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## Material properties

# Properties of immiscible and ethylene-butyl acrylate-glycidyl methacrylate terpolymer compatibilized poly (lactic acid) and polypropylene blends

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## ABSTRACT

Poly(lactic acid) (PLA) and polypropylene (PP) blends of various proportions were prepared by melt-compounding. The miscibility, phase morphology, thermal behavior, and mechanical and rheological properties of the blends were investigated. The blends were immiscible systems with two typical morphologies, spherical droplet and co-continuous, and could be obtained at various compositions. Complex viscosity, storage modulus and loss modulus depend on the PP content. Thermal degradation of all blends led to two weight losses, for PLA and PP. The incorporation of PP improved the thermal stability of the blend. The effect of compatibilizer (ethylene-butyl acrylate-glycidyl methacrylate terpolymer, EBA-GMA) on the morphology and mechanical properties of 70/30 w/w PLA/PP blends was investigated. The tensile strength of these blends reached a maximum for 2.5 wt% EBA-GMA, and impact strength increased with increasing EBA-GMA content, suggesting that EBA-GMA is an effective compatibilizer for PLA/PP blends.

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## 1. Introduction

Growing concerns about environmental protection and energy security, have led to biocompatible and biodegradable polymers attracting significant attention over recent decades [1–4]. Poly(lactic acid) (PLA) is a degradable, linear, aliphatic, thermoplastic polyester that can be derived from the starch of crops such as corn and sugar beet [5], and has been much-researched as a biodegradable polymer [6]. However, the mechanical and thermal properties of PLA are often inferior to those of conventional petroleum-based thermoplastic polymers, such as polypropylene (PP), polystyrene and polyethylene, thus limiting its applications [7–9].

Polymer blending is a practical, economical protocol to achieve a desirable combination of properties that are often absent in single-component polymers [10,11]. PLA blends with biodegradable or non-biodegradable polymers, and characterization of the blends with regard to miscibility, morphology, and mechanical and thermal properties, have been reported. Wu et al. [12] prepared PLA and polycaprolactone blends of various compositions via melt mixing, and observed three typical immiscible morphologies: spherical droplet, fibrous and co-continuous structures. Xiong et al. [13] prepared entirely biosourced PLA/starch blends with epoxidized soybean oil as a reactive compatibilizer, and studied the morphology, thermal properties and mechanical properties of the blends. Zou et al. [14] investigated the crystallization, hydrolytic degradation and mechanical properties of poly(trimethylene terephthalate) and PLA blends. Moreover, to reduce cost, Biresaw et al. [15]

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blended PLA with polystyrene and investigated the interfacial tension between the components.

Polypropylene is one of the cheaper synthetic polymers. Because of its many advantages, such as low density, low cost, high softening point and easy processing, it is widely used in flexible packaging and containers, as well as in engineering applications when reinforced [16–18]. The automotive industry, for instance, uses unfilled or filled PP for batteries and bumpers [19,20]. Hamad et al. [21] blended PLA with PP, and studied the rheological and mechanical properties. However, PP is non-degradable. Reddy et al. [22] also prepared PLA/PP fibers, and found that blending PLA with PP not only improves the resistance of PLA to hydrolysis, but also enhances the degradability of PP. Kim et al. [23] investigated the effects of compatibilizers and hydrolysis on the mechanical properties, interfacial tension, and morphology of PP/PLA (80/20 w/w) blends. They found that maleic anhydride-grafted polypropylene (PP-g-MAH) is an effective compatibilizer for improving the tensile strength of PP/PLA (80/20 w/w) blends, and that maleated styrene-ethylene/butylene-styrene (SEBS-g-MAH) is an effective impact-modifier for improving the impact strength of PP/PLA (80/20 w/w) blends.

In this study, immiscible PLA/PP blends of different compositions were prepared and studied. The effect of compatibilizer (ethylene-butyl acrylate-glycidyl methacrylate terpolymer) loading on the blends' morphology and mechanical properties was also investigated. Surprisingly, a co-continuous phase morphology was observed in the PLA/PP blend, which has never been reported before, and could lead to a number of specific end-use applications.

