PROPERTIES OF FLAT-PRESSED WOOD-POLYMER COMPOSITES MADE USING SECONDARY POLYETHYLENE

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ABSTRACT

The influence of wood filler type (particles, flour, mixture of waste chips and sawdust at ratio 10:90), its content (20, 40, and 60 %), board density (800, 900, and 1000 kg·m⁻³), pressing temperature (160, 180, and 200 °C) and pressing time (6.4, 8.0, and 9.6 min) on the properties of flat-pressed wood-polymer composites (WPC) using a conventional flat-press process was investigated in this study. Dry-blending method was applied for mixing wood fillers and secondary polyethylene (sPE). The highest strength and water resistance were observed in flat-pressed WPC made using wood particles. The highest values of bending strength were noticed in WPC with 40 % content of wood particles and 60 % content of sPE. The further increase or decrease of flat-pressed WPC improves with the increasing of secondary polyethylene contents from 40 % to 80 %. With increasing board density from 800 to 1000 kg·m⁻³ the properties of flat-pressed WPC also improved. With increasing pressing temperature from 160 to 200 °C and pressing time from 6.4 to 9.6 min the strength of flat-pressed WPC noticeably increased and water resistance considerably improved.

Keywords: wood-polymer composites, mechanical properties, physical properties, wood filler, secondary polyethylene.

INTRODUCTION

During the last few decades many investigations were carried out to create nontoxic wood composites materials. Wood-polymer composite (WPC) is one of such materials. About 90 % of WPC in the USA are produced using polyethylene (BLEDZKI *et al.* 2002, KLYOSOV 2007, NISKA and SAIN 2008). In Europe, the use of polyethylene is a little bit lower – about 70 %; polypropylene – 11 % and polyvinyl chloride – 9 % (EDER 2010, ROWELL 2005). Basically, virgin polymers are used for manufacture of WPC but recycled polymers can also be used (EDER 2010). For example, 50 % of virgin and 21 % of recycled PP, 41 % of virgin and 26 % of recycled PE were used for manufacture of WPC in 2009 in Europe (EDER 2010). In addition, it is predicted that in 2015 the accumulation of plastic wastes such as LDPE, HDPE and PP will be more 10000 kt, 5000 kt, and 4000 kt, respectively (DELGADO *et al.* 2007). The packaging sector continues to be the major consumer of plastics, with a stable share in total plastic consumption at just over 37 % in 2002 and 2003. Packaging is relatively short-lived and therefore packaging waste can be assumed to correspond roughly to the amount of packaging put on the market annually. Plastic packaging flowing into the household waste stream has been estimated by several

sources to amount for 65–75 % by weight of total plastic packaging (APME 2001). The prospective for total plastic packaging waste generated is expected to amount for 19314 kt, and the plastic packaging waste recovered to 13859 kt (DELGADO *et al.* 2007) and moreover combustion is undesirable (PÓŁKA *et al.* 2011). The packaging is mainly produced from LDPE, HDPE and PP. All this polymer wastes can be used for manufacture of WPC. Therefore, these polymeric raw materials for manufacture of WPC are virtually inexhaustible.

Such considerable percentage of using these thermoplastic polymers can be explained by their low melting temperature (110-160 °C). The thermal stability of wood becomes worse when higher temperatures (more than 200 °C) are used (KLYOSOV 2007). The further increase of temperature causes the decomposition of wood macromolecules and negative changes of wood properties.

Usually, wood flour is used as a filler for making WPC (BOTROS 2003, RADOJEVIĆ *et al.* 2006, SIMONSEN *et al.* 2004, STARK 1997, WINANDY *et al.* 2004), although wood sawdust (STARK 1997, WINANDY *et al.* 2004), wood fiber (H'NG *et al.* 2008, H'NG *et al.* 2010, IBACH and CLEMONS 2006, LIANG *et al.* 1994, YOUNQUIST 1995) and shaving (SALIKLIS and WHITE 2008, SEGERHOLM 2007) are also widely applied. Except wood particles, other agricultural residues, in particular wheat straw (DIGABEL *et al.* 2004) MENGELOGLU and KARAKUS 2008, SARDASHTI 2009) and rice straw (BUZAROVSKA *et al.* 2008, YAO 2008) are widely used as a filler to make WPC.

