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Color Change of *Pinus densiflora* Thermally Modified with Oil and Convection Air

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Color changes of *Pinus densiflora*, oil heated at the temperature of 180ºC and 200ºC, and convection air heated at the temperature of 200ºC were investigated. Belgian flaxseed oil was used for oil heating treatment. Color indexes of CIE $L^*a^*b^*$ were measured by using HunterLab MiniScan XE Plus. It was revealed that heating temperature was more influencing factor than heating media, such as oil and convection air. Oil heating at the temperature of 200ºC resulted in more uniform and effective color change of *Pinus densiflora* specimens than the convection air heating treatment at the temperature of 200ºC and the oil heating treatment at the temperature of 180ºC.

Key words: Oil heating, Convection air heating, CIE $L^*a^*b^*$, Color index, *Pinus densiflora*

INTRODUCTION

According to heating method, the heating treatment technology of wood could be classified to the oil heating method of Germany, the Plato method of Netherlands, the nitrogen heating method of France and the convection air heating method of Finland (Rapp, 2001). The heat treatment is a technology to heat wood in the temperature range from 160ºC to 250ºC, resulting to increase the dimensional stability, weather resistance, and color uniformity by darkening (Yilgor and Kartal, 2010; Korkut *et al*., 2010; Borrega and Karenlampi. 2010; Poncsak *et al*., 2011)

The following problems were found after convection air method was applied in mass production: the dust, smoke and unpleasant smell occurred during carbonizing process, a fire was easily caused by overheat, energy efficiency was low because the temperature of wood was raised by air with low heat capacity (Sidorova *et al*., 2010). However, oil heating method is carbon neutral because that recyclable natural plant oil is used and that smoke and fire are not generated. Moreover, its energy efficiency is high due to using the oil with high heat capacity (Steele, 2012). In New Zealand, the effects of oil heating on color change, dimensional stability, oil absorption rate, coefficient of anti–swelling, UV degradation, weatherability of *Pinus radiata* have been investigated intensively (Dubey *et al*., 2010; 2011; 2012ab; 2014).

Coniferous species, such as *Pinus densiflora*, *Pinus rigida*, *Pinus Koraiensis* etc., are susceptible to be discolored by fungi, which degrades their values. Deciduous trees infected by bacteria are also discolored. Heating treatment make the color of wood dark and uniform. In this study the effects of oil and convection air heating on the color of *Pinus densiflora* are investigated.

MATERIALS AND METHODS

Sample specimen preparation

The dTen pieces of flat–grained *Pinus densiflora* specimens with the dimension of 20 mm in thickness, 100 mm in width and 150 mm in length, and with average moisture content of 10 percent were prepared for each treatment. Three treatments (oil heating at 180ºC and 200ºC and convection air heating at 200ºC) were conducted, thus the total number of specimens was 30 pieces.

Measuring oven–dry densities Oil heat treatments (OL–180 and OL–200)

The oil heating treatments was conducted in an autoclave (350 mm in diameter and 600 mm in height) filled with Belgian flaxseed oil with the a specific gravity of 0.92~0.93 at 23ºC. The treatment time was 2 hours, which began after the core of a specimen reached the target temperatures. A data logger (Hydra 2625 A, 20 ch, Fluke, USA) and copper–constantan thermocouples were used to measure oil and specimen temperatures. The target temperatures were 180ºC (OL–180) and 200ºC (OL–200).

Convection air heating treatment

Convection air heating treatment was conducted in a dry kiln that had a stacking space of 700 mm in width, 800 mm in length and 600 mm in height, with a air velocity of 4 m/s. The time schedule for convection air heating treatment is shown in Figure 1. Duration for increasing the temperature in a kiln from room temperature to 100ºC was 1 hour with the heating rate of 1.3ºC/min. The temperature in a kiln increased from 100ºC to 200ºC for 5 hours with the heating rate of 0.3ºC/min. Then the

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target temperature of 200°C lasted for 2 hours. Then the heater was turned off and the kiln was cooled down to room temperature for about 4 hours.

