

# Biological Characterization of Panels Manufactured from Recycled Particleboards using Different Adhesives

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Transforming waste or recycled materials into value-added products is of high priority today. Wood plastic composites (WPCs) show high potential for the use of recycled materials in making durable composites. The applicability of WPC panels produced from recycled materials (ultralight foam core particleboards) for exterior building application was tested using wood-destroying basidiomycetes. The results showed that the panels were fully resistant against *Coniophora puteana* (Cp) and *Gloeophyllum trabeum* (Gt), but not very resistant against *Pleurotus ostreatus* (Po). The decay susceptibility index of Po-exposed specimens showed that the polystyrene-bonded (PS) samples were more resistant than solid beech wood samples that were used as references, followed by melamine-urea formaldehyde-bonded samples. A comparison with the reference samples also showed that the panel density had a significant influence on the panel's resistance against basidiomycetes. The higher the panel density, the more resistance will be achieved in the panel.

*Keyword: WPCs; Recycled materials; Basidiomycetes; Outdoor application*

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## INTRODUCTION

Using production wastes is of economic and ecological importance in the particleboard industry, as it combines the chance to act in a highly competitive market with an almost perfected technology. Waste trimmings and production rejects from particleboard productions are usually burned for power generation or further processed and used again as core layer material (Irlé and Barbu 2010). A new technology to produce lightweight foam core particleboard has been developed recently, derived from the conventional production line of particleboard (Luedtke 2011). The produced ultralight panels (< 400 kg/m<sup>3</sup>) consist of resinated wood particles for the facing and expanded polystyrene foam in the core layer (Shalbafan *et al.* 2012). An additional innovative characteristic of this ultralight particleboard is that its continuous manufacturing occurs in a one-step process, which makes it possible to produce foam core panels using a conventional double-belt press. However, in contrast to common particleboard production, waste trimmings and production rejects from ultralight foam core particleboards cannot be directly recycled because the thermoplastic foam material would land in the surface layers and could complicate the process.

Wood plastic composites (WPCs) were introduced to the market in the 1990s. A WPC is a homogeneous material comprised primarily of wood particles, a thermoplastic polymer, and, depending on the intended use, various additives (Wolcott 2003; Segerholm

*et al.* 2012; Rahman *et al.* 2013). This technology shows a good potential for using waste or recycled wood and polymers to make durable composites, which in turn are potentially recyclable (Rowell *et al.* 1991). The use of waste as a raw material for WPC production has been investigated by a number of authors (Rowell *et al.* 1991; Chaharmahali *et al.* 2008; Poletto *et al.* 2011). The foam core layer of the recycled ultralight panels can be a possible material for WPC production, since it has a thermoplastic nature. Shalbafan *et al.* (2013a) have shown that producing durable and water-resistant WPC panels is possible using dry-blended residues of ultralight foam core particleboards.

The main application of WPC products is for outdoor use (*e.g.*, decking and fencing), and the products can thus be infected by wood-degrading fungi. Hence, resistance against destructive basidiomycetes is one of the most important characteristics for WPC products (Yap *et al.* 1990). The risk of fungal influx can be also changed when the wood content is varied (Krause and Gellerich 2014). Benthien *et al.* (2012) compared the fungal decay resistance of wood with WPC and found that WPC panels are more durable than wood samples. Curling and Murphy (2002) recommend the use of decay susceptibility index (DSI) to compare decay tests carried out in different laboratories, periods and conditions, or to compare samples of different sizes and densities. Defoirdt *et al.* (2010) mentioned that comparing the biological durability of WPC usage tests that were developed for wood or wood-based panels is only justified if their moisture behavior is similar. Several publications recommend a moistening method for WPC panels to achieve a minimum level of moisture content for fungal attack (Rowell 2007; Kim *et al.* 2008; Defoirdt *et al.* 2010).

