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ORIGINAL ARTICLE

Kinetic analysis of color changes in keyaki (*Zelkova serrata*) and sugi (*Cryptomeria japonica*) wood during heat treatment

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Abstract The kinetics of color changes in keyaki (*Zelkova serrata* Makino) and sugi (*Cryptomeria japonica* D. Don) wood during heat treatment were examined. The color of wood specimens treated at 90, 120, 150, and 180 °C was measured by an imaging spectrophotometer and expressed using CIELAB color parameters. At any treatment temperature, values for L^* and ΔE^*_{ab} decreased and increased in both wood species, respectively, with increased treatment time. Changes in a^* and b^* varied depending on wood species and treatment temperature. The color changes were successfully analyzed using the kinetic approach applying time–temperature superposition method. This approach elucidated and accurately predicted color changes during heat treatment.

Keywords Heat treatment · Color · Kinetic analysis · Keyaki (*Zelkova serrata* Makino) · Sugi (*Cryptomeria japonica* D. Don)

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Introduction

Heat treatment of wood, particularly at temperatures below the thermal decomposition point of each wood component, has been widely studied for various purposes, including wood modification, wood drying, and accelerated aging [1, 2]. Among the various properties of wood, color change during heat treatment has been frequently examined to develop chemical-free coloration methods [3] or to prevent staining and discoloration of wood during heating [4, 5] because color is esthetically important and affects the commercial value of wood. Many previous studies examined the effects of factors affected by heating processes such as temperature, duration, and moisture. However, individual phenomena in specific treatment conditions were described; no comprehensive study has been conducted to examine the color change behavior of wood during heat treatment. Color has also been studied as a nondestructive criterion for monitoring changes in other wood properties of wood. Previous studies investigated treatment duration and temperature within a limited range and for only a few wood species. Thus, the potential of color as a criterion remains to be determined. Some researchers found clear relationships between color and changes in other properties such as weight and mechanical properties during heat treatment [6–8], whereas others asserted that color is not suitable as a criterion for wood evaluation purposes, or that assessment techniques must be improved because results obtained in previous studies were unclear or occasionally contradictory [9–11].

In our previous studies [12, 13], color changes in hinoki wood (*Chamaecyparis obtusa* Endl.) and cellulose filter paper as a major component of wood were successfully analyzed using a kinetic approach, which allowed precise prediction of color changes at any temperature and time within the experimental ranges. Furthermore, the obtained

results of hinoki wood were expanded to elucidate color changes during natural aging [14], leading to the conclusion that a uniform reaction (thermal oxidation) causes color changes during both natural aging and heat treatment.

In the present study, color changes in keyaki (*Zelkova serrata* Makino) and sugi (*Cryptomeria japonica* D. Don) were comprehensively investigated. These typical examples of hardwood and softwood, respectively, are widely used in wooden buildings in Japan, such as hinoki wood. Specimens were heat-treated at various treatment durations and temperatures. A kinetic approach was used to analyze the temperature dependency of color changes in these two species. The results were then compared with those of hinoki wood and cellulose filter paper to elucidate changes in the color of wood during heat treatment.

Materials and methods

Samples

A 100-year-old keyaki and a 180-year-old sugi tree were used. The trees were harvested in Japan and the wood was dried for more than 7 years under ambient room temperature conditions. A sample block of homogenous grain for each species was carefully selected from the area near the outermost part of the heartwood. The size of specimens was 60 mm (longitudinal direction) × 10 mm (radial direction) × 2 mm (tangential direction). Air-dried density and annual ring width of each species are summarized in Table 1. The specimens were dried in an air-circulating oven and a vacuum oven at 60 °C for 24 h each. Then, the specimens were heated in an air-circulating oven at 90, 120, 150, and 180 °C for durations ranging from 10 min to approximately 1.4 years (Table 2). Heating times were selected on the assumption that a 10 °C increase in reaction temperature halves the reaction rate, to obtain a similar degree of reaction intensity despite the differences in temperature [15].

Color measurement

Despite careful selection of samples, the density varied more widely and the annual ring width were larger than those of hinoki wood used in our previous study [12]. To reduce within-specimen variations in color caused by less homogeneity in tissue structure, specimen color was measured with a spectroscopic imaging system. The system consisted of an imaging spectrophotometer (ImSpector V8E, JFE Techno-Research Corporation, Tokyo, Japan), a monochrome camera with a charge-coupled device, xenon light source, stepper table, and controlling computer. This system provided scanned digital images of all specimens. Each pixel in the images contains spectroscopic intensity data, which can be converted to various color parameters using spectrum-analyzing software (ImSpector Spectrum Analyser, JFE Techno-Research Corporation, Tokyo, Japan). An area of 50 mm (longitudinal direction) × 8 mm (radial direction) in each specimen was scanned at a scanning resolution of approximately 0.19 mm/pixel (134 dpi). The color value of each specimen was calculated as the average of all pixels in the measured area, which reduced the variations in color.

