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BONDING AND SURFACE FINISHING OF WOOD WITH LIQUEFIED WOOD

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Abstract

This paper is an overview of our recent investigations of liquefied wood (LW) for preparation and application of wood adhesives and wood surface finishes. For the purpose of wood bonding, poplar wood (Populus nigra L.) was liquefied with ethylene glycol (EG) and sulphuric acid. The process of the curing of LW at different conditions was characterized using differential scanning calorimetry (DSC). The effect of the press temperature and the pressing time on the shear strength of bonds was monitored after the bonding of 5mm thick beech (Fagus sylvatica L.) lamellas. According to the results of DSC measurements, the curing process of the LW took place in two phases. In the initial physical phase the removal of water and EG from the LW occurred. This phase was followed by an exothermic chemical reaction. It was found that the optimum conditions for bonding beech lamellas were: a ratio between poplar and EG of 1:1, a press temperature of 180°C, and a pressing time of 12 minutes. Using such conditions it was possible to obtain bonds with a dry shear strength of about $7N/mm^2$*. For preparation of polyurethane wood coatings, poplar wood was liquefied with diethylene glycol (DEG) and the excessive DEG was removed from the reaction mixture prior to curing with isocyanate type hardener. For the purpose of preparation of light-coloured polyurethane coatings from bleached liquefied wood, liquefaction of wood was conducted with a mixture of polyethylene glycol (PEG) and glycerol (GLY) at a 9:1 ratio. Both LW based PU coatings and bleached LW based PU finishes were in the most relevant properties comparable to commercial polyurethane wood coatings.*

Key words: *liquefied wood; bonding; shear strength; polyurethane coating; bleached liquefied wood.*

BONDING AND SURFACE FINISHING WITH LIQUEFIED WOOD

In the last decade, liquefied wood (LW) has been intensively investigated as a natural-based material that has the potential to be used for various applications. It is obtained in a liquefaction process where wood particles are dissolved at moderate temperatures (100-250°C) in organic solvents (e.g. phenol, polyhydric alcohols, cyclic carbonates, or ionic liquids), whereby partly catalysts can be added. During liquefaction the basic wood components (i.e. cellulose, lignin, and hemicelluloses) are degraded into oligomers or even monomers. In the area of wood science and technology, the major emphasis has been given on applications of liquefied lignocellulosics for wood adhesive resins and adhesives themselves (Tohmura *et al.* 2005, Lee and Lin 2008, Juhaida *et al.* 2010). For instance, the liquefied spruce wood was used as adhesives for wood particle boards (Antonović *et al*. 2010, Kunaver *et al.* 2010). There have been quite less reports on possible uses of liquefied wood and other lignocellulose sources in paint and especially in wood coating formulations. For example, Abdel-Mohdy and co-authors (2009) used a liquefied rice straw as a new resource for paint formulations, and it was also shown that liquefied wood could be utilized for preparation of polyurethane films (Lee *et al.* 2014).

OBJECTIVES

The most important objectives of our several year investigations in the field of wood liquefaction and utilization of liquefied wood were to find optimal liquefaction parameters, to study applications of liquefied wood in wood bonding and to prepare wood finishes on the basis of liquefied wood. So, the process of the liquefaction of black poplar (*Populus nigra* L.) using various polyalcohols (like ethylene glycol (EG), diethylene glycol (DEG, polyethylene glycol (PEG) etc.) as a solvent and sulphuric(VI) acid as a catalyst was optimized. The influence of press temperature and pressing time on the shear strength of LW bonds was determined as well, and the curing process of LW was characterised using differential scanning calorimetry (DSC). Preparation of polyurethane wood coatings based on liquefied wood and determination of their properties was also an important topic of our research. Because liquefied wood based products are of a dark brown or black colour, recently, polyurethane wood coatings from bleached liquefied wood, of an aesthetically acceptable light colour, were prepared and characterised.

MATERIALS AND METHODS

Liquefied wood for bonding

Before liquefaction, poplar wood was milled using a rotary mill in order to obtain fine fractions (<1mm), and dried for 24 hours at (103±2)°C. During the process of the optimization of liquefaction in a glass reactor, the wood particles were mixed with EG in five different mass ratios between poplar and EG: 1:1, 1:2, 1:3, 1:4 and 1:5. In all five cases the addition of sulphuric acid was 3 % based on the mass of the EG, and the temperature of the liquefaction was 180°C. The minimal liquefaction time for obtaining the best liquefaction yield was determined, as well as the optimal ratio between poplar and EG. After liquefaction, the LW was diluted with a mixture of 1,4-dioxane and water to ensure easier filtration. After filtration, the mixture of 1,4-dioxane and water was removed at 55°C and with a gradual rise in the reduced pressure from room pressure to 10 mbar, and then part of the EG was additionally removed at 120°C and at a reduced pressure of 10 mbar (Ugovšek *et al.* 2011).

