PARTICLEBOARD FROM RUBBER WOOD FLAKES WITH POLYMERIC MDI BINDER

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ABSTRACT

The use of rubber wood (Hevea brasiliensis) flakes for particleboard with polymeric diisocyanate (pMDI) as a binder was investigated. The effects of binder content in surface/core layers, cure temperature, cure time, and flake moisture on the properties of rubber wood particleboards were studied. The properties of the particleboard were compared with those of phenol-formaldehyde (PF) binder. The particleboard prepared from rubber wood flakes with pMDI binder gave high quality particleboards that had durability under severe conditions, low water absorption and thickness swelling, and a high modulus of rupture, modulus of elasticity, and internal bond strength. Moreover, pMDI binder could be cured at low temperatures, within a short time, and the binder consumption was small. The scanning electron micrographs of the rubber wood particleboard prepared from the high moisture content flakes showed the best, very tight packing characteristics.

Keywords: Particleboard, polymeric MDI, wood flakes, binder.

INTRODUCTION

Wood is a preferred building and engineering material because it is economical, especially in terms of low processing energy, renewable, strong, and aesthetically pleasing (Youngquist 1992). Nowadays, the forests of the world are progressively dwindling as the countries of the world are rapidly developing and their populations fast increasing. The demand for wood is on the increase, and the supply is unable to keep pace with the demand. In this respect, there are cheap sources of wood to substitute effectively for wood panel industries, such as plywood, fiberboard, and particleboard. Industrial wood residues, such as sawmill sawdust, plywood trim, plywood dust, wood chips, wood flakes etc., when suit-
ably processed, can be utilized in the manufacture of particleboard (Akers 1966). However, these wood residues can be bonded only with great difficulty or not at all, using the existing common aqueous binders. Through the development of polyisocyanate binders, it has become possible to produce particleboards and molded parts from these materials for interior and exterior uses. Under pressure and heat, polyisocyanates are able to penetrate the hydrophobic layer of wax and silicate that exists in such a natural product (Franke et al. 1994).

Panels and molded parts from wood chips, fibers, and veneer are produced under heat and pressure using a variety of organic binders. In the past, these binders were primarily aqueous condensation polymers based on formaldehyde and urea, melamine, or various phenols. Today, polyisocyanate bonding agents are replacing these binders in many applications. Because curing takes place when moisture reacts with polyisocyanates, polyureas are formed. Polymeric diphenylmethane diisocyanates (pMDI) have found increasing use because of their inherent value as bonding agents. The pMDI binders offer significant advantages in terms of processing and physical properties. These advantages more than offset the higher cost of polyisocyanate binders. Formaldehyde binders have long pressing times, intensive energy consumption, and health risk for workers. The demand for control of formaldehyde emissions further intensifies the interest in formaldehyde-free polyisocyanate binders.

In general, the intrinsic toxicity of the isocyanates and polyols, by ingestion, absorption or implantation, is low although some isocyanates can cause skin irritation. The main hazard arises from the possible inhalation of isocyanate vapor or aerosol droplets. All isocyanates are potential sensitizers and may cause respiratory irritation—that is, they may cause allergic symptoms in susceptible people. Additionally, from more than 30 years of polyurethane industrial experience handling isocyanates, there is nothing to suggest that they are human carcinogens (Wood 1990). The objectives of handling procedures are to avoid the inhalation of isocyanate vapor or aerosols and to protect eyes and skin from contact with isocyanates.

As the use of waste wood products for particleboard appears to gain importance, research work on various types of binders is of interest. Some investigators studied new difurfuryl diisocyanate adhesives derived from renewable resources. Ethylidenebis (2,5-furandiy lmethylene) diisocyanate (EDFI) was proven to be an excellent adhesive for bonding wood composites (Holfinger et al. 1993). A 99% 15N-labeled polymeric methylenebis (phenylisocyanate) (pMDI) resin was used to make a series of wood-15N-pMDI composites cured as a function of temperature and time (Wendler and Frazier 1996). The pMDI particleboard had superior performance compared with conventionally bonded board, and the quality of pMDI bonding was affected by the species of the wood being bonded (Franke et al. 1994). The diisocyanate-based binders were modified with natural polymers of tannins, starches, or proteins. A modified emulsifiable diisocyanate gave plywood good strength and less wood failure (Dix and Marutsky 1989). The oxidized lignosulfonate adhesive (LS-OB) was also studied as a partial substitute for urea-formaldehyde in particleboard, and a positive effect on the curing of resin was observed (Chen and Wu 1994).

