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In situ compatibilization of polypropylene and poly(butylene terephthalate) polymer blends by one-step reactive extrusion

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This paper concerns the *in situ* compatibilization of immiscible polypropylene/poly(butylene terephthalate) (PP/PBT) blends by a one-step reactive extrusion process. By *one-step reactive extrusion*, we are referring to functionalization of the PP with a monomer bearing a desired functional group and the subsequent interfacial reaction of this functionalized PP with the PBT, where the whole operation is carried out in *a single extrusion process.* Three monomers, acrylic acid (AA), maleic anhydride (MA) and glycidyl methacrylate (GMA), which are potentially reactive towards the carboxylic and/or hydroxyl groups at the chain ends of the PBT, were melt grafted on to the PP by free-radical reactions. A comparative study showed that GMA is more effective in compatibilizing the PP/PBT blends. Optimization of chemical parameters (initial concentrations of GMA and peroxide) and processing conditions (the sequence of addition of the materials, devolatilization of residual GMA, etc.) allowed us to obtain compatibilized PP/PBT blends with a fifteen- to twentyfold improvement in elongation at break and impact strength over an uncompatibilized PP/PBT blend. The *in situ* compatibilization performance of this one-step reactive extrusion process was comparable with that of a classical two-step process. In this latter case, the first extrusion step was to functionalize the PP with GMA, and the second one being the interfacial reaction between the functionalized PP and the PBT. Copyright © 1996 Elsevier Science Ltd.

(Keywords: polymer blends; reactive extrusion; compatibilization)

INTRODUCTION

The mechanical properties of a polymer blend are directly associated with its morphology and the interfacial adhesion between the various phases. A desired and stabilized morphology of an immiscible polymer blend with sufficiently strong interfacial adhesion often requires the presence of an appropriate interfacial agent (compatibilizer). Block or graft copolymers are well known compatibilizers which locate themselves preferentially at the interface. As a result, they tend to reduce the interfacial tension, strengthen the interfacial adhesion and prevent coalescence[1]. However, if a particular compatibilizer had to be synthesized separately for every pair of immiscible polymers, the use of the term 'polymer blend' would be nothing more than academic. This is due primarily to the difficulties involved in designing and synthesizing these particular molecules on an industrial scale.

Recently increasing efforts have been directed towards *in situ* compatibilization of immiscible polymer blends by reactive extrusion[2,3]: instead of synthesizing the compatibilizers in a separate step, they are created during extrusion through interfacial reactions between selected functional polymers. However, not all polymers contain functional groups. When a polymer bearing functional groups is to be mixed with another polymer which is chemically inert with respect to these functional groups, this latter material needs to be functionalized. Free-radical grafting of a vinyl monomer bearing the desired functional groups is the usual method employed for functionalization. In this case, two reactive blending processes are conceivable: (a) the chemically inert polymer is functionalized as a separate extrusion step, and is then blended with the functional polymer in a second extrusion step; (b) both the functionalization and the reactive blending steps are executed in the same extrusion process. For example, functionalization can be carried out in the first section of the extruder, followed by subsequent interfacial reaction between the functional and functionalized polymers. Process (a) is called *in situ* compatibilization of immiscible polymers by *two-step reactive extrusion*, with process (b) being called *one-step reactive extrusion.* To some extent, it can be said that a one-step reactive extrusion process is easier to control from a technological point of view, but is less profitable economically due to the additional passage of materials through the extruder.

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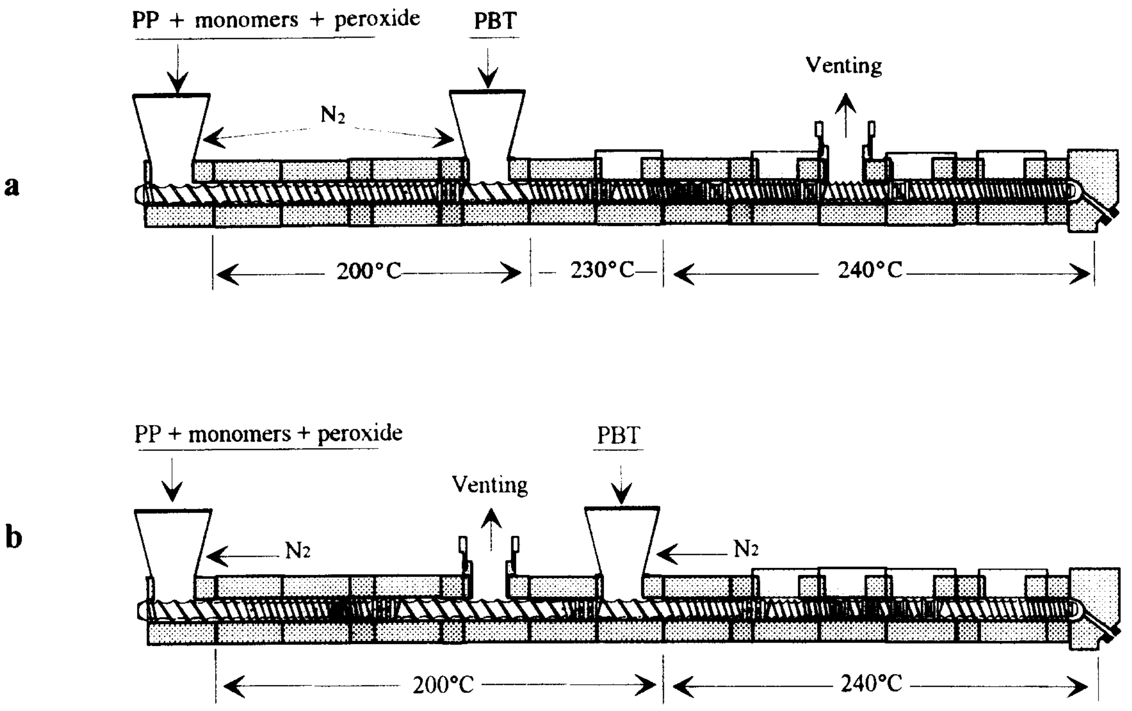

