Effect of Wood Particle Size on Selected Properties of Neat and Recycled Wood Polypropylene Composites

Vedat Çavuş a,*, and Fatih Mengeloğlu b

Neat polypropylene (PP)- and post-industrial recycled polypropylene (rPP)-based wood-plastic composites (WPC) were manufactured using 40% mahogany wood flour (WF). The effect of particle size (0.074 to 0.149 mm, 0.177 to 0.250 mm, and 0.400 to 0.841 mm) on the selected properties of PP and rPP composites was studied. The influence of 3% maleic anhydride grafted polypropylene (MAPP) presence in the formulation was also evaluated. Test specimens were manufactured using a combination of extrusion and injection molding processes. The density and mechanical properties, such as flexural strength, flexural modulus, tensile strength, tensile modulus, elongation at break, hardness and impact strength values were determined. Morphology of the manufactured composites was also studied using scanning electron microscopy (SEM) analysis. Results showed that the particle size, polypropylene type (neat or recycled), and presence of MAPP had important effects on WPC's properties. Density, flexural modulus, tensile modulus, and impact strength values increased with decreased particle size regardless of the presence of MAPP. Flexural strength values increased with decreased particle size without MAPP. Regardless of particle size, addition of MAPP in composites provided higher flexural strength, flexural modulus, tensile strength, and tensile modulus values but lower elongation at break values compared to composites without MAPP.

Keywords: Particle size; Mechanical properties; Density; Recycled polypropylene; Neat polypropylene; MAPP

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INTRODUCTION

According to the European Committee for Standardization, wood-plastic composites (WPCs) are materials or products consisting of one or more lignocellulosic fibres, flours, and one or a mixture of thermoplastic resins, such as polypropylene (PP), polyethylene (PE), or polyvinyl chloride (PVC). Lignocellulosic fillers and polymeric matrices may come from different sources such as wood waste, unused natural resources, and neat or recycled thermoplastics, as per the requirements of CEN EN 15534-1(2014) (Moreno and Saron 2017). WPCs are widely used in floorboards, house roofs, doors, and window frames (Wechsler and Hızıroğlu 2007). The usage of lignocellulosic fibers or flours in WPCs have some advantages, such as coming from bio-based resources, being inexpensive, wide availability, recyclability, biodegradability, low density, flexibility, water resistance, providing high filling levels of lignocellulosic material, and various specific (strength, stiffness, and resistance to wear) properties (Youssef et al. 2019). The use of recycled polymers instead of neat polymers for industrial purposes is one of the most promising techniques for lowering the environmental impact and the expenses associated
with the scrapping of components (Galve et al. 2019; Momanyi et al. 2019). The production of WPCs can be one of the approaches used to achieve sustainable consumption and production using plastic waste as a resource. Recycled plastic (rPP) could be a promising raw material for WPC manufacturing and a proper way to dispose of plastic wastes (Gulitah and Liew 2019). The source of the recycled polymer can affect the final composite properties. Post-industrial (PI, regrinding of production waste) recycled polymer provides more controllable and stable properties compared to the post-consumer (PC, regrinding of used consumer products) recycled polymer, since the used consumer products are coming from unknown sources (Hubo et al. 2015).

The performances of WPCs can be affected by the amount, chemical composition (cellulose, lignin, and extractive contents), particle geometry, surface characteristics, and particle size (PS) of wood flour (WF). In addition, interfacial properties between the wood and polymer matrix as well as polymer structure, molecular weight, and additives used influence the physical and mechanical properties of the WPC (Bledzki et al. 1998; Shebani et al. 2009; Izekor et al. 2013). Through using various additives in WPC formulations, the physical, mechanical, and chemical properties as well as the expected lifetime of the WPC can be improved, and their application areas can be expanded (Mengeloğlu and Çavuş 2019). Several studies were conducted, and different methods and chemicals were utilized to improve the compatibility of lignocellulosic filler and PP matrix (Mengeloğlu and Çavuş 2019). Among them, MAPP was the most preferred one, since MAPP can directly be mixed with polymer and filler during processing. The optimum amount of MAPP required for PP based lignocellulosic filler has been intensively studied. It was reported by Daghigh et al. (2018), Huang et al. (2018), Mutjé et al. (2006) and Pimenta et al. (2008) that the optimum amount was 2%, 3 to 5%, 4%, and 6%, respectively. The majority of the studies on lignocellulosic material filled PP composites, on the other hand, have utilized 3% MAPP coupling agent to improve the compatibility between polymer matrix and filler (Keener et al. 2004; Zampaloni et al. 2007; Huang et al. 2018b).