The CIELAB color parameters (L^*, a^*, and b^*) were used to express color changes. The differences in parameters ΔL^*, Δa^*, and Δb^* and the total color differences ΔE^*_{ab} were calculated using the following formulae [16, 17]:

$$\begin{aligned} \Delta L^* &= L^* - L_0^* \\ \Delta a^* &= a^* - a_0^* \\ \Delta b^* &= b^* - b_0^* \end{aligned} \quad (1)$$

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

Table 1 The average air-dried density and annual ring width with their standard deviations in parentheses

Sample	Air dried density (g/cm^3)	Annual ring width (mm)	n
Keyaki wood	0.66 (0.02)	4.40 (1.35)	255
Sugi wood	0.36 (0.02)	1.50 (0.31)	243

n is the number of specimens used to calculate the average value

Table 2 Treatment temperature and time for keyaki and sugi wood

	90 °C	120 °C	150 °C	180 °C
Treatment time (h)	12	1	40 min	
		3		
	85.3	10.7	1.33	10 min
	171	21.3	2.67	20 min
	256	32	4	30 min
	384	48	6	
	524	64	8	1
				1.5
	1024	124	16	2
	3072	384	48	6
	6144	768	96	12
	9266			
	12288	1536	192	24
		3136	384	48
		5440		84
		7680	960	120

Table 3 The average initial color values L_0^*, a_0^*, and b_0^* with their standard deviations in parentheses

Sample	L_0^*	a_0^*	b_0^*	n
Keyaki wood	59.69 (1.78)	11.79 (0.88)	31.96 (0.98)	155
Sugi wood	56.25 (2.43)	11.63 (1.02)	21.27 (1.03)	90

n is the number of specimens used to calculate the average value

where L^* is the lightness and a^* and b^* are the color coordinates under any treatment condition, respectively, and L_0^*, a_0^*, and b_0^* are the corresponding initial values obtained as the average of untreated specimens. The values of L_0^*, a_0^*, and b_0^* are summarized in Table 3. Four to five specimens were tested under each treatment condition and their average values and standard deviations calculated.

Kinetic analysis

A chemical reaction in a uniform phase can be described by the Arrhenius equation:

$$k = A\exp\left(-\frac{E_a}{RT}\right) \tag{3}$$

where k is the reaction rate constant, A is the frequency factor, E_a is the apparent activation energy, R is the gas constant, and T is the absolute temperature at which the reaction occurs. In general, the temperature dependence of the reaction rate is expressed as E_a. When changes in material properties were measured at different T, the values of k were obtained from their reaction rates. By plotting the logarithm of k versus the reciprocal of T, the values of E_a can be obtained from the slope of the regression line for the plots, which is called the Arrhenius plot [18]. Interpolating or extrapolating the regression line of the Arrhenius plot allows us to determine the reaction rate at any given temperature, which is useful not only in understanding the reaction, but also in predicting changes in the property under any conditions.

To determine the activation energy more accurately, a time–temperature superposition (TTSP) method has frequently been used for polymeric materials [19–22]. An Arrhenius approach applying the TTSP method is briefly described as follows. When changes in material properties at different treatment temperatures are plotted as functions of logarithmic treatment time and the plotted curves show a similar shape irrespective of treatment temperature, the plotted curves can be superposed to a single curve at a fixed reference temperature T_{ref} along the logarithmic time axis. This shift distance used to superpose the curves is called the time–temperature shift factor a_T and is given as follows:

$$a_T = \frac{t_T}{t_{ref}} \tag{4}$$

where t_{ref} is the treatment time at T_{ref}, and t_T is the time required to obtain the same response at treatment temperature T. Combining Eqs. (3) and (4) thus gives:

$$a_T = \exp\left[\frac{E_a}{R}\left(\frac{1}{T}-\frac{1}{T_{ref}}\right)\right] \tag{5}$$

where both T and T_{ref} are absolute temperatures. Plotting the logarithm of a_T versus the reciprocal of T is another way to calculate E_a. Ding et al. [23] compared the TTSP method with the conventional method in Eq. (3) by analyzing several sets of data for cellulose degradation from literature. They concluded that the TTSP method is more accurate than the conventional one. In our previous study, the color changes of hinoki wood [12] and cellulose filter paper [13] were successfully analyzed using a combination of kinetic analysis and TTSP.

