DIMENSIONAL STABILITY AND BIOLOGICAL RESISTANCE OF PARTICLEBOARD MADE FROM ACETYLATED PINE WOOD CHIPS

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1 INTRODUCTION

Wood is a preferred material for many applications. As high quality, full size timber is becoming more scarce reconstituted wood products, e.g. particleboards, are finding a growing market. All wood products have the disadvantages, among others, of dimensional instability and susceptibility to biological degradation, which, however, can be controlled by means of chemical modification of the wood constituents (1).

In board products thickness swelling is a great problem. Water sorption causes both irreversible swelling due to release of residual compressive stresses imparted to the board during the pressing process, and reversible swelling due to water uptake in the wood cell wall.

In a preliminary study wood flakes were made to react with liquid acetic anhydride diluted with xylene, and made into flakeboard (3). The purpose of the present study was to perform the acetylation with acetic anhydride vapor, which eliminated the organic cosolvent used in the earlier study, and greatly simplified the recovery of excess chemicals.

Particleboards made from vapor phase acetylated chips were evaluated with respect to rate and extent of swelling, equilibrium moisture content and biodegradeability. For comparison boards were also made from liquid phase acetylated chips.

2 EXPERIMENTAL

2.1 Wood chips

Commercial board chips of pine wood (Pinus Sylvestris) containing a small amount of bark were used. The chips were passed over a 3 x 3 mm screen to remove fines, and oven-dried at 105°C before use.

2.2 Reaction of chips with acetic anhydride

The chips (200 g) were placed in a stainless steel reactor in a mesh container to keep them off the bottom of the reactor. A vacuum was applied for 1 h at 105°C, then 120 ml of acetic anhydride was drawn inside the bottom of the reactor. The temperature was raised to 120°C and maintained for various lengths of time, from 4 to 48 h. After the reaction, the excess acetic anhydride, together with by-product acetic acid, was drained from the reactor and a vacuum applied for 1 h at
105°C. The chips were removed from the reactor and oven-dried for 24 h at 105°C. The weight gain was calculated as a percent by weight based on oven-dried (o.d.), untreated chips.

Oven-dried chips were also treated with refluxing acetic anhydride/xylene (1/1, v/v) in a glass reactor equipped with a condenser. After a reaction time of 0.5 to 48 h, the excess chemical was drained from the reactor and a vacuum applied for 1 h at 105°C.

2.3 Particleboard making

Two different adhesives were used to make particleboards: phenol/formaldehyde (PF) and melamine/urea/formaldehyde (MUF) resin.

Phenol/formaldehyde adhesive

Treated chips (180 g o.d. chips) were pressed into a board approximately 1.25 x 15 x 15 cm in size. Each board was made with a density of approximately 640 kg/m³, with a resin solids content of 6% (based on o.d. treated chips). The adhesive used was a 43.5% aqueous solution of a PF resin. No catalyst or wax was added. The mat moisture content was 12 to 13%. Pressing lasted for 10 min at 177°C.

Melamine/urea/formaldehyde adhesive

Treated chips (196 g o.d. chips) were pressed into a board approximately 1.0 x 17.5 x 17.5 cm in size. Each board was made with a density of approximately 700 kg/m³, with a resin solids content of 8% (based on o.d. chips). An aqueous solution of an MUF resin was used, with a resin concentration of 59%. 3.1% (based on dry resin weight) NH₄Cl was added as a catalyst. The mat moisture content was about 5%. Pressing lasted for a total time of 2 min at 190°C.

2.4 Water swelling rate tests

Each particleboard specimen to be tested was placed in a 10 x 10 cm container, 5 cm deep. The container was on a flatbed micrometer for the thickness measurements. Water was added to the container, and the thickness was recorded periodically for 5 days.
2.5 Water soaking tests

Repeated water soaking tests were run as previously described (2). Each cycle consisted of water soaking for 5 days followed by oven-drying for 2 days. Six cycles were run. Thickness swelling was calculated as a percentage of the original thickness (o.d. board).

2.6 Humidity tests

Oven-dried specimens were placed in constant humidity rooms at 30, 65 and 90% relative humidity (RH) and 27°C. After 21 days the specimens were weighed and the equilibrium moisture content (EMC) was determined.

Separate specimens were placed in a humidity room at 90% RH and 27°C. The thickness was determined after 21 days. The specimens were then placed in a humidity room at 30% RH and 27°C for another 21 days, whereafter the thickness was measured. This procedure was repeated for a total of four cycles of 90 to 30% RH. The specimens were then oven-dried and the thickness determined. Changes in thickness were determined as a percentage of the original thickness (o.d. board).

2.7 Exposure in unsterile soil

Specimens were incubated for five months at approximately 25°C in mist unsterile soil. Among the microorganisms present in the soil were brownrot and softrot fungi and wood destroying bacteria.