In this study, flat-pressed WPC panels made of milled foam core particleboards residues were characterized regarding their resistance against biological degradation by investigating the samples' mass loss and decay susceptibility index (DSI) after fungal decay. Melamine urea formaldehyde (MUF)-bonded particle materials were used as reference material. The effects of wood flour (WF) content and binder type were studied. Before testing, samples were leached by immersion in water according to EN 84 (1997). It was the aim of this study to evaluate the durability of WPC material with respect to moisture resistant particle-based panel material.

## EXPERIMENTAL

### Panel Production

Two types of panels, both manufactured from recycled particleboards but differing in adhesive, either polystyrene (PS) or MUF, were compared in this study. The press temperature and specific pressure were set at 210 °C and 1 N/mm<sup>2</sup>, respectively. The hot pressing time was 400 s for reaching to the desired nominal thickness of 10 mm. At the end of the pressing cycle, cooling of the panels was performed under pressure (inside the press) by internal cooling of the press plates for the next 400 s to reach the ambient temperature. More details regarding the panel production process is explained by Shalbafan *et al.* (2013a).

Composite panels containing PS were produced in three steps, following previous studies (Shalbafan *et al.* 2012, 2013b). Step one was the production of foam core particleboards that consisted of softwood particles (spruce and pine) for the surface layers, which included urea formaldehyde resin (UF) and ammonium sulfate (AS) as additives, and expandable polystyrene beads (Sunpor GmbH, Austria) in the core layer. Second, the

foam core particleboards were crushed into a powder blend (mainly between 0.1 and 0.5 mm), and, third, the WPC panels were produced from the powder blend. In this study, the composite panels were produced without adding a coupling agent or further compounding with the extruder, as Shalbafan *et al.* (2013a) showed that such parameters had no positive or significant effect on the panel properties. PS played a role as an adhesive in the composite panels because of its thermoplastic nature. The contents of PS as adhesive in the final mixtures were 12% and 24% based on the oven dry mass of the recycled wood particles. Both types were named according to the adhesive amounts, as PS12 and PS24, respectively. Composite panels with a target density of 1000 kg/m<sup>3</sup> and a target thickness of 10 mm were produced from the crushed powder blend.

The second type of adhesive, used as a reference, was MUF. Composite panels containing MUF were also produced in three steps, which were as similar as possible to the steps of the composite panels containing PS to ensure the maximum comparability. Step one was the production of particleboards using softwood particles, UF and AS, according to Shalbafan *et al.* (2012) but without a PS foam core. Step two consisted of crushing of the particleboards to a powder blend. Step three, the production of composite panels from the powder blend, was carried out according to Shalbafan *et al.* (2013a), but with an additional spraying of MUF into the powder blend. The powder blends were mixed with 12% (MUF12) and 24% (MUF24) MUF resin based on the oven dry mass of the wood particles. The adhesive was sprayed onto the particle furnish tumbling in a rotating drum-type blender using a compressed air spray head. The MUF resin called Prefere 10H116 was supplied from Dynea Austria GmbH (Krems, Austria). The panel density and thickness were kept constant at 750 kg/m<sup>3</sup> and 10 mm, respectively. A higher panel density than 750 kg/m<sup>3</sup> was hard to achieve as the particle mat moisture content reached levels that were not manageable in the case of high resin content samples. Three panels were produced from each panel variation. Three samples of each replicate (resulting in n = 9) were randomly selected and tested. Table 1 shows the composition of the produced WPC panels.

**Table 1.** Composition of the WPC Panels

Sample	Wood content (%)	Adhesive content (%)	Adhesive type*	Panel density (kg/m <sup>3</sup> )	Repetitions
PS12	76	24	PS	1000	3
PS24	88	12	PS	1000	3
MUF12	76	24	MUF	750	3
MUF24	88	12	MUF	750	3

\*PS: Polystyrene; MUF: Melamine urea formaldehyde

### Test Specimen Preparation

Testing methods for the characterization of WPC panels are described in the EN 15534-1 (2012). Composite panels were cut into test specimens with dimensions of 250 x 50 x 10 mm<sup>3</sup> for the flexural tests and 50 x 50 x 10 mm<sup>3</sup> for all other tests. Surface layers of test specimens were not further processed. Prior to testing, specimens were conditioned at 65% relative humidity and 20 °C for two weeks to reach equilibrium moisture content (EMC).

