

## PROPERTIES OF PARTICLEBOARDS MADE BY USING AN ADHESIVE WITH ADDED LIQUEFIED WOOD

### LASTNOSTI IVERNIH PLOŠČ, IZDELANIH Z UPORABO LEPILA Z DODANIM UTEKOČINJENIM LESOM

Nataša Čuk<sup>1,2</sup>, Matjaž Kunaver<sup>1,2</sup>, Sergej Medved<sup>3</sup>

<sup>1</sup>National Institute of Chemistry, Hajdrihova 19, SI-1000 Ljubljana, Slovenia

<sup>2</sup>Center of Excellence for Polymer Materials and Technologies, Tehnološki Park 24, SI-1000 Ljubljana, Slovenia

<sup>3</sup>Biotechnical Faculty, Department of Wood Science and Technology, Rožna dolina, Cesta VIII/34, 1000 Ljubljana, Slovenia  
natasa.cuk@ki.si

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In this study, three-layer particleboards were produced using liquefied wood in an adhesive mixture. The influence of two different formaldehyde resins: melamine-formaldehyde and melamine-urea-formaldehyde resin and two various catalysts: ammonium sulfate and ammonium formate on the particleboard properties was investigated. There were also two pressing parameters examined: temperature and time. The following physical and mechanical properties of particleboards were measured: board thickness, density, moisture content, bending strength and modulus of elasticity, internal bonding strength, surface soundness, thickness swelling and formaldehyde content. The results showed that properties of produced particleboards were better when melamine-formaldehyde resin and ammonium formate as a catalyst were used in combination with liquefied wood in the adhesive mixture. Also, mechanical properties were improved as the press time and press temperature increased. The optimal mechanical properties of particleboards made with the utilization of the liquefied wood in the adhesive mixture were achieved at 3 min press time and 180 °C press temperature using melamine-formaldehyde resin and 3 % of ammonium formate as a catalyst. A very important characteristic is the low formaldehyde content and that is extremely important in the provision of better quality of life. With the liquefaction of wood and the application of liquefied wood in the particleboard production wood biomass exploitation could be significantly increased. We are able to produce new materials from renewable resources that can be a great alternative to the raw materials, originating from crude oil.

Keywords: liquefied wood, renewable resources, particleboards, mechanical properties

Izdelali smo trislojne iverne plošče, kjer smo za pripravo lepila uporabili utekočinjen les. Ugotavljali smo, kako v kombinaciji z utekočinjenim lesom na lastnosti ivernih plošč vplivata dve različni formaldehidni smoli: melamin-formaldehidna in melamin-urea-formaldehidna smola, ter dva različna katalizatorja: amonijev sulfat in amonijev formiat. Ugotavljali smo tudi vpliv dveh parametrov stiskanja, in sicer temperature stiskanja in časa stiskanja. Fizikalne in mehanske lastnosti, ki smo jih ivernim ploščam določili, so bile: debelina plošče, gostota, vsebnost vlage, upogibna trdnost in modul elastičnosti, razslojna trdnost, čvrstost površine, debelinski nabrek in vsebnost prostega formaldehida. Rezultati so pokazali, da smo izboljšali lastnosti ivernih plošč, ko smo v kombinaciji z utekočinjenim lesom uporabili melamin-formaldehidno smolo in amonijev formiat kot katalizator. Ugotovili smo tudi, da so se lastnosti izdelanih ivernih plošč s časom in temperaturo stiskanja izboljšale. Optimalne lastnosti ivernih plošč, pri katerih smo lepilni mešanici dodali utekočinjen les, smo dosegli pri času stiskanja 3 min in temperaturi stiskanja 180 °C, pri uporabi melamin-formaldehidne smole in 3 % amonijevega formiata kot katalizatorja. Zelo pomembna pri izdelanih ivernih ploščah je tudi majhna vsebnost prostega formaldehida, kar močno vpliva na izboljšanje kvalitete bivanja. Z utekočinjanjem lesa in njegovo uporabo pri izdelavi ivernih plošč povečamo izrabo lesne biomase. Iz obnovljivih virov tako lahko sintetiziramo nove materiale, ki so odlična alternativa materialom, ki jih sicer pridobivamo iz surove nafte.