3 RESULTS AND DISCUSSION

The reaction in refluxing liquid was much faster than the vapor phase reaction. A weight gain of 17.5% was obtained in 4 h in the refluxing liquid, while in the vapor phase reaction it took 21 h to obtain the same weight gain. However, the same weight gain, 24%, was obtained for both treatments in 48 h.

When chips which had been acetylated to various weight gains in the vapor phase system were subjected to water soaking, the amount of water-soluble components decreased with increased level of acetylation, with the highest value found at a weight gain of 6.7% (fig.1). At weight gains >10%, the amount of leachable components was lower than it was for untreated chips. It is possible that up to a certain treatment level, acid
hydrolysis of wood components produced water-soluble compounds, while on further acetylation these were transformed to less hydrophilic substances. The greatest amount of leachable carbohydrates was also found at a weight gain of about 7%.

Fig. 1. Weight loss and amount of water-soluble carbohydrates based on the weight of o.d. treated wood, as a function of the weight gain.

○ Weight loss, △ water-soluble carbohydrates

(The chips were leached in distilled water for 4 days and the weight loss was determined. The leachate was hydrolyzed in 0.5 M H2SO4 for 4 h at 100°C. The carbohydrates were determined as monosaccharides by liquid–partition chromatography).

Two different adhesives were used, both commonly used for exterior wood applications. The PF adhesive is more water resistant than the MUF adhesive. The level of MUF adhesive used was approximately 2/3 of that used in the particleboard industry, while the level of the PF adhesive was about the same as the one used industrially.

Figure 2 shows the rate of swelling in liquid water for the MUF boards. It is interesting to note that the board made from chips of the lowest weight gain (6.7%) swelled faster and to a greater extent than the control did. When chips of increasing level of acetylation were used, however, the corresponding particleboards showed a decreasing rate and extent of swelling. The rate of swelling was higher for the PF control board as compared to that of the MUF control, but the final extent of swelling was about the same for both controls (fig. 3). Irrespective of which acetylation treatment and adhesive were used, high levels of wood acetylation reduced the extent of board swelling to less than 10%, while
the two control boards made from untreated chips swelled approximately 50%.

Fig. 2. Swelling in liquid water of particleboards made from acetylated chips. (8% MUF adhesive).
- Control, + Vapor 6.7 WPG, * Vapor 11.5 WPG ▼ Vapor 17.9 WPG △ vapor 22.1 WPG, ◇ vapor 24.3 WPG, □ Liquid 23.0 WPG

Fig. 3. Swelling in liquid water of particleboards made from acetylated chips. (6% PF adhesive).
- Control, △ Vapor 23.7 WPG, □ Liquid 24.0 WPG

The thickness changes during the repeated water soaking test for the MUF particleboards are shown in fig. 4. The board made from vapor phase acetylated chips with a weight gain of 6.7% swelled much more than the control did, while the board made from chips with a weight gain of
11.5% swelled about the same as the control. This shows that low levels of acetylation caused more swelling to occur, and this trend was not reversed until a weight gain of at least 12% was achieved. Even at the highest levels of both vapor and liquid phase acetylation, the swelling of the boards was not reduced to less than 20% after six o.d./wet cycles. This may be a result of the low amount of MUF adhesive used, or may indicate that the adhesive breaks down under the test conditions.

Figure 5 shows the thickness changes for the boards made with the PF adhesive. Liquid phase acetylation to a weight gain of 24% gave a swelling of the board which was less than 15% after six o.d./wet cycles, while the board made from vapor phase acetylated chips of a somewhat lower weight gain was not as good.

Change in thickness in repeated water soaking test of particleboards made from acetylated chips. (Open symbols 8% MUF adhesive, filled symbols 6% PF adhesive).

○ Control, + vapor 6.7 WPG, X Vapor 11.5 WPG ▼ Vapor 17.9 WPG
▲ Vapor 22.1 WPG  □ Liquid 23.0 WPG
● control, A vapor 20.2 WPG, ■ Liquid 23.0 WPG
Table 1 shows that for both adhesives used the equilibrium moisture content (EMC) at each relative humidity decreased due to acetylation. The lowest EMC was obtained for the MUF board made of liquid phase acetylated chips, where the EMC was reduced over 65% at 90% RH and over 75% at 30% RH. The reduction was lower for the PF boards.

**TABLE 1**

Equilibrium moisture content (EMC) of particleboards made from acetylated pinechips.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Adhesive</th>
<th>WPG</th>
<th>EMC at 30% RH</th>
<th>65% RH</th>
<th>90% RH</th>
</tr>
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<tbody>
<tr>
<td>Control</td>
<td>8% MUF</td>
<td>0</td>
<td>6.1</td>
<td>9.8</td>
<td>18.3</td>
</tr>
<tr>
<td>Vapor</td>
<td>&quot;</td>
<td>20.0</td>
<td>2.7</td>
<td>5.0</td>
<td>9.1</td>
</tr>
<tr>
<td>Liquid</td>
<td>&quot;</td>
<td>23.0</td>
<td>1.4</td>
<td>3.7</td>
<td>6.7</td>
</tr>
<tr>
<td>Control</td>
<td>6% PF</td>
<td>0</td>
<td>6.8</td>
<td>10.5</td>
<td>19.6</td>
</tr>
<tr>
<td>Vapor</td>
<td>&quot;</td>
<td>20.2</td>
<td>3.2</td>
<td>6.5</td>
<td>14.2</td>
</tr>
<tr>
<td>Liquid</td>
<td>It</td>
<td>23.0</td>
<td>2.0</td>
<td>5.1</td>
<td>12.4</td>
</tr>
</tbody>
</table>