The particle size (PS) is generally used to characterize the shape of WF used in WPC (Chaudemanche et al. 2018). The major advantages of the WF are its low density, low cost, high strength, renewability, biodegradability, and wide availability. In contrast, the incompatibility of hydrophilic filler and hydrophobic polymer matrix is the main disadvantage of WF utilization in WPC manufacturing (Habibi et al. 2008; Gallagher and McDonald 2013; Poletto 2017). Mahogany (Swietenia mahagoni) is widely grown in South America, in Brazil, Bolivia, Peru, and up through Central America to Mexico (Mejía et al. 2008). It has an air-dry density of 0.60 g/cm³. It is a reasonably durable wood. It is easy to work with when using hand tools, finishes to a smooth surface, and has good gluing and nailing properties. Mahogany wood has attractive surfaces and is particularly valued for high-class furniture and cabinetry work. It has also been used in interior paneling, joinery work, turnery, plywood, woodwork, such as models and patterns, instrument cases, clocks, printer’s blocks, parts of musical instruments, and heavy construction work (Pennington 2002; Mejía et al. 2008; Langbour et al. 2011). While producing timbers with a desired shape and size, wood wastes are also generated. The amount of waste generated depends on the number of operations performed and the thickness of the cutter.

This study investigates the effect of wood flour particle size on the selected properties of neat and recycled PP-based composites. The effect of MAPP presence in composites is also evaluated.
EXPERIMENTAL

Materials
Mahogany wood particles were supplied by a Yacht workshop in İzmir city (Turkey). They were granulated into flour form using a Wiley mill (Altundal, Kahramanmaraş, Turkey) and classified into three particle size groups (from smaller to bigger particle sizes): particles that passed through a 0.400 mm screen and stayed on a 0.841 mm screen, those that passed through a 0.177 mm screen and stayed on an 0.250 mm screen, and those that passed through a 0.074 mm screen and stayed on a 0.149 mm screen. The microscopic (Leica, Apo 8 stereo microscope) views of WFs are presented in Fig. 1 as 0.074 to 0.149 mm (A), 0.177 to 0.250 mm (B), and 0.400 to 0.841 mm (C), respectively. The PP (MH 418) was obtained from Petkim Petrochemical Co. (melting point: 163 °C, melt flow index (MFI): 4.5 g/10 min, and density: 0.905 g/cm³). Post-industrial (PI) waste pipes were reground into pellets and donated by Egeplast in İzmir City, Turkey. The MAPP (Licomont AR 504 by Clariant, Berlin, Germany) was used as a coupling agent (density: 0.91 g/cm³, softening point: 156 °C). Paraffin wax (K.130.1000) was used as a lubricant (density: 0.93 g/cm³, softening point: 56 to 58 °C).

Composite Manufacturing and Testing
The manufacturing recipe of the study is given in Table 1. The mahogany WF was first desiccated in an oven for 24 h at 103 °C (± 2 °C). The moisture content of WF was reduced to below 1%. Later, depending on the recipe used, the required amount WF, PP or rPP, lubricant, and MAPP were dry-mixed in a high-intensity mixer for 5s (900 to 1000 rpm) to produce a homogeneous blend. These blends were compounded using a single-screw extruder (TTB 30; Teknomatik, İstanbul, Turkey) at 40 rpm screw speed. The temperature ranged from 170 to 200 °C from barrel to die. Extruded specimens were cooled in a water pool (23 °C ± 2) and then pelletized with a grinding machine. The pellets were desiccated in the oven at 103 °C (± 2) for 24 h. The moisture content of dried wood-plastic pellets was below 1% before the injection molding. The injection molding machine (HAIDAHDX–88, Ningbo Haida Plastic Machinery Co., Ltd., Ningbo, China) was used for manufacturing test specimens. The temperature of injection molding machine varied from 180 to 200 °C (from feed zone to die). The injection pressure, injection speed, and cooling speed were 5 to 6 MPa, 80 mm/s, and 30 s, respectively.

