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Environmental effects on the color of sugi (*Cryptomeria japonica* D. Don) heartwood

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Abstract Sugi (*Cryptomeria japonica* D. Don) is one of the most important planted trees in Taiwan. The normal heartwood of sugi has a red to pinkish rose color. Unfortunately, this pleasant appearance is susceptible to discoloration after environmental exposures. This discoloration is a serious defect that decreases the value of sugi products. The objective of this study was to investigate the effects of environmental factors such as moisture, oxygen, and lights of different wavelengths on the discoloration of sugi. The results show that under the combined effects of light and oxygen the color of heartwood changes from red (or rose-pink) to bluish green. The color further darkens and eventually turns to black with the presence of moisture. As for the effect of light wavelengths on the discoloration of sugi heartwood, it was found that the red color was enhanced after being irradiated with light of wavelengths above 600 nm. Both Δa^* and Δb^* values increased significantly as a result of such exposure.

Key words *Cryptomeria japonica* · Heartwood · Discoloration · Environmental factors · Light wavelengths

Introduction

Color is one of the most distinctive properties of wood. It is of considerable aesthetic importance for wood panels used for decorative purposes. Color also affects human moods in various ways. Red, for example, is a passionate color. The normal heartwood of sugi (*Cryptomeria japonica* D. Don) is red to rose-pink. Among woods, sugi and hinoki have the highest value as building materials in Japanese-style houses. They are also widely used for ceiling board, wall paneling,

and posts.¹ It is well known that the color of some sugi changes from reddish brown to black in a matter of few hours after being logged. There have been many reports discussing this phenomenon,^{2–6} and study of this problem is drawing the attention of many researchers.

Sugi and taiwania (*Taiwania cryptomerioides* Hayata), in the Taxodiaceae class, are two of the most important planted tree species in Taiwan. Taiwania heartwood is yellowish red with distinguished purplish pink streaks; and the heartwood of sugi has a red to pinkish rose color. Both species are fascinating and loved by Taiwanese of traditional tastes. Unfortunately, the color of both heartwoods, after logging, is susceptible to blackening after exposure in natural environment. Hence we were interested in the color changes of both wood lumbers after they were exposed to environmental conditions, such as moisture, oxygen, and light of different wavelengths. In one of our previous study,⁷ it was proven that this pronounced discoloration of taiwania heartwood was mainly induced by light of wavelengths in the range of 350–450 nm. In contrast to the results of this study,⁷ a surprising yet fascinating result was obtained when sugi red heartwood was irradiated with light of different wavelengths. Some of the results of this study have already been reported,⁸ and the complete results are reported herein.

Materials and methods

Thirty-year-old planted sugi (*Cryptomeria japonica* D. Don) from the Experimental Forest of National Taiwan University was used in this study. Blocks of sugi reddish heartwood with dimensions of $2.5 \times 2.5 \times 1.0$ cm³ were prepared from a freshly cut log. All specimens were air-dried in a dark room at constant temperature (20°C) and moisture (relative humidity 65%). Myriad exposure conditions with various environmental factors including light, moisture, and oxygen was used (Table 1). Three specimens having a homogeneous color were used for each exposure condition. The air-dried specimens for “vacuum” were cov-

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Fig. 1. Transmission curves of various light filters