Figure 1 The two screw configurations used for the *in situ* compatibilization of PP/PBT blends by one-step reactive extrusion

The objective of this study was twofold: the *in situ* compatibilization of PP/PBT blends by one-step reactive extrusion, and the comparison of the *in situ* compatibilization performance of one-step reactive extrusion with that of two-step reactive extrusion. From the chemistry viewpoint, maleic anhydride (MA), acrylic acid (AA) and glycidyl methacrylate (GMA) are potentially reactive towards the carboxylic and/or hydroxyl groups of the chain ends of the PBT[3-7]. From an engineering viewpoint, the performance of *in situ* compatibilization by one-step reactive extrusion depends on the ability to control the chemical parameters (initial GMA and peroxide concentrations) and the processing conditions (the sequence of addition of the materials, devolatilization of residual monomers, screw speed, feed rate, etc.) in favour of the functionalization of the PP and the interfacial compatibilization reaction between the functionalized PP and the PBT.

EXPERIMENTAL

Materials

The polypropylene (PP) used in this study was a commercial product of Himont, i.e. Valtec CL101D. This was in the form of porous pellets which were capable of absorbing larger amounts of liquid monomers and peroxides than normal PP pellets. Another commercial polypropylene (Statoil, P401H) was also used in some of the two-step reactive extrusion runs. This was in the form of normal pellets. The poly(butylene terephthalate) (PBT) used was a commercial product of DSM, i.e. Arnite T08-200. This PBT contained at its chain ends 0.032 eq kg^{-1} of hydroxyl groups and 0.040 eq kg^{-1} of carboxylic groups. Selected characteristics of these polymers are given in *Table 1*.

Table 1 Characteristics of the PP and PBT polymers used in this study

Polymer	$\bar{M}_w$ (g mol^{-1})	$\bar{M}_n$ (g mol^{-1})	*MFI*[a]	η_0^b (Pa s)	m.p. (°C)
PP (Valtec CL101D)	654 000	141 000	0.30	1.0×10^5	165
PP (Statoil P401H)	484 000	88 000	0.35	6.5×10^4	165
PBT (Arnite T08-200)	65 000	28 000	–	1.0×10^3	223

[a] Melt flex index (in units of g per 10 min); ASTM 1238L, 230°C and 2.16 kg
[b] Zero shear rate viscosity of polymer at 240°C

The functional monomers, maleic anhydride (MA, purity 99%), acrylic acid (AA, purity 99%) and glycidyl methacrylate (GMA, purity 97%), were purchased from Aldrich and used as received. Styrene (purity 99%) was used as a comonomer to assist the free-radical grafting of the functional monomers on to PP. This was also purchased from Aldrich and used without further purification. The initiator was 1,3-bis(tert-butylperoxyisopropyl)benzene, a commercial product from Akzo (Perk-14, purity 90%). Its half-life time was ca. 30 s at 200°C and 2 s at 240°C.

One-step reactive extrusion processes

A co-rotating self-wiping twin screw extruder of the type Werner & Pfleiderer ZSK-30 was used (screw diameter $d = 30$ mm; length-to-diameter ratio $l/d = 42$). Two different screw configurations were chosen to process the PP/PBT blends (*Figure 1*). In screw configuration (a), the PP pellets, the monomers (GMA and styrene) and the peroxide were fed into the first hopper while the PBT pellets were fed from a second hopper further downstream. In this way, the functionalization of the PP occurred almost exclusively in the first zone between the first and second hoppers, after which (from the second hopper to the die) the interfacial reaction started between the functionalized PP and the PBT[8].

These two zones were demarcated by a kneading block (KB90/5/28), together with a left-handed screw element (−20/10). These two special elements were used in order to promote a rapid and complete melting of the PP at that particular location. By considering the short half-life time of the peroxide, the barrel temperature in the first zone was set at 200°C. The temperature for the next barrel was raised to 230°C and then 240°C (melting point of the PBT, 223°C) in order to promote the interfacial reaction leading to the formation of the desired grafted copolymer, PP-*g*-PBT, as the compatibilizer. Six kneading blocks (KB90/5/28 and KB 45/5/20) were placed after the feeding point of the PBT in order to provide efficient mixing. A devolatilization zone was placed prior to the pumping zone, close to the die. The pressure in this devolatilization zone was ca. 25 mmHg, achieved by the use of a vacuum pump.

The extruder configuration (b) differed from (a) in that the devolatilization zone was shifted upstream so as to precede the PBT feeding point. Moreover, the devolatilization zone in the screw configuration (b) was longer than that in configuration (a). In this way, the unreacted monomers could be removed more efficiently, therefore reducing the undesired reaction between the unreacted GMA and the PBT.

Before being fed into the extruder, a liquid mixture of GMA/styrene/Perk-14 of a chosen proportion was absorbed by the porous PP pellets at room temperature for ca. 1 h, while the PBT pellets were dried overnight at 105°C under hot air circulation conditions. The mixture of PP/monomers/Perk-14 and the dried PBT pellets were fed to the extruder separately through two screw feeders (K-Tron K10S). In this present study, the total feeding rate of the two polymers was always 5.0 kg h^{-1}, and the screw rotating speed was 150 rpm. The extrusion processes were carried out under a constant nitrogen purge. The extrudates were cooled in water and then pelletized for further characterization.

Two-step reactive extrusion process

The first extrusion step was to graft GMA on to the PP (Valtec CL101D), while the second step was to use the GMA-grafted PP thus obtained as a compatibilizer precursor for the PP/PBT blends. A virgin PP, GMA-grafted PP and PBT pellets were mixed before they were fed to the extruder through the same hopper. The screw configuration for the second extrusion step is the same as screw configuration (a), except that the barrel temperatures were kept at 240°C throughout.

Determination of the GMA grafting yield

The GMA grafting yield on to the PP was measured by a Fourier transform infra-red spectrometer using a pre-established calibration curve. The experimental details have been presented elsewhere[9].

