Investigation of the effects of raw material density and molecular weight of phenolic resin binders on the properties of lightweight panels based on *Paulownia* wood

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List of abbreviations

MOR modulus of rupture

MOE modulus of elasticity

IB internal bond strength

TS thickness swelling

WA water absorption

SWR screw withdrawal resistance

HB hardness Brinell

BD bulk density

VDP vertical density profile

PD peak density

PF phenol formaldehyde

LMW low molecular weight

HMW high molecular weight

OSB oriented strand boards

SBs strand boards

pMDI polymeric diphenylmethane diisocyanate

WPG weight percentage gain

WPC wood polymer composite

VSB veneer strand board

MC moisture content

SEM scanning electron microscopy

X-ray uCT X-ray microtomography

n number of specimens

K kiri

B birch

V veneer

VSB veneer strand boards

DBH diameter at breast height

M_w molecular weight

Abstract

This thesis focuses on different production process conditions of strand boards (SBs), especially with wood species (*Paulownia tomentosa*) by various adhesive binders (pMDI) and their influence on the properties of strand boards. The kiri tree is an extremely fast-growing, ring-porous tree with a low density of about 280 kg m^{-3} and it is mainly grown in plantations. Based on that, this study focused on the production of lightweight strand boards under 500 kg m^{-3}.

In the first part of the study, lightweight strand boards were produced at a target density of 300 kg m^{-3} and 400 kg m^{-3} with three wood species including low-density kiri wood (*Paulownia tomentosa*), medium-density pine wood (*Pinus sylvestris*) and high-density beech (*Fagus sylvatica* L.). However, beech strands did not glue together at these low board densities because of the low compaction ratio. The cohesion of beech strand boards was insufficient for further testing, which is attributed to the lack of bonding contact points for a high-density wood strand at low board density. The higher surface area of kiri SBs only indirectly led to a steep vertical density profile and improved mechanical properties including modulus of rupture (MOR), modulus of elasticity (MOE) and internal bond strength (IB). However, as the total surface area of strand mat increases, the thickness swelling of kiri SBs increases because a higher compaction ratio causes spring-back effects.

In the second phase of the research, strands were blended with low (LMW) and high molecular weight (HMW) PF resin and its mixture 50-50%. LMW acts not only as a treatment agent but also as an adhesive. SBs from kiri timber were produced with densities of 400 kg m^{-3} and 500 kg m^{-3}. At target density 400 kg m^{-3}, the modulus of rupture (MOR) and modulus of elasticity (MOE) of SBs containing LMW PF resin were slightly higher than those of HMW PF at both adhesive contents. Internal bond (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} target density, MOR and MOE of SBs for both PF were approximately similar, while IB of SBs containing LMW PF was about two times as high as that of SBs bonded with HMW PF. This effect was observed at 10% adhesive content but was more pronounced with 20% adhesive content. TS of SBs containing LMW PF was reduced by about 100% (at 20% adhesive content) and 50% (at 10% adhesive content) compared to SBs containing HMW PF. The improved physical and mechanical properties of kiri SBs based on LMW PF might be associated with cell wall matrix stiffening, deeper penetration and larger resin area coverage. Further characterization of LMW and HMW PF resin granules were implemented with various

methods: (1) X-ray microtomography (2) SEM scanning electron microscopy (3) water-proof and filter papers aim to investigate the mechanism of resin expansion and penetration. The enhancement in the internal bond strength and the reduction of thickness swelling of kiri SBs based on LMW PF resin is highly linked to the expansion and penetration abilities of low resin molecules, which are associated with a high compaction ratio of kiri strands resulting in better bonding quality.

At the third phase of the research, an innovative lightweight VSB was manufactured by combining kiri and birch veneers on the surface and inside the panel in order to enhance the mechanical properties of the panel. The results revealed that the MOR and MOE values of VSB meet not only the requirement for OSB\4 but also the requirement for plywood. In contrast, the internal bond strength was reduced, which is attributed to no glue applied on the veneer surface resulting in a weak bonding formation between strands and veneers. The improved mechanical properties might be assigned to the bonding reinforcement between veneers and strands, moreover, the longitudinal grain of veneer was arranged parallel with the orientation of strands.

The fourth phase of research investigated the compaction behavior of wood strands from different wood species associated with different molecular weight phenol-formaldehyde (PF). Strand boards were manufactured at a target density of 500 kg m^{-3} from kiri, beech, pine strands blended with low and high molecular weight PF resin. The IB value of kiri SBs containing LMW PF resin was ten times higher than those of pine and beech. Also, bending properties and screw withdrawal resistance increased linearly with the compaction ratio for both resins. HMW PF resin can only contribute to IB performance until a certain level of compaction ratio (CR) is reached, whereas LMW PF resin starts producing bonding contacts if the compaction ratio goes higher than 1.0 resulting in deformed wood cells on the strand surface. The deformation on the strand surface occurred causing more contact points between strands, which leads to more effective glue utilization.

In the fifth phase of research, the effects of pressing time on the properties and formaldehyde emission of strand boards treated with different molecular weight PF resin were investigated. No clear impact of resin molecular weight and pressing time were observed for bending strength and water-related properties. IB of SBs based on LMW PF was significantly higher than those of HMW PF; however, IB value decreased considerably with increasing press time. The formaldehyde emission decreased linearly with the increasing press time, which might be attributed to the progressive condensation of the resin.

Zusammenfassung

Diese Dissertation befasst sich mit unterschiedlichen Produktionsprozessbedingungen von Spanplatten (SBs), insbesondere mit der neuen Holzart Kiri (*Paulownia tomentosa*) mit verschiedenen Klebstoffen und deren Einfluss auf die Eigenschaften von Spanplatten. Der Kiri-Baum ist ein extrem schnell wachsender, ringporiger Baum mit einer geringen Dichte von ca. 350 kg m-3 und wird hauptsächlich in Plantagen angebaut. Ausgehend davon konzentrierte sich diese Studie auf die Herstellung von leichten Spanplatten unter 500 kg m-3.

Im ersten Teil der Studie wurden leichte Spanplatten mit einer angestrebten Dichte von 300 kg m-3 und 400 kg m-3 mit drei Holzarten hergestellt, darunter Kiri-Holz niedriger Dichte (*Paulownia tomentosa*), Kiefernholz mittlerer Dichte (*Pinus sylvestris*) und Buche hoher Dichte (*Fagus sylvatica* L.). Buchenstrands verkleben jedoch bei diesen niedrigen Plattendichten wegen des niedrigen Verdichtungsverhältnisses nicht miteinander. Die Kohäsion von Buchenspanplatten war für weitere Tests unzureichend, was auf das Fehlen von Verleimungskontaktpunkten der hochdichten Buchenstrands bei niedriger Plattendichte zurückzuführen ist. Die größere Oberfläche von Kiri SBs führte zu einem steilen vertikalen Dichteprofil und verbesserten mechanischen Eigenschaften, einschließlich Biegefestigkeit (MOR), Elastizitätsmodul (MOE) und innere Quezugfestigkeit (IB). Mit zunehmender Gesamtoberfläche der Spanmatte nimmt jedoch auch die Dickenquellung von Kiri SBs zu, da ein höheres Verdichtungsverhältnis zu akkumulierten spring-back-Effekten führt.

In der zweiten Phase der Forschung wurden SBs mit PF-Harz mit niedrigem (LMW) und hohem Molekulargewicht (HMW) bei einem Mischungsverhältnis von 50-50% gemischt. LMW wirkt nicht nur als Modifizierungschemikalie, sondern auch als Klebstoff. SBs aus Kiriholz wurden mit Dichten von 400 kg m-3 und 500 kg m-3 hergestellt. Bei der angestrebten Dichte von 400 kg m-3 waren der Bruchmodul (MOR) und der Elastizitätsmodul (MOE) von SBs, die LMW PF-Harz enthielten, bei beiden Klebstoffgehalten etwas höher als die von HMW PF. Innere Haftfestigkeit (IB), Schraubenauszugswiderstand (SWR) und Dickenquellung (TS) von SB, die LMW PF enthalten, waren bei 10% Klebstoffgehalt signifikant höher als die von HMW PF, und die Unterschiede nahmen leicht ab, als der Klebstoffgehalt auf 20% stieg. Bei 500 kg m-3 Zieldichte waren MOR und MOE von SB für beide PF ungefähr ähnlich, während IB von SB, die LMW PF enthielten, etwa doppelt so hoch war wie die von SB, die mit HMW PF verklebt waren. Dieser Effekt wurde bei 10% Klebstoffgehalt beobachtet, war jedoch bei 20% Klebstoffgehalt ausgeprägter. Der TS von SB wurde bei SWR durch LMW PF im Vergleich zu HMW PF bei beiden Klebstoffgehalten um etwa 100% (bei 20% Klebstoffgehalt)

und 50% (bei 10% Klebstoffgehalt) erhöht. Die verbesserten physikalischen und mechanischen Eigenschaften von Kiri-SBs auf der Basis von LMW PF könnten mit einer Versteifung der Zellwandmatrix, einer tieferen Penetration und einer größeren Harzflächenabdeckung zusammenhängen. Eine weitere Charakterisierung der LMW- und HMW PF-Harzgranulate wurde mit verschiedenen Methoden durchgeführt: (1) Röntgenmikrotomographie (2) REM-Rasterelektronenmikroskopie (3) Wasserdichte und Filterpapiere zielen darauf ab, den Mechanismus der Harzausdehnung und -penetration zu untersuchen. Die Verbesserung der inneren Haftfestigkeit und der Dickenquellung von Kiri-SBs auf der Basis von LMW PF-Harz ist eng mit den Ausdehnungs- und Penetrationsfähigkeiten niedriger Harzmoleküle verbunden, die mit einem hohen Verdichtungsverhältnis der Kiri-Stränge verbunden sind, was zu einer besseren Bindungsqualität führt.

In der dritten Phase der Forschung wurde ein innovatives leichtes VSB durch Kombination mit Kiri- und Birkenfurnieren auf der Oberfläche und im Inneren der Platte hergestellt, um die mechanischen Eigenschaften der Platte zu verbessern. Die Ergebnisse zeigten, dass die MOR- und MOE-Werte von VSB nicht nur die Anforderung für OSB\4, sondern auch die Anforderung für Sperrholz erfüllen. Im Gegensatz dazu war die innere Haftfestigkeit reduziert, was darauf zurückzuführen ist, dass kein Leim auf die Furnieroberfläche gegeben wurde, was zu keiner Verbindungsbildung zwischen Strands und Furnieren führte. Die verbesserten mechanischen Eigenschaften könnten der Verleimungsverstärkung zwischen Furnieren und Strands zugeschrieben werden, zudem wurde die Längsmaserung der Furniere parallel zur Orientierung der Strands angeordnet.

Die vierte Forschungsphase untersucht das Verdichtungsverhalten von Holzspänen aus verschiedenen Holzarten in Verbindung mit Phenol-Formaldehyd (PF) mit unterschiedlichem Molekulargewicht. Es wurden Spanplatten mit einer Zieldichte von 500 kg m-3 aus Kiri-, Buchen- und Kiefernholzstrands hergestellt, die mit nieder- und hochmolekularem PF-Harz gemischt wurden. Der IB-Wert von Kiri SBs mit LMW PF-Harz war zehnmal höher als der von Kiefer und Buche. Auch die Biegeeigenschaften und der Schraubenauszugswiderstand nahmen linear mit dem Verdichtungsverhältnis zu beide Harze. HMW PF-Harz kann nur so lange zur IB-Leistung beitragen, bis ein bestimmtes Niveau des Verdichtungsverhältnisses (CR) erreicht ist, wohingegen LMW PF-Harz beginnt, Bindungskontakte herzustellen, wenn das Verdichtungsverhältnis höher als 1,0 geht, was zu verformten Holzzellwänden auf der Spanoberfläche führt. Die entstandene Verformung auf der Spanoberfläche verursacht mehr Kontaktpunkte zwischen den Spänen, was zu einer effektiveren Leimausnutzung führt.

In der fünften Forschungsphase wurden die Auswirkungen der Presszeit auf die Eigenschaften und die Formaldehydabgabe von Spanplatten untersucht, die mit PF-Harz mit unterschiedlichem Molekulargewicht behandelt wurden. Es wurden keine eindeutigen Auswirkungen des Molekulargewichts des Harzes und der Presszeit auf die Biegefestigkeit und die wasserbezogenen Eigenschaften beobachtet. Der IB-Wert von SBs auf der Basis von LMW PF war signifikant höher als der von HMW PF; der IB-Wert nahm jedoch mit zunehmender Presszeit erheblich ab. Die Formaldehyd-Emission nahm mit zunehmender Presszeit linear ab, was auf das tiefere Eindringen und die größere Flächendeckung des LMW PF-Harzes zurückzuführen sein könnte.

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1. General introduction

1.1. Kiri tree (*Paulownia tomentosa*)

1.1.1. Original and cultivation conditions of Paulownia tree

The kiri tree (*Paulownia tomentosa*) is a fast-growing, ring-porous tree with very light timber, with an average oven-dry density of approx. 300 kg m^{-3} (Olson and Carpenter 1985; Akyildiz and Kol Sahin 2010) to 350 kg m^{-3} (Flynn and Holder 2001). There are two other species, *Paulownia elongate* and *Paulownia fortunei*, which also grow rapidly but are hardly cultivated in block plantations. The genus *Paulownia* is natively distributed in East Asia, which has in temperate subtropical and tropical climate (Hua et al. 1986; Cossalter and Pye-Smith 2003). Nevertheless, they have high adaptability to different climate conditions (Zuazo et al. 2013). A *Paulownia* tree, which could grow up to 3 m in a year under ideal conditions, can reach a height of 10-20 m (Kalaycioglu et al. 2005; Si et al. 2008).

Flowers of *Paulownia* usually bloom in May and June with colour from white to light purple, while the fruit ripens in autumn (Barton et al. 2007; Si et al. 2008). A stem of a 10-year-old tree can reach 30-40 m in diameter at breast height (DBH) (Kalaycioglu et al. 2005b; Barton et al. 2007; Ates et al. 2008; Yadav et al. 2013). Also, the cultivation of *Paulownia* is identified as an effective tool to reduce climate change because of the high absorption of CO_2 from the air (Zuazo et al. 2013). The *Paulownia* tree, which can be harvested after 15 years for valuable timber, could produce approximately a cubic meter of wood at aged 5-7 years old with a density of 2000 trees/ha providing a total production of 330 t/ha (Gonçalves et al. 2008; Si et al. 2008).

1.1.2. Properties of kiri wood

Paulownia timber, which is reported with an averages wood density of approx. 300 to 350 kg m^{-3}, is considered as soft, lightweight, ring-porous, straight-grained, low thermal conductivity, free of warping and knot-free wood; however, kiri wood exhibits very low hardness and strength (Kalaycioglu et al. 2005b; Ates et al. 2008; Candan et al. 2013; Yadav et al. 2013; Akyildiz 2014; Koman et al. 2017). Due to the low density, wood of *Paulownia* can dry quickly in open-air conditions without causing any serious drying defects and has excellent machining and finishing properties (Akyildiz and Kol Sahin 2010). Kiri wood has been suggested not to be suitable for load-bearing applications as heavy construction which usually requires high bending strength (Akyildiz and Kol Sahin 2010; Koman et al. 2017). Nevertheless, kiri wood, which has lower shrinkage and swelling properties than other species, might be used as a material for building components where non-load bearing isolation is needed.

Under the flame condition, the porous wood structure of *Paulownia* becomes easily carbonized; however, it is difficult to ignite because oxygen is insufficiently supplied in the big and independent vessel structure (Li and Oda 2007). Low-quality kiri tree (*Paulownia tomentosa*) has proven the potential as raw material to produce particleboard with a density of 550 and 650 kg m^{-3}, which meets the minimum strength requirements of the European Standards for general uses (Kalaycioglu et al. 2005b). Also, kiri wood has been used to manufacture particleboard with a density of 350, 550 and 650 kg m^{-3}; however, particleboard with a density of 650 kg m^{-3} caused blisters after hot pressing (Nelis et al. 2018). Kiri timber has been displayed as a potential raw material for the particleboard industry. Thin cell walls and very large vessels of kiri wood were showed in Figure 1.

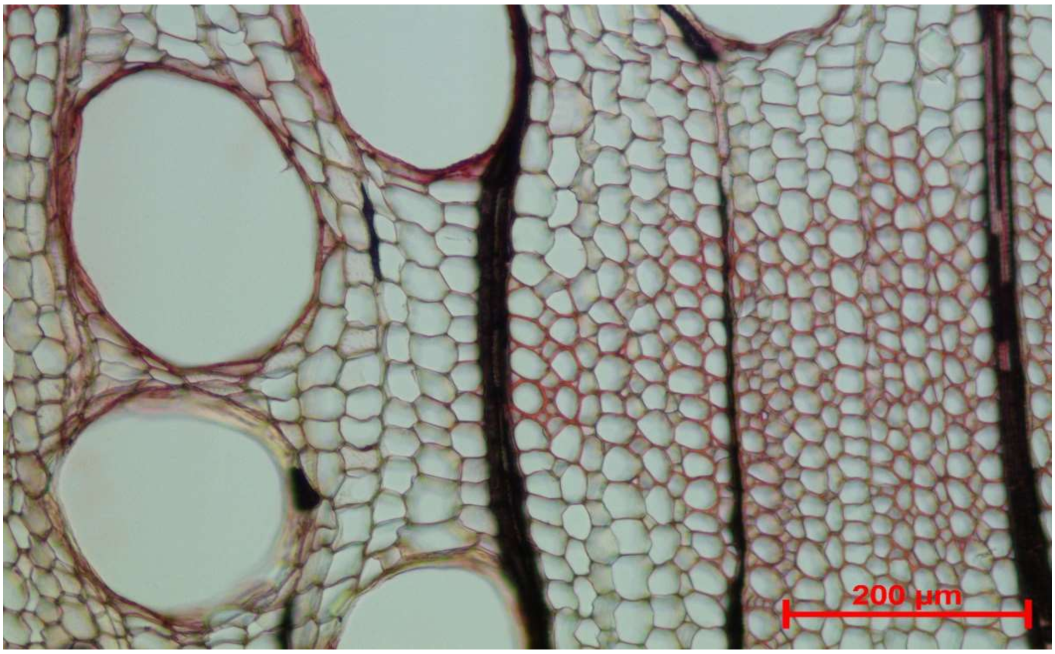

Figure 1 Transverse sections of kiri wood showing cell walls and lumina (Scale bar 200 µm).

1.1.3. Potential biomass and worldwide interest

The demand for global bioenergy is rapidly increasing, hence the European foresters are expanding progressively the cultivation of energetic plants. In addition, the biomass demand is anticipated to grow by 50% in the year 2020 in Europe (Zuazo et al. 2013). Among of main sources of global bioenergy, the fast-growing woody crops play an essential part in the generation of bioenergetics biomass. *Paulownia* is one of the fastest-growing species in the world with a low concentration of ash, sulphur and bioethanol, which will be an important plant of the global bioenergy sector (Zuazo et al. 2013; Yadav et al. 2013). With a rapid growth rate, *Paulownia* wood has a high cellulose concentration up to 440 g cellulose/kg that might be suitable for solid biofuel and cellulose pulp industry (Ates et al. 2008; Yadav et al. 2013). A trihybrid clone *Paulownia fortunei* × *tomentosa* × *elongate* has been investigated for the pulping production process, which has ash content (0.89%) and cellulose content (44.0%)

Many European countries and the United States have put the effort to cultivate the *Paulownia* tree for timber as it is not very competitive in closed forests. Brazil, Paraguay, Argentina and Australia have developed this species in plantations; however, there is only limited success (Cossalter and Pye-Smith 2003). These include both short-rotation plantations for the production of biomass and plantations for the production of quality timber with rotation ages of ca. 20 years (Barton et al. 2007). The utilization of these low-density wood species for the manufacture of the light-weight wood-based panel provides an opportunity to convert a low-value raw material into an added-value product. In the USA, *Paulownia* plantations exist since the 1970ies, while there is recently an increasing demand for wood raw materials in Europe (Barton et al. 2007). *Paulownia* tree offers great potential for plantation management in the area of Europe. A cultivation project for sustainable wood production was developed by a company since 2009 (WeGrow GmbH, Saxer 2018).

1.2. Oriented strand boards (OSB)

Oriented strand boards were first produced in the 1950s by utilizing a low-quality resource; however, it has been introduced to the market only in the late 1970 but widely used nowadays as an important component of the construction industry (Kloeser et al. 2007). As a low-cost substitution for plywood, OSB is manufactured from wood stems shredded into rectangular shape flakes with a length of few centimeters, the thickness of circa 1 mm, and width from 10 to 50 mm. These strands are generally laid in a crosswise direction in the core layers, whereas in the face layers strands are oriented longitudinally in line with the panel length (Brinkmann 1979; Barnes 2002a, b; Lee et al. 2006).

1.2.1. Strand dimension and its impact on panel properties

Longer strands improved the bending properties of OSB, whereas shorter strands resulted in a better internal bond strength (IB) property (Barnes 2001; Beck et al. 2009). OSB made from Betung bamboo with a strand length of 70 mm had higher values of MOR and MOE compared to 60 and 50 mm strand length (Febrianto et al. 2012b). The orientation of face strands is one of the key factors influencing the properties of OSB (McNatt et al. 1992a; Meyers 2001; Nishimura et al. 2001a; Barnes 2002a, b; Chen et al. 2008). A weak relationship between the MOR and alignment angle of the strand was found by using an image analysis method (Nishimura et al. 2001b). In contrast, McNatt found that alignment of the strand panel on the surface improved the bending strength and stiffness of OSB; however, no impact was identified for either IB or thickness swelling (TS) (McNatt et al. 1992). Analysis of the strand mat structure indicated that strands of the upper side are bigger than the strand of the lower side, which is attributed to the vibration of the strand mat (Nishimura et al. 2002). Hence, the arrangement of the wood strand needs to be optimized to ensure the output quality of OSB products.

Image analysis is considered as a powerful method to determine the distribution of strand orientation by using a CCD (charge coupled device) digital camera (Nishimura et al. 2001). Alignment of strands is primarily a surface layer phenomenon of panel structure that contributes much more to parallel MOE, although the improvement of MOE is a variable that depends on face/core ratio and strand angle distribution (Chen et al. 2008). The effect of thin hinoki strands with various face:core:face ratios (Japanese cypress, *Chamaecyparis obtuse* Endl.) on the properties of three-layered OSB also has been studied (Zhang et al. 1998). The results showed that 10-20% of thin strands in the surface layer significantly improved MOR and MOE by about 47%-124% and 30%-65% respectively. This improved property is attributed to the peak density of the vertical density profile shift to the board surface as the face/core ratio increased.

1.2.2. Raw material density and its impact on panel properties

OSB is a wood engineering product in the European Union as its first standard which was defined in 1997 (USDA 1999). Nevertheless, OSB has been developed with trembling aspen in Canada for a long time, while southern pine and aspen are widely utilized in North America (Canadian Forest Service 1995; Wang and Winistorfer 2000). However, as a major structural material of the construction industry, the demand for the OSB market has rapid growth to levels that are not sustainable with only aspen as a raw material (Canadian Forest Service 1995). The production of OSB in North America and Europe gradually grows because there is an increasing demand by the construction producer (Meil et al. 2007).

Many researchers have put their effort into identifying other alternative wood raw materials for OSB production. Hybrid poplar (*populous* × *euramericana*) has been used to produce OSB and the study found that increasing panel density resulted in a higher MOR, MOE, IB, and SWR but a negative impact on thickness swelling (Zhou 1990). Southern pine (*Pinus spp.*) was used to manufacture OSB but the strands were impregnated with low molecular weight PF resins to improve dimensional stability (Wan and Kim 2007). They pointed out that strand geometry and species had no clear impact on IB but an influence on bending properties.

Beech (*Fagus sylvatica*) and poplar (*Populus tremula*) wood are considered as feasible materials to reduce the dependence on pine (*Pinus sylvestris*) for the production of OSB at a target density of 720 kg m^{-3} (Akrami et al. 2014a, b, c, 2015; Akrami and Laleicke 2017). Low-density kiri wood (*Paulownia tomentosa*) improved the performance of OSB with target densities of 300 and 400 kg m^{-3} compared to pine (*Pinus sylvestris*) and beech (*Fagus sylvatica*) (Pham et al. 2019). Three fast-growing wood species in Romania including birch, willow and poplar were used to produce OSB with a density of 610 kg m^{-3}, which meet the standard requirements for OSB/2 EN 300 (2006) (Dumitrascu et al. 2020).

In Asia, bamboo has widely been used as a very promising raw material to produce OSB because it is abundantly available in many tropical countries (Sumardi et al. 2007; Malanit et al. 2011; Febrianto et al. 2012a, 2015; Yong et al. 2012; Sumardi and Suzuki 2013). Strands from Betung bamboo (*Dendrocalamus asper* (Schultes.f) Backer ex Heeyne) with a length of 50, 60 and 70 mm were manufactured by using a wood planner. It showed that MOR and MOE of OSB made from strands with 70 mm strand length were higher than those of 50 and 60 mm (Febrianto et al. 2012). The bending strength of OSB containing the mixture of bamboo and poplar reached the maximum value at the ratio of 50:50, while IB decreased with the decreasing proportions of bamboo (Yong et al. 2012).

The compaction ratio between board density and wood density is considered as one of the most important factors for influencing the final properties of particle-based panels (Kelly 1977). Wood raw-material density increases resulting in less contact between strands, which is considered as one of the most important variables in wood composites (Dai et al. 2007). Kiri wood as a very low wood density species has been used to manufacture particleboard with a density of 350, 500 and 650 kg m^{-3}; however, particleboard with a density of 650 kg m^{-3} exhibited blisters after hot pressing at temperature 200°C (Nelis et al. 2018). These blisters can be explained by the low in-plane permeability of particleboard because the soft and low-density kiri wood caused too high compaction ratio at a board density of 650 kg m^{-3}. In the early stage of hot pressing, the heat is transported by conduction as a result of the direct contact between the hot-plates and the wood particles (Hood et al. 2005). The transverse permeability is reduced as the mat is compressed to the board target thickness, which leads to the build-up of internal gas pressure (Fakhri et al. 2007). As the pressing process is continuing, heat transport is transferred by convection and bulk flow of moisture (Dai and Yu 2004a; Fakhri et al. 2005; Hood et al. 2005a; Fakhri et al. 2007; Zhang and Smith 2009). The combination of convective heat transfer and in-plane flow of moisture is dependent on the available pathways inside the mat which might be rather controlled by the compaction ratio of the panel and the hardness of the particle.

1.2.3. Application of oriented strand boards

Wood-based panels serve as the main raw material for the furniture and construction industry. Among engineered structural wood products, OSB is less costly and provides sufficient strength properties for the residential construction in North America compared to other structural panels such as plywood (Spelter et al. 1997; Brochmann et al. 2004; Gu et al. 2005). In the United States housing market, the demands of OSB in housing construction has rapidly been growing even faster than the domestic production capacity of OSB (Wear et al. 2016). In 2011, the consumption of OSB in the United States market has reached 11.2 million m^3 (constituting 60% of the structural panel market) (Howard and McKeever 2013). In the next year, 9.8 million m^3 of OSB were manufactured; however, the growing residential construction sector is anticipated to increase OSB consumption in 2013 to about 16 million m^3 (Howard and McKeever 2013).

1.3. Lightweight board structure

The lightweight panel's topic has been discussed for more than one decade. In general, there is no proper definition for the lightweight board, however, it can be stated that lightweight board densities are less than 500 kg m^{-3} (Poppensieker and Thömen, 2005). Reduction of the density is being implemented in various ways by using low-density wood species, lower densification of the material, adding light materials and a low-density core structure (Nilsson et al. 2013). In low-density structure, the weight of materials is reduced by replacing heavy core layers with various types of lightweight materials such as low-density wood, hollow materials, honeycomb paper, polystyrene foam, polyurethane foam, polycarbonate, polypropylene, ultra-lightweight foam, and wood fiber-based, maize granular (Karlsson and Tomas\AAström 1997; Jivkov et al. 2012; Shalbafan et al. 2013; Burnett and Kharazipour 2018).

1.3.1. Sandwich structure

The concept of sandwich construction was first stated by Noor, Burton and Bert in England in 1849 (Allen 1969). Nowadays, the trend of using the sandwich structure is rapidly developing in many sectors such as satellites, aircraft, ships, automobiles, rail cars, wind energy systems, and bridge construction (Thomsen et al. 2005). A typical sandwich structure consists of three layers with two outer facings which are separated by a thick middle-layer of low-density materials (Allen 1969; Birman and Kardomateas 2018). The sandwich structure has also been employed in the wood industry to achieve low-density panels with the use of plywood, thin particleboard and medium-density fibreboard (MDF) as faces and various core materials as honeycombs, expanded foams and end-grain balsa wood in the multi-stage process (Shalbafan et al. 2012b). The honeycomb sandwich panels have been developed to offer cost savings for furniture applications and the use of honeycombs in sandwich construction provides the highest strength to weight and stiffness ratio (Pflug et al. 2004).

The choice of face and core materials of the sandwich structure depends on the function of the products; however, face material might be related to standard thicknesses while core materials might be restricted in the choice of thickness and density (Allen 1969; Birman and Kardomateas 2018). The use of material combinations might be required to optimize the function of each component in order to achieve optimal structural performance (Pflug et al. 2004).

1.3.2. Core materials

The core layer of wood-based panels did not considerably contribute to in-plane stresses when the panel is loaded, thus it can be produced from lighter and less expensive materials (Pflug et al. 2004). The core layer can be very light because its main function is to keep the facings at a certain distance. In contrast, the materials of surface layers should be strong and stiff to maintain bending strength and resist normal loads (Allen 1969; Mohammadabadi et al. 2018). This lightweight structure offers a possibility to save material because of its three-layered composition consisting of two thin outer layers which are separated by a light core (Shalbafan et al. 2012c). Popcorn sandwich panels have been produced with popcorn in the core layer and poplar plywood as face materials, however, the mechanical properties of this composite are limited (Burnett and Kharazipour 2018). Another solution that lightweight products could offer is the application of wood-based foam core panels for the furniture industry, which may meet the property requirement of particleboard while reducing the amount of raw material input (Shalbafan et al. 2012c). In the furniture industry, paper honeycomb panels are the most-used common ones which could provide relatively good strength properties and price-thickness ratio when the thickness is above 25 mm (Jivkov et al. 2012).

The choice of core materials for the sandwich structure depends on the function of the products and it might be restricted in the choice of thickness and density (Allen 1969; Birman and Kardomateas 2018). The optimization of the sandwich structure is based on a given ratio between core depth and face thickness. The core materials might be mainly divided into four types: foam or solid core, honeycomb core, truss core, web core (Thomsen et al. 2005). The honeycomb core material can provide sufficient compression properties at a low density (Pflug et al. 2004). On the other hand, foam core may offer thermal, acoustic insulation and fire resistance properties (Barbu 2015). The application of lightweight material for construction, transportation and furniture industry in the future is needed as the social trend of human mobility will prompt manufacturers of furniture to design them in a way that they are flexible in terms of mobility (Poppensieker and Thömen 2005; Lüdtke 2011). Nevertheless, the use of low-density structures in the wood-based panel industry has developed pretty slow because of the high costs of core material and processing (Shalbafan et al. 2012a).

1.3.3. Surface materials

The surface material of the sandwich structure must be much stiffer than the core material. Principally, the axe of the face material must be located parallel with the sides of the panel (Allen 1969). Fiber-reinforces polymers, fiberglass-reinforced polyester, thick veneer, thick plywood, particleboard and high-density fiberboard are commonly used as face materials in a lighter wooden structure (Karlsson and Åström 1997; Barbu 2015). The choice of face material is usually restricted because mechanical performance is significantly depending on surface quality (Pflug et al. 2004). Bending properties, MOR and MOE, of the sandwich structure relies to a large extent on the stiffness of the face materials (Allen 1969).

1.3.4. Lightweight wood-based panels

Wood raw-material density and compaction ratio of the mat are regarded as two of the most important factors for controlling the final density of particle-based panels (Kelly 1977). The average density of conventional OSB is significantly dependent on the wood species which is used for the manufacture (Chen and Wellwood 2010). Multilayered panels that have low weight and high dimensional stability and screw withdrawal resistance, have been produced from several light species such as albizia, balsa, ceiba, fuma, ilomba for the door and furniture industry (Barbu 2015). Until now there is no standard to categorize the wood-based panels in terms of their weight. The lightweight panels were widely known and mainly classified in terms of their density: lightweight with a density below 500 kg m^{-3}; very lightweight with a density below 350 kg m^{-3} and ultra-lightweight with a density below 200 kg m^{-3} (Grbac 2012).

1.3.5. Vertical density profile and peak density

Vertical density profile (VDP), which is formed with a typical U-shape with high-density surfaces and low-density core, describes the board density at each constituent layer of board thickness (Jin et al. 2009). VDP shapes affect the mechanical properties of wood-based panel products (Kelly 1977; Xu and Suchsland 1997a; Xu 1998; Wong et al. 1999a). In addition, several studies investigate the effect of heat transfer, moisture movement, pressing strategies on the formation of VDP (Kelly 1977; Wang and Winistorfer 2000; Houts et al. 2004; Dai et al. 2007b, 2008; Jin et al. 2009; Li et al. 2009).

It was demonstrated that the MOE of the board increased linearly as the slope of VDP and peak density increased (Xu and Suchsland 1997b; Jin et al. 2009). MOR and MOE of boards with conventional density profiles were higher than those of homo-profile. Remarkably, it is stated that maximum MOE occurs when peak density is located at approximately 14% of the board thickness under the board surface (Jin et al. 2009). When OSB producers try to make a board density below 560 kg m^{-3}, problems will occur because of the porous appearance of the core layer causing lower strength properties. A homogenous vertical density profile was formed by preheating the wood strand and controlling the moisture content of the strand mat before it is going to pressing to lower the conventional OSB density (Chen and Wellwood 2010).