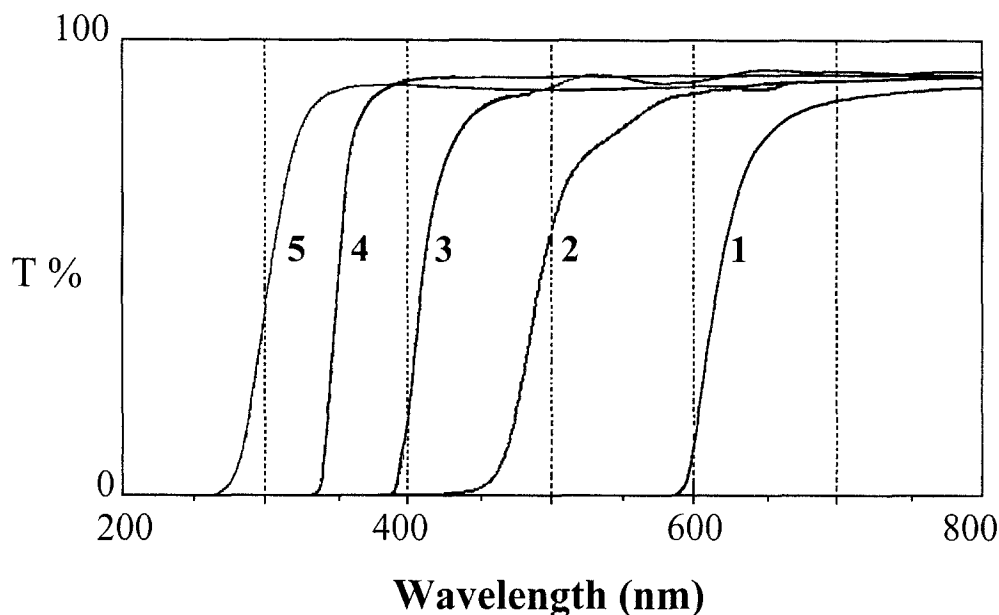


Table 1. Control environmental conditions for the exposure study of sugi heartwood

Abbreviation	Environmental factors		
	Oxygen	Light	Moisture
Vacuum			
Indoor	✓	✓	
Outdoor	✓	✓	✓
W			✓
WO	✓		✓
O	✓		
L		✓	
LO	✓	✓	

W, moisture; O, oxygen; L, light irradiation; WO, moisture + oxygen; LO, light irradiation + oxygen

ered with aluminum foil and kept in the vacuum desiccator. The “outdoor” and “indoor” specimens were air-dried and then exposed on an outdoor weathering rack and to the ambient indoor condition, respectively. The light used for the “indoor” test came from fluorescent lamps. The moisture- plus oxygen-exposed (“WO”) specimens and moisture-exposed (“W”) specimens were saturated with water and then put into a chamber that was purged with oxygen and nitrogen, respectively. Meanwhile, for blocking light irradiation both chambers were covered with aluminum foil. The air-dried oxygen-exposed (“O”) specimens were placed in a dark chamber purged with oxygen. The air-dried light-irradiated specimens (“L”) were placed in a chamber equipped with plant lamps and also purged with nitrogen. The air-dried light-irradiated plus oxygen-exposed specimens (“LO”) were placed in a chamber equipped with plant lamps and also purged with oxygen. To elucidate the effects of light wavelengths on discoloration, five filters with different light transmissions were used to cover the surface of specimens. Figure 1 shows the transmis-

sion curves of the various light filters. With filter 1, for example, only light with wavelengths above 600 nm pass through the filter. Sugi specimens covered with different filters were then exposed to sunlight at ambient conditions. The distance between the specimens and filters is 1 mm.

Color measurements of the specimens were conducted using a color and color difference meter (Dr Lange Co. LMG082). The tristimulus values X , Y , and Z of all specimens were obtained directly from the colorimeter. The recommended CIE (Commission International del'Eclarirange) L^* (psychometric lightness), a^* , and b^* (psychometric chroma coordinates; roughly speaking, a^* is along the X axis red to green, and b^* is along the Y axis yellow to blue) color parameters were then computed to calculate the color difference (ΔE^*) based on the following formula:

$$\Delta L^* = L_t^* - L_o^*, \Delta a^* = a_t^* - a_o^*, \Delta b^* = b_t^* - b_o^*, \\ \Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2}$$

where the subscripts o and t indicate the before (o) and after (t) exposure values.