*Biological properties*

The resistance against wood-destroying basidiomycetes in the form of loss of mass and the decay susceptibility index (DSI) was determined by testing according to ENV 12038 (2002) with modifications according to EN 15534-1 (2012) and an aging procedure according to EN 84 (1997). The DSI is calculated using following equation,

$$DSI(\%) = \frac{T}{S} \times 100 \quad (1)$$

where  $T$  is loss in mass of an individual test specimen (%) and  $S$  is loss in mass of the appropriate set of size control specimens (%). Loss in mass was calculated as the percentage of the entire initial test specimen mass minus the mass of its adhesive (12 or 24 wt%). The aging procedure of pre-conditioning, which was performed to ensure optimal growth conditions for *Coniophora puteana* (Cp) and *Gloeophyllum trabeum* (Gt), was unnecessary for *Pleurotus ostreatus* (Po) because test specimens had to be embedded into water-impregnated vermiculite anyway.

A total of 30 test specimens per composite panel type (PS24, PS12, MUF24, and MUF12) were spread over five test groups (Cp, Gt, Po, moisture content check, and wetting check), each with six test specimens. Twenty-four control specimens consisting of Scots pine sapwood (*Pinus sylvestris*) were spread over four test groups (virulence control and size control of the test fungi Cp and Gt, which causes a decay of brown rot type), each with six test specimens. Twelve control specimens consisting of beech (*Fagus sylvatica*) were spread over two test groups (virulence control and size control of the test fungus Po, which causes a white rot type of decay), each with six test specimens.

*Physico-mechanical properties*

Flexural strength and flexural modulus of elasticity (three point bending test) were determined according to EN 310 (1993) using a 200-mm support span with a crosshead speed of 5 mm/min. Internal bond strength (tensile strength perpendicular) of the test pieces was also determined according to EN 319 (1993). Square test pieces of 50\*50 mm<sup>2</sup> for IB were prepared from each of the panel replicates ( $n = 15$ ). Density was determined by testing 45 test specimens per composite panel type, which were later used for testing resistance against fungal attack, according to EN 323 (1993). Five samples of each replicate (resulting in  $n = 15$ ) were randomly selected and mechanically tested.

**Table 2.** Test Specifications

Test	Standard type	Sample size (mm)	Repetitions
Density	EN 323	50 x 50 x 10	45
<b>Biological properties</b>	ENV 12038	50 x 50 x 10	30
Thickness swelling	EN 317	50 x 50 x 10	9
Internal bond strength	EN 319	50 x 50 x 10	15
Bending properties	EN 310	250 x 50 x 10	15

Water absorption and thickness swelling were determined after 24, 48, and 672 h of water immersion by testing nine test specimens per composite panel type according to EN 317 (1993). Water absorption is shown as the percentage of the entire initial test

specimen mass and not as the percentage of only the initial mass of the wood part. Detailed information for testing specifications is shown in Table 2.

### Statistical Analysis

To evaluate the significance of differences among mean values of PS24, PS12, MUF24, and MUF12, a single-factor analysis of variance (ANOVA) and a Tukey honest significant difference (HSD) test were conducted using the analysis tool of SAS JMP (SAS Campus Drive, Cary, USA). The null hypothesis, no effect, was accepted if the p-value exceeded the  $\alpha = 0.05$  significance level.

## RESULTS AND DISCUSSION

### Biological Properties

According to ENV 12038 (2002), the loss in mass of the virulence control and size control specimens are the basis for any further appraisal of test specimen results regarding the resistance against wood-destroying basidiomycetes. An overview of the results for the control specimens is presented in Table 3. Mean losses in mass of virulence control and size control specimens within each test fungus group were always similar and therefore indicated a low influence of the difference in form between virulence control (50 x 25 x 15 mm<sup>3</sup>) and size control specimens (50 x 50 x 10 mm<sup>3</sup>). For virulence control specimens, according to ENV 12038 (2002), a minimum loss in mass of 20% is required after 16 weeks of fungal attack exposure to accept the fungi test results as valid. This minimum level was exceeded by Cp (25.77%) and Gt (31.07%) but not by Po (14.94%). Failed virulence control limits of Po were also reported by Brischke *et al.* (2014) as a result of strain weakness. However, in this study, even the Po results were considered further, taking into account that this test fungus led only to a relatively low mass loss. Mean mass losses of size control specimens, also including Po results, were used to calculate the DSI.