Though the curves plotted as a function of logarithmic treatment time are similar in shape, these curves cannot always be completely superposed, namely, a single smooth curve cannot be obtained only by a horizontal shift of data along the logarithmic time axis. In such cases, in addition to a horizontal shift, the data set may be shifted vertically along the logarithmic property axis to obtain a smooth superposed curve. This shift distance is called vertical shift factor b_T. Vertical shift has been used to explain the influences of temperature on the viscoelastic behaviors of thermorheologically complex materials [24–26]. Though application of b_T in the context of color changes during heat treatment has not been attempted, the TTSP method was modified in the present study using b_T formulated as follows:

$$b_T = \frac{P_T}{P_{ref}} \tag{6}$$

where b_T is the vertical shift factor at treatment temperature T, P_T the property at T, and P_{ref} the property at T_{ref}.

To calculate a_T and b_T, a temperature of 180 °C was chosen as T_{ref}. Nonlinear regressions were performed to find the best models that fitted well to changes in color parameters at 180 °C, expressed as a function of t.

All parameters of the model functions and kinetics were estimated using a nonlinear iterative curve-fitting method. The coefficient of determination (R^2) and the root mean-square error (RMSE) were used as criteria for the fitting of the tested models to the experimental data:

$$R^2 = \frac{\sum_{i=1}^{N}(\hat{Y}_i - \bar{Y})^2}{\sum_{i=1}^{N}(Y_i - \bar{Y}_i)^2} \tag{7}$$

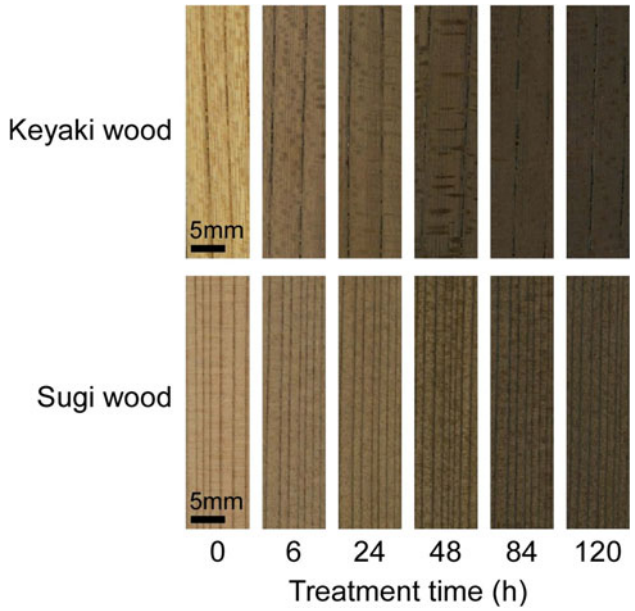

Fig. 1 Color changes of specimens treated at 180 °C in order of increasing treatment time

$$\mathrm{RSME} = \sqrt{\frac{\sum_{i=1}^{N}(Y_i - \hat{Y}_i)}{N}}, \quad (8)$$

where $\hat{Y}_i$ and Y_i are the measured and modeled values, respectively, $\bar{Y}$ is the average of the measured data, and N is the total number of observations. The higher the value of R^2 and the lower the value of RSME, the better did the model fit the data. The FlexPro 8 software package (Weisang GmbH, Germany) was used for all data analysis.

Results and discussion

Color changes during heat treatment

Figure 1 shows the images of representative specimens. The specimens turned darker and browner with increased treatment time. The color changed uniformly from the outside to the middle of the specimens. Figures 2 and 3 show changes in L^*, a^*, b^*, and ΔE^*_{ab} of keyaki and sugi