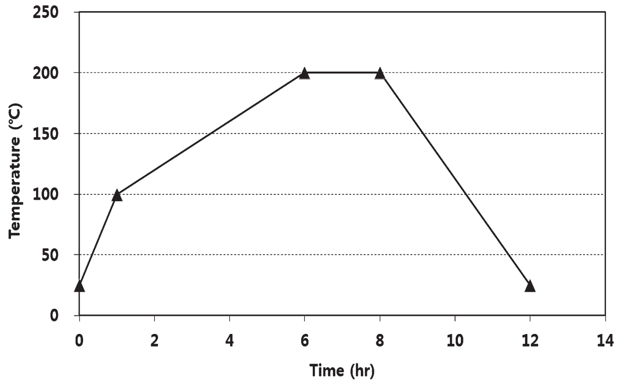

Fig. 1. Time schedule for convection air heating treatment.

Color measurement

Color indexes of CIE $L^*a^*b^*$ were measured by using a color–difference meter (HunterLab MiniScan XE Plus). A source of light was Xenon flash lamp in the wavelength range of 400~700 nm. Because excess oil on the surfaces of specimens influenced on color measurement, specimens were surfaced with an electric planer. Planning reduces the thickness of a specimen about 0.5 mm each. Color measurements were conducted at 1.0 mm and 4.0 mm below the initial thickness. Three randomly distributed positions on each planed surface were measured and averaged. The color difference index was calculated by the equation (1).

$$\Delta E^* = \sqrt{(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2} \qquad (1)$$

Here, ΔE^* is a color difference index, L^* is a lightness index, a^* is a redness index, and b^* is a yellowness index.

Sensitive expressions according to ΔE^* are as Table 1, which is provided by National Bureau of Standards of U.S.A. ΔE^* should be larger than 6.0 for two samples to be appreciably different in color.

Table 1. Sensitive expression of a color difference index (National Bureau of Standards)

ΔE^*	Sensitive expression
0~0.5	Trace
0.5~1.5	Slightly
1.5~3.0	Noticeable
3.0~6.0	Appreciable
6.0~12.0	Considerably
Above 12.0	Extremely

RESULTS AND DISCUSSION

Eyeball observation

The colors of the specimens heat–treated by oil and convection air were compared with the control in Figure 1. The photos were taken at the depths of 1mm below the initial thicknesses. Their colors look obviously different from each other. The grains on oil heated specimens are more noticeable than the others.

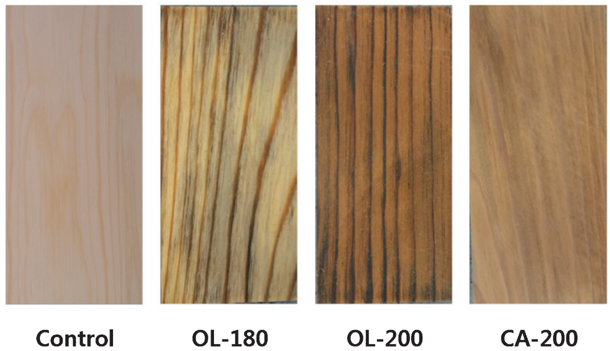

Fig. 2. Photos of *Pinus densiflora* specimens heat–treated at 180°C and 200°C.

Color indexes

The lightness indexes (L^*) of all specimens after heating treatment are shown in Figure 3. The lightness index of the control was the average of those obtained before heat treatments. At 1mm depth, the average lightness index of OL–180 specimens was the highest, followed by CA–200 and OL–200 specimens, while at 4 mm depth those of OL–180 and CA–200 specimens were almost equal and much higher than OL–200. For OL–180 and CA–200 specimens the average lightness indexes at 4 mm depth were definitely higher than those at 1 mm depth, which means that their colors were not uniformly darkened along the thickness. However for OL–200 specimens the average lightness index at 4mm depth was almost the same as that at 1 mm depth. Comparing the specimens heated at 200°C, it could be concluded that oil heating was more effective than convection air heating to change wood color although the treatment time of the former was shorter than that of the latter.

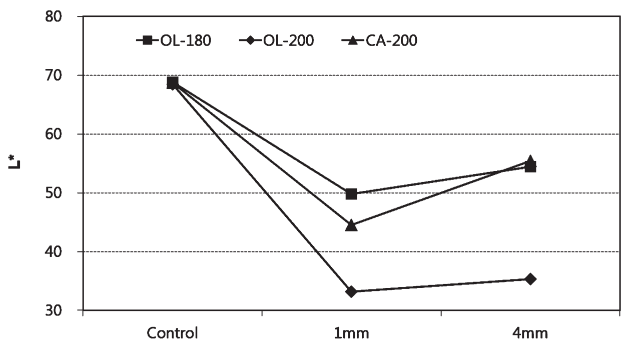

Fig. 3. Average lightness indexes (L^*) at the depth of 1 mm and 4 mm below the initial surfaces of *Pinus densiflora* specimens heat–treated at 180°C and 200°C.