At present, Thailand is the world’s largest producer of natural rubber and the biggest exporter of natural rubber latex. The average economic life of rubber trees is 25 years. The cut-down rubber tree can be used for substitution of import sawlogs and sawntimber from neighboring countries. Since 1989, the manufacturers have found that rubber wood can substitute for natural wood from natural forests, and the export market prefers furnitures made from rubber wood. Therefore, the rubber-growing countries of the world could develop wood-based industries to meet their own requirements and to supply neighboring countries, in competition with any of the highly
developed countries specializing in these industries. Thus, the use of wood chips or particle from rubber wood in particleboard is one of the methods to maximize the utilization of rubber wood.

The aim of this research was to fabricate particleboard from rubber wood flakes with polymeric diphenylmethane diisocyanate (polymeric MDI or pMDI) binder by hot-pressing. The effects of pMDI content in surface and core layers, cure temperature, cure time, and flake moisture on the properties of particleboard were investigated.

EXPERIMENTAL

Materials and samples

Polymeric MDI for this study was Raycore B9001 supplied by Thai Petrochemical Industry Co., Ltd. The phenolic resin was Dynosol S-576 supplied by Thai Plywood Industry Co., Ltd. The rubber wood (Hevea brasiliensis) flakes were obtained from Vanachai Group Public Co., Ltd. The flakes were divided into two groups, the coarse and fine flakes of ASTM mesh size ranges of -5 to +60 and -20 to +100, respectively.

For each board, the appropriate quantity of flakes was weighed and placed in a rotary drum blender. The resin (as a percentage based on the oven-dried weight of the flakes, 100% solid) was sprayed onto the flakes from a sprayhead mounted at the center of the blender drum. Compressed air was used to atomize the resin. After blending, the flakes were matted by hand on caulcs with a releasing agent. In all cases, the mat was formed into 3-layer particleboard with a required board density of 750 kg/m³. It was formed within the forming case sized 35 × 35 cm. Then 250 g of fine flakes were placed on the treated lower caul for each surface, and 500 g of coarse flakes were used for the core. Then the completely formed mat was prepressed to consolidate the mat for subsequent handling, covered with the treated upper caul, and transferred to the hot press. The 10-mm-thick stopper was used to control the thickness of the mat. The resin was cured at the selected temperature. The mat was pressed at a pressure of 140–160 kg/cm² for a definite time. After pressing, the particleboards were allowed to cool at room temperature, and then stored at room conditions at 27°C until testing.

Investigation method

The water absorption, thickness swelling, internal bond strength (dry), modulus of rupture (dry), and modulus of elasticity (dry) were determined according to TIS 867-2532 (Thai Industrial Standard 1989). The internal bond strength (wet) was determined according to DIN 68763 (Deutsch Norm 1990). The modulus of rupture (wet) was determined according to CAN 3-0188.0-M78 (Canadian Standards Association 1978).

The fracture surfaces of internal bond test specimens were observed by a JOEL JSM-6400 scanning electron microscope. The samples were coated with gold before scanning observations.

RESULTS AND DISCUSSION

Effect of pMDI content in surface and core layers

Results on the effect of pMDI content in surface and core layers on the mechanical properties of particleboard are presented in Table 1. The water absorption decreased with increasing pMDI content in the core layer. The modulus of rupture and internal bonding both in dry and wet conditions were almost unchanged with increasing pMDI content in the core layer. The properties of the particleboard surface were improved by increasing the pMDI content in the surface layer. The particleboard with pMDI content at the surface layer of 7% and at the core of 4–5% gave excellent properties, from which the internal bond strength was about 3 times the standard value and the thickness swelling was about half of the standard value of TIS 867-2532 (Thai Industrial Standard 1989).

Effect of cure temperature and cure time

The effects of the cure temperature and cure time on the mechanical properties are given in
Table 1. Effect of pMDI content on properties of particleboard prepared at cure temperature of 160°C and cure time of 5 min.

<table>
<thead>
<tr>
<th>PMDI content at surface/core (%)</th>
<th>Water absorption (%)</th>
<th>Modulus of elasticity (MPa)</th>
<th>Modulus of rupture (MPa)</th>
<th>Internal bonding (MPa)</th>
<th>Thickness swelling (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>(dry) (MPa)</td>
<td>(wet) (MPa)</td>
<td>(dry) (MPa)</td>
<td>(wet) (MPa)</td>
</tr>
<tr>
<td>7/3</td>
<td>24.2</td>
<td>2.721</td>
<td>19.6</td>
<td>7.6</td>
<td>1.00</td>
</tr>
<tr>
<td>7/4</td>
<td>18.5</td>
<td>2.142</td>
<td>19.3</td>
<td>7.5</td>
<td>1.07</td>
</tr>
<tr>
<td>7/5</td>
<td>17.3</td>
<td>2.334</td>
<td>18.0</td>
<td>7.3</td>
<td>1.07</td>
</tr>
<tr>
<td>6/6</td>
<td>16.4</td>
<td>2.678</td>
<td>19.6</td>
<td>NA</td>
<td>1.15</td>
</tr>
<tr>
<td>*</td>
<td>40 (max)</td>
<td>2,000 (min)</td>
<td>13.8 (min)</td>
<td>5.6 (min)**</td>
<td>0.34 (min)</td>
</tr>
</tbody>
</table>

