

# Study on Poly(propylene)/Ammonium Polyphosphate Composites Modified by Ethylene-1-octene Copolymer Grafted with Glycidyl Methacrylate

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**ABSTRACT:** The compatibilization effect of ethylene-1-octene copolymer grafted with glycidyl methacrylate (POE-g-GMA) as an interface compatibilizer on the mechanical and combustion properties, and the morphology and structures of the cross sections of ammonium polyphosphate (APP)-filled poly(propylene) (PP) were investigated by thermogravimetry, dynamic mechanical analysis, and differential scanning calorimetry. The results indicated that the toughness of the PP/APP composites increased rapidly with adding POE-g-GMA; the dynamic mechanical spectra revealed that the increase of the toughness was closely related to the peaks of loss modulus ( $E''$ ) and mechanical loss ( $\tan \delta$ ). The improvement of the dispersion of APP in the PP matrix was attributed to the addition of POE-g-GMA; it was found that the interfacial adhesion between the filler and matrix

was enhanced when the grafting material was added to the composites. Under such circumstances, the ratio of char formation was increased when the PP composites were heated, although the content of flame retardant was not changed, so the flame retardance of the material was improved. The addition of POE-g-GMA increased the rate of crystallization. At the same time, the degree of crystallinity and the temperature at the beginning of crystallization were decreased, although exerting little influence on the melt behavior of the crystallization of the composites. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 93: 412–419, 2004

**Key words:** poly(propylene) (PP); POE-g-GMA; ammonium polyphosphate (APP); flame retardance; toughness

## INTRODUCTION

Critical fire retardance standards and the expansion of plastic markets make the application of flame-retardant polymer materials increasingly widespread in the construction and automobile industries, home appliances, electrical equipment, and so on.<sup>1</sup> It is effective to add halogen-containing flame retardants into polymer materials to improve the flame-retardant property. However, halogen-containing materials give off thick smoke and contribute to hazardous gas during burning, which may increase the potential for injury to people and property. Recently, the discussion on toxicological effects of certain halogenated fire retardants and the resulting legislative restrictions have fostered the development of phosphorus-containing compounds as alternative halogen-free flame-retardant additives. Phosphorus-containing compounds include red phosphorus, inorganic phosphates, and organic phosphorus compounds.<sup>2</sup> Ammonium polyphosphate (APP), as a component of intumescent flame-retardant formulations, has been

used in poly(propylene) (PP).<sup>3–6</sup> However, satisfactory flame retardance of PP requires high contents of APP, which leads to a marked deterioration in certain physical properties of the materials, particularly their toughness.<sup>7,8</sup>

Now, the key problem in the preparation of APP/PP composites is how to solve the problem of the incompatibility of the hydrophilic inorganic filler and the hydrophobic PP matrix and then improve the properties of compounds.<sup>9</sup> Interfacial action is very important in a filled polymer system: strong interfacial action can achieve good interfacial cementation and effective stress transfer, which can lead to the improvement of properties of the composites. Because of the thermodynamic incompatibility between nonpolar PP and polar APP, use of a compatibilizer is an effective way of preparing PP/APP composites.

PP can be functionalized if a polar group is grafted through chemical reaction. Mai et al.<sup>10</sup> and Ma et al.<sup>11</sup> studied the effects of PP-g-AA (PP grafted with acrylic acid) and PP-g-MAH (PP grafted with maleic anhydride) on the properties of PP/intumescent flame-retardant composites. However, the experiments showed that during the PP grafting reaction, the PP main chain easily degraded, leading to a rapid decrease of molecular weight and low grafting ratio.

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Therefore, it is difficult to obtain a grafted PP with high grafting ratio and low melt flow index.

POE, commercially named Engage, is a saturated ethylene-1-octene copolymer. It has good thermal stability, weather resistance, ageing resistance, excellent toughness, and good toughening modification effect on PP. The methacrylate group in glycidyl methacrylate (GMA) has high reactivity and grafting efficiency. The polar group in GMA gives good interfacial affinity with polar APP flame retardant. The effect of POE-g-GMA on the flame-resistant property, mechanical properties, dynamic mechanical properties, melting and crystalline behavior of PP/APP composites, and its mode of action are reported and discussed in this article.

