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Effect of the Percentage of MUF Adhesive Coverage on Shear Strength When Bonding Different Wood Species

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Due to climate changes, it is necessary to consider the use of other wood species to replace currently used woods. This work deals with the determination of the shear strength of bonded veneers (eight European wood species: spruce, larch, pine, beech, oak, poplar, birch, and alder) with Silekol® 311 melamine-urea-formaldehyde adhesive (MUF) with a variable coverage on the surface of the samples: 10, 15, 20, 25, 30, 50, 75, and 100%. The Automated Bonding Evaluation System (ABES) was used to evaluate and compare adhesive bond strengths. The larch, beech, and oak samples exhibited greater single-lap shear strength than the control samples from spruce. There was no statistically significant difference in shear strength regarding the adhesive coverage from 100% to 20% on the surface of the samples, for almost all wood species. The results of the project provide basic information about the bonding strengths with different coverage in the adhesive layer, comparing non-commonly used wood species in wood-based composites such as oriented strand board and particleboard.

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Keywords: Wood-based panels; Lap shear strength; Percentage of adhesive coverage; Melamine-urea-formaldehyde adhesive; Strand boards; Automated Bonding Evaluation System (ABES)

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INTRODUCTION

In Europe, wood-based composites, such as particleboard (PB), medium-density fiberboard (MDF), and oriented strand board (OSB), are usually produced from Norway spruce (*Picea abies*). Extreme conditions, such as winter storms (Seidl *et al.* 2014) and wind damage, can be mitigated by decreasing the proportion of Norway spruce, limiting stand age, and reducing timber stock (Pasztor *et al.* 2015). The most significant impact on the spruce forest is climate change, which is accompanied by changes in the temperatures and drying of the forest stands (Allen *et al.* 2010; Hanewinkel *et al.* 2013). Changes in the climate conditions can cause shifts of the tree species to higher altitudes and complete changes in the forest distribution (Küchler *et al.* 2015).

Based on the changes in the forest distribution related to climate changes, the utilization of other wood species in the production of wood-based composites is a possibility worth investigating. Research has focused on the possibilities of utilization of the softwood (larch, pine) and hardwood (oak, beech, birch, alder, poplar) species (Akrami *et al.* 2014; Ciobanu *et al.* 2014; Akyildiz *et al.* 2018). The utilization of the deciduous and coniferous wood species in wood-based composites is influenced by many

different factors: anatomical structure, adhesive penetration, wettability, and adhesive curing, among others.

The wood of deciduous and coniferous trees differs mainly in anatomical structure, especially the different pore systems (*e.g.*, diffuse- and ring-porous) or in surface chemistry; they exhibit different and provide wide ranges of swelling. The resulting strength of the bonded joint is influenced by the penetration of the adhesive to the wooden structure (Kamke and Lee 2007). Many factors influence the penetration behavior of the adhesive. These factors are related to the liquid properties of the adhesive, the anatomical properties of wood (roughness of the surface), the permeability of the wood, and, finally, the processing conditions. In addition, the type of penetration (lumen penetration *vs.* cell wall penetration) may have a different origin and function for the bond. The gradients of resin movement into the cell lumen are hydrodynamic flow and capillary action, contributing to the mechanical connection. In contrast, penetration into the cell wall is influenced by the diffusion gradient and by chemistry (Kamke and Lee 2007; Frihart 2009; Gavrilović-Grmuša *et al.* 2016).

In addition to adhesive penetration, wettability is crucial for good adhesion in wood bonding. The adhesive wettability of wood is usually evaluated by contact angle measurement (Shi and Gardner 2001). The contact angle is defined as the angle that occurs between the planes of solid substance, the tangent plane to the surface of the drop on the solid interface with the surrounding atmosphere, and the droplet. The contact angle is a function of the liquid's surface tension and the solid's surface energy (Chan 1994). The contact angle of the small droplets is more relevant for the wood-based panel industry.

An adhesive is commonly applied to wood strands as small droplets during the manufacture of OSB and other wood-based panels. The quantity of adhesive applied is low, and the coverage of strands is incomplete. During the pressing, strands come into contact, and bonding between strands may occur as a result of adhesive-to-wood and adhesive-to-adhesive contact (Smith 2005). Process optimization has mostly been done based on trial and error. Therefore, the development of predictive models that correctly describe the adhesive curing process is important to promote adequate bond strength and improve long-term performance (Carvalho 2008). The dynamics of adhesive curing and the development of adhesive bond strength during hot pressing affect production speed, energy consumption, and product quality.

