

# Effect of Nylon 6 on Fracture Behavior and Morphology of Tough Blends of Poly(2, 6-dimethyl-1,4-phenylene oxide) and Maleated Styrene-Ethylene-Butadiene-Styrene Block Copolymer

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**ABSTRACT:** Ternary blends of poly(2,6-dimethyl-1,4-phenylene oxide) (PPO), nylon 6, and styrene-ethylene-butadiene-styrene block copolymer grafted with maleic anhydride (SEBS-g-MA) were prepared via a melt extrusion, and the fracture behavior, morphology, mechanical properties, and rheology were studied. The compatibilization of the blended components was confirmed by differential scanning calorimetry (DSC) analysis. Mechanical properties evaluation demonstrated that incorporation of nylon 6 resulted in an improvement of the tensile strength, but reduction of both the notched Izod impact strength and elongation at break. Transmission electron microscopy (TEM) observation revealed that the network structure of SEBS-g-MA domain

was gradually destroyed by incorporating the nylon 6. A conversion of SEBS-g-MA domain from the network to the irregular dispersed phase took place when the nylon 6 content reached 20 wt %, which resulted in a reduction of the impact strength. Fracture morphology implied that increase of the tensile strength was caused by the plastic deformation of matrix. Rheology investigation indicated that the melt viscosities could be reduced significantly with increasing the content of nylon 6; thus, the processability was improved. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 99: 3336–3343, 2006

**Key words:** PPO; nylon 6; SEBS-g-MA; blends; fracture behavior; morphology

## INTRODUCTION

Blending of polymers is the most widely used method to obtain new polymeric materials. It is also considered as an efficient way of upgrading the properties.<sup>1</sup> In recent decades, polymer blends are receiving increasing attention from both the scientific and industrial communities, as they offer an attractive low-cost substitute to the development of entirely new macromolecules. Most commercial multicomponent polymer systems are two-phase blends that display advantages over the single-phase systems.<sup>2</sup> There is a general agreement that the properties of polymer blends are usually controlled by the properties of the components, morphology of the blends, and interaction between components in the blends.<sup>3–5</sup> The morphology of polymer blends is intimately correlated with the properties of the blending system. The domain size is often used to indicate the extent of compatibility of multiphase polymer systems, i.e., the smaller the domain size, the more compatible are the systems and the better are the mechanical properties.<sup>6–9</sup> The de-

tailed investigation on phase morphological development, phase continuity, and phase stability of polymer blends has been undertaken. The correlation between morphology and mechanical properties has been established.<sup>10,11</sup> Polymer toughening has been a major goal of the research of the polymer blending. Blending with suitable elastomeric materials has been considered as one of the most effective methods to improve the toughness of a brittle polymer.<sup>12</sup> The toughening of a rubber-toughened polymer is strongly influenced by the amount, the particle size, and properties of the elastomers, as well as the interactions between phases determining the mechanical properties of the toughened plastics.<sup>4,10,11,13</sup>

Poly(2,6-dimethyl-1,4-phenylene oxide) (PPO), one of the most attractive engineering materials, displays a high strength, excellent heat resistance, high glass-transition temperature ( $T_g$ ) of 210°C, and good dimensional stability. However, deficiencies, such as natural brittleness, extremely bad melt-processability, and poor solvent resistance, prohibit its broader application.<sup>14,15</sup> Blends of polyamide 6 (PA-6) and poly(phenylene oxide) (PPO) are of interest as a high-performance alloy, and has attracted great interest from both industry and academic.<sup>16,17</sup> PA-6 has a number of interesting properties such as a good melt processabil-

