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IN SITU COMPATIBILIZATION OF PBT/PPO BLENDS

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Abstract—A reactive copolymer, styrene-glycidyl methacrylate (SG) has been used as an *in situ* compatibilizer for the polymer blends of PBT and PPO. The epoxy groups from the SG copolymer are able to react with PBT endgroups (—OH or —COOH) in melt to form an SG-g-PBT copolymer which can function as phase compatibilizer of the PBT/PPO blends. The optimized glycidyl methacrylate (GMA) content in SG copolymer and its optimized quantity in PBT/PPO blends to achieve maximum property improvement have been thoroughly investigated in this study. The presence of this *in situ* reactive compatibilizer improves the processability, impact strength, tensile properties, heat distortion temperature, and phase stability in thermal annealing of the incompatible blends of PBT/PPO.

INTRODUCTION

Poly(2,6-dimethyl-1,4-phenylene oxide) (PPO) is a material with high T_g, rigidity, good electric and chemical resistance, low water absorption, and good dimensional stability. Since PPO has high T_g, a higher processing temperature is necessary which leads to thermal degradation problems of the material. Miscible polymer pairs of PPO and polystyrene (PS) or other styrenic polymers have been intensively investigated [1–5]. The driving force for miscibility of PPO/PS blends has been determined as the van der Waals interactions between the aromatic rings of PPO and PS based on spectroscopic studies [2]. The miscibility of the blend PPO and styrene-maleic anhydride (SMA) copolymer was found depending on the MA content of the SMA [6].

Only a limited number of published papers on polymer blends of PPO and the polyester poly(-butylene terephthalate) (PBT) have been reported in open literature [7–9] possibly due to the incompatibility between these two classes of polymers. PBT is also classified as a major engineering thermoplastic with excellent properties in terms of electric insulation and solvent resistance. Yamamoto reported significant processability improvement of the PBT/PPO blends by using an ethylene-glycidyl methacrylate graft polystyrene copolymer (EGMA-g-PS) as reactive compatibilizer [7]. Hobbs *et al.* reported the blends of the unfunctionalized and functionalized PPO with PBT or with PBT containing also a styrene-ethylene/butylene-styrene (SEBS) impact modifier [8, 9]. The SEBA rubber is selectively incorporated in the PPO phase when using the unfunctionalized PPO while a significant fraction of the rubber remained encapsulated in the PBT phase when using the functionalized PPO. A few Japanese companies have also claimed that certain GMA-containing copolymers are capable of compatibilizing PBT/PPO blends [10, 11].

Considerable emphasis has been placed on how to compatibilize incompatible blends which have been the subject of several recent reviews [12–15]. Non-reactive block or graft copolymers capable of specific interactions with the blend constituents are the commonly employed approaches. Reactive copolymers of the type $C—X$ may compatibilize an incompatible blend of A and B provided that C is miscible with A and X is capable of reacting or strongly interacting with B [15]. Reactive copolymers containing maleic anhydride have been the focus for reactive compatibilization in the literature. Applications of epoxy-containing polymers or copolymers as reactive compatibilizers have also attracted great attention recently. Chung *et al.* reported that styrene-GMA-methyl methacrylate copolymer acts as a reactive compatibilizer for PC/PET/rubber blends [16]. The styrene-acrylonitrile-GMA (SAG) copolymer has been used as a reactive compatibilizer for PBT/ABS [17, 18] and phenoxy/ABS [19] blends. Ethylene-glycidyl methacrylate (EGMA) copolymer was also used as a reactive compatibilizer in the polycarbonate (PC)/PET [20] and various polyolefins [21] blends. EGMA has also been patented as a reactive compatibilizer for blends of PC/ABS [22], polypropylene (PP)/PBT [23], and PP/PA [24]. The SG copolymer has also been employed as an *in situ* reactive compatibilizer for the PS/PA [25] and PS/PET [26] blends. The epoxy groups in these copolymers are capable of chemically reacting with one or both of the blend components during melt processing provided that the blend constituents contain —OH, —COOH, or amine group. Ethyltriphenyl phosphonium salts have been used commercially as catalyst to manufacture solid epoxy resins [27, 28] and epoxy ester resins [29] which involve reactions of epoxy with —OH and —COOH groups. The possible reaction mechanisms of these catalyzed reactions were described previously [30–32]. Polymer blends of PBT and PPO are considered as incompatible. Copolymers of SG with various GMA contents have been employed in this

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study to compatibilize the polymer blends of PBT and PPO. The epoxy groups in this SG copolymer can react with PBT endgroups to form the SG-g-PBT copolymers which can act as phase compatibilizer for the PBT/PPO blends. The non-reactive segment in this SG copolymer, polystyrene, is known to be fully miscible with PPO. Therefore, the selection of an SG copolymer seems to be an ideal choice as a potential compatibilizer for the polymer blend of PBT and PPO.

EXPERIMENTAL

Materials

The materials used were: (a) a natural grade PBT, from the General Electric, Valox 325; (b) an unmodified PPO, UPO2, an experimental product from Catalyst Research Center of Taiwan; (c) styrene-GMA (SG) copolymers with various GMA contents were prepared by suspension polymerization according to the procedures previously reported [26]; (d) the catalyst employed in this study was ethyltriphenyl phosphonium bromide, obtained from Merck.

Extruder blending and injection moulding

One-step melt blending was carried out by using a 20 mm counterrotating intermeshing twin screw extruder from Welding Engineers. The extruder was operated at 250 rpm and the barrel sectional temperatures were set at 180, 230, 240, 245, 250 and 260°C, respectively. Standard ASTM specimens (tensile, flexural, and falling weight impact) were produced by injection moulding using a machine from Toshiba Co.