Fig. 1. Microscopic view of WF; A: 0.074 to 0.149 mm, B: 0.177 to 0.250 mm, C: 0.400 to 0.841 mm
Prior to testing, specimens were conditioned at a relative humidity of 65 ± 2% and temperature of 23 ± 2 °C for a week. The density was determined by a water displacement technique according to the ASTM D792 (2007) standard. Flexural strength (FS), flexural modulus (FM), tensile strength (TS), tensile modulus (TM), elongation at break (EatB), hardness (H), and impact (IS) strength (notched) values were determined according to ASTM D790 (2003), ASTM D638 (2001), ASTM D256 (2000), ASTM D2240 (2010), respectively. The dimensions (length x width x thickness) of the test specimens for density, flexural properties, tensile properties, impact strength, and hardness were 13 mm x 13 mm x 4 mm, 165 mm x 13 mm x 4 mm, 165 mm x 13 mm (narrow section) x 4 mm (Dog Bone shape), 65 mm x 13 mm x 4 mm, respectively. FS, FM, TS, TM and EatB testing were implemented on Zwick 10 KN (Ulm, Germany), while a HIT5, 5P (Zwick) was used for IS testing on notched specimens. Notches were added by a RayRan™ Polytest notching cutter (London, UK). Design-Expert® version 7.0.3 statistical software (Minneapolis, USA) was used for statistical analysis. Morphological properties of test specimens were observed using scanning electron microscopy (SEM) (EVO LS10; Zeiss, Jena, Germany). Test specimens for SEM analysis were dipped into liquid nitrogen for 5 min and then broken in half with hammer for a clear fractured surface. The specimens were placed on a specimen holder and sputtered with gold (Sputter Coater 108Auto; Cressington, London, England) to prevent charge accumulation of the electron absorbed by the specimens with 10 mA in 120 s.

RESULTS AND DISCUSSIONS

In this study, the effect of particle size (PS: 0.074 to 0.149 mm; 0.177 to 0.250 mm; 0.400 to 0.841 mm), polymer type (PT: PP, or rPP), and the presence of MAPP (0% or 3%) on the selected properties of WPCs was investigated. Density, FS, FM, TS, TM, (EatB), HS, IS and morphology of the manufactured composites were determined. The values obtained from the test specimens are presented in Table 2.
Density is an important characteristic for WPCs. The densities obtained from the test specimens of PP and rPP-based WPCs were in the range of 1.00 to 1.05 g/cm³. All composites provided higher densities regardless of the presence of PS and MAPP. It is believed that higher cell wall density of lignocellulosic materials (~ 1.5 g/cm³) increased the final composite densities when incorporated in the polymer mixture (Stokke et al. 2014; Karakuş and Mengeloğlu 2016; Mengeloğlu and Çavuş 2019). The interaction graphs of the density are presented in Fig. 2a and 2b. The letters a and b denote composites with no MAPP and 3% MAPP coupling agent, respectively. Statistical analysis showed that PS and PT had a significant effect on the density values of WPCs (P = 0.0098 and P = 0.0002). In contrast, overall the presence of MAPP in WPC did not have a significant influence on density (P = 0.2705). However, statistical analysis showed significant interaction between PS and MAPP (P = 0.0135) and PT and MAPP (P < 0.0001). This means the dependence of MAPP on density was different for PT and PS. The MAPP was more effective in PP-based WPCs compared to rPP-based WPCs. Similarly, MAPP caused an increase in density while PS was low. Higher densities with the use of small particle size filler were also reported by others (Behazin et al. 2017; Chaudemanche et al. 2018).

