Effect of Poplar Cultivar “Hybrid 275” Fiber Impregnation with 1,3-Dimethylol-4,5-dihydroxyethyleneurea on the Properties of High Density Fiberboards

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Wood from the fast-growing poplar cultivar “Hybrid 275” (P. maximowiczii x trichocarpa) was defibrated under industrial conditions, and the resulting fibers were subjected to impregnation modification with 5% or 10% 1,3-dimethylol-4,5-dihydroxyethyleneurea (DMDHEU). The modified fibers were used to produce 3-mm thick high density fiberboards (HDF). The mechanical testing revealed that DMDHEU provided an increase in modulus of elasticity (MOE) and a decrease in modulus of rupture (MOR); however, all panels met the requirements of the European Standard EN 310 (1993). Increases of 1.5-fold and 2.5-fold in internal bonding (IB) compared to the unmodified reference panels were observed. The modification resulted in reduced HDF interactions with water, manifested by decreased water absorption, thickness swelling, and reduced water wetting. The modification also provided the panels with an improved resistance to moulds.

Keywords: DMDHEU; High density fiberboard, Fast-growing poplar; Modification

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INTRODUCTION

Due to the dwindling round wood supply for the furniture and panel industry, non-classical raw materials such as waste wood, annual crops, or fast-growing species are feedstocks that are receiving greater attention (Abdul Khalil et al. 2010; Gatani et al. 2013; Varanda et al. 2013; Mirski et al. 2017). Plantations of fast-growing trees are a solution that may possibly quench the demand for wood, especially for grades other than timber (Balducci et al. 2008; Carle and Holmgren 2009). Poplar species have fast growth, great gain in volume, and short rotation periods (Deswal et al. 2014).

Fast-growing wood material usage is a trend in wood-based composites industrial development. This is an alternative way to overcome deficits in the long-term logistic planning of raw materials and is low in cost (Boruszewski et al. 2016). Poplar, willow, maple, birch, spruce, larch, Douglas fir, sweet cherry, and linden are the main tree species grown in forestry plantations around Europe. Various composite materials and wood-based panels production are made using fibers from fast-growing wood, especially poplar and willow species (Szostak et al. 2013), although the mechanical properties of fast-growing wood and its natural resistance against fungi are low (Strauss et al. 2004; Gao et al. 2017).
From 1960 to 1980, there was a large-scale establishment of poplar plantations (Populus spp.) in Poland for the paper industry (Przybysz and Przybysz 2013). Poplar trees are also widely used in bioenergy and environmental fields such as phytoremediation, soil carbon sequestration, and watershed protection. The poplar tree grows 1.5 m to 3.1 m per year. However, the growth rate depends on the location of the plantation and also determines the rotation time. On average, trees can be harvested every 2 years to 7 years (Stobrawa 2014). In North America, poplar wood is exploited in primary and secondary forest productions, including products such as lumber, composite panels, pallets, pulp and paper, furniture components, chopsticks, and fruit baskets. In Europe it is considered a valuable resource of fiber for the wood panel and paper industries (Balatinecz and Kretchmann 2001; Przybysz et al. 2018).

Currently, medium density fiberboard (MDF) and high density fiberboard (HDF) play an important role in various industrial applications, and their global production reached 98.4 million m$^3$ in 2015 (FAO 2015). HDF is known for its dense and smooth texture, easy processing, high stability, and suitability for surface applications. These advantages make HDF a convenient substrate for operations such as laminating, painting, varnishing, and veneering. HDF is widely used in fields such as furniture, flooring, outdoor and indoor decoration, hardboard substitution, electronic industry, wall panel, doors, and partitions (Ayrilmis 2007). However, the main raw material for HDF is pine wood, which is becoming tight in supply. Thus, it is reasonable to use fast-growing poplar in HDF manufacturing. As HDF is often a part of furniture that service under increased-humidity conditions, such as the bathroom and kitchen, the fiber needs to be modified to obtain higher water resistance.

To enhance properties of the HDF, the fiber was modified via impregnation with 1,3-dimethylol-4,5-dihydroxyethyleneurea (DMDHEU) resin, which is an efficient modifying agent for wood (Xie et al. 2013; Yuan et al. 2013; Mamiński et al. 2016; Han et al. 2017). Two N-methylol and two secondary hydroxyl groups per DMDHEU molecule are reactive towards hydroxyl groups in cellulosic fiber (Militz 1993; Yang et al. 2009). DMDHEU is effective in preventing degradation and fungal penetration into wood tissue, increasing water resistance, and improving durability and wood properties (Dieste and Krause 2009; Xie et al. 2013; Mamiński et al. 2016). In this work, the effect of modification of poplar fiber with DMDHEU on the physiochemical, mechanical, and biological properties of the resultant HDF panels was investigated.

