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Influence of poly(butylenes adipate-co-terephthalate) on the properties of the biodegradable composites based on ramie/poly(lactic acid)

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ABSTRACT

Ramie/poly(lactic acid) (PLA) composites were prepared by using two-roll mill. Poly(butylenes adipate-co-terephthalate) was added into the composites, and the effect of PBAT on the mechanical properties and thermal properties of the composites was investigate. The tensile and flexural strength decreased with the increase of PBAT content, and the addition of a small amount of PBAT increased the impact toughness. Microscopic study of the fracture surfaces revealed deteriorated interfacial adhesion between the fiber and the matrix in presence of PBAT. DSC results showed that PBAT lead the glass transition temperature (T_g) and the percentage crystallinity (X_c) decrease. Thermogravimetric analysis (TGA) thermograms showed improved thermal stability as compared with ramie/PLA composites.

1. Introduction

In recent years, interest has been growing in developing biopolymers to reduce green-house gas emissions and to conserve petroleum resources [1–3]. Natural fiber (NF) was widely used as reinforcement to replace the conventional inorganic fibers in polymer matrix composites due to their possessing many advantages such as low weight, recyclability, biodegradability and high specific strength and modulus. [4–6]. Natural fibers have different origins such as ramie, jute, hemp, sisal, and flax [7]. These fibers are mainly made of cellulose, hemicelluloses, lignin and pectins, with a small quantity of extractives [8]. The exact percentage of each chemical composition depends on the nature of the plant, location in which it is grown, age of the plant and extraction methods, etc. During the last decade, many research activities have been focusing on the natural fiber reinforced thermoplastic composites, especially polypropylene (PP) matrix composites are gaining increasing interest. As a result, natural fiber/PP composites have acceptable properties and have found wide use in the automobile industry.

Many studies have been made to use renewable polymer as matrix for natural fibers [9,10]. Poly(lactic acid)(PLA) as a linear aliphatic thermoplastic polyester produced from renewable resources has received much attention, which is produced either by ring-opening polymerization of lactide or by polycondensation of the lactic acid monomers, and the monomer is obtained from the fermentation of corn etc. [11,12]. For many applications, the mechanical properties of PLA such as tensile strength and stiffness can be much improved by natural fibers at lower cost, but the brittleness of PLA and NF/PLA composites limit their applications. Plackett et al. [13] determined the mechanical properties of the PLA/jute fiber composites with the results showing that the tensile strength of composites was significantly higher than that of PLA. But according their results the elongation at break of the composites is still very low at about 2%.

Poly(butylenes adipate-co-terephthalate) (PBAT) is an aliphatic–aromatic copolyester, which is fully biodegradable [14,15]. And PBAT is a flexible plastic designed for film extrusion and extrusion coating which degrades within a few weeks by the aid of naturally occurring enzymes. In the view of its high toughness and biodegradability, PBAT is considered as a good candidate for the toughening of PLA [16,17]. PLA/PBAT blends were studied by Jiang et al. [18] The blends showed decreased tensile strength and modulus, however, elongation and toughness were dramatically increased. The failure mode changed from brittle fracture of the neat PLA to ductile fracture of the blends.

The aim of this study was to toughen PLA matrix by using PBAT and achieve ramie/PLA/PBAT ternary composites. The influence of PBAT on the mechanical and thermal properties of ramie/PLA composites was investigated.

2. Experimental

2.1. Materials

Poly(lactic acid)(PLA) (M_w = 140,000) was supplied by Shanghai Tong-Jie-Liang Biomaterial Co. Ltd., China. Ramie fiber was purchased from Jinlan Ramie Fiber Co. Ltd., China. Ramie fiber

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Table 1
Raw materials used for composites.

Materials	Density (g/cm^3)	Melt temperature (°C)	Fiber length (mm)
PLA	1.25	165	–
PBAT	1.26	115	–
Ramie	1.35	–	10

Table 2
Formulations of the composites.

Materials	PLA (wt%)	PBAT (wt%)	Ramie (wt%)
PLA	100	–	–
Ramie/PLA	70	–	30
Ramie/PLA/PBAT-5	66.5	3.5	30
Ramie/PLA/PBAT-10	63	7	30
Ramie/PLA/PBAT-15	59.5	10.5	30

(diameter: about 20 μm)was cut to 10 mm in average length. Firstly, ramie fibers were immersed in sodium hydroxide solution (5% w/v) for 3 h at room temperature. Then the fibers were washed with distilled water containing a few drops of acetic acid. Next, the fibers were washed with distilled water until alkalinity is no longer detected. Finally, the fibers were dried in air at 80 °C for 72 h. Poly(butylenes adipate-co-terephthalate) (PBAT, $T_g = -29$ °C, $T_m = 115$ °C, Ecoflex F BX7011) was supplied by BASF Company. The properties of PLA, PBAT and ramie were shown in Table 1.

