Effects of Drying Temperature for Cryptomeria japonica on the Permeability of Wood Preservative. I: The Permeability of Dried Logs

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Wood preservative treatments are indispensable for wood used in severe environmental conditions. Decay occurs in preservative-treated woods due to the poor impregnation of sapwood; this problem has recently gained attention for Cryptomeria japonica kiln-dried logs. To clarify the causes of this phenomenon, the influence of drying temperature on the penetration of preservative into sapwood logs was investigated. Sapwood samples taken from logs dried at 20 °C to 120 °C were impregnated with copper azole (CuAz). The bordered pits of these samples were observed by scanning electron microscopy (SEM). These results revealed that CuAz absorption decreased with increased drying temperature. The CuAz penetration was deepest for the samples dried at 20 °C. The occurrence of neutral-position bordered pits tended to decrease with increasing drying temperature. These results indicated that there is a strong relationship between the drying temperature and the appearance of bordered pits. Furthermore, the preservative permeability decreased with increasing drying temperature. This result implies that one factor restraining fluid permeability is the aspiration of bordered pits.

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

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INTRODUCTION

Cryptomeria japonica D. Don is the most popular domestic commercial tree in Japan. Effective utilization of wood is important in Japan because the number of mature trees is increasing due to long rotation. C. japonica wood is mainly used in construction for structural timbers (e.g., posts and beams) and interior wood products. To accelerate its abundant consumption and reduce CO₂ emissions, small logs as well as sawn timbers have been used in civil engineering applications as guardrails (Kamiya 2003; Kubojima et al. 2012) and check dams instead of steel and concrete (Noda et al. 2014). However, the introduction of woods for these purposes is not efficient when the costs of building and long-term maintenance are more than those of ordinary materials.

Untreated and non-naturally durable wood materials are easily decayed by fungi, attacked by termites and insects, and degraded by the sunlight when they are exposed to severe environmental conditions. To prevent these effects, it is preferable to treat timbers
with a wood preservative. However, it is difficult to impregnate the timber evenly with the preservative due to its low permeability.

Fluid permeability in heartwood is lower than in sapwood because bordered pits act as obstacles. Bordered pits positioned between tracheids play an important role in transporting air and liquid within the tree (Stamm 1967). The causes of low permeability in heartwood include the aspiration of bordered pits and the incrustation of these pits with heartwood substances (Matsumura et al. 1995; Fujii et al. 1997; Matsumura et al. 2005). Bordered pits aspire when they lose moisture in the intermediate wood, where heartwood starts to form (Nakada and Fukatsu 2012). Additionally, embedded bordered pits in heartwood also hinder fluid permeability, even if the pits are opened (Fujii et al. 1997).

Fluid permeability and impregnation in sapwood is much better than in heartwood (Sandberg and Salin 2012; Sedighi-Gilani et al. 2014). Dried, preservative-treated C. japonica logs decay because of poor impregnation in sapwood; this problem has recently gained attention in Japanese companies. By comparing the preservative penetration in air-dried and kiln-dried C. japonica logs, Momohara et al. (2009) found that the preservative penetration of air-dried logs was significantly better. While poor penetration may be due to the different drying conditions, the details are unclear.

The characteristics of fluid permeability in wood prepared under different drying conditions have been examined. In some reports, the permeability of wood that had been kiln-dried at high temperatures is higher than that of air-dried or conventionally dried wood. Booker and Evans (1994) reported that the higher radial permeability of Pinus radiata D. Don dried at a high temperature is caused by the movement and modification of resin in the canals. Terziev (2002) also compared the effect of drying methods on the permeability of the preservative, finding better penetration for high-temperature dried wood. Further reports found damaged apertures in some of the bordered pits and nano- and micro-checks in the warty and $S_3$ layers of the cell walls (Terziev and Daniel 2002). A rapid rise to a high temperature increases the permeability because of ruptured pits (Zhang and Cai 2008). In contrast, some reports show higher permeability for woods dried at a lower temperature. Comstock and Côté (1968) investigated the gas permeability of eastern hemlock dried at -18 °C, 20 °C, 60 °C, 100 °C, and 140 °C. Higher gas permeability coincided with decreasing drying temperature, resulting from contributions from pit aspiration. According to Thompson (1969), the retention of creosote in southern pine poles dried at 152 °F (67 °C) was slightly higher those dried at 182 °F (83 °C).

