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The influence of accelerated weathering on the mechanical and physical properties of wood-plastic composites

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Abstract. This research concentrates on the effect of moisture absorption and UV radiation on the mechanical and physical properties of wood-plastic composites (WPC). The goal is also to evaluate the importance of wood flour fraction size on the mechanical properties of WPC and their influence on the accelerated weathering results. Wood flour reinforced composites with three different fractions of wood flour made from birch (*Betula*) chips were prepared. Additionally, Bleached-Chemi-Thermo-Mechanical aspen (*Populus tremula*) pulp (Aspen BCTMP) was used. Thermoplastics (LLDPE-g-MAH, PP) were used to prepare composites. Wood flour and BCTMP surface were treated with two different coupling agents: 3-aminopropyltriethoxysilane (APTES) and polyvinyalcohol (PVA). The WPC specimens were prepared by injection molding. Accelerated weathering tests were carried out to evaluate the influence of weathering on the mechanical and physical properties of composites. Three-point bending test and Charpy impact test were used to test mechanical properties. The test results showed that using wood flour as a filler material in composites made the WPC material more rigid and brittle. Due to the water absorption and swelling of WPC, the flexural modulus (MOE) and strength decreased and impact strength increased by making the material weaker with increasing the deflection of the WPC. The UV radiation decreased the composites flexural strength and MOE, while impact strength was increased. After the accelerated weathering cycles, cracks and voids were found on the surface of the WPC materials. After the UV radiation treatment, also the WPC colour was lightened.

Key words: wood-plastic composites, wood flour, coupling agent, mechanical properties, physical properties, weathering.

1. INTRODUCTION

Wood-plastic composites (WPC), based on wood flour and polyolefins, have been of interest scientifically as well as commercially, because WPC combines the best properties of both polyolefins and wood particles. For example, compared to wood, WPC has higher moisture resistance, rot resistance, split resistance, distortion resistance, UV resistance and does not need to be painted. WPC also has better durability (8–15 years) compared to wood (3 years) [1]. WPC properties depend on the features of their constituents like the wood particle size and binding the wood particles to the polymer matrix using coupling agents. WPC material properties are also influenced by different polymer types, wood filler content, additives and processing technologies.

WPC mechanical properties are influenced by weathering conditions (moisture and UV radiation effect) and therefore using this material outdoors makes it sensitive to changes in the environment. The natural weathering or accelerated weathering of WPC materials can change their colour and mechanical properties [2]. WPC two major components, wood and polymer, are both vulnerable to weathering and, therefore, it is necessary to evaluate water absorption and UV radiation effect on WPCs.

WPC consists of hydrophobic polymer matrix and hydrophilic wood, in form of flour, fibers or particles [3]. Due to the wood hydrophilic nature, moisture absorption of WPCs will mainly occur in the wood component [3,4]. Therefore, it is very important to have

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strong bonding between wood fibers and thermoplastic polymers in order to avoid unwanted effects on the WPCs dimensional stability and mechanical properties [4]. For that reason, different surface treatments are used before improving wood/polymer adhesion in WPCs [5]. Maleic anhydride is typically used as a coupling agent in composites and different silane coupling agents are used for wood fibre surface treatments [6]. Also, polyvinyl alcohol (PVA) has been used for wood fibre surface treatment due to the strong bonding with wood [7,8]. This improves the adhesion and wetting between the polymer matrix and wood filler in the composites. On the surface of the WPC, wood particles are covered with a thin layer of the polymer matrix by making the WPC relatively water-resistant material. However, UV radiation can cause degradation of this thin polymer surface layer resulting in a decreased water resistance [2,4]. Due to the degradation of this thin surface layer, water absorption into the composites can occur through gaps and flaws at the interfaces between wood fibres and thermoplastic matrix resulting in the reduction of the mechanical properties of WPCs [4].

WPCs are also very susceptible to UV radiation degradation. Photodegradation affects all the main components of wood such as cellulose, hemicellulose, lignin, and extractives. Lignin is found to be more sensitive to degradation than other wood constituents (lignin absorbs from 80% to 95% of UV light). Photodegradation of lignin results in the discoloration of wood. Photochemical reactions in wood are initiated by the UV radiation causing the formation of free radicals, which causes the degradation of lignin and photo-oxidation of cellulose and hemicelluloses, leading to discoloration of wood [9]. In WPCs, UV radiation also causes photodegradation of polyolefins through the radical-based process. Due to the photodegradation, polymer dimensions change in all scales (monomer unit, chain, morphology, and on the macroscopic scale). Therefore cracks can occur and propagate on the composite surface which result in light diffusion and decrease of mechanical properties [9,10].

The aim of this study was to investigate the influence of wood particle size, UV radiation, and moisture absorption to the WPC structure, mechanical and physical properties. Therefore WPCs were made using three different wood particle fraction sizes. Wood particle surface was treated with different coupling agents (silane or polyvinyl alcohol). WPC test specimens were made by injection molding. The behaviour of these materials was studied after accelerated weathering: bending and impact tests were made to evaluate mechanical properties of the composites, and surface changes were observed by SEM and colour measurement.