Ključne besede: utekočinjen les, obnovljivi viri, iverne plošče, mehanske lastnosti

## 1 INTRODUCTION

Wood is recyclable, renewable, biodegradable and one of the most abundant natural polymers. It can be used for many different purposes and converted to many useful industrial chemicals. Nowadays, it can also be liquefied using different polyhydric alcohols and acid catalysts. Cellulose, hemicellulose and lignin are the main wood components and during liquefaction these components are decomposed – depolymerized. The final product has lots of free hydroxyl groups and can therefore be used to prepare an adhesive for gluing the wood and wood composites. Particleboards for which we use 10–15 % of adhesive (based on the weight of particles) represent almost 60 % of the wood composites production.

To produce particleboards wood is broken down into particles of various size and glued together. Wood particles are bonded using synthetic adhesives and pressed into boards at high temperature and high pressure. The pressing operation provides increased density and strength.<sup>1</sup>

The properties of particleboards are engineered in a manner to meet many service requirements. Particleboards can be made in large panel sizes, with a full range of thicknesses. With the introduction of continuous presses, endless boards emerge and are subsequently cut into any length desired.<sup>2</sup> There are many factors affecting the properties of the particleboards and the most important are:

- species of wood, fiber structure, density, hardness, compressibility;

- form and size of raw wood;
- share of bark;
- non-wood lignocellulose materials;
- type and size of particles;
- method of particle drying;
- particle screening and separating, particle size distribution;
- type and amount of binding agents;
- method of mat formation, structure of particleboard;
- moistening of particles prior to pressing, final moisture content of board, conditioning;
- curing conditions;
- thickness of board;
- sand content of particleboard;
- surface quality;
- primings, coats of varnish or lacquer;
- laminating, veneering, overlaying.<sup>3</sup>

Over the years many different lignocellulosic materials beside wood were used in particleboard production: coconut chips,<sup>4</sup> paper sludge,<sup>5</sup> waste tea leaves,<sup>6</sup> castor stalks,<sup>7</sup> wheat straw,<sup>8</sup> flax shiv,<sup>9</sup> kenaf stalks,<sup>10</sup> needle litter,<sup>11</sup> waste grass clippings,<sup>12</sup> bagasse,<sup>13</sup> saline creeping wild rye,<sup>14</sup> peanut hull,<sup>15</sup> cotton carpel,<sup>16</sup> vine prunings,<sup>17</sup> kiwi prunings,<sup>18</sup> waste tissue paper and corn peel,<sup>19</sup> almond shell<sup>20</sup> and others.

Next to the commonly used urea-formaldehyde resin for particleboard manufacturing, there were also other adhesives applied: melamine-modified urea-formaldehyde resin,<sup>21</sup> phenol-formaldehyde resin and melamine-urea-phenol-formaldehyde resin,<sup>22</sup> PTP resin,<sup>23</sup> soy-protein adhesive,<sup>8, 24</sup> cement,<sup>25</sup> rice bran adhesive,<sup>26</sup> EMDI isocyanate resin,<sup>4</sup> MDI resin.<sup>8</sup>

Lee and Liu prepared resol resin based on liquefied bark and used it for particleboard production.<sup>27</sup> Particleboard made from liquefied Taiwan acacia bark, using H<sub>2</sub>SO<sub>4</sub> as a liquefaction catalyst, had the best properties in bending strength, internal bonding strength and thickness swelling. Liquefied wood was also used in preparation of phenol-formaldehyde resin,<sup>28</sup> polyurethane resin,<sup>29</sup> isocyanate adhesive<sup>30</sup> and epoxy resin.<sup>31</sup>

Since we have already proved in our previous research that liquefied wood can be used in the adhesive mixture for particleboard production,<sup>32</sup> the aim of this research was to evaluate the influence of some factors on the properties of particleboards made from liquefied wood. We investigated the impact of two different formaldehyde resins and two various catalysts used in combination with liquefied wood in the adhesive mixture on the properties of particleboards. We also examined the influence of two pressing conditions: temperature and time. Other parameters such as liquified wood loading, resin content and press pressure were held constant.