The size of filler particles, its type and the type of the thermoplastic polymer usually determine the method of WPC manufacture. There are many methods of WPC manufacture, as: extrusion, injection and compression moulding etc., (BLEDZKI *et al.* 2002, EDER 2010, KLYOSOV 2007, NISKA and SAIN 2008, ROWELL 2005). Extrusion is the predominant technology for manufacturing WPC both in the USA and in Europe (BLEDZKI *et al.* 2002, EDER 2010, KLYOSOV 2007). Extrusion gives an opportunity to obtain composites of endless profiles with high durability, water resistance, chemical resistance and ability to re-use (EDER 2010, RADOJEVIĆ *et al.* 2006, ROWELL 2005). Alternative technology is making WPC in flat presses. This technology is less studied and less explored but it has several advantages over extrusion: a relatively lower pressure is required compared to extrusion, the saving of wood raw material due to lower density of composite panels, higher productivity of pressing technology compared to the extrusion.

Such composites as particleboard, MDF and OSB are produced in flat presses. However, these composites are harmful, have insufficient water resistance and they are quite difficult to re-use. It limits the scope of their use. The thermosetting (UF and PF) resins are usually used to produce such composites, which lead to the emission of free formaldehyde under pressing of composites and during its operation. Therefore, WPC can be a serious competitor for particleboards, MDF and OSB if the technology for manufacturing wood-based boards will be adapted for producing WPC.

Few works were devoted to the production of WPC by flat pressing. It can be explained by the complication of getting homogeneous mixtures in ordinary mixer and by the retaining of the thermoplastic polymer on the surface of wood filler. The works (H'NG *et al.* 2008 and 2010) describe the making of flat WPC boards using extruder for mixing components and hot press for flat pressing. In other works (AYRILMIS *et al.* 2011 and 2012, AYRILMIS and JARUSOMBUTI 2011) the mixing of wood filler and thermoplastic polymer was accomplished using extruder and commercial PP Homopolymer G452 (H'NG *et al.* 2008 and 2010) or maleic anhydride-grafted PP (AYRILMIS *et al.* 2011 and 2012, AYRILMIS and JARUSOMBUTI 2011) as the matrix. However, application of these polymers leads to a significantly increased cost of finished products. Besides, the stitching of maleic anhydride to the chain of PP is needed for obtaining maleic anhydride-grafted PP, followed by

activation of the copolymer by heating (at the temperature of 170 °C) (H'NG *et al.* 2008, PROSZYK 1996). Therefore, the use of secondary polyethylene will reduce production costs and will allow WPC to compete with particleboard, MDF and OSB. Using secondary polyethylene, except for the economic effect has some other advantages. In particular, the destruction of the main chain with formation of simple and complex etheric bonds and hydroxyl groups occurs during the life of the polyethylene products, as a result of UV rays and oxygen. The presence of these groups enables to create chemical bonds between the reaction centers of wood and polyethylene. Accordingly, it will promote increasing of the bending strength and water resistance of WPC based on polyethylene.

The objective of this study was to investigate the possibility of WPC production by using secondary polyethylene and conventional flat press method, which is widely used in manufacturing wood-based panels such as particleboards, MDF and OSB. In addition, the effect of different types of wood filler, its content, density of panels and pressing parameters on the physical and mechanical properties of WPC boards was evaluated.

MATERIALS AND METHODS

The following particles were used in this study: secondary polyethylene and wood fillers of various broadleaved and coniferous species as flour (WF), particles (WP), mixture of waste chips and sawdust (WS&SD) at ratio 10:90) with moisture content of 2-3 %. Their fraction analysis is presented in Table 1. The particles are shown in Figure 1. Experiments were carried out in two series, using 12 testing samples for each test.



Fig. 1 Secondary polyethylene and wood fillers.

Components	Screen hole size (mm)						
	-/5	5/4	4/2	2/1	1/0.63	0.63/0.315	0.315/0
Wood particles	4.75	12.2	15.79	40.28	15.67	9.13	2.18
Mixture of waste chips and sawdust (10:90)	1.2	3.6	9.6	17.3	36.7	20.1	11.2
sPE	9.53	3.04	53.14	32.45	1.83	-	-

Tab. 1 Fraction analysis (by % weight) of particles.

Test 1: Effect of wood filler type on the properties of flat-pressed WPC

Three types of wood fillers were chosen for this test: flour, particles, mixture of waste chips and sawdust (10:90). The wood filler to secondary polyethylene ratio was of 40/60. Wood filler and secondary polyethylene in the solid dry state were mixed by hand during 10 minutes. Then the mixture was formed into a mat in an open press-form. The boards with dimensions 250 mm × 230 mm × 8 mm were pressed by pressure of 3.5 MPa, temperature (*T*) of 180 °C and time (τ) of 8.0 min. The used press form enables to produce boards with equal density within the range of \pm 2.5 %. At the end of the press cycle, the board was removed from the press for cooling to the temperature 30–40 °C.

