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APPLICATION OF SELECTED AGENTS FOR WOOD LIQUEFACTION AND SOME PROPERTIES OF PARTICLEBOARDS PRODUCED WITH THE USE OF LIQUEFIED WOOD

This work presents the characteristics of wood liquefied using different types of solvents in terms of its application for binding particleboards. Standard pine particles from barked wood were used for the liquefaction experiments. The liquefaction reaction was carried out in high temperature conditions using a mixture of solvents from the polyhydroxy alcohol group, including glycerine, ethylene glycol, propylene glycol, diethylene glycol, and dipropylene glycol. The microstructure of both the liquefied wood and the liquefaction residues was determined by means of optical microscopy analysis. The basic parameters of the adhesive mixture modified with the liquefied wood, such as viscosity, pH and gel time were determined. Particleboards containing liquefied wood were produced. The following physicochemical and mechanical properties of the particleboards were measured: tensile strength, bending strength, modulus of elasticity, and formaldehyde content. The influence of the liquefying agent on the board properties was investigated. In all the tests, a control particleboard, bonded with a urea-formaldehyde adhesive resin with no inclusion of liquefied wood, was used for the purposes of comparison.

Keywords: liquefied wood, adhesive, particleboard properties, confocal microscopy

Introduction

Recently there has been growing interest in research on liquefied wood and its possible applications. Thus far, liquefied wood has been used, among other things, in the production of polyurethane foams [Ertaş et al. 2014, Čuk et al. 2015a] and coatings [Hrastnik et al. 2014, Cheumani-Yona et al. 2015], for the preparation of activated carbon fibres [Li and Ma 2013, Liu et al. 2015], as

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a fuel [Seljak et al. 2012] and as a urea- and melamineureaformaldehyde resin substitute or modifier [Kunaver et al. 2010, Esteves et al. 2015, Janiszewska et al. 2016]. Recently Čuk et al. [2015b] carried out research which involved producing particleboards bonded with melamine-formaldehyde resin modified with liquefied wood. The research indicated that particleboards made with an adhesive mixture containing even 30% liquefied wood displayed the same desired parameters as a board made without the inclusion of liquefied wood. Other similar research has also indicated that liquefied wood affects the emission of formaldehyde from particleboards. Medved et al. [2009] and Kunaver et al. [2010] have proved that a 30% substitution of synthetic resin with liquefied wood can decrease formaldehyde emission by ca 40%. Of the best-known liquefying agents from the polyhydroxyl alcohols, the most used is a mixture of glycerine and diethylene glycol. In this study, different types of liquefying agents were used, including the less toxic propylene glycol, in order to investigate the influence of the tested liquefying agent on the properties of particleboards produced using liquefied wood. A decision was also taken to assess the suitability of laser scanning confocal microscopy, which as yet has not been fully described in the research on liquefied wood.

The aim of the research was to determine selected physicochemical properties, including confocal microscopy analysis of wood liquefied using different types of polyhydroxyl alcohol group solvents and liquefaction residues, as well as to investigate the standard properties of particleboards produced with the addition of liquefied wood.

Materials and methods

The raw wood material consisted of standard pine particles from barked wood. The particles were sorted using an Allgaier vibration screening machine with a set of screens with mesh diameters of 8, 2, 1 and 0.5 mm, respectively. Particles ≤1 mm and ≥0.5 mm were selected as the usable fraction for the liquefaction experiments. Before use, the particles were dried at 103°C for 24 h. The polyhydroxyl alcohol mixture of glycerine-G, ethylene glycol-EG, propylene glycol-PG, diethylene glycol-DEG, and dipropylene glycol-DPG were used as a liquefying agent. *P*-toluenesulfonic acid acted as a catalyst. The glycerol, ethylene glycol, propylene glycol, diethylene glycol, and 1,4-dioxane were obtained from Chempur, while the dipropylene glycol and *p*-toluenesulfonic acid (monohydrate) were acquired from Alfa Aesar. All the chemicals and solvents were of synthesis grade and were used without further purification. The mixture of glycerine and ethylene glycol G-EG (G-PG, G-DEG etc.) 1:1 by wt and *p*-toluenesulfonic acid (3% by wt for the liquefying agent) were placed into a 2000 cm^3 three-necked glass reactor equipped with a mechanical stirrer. The mixture was heated to 130°C and was stirred constantly. Wood particles were then gradually added. The liquefaction reaction

