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Striking effect of epoxy resin on improving mechanical properties of poly(butylene terephthalate)/recycled carbon fibre composites

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ABSTRACT

Recycled carbon fibre (RCF) reinforced poly(butylene terephthalate) (PBT) composites with high mechanical properties have been prepared through melt compounding. Here three epoxy resins (EP) with different molecular weights and epoxy values were used as in-situ coupling agent to improve the interfacial adhesion between RCF and PBT. The morphologies of impact fractured surfaces were studied to compare the effects of three kinds of EP on the interfacial adhesion between RCF and PBT. The optimal strengthening effect was achieved in the case containing EP with the highest molecular weight. The strengthening mechanism of EP on the mechanical properties of PBT/RCF composites was studied. The results showed that the improved mechanical properties of PBT/RCF composites resulted from the improvement for the interfacial adhesion between PBT and RCF and the in-situ extended chain reaction of PBT matrix with EP.

1. Introduction

Considering sustainable development in our modern society, the effective recycle of various materials is an important issue and a challenge. With development of recovering technology of carbon fibre (CF) from waste CF reinforced composites, the recovery process provides recycled CF (RCF) with high quality and good mechanical property retention [1–3]. It is benefit able to use RCF to prepare composites instead of the virgin CF in environmental protection and low cost [4]. A few studies have been reported on the application of RCF in some applications such as electromagnetic shielding materials [5], thermally conductive fabric [6], electrode materials [7] et al. There are some studies focusing on exploring a reinforcing potential of RCF in thermoplastic composites [8,9]. However, the reinforcing potential of RCF was closely related to the dispersion state of RCF and the interfacial adhesion between RCF and polymer matrix. In order to obtain good performance, the addition of coupling agent or appropriate surface treatment for RCF is necessary. Han et al. [10] immersed RCF in concentrated nitric acid to clean and polarize the surface of RCF and then treat with silane coupling agent, and found that the surface treatment enhanced the interfacial adhesion between RCF and matrix and thus significantly improved mechanical properties of poly(L-lactic acid)/RCF composites. Wong et al. [11] found that the mechanical properties of the polypropylene(PP)/RCF composites were effectively improved by maleic anhydride grafted polypropylene (PPMA) due to the improvement of compatibility between the RCF and PP, and the compatibility was strongly dependent on the molecular weight and the amount of anhydride groups of PPMA. Recently we have found there are more hydroxyl and carboxy groups on the surface of RCF compared to of virgin CF (without sizing) [1–3], thus the RCF has higher surface activity. In the RCF reinforced composites, it is expected that the coupling reaction between RCF and matrix is implemented with the aid of these hydroxyl and carboxyl groups.

Poly(butylene terephthalate) (PBT) is an important engineering plastics with good mechanical properties, solvent resistance, dimension stability and processability. Glass fibres reinforced PBT composites have been widely used as structural materials. Comparatively, there are only a few studies on CF (or RCF) reinforced PBT composites [12–17]. Some reports studied the properties of woven CF fabric reinforced PBT composites [12,13]. However, the woven CF fabric is different from the short cut CF and RCF. Ng

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et al. [14] found that the electrical conductivity of composites significantly was reduced and the thermal conductivity did not improve, when the boron nitride (BN) and carbon fiber (CF) were added into PBT. Bardash et al. [15] synthesized cPBT from cyclic butylene terephthalate oligomers, and investigated the effects of CF on electrical properties of the cPBT/CF composites. The percolation threshold was observed in the case containing 3 wt% of CF. However, the interfacial interaction between CF and PBT was not studied in these literature.

Gamze et al. [16] studied the influences of sizing components on the properties of PBT/CF composites, in which different polymers (polyurethane (PU), polyamide, epoxy/phenoxy, etc.) were used to size CF before compounding PBT/CF composites. The results found that the CF sized by PU is the best to reinforce PBT as the best wetting by PBT matrix was observed in the case of PU sizing material. Chen et al. [17] treated RCF with concentrated nitric acid and diglycidyl ether of bisphenol A, and investigated the effect of surfaced-treatment RCF on PBT-based composites. It was found that the reinforcing potential of RCF was enhanced substantially by improving the interfacial adhesion between the RCF and PBT due to the surfaced-pretreatment of RCF before melt blending. However, the process of RCF surfaced-pretreatment is cumbersome. A simple method for improving the interfacial adhesion between RCF and PBT is highly expected.