Fiberboard and particleboard with a flat, homogeneous (homo-profile) and typical U-shape (conventional density profile of wood-based panel) along the thickness have been investigated by using the manipulation of mat moisture content distribution, press closing speed and hot pressing (Wong et al. 1998, 1999, 2000, 2003). Peak density of fiberboard at a target density of 500 kg m^{-3} increased from 0.5 to 1.1 kg m^{-3}, resulting in an increase for MOR and MOE up to 67% and 62% respectively (Wong et al. 2000). Similar results were observed at a board density of 700 kg m^{-3}. Therefore, it is concluded that the value of peak density shows a linear correlation with the bending strength of wood-based panels. However, the IB value of the conventional density profile was lower than those of homo-profile because of the low core density (Wong et al. 1999).

1.4. Modification of wood composites

The wood-based panel responds sensitively to variations of temperature and relative humidity with changes in moisture content. The low dimensional stability might limit the service-life of wood-based panels greatly, especially when applied in exterior conditions. In order to improve the service life of wood-based panels, wood particles may be impregnated with resins or other chemicals. As another alternative, thermal modification of strands at high temperatures provides decay resistance and dimensional stability; however, it results in a reduction in the bending properties of the panel.

1.4.1. Chemical modification of wood-based panels

By combining three percent phenol-formaldehyde adhesive and seven percent impregnating phenol-formaldehyde resin, a reduction in thickness swelling of particleboard can be observed (Haygreen and Gertjejansen 1971). Spruce (*Picea abies*) strands were acetylated by reaction with acetic anhydride at 120°C for 30 and 60 min, which significantly reduced the thickness swelling and water absorption of OSB; however, IB decreased about 19% compared to control boards (Papadopoulos and Traboulay 2002). Two forms of maleic anhydride polypropylene (powder and emulsion) were added with polymeric diphenylmethane diisocyanate (pMDI), which considerably causes a reduction in mechanical properties of OSB (Ruffing et al. 2009).

1.4.2. Thermal modification of wood-based panels

Thermal modification of wood-based panels, which can be categorized into two approaches: pre-treatment and post-treatment, has been used to improve dimensional stability. Thermal modification normally starts applying the heat from temperature 160°C to 260°C (Direske et al. 2018). Pine strands were hydrothermally treated at 170°C for 7 and 21 min, which efficiently reduces the equilibrium moisture content of strand and improves the dimensional stability of boards; however, MOE did not alter in all treatment and MOR was higher in the panels containing strands treated at 170°C for 7 and 21 min (Carvalho et al. 2018). Physical properties of OSB as thickness swelling and water absorption was significantly improved by thermal treatment. Nevertheless, at a higher temperature 240°C within 60 minutes a negative effect on mechanical properties was witnessed (Farinassi Mendes et al. 2013).

A similar reduction in mechanical properties of thermally treated panels was observed in several studies (Paul et al. 2007; Menezzi et al. 2009; Mendes et al. 2013a; Andrade et al. 2016; Direske et al. 2018; Pipíška et al. 2020). One of the simple reasons for the reduction in strength may be the loss of substance that normally occurs during thermal treatment. Degradation of hemicelluloses and hydrolysis of cellulose (in presence of acetic acid) in the wood cell could occur, resulting in a weaker wood structure, which might explain the reduction in mechanical properties of OSB (Aro et al. 2014). In contrast, the moisture-related properties of the thermally treated wood-based panel were considerably improved and the pre-treatment showed a more pronounced effect on physical properties than post-treatment methods (Mendes et al. 2013).

1.5. Influence of resin on the properties of wood composites

Higher loading contents of polymeric 4,4´-methylene di-phenyl-de-isocyanate (pMDI) resin could enhance the properties of wood-based panels; for instance, thickness swelling reduced 30% when loading resin increased from 2% to 6% (Buckley et al. 2002). The physical and mechanical properties of wood-based panels are mainly depending on the resin content and wood material density (Barbuta et al. 2011; Febrianto et al. 2015).

1.5.1. Low and high molecular weight phenol-formaldehyde resin

Phenolic resin (phenol-formaldehyde PF), which is easily recognized by the dark color of the bonding line and on the board surface, exhibits high resistance to hydrolysis of the C-C bond (Pizzi 1983; Pocius and Dillard 2003). Therefore, they can be used for water-resistant bonding lines to manufacture wood-based panels such as OSB or plywood for exterior applications. Sodium hydroxide (NaOH) is usually used for resoles during the process of phenolic resin synthesis which contains oligomeric and polymeric chains as well as monomeric methylol-phenol, free formaldehyde and unreacted phenol (Pizzi 1983; Knop and Pilato 1985a; Gardziella et al. 2000). The curing reaction of PF can only be initiated by heat with the transformation of molecules of various sizes via chain lengthening, branching, and cross-linking to a three-dimensional network (Pizzi and Mittal 2018a). Under hot-pressing in composite manufacture, the hardening behavior of PF resins significantly depends on lignocellulosic substrates whereby the available heat of the resin hardening process was much lower than for the resin alone. The catalytic reaction of the PF condensation might be activated by carbohydrates (Knop and Pilato 1985b; Gardziella et al. 2000; Pocius and Dillard 2003; Pilato 2010). Covalent bonds play only a minor role in bonding performance, which might be formed between the PF resin and the various wood components (Pizzi and Mittal 2018a).

Aqueous phenol-formaldehyde has commonly been used as a wood treatment agent and as an adhesive binder in the woodworking industry (He and Riedl 2004). Wood composites manufactured with phenol-formaldehyde resin as a binder displayed excellent properties such as IB, MOR, MOE, TS (Kajita and Imamura 1991; Barbuta et al. 2011b; Hong et al. 2018). A mixture of low and high molecular weight PF resin might be a promising option to produce wood-based panels with improving physical and mechanical properties. Haygreen and Gertjejansen (1971) manufactured particleboards from aspen with a combination of PF adhesive and PF impregnating resin, improving significantly the thickness swelling and spring back of panels. The influence of the molecular weight of PF adhesives on the properties of MDF from commercial softwood was investigated by mixing low and high molecular weight in different proportion LMW:HMW from 100:0, 80:20, 60:40, 40:60, 20:80, 0:100.

Japanese cedar (*Cryptomeria japonica D. Don*) particles were dipped into aqueous resin solution or sprayed with low molecular weight PF resin solution (M_w = 390) before spraying the high molecular weight PF adhesive. The effect of resin molecular weight on the bonding performance of aspen flake boards was studied (Stephens and Kutscha 1986). High molecular weight (M_w >1000) performed as conventional adhesives although dimensional stability of aspen flake boards was poor, while low molecular weight alone (M_w <1000) exhibited a poor bonding quality as a binder.

1.5.2. Strand deformation and compaction ratio of strand boards

The properties of strand boards are a result of various important factors such as wood material density, particle production parameters, board density, adhesive, pressing parameters. Many factors explain the differences in the board properties of the various wood species. However, the oriented strand board properties can be explained by the strand deformation and compaction ratio between board density and wood material density. High-density wood species led to higher bending strength when used to produce wood-based panels at the same compaction ratios than low-density wood species; however, resulted in lower bending strength at the same level of board density (Kelly 1977; Xu and Suchsland 1997a). Strand deformation, which is dependent on the compression strength, might contribute to the adhesive bonding performance.

1.5.3. Influence of resin distribution on the board performance

Adhesive is probably adjustable processing variants, which plays an important part in the wood panel production cost. Hence, understanding the adhesive distribution is of great importance not only to optimize the production parameter but also to minimize production costs (Dai et al. 2005a). Various studies (Johnson and Kamke 1994; Kamke et al. 1996; Buckley et al. 2002; Grigsby and Thumm 2004, 2012a; Dai et al. 2005b; Grigsby et al. 2007; Loxton et al. 2007) have examined the effects of resin flow and its distribution on the properties of wood composites. Bonding performance is determined by the resin distribution and resin coverage of particle surface (Grigsby and Thumm 2012b).

The effect of resin distribution on the bonding performance of wood composites has been extensively studied over the past few decades. Adhesive penetration is identified into two categorizations consisting of gross penetration and cell-wall penetration. Gross penetration occurs when the resin flows into large voids of the porous structure of wood due to hydrodynamic flow and capillary action (Johnson and Kamke 1992a; Kamke and Lee 2007). The mechanism of resin flow movement through the cell lumen was only due to the capillary action of resin at low viscosity, while cell wall penetration only occurs with a resin having a low molecular weight (Buckley et al. 2002; Kamke and Lee 2007). Deeper adhesive penetration could improve the bonding strength and dimensional stability of plywood under various conditions (Li et al. 2020). Resin with low molecular weight can penetrate 1 to 2 cells deep when resin drops were placed on aspen, whereas high molecular weight resin is trapped on lumen surface, which results in no penetration for the high molecular weight resin (Christiansen and Gollob 1985; Stephens and Kutscha 1986; Johnson and Kamke 1992b, 1994; Furuno et al. 2004a; Kamke and Lee 2007).

While resin distribution and penetration are mostly determined by the light microscopy image analysis; however, the spreading of resin must be difficult to detect. Unfortunately, little information is given in the literature about the spreading ability of resin, especially for PF resins with different molecular weights. The resin area coverage generally increased by small resin spots rather than the big spots. Resin spreads mainly in the parallel to wood grain direction, which results in increasing resin area coverage after hot pressing; however, the increase slows down at high resin contents (Dai et al. 2011). Hence, one of the goals of this study is to investigate the resin expansion ability and the expansion mechanism of resin granules from different molecular weight PF resin.

2. Objectives and scope of this thesis

The objective of this study is to evaluate the impact of wood raw material especially from low-density wood species, different types of resin (pMDI, PF) and various board structures on the performance and properties of the lightweight strand boards.

Kiri wood (*Paulownia tomentosa*) is well-known in the Asian area for a long time as the low density wood species but newly planted and studied in Europe. However, the performance of kiri wood and its link to the specific application and various process conditions as raw material for the production of the wood-based panel are not fully studied. This issue might be partly explained by the lack of kiri wood raw material availability in Europe in the market, which now, after several years of kiri plantation project in Germany, has result in new questions to be answered. In addition, kiri has a very low density and might be thus suitable for the production of light solid wood products; however, low wood density leads to a low surface hardness which limits the possible applications of solid wood for furniture or construction. Up to today, there is still a lack of scientific research on the development of new technologies to enable the use of low-density kiri wood for the production of wood-based panels, particularly oriented strand boards.

The investigations of this present study are divided into three parts:

The first part of this thesis (**publication I-IV**) investigates the influences of different wood species such as kiri, pine and beech strands. It focuses on the impact of strand surface area from these three wood species on the properties of lightweight strand boards. A special emphasis is on the total strand area for each wood species calculated by scanning and image analysis. The relation of compaction ratio (between wood material density and panel density) and resin molecular weight were investigated. Such knowledge might enable the wood-based panel producer to obtain expected board properties with the minimum cost of adhesives and wood materials.

The second part of this thesis (**publication II-V-VI**) investigates the problems in thickness swelling of the kiri strand board which arises from the first part. To evaluate these issues, two PF resins with different molecular weight were investigated with regard to: (1) the influence of phenol-formaldehyde of low and high molecular weight on the properties of strand board, (2) effects of resin molecular weight on the resin spreading and penetration and its influence on internal bond strength and dimensional stability of strand boards from kiri wood (*Paulownia tomentosa*), (3) effect of pressing time on properties and formaldehyde emission of strand boards.

The third part of this thesis (**publication III**) investigates a new veneer strand board (VSB) which was developed by combining strands and low-quality veneer on the surface and inside the panel. The low panel density results in low bending strength. This combination provides the possibility to enhance the bending strength of conventional OSB aim to compete with plywood in the construction industry where the demand for the lightweight wood-based panel is growing.

3. List of publications

The thesis is based on the following publications in peer-reviewed journals.

Publication I

Effect of wood and panel density on the properties of lightweight strand boards

Tien Van Pham, Christian Schöpper, Andre Klüppel & Carsten Mai; Wood material science & Engineering (2019), *in press*

Publication II

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

Tien Van Pham, Vladimirs Biziks, Carsten Mai; Wood material science & Engineering (2021), *in press*

Publication III

Performance properties of light-weight strand boards reinforced with veneers and veneer strips

Tien Van Pham, Carsten Mai; (submitted)

Publication IV

Influence of compaction ratio and resin molecular weight on the mechanical and water-related properties of strand boards

Tien Van Pham, Carsten Mai; (submitted)

Publication V

Effect of pressing time on physical-mechanical properties and formaldehyde emission of strand boards

Tien Van Pham, Carsten Mai; (manuscripts in preparation)

Publication VI

Assessment of Phenol-formaldehyde resin distribution on the strand surface by light microscopy

Tien Van Pham, Vladimirs Biziks, Carsten Mai; (submitted)

4. Experimental summary

The detailed description of the experimental and analytical methods in this study can be found in the scientific paper. A summary of experimental and analytical methods was showed in Table 1 below (**I to VI**: scientific papers).

Table 1 Summary of wood-based panel's standards and analytical methods (**I to VI** publications)

EN 300 (2006)	Oriented Strand Boards (OSB) - Definitions, classification and specifications.
EN 310 (1993a)	Wood-based panels - Determination of modulus of elasticity in bending and of bending strength.
EN 317 (1993b)	Particleboards and Fiberboards - Determination of swelling in thickness after immersion in water.
EN 319 (1993c)	Particleboards and Fiberboards - Determination of tensile strength perpendicular to the plane of the board.
EN 323 (1993d)	Wood-based panels – Determination of density.
EN 320 (1993e)	Particleboards and Fiberboards - Determination of resistance to axial withdrawal of screws.
EN 1534 (2010)	Wood and parquet flooring – Determination of resistance to indentation (Brinell) – Test method.
EN 717-2 (1994)	Wood-based panels - Determination of formaldehyde release – Part 2: Formaldehyde release by the gas analysis method.
EN 636 (2015)	Plywood - Specifications.
EN ISO 178 (2013)	Plastics – Determination of flexural properties.
Pull off test	PosiTest-AT-20 (DeFelsko Corporation, New York, USA)
Vertical density profile	X-ray densitometer (DAX, Fagus-Grecon GmbH & Co. KG, Alfeld, Germany.
Light microscopy	Eclipse E600 microscope (Nikon, Japan).
Contact angle	Krüss GS 10 (Krüss GmbH, Hamburg, Germany)
SEM	LEO supra 35 high-resolution filed scanning electron microscopy (Carl Zeiss AG, Germany)
Data analysis	ANOVA and Tukey HSD test (Origin 8.5) ImageJ (image analysis)

5. General discussion and conclusions

This chapter connects the individual discussion in **publication I-VI** with the objectives to go beyond a general summary of the previous chapters in order to provide not only an overview of background information but also several further details.

5.1. Strand dimensions and manufacturing parameters of strand boards

5.1.1. Influence of strand dimensions on the properties of strand boards

Factors include strand thickness, total surface strand area of the mat, resin distribution, pressing temperature, size and thickness of resin spots have effects on resin efficiency inside the wood strand mat (Dai et al. 2011). As the strand dimension decreases, the total surface area of strand mat increases; however, the experimental difficulty of quantitatively determining the surface area of a normal furnish is enormous (Kelly 1977). To quantify the total surface strand area, which could be generated for each wood species, might contribute to the fundamental knowledge for OSB manufacturers. The results of **publication I** clearly demonstrate that the total strand area of low-density wood species results in high amount of bonding contact points between strands at the low-density board, which helps to reduce the weight of the panel but the properties of the panel still meet the standard requirements.

Longer and wider strands might result in higher maximum core temperature, higher core gas pressure and higher core moisture content (Dai et al. 2005c). Transverse and in-plane permeability of the strand mat is considerably dependent on the thickness of the strand (Hood et al. 2005). Strand thickness, which apparently has a considerable influence on the three pressing parameters, is a dominant factor in determining mat permeability compared to strand width and length (Dai et al. 2005c). Thicker strands lead to higher mat permeability, even at low mat density (Hood et al. 2005b). A model, which was developed to simulate behaviors of individual strands in the mat-forming process of OSB, found that increased strand thickness results in decreasing the permeability (Li et al. 2008).

5.1.2. Influence of manufacturing parameters of strand boards

Major manufacturing parameters including wood material, resin, board density, thickness, platen temperature, pressing pressure, pressing time are key factors to determine the board properties, which is a complex process where several parameters interact simultaneously. Wood-based composites are usually produced by consolidating mats of resinated wood furnishes under heat and pressure for a certain duration which allows the resin to cure to form the bonding contact between wood furnishes. The transverse permeability of OSB mats decreased rapidly as the compaction ratio is increased; however, there is no difference in permeability as the compaction ratio reaches higher than 1.3 (Hood et al. 2005). The internal conditions of the mat during hot pressing, which includes complex movement of heat conduction and moisture flow, change rapidly (Zombort et al. 2003). Understanding the wood raw material and the mechanisms of the hot pressing process will help to establish the pressing parameters of wood-based panels (Haselein 1998; Zombort et al. 2003; Dai and Yu 2004b; Dai et al. 2005c; Harless et al. 2007).

Typical hot pressing temperatures of wood-based panels range from 150 to 220°C. The condensation reaction of phenol-formaldehyde adhesives requires the lowest temperature approximately 100°C (Pizzi and Mittal 2018a). The too-high gas temperature of the internal core layer, which accumulates by moisture and heat, can cause the blister after hot pressing (Figure 2). It is apparent that the pressing temperature can explain the reason that led to the blister of the kiri board whereby the board surface gets slightly burn.

Figure 2 Blisters of kiri strand board after hot pressing and burned board surface

Wood-based composites compressed under conditions of high temperature and low moisture content. The compressive stresses are imparted to the individual wood particles leading to damaged wood cell on the particle surface and strength losses during hot pressing (Hse et al. 1973; Geimer and Loehnertz 1985). In contrast, pressing at higher moisture content will not be a simple task as the high internal-steam gas rises associated with excessive adhesive migration from the glue-line (Christiansen et al. 1993), although the existence of capillary water associated with residual liquid water may contribute to heat transfer inside the mat (Pichelin et al. 2001). Moreover, internal-gas steam can easily escape when the size of the panel is small (Pichelin et al. 2002).

During the stage of hot pressing, the heat is transferred by conduction as a result of the direct contact between the hot-platens and the wood particles (Hood et al. 2005). The transverse and in-plane permeability are reduced as the mat is consolidated to the board target thickness, which leads to the build-up of internal gas pressure (Fakhri et al. 2007). The compaction ratio between board density and wood density is considered as one of the most important factors for controlling the transverse and in-plane permeability (Kelly 1977; Dai et al. 2005b, a). Low-density kiri strands might cause low in-plane permeability of the strand mat at high board density. Therefore, the blister of kiri strand boards occurs when the low permeability of the mat is associated with high pressing temperature and pressing time. The pressing parameter of strand boards producing with pMDI and PF were summarized in Tables 2 and 3.

Table 2 Overall pressing parameters of strand boards manufacturing with pMDI adhesive in this thesis

Panel dimensions	450 × 450 × 18 mm^3
Pane densities	300 kg m^{-3}; 400 kg m^{-3}
Strand fractions	1:1:2 (10-30 mm:30-50 mm:>50 mm)
Wood species	Kiri (*Paulownia tomentosa*)
	Pine (*Pinus sylvestris*)
	Beech (*Fagus sylvatica* L.)
Strand moisture content	3-5%
Target mat moisture content	8-10%
Resin type	pMDI I-Bond PB PM 4350
Resin mass content	5%
Blender rotation speed	30 rounds min^{-1}
Press temperature	200°C
Press time	15 s mm^{-1}

Table 3 Overall pressing parameters of strand board manufacturing with phenol-formaldehyde adhesive in this thesis

Panel dimensions	450 × 450 × 12 mm^3
Pane densities	400 kg m^{-3}; 500 kg m^{-3}
Strand fractions	4:5:1 (10-30 mm:30-50 mm:>50 mm)
Wood species	Kiri (*Paulownia tomentosa*)
	Pine (*Pinus sylvestris*)
	Beech (*Fagus sylvatica* L.)
Strand moisture content	3-5%
Target mat moisture content	8-10%
Resin type	LMW (M_w=400-500 g mol^{-1}; solid content 53%)
	HMW (M_w=1000-1200 g mol^{-1}; solid content 70%)
Resin mass content	10%; 20%
Blender rotation speed	30 rounds min^{-1}
Press temperature	140°C
Press time	90 s mm^{-1}; 135 s mm^{-1}; 180 s mm^{-1}

5.1.3. Reduction of the density of strand boards

The typical density of oriented strand board ranges from 600 to 650 kg m^{-3}, which is considered higher than that of typical plywood in the range from 450 to 520 kg m^{-3}. As a result, the weight of OSB panels is significantly higher, which could cause various problems when used in construction (Go and Cacchione 2003). Studying the effect of wood material density on the resulting strand board properties requires a suitable basis for comparison. Keeping all manufacturing process parameters constant and only changing the board density might be the simplest approach. However, it is generally known that reducing the target board density lowers the mechanical properties of the board (Go and Cacchione 2003; Chen and Wellwood 2010; Mirski and Dziurka 2015; Shalbafan et al. 2016b; Monteiro et al. 2016). However, when reduced board density, the resin efficiency decreases which is attributed to less bonding efficiency between particles, not to resin distribution (Kelly 1977).

5.2. Improving the properties of lightweight strand boards

The results of **publication I** apparently display that the total strand area of low-density wood species leads to more strand area and the compression causes more bonding contact points between strands at the low-density board, which enhances the mechanical properties of the board. During hot pressing as the strand mat is consolidated, compressive stresses are imparted to the individual wood strands leading to compressed wood cells on the strand surface (Hse et al. 1973; Geimer and Loehnertz 1985). A higher amount of compressed wood cells could be a factor particularly attribute to the higher spring-back effect. Also, it should be noted that pMDI adhesive is known as a water-resistance adhesive in the OSB production industry. Therefore, the author assumed that those compressed wood cells on the strand surface are without adhesive, which easily springs back when exposed to moisture conditions. That is the reason to use an adhesive that could have a better penetration to bond the compressed wood cell walls on the strand surface particularly in the case of pMDI. The investigations performed in **publications II and VI** provide explanations about why the performance of kiri strand boards containing LMW PF is better than those containing HMW PF in some of the physical and mechanical properties. These publications study the effect of resin molecular weight on the bonding performance between kiri strands.

5.2.1. Assessment of Phenol-Formaldehyde resin distribution

Adhesive distribution is one of the most important factors to influence the bonding performance of wood-based panels. As can be seen in Figure 3, the resin was equally distributed by spraying over the strand surface. Resin efficiency is primarily affected by resin distribution, resin coverage and board density (Lehmann 1970; Vital et al. 1974). In addition to the adhesion strength, the penetration of LMW PF resin into the wood strand was observed in **publication VI** and its effects on the properties of kiri strand boards displayed in **publication II**. In general, the penetration depth of the phenolic resin into solid wood can be observed by light microscopy (Furuno et al. 2004a; Biziks et al. 2019). However, the wood strand with uneven surface seems to be difficult to be observed by light microscopy.

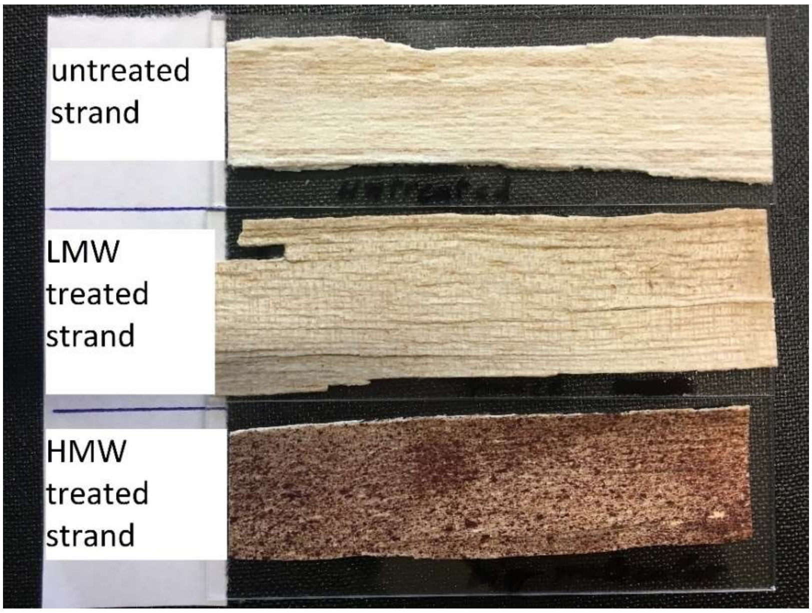

Figure 3 Image of blended kiri woos strands with LMW and HMW PF

Cell wall matrix stiffening (pull-off strength)

Pull-off tests were conducted with two molecular weight PF resins. The cell wall matrix stiffening effect caused by the penetration of LMW PF resin during hot pressing reduces the swelling of compressed cell material. Also, the increase in IB of strand boards might be attributed to the greater gluing area. Resin penetration into the cell wall matrix can play some roles in the strength of adhesive bonds; however, little is known about the method to determine that role (Frihart 2006a). The penetration ability of resin into the cell wall matrix and other voids is significantly dependent on the resin molecular weight (Shams and Yano 2011). LMW resin could easily penetrate cell walls resulting in softening of the wood cell which thus induces less damaging the wood cell under pressing pressure, while HMW PF resin does not penetrate into the walls (Knop and Pilato 1985; Gardziella et al. 2000; Furuno et al. 2004; Frihart 2006b; Kamke and Lee 2007).

Bond formation

The bonding performance of adhesive, which takes place in the interphase region between wood elements, is considerably affected by the degree of adhesive penetration into the porous network of interconnected cells (Ülker 2016). The process of bond formation involves several steps from macroscopic wetting to polymerization under the heat of hot pressing (Frihart 2006). The flow of resin, which was related to resin molecular size, increases with decreasing resin molecular weight (Frihart 2006). A mixture of resin containing 20% LMW PF and 80% HMW PF caused the highest IB and dimensional stability of OSB (Knop and Pilato 1985a;

Gardziella et al. 2000). The penetration behavior of resin into a porous wood structure is influenced by various factors such as wood substrate species, amount of resin expansion, pressing parameters (Pizzi and Mittal 2018). Larger molecules of phenolic resin and a high viscosity might limit the speed of resin penetration (Pizzi 1983). As can be seen in Figure 4 with the average grey value per slice, the resin of HMW PF was mostly distributed by spraying over the strand surface 0.085 mm, while LMW PF resin has a similar grey value as control. This effect might show that HMW PF resin granules mainly were trapped on the wood lumens, which witnesses no penetration into the wood cell wall. Similar results have been reported by (Laborie 2002; Furuno et al. 2004b; Kajita et al. 2004; Wan and Kim 2007; Shams and Yano 2011).

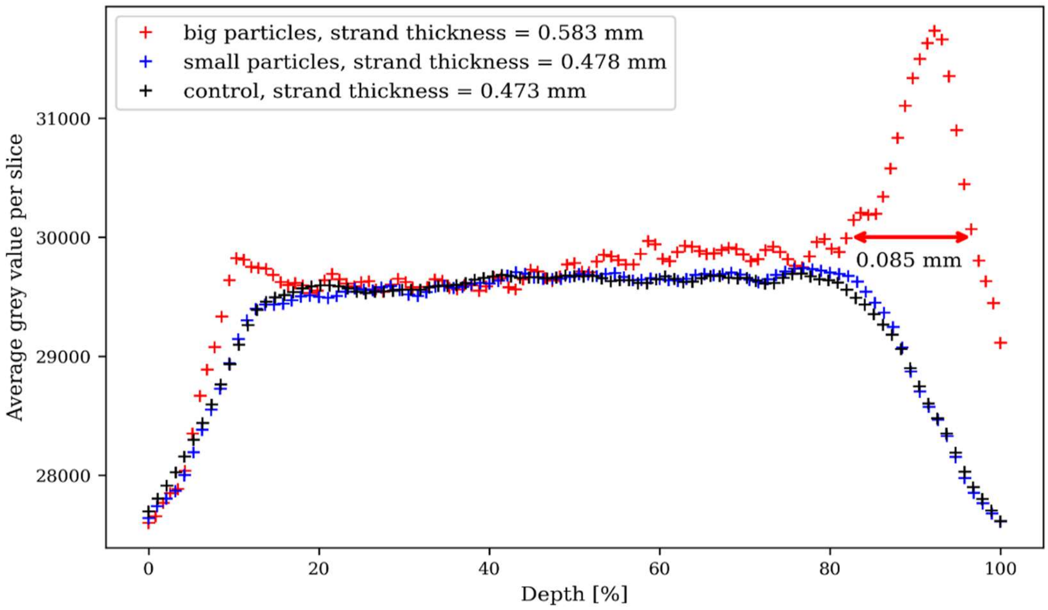

Figure 4 Average grey value per slice of kiri woos strands; control (black); LMW PF (blue); HMW PF (red)

5.2.2. Effects of resin molecular weight spreading on adhesion quality

An important new aspect of resin properties found in this study is resin expansion to improve the bonding quality by increasing the resin area coverage. As shown in **publication VI**, resin expansion of LMW PF might display a great improvement for the internal bonding strength and dimensional stability. The investigation of resin granules expansion was limited because there are not any visible differences between the untreated strand and LMW treated strand on the kiri strand surface as a substrate. White filter paper is considered as an alternative cellulosic substrate, which allows us to visualize the LMW resin. Besides the positive impact of LMW PF on the properties of strand boards, LMW resin does not change the original color of the wood strand. The gap between resin granules can be also clearly observed, which causes non-continuously resin glue lines and may result in decreasing the bonding quality in wood-based panels.

The movement of resin drops has been recorded by two stages during adhesive application. Water-proof papers and filter papers were set up to record those stages. While water-proof papers record the original shape of PF resin granules as it hits the substrate, the filter paper will display the spreading ability of resin. LMW PF resin droplets were enlarged to fill the gap between two adjacent resin droplets significantly increasing the resin area coverage, while HMW PF resin droplets were maintained as its initial shapes when it hits the substrate.

The increase in the IB of strand boards containing LMW PF might be linked to the expansion ability of low resin molecules as the resin area coverage and bonding quality are strongly correlated to the tensile strength. The penetration behavior of resin into the porous wood structure is affected by various factors such as wood substrate species, amount of resin expansion, pressing parameters (Pizzi and Mittal 2018). It is thus concluded that the spreading ability of resin, which can be observed in Figure 6, was also related to the penetration of resin.

Resin viscosity, which is an important variable, influences the wetting and flow of resin into the wood cell wall (Frihart 2006). The flow of resin was obviously controlled by factors such as resin molecular size, shape and viscosity (Knop and Pilato 1985; Gardziella et al. 2000; Frihart 2006). Therefore, lower viscosity and low molecular weight resin can be easily spreading out on the substrate surface (as can be observed in Figure 6 a, b), which significantly influences the resin coverage area. Such spreading effects of LMW PF might be assigned to the higher IB properties of kiri strand boards but lower IB for spruce and beech strand boards.

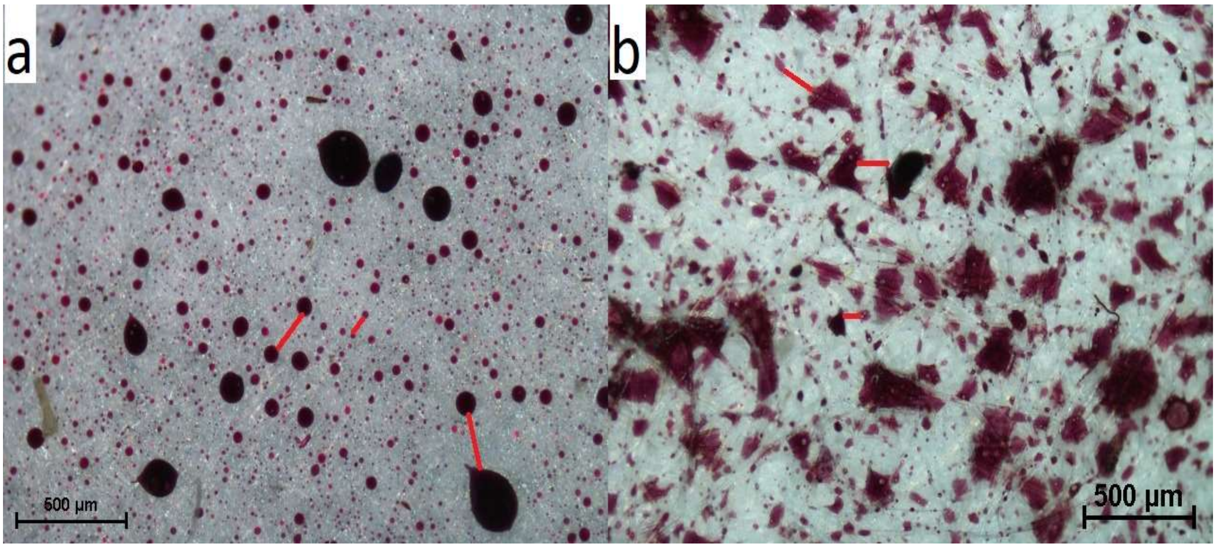

Figure 5 Expansion of HMW PF resin granules; (a) initial size of HMW resin granule on the surface of the water-proof paper (b) expanded the size of HMW resin granule on the surface of filter paper (red line is the gap between adjacent resin granules)

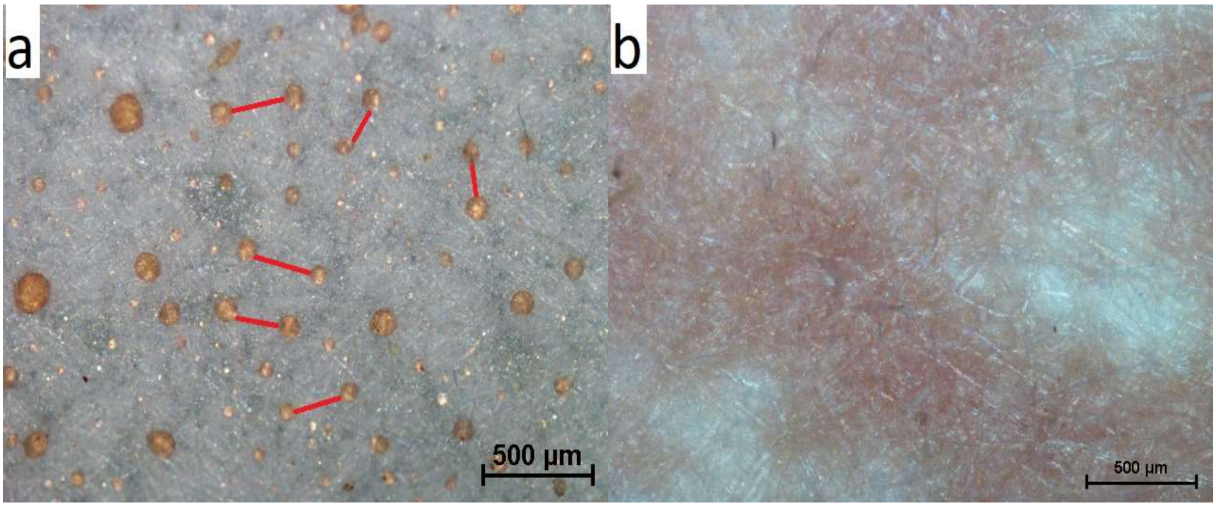

Figure 6 Expansion of LMW PF resin granules; (a) initial size of LMW resin granule on the surface of the water-proof paper (b) expanded the size of LMW resin granule on the surface of filter paper (red line is the gap between adjacent resin granules)

5.3. Influence of compaction ratio on the adhesion bonding quality

As can be seen in Figure 7, the cross-section of the kiri wood strand contains big vessels and approximately 22 wood cells through the strand thickness. A fundamental understanding of the porous matrix structure of the wood substrate is needed to perceive physical changes in wood structure as well as different wood species. Characterization of bond lines may assist in the design of a wood-based panel manufacturing system.