The adhesive bond strength development can be assessed with an Automated Bonding Evaluation System (ABES). The ABES (Humphrey 1993) is a powerful and versatile technique for the evaluation of the “mechanical cure” of adhesive. This apparatus is based on a single lap shear test, but the joint is usually small (60 to 100 mm²), and the pressing conditions, such as temperature, pressing time, and pressure, can be adjusted. The overlapped strips could also be cooled before testing in shear mode, making ABES a suitable apparatus for the determination of shear strength as a function of adhesive type, catalytic system, adhesive application rate, cure temperature, time, substrate, *etc.*, in wood-based composite research (Humphrey 2009; Martins *et al.* 2012). The ABES simulates the bond strength development that occurs inside the mat during the hot pressing of wood-based composites. The shear strength of an adhesive joint depends on the number of bonds formed between the wood and the thermosetting resin. These bonds are formed during the adhesive cure. Therefore, adhesive cure depends on the temperature attained inside the mat, while the maximum shear strength is dependent on the number of wood-adhesive-wood bonds, which is intrinsically related to the amount of adhesive per area (resin load). Shear strength is extremely dependent on the resin load in the bonded joint. Increasing the

adhesive load increases the maximum shear strength. However, above 100 g per square meter, increasing adhesive load led to an adverse effect on shear strength. The ideal adhesive load for bonding evaluation was found to be around 100 g/m² (Cost *et al.*, 2013).

A comprehensive comparison of the different wood species with the different coverage of the adhesive has not been done. Melamine-urea-formaldehyde (MUF) resin, as a typical example of a wood-based composite adhesive, was used in this experiment. Seven different wood species were compared with the spruce, and the results show the similarities and differences in the utilization of the less-known wood species in wood-based composites. The main aim of this research was a comprehensive comparison of the shear strength achieved with different wood species at different coverage of the MUF adhesive. The percentage coverage of the surface was defined depending on the application of adhesive with stamps, *i.e.* percentage of coverage was during the adhesive application. The percentage of coverage of the adhesive in the final bond line (after hot pressing) was not the aim of the study.

The main motivation of the study was to determine the value at which the decrease of the bonding strength started and the determination of the minimal amount of the adhesive for different wood species for wood-based composite manufacturing. Establishing the methodology can aid in discovering the effect of different adhesives on underutilized wood species.

EXPERIMENTAL

Materials

Eight different adherends (spruce, pine, larch, beech, oak, poplar, birch, and alder veneers) were supplied by the company Jan Ficek Dřevovýroba s.r.o. The plain sliced (flat cut), radial pattern veneer sheets were cut into 117 × 25 mm strips and stored in an environmental chamber at 20 °C and 65% RH. Silekol® 311 melamine-urea-formaldehyde adhesive (MUF) (with 5% of melamine) was supplied by DDL Lukavec. Stamps with an area of 50 × 25 mm were used to apply the adhesive to the veneer strips. A grid with a mesh size of 1 mm² was drawn on the given surface. The raster was subsequently adjusted so that the number of overlaps with an edge size of 1 mm and the space between them formed the required percentage of the total area. Stamps were made for adhesive application of 10, 15, 20, 25, 30, 50, and 75% (Fig. 1).

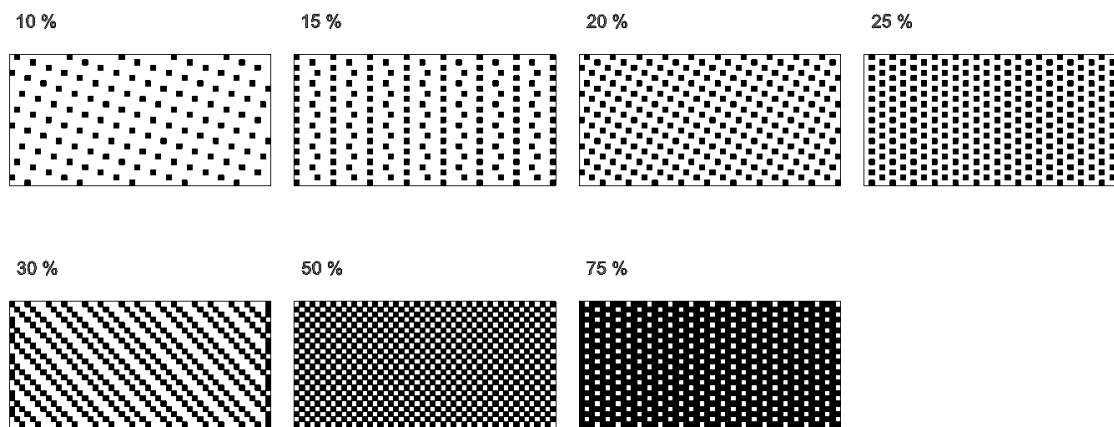


Fig. 1. Grids for variations of adhesive coverage of MUF resin on the veneer surface

A glass plate and an Elcometer 3540/3 manual film applicator with a thickness of 150 μm were used to precisely define the thickness of the adhesive layer. The samples were tested on the Automated Bonding Evaluation System (ABES).