The curing process of the LW was monitored with a high-pressure differential scanning calorimeter (HP DSC 1, Mettler Toledo). The measurements were carried out in 30 µl platinum crucibles in a high-pressure cell with a nitrogen atmosphere. STAR® software (Mettler Toledo) was used for the analysis of the thermograms and the data calculations (Ugovšek and Sernek 2013a).

Investigation of the influence of various parameters on the shear strength of bonds bonded with LW took place in a conventional hot press. The influence of press temperature was studied at temperatures of 150°C, 160°C, 170°C, 180°C, 190°C and 200°C for 15min, and the influence of pressing times of 3, 6, 9, 12, 15 and 18 minutes at 180°C was also investigated. Bonding of the 5mm thick beech lamellas and testing of the shear strength of the bonds was carried out in accordance with the requirements of the standard EN 205 (Ugovšek and Sernek 2013b).

Liquefied wood for wood finishing

The dust (0.24mm fraction) of black poplar wood (*Populus nigra* L.) was liquefied in 1000mL reactor (T= 180°C), where the following quantities of reactants were added: 100 g of wood, 500g of DEG and 15g of a catalyst – concentrated H_2SO_4. The yields of liquefaction reaction carried out for different periods were determined and the following step was removal of the excessive, unreacted DEG by rotary evaporation under reduced pressure (0.6 mbar) (Cheumani-Yona *et al*. 2012).

Liquefied wood without excessive DEG was used as the first component for preparation of polyurethane coatings (PU). Its viscosity was reduced by addition of water (3 mass portions of water per 1 portion of the liquefied wood). After 30 minutes of dispersion (15000rpm), the commercial YCM402 isocyanate hardener solution was added and 1,4-diazabicyclo [2.2.2] octane (DABCO) as a curing catalyst. The investigations were performed with two new formulations: the System 1, consisting of the mixture of diluted liquefied wood and the commercial solution of the hardener (1/1 w/w) and of 2% of DABCO; and the System 2 (liquefied wood/hardener 1/1.5, 2% of DABCO). For the purpose of comparison, a commercial water-borne two-component transparent polyurethane coating was used (the System 3). Adequately prepared solid beech wood plates were coated by conventional spraying. The Systems 1 and 2 were applied in 2 layers and the System 3 in 3 layers. Drying was executed at normal laboratory conditions. Properties of the films were determined 21 days after application. FT-IR spectra in the Attenuated Total Reflectance (ATR) technique were recorded by the Spectrum one (Perkin Elmer Instruments) spectrometer. Surfaces of the dried films of the Systems 1 and 3 were observed with the LV SEM (JEOL 5500 LV). It has to be noted that for the purpose of SEM observations the coatings were dried on aluminium foil rather than on solid wood substrates. The following properties were determined: resistance against cold liquids (SIST EN 12720:97), resistance against dry (SIST EN 12722:97) and wet heat (SIST EN 12721), flexibility (DIN 53155:1971), gloss

(SIST EN ISO 2813, for the Systems 1 and 2 only), and scratching resistance (SIST EN ISO 1518) (Cheumani-Yona *et al*. 2012).

