

PHYSICAL, MECHANICAL AND THERMAL PROPERTIES OF WOOD/ ZEOLITE/PLASTIC HYBRID COMPOSITES

Alperen Kaymakci^{1,}, Turker Gulec², Seyyed Khalil Hosseinihashemi³, Nadir Ayirmis⁴*

ABSTRACT

Effect of zeolite content on the physical, mechanical and thermal properties of wood plastic composites was investigated in this study. To meet this objective, pine wood flour (0, 10, 20, 30, 40, 50 wt%) with compatibilizing agent, zeolite (0, 10, 20, 30, 40, 50 wt%), and polypropylene were compounded in a twin screw co-rotating extruder. The mass ratio of the wood flour to polypropylene was 50/50 (w/w) in all compounds. Test specimens were produced using injection molding machine from the pellets. Physical and mechanical, and thermal (Thermogravimetry Analysis/Differential Scanning Calorimetry) properties of the wood plastic composites were determined. The water absorption and thickness swelling properties of wood plastic composites improved with increasing zeolite content. The flexural and tensile properties of the wood plastic composites decreased with increasing zeolite content. All the wood plastic composites provided the values of flexural strength (58,4-72,9 MPa) and flexural modulus (2718-5024 MPa) that were well over the requirement by the standard specified in ASTM D 6662. The mass loss rates values of wood plastic composites increased with increasing zeolite contents. The Differential Scanning Calorimetry analysis showed that the melt crystallization enthalpies and degree of crystallization of wood plastic composites decreased with increasing zeolite content. The decrease in the T_c and X_c indicated that zeolite was the poor nucleating agent for the wood plastic composites.

Keywords: *Pinus sylvestris*, swelling, thermoplastic composites, technological properties, thermal stability.

INTRODUCTION

New kinds of biobased materials produce from renewable resources, such as lignocellulosic material are becoming more attractive. The principal reason for this can be related to global environmental problems and the need for a green development in society (Naik *et al.* 2007). Green development can only be probable when production uses renewable materials or materials recycled from wastes. As a consequence of the increasing demand for eco-friendly materials and the high cost of synthetic fibres, new biobased materials containing natural fibres were developed (Kidalova *et al.* 2012, Yorur 2016, Mengelolu and Karakus 2008, Kordkheili *et al.* 2013, Safdari *et al.* 2011).

Wood plastic composites (WPC) define as composite materials containing lignocellulosic (in various forms) and thermoplastic materials are a relatively new family of composite materials. In such

¹Kastamonu University, Forestry Faculty, Department of Wood Mechanics and Technology, Kastamonu, Turkey.

²Artvin Coruh University, Forestry Faculty, Department of Wood Product Chemistry and Technology, Artvin, Turkey.

³Department of Wood Science and Paper Technology, Karaj Branch, Islamic Azad University, Karaj, Iran.

⁴Department of Wood Mechanics and Technology, Forest Faculty, Istanbul University, Turkey.

*Corresponding author: akaymakci@kastamonu.edu.tr

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composites, a natural fiber/filler (such as kenaf fiber, wood flour (WF), hemp, sisal, etc.) is mixed with a thermoplastic (e.g., polyethylene, polypropylene, PVC, etc.) to produce a WPC (Najafi *et al.* 2005, Ayrlmis *et al.* 2013, Yemele *et al.* 2010, Ayrlmis and Kaymakci 2013, Bledzki and Gassan 1999, Demir *et al.* 2006, Valles-Rosales *et al.* 2016b). Generally, WPC formulation consists of over 50% WF, thus its characteristics play a critical role in determining the WPC processing parameters as well as final product properties. To produce WPCs, wood residuals, e.g. sawdust, wood shavings or WF, are mixed with a thermoplastic polymer and melt processed (or thermoformed) into its final shape, either as a continuous profile forming through extrusion or as a three dimensional form through injection molding. During recent decades, WPCs have rapidly increased their market share as a building material (Carus and Gahle 2008). Despite the recent economic downturn, which has leveled off this growth, a continued global market advance is predicted (Anonymus 2011, Bledzki *et al.* 1998). This success may partly be attributed to the fact that WPCs are a competitive alternative to tropical hardwoods and that they are considered to require less maintenance than conventional wood products. Using WPCs, it is also possible to manufacture more complex shapes than with solid wood products, with a raw material yield close to 100%. A growing demand for WPCs has led to continuous efforts to find new resources as alternative to wood. With the increasing population of the world, the sustainable utilization of forest resources has been adversely influenced.

