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The role of poly(ethylene terephthalate-co-isophthalate) as interfacial agent in polypropylene–matrix composites

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Abstract The effect of modifying the particle/matrix interfacial region on the morphology and tensile behaviour of glass bead-filled polypropylene (PP) composites was studied. The interface modification was promoted by blending PP with a small concentration (5% by weight) of poly(ethylene terephthalate-co-isophthalate) (co-PET). Ten different PP/co-PET/glass beads ternary composites were prepared, characterized and compared with the homologous PP/glass beads binary ones. Maleic anhydride-grafted PP was added as a compatibilizing agent for PP and co-PET in some of the studied formulations, and its effect studied. Furthermore, four different silane-treated glass beads were used to prepare the composites (50 wt.%). Results showed that three different interfaces, corresponding to three different levels (low, middle and high) of particle/matrix adhesion, could be obtained in these composites by varying the matrix composition and the silane coupling agent on the glass bead surface, which resulted in a wide range of tensile properties, from ductile composites with low tensile strength and high elongation to brittle ones with high tensile strength. It was found that co-PET embeds glass bead surface independently of the silane coupling agent employed. Finally, the adhesion degree differences between the different composite phases seemed to be the main cause to explain the differences found in the sensitivity of the composite tensile characteristics to the strain rate.

Introduction

It is well known that mechanical behaviour of polypropylene (PP) composites is greatly affected by the physical characteristics of the interfacial region. One of the most important factors affecting the interface properties is the adhesion between the polymer and the surface of filler particles or fibres [1–3].

Usually, the way to achieve good adhesion between mineral fillers or glass fibres and PP involves actions concerning both the filler and the polymer. On the one hand, glass fillers (beads, flakes or fibres) are usually surface-treated with organofunctional silanes [4] or other coupling agents. Silanes may chemically bond to the glass surface through condensation reaction. Under certain conditions functionalized silane may react chemically with PP [5]. Silanes do not form physical entanglements that could improve the cohesive strength of the material [6]. High molecular weight adhesion promoters are then preferred to promote physical entanglements with the matrix in the bulk. On the other hand, reactive groups in functionalized PP (e.g. maleic anhydride-grafted PP (MAPP) [6, 7]) may absorb on the high energy surface of the filler and react chemically. However, this is not the only way if PP is effectively compatibilized with other functional polymers. It was proposed the use of some polar thermoplastic polymers, such as polyamide [8] or

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polycarbonate [9], as efficient adhesion promoters in filled PP composites. They increase the surface energy of the polymer matrix. In addition, it has also been proposed that they could bond to the glass surface through condensation reaction with the hydroxyl groups. Ideally, in these cases, the treatment of the filler particle surface with silane could not be required to improve the interfacial adhesion.

Following the former idea, we analyse in this paper the use of a poly(ethylene terephthalate-co-isophthalate) (co-PET) to modify the mechanical properties of glass bead-filled PP composites. Depending of the adhesion level promoted different tensile behaviour can be observed in these composites, from ductile one having low tensile strength to brittle one with high tensile strength, even slightly higher than that of the unfilled polymer.

The affinity of PET for glass surface has been reported [10]. So, it could be suitable to modify the interface characteristics and/or the adhesion degree in glass bead-filled PP composites. However, the addition of PET into a PP matrix has some drawbacks due to the poor compatibility between both polymers [11–13]. This can be overcome by adding into the blend a compatibilizing agent. The addition of several types of PP graft copolymers, like those having acrylic acid [13], maleic anhydride groups [14–17], or glycidil methacrylate [18] resulted in finely dispersed phases in the PP/co-PET blend, showing that these copolymers were able to compatibilize the blend. This compatibilizing efficiency has also been displayed by styrene-b-(ethylene-co-butylene)-b-styrene (SEBS) grafted with maleic anhydride groups (SEBS-g-MAH) [19, 20]. Lepers et al. [21] found that SEBS-g-MAH prevented of coalescence of the dispersed phase in the PP/PET blend.

In general, glass bead-filled composites are easily processed and have small and well-distributed internal stress, high dimensional stability and good service performance. Most of the published papers dealing with glass bead-filled PP composites were focused on aspects related with the particle size and the filler concentration effects [22]. So, increasing the glass bead diameter led to a build-up in the composite stiffness [23], although the tensile strength, the elongation at break [23, 24], and the fracture toughness [25] diminished. Similarly, increasing the glass bead concentration resulted in a stiffness increase and in both tensile strength and elongation at break decrease [23, 24, 26]. Izod impact strength was found to be reduced [24]. A brittle-ductile transition located at 10% by volume of glass beads was also reported [27].

The aim of the present paper is to show the effects of blending PP with a small percentage of co-PET on the morphology and tensile behaviour of glass bead-filled PP composites. Both untreated and different silane-treated glass beads were employed. The effects of a compatibilizer agent (MAPP) and four different silane coupling agents were analysed. The crystalline microstructure and some thermomechanical properties of the blend PP/co-PET (95/5) are shown elsewhere [28].

