Discoloration Control and Durability Improvement of Cedar Wood through High-temperature and Hot-water Treatments

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Abstract

Korean cedar tree frequently experiences a darkening phenomenon in which its normally red heartwood turns black. Moreover, while cedar wood is known as a resistant species of trees against decay, its sapwood is weak against decay, in contrast to its heartwood. The blackening phenomenon reduces the esthetic value of the wood, and the considerable durability difference between the sapwood and the heartwood create an unpredictable decay phenomenon. To solve these problems, a heat treatment and a hot-water treatment were applied separately to cedar wood. The color value, moisture content, shrinkage, and mass loss rate were measured during the heat treatment. The contact angle of water on the surface of wood that had been heat-treated was also measured after the treatment. The movement of extractives and the constituents of these extractives in the wood during the hot-water treatment were analyzed through a yield estimation test and via a GC/MS analysis. Heat treatment in excess of 180°C had an effect on the color unification properties between sapwood and heartwood, as it changed the color of the surface of the cedar. In addition, it was found the heat treatment caused an increase in the hydrophobicity and a reduction of the EMC of the cedar wood. With a hot-water treatment over 70°C, extractives in the heartwood migrate to the sapwood. This movement of the extractives not only reduces the color difference between the sapwood and the heartwood, but also improves the durability of the sapwood.

Keywords: color unification, cedar, heat treatment, hot-water treatment, durability
Introduction

Although the heartwood of Cedar appears usually red color series, blacken parts are often observed. This blackening is known to be developed by oxidation reaction of phenolic substance in the heartwood in weak alkali condition (Kawazumi et al, 1991). In addition, there is report that black heartwood by blackening has larger content of ash, metal ions, and extractives and is often alkali compared with general red series heartwood (Kubo and Ataka, 1998). Then, Abe et al(1994) reported that hydrocarbonate is a substance to change the Cedar heartwood from red to black. The study on color changing mechanism of Cedar blackening was further progressed and reported that the hydrocarbonate was not major substance of the blackening and conversion of Norlignans in the heartwood into a substance appearing black color series by KHCO₃ in alkali condition induced the blackening. And then, Takahashi and Mori(2006) reported that as Sequrin-C among the Norlignans in Cedar heartwood changed into deep purple series substance in weak alkali condition and absorption spectrum resulted that this corresponded with the spectrum of Cedar black heartwood showing peaks at 450 and 525nm, Sequrin-C was the most important substance in the blackening.

Besides, Aydin and Colakoglu(2005) assessed color change of high temperature kiln drying for alder tree and beech. Ishiguri et al(2000, 2003) reported that heating and smoke-heating induced acidification of Cedar to inhibit blackening of Cedar and make its color into lighter (yellow-white). Studies concerned with wood color change has been continued also in other species, and especially a study on wood color control among essential processes for color unification to make wood high value added is being progressed actively. For instance, studies on wood color change according to internal temperature, water content, and extractives ingredients of maple tree (Yeo and Smith, 2004) and Pitch Pine (Yeo et al. 2007) during kiln-drying have been performed. As a study relating to wood color control by heat treatment, changes in color and surface roughness of alder tree and beech by kiln-drying at 110 °C and 180 °C have been reported (Aydin and Colakoglu, 2005). The high temperature heat treatment of wood is spotlighted as an eco-friendly antiseptic and anti-insect treatment technology by increase of concern for environmental load and human harmfulness of chemical antiseptic and insecticide agent. Militz (2002) reported that when proper process control was done according to tree species within temperature range between 160 ~ 260 °C, increase of dimensional stability and decay resistance is possible, as results of comparison assessment on high temperature heat treatment processes in Finland(Thermo Wood process), Netherland (Plato process), France (Rectification process and Boisperdure process), and Germany (OHT-Oil Heat Treatment process). Also, it was also reported that French type heat treatment for pine tree, fir tree, and beech improved durability, but reduced by about 10~50% of flexural strength (Kamdem, 2002). Besides, Esteves et al(2007) reported that when eucalyptus was treated with heat at 170~200 °C dimensional stability increased and strength decreased by decrease of equilibrium moisture content and mass loss.
Material & Method

Material
Cedar boards 150mm (width, radial direction) × 50mm (thickness, tangential direction) × 3700mm (length, longitudinal direction) in size, were created by lumbering to locate black heartwood on one end, normal heartwood in the center, and sapwood on the other end in the width direction. The cedar tree was cut down on Jeju Island in Korea.