## EXPERIMENTAL

### Materials

Poly(propylene) (PP2401, melt index 2.5 g/10 min, density 0.91 g/cm<sup>3</sup>) was supplied by Sinopec Yanshan PetroChemicals (Beijing, China). POE (Engage8100, melt index 1.0 g/10 min, density 0.87 g/cm<sup>3</sup>, octene content 24%) was obtained from DuPont Dow Elastomers (Wilmington, DE). Ammonium polyphosphate (Exolit AP 750) was produced by Clariant International (Gersthofen, Germany). GMA was a product of Hengshui Chemical (Luoyang, China). Chemical-grade dicumyl peroxide (DCP), used as an initiator for the GMA grafting reaction, was a commercial product of the Beijing Xizhong Chemical Factory (Beijing, China).

### Preparation and grafting rate of POE-g-GMA

The fixed quantity of DCP was dissolved thoroughly in GMA and then blended with PP pellets in a high-speed mixing machine. The blended materials were extruded by a WP ZSK-25 corotating twin-screw extruder (Werner & Pfleiderer GmbH, Dinkelsbühl, Germany) with a barrel temperature of 190°C and screw speed of 350 rpm to obtain the GMA grafted POE.

About 2 g grafted product and 150 mL methylbenzene were charged to a vessel with a reflux condenser, refluxed about 2 h at 110°C, then cooled to room temperature. Ethyl acetate was added slowly with agitation. The sediment was filtered out, ethyl acetate was added to the sediment, mixed, and the sediment filtered out again. The washing procedure was repeated twice, after which the sediment was dried in a vacuum at 60°C for 2 h, to remove the GMA monomer that was not grafted to POE.

A 1-g sample of dried sediment was dissolved in 100 mL methylbenzene and a measured quantity of trichloroacetic acid was added, heated to 110°C and

held 1 h, then cooled to room temperature, after which an indicator amount of phenolphthalein was added. A measured amount of NaOH/CH<sub>3</sub>OH was used to titrate the content of GMA in the grafted product, and from this amount the grafting rate could be calculated.

The FTIR spectra of POE and purified POE-g-GMA were recorded by a Magna-IR760 infrared spectrometer (Nicolet Analytical Instruments, Madison, WI).

### Preparation of specimens

PP, APP, and POE-g-GMA were mixed in a high-speed mixing machine, and then the composites were prepared on a WP ZKS-25 corotating twin-screw extruder with a barrel temperature of 200°C and screw speed of 350 rpm. The pelletized PP/APP/POE-g-GMA composites were dried at 80°C for 4 h before injection molding. Specimens for tensile strength test (GB1040-92, 170 × 10 × 4 mm<sup>3</sup>), flexural strength tests (GB9341-2000, 120 × 10 × 4 mm<sup>3</sup>), Charpy impact strength tests (GB1043-93, 100 × 10 × 4 mm<sup>3</sup>), and limiting oxygen index (LOI) tests (ASTM D 2683-70, 150 × 6 × 3 mm<sup>3</sup>) were injection-molded using an HT125 injection-molding machine. The barrel and the nozzle temperatures of the injection-molding machine were 200 and 215°C, respectively. A single-edge 45° V-shape notch (tip radius = 0.25 mm; depth = 2.5 mm) was milled in the specimens for the impact test.

### Experimental measurements

Tensile properties were determined on an LJ-10000N tensile machine (50 mm/min) (Chengde, China). The flexural strength and modulus tests were determined on a CMT6000 electronic universal testing machine (Shengzhen, China). The notched Charpy strength was determined on an XJJ-5 impact-testing machine (Chengde, China). The LOI was measured on an HC-2 oxygen index tester (Nanjing, China).

Thermal degradation of the samples was investigated with a Perkin-Elmer TG-7 thermogravimetric analyzer (Perkin Elmer Cetus Instruments, Norwalk, CT). Thermogravimetric analysis (TGA) was performed at a heating rate of 10°C/min and a nitrogen flow rate of 50 mL/min.

Dynamic mechanical spectra were performed on a Netzsch DMA242 dynamic mechanical analysis device (Netzsch-Gerätebau GmbH, Bavaria, Germany) from -130 to 150°C, at a heating rate of 5°C/min and frequency of 5 Hz.

