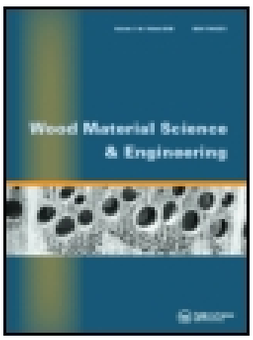

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ORIGINAL ARTICLE

Effects of low and high molecular weight of phenol-formaldehyde (PF) on the properties of strand boards from kiri wood (*Paulownia tomentosa*)

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ABSTRACT

Kiri (*Paulownia tomentosa*) wood is a promising material for lightweight strand boards (SBs); however, kiri SBs have displayed a limited dimensional stability. The aim of this study was to evaluate the possibility of using low molecular weight (LMW) phenol-formaldehyde (PF) resin not only as an adhesive but also as a treatment (impregnating) agent to manufacture SBs. SBs from kiri wood were manufactured with densities of 400 kg m^{-3} and 500 kg m^{-3}. PF resin with low and high molecular weight as well as its 50-50% mixture was studied at two adhesive formulation contents of 10% and 20% related to the strand mass. At 400 kg m^{-3} density, internal bond strength (IB), screw withdrawal resistance (SWR) and thickness swelling (TS) of SBs containing LMW PF were significantly higher than those of HMW PF at 10% adhesive content and the differences slightly decreased as the adhesive content increased to 20%. At 500 kg m^{-3} density, IB, TS and SWR of SBs were considerably enhanced by LMW PF at both adhesive contents. We concluded that using LMW PF may cause higher strength and dimensional stability, at least, when the strand material and the SBs exhibits very low density, which is highly compressed during SBs manufacturing.

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Introduction

Oriented strand boards (OSB) are mainly utilized for construction purposes such as walls, roof sheathings, I-beams, or single-layer flooring (USDA 1999). Strength properties such as MOR and MOE of OSB, however, can be gradually reduced when the products are exposed to a moist environment during service (Wu and Suchsland 2007). This is mostly attributed to dimensional instability, as swelling and shrinking of the strands may reduce adhesion and induce cracking of the panels. Therefore, there is a need to improve the dimensional stability of OSB under moist conditions, especially in exterior applications. The application of adhesives, which are stable towards hydrolysis, is a prerequisite of high dimensional stability and strength maintenance of OSB and other wood-based panels under outdoor conditions. In Europe, OSB is therefore produced with isocyanate adhesives such as polymeric diphenylmethane diisocyanate (pMDI), while the OSB industry in other geographic regions also applies phenol-formaldehyde (PF) resins. Given such a hydrolysis stable adhesive, there are two main factors determining the thickness swelling of OSB: swelling of wood strands itself and the spring-back effect after the stress is released and the board gets in contact with water (Menezzi and Tomaselli 2006). In addition, the water uptake of solid wood can be reduced by using various wood modification approaches, where the wood cell wall is altered in different ways. These approaches involve thermal modification, where the number of OH groups is reduced or chemical wood modification such as acetylation, where the OH groups are substituted with more bulky acetate groups (Hill 2006). Thermal modification is acknowledged as an effective technique to enhance the dimensional stability of particleboards (Boonstra *et al.* 2006) and OSB (Paul *et al.* 2007), where modification at a temperature above 190 °C and 220°C, however, led to a slight reduction of MOR, MOE and IB in OSB (Menezzi *et al.* 2009). OSB from Scots pine (*Pinus sylvestris* L.) in a thermal post-treatment process, showed that mechanical strength (MOR, IB) decreased significantly, while MOE did not change significantly (Direske *et al.* 2018). Pipíška *et al.* (2020) reported, that the thermal modification of Norway spruce (*Picea abies L. Karst*) strands at 180°C prior to panel production cause significant mechanical strength loss.