Heat treatment
An air-dried cedar board was manufactured into sample pieces 150mm (width) × 50mm (thickness) × 200mm (length) in size. 6 sample pieces were treated at 160, 180, 200, and 220 °C, respectively. In addition, pieces were painted on their end cross-section to prevent rapid movement of moisture and heat through the section under the substantial processing manufacturing condition generally used to create longer boards. The treatment time was 6 hours. The mass change, linear length change, color value of the black heartwood and the sapwood, and the overall surface color change per hour were measured.

In order to assess the hydrophobic properties of the heat-treated wood surface, the contact angles of 3 sections (a cross-section, a tangential section, and a radial section) against distilled water were measured using a contact angle analyzer (Surface Electro Optics Inc., Phoenix 330). The contact angles 5 sec after contact with distilled water were measured and the average values were obtained after five repeated measurements at each temperature. These values were then compared and assessed.

Hot-water treatment
In the case of cedar, extractives in the heartwood cause color and durability differences between the heartwood and the sapwood. Thus, the movement of extractives in the heartwood from black heartwood to sapwood through the hot-water treatment enhanced the wood color unification and decay resistance of the cedar. At this time, both end cross-sections of the sample were sealed with a double wrapping of plastic wrap to prevent rapid secession of the extractives. In each temperature condition of 70, 80, and 90 °C, the color values of the black heart wood and the sapwood in the water bath were measured per hour for 6 hours.

To identify that the extractives in heartwood had moved into the sapwood, a GC/MS (Agilent, 6890) analysis was conducted. After grinding untreated air-dried black heartwood and sapwood of cedar and hot-water-treated black heartwood and sapwood into 400-mesh powder, the GC/MS analysis was performed. The extract content by regions and types of extractive that moved from the black heartwood to the sapwood by hot-water treatment were identified.

To confirm the effect of the hot-water treatment on the decay resistance, 6 cubes of air-dried untreated sapwood and hot-water-treated cedar sapwood 20mm x 20mm x 20mm in size (a total of 12 cubes) were prepared. Each of the three samples was put into a culture bottle in which Fomitopsis palustris and Trametes versicolor were cultivated to perform the decay test, which
lasted 12 weeks. After 12 weeks, the fungus body was removed from the samples and the mass reduction rate (%) was calculated to evaluate the decay resistance effect of the treatment.

**Measurement of the color value and difference in the color value**

To increase the reliability of the color measurement, the L*a*b* color values of 6 sides of black heartwood and sapwood cubes were measured with a spectrophotometer (Konica-Minolta, CM-2600d) three times successively per hour. The average value of 18 measurement values was then used as a representative value. The color difference between the black heartwood and the sapwood was determined by \( \Delta E \) expressed as the distance between 2 points in the L*a*b* 3-d chroma color space.

\[
\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}
\]

\[
= \sqrt{(L_{\text{heart}} - L_{\text{sap}})^2 + (a_{\text{heart}} - a_{\text{sap}})^2 + (b_{\text{heart}} - b_{\text{sap}})^2}
\]

As the color difference between the heartwood and the sapwood before treatment varied, in order to revise this, the color difference reduction rate was defined as the “loss of color difference / color difference between the heartwood and the sapwood before treatment x 100” and the color difference reducing effects at each temperature were compared.

\[
\text{Color difference reduction rate} = \frac{\Delta E^{(\text{before})} - \Delta E^{(\text{after})}}{\Delta E^{(\text{before})}}
\]

**Result**

**Color value of heat- and hot-water-treated wood and the color difference reduction rate**

As a result of the heat treatment at 220°C, it was found that the color value difference between the black heartwood and sapwood (\( \Delta E \)) decreased by 24.21 from 27.37 before the treatment to 3.16 after the treatment, reducing the black heartwood to the same color as the sapwood. For the heat treatment at 200°C, it was reduced by 18.79 from 32.76 to 13.97, and at 180°C, it was reduced by 15.42 from 39.13 to 23.71. However, the heat treatment at 160°C resulted a reduction of 3.26 from 25.89 to 22.63. The color difference of the black heartwood and the sapwood before and after the treatment was nearly identical. It was identified that there was little wood color control effect below 160°C. The color difference reduction rates by a heat treatment under each condition of 220°C, 200°C, 180°C, and 160°C were 88.45%, 57.36%, 39.41%, and 12.59%, respectively.

