



# Study on the effect of dispersion phase morphology on porous structure of poly (lactic acid)/poly (ethylene terephthalate glycol-modified) blending foams



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## ABSTRACT

A methodology for blending foam of poly (lactic acid) (PLA)/poly (ethylene terephthalate glycol-modified) (PETG) was proposed. PLA/PETG blends were prepared through a melt blending method, using multiple functionality epoxide as reactive compatibilizer. The effects of blending ratio and compatibilizer content on the dispersion morphology, molecular structure, mechanical properties, and rheological behavior of PLA/PETG blends were studied. Then PLA/PETG blends were foamed using supercritical CO<sub>2</sub> as physical blowing agent, and their porous structure, pore size, as well as pore density were investigated. The results showed that the mechanical properties and rheological parameters such as melt strength and melt elasticity, as well as the porous structure of the foams dispersion morphology of PLA/PETG blends were affected strongly. The melt elasticity of PLA/PETG blends increased with increasing compatibilizer content. Dispersion phase morphology of PLA/PETG blends also had a significant effect on the pore density of all the samples. The results indicated that homogeneous and finer porous morphology of PLA/PETG foams with high expansion ratio could be achieved with a proper content of compatibilizer in the blends.

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## 1. Introduction

The research of polymer blends had attracted increasing attention because of the following advantages [1–8]. First, it could balance the properties of various polymeric components and eliminate the weakness on some properties of one polymeric component. Second, it could improve the processability and be helpful for developing a lot of novel polymer products. Third, the blending method was able to reduce the cost of developing new polymer. As most commonly used biopolymers, poly (lactic acid) (PLA) is often blended with other polymers to reduce the cost and overcome the disadvantages of PLA, such as low glass transition temperature ( $T_g$ ), poor toughness, and poor melt strength, which limit its applications [9–11].

Due to the poor compatibility and the inferior mechanical property of common PLA blends, compatibilizer was often used in the blending process to improve the properties. Wang et al.

prepared PLA/poly ( $\epsilon$ -caprolactone) (PCL) blends by melt blending method using triphenyl phosphite as reactive compatibilizer. They found that triphenyl phosphite was an effective coupling agent in the reactive compatibilization of PLA and PCL, and the elongation at break of blends was obviously improved [12]. Zhang et al. fabricated PLA/PBAT blends by twin screw extrusion using glycidyl methacrylate (GMA) as reactive processing compatibilizer. The results revealed that the rheological properties, blending morphology, and miscibility of PLA/PBAT blends could be improved by the introduction of GMA, resulting in increased tensile toughness of blends [13].

Besides these, in order to further efficiently improve the toughness of PLA and widen the application of PLA, efforts were continuously made to research PLA foaming technology, which was a well-known process to enhance the ductility and impact resistance by providing a significant expansion ratio and weight reduction [14,15]. The high expansion ratio induced by foaming generally could reduce the material cost and consumption in mass-produced plastic parts without significantly compromise in other properties.

In PLA foaming process, inorganic particle was usually considered as a good porous nucleation sites, which could enhance the

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pore nucleation number; thus it could improve porous structure as well as the properties of PLA foam [16,17]. In reality, the poor compatibility and dispersibility of inorganic particles in PLA matrix were easily resulted in the non-uniform pore size and distribution, finally affecting the comprehensive performance of PLA foam. However, for the polymeric blends, the dispersion phase could also act as an efficient porous nucleation agent since it would decrease the activation energy of porous nucleation. Unlike the inorganic particle, dispersion phase in polymer blends characterized with better compatibility and dispersibility in matrix phase. Consequently, the finest micro-size pores and uniform porous structure could be obtained by tunable dispersion morphology.

So far, there is little literature reported about PLA blending foam using the dispersion phase as heterogeneous bubble nucleation agent in its blends, and the nucleation mechanism in the interface of polymer blends. In this paper, poly (ethylene terephthalate glycol-modified) (PETG) was blended with PLA. PETG is an amorphous random copolymer, and has a glass transition temperature around 80 °C. Mechanical properties of the PETG are close to those of PET. This advantage makes the PETG has noticeable toughness, flexibility, and high processability. PLA blending with PETG can be effective to overcome the drawbacks of PLA, resulting in PLA/PETG blends with good toughness and flexibility, which can be widely used in packaging industries. In this blending, PETG acting as organic dispersion phase was expected to replace the inorganic filler as heterogeneous bubble nucleation agent and impact modifier in foaming process, due to its noticeable toughness, high processability, and well compatibility with PLA [18–20]. Compared with inorganic filler, organic dispersion phase could be dispersed better in PLA and possessed smaller size and increased quantity, leading to abundant interface between PLA and PETG that was potential to become the pore nucleation source. In addition, epoxy-based compatibilizer was used to improve the compatibility of PLA/PETG blends. The aim of this work was to investigate the effect of tunable PLA/PETG blending morphology on the porous structure of its foam. The dynamic rheological properties and mechanical properties were also studied.

