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PARTICLEBOARDS WITH PARTIALLY LIQUEFIED BARK OF DIFFERENT PARTICLE SIZES

This paper presents a novel method of partially liquefying bark sawmilling waste for use in making particleboards. Maritime pine (Pinus pinaster Ait.) bark of different particle sizes (fine, medium, coarse, and mixed) was partially liquefied in the presence of ethylene glycol as a solvent and sulphuric acid as a catalyst at 180°C for 30 minutes. Single-layer particleboards were prepared by mixing partially liquefied bark (PLB) and wood chips at a ratio of 0.25 with no adhesives (group A) and at ratios of 0.25 or 0.1 with melamine-urea-formaldehyde (MUF) adhesives for additional bonding (groups B and C respectively). Mechanical and physical properties of the particleboards were tested according to European standards. The results showed that the boards in group A had lower densities, inferior mechanical properties and higher moisture content than those in groups B and C. Bark particle size had a significant effect on the mechanical properties of particleboards within each group. Additional MUF bonding and avoidance of coarse bark particles had a positive effect on mechanical properties. The thickness swelling (TS) and water absorption (WA) values of MUF-bonded boards were lower than those of boards without MUF, and greater addition of PLB produced particleboards with better water resistance. Bark particle size was not as critical for TS and WA as for mechanical properties. The overall results suggested using a bark particle size of < 2 mm for further studies.

Keywords: bark, ethylene glycol, liquefaction, maritime pine, MUF resin, particleboard

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Introduction

Particleboard is a wood-based composite made from wood chips (particles) and some other lignocellulosic materials bonded with synthetic adhesives (e.g., urea--formaldehyde, melamine–urea–formaldehyde, and isocyanates). Particleboard is mainly used by the furniture industry because of its low weight, economical price, and existing processing technology [Barbu et al. 2014]. Competition for raw materials with other industry sectors (especially the bioenergy sector), sustainable forest management issues, and increasing transport costs have all led to an increase in the price of wood raw materials for the wood panel industry. It is reported that the global demand for particleboards for furniture and flooring has experienced a strong recovery since 2012, and an expansion in particleboard production is forecast to continue [BIS Shrapnel 2013; FAO 2017]. The demand for wood panels is constant in Europe, while it is expected to increase with an average annual growth rate of 1.05% from 2015 to 2030 in China, which is a large consumer of panels [Barbu et al. 2014; Yildirim 2019].

There has been a trend in using agricultural residues, industrial wastes like bark, and other lignocellulosic materials such as bamboo for producing particleboards [Blanchet et al. 2000; Papadopoulos et al. 2004; Yemele et al. 2008b; Madurwar et al. 2013]. Bark as sawmill residue has been studied as a raw material in wood-based panels as well as insulation boards for construction [Gao et al. 2011; Kain et al. 2012; Feng et al. 2013; Aydin et al. 2017]. Blanchet et al. [2000] prepared particleboards from black spruce bark using urea–formaldehyde resins. They reported that particleboards with a 1:1 ratio of wood to bark particles, 14% resin content in the surface layer, and 2-6 mm bark in the core layer had the best mechanical properties, including modulus of elasticity, modulus of rupture, and internal bond. Yemele et al. [2008a] investigated the effects of bark content and particle geometry on the physical and mechanical properties of particleboard made from black spruce and trembling aspen. Their results showed that increasing bark content led to decreased mechanical properties, and slightly increased linear expansion and thickness swelling. The internal bond strength of particleboard made with 50% bark content decreased with increasing particle size. As reported by some others, the use of bark in wood-based panels can cause a decrease in density, which accounts for the decrease in mechanical properties [Nemli and Çolakoğlu 2005; Yemele et al. 2008b; Kain et al. 2012].

Due to the existence of tannin in the bark chemical composition, it is possible to use bark for binderless boards [Gupta et al. 2011]. Thermo--mechanical refining of black spruce bark led to hydrolysis of hemicelluloses and plasticization of lignin for the preparation of fibres for self-bonding under a hot press [Velásquez et al. 2002; Gao et al. 2011]. Geng et al. [2006] also reported a pretreatment of bark using an alkaline solution followed by steam--pressurized refining to prepare fibres for fibreboards. After pretreatment and refining, the condensed polyphenolics, including condensed tannins and hydrolysed hemicelluloses, acted as binders for the fibreboards during hot-pressing.

Bark can also be used for other value-added applications such as tannin-based adhesives, preservatives and foams, and resins based on liquefaction or pyrolysis products [Feng et al. 2013]. A series of papers has been published on the liquefaction of bark and other lignocellulosic materials for application in wood adhesives [Pan 2011; Feng et al. 2013; Jiang et al. 2018]. Lee and Liu [2003] prepared particleboards bonded with phenol-formaldehyde resins based on liquefied bark. Specifically, they liquefied barks of Taiwan acacia and fir in phenols catalysed by sulphuric acid and hydrochloric acid. The results showed that the viscosities and thermal properties of resol resins were affected by the catalyst type and bark origin. Particleboard made with resol resin based on the liquefied Taiwan acacia bark catalysed by sulphuric acid had the best mechanical properties and the lowest thickness swelling among all particleboard types. Janiszewska [2018] liquefied bark in polyhydric alcohols and *p*-toluenesulphonic acid at 120°C for 2 hours, and then prepared three-layer particleboards using adhesives by combining commercial melamine-urea-formaldehyde (MUF) and liquefied bark (LB). It was also found that replacing MUF with LB led to a slight reduction in the formaldehyde content.