The melting and crystallization behaviors of the samples were studied by differential scanning calorimetry (DSC) using a TA DSC Q100 (TA Instruments, New Castle, DE). General scan procedures were that samples were heated at 210°C for 3 min to eliminate any thermal history of the material, and then cooled to

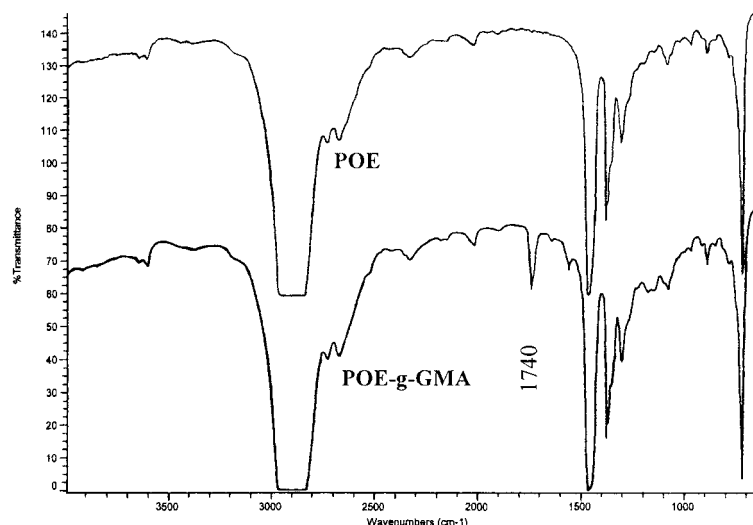


Figure 1 FTIR spectra of POE and POE-g-GMA.

40°C at a rate of 10°C/min with the nonisothermal crystallization process recorded, and held for 2 min at 40°C. The samples were then heated from 40 to 210°C at a heating rate of 10°C/min, and the second heating run was also recorded to describe the melting process. All these procedures were carried out in a dry nitrogen atmosphere.

The fracture morphology of PP/APP and PP/APP/POE-g-GMA composites was observed with a FEI XL-30 scanning electron microscope (SEM; FEI Co., Urbana, IL). The fracture surfaces of the specimens were broken rapidly after refrigeration in liquid nitrogen for 15 min. The fracture surfaces were then gold-coated before observation.

## RESULTS AND DISCUSSION

### Characterization of POE-g-GMA

Under the action of initiator (DCP) and high temperature, the grafting reaction may occur between the POE molecule and the GMA monomer. The FTIR spectra of the purified reaction product can be analyzed to verify whether the grafting reaction occurred

through examining the existence of characteristic functional groups.

Figure 1 shows the FTIR spectra of POE and POE-g-GMA. Different from the FTIR spectra of POE, a new absorption peak appears at the 1740  $\text{cm}^{-1}$  band, which is the characteristic absorption peak of C=O. As described earlier, the grafted product had been purified before the FTIR spectra were recorded; thus the unreacted GMA monomers and reacted autopolymers had been removed, so this C=O peak is produced by POE-g-GMA. We can then conclude that GMA monomers have been grafted onto the POE molecules.

The grafting ratio of POE to GMA, 3.85%, was determined by means of the chemical titration.

### Effect of POE-g-GMA on mechanical properties of composites

The mechanical properties of the POE-g-GMA modified PP/APP composites are presented in Table I. As seen from Table I, the Charpy strength decreased drastically to 3.2  $\text{kJ/m}^2$  when the content of APP is 30%. The toughness of the composites increases after the

TABLE I  
Mechanical Properties of PP/APP/POE-g-GMA Composites

Property	PP/APP/POE-g-GMA						
	100/0/0	96/0/4	70/30/0	68/30/2	66/30/4	64/30/6	62/30/8
Tensile strength (MPa)	34.8	34.5	32.2	29.4	28.1	27.1	26.3
Elongation at break (%)	165.2	192.3	65.6	138.8	140.8	190.8	206.8
Flexible strength (MPa)	42.13	47.68	53.69	46.5	45.2	42.6	39.9
Flexible modulus (GPa)	1.64	1.93	2.97	2.53	2.43	2.24	2.14
Charpy strength ( $\text{kJ/m}^2$ )	4.7	5.5	3.2	4.6	4.9	5.4	6.4

TABLE II  
Limiting Oxygen Index (LOI) of PP/APP/POE-g-GMA Composites

LOI (%)	PP/APP/POE-g-GMA						
	100/0/0	96/0/4	70/30/0	68/30/2	64/30/4	64/30/6	62/30/8
	19.0	19.0	36.0	36.5	37.0	37.5	37.5

addition of POE-g-GMA. The greater the content of added POE-g-GMA, the greater the increase of the toughness of composites. The Charpy strength is 4.9 kJ/m<sup>2</sup> when the content of POE-g-GMA is 4%, which is even higher than that of the pure PP resin.