The results of the 70°C hot-water treatment showed that the color value difference between the black heartwood and the sapwood (\( \Delta E \)) decreased by 11.95 from 32.43 before the treatment to 20.48 after the treatment. In the heat treatment at 80°C, it was reduced by 16.15 from the initial
value of 37.96 to 21.81, and in the heat treatment at 90 ℃, the result was a reduction of 15.93 from the initial value of 36.55 to 20.62. It was identified that the color difference reducing effect depending on the temperature gap was relatively small. There were no significant differences between the results at temperatures of 70 ℃, 80 ℃, and 90 ℃. Moreover, the hot-water treatment at 90 ℃, 80 ℃, and 70 ℃ resulted in reduction rates of 36.85%, 42.54%, and 43.58%.

![Graphs showing color difference and color difference reduction rate by the heat treatment and the hot-water treatment.](image)

**Figure 1.** Color difference and color difference reduction rate by the heat treatment and the hot-water treatment, (a) color difference by a high-temperature heat treatment, (b) color difference reduction rate by a high-temperature heat treatment, (c) color difference by a hot-water treatment, (d) color difference reduction rate by a hot-water treatment

### Changes of moisture content, mass loss, and line/volume shrinkage by heat treatment

When a heat treatment was introduced as a practical process, more rapid moisture movement and a higher mass loss at a higher temperature were noted. The moisture content and mass loss rate (% of mass loss against the mass of the initial sample) were measured to determine
the appropriate treatment time. During the heat treatment at 220°C, the MC of the specimen reached 0% after 4 hours. In addition it reached 0% after 5 hours at 200°C. Thus, a superior drying effect was determined. However, for the heat treatments at 220°C and 200°C, continuous mass loss, even after 0% of water content, was confirmed. Hence, it was judged that this result can be attributed to the deterioration of cellulose and hemicellulose in the wood due to the high temperature. In addition, although surface splitting occurred on the section by drying stress due to the rapid moisture movement, the degree of this splitting was not severe. As a result, it is expected that an oven heat treatment process can be used to shorten the drying time and as a process of wood color control for cedar.

Table 1. Mass loss and MC by heat treatment

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Mass Loss(%)</th>
<th>Start</th>
<th>1hours</th>
<th>2hours</th>
<th>3hours</th>
<th>4hours</th>
<th>5hours</th>
<th>6hours</th>
</tr>
</thead>
<tbody>
<tr>
<td>200°C</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oven-heat</td>
<td>Mass Loss(%)</td>
<td>0</td>
<td>6.56</td>
<td>13.14</td>
<td>18.83</td>
<td>22.44</td>
<td>24.68</td>
<td>26.38</td>
</tr>
<tr>
<td></td>
<td>MC(%)</td>
<td>35.26</td>
<td>26.38</td>
<td>17.49</td>
<td>9.79</td>
<td>4.91</td>
<td>1.88</td>
<td>0</td>
</tr>
<tr>
<td>220°C</td>
<td>Mass Loss(%)</td>
<td>0</td>
<td>8.73</td>
<td>16.42</td>
<td>24.00</td>
<td>28.57</td>
<td>31.30</td>
<td>33.01</td>
</tr>
<tr>
<td>Oven-heat</td>
<td>MC(%)</td>
<td>43.95</td>
<td>31.38</td>
<td>20.31</td>
<td>9.40</td>
<td>2.82</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

With an increase in the treatment temperature of the heat treatment in an effort to control the wood color of cedar, it was found that the shrinkage in the thickness, width and length directions increased. However, the shrinkage levels in the thickness direction and in the width direction were much greater than that in the length direction. The shrinkage in the length direction was slightly less than 0.5%, but the shrinkage in the thickness and width directions differed depending on whether an edge grain or flat grain was processed during the lumbering process. Moreover, volume reduction by linear shrinkage also developed, and it was found that the volume shrinkage increased with an increase in the treatment temperature. This increase was 6.93% for the heat treatment at 220°C. It is expected that the observed linear/volume shrinkage in an oven heat treatment may be useable as data for lumbering dimensions and yields to obtain accurate dimensions of the final wood product in practice.
Increase of hydrophobicity of heat-treated wood

The contact angle of water on a tangential section of an untreated sample obtained from an image captured by the contact angle analyzer was 0°, indicating that it was hydrophilic. However, the contact angles on a tangential section of heat-treated samples at all temperatures were greater than 45°. This shows that their hydrophobicity values increased.

