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A Comparative Study of Morphology and Structure related Properties of Saturated Olefinic Thermoplastic Elastomer Blends of EPDM/PP/Oil and SEBS/PP/Oil

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ABSTRACT : This work presents a comparative study of the morphology and structure related properties of thermoplastic elastomer blends based on SEBS/PP/Oil and dynamically vulcanized EPDM/PP/Oil. A combination of ruthenium oxide staining and low voltage scanning electron microscopy (LVSEM) was found to be suitable for the study of morphology of these highly oil extended blends. A close analogy was found in the mechanical, thermal and rheological properties of the two systems made in an internal Brabender mixer and co-rotating twin screw extruder. The morphology of the blends, as made by the two techniques, was found to be significantly different. In the case of TPVs, the blends made in the extruder had smaller EPDM domains and better tensile properties. In the case of SEBS, the blends made in the Brabender had more co-continuous phases and showed better tensile properties. Crystallization behavior of the isotactic polypropylene in the blends was found to be influenced by the type of rubber. Blends of SEBS/PP crystallized at a lower temperature than the TPVs. These differences were probably caused by differences in the nucleating ability of the two rubbers.

Keywords : thermoplastic elastomer, morphology, EPDM, PP, properties

Ⅰ. Introduction

Thermoplastic elastomers (TPE) based on blends of a semicrystalline isotactic polypropylene (iPP) and an amorphous rubber, particularly ethylene propylene diene terpolymer (EPDM) rubber, have shown an impressive growth in consumption over the last decades. The more so after the pioneering work on dynamic vulcanization by Coran, Das and Patel[1] and the discovery of the preferred phenolic resin curative system by Abdou-Sabet and Fath.[2] These blends also known as Elastomeric Alloys (EA), have a close resemblance in properties and applications to blends of iPP and polystyrene-block-poly-(ethylene-stat-butylene)-block polystyrene (SEBS). Both commercial blends usually contain a significant amount of paraffinic oil. The oil is added with two objectives: I) to enhance processability; II) to reduce the hardness.

The blends can be prepared on lab scale using batch internal mixers such as a Brabender Plasticorder, or on a large scale using co-rotating twin screw extruders. Although investigations, that use both Brabender and twin screw extruders to prepare

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the thermoplastic elastomer blends, are common, there are just a few studies[10-13] that directly discuss the blend morphology resulting from these two processing techniques. A major problem in this context is to characterize the morphology of these blends (due to a high Rubber/PP ratio and Rubber/Oil ratio) with common microscopic techniques. Morphological pictures are commonly difficult to interpret, because of the high rubber content of the blends. Additionally, the oil interferes with the experiment resulting in poor quality images.

The striking analogy in the properties of TPV blends based on iPP/EPDM/Oil and iPP/SEBS/Oil has not been studied before. In this paper, the morphology of iPP/EPDM/Oil elastomeric alloys is compared with the morphology of iPP/SEBS/Oil blends. The influence of mixing procedure - batch mixing versus continuous mixing on the blend morphology is reported as studied by different microscopic techniques. In this comparative study of the two blend systems, the compositions were kept similar as much as possible, to eliminate the possibility of morphological changes as a function of composition. A comparative study of the mechanical and thermal properties of these systems is also presented.

Ⅱ. Experimental

1. Materials

The polymers that were used in the present work were ethylene-propylene-diene terpolymer rubber (EPDM; DSM Elastomers B.V.; 63 wt % ethylene, 4.5 wt% 5-ethylidene-2-norbornene (ENB) termonomer) and extended with 50 wt% (100 parts per hundred rubber) of paraffinic oil; polypropylene (PP; DSM Polypropylenes B.V.) and styrene/(ethylene-butylene) based block copolymer (SEBS; Kraton™ Polymers). The PP used was an extrusion grade with a melt flow index (at 230℃, 2.16 kg) of 0.3 g/10 min. SEBS had a styrene to rubber ratio of 32/68 wt %. The oil used was a nonaromatic mineral oil containing paraffinic and napthanic hydrocarbons. For the EPDM formulation a phenolic resin was used as curing agent along with stannous chloride dihydrate ($SnCl_2.2H_2O$; Merck) as catalyst and zinc oxide (rubber grade; Merck) as acid scavenger during dynamic vulcanization. Additionally, stabilizers Irganox® 1076 (primary phenolic type antioxidant; Ciba Geigy) and Irgafos® 168 (secondary phosphite type antioxidant, Ciba Geigy) were used.

