Influence of Urea-formaldehyde Adhesive Modification with Beech Bark on Chosen Properties of Plywood

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The influence of beech bark concentrations as filler in urea-formaldehyde (UF) adhesives was investigated relative to the composite forming process and selected properties of final 5-layer beech plywood. Beech bark was used as filler to lower the wood processing waste production and decrease formaldehyde emissions. A combination of UF adhesives filled with different beech bark concentrations as the adhesive was used. Three different concentrations of beech bark, 15 wt.%, 20 wt.%, and 25 wt.% were used in the experiment. Urea-formaldehyde adhesive filled with 20 wt.% technical flour was used as a reference sample. The effect of the filler was studied *via* its temperature profile during pressing, mechanical properties in bending, water absorption, thickness swelling, and formaldehyde emissions after pressing. The time needed to reach the temperature between the beech veneers, at least 105 °C, which was equal to the final temperature filler-adhesive-wood matrix cross-linking, was also investigated during the pressing process. The measurements of the free formaldehyde emissions showed that for samples with non-zero bark concentrations there was a decrease of formaldehyde emissions by at least 46%.

Keywords: Beech bark; Beech plywood; Physical and Mechanical properties, UF adhesive modification

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INTRODUCTION

Plywood is one of the most important wood-based composites, with more than 90 million m^3 produced and a market value of approximately 20 billion U.S. dollars in exports and imports in 2014. In the last six years, a 40% production increase of plywood worldwide has been observed, and Slovakia has been an important manufacturer of shaped plywood. Currently, most of the plywood adhesives are formaldehyde-based; urea formaldehyde (UF), phenol formaldehyde, and melamine-formaldehyde resins are used widely in the plywood manufacturing industry, despite being non-renewable materials. The UF resin generates harmful formaldehyde emissions. Emissions occur mainly from the breakdown of formaldehyde-based resin in wood panels, and this poses a great hazard to human health by emitting carcinogens into the environment (Luo *et al*. 2015).

Formaldehyde emissions can be lowered by several methods, such as reducing the formaldehyde ratio during composition and the addition of more formaldehyde scavengers to the resin (Dongbin and An 2006; Hematabadi and Behrooz 2012; Costa *et al*. 2013).

However, the other physical and mechanical properties of wood panels are affected (Que *et al.* 2007; Mao *et al.* 2013). Formaldehyde emissions from wood-based products can be reduced during the manufacturing process, by a post-treatment of the wood-based products and a surface treatment. The surface treatment can both physically and chemically minimize formaldehyde diffusion. The most efficient route is the modification of the adhesives chosen (Kim *et al.* 2006; Bekhta *et al.* 2016).

Resin and filler are the main compounds in the adhesive formulation of plywood. Adhesive are often mixed with filler to increase the viscosity, control rheology, and reduce raw material cost. Wood's surface consists of small pores, and filler material is used to fill the holes, which helps to increase the bonding between the components. Filler is also used to reduce the penetration of resin into the small pores of wood (Pizzi 1994). Many types of natural fillers have been used by researchers, *i.e.*, cornstarch flour, tapioca flour, wheat flour, soybean, and polyvinyl acetate (PVAC) (Hojilla-Evangelista 2010; Hojilla-Evangelista and Bean 2011; Ong *et al.* 2012a, b; Bekhta *et al.* 2015). The high protein content in some of these fillers can enhance the bonding interaction between the wood and the adhesive.

