



Toughening modification of polycarbonate/poly(butylene terephthalate) blends achieved by simultaneous addition of elastomer particles and carbon nanotubes



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ABSTRACT

Small quantities of maleic anhydride grafted styrene-ethylene-butylene-styrene (SEBS-g-MAH) copolymer and carbon nanotubes (CNTs) were introduced into polycarbonate (PC)/poly(butylene terephthalate) (PBT) blends. The results demonstrated that simultaneously adding SEBS-g-MAH and CNTs greatly enhanced the fracture toughness of the samples and the impact strength increased with increasing CNT content. The morphologies, the dispersion of CNTs, the relaxation behaviors and the crystallization behaviors of samples were systematically investigated. SEBS-g-MAH formed the dispersed particles in the system. The particle diameter was decreased in the blend composites. CNTs exhibited homogeneous dispersion in the blend composites and they also formed a percolated network structure at relatively high content. The transesterification between PC and PBT components was suppressed by SEBS-g-MAH, and the crystallization ability of the PBT component was greatly enhanced. The toughening mechanisms were mainly related to the suppressed transesterification, the decreased elastomer particle size, and the formation of a CNT network structure.

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1. Introduction

Polymer blending has already been demonstrated an efficient way to prepare new materials with promising chemical and physical properties. Most of the polymer blends are immiscible due to their high molecular weight and weak interaction between molecular chains of the components. The weak interfacial interaction between components usually cannot prevent the initiation and propagation of cracks along the interface and consequently, the blends usually exhibit poor fracture toughness. Many strategies have been developed to further improve the mechanical properties of the immiscible blends. Among these strategies, the most widely used strategy is introducing a third component, such as a compatibilizer or nanofiller. The properties of the modified blends are influenced by many factors. For example, in the compatibilized blends, the mechanical properties depend not only on the compatibilizing efficiency and the content of the compatibilizer but also on the morphology of the blends [1–12]. However, in the blend composites, besides the determinable role of blend morphology,

the mechanical properties are also influenced by the dispersion and microstructure of nanofillers [13–15].

As one of the engineering plastics that exhibit good processing ability and good general properties, polycarbonate (PC) exhibits great potential application. So far, different kinds of PC-based articles have been widely used in many fields ranging from packages, toys, and electronic elements to automobile applications. PC-based blends have also been widely investigated since PC has been synthesized, and different kinds of PC blends have already been developed. Among these PC blends, the blends with poly(butylene terephthalate) (PBT) attract much attention from researchers [16–24]. PBT is a semicrystalline polymer with good chemical resistance and thermal stability. Therefore, the PC/PBT blends exhibit versatile properties, such as good chemical resistance, high heat resistance, and excellent esthetics and good flow characteristics. Different from other polymer blends without any chemical reaction between components, the most important characteristic of the PC/PBT blends is that there is a transesterification reaction between PC and PBT molecular chains during the melt compounding processing [16–18]. As a consequence, the intermolecular interaction between PC and PBT is enhanced and the blends exhibit good miscibility in the amorphous state. For example, the blends exhibit only one glass transition temperature (T_g) [12]. However,

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the progressive transesterification results in a change of initial homopolymers into block copolymers and finally into random copolymers [19–21]. On one hand, the crystallization ability of PBT component is greatly reduced [12], which deteriorates the chemical resistance and thermal stability of the blends. On the other hand, it has been demonstrated that the transesterification also reduces the fracture toughness of the blends [22]. Therefore, to obtain the PC/PBT blends with promising properties, the transesterification should be completely suppressed. Otherwise, other modification strategies should be developed to compensate the performance loss induced by the transesterification since the transesterification is hardly suppressed [23].

Introducing stabilizer or inhibitor is an efficient way to suppress the transesterification [21,24]. For example, Lei et al. [23] introduced silicone phosphate into PC/PBT blends. They found that the content of copolymer was decreased and the crystallization ability of PBT was greatly enhanced compared with the binary PC/PBT blends. Their observation demonstrated that silicone phosphate acted as the inhibitor to suppress the transesterification between PC and PBT components. However, Pompe et al. [18] demonstrated that the stabilizer could not completely suppress the transesterification because at the beginning the transesterification rate was higher than the reaction rate of stabilizer. Furthermore, they demonstrated that the annealing time of the thermal treatment in the melt compounding processing determined the degree of the transesterification.

