EFFECTS OF WOOD PARTICLE SIZE AND TEST SPECIMEN SIZE ON MECHANICAL AND WATER RESISTANCE PROPERTIES OF INJECTED WOOD–HIGH DENSITY POLYETHYLENE COMPOSITE

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Abstract. The objective of this study was to evaluate mechanical (tensile modulus and strength and flexural modulus and strength) and physical (water absorption and thickness swelling) properties of wood–plastic composites made from industrial wood particles used for manufacturing three-layer particleboards and high-density polyethylene. The effects of wood particle (WP) size (0.25-0.5, 0.5-1, and 1-2 mm) and test specimen cross section (4×10, 6×15, and 8×20 mm^2) were investigated. Mechanical properties increased with increasing WP size because of the higher aspect ratio of larger WPs. Composites with larger WPs had lower water resistance. The values of both mechanical and water absorption properties derived from specimens with a larger cross-sectional area were lower than those derived from specimens with a smaller cross-sectional area.

Keywords: High-density polyethylene, mechanical properties, particle size, physical properties, wood particle, wood–plastic composite.

INTRODUCTION

Because of their numerous advantages, wood–plastic composites (WPC) have been increasingly used for many applications (Clemons 2002; Jacob 2006; Klyosov 2007; Ashori 2008; Stark et al 2009; Thomson et al 2010). This results in growth requirements for these composites. One of these requirements may be superior mechanical properties. The use of large wood particles (WPs), larger than particles of wood flour and short wood fibers, can be particularly effective in achieving this purpose.

The effect of WP size on WPC mechanical properties was primarily evaluated for conventional WPC with wood flour or short wood fibers. The smallest WPs were taken into account by Gallagher and McDonald (2013). They studied the effect of WP size on the flexural properties of WPC made from WP of less than 74-177 μm and high-density polyethylene (HDPE). This effect depended on WP content (30%, 40%, and 50% by weight) and the presence of a coupling agent. Flexural modulus generally increased with increasing WP size regardless of WP content and presence of coupling agent, whereas the effect of WP size on flexural strength was dependent on WP content. This strength increased with increasing WP size for WPC with 30 wt% wood content and decreased for WPC with 50 wt% wood content. Zaini et al (1996) investigated the effect of WP size on tensile strength and flexural modulus of WPC from WP and polypropylene (PP) with 40 wt% WP content and with WP of 63-250 μm. They found that both mechanical properties increased considerably when WP size increased. A similar range of WP size, from 100 to 300 μm, was taken into account by Khalil et al (2006). They showed that the tensile modulus and strength

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and flexural modulus of WPC consisting of WP (10, 20, 30, 40, and 50 wt%) and recycled PP decreased with increasing WP size. Stark and Berger (1997) and Stark and Rowlands (2003) concluded that the tensile modulus and strength and flexural modulus and strength of WPC made from WP (40 wt%) and PP increased slightly with increasing WP size from 64 to 215 μm and then decreased when WP size reached 513 μm. Large WP of 150-850 μm were used to prepare WPC consisting of 25, 35, and 45 wt% WP and HDPE by Bouafif et al (2009). They found that tensile modulus and strength and flexural modulus and strength of WPC increased with increasing WP size regardless of WP content. Takatani et al (2000) studied flexural strength of WPC made from softwood flour, steam-exploded beech flour, polyvinyl chloride (PVC), and polystyrene using two WP sizes: 125 and 841 μm. They observed that WPC with WP of 125 μm had greater strength when softwood flour was the wood component and conversely had less strength when a mix of softwood and steam-exploded beech flour was the wood component.