was carried out for 2 h at 130-140°C. The mixture was diluted with a dioxane/ water solution (4:1 v/v) after the reaction was finished. The product was separated from the solid residues by vacuum filtration. The residues were rinsed with the dioxane and oven dried at 103°C for 24 h. The water and dioxane were evaporated under reduced pressure.

The microscopic investigations were conducted in the Department of the Physics of Liquid Crystals at the Institute of Molecular Physics of the Polish Academy of Sciences. A drop of liquefied wood and dry liquefication residues were put onto the microscopic slides. The microstructural analysis of the samples was performed using an Olympus BX53 polarizing microscope (PM) and an Olympus FluoView FV1200 IX83 laser scanning fluorescence confocal microscope (LSFCM). For all the studied samples, a magnification of 10× was used. As a source of coherent light, a diode laser with two excitation beams of 559 nm and 635 nm was used.

A mixture of an industrial urea-formaldehyde resin (80%) and liquefied wood (20% relative to the dry weight of the resin) was prepared to use as the binder. The industrial urea-formaldehyde (UF) glue resin was characterised by the following parameters: a gel time of 75 seconds, a viscosity of 336 mPa·s, a dry mass content of 69.4%, and a pH of 7.3. Urea-ammonium nitrate solution (46%) was used as a curing agent, constituting 1% of the dry weight of the resin. The viscosity, pH and gel time at 100°C characterising the adhesive mixture were determined according to the following standards: EN 12092:2004, EN 1245:2011 and PN-C-89352-3:1996. Each value is an average from three parallel experiment.

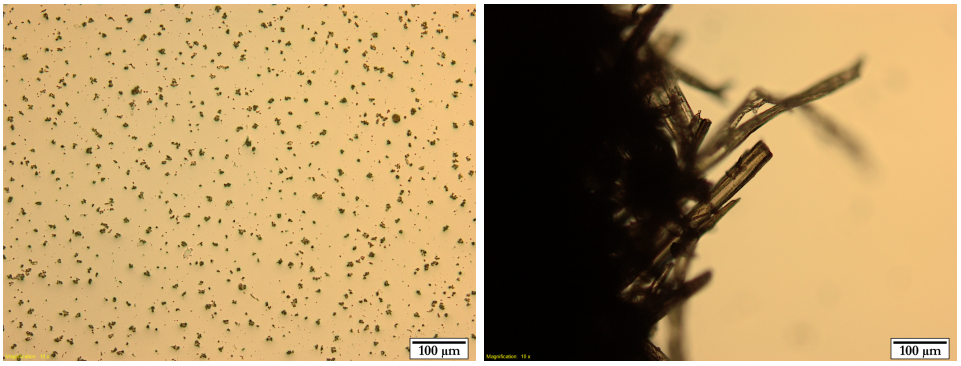
The particle fraction ≤8 mm and ≥1 mm was intended for particleboard production. In order to characterize the raw wood material, a determination of the fraction composition and a measurement analysis of the particles were carried out. After the sorting process, the determination of the fraction composition of the particles was carried out on the particles dried to a moisture content of approx. 8-10%. The test was conducted using a Fritsch screening machine. Approx. 200 g of raw material was taken randomly from a given particle portion. The mean share of the fraction was as follows: 4.00 mm – 4.6%, 2.00 mm – 18.3%, 1.00 mm – 49.4%, 0.50 mm – 25.6%, 0.25 mm – 1.6%, and <0.25 mm – 0.5%. Approx. 200 pieces of raw material were taken randomly from a given particle portion in order to undergo dimensional analysis. The average dimensions of the particles were: length 9.58 mm, width 1.82 mm, thickness 0.83 mm, slenderness coefficient 11.5 and flatness coefficient 2.2.