Nowadays, most commercial particleboards are manufactured with formaldehyde-based adhesives, which can result in environmental issues and health concerns [Abuarra et al. 2014; Solt et al. 2019]. Research on liquefied biomass-based adhesives has received remarkable attention, and although these alternatives have shown competitive properties, they have not entered commercial use. A better understanding of reaction pathways and optimization of the liquefaction processes are prerequisites for further development [Jiang et al. 2018]. To minimize the negative effect of bark on the mechanical strength of particleboards, and to utilize the chemical characteristics of bark, this paper presents a novel approach in using partially liquefied bark (PLB) for particleboard production. Partial liquefaction of biomass represents a new route in this area and has not been reported previously by other researchers. Bark was partially liquefied in ethylene glycol catalysed by sulphuric acid. The obtained PLB, which could serve as a furnish material with binding ability, was mixed with wood chips for making single-layer particleboards with the addition of MUF adhesives or without. It was hypothesized that PLB particles with chemical-activated surfaces can create chemical linking with wood chips, thus forming a homogeneous and interconnected furnish. The effects of bark particle sizes on the physical and mechanical properties of the particleboards were investigated.

Materials and methods

Materials

Bark of maritime pine (*Pinus pinaster* Ait.) was used for liquefaction. Bark was provided by BVB Substrates (De Lier, Netherlands). Bark was milled with a CSK 360/N1 Condux mill (Hanau, Germany) and screened to three different sizes: fine (< 1 mm), medium (1-2 mm), and coarse (> 2 mm). Chemicals used for liquefaction were ethylene glycol (Honeywell, Germany) as a solvent and 96% sulphuric acid (KEMIKA d.d., Croatia) as a catalyst. Fresh wood chips from spruce (*Picea* spp.) were collected from a local sawmill in Ljubljana, Slovenia. Melamine-urea-formaldehyde (MUF) resin H97 was provided by Melamin Kočevje d.d., Slovenia. Ammonium sulphate with 20% solid content was used as a hardener for MUF, and 1,4-dioxane purchased from VWR (Stockholm, Sweden) was used for the purification of liquefaction products.

Partial liquefaction

The bark fractions were dried in an oven at 105°C for 24 h before liquefaction. Bark liquefaction was carried out in a 1000 ml three-neck glass reactor, equipped with a stirrer and a condenser. Four bark particle size fractions, of all sizes (mixture), fine (< 1 mm), medium (1-2 mm), and coarse (> 2 mm), were used for the partial liquefaction. Four liquefactions were carried out, by loading fine, medium or coarse bark or a mixture thereof (an equal oven-dry weight of each size fraction) with solvents and catalysts into a reactor. For each liquefaction of the bark fractions, the ratio of solvent to bark was 3:1 (w/w) and the catalyst concentration was 3% based on the solvent mass. The reactor was immersed in an oil bath and kept at 180°C for 30 min with continuous stirring. The selected liquefaction method allowed only partial liquefaction (activation of bark surface) of the bark materials. As described in previous work [Jiang et al. 2020], the partially liquefied bark (PLB) product is a wet material with a solid content of 41% (oven-dried mass of solids divided by total PLB mass) containing unreacted solvent and liquefaction derivatives. The four categories of PLB obtained from partial liquefaction of different bark fractions were marked respectively as PLB-A, PLB-F, PLB-M, and PLB-C.

Particleboard manufacture

Wood chips were hammer-milled and then dried (16 hours at 70°C) to a moisture content of less than 4%. A single-layer particleboard was used as a control board and was made from wood chips and 10% MUF resin. Twelve single-layer experimental particleboards were made from PLB and wood chips with or without adding MUF resin. All boards were prepared with a target thickness of 8 mm. Wood chips together with each PLB category and MUF resin were blended and hand-formed into a 500 × 500 mm^2 particle mat. The mats were hot-

-pressed at 180°C under a maximum pressure of 3.0 N·mm^{-2}, which was afterward adjusted to enable vapor degassing (the degassing stage lasted 150 s, and pressure of 1 N·mm^{-2}), followed by a final mat densification and curing stage (180 seconds). A pressing diagram can be seen from the previous paper [Jiang et al. 2020]. Due to the high moisture in the mats, the total pressing time was 7 minutes for each board (pressing factor 52.5 s·mm^{-1}). Table 1 shows the manufacturing parameters of the particleboards, which were divided into four groups. Boards in group A were made by mixing PLB from partial liquefaction of the different bark particle size categories and wood chips at a weight ratio of 0.25 without using MUF resin for additional bonding. Boards in group B were made with PLB and wood chips at the same ratio of 0.25, and 10 wt% of MUF based on the weight of oven-dried wood chips and wet PLB. Boards in group C were made with a PLB-to-wood weight ratio of 0.1 and an additional 10 wt% of MUF.

