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Material properties

Revealing toughening mechanism for alternating multilayered polypropylene/poly(ethylene-*co*-octene) sheets

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ABSTRACT

Alternating multilayered (16, 32, and 128 layers) polypropylene/poly(ethylene-*co*-octene) (PP/POE) sheets were prepared via multilayer co-extrusion. Impact (−20°C), tensile, and dynamic rheological tests were carried out on the prepared multilayered samples. The toughening mechanism for the multilayered samples was revealed via investigating their fracture surface morphology and analyzing the work of fracture. The results showed that the impact energy for the notched multilayered samples is mainly absorbed by the continuous thin ductile POE layers due to cavitation, whereas for the unnotched multilayered samples the continuous thin POE and PP layers both contributed significantly to the total work of fracture due to multiple crazing, cracking and distinct plastic deformation in both PP and POE layers, and thus significantly extended crack propagation paths. Hence, the multilayered samples possessed much higher unnotched impact strength than notched impact strength. Moreover, the multilayered samples exhibited slightly increased notched impact strength and obviously increased unnotched impact strength with increasing layer number. Interestingly, the multilayered samples exhibited lower notch sensitivity than the PP sample. In addition to significantly improved low temperature impact toughness, the multilayered samples maintained the strength and stiffness as well as having superior extensibility to that of the PP sample.

1. Introduction

Toughening is one of basic and important topics for brittle polymers. Adding elastomers into brittle polymers has been proved to be an effective approach for improving their impact toughness [1–6]. Generally, relatively high contents of elastomer must be added into the brittle polymers to improve their impact toughness, especially at low temperatures [7]. However, the addition of elastomers often results in sharp weakening in the strength and stiffness of the obtained blends. To overcome the drawbacks resulting from adding elastomers, the general method is to add rigid fillers, such as calcium carbonate [8], silica [9], carbon nanotubes [10] and clay [11], into the elastomer-toughened polymers to obtain ternary composites with enhanced impact toughness and adequate strength and stiffness.

It is well known that the ultimate properties of polymer blends and their composites depend significantly on the morphology of the dispersed phase. Several researches found that a stratified structure such as a co-continuous structure endows the blends with balanced mechanical properties [12,13]. However, the co-continuous structure is usually not easy to obtain in blends with low minor phase content (such as 20 wt% used in this work) using conventional methods [14], such as melt blending via single- and twin-screw extruders. Fortunately, multilayered co-extrusion technology can be used to prepare polymer

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parts with alternating multilayered structure (a special stratified structure), in which each layer is continuous along the extrusion direction [15]. Multilayered co-extrusion technology has a number of applications, such as enhancing barrier properties [16,17], improving filler dispersion [18] and manufacturing functional optical films [19,20]. The final properties of multilayered polymer parts can be tailored via precisely controlling the number and thickness of the layers.

Mechanical properties are very fundamental and critical for polymer parts. Ivan'kova et al. [21] found that multilayered samples made of two brittle incompatible polymers – polystyrene (PS) and polymethylmethacrylate (PMMA) – exhibit a noticeable increase in toughness with increasing layer number due to massive cooperating crazing and yielding of both PS and PMMA layers. The toughness of the multilayered samples made of brittle polycarbonate (PC) and PMMA also increases with increasing layer number [22]. Shen et al. [23] studied the mechanical properties of multilayered propylene-ethylene copolymer/poly(ethylene-co-octene) (PPE/POE) samples via theoretical model prediction and experimental verification. The results showed that the multilayered samples possess higher yield strengths and storage moduli than those of conventional blends with the same POE content. Most of the previous works focused on the tensile properties of the multilayered samples, and less emphasis was given to their impact toughness. Hsieh and Song [24] revealed that the multilayered PC/PMMA sheets exhibit high ballistic impact behavior, which increases with increasing layer number. Recently, Wang et al. [25] found that the alternating multilayered polypropylene (PP) and PP/POE samples possess enhanced low temperature impact toughness and adequate rigidity. In their work, the POE phase was previously dispersed in the PP matrix as blend layers rather than directly co-extruded with PP as continuous pure POE layers. They focused on the effect of POE content rather than the layer number on the notched impact toughness of the multilayered samples. However, there is still a lack of toughening mechanism analyses for multilayered brittle polymer/elastomer samples. Moreover, to the best knowledge of the authors, no report has been published on the unnotched impact toughness for multilayered samples.

