Poly(ethylene terephthalate) (PET) based blends were obtained by melt blending PET with up to 30 wt% poly(ethylene-octene) either modified with maleic anhydride (mLLDPE) or not (LLDPE). Both PET/LLDPE and PET/mLLDPE blends were immiscible. The dispersed phase particle size was large in LLDPE blends, but upon mLLDPE addition, it decreased to a small (submicron) and rather constant value with composition. This indicated compatibilization, and was attributed to specific interactions between the ester and maleic groups of PET and mLLDPE, respectively, rather than grafting reactions between components. Linear decreases in Young’s modulus and yield stress, and ductility increases were observed in blends with mLLDPE. Super-toughness was achieved in blends with mLLDPE, which took place when the critical interparticle distance (IDc) was below 0.17 μm and with only half the cross section of the specimens broken. The IDc of these blends and those of other blends from bibliography were compared with the adhesion levels estimated from the expected main interactions between the components of the blends. This comparison strongly indicated that, at least through an adhesion range, IDc depends on the adhesion level, and that IDc decreases as the adhesion level increases.

INTRODUCTION

It is known that the applications of many semicrystalline thermoplastics are often limited by their low notched impact strength. This is the reason for the efforts devoted to obtain impact resistant polymers by blending them with impact modifiers, mainly rubbers. Recently, very high impact strength of the order of 10–20 fold that of the matrix (super-toughness) [1–7] has been obtained. The transition to super-toughness is mostly characterized by the critical interparticle distance (IDc) [1–3, 8–11] because the impact modifier particle size at which super-toughness appears depends on the impact modifier content but IDc does not [8, 12]. Super-toughness has been observed in thermoplastics that include poly(butylene terephthalate) (PBT) [1, 4, 8, 9, 13–15], poly(ethylene terephthalate) (PET) [12, 16–22], nylon [5, 6, 10, 23], polysulphone (PSU) [7], and polyolefins [2, 3], mostly thanks to the chemical modification of the impact modifier or the use of compatibilizers. A lot of different impact modifiers have been used for this purpose; mainly ethylene [1, 2, 6, 7, 12, 16, 17, 23] and styrene copolymers [3, 22] and ABS [4, 18].

When the parameters that influence IDc were studied, the influence of extrinsic parameters such as temperature [11, 24–27], strain rate [24, 27, 28], or notch radius [25] was claimed. However, the possible influence of intrinsic parameters is more difficult to study because the change in one of them is usually accompanied by a change in at least a second parameter. Parameters such as the modulus of the matrix and the impact modifier [13,14,29,30], the modulus ratio [8, 13, 14], or the adhesion between matrix and impact modifier [1, 9, 13, 15, 31] have been proposed to influence IDc. The dependence on the product of the yield strength and yield strain of the matrix has also been tentatively stated [32]. Literature results on adhesion are not, however, conclusive. When adhesion increased, IDc decreases were observed in several PBT/impact modifier [1, 9, 13, 15] and PVC/NBR blends [31]. In PBT/ethylene-propylene rubber [33] and in blends of semicrystalline PET with various modifiers [17], different levels of compatibilizer did not lead to changes in IDc. However, although adhesion probably changed, it was neither measured nor estimated. Therefore, much more work is necessary to find out whether and how adhesion influences IDc.

Thermoplastic polyesters have many applications as engineering polymers because of their combination of properties such as rigidity, solvent resistance and low permeability, and price. Among them, PET is the most used. To increase its low notched impact strength, it has been modified with ABS [18, 34], EPDM [35], SEBS [22, 36], EPR [12, 17, 19, 37], SBR [38], LLDPE [16, 39–41], and several copolymers [16, 20, 21]. Medium or low impact strength...
values were obtained when modified with EPR [12, 17, 19], ABS [18], SEBS [22], LLDPE [41], and ethylene-based terpolymers [20]. Very high (60-fold) increases were seen in PET with glycidyl methacrylate-grafted LLDPE (GMA-LLDPE) and with other impact modifiers [16], and also with a core-shell modifier [21]. Both amorphous [16, 18, 36] and semicrystalline [12, 17, 35, 40] PET were used. The influences on super-toughness of the crystallinity and the molecular weight of the matrix [12], the impact modifier content and particle size [12, 16–22], the interfacial characteristics [12, 16, 17, 19, 21, 22], and the deformation mechanisms [16, 17, 19, 22] have been discussed. The ID$_c$ for super-toughness to appear in PET has not been clearly determined, as a maximum impact strength was not apparently reached in PET/EPR [12, 17] blends and ID$_c$ was not accurately determined in amorphous PET modified with GMA-LLDPE and with other impact modifiers [16].