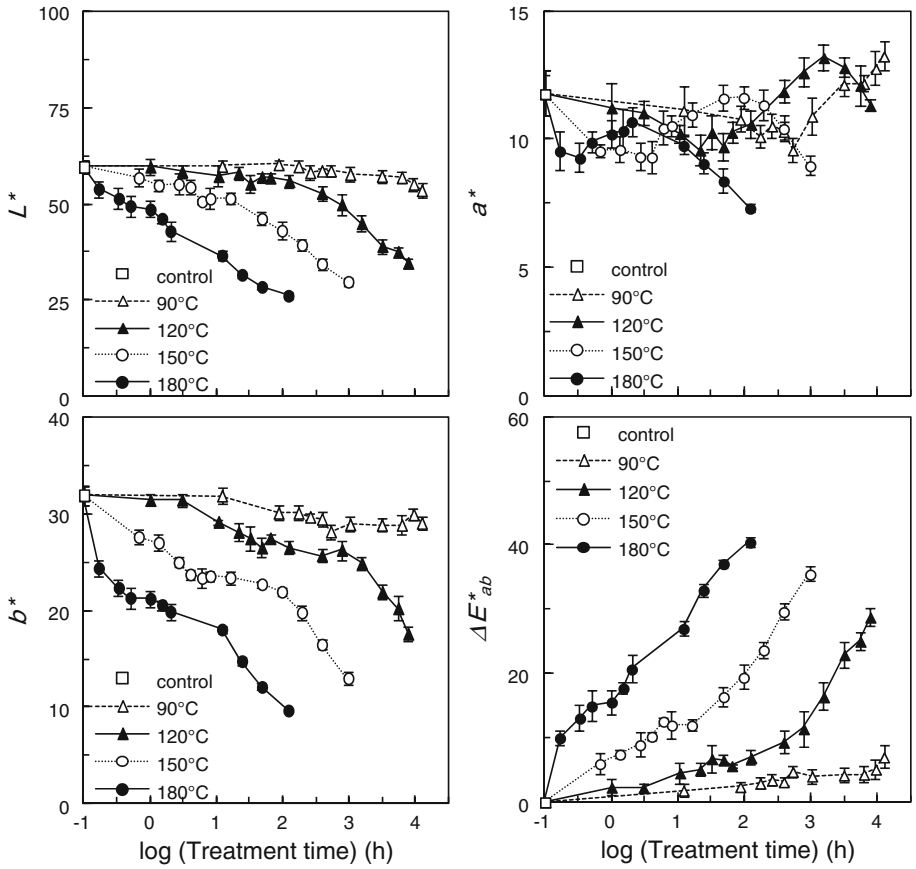

Fig. 2 Changes in L^*, a^*, b^*, and ΔE^*_{ab} of keyaki wood during heat treatment as a function of time and temperature (*error bar* standard deviation)

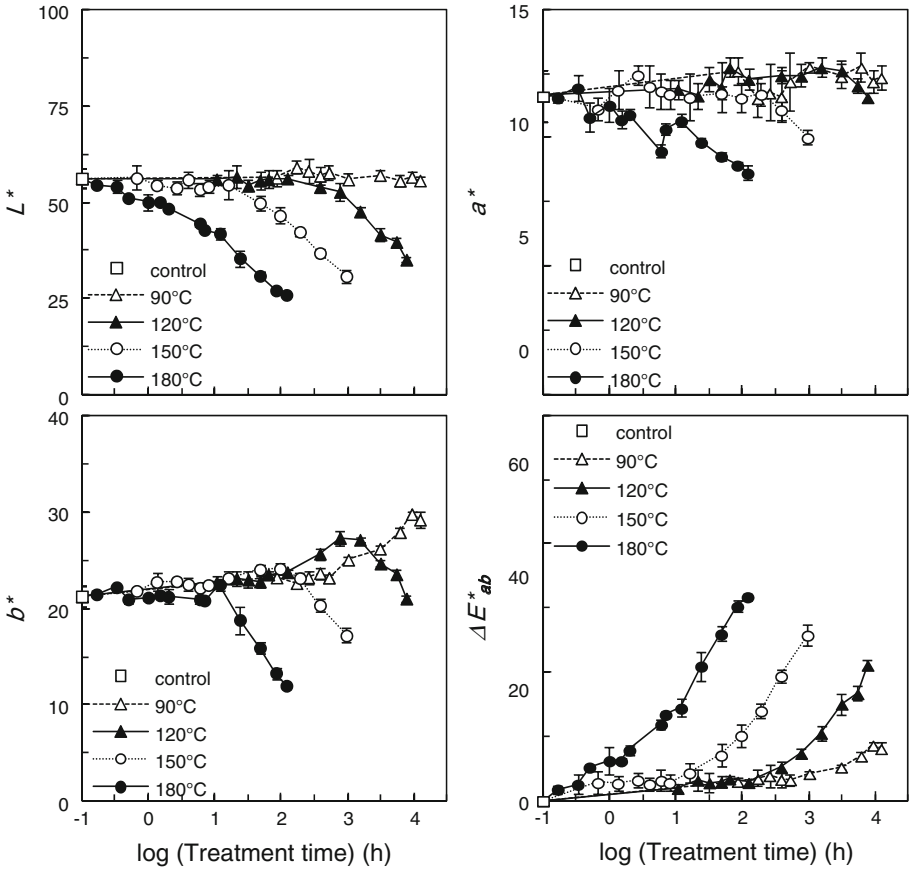

Fig. 3 Changes in L^*, a^*, b^*, and ΔE^*_{ab} of sugi wood during heat treatment as a function of time and temperature (*error bar* standard deviation)