AES_CONTROL_36A software was used to set the parameters of the test, control the device, and display the measured values.

Preliminary Tests

To determine the most suitable pressing time, beech veneers (5 test samples for each variant) with 100% adhesive coverage on the surface were used. The adhesive-coated samples were placed in the holders of the ABES test equipment. The samples were pressed in a small ABES press at a pressure of 2,69 MPa and a temperature of 180 $^{\circ}\text{C}$. The influence of pressing time on the strength of the bonded joint was monitored for pressing times 60, 120, 180, 240, 300, and 360 seconds (Fig. 3). The pressing time with the highest strength was then used throughout the entire experiment.

The pressing temperature was set at 180 $^{\circ}\text{C}$ during the entire experiment; the given temperature was identical to the temperature used in the production of OSB. The aim of the preliminary test was to determine the influence of the thickness of the veneers and the type of wood on the speed of heat transfer into the bonded joint of the samples, especially the time to reach 100 $^{\circ}\text{C}$ in the bond line. A thermocouple data logger probe (USB TC-08) from Pico Technology was placed between the samples with adhesive, which recorded the temperature during the test cycle in seconds, to determine the effect of the wood (Fig. 4).

Contact Angle

A water-based adhesive was used for the experiment; therefore demineralized water was used to determine wettability. To determine the surface wettability of the veneers used for testing, the method of measuring the contact angle of a water drop was applied to the surface of the samples (*i.e.*, sessile drop). Using a pipette (VWR EHP Pipettor, 1ch, 0.1-2 μL) on the samples placed on the positional table of the optical device (see System E/S, AdveX Instruments), a water drop (size: 1 μL) was applied. Using the See System software, droplet sizes were recorded in steps of 5 seconds (0 to 25 seconds); after determining the points on the drop, the software calculated the contact angle.

Methodology

The adhesive was applied to the glass plate, and a 150 μm thick layer was formed with an Elcometer 3540/3 applicator. A stamp was soaked in this layer of adhesive, and it was applied to one surface of the pairs of veneers 4 mm from the front edge of the sample (Fig. 2).

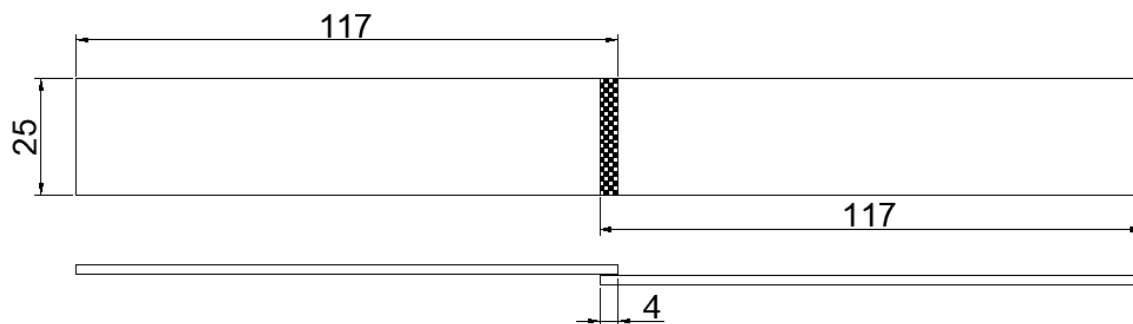


Fig. 2. Lap shear specimens for testing single lap shear strength on the ABES

To apply a 100% spread, the veneer samples were directly dipped into the created layer of adhesive. For each variant (wood and amount of adhesive), 10 samples were tested. The samples were fixed in the jaws of the testing device and pressed from both sides with heated pressing elements at a pressure of 1.37 MPa for softwoods and 2.69 MPa for hardwoods. Pressing was carried out at a temperature of 180 °C for 180 seconds. Then, the press was opened, and a shear test of the bonded joint was conducted (4 s delay). After testing, the maximum force required to break the bonded joint was recorded, and the area of the bonded joint was measured on the samples to calculate the shear strength.

Statistical Analysis

The data were processed in STATISTICA 10 software (StatSoft Inc., USA) and evaluated using a one-factor analysis of variance (ANOVA), completed with Tukey's honest significance test (HSD test).

RESULTS AND DISCUSSION

Preliminary Tests

The highest strength for beech veneers bonded with 100% adhesive coverage was achieved at 180 seconds of pressing time (Fig. 3).

