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Effects of impregnation and heat treatment on the physical and mechanical properties of Scots pine (Pinus sylvestris) wood

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Abstract

Wood modification, of which thermal modification is one of the best-known methods, offers possible improvement in wood properties without imposing undue strain on the environment. This study investigates improvement of the properties of heat-treated solid wood. Scots pine (Pinus sylvestris) was modified in two stages: impregnation with modifiers followed by heat treatment at different temperatures. The impregnation was done with water glass, melamine, silicone, and tall oil. The heat treatment was performed at the temperatures of 180°C and 212°C for three hours. The modified samples were analyzed using performance indicators and scanning electron microscope micrographs. The mechanical and physical properties were determined with water absorption, swelling, bending strength, and impact strength tests. All the modifiers penetrated better into sapwood than hardwood; however, there were significant differences in the impregnation behavior of the modifiers. As regards the effect of heat treatment, generally the moisture properties were improved and mechanical strengths impaired with increasing treatment temperature. In contrast to previous studies, the bending strength increased after melamine impregnation and mild heat treatment. It is concluded that the properties of impregnated wood can be enhanced by moderate heat treatment.

Keywords: Impregnation, mechanical properties, physical properties, Scots pine, thermal modification

Introduction

Wood is a widely used natural structural material in many applications. The modification of wood, for example, by impregnation or thermal treatment, is a possible option for improving the performance of wood material, thus permitting its utilization in a wider variety of applications. Impregnation modifies wood properties by filling the wood substance with an inert material. Thermal modification causes chemical changes in the wood material by heat (Hill 2006).

The exposure of wood to a controlled elevated temperature has been known to modify its properties since the mid-twentieth century (Stamm et al. 1946). The thermal modification of wood reduces its hygroscopicity (Borrega and Kärenlampi 2010, Hill et al. 2012), thus improving its dimensional stability (Korkut and Bektas 2008, Kocaefe et al. 2008a). The color of thermally modified wood is darker (Esteves et al. 2011, Poncsak et al. 2011, Akgül and Korkut 2012) and the strength properties are reduced (Kocaefe et al. 2010, Pfiem et al. 2010, Rowell et al. 2013). The treatment of wood at elevated temperatures causes mass loss because wood cell wall polymers degrade thermally. Mass loss is an important variable in the heat treatment of wood (Esteves and Pereira 2008). Hemicelluloses degrades earlier than other wood cell wall compounds with increasing temperature (Esteves et al. 2011), and hence hemicellulose degradation has a significant impact on the strength and dimensional stability properties of heat-treated wood (Müller et al. 2003, Weiland and Guyonnet 2003, Kocaefe et al. 2008b, Rowell et al. 2009, Weigl et al. 2012).

Thermal modification is the most advanced wood modification process commercially (Hill 2006) and various thermal modification processes have become widespread over the past decade, for example, the ThermoWood (Finland), Plato (Netherlands), Retification (France), and Oil Heat Treatment (OHT) (Germany) processes. A common factor in these
processes is thermal treatment at elevated temperatures with low oxygen content. The oxygen content is minimized in a variety of ways, for example, by nitrogen in the Retification process or by oil in the OHT process (Rapp 2001, Militz 2008). The ThermoWood (2003) process operates with the presence of water vapor, which also protects the wood. The thermal modification of wood has been studied with various wood species; pine wood is a commonly used material in such research.

To meet social demands, the wood products industry needs new innovative products that are based on environmentally nontoxic agents and methods. Research into novel wood modification approaches contributes to the removal of barriers to new applications for solid wood products. The effects of thermal modification on the properties of pine are well known, but combining such modification with another treatment has not been widely researched. However, some treatment combinations have recently been tested, for example, heat treatment and borate impregnation (Awoyemi and Westmark 2005) and preboiling (Awoyemi et al. 2009) as a pretreatment. Sun et al. (2013) have studied the combination of melamine-urea-impregnation and heat treatment.

The aim of this study is to investigate the influence of impregnation on subsequently heat-treated pine wood. The impregnation pretreatment was performed with four different solutions (water glass, melamine, silicone, and tall oil) and the heat treatment was performed at two temperatures. The impregnation solutions were selected focusing on environmentally nontoxic agents with different compositions. The selected solutions have previously been found to give improved wood properties (Hansmann et al. 2006, Temiz et al. 2008, Ghosh et al. 2009, Pfeffer et al. 2010). The major motivation for this study was enhancement of the mechanical properties of heat-treated wood.