**Table 3.** Biological Properties of Virulence Control and Size Control Specimens

	Scots pine sapwood				Beech			
	Virulence control		Size control		Virulence control		Size control	
	MV*	SD*	MV	SD	MV	SD	MV	SD
Loss in mass (%)								
<b><i>Coniophora puteana</i></b>	25.77	5.43	23.77	4.85	-	-	-	-
<b><i>Gloeophyllum trabeum</i></b>	31.07	4.32	32.08	1.34	-	-	-	-
<b><i>Pleurotus ostreatus</i></b>	-	-	-	-	14.94	4.00	15.00	5.29

\*MV: mean value; SD: standard deviation

An overview of the biological properties of the test specimens is given in Table 4. Regarding the mass loss results, fungi causing brown rot decay, Cp and Gt, led to a mass loss clearly under 3%. The fungus causing white rot decay, Po, led to a mass loss of more than 3% in all test specimens. The high mass loss caused by Po was unexpected because of the failed virulence control limit and the low share of hardwood in the composite panels. Schmidt (2006) showed that white-rot fungi such as Po prefer hardwoods, whereas brown-rot fungi like Cp and Gt grow better on softwoods. Köse *et al.* (2011) earlier also reported findings similar to those reported here (Po fungi have a preference to grow on softwood).

Köse *et al.* (2011) showed that in case of Po, the mass loss of particleboard and fiberboard is higher than that of Gt. The results may be due to the processing conditions of wood and wood-based panels. Wood structures and their compositions can be changed during the production process of wood-based panels; hence, different biological characteristics were achieved. Deng *et al.* (2006) also stated that the process conditions can significantly contribute to the mold resistance of fiberboard.

The lowest mass loss values (%) for Po were obtained for the PS24 (3.55%) followed by the PS12 (4.02%). The panel types MUF24 and MUF12 had a mass loss of 21.01% and 16.9%, respectively. For Po, there were statistically significant differences in mass losses between PS and MUF composites. The higher share of adhesive in the MUF24 specimens may have led to their higher mass loss caused by Po then for the MUF12 specimens. This was unexpected because a higher share of adhesives (at MUF24) should result in increased moisture resistance of the specimens and, accordingly, lower fungal-induced mass losses (because of the protective effect of water absorption). However, because of the high standard deviation of the mass losses of MUF specimens (Po), such results were not significant and it could not be concluded that testing characteristics caused the results.

**Table 4.** Biological Properties of Test Specimens

	PS12			PS24			MUF12			MUF24		
	MV*	SD*	HG*	MV	SD	HG	MV	SD	HG	MV	SD	HG
<b>Loss in mass (%)</b>												
<i>Coniophora puteana</i>	1.16	0.53	a	1.37	0.21	a	0.79	0.42	a	0.88	0.20	a
<i>Gloeophyllum trabeum</i>	0.43	0.12	b	0.70	0.11	a	0.64	0.10	a	0.51	0.14	a,b
<i>Pleurotus ostreatus</i>	4.02	0.50	b	3.55	0.24	b	16.9	7.96	a	21.0	10.0	a
<b>Decay Susceptibility Index (-)</b>												
<i>Coniophora puteana</i>	Fully resistant			Fully resistant			Fully resistant			Fully resistant		
<i>Gloeophyllum trabeum</i>	Fully resistant			Fully resistant			Fully resistant			Fully resistant		
<i>Pleurotus ostreatus</i>	26.8	3.34	a	23.7	1.57	a	113	53.1	b	140	66.7	b

\*MV: mean value; SD: standard deviation; HG: homogeneous group (Within a row, groups with the same letter are not statistically different;  $\alpha = 0.05$ )