**EXPERIMENTAL**

Poplar cultivar "Hybrid 275" P. maximowiczii x trichocarpa is grown on plantations, managed by State Forest National Forest Holding in Poland (wood density 365 kg/m$^3$, tree age 13 to 28 years, trunk diameter 16 cm to 18 cm). Defibration was performed in a fiberboard plant in Poland at typical industrial settings (pre-heating at 90 °C, steam pressure 8.5 bar, gap 0.22 mm). The fractional composition is shown in Fig. 1.
Fig. 1. Fiber fractional composition. Dimensions denote width of oblong hole in sieve that the fraction passes through.

Commercial urea-formaldehyde resin (solids 65%, viscosity 230 mPas at 20 °C) and 1% ammonium sulfate (based on resin solids) were used as a binder and hardener, respectively. Aqueous DMDHEU stock solution (pH 4.3; solids 34%; viscosity 27 mPas manufactured by BASF Chemicals, Ludwigshafen, Germany) was used as the modifying agent. The impregnation was done by soaking poplar fiber into 5% solution DMDHEU or 10% solution DMDHEU at room temperature for 30 min, and subsequently on the drying rack at ambient temperature overnight. Prior to HDF manufacturing, the impregnated poplar fiber was dried at 40 °C for 30 min to obtain a moisture content below 5%. The fiber was resinated with 10% (based on fiber dry weight) of the UF binder. Fibrous mats were hot-pressed at 180 °C with maximum unit pressure 2.5 MPa at 18 s/mm press factor to produce HDF panels with dimensions 300 mm × 300 mm × 3 mm (length × width × height) and approximately 850 kg/m³ density.

The produced panels were stored under room conditions (20 ± 2 °C, 65 ± 5% relative humidity) for 7 days prior to analysis. Modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB) were tested according to European standards EN 310 (1993) and EN 319 (1993). Mechanical tests were done using Instron 3369 universal testing machine (Instron Corp., Norwood, MA, USA). Density profile measurements were performed on an X-ray density analyzer GreCon Da-X (Fagus-Grecon Greten GmbH & Co. KG, Alfred-Hannover, Germany) with a scanning speed of 0.05 mm/s.

The samples were cut into specimens of dimensions 50 mm × 50 mm × 3 mm (length × width × height) and 3 mm × 50 mm × 150 mm (height × width × length) for density profile and for both MOR and MOE measurements, respectively. The IB test was done on specimens of dimensions 50 mm × 50 mm × 3 mm (length × width × height). Water contact angle was measured using a Phoenix 300 contact angle analyzer (Surface Electro Optics Co., Ltd., Suwon City, Korea) equipped with CCD camera and microscopic lenses. For this measurement, the sessile droplet method was done with the means of 20 measurements of average contact angles as well as spreading curves. Thickness swelling (TS) was measured according to EN 317 (1999). Water absorption (WA) was computed from the following Eq. 1,
\[ WA = \frac{m_f - m_i}{m_i} \times 100\% \]  

where WA (%) is water absorption, \( m_f \) is sample weight after soaking (g), and \( m_i \) is initial weight before soaking (g).

Preliminary fungal tests were performed on HDF specimens of dimensions 30 mm × 30 mm × 3 mm. Eight specimens were tested in each series: HDF from fibers impregnated in 5%-DMDHEU solution, HDF from fibers impregnated in 10%-DMDHEU solution, HDF untreated, and two series of untreated solid wood for comparison purposes: Scotch pine (Pinus sylvestris L.) sapwood and poplar (Populus tremula L.). In order to equalize the initial moisture content in all the specimens, sterilized specimens were soaked in demineralized water for 60 min and deposited on Petri dishes filled with Czapke Dox Agar medium enriched with 1% maltose. Glass spacers were applied for physical separation of the specimen from agar to avoid any migrations of nutrient ingredients from the substrate to the samples (Fig. 2).

![Fig. 2. Fungal test specimen arrangement](image)

The samples were inoculated by spraying with an aqueous suspension of a mixture of spores: Chaetomium globosum Kunze, Trichoderma viride Pers., Alternaria alternata (Fr.) Keissler, Aspergillus niger van Tieghem, Paecilomyces variotii Bainier, and Penicillium funiculosum Thom.