The redness indexes (a^*) of all specimens were substantially increased as shown in Fig. 4. The redness index of OL–180 specimens was less changed than those of OL–200 and CA–200 specimens which could be attributed to its low treatment temperature. The redness indexes of OL–200 and CA–200 specimens presented almost same whether at the depths of 1 mm or 4 mm.

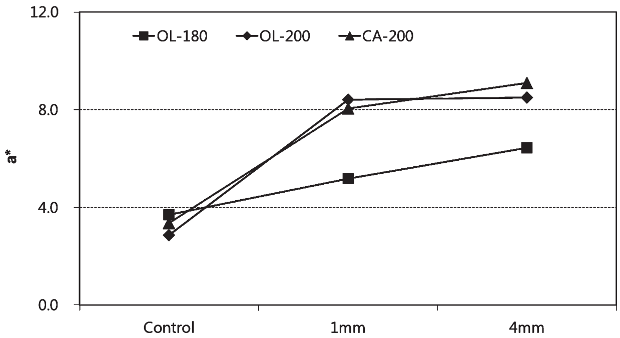

Fig. 4. Average redness indexes (a^*) at the depth of 1 mm and 4 mm below the initial surfaces of *Pinus densiflora* specimens heat–treated at 180°C and 200°C.

The yellowness indexes (b^*) of all specimens after treatment were slightly reduced compared to that before heat treatment at the depth of 1 mm, but those were increased at the depth of 4 mm, as shown in Fig. 5. The yellowness index of CA–200 specimens was almost unchanged or somewhat lower than that of control specimens, which was different from the result of a previous study (Lee *et al.*, 2015): the yellowness index of oil heated *Pinus Koraiensis* specimens was increased at the depth of 1 mm while that was reduced at the depth of 4 mm.

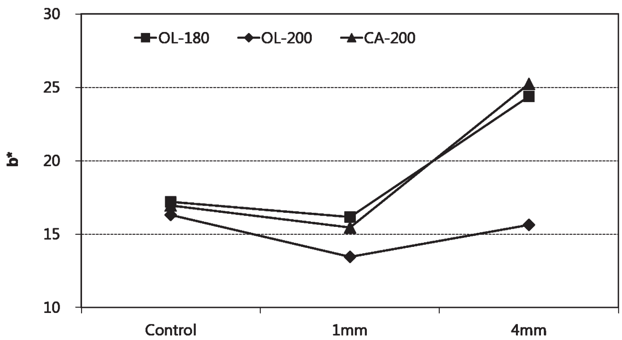

Fig. 5. Average yellowness indexes (b^*) at the depth of 1 mm and 4 mm below the initial surfaces of *Pinus densiflora* specimens heat–treated at 180°C and 200°C.

The yellowness indexes of OL–180 and CA–200 specimens were similarly higher than that of OL–200 specimens at the depth of 4 mm. Lower the yellowness index of specimen was, more the wood color of the specimen was changed, thus, it could be considered that effect of oil heating treatment on yellowness indexes was larger than that of convection air heating treatment under the same temperature of 200°C. The changes of redness index and yellowness index and the relation between them can be easily found in Fig. 6. The distribution range of equivalent of control specimens became narrow after heating treatment although that was widely distributed before heating treatment. The distribution range of yellowness index of heated specimens was not far off that of control specimens. The redness indexes were arranged in the order of OL–200, CA–200, OL–180.

