Physical and Mechanical Properties of Recycled Polypropylene Composites Reinforced with Rice Straw Lignin

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Recycled polypropylene (RPP) and lignin represent by-products produced in enormous amounts worldwide that remain underutilized. This study used rice straw lignin as a filler at various concentrations (0% to 70% w/w) in RPP and virgin polypropylene (PP) composites by melt blending. Structural and morphological alterations of lignin were analyzed by Fourier transform infrared (FTIR) spectroscopy and scanning electron microscopy (SEM), respectively. Mechanical properties were evaluated using a universal testing machine (UTM). Results revealed that the tensile strength of the composites decreased as the lignin content increased, presumably due to the low of compatibility degree of lignin and MAPP, as well as the crack formation due to the agglomeration of lignin. However, composites with lignin as a filler showed higher moduli and water absorption capacities, as well as thickness swelling; using lignin as a filler caused a drastic reduction of the elongation at break values. The results indicated that the physical and mechanical properties of RPP and its virgin PP composites had no substantial differences. This indicated that virgin PP could be substituted by recycled polypropylene (RPP) for composite applications with the addition of MAPP.

INTRODUCTION

After cellulose, lignin is one of the most available sources of biomass for renewable polymer production. Polymer products can be derived from lignocellulosic sources such as trees, grasses, and agricultural crops (Hatakeyama and Hatakeyama 2009). In the pulp and paper industries, the separation of cellulose from wood generates approximately 50 million tons of lignin annually as wastes (Norgren and Edlund 2014). In addition, the emerging utilization of biomass for bio-energy liberates a large amount of lignin. Even though considered as a waste and the major application of lignin is burned to produce energy (Mu *et al.* 2014; Su and Fang 2014), lignin has potential as a reinforcer or filler for polymer composites due to the compatibility property with chemicals, and it possesses the reactive functional groups (Thakur *et al.* 2014).

To find alternative resources for petroleum-derived composites, lignocellulosic fillers are being increasingly used to reinforce thermoplastics (Azwa *et al.* 2013; El-Sabbagh 2014; Marques *et al.* 2014; Xie *et al.* 2014), as well as thermosetting plastics (Thakur and Thakur 2014).

Lignin has been investigated for sugarcane-based fiber composites and resulted in effective water barrier capability (Doherty *et al*. 2007). Lignin was blended together with epoxy resin and polyamine; the resulting composite showed good interfacial adhesion properties (Yin *et al.* 2012). In addition, lignin was blended with polyvinyl chloride and resulted in renewable and biodegradable materials with increased surface reflectance characteristics as the amount of lignin increases (Mishra *et al*. 2007). When lignin was blended with poly(L-lactic) acid, the impact strength of poly(L-lactic) acid was improved appreciably, as well as its toughness; the resulting composite was less expensive compared to neat poly(L-lactic) acid plastic (Mu *et al*. 2014). The impact strength and structural properties were also improved when lignin was added to a hemp/epoxy resin composite matrix (Wood *et al*. 2011).

Lignin is also feasible as a compatibilizer, as a fire retardant, and as a toughening agent, when it is used in conjunction with jute and polypropylene (Acha *et al*. 2009). The mechanical properties of epoxy resin improved when it was cured with 25% lignin obtained from empty fruit bunch biomass (Abdul Khalil *et al.* 2011a,b). However, when lignin was incorporated with a polysiloxane matrix, the resulting mechanical and thermo-mechanical properties of the composite did not change (Stiubianu *et al*. 2009). Lignin was used due to its low weight, biodegradability, abundancy, and underutilization. Therefore, this natural resource has potential as a filler for composites (Thakur *et al*. 2014).

Most of the studies that have been conducted related to the incorporation of lignin with the neat polymer. However, limited research has been reported on lignin utilization in conjunction with recycled polymers. Besides, plastic recycling is of particular interest and has intensified worldwide in an attempt to overcome pollution issues (Yang *et al.* 2004). Therefore, the utilization of recycled plastics for eco-composites has received much attention.