The moulds were allowed to grow at 27 °C for 10 days. The mycelium growth on the surface of samples was then examined according to visual assessment on a 10 point scale with 2-point intervals: 0, a specimen free from fungi; 2, below 10% of surface area infected, visibility only under microscope, and with possible dispersed single fruiting bodies of moulds; 4, below 50% of surface area infected, mycelium visible in naked eye, and dispersed grouped fruiting bodies of moulds; 6, up to 100% of surface area infected, numerous grouped fruiting of moulds visible in naked eye, and covering up to 50% of surface area; 8, 100% of surface area grown with mycelium, and 20% to 50% covered with abundant fruiting bodies of moulds; 10, 100% of surface area grown with mycelium and above 50% covered with abundant fruiting bodies of moulds. Midway performance was assigned in-between degrees.

Statistical analysis

The significance of the differences between average shear strengths was calculated using a Student’s t-test at a 95% confidence interval.
RESULTS AND DISCUSSION

X-Ray Density Profile

Figure 3 presents X-ray density profiles of the studied HDF panels. The profiles reveal changes in average density, which were 928 kg/m³, 872 kg/m³, and 784 kg/m³ for 0%, 5 %, and 10 % DMDHEU fiber treatment, respectively. There was a U-shaped curve for fiberboard. Higher densities at the beginning and end of the profile are due to higher compression of face layers, which is typical for fiberboards (Xie et al. 2011; Boruszewski et al. 2016). The lower density found for the panels made of DMDHEU-treated fiber is probably because of the higher density of a single fiber after impregnation, which lead to a reduced bulk density of a panel, increased fiber stiffness, and lesser susceptibility of mat to compression. These changes affected the bending properties of the panels, as shown in Fig. 3.

![X-ray density profile](image)

**Fig. 3.** X-ray density profiles of HDF panels made of the fiber modified with different DMDHEU concentrations

Physical Properties of Modified HDF

A commonly known drawback of HDF is low dimensional stability when exposed to high humidity conditions. The boards deteriorate due to swelling, which renders loss in coherence and, subsequently, loss in mechanical properties and damage. To overcome this disadvantage, a DMDHEU impregnation agent was used to act as a cross-linker for cellulose chains. The data in Table 1 indicates a reduction in TS and WA after 2 h and 24 h as DMDHEU concentration increased. The low hygroscopicity of wood fibers might be due to the cross-linking formation of hydroxyl groups between DMDHEU solution and wood fibers (Yusuf et al. 1995).

As cross-linking occurred, crystallinity increased, while amorphous region of the sample decreased. This resulted in reduced TS and WA (Yildiz and Gumuskaya 2006; Mohebby et al. 2008). As the concentration of DMDHEU increased, the TS and WA for the panels decreased. The results showed a reduction of up to 54% and 32% for TS and WA, respectively, after impregnation with 10% DMDHEU. This result is explained by DMDHEU penetration into fiber and cross-linking, the latter of which reduced the
interaction with water and enhanced fiber dimensional stability. A reduction in TS and WA was correlated with a decrease in water wetting (Yuan and Lee 2013). The observed water contact angles (Table 1) remained in accordance with those findings. Statistical analysis showed that the values observed for DMDHEU-modified panels were noticeably different from those for the controls.

Table 1. Physicochemical Properties of the Modified HDF

<table>
<thead>
<tr>
<th>Panel Type (% DMDHEU)</th>
<th>Thickness Swelling (%)</th>
<th>Water Absorption (%)</th>
<th>Water Contact Angle (*)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2 h</td>
<td>24 h</td>
<td>2 h</td>
</tr>
<tr>
<td>0</td>
<td>18.4 ± 1.3</td>
<td>27. ± 1.6</td>
<td>56.5 ± 5.1</td>
</tr>
<tr>
<td>5</td>
<td>10.5 ± 0.3</td>
<td>13.5 ± 0.4</td>
<td>44.9 ± 3.4</td>
</tr>
<tr>
<td>10</td>
<td>10.3 ± 0.5</td>
<td>12.6 ± 1.3</td>
<td>42.3 ± 2.2</td>
</tr>
</tbody>
</table>

Bending Properties of Modified HDF

Bending properties, such as MOE and MOR, are strongly dependent on the average density of the fiberboard (Halvarsson et al. 2008). A lower average density results in lower apparent mechanical properties, as illustrated in Fig. 4. However, due to the differences in densities of the produced panels, the specific MOE and MOR are more representative measures. The MOE and MOR for 0%, 5%, and 10% DMDHEU-treated HDF were 4.0 MPa/kg·m³, 4.2 MPa/kg·m³, and 4.2 MPa/kg·m³ and 0.059 MPa/kg·m³, 0.044 MPa/kg·m³, and 0.046 MPa/kg·m³, respectively. These values show that the modification increased MOE and decreased MOR. It has been documented that cross-linking within a fiber renders increased stiffness (Xie et al. 2007). The reduction in MOR observed for the modified boards was remarkable. Although the bending properties were affected by the modification, all modified HDFs exceeded the standard requirements for MOE and MOR, which are 2700 MPa and 23 MPa, respectively, according to EN 310 (1993).

