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Stiffer and super-tough poly(butylene terephthalate) based blends by modification with phenoxy and maleated poly(ethylene–octene) copolymers

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Abstract

New super-tough poly(butylene terephthalate) (PBT) materials were obtained by melt blending PBT with both 20 wt% phenoxy (Ph) and 0–30 wt% maleic anhydride grafted poly(ethylene–octene) (mPEO) copolymers with different grafting levels. Ph was completely miscible in the PBT matrix. The presence of mPEO did not influence either the nature of the PBT–Ph matrix or the crystallization of PBT. The overall decrease in particle size and in interfacial tension upon grafting indicated that compatibilization had taken place. Super-tough (impact strength 23-fold that of the PBT) and stiffer PBT based blends were obtained at mPEO contents equal to or higher than 15%. The dependence of the critical inter-particle distance (τ_c), on both adhesion measured by means of the interfacial tension, and on the relation between the modulus of the matrix and that of the rubbery dispersed phase (E_m/E_d), is proposed.

Keywords: Poly(butylene terephthalate); Super-toughness; Critical inter-particle distance

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1. Introduction

Rubber toughening of thermoplastic polymers is extensively used to improve the low impact strength of polymers [1]. This technique has been applied not only to inherently brittle polymers, but also to ductile notch-sensitive polymers. Recently, impact strength improvements of 15- or even 20-fold that of the matrix have been achieved by compatibilizing thermoplastic/rubber blends. Some examples are poly(butylene terephthalate) (PBT)/poly(ethylene–octene) (PEO) [2–5], PBT/poly(acrilonitrile-*co*-butadiene-*co*-styrene) (ABS) [6,7], PBT/ethylene–propylene–diene (EPDM) [8], Nylon/PEO [9–13] and Nylon/EPDM [14]. The most common functional groups used for compatibilization of these blends were maleic anhydride (MA) [2,9–13] and epoxy [3–14].

Nowadays, the inter-particle distance (τ) [15] is widely accepted as the parameter that controls super-toughness in rubber-modified compatibilized thermoplastic blends [2–4, 16–18]. Among the parameters that influence τ_c, the influence of extrinsic parameters such as the test temperature [17,19–21], strain rate [22,23] and mode of deformation [20] is clear. The possible influence of intrinsic parameters such as the presence of a plasticizer [24], degree of crystallinity [17], the type and modulus [17,18,25] of the rubber, the E_m/E_d relation [2], and the adhesion between the components [3,4,26] is more difficult to define, with the exception of adhesion. This is because, the change of one intrinsic parameter usually gives rise to the change of a second.

It has been seen that the so-called super-tough PBT blends can be obtained by blending PBT with PEO either grafted with MA (mPEO) [2] or modified by a difunctional epoxy resin [3,4]. In the first study [2], the influence of the relation between the modulus of the matrix and that of the rubbery dispersed phase (E_m/E_d) on τ_c was tested, while in the second the influence of adhesion between the components [3,4] on τ_c was stated. However, more experimental work is necessary to gain insight which are the intrinsic parameters that influence τ_c in thermoplastic/rubber blends.

In this study the PBT matrix of super-tough PBT/mPEO [2] blends was modified with Phenoxy (Ph), which is miscible with PBT [27,28], to test the possibility of achieving super-toughness in stiffer materials. Moreover, the fact that both E_m/E_d and adhesion between the components should change upon Ph addition, will allow the study of whether τ_c depends on either E_m/E_d or adhesion, or both factors.

With these aims, (PBT–Ph)/mPEO blends with an 80:20

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PBT–Ph ratio and with mPEO contents from 0 to 30 wt% were obtained by extrusion and subsequent injection moulding. The grafting level of PEO changed from 0 to 1.80%. The blends were characterized by differential scanning calorimetry (DSC), dynamic mechanical analysis (DMTA), contact angle measurements, Fourier transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM). Their mechanical properties were measured by means of tensile and notched impact tests. The τ_c–E_m/E_d, and τ_c-adhesion pairs of this blend are compared with those collected from previously studied thermoplastic/rubber blends.

2. Experimental

The PBT used in this work was CRASTIN S600F10 (DuPont), PEO was ENGAGE EG8200 (DuPont–Dow) and Ph was PKHH from Union Carbide (supplied by Quimidroga, S.A., Barcelona, Spain). The proportion of octene in the PEO was 24 wt%. PBT (4 h at 120 °C), PEO (6 h at 60 °C), and Ph (16 h at 80 °C) were dried to avoid possible moisture-degradation reactions in an air oven (PBT and PEO) and in vacuo (Ph), respectively. The mPEO was that of Ref. [2]. It was obtained by mixing MA (from 0.5 to 3 wt%) with a constant amount of peroxide initiator (dicumyl peroxide, 0.05 wt%) in a Collin twin-screw extruder–kneader. The obtained amount of MA grafted in PEO was measured by means of the titration with 0.01 M KOH. The temperature profile along the extruder was 50, 140, 155, 170, 170 and 175 °C and the rotor speed was 70 rpm. PEO-0.63 indicates that the amount of grafted MA of the PEO is 0.63%.

Blending of PBT, Ph and mPEO at the desired compositions was carried out in a Collin twin-screw extruder–kneader (type ZK25, *L/D* ratio 24, screw diameter 25 mm). All the blends were processed at 250 °C, with the exception of the (PBT–Ph)/mPEO 70/30 blends that were processed at 225 °C because of the low melt strength at 250 °C. The rotor speed was 50 rpm. The rod extrudate was cooled in a water bath, and then pelletized. Injection moulding was carried out in a Battenfeld BA230E reciprocating screw injection moulding machine to obtain tensile (ASTM D638, type IV, thickness 3.2 mm) and impact (ASTM D256, 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 260 °C (190 °C for the neat PEO) and the mould temperature 60 °C (15 °C for the neat PEO). The injection speed and pressure were 7 $\mathrm{cm^3/s}$ and 120 MPa, respectively.