* Standard values for flat press particleboard according to the Thai Industrial Standard, TIS 876-2532
** According to CAN 3-0188.0-M78
*** According to DIN 68763, V100

Table 2. The water absorption and thickness swelling increased with increasing press temperature. The internal bond strength, both dry and wet, was not significantly different. The particleboard pressed at 160°C gave the highest value of the modulus of rupture (MOR) and modulus of elasticity (MOE) under both dry and wet conditions. The particleboard pressed at 120°C exhibited the lowest water absorption, thickness swelling, and modulus of rupture. The low cure temperature range of 120–160°C was appropriate for use with pMDI. Compared to the use of the conventional binder (phenol-formaldehyde), the high cure temperature of 180–200°C is required for particleboard preparation, because the cure rate of the phenolic group is increased by the higher temperature. For the use of pMDI, the low press temperature can be used rather successfully, since heat from the press is also supplemented from the heat developed by the condensation reaction of the resin cure (Wendler and Frazier 1996). The particleboard using pMDI could be cured at a lower temperature (120°C) than that for the conventional binder. Wendler and Frazier (1996) reported that at a low moisture content, the low cure temperature promoted the urea and biuret formation, but biuret appeared to be dominant at the high cure temperature.

For the cure temperature of 160°C, the highest value of internal bond strength at the dry condition was achieved at the cure time of 3 minutes. Therefore, a good quality of particleboard can be achieved at the low cure time of 3 minutes. Wendler and Frazier (1996) also reported that when pMDI was cured at 120°C with 4.5% wood moisture, the network chemistry did not change significantly with time. However, at a low cure time, high values of thickness swelling and water absorption were achieved.

Effect of flake moisture

Table 3 shows the effect of flake moisture on the modulus of elasticity, modulus of rupture, and internal bond strength. The MOE,
TABLE 3. Effect of flake moisture and binder type on properties of particleboard prepared with binder content of 7% in surface and 5% in core at cure temperature of 160°C and cure time of 5 min.

<table>
<thead>
<tr>
<th>Binder</th>
<th>Flake moisture (%)</th>
<th>Water absorption (%)</th>
<th>Modulus of elasticity (MPa)</th>
<th>Modulus of rupture (dry) (MPa)</th>
<th>Modulus of rupture (wet) (MPa)</th>
<th>Internal bonding (dry) (MPa)</th>
<th>Internal bonding (wet) (MPa)</th>
<th>Thickness swelling (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pMDI</td>
<td>2</td>
<td>17.4</td>
<td>1,807</td>
<td>12.5</td>
<td>4.8</td>
<td>1.07</td>
<td>0.23</td>
<td>4.22</td>
</tr>
<tr>
<td>pMDI</td>
<td>7</td>
<td>17.3</td>
<td>2,334</td>
<td>18.0</td>
<td>7.4</td>
<td>1.07</td>
<td>0.26</td>
<td>5.60</td>
</tr>
<tr>
<td>pMDI</td>
<td>12</td>
<td>12.5</td>
<td>2,974</td>
<td>23.3</td>
<td>10.3</td>
<td>2.09</td>
<td>0.45</td>
<td>4.45</td>
</tr>
<tr>
<td>pMDI</td>
<td>17</td>
<td>11.2</td>
<td>3,186</td>
<td>25.3</td>
<td>9.9</td>
<td>1.48</td>
<td>0.34</td>
<td>3.73</td>
</tr>
<tr>
<td>pMDI</td>
<td>22</td>
<td>14.8</td>
<td>3,111</td>
<td>23.7</td>
<td>11.1</td>
<td>1.09</td>
<td>0.36</td>
<td>3.52</td>
</tr>
<tr>
<td>PF/pMDI</td>
<td>7</td>
<td>32.1</td>
<td>2,484</td>
<td>20.0</td>
<td>8.3</td>
<td>1.29</td>
<td>0.29</td>
<td>12.0</td>
</tr>
<tr>
<td>PF</td>
<td>7</td>
<td>48.6</td>
<td>2,385</td>
<td>19.4</td>
<td>9.3</td>
<td>1.13</td>
<td>0.19</td>
<td>15.6</td>
</tr>
</tbody>
</table>

* Standard values for flat press particleboard according to Thai Industrial Standard, TIS 876-2532.
** According to CAN 3-0188-0-M78.
*** According to DIN 68763, V100.

