

# Effects of the compatibilizer PP-g-GMA on morphology and mechanical properties of PP/PC blends

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Effects of the compatibilizer polypropylene grafted with glycidyl methacrylate (PP-g-GMA) on the morphology, thermal, rheological and mechanical properties of polypropylene and polycarbonate blends (PP/PC) were studied. It was found that the addition of PP-g-GMA significantly changed their morphology. The mean size of domains reduced from 20  $\mu\text{m}$  to less than 5  $\mu\text{m}$ . The dispersed domain size is also strongly dependent upon the content of PP-g-GMA. The interfacial tension of PP/PC/PP-g-GMA (50/30/20) is only about one-tenth of PP/PC (70/30). The crystallization temperature of PP in PP/PC/PP-g-GMA is 5–8°C higher than that of PP in PP/PC blends. Characterization studies based on mechanical properties, differential scanning calorimetry, rheology and morphological evidence obtained by using scanning electron microscopy support the hypothesis that an in-situ copolymer PP-g-PC was formed during the blending process. © 1997 Elsevier Science Ltd.

(Keywords: polypropylene; polycarbonate; compatibilizer)

## INTRODUCTION

Blending of polyolefins with engineering plastics such as polyesters and polyamides could be a useful method for upgrading the properties of polyolefins. The major problem is the lack of miscibility and, consequently, surface active substances are needed to compatibilize the constituents in the blend which are mutually immiscible. Another approach is reactive compatibilization, for example by functionalizing the polyolefins. In the past two decades considerable efforts have been reported in the literature concerning reactive compatibilization of polyolefins and blending with engineering plastics<sup>1–7</sup>. A well-known procedure for functionalizing polyolefins is to graft maleic anhydride, methacrylic acid etc. onto polyolefins<sup>8–12</sup>.

In recent years glycidyl methacrylate (GMA) has attracted much attention for functionalizing polyolefins, since the epoxy groups of GMA can react with the corresponding reactive groups, such as –OH, –COOH, –NH<sub>2</sub> etc., of other polymers to form new covalent bonds across the interfaces. Chung and Carter<sup>11</sup> patented styrene–glycidyl methacrylate–acrylonitrile (SGA) copolymer which was used as a compatibilizer of PC and ABS. Akkapeddi *et al.*<sup>13</sup> reported that polyethylene-g-GMA acts as a good compatibilizer of blends of PC with PET and various polyolefins. F. C. Chang and his co-workers<sup>14–16</sup> have reported a series of reactive compatibilizations of many blending systems based on GMA-containing copolymers.

The mechanical properties and morphology of PP/PC blends, without addition of any compatibilizer, obtained in

an internal mixing chamber and also in a twin-screw extruder have been examined by Favis and co-workers<sup>17–19</sup>. The morphology of samples with different compositions and obtained with various processing parameters was estimated on the basis of optical and electron microscopy. The authors found that the size and shape of PC particles dispersed in a PP matrix depended significantly on processing conditions as well as on the contribution of particular components. The crystallization of PC/PP blends was investigated by wide angle X-ray scattering<sup>20</sup>.

Here, we report the effects of compatibilizer PP-g-GMA on morphology, interfacial behaviour, rheological and mechanical properties of PP/PC blends.