Single-layer particleboards measuring 510 × 510 × 10 mm with a 20% share of liquefied wood were produced. Pressing was performed at 200°C, at a unit pressure of 2.5 N/mm^2 reduced to 1 N/mm^2 after 41 s, to 0.5 N/mm^2 after 50 s and to 0 N/mm^2 at the end of the pressing process. The pressing time coefficient was 6.5 s/mm, while the resination rate was 10%. The nominal density of the panels was 650 kg/m^3. The standard mechanical and physicochemical properties

of the particleboards were examined according to the standards: EN 323:1999, EN 310:1994, EN 319:1999, and EN 120:1994. Two parallel tests of one type of the board were carried out. A particleboard bonded with urea-formaldehyde adhesive resin without a share of liquefied wood (Control) was used for comparison purposes in all the tests. The boards were conditioned at 20°C and at 65% relative humidity.

Results and discussion

The optical microscopy studies conducted on the pine particles liquefied with the G-PG mixture (fig. 1a), as well as for the liquefaction residues (fig. 1b), showed differences in the images of the microstructures of the samples. The microstructure images of the wood liquefied with the aid of propylene glycol and registered by means of a confocal fluorescence microscope (fig. 2a) indicated the liquefaction of almost all the lignin and remaining of the cellulose in the residues. The structure of the liquefied wood was crosslinked and showed strong fluorescence in regions where no objects observed by optical microscope were present. In contrast, the sample of the residue showed poor luminescence in all the investigated regions, indicating a high cellulose content (fig. 2b).

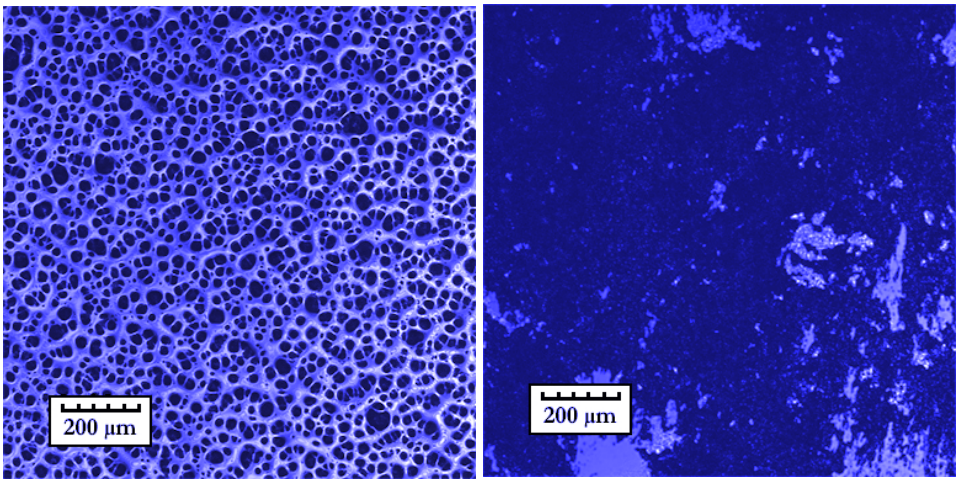


a b

Fig. 1. Optical microscopy image of G-PG; a – liquefied wood, b – residue (886 × 670 µm), magnification 10×

The physicochemical properties such as the viscosity, pH and gel time of the adhesive mixture based on liquefied wood are given in table 1.