Generally, the increase of toughness is always accompanied by the loss of tensile strength, flexible strength, and modulus of elastomer particle toughened PP blends. Clearly corresponding to the change of POE-g-GMA content, this loss is listed in Table I. The tensile strength, flexible strength, and modulus decrease gradually. However, even when the content of POE-g-GMA reaches 8%, the composites still retain good mechanical properties and have practical utility.

#### Effect of POE-g-GMA on flame retardance of the composites

The effect of POE-g-GMA on the flame retardance of the APP-filled PP composites is shown in Table II. Although the addition of POE-g-GMA has no influence on the LOI of pure PP, POE-g-GMA can increase the LOI of the PP/APP composites. When the content of APP is fixed in the composites, while the content of POE-g-GMA is changed, we can see that the LOI increases with the increased content. The LOI of the composites reaches 37.5% when the content of POE-g-GMA is 6%; it is 1.5% higher than that of the composites having no POE-g-GMA. It is clear that the addition of POE-g-GMA can improve the flame retardance of the PP/APP composites, which may be attributed to the better dispersion of the APP particles in the PP matrix. With the improvement of the flame-retardant efficiency of the APP, the flame retardance of the composites increases. Moreover, the interfacial interaction between APP and PP, which is enhanced with the addition of POE-g-GMA, may contribute to the increase of the LOI of the composites.

#### Thermogravimetric analysis

The PP composites filled with flame retardant APP can promote the carbonification during the process of combustion or thermal degradation, so the residual mass of the thermal decomposition increases.<sup>12,13</sup> The TG behavior reflects the carbonification of the flame-retardant materials in the process of heating under nitrogen. It can be seen from the TG results shown in

Figure 2 that the pure PP begins to lose weight at 340°C, loses 98% weight at 450°C, and decomposes almost completely at 470°C. There is no residual char formed in the process. When the PP/APP composite (30% APP content) is heated from room temperature, at a heating rate of 10°C/min, it begins to lose weight at 250°C (the beginning decomposition temperature of APP is 250°C), the residual mass is 18.4% at 500°C, and changes little until the temperature reaches 700°C. This means that the carbonification has taken place on the PP of the composite during the heating process and some char has formed.

Under the condition of the same flame-retardant APP content, POE-g-GMA was added to the composites. The TG curve is shown in Figure 2(c), from which it can be observed that the beginning and ending temperatures of the TG curve are identical to those of Figure 2(b), where no POE-g-GMA was added to the composites. However, there is some difference that the TG curve becomes gradual during the range of the main decomposition process. This indicates that the addition of the grafted material can help the APP fully exert its flame-retardant property more uniformly during the decomposition temperature region of PP and the flame-retardant efficiency of APP is improved. Furthermore, the residual mass of the composite improved by 1.2%. This shows clearly that the addition of POE-g-GMA promoted the carbonization and char formation of the composites during the heating process. The improvement of the mass of char formation is favorable to the flame resistance of the PP composites.

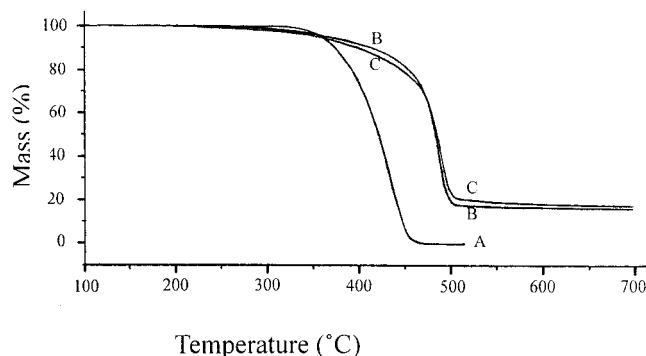
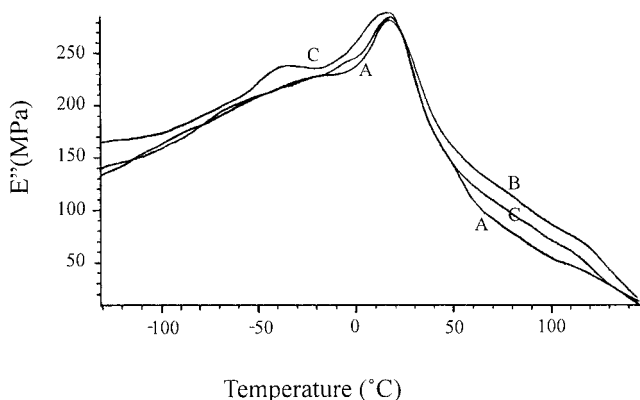


Figure 2 TG curves of the PP/APP/POE-g-GMA composites at various ratios: (A) 100/0/0; (B) 70/30/0; (C) 66/30/4.