2.2. Preparation of PLA/PBAT blends

To prepare ramie/PLA/PBAT composites, a two-step process was used. Firstly, PLA/PBAT blends as the intermediate products were prepare by using a twin-screw extruder. Then PLA/PBAT blends and ramie were mixed using a two-roll plastics mill. The processing of PLA/PBAT blends, ramie/PLA composite and ramie/PLA/PBAT composites were shown in Fig. 1.

PLA/PBAT blends were prepared by melt mixing 85–95 wt% PLA and 5–15 wt% PBAT using a twin-screw extruder with a screw diameter of 27 mm and an L/D ratio of 42. Before extrusion, both PLA and PBAT pellets were dried under vacuum at 80 °C for 12 h. The extrusion temperature was independently controlled at eight zones along the extruder barrel and a strand die to achieve a temperature profile in the range of 155–175 °C. The screw speed was set at 100 rpm and feed rate was 30 g/min.

2.3. Composites preparation

The ramie fibers and PLA/PBAT blends were mixed using a two-roll plastics mill at 140 °C for 5 min. The volume percent of the ramie fibers was 30%. Formulation of the composites prepared for this study is given in Table 2. The composites obtained were then molded into sheets by hot pressing at 170 °C and 20 MPa for 4 min followed by cooling to room temperature at 5 MPa. The sheets were prepared for structure characterization and properties measurement.

2.4. Characterization

2.4.1. Mechanical properties measurement

The tensile properties of the composites were tested according to GB 13022-91 standard using a CMT5105 Machine (Shenzhen Sansi Material Instruments Ltd., China). Crosshead speed was set at 5 mm/min.

The flexural properties of the composites were tested under three point bending in a DXLL-5000 machine (Shanghai Jiedeng Instruments Ltd., China) in accordance with GB 1449-83. The size of the flexural testing samples used was 65 mm × 10 mm × 3.5 mm. The machine was operated at a crosshead speed of 1.2 mm/min and a span of 60 mm.

The impact strength of the composites was determined from the specimens having dimensions of 65 mm × 10 mm × 3.5 mm. The test was carried out in a XCJ-50 test machine (Chengde test Instruments Ltd., China) according to GB 1451-83.

At least five specimens were used for each condition.

2.4.2. SEM

The morphologies of the impact fractured surfaces of the composites were observed and analyzed by scanning electron microscope (SEM) (Quanta 200 FEG, FEI Company) at room temperature. The samples were coated with gold using a vacuum sputter coater. The samples were viewed perpendicular to the fractured surface.

2.4.3. DSC

Thermal properties of the composites were analyzed using TA Q20 Differential Scanning Calorimetry (DSC) thermoanalyzer. The

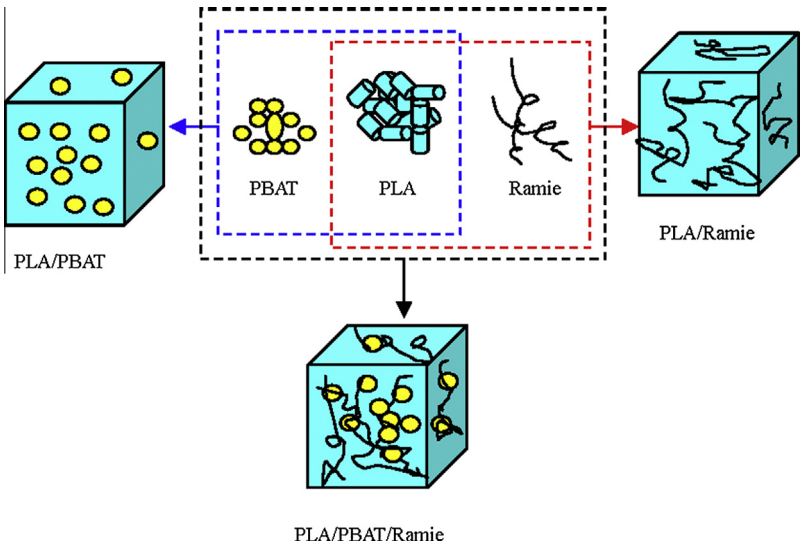

Fig. 1. Manufacture process of ramie/PLA composites without and with PBAT. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)