For the matter of preparation of polyurethane coatings from bleached liquefied wood, liquefaction of wood was conducted in a 1-L reactor using a mixture of polyethylene glycol #400 (PEG) and glycerol (GLY) at a 9:1 mass ratio as the reactive solvent and sulphuric acid as the catalyst. The wood-to-solvent mass ratio was 1:3, and the catalyst-to-solvent mass ratio was 3:100. GLY, PEG #400, sulphuric acid, and wood sawdust were charged into the reactor and refluxed under continuous mechanical stirring for 120min at 150°C. After this time, the liquefied mixture was cooled down and diluted with a 1,4-dioxane/water mixture (4:1, v/v). The residue from liquefaction was removed by filtration under vacuum through a filter disk (Sartorius 388 grade, 12 to 15µm particles retention). Further on, a two-stage bleaching process was chosen. First, 100g of LW was diluted with 300g of a 1,4-dioxane/water binary mixture (4:1, v/v) in a 2-L reactor. Then, 33.33g of a 30% aqueous hydrogen peroxide solution (corresponding to a hydrogen peroxide-to-LW mass ratio of 10:100) was diluted three times in a 1,4-dioxane/water binary mixture (4:1, v/v), then added dropwise to the diluted LW for 24h at room temperature. The decomposition of hydrogen peroxide, was controlled by pH, i.e., by sulphuric acid (the residue of the catalyst used for liquefaction) and carboxylic acids formed during the liquefaction reaction. After the first 24h, the pH of the reaction mixture was adjusted to 8.5 with a 40% aqueous sodium hydroxide solution to allow the decomposition of the residual peroxide. The reaction mixture was kept at room temperature for another 24h. This step was important because of the high stability of hydrogen peroxide in acidic media. The direct evaporation of the solvent under reduced pressure after this step could be especially dangerous because of the (explosive) decomposition of the residual hydrogen peroxide during the process. After the second 24h of treatment, the residual sodium salts were removed by filtration under vacuum through a filter disk (Sartorius 388 grade, 12 to 15µm particles retention), because the presence of these salts could have a negative impact on the properties of cured polyurethane films on the basis of liquefied wood (Cheumani-Yona *et al*. 2015).

Pure liquefied wood or bleached liquefied wood were dosed with an appropriate amount of polyisocyanate (given by the -NCO/-OH ratio), as described by Cheumani-Yona *et al.* (2015) and ethyl acetate (EA), then thoroughly mixed in a polyethylene cup with a glass rod. The mass of EA added in each run was 10% of the overall mass of LW and polyisocyanate. The mixtures were applied to a glass plate and to a beech wood substrate using a film applicator. The thickness of the PU films was 120µm. Curing of the films was performed at room temperature. All of the films were characterised after seven days of curing, similarly as described above and by Cheumani-Yona *et al*. (2015).

RESULTS AND DISCUSSION

Optimization of liquefied wood for bonding

It was found that poplar wood can be successfully liquefied using EG as a solvent and sulphuric acid as a catalyst. Under optimal conditions, the liquefaction yield was more than 90%. The optimal parameters for the liquefaction of poplar with EG were as follows: a mass ratio of 1:3 between poplar and EG, and a pressing time of 120 min at 180°C. After the liquefaction process, which was followed by filtration, two thirds of the initial quantity of EG was removed by evaporation at 120°C, at a reduced pressure of 10 mbar. By means of this procedure the final LW with a mass ratio between poplar and EG of 1:1 was obtained (Ugovšek *et al.* 2011). A lower percentage of EG in the LW had a positive effect on the final shear strength of bonds compared to LW with a ratio of 1:3. The shear strength of the bonds after 50 days of conditioning in a standard climate was greater in the case of LW with a ratio of 1:1 (6.4N/mm^2) than in the case of LW with a ratio of 1:3 (5.6N/mm^2). This optimized LW with a mass ratio between poplar and EG of 1:1 was used for further studies.

Curing of liquefied wood

A DSC thermogram of the curing of LW is presented in Fig. 1. The curing process of the LW depended on the elimination (evaporation) of water (first endothermic signal) and EG (second endothermic signal). After the sufficient elimination of water and EG (185°C) the chemical reaction occurred. The latter is represented as an exothermic signal betweeen 185°C and 290°C, with a peak at around 260°C. At higher temepratures, decomposition of the LW occurred (Ugovšek and Sernek 2013a).

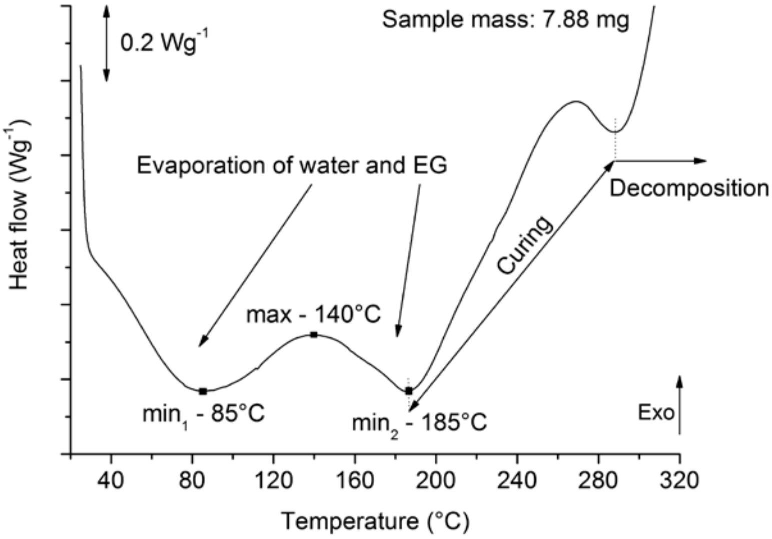

Fig. 1.
Analysis of the physical transitions and chemical changes taking place during the curing of LW (Ugovšek and Sernek 2013a)

