Selected Properties of Flat-Pressed Wood-Polymer Composites for High Humidity Conditions

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This study investigated the possibility of applying flat-pressed wood-polymer composites in conditions of high humidity. The experiment involved three variants of wood-polymer composite panels 16 mm thick, and 680 kg per m^3 density. The wood particles were bonded with polyethylene. The share of polyethylene in the core layer was fixed at 50%, while in the face layers the content was varied (40%, 50%, or 60%). The following parameters were examined: modulus of rupture (MOR), modulus of elasticity (MOE), internal bond (IB), screw holding (SH), thickness swelling (TS), water absorption (WA), susceptibility to drilling and milling, wettability and surface free energy, and resistance to mold. The results were compared to particleboard glued with urea-formaldehyde resin. The wood-polymer composite had lower MOR and MOE values and similar IB and SH values. The panels indicated a remarkably higher water resistance (lower TS and WA values) with good surface wettability and high resistance to mold fungi. Additionally, the composites were easier to machine, *e.g.* drilling or milling, in comparison to standard particleboards.

Keywords: Wood-polymer composite; Mechanical and physical properties; Machining; Resistance to mold

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INTRODUCTION

The range of wood-polymer composites (WPC) applications in new material solutions has been continually expanding. Aside from the standard WPC composites produced by extrusion or injection, the concept of the bonding of wood chips with use of thermoplastics has been developed. Research conducted in this field indicates the possibility of producing boards with favorable operational and quality parameters using methods similar to wood-based board pressing technology (Youngquist *et al.* 1994, 1995; Boeglin *et al.* 1997; Borysiuk 2004, Borysiuk *et al.* 2004, 2006). The strength of wood-polymer composites is determined by the quantitative share of wood and thermoplastic particles, the size of the wood particles, the type of thermoplastic, the addition of bonding substances, and the manufacturing methods (Stark and Berger 1997; Błędzki and Faruk 2004). The composites achieve optimal resistance to bending forces at a wood particle content in the range of 40 to 60% (Stark and Berger 1997; Borysiuk *et al.* 2004; Chen *et al.* 2006; Djiporovic *et al.* 2006; Borysiuk *et al.* 2008). In general, WPC composites have lower MOR and MOE values and comparable tensile and compressive strength values. An important advantage of wood-polymer composites over other wood-based panels is their water resistance (Falk *et al.* 1999; Sellers *et al.* 2000). The hydrophobic properties deteriorate with the increase in the proportion of wood particles in the composite

(Zajchowski *et al.* 2005) and the increase in porosity of its structure (*e.g.*, as a result of foaming). Along with the increasing humidity, WPC composites are more susceptible to attack by both home-grown fungi (basidiomycetes) and mold fungi. The degradation effect is dependent on the weight of wood particles, their size, and the species from which they have been manufactured, as well as from the possible use of other additives (Verhey and Laks 2002; Barton-Pudlik *et al.* 2017).

The appropriate selection of materials to produce elements such as bathroom furniture, is important due to their cyclic contact with high humidity air, temperature variability, as well as frequent contact of the material with liquid water. Wood-based materials selected for these constructions must meet several requirements, the most important being resistance to water. Wood-based materials show high hygroscopicity, which results in the free exchange of water vapor contained in the material with the environment. The use of traditional boards with resistance to water does not guarantee sufficient protection against the destructive effects of moisture and related degradation factors (Thoemen *et al.* 2010; Niemz and Sonderegger 2017). A good solution for applications in an environment with high relative humidity (*e.g.*, as elements of bathroom or kitchen furniture) may be wood-polymer composites.

The research evaluates selected properties determining the applicability of wood-polymer composites in production of furniture elements exposed to high humidity or liquid water.

EXPERIMENTAL

Materials

Tests were carried out using industrial coniferous particles applied to the face and core layers of the particleboards. The wood particles moisture content was 8%. The panels fractional composition is shown in Table 1.