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ity and an excellent solvent resistance. However, its heat distortion temperature is low, and it has a poor dimensional stability and low impact strength. The inherent properties of PPO and nylon 6 suggest that a combination of PPO and nylon 6 should provide a useful polymer alloy with the balanced properties and the advantages from individual components, since the advantages of one component can compensate for the deficiencies of the other.<sup>18,19</sup> As an engineering thermoplastic alloy with high performance, the PPO/nylon 6 blend exhibits excellent chemical resistance, high-dimensional stability, good thermal properties, and good melt-processability, and has found wide application in the fields of mechanics, electronic applications, and automobile package materials and body panels. However, PPO is incompatible with nylon 6, and the blend of PPO and nylon 6 generally shows the deterioration in impact and tensile strength.<sup>20</sup> Such a reduction in properties is frequently a reflection of poor interfacial adhesion between dispersed and continuous matrix that leads to rapid initiation and growth of a crack.<sup>21,22</sup> Thus, it is reasonable to expect that some form of compatibilization would be required to prepare a useful alloy. A large number of studies employed maleic anhydride (MA) functionalized copolymers as compatibilizers for the alloy of PPO and nylon 6.<sup>23,24</sup> Brown reported that the reaction of an aryloxytriazine-functionalized PPO with nylon 6 resulted in a copolymer, which was used as an *in situ* reactive compatibilizer for the PPO/nylon 6 alloys.<sup>25</sup> Jo and Kim reported that the styrene-acrylic acid random copolymer with a high acrylic acid content was an effective interfacial linker for PPO/nylon 6 blends.<sup>26</sup> Lai synthesized a PPO-nylon 6 grafted copolymer with well-defined structure, and used this copolymer as the compatibilizer for PPO/nylon 6 blends, which showed satisfactory mechanical properties, except tensile strength.<sup>18</sup> Wang et al. prepared PPO/nylon 6 blends compatibilized with a poly(ethylene-1-octene) grafted with MA (POE-g-MA) and obtained improved toughness.<sup>27</sup> PPO grafted with MA (PPO-g-MA) was also reported as a good reactive compatibilizer for PPO/nylon 6 alloys.<sup>28–32</sup> A PPO/nylon 6 blend at a weight ratio of 70/30 provided notched impact strength of only 25 J/m.<sup>33</sup> However, when half of the PPO was replaced by PPO-g-MA, 49 J/m was achieved.<sup>34</sup> Although improvement was moderate, this method provided a basis for further toughening.

In our former study,<sup>35</sup> the styrene-ethylene-butadiene-styrene block copolymer grafted with MA (SEBS-g-MA) was found to be an excellent impact modifier for PPO, and the PPO/SEBS-g-MA blend (80/15, wt/wt) could achieve a high toughness with impact strength of 950 J/m. In that blending system, the SEBS-g-MA is discovered to provide an optimum toughening for PPO by a bicontinuous morphology,

where the SEBS-g-MA forms a network in PPO matrix. The goal of the present work is to attempt to improve the processability by incorporating nylon 6 into the tough PPO/SEBS-g-MA blend. For this purpose, the ternary PPO/SEBS-g-MA/nylon 6 blends were prepared via a melt extrusion. The effect of nylon 6 on fracture behavior and morphology was studied. The mechanical and rheological properties were also investigated, and the correlation between the mechanical properties and morphology was established.

## EXPERIMENTAL

### Materials

PPO (646–111 grade) was purchased from GE Plastics Company, USA, which had an intrinsic viscosity of 0.43 dL/g measured in chloroform at 25°C. Nylon 6 (1013B) with a number-average molecular weight of 25,000 was supplied by UCB Chemical Co., Japan. SEBS-g-MA (Kraton FG 1901) was obtained commercially from Shell Chemical Company, USA, which contained 29 wt % of styrene, and had a number-average molecular weight of 7500 in the polystyrene (PS) blocks and 37,500 in the polyolefin blocks. The weight percent of MA grafted onto this elastomer was 1.7–2.0 wt %.

### Preparation of blends

The pellets of all materials were dried in a vacuum oven at 90°C overnight before using. The constituents of the blends with various weight ratios were blended using a corotating and intermeshing ZSK-25 twin-screw extruder (Werner and Pfleiderer Corp.) with a screw configuration adapted to PPO. All the ingredients were tumble-mixed and fed through the throat of an extruder. The temperatures along the barrel were set from 240 to 260°C, and the rotation speed of the screw was 180 rpm. The extruder was equipped with a high efficient vacuum venting to remove nonreacted species and reaction by-products. The samples were passed through a cooling water bath, and the pelletizing was subsequently carried out. The extrusion parameters were changed very little from one composition to another. The blends were further dried at 100°C before injection-molding the test specimens for the determination of mechanical properties.

### Measurements of mechanical properties

The notched impact and tensile test bars were prepared via injection-molding using a reciprocating screw injection machine (CJ150NC-2); the barrel temperature was 270°C, and the mold temperature was 90°C. The tensile properties were determined with an Instron-1185 universal testing machine, using a 5000

Newton load transducer, according to the standard of ASTM D-638. Notched Izod impact strength was measured with a SUMITOMO impact tester according to ASTM D-256. The thickness of Izod impact specimens was 1/8 in., and impact energy was 4 J. All the experiments were at room temperature and five measurements were carried out for each data point.