Test 2: Effect of pressing factors on the properties of flat-pressed WPC

Wood particles were chosen after test 1 for the next test. The WPC composition was prepared in the same way as described in Test 1. The pressing factors were: board density $(\rho) - 800$, 900 or 1000 kg/m³; wood particles content (*C*) - 20, 40 or 60 %: pressing temperature (*T*) - 160, 180 or 200 °C; pressing time (τ) - 6.4, 8.0 or 9.6 min.

Bending strength and screw withdrawal resistance of WPC boards were determined according to EN 310: 2003 and EN 320: 1993, respectively. Water resistance of the boards as thickness swelling (TS) and water absorption (WA)) was evaluated according to EN 317: 1993.

RESULTS AND DISCUSSION

The highest values of bending strength and better values of water resistance were observed in WPC made using wood particles. Specifically, the bending strength of WPC made using wood particles was two times higher in comparison with using WC&SD and 2.76 times higher in comparison with using wood flour (Figure 2, a). The screw withdrawal resistance for WPC made using WC&SD and wood flour was respectively 1.16 and 1.31 times lower in comparison with WPC made using wood particles (Figure 2, b). But, it should be noted that the bending strength values of flat-pressed WPC boards with all wood filler types don't meet standards for particleboard (12.5 MPa minimum requirements of EN 312), MDF (23 MPa minimum requirements of EN 622-5) and OSB (20 MPa minimum requirements of EN 300).

Although, higher values of bending strength for extruded WPC boards made using wood flour were observed by other researchers (SIMONSEN *et al.* 2004). This can be explained by the fact that blending of wood particles with melted thermoplastic polymer occurred in the extruder. Under such conditions the thermoplastic polymer is able to cover more specific area of wood particles and to provide greater composite strength. In the case of dry blending the particles of thermoplastic polymer are unable to cover such a large number of surfaces of wood particles. This is also confirmed in work (H'NG *et al.* 2008), where blending of components occurred in the extruder, and pressing in the hot flat press. However, the use of such method of

manufacturing WPC has minor problems, including difficulties in the formation of products with a large area due to the rapid transition of thermoplastic polymer into crystalline state, and constant heating of mold to the temperature above its melting point.

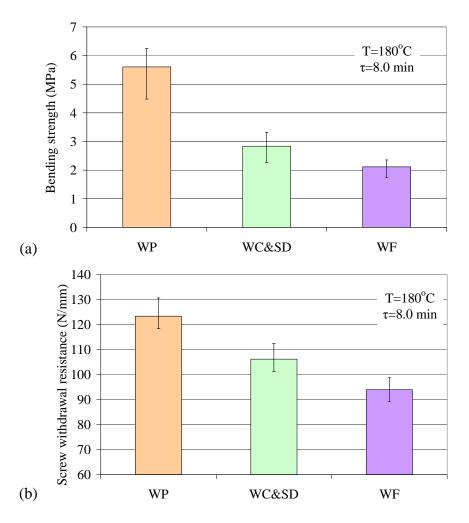


Fig. 2 Effect of wood fillers on the bending strength (a) and screw withdrawal resistance (b) of flatpressed WPC boards.

It should also be noted that the maximum bending strength values during the production of WPC by extrusion are observed for wood flour particles of size 0.2–0.3 mm (STARK 1997) A further increase in particles size leads to a gradual decrease in this parameter. The values of screw withdrawal resistance are similar to those in work (AYRILMIS and JARUSOMBUTI 2011).

Water absorption and thickness swelling values of WPC boards made using WC&SD were 1.56 and 1.4 times higher than those of WPC boards made using wood particles. But the water absorption and thickness swelling of WPC boards made using wood flour were respectively 1.49 and 1.22 times higher than those of WPC boards made using wood particles (Figure 3, a, b). Hereby, better water resistance values were observed in WPC boards made using wood particles. Thickness swelling values of WPC boards for all wood fillers were less than those of particleboard Type P4-P7 (9–16 % maximum requirements of EN 312), MDF (12–17 % maximum requirements of EN 622-5) and OSB (12–25 % maximum requirements of EN 300).

As it is known, wood flour is the most suitable filler for manufacturing WPC boards by extrusion. However, as it can be seen from Figures 2 and 3, it is necessary to apply larger particles for the manufacture of WPC boards in flat presses.

The WPC boards with wood particle to secondary polyethylene ratio of 40/60 had the highest values of bending strength (Figure 4). Similar dependences are noticed at other pressing temperatures and times.

