



Poly (lactic acid) fibre reinforced biodegradable composites



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ABSTRACT

This study focuses on the development of fully biodegradable poly (lactic acid) (PLA) fibre reinforced PLA and poly (butylene succinate) (PBS) matrix composites in order to reduce the impact on the environment by long-lasting plastics based composites. PLA self-reinforced composite (SRC) and the PLA reinforced PBS composite (PLA–PBS) composites were produced by film-stacking method. With the increasing of the loading direction fibre content, the composite moduli significantly increase by between 70% up to 6 times higher than those of the matrix films. Higher tensile strength and Young's modulus have been found in PLA–SRC than in PLA–PBS composite. This is due to better tensile properties of the PLA film and better interfacial adhesion in PLA–SRC.

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1. Introduction

1.1. Background

Composite materials are used in an ever increasing range of applications, including airplanes and automobiles. However, most of the commercial composites are carbon or glass fibre reinforced epoxy composites which are fossil fuel based synthetic materials. Due to the difficulties of recycling and disposing these materials at the end their life cycle and limitation of fossil fuel, a series of new regulations has been introduced to reduce their impact on the environment. Greater efforts are being made to seek more eco-friendly materials which could replace fossil fuel based polymeric materials. One solution is to replace the synthetic reinforcing material with natural fibres such as bast fibre. However, most of these natural fibre reinforced composites still use non-biodegradable petroleum-derived polymer matrix, such as polypropylene (PP) and polyethylene (PE). In order to produce fully biodegradable composites, there have been increasing interests in the development of composites based on biodegradable matrix materials such as poly (lactic acid) (PLA) and poly (butylene succinate) (PBS).

1.2. Biodegradable polymer and composite

PLA, a thermoplastic polymer, is derived from renewable resources that contain rich carbohydrates, such as core and sugar cane [1–3]. Because of its good biodegradability and biocompatibility, PLA has been used in biomedical materials for many years. Similar to PLA, PBS is a biodegradable thermoplastic polymer and produced by polycondensation of glycol and succinic acid, which are available from bio-based renewable resources [3–6]. Because of the high market demand of biodegradable based products in recent years, both PLA and PBS based products are manufactured commercially in many countries. Their applications include film, food packaging, textiles, disposal bottle and tableware [1,2,7–10]. Because PBS has a lower melting temperature (100–120 °C) than PLA [11,12], it provides easier composite processing conditions in the hot compression process.

The attempts of reducing the weight and cost, as well as improving the sustainability of a vehicle have led to more attention on seeking alternative sustainable materials in the automobile industry. Natural fibre reinforced composites have become the best candidates, owing to their low density, cheap resource and comparable mechanical properties. Recently, natural fibre reinforced composites have been used widely as interior and exterior materials with multiple functions in vehicles [13–15]. Several studies [16–19] have found that natural fibre/PLA composites had higher mechanical properties, especially in tensile modulus, compared to natural fibre/PP or PE composites.

A large number of studies have focused on replacing non-biodegradable polymer matrix with PLA to create fully biodegradable

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composites by using various natural fibres. Bax and Müssig [20] showed flax/PLA and cordenka/PLA composites had higher Young's modulus than pure PLA. Bajpai et al. [17] and Granpner et al. [21] produced a range of natural fibre (cotton, hemp, kenaf, sisal, nettle and man-made cellulose fibres) reinforced PLA composites by compression moulding. Most of the composites showed higher tensile properties than pure PLA. However, poor interfacial bonding have been found to be a limitation between PLA matrix and other natural fibres such as jute [22,23], hemp, [24–26], kenaf [19,27–29] although the mechanical properties of these natural fibre/PLA composites can be improved by the optimization of the fibre–matrix interactions by adding adhesion promoters, coupling agents or plasticisers [24,25,29]. On the other hand, Wu [30] found modified PLA matrix improved the adhesion between sisal fibre and PLA matrix, while deteriorate the biodegradation rate when the resulting composite was buried in soil.

There is also a considerable amount of literature published on a wide range of natural fibre reinforced PBS biodegradable composites, including jute [4,5], silk [8,31], kenaf [32] bamboo [6,33], and bio-flour [34,35]. For PBS matrix composites, Liu et al. [4] found that the poor interfacial adhesion in untreated jute/PBS composites could be improved by modifying jute surface. Liu et al. [5] also found that the biodegradability of jute/PBS composite was reduced after fibre surface modification by alkali and coupling agent. Similarly, Lee and Wang [6] showed that adding bio-based coupling agent to bamboo fibre reinforced PLA/PBS composites led to better mechanical properties but lower enzymatic degradability.