### Thermal analysis

Thermal analysis was carried out using a Perkin-Elmer Pyris-1 differential scanning calorimetry (DSC) apparatus. All measurements were made under a nitrogen gas atmosphere at heating rate of 10°C/min on samples weighing about 10 mg. Before the formal measurements, the first heating scan was carried out from room temperature to 250°C at heating rate of 10°C/min, and the samples were held at this temperature for 3 min to diminish the thermal and processing history effects, and then were quenched. The temperature ranges used for measuring the values of melt point ( $T_m$ ) and heat fusion ( $\Delta H_m$ ) were normally from 70 to 250°C.

### Scanning electron microscopy

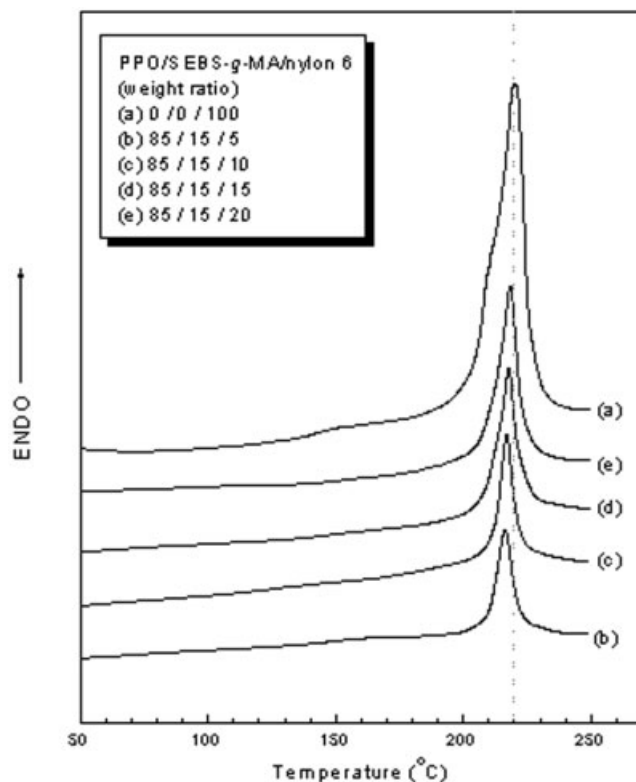
Scanning electron microscope (Cambridge S250) was employed to observe and record the fracture surface of the blending samples. The fractured specimen surfaces from the tested tensile bars were coated with a thin layer (10–20 nm) of gold–palladium. The coating was carried out by placing the specimen in a high vacuum evaporator and vaporizing the metal held in a heated tungsten basket. Scanning electron microscopy (SEM) measurements were performed only on the fractured surface of all the blends.

### Transmission electron microscopy

Morphology was determined by transmission electron microscopy (TEM). Samples were cryogenically microtomed from Izod impact bars perpendicular to the flow direction using a diamond knife. PPO/nylon 6/SEBS-*g*-MA system was stained with osmium tetroxide ( $\text{OsO}_4$ ). A Hitachi H-800 transmission electron microscope was used to view and photograph these samples, which are only of to order of 100–150 nm in thickness.

### Rheology measurements

The apparent viscosities at various shear rates were determined using a Capirograph-1B capillary rheometer (Toyoseiki Manufacture Company) with a capillary diameter of 1.262 mm and an L/D ratio of 50.51. Since the L/D ratio of the capillary die used in this study is large than 40, the errors between the tested



**Figure 1** DSC heating traces of nylon 6 and its blends with PPO and SEBS-*g*-MA.

shear rates and the real Newtonian shear rates can be negligible. The measurement was carried out at 270°C under the shear rates ranging from 1 to 10<sup>4</sup> 1/s.

## RESULTS AND DISCUSSION

### Thermal behavior

Blends of PPO and SEBS-*g*-MA were partially compatible blending systems, as the PS blocks in SEBS-*g*-MA chains are demonstrated to directly interact with PPO. However, when nylon 6 is incorporated into PPO/SEBS-*g*-MA blend, the incompatibility may arise due to the thermodynamic immiscibility of PPO and nylon 6. The thermal behaviors of the nylon 6 and PPO/SEBS-*g*-MA/nylon 6 ternary blends were investigated by DSC. Figure 1 shows DSC thermograms of these samples, and the  $T_m$  and the heat fusion ( $\Delta H_m$ ) were obtained from DSC analysis, which were corresponding to the nylon 6 phase in blends, and summarized in Table I. Pure nylon 6 displays a melting peak at about 220°C and a shoulder at 212°C, which is referred to the  $\beta$ -form crystals of the nylon 6. The thermal behaviors of the PPO/SEBS-*g*-MA/nylon 6 ternary blends are clearly affected by the presence of the SEBS-*g*-MA. One may observe a reduction of the  $T_m$  for the ternary blends containing 5 wt % of nylon 6, which implied a compatibility of the nylon 6 with the other compo-

