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## Preparation of Polymer Blends Between Poly(lactic acid) and Poly(butylene adipate-co-terephthalate) and Biodegradable Polymers as Compatibilizers

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### Abstract

Biodegradable polymers having good toughness were used as compatibilizers for the polymer blend system of poly(lactic acid) (PLA) and poly(butylene adipate-co-terephthalate) (PBAT) to replace the inorganic or non-degradable compatibilizers. Biodegradable polymers such as poly(butylene succinate) (PBS), poly(butylene adipate-co-terephthalate) (PBAT), and poly(hydroxybutylate-co-valerate) (PHBV) have capability to mix with PLA. Polymer blends of PLA/PBAT (70:30) were added with 1, 3 and 5 phr of PBS, PBSA and PHBV. It was found that thermal property of polymer blends slightly decreased, melting temperature ( $T_m$ ) of the compositional polymers were almost not changed which indicated the phase separation of compositions. The melt flow of polymer blends increased with increasing amount of PBS while it was not changed with addition of PBSA and PHBV. Modulus of polymer blend with addition of 1 wt% PHBV was slightly increased while modulus of most of the polymer with addition of PBS and PBSA decreased. The results confirmed the phase separation of polymer components.

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## 1. Introduction

A biodegradable poly(lactic acid) (PLA) is a thermoplastic polyester produced from lactic acid, which is derived from the fermentation of sugar, cassava, corn starch and other polysaccharide sources [1]. PLA is becoming increasingly popular as a biodegradable engineering plastic owing to its high mechanical strength. However, PLA is brittle, softens at temperatures around 60°C and has lower water vapour and gas barrier properties compared to equivalent petroleum based polymers [2,3,4]. Therefore, it is needed to adjust the property of PLA for the advanced processing and product applications. Blending of PLA with PBAT has been studied by researcher and The convenience method to modify the PLA property is blending PLA with other biodegradable polymer having higher toughness and will enhance the fabrication processing. Poly(butylene adipate-*co*-terephthalate) (PBAT) is a biodegradable aliphatic–aromatic copolyester and is a commercial product with trademark Ecoflex from BASF suitable to blend with PLA. Moreover, PBAT is flexible and has a higher elongation at break than most biodegradable polyesters and more suitable for packaging films [5]. The main limitations toward wider industrial applications of PBAT are its poor thermal and mechanical resistance that limits its application to packaging. Nevertheless, the above drawbacks could be overcome by enhancing thermal and/or mechanical properties through filling techniques [6-9]. The drawback of polymer blends of PLA/PBAT is the phase separation which is affected the product mechanical properties. In this work we have studied the addition of functional biodegradable polymers as compatibilizers to enhance the mixing of PLA and PBAT as well as assist the biodegradability of the products. The compatibilizers studied included poly(butylene succinate) (PBS), poly[(butylene succinate)-*co*-adipate] (PBSA), and poly(hydroxybutyrate-*co*-hydroxyvalerate) (PHBV) [10-14]. The physical and mechanical properties of the polymer blends were measured and reported herein.

## 2. Experimental

### 2.1. Materials

PLA grade 2002D was purchased from Natureworks Co., Ltd. PBAT used are grade Ecoflex FBX 7011 from BASF Corp., PBS grade 1001MD and PBSA grade 3001MD was purchased from Showa High Polymer Co., Ltd., (Bionolle®). PHBV grade Y1000P was purchased from Tianan Biologic Material Co., Ltd. The properties of all biodegradable polymers are shown in Table 1.

Table 1. Polymers physical and mechanical properties

	PLA	PBAT	PBS	PBSA	PHBV
density	1.24	1.25	1.26	1.23	1.24
MFR	4-8	2.7–4.9	1-3	1-3	1.5-2.5
Tm (°C)	150-160	110-120	114	94	170-175
Tensile Strength @ Break, (MPa)	53	-	62	40	-
Tensile Yield Strength, (MPa)	60	35/44	31	18	33
Tensile Modulus,	3.5 GPa	-	470 MPa	320 MPa	1.4 GPa
Tensile Elongation, %	6.0	560/71	660	780	2.28
Notched Izod Impact,	12.81 J/m	-	24 kJ/m	29 kJ/m	7.5 kJ/m

### 2.2. Preparation of PLA /PBSA/PBAT blends

Polymer blends of PLA/PBAT (70:30) were added with 1, 3 and 5 phr of PBS, PBSA and PHBV. Before extrusion, PLA, PBAT, PBS, PBSA and PHBV resin were dried at 80 °C for 8 hours in an oven in

order to remove any trace of moisture to prevent potential hydrolytic degradation during the melt processing in the extruder. The amount of PBS, PBSA and PHBV are 1, 3, 5 phr. The polymers were blended using a twin screw extruder at 150-170 °C with screw speed 80 rpm, pressure 4.5 bar. The polymer blend was cut into pellets and dried at 80 °C for 8 h.