Table 1. The Fractional Composition of Particles Used in this Study

Fraction (mm)	Fractional composition (%)		
	Wood particles		Thermoplastic particles
	Face layer	Core layer	
6.0	13.0	-	0.2
4.0	19.0	-	5.7
2.0	51.0	0.6	37.3
1.25	13.0	14.3	39.7
1.00	2.6	25.5	4.7
0.63	1.0	30.0	8.4
0.49	0.2	11.4	0.9
0.385	0.1	7.8	0.9
< 0.385	0.1	10.4	2.2

Post-consumer HDPE polyethylene obtained from used film and packaging was applied in the tests. The raw material was ground to a similar size as the particles used in the face and core layers, respectively. Grinding of the thermoplastic was carried out using a laboratory mill. The average melting point of polyethylene was 120 °C. Due to the manufacturing method of the boards (cold forming), similarly to Rahman *et al.* (2013) and Lyutyy *et al.* (2018) no compatibilizer was applied.

Boards

A three-layer wood-polymer board with dimensions of 330 by 330 by 16 mm^3 and a density of 680 kg per m^3 was produced. Individual panel variants were characterized by a variable contribution of thermoplastics in the face layers (Table 2). The reference material was a particleboard glued with UF resin. Commercially available urea-formaldehyde resin with a U per F molar ratio equal to 1 to 1.2, 65 wt% solids content, and a viscosity of 230 MPa at a temperature of 20 °C was used as the binder.

Table 2. Characteristics of Panel Construction

Parameters	Value
Face layers contribution	34%
Wood-polymer composites (variant A, B, and C)	
Contribution of polyethylene in the core layers	50%
Contribution of polyethylene in the face layers	
Variant A	40%
Variant B	50%
Variant C	60%
Particleboards glued with UF resin - variant D	
Degree of sealing of core layers	8%
Degree of sealing of the face layers	10%

The appropriate proportions of raw materials (thermoplastic and wood particles) for wood-polymer composites were cold mixed. The mats were then formed manually from the obtained mixtures. The boards were manufactured in an electrically heated single-shelf laboratory press. The pressing parameters are presented in Table 3. In the first stage, the mats were hot pressed. After plasticizing the polyethylene, the mats were transferred into the cold press for cooling. Hot and cold pressing was carried out using spacers to determine the thickness of the plates. Finally, the manufactured WPC boards were air-conditioned for 7 days under laboratory conditions (20 ± 2 °C and 65 ± 5% humidity).

Table 3. Parameters for Pressing the Plates

Parameters	Variant A, B, and C	Variant D
Maximum specific pressure	2.5 MPa	2.5 MPa
Temperature of hot pressing	200 °C	200 °C
Temperature of cold pressing	20 °C	-
Time of hot pressing	300 s	288 s
Time of cold pressing	300 s	-

Mechanical and Physical Properties

Several physical and mechanical properties of the boards were tested in this study. The density was tested according to the EN 323 (1999) standard and the density profile using the Laboratory Density Analyzer DAX GreCon. The density measurement was made every 0.02 mm at a measurement speed of 0.05 mm per s. The modulus of rupture (MOR) and modulus of elasticity (MOE) were tested according to the EN 310 (1994) standard. The internal bonding (IB) was tested according to the EN 319 (1999) standard. The screw holding (SH) was tested according to the EN 320 (2011) standard. Thickness swelling (TS) and water absorption (WA) after immersion in water for 2h and 24h were tested according to the EN 317 (1999) standard. Every test included ten replicates of each variant. Additionally, the susceptibility to drilling and milling, contact angle and surface free energy, and resistance to biodegradation were evaluated for the manufactured panels.

Susceptibility to Drilling and Milling

Plate machinability tests were carried out using a Busellato Jet 130 CNC machining center (Casadei-Busellato, Thiene, Italy). Through-hole drilling (throughout the entire thickness of the plate) was done with a new, 8 mm diameter, single-edge, polycrystalline DPI diamond drill (Leitz, GmbH and Co. KG, Stuttgart, German). The rotational speed was set to 6000 rpm, the feed speed was set to 1.2 m per min, and the feed per revolution was set to 0.2 mm. During drilling, F_z axial force signals were recorded using a Kistler 9345A piezoelectric force sensor (Kistler Group, Winterthur, Switzerland) with a sampling frequency of 12 kHz. For each variant, ten cuts were made. The RMS of axial force signals was evaluated.