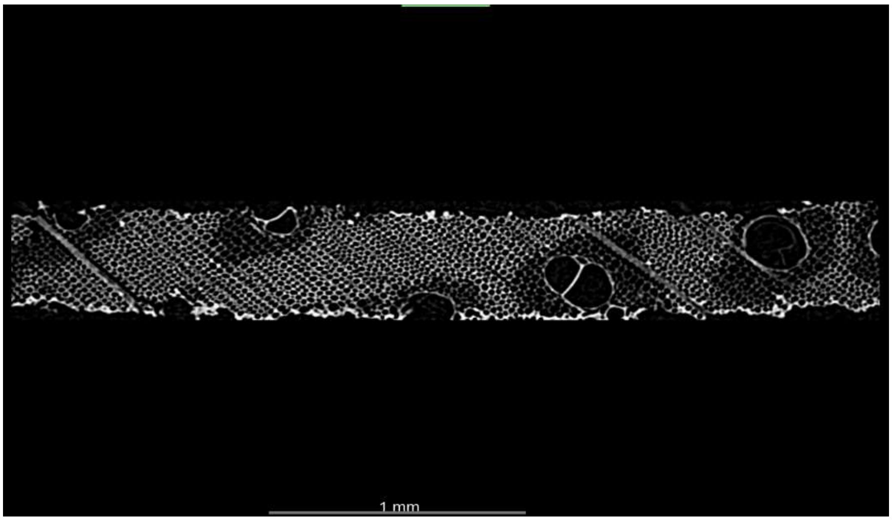

Figure 7 X-ray tomography image of a cross-section of kiri wood strand

Penetration behavior of resin into the porous wood structure is affected by various factors such as wood substrate species, resin expansion, pressing parameters (Pizzi and Mittal 2018). Particularly, the penetration ability of resins into the wood, which is highly dependent on the resin molecular weight, contributes significantly to compressed dimensional stable resin-impregnated wood (Shams and Yano 2011). LMW resin could easily penetrate cell walls, resulting in softening of the cell wall which thus induces less damaging the wood cell wall under pressing pressure (Furuno et al. 2004a). Nevertheless, over-penetration, which causes starved bonding lines between particles, can easily occur with LMW resin (Kamke and Lee 2007; Pizzi and Mittal 2018b). As the deformation occurred on the wood strand surface, a significant increment of density might be observed for LMW PF resin compared to HMW PF resin (Shams and Yano 2004).

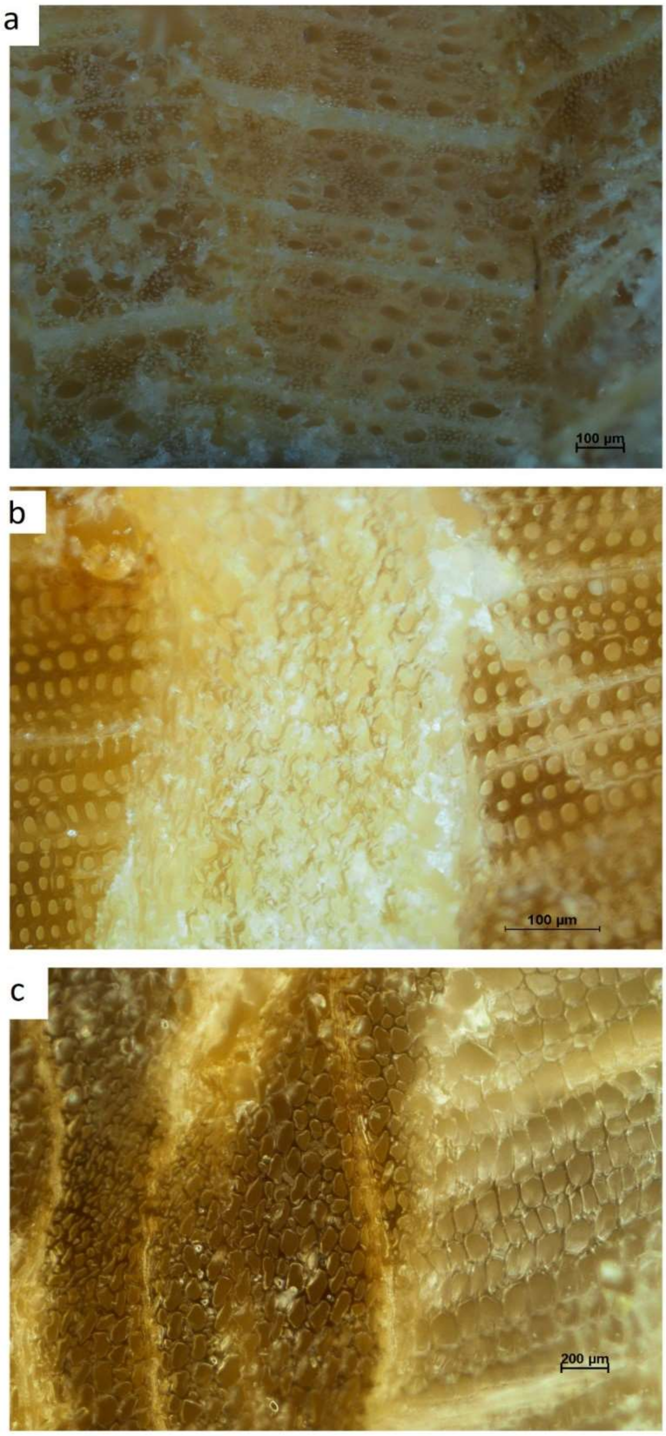

Figure 8 Light microscopy images of strand boards; (a) beech; (b) pine; (c) kiri

The effect of resin molecular weight as well as the structure of wood strand substrate are needed to examine the bonding performance. The adhesive bond lines of OSB thin cross-sectional slices, which were produced from three wood species including beech, pine and kiri, can be obtained by using a microtome (Figure 8 a,b,c). A closer view of the compressed kiri wood cell between strands is more easily observed, but it is not clear because of the uneven surface (Figure 8c). SEM might be a better technique to obtain a clear observation of the glue line between strands, which was successful used for detecting resin granules in a single strand in the scope of this study. During hot pressing as the kiri strand mat is consolidated, compressive stresses are imparted to the individual wood strands leading to deformed wood cells on the kiri strand surface. The deformation behavior of wood is mainly dependent on wood species as well as the anatomical features of species (Shams et al. 2006). Kiri strand, which exhibit a very low compression strength, could be easily deformed under pressing pressure. The deformed cells of kiri wood can be observed in Figure 8c where bond lines between kiri strands were recorded by light microscopy. Adhesives bonding lines of beech and pine strands exhibited in Figure 8a, b in which no deformation of wood cell in the interphase region was observed. It is thus concluded that deformation of wood strands might be considered as one indicator of bonding performance for strand boards, which is caused by the compression.

Compaction ratio (board density/wood density) has a linear relationship with the mechanical properties of panels up to 1.5 maximum (Vital et al. 1974; Hse et al. 1975). However, a reduction in IB at the same board density was reported as the compaction ratio increased from 1.2 to 1.5, which was attributed to the high amount of damaged flakes at a high compaction ratio (Vital et al. 1974). Such damaged flakes at a high compaction ratio might be assigned to the higher compression. Hence, the compaction ratio alone is not the only factor affecting the bonding performance. Pressing parameters, adhesive content and many other processing factors are very important (Kelly 1977).

5.4. Effects of pressing time on the properties of strand boards

Phenolic resins, which contains oligomeric and polymeric chains as well as monomeric methylol-phenol, free formaldehyde and unreacted phenol, can initiate the curing process by heat with the transformation of molecules of various sizes via chain lengthening, branching and cross-linking to a three-dimensional network (Pizzi 1983; Knop and Pilato 1985a; Gardziella et al. 2000; Pizzi and Mittal 2018a). 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).

As discussed above, the penetration ability of resin into the cell wall matrix is significantly dependent on the resin molecular weight (Shams and Yano 2011). LMW resin can easily penetrate cell walls resulting in softening of the cell, while HMW PF resin is blocked up in the walls (Knop and Pilato 1985; Gardziella et al. 2000; Furuno et al. 2004; Frihart 2006b; Kamke and Lee 2007). This deep penetration of LMW PF might not explain the higher free formaldehyde in the board containing LMW PF. HMW PF remains on the lumen surface, so that formaldehyde easily evaporates during hot pressing resulting in low free formaldehyde content. In contrast, LMW PF resin penetrates deeply into the wood cell wall and reacts with phenol, which is more difficult to condense during hot pressing than HMW PF resin. Moreover, larger bonding coverage of LMW PF must be also taken into account resulting in better bonding between strands, which might block the pathway to release free formaldehyde.

5.5. Assessment of phenol-formaldehyde resin droplets on wood strand surface

Many methods of direct observation have been carried out in this study such as light microscopy, fluorescence microscopy and electron microscopy. However, it is difficult to generate the strand specimen for those methods because kiri strands were thin and soft, especially after it was blended and dried the strands become brittle. Furthermore, the strand surface is uneven, which causes a lot of blurry points on images of light microscopy. Scanning electron microscopy has been used in this study to observe the appearance of strand surfaces in order to understand how the resin granules look. The resin solution was evenly distributed by spraying. The kiri strand surface has many cells that are split open for better access to the binder (Figure 10a, b).

X-ray computed tomography images

An attempt has been made to visualize the phenolic resin granules in the porous matrix structure of the wood strand by x-ray tomography. The methods involve using a contrast agent that includes an x-ray active cationic label in the manufacture of the composite product. The phenolic resin might easily absorb the X-ray radiation which increases the contrast level between wood structure and phenolic resin. However, as can be seen in Figure 9, there is no clear difference between a (control) and b (blended strand with LMW PF), while HMW PF resin granules were easily visualized (Figure 9 c). This effect can be attributed to the fact that used 4% of 4-Iodophenol in phenolic resin solution was too less in order to produce sufficient contrast for x-ray radiation. Therefore, it can be concluded that x-ray tomography did not indicate the difference between HMW PF and LMW PF treated strands.

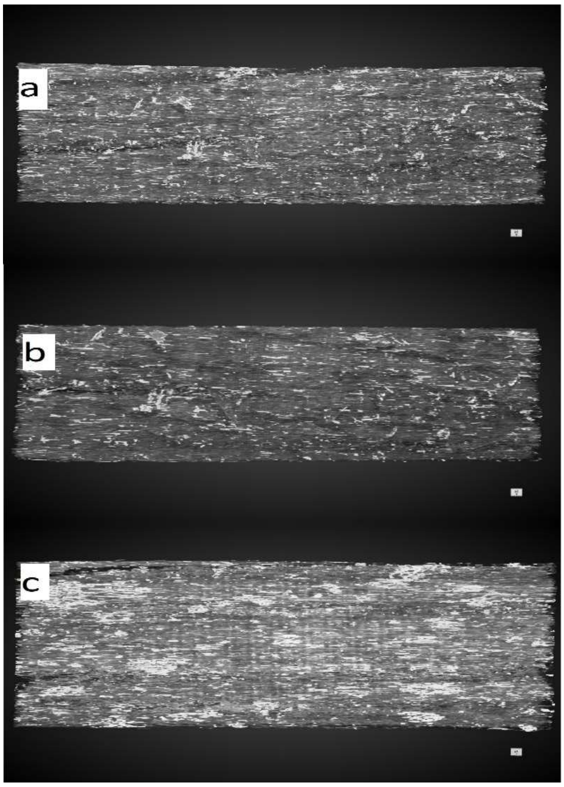

Figure 9 X-ray tomography images of kiri wood strands; (a) control; (b) LMW PF and (c) HMW PF

Scanning electronic microscopy (SEM)

Phenolic resins contains oligomeric and polymeric chains as well as monomeric methylol-phenol, free formaldehyde and unreacted phenol, (Pizzi 1983; Knop and Pilato 1985a; Gardziella et al. 2000; Pizzi and Mittal 2018a). Sodium hydroxide (NaOH) is preferably known as an activating catalyst for the phenol-formaldehyde adhesive manufacture (Pizzi 1983; Knop and Pilato 1985a; Gardziella et al. 2000; Chiu et al. 2003). Sodium hydroxide does not form the crystal molecules but reacts to phenol ions, which was clearly observed inside the HMW PF resin granules (Figure 10 f).

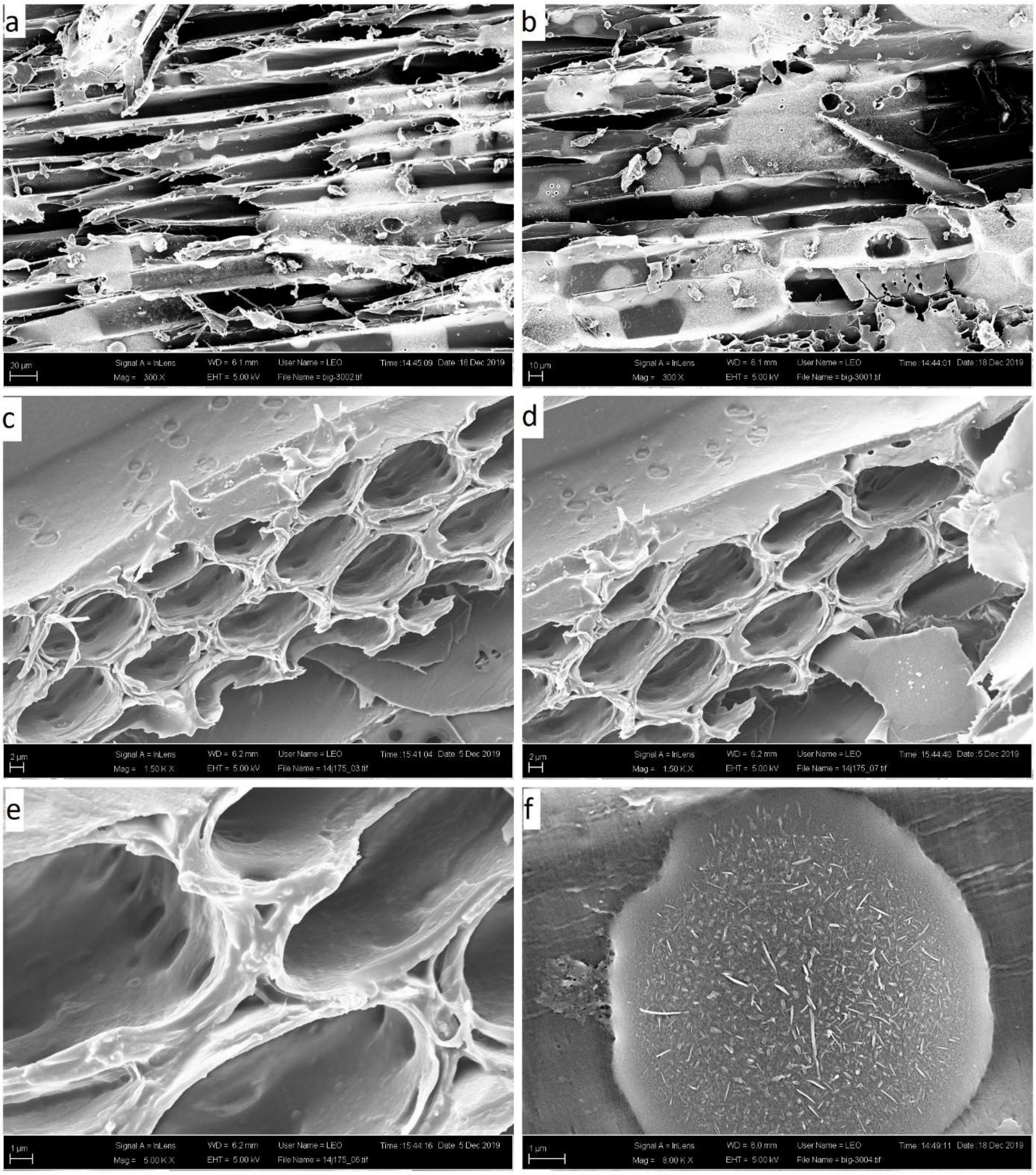

Figure 10 SEM images of kiri wood strands with HMW PF resin granules; scale bar (a) 20µm; (b) 10 µm; (c,d) 2 µm; (e,f) 1µm

The spray process is a widely used method to apply the adhesive on the "woody elements" such as wood fibers, particles or strands for the manufacture of a range of wood-based panel products, fiber or particle, or orientated strand boards (OSB). During the spraying process, the adhesive is pushed through the nozzle by using overpressure, and different sizes of droplets are produced. The size (volume) and distribution of the resin droplets sprayed out depend on applied pressure during spraying, as well as from spray air cap and material control nozzle. Principally during blending between the resin and kiri strands, the inelastic collision (as a mass of resin droplet is in smaller as a mass of strand) of resin droplet with the strand take part, which occurs in a few milliseconds. Three interaction stages of the rein on the surface of the strand during a collision can be observed; the first stage is the contact between droplet and strand, the second stage is the rapid expansion of droplet outwards along the plane, in the third stage resin continue to expand. The first stage is the dominant phase, for which the viscous forces have little influence, the next followed the viscosity dominant phase and gravity forces (Charalampous and Hardalupas 2017). Hence, the penetration depth, film thickness, shape, and size distribution on the surface of the strands depend on impact forces created during the spraying, viscosity of the resin and surface energy of wood are considered as a significant factor influencing the properties of OSB.

Although the color of the wood strand treated with LMW PF resin was slightly changed, control and treated strands did not show any visual color difference after curing at temperature 120°C (Figure 3). Adhesive interphase is critical to bonding performance, so there is a need for methods that can visualize the phenolic resin granules (Laborie 2002). The wood adhesive interface region can be observed on the micro-level within the cell wall without interfering specimens by SEM. SEM could detect the resin granules of LMW PF that did not significantly alter the appearance of strand surface from the macroscopic scale. The presence of LMW phenolic resin might be observed on the cell wall surface in the shapes of granules, which were equally distributed through the entire wall (Figure 11 a, b, c and e). In addition, round minute resin granules were found on the lumen wall (Figure 11 d, f), which can be linked to the penetration of LMW resin into cell walls. Such penetrated resin granules were formed as a result of releasing the vapor during hot pressing or drying in an oven.

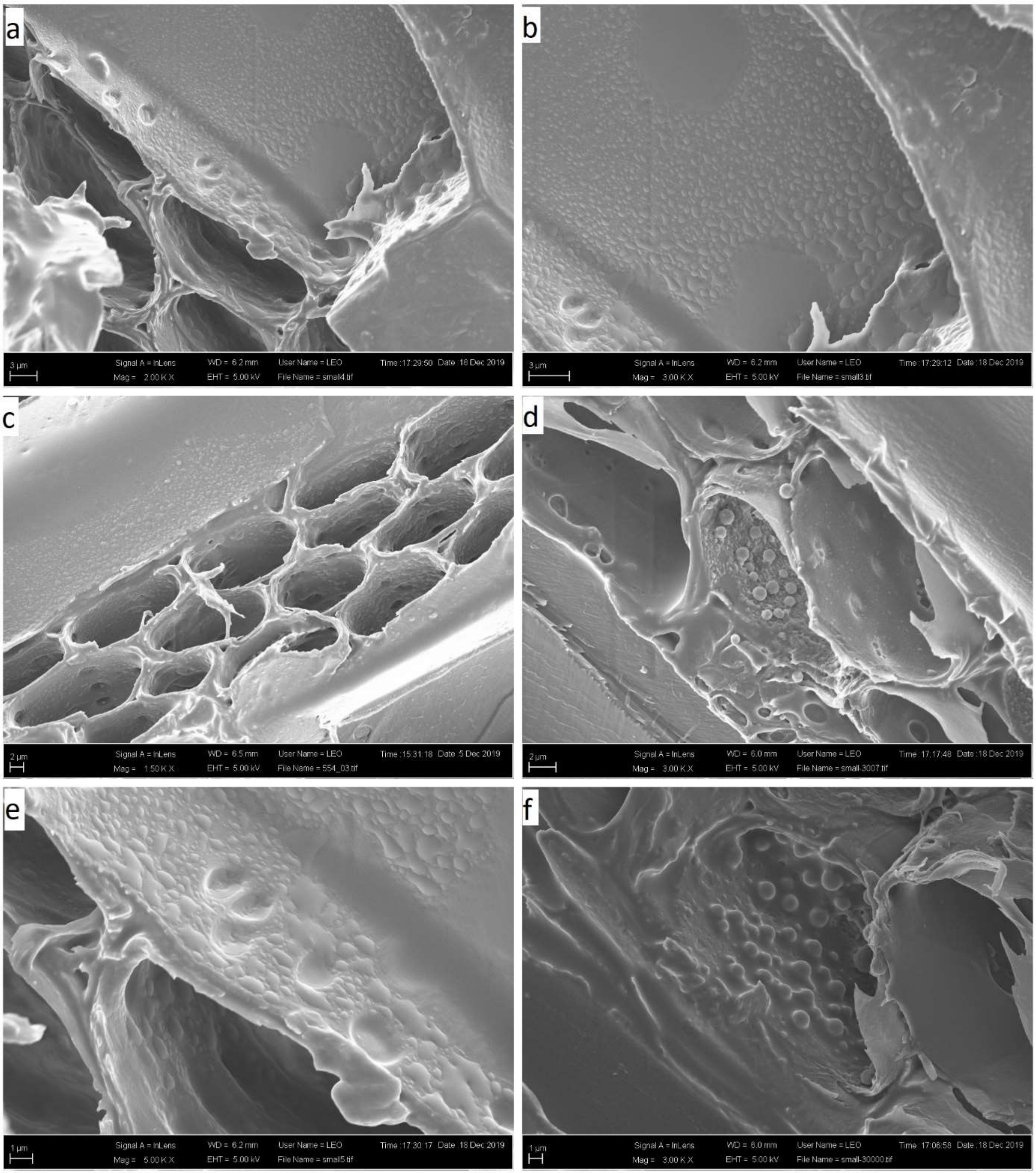

Figure 11 SEM images of kiri wood strands with LMW PF resin granules; scale bar (a,b) 3µm; (c,d) 2µm; (e,f) 1µm

5.6. Performance of strand boards reinforced with veneer and veneer strips

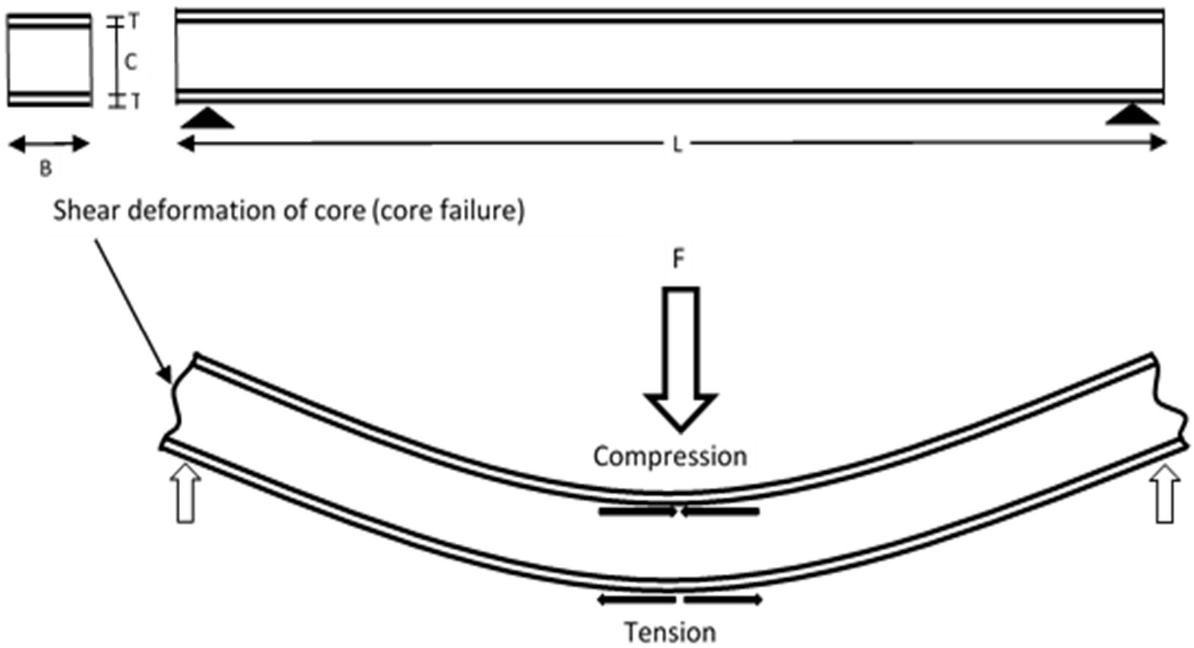

Figure 12 Compression and tension load of the panel under the applied force

This study aimed to improve the bending strength of light-weight strand boards (SBs). In this study, the results cannot be compared to standard requirements for oriented strand boards, because board properties are strongly influenced by strand orientation. Producing SBs with the same target density, but using veneer materials of different densities consequently results in the enhancement of bending properties. Veneer strand boards with kiri and birch veneers at a target density of 500 kg m^{-3} can be produced under laboratory conditions without technical problems. The arrangement of kiri veneers led to a 45% and 39% increase whereas birch veneers imparted gains of 80% and 77% increase in MOR and MOE respectively. Alignment between veneers strands might have a great potential to improve the bending strength of SBs. The results showed that light-weight VSBs (Veneer strand boards) could be a promising new innovative product that might even compete with plywood in the light-weight construction industry. We conclude that the new VSB is feasibly applicable in the wood-based panels industry to improve the mechanical properties of boards by the arrangement of veneers. The properties of VSBs still could most likely be improved by applying resin on veneers and orientating strands before hot-pressing.

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7. Appendix

Publication I

Effect of wood and panel density on the properties of lightweight strand boards

Publication II

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

Publication III

Performance properties of light-weight strand boards reinforced with veneers and veneer strips

Publication IV

Influence of compaction ratio and resin molecular weight on the mechanical and water-related properties of strand boards

Publication V

Effect of pressing time on physical-mechanical properties and formaldehyde emission of strand boards

Publication VI

Assessment of phenol-formaldehyde resin distribution on the strand surface by light microscopy

Publication I

Effect of wood and panel density on the properties of light weight strand boards

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Effect of wood and panel density on the properties of lightweight strand boards

Abstract

The objective of this study is to evaluate the effect of wood and panel density on the properties of lightweight strand boards. For this purpose, we compared lightweight SBs made of low-density kiri wood (*Paulownia tomentosa*), medium-density pine wood (*Pinus sylvestris*) and high-density beech wood (*Fagus sylvatica*). Single-layer non-oriented SBs were manufactured with target densities of 300 kg m^{-3} and 400 kg m^{-3}. The cohesion of beech boards was insufficient for further testing. The larger compaction ratio of kiri boards resulted in a steep density profile and mechanical properties, namely modulus of rupture (MOR), modulus of elasticity (MOE) and internal bond (IB), superior to those of pine boards. However, Kiri panels exhibited a higher thickness swelling (TS). At 400 kg m^{-3}, IB and TS of kiri boards met the requirements for OSB/1 according to the European standard EN 300. MOR of kiri boards was 2.6 and 1.6 times higher than that of pine boards at 300 kg m^{-3} and 400 kg m^{-3}, respectively, while these ratios were only 1.8 and 1.2 for MOE. We conclude that the effect of wood density on bending properties reduces as panel density increases and that the compaction ratio affects MOR more than MOE, which seems to be mainly determined by panel density. Hence, utilization of kiri wood allows the reduction of board density with MOE being the limiting factor.

Keywords: bulk density; compaction ratio; kiri wood; strand board

Introduction

Oriented strand boards (OSB) are wood-based panels designed as a cheaper replacement for plywood. Their structure optimizes the mechanical properties of boards that can be manufactured from low quality (small diameter) logs. The wood elements, strands, are approximately 15…25 mm wide, 75…150 mm long and 0.3…0.7 mm thick. The high aspect ratio (strand length divided by thickness) and horizontal alignment in the flat-pressed boards result in large contact areas between overlapping strands on which adhesive bonds can form. Due to their relatively large size, the surface area of the strands is smaller than the area of elements in particleboards, so that a lower adhesive content is needed for the same amount of adhesive per wood surface area. Additionally, the orientation of the strands in the surface layers increases strength in the respective direction. For these reasons, the mechanical properties of OSB are superior to most other wood-based panel products for a given adhesive content and board density (Dinwoodie 2000, p. 232 ff.). Accordingly, OSB is mainly used for load-bearing applications such as sub-flooring, roof sheathing, wall and ceiling elements as well as for packaging (EPF 2017). These applications require a high strength-to-weight ratio (i.e. specific strength) and, thus, further increasing the specific strength would add value to the product. This is especially true for applications in the transport sector where lightweight allows fuel-saving and easier handling.

In general, there is no proper definition of the term ‘lightweight board’. However, it can be stated that lightweight boards exhibit densities of less than 500 kg m^{-3} (Poppensieker and Thömen 2005). The density of wood-based boards can be reduced in various ways: by using low-density wood species, by densifying the material to a lower degree, by adding light non-wood materials or by employing a sandwich structure with a low-density core (Nilsson et al. 2013). For this low-density core various types of lightweight materials are used such as an extremely low-density wood, hollow materials, honeycomb paper, polystyrene foam, polyurethane foam, polycarbonate, polypropylene, ultra-lightweight foam, and wood fiber-based, maize granular (Burnett and Kharazipour, 2018; Jivkov et al., 2012; Karlsson and Tomas\AAström, 1997; Shalbafan et al., 2013). The disadvantage of combining wood-based boards with other materials is that these materials are often non-renewable and that mixtures are usually more difficult to recycle. Additionally, sandwich boards allow little flexibility during the manufacture of furniture, because reinforcements are required at the board edges and for joints and fittings.

The utilization of low-density wood species and reduced densification is the other possible approaches to manufacture lightweight boards with acceptable strength properties. The good bonding between the wood particles that make up the board requires a minimum compaction ratio (ratio of board density to wood density). For a given board density, therefore, strength properties are generally expected to increase as the density of the wood raw material decreases (Klauditz 1952). The compaction ratio of particleboards is typically around 150% (Irle and Barbu 2010, p.7). Zhou (1990) suggested a density of 650 kg m^{-3} for OSB from hybrid poplar (density ca. 400 kg m^{-3}), which corresponds to a similar compaction ratio. However, the industry also produces particleboards and OSB with a distinctly lower compaction ratio. Chen *et al.* (2010) found that strength properties of aspen OSB increase with board density but this increase gradually lowered as density continued to rise. Properties acceptable for many applications, however, may be achievable at much lower densification ratios (< 100%), if high-quality strands are used (Mirski and Dziurka 2015).

The objective of this study is to evaluate the possibility of using low-value and low-density *Paulownia* wood to produce light-weight OSB. This includes investigating the effect of low compaction ratios. Randomly-oriented strand boards (SB) were used as a model for OSBs to assess the effect of wood density on the properties of light-weight boards. To investigate the effect of wood density on the production of OSB, kiri SBs were compared with SBs made of the medium-density species pine (*Pinus sylvestris*) and the high-density species beech (*Fagus sylvatica*).

Material and methods

Strand production

Strands were produced from kiri, pine and beech logs, with the diameter of the kiri logs ranging from 12 to 20 cm and that of the beech and pine logs ranging from 30 to 35 cm. All logs were 2 m long and were manually debarked before stranding. The strands were manufactured by a knife ring flaker PZUL 8-300 (Pallman Company, Zweibrücken, Germany) at Fraunhofer WKI (Braunschweig, Germany) with a target length of 110 mm, a target thickness of 0.5-1 mm and a target width of 10-50 mm. A single wood stem was firmly held by a hydraulic clamp and moved towards the knife ring. For pine and beech, the feeding speed was 40 mm s^{-1}. For *Paulownia*, it was reduced to 36 mm s^{-1}, because faster feeding resulted in a high amount of damaged strands. All strands were directly sorted by a sieving shaker into 3 fractions: 10-30 mm, 30-50 mm and bigger than 50 mm. Sieve fractions smaller than 10 mm were discarded as fines. To avoid mould growth, the strands were immediately dried at 70 °C to a moisture content of 3% to 5% in a drying oven. Then, strands were stored in plastic bags to keep the moisture content low. For panel manufacturing, the strand fractions were mixed in a ratio of 1:1:2 (10-30 mm:30-50 mm:> 50 mm).