The crystalline structure of PBT and of the blends was studied by DSC using a Perkin–Elmer DSC-7 calorimeter and indium as reference. The samples were first heated from 10 to 270 °C at 20 °C/min. The melting temperature and enthalpy were determined from the maxima and the areas of the corresponding peaks, respectively. The crystallinity of PBT was calculated supposing a melting heat of 142 J/g [29]. Dynamic mechanical thermal analysis was performed on a Polymer Laboratories DMTA that provided the loss tangent (tan δ) and the storage moduli (E') as a function of temperature. The scans were carried out in single cantilever mode at a constant heating rate of 2 °C/min and at a frequency of 1 Hz, from −130 °C until roughly 120 °C.

The possible reactions between PBT and Ph were studied by FTIR, using a Nicolet 5 DXC spectrophotometer. The contact angle measurements were carried out on a CAM 100 goniometer (KSV) on injection moulded specimens, using water and ethylene glycol. The mean standard deviation of the measurement was 2–3° which gave rise to an error in the interfacial tension values of approximately 20%.

The surfaces of cryogenically 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 rubber particle diameter was measured in representative zones of the cryogenically fractured specimens. The weight-average particle size, $\bar{d}_w$, was calculated from a minimum of 200 particles by means of [1,19,21,22]

$$\bar{d}_w = \frac{\sum n_i d_i^2}{\sum n_i d_i} \tag{1}$$

where n is the number of particles with size d, assuming a spherical geometry. The inter-particle distance, τ, was calculated by means of the equation proposed by Wu [15]

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

where ϕ is the volume fraction of the rubber.

The tensile tests were carried out using an Instron 4301 tensile tester at a cross-head speed of 10 mm/min and at 23 ± 2 °C on specimens according to ASTM D638 type IV. The Young's modulus was determined by means of an extensometer. The yield stress and ductility, measured as the break strain, were determined from the load–displacement curves. The elongation at break was determined assuming a gauge length of 64 mm. Izod impact tests were carried out on notched specimens (ASTM D256) using a CEAST 6548/000 pendulum. The notch (depth 2.54 mm and radius 0.25 mm) was machined after injection moulding. A minimum of five tensile specimens and ten impact specimens were tested for each reported value.

3. Results and discussion

3.1. Phase structure

The DSC did not provide information on the T_g behaviour of the blends, because the T_g of the miscible PBT–Ph matrix overlapped with the T_m of PEO. For this reason the phase structure of the blends will be discussed by

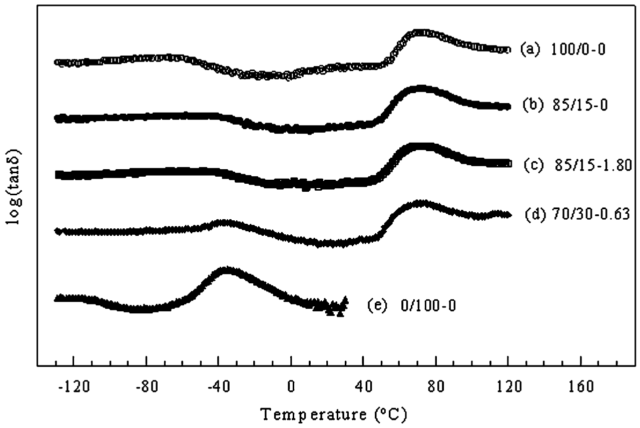

Fig. 1. DMTA log(tan δ) vs. temperature, 80/20 PBT–Ph matrix (a), and (PBT–Ph)/mPEO 85/15-0 (b), 85/15-1.80 (c), 70/30-0.63 blend (d) and of the neat PEO (e). To aid clarity, the curves are shifted on the vertical axis.

reference to the DMTA data. Fig. 1 shows the DMTA scans of the 80/20 PBT–Ph matrix (a), 85/15 (PBT–Ph)/mPEO blend with grafting levels of 0 (b) and 1.80% (c), the 70/30-0.63 blend (d) and neat PEO (e). As can be seen, the curve (a) of the PBT–Ph 80/20 blend shows a single T_g (at roughly 72 °C) at temperatures intermediate to those of the neat components (58 and 88 °C, respectively, for PBT and Ph from Refs. [2,30]). This indicates, in agreement with former results [27], that the blend is fully miscible. A possible reaction between PBT and Ph was ruled out by FTIR as no difference was observed between the weighted sum of the spectra of the pure PBT and Ph components and that of the 80/20 PBT–Ph blend.

As can also be seen in Fig. 1, when the scans of the PBT–Ph matrix (a) and that of the 70/30-0.63 blend (d) are compared, the position of the T_g of PBT–Ph did not change significantly. Moreover, when the scan of the 85/15-0 blend (curve b) is compared with that of the 85/15-1.80 blend (curve c), the maximum grafting level of PEO, the T_g of the PBT–Ph matrix also remained practically constant. Therefore, neither the presence of PEO nor that of maleic anhydride influenced the nature of the matrix of the (PBT–Ph)/mPEO blends, which is composed of miscible 80/20 PBT–Ph blend. Finally, the intensity of the T_g of PEO is low. Its position, which can be accurately determined only in the 30% mPEO blend of Fig. 1d, did not change with respect to that of neat PEO, indicating its pure PEO nature.

The melting behaviour of (PBT–Ph)/mPEO blends was studied by DSC, and in Fig. 2 the corresponding results of the first scan of the 80/20 PBT–Ph matrix (a), the 85/15 (PBT–Ph)/mPEO blends with grafting levels of 0% (b) and 1.80% (c), and the 70/30-0.63 blend (d) are shown. As can be seen, no crystallization exotherm was observed during the heating scan, in spite of the fast cooling in the injection mould. Moreover, as seen in curve (a) of Fig. 2, the T_m (228 °C) of PBT (34%) did not change (as did crystallinity) with the addition of Ph. As can also be seen when the (b) and (c) curves of the 85/15 blends with 0 and 1.80% grafting level are compared, the T_m of PBT remained practically constant (as did crystallinity) with the grafting level. Finally, as can also be seen in Fig. 2 when curves (b) and (d) are compared, the T_m of PBT did not change significantly (as did crystallinity) with the PEO content. This indicates that neither the addition of either Ph or PEO, nor the grafting level of PEO disturbed the crystallization process of PBT, as was previously seen in PBT/PEO blends [2,3] and in other semicrystalline matrix/elastomer blends [31,32].

