EFFECTS OF CHEMICAL COMPOSITION OF WOOD AND RESIN TYPE ON PROPERTIES OF PARTICLEBOARD

Bünyamin Sarı, a Nadir Ayrilmis, b, * Gökay Nemli, a Mehmet Baharoğlu, a Esat Gümüşkaya, c Selahattin Bardak d

The effects of chemical composition of various wood species and resin type on the dimensional stability, mechanical properties, and formaldehyde emission of particleboard were investigated. The solubility in cold and hot water, NaOH, and alcohol-benzene were evaluated, as well as the amount of cellulose, α-cellulose, holocellulose, hemicelluloses, and lignin and the acidity (pH) of the wood particles after the chipping process. The chemical compositions of wood and resin type were the main parameters influencing physical properties, mechanical properties, and formaldehyde emission of particleboard. While high cellulose and α-cellulose content resulted in superior mechanical properties, high hemicelluloses content was detrimental to the mechanical properties and dimensional stability. The extractives dissolving in cold and hot water decreased the formaldehyde emission of particleboard. Hemicelluloses were found to be effective at lowering formaldehyde emission. High hemicelluloses content caused lower formaldehyde releasing. Extractives dissolving in the NaOH and alcohol-benzene positively affected the dimensional stability of particleboard panels. Resin type was found to have an effect on all of the properties of particleboard. Particleboards produced with melamine-urea formaldehyde resin showed better quality properties and lower formaldehyde emission compared to the particleboards produced with urea-formaldehyde resin.

Key words: Particleboard; Mechanical properties; Physical properties; Chemical compounds; Formaldehyde emission; Resin type

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INTRODUCTION

Particleboard is an engineered wood-based panel composite manufactured from wood particles or other lignocellulosic fibrous materials with the addition of a resin. It is one of the most popular materials used in furniture, counter- and desktops, insulators, cabinets, wall and ceiling panels, flooring, bulletin boards, office dividers, building, and packing materials. The demand for particleboard has recently increased throughout the world, due to the fact that particleboard is a homogenous material suitable for both industrial production and construction. Although particleboards have some advantages,
Lignocellulose

they have four important disadvantages such as formaldehyde emission, water absorption, wood-attacking insects, and flammability (Sellers 2000; Nourbakhsh 2010). For example, the emission levels of formaldehyde fumes from wood-based panels manufactured using UF resins has now become one of the major concerns of the panel and wood adhesives industries. Recently, there have been many concerns about human health and the environment. The impact of the increased consciousness about indoor environment has created a demand for low-emitting (healthy) particleboards for industrial furniture.

A considerable amount of work has been carried out, studying the effects of some manufacturing factors and properties of raw materials related to the physical and mechanical quality properties and formaldehyde emission of particleboard, such as amount of resin (Nemli and Demirel 2007), collection time of raw materials (Nemli and Aydin 2007), density profile (Nemli and Aydin 2007), sanding factors (grit sizes, feeding speed, and feed powder) (Nemli et al. 2007a), moisture content of the mat, wood dust usage, press time (Nemli et al. 2007b), press temperature, pressure, panel density, shelling ratio (Kalaycioglu and Nemli 2006), impregnation with bark extractives (Nemli et al. 2006), surface coating applications (Nemli and Hiziroglu 2009; Nemli et al. 2003), residue type (Nemli et al. 2004), permeability of wood (Lynam 1969), resin type (Geimer et al. 1973), dimensions of the particles (Vital et al. 1980), moisture content of the particleboard (Halligan and Schiewind 1974), waste sanding dust usage (Bardak et al. 2010), and formaldehyde/urea molar ratio of resin (Akbulut 1995). Nevertheless, the problems related to the physical and mechanical properties of particleboard still exist.