**TABLE 1**  
The Thermal Properties of the Nylon 6 and Its Blended Samples

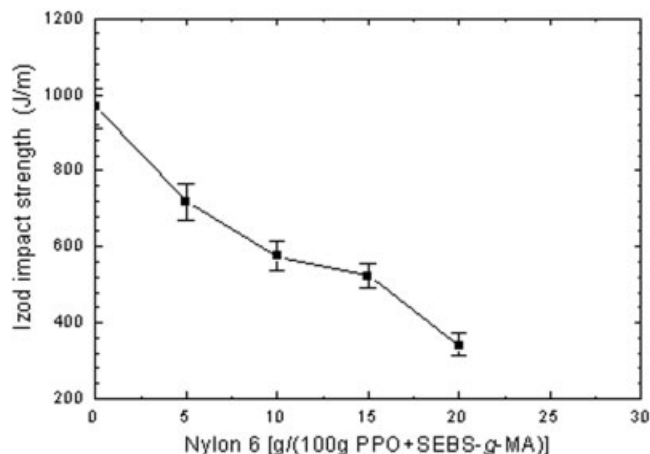
Sample (weight ratio) PPO/SEBS-g-MA/nylon 6	$T_m$ (°C)	$\Delta H_m^a$ (J/g)
0/0/100	220.18	68.32
85/15/5	216.35	62.76
85/15/10	217.94	64.12
85/15/15	218.28	65.47
85/15/20	218.63	65.86

<sup>a</sup> Values normalized to the amount of the nylon 6 phase.

nents. The values of  $\Delta H_m$  for the ternary blends were normalized to the amount of the nylon 6 phases under consideration of the concentration of the PPO and the SEBS-g-MA. It was found that the  $\Delta H_m$  decreased significantly. In fact, the temperatures and the enthalpies associated with the transition of the phase of the nylon 6 are lowered with respect to pure nylon 6 in all blending ranges. The decrement in  $T_m$  and  $\Delta H_m$  strongly depends on the relative concentration of the SEBS-g-MA to nylon 6. The higher the relative concentration of the SEBS-g-MA, the greater is the decrement. Above-mentioned results are indicative of interaction between the phases of the nylon 6 and the SEBS-g-MA. It is understandable that the grafted MA moieties on SEBS may react with the amine moieties on nylon 6, and thus generates bridges between the two polymers as shown in Figure 2. This correlated interfacial reaction between nylon 6 and MA-functionalized copolymer have been widely reported, and will be verified in following discussion regarding rheological properties.<sup>36–38</sup> The mentioned interfacial reaction also disturbs the crystallization of the nylon 6 and results in a decrease of its crystallinity.

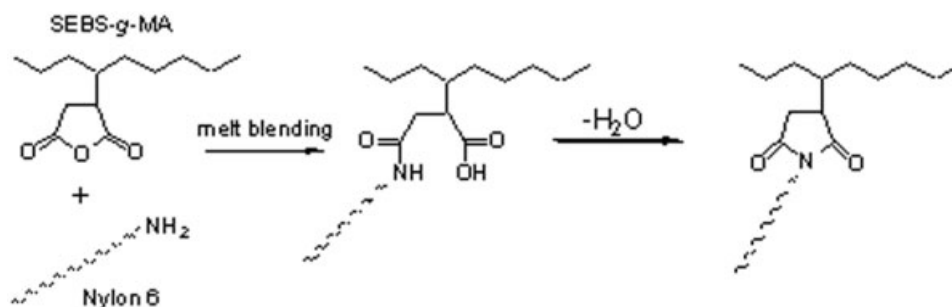
### Tensile and impact properties

Notched Izod impact strength, tensile strength, and elongation at break of the PPO/SEBS-g-MA/nylon 6 blends as a function of the nylon 6 content are presented in Figures 3 and 4. The binary blend of PPO/

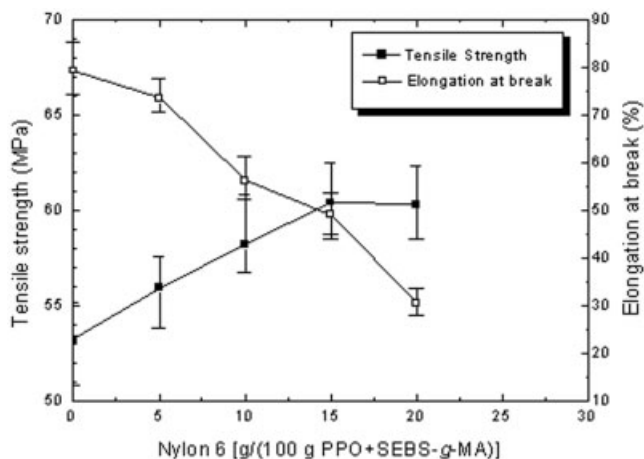


**Figure 3** Effect of the nylon 6 content on the notched Izod impact strength of PPO/SEBS-g-MA (weight ratio: 85/15) blends.

