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Changes in the physical and chemical properties of six Japanese softwoods caused by lengthy smoke-heating treatment

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Abstract The effects of prolonged smoke-heating treatments on wood quality were investigated. Six Japanese softwoods were smoke-heated for 100 and 200 h at a temperature of 75° ± 5°C, which was recorded inside the log. After smoke heating, wood quality, including moisture content, amounts of chemical components, relative degree of crystallinity (RDC) of cellulose, and sapwood color were examined. Moisture content decreased as a result of smoke heating, especially in sapwood, leading to a uniform distribution of moisture content within a log. Almost no difference was found in the amounts of chemical components between the control woods and the woods that were smoke-heated for 100 h. However, in the wood that was smoke-heated for 200 h, the amounts of holocellulose decreased, suggesting that thermal deterioration and/or degradation of hemicelluloses had occurred. We assume that the increase in RDC was caused by smoke heating with the crystallization of cellulose and/or thermal degradation of hemicelluloses. Almost no differences were found in sapwood color between the control woods and the woods that were smoke-heated for 100 h. In the wood that was smoke-heated for 200 h, however, ΔL^*decreased, whereas Δa^* and Δb^* increased. As a result, ΔE^*ab, showing the total color change, increased, resulting in a deeper color. These results suggest that thermal degradation of hemicelluloses was caused by smoke heating for over 100 h. Therefore, smoke heating of softwood logs using a commercial-scale kiln should not exceed 100 h.

Key words Smoke heating · Moisture content · Chemical components · Degree of crystallinity · Sapwood color

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Introduction

During the past decade, several researchers have tried to improve wood quality with smoke-heating treatments.[1–11] With smoke heating, the inside of a log is generally heated to a temperature of 80°C for 35–40 h by circulating smoke, which is generated by burning wood. When logs are satisfactorily smoke-heated, improvements in wood quality, such as reduction of growth stresses, decrease in moisture content, uniform distribution of moisture content within a log, and increase in sawing yields, can be obtained. However, deterioration or degradation of hemicelluloses, surface checks, and carbonization are frequently caused by excessive treatment, which is characterized by high temperatures and lengthy exposure. Smoke heating of logs, therefore, has been generally conducted for less than 40 h at a temperature inside the log of 80°C.

On the other hand, Tejada et al.[4] reported that almost no reduction of static bending strength occurred as a result of prolonged smoke heating for 70 h at a temperature inside the log of 80°–100°C. We also reported that almost no thermal deterioration of wood qualities occurred even after smoke heating for 70 h at a temperature inside the log of 75° ± 5°C.[7] However, we pointed out that slight degradation of hemicelluloses might have been caused by smoke heating for 100 h at 100°C because the sapwood color became significantly darker.[9] The color change has so far been determined to be a result of thermal deterioration or degradation of hemicelluloses.[12] Unfortunately, we did not analyze the changes in the amounts of chemical components in wood that was smoke-heated for 100 h at 100°C. Thus, the influence of prolonged smoke heating on the chemical and physical properties of wood has not yet been determined.

In the present study, six Japanese softwood logs were smoke-heated for 100 and 200 h at a temperature inside the log of 75° ± 5°C. After smoke heating, the moisture content, chemical components, relative degree of crystallinity, and sapwood color were examined. Based on the results obtained, the effects of the duration of smoke heating on the quality of softwood are discussed.

Materials and methods

Materials

The following six Japanese softwoods were used in the present study: Japanese cedar (*Cryptomeria japonica* D. Don), Japanese larch (*Larix kaempferi* Carriere), Japanese red pine (*Pinus densiflora* Seib. et Zucc), Jpanese cypress (*Chamaecyparis obtusa* Sieb. et Zucc), Saghalin spruce (*Picea glehnii* Mast.), and Sakhalin fir (*Abies sachaliensis* (fr. Schm.) Mast.). Green logs (3 m in length and ca. 20 cm in end diameter) were harvested from these trees, and each was then cut into three 1-m lengths. The first and second logs from the bud end were used for smoke heating for 100 and 200 h, respectively, and the third one was used as a control.

Smoke heating

Smoke heating of logs was conducted by using a kiln.[8] The temperature inside the log was controlled at 75° ± 5°C for 100 and 200 h continuously by circulating smoke gas at 120°–130°C in the heating chamber. The changes in temperature within a log were monitored with thermocouples inserted 10 cm in depth from the surface and were recorded with a data recorder (Hakusan, LS-3000).