Another promising technique for solid wood treatment involves the use of synthetic resins (PF, melamine-formaldehyde, DMDHEU) (e.g. Verma *et al.* 2009). The impregnation of wood with PF and other resins has been previously studied, largely focusing on dimensional stability (Stamm and Seborg 1939, 1941, Stamm 1955, Stamm and Baechler 1960) and decay resistance (Stamm and Seborg 1939, Stamm and Baechler 1960). Stamm and Seborg (1939) tested different resin forming materials such as PF, urea-formaldehyde (UF), and methacrylate intermediates, of which they selected PF-based resins as being the most promising for treating veneers for plywood manufacture. Later, several authors reported that the molecular weight of the resin is crucial for the penetration into the wood cell wall (Smith *et al.* 1985, Imamura *et al.* 1998, Furuno *et al.* 2004). High molecular

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weight (HMW) resins penetrate the lumens and may prevent capillary water uptake by physically blocking the flow paths of water into the wood. Low molecular weight (LMW) resins may additionally penetrate the cell wall matrix of wood. They concluded that resin located in the cell wall would alter the wood properties to a greater extent than on located in the cell lumen. Furuno *et al.* (2004) showed that resins with an average molecular weight ranging from 290 to 470 g mol^{-1} penetrated the cell walls of Japanese cedar wood (*Cryptomeria japonica* D. Don) and improved the dimensional stability. A higher proportion of resin penetration increased cell wall bulking and respectively reduced water absorption.

PF resin has been used to treat wood elements for exterior application to enhance the dimensional stability of plywood (Stamm and Seborg 1939), laminated veneer lumber (LVL) (Bicke and Militz 2014, Hong *et al.* 2018), and particleboards (Kajita and Imamura 1991). Haygreen and Gertjejansen (1971) partially replaced HMW PF "bonding" resin with LMW PF "impregnating" resin by mixing the two resin types and applied it in a laboratory blender to produce particleboards. Addition of impregnating resin significantly enhanced the dimensional stability compared to panels containing bonding resin only, while mechanical properties were found to be similar for panels containing both resin types and those containing only bonding resin. In a similar approach, particles were treated by dipping into aqueous solutions of LMW PF resin with a number-average molecular weight of 390 g mol^{-1} and subsequently dried at 60 °C (Kajita and Imamura 1991). Then, the particles were sprayed with an HMW PF adhesive of a number-average molecular weight of 960 g mol^{-1}, or directly sprayed with a mixture of LMW PF resin and the adhesive PF at one single step. The treated particleboards displayed higher dimensional stability and decay resistance as well as internal bond strength (IB) than conventional panels bonded only with PF adhesive. Stephens and Kutscha (1986) investigated aspen particleboards bonded with PF resins of different molecular weight and found that the best panel performance for IB and TS was achieved by combining both HML and LMW fractions.

It is desirable to develop lightweight wood-based panels based on renewable and sustainable resources such as light and fast-growing wood species (Barbu 2015). The kiri tree (*Paulownia tomentosa*) has a very high growth rate and short rotation time (Icka *et al.* 2016). Previous studies revealed that kiri wood is a promising raw material for the wood-based panel industry. Particleboards of kiri wood with a density of 350–500 kg m^{-3} showed higher bending strength than those of conventional industry particles (Nelis *et al.* 2018). Van *et al.* (2019) produced lightweight strand boards (SBs) from kiri with target densities of 300 and 400 kg m^{-3} which have shown higher MOR, MOE and IB than those of SBs made of pine (*Pinus sylvestris*). Nevertheless, kiri SBs showed a strong thickness swelling, which was attributed to a high compaction ratio and a resulting higher spring-back effect. This low dimensional stability might limit the application of kiri wood panels for outside application. The working hypothesis was that using LMW PF resins dimensionally stabilize SBs compared to using PF resins of HMW and, thus, make them better suitable for outdoor application. To overcome this drawback, this work intends to improve the dimensional stability of SBs and assesses the influence of the molecular weight of PF resin on the water-related properties (TS, water uptake) of SBs from kiri wood. In addition, it assesses the effect of LMW and HMW PF resins on IB, bending properties (MOR, MOE), and SWR.

Materials and methods

Strand production

Kiri (*Paulownia tomentosa*) trees were harvested near Bonn, Germany. Logs with diameters from 12 to 20 cm were cut into 200 cm long sections and debarked by hand before stranding. The strands were produced by a knife ring flaker PZUL 8–300 (Pallmann Maschinenfabrik, Zweibrücken, Germany). The average strand size was 110 mm in length, 10-50 mm in width and 0.5-1 mm in thickness. Strands were sorted into 3 fractions: 10-30 mm, 30-50 mm and bigger than 50 mm by a sieving shaker and sieve fractions smaller than 10 mm were categorized as fines. After sorting, the strands were dried in a drying oven at 70°C to reach the target moisture content of 3% to 5%.