**Figure 3** Temperature dependency of  $E''$  for PP/APP/POE-g-GMA composites at various ratios: (A) 100/0/0; (B) 70/30/0; (C) 66/30/4.

#### Effect of POE-g-GMA on DMA of the composites

Figure 3 displays the loss modulus ( $E''$ ) data of PP and PP composites versus temperature. The toughness of a polymer material is determined by the capacity of the material to dissipate the impact energy. The higher the material's  $E''$ , the better its toughness, and the  $E''$  value at low temperature is more important in determining toughness.<sup>14</sup> It can be seen from Figure 3 that there is no peak of  $E''$  in the PP and PP/APP composites at low temperature, and a new peak of  $E''$  ( $-34.8^\circ\text{C}$ ,  $E'' = 236.9$  MPa) has been created after the POE-g-GMA was added. This indicates that the compatibilizer improves the toughness of the PP composites.

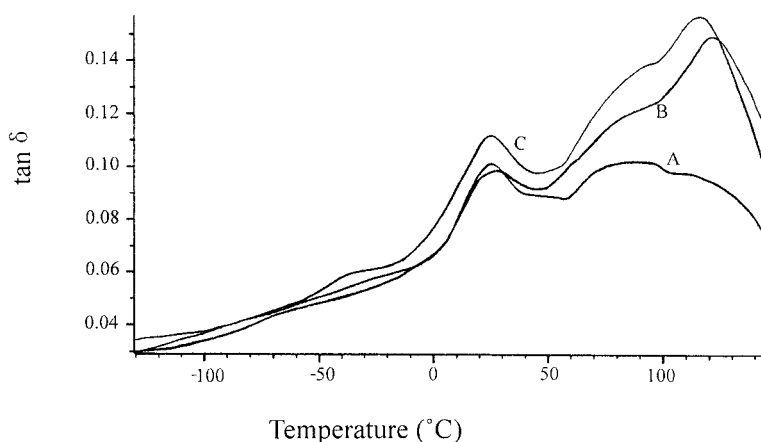
In addition, the results of Figure 3 show that the peak temperature of  $E''$  at room temperature was decreased about  $1.4^\circ\text{C}$  after 4% POE-g-GMA was added to the PP/APP composites. If the APP particle has a local motion-resistant effect on the elastic POE-g-GMA molecule dispersed in the composites, then the entan-

glement potency of the POE-g-GMA molecule in the blocked domain may be weakened, thus leading to a shortened relaxation time for this chain segment of the molecule, which can achieve transition from frozen to free at lower temperature. Therefore, this result may be the direct reason that the PP/APP composites have been toughened by POE-g-GMA.

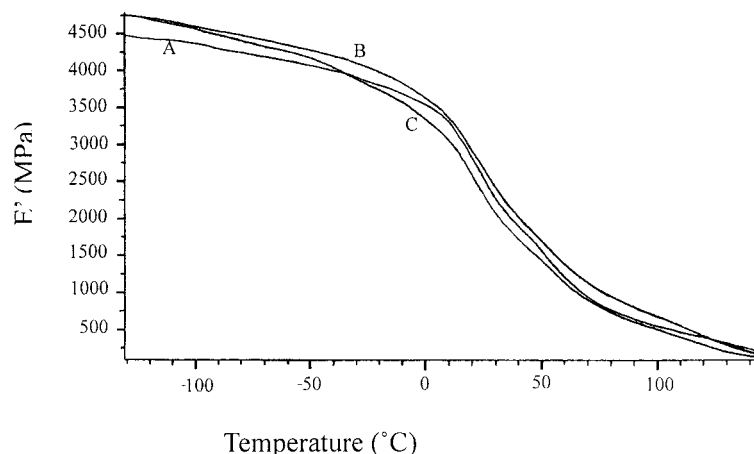
All polymer material has viscoelasticity. The strain lags the cyclical sinusoidal stress exerted in the DMA experiment. The inner loss ( $\tan \delta$ ) is expressed by the proportion of  $E''$  and storage modulus ( $E'$ ), that is,  $\tan \delta = E''/E'$ .<sup>14</sup> The temperature dependency of  $\tan \delta$  for PP/APP/POE-g-GMA composites is shown in Figure 4.

