



Carbon nanotubes induced microstructure and property changes of polycarbonate/poly(butylene terephthalate) blend



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ABSTRACT

Polycarbonate/poly(butylene terephthalate) (PC/PBT) blends exhibit wide applications in many fields. However, the blends usually exhibit poor mechanical and thermal properties because of the transesterification reaction occurred between molecules of the blend components. In this work, a small quantity of carbon nanotubes (CNTs) was introduced into the PC/PBT blend to prepare the ternary blend composites with balanced macroscopic properties. The dispersion of CNTs in the blend composites and the microstructural changes induced by CNTs were comparatively investigated. The results demonstrated that CNTs exhibit relatively homogeneous dispersion in the blend composites. Specifically, at relatively high content, CNTs form the percolated network structure. The crystallization ability of the PBT component is greatly enhanced with the aid of the nucleation effect of CNTs. Mechanical property measurements showed that CNTs exhibit strengthening and toughening effects on the PC/PBT blend, especially incorporating 2 wt% CNTs leads to the dramatic increase of tensile ductility. Further results showed that the blend composites also exhibit excellent antistatic performance. It is believed that the simultaneously improved fracture toughness and antistatic performance endow the blend composites with more potential applications.

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1. Introduction

As one of the engineering plastics which exhibit good processing ability and good comprehensive properties, polycarbonate (PC) has wide applications in many fields ranging from consumption electronics, automobile, medical apparatus and instruments, and building to packages, etc. To further enlarge the application fields, different types of PC-based materials have been developed through chemical or physical modification methods. Among these methods, blending PC with other polymers, such as polyesters, polyolefins and elastomers, attracts much attention of researchers [1–6]. Due to its good chemical resistance and thermal stability, poly(butylene terephthalate) (PBT) is believed to be a promising modifier in improving the general properties of the PC-based materials [7–9]. For example, the PC/PBT blends exhibit versatile properties, such as good chemical resistance, high heat resistance, excellent esthetics and good flow characteristics, etc.

However, it is worth noting that the PC/PBT blends usually

exhibit poor fracture toughness [10], which greatly restricts the application of the blends in the area in which articles usually experience impact load condition. Different from the other polymer blends that the poor fracture toughness is mainly related to the poor interfacial interaction between components, it is suggested that the poor fracture toughness of the PC/PBT blends is mainly originated from the excessive transesterification reaction between PC and PBT molecular chains during the melt compounding processing [10]. Although the transesterification reaction facilitates the improvement of miscibility, the progressive transesterification results in a continuous change of initial homopolymers into block copolymers and finally into random copolymers [11,12], which greatly reduces the crystallization ability of the PBT component and consequently, leads to the great deterioration of chemical resistance and thermal stability of the materials.

There are two strategies which can be used to prepare the PC/PBT blends with promising properties. One is suppressing the transesterification between PC and PBT components during the melt compounding processing through incorporating stabilizer or inhibitor [12,13]. The other is introducing the third component, such as rubber or impact modifier, compatibilizer and fillers [14,15],

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etc. to compensate the performance loss induced by the transesterification since the transesterification is hardly suppressed. Sometimes, the third component also suppresses the transesterification between PC and PBT components [16]. However, it is worth noting that the researches relating to introducing fillers into the PC/PBT blend are mainly focused on studying the reinforcement effect of fillers in the blend composites and less attention has been paid to the toughening effect of fillers [15,17–19].

In this work, we attempt to introduce carbon nanotubes (CNTs) into the PC/PBT blend to prepare the blend composites with promising properties. Factually, to date, various kinds of blend composites modified by adding CNTs have already been prepared. Most of researches are focused on preparing the conductive composites [20–23] and a “double-percolation conductive mechanism” has been proposed to explain the largely enhanced electrical conductivity at relatively high CNT content. Other researches are focused on studying morphological changes of the blend composites induced by CNTs due to the variation of viscosity ratio between polymer components and the formation of CNT network structure in the composites [24–26]. Furthermore, CNTs have already been widely demonstrated to be an efficient nucleating agent in promoting the crystallization of semicrystalline polymers. Even in the miscible blend in which the crystallization of the crystalline component is greatly restricted by the other amorphous component, the presence of CNTs still promotes the crystallization of the crystalline component [27–29]. Specifically, it has been demonstrated that CNTs also exhibit a role of impact modifier and they can improve the impact toughness of some immiscible polymer blends, such as PC/poly(L-lactide) (PLLA) [30], polypropylene (PP)/ethylene-co-vinyl acetate (EVA) [31], etc. The toughening mechanisms are suggested to be related to the morphological change induced by adding CNTs, the bridging effect of CNTs at the blend interface which facilitates the stress transferring between components, and the formation of CNT network structure [32–34]. In this work, it is expected that the presence of CNTs can promote the crystallization of the PBT component on the one hand. This possibly results in the change of mechanical properties of the blend composites. On the other hand, the presence of CNTs in the blend composites possibly endows the material with excellent antistatic performance because of the intrinsic high electrical conductivity of CNTs. Obviously, it can be expected that if the promising mechanical and antistatic properties can be simultaneously achieved, the PC/PBT/CNT blend composites may exhibit more potential applications.

