Selected Properties of Mahogany Wood Flour Filled Polypropylene Composites: The Effect of Maleic Anhydride-Grafted Polypropylene (MAPP)

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The aim of this study was to produce mahogany (*Swietenia macrophylla*) wood flour filled polypropylene (both recycled and virgin) composites and to determine the effects of maleic anhydride-grafted polypropylene (MAPP) on the interfacial compatibility, density, and other mechanical properties of the resulting composites. Approximately 40 wt% of mahogany wood flour, and 60 wt% of polypropylene, 3 wt% of MAPP, 1.5 wt% of zinc stearate, and 1.5 wt% of wax were used during composite manufacturing. Test specimens were manufactured using extrusion and injection molding processes. The flexural, tensile, impact, and hardness properties of all specimens were determined. Scanning electron microscopy was used to study the morphology and interfacial compatibility of the filler in prepared composites. The test results showed that MAPP use and the filler rates affected the density and mechanical properties of test specimens.

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INTRODUCTION

Big leaf mahogany (*Swietenia macrophylla*) belonging to the family of Meliaceae is one of the most commercially important timber tree species in neo-tropical forests. It is naturally grown from southern Mexico to the southern Amazon basin of Bolivia, Brazil, and Peru (Anoop *et al.* 2014). Mahogany wood with the density of 0.55 g/cm^3 is used for various purposes including furniture, paneling, parquet and doors, buildings, interior and exterior, aircraft, wagon, small ship, violin making, marquetry, turning, engraving, *etc.* (Bozkurt and Erdin 1998; Lemmens 2005; Sharmin *et al.* 2015). During the conversion of the mahogany wood material from the logs to the desired shapes and sizes, a lot of waste is produced. The amount of waste varies according to the number of cuts, the number of operations performed, and the thickness of the cutter.

Wood plastic composites (WPC) consist of wood fibers or flours (WF) and polymers. WPC is manufactured with specific heat and pressure conditions with extruder and injection machines. The WF can be generated from various sources of fiber or flour. Polymers used are either in virgin or recycled form. Currently, the main applications of WPC products are cladding, decking, fencing, paneling, and furniture (Çetin *et al.* 2014; Özmen *et al.* 2014; Çavuş and Mengeloğlu 2016; Martikka and Karki 2019; Mengeloğlu and Çavuş 2019). Good adhesion of matrix with WF improves the mechanical properties of WPCs. Unfortunately, both components are naturally incompatible due to the hydrophilic nature of wood materials and hydrophobic nature of polymers. By using either physical or chemical modification, compatibility of the polymer matrix and WF in

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composites is improved. Compatibilizer/coupling agents can also considerably improve the performance of WPC (Klyosov 2007; Kim and Pal 2011). The preferred coupling agents for polyolefin are maleic anhydride-grafted polyethylene (MAPE) or propylene (MAPP).

During WPC production, the anhydride of the maleated polyolefins (MAPP or MAPE) reacts with hydroxyl groups on wood surface to form ester linkages. While the maleated ends of MAPP/MAPE bond to WF, the polyolefin side of the coupling agent interacts with the melted thermoplastic matrix, producing a bond between the hydrophilic WF and hydrophobic thermoplastic (Kattas *et al.* 2000; Rowell 2006; Yang *et al.* 2007). In actual production units, to improve the ease of processing and interfacial compatibility of WPCs, some lubricants, compatibilizer, or coupling agents are used during compounding. The addition of an internal lubricant is effective in reducing the apparent viscosity at high WF loadings. The apparent shear viscosity of the WPC melts decreases gradually with increased lubricant content (Li *et al.* 2004; Adhikary *et al.* 2011; Mengeloğlu and Çavuş 2019).

In this study, the effect of MAPP on the density and some mechanical properties of polypropylene (PP) composites were determined.

EXPERIMENTAL

Materials

Mahogany wood flour (WF) was used as a lignocellulosic filler. Mahogany flour was supplied by Yatch factory (İzmir, Turkey). Polypropylene (MH 418; density 0.905 g/cm^3, MFI 4.5 g/10 min, melting point 163 °C) was obtained from Petkim Petrochemical Co. (Turkey). Recycled polypropylene (rPP) was purchased from seller in Izmir (Turkey). Paraffin wax (K.130.1000) was used as a lubricant (softening point 56 °C to 58 °C, density 0.93 g/cm^3). Zinc stearate, a lubricant and mold discharging agent, (white powder, melting point 130 °C, density 1.1 g/cm^3) and MAPP (Licomont AR 504 by Clariant) were utilized as coupling agents (softening point 156 °C, density 0.91 g/cm^3) for improved performances of WPCs.