Urea-formaldehyde (UF) resin-based resins are used extensively in the production of wood-based panels because of their excellent cohesion and adhesion, lack of color in the finished product, and low cost. Nonetheless, formaldehyde can be irritating to the eyes, nose, and throat. The eyes are most sensitive to formaldehyde exposure. Over the past several decades, air pollution in residential buildings has become a matter of increasing concern. Pollutants, such as formaldehyde, are emitted into indoor air from the building materials and the wood-based construction products. It is a suspected human carcinogen that is known to be released from pressed wood products used in home construction, including products made with UF resin (e.g., particleboard, plywood, and fiberboard). Emission of formaldehyde adversely affects indoor air quality. The toxicity and health hazards of wood-based panels due to the emission of formaldehyde could act as an obstacle to their acceptance by the public, given the prevailing climate of environmental awareness and concern (Kim et al. 2006a). There has been increasing focus on formaldehyde as it relates to cancer and asthma (Natz 2007; Kim et al. 2006a; Uchiyama et al. 2007; Pizzi 1994a). Many studies have been carried out on the effects of some factors on formaldehyde emission, such as formaldehyde/urea molar ratio, panel density, shelling ratio, waste screen dust usage (Sari et al. 2011), bio-scavengers (tannin, wheat flour, rice husk flour, and charcoal) usage (Kim et al. 2006b), pine cone flour content (Ayrilmis et al. 2009), volcanic pozzolan, ammonia, ammonium salts, organic amines, ester usage (Kim 2009; Sundin et al. 1997), urea and melamine addition to UF resin (Tsai 1984), reaction of ammonia with formaldehyde (Myers 1986), coating of particleboard surfaces with decorative papers (Groah et al. 1984), press temperature and time (Kollman et al. 1975), hardener type, and impregnation of particles with nitric acid (Mari et al. 1987). This study will focus on using melamine-urea formaldehyde (MUF)
resin as a binder for particleboard to evaluate its effectiveness at reducing formaldehyde emission.

Particleboard is comprised of about 90% (by weight) wood material, which has a significant effect on its quality properties. The chemical properties of wood, in particular extractives and pH, significantly affect the cure cycle of UF resin, as well as swelling and mechanical properties of particleboard. The effects of the chemical composition of wood on the properties of particleboard have not been extensively investigated in the literature. Therefore, the aim of the present study was to determine the effects of the chemical composition of wood and resin type on the physical properties, mechanical properties, and formaldehyde emission of particleboard.

**MATERIALS AND METHODS**

**Materials**

Pine (*Pinus sylvestris* L. and *Pinus palustris* Mill.), beech (*Fagus orientalis* L.), and poplar (*Populus tremula* L.) species obtained from the city of Bursa on the northwestern side of Turkey were used in this study. The wood of each species was processed into particles by passing through a chipper and a ring flaker. The particles were then dried to 1% moisture content prior to the resin application. The wood particles passing the 1 mm screen but retained by the 0.25 mm screen were used for surface layers, while the wood particles retained by a pneumatic system on the 1 mm screen were used for the core layer. Commercial E1 grade UF and MUF resin were used in this study. Resins were water dispersed with a solids content of 65%. They were sprayed onto the particles in a blender. The resin contents of 9 and 11% were used for the core and surface layers, respectively, based on the oven-dry weight of the particles. Ammonium sulphate and urea were added to the resin as hardener and formaldehyde scavenger. For the surface layers 0.5% ammonium sulphate (10% solution) and for the core layer 2.5% ammonium sulphate (25% solution) were used, based on the solid weight of the resin. As a hydrophobic substance, a paraffin emulsion with a solids content of 32% was used. Paraffin contents of 1.5% and 1% were used for the core and surface layer, respectively, based on the oven-dry weight of the particles. A hot press was used to manufacture the boards. The panels were pressed at 220 °C for 100 sec under 2.5 N/mm² pressure. The resulting panels were sanded with a sequence of 40, 60, 80, 100, and 120 grit sandpapers. The ratio of the face thickness to the total thickness was 0.34 for all boards. The target density and dimensions of the panels were 0.665 g/cm³ and 183 × 366 × 1.8 cm, respectively. A total of 6 experimental panels, two for each type of panel, were manufactured. The raw material formulations used for the particleboards are presented in Table 1.
Table 1. The Raw Material Formulations Used for the Particleboards

