Synchronous Toughening and Reinforcing of Polypropylene with Ultrahigh-Molecular-Weight Polyethylene via Melt Blending: Mechanical Properties, Morphology, and Rheology

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ABSTRACT: Blending systems based on polypropylene (PP) and ultrahigh-molecular-weight polyethylene (UHMWPE) were prepared via a melt extrusion by the four-screw and the twin-screw extruders, respectively. The mechanical evaluation demonstrated that the synchronous toughening and reinforcing effects could be achieved from the combination of the PP and the UHMWPE, in which the toughness and the tensile properties could be improved with increasing the UHMWPE content, and achieved optimal values at a weight ratio of PP/UHMWPE (85/15). EPDM can be used as a compatibilizer to improve the compatibility and the interfacial adhesion between the PP and the UHMWPE. This resulted in more effective toughening and reinforcing effects. In contrast, for the PP/UHMWPE blends prepared by the normal twin-screw extruder, the poor dispersion capacity for the UHMWPE resulted in a deterioration of all mechanical parameters. Morphological observation revealed that the UHMWPE domain was well distributed as tiny particles in the PP matrix, which was confirmed by the differential scanning calorimetry analysis. The toughening effect was attributed to the energy dissipation caused by these rigid tiny particles that detached from the matrix to initiate the local matrix shear yield and formed the void. Rheological investigation demonstrated that there was an interesting composition dependence of viscosity, for which the melt viscosities of the PP/UHMWPE blends decreased when 5 wt % UHMWPE was added, and then began to increase as the UHMWPE content continued to increase. However, this dependence on composition became weaker because of the compatibilization of the EPDM. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 100: 3498–3509, 2006

Key words: UHMWPE; PP; toughening; morphology; mechanical properties

INTRODUCTION

The growing importance of polymer blending is widely accepted as an attractive low-cost substitute for the development of entirely new polymeric materials as well as an efficient way of providing unusual combinations of properties for materials. Polymer blends are receiving increasing attention from both the scientific and industrial communities, in resent years.[1] Most commercial multicomponent polymer systems are two-phase blends that have advantages over the single-phase systems.[2] There is a general agreement that the properties of polymer blends are usually controlled by the properties of the components, the morphology of the blends, and the interaction between the components in the blends.[3–5] 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.[6–9] A detailed 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.[10–13] Polymer toughening through a melt extrusion is a major goal in 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.[14,15] The toughening of rubber-toughened polymer is strongly influenced by the amount, the particle size, and the properties of the elastomers, as well as the interactions between the phases that determine the mechanical properties of the toughened plastics.[16,17] However, while a good toughness is achieved for rubber-toughening systems, there is a penalty of the tensile strengths and stiffness, which is resulted by the incorporation of the soft materials like rubbers. Therefore, the realization to synchronous toughening and reinforcement of the brittle polymers becomes the pursuing target of many researchers.

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Impact toughness of ductile polymers can also be improved by blending with rigid particles, usually brittle polymer component like polycarbonate or poly-(styrene-*co*-maleic anhydride) for toughening nylon 6.[18,19] Though the enhancement of toughness in these systems is lower than that in systems toughened by elastomers, the advantage of rigid-brittle toughening is a simultaneous increase in toughness and stiffness, whereas reduction of these parameters is a disadvantage of the addition of elastomers. From the articles dealing with rigid polymer toughening brittle polymer systems having well-balanced mechanical properties, it is clear that such a system must have a sufficiently fine phase structure and high interfacial adhesion.[20–22] It was shown that, in this case, the loading also causes plastic deformation of dispersed rigid particles with a ductile matrix. As a result, the rigid particle absorbs mechanical energy instead of brittle fracture occurring in the bulk.

As one of the most important general plastics, polypropylene (PP) exhibits many merits on the strength, the thermal stability, the processability, the solvent resistance, etc. However, deficiencies, such as natural brittleness, prohibit its broader application.[23,24] Numerous investigations on the toughening blends of PP with various elastomers, such as ethylene–propylene–diene block copolymer (EPDM), poly-(ethylene-*co*-1-octene) (POE), styrene–butadiene–styrene block copolymer (SBS) and so on, have been reported in the past decades.[25–27] Although a satisfied toughening effect comes from these systems, the deterioration in tensile strengths and stiffness also fazes the developers. The current research on PP was mainly focused on the blends of ultrahigh-molecular-weight polyethylene (UHMWPE) with PP. Mironi-Harpaz and Narkis[28] investigated the electronic behavior and structure of the PP/UHMWPE blends filled with carbon black, and showed that UHMWPE was chosen as the dispersed phase because of its universal structure and extremely high viscosity. Wang and Zhou[29] prepared the PP/UHMWPE blends via a melt-vibration blending, and provided a novel processing method for such an extremely viscous blending system. Liu et al.[30–32] reported several processing technologies regarding the preparation of the PP/UHMWPE blends, and found that the incorporation of PP into UHMWPE can reduce the melt viscosity and improve the processability of the UHMWPE.

In this study, we employed the UHMWPE as an impact modifier for the PP to prepare the toughened blends. UHMWPE is characterized by a number of important properties, including high notched impact strength, tensile strength, energy absorption capacity at high loading rates, resistance to stress cracking, and extremely low embrittlement temperatures. Thus, it is reasonable to expect that a combination of PP and UHMWPE should provide the advantages from individual components, based on the inherent properties of these two polymers, and may give a synergistic effect on toughness and stiffness. However, UHMWPE, at such a high molecular weight, displays an extremely high viscosity in the melt that makes it unsuitable for conventional processing operations, with the exception of compression molding and ram extrusion. Thus, we employed a corotating four-screw extruder to prepare the PP/UHMWPE, and evaluated the mechanic properties through comparing with that of the corresponding blends prepared by the general twin-screw extruder. The morphology, the thermal properties, and the rheology of the blends were also investigated.

EXPERIMENTAL

Materials

UHMWPE was purchased from Beijing No. 2 Auxiliary Agent Company, China. It has a density of 0.64 g/cm^3 and an average molecular weight of 2,500,000 determined from the intrinsic viscosity measurement and the calculation using the Mark–Houwink equation. PP (1300, injection-molding grade) with a melt flow index (MFI) of 1.5–2.0 g/10 min and a density of 0.91 g/cm^3 was supplied by Beijing Yanshan Petrochemical Company, China. EPDM (IP 3725P), with a density of 0.84 g/cm^3 and Mooney viscosity ML_{1+4} of 40 at 125°C was commercially obtained from Dupont-Dow Elastomer Company (Wilmington, DE).