Considerable research has focused on composites made from recyclable polymers combined with kenaf fibers (Srebrenkoska *et al*. 2009). It was reported that thermal stability and flexural modulus of the composite improved when 5% esterified lignin was added to the recycled polystyrene (Pérez-Guerrero *et al*. 2014); similar results have been reported with the addition of unmodified kraft lignin (Schorr *et al*. 2015). However, there are no current studies that have reported on the utilization of rice straw lignin with recycled polypropylene (RPP).

Accordingly, recycled polypropylene (RPP) composites containing various concentrations of rice straw lignin were prepared to evaluate their physical and mechanical properties. It is well known that the adhesion between thermoplastic matrices (hydrophobic) and lignocellulose (hydrophilic) are poor. The different properties of the materials provide the blended composites with inferior moldability and low reinforcement (Febrianto *et al*. 2006). Therefore, maleic anhydride polypropylene (MAPP) was used for the current study. Moreover, MAPP is cost effective and provides dimensional stability of the composites (El-Sabbagh 2014).

The compound MAPP acts as a compatibilizer for polymer composites reinforced with lignocellulosic fibers. Maleic anhydride itself plays an important role as an effective adhesion proponent. This study aims to evaluate the physical and mechanical properties of rice straw lignin filled-recycled polypropylene composites utilizing MAPP as a compatibilizer.

EXPERIMENTAL

Materials

Composite matrices were produced from virgin polypropylene (PP J150 GEB 267) and recycled polypropylene (RPP) that was obtained from a local, post-consumer recycling plant of plastics located in the Bandung Province of West Java, Indonesia. Maleic anhydride grafted polypropylene (MAPP; Toyotac M-300) with a melt flow rate of 13 g/min and a melting point of 160 °C was donated by Toyo Seiki Kogyo Co., Ltd. (Nagano, Japan) and was used as a compatibilizer between lignin and PP or RPP. Alkali black liquor from rice straw pulping was obtained from the Padalarang Paper Mill located in West Java, Indonesia.

Methods

Lignin precipitation and purification

The initial pH of black liquor was in the range of 12 to 13. Sulfuric acid (25% concentration) was slowly added while stirring, and the pH was adjusted to 2 to 3. The color of the black liquor solution changed from black to brown. Excess water was decanted from the resulting precipitate. The precipitate was filtered through a pre-weighed filter paper, and washed with water. The lignin precipitation was then air-dried overnight, and it was then oven-dried at 105 °C. Lignin impurities were removed by the modified method described by Suhas *et al.* (2007). Approximately 20 g of oven-dried lignin and 150 mL methanol were stirred for 20 min; afterwards, the mixture was left to settle for 10 min. Excess methanol was decanted from the remaining solids; this was repeated again with the solids until the supernatant (*i.e.*, methanol) was light brown in color. The methanol fractions were evaporated and then oven-dried at 105 °C for 3 h.

Composites compounding

PP or RPP was placed in a double screw of a Laboplastomill 30R150 mixer (Toyo Seiki Seisaku-Sho, Ltd., Tokyo, Japan) at 180 °C, 10 rpm for 3 min. MAPP was added into the molten PP or RPP for 2 min. The concentration of MAPP was 2.5% with respect to the weight of RPP or PP used, which was based on the previous works of Karina *et al.* (2007). The PP and RPP were then thoroughly mixed with various lignin amounts (0% to 70%). The melt mixing was continued with a screw rotation rate of 30 rpm for 3 min. The total mixing time was 8 min.

Composite sheet preparation

Granulated composite samples were molded into a sheet by hot pressing. Blended samples (10 g) were placed between a pair of glossing plates with a 3-mm thick spacer. The press temperature was set at 180 °C and the granular sample pressed at 75 kgf/cm^2 for 30 min, which was then followed by cold pressing at the same pressure for 10 min.