Strand characterization

For strand characterization, 100 g of each strand fraction were randomly selected. The strands were placed on a flatbed scanner (Expression 11000XL, Epson, Suwa, Japan) and carefully separated to make sure that they did not overlap. For every fraction, several images were recorded, because the scanning area was too small for all strands. A white reference paper of 50 × 50 mm^2 was scanned with every image. To create a dark background with high contrast to the strands, the scanner was not closed but it was covered with a carton. Images were processed with the program Image J (Rueden *et al.* 2017): Greyscale images were converted to binary ones by automatic thresholding so that the images were segmented into strands and background

(Figure 1). The scale of the image was set based on the reference paper. The area and minimum caliper diameter (Feret's diameter) of each strand were recorded using the software's integrated analysis functions. In this study, the minimum Feret diameter served as a measure for the strand width. Assuming a perfectly rectangular shape, strand length was calculated on the basis of area (A) and width (W). In addition, the thickness of 20 randomly chosen strands from each fraction was measured manually with a dial gauge.

To calculate strand density, mass and volume of 20 strands were determined. The volume was determined by the submersion method. Each strand was attached to a sample holder and immersed in a beaker filled with water without contacting the inner walls or the bottom of the beaker. Before submersion, the beaker was placed on a scale and the scale was tared to the weight of the beaker (including water) with the immersed sample holder. The weight shown by the scale after immersion of the strand equals the mass of water displaced by the strand. As the density of water is ca. 1 g cm^{-3}, the value is equivalent to the volume of the strand in cm^3.

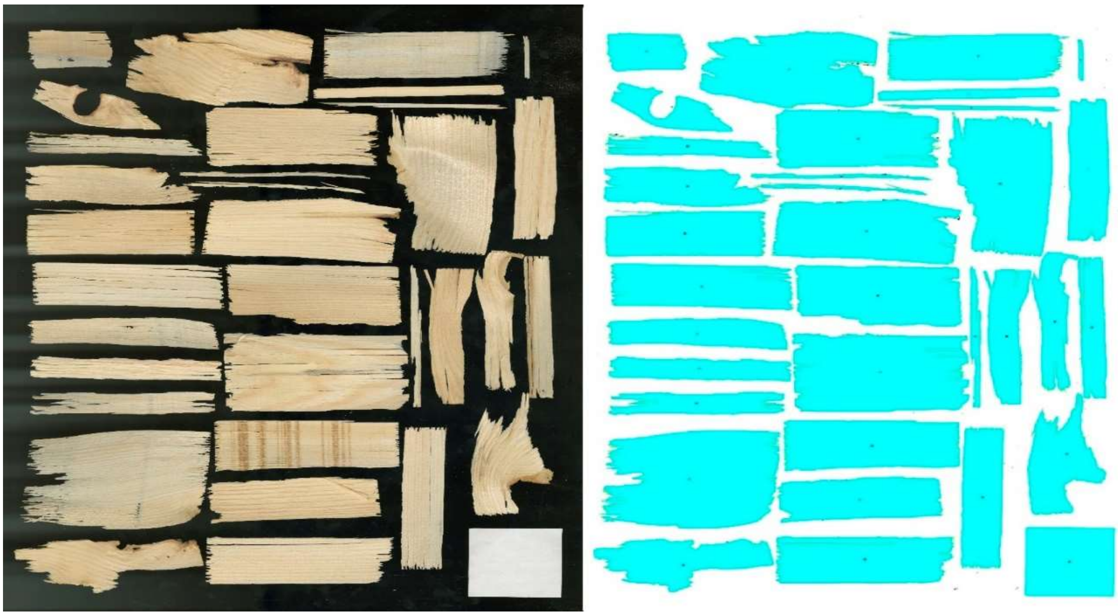

Figure 1 Example for the scanned image before (left) and after (right) segmentation

Determination of mat density

Mat density was determined by using unresinated strands. The mats were formed in the same way as for panel manufacturing using the same mixture of strand fractions and single-strand fractions. The number of strands used to produce the mat was the same as that needed to produce a board with a density of 300 kg m^{-3}. The height of the mat was recorded for volume calculation (3 times for each fraction and species).

OSB panels manufacturing and testing

The strand fractions were mixed in a ratio of 1:1:2 (10-30 mm:30-50 mm:> 50 mm) and directly blended with 5% (m/m based on oven-dry wood) pMDI (polymeric methylene diphenyl diisocyanate, I-Bond PB PM 4350, Huntsman, Everberg, Belgium) in a rotating drum blender at a speed of 30 rounds per minute. After mixing, the strand mat was manually formed in a lab-size forming box of 450 mm × 450 mm without orientation. The moisture content of the mat was adjusted to 10% by spraying water during blending. The mat was pre-pressed and directly placed into a single opening hot-press. A target thickness of 18 mm was established using two iron-distance bars. The boards were pressed at a nominal pressure of 6.3 N mm^{-2} and a temperature of 200 °C for 15 s mm^{-1}. Target densities were 300 kg m^{-3} and 400 kg m^{-3}. After pressing, all boards were conditioned at 20 °C and 65 % RH for at least 2 weeks to reach the constant mass. Three replicate boards were produced for each factor combination (wood species and board density). The vertical density profile of each board was recorded with an x-ray densitometer (DAX, Fagus-Grecon GmbH & Co. KG, Alfeld, Germany). Bending strength (modulus of rupture, MOR) and modulus of elasticity in bending (MOE) was determined according to EN 310 (1993; 7 replicates per board, n = 21), internal bond strength according to EN 319 (1993; 5 replicates per board, n = 15). Thickness swelling was assessed after 24 h immersion in water at 20 °C according to EN 317 (1993; 5 replicates per board, n = 15).

Statistical analysis

Fifth and 95th percentiles of panel properties were calculated by

$$P_\alpha = \mu + t_{n-1,\alpha}\sigma \quad \textbf{(1)}$$

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where P_α: α^{th} percentile; μ: sample mean; σ: sample standard deviation, $t_{n-1,\alpha}$: α^{th} percentile of the t-distribution with n-1 degrees of freedom; n: sample size.

Results and Discussion

Strand geometry and panel structure

The average density of kiri, pine and beech strands was 193…238…296 kg m^{-3}, 358…495…604 kg m^{-3} and 538…605…765 kg m^{-3} (min…mean…max), respectively. Kiri strands (100 g) covered the largest area on the scanner followed by pine and beech strands (Figure 2). The difference between pine and beech was relatively small, but consistent for all sieve fractions, with the area slightly decreasing as the sieve fraction increased (Figure 2). Distributions of strand length and width differed only slightly between wood species (Figure

3). Strands longer than 90 mm covered only a minor proportion of the total area so that most strands were shorter than the target length. Length distribution was very wide with two peaks for the smallest sieve fraction, while it was narrower for the larger fractions. The maximum strand width corresponded to the upper mesh size, but all sieve fractions included strands that were narrower than the lower mesh size. Accordingly, width distribution increased with increasing sieve fraction. Strand thickness varied widely and it seemed to depend on the wood species, with kiri strands being the thickest followed by pine and beech (Figure 4). Also, strands in the smaller sieve fractions appeared to be slightly thinner than those in the larger fractions. The thickness of most strands, however, was within the target range of 0.5 mm to 1 mm.

The most important difference between the wood species used in this study is their density. Strand densities determined for kiri and pine are similar to values given in the literature. That of beech is unexpectedly low but within the range reported by Kollmann (1951). Calculating the wood density based on strand area (of 100 g, Figure 2) and mean thickness (Figure 4) yielded the following values (average of three fractions): 175 kg m^{-3} (kiri), 332 kg m^{-3} (pine) and 448 kg m^{-3} (beech). These values are much lower than those found by the submersion method. Measured thickness values accordingly seemed to be too high. This may be due to surface roughness or deformation of strands. Therefore, thickness values may be used only for comparisons.

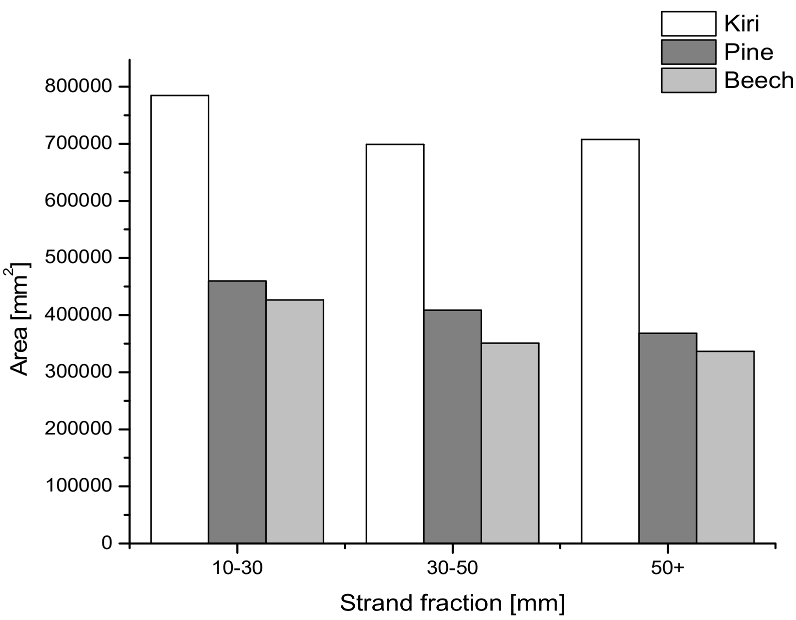

Figure 2 Total area of 100 g of strands for different strand fractions and species

The area of a specific mass of strands is affected by both wood density and strand thickness. While wood density determines the overall wood volume, strand thickness affects how this volume is distributed in the vertical and horizontal direction. Consequently, a low wood density and thin strands result in a large strand area. Strand thickness is mainly controlled by the projection of the knives in the flaker and the feeding speed (Kruse *et al.* 2000). When logs of different densities are used to produce strands with identical settings, the low-density wood typically yields thicker strands. The reason is that the wood is compressed during the cutting process and expands afterward. Due to the lower strength, this compression is higher in low-density woods. In the present study, a slower feeding force was used for stranding kiri wood as for pine and beech, which generally results in thinner strands. Still, this adjustment did not completely compensate for the density effect, so that strand thickness increases as wood density decrease. Comparing pine and beech, the effect of strand thickness on the strand area obviously compensated for the effect of wood density. For the strand area of kiri, the low wood density is the determining factor. The decrease of the strand area with increasing sieve fraction is also related to strand thickness. The latter increases with sieve fraction, because thick particles are less likely to break into smaller parts during strand generation and handling.

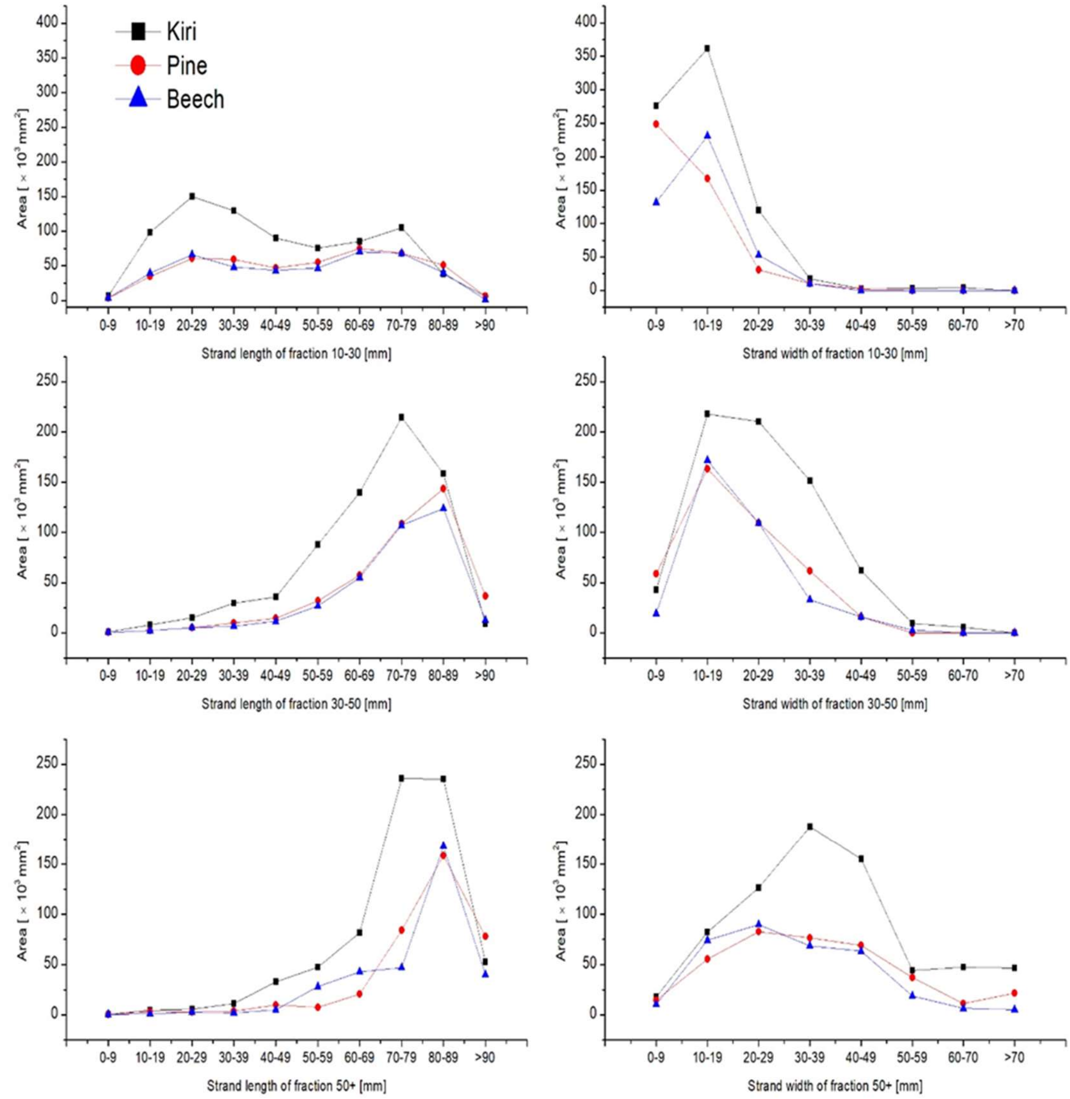

Figure 3 Distribution of strand area according to the length and width of the strand in each fraction

The width and length of strands were very similar for the different species within each sieve fraction. Accordingly, strand shape is assumed not to cause differences in board properties. Target strand length is set by the distance of the scoring knives in the flaker and usually can be controlled accurately. The reason for the short length of strands in this study probably is that the calculation method underestimates maximum length in fiber direction due to the underlying assumption that strands represent perfect rectangles. In contrast, the minimum caliper diameter is assumed to correspond to the widest part of an irregularly shaped strand, so that the modeled strand with the identical area is slightly wider and shorter than the real one. Distribution of strand width is usually broader than that of strand length because it results from a fracture along the grain direction of the wood sheets immediately after cutting due to a bending force. The width distributions of single sieve fractions show that the vibratory screener mainly sorts strands by widths. In doing so, it effectively retains strands wider than the mesh size, but also narrower ones.

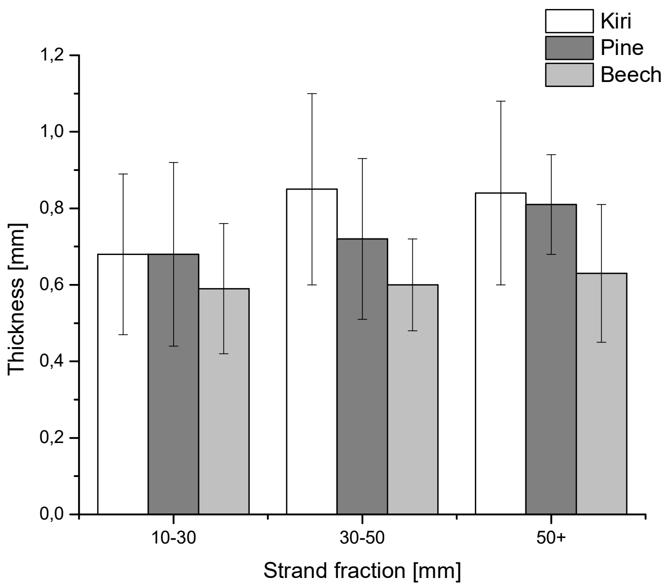

Figure 4 Strand thickness; columns: mean values; error bars: total span; (n = 20)

The bulk density of kiri strands was lowest followed by that of pine and beech strands, with the latter two being quite similar (Figure 5). It slightly increased with strand size, but the recorded density of the mixture did not obviously depend on a particular sieve fraction. The mixture of beech strands used for panel manufacturing had a bulk density of ca. 100 kg m^{-3}. Accordingly, the mat was densified 3-fold or 4-fold during hot pressing of panels with target densities of 300 kg m^{-3} and 400 kg m^{-3}, respectively. Nevertheless, the obtained boards were not sufficiently solid for handling and cutting of specimens, so that investigating panel properties was not possible. Pine boards had a more solid structure than beech panels (Figure 6), although the bulk density of the mat was only slightly lower (Figure 5). Compared with kiri panels, pine panels contained more interparticle voids resulting in more porous edges and inhomogeneous panel structure (Figure 6). This was confirmed by the respective density profiles (Figure 7), which showed a jagged, horizontal line for pine panels. In contrast, the density profiles of kiri panels exhibited a smooth line with peaks near the surface.

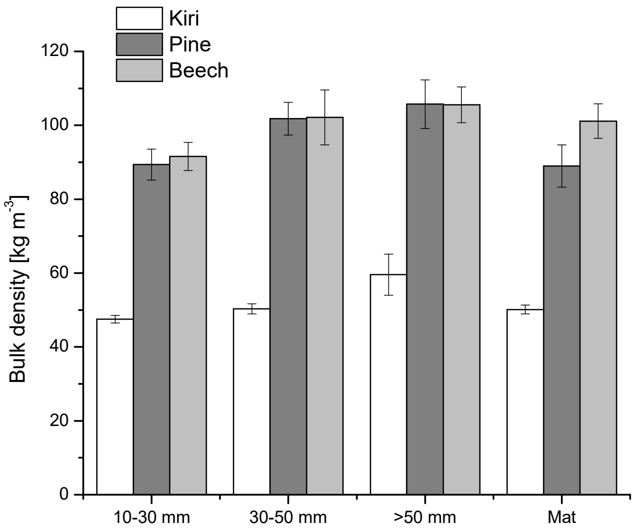

Figure 5 Bulk density of strand mats consisting of different species, sieve fractions and the mixture of fractions used for panel manufacturing; columns: middle value; error bars: from lower to upper value; (n = 3)

A strand mat contains wood and voids. Thus, its bulk density depends on the wood density and shape of the strands, because the latter determines the void volume fraction of the mat. In this study, bulk density correlates remarkably close with strand surface area. Besides wood density, thus, strand thickness seems to be the crucial factor that determines mat density. Obviously, a decreasing strand thickness results in a mat with more horizontal layers, which increases the void volume in the mat. As for the strand surface area, the low thickness of beech strands seems to compensate for the lower density of pine resulting in a similar bulk density. In the same way, thicker strands in larger sieve fractions seem to cause the respective increase in mat density. The effect of strand width and length on mat density, the main difference between sieve fractions, seems to be negligible for flat strands. This effect contrasts with the wood furnish materials used for particleboards, the bulk density of which reduces as particle size increases (Schmutzler 1963, Shang *et al.* 1999). Also different from the findings in the present study, the pore structure of wood furnish for particleboards is mainly determined by the smallest fraction, because the smaller particles fill up the spaces between larger particles (Shang *et al.* 1999, Thömen and Klüppel 2008).

Figure 6 Representatives samples of SBs panels from kiri, pine, beech (top to bottom) with a target density of 300 kg m^{-3} (left side) and 400 kg m^{-3} (right side)

Upon densification, the void volume of the mat decreases and strands get closer to each other, because they are rearranged and deformed. As soon as continuous vertical strands columns are formed, wood is compressed. At low compaction ratios, movement and deformation of strands predominate and, thus, compression resistance of the mat is low. As the compaction ratio increases, the densification of wood substance occurs and requires much higher forces so that compression resistance progressively increases (Zombori *et al.* 2001, Benthien *et al.* 2018). The present results show that the structure of the finished panel depends on wood density rather than mat density because the former determines the inter-particle void volume fraction: In beech panels, this fraction is so large that there is an insufficient contact area between strands for adhesive bonding. For pine panels, the ragged density profile indicates small-scale density variations, i.e. alternation of wood substance and voids. In kiri panels, the strands seem to be in close contact.

Vertical density profiles of wood-based panels typically show peaks near the surfaces with a lower density in the core. This profile is predominantly formed during the closing of the hot press. The stresses applied to each layer are the same and equal the mat pressure. Surface layers, however, heat up first and to a higher temperature than the core resulting in a lower compression modulus and higher densification. The effect of temperature certainly depends on the densification mechanism. It is obviously negligible for the rearrangement of particles, but crucially affects deformation and compression of wood. Furthermore, compression of wood particles is probably more sensitive to temperature changes than deformation, as MOE and compressive strength perpendicular to the grain is the most temperature-sensitive wood properties (Gerhards 1982). This is confirmed by the finding of v. Haas and Frühwald (2000) that the effect of temperature increase as mat density increases. Consequently, we assume that the vertical density profile of kiri panels showed distinct surface peaks because the low wood density required considerable densification of strands during hot pressing. In contrast, densification of the pine mat mainly involved strand rearrangement and deformation. The general finding that the utilization of low-density wood supports the formation of a more pronounced density profile accords with previous studies (Akrami *et al.* 2014, Beck *et al.* 2009). Also, the very narrow surface densities peaks are typical for boards made of low-density wood (Wang and Winistorfer 2000).

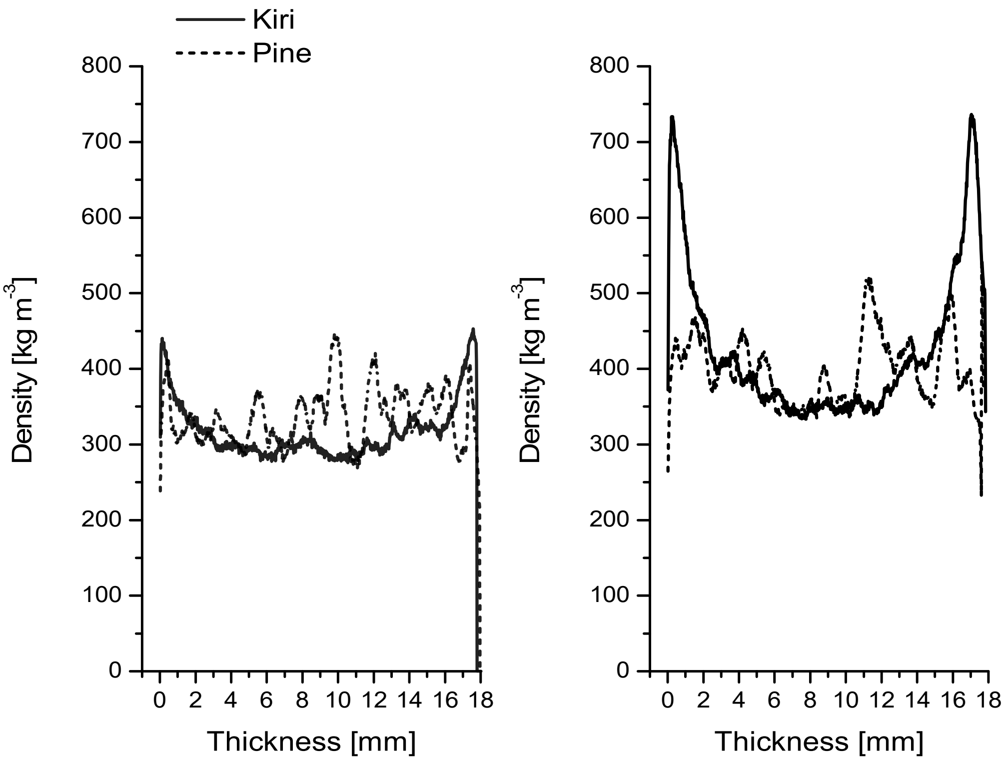

Figure 7 Representative vertical density profiles of pine and kiri boards with a target density of 300 kg m^{-3} (left) and 400 kg m^{-3} (right)

Panel properties

Strength values, i.e. MOR, MOE and IB, increased with board density and they were higher for kiri boards than for pine boards (Figure 8 and Figure 9). At a low panel density (300 kg m^{-3}) the difference between kiri and pine panels was larger than at higher panel density (400 kg m^{-3}); for MOR and IB it was larger than for MOE: MOR of kiri boards was 2.6 and 1.6 times higher than that of pine boards at 300 kg m^{-3} and 400 kg m^{-3}, respectively, while these ratios were only 1.8 and 1.2 for MOE. Especially the internal bond strength of low-density pine panels was remarkably low. Thickness swelling of kiri boards exceeded that of pine boards, while water uptake during the test was similar for both species (Figure 10). While thickness swelling was higher at a high density, the percentage of water uptake was higher for low-density boards. IB and TS of kiri boards met the standard requirements at 400 kg m^{-3} (panel thickness: 18…25 mm; EN 300:1997).

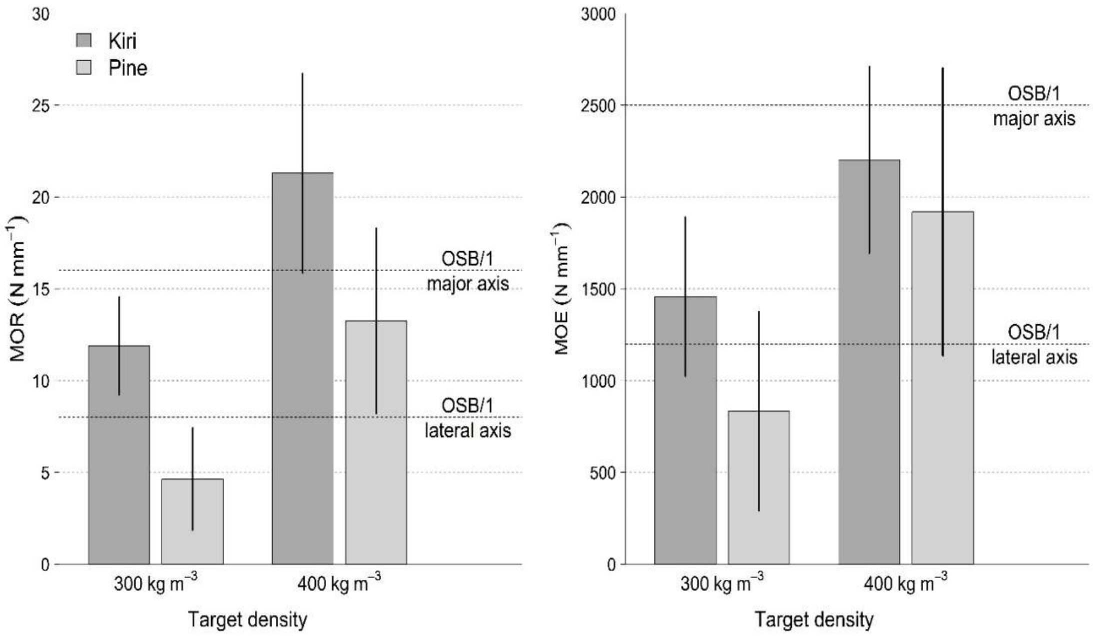

Figure 8 MOR (left) and MOE (right) of strand boards made of kiri and pine; columns: mean values; error bars: 5th to 95th percentile; dashed lines: 5th percentile requirements for OSB/1 according to EN 300

The mechanical properties of wood-based panels depend on the strength of individual wood particles and the adhesive bonds between them. The strength of wood particles generally increases with density, which depends on species (initial density) and densification during hot pressing. However, densification can also damage particles and, thus, decrease their tensile strength (Price 1976, Geimer *et al.* 1985). The bonding strength between the particles is affected by resin area coverage and strand contact area (He *et al.* 2007). For a given mass fraction of resin, a large surface area of low-density wood particles results in low resin area coverage. In contrast, strand area contact increases as wood density reduce, because of the more solid board structure described above. Due to the development of a vertical density profile, board density and compaction ratio are not uniform within the board and the effects of wood density are not identical for different board properties. While bending properties are mainly determined by the surface layers, internal bond strength predominantly depends on the core layer. The steep profile caused by low-density wood species tends to enhance bending properties but to lower internal bond strength (Jin, J. *et al.* 2009).

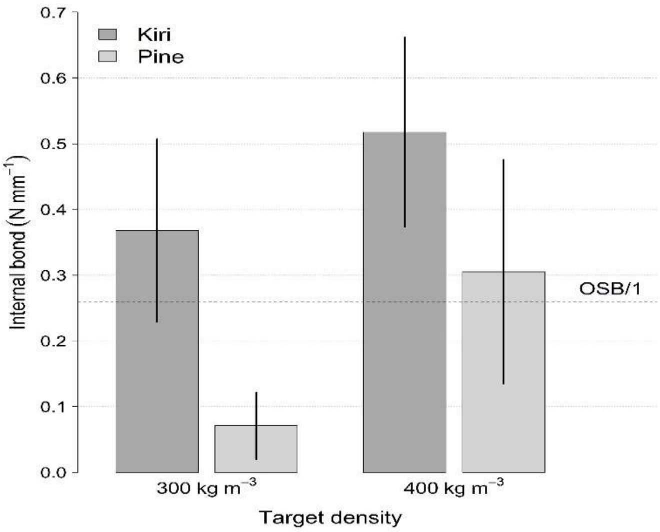

Figure 9 Internal bond strength of kiri and pine strand boards at densities of 300 kg m^{-3} and 400 kg m^{-3}; columns: mean values, error bars: 5th to 95th percentile; dashed lines: 5th percentile requirements for OSB/1 according to EN 300

In the present study, differences between the mechanical properties of kiri and pine boards seem to be predominantly caused by the different strand contact area and, thus, inter-particle bonding strength. Accordingly, internal bonding strength, a parameter that serves as a measure of inter-particle adhesion, shows the largest differences between the species, although board density of the core layers is similar. Additionally, the fact that the adhesive bonding of beech boards was insufficient for handling confirms this finding as well as several earlier studies (e.g. Klauditz and Stegmann 1958, Zhou 1990). Despite the high surface layer density of kiri boards, bending properties varied less than internal bond according to species. This was especially true for MOE. This result suggests that MOR is mainly determined by adhesive bonding, while MOE is primarily determined by the properties of the individual strands. This finding accords with Hse (1975), who compared black tupelo flakeboards with red maple flakeboards. The cross-grained black tupelo wood exhibited a similar density as the red maple wood but a lower MOR and MOE. While the low MOE of the black tupelo wood was transferred to flakeboards made thereof, the MORs of the boards were similar. The reason might be that the glue joints behave more brittle (higher MOE, lower MOR) than the solid wood, so that elastic deformation occurs mainly in the wood elements of the board, while adhesive bonds might fracture first.

When exposed to water (liquid or vapour), strand boards swell. Thickness swelling is distinctly larger than in-plane swelling. It involves three components: swelling of the strands; partial reversal of densification of strands that were compressed during hot pressing (spring-back); development of inter-particle voids due to breaking of adhesive bonds by internal stresses. The higher the wood density, the more the wood swells. The higher the compaction ratio, the higher are the spring-back and internal stresses, but also the internal bond strength that can take up internal stresses. Overall, the low density, i.e. high compaction ratio, of kiri wood resulted in higher thickness swelling of the boards than those of pine.

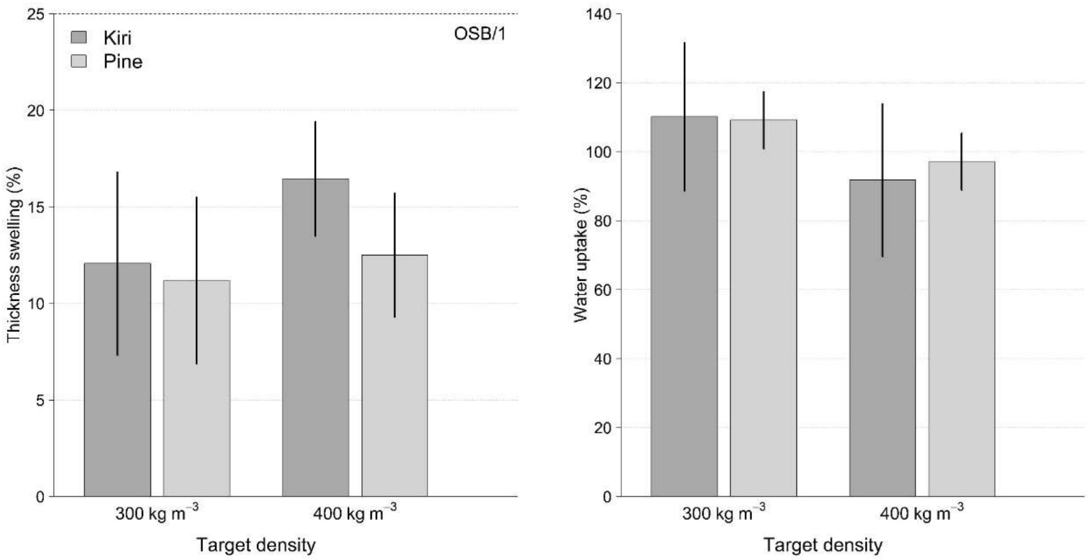

Figure 10 Thickness swelling (left) and water uptake (right) of kiri and pine panels at densities of 300 kg m^{-3} and 400 kg m^{-3} after 24h of immersion; columns: mean values; error bars: 5th to 95th percentile; dashed lines: 95th percentile requirements for OSB/1 according to EN 300

Water is taken up by cell walls, intra-particle and inter-particle voids. Therefore, the absolute amount depends on board density rather than board structure. However, a solid structure slows down the rate of water uptake. The larger swelling allowed kiri boards to take up more water than pine boards, while their structure caused a slower rate. Present results suggest that both effects are compensated for each other.