2. MATERIALS AND METHODS

2.1. Polymers and wood fillers

Two different thermoplastics were used: pelletized polypropylene (PP) from the Borealis Polymers OY company and Fusabond E (MB226DE) from the DuPont enterprise. PP was heterophasic copolymer (block copolymer) and Fusabond E was maleic-anhydride-grafted linear low-density polyethylene (LLDPE-g-MAH). Physical properties of the polymers used are given in Table 1.

Two different types of wood raw material were used as a reinforcing filler in the composites. Birch (*Betula*) chips were brought from the UPM Kymmene Otepää AS firm and then refined to appropriate fractions using disintegrator device DS-A. After the refining process, three different fraction sizes of birch wood particles were obtained (Table 2). Additionally Bleached-Chemi-Thermo-Mechanical aspen (*Populus tremula*) pulp (Aspen BCTMP) from company AS Estonian Cell was used. Quality of Aspen BCTMP was HB 450/80. As Aspen BCTMP was in the form of large chips, it was ground to the fine flour (particle size ≤0.126 mm) using cutting mill Retsch SM100.

2.2. Coupling agents

Three different coupling agents were used to improve adhesion between the polymer matrix and the filler. LLDPE-g-MAH was added to the PP composites, 5% of the wood fibre content (Table 3). Wood flour was treated with two different coupling agents: 3-aminopropyltriethoxysilane (APTES) and PVA. Both coupling agents bond to the wood through OH-groups creating a hydrogen bond. APTES was mixed (5% of the wood fibre content) with solution of ethanol and destilled water (ratio of the substances was 9 : 1) and sprayed over wood flour. For PVA, water solution was made, 5% PVA was added to the wood fibre content and solution was sprayed on wood flour.

Table 1. Physical properties of polymers

Properties	Fusabond E (MB226DE)	PP (BC245MO)
Density, g/cm^3	0.93	0.905
Melt flow rate, g/10 min	1.75	3.5
Metling point, °C	120	166
Conditions of use, °C/2.16 kg	190	230

Table 2. Measurements of birch wood particles

Fraction	Size, mm
I	≤0.63
II	0.63–1.25
III	1.25–2

Table 3. Composition of WPC samples

Sample	Polymer	Coupling agent	Polymer coupling agent, wt%	WF fraction, mm	WF coupling agent	WF coupling agent, wt%	Polymer/ wood, %
PP1	PP	LLDPE-g-MAH	5				100
PP2	PP	LLDPE-g-MAH	5	BCTMP			80/20
PP3	PP	LLDPE-g-MAH	5	BCTMP	APTES	5	80/20
PP4	PP	LLDPE-g-MAH	5	BCTMP	PVA	5	80/20
PP5	PP	LLDPE-g-MAH	5	I	APTES	5	65/35
PP6	PP	LLDPE-g-MAH	5	II			65/35
PP7	PP	LLDPE-g-MAH	5	II	APTES	5	65/35
PP8	PP	LLDPE-g-MAH	5	II	PVA	5	65/35
PP9	PP	LLDPE-g-MAH	5	III	APTES	5	65/35
LLDPE1	LLDPE-g-MAH						100
LLDPE2	LLDPE-g-MAH			BCTMP			80/20
LLDPE3	LLDPE-g-MAH			BCTMP	APTES	5	80/20
LLDPE4	LLDPE-g-MAH			BCTMP	PVA	5	80/20
LLDPE5	LLDPE-g-MAH			I	APTES	5	65/35
LLDPE6	LLDPE-g-MAH			II			65/35
LLDPE7	LLDPE-g-MAH			II	APTES	5	65/35
LLDPE8	LLDPE-g-MAH			II	PVA	5	65/35
LLDPE9	LLDPE-g-MAH			III	APTES	5	65/35

2.3. Composite processing

After mixing the wood particles with coupling agents, wood flour was dried at 110 °C in an air-circulating oven for 2 h and then mixed thoroughly with polymer matrix in mortar. All composites made of wood flour as a filler were prepared with 65% polymer and 35% wood flour (WF). Aspen BCTMP composites were prepared with 80% polymer and 20% wood flour. Also 3% (by weight of the mixture) calcium stearate and silicone oil were added to the mixture in order to improve the fluidity of the mixture in the extruder. Mixture of composites is shown in Table 3.

The mixture was then compounded in twin-screw extruder Brabender Plasti-Conder PLE 651 at 175 °C for LLDPE-g-MAH and 185 °C for PP to produce homogeneous composite pellets. The screw speed was 30–40 rpm. Test samples were prepared by injection molding of the WPC pellets according to ISO 178:2010. Battenfeld BA 230 E device was used for injection molding. Test samples dimensions were 60 mm × 10 mm × 4 mm, cross-section area was 40 mm^2.

2.4. Methods of testing

2.4.1. Weathering

Water absorption, swelling, and resistance to the UV radiation of the composites were tested. For testing the resistance to UV radiation, at least five test samples were selected from each experimental group. Composite resistance to the UV radiation was tested according to the EN ISO 4892-3:2006. The composite samples were placed in the UV radiation chamber that was equipped with two UVC radiation lamps: Philips 30 W, G30T8 UV-C and 15 W, G15T8 UV-C. Wavelength of UV lamps was in the range of 250 nm. Test was carried out at room temperature, 23 °C. Composite samples were placed in the UV radiation chamber and the exposure cycle was carried out for 3 weeks (500 h). After the exposure cycle ended, the samples were removed for colour measurements and mechanical testing.