Materials, compounding and specimens

Polypropylene was provided by *Repsol-YPF* (Puertollano, Spain). It was an homopolymer grade (*Isplen PP050*) with melt flow index (conditions: 230 °C and 2,160 g) 5.0 g/10 min. Glass beads with average particle size of 20 μm were employed as filler, being kindly provided by *Sovitec Ibérica, S.A.* (Castellbisbal, Spain). *Eastman Chemical* (Madrid, Spain) supplied a commercial grade (*Epolene G-3003*) of maleated polypropylene (MAPP) of acid number 8. *Extrupet EW36* was a poly(ethylene terephthalate-co-isophthalate) (co-PET) manufactured by *Catalana de Polímers S.A.* (El Prat de Llobregat, Spain), with intrinsic viscosity 0.8 dL/g.

Four different series of composites were prepared with four different matrixes: Series A was the 100% PP-based series; the matrix of series B was a blend PP/MAPP (97/3); composites of series C were prepared with a blend PP/co-PET (95/5) and series C with the blend PP/MAPP/co-PET (92/3/5). All the blends and composites were prepared by melt extrusion. The neat PP was also extruded in the same conditions to comparison proposals.

Both untreated- and silane-treated glass beads were employed as filler (50% by weight in the composite). In order to analyse the eventual influence of adhesion promoters on the glass surface on the composites morphology and properties, four organosilanes with different reactivity for the polymer matrix were tested (Table 1). Trialkoxysilanes, $RSi(OR')_3$, hydrolyse stepwise in water to give the corresponding silanols, which ultimately condense to siloxanes. The hydrolysis is relatively fast, while the condensation reaction is much slower. When the prehydrolysed silane is applied to siliceous surfaces, silanol groups are also expected to condense with hydroxyl groups of the filler surface during drying operations. According to Plueddemann [4] the siloxanes formed on the glass surface having small alkyl substituents (methyl, vinyl, propyl, ...)

Table 1 Reference and type of silane on the glass bead surface.

GB surface treatment reference	Silane-coupling agent	Commercial name
1	–	–
2	3-methacryloxypropyltrimethoxy silane	Z-6030[a]
3	3-mercaptopropyltrimethoxy silane	A-189[b]
4	*N*-(2-aminoethyl)-3-aminopropyl trimethoxy silane	Z-6020[a]
5	*N*-(2-(vinylbencylamino)-ethyl)-3-aminopropyltrimethoxy silane	Z-6032[a]

Organosilanes supplied by [a] Dow Corning and [b] Witco

normally condense to highly cross-linked insoluble siloxane films; however, siloxanes having larger organic groups on the silicon, like those used here, are more highly cyclized and less highly cross-linked, and therefore are expected to be soluble, fusible and easier to interdiffuse with the polymer matrix.

Silane Z-6030 (methacrylate) is among the best known silane-coupling agents for polyesters. Polyolefins filled with siliceous minerals have shown improvement through the addition of methacrylate-functional silane [29]. It is believed that methacrylate silanes and aminoalkyl silanes form interpenetrating polymer networks with polyolefins at the interface [30], while vinyl silanes have ability to graft to organic polymers during the high temperature shear [29], and they copolymerize readily with maleic anhydride. Vinylbenzyl cationic silane (Z-6032) also may homopolymerize to form interpenetrating polymer networks. This silane is known to be an effective adhesion promoter for virtually all polymers to most mineral fillers. It has been observed that mechanical properties of Z-6032-treated mineral-filled PP increased with increasing adhesion [31]. Aminoalkyl silanes are used with a high number of polymers, including condensation-thermoplastics like linear polyesters and polyamides. They react normally with cyclic carboxylic anhydrides to form the corresponding amic acids [4]. Finally, the mercaptofunctional silanes (A-189) react readily to acrylates, methacrylates, itaconates, fumarates, etc; therefore it would be expected to add to MAPP.

The following procedure was used to homogeneously coat the glass bead surface with the silane: a solution containing 30 mL of silane, 250 mL of methanol, 60 mL of water and 5 mL of acetic acid was prepared by 1.5 kg of glass beads. This solution was stirred for 20 min to assure silane alkoxy group hydrolysis. The solution was transferred into a flask and glass beads were then added gradually while stirring. After decantation the mixture was heated up to 40 °C and maintained at this temperature for 24 h to evaporate the residual solvent. Stirring was continued as long as the viscosity of the mixture was low enough. The non-reacted silane was released by washing out three times with methanol. Under these conditions, an average concentration of silane of 1.8 wt.% was determined on the glass bead surface from ignition loss measures. Based on this concentration and on silane and microsphere characteristics an average surface coverage could be estimated to be comprised between 10 and 20 monolayers.

Composites were prepared using a Collin ZK-35 co-rotating twin-screw extruder (screw diameter 25 mm and L/D ratio 36). The co-PET was previously dried for a minimum of 4 h at 160 °C employing air of dew point –40 °C. The extrusion temperature profile was from 150 °C at the entrance to 250 °C at the die, and the screw speed was fixed at 120 rpm. Vacuum devolatilizing was applied. A circular cross-section die of 3 mm diameter was employed, and the extrudate obtained was cooled in a water bath and pelletized.

Tensile dumbbell-shaped specimens (type I according to ASTM D638) were injection-moulded in a multipropose mould (Fig. 4 of the ASTM D-647 standard) using a *Mateu & Solé 440/90* injection-moulding machine. The temperature profile in the cylinder was 150–220–230–240–250 °C, the mould temperature was fixed at 60 °C and the nominal injection pressure was 100 MPa. All the specimens were annealed at 110 °C for 24 h.