Scanning electron microscopy (SEM)

The morphology was examined by SEM, Model S-570, Hitachi Co. from cryogenically fractured specimens in the plane perpendicular to flow direction of injection moulding. The specimens were etched with chloroform for 10 min to extract the PPO phase from the blends.

Melt properties

The torque evolution with time for the melting operation was monitored at 270°C and 30 rpm in a Brabender Plastic-Corder. The melt flow rate (MFR) values were obtained at 250°C with 2.16 kg load for the PBT/PPO = 70/30 blend series and at 260°C with 3.8 kg for the PBT/PPO = 50/50 blend series using an Automatic Flow Rate Timer from Ray-Ran Co.

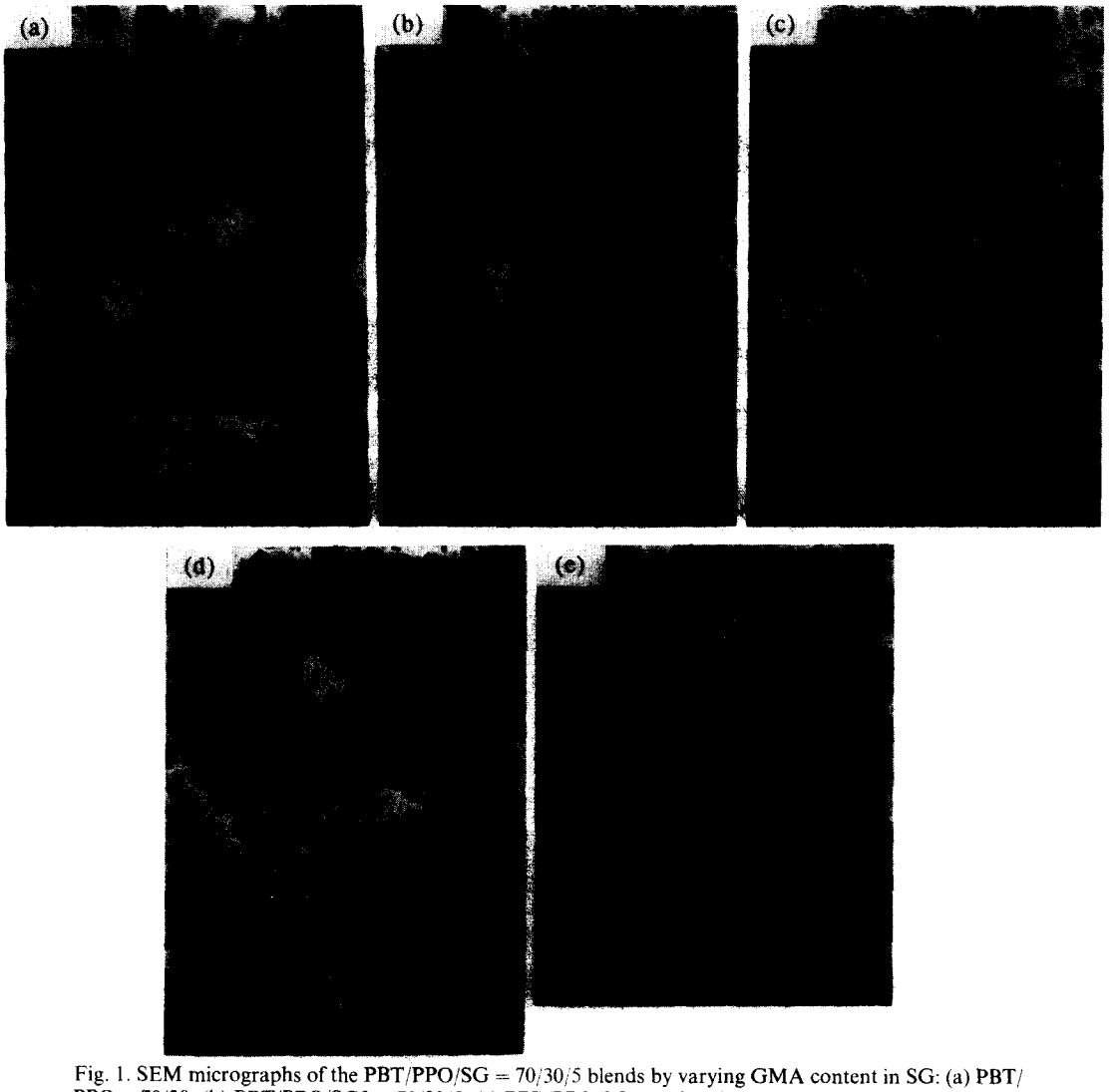

Fig. 1. SEM micrographs of the PBT/PPO/SG = 70/30/5 blends by varying GMA content in SG: (a) PBT/PPO = 70/30; (b) PBT/PPO/SG2 = 70/30/5; (c) PBT/PPO/SG5 = 70/30/5; (d) PBT/PPO/SG10 = 70/30/5; (e) PBT/PPO/SG15 = 70/30/5. The number after SG refers to the %GMA in the compatibilizer.

Mechanical properties

Impact and tensile tests were carried out at ambient temperature. Dynamic mechanical measurements were made on a Rheometric Dynamic Spectrometer (RDS) operating in a standard torsion mode at a frequency of 6.28 rad/sec and 4°C/min heating rate.