Table 1. Experimental design

Group	Particleboard	Bark particle size*	PLB-to-wood weight ratio	MUF (wt%)
Control	–	–	–	10
A	25PLB-A	All sizes (A)	0.25 (1:4)	0
	25PLB-F	Fine (F)	0.25	0
	25PLB-M	Medium (M)	0.25	0
	25PLB-C	Coarse (C)	0.25	0
B	25PLB-A-MUF	A	0.25	10
	25PLB-F-MUF	F	0.25	10
	25PLB-M-MUF	M	0.25	10
	25PLB-C-MUF	C	0.25	10
C	10PLB-A-MUF	A	0.10 (1:10)	10
	10PLB-F-MUF	F	0.10	10
	10PLB-M-MUF	M	0.10	10
	10PLB-C-MUF	C	0.10	10

*Bark size denotes the size of bark particles used for liquefaction.

Particleboard testing

All particleboards were tested for physical and mechanical properties according to the European standards concerning board thickness (EN 324-1:1993, 6 samples), density (EN 323:1993, 6 samples), moisture content (EN 322:1993, 4 samples), modulus of elasticity (MOE) and rupture (MOR) in bending (EN 310:1993, 5 samples), internal bond (IB) strength (EN 319:1993, 8 samples), and thickness swelling (TS) and water absorption (WA) after water immersion

(EN 317:1993, 8 samples). A tensile test parallel to the surface of the board was also applied with six samples of 25 × 210 mm^2 (width × length) from each board. Mechanical tests were performed using a Zwick/Roell Z005 universal testing machine (Zwick/Roell GmbH, Ulm, Germany).

ANOVA analysis

SPSS version 25.0 (IBM Corp., Armonk, NY, USA) was used for statistical analysis of the test results. One-way ANOVA was performed on the physical and mechanical results for the analysis of variance at a 95% confidence interval ($p < 0.05$). The statistical differences between mean values within the different groups were assessed using the Tukey test.

Fourier Transform Infrared (FTIR) spectroscopy

The chemical structure of raw bark, crude PLB, oven-dried PLB (O-PLB), and oven-dried purified PLB (OP-PLB) was analysed with a Fourier Transform Infrared spectrometer (Alpha FTIR spectrometer, Bruker, Karlsruhe, Germany) with a versatile high-throughput ZnSe ATR crystal. Purified PLB was prepared by first dissolving crude PLB in a solvent mixture of 1,4-dioxane and water at a mass ratio of 4:1, and then centrifuging the solutions at 1000 rpm for 10 min to remove the residual solvents and intermediate chemicals, as described previously [Jiang et al. 2020]. The FTIR analysis was performed in a wavelength range from 4000 to 800 cm^{-1} at room temperature, accumulating 64 scans with a resolution of 4 cm^{-1}.

Results and discussion

The physical properties of all particleboards are given in Table 2. Using a PLB-to-wood ratio of 0.10 with the presence of MUF adhesives (group C) did not cause large differences in the density and thickness, but led to lower moisture content in the boards compared with the control. However, when using a higher loading of PLB, at a PLB-to-wood ratio of 0.25 with or without MUF, the boards in groups A and B had much lower density, larger thickness, and higher moisture content than both group C and the control board.

The particleboards in group A made with a PLB-to-wood ratio of 0.25 with no adhesives were found to have lower average densities than those in group B and C (Table 2). The reason for such results may be related to a larger springback effect due to the lack of adhesive bonding between the wood chips and partially liquefied bark in the boards of group A compared with groups B and C. Hence, relaxation occurred, leading to a higher thickness and lower density. The addition of MUF adhesives in the particleboards (group B and C) contributed to a more stable bond between wood chips and partially liquefied bark, thus resulting in lower springback and hence lower thickness and higher density.

Table 2. Physical properties of particleboards made with partially liquefied bark of different sizes (mean values ± standard deviations)

Group	Particleboard	Density (kg/m^3)	Thickness (mm)	Moisture content (%)
Control	–	754.58 ±17.32	7.63 ±0.09	8.66 ±0.07
A	25PLB-A	553.70 ±17.52	8.32 ±0.27	14.30 ±0.98
	25PLB-F	603.29 ±10.31	8.47 ±0.12	13.34 ±0.53
	25PLB-M	602.62 ±53.51	8.13 ±0.15	12.99 ±0.90
	25PLB-C	436.52 ±45.19	8.52 ±0.12	13.89 ±0.68
B	25PLB-A-MUF	685.89 ±61.78	7.79 ±0.11	15.79 ±3.47
	25PLB-F-MUF	616.96 ±97.92	7.85 ±0.07	12.27 ±0.71
	25PLB-M-MUF	633.16 ±42.18	7.88 ±0.06	11.77 ±0.42
	25PLB-C-MUF	628.92 ±36.17	7.89 ±0.09	12.65 ±0.83
C	10PLB-A-MUF	728.03 ±19.38	7.60 ±0.15	6.46 ±0.33
	10PLB-F-MUF	759.12 ±34.83	7.64 ±0.12	5.54 ±0.26
	10PLB-M-MUF	717.32 ±61.13	7.64 ±0.06	7.58 ±0.36
	10PLB-C-MUF	751.62 ±65.10	7.77 ±0.06	6.80 ±0.30