These contradictory results are influenced by many factors. Some factors are related to various structures in different wood species, including vessels or resin canals that enhance the permeability of fluids and the stiffness of bordered pits associated with pit aspiration. Other factors include the size of samples, the drying rate, and temperature. For the practical use of a target species, it is important to clarify the reason for low permeability, as different conditions produce conflicting results even within the same species. In this study, the influence of the drying temperature on the preservative penetration in C. japonica sapwood was investigated, and the relationship between the drying temperature and bordered pit aspiration was examined. To eliminate the irrelevant factors derived from dispersion within the same species, corresponding samples were used from end-matched logs from the same tree, and the permeability and bordered pit aspirations were compared for different drying temperatures.
EXPERIMENTAL

Materials

A Cryptomeria japonica tree was harvested from the Kasuya Research Forest of Kyushu University in Fukuoka Prefecture, Japan. The height of the tree was 21 m, and the diameter at breast height was 26 cm. The number of growth rings at breast height was sixty. Seven logs of 50-cm length, labeled from 1 to 7, were derived in sequence above 4.75 m from the ground level (Fig. 1).

Four discs, which were used for measuring the green moisture content, were taken from the same section of the tree. The diameters of the logs at the lowest (No. 1) and highest (No. 7) positions were 21 cm and 19 cm, respectively. The green moisture content of sapwood and heartwood from the four discs was determined from the weight of samples after oven drying. They were averaged from four uneven split small samples without intermediate wood in each place of discs.

Fig. 1. Diagram of the seven logs used for testing different drying temperatures and the four discs used for the green moisture content measurement

Drying of Logs

Both ends of all seven green logs were sealed with epoxy resin to prevent excessive drying from the end grain after the bark was removed from the logs. Two of the seven logs were dried at 20 °C and 65% relative humidity for 20 months in a controlled-environment room. The rest were dried at temperatures of 40 °C, 60 °C, 80 °C, 100 °C, or 120 °C (Table 1) until they reached approximately 10% of the original moisture contents, which were calculated from the green moisture content of the discs. The weights of the logs were measured daily.

After the logs were dried, they were stored at 20 °C and 65% relative humidity, where the equilibrium moisture content was around 12%. The drying rate of each log was calculated from the change in moisture content, with the final moisture content being determined from 1-cm thick discs removed from all logs.
Table 1. Drying Conditions of *C. japonica* Logs

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Temperature (°C)</th>
<th>Relative Humidity (%)</th>
<th>Duration of Drying (d, days; m, months)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>40</td>
<td>62</td>
<td>34 d</td>
</tr>
<tr>
<td>7</td>
<td>60</td>
<td>66</td>
<td>28 d</td>
</tr>
<tr>
<td>5</td>
<td>80</td>
<td>74</td>
<td>24 d</td>
</tr>
<tr>
<td>3</td>
<td>100</td>
<td>80</td>
<td>15 d</td>
</tr>
<tr>
<td>1</td>
<td>120</td>
<td>42</td>
<td>2 d</td>
</tr>
<tr>
<td>2</td>
<td>20</td>
<td>65</td>
<td>20 m</td>
</tr>
<tr>
<td>4</td>
<td>20</td>
<td>65</td>
<td>20 m</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 the logs. Twelve clear sticks were selected from each log. Different sized samples were derived from these sticks: 15-cm samples for permeability experiments, a sample several centimeters in size for the moisture content measurement when the wood preservative was absorbed, and small samples for scanning electron microscopy (SEM) (Fig. 2).