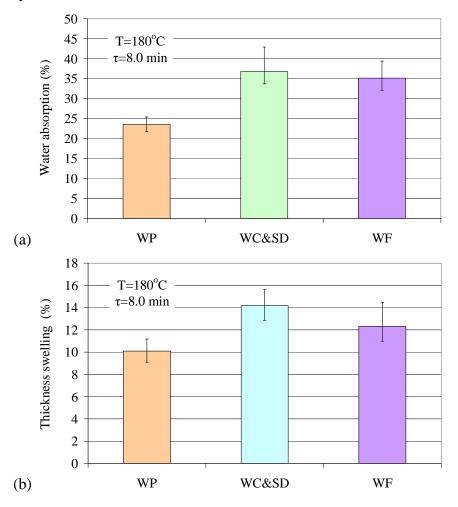


Fig. 3 Effect of wood fillers on the water absorption (a) and thickness swelling (b) of flat-pressed WPC boards.

When content of wood particles isn't high, the thermoplastic zones which are not connected to the wood particles appear. In such case, wood particles perform as modifying additives. More homogeneous wood-polymer mixture with stronger wood-polymer connections is created with increasing content of wood particles. The non-gluing zones were formed with increasing above 40 % content of wood particles, because specific surface area of wood particles increased. This surface area must be covered by a thermoplastic polymer. But under loading these non-gluing zones will be damaged with forming cracks which results in decreasing of bending strength. The same dependencies were observed in the work (AYRILMIS *et al.* 2011). But higher values of bending strength were observed when 50 % content of wood particles and polypropylene was used. This can be explained by the use of polypropylene and maleic anhydride-grafted PP as a matrix that ensures formation of strong wood-polymer connection. Moreover, the rubberwood fibers were used as a filler in that study (AYRILMIS *et al.* 2011).

Polyethylene, having high water resistance, forms water resistance film on the surface of wood particles. This film doesn't allow water to penetrate into capillary and porous structure of wood particles and doesn't allow it to absorb water and to swell. Polyethylene at the content of

80 % covers more specific surface area of wood particles that results in forming of the water resistance framework. Besides, melted thermoplastic polymer fills voids between wood particles which prevents water penetration into material.

Pressing temperature and time also have essential influence on the properties of flatpressed WPC. The increase of pressing temperature from 160 to 200 °C and time from 6.4 to 9.6 min leads to the increased bending strength of WPC (Figure 5). The mat of wood-polymer mixture doesn't heat at the pressing temperature of 160 °C and time of 6.4 min to the temperature necessary for enough fluidity of polyethylene. In such conditions, polyethylene cannot cover the necessary amount of wood particles surfaces for providing high strength. As a result zones with the uncovered wood particles by a thermoplastic polymer appear. These zones will be damaged in the first place. Similar dependences are noticed at the wood particles content of 20 and 60 % and board density of 800 and 1000 kg·m⁻³.

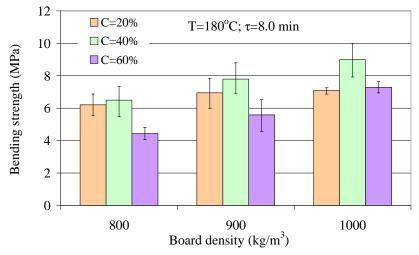


Fig. 4 Effect of wood particles content and board density on the bending strength of flat-pressed WPC boards.

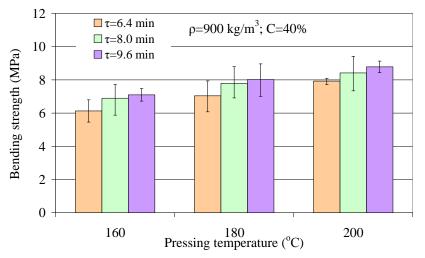


Fig. 5 Effect of pressing temperature on the bending strength of flat-pressed WPC boards with different pressing time.

The water resistance of WPC becomes higher with increasing of the board density (Figure 6, a, b), as a result of closer contact between wood particles and thermoplastic polymer. Water absorption and thickness swelling of WPC reduce with the decrease of wood particles content from 60 to 20 %. These results are similar to the results obtained in the works

(AYRILMIS *et al.* 2011). Similar results also were obtained in the work (AYRILMIS and JARUSOMBUTI 2011). However, water resistance values were significantly higher in that study. In particular, water absorption was lower by about 8.2 % and swelling in thickness was lower at 2.87 % for the density of 800 kg/m³ board and wood particles content of 40 %. Thus, it can be concluded that the use of recycled polyethylene also allows providing high water resistance of WPC. And water resistance of WPC made using secondary polyethylene can compete with water resistance of WPC made using maleic anhydride-grafted PP and PP.

