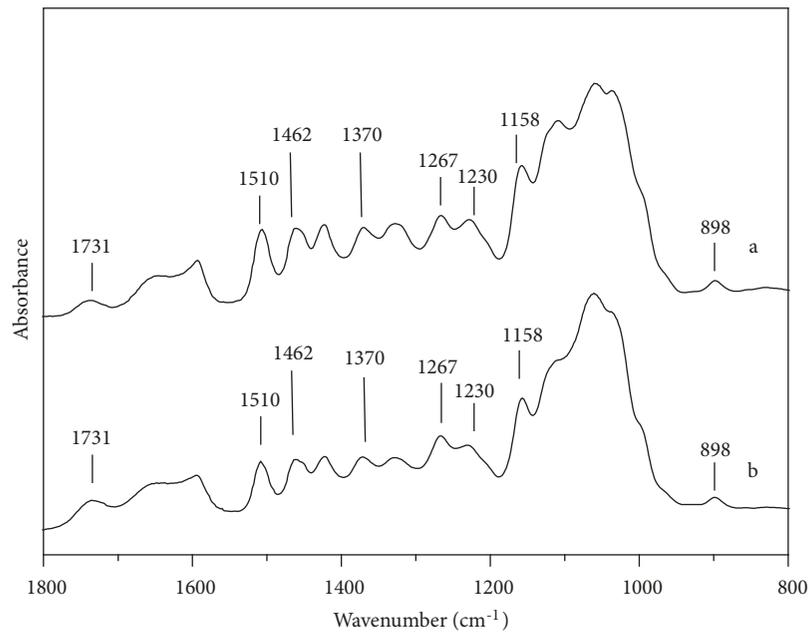


FIGURE 2: FT-IR spectra of the wood powder after extraction of African padauk (a) before and (b) after natural sunlight exposure.

great and remarkable color difference and soon reached 28. With an increased irradiation time, the ΔE^* value increased a little and remained at approximately 30 after 80 d.

Because of uncontrollable factors in natural sunlight, an accelerated lightfastness test was performed. The ΔE^* values of the specimens during the UV radiation process were calculated and are shown in Figure 4. The specimens S1-UV, S2-UV, and S3-UV were three duplicate specimens of African padauk that underwent UV irradiation. With an increase in the irradiation time, the overall ΔE^* values of S1-UV, S2-UV, and S3-UV increased gradually with a similar

trend. During the first 32 h of UV irradiation, the ΔE^* values increased rapidly and soon reached approximately 25. After 32 h of UV irradiation, the value of ΔE^* tended to level off and ultimately remained at approximately 30 after 128 h of UV irradiation. This confirmed that UV radiation is one of the most important environmental factors that cause severe wood surface discoloration.

3.3. Inhibitory Effect of the Red Iron Oxide Coating on Wood Discoloration. The discoloration of the African padauk was mainly caused by UV light degradation. To investigate the