3.2. Morphology

The cryofractured surfaces of the injection moulded impact specimens were observed by SEM. As in other blends [2–4,33,34], a fine layer (100–150 μm) with elongated rubber particles covered the specimens. However,

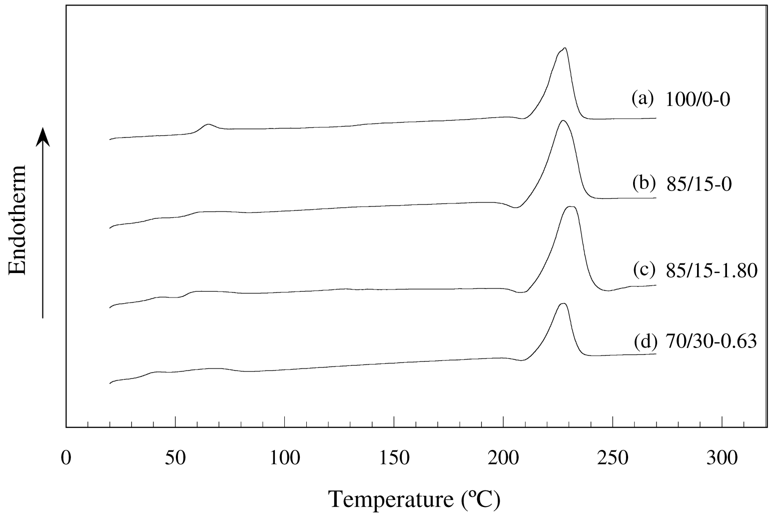

Fig. 2. DSC heating scans of the 80/20 PBT–Ph matrix (a), and the (PBT–Ph)/mPEO 85/15-0 (b), 85/15-1.80 (c) and 70/30-0.63 blends (d). To aid clarity, the curves are shifted on the vertical axis.

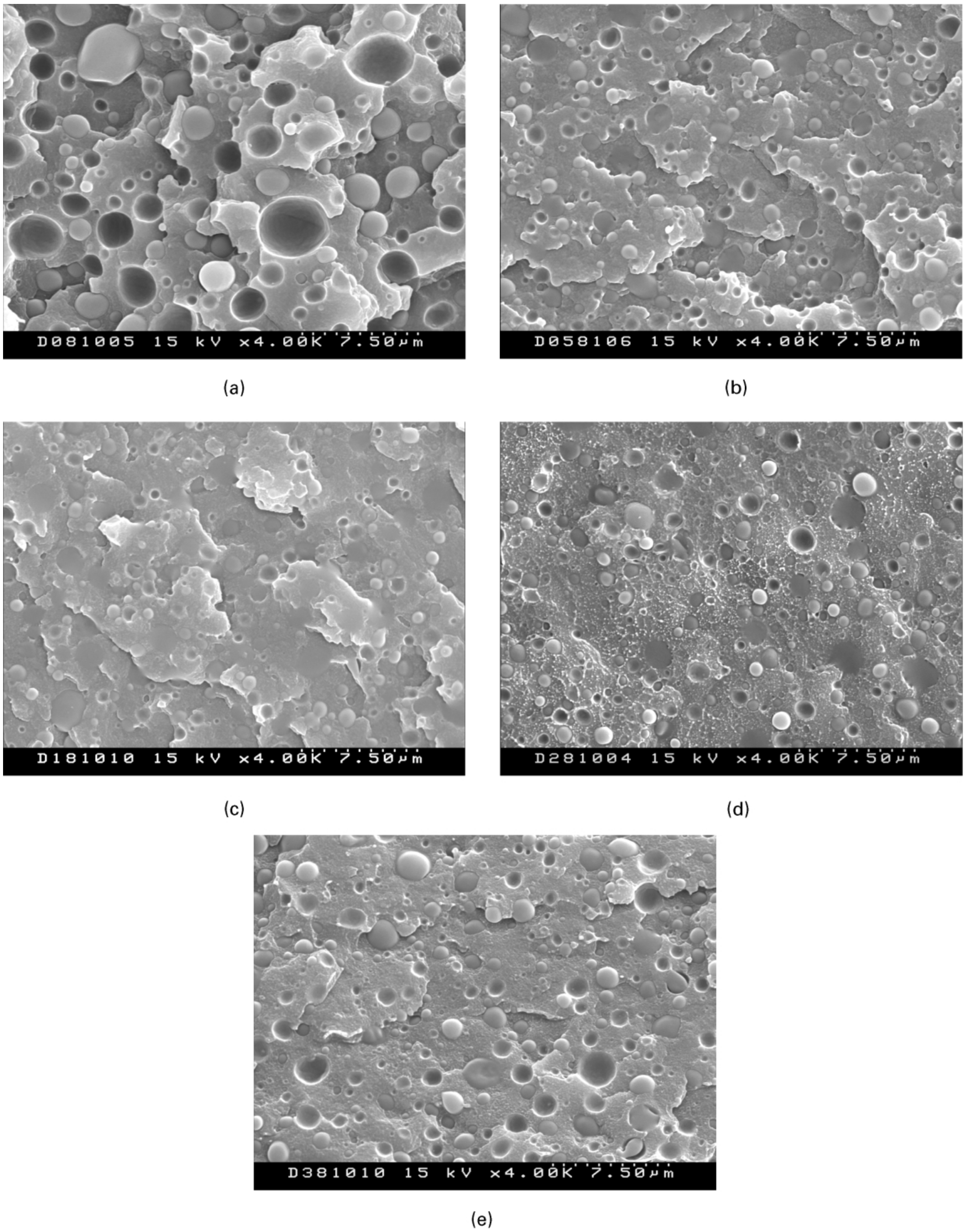

Fig. 3. Cryofractured surfaces of the inner zone of injection moulded impact specimens with 15% mPEO and 0 (a), 0.32 (b), 0.63 (c), 1.14 (d) and 1.80% (e) grafting level.

as a consequence of its low thickness and of its lack of effect on mechanical properties, it will not be discussed. In the rest of the transverse section, the morphology was slightly finer in the outer than in the inner part of the specimen (roughly two-thirds of the specimen). The morphological changes of the two parts followed the same trends. Therefore, only the morphology of the inner part of the specimens will be shown.