Testing procedure

Tensile tests

Tensile tests were carried out at different crosshead speeds (1, 5, 10, 50, 100 and 500 mm/min) on a *Galdabini Sun 2500* universal testing machine. A video-extensometer was adapted for the elongation measurements. The temperature of the test was 20 ± 2 °C. From the obtained tensile curves, Young's modulus (E), tensile strength (σ_{max}) and elongation at break (ε_b) average values were compiled. At least six specimens were tested for each composition.

Scanning electron microscopy

To investigate morphological aspects associated to the phase composition, as well as with the deformation and the fracture processes, a *Jeol-820* scanning electron microscope (SEM) was employed. Vacuum coated with gold was applied on the fracture surfaces to achieve optimal conductivity.

Raman spectroscopy

Raman spectra of unfilled blends were recorded on a *Dilor XY* Raman spectrometer powered by a He/Ne laser and operating at a wavelength of 632.8 nm. The laser was polarized vertically and the instrument operated exclusively in backscatter.

Results and discussion

Young's modulus

In the unfilled blend, the co-PET presence contributed to increase slightly the stiffness (Table 2), because of its higher Young's modulus than that of the neat PP. The addition of MAPP did not result in further stiffness build-up as would be expected from the compatibilizing effect of the MAPP. Champagne et al. [18] found similar rigidity values in blends PP/PET with and without PP-g-GMA as compatibilizer, concluding that both blends displayed similar elastic properties.

As expected, from the Young's modulus values of the glass bead-filled composites it is noticed that the addition of glass beads results in a remarkable increase of rigidity if compared with that showed by the unfilled PP (E = 1.8 GPa), which is obviously due to the higher rigidity (E = 72.6 GPa) of glass beads. Silane-based surface treatment of glass bead did not lead to significant differences in the Young's modulus values. Depending on the type of filler and its surface treatment some contradictory results can be found in the literature. So, enhanced interfacial PP/mica adhesion was reported [32] due to mica surface treatments, leading to increased rigidity. In other work [33] no differences in the Young's modulus of a series of magnesium hydroxide-filled PP are reported when the filler was surface-treated with stearic acid.

Furthermore, no remarkable differences are found in the composites due to the different polymer/matrix composition. Thus, it is deduced that the high filler content rules the rigidity of the composite, hiding aspects related to the matrix modification.

Tensile strength and elongation at break

The tensile strength is the most dependent mechanical property of the interfacial adhesion [34] in a composite

Table 2 Average values and standard deviation of the composite tensile properties

Sample	Young's modulus, E^a (GPa)	Tensile strength, σ^b_{max} (MPa)	Elongation at break, ε^b_b (%)
PP	1.5 (0.1)	37.1 (0.3)	75.0 (9.1)
A1	3.7 (0.1)	21.3 (0.1)	58.3 (5.4)
A2	3.6 (0.2)	20.7 (0.1)	55.0 (3.7)
A3	3.6 (0.1)	20.2 (0.1)	59.3 (5.0)
A4	3.2 (0.2)	22.2 (0.1)	31.0 (3.6)
A5	4.2 (0.4)	29.1 (0.8)	2.1 (0.1)
PP/MAPP	1.5 (0.1)	36.8 (0.2)	70.1 (8.0)
B1	3.1 (0.4)	29.5 (0.1)	2.5 (0.2)
B2	3.3 (0.1)	26.7 (0.1)	2.9 (0.3)
B3	3.7 (0.1)	25.5 (0.1)	3.0 (0.6)
B4	3.8 (0.2)	37.6 (0.1)	5.0 (0.1)
B5	3.6 (0.3)	38.0 (0.1)	5.5 (0.1)
PP/co-PET	1.7 (0.2)	36.8 (0.2)	31.5 (3.6)
C1	3.5 (0.1)	20.1 (0.1)	33.5 (4.1)
C2	3.2 (0.3)	20.8 (0.1)	44.2 (2.1)
C3	3.2 (0.1)	19.5 (0.1)	46.9 (5.2)
C4	3.2 (0.4)	19.7 (0.1)	50.6 (2.2)
C5	3.1 (0.2)	21.4 (0.2)	17.9 (2.5)
PP/MAPP/co-PET	1.7 (0.4)	38.6 (0.5)	29.1 (8.8)
D1	3.2 (0.1)	37.5 (0.3)	3.3 (0.3)
D2	3.2 (0.1)	38.4 (0.1)	3.9 (0.3)
D3	3.1 (0.3)	36.3 (0.1)	3.1 (0.2)
D4	3.1 (0.2)	39.0 (0.1)	6.4 (0.6)
D5	2.91 (0.2)	37.8 (0.2)	4.0 (0.1)

[a] Measured at 1 mm/min

[b] Measured at 50 mm/min

material. The values of tensile strength and elongation at break are compiled in Table 2.

In the unfilled non-compatibilizing blend, the addition of co-PET to PP resulted in decreased tensile strength and elongation at break, due to the poor compatibility between both polymers [12, 14]. So, in Fig. 1a it can be appreciated that the dispersed co-PET domains completely debonded from PP matrix. The compatibilizing role of MAPP resulted in slightly increased tensile strength if compared the blend with pure PP. A similar result was reported for PP/PET blends having small amounts of methylmethacrylate or ethylacrylate-PP [35]. The elongation at break diminished as a result of the immobilized PP chains near the interface. In Fig. 1b it can be appreciated that the co-PET domains are being well bonded to the PP (compatibilized).