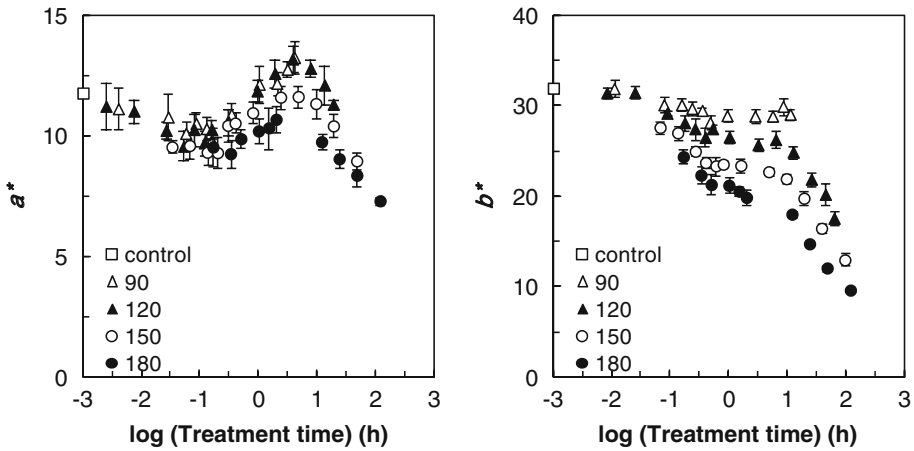

Fig. 4 Changes in a^* and b^* of keyaki wood superposed using only horizontal shift factor a_T (*error bar* standard deviation)

wood, respectively, according to the treatment time and temperature. The values of L^* decreased in both keyaki and sugi wood, indicating that they became darker. This result corresponded to the increase in ΔE^*_{ab}, which was largely dominated by decreasing L^*. Decreases in L^* and increases in ΔE^*_{ab} during heat treatment have been well documented in various wood species [1–12]. In addition, changes in a^* and b^* differed depending on the wood species. In keyaki wood, the value of a^* initially decreased, then increased to a peak and decreased again, whereas the value of b^* decreased constantly with a shoulder. In sugi wood, the value of a^* decreased slightly, whereas the value of b^* initially increased and then decreased. This increase and subsequent decrease in a^* and b^* have been reported for

some other wood species such as beech, spruce, and pine [6, 7, 27], whereas a monotonous decrease in a^* and b^* has been reported for pine and eucalypt wood [3]. No significant change was previously reported in an analysis of sugi wood [28]. This inconsistency may be explained by the fact that treatment duration was insufficient in these reports.

Comparing the results among treatment temperatures in each species, changes in L^* and ΔE^*_{ab} showed similar behavior irrespective of treatment temperature, whereas changes in a^* and b^* were temperature dependent. As shown in Figs. 2 and 3, the lower the treatment temperature, the higher the peak values of a^* for keyaki and b^* for sugi, indicating that treatment at lower temperatures enhanced the increase in these chromatic parameters. Furthermore, the lower the treatment temperature, the smaller the decreases in b^* of keyaki and a^* of sugi wood, indicating that heat treatment at lower temperatures moderated the reduction in these chromatic parameters. No such temperature dependencies were observed in hinoki wood and cellulose filter paper [12, 13]. Thus, the results of our studies on heat treatment in several wood species at a wide range of durations and temperatures revealed that color changes in complex ways depending on the duration of heat treatment, temperature, and wood species. The results would contribute to the simple assessment of wood quality and understanding the empirical knowledge about quality changes. Almost all the main components of wood, i.e., cellulose, hemicelluloses, lignin, extractives, and ash contents are related to the color of wood and its changes [29]. Our previous study showed that cellulose is a major component that contributes to color changes of hinoki

Table 4 Coefficients of determination (R^2) and root mean-square errors (RMSE) of regression curves using Eq. (9) for color changes of keyaki and sugi wood at 180 °C