The influence of press temperature and pressing time on the shear strength of LW bonds

Both press temperature (Fig. 2) and pressing time (Fig. 3) significantly influenced the shear strength of bonds made with LW. It was found that a temperature of 180°C and a pressing time of 12min were optimal for the bonding of 5mm thick beech lamellas with the LW used in this study. Shear strength of the LW bonds in dry conditions of around 7 N/mm^2 was ensured, whereas the percentage of wood failure was high (around 100%). However, the shear strength values of the bonds did not satisfy the requirements of standard EN 12765:2002 for thermosetting adhesives for non-structural applications in dry conditions (Class C1), which requires shear strength of 10N/mm^2. The LW bonds also showed poor resistance to water and humidity (Ugovšek and Sernek 2013b).

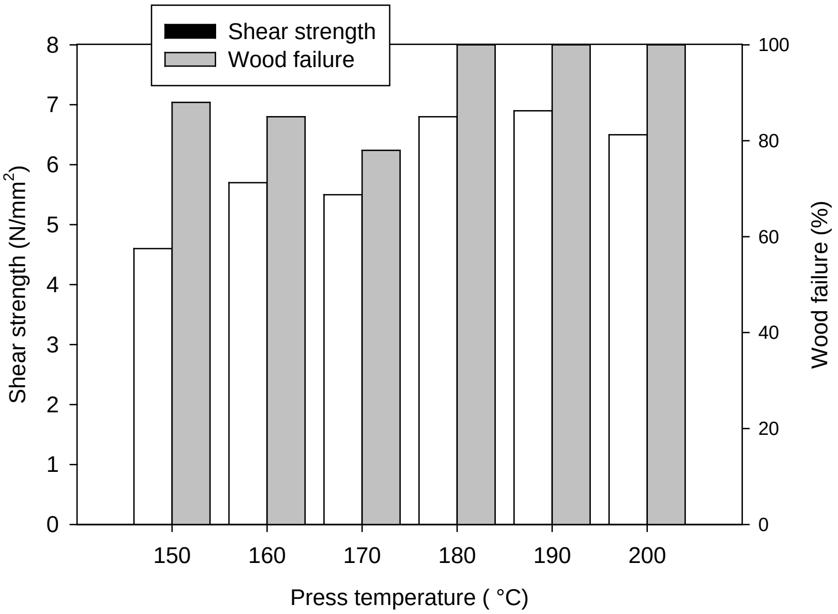

Fig. 2.
Influence of press temperature on shear strength and wood failure of specimens bonded with liquefied wood after 7 days of exposure to standard climate (Ugovšek and Sernek 2013b)

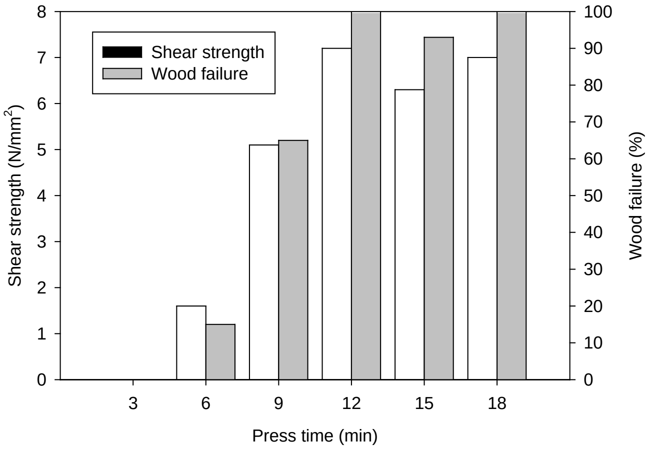

Fig. 3.
Influence of pressing time on shear strength and wood failure of specimens bonded with liquefied wood after 7 days of exposure to standard climate (Ugovšek and Sernek 2013b)