## EXPERIMENTAL

### Materials and preparation

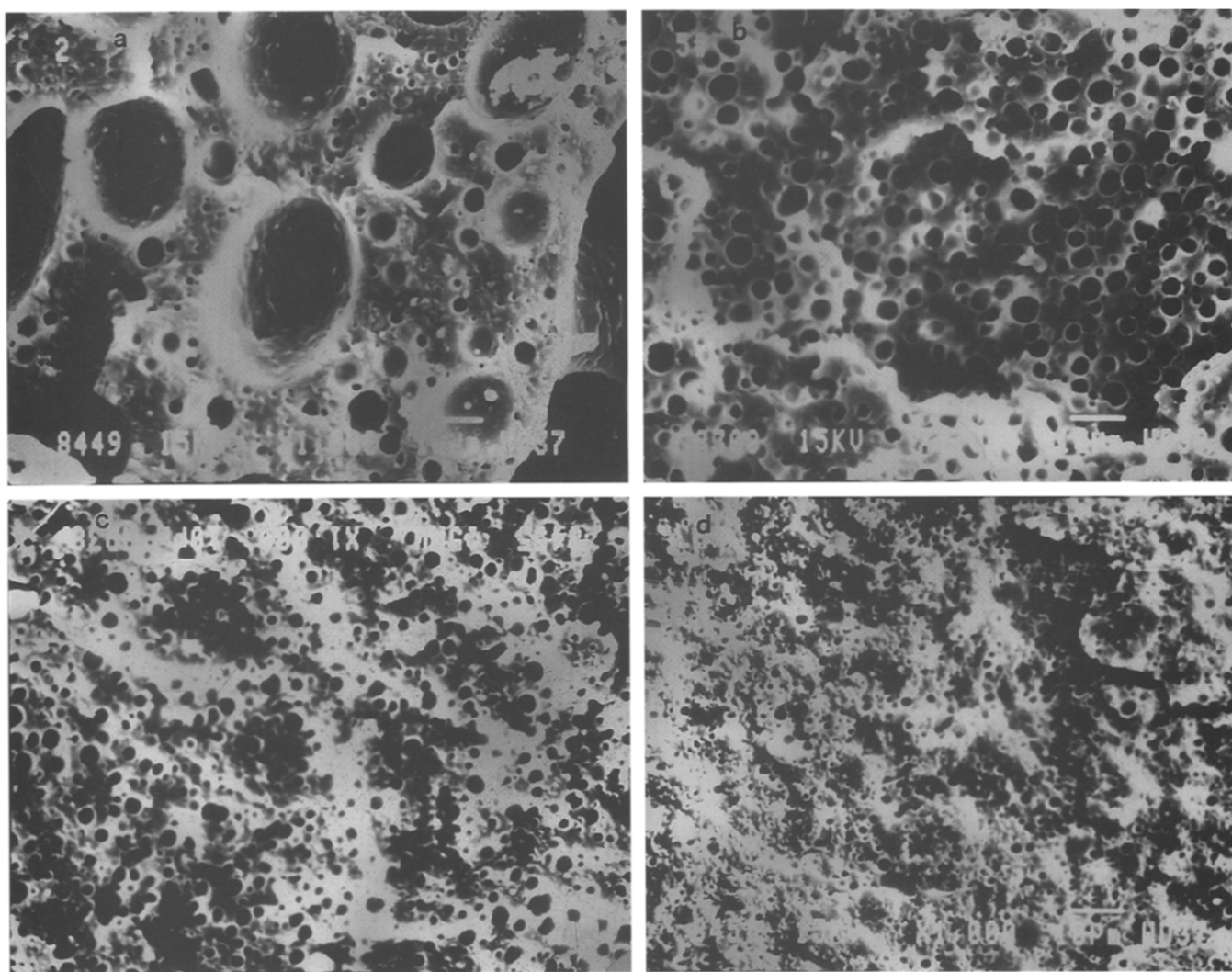
PP (more precisely, a propylene/ethylene random copolymer containing 8% mol ethylene) used in this work was offered by Pan Jing Petrochemical Co. Ltd (China). Its melt index is 1.0 g/10 min (230°C, 2.16 kg). PC was purchased from Mitsubishi P. Co. Ltd (Japan), commercial model MS 3000. PP-g-GMA was prepared in this laboratory with a GMA content of 0.46 mol%. Details of preparation of PP-g-GMA were reported in a previous publication<sup>21</sup>.

Blends of PP/PC and PP/PC/PP-g-GMA were prepared using an SHJ-30 corotating twin-screw extruder ( $\phi = 30$  mm,  $L/D = 32$ ). Before blending, PC was dried for 12 h at 90°C. The rotating rate of the screw is 100 rpm, and the blending temperature is 250°C. Compositions of PP/PC were 100/0, 90/10, 80/20 and 70/30 (weight percent); the compositions of PP/PC/PP-g-GMA are listed in *Table 1*.