A Faba single-edge milling head (Faba S.A., Baboszewo, Poland) with a 40 mm diameter WC-Co cemented carbide knife was used for milling. During the test, grooves 40 mm wide (tool diameter) and 5 mm deep were milled. The rotational speed was set to 18000 rpm, the feed speed was set to 2.7 m per min, and the feed per revolution was set to 0.15 mm. Two components of cutting force were recorded during milling (F_x was in accordance with the tool feed direction, and F_y was perpendicular to F_x) using a Kistler piezoelectric force sensor with a sampling frequency of 12 kHz. The resultant cutting force F_w (geometric sum of the vectors F_x and F_y) was determined based on two perpendicular components. For each variant, seven cuts were made. The mean value of the feed force signals was evaluated.

Wettability (Contact Angle) and Surface Free Energy

The contact angle (θ) was based on the sessile drop method and performed on a Phoenix 300 (Surface Electro Optics, Suwon City, Korea) contact angle analyzer, equipped with microscopic lenses and a digital camera. The distilled water and di-iodo-methane were used as reference liquids for the wettability calculations. The angles were determined 60 s after the drops of liquid were applied onto the surface of the reference (the water). For each type of board (including the right-side A and left side B), ten droplets were measured.

The surface free energy was assayed according to the Owens-Wendt (1969) method. The method consists in determining the contact angles for two measuring liquids (water and di-iodo-methane), and the free surface energy (γ_s) is equal to the sum of dispersion (γ_{sd}) and polar (γ_{sp}) components (Wolkenhauer *et al.* 2009).

Resistance to Molds

The resistance to mold was evaluated using the test specimens of dimensions 50 by 50 by 16 mm^3. The test samples were superficially sterilized by spraying all surfaces with 70% ethanol. They were placed in sterile glass vessels for 24 h at 65 °C. After cooling the samples for the next 24 h, they were exposed to pure cultures of *Trichoderma virens* (strain BAM 34) fungus (growing on 2% MEA nutrient medium (OXOID)).

The study was carried out in petri dishes with a diameter of 200 mm and a height of 30 mm. Inoculation was performed on the surface of the nutrient medium using fungus by spraying the spore suspension. The samples were immediately placed in vessels. Four samples were placed in each vessel, namely one sample from each WPC variant (A was 40%, B was 50%, and C was 60% polymer content) and one control sample (D was 0% polymer content). The samples of materials were placed directly on the nutrient agar medium to ensure good moisture saturation.

The growth of fungus was conducted in incubator chambers for 22 days at 26°C. The degree of fungus overgrowing was determined periodically by taking high resolution laboratory photos, while using the cabinet station for documentation purposes. The degree of mycelium development on the samples was expressed as a percentage of the area covered in relation to the total upper surface of samples. The percentage value overgrowth of samples was determined with an accuracy of up to 5% with Image J2 - Fiji v1.52i image analysis software (Schindelin *et al.* 2012; Tinevez *et al.* 2017).

The resistance of the materials tested with the *T. virens* fungus was presented graphically by comparing the dynamics of mycelium growth on samples with different contents of the WPC polymer.

Statistical Analysis

Statistical analysis of the results was carried out using Statistica 13 (TIBCO Software Inc.) The analysis of variance (ANOVA) was used to test (α = 0.05) for significant differences between factors. A comparison of the means was performed using a Tukey test with α equal to 0.05.