## 2. Raw materials

Commercial grade PLA (PLA 4032D), with glass transition temperature  $T_g \approx 60$  °C and melting temperature  $T_m \approx 170$  °C, was obtained from Natureworks, LLC (USA). The isotactic polypropylene sample (model T30S), with a melt flow rate of 3 g/10 min (230 °C, 21.6 N),  $M_w = 39.9 \times 10^4$  g mol<sup>-1</sup>, and  $M_w/M_n = 4.6$ , was purchased from Maoming Petrochemical Co., Ltd. (Guangdong, China). Ethylene-butyl acrylate-glycidyl methacrylate terpolymer pellets (Elvaloy PTW, designated 'EBA-GMA') with a melt index of 12 g/10 min (190 °C, 2.16 kg), were provided by DuPont Co.; its E/BA/GMA monomer ratio is 66.75/28/5.25 (w/w/w).

## 3. Experimental

### 3.1. Preparation of PLA/PP blends and PLA/PP/EBA-GMA blends

Before melt blending, the PLA and PP pellets were dried in vacuum at 80 °C for 4 h. PLA/PP blends of different compositions (100/0, 90/10, 70/30, 50/50, 30/70, 10/90, 0/100) were prepared in a Brabender counter-rotating twin-screw extruder (Germany) with a screw diameter of 25 mm and a length/diameter ratio of 20:1. The profile temperatures were 170, 190, 205, 205, 205, 210, 210, and 210 °C, and the screw speed was 60 rpm. To investigate the effect of compatibilizer (EBA-GMA) on the morphology and

properties of the blends, PLA/PP/EBA-GMA blends were melt-compounded using the same process parameters. All extruded blends were immediately cooled in a water bath, and a pelletizer was used to cut the extruded blends into pellets. These pellets were dried at 80 °C for more than 4 h. After being oven-dried, the extruded pellets were injection-molded into ISO527-standard specimens (EC-75N-II, TOSHIBA Machine Co. Ltd., Japan). The temperature profile of the injection barrels was 185–200–210–210 °C from the first heating zone to the nozzle. The injection pressure was set at 50 MPa. All test specimens were conditioned for 7 days at 25 °C and 50% relative humidity prior to testing and characterization.

### 3.2. Characterization

The fracture surfaces of the blends were studied using a scanning electronic microscope (SEM, Quanta 200 FEI Co.). Specimens (4-mm thick) were submerged in liquid nitrogen for ~ 15 min and fractured to expose the internal structure. Before recording the morphological observations, the sample surfaces were sputter-coated with Au to prevent build-up of electrostatic charge during observations.

Melt rheological behavior of the blends was studied using a MCR302 rheometer (Anton Paar, Austria) in dynamic oscillation mode with a parallel-plate geometry (diameter = 25 mm, gap = 1 mm). Frequency-sweep tests were performed for 0.01–100 rad s<sup>-1</sup> at 210 °C. To maintain the response of materials in the linear viscoelastic regime, an amplitude of 1% was used.

Thermogravimetric analysis (TGA) was performed on  $\approx 10$  mg samples using a Netzsch TG209 instrument over 30–500 °C in a N<sub>2</sub> atmosphere (250 ml/min) with a 10 °C min<sup>-1</sup> heating ramp. All thermal parameters were determined as an average of three experiments.

Dynamic mechanical analysis (DMA) was performed using a Netzsch DMA242c instrument at a fixed frequency of 3 Hz and oscillation amplitude of 0.15 mm. The temperature range studied was –40 to +100 °C with a heating rate of 3 °C min<sup>-1</sup>.

Tensile tests to determine the modulus, yield strength, and elongation at breaking were carried out using an Instron universal machine (model 5566, USA), in accordance with ISO527, under tension mode at a single-strain rate of 20 mm min<sup>-1</sup> at room temperature. An Instron POE2000 pendulum impact tester was used for impact testing.

## 4. Result and discussion

### 4.1. Miscibility and phase morphology of PLA/PP blends

Fig. 1 shows the dynamic mechanical traces of PLA/PP blends of various compositions. Plots of dynamic loss (tan  $\delta$ ) as a function of temperature for pure PLA, pure PP and PLA/PP blends are given in Fig. 1(a–c). Pure PLA and pure PP show glass transition peaks at 69.2 °C and 15.3 °C, respectively. The glass transition temperature ( $T_g$ ) of the PLA/PP blends, obtained from the tan  $\delta$  peak temperature, are summarized in Table 1. As observed from Fig. 1(a) and Table 1, two glass transition peaks are obtained for each blend,

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