Fig. 3 shows the cryogenically fractured surfaces of the core of the 15% mPEO blends with grafting levels of 0 (a), 0.32 (b), 0.63 (c), 1.14 (d) and 1.80% (e). The correspondent $\bar{d}_w$ values of the blends, calculated by means of Eq. (1), are shown in Table 1. As can be seen both in Fig. 3 and in Table 1, the particle size clearly decreased when the grafting level increased from 0 to 0.32%. However, as the grafting level further increased, the particle size remained practically

Table 1
The weight-average particle size, $\bar{d}_w$, of (PBT–Ph)/mPEO 85/15 blends at different grafting levels

Grafting level (%)	$\bar{d}_w$ (μm)
0	1.90 ± 0.22
0.32	0.98 ± 0.13
0.63	1.02 ± 0.12
1.14	1.05 ± 0.11
1.80	1.17 ± 0.13

constant, and at the maximum grafting level tested (1.80%) $\bar{d}_w$ increased slightly. The decrease in $\bar{d}_w$ as a consequence of grafting indicates compatibilization, probably by means of interactions or grafting reactions at the interface. The slight $\bar{d}_w$ increase at high grafting levels was previously seen in PBT/mPEO blends [2], and will be discussed later.

As can also be seen in Fig. 3a, the adhesion level was poor. This is because the fracture was always adhesive and the fracture surface of both particles and holes appeared clear. However, when mPEO was used (Fig. 3b–e) a number of particles broke just at the level of the fracture surface of the matrix, indicating that interfacial adhesion improved.

With respect to the effect of the PEO content, Fig. 4 shows the morphology of the PEO-0.63 blends with 10 (a), 20 (b) and 30% (c) mPEO contents. As can be seen, the particle size was rather homogeneous up to 20% mPEO. However, in the 30% mPEO blend, the heterogeneity clearly increased. Moreover, as is usual in rubber-toughened blends [2–4,13,33], when the mPEO content increased from 10 to 30%, the mean particle size increased from 0.72 to 3.04 μm. In the 30% mPEO blend of Fig. 4c, the dispersed rubber particles were even large enough to include some smaller particles of obvious PBT–Ph nature inside. Broken particles similar to those of Fig. 3b–e appeared in these blends mainly at rubber contents equal to or higher than 20%.

In order to prove the improved compatibility upon mPEO addition, the interfacial tensions between PBT–Ph and both PEO-0 and mPEO were measured by means of contact angle measurements. The interfacial tension between PBT–Ph and PEO-0 was 1.74 mN/m, whereas that between PBT–Ph and mPEO was 0.55 mN/m, whatever the grafting level. Thus, this decrease in the interfacial tension indicates that the grafted MA compatibilized the blends, and was the reason for the observed particle size decrease.

The observed increase in $\bar{d}_w$ at high grafting level was unexpected, because more functional groups should lead to higher interaction, and therefore, to smaller interfacial tension and particle size. This did not take place because, as