Following, the results of the glass bead-filled composites are presented and discussed. For simplicity proposals they have been divided into two groups, depending on the presence or not of co-PET in the matrix composition:

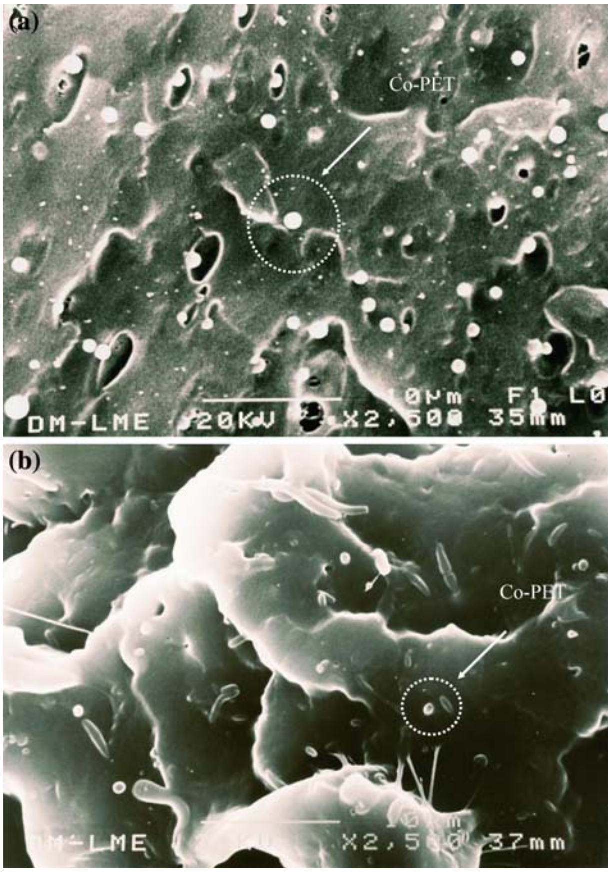

Fig. 1 Fracture surfaces of the unfilled samples: (**a**) PP/co-PET and (**b**) PP/MAPP/co-PET

PP/glass microspheres composites

At sight of the tensile strength values compiled in Table 2, three different degrees of particle/matrix adhesion [1] seem to be in these composites (Fig. 2a). The low interfacial adhesion level (poor adhesion) correspond to low values of tensile strength along with

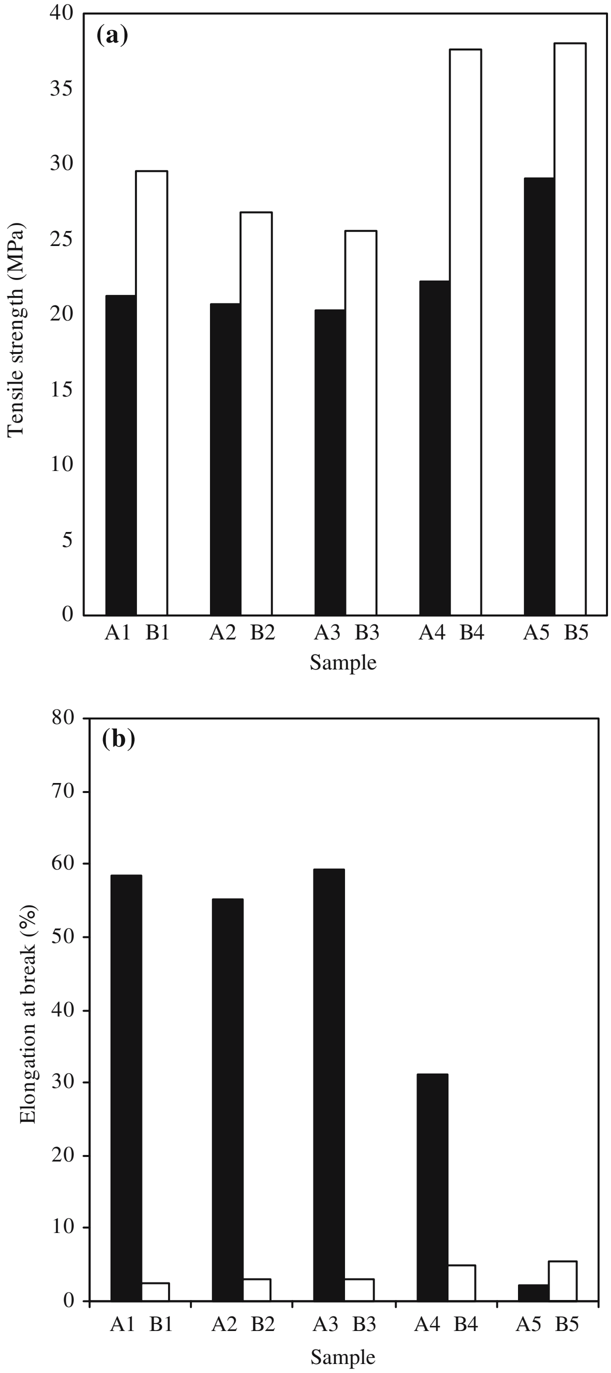

Fig. 2 Tensile strength (**a**) and elongation at break (**b**) of PP (■) and PP/MAPP (□) matrix-based composites