	Keyaki wood		Sugi wood	
	R^2	RMSE	R^2	RMSE
L^*	0.9938	0.294	0.9962	0.634
a^*	0.9957	0.721	0.9142	0.307
b^*	0.9970	0.335	0.9740	0.542
ΔE^*_{ab}	0.9964	0.743	0.9967	0.614

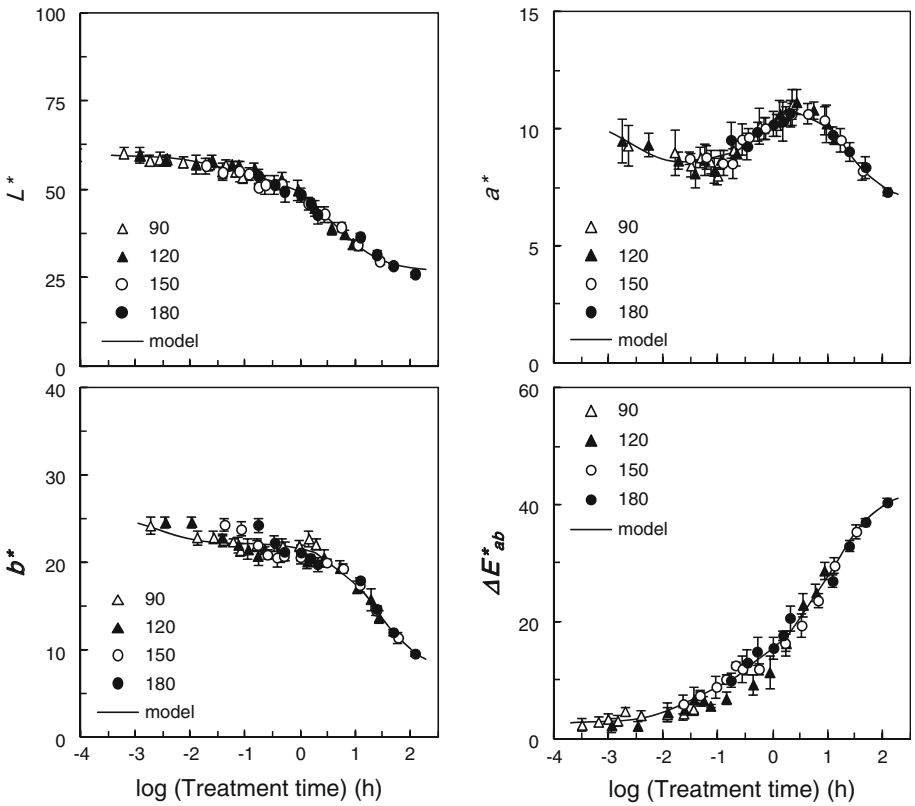

Fig. 5 Changes in color parameters of keyaki wood: best-fit model [Eq. (9)] and superposed data using only horizontal shift factor a_T for L^* and ΔE^*_{ab} and using both a_T and vertical shift factor b_T for a^* and b^* (*error bar* standard deviation)

wood, while other components also have effects on color changes [13]. Extractives of each wood species also make the characteristic color [30–32] mainly expressed by chromatic parameters, a^* and b^*. The different behavior of different color parameters would be caused by chemical changes of each component. However, CIELAB color parameters do not necessarily reflect a particular chemical change in wood. To refer to chemical changes, qualitative and quantitative analysis of different wavelength spectra and chemical components will be needed.

Kinetic analysis

In keyaki and sugi wood, changes in L^* and ΔE^*_{ab} were analyzed using the TTSP method and by applying only the horizontal shift factor a_T. As shown in representative data for a^* and b^* in Fig. 4, single smooth superposed curves were not obtained for a^* and b^* by shifting only along the logarithmic time axis, though overlap of the inflection points of each curve was observed. This implies that application of the shift factor, a_T, only was insufficient for accurate prediction of color changes in keyaki and sugi wood and that temperature dependencies should also be considered. Therefore, the vertical shift factor b_T was applied in addition to a_T to analyze the changes in a^* and b^*.

Based on the values of R^2 and RMSE, the following was selected as the best-fit function:

$$P = \frac{C_0 + C_1 t + C_2 t^2 + C_3 t^3}{1 + C_4 t + C_5 t^2 + C_6 t^3}, \tag{9}$$

where C_i ($i = 0, \ldots, 6$) is a constant. Note that this function was applied for numerical analysis of color changes; it may have no meaning in terms of the reaction mechanism.