## 2 EXPERIMENTAL

### 2.1 Wood liquefaction

Sawdust of spruce wood (*Picea abies* (L.) Karst.) was liquefied using the mixture of glycerol and diethylene glycol (mass ratio 80/20,) as a liquefying reagent and p-toluenesulphonic acid as a catalyst. The mass ratio of wood sawdust to glycols was 1 : 3 and the amount of p-toluenesulphonic acid was 3 % based on the mass of glycols.

The mixture of glycols and acid was charged into three neck glass reactor equipped with mechanical stirrer, thermometer, condenser and external heating. When the mixture in the reactor reached temperature 160 °C wood sawdust was added. After all the wood sawdust was added the temperature was elevated to 180 °C and maintained for 90 min to carry out the liquefaction reaction. After the liquefaction the liquefied product was cooled down.

### 2.2 Adhesive preparation

The adhesive was prepared with the addition of liquefied wood (LW) to the melamine-formaldehyde (MF) or the melamine-urea-formaldehyde (MUF) resin. Both resins, MF and MUF, were received from Melamin Kočevje where they also measured the properties. The properties of MF and MUF resin are presented in **Table 1**. The liquefied wood loading was 30 % based on the mass of adhesive needed (dry content based), other 70 % was either MF or MUF resin. The mixture was thoroughly blended, then water and catalyst were added. The catalyst was only added into the adhesive mixture for the core layer. Catalysts used were 20 % solution of ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) and 10 % solution of ammonium formate (NH<sub>4</sub>HCO<sub>2</sub>). The catalyst content was 3 % based on the weight of the adhesive mixture, except when the influence of catalyst on the properties of particleboards was investigated. The whole resin mixture was blended for a few minutes before use.

**Table 1:** Characteristics of MF and MUF resin  
**Preglednica 1:** Karakteristike MF in MUF smole

Properties	MF	MUF
Solid content	(58 ± 1) %	(63 ± 2) %
Viscosity*	18–22 s	80–200 s
Free formaldehyde content	below 0.2 %	max. 0.5 %
pH value	9–10	9.2–9.5
Stability (at 20 °C)	20 d	2 months

\*viscosity was determined according to the standard DIN EN ISO 2431.  $\phi = 4$  mm, 20 °C

### 2.3 Particleboard production

Three-layer particleboards of 500 mm by 500 mm in size, 16 mm of thickness and 0.600 g/cm<sup>3</sup> of target density were produced in laboratory conditions. The

resin content was 10.50 % in the surface layer and 6.50 % in the core layer. The resin content was calculated based on the dry weight of wood particles.

Wood particles mixture composed from 75 % of spruce wood (*Picea abies* (L.) Karst.) and 25 % of beech wood (*Fagus sylvatica* L.) was first dried at 70 °C to obtain the moisture content approximately 4 %, then the resin was added. After blending for several minutes the glued wood particles were hand-formed into a mat using wood mold. For the determination of resin and catalyst type impact on the particleboard properties, the wood mat was hot-pressed for 3 min at temperature 180 °C. Later the press times were (2, 2.5, 3 and 3.5) min at the press temperature 180 °C and the press temperatures were 140 °C, 160 °C, 180 °C and 200 °C at the press time of 3 min. Before testing, all particleboards were conditioned for 4–5 d at temperature 20 °C and relative humidity 65 %.

#### 2.4 Particleboard testing

The physical and mechanical properties of produced particleboards were tested according to the European standards: the board thickness EN 324, the density EN 323, the moisture content EN 322, the bending strength and the modulus of elasticity EN 310, the internal bonding strength EN 319, the surface soundness EN 311, the thickness swelling EN 317 and the formaldehyde content EN 120.

### 3 RESULTS AND DISCUSSION

The results of the physical and mechanical properties of laboratory made particleboards bonded with LW combined with the MF and the MUF resin are presented in **Table 2**. The results indicate that mechanical properties of particleboards were up to 25 % better when MF resin was used. The reason may be that the resin containing more melamin is more reactive. Also, the cyclic structure of the melamine imparts greater stability to the resulting linkages. The three amine groups assure a three-dimensional, cross-linked molecular structure when fully cured.<sup>33</sup> Consequently, mechanical properties of particleboards using LW and MF resin were better in comparison to the particleboards where LW and melamine enhanced UF (MUF) resin was used. Nevertheless, in both cases, the properties satisfied the standard requirements. There was no major difference in thickness swelling with regard to the resin type used.

