Mechanical, Thermal and Morphological Properties of PLA/PP Melt Blends

Mohd Bijarimi[1,2], Sahrim Ahmad[2], Rozaidi Rasid[2]

***Abstract*—** In this study, polylactic acid (PLA) was melt blended with polypropylene (PP) and liquid natural rubber (LNR) with the ratio of PLA/PP (90/10) and PLA/PP/LNR (90/10/10) in the Haake Rheomix internal mixer. The mechanical properties of such as stress-strain, flexural and impact were studied. It was found that the elongation at break, flexural and notched impact strength increased significantly for the LNR compatibilized PLA/PP blend. The DSC and FTIR showed the PLA/PP and PLA/PP/LNR were not miscible.

***Keywords*—**PLA, PP, blend, liquid natural rubber.

I. Introduction

Polylactic acid (PLA) is gaining popularity due to its biodegradable, renewability and comparable properties with petroleum-based polymers. One of the major concerns of using non-biodegradable polymer is disposal after end the product life cycle. The emergence of biodegradable plastics promises an alternative solution for this disposal problem.

PLA possesses the tensile strength and stiffness similar to polyethylene terephtalate and processing characteristics of polystyrene, but it suffers low impact resistance. The poor toughness limits its use in the applications that need plastic deformation at higher stress level [1]. Blends of PLA with other polymer is the most attractive and practical route towards modifying its physical properties. Polypropylene (PP) is an excellent polymer with a combination of outstanding physical, chemical, mechanical, thermal and electrical properties. It was therefore not surprising that PP has been blended with other polymers such as polyethylene (PE) and natural rubber (NR) for physical property modification. This work reports the melt blend of PLA/PP and PLA/PP with liquid natural rubber (LNR) compatibilizer.

II. MATERIALS AND METHOD

Polylactic acid of Natureworks Ingeo™ Biopolymer 2002D grade supplied by Unic Technology Ltd, China was a thermoplastic resin used in this study. It has a density of 1.24 g/cm^3 and melt flow index of 5-7 g / 10 min. (190°C / 2.16 kg) and melting temperature between 160-170^{o}C. As for the PP, an injection molding grade with a melt flow index of 7.5 g / 10 min was used. Natural rubber of SMR L type with a density of 0.91 g/cm^3 was a product from Malaysia Rubber Board. Liquid natural rubber (LNR) was synthesized using a photochemical oxidation technique on SMR L in our laboratory [2, 3]. Other chemicals were used as received. All melt blends prepared in a laboratory mixer (Haake Rheomix 600p) at 180^{o}C with a capacity of 60 g. Blending was carried out with a rotor speed of 50 rpm for 15 min. The PLA was initially melted for 120 s and subsequently LNR (if applicable) and PP were incorporated after 20 seconds. The blend was removed from the internal mixer and then molded at 180^{o}C under 45MPa of pressure for 13 minutes using a hot press into thin sample sheets (150 mm x 150 mm) for test specimens.

All compositions of blend were tested and compared in terms of their mechanical properties. Tensile test was carried out according to ASTM D638 using a Testometric under ambient conditions with crosshead speeds of 50 mm min^{-1}. The flexural strength and modulus were also measured on the same tensile machine according to ASTM D790 with a 3mm/min strain rate. The Izod impact properties of the blends were determined by the Ray-Ran Impact tester on notched specimens. The glass transition temperature (T_g) and melting temperature (T_m) of the blend components were characterized with a Mettler Toledo DSC 822 on compression molded specimens. As for thermal stability of PLA and blends, a Mettler Toledo TGA/SDTA 851 apparatus was used. The chemical changes after blending were monitored by FT-IR spectroscopy. The IR spectra were recorded using a Shimadzu 8400 M FT-IR spectrometer with 4 cm^{-1} resolution and 10 scans. All spectra were recorded in the absorbance mode in the 3500–600 cm^{-1} region.