### Table 2. Properties of Test Specimens

<table>
<thead>
<tr>
<th>ID</th>
<th>Density (g/cm³)</th>
<th>FS (MPa)</th>
<th>FM (MPa)</th>
<th>TS (MPa)</th>
<th>TM (MPa)</th>
<th>EatB (%)</th>
<th>H (Shore D)</th>
<th>IS (kJ/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP1-0</td>
<td>1.03 (0.01)</td>
<td>34.45 (0.87)</td>
<td>1910.01 (71.57)</td>
<td>18.82 (0.52)**</td>
<td>753.99 (10.14)</td>
<td>5.41 (0.42)</td>
<td>71.94 (0.92)</td>
<td>1.78 (0.10)</td>
</tr>
<tr>
<td>PP2-0</td>
<td>1.02 (0.02)</td>
<td>33.80 (0.83)</td>
<td>1992.54 (95.68)</td>
<td>18.67 (0.78)</td>
<td>783.47 (21.87)</td>
<td>4.78 (0.55)</td>
<td>73.04 (1.14)</td>
<td>1.56 (0.10)</td>
</tr>
<tr>
<td>PP3-0</td>
<td>1.00 (0.01)</td>
<td>37.32 (0.59)</td>
<td>2178.12 (88.73)</td>
<td>19.48 (0.57)</td>
<td>834.51 (21.89)</td>
<td>4.40 (0.73)</td>
<td>74.76 (0.38)</td>
<td>1.80 (0.33)</td>
</tr>
<tr>
<td>PP1-3</td>
<td>1.05 (0.01)</td>
<td>41.74 (1.01)</td>
<td>2146.12 (86.86)</td>
<td>24.25 (0.62)</td>
<td>826.04 (125.79)</td>
<td>4.56 (3.20)</td>
<td>74.72 (0.96)</td>
<td>1.62 (0.31)</td>
</tr>
<tr>
<td>PP2-3</td>
<td>1.04 (0.01)</td>
<td>41.81 (0.33)</td>
<td>2271.75 (36.98)</td>
<td>24.50 (1.49)</td>
<td>826.21 (68.1)</td>
<td>4.59 (0.24)</td>
<td>74.10 (0.79)</td>
<td>1.70 (0.06)</td>
</tr>
<tr>
<td>PP3-3</td>
<td>1.03 (0.01)</td>
<td>41.86 (0.66)</td>
<td>2343.93 (33.70)</td>
<td>24.03 (0.34)</td>
<td>825.21 (38.94)</td>
<td>5.64 (0.34)</td>
<td>75.12 (0.50)</td>
<td>2.02 (0.15)</td>
</tr>
<tr>
<td>rPP1-0</td>
<td>1.04 (0.00)</td>
<td>34.42 (0.47)</td>
<td>1927.06 (22.30)</td>
<td>18.97 (0.31)</td>
<td>791.28 (17.87)</td>
<td>6.56 (0.41)</td>
<td>72.54 (0.48)</td>
<td>1.60 (0.09)</td>
</tr>
<tr>
<td>rPP2-0</td>
<td>1.01 (0.02)</td>
<td>34.32 (1.38)</td>
<td>1997.30 (125.17)</td>
<td>19.04 (0.71)</td>
<td>802.33 (34.86)</td>
<td>5.14 (0.84)</td>
<td>73.82 (2.06)</td>
<td>1.85 (0.05)</td>
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<tr>
<td>rPP3-0</td>
<td>1.04 (0.00)</td>
<td>36.48 (0.44)</td>
<td>2141.91 (38.44)</td>
<td>20.26 (0.35)</td>
<td>800.23 (11.97)</td>
<td>4.96 (0.55)</td>
<td>73.38 (0.67)</td>
<td>1.96 (0.05)</td>
</tr>
<tr>
<td>rPP1-3</td>
<td>1.03 (0.01)</td>
<td>40.47 (1.02)</td>
<td>2126.42 (82.25)</td>
<td>22.68 (0.17)</td>
<td>858.67 (32.95)</td>
<td>3.89 (0.22)</td>
<td>75.18 (1.29)</td>
<td>1.69 (0.86)</td>
</tr>
<tr>
<td>rPP2-3</td>
<td>1.02 (0.01)</td>
<td>40.17 (0.68)</td>
<td>2205.59 (47.68)</td>
<td>23.06 (0.27)</td>
<td>865.36 (23.44)</td>
<td>4.08 (0.18)</td>
<td>74.08 (1.71)</td>
<td>1.76 (0.14)</td>
</tr>
<tr>
<td>rPP3-3</td>
<td>1.03 (0.02)</td>
<td>39.83 (0.68)</td>
<td>2217.72 (83.41)</td>
<td>23.47 (0.33)</td>
<td>876.03 (45.06)</td>
<td>4.41 (0.38)</td>
<td>74.62 (2.08)</td>
<td>1.59 (0.17)</td>
</tr>
</tbody>
</table>