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**Table 1** Thermal parameters of PP/PC and PP/PC/PP-g-GMA blends

PP (wt%)	PC (wt%)	PP-g-GMA (wt%)	$T_g$ of PC (K)	$T_c$ of PP (K)	$T_m$ of PP (K)	Crystallinity of PP (%)
100	0	0		386.6	436.8	43.5
90	10	0	431.2	386.4	436.8	41.7
80	10	10	429.1	392.4	436.7	41.9
80	20	0	431.2	386.5	436.7	40.2
70	20	10	429.2	392.8	436.6	40.1
70	30	0	431.2	386.6	436.8	38.4
67.5	30	2.5	430.9	391.5	436.6	38.9
65	30	5	429.6	392.4	436.8	39.6
60	30	10	428.8	394.3	436.8	39.9
50	30	20	428.6	394.6	436.8	38.8



**Figure 1** SEM micrographs of PP/PC and PP/PC/PP-g-GMA blends: (a) PP/PC (70/30 wt%); (b) PP/PC/PP-g-GMA (67.5/30/2.5 wt%); (c) PP/PC/PP-g-GMA (65/30/5 wt%); (d) PP/PC/PP-g-GMA (60/30/10 wt%)

*Characterization and measurements*

The morphologies of PP/PC and PP/PC/PP-g-GMA samples were observed by using a JEOL JXA-840 scanning electron microscope (Japan). Before observation, the examined sections were etched with dichloromethylene for 4 h to increase the contrast.

The thermal behaviour of two blending systems was investigated using a Perkin-Elmer DSC-2 with a heating rate of 10°C min<sup>-1</sup>. All measurements were performed under a nitrogen atmosphere.

A cone-plate rheometer, Contrive Rheomat 120, was used to measure the apparent melting viscosities of blending samples. The diameter of the cone plate is 20 mm and cone angle is 5°. Test specimens about 0.2 mm thick were prepared by moulding. Experiments were carried out under a nitrogen atmosphere.

Wide angle X-ray diffraction profiles of PP/PC and PP/PC/PP-g-GMA blends were obtained in a PW1700 Philips diffractometer with Cu target K $\alpha$ , Ni-filtered radiation. The tube voltage is 40 kV and the current is 30 mA.

Specimens used for mechanical measurements were prepared using a JSW17SA injector (Japan) with a barrel temperature of 220–250°C and an injection pressure of 60 MPa. Before testing, all specimens were completely dried for 18 h at 90°C. Izod impact strengths of specimens were measured with an XJU-22 impact tester (Chen De, China) according to the standard ISO/R 180–1961. Measurements of tensile properties of specimens were performed on an Instron 1121 tester to the standard ISO/R527–1966E.

## RESULTS AND DISCUSSION

### Morphology and structure

SEM photomicrographs of sections of PP/PC (70/30) and

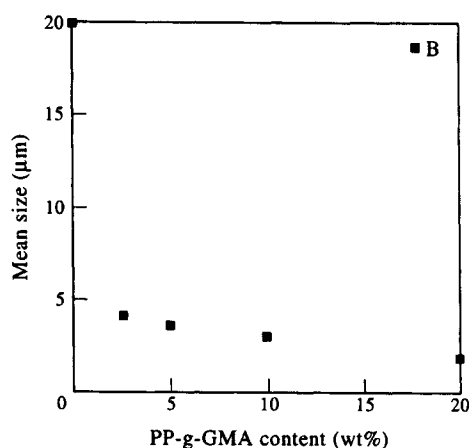


Figure 2 Mean size of PC domains versus PP-g-GMA content

PP/PC/PP-g-GMA (67.5/30/2.5, 65/30/5, 60/30/10) specimens are shown in Figure 1. The smooth holes visible in Figure 1a were left by etched PC domains and are 3–30 μm in diameter. This feature suggested that the interfacial adhesion between PP matrix and PC domains was very poor: they are completely immiscible. On the other hand, the holes shown in Figure 1b become smaller and more homogeneous than those in Figure 1a. The diameters of PC domains are 2–5 μm. With increasing content of PP-g-GMA in ternary blends, the size of PC domains decreases further. Even PC particles trapped in the PP matrix can be discovered.

The relationship between mean size of PC domains and PP-g-GMA content is shown in Figure 2. These results indicate that PP-g-GMA can act as a compatibilizer of PP/PC binary blends.