Methods

The mahogany wood particles were turned into flour form using a Wiley mill. These flours were classified as 40–60–80–100 to 200 mesh sizes. Mahogany WF passing through 60 mesh screen and staying on 80-mesh screen were used. The WF was dried in an oven for 24 h at 103 ± 2 °C). The moisture content of dried WF was below 1%.

The procedure for the manufacturing is given in Table 1. Depending on the method, WF, PP, rPP, MAPP, and wax were mixed in a high intensity mixer (900 to 1000 rpm in 2 s) to supply a homogeneous blend. These blends were compounded in a single-screw extruder at 40 rpm screw speed in the temperatures range of 170-180-185-190 to 200 °C (from barrel to die). The extruded compounds were first cooled in a water pool (23 °C ± 2 °C). Cooled extruded compounds were granulated into pellets by Wiley mill. The pellets were dried in oven at 103 ± 2 °C) for 24 h. The moisture content of pellets was reduced below 1% before the injection molding. The injection-molding machine (HAIDA HDX-88, Ningbo, China) was used for manufacturing specimens. Injection pressure was set as 5 to 6 MPa. The temperatures of machine were set as 180 °C to 200 °C from feed zone to die zone.

Table 1. Manufacturing Compositions

No	ID	PT	PR (%)	WF (%)	MAPP (%)	Wax (g)	Zn (g)
1	PP-0-0	PP	100	0	0	1.5	1.5
2	PP-20-0	PP	80	20	0	1.5	1.5
3	PP-40-0	PP	60	40	0	1.5	1.5
4	PP-0-3	PP	100	0	3	1.5	1.5
5	PP-20-3	PP	80	20	3	1.5	1.5
6	PP-40-3	PP	60	40	3	1.5	1.5
7	rPP-0-0	rPP	100	0	0	1.5	1.5
8	rPP-20-0	rPP	80	20	0	1.5	1.5
9	rPP-40-0	rPP	60	40	0	1.5	1.5
10	rPP-0-3	rPP	100	0	3	1.5	1.5
11	rPP-20-3	rPP	80	20	3	1.5	1.5
12	rPP-40-3	rPP	60	40	3	1.5	1.5

PT: Polymer type; PR: Polymer rate; WF: Wood Filler; Zn: Zinc stearate ($C_{36}H_{70}O_4Zn$).

Test specimens were first conditioned at room temperature (23 ± 2 °C) and $65 \pm 2\%$ relative humidity. The density was determined by a water displacement technique according to the ASTM D792 (2007) standard. Flexural, tensile, impact properties, and hardness of all specimens were determined according to ASTM D790 (2003), ASTM D638 (2001), ASTM D256 (2000), and ASTM D2240 (2010), respectively. Flexural and tensile testing were implemented on Zwick 10 KN (Ulm, Germany), while a HIT5, 5P (Zwick) was used for impact property testing on notched specimens. The notch was added using a RayRan™ Polytest notching cutter (London, UK). For statistical analysis, Design-Expert® version 7.0.3 statistical software (Minneapolis, USA) was used. Scanning electron microscopy (SEM) was used to determine the morphological properties of produced samples. They were dipped in to liquid nitrogen and snapped in half to attain clean surfaces. Before taking images at 20 kV and 100X magnification, the fractured surfaces were sputtered with gold.

