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Evaluation of the aging wood from historical buildings as compared with the accelerated aging wood and cellulose -Analysis of color properties-

Miyuki MATSUO[1], Misao YOKOYAMA[1], Kenji UMEMURA[1], Junji SUGIYAMA[1], Shuichi KAWAI[1], Joseph GRIL[1,2],

Ken'ichiro YANO[3], Shigeru KUBODERA[4], Takumi MITSUTANI[5], Hiromasa OZAKI[6], Minoru SAKAMOTO[7] and

Mineo IMAMURA[7]

[1]Research Institute for Sustainable Humanosphere, Kyoto University, Japan
[2]Laboratoire de Mécanique et Génie Civil, Université Montpellier 2, France
[3]Tokyo University of the Arts, Japan
[4]The Japanese Association for Conservation of Architectural Monuments, Japan
[5]National Research Institute for Cultural Properties, Nara, Japan
[6]Paleo Labo Co., Ltd., Japan
[7]National Museum of Japanese History, Japan

Abstract

Color changes of wood during natural aging and during heat treatment were elucidated to determine whether they could be explained as the result of a mild thermal oxidation process at ambient temperatures. The results of kinetic analysis employing the time-temperature superposition method showed that the color changes during natural aging were mainly the result of an oxidation process. Color changes of heat-treated cellulose were also compared with that of wood and indicated that cellulose is responsible for the color changes of wood during natural aging and during heat treatment.

1. INTRODUCTION

The deterioration of wood as a material is due to biodegradation, weathering, and aging. In conditions under which biodegradation and weathering can be avoided the service life of wood can exceed thousand years, and aging becomes the major source of wood deterioration. The elucidation of the wood aging mechanism is important not only for the preservation and restoration of wooden historical buildings but also for the purposes of basic wood research. This study aims to elucidate wood aging mechanism by examining changes of wood properties that occur during aging.

Sculptors, carpenters, and restorers of wooden cultural properties and historical buildings have expertise to evaluate the aging of wood. They use the color of wood as an important aging criterion. This may be because the color properties changes directly express chemical changes that occur in wood during aging. In this study we elucidate wood aging mechanism by using color properties.

Though some empirical data had suggested that wood aging is a mild thermal oxidation at room temperature, few papers had evaluated it theoretically or reported on its detailed mechanisms. Our previous study successfully employed kinetic analysis to color value L^* and color difference ΔE^*_{ab} to theoretically predict and elucidate natural aging by comparing it to the accelerated aging [1]. The present study reports on the inclusive analysis of color values expressed by the CIELAB color system.

Furthermore, we also measured the color properties of heat-treated cellulose. Several wood components such as cellulose, lignin, and extractive are responsible for wood color changes. This paper examines the contribution of color changes in cellulose to the changes in wood.

2. MATERIALS AND METHODS

Hinoki (*Chamaecyparis obtusa* Endl.), the typical wood for traditional buildings and Buddhist sculptures in Japan, was chosen as wood species. All samples were carefully selected so that they had homogeneous grain and so that they were free from bio-degradation, weathering and stain and defect.

Natural aging wood: Eight natural aging wood samples collected from historical buildings were used. The wood formation year, ranging from A.D. 434 to A.D. 1438, were determined by dendrochronology and radiocarbon dating which agreed well with each other and agreed well with corresponding historical documents. Aging heartwood specimens were cut out with the dimensions of 60mm (L) x 10mm (R) x 2mm (T). Yokoyama et al. [2] reported in detail on these samples.

Heat-treated wood: 360-years-old wood from Kiso, Japan was used. The tree was harvested in 1988 and the wood had been dried for 18 years in ambient condition in shed. Specimens were cut out with the dimensions of 120 mm (L) x 20 mm (R) x 4 mm (T) from near the outermost part of the heartwood and then dried in an air-circulating oven and in a vacuum oven at 60 °C and for 24 hours for each. Dried specimens were heated in an air-circulating oven at 4 temperatures levels ranging from 90 to 180 °C for a duration ranging from 0.5 hour to approximately 2 years.

Heat-treated natural aging wood: Specimens aged for 921 years, cut from one of natural aging samples, were heated with treatment temperature and time at 3 temperatures levels ranging from 120 to 180 °C for a duration ranging from 4 hour to approximately 110 days.

Cellulose: Specimens of cellulose filter paper made from cotton (Whatman No.1) were heated at 180 °C for a duration ranging from 1 to 120 hours.

The color properties of the specimens were measured with a spectrophotometer (KONICA MINOLTA CM-2600d) and was expressed using the CIELAB color parameters (L^*, a^*, b^*). The total color differences ΔE^*_{ab} were calculated using the reference color parameters obtained as the average of 83 untreated specimens.