Material and methods

Scots pine (Pinus sylvestris L.) wood samples were first impregnated with the solutions studied, followed by treatment of the impregnated samples. Heat treatment was carried out at two different temperatures: 180°C and 212°C. Density, moisture content, heartwood ratio, and the rate of growth were measured before the treatment. The values are presented in Table I.

The samples were cut and planed into the size 20 × 95 × 1000 mm, and impregnated with four different modifiers. The impregnation was carried out using registered pressure apparatus, at a pressure of 10 bars for 120 min. The capacity of the pressure apparatus is 600 l, and it contains an individual bin into which the test samples are placed with the impregnation solution.

After impregnation, the samples were dried in an oven at 103°C for 24 h, with the exception of the melamine- and tall oil-treated samples which have a more responsive flash point and thus required slightly different treatment. The melamine-treated samples were dried at 90°C for 24 h, and the tall oil-treated samples were allowed to stabilize for a day before drying, ensuring that the responsive agent had evaporated. The result of the impregnation was measured by weight percent gain (WPG).

Impregnation was performed with different modifiers, and each treatment had 15 samples, consisting of both sapwood and heartwood. The modifiers used were water glass, melamine, silicone, and tall oil. Water glass, specifically sodium silicate (ZEOPOL 33), was obtained from J.M. Huber Finland Oy (Hamina, Finland) and diluted 1:1 with water. Melamine (Prefere 70 0592 L) was supplied by Dynea Chemicals Oy (Hamina, Finland). The silicone emulsion was made of silicone (001 7100) from Tikkurila Oy (Vantaa, Finland) and water, mixed at the ratio 1:9. Tall oil (EP608) was acquired from Ekopine Oy (Oulu, Finland) and diluted 1:1 with wood turpentine (T2501) from Kiilto Oy (Lempäälä, Finland). The properties of the impregnation solutions are presented in Table II.

The heat treatment was performed based on the ThermoWood process, but without water vapor in the process. The samples were treated in a heating oven, at temperatures of 180°C and 212°C for 180 min.

Table I. Average properties of samples (62 pieces) before treatment.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value (± standard deviation)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (kg/m³)</td>
<td>447.6 (46.9)</td>
</tr>
<tr>
<td>Moisture content (%)</td>
<td>11.7 (2.5)</td>
</tr>
<tr>
<td>Rate of growth (mm/a)</td>
<td>2.7 (1.5)</td>
</tr>
<tr>
<td>Heartwood ratio (%)</td>
<td>45.7 (43.3)</td>
</tr>
</tbody>
</table>

Table II. Properties of impregnation solutions.

<table>
<thead>
<tr>
<th>Solution</th>
<th>Water glass</th>
<th>Melamine</th>
<th>Silicone</th>
<th>Tall oil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm³)</td>
<td>1.38 ± 0.015</td>
<td>1.19</td>
<td>1.00</td>
<td>0.86</td>
</tr>
<tr>
<td>pH</td>
<td>11.2</td>
<td>9.4-10.0</td>
<td>c. 7</td>
<td></td>
</tr>
<tr>
<td>Viscosity (mPas)</td>
<td>250</td>
<td>8-14</td>
<td>–</td>
<td>4000</td>
</tr>
</tbody>
</table>

*Relative density.
After the heat treatment, the mass loss was determined.

The impregnated heat-treated specimens were examined with a scanning electron microscope (SEM), Jeol JSM-5800 LV. Prior to the analysis, the surfaces of the samples were covered with a layer of gold using a sputter coater. In view of the poor impregnation of the heartwood, only the treated sapwood specimens were analyzed by SEM microscopy.

The moisture properties were determined with thickness swelling and water absorption tests. The size of the test pieces was 20 × 20 × 30 mm. The test pieces were conditioned at 20°C and 65% relative humidity before testing. The weights and dimensions of the pieces were measured before immersion into water for a duration of 28 days. The pieces were periodically taken out of the water, surface-dried with absorbent paper, re-measured, and returned to the water immediately.

Swellings were determined in the radial and tangential directions. Swelling in the axial direction has been noted to be insignificant (Poncsak et al. 2011). Swelling (S) was calculated according to the following formula:

\[ S = \frac{T_1 - T_2}{T_2} \times 100 \]

where \( T_1 \) is the thickness of the specimen after immersion, and \( T_2 \) is the thickness of the specimen before immersion.

The water absorption values (WA) were calculated as follows:

\[ WA = \frac{W_1 - W_2}{W_2} \times 100 \]

where \( W_1 \) is the mass of the specimen after immersion, and \( W_2 \) is the mass of the specimen before immersion.