From previous research we have already found out that the addition of LW in the adhesive mixture reduces the content of formaldehyde in the boards.<sup>32</sup> In this research we have again obtained low formaldehyde content. We observed that formaldehyde content was lower when MF resin was used. Lower formaldehyde content at use of MF resin, compared to formaldehyde content at use of MUF is related to the nature of reaction between formaldehyde and melamine or formaldehyde

and melamine-urea, as it was reported by Eom et al.<sup>34</sup> Such boards can easily be used in dry conditions for general and interior purposes.

**Table 2:** Properties of particleboards produced by using an adhesive mixture made of LW combined with MF or MUF resin

**Preglednica 2:** Lastnosti ivernih plošč, izdelanih iz utekočinjenega lesa v kombinaciji z MF in MUF smolo v lepilni mešanici

Particleboard properties	LW-MF	LW-MUF
Board thickness (mm)	15.71	16.18
Density (g/cm <sup>3</sup> )	0.627	0.610
Moisture content (%)	7.98	7.70
Bending strength (N/mm <sup>2</sup> )	13.42	11.06
Modulus of elasticity (N/mm <sup>2</sup> )	2322	1823
Internal bonding strength (N/mm <sup>2</sup> )	0.47	0.45
Surface soundness (N/mm <sup>2</sup> )	1.28	1.18
Thickness swelling (%)	19.51	19.97
Formaldehyde content (mg/100 g of dry board)	1.40	2.55

**Table 3:** Properties of particleboards produced by using an adhesive with added LW and ammonium formate or ammonium sulfate as a catalyst

**Preglednica 3:** Lastnosti ivernih plošč, izdelanih iz utekočinjenega lesa, dodanega lepilni mešanici pri uporabi amonijevega formiata in amonijevega sulfata kot katalizatorja

Particleboard properties	NH <sub>4</sub> HCO <sub>2</sub>				(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>			
	1 %	2 %	3 %	4 %	1 %	2 %	3 %	4 %
Catalyst content (%)	1 %	2 %	3 %	4 %	1 %	2 %	3 %	4 %
Board thickness (mm)	15.58	15.70	15.71	15.69	16.04	15.83	15.53	15.71
Density (g/cm <sup>3</sup> )	0.658	0.643	0.627	0.660	0.637	0.642	0.659	0.651
Moisture content (%)	7.80	7.55	7.98	8.27	8.23	7.49	7.18	7.08
Bending strength (N/mm <sup>2</sup> )	12.37	12.71	13.42	12.70	10.48	11.96	12.98	11.71
Modulus of elasticity (N/mm <sup>2</sup> )	2286	2327	2322	2240	2091	2193	2404	2321
Internal bonding strength (N/mm <sup>2</sup> )	0.40	0.54	0.47	0.47	0.34	0.40	0.46	0.39
Surface soundness (N/mm <sup>2</sup> )	1.22	1.22	1.28	1.18	1.01	1.09	1.14	1.24
Thickness swelling (%)	20.82	18.77	19.51	18.20	21.62	20.74	21.33	22.00
Formaldehyde content (mg/100 g of dry board)	2.51	2.06	1.40	1.22	3.44	2.85	2.71	2.25

We also investigated the impact of type and amount of catalyst on the particleboard properties. We used two different ammonium salts: ammonium formate (NH<sub>4</sub>HCO<sub>2</sub>) and ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>). The results of particleboard testing are shown in **Table 3**. It can be seen that the properties of particleboards were better when ammonium formate was used. We also varied the quantity of the catalyst from 1 % to 4 %. The properties of particleboards were the best when the 3 % loading of each catalyst was applied. When comparing the efficiency of both catalysts and the influence of the amount of the catalyst the difference in thickness swelling is not very distinctive. Nevertheless, there are some differences in formaldehyde content. With higher amount of catalyst the formaldehyde content was lower

for the both catalysts. Furthermore, the formaldehyde content was higher when ammonium sulfate was used.