Fourier transform infrared spectroscopy (FTIR)

The FTIR analyses were performed on a Shimadzu FTIR 4300 (Shimadzu Corp., Kyoto, Japan). Each of the composites samples was ground in a mortar with KBr pellets, and the resulting powder was pressed with a hydraulic press (400 kgf/cm^2) into a transparent disk. The FTIR spectra were recorded in a spectral range of 400 cm^{-1} to 4200 cm^{-1} at a resolution of 2 cm^{-1}.

Mechanical properties test

Mechanical properties of the composite sheets were measured in accordance with ISO 527-2 (2012). Prior to mechanical testing, the sample sheets were shaped into a dumbbell form in accordance with the ISO 527-5 (2012) standard, then conditioned at 23 °C and 50% relative humidity (RH) for a minimum of 40 h. Tensile tests were performed in accordance to ISO 527 (2012) using a universal testing machine (Orientec UCT-5T; Orientec Corp., Ltd., Toyko, Japan) in a room conditioned at 23 °C and 50% RH.

Morphological evaluation

Composite morphologies were visually observed using a Hitachi SU3500 SEM (PT. Hitachi High-Technologies, Jakarta Pusat, Indonesia). The specimen was cut and mounted in an aluminum holder with double-sided carbon tape. The specimen was sputter coated with a thin gold layer (JEOL type JFC-1100 E, Tokyo, Japan), and then placed into the SEM. The SEM was operated at low vacuum mode with an acceleration voltage of 5 kV to 10 kV.

Water absorption and thickness swelling

The water absorption and thickness swelling were measured in accordance with the procedure given by Febrianto *et al.* (2006). Specimens (50 mm x 25 mm x 3 mm) were prepared from the composites for five replications. The specimens were vacuum oven-dried at 60 °C overnight and stored in a desiccator. The weight and the thickness were measured (W1 and TS1) in a conditioned room (23 °C and 50% RH). Water absorption and thickness swelling was carried out by submerging the specimens in water for 24 h. The specimens were wiped and measured for water absorption and thickness swelling (W2 and TS2). The specimens finally were oven-dried in 60 °C overnight to obtain the constant weight. Water absorption and thickness swelling were calculated based on the following equation:

$$\text{Water absorption} = (W_2 - W_1)/W_1 \text{ x } 100\% \tag{1}$$

$$\text{Thickness swelling} = (TS_2 - TS_1)/TS_1 \text{ x } 100\% \tag{2}$$

RESULTS AND DISCUSSION

Fourier Transform Infrared Spectroscopy

The FTIR analyses were used to analyze the raw materials and the structural changes that occurred after composite compounding. Comparisons of 10% *versus* 70% lignin content in the composites are discussed. Figures 1 and 2 show the FTIR absorption peaks that corresponded to the molecular vibrations for the lignin alone, RPP, or PP alone, and lignin-RPP or lignin-PP composites. The FTIR spectra of lignin-RPP and lignin-PP composites had prominent absorption peaks for O-H and C-H stretching. Lignin had an absorption at 3505 cm^{-1}, which was attributed to the hydroxyl groups in the phenolic and aliphatic of structures of lignin, at 2976 cm^{-1}; this was characteristic of methoxy groups, and bands at 1519 cm^{-1}, which was assigned to aromatic ring vibrations (Rodrigues *et al.* 2002). As observed in these figures, the absorption peak at approximately 2976 cm^{-1} in PP and RPP was attributed to -CH$_3$ stretching (Devasahayam *et al.* 2013), whereas the peak at 1718 cm^{-1} in PP was assigned to the carbonyl groups.

