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Test Method

Characteristics of the brittle/tough transition of poly(butylene terephthalate)/maleinized poly(ethylene-octene) blends determined by the essential work of fracture procedure

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ABSTRACT

Blends of poly(butylene terephthalate) (PBT) with varying contents of a maleic anhydride modified poly(ethylene-octene) were obtained by melt processing. The comparison of the standard Izod impact test with the essential work of fracture (EWF) procedure against either the blend composition or the inter-particle distance (τ) revealed that the EWF steadily increased with the rubber content, and the blend composition causing the brittle/tough transition obtained by the standard Izod technique was similar to that obtained using the non-essential work of fracture ($\beta\, w_p$).

The slight increase in toughness in the brittle zone of the impact strength curve when compared with either the rubber content or τ does not appear in the curve for the $\beta\, w_p$. In the tough zone of the EWF curve, unlike that of the impact test, toughness increases of up to 30% and some change of morphology of the fracture surfaces were seen.

1. Introduction

It is commonly accepted that toughness is, probably only second to stiffness, the most relevant mechanical property of polymers. Its value clearly depends, among others, on the fracture method, test speed and specimen characteristics. The Izod test is one of the most common impact methods. However, pendulum tests such as the Izod or Charpy methods have serious limitations, mainly related with the notch geometry, which renders the results somewhat limited in their applicability [1,2]. These limitations seriously reduce the significance of the test [1], mainly as regards design, because the test does not provide any value that might be termed fundamental.

To overcome these limitations, the essential work of fracture (EWF) procedure has been proposed as a valuable method to assess toughness in ductile polymeric materials. The EWF procedure involves fracture of a series of specimens where the ligament length changes slowly enough to extrapolate the results accurately. The EWF procedure also involves the need to obtain force-deformation plots during the impact test; this complicates the experimental method and makes it expensive. Consequently, toughness of ductile polymer materials has, up to now, been mostly characterized in the open literature by standard pendulum impact tests. This is with the exception of blends of polyamide 6 with poly(ethylene-octene) (PEO) [3], acrylonitrile-butadiene-styrene (ABS) [4–6], maleated ethylene-propylene rubber (mEPR) [7], and glass fiber/mEPR [7], of polycarbonate [8] and poly(butylene terephthalate) (PBT) [9] with ABS, and of amorphous polyamide with PEO [3].

Thus, the advantages of using standard pendulum impact tests are clear but their significance is limited. It would be of clear interest to find out to what extent the results of the Izod tests are similar to those provided by the EWF procedure. This information would improve the

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significance of the results of the Izod tests as related with results closer to design such as those provided by the EWF procedure.

In this paper we have characterized a rubber toughened polymer blend both through standard Izod tests and the EWF procedure. The blend tested was PBT/poly(ethylene-octene) modified with maleic anhydride (mPEO), where the mPEO content changed from 0 to 30% to ensure the brittle/tough transition took place, and was obtained in the melt state and characterized through dynamic-mechanical analysis (DMA), differential scanning calorimetry (DSC) and scanning electron microscopy (SEM). Subsequently, both simple and instrumented Izod impact tests were performed and the comparative results discussed. The aim was to find out the differences between the two testing procedures and, thus, to enhance existing information on the practical significance of the already widespread results from standard Izod tests.

2. Experimental

The poly(butylene terephthalate) PBT used in this work was CRASTIN S600F10 (DuPont) and the PEO rubber was ENGAGE EG 8200 (DuPont–Dow). The weight proportion of octene in the PEO was 24%. The reactive monomer used for grafting was a commercial maleic anhydride (MAH, 98% purity) and the peroxide initiator was dicumyl peroxide (DCP) (Aldrich). The PBT (4 h at 120 °C), the PEO (6 h at 60 °C) and the mPEO (2–3 days at 60 °C) were dried before processing in an air oven to avoid possible moisture induced-degradation reactions.