Zeolites are hydrated crystalline aluminosilicates with microporous nanostructures and pore sizes ranging from about 3-15 Å. The structure of zeolites consists of 3-dimensional frameworks made up by SiO₄ and AlO₄ tetrahedra. They possess permanent negative charges in their structural framework, which has the capability of adsorbing or rejecting molecules, thus they are widely used in catalysis, separation, adsorption and ion-exchange (Wang *et al.* 2015). Zeolites can be used to reinforce composites or as agents to improve their mechanical properties and crystallinity behavior of polymer (Chang *et al.* 2013). The addition of zeolites to natural-flour-filled thermoplastic polymer composites is an effective method of reducing their odor and VOC emissions without any degradation of their mechanical and thermal properties (Ki and Kim 2008).

There is a lot of studies on the effects of filler such as WF, wheat straw, nanoclays and carbon nanotube on the physical, mechanical and thermal performance of WPCs. (Khonsari *et al.* 2015, Samariha *et al.* 2015, Aydemir *et al.* 2016, Valles-Rosales *et al.* 2016a, Nafchi *et al.* 2015). But, currently there is little information available concerning on effect of zeolites mining on mechanical, physical, and thermal properties of WPCs. For this reason, this study was to examine effect of zeolite as well as compatibilizer agent (MAPP) used in experimental polypropylene (PP) and pine wood flour (WF) composites on their physical, mechanical and thermal properties. The major weakness is a poor compatibility between wood and plastic. The effect of the zeolite powder on the interfacial bonding between wood flour and polymer matrix was also investigated in this study.

EXPERIMENTAL

Materials

Pine WF (*Pinus sylvestris* L.) with mesh size of 60 was provided by Wood Plastic Decking Factory, Tekirdağ, Turkey. Polypropylene (PP) with a density of 0,9 g/cm³ was purchased from Borealis Incorp in Austria. It has a melt point 170 °C and a melt flow index of 2,5 g/10 min at 230 °C. Zeolite was obtained from Rota Mining Co. İstanbul, Turkey. The compatibilizing agent, maleic anhydride grafted polypropylene (MAPP) (Optim-425) was supplied by Pluss Polymers Pvt. Ltd. Gurgaon, India. It has a density 0,91 g/cm³ and a melt flow index about 120 g/10 min at 190 °C.

WPC preparation

Before manufacturing processes, the WF was dried in a vacuum oven at 103±2 °C for 24-h to a moisture content of 0-1% based on the oven-dry weight of wood. The dried WF was stored in a plastic bag. Then the PP, WF, zeolite, and MAPP were weighed and bagged according to formulations given in Table 1.

Table 1. Compositions of the injection molded WPC formulations.

| Composite type | Composite composition | | | |
|----------------|-----------------------|----------------|-------------------------|------------------------------------|
| | Wood flour (wt %) | Zeolite (%) | Polypropylene (wt %) | Coupling agent (MAPP) (wt %) |
| A | 50 | - | 50 | 3 |
| B | 40 | 10 | 50 | 3 |
| C | 30 | 20 | 50 | 3 |
| D | 20 | 30 | 50 | 3 |
| E | 10 | 40 | 50 | 3 |
| F | - | 50 | 50 | 3 |

The PP, WF, zeolite and MAPP granulates were processed in a 30 mm co-rotating twin-screw extruder with a length-to-diameter (L/D) ratio of 30:1. The barrel temperature of the extruder was controlled at 175-190 °C. The extruded strand passed through a water bath and was subsequently pelletized. The pellets were dried again at 103 ± 2 °C for 2 h before the injection molding. From the compounds which had been granulated, specimens were injection molded by injection molding machine (TSPX 60, China) at molding temperature of 180 °C. The injection pressure was 3-5 MPa. The test specimens were conditioned at 23 °C and a relative humidity of 50% in a climate room until they reached constant weight. This was important because all the specimens should reach equilibrium moisture content prior to testing.