Higher loading of PLB in groups A and B compared with group C led to a decrease in the density, because of the high amount of unreacted solvents and aqueous intermediate fragments from the liquefaction, such as water, furfurals, acids, and aldehydes, in the PLB [Yamada and Ono 2001; Zou et al. 2009; Dussan et al. 2015]. These liquids evaporated during the pressing process. An analysis of the chemical composition of bark and PLB products based on FTIR is shown in Fig. 1. Ethylene glycol exhibited typical absorption bands in the FTIR spectrum as follows: strong OH stretching at 3300 cm^{-1}; symmetric and asymmetric CH stretching vibrations at 2935 and 2872 cm^{-1}; weak bands corresponding to C–O vibrations between 1456 and 1330 cm^{-1}; strong bands at 1078, 1037, and 874 cm^{-1} corresponding to CH_2 groups [Chirea et al. 2011]. All the above absorption bands were observed in crude PLB but not in O-PLB or OP-PLB, which proved the presence of unreacted ethylene glycol in the crude PLB that can be removed by oven-drying or purification. The PLB residues, presented as O-PLB and OP-PLB, exhibited different chemical structures than raw bark. A broad band corresponding to C–O linkage between 1266 and 1030 cm^{-1} was found in O-PLB and OP-PLB but not in raw bark, indicating the formation of new substances, as described in previous work [Jiang et al. 2020]. A band shift from 1743 cm^{-1} in the raw bark to 1720 cm^{-1} in O-PLB and

OP-PLB was observed and was attributed to the hydrolysis of hemicellulose and the solvolysis of cellulose to form levulinic acid esters [Yamada and Ono 2001; Kobayashi et al. 2004]. The changes in the chemical structure of bark before and after partial liquefaction confirmed the initiation of the liquefaction process. The unreacted solvent and intermediate chemicals may prolong the liquefaction and promote further recondensation reactions during the pressing of particleboards, since the required liquefaction temperature was the same as the pressing temperature of 180°C.

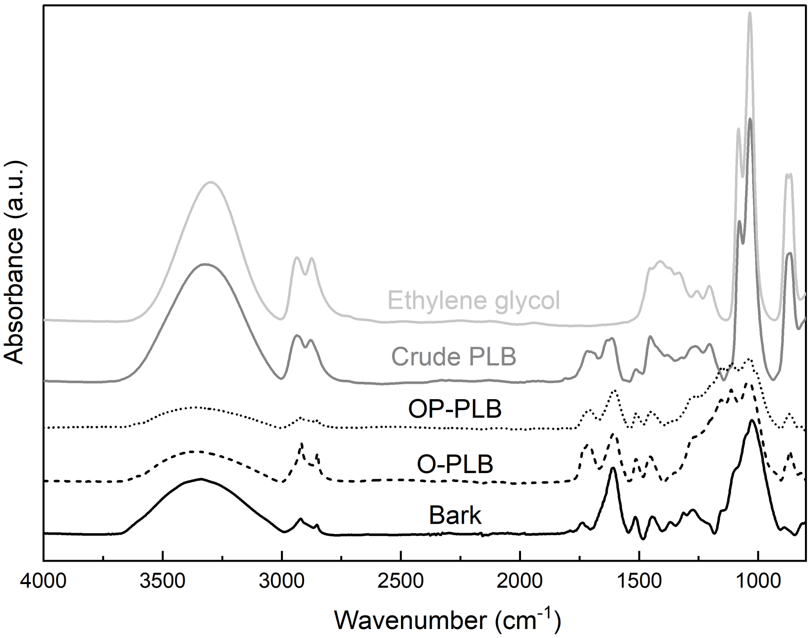

Fig. 1. Fourier transform infrared (FTIR) spectra of raw bark, crude partially liquefied bark (PLB), oven-dried PLB (O-PLB), oven-dried purified PLB (OP-PLB), and ethylene glycol

As shown in Table 2, the bark particle size used at liquefaction seemed to affect the physical properties only in the case of boards made without MUF (group A), where the particleboards made with PLB from mixed-size or coarse bark had lower densities and higher moisture content than those with PLB from fine bark or medium bark. Due to their higher density values, particleboards of groups B and C had higher MOR, MOE, IB, and tensile strength than those of group A, but only group C attained mechanical strength comparable to that of the control board (Table 3). This was expected, as MUF provided sufficient bonding to the wood chips, but a limited portion of PLB should be used. The statistical analysis (noted in Table 3) showed that the bark particle size used at liquefaction had a significant effect ($p < 0.05$) on the mechanical properties of particleboards within each group. An exception was noted for the MOR of particleboards made with a PLB-to-wood chips ratio of 0.25 and 10 wt% MUF (group B), where no significant differences related to bark particle size could be observed.

Table 3. Mechanical properties of particleboards made with partially liquefied bark of different sizes (mean values ± standard deviations)