Characterization of PP/PBT blends

The performance of the *in situ* compatibilization was evaluated by the mechanical properties in terms of the elongation at break (by tensile testing, ASTM 638-71A) and the impact strength (falling weight, ISO 6603/2) of the resulting PP/PBT blends. The experimental details can be found elsewhere[3]. The morphology of some of the PP/PBT blends was also examined by scanning electron microscopy (SEM) on the freshly fractured and gold-sputtered surfaces of the specimens to be used for the tensile testing. The middle part of the cross-section of the specimens was chosen as being representative of the overall morphology.

RESULTS AND DISCUSSION

Previous studies[8,9] showed that the presence of styrene not only improves the GMA grafting yield but also reduces the PP chain degradation. At a given initial GMA concentration, the GMA grafting yield increases with increasing $[\text{styrene}]_i/[\text{GMA}]_i$. In order to graft enough GMA on to the PP, the $[\text{styrene}]_i/[\text{GMA}]_i$ ratio was fixed at 1.40 mol mol^{-1}, with the same styrene/monomer ratio being retained for the grafting of AA and MA.

Effect of the nature of functional monomers

In order to compare the effectiveness of these three functional monomers, i.e. MA, AA and GMA, in the *in situ* compatibilization of the PP/PBT blend, their initial molar concentrations were identical, being 0.01 mol per 100 g of PP or 0.01 mol per 100 parts of resin (mhr). The initial peroxide concentration was 0.30 g per 100 g of PP or 0.3 parts per 100 parts of resin (phr). The weight proportion of PP/PBT was 70/30, and screw configuration (a) was used (*Figure 1*). The grafting yields of these functional monomers are shown in *Table 2*, as well as the mechanical properties of the resulting PP/PBT blends. There is clearly, a great increase in the elongation at break and impact strength for the GMA compatibilized PP/PBT blend compared to the mechanical blend. The MA and AA compatibilized PP/PBT blends, on the other hand, do not show any improvement in mechanical

Table 2 Comparison of the effectiveness of MA, AA and GMA as compatibilizer precursors for the PP/PBT (70/30) blend obtained by one-step reactive extrusion

System	$[M]_g^a$ (mhr)	E^b (MPa)	σ_y^c (MPa)	ε_b^d (%)	Impact strength at 0°C (*J*)
PP	–	1190 ± 30	27.2 ± 0.7	146 ± 15	24 ± 1.7
PBT	–	2570 ± 60	52.8 ± 0.4	>260	20 ± 1.0
PP/PBT, mechanical mixture	–	1450 ± 70	29.6 ± 0.6	29 ± 5	2 ± 0.4
PP/PBT, reactive blending with MA	0.006	1400 ± 30	27.6 ± 0.2	28 ± 5	2 ± 0.2
PP/PBT, reactive blending with AA	0.006	1440 ± 20	27.9 ± 0.2	32 ± 5	1.5 ± 0.6
PP/PBT, reactive blending with GMA	0.003	1320 ± 60	25.1 ± 0.2	110 ± 16	19 ± 3.0

[a] Grafting yield of a functional monomer on to PP
[b] Young's modulus
[c] Yield strength
[d] Elongation at break