Methods

Physical Tests

Test samples measuring 2 mm x 50 mm x 50 mm were used for the thickness swelling (TS) and water absorption (WA) determination according to ISO 62 procedures. The conditioned specimens were entirely immersed for 1-day, 7-days, and 14-days in a container of water at 23 ± 2 °C. At the end of each immersion time, the samples were taken out from the water and all surface was removed with a clean dry cloth. The specimens were weighed to the nearest 0,01 g and measured to the nearest 0,001 mm immediately. Twelve replicate samples were tested for each wood plastic composite formulation.

Mechanical tests

The flexural and tensile tests were measured according to the ISO 178 and ISO 527, respectively, using a Lloyd universal testing machine (Model LS 100, England). The tests were performed at crosshead speeds of 5 mm/min. Twelve replicate specimen were tested for both flexural and tensile strength measurements.

Thermogravimetry Analysis (TGA) and Differential Scanning Calorimetry (DSC)

The injected pellets were grinded with a Willey mill prior to analysis. Thermo-gravimetric analysis of the samples was done by using the Perkin Elmer STA 6000 thermal analyzer. For the TGA test, the specimens having a weight between 17-20 mg were heated from 50 to 600 °C at a heating rate of 20 °C/min. Additionally, melting and crystallization behavior of the WPNs were studied in a heat-flux type differential scanning calorimeter (DSC, Pelkin Elmer DSC 4000) according to ASTM D3418. The test samples weighing about 9-10 mg in an aluminum crucible were heated up to 250 °C with the heating rate of 10 °C/min and kept at this temperature for 2 min to remove thermal history. Then the samples were cooled down to 0 °C with the cooling rate of 10 °C/min by an electrical cooling device. All heating-cooling runs in melting and crystallization studies were carried out under nitrogen (N₂) atmosphere at a flow rate of 30 ml/min to prevent oxidation of the samples.

Statistical analysis

An analysis of variance, ANOVA, was conducted ($p < 0,05$) to evaluate the effect of the zeolite content on the physical, mechanical and thermal properties of the wood polymer composites. Significant differences among the average values of the wood polymer nanocomposites types were determined using Duncan's multiple range tests.

RESULT AND DISCUSSION

Dimensional Stability

The results of air-dry density, TS and WA performed on the WPC samples are presented in Table 2. The air-dry density of the composites ranged from 1,03 to 1,17 g/cm³. As compared to the wood flour, the TS and WA of the WPCs significantly improved with increasing amount of the zeolite. The integration of WF into the plastic matrix resulted in high TS and WA values due to the hydrophilic nature of WF, as estimated. The specimen types presenting significant differences with further groups according to Duncan's multiple-range tests are shown by superscript letters in Table 2. The lowest TS value was obtained to be 0,06% for the specimens containing 50 wt% zeolite and MAPP after 24 h (1 day) of submersion in water, whereas the highest TS value was obtained to be 0,98% for the samples containing 50% WF and MAPP after 14 day of submersion in water.

Table 2. Physical properties of the WPCs filled with wood flour and zeolite.

| WPC Type ¹ | Water Resistance | | | | | | |
|-----------------------|------------------------------|----------------------|------------------|------------------|------------------------|-------------------|------------------|
| | Density (g/cm ³) | Water Absorption (%) | | | Thickness Swelling (%) | | |
| | | 1-d | 7-d | 14-d | 1-d | 7-d | 14-d |
| A | 1,03 a (0,004) | 0,21 a (0,10) | 0,48 a (0,06) | 0,59 a (0,12) | 0,48 a (0,05) | 0,66 a (0,12) | 0,98 a (0,04) |
| B | 1,07 b (0,005) | 0,16 b (0,04) | 0,43 b (0,06) | 0,55 a (0,08) | 0,43 a (0,13) | 0,60 ab (0,38) | 0,91 b (0,05) |
| C | 1,09 c (0,004) | 0,12 bc (0,07) | 0,33 c (0,02) | 0,41 b (0,04) | 0,34 b (0,06) | 0,47 bc (0,04) | 0,85 c (0,04) |
| D | 1,10 d (0,01) | 0,10 bc (0,03) | 0,26 d (0,07) | 0,37 b (0,05) | 0,29 bc (0,07) | 0,39 cd (0,12) | 0,75 d (0,04) |
| E | 1,17 e (0,006) | 0,08 c (0,02) | 0,19 e (0,04) | 0,26 c (0,07) | 0,26 c (0,05) | 0,32 cd (0,03) | 0,64 e (0,08) |
| F | 1,15 f (0,013) | 0,03 d (0,02) | 0,06 f (0,02) | 0,12 d (0,02) | 0,06 d (0,03) | 0,22 d (0,02) | 0,46 f (0,06) |