Strand boards manufacturing

For panel manufacturing, the strand fractions were mixed in a ratio of 4: 5: 1 (10-30 mm: 30-50 mm: >50 mm). The panel target densities were 400 kg m^{-3} and 500 kg m^{-3}. The strands were loaded in a drum blender with a rotating speed at 30 rounds min^{-1}. PF resin with low and high molecular weight was used as adhesive with the amount of 10% and 20% based on the oven-dry weight of the strands. The low molecular weight (LMW) PF resin exhibited an M_w from 400 to 500 g mol^{-1} and solid content of 53% (Surfactor GmbH, Schöppenstedt, Germany). In addition, the high molecular weight (HMW) PF resin exhibited an M_w from 1000 to 1200 g mol^{-1}, solid content of 70% and was provided by Prefere Resins Germany GmbH (Erkner, Germany). Basic properties of phenol-formaldehyde (PF) resin are listed in Table 1. The resinated strands were manually formed into a mat (450 × 450 mm^2) in a cold pre-press prior to hot-pressing at 140°C for 90 s mm^{-1} using metal bar stops to reach a panel target thickness of 12 mm. In total, 28 panels were produced; two SBs were produced for each of the ten variants and four SBs were produced for each of the two variants of LMW and HMW PF with target density 500 kg m^{-3} at adhesive content 10%.

The actual amount of PF (M_{PF}) on strands was calculated based on the oven-dry mass of strands and solid content of

Table 1. Basic properties of phenol-formaldehyde (PF) resins.

Type of resin	LMW PF	HMW PF
Molecular weight [g mol^{-1}]	400–500	1000–1200
Form	Liquid	Liquid
Color	Light brown, transparent	Red, dark
Solid content at 2 h/120°C [%]	53	70
Viscosity at 20°C [mPa*s]	37.4	240–300
Density at 20°C [g cm^{-3}]	1.1-1.3	1.21

resin stock solution, according to Equation (1):

$$M_{PF} = (M_s \times R_s \times S_c)/100 \quad (1)$$

Where M_s is the oven-dried mass of the strands [g], R_s is the percentage of used resin based on oven-dried mass of the strands [%] and S_c is the solid content of resin [%].

The weight percentage gain (WPG) of strands was evaluated based on the oven-dry mass of strands and amount of PF (M_{PF}) which was sprayed on the strands, according to the Equation (2):

$$WPG = (M_{PF}/M_m) \quad (2)$$

Where M_{PF} is the amount of phenol-formaldehyde of strands [g] and M_m is the oven-dried mass of the strands [g].

Mechanical properties

All specimens were conditioned at 20°C and 65% relative humidity for at least 2 weeks to reach equilibrium moisture content. The density of all specimens was determined according to EN 323 (1993). Three-point bending strength of boards (modulus of rupture (MOR)) and modulus of elasticity in bending (MOE) were determined following EN 310 (1993) on specimens with dimensions of $400 \times 50 \times 12$ mm^3 (seven replicates for each panel, $n = 14$). The test was conducted on a universal testing machine (Zwick Roell Z010, Zwick, Ulm, Germany). The specimens were placed on the center of the supports and the distance between two supports was adjusted to 240 mm. The rate of loading was applied with a testing speed adjusted to cause the failure of the samples within 60 ± 30 s. The failure was defined as a load decrease of 10% or more of the maximum load. The MOE of each specimen was calculated according to Equation (3):

$$E_{MOE} = \frac{L^3(F_2 - F_1)}{4bh^3(\sigma_2 - \sigma_1)} \quad (3)$$

Where: L is the distance between the centers of the supports (mm), b is the width of the specimen (mm), h is the thickness of the specimen (mm), $F_2 - F_1$ is the increment of load on the straight line portion of load-deflection curve (N), $\sigma_2 - \sigma_1$ is the increment of deflection at the mid-length of the specimen (N).