In accordance with ENV 12038 (2002), the DSI was only calculated if the mean mass losses were greater than 3%, and the rest were designated as fully resistant to attack by wood-rotting basidiomycetes. Thus, only DSIs for Po specimens were calculated to allow a comparison between test specimens (composites) and size control specimens (beech). The PS composites with a DSI below 100 (PS24 at 23.69 and PS12 at 26.78) were more resistant than beech, whereas the MUF specimens with a DSI above 100 (MUF24 at 140.11 and MUF12 at 112.67) were less resistant than beech (Table 4). Higher resistance of the PS specimens compared to those of the MUF and beech specimens may have resulted from the inherent nature of the polymer and possible chemical action taking place in the recycled PS samples.

## Physico-Mechanical Properties

An overview of the achieved mechanical properties is given in Table 5. Many of the PS samples failed during the perpendicular tensile strength tests (internal bond strength) because of the failure at the adhesive joint between the test specimen and the test blocks. The weakest point in the arrangement of internal bond strength tests was the adhesive joint in 14 out of 15 PS24 samples and 4 out of 15 PS12 samples (PS24: 1.50 N/mm<sup>2</sup> and PS12: 1.50 N/mm<sup>2</sup>). This showed that the results obtained for the PS samples were approximately the perpendicular tensile strength of the adhesive joint.

The fact that 14 PS24 and only 4 PS12 specimens were too strong for IB measurements permit the assumption that the true tensile strength perpendicular to PS24 was higher than those of PS12, without indicating the precise amount. However, this problem in estimating the perpendicular tensile strength of strong composites also arose when using another type of adhesive, polyurethane adhesive, for fixing specimens on the plywood test blocks. Taking the adhesive problems into account, it could be generalized that PS specimens (1.50 N/mm<sup>2</sup>) had a higher perpendicular tensile strength than MUF specimens (MUF24 at 1.44 N/mm<sup>2</sup> and MUF12 at 1.05 N/mm<sup>2</sup>). Moreover, it was likely that for both materials—PS composites and MUF composites—a higher adhesive share (PS or MUF) led to a higher perpendicular tensile strength.

A ranking similar to that for internal bond strength was observed for the flexural properties; PS24 always had the highest mean values, followed by PS12, MUF24, and MUF12. Significant differences were observed for both the MOE and the MOR between the corresponding specimens of PS and MUF samples. Accordingly, higher adhesive share (PS or MUF) led to increased flexural properties (MOE and MOR). The higher flexural properties of the PS samples occurred because of the nearly 30% higher panel density compared with those of MUF panels. Flexural properties in wood and wood-based composites are a function of its density (Kretschmann 2010). On the other hand, a reduction of polymer content (from 24% to 12%) results in a reduced binding between the wood flours and, adversely, leads to the lowering of flexural properties (Sanadi *et al.* 2001). In WPC panels with high WF content (> 50%), polymer acts as an adhesive to bond wood flour together. Reduction of the polymer amount leads to the weak bonding of wood flour and consequently reduces bending properties (Shalbafan *et al.* 2013a).

**Table 5.** Mechanical Properties of Test Specimens

	PS12			PS24			MUF12			MUF24		
	MV	SD	HG	MV	SD	HG	MV	SD	HG	MV	SD	HG
<b>Internal bond</b>	1.50	0.2	a	1.50	-	a:b	1.05	0.26	b	1.44	0.35	a
<b>Flexural test</b>												
<b>MOE</b>	5120	720	a	5690	550	a	1660	390	b	1900	630	b
<b>MOR</b>	29.3	4.7	b	40.1	5.7	a	11.0	3.3	c	14.1	4.9	c

MV: mean value; SD: standard deviation; HG: homogeneous group (Within a row, groups with the same letter are not statistically different;  $\alpha = 0.05$ )