Moisture content

After smoke heating, a 3-cm thick disk was taken from the center of each smoke-heated log. To determine the moisture content (MC), 1.5-cm wide strips were taken through pith to bark from each disk. The MC was determined at every three annual rings from pith to bark by oven drying.

Chemical components

Wood meal (42–80 mesh), which was prepared from the sapwood of the central part of each log, was used to determine the chemical components. Amounts of extracts (hot water, 1% NaOH, and ethanol–toluene) and main components (holocellulose, α-cellulose, and lignin) were determined according to a method described previously.[10]

Relative degree of crystallinity

To determine the relative degree of crystallinity (RDC), wood meal (80–120 mesh) was taken from the sapwood of the central part of both the control and smoke-heated woods. Wood meal was conditioned at 20°C and 65% relative humidity. The RDC was determined from an X-ray intensity curve which was obtained by using an X-ray apparatus (JEOL JDX-12VA). An X-ray diffraction analysis was conducted at 20°C and 65% relative humidity. The measuring conditions were as follows: characteristics of X-ray, CuKα; divergence slit angle, 1/2 degree; scatter slit angle, 1/2 degree; receiving slit, 0.2 mm; tube voltage, 40 kV; tube current, 100 mA; time constant, 1 s; scanning speed, 2 degrees/min.

Sapwood color

Wood meal (42–80 mesh) was used for measurement of the sapwood color. The sapwood color was measured using a colorimeter (Minolta, CR-200) and evaluated with the $L^*a^*b^*$ system based on the JIS Z8729. The total color difference (ΔE^*ab) was calculated from the following equation:

$$\Delta E^*ab = \left(\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}\right)^{1/2}$$

where, ΔL^*, Δa^*, and Δb^* are differences of obtained values in each color index between control and smoke-heated woods.

Results and discussion

Moisture content

Changes in the MC within logs subjected to smoke heating for 100 and 200 h are shown in Fig. 1. The MC decreased in all species, and the decrease ratio was greater in the sapwood than in the heartwood. As a result, the difference in

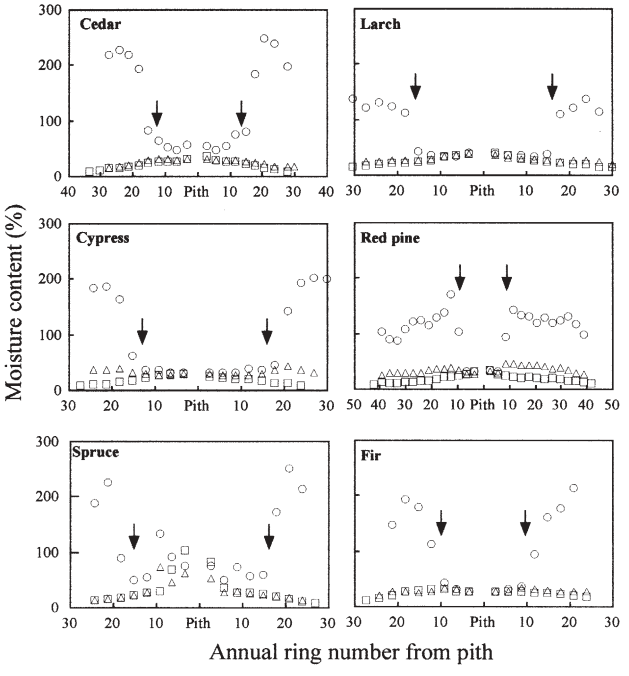

Fig. 1. Changes in moisture content as a result of smoke heating of six Japanese softwoods. *Open circles*, control; *open triangles*, 100-h treatment; *open squares*, 200-h treatment. *Arrows* indicate boundary between heartwood and sapwood

the MC between the heartwood and sapwood decreased, resulting in an almost uniform distribution of the MC within a log.