*: average values, **: standard deviation
Fig. 2. Interaction graphs of density; (a): PS+MAPP-0, (b): PS+MAPP-3

The FS and FM are important mechanical properties affecting WPCs application areas and performances. The interaction graphs of the FS are presented in Figs. 3a and 3b. The FS values obtained from the test specimens of WPCs were in the range of 33.80 to 41.86 MPa. Statistical analysis showed that all factors investigated (the presence of PS, PT, and MAPP) had statistically significant effects on the FS values of the manufactured WPCs. Based on the results, regardless of PT, composites having particle size of 0.400 to 0.841 mm provided the highest FS values among composites having no MAPP in the formulations. However, when MAPP was present in the formulation, all three PS types provided similar FS values. This result can be explained by the improved adhesion strength between filler and matrix surfaces (Stark and Rowlands 2003; Rude and Laborie 2008; Gallagher and McDonald 2013). It should also be noted that PP composites provided better overall FS values compared to rPP composites, especially when MAPP was present in the formulations. This might be caused by the chain length reduction of the polymer during the recycling process. Reduction of molecular weight during the recycling process was also reported by Mantia and Gardette (2002).

Fig. 3. Interaction graphs of Flexural strength; (a): PS+MAPP-0, (b): PS+MAPP-3
The interaction graphs of the FM of the composites without and with MAPP are presented in Figs. 4a and 4b, respectively. The FM values of the manufactured WPCs were in the range of 1910 to 2344 MPa, respectively. Statistical analysis showed that the PT did not have a significant effect on FM values (P > 0.05). However, both the PS and MAPP presence had a statistically significant effect on FM value of the manufactured WPCs (P < 0.0001). The FM values steadily increased with increased PS. Addition of MAPP in the formulations resulted in an almost 10% improvement on FM values. It is believed that this result was achieved by the improvement of fiber-matrix interfacial bonding between the polymer matrix and filler with MAPP (Kim et al. 1994; Shesan et al. 2019).

![Interaction graphs of flexural modulus](image)

*Fig. 4. Interaction graphs of flexural modulus; (a): PS+MAPP-0, (b): PS+MAPP-3*

The interaction graphs of the TS, TM and EatB are presented in Fig. 5a-b, Fig. 6a-b, and Fig. 7a-b respectively. In all three figures, (a) denotes the sample with no MAPP and (b) shows samples with MAPP in the formulations. The TS values of the manufactured WPCs were in the range of 18.7 to 24.5 MPa. Regardless of particle sizes, WPCs produced with MAPP provided higher TS values than those without MAPP due to the improved adhesion between hydrophilic filler and hydrophobic polymer. In addition, the highest TS values were provided by the samples produced with higher PS (0.400 to 0.841 mm). Once again, with the addition of MAPP, composites produced with different particle sizes provided similar TS values. The MAPP usage improved the TS values regardless of PS and PT.

For TM values of manufactured WPCs, they were in the range of 754 to 876 MPa. Addition of WF with higher cell wall density in the polymer matrix usually increased the modulus values of the resulting composites. This can be explained by the rule of mixtures (Matuana et al. 1998). Moreover, rPP-based composites provided higher TM values compared to PP-based composites. It was reported that polymer chain length is reduced and crystallinity is increased during the recycling process (Mantia and Gardette 2002). It should be further investigated which (chain length or crystallinity) factor has more impact on which properties. There was some increase on TM values with increasing PS. Other studies reported a steady increase in TM with increasing PS (Zaini 1996; Stark and Berger 1997; Stark and Rowlands 2003; Bouafif et al. 2009).
Fig. 5. Interaction graphs of tensile strength; (a): PS+MAPP-0, (b): PS+MAPP-3