Preparation of blends

The pellets of all the components were dried in a vacuum oven at 80°C overnight before using. The constituents of the blends with various weight ratios were blended using a ZSK-25 twin-screw extruder (Werner and Pfleidere Company, Germany) and a 4SJ-40 four-screw extruder (Chenguang Plastics Machine Company, China), respectively. In the four-screw extruder, the corotating four screws were specially designed in an array of the "V" type with a screw diameter of 40 mm and a length–diameter ratio of 28. It was an enhanced version of the general twin-screw extruder. The screw configurations for both of the extruders were set to be adapted to the PP. All the ingredients were tumble-mixed and fed through the throat of the extruder. The temperatures along the barrel were set from 205 to 240°C, and the rotation speed of the screw was 150 rpm. 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 80°C before injection molding the test specimens for determination of mechanical properties.

Measurements of mechanical properties

The notched impact and tensile test bars were prepared via injection molding using an SZ-250 reciprocating screw injection machine; the barrel temperature was 230°C and the mold temperature was 90°C. The tensile properties were determined with an Instron-1185 universal testing machine using a 5000 N load transducer, according to the standard of ASTM D638. Notched Izod impact strength was measured with a Sumitomo impact tester, according to the standard of ASTM D256. The thickness of Izod impact specimens was 1/8 in., and impact energy was 4 J. All the tests were done at room temperature and five tests 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. The first heating scan was carried out from 20 to 250°C and the sample was held at this temperature for 3 min, to diminish the thermal and processing history effects, before the formal measurements.

Scanning electron microscopy

A Cambridge S250 scanning electron microscope was used to study and record the fracture surface of the blending samples. The fractured specimen surfaces from tested impact bars were etched with boiling toluene for 2 h, and then 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

The specimens for transmission electron microscopy (TEM) were cryogenically microtomed from Izod impact bars perpendicular to the flow direction, using a diamond knife, and then were stained with ruthenium tetroxide (RuO_4). A Hitachi H-800 transmission electron microscope was used to observe and photograph these specimens, which are only of the order of nanometers, in thickness.

Measurements of rheological properties

The apparent viscosities at various shear rates were determined using a Capirograph-1B capillary rheom-
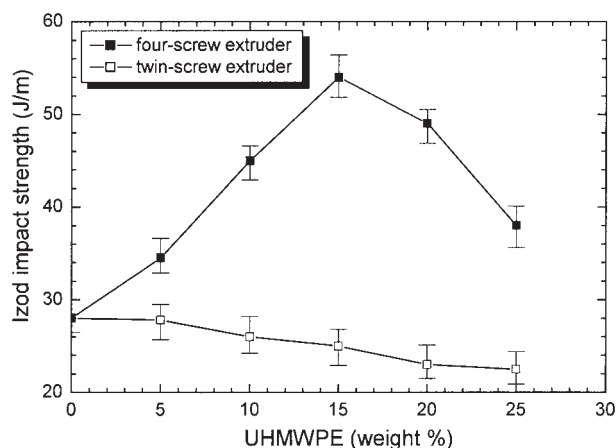

Figure 1 Notched Izod impact strength of PP/UHMWPE binary blends as a function of UHMWPE content.

eter (Toyoseiki Manufacture Company) with a capillary diameter of 1.262 mm and an L/D ratio of 50.51. The tests were carried out at 200, 220, and 240°C under the shear rates ranging from 1 to 10^4 s^{-1}.

RESULTS AND DISCUSSION

Mechanical properties of PP/UHMWPE blends

Figure 1 illustrates the notched Izod impact strength of PP/UHMWPE binary blends prepared by the twin-screw and the four-screw extruders, respectively, and indicates that the UHMWPE exhibits a significant toughening effect on PP for the blends prepared by the four-screw extruder, and that the Izod impact strength of these blends increases as the UHMWPE content increases. The impact strength achieves a maximum value of 54.8 J/m for the blend containing 15 wt % UHMWPE, which increases almost by a factor of 2 in comparison with that of pure PP. However, the impact strength begins to drop after the UHMWPE content exceeds 15 wt %. On the other hand, it is found that the toughening effect is negative for the binary blends prepared by the twin-screw extruder. The Izod impact strength decreases, as the UHMWPE content increases. A similar tendency can be observed from the variation of the tensile properties of the binary blends as a function of the UHMWPE content, as shown in Figures 2 and 3, respectively. It is found that, for the PP/UHMWPE blends prepared by the four-screw extruders, both the tensile strength and the elongation-at-break increase, as the content of UHMWPE increases, and achieves the optimal value when the UHMWPE content is 15 wt %. However, the binary blends prepared by the twin-screw extruder present a slight reduction in all the parameters (i.e., tensile strength and elongation-at-break). As we know, the UHMWPE has an extremely high viscosity in the melt state, and is very difficult to be dispersed in the PP matrix by the
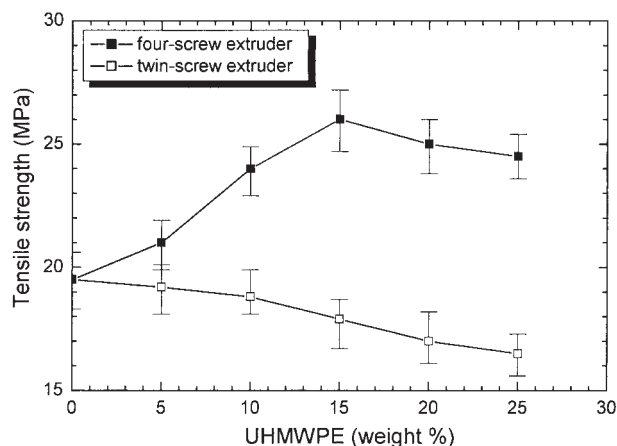
Figure 2 Tensile strength of PP/UHMWPE binary blends as a function of UHMWPE content.

normal twin-screw extruder. It can be easily concluded that a negative deterioration in mechanical properties of the binary blends is resulted from poor dispersion of the UHMWPE caused by the twin-screw extruder. Nevertheless, the four-screw extruder used in this study was configured with four corotating and intermeshing screws, which triplicates the mixing effect in comparison with that of the twin-screw, at the same extrusion condition (screw speed, configuration of screw elements, etc.); therefore, it can exhibit a powerful process capability for the high-viscosity polymers like UHMWPE. It is well known that the dispersed-phase component undergoes the drop break-up and coalescence in multiphase fluid during melt extrusion, and the final morphology (or say "dispersion extent") of a blend represents the balance of the drop break-up and coalescence rates at the end of the mixing process.[33] For the polymers with extremely high viscosity, like UHMWPE, the drop break-up is so difficult that the bulk of UHMWPE still remains under the extrusion, with the general twin-screw extruder. However, the four-screw extruder can provide long-term shearing and kneading effects to facilitate the drop break-up of the UHMWPE. As a result, the UHMWPE achieves a much better dispersion in the PP matrix by using the four-screw extruder than by the twin-screw one. This can be considered as a reasonable explanation that the mechanical properties are significantly improved for the corresponding binary blends.