It is well known that the MC within a log is decreased and made uniform by smoke heating.[1,2,4,6,9,10] However, it has been pointed out that surface checks frequently occur when the MC decreases below the fiber-saturation point.[2] In a previous study,[10] Japanese cedar logs were smoke-heated at 80°C using a laboratory-scale kiln; an almost uniform distribution of MC within a log was obtained with a 60-h smoke-heating treatment, whereas many surface checks occurred when the treatment exceeded 60 h. We concluded, therefore, that the limit of smoke-heating duration at 80°C inside a log was about 60 h based on the relationship between the MC and the frequency of surface checks.[10] The present study shows that the MC of the sapwood in logs smoke-heated for 100 h decreased to around the fiber-saturation point. In logs smoke-heated for 200 h, however, the MC of the sapwood decreased below the fiber-saturation point. Thus, in all species, many surface checks occurred in the logs that were smoke-heated for 200 h. These results suggest that smoke-heating of logs using a commercial-scale kiln should not exceed 100 h at this temperature, considering the frequency of surface checks.

Chemical components

Table 1 shows changes in the amount of chemical components caused by smoke heating for 100 and 200 h. Almost no differences were found in the amounts of α-cellulose, lignin, and ethanol–toluene extracts between the control and smoke-heated woods. The amounts of holocellulose decreased apparently as a result of smoke heating for 200 h. In contrast, the amount of hot-water and 1% NaOH extracts increased with an increase in the length of the smoke-heating treatment.

It has been reported that, when wet wood is heated, a loss of mechanical strength occurs, probably due to an increase in the amount of acetic acid derived from the degradation of hemicelluloses during heating.[13] Tejada et al.[5] also reported that heat treatment of wood in wet conditions produces a greater reduction of hemicellulose, whereas the α-cellulose content does not change. Smoke heating is a method of heating wood in wet conditions. It has been considered, therefore, that thermal degradation of hemicelluloses in wood occurs as a result of smoke heating for longer than 40 h even at temperatures as low as 80°–100°C.[3] However, almost no thermal degradation of wood chemical components in the logs of Japanese larch occurred, even when the duration of smoke heating exceeded 70 h.[7] It has also been reported that thermal degradation of hemicelluloses might not be caused by smoke heating within 100 h at a temperature inside the log below 100°C.[9]

In the present study, almost no differences were found in the amounts of chemical components between the control woods and the woods subjected to smoke-heating treatment for 100 h. In contrast, the amounts of holocellulose decreased after smoke heating for 200 h, resulting in an increase in the amounts of hot-water and 1% NaOH extracts. This observation was also made by Okuyama et al.[3] As a result, in all species, the amounts of hemicellulose (A–B), which may contain β-cellulose and low molecular weight cellulose, decreased after smoke heating for 200 h compared with controls by: cedar, 4.6%; larch, 4.5%; cypress, 4.3%; red pine, 7.6%; spruce, 4.1%; fir, 6.5% (see Table 1). We assume, therefore, that the thermal degradation of wood chemical components, especially hemicelluloses, might have occurred as a result of smoke-heating treatment for 100–200 h at a temperature inside the log of about 75° ± 5°C. Further research is needed concerning the treatment time at which the thermal degradation of hemicelluloses occurs.

Table 1. Changes in amounts of chemical components as a result of smoke heating for 100 and 200 h

Species	Treatment	Main components (%)				Extracts (%)		
		Holocellulose (A)	α-Cellulose (B)	A − B	Lignin	Hot water	1% NaOH	Ethanol–toluene
Cedar	Control	79.6	48.5	31.1	33.4	0.9	8.1	1.9
	100 h	76.6	46.2	30.4	33.3	0.7	10.3	1.3
	200 h	73.5	47.0	26.5	32.6	2.3	11.7	1.6
Larch	Control	75.2	46.1	29.1	25.7	0.8	10.5	2.0
	100 h	79.6	49.9	29.7	27.6	1.6	13.5	2.2
	200 h	76.8	52.2	24.6	26.9	4.3	15.4	2.8
Cypress	Control	80.0	52.0	28.0	31.3	0.7	10.2	1.3
	100 h	78.0	48.4	29.6	31.9	1.9	11.3	2.0
	200 h	71.3	47.6	23.7	32.3	3.2	12.6	1.4
Red pine	Control	81.1	49.6	31.5	27.9	1.4	10.9	1.6
	100 h	81.5	50.8	30.7	27.9	0.2	12.0	2.2
	200 h	75.7	51.8	23.9	28.0	2.1	14.4	2.5
Spruce	Control	79.9	50.2	29.7	29.2	1.2	10.3	1.8
	100 h	73.6	48.1	25.5	30.0	1.1	11.9	1.8
	200 h	73.8	48.2	25.6	29.0	2.5	12.3	1.9
Fir	Control	80.5	47.4	33.1	27.8	1.9	10.6	2.7
	100 h	78.1	45.7	32.4	27.6	2.3	12.1	1.7
	200 h	73.9	47.3	26.6	27.7	2.3	12.6	2.4

Relative degree of crystallinity

Changes in the RDC caused by smoke heating for 100 and 200 h are shown in Fig. 2. In all species, RDC values were increased by smoke heating.