Thermal properties

Heat deflection temperature was determined according ASTM-D648 with 66 psi loading. Differential scanning calorimetry (DSC) analysis was carried by a heating rate of 10°C/min (air atmosphere) by using a Seiko I SSC-5000 instrument.

Annealing

Specimens were placed in an air draft oven at 200 and 220°C for different annealing times. The annealed specimens were then broken cryogenically and solvent etched prior to the SEM examination.

RESULTS AND DISCUSSION

SEM morphologies

Figure 1 shows the chloroform etched SEM morphologies of the PBT/PPO/SG = 70/30/5 blends with various GMA content in the SG copolymer. The visible and evenly distributed holes correspond to the PPO phase as this is the minor component in these blends. The non-compatibilized blend [Fig. 1(a)] has the largest PPO domain size. The size of the PPO phase decreases with the increase of GMA content and reaches a minimum at SG5 (5% GMA in the styrene-GMA copolymer) [Fig. 1(c)]. Too high GMA content in styrene-GMA copolymer results in a less effective compatibilizer as mentioned previously [18, 26], which may result from the lower miscibility between PPO and the PS segment in the copolymer. A similar phenomenon has been observed in blends of PPO and styrene-maleic anhydride copolymer (SMA) [6], which are miscible only if the maleic anhydride content in SMA is low. The effect of increasing the amount of SG5 compatibilizer can be revealed by comparing the corresponding morphologies [Figs 1(a), 2(a), 2(b), 1(c) and 2(c)]. The size of the dispersed PPO phase becomes smaller with increasing the SG5 content in the blend.

Processability

The PPO-rich compositions (PPO >50%) could not be processed due to high melt viscosity of these blends at a maximum allowable process temperature of 260°C. Temperatures higher than 260°C caused significant PPO degradation. Melt blending of the non-compatibilized PBT/PPO blends by extrusion gave rise to severe die swelling and melt fracture problems. The presence of the styrene-GMA reactive compatibilizer, on the other hand, significantly decreased die swelling and essentially eliminated melt fracture.

Torque vs time

Figure 3 illustrates plots of the melting torque vs time for the individual components and the corresponding blends. Significantly higher torque values are observed for PPO than PBT at the processing temperature (270°C). The PBT/PPO = 50/50 blend shows a torque significantly lower than the average values recorded for PBT and PPO, which are only slightly higher than PBT. The torque of the SG5

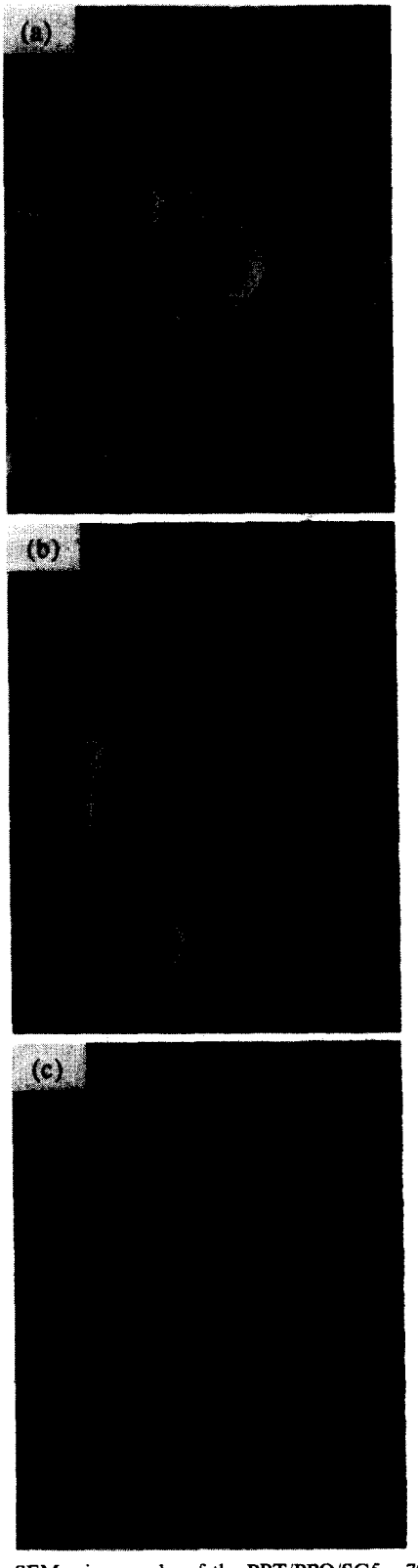

Fig. 2. SEM micrographs of the PBT/PPO/SG5 = 70/30/X blends by varying SG5 content: (a) PBT/PPO/SG5 = 70/30/1; (b) PBT/PPO/SG5 = 70/30/3; (c) PBT/PPO/SG5 = 70/30/10.