SEBS-g-MA at weight ratio of 85/15 exhibits a super toughness with a notched Izod impact strength of 969 J/m by comparison of the value (76 J/m) of pure PPO. However, its tensile strength drops slightly to 68 MPa and elongation at break increases to 92% from the values (74 MPa and 83%) of the pure PPO, respectively. When nylon 6 as a processability modifier was incorporated into the tough PPO/SEBS-g-MA blends, the tensile strength increased because of the increase of the plastic phases and the compatibility of the ternary blending systems. But, the introduction of nylon 6 results in a reduction of the impact strength and elongation at break. The higher the content of nylon 6, the greater is the tensile strength. The contrary results are observed for the variation of the impact strength and elongation at break. Both the increment in tensile strength and decrement in impact strength and elongation at break strongly depend on the concentration of the nylon 6. As a highly crystallized polymer, incorporation of nylon 6 causes an increase of the concentration of thermoplastic materials in the ternary blends, and, consequently, a reduction of impact strength and elongation at break even if the PPO/SEBS-g-MA/nylon 6 blend is a compatibilizing sys-



**Figure 2** Scheme of reactive compatibilization of SEBS-g-MA and nylon 6 during melt extrusion.



**Figure 4** Effect of the nylon 6 content on the tensile properties of PPO/SEBS-g-MA (weight ratio: 85/15) blends.

tem. Moreover, the crystallinity of nylon 6 phases in the matrix may also affect the toughness of blends, and lower crystallinity will favor a higher toughness. As discussed in last section, the  $\Delta H_m$  data for the nylon 6 phases revealed that the relative crystallinity of nylon 6 phases decreased with an increase of nylon 6. However, the effect of the decreasing crystallinity could not compensate the deterioration of toughness by incorporating the increasing nylon 6 phases with the crystallized zone. As a result, the dependence of impact strength of the blends on the nylon 6 content is negative. In general, a balance in mechanical properties as well as processability is necessary for high-performance blending materials. Although the incorporation of nylon 6 deteriorates the impact strength and elongation at break, the mechanical properties of the ternary blend compositions still indicate a favorable combination of toughness, strength, and rigidity.

### Morphology

To examine the influence of morphology on the mechanical properties, TEM studies on the specimens were carried out. The TEM image of PPO/SEBS-g-MA/nylon 6 blends, as shown in Figure 5, could be identified with the aid of the  $\text{RuO}_4$  staining, because the SEBS-g-MA component in the blends could be stained with  $\text{OsO}_4$  due to the double bond in SEBS molecular chain.

