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HDPE/PA-6 blends: Influence of screw shear intensity and HDPE melt viscosity on phase morphology development

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Blending is an economically viable, versatile method of manufacturing new materials with a wide range of properties. The parameters most frequently targeted for the improvement by blending are: the mechanical properties, impact strength, processability, heat deflection temperature, and the cost-to-performance ratio.

When designing a blend, first the polymers and their approximate concentrations must be selected, then the most appropriate blend morphology for the envisaged application. For example, the need to improve impact properties implies that the minor phase ought to be dispersed as spherical droplets with micron or submicron diameter. On the other hand, if the material is to be used in vapor on solvent barrier applications, the minor phase should be dispersed in the form of relatively large, thin lamellas. The desired morphology is obtained by selecting the appropriate compatibilization and compounding/processing methods [1].

The studies on the morphology development of polymer blends in the single screw extruders [2] and twin screw extruders [3–5] demonstrated that significant changes in morphology occur in the melting zone and only comparatively minor changes take place in the following metering zone. The objective of the present work was to examine, using a single screw extruder combining with a convergent die, the effect of screw design of the metering zone on the morphology of HDPE/PA-6 blends with different HDPE melt viscosity.

Three commercial grades of HDPE were used as the matrix phase: HHM TR-144 (HDPE1), 60550AG (HDPE2), and HD5218EA (HDPE3). The HDPE1, HDPE2, and HDPE3, with a melt index of 0.18, 7.2, and 15 g/10 min, were manufactured by Phillips Petroleum Singapore Chemicals, Petrochina Dushanzi Petrochemical Co., and Petrochina Lanzhou Petrochemical Co., respectively. The PA-6 1030B manufactured by UBE Industries Ltd. was used as the dispersed phase. This PA-6 is a blow molding grade resin with a high melt viscosity. For the compatibilizer, Polybond 3009 manufactured by Uniroyal Chemical, a maleic anhydride grafted polyolefin copolymer, was used. The weight content of HDPE, PA-6, and compatibilizer is 85, 12, and 3% in the blend, respectively.

The melt shear viscosity measurements of individual resins were made using a slit die rheometer directly connected to an extruder with a screw diameter of 25 mm. The melt shear viscosity curves for three HDPEs and PA-6 at 240 °C are presented in Fig. 1. The corresponding melt viscosity ratio (VR) of the dispersed phase to the matrix phase is shown in Fig. 2. As can been seen, the VR for HDPE1/PA-6, HDPE2/PA-6, and HDPE3/PA-6 blends is about 0.46–0.94, 3.1–3.5, and 5.9–6.9 in the shear rate range of 20–400 s^{-1}, respectively.

The single screw extruder used has a screw diameter of 45 mm and a length-diameter ratio of 25:1. In order to investigate the influence of the screw elements in the metering zone on the morphology development of blends, the screw is constructed in a modular form. The core shaft of its metering zone can accommodate different screw elements to change the mixing type and intensity. Two different screw geometries, conventional metering and mixing screws, were employed. The mixing screw is outfitted with a fluted mixing element with a length of two times of screw diameter. Mixing screws in turn included two kinds, one with low shear intensity, and another with high shear intensity. If being approximatively estimated by the Couette shear rate, the shear rate in the channel of the metering screw, at a 40 rpm screw speed, is about 31 s^{-1}; in the barrier clearance of low and high shear intensity fluted mixing element, it is about 157 and 314 s^{-1} at the same speed, respectively.

The slit die mounted on the extruder exit to yield ribbons was a convergent one, having a 1.5 mm × 20 mm slit dimension.

The PA-6 and compatibilizer were dried at 100 °C for 12 hr and 85 °C for 4 hr in a oven, respectively. The HDPE and the dried components of PA-6 and compatibilizer were dry mixed thoroughly and then fed into the extruder. The extruder was operated at 185 °C in the feeding zone and at 240 °C toward the extrusion

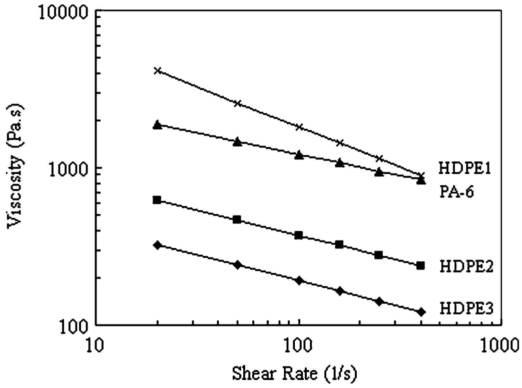

Figure 1 Shear viscosity vs. shear rate for three HDPEs and PA-6 at 240 °C.