The grafting of PEO was made using the procedure described in a previous work [10]. The amount of MAH grafted in the mPEO was 0.61%. Blending of PBT and mPEO was carried out in a Collin twin screw extruder–kneader (type ZK 25) with L/D ratio of 30 and screw diameter of 25 mm. PBT/mPEO blends with mPEO contents ranging from 5 to 30% were processed at 250 °C and at a rotor speed of 50 rpm. The rod extrudate was cooled in a water bath, and then pelletised.

Injection moulding was carried out in a Battenfeld BA230E reciprocating screw injection moulding machine to obtain impact (ISO 180, thickness 3.2 mm) specimens. The screw had a diameter of 18 mm and an L/D ratio of 17.8. The melt temperature was 250 °C (190 °C for pure mPEO), the injection pressure was 120 MPa, and the mould temperature was 60 °C. The notches of the impact specimens (depth 1.5–7.5 mm and radius 0.25 mm) were machined after injection moulding. A sharp notch was made by inserting a fresh razor blade into the root of the notch. This led to ligament lengths ranging from 3 to 9 mm.

The thermal behaviour of the blends and pure components was studied by differential scanning calorimetry (DSC) using a Perkin Elmer DSC-7 calorimeter. The samples were heated from 25 to 280 °C at 20 °C/min. The crystallization and melting temperatures and heats were calculated from the maxima and from the areas of the corresponding peaks, respectively. The phase structure was studied by DMA analysis using a TA Q800 that provided the loss tangent ($\tan\delta$) and the storage modulus (E') against temperature. The scans were carried out in bending mode from −135 °C to roughly 120 °C at a constant heating rate of 4 °C/min and a frequency of 1 Hz.

The surfaces of both the cryogenically fractured and the impact test fractured specimens were observed by SEM after gold coating. A Hitachi S-2700 electron microscope was used at an accelerating voltage of 15 kV. The weight-average particle size, $\overline{d}_w$, was calculated from a minimum of 200 particles as

$$\overline{d}_w = \frac{\sum_i n_i d_i^2}{\sum_i n_i d_i} \tag{1}$$

where n_i is the number of particles with size d_i. The inter-particle distance (τ) was calculated by means of

$$\tau = \overline{d}_w \left[\left(\frac{\pi}{6\phi} \right)^{\frac{1}{3}} - 1 \right] \tag{2}$$

where ϕ is the volume fraction of the matrix.

Instrumented Izod impact testing was performed on the notched specimens using a CEAST 6548/000 pendulum with a data acquisition system DAS 8000. Some specimens did not fully break after the impact test due to their highly tough nature; in these cases, to calculate the relative energy of fracture, the actual broken ligament length area was used instead of the initial ligament length area [6]. In the EWF procedure, the total fracture energy (W_f) (calculated from the area under load-displacement curves) is the addition of the surface related essential work (W_e), and the volume related non-essential work (W_p). W_e is the measure of the energy needed to create new surface, and W_p is the measure of the energy-absorbing process surrounding the fracture surface. Using specific energies (w_f, w_e and w_p):

$$w_f = w_e + w_p \beta l \tag{3}$$

where l is the ligament length and β is a shape factor of the plastic zone. According to Eq. (3), the straight line obtained by plotting w_f (obtained from the impact test and the ligament area) versus l for different ligament lengths, will provide us with w_p β (the slope) and w_e (the y-intercept).

3. Results and discussion

3.1. Phase structure

When the melting behaviour of the blends was studied by DSC, the T_m (229 °C) and the crystallinity (30%) of PBT remained constant with the addition of mPEO, indicating that the rubber phase did not disturb the crystallization process of PBT. When the phase structure of the blends was studied by DMA, the T_g of the PBT did not change. The slight decrease in the low intensity T_g of mPEO in the blends was previously observed in PBT/mPEO [10] and PP/PEO [11] blends, and was attributed to the different processing conditions used in both the blends and in the pure mPEO. These results indicate the presence of two pure amorphous phases in the blends.