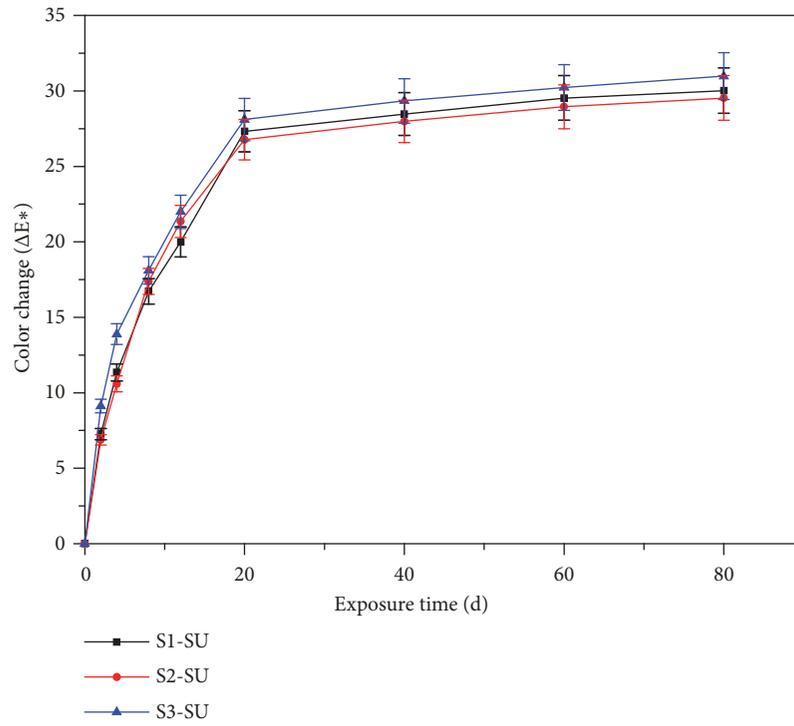


FIGURE 3: Color change of the African padauk after being exposed to natural sunlight.

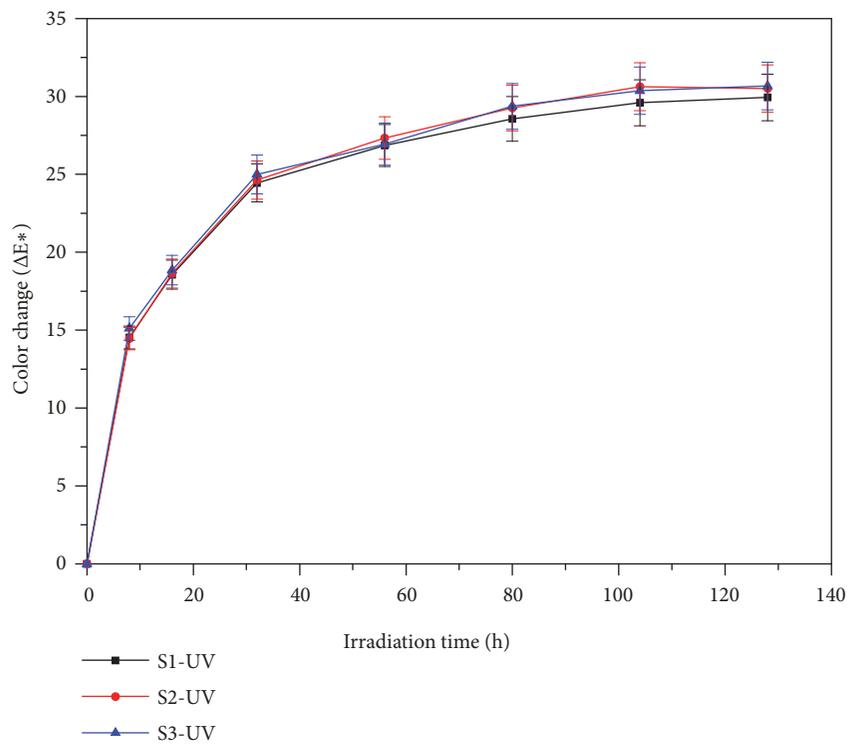


FIGURE 4: Color change of the African padauk after being exposed to UV radiation.