Conclusions

This study proved the feasibility of producing strand boards from the low-density kiri wood with only minor adjustments to the production process, e.g. the strand generation. Due to the low density of kiri wood, the bulk density of the kiri strand mat was much lower than that of pine and beech mats. However, the density of the beech mat was similar to that of the pine mat, because the beech strands were thinner than the pine strands. The fact that it was possible to produce pine boards but beech strands did not form a cohesive board at the target densities, confirms that wood density rather than mat density determines board properties and that increasing mat density by shaping strands has little effect. The lower the wood density, the better the mechanical properties of the boards. Low wood density, however, resulted in higher thickness swelling, because the densification of wood strands is reversed during swelling.

Results from this study on randomly oriented strand boards cannot be compared to standard requirements for oriented strand boards, because strand orientation crucially affects board properties. Still, present results suggest that it is possible to meet IB- and TS-requirements for OSB/1 at a board density of 400 kg m^{-3}, corresponding to a compaction ratio of ca. 1.6. A comparison with standard requirements reveals that low-density boards generally exhibit low thickness swelling. In contrast, MOE seems to be generally low for lightweight boards. This is remarkable because the large compaction ratio resulted in a steep vertical density profile, the high surface densities of which generally improve bending properties. Obviously, MOE is primarily determined by the properties of the (densified) wood strands, which correlate with panel density, while MOR and IB mainly depend on inter-particle adhesive bond strength, which correlates with the compaction ratio.

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Publication II

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

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

Abstract

Kiri (*Paulownia tomentosa*) wood is a promising material for lightweight strand boards (SBs); however, kiri SBs have displayed 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.

Keywords: low molecular weight; high molecular weight; strand boards; kiri wood; *Paulown*

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, two main factors are 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 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 to 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 applications. 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 rotating speed at 30 rounds min^{-1}. PF resin with low and high molecular weight was used as adhesive with the amount 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). 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 a 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 resin stock solution, according to the Eq.1:

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

Where $\boldsymbol{M_s}$ is the oven-dried mass of the strands [g], $\boldsymbol{R_s}$ is the percentage of used resin based on the oven-dried mass of the strands [%] and $\boldsymbol{S_c}$ 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 Eq.2:

$$WPG = (M_{PF}/M_m) \times 100 \quad (2)$$

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

Table 1. 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

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 × 50 × 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 Eq. 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 the 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 × 50 × 12 mm^3, the surface of the 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 the 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 the specimen.

Brinell hardness (BH)

For BH, dimensions of test specimens were 50 × 50 × 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 Eq. 4:

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

Where D is ball diameter, F is the 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 in the middle of the specimen's surface and 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-liter 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. In the end, the formaldehyde concentration was determined by the acetylacetone 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 (Fig. 1), although the weight percent gains (WPG) of panels manufactured with LMW PF was lower than that of respective SBs made with HMW PF (Table 1). 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 (Fig. 1). 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 was approximately 30% higher than those containing HMW PF resin.

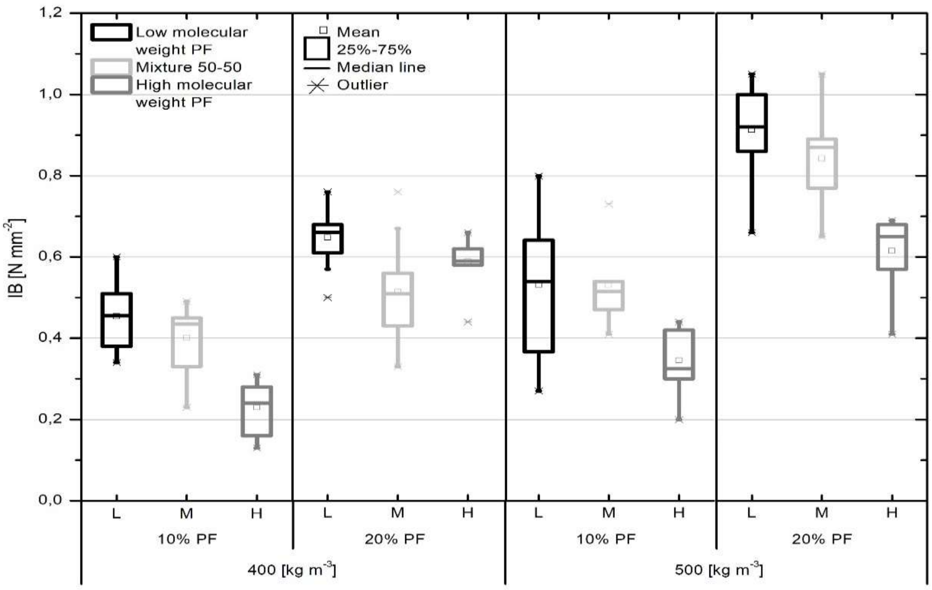

Figure 1 Internal bonding strength (IB) of kiri strand boards with a target density of 400 kg m^{-3} and 500 kg m^{-3} (n=10)

As observed for IB, both MOR and MOE increased with 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).

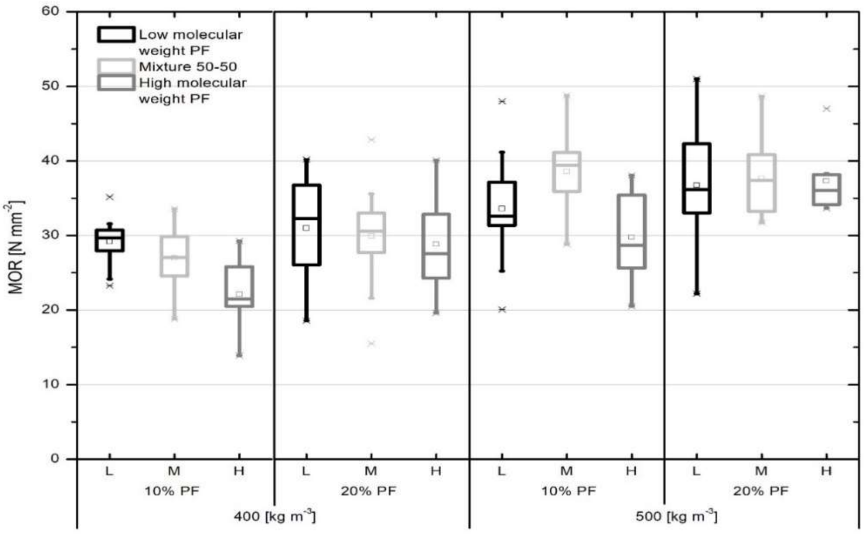

Figure 2 Modulus of rupture (MOR) of kiri SBswith target density of 400 kg m^{-3} and 500 kg m^{-3} (n=14)

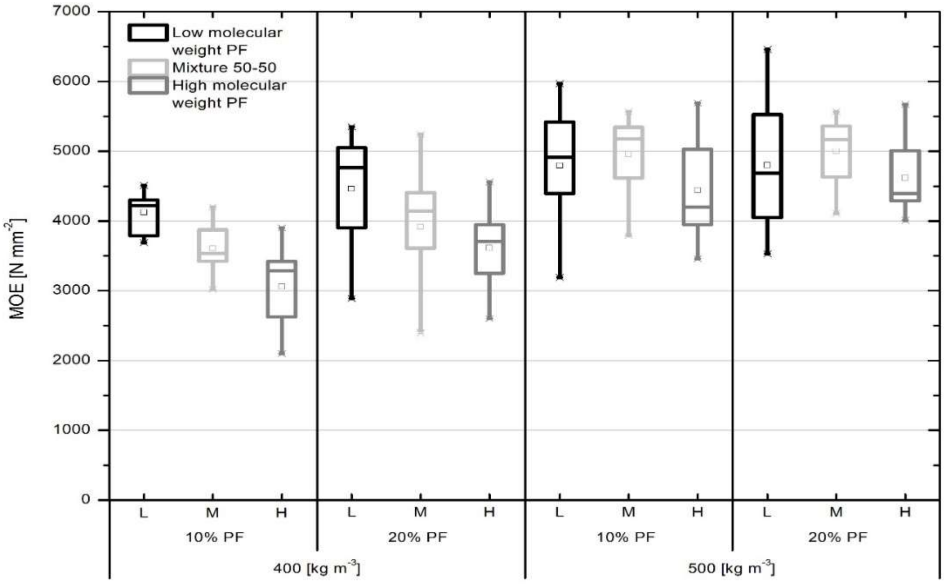

Figure 3 Modulus of elasticity (MOE) of kiri SBswith target density of 400 kg m^{-3} and 500 kg m^{-3} (n=14)

As can be seen in Fig. 4, at the 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 and resin content was apparent (Fig.4).

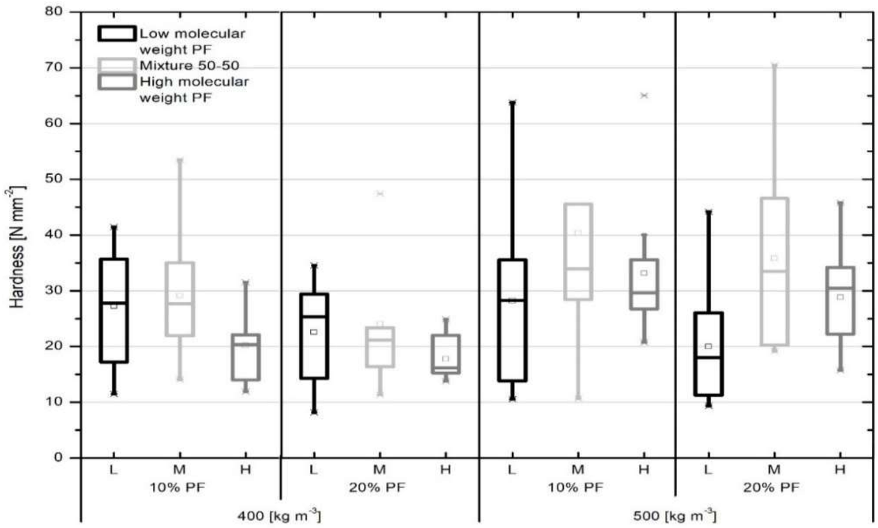

Figure 4 Brinell hardness of kiri strand boards with target density 400 kg m^{-3} and 500 kg m^{-3} (n=10)

Water-related properties

Thickness swelling (TS) increased with increasing density of the panels (Fig. 5), which is in accordance with earlier 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.

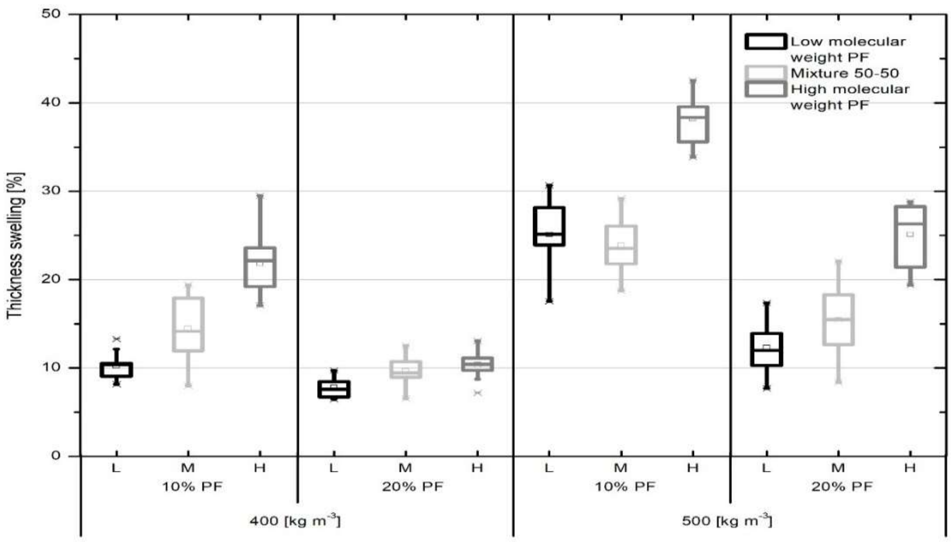

Figure 5 Thickness swelling of kiri strand boards with target density 400 kg m^{-3} and 500 kg m^{-3} after 24 hours (n=10)

In reverse to TS, relative water absorption (WA) linearly decreased with increasing panel density (Fig. 6), while the absolute WA (related to panel volume) was similar for both densities (not shown). For all SBs variants, the WA of LMW PF boards was significantly lower than that of the respective HMW PF panels (Fig. 6). 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.

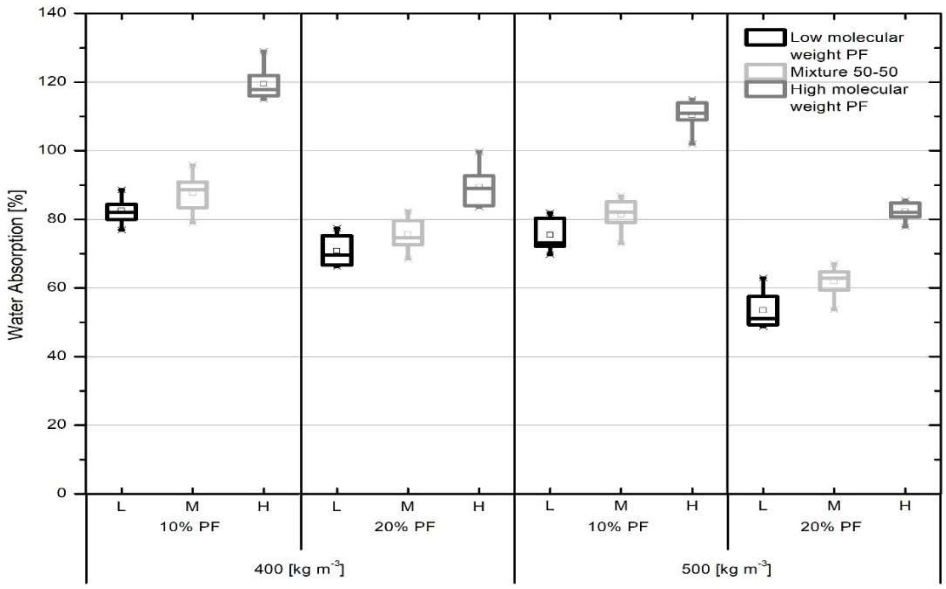

Figure 6 Water absorption of kiri strand boards with target density 400 kg m^{-3} and 500 kg m^{-3} after 24 hours (n=10)

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 the 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 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 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 a 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 that 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 (Fig. 7). The appearance of respective panel specimens is shown in Figure 1. The values of ITS were somewhat lower than TS after 24 h immersion in water (Fig. 7).

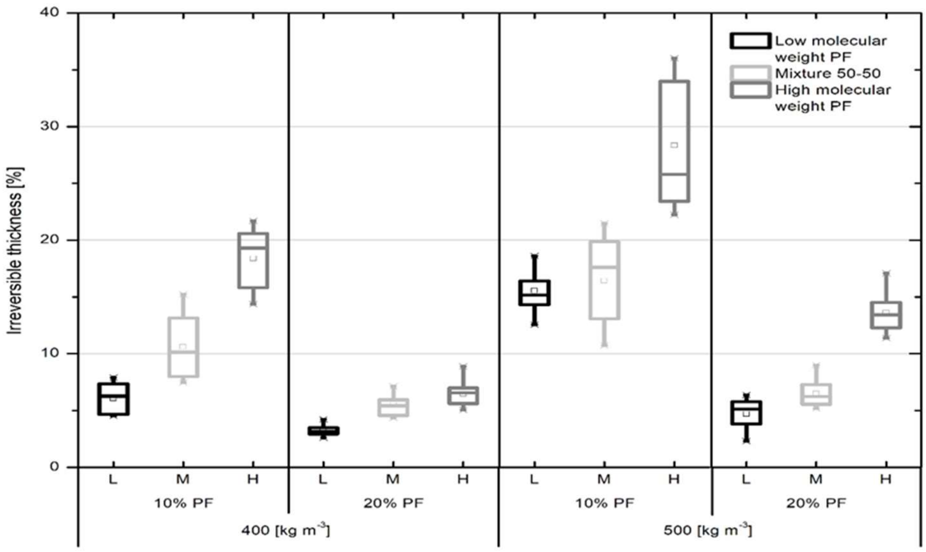

Figure 7 Irreversible thickness swelling with a vacuum at the first hour after immersion in water for 24, 48, 96h and dried 24h for each temperature 40^0C, 60^0C, 80^0C and at 103^0C until certain mass (n=10)

The water absorption after vacuum (WA-v) and 96 h immersion in water were about 2.3 times higher than that after 24 h water immersion (WA; Fig. 8 and 9), 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.

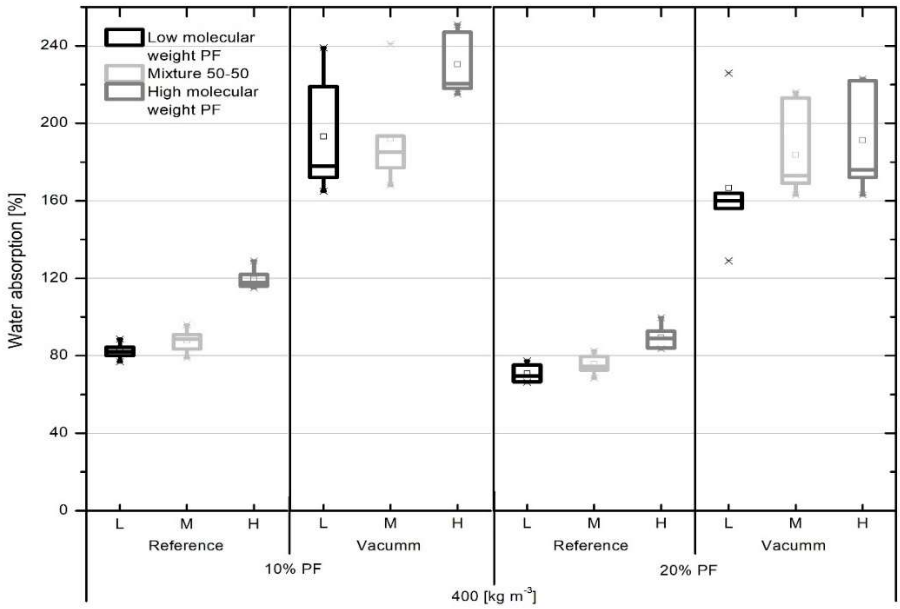

Figure 8 Water absorption of OSB samples with board density 400 kg m^{-3} after vacuum at first hour and immersion in water for 24, 48, and 96 h and dried 24h for each temperature 40^0C, 60^0C, 80^0C and at 103^0C until certain mass (n=10)

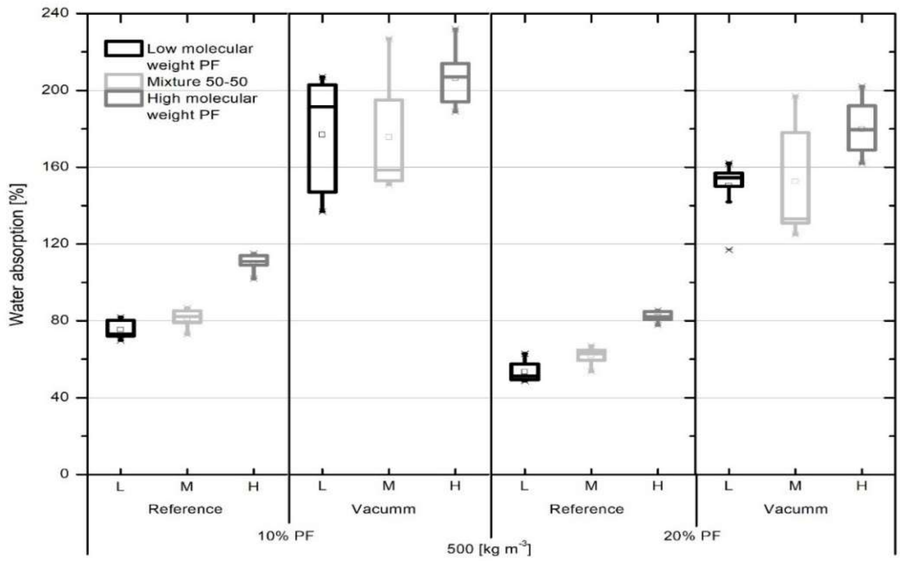

Figure 9 Water absorption of OSB samples with board density 500 kg m^{-3} after vacuum at the first hour and immersion in water for 24, 48, and 96 h and dried 24h for each temperature 40^0C, 60^0C, 80^0C and at 103^0C until certain mass (n=10)

The mass loss (leaching) 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 (Fig. 10). Panels with lower density underwent higher relative ML than those of higher density.

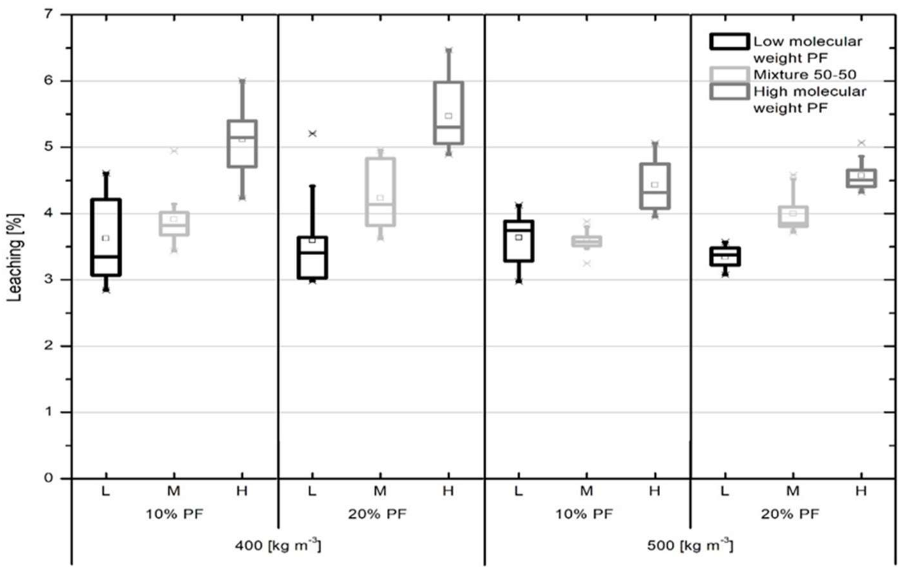

Figure 10 Leaching (mass loss) of SBs samples after vacuum at the first hour and immersion in water for 24, 48, and 96 h and dried 24h for each temperature 40^0C, 60^0C, 80^0C and at 103^0C until certain mass (n=10).

Figure 11 Irreversible thickness swelling SBs samples with high molecular weight PF (left); low molecular weight PF (right) after vacuum at the first hour, then immersion in water for 24, 48, 96h and dried 24h for each temperature 40^0C, 60^0C, 80^0C and at 103^0C until certain mass; initial thickness (middle).

The contact angle (CA) of water on kiri strand surfaces was determined after 2 and 4 s (Fig. 12). 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. The higher surface energy of the 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 was slightly higher compared to the untreated control (Fig. 13).

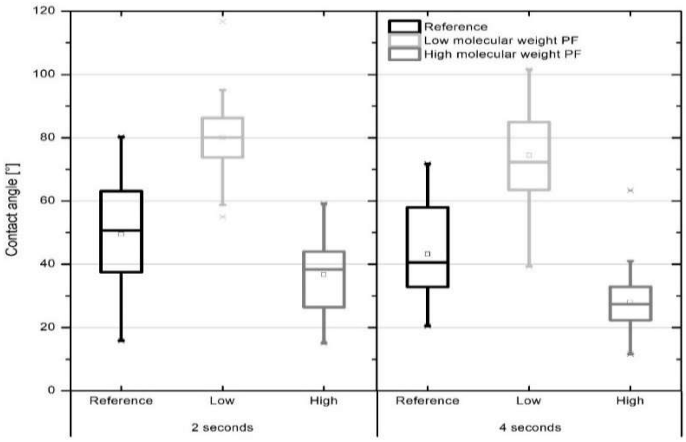

Figure 12 Contact angle on kiri strands treated with low and high molecular weight PF and non-treated kiri strands. Droplets were measured at 2 and 4 seconds (n=20)

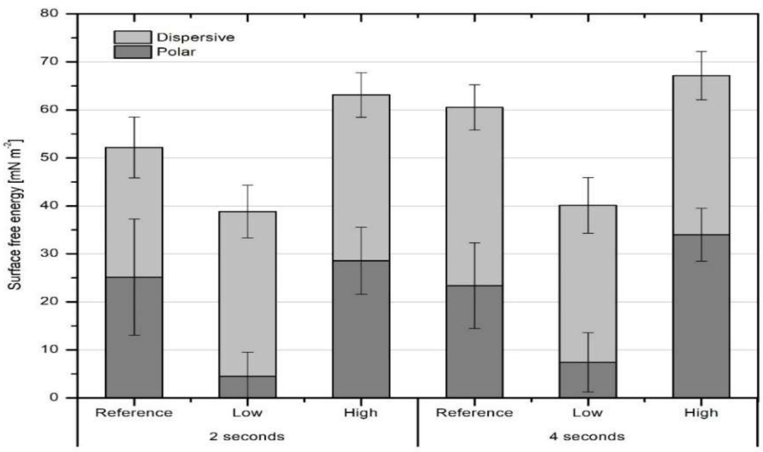

Figure13 Surface free energy of kiri strands treated with low and high molecular weight PF and non-treated kiri strands. Droplets were measured at 2 and 4 seconds (n=20)

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 (Fig. 14). 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 the emission of free FA. At equal pressing temperature, the HMW PF resin needs a considerably 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.

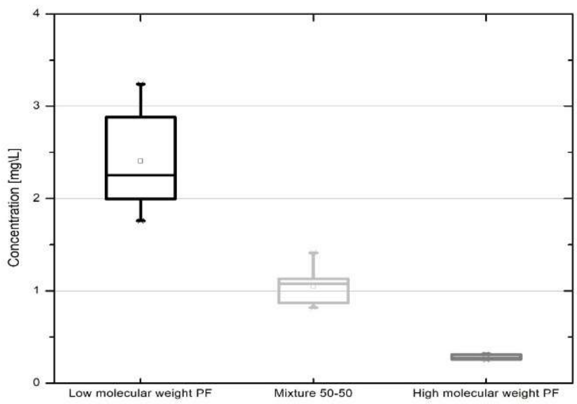

Figure 14 Free formaldehyde emission of kiri strand boards with a target density 500 kg m^{-3} at resin content 10% following standards EN 717-2 (n=2)

Conclusion

Utilization of LMW PF as an 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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Disclosure statement

No potential conflict of interest was reported by the authors.

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Publication III

Performance properties of light-weight strand boards reinforced with veneers and veneer strips

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Performance properties of light-weight strand boards reinforced with veneers and veneer strips

Abstracts

Veneers and veneer strips were used as a reinforcement layer on the surfaces and between wood strands to improve the properties of strand boards. These veneer strand boards (VSB) of 500 kg m^{-3} density were produced from kiri (*Paulownia tomentosa*) and beech (*Fagus sylvatica*) strands as a core material with veneers or veneer strips from kiri and birch wood (*Betula pendula*) as face layers in one production step. Only the strands (core material) were resinated with phenol-formaldehyde adhesive. The obtained sandwich structure significantly improved the mechanical properties of the panel. With respect to VSBs with a kiri core, the utilisation of kiri veneers led to a 45% and 39% increase in MOR and MOE, whereas birch veneers induced 80% and 77% increase, respectively, compared to kiri strand board without veneer. The sandwich structure makes it possible to produce low-density boards bearing high bending strength for furniture and construction purposes with low technical effort.

Keywords: veneer strand board, sandwich structure, veneer, veneer strips, *Paulownia* sp.

Introduction

In recent years the application of light-weight wood-based panels in the furniture and construction industry is emerging as a topical issue in many developed countries (Jivkov et al. 2012). Regarding wood-based panel production, raw materials and transportation costs are essential economic influencing factors for producers and end-users. As such, the lighter the products and raw materials, the more ecological and economical transportation solutions become (Barbu 2015). In general, there is no proper definition of a light-weight board, but it is widely recognized to categorize the light-weight panels in terms of their density: light-weight, very light-weight and ultra-lightweight with a density below 500 kg m^{-3}, 350 kg m^{-3}, 200 kg m^{-3}, respectively (Jivkov et al. 2012). One solution to reduce the density of products is to use a sandwich structure with various core materials such as homogeneous, inhomogeneous or hollow material (Nilsson et al. 2013).

The use of the sandwich structure provides distinct advantages through efficient design to utilize each material to its ultimate limit (Nilsson et al. 2013). Several scientists have investigated reducing the weight of panels by using different light-weight materials as core layers such as popcorn (Burnett and Kharazipour 2018), expandable polystyrene granules (Shalbafan et al. 2016), natural formable resins from sour cassava starch (Monteiro et al. 2016), or using paper honeycomb with hexagonal and auxetic cells (Smardzewski 2013). A technique for continuous production of a polyurethane foam core panel with wood-based facings has been introduced at a laboratory scale (Barbu et al. 2010), which obtained a weight reduction of 30-50%. The disadvantage of using synthetic materials for wood-based panels is that these materials are non-renewable and that mixtures are often more challenging to recycle. Moreover, the costs of raw materials such as foams and expandable materials are usually higher than those of standard panels (Barbu et al. 2010). Other aspects, such as material saving and resource efficiency as well as the ethical aspect that some materials are food products, are becoming more relevant for the application of light-weight materials. In addition, a negative consequence of weight reduction might be a decrease in mechanical properties (Nilsson et al. 2013).

Plywood is manufactured from an odd number of thin veneers, where each veneer is laid down with its grain perpendicular to its adjacent veneer (Thoemen et al. 2010). Plywood has been promoted for residential construction in the 1950s, and its production had reached 16 million m^3 by 1980 in North America. Since then, the oriented strand board (OSB) was introduced, which has considerably displaced plywood as a structural wood panel in the construction industry (Shi and Walker 2006). One of the reasons for introducing OSB to the market and to replace plywood is its productivity, as plywood production requires about three hours per cubic metre while OSB only requires around one hour per cubic metre (Spelter 1988). Also, the raw material of plywood needs to adhere to specifications regarding density, ease of peeling or slicing, drying without wrinkling and volume, while the OSB process is not dependent on the large diameter and old-growth logs (Spelter et al. 1997; Shi and Walker 2006). Hence, the combination of veneers and strands would be a promising solution for structural wood-based panels industry. Weight and Yadama (Weight and Yadama 2008) produced an unveneered thin wood strand composite of ponderosa pine (*Pinus ponderosa*), 3.2 mm thick, as a veneer substitute. The development of a low-density OSB was described in the patent literature by combining wood strands with a thermosetting resin, which is preheated before hot-pressing to achieve a homogenous vertical density profile (Chen and Wellwood 2010). The hygro-mechanical properties of these panels were acceptable but should reportedly be improved.

The objective of this study was to evaluate the possibility of using peeled veneers and low-density *Paulownia* wood strands to produce new light-weight products, which might be an alternative for plywood in light-weight construction. Various combinations of kiri (*Paulownia tomentosa*) (as a low-density wood species) and beech (*Fagus sylvatica*) strands were used for the core layer and kiri and white birch (*Betula pendula*) veneers for the surface layer to investigate the effects of the board structure and wood density on the hygro-mechanical properties of the sandwich boards.

Materials and methods

Strands and veneers production

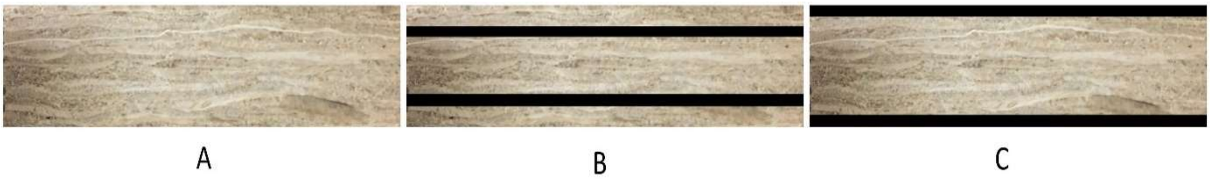
Kiri trees (*Paulownia tomentosa* (Thunb.) Steud.) were planted and harvested near Mannheim (Germany). They were provided by the company WeGrow GmbH (Tönisvorst, Germany). Trees were cut into logs with diameters of 12-20 cm for kiri and 30-35 cm for beech (*Fagus sylvatica* L.), which were stranded at Fraunhofer WKI (Braunschweig, Germany). The logs were 2 m long and debarked manually before loading to a knife ring flaker PZUL 8-300 (Pallman Company, Zweibrücken, Germany) with target stranding dimensions of 110 mm length, 0.5-1 mm thickness and 10-50 mm width. The log was firmly held by a hydraulic clamp and moved towards the knife ring with a speed of 36 mm s^{-1} (kiri) and 40 mm s^{-1} (beech). The classification of the strands was carried out by a sieving shaker. Three fractions of strands sorted by width: 10-30 mm, 30-50 mm and bigger than 50 mm and sieve fractions smaller than 10 mm were categorized as fines. Then, the kiri and beech strands were immediately dried at 70°C to a moisture content of 3% to 5% in a drying oven. To avoid moisture absorption, the strands were stored in plastic bags during transport to University of Göttingen, (Germany) for panel manufacturing. White birch (*Betula pendula*) and kiri (*Paulownia tomentosa*) veneers exhibited a thickness of 1.5 and 2.2 mm, respectively. They were trimmed to dimensions of 430 × 430 mm with a density of approximately 570 kg m^{-3} for birch veneer and 250 kg m^{-3} for kiri veneer. Birch and kiri veneers were also cut into strips with dimensions of 430 × 65 mm. The veneers and strips were dried at 70°C in an oven for 24 h.