2. Melt blending

2.1 Brabender mixer

Ternary blends with a composition of EPDM- or SEBS- rubber/i-PP/Oil 28/33/39 (wt%) were prepared using a Brabender Plasticorder type- 350S with a mixer chamber volume of 390 ml and fitted with Banbury type rotors. To improve dispersion, the oil was preblended with the rubber. In the case of EPDM, the extra oil (40 parts per hundred rubber) was added on a two-roll mill. In the case of SEBS, the oil was added to and soaked up directly into the porous SEBS material.

To make TPVs, EPDM rubber with preblended oil was mixed with polypropylene at 180℃ and 80 rpm rotor speed. One minute after the melting of the polypropylene/stabilizers in the mixer, the EPDM/Oil preblend and zinc oxide was added. Four minutes later, curatives and the catalyst were added. Mixing was continued for another 5 mins after the last ingredient had been added. The complete cycle took 10 mins.

For making iPP/SEBS/Oil blends, the mixing time was increased to 30 mins. One minute after the melting of polypropylene/stabilizers in the mixer, the preblend of SEBS/Oil was added. Mixing was continued till torque consistency was obtained.

2.2 In Twin screw extruder

Extruded blends were prepared with a ZSK co-rotating twin screw extruder. Temperatures in the melt ranged from 200 to 260℃; the average residence time varied between 2 and 6 mins.

3. Compression moulding

After mixing the materials were compression molded at 200℃ for 3 mins under a pressure of 10 MPa and then cooled under pressure to about 30℃. The molded sheet thickness was about 2mm. The sheets were punched into dumbells for tensile measurements.

4. Sampling of blends for morphological characterization

After the blends had been compression molded, small samples were taken for electron microscopic examination. The samples were microtomed at - 130℃ using a diamond knife. Three different microscopic methods - Scanning electron microscope (SEM), Transmission electron microscope (TEM) and Atomic force microscopy (AFM) were used to study the morphology of the blends. For both SEM and TEM, the samples were stained after microtoming using a 1 % solution of ruthenium tetraoxide vapors. For TEM, thin sections (below 80 nm) were cut and vapor stained for 10 mins. For SEM the staining time was increased to 30 mins. The duration of the vapor staining was selected to only stain the rubber phase in both the blends.

5. Morphological characterization

The blends were first characterized by using conventional scanning electron microscope (Hitachi S800 FEG) after coating the microtomed surfaces with gold. The images obtained using secondary electrons and taken at an operating voltage of 3-6 keV were of poor quality due to sample charging. Hence, later on the measurements were made at 1 keV beam voltage using a LEO 1500 Scanning electron microscope (LVSEM) without the need for gold coating.

TEM measurements were made using a Philips CM 30 transmission electron microscope. Atomic force microscopy (AFM) measurements were performed in tapping mode using a Nanoscope III scanning probe microscope (Digital instrument). The instrument was equipped with a E-scanner. Commercially available Si_3N_4 cantilevers were used.

6. Mechanical properties

Tensile stress-strain curves were determined at room temperature according to ISO -37 using a Zwick Z020 universal testing machine equipped with a 5 N load cell. Crosshead speed of 50 mm/min was used to measure the E-modulus and 500 mm/min to measure the ultimate tensile strength and elongation at break. The reported results of each blend are the average of at least four measurements.

7. Thermal analysis

The thermal behavior of the blends was studied by means of a Perkin Elmer DSC 7 differential scanning calorimeter (DSC). The samples (about 5-10 mg) were heated at a rate of 10℃/min from 25℃ up to 200℃, maintained at this temperature for 5 mins. and then cooled down to - 100℃ at the same rate. At - 100℃ they were kept for 5 mins. and then reheated to 25℃ at a rate of 5℃/min. The crystallization temperatures and enthalpy of fusion of iPP were calculated from the cooling curve.

