Effect of Short-term Thermomechanical Densification of Scots Pine Veneers Bonded with Different Adhesives on Shear Strength of Plywood

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The effect of short-term thermomechanical (STTM) densification on shear strength of plywood produced with Scots pine (*Pinus sylvestris* L.) veneers was investigated. The veneer sheets were densified under temperatures of 20 °C, 120 °C, and 180 °C, and pressure of 1 MPa for 2 min. There were four polyvinyl acetate (PVAc) adhesives, 1 emulsion polymer isocyanate (EPI)-adhesive, and 2 resins for comparison reasons applied. The samples were stored for 7 d under the normal climate temperature of 22 °C and relative humidity of 65%. As a result of STTM densification, the plywood had higher density but lower shear strength. The plywood made with these veneers and EPI-adhesives showed the highest shear strength values. There was a slight difference of the final product's shear strength between the used adhesives, but it was not noticeable in tested conditions.

Keywords: STTM; Thermomechanical densification; Veneer; Plywood; Pinus sylvestris; PVAc; EPI adhesives

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INTRODUCTION

Among all methods of wood modification, thermomechanical modification has shown increasing importance. Over the last decades, there has been a rapid increase of application of different thermo modifications (thermal, thermomechanical, and thermo-hydro-mechanical) of wood to improve its properties. According to Niemz and Sonderegger (2017), thermal modification of wood has reached an industrial market volume, but it is still relatively low. Advantages of the use of thermally modified wood is the improvement of physical, mechanical, and aesthetic properties (Hill 2006; Navi and Sandberg 2012; Bekhta *et al.* 2015).

There have been studies considering the reduction of duration of the thermal modification. This process is called short-term thermomechanical (STTM) densification. The concept of thermal wood treatment dates back to the beginning of 1900s (Kollmann *et al.* 1975). One of the main goals of the densification is to increase mechanical strength properties, but also thermal conductivity (Asako *et al.* 2002). A good indication was established by Jennings (1993), Jennings *et al.* (2005, 2006), Kamke (2006, 2007), Kutnar *et al.* (2008a, 2008b, 2009), and Sernek *et al.* (2004). According to Bekhta *et al.* (2012), the densification of veneer to be used for plywood allows up to 40% reduction of glue spread and up to 45% reduction in pressing pressure. Furthermore, STTM densification of veneers causes irreversible changes in their morphology (Bekhta and Krystofiak 2016). The STTM-densified veneer surfaces become smoother, more hydrophobic, and the roughness and thickness values decrease significantly (Bekhta *et al.* 2015). A further advantage is the increase of mechanical performance of plywood (Bekhta *et al.* 2019). This type of modified wood can be used in production of wood components such as plywood and laminated veneer lumber (LVL) (Bekhta *et al.* 2015).

The most common adhesives used in the production of plywood or LVL are phenol formaldehyde resin (PF), urea-formaldehyde resin (UF), and melamine-urea-formaldehyde resin. They have one major disadvantage – they release formaldehyde (Dunky and Niemz 2002). The European Commission in 2001 classified formaldehyde as a suspected carcinogen (Directive 2001/58/CEE). Therefore, the biggest furniture producers are looking for a substitute for formaldehyde resins.

That is also the reason why the use of polyvinylacetate (PVAc-adhesive) and emulsion-polymer-isocyanate adhesive (EPI-adhesives) in the production of plywood based on STTM-densified veneers was investigated.

EXPERIMENTAL

Materials

Densification was conducted on Scots pine (*Pinus sylvestris* L.) with an average density of (490 ± 30) kg/m^3 and a moisture content (MC) of (10 ± 0.5%). The veneers used were without wood defects to obtain reference results. The veneers (310 mm × 310 mm × 1.5 mm) came from Sklejka Multi S.A. Comp., Bydgoszcz, Poland.

For bonding of veneers, the formaldehyde-free adhesives PVAc and EPI were from Jowat AG Comp (Detmold, Germany), and the formaldehyde-based resins UF and PF were from Silekol Comp. (Kędzierzyn-Koźle, Poland). The properties of these adhesives are listed in Table 1.