¹ See Table 1 for WPC formulation.

The value in parenthesis is the standard deviation, Groups with same letters in column indicate that there is no statistical difference ($p < 0,05$).

The TS percent for the 24-hour water-immersed specimen varied from 0,48% to 0,06% for the WPC specimens. These values increased after 14 days of immersion, varying from 0,98% to 0,46% for the WPC samples. Similar to TS properties, the lowest WA value was obtained to be 0,03% for the samples including 50 wt% zeolite and MAPP after 24 h (1 day) of submersion in water, whereas the highest WA value was obtained to be 0,59% for the samples containing 50% WF and MAPP after 14 day of submersion in water. The WA percent for the 24-hour water-immersed samples varied from 0,21% to 0,03% for the WPC specimens. These values increased after 14 days of immersion, changing from 0,59% to 0,12% for the WPC samples.

Due to the hydrophilic nature of the wood, the moisture absorption is high, which leads to weak interfacial adhesion between the WF and polypropylene, which makes debonding. For improving interfacial bonding between the WF and polymer matrix, the MAPP which was one of the efficient compatibilizer agents in WPC industry was used in the production of the composites (Demir *et al.* 2006). Anhydride part of the MAPP forms ester bonds with wood's hydroxyl groups and the long hydrophobic polymer incorporates into the polymer network. MAPP is therefore a functional modifier. Consequently, the wood fibers and the polymer matrix become bonded together, resulting in enhanced mechanical properties and reduced moisture absorption (Väisänen 2016).

The TS and WA decreased with increasing zeolite content in the polypropylene composites - a trend that is expected for 1 day and 7 and 14 days water immersion tests. With the increase in the zeolite content, there are less water residence spaces, thus less water is absorbed. However, the samples containing a high content of WF have more hydroxyl groups reacted with water molecules, which results higher TS and WA. The lowest TS and WA value were found in the specimens containing 0 wt% zeolite after 1-day submersion in water, whereas the highest TS and WA values were obtained for the samples containing 50 wt% WF after 14-days of submersion in water. The samples produced with lower content of the zeolite had lower TS and WA values.

Mechanical Properties

The significant differences ($p < 0,05$) were found in the values of the flexural strength and modulus of elasticity (MOE). The significant differences between the composite groups are displayed in Table 3.

Table 3. Mechanical properties of the WPCs filled with wood flour and zeolite.

| WPC type ¹ | Mechanical properties | | | |
|-----------------------|----------------------------|--|------------------------|--|
| | Flexural strength (MPa) | Modulus of elasticity in bending (MPa) | Tensile strength (MPa) | Modulus of elasticity in tensile (MPa) |
| A | 70,8 (2,79) a ² | 3200 (743) ad | 41,5 (0,77) a | 1336 (206) a |
| B | 72,9 (3,38) a | 5024 (478) b | 42,1 (0,54) a | 1411 (121) a |
| C | 64,9 (2,82) b | 4372 (1005) c | 34,8 (0,49) b | 1173 (117) b |
| D | 62,2 (2,87) bc | 4061 (254) c | 32,0 (0,57) c | 1121 (163) bc |
| E | 59,5 (0,90) cd | 3602 (457) cd | 31,47 (1,18) cd | 1061 (55) bc |
| F | 58,4 (1,94) d | 2718 (310) a | 30,94 (0,98) d | 1008 (118) c |

¹Table 1 for WPC formulation.