8. Rheological properties

Dynamic rheological properties were measured using a Rheometrics mechanical spectrometer (RMS 800) with parallel plate geometry. The blends were measured in a temperature range from 130 to 240℃. On these measurements time temperature superposition could be applied to create a master curve with 200℃ as reference temperature.

Ⅱ. RESULTS AND DISCUSSION

1. Morphology

Figure 1 shows the image artifact for the oil extended TPV made in the Brabender obtained with conventional SEM measured at a voltage of 3 kV. The picture is blurred due to sample charging caused

by the interaction of the electron beam with the oil that exudes from the sample during measurement in the vacuum environment. It might also be due to inelastic scattering of electrons induced by oil, caused by radiation damage at the high voltages. Such problems are common in SEM analysis of biological, hydrated or organic samples. Charging effects such as bright spots in the image arise due to build up of an electric negative charge on the specimen surface. This occurs at high acceleration voltages, if the number of electrons leaving the surface is not sufficient to compensate for the incoming ones. Charging can be avoided by applying a conductive coating or by working at low acceleration voltages (LVSEM), also known as crossover voltages, where the charges balance each other.

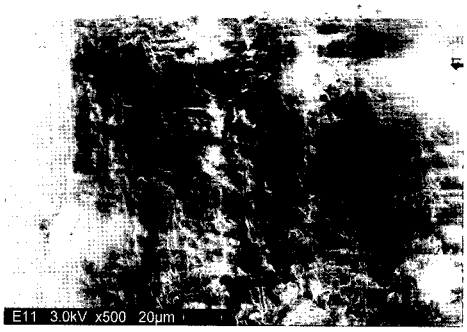


Figure 1. SEM image of Brabender made TPV, showing charging problem.

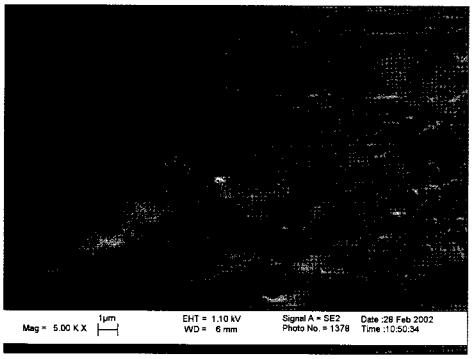


Figure 2a. LVSEM image of isotactic polypropylene.

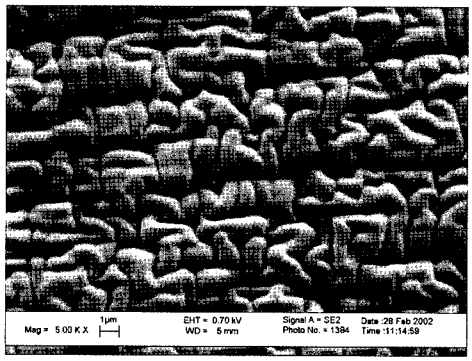


Figure 2b. LVSEM image of RuO_4 stained EPDM/Oil (50:50 w/w) blend.

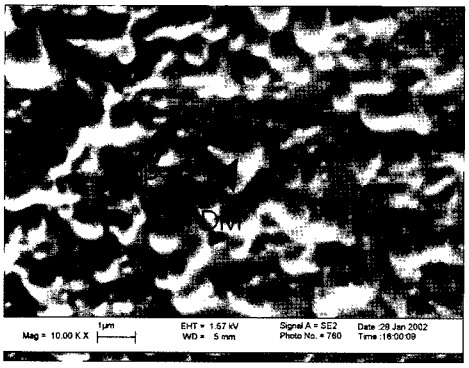


Figure 2c. LVSEM image of RuO_4 stained blend of TPV made in the extruder.

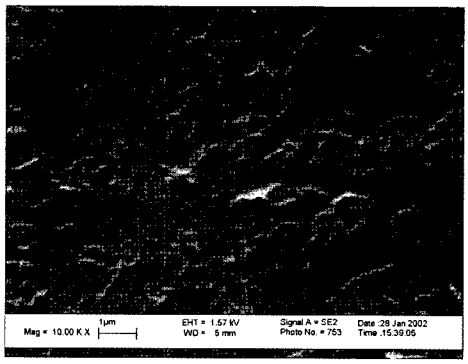


Figure 2d. LVSEM image of RuO_4 stained blend of TPV made in the Brabender.