RESULTS AND DISCUSSION

The effect of WF rates and MAPP use on the density and some mechanical properties of PP- and rPP-based composites were studied. The density, tensile strength (TS), tensile modulus (TM), elongation at break (EatB), flexural strength (FS), flexural modulus (FM), impact strength (IS), and hardness (H) values were determined. The densities of PP and rPP specimens were in the range of 0.884 g/cm^3 to 1.04 g/cm^3 and 0.90 g/cm^3 to 1.02 g/cm^3, respectively. The density values are presented in Fig. 1. Statistical analysis showed that WF rates had significant effect on density ($P < 0.0001$). This increase was believed to be due to the higher cell wall density of lignocellulosic materials (Mengeloğlu and Karakuş 2012; Karakuş and Mengeloğlu 2016). This was because the density of the wood substance in the cell wall is about 1.5 g/cm^3 (Mäkinen *et al.* 2002). The bulk density of wood flour depends on factors such as moisture content, particle size, and species (Clemons 2010). The MAPP use also had an effect on densities but this effect was less pronounced compared to WF content ($P = 0.0303$). It should be noted that the effect of MAPP on density was more obvious at high WF content (40%). Polymer type (PT), on the other hand, had no significant effect on densities. Thus, both PP and rPP composites provided similar results ($P = 0.5999$).
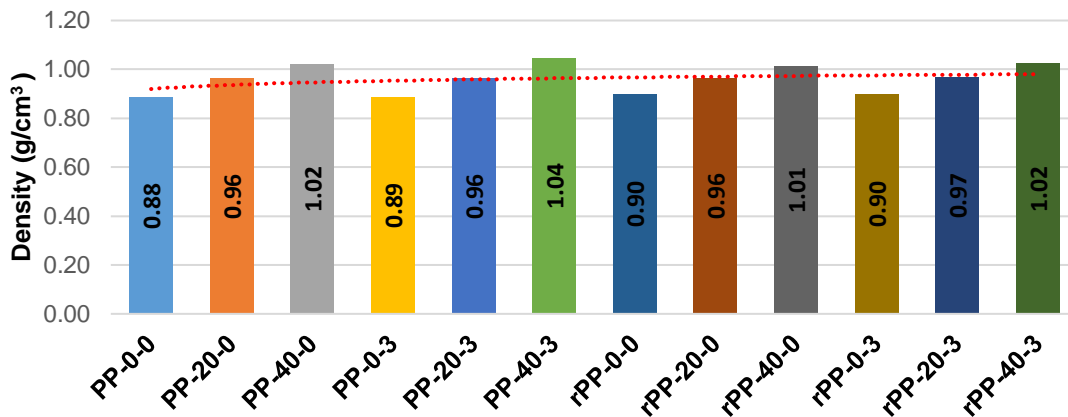
Fig. 1. Densities of manufactured PP and rPP composites 0 and 3% MAPP

The tensile strength values are presented in Fig. 2. The TS of PP and rPP specimens were in range of 18.7 MPa to 28.7 MPa and 19.0 MPa to 27.6 MPa, respectively. Statistical analysis showed that the WF rates were significantly important for TS ($P < 0.0001$). Regardless of having MAPP or PT, the TS values were reduced with increased concentration of WF. The usage of MAPP also had significant effect on TS of the composites ($P < 0.0001$). Both rPP and PP composites provided significantly higher TS values compared to composites having no MAPP in the formulations. This improvement was more pronounced in rPP based specimens. Statistical analysis showed that PT also had a significant effect on TS ($P < 0.0001$). The PP composites provided better TS than recycled ones, especially when MAPP was included in the formulation. The molecular chain length of PP might be reduced during recycling process, resulting in weaker properties.
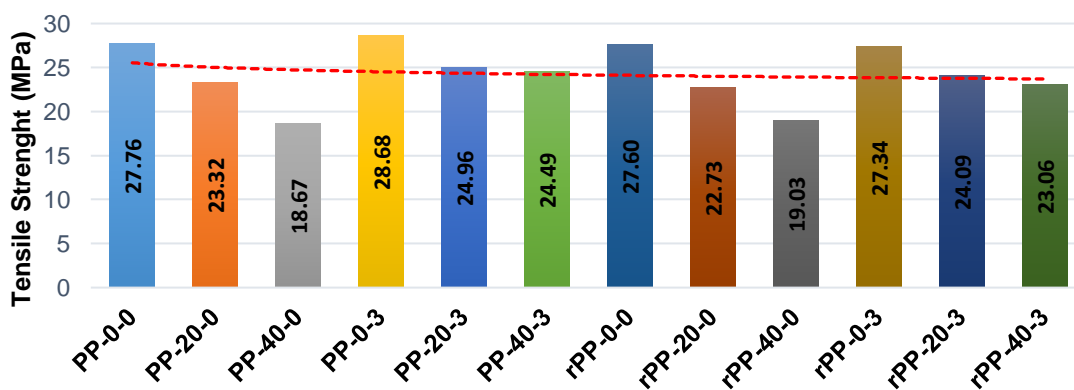