Wu<sup>22</sup> proposed a simple relation to correlate the minimum domain size ( $d_{\min}$ ) with interfacial tension ( $\sigma$ ) for binary blends:

$$d_{\min} = \frac{4\sigma}{\eta\dot{\gamma}}$$

where  $\dot{\gamma}$  is the shear rate adopted in preparing the blend, and  $\eta$  is the melt viscosity of the matrix phase. This expression was derived on the basis of the balance between the shearing force, which tends to deform the minor phase into droplets, and the interfacial tension, which tends to restore the spherical shape of the minor phase. In preparations of blends of PP/PC and PP/PC/PP-g-GMA the same mixing process was adopted. This meant that the parameters  $\dot{\gamma}$  and  $\eta$  could be assumed as constants. Therefore, the interfacial tensions of two kinds of blends are

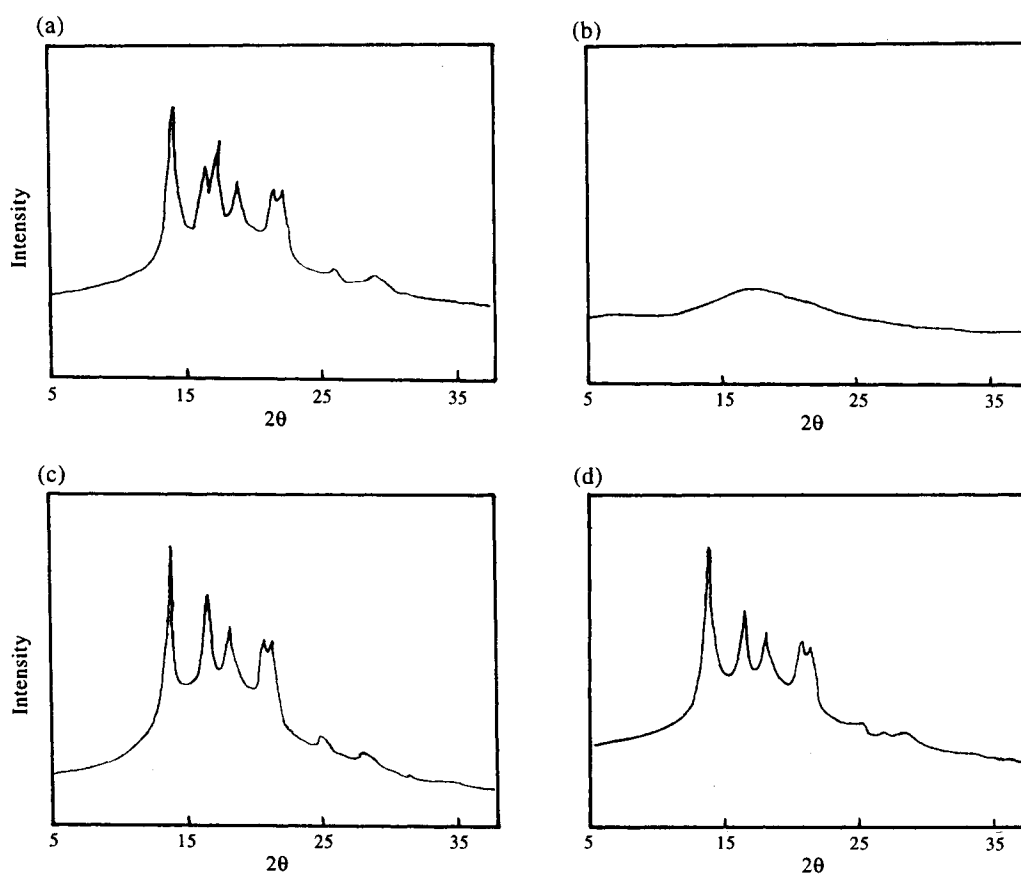


Figure 3 WAXD patterns of (a) PP; (b) PC; (c) PP/PC (70/30 wt%); (d) PP/PC/PP-g-GMA (60/30/10 wt%)

proportional to the parameter  $d_{\min}$ , which provides a useful approach for quantitative measurements of interfacial tension. In this work the mean interfacial tension is about  $45 \text{ mN m}^{-1}$  of PP/PC (70/30) and less than  $4.0 \text{ mN m}^{-1}$  for PP/PC/PP-g-GMA (50/30/20) blends.

Wide angle X-ray diffraction (WAXD) patterns of PP, PC, PP/PC (70/30) and PP/PC/PP-g-GMA (60/30/10) are presented in Figure 3a, 3b, 3c and 3d, respectively. The diffraction pattern of PC shows that PC is completely amorphous. In Figure 3a, the diffraction peak at  $2\theta = 15.9^\circ$  corresponds to the 300 diffraction plane of the  $\beta$  crystal form (hexagonal). However, it disappeared after blending with PC, as shown in Figure 3c, d. This feature suggested that the mixture of  $\alpha$  and  $\beta$  crystal forms of plain PP is replaced by the pure  $\alpha$  crystal form in PP/PC blends. Addition of the compatibilizer PP-g-GMA did not affect the crystal structure of PP in blends.

