Effects of Drying Temperature for *Cryptomeria japonica* on the Permeability of Wood Preservative. II: The Permeability of Dried, Split Log Pieces

Hiroki Sakagami,a,* Atsuro Tokunaga,b Noboru Fujimoto,a Shinya Koga,c Isao Kobayashi,d and Ikuo Momohara e

Poor impregnation of sapwood from *Cryptomeria japonica* kiln-dried logs is a problem for preservative treatment in Japan. The permeability of copper azole (CuAz) into sapwood was reported to decrease with an increase in the drying temperature of logs, due in part to the presence of bordered pits. However, damaged and aspirated bordered pits appeared abundantly at 100 °C and 120 °C, although the difference in permeability was very little. To investigate this phenomenon, two types of smaller split log pieces, one containing both heartwood and sapwood, and the other containing sapwood without heartwood, were dried at 20 °C to 120 °C to test higher drying conditions. Results were similar to those of the dried logs. However, the impregnation and penetration at 80 °C were the lowest, and those at 100 °C and 120 °C were greater than the dried logs. Additionally, the number of damaged bordered pits on dried, split samples was generally higher than that of dried logs, as observed with scanning electron microscopy.

Keywords: Permeability; Absorption; Preservative; Dry; CuAz; Cryptomeria japonica

**INTRODUCTION**

_Cryptomeria japonica_ D. Don is the most popular domestic commercial wood in Japan, and its effective utilization is important. Recently, small logs and sawn timbers have been used for civil engineering (Kamiya 2003; Kubojima et al. 2012; Noda et al. 2014). Most woods destined for these uses are treated with wood preservative. However, it is difficult to impregnate the timber evenly with preservative due to non-uniform permeability by some factors. (Erickson 1970; Flynn 1995). Problems with sapwood permeability for _C. japonica_ kiln-dried logs have gained special attention due to deterioration of durability in Japan. The preservative penetration of air-dried logs is significantly better than that of the kiln-dried logs (Momohara et al. 2009). Although the details are not specified, the cause is likely the different log drying conditions.

Fluid flow in softwoods, both sapwood and heartwood, is generally related to the shape, size, and conditions of pits in tracheids. According to Fujii et al. (1997), air permeability has an obvious negative correlation with the pit aspiration ratio in samples collected from sapwood, transition zone between sapwood and heartwood, and heartwood.
Fluid flow is clearly influenced by drying conditions. Studies on the relationship between fluid flow and drying conditions have contradictory results after high temperature drying, reporting enhanced permeability (Booker and Evans 1994; Terziev 2002; Terziev and Daniel 2002; Zhang and Cai 2008; Taghiyari 2013) or poor permeability (Comstock and Côté 1968; Thompson 1969; Momohara et al. 2009; Taghiyari et al. 2014). Even by thermal treatment at high temperature, the opposite results were reported (Ahmed and Morén 2012; Ahmed et al. 2013a,b). Therefore, it is impossible to determine the fluid flow characteristics based on only these results because there may be multiple factors. Wood structures or chemical components vary in species and position within a log or timber, and drying conditions are related to the sample size and drying temperature.

To characterize fluid flow, we previously investigated the absorption and penetration of wood preservative into the sapwood of *C. japonica* logs dried from 20 °C to 120 °C at every 20 °C (Sakagami et al. 2016). Additionally, the relationship between drying temperature and bordered pit conditions classified into five categories with scanning electron microscopy was investigated. The absorption and permeability of sapwood decreased with increasing drying temperature; the temperatures producing the lowest values were 100 °C and 120 °C. With increasing temperature of drying there was a decrease in the proportion of bordered pits classified into the categories of (a) “neutral” and (b) “between neutral and aspiration.” On the other hand, there were increases in the categories of pits classified as (c) “aspiration,” (d) “crack appearance,” and (e) “exfoliation,” all of which were assumed to hinder liquid flow. In sum, fluid flow was controlled by the structure of bordered pits, which was triggered by heat and rapid evaporation, or by faster water movement due to higher drying temperature. However, we were interested in what increased the number of damaged or aspirated bordered pits, which showed different effects on fluid flow, when wood samples were dried under extreme drying conditions; the proportions of exfoliation and aspiration were highest at 100 °C and 120 °C, respectively. We speculated that there was an inflection point of fluid flow if damaged bordered pits increased.