The value in parenthesis is the standard deviation, Groups with same letters in column indicate that there is no statistical difference ($p < 0,05$).

The flexural strength and MOE values of the WPC specimens decreased with increasing zeolite content from 10 to 50% in the composite. For example, the average flexural strength and modulus of elasticity values of the WPCs containing 50 wt% WF (Group A) were found to be 70,8 and 3200 MPa as compared to the WPCs containing 50 wt% zeolite (Group F), which were about 58,4 and 2718 MPa, respectively. However, for polyolefin-based plastic lumber decking boards, ASTM D 6662 standard requires the minimum flexural strength of 6,9 MPa and flexural modulus of 340 MPa. All the composites produced in this study provided flexural strength values (58,4-72,9 MPa) and flexural modulus (2718-5024 MPa) that are well over the requirement by the standard. In a former report, the modulus of elasticity values were obtained to be 5336 MPa for WPCs produced from 47% PP and 50% hardwood WF respectively (Ayrilmis and Kaymakci 2013).

Wood is a lignocellulosic material made up of three major constituents (42-44% cellulose, 27-28% hemicelluloses, and 24-28% lignin). The major percentage of wood is cellulose. The aligned fibril structure of the cellulose, along with strong hydrogen bonds, has a high stiffness; thus, the addition of WF can increase the stiffness of thermoplastic composites (Walker 2006, Uysal and Yorur 2013). As shown in Table 3, the flexural strength and modulus of elasticity values of the composites containing WF were significantly higher than those in the composites containing zeolite. This could be explained by the improved interfacial adhesion between polypropylene and the WF due to the compatibilizing

agent (MAPP). According to Bledzki and Gassan (1999) an increase in the wood plastic composite strength could be related with higher cellulose and lignin contents. This provide better dispersion and adhesion to the polymer matrix. The lower flexural strength and modulus of elasticity values of the composites containing high zeolite contents were also attributed to the lower fibrous zones in the zeolites. The observed decrease in flexural strength and modulus of elasticity indicated that the zeolites could not act as a reinforcing agent inside the WPC matrix. These findings propose that the desirable content of zeolites was 10% of the production of WPCs (Group B). Similar results were reported by Wang *et al.* (2015). They stated that extreme zeolite content could form agglomerates inside the WPC matrix and act as micro crack initiator, thus playing an adverse role in the mechanical performance.

The findings of the tensile properties of the samples with WF and zeolite are given in Table 3. Adding zeolite to the polypropylene significantly decreased the tensile properties of the WPC samples. The tensile strength and tensile modulus values of the WPC specimens decreased with increasing zeolite content from 10 to 50 wt% in the WPC. For example, the average tensile strength and tensile modulus values of the WPCs containing 50 wt% WF (Group A) were 41,5 MPa and 1336 MPa as compared to composites containing 50% zeolite (Group F), which were about 30,94 MPa and 1008 MPa, respectively. The decrease in the tensile properties was mainly attributed to the weak interfacial adhesion between the WF, zeolite and the polypropylene matrix, as reported in previous paper Wang *et al.* (2015). In all the WPC groups, the WPCs with the zeolite had lower tensile strength and tensile modulus (except composite group B). Ashori and Nourbakhsh (2009) reported that the strength of the wood plastic composites was mainly influenced by the interfacial adhesion between wood and polymer matrix. Better interfacial adhesion between particles and matrix results into more restriction to deformation capacity of the matrix in the elastic zone and increased strength and modulus (Ashori and Nourbakhsh 2009).

TGA and DSC analysis

TGA test curves of WPCs filled with WF and zeolite samples are shown in Figures 1 and 2 (Table 4). From the figures, it was observed that zeolite could change the thermal stability of the WPCs. As can be clearly seen from Figures 1 and 2, the thermal degradation of all WPCs was comprised of two stages. All the WPC groups were moderately stable up to the temperature above 200 °C, which was higher than the process temperatures for the twin-screw extruder and injection molding (Figure 1). The first peaks were observed around 230 °C for WPCs filled with the WF and zeolite while the second peaks were around 420 °C for wood plastic composites. As shown in Figure 1, the highest weight loss was found in the specimens containing 50 wt% WF while the lowest weight loss was found in the specimens containing 50 wt% zeolite.