2. Experimental part

2.1. Materials

PC (S-2001R) was purchased from Mitsubishi Engineering-Plastics Corp. It had 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. PBT (SABIC VALOX 325-1001) was purchased from Saudi Basic Industries Corp. It had the MFR of 50 g/10 min (250 °C/2.16 kg) and the density of 1.31 g/cm³. CNTs (TNIMC4) were obtained from Chengdu Institute of Organic Chemistry, Chinese Academy of Science (Chengdu, China). The content of carboxyl groups was about 1.55 wt%. The length of a single CNT was about 10–20 μm and the outer diameter of the CNT was 10–30 nm. The purity of the CNTs was above 95%.

2.2. Sample preparation

Sample was prepared through melt-compounding processing which was conducted on a twin-screw extruder SHJ-30 (Nanjing

Ruiyu, China). During the extrusion processing, the melt temperatures were set at 210–230–240–250–250–250–245 °C from hopper to die and the screw speed was kept at 150 rpm. After being granulated, the pellets were injection-molded on an injection-molding machine EM80-V (Chen Hsong Machinery, China) to obtain a dumbbell specimen with a width of 10 mm and a thickness of 4.2 mm. During the injection-molding processing, the melt temperatures were set at 240–250–245 °C from hopper to nozzle and the mould temperature was maintained at 25 °C. For the blend composites, the weight ratio of PC and PBT components was maintained at 80/20 while the contents of CNTs were varied from 0.5 to 2.0 wt%. The sample notation was defined as C80B20Cx, where x presented the CNT content. For comparison, the binary PC/PBT blend (80/20, wt/wt) was also prepared, and the sample notation was defined as C80B20.

2.3. Transmission optical microscopy (TOM)

The dispersion of CNTs in the blend composites was characterized using a transmission optical microscope (TOM) (ZEISS, Germany). The sample film with a thickness of about 0.1 mm was prepared through compression-molding processing which was carried out at a melt temperature of 250 °C and a pressure of 5 MPa.

2.4. Scanning electron microscopy (SEM)

The phase morphologies of samples were characterized using a scanning electron microscope (SEM) FEI Inspect (FEI, the Netherlands), which was operated at a voltage of 20 kV. Before SEM characterization, the sample was cryogenically fractured in liquid nitrogen, and then the cryo-fractured surface was coated with a thin layer of gold.

2.5. Rheological measurements

Rheological measurements were conducted on a stress-controlled rheometer DHR-1 (TA Instrument, USA). The plate sample was prepared through compression-molding processing, which was carried out at a melt temperature of 250 °C and a pressure of 5 MPa. The plate sample had a diameter of 25 mm and a thickness of 2 mm. The measurements were carried out at a melt temperature of 250 °C and a frequency sweep of 0.01–100 Hz. All the measurements were carried out in nitrogen atmosphere.

2.6. Differential scanning calorimetry (DSC)

The crystallization and melting behaviors of samples were investigated using a differential scanning calorimeter (DSC) STA 449C Jupiter (Netzsch, Germany). A sample of about 8 mg was heated from 30 °C to 250 °C at a heating rate of 10 °C/min and maintained at 250 °C for 3 min to erase any thermal history, and then the sample was cooled down to 30 °C at a cooling rate of 5 °C/min. All the measurements were carried out in nitrogen atmosphere. The degree of crystallinity (X_c) of the PBT component was calculated according to the following relation:

$$X_c = \frac{\Delta H_m - \Delta H_{cc}}{\Delta H_m^0 \times \phi} \times 100\% \quad (1)$$

Where, ΔH_m was the value of fusion enthalpy measured through DSC, ΔH_{cc} was the cold crystallization enthalpy obtained during the DSC heating process, ΔH_m^0 was the fusion enthalpy of the completely crystalline PBT, and ϕ was the relative weight fraction of the PBT component in the samples. Here, ΔH_m^0 was taken as 142 J/g [35].

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