Table 1. Properties of Adhesives Used

Adhesive (trade name)	Base	Durability Class (DIN EN 204 (2016)	Density (g/cm^3)	Viscosity (mPa*s)	Open Time (min)	Adhesive Amount (g/m^2)
Jowacoll 104.20	PVAc	D2	1.07	10500	8 to 11	180
Jowacoll 103.05	PVAc	D3	1.08	11000	5 to 8	180
Jowacoll 107.20	PVAc	D4	1.05	6000	10	180
Jowacoll 102.49	PVAc	D2	1.50	11000	8 to 12	180
Jowacoll 102.49 + 195.60	EPI	D4	1.50	11000	9 to 12	180
Silekol M-2	UF	D2	1.28	1200 to 1800	2 to 3	180
K-48	PF	D4	1.10 to 1.23	200 to 3000	30	180

Methods

Veneer preparation

Veneer sheets (50 pieces of 310 mm × 310 mm × 1.5 mm) with consideration of wood defects were carefully selected and cut in four equal-sized veneers (150 mm × 150 mm × 1.5 mm).

STTM densification

Each veneer was densified separately through heat and pressure. Between the heated plates of the laboratory press (HLP 350; HÖFER Presstechnik GmbH, Taiskirchen, Austria) and a specimen every time there was a clean anti-adhesive paper. The experiments were conducted under the following parameters: pressure of 1 MPa, temperature of 20 °C, 120 °C, and 180 °C, and time of 2 min

After densification, the specimens were removed from the heated press and naturally cooled down to room temperature. Then the veneers were stored in a conditioning chamber at 20 °C and a relative humidity of 65% until they reached constant mass weight.

Veneer thickness

The method of thickness measurement was based on the literature (Navi and Giradet 2000; Welzbacher *et al.* 2008; Gong *et al.* 2010; Fang *et al.* 2012; Tu *et al.* 2014; Bekhta *et al.* 2015). Veneer thickness at five measuring points (Fig. 1) was measured before (T_B) and 24 h after (T_A) the STTM densification.

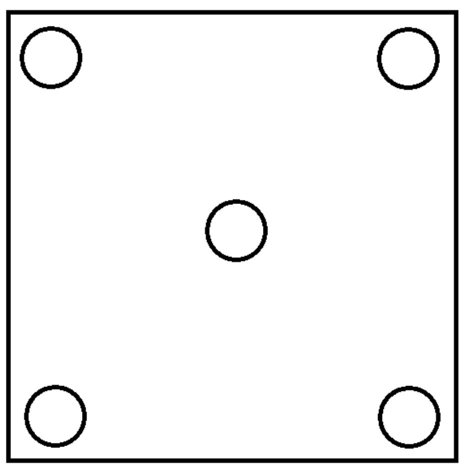

Fig. 1. Measuring points by the measurement of compression ratio

The compression ratio (CR) is defined as a degree of difference of thickness to original thickness and was calculated based on Eq. 1,

$$CR = \frac{T_B - T_A}{T_B} \times 100\% \qquad (1)$$

where T_B is the veneer thickness (mm) before densification and T_A is the veneer thickness (mm) after densification.

Shear strength and wood failure

The measurement of shear strength and mean wood failure (WF) percentage were carried out according to EN 314-1 (2004). There were 10 specimens (Fig. 2) of each variant that were prepared and stored in a climate chamber for 7 d to obtain reference results. Authors were interested only in plywood stored in dry conditions. For the experiments, adhesives with the different durability class were used. Water resistance and thermo-resistance tests will be a topic of the next investigations.