For water absorption and swelling experiments a minimum of five samples were tested for each composite. Tests were carried out according to EN ISO 322 and EN ISO 317. First, samples were weighed and measurements were taken and then immersed in distilled water at room temperature for 3 weeks. Samples were weighed and measured each week. Before weighing and measuring, the samples were wiped with paper tissue to eliminate the excess of water on the sample surfaces. The change in mass of the samples was measured to an accuracy of 0.01 g and their dimensions to an accuracy of 0.01 mm. After the soaking cycle ended, the samples were removed for colour measurements and mechanical testing.

2.4.2. Mechanical testing

Flexural tests were carried out with three-point loading system according to ISO 178:2010. Flexural strength was measured at room temperature with crosshead speed of 20 mm/min, test span 60 mm. For testing, Instron 5866 tensile tester was used. Five test samples were used for every composite. Before testing, width and thickness of each test sample were measured. The

flexural strength and modulus of elasticity (MOE) were calculated during the bending testing.

Also Charpy impact strength for single-notched samples were tested in edgewise position according to ISO 179-1. Test was carried out with a Zwick 5102 pendulum impact tester at room temperature. The nominal pendulum energy was 4 J. Thickness and remaining width in the centre from the notch, to the nearest 0.02 mm was measured before Charpy impact test was performed. Energy, absorbed by breaking the test sample, was measured and Charpy impact strength was calculated.

2.4.3. Colour analysis

Surface colour was measured to evaluate the colour changes of the treated and untreated composites. Colour was measured with Minolta Chroma Meter CR-121, according to EN ISO 4582. Composites surface colour was measured by using the CIELAB colour system. In CIELAB system, colour is measured in L^*, a^*, b^* coordinates, where L^* is the lightness coordinate in the range between 0 (black) and 100 (white), a^* is red/green coordinate ($+a^*$ to red and $-a^*$ to green), b^* is yellow/blue coordinate ($+b^*$ to yellow and $-b^*$ to blue). The colour difference is expressed as a single numerical value, ΔE^*, which indicates the size of the colour difference. ΔE^* is defined as

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2}, \quad (1)$$

where ΔL, Δa, and Δb are the differences between the initial and after weathering values of L^*, a^*, and b^*, respectively.

3. RESULTS AND DISCUSSION

3.1. Flexural properties

The changes in flexural strength of different wood flour fraction size (modified with APTES) and polymers (LLDPE-g-MAH and PP) are described in Fig. 1. The results show that addition of wood flour to the LLDPE-g-MAH increased flexural strength twice. However, addition of wood flour to the PP reduces the flexural strength, because PP is a more rigid polymer than LLDPE-g-MAH and therefore wood flour makes PP more flexible. Test results show that biggest flexural strength results were obtained with wood flour fraction I. Increasing the wood flour fraction size decreased the flexural strength of WPCs slightly up to 8%. A similar trend has also been shown in other studies [11,12]. Decreasing flexural strength with increasing the wood flour fraction size can be explained with the fact that composites with smaller wood particles were more homogeneous than composites with larger wood fibres. WPC mechanical properties are more influenced by the fibre length to diameter ratio, which significantly improves the flexural strength [11,12].

The change in flexural MOE according to wood flour fraction size is shown in Table 4. The results demonstrate, that increasing the wood flour fraction size has little effect on flexural MOE (best results are obtained with fraction I). It is seen that the addition of wood flour to the polymer matrix increases the flexural MOE of the composite and makes the material more brittle (up to 80%).

For improving the mechanical properties of WPCs, it is very important to have strong bonding between the polymer matrix and wood filler. Therefore, the influence of coupling agents on the mechanical properties was investigated and the results are given in Table 4 and Fig. 2. From Fig. 2 it can be seen that

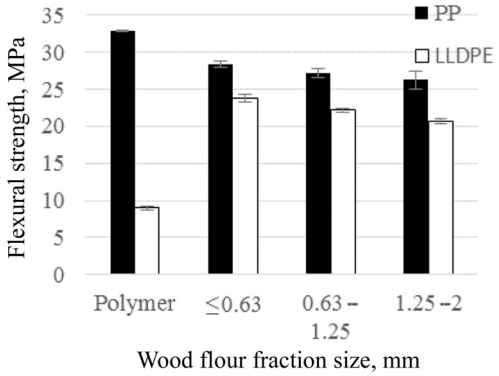

Fig. 1. APTES modified wood flour fraction size impact to the flexural strength of WPCs.

Table 4. Flexural and impact properties of WPCs

Sample	Flexural strength, MPa	Flexural MOE, GPa	Impact strength, kJ/m^2
PP1	32.8	1.01	14.53
PP2	23.5	1.30	6.41
PP3	23.8	0.87	7.26
PP4	25.4	1.20	7.69
PP5	28.3	2.34	4.03
PP6	25.6	1.82	5.57
PP7	27.1	2.07	4.99
PP8	27.4	1.74	8.08
PP9	26.2	1.91	4.67
LLDPE1	9.0	0.23	74.59
LLDPE2	20.7	0.76	8.13
LLDPE3	16.3	0.51	16.97
LLDPE4	15.3	0.49	14.71
LLDPE5	23.8	1.00	7.57
LLDPE6	18.2	0.65	11.31
LLDPE7	22.2	0.92	9.25
LLDPE8	21.8	0.91	10.29
LLDPE9	20.7	0.83	8.59