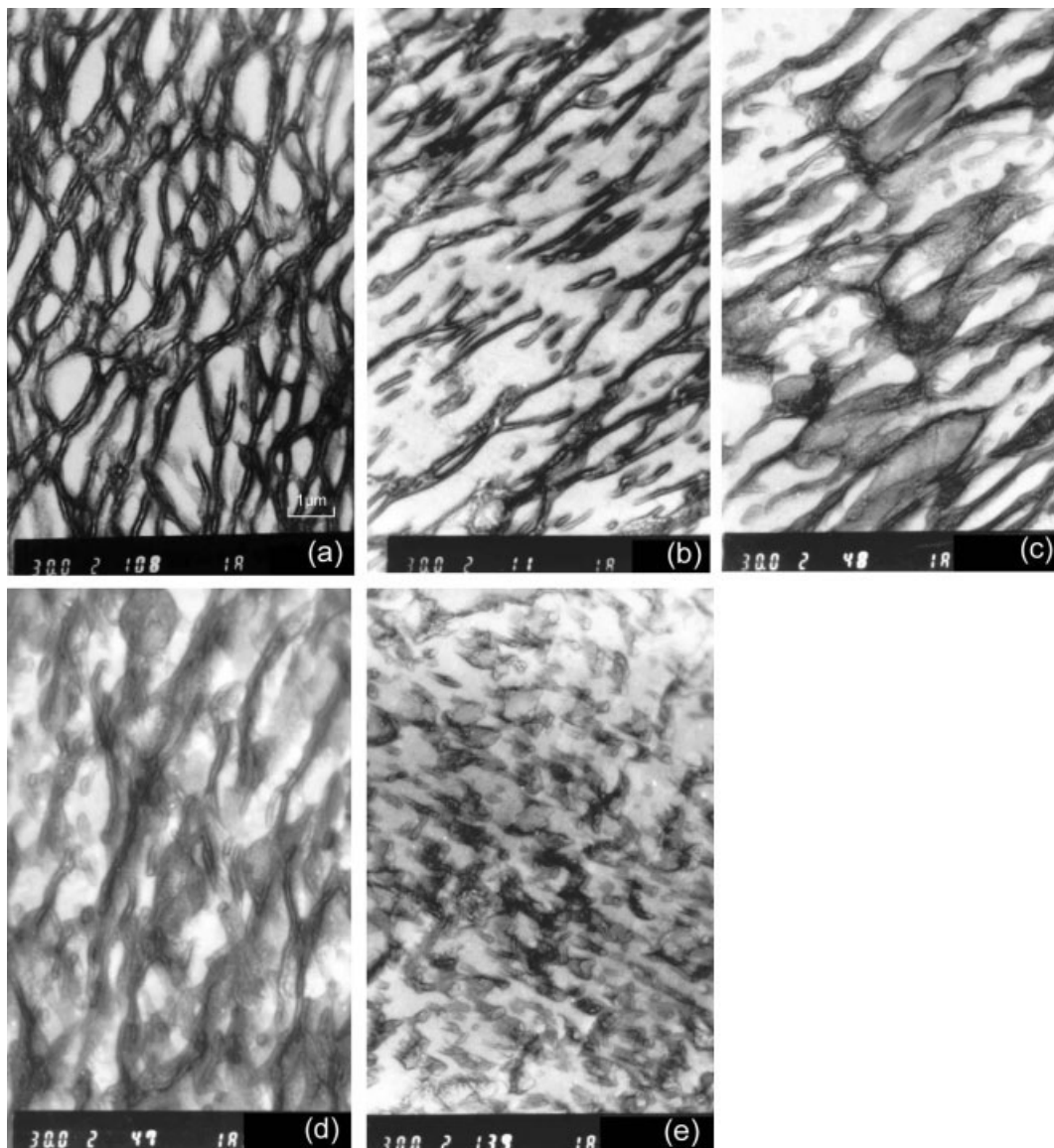
Figure 5(a) clearly displays a bicontinuous-phase morphology, in which SEBS-g-MA forms a network domain in PPO matrix. This bicontinuous structure was caused by aggregation of SEBS segments. The network of SEBS-g-MA domain was considered to be more effective than the individual particles in stopping the growth of the crazing and shear yields, which was attributable to the toughening of PPO matrix.<sup>36</sup>

From Figures 5(b)–5(e), it was observed that the perfection of the network structure was gradually destroyed with increasing the nylon 6 content, which indicated that the incorporation of nylon 6 into PPO/SEBS-g-MA blend generated a significant effect on the morphology. When the content of nylon 6 reached 20 wt %, the network representing SEBS-g-MA domain was completely broken and irregular dispersed phases were presented instead. It has been reported in our former study that the bicontinuous rubbery network exhibited a much higher toughening efficiency than the dispersed particles did.<sup>36</sup> From variation of the toughness of the blends, it can be stated that conversion of SEBS-g-MA domain from the network to the irregular dispersed phase results in the reduction of the impact strength.

### Fracture behavior and morphology of macroscopic surface

Stress–strain curves for the PPO/SEBS-g-MA/nylon 6 blends were measured by tensile experiments and plotted as a function of nylon 6 content in Figure 6. SEM micrographs of the macroscopic fracture surfaces obtained from tensile experiments are presented in Figure 7. A typical ductile plastic fracture behavior with a yield stress and following large strain was found in the curve of the PPO/SEBS-g-MA binary blend. The incorporation of nylon 6 changes the nature of the curves considerably. It could be noted that the stress–strain curve for the ternary blend containing 5 wt % nylon 6 presented a higher yield stress, but a lower strain by comparison of those of the binary blends. The yield stress continuously increased with increasing the content of nylon 6. Simultaneously, the strain maintained a lower value than that of binary blend. It is obvious that there are two plastic phases in the ternary blends. Owing to the role of SEBS-g-MA as a compatibilizer between them, these two plastic phases possess high interfacial affinity to achieve a higher yield stress. However, the increase of the plastic phase resulted in stiffness, and, consequently, a reduction of the strain values.