## 2. Experimental

### 2.1. Materials

The biodegradable linear PLA (2002D) with the melt flow rate of 5.6 g/10 min was purchased from NatureWorks. It's a semi-crystalline polymer containing approximately 4% D-lactic. PETG (S2008) with number-averaged molecular weight ( $M_n$ ) of 26,000 g/mol, intrinsic viscosity of 0.78 dl/g, and  $T_g$  of 80 °C was supplied by SK chemical corp (Seoul, South Korea). Reactive compatibilizer (CESA extend BL 10069 N), containing 10 wt% multi-functional epoxide in a PLA matrix resin, was supplied by Clariant Chemical Company, USA. The multi-functional epoxy-based compatibilizer was a styrene-acrylic oligomer with  $M_n$  less than 3000, and its' average functionality was more than four. The chemical structure was showed in Fig. 1.

### 2.2. Preparation of PLA/PETG blends

The PLA/PETG and compatibilized-PLA/PETG blends were prepared in a Haake internal mixer at 190 °C, with a mixing time of 10 min and mixing speed of 50 rpm. Prior to melt mixing, PLA, PETG, and compatibilizer were dried in an oven at 60 °C for 12 h to remove excess moisture. Afterward, the blending samples were compression molded into sheets of 2 mm in thickness for subsequent characterizations and solid-state batch foaming. The samples formula was shown in Table 1. All components used the unit of "parts per hundred resin" (phr) for their formula.

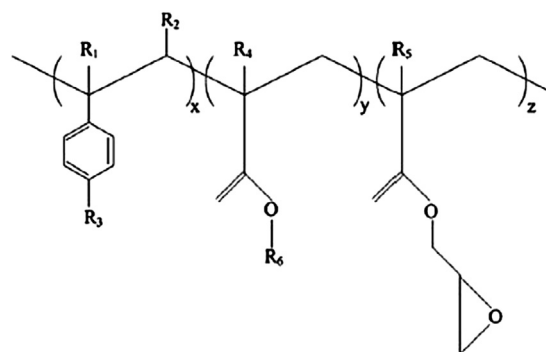


Fig. 1. The chemical structure of multi-functional epoxy-based compatibilizer.

### 2.3. Batch physical foaming

The PLA/PETG blend foams were prepared by the batch foaming method using supercritical CO<sub>2</sub> as physical blowing agent. In order to investigate the differences of foaming behaviors and dispersion phase morphologies of various PLA/PETG blends and compatibilized-PLA/PETG blends, the samples were prepared and foamed under the same conditions. First, the samples were put into autoclave at a temperature of 170 °C and a high pressure of 20 MPa for 4 h. After the CO<sub>2</sub> was fully diffused and dissolved in the polymer melt matrix, the samples were cooled to the foaming temperature of 130 °C. An instantaneous pressure drop by release of CO<sub>2</sub> from 20 MPa to 0.1 MPa provided the driving force for porous nucleation and growth. At last, the foaming samples were prepared for further characterizations.

### 2.4. Characterizations

#### 2.4.1. Dynamic rheometer

Dynamic rheological behaviors of various samples were tested using a strain-controlled rheometer (ARES Rheometer, TA, USA) at 190 °C, with parallel plates (20 mm in diameter with a gap of 1.0 mm). The frequency range was 0.1–100 rad/s, and the maximum strain was fixed at 5%, to confirm that these conditions were within the linear viscoelastic region under nitrogen. The complex viscosity ( $\eta^*$ ), storage modulus ( $G'$ ), and loss modulus ( $G''$ ) were measured at various frequencies.

#### 2.4.2. Fourier transformation infrared spectroscopy (FTIR)

Infrared spectra of compatibilized-PLA/PETG blends were obtained using an FTIR (Nicolet iS10 thermo scientific spectrometer) in transmission mode. Each spectrum was obtained within the range of 3800–480 cm<sup>-1</sup> with a wavelength resolution of 4 cm<sup>-1</sup>.

#### 2.4.3. Differential scanning calorimetry (DSC)

The glass transition regions of blend samples were characterized by a DSC (Q100, TA, USA) purged with nitrogen. The samples were heated to 200 °C for 5 min to remove the heating history. Then, the

Table 1  
The weight ratio of each component of the various PLA/PETG blends.

Serials No.	PLA/phr	PETG/phr	Compatibilizer/phr
1	90	10	0
2	80	20	0
3	70	30	0
4	80	20	1
5	80	20	3
6	80	20	5
7	80	20	7

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