RESULTS AND DISCUSSION

Mechanical and Physical Properties

The average density of the manufactured panels ranged between 699 to 723 kg per m^3. The density profiles of individual panel variants were characterized by a typical U-shaped course (Fig. 1). There were no significant differences between the course of density profiles for individual panel variants. Regardless of the panel variant, the differences between the densities of the face and core layers were in the range of 224 to 296 kg per m^3. Wong *et al.* (1998, 1999, 2003) and Treusch *et al.* (2004) reported that the density distribution is clearly correlated with the basic properties of particleboard, such as the MOR, MOE, and IB.

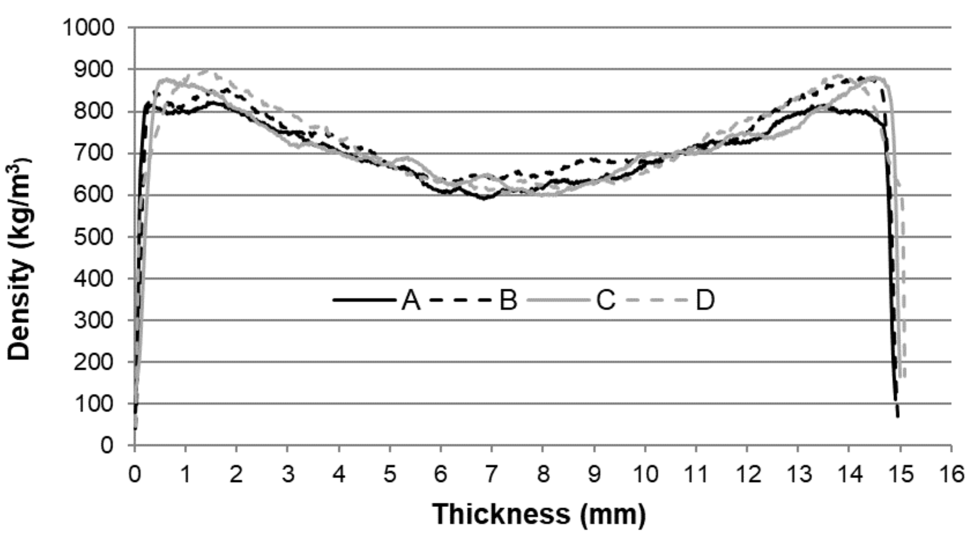

Fig. 1. Density profiles of tested panels

Wood-polymer composites manufactured with the contribution of a thermoplastic (variants A, B, and C) were characterized by a significant decrease in MOR and MOE values when compared to the control variant (option D) (Table 4). The decrease in panel strength of panels with thermoplastic was 25 to 28% in the case of MOR and 52 to 65% in MOE (Table 4). According to Falk's study (1999), WPC composites with 20% and 60% wood flour content have lower MOR and MOE values than traditional wood-based materials. At the same time, the authors indicated that WPC composites achieve comparable values of tensile and compressive strength as well as hardness in comparison to traditional wood materials. Sellers (2000) obtained similar strength properties to particleboards by examining flat-pressed boards of wood fibers and kenaf bonded with polyethylene or polystyrene (wood per polymer content was 50% per 50%) with densities of 600 to 900 kg per m^3.

Table 4. MOR, MOE, and IB values of the Tested Panels

Variant	MOR (N per mm^2)		MOE (N per mm^2)		IB (N per mm^2)	
	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.
D	13.55^a	0.95	2309^a	135	0.88^a	0.07
A	9.94^b	0.98	1110^b	113	0.81^a	0.07
B	10.17^b	0.90	1009^b	72	0.85^a	0.08
C	9.73^b	0.50	807^c	68	0.88^a	0.08

*Note: ^{abc} is the homogeneous groups by the Tukey test with α equal to 0.05, Avg. is the average value, and St. Dev. is the standard deviation.

Table 5. SH Values of the Tested Plates

Variant	SH ┴* (N per mm)		SH ǁ* (N per mm)	
	Avg.	St. Dev.	Avg.	St. Dev.
D	82.12^a	8.31	55.00^a	6.44
A	82.76^a	8.89	51.70^a	4.54
B	71.76^b	7.55	39.86^c	3.57
C	77.12^{ab}	6.80	44.33^b	4.91

*Note: ^{abc} is the homogeneous groups by the Tukey test with α equal to 0.05, ┴ means perpendicular to the surface, ǁ means parallel to the surface, Avg. is the average value, and St. Dev. is the standard deviation.