Figure 7(a) gives an overview of fracture surface of the PPO/SEBS-g-MA binary blend, which presents the planar and small-deformed morphological feature. However, the fracture surfaces from the ternary blends exhibited a different macroscopic feature as shown in Figures 7(b)–7(f). One could observe a large-scale plastic deformation of the matrix from its morphological feature generated in the tensile experiment. The plastic deformation was distributed in an annular-layered structure, and the largest deformation was observed in the central part of the fracture surface. It is acceptable that the plastic deformation corresponding to the macroscopic stress-whitening observed during the tensile experiment is indicated by shear flow zones

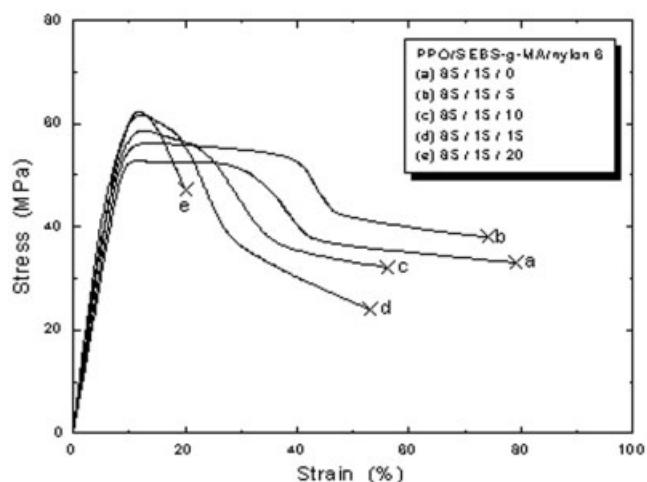


**Figure 5** TEM photographs of PPO/SEBS-g-MA/nylon 6 blends at weight ratio of: (a) 85/15/0, (b) 85/15/5, (c) 85/15/10, (d) 85/15/15, and (e) 85/15/20.

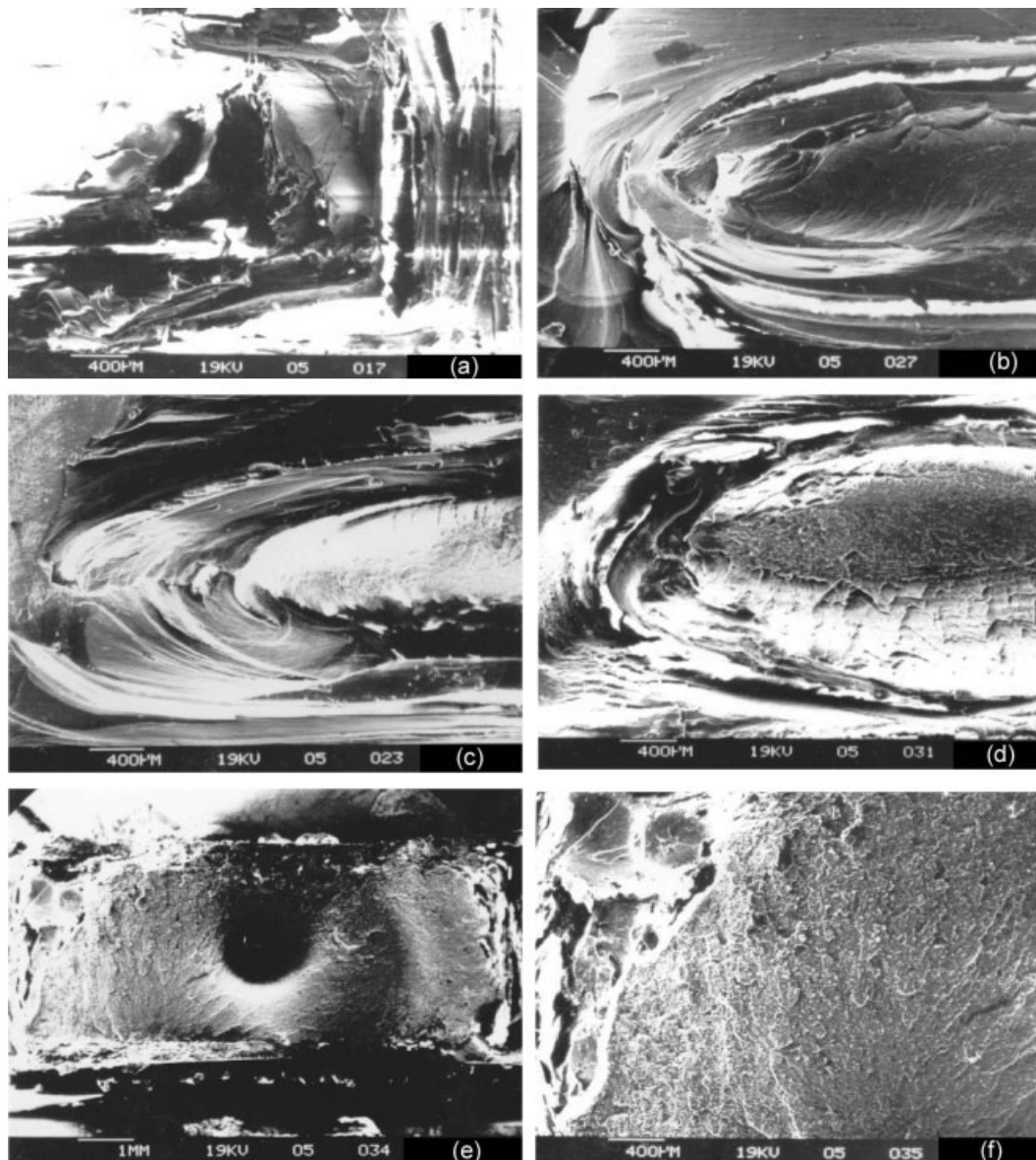
by shear lip.<sup>37</sup> The improvement in the yield stress of the ternary blends was attributed to this large-scale plastic deformation.