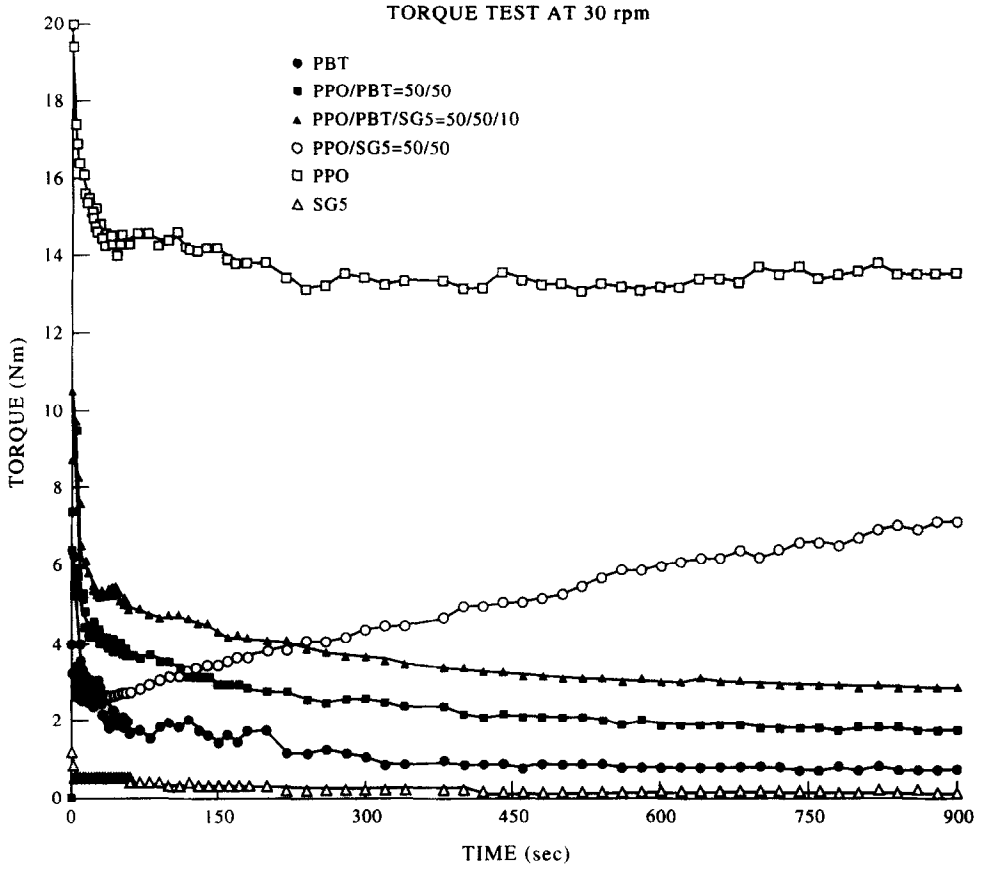

Fig. 3. Torque vs time for individual components and respective blends.

compatibilizer is extremely low at this temperature. The compatibilized blend, PBT/PPO/SG5 = 50/50/10, shows a slightly higher torque value than the corresponding non-compatibilized blend due to the possible occurrence of reactions between SG5 with the blend components. The reaction between SG copolymer with endgroups (—COOH or —OH) of polyesters has, in fact, been well-documented

Table 1. Summarized data on MFR, HDT, and various mechanical properties

			Impact strength		Tensile	
Composition	MFR* (g/10 min)	HDT (°C)	Notched (J/M)	Unnotched (J/M)	Strength (MPa)	Elong. (%)
PBT/PPO = 70/30	7.42	88.9	55	130	33.5	5.74
PBT/PPO/SG2 = 70/30/1	6.13	94.7	58	144	38.8	6.43
PBT/PPO/SG2 = 70/30/3	5.67	95.5	59	172	47.5	8.55
PBT/PPO/SG2 = 70/30/5	5.36	100.6	58	186	50.4	9.57
PBT/PPO/SG2/Cat = 70/30/5/0.05	—	103.5	59	230	50.1	11.98
PBT/PPO/SG2 = 70/30/10	3.81	109.8	59	230	54.4	10.48
PBT/PPO/SG5 = 70/30/1	6.24	90.3	60	164	42.5	7.87
PBT/PPO/SG5 = 70/30/3	5.57	95.4	56	192	51.0	9.74
PBT/PPO/SG5 = 70/30/5	4.27	103.7	56	217	53.0	10.91
PBT/PPO/SG5/Cat = 70/30/5/0.05	—	105.2	61	237	51.4	10.47
PBT/PPO/SG5 = 70/30/10	1.97	100.1	60	281	55.3	10.96
PBT/PPO/SG10 = 70/30/5	4.07	97.4	55	172	48.4	8.81
PBT/PPO/SG10/Cat = 70/30/5/0.05	—	97.9	59	237	51.9	11.11
PBT/PPO/SG15 = 70/30/5	1.73	89.1	60	192	48.9	9.51
PBT/PPO = 50/50	5.03	113.4	55	115	24.1	4.14
PBT/PPO/SG2 = 50/50/5	4.44	121.8	55	180	41.9	7.08
PBT/PPO/SG5 = 50/50/1	3.61	116.5	55	154	35.1	5.96
PBT/PPO/SG5 = 50/50/3	3.17	116.7	55	174	40.2	6.72
PBT/PPO/SG5 = 50/50/5	4.73	126.1	56	195	44.2	7.61
PBT/PPO/SG5 = 50/50/10	3.05	124.7	55	177	36.7	6.29
PBT/PPO/SG5 = 70/30/20	1.76	111.7	56	146	45.1	7.55
PBT/PPO/SG10 = 50/50/5	4.89	120.5	55	152	30.8	5.32
PBT/PPO/SG15 = 50/50/5	5.91	118.6	57	144	38.8	6.54

*The MFR conditions are: PBT/PPO = 70/30 blends: 250°C and 2.16 kg load; PBT/PPO = 50/50 blends: 260°C and 3.80 kg load.