Results and discussion

Understanding the color variations caused by environmental factors is of considerable importance for woods being used as decorative materials. In our previous studies^{7,9} we proved that the color change in taiwania from yellowish red to bluish green was caused by the combined effects of light and oxygen. The color subsequently darkened and eventually turned to black in the presence of moisture. Sugi also is one of important planted tree species in Taiwan. To understand the discoloration of sugi reddish heartwood, we designed an experiment using several exposure conditions combined with various environmental factors, such as light,

Table 2. Color changes of sugi reddish heartwood after exposure for 48 days in various environmental factors

Parameter	Vacuum	Outdoor	Indoor	W	WO	O	LO	L
ΔE^*	1.6	13.9	5.1	26.3	24.5	1.0	4.8	4.3
ΔL^*	1.2	-2.2	-3.4	-24.2	-22.6	-0.8	-2.7	-2.7
Δa^*	-0.9	-12.1	2.1	5.5	5.8	-0.5	-1.5	-2.3
Δb^*	-0.2	-6.0	1.9	-8.2	-5.2	-0.3	2.2	1.1

moisture, oxygen, and light wavelengths. The results of the experiment are presented below.

Effects of moisture

To study the effects of moisture on the discoloration of reddish heartwood of sugi, we exposed sugi specimens under high moisture conditions, including "outdoor," "WO," and "W." Moisture, oxygen, and light are the three major factors causing the discoloration when specimens were exposed to the "outdoor" exposure condition (Table 1). For "WO" the specimens were exposed to a condition of high moisture and oxygen without light. For the "W" the specimens were exposed to a condition with high moisture without light and purging with nitrogen. Table 2 shows the changes in color of sugi heartwood after 48 days of exposure under various environmental conditions. The results clearly show that moisture caused significant color changes. The ΔE^* values of specimens exposed to light with the presence of moisture were all over 13.9; for "outdoor" the ΔE^* value was 13.9, for "W" it was 26.3, and for "WO" it was 24.5. The ΔE^* values in other conditions without moisture were all below 5.1. The results make it clear that moisture enhances discoloration of sugi heartwood.

Figure 2 shows the effect of moisture on discoloration of sugi heartwood. In the presence of moisture, the ΔL^* values of sugi specimens were clearly decreased, which caused the color of sugi to darken and eventually turned to black. There were, however, no statistically significant differences in Δa^* and Δb^* values when specimens were exposed to high moisture conditions without light ("WO" and "W").

Effects of light and oxygen

It has been proven that light and oxygen are important factors causing photodegradation of wood.¹⁰ In fact, the photodegradation of wood is essentially a photochemical oxidation process. Oxygen is an important element in the promotion of free-radical formation. Peroxide is generated from the interaction of free radicals and oxygen molecules.¹¹

In this study, "O" represents the samples exposed to oxygen atmosphere only (without light or moisture). Its color change was slight after 48 days of exposure, and the ΔE^* value was 1.0 (Table 2). The same results were found in Kondo et al.'s study¹² on the discoloration of tagayasan wood (*Milletia* sp.) and in our previous study on taiwania

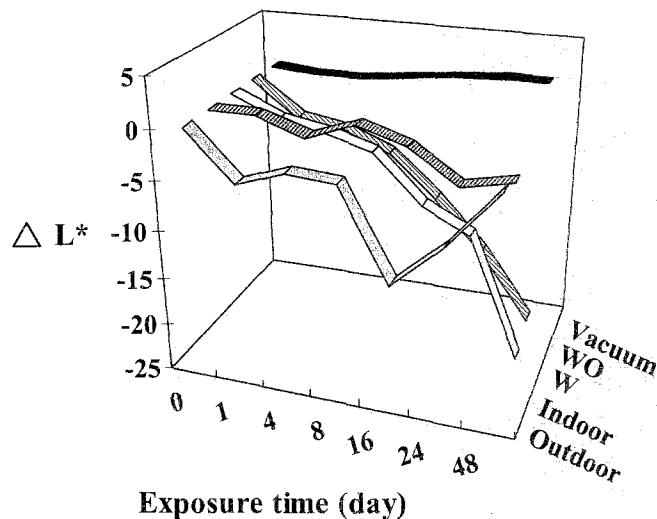


Fig. 2. Effect of moisture on the discoloration of sugi reddish heartwood. □: outdoor; ▨: indoor; ▤: W (moisture); ▩: WO (moisture + oxygen); ■: vacuum