Panel manufacturing

Twelve variants of veneer strand boards (VSB) were produced with a target density of 500 kg m^{-3} (including the veneers). In total, 28 panels were produced, which consisted of two VSB for each of ten variants and four strand boards (SB) for each of two reference strand board variants without veneer (Table 1). The strand fractions were mixed in a ratio of 4:5:1 (10-30 mm: 30-50 mm: >50 mm). The strands were directly blended with an amount of 10% phenol-formaldehyde (PF) adhesive solution based on the oven-dry weight of strands (in a drum blender with a rotating speed at 30 rounds min^{-1}). PF adhesive was provided by Prefere resins Germany GmbH (Erkner, Germany) with an average M_w of 1000-1200 g mol^{-1} and 70% solid content. The resinated strands were formed into a mat without orientation (450 × 450 mm^2) in a cold pre-press before hot-pressing at 140 °C for 90 s mm^{-1} using metal bar stops to ensure a panel target thickness of 12 mm to produce the reference boards (Table 1A). To produce sandwich VSB, a veneer was placed at the bottom and the top of the resinated strand mat (Table 1C). In addition, the second variant of VSB was produced with veneers and veneer strips placed 3 mm under the panel surface at both sides ("veneer inside", Table 1B). After hot-pressing, the VSB was cooled to room temperature for 48 h.

Table 1

Structure layup of mats for twelve panel variants. Each mat consisted of two veneers that had a parallel grain direction.



Panel type	Board structure		
	Veneer	Position of veneer	Strand
1.	Birch	C	Kiri
2.	Birch	B	Kiri
3.	Kiri	C	Kiri
4.	Kiri	B	Kiri
5.	Birch	C	Beech
6.	Birch	B	Beech
7.	Kiri	C	Beech
8.	Kiri	B	Beech
9.	Birch strips	B	Kiri
10.	Kiri strips	B	Kiri
11.	-	A	Kiri
12.	-	A	Beech

Physical and mechanical properties

All specimens were conditioned at 20 °C and 65% relative humidity for at least two weeks to reach equilibrium moisture content. The density of all specimens was determined according to EN 323 (1993a). Bending strength (modulus of rupture, MOR) and modulus of elasticity in bending (MOE) were assessed according to EN 310 (1993b) (7 specimens per board, n = 14) with the longitudinal grain direction of the veneers parallel to the length of specimens. Internal bond strength (IB) was determined according to EN 319 (1993c) (5 specimens per board, n = 10). Thickness swelling (TS) was measured after 24 h immersion in water at 20°C according to EN 317 (1993d) (5 specimens per board, n = 10). During TS testing, the water absorption (WA) was assessed after 24 h immersion in water.

Vertical density profiles (VDP) and peak density (PD)

Vertical density profiles (VDP) of each VSB was recorded with an x-ray densitometer (DAX, Fagus-Grecon GmbH & Co. KG, Alfeld, Germany). The specimen size was 50 × 50 × 12 mm^3. The scanning speed of the x-ray device was set to 1 mm s^{-1} with a resolution of 0.02 mm. Peak density (PD) is defined as the highest density point(s) of the panel within the VDP, which is typically located close to the panel surfaces (Jin et al. 2009a). Five specimens per board (n=10) were used to assess VDP and PD.

Determination of MOR and MOE of veneers

Kiri and birch veneers were cut into strips with a dimension of 100 mm length and 10 mm width. The cutting procedure ensured that the specimens of the two categories are always adjacent to minimize the variation. All veneer strips were conditioned to constant mass at 20°C and 65% relative humidity. Bending strength (modulus of rupture, MOR and modulus of elasticity, MOE) was determined according to EN ISO 178 (2006) (Table 2).

Table 2

MOR and MOE of kiri and birch veneers (n = 10 ± SD).

Type of veneers	MOR [N mm^{-2}]	MOE [N mm^{-2}]
Kiri	36 ± 12	3284 ± 1248
Birch	94 ± 10	7898 ± 741

Results and discussions

Mechanical properties

All VSBs variants with beech and kiri strands showed a significantly higher modulus of rupture (MOR) and modulus of elasticity (MOE) than reference SBs (Fig. 1, 2) except variants with birch veneer located inside panels and with a beech core and birch veneers (Fig. 2a). The higher bending strength of VSBs is attributed to the panel structures and force distribution. In bending, maximum strain and stress occur in the surface layers: the top layer is compressed and the bottom layer is elongated. As in compression a larger deformation occurs, a board usually breaks due to tensile failure of the bottom layer. The longitudinal grain direction of veneers was parallelly arranged with the length of the specimens. In this orientation, the veneer at the bottom side has a higher tensile strength and thus enhances the bending strength of the composite. The increase in strength of the SBs might be also related to the higher tensile strength of veneers itself compared to the strands. Hence, MOR and MOE of SBs might be considerably enhanced by using veneers in the surface layers at different locations. For kiri core layers and kiri veneers, no significant difference in MOR was observed between locations of veneers on the board surface (Table 1C) and 3 mm under the surface (Table 1B). On the other hand, the MOE of kiri but not of beech VSBs with birch veneers located on the panel surface was higher than that with veneers located inside the panel (Fig. 2b). With respect to panels with kiri veneers, no significant difference in MOE was observed between VSBs with veneers located on the inside and surface of the panel – both for kiri and beech cores.

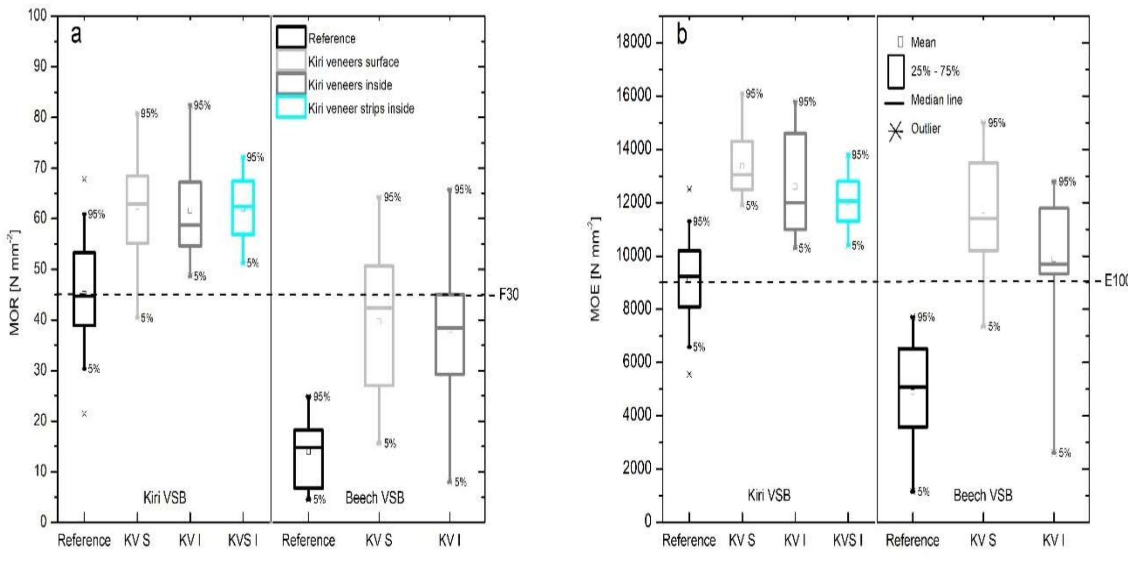


Fig. 1. (a) Modulus of rupture (MOR); (b) modulus of elasticity (MOE) of kiri and beech strand boards without veneering (reference), with kiri veneers on the surface (KV S) as well as with kiri veneers (KV I) and kiri veneer strips (KVS I) 3 mm below the surface (inside) at target density 500 kg m^{-3} (n = 14). *Horizontal dashed lines* indicate the minimum requirement of bending strength for class F 30 of plywood and bending modulus for class E 100 of plywood according to EN 636 (2015). The components of the boxplots are indicated in Fig. 1 b; the whiskers show the 5% and 95% quartile.

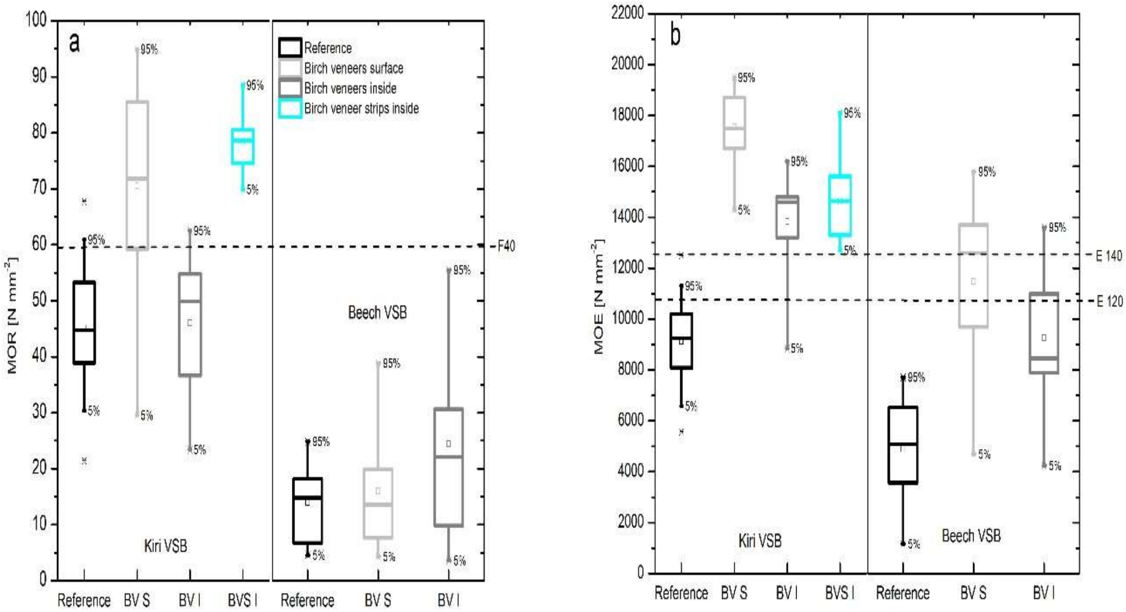


Fig. 2. (a) Modulus of rupture (MOR); (b) modulus of elasticity (MOE) of kiri and beech strand boards without veneering (reference), with birch veneers on the surface (BV S) as well as with birch veneers (BV I) and Birch veneer strips (BVS I) 3 mm below the surface (inside) at target density 500 kg m^{-3} (n = 14). *Horizontal dashed lines* indicate the minimum requirement of bending strength for class F 40 of plywood and bending modulus for class E 120 and E 140 of plywood according to EN 636 (2015). The whiskers show the 5% and 95% quartile.

MOR and MOE of VSBs and SBs containing kiri strands were much higher than those containing beech strands (Fig. 1, 2). Furthermore, birch veneers were delaminated from the beech core layer and hardly enhanced MOR of VSBs, despite increased MOE. As previously shown, kiri strands or particles cause high strength properties of SBs (Pham et al. 2019) and particleboards (Nelis and Mai 2019a) due to a high compaction ratio resulting in larger strand/particle contact area compared to high-density species such as beech. MOE of kiri VSBs containing birch veneers was significantly higher than those containing kiri veneer. Interestingly, the 5%-quantile of MOE of kiri VSBs with birch veneer on the surface was above the highest class E 140 threshold (with a minimum value of 12,600 N mm^{-2} for the 5% quantile) of the plywood standard EN 636 (2015). In addition, VSB variants with kiri strands containing kiri veneers and veneer strips meet the MOR for F 30 (45 N mm^{-2}) and MOE for class E 100 (9,000 N mm^{-2}), while variants with birch veneer strips even fulfilled the MOR for F 40 (60 N mm^{-2}) and MOE for class E 120 (10,800 N mm^{-2}) of plywood specification requirements EN 636 (2015). The higher values of the variant with a beech core layer and birch strips inside and the respective variant with birch veneers inside (Fig. 2) can be attributed to a more flexible structure caused by the strips, which allow a better adaptation to the strand surfaces and thus larger gluing areas.

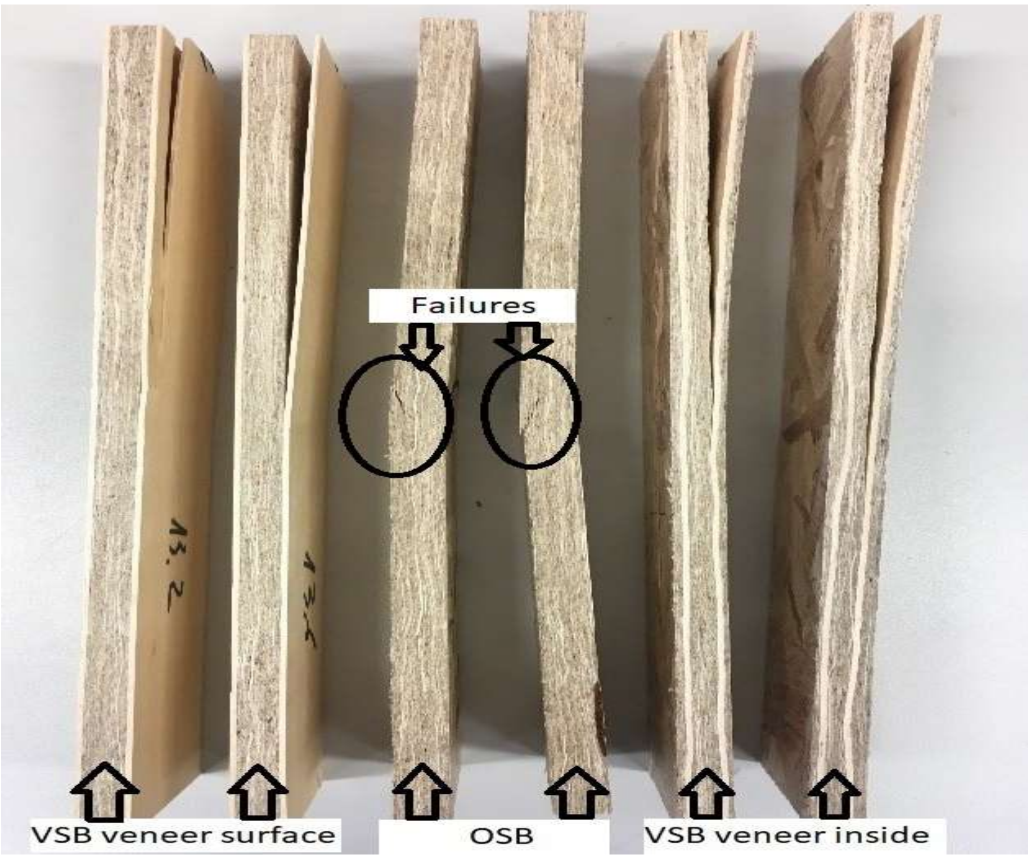


Fig. 3. Failure modes of bending strength specimens after testing (2 left: VSBs with birch veneer on the surface; 2 middle: SB references; 2 right: VSBs with birch veneer 3 mm below the surface).

The failure of VSBs during MOR testing occurred at the interface of strands and veneers and resulted in delamination (Fig. 3), while the SBs directly failed at the bottom of the specimen (tensile side). Thus, MOR and MOE were considerably influenced by the surface layer of the panels. This shear failure mode of VSBs can be attributed to the weak bonding strength at the interface between strands and veneers because no resin was separately applied to the veneers. The longitudinal grain direction of veneers was arranged parallel with the length of the specimens, which causes higher MOR than perpendicular grain orientation (Ashby and Gibson 1997). The grain angle of the face veneer is also one of the critical factors influencing the MOE of the panel (Hu et al. 2005).

The MOR and MOE of birch veneer were significantly higher than those of kiri (Table 2) attributable to the higher density of birch veneer. This effect may explain the higher bending strength of VSB with a kiri core layer and birch veneers despite the stronger compression of the kiri veneers during hot pressing. As reported previously, the bending strength of the panel considerably depended on the stiffness and density of the surface layer (Wong et al. 1999; Nemli and Demirel 2007). On the other hand, the MOR of VSB with a beech core layer was higher when planked with kiri veneers compared to birch veneers. This finding is attributed to the fact that no adhesive was applied to the veneers but only to the strands. Due to the higher compaction of the kiri strands, the area, that is available for gluing, is larger than for beech strands and enough to exploit the higher bending strength of the birch veneers. In contrast, the beech strands are much less compressed than kiri strands, which results in a much smaller gluing area. Kiri veneers, therefore, cause a higher bending strength on the beech core because they are more easily deformed and compressed than birch veneers and cause a larger gluing area. This means that on the kiri core the higher stiffness and bending strength of the birch veneers had a larger influence than the stronger compressibility of the kiri veneers. On a kiri core, it was *vice versa.*

Internal bond strength (IB) and pull-off strength (PoS)

SBs with kiri strands exhibited considerably higher internal bond strength (IB) than those based on beech strands (Fig. 4), which can be attributed to the higher compaction ratio and the larger contact area between strands. Similar results were previously found for SBs (Pham et al. 2019) and three-layered particleboards (Nelis and Mai 2019b). Determining IB of the VSB specimens was impossible because all failed at the interface between the veneers and strands because of a lack of resin added to the veneers. Thus, the values assessed for VSBs in the test indicate the pull-off strength (PoS) of the veneers rather than IB of the core layer. As observed for MOR, the PoS was higher for VSBs with a kiri core layer than for the beech core due to higher compression and deformability (ductility) on the entire core layer surface, which leads to larger gluing areas. For the same reason, kiri veneers caused higher PoS on a kiri core than birch veneers, while no clear effect was apparent on beech cores. Additionally, the PoS of VSBs with birch veneer strips on kiri cores was much higher than those with complete birch veneers (Fig. 4b), most probably due to the easier adaption to the strands' surfaces resulting in a better bonding quality.

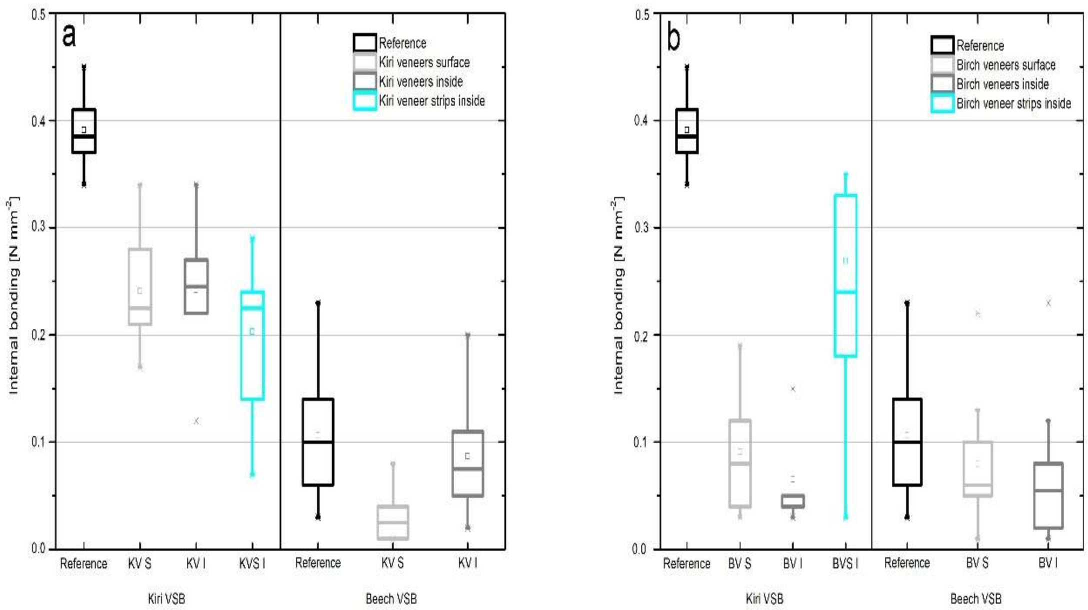


Fig. 4. Internal bonding strength (IB) or pull-off strength of kiri and beech strand boards without veneering (reference), with kiri (a) and birch (b) veneers on the surface (KV S/ BV S) as well as with veneers (KV I, BV I) and veneer strips (KVI S, BVS I) 3 mm below the surface (inside) at target density 500 kg m^{-3} (n = 10).

Water-related properties

Thickness swelling (TS) and water absorption (WA) of VSBs with kiri veneers after 24 h immersion in water was slightly but not significantly lower than that of SBs (Fig. 5). A significant difference was also not observed between kiri and beech VSBs and no apparent effect of veneer localisation on TS of panels was found. The kiri veneers did not peel off the core layer during the test, while the birch veneers of respective VSBs become detached after 2 h immersion. The reason for this behaviour is again attributable to the better gluing when kiri strands and veneers are involved as described with respect to the mechanical properties.

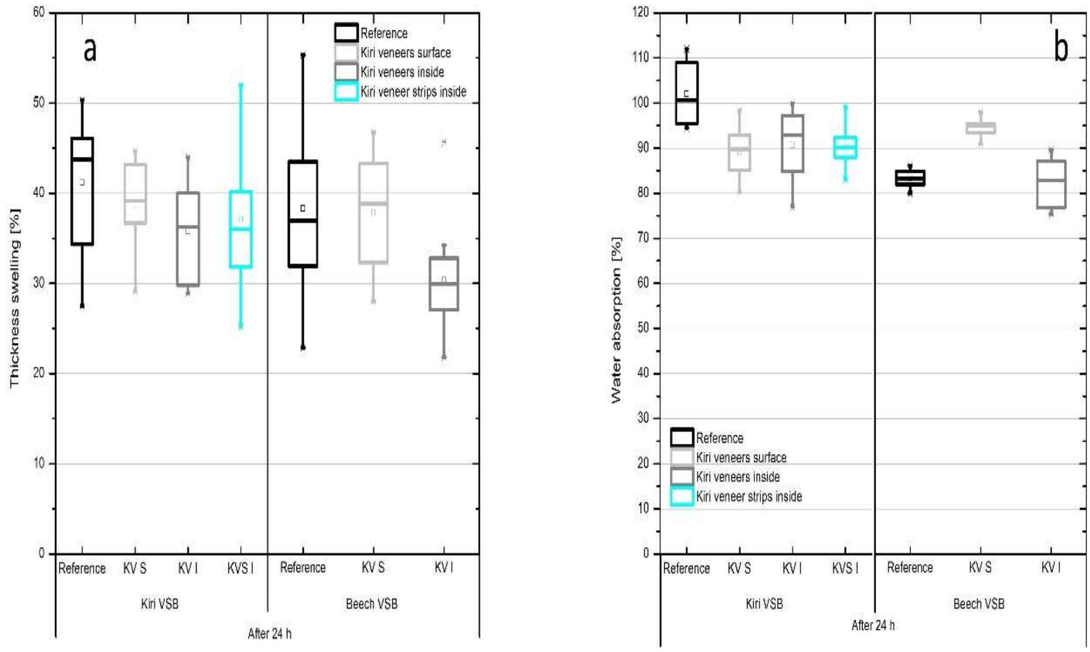


Fig. 5. (a) Thickness swelling (TS); (b) water absorption (WA) after 24 h immersion in water of kiri and beech strand boards without veneering (reference), with kiri (a) and birch (b) veneers on the surface (KV S/ BV S) as well as with veneers (KV I, BV I) and veneer strips (KVI S, BVS I) 3 mm below the surface (inside) at target density 500 kg m^{-3} (n = 10).

Vertical density profile (VDP) and peak density (PD)

The vertical density profile (VDP) is considered as one of the key factors directly influencing the properties of OSB (Gu et al. 2005; Xu 1998; Xu and Suchsland 1997; Jin et al. 2009b). Most notably bending strength but also IB is significantly affected by the VDP (Kelly 1977). The VDP may vary considerably depending on the particle configuration, moisture distribution, pressing temperature and pressure, as well as reactivity of the resin used. All VSBs and SBs variants exhibited a 'U-shape' vertical density profile, which is determined by the factures mentioned above (Wong et al. 1999; Chen et al. 2010). VSBs with veneers on the surface exhibited a sharp linear (birch veneer due to higher density than kiri) to more gradually decreasing, slightly concave (kiri veneer) descent towards the core layer, while SBs exhibit a convex descent of the surface layer (Fig. 6 a). VSBs with veneers located 3 mm below the board surface also showed higher surface densities than SBs (Fig. 6 b). The VSBs with birch veneer strips display a clear increase in density with a peak at 3 mm below the surface, while VSBs with kiri veneer strips did not show such a peak. This is attributed to the different densities of the respective veneers.

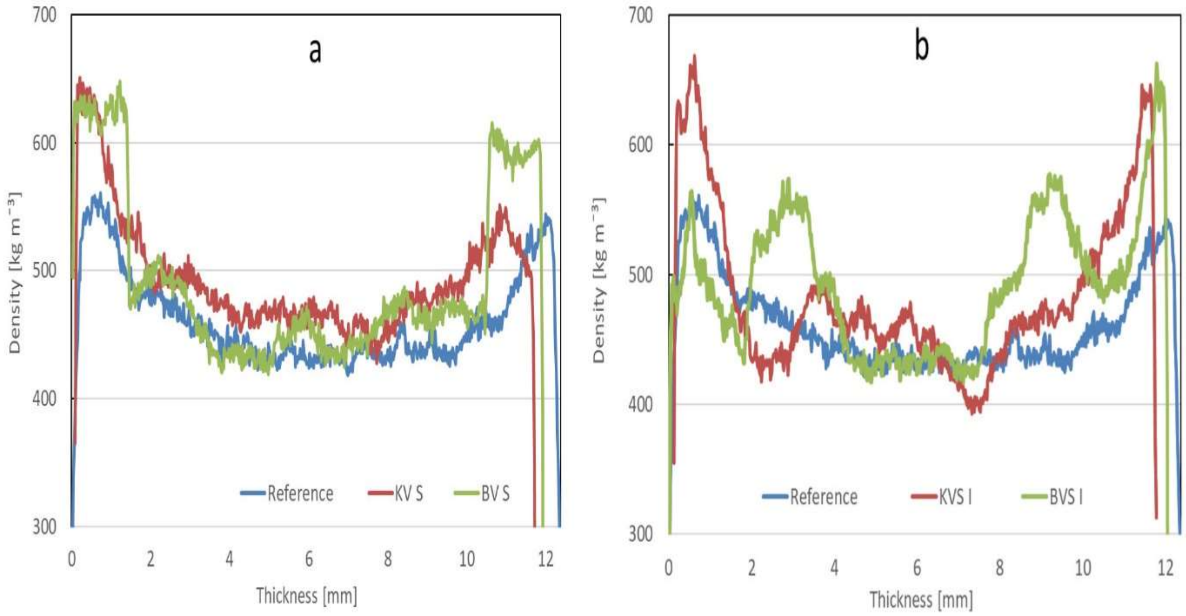


Fig. 6. Representative vertical density profile of kiri SBs and VSBs with birch and kiri veneers at target density 500 kg m^{-3}; (a) SBs without veneering (reference) and with kiri and birch veneers located on the surface of the panel (KV S/ BV S); (b) veneer strips located 3 mm below the surface (KVS I/ BVS I).

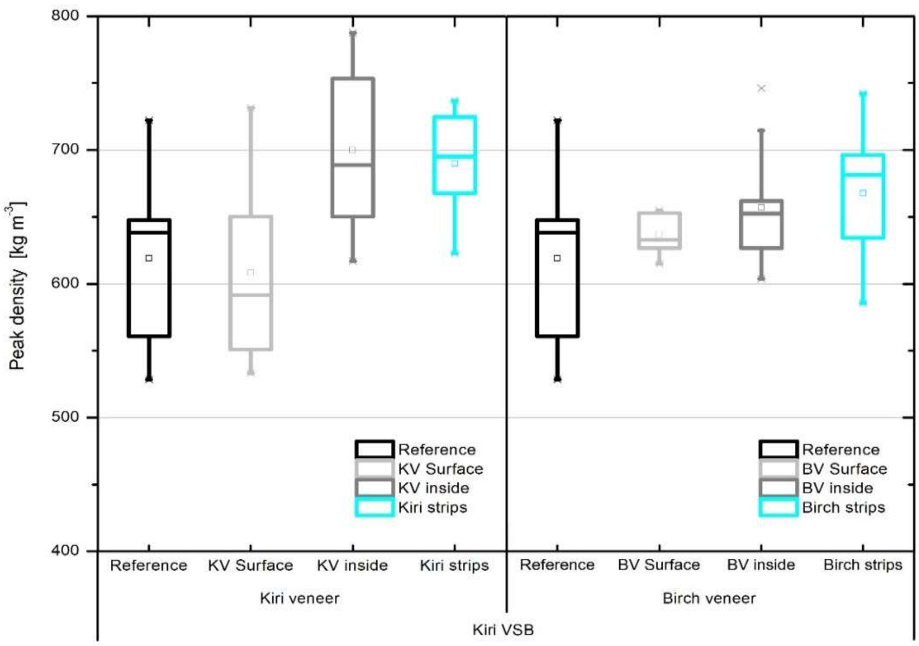


Fig. 7. Peak density of kiri strand boards with birch and kiri veneers on the surface and 3 mm below the surface (inside) at target density 500 kg m^{-3} (n = 10).

One of the essential characteristics of the vertical density profile is the peak density, which influences the MOR and MOE (Wong et al. 1999). The highest bending MOE occurs, if the peak density is located close to but not right at the surface of the panel (Xu 1998). In general, VSBs showed a higher peak density than SBs (Fig. 7). The linear to steep gradients towards the core layer of VSBs with birch veneer on the surface is assigned to the high density of birch, while the concave descent of VSBs with kiri veneer is due to the low density of kiri, which was considerably compressed (about 4 times compared to its initial thickness). In addition to the different force distribution through the veneers (see above), the high PD of VSBs with birch veneer on the surface explains the high MOR and MOE of VSBs (Fig. 2). In addition, mean density of various layers was shown in table 3.

Usually, the VDP can be used to predict the IB of a panel because the failure occurs at the site of the lowest density (Nemli and Demirel 2007). The VSBs, however, failed at the interface between the veneers and the core layer, which cannot be predicted from the VDP.

Table 3

Mean density of surface and core layers obtained from mean density profiles of VSBs and two variants of SBs (n = 10).

	Mean density of various layers (kg m^{-3})						
	Reference	Kiri strips	Birch strips	BV* surface	BV* inside	KV* surface	KV* inside
Upper surface	546	552	561	553	578	516	555
Core layer	486	483	508	485	503	498	491
Lower surface	515	540	557	547	574	512	556

*BV: birch veneer; KV: kiri veneer

Conclusion

VSB with kiri and birch veneers at a target density of 500 kg m^{-3} can be produced in a laboratory press without technical problems. Veneering of strand boards significantly enhances the MOR and MOE without increasing the density compared to "pure" strand boards. The increase in strength of the VSBs by the reinforcement of veneers is consistent, particularly with the high density of birch veneers. The strengthening of VSBs with veneers on the surface is higher than when the reinforcement is located 3 cm below the surface. In this approach, the PF adhesive was only applied to the strands but not on the veneers. Strength testing, however, revealed that this approach leads to failure of the bonding between the strand core and the veneers and, thus, to the delamination of the veneers. Delamination is the more pronounced, the higher the density of the veneer and the stiffer it is. Thus, an additional application of glue is necessary but will make the production process more laborious and costly. Still, the results principally show that lightweight VSBs can be a product that might even compete with plywood for light-weight constructions but the production requires less effort than plywood production.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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Publication IV

Influence of compaction ratio and resin molecular weight on properties of strand boards and resin efficiency

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Influence of compaction ratio and resin molecular weight on the mechanical and water-related properties of strand boards

Abstracts

This study assesses the influence of the compaction behavior of wood strands and the bonding performance of a low- and high-molecular-weight phenol-formaldehyde (PF) resin on the mechanical and water-related properties of strand boards (SBs). Strands of three wood species with increasing density - kiri (*Paulownia tomentosa)*, pine (*Pinus sylvestris* L.) and beech (*Fagus sylvatica* L.) - were used to produce SBs at a density of 500 kg m^{-3} with low (LMW) and high molecular weight (HMW) PF. With LMW PF resin, the internal bond strength (IB) of SBs from pine and beech was ten times lower than that of kiri SBs, probably because of insufficient amounts of resins on the surface of pine and beech strands. HMW PF resin induced clearly higher IB of pine and beech SBs than LMW PF resin, while in the case of kiri SBs LMW PF resin caused higher IB than HMW PF resin. Bending properties and screw withdrawal resistance increased with compaction ratio (CR) for both resins. A high CR is assumed to be associated with more adhesion contact points between the strands. Only in the case of the low-density kiri strands, this led to higher strength and dimensional stability with LMW PF than with HMW PF.

Keywords: resin molecular weight; strand boards; *Paulownia tomentosa*; compaction ratio

Introduction

Wood species, the resulting bulk density of particles, board density, compaction ratio and wood surface chemistry are considered as key factors influencing the mechanical properties of wood-based panels because these factors influence the binder consumption (Vital et al. 1974; Wang and Winistorfer 2000). For a given particleboard density of 500 kg m^{-3}, the use of low-density wood species led to a higher strength than using beech particles of higher bulk density (Nelis and Mai 2019a). Strength differences between kiri and beech particleboards, however, decreased with increasing panel density to 650 kg m^{-3} (Nelis and Mai 2019a, b). The bending strength of urea-formaldehyde-bonded particleboards increased with increased wood density; however, (Vital et al. 1974) found that a low compaction ratio exerted higher IB than those of high compaction ratio. The authors explained this with an increasing amount of crushing and flake damage occurring at the high-compression level.