It is well known that the degree of crystallinity (DC) increases during the initial process when wood is heated at a temperature in excess of 100°C.[14–17] Bhuiyan et al.[17] found that crystallization occurs in wood cellulose after heat treatment in high-moisture conditions, at a rate almost twice that observed in oven-dried conditions. They also observed that more crystallization occurred in wood cellulose than in pure cellulose. As a result, they concluded that other components accompanying wood cellulose are involved in the increase of crystallinity after heat treatment and that wood cellulose contains more quasicrystalline regions than does pure cellulose. In addition, Hirai et al.[14] reported that the increase of DC was due to the decreased amorphous region by thermal decomposition in addition to crystallization of the amorphous region.

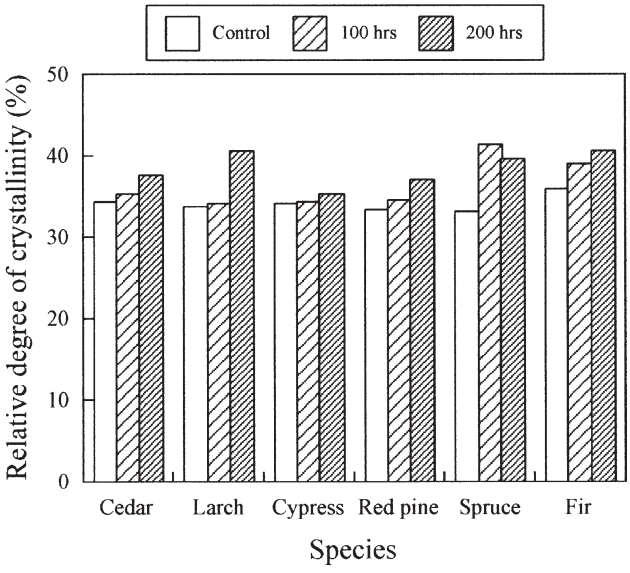

Fig. 2. Changes in relative degree of crystallinity as a result of smoke-heating of six Japanese softwoods

Smoke heating of wood, which is a heating treatment in wet conditions as described above, increased the DC and RDC.[4,7,9,10] Tejada et al.[4] reported that the DC increased significantly in Japanese cedar, larch, and fir when logs were smoke-heated for 70 h at a temperature inside the logs of 80°–100°C. It was also found that the RDC of Japanese cedar logs increased significantly after smoke heating at a temperature inside the log of 80°C for 30–40 h.[10] In the present study, the RDC also increased as a result of smoke heating with an increase in the length of the treatment, indicating that crystallization of cellulose in the non crystalline regions occurred. On the other hand, as shown in Table 1, the amounts of holocellulose decreased by about 5% in woods that were treated for 200 h, whereas the α-cellulose did not increase significantly. This fact suggests that the increase in the RDC might be accompanied by a decrease in the amounts of hemicellulose, in addition to the crystallization of cellulose amorphous regions.

Sapwood color

Changes in sapwood color as a result of smoke heating for 100 and 200 h are shown in Table 2. Almost no differences were observed in sapwood color between the control woods and the woods of all species that were smoke-heated for 100 h. However, in all of the woods that were smoke-heated for 200 h, ΔL^* decreased and Δa^* and Δb^* increased. Fur-