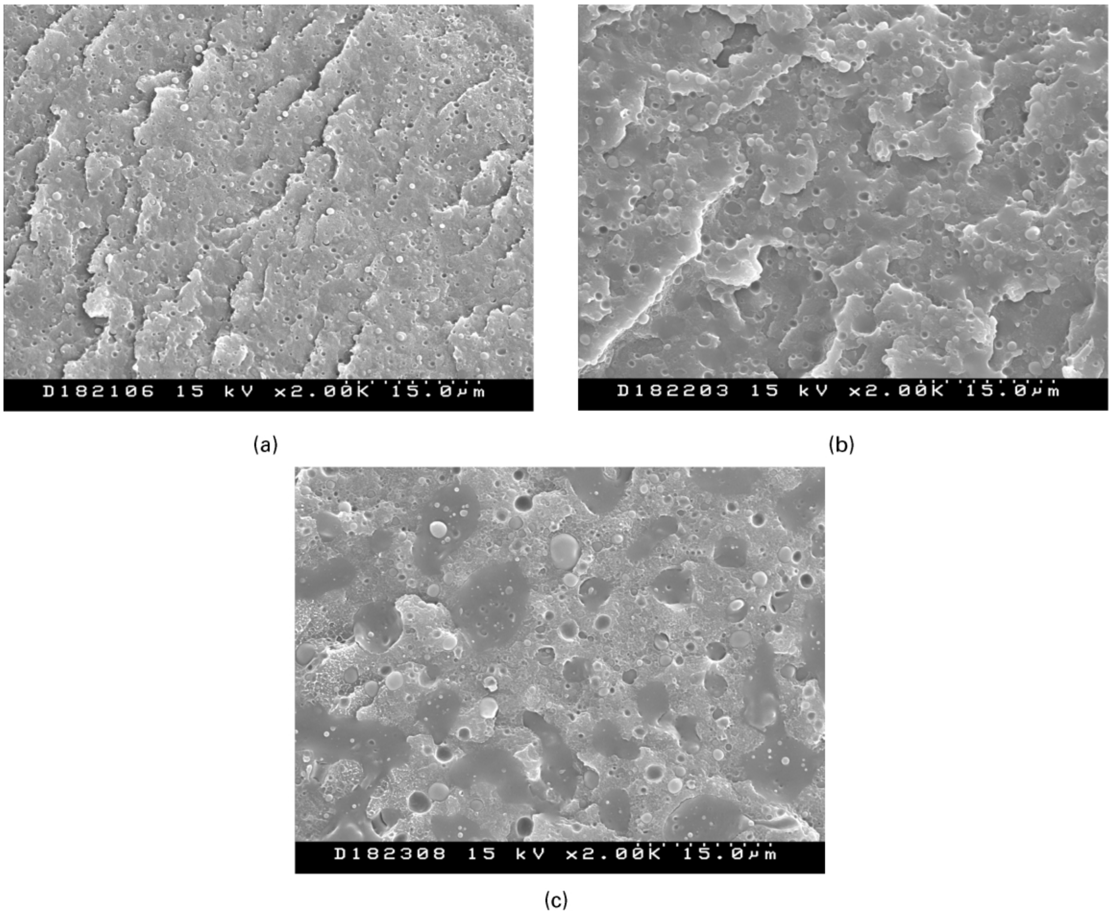

Fig. 4. Cryofractured surfaces of the inner zone of injection moulded impact specimens of (PBT–Ph)/PEO-0.63 90/10 (a), 80/20 (b) and 70/30 (c) blends.

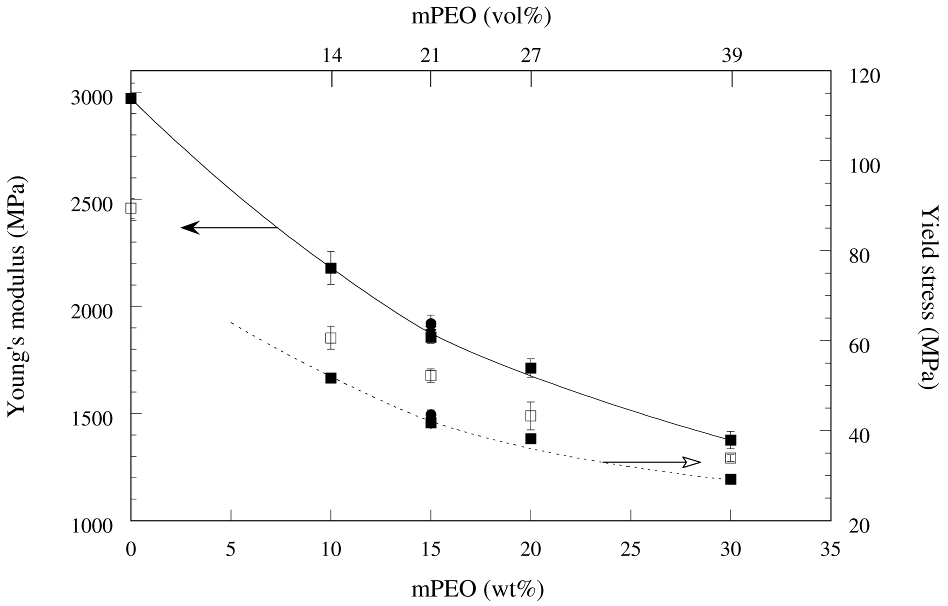

Fig. 5. Young's modulus and yield stress of (PBT–Ph)/mPEO blends as a function of PEO-0.63 content. Those of the 15% mPEO blends are also plotted at different grafting levels: 0 (●), 0.32 (▲), 0.63 (■), 1.14 (▼) and 1.80% (◆). Empty squares correspond to the Young's modulus of PBT/mPEO blends of Ref. [2].

seen before [2], at high grafting levels, the viscosity of mPEO increases, leading to a larger particle size.

3.3. Mechanical properties

Fig. 5 shows the Young's modulus and the yield stress of the blends both against the PEO-0.63 content, and also at different grafting levels in the case of the 15% mPEO blends. The empty squares of Fig. 5 correspond to the Young's modulus of PBT/mPEO blends of Ref. [2], which are plotted as a reference. The Young's modulus and the yield stress, as well as the other mechanical properties, were plotted against both the weight and volume compositions as the densities of PBT (1.31 g/cm^3), PEO (0.87 g/cm^3) and Ph (1.17 g/cm^3) are very different. The 80/20 PBT–Ph matrix broke before yielding at 62 MPa.

As can be seen, both the Young's modulus and the yield stress of the 15% mPEO blend at different grafting levels were almost identical. Therefore, as found in the previous works [2,3,34,35], the improved compatibility had no significant effect on these low-strain properties. As can also be seen, and as a consequence of the elastomeric nature of PEO [2,4,9,36], both the Young's modulus and the yield stress decreased when the mPEO content in the blend increased. However, the Young's moduli of these (PBT–Ph)/mPEO blends are clearly higher than those of the corresponding PBT/mPEO blends [2] (empty squares) with the same PEO content, due to the higher (approx. 20%) modulus of their matrix. This will allow us to test whether the modulus of the matrix has an effect on the τ_c values.

The effect of the PEO-0.63 content on the ductility of the blends is shown in Fig. 6. In the case of the 15% mPEO blend, the ductility of blends with different grafting levels is also shown. As can be seen, in the 85/15 blend, the ductility improved with the grafting level, from 10% in the PEO-0 blend to 50% when PEO-0.32 was used, but more grafting did not influence ductility. As can also be seen, the ductility increased slightly up to 10% mPEO, then it clearly increased, and finally it remained practically constant at roughly 40%. The ductility increases observed upon mPEO addition in Fig. 6 indicate that mPEO compatibilized the blends. This is despite the low ductility of the PBT–Ph matrix of this work, which is smaller than that of a previous work [27] (obtained with a different processing method and conditions) and much lower than that of the pure PBT (200%) of a previous work [2].

Fig. 7 shows the impact strength of (PBT–Ph)/mPEO blends as a function of the PEO-0.63 content. In the case of the 15% mPEO blend, the impact strength of blends with different grafting levels is also shown. All the blends showed a stress-whitening zone after fracture, that in the blends with low impact strength extended only around the notch, whereas in super-tough PBT it extended along the whole fracture surface. As can be seen, the impact strength of the PBT–Ph matrix improved from roughly 10 to 110 J/m upon the addition of 10% mPEO. But slightly larger mPEO contents increased greatly the impact strength to 600 J/m. Further increases in the mPEO content led to slight increases in impact strength, reaching 700 J/m in the 30% mPEO blend.