Argon and Cohen have presented a rigid-particle toughening mechanism, which is governed by the debonding or cavitation process and works through the principle of crack tip shielding, often in more than one form.[34,35] For the PP/UHMWPE binary blending system in this study, the UHMWPE has a much greater Young's modulus than the PP matrix, and so it will not basically deform, when the blending speci-

mens are subjected to external loading. Moreover, the PP is a semibrittle thermoplastic, and thus, the plane strain state cannot be released, making matrix deformation difficult. Therefore, the main toughening effect would come from debonding at the interface, which occur first, leading to the formation of microvoids or say "crack," when the stress applied exceeds the interfacial adhesion strength between the UHMWPE and the PP matrix. In this case, the deformation restraint of the matrix around the UHMWPE is released, resulting in the production of extensive elastic deformation and absorbing strain energy and the promotion of the enhancement in toughness as well as the tensile properties.[36] The relatively higher UHMWPE content can result in more effective deformation, and thus, more absorbing energy. This makes the impact strength increase as a function of the UHMWPE content and so do the tensile properties. However, it is noticed that the mechanical properties begin to drop after undergoing an increase and optimum with an increase of the UHMWPE content. As mentioned previously, although the four-screw extruder can provide much stronger dispensability than the twin-screw one through facilitating the drop break-up of the UHMWPE, this capacity of the drop break-up is dependent on the UHMWPE content. It is acceptable that the higher the UHMWPE content, the more difficult is the drop break-up process. When the balance of the drop break-up and coalescence rates is broken at a point of the UHMWPE content (about 20 wt % in this study), the UHMWPE phases will greatly aggregated. This means that even the four-screw extruder cannot make too much UHMWPE be dispersed in the PP matrix. Therefore, the mechanical properties begin to reduce when the UHMWPE content exceeds 15 wt %.

For the rigid-particle toughening system, like the PP/UHMWPE blends, although this kind of toughening effect is relatively minor in comparison with rubber-toughening systems, it should be noticed that it
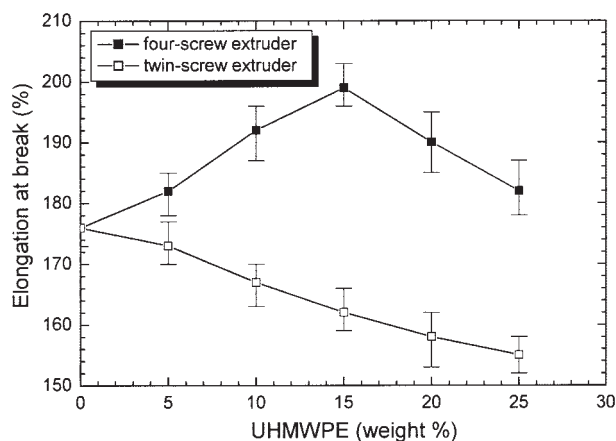

Figure 3 Elongation-at-break of PP/UHMWPE binary blends as a function of UHMWPE content.
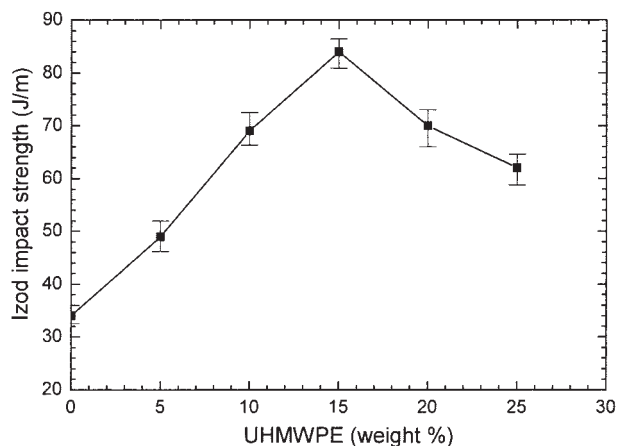
Figure 4 Effect of EPDM on notched Izod impact strength of PP/UHMWPE blends as a function of UHMWPE content.

should be more beneficial to use rigid plastics, since they would also improve tensile properties of the overall composite, roughly by as much as rubbery particles would compromise them.

Mechanical properties of PP/UHMWPE/EPDM blends

EPDM has been widely used as an impact modifier for PP.[37] On the other hand, the EPDM comprises the ethylene segments and propylene segments within its chain, and this makes the EPDM miscible with both PP and polyethylene (PE). Therefore, the EPDM can also be employed as a compatibilizer for PP/PE blends, where the EPDM bridges the PP and the PE through the physical interaction from this miscibility.[38–40] The effects of EPDM on the toughness and the tensile properties are presented in Figures 4 and 5, respectively. Note that the incorporation of 5 wt % EPDM into pure PP results in a slight improvement in toughness and elongation-at-break, but a moderate reduction in tensile strength. When 5 wt % EPDM was incorporated into the PP/UHMWPE blends, the ternary blends exhibited a much higher impact strength than that of the PP/UHMWPE binary blends at constant UHMWPE content. For the ternary blends containing 15 wt % UHMWPE, the impact strength reaches an optimal value of 84.3 J/m by a factor of 3 in comparison with the pure PP. Notice that the tensile properties were also enhanced by incorporating EPDM into the PP/UHMWPE blends, and their tendency as a function of UHMWPE content was similar to those observed for the impact strength.

It is evident that the enhancement in toughening effect arises not from the toughening of EPDM, but from the compatibilization of EPDM between the PP and the UHMWPE, because the former is quite limited

and follows a penalty in the tensile strength. As mentioned previously, the mechanism for the UHMWPE toughening PP was attributed to the debonding or cavitation process. It is acceptable that the properly stronger interfacial adhesion is in favor of the initiation of the matrix shear yielding, while the UHMWPE particles detaches from the PP matrix. Moreover, many studies demonstrated that multiple-crazing in the polymer matrix also exist at the same time; for strong interfacial adhesion, the crazing is favored. As a result, the toughening effect is much better. For the PP/UHMWPE/EPDM ternary blends, the EPDM plays a role of compatibilizer for the PP and the UHMWPE and improves the interfacial adhesion between them, and thus enhances the toughening effect for the UHMWPE. On the other hand, as an inherent strong rigid material with the superlonger chain, the well-dispersed UHMWPE can act as a backbone for the PP matrix, which results in an increase in the tensile properties. It is evident that the stronger interfacial adhesion redounds to this reinforcing effect.