MOR, and internal bond strength increased with increasing flake moisture, reached a maximum value, and then decreased with flake moisture. The maximum internal bond strength was achieved at the flake moisture of 12%. The chemical reactions of isocyanate with wood are the bonding reactions between isocyanate and water as follows:

\[
\begin{align*}
R-\text{NCO} + \text{H}_2\text{O} \rightarrow R-\text{N}-\text{C}=\text{O} + \text{NH}_2 \quad \text{Carbamic acid} \\
R-\text{NH}_2 + R-\text{NCO} \rightarrow R-\text{N}-\text{C}+\text{R} \quad \text{Urea} \\
R-\text{NH}_2 + \text{R-NCO} \rightarrow R-\text{N}=\text{C}+\text{R} \quad \text{Biuret}
\end{align*}
\]

At high temperature, polyisocyanates were transformed into polyureas in the presence of moisture. The flake moisture content must be controlled so that it gives the optimum number of urethane linkages to create adequate bonding between the wood surface and the binders. The high mat moisture content of surfaces also assisted in plasticizing the wood particles and provided tight and hard surfaces. The various moisture contents and the distribution of moisture content throughout the mat had a pronounced effect on the final particleboard density and mechanical properties. The problems resulting from a low mat moisture content are high water absorption in the board, a rough surface due to the decrease in the wood plasticization, and slow heat transfer to the core.

The particleboard using pMDI was compared with the particleboard having phenol-formaldehyde (PF) in both layers and that having PF in the surface layers and pMDI in the core layer (PF/pMDI). From Table 3, the MOR, MOE, and internal bond strength of particleboard (7% flake moisture) with PF and PF/pMDI binders were slightly higher than those with pMDI binder. However, the water
absorption property of particleboard using pMDI binder was better than that for PF and PF/pMDI binders. For the particleboard using PF resin, the thickness swelling value was higher than the standard value.

These results demonstrated that pMDI could be used well with high flake moisture and it gave particleboard of better properties than those for conventional binders. The particleboard using pMDI in the core layer and PF in the surface layers also exhibited good performance. Thus, polymeric MDI could partially replace PF in the manufacture of wood products. The pMDI can be used on existing machines for making particleboard with little or no modification. Though the pMDI cost is higher than PF, the cost of particleboard should be reduced considerably if pMDI content used is less than PF. The operating cost can be reduced to a minimum by the decrease of pressing time and temperature. Optimization with respect to process conditions and pMDI content will no doubt reduce the particleboard cost; therefore, the production of particleboard should possibly be feasible in the next decade in the country where control of formaldehyde emission is stringent.

*Fracture surfaces of particleboards*

Scanning electron microscopy was employed to investigate the fracture surfaces of samples from the internal bond test. In Figs. 1a and 1b, the SEM micrographs of particleboard with 2% flake moisture and 7/5 pMDI content show poor interfacial adhesion and a loose packing of flakes. The particleboard with 12% flake moisture had the best, very tight packing characteristics. The packing behavior supported the results of mechanical tests that the particleboard prepared from the flakes with 12% moisture content had the highest internal bond strength. Furthermore, this confirmed the effect of the moisture content on the compressibility of the flakes: the higher the flake moisture, the greater the compressibility and the internal bond strength.

**CONCLUSIONS**

The manufacture of particleboard from rubber wood flakes with pMDI binder appears to be technically feasible. The properties meet the industry-level requirements for commercial particleboard. The appropriate conditions
for the particleboard preparation are pMDI contents of 7% in surface and 5% in core layers, flake moisture of 12%, a cure temperature of 160°C, and a cure time of 5 min. Under these conditions, the highest-performance particleboard was achieved with the lowest water absorption, low thickness swelling, the highest modulus of rupture and modulus of elasticity, and the highest internal bonding strength, both under dry and wet conditions.

The properties of the particleboards prepared with pMDI and PF resin were significantly different. At the same binder content, pMDI binder gave a higher performance than PF resin, especially for a high flake moisture. The case of using pMDI in the core layer and PF resin in the surface layers indicated that the good properties of rubber wood particleboard could be achieved and conventional releasing agents could be used. The interfacial adhesion could be confirmed via the observation of fracture surfaces by SEM. The scanning electron micrographs of the rubber wood particleboard prepared from flakes of high moisture content showed the best, very tight packing characteristics. This was also supported by the results on the effect of moisture content on the internal bond strength of particleboard.

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