Internal bond strength (IB)

To test IB, the specimens were randomly selected by density group (400 kg m^{-3} ±10%, 500 kg m^{-3} ±10%). IB of the specimens was determined according to EN 319 (1993), with specimen size $50 \times 50 \times 12$ mm^3, the surface of specimen glued onto metal blocks with a hot melt adhesive at 140°C (five replicates for each panel, $n = 10$). IB test was conducted on a universal testing machine (Zwick Roell Z010, Zwick, Ulm, Germany) and tensile load was applied at a speed of 2 mm min^{-1}. The IB value was calculated by the ratio between the maximum load and the area of specimen.

Brinell hardness (BH)

For BH, dimensions of test specimens were $50 \times 50 \times 12$ mm^3. The hardness was assessed following EN 1534 (2000). The maximum test force was 1000 N. The BH (N mm^{-2}) was calculated according to Equation (4):

$$Hb\frac{2F}{\pi D\left(D - \sqrt{D^2 - d^2}\right)} \quad (4)$$

Where D is ball diameter, F is maximum force (N), and d is impression diameter (mm).

Screw withdrawal resistance (SWR)

SWR was determined following EN 320 (1993) but SPAX universal screws (SPAX international GmbH & Co. KG, D-58256 Enneptetal, Germany) were used with dimensions of 4 mm (d) × 35 mm (l). Specimens with dimensions of 50 × 50 × 12 mm^3 were pre-drilled on two sides: screws were located on the middle of specimen's surface and on the side. The crews were manually screwed into the specimen's side until they reached a depth of 15.0 ± 0.5 mm. The screws were pulled out by a clamp on a universal testing machine (Zwick Roell Z010, Zwick, Ulm, Germany). The SWR was the maximum load needed until the failure occurred.

Thickness swelling (TS) and water absorption (WA)

TS and WA were determined after 24 h immersion in water at 20°C according to EN 317 (1993) with samples 50 × 50 × 12 mm^3 (five replicates for each panel, n = 10). Samples were soaked in a water bath with their face vertical. The temperature of the water bath was maintained at 20°C throughout the test period. During the test, specimens were separated from each other by a plastic frame. After 24 h immersion in water, the specimens were taken from the water bath, to remove excess water and measure the dimension and mass of each test specimen.

Irreversible thickness swelling (ITS)

Tests on dimensional stability were conducted with sample dimensions of 50 × 50 mm^2. The strand board specimens were first dried at 103°C to constant mass. The specimens were immersed in water and a vacuum was applied for 60 min at 100 mbar. The thickness and mass of specimens were measured after 24, 48, and 96 h. The specimens were subsequently oven-dried for 24 h at 40, 60, 80, 103°C to determine the dry weight. The leaching percentage was calculated as the ratio between the oven-dry weight of the specimen prior to water-soaking and the oven-dry weight of the specimen after water-soaking.

Contact angle and surface free energy

Strands were resinated by spraying LMW and HMW PF resin in a drum blender with a rotating speed at 30 rounds min^{-1}. A minor amount of the resinated strands was dried at 103°C in a drying oven for 24 h in order to cure the resin. The contact angle was evaluated by the sessile drop technique using drop shape analyzer Krüss GS 10 (Krüss GmbH, Hamburg, Germany) and the corresponding software Krüss DSA 1. The

surface free energy was measured by the methods of Kaelble (1970) and Owens and Wendt (1969). Three treatment variants of strands were randomly chosen; these were strands treated with no resin, with LMW and with HMW PF resin (twenty replicates for each treatment).

Free formaldehyde emission (EN 717-2)

Free formaldehyde emission was determined by gas analysis according to EN 717–2 (1994). The test specimen dimension was 400 × 50 × 12 mm; only specimens of 500 kg m^{-3} prepared with 10% PF resin were tested. A dual-chamber device GA 5000 (Fagus Grecon, Alfeld, Germany) was employed. The edges of the specimens were sealed with aluminum tape before it was placed in a 4-litre cylindrical chamber with controlled temperature (60.0 ± 0.5°C), airflow (60 ± 3 l h^{-1}) and pressure (from 967 to 1054 Pa). The air in the device is conducted into wash bottles in which the free formaldehyde dissolves in water. The actual free formaldehyde value was the average of two specimens after 4 h. At the end, the formaldehyde concentration was determined by the acetyl acetone method (EN 717–2 1994).