Apart from small WP, short wood fibers are used to manufacture WPC. Fiber length is taken as a measure of fiber size. Migneault et al (2008, 2009) concluded that tensile modulus and strength and flexural modulus and strength of WPC made from wood fibers (40 wt%) and HDPE increased with increasing wood fiber length from 196 to 481 μm. The reverse effect of wood fiber length on flexural modulus and strength was found by Cui et al (2008) regarding WPC containing 10, 20, 35, and 50 wt% wood fibers and HDPE. These properties decreased regardless of wood fiber content when wood fiber length increased from 75 to 900 μm. Kumari et al (2007) observed that flexural modulus and strength of WPC made from wood fibers (80 wt%) and PP increased with increasing wood fiber length from 120 to 300 μm and next decreased with increasing wood fiber length to 900 μm. Liew et al (2000) showed that tensile and flexural strengths of WPC consisting of wood fibers (40 wt%) and PP increased with increasing wood fiber length from 0.5 to 2.0 mm. Lee at al (2001) observed a considerable increase in tensile modulus and strength and flexural strength of similar WPC when wood fiber length increased from 1.7 to 3.2 mm.

The results of the presented studies show that the effect of WP size and wood fiber length on WPC mechanical properties varied. These properties increased considerably because of increasing WP size and wood fiber length, but in some cases, they increased only slightly or even decreased. The differences in these results were caused by such factors as thermoplastic type, wood content, WP geometry, coupling agent presence, and processing method.

The mechanical properties of WPC with WP larger than particles of wood flour have seldom been studied. Bledzki and Faruk (2003) found that tensile modulus and strength and flexural modulus and strength of wood (50 wt%)–PP composites made with large industrial WP and typical wood flour were almost the same. Chen et al (2006) studied the effect of WP size on flexural modulus and strength of WPC made from WP (50, 60, 70, and 75 wt%) of a size from less than 0.59 mm to more than 1.18 mm and HDPE. They concluded that increasing WP size improved these properties. Najafi and Khademi-Eslam (2011) investigated flexural modulus and strength of lignocellulosic filler (60 wt%)–recycled HDPE composites made with agricultural residue and industrial wastes such as flour of rice hulls, wood sawdust, sanding flour from medium-density fiberboard, and particleboard sawdust. They found that the properties of these WPC differed considerably because of differences in the particle size of lignocellulosic fillers. In recent studies, the effect of WP size on tensile modulus and strength and flexural modulus and strength of WPC made from industrial WP (40 wt%) used for producing particleboards and PP (Gozdecki et al 2012), PVC (Kociszewski et al 2012), and low-density polyethylene (LDPE) (Gozdecki et al 2011) was investigated. Tensile and flexural properties of WPC increased considerably with increasing WP size from 0.25-0.5 mm to 1-2 mm and decreased slightly when WP size increased to

2-4 mm. The increases in these properties differed greatly, ranging from 9% for tensile strength of WPC with PVC to 41% for flexural modulus of WPC with LDPE.

WP size and wood fiber length also affected water absorption and thickness swelling of WPC. Steckel et al (2007) investigated the process of water diffusion into WPC containing small WP (40 wt%) and PP and stated that water absorption of WPC with WP from 150 to 180 μm after 238 da of immersion was greater than that of WPC with WP of less than 150 μm. Khalil et al (2006) showed that water absorption of WPC consisting of WP (10, 20, 30, 40, and 50 wt%) and recycled PP increased considerably after 100 da of immersion with increasing WP size from 100 to 300 μm. Bouafif et al (2009) investigated the effect of WP size on water absorption of WPC made from WP (25, 35, and 45 wt%) and HDPE. They observed an increase in water absorption after 28 da of immersion when WP size increased from 150 to 850 μm. However, this increase was significant only for WPC with a low content of WP. Chen et al (2006) studied the effect of WP size on water absorption and thickness swelling of WPC made from WP (50, 60, 70, and 75 wt%) of a size from less than 0.59 to more than 1.18 mm and HDPE. They concluded that after 24 h of immersion, these properties generally increased with increasing WP size regardless of WP content. Studies by Najafi and Khademi-Eslam (2011) focused on different lignocellulosic fillers (60 wt%) of recycled HDPE. They found that both short- (24 h) and long-term (84 da) water absorption and thickness swelling of WPC depended on the filler type. WPC containing larger particles of sanding flour from medium-density fiberboard and wood flour showed greater water absorption and thickness swelling, both after 24 h and 84 da, than smaller particles of flour of rice hulls and particleboard sawdust. The effect of wood fiber length on water absorption and volumetric swelling of WPC made from wood fibers (40 wt%) and HDPE using two processing methods (extrusion and injection molding) was studied by Migneault et al (2008, 2009). They showed that the values of these properties increased with increasing wood fiber length from 196 to 481 μm for different immersion times (250, 500, 1000, 1500, and 2500 h).