LVSEM is a well-established technique for analysis of non-conducting polymers. Details of its operating principles and applications can be readily found in literature.[3-5] Because of the better performance of LVSEM, results of analysis of the morphology in TPV and SEBS blends using this technique are described below.

1.1 Morphological features of the TPV blends

LVSEM micrographs of iPP and EPDM/Oil (50:50 w/w) are shown in figures 2a and 2b respectively. The micrographs of the TPVs made with the extruder and Brabender are shown in figures 2c and 2d respectively. The rubber and the polypropylene phases in the blend can be identified after comparison with the SEM micrographs of the individual components. In both blends crosslinked rubber particles are dispersed in the polypropylene matrix. The disperse morphologies of the EPDM phase are consistent with those described by Coran[1], Sabet[2], Radusch[6] and many others for binary blends of dynamically cured EPDM/PP TPVs.

The particles in the figure 2c correspond to the EPDM phase and the smooth areas to the PP phase. When compared to the blend made in the Brabender, the particles are more spherical. It is difficult to judge using only SEM whether these particles are bumps or pits. The EPDM domains in the extruded sample have an average size of about 0.5 μm.

The EPDM phase looks different in the blend made in the Brabender (figure 2d). The PP matrix contains roughly elliptical shaped elastomeric domains, ranging in size from approximately 2 μm to 10 μm (in their larger dimensions). Most domains contain many sub-domains ranging in size from 0.1 μm-0.8 μm. The interface between EPDM and PP also looks different in the two pictures.

The LVSEM images shown above are compared with the TEM for these blends as shown in the figures 3a and 3b for the TPVs. The two-phase structure of the blends is also seen by TEM. The TEM image of extruded blends also show talcum particles, which were added to the extruded blends for free flowing of the ingredients. Unlike the LVSEM images the phase contrast in TEM is not as sharp, probably due to thick sections or insufficient staining. The continuity of the PP phase is clearly visible but the morphological differences seen with LVSEM between the two blends can not be confirmed. Further experiments to improve the

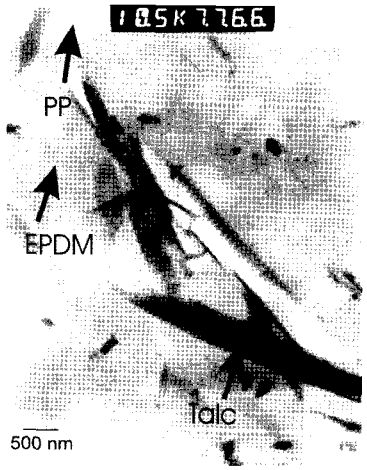


Figure 3a. TEM image of a RuO_4 stained blend of TPV made in the extruder.

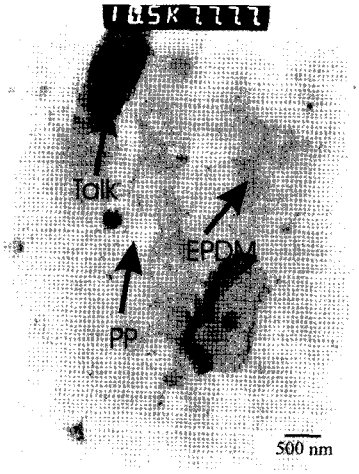


Figure 3b. TEM image of a RuO_4 stained blend of TPV made in the Brabender.

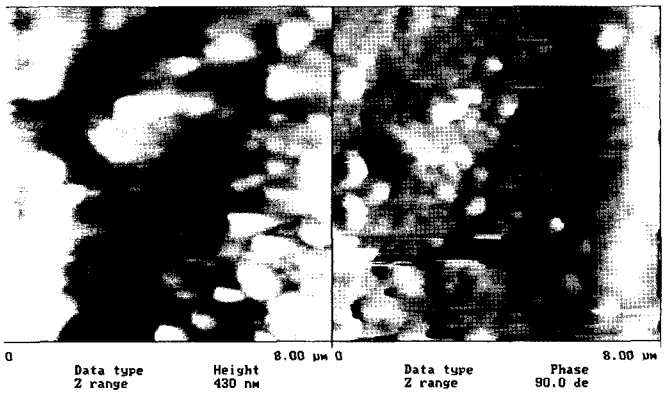


Figure 4a. AFM image of TPV made in the extruder. The image on left is the height image and the one on right is the phase image. In the height image, the white parts are higher.