Thermal analysis

The thermal behaviors of the PP, the UHMWPE, and their blends were investigated using DSC. The DSC thermograms of the pure resins and their blends prepared by the twin-screw and the four-screw extruders are illustrated in Figures 6 and 7, respectively. The melt points (T_m's) and the heat fusions (ΔH_m's) obtained from DSC analysis were summarized in Table I. The values of the ΔH_m's were normalized to the weight percents of the PP and the UHMWPE phases in blends, respectively. The thermal data confirm a high crystallinity for both the PP and the UHMWPE with T_m's of 165.1 and141.4°C, and ΔH_m's of 137.7 and 189.3 J/g, respectively. For the binary blend containing 5 wt % UHMWPE prepared by the four-screw extruder, it
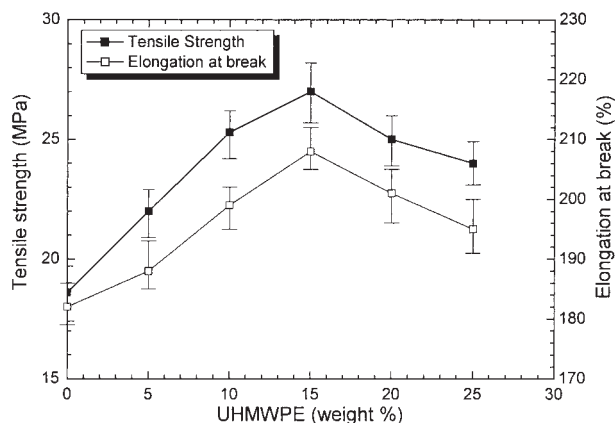

Figure 5 Effect of EPDM on tensile strength and elongation-at-break of PP/UHMWPE blends as a function of UHMWPE content.
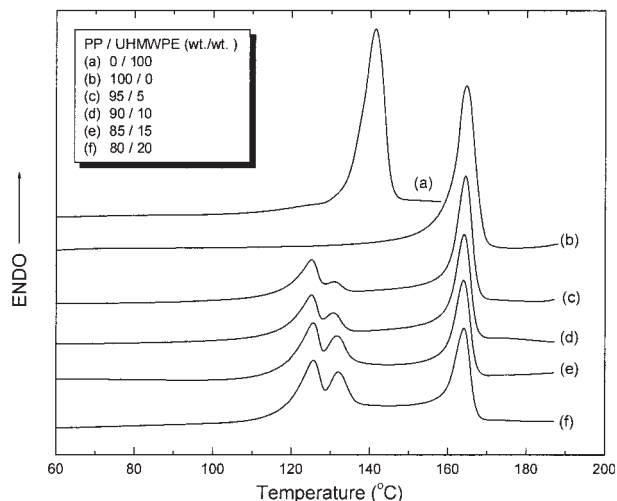
Figure 6 DSC heating traces of PP, UHMWPE, and their blends prepared by the four-screw extruder.

is seen that there are two peaks, whose transition temperatures correspond to T_m's of the PP and the UHMWPE phases in the blends. It can be easily observed that both the T_m and the ΔH_m of the PP phase in the binary blends are almost not affected by incorporation of the UHMWPE. This implied immiscibility of the PP and the UHMWPE. However, the two small peaks associated with the transition of the UHMWPE phase shifts to the direction of lower temperature. The corresponding T_m reduces to 123.2°C with a decreasing ΔH_m in comparison with those of the pure UHMWPE, which indicates that the crystallinity of the UHMWPE phase decreases in presence of the PP matrix and two crystal structures exist at least. Owing to its extremely high viscosity, after being dispersed in the PP melt, the UHMWPE chains are surrounded by the PP melt and are difficult to aggregate to form the bulk crystal, like its pure resin. Therefore, it can be deduced that the UHMWPE phase presents in the formation of the small crystal in the PP matrix. A slight increase in the T_m and the ΔH_m for the UHMWPE phase is observed with an increase in the UHMWPE content. This result is caused by the increase in the crystal size and the crystallinity of the UHMWPE phase. Moreover, no influence of the UHMWPE content on both the T_m and the ΔH_m of the PP phase can be found.

On the other hand, for the PP/UHMWPE blends prepared by the twin-screw extruder, a reduction in the T_m and the ΔH_m only for the UHMWPE phase is also observed, which indicated the decrease in the crystal size and the crystallinity. In fact, the decrements of the temperatures and the enthalpies associated with the transition of the phase of the UHMWPE phase are less than those of the blends prepared by the four-screw extruder. This verified that the dispersing capability of the twin-screw extruder is weaker than

that of the four-screw extruder, which results in a larger UHMWPE domain.

Morphology of PP/UHMWPE blends

Figure 8 shows the SEM micrographs of the fracture surface of the PP/UHMWPE binary blends prepared by the twin-screw extruder, which revealed a similar morphology of the fracture surface for the pure PP and the binary blends. It can be observed from Figure 8(a) that the fracture surface of the pure PP is smooth. It is clear that seldom deformations of the matrix occurred during the break, and the plastic phase was inactivated. From Figure 8(b–d), one can find a somewhat larger plastic deformation on the fracture surface of the binary blends than that of the pure PP; however, the interface between two phases can hardly be distinguished. Although clear phase separation is not observed throughout the blended composition in this study, it can still be assumed that the UHMWPE only dispersed as an aggregated bulk domain in the PP matrix because of the poor dispersion capability of the twin-screw extruder. This deduction has also been confirmed by DSC analysis and the fairly significant impairment of the mechanical properties with increasing the UHMWPE content for the binary blends prepared by the twin-screw extruder.

For the binary blends prepared by the four-screw extruder, a completely different phase structure for the fracture surface is discovered, as shown in Figure 9. Morphological observation of the blend containing 5 wt % UHMWPE shows that a large number of tiny particles are dispersed uniformly on the fracture surface, and some small scale-shape deformation can also be observed around these tiny particles. The number and the size of the tiny particles increase as the UH-
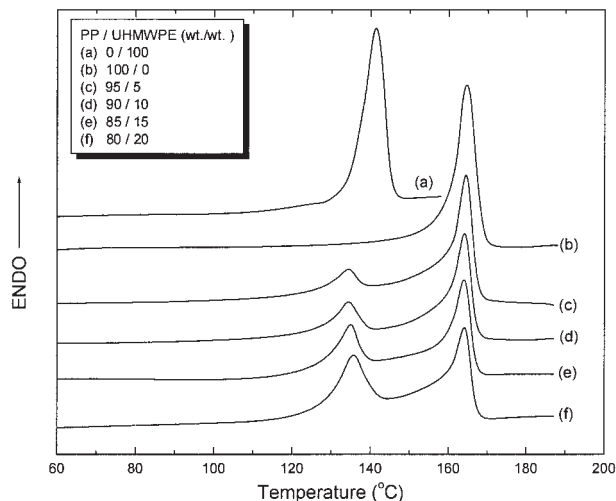

Figure 7 DSC heating traces of PP, UHMWPE, and their blends prepared by the twin-screw extruder.