The tested wood-polymer composites were characterized by similar IB values (insignificant differences) in relation to the control particleboards (variant D). The decrease in MOR and MOE values was an outcome of polyethylene presence in the face layers. Polyethylene is a material with much lower elastic properties than wood (Seachtling and Woebcken 1995).

The increase in thermoplastic content in the face layers from 40% (variant A) to 60% (variant C) did not have a significant effect on the MOR properties (Table 5). However, a significant decrease by approximately 20% in MOE values was observed when the thermoplastic content in the face layers increased from 50% (variant B) to 60% (variant C). The obtained dependencies corresponded to the data presented in the literature. Stark and Berger (1997), Błędzki and Faruk (2004), Lee *et al.* (2004), or Cui *et al.* (2008) reported that as the content of wood particles in the composite decreases (regardless of their size), the MOR and MOE values decrease, whereas the tensile strength increases.

The produced wood-polymer composites (variant A, B, and C) when compared to the control particleboards (variant D) were characterized by a decrease in SH values both in the perpendicular and parallel tests (Table 5). However, it should be noted that in the case of SH in the perpendicular system, the registered differences were insignificant (Table 5). In the parallel system, the maximum decrease of 28% in the SH value was recorded for variant B. The literature report that SH in the WPC composites is comparable or higher than in solid wood or wood-based materials (Falk *et al.* 1999; Carroll *et al.* 2001; Kociszewski *et al.* 2007; Gozdecki and Kociszewski 2008). However, the literature data related mostly to extruded composites, which have a more even and uniform internal structure in comparison to the analyzed panels produced by pressing. It is also important to note that the obtained SH values for the tested wood-polymer composites were in the range of values provided among others for traditional particleboard (30 to 75 N per mm) (Niemz and Sonderegger 2017).

The produced particle-polymer boards compared to the control particle boards were characterized by much higher moisture resistance. The decrease in TS (soaking after 2 and 24 h) was in the range of 74% to 86%, while the decrease in absorptivity (after 2 and 24 h of soaking) was between 58% and 64%.

Table 6. Physical Properties of the Tested Panels

Variant	TS 2 h (%)		TS 24 h (%)		WA* 2 h (%)		WA* 24 h (%)	
	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.
D	25.39[a]	1.87	29.95[a]	2.29	76.29[a]	3.88	91.75[a]	3.73
A	5.88[b]	0.51	7.80[b]	0.49	29.63[b]	2.30	38.22[b]	2.51
B	5.10[b]	0.33	6.75[b]	0.48	29.31[b]	1.70	37.09[b]	1.95
C	3.43[c]	0.23	4.91[c]	0.47	27.26[b]	1.73	33.55[c]	2.07

*Note: [abc] is the homogeneous groups by the Tukey test with α equal to 0.05, Avg. is the average value, and St. Dev. is the standard deviation.

The high resistance of WPC composites to water in relation to other wood-based panels is also confirmed by the literature data (Falk *et al.* 1999; Sellers *et al.* 2000). Synthetic polymers in general have low water absorption (less than 1%) (Saechtling 2000) and act as a hydrophobic agent in WPC composites. They mechanically block the access of moisture to wood particles in the composite. Hydrophobic properties deteriorate with the increase in the contribution of wood particles in the composite (Zajchowski *et al.* 2005) and the increase in the porosity of its structure.

The increase in the thermoplastic content in the face layers from 40% (variant A) to 60% (variant B) reduced the penetration of moisture into the structure of the boards (significant decrease in swelling and water absorption) (Table 6). This is a favorable effect, if one assumes the use of the composite in an environment with a high relative humidity (*e.g.*, as elements of bathroom furniture).