III. RESULTS AND DISCUSSION

A. Mechanical Properties

The tensile properties of the PLA matrix and PLA/PP blends are shown in the Table 1. It is shown that the neat PLA with tensile strength of 69.91 MPa, Young's modulus of 1968 MPa and elongation at break at 3.8%. In general, the incorporation of PP has reduced the tensile strength and Young's modulus of the PLA matrix but with improved elongation at break. This can be explained by incompatibility of PP and PLA due to the difference in polarity. However, in the LNR compatibilized system, i.e. PLA/PP/LNR (90/10/10), the Young's modulus is comparable to PLA/PP (90/10)

Mohd Bijarimi is with the [1]Universiti Malaysia Pahang, Kuantan Pahang Malaysia, (corresponding author phone:6095492918 ; fax: 6095492889 ; e-mail: bijarimi@ump.edu.my).
Sahrim Ahmad & Rozaidi Rasid are with [2]Universiti Kebangsaan Malaysia, Bangi, Malaysia. (e-mail; sharim@ukm.my, rozaidi@ukm.my)

uncompatibilized blend system. As expected the tensile strength has reduced from 32.2 MPa to only 18.5 MPa; a 42% reduction from the uncompatibilized system. On the contrary, we saw a marked improvement ca. 46% in the elongation at break in the compatibilized blend system.

The effect of flexural properties of the PLA/PP blends is shown in the Table 1. The neat PLA showed a flexural strength of 94 MPa. With the incorporation of 10 wt. % PP, the value dropped tremendously from 94 MPa to 12.5 MPa only; indicating an immiscible PLA/PP blend. Nevertheless, the flexural strength increased by 224% i.e. from 12.6 MPa to 40.7 MPa when the 10 wt. % of LNR was introduced in the PLA/PP (90/10) blend. This can be explained by the role of LNR which acts a compatibilizer in the PLA/PP phase. On the other hand, the flexural modulus dropped about 21.7 % i.e. from 3278 MPa to 2567 MPa.

The notched Izod impact strength of the neat PLA shows an impact strength of 2.44 kJ/m^2; indicating a very brittle polymer. In the PLA/PP (90/10) system, it was found that the value dropped slightly to 2.33 kJ/m^2; shows that incorporation of PP has not altered the impact property of PLA. However, when the 10 % wt. of LNR was added into the PLA/PP (90/10) blend system, the impact strength increased significantly i.e. from 2.44 kJ/m^2 to 5.72 kJ/m^2 for neat PLA and PLA/PP/LNR (90/10/10) respectively. .

B. Thermal Properties

Table II illustrates the T_g, T_m and percentage of crystallinity of the PLA and PLA/PP blends. It is evidently the neat PLA has a T_g of 62.8 ^oC and T_m of 167.9 ^oC. The incorporation PP part has not significantly changed the T_g and of the PLA/PP (90/10) blends system. Nevertheless when the LNR was added to the blend system, we saw a reduction of glass transition temperature from 62.0 of neat PLA to 55.6 ^oC as shown in the Table II. The thermal degradation temperature of PLA and blends showed that there were no significant changes to the peak and end degradation temperatures except a slight drop on the onset degradation temperature (333.0^{o}C vs. 331.8^{o}C) for the PLA/PP/LNR (90/10/10) blend.

C. Morphology

The studies on morphology of the polymer blend are of paramount importance for determining the structure and property relationship. The morphology characterization will provide insights, among others, such as particle size, distribution of the rubbery impact modifiers in a polymer matrix, the distribution of components in a binary blend, the effect of interfacial addition on the particle size, the crystalline phase, dispersion/agglomeration of particles, as well as distribution of fillers in the polymer blend [4]. Fig. 1 & 2 illustrate the SEM micrograph examined from the tensile fracture surfaces for neat PLA/PP and PLA/PP/LNR blends respectively. It can be seen that there was a phase separation between the PLA and PP particles as shown in the Fig. 1. However, when LNR was added, the morphology of the blend showed a more homogenized and refined structure as evident in the Fig. 2. This can be explained by the role of LNR in compatibilizing the PLA and PP phases which resulting the better elongation at break, flexural and impact strength as discussed earlier. Liquid natural rubber is a chemically modified NR generated via oxidative degradation of NR. The PLA and PP are immiscible with LNR as confirmed by the FTIR analysis shown in Fig. 3.

TABLE I
MECHANICAL PROPERTIES OF PLA AND BLENDS

Property	PLA	PLA/PP (90/10)	PLA/PP/LNR (90/10/10)
Stress at Break, MPa	69.9	32.2	18.5
Elongation at Break, %	3.8	7.8	21.3
Young's Modulus, MPa	1968	1174	1178
Flexural Strength, MPa	94.0	12.5	40.7
Flexural Strain, %	5.5	12.6	18.9
Flexural Modulus, MPa	3536	3278	2567
Notched Impact Strength, kJ/m^2	2.44	2.33	5.72