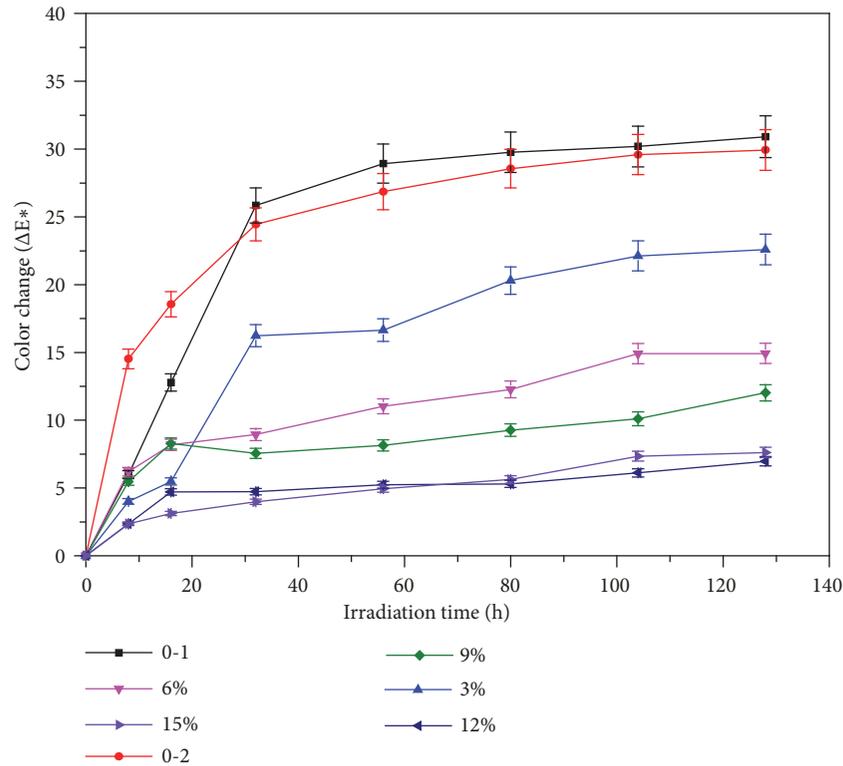


FIGURE 5: ΔE^* changes of 3% to 15% concentration of red iron oxide coated specimens in the UV exposure condition.

influence of the red iron oxide coating on the color change of the African padauk, different red iron oxide concentrations were evaluated. The extensive experiments showed that a concentration of red iron oxide less than 15% had little effect on the initial color change of the African padauk. The results in Figure 5 revealed that all of the different red iron oxide concentrations had varying degrees of protective effect. The 0-1 group represented the untreated African padauk and the 0-2 group represented the African padauk with 0.2 g of megilp. With an increase in the irradiation time, all of the overall ΔE^* values exhibited an increasing trend. In the 0-1 and 0-2 groups, the color change trend was almost the same. The ΔE^* values increased rapidly to approximately 25 during the first 32 h of UV irradiation and then reached approximately 30 after 132 h of UV irradiation, which revealed that the solvent had no effect on preventing surface discoloration of the African padauk caused by UV light. However, as the red iron oxide concentration increased, the ΔE^* values decreased gradually. These results showed that when the red iron oxide concentration was higher, the ΔE^* values were lower. Compared with the 3%, 6%, and 9% red iron oxide concentrations, the ΔE^* appeared to be smaller and more stable with the 12% and 15% concentrations. The ΔE^* values were approximately 7 after 132 h of UV irradiation. Thus, these results showed that red iron oxide has a positive effect on preventing discoloration of African padauk.

To more clearly show the effect of red iron oxide on preventing UV light-induced discoloration, the inhibition ratio parameter was introduced. The results that are presented

in Figure 6 revealed that all of the different red iron oxide concentrations showed some degree of inhibitory effect on discoloration. Specifically, when the red iron oxide concentration was 3%, the inhibition ratio sharply declined and remained at approximately 30%. When the red iron oxide concentration was 6% to 15%, the trend remained consistent. The photochromic inhibition rate remained relatively stable during the first 32 h of exposure to UV radiation and slightly decreased after that point. Moreover, when the red iron oxide concentrations were 12% and 15%, the inhibition ratio was higher than that of the other concentrations and reached approximately 80. These findings demonstrated that the photochromic inhibitory effect was superior when the concentration of red iron oxide was 12% to 15%, which was consistent with the results that are shown in Figure 5.