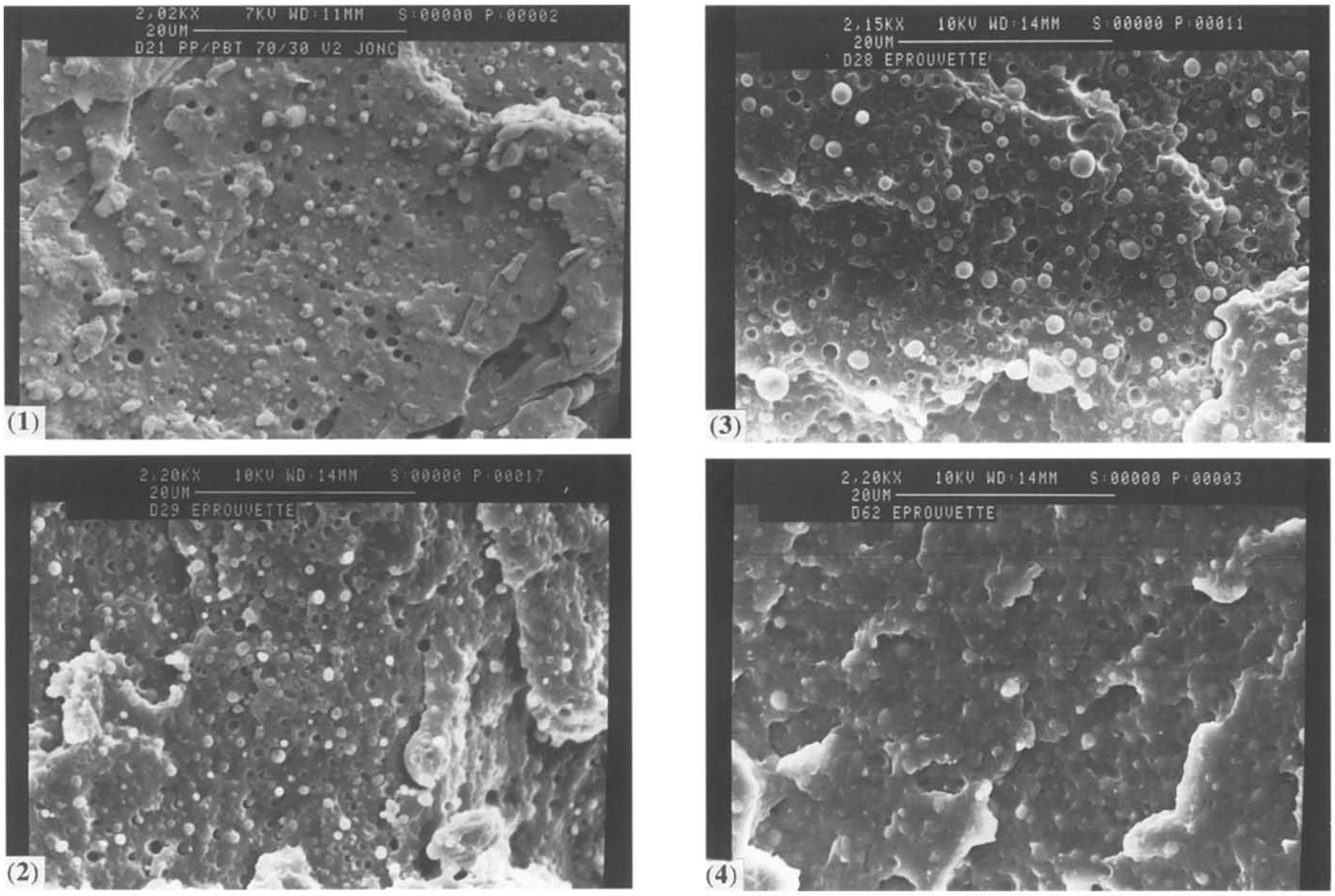

Figure 2 Scanning electron micrographs of the PP/PBT (70/30) blends compatibilized with various functional monomers: (1) mechanical mixture; (2) blend compatibilized with MA/styrene; (3) blend compatibilized with AA/styrene; (4) blend compatibilized with GMA/styrene

properties, despite the greater grafting yields (0.006 mhr) compared to that of GMA (0.003 mhr).

In fact, this disparity in compatibilization effectiveness among these three functional monomers is in line with the morphology observations. As shown in *Figure 2*, the morphology of the MA or AA compatibilized PP/PBT blends is basically the same as that of the mechanical blend, at least as far as the size of the dispersed phase and the interfacial adhesion are concerned. The particle size of the GMA compatibilized PP/PBT blend is somewhat smaller and the interfacial adhesion appears to be stronger.

The compatibilization performance difference between these three functional monomers lies in their functional reactivity towards the carboxylic and/or hydroxyl groups at the chain ends of the PBT. The MA-grafted PP is expected to react with the hydroxyl group of the PBT to form the desired compatibilizer PP-*g*-PBT. This reaction is reversible and its equilibrium is highly shifted to the reactant side with increasing temperature[10]. A temperature as high as 240°C certainly corresponds to an equilibrium which is unfavourable for the formation of the PP-*g*-PBT. The GMA-grafted PP reacts primarily with the carboxylic group of the PBT. This reaction is also reversible, but its equilibrium is shifted to the product side with increasing temperature[11]. In the case of AA, the main reaction should be the esterification of the AA-grafted PP with the hydroxyl group of the PBT. This reaction is know to be much slower kinetically than the other two reactions.

Optimization of the initial GMA concentration

The GMA grafting yield is far below 100% with respect to the initial GMA concentration. In other words, there is always unreacted or residual GMA left after the free-radical grafting process. An increase in the initial GMA concentration increases not only the amount of the GMA grafted on to the PP but also that of the residual GMA[3,8,9]. The former favours the formation of the desired graft copolymer PP-*g*-PBT, whereas the latter achieves the opposite effect. This is because the residual GMA monomer can also react with the PBT, probably at a higher rate than the GMA-grafted PP, due to its much greater mobility, therefore reducing the formation of the PP-*g*-PBT copolymer. The competition between the GMA-grafted PP and the residual GMA monomer in terms of their reactivities towards PBT implies the existence of an optimum initial GMA concentration for achieving the highest possible compatibilization performance of PP/PBT blends under the given processing conditions.

In order to verify this point, various PP/PBT (70/30) blends were processed using screw configuration (a) by varying the initial GMA concentration $[GMA]_i$, from 0 to 7.0 phr. The initial peroxide concentration $[Perk\text{-}14]_i$ was 0.30 phr in all cases. *Figure 3* shows the elongation at break and impact strength as a function of $[GMA]_i$. Also plotted in this figure is the GMA grafting yield as a function of $[GMA]_i$. Indeed, the elongation at break and

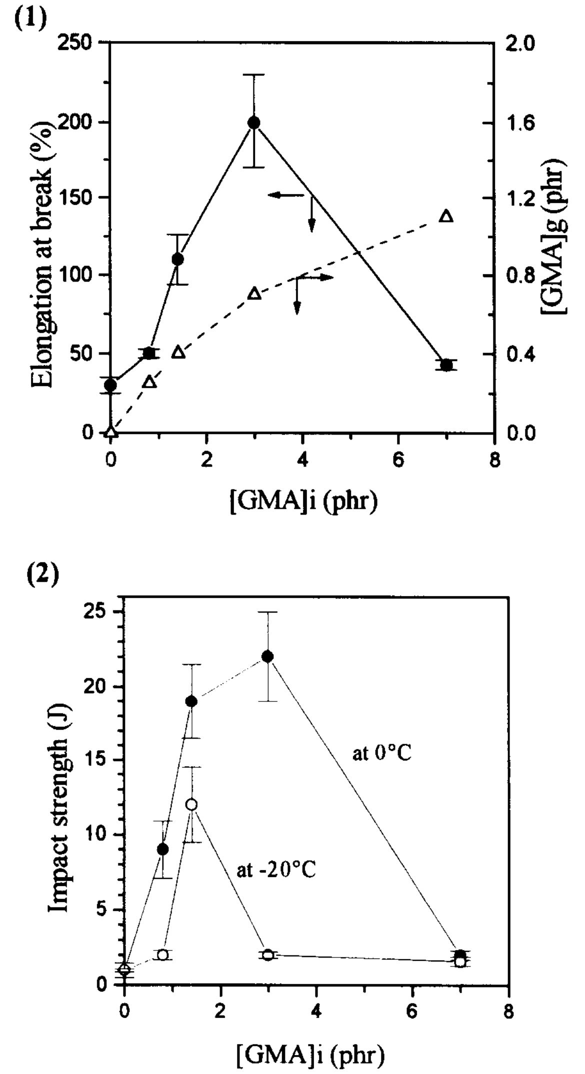

Figure 3 Effect of the initial GMA concentration on the elongation at break (1) and impact strength (2) of the compatibilized PP/PBT blend using screw configuration (a)

impact strength of the PP/PBT blend increase with increasing $[GMA]_i$, reach a maximum when $[GMA]_i$ is equal to 3.0 phr, and then decrease with further increases in $[GMA]_i$. In addition, when $[GMA]_i$ is increased from 0.8 to 7.0 phr, the GMA grafting yield prior to the feeding point of the PBT, $[GMA]_g$, is increased from ca. 0.3 to 1.1 phr. Correspondingly, the residual GMA monomer concentration, $[GMA]_r$, is increased disproportionally from 0.5 to 5.9 phr.