The alternative use of biomass-based, biodegradable adhesives can solve the negative environmental impact by the wood processing industry, as they can be degraded within the environment by moisture and microorganisms (Hongyan *et al.* 2014). One of the possible available materials for the production of adhesive mixture fillers is the bark of domestic wood materials. Wood bark is the waste product during wood raw material processing. Depending on the type of wood, the wood bark for thin crust wood represents 6% to 9% of the dry mass (Račko and Čunderlík 2007). The annual wood mining volume in Slovakia is approximately 9.4 x 10^6 m^3 per year. The higher percentages of species harvested in Slovakia are the spruce (46.5%) and beech (32.4%). Wood processing in Slovakia produces approximately 7.5 x 10^5 m^3 of waste bark annually. Currently, more than half of the bark is used primarily as a cheap source of energy in pulp mills. Both bark incineration and landfilling can lead to environmental problems. Due to the abundance of ash in bark and the low sintering point of bark ash, the combustion of bark can lead to fouling, which damages the combustors and makes bark non-ideal fuel for energy production (Feng *et al.* 2013). Over the last few decades, bark-based panels bonded using urea formaldehyde (UF) resin have been highly investigated. In Pedieu *et al*. (2009), white birch inner bark particles were applied in the core layer of particleboard with wood fiber-reinforced surfaces. They discovered that almost 70% of wood fibers could be replaced by white birch inner bark while maintaining the required mechanical and physical properties. The best mechanical properties showed panels that were manufactured from 25% of wood fiber on the surface area and 9% wood fibers in the core layer. Another study (Pedieu *et al*. 2008) showed that the panels containing approximately 45% white birch outer substitution of wood particles maintained the mechanical and physical properties. Bark in the core layer significantly lowered formaldehyde emission and the thickness swelling of particleboard (Aydin *et al.* 2016). It was also shown that amounts of bark higher than 12.25% significantly worsened the mechanical strength, formaldehyde release, and the thickness swelling. Another study (Umemura *et al.* 2012) showed that the physical and mechanical properties required for furniture manufacturing could be achieved with 30% bark content.

The disadvantage of bark-based wooden panels is that below 200 °C the bark softens, while above this temperature, polymerization and partial degradation of bark components mainly contribute to the bonding (Marashdeh *et al*. 2011). Previous studies (Gao *et al*. 2011) showed that particleboard panels pressed at 230 °C had strength values

that were 9 times (molding´s modulus of rupture (MOR)) and 3 times (internal bonding strength (IBS)) higher than panels pressed at 170 °C. Bark panels are not commercially available due to their deteriorated appearance, inferior performance in other physical tests (large linear expansion and thickness swelling), and the extremely high temperatures and long press times that are needed for the panel production, which results from the poor thermal conductivity of bark (Blanchet *et al*. 2000). With the use of bark as the adhesive filler, these negatives can be overcome. Bark in the liquefied form has been also applied as a UF modifier and tested as a binder for particleboards production (Janiszewska *et al.* 2016)

In addition to input raw materials, other key parameters that influence the final composite properties are the composition of adhesive mixtures, amount of adhesive, and the parameters of the pressing process (temperature, pressure, pressing time) (Igaz *et al*. 2015, 2016). Optimization of the pressing process can increase the manufacturing speed, decrease expenses, reduce energy, and lower emissions (Thoemen *et al.* 2006). During the pressing process, complicated processes occur, including heat combination (fast heating and slow heating phases during plywood hot pressing) and moisture and pressure transfer (Liu *et al*. 2013). Their effect on the composite formulation is a function of the type and density of the wood and also of the wood fibers orientation (Carvalho *et al.* 2010; Gaff *et al.* 2015; Kminiak and Gaff 2015; Kvietková *el al.* 2015; Gaff *et al.* 2016). Key aspects here are also the adhesive properties, mainly temperature and the hardening time (Irle *et al*. 2012). With modification of the adhesive mixture, it is possible to drastically affect the pressing process and, therefore, the final composite properties. Research on plywood and other wood composites (mainly agglomerated), their properties, and the modification and optimization of the pressing process is applied globally. The research considers the effect of adhesives on heat transfer during the pressing process (Aydin 2014); influence of veneer modification on the mechanical properties of plywood (Aro *et al*. 2014); the optimized parameters of manufacture in the scope of the final composite mechanical properties (Demirkir *et al*. 2013; Gaff *et al*. 2017a,b); and the influence of adhesives based on carbon fibers (Aziz *et al*. 2015) or bark (Bai *et al*. 2012) on the plywood properties.