TABLE I
Thermal Properties of PP, UHMWPE, and Their Blends from DSC Analysis

Blending sample (wt/wt) PP/UHMWPE	$T_{m, PP}$ (°C)	$\Delta H_{m, PP}$ [a] (J/g)	$T_{m, UHMWPE}$ (°C)	$\Delta H_{m, UHMWPE}$ [a] (J/g)
100/0	165.1	137.69	—	—
0/100	—	—	141.4	189.29
95/5[b]	165.2	136.92	123.2	126.47
90/10[b]	164.9	134.85	123.7	128.15
85/15[b]	165.3	136.87	124.3	131.79
80/20[b]	164.4	135.69	124.9	135.33
95/5[c]	164.6	134.62	132.8	149.12
90/10[c]	164.8	132.58	133.2	151.47
85/15[c]	164.3	135.73	133.9	153.86
80/20[c]	164.5	133.29	134.4	158.72

[a] All values were normalized to the amounts of the PP and UHMWPE phases in the blends, respectively.
[b] The blends were prepared by the four-screw extruder.
[c] The blends were prepared by the twin-screw extruder.

MWPE content increases, and the particle size is around 0.2–0.3 μm by eyeballing from the magnified SEM micrograph [Fig. 9(e)] when the UHMWPE content reaches 15 wt %. These tiny particles are reasonably considered as the UHMWPE domain, which act as rigid particles to toughen and reinforce the PP. However, from Figure 9(f), it can be seen that, when the UHMWPE content exceeds 20 wt %, the particle size increases significantly, and the distribution of the particle size also becomes uneven. As discussed previously, the higher UHMWPE content results in a greater difficulty in the drop break-up. A new balance of the drop break-up and coalescence rates at 20 wt %

UHMWPE content makes the UHMWPE phases greatly aggregated, and results in a worse dispersion of the particles, which caused a lower toughening effect.

The morphology of the binary blends prepared was also observed by TEM. The TEM image of PP/UHMWPE systems could be identified with the aid of the RuO$_4$ staining. Although both PP and UHMWPE can be stained with RuO$_4$, the staining rates are very different.[41] It was found that the staining rate of the amorphous part of UHMWPE was fastest than that of PP, in our experiments. The UHMWPE appears in the images as dark region, and PP appears as light gray
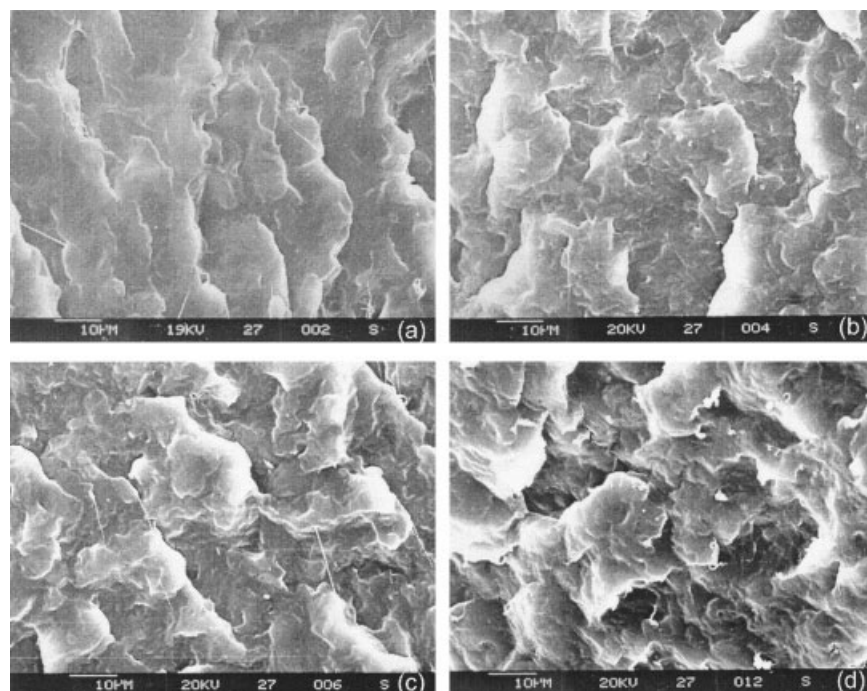

Figure 8 SEM micrographs of the PP/UHMWPE binary blends prepared by the twin-screw extruder at weight ratios of: (a) 100/0, (b) 90/10, (c) 85/15, and (d) 80/20.
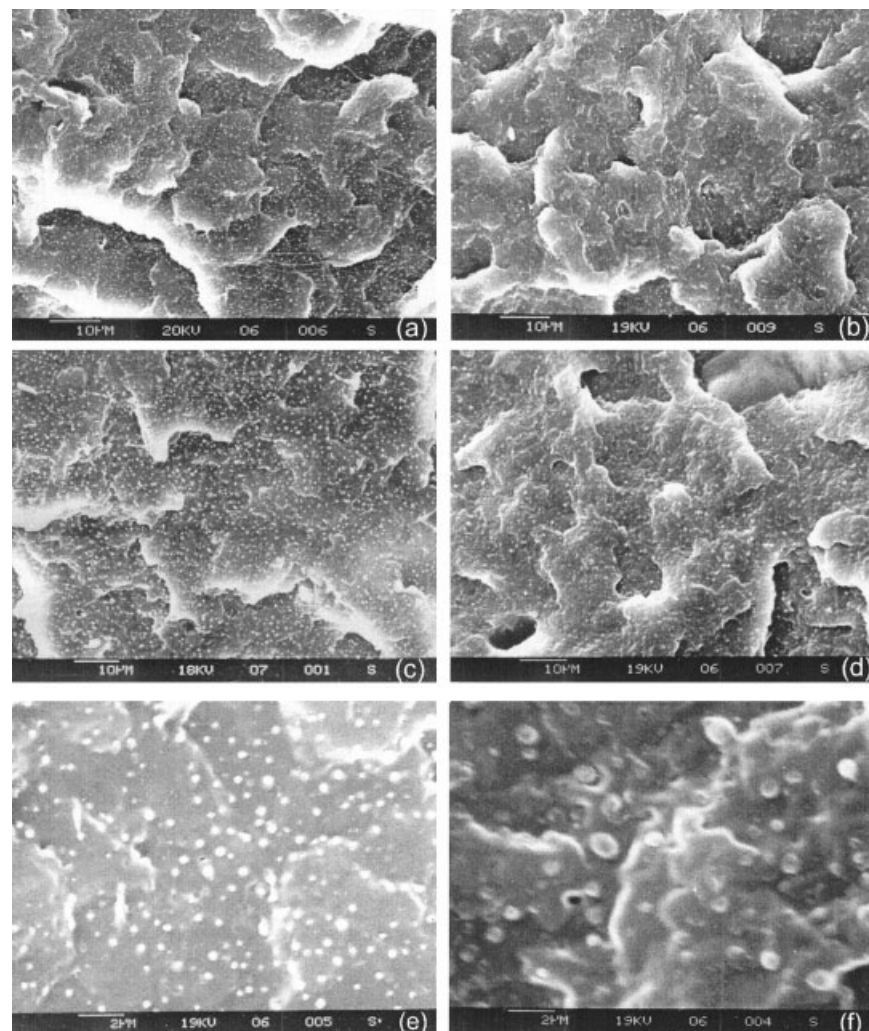
Figure 9 SEM micrographs of the PP/UHMWPE binary blends prepared by the four-screw extruder at weight ratios of: (a) 95/5, (b) 90/10, (c, e) 85/15, and (d, f) 80/20.