An overview of the achieved physical properties is given in Table 6. Significant differences between the densities of PS and MUF specimens were generally expected because of their different target densities. However, in contrast to the density of MUF specimens (MUF24: 751 kg/m<sup>3</sup> and MUF12: 732 kg/m<sup>3</sup>), which differed in a small range around their target density (750 kg/m<sup>3</sup>), the density of PS specimens (PS24: 1,114 kg/m<sup>3</sup> and PS12: 1,072 kg/m<sup>3</sup>) significantly differed from one another and were clearly above the

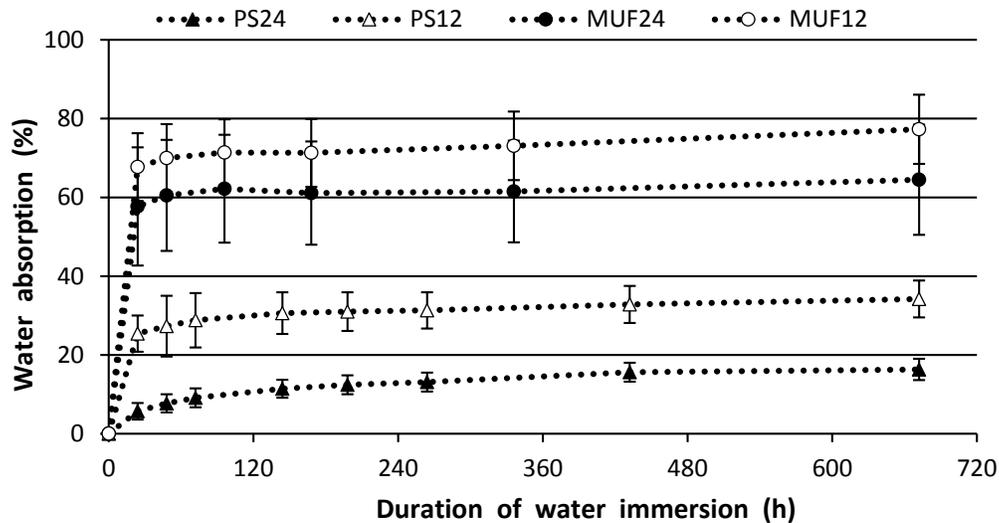
target density of PS specimens (1,000 kg/m<sup>3</sup>). The density was higher for samples with higher adhesive content (PS24 and MUF24). This can be explained by the higher compressibility of the samples with higher amounts of adhesives (PS and MUF). However, these density differences were very likely a result of manual spreading.

**Table 6.** Physical Properties of Test Specimens

	PS12			PS24			MUF12			MUF24		
	MV	SD	HG	MV	SD	HG	MV	SD	HG	MV	SD	HG
<b>Density (kg/m<sup>3</sup>)</b>	1114	61	a	1072	78	b	732	59	c	751	77	c
<b>Water absorption (%)</b>												
24 h immersion	25.4	4.6	b	5.7	2.1	c	67.7	8.6	a	57.7	15	a
48 h immersion	27.3	7.7	b	7.7	2.3	c	70.0	8.6	a	60.5	14	a
672 h immersion	34.2	4.7	c	16.3	2.7	d	77.3	8.8	a	64.5	14	b
<b>Thickness swelling (%)</b>												
24 h immersion	12.5	2.3	a	2.2	0.5	d	9.1	0.5	b	6.2	0.7	c
48 h immersion	14.9	3.0	a	3.2	1.1	d	9.4	0.6	b	6.6	0.7	c
672 h immersion	20.1	2.5	a	7.1	0.8	c	10.5	0.9	b	7.4	0.7	c

MV: mean value; SD: standard deviation; HG: homogeneous group (Within a row, groups with the same letter are not statistically different; α = 0.05)

Rankings of water absorption were unchanged during the whole period (672 h) of water immersion; PS24 always had the lowest mean values, followed by PS12, MUF24, and MUF12 (Fig. 1). Thus, the PS specimens had lower water absorptions than the MUF specimens, and specimens with a high share of adhesive had lower values than those with a low adhesive share. At the end of the treatment, after 672 h of immersion in water, statistically significant differences existed between all tested materials.