Resin penetration into the structure of the wood elements as well as its distribution over the surfaces has a significant influence on the adhesive bond performance of wood-based composites (Kamke and Lee 2007). Several studies have investigated the bonding performance of PF resin. Sernek studied PF bond performance with solid wood of yellow poplar and southern pine, and deeper penetration took place in yellow poplar than southern pine (Sernek 2002). The study, however, did not reveal any correlation between bond performance and resin penetration. A previous study reported on the effect of temperature and humidity on PF resin bonding of two flakes in a lap-shear configuration (Wang et al. 1995) but did not show an influence of these factors on resin penetration. Finer resin droplets and longer blending time reportedly result in a continuous and uniform distribution of resin, thus improving bonding strength in particleboards (Klauditz 1962). The small size of resin droplets on wood particles after spraying might be an indicator of a more homogeneous distribution to create continuous bonding lines (Lehmann 1970; Altgen et al. 2019). Steam-injection pressing was suggested to increase the penetrability of PF into yellow poplar strands (Johnson and Kamke 1994); however, the influence of the molecular weight of PF on the mechanical properties of particleboards was not studied.

The literature on the effect of compaction ratio in relation to the resin molecular weight on strand board properties is limited. Resin distribution and penetration into the flakes play a crucial role in the performance of wood-based panels; previous studies provided general knowledge about fundamental theories of resin distribution (Dai et al. 2005; Altgen et al. 2019). Hence, it is necessary to study the effect of resin molecular weight with respect to the compaction ratio of the panel. This effect is associated with the resin distribution and penetration to form contacts for bonding between wood particles. PF resins of low molecular weight require a longer pressing time for curing at a given press temperature (Pham et al. 2019). The aim of this study is to determine the effect of the resin's molecular weight and the compaction ratio on the hygro-mechanical properties of SBs.

Materials and methods

Strands production

Kiri trees were harvested near Mannheim (Germany) and provided by the company WeGrow GmbH (Tönisvorst, Germany). Logs of Kiri (*Paulownia tomentosa* (Thunb.) Steud.), pine (*Pinus sylvestris* L.) and beech (*Fagus sylvatica* L.) exhibited a mean density of 300, 510, 720 kg m^{-3}, respectively, and diameters of 12-20 cm (kiri) and 30-35 cm (pine and beech). Manually debarked 2-meter-long logs were firmly held by a hydraulic clamp and moved towards a knife ring flaker PZUL 8-300 (Pallman Company, Zweibrücken, Germany). For pine and beech logs, the feeding speed was 40 mm s^{-1}, while it was reduced to 36 mm s^{-1} for kiri logs because faster feeding speed led to a high amount of damaged strands. Target strands were up to 110 mm long, 0.5-1.0 mm thick and 10-50 mm wide. These strands were further sorted out by a sieving shaker. Three fractions of strand were obtained categorized by width: 10-30 mm, 30-50 mm and bigger than 50 mm; sieve fractions smaller than 10 mm were discarded as fines. After sorting, the strands were immediately dried at 70°C to a moisture content of 3- 5% in a drying oven. In order to keep the moisture content low, the strands were stored in plastic bags until panel manufacturing.

Panel manufacturing

Six variants of single-layered SBs were produced at a target density of 500 kg m^{-3} with the three wood species kiri, pine and beech. The strand fractions were mixed in a ratio of 4:5:1 (10-30 mm : 30-50 mm : >50 mm width). The strands were resinated with phenol-formaldehyde (PF) resin in a drum blender with a rotating speed at 30 rounds min^{-1}. Two types of PF resins with low (LMW) and high molecular weight (HMW) were used as adhesive with the amount of 10% stock solution based on the oven-dry weight of strands. The LMW PF resin was provided by Surfactor GmbH (Schöppenstedt, Germany) with an average molecular weight (M_w) from 400 to 500 g mol^{-1} and 53% solid content. HMW PF resin was provided by Prefere resins Germany GmbH (Erkner, Germany) with an average M_w from 1000 to 1200 g mol^{-1} and 70% solid content. The resinated strands were formed into a mat (450 × 450 mm^2) in a cold pre-press before hot-pressing at 140 °C for 90 s mm^{-1} using metal bars stops to ensure a panel target thickness of 12 mm. In total, 24 panels were produced, which consists of four SBs for each of six variants (Table 1).

Table 1 Parameters of panel manufacturing

Panel dimensions	450 × 450 × 12 mm^3
Wood species	kiri, pine, beech
Target mat moisture content	8-10%
Resin type	LMW PF (M_w=400-500 g mol^{-1}; solid content
	HMW PF (M_w=1000-1200 g mol^{-1}; solid content
Resin mass content	10 wt% based on the oven-dry weight of strands
Blender rotation speed	30 rounds min^{-1}
Press temperature	140°C
Press time	90 s mm^{-1}
Replicates	4

Physical and mechanical properties

All specimens were conditioned at 20 °C and 65 % relative humidity for at least two weeks to reach equilibrium moisture content. The density of all specimens was determined according to EN 323 (1993a). Bending strength (modulus of rupture, MOR) and modulus of elasticity in bending (MOE) according to EN 310 (1993b) (7 specimens per board, n = 28), and internal bond strength according to EN 319 (1993c) (5 specimens per board, n = 20). Screw withdrawal resistance (SWR) was determined following EN 320 (1993d) but SPAX universal screws (SPAX international GmbH & Co. KG, D-58256 Enneptetal, Germany) were used with dimensions of 4 mm (d) × 35 mm (l) (5 specimens per board, n = 10). All test specimens were randomly selected by their density variation (500 kg m^{-3} ± 10%). All tests were performed using a universal testing machine (ZwickRoell GmbH & Co. KG, Ulm, Germany). Thickness swelling was measured after 24 h immersion in water at 20 °C according to EN 317 (1993e) (5 specimens per board, n = 10). Also, the water absorption (WA) was assessed after 24 h immersion in water.

Table 2 Compaction ratio of kiri, pine, beech strand at board target density 500 kg m^{-3}

Wood species	Compaction ratio at panel density of 500 kg m^{-3}
Kiri	500/300 = 1.7
Pine	500/510 = 1.0
Beech	500/720 = 0.7

Results and discussion

Internal bond strength (IB)

Kiri SB variants containing LMW PF displayed a significantly higher internal bond strength (IB) than those with HMW PF. In contrast, IB values of pine and beech SB variants containing LMW PF were considerably lower than those of HMW PF (Fig. 1a). IB values of pine and beech SBs containing LMW PF were below 0.1 N mm^{-2} and indicated insufficient bonding. Respective values of SBs containing HMW PF were about 0.4 and 0.1 N mm^{-2}, which is significantly higher than those of panels bonded with LMW PF. The higher IB of these panels can mainly be attributed to sufficient HMW PF adhesive between the strands due to a minor degree of penetration into the wood. It was previously shown for kiri SBs with lower densities (300 and 400 kg m^{-3}) that the IB was higher than that of pine SBs due to the higher compaction ratio of kiri panels (Pham et al. 2019, 2021). At panel density of 500 kg m^{-3}, however, the influence of the compaction ratio was lower resulting in similar IB for kiri and pine SBs. These findings are confirmed in this study. Furuno reported that LMW PF resins (number average molar mass (M_n) of 290 and 470 g mol^{-1}) penetrated easily into the wood cell walls, while HMW resin (M_n of 1000-1200 g mol^{-1}) mostly covered the cell wall lumens (Furuno et al. 2004). LMW PF in this study must mainly act as an adhesive on the strand surface rather than a modifying agent; thus excessive penetration might result in too little adhesive at the interface between the strands, which would lead to starved bond areas (Kamke and Lee 2007). Stephens and Kutscha pointed out that the highest mechanical strengths of aspen flake boards are obtained by combining HMW and LMW PF resins (Stephens and Kutscha 1986). The low IB of pine and beech SBs in this study can thus be assigned to the lack of available adhesive on the strand surfaces because of excessive penetration of LMW PF.

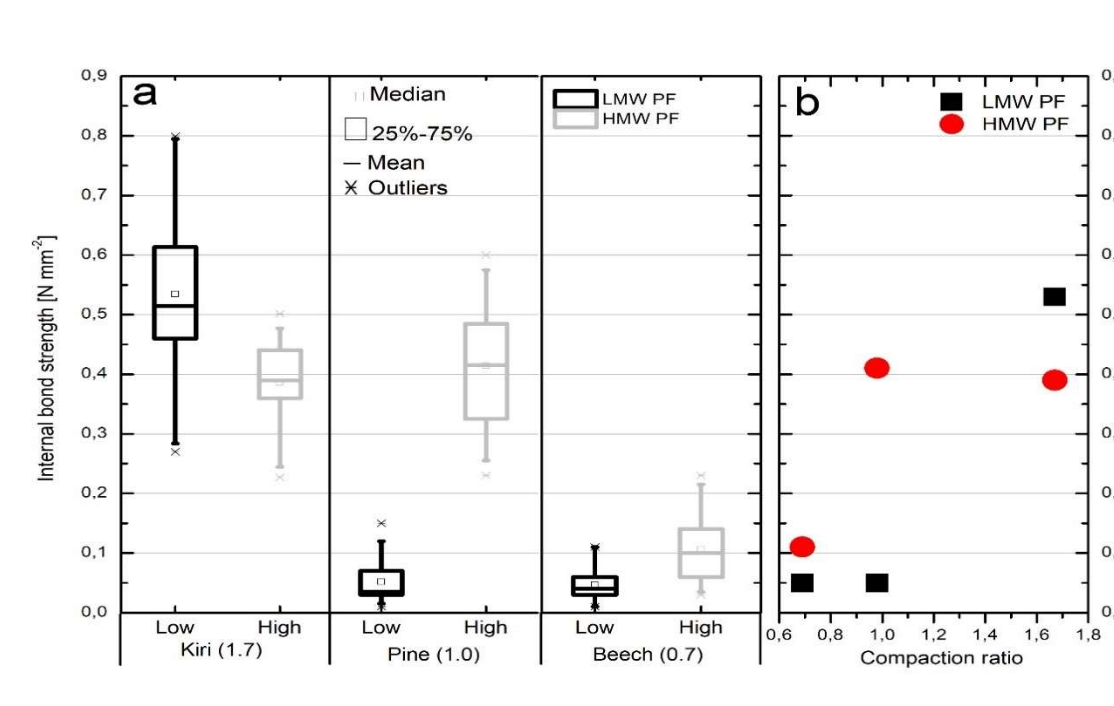

Fig. 1 (a) Internal bond strength (IB) of kiri, pine, beech SBs at 500 kg m^{-3} target density (n = 20); "Low" indicates LMW PF resin; "High" HMW PF resin; and (b) its relation to the compaction ratio depending on resin's molecular weight. The components of the boxplots are indicated in Fig.1a; the whiskers show the 5% and 95% quartile.

Fig. 1b displays the relationship between compaction ratio (CR, Table 2) and its influence on the IB depending on the resin's molecular weight. It shows that LMW PF resin only induces high IB at the highest CR of 1.7. In contrast, HMW PF resin can cause considerable IB at CR of 1.0 and 1.7 but it is clearly lower at 0.7. In contrast to this study, Vital et al. found a low IB at a higher compaction ratio but attributed this finding to an increasing amount of crushing and flake damage at high compaction (Vital et al. 1974). Kim (Kim et al. 1994) studied the effect of PF resins of higher molecular weight than those used in this study. At a target density of 721 kg m^{-3} at 4% PF content, the high molecular weight PF resin (M_w of 3487 g mol^{-1}) resulted in a lower IB than the low molecular weight PF resin (M_w of 2179 and 2289 g mol^{-1}). This was attributed to limited resin flow during hot pressing because of too high molecular weight. These findings, however, cannot be compared to this study.

Modulus of rupture (MOR), and modulus of elasticity (MOE)

MOR and MOE of SBs increased with decreasing strand density (Fig. 2) and, thus, the compaction ratio (Fig. 3); kiri variants displayed significantly higher MOR and MOE than those of pine and beech strands (Fig. 2, 3). This result is in accordance with a previous study which showed that MOR and MOE of pMDI bonded kiri SBs were higher than those of pine SBs at a density of both 300 and 400 kg m^{-3} (Nelis and Mai 2019b; Pham et al. 2019). While the PF resin's molecular weight did not show apparent influence on MOE, MOR of pine and beech SBs containing HMW PF was considerably higher than of those containing LMW PF. This effect was already reported by Haygreen and Gertjejansen (Haygreen and Gertjejansen 1971) with respect to particleboards. In contrast, kiri SBs containing LMW PF and HMW PF did not exhibit differences in MOR. The increase of MOR and MOE with CR (Fig. 3) was previously shown for SBs bonded with pMDI (Pham et al. 2019), pMDI-bonded particleboards (Wong et al. 1999) and urea-formaldehyde-bonded particleboards (Vital et al. 1974; Nelis and Mai 2019b, a). At a low CR (pine and beach), the MOR of SBs containing LMW PF is significantly lower than those of HMW PF. As with IB, this is attributed to the fact that less resin is available on the strand surface when LMW PF is used as the binder. Due to lower densification, this results in smaller bonding areas than when using HMW PF. Differences between LMW and HMW PF were less pronounced at the highest CR (kiri).

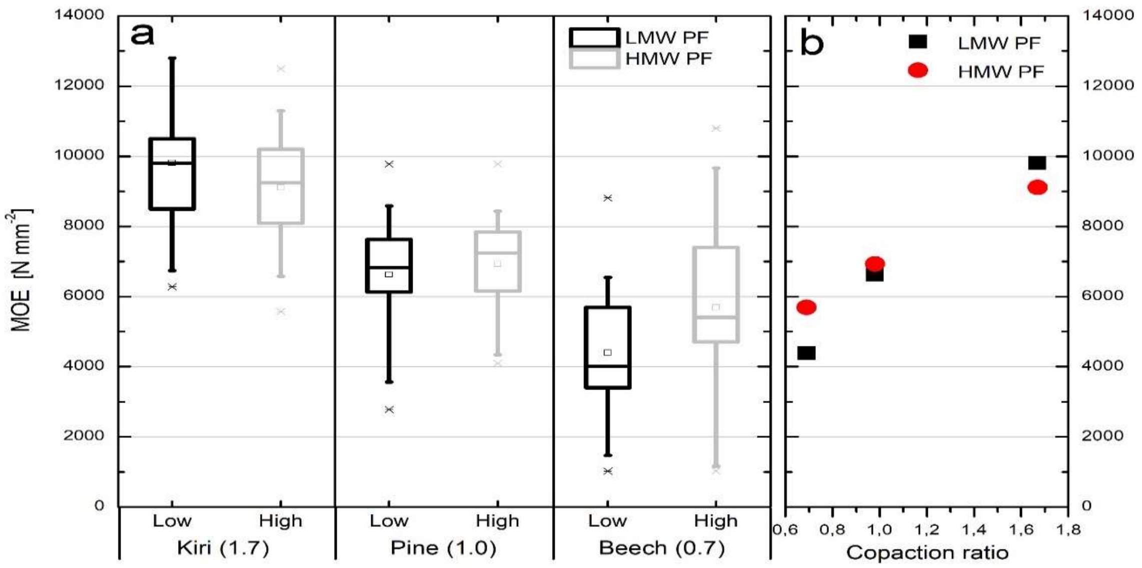

Fig. 2 (a) Modulus of rupture (MOE) of kiri, pine, beech SBs at 500 kg m^{-3} target density (n = 28); (b) its relation to the compaction ratio depending on resin´s molecular weight. "Low" indicates LMW PF resin; "High" HMW PF resin. The components of the boxplots are indicated in Fig.1a; the whiskers show the 5% and 95% quartile.

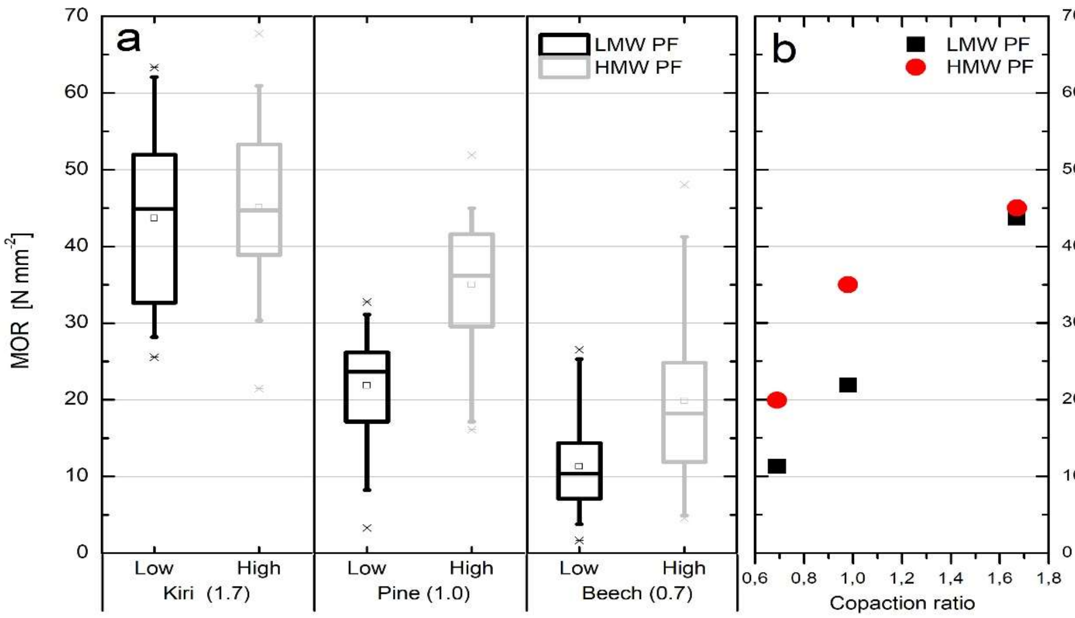

Fig. 3 (a) Modulus of rupture (MOR, mean value) of kiri, pine, beech SBs at 500 kg m^{-3} target density (n = 28); (b) its relation to the compaction ratio depending on resin´s molecular weight.

"Low" indicates LMW PF resin; "High" HMW PF resin. The components of the boxplots are indicated in Fig.1a; the whiskers show the 5% and 95% quartile.

Screw withdrawal resistance (SWR)

For all variants of face orientation of the screw, SWR did not indicate significant differences irrespective of the wood species and the resin's molecular weight (Fig. 4a). This result was expected because the screws were positioned perpendicular to the strand surface, so the bonding quality of SBs did not contribute to SWR. For the side orientation of the screw, SWR revealed a similar trend as observed for IB. The highest SWR was observed for LMW-PF-bonded kiri SBs, while LMW-PF-bonded beech SBs exhibited the lowest SWR (Fig. 4b). The SWR of kiri SBs showed only a minor difference between the two resin types but there was a trend for higher SWR with respect to LMW-PF-bonded kiri SBs. In general, the SWR of side orientation depends on IB because both properties reflect the density and the bonding quality in the core of the panels, the site of lowest strength.

Correspondingly, both IB as well as SWR (side orientation) of kiri and pine SBs containing HMW PF were not significantly different. As for IB, the low SWR of pine and beech SBs containing LMW PF might be attributed to the lack of adhesive on the strand surfaces due to strong resin penetration and concomitant low CR.

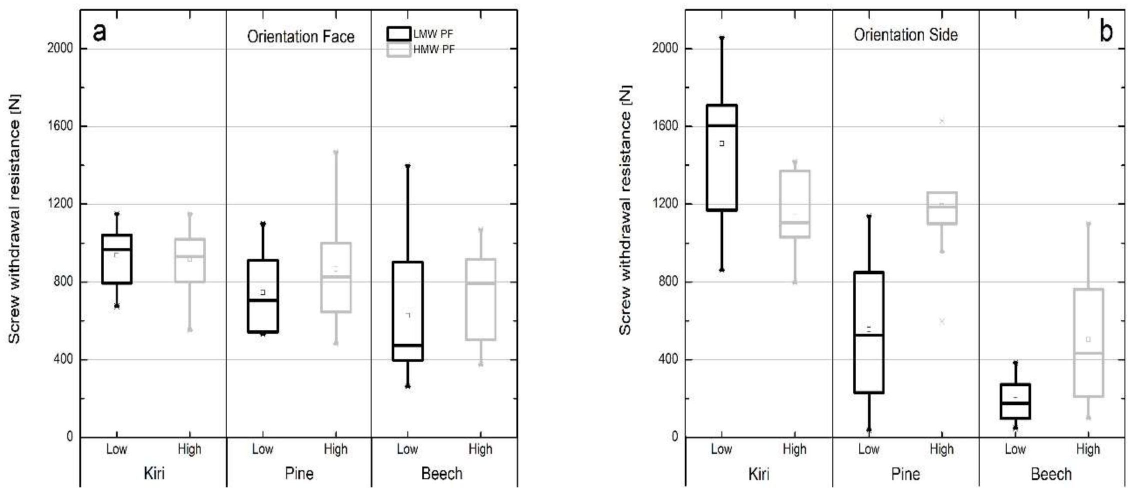

Fig. 4 Screw withdrawal resistance (SWR) of kiri, pine, beech SBs at 500 kg m^{-3} target density with the face (a) and side (b) orientation of the screw (n = 10); "Low" indicates LMW PF resin; "High" HMW PF resin. The components of the boxplots are indicated in Fig.1a; the whiskers show the 5% and 95% quartile.

Water-related properties

Statistical analysis revealed that for all variants, except beech SBs, thickness swelling (TS) was considerably affected by resin molecular weight (Fig. 5). Kiri SBs bonded with LMW PF exhibited lower TS than those bonded with HMW PF. The opposite was true for pine SBs. Beech SBs containing LMW PF and HMW PF showed similar TS values. With respect to LMW PF, TS after 2 h of kiri SBs was significantly lower than that of pine and beech SBs; however, the differences decreased after 24 h. The lower TS of kiri SBs is attributed to better adhesion (indicated by higher IB) with LMW PF. Previous studies, however, revealed that pMDI-bonded kiri SBs with a high compaction ratio resulted in higher TS compared to pine panels with lower CR (Pham et al. 2019). This might be explained by the better gluing properties of pMDI as high adhesive strength (indicated as high IB) leads to lower TS and thus offsets the effect of compaction and resulting spring-back on TS. Particularly, pine and beech SBs bonded with LMW PF showed poor IB and thus high TS. Poor water-related properties of LMW-PF-bonded pine SBs were observed by Wan and Kim (Wan and Kim 2007), who impregnated southern pine strands with LMW PF to enhance dimensional stabilization. Regarding HMW PF, TS of kiri SBs was considerably higher than that of pine panels as reported previously (Pham et al. 2019).

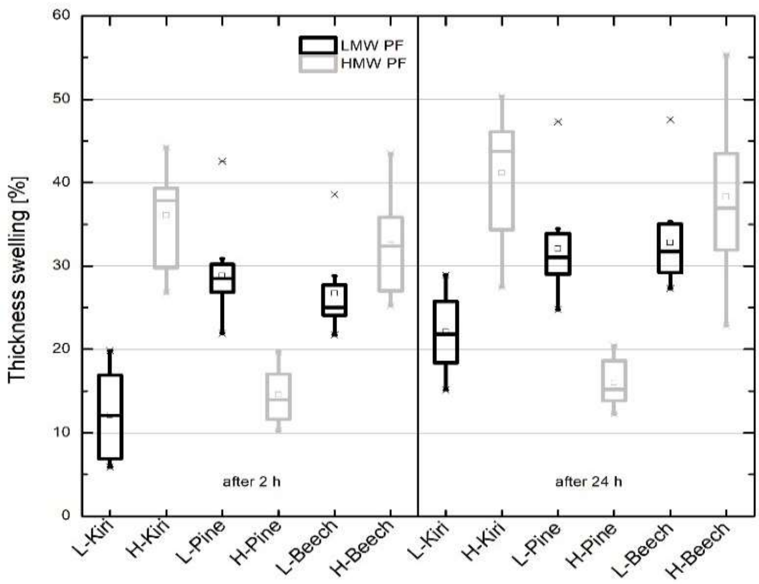

Fig. 5 Thickness swelling (TS) of kiri, pine, beech strand boards at 500 kg m^{-3} target density after 2 h and 24 h immersion in water (n = 10); "L" indicates LMW PF resin; "H" HMW PF resin. The components of the boxplots are indicated in Fig.1a; the whiskers show the 5% and 95% quartile.

Similar tendencies as described for TS were also observed for water absorption (WA). Kiri SBs containing LMW PF revealed the lowest relative WA (based on the mass of the SBs) after 2 and 24 h immersion in water (Fig. 6). WA of kiri SBs containing LMW PF after 2 h and 24 h immersion was significantly lower than that of respective panels bonded with LMW PF. This is attributed to the lower TS of the latter boards due to better adhesion (higher IB) of the former. Accordingly, pine SBs containing LMW PF exhibited higher WA after 2 h and 24 h than pine SBs with HMW PF due to respective differences in TS. Interestingly, kiri SBs bonded with LMW PF displayed similarly low TS as pine SBs bonded with HMW PF (Fig. 5). After 24 h immersion, the TS of the latter was even lower. WA of pine SBs bonded with HMW PF, however, was significantly higher than that of kiri SBs bonded with LMW PF – almost twice as high. This indicates that WA does not only depend on the swelling behavior determined by the adhesive strength (IB) and spring-back but also on the water penetration into the composite and the wood cell wall. LMW PF might block some penetration pathways for water into voids between the strands and into the cell walls more effectively than HMW PF.

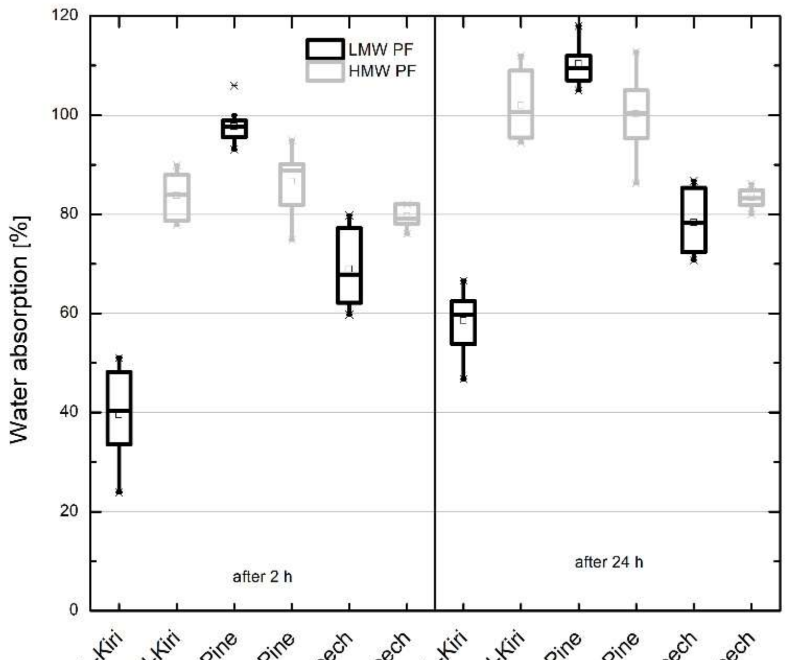

Fig. 6 Water absorption (WA) of kiri, pine, beech strand boards at target density 500 kg m^{-3} after 2 h and 24 h immersion in water (n = 10), “L” indicates LMW PF resin; “H” HMW PF resin. The components of the boxplots are indicated in Fig.1a; the whiskers show the 5% and 95% quartile.

Mode of action of LMW and HMW PF resins

Earlier studies revealed that LMW PF may penetrate the wood cell wall under the conditions of vacuum-pressure impregnation, while only a minor proportion of the molecules in HMW PF can diffuse into the cell wall under these conditions (Stamm and Seborg 1939; Kajita et al. 2004; Kielmann et al. 2018). Accordingly, it is assumed that also in our study a considerable proportion of the LMW PF resin diffused into the wood matrix after spraying and is therefore not available on the surface of the strands. In contrast, HMW PF resin mainly remains on the stands' surface and may cause bonding. During spraying of the resins in a glue-drum, a wiping effect occurs and distributes the resins on the strand surface. Further spreading of the resin occurs during hot-pressing. The LMW PF resin might create a more homogeneous distribution on the strand surface but less resin is available for gluing than in the case of HMW PF. In addition, the application of aerosol spray produces small resin droplets, which improves the mechanical properties of the panels (Lehmann 1970). At a given resin, the use of the plasma pre-treatment of wood particles was suggested to create a high number of small resin droplets, which form a continuous film that indicates a more homogeneous distribution (Altgen et al. 2019). On the other hand, deeper penetration of LMW PF might cause a stronger anchoring of the resin in the strand and thus higher strength properties. This effect only becomes important, if the adhesion area and thickness between the strands is sufficient. The results related to the strength properties of kiri SBs indicate that the advantages of LMW PF, i.e. homogeneous distribution and strong anchoring in the wood surface, lead to higher bonding strength than HMW PF due to the high compaction ratio of the panels (Pham et al. 2019). In the case of pine and beech strands, however, the compaction ratio is not sufficient to generate enough contact areas and adhesive thickness with LMW PF compared to HMW PF. Thus, the adhesion efficiency of HMW PF is higher with HMW PF resins.

Conclusions

Bending strength (MOR) and stiffness (MOE) of SBs increase up to a certain point with increasing compaction ratio (CR), when sufficient adhesive is available on the strand surface. For HMW PF resin, internal bond strength (IB) did not increase at CR above 1. Using LMW PF resin can only enhance IB, screw withdrawal resistance (SWR) and dimensional stability (low swelling rate) at high CR. This means that for the production of low-density SBs with LMW PF resin, wood species should exhibit a very low density – in this study roughly below 300 kg m^{-3}. If this is not the case, the amount of LMW PF resin on the strand surface is not sufficient for causing appropriate bonding between the strands and results in "starved glue lines". This is most probably due to the penetration of the resin into the wood matrix. At appropriately high CR, LMW PF resins do not only enhance the mechanical properties but also improve dimensional stability. This makes these SBs more suitable for moist and outdoor conditions than SBs produced with HMW PF resin.

Acknowledgments

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Disclosure statement

The authors reported no potential conflict of interest.

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Publication V

Effect of pressing time on physical-mechanical properties and formaldehyde emission of strand boards

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Effect of pressing time on physical-mechanical properties and formaldehyde emission of strand boards

Abstracts

In order to reduce the formaldehyde emission of strand boards produced with different molecular weight phenol-formaldehyde resin, pressing time was investigated and its effect on formaldehyde emission. Strand boards (SBs) from kiri wood (*Paulownia tomentosa*) were manufactured with PF resin at a target density of 500 kg m^{-3}. PF resins with low (LWM) and high molecular weight (HMW) were studied with adhesive content of 10% related to strand mass. Physical-mechanical properties of strand boards produced were determined. Internal bond strength (IB) of SBs containing LMW PF was considerably higher than those of HMW PF; however, with increasing pressing time IB decreased significantly. Accordingly, the formaldehyde emission of SBs based on LMW PF was significantly higher than that of panels produced with HMW PF resin; however, the formaldehyde emission generally decreased linearly with the increasing pressing time. No effect of resin molecular weight and pressing time was found for MOR and MOE.

Introduction

Oriented strand boards (OSB) are manufactured in Europe mainly with isocyanate adhesives such as polymeric diphenylmethane diisocyanate (pMDI), whereas the OSB industry in other geographic regions also employs phenol-formaldehyde (PF) adhesives. OSB is considered as lower cost in residential buildings compared to structural plywood; however, OSB swells when exposed to environmental moisture, causing a loss of structural strength (Brochmann et al. 2004). Phenol formaldehyde has been used as waterproof adhesives which are commonly used in structural wood-based panels such as plywood, oriented strand boards, wafer boards, and exterior particleboards (Meyer et al. 1986; Wescott and Frihart 2004). Phenol-formaldehyde resins, which contains oligomeric and polymeric chains as well as monomeric methylol-phenol, free formaldehyde, and unreacted phenol, might only initiate the curing process under heating conditions with the transformation of molecules of various sizes via chain lengthening, branching, and cross-linking to a three-dimensional network (Pizzi 1983; Knop and Pilato 1985; Gardziella et al. 2000; Pizzi and Mittal 2018).

Low molecular weight phenol-formaldehyde (PF) resin has been successful employed to enhance dimensional stability by penetrating and bulking the cell wall (Stamm and Seborg 1941; Ohmae et al. 2002; Rowell 2012). Nevertheless, LMW PF resin contains a higher amount of both free formaldehyde 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). Composites treated with LMW PF resin were commercially produced, which exhibits significantly improved dimensional stability with anti-swelling efficiency (ASE) values of 75 and 95% (Stamm 1959). The efficiency of PF resin depends on the penetration of the cell wall, which is influenced by the average molecular weight of the resin (Gabrielli and Kamke 2010). PF resin with a number-average molecular (M_n from 290 to 480 g mol^{-1}) can penetrate the cell walls resulting in increased stability, whereas PF resin with M_n of 820 g mol^{-1} remained in the cell lumens (Furuno et al. 2004).

Formaldehyde is considered as a dangerous substance, which is strictly limited for the indoor wood products (Salem et al. 2011). However, very few publications have determined the influences of manufacturing conditions on the emission of formaldehyde (Carlson et al. 1995). Increasing pressing time during hot pressing might be one useful method to reduce the formaldehyde emission of wood-based panels. The purpose of this study was to study the effects of pressing time on the formaldehyde emission of phenol-formaldehyde resin and the properties of kiri SBs.

Materials and methods

Strands production

Kiri trees (*Paulownia tomentosa* (Thunb. Steud.) were harvested near Mannheim (Germany) and provided by the company WeGrow GmbH (Bonn, Germany). Logs of 2 m length were manually debarked with diameters of 12-20 cm and firmly held by a hydraulic clamp, moved towards a knife ring flaker PZUL 8-300 (Pallman Company, Zweibrücken, Germany). The feeding speed was 36 mm s^{-1} to obtain a target strand length of 110 mm, a thickness of 0.5-1.0 mm, and a width of 10-50 mm. These strands were further sorted out by a sieving shaker. Three fractions of strands were categorized by width: 10-30 mm, 30-50 mm, and bigger than 50 mm and sieve fractions smaller than 10 mm were discarded as fines. To avoid mould growth, the strands were immediately dried at 70°C in a drying oven to a target moisture content of 3% to 5%. In order to maintain moisture content low, the strands were stored in plastic bags and then delivered for panel manufacturing.