*Thermal behaviour*

The thermal properties of PP/PC and PP/PC/PP-g-GMA

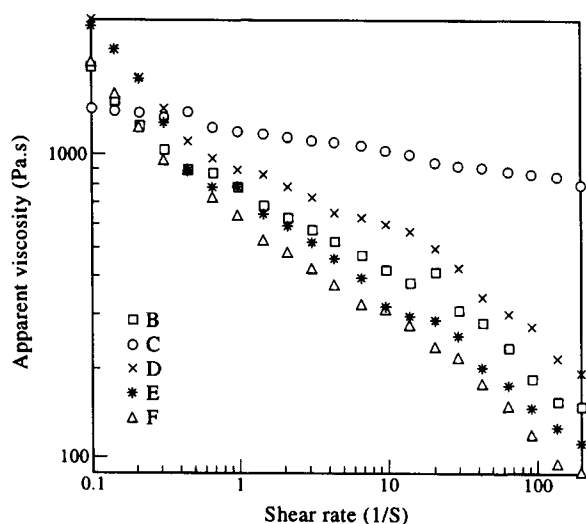


Figure 4 Relationship of the apparent viscosities of PP (\*), PC (O), PP-g-GMA (-), PP/PC/(70/30) ( ) and PP/PC/PP-g-GMA (60/30/10) ( x ) versus shear rates

blending systems are shown in Table 1. The melting temperature ( $T_m$ ) of PP remains constant for all blends. However, the crystallization temperature of PP in the ternary blending system is 5–8°C higher than that of PP in the binary blends. This feature can be tentatively explained as being that GMA in the compatibilizer PP-g-GMA acts as a nucleating agent in the PP crystallization process. As shown in Table 2, the crystallinity of PP decreased with addition of PC. There was no obvious effect of the compatibilizer, PP-g-GMA, on the crystallinity of PP in blends. In this work,  $209 \text{ J g}^{-1}$  was adopted as the melting enthalpy of fully crystalline PP<sup>23</sup>.

From Table 1, it can be seen that the glass transition temperature of PC remained constant in the PP/PC binary blends, but shifted to lower temperature with increasing PP-g-GMA content in the ternary blends. This feature offered us another direct evidence that the copolymer PP-g-GMA is an effective compatibilizer for the PP/PC blending system.

*Rheological properties*

Apparent viscosities of PC, PP, PP-g-GMA, PP/PC (70/30) and PP/PC/PP-g-GMA (60/30/10) as a function of shear rates at 250°C are shown in Figure 4. The apparent viscosity of the PP/PC/PP-g-GMA blend is higher than the value of the PP/PC sample. As indicated in their compositions, the ratio of PP (including PP-g-GMA in PP) to PC is nearly the same for the two blends. The difference in their apparent viscosities may result from the possible coupling reaction between PP-g-GMA and PC. The epoxy group of PP-g-GMA could react with the terminal hydroxyl group of PP as follows:

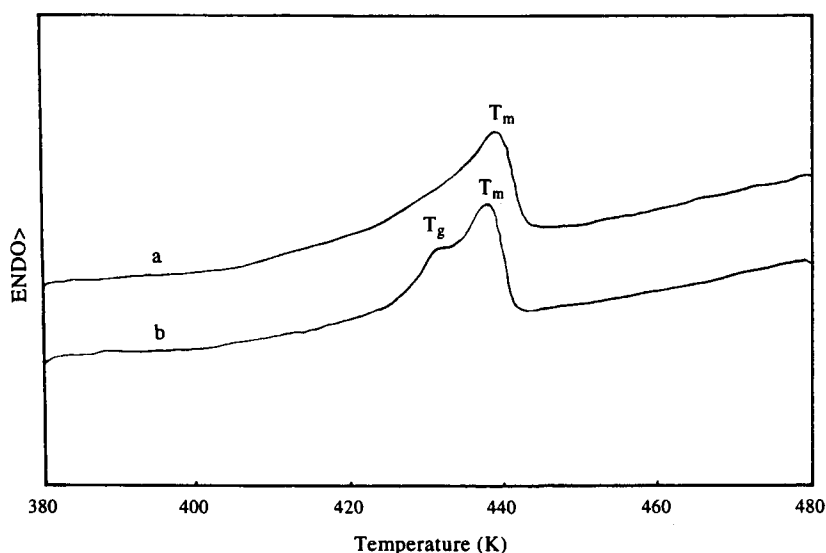
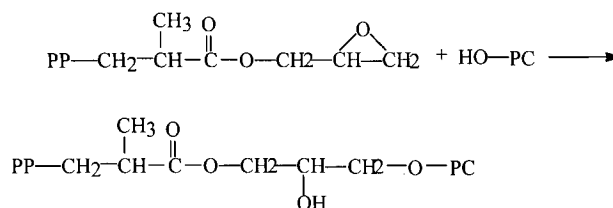