![Fig. 4. MOE (a) and MOR (b) of the panels made of the fiber modified at different DMDHEU concentrations](image)

Internal Bonding (IB)

As the data in Fig. 5 indicate, the internal bonding (IB) of the modified HDF was apparently higher than that of the control series. The IB reflects homogeneity of the adhesion between the fibers, which are bonded by an adhesive, and also indicates single
fiber strengths against applied load during the test (Mohebby et al. 2008).

The IB required by EN 622 standard (2009) is 0.65 MPa for 3.0 mm thick general use boards. Thus, the IB for the modified HDF greatly exceeded that requirement, i.e. 1.0 MPa and 1.6 MPa for 5% DMDHEU and 10% DMDHEU, respectively. Regardless of low density, 10% DMDHEU HDF exhibited the highest IB, exhibiting enhanced mechanical properties of boards made of the modified fiber. Therefore, it is apparent that the impregnation of fibers in DMDHEU contributed to increased adhesive interactions between fiber and adhesive, which is consistent with published data (Han et al. 2015). Moreover, the mechanical properties of a composite made of a resin-impregnated woody material are often improved compared with the non-modified material (Manosuri et al. 2006). The enhancement results from the intercalation of cellulose macromolecules (Yang et al. 2009; Dong et al. 2015; Rahman et al. 2017).

![IB of panels made of the fiber modified at different DMDHEU concentrations](image)

**Fig. 5.** IB of panels made of the fiber modified at different DMDHEU concentrations

**Resistance to Fungal Attack**

The results of the HDF fungal test are shown in Table 2. Fiber impregnation with DMDHEU increased resistance of HDF to the attack of moulds. There is a positive effect of modification with DMDHEU on the resistance to biodeterioration of woody materials (Xie et al. 2005; Pfeffer et al. 2011).

**Table 2.** HDF Average Infection Degree after 10-day Mould Growth

<table>
<thead>
<tr>
<th>Specimen Type</th>
<th>Average Infection Degree (scale = 0 to 10)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDF 0% (untreated)</td>
<td>9</td>
</tr>
<tr>
<td>HDF treated with 5% DMDHEU</td>
<td>7</td>
</tr>
<tr>
<td>HDF treated with 10% DMDHEU</td>
<td>2</td>
</tr>
<tr>
<td>Scotch pine wood</td>
<td>1</td>
</tr>
<tr>
<td>European aspen wood</td>
<td>1</td>
</tr>
</tbody>
</table>
The present studies indicated that there is a threshold above which the protective effect becomes manifested. Impregnation with 5% DMDHEU gave a slight improvement in resistance to fungi (degree 7), while 10% DMDHEU resulted in an apparent inhibition of mould growth (degree 2); however, deterioration was not completely eliminated. Thus, it seems very likely that a DMDHEU concentration above 10% will provide full protection against moulds.

CONCLUSIONS

1. Poplar cultivar "Hybrid 275" (P. maximowiczii x trichocarpa) fiber was modified by impregnation with 1,3-dimethylol-4,5-dihydroxyethyleneurea (DMDHEU) and subsequently used in the manufacture of high density fiberboard (HDF).

2. Mechanical properties of the resultant HDF were affected by the modification. There was an increase in specific MOE and a reduction in specific MOR in comparison to the unmodified reference HDF. The determined mechanical properties meet the requirements of the EN 310 (1993) standard.

3. The internal bond (IB) of the modified HDF exceeded the requirements of EN 622 (2009) by 1.5-fold and 2.5-fold for 5% DMDHEU and 10% DMDHEU, respectively. This indicated that DMDHEU deposited within fibers significantly contributed to increased adhesive interactions within the panel.

4. Modifying the HDF with DMDHEU improved water resistance, which manifested in reduced thickness swelling, reduced water absorption, and increased water contact angle.

5. Fungal tests showed that DMDHEU improved fiberboard resistance to moulds.

6. The overall assessment of the results allow to conclude that the presented approach is a promising tool to develop new types of fiberboards of increased durability under severe conditions, and that fast-growing poplar species can be a convenient raw material for fiberboard industry.

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