Results and discussion

Mechanical properties

All SBs variants containing LMW resin displayed a significantly higher internal bond strength (IB) than those with HMW PF (Table 2), although the weight percent gains (WPG) of panels manufactured with LMW PF was lower than that of respective SBs made with HMW PF (Table 3). With increasing adhesive content, the IB significantly increased as reported previously (Beck *et al.* 2010) for oriented strand board made of trembling aspen (*Populus tremuloides*) and paper birch (*Betula papyrifera*). As expected, the IB of all 500 kg m^{-3} SBs variants was higher than the corresponding 400 kg m^{-3} variants at the same adhesive content (Table 2). The IB values of all SBs variants increased with increasing adhesive content. Similar results were observed for OSB made of Scots pine (*Pinus sylvestris*) (Gündüz *et al.* 2011). At target density of 400 kg m^{-3}, SBs resinated with LMW PF and the mixture of LMW and HMW PF showed a significantly higher IB than those with HMW PF. Remarkably, IB values of SBs containing 10% LMW PF adhesive were about two times higher than those of HMW PF; however, these differences were minor at 20% adhesive content. At target density of 500 kg m^{-3}, clearly higher IB values were observed for SBs containing LMW PF at 10% and 20% adhesive content; in both cases, the IB of panels with LMW PF were approximately 30% higher than those containing HMW PF resin.

As observed for IB, both MOR and MOE increased with the increasing density of the SBs variants (Table 2). The differences between LMW and HMW PF were more pronounced at lower density; LMW PF imparted higher MOR than HMW PF. The Tukey test showed that MOR was only statistically different between LMW and HMW PF at 10% adhesive content for both densities tested. At 20% adhesive content, only slight differences in MOR were observed for the three resin formulations at both densities. The effect of the PF mixture on MOR was not consistent over all variants.

Similar tendencies as described for MOR were also observed for MOE, due to the strong correlation between bending strength and stiffness (Table 2). As observed for IB and MOR, the SWR increased with increasing adhesive content, but there was a correlation between density and SWR. In addition, the SWRs at side orientation was about 30% higher than those of the face. A statistically significant influence of the molecular weight of the PF resin was found for SWR at both face and side orientations except for SBs with a density of 500 kg m^{-3} at 20% adhesive content. The SWR values were highest for panels containing LMW PF, followed by those containing the resin mixture and those containing HMW PF (Table 2).

At target density of 400 kg m^{-3}, Brinell hardness (BH) was higher for panels treated with LMW PF, while at 500 kg m^{-3} target density, no effect of the resin's molecular weight was apparent (Table 2).

Water-related properties

Thickness swelling (TS) increased with increasing density of the panels (Table 4), which is in accordance with earlier

Table 2. Internal bond strength (IB, $n = 10$), modulus of rupture (MOR, $n = 14$), modulus of elasticity (MOE, $n = 14$) screw withdrawal resistance (SWR,) at face and side orientation (each $n = 10$), Brinell hardness (BH, $n = 10$) of kiri strand boards with target densities of 400 kg m^{-3} and 500 kg m^{-3}; mean value ± standard deviation.