It was confirmed that all contact angles on three sections of untreated air-dried samples were 0°, indicating that they were hydrophilic. However, all contact angles on three sections of heat-treated samples were increased compared to those of untreated samples. This shows that the heat treatment increased the hydrophobicity of cedar. Through these results, it is expected that a heat treatment can increase the decay resistance due to the reduction of the water absorption characteristics according to the increase in the hydrophobicity.

Movement of extractives by hot-water treatment

To confirm the movement of the extractives, the yields of volatile extractives in untreated sapwood and 80°C hot-water treated sapwood were measured. Figure 4 shows that the extractives yield of hot-water treated sapwood is 3.41%, two times higher than 1.74%, the extract yield of untreated sapwood. This indicates that the extractives in the heartwood moved to the sapwood.
To confirm the movement of extractives from heartwood to sapwood by the hot-water treatment, a GC/MS chemical analysis was performed. As a result, it was identified that terpene series extractives of cadinene and copeane, which did not exist in the untreated air-dried sapwood, existed in the hot-water treated sapwood. In Figures, the Y axis indicates the amount of the chemical. The Y axis scale of the hot-water treated sapwood shows 4,500,000, 10 times higher than 450,000, the scale of the untreated sapwood.
Decay resistance of hot-water treated wood

Figure shows the results of the decay resistance test (KS F 2213) using the air-dried sapwood and the 80°C hot-water treated sapwood. While the untreated air-dried sapwood resulted in a mass loss of 21% by *Fomitopsis palustris* (FPA) and 36% by *Trametes versicolor* (TRA) at 12 weeks, the 80°C hot-water treated cedar sapwood resulted in 7% and 10% mass loss by each respective fungus. From the 12-week decay resistance test, it was determined that the decay resistance results of an oven heat-treated sample at 200°C far exceeded those of untreated samples, at 67% \[\{(21\%-7\%)/21\%\} \times 100\%\] in the case of *Fomitopsis palustris* (FPA) and 72% \[\{(36\%-10\%)/36\%\} \times 100\%\] in the case of *Trametes versicolor* (TRA). Through these results, it was identified that the hot-water treatment increases the decay resistance of sapwood.

The color difference reduction rates after the six-hour heat treatment were 39%, 57% and 88%, respectively, at 180°C, 200°C, and 220°C. Color unification of cedar wood was accomplished by a heat treatment at the appropriate treatment level. In addition, the results showed a hydrophobicity increase and a reduction of the EMC on the surface of the cedar wood due to the heat treatment. As a color control effect after 6 hours of a hot-water treatment, it was found that there were 37%, 43% and 44% color difference reductions, respectively, at 70°C, 80°C, and 90°C. In addition, it was determined that the terpene-type extractives in the heartwood were moved to...
the sapwood as a result of the hot water. Moreover, it was identified that the decay resistance of hot-water-treated sapwood against brown-rot fungi and white-rot fungi were improved by as much as 67% and 72%, respectively, compared that of untreated sapwood. Through the results of this study, it is expected that the heat and hot-water treatment can be applied as a color control process to reduce the color difference between the heartwood and sapwood, which is a major cause of the undervaluing of this wood. This is an eco-friendly preservative process that does not involve a chemical treatment for enhancing the relatively low decay resistance of sapwood.

**Current research**

Despite the success of this study, a separate application of a hot water treatment and a high-temperature treatment is associated with a process time loss, energy loss and uncontrollable carbonization. To overcome these limitations, we designed and created equipment that could undertake the heat treatment and hot-water treatment continuously in a pressure chamber. First, cedar was treated in hot water under an alternative pressure (8~0.5atm) to migrate the extractives from the heartwood to the sapwood. After the hot-water treatment, a high-temperature treatment over 200°C was conducted for control of the surface and to increase hydrophobicity. Through this complex process, the durability of the cedar increased more than it did through separate processes. In addition, the color differences of the cedar between the sapwood and the heartwood both on the surface and in the inside the wood were decreased by this type of treatment.

**References**