Typical test specimens for determining tensile and flexural properties of plastics have a cross section of 4×10 mm^2. Such specimens are also used for determining properties of WPC. This size is justified with regard to WPC with wood flour containing small WP. However, greater cross section appears to be advantageous in the case of WPC with larger WP, which makes the WPC more heterogeneous. Gozdecki et al (2012) and Kociszewski et al (2012) found that the values of WPC mechanical properties determined on specimens with larger cross section, eg, 6×15 mm^2, were lower than the values received for specimens with a cross section of 4×10 mm^2.

The objective of this study was to investigate the effect of WP size on mechanical and water absorption properties of WPC made from industrial WP used for manufacturing particleboards and HDPE. In addition, the effect of cross-section size of test specimens made by injection molding was evaluated.

MATERIALS AND METHODS

Materials

The HDPE used in this study was Tipelin 550-13 in granule form obtained from Basell Orlen Poliolefins (Plock, Poland) with density of 956 kg/m^3 and melt flow index of 0.35 g/10 min (190°C/2.16 kg). Raw wood materials were industrial softwood particles used for manufacturing three-layer particleboards. Fine particles were used for face layers and coarse particles for the core layer. They were provided by Kronospan Szczecinek (Szczecinek, Poland). WPs were screened to obtain three WP sizes: small (S), 0.25-0.5 mm, and medium (M), 0.5-1 mm, produced from fine particles and large (L), 1-2 mm, from coarse particles (Fig 1). A total of 200 randomly selected WPs of each size were measured,

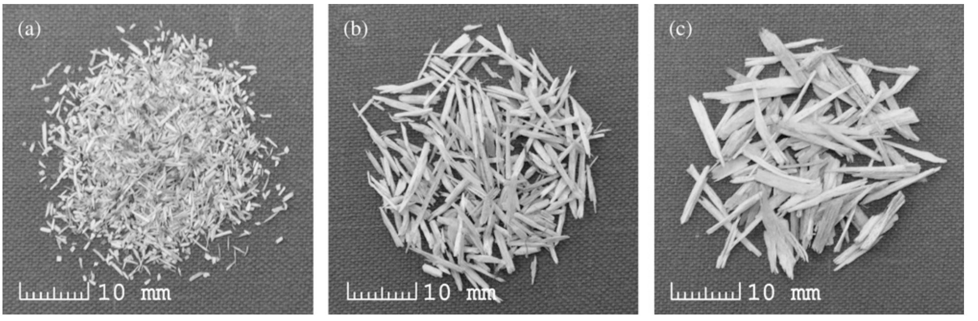

Figure 1. Screened WPs: (a) S, (b) M, and (c) L.

Table 1. Length, length–thickness, and width–thickness ratios of WP.[a]

WP size	Length (mm)	Length–thickness	Width–thickness
S	2.2 (0.9)	11.2 (2.9)	2.3 (0.6)
M	4.6 (1.3)	15.4 (3.7)	2.4 (1.0)
L	12.7 (5.8)	21.2 (5.9)	2.6 (1.3)

[a] Standard deviations in parentheses.