![Fig. 2. Preparation of samples](image)

Treatment of Samples by Wood Preservative

After the volume of each sample was measured, one cross surface was left for liquid penetration, and five surfaces of the samples were sealed with epoxy resin. The copper azole (CuAz, Lonza, Tokyo, Japan) wood preservative in which the concentration of copper(II) oxide and cyproconazole were 11.6% and 0.139% was diluted 1:30 with distilled water, and the wood penetration area was distinguished by a color change. Samples were placed in a vacuum chamber (1.3 × 10³ Pa) for 5 min, and then the preservative was added to the chamber for 5 min of soaking under normal atmospheric pressure. This moderate condition allowed the liquid to sufficiently penetrate throughout the 15-cm length of the samples. The amount of impregnation was calculated from the weights of samples before and after absorption.

Measurement of Penetration Area

Samples impregnated with CuAz were cut every 1 cm from the cross surface where the liquid penetrated. A 0.5% solution of Chrome Azurol S (MP Biomedicals, Santa Ana, CA, USA) in 1% sodium acetate was used to reveal the penetration area.
The penetration area was calculated by the binary method with winROOF software ver.6.3.0 (Mitani Corporation, Tokyo, Japan) after each cross surface was scanned with a flatbed scanner (GT-X970, Seiko Epson Corporation, Suwa, Japan).

**SEM Observation of the Bordered Pits**

Fluid flow is influenced by bordered pit aspiration. The aspirated conditions of the bordered pits for the different drying temperatures were compared via SEM. Two or three samples were collected from the rest of the sticks for each drying temperature (Fig. 2). After being sputter coated with gold, the bordered pits in the earlywood on the radial surface were observed by SEM (JSM 5600LV, JEOL Ltd., Akishima, Japan). The bordered pits were placed into five categories: (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 (200 to 700 for each sample). Freeze-dried bordered pits in green condition were also observed and counted to confirm the aspiration in the green condition.

**RESULTS AND DISCUSSION**

**Moisture Content**

The green moisture content of the four sapwood and heartwood discs (Fig. 1) is shown in Table 2. The average moisture content in sapwood and heartwood was 185.1% and 46.7%, respectively. As shown by the standard deviation of 19.88 for sapwood and 1.21 for heartwood, there was little variation in the moisture condition of different logs.

<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>Average</td>
</tr>
<tr>
<td></td>
<td>Standard deviation</td>
</tr>
<tr>
<td>Heartwood</td>
<td>Average</td>
</tr>
<tr>
<td></td>
<td>Standard deviation</td>
</tr>
</tbody>
</table>

**Drying of Logs**

The change in moisture content during drying is shown in Fig. 3. The moisture content of the two logs dried at 20 °C decreased slowly, reaching approximately 11% after 20 months had passed. However, the moisture content of the log dried at 120 °C quickly reached below 10% within 48 h. Thus, logs dried faster at higher temperatures.
Fig. 3. Changes in moisture content during drying. The upper graph includes drying temperatures from 20 °C to 100 °C and drying time measured in days; the lower graph shows the drying temperature of 120 °C and drying time measured in hours.

Impregnation and Penetration of CuAz

CuAz was absorbed from the one cross surface, under the condition where the other five surfaces were sealed by epoxy resin. The amount of impregnation calculated from the difference in weight before and after the absorption for all drying temperatures is shown in Table 3. The condition of the impregnation in this study was much milder than the actual treatment, in order to clarify the differences in absorption for the different drying temperatures. The average moisture contents of four randomly selected samples were 9.7% for 20 °C and 40 °C, 9.5% for 60 °C, 9.1% for 80 °C, 8.3% for 100 °C, and 6.8% for 120 °C. The samples dried at 100 °C and 120 °C were excessively dried. Thus, impregnation decreased with increased drying temperature. The highest amount of CuAz impregnation was around 400 kg/m³ for the samples dried at 20 °C. The lowest amount of impregnation was less than 100 kg/m³ for the samples dried at 100 °C and 120 °C; as determined by t-test, there was no statistical difference between these two samples.