To investigate drying conditions on fluid flow, the remaining logs used in the previous study were dried at different temperatures. Smaller split samples from logs were used to test stronger drying conditions, preservative impregnation, and bordered pit conditions using methodologies similar to the previous study. This study used two types of samples: a split sample of logs, including both heartwood and sapwood, and a split sample with the heartwood removed.

**EXPERIMENTAL**

**Materials**

Seven logs of 50-cm length, labeled from 1 to 7, were derived in sequence from 0.65 m from ground level to 4.75 m in the species *Cryptomeria japonica* (Fig. 1). The bark of each log was removed. The upper region (above 4.75 m) was used in the previous study (Sakagami et al. 2016). Damaged logs, RM1 for 10-cm length and RM2 for 30-cm length, were removed. Four discs, which were used for measuring the green moisture content, were taken from the same section of the tree. The moisture content was calculated from the weight of water in wood based on oven-dry wood weight. The diameter of the logs at the lowest (No. 1) and highest (No. 7) positions were 26 cm and 21 cm, respectively. The green moisture content of sapwood and heartwood for each of the four discs, excluding
intermediate wood, was measured by uneven split small samples based on their green and oven dry weight. Each log was divided into eight pieces through the pith with a band saw. Half of them contained both heartwood and sapwood (HSW). The rest were sapwood specimens (SW) with heartwood portions removed by dividing on the intermediate wood.

![Diagram of log division and drying process]

**Fig. 1.** Collection of seven logs for different drying temperatures and four discs for green moisture content measurement

### Drying of Split Logs

Surfaces excluding the bark side of all split pieces from seven logs were sealed with epoxy resin to limit the evaporation surface to only the bark side. Split pieces of two logs (No. 2 and No. 4) were dried at 20 °C and 65% relative humidity for a year in a controlled environment room. The remaining split pieces of each log were dried at 40 °C, 60 °C, 80 °C, 100 °C, and 120 °C (Table 1) until they reached approximately 10% moisture content, which was measured by the green moisture content of the slice removed from the pieces. Two split pieces (SW and HSW) of the eight derived from a log were used for moisture content measurement. The weights were measured every day, except for those dried at 120 °C, which measured every 3 h because of drying within a day. The weights of pieces dried at 20 °C were measured every several days. After the moisture contents in the pieces reached approximately 10%, they were kept at 20 °C and 65% relative humidity, where the equilibrium moisture content was approximately 12%. The drying rate of each split piece, SW and HSW, was calculated from the change in moisture content based on oven-dry weight.

### Table 1. Drying Conditions Applied to Split Samples of Seven Logs

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Relative Humidity (%)</th>
<th>Duration of Drying (d = days, y = year)</th>
<th>Sample No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>65</td>
<td>1 y</td>
<td>2</td>
</tr>
<tr>
<td>20</td>
<td>65</td>
<td>1 y</td>
<td>4</td>
</tr>
<tr>
<td>120</td>
<td>42</td>
<td>0.75 d</td>
<td>1</td>
</tr>
<tr>
<td>100</td>
<td>80</td>
<td>6 d</td>
<td>3</td>
</tr>
<tr>
<td>80</td>
<td>74</td>
<td>7 d</td>
<td>5</td>
</tr>
<tr>
<td>60</td>
<td>66</td>
<td>11 d</td>
<td>7</td>
</tr>
<tr>
<td>40</td>
<td>62</td>
<td>15 d</td>
<td>6</td>
</tr>
</tbody>
</table>

Preparation of Samples for Permeability Testing
Sticks of 2 cm (radial) × 2 cm (tangential) × 45 cm (longitudinal) were sawn from the sapwood of split pieces derived from logs. A detailed schematic diagram is shown in the previous study (Sakagami et al. 2016). Six clear sticks were selected from the pieces of SW and HSW. From these sticks, 15-cm samples for permeability experiments, a sample several centimeters in size for moisture content measurement when wood preservative was absorbed, and small samples for scanning electron microscopy (SEM) observation were derived.

Treatment of Samples by Wood Preservative
The same wood preservative treatment (Sakagami et al. 2016) was used. Diluted copper azole wood preservative (CuAz; Lonza, Tokyo, Japan) was impregnated into 15-cm samples, of which five surfaces were sealed with epoxy resin. The amount of impregnation by volume was calculated.

Measurement of Penetration Area
The penetration area of the cross surface at every 1 cm from the top surface of samples impregnated with CuAz was measured as described previously (Sakagami et al. 2016). A 0.5% solution of Chrome Azurol S (MP Biomedicals, Santa Ana, CA, USA) in 1% sodium acetate was used to determine the penetration area.