The mechanical strengths of the heat-treated wood were determined by three-point bending and impact strength tests. The bending strength was determined with a Zwick Roell Z020 testing machine in accordance with ISO 3133. Fifteen test samples of each category, with dimensions 20 × 20 × 380 mm, were tested. Before the test, the specimens were conditioned at 20°C and 65% relative humidity for over 24 h. The impact strength was determined with a Zwick 5102 Model impact tester in accordance with EN ISO 179. The impact strength testing was carried out with 15 samples with the dimensions 4 × 10 × 80 mm. The test pieces were conditioned at 23°C and 50% relative humidity for over 24 h before the impact test was performed.

**Results and discussion**

Results for the control and reference samples are presented below. The control sample was not subjected to elevated temperature heat treatment, only kiln-dried at the temperature of 103°C for 24 h. The reference samples were heat-treated, but not impregnated. The mechanical properties data contain also the results of impregnated samples that were not heat-treated but only kiln-dried as described above.

**Treatment**

The impregnation results are presented in Table III by WPG. Results for the sapwood specimens are presented separately as they clearly differ. Some samples exhibited no impregnation, which, after drying, gives negative results for WPG in a few categories. The results show that the melamine samples were impregnated best, especially in sapwood. The effects of heat treatment are presented in Table IV by mass loss. It can be seen that the higher temperature causes greater mass loss.

The effects of the heat treatment are also presented in Figures 1–5, which show SEM micrographs of the surface of the samples. Cracks can be seen in the samples after heat treatment, especially in the water glass-impregnated sample treated at the higher temperature (Figure 2). Based on Figure 3, melamine seems to have melted into the wood during the heat treatment. The tall oil seems to have boiled during heat treatment at the temperature of 212°C (Figure 5).

It can be seen in Table III that the sapwood specimens have been well impregnated. The solutions penetrate into sapwood better due to a lower proportion of extractives, and because the pit

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Water glass</th>
<th>Melamine</th>
<th>Silicone</th>
<th>Tall oil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average (%)</td>
<td>2.76 (6.24)</td>
<td>38.71 (24.68)</td>
<td>6.85 (8.72)</td>
<td>7.28 (5.29)</td>
</tr>
<tr>
<td>Sapwood average (%)</td>
<td>3.48 (4.75)</td>
<td>57.04 (15.60)</td>
<td>12.09 (6.70)</td>
<td>7.73 (3.26)</td>
</tr>
</tbody>
</table>

Values in parentheses indicate standard deviations.
membranes are not aspirated in sapwood (Archer and Lebow 2006). The excellent impregnation of the melamine samples may be due to the lower viscosity of the modifier. Viscosity has been found to affect the uptake of a solution (Larnøy et al. 2005, Dubey et al. 2011), especially in pine wood (Tondi et al. 2013).

The results show that increased treatment temperature enhances mass loss, as presented in previous studies (Welzbacher et al. 2007, Metsä-Kortelainen and Viitanen 2010). In addition to the temperature, longer treatment time has also been found to increase mass loss (Alén et al. 2002, Esteves et al. 2007). Density, which has been found to correlate with mechanical properties (Mahnert and Militz 2012), is also reduced by heat treatment (Boonstra et al. 2007a, Metsä-Kortelainen and Viitanen 2010). The mass loss in heat treatment is caused by degradation of the hemicelluloses (Esteves and Pereira 2008, Esteves et al. 2011).

Values in parentheses indicate standard deviations.

Table IV. Average mass loss and standard deviations.

<table>
<thead>
<tr>
<th></th>
<th>Reference</th>
<th>Water glass</th>
<th>Melamine</th>
<th>Silicone</th>
<th>Tall oil</th>
</tr>
</thead>
<tbody>
<tr>
<td>180°C</td>
<td>7.49 (0.78)</td>
<td>8.20 (0.93)</td>
<td>8.14 (0.54)</td>
<td>7.21 (0.48)</td>
<td>7.33 (0.53)</td>
</tr>
<tr>
<td>212°C</td>
<td>10.24 (1.00)</td>
<td>13.00 (1.63)</td>
<td>11.00 (0.80)</td>
<td>10.44 (0.78)</td>
<td>11.01 (1.67)</td>
</tr>
</tbody>
</table>

Moisture properties

Data on the moisture properties are presented in Figures 7–10. The water absorption properties are presented as a point chart in Figures 7 and 8, to which a trend line has been added between the points using a two-period moving average. The results for water absorption are presented separately.