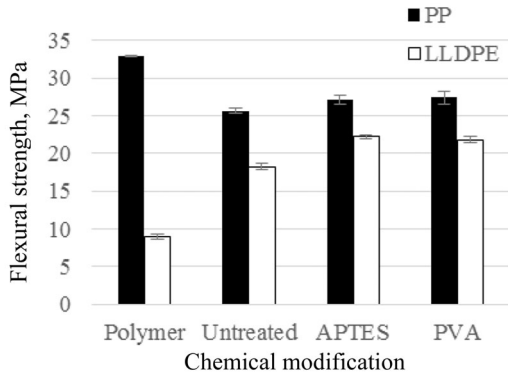

Fig. 2. Coupling agent influence to the flexural strength of WPC (fraction II).

chemically pretreated wood flour gives better coupling with the polymer matrix and thereby better flexural strength values than composites with untreated wood flour. Lower flexural strength values with untreated wood flour composites can be explained by poor bonding between hydrophilic wood flour and hydrophobic polymer and therefore by poor wood flour dispersion into the polymer matrix [13]. Poor coupling of the polymer and wood flour may also be the reason why the flexural strength decreased with increasing the dimensions of the wood fibres.

With consideration of the flexural strength and influence of additives, the wood flour processed with APTES gives WPC the best flexural strength values. The results from Table 4 and Fig. 2 show that the addition of APTES to the wood flour increased the flexural strength 6% of PP/wood flour composite and 22% of LLDPE-g-MAH/wood flour composite. The flexural MOE also increased with APTES modified wood flour composites resulting in a stiffer material (Table 4). With APTES, flexural strength increased most (22%) with the flour/LLDPE-g-MAH composite. PVA gave best flexural strength increase of 7% with wood flour/PP composite. However, flexural MOE decreased while modifying wood flour with PVA.

WPCs were also prepared with Aspen BCTMP. From Table 4 it can be seen that Aspen BCTMP gave similar results to wood flour composites. Adding 20% of Aspen BCTMP to LLDPE-g-MAH polymer matrix increased flexural strength of composite for 23%, almost as much as the wood flour fraction I. Since Aspen BCTMP was refined to very fine flour (≤0.126 mm), then it was distributed evenly into the polymer matrix and therefore flexural properties increased significantly. Previous studies have shown that wood particle size over 1 mm increases significantly mechanical properties of WPCs and size under 1 mm decreases the influence on mechanical properties [14].

3.2. Charpy impact strength

The results of Charpy impact strength are shown in Figs 3 and 4 and Table 4. The results show that addition of wood flour to the polymer matrix decreased impact strength more than 7 times, which correlates with other studies [15,16]. The addition of wood flour to the polymer generates stress concentration regions in composites which require less energy for cracking the composite, and thereby reducing the impact strength. Therefore, the addition of wood flour makes the composite material more brittle.

WPC material impact properties are influenced by the size of wood fibres. From Fig. 3 it can be seen that the lowest impact strength was with wood flour fraction I. Highest impact strength values were obtained with wood flour fractions II and III, which correlates with other studies [15–19]. This can be explained by the fact that larger wood particles have greater strength and thereby the strength of the composite increases. The results of this research suggest that addition of wood flour to the LLDPE-g-MAH makes it almost as brittle as the PP composite. In the composite, wood flour reduces the mobility of polymer chains, thereby reducing their energy-absorption capability by impact test. Poor linkage

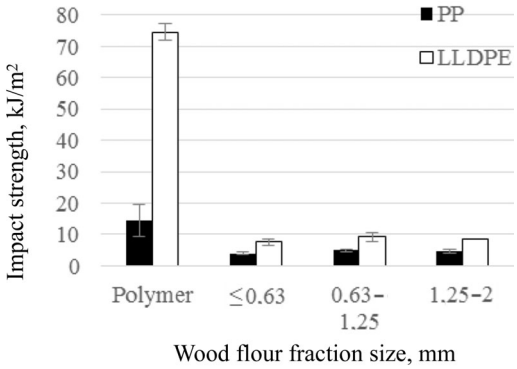

Fig. 3. APTES modified wood flour fraction size influence to the impact strength of WPCs.

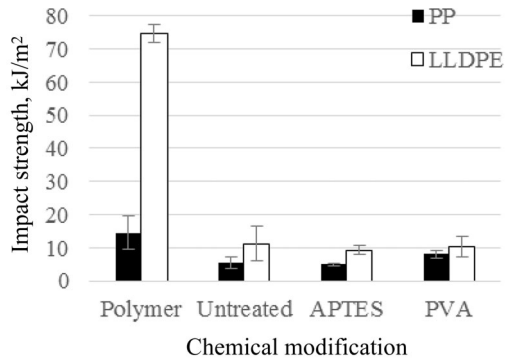

Fig. 4. Coupling agent influence to the impact strength of WPC (fraction II).

between the wood fibres and polymer matrix leads to microcracks in the impact test, which grow rapidly into large cracks leading test samples to break. Therefore it is important to have strong bonding between wood fibres and polymer matrix, what can be improved with coupling agents. Figure 4 shows that wood flour, processed with APTES, reduces impact strength the most, 10% for the PP composite and 17% for the LLDPE-g-MAH composite. PVA influenced less, reducing impact strength up to 10%. The largest decrease in impact strength of APTES modified wood flour composites is due to the fact that APTES bonds wood fibres with the polymer matrix better than PVA. With weaker bonding between polymer and wood fibres, impact force influenced more the polymer matrix. Therefore impact strength with PVA was lower than with APTES. Increasing impact strength values of PVA modified wood flour/PP composites are due to the poor interfacial bonding between polymer and wood flour, which results in different cracks in the composite during the impact testing (Fig. 4) [20].