Group	Panel type	MOR (MPa)	MOE (MPa)	IB (MPa)	Tensile strength (MPa)
Control		20.97 ±2.85	2643 ±223	1.02 ±0.25	10.29 ±0.85
A	25PLB-A	2.37 ±0.88a	737 ±297a	0.06 ±0.02a	0.98 ±0.30ab
	25PLB-F	2.57 ±1.22a	826 ±366a	0.08 ±0.02b	1.26 ±0.06a
	25PLB-M	2.72 ±0.74a	813 ±258a	0.10 ±0.01b	0.75 ±0.16b
	25PLB-C	0.85 ±0.10b	152 ±26b	0.01 ±0.01c	0.30 ±0.07c
	Sig.	0.003	0.001	0.000	0.000
B	25PLB-A-MUF	13.84 ±5.03a	2308 ±771a	0.71 ±0.33a	4.15 ±1.79ab
	25PLB-F-MUF	13.33 ±2.45a	2214 ±255a	0.46 ±0.18bc	6.74 ±2.30ac
	25PLB-M-MUF	8.95 ±3.20a	1422 ±432b	0.49 ±0.12bc	3.12 ±2.05b
	25PLB-C-MUF	11.25 ±1.86a	1736 ±254ab	0.43 ±0.10c	7.72 ±0.82c
	Sig.	0.077	0.014	0.045	0.001
C	10PLB-A-MUF	30.87 ±3.94a	3837 ±474a	2.07 ±0.33a	7.37 ±1.07a
	10PLB-F-MUF	28.80 ±4.60ab	7193 ±690b	0.87 ±0.13b	11.17 ±1.56b
	10PLB-M-MUF	23.95 ±3.80b	3295 ±484a	0.79 ±0.15b	7.48 ±2.34ac
	10PLB-C-MUF	16.81 ±1.85c	2316 ±200c	0.83 ±0.17b	4.78 ±1.54c
	Sig.	0.000	0.000	0.000	0.000

Note: For each group, values followed by a different letter within a column are statistically different at $p < 0.05$ (ANOVA and Tukey HSD test).

In group A, boards with PLB from coarse bark (25PLB-C) had significantly lower MOR, MOE, IB, and tensile strength. Boards in the other bark categories (25PLB-A, 25PLB-F, and 25PLB-M) showed no statistically significant differences in bending properties (MOR, MOE). PLB made from coarse bark gave almost zero IB strength in the board 25PLB-C, while the board 25PLB-A with mixed bark had significantly lower IB than boards with medium and fine bark (25PLB-M and 25PLB-F). PLB from fine and mixed bark (boards 25PLB-F and 25PLB-A) gave higher tensile strength than the material with the medium fraction (25PLB-M).

As mentioned before, MOR was not found to differ with the bark particle size fraction for particleboards in group B made with a PLB-to-wood ratio of 0.25 and 10 wt% MUF. The board with the medium bark fraction (25PLB-M-

-MUF) had lower bending properties, but only MOE was significantly different from the other boards. For group B, mixed bark (25PLB-A-MUF) gave the best IB results, with a significantly higher mean value than the other individual bark categories. Boards made with PLB from medium bark (25PLB-M-MUF) exhibited inferior tensile strength compared with the mixed (25PLB-A-MUF) and fine (25PLB-F-MUF) categories, while the coarse (25PLB-C-MUF) fraction had significantly higher tensile strength.

When the PLB-to-wood chip ratio was 0.1, in group C, the board with PLB from mixed bark (10PLB-A-MUF) had better mechanical performance overall, especially in terms of MOR and IB. Boards based on PLB from fine and medium fraction bark (10PLB-F-MUF and 10PLB-M-MUF) also performed comparably to 10PLB-A-MUF as well as the control board. The board with PLB made from bark with coarse particle size (10PLB-C-MUF) had lower MOR, MOE, and tensile strength than the other categories, and the differences were statistically significant.

Thickness swelling (TS) and water absorption (WA) of the particleboards were tested after 2 and 24 h immersion in water at a temperature of 20°C. The TS after 2 and 24 hours of the particleboards made without MUF (group A) was above 20%, while the particleboards made with MUF (groups B and C) had much better water resistance, with TS values below 20% (Fig. 2). The higher swelling of group A may be related to lower density and lower bond strength (IB and tensile strength). Fig. 2 also shows that groups B and C exhibited lower values of TS and WA than the control board, while boards in group A had slightly higher values of thickness swelling after 2 and 24 h, and comparable water absorption, relative to the control. This indicates that PLB may act as an excellent water-resistant reagent in particleboards. A reasonable explanation was given in a previous publication, namely that PLB had an activated surface and a less porous structure than wood chips, which can enhance the compatibility of bark with wood chips and result in a reduction of water penetration [Jiang et al. 2020].

As shown in Fig. 2, bark particle size had a significant influence ($p < 0.05$) on the TS and WA of group A, 24 h TS and 24 h WA of group B, and WA of group C. For all cases, TS values were comparable between 2 h and 24 h. In all groups, TS changed little with varying bark particle size. Only in group A, 25PLB-C exhibited relatively higher TS than the other boards. As expected, particleboards made without using MUF (group A) absorbed more water than the MUF-bonded boards (groups B and C). It is surprising to observe, when comparing the boards made from PLB from the same bark particle size in groups B and C, that boards made with a PLB-to-wood ratio of 0.25 had lower TS and WA than those with a PLB-to-wood ratio of 0.1. This indicates that PLB also acted as a water repellent component. For groups A and B, the TS and WA of boards made from fine and medium bark had no statistically significant