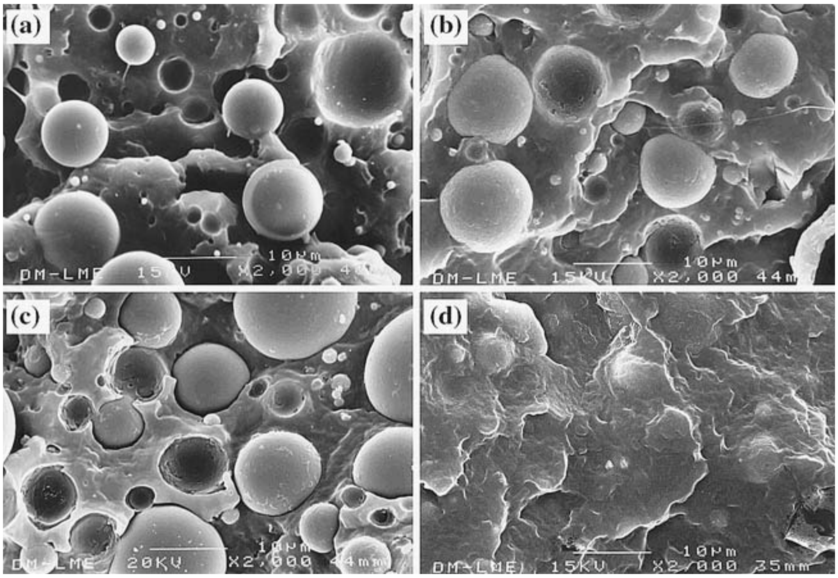

Fig. 3 Fracture surfaces of (**a**) A1, (**b**) A5, (**c**) B1 and (**d**) B4 composites

high values of deformation at break, and it is accomplished by composites having low debonding strength (Fig. 3a). This behaviour is displayed by the composites based on pure PP matrix. Only silane Z-6032 was able to slightly improve the interfacial adhesion degree up to the intermediate adhesion level. The relatively high plasticity displayed by these composites would come as result of a plane stress state localized between adjacent microspheres once particle debonding occurs [23].

The intermediate adhesion level is characterized by a less marked tensile strength drop with respect to the pure PP, as well as by a quasi-brittle failure. This behaviour is accomplished by composites A5, B1, B2 and B3. As noticed before, only Z-6032 silane promoted some improved adhesion between glass surface and pure PP, probably due to silane grafting carried out during the melt compounding by a mechanism through radicals, involving the vinyl group of this silane. It has been reported [5, 36] the fast reaction kinetics of silane Z-6032 amino groups with PP chains, as well as the general efficiency of silanes containing primary amine groups. In the present research, a silane containing primary amine group (Z-6020) different from Z-6032 did not result efficient in promoting adhesion between PP and glass surface, although it displayed the highest tensile strength values and the lowest elongation at break within the composites accomplishing the low adhesion level.

In B1, B2 and B3 samples carboxyl groups of MAPP would interact with hydroxyl groups of untreated glass bead surface and with methacryloxy and even with mercapto groups of silanes Z-6030 and A-189, respectively. These interactions would not be strong enough to reach in these composites the tensile strength levels of neat PP, although the elongation at break values fell down, resulting in quasi-brittle failure. As it can be observed in Fig. 3b and c, the samples that satisfy the medium adhesion level (A5 and B1) show less debonded microspheres than the samples with low interfacial adhesion (Fig. 3a), and the microspheres appear more coated by the matrix, which is an evidence of stronger interface.

Finally, two samples (B4 and B5) exhibited tensile strength values similar to those of pure PP, revealing high adhesion level and the strongest interface. The fracture surfaces of these composites (Fig. 3d) show a cohesive-type fracture through the matrix, with no microspheres debonded. The amine groups in the silane of these two samples would react with carboxyl groups of MAPP [36, 37] creating amide bonds that contribute to a very resistant interface.

PP/co-PET/glass microspheres composites

This group of composites displayed a very different tensile behaviour depending if MAPP was added or not to the matrix formulation. When MAPP was not added (C composites) low tensile strength values resulted (Fig. 4a).

It would be expected that glass beads appear debonded from matrix in the fractographies. In fact, they appear in this way (Fig. 5a), although the microspheres surface is covered by a thin layer of polymer. To elucidate the nature of this polymeric layer, two different experiences were carried out on C1 sample. First, fracture surface of this composite was immersed for 24 h in trifluoroacetic acid to extract the co-PET.