<table>
<thead>
<tr>
<th>Panel type</th>
<th>Particleboard composition (by weight)</th>
<th>Resin type</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pinus sylvestris L. (%)</td>
<td>Pinus palustris Mill. (%)</td>
</tr>
<tr>
<td>A</td>
<td>90</td>
<td>0</td>
</tr>
<tr>
<td>B</td>
<td>0</td>
<td>90</td>
</tr>
<tr>
<td>C</td>
<td>90</td>
<td>0</td>
</tr>
</tbody>
</table>

UF: urea-formaldehyde resin, MUF: melamine-urea formaldehyde resin

Methods

The modulus of rupture (MOR), modulus of elasticity (MOE), internal bond strength (IB), and thickness swelling (TS) were prepared and tested in accordance with European norm (EN) standards 310 (1993), EN 319 (1993), EN 317 (1993), and EN 120 (1992), respectively. Twenty replicates were used for each physical and mechanical property. The TS was measured after 24 h immersion in distilled water at 20 °C.

Formaldehyde emissions (FE) of the samples were determined using the perforator method based on standard EN 120 (1992). Twenty samples (20 mm × 20 mm) were randomly taken from each type of particleboard for formaldehyde emission calculation. The perforator method involves reflux in boiling toluene with approximately 110 g small cube samples. As soon as refluxing of the toluene occurred, the refluxing speed of the system was adjusted to 30 mL/min (50-70 drop/min). The duration of extraction was 2 h. Extraction of water with formaldehyde was carried out by adding 50 mL of iodine solution and 20 mL of sodium hydroxide in a dark room for 15 min. A 10 mL mixture of sulfuric acid and sodium thiosulfate solution was applied to water until its color changed from light brown to light yellow. This method led to the determination of the perforator value, which is expressed in milligrams of formaldehyde per 100 g of dry sample.

For the determination of the chemical properties of raw materials, preparation of the test specimens was carried out according to TAPPI standard T m-45 (1992). Alcohol-benzene (TAPPI T 204 cm-97 1992), dilute alkali (1% NaOH) (TAPPI T-212 om-98 1992), hot/cold water solubility (TAPPI T 207 om-88 1992), α-cellulose (TAPPI T 429 cm-84 2000), pH (Browning 1967), holocellulose (Wise and Karz 1962), cellulose (EPF 1969), and lignin (TAPPI T 222 om-98 1998) were determined. Data from each test were statistically analyzed using T-tests at 95% confidence level.

RESULTS AND DISCUSSION

Chemical Properties

Table 2 displays the average chemical properties of raw materials and results of statistical analyses. According to the results, there were no statistical differences between the pH, lignin, and holocellulose contents of the raw materials. The wood species had a statistically significant effect on the cellulose, α-cellulose, and solubility. While the highest values for cellulose, α-cellulose, NaOH, and alcohol-benzene solubility were
obtained from *Pinus sylvestris* L. wood, *Pinus palustris* Mill. had higher cold and hot water solubility.