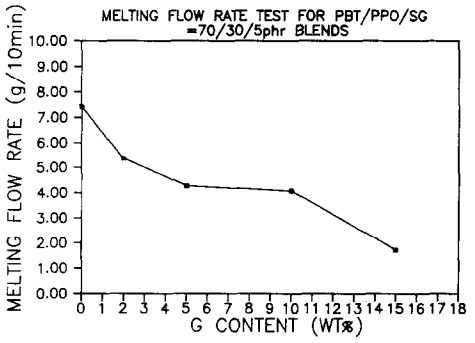

Fig. 4. Effect of GMA content in SG on the melt flow rate of the PBT/PPO/SG = 70/30/5 blends.

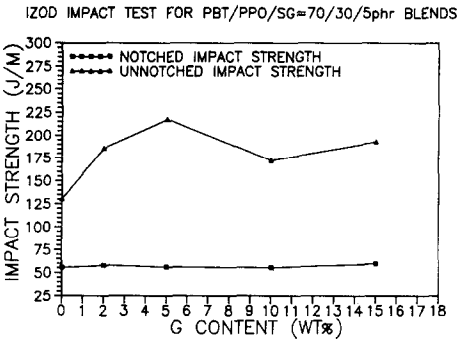

Fig. 6. Effect of GMA content in SG on Izod impact strength of the PBT/PPO/SG = 70/30/5 blends.

[11, 20, 26]. The torque increase observed for the PPO/SG5 = 50/50 blend indicates that reactions between SG and the phenolic—OH endgroups from PPO are also occurring during melting.

Melt flow rates (MFR)

The melt flow rates from all the blends investigated are summarized in Table 1. Figure 4 clearly demonstrates that the higher GMA content in SG copolymer results in lower MFR of the blend, while the presence of greater amounts of compatibilizer also results in lower MFR values for the blends as would be expected from the recorded torque values during blending.

Heat distortion temperatures (HDT)

Figure 5 shows the effect of the GMA content in the SG compatibilizer on the HDT value of the PBT/PPO/SG = 70/30/5 blend. The presence of only 5 phr of SG2 increases the HDT by 11°C over the corresponding non-compatibilized counterpart. The maximum HDT is obtained at 5% GMA in the SG compatibilizer (SG5). As mentioned earlier, too high GMA amount in the SG copolymer tends to reduce the compatibilization efficiency. Comparing the HDT results with the morphologies of the same materials it is revealed that the blend with SG5 has the smallest particle size of the dispersed phase and also has the highest HDT. The general trend is that the blend containing more compatibilizer exhibits higher HDT value. The presence of catalyst produces further rises in HDT values but these are not very large, as shown in Table 1. Very similar HDT results are also obtained for PBT/PPO = 50/50 blends containing varying amounts of compatibilizer (Table 1).

Izod impact strengths

PBT and PPO are both brittle polymeric materials when tested with notched specimens, hence the PBT/PPO blends are also expected to be brittle. The notched impact strength of PBT/PPO blends was found to remain essentially unchanged even in the presence of the reactive compatibilizer as shown in Table 1 and Fig. 6. Figure 6 also shows the effect of GMA content in SG reactive compatibilizer on the unnotched impact strength and reveals that the blend containing SG5 as compatibilizer has the highest unnotched impact strength. The addition of catalyst further increases the unnotched impact strength, as shown in Table 1.

Tensile and falling weight impact properties

The trend for the tensile test results in terms of tensile strength and elongation to break is about the same as for HDT and unnotched impact strength. Figure 7 shows the plot of tensile strength vs GMA content of the SG copolymer for the PBT/PPO/SG = 70/30/5 blends. The highest tensile strength values are obtained for blends containing the SG5 compatibilizer (Table 1). Figure 8 shows a very similar trend for the elongation at break. Although the tensile modulus measurements were usually not very accurate however, a similar trend as tensile strength and elongation could be deduced (data not shown). Figure 9 shows the trend obtained from the falling weight impact is similar to that from the Izod impact.

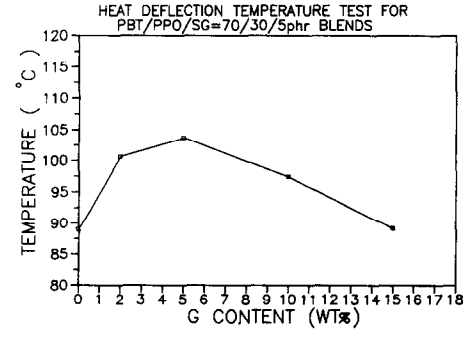

Fig. 5. Effect of GMA content in SG on HDT of the PBT/PPO/SG = 70/30/5 blends.

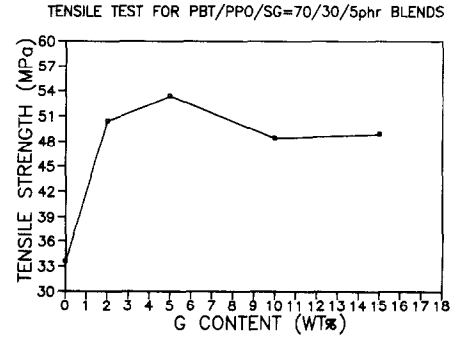

Fig. 7. Effect of GMA content in SG on tensile strength of the PBT/PPO/SG = 70/30/5 blends.

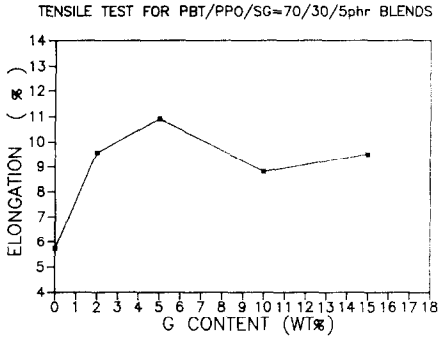

Fig. 8. Effect of GMA content in SG on tensile elongation of the PBT/PPO/SG = 70/30/5 blends.