The main aim of this study was to investigate the influence of beech bark concentrations as the filler of urea-formaldehyde (UF) adhesives on the composite forming process and selected properties of final 5-layer beech plywood. The originality of this study is that we studied the effect of the filler *via* its temperature profile during pressing, mechanical properties in bending, water absorption, thickness swelling, and formaldehyde emissions after pressing.

Other studies have considered the advantages of the use of the milled bark as a filler in the adhesive compositions in terms of improving of the physical and mechanical properties of plywood. Investigations have been so far focused only on the bark of selected trees. Beech bark was processed in the framework of this research. This paper considers as well the various grits made with bark and its impact on the reduction of leakage of free formaldehyde adhesive compositions.

EXPERIMENTAL

Materials

As the basic material for the manufacture of the tested samples, beech (*Fagus sylvatica* L.) from central region Polana in Slovakia was used. The cuttings were hydrothermally modified and then processed into veneer form. After drying and

conditioning, they reached an equilibrium moisture content of 8% ± 1%. For the samples preparation, only defect-free veneers were used. The average thickness of veneers used was 1.23 mm. Veneers were cut to the dimensions of the pressing machine plates equal to 480 mm x 480 mm. From the veneers, files of five veneers were prepared so that the neighboring layers of veneers were rotated by 90°. After pressing, 28 pieces of plywood were produced.

Urea formaldehyde (UF) adhesive Kronocol U 350 (Supplier Diakol, Strážske, Slovakia) was used in powder form, and it was transformed to the liquid state according to the supplier's instructions. For the gluing of the reference samples, an adhesive mixture of UF adhesive filled with technical flour was used in the ratio of 20 g of filler to 100 g of adhesive. In the modified experimental samples, disintegrated beech bark was used as filler, with particles on average lower than 0.2 mm. The adhesive spread was, in all cases, equal to 180 g/m^{-2}. The gluing mixture was applied manually on the veneers from one side only with use of a roller. Beech bark was added to the UF adhesive in the 15, 20, and 25 weight percentages (wt.%), marked as 15 wt.%, 20 wt.%, and 25 wt.%. As a reference sample, urea-formaldehyde adhesive-filled was used with 20 wt.% of technical flour and marked as REF. Adhesive in powder form was used due to the fact that research results should be used in practice in the manufacturing plant based in the nearby of the Technical University in Zvolen. The manufacturing plant handled the entire volume of the adhesive for the purposes of the research sponsorship.

Beech bark was collected from the harvesting of beech raw material from a local forest enterprise. Bark was fresh, so it had been dried to a moisture content of 6 +/- 2%. Then, bark was directly ground in the chipper for particle production, and fine fraction was obtained by bark grinding with an electric coffee beans grinder. Bark powder was individually classified on a sieve shaker for 30 minutes. The marking and compositions of the gluing mixtures used in the experiments are shown in Table 1.

Table 1. Used Gluing Mixtures

	REF	15 wt.%	20 wt.%	25 wt.%
UF Adhesive (g)	100	100	100	100
Filler (g)	20 a	15 b	20 b	25 b
Hardener (g)	10	10	10	10

Note: a, technical flour as filler; b, defragmented beech bark as filler

Veneer pieces were pressed at 120 °C and the specific pressure of 1.8 MPa for 320 s. The pressing time computation was based on the condensation time needed for the UF adhesive cross-linking (180 s) and the theoretical time needed for heat transfer to the middle adhesive line (approximately 140 s). The theoretical time needed for heat transfer to the 2.3 mm depth was computed directly proportional to the time needed for heat transfer to 1 mm depth, which was 60 s.