Optimization of devolatilization of the residual GMA

It should be noted that when 1.4 phr GMA is added, the concentration of the residual monomer, $[GMA]_r$, is four times the molar concentration of the carboxylic group, [–COOH], at the chain end of the PBT. Moreover, it is found to be as high as seventeen times that of [–COOH] when $[GMA]_i$ is 7.0 phr. This clearly shows the need to remove the residual GMA effectively from the system before the PBT is fed in order to promote the desired interfacial reaction between PP-*g*-GMA and PBT–COOH[3]. Thus, we have used screw configuration (b), rather than configuration (a).

The performance of screw configuration (b) was compared with that of configuration (a) in terms of the impact strength of the PP/PBT (70/30) blends compatibilized with 3.0 and 7.0 phr GMA, respectively. With 3.0 phr GMA, the impact strength of the blend at 0°C is 22 and 27 J for screw configurations (a) and (b), respectively. When the testing temperature is −20°C, the impact strength is only about 2 J with screw configuration (a), whereas it is as high as 19 J with configuration (b) (*Figure 4*). The greater impact strength with screw configuration (b) is undoubtedly due to the more efficient and earlier removal of the residual GMA monomer. Actually, this is also supported by the experimental fact that the grafted GMA concentrations prior to the feeding point of the PBT are very similar (0.6–0.7 phr) with both screw configurations, whereas $[GMA]_r$ is reduced by a factor of two when using screw configuration (b).

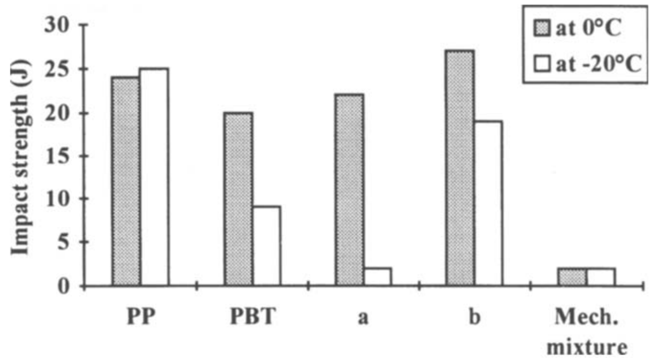

Figure 4 Effect of the devolatilizing residual GMA on the impact strength of the PP/PBT blends: (a) blend made by screw configuration (a) with a devolatilization zone positioned close to the die; (b) blend made by screw configuration (b) with a devolatilization zone located upstream just after the GMA grafting zone ($[GMA]_i = 3.0$ phr; PP/PBT = 70/30)

The impact strength of the blends obtained with 7.0 phr GMA is weak (~ 2 J) for both screw configurations. The difference, in terms of impact strength, between 3.0 and 7.0 phr GMA implies that although $[GMA]_g$ is somewhat higher with 7.0 phr GMA, this gain in $[GMA]_g$ does not compensate for the negative effect of the concomitant increase in $[GMA]_r$. This is true for both screw configurations, indicating that a high compatibilization performance can be achieved only when the competition between $[GMA]_g$ and $[GMA]_r$ is so balanced that formation of the compatibilizer PP-*g*-PBT is the greatest. Thus, an increase in $[GMA]_i$ should be accompanied by an enhanced devolatilization capability so as to satisfy this balance.

Effect of the peroxide concentration

Previous studies[8,9] showed that when the initial peroxide concentration increases, the GMA grafting yield increases while the weight-average molecular weight of the GMA-grafted PP, $\bar{M}_w$, decreases. An increase in $[GMA]_g$ favours the formation of the PP-*g*-PBT copolymer, therefore improving the elongation at break and impact strength of the PP/PBT blend, whereas a decrease in $\bar{M}_w$ of the PP leads to a deterioration in these properties. Thus the effect of varying the peroxide concentration may go both ways.

A PP/PBT (70/30) mixture was processed with screw configurations (a) and (b), respectively. In the case of screw configuration (a), a relatively low initial GMA concentration (1.4 phr) was added in order to reduce the effect of the residual GMA monomer. The initial perox-

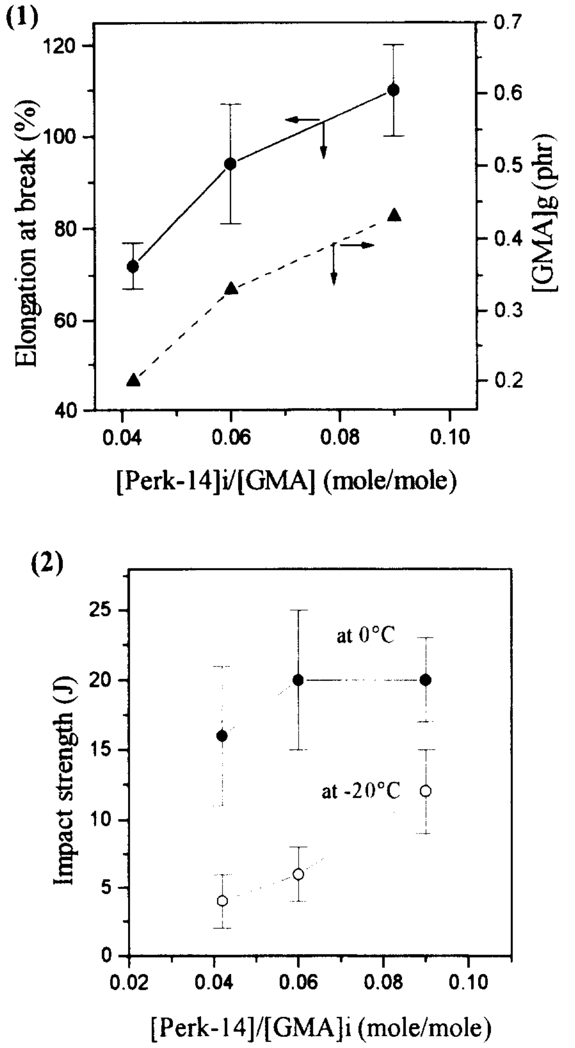

Figure 5 Effect of the initial peroxide concentration on the elongation at break (1) and impact strength (2) of compatibilized PP/PBT (70/30) blends using screw configuration (a) ($[GMA]_i = 1.4$ phr)