Table 2. Changes in sapwood color as a result of smoke heating for 100 and 200 h

Species	Treatment	Color index						
		L^*	ΔL^*	a^*	Δa^*	b^*	Δb^*	ΔE^*ab
Cedar	Control	75.0	–	5.0	–	18.9	–	–
	100 h	74.3	−0.6	5.5	0.5	20.9	1.0	2.2
	200 h	73.7	−1.3	6.3	1.3	21.9	2.0	3.6
Larch	Control	76.0	–	4.3	–	19.6	–	–
	100 h	74.1	−1.9	5.6	1.3	20.4	0.8	2.4
	200 h	71.8	−4.2	5.5	1.2	21.4	1.8	4.7
Cypress	Control	74.4	–	4.5	–	19.7	–	–
	100 h	76.9	−2.5	4.7	0.2	21.9	2.2	3.3
	200 h	71.6	−2.8	6.1	1.6	22.7	3.0	4.3
Red pine	Control	77.1	–	4.0	–	19.4	–	–
	100 h	78.3	−1.2	3.5	−0.5	22.3	2.9	3.3
	200 h	73.8	−3.3	5.2	1.2	23.5	4.1	5.4
Spruce	Control	78.4	–	3.6	–	18.1	–	–
	100 h	78.6	0.2	3.4	−0.2	20.3	2.2	2.2
	200 h	76.9	−1.5	4.1	0.5	22.0	3.9	4.2
Fir	Control	80.5	–	2.5	–	16.5	–	–
	100 h	81.3	0.8	2.8	0.3	19.4	2.9	3.1
	200 h	77.8	−2.7	3.9	1.4	21.4	4.9	5.8

thermore, the increase of Δb^* was greater than that of Δa^*. As a result, the sapwood color of woods that were smoke-heated for 200 h became darker. In all species, the ΔE^*ab values showing a total color change were greater in the 200-h treatment than in the 100-h treatment, the values being 3.6–5.8. These values indicate appreciable color changes according to the standard of color change in the $L^*a^*b^*$ system.

In general, the L^* value decreased, and the a^* and b^* values increased in the color change of sapwood as a result of smoke-heating treatments.[3,4,9] Almost no changes in sapwood color were reported to occur at a treatment temperature of 60°C, when cedar and larch logs were smoke-heated for 20–100 h at 60°, 80°, and 100°C with a laboratory-scale kiln.[9] However, at 80° and 100°C, the ΔE^*ab increased with an increase in the length of the smoke-heating treatment to values above 5, indicating an appreciable color change. In the present study, the ΔE^*ab value also increased with an increase in the duration of the smoke-heating treatment. We assume that the increases of ΔL^* and Δb^* are the main contributors to the change in sapwood color that occurs as a result of smoke-heating treatments.

On the other hand, it is well known that wood color becomes darker with the hydrolysis of hemicelluloses subjected to heat treatment.[12] Terziev et al.[18] reported that yellowness in the sapwood of *Pinus sylvestris* L. increased with an increase in the amount of low molecular weight sugars derived from the thermal deterioration or degradation of hemicelluloses. Thus, thermal deterioration or degradation of hemicellulose is closely related to the deeper color change of the sapwood. In the present study, as shown in Table 1, the content of holocellulose decreased when subjected to smoke heating for 200 h, corresponding to an increase in the amounts of hot-water and 1% NaOH extracts. These results indicate that hemicelluloses might deteriorate or degrade when subjected to prolonged smoke-heating treatments. The results obtained suggest that sapwood color is greatly affected by thermal deterioration or degradation of hemicelluloses subjected to smoke-heating treatments.

Conclusion

The present study is an investigation of the effects of prolonged smoke-heating treatments on the quality of six Japanese softwoods. Logs were smoke-heated for 100 and 200 h at a temperature inside the log of 75° ± 5°C. After smoke heating, the changes in the quality of the wood were examined. The following results were obtained:

1. The difference in the MC between the heartwood and sapwood decreased as a result of smoke heating. In all species, however, many surface checks occurred in the logs that were smoke-heated for 200 h.
2. Almost no differences were found in the amounts of chemical components when comparing the control woods and the woods that were smoke-heated for 100 h. In contrast, the amounts of holocellulose clearly decreased as a result of smoke heating for 200 h.
3. The RDC values increased in all species as a result of prolonged smoke-heating treatments.
4. In the wood that was smoke-heated for 200 h, the ΔL^* decreased, and the Δa^* and Δb^* increased. As a result, the ΔE^*ab values were 3.6–5.8, indicating appreciable color changes according to the evaluation of color change in the $L^*a^*b^*$ system.

These results suggest that thermal deterioration or degradation of hemicellulose occurred as a result of smoke heating for longer than 100 h. Therefore, smoke heating of softwood logs using a commercial-scale kiln should not exceed 100 h.

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