SEM Observation of the Bordered Pits
The aspirated conditions of the bordered pits for the samples dried at different temperatures were observed by SEM (JSM 5600LV, JEOL Ltd., Akishima, Japan) and classified into five categories, as in the previous study (Sakagami et al. 2016): (a) neutral position, (b) between neutral position and aspiration, (c) aspiration, (d) crack appearance along pit aperture, and (e) exfoliation along pit aperture. The probability of each category was calculated from the number of bordered pits in each category divided by the total number of bordered pits counted. The total counts in each sample were 200 to 700.

RESULTS AND DISCUSSION

Moisture Content
The green moisture content of sapwood and heartwood for the four discs (Fig. 1) is shown in Table 2. The average moisture content in sapwood and heartwood was 210.0% and 53.2%, respectively. These moisture contents were higher than the previously reported values of 185.0% for sapwood and 46.7% for heartwood (Sakagami et al. 2016).

Table 2. Green Moisture Content

<table>
<thead>
<tr>
<th>Wood Sample</th>
<th>Moisture Content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Disc 1</td>
</tr>
<tr>
<td>Sapwood</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>252.0</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>14.05</td>
</tr>
<tr>
<td>Heartwood</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>61.7</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>3.75</td>
</tr>
</tbody>
</table>
Drying of Split Log Pieces

The change in moisture content of the split pieces from each log is shown in Fig. 2. Initial moisture contents of HSW and SW pieces were approximately 91 to 126% and 130 to 202%, respectively. The reason for the high variance in SW seemed to be the portion of intermediate wood having low moisture content. The moisture content of the split pieces dried at 20 °C decreased slowly, and they dried quickly, with increasing drying temperature. Comparing HSW and SW, the higher moisture content of SW decreased faster because their smaller shape and faster water movement likely accelerated drying. All pieces reached below 10% in a controlled environment room in the end.

![Graph showing moisture content over time for different temperatures and moisture conditions](image)

**Fig. 2.** The change in moisture content of split samples during drying

Impregnation and Penetration of CuAz

The amount of impregnation of the 15-cm samples by volume for HSW and SW is shown in Table 3. The impregnation conditions were previously described (Sakagami et al. 2016). This mild condition is suitable for distinguishing the difference in absorption among samples dried at different temperatures. The average moisture contents of four or five randomly selected samples when CuAz was absorbed are shown in Table 4. The moisture content of the specimens was approximately 9%.

**Table 3.** Impregnation of CuAz

<table>
<thead>
<tr>
<th>Drying Temperature (°C) (Log Number)</th>
<th>20 (2)</th>
<th>20 (4)</th>
<th>40 (6)</th>
<th>60 (7)</th>
<th>80 (5)</th>
<th>100 (3)</th>
<th>120 (1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Absorption by HSW (kg/m³) (Standard deviation)</td>
<td>379.5 (65.4)</td>
<td>328.0 (88.0)</td>
<td>287.9 (78.9)</td>
<td>185.7 (15.0)</td>
<td>101.2 (17.6)</td>
<td>176.8 (35.6)</td>
<td>137.6 (13.9)</td>
</tr>
<tr>
<td>Absorption by SW (kg/m³) (Standard deviation)</td>
<td>372.9 (65.3)</td>
<td>338.5 (68.8)</td>
<td>304.3 (24.4)</td>
<td>143.5 (30.0)</td>
<td>112.3 (18.1)</td>
<td>166.6 (34.3)</td>
<td>109.2 (26.0)</td>
</tr>
</tbody>
</table>
The highest amount of impregnation of CuAz was 379.5 kg/m³ for HSW dried at 20 °C, which was lower than 431.5 kg/m³ for the log dried at 20 °C (Sakagami et al. 2016). However, the lowest impregnation was 101.2 kg/m³ for HSW dried at 80 °C. This value was higher than 84.8 kg/m³ for the log dried at 100 °C (Sakagami et al. 2016). It seemed that sample size affected the liquid impregnation.

| Table 4. Moisture Contents of Specimens when CuAz was Absorbed |
|-----------------|-------|-------|-------|-------|-------|-------|-------|-------|
| Drying temperature (°C) | 20 (2) | 20 (4) | 40 (6) | 60 (7) | 80 (5) | 100 (3) | 120 (1) |
| Moisture content of HSW | 8.91 | 9.42 | 9.30 | 8.81 | 8.33 | 8.71 |
| Moisture content of SW | 8.88 | 9.03 | 8.73 | 8.99 | 8.73 | 8.10 | 7.63 |