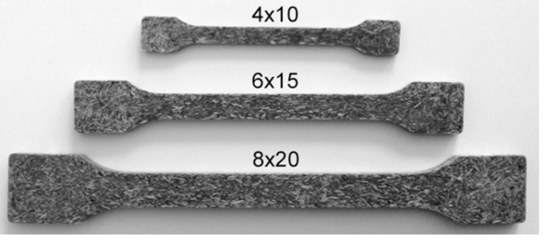

Figure 2. Test specimens of different size.

and length-to-thickness ratio (aspect ratio) and width-to-thickness ratio were calculated to obtain WP geometric characteristics (Table 1).

Sample Preparation

Before the mixing process, WPs were dried to obtain 1-2% MC. Next, WP and HDPE were dispensed into the hopper using two DSH screw feeders (Hydrapress, Biale Blota, Poland) and then mixed at a ratio of 40:60 by weight in a one-step mixing process in the feed zone of an AH-80 injection molding machine (Metalchem, Poznan City, Poland). Test specimens were made by injection molding using a temperature of 120-180°C from feed zone to die zone. Diameters of the injection die and sprue bush were enlarged to 4.5 and 8 mm, respectively, to minimize mechanical degradation of WP during molding. Cross sections of the runner and gate were 10×10 and 6×6 mm^2, respectively.

Three cross-sectional areas of test specimens were assumed: 4×10, 6×15, and 8×20 mm^2. The length of those with cross section of 4×10 mm^2 was 150 mm and was consistent with EN ISO (1996), and the lengths of those with cross sections of 6×15 and 8×20 mm^2 were sufficiently larger amounting to 230 and 310 mm, respectively. The specimens (Fig 2) were conditioned at constant temperature of 25°C and 65% RH for 2 wk before testing.

Experimental Design

Experiments were conducted according to a 3×3 complete factorial design. Ten specimens were produced for each WPC combination and group of WPC properties, giving a total of 270 specimens (Table 2).

Table 2. Experimental design and number of specimens.

WPC	WP size	Cross-sectional area (mm^2)	Number of specimens: Tensile properties	Flexural properties	Physical properties
1	S	4×10	10	10	10
2	M	4×10	10	10	10
3	L	4×10	10	10	10
4	S	6×15	10	10	10
5	M	6×15	10	10	10
6	L	6×15	10	10	10
7	S	8×20	10	10	10
8	M	8×20	10	10	10
9	L	8×20	10	10	10

Mechanical Testing

The mechanical properties of tested WPC were evaluated in relation to tensile (modulus and strength) and flexural (modulus and strength) properties. Tensile and flexural tests were conducted with a crosshead rate of 2 mm/min in accordance with EN ISO (1996) and EN ISO (2005), respectively, using an Instron (Norwood, MA) 3367 machine.

Physical Testing

Determined properties characterizing the water resistance of WPC were water absorption and thickness swelling. The specimens were dried in an oven for 24 h at 100°C, and the weight and thickness of oven-dried specimens were measured. The specimens were then immersed in distilled water at room temperature for 28 da, and their weight and thickness were measured at different times *t*. The values of water absorption and thickness swelling were calculated according to the following formulas:

$$\mathrm{WA}(\%) = \frac{W_t - W_0}{W_0} \times 100 \qquad (1)$$

$$\mathrm{TS}(\%) = \frac{T_t - T_0}{T_0} \times 100 \qquad (2)$$

where W_t is the specimen weight at time t, W_0 is the oven-dried weight, T_t is the specimen thickness at time t, and T_0 is the oven-dried thickness.

Water absorption and thickness swelling after two selected times of specimen immersion were assumed as specifying WPC water resistance: after 24 h for short-term water diffusion into WPC and after 28 da for long-term water diffusion.

Statistical Analysis

The obtained data were statistically analyzed using Statistica version 10 (StatSoft, Inc., Tulsa, OK). To evaluate the significance of the effects of WP size and cross-section size on WPC mechanical and physical properties, two-way analysis of variance (ANOVA) was conducted. Significance among average values of WPC properties was determined using Tukey's test.

RESULTS AND DISCUSSION

The results of the ANOVA showed that both mechanical and physical properties of WPC depended significantly on WP size and specimen cross-section size (Table 3).