The results of Aspen BCTMP are shown in Table 4 and are similar to the wood flour based composites. Adding only 20% of Aspen BCTMP fine flour, decreased impact strength almost as much as adding 35% wood flour.

3.3. Influence of water absorption and swelling

Water absorption and swelling of the composites during the accelerated weathering are shown in Table 5. The highest water absorption values were obtained with wood flour fraction II, 2.8% for PP composite and 1.2% for LLDPE-g-MAH composite. Water absorption was lowest with wood flour fraction I. Higher water absorption values with PP composite can be explained with the fact that the PP composite had lower amount of MAH coupling agent than the LLDPE-g-MAH. Therefore PP composites had weaker bonding between the polymer matrix and wood flour, which allowed water more easily to impact wood fibres in the composite. The results show that increasing the wood flour fraction size increases the water absorption of the composites, which also correlates with other studies [21,22]. Larger wood particles have an uneven interfacial layer what makes them more vulnerable to moisture.

Table 5. Water absorption and swelling of WPCs after 3 weeks of soaking in water

Sample	Absorption, %			Swelling, %		
	After 1 week	After 2 weeks	After 3 weeks	After 1 week	After 2 weeks	After 3 weeks
PP3	0.0	0.0	0.9	2.5	2.4	2.2
PP4	0.3	0.1	0.9	2.1	2.1	1.9
PP5	1.5	2.3	2.4	2.3	2.6	2.5
PP7	1.6	2.1	2.8	2.9	3.4	2.7
PP8	1.5	2.6	3.1	3.2	3.4	2.7
PP9	1.1	1.8	2.1	2.9	3.4	2.9
LLDPE3	0.5	0.3	0.7	0.6	1.2	0.8
LLDPE4	0.6	1.0	0.7	1.8	1.6	1.6
LLDPE5	0.2	1.4	1.1	1.9	2.0	1.8
LLDPE6	0.3	1.3	1.2	1.4	1.8	1.7
LLDPE7	0.6	1.1	1.2	1.8	1.3	1.5
LLDPE9	1.1	1.0	1.0	1.3	0.8	1.0

In Table 5, there is a clear difference in swelling the two different polymers. With PP composites the swelling increases when wood flour fraction size increase. Highest swelling of 2.9% was obtained with wood flour fraction III. However, with LLDPE-g-MAH there is the opposite situation. This can be explained by the inhomogeneity of LLDPE-g-MAH samples.

Table 5 shows that PVA modified wood flour/PP composite swelling increased 3% and water absorption up to 10% more than APTES modified wood flour/PP composites. The reason for this is that, PVA is a water-soluble polymer, which links with water through hydrogen bonding, and therefore reduces moisture resistance of composites. APTES molecules can react with cellulose OH group through amine groups, resulting in a strong bond [23]. To improve moisture resistance of WPC, it is very important to have strong bonding between wood fibres and polymer matrix.

Moisture affects also the mechanical properties of WPC. Table 6 shows that flexural strength and MOE decrease during the water soaking cycle. After 3 weeks of water soaking, the WPCs flexural strength decreased for 3% and flexural MOE for 14%. It shows that flexural strength decreased most with the PVA modified wood flour fraction II/LLDPE-g-MAH composite, about 5%. Best results were obtained with APTES modified wood flour fraction I composites, 1.2% for PP composite and 2.1% for LLDPE-g-MAH composite. It can be concluded that APTES binds wood fibres through OH groups and makes them hydrophobic, thus the moisture resistance is greater than the PVA modified wood flour composites. Moisture causes swelling of wood particles in WPC which then causes microcracks in the matrix, and therefore the flexural strength and MOE decrease.

Moisture also affects the impact properties of WPCs. Based on the results of Table 6, it can be seen that impact strength increases 17% on average after 3 weeks of water soaking cycle. This can be explained by the plasticizing effect on wood fibre and polymer interface during the moisture impact [4]. During the further absorption of water into composite, the wood fibre and polymer interface weakens, and as a result the impact strength also starts to decrease. Figure 5 shows scanning electron microscope (SEM) micrograph of the surface change of a WPC sample after 3 weeks of water soaking. Micrograph shows separation between wood flour and polymer, which indicates the lack of interfacial adhesion. Figure 5b shows a WPC specimen after 3