The structure of the bond resulting from the bonding of beech lamellas with LW was found to be very atypical compared to the structure of bonds obtained when bonding with synthetic adhesives for wood. It was found that the LW caused changes in the chemical structure of the cell walls of the wood cells of the beech lamellas that were in contact with the LW. In the initial stage of the bonding, in the case of lower press temperatures (up to 160°C) and a shorter pressing time (up to 6min) a phenomenon similar to that which takes place in the process of the liquefaction of wood occurred: a degradation of lignin or partial delignification, whereas cellulose remained the basic component of the cell walls. In the following phase of bonding (at temperatures above 180°C), these partially delignified cells underwent partial carbonisation due to the presence of sulphuric acid in the LW and a reduced amount of oxygen in the bond. The bond formed during the bonding of wood with LW therefore consisted of two parts. The central part of partially carbonised wood cells of the bonded lamellas and the products generated during the curing of LW, and a narrow layer of partially delignified cells at the boundary between the intact tissue of the bonded wood lamellas and the central carbonised layer. The reason for the low shear strength at a relatively high percentage of wood failure, and for the poor resistance of the bonds to water, was the already mentioned boundary layer of partially delignified cells, which mainly consisted of cellulose (Ugovsek *et al.* 2013).

Properties of polyurethane coatings made from poplar wood, liquefied with DEG

It was shown (Cheumani-Yona *et al*. 2012) that liquefied poplar wood could be used to prepare polyurethane wood coatings that are by their properties comparable to those of commercial ones Table 1). It was succeeded to remove the major part of the excessive reactive solvent – diethylene glycol from the liquefaction mixture, with a consequence of lower amount of the isocyanate hardener needed. This represents a novelty in research of preparation of polymers on the basis of liquefied wood.

Table 1
Average assessments of resistance of the coating Systems 1, 2 and 3 against various agents, wet and dry heat, scratching, their gloss and flexibility

<table>
<tr><th>Resistance against</th><th>Time of exposure</th><th colspan="4">System 1</th><th colspan="4">System 2</th><th colspan="4">System 3</th></tr>
<tr><td>Distilled water</td><td>24 hours</td><td colspan="2">2/3</td><td colspan="2">2</td><td colspan="2">2</td><td colspan="2">2</td><td colspan="2">5</td><td colspan="2">5</td></tr>
<tr><td>Oil</td><td>24 hours</td><td colspan="2">5</td><td colspan="2">5</td><td colspan="2">5</td><td colspan="2">5</td><td colspan="2">5</td><td colspan="2">5</td></tr>
<tr><td>Coffee</td><td>1 hour</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">5</td><td colspan="2">5</td></tr>
<tr><td>[redacted]</td><td>1 hour</td><td colspan="2">3/4</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td></tr>
<tr><td>[redacted]</td><td>2 min</td><td colspan="2">5/4</td><td colspan="2">5/4</td><td colspan="2">4/3</td><td colspan="2">4/3</td><td colspan="2">3/4</td><td colspan="2">3</td></tr>
<tr><td>[redacted]</td><td>20 min</td><td colspan="2">5</td><td colspan="2">5</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">5</td><td colspan="2">5</td></tr>
<tr><td>[redacted]</td><td>20 min</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">3</td><td colspan="2">4</td><td colspan="2">4</td></tr>
<tr><td>[redacted]</td><td></td><td colspan="4">5.3</td><td colspan="4">10.0</td><td colspan="4">Not measured</td></tr>
<tr><td>[redacted]</td><td>Force (N)</td><td>5</td><td>10</td><td>15</td><td>20</td><td>5</td><td>10</td><td>15</td><td>20</td><td>5</td><td>10</td><td>15</td><td>20</td></tr>
<tr><td></td><td>Track (mm)</td><td>0</td><td>0</td><td>0</td><td>0.5</td><td>0</td><td>0</td><td>0.3</td><td>0.5</td><td>0</td><td>0.3</td><td>0.4</td><td>0.6</td></tr>
<tr><td>[redacted]</td><td></td><td colspan="4">1</td><td colspan="4">1</td><td colspan="4">1</td></tr>
</table>

Properties of polyurethane coatings made from bleached liquefied wood

The bleaching treatment converted liquefied wood, originally dark brown, to a yellowish product, as shown by the CIE *L*a*b** parameters (Table 2) (Cheumani-Yona *et al*. 2015).