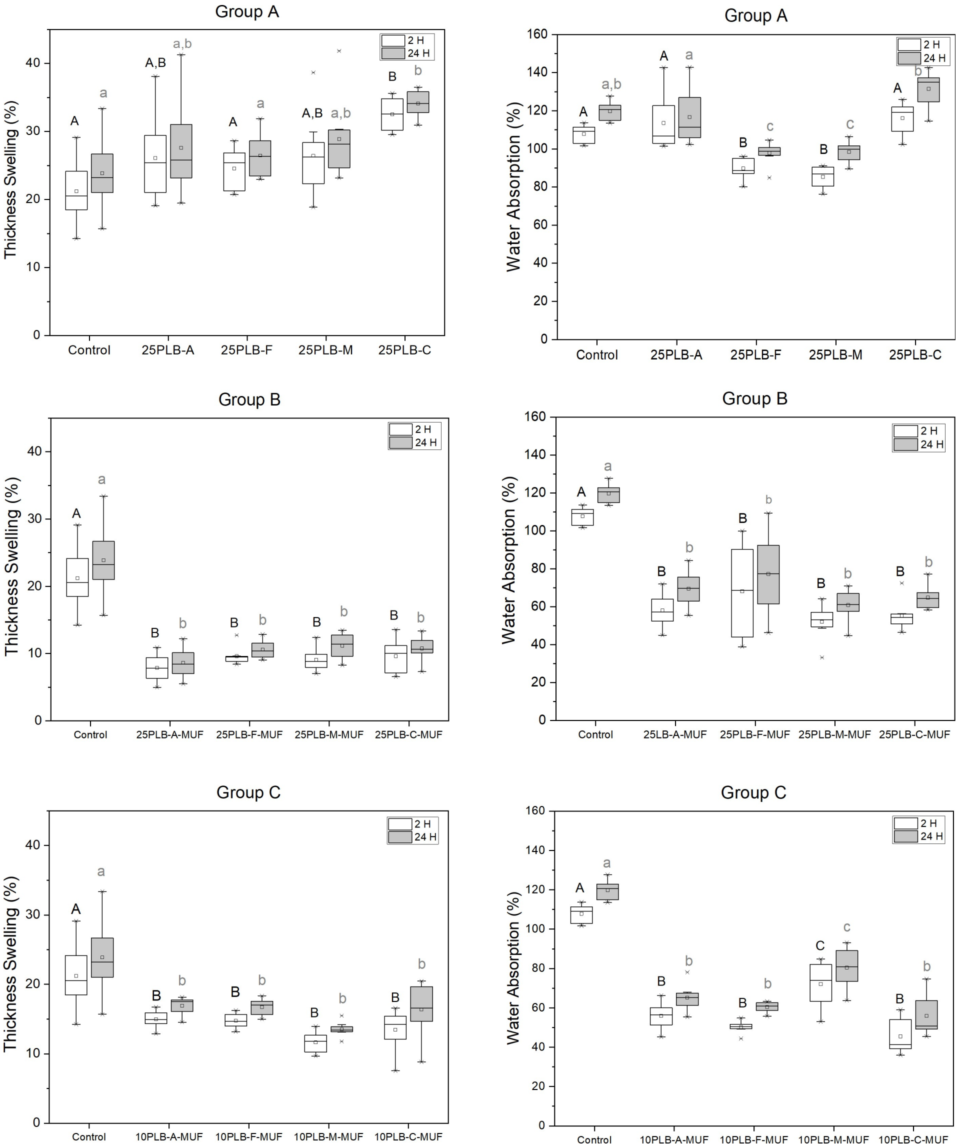

Fig. 2. Thickness swelling and water absorption (2 and 24 h) of particleboards made with partially liquefied bark of different sizes. Error bars represent standard deviations. Values labelled with different letters are statistically different at $p < 0.05$ (ANOVA and Tukey test)

differences, which suggests the potential use of a mixture of fine and medium bark for further studies. For group C, no statistical difference was found between the boards, but they all had significantly lower values than the control. Values of WA for coarse bark board (10PLB-C-MUF) in group C were significantly different from those for 10PLB-M-MUF, but not from those for 10PLB-A-MUF and 10PLB-F-MUF.

MUF adhesives were expected to provide sufficient bonding to the wood chips. Based on previous studies on the liquefaction of different types of biomass, the liquefied products exhibited no polymerization properties or cohesion properties [Budija et al. 2009; Ugovšek and Sernek 2013]. However, an inspiring outcome was the possible production of particleboard from PLB and wood chips without any commercial resin. As explained above, this was possible because PLB containing unreacted solvent and intermediate chemicals formed chemical linking with wood chips. Nevertheless, group A exhibited inferior properties overall compared with groups B and C. The overall results suggest that bark particle size has a significant effect on the physical and mechanical properties of particleboards. The coarse fraction does not promote bonding, possibly due to a lesser degree of partial liquefaction, but interestingly the bark mixture, despite the presence of coarse bark particles, seems to be quite effective (see Table 3 and Fig. 2). A reduction in particle size helps to increase the accessibility of biomass and enables better heat transfer during liquefaction [Akhtar and Amin 2011], hence better liquefaction efficiency. Thus, a bark particle size of less than 2 mm should be considered for further studies (in our case, sizes F and M). It is known that particle dimensions strongly affect the properties of particleboards [Mundy and Bonfield 1998]. In this respect, it is evident that an optimized mixture of wood chips and PLB with appropriate size must be established to provide a more uniform and compact board structure, which is required for adequate IB strength and lower thickness swelling [Dai et al. 2008; Nazerian et al. 2011]. Interactions with MUF adhesives should also be taken into account, since for example, a higher share of finer particles would tend to absorb an unpredictable amount of resin, leaving less resin available for particle bonding [Sackey et al. 2008; Wan Abd Rahman et al. 2019].