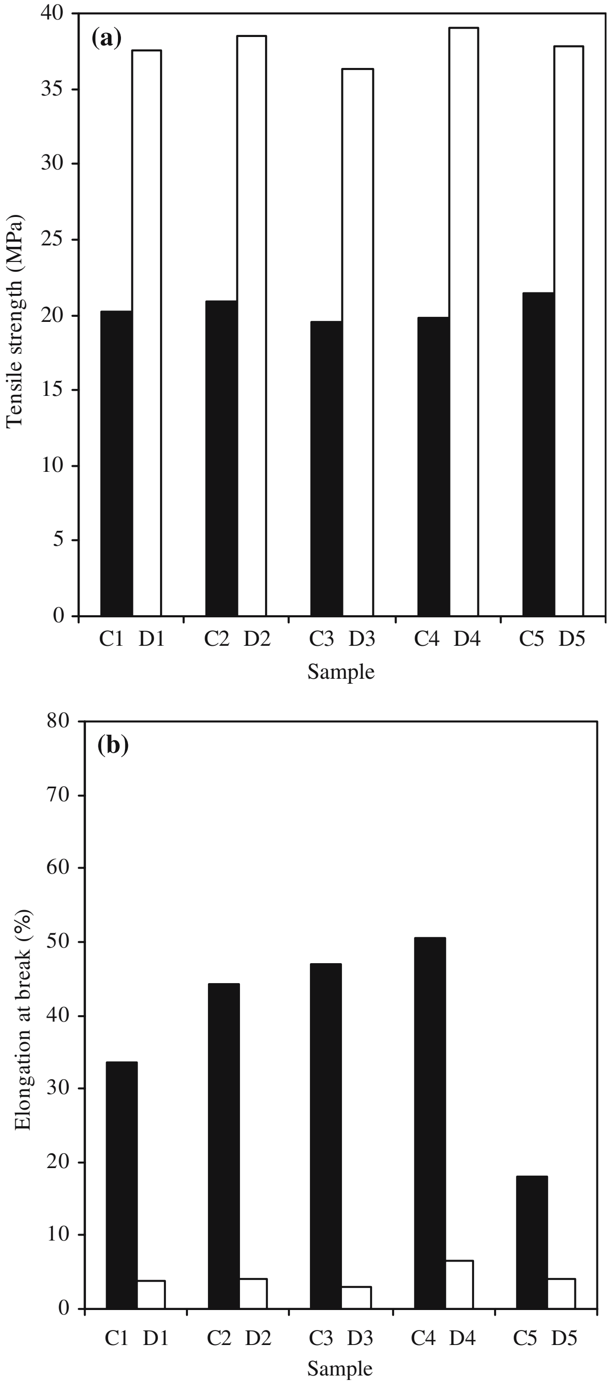

Fig. 4 Tensile strength (**a**) and elongation at break (**b**) of PP/co-PET (■) and PP/MAPP/co-PET (□) matrix-based composites

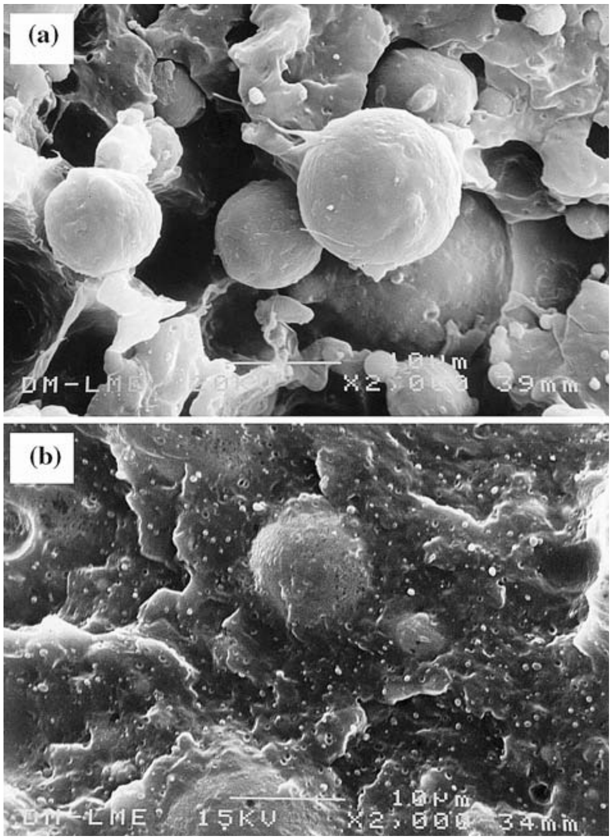

Fig. 5 Fracture surfaces of (**a**) C1 and (**b**) D1 composites

The subsequent observation by SEM showed smooth glass bead surface (Fig. 6), indicating that the polymer encapsulating the glass bead surface was extracted. Second, Raman spectra were taken from the composite fracture surfaces. The laser beam was focused on the matrix and on the microspheres to obtain information about the polymer composition in both zones. When the laser beam fixed on the glass bead surface, the laser intensity was reduced as low as possible with the aim of recording only the signal of the polymeric layer. The relative intensity of the characteristic Raman vibration band at 1,613 cm^{-1}, assigned to the stretching of C=O bond, in the microspheres surface was much higher that the one recorded from the whole matrix (Fig. 7), confirming the tendency pointed before of co-PET to

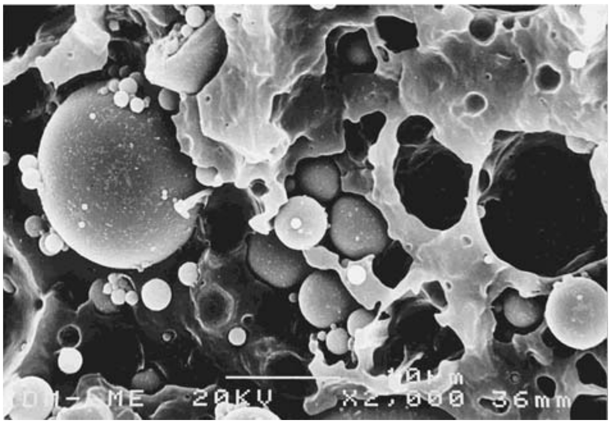

Fig. 6 Fracture surface of composite C1 after extracting co-PET phase with trifluoroacetic acid