**Fig. 1.** Water absorption of test specimens (mean values and standard deviation)

Water absorption (WA) was influenced by the panel density. Higher panel density was correlated with lower WA. It is difficult for water molecules to permeate samples with higher density. The same was true when the amount of adhesive share was increased (Yap

*et al.* 1990; Shalbfan *et al.* 2013a). A good correlation ( $R^2=83\%$ ) was found between the amount of water absorption and mass loss for the white rot decay (Po), implying that the penetration of fungus mycelium into the specimens followed a similar pattern as the penetration of water molecules (Bari *et al.* 2014). Such correlation was not observed for the Cp and Gt fungi.

Thickness swelling (TS) values after long-term immersion (672 h) are presented in Fig. 2. Another ranking was observed for TS compared to WA; PS24 had the lowest mean values, followed by MUF24, MUF12, and PS12. It was striking that the MUF specimens, which had higher water absorptions than PS specimens, showed thickness swellings in between and not higher than those of PS specimens. This could have been caused by the lower density of MUF24 and MUF12, which are associated with a higher amount of hollow spaces in the composite and could partially allow water absorption without swelling. It was also concluded that the MUF adhesive had a better covering and binding effect on wood flour than did the PS adhesive. This was attributed to the decreased swelling of wood parts in the MUF specimens. It was concluded that a minimum amount (24%) of adhesive for recycled WPC panels was necessary to achieve comparable physical properties. However, a higher share of adhesive also led to lower thickness swelling.

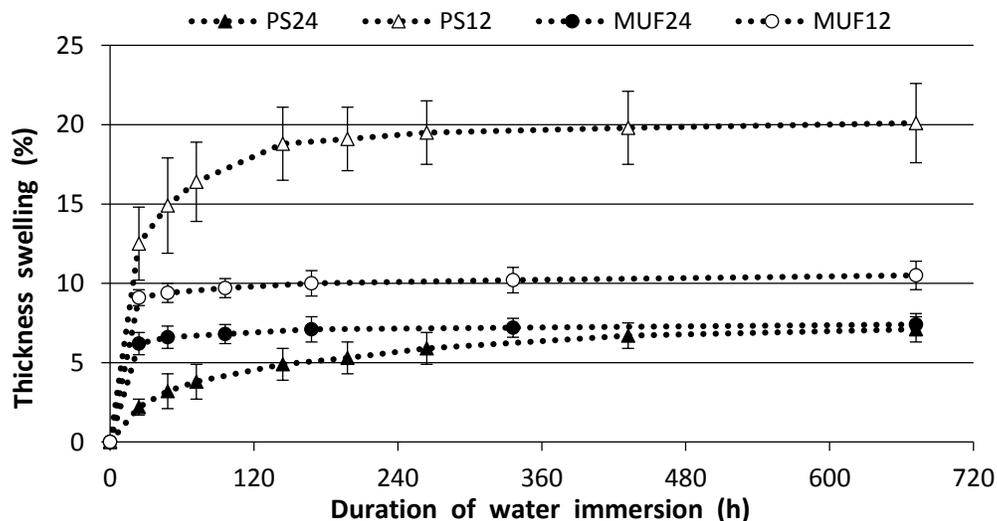


Fig. 2. Thickness swelling of test specimens (mean values and standard deviations)

## CONCLUSIONS

1. The study showed that utilization of recycled and waste materials in the value-added and a durable WPC panel for outdoor applications was possible.
2. WPC and reference panels were fully resistant to attack by wood-rotting of Cp and Gt, while the panels were not resistant against Po.
3. The decay susceptibility index (DSI) of Po specimens showed that the PS samples were more resistant than beech wood, followed by MUF samples. Such trends can be explained by the higher density of PS composites.
4. The mechanical properties (MOE, MOR, and IB) were raised at higher panel density and the performance ranking with this regard was PS24 > PS12 > MUF24 > MUF12.

5. Physical properties (WA and TS) showed that the amount of adhesive content should be at least 24% to achieve a durable panel for outdoor application.
6. As a final conclusion, it can be said that the recycled WPC panels were produced without a coupling agent and even extra compounding process (as they are too costly), which has good potential to be used for exterior application.

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