A similar behaviour is seen in Fig. 7 when the grafting level of the 15% mPEO blend changed. Thus, the impact strength increased rapidly from 95 J/m in the blend with PEO-0 up to at least 600 J/m (the specimen did not break) in the blend with PEO-0.32. Further increases in the grafting level up to 1.14% had no significant influence on the impact

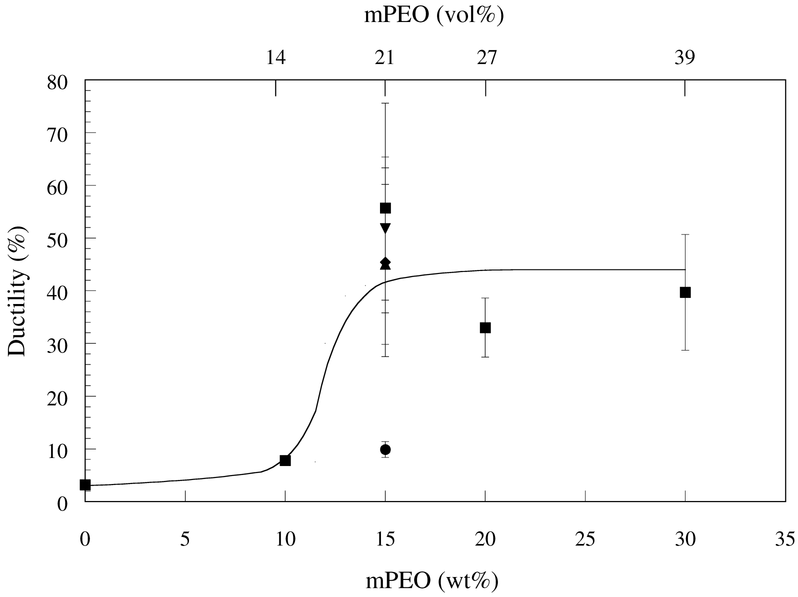

Fig. 6. Ductility of (PBT–Ph)/mPEO blends as a function of mPEO content. Symbols as in Fig. 5.

strength. When the grafting level increased to 1.80%, the impact strength decreased, probably due to the $\bar{d}_w$ increase observed before in Fig. 3e and in Table 1 at 1.80% grafting level.

The impact strength values of Fig. 7 are typical of super-tough blends, because they are about 6-fold that of the corresponding blend with ungrafted PEO and more than 23-fold that of PBT. Furthermore, the impact strength value of the super-tough blends of Fig. 7 are minimum values. This is because the specimens did not break completely under testing, and roughly 20, 30 and 35% of the transversal area of the specimens with 15, 20 and 30% mPEO, respectively, remained unbroken.

The impact modification of PBT by compatibilization with rubber-like materials has been studied by different authors. Although the results are often not strictly comparable despite the use of the same standard (the processing conditions also play a role through the structures obtained), in Table 2 the results of works on both PBT and PEO are compared together. The impact strength, the stiffness, the rubber content at which maximum toughness was achieved, and the thickness of the specimens (that leads to either plane stress or strain conditions, thus influencing impact strength) are given.

As can be seen, the maximum impact strength attained corresponds to the addition of the tough PC to PBT in blends with a core–shell modifier [37]. Another study now in progress in our laboratory on the impact strength

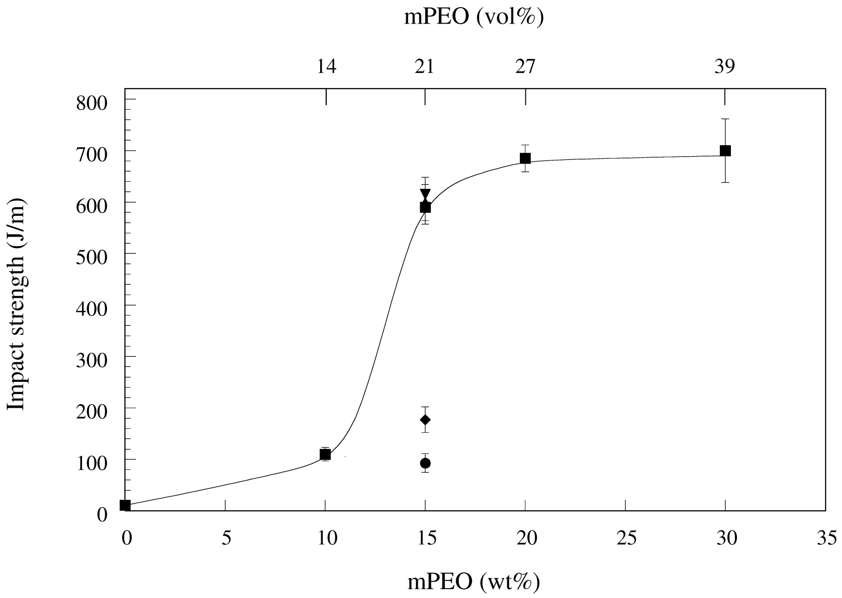

Fig. 7. Impact strength of (PBT–Ph)/mPEO blends as a function of mPEO content. Symbols as in Fig. 5.

Table 2
Rubber content at which maximum toughness appears, impact strength, stiffness, thickness of the specimens and testing method used for compatibilized thermoplastic/rubber based blends based on either PBT or PEO

Materials	Rubber content (wt%)	Impact strength (J/m)	Modulus of elasticity (GPa)	Thickness (mm)	Testing method	Reference
(PBT–Ph)/mPEO	20	700	1.7	3.2	ASTM D256	This work
PBT/mPEO	20	640	1.5	3.2	ASTM D256	[2]
PBT/PEO-epoxy	20	560	1.5	3.2	ASTM D256	[4]
(PBT-PC)/core–shell	20	1000	1.7	3.18	ASTM D256	[37]
PBT/ABS/MGE	30	700	1.7	–	ASTM D256	[7]
Nylon 6/mPEO	20	500	1.4[a]	–	National Standard GB 183-80[b]	[9]
PETG/mPEO	15	800	1.2[a]	–	National Standard GB 183-80[b]	[36]

The values corresponding to PBT/epoxy-EPDM (60 kJ/m^2) blends of Ref. [8] are not given due to the different units used.