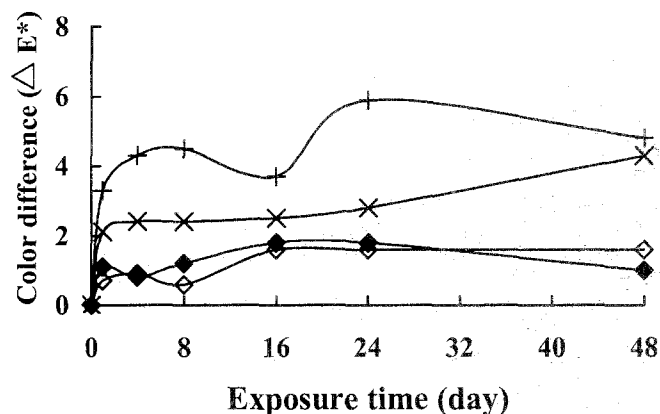


Fig. 3. Effect of light and oxygen on the discoloration of sugi reddish heartwood. ◆: O (oxygen); ×: L (light irradiation); +: LO (light irradiation + oxygen); ◇: vacuum

heartwood.⁷ Figure 3 shows the effect of light and oxygen on the discoloration of sugi heartwood. The "LO" samples (exposed to high oxygen concentration and irradiated with plant lamps) showed the most significant color change (ΔE^* 6.0) after exposure for 24 days. However, the ΔE^* was reduced to 4.8 when the exposure was extended to 48 days. On the other hand, the ΔE^* value of sugi heartwood speci-

mens irradiated with plant lamps at ambient condition ("L") was 2.8 after exposure for 24 days, and it increased to 4.3 after 48 days of exposure. This outcome indicates that the color change is more intense when the specimens are both light and oxygen exposed to up to 24 days. However, the color changes of the "LO" and "L" specimens became equal after an extended exposure. It is believed that the ΔE^* reduction for "LO" after extended exposure is probably due to photo bleaching during the high oxygen concentration condition. Figure 4 shows the effect of light and oxygen on the ΔL^* change of sugi heartwood. The ΔL^* value for "LO" specimens was increased from 24 to 48 days of exposure. The color change during this period also tends to be brighter.

Effects of light wavelengths

Light is an aggregate of photons, the energy of which is wavelength dependent. Light can provide the necessary energy for electron excitation and transition. The photo-discoloration of wood is a chemical phenomenon during which the constituent molecules continuously absorb light energy and convert to colored products. To study the effects of light of different wavelengths on the discoloration of sugi reddish heartwood, five filters were used to truncate different transmission ranges of light. Blocks of sugi reddish heartwood (a^* 15.9–17.6) (Table 3) covered with different filters were exposed to sunlight. To achieve consistent light intensity absorbed by the specimens during irradiation all specimens were irradiated under sunlight from 10:00 a.m. to 2:00 p.m. every day, and the total exposure time was 12 days. Table 3 shows the color parameters of sugi specimens

irradiated for 48 h. It is clear that the degree of discoloration is highest >600 nm (ΔE^* 9.5), followed by above 280 nm (ΔE^* 8.3), above 340 nm (ΔE^* 5.8), above 400 nm (ΔE^* 3.2), and above 450 nm (ΔE^* 2.8). Considering the energy absorbed by the wood specimens, the tendency of color change is reasonable except for the specimen exposed to light with wavelengths above 600 nm. Further examination of the color parameters L^* , a^* , and b^* revealed that all specimens had the same tendency to change color except the one exposed to light of wavelengths above 600 nm. That is, the L^* and a^* values decreased and the b^* value increased or remained the same after 48 h of irradiation. As expected, the discoloration of specimens irradiated with light wavelengths above 450 nm and above 400 nm was slight. However, the discoloration of specimens irradiated with light wavelengths above 600 nm was totally opposite to what we expected: Although it received the lowest energy, the discoloration was the highest. Figure 5 shows the changes in Δa^* and Δb^* of sugi specimens irradiated with light of various wavelengths for 48 h. In contrast to others, sugi specimens changed their color to red after irradiation with light of wavelengths above 600 nm. In addition, the Δa^* and Δb^* of sugi specimens irradiated with light of wavelengths above 600 nm exhibited a significant increase. As a result sugi wood specimens became redder than nonirradiated wood after exposure to light of wavelengths above 600 nm.