Effect of WP Size

The mechanical properties of WPC as a function of WP size are given in Fig 3. Specimens of different cross-section size and with the same WP size were included in one group. Mean values of mechanical and physical properties for these groups as well as the results of Tukey's test are listed in Table 4. Values marked by different letters for a given property differed significantly at the 5% significance level.

WPC tensile and flexural properties increased with increasing WP size. Increases in tensile modulus, flexural modulus, tensile strength, and flexural strength with increasing WP size from S to L were on average 20%, 19%, 25%, and 24%, respectively. Increases in WPC mechanical properties with increasing WP size from M to L were greater than those with increasing WP size from S to M. Increases in tensile and flexural strengths depended on specimen cross-section

Table 3. Two-way ANOVA test on the effects of WP size and cross-section size on wood–plastic composite mechanical and physical properties (p-values).

	Tensile modulus	Flexural modulus	Tensile strength	Flexural strength	WA 24 h	WA 672 h	TS 24 h	TS 672 h
WP size	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
Cross-section size	<0.0001*	0.0192**	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
WP size × cross-section size	0.1292^{ns}	0.9961^{ns}	0.0194**	0.0097*	0.0003*	0.1204^{ns}	0.7814^{ns}	0.0523^{ns}

* Significant at 0.01; ** significant at 0.05; ^{ns} nonsignificant at 0.05; WA, water adsorption; TS, thickness swelling.

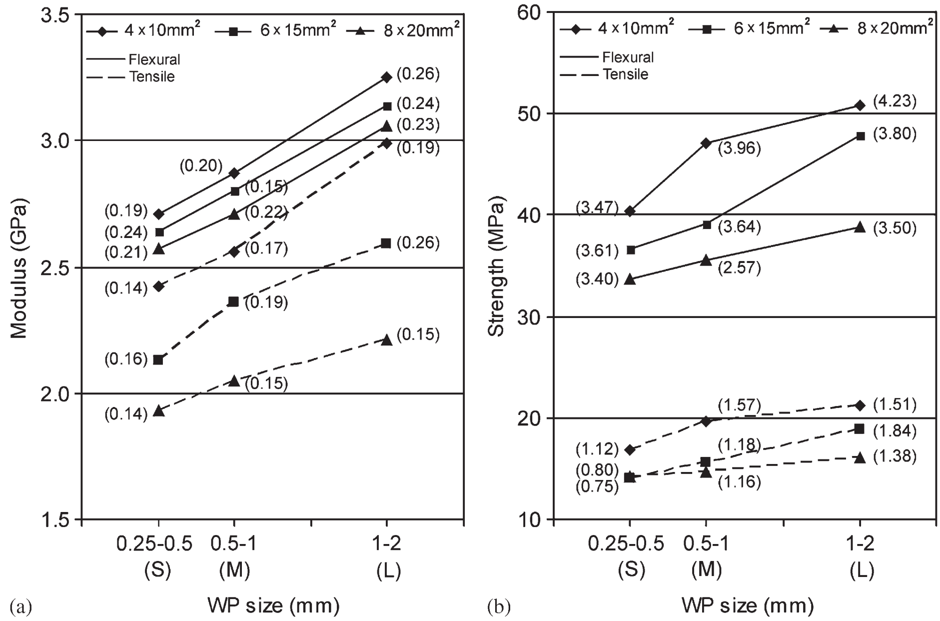

Figure 3. Effect of WP size on (a) tensile and flexural moduli of WPC, (b) tensile and flexural strengths of WPC. Standard deviations in parentheses.

Table 4. Effect of WP size on wood–plastic composite mechanical and physical properties.