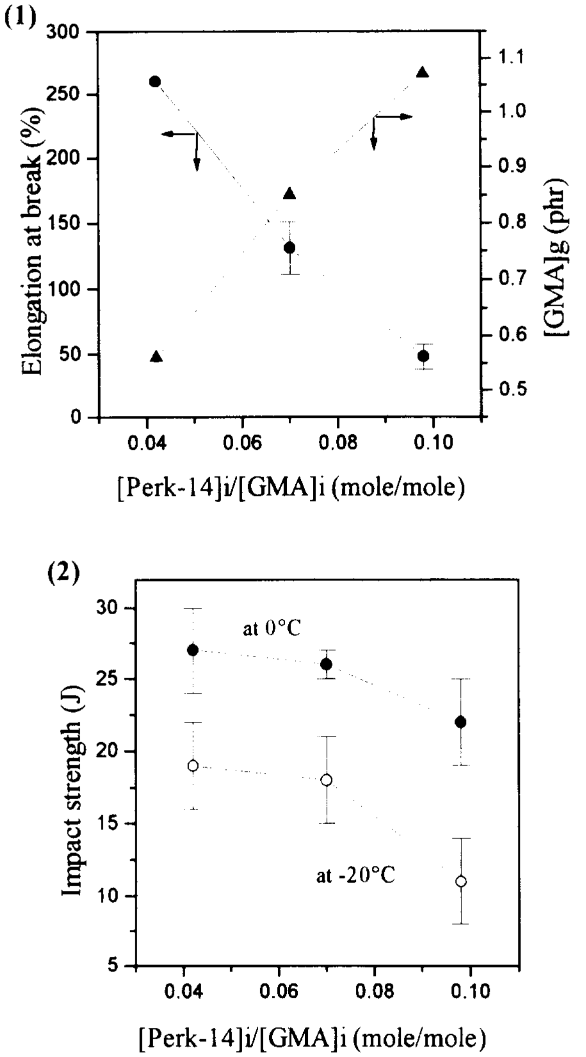

Figure 6 Effect of the initial peroxide concentration on the elongation at break (1) and impact strength (2) of compatibilized PP/PBT (70/30) blends using screw configuration (b) ($[GMA]_i = 3.0$ phr)

ide concentration $[\text{Perk-14}]_i$ ranged from 0.14 to 0.30 phr. In the case of screw configuration (b), which has a greater ability to remove the residual GMA, the initial GMA concentration was fixed at 3.0 phr. The initial peroxide concentration, $[\text{Perk-14}]_i$, varied from 0.30 to 0.70 phr.

Figure 5 shows plots of the GMA grafting yield on to PP, i.e. the elongation at break and the impact strength of the corresponding PP/PBT blend, as a function of $[\text{Perk-14}]_i/[\text{GMA}]_i$ for screw configuration (a). An increase in $[\text{Perk-14}]_i$ tends to increase $[\text{GMA}]_g$ and improves the mechanical properties of the PP/PBT blends in terms of the elongation at break and impact strength. In contrast, in the case of screw configuration (b), while an increase in $[\text{Perk-14}]_i$ also increases $[\text{GMA}]_g$, the elongation at break and impact strength of the resulting blend both decrease (*Figure 6*). These opposing trends, with respect to the mechanical properties of the blend for these two screw configurations, suggest that in the case of screw configuration (a), when increasing $[\text{Perk-14}]_i$, the positive effect of increasing $[\text{GMA}]_g$ dominates the negative effect of decreasing the grafted PP molecular weight; in the case of screw configuration (b), due to the greater GMA grafting yields, the positive effect of further increasing $[\text{GMA}]_g$ does not compensate for the negative effect of decreasing the grafted PP molecular weight.

Effect of the PBT fraction

Three PP/PBT blends containing, respectively, 30, 50 and 70 wt% PBT were prepared using screw configuration (a). The initial concentrations of GMA, styrene and Perk-14 were 3.0, 2.0 and 0.30 phr, respectively. Under these conditions, the GMA grafting yield prior to the PBT feeding point was found to be 0.7 phr and the number-average molecular weight of the grafted PP was ca. 106 000 g mol^{-1}. This means that on the average, there were about five GMA moieties per PP chain.

The elongation at break and the impact strength at 0°C of the compatibilized PP/PBT blend are plotted in *Figure 7* as a function of the PBT fraction. The results obtained for the compatibilized PP/PBT (70/30) blend using screw configuration (b) (the removal of the residual GMA is improved in this case) are also shown for comparison purposes. As expected, the compatibilized PP/PBT blends display much better mechanical properties (elongation at break and impact strength) than the uncompatibilized ones, regardless of the PBT fraction. In the case of the PP/PBT (70/30) blend, the improvement in these mechanical properties is even greater with screw configuration (b) than with configuration (a) due to a much more efficient removal of the residual GMA in the