Table 6. Accelerated weathering influence on the mechanical properties of WPCs

Sample	Flexural strength, MPa			Flexural MOE, GPa			Impact strength, kJ/m^2		
	Untreated	After 3 weeks of water soaking	After 3 weeks of UV radiation	Untreated	After 3 weeks of water soaking	After 3 weeks of UV radiation	Untreated	After 3 weeks of water soaking	After 3 weeks of UV radiation
PP3	23.80	23.42	25.54	0.87	0.90	0.97	7.26	8.94	8.81
PP4	25.42	25.13	26.00	1.20	0.97	1.26	7.69	8.01	7.11
PP5	28.33	27.97	27.55	2.34	2.18	2.41	4.03	4.61	4.17
PP7	27.08	26.45	25.38	2.07	1.67	2.08	4.99	6.65	4.66
PP8	27.41	26.71	25.85	1.74	1.49	1.74	8.08	7.19	6.56
PP9	26.22	25.04	24.79	1.91	1.68	1.88	4.67	5.26	4.52
LLDPE3	16.27	14.18	15.99	0.51	0.46	0.48	16.97	22.07	17.84
LLDPE4	15.31	13.38	15.02	0.49	0.42	0.46	14.71	15.19	13.55
LLDPE5	23.78	23.27	22.22	1.00	0.53	0.88	7.57	9.08	8.28
LLDPE6	22.17	21.40	21.86	0.92	0.89	0.89	9.25	11.36	9.48
LLDPE7	21.76	20.76	21.17	0.91	0.87	0.88	10.29	10.64	8.84
LLDPE9	20.71	20.16	20.42	0.83	0.77	0.84	8.59	9.88	9.60

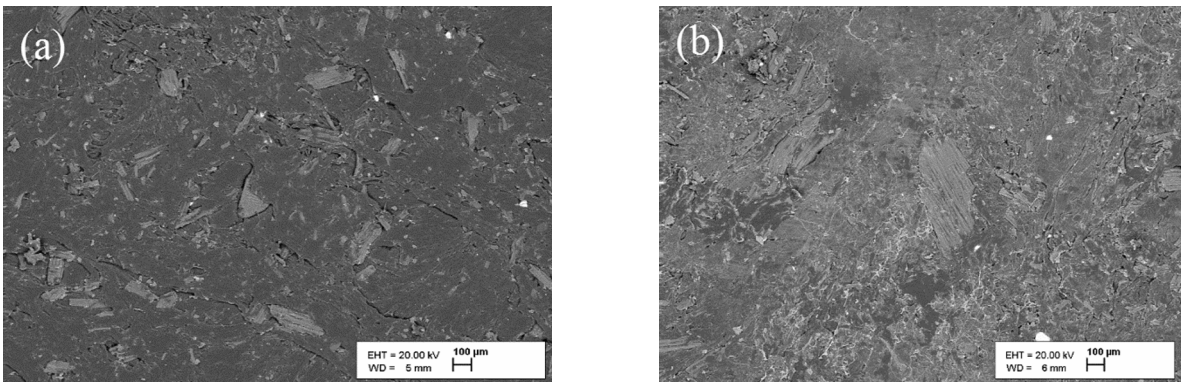

Fig. 5. SEM micrographs of the WPC surface: composite LLDPE-g-MAH surface (a), and composite LLDPE-g-MAH surface after water soaking (b).

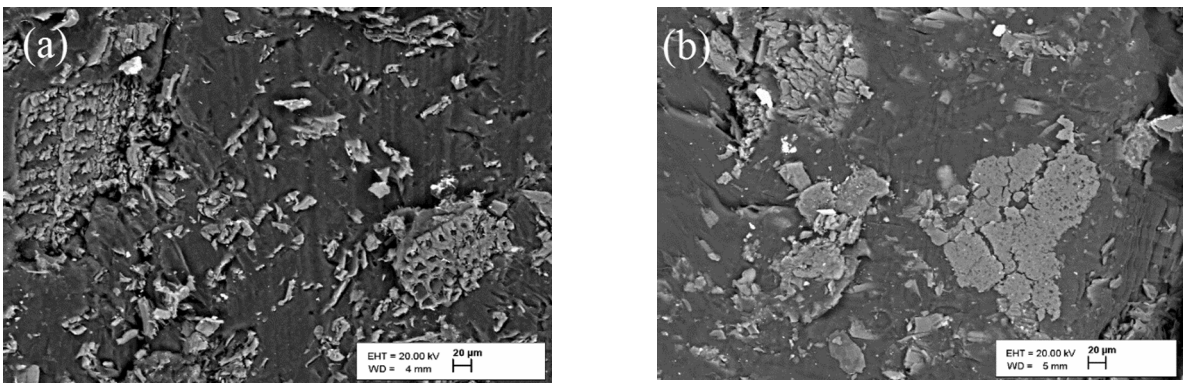

Fig. 6. SEM micrographs of WPC cross-section: composite PP cross-section (a), and composite LLDPE-g-MAH cross-section (b).

weeks of water soaking, where cracks can be observed in the polymer matrix as well as wood-matrix delamination. Figure 6 shows cross-section areas of PP and LLDPE-g-MAH composites. It can be seen that the LLDPE-g-MAH composite has smoother surface than the PP composite and it is caused by better interfacial adhesion between wood filler and polymer matrix on the LLDPE-g-MAH composite.

In addition, Aspen BCTMP (20% of Aspen BCTMP fine flour added to the polymer) composites were also tested, which gave similar results to the 35% wood flour based composites. This indicates that fine flour composite properties are more homogeneous. The results of the tests are shown in Table 6. It can be concluded that after 3 weeks of water soaking cycle, WPC becomes weaker and more elastic.