Property	Panel den-sity [kg m^{-3}]	Adhesive added related to strand mass					
		10%			20%		
		LMW	Mixture	HMW	LMW	Mixture	HMW
IB [N mm^{-2}]	400	0.45 ± 0.08	0.40 ± 0.08	0.23 ± 0.06	0.65 ± 0.07	0.51 ± 0.13	0.59 ± 0.06
	500	0.53 ± 0.18	0.53 ± 0.09	0.35 ± 0.08	0.91 ± 0.11	0.84 ± 0.11	0.62 ± 0.08
MOR [N mm^{-2}]	400	29.1 ± 3.0	27.0 ± 3.8	22.1 ± 4.7	31.0 ± 7.0	29.9 ± 6.5	28.8 ± 5.9
	500	33.6 ± 6.8	38.6 ± 5.1	29.8 ± 5.4	36.7 ± 8.6	37.6 ± 4.9	37.3 ± 4.1
MOE [N mm^{-2}]	400	4126 ± 261	3608 ± 318	3058 ± 539	4459 ± 764	3917 ± 794	3613 ± 536
	500	4794 ± 789	4954 ± 582	4440 ± 685	4801 ± 961	5002 ± 436	4616 ± 515
SWR-face [N]	400	1211 ± 183	1057 ± 178	893 ± 135	1472 ± 337	929 ± 275	1104 ± 251
	500	1383 ± 134	1142 ± 234	1019 ± 217	1088 ± 260	1325 ± 196	1026 ± 251
SWR-side [N]	400	1574 ± 189	1290 ± 200	1067 ± 195	2014 ± 533	1312 ± 438	1531 ± 342
	500	1699 ± 190	1495 ± 338	1052 ± 228	1561 ± 563	1801 ± 241	1210 ± 193
BH [N mm^{-2}]	400	27.2 ± 10.1	29.1 ± 10.7	20.2 ± 6.2	22.6 ± 8.7	24.0 ± 11.9	17.8 ± 3.6
	500	28.2 ± 15.2	40.4 ± 20.8	33.1 ± 12.0	20.0 ± 10.5	35.8 ± 16.2	28.8 ± 8.7

Table 3. Amount of phenol-formaldehyde (PF) on the strands and weight percent gain of PF resin on strands (WPG).

Board density [kg m^{-3}]	Amount of adhesive per batch [%]	Oven-dry strand weight per batch [g]	Amount of PF per batch [g]			WPG on strand [%]		
			LMW	Mixture	HMW	LMW	Mixture	HMW
500	10	2433	128.8	149.5	170.1	5.3	6.1	6.9
	20	2433	258.1	299.6	340.9	10.6	12.3	13.9
400	10	1947	103.4	120	136.5	5.3	6.2	7.0
	20	1947	206.1	239.3	272.3	10.6	12.3	13.9

studies (Van *et al.* 2019); e.g. Geimer (1982) also showed the effect for flakeboards. In contrast, Chen *et al.* (2009) found a decrease in TS of OSB made from aspen with increasing board density. In the case of SBs from kiri wood, the higher TS was attributed to a high compaction ratio, which results in a stronger spring-back upon water uptake (Van *et al.* 2019). At all variants, panels with higher adhesive content exhibited lower TS than the respective panels at lower adhesive content. At 400 kg m^{-3} target density, TS increased with the proportion of HMW PF in the panels; this effect was much more pronounced at 10% adhesive content than at 20%. The difference between LMW and HMW PF was larger at 500 kg m^{-3} density than at 400 kg m^{-3} density; TS of panels containing HMW PF was 1.8 (10% PF) and 2.0 (20% PF) times higher than that of boards containing LMW PF, while TS of boards containing the PF mixture was more similar to those containing LMW PF. The Tukey HSD test revealed significant differences with respect to the molecular weight of the PF resin.

In reverse to TS, relative water absorption (WA) linearly decreased with increasing panel density (Table 4), while the absolute WA (related to panel volume) was similar for both densities (not shown). For all SBs variants, WA of LMW PF boards was significantly lower than that of the respective HMW PF panels (Table 4). Equivalent results were reported previously for particleboards and SBs from kiri (Nelis *et al.* 2018; Van *et al.* 2019) and aspen OSB (Chen *et al.* 2009). The difference in WA between panels containing LMW and HMW PF was similar for both densities at 10% adhesive content but at 20% adhesive content these difference were significantly larger at 500 kg m^{-3}; WA of panels containing HMW PF was 26% (400 kg m^{-3}) and 54% (500 kg m^{-3}) higher than that of boards containing LMW PF. WA of boards containing the PF mixture was only slightly higher than that of boards containing LMW PF but significantly lower than that of boards containing HMW PF.