The beech plywood after pressing were conditioned for 4 weeks at 20 °C ± 2 °C and a relative moisture content of 65% ± 5%. The final moisture contents of plywood after conditioning were computed according to ISO 13061-1 (2014) with the use of Eq. 1,

$$w = \frac{m_w - m_0}{m_0} \times 100 \quad (1)$$

where w is the sample moisture content (%), m_w is the mass of the tested sample with moisture content w (kg), and m_0 is the mass of the dry sample (kg). The drying of the

samples for the computation of the moisture content was performed according to ISO 13061-1 (2014). The samples were weighed and dried at 103 °C ± 2 °C. Samples were considered dry when the mass difference during the time interval of 6 h did not exceed 0.5% of the sample mass. After the samples finished drying, the samples were cooled in a desiccator and subsequently weighed rapidly; therefore, the changes in the moisture content can be neglected. The final moisture content was rounded to a precision of 0.5%. The average moisture content of the samples after conditioning was 12% ± 1%.

Methods

Temperature-time dependence in the adhesive line during the pressing process

Due to the heat transfer research in the pressing process, the temperature-time dependence during the pressing process was monitored. The temperature measurement was performed using 12 pieces of K thermocouples located at the surface of the top veneer and next to two neighboring adhesive lines, GL1 and GL2, as shown in Fig. 1b. Because of the symmetric heating from both pressing plates, the thermocouples were localized only in the upper half of the pressed pieces thickness. In every measured surface 4 thermocouples were localized, according to Fig. 1a. Thermocouples were installed onto the adhesive line after binding and before the completion of the pressing piece. Data acquisition in the pressing process was performed with a multimeter GW Instek GDM-8255A (Good Will Instrument Co., Taipei, Taiwan) with a 16-channel scanning cart, GDM-SC1.

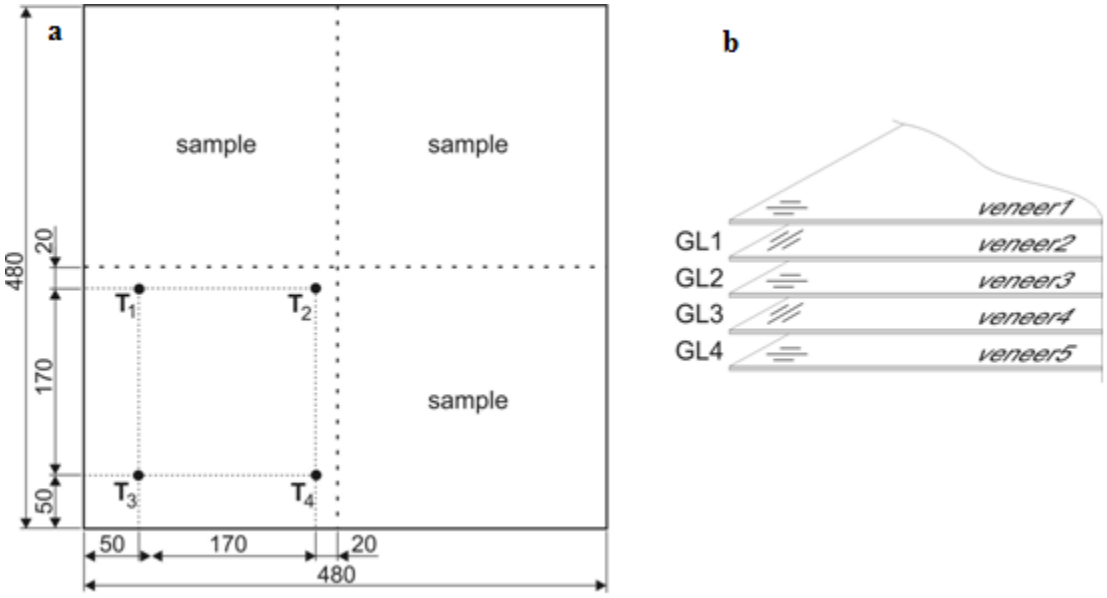

Fig. 1. (a) Denomination of thermocouples position, denomination of glue line, and **(b)** the scheme of sample preparation

The effect of beech bark concentration on the relaxation time *r*, and on the time needed to reach 105 °C in the adhesive line, denominated as t_{105}, was evaluated. The ambient temperature (T_{amb}) in all experiments was 18 °C, and the temperature of the pressing plates T_{max} was 120 °C. The temperature function can be found in Eq. 2,

$$T = T_{max} - dT_{max} \times e^{-1.t/\tau} \quad (2)$$

where T_{max} is the temperature of the pressing plates (°C), dT_{max} is the maximal temperature difference (°C), and r is the relaxation time (s).