1 Weight percent gain.

Figure 6 shows the thickness changes of MUF boards at 30 and 90% RH. In contrast to the behavior in the repeated water soaking test, the board made from vapor phase acetylated chips of the lowest weight gain swelled about the same as the control did. An increased acetyl content decreased the extent of swelling. For the board made from liquid phase acetylated chips of a weight gain of 23%, the maximum swelling was reduced to about 5%. This value is lower than that obtained for the board made from vapor phase acetylated chips of almost the same weight gain.

With the use of PF adhesive and chips acetylated to 23% weight gain in the liquid phase, the board showed less than 5% swelling in the four cycle test (fig. 7). Essentially no swelling occurred in this board at 30% RH.
Change in thickness at 30 and 90% relative humidity (27°C) of particleboards made from acetylated chips. (Open symbols 8% MUF adhesive, filled symbols 6% PF adhesive).

- Control,
- + Vapor 6.7 WPG,
- × Vapor 11.5 WPG,
- ▽ Vapor 17.9 WPG,
- △ Vapor 22.1 WPG,
- □ Liquid 23.0 WPG,
- ○ Control,
- ▲ vapor 20.2 WPG,
- ■ Liquid 23.0 WPG

In both the water soaking and the humidity tests, the results show that irreversible swelling was evident in the early cycles. There was an increase in permanent swelling, however, even during later cycles which was more prominent in the case of the MUF boards also at high levels of wood acetylation. This observation is probably partly due to adhesive and wood failures resulting from harsh test conditions, but may also indicate that the MUF adhesive is less compatible with highly acetylated wood than is the PF adhesive.

Table 2 shows that at weight gains above 18% no biological decomposition took place in five months exposure in unsterile soil.
TABLE 2
Biological decomposition of particleboards made from acetylated chips (8% MUF adhesive) after five months exposure in unsterile soil.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>WPG¹</th>
<th>Rating²</th>
</tr>
</thead>
<tbody>
<tr>
<td>control</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>Vapor</td>
<td>6.7</td>
<td>4</td>
</tr>
<tr>
<td>&quot;</td>
<td>10.9</td>
<td>2</td>
</tr>
<tr>
<td>&quot;</td>
<td>18.3</td>
<td>0</td>
</tr>
<tr>
<td>&quot;</td>
<td>21.1</td>
<td>0</td>
</tr>
<tr>
<td>&quot;</td>
<td>24.5</td>
<td>0</td>
</tr>
<tr>
<td>Liquid</td>
<td>23.0</td>
<td>0</td>
</tr>
</tbody>
</table>

¹ Weight percent gain.
² Rating system: 0 - no attack, 1 - slight attack, 2 - moderate attack, 3 - severe attack, 4 - destroyed.

CONCLUSIONS

- Chemical modification by both liquid and vapor phase acetylation of wood chip before pressing into a particleboard greatly reduce the rate and extent of thickness swelling due to water sorption.
- Vapor phase acetylation seems to be less effective than liquid phase acetylation at the same weight gain in reducing the rate and extent of thickness swelling.
- Low levels of chip acetylation result in a greater thickness swelling of the board as compared to that obtained when using untreated chips.
- MUF boards swelled at a lower rate but continued to swell to a greater extent, than did the PF boards.
- Particleboards made from acetylated chip are stable towards a broad range of wood destroying microorganisms.
ACKNOWLEDGEMENT

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SUMMARY

Pine wood chips were made to react with acetic anhydride vapor or with liquid acetic anhydride diluted with xylene. The rate of acetylation was much lower for the vapor treatment than for the liquid phase treatment. High levels of acetylation (>20% weight gain) were achieved by both methods. Particleboards made from highly substituted chips swelled at a lower rate and to a lower extent than did the control boards made from untreated chips. At low levels of acetylation, on the other hand, the rate and extent of swelling were higher than those found for the controls.

With increased level of wood acetylation, the hygroscopicity of the resulting particleboards decreased. The equilibrium moisture content was the lowest at each relative humidity tested, when the particleboards were made from liquid phase acetylated chips. Boards made with a melamine-urea-formaldehyde adhesive swelled at a lower rate but continued to swell to a greater extent, than did particleboards containing a phenol-formaldehyde adhesive.

Boards made from highly substituted chips were stable towards a broad range of wood-destroying microorganisms.

LITERATURE