Among the impact modifiers used to modify PET, poly(ethylene-octene) (LLDPE) is a recently developed metalloenic impact modifier that has been successfully used to toughen, among others, PSU [7], polyamide 6 [42, 43], PBT [8, 13, 15], and PET [16, 40].Compatibilization is usually necessary and mostly attained through grafting reactions that result from the functionalization of the impact modifier [5–8, 13, 15, 16] or from the addition of a compatibilizer [9, 13, 15, 16, 40]. In the works devoted to PET/LLDPE blends, the micromechanical behavior [39], the modification of the blend was measured, and the adhesion estimated from the possibility of interaction based on the chemical structure. This is an alternative to the contact angle technique, which is an indirect measurement that can lead to a high experimental error. The ID$_c$ obtained and that of other blends in the bibliography, together with the estimated adhesion level, will be used to discuss whether the interfacial adhesion influences ID$_c$.

**EXPERIMENTAL PROCEDURES**

The poly(ethylene terephthalate) (PET) used in this work was BRIPET BST (Brilen Uldesta) ($M_n = 35,000$ and $M_w = 65,000$, approximately) and the impact modifier was an ethylene–octene copolymer, ENGAGE EG 8200 (Dupont-Dow) (LLDPE). Grafting of LLDPE was carried out with a commercial maleic anhydride (MA), and the peroxide initiator was dicumyl peroxide (Aldrich).

The grafting of LLDPE was carried out in a Collin twin-screw extruder-kneader (type ZK25) with 1–3 wt% MA, and with 0.05% of dicumyl peroxide. The temperature profile along the extruder was 50/140/155/170/170/175°C and the screw rotation speed was 70 rpm. The L/D ratio and the diameter of the screw were 24 and 25 mm, respectively. The rod extrudate was cooled in a water bath and then pelletized. To remove the unreacted MA from mLLDPE, the pellets were placed in methanol for 48 h [44] and then dried at 60°C for 2 days.

To determine the amount of grafted MA in mLLDPE, the titration method was used [38, 44–46]. The mLLDPE was dissolved in toluene at 100°C and then distilled water was added. An aqueous, a gel, and an organic phase containing the mLLDPE appeared. A known volume of this organic phase was titrated with 0.02 M KOH in ethanol. At least three measurements were made for each determination. In agreement with previous results [7, 8], the amount of mLLDPE was found to increase almost linearly with the amount of MA added, in the range studied. The most reacted mLLDPE (3 wt%) was selected.

PET and the modified (mLLDPE) and pure LLDPE were dried before processing in a dehumidifier for 12 h at 150°C in the case of PET, and in an air circulating oven for 6 h at 60°C in the case of both LLDPE and mLLDPE.

The PET/LLDPE and PET/mLLDPE blends were obtained in the twin screw extruder-kneader used for the grafting of LLDPE at a melt temperature of 275°C and a rotor speed of 70 rpm. Both the LLDPE and the mLLDPE content in the blends varied from 0 to 30 wt%. The rod extrudates were cooled in a water bath and then pelletized. The blends will be named according to their LLDPE or mLLDPE weight content. The volume content will be used in the case of mechanical properties due to the significant difference in density of the two components of the blends. The kneading torque of the 30 wt% LLDPE and mLLDPE blend was measured in a Brabender Plasticorder at 275°C and 30 rpm.