Fig. 6. Interaction graphs of tensile modulus; (a): PS+MAPP-0, (b): PS+MAPP-3

Fig. 7. Interaction graphs of elongations at break; (a): PS+MAPP-0, (b): PS+MAPP-3
Another property determined was EatB. Manufactured WPCs had EatB values in the range of 3.89 to 6.56%. The PT had a significant effect on EatB values. EatB values of rPP were almost 80% higher than the values for PP. However, in composite samples there was not much of a difference. With the WF usage, the EatB values were dramatically reduced to 5 to 7%. Test specimens without MAPP provided higher EatB values. Regardless of PT, the highest EatB values were obtained when 0.074 to 0.149 mm PS was used. In addition, similar EatB results were observed for PP- and rPP-based composites if MAPP was present in the formulation.

The interaction graphs of the HS are presented in Figs. 8a and 8b. The figures show the composite without MAPP and with MAPP, respectively. The average HS values obtained for test specimens were in the range of 71.9 to 75.2 Shore D values, respectively.

The interaction graphs of the IS are presented in Fig. 9a and 9b respectively. The average IS values obtained for test specimens were in the range of 1.56 to 2.02 kJ/m². This IS usually reduces with increasing brittleness of the composite material (Mengeloglu and Karakus 2008). Overall, rPP composites provided higher IS values compared to PP-based composites. However when MAPP was utilized in composites, this difference of IS values
between rPP- and PP-based composites was reduced. Similar results were reported by others (Bourmaud et al. 2011; Pickering et al. 2016; Abdullah and Aslan 2019; Hubbe and Grigsby 2020).

The morphology of the test specimens is presented in Fig. 10. Parts (A), (C), (E), (G), (I), and (K) of the figure show SEM images of the PP and rPP samples without MAPP, while parts (B), (D), (F), (H), (J), and (L) of the figure show SEM images of the rPP samples with MAPP in the formulations. The PS of the fillers is denoted under the images. For the ease of visual comparison all images were taken using the same magnification factor. Particle size differences can clearly be seen on images. The use of MAPP provided some improvement in the particle distribution in the polymer matrix. It should also be noted that the number of standing particles and holes left by pulling out particles was reduced when MAPP was present in the formulations.

Higher mechanical properties with MAPP usage have also been reported and believed that MAPP works to minimize the creation of microcracks between the dissimilar polar wood filler and non-polar PP matrix (Myers et al. 1991; Clemons 2010; Rodríguez-Llamazares et al. 2011; López et al. 2012; Clemons et al. 2013; Tisserat et al. 2014; Kusumoto et al. 2016).
**CONCLUSIONS**

In this study, neat polypropylene (PP-) and recycled polypropylene (rPP)-based wood-polymer composites (WPCs) were successfully produced using three different particle sizes of mahogany wood flour (WF). The composites were manufactured with both 0% and 3% maleic anhydride polypropylene (MAPP) coupling agent. The following conclusions can be drawn:

1. WPCs produced with post-industrial waste recycled polymer (rPP) provided excellent properties which is comparable to the ones produced with neat PP.

2. MAPP utilization significantly improved the properties of both PP- and rPP-based WPCs. This effect was more pronounced in PP-based composites compared to rPP-based ones.
Regardless of PP type, utilization of higher particle size corresponded to an increase in the tensile strength and flexural strength values but a reduction in EatB values of composites without MAPP utilization.

MAPP presence in composites minimized the effect of particle size on the flexural and tensile properties of WPCs.

SEM images of composites having MAPP confirmed that improved adhesion between the WF and the polymer matrix and better distribution of WF were present.

Hardness values of the all manufactured composites were in the “extra hard material” class.

ACKNOWLEDGMENTS

This research was supported by the Scientific Research Project Fund of İzmir Katip Çelebi University (Project Number: 2019-GAP-ORMF-0005). Authors would like to thank Graduate Students working at the Wood Composite Testing Laboratory and Wood Composite Manufacturing Laboratory of the Kahramanmaraş Sutcu Imam University.

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Article submitted: December 19, 2019; Peer review completed: February 29, 2020; Revised version received and accepted: March 16, 2020; Published: March 26, 2020. DOI: 10.15376/biores.15.2.3427-3442