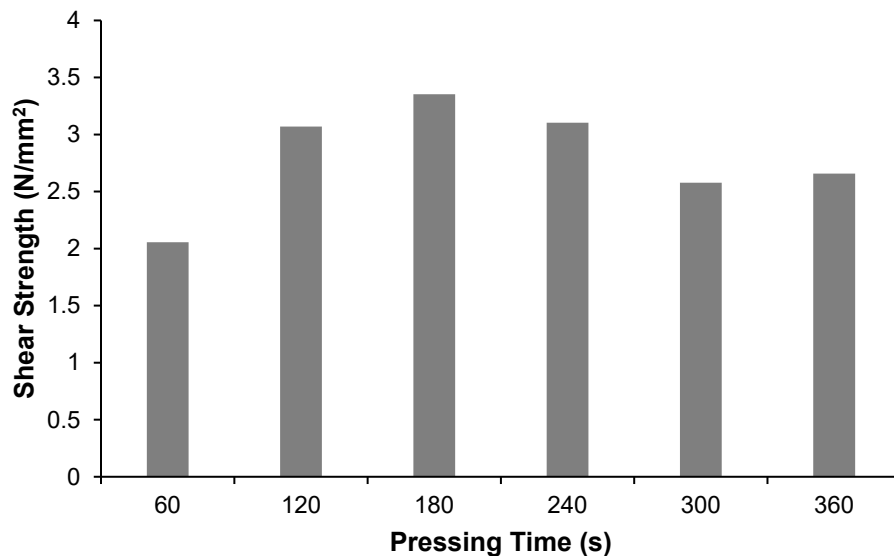


Fig. 3. Shear strength of MUF adhesive joint between two beech veneers for determining the optimal pressing time at a pressing temperature of 180°C

This pressing time was used throughout the further experiments on the ABES device. The rate of heat transfer to the bonded joint showed that the variance of the time to reach 100 °C in the bond line, depending on the thickness and type of the wood of the veneer strips, is 5 seconds, which does not have a significant impact on the total pressing time; therefore, the pressing time remains 180 seconds (Fig. 4).

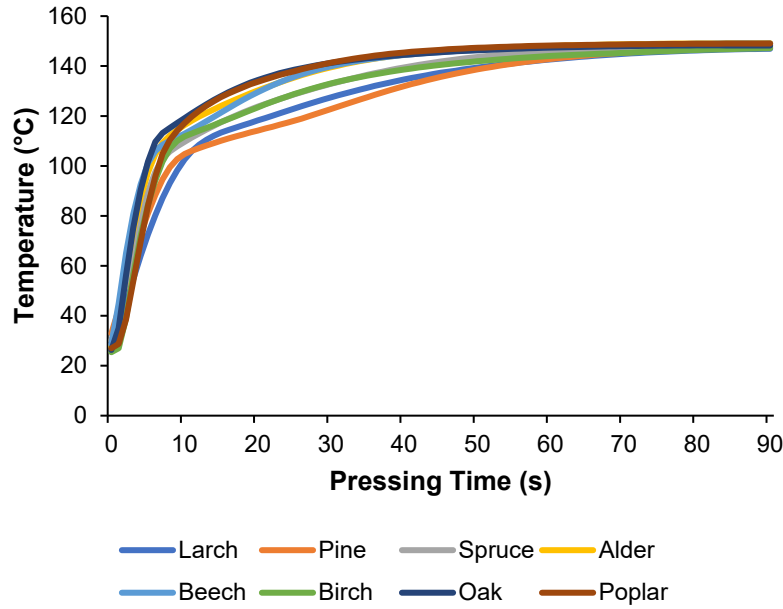


Fig. 4. The effect of the type of wood veneer on the rate of heat transfer to the bonded joint

Contact Angle of Wood Species

Figure 5 shows that all wood species have a hydrophilic character ($\theta < 90^\circ$) (Martines *et al.* 2005) and that proper wetting of the surface by water occurs. The decrease in wetting over time is not significant for spruce and pine samples due to the resin content and birch, which has a uniform wood structure.