one via controlling the staining time, as shown in Figure 10(a,b). From Figure 10(c–e), it is observed that the dark region is dispersed widely in the image, and its area increases as the UHMWPE content increases. The dark region occupies a much larger area than the tiny particles in the SEM micrographs, at the constant UHMWPE content. It can be deduced that the tiny particles in SEM micrographs reflect the crystal aggregation of the UHMWPE and the dark region in TEM micrographs represent the amorphous part of the UHMWPE domain, which connects with the crystal aggregation. The superlong chains of the UHMWPE result in a poor movability in the PP melt, and thus, in a lower crystallinity for the UHMWPE domain, which confirms the DSC analysis results. The TEM micrographs have verified once more that the four-screw extruder makes the UHMWPE well dispersed in the PP matrix via a melt blending, which has been probed to be a key factor that determines the toughening effect for the blends.

Morphology of PP/UHMWPE/EPDM blends

The fracture surface morphology of the PP/UHMWPE/EPDM ternary blends is displayed in Figure 11 as SEM micrographs, in which one may observe a very interesting phase structure. All of the images indicated that an orientation occurs for the primary particles representing the UHMWPE domain. From the magnified micrograph in Figure 11(d), one can observe clearly that the UHMWPE particles are stretched into long rods with a size of around 1 μm. There is a general agreement that the EPDM can enhance the compatibility and the interfacial adhesion between the PP and the UHMWPE.[38–40] When the tested specimens containing the EPDM were impacted, the particles of the UHMWPE in the matrix were drawn, and the strong interfacial adhesion can keep the entire UHMWPE domain stretching into elongated rod during the debonding. It is clear that this process will result in much more energy dissipation. Otherwise,
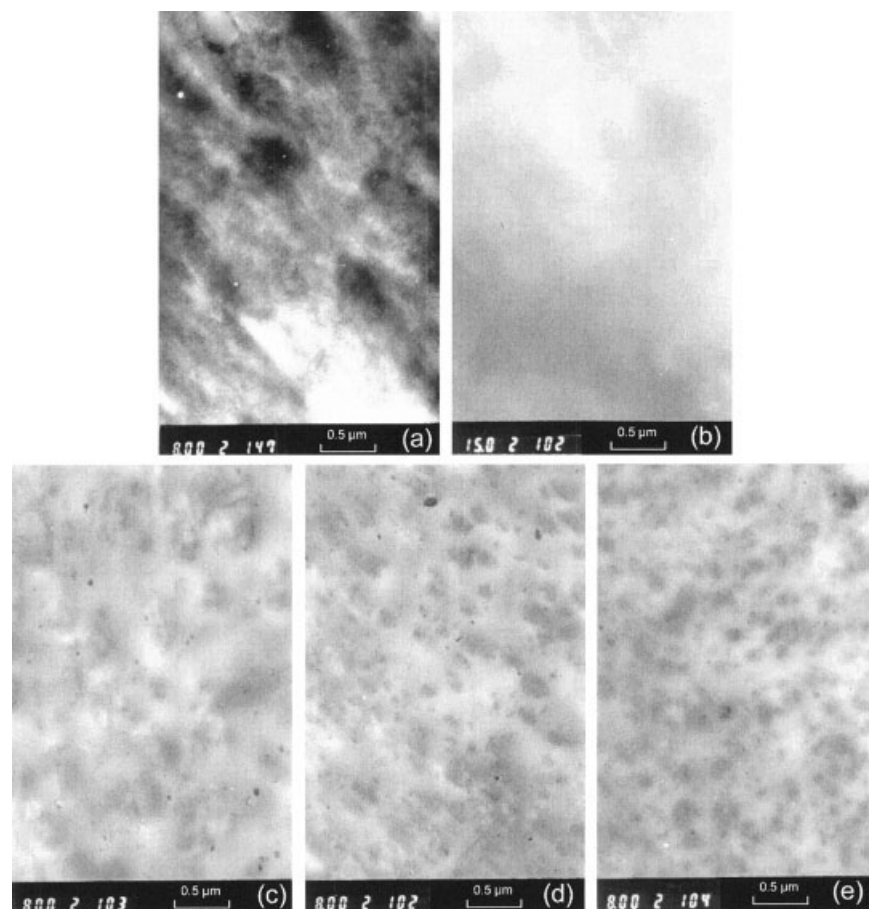
Figure 10 TEM micrographs of the PP/UHMWPE binary blends prepared by the four-screw extruder at weight ratio of: (a) 100/0, (b) 0/100, (c) 95/5, (d) 85/15, and (e) 80/20.

owing to the relatively worse interfacial adhesion for the PP/UHMWPE binary blends, the UHMWPE domain is easily broken while the particles of the UHMWPE in the matrix are drawn during the fracture. As a result, only spherical particles present in the fracture surface, as shown in Figure 10. This is the reason why all ternary blends exhibit much higher toughness and the other mechanical parameters, than the binary blends at constant UHMWPE content.

We also observed the morphology of the EPDM and its ternary blends by TEM at the same staining conditions. The TEM micrographs are presented in Figure 12. It can be found that the staining rate of pure EPDM is faster than that of the pure PP but slower than that of the UHMWPE [Fig. 12(a)], and so, the EPDM domain appears slight dark gray in the TEM micrograph. The EPDM domain can be still distinguished from the TEM micrographs for the ternary blends. Note that the slight dark gray region only exists around the dark particles. It is found that the EPDM is not dispersed as individual particles but as transition layers between the UHMWPE particles and the PP matrix. This verifies that EPDM plays the role of a compatibilizer instead of an impact modifier.