Tensile (ASTM D638 type IV) and impact (ASTM 256) specimens with 2 and 3.2 mm thickness, respectively, were obtained in a Battenfeld BA230E reciprocating screw injection molding machine. The screw had a diameter of 18 mm and a L/D ratio of 17.8. The temperature profile was 275/275/275°C and the injection speed and pressure were 20.4 cm$^3$/s and 2600 bar, respectively. The mould temperature was held at 15°C.

The phase structure of the PET/LLDPE and PET/mLLDPE blends was studied by dynamic mechanical analysis on a TA Instruments’ DMTA that provided the plots of the loss tangent (tan δ) and the storage moduli (E‘) against temperature. The scans were carried out in the bending mode at a constant heating rate of 4°C/min and a frequency of 1 Hz from −110 to 110°C. The crystallization degree was determined both by X-ray diffraction and density measurements. X-ray diffraction patterns were recorded in a Phillips PW
1729 GXRD X-ray diffractometer that worked at 45 kV and 50 mA, using a Ni-filtered Cu Ka radiation source. The scan speed was 0.5°/min. Density measurements were carried out by the displacement method in n-butyl alcohol. A Hitachi S-2700 electron microscope at an accelerating voltage of 15 kV was used. The impact modifier particle size was measured in representative zones of the cryogenically fractured impact specimens. The weight average particle size \( d_w \) was calculated from approximately 200 particles as follows

\[
d_w = \frac{\sum n d^2}{\sum n d}
\]

where \( n \) is the number of particles with size \( d \). This average particle size was used to calculate the average ID by the equation for dispersed spherical particles \[10\]

\[
\text{ID} = \frac{d}{\phi k } \left( \frac{\pi}{\phi} \right)^{\frac{1}{3}} - 1
\]

where \( \phi \) is the impact modifier volume fraction and \( k \) a geometric constant (equal to 1 for a cubic lattice).

The tensile tests were carried out in an Instron 4301 tensile tester at a cross-head speed of 10 mm/min and at (23 ± 2)°C. The mechanical properties (Young’s modulus, \( E \); yield stress and ductility, measured as the percent elongation at break) were determined from the load-displacement curves. The Young’s modulus was determined using an extensometer. Izod impact tests were carried out on notched specimens using a CEAST 6548/000 pendulum. The notches (depth 2.54 mm and radius 0.25 mm) were machined after injection molding. A minimum of five tensile and eight impact specimens were tested for each reported value.

**RESULTS AND DISCUSSION**

*Phase Structure*

The phase structure of the blends was studied by DMTA. Figure 1 shows the tan \( \delta \) vs. temperature plots of pure PET and mLLDPE and those of the 10, 20, and 30 wt% mLLDPE blends. The scans of the 5, 15, and 25 mLLDPE blends showed a similar behavior. The scans of the LLDPE blends were similar to those of Fig. 1, indicating that the phase behavior of the LLDPE and mLLDPE blends was the same. As can be seen, the pure PET showed two main transition peaks, the \( T_g \) at approximately 80°C, and a very broad and weak \( T_m \) centred at −55/−60°C. This \( T_m \) peak can hardly be observed in Fig. 1 due to the large intensity of the \( T_g \) peak.

Pure mLLDPE showed a clear \( T_g \) peak centered at approximately −38°C.

As can be seen in Fig. 1, the blends showed two main transition peaks. The high temperature one was centered, within the experimental error, at the \( T_g \) of PET. This indicates the presence in the blends of a pure amorphous PET phase. As can also be seen in Fig. 1, the low temperature peak appeared at a temperature range similar to that of the \( T_g \) of mLLDPE, indicating the presence of an amorphous mLLDPE phase. However, the peak extended towards temperatures below that of pure mLLDPE. As the peak slip was larger at increasing PET contents, it was attributed to the \( T_g \) peak of PET that was located at −55°C. The partial superposition of the \( T_g \) of mLLDPE and the \( T_g \) of PET in the blends complicated an accurate determination of the position of the \( T_g \) peak of mLLDPE. However, the position of the \( T_g \) peak of mLLDPE in the blends has to be that of pure mLLDPE. This is because when the composition-weighted addition of the tan \( \delta \) plots of pure PET and mLLDPE was calculated, the plots were very similar to those of Fig. 1. This indicated the presence of a pure mLLDPE phase. Although a decrease in the \( T_g \) of PET was observed in modified PET/EPR blends \[37\], the presence of two pure amorphous phases is the usual behavior in toughened thermoplastic blends, and in particular, in PET/impact modifier blends \[18,37\].