Conclusions

The present study has investigated the effect of the particle size of bark undergoing partial liquefaction, and later used for particleboard manufacture, on the physical, mechanical, and water-related properties of the particleboards. The use of PLB to manufacture particleboards without MUF resin was possible, but the density and mechanical strength of the boards were inferior to those of particleboards made with the addition of MUF. Bark particle size was shown to be an influencing factor on the MOR, MOE, IB, and tensile strength of particleboards made with the same PLB content. However, it was not possible to draw a definite conclusion on which specific bark particle size had the largest influence on the mechanical properties. PLB prepared from a mixture of bark particles with a size of less than 2 mm (i.e. avoiding coarse particles) seemed to ensure good mechanical behaviour of the particleboards. The results for TS and

WA implied that the addition of MUF is necessary, and that bark particle size is not as critical here as in the case of mechanical properties.

References

Abuarra A., Hashim R., Bauk S., Kandaiya S., Tousi E.T. [2014]: Fabrication and characterization of gum Arabic bonded *Rhizophora* spp. particleboards. Materials & Design 60: 108-115. DOI: 10.1016/j.matdes.2014.03.032

Akhtar J., Amin N.A.S. [2011]: A review on process conditions for optimum bio-oil yield in hydrothermal liquefaction of biomass. Renewable and Sustainable Energy Reviews 15 [3]: 1615-1624. DOI: 10.1016/j.rser.2010.11.054

Aydin I., Demirkir C., Colak S., Colakoglu G. [2017]: Utilization of bark flours as additive in plywood manufacturing. European Journal of Wood and Wood Products 75: 63-69. DOI: 10.1007/s00107-016-1096-0

Barbu M.C., Reh R., Irle M. [2014]: Wood-based composites. In: Aguilera A., Davim J.P. (eds.), Research Developments in Wood Engineering and Technology. IGI Global. Hershey

Blanchet P., Cloutier A., Riedl B. [2000]: Particleboard made from hammer milled black spruce bark residues. Wood Science and Technology 34 [1]: 11-19. DOI: 10.1007/s002260050003

BIS Shrapnel [2013]: Particleboard and medium density fibreboard in the Pacific Rim and Europe 2009-2013. BIS Shrapnel Business Research and Forecasting. Sydney. [accessed: 08.06.2021]. Available from: https://www.compositepanel.org/userfiles/filemanager/52950202ad814

Budija F., Tavzes Č., Zupančič-Kralj L., Petrič, M. [2009]: Self-crosslinking and film formation ability of liquefied black poplar. Bioresource technology 100 [13]: 3316-3323. DOI: 10.1016/j.biortech.2009.02.004

Chirea M., Freitas A., Vasile B.S., Ghitulica C., Pereira C.M., Silva F. [2011]: Gold nanowire networks: synthesis, characterization, and catalytic activity. Langmuir 27 [7]: 3906-3913. DOI: 10.1021/la104092b

Dai C., Yu C., Jin J. [2008]: Theoretical modeling of bonding characteristics and performance of wood composites. Part IV. Internal bond strength. Wood and Fiber Science 40 [2]: 146-160

Dussan K., Girisuta B., Lopes M., Leahy J.J., Hayes M.H. [2015]: Conversion of hemicellulose sugars catalyzed by formic acid: kinetics of the dehydration of D-xylose, L-arabinose, and D-glucose. ChemSusChem 8 [8]: 1411-1428. DOI: 10.1002/cssc.201403328

FAO [2017]: 2016 Global forest products facts and figures. [accessed: 30.10.2019]. Available from: http://www.fao.org/3/i7034en/i7034en.pdf

Feng S., Cheng S., Yuan Z., Leitch M., Xu C.C. [2013]: Valorization of bark for chemicals and materials: A review. Renewable and Sustainable Energy Reviews 26: 560-578. DOI: 10.1016/j.rser.2013.06.024

Gao Z., Wang X., Wan H., Brunette G. [2011]: Binderless panels made with black spruce bark. BioResources 6 [4]: 3960-3972

Geng X., Zhang S.Y., Deng J. [2006]: Alkaline treatment of black spruce bark for the manufacture of binderless fiberboard. Journal of Wood Chemistry and Technology 26 [4]: 313-324. DOI: 10.1080/02773810601076857

Gupta G., Yan N., Feng M.W. [2011]: Effects of pressing temperature and particle size on bark board properties made from beetle-infested lodgepole pine (*Pinus contorta*) barks. Forest Products Journal 61 [6]: 478-488. DOI: 10.13073/0015-7473-61.6.478

Janiszewska D. [2018]: Bark liquefaction for use in three-layer particleboard bonding. Drewno 61 [202]: 119-127

Jiang W., Adamopoulos S., Hosseinpourpia R., Žigon J., Petrič M., Šernek M., Medved S. [2020]: Utilization of Partially Liquefied Bark for Production of Particleboards. Applied Sciences 10 [15]: 5253. DOI: 10.3390/app10155253

Jiang W., Kumar A., Adamopoulos S. [2018]: Liquefaction of lignocellulosic materials and its applications in wood adhesives – A review. Industrial Crops and Products 124: 325--342. DOI: 10.1016/j.indcrop.2018.07.053