[a] Flexural modulus.

[b] Similar to ASTM D256.

of PBT–PAr/mPEO blends, shows impact strength values similar to those of Ref. [37]. Moreover, the highest impact strength also corresponds to a matrix modified blend. Therefore, the maximum impact strength appears to be related to matrix modification. Finally, the results of this study and those of Refs. [7,37] link super-toughness with higher modulus of elasticity. The unknown thickness of the specimens of both Nylon 6 and PETG modified prevents the study of the effects of the nature of the matrix.

3.4. Inter-particle distance

It is known that τ_c is increasingly accepted [15] as the parameter that controls super-toughness in rubber modified thermoplastics, in preference to the minimum rubber particle diameter (d_c) [1,38], that should be used only in blends of the same composition. This is clear in the blends of this study, because the 85/15-1.80 blend (mean particle diameter 1.17 μm) was brittle, whereas the 80/20-0.63 blend with the same diameter (1.14 μm) was super-tough. Therefore, τ_c is used to characterize the impact behaviour of the blends.

The impact strength of the blends studied are plotted as filled squares against the τ values, calculated by means of Eq. (2), in Fig. 8. The values that correspond to the same blend without Ph [2] are also shown in Fig. 8 (empty squares and broken line) as a reference. As can be seen, a sharp increase in the impact strength of the blends took place at a τ value in the region of 0.39 μm. Thus, blends with $\tau > 0.40$ μm (90/10-0.63 for instance) showed low impact strength, whereas blends with $\tau < 0.37$ μm (85/15-1.14 for instance) showed super-tough behaviour. Thus, a rather accurate τ_c value of 0.39 ± 0.01 μm for these blends is inferred from Fig. 8.

Although τ_c was firstly considered [15,16,20] a characteristic of a given matrix, subsequent studies showed that the τ_c of a blend depended on extrinsic parameters [17,19–23]. Moreover, it has been proposed that intrinsic parameters such as the use of a plasticizer [24], crystallinity content

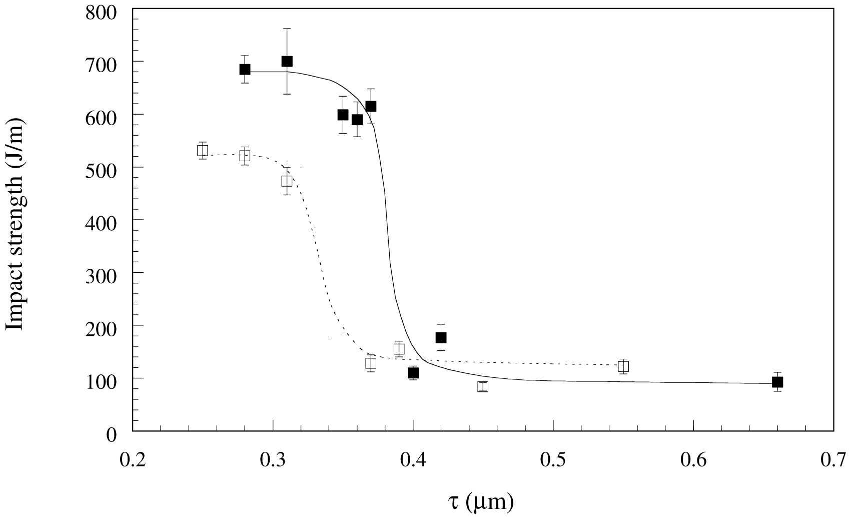

Fig. 8. Impact strength as a function of inter-particle distance for (PBT–Ph)/mPEO blends (■). Empty squares correspond to PBT/mPEO blends collected from Ref. [2].

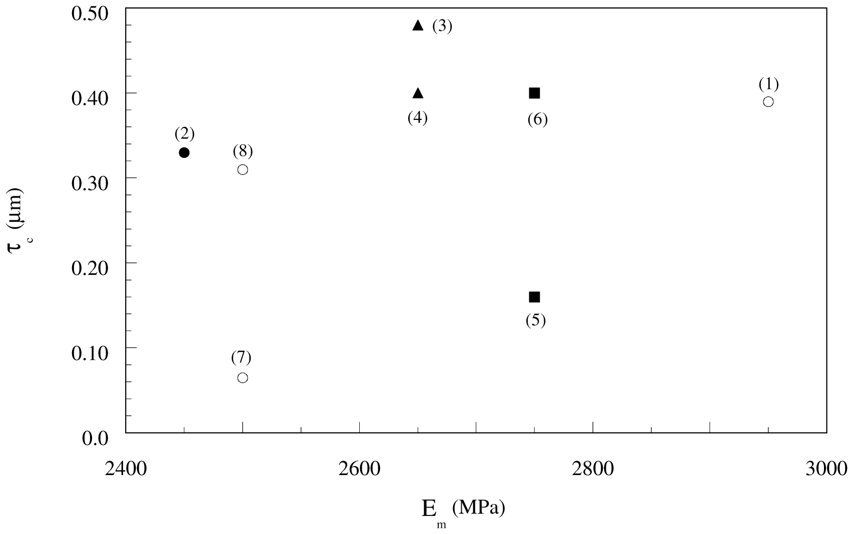

Fig. 9. τ_c as a function of E_m for (PBT–Ph)/mPEO (1), PBT/mPEO (2), PBT/PEO-epoxy (3), PBT/PEO-EGMA (4), PBT/SEBS (5), PBT/ethylene olefin (6), Nylon 6/silica (7) and Nylon 6/EPDM (8) blends that correspond to this work and to Refs. [2,4,5,17,39,21], respectively.