Figure 4b. AFM phase image of TPV made in the Brabender.

contrast in these images by optimization of section thickness and staining time are in progress.

AFM images of the two blends are shown in figure 4a and 4b respectively. Although the pictures show artifacts due to contamination of the AFM-tip by oil, the morphological differences observed with LVSEM are reconfirmed. The darker grey areas correspond to the rubber phase and the white areas correspond to the PP. In both LVSEM and AFM images, more spherical rubber domains are seen throughout the PP matrix. Both techniques prove that the extruded sample shows better-dispersed rubber particles than the sample made in the Brabender. The extruded sample shows particles of roughly spherical shape and size around 1 μm in diameter, while in the Brabender sample the rubber particles are much bigger. From figure 2c it is difficult to determine whether the small round structures are bumps or pits. Since AFM data also contain the height information, the determination whether a feature is a bump or a pit is very straightforward. From the height image corresponding to the phase image shown in figure 4a it is concluded, that the round features in the SEM image (figure 2c) are bumps.

1.2 Morphological features of the SEBS blends

LVSEM micrographs of iPP and SEBS/Oil (50:50 w/w) are shown in figures 5a and 5b, respectively. The micrographs of the SEBS/PP/Oil blends made with the extruder and Brabender are shown in figures 5c and 5d, respectively.

The SEBS and the polypropylene phases in the blends can be identified after comparison with SEM micrographs of the individual components. The roughly elliptically shaped particles in figure 5c correspond to SEBS particles and the smooth areas in between correspond to the PP phase. The average domain size of SEBS is about 0.5 μm. Previous studies on these blends by Ohlsson, Hassander and Tornell[7] have shown that the three components SEBS/PP/Oil form co-continuous structures in the composition range from about 10 to 55% by weight of polypropylene. From the SEM image of the extruder made blend, the co-continuity of both phases can not be confirmed. Solvent extraction experiments are in progress to determine whether the phases are indeed co-continuous or disperse.

The blend made in the Brabender (figure 5d) shows more co-continuous SEBS and PP phases. The rubber domains look bigger when compared to extruded blends. This implies that the SEBS blends made in the internal mixer have a larger tendency to form a co-continuous morphology than blends made in the extruder. Interestingly, this observation is in agreement with the results of solvent extraction

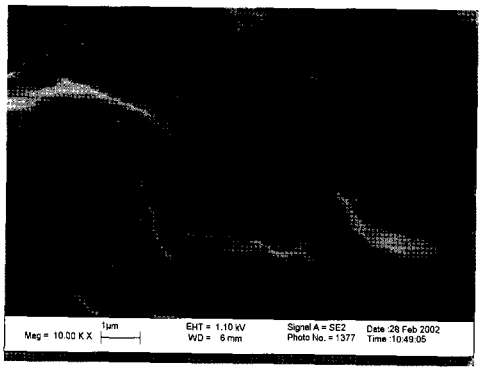


Figure 5a. SEM image of iPP.

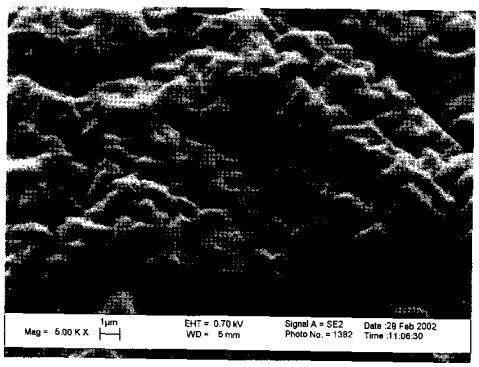


Figure 5b. SEM image of SEBS/Oil (50:50 w/w) blend.