Table 4 summarizes the values of R^2 and RMSE for Eq. (9) fitted to the data set of color changes at 180 °C, indicating that Eq. (9) fitted well to the data set. Figures 5 and 6 show the well-superposed data with the regression curves of keyaki and sugi wood, respectively, using only a_T for L^* and ΔE^*_{ab} and both a_T and b_T for a^* and b^*. Figure 7 shows the Arrhenius plots for L^* of keyaki wood as an example. The regression lines of the Arrhenius plots for all color parameters showed good linearity ($R^2 > 0.980$), suggesting that color changes at 90–180 °C were caused by an apparently similar reaction. The apparent activation energies (E_a) calculated from each color parameter were higher than those of hinoki wood and cellulose filter paper [12, 13], particularly for L^* and ΔE^*_{ab} (Table 5). This result indicated that color change reactions of keyaki and sugi wood apparently have higher energy barriers than those of hinoki wood and cellulose. Considering sugi and hinoki are

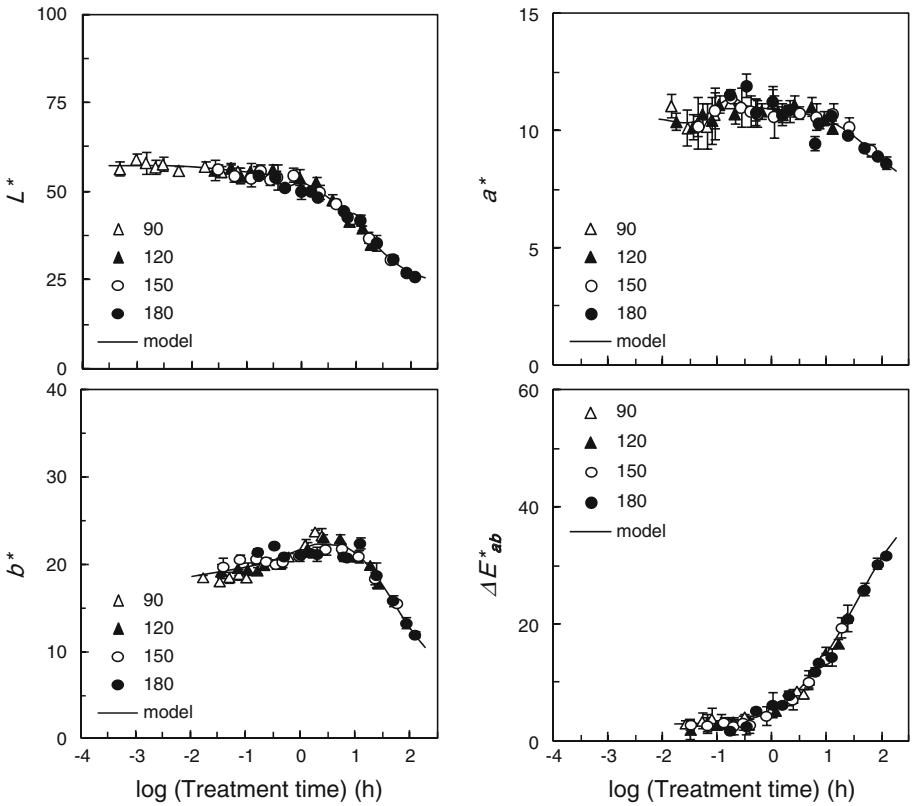

Fig. 6 Changes in color parameters of sugi wood: best-fit model [Eq. (9)] and superposed data using only horizontal shift factor a_T for L^* and ΔE^*_{ab} and using both a_T and vertical shift factor b_T for a^* and b^* (*error bar* standard deviation)

softwoods and keyaki is hardwood, the present results differ from those of previous studies, which suggested that the thermal degradation of hardwood proceeds faster than that of softwood because some components of hardwood are more reactive than those of softwood [33, 34]. The differences in E_a values among color parameters may therefore be interpreted as differences in reactivity of the chemical components responsible for each color parameter.