Carbonyl groups could have been generated from the stabilizer that is present in PP (Barbeś *et al.* 2014). This peak was also found in the RPP with a lower intensity. The peaks found at 1165 cm^{-1}, 987 cm^{-1}, and 825 cm^{-1} were ascribed to a -CH$_3$ symmetric deformation vibration and -CH$_3$ rocking vibration (Chun *et al.* 2013). These three dominant peaks were also detectable in RPP. When compared to the spectrum of RPP or PP, the FTIR spectra of lignin-RPP or lignin-PP composites (Figs. 1 and 2) had vibration peaks for -CH$_3$ at 987 cm^{-1} and 825 cm^{-1} that became smaller and the –CH$_3$ vibration almost disappeared. This observation was ascribed to the incorporation of lignin into the plastic composite matrix.

Mechanical Properties

Table 1 shows the mechanical properties of lignin-RPP and lignin-PP composites. The test results revealed that the tensile strengths for the RPP-based composites were close to those for the PP composites. A slightly higher tensile strength value from the RPP than that of PP composites might be due to a kind of "crosslink", which is probably generated during the production of RPP. However, the tensile strength of the composites made from both RPP and virgin PP decreased as the amount of lignin increased. This tensile strength reduction could be attributed to the low compatibility of lignin with the PP matrices of the composites.

Compatibility of hydroxyl groups of lignin and MAPP will affect the tensile property of composites. However, this result showed that the amount of 2.5% MAPP was somewhat too low and inadequate to improve the tensile strength of the composites. Lower tensile strength properties with composites can also be associated with lignin agglomerations that create cracks inside the composites at higher filler amounts of lignin. The tensile strength value from RPP-lignin and PP-lignin composites showed no substantial differences. This result indicated that RPP is a possible replacement for PP in composite application.
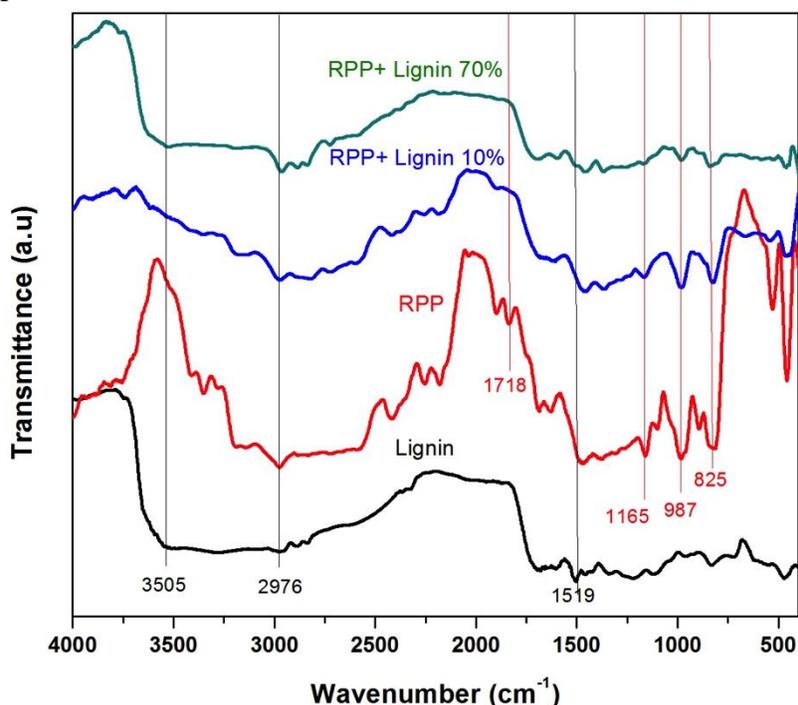