### 2.3 . Thermal property analysis

The thermal properties of the polymers were determined by differential scanning calorimetry (DSC). Polymer sample 5–10 mg was sealed in aluminum pans. The heating rate was 20 °C/min under nitrogen.

### 2.4 . Rheological property analysis

The melt flow of polymer melt was measured according to ASTM D1238 type 6452. The flow rate is determined as a function of applied load. Melt flow index (MFI) is the mass flow rate expressed in gram mass per 10 minutes. The sample of 5 g as a power or a pellet is heated for 5 minutes in the barrel and extruded through the die under a constant load 2.16 kg. The melt flow index of PLA /PBAT blends and polymer blends with PBS, PBSA and PHBV were measured at temperature of 190 °C.

### 2.5 . Mechanical property analysis

The tensile properties were determined according to ASTM D638 (Type I) using dumbbell shape specimens. The test was carried out on a universal testing machine (LLOYD Instruments LR 10K Plus) with a crosshead speed of 50 cm/min. The elongation of specimens was obtained from the extensometer. The Izod impact strength of was determined according to ASTM D256 by using the Impact Tester GOTECH GT 7045. The report value was the average of three replicates for each property test.

## 3. Results and Discussion

### 3.1 . Melt flow property of polymer blends

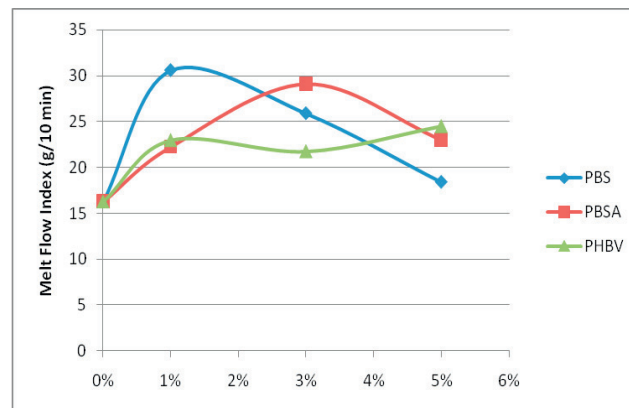


Fig. 1. Melt Flow Index of PLA/PBAT with PBS, PBSA and PHBV at 1, 3 and 5 phr

Addition of PBS, PBSA and PHBV to the blends of PLA/PBAT (70:30 by weight) increased the melt flow of the polymers. With 1 phr of PBS the melt flow increased obviously but decreased with more

amount of compatibilizer. Less amount of PBS may combine PLA and PBAT while more amounts of small molecules of PBS may act as the nucleating agent promoted crystallinity of polymer blends and phase separation may occur. The results are in agreement with tensile strength and sharp melting temperature peaks from DSC measurement of 3 and 5 phr PBS. Melt flow of blends with addition of PHBV increased constantly while the case of PBSA showed highest value at 2 phr. The results confirm the assist of compatibilizers to mixing of PLA and PBAT.

### 3.2. Mechanical property analysis

The tensile properties of polymer blends of PLA/PBAT (70:30) with addition of 1, 3 and 5 phr of PBS, PBSA and PHBV were determined according to ASTM D638. The results are shown in Fig. 2.

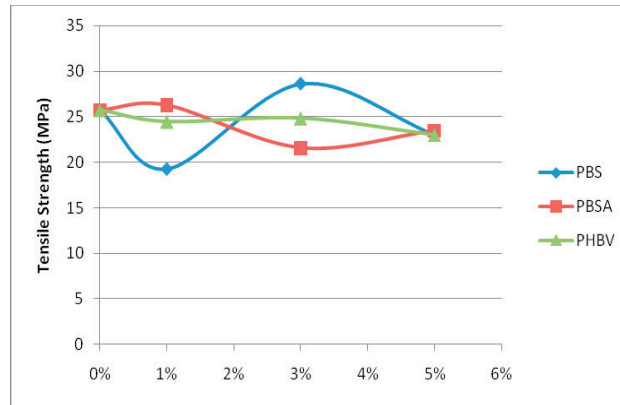


Fig. 2. Tensile strength of polymer blends PLA/PBAT with PBS, PBSA and PHBV 1,3,5 phr

Addition of 3 wt% PBS increased the tensile strength of polymer blend but decreased with 1 wt%. PHBV did not show variation of tensile strength. Similar results were seen for PBSA.