Table 2

Change of Colour of Liquefied Wood after Hydrogen Peroxide Treatment

Sample	Colour parameter			
	*L**	*a**	*b**	*ΔE**[1]
Liquefied wood (LW)	*28.3*	*0.2*	*0.1*	*0.0*
Bleached liquefied wood (LWB)	*54.3*	*8.5*	*40.7*	*48.9*

[1] *ΔE**: colour difference between the liquefied wood (LW) and of the bleached liquefied wood (LWB)

The mechanical properties and resistance to cold liquids (water and alcohol) of PU films prepared from liquefied wood and bleached liquefied wood are presented in Table 3. It has to be noted that there are only few reports on liquefied wood based polyurethane films in literature (for instance Lee *et al.* 2014) and up to our best knowledge, this was (Cheumani-Yona *et al.* 2015) the first report on polyurethane films made from bleached liquefied wood. The properties of the bleached liquefied wood based PU films were all comparable to the properties obtained with non-bleached liquefied wood based films. Generally, these results suggest that the bleaching pretreatment could be applied to liquefied wood before its use in the preparation of PU without any harmful effects on the properties of the cured PU coatings based on liquefied wood, what was believed to be an important new finding.

The mechanical properties of the TMP/TDI-based PU films were acceptable for coating applications, especially the films prepared with the -NCO/-OH ratio of 1.25. Quite high pendulum hardness values (> 78s) were measured for these films. The scratching resistance values of the films were above 16N. For TMP/TDI (at a ratio of 1.25) LW- and LWB-based PU films (Table 3), the elasticity was optimal, and, at the same time, the adhesion strength on the beech substrate was higher than 3MPa.

Table 3

Sample[1]	Pendulum hardness (s)	Resistance to scratching (N)	Elasticity[2]	Adhesion strength[3] (MPa)	Resistance to liquid[4]	
					Water (1 h)	Alcohol (1 h)
LW/PMDI 1	8.4	3	1.1	2.27A	3	3
LW/PMDI 1.25	14.0	14	1.1	3.77A	2	2
LW/TMP/TDI 1	37.7	18	1.1	3.97A	3	3
LW/TMP/TDI 1.25	78.1	20	1.1	3.90A	3	3
LWB/PMDI 1	12.7	9	1.1	2.8A	3	3
LWB/PMDI 1.25	12.6	14	1.1	4.14A	3	3
LWB/TMP/TDI 1	38.1	14	1.1	4.42 (A)	3	3
LWB/TMP/TDI 1.25	85.9	16	1.1	3.38A	3	4

[1] LW:liquefied wood based film; LWB: bleached liquefied wood based film; PMDI or TMP/TDI: isocyanate-type curing agent; 1 or 1.25: the NCO/OH ratio

[2] Assessment of elasticity from 1.1 (the best - large elasticity) to 3 (no elasticity - high hardness)

[3] A: adhesive type of failure between the coating and the substrate occurred

[4] Assessment of the resistance to liquids from 1 (no resistance) to 4 (acceptable resistance) and 5 (the best resistance)

CONCLUSIONS

It was determined that the mass ratio between ethylene glycol and wood of 1:3 during liquefaction for 120 minutes at 180°C was optimal in terms of the highest liquefaction yield achieved. The preparation of low solvent liquefied wood was performed with ethylene glycol evaporation at 120°C and at reduced pressure. The final mass ratio obtained between the solvent and wood was 1:1 and this liquefied wood was most promising for bonding and further studies. Differential scanning calorimetry revealed that the curing of liquefied wood can be divided into two parts. In the first part, water and ethylene glycol were evaporated, whereas in the second part a chemical reaction of the liquefied wood occurred. It was found that the pressing time of 12 minutes at 180°C was determined to be optimal for the bonding of the 5mm thick beech wood lamellas with LW used in this study.

Polyurethane wood coatings were also successfully prepared from liquefied wood, with their most important properties comparable to those of commercial coatings. When diethylene glycol (DEG) is used as a reactive solvent, this can be removed by vacuum evaporation after liquefaction. This results in decreased amounts of isocyanate hardeners needed for preparation of polyurethane films, but without a negative influence on their properties.

Further on, liquefied black poplar wood was obtained by solvolysis in a polyethylene glycol/glycerol mixture, and it was bleached with hydrogen peroxide. The bleaching treatment converted liquefied wood from a dark brown to a yellowish product. Polyurethane films were prepared by the curing of liquefied wood with polymeric diphenylmethane diisocyanate or trimethylolpropane toluene diisocyanate prepolymer (TMP/TDI) isocyanate-type hardeners, and the selected properties of the films prepared from the bleached liquefied wood were, in general, equivalent to those prepared from unbleached liquefied wood.

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