Figure 4 shows that the  $\tan \delta$  peak of PP in the  $\tan \delta - T$  DMA curve appears at room temperature. The intensity and location of the peak can be used to evaluate the toughness of polymer composites. Generally, the higher the intensity and the lower the temperature of the  $\tan \delta$  peak, the better the toughness of the composites. It can be observed that the  $\tan \delta$  peak of PP is at  $24.6^\circ\text{C}$  and the value of  $\tan \delta$  is 0.101; the  $\tan \delta$  peak of PP/APP composites is at  $26.4^\circ\text{C}$  and  $\tan \delta = 0.099$ ; the  $\tan \delta$  peak of PP/APP/POE-g-GMA is at  $24.4^\circ\text{C}$  and  $\tan \delta = 0.112$ ; these values indicate that the toughness of PP is deteriorated by APP. After adding 4% POE-g-GMA to the PP/APP composites, the  $\tan \delta$  peak shifts to lower temperature and the intensity of the peak is also increased, corresponding to the significant improvement of the toughness of the material.

From the perspective of molecular motion, the improvement of the material's toughness is attributed to the fact that the mobile building block of the macromolecule can create deformation and absorb energy when the polymer is subjected to external force.<sup>15</sup> Under the condition of service, although the backbone movement of the PP macromolecule is frozen, some of



**Figure 4** Temperature dependency of  $\tan \delta$  for PP/APP/POE-g-GMA composites at various ratios: (A) 100/0/0; (B) 70/30/0; (C) 66/30/4.



**Figure 5** Temperature dependency of  $E'$  for PP/APP/POE-g-GMA composites at various ratios: (A) 100/0/0; (B) 70/30/0; (C) 66/30/4.

the units (such as shorter segments, lateral groups, and atomic groups, etc.) have the ability to move. So the energy can be absorbed by the deformation (which is much greater than the glass transition) created under external force. This secondary transition is very important to the toughness of polymer material. Generally speaking, the macromolecule having no secondary transition at low temperature is relatively brittle. If there is a larger secondary transition at a temperature lower than room temperature, the polymer has good toughness. Thus, the  $\tan \delta$  peak located at  $-36^\circ\text{C}$  is an important factor that is favorable to the toughness.

Figure 5 shows the dynamic mechanical storage modulus ( $E'$ )- $T$  spectra of PP and PP composites.  $E'$  can describe the stiffness of material. From the curves we can see that the  $E'$  of PP/APP composites is increased by the addition of APP, and APP particles play the role of physical crosslinking points in the system. After POE-g-GMA is added in, the  $E'$  of the composites changes little at low temperature ( $< -105^\circ\text{C}$ ), but the  $E'$  clearly decreases with the temperature increased after  $-105^\circ\text{C}$ ; the  $E'$  is even lower than the  $E'$  of pure PP after  $-35^\circ\text{C}$ . Thus we should not add too much POE-g-GMA to the composites to keep the stiffness within a useful range.

### Crystallization and melting behavior

According to the proposed theory, the improvement of the compatibility between polymer and filler can reduce the free energy of a nucleation process, which is favorable to the nucleation process at the phase boundary.<sup>16</sup> An abundance of experimental results supports this theory in polymer systems. For example, Velasco et al.<sup>17</sup> found that the nucleation action of talc on PP improved after the talc was modified by silicone hydride. The study by Liu and Gilbert<sup>18</sup> indicated that

the addition of talc modified by phosphoric ester distinctly increased the crystallization temperature and crystallinity. At the same time, there are many experiments that showed that the filler's nucleation action was reduced after surface modification, especially when modified by a coupling agent having a long fatty chain. Mitsubishi et al.<sup>19</sup> modified  $\text{CaCO}_3$  with a series of phosphoric esters formed at the surface of  $\text{CaCO}_3$ , which provided a coating that diluted the PP molecule chain near the  $\text{CaCO}_3$  particle, so that the nucleation action decreased.