Fig. 2. Tensile strength of manufactured PP and rPP composites 0 and 3% MAPP

The morphology of the samples was studied using scanning electron microscopy (SEM) (Fig. 3). There were more holes caused by pulled out fibers and standing fibers in samples with no MAPP in the formulations. The rPP composites had more holes than the PP composites. After using 3% MAPP coupling agent, the number of holes and standing fibers were considerably reduced because of the improved adhesion between the polymer and WF. In previous studies (Clemons 2010; Lopez *et al.* 2012), MAPP composites have shown higher mechanical properties because MAPP minimizes the creation of microcracks between the dissimilar polar WF and non-polar PP matrix.
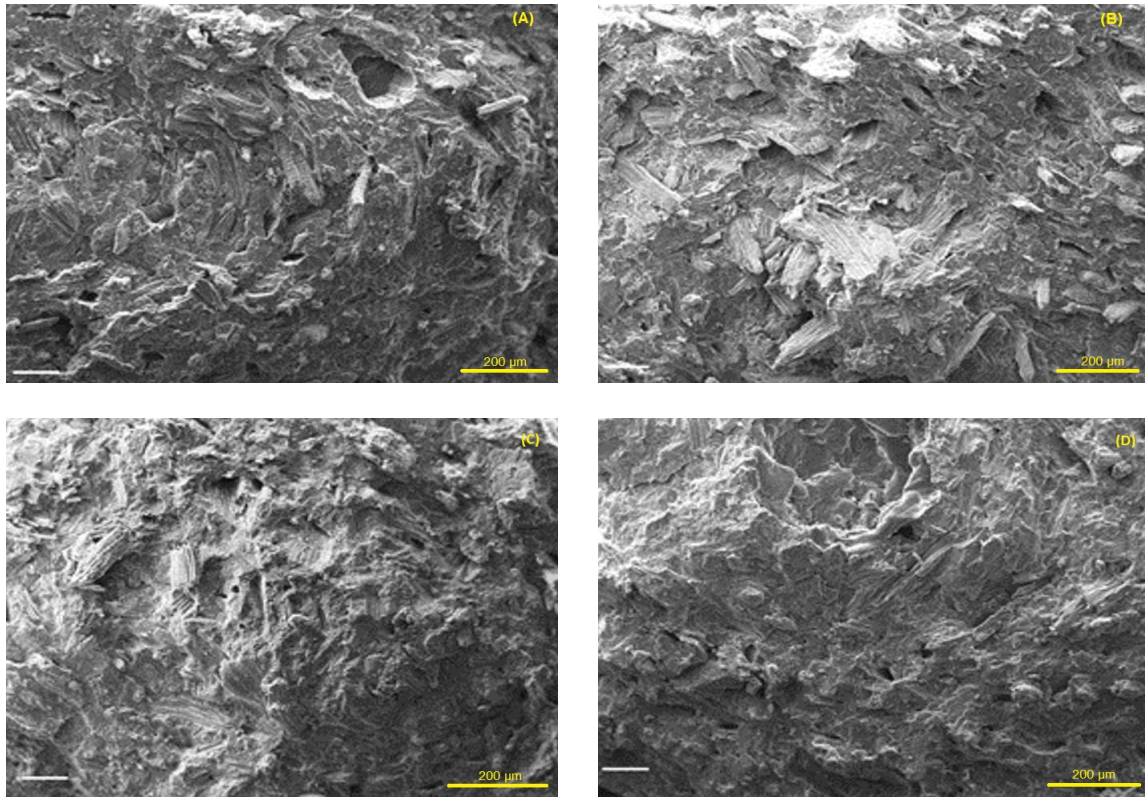
Fig. 3. SEM images of the manufactured composites; (A): PP-40-0; (B): rPP-40-0; (C): PP-40-3; (D): rPP-40-3