**Table 2. The Average Chemical Properties of Raw Materials and Results of Statistical Analysis**

<table>
<thead>
<tr>
<th>Properties / Raw materials</th>
<th><em>Pinus sylvestris</em> L.</th>
<th><em>Pinus palustris</em> Mill.</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>4.87 (0.06) a</td>
<td>4.83 (0.04) a</td>
</tr>
<tr>
<td>Holocellulose (%)</td>
<td>71.61 (0.18) a</td>
<td>71.59 (0.14) a</td>
</tr>
<tr>
<td>Hemicellulose* (%)</td>
<td>16.13 (0.35) a</td>
<td>21.0 (0.26) b</td>
</tr>
<tr>
<td>Cellulose (%)</td>
<td>55.48 (0.90) a</td>
<td>50.58 (0.51) b</td>
</tr>
<tr>
<td>α-cellulose (%)</td>
<td>48.94 (0.17) a</td>
<td>41.59 (0.09) b</td>
</tr>
<tr>
<td>Lignin (%)</td>
<td>26.71 (0.13) a</td>
<td>26.68 (0.11) a</td>
</tr>
<tr>
<td>Cold water solubility (%)</td>
<td>3.14 (0.13) a</td>
<td>5.86 (0.26) b</td>
</tr>
<tr>
<td>Hot water solubility (%)</td>
<td>4.08 (0.05) a</td>
<td>6.97 (0.08) b</td>
</tr>
<tr>
<td>NaOH solubility (%)</td>
<td>17.88 (0.11) a</td>
<td>14.89 (0.07) b</td>
</tr>
<tr>
<td>Alcohol-benzene solubility (%)</td>
<td>6.45 (0.15) a</td>
<td>4.68 (0.06) b</td>
</tr>
</tbody>
</table>

Note: Different letters in the same line represent statistical differences at 95% confidence level, numbers in the parenthesis are standard deviations, *Hemicelluloses are the minus between the holocellulose and cellulose.

**Physical Properties, Mechanical Properties, and Formaldehyde Emission**

The average values of the MOR, MOE, IB, TS, and FE and results of the statistical analyses are given in Table 3. The flexural properties of the MUF resin-bonded particleboards were significantly better than those of UF resin-bonded particleboards. The MOR and MOE values of the UF resin-bonded particleboards (type A) were 13.62 and 1738.8 N/mm², while they were found to be 16.78 and 2579.3 N/mm² for the MUF resin-bonded particleboards, respectively. Concerning tree species, the flexural properties of the particleboards made from *Pinus sylvestris* were better than that of particleboard made from *Pinus palustris* (Table 3). Based on standard EN 312 (2004), 11.5, 13.0, and 16.0 N/mm² are the minimum MOR requirements of particleboard for general uses, interior fitments (including furniture), and load bearing boards in wet conditions, respectively, while the minimum MOE requirements for furniture used in indoor and load bearing applications in wet conditions are 1600 and 2400 N/mm², respectively. The MUF-bonded particleboards met the minimum MOR and MOE requirements of EN 312 for load bearing applications in wet conditions. The particleboards made from *Pinus sylvestris* met the minimum MOR and MOE requirements of EN 312 for interior general purpose applications, while the particleboards made from *Pinus palustris* did not meet the minimum MOE requirement (1600 N/mm²) of EN 312.
The IB strength of the particleboards showed a similar trend to flexural properties. The IB of the MUF resin-bonded particleboards was significantly better than those of UF resin-bonded particleboards. The average IB value of the UF resin-bonded particleboards was 0.39 N/mm², while it was 0.50 N/mm² for the MUF resin-bonded particleboards. The IB strength of the particleboards made from *Pinus sylvestris* was better than that of the particleboard made from *Pinus palustris* (Table 3). The minimum requirements of IB strength for general purpose, furniture manufacturing, and load bearing applications in wet conditions are 0.24, 0.35, and 0.45 N/mm², respectively (EN 312 2004). According to the test results, MUF resin-bonded particleboards met the minimum IB strength requirement of EN 312 for particleboards used in load bearing applications in wet conditions, while the UF resin-bonded particleboards made from *Pinus sylvestris* met the minimum requirement for load bearing applications in dry conditions. The IB strength of the particleboards made from *Pinus palustris* met the minimum requirements of general purpose particleboard for dry conditions.