A strong and stable interface between the fibre and the matrix could result in high mechanical properties of a composite. However, in a traditional fibre reinforced composite, the fibre/matrix chemical difference, and the distinct surface energies and properties limit the fibre/matrix interfacial adhesion [36]. Moreover, due to rapid growth in the demand of reinforced composites, the disposal and recycling of these composites at their end-of-life have become serious concerns [37]. The recycling process of a reinforced composite usually requires advanced technology and high cost due to the difficulty in separating the fibre from the polymer matrix [38].

1.3. Self-reinforced composite

Capiati and Porter [36] in 1975 first introduced the concept of self-reinforced polymer composites (SRC) to solve the problems in traditional composites. They defined self-reinforced composite as a composite in which the matrix and reinforcement are made from similar or identical polymer but in different morphologies. The concept of SRC is widely used for thermoplastic polymer composites because thermoplastic polymers have characteristics of reversible and rapid processing, and higher toughness and durability. Most of the earlier SRC studies [39–41] used PE owing to the high modulus of the ultra-high molecular weight PE fibre. The high modulus of the all-PE composite led to a wide range of applications, but the low melting temperature (about 110 °C) of PE restricts utilization under high temperature conditions. In the last decade, polypropylene (PP) polymer has gradually become an important polymer for SRC because of its relatively high thermal stability [42–48]. For even higher temperature applications, poly (ethylene terephthalate), with a melting temperature of approximate 260 °C, is becoming more attractive [38,49]. However, none of these SRC composites is biodegradable.

In this study, the biodegradable polymer, PLA, is used to create a biodegradable self-reinforced composite, combining the concepts of self-reinforcement composite and biodegradable composite. The production of SRC usually requires a large difference in melting temperature of matrix and fibre, around 15 °C, to allow the matrix to melt sufficiently and to maintain the mechanical properties of

oriented fibre. The lower limit of the processing temperature window is usually the minimum temperature required for the onset of adhesion for SR composite; and the upper limit refers to the onset of the melting peak of the fibre [49]. The melting temperature difference between the two PLA polymers is achieved by using highly oriented PLA fibres (semi-crystalline) as the reinforcement, and amorphous PLA film as the matrix. PBS, which has lower melting temperature, is also employed as the matrix of the PLA fibre reinforced PBS composite to compare with the PLA self-reinforced composite. The two types of PLA fibre reinforced composites are produced by film stacking following by hot pressing. The mechanical performances of the composites are assessed. The interfacial adhesion between PLA fibre and the PLA/PBS matrix is also examined.

2. Materials and methods

2.1. Materials

An untwisted PLA filament yarn was used as the reinforcement of the biodegradable composites. Two matrix systems were used; one is a PLA film and the other is a PBS film. The details of the materials are given in Table 1. To evaluate the effects of composite manufacturing conditions on the tensile properties of the PLA fibre, PLA film and PBS film, these raw materials were processed under the same conditions of composite manufacturing and their properties were tested before and after the processing for comparison.

2.2. Differential scanning calorimeter (DSC) and wide-angle X-ray diffraction (WAXD)

The successful production of a thermoplastic composite by the hot pressing method is dependent on the capacity of the film matrix to flow around and satisfactorily bond to the fibres without notable effect on the fibre physical properties. Thus, an appropriate processing temperature should be based on the thermal properties of fibre and matrix. The processing temperature for composite manufacturing is usually equal to or higher than the melting point of the matrix, but below that of the fibre. To obtain a suitable processing temperature, DSC is carried out by using a calibrated TA Instruments DSC Q100 for the fibre and matrix materials. Samples from the PLA yarn, PLA and PBS films were cut into small pieces (about 1–2 mg) and sealed in hermetic pans and lids. All the samples experienced a heat-cool cycle with a heating and cooling rate of 10 °C/min. A Philip X'pert X-ray diffractometer (copper K α , 1.5 Å) was employed to evaluate the crystallinity of the PLA fibre, PLA film and PBS film.

2.3. Tensile testing

Instron 4411 with a load cell of 0.5 kN was used to test the tensile properties of untreated and treated PLA yarn at a constant crosshead speed of 250 mm/min (ISO 2062). Five dumb-bell shaped untreated and treated PLA and PBS films were tested at 2 mm/min for PLA film and 5 mm/min for PBS film (ISO 527). PLA fibre reinforced composites tensile testing was carried out at crosshead speed of 2 mm/min using Instron 5569 equipped with 1 kN load cell at room temperature (ISO 527). The dimensions of the stripe specimen were 25 mm \times 200 mm \times (0.17–0.29)mm.

2.4. Scanning electron microscope (SEM)

The cross-sections and the tensile fracture surfaces of the composites were examined using a Zeiss EVO 50 scanning electron microscope. To reduce the charging effect, carbon coating was

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