Figure 5 Thermograms of (a) PP/PC (70/30 wt%) and (b) PP/PC/PP-g-GMA (50/30/20 wt%) after extraction with dichloromethylene for 8 h

**Table 2** Mechanical properties of PP/PC and PP/PC/PP-g-GMA blends

PP (wt%)	PC (wt%)	PP-g-GMA (wt%)	Young's modulus $E_y$ (MPa)	Tensile strength $\sigma_b$ (MPa)	Elongation at break $\epsilon_b$ (%)	Izod impact strength ( $J m^{-1}$ )
90	10	0	702	26.4	122	109.8
80	10	10	844	32.8	197	134.5
80	20	0	674	24.3	31.4	58.3
70	20	10	799	30.7	64.5	67.2
70	30	0	625	22.5	5.7	47.3
67.5	30	2.5	660	27.7	14.8	51.1
65	30	5	725	30.5	32.6	56.3
60	30	10	803	36.2	49.7	67.4
50	30	20	874	42.8	69.4	78.9

The above possible coupling reaction was verified by the following experiment. Figure 5 shows the thermograms of PP/PC and PP/PC/PP-g-GMA blends in which PC was extracted out with the solvent dichloromethylene. Thermogram (a) corresponds to the extracted insoluble residue of the PP/PC blend. The fusion peak at 438 K can be attributed to  $T_m$  of PP. In thermogram (b), except for the melting peak of  $T_m$  of PP at the same temperature, a transition temperature corresponds exactly to the  $T_g$  value of PC. This means that there was still some insoluble PC left in the insoluble residue of the PP/PC/PP-g-GMA blend. It is possible that the coupling reaction between PP-g-GMA and PC makes it impossible to extract all the PC phase from the ternary blend.

#### Mechanical properties

The mechanical properties of dried PP/PC and PP/PC/PP-g-GMA samples are listed in Table 2. Compared with the binary PP/PC blends, Young's modulus, tensile strength, elongation at break and Izod impact are significantly improved. For example, the tensile strength and the Izod impact strength of PP/PC/PP-g-GMA (50/30/20) are 90% and 67% higher than the values for PP/PC (70/30), respectively. The modification of mechanical properties of the ternary blends can be attributed to the contribution of the compatibilizer, PP-g-GMA, which acts as an 'interfacial agent' and reduces the interfacial tension, minimizes and homogenizes the domain size, and increases the interfacial adhesion.

#### CONCLUSIONS

PP-g-GMA is an effective compatibilizer for the PP and PC blending system. With addition of PP-g-GMA, the size of PC domains is significantly reduced and the interaction between different phases is greatly improved. The crystal form of crystalline PP changes from a mixture of  $\alpha$  and  $\beta$  into a unique  $\alpha$  crystal form when PP is incorporated with PC. Thermal analysis data show that GMA grafted onto PP molecular chains can act as a nucleating agent which enhances the crystallization temperature of PP. Due to the compatibilization of PP-g-GMA, the glass transition temperature of PC in the ternary blends shifts towards lower temperature. The apparent viscosities and mechanical properties, such as Young's modulus, tensile strength,

elongation at break and Izod impact strength, of PP/PC/PP-g-GMA are much higher than the values for PP/PC at the same content of PC in both blending systems, and increase with increasing content of PP-g-GMA. These features can be tentatively attributed to the fact that the epoxy group of PP-g-GMA reacts with the terminal hydroxyl group of PC to form coupling structures, PP-g-GMA-PC, during the blending process, which reduce the interfacial tension and promote the adhesion of the two phases.

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