Further on, we evaluated the influence of pressing conditions, i.e. time and temperature, on the properties of particleboards made by using an adhesive with added liquefied wood. The results of testing particleboards pressed at different times are presented in **Table 4**. The prolongation of the press time resulted in an improvement of mechanical properties. The best properties were achieved at press time of 3 min. The time of pressing can be shorten to 2.5 min and consequently, the energy could be saved significantly. Namely, the reduction of press time means greater level of production, i.e. increased output, and consequently, the reduction of the unit production costs. Of course, press time reduction must be carefully watched because of the impact on the board quality. The press time had no larger effect on the thickness swelling and the formaldehyde content.

**Table 4:** Properties of particleboards produced by using an adhesive with added LW pressed at different times

**Preglednica 4:** Lastnosti ivernih plošč, izdelanih iz utekočinjenega lesa, dodanega lepilni mešanici pri različnih časih stiskanja

Particleboard properties	2 min	2,5 min	3 min	3,5 min
Board thickness (mm)	16.35	16.15	16.12	15.85
Density (g/cm <sup>3</sup> )	0.614	0.595	0.596	0.624
Moisture content (%)	6.18	5.97	5.43	5.53
Bending strength (N/mm <sup>2</sup> )	8.53	9.53	10.62	9.65
Modulus of elasticity (N/mm <sup>2</sup> )	1872	1806	2106	1904
Internal bonding strength (N/mm <sup>2</sup> )	0.37	0.52	0.46	0.48
Surface soundness (N/mm <sup>2</sup> )	0.95	0.94	1.03	0.93
Thickness swelling (%)	19.58	19.59	19.75	17.82
Formaldehyde content (mg/100 g of dry board)	3.49	2.94	2.83	3.02

**Table 5:** Properties of particleboards produced by using an adhesive with added LW pressed at different temperatures

**Preglednica 5:** Lastnosti ivernih plošč, izdelanih iz utekočinjenega lesa, dodanega lepilni mešanici pri različnih temperaturah stiskanja

Particleboard properties	140 °C	160 °C	180 °C	200 °C
Board thickness (mm)	17.51	16.47	16.12	16.27
Density (g/cm <sup>3</sup> )	0.592	0.629	0.596	0.628
Moisture content (%)	6.03	5.63	5.43	4.77
Bending strength (N/mm <sup>2</sup> )	8.10	9.34	10.62	10.99
Modulus of elasticity (N/mm <sup>2</sup> )	1655	1974	2106	2037
Internal bonding strength (N/mm <sup>2</sup> )	0.26	0.44	0.46	0.43
Surface soundness (N/mm <sup>2</sup> )	0.94	0.99	1.03	0.96
Thickness swelling (%)	19.86	20.56	19.75	20.44
Formaldehyde content (mg/100 g of dry board)	2.74	2.76	2.83	2.71

The results of testing of particleboards which were pressed at various temperatures are presented in **Table 5**. When the press temperature was elevated, the properties of particleboards were improved. The optimal properties were obtained at press temperature of 180 °C, while the

temperature of 140 °C was too low to achieve required strength and the temperature of 200 °C did not provide better quality of the board than of the 180 °C. The press temperature can be lowered to 160 °C and the properties would still satisfy the European standards requirements. The press temperature did not have much effect on both the thickness swelling and the formaldehyde content, what was also observed when evaluating the influence of the press time.

## 4 CONCLUSIONS

The three-layer particleboards were successfully produced using adhesive with the addition of 30 % of liquefied wood. The physical and mechanical properties of produced particleboards met the European standard requirements and were better when using melamine-formaldehyde resin and 3 % of ammonium formate. The optimal properties of particleboards were achieved at the press time of 3 min and the press temperature 180°C. By shortening the press time to 2.5 min or by decreasing the press temperature to 160°C reduction of the energy costs could be achieved. Nevertheless, the additional research will be needed to confirm these findings. Particleboards made with the adhesive with added liquefied wood have low formaldehyde content, and that is of special importance when using particleboards for interior applications. With the liquefaction of wood and application of liquefied wood in particleboard production the wood biomass exploitation can be increased. That would enable efficient lignocellulosic waste recycling, increase renewable resources usage and reduce the crude oil consumption.

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