4. Conclusions

Ultraviolet light is the main cause of wood discoloration. Rapid color changes in the samples took place in the beginning of radiation exposure and the ΔE^* value ultimately reached approximately 30 for both natural and artificial UV light. Discoloration occurred along with fading of the African padauk and degradation of the wood compositions. The FT-IR spectroscopic analysis of the African padauk demonstrated that the changes in the chemical characteristics occurred after irradiation. The photodegradation of the lignin and extractives resulted in the formation of chromophoric carbonyl groups. As the extractives contain large amounts of

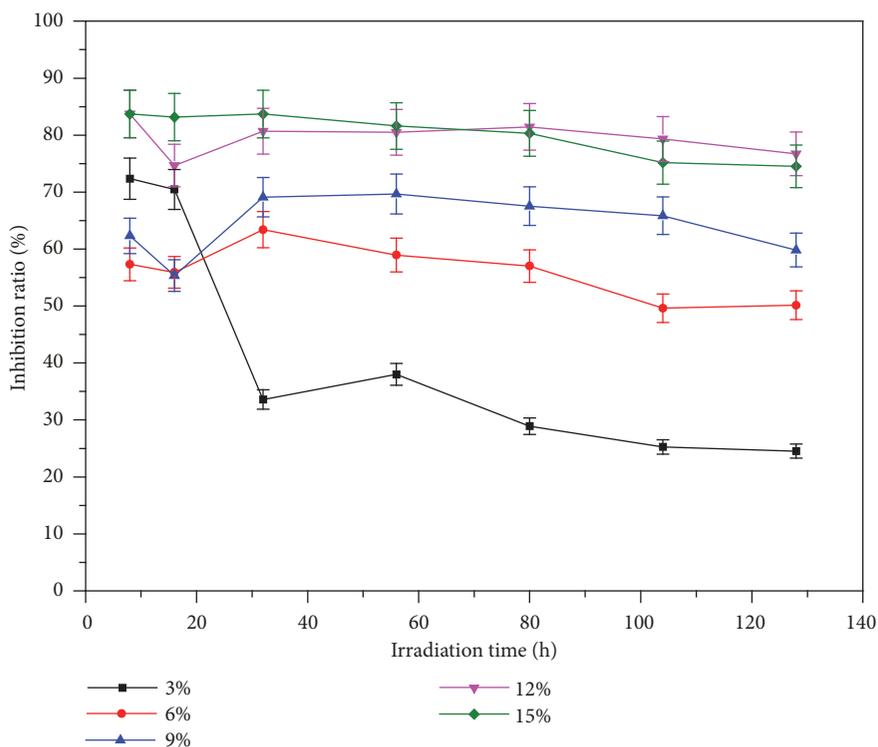


FIGURE 6: Inhibition ratio changes of 3% to 15% concentrations of red iron oxide coated specimens in the UV exposure condition.

phenolic compounds, the presence of extractives accelerated the photodegradation of lignin. The degree of discoloration of the African padauk was greatly influenced by the red iron oxide concentration.

The ΔE^* remained at approximately 7 after 128 h of UV irradiation with the tested red iron oxide concentrations of 12% and 15%. The inhibition ratio in the specimens was also higher with the tested concentrations of 12% and 15% than the concentrations of 3% to 9%. The highest inhibition ratio in the specimens reached approximately 80% when the red iron oxide concentration was 12% and 15%. This was sufficient to prevent discoloration of the African padauk by covering them with a film of red iron oxide and showed an excellent light inhibitory performance.

However, because of the low price and outstanding property of photoprotection, red iron oxide has great potential in wood protection. Until now iron oxide properties have not been fully investigated in conjunction with wood surfaces and this will become a focus of future study.

Data Availability

The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Conflicts of Interest

The authors declare no conflicts of interest.

Authors' Contributions

Chen Wang and Ye Qin performed the experiments, analyzed the data, and wrote the manuscript; Fang Wang and Anmin Huang conceived and designed the experiments; Zhiping Wang contributed to the materials and analysis tools.

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