3. RESULTS AND DISCUSSION

3.1. Color Changes in the Specimens

Figure 1 shows color changes that occur during aging and heat treatment for each sample. The specimens became darker with aging time and treatment time.
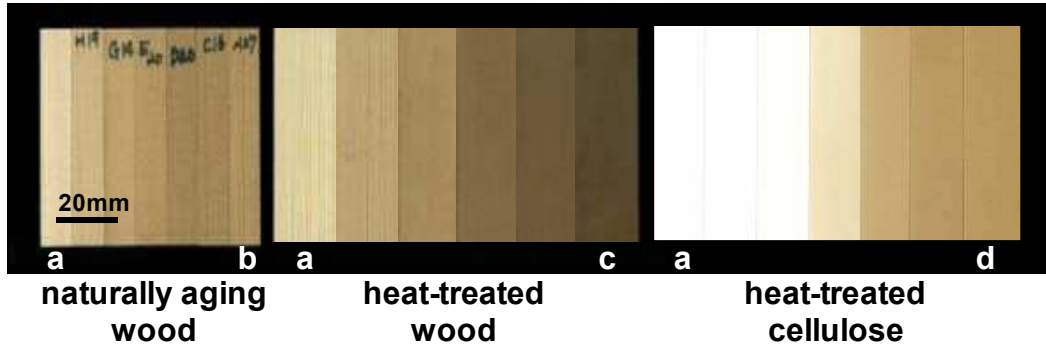
Fig. 1 Color changes of naturally aging wood, heat-treated wood, and heat-treated cellulose (a: control (untreated wood or untreated cellulose), b: wood aged for 1573 years, c and d: wood and cellulose treated at 180°C for 120 hours, respectively).

3.2. Kinetic Analysis for Wood Specimens

Figure 2 shows ΔE^*ab value change of naturally aging wood and heat-treated wood. Changes of the color value that occur during aging were similar to changes that occur during heat treatment. The same behavior was observed in other color values, L^*, a^*, and b^*. This suggested that heat treatment could accelerate color changes that occur during aging.
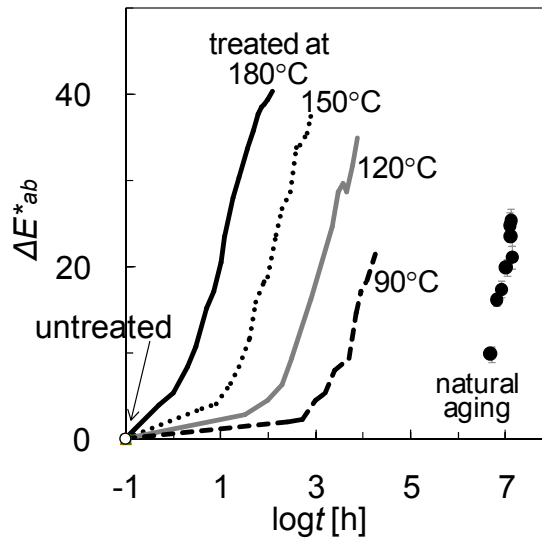

Fig. 2 Changes in ΔE^*ab value during heat treatment and natural aging. (t: treatment time or aging time)

Kinetic analysis using the time-temperature shift factor was adopted to predict the color change during natural aging. Arrhenius equation and the shift factor (a_T) are given by

$$k = A \exp\left(-\frac{E_a}{RT}\right) \tag{1}$$

and

$$a_T = t_T / t_{ref} \tag{2}$$

where k is the rate constant of the chemical reactions, A the frequency factor, E_a the apparent activation energy, R the gas constant, T the absolute test temperature, t_{ref} is the test time at a reference temperature T_{ref}, and t_T is the time required to give the same response at the test temperature T. Combining Eqs. (1) and (2) gives

$$a_T = \exp\left[\frac{E_a}{R}\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right] \tag{3}$$

which allows to use the whole data set observed during accelerated aging treatment and natural aging [3][4][5][6]. Plotting $\ln(a_T)$ vs. $1/T$ is a way to calculate E_a and to predict changes in properties under ambient conditions. E_a was calculated by using a_T of (i)heat-treated wood, (ii)naturally aging wood and heat-treated wood, and (iii)heat-treated naturally aging wood. Table 1 shows the values of the apparent activation energies for each color values.