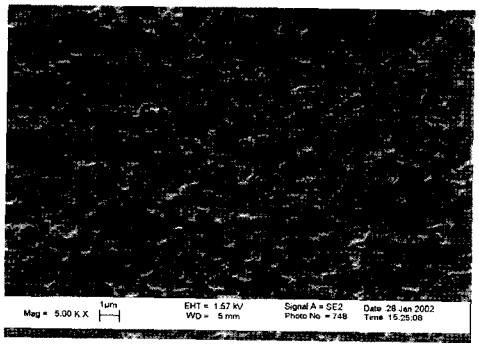


Figure 5c. SEM image of RuO_4 stained blend of SEBS/PP/Oil made in the extruder.

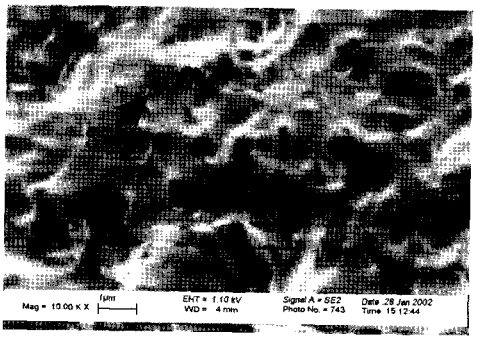


Figure 5d. SEM image of RuO_4 stained blend of SEBS/PP/Oil made in the Brabender.

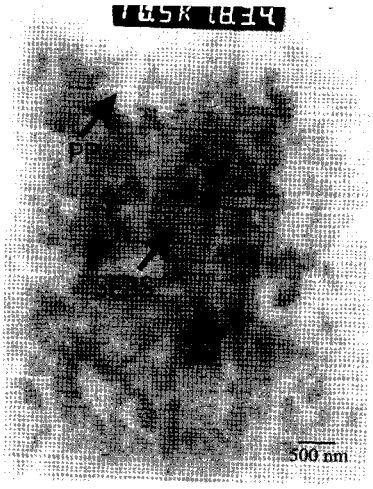


Figure 6a. TEM image of a RuO_4 stained blend of SEBS/PP/Oil made in the extruder.

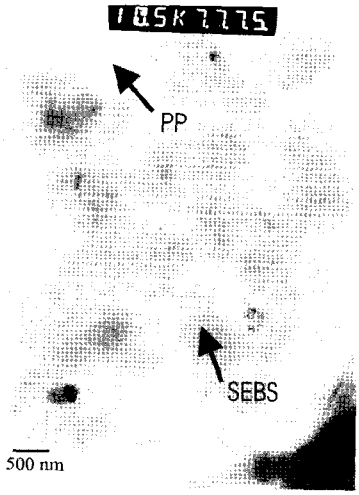


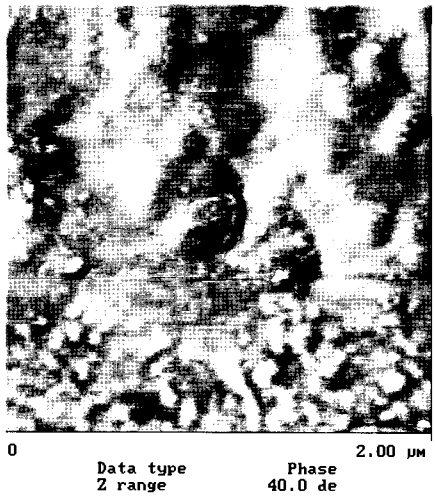
Figure 6b. TEM image of a RuO_4 stained blend of SEBS/PP/Oil made in the Brabender.

experiments as reported by Ohlsson et. al.[7].

Stained TEM micrographs of thin sections of these two blends are shown in figures 6a and 6b, respectively. The bright areas correspond to the polypropylene phase and the dark areas to the SEBS. These TEM micrographs again reveal the two-phase structure of these blends, but due to variations in sample thickness and staining the images are less informative than the LVSEM images.

AFM images of the two systems are shown in figures 7a and 7b respectively. The darker areas are the elastomer and the lighter areas are the PP. Com--parison of figure 7a with figure 7b shows several differences. First, the PP in figure 7a contains roughly spherical elastomer domains which indicates that the SEBS phase is less co-continuous in the blend made in the extruder than in the blend made in the Brabender (figure 7b). Furthermore, the size distribution of the dark SEBS domains is not uniform. In the extruded blend the particles are smaller (0.1-0.2 μm) and have more round edges than the blends made in the Brabender. Thus, using alternative microscopic techniques the various features in the phase morphology of SEBS/PP/Oil blends made in the extruder and in the Brabender do confirm each other.