The crystallization temperature  $T_c$ , the width at half-height of peak  $B_{1/2h}$ , the peak temperature of the crystallization  $T_{cp}$ , the enthalpy of crystallization  $\Delta H_c$ , and the crystallinity  $X_c$  are listed in Table III, from which it may be observed that the addition of POE-g-GMA into the PP matrix reduced the  $T_c$  and widened the  $B_{1/2h}$ . The  $B_{1/2h}$  is a parameter associated with the rate of crystallization, so that when the  $B_{1/2h}$  becomes narrower, the speed of crystallization becomes faster. The main chain of POE-g-GMA entwines the molecule of PP, which may hinder the rate of crystallization, and  $X_c$ ,  $T_{cp}$  and  $X_c$  are clearly increased by APP. The  $B_{1/2h}$  decreases from 6.69 to 4.26°C, so the rate of crystallization is also distinctly improved. These results make clear that APP takes part in the role of nucleation agent for PP, so it can induce the PP molecule to crystallize at higher temperature. It can be conjectured that the dimensions of the PP crystal become smaller, with the same degree of crystallinity, with the increasing number of crystal nuclei after the APP is added to the PP matrix.

Although the  $T_c$  and  $T_{cp}$  are both slightly decreased, the  $B_{1/2h}$  is reduced when POE-g-GMA is added to the PP/APP composites, and the peak becomes narrower when the content of POE-g-GMA increases, which is different from the PP/POE-g-GMA blend. The main

TABLE III  
DSC Results of PP/APP/POE-g-GMA Composites by Crystallization (Cooling)<sup>a</sup>

DSC Parameter	PP 100	PP/POE-g-GMA 96/4	PP/APP 70/30	PP/APP/POE-g-GMA	
				66/30/4	62/30/8
$T_c$ (°C)	118.85	118.50	119.27	118.88	118.71
$B_{1/2h}$ (°C)	6.69	6.92	4.26	3.89	3.85
$T_{cp}$ (°C)	111.88	110.18	122.91	122.11	121.89
$\Delta H_c$ (J/g)	-103.33	-101.37	-110.74	-108.62	-107.72
$X_c$ (%)	49.88	48.93	53.46	52.43	52.00

<sup>a</sup> The enthalpy of 100% crystallized PP is 207.15 J/g.<sup>20</sup>

reason for this phenomenon is that the compatibility between PP and APP has been enhanced because of the action of POE-g-GMA. POE-g-GMA is favorable to the contact of the APP particles with the PP matrix and activates the nucleation points on the surface of APP. The rate of crystallization increases with the increasing number of APP particles, which can have the function of forming crystals induced by the nucleation process at the phase boundary. Although the long flexible chain of POE can lower the  $T_c$  of PP, it does so only very slightly.

The melting DSC data of the PP/APP/POE-g-GMA composites are shown in Table IV. It is observed that the effect of POE-g-GMA on the melting temperature ( $T_m$ ) and the peak temperature of the melting ( $T_{mp}$ ) of the pure PP and APP filled PP is very small. Even though POE-g-GMA is added in, the melting point of the blend remains almost the same, such that the heat resistance is not deteriorated. This addition also has scant influence on the rate of melting.

### SEM analysis

The mechanical properties, especially the toughness of the filled polymer system, are influenced by the shape, dimension, and dimension distribution of the filler particles, the filler content, and the physical properties of the filler and polymer, for example.<sup>21,22</sup> Without doubt, the interfacial interaction between the polymer and the filler is one of the most important factors. The interfacial interaction between the polymer and filler not only can change the local deformation and micro-mechanism of the local deformation and the breaking

process, but also can influence the crystalline behavior of the polymer, both of which influence the mechanical properties of the composites.