The maximal temperature difference in the selected adhesive line can be found as the difference between the maximal temperature in adhesive line T_{max} and the ambient temperature T_{amb}, *e.g.*, Eq. 3,

$$dT_{max} = T_m - T_{amb} \quad (3)$$

and according to the fact that the only heat sources in the experiments were pressure plates, it is equal to Eq. 4,

$$dT_{max} = T_{max} - T_{amb} \quad (4)$$

Thus, the temperature-time dependence can be found in Eq. 5,

$$T = T_{max} - (T_{max} - T_{amb}).e^{-1.t/\tau} \quad (5)$$

from which 105 °C was reached in the time t_{105}, and is equal to Eq. 6,

$$t_{105} = \ln\left(\frac{T_{max} - T_{amb}}{T_{max} - 105}\right) \times \tau \quad (6)$$

Lastly, the total time needed to reach 105 °C in the adhesive line can be found in Eq. 7,

$$t_0 = t_{105} + t_{onset} \quad (7)$$

where t_{onset} is the time (in seconds) needed to transfer heat to the selected adhesive line and is lower when the distance to the heat source is lower.

Fitting procedure

The values of parameters T_{max} and r were acquired by a parametric non-linear fitting procedure based on the minimal least squares method. In this method, parameters values are increased by a defined step and an algorithm finds the best parameter values (minimal difference and highest correlation between the measured and theoretical values).

The quality of fit was studied in terms of the statistical parameters *RMSE* and *r*. These are the root mean square error and the average temperature difference between the measured data and theoretical data; both parameters should be as small as possible. The coefficient of correlation, *r*, is between the experimental and theoretical values, and should be close to 1. It also means that with high values of *r*, larger than 0.98, the experimental data can be described by Eq. 5.

Formaldehyde release

The emission of free formaldehyde was determined *via* JIS A 1460 (2001). The test principle is measurement of free formaldehyde emission from plywood samples absorbed by distilled water during 24 hours in the glass desiccator.

The samples tested were prepared from every type of plywood with dimensions (150 ± 1) x (50 ± 1) mm (length x width) with a total of 1735 cm^2 (10 pieces). They were then placed in a desiccator with 300 mL of distilled water. After 24 h, the samples were removed from the distilled water and prepared for spectroscopy. To prepare the tested solution, 25 mL of distilled water from the desiccator was mixed with 25 mL of acetylacetone and ammonium acetate solution. The samples were then placed into a 65 °C ± 2 °C water bath for 10 min and subsequently cooled to ambient temperature. The

formaldehyde content was determined by a SPEKOL 221 spectrophotometer (Carl Zeiss, Jena, Germany) *via* the measurement of water absorption at wavelength 415 nm.

Bending strength and Young's modulus of elasticity in bending

The effect of the adhesive mixture modification, with the addition of beech bark, on the mechanical properties was tested by the three points bending test according to STN EN 310 (1998). The samples were prepared according to EN 326-1 (2014).

From experimentally obtained plywood *via* pressing, the samples were prepared for mechanical testing in the longitudinal and transverse directions. The dimensions of the tested samples were 50 mm ± 1 mm x 165 mm ± 1 mm (width x length) and were measured with a 0.01 mm accuracy (thickness, width) and 0.1 mm (length). The mechanical properties were tested using the TIRAtest 220 (Tira, Schalkau, Germany), with 90 s ± 30 s intervals.

From the strength-deformation curve a maximal force *Fm* was reached. The breaking and bending strength (σm) were then computed according to STN EN 310 (1998), and using Eq. 8,

$$\sigma_m = \frac{3F_m l}{2bh} \quad (8)$$

where σ_m is the bending strength (MPa), F_m is the maximal force before sample break (N), l is the distance between supports (mm), b is the width of the tested samples (mm), and h is the thickness of the tested sample (mm).