The gel time of the adhesive mixture was in the range of 41-55 s for all the liquefying agents. The inclusion of liquefied wood in the adhesive mixture decreased the gel time by ca 30 s. Additionally, the pH value of the liquefied wood itself was between 2-3, which may increase the adhesive crosslinking process [Medved et al. 2009]. The greatest increase was observed in the viscosity of the prepared adhesive mixture G-DPG in comparison to the standard UF resin.



a **b**

Fig. 2. LSFCM image of G-PG; a – liquefied wood (1276 × 1276 µm), b – residue (3140 × 3140 µm), magnification 10×

Table 1. Physicochemical properties of adhesive mixture based on liquefied wood

Adhesive mixture modified with 20% liquefied wood	pH [-]	Viscosity [mPa·s]	Gel time [s]
G-EG	5.6	299	49
G-PG	5.4	335	41
G-DEG	5.9	345	55
G-DPG	5.7	388	50

Gel time for adhesive mixture without liquefied wood: 75 s

The properties of wood-based panels based on liquefied wood are presented in table 2.

For all tested liquefying agents, it was observed that the tested particleboards displayed desirable properties in comparison to the control particleboard produced without a share of liquefied wood. The density profile of the standard board and the boards prepared with the use of liquefied wood was similar, therefore the influence of that parameter on the mechanical properties of the board was excluded. The research conducted showed that the replacement of urea-formaldehyde resin with 20% of liquefied wood did not cause an increase in the formaldehyde content.

Table 2. Properties of wood-based panels based on liquefied wood

Tested property	Value	Measure unit	Board symbol				
			Control	G-EG	G-PG	G-DEG	G-DPG
Tensile strength	x	N/mm^2	0.86	0.82	0.81	0.73	0.74
	s		0.06	0.12	0.12	0.08	0.08
	v	%	7.0	14.6	14.8	11.0	10.8
	n	pcs	8	7	8	8	8
Sample's density for tensile strength test	x	kg/m^3	654	653	652	627	654
	s		14	21	9	13	11
	v	%	2.1	3.2	1.4	2.1	1.7
	n	pcs	8	7	8	8	8
Bending strength	x	N/mm^2	15.6	15.5	15.9	15.0	13.8
	s		2.9	2.0	1.6	1.0	0.7
	v	%	18.6	12.9	10.1	6.6	5.1
	n	pcs	8	8	8	8	8
Modulus of elasticity	x	N/mm^2	2663	2797	2780	2723	2584
	s		180	375	182	166	95
	v	%	6.8	13.4	6.5	6.1	3.7
	n	pcs	6	7	6	7	7
Sample's density for bending strength and modulus of elasticity tests	x	kg/m^3	642	629	653	637	630
	s		14	17	20	10	14
	v	%	2.2	2.7	3.0	1.5	2.2
	n	pcs	6	6	6	6	5
Formaldehyde content	mg/100g dry board		8.1	6.4	8.1	7.2	7.1
Moisture content	%		5.4	5.2	5.4	5.1	5.5

x – mean value, s – standard deviation, v – coefficient of variation, n – number of samples taken for the test

Conclusions

The preliminary experimental results showed that the tested particleboards based on liquefied wood displayed mechanical properties which were comparable to the control board without liquefied wood, irrespective of the type of liquefying agent used. A slight decrease in the bending strength was observed when G-DPG

was used as the liquefying agent, but this parameter still fulfilled the demands of the European quality standard. In addition, the usefulness of laser scanning confocal microscopy in the research on liquefied wood was demonstrated. The microstructural image of the wood liquefied with the aid of glycerol and propylene glycol, registered by means of a confocal fluorescence microscope showed strong fluorescence, which indicated that almost all the lignin was liquefied and the cellulose remained in the residues.

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List of standards

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EN 1245:2011 Adhesives. Determination of pH
PN-C-89352-3:1996 Kleje-Oznaczanie czasu żelowania (Adhesives. Determination of gel time)
EN 310:1994 Wood-based panels. Determination of modulus of elasticity in bending and of bending strength
EN 319:1999 Particleboards and fibreboards. Determination of tensile strength perpendicular to the plane of the board
EN 323:1999 Wood-based panels. Determination of density
EN 120:1994 Wood-based panels. Determination of formaldehyde content

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