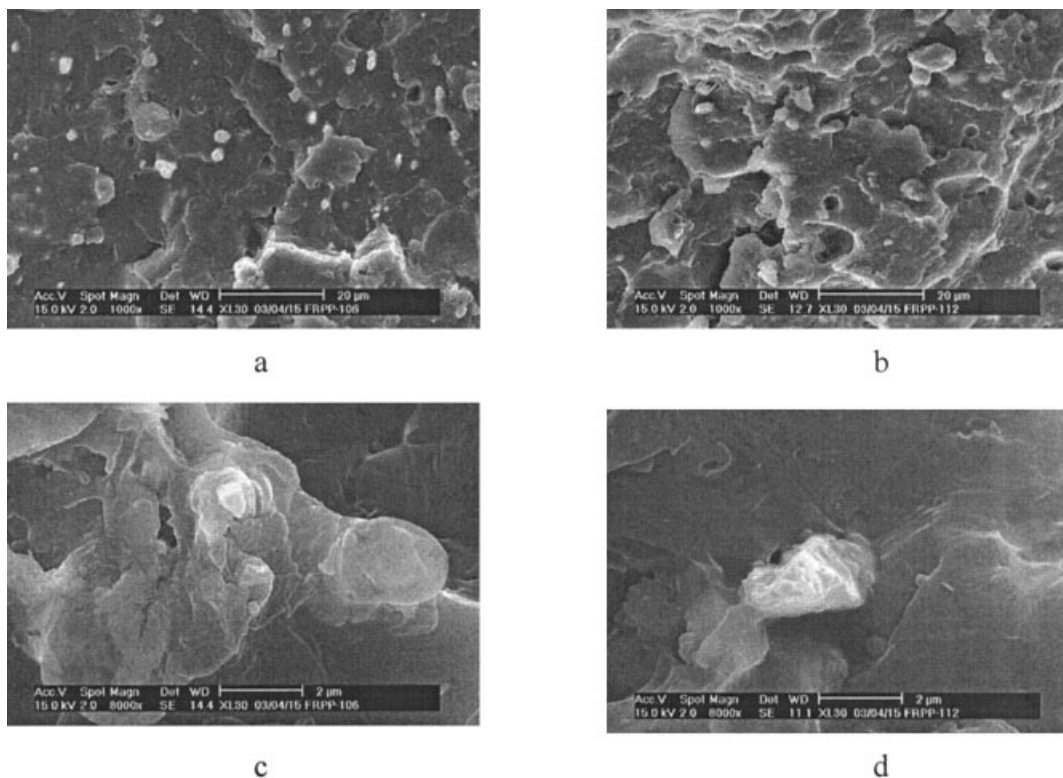
Figure 6 shows SEM micrographs of unmodified PP/APP and POE-g-GMA modified PP/APP composites. There is no POE-g-GMA in Figure 6(a) and (b). As seen from the SEM micrographs, the PP and APP are blended by simple extrusion, the interface between the two phases is neat and clear, and the acting force of the interface is weak. In some domains, larger particles of agglomerated APP filler may be observed; these larger particles can lead to a deterioration of toughness. After POE-g-GMA is added to the composites, as shown in Figure 6(c) and (d), APP particles are dispersed more uniformly, the interface becomes nebulous, and this means that POE-g-GMA plays an important role of compatibilizer. The APP particles are covered first by a layer of POE-g-GMA and then blend with the PP matrix. The interfacial adhesion between PP and APP is improved as a result of the interfacial interaction of POE-g-GMA in the composites. As a result, the toughness of the composites is improved.

### CONCLUSIONS

POE-g-GMA is a true compatibilizer for APP-filled PP flame-retardant composites and can improve the toughness of the PP/APP composites. Dynamic mechanical analysis revealed that the addition of POE-g-GMA not only improved the peak of  $E''$  and  $\tan \delta$  at room temperature, but also created new peaks of  $E''$  and  $\tan \delta$  in the low-temperature region; both of these phenomena are related to the improvement of the

TABLE IV  
DSC Results of PP/APP/POE-g-GMA Composites by Melt (Second Heating)

DSC Parameter	PP 100	PP/POE-g-GMA 96/4	PP/APP 70/30	PP/APP/POE-g-GMA	
				66/30/4	62/30/8
$T_m$ (°C)	154.72	154.35	157.39	157.20	157.14
$T_{mp}$ (°C)	161.79	161.38	161.72	161.58	161.45
$B_{1/2h}$ (°C)	8.53	8.49	5.32	5.47	5.48



**Figure 6** SEM micrographs of the PP/APP and PP/APP/POE-g-GMA composites at various magnifications: (a) PP/APP ( $\times 1000$ ); (b) PP/APP/POE-g-GMA ( $\times 1000$ ); (c) PP/APP ( $\times 8000$ ); (d) PP/APP/POE-g-GMA ( $\times 8000$ ).

toughness of the composites. The presence of POE-g-GMA appears to have improved the filler dispersion and the interfacial interaction between APP particles and the PP matrix. It can also influence the thermodegradation of PP and improve the percentage of char formation. Thus the flame retardance of the composites is improved at the same APP content. POE-g-GMA can improve the crystallization rate and decrease the initial crystallization temperature and the crystallinity, but it has little effect on the melting behavior of PP.

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