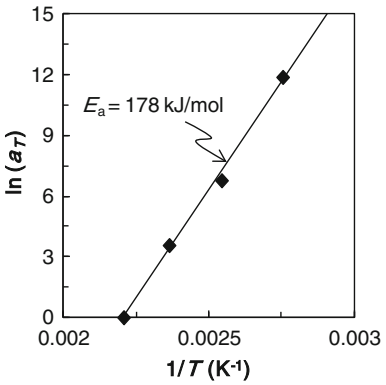

Fig. 7 Arrhenius plots for L^* of keyaki wood, regression line ($R^2 = 0.997$), and calculated apparent activation energy (E_a)

Table 5 Comparison of apparent activation energies (E_a) calculated using the modified Arrhenius equation [Eq. (5)] among several wood species [12] and cellulose filter paper [13]

	E_a (kJ/mol)			
	Keyaki wood	Sugi wood	Hinoki wood [12]	Cellulose filter paper [13]
L^*	178	180	117	125
a^*	132[a]	135[a]	95	124
b^*	133[a]	129[a]	114	118
ΔE^*_{ab}	188	123	113	120

[a] Values of E_a were calculated by applying both a_T and b_T for a^* and b^* of keyaki and sugi wood

These results implied that reactivity may differ according to species and physical properties. However, further investigations including chemical analysis are needed to elucidate the E_a values obtained in this study.

The relationships between b_T and treatment temperature for a^* and b^* are shown in Fig. 8. As the reference temperature was 180 °C, b_T at 180 °C was 1.0. The values of b_T represent the difference in the extent of decrease or increase in a^* or b^* between treatment at 180 °C and that at other temperatures. The values of b_T decreased with increasing temperature toward 1.0 at 180 °C in both a^* and b^*. In Figs. 2, 3, and 8, specimens were more reddish and yellowish at lower temperatures and b_T expressed its degree. The relationship between b_T and temperature was linear from 120 to 180 °C, but b_T seemed to be saturated at temperatures less than 120 °C. This result indicates that treatment at lower temperatures keep colorimetric parameters larger and it may have an upper limit. Studies examining color changes at lower temperatures than those used in this experiment will allow further evaluation of b_T.

In the present study, the factors affecting temperature dependency expressed by E_a and b_T of color changes in keyaki and sugi wood could not be determined. However, the temperature dependency may have been due to the extractives responsible for the characteristic color of wood, which are usually expressed as chromatic parameters such as a^* and b^* [29]. Characteristic extractives possibly give different E_a to different wood species and to different color parameters such as L^*, a^*, b^*, and ΔE^*_{ab}.

From the results of this study, we can conclude that color changes in keyaki and sugi wood during heat treatment can be successfully analyzed using a kinetic approach combined with the TTSP method. This strategy allows prediction of color changes at any of the temperatures and durations used in the present study using E_a and a_T. Temperature dependencies expressed by b_T must be considered when needed.

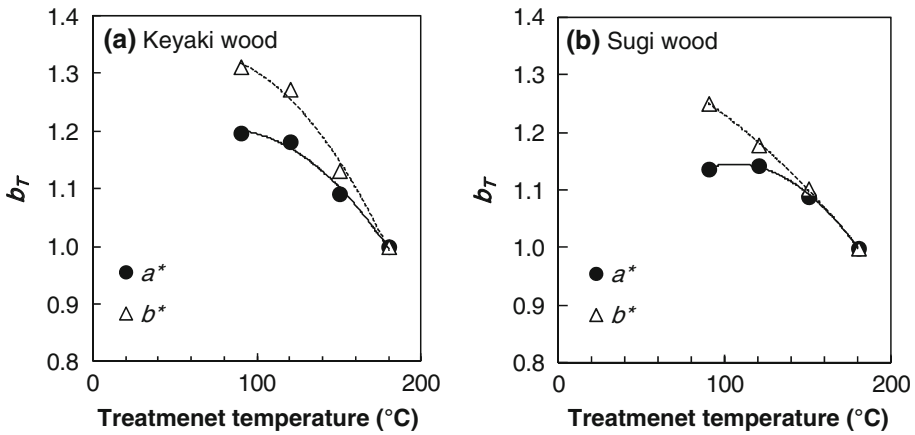

Fig. 8 Relationship between vertical shift factor b_T and treatment temperature for a^* and b^*. **a** Keyaki wood and **b** sugi wood

Conclusions

Color changes in keyaki and sugi wood during heat treatment at 90–180 °C were investigated in this study. The values of L^* and ΔE^*_{ab} decreased and increased, respectively, in both wood species at all treatment temperatures, whereas the values of a^* and b^* showed species- and temperature-dependent behaviors. Kinetic analysis was successfully applied to describe these color changes. Calculating the values of apparent activation energy and shift factors for time–temperature superposition allowed prediction of color changes in the present experimental conditions. The present study revealed and quantitatively evaluated differences in color change behavior between species and temperatures. The results provide a more comprehensive understanding of color changes in wood as a function of time and temperature.

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