Panel manufacturing

Single-layered strand boards (SBs) were produced at a target density of 500 kg m^{-3}. The strand fractions were mixed in a ratio of 4:5:1 (10-30 mm: 30-50 mm : >50 mm). The strands were resonated in a drum blender with rotating speed at 30 rounds min^{-1}. Two types of phenol-formaldehyde resins (PF) with low (LMW) and high molecular weight (HMW) were used as adhesive with the amount 10% stock solution based on the oven-dry weight of strands. LMW PF resin was provided by Surfactor GmbH (Schöppenstedt, Germany) with M_w from 400 to 500 g mol^{-1} and solid content 53%. HMW PF resins were provided by Prefere resins Germany GmbH (Erkner, Germany) with M_w from 1000 to 1200 g mol^{-1} and solid content 70%. The resinated strands were formed into a mat (450 × 450 mm^2) in a cold pre-press before being delivered to hot-pressing at 140 °C for 90 s mm^{-1} using metal bars stops to ensure a panel target thickness of 12 mm. In total, 12 panels were produced, which is consists of four SBs for each of two variants LMW and HMW PF at pressing time 90 s mm^{-1} and two SBs for each of two variants of 135 s mm^{-1} and 180 s mm^{-1}.

Table 1 Parameters of panel manufacturing

Panel dimensions	450 × 450 × 12 mm^3
Wood species	Kiri (*Paulownia tomentosa*)
Target mat moisture content	8-10%
Resin type	LMW PF (M_w=400-500 g mol^{-1}; solid content HMW PF (M_w=1000-1200 g mol^{-1}; solid content
Resin mass content	10%
Blender rotation speed	30 rounds min^{-1}
Pressing temperature	140°C
Pressing time	90 s mm^{-1}; 135 s mm^{-1}; 180 s mm^{-1}

Physical and 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 (1993a). Bending strength (modulus of rupture, MOR) and modulus of elasticity in bending (MOE) according to EN 310 (1993b) (7 replicates for each panel, n = 14), and internal bond strength according to EN 319 (1993c) (5 replicates for each panel, n = 10). Thickness swelling (TS) and water absorption (WA) were determined after 24 h immersion in water at 20°C according to EN 317 (1993d) (5 replicates for each panel, n = 10). The temperature of the water bath was maintained at 20°C throughout the test period. After 2 h and 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. All test specimens were randomly selected by their density variation (500 kg m^{-3} ± 10%). All tests were performed using a universal testing machine (ZwickRoell GmbH & Co. KG, Ulm, Germany).

Formaldehyde emission (EN 717-2)

Free formaldehyde emission was assessed by gas analysis according to EN 717-2 (1994). The test specimen dimension was 400 × 50 × 12 mm. A dual-chamber device GA 5000 (Fagus Grecon, Alfeld, Germany) was used. The edges of the specimens were sealed with aluminum tape before it was placed in a 4-liter cylindrical chamber with controlled temperature (60.0 ± 0.5°C), airflow (60 ± 3 l h^{-1}), and pressure (from 978 to 1092 Pa). The air in the device is conducted into wash bottles in which the free formaldehyde dissolves in water. The amount of free formaldehyde was calculated as the average of two specimens after 4 h. At the end of the process, the formaldehyde concentration was assessed by the acetylacetone method (EN 717-2 1994).

Results and discussion

Internal bond strength (IB)

SBs variants bonded with LMW PF displayed a higher internal bond strength than those with HMW PF except SBs at pressing time of 180 s mm^{-1} (Fig. 1). This effect can be attributed to the resin penetration and expansion of LMW PF created smaller resin granules. On the other hand, HMW PF produced bigger resin granules on strands and these molecules might be too big to penetrate into wood strand cells although it was equally distributed over strand surfaces. LMW resin was able to penetrate deeper into wood strand cells, which was fixed inside the cell wall by heat during hot pressing. This effect might be additionally enhanced by the expansion and cross-linking reaction of LMW resin. LMW resin was able to spread over the strand surface after resin droplets hit the strands, which creates a continually bonding line on the strand surface leading to a larger resin coverage. Accordingly, this expansion on the strand surface can be linked to the stronger bonding strength of SB panels based on LMW resin. However, a larger resin coverage and greater penetration of LMW resin granules, which is cured during hot pressing, might block the way to release free formaldehyde. Therefore, it can be concluded that LMWPF has formed the stronger bonding contact between strands of kiri SBs; however, resulting in lower permeability inside SB panels. With increasing pressing time, the IB strength of SBs containing LMW PF is considerably decreasing and this result was found similar to the study by (Kusumah et al. 2017) for particleboards made from sweet sorghum bagasse bonded with citric acid.

Statistic analysis revealed that IB of kiri SBs blended with LMW PF resin was highly affected by pressing time. Pressing time negatively influenced IB of kiri SBs containing LMW PF resin. It is assumed that a longer pressing time under high temperature causes too dry wood strand inside the mat resulting in brittle bonding lines, which leads to a decrease in the IB of SBs. Too dry particles have a negative effect on particle bonding quality (Kollmann et al. 1975). It is concluded that longer pressing times negatively influence IB properties of kiri SBs bonded with LMW PF resin.

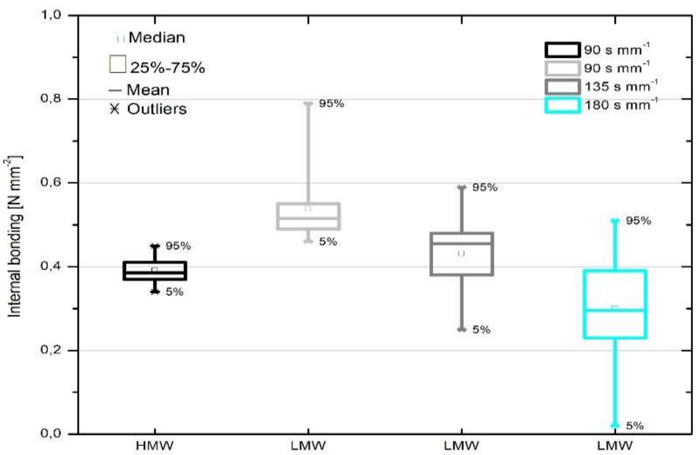

Fig. 1. Internal bond strength (IB) of strand boards at target density 500 kg m^{-3} (n = 10). The components of the boxplots are indicated as insert; the whiskers show the 5% and 95% quartile.

Unlikely IB, statistical analysis revealed that the variants with different molecular weight resin did not show the significant influence of pressing time on MOR, and MOE. For SBs pressed at 180 s mm^{-1}, a slightly higher MOE and MOR were not expected. It is presumed that kiri wood strands get stiffer and more densified after a long time pressing under high compression. However, no clear effect of the PF resin´s molecular weight and pressing times on MOR and MOE of SBs was apparent (Fig. 2). This effect indicates that the reduction in IB due to longer pressing time has no relevant influence on the bending strength of SBs.

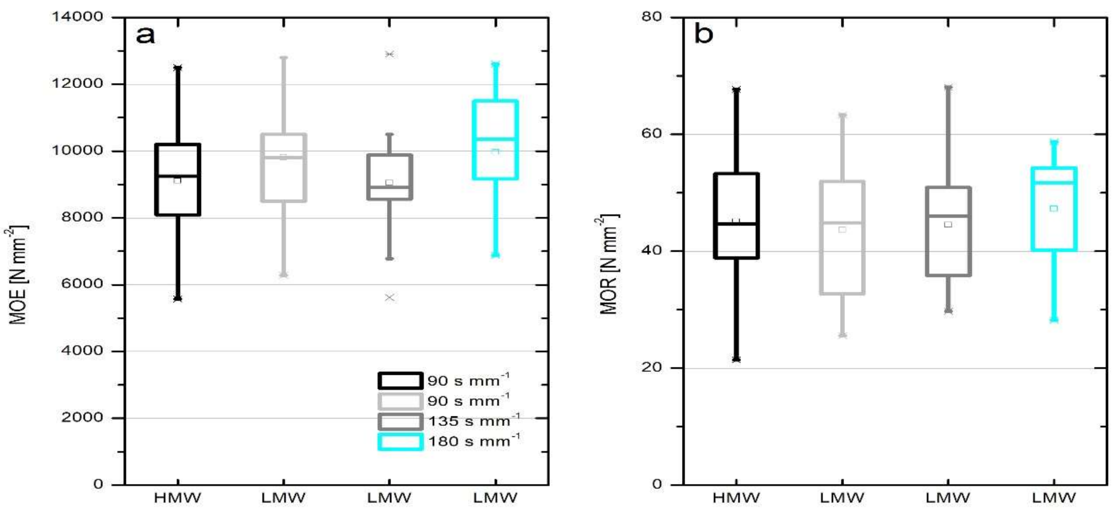

Fig. 2. (a) Modulus of elasticity (MOE); (b) modulus of rupture (MOR) of strand boards at target density 500 kg m^{-3} (n = 14). The components of the boxplots are indicated in Fig. 1; the whiskers show the 5% and 95% quartile.

Formaldehyde emission (FE)

Phenolic resins (phenol-formaldehyde PF), which distinctly requires longer pressing time necessary for curing when compared with urea-formaldehyde (UF) and melamine-urea formaldehyde (MUF), displays high resistance to hydrolysis of the C-C bond between the aromatic nucleus and the methylene bridge. Hence, they could be used for water resistance bonding to manufactures wood-based panels for exterior use or in humid conditions (Pizzi 1983; Pocius and Chaudhury 2003). Phenolic resins, which contains oligomeric and polymeric chains as well as monomeric methylol-phenol, free formaldehyde, and unreacted phenol, can only initiate the curing process by heat with the condensation of the precursers resulting in lengthening, branching, and cross-linking to a three-dimensional network (Pizzi 1983; Knop and Pilato 1985; Gardziella et al. 2000; Pizzi and Mittal 2018). The quantity of free formaldehyde content considerably depends on the use of formaldehyde to phenol mole ratios in resin synthesis (Meyer et al. 1986).

Pressing temperature to cure the phenolic resin should usually be in the range of 130°C to 220°C (Kollmann et al. 1975; Meyer et al. 1986). Also, sodium hydroxide (NaOH) is usually employed as a reaction catalyst during the synthesis of phenolic resin (Pizzi 1983; Knop and Pilato 1985; Gardziella et al. 2000). In this study, the formaldehyde emission (FE) of SBs based on LMW PF resin was considerably higher than that of boards manufactured with HMW PF resin.

Nevertheless, FE generally decreased continuously with increasing pressing time. LMW PF is condensed at a lower degree than HMW PF, i.e. it contains a higher amount of both free formaldehyde (FA) and methylol groups and less methylene and methylene-ether bonds than HMW PF (Hultzsch 1950). The formaldehyde emission is influenced by pressing conditions such as mat moisture content, pressing temperature, and time (Carlson et al. 1995). In general, a longer pressing time reduces the formaldehyde emission of kiri strand boards.

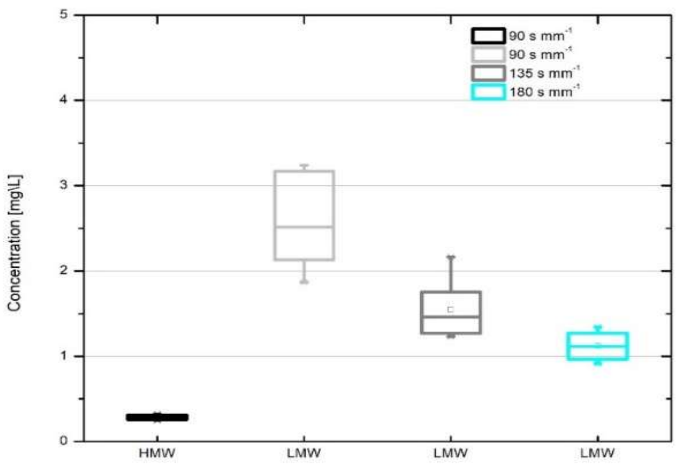

Fig. 3. Formaldehyde emission according to EN717-2. The components of the boxplots are indicated in Fig. 1; the whiskers show the 5% and 95% quartile.

Water-related properties

Boards containing LMW PF resin with various pressing times exhibited significantly lower TS and WA after 2 h and 24 h immersion in water than those of HMW PF resin (Fig. 4). Nevertheless, statistical analysis revealed that variants with different molecular weight resin did not show significant differences with respect to pressing time, TS and WA. LMW PF-boards pressed with the longest pressing time 180 s mm^{-1} showed a slightly higher value of TS after 24h than those with shorter pressing time. After 2 h immersion, boards pressed in 90 and 135 s mm^{-1}. Comparing TS and WA after 24 h immersion, a similar TS of SBs containing LMW PF resin pressed at 90 and 135 s mm^{-1} caused different WA. For SBs pressed at 180 s mm^{-1}, a higher TS was expected due to a longer pressing time. Phenol-formaldehyde binders experience a slower transition; hence, they require longer pressing times which is suggested from 8 to 30 minutes for board thickness of 19 mm (Kollmann et al. 1975). Therefore, a pressing time of 180 s mm^{-1}, which significantly reduces formaldehyde emission, could be too long resulting in a reduction in bonding performance when exposed to water.

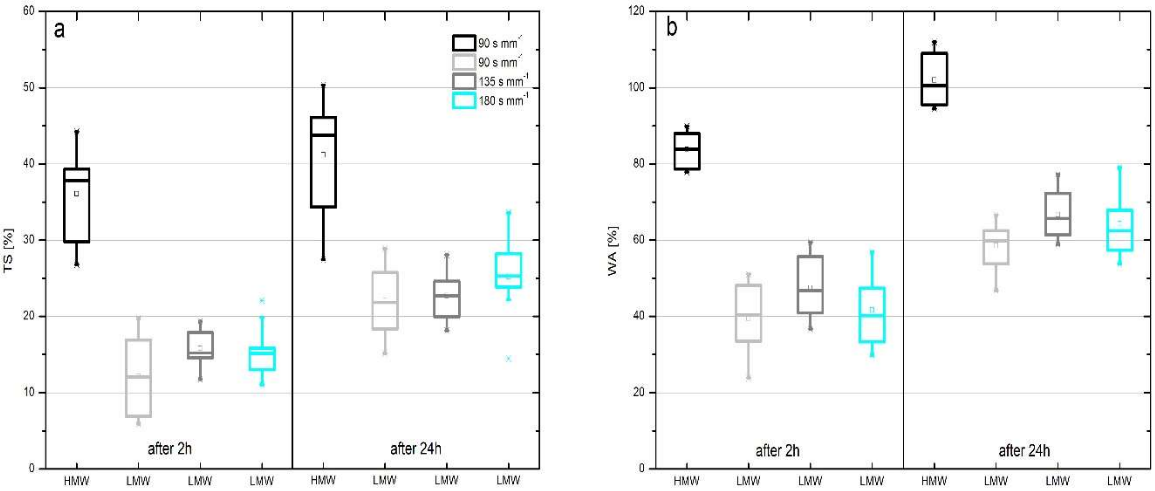

Fig. 4 (a) Thickness swelling (TS); (b) water absorption (WA) of strand boards at a target density 500 kg m^{-3} (n = 10) after 2h and 24h immersion in water. The components of the boxplots are indicated in Fig. 1; the whiskers show the 5% and 95% quartile.

Conclusion

LMW PF resin was successfully used as an adhesive to increase the IB and dimensional stability of SBs from kiri wood. SBs containing LMW PF pressed with longer pressing time exhibited lower formaldehyde emission; however, it also displayed a significant reduction in IB. This effect indicates that IB and formaldehyde emission of LMW PF more dependent on the pressing time than HMW PF. This can be attributed to deeper penetration and larger coverage area of LMW PF resin causing lower permeability of strand mat during hot pressing, which leads to that formaldehyde emission of the strand mat needs longer pressing time to release during hot pressing. It cannot be considered an optimal method to reduce formaldehyde emission because it increases pressing time resulting in low manufacture productivity.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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Publication VI

Assessment of phenol-formaldehyde resin distribution on the strand surface by light microscopy

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Assessment of phenol-formaldehyde resin distribution on the strand surface by light microscopy

Abstract

This study reports on a new approach for the detection of phenol-formaldehyde (PF) resin distribution on the strand surface after air-spray application in a glue drum using reflecting light microscopy (LM). The method allows visualization and quantification of PF resin distribution on wood strands by using either waterproof paper or 200 µm thick microtomed kiri veneer slices, which were clipped with staples on conventional strands typically used for the production of oriented strand boards (OSB). These strands were together mixed with other conventional OSB strands and two types of molecular weight PF resins were sprayed on the strand mixture. While the total area of resin coverage was similar for both resins, the low molecular weight (LMW) PF resin formed a higher number of resin droplets than the high molecular weight (HMW) PF resin. The study offers a new, easy applicable approach for a qualitative investigation of resin distribution on the strand surface.

Keywords: resin droplets; low molecular weight; high molecular weight; waterproof paper; veneer slices

Introduction

Phenol-formaldehyde (PF) adhesive is the most used adhesive for exterior-grade plywood and, in addition to polymeric methyl diphenyldiisocyanate (pMDI), oriented strand boards (OSB) (Stamm and Seborg 1939; Mendes et al. 2013; Bufalino et al. 2015; Pham et al. 2019, 2021; Grinins et al. 2021). This high share of application is basically due to the high resistance of the adhesive to hydrolytic degradation. It is well known that the performance of OSB depends on both the properties of the PF adhesive such as viscosity and the processing variables such as pressing parameters (Park et al. 1998). Additional important factors are the resin distribution, the thickness of the "glue-line" and the area covered by resin on the strand surface. Modulus of rupture (MOR), internal bond strength (IB) and dimensional stability reached high values when fine droplets were generated during the spray application and the resin was thus uniformly distributed (Lehmann 1970; Zhang et al. 2009). Therefore, assessing the adhesive distribution and droplet size distribution on the strands before hot pressing can indicate their influence on the OSB properties. Transmitted light, fluorescence and electron microscopy in combination with microtome techniques, staining, and chemical labeling are common methods to study the distribution on the strand or flake surface and the penetration depth of thermosetting resins (PF, urea-formaldehyde -UF- or melamine-urea-formaldehyde -MUF) into the wood tissue (Xing et al. 2005; Stephens and Kutscha 2007; Pakdel et al. 2008; Mahrdt et al. 2015, 2017). Advanced methods for resin detection in wood tissues are confocal laser scanning microscopy (CLSM) and X-ray microcomputed tomography (X-ray µCT) (Pakdel et al. 2008; Riegler et al. 2012; Paris et al. 2014; Evans et al. 2015). In contrast to light microscopy, most of the other above-mentioned methods are relatively laborious and often cause artifacts and misleading observations. Stains or chemically bonded dyes (for fluorescence microscopy) are mostly used to enhance the contrast between the adhesive and wood.

LMW PF resin drops are more transparent than HMW and thus more difficult to detect due to the minor contrast of the former to wood tissue, particularly after resin curing inside the wood tissue. In addition, the bonding performance of wood-based panels depends considerably on adhesive distribution (Riegler et al. 2012). Therefore, the aim of this study is to introduce a new method for detecting PF resin distribution on the surface of wood strands directly after blending (spray application of resin) without additional staining. As well, to assess the influence of the type of PF resin on the distribution and provide clues to the interaction between the flake board properties and resin distribution.

Materials and methods

Veneer slices

Slices of kiri wood with dimensions of approximately 25 x 70 x 0.2 mm (tang. x long. x rad.) were prepared in a microtome Mod. 81A80 (Sartorius GmbH, Göttingen, Germany) using metal blades type C (Leica Instruments GmbH, Nußloch, Germany). Prior to slicing, solid kiri wood pieces were fully saturated with water using a vacuum of 60-80 mbar.

Strand production

The strands were manufactured by a knife ring flaker PZUL 8-300 (Pallman Company, Zweibrücken, Germany) prepared at Fraunhofer WKI (Braunschweig, Germany) with a target length of 110 mm, a target thickness of 0.5-1.0 mm and a target width of 10-50 mm. A single wood stem was firmly held by a hydraulic clamp and moved towards the knife ring with the feeding speed of 36 mm s^{-1}. All strands were directly sorted by a sieving shaker into 3 fractions: 10-30 mm, 30-50 mm and bigger than 50 mm. Sieve fractions smaller than 10 mm were discarded as fines. To avoid mould growth, the strands were dried at 70°C to a moisture content of 3% to 5% in a drying oven. Strands were stored in plastic bags to keep the moisture content low. However, in this study only strands with a width of 30-50 mm were used.

Resin blending

Two different PF resins were used as adhesive with the amount of 20 % based on the oven-dry weight of the wood strands. The low molecular weight (LMW) PF resin displayed a molecular weight (M_w) of 400–500 g mol^{-1} and solid content of 53% (Surfactor GmbH, Schöppenstedt, Germany), whereas the high molecular weight (HMW) PF resin displayed an M_w of 1000-1200 g mol^{-1} and a solid content of 70% (Prefere Resins Germany GmbH, Erkner, Germany). The solid content of LMW resin was increased up to 65% by removing the water in a rotary evaporator. Both resins were air-sprayed at 8 bar pressure through the nozzle (air cap V10 700 35 xx8) using a Walter Pilot WA700 gun (Walter Spritz- und Lackiersysteme GmbH, Neunkirchen Struthütten, Germany). Twenty waterproof papers (3M Germany GmbH, Neuss, Germany) and veneer slices were attached by staples on both sides of kiri strands (Fig. 1b, e). These strands clipped with veneer slices and waterproof paper were then together mixed with 200 g pre-dried strands with a width of 30-50 mm for resin blending. About 48-50 g of resin per experiment was sprayed for 2 min. The strand mixture was sprayed with PF adhesive in a drum blender with a rotating speed of 30 rounds min^{-1}. Afterwards, the adhesive droplet size distribution and area coverage on the strand surfaces was evaluated using reflecting light microscopy. Subsequently, the strands with veneer slices and waterproof papers were sorted out from the strand mass and dried in a kiln at 140°C for 30 min to cure the PF resin. The dark purple HMW PF resin exhibited a strong contrast with the light kiri wood surface, while LMW PF resin had to be darkened by adding a black dye with 0.005% based on resin weight (Kremer Pigmente GmbH & Co.KG, Aichstetten, Germany).

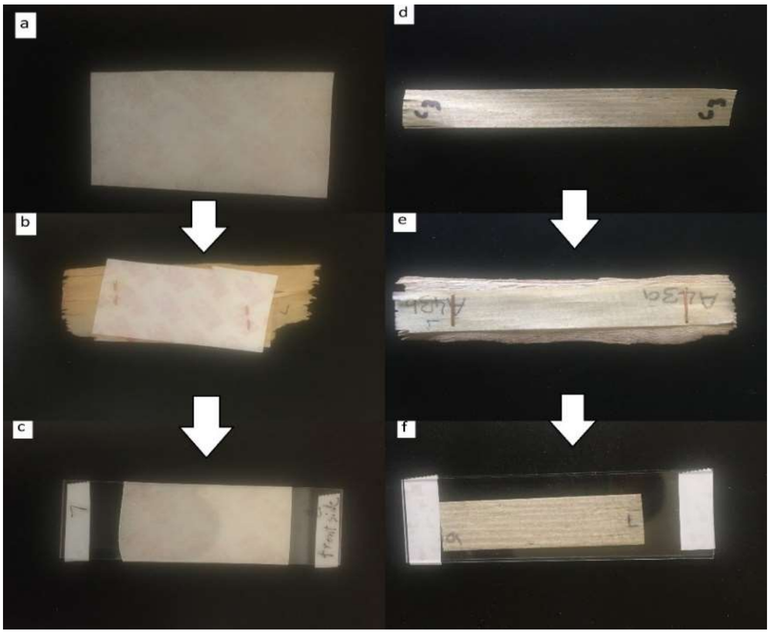

Fig. 1. Sample preparation of waterproof papers (a, b, c) and veneer slices (d, e, f) for the resin blending (b, e) and light microscopy (c, f).

Light microscopy (LM) evaluation

The water proof paper and veneer slices were removed from the strands and covered by glass slices on the both surfaces of the specimen in order to generate a better surface quality for light microscopy (Fig. 1c, f). Images (2D) were taken with an Ellipse E600 microscope (Nikon Corporation, Tokyo, Japan) with a digital camera (Nikon DS-Filc), which was connected to a computer. Image processing was performed by using NIS-elements software (4.10) (Nikon Instrument, New York, U.S.A). For measurement of resin area coverage, five of each resinated waterproof papers and veneer slices were randomly selected. On each of these specimens, five locations were taken by using 10 × magnification. In total, 25 images per resin treatment were recorded. For the image analysis, an open-source software ImageJ was used (Schindelin et al. 2012; Rueden et al. 2017). The following processing steps were applied: conversion of JPG into 8-bit greyscale image and was split into channels (blue, red, green), the brightness and contrast of the green or blue channel image were set (depending on which channel brought the best result) and a manual threshold was applied. Afterward binary images were used to apply a watershed algorithm to estimate the resin droplet size distribution and area coverage, automatically. Feret´s diameter was determined to describe the distribution of the resin droplets (Fig. 2, 3 and 4). It is the longest distance between any 2 points along the resin droplet´s boundary, as known as maximum caliper.

Results and discussions

At the beginning of this study, we used filter paper clipped on the strands to detect the resin droplets; however, the resin droplets of LMW PF could not be detected due to the strong spreading of the resin spots on the filter paper. Waterproof papers and veneer slices proved to be better suited for microscopic evaluation of the resin droplets than the strands themselves due to the uneven surface of the latter. In this study, the adhesive droplets were detected by LM on specifically prepared kiri wood slices with a smoothed surfaces. The distribution of resin spots for the high molecular weight (HMW) PF is shown with respect to the raw (Fig. 2a) and binary data (Fig. 2b). Spraying of HMW and low molecular weight (LMW) PF resin resulted in a different resin droplet size distribution on the waterproof paper (Fig. 2c). The image analysis did not reveal significant differences in the total area coverage between the resins of different molecular weights. In addition, no clear trend was found with regard to the influence of molecular weight on droplet size distribution on the waterproof paper.

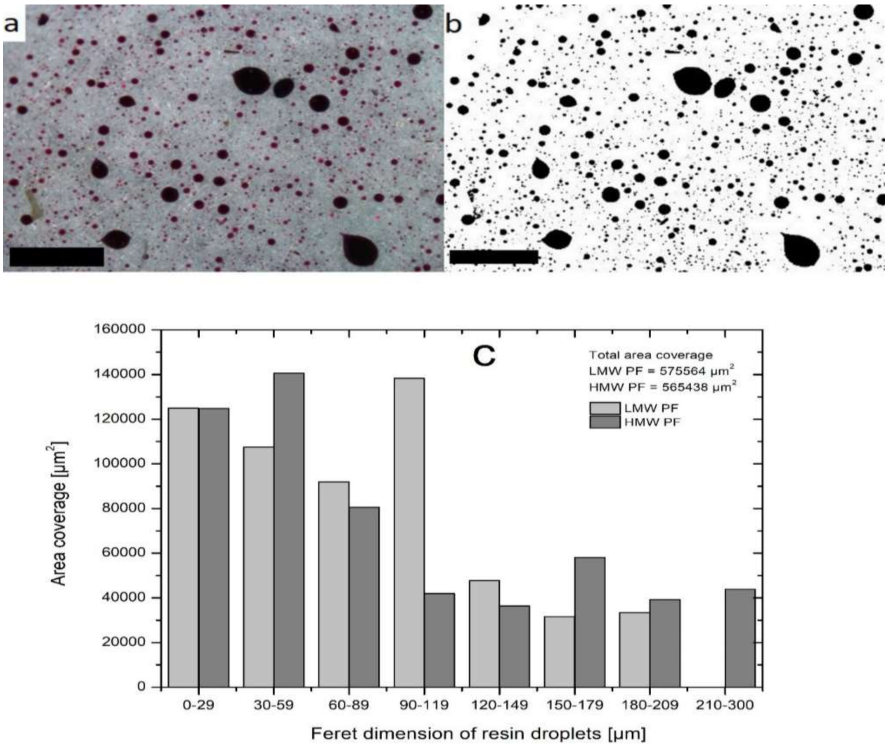

Fig. 2. Distribution and area coverage of HMW resin droplets on waterproof paper after spraying: (a) raw and (b) binary image (scale bar 500 µm) and (c) histogram of droplet size distribution.

As observed for the waterproof filter paper (Fig. 2c), both resin types showed similar total area coverage directly on the kiri wood strands (Fig. 3e) but the resin droplet sizes and the number of droplets were different. In addition, the resin droplets of the waterproof paper exhibited another shape and were more round than those on the wood strands. Spraying the LMW PF on the wood strands resulted in a clearly larger number of droplets than when using HMW PF, from which it follows that the average drop size of these droplets is significantly smaller. This difference is due to the smaller size of resin droplets formed during air-spray (atomization) and can be attributed to the lower viscosity of the LMW PF.

The use of waterproof paper clipped on the strand surface is easier and faster than using veneer slices, the production of which is very time-consuming and requires the appropriate equipment. In addition, LMW PF resin droplets can be observed on the surface of the waterproof paper without adding a dye, which is impossible on the veneer and strand surface. Still, veneer slices exhibit almost the same structure and chemical composition as the strand surfaces and therefore the resin distribution on veneer slices is more similar to that on the strand surfaces than that on the waterproof paper.

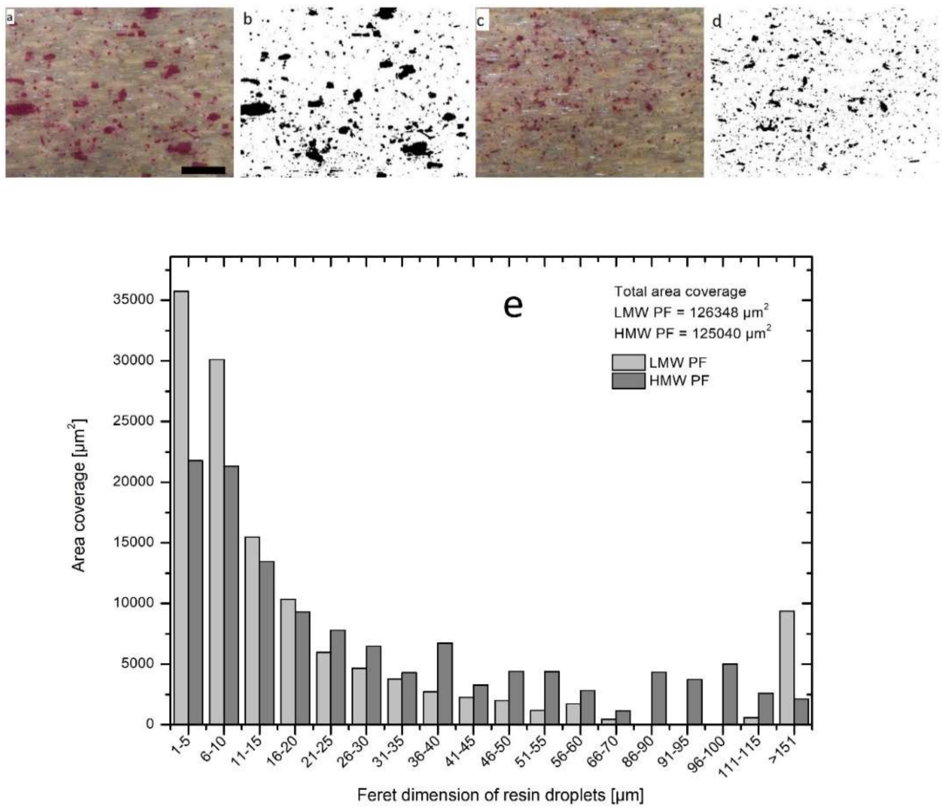

Fig. 3. Distribution and number of resin droplets of HMW (a, b) and dyed LMW (c, d) PF resin on kiri wood strands. The (a, c) raw and (b, d) binary image and (e) histogram of droplet size distribution after the spraying. Scale bar 500 µm.

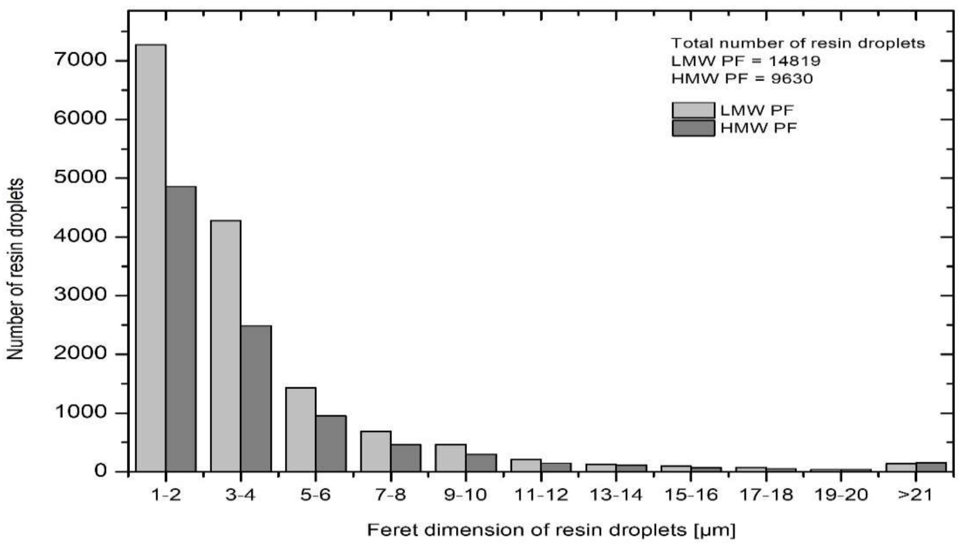

Fig. 4. Number of HMW and dyed LMW PF resin droplets on kiri wood strands.

Conclusions

The newly developed method is easily applicable and useful to monitor the spraying quality on strands surfaces by light microscopy. The method allows to identify the differences in droplet size distribution influenced by the viscosity of the resin (here due to differences in molecular weight) and can provide indications on the properties of strand boards such as OSB. Clipping water-proof paper on the strand surfaces is easy to perform but using veneer slices provides information on the resin distribution, which is more similar to that on strand surfaces.

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