The temperature of the heat treatment plays significant role; for instance, the tall oil-impregnated heat-treated samples seem to undergo a boiling effect at the temperature of 212°C (Figure 5c) but not at 180°C (Figure 5b). The melamine samples that were treated at the temperature of 212°C have unusual patterns on the surface of the samples (Figure 6a, 6b) and there are areas with no patterns (Figure 6c). The patterns appear to occur randomly on the surface (Figure 6a, 6b) and resemble condensation on the surface of the wood.
at the different temperatures. The samples treated at
the higher temperature absorbed less water during
the first 7 days of the water absorption test than the
samples treated at the lower temperature. Some
samples absorbed the same amount or more water
than the reference samples regardless of the treat-
ment temperature. The melamine-impregnated
heat-treated samples absorbed least water through-
out the test at both temperatures. The silicone-
impregnated heat-treated samples absorbed water
with restraint at the beginning of the test. Water
glass impregnation followed by heat treatment had a
negative effect on water absorption, i.e. more water
was absorbed than the untreated reference sample.

The swelling properties are presented as bar charts
in Figures 9 and 10. The swelling is presented
separately for the tangential and radial directions.
The higher treatment temperature restrains swelling
in the tangential and radial directions. The benefit of
impregnation can be seen at the lower temperature
in the tangential directions, where all impregnated
heat-treated samples have swelled less than the
reference samples. The samples heat-treated at
212°C swelled similarly despite the impregnation.
The melamine samples swelled most in the radial
direction. By comparison with the data in Table IV,
it can be seen in Figures 7–10 that mass loss is
related to the moisture properties of the wood.
The higher treatment temperature stabilized the moisture properties of the samples, which is in agreement with earlier studies (Welzbacher et al. 2007, Gündüz et al. 2008, Korkut and Bektas 2008, Jalaludin et al. 2010, Cao et al. 2012, Priadi and Hiziroglu 2013). Longer treatment time could restrain moisture absorption, but not as efficiently as increased temperature (Cao et al. 2012, Priadi and Hiziroglu 2013).

Improvement in the moisture-resistance properties with increasing temperature and time is not always unambiguous. Metsä-Kortelainen et al. (2006) noted that the water absorption of pine sapwood increased after heat treatment at the temperatures of 170°C, 190°C, and 210°C compared to normally kiln-dried reference samples when the specimens were floated for 146 hours. Only heat treatment at the temperature of 230°C reduced the water absorption of pine sapwood. In addition, the differences in water absorption between pine sapwood and heartwood were significant in this study, which could be explained by the migration of extractives onto the wood surface (Metsä-Kortelainen and Viitanen 2012). Nuopponen et al. (2003) found that during heat treatment at a low temperature the extractives moved onto the edges of pine sapwood. Extractives were not detected in the sapwood edges after the heat treatment at a higher temperature in this study, which could be due to degradation and evaporation of the extractives. Another option could be softening of the lignin that enables the diffusion of the extractives deeper into the battens (Nuopponen et al. 2003).

In untreated wood, tangential swelling is usually twice that of radial swelling (Rowell 2013), which appears to be valid also in heat-treated Scots pine (Korkut and Bektas 2008). However, the dissimilar anisotropy of different wood species together with the different treatment temperatures and times affects the swelling ratio (Welzbacher et al. 2007, Priadi and Hiziroglu 2013). The melamine-impregnated heat-treated samples swelled about one and a half times in...
the tangential direction compared to the radial direction. The swelling and absorption are lower after heat treatment because the amount of wood hydroxyl groups are reduced (Gündüz et al. 2008) due to the formation of an ether linkage by the splitting of two adjacent hydroxyl groups (Kamdem et al. 2002). Furthermore, hemicellulose degradation in heat-treated wood reduces the hygroscopicity of the wood, because hemicellulose is the hydrophilic component in the wood cell wall (Walker 2006). In addition, depolymerization of hemicelluloses increases the relative proportion of crystalline cellulose and cross-linking of the lignin network, which inhibits the encounter of water and hydroxyl groups (Boonstra et al. 2007b).

**Mechanical properties**

The mechanical properties are presented in Figures 11 and 12. The average results are presented as bar charts with standard deviations added as error bars. In addition, polynomial trend lines have been added to the figures.

The higher temperature usually decreases the bending strength. None of the treated samples achieved the same or better bending strength than the normal kiln-dried sample. Melamine improved the bending strength after heat treatment. Melamine-impregnated samples heat-treated at 180°C had 10.0% better bending strength compared to the reference samples treated at the same temperature. The melamine-impregnated samples heat-treated at 212°C had 21.7% better bending strength compared to the reference sample treated at the same temperature. The bending strengths of the samples impregnated with silicone and tall oil and heat-treated at 212°C were 9.2% and 8.2% better than that of the reference sample after heat treatment at the same temperature. The bending strengths of the water glass-impregnated heat-treated samples were...
reduced close to half after heat treatment at the higher temperature. The water glass-impregnated samples achieved 51.7% of the bending strength of the control sample after 212°C heat treatment. The bending strengths of silicone- and tall oil-impregnated heat-treated samples were not increased, but the reduction appeared to be smaller than that of the reference samples.