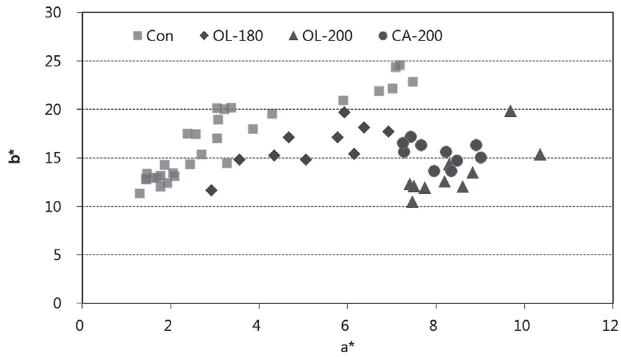

Fig. 6. Plots of yellowness index (b^*) vs. redness index (a^*) at the depth of 1 mm blow the initial surfaces of *Pinus densiflora* specimens heat–treated at 180°C and 200°C.

Color difference index

The color difference indexes (ΔE^*) calculated by the equation (1) at the depth of 1mm below the surface of specimens before and after heating treatments are shown in Fig. 7.

The wood color of all specimens was extremely changed after heating treatment at the depth of 1 mm because the color difference indexes of all specimens were far above 15. Especially the color difference indexes of OL–200 and CA–200 specimens were higher than OL–180, which proved that the temperature of heating influenced more for the color change of wood than the kind of heating medium. The color difference index of OL–200 specimens was much higher than that of CA–200 specimens, which indicated that oil heating treatment was more effective to change the wood color than convection air heating treatment.

The color differences between the depths of 1 mm

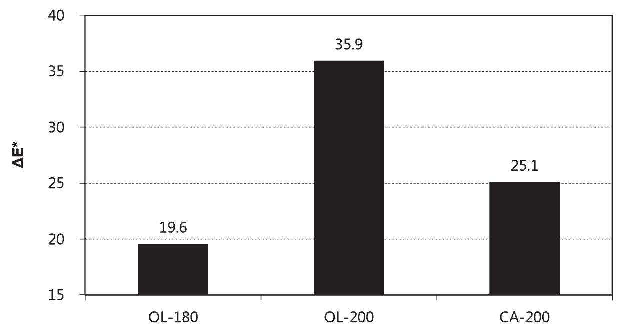

Fig. 7. Average color difference indexes (ΔE^*) at the depth of 1 mm below the initial surfaces of *Pinus densiflora* specimens heat–treated at 180°C and 200°C, compared with the control.

and 4 mm were evaluated by calculating color difference indexes (Fig. 8). The color difference index of OL–200 specimens was the smallest (=4.9) among all specimens. According to Table 1, the color difference index of OL–200 specimens was expressed as appreciable, while the color difference indexes of CA–200 and OL–180 specimens was as extremely severe.

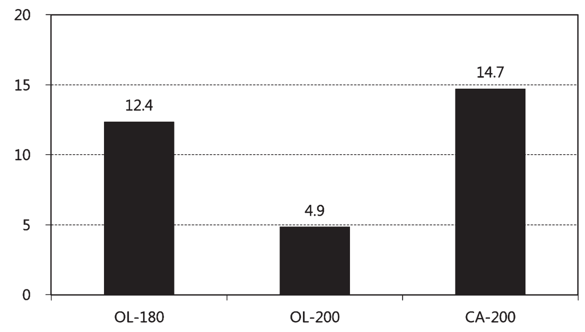

Fig. 8. Average color difference indexes (ΔE^*) at the depth of 4 mm below the initial surfaces of *Pinus densiflora* specimens heat-treated at 180°C and 200°C, compared with at the depth of 1 mm of the same specimens.

CONCLUSIONS

The color change of *Pinus desiflora* specimens, heat–treated by oil at the temperatures of 180°C and 200°C, and by convection air at the temperature of 200°C, was investigated. The results are as followings:

1. There was a little differences in the lightness indexes of oil heated and convection air heated specimens at the depth of 1mm but there was large differences in the lightness indexes at the depth of 4mm.
2. The redness indexes of all specimens were substantially increased and presented almost same values for OL–200 and CA–200 specimens whether at the depth of 1mm or 4mm.
3. The yellowness indexes of all specimens were slightly reduced at the depth of 1 mm, but those were increased at the depth of 4 mm.
4. Among three heating treatments oil heating at the temperature of 200°C resulted in most uniform and effective color change of *Pinus densiflora* specimens

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Author Contributions: Kang, H. Y. designed the study, performed the air heat and oil heat treatment experiments, and compared two methods. Kang, C. W. analyzed data and wrote the paper. Matsumura, J. provided valuable opinions on the entire paper. All authors assisted in editing of the manuscript and approved the final version.