Fig. 1. FTIR spectra of lignin-RPP composites *versus* lignin and RPP components
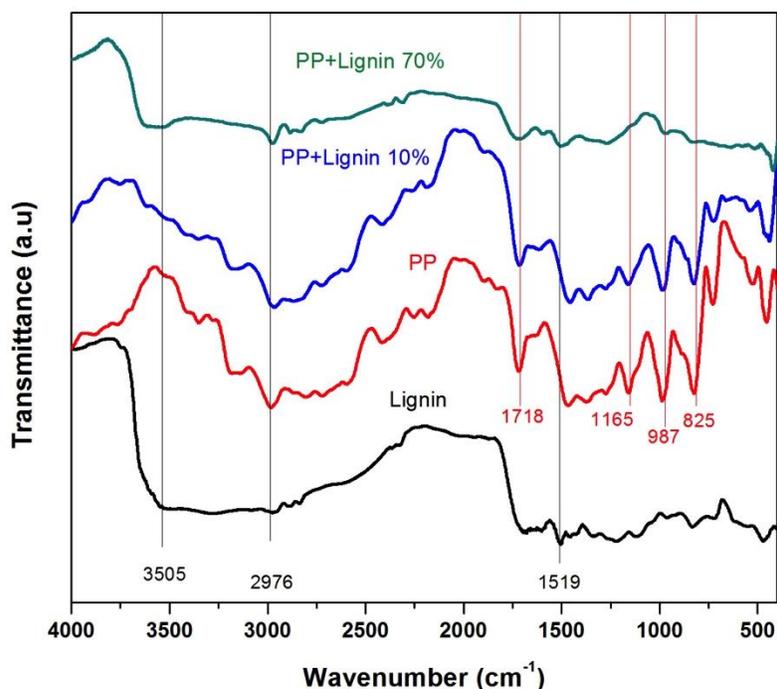
Fig. 2. FTIR spectra of lignin-PP composites *versus* lignin and PP components

The rigid characteristic that lignin imparts to plants (Feldman *et al.* 1987) would be expected to improve Young's modulus of RPP and neat PP composites. The addition of lignin improved the Young's modulus of the composites; in other words, the lignin addition improved the rigidity of the composites.

The elongation at break of the composites with RPP and with virgin PP decreased drastically as the amount of lignin filler increased. Also, the elongation at break of the composites with RPP and with virgin PP decreased drastically as the amount of lignin filler addition increased (Table 1). The rigidity of lignin may cause the elongation at break to decrease with the composites. Similar elongation at break values for RPP and PP composites suggested that the substitution of RPP for PP was feasible.

Table 1. Mechanical Properties of Lignin-RPP and Lignin-PP Composites

Lignin Content (%)	Tensile Strength (MPa)		Modulus (GPa)		Elongation at Break (%)	
	RPP	PP	RPP	PP	RPP	PP
0	37.8 ± 0.6	34.0 ± 0.5	1.22 ± 0.01	1.06 ± 0.02	44 ± 8	700 ± 40
10	24.3 ± 0.9	23.4 ± 0.5	1.24 ± 0.05	1.08 ± 0.04	6.9 ± 1.7	7.5 ± 0.9
20	18.2 ± 0.6	19.1 ± 0.4	1.25 ± 0.04	1.14 ± 0.02	4.1 ± 0.7	5.6 ± 1.0
30	15.2 ± 0.1	16.0 ± 0.6	1.32 ± 0.06	1.25 ± 0.05	3.5 ± 1.1	5.2 ± 1.3
40	12.5 ± 0.5	13.1 ± 0.3	1.36 ± 0.08	1.29 ± 0.05	1.9 ± 0.2	4.4 ± 2.5
50	10.4 ± 0.6	10.8 ± 0.4	1.36 ± 0.08	1.35 ± 0.03	1.6 ± 0.3	1.6 ± 0.4
60	8.2 ± 0.4	8.9 ± 0.3	1.37 ± 0.10	1.35 ± 0.07	1.4 ± 0.0	1.3 ± 0.1
70	7.1 ± 0.5	6.8 ± 0.6	1.44 ± 0.07	1.38 ± 0.09	0.7 ± 0.3	0.7 ± 0.1

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Water Absorption and Thickness Swelling

Table 2 shows the water absorption and thickness swelling values of the lignin-RPP and the lignin-PP composites after immersion in water for 4 weeks. The values for water uptake and swelling of the RPP and PP was negligible; this observation could have been attributed to the hydrophobic nature of these homopolymers. Thus, it could be assumed that all the water absorption and thickness swelling of the composites was caused by lignin and not by the RPP or PP. The dispersion of lignin in RPP or PP may have created voids which allowed water to absorb.