Table 3. Impregnation of CuAz

<table>
<thead>
<tr>
<th>Drying temperature (°C)</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 (kg/m³)</td>
<td>431.5 (29.4)</td>
<td>412.3 (42.2)</td>
<td>266.4 (49.6)</td>
<td>167.6 (44.2)</td>
<td>139.0 (37.9)</td>
<td>84.8 (11.1)</td>
<td>91.4 (23.1)</td>
</tr>
</tbody>
</table>

The distribution of CuAz and the percentage of the stained area of samples that were cut every 1 cm are shown in Figs. 4 and 5, respectively. The CuAz was stained with Chrome Azurol S. For different drying temperatures, differences in the stained area as a function of depth into the sample are obvious, though the stained area of the cross surface

at 1 cm for all of the drying temperatures was approximately 100%. For the logs dried at 20 °C, at which temperature the most CuAz was absorbed, the penetration area remained high. The penetration area was above 70% even at a depth of 10 cm. However, the penetration areas for the logs dried at 100 °C and 120 °C, which showed lower absorption, were only half at 3-cm depth. At 6-cm depth, there was very little penetration. From the results of the impregnation and penetration, it is clear that higher drying temperatures restrain the liquid flow in logs of dried *C. japonica*.

Fig. 4. Penetration of preservative on the cross surface at 1-cm intervals. Representative samples were chosen. Stronger color shows the penetration area; 0 cm indicates the place where liquid penetrated.

Fig. 5. The penetration area of CuAz in cross cut surfaces at 1-cm intervals from the penetrative surface. The averages of 12 samples are shown.
Observation of Bordered Pits

Bordered pits on the radial surface were classified into five categories, as shown in Fig. 6. The rates of appearance by total counts were calculated in each category. The proportion of aspirated bordered pits is shown in Fig. 7. The proportions of bordered pits exhibiting the (a) neutral position for the 20 °C drying temperature were 25.3% (No. 2) and 21.6% (No. 4), compared with above 90% for the freeze-dried bordered pits in the green condition, which are assumed to show the highest fluid flow. However, the rates of neutral position for the samples dried at 100 °C and 120 °C were only 2.1% and 3.6%, respectively. The higher rates of (c) aspiration, which is assumed to be an obstacle for the transport of fluid, were 33.8% for 100 °C and 76.7% for 120 °C, although the rates of aspiration for the samples dried at 20 °C were 22.5% and 19.5%.

As previously mentioned, the results on the effect of drying temperature on liquid/gas permeability in woods have been contradictory. In this study, higher drying temperatures inhibited the absorption and permeability of the wood preservative, which was mostly due to pit aspiration (Comstock and Côté 1968). The condition of bordered pits affects the fluid flow; in particular, aspirated and incrusted bordered pits remarkably restrain the liquid and gas flow against open pit permeability. According to Fujii et al. (1997), air permeability shows an obvious negative correlation to the pit aspiration ratio derived from air-dried sapwood, air-dried transwood, air-dried heartwood, freeze-dried sapwood, freeze-dried transwood, and freeze-dried heartwood, where aspiration widely varied between wood types. This study confirmed that pit aspiration was caused by drying because the ratios of the neutral positioned bordered pits were 25.3% and 21.6% for the samples dried at 20 °C, compared with above 90% for the freeze-dried condition.