WP size	Tensile modulus GPa	Flexural modulus GPa	Tensile strength MPa	Flexural strength MPa	WA 24 h %	WA 672 h %	TS 24 h %	TS 672 h %
S	2.16^{a}	2.64^{a}	15.0^{a}	36.8^{a}	2.08^{a}	5.63^{a}	1.67^{a}	3.42^{a}
M	2.33^{a}	2.79^{b}	16.6^{b}	40.6^{b}	2.18^{ab}	6.20^{b}	1.72^{ab}	3.69^{ab}
L	2.59^{b}	3.15^{c}	18.7^{c}	45.8^{c}	2.51^{b}	6.82^{c}	1.97^{b}	4.02^{b}

Values marked by different letters (a, b, or c) for a given property differed significantly at the 5% significance level.
WA, water adsorption; TS, thickness swelling.

size. They were smallest when the strengths were derived from the specimens with cross section of 8 × 20 mm^2.

The improvement in mechanical properties of WPC from the use of larger WP was a result of WP geometry. As the mechanics of fiber-reinforced composite materials show, one of the important factors affecting their mechanical properties is aspect ratio. Fibers with a greater ratio intensify stress transfer from the polymer matrix to the fibers, which results in improved mechanical properties of the composite. The validity of this rule with regard to WPC was indicated by several researchers (Zaini et al 1996; Stark and Rowlands 2003; Bouafif et al 2009; Ashori et al 2011; Gozdecki et al 2012; Kociszewski et al 2012). The values of this ratio for the WP used in this study were 11.2, 15.4, and 21.2 for S, M, and L particles, respectively (Table 1).

A similar effect of WP size on mechanical properties of WPC with WP larger than those contained in wood flour were found in other studies. Bouafif et al (2009) observed an increase of about 20% in flexural modulus and strength of WPC made from WP (45 wt%) and HDPE by injection method when WP size increased from 150 to 850 μm. Chen et al (2006) found an increase of about 15% in flexural modulus and strength of WPC made from WP (50 wt%) and HDPE by compression method when WP size increased from 0.59 to 1.18 mm. Unfortunately, the values of aspect ratio were not given in either of these studies. Presumably, larger WP had greater aspect ratio. Gozdecki et al (2012) studied the properties of WPC made from WP (40 wt%) and PP by injection method and reported that tensile and flexural strengths of WPC with WP of 1-2 mm were about 28% and 25%, respectively, greater than those of WPC with WP of 0.25-0.5 mm.

The water absorption and thickness swelling of WPC after 24 h and 28 da as a function of WP size are shown in Fig 4, and the results of Tukey's test for groups of specimens of different cross-section size and with the same WP size are given in Table 4. Both water absorption and thickness swelling increased steadily with increasing WP size. Increase in water absorption after 24 h depended on specimen cross-section size. It was the greatest when water absorption was derived from specimens with cross section of 4×10 mm^2. Increases in water absorption and thickness swelling with increasing WP size from S to L were on average 21% and 18%, respectively, both after 24 h and 28 da. Such effect of WP size on water absorption can be explained in two ways: 1) larger WP led to greater hydrophilic exposed surfaces and 2) poor adhesion between WP and the polymer matrix generated void spaces around the WP, and these voids in the bulk matrix were readily filled with water (Bouafif et al 2009). Increase in thickness swelling of WPC is a natural consequence of increase in water absorption.

The rule, the larger the WP size, the greater the water absorption, was found in other studies on WPC with WP larger than those of wood flour. Bouafif et al (2009) observed an increase of about 13% in water absorption (after 28 da of immersion) of WPC made from WP (25 wt%) and HDPE by injection method when WP size increased from 150 to 850 μm. Chen et al (2006) concluded that water absorption (after 24 h of immersion) of WPC made from WP (50 wt%) and HDPE by compression method with WP