As mentioned above, wood with a red color is often more popular and valuable. Thus, from a utilization point of view, red color enhancement of sugi heartwood by light irradiation with wavelengths above 600 nm has a potential to increase the economic value of sugi wood. Further studies on the fastness of this enhanced red color and the mechanisms of the enhancement are in progress. The results will be reported in the near future.

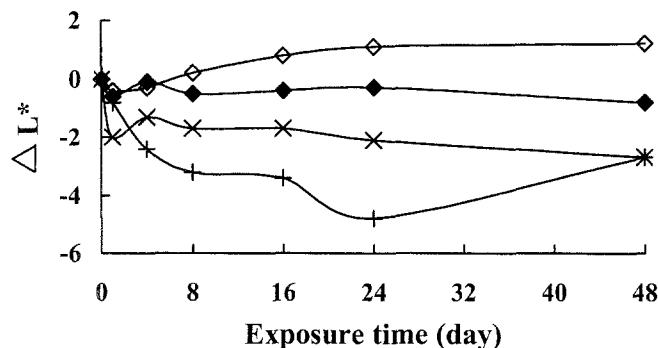


Fig. 4. Effect of light and oxygen on the ΔL^* change of sugi reddish heartwood. \blacklozenge O (oxygen); \times , L (light irradiation); $+$, LO (light irradiation + oxygen); \diamond : vacuum

Conclusions

To understand the environmental factors and light wavelengths that influence the discoloration on sugi reddish heartwood, several exposure conditions combined with various environmental factors, such as light, moisture, oxygen, and light wavelength, was conducted. The experimental results can be summarized as follows.

1. The color of sugi reddish heartwood darkens and eventually turns to black owing to the existence of moisture.

Table 3. Color variation of sugi reddish heartwood irradiated with light of different wavelengths for 48 h

Sample time	Above 600 nm			Above 450 nm			Above 400 nm			Above 340 nm			Above 280 nm		
	L^*	a^*	b^*	L^*	a^*	b^*	L^*	a^*	b^*	L^*	a^*	b^*	L^*	a^*	b^*
Before irradiation	67.6	17.0	19.4	68.8	16.1	20.0	67.7	15.9	19.8	67.0	17.6	20.0	66.8	17.0	20.1
Irradiation for 48 h	63.9	22.8	26.0	67.2	14.1	21.6	67.2	12.7	19.8	64.1	14.8	24.4	63.0	14.6	27.1
ΔE^*		9.5			2.8			3.2			5.8			8.3	