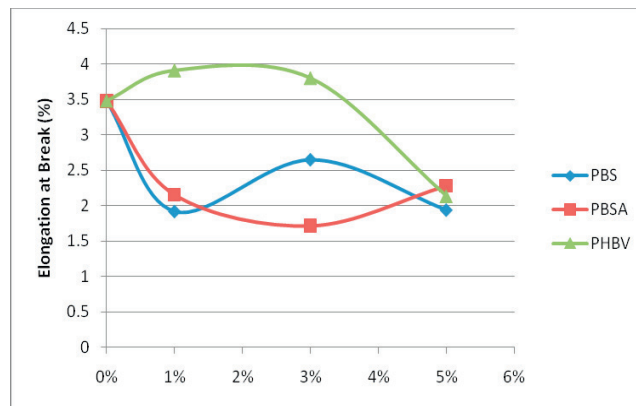


Fig. 3. Elongation at break of polymer blends PLA/PBAT with PBS, PBSA and PHBV 1,3,5 phr

Polymer blends with addition of PBS and PBSA show decrease in the elongation at break. With 1 and 3 wt% of PHBV, the elongation at break increased but decreased with 5 wt%. The small amount of PBS and PBSA may acts as nucleating agent and increased the crystallinity of the polymer components and results in phase separation and the decreased in elongation.

The modulus of the polymer blends of PLA/PBAT (70:30) with addition of 1,3,5 phr of PBS, PBSA and PHBV are shown in Fig. 4.

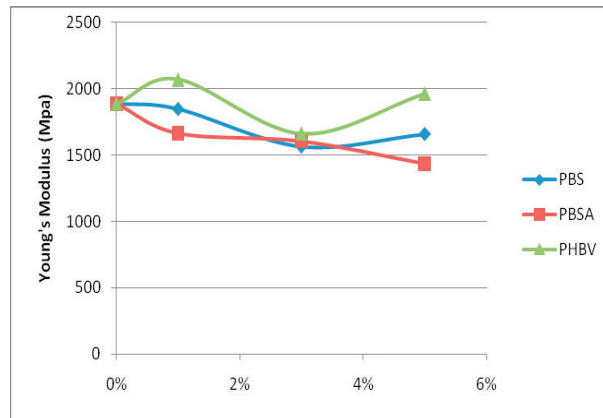


Fig. 4. Modulus of polymer blends PLA/PBAT with PBS, PBSA and PHBV 1,3,5 phr

Modulus of polymer blend with addition of 1 wt% PHBV was slightly increased while modulus of most of the polymer with addition of PBS and PBSA decreased. The results confirmed the phase separation of polymer components.

The impact test of polymer blends was analyzed and reported in Fig. 5.

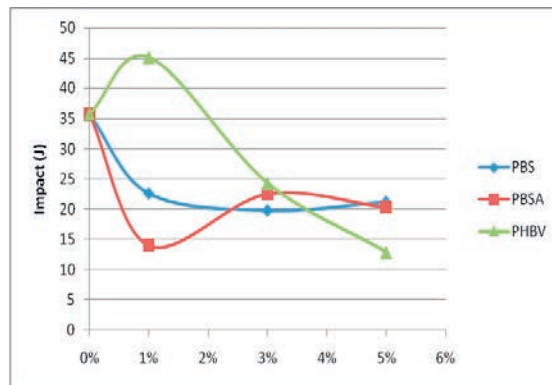


Fig. 5. Impact strength of polymer blends of PLA/PBAT with PBS, PBSA and PHBV 1,3,5 phr

The impact strength of polymer blends with PHBV 1 phr increased but decreased when added more amount of compatibilizer. PBA and PBSA decreased the impact strength to all components.

#### 4. Conclusion

Biodegradable polymers, poly(butylene succinate) (PBS), poly(butylene adipate-co-terephthalate) (PBAT), and poly(hydroxybutyrate-co-valerate) (PHBV) were used as compatibilizer for polymer blends of poly(lactic acid) (PLA) and poly(butylene adipate-co-terephthalate) (PBAT). The ratio of PLA:PBAT studied was 70:30 by weight. The amounts of PBS, PBSA and PHBV as compatibilizer were 1, 3 and 5 phr. The thermal property of polymer blends slightly decreased while melting temperature ( $T_m$ ) of the compositional polymers were almost not changed which indicated the phase separation of compositions. The melt flow of polymer blends increased with increasing amount of PBS while it was not changed with addition of PBSA and PHBV. Modulus decreased with 10 phr of PBS, PBSA and PHBV but stable when increased the amount of biodegradable polymers.

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