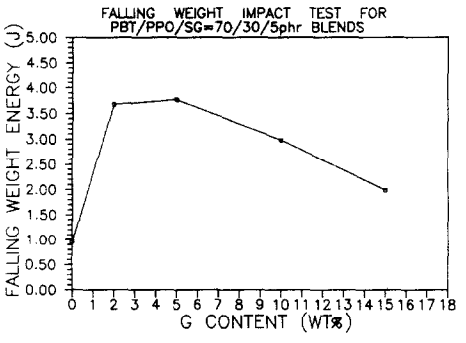

Fig. 9. Effect of GMA content in SG on falling weight impact energy of the PBT/PPO/SG = 70/30/5 blends.

Dynamic mechanical analysis

Dynamic mechanical analysis is relatively more sensitive than DSC in locating the polymer transitions, especially the secondary transitions. Plots of tan δ vs temperature for the PBT/PPO/SG5 = 50/50/X blends with varying SG5 content are given in Fig. 10. The glass transition temperatures (T_g) of PBT in the PBT/PPO blends is located around 50°C and is found to increase slightly with increasing SG5 content in the blend. The uncompatibilized PBT/PPO blends show a broad T_g peak for the PPO phase at 225°C and a T_m peak for PBT at 236°C. Both the T_g of PPO and the T_m of PBT for the PBT/PPO blend gradually decrease with increasing SG5 amounts. A broad peak at 125°C is found for the blends with high SG5 content which is believed to be due to the glass transition of the SG5. The T_g of SG5 was found to be 110°C by DSC analysis (data not shown). The miscibility of PPO and SG depends on GMA content, similar to that between PPO and SMA [6]. The observed T_g decrease for the PPO phase may be attributed to the *in situ*-formed SG-g-PPO copolymer and the partial miscibility between PBT and PPO.

Differential scanning calorimetry (DSC)

Figure 11 shows the DSC thermograms of PBT, PPO, and the PBT/PPO blends. The PPO obtained in powder crystalline form, presumably recovered from

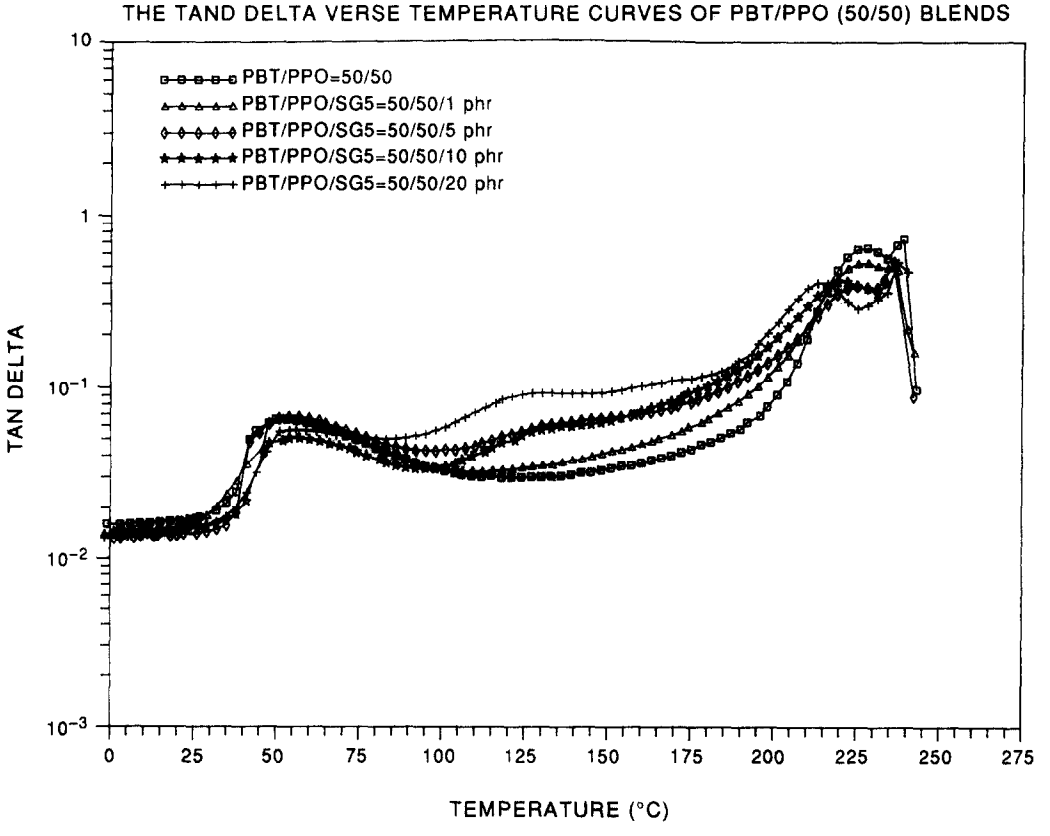

Fig. 10. Plots of tan δ vs temperature of the PBT/PPO/SG5 = 50/50/X blends by varying SG5 quantity.