Figure 2 shows the X-ray diffraction patterns of the PET, mLLDPE and the 30 wt% mLLDPE blend as an example. As can be seen, no peak was observed in the pattern of the pure PET. This indicates its amorphous state, that is a consequence of the low mold temperature (15°C) and consequent rapid cooling in injection molding. The X-ray pattern of the pure mLLDPE showed a broad peak at 2θ values approximately between 10° and 30°, which indicates crystallinity. In the pattern of the blend no peak that could correspond to PET appears, indicating that the PET was also amorphous in the blends. A nucleation activity of the modifier particles has been observed in PET/impact modifier blends \[17, 34, 36, 37\]. The amorphous nature of PET was confirmed by density measurements of pure PET, LLDPE, and the blends because the density of the blends was close but below additivity. The crystallinity of mLLDPE was...
retained in the blends because the area of their diffraction peak corresponded to the composition-weighted addition of the peak of mLLDPE and the amorphous halo of pure PET.

**Morphology**

Figure 3a and b shows the main morphology of the cryogenically fractured impact specimens with respectively 20 and 30 wt% unmodified LLDPE. The 25 wt% LLDPE compositions showed similar morphologies. In the core of the specimens, larger particles than those of Fig. 3 appeared and the small particles almost disappeared. The absence of small particles indicated coalescence in this zone of the specimen, in which cooling was slower. Table 1 shows the weight average particle diameter of both LLDPE and mLLDPE blends.

As can be seen in Fig. 3 and also in Table 1, in the PET/unmodified LLDPE blends, the average particle size was very large at LLDPE contents above 15 wt%. This large particle size indicates a high interfacial tension and that compatibilization will be needed.

Figure 4a–c shows representative morphologies of cryogenically broken impact specimens of the 10, 20, and 30 wt% mLLDPE blends. The rest of the compositions showed similar morphologies. In the mLLDPE blends (Fig. 4 and Table 1), the average particle size was small (submicron) and rather constant with composition. The smaller particle size of the blends with mLLDPE indicated that the interfacial tension decreased and that compatibilization had taken place. Although, a reduction in particle size is increasingly difficult at low particle sizes, the particle size was not affected by compatibilization up to 15% LLDPE addition. A reduction in coalescence may have also taken place [47] in the mLLDPE blends. This is supported by the fact that, while small particles were present in the center of the specimens of the blends with mLLDPE, just in the center of the LLDPE blends the presence of small particles was rare, indicating that coalescence had occurred.

Consequently, adhesion in the solid state increased in mLLDPE blends. It is known that low adhesion as that provided by Van der Waals attraction is enough for toughening [10, 23]. In this way many LLDPE particles are pulled out in the mLLDPE modified blends; however, many other

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**TABLE 1.** Weight average particle diameter (Eq. 1) (μm) in PET/LLDPE and PET/mLLDPE blends.

<table>
<thead>
<tr>
<th>Wt%</th>
<th>PET/LLDPE blends</th>
<th>PET/mLLDPE blends</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>0.70</td>
<td>0.65</td>
</tr>
<tr>
<td>10</td>
<td>0.70</td>
<td>0.71</td>
</tr>
<tr>
<td>15</td>
<td>1.00</td>
<td>0.78</td>
</tr>
<tr>
<td>20</td>
<td>2.00</td>
<td>0.73</td>
</tr>
<tr>
<td>25</td>
<td>2.60</td>
<td>0.76</td>
</tr>
<tr>
<td>30</td>
<td>3.00</td>
<td>0.85</td>
</tr>
</tbody>
</table>

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appeared broken (Fig. 4c mainly), indicating cohesive fracture. This indicates that in these blends the adhesion level is not as low as usual in impact modified blends. The presence of an intermediate adhesion level in these blends was corroborated by the elongation at break results. As a small particle size is definitive to achieve good mechanical properties, the maleinization of LLDPE appears to be necessary in these blends. Particle sizes from roughly 2 to 5 μm were reported in the bibliography in other unmodified PET/impact modifier blends at impact modifier contents above 5%, depending on the processing procedure [12, 17, 19]. However, when compatibilized, the particle size was typically below 1 μm [12, 16, 17, 19, 36].