Fluid permeability clearly distinguished the aspirated/incrusted pits from the open ones, whereas the influence of an ambiguous position of the bordered pits on fluid flow is unclear, as shown in Figs. 6(b), 6(d), and 6(e). There has been little research on the permeability of fluid through ambiguous positions, while most research focuses only on aspirated/incrusted or open pits. Matsumura et al. (2005) described this ambiguous condition as an incompletely aspirated pit but did not include them in the category of aspirated pits. If it were hypothesized that the fluid flow is restricted to the area of the pit aperture, it would be assumed that the condition (b) in Fig. 6 is a somewhat fluid-permeable condition similar to the neutral position (a), as fluid could pass through the pit aperture even in the ambiguous position. In contrast, it is assumed that the bordered pits in conditions (c) and (d) successively restrain liquid flow compared with (a) and (b), though some liquid passes through the rupture along the pit aperture in (d) and (e). When the bordered pits are divided into two categories of high or low permeability, the former contains conditions (a) and (b), and the latter, (c), (d) and (e). From the result of the probability of occurrence of different types of bordered pits in Fig. 7, the total appearance rates of the former, (a) and (b), decreased and the total appearance rates of the latter, (c), (d) and (e), increased with increased drying temperature. This phenomenon strongly correlates with the results of the penetration and absorption of CuAz.

The trigger for bordered pit movement is its stiffness and the degree of external force. According to Lü et al. (2007), one reason for pit aspiration is that a high temperature decreases the rigidity of the pit membrane; in this study, the rigidity decreased with increasing drying temperature. The other cause of aspiration is the rapid evaporation of water at a high temperature, and the surface tension causes the pit torus to move to the aspirated position.
Fig. 6. Five types of bordered pit of *C. japonica*. (a) Neutral position, (b) between neutral position and aspiration, (c) aspiration, (d) crack appearance along pit aperture, and (e) exfoliation along pit aperture
Fig. 7. The relative occurrence probability of bordered pits

The change in moisture content during drying by day is shown in Fig. 8, which was derived from Fig. 3. The lowest degree of inclination was shown in a log dried at 20 °C. Hence, when the drying temperature was high, the degree of inclination was large. This result implies more rapid evaporation or faster water movement within wood fiber with an increased drying temperature. Taken together, the data suggests that bordered pits aspirate at a higher drying temperature.

Fig. 8. The change in moisture content of logs per day. Only two measurements were performed at a drying temperature of 120 °C because of the short drying time
SEM images showed no remarkable micro-cracks near the bordered pits or cell walls, which could otherwise have contributed to gas/liquid permeability. Cracks along the pit aperture (Figs. 6(d) and 6(e)) were associated with damage due to drying and were observed at a higher drying temperature. However, no effect on the permeability was determined in this study. This phenomenon may assume nascent damage under high temperature drying. It is possible that fine cracks emerge that have no effect on the liquid permeability under the low drying rate because the greater log dimensions disperse heat more evenly than in sawn timber. Severe factors that accelerate drying, such as a higher temperature or smaller samples, affect the wood fibers, allowing damage to occur in the structure of the wood and improving fluid permeability.

CONCLUSIONS

1. The absorption of CuAz decreased with increasing drying temperature. The highest amount of CuAz impregnation was in the samples dried at 20 °C. The lowest amount of impregnation was in the samples dried at 100 °C and 120 °C, for which there was no statistically significant difference.

2. The differences in the CuAz stained area into the depth of the sample were obvious among the drying temperatures. The penetration was the deepest for the samples dried at 20 °C, and the area of penetration on the cross surface remained high. The penetration area on the cross surfaces dried at 100 °C and 120 °C was less, and there was little penetration at a 6-cm depth from the surface where the liquid was absorbed.

3. The probabilities of a neutral and between neutral and aspiration position of a bordered pit, which were assumed to fluid-permeable condition, inclined to decrease with increased drying temperature.

4. There was a strong relationship between the drying temperature and the appearance of bordered pits in C. japonica logs. Additionally, the permeability of the preservative decreased with increased drying temperature. This result implies that the aspiration of bordered pits restrains fluid permeability.

ACKNOWLEDGMENTS

The authors thank the staff of the Kasuya Research Forest of Kyushu University for providing materials and Kyushu Mokuzai Kougyou Co., Ltd. for the gift of CuAz. This work was supported by JSPS KAKENHI Grant Number 24248031.

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Article submitted: December 16, 2015; Peer review completed: March 18, 2016; Revised version received: March 22, 2016; Accepted: April 3, 2016; Published: April 13, 2016. DOI: 10.15376/biores.11.2.4781-4793