3.4. Influence of UV radiation

UV radiation influence on the flexural and impact properties of WPCs are shown in Table 6. The results show that after the UV radiation exposure for 3 weeks (500 h), flexural strength of WPCs decreases about 4%. The greatest flexural strength decrease, of 6.5%, was obtained with APTES modified wood flour fraction I/LLDPE-g-MAH composite. The smallest change in flexural strength, 1.4%, was obtained with the APTES modified wood flour fraction II and II/LLDPE-g-MAH composites. However, the results show differences in flexural strength changes for PP and LLDPE-g-MAH composites after UV radiation exposure. UV radiation has strong influence on the wood fibres, which causes photodegradation, causing a decrease in WPCs mechanical properties.

Table 6 shows that flexural MOE decreases after UV radiation exposure to LLDPE-g-MAH composites. However, for PP composites, flexural MOE slightly increases after exposure. This difference may be due to the fact that, when composites are exposed to UV radiation, the crystallinity of PP initially increases, then decreases [24]. Therefore, the results show that PP composite structure tolerates the influence of UV radiation much longer than the LLDPE-g-MAH composite, which will degrade already after 500 h. For different wood flour fraction sizes, best results were achieved with fraction I and UV radiation influence increase when the fraction size was increased. Results also show that PVA modified wood flour composites had slightly better resistance to UV radiation exposure than the APTES modified wood flour composites.

As for the impact strength, the UV radiation exposure increased the impact strength of composites. Impact strength increased most (11.8%) with wood flour fraction III and LLDPE-g-MAH based composites.

In addition, Aspen BCTMP based composites were also tested. The results were similar to the wood flour based composites. Based on this research, it can be said that UV radiation exposure makes WPC material more elastic.

3.5. Colour analysis

The lightness (L^*) and colour coordinates (a^* and b^*) as well as the total change of colour ΔE^* of the WPCs are shown in Figs 7 and 8. Figure 7 shows that lightness of the composites increased for most of the composites after 3 weeks of the water soaking cycle. The biggest lightness change occurred in the composite LLDPE9, the most significant total colour change occurred in the composite LLDPE6, where colour changed from yellow to blue (9.9 units). Colour measuring shows the loss of red and yellow colour during water soaking, which makes composites faint and turn into grey tone. However, some test samples shifted towards yellow and red, and made composites darker.

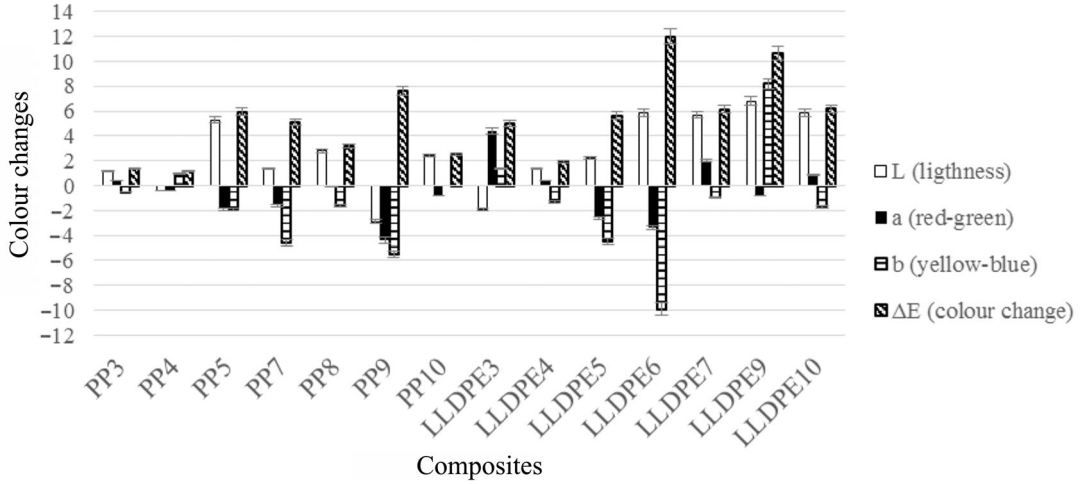

Fig. 7. Colour changes after 3 weeks of water soaking.

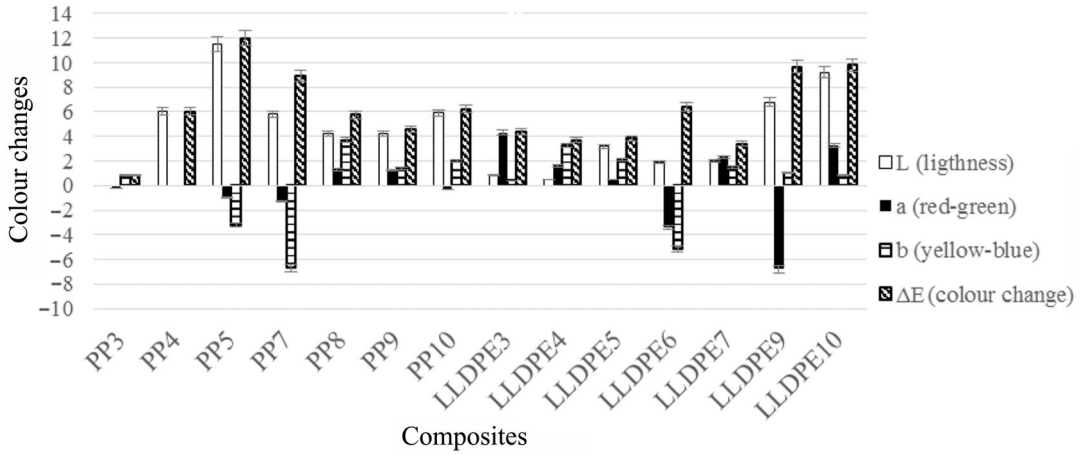

Fig. 8. Colour changes after 3 weeks of UV radiation exposure.