The maximum thickness swelling of particleboards for load bearing applications in wet conditions after 24 h immersion should be 10%, according to standard EN 312. According to the test results, panels produced with the MUF resin had the required level of TS. The chemical composition of wood species had a statistically significant effect on all of the properties of particleboards. According to the statistical analysis results, particleboard panels made from *Pinus sylvestris* L. showed better mechanical properties than those of the panels made from *Pinus palustris* Mill. As can be seen in Table 2, the cellulose and α-cellulose contents of *Pinus sylvestris* L. are higher than *Pinus palustris* Mill. The mechanical properties of wood species that have higher amounts of cellulose and α-cellulose are better than those of wood species that have low amounts of cellulose and α-cellulose (Pettersen 1984). Hot and cold water solubility of *Pinus palustris* Mill. wood are higher than those of *Pinus sylvestris* L. Extractives dissolving in the cold and hot water, such as acetic and uronic acids, break down the linkage between the wood and resin during hot pressing. They decrease the pH of wood and cause pre-curing of the UF resin. Pre-curing of the resin decreases the internal bond between the particles (Foster 1967). There are no statistical differences between the hemicellulose contents of wood species. Hemicellulose is the total amount of cellulose and hemicelluloses. The cellulose content was higher for *Pinus sylvestris* L. wood. Therefore, the hemicelluloses content of

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**Table 3. Average Values of Modulus of Rupture, Modulus of Elasticity, Internal Bond Strength, Thickness Swelling, and Formaldehyde Emission, and Results of Statistical Analysis**

<table>
<thead>
<tr>
<th>Particleboard type</th>
<th>MOR (N/mm²)</th>
<th>MOE (N/mm²)</th>
<th>IB (N/mm²)</th>
<th>TS (%)</th>
<th>FE (mg CH₂O/100 g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>13.62 (0.44) a</td>
<td>1738.8 (64.67) a</td>
<td>0.39 (0.025) a</td>
<td>14.43 (0.41) a</td>
<td>7.84 (0.07) a</td>
</tr>
<tr>
<td>B</td>
<td>12.36 (0.48) b</td>
<td>1511.0 (67.28) b</td>
<td>0.29 (0.035) b</td>
<td>16.77 (0.50) b</td>
<td>6.76 (0.09) b</td>
</tr>
<tr>
<td>C</td>
<td>16.78 (0.62) c</td>
<td>2579.3 (86.45) c</td>
<td>0.50 (0.042) c</td>
<td>9.03 (0.23) c</td>
<td>5.03 (0.05) c</td>
</tr>
</tbody>
</table>

Note: Different letters in the same column represent statistical differences at 95% confidence level. MOR: modulus of rupture, MOE: modulus of elasticity, IB: internal bond strength, TS: thickness swelling, FE: formaldehyde emissions.
the *Pinus palustris* Mill. was higher than *Pinus sylvestris* L. Hemicelluloses are weak compounds of the wood and their polymerization degree is lower than cellulose. A low polymerization degree causes poor mechanical strength properties. Hemicelluloses dissolve in alkali solutions and hydrolyze in acids (Pettersen 1984). For these reasons, particleboards made from *Pinus palustris* Mill. particles showed poorer mechanical strength properties.

Particleboards made from *Pinus sylvestris* L. showed lower thickness swelling than the panels from *Pinus palustris* Mill. There are two reasons for low thickness swelling values of the panels made from *Pinus sylvestris* L. One of these reasons is the hemicelluloses content. Hemicelluloses absorb a greater amount of water than cellulose. Particleboards made from *Pinus palustris* Mill. had higher thickness swelling values due to a higher amount of hemicelluloses. Another reason is attributed to the presence of more extractives in *Pinus sylvestris* L. wood that dissolve in NaOH and alcohol-benzene, as can be seen in Table 2. The extractives, such as wax and lipophilic extractives, make the particleboard more waterproof (Maloney 1993). The positive effect of these extractives on the resistance to water and humidity was mentioned in a previous work (Marshall et al. 1974).