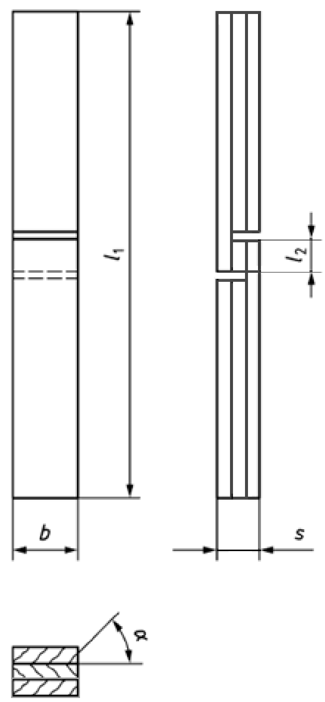

Fig. 2. Schematic of a tested sample (adapted from EN 314-1 (2004))

RESULTS AND DISCUSSION

Influence of STTM on Compression Ratio

Figure 3 presents the compression ratio in the function of the temperature of STTM densification. With 99% confidence, based on a two-way test, the value of CR resulting from densification at 120 °C (1.10% to 3.13%) was higher than the value of CR resulting from densification at 20 °C (10.53% to 15.22%). Furthermore, with 90% confidence, based on a two-way test, the value of CR resulting from densification at 180 °C (11.62% to 14.13%) was higher than the value of CR resulting from densification at 120 °C (14.20% to 16.86%).

The investigations showed that the pressing at 20 °C and 1 MPa pressure led to the increase of compression ratio of 2%. The surface of veneers at this temperature was smoother and that explained why the veneer was thinner. At 20 °C there were no chemical changes in wood (Windeisen and Wegener 2008). The increase of temperature during the STTM densification resulted in the growth of compression ratio remarkably. The highest compression ratio of 15% was reached at the highest temperature of 180 °C.

The applied pressure of 1 MPa was lower than the compressive strength of pine (⊥) of 7.7 MPa. However, due to the reduction of proof stress caused by increased temperature and also water present in the adhesive, veneers become thinner (Niemz and Sonderegger 2017).

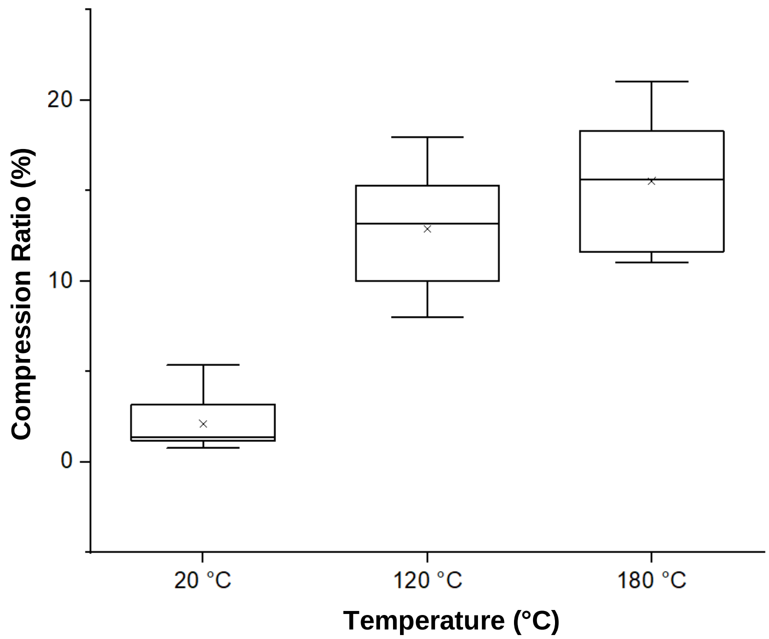

Fig. 3. Influence of the pressing temperature during the STTM densification with pressure of 1 MPa on the pine veneers' compression ratio

Furthermore, this behavior might be attributed to chemical changes of lignin. Lignin softens at 70 to 80 °C, and then radicals are formed in the depolymerization reactions (120 to 130 °C) that in turn are recombined (140 to 200 °C) to compounds that presumably have lower polarity (Windeisen and Wegener 2008).

Influence of STTM on Density

Figure 4 displays the density values in dependence of the temperature of STTM densification.