The TM of PP and rPP values were in the range of 453 MPa to 826 MPa and 503 MPa to 865 MPa, respectively. Figure 4 shows the TM values of test specimens. Statistical analysis showed that the WF amounts significantly affected the TM values ($P < 0.0001$). Regardless of PT and MAPP use, the TM values were increased with the rising concentration of WF.
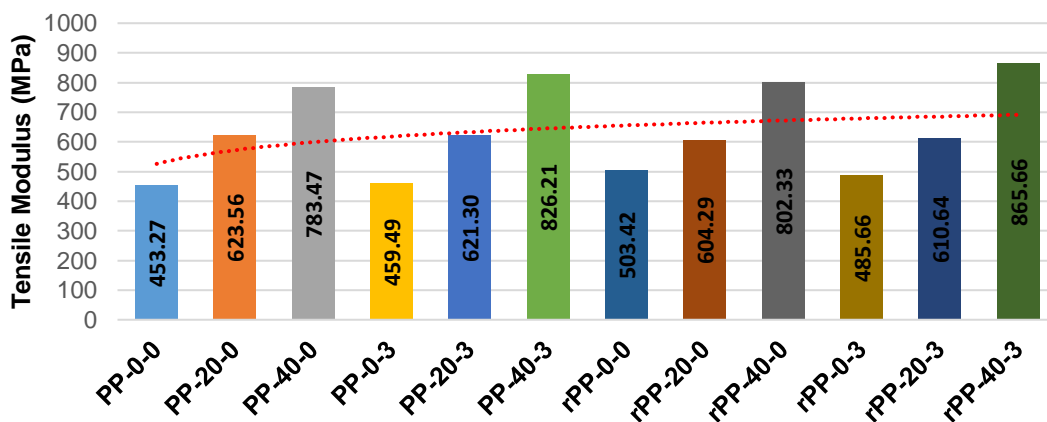

Fig. 4. Tensile modulus of manufactured PP and rPP composites 0 and 3% MAPP

The WF indicated higher modulus when compared to polymer matrix; as a result, their mixture showed modulus values higher than the polymer itself (Matuana *et al.* 1998;

Clemons 2010; Mengeloğlu *et al.* 2015). The MAPP content had a significant effect on TM of the composites (P = 0.0155). This improvement was more pronounced at high concentrations of WF.

The EatB values of PP- and rPP-based composites were in the range of 3.88% to 452.8% and 3.88% to 454%, respectively. The EatB values are presented in Fig. 5. Statistical analysis showed that the WF rates had a significant effect on EatB (P < 0.0001). Regardless of having MAPP or PT, the EatB values were reduced with increasing concentration of WF. The MAPP use also had a significant effect on EatB of the composites (P < 0.0001). Both rPP and PP composites provided significantly lower EatB values compared with composites having no MAPP in the formulations. Polymer composites became stiffer when the amount of WF in the matrix was increased. As a result, the EatB values were reduced. The EatB values are usually reduced with the increased modulus of the composites having WF, as reported for composites produced with various wood flours (Li and Matuana 2003; Mengeloğlu and Karakuş 2008).
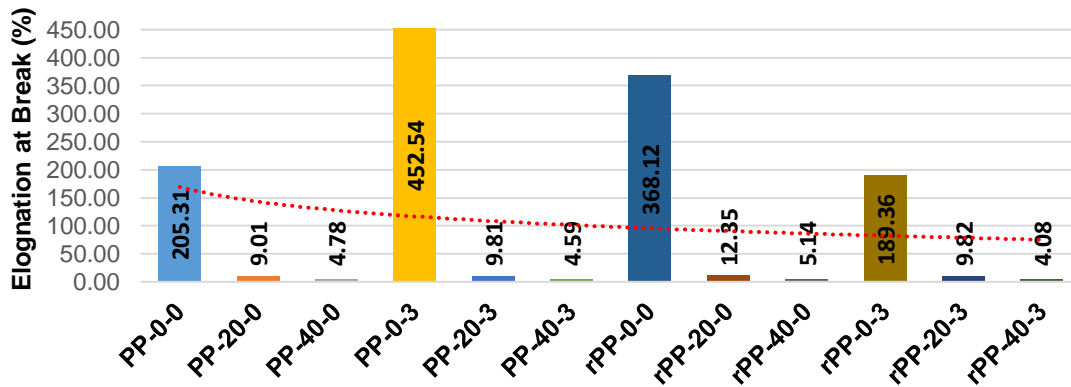