Susceptibility to Drilling and Milling

The produced wood-polymer composites (variants A, B, and C) compared to the control particleboards (variant D) were easier to process. Both axial forces during drilling (Fig. 2) and the resultant cutting forces during milling (Fig. 3) were approximately 10 to 20% lower than the forces recorded during the processing of the control particleboards. A significant decrease in axial forces was noted for drilling (variants B and C). Better machinability of the particleboard with thermoplastic in terms of cutting resistance reported Wilkowski *et al.* (2013). Zbieć *et al.* (2010) examining the parameters of the WPC board cutting process (30% polyethylene) found that they are comparable to cutting parameters of particleboards with similar densities and glued with a UF resin. At the same time, the polyethylene that was part of the wood-polymer composite affected tool wear approximately 10 times slower compared to the traditional particleboard.

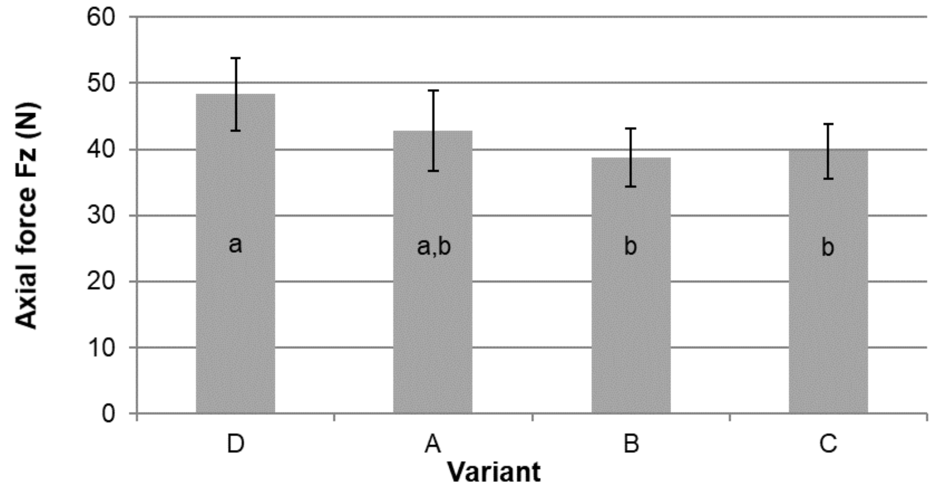

Fig. 2. Axial force values during plate drilling (ab is the homogeneous groups by the Tukey test)

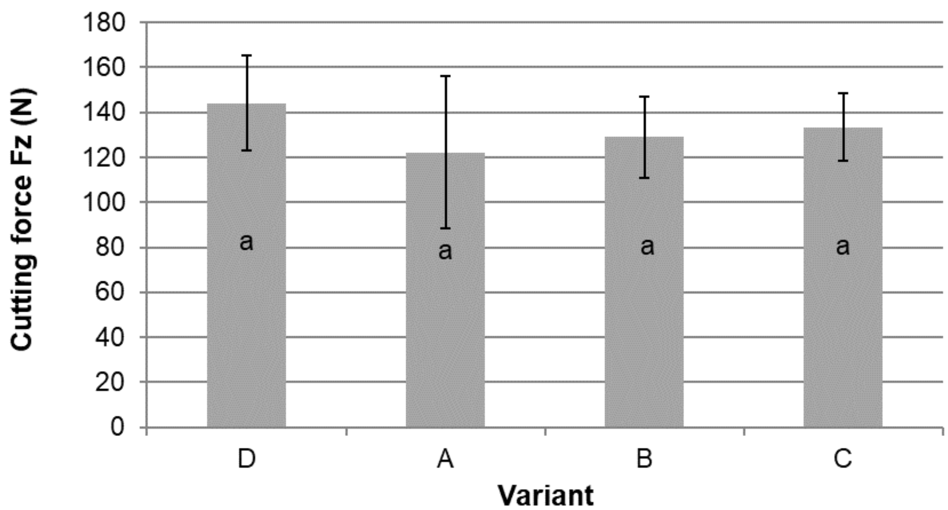

Fig. 3. Cutting force values (*Fw*) when milling boards (a represents the homogeneous groups by the Tukey test)

Buchlmann *et al.* (2001) reported that increased pigmentation of WPC composites may promote wear of cutting tools. In the present experiment, an increase in polyethylene content in face layers ranging from 40 to 60% did not significantly affect the values of the tested forces (Fig. 3).