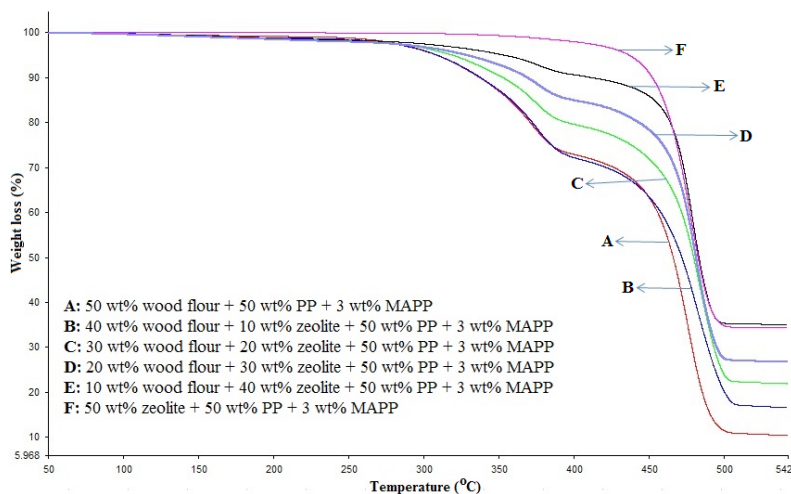


Figure 1. TGA results of wood plastic composites.

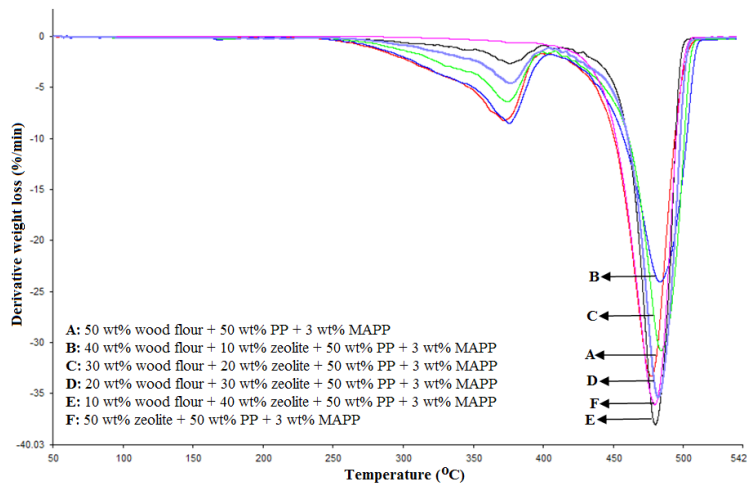


Figure 2. Derivative weight losses result of wood plastic composites.

Table 4. The results of Thermogravimetric analysis (TGA).

| WPC code ¹ | 1st region | | | 2nd region | | | Residue at 520 °C (%) |
|-----------------------|---------------|-----------|-----------------|---------------|-----------|-----------------|-----------------------|
| | Range (°C) | Peak (°C) | Weight loss (%) | Range (°C) | Peak (°C) | Weight loss (%) | |
| A | 238,15-395,10 | 318,48 | 36,669 | 405,31-511,22 | 452,86 | 33,483 | 10,736 |
| B | 233,00-402,93 | 319,29 | 41,854 | 408,46-515,09 | 458,46 | 32,900 | 16,976 |
| C | 233,34-400,02 | 321,06 | 30,582 | 409,33-512,56 | 462,35 | 41,028 | 22,214 |
| D | 241,91-401,10 | 330,26 | 19,866 | 410,86-510,19 | 462,20 | 43,438 | 26,914 |
| E | 240,51-398,19 | 328,23 | 11,530 | 404,11-504,30 | 461,38 | 56,726 | 35,179 |
| F | - | - | - | 385,49-505,91 | 452,57 | 82,222 | 34,442 |

¹ See Table 1 for WPC formulation.