Other strategies reported in the literature are mainly related to the introduction of the third component, such as rubber or impact modifier, compatibilizer and fillers. Wu et al. [22,25] intensively investigated the microstructure-performance relationship of the PC/PBT blends with impact modifier. Tseng et al. [9] introduced a special impact modifier (MBS) that consisted of three layers, in which the inner, middle and outer layers were styrene butadiene rubber (SBR) latex, polystyrene, and a copolymer of the methacrylic acid (MMA) and the functional group containing vinyl monomer, respectively. Bai et al. [10,11] investigated the effect of ethylene-butylacrylate-glycidyl methacrylate copolymer (PTW) and ethylene-1-octylene copolymer (POE) on the fracture toughness of PC/PBT blends. They found that the simultaneous addition of PTW and POE resulted in remarkable increases in the impact strength, elongation at break, and Vicat temperature. So far, the fillers that have been used to modify PC/PBT blends are mainly glass fiber (GF) [26,27] and talc [28]. For example, Khamsehnezhad et al. [26,27] introduced short GF into PC/PBT blends to improve the flexural and tensile strength of the blends. DePolo et al. [28] investigated the effects of particle size (nanotalc versus fine talc) and content on the dimensional stability and mechanical properties of PC/PBT. Wang et al. [29] introduced GF into the PC/PBT blends with a reactive elastomer, ethylene-co-glycidyl methacrylate (E-GMA).

In this work, we attempt to simultaneously introduce small quantities of elastomer, i.e. maleic anhydride grafted styrene-ethylene-butylene-styrene (SEBS-g-MAH) copolymer, and carbon nanotubes (CNTs) into the PC/PBT blends. CNTs have already been demonstrated an efficient impact modifier in some immiscible polymer blends, whose toughening mechanisms are related to the bridging effect of interfacial-localized CNTs that prevents the initiation and propagation of cracks along the blend interface and facilitates the stress distribution between components, and the formation of CNT network structure that also facilitates the stress transferring in the whole sample and avoids the stress concentration resulting in the failure of sample [30]. Jamaludin et al. [31] already demonstrated that SEBS-g-MAH was an efficient compatibilizer for the poly(ethylene terephthalate) (PET)/PC blend composites with halloysite nanotubes. They found that SEBS-g-MAH not only improved the fracture toughness of the blend composites

but also improved the dispersion of nanofillers. Here, the effects of SEBS-g-MAH and CNTs on the mechanical properties, phase morphologies of the blend composites and the crystallization ability of the PBT component are comparatively investigated. It is interesting to observe that the impact strength of the PC/PBT blend is greatly enhanced by simultaneous addition of SEBS-g-MAH and CNTs. The toughening mechanisms are then analyzed.

2. Experiments

2.1. Materials

All the materials used in the present work are commercially available. PC (S-2001R, with a viscosity-average molecular weight of 2.3×10^4 g/mol, a melt flow rate (MFR) of 7.5–10.5 g/10 min (300 °C/1.2 kg) and a density of 1.2 g/cm³) was purchased from Mitsubishi Engineering-Plastics Corp. PBT (SABIC VALOX 325-1001, with the MFR of 50 g/10 min (250 °C/2.16 kg) and the density of 1.31 g/cm³) was purchased from Saudi Basic Industries Corp. SEBS-g-MAH (FG1901GT, with the density of 0.91 g/cm³ and the MFR of 22 g/10 min (230 °C/5 kg)) was obtained from KRATON, USA. The grafting ratio of MAH was about 1.4–2.0 wt%. CNTs (TNIMC4, with carboxyl groups of about 1.55 wt%) were obtained from Chengdu Institute of Organic Chemistry, Chinese Academy of Science (Chengdu, China). The length of a single CNT and the outer diameter of the CNT were 10–20 μm and 10–30 nm, respectively. The purity of the CNTs was above 95%.

2.2. Sample preparation

Before sample preparation, all the pristine materials were dried in an oven set to 70 °C for 10 h. Samples were prepared through melt compounding processing, which was conducted on a twin-screw extruder SHJ-30 (Nanjing Ruiya, China). During the extrusion, the melt temperatures were set to 210 °C (hopper, region I), 230 °C (region II), 240 °C (region III) and 250 °C (region IV–VI) and 245 °C (extrusion head, region VII) as schematically represented in Fig. 1a, and the screw speed was maintained at 150 rpm. After being granulated, the pellets were injection-molded to obtain the standard bars with a thickness of 4.2 mm and a width of 10 mm. The injection molding was conducted on an injection-molding machine EM80-V (Chen Hsong Machinery, China), and the melt temperatures were set to 240 (hopper, region I), 250 °C (region II–III), and 245 °C (nozzle, region IV) as shown in Fig. 1b. The mould temperature was maintained at 25 °C. In this work, the weight ratio of the ternary PC/PBT/SEBS-g-MAH blend was maintained at 50/50/5, and the content of CNTs was varied from 0.5 to 2 wt%. The sample notation was defined as C50B50S5Cx, where x represented the content of CNTs. For comparison, the binary PC/PBT (50/50, wt/wt) and the ternary PC/PBT/SEBS-g-MAH were also prepared, and the sample notations were C50B50 and C50B50S5, respectively.

2.3. Mechanical properties

Tensile properties were measured using a universal tensile machine AGS-J (SHIMADZU, China) according to ASTM D638. The gauge distance was set to 50 mm and a cross-head speed of 50 mm/min was adopted. During the measurements, the load-displacement curves were firstly obtained, and then converted to the stress-strain curves according to the measurement standard. The fracture energy was determined from the integral area. The load was measured by the testing machine load cell while the displacement was obtained through an extensometer positioned within the gauge length. The notched Izod impact strength was

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