The FE of the UF resin-bonded particleboards was significantly higher than that of the MUF resin-bonded particleboards, as was expected (Table 3). The maximum permissible formaldehyde content for E1 quality particleboard is 8 mg HCHO/100 g dry sample (EN 312-1 1997). The results showed that all of the panel types were within the accepted level of formaldehyde emission for indoor uses. Wood species statistically affected the formaldehyde emission of particleboard. The FE (7.84 mg HCHO/100 g) of the particleboards made from *Pinus sylvestris* was significantly higher than (6.76 mg HCHO/100 g) the particleboards made from *Pinus palustris*. Acetyl groups in the wood play a major role in formaldehyde emission. These groups change to acetic acid during hot pressing and fix the formaldehyde. Hemicelluloses contain a higher amount of acetyl groups than cellulose. According to the test results (Table 2), *Pinus palustris* Mill. contained more hemicellulose than *Pinus sylvestris* L. Another reason for lower formaldehyde emission is extractives dissolving in cold and hot water. Dissolved extractives, such as tannin and phenolic compounds, fix the formaldehyde as a formaldehyde scavenger (Akbulut 1995). As shown in Table 2, *Pinus palustris* had greater cold and hot water solubility values than those of *Pinus sylvestris* L. wood.

The resin type had a statistically significant effect on all of the properties of the particleboard. The MUF resin-bonded particleboards had higher mechanical strength (MOR, MOE, and IB) properties, but lower thickness swelling and formaldehyde emission values compared to UF resin-bonded particleboards. As with urea, formaldehyde first attacks the amino groups of melamine, forming methylol compounds. The condensation reaction of melamine with formaldehyde is similar to that between urea and formaldehyde. However, the addition of formaldehyde to melamine occurs more easily and completely than to urea. The amino group in melamine easily accepts up to two molecules of formaldehyde. Thus, complete methylolation of melamine is possible, which is not the case with urea. Up to six molecules of formaldehyde attach to a molecule of melamine. The methylolation step then leads to a series of methylol compounds with two to six methylol groups. Because melamine is less soluble than urea in water, the
hydrophilic stage proceeds more rapidly in MUF resin formation. Therefore, hydrophobic intermediates of the MUF resin condensation appear early in the reaction (Kim and Kim 2005).

UF resins have linear linkages, but MUF resins have cross linkages. Cross linkages give more stable mechanical properties to particleboard. Melamine is an ideal chemical to fortify UF resins primarily due to melamine’s high functionality, stable molecular structure in comparison to urea, and reaction mechanisms similar to those of urea with formaldehyde (Dunky 2003). The higher melamine content enhances the bond strength of boards because of the higher functionality and rigid structure of melamine (Young and Kim 2007). The formaldehyde emission of the MUF resin-bonded particleboards was significantly \( p < 0.05 \) lower than that of the UF resin-bonded particleboards (Table 3). This is because the addition of formaldehyde to melamine occurs more easily and completely than to urea, even though the condensation reaction of melamine with formaldehyde is similar to the reaction between urea and formaldehyde.

CONCLUSIONS

1. This study presented the effects of the chemical composition of wood on the physical properties, mechanical properties, and formaldehyde emission of particleboards. The chemical composition of wood and resin type were main parameters influencing physical properties, mechanical properties, and formaldehyde emission of particleboard.
2. While high cellulose and \( \alpha \)-cellulose contents caused superior mechanical properties, high hemicelluloses content decreased the mechanical properties and increased the thickness swelling.
3. Extractives that dissolve in cold and hot water decreased the formaldehyde emission of particleboard. Extractives that dissolve in NaOH and alcohol-benzene positively affected the dimensional stability of particleboards.
4. Hemicelluloses were found to be effective on the formaldehyde emission. High hemicelluloses content caused lower formaldehyde releasing.
5. MUF resin-bonded particleboards showed superior quality properties, compared to the UF resin-bonded particleboards.

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