Rheological properties

The apparent melt viscosities of the PP/UHMWPE blends based on various UHMWPE contents were measured in the capillary rheometer at 200, 220, and 240°C, and were shown in Figure 13. The rheology behavior of polymer blend is generally a reflection of the change in molecular weight and the interaction among the components. It is found that the melt viscosity of the binary blend containing 5 wt % UHMWPE exhibits a slight reduction in comparison with that of the pure PP at these three testing temperatures, which is due to the immiscibility between these two polymers. However, a strange composition dependence of the viscosity can also be observed as a function of the UHMWPE content, in which, the viscosities begin to increase as the UHMWPE content continues to increase. Rheological behaviors of immiscible polymer blends having minor dispersed phases are different from those of homogeneous polymers, owing to the contributions of the minor separated phase and of the interface between the two component phases.[42] Repulsion resulted from the immiscibility between the PP ma-
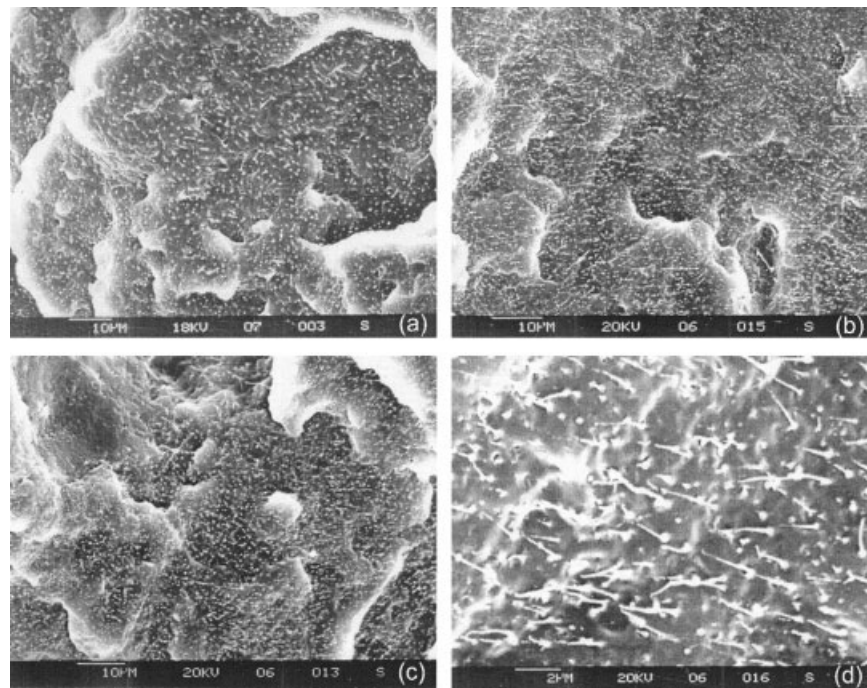
Figure 11 SEM micrographs of the PP/UHMWPE/EPDM ternary blends prepared by the four-screw extruder at weight ratio of: (a) 95/5/5, (b) 90/10/5, and (c, d) 85/15/5.

trix and the UHMWPE domain caused this negative deviation in the melt viscosity. However, the UHMWPE has a long chain and an extremely high inherent viscosity. When the UHMWPE content increases, the intermolecular entanglement becomes greater. As a result, the viscosity begins to increase. On the other hand, the viscosity dependence of composition is also affected by shear rate and temperature. It is well known that the intermolecular entanglement and disentanglement determine the melt viscosity of the polymer blend.[43] The repulsion arising from the immiscibility between the PP seg-

ments and the UHMWPE segments can cause this disentanglement, while the shearing results in the entanglement. It is evident that the disentangling effect is much stronger at low shear rate, and as a result, the decrement in the viscosity at low shear rate is greater than that at higher one. Moreover, the temperature can generate a positive influence on the composition dependence of the viscosity. Thus, the higher temperature results in a more active molecular motion, which favors the disentanglement. Therefore, at the higher temperature, the higher the UHMWPE content, the decrement in the viscosity is
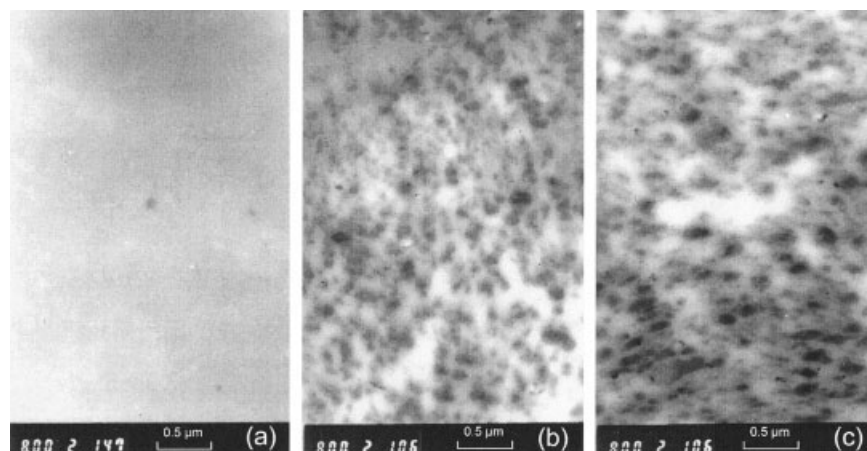

Figure 12 TEM micrographs of the PP/UHMWPE/EPDM ternary blends prepared by the four-screw extruder at weight ratio of: (a) 0/0/100, (b) 90/10/5, and (c) 80/20/5.
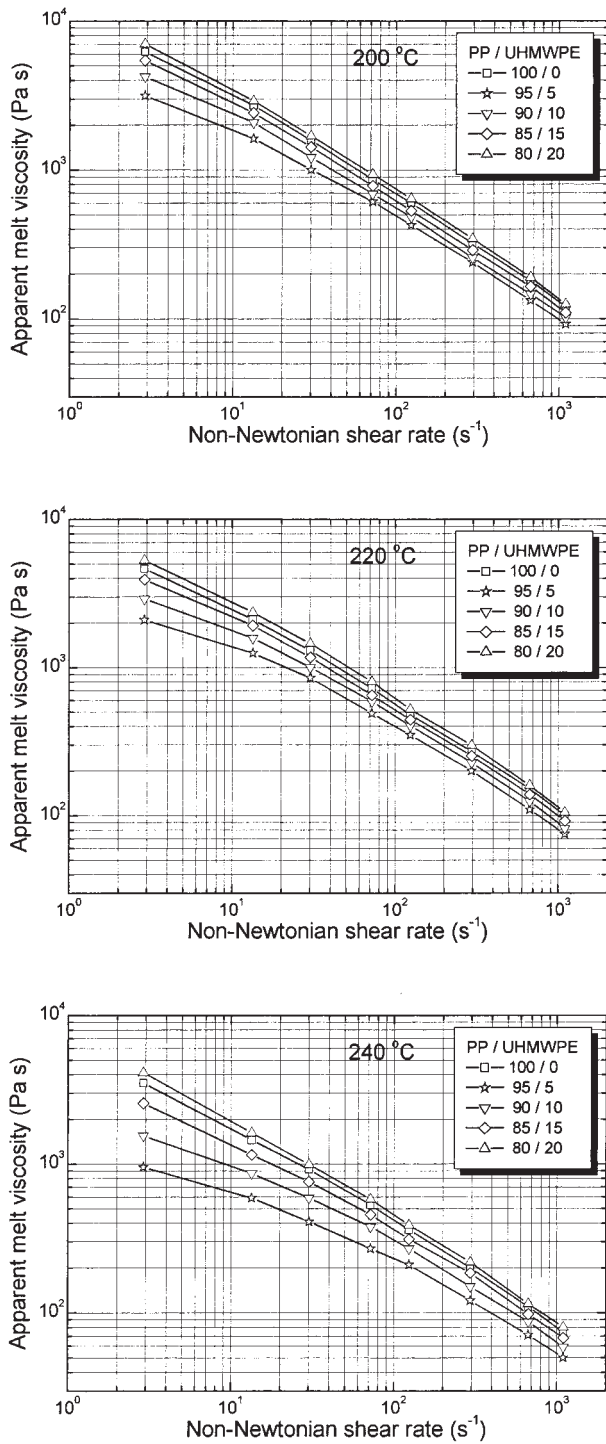
Figure 13 Plots of apparent melt viscosity versus non-Newtonian shear rate for PP/UHMWPE binary blends at various temperatures.