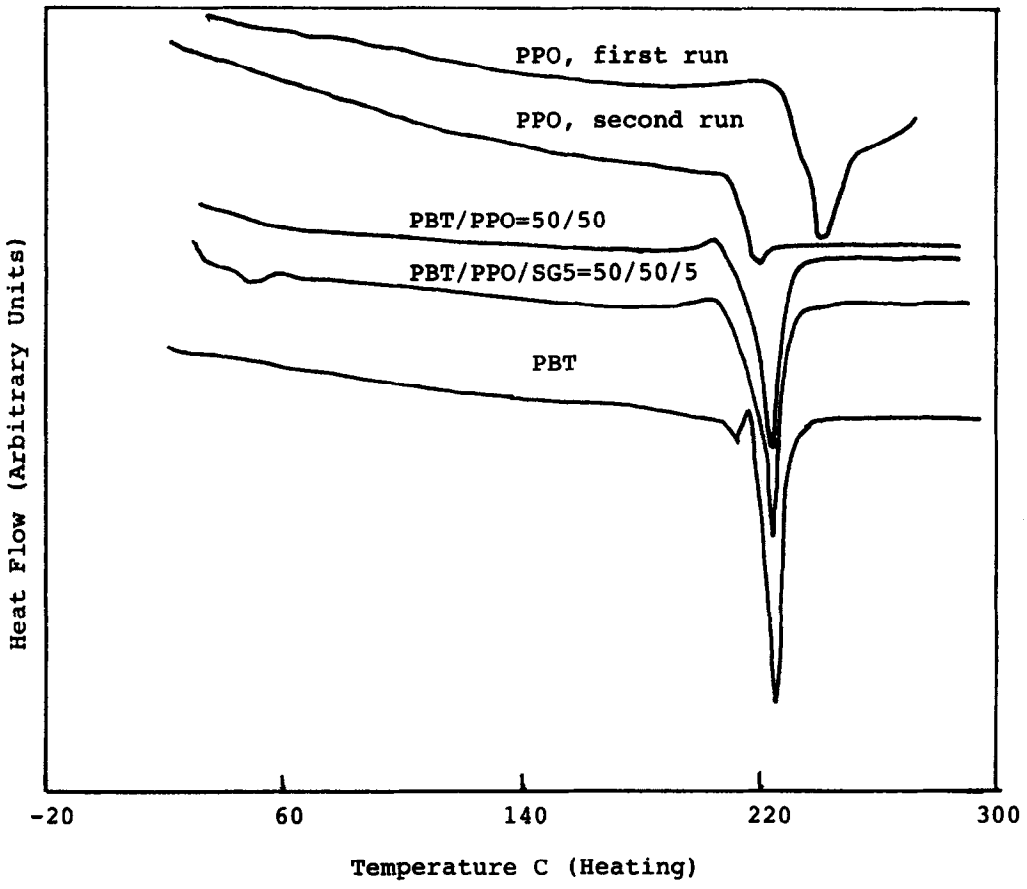

Fig. 11. DSC thermograms of PBT, PPO, and PBT/PPO blends.

solvent, has a T_m of 241°C. The quenched PPO (second run) shows a T_g of about 216°C which is identical to the value reported by Hseih and Peiffer [5]. The T_g of PBT (about 45°C) cannot be detected by the DSC analysis due to its high degree of crystallinity, while the T_m of the crystalline PBT is located at about 228°C. The PBT/PPO = 50/50 blend shows a relatively broad peak at 224°C resulting from an overlap between the glass transition of PPO and the melting point of the crystalline PBT. T_g, T_m and melting enthalpy (ΔH_m) of the blends investigated are summarized in Table 2. The expected effects of the compatibilizer on the crystallinity level of the PBT phase, however, is not borne out in the results observed.

Table 2. Summarized data on DSC analyses

Composition	T_g (°C)	T_m (°C)	ΔH_m (mJ/mg)
PBT	44.1	228.4	39.5
PPO (first run)	—	241.4	15.3
PPO (quenched)	216.1	—	—
PBT/PPO = 70/30	45.5	224.2	31.0
PBT/PPO/SG2 = 70/30/1	48.2	224.9	28.4
PBT/PPO/SG2 = 70/30/3	47.5	224.2	30.6
PBT/PPO/SG2 = 70/30/5	48.9	224.8	32.1
PBT/PPO/SG2 = 70/30/10	49.3	224.3	26.8
PBT/PPO/SG5 = 70/30/5	47.2	224.2	31.0
PBT/PPO/SG10 = 70/30/5	46.4	224.2	27.5
PBT/PPO = 50/50	50.2	224.2	23.9
PBT/PPO/SG2 = 50/50/5	52.1	222.7	20.1
PBT/PPO/SG5 = 50/50/1	51.9	223.6	24.0
PBT/PPO/SG5 = 50/50/3	52.5	222.8	19.6
PBT/PPO/SG5 = 50/50/5	52.7	224.2	22.4

Except one PPO test, all data were obtained from the second run after quenching.

Annealing

Figure 12 shows the SEM micrographs of the non-compatibilized PBT/PPO = 50/50 blend before and after annealing for 60 min at 200 and 220°C. It can be clearly seen that the phase morphology becomes coarser after annealing. The interfacial tension, in fact, provides the driving force for coarsening of the phases for an immiscible blend system when it is annealed near the T_g of the blend components. Figure 13 shows that the compatibilized PBT/PPO/SG5 = 50/50/5 blend, on the other hand, displays no change in morphology after annealing. The presence of the reactive compatibilizer is able to stabilize the phase morphology during annealing by reducing the interfacial tension.