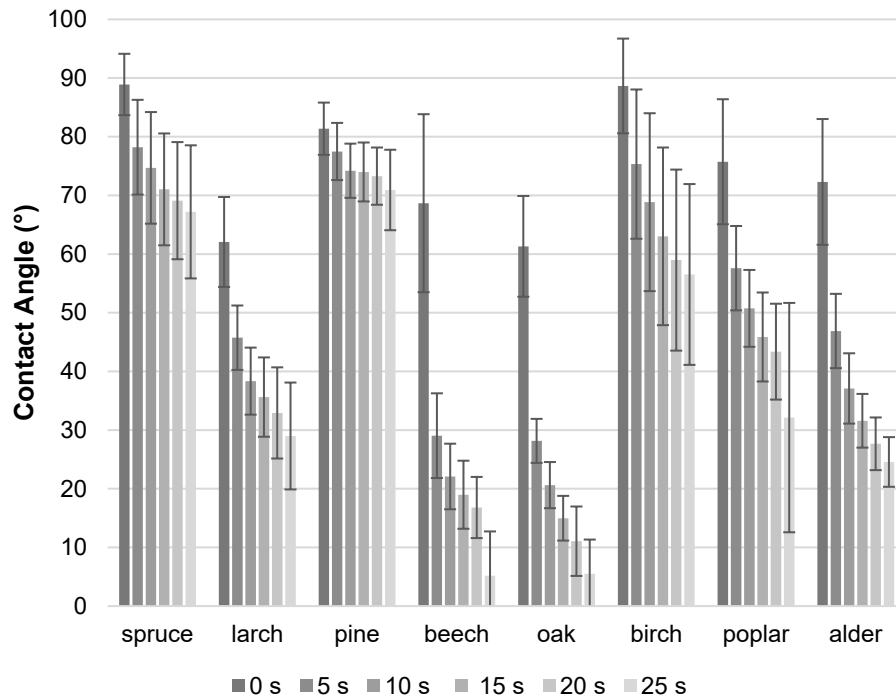


Fig. 5. Time dependence of the contact angle during wetting time for water on eight wood surfaces. Error bars represent standard deviation

The same results for birch veneers were shown in research focused on measuring the contact angle for the loose sides of rotary-peeled birch veneers (Bekhta *et al.* 2018). A significant decrease in wettability occurs for poplar and larch samples, when the contact angle after 25 seconds is below the limit defining super hydrophilic character ($\theta < 30^\circ$) (Martines *et al.* 2005). The contact angle achieved the limit defining super hydrophilic character within 5 seconds of the application of drops on the surface of beech and oak. In general, it can be concluded that the best bonding results would be obtained (in descending order) from oak, beech, larch, alder, poplar, birch, spruce, and pine veneers, because the contact angle was measured on the water and water-based adhesives were used in the experiment.

Adhesive Bond Performance

The average values for the density and single lap shear strength of eight wood species and different percentages of the coverage on the surface are shown in Table 1.

Table 1. Average Values for Density and Single Lap Shear Strength of Eight Wood Species and Different Percentages of the Coverage on the Surface

Species	100%			75%		50%		30%	
	Density	Shear strength	Tukey's test	Shear strength	Tukey's test	Shear strength	Tukey's test	Shear strength	Tukey's test
	(kg/m ³)	(N/mm ²)		(N/mm ²)		(N/mm ²)		(N/mm ²)	
Spruce	455	3.6 (0.6)	A,B,C	4.1 (0.8)	C, D	4.2 (0.4)	D	4 (0.3)	B,C,D
Larch	636	4.4 (1.3)	C,D	4.7 (0.7)	D	4.9 (0.4)	E,F	5 (0.4)	E
Pine	583	2.9 (0.8)	A	2.8 (0.5)	A, B	2.8 (0.5)	A	2.7 (0.9)	A
Beech	796	4.7 (0.2)	D	4.6 (0.4)	D	4.9 (0.6)	F	4.7 (0.5)	D,E
Oak	725	4.1 (0.3)	B,C,D	4.4 (0.3)	D	4.3 (0.5)	D, E	4 (0.4)	C,D
Birch	704	2.9 (0.2)	A	2.8 (0.4)	A	3 (0.3)	A, B	2.9 (0.6)	A
Poplar	496	3.5 (0.2)	A,B	4 (0.3)	C, D	4 (0.3)	C, D	3.9 (0.2)	B,C
Alder	585	3.7 (0.3)	A,B,C	3.5 (0.4)	B, C	3.4 (0.2)	B, C	3.4 (0.3)	A,B

Species	25%			20%		15%		10%	
	Density	Shear strength	Tukey's test	Shear strength	Tukey's test	Shear strength	Tukey's test	Shear strength	Tukey's test
	(kg/m ³)	(N/mm ²)		(N/mm ²)		(N/mm ²)		(N/mm ²)	
Spruce	455	3.8 (0.4)	B,C	3.6 (0.6)	C, D	3.2 (0.7)	B, C	2.4 (0.9)	A,B
Larch	636	4.9 (0.5)	D	4.5 (0.5)	E	4.1 (0.8)	D	3.7 (1)	C
Pine	583	2.7 (0.9)	A	2.6 (0.5)	A	2.2 (0.7)	A	2.2 (0.4)	A
Beech	796	4.7 (0.4)	D	4.4 (0.3)	E	3.8 (0.5)	C, D	3 (0.6)	A,B,C
Oak	725	4.3 (0.3)	C,D	4.2 (0.4)	D, E	3.6 (0.4)	C, D	3.2 (0.7)	B,C
Birch	704	2.9 (0.4)	A	2.8 (0.2)	A, B	2.6 (0.2)	A, B	2.2 (0.3)	A
Poplar	496	3.8 (0.3)	B,C	3.6 (0.3)	C, D	3.2 (0.5)	B, C	2.8 (0.5)	A,B,C
Alder	585	3.4 (0.3)	A,B	3.2 (0.4)	B, C	2.7 (0.3)	A, B	2.7 (0.3)	A,B