Impregnation tended to decrease with increasing drying temperature, up to 80 °C, whereas the impregnation of samples dried at 80 °C was lower than that at 100 °C and 120 °C in HSW and 100 °C in SW. The impregnation of both HSW and SW samples dried at 100 °C was especially higher. A two-way factorial analysis of variance was conducted for impregnation of CuAz. The difference among drying temperatures was significant at 1%, and there was no difference between HSW and SW. Therefore, it was obvious that the drying temperature of samples affected the impregnation of liquid into sapwood. However, the presence of heartwood did not influence fluid flow. Thus, there was little possibility for extractives in heartwood to move to sapwood with water movement via bordered pits or ray parenchyma cells during drying.

Cross surfaces were observed every 1 cm from the penetrative surface of the specimens impregnated with CuAz (Fig. 3). Representative specimens showing the average impregnation of HSW and SW were selected from each condition. The averaged stained areas for each cross surface are shown in Fig. 4. The penetration of samples dried at 20 °C was the deepest, whereas the penetration depth of samples dried at higher temperatures was drastically reduced, by approximately 4 cm or 5 cm. This trend was similar to previous results (Sakagami et al. 2016). However, there were slight differences. The lowest stained area occurred in split samples dried at 80 °C, and more stained areas were recognized at 100 °C and 120 °C. This was similar to the impregnation results in Table 3, while the lowest for dried logs occurred at 100 °C and 120 °C (Sakagami et al. 2016). Altogether, these results indicate that higher drying temperature hindered the liquid penetration and impregnation of *C. japonica* sapwood; the wood qualities deteriorated when treated with performance enhancing chemicals such as wood preservative and fire retardant.

**Observation of Bordered Pits**

The percentages of appearance by total count in five categories of HSW and SW are shown in Fig. 5. The proportions of bordered pits in the neutral position of category (a) was only 11.2% (No. 2) and 11.9% (No. 4) for HSW, and 13.2% (No. 2) and 11.1% (No. 4) for SW dried at 20 °C, compared with 25.3% and 21.6% for the log samples (Sakagami et al. 2016). Due to the decrease in category (a) dried at 20 °C, more bordered pits were classified as exfoliation of category (e), as compared to logs (Sakagami et al. 2016). As with dried logs (Sakagami et al. 2016), the number of category (a) decreased, and category (e) increased, with elevated drying temperature. Additionally, the number of category (e) for dried split samples was higher than that of dried logs for all temperatures (Sakagami et al. 2016).
**Fig. 3.** Penetration of CuAz on the wood cross surface at 1-cm intervals. The upper image shows the HSW samples. The lower image shows the SW samples. Representative samples, showing average absorption, were chosen. Blue color shows the penetration area; 0 cm indicates the place where liquid penetrated.
Fig. 4. The percentage of CuAz penetration area in cross cut surfaces at 1-cm intervals from the penetrative surface. The averages of six samples are shown. The upper image shows the results of HSW samples. The lower image shows the results of SW samples.

Fig. 5. The relative occurrence probability of bordered pits, as classified by SEM observations. Bordered pits were classified into five categories (Sakagami et al. 2016). The left and right graphs show HSW and SW, respectively.
These results suggest that a higher drying temperature and smaller sample dimension result in greater stress effects on bordered pits. These morphological changes in bordered pits seemed to hinder liquid absorption.

In previous research (Sakagami et al. 2016), we confirmed the poor impregnation of wood preservative into dried log sapwood with a focus on the drying temperature because it was relevant for industry in Japan. The total proportions of bordered pits that were classified into categories (c), (d), and (e), which were assumed to hinder liquid flow, increased. The categories (a) and (b), which were assumed as fluid-permeable conditions, decreased with drying temperature. The softened pit membranes and rapid evaporation, or faster water movement, were caused by high temperature in terms of drying rate. However, the total proportions of categories for (a) (b) and (c) (d) (e) in this study were approximately the same as in the previous report, regardless of the smaller samples.