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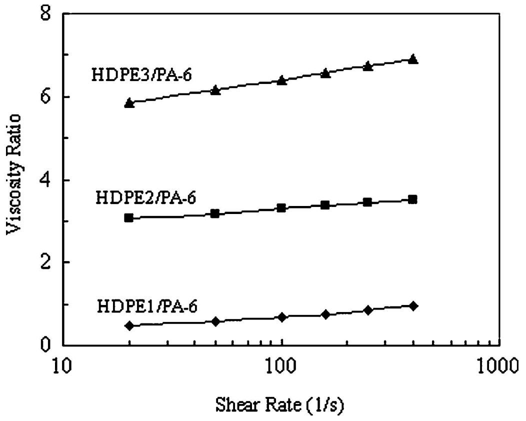

Figure 2 Viscosity ratio vs. shear rate for HDPE/PA-6 blends at 240 °C.

die. The screw speed was set at 40 rpm. The die was set at a temperature of 230 °C. The extruded ribbons were uncalibrated and cooled in water.

The observation of the morphology of the extruded ribbons was carried out on the surface in the center along the extrusion direction. The ribbons were freeze-fractured in liquid nitrogen and extraction of the PA-6 phase was performed by immersing them in formic acid for about 3 hr at room temperature. Later they were washed, dried, and subsequently coated with a conductive gold layer. The morphology was examined using a scanning electron microscope (SEM, Philips XL-30 FEG), operated at an accelerating voltage of 15 kV.

Fig. 3 compares the morphology of PA-6 phase in three different blends prepared by using different screw geometries. It can be seen that the type and shear intensity of the screw play a key role in the morphology, that is, the distribution and size of dispersed (PA-6) phase in the matrix (HDPE).

For the HDPE1/PA-6 blend, the morphology, shown in Fig. 3-a1, is some irregular when using a metering screw. Most of PA-6 phase exhibits spherical or ellipsoidal particles, some with small diameter, about 0.4 μm, and some with large diameter, about 1.5 μm. A few of PA-6 particles were stretched to big sheets, with a thickness of about 1.3 μm and a length of about 5 μm, along the flow direction. This result may be attributed to the non-uniform mixing in the metering screw. When the mixing screws involving shearing elements were used, the resulting morphology is more uniform, as shown in Figs 3-1b and c. In the case of the mixing screws with low and high shear intensity, the spherical PA-6 particles have an average diameter of about 0.35 and 0.25 μm, respectively. Some decrease in the domain size is due to the increased deformation and consequent break-up of PA-6 particles with increasing shear rate.

The above results can be elucidated as follows briefly. Shear stresses are very effectively transferred towards the dispersed phase and droplet break-up is enhanced with a highly viscous matrix. So, the minor component is finely dispersed.

For the HDPE2/PA-6 blend, clear laminar morphology with long PA-6 layers, some of which are extended