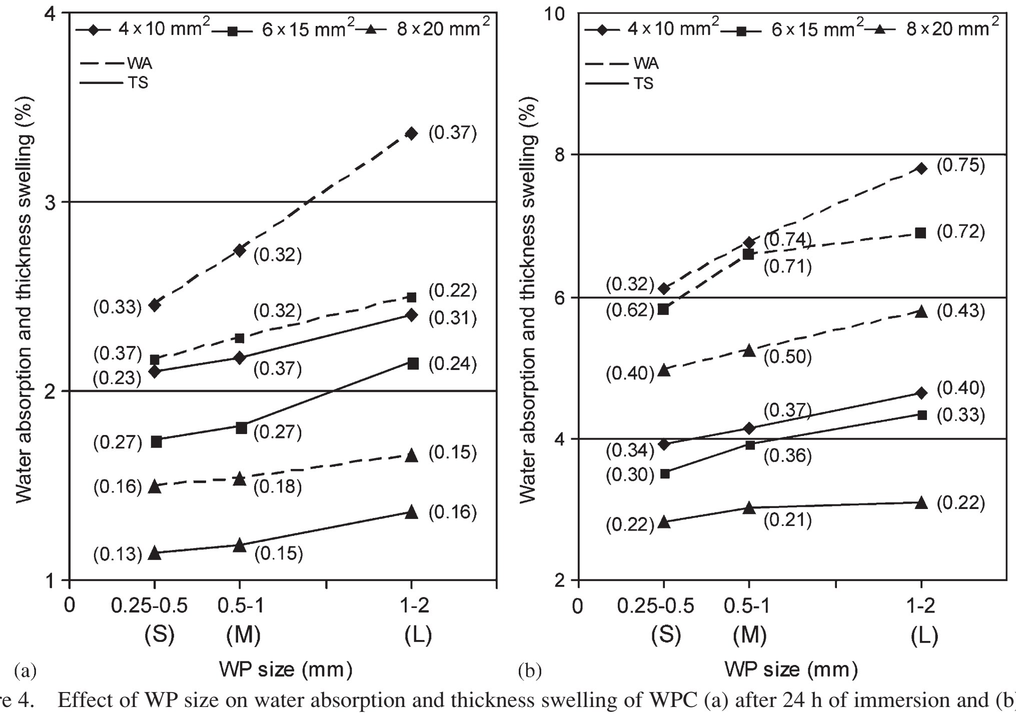

Figure 4. Effect of WP size on water absorption and thickness swelling of WPC (a) after 24 h of immersion and (b) after 28 da of immersion. Standard deviations in parentheses.

from 0.74 to 1.18 mm was greater by about 12% than that of WPC with WP of less than 0.59 mm.

Effect of Cross-Section Size

The effect of specimen cross-section size on WPC mechanical properties is shown in Fig 3. In general, properties that were determined with the use of specimens with greater cross-section had lower values. WPC tensile modulus, flexural modulus, tensile strength, and flexural strength derived from specimens with cross sections of $8 \times 20\ mm^2$ were on average 22%, 6%, 22%, and 21% smaller, respectively, than those derived from specimens with cross sections of $4 \times 10\ mm^2$. Decreases in tensile and flexural strengths depended on WP size. Decreases were smallest for WPC with S particles.

The phenomenon of lower strength of material when it is determined by means of larger specimens is known in the mechanics of materials. The greater the volume of material, the higher the probability of critical flaws and stress concentrations is. The voids between WP and polymer matrix are typical flaws in the case of WPC. A similar effect of specimen cross-section size on tensile and flexural strengths of WPC was found in previous studies on the properties of WPC made from industrial WP (40 wt%) and PP (Gozdecki et al 2012) or PVC (Kociszewski et al 2012) by injection molding method.

Lower values of water absorption and thickness swelling were derived from specimens with greater cross section (Fig 4). These properties derived from specimens with cross sections of $8 \times 20\ mm^2$ after 24 h of immersion were on average 44% and 45% smaller, respectively, than those derived from the specimens with cross sections of $4 \times 10\ mm^2$. After 28 da of immersion, water absorption and thickness swelling determined with the use of specimens with cross sections of $8 \times 20\ mm^2$ were on average 22% and 29% smaller, respectively, than those determined with the use of specimens with cross sections of $4 \times 10\ mm^2$.