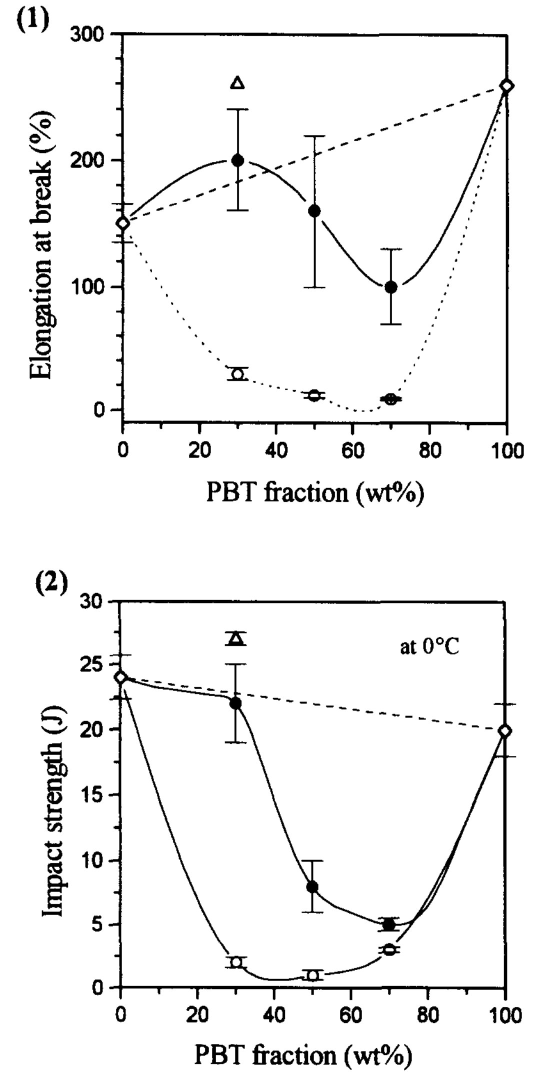

Figure 7 Elongation at break (1) and impact strength at 0°C (2) of the compatibilized PP/PBT blend as a function of the PBT weight fraction: (-◇-) additivity line of the pure PP and PBT; (○) mechanical mixtures; (●) compatibilized blends using screw configuration (a); (△) compatibilized blends using screw configuration (b)

former screw configuration. Also noteworthy is that an increase in the PBT fraction tends to worsen these mechanical properties of the PP/PBT blend. This may be attributed to possible crosslinking of the blend when the PBT fraction becomes more significant.

In fact, the degree of crosslinking of the three blends obtained by using screw configuration (a) was examined by dissolving them in boiling 1,1,2,2-tetrachloroethane, in which both the pure PP and PBT are soluble. These blend samples weighed 3% with respect to the solvent. It turned out that after ~2 h of heating the blends containing 30 and 50 wt% PBT were completely dissolved, forming transparent solutions. However, the blend with 70% PBT was only partly dissolved, with particles being suspended in the system. This was the case, even after 8 h of treatment in this way, thus suggesting that this blend was slightly crosslinked.

We should bear in mind that the concentration of the carboxylic group is 0.040 eq kg^{-1} and that of the hydroxyl group is 0.032 eq kg^{-1} for the PBT used in this study. This means that roughly speaking, 90% of the PBT chains contain one carboxylic group at one extreme of the chains and one hydroxyl group at the other extreme. The other 10% have carboxylic groups at both extremes. It is known that both carboxylic and hydroxyl groups can react with an epoxy group, with the former being more reactive (by a factor of ten to twenty). If both terminal groups of the PBT chains have reacted with the GMA-grafted PP chains, then crosslinking has occurred. Obviously, the degree of crosslinking is expected to increase with an increasing PBT fraction in the PP/PBT blend.

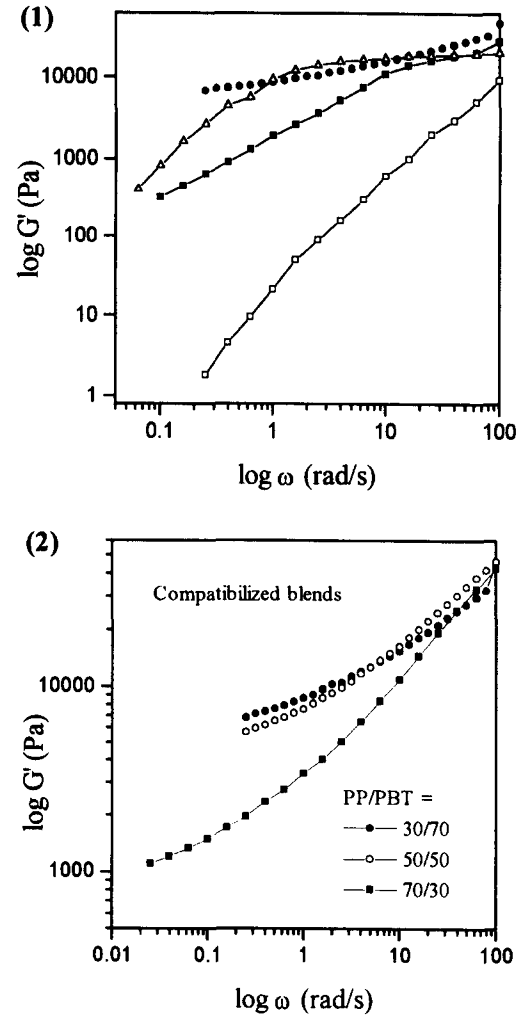

Figure 8 shear modulus (log G') of various PP/PBT blends as a function of deformation frequency (log ω). (1) (-△-) PP; (-□-) PBT; (-■-) mechanical PP/PBT (30/70) blend; (●) compatibilized PP/PBT (30/70) blend. (2) Compatibilized PP/PBT blends with various PBT contents: (-■-) 30; (-○-) 50; (-●-) 70 wt%

Rheological behaviour of the compatibilized PP/PBT blends

The rheological properties of the compatibilized PP/PBT blends were examined at 240°C by using a mechanical spectrometric analyzer (type RMS 605, Rheometrics). As shown in *Figure 8*, the logarithm of the dynamic modulus G' of the PP/PBT (30/70) mechanical blend as a function of the logarithm of the deformation frequency ω is located in between those of the PP and PBT. The log G' of the compatibilized blend is consistently higher than that of the mechanical one, confirming that there is a molecular buildup resulting from the interfacial reaction between the GMA-functionalized PP and the PBT. *Figure 8* also shows plots of $\log G'$ as a function of $\log \omega$ for the three compatibilized PP/PBT blends containing 30, 50 and 70 wt% PBT, respectively. It is seen that the $\log G'$ values of the compatibilized PP/PBT blends increase with increasing PBT fraction. If there was no molecular-weight buildup caused by the interfacial reaction between the GMA-grafted PP and the PBT, the $\log G'$ values of these blends would have decreased with increasing PBT fraction because the $\log G'$ of the PBT itself is much smaller than that of the PP.