The decrease in interfacial tension in the blends with mLLDPE can be due to (i) reactions between the matrix and the impact modifier, giving rise to graft copolymers, (ii) specific interactions between the ester and maleic groups of PET and mLLDPE, respectively or (iii) both of them. When grafting reactions take place, the melt viscosity and therefore the torque of kneading should increase along time. This is why the kneading torque of the 30 wt% LLDPE and 30 wt% mLLDPE blends was measured at 275°C for 12 minutes. A slight torque increase appeared in mLLDPE blends after 8 min, which is less than the estimated residence time in the extrusion/injection process. However, a similar torque increase was also seen in the LLDPE blends. As the torque increase was small, the occurrence of cross-linking that could have serious consequences for properties is discarded. The similarity in torque behavior rules out the possibility of grafting reactions and points to degradation reactions as the origin of the torque increase in the blends. Furthermore, as no significant grafting was observed by FTIR in the chemically similar PBT/mLLDPE blends [8], chemical interactions between PET and the mLLDPE are proposed as the reason for the interfacial tension decrease.

The fracture surface of the center of the 25% mLLDPE impact specimens is shown in Fig. 5. Fracture propagated from the top to the bottom of Fig. 5. The fracture characteristics are those most usually seen at the fracture surface. In zones far from the notch the fibrillation was more devel-
oped, while close to the notch the fracture surface was less fibrillated than in Fig. 5. These differences can be attributed to a different crack propagation speed. The fracture surfaces of both pure PET and the blends with low rubber content were brittle. As can be seen in Fig. 5, the fracture surface was highly irregular. The fractured matrix was extensively deformed and was slightly fibrillated. All these characteristics are those of a ductile failure. They indicate that a large impact energy was involved to deform the fracture area and to create such a large new surface. Although the highly deformed matrix could occlude dispersed particles, the cavitation of the rubber particles was a generalized process as a large number of holes appeared. However, the surface of the holes, not clearly observed in Fig. 5, was smooth, in agreement with the stated not very low adhesion. The cavities are far from spherical, indicating that they deformed during the impact test.

**Tensile Properties**

Figures 6 and 7 show, respectively, the Young’s modulus and the yield stress of the blends vs. composition. The values of the LLDPE blends appear slightly above those of the mLLDPE blends, but the differences are less significant because most of them were less than the experimental error. Moreover, it is known that adhesion (maleinization) does not affect either the Young's modulus or significantly the yield stress because of the low strains at which they are measured. As can be seen, the two properties decreased with the impact modifier content because of the elastomeric nature of the dispersed phase. As can also be seen, the modulus behavior was rather close to the direct rule of mixtures. The lack of defined yield point in the case of LLDPE prevented the application of the rule of mixtures to the yield stress. The usual modulus behavior of other PET/impact modifier blends [17, 18] follows the direct rule of mixtures, but in previous PET/GMA-LLDPE blends [16] it was on average 10% below additivity. To discover the structural origin of the modulus values, the main parameters that influence the modulus of elasticity, i.e., the mixing-induced change of specific volume and of orientation were measured. When the phase behavior was studied, it was seen that the density of the blends was below but close to linearity (mean deviation, 0.017 g/cm³). When the orientation was measured, no significant change was observed. Therefore, the lack of significant deviation from linearity of the modulus is a consequence of the lack of change of free volume and orientation of the blend components upon melt mixing.