CONCLUSIONS

Polyblends of PBT and PPO are immiscible and incompatible with poor interfacial adhesion and large phase domains and are difficult to process. The SG copolymer itself does not function as a compatibilizer, but will become one after reacting with PBT

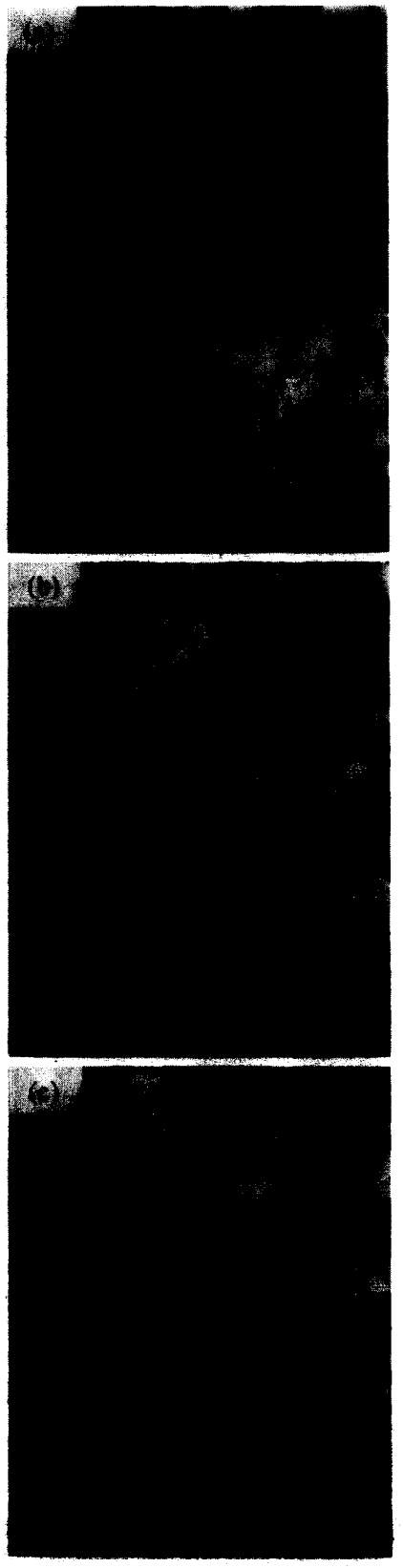

Fig. 12. SEM micrographs before and after annealing of the non-compatibilized PBT/PPO = 50/50 blend: (a) before annealing; (b) after annealing at 200°C for 60 min; (c) after annealing at 220°C for 60 min.

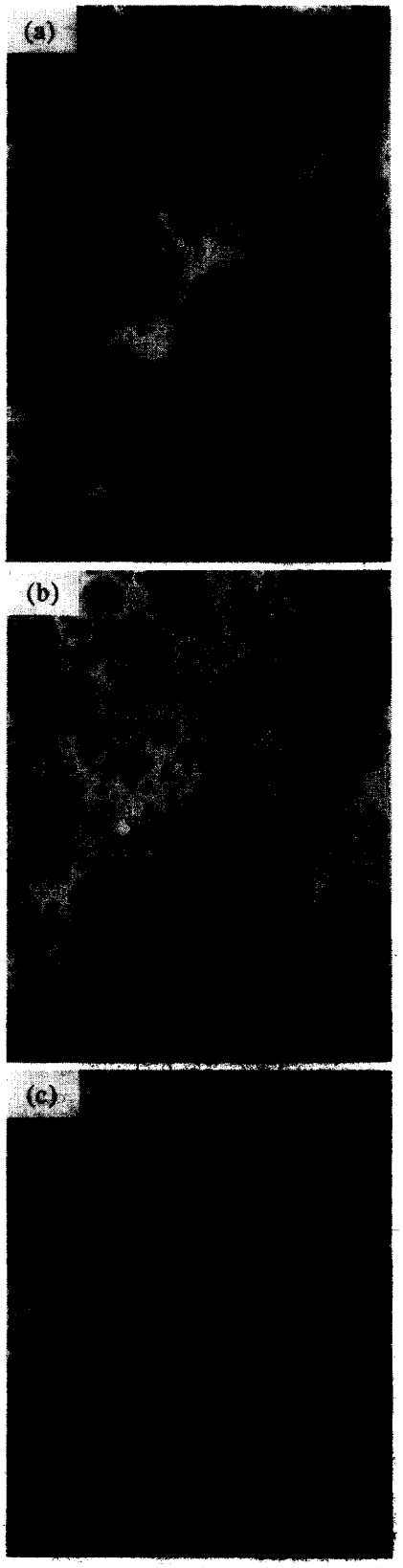

Fig. 13. SEM micrographs before and after annealing of the compatibilized PBT/PPO/SG5 = 50/50/5 blend: (a) before annealing; (b) after annealing at 200°C for 60 min; (c) after annealing at 220°C for 60 min.

endgroups (—OH or —COOH) to form the SG-g-PBT copolymer during melt blending. The presence of a small amount of ethyltriphenyl phosphonium bromide catalyst accelerates the graft reaction and results in a better compatibilized blend. The maximum efficiency of compatibilization was found by adding about 5–10% SG copolymer with 5% GMA in the copolymer. The compatibilized PBT/PPO blend has a smaller phase domain and a slightly higher melt viscosity then the corresponding non-compatibilized blend. The compatibilized PBT/PPO blend also shows substantial improvements in mechanical properties, heat distortion temperature, and phase stability during thermal annealing over the non-compatibilized counterpart.

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