For the aforementioned reasons, thick alternating multilayered PP/POE sheets were prepared using a multilayered co-extrusion system designed by our laboratory. Aiming at revealing the toughening mechanism for the multilayered samples, notched and unnotched impact strengths at low temperature (−20°C) were tested and fractured surface morphologies were observed. Moreover, total work of fracture was analyzed for notched and unnotched samples.

2. Experimental

2.1. Materials and sample preparation

The polymers used in this work were commercial PP (grade J501, Sinopec Group Guangzhou, China) and POE (grade Engage 8150, DuPont-Dow Chemical Company, USA). The PP has a melt flow index (MFI) of 2.7 g/10 min (at 230°C and 2.16 kg). The POE consists of 25 wt% octene and has an MFI of 0.5 g/10 min (at 190°C and 2.16 kg).

A multilayered co-extrusion system shown in Fig. 1, which was designed by our laboratory, was used to prepare multilayered PP/POE (80/20, w/w) sheets with a thickness of about 3 mm and a width of about 40 mm. The system included two single-screw extruders (SSEs), a feedblock, an assembly of layer multipliers and a die. One SSE had a screw diameter of 45 mm (SSE-1) and the other SSE had a diameter of 30 mm (SSE-2). They were used to extrude the PP and POE, respectively. The assembly with n layer multipliers could prepare a sheet with $2\times2^{n+1}$ layers. As an example, Fig. 2 schematically shows the layer multiplying process of the sheet with 16 layers. Multilayered PP/POE sheets with 16, 32, and 128 layers (corresponding to 2, 3, and 5 layer multipliers) were prepared and denoted as MC-16, MC-32, and MC-128, respectively. The barrel temperatures along SSE-1 and SSE-2 were set at 120/220/220/220°C and 120/220/220°C, respectively. The temperatures of the feedblock and layer multiplier were set at 220°C. The screw speed of the two extruders was fixed at 20 rpm and the total feeding rate was set at 2 kg/h. For comparison, PP and POE sheets were also prepared via SSE-1 at the same processing conditions without the layer multiplier. The molten PP, POE, and multilayered PP/POE sheets were collected from the die exit and immediately compressed at 30 MPa for 2 min using a compression molding machine to prepare Izod impact, tensile, and rheological test samples.

2.2. Tests and characterization

A rotational rheometer (Bohlin Gemini 200, Malven Instruments, UK) with a parallel-plate (diameter: 25 mm) was used to investigate the rheological behavior of the PP, POE and multilayered PP/POE sheets. Disk samples with a thickness of 1.5 mm and a diameter of 25 mm were tested. An oscillatory mode was used to conduct a dynamic frequency sweep. Dynamic complex viscosity (η^*) and storage modulus (G') with angular frequency (ω) ranging from 0.01 to 100 rad/s were measured for the samples at 220°C. A fixed strain of 1% was used to ensure that tests were carried out within the linear viscoelastic range of the samples investigated.

Notched and unnotched Izod impact tests were performed at −20°C with a pendulum impact tester (XJU−22, Chengde tester, China) following ISO-180-2000. The impact tests were carried out in the transverse direction, that is, the impact force was applied parallel to the microlayer surface, as schematically shown in Fig. 3. The size of the

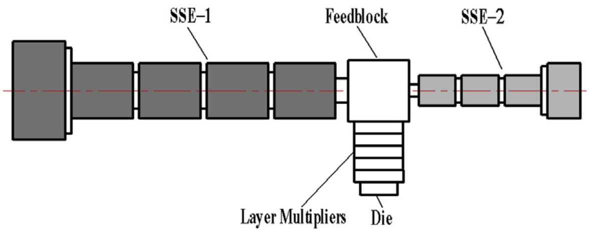

Fig. 1. Schematic of multilayered co-extrusion system used in this work.