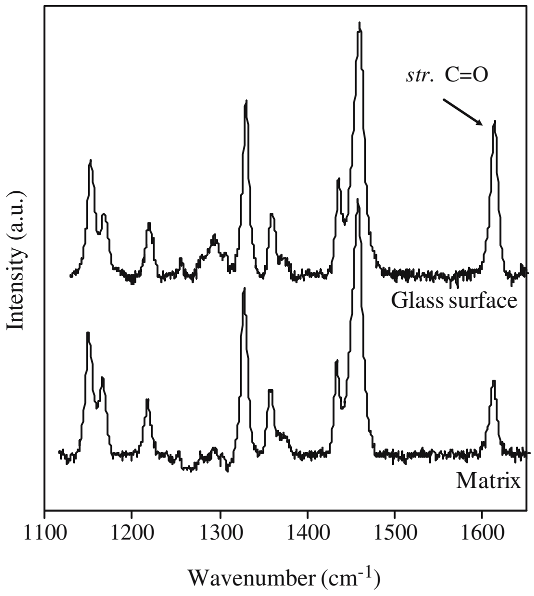

Fig. 7 Raman spectra of C1, obtained pointing the laser in the surface of the sphere (*upper spectrum*) and in the matrix (*inferior spectrum*), respectively

encapsulate glass beads in these composites. Possible interactions between hydroxyl and carboxyl co-PET groups either with hydroxyl groups of untreated glass surface and with functional groups of silanes should be investigated to help to elucidate the co-PET tendency to embed glass particles.

These composites failed in a ductile manner with lower values of both tensile strength and elongation at break (Fig. 4b) than those of composites of series A. Two combined effects would explain these low mechanical properties. First, the co-PET tendency to encapsulate glass beads would result in increased glass bead apparent volume, and thus in reduced matrix effective cross-section. Second, the poor compatibility between co-PET and PP would lead to an easy debonding of these two polymers [15, 37, 38].

All the composites of the D series satisfied high adhesion level. They displayed the highest tensile strength values (Fig. 4a). In these composites there is a strong interface between matrix and microspheres, because of the compatibilizing effect of MAPP for co-PET and PP [16, 20, 37]. As it can be seen in Fig. 5b the glass beads are hidden, fully covered by the matrix. In this case, similarly to composites of the C series, glass beads are encapsulated by co-PET. However, the difference between C and D composites is that the co-PET encapsulating the glass beads is now compatibilized, strongly bonded to PP, while in composites of the C series it was not bonded to PP (Fig. 5a). Furthermore, as it can be seen in Fig. 5b, the excess of PET that is not encapsulating the glass microspheres appear very well dispersed within the PP matrix in the form of very small droplets. A further study focusing on the concentration of co-PET and MAPP would be necessary to optimize the interface in these composites.

Effect of the strain rate

The effect of the strain rate on the composite tensile properties was studied, and it was found that Young's modulus, tensile strength and elongation at break values followed logarithmic functions with the strain rate ($\dot{\varepsilon}$), as follows:

$$M = A \log \dot{\varepsilon} + B \tag{1}$$

In the above equation, M refers to the tensile property and A is a parameter indicative of the variation sensitivity or time dependency of the composite. The values of A for all the composites have been summarized in Table 3 and the fit plots are shown in Fig. 8.

As expected, both Young's modulus and tensile strength increased with the strain rate, whereas the elongation at break diminished. This is the general behaviour of the thermoplastic matrix-based composites due to the viscoelasticity [39]. Nevertheless,

Table 3 Strain rate sensitivity parameter (A in Eq. 1) of the indicated mechanical properties

Sample	A (M = Young's modulus)	A (M = Tensile strength)	A (M = Elongation at break)
PP	0.21	3.80	–
A1	0.42	1.53	–
A2	0.40	1.79	–
A3	0.43	1.79	–
A4	0.45	1.48	–
A5	0.40	1.51	–0.53
PP/MAPP	–	–	–
B1	0.50	3.81	–1.61
B2	0.50	2.19	–1.49
B3	0.46	2.24	–1.79
B4	0.46	2.63	–0.52
B5	0.46	2.80	–0.51
PP/co-PET	0.23	3.40	–
C1	0.78	1.66	–
C2	0.75	1.76	–
C3	0.73	1.51	–
C4	0.77	1.64	–
C5	0.77	1.80	–
PP/MAPP/co-PET	0.23	3.13	–
D1	0.97	3.36	–0.10
D2	0.98	3.47	–0.22
D3	0.97	3.16	–0.04
D4	0.98	3.21	–0.19
D5	0.98	3.12	–0.18