Swelling of SBs involves three processes: swelling of the strands itself; release of stress on the densified strands that were compressed during hot pressing (spring-back); developments of voids caused by breaking of adhesive bonds between the strands. Considerable changes in TS and WA may be assigned to the deeper penetration of the LMW resin into voids between the strands, in the lumens and in the cell wall due to a lower viscosity and lower molecular weight. Three effects may be considered if it is assumed that both resin types are stable towards hydrolysis. Firstly, LMW imparted higher IB which indicates that bonding is stronger in these panels. Smaller resin granules staying on the strand surface might reduce the gaps between strands, which might create a better continuously bonding line of SBs. Secondly, LMW PF can better fill voids around the strands in the panels than HMW PF. More and bigger voids enable more capillary water uptake. Penetration into and filling these voids may lead to adhesion between areas that do not occur with HMW PF. Thirdly, LMW may penetrate in the cell wall of wood and cause cell wall bulking, which imparts higher dimensional stability of the strands than HMW PF, which is supposed to remain located at the lumen surface of the cell walls (Kajita and Imamura 1991).

Irreversible thickness swelling (ITS) was determined after water impregnation using vacuum to obtain maximum swelling and subsequent drying of the specimens (Bonigut *et al.* 2014). Generally, ITS was higher for panels of higher density. The values for specimens prepared with LMW PF were significantly lower than for panels prepared with HMW PF, while the ITS of panels containing a mixture of LMW and HMW PF was between those of the other two PF variants (Table 4). The appearance of respective panel

Table 4. Thickness swelling (TS) and water absorption (WA) after 24 h (both $n = 10$), irreversible thickness swelling (ITS) after 1 h vacuum, immersion in water for 96 h and drying ($n = 10$), water absorption (WA-v) and mass loss (ML) after 1 h vacuum, immersion in water for 96 h (both $n = 10$) of kiri strand boards with target densities of 400 kg m^{-3} and 500 kg m^{-3}; mean value ± standard deviation.

Property	Panel desity [kg m^{-3}]	Adhesive added related to strand mass					
		10%			20%		
		LMW	Mixture	HMW	LMW	Mixture	HMW
TS [%]	400	10.2 ± 1.5	14.5 ± 3.5	21.8 ± 3.5	7.8 ± 1.1	9.7 ± 1.7	10.4 ± 1.5
	500	21.5 ± 3.9	23.8 ± 3.3	38.2 ± 2.7	12.3 ± 2.7	15.4 ± 4.2	25.1 ± 3.3
WA [%]	400	82.5 ± 3.2	87.7 ± 5.6	120 ± 4.1	70.7 ± 4.0	75.7 ± 4.5	89.3 ± 4.9
	500	75.4 ± 4.5	81.3 ± 4.4	110 ± 3.8	53.4 ± 4.8	61.9 ± 3.8	82.2 ± 2.6
ITS [%]	400	6.1 ± 1.2	10.6 ± 2.7	18.4 ± 2.7	3.2 ± 0.5	5.5 ± 1.0	6.5 ± 1.0
	500	15.5 ± 1.8	16.4 ± 3.7	28.3 ± 5.5	4.7 ± 1.3	6.5 ± 1.1	13.6 ± 1.6
WA-v [%]	400	193 ± 27	192 ± 21	230 ± 15	167 ± 30	184 ± 21	191 ± 24
	500	110 ± 4	177 ± 29	176 ± 28	150 ± 12	153 ± 27	180 ± 12
ML [%]	400	3.6 ± 0.6	3.9 ± 0.4	5.1 ± 0.6	3.6 ± 0.7	4.2 ± 0.5	5.5 ± 0.5
	500	3.6 ± 0.3	3.6 ± 0.2	4.4 ± 0.4	3.3 ± 0.2	4.0 ± 0.3	4.6 ± 0.2

Figure 1. Photo of representative SB test specimens before (untreated control, middle) and after assessing irreversible thickness swelling (ITS) at 1 h vacuum, immersion in water for 96 h and drying; left: panel produced with HMW-PF; right: panel produced with LMW-PF.

specimens is shown in Figure 1. The values of ITS were somewhat lower than TS after 24 h immersion in water (Table 4).

The water absorption after vacuum (WA-v) and 96 h immersion in water was about 2.3 times higher than that after 24 h water immersion (WA; Table 4), although the respective TS was similar. This indicates that maximum swelling was nearly reached after 24 h, but that many emerging voids in the swelling panel were not filled with water. Additional WA under vacuum-assisted conditions did not significantly contribute to further swelling, because maximum spring-back was reached. Despite lower TS, the relative WA-v was higher for panels of lower density, but the absolute WA-v was higher for panels with higher density (not shown), as already shown for particleboards made of kiri wood (Nelis *et al.* 2018). Panels produced with LMW PF exhibited lower WA-v than those produced with HMW PF, predominantly due to the lower TS of the former.