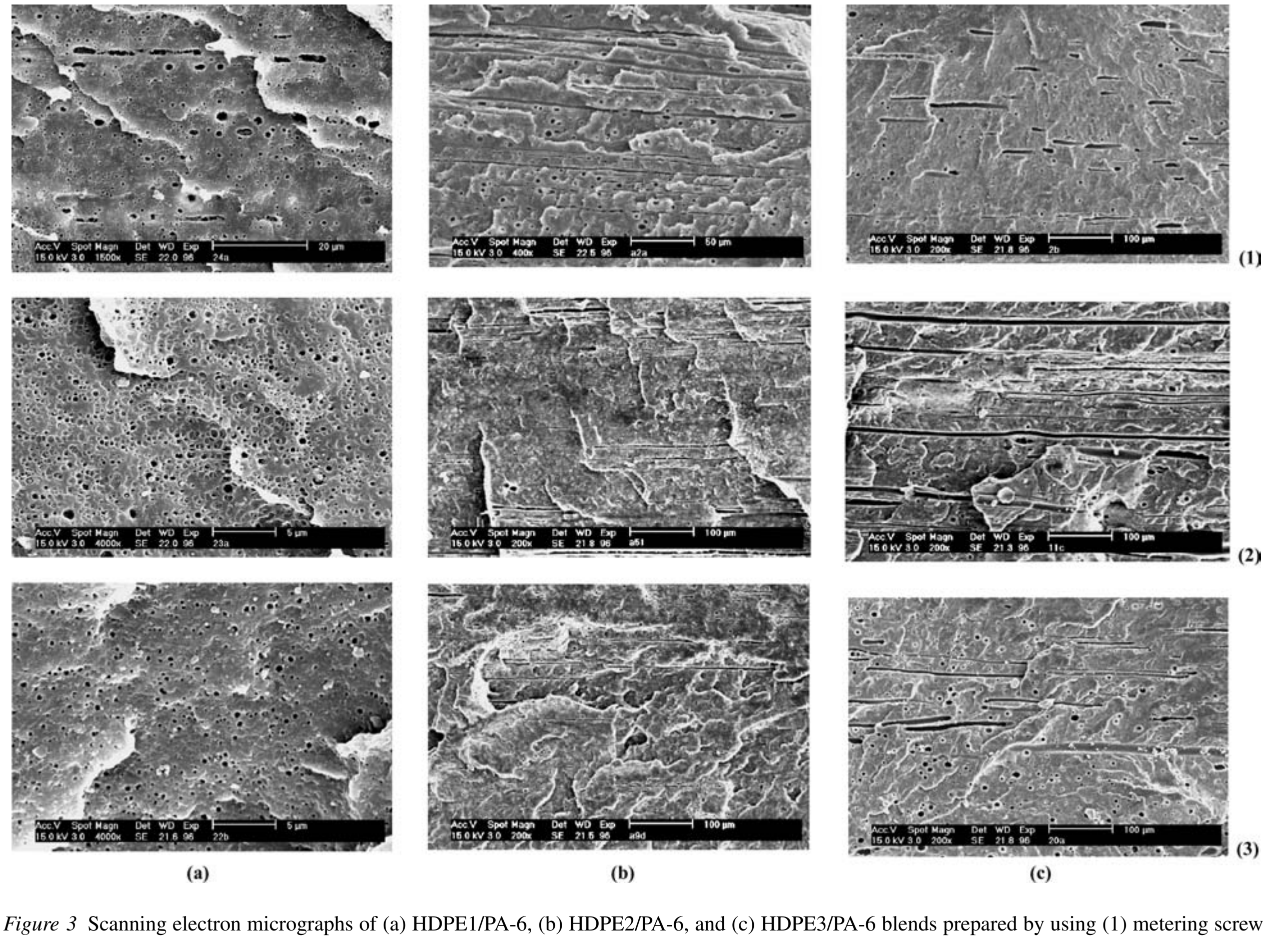

Figure 3 Scanning electron micrographs of (a) HDPE1/PA-6, (b) HDPE2/PA-6, and (c) HDPE3/PA-6 blends prepared by using (1) metering screw and mixing screws with (2) low and (3) high shear intensity.

to cover the whole micrograph, is yielded when a metering screw was used (Fig. 3-b1). Some small spherical PA-6 particles also exist in this case. However, the laminar structure diminishes gradually with increasing shear intensity in the mixing screw (Figs 3-b2 and 3).

For the HDPE3/PA-6 blend, no completely extended particles of PA-6 align in the flow direction when using a metering screw (Fig. 3-c1). Using mixing screws results in a laminar structure (Figs 3-c2 and 3). Pronounced laminar morphology with a large number of thinner and longer PA-6 layers, having an aspect ratio of about 100, is obtained in the case of the low shear intensity screw (Fig. 3-c2). Most of PA-6 layers are extended to cover the whole micrograph. However, the high shear intensity screw leads to a notable decrease in the length of PA-6 platelets (Fig. 3-c3). Some small spherical PA-6 particles also exist, especially in the case of the mixing screws with high shear intensity.

Contrary to what is often claimed and accepted in the literature [6–11], the VRs close to unity are more likely to produce laminar morphology, the above results reveal that it is feasible to yield a well-developed laminar structure from the HDPE/PA-6 blends with VRs larger than one by controlling the shear intensity of the screw. This can be explained as follows briefly. As the VR of the blend was increased, that is, the viscosity of the matrix phase was decreased, the shear stress imposed by the matrix to the dispersed phase decreased. Thus, higher shear intensity should be used to enhance the deformation and extension of the dispersed phase to yield a laminar structure.

In summary, immiscible HDPE/PA-6 blends were prepared by combining a single screw extruder with a convergent die, to examine the influence of the shear intensity in the metering zone of the screw and matrix phase (HDPE) melt viscosity on the phase morphology. The results showed a clear dependence of the morphology on the screw shear intensity and viscosity ratios (VRs). For the blend with a VR less than 1, PA-6 particles were dispersed as spherical droplets. For the blend with a VR between 3 and 7, it is possible to develop a laminar structure of PA-6 in HDPE matrix by controlling the screw shear intensity of metering zone. In other words, the screw shear intensity plays a distinct role in controlling the blend morphology. This is contrary to what is reported in some literature.

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