Means with the same letter in the column do not differ statistically by the Tukey's test ($\alpha = 0.05$). Numbers in parentheses represent standard deviation.

The highest shear strength for spruce veneers (control specimens) was obtained at 50% coverage on the surface of the samples, at 4.19 N/mm², but the graph (Fig. 6a) shows a slight increase from full coverage (100%) to 50% coverage and a subsequent decrease. According to Tukey's range test, there were no statistically significant differences between a coverage on the surface up to 100% and a coverage on the surface of 15%. The spruce samples bonded with MUF according to the EN 301 standard showed a shear strength of 6.5 N/mm², and EN 302 showed a shear strength of 5.8 N/mm², which was slightly higher because the adhesive was tested after fully conditioned and cured (Konnerth *et al.* 2006; Konnerth *et al.* 2016).

The highest shear strength for larch veneers was achieved when 30% of the total surface (100 mm²) was covered, which is 5 N/mm² (an increase of 19% related to the control specimen). According to the result of Tukey's test, there was no statistically significant difference between the strength of adhesive coverages from 100% to 10%. From the research according to the EN 301 standard (Konnerth *et al.* 2016; Liu *et al.* 2020), the shear strength of larch specimens was 9.6 N/mm², which is reasonable compared to the ABES results where specimens were tested hot.

The results of pine specimens (Fig. 6c) showed the highest strength, at 2.85 N/mm², for 100% adhesive coverage (a decrease of 36% related to the control specimen), but Tukey's test did not show any statistically significant difference between the other coverages. In a study according to the EN 302 standard (Wang *et al.* 2016), the shear strength of pine specimens was 9.8 N/mm². However, the study of Sahin Kol *et al.* (2015) showed a strength of 6.16 N/mm² (according to EN 205). Both studies were based on the lap shear tests of fully cured adhesive after conditioning, which is not the case in the ABES measurement.

It is clear from the graph (Fig. 6d) that the highest shear strength was obtained for the beech samples with a coverage of 50% (*i.e.*, 4.93 N/mm²), which is an increase of 18% related to the control specimen). Statistical evaluation of Tukey's test showed significant differences in the samples with a coverage on the surface of 15 % and 10%. The study of beech veneers, on ABES (ASTM D7998-19), bonded with UF resin at a strength of 5.7 N/mm² (Stöckel *et al.* 2010; Costa *et al.* 2013), determined the strength of beech samples bonded with UF resin to be 5 N/mm² and MUF resin to be 5.8 N/mm²; these results are comparable with the results made in this experiment. In contrast, the shear strength (EN 302) of beech specimens bonded with MUF was 12.3 N/mm² (Bachtiar *et al.* 2017), 10 N/mm² (Konnerth *et al.* 2006), and 11.1 N/mm² (Konnerth *et al.* 2016). The differences between the results of ABES (ASTM D7998-19) were only a portion of the results measured according to EN 302, because the ABES specimens were tested hot, immediately after bonding, and without any conditioning.