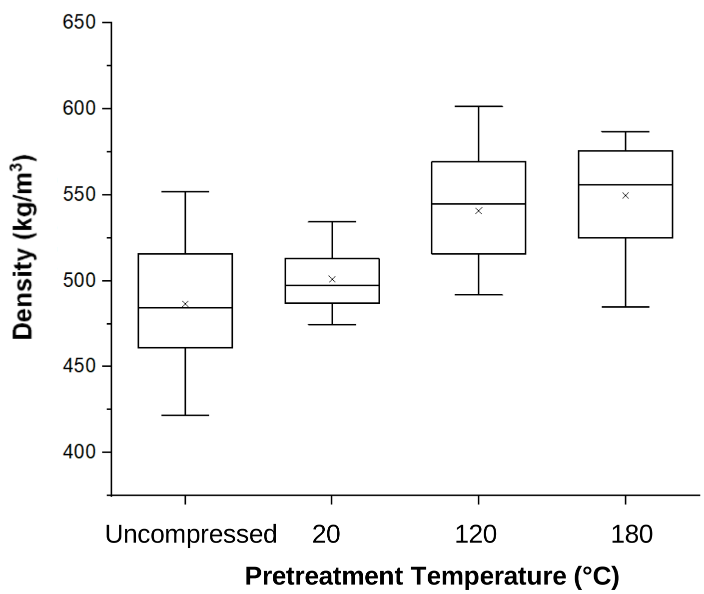

Fig. 4. Influence of the STTM densification on density of Scots pine veneers

The density of the uncompressed wood varied more noticeably. The STTM densification caused increased density values. With 90% confidence, based on a two-way test, the value of density after densification at 20 °C and 1 MPa (479.36 kg/m^3 to 493.60 kg/m^3) was higher than the value of density before densification (493.88 kg/m^3 to 508.08 kg/m). The increase of the temperature during the STTM densification accomplished significant growth of the density. Due to the reduction of the compressive strength of pine, the highest density of 550 kg/m^3 was reached at the highest temperature of 180 °C. The reasons for this behavior were also the wood structural changes mainly induced by the high temperature.

Influence of STTM on Shear Strength and Wood Failure

Figure 5 shows the shear strength values in the function of the temperature of STTM densification.