In case of UV radiation exposure, all the test samples turned lighter, as shown in Fig. 8. This is also in accordance with other studies [24,25]. During the UV radiation, most of the samples also shifted to more red. There is also a noticeable change towards green colour in many composites. The lightness and yellowness of the composites during weathering is caused by the lignin component decomposition in wood fibres. Redness is determined with the extractives content of the wood. Discoloration of WPCs during UV radiation exposure is a combination of both chemical and physical changes. Photooxidation of wood components initiate chemical changes in WPCs, which determine the primary colour change. If the adhesion between wood flour and polymer is strong enough to anticipate surface erosion and removal of wood particles, then composites turn darker and yellowish. However, the loss of the degraded wood component and combination with degradation of the polymer matrix leads to fading of the WPC surface [9].

Based on colour analysis results, visual colour changes were not observed, but with colorimeter the colour changes were present.

4. CONCLUSIONS

The influence of UV radiation and moisture content on the mechanical and physical properties of WPCs were investigated. The effect of coupling agents and wood flour fractions size on the mechanical and physical behaviour of WPCs were also experimentally determined. The following conclusions can be drawn from the study.

- WPC mechanical properties depend mostly on the wood fraction size. Smaller wood fraction size gives better flexural properties than the larger wood fraction size.
- It is very important to choose right coupling agent. Best test results were obtained with composites made of APTES modified wood flour and LLDPE-g-MAH.
- Adding wood flour to the thermoplastic polymers decreased impact strength and made WPC more rigid and brittle.
- Water absorption and swelling increase with increasing the wood flour fractions size in the composites because of the decreasing interface between the polymer matrix and wood flour.
- Water absorption and swelling of WPC decreased the flexural MOE and strength while impact strength increased by making material weaker and increasing the deflection of WPC materials.
- UV-radiation decreases the flexural MOE and strength, while impact strength increases by making WPC material properties weaker.
- Weathering of WPC also changes material colours. UV-radiation increased the lightness of all specimens. However, SEM analysis showed cracks and voids on the cross-section area of WPC specimens after weathering tests.

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Kunstliku vanandamise mõju puitplastkomposiitide mehaanilistele ja füüsikalistele omadustele

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Käesolevas töös uuriti kunstliku vanandamise (niiskuse ja UV-kiirguse) mõju puitplastkomposiitide (PPK) mehaanilistele ning füüsikalistele omadustele. Uurimistöö eesmärgiks oli hinnata ka puidujahu fraktsiooni suuruse mõju PPK mehaanilistele omadustele ja kunstlikule vanandamisele. Komposiitide valmistamiseks kasutati kolme erinevat kase (*Betula*) puidujahu fraktsiooni ja kemi-termo-mehaanilist (KTM) haava (*Populus tremula*) puitmassi. Polümeerse maatriksina kasutati kaht erinevat termoplastilist polümeeri: Fusabond E MB226DE (maleiinhappe anhüdriidiga modifitseeritud LLDPE) ja polüpropüleeni (PP). Puidujahu ja KTM-i modifitseerimiseks kasutati kaht erinevat sidusagensit: 3-aminopropüül-trietüksüsilaani (APTES) ning polüvinüülalkoholi (PVA). Komposiidid valmistati

kaheteolises ekstruuderis ja katsekehad tehti survevalumeetodiga. Viidi läbi kiirendatud vanandamise katsed (UV-kiirguskamber ja vees leotamine), et hinnata vanandamise mõju PPK omadustele. Mehaanilisi omadusi testiti kolme punkti paindekatsega ja Charpy pendliga löögikatsega. Lisaks uuriti ka PPK värvuse muutust peale vanandamist.

Mehaanilistest katsetest järeldus, et PPK materjali mehaanilised omadused sõltuvad kõige rohkem puidujahu sisaldusest ja fraktsiooni suurusest. Puidu lisamine tõstis materjali paindeelastsusmoodulit ja vähendas löögitugevust, muutes materjali jäigemaks ning rabedamaks. Kõige paremad paindeomadused andis puidujahu fraktsioon I, sest see oli komposiidis kõige ühtlasemalt jaotunud. Puiduosakeste suuruse kasvades muutus materjal elastsemaks, vähenes paindetugevus ja kasvas löögitugevus. Niiskus vähendas puidukiudude tugevust ja muutis need elastsemaks, mistõttu PPK tugevusomadused muutusid nõrgemaks ning läbipaine suurenes. UV-kiirguse toimel vähenesid mehaanilised omadused, paindeelastsusmoodul ja paindetugevus, löögitugevus kasvas. PPK materjal muutus elastsemaks. Lisaks muutus PPK materjal UV-kiirguse toimel heledamaks.