Contact Angle and Surface Free Energy

The tested wood-polymer composites were characterized by reduced surface wettability (greater hydrophobicity). Regardless of the thermoplastic contribution in the face layers (variants A, B, C), the average contact angles of the surface with water were from 42% to 48% larger than the average contact angle of the control boards (option D) (Table 7). At the same time, wood-polymer composites presented 25% to 28% lower free surface energy (Table 7). Wettability is crucial with respect to the gluing process and surface finishing. The reduced wettability in variants A, B, and C impedes covering the surfaces with an aqueous solution (*e.g.*, glue or paint), and on the other hand, it decreases the penetration of moisture it the control board (variant D). According to literature data, contact angles below 90° indicate good wetting of the surface by the liquid (Baharoğlu *et al.* 2012). Buyuksari *et al.* (2010), Baharoğlu *et al.* (2012), and Sari *et al.* (2013) reported that contact angles for various particleboard variants are in the range of 83° to 116°. In turn, Ayrilmiss *et al.* (2012) found that the contact angles of the surface of flat WPC composites (depending on the size and content of wood particles, density of composites, and pressing temperature) were in the range of 70.9° to 102.4°. Jaunslavietis *et al.* (2018) indicated that the free surface energy of WPC composites made based on polypropylene with the participation of 50% wood particles is below 30 mN per m. In general, it can be stated that all tested wood-polymer composites (variants A, B, and C) revealed comparable or better surface wettability (smaller contact angle and higher free surface energy) in reference to data presented in the literature.

Table 7. Value of Contact Angle and Surface Free Energy

Variant	Wettability (contact angle*) (°)				Surface free energy** (mN / m)
	Water		Diiodomethane		
	Avg.	St. Dev.	Avg.	St. Dev.	
D	55,66^{a}	11,2	30,87^{a}	2,1	57,12
A	82,29^{b}	6,6	38,39^{ab}	3,5	43,04
B	79,96^{b}	6,4	43,39^{ab}	7,2	41,71
C	79,20^{b}	3,8	45,74^{b}	7,6	41,00

*Note: the contact angle was determined for 2 s, while in the subsequent seconds the drop completely dispersed, and the **surface energy was calculated by the Owens-Wendt method. ^{ab} is the homogeneous groups determined by the Tukey test with α equal to 0.05, Avg. is the average value, and St. Dev. is the standard deviation.

Resistance to Mold

The tested wood-polymer composites exhibited higher resistance to mildew (Fig. 4). The result of mold growth on the samples surface is illustrated in Fig. 5. Schirp *et al.* (2008) reported that WPC is susceptible to mold fungi. However, there is a lack of detailed information on this subject in the literature. Variant A of the wood-polymer composites (with a 40% thermoplastic content in the face layers) indicated a 25% lower tendency to surface fouling in comparison to the control samples (variant D). An increase in thermoplastic content in the face layers of up to 50% (variant B) increased the rate of growth of mold by 3.5-fold. In the case of wood-polymer composites (variant C), only 64% of the samples surface was covered after 22 days of exposure to the *Trichoderma virens* mold fungi (Fig. 4). Vidholdová *et al.* (2015) reported that traditional particleboard with uncovered surfaces are easily overgrown by mold fungi. WPCs with larger wood particles and their greater contribution are more susceptible to mold fungi (Schirp *et al.* 2008; Kartal *et al.* 2013; Feng *et al.* 2014). WPC susceptibility to molding is also dependent on the species of wood used as the filler (Feng *et al.* 2016). Klyosov (2007) reported that mold fungi reduce the aesthetics of WPC products by changing their color and decomposition. They also have a harmful effect on human and animal health (Jaakkola *et al.* 2013; Hernberg *et al.* 2014).