Figure 8 shows the ductility of LLDPE and mLLDPE blends against the impact modifier content. As can be seen, the ductility of the unmodified LLDPE blends was initially slightly higher than that of pure PET because of the presence of LLDPE, and the small particle size, but then abruptly decreased at LLDPE contents above 20% (particle size approximately 2 μm). Decrease in ductility on impact modifier addition have been observed in uncompatibilized PET/impact modifier [17] and PET/LLDPE [16] blends. In other PET/impact modifier blends [17] ductility decreased at different particle sizes. Therefore, besides the dispersed particle size, other parameters must influence ductility.
In the mLLDPE blends, ductility increased above 5 wt% mLLDPE and furthermore was sustained at higher LLDPE contents (particle size always below 1 μm). Therefore, as expected, maleinization and the concomitant decrease in both interfacial tension and particle size, clearly favored the deformation ability of the blends. Upon compatibilization, ductility values higher than that of the pure PET matrix were obtained in PET/impact modifier [16, 36] blends (particle size below 0.40 μm), and a high ductility independent of the impact modifier content in amorphous PET/GMA-LLDPE blends [16] (particle size below 0.45 μm).

**Notched Impact Strength**

Figure 9 shows the Izod notched impact strength of both LLDPE and mLLDPE blends vs. the impact modifier content. As can be seen in uncompatibilized LLDPE blends, although increases up to 170 J/m were obtained, super-toughness was not achieved. However, in the case of mLLDPE blends, the impact strength increased at impact modifier contents above 20 vol%, up to typical super-tough values (800 J/m). This is 60-fold the impact strength of pure PET. Moreover, the 25 and 30 wt% mLLDPE specimens of this study did not completely break (the unbroken area was higher than 50% in the 30 mLLDPE blend). Therefore, the values of this study are minimum values, because additional energy would be necessary to fully break the specimens. Although only a 10-fold increase in impact strength was obtained in previous PET/LLDPE blends [41], the usual increase in PET/impact modifier blends was 15–35 fold [12, 17–20]. The minimum impact modifier content to attain super-toughness was similar to that of this work. In PET/GMA-LLDPE [16] and PET/core-shell polyester [21] blends, 60-fold increases were seen, but the specimens broke completely [21], or had a broken area above 90% [16]. This indicates an impact strength smaller than that of the blends of this study. The impact modifier content (and consequent modulus of elasticity) at which super-toughness occurred in this study was similar to that of PET/GMA-LLDPE [16] and smaller (higher modulus of elasticity) than in PET/core-shell polyester blends (35wt%) [21].

In this study the modifier particle size was almost the same (0.65–0.85 μm) in all mLLDPE and low-impact modifier content LLDPE blends. However, only the 25 and 30 mLLDPE compositions showed super-toughness. For example the 15 and 25% mLLDPE blends have the same particle size (0.78 and 0.76 μm, respectively), but the impact strength changed from 90.7 to 741 J/m proving that the particle size is not a parameter that plays a major role in the impact strength of these blends. Therefore, the IDc [10,11], whose critical value (IDc) is independent of the impact modifier content, was measured and is shown vs. the impact strength in Fig. 10. As can be seen, a clear transition (inflexion point) to super-toughness took place at IDc = 0.17 μm in mLLDPE blends. In a previous work on blends of PET with LLDPE [16], IDc was below 0.10 μm. However, this value was obtained from blends with different rubbers and modifiers and therefore, probably with different interfacial adhesion levels. The interfacial adhesion has been shown to affect IDc [1,9,13,15,31]. It should affect the stress transmission through the interphase and therefore, the impact modifier particles cavitation, that is a condition for toughening [48]. In blends of PET with various modifiers [17], the reported IDc of PET was 0.10 μm. This difference would be higher if the IDc were measured at the inflexion point, as in this paper, instead of at the onset of the brittle-
tough transition. The PET crystallinity, the impact modifier, and the compatibilization method (and therefore, respectively, the modulus of the matrix and of the dispersed phase and, probably, the adhesion), were different in the study of Loyens and Groeninckx [17] and in this study. All these parameters have been proposed to influence IDc [8,13–15,29–31], either directly or through $E_m/E_d$ and will account for the observed difference.