Fig. 5. Elongation at break of manufactured PP and rPP composites 0-3% MAPP

The FS values of PP- and rPP-based composites were in the range of 33.0 to 42.3 MPa and 32.4 to 40.9 MPa, respectively. Figure 6 presents FS values of the test specimens. Statistical analysis showed that WF rates had a significant effect on FS (P < 0.0001). Although addition of 20% WF showed some increase, FS decreased with 40% WF in composites without MAPP coupling agent. However, in composites with MAPP coupling agents, both PP- and rPP-based composites provided higher FS with increased WF content.
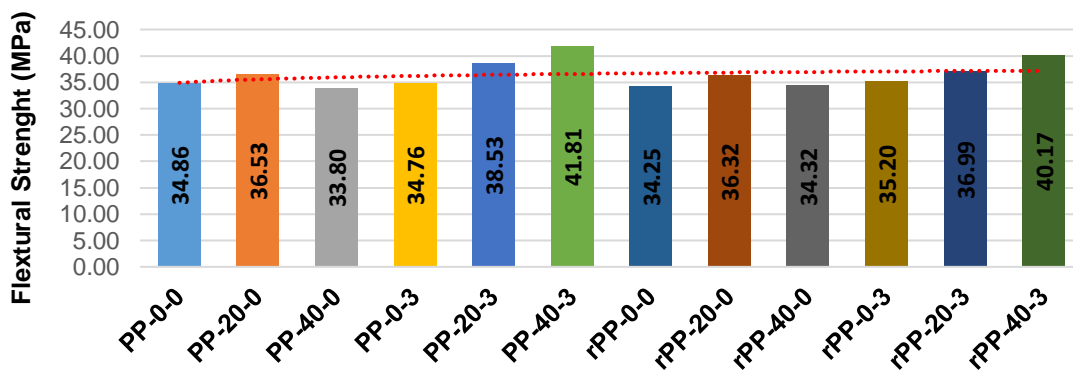

Fig. 6. Flexural strength manufactured of PP and rPP composites 0-3% MAPP

The MAPP use had significant effect on FS of the composites (P < 0.0001). Statistical analysis also showed that PT also had a significant effect on TS (P < 0.0001). The PP composites provided higher FS compared to recycled ones especially for formulation having MAPP in it.

The FM values of PP- and rPP-based composites were in the range of 996.5 to 2309 MPa and 1026 to 2260 MPa, respectively. The FM are presented in Fig. 7. Similar to the FS, FM was significantly affected by WF rates (P < 0.0001). The FM values showed improvement with WF, regardless of PT. The WF had higher modulus values than polymer (Bouafif 2009; Chaharmahali *et al.* 2010). The resulting composites of WF and polymer provide higher modulus values than the polymer itself (Clemons 2010; Lopez *et al.* 2012; Mengeloğlu *et al.* 2015).
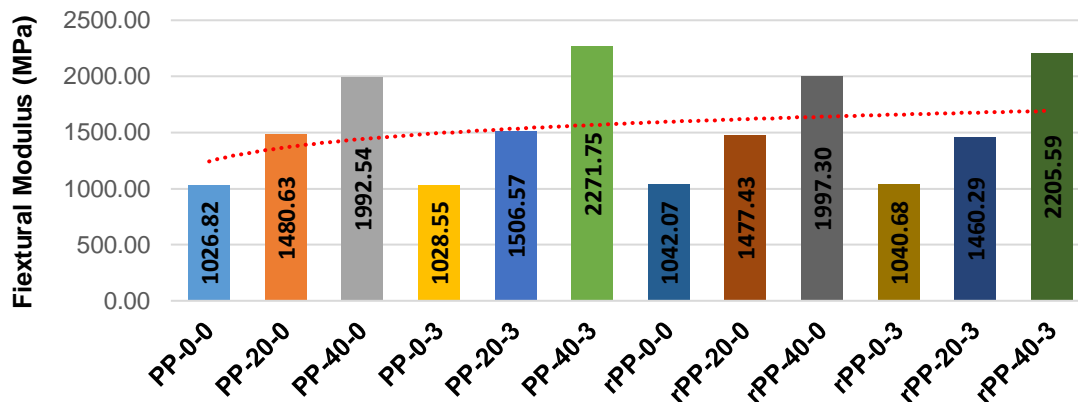