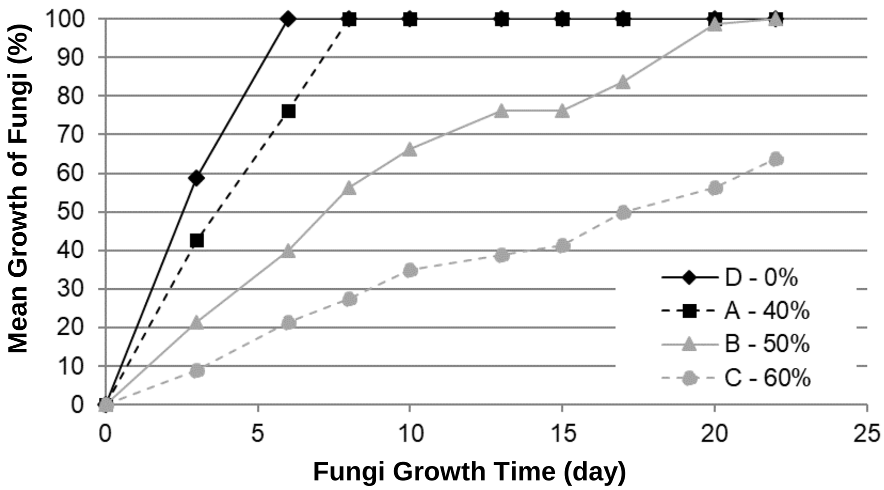

Fig. 4. *Trichoderma virens* mold fungal surface growth rate

Schirp *et al.* (2008) reported that the effect of staining fungi and mold fungi on WPC has been characterized only by the method of visual evaluation of microbial growth on the material. In this research, a computer analysis was carried out for the mold growth image on the samples surface.

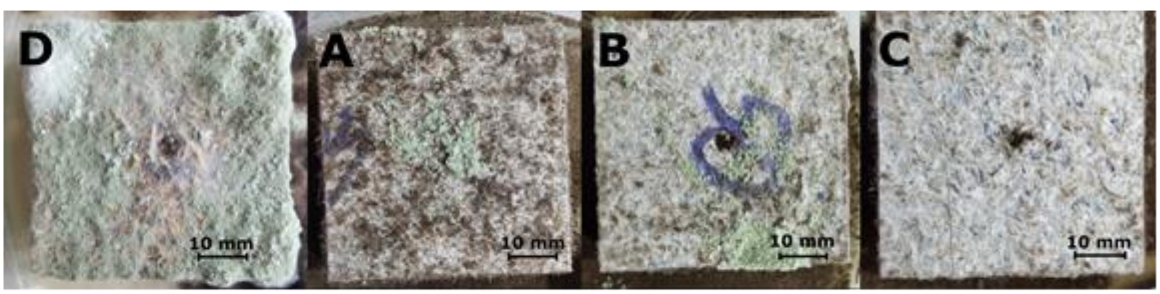

Fig. 5. Images of panel samples after 22 days of exposure to *Trichoderma virens* molds

CONCLUSIONS

1. Flat pressed wood-polymer composites combine selected properties of both traditional particleboard and WPC. In comparison to particleboard, wood-polymer composites present lower MOR and MOE values, but similar IB and SH values.
2. Wood-polymer composites are easier to machine (drilling and milling) than traditional particleboard.
3. Wood-polymer composites characterized high moisture resistance and the associated dimensional stability.
4. The composites indicated increased hydrophobicity of the surface, but simultaneously revealed a high wettability (contact angle below 90°) that should not unduly hinder the finishing process.
5. Although the wood-polymer composites presented high resistance to mold fungi, the

resistance increased as the thermoplastic content in the face layers increased.

6. The tested panels can be used in conditions of increased humidity, *e.g.*, as bathroom furniture elements.

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