Considering the bibliography on toughened LLDPE blends, there are a number of matrices where super-toughness was achieved at similar extrinsic parameters, but IDc was different to that of this study. Moreover, the matrices have a similar modulus to that of PET. Therefore a possible influence of the modulus of elasticity [13,14] or parameters related to it, as the rigidity of the chain [49] whose relation with the IDc is not clear [13], can be left out. Therefore, as the intrinsic parameters considered to possibly influence IDc (modulus of the matrix, of the modifier and their ratio) are the same with the exception of adhesion, this last one appears as the only parameter that could lead to a change in IDc. Consequently, the IDc of these blends offers a clear opportunity to study the dependence of IDc on adhesion. The main interactions (and therefore, adhesion) will be estimated from the chemical structure of the components of the blends and will be compared with the observed change of IDc. These matrices are PBT [8], Polysulphone of Bisphenol A (PSU) [7], and PBT mixed with either Polyarylate of Bisphenol A (PAR) [15] or Poly(hydroxy ether of Bisphenol A) (Ph) [13]. In PBT-Ph/mlLDPE blends, reactions are fast and occur between the two components of the matrix, therefore, the adhesion between the matrix and the dispersed phase is difficult to evaluate and will not be discussed. There was no reaction in PBT/PAR blends after 100 min, in the melt state [50], and therefore, they will be discussed.

In the case of PBT/mlLDPE blends [8], the IDc was clearly higher (0.33 μm) than that of this study (0.17 μm). The more rigid molecular backbone of PET may account for the change of IDc [49]. However, another molecular characteristic, such as the crystallinity level, may also have an influence. It seems that both characteristics have comparable effects because the modulus of elasticity of PET and PBT are very similar. Looking at other possible reasons, both in PET/mlLDPE and PBT/mlLDPE blends, the interaction must be between the ester groups of either PET or PBT and the maleic groups of the mlLDPE. As the relative amount of ester groups is higher in PET, in PET/mlLDPE blends the adhesion should be larger. This is in agreement with previous results in PBT blends [1,8], where a higher adhesion led to a lower IDc.

Comparing the estimated interacting ability of PBT with mlLDPE (IDc = 0.33 μm) and with the 90/10 PBT/PAR matrix (IDc = 0.43 μm) [15], both PBT and PAR have two interacting ester groups per repeating unit. However, the volume of the Bisphenol A unit of the PAR is larger than that of the butanediol of PBT. This leads to a decrease in the density of interacting groups per volume unit. Therefore, both the interactions and the adhesion should be larger in the PBT/mlLDPE blends, just where IDc was smaller.

In the case of PSU/mlLDPE blends [7], the interacting ability of the sulfone groups with mlLDPE seems to be lower than that of the ester groups. Moreover, the volume of the repeating unit is larger than that of PBT. These two facts point to the PSU-mlLDPE interactions being the weakest in the blends considered. As the IDc = 0.57 μm of these blends is the highest, the proposition of a higher adhesion leading to a lower IDc is additionally supported.

CONCLUSIONS

In PBT/LLDPE blends, both maleinized (mlLDPE) and nonmaleinized, two pure amorphous phases were present and the presence of the impact modifier particles did not affect the amorphous nature of PET. In PBT/mlLDPE blends, the particle size was smaller and rather constant with composition, indicating compatibilization and an enhanced matrix/impact modifier adhesion. Compatibilization by LL-
DPE maleinization was attributed to specific interactions between the matrix and the impact modifier rather than grafting reactions as no torque increase was seen.

Compatibilized PET/mlLDPE blends showed super-toughness (impact strength >800 J/m and 60-fold that of the matrix) when ID decreased below a critical value IDc = 0.17 μm. Moreover, contrary to previous works, a large part of the cross section of the specimens (roughly 50%) remained unbroken. When the IDc of this study and those of other thermoplastic/mlLDPE blends were compared with the corresponding adhesion levels estimated from the chemical interactions, it appeared that increases in the adhesion level came accompanied by decreases in IDc. Thus, it is proposed that, at least within an adhesion range, IDc depends on the adhesion level and that a higher adhesion leads to a smaller IDc.

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