TABLE II
THERMAL PROPERTIES OF PLA AND BLENDS

Property	PLA	PLA/PP (90/10)	PLA/PP/LNR (90/10/10)
DSC			
T_g	62.0	61.3	55.6
T_m	170.7	157.0	158.9
Crystallinity, %	11.7	25.0	11.9
TGA			
Onset Temp., ^oC	333.0	333.9	331.8
Peak Temp., ^oC	365.1	364.8	364.7
End Temp. ^oC	384.7	384.0	383.5

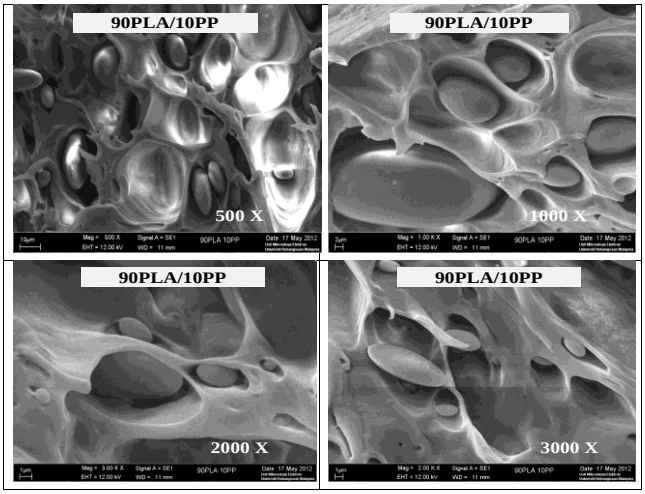

Fig. 1. SEM micrographs of tensile fractured specimen for PLA/PP (90/10) blend.

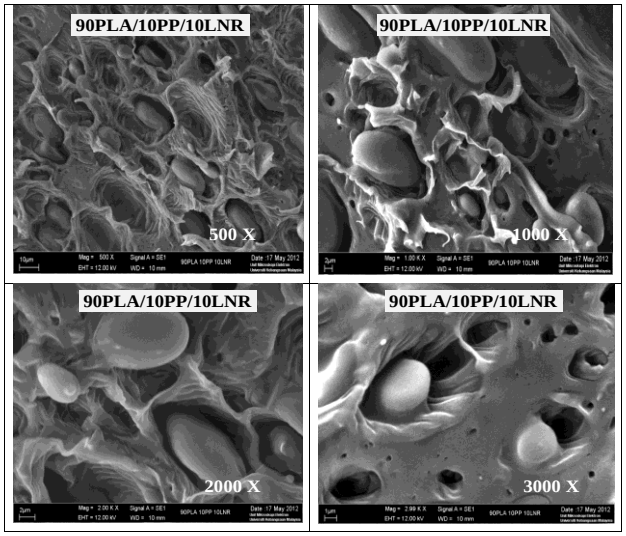

Fig. 2. SEM micrographs of tensile fractured specimen for PLA/PP/LNR (90/10/10) blend.

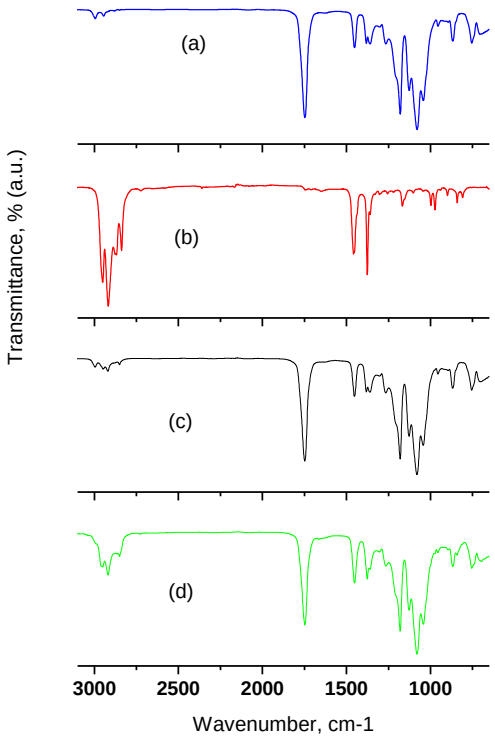

Fig. 3. FT-IR spectra of (a) PLA (b) PP (c) PLA/PP (90/10) (d) PLA/PP/LNR (90/10/10)

IV. Conclusion

In this study, we have shown that PLA toughness could be improved by direct melt blending with PP and LNR. The improvement of the elongation at break, impact strength and flexural properties were contributed by the LNR compatibilizer as evident by a more homogenized and refined blend morphology by SEM observation.

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