Less water absorption of thicker specimens can be explained on the basis of water diffusion theory. For short times, when $M_t/M_\infty \le 0.5$, the following equation, based on Fick's theory, can be used (Espert et al 2004):

$$\frac{M_t}{M_\infty} = \frac{4}{L}\left(\frac{D}{\pi}\right)^{0.5} t^{0.5} \qquad (3)$$

where M_t is the MC at time t, M_∞ is the MC at the equilibrium state, D is the diffusion coefficient, and L is the thickness of the WPC specimen. The plots of water absorption vs $\sqrt{t}$ for WPC with large particles are given in Fig 5. The initial portion of the graphs is straight lines according to Fick's theory. Assuming that M_∞ as well as D is the same for a given WPC regardless of the specimen thickness L, it can be said that M_t is inversely proportional to L. The water absorption in this study, according to definition (Eq 1), is the same physical quantity as the MC. Thus, for short-term water absorption, the value of the ratio of water absorption determined on thicker specimens to water absorption determined on thinner specimens should be equal to the ratio of smaller to greater thickness. This ratio should

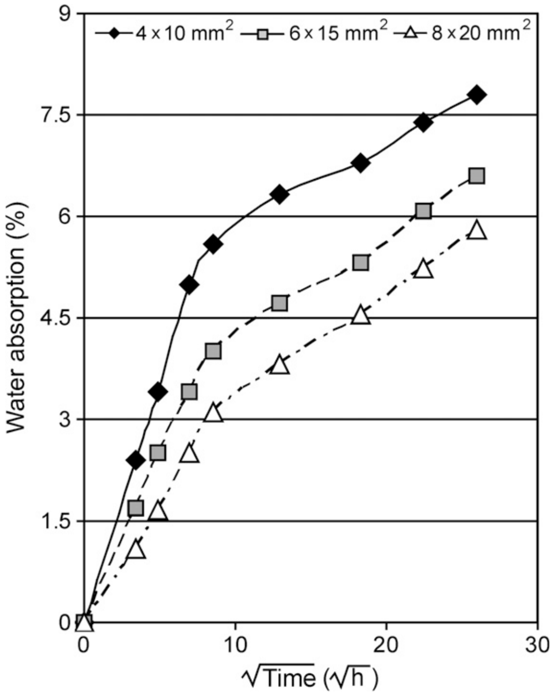

Figure 5. Water absorption as a function of square root of time for WPC with large particles and different cross sections of WPC specimens.

equal 1:2 when specimen thicknesses are 4 and 8 mm. Actual values of the ratio of water absorption determined on specimens with thicknesses of 4 and 8 mm, after 24 h of immersion, were 1:1.67; 1:1.79, and 1:1.98 for WPC with S, M, and L WP size, respectively. Therefore, values of this ratio were closer to 1:2 with larger WP in WPC.

CONCLUSIONS

Mechanical properties, including tensile modulus, tensile strength, flexural modulus, and flexural strength, and water resistance properties, including water absorption and thickness swelling, of WPC made from HDPE and industrial WP used for manufacturing particleboards depended significantly on WP size and cross-section size of injection-molded specimens. The large-sized WP with greater aspect ratio, used for manufacturing a core layer of particleboard, provided better mechanical properties than the small-sized WP used for manufacturing face layers of particleboard. The tensile and flexural moduli of WPC with large WP were greater by about 20% and the tensile and flexural strengths were about 25% greater than those of WPC with small WP. However, WPC with larger WP had lower quality water resistance properties. Water absorption and thickness swelling of WPC with large WP were greater by about 20% than those of WPC with small WP. Values of both mechanical and water absorption properties derived from specimens with a larger cross-sectional area were lower than those derived from specimens with a smaller cross-sectional area. WPC tensile modulus, flexural modulus, tensile strength, flexural strength, water absorption, and thickness swelling (after 28 da of immersion) derived from specimens with cross sections of 8×20 mm^2 were on average 22%, 6%, 22%, 21%, 22%, and 29% smaller, respectively, than those derived from specimens with cross sections of 4×10 mm^2.

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