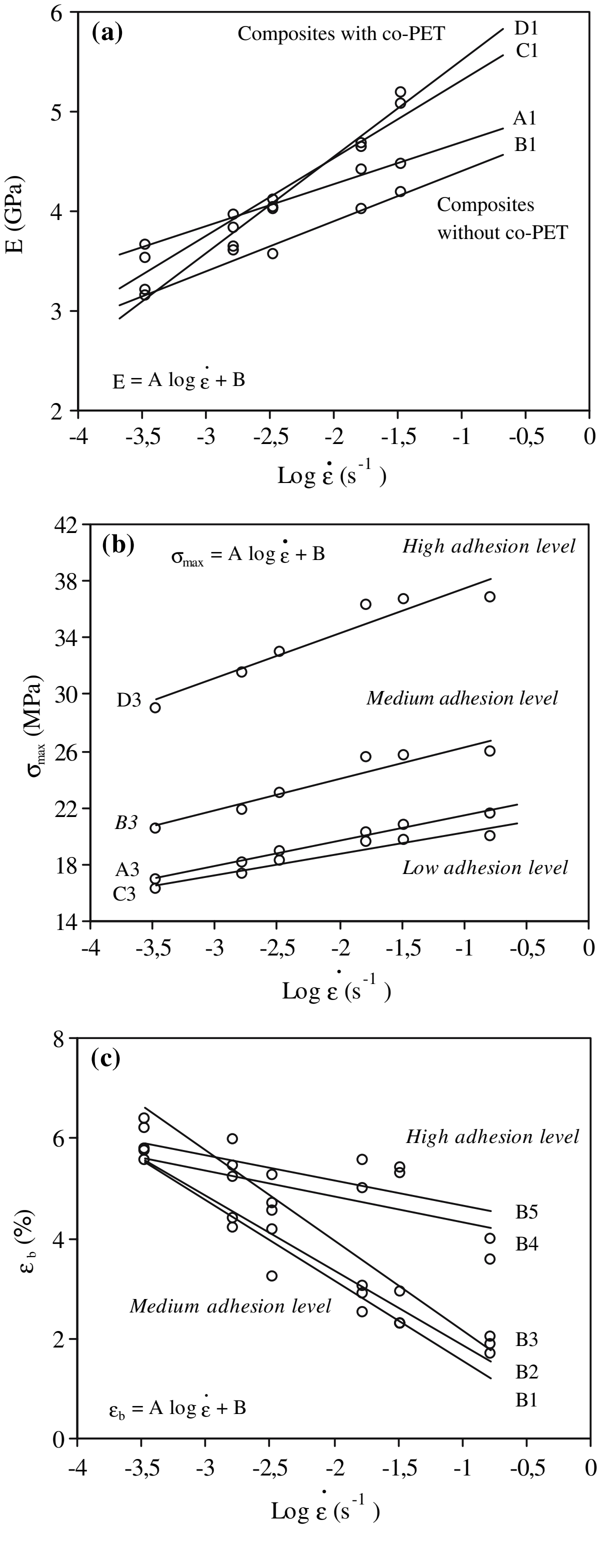

Fig. 8 Dependency functions of Young's modulus (**a**), tensile strength (**b**) and elongation at break (**c**) with respect to the deformation rate

remarkable differences could be appreciated in the sensitivity parameter (A) of the composites studied in this paper; as follows:

By one hand, when co-PET and/or MAPP was added in the polymer matrix, significant differences of strain rate sensitivity on the Young's modulus resulted, which should be related with changes of the interfaces elastic properties, as their addition leads to dramatic changes in the adhesion level between phases, and even in the extent or thickness of the interface. It was observed that the incorporation of 5 wt.% of co-PET into PP slightly modified the sensitivity parameter (A passed from 0.21 to 0.23), whereas the addition of co-PET in glass bead-filled PP provoked a synergic effect (A increased from 0.42 to 0.78). This synergic effect was even improved with the addition of MAPP, resulting A values near to 1.

On the other hand, the strain rate sensitivity of the composite tensile strength seemed to be related with both the interfacial adhesion level and the material composition. It was appreciated that the addition of glass beads into PP resulted in a marked reduction of the parameter A, whereas a higher adhesion level resulted in values of A close to that of unfilled PP. The most alike values were found for composites of D series, having co-PET and MAPP.

Finally, it was noticed that the composite elongation at break with a medium adhesion level showed a higher dependence on the strain rate, whereas in composites with high interfacial adhesion the strain rate sensitivity was lower. This could be explained on a basis of more viscous dissipation phenomena, such as microspheres debonding and matrix plastic flow.

Conclusions

Different phase morphology, tensile behaviour and failure patterns have been shown in glass bead-filled PP composites by adding or not co-PET and/or MAPP, and also depending on the type of silane-coupling agent employed for the glass bead surface treatment. As a general rule, good interfacial adhesion made the composite tensile strength to be high, even slightly higher than that of unfilled PP. In the PP composites, this was achieved when both MAPP and silane-coupling agents Z-6020 or Z-6032 were used. The addition of MAPP in combination with co-PET leaded to high interfacial adhesion independently on the surface treatment applied on the glass surface. The compatibilization of the blend co-PET/PP by adding MAPP guarantee a strong interface between them. In addition, a high affinity of co-PET towards the glass bead surface (untreated or silane-treated), higher than that of the pure PP, was shown. This higher affinity promoted encapsulation of the glass microspheres by co-PET during the melt processing. The adhesion at glass/co-PET interface resulted strong independently of the silane nature and even without silane treatment, which seems to be evidence that co-PET chemically bond to the glass surface through surfacial hydroxyl groups.

Higher the interfacial adhesion level lower the extent of plasticity developed in the composites. Both the stiffness and the tensile strength rose with the strain rate, while the elongation at break diminished. The modification of the interfacial adhesion significantly modified the sensitivity of the mechanical properties with the strain rate variation.

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