Comparison between one-step and two-step reactive extrusion processes

So far, we have shown that the one-step reactive extrusion process developed in this study has yielded PP/PBT blends with highly improved mechanical properties in terms of the elongation at break and impact strength. Below we examine whether a classical two-step reactive extrusion process would do a better job. In order to carry this out, the first extrusion step was to graft GMA ($[\mathrm{GMA}]_i = 3.0$ phr) on to PP in the presence of Perk-14 (0.3 phr); the second step was to compatibilized the PP/PBT blends by using the GMA-grafted PP thus obtained as a compatibilizer precursor. Three PP/PBT (70/30) blends were then prepared with three different types of PP (*Table 3*). For a better comparison between the two-step process and the one-step process, the first extrusion stage in the two-step process was optimized so that it allowed us to produce GMA-grafted PP samples with characteristics (e.g. GMA grafting yield and molecular weight of the modified PP) close to those of the GMA-grafted PP obtained by the one-step reactive extrusion process (*Table 4*). *Table 5* shows some selected mechanical properties of these three compatibilized PP/PBT (70/30) blends. Clearly, they all display highly improved mechanical properties in terms of the elongation at break and impact strength when compared to the mechanical blend. Moreover, when the PP is composed of the GMA-grafted PP alone, the said mechanical properties are worse than those in which half of the PP-*g*-GMA is replaced either by the raw PP (Valtec) or PP (P401H) resin. This can be explained by a decrease in the molecular weight of the PP (Valtec) when this latter material is used in the GMA grafting process.

Table 3 Compositions of the three PP/PBT (70/30) blends used in the two-step extrusion process (values in wt%)

Run	PBT	PP-*g*-GMA	PP (Valtec)	PP (P401H)
1	30	70	0	0
2	30	35	35	0
3	30	35	0	35

Table 4 Characteristics of the GMA-grafted PP (Valtec) obtained by the one-step and two-step reactive extrusion processes

Process	$[\mathrm{GMA}]_i$ (phr)	$[\mathrm{GMA}]_g$ (phr)	$[\mathrm{GMA}]_r$ (phr)	$\bar{M}_w$ ($\mathrm{g\,mol^{-1}}$)	M_n ($\mathrm{g\,mol^{-1}}$)
Two-step	3.0	0.6	1.0	309 000	53 500
One-step	3.0	0.6	1.1	317 000	87 300

Table 5 Mechanical properties of the three compatibilized PP/PBT (70/30) blends obtained by using the two-step reactive extrusion process

Run	E^a (MPa)	σ_y^b (MPa)	ε_b^c (%)	Impact strength (J)	
				0°C	−20°C
1	1580 ± 40	26.0 ± 0.2	170/ ± 40	16 ± 8	9 ± 8
2	1590 ± 60	27.0 ± 0.3	>260	22 ± 1	7 ± 6
3	1430 ± 50	27.6 ± 0.1	>260	25 ± 1	12 ± 7

[a] Young's modulus
[b] Yield strength
[c] Elongation at break

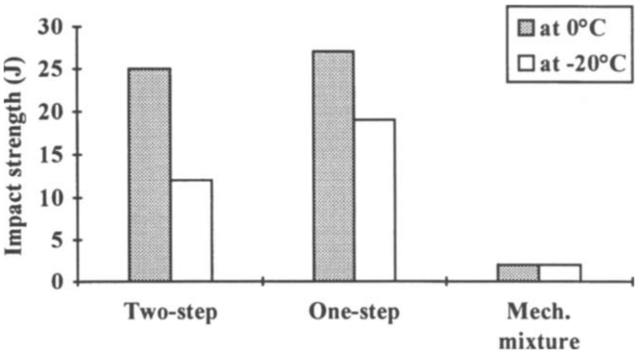

Figure 9 Comparison between the highest impact resistance values of the PP/PBT (70/30) blends obtained from the one- and two-step reactive extrusion processes

A comparison was also made of the highest impact strengths of the PP/PBT (70/30) blends obtained by the one-step and two-step reactive extrusion processes (*Figure 9*). Both processes have yielded a fifteen- to twentyfold improvement in impact strength with respect to the corresponding uncompatibilized blends. More importantly, the performance of the one-step reactive extrusion process is comparable to, if not better than, that of the two-step reactive extrusion process.

CONCLUSIONS

In this study, polypropylene/poly(butylene terephthalate) (PP/PBT) blends have been compatibilized by using a *one-step reactive extrusion process*. This process refers to the sequential execution of the grafting of the PP with a monomer bearing a desired functional group, and the subsequent interfacial reaction of this functionalized PP with the PBT through a single passage through the extruder. Among the three functional monomers, i.e. acrylic acid, maleic anhydride and glycidyl methacrylate (GMA), which are potentially reactive towards the carboxylic and/or hydroxyl groups at the chain ends of the PBT, GMA has yielded by far the greatest compatibilization performance. When optimized by taking into consideration the effects of various chemical parameters (initial concentrations of GMA and peroxide) and the processing conditions (sequence of addition of the materials, devolatilization of the residual GMA, etc.), this one-step reactive extrusion process has produced a compatibilized PP/PBT (70/30) blend with a fifteen- to twentyfold improvement in impact strength and elongation at break when compared with the uncompatibilized PP/PBT blend. A further increase in the PBT fraction tends to reduce the impact strength and elongation at break of the compatibilized PP/PBT blend, as a result of possible crosslinking which is associated with the reactivity of the second functional (hydroxyl and/or carboxylic) group located at one end of the PBT chains. Rheological characterization has confirmed a molecular-weight buildup due to the interfacial reaction between the GMA-grafted PP and the PBT.

A comparative study has shown that the mechanical properties (i.e. elongation at break and impact strength) of the PP/PBT blend obtained by the one-step reactive extrusion process are comparable to, if not better than, those of the blend obtained by a two-step process.

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