Young´s modulus at bending was computed from the force-deformation diagram according to STN EN 310 (1998) and STN EN 49 0116 (1980) with the use of Eq. 9,

$$E = \frac{l^3(F_2 - F_1)}{4bh^2(a_2 - a_1)} \quad (9)$$

where E is the Young´s modulus of elasticity in bending (MPa), l is the distance between supports (mm), b is the width of the tested samples (mm), h is the thickness of tested sample (mm), $F_2 - F_1$ is the force increase in the linear part of the force-deformation curve, and $a_2 - a_1$ is the change of deformation in the middle of sample corresponding to the force change ($F_2 - F_1$).

Water absorption

The water absorption of plywood was specified according to STN EN 49 0164 (1980). For the water absorption, test samples were prepared with 50 mm ± 0.1 mm x 50 mm ± 0.1 mm (width x length) dimensions. The samples' masses were set with 0.01 g accuracy. Tested samples were not in contact with each other or with the wall of the container. Samples were submerged 10 mm beneath the water surface. During the test the water temperature was 20 °C ± 1 °C.

After 2 h, the samples were removed from the water and subsequently dried with filtration paper and weighed with 0.01 g accuracy. Once the mass was constant, the samples were again placed in water. The process of mass determination was repeated after 24 h from the first placement in the water, and the samples were themselves weighed. The water absorption was then computed with 0.1% accuracy according to Eq. 10,

$$n_a = \frac{m_w - m_k}{m_w} \times 100 \quad (10)$$

where n_a is the water absorption (%), m_w is the mass of the sample after removal from water (kg), and m_k is the mass of the sample before placement in water (kg).

Thickness swelling

The thickness-swelling test was performed according to STN EN 317 (1995). For the thickness-swelling test, samples were prepared with the dimensions 50 mm ± 0.1 mm x 50 mm ± 0.1 mm (width x length). The thickness of the samples was determined with 0.01 mm accuracy. Tested samples were not in contact with each other or with the wall of the vessel. Samples were submerged 10 mm beneath the water surface, and the temperature of the water was 20 °C ± 1 °C.

Tested samples were removed from the water after 2 h, and they were subsequently dried with filtration paper and measured (thickness) with 0.01 mm accuracy. After the thickness set, samples were again placed in the water. The process of thickness determination was repeated after 24 h from the first placement in water and the samples thickness was measured. The thickness-swelling were then computed with 0.1% accuracy according to Eq. 11,

$$\beta_{hw} = \frac{h_w - h_k}{h_w} \times 100 \quad (11)$$

where β_{hw} is the thickness swelling (%), h_w is the thickness after removal from water (mm), and h_k is the thickness of the sample before placement in water (mm).

RESULTS AND DISCUSSION

Thermal characteristics of mixing process

The following tables present the effects of beech bark concentration on the relaxation time *r*, and on the total time needed to reach 105 °C t_0, and the maximum temperature in the adhesive line T_{max} for all studied adhesive lines GL1, GL2, and GL3. In Table 2, the results for beech plywood with 0 wt.%, 15 wt.%, 20 wt.%, and 25 wt.% are presented. Table 3 presents the comparison of time t_0 and T_{max} for beech bark concentrations of 15 wt.%, 20 wt.%, 25 wt.%, and the reference sample (REF). Table 4 presents the statistical parameters *RMSE*-average value for the chosen wt.% and the *r*-average value for the chosen wt.%. Table 5 presents the increase of total time t_0 of the selected samples with 15 wt.%, 20 wt.%, and 25 wt.% compared to reference sample, REF.