The heat treatment impaired the impact strength of the wood. The melamine-impregnated and lower temperature heat-treated samples were able to maintain the same level of impact strength as the melamine-impregnated sample without heat treatment, but it was weaker by a half compared to the control sample. On the basis of Figure 12, it can be concluded that the impact strength of wood is impaired with increasing heat treatment temperature.

Impregnation makes the wood more fragile because the mobility of the cell wall components is reduced by the impregnation solution (Dieste et al. 2008). The bending strength of the melamine-impregnated samples was improved after heat treatment at 180°C. The study of Deka and Saikia (2000) indicated that the bending strength increases by 12–20% due to the thermosetting resin. The study of Boonstra et al. (2007b) gives impact strength results similar to the result for the untreated control samples in this study.
The higher treatment temperature reduced the bending strength, in agreement with previous studies (Kocaefe et al. 2010, Poncsak et al. 2011, Welzbacher et al. 2011, Surini et al. 2012). In addition to temperature, the treatment time and atmosphere affect the bending strength (Rowell et al. 2009), as does the wood species (Shi et al. 2007). For example, hardwood is more sensitive to mechanical changes than softwood (Kamdem et al. 2002), which is thought to be due to lignin condensation (Wikberg and Maunu 2004). A slight increase in the bending strength after mild heat treatment may be due to the decreasing moisture content (Zhang et al. 2013). Kocaefe et al. (2010) have found that heat treatment up to 160°C temperature improves bending strength. Sun et al. (2013) impregnated Eucalyptus pellita wood with melamine–urea–formaldehyde resin and then heat-treated it at different temperatures. They noted that the changes in the mechanical properties were minimal below 200°C, which is in agreement with the present study. The impact strength is strongly dependent on the treatment method. Heat treatment in an air atmosphere decreases the impact strength more than to oil heat treatment or heat treatment in nitrogen air (Kubojima et al. 2000, Rapp 2001). Winandy et al. (1983) state that higher retention of an impregnation solution decreases toughness, which is congruent with our results. Well-penetrated melamine impaired the impact strength of the wood.

Degradation of hemicellulose between microfibrils is the main mechanism of strength loss in wood (Sweet and Winandy 1999), and the reduced strength properties of heat-treated wood are attributed to the loss of hemicelluloses (Rowell et al. 2009). When flexibility decreases, the hemicellulose–cellulose–hemicellulose bond is replaced with a more rigid cellulose–cellulose bond (Boonstra et al. 2007b, Kocaefe et al. 2008b). The reduction in bending strength is correlated with the degradation of hemicelluloses (Weigl et al. 2012). Boonstra et al. (2007b) have characterized that the impact strength is determined by secondary bonds between cellulose and hemicelluloses, therefore degradation of hemicelluloses is also responsible for the decrease in impact strength. The mass losses were similar in every sample at the same temperature, so it can be assumed that the degradation of hemicelluloses is similar after impregnation. The better bending strength of the melamine-impregnated heat-treated samples thus is unlikely to be due to the minor degradation of hemicellulose. In addition to the degradation of hemicellulose, previous studies have assumed that the changed mechanical properties may be explained by the increasing crystalline cellulose content (Boonstra et al. 2007b, Kocaefe et al. 2008b). Furthermore, heat-treated wood has lower plastic ductility, which makes crack initiation easier (Majano-Majano et al. 2012).
Conclusions

The effect on pine wood of impregnation with different modifiers together with heat treatments was investigated in this work. The combination of two treatment methods can improve some properties of wood, but the type of modifier and the treatment conditions affect the results.

On the basis of the tests, the moisture properties of wood were improved by the heat treatment, with greater improvement found at higher treatment temperature. It was found that the mechanical properties decreased after impregnation compared to untreated wood but the impregnation solutions had a favorable effect on the mechanical properties after heat treatment at a low temperature. The most significant property change was the bending strength of melamine-impregnated wood improved with the mild heat treatment temperature.

This study shows that moderate heat treatment of impregnated solid wood can improve, or at least will not reduce, the mechanical properties of Scots pine wood. Solid wood treated in this way may be suitable for various uses, for example, in outdoor use and furniture. The promising results for melamine-impregnated wood suggest that future research should focus on more detailed study of its properties and suitability for various applications.

References


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