Fig. 7. Flexural modulus of manufactured PP and rPP composites 0 and 3% MAPP

The hardness values (Fig. 8) of PP- and rPP-based composites were in the range of 59.4 to 75.1 (Shore D) and 63.3 to 75.6 (Shore D), respectively. Statistical analysis showed that WF rate and MAPP use had significant effect on H values (P < 0.0001). The effect of WF on H values was more pronounced than that of composites with MAPP. The PT on the other hand had no significant effect on H values. For the material characterization standpoint, the hardness values of all manufactured composites provided Shore D values of around 60 and above, which are classified as "extra hard" material. Ibach and Rowell (2013) showed that a higher polymer loading yields a higher WPC hardness.
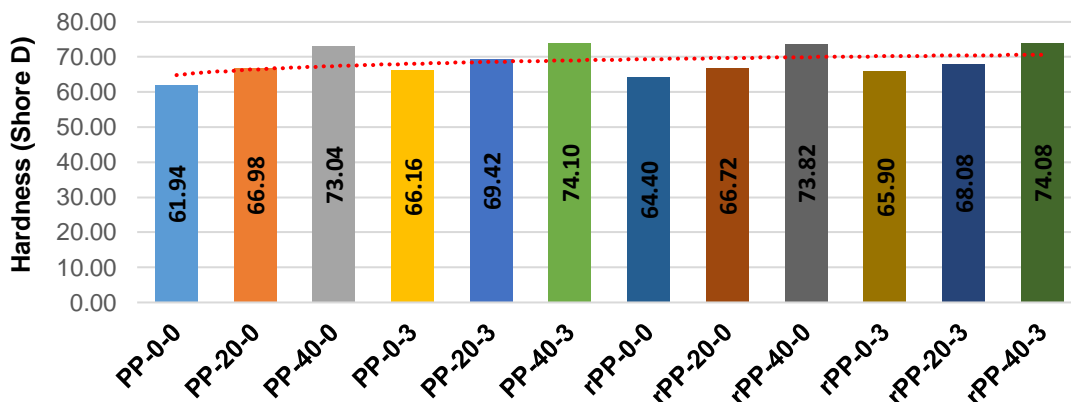

Fig. 8. Hardness properties of manufactured PP and rPP composites 0 and 3% MAPP

In the case of IS, manufactured PP- and rPP-based composites provided values in the range of 1.41 kJ/m^2 to 2.03 kJ/m^2 and 1.26 kJ/m^2 to 2.13 kJ/m^2, respectively. The notched IS are presented in Fig. 9. There were some differences on IS values of composites; however, statistical analysis showed that these differences were not significant for the factors PT (P = 0.2117), WF amount (P = 0.6242), and MAPP presence (P = 0.7636).
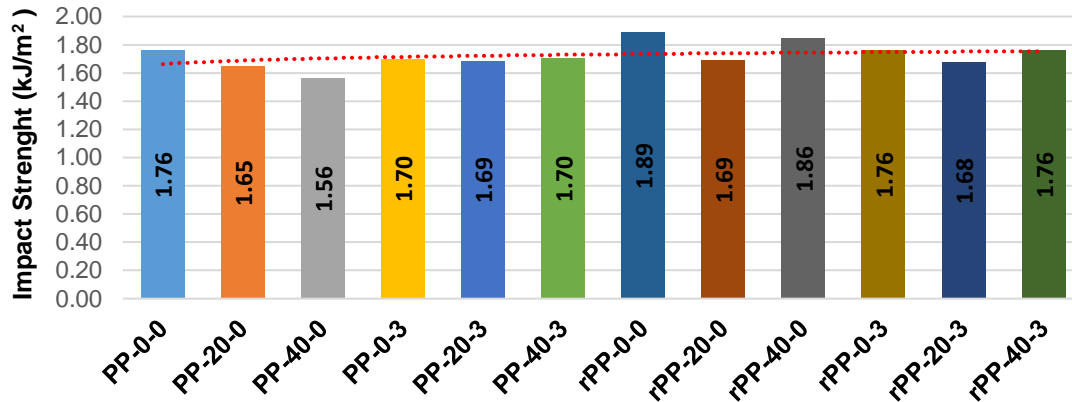

Fig. 9. Impact strength properties of manufactured PP and rPP composites 0 and 3% MAPP

CONCLUSIONS

1. The WF content and the presence of MAPP in composites played an important role on density and mechanical properties.

2. Addition of both WF and MAPP in PP- and rPP-based composites has increased density values.

3. The MAPP presence in both PP- and rPP-based composites provided improved TS and TM values with adding wood flours.

4. Regardless of PT, WF improved both FS and FM values of the composites having MAPP as coupling agent.

5. Overall, the mahogany WF can be utilized in both PP- and rPP-based composites when manufacturing with MAPP as coupling agent.

ACKNOWLEDGMENTS

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