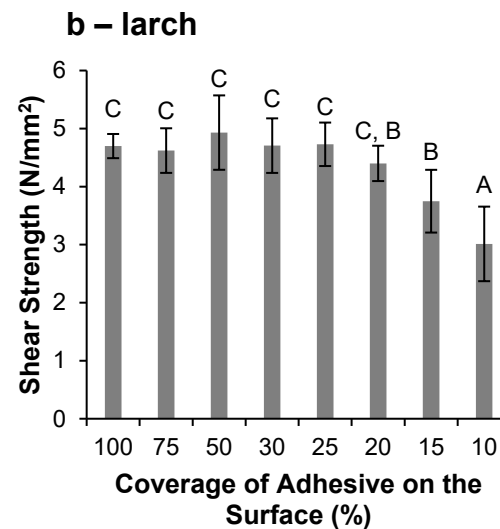
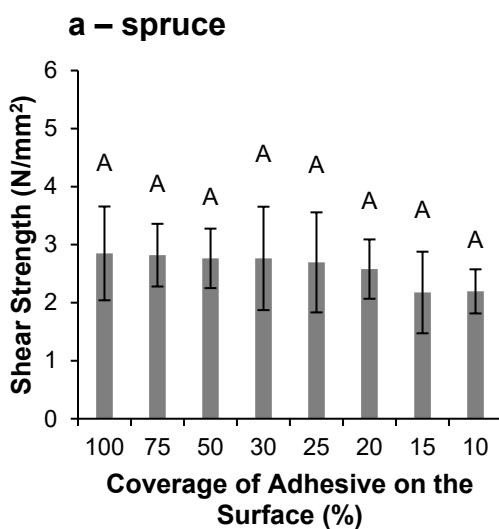
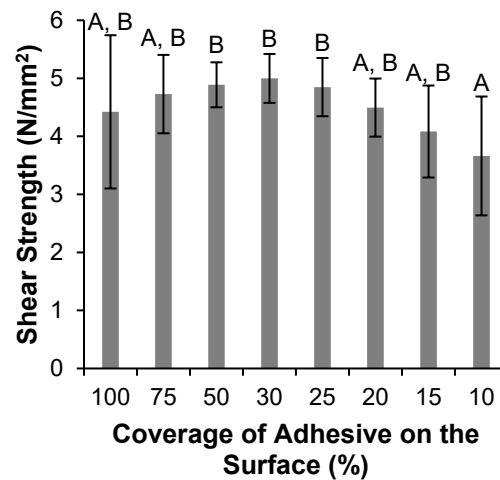
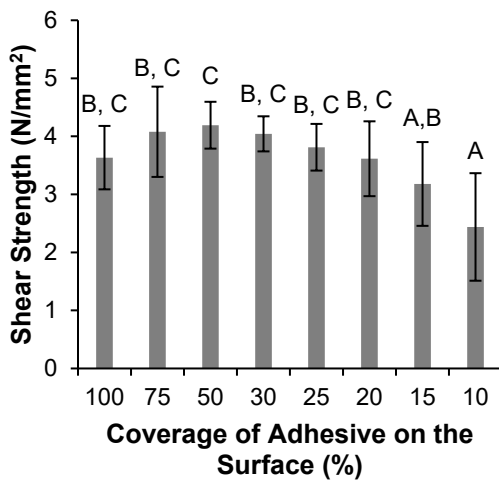
The highest strength was shown for oak samples (Fig. 6e) with 75% adhesive coverage at 4.44 N/mm² (an increase of 6% related to the control specimen); the samples with adhesive coverage of 15% and 10% showed significant decreases. In one study (Sahin *et al.* 2015), the shear strength was 8.74 N/mm²; in another (Konnerth *et al.* 2016), the resulting strength for oak samples was 10.25 N/mm² measured on the ABES.

The highest shear strength for birch veneers was achieved with 50% of coverage, 2.95 N/mm² (a decrease of 30% related to the control specimen); according to Tukey's test, there is no statistical difference between the coverage from 100% to 15%. Another study (Konnerth *et al.* 2016) determined, according to the EN 301 standard, that the strength of birch samples bonded with MUF resin was 11.5 N/mm², which is four times higher than the results in this experiment. The main reason for the difference is, as mentioned before:

standardized testing requires conditioning of the specimens and that the substrate be solid wood instead of veneer.

The highest shear strength for poplar veneers was achieved on 75% of the adhesive applied on the surface (4.01 N/mm²), which is a decrease of 4% related to the control specimen; according to Tukey’s test, there is no statistical difference between the coverages from 75% to 20%. In the research of Konnerth *et al.* (2016), according to the EN 301 standard, the strength for poplar samples bonded with MUF resin was 5.5 N/mm², which is comparable with our results because the ABES specimens were tested immediately, and the adhesive was not fully cured.

The highest shear strength for alder veneers was achieved on 100% of adhesive coverage (3.65 N/mm²); according to Tukey’s test, there was no statistically significant difference between coverages from 100% to 20% (a decrease of 13% related to the control specimen).