larger at the same shear rate. As a result, the differences in the viscosity of the blends tended to increase with a rise in the temperature.

Figure 14 displays the apparent melt viscosities of the PP/UHMWPE/EPDM ternary blends as a function of the UHMWPE content at three different tem-

peratures. It can be observed from this figure that the composition dependence of the viscosity becomes much weaker in comparison with that for the binary blends. As discussed previously, the incorporation of EPDM enhances the compatibility of the PP matrix and the UHMWPE domain. This can
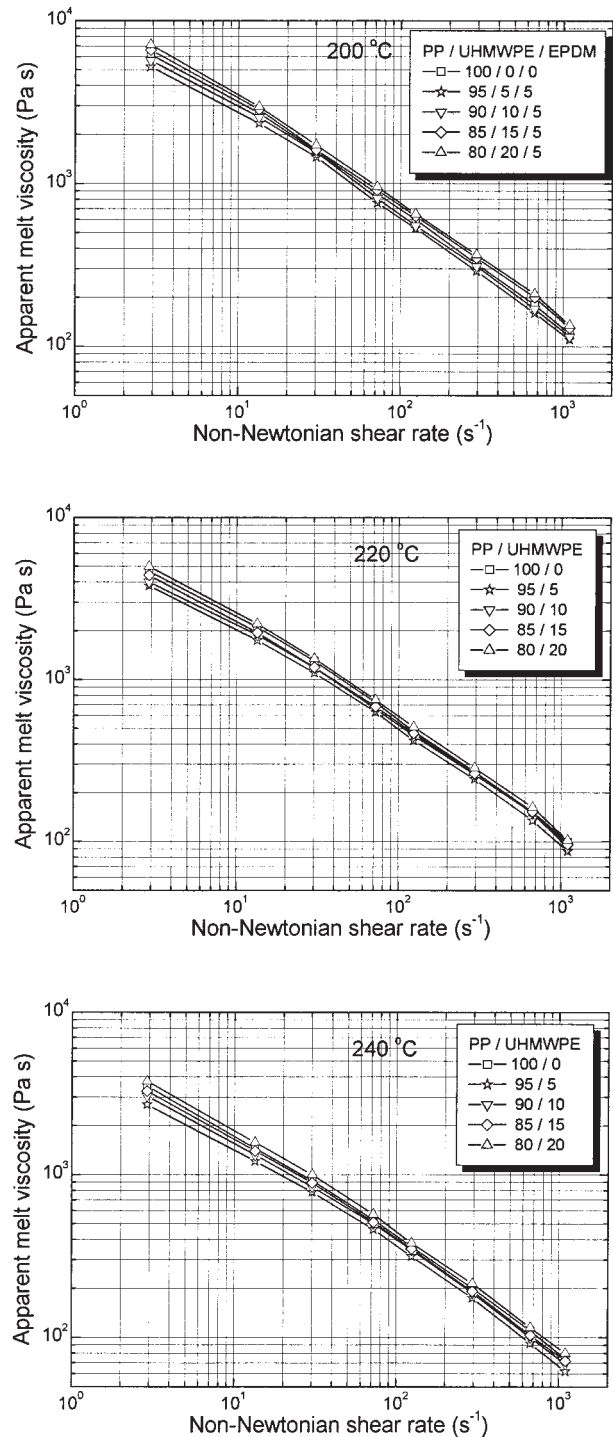

Figure 14 Plots of apparent melt viscosity versus non-Newtonian shear rate for PP/UHMWPE/EPDM ternary blends at various temperatures.

weaken the repulsion between these two polymers, and thus, the decrement in viscosity becomes less when the EPDM is introduced into the PP/UHM-WPE blends. It is also observed that the differences in the viscosity of ternary become less pronounced when EPDM is incorporated as a compatibilizer. It can be deduced that the repulsion between the molecules of PP and UHMWPE arising from the immiscibility is greatly weakened by introducing the EPDM. This reduces the difference in the interaction among the different molecules, and makes the activity of molecular motion for the blends with the different compositions become similar. As a result, the influence of temperature on the composition dependence of the viscosity of the ternary blends became less significant. The rheological property investigation indicates that the PP/UHMWPE blending system is still processable, though the UHMWPE with extremely high melt viscosity itself are hardly processed.

CONCLUSIONS

The PP/UHMWPE blends can be well prepared via melt extrusion by a specially designed four-screw extruder. The synchronous toughening and reinforcing effects could be achieved from the combination of these two polymers. The notched Izod impact strength, the tensile strength, and the elongation-at-break could be improved with increasing UHMWPE content, and achieved optimal values at an 85/15 weight ratio of PP/UHMWPE. EPDM can be used as a compatibilizer to improve the compatibility and the interfacial adhesion between the PP and the UHM-WPE, which resulted in more effective toughening and reinforcing effects. In contrast, for the PP/UHM-WPE blends prepared by the normal twin-screw extruder, the poor dispersion capacity for the UHMWPE resulted in deterioration of all mechanical parameters. Morphological observation revealed that the UHM-WPE domain was well distributed as tiny particles in the PP matrix. The toughening effect was attributed to the energy dissipation caused by these rigid tiny particles that detached from the matrix to initiate the local matrix shear yield and formed the void. Strong interfacial adhesion by incorporation of the EPDM could result in a stretch of the rigid UHMWPE particles, and thus, in more energy dissipation. Rheological investigation demonstrated that there was a strange viscosity dependence of composition, in which the melt viscosities of the PP/UHMWPE blends decreased when 5 wt % UHMWPE was added, and then began to increase as the UHMWPE content continued to increase. However, this viscosity dependence became weaker due to

the compatibilization of the EPDM. The processability of the blends is still acceptable according to the rheological results.

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