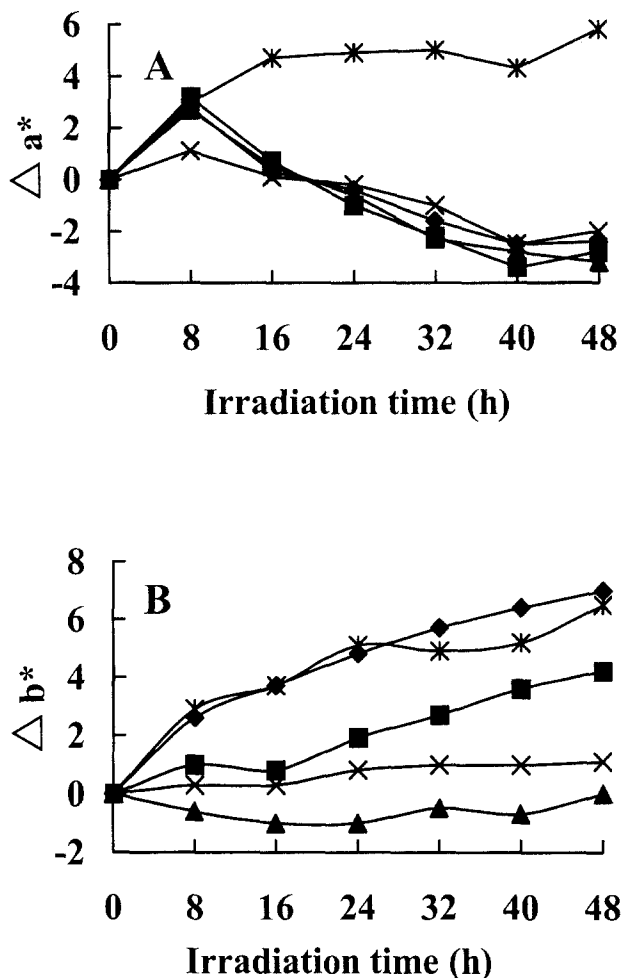


Fig. 5. Changes in Δa^* (A) and Δb^* (B) of sugi reddish heartwood irradiated with light of different wavelengths for 48 h. *, wavelengths above 600 nm; ×, wavelengths above 450 nm; ▲, wavelengths above 400 nm; ■, wavelengths above 340 nm; ◆, wavelengths above 280 nm

- The color of the heartwood changes from red (or rose-pink) to bluish green under the combined effects of light and oxygen.
- The red color of sugi heartwood is enhanced after irradiation with light of wavelengths above 600 nm. The Δa^* and Δb^* of specimens increased significantly when they were irradiated with light of wavelengths above 600 nm.

- The appearances of sugi heartwood specimens were redder than nonirradiated wood after exposure to light of wavelengths above 600 nm.

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References

- Hon DNS, Minemura N (1991) Color and discoloration. In: Hon DNS, Shiraish N (eds) Wood and cellulosic chemistry. Marcel Dekker, New York, pp 395-454
- Abe Z, Oda K, Matsumura J (1994) The color change of sugi (*Cryptomeria japonica* D. Don) heartwood from reddish brown to black. I. The color changes and its causes (in Japanese). Mokuzaï Gakkaishi 40:1119-1125
- Abe Z, Oda K (1994) The color change of sugi (*Cryptomeria japonica* D. Don) heartwood from reddish brown to black. II. Identification of potassium hydrogen carbonate as one of the causative materials (in Japanese). Mokuzaï Gakkaishi 40:1126-1130
- Takahashi K (1996) Relationships between the blackening phenomenon and norlignans of sugi (*Cryptomeria japonica* D. Don) heartwood. I. A case of partially black heartwood (in Japanese). Mokuzaï Gakkaishi 42:998-1005
- Takahashi K (1998) Relationships between the blackening phenomenon and norlignans of sugi (*Cryptomeria japonica* D. Don) heartwood. II. On blackening heartwood containing two main norlignans, sugiresinal and hydroxysugiresinol (in Japanese). Mokuzaï Gakkaishi 44:125-133
- Kubo T, Ataka S (1998) Blackening of sugi (*Cryptomeria japonica* D. Don) heartwood in relation to metal content and moisture content. J Wood Sci 44:137-141
- Wang SY, Chang ST, Su YC (1994) The effect of environmental factors on the discoloration of *Taiwania cryptomerioides* heartwood (in Chinese). For Prod Ind 13:351-361
- Chang ST, Wang SY, Cheng SS (1999) Red color enhancement of sugi (*Cryptomeria japonica* D. Don) heartwood by light irradiation. J Wood Sci 45:271-273
- Chang ST, Wang SY, Su YC, Huang SL, Kuo YH (1999) Chemical constituents and mechanisms of discoloration of taiwania (*Taiwania cryptomerioides* Hayata) heartwood. I. The structure, reconfirmation and conversion mechanism of taiwanin A. Holzforschung 53:142-146
- Hon DNS (1991) Photochemistry of wood. In: Hon DNS, Shiraish N (eds) Wood and cellulosic chemistry. Marcel Dekker, New York, pp 525-555
- Chang ST, Wang SY, Su YC (1993) Mechanisms of photoinduced discoloration of wood extractives and its prevention (in Chinese). Q J Chin For 26(2):113-126
- Kondo R, Mistsunage T, Imamura H (1986) The chemistry of color of wood. II. The effect of environmental factors on the coloration of murasakitagayasan (*Millettia* sp.) heartwood. Mokuzaï Gakkaishi 32:462-466