The crystallization peak temperature (T_c) of the WPCs with the WF was higher than that of those with zeolite, indicating that the WF which was a conductive filler could act as nucleating agents (Table 5).

Table 5. The results of differential scanning calorimetry (DSC) analysis.

| WPC code ¹ | $T_{m,1}$ ^b (°C) | $T_{c,o}$ ^c (°C) | $T_{c,p}$ ^d (°C) | ΔH_c ^e (J/g) | $T_{m,II}$ ^f | ΔH_{II} ^g (J/g) | X_c ^h (%) |
|-----------------------|-----------------------------|-----------------------------|-----------------------------|---------------------------------|-------------------------|------------------------------------|------------------------|
| A | 165,2 | 131,0 | 127,5 | 48,9 | 166,2 | 33,2 | 31,8 |
| B | 165,8 | 130,0 | 126,3 | 55,6 | 165,6 | 42,7 | 40,8 |
| C | 165,2 | 129,3 | 125,9 | 41,7 | 165,5 | 32,5 | 31,1 |
| D | 165,6 | 126,5 | 127,8 | 40,4 | 163,7 | 32,2 | 30,8 |
| E | 165,3 | 131,1 | 127,3 | 39,1 | 166,0 | 31,1 | 30,1 |
| F | 165,5 | 127,7 | 124,4 | 32,5 | 164,5 | 22,5 | 21,5 |

¹ See Table 1 for WPC formulation.

^b Peak temperature of first melting.

^c Onset temperature of crystallization.

^d Peak temperature of crystallization.

^e Enthalpy of crystallization.

^f Peak temperature of second melting.

^g Enthalpy of second melting.

^h Degree of crystallization calculated with the enthalpy of second melting.

Higher peak T_c was due to faster crystallization of polymer chains upon cooling. In general, the polymer molecular chains can crystallize by themselves thorough a self- nucleation effect or by introducing a nucleating agent (Cheng 1985, Demir *et al.* 2005, Mohanty *et al.* 2006, Doan *et al.* 2007). The differences in the T_c values of WPCs were mainly due to use of lignocellulosic filler, coupling agent or zeolites.

The degree of crystallinity decreased increasingly from 20 to 50 wt% zeolite content, the highest crystallinity value being recorded for WPCs at 50 wt% WF, 10 wt% zeolite and 3 wt% MAPP (40,8 %). This may be due to the fact that WF (40 wt%) and zeolite (wt% 10) introduced in the PP matrix can serve as a nucleating agent. The existence of filler can have an effect on the crystallinity behavior of the plastic composites. Accordingly, in this study, the effect of the zeolite content on the crystallization degree of the WPC samples was significant.

The second melting enthalpy of the control group (Group A) was found to be 33,2 J/g. Except the group B (50wt% PP+ 50wt% WF+ 10wt% zeolite+ 3wt% MAPP) the enthalpy of second melting of the samples decreased with increasing content of the zeolite flour. The second melting enthalpy of the WPCs decreased from 32,5 to 22,5 J/g as the zeolite content increased from 20 to 50 wt%. This result indicated that thermal stability of the wood/polypropylene composites increased with increasing the zeolite content. The zeolite absorbed more heat energy in the melting of the plastic composites.

CONCLUSIONS

The results of the present study revealed that water resistance of the WPCs improved with increasing zeolite content while the flexural and tensile properties of WPCs decreased. The lower flexural and tensile properties might be due to poor dispersion of zeolite in the polypropylene matrix. This is an evidence of poor adhesion and weak stress transfer efficiency.

The thermal stability of the wood/polypropylene composites increased with increasing the zeolite content. It should be noted that WPCs having a higher amount of the zeolite had higher residues at 500 °C due to the presence of silicates in the surface region of the zeolites. The DSC analysis showed that the melt crystallization enthalpies and degree of crystallization of WPCs decreased with increasing zeolite content. The decrease in the T_c and X_c indicated that zeolite was the poor nucleating agent for the WPCs. Based on the findings obtained from the present study, a 40/10/50/3 formulation of the WF/zeolite/polypropylene/MAPP can be used in manufacturing WPC with satisfactory service results.

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