### Rheological properties

Apparent viscosities of PPO/SEBS-g-MA/nylon 6 blends based on various content of nylon 6 were measured in the capillary rheometer at 270°C and shown in Figure 8. The rheology behavior of polymer blend is generally a reflection of the change in molecular weight and the interaction among the components. It was found that the PPO/SEBS-g-MA binary blend possessed extremely high viscosities in all range of the shear rates carried out in this experiment. The viscosities were reduced remarkably by incorporating nylon

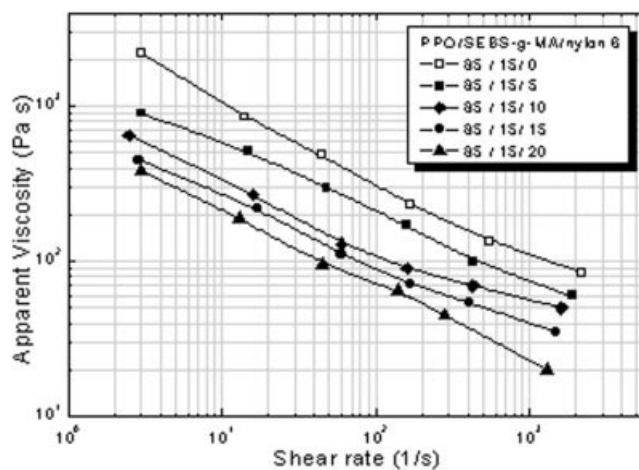


**Figure 6** Plots of stress versus strain for PPO/SEBS-g-MA/nylon 6 blends.



**Figure 7** SEM micrographs of the fractured surfaces of PPO/SEBS-g-MA/nylon 6 blends at weight ratio of: (a) 85/15/0, (b) 85/15/5, (c) 85/15/10, (d) 85/15/15, and (e, f) 85/15/20.

6, and kept decreasing with an increase of the content of nylon 6. The decrease in viscosity was mainly due to low viscosity of nylon 6 by comparison of that of PPO. The reaction of MA moieties on SEBS with the amine groups of nylon 6 may increase the molecular weight and the degree of branching, and both increase the viscosity of the blends; however, this effect is found to be less significant from our tested results. It is well-known that nylon 6 is thermodynamically immiscible with PPO or SEBS. Therefore, the molecular chains of various components are easily disentangled in melts by shear effect; thus, the viscosities were reduced.<sup>39,40</sup> The rheology study indicated an improvement of processability of the PPO/SEBS-g-MA blends by incorporation of the nylon 6.



**Figure 8** Plots of apparent viscosity versus non-Newtonian shear rate for PPO/SEBS-g-MA/nylon 6 blends.

## CONCLUSIONS

To improve the processability of the rubber-toughened PPO blends, blends of PPO, SEBS-g-MA, and nylon 6 were prepared via a melt extrusion. Incorporation of nylon 6 resulted in an improvement of the tensile strength, but reduction of both the notched Izod impact strength and elongation at break. The deterioration in toughness was caused by variation of rubbery phase structure, in which a network of SEBS-g-MA was destroyed because of the intervention of nylon 6. Increase of the tensile strength was resulted from the plastic deformation of matrix. The melt viscosities could be reduced significantly with increasing the content of nylon 6; thus, the processability was improved.

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