Higher water absorption and thickness swelling values were observed when the lignin content in the composite was increased and/or the soaking time was increased. These values increased linearly as the addition of lignin increased. These results are similar to other natural fiber additives as enforcing agents for composites (Karina *et al*. 2007; Khalil *et al*. 2007). The addition of 10% lignin to PP or RPP resulted in composites with minimal water absorption and thickness swelling when compared to the other composites (Table 2). Both composites exhibited low water absorption and thickness swelling. These results could be a constraint for their application. These findings suggested that hydrophobic properties of polypropylene were unaltered as it was recycled. Thus, this finding suggested that RPP was able to replace PP for polypropylene-based composites.

Table 2. Water Absorption and Thickness Swelling Values of Lignin-RPP and Lignin-PP Composites

Lignin Content (%)	Water Absorption (%)				Thickness Swelling (%)			
	Immersing time (h)				Immersing Time (h)			
	0		24		0		24	
	RPP	PP	RPP	PP	RPP	PP	RPP	PP
0	0.45	0.38	0.45	0.38	0.86	0.87	0.86	0.92
10	0.49	0.49	0.49	0.50	0.97	1.05	1.02	1.10
20	0.59	0.52	0.60	0.59	1.02	1.09	1.07	1.15
30	0.59	0.52	0.60	0.60	1.07	1.10	1.13	1.16
40	0.64	0.52	0.65	0.62	1.11	1.15	1.17	1.21
50	0.65	0.59	0.66	0.67	1.13	1.19	1.19	1.25
60	0.74	0.64	0.75	0.72	1.22	1.45	1.28	1.53
70	0.78	0.70	0.76	0.74	1.25	1.58	1.32	1.66

Morphological Evaluation

To clarify the observed results in the tensile strength properties, the fracture surfaces of the test specimens were examined by SEM. The SEM micrographs of lignin-RPP and lignin-PP composites that contained 10% and 70% lignin loading are shown in Fig. 3. The RPP composite had a rough surface (Fig. 3a), whereas the PP composite had a smooth morphology (Fig. 3b). The rough surface of RPP was probably due to by product formation of "crosslink" during the production process of RPP. As shown in Figs. 3c and 3d, there were voids inside the composites where lignin was added into. Higher amounts of lignin resulted in more void spaces (Figs. 3e and 3f). This could have been attributed to the dispersion of lignin, which was less homogeneous, and attributed to the agglomeration of lignin particles inside the matrices. The results from the SEM