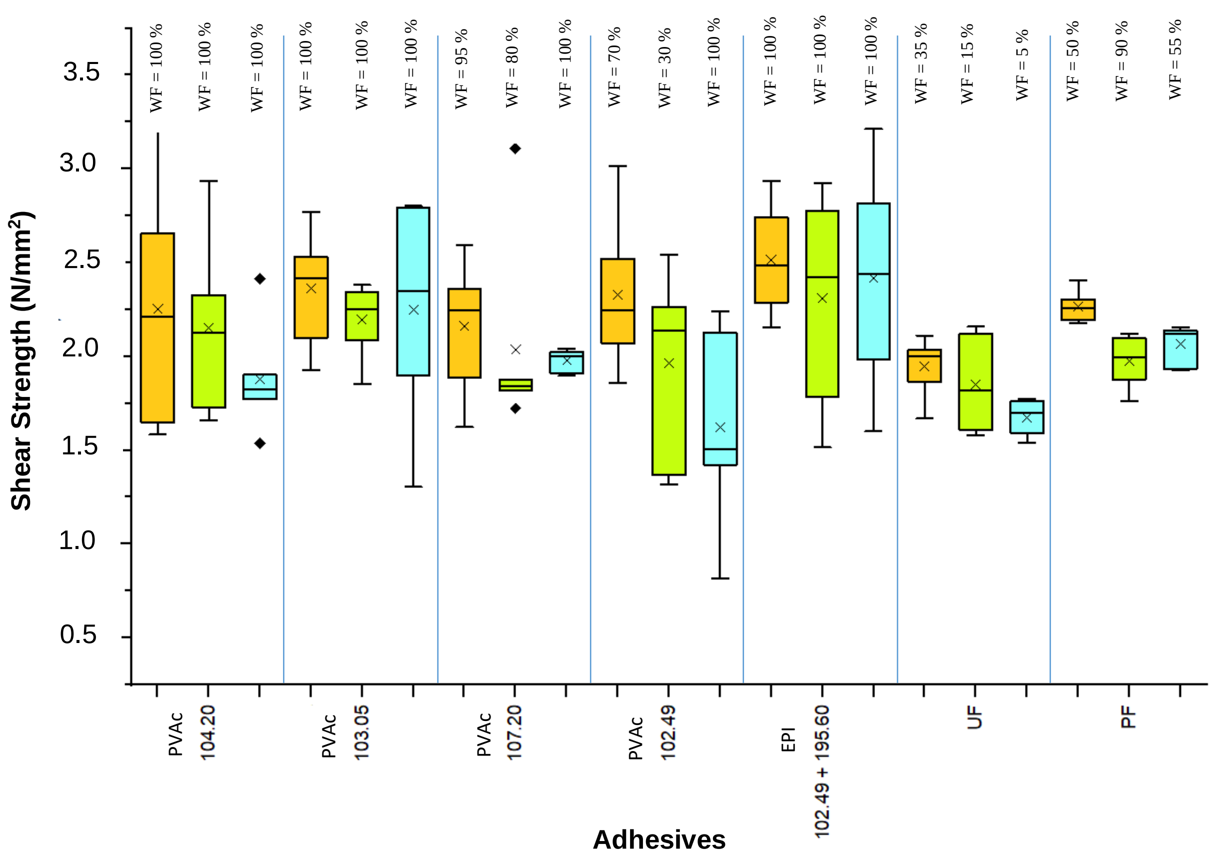

Fig. 5. Shear strength of plywood bonded with different adhesives after STTM densification at 20 °C (orange), 120 °C (green), and 180 °C (blue)

It was noticeable that the STTM densification caused a reduction of shear strength parallel to the fibers. All tested specimens had remarkably passed the required shear strength of $\leq$ 1 N/mm^2 according to EN 314-2 (1993). The results of plywood bonded with all types of adhesives were similar. Wood failure for PVAc- and EPI-adhesives during densification at 20 °C occurred 100% of the time. Meanwhile the wood failure for UF- and

PF-resin oscillated between 30% and 50%. With the increase of temperature of STTM densification the reduction of wood failure was noticeable.

The use of filled PVAc adhesive Jowacoll 102.49 (filler portion: 40%) without crosslinker prompted a reduction of shear strength by 30% and to a noticeable reduction of wood failure from 100% to 30%. However, the addition of an isocyanate-crosslinker to the PVAc adhesive in an amount of 15% led to a large increase of shear strength to 2.31 N/mm^2 to 2.51 N/mm^2. The EPI-adhesive obtained the highest shear strength and wood failure (100% by all densification temperatures) values.

According to Zeppenfeld and Grunwald (2005), isocyanate links up not only as a hardener with adhesive, but also with hydroxyl groups of wood. The wood surface hardens at the same time. That is the reason why EPI-adhesives are used for bonding of problematic wood with low surface energy.

The increased densification temperature led to a decreased shear strength. The water present in the adhesive might be attributed to plasticize of the veneer surfaces.

CONCLUSIONS

1. The temperature and the pressure during the STTM densification had a direct influence on the compression degree and the density of pine veneers. As a result of STTM densification under 180 °C and 1 MPa pressure, the plywood achieved the highest compression degree of 15 % and the highest density of 550 kg/m^3.
2. The use of unfilled PVAc adhesives helped to achieve relatively high shear strength values (1.9 N/mm^2 to 2.2 N/mm^2) and relatively high wood failure (80% to 100%). The application of condensation resins resulted in substantially lower shear strength values (UF-resin: 1.7 N/mm^2 to 2.0 N/mm^2; PF-resin: 2.0 N/mm^2 to 2.1 N/mm^2) and lower wood failure (UF-resin: 10 %; PF-resin: 70 %) due to their restricted penetration depth in the wood.
3. The addition of filler to PVAc adhesive led to the reduction of shear strength of the plywood made with STTM-densified pine veneers.
4. The addition of isocyanate-crosslinker to the PVAc adhesive created EPI-adhesive**,** which prompted a noticeable increase of shear strength value to approximately 2.5 N/mm^2.
5. Although there was a growth of density in STTM-densified pine veneers compared to undensified wood, the results came in a compensation of loss in shear strength of plywood.

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