![Fig. 6. Moisture content of split log pieces. The upper graphs show HSW and SW. The result of drying at 120 °C is shown on the lower graph due to its drastically high value.](image-url)
To confirm the drying rate of split samples, the change in moisture content during drying was calculated (Fig. 6). It was obvious that increased drying stress in wood affected the impregnation because inclination of each drying temperature was larger compared with the results of logs (Sakagami et al. 2016). As mentioned in the introduction, there are contradictory research results about fluid permeability when woods are dried at high temperature. One factor affecting fluid permeability is damaged or ruptured tissues and bordered pits due to drying stress at high temperatures and vaporization forces (Terziev and Daniel 2002; Zhang and Cai 2008; Ahmed and Morén 2012; Taghiyari 2013). Increased drying stress was possible to act on the bordered pits because composition of Cryptomeria japonica related to water or air pathway were only tracheid. The conduits like resin canals for Pinus and vessels for hardwood do not exist. Considering this effect and the drying rates found in this study, it was assumed that the increases in category (e) in all temperatures were due to some greater drying stress than those of logs. One of the reasons for inflating drying stress is the faster heat conduction into the center of smaller samples. This tendency was also observed when comparing HSW and SW, though the difference was not so remarkable. As a result, the number of damaged bordered pits exceeded the number of retained bordered pits, and the split samples dried at a higher temperature (100 °C and 120 °C in this study) absorbed more CuAz than those dried at 80 °C. This result is supported by the comparison of results between 100 °C and 120 °C. Thus, higher impregnation at 100 °C was caused by the higher proportion of category (e), though it was not clear why more exfoliation appeared at 100 °C than 120 °C.

In both this and the previous study (Sakagami et al. 2016), the bordered pits were vulnerable to heat stress and evaporation or water movement in terms of drying temperature, and they interfered with fluid permeability. The potential mechanism affecting fluid flow in Cryptomeria japonica is as follows. The resistance of fluid flow is low when larger pieces of wood dry at lower temperatures because bordered pits remain neutral under a rigid pit membrane, low evaporation pressure, and slow water movement. However, fluid flow is hindered by moving bordered pits due to increasing temperature, higher evaporation pressure, and faster water movement. First, the torus of the bordered pit begins to close toward the pit aperture with increased pressure or water movement. Then, the fluid flow is greatly affected with a higher number of aspirated bordered pits. However, the decreased fluid flow is improved with increasing temperature, excessive vapor pressure, and water movement because damaged bordered pits such as category (e) begin to appear.

Finally, enhancing fluid flow may afford the same permeability with the condition of lower drying temperature, or exceed this permeability by increasing extremely damaged bordered pits or occurring micro cracks, thereby producing larger channels. The contradictory types of fluid permeability for woods dried at high temperatures, as mentioned before, are described in the hypothesis. In the previous study using dried logs, poorer permeability caused by milder drying of larger samples compared with the small dried split log pieces, even if they were dried at high temperatures. Impregnation and penetration into samples in this study dried at 100 °C and 120 °C could be increased; whereas at 80 °C, impregnation was the lowest and could not reach the level of samples dried at 20 °C. Little cell damage was observed with SEM. If any methods to widen the flow path rather than the pit aperture could be developed like other reports (Terziev and Daniel 2002; Zhang and Cai 2008; Ahmed and Morén 2012; Taghiyari 2013), poor permeability issues would be resolved, and the production of wood materials with high permeability would be possible.
The results in this study were insufficient because there were no evidence directly connecting the liquid pathway with the condition of bordered pits, and the main factors affecting bordered pit movement and rupture were not identified. Additionally, it is unclear how the damaged bordered pits contribute to liquid permeability because the thermally modified samples had lower preservative uptake though some damages were observed with a microscope (Ahmed et al 2013b). However, characterization of these factors and finding permeable conditions are indispensable for the wood industry to extend the service life.

CONCLUSIONS

1. The impregnation of CuAz into C. japonica sapwood decreased with increasing drying temperature, from 20 °C to 80 °C in the shape of split log pieces. The highest impregnation of CuAz was observed for the samples dried at 20 °C. The amount of impregnation for samples dried at 100 °C and 120 °C for HSW, and 100 °C for SW, was higher than for those dried at 80 °C, at which the lowest absorption was shown.

2. There was no difference in impregnation between HSW and SW. The presence of heartwood did not influence the impregnation of CuAz at all.

3. The CuAz penetration of samples dried at 20 °C were the deepest, whereas those dried at higher temperatures were drastically hindered at approximately 4 or 5 cm of depth for both HSW and SW. However, the penetration of samples dried at 100 °C and 120 °C was deeper than that at 80 °C.

4. The occurrence of neutral bordered pits decreased, and exfoliation increased, with higher drying temperatures. The neutral bordered pit numbers of split logs were lower, and exfoliation numbers were higher at each temperature than those of logs.

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