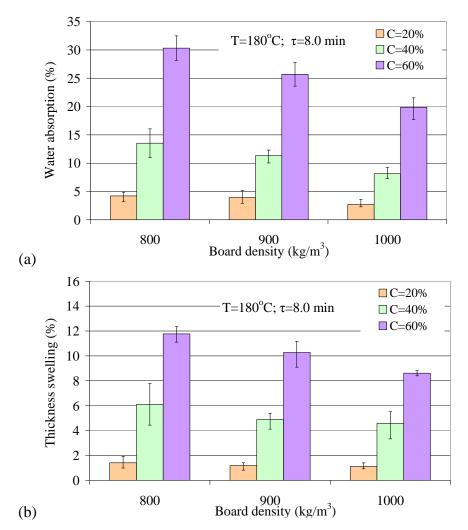


Fig. 6 Effect of wood particles content and board density on the water absorption (a) and thickness swelling (b) of flat-pressed WPC boards.

The increase of pressing temperature and time in the investigated levels considerably influences water absorption and thickness swelling of WPC (Figure 7, a, b). At the pressing temperatures of 180–200 °C and times of 8.0–9.6 min the sPE can easily penetrate into the capillary and porous structure of wood particles. The thermoplastic film created on the surface of wood particles prevents the co-operation between molecules of water and components of wood that reduces thickness swelling and water absorption by wood. Besides, thermoplastic polymer fusion spreads on all volumes of WPC with formation of composites with homogeneous properties.

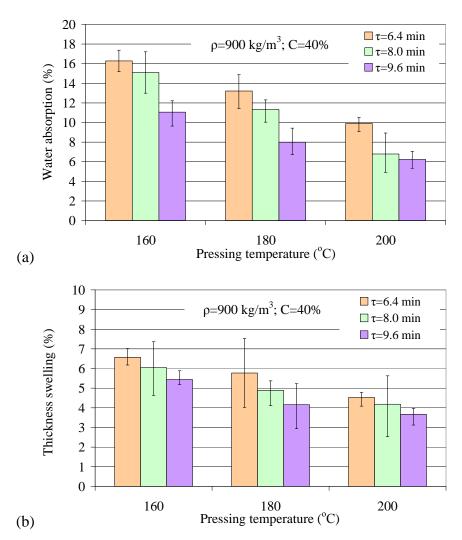


Fig. 7 Effect of pressing temperature on the water absorption (a) and thickness swelling (b) of flatpressed WPC boards with different pressing time.

However, partial destruction of extractives and pentosanes starts at these pressing parameters. This is accompanied by the appearance of characteristic smell and darkening of WPC. Similar dependences of WPC water resistance are noticed at all other levels of investigated factors.

Thus, flat-pressed WPC are characterized by satisfactory properties at about 40 % content of wood particles. The further increase of wood particles content leads to the decline of water resistance and bending strength of WPC.

CONCLUSIONS

Physical and mechanical properties of the flat-pressed WPC boards made using secondary polyethylene were significantly affected by the type of wood fillers. The highest bending strength and screw withdrawal resistance values were noticed in WPC boards made using wood particles. Better water resistance values were also observed in WPC boards made using wood particles.

The properties of the flat-pressed WPC boards were also significantly affected by the wood particle/secondary polyethylene ratio. The WPC boards with wood particle to

polymer ratio 40/60 had the highest values of bending strength. The further increase or decrease of their content results in the decline of the bending strength. Water resistance improves with increasing of secondary polyethylene.

Water resistance and bending strength of WPC boards were also improved with the increase of board density from 800 to $1000 \text{ kg} \cdot \text{m}^{-3}$.

The properties of WPC boards significantly improved with the increase of pressing temperature from 160 to 200 °C and time from 6.4 to 9.6 min. It can be explained by the fact that wood-polymer composition is better heated and this causes lowering viscosity of sPE and better bonding. But partial destruction of wood extractives and some pentosanes begins at the pressing temperature of 180–200 °C and time of 9.6 min. This is accompanied by the appearance of characteristic smell of slightly burnt and darkening of WPC.

Conventional flat-press process for manufacturing WPC boards under laboratory conditions showed that WPC boards made using secondary polyethylene had better water resistance and screw withdrawal resistance than those of the conventional wood-based boards (particleboard, MDF or OSB). However, the values of bending strength for flat-pressed WPC boards were lower than the standard requirements for particleboard, MDF and OSB, and further studies are needed to improve this parameter.

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