Epoxy resin (EP) is often added into PBT-based composites as in-situ compatibilizer to improve the interfacial adhesion between PBT and fillers (or other polymers) [18–20]. Wu et al. [18] used EP as a compatilizer to prepare PBT/clay nanocomposites through three different blending sequences in melt mixing, and found that the mixing sequence remarkably influenced the dispersion of clay in the matrix. An et al. [19] investigated the effect of EP on compatibility and mechanical properties of PBT/nylon 6 (PA6) blends, and found when the small amount of EP was added, the impact strength and flexural strength of the blends were increased significantly due to improving compatibility of the PBT/PA6 through the reactions of epoxy groups with both PBT and PA6. However, few studies were reported on in-situ compatibilization of EP for PBT/CF (or RCF).

Table 1
The average length and tensile properties of RCF.

Number average length (mm)	Average tensile strength (GPa)	Average tensile modulus (GPa)	Strain at failure (%)
3.2	3.80 ± 1.80	254 ± 61	1.54 ± 0.72

Table 2
The surface elemental composition of RCF.

Surface elemental composition (%)				Oxygenated groups (%)			
C	N	O	O/C	C–C	C–OH	C=O	COOH
81.09	1.31	17.60	0.217	78.71	14.81	0.00	6.48

In this study, EP as an in-situ coupling agent was incorporated into the compounding process to react with the hydroxyl and carboxyl of RCF surface and strengthen the interfacial interaction between PBT and RCF. Three EPs with different molecular weights and epoxy values were used to study the influences of EP molecular weights and loadings on the morphology and mechanical properties of PBT/RCF composites. The effects of EP on mechanical properties of PBT matrix and reaction between PBT and EP were investigated. Finally enhancement mechanism of composites was discussed.

2. Experimental

2.1. Materials

Poly(butylene terephthalate) (PBT, ST800123 NC010) was purchased from E. I. Du Pont de Nemours and Company. The RCF was recycled from the carbon fibre (PAN-based CF, T300, 3k, from Toray Industries, Inc.) reinforced epoxy resin composites using molten potassium hydroxide process [2]. The RCF was cut short for the purpose of convenient feeding. The average length of RCF was quantified using an image analysis technique. The RCF was dispersed in ethanol and then coated on glass slide. Images of various fiber lengths were taken by a Panasonic WV-CP280 Closed-circuit surveillance camera equipped on Shanghai Jinke XTZ-E continuous variable times microscope. The images were then analyzed using Nano Measurer processing software. A series of 1000 samples were measured for RCF. The number average length of fiber was calculated through the formula: $L_N = \frac{\sum N_i L_i}{\sum L_i}$.

The number average lengths and tensile properties of RCF were shown in Table 1. The surface elemental composition and percentages of various oxygenated functional groups of RCF were shown in Table 2.

Three epoxy resin (EP, diglycidyl ether of bisphenol-A), with the trade name of E12 (solid, epoxy value 0.12 mol/100 g, M_w = 1667 Da), E20 (solid, 0.20 mol/100 g, M_w = 1000 Da) and E51 (Sticky liquid, 0.52 mol/100 g, M_w = 385 Da), were supplied by Bluestar New Chemical Materials Co. The chemical structures of PBT and EP were shown in Fig. 1. Before using, all the materials were dried under vacuum at 85 °C for 12 h.

2.2. The preparation of PBT composites

The PBT/EP blends and PBT/EP/RCF composites were prepared in a Banbury mixer (XSS-300 torque rheometer, Shanghai kechuang rubber and plastic machinery equipment co.) at 250 °C with a rotating speed of 60 rpm. In a typical preparation method, PBT was melted, and then EP and SCF at 2 min were in turn added to the PBT melt. The mixture was removed at 7 min, and then cut into small pieces and cooled. The content of RCF was kept at 20 wt.% in the composites. The obtained composites were named as PBT-RCF-E*xy*,

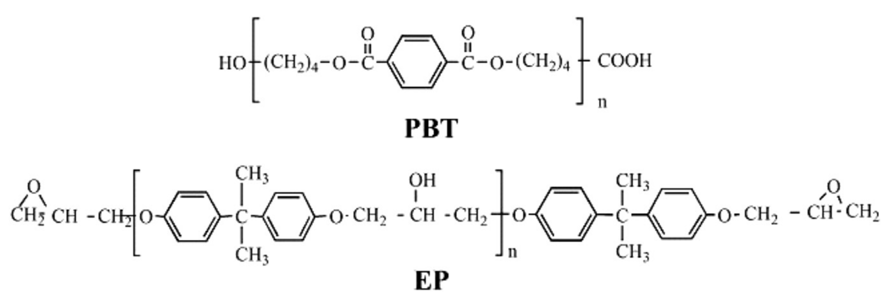

Fig. 1. The chemical structure of PBT and EP.