The mass loss (ML) after vacuum-assisted immersion in water was lowest for panels produced with LMW PF and highest for those with HMW PF; ML of panels produced with resin the mixture lay in between (Table 4). Panels with lower density underwent higher relative ML than those of higher density.

The contact angle (CA) of water on kiri strand surfaces was determined after 2 and 4 s (Table 5). Treated kiri strands originated from the same blending batch of the SBs' manufacturing process, to minimize variations. The water CAs of strands treated with LMW PF were higher than those on the untreated kiri controls and those treated with HMW PF indicating a more hydrophobic surface. A higher surface energy of strand surface leads to a lower hygroscopicity (Kamke and Lee 2007). LMW PF decreased, while HMW PF increased the surface energy compared with the untreated control. The polar part of the surface free energy with LMW-treatment was significantly lower and the dispersive part slightly higher compared to the untreated control.

Table 5. Water contact angle and surface free energy of kiri strands treated with low (LMW-PF) and high molecular weight PF (HMW-PF) and untreated kiri strands. Water droplets were measured at 2 and 4 s ($n = 20$), mean value ± standard deviation.

	Contact angle [°]		Surface free energy [mN m^{-2}]			
	2 s	4 s	2 s		4 s	
Treatment			polar	dispersive	polar	dispersive
Untreated	49 ± 17	46 ± 14	25 ± 12	27 ± 16	23 ± 9	37 ± 5
LMW PF	80 ± 14	76 ± 11	5 ± 5	34 ± 5	7 ± 6	33 ± 6
HMW PF	37 ± 11	27 ± 8	29 ± 7	35 ± 5	34 ± 6	33 ± 5

Table 6. Free formaldehyde emission of kiri strand boards with a target density 500 kg m^{-3} at adhesive content 10% following the European standards EN 717–2 ($n = 2$), mean value ± standard deviation.

	Concentration [mg dm^{-3}]		
Panel density [kg m^{-3}]	LMW	Mixture	HMW
500	2.4 ± 0.5	1.0 ± 0.2	0.3 ± 0.0

Formaldehyde emission

The formaldehyde emission of SBs produced with LMW PF resin was significantly higher than that of panels produced with HMW PF resin (only panels with 500 kg m^{-3} target density and 10% adhesive content were assessed). Panels containing mixtures of both resin types lay in between. This indicates that formaldehyde emission increases with an increasing ratio of LMW resin (Table 6). LMW PF contains a higher amount of both free formaldehyde (FA) and methylol groups and respectively less methylene and methylene-ether bonds than HMW PF due to the lower degree of condensation of LMW PF (Hultzsch 1950). During hot-pressing, the resins undergo further condensation but also cleavage of methylol groups and methylene-ether bonds occur and thus emission of free FA. At equal pressing temperature, the HMW PF resin needs considerable shorter pressing time to complete condensation, which results in a minimum emission of free FA. The high free FA emission of specimens containing LMW PF resin indicates incomplete condensation of these resins. Because the concentration of methylol groups is still high, they can be split off and emitted free FA (Christiansen and Gollob 1985). This indicates that the LMW PF resin in these panels was not fully cured and require considerably longer pressing times than HMW PF. This lowers the industrial feasibility of using LMW PF resins because of a lower reachable production volume.

Conclusion

Utilization of LMW PF as adhesive for SBs made of kiri strands results not only in higher dimensional stability (TS, ITS) but also in better strength properties (particularly IB, SWR) compared to a respective HMW PF, when the same board density is compared. Thus, the amount of LMW PF on the strand surface is sufficient to cause adhesion between the strands. However, using LMW PF requires considerably more severe pressing parameters (higher pressing temperatures and/or longer pressing times) to meet the standard thresholds with respect to formaldehyde emission. These requirements might limit the viability of using LMW PF as

adhesive for OSB production, because of higher energy consumption and longer production times. Using the mixture of LMW and HMW PF could be an option to enhance the properties of SBs based on kiri strands while limiting the formaldehyde emission caused by LMW PF.

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