Table 2. Effect of Beech Bark (0 wt.%, 15 wt.%, 20 wt.%, 25 wt.%) Concentration on the Chosen Properties

REF	*r* (s)	t_0 (s)	T_{max} (°C)
GL2 (0%)	24.23 ± 1.99	61.85 ± 0.85	117.60 ± 2.41
GL3 (0%)	31.79 ± 1.98	79.24 ± 4.63	
GL2 (15%)	21.64 ± 1.64	42.63 ± 4.23	123.61 ± 0.95
GL3 (15%)	38.47 ± 3.26	68.85 ± 4.51	
GL2 (20%)	23.64 ± 3.20	56.64 ± 10.06	122.90 ± 4.13
GL3 (20%)	41.11 ± 5.72	84.02 ± 9.55	
GL2 (25%)	22.10 ± 1.97	47.48 ± 2.58	122.92 ± 3.55
GL3 (25%)	45.33 ± 9.65	81.52 ± 10.94	

Table 3. Comparison of Chosen Properties

Sample	REF	15 wt.%	20 wt.%	25 wt.%
t_0 (s)	79.24	68.85	84.02	81.52
T_{max} (°C)	117.60	123.61	122.90	122.92

Table 4. Statistical Parameters

Sample	REF	15 wt.%	20 wt.%	25 wt.%
RMSE (°C)	2	3.64	4.24	4.05
r (-)	0.990	0.992	0.989	0.990

Table 5. Effect of Beech Bark Concentration on the Total Time dt_0 Increase

Sample	REF	15 wt.%	20 wt.%	25 wt.%
dt_0 (%)	-	-13.11%	6.03%	2.88%

Based on the average values of *RMSE* shown in Table 4 and the results of T_{max} (°C) in Table 3, it was concluded that the fitting procedure from the experimental temperature-time dependences resulted in the set value of the temperature of pressing plates. According to correlation coefficient values, it was concluded that Eq. 5 was valid.

From Table 5, it is evident that the addition of beech bark above 15 wt.% resulted in the decrease of total time needed for filler-adhesive-wood matrix cross-linking.

In the frame of the results of the chosen properties, it was concluded that the pressing process did not result in a homogenous beech bark distribution in the adhesive line and, therefore, to the high variability of time t_0 and .

Formaldehyde release

Formaldehyde release was performed under JIS A 1460 (2001). The results for samples REF and samples with beech bark concentrations 15 wt.%, 20 wt.%, and 25 wt.% are plotted in Fig. 2.

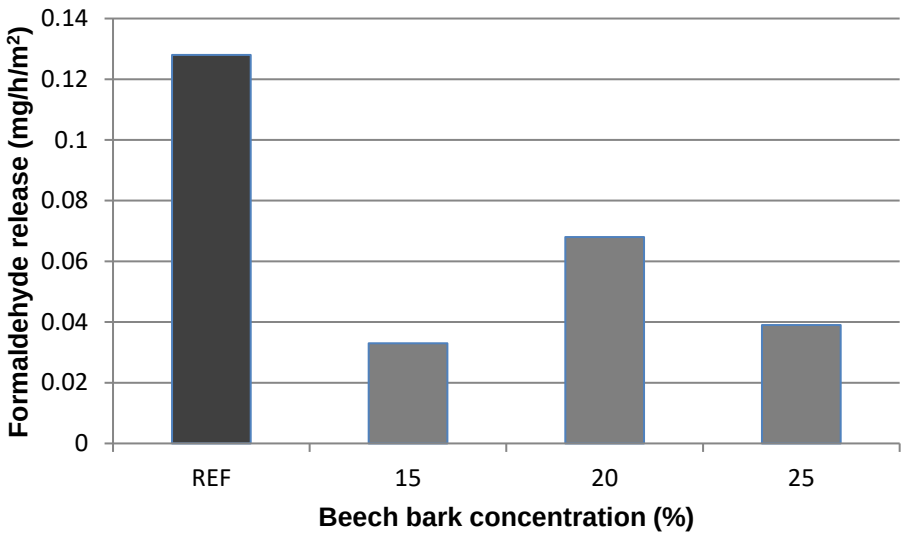

Fig. 2. Effect of bark concentration on formaldehyde release

Mechanical properties of mixtures

The samples of plywood prepared with different beech bark concentrations were mechanically tested *via* the three point bending test. The test method determined two important characteristics of mechanical behavior of plywood, namely the Young´s modulus of elasticity and bending strength, according to Eqs. 8 and 9. From these results, two mechanical properties evaluated the mixing process in such way that mixtures with bad adhesion between layers had lower values of these mechanical properties. Table 6 shows the results of the mechanical properties.