Kain G., Barbu M.C., Teischinger A., Musso M., Petutschnigg A. [2012]: Substantial bark use as insulation material. Forest Products Journal 62 [6]: 480-487. DOI: 10,13073/FPJ-D-12-00052.1

Kobayashi M., Asano T., Kajiyama M., Tomita B. [2004]: Analysis on residue formation during wood liquefaction with polyhydric alcohol. Journal of Wood Science 50: 407-414. DOI: 10.1007/s10086-003-0596-9

Lee W.J., Liu C.T. [2003]: Preparation of liquefied bark-based resol resin and its application to particle board. Journal of Applied Polymer Science 87 [11]: 1837-1841. DOI: 10.1002/app.11917

Madurwar M.V., Ralegaonkar R.V., Mandavgane S.A. [2013]: Application of agro-waste for sustainable construction materials: A review. Construction and Building Materials 38: 872-878. DOI: 10.1016/j.conbuildmat.2012.09.011

Mundy J.S., Bonfield P.W. [1998]: Predicting the short-term properties of particleboard using composite theory. Wood Science and Technology 32 [3]:237-245

Nazerian M., Ghalehno M.D., Shojaiishad M., Sharifpoor H., Taftiyan M.H. [2011]: Properties of three-layer particleboard made from wood of Athel (*Tamarix aphylla*) and pruning particles of almond (*Amygdalus communis*) and pistachio (*Pistacia vera*). Journal of Basic and Applied Scientific Research 1 [8]:837-843

Nemli G., Çolakoğlu G. [2005]: Effects of mimosa bark usage on some properties of particleboard. Turkish Journal of Agriculture and Forestry 29: 227-230

Pan H. [2011]: Synthesis of polymers from organic solvent liquefied biomass: A review. Renewable and Sustainable Energy Reviews 15 [7]: 3454-3463. DOI: 10.1016/j.rser.2011.05.002

Papadopoulos A.N., Hill C.A.S., Gkaraveli A., Ntalos G.A., Karastergiou S.P. [2004]: Bamboo chips (*Bambusa vulgaris*) as an alternative lignocellulosic raw material for particleboard manufacture. Holz als Roh-und Werkstoff 62: 36-39. DOI: 10.1007/s00107-003-0447-9

Sackey E., Semple K., Oh S-W., Smith G. [2008]: Improving core bond strength of particleboard through particle size redistribution. Wood and Fiber Science 40 [2]: 214--224

Solt P., Konnerth J., Gindl-Altmutter W., Kantner W., Moser J., Mitter R., van Herwijnen H.W. [2019]: Technological performance of formaldehyde-free adhesive alternatives for particleboard industry. International Journal of Adhesion and Adhesives 94: 99-131. DOI: 10.1016/j.ijadhadh.2019.04.007

Ugovšek A., Sernek M. [2013]: Characterisation of the curing of liquefied wood by rheometry, DEA and DSC. Wood Science and Technology 47: 1099–1111. DOI: 10.1007/s00226-013-0565-4

Velásquez J.A., Ferrando F., Salvadó J. [2002]: Binderless fiberboard from steam exploded Miscanthus sinensis: the effect of a grinding process. Holz als Roh-und Werkstoff 60: 297-302

Wan Abd Rahman W., Md Yatim A., Mat Zlan A., Kasim J., Mohd Yunus N. [2019]: Effects of the resin content and particle size on the properties of particleboard made of Neolamarckia and Leucaena particles. BioResources 14 [3]: 6079-6087

Yamada T., Ono, H. [2001]: Characterization of the products resulting from ethylene glycol liquefaction of cellulose. Journal of Wood Science 47 [6]: 458-464. DOI: 10.1007/BF00767898

Yemele M.C., Blanchet P., Cloutier A., Koubaa A. [2008a]: Effects of bark content and particle geometry on the physical and mechanical properties of particleboard made from black spruce and trembling aspen bark. Forest Products Journal 58 [11]: 48-56

Yemele M.C., Cloutier A., Diouf P.N., Koubaa A., Blanchet P., Stefanovic T. [2008b]: Physical and mechanical properties of particleboard made from extracted black spruce and trembling aspen bark. Forest Product Journal 58 [10]: 38-46

Yildirim H.T. [2019]: Raw material demand-supply and policy recommendations of Turkish wood-based panel industry. In: Timber buildings and constructions. IntechOpen. DOI: 10.5772/intechopen.82627

Zou X., Qin T., Huang L., Zhang X., Yang Z., Wang Y. [2009]: Mechanisms and main regularities of biomass liquefaction with alcoholic solvents. Energy & Fuels 23 [10]: 5213-5218. DOI: 10.1021/ef900590b

List of standards

EN 310:1993 Wood-based panels – Determination of modulus of elasticity in bending and of bending strength

EN 317:1993 Particleboards and fibreboards – Determination of swelling in thickness after immersion in water

EN 319:1993 Particleboards and fibreboards – Determination of tensile strength perpendicular to the plane of the board

EN 322:1993 Wood-based panels – Determination of moisture content

EN 323:1993 Wood-based panels – Determination of density

EN 324-1:1993 Wood-based panels – Determination of thickness, width and length

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