The color change in the ambient condition was predicted by using E_a of heat-treated wood and compared with the change measured in naturally aging wood. That is, the Arrhenius plots were extrapolated to the temperature of 15 $^{\circ}$C, the annual mean air temperature in Nara, Japan. Then the a_T (T=15 $^{\circ}$C) calculated from the extrapolation was compared with the a_T (T=15 $^{\circ}$C) determined from empirical data. The a_T of the natural-aging data was positioned near the regression line for the heat-treatment data on the Arrhenius plot. The regression lines for the plots including both heat treatment and natural aging showed good linearity ($R^2 > 0.98$). These results indicated that the color change during natural aging was, as with the heat treatment, mainly due to thermal oxidation. E_a of naturally aging wood and heat-treated wood lower than that of heat-treated wood meant that the natural color change was somewhat faster than the changes due to only thermal oxidation. This result suggests that, the color change could be accelerated by other factors in addition to thermal oxidation, such as moisture in the air, the repetition of drying and wetting, and the repetition of hot and cold periods. Although E_a of heat-treated naturally aging wood were somewhat higher than the others, those of L^* and ΔE^*_{ab} were within the reported values [7][8]. The higher energies of a^* and b^* means that they change more slowly than L^* and ΔE^*_{ab}. However, the range of these data was not sufficient to accurately evaluate them.

Table 1 Apparent activation energies calculated from a_T of (i)heat-treated wood, (ii)naturally aging wood and heat-treated wood, and (iii)heat-treated naturally aging wood, for each color values.

	E_a [kJ/mol]		
	(i)	(ii)	(iii)
L^*	117	90	135
a^*	95	84	154
b^*	114	99	153
ΔE^*_{ab}	113	90	133

3.3. Color changes of heat-treated cellulose

Figure 3 and 4 compare changes in color values of heat-treated wood with that of heat-treated cellulose. Cellulose color changes exhibit the same behavior with that of wood in L^* and ΔE^*_{ab} values, while the changes were somewhat different in a^* and b^* values. This result indicates that cellulose is one of major components that contribute wood color changes during aging and heat treatment, although other components also have effects on color changes that are expressed by a^* and b^*.
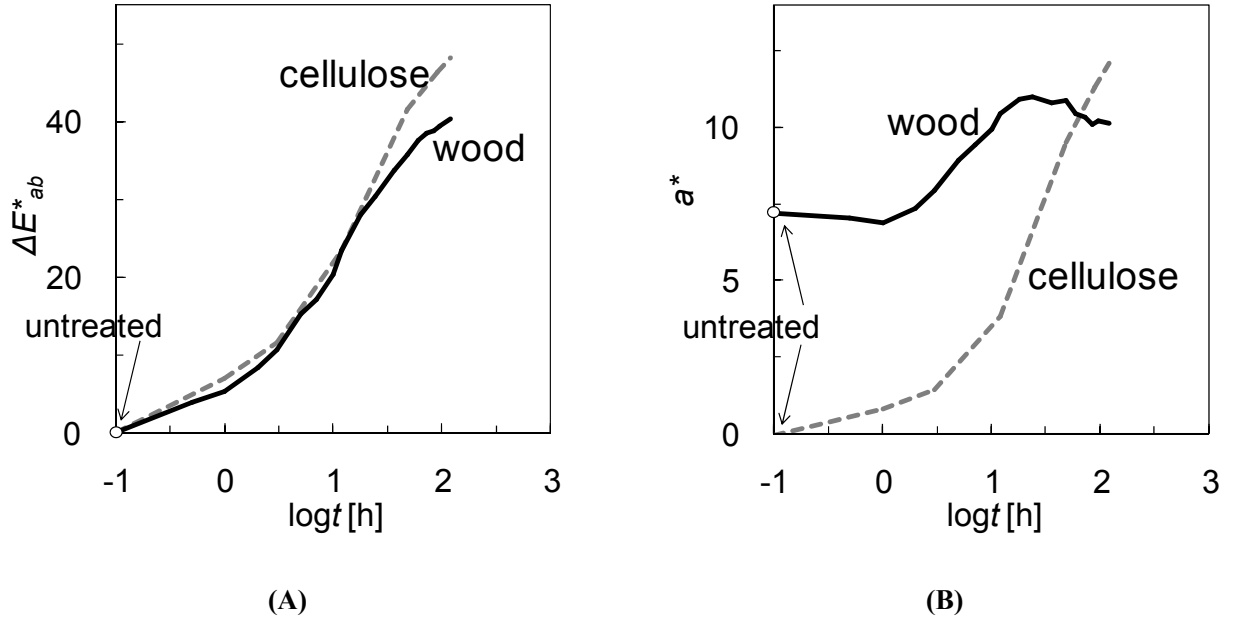

(A) **(B)**

Fig. 3 Changes of **(A)** ΔE^*_{ab} and **(B)** a^* value in wood and cellulose that occur during heat treatment at 180 °C.

4. CONCLUSIONS

Wood color changes that occur during aging show the same behavior with changes that occur during heat treatment in all measured color values. The values of L^* decreased and a^*, b^*, and ΔE^*_{ab} increased with the aging or treatment time. The kinetic analysis indicated that the color changes during natural aging were mainly explained as a result of mild thermal oxidation. This conclusion was also supported by analyzing the color changes of heat-treated natural aging wood. Color changes of heat-treated cellulose suggested that cellulose is one of wood components that are mainly responsible to color changes of heat-treated and naturally aging wood.

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