Table 6. Comparison of Chosen Mechanical Properties

Sample	REF	15 wt.%	20 wt.%	25 wt.%
σ_m (MPa)	98.70 ± 11.50	113.90 ± 3.41	107.50 ± 6.85	105.25 ± 3.59
E (GPa)	8.71 ± 0.77	10.34 ± 0.54	9.98 ± 0.95	9.60 ± 0.85

From Table 6, it was evident there was a decrease in the studied mechanical properties with raised beech bark concentration, which was also observed in the thermal characteristics. It was also evident that the mechanical properties of composites with beech bark were higher than for the reference sample with 0 wt.% of beech bark; thus the addition of beech bark improved the mechanical properties. The decrease of the studied mechanical properties of composites with beech bark concentration was attributable to the poorer adhesion of layers, which negatively affected the strength and modulus of elasticity, and thus the final composite.

Thickness swelling and water absorption test

The prepared samples of plywood with different beech bark concentrations were also tested for water absorption after 2 h and 24 h and thickness-swelling. The average values of water absorption and thickness swelling were computed according to Eqs. 10 and 11. Table 7 shows the results of water absorption and thickness-swelling.

Table 7. Comparison of Chosen Mechanical Properties

Sample	REF	15 wt.%	20 wt.%	25 wt.%
n_2 (%)	17.29 ± 1.36	17.03 ± 0.66	17.08 ± 0.31	18.52 ± 0.49
n_{24} (%)	41.94 ± 3.71	37.75 ± 2.72	33.22 ± 1.12	36.54 ± 0.86
β_2 (%)	2.94 ± 0.35	5.11 ± 0.94	4.47 ± 0.54	5.31 ± 0.08
β_{24} (%)	6.51 ± 0.67	9.10 ± 1.05	9.04 ± 1.19	10.19 ± 0.94

From Table 7 it is evident that the water absorption and thickness-swelling did not change noticeably with rising beech bark concentration. It was also evident there was decrease of water absorption with rising beech bark concentration, and an increase in thickness-swelling with rising beech bark concentration.

CONCLUSIONS

1. The replacement of technical flour as a filler with disintegrated beech bark led to a decrease of free formaldehyde emissions in the final plywood regardless of beech bark weight percentage. The free formaldehyde emissions were decreased from 46% to 75%.
2. The replacement of technical flour with beech bark in the adhesive mixture at all investigated concentrations increased the observed mechanical properties of plywood.
3. The most noticeable increase of bending strength and Young´s modules of elasticity was recorded at 15 wt.% beech bark in the adhesive mixture ($\Delta\sigma_m$ = 15.4%, ΔE = 18.7%). With an increased concentration of beech bark the observed mechanical properties decreased.
4. The most noticeable decrease of the total time of plywood preparation was recorded at 15 wt.% beech bark in the adhesive mixture (Δt = -13.1%). With an increased concentration of beech bark, the observed time increased.
5. The water absorption of the final plywood with beech bark as filler was lower, but the decrease was not statistically significant.
6. The use of beech bark in the adhesive mixture for all investigated concentrations led to a dramatic increase of the thickness-swelling ($\Delta\beta$ = 39% to 80%).
7. Based on the experimental measurements, it was possible to conclude that with the exception of the thickness swelling results, the replacement of technical flour with disintegrated beech bark leads to maintained or improved 5-layer beech plywood properties.
8. The total or partial replacement of technical flour with disintegrated bark has the potential to decrease negative environmental impact produced by the wood processing industry and gives possibilities for valuation of waste products produced by wood raw material processing.

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