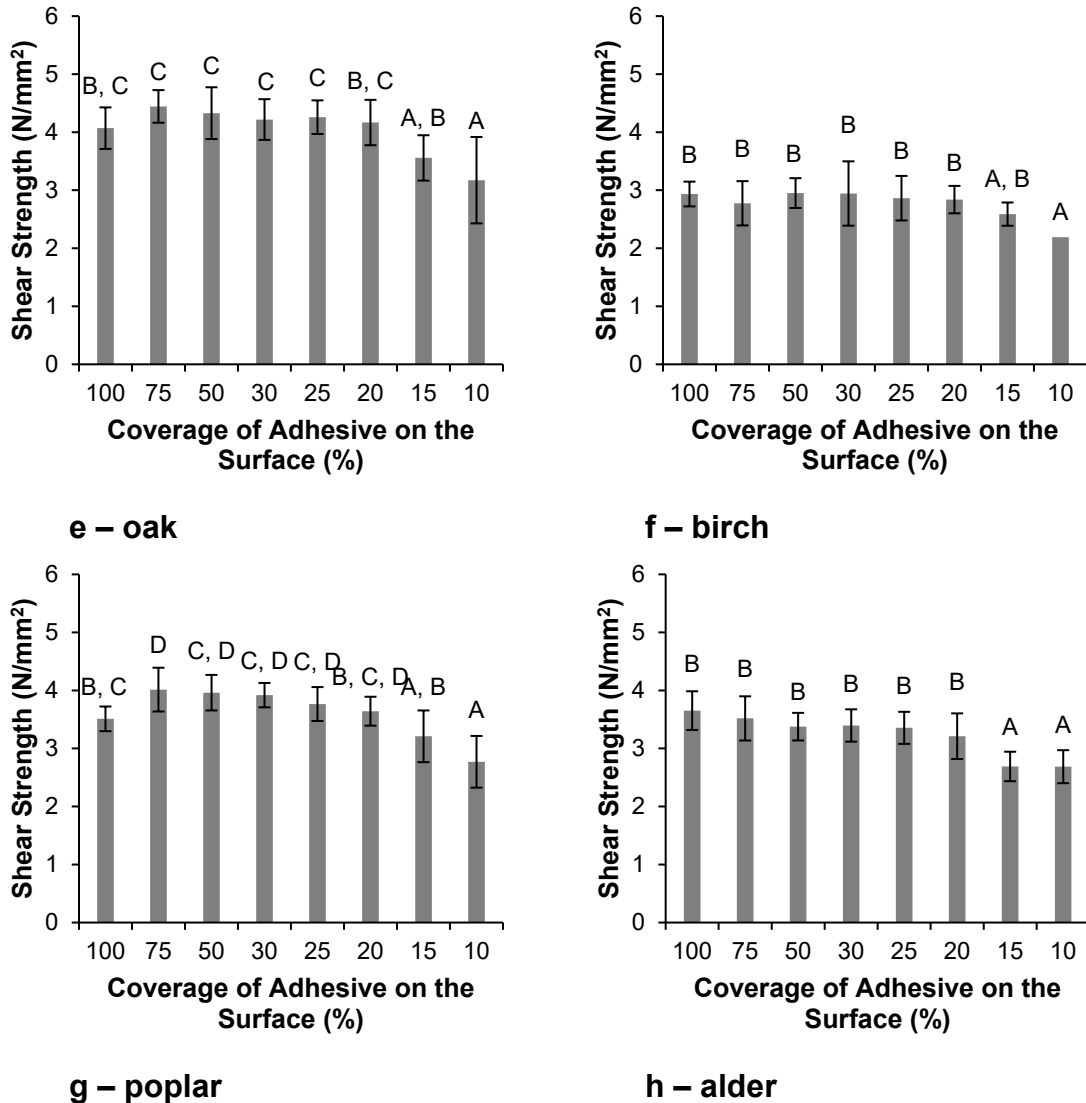


Fig. 6. The influence of the percentage of MUF adhesive coverage on bond strength for different wood species.

CONCLUSIONS

1. The resulting shear strength determined by the Automated Bonding Evaluation System (ABES) showed lower values than the results of other research obtained according to EN standards. This is related to the conditions of the ABES test; adhesive bonds were made of veneers instead of solid wood, and they were tested hot, and not fully cured, as compared to EN standard tests.
2. Based on the average values of shear strength, a slight increase was observed from 100% to 50% of adhesive coverage, with maximum at 50% of adhesive coverage, and a subsequent decrease in the shear strength to 10% of adhesive coverage for all tested wood species.

3. There was no statistically significant difference in the shear strength values in regard to adhesive coverage from 100% to 20%. Adhesive coverage of 20% was found to be sufficient for effective shear strength for all studied wood species, which showed that for the utilization of the studied wood species, it is not important to increase the resin consumption for the appropriate bonding properties for the wood-based panel industry.
4. Pine, birch, and alder did not reach at least the same values of shear strength as spruce (control) for this type of adhesive. It can be expected that wood-based panels from these wood species will not fulfill the standard requirements. The most promising results are for beech and larch and these two species can be the future leader in the single-species wood-based panels.
5. Based on the ABES results, there is a great potential for the replacement of the spruce with other deciduous or coniferous wood species in the production of wood-based panels with conventional adhesives.

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