confirmed the presence of voids, which likely contributed to lowering the tensile strength of the composites. In addition, the lignin-RPP and lignin-PP composites exhibited similar morphology.
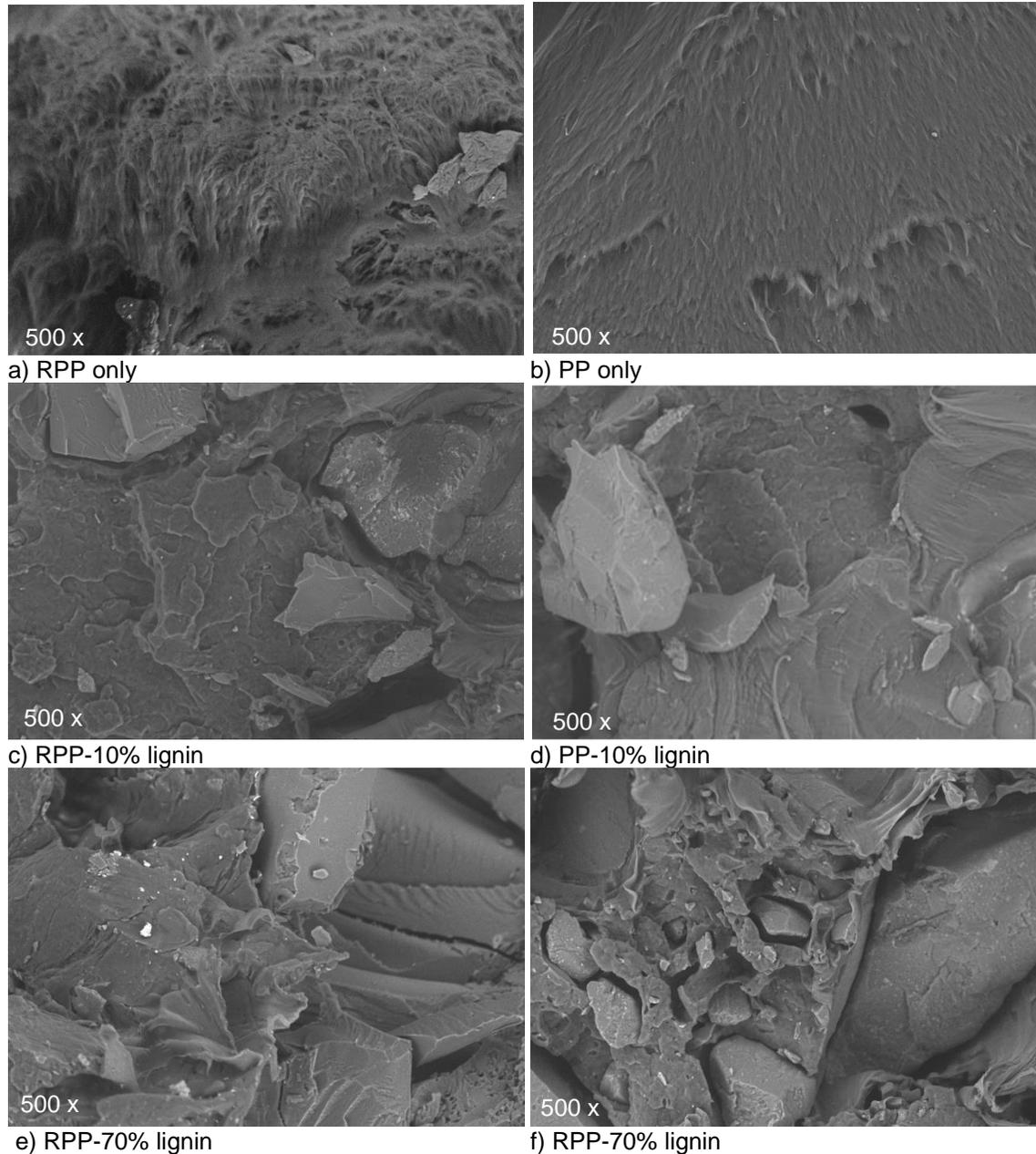

a) RPP only   b) PP only

c) RPP-10% lignin   d) PP-10% lignin

e) RPP-70% lignin   f) RPP-70% lignin

Fig. 3. Morphology of lignin-RPP and lignin-PP composites

CONCLUSIONS

1. Tensile strength and elongation at break properties decreased as the amount of lignin filler to the polypropylene (PP) matrix increased. However, composites with lignin as

filler showed higher moduli properties, water absorption capacities, and thickness swelling values.

2. This work indicated that the physical and mechanical properties of composites made from a combination of recycled polypropylene (RPP) and maleic anhydride polypropylene compatibilizer (MAPP) had no substantial differences when compared to PP+MAPP-based composites. Thus, RPP has the potential to substitute virgin PP in composites with additional MAPP applications.

ACKNOWLEDGMENTS

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