[17], type and modulus of the rubber [17,18,25], E_m/E_d [2] and adhesion between the components [3,4,26] also influence the τ_c value. Comparing τ_c of this study on (PBT–Ph)/mPEO blends and that of the former study of Ref. [2] on PBT/mPEO blends ($\tau_c = 0.33$ µm), the values are clearly different. Both studies were carried out under the same experimental conditions, and with the same dispersed phase. The nature of the matrix changed. This could indicate that τ_c depends either on the modulus of the matrix, or on parameters related to it such as E_m/E_d. The possibility of E_m being the parameter that controls toughness was studied in Fig. 9. As can be seen, the clear scattering of the data indicates that there is no direct relation between E_m and τ_c.

With respect to E_m/E_d, it is not the only parameter to change when Ph is added to PBT/mPEO blends. This is because, as a consequence of the presence of Ph in the matrix, and therefore, of the larger amount of hydroxyl groups, the interactions with mPEO will change, and therefore, adhesion, which can also influence τ_c, should change too. This was confirmed by the different interfacial tension between the two components of the blend of this study (0.55 mN/m) and that of Ref. [2] (0.25 mN/m).

Thus, the different τ_c of this study to that of Ref. [2] can be due to either the change in E_m/E_d alone, the change in adhesion (measured by the interfacial tension) alone, or both. To find out which of these three possibilities is most likely, the τ_c–E_m/E_d and the τ_c-interfacial tension relations of this study are compared in Fig. 10a and b, respectively, with other values obtained from the literature. Although the additional influence of interfacial tension should lead to scattering in the τ_c values, as can be seen in Fig. 10a, the relation between τ_c and E_m/E_d proposed in this work and in Ref. [2] is valid not only for the previous values (points 2, 5–8), but also for that of this study (point 1) and also for those of recent papers (points 3 and 4). This supports the proposed dependence of τ_c on E_m/E_d.

When the τ_c-interfacial tension data are shown in Fig. 10b, despite the usually not very accurate γ_{12} values, the relationship between τ_c and interfacial tension is also clearly depicted. Furthermore, in the τ_c and γ_{12} range studied, the relationship is reasonably linear. Thus, the dependence of τ_c on interfacial tension (adhesion) is also inferred.

4. Conclusions

Super-tough PBT based (PBT–Ph)/mPEO blends with notched toughness values higher than those of PBT/mPEO blends and 23-fold that of PBT, and with higher stiffness, were obtained by means of the addition of 20% Ph that miscibilized in the PBT matrix.

The minimum grafting level (0.32%) used is enough to achieve super-toughness (600 J/m) in the 15% mPEO blend, that showed a modulus of elasticity of 1.9 GPa. 15% mPEO is also the minimum rubber content required to achieve super-toughness. The maximum toughness (at least of 700 J/m) appeared at 20% mPEO with a modulus of elasticity of 1.7 GPa.

From an analysis of the τ_c-interfacial tension, and τ_c–E_m/E_d pairs from this study and from previous papers, the dependence of τ_c on both E_m/E_d and adhesion, measured by means of the interfacial tension between the components of the blends, is inferred.

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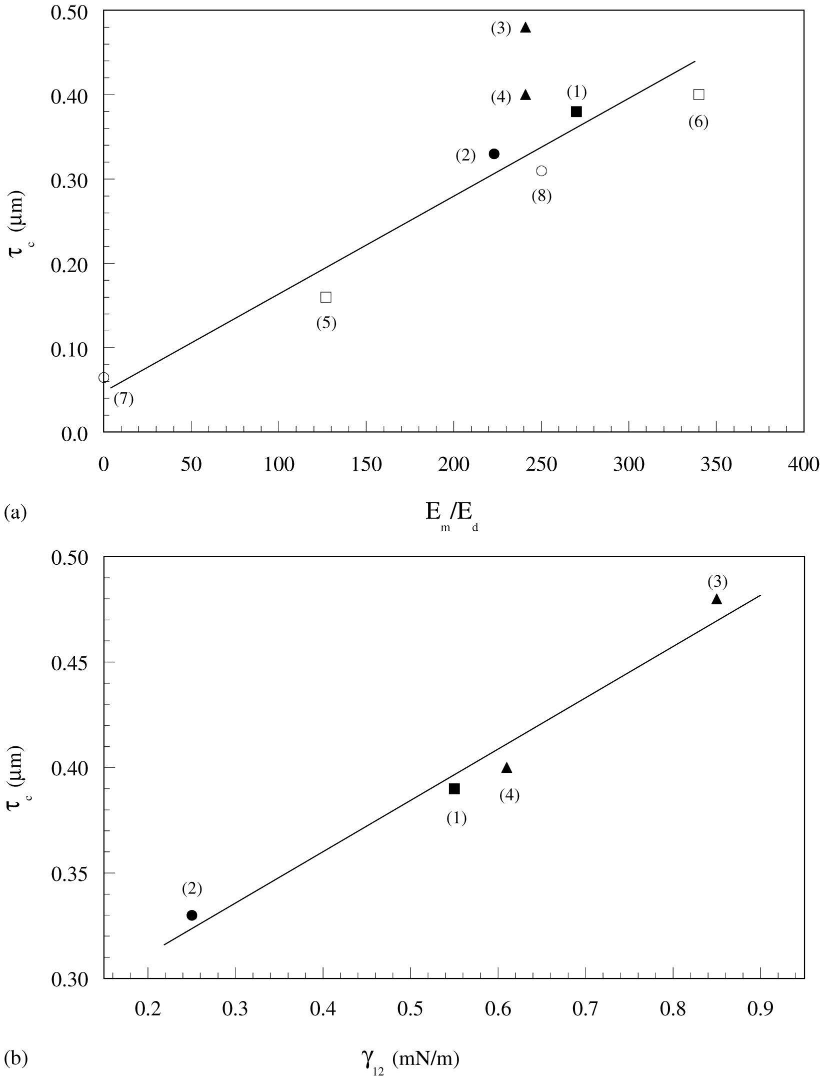

Fig. 10. τ_c as a function of E_m/E_d relation (a), and interfacial tension (b). Symbols and numbers as in Fig. 9.

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