Comparison of the blends obtained in the Brabender and extruder for both systems shows that the overall blend morphologies are very similar. The extruder made blends show smaller particles than the Brabender blends. Previous studies on various binary immiscible polymer blends by Scott et.al[10-11] and Sundararaj[12] have shown that the mechanism of morphology development in both devices is similar. In both mixing devices the polymers simultaneously undergo a complex combination of shear and extensional flow. During melting in both the twin screw extruder and the Brabender the dispersed phase (the phase that melts later) is stretched into sheets. These sheets develop into cylinders and the cylinders ultimately break into spherical droplets via Rayleigh type instabilities. At high shear rates in the twin screw extruder, the sheets are thinned out and breakup into small particles by hole formation, whereas at low shear rates in the Brabender the sheets are stretched out into cylinders or a long ribbons and cannot be broken up efficiently. This explains why smaller and more round particles are obtained in the case of blends made in an extruder.



Figue 7a. AFM image of SEBS/PP/Oil blend made in extruder.

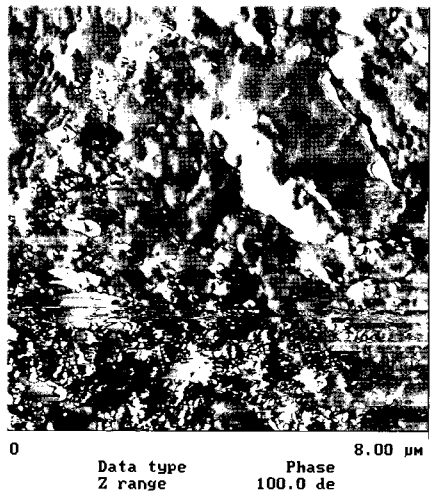


Figure 7b. AFM image of SEBS/PP/Oil blend made in Brabender.

2. Mechanical Properties

The differences in the stress-strain behavior of the samples are shown in figure 8. The differences, although small, are reproducible and can be related to the morphology of these blends. In case of TPVs, the blends made in the extruder reaches higher tensile strength at the same elongation at break as compared to the blends made in the Brabender. One reason for the high tensile strength of the blends made in the extruder could be a smaller particle size of the EPDM phase. The increase in tensile strength with decrease in particle size of the EPDM phase in dynamically vulcanized PP/EPDM blends has been reported before.[1] In case of SEBS, the blend made in the Brabender having a more co-continuous phase structure shows a somewhat better property.

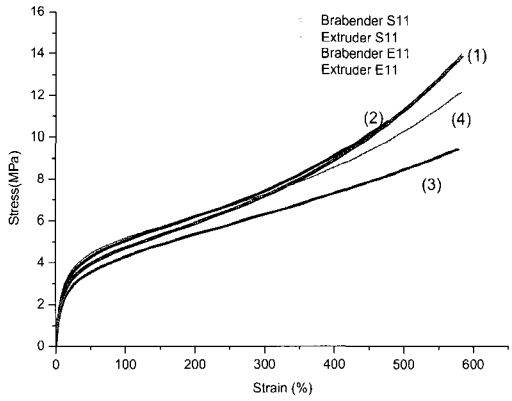


Figure 8a. Stress-strain curves. (1) Brabender made SEBS/PP/Oil blends (2) Extruder made SEBS/PP/Oil blend (3) Brabender made TPV (4) Extruder made TPV.

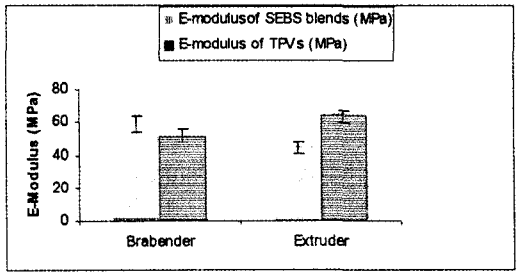


Figure 8b. E-moduli of SEBS/PP/Oil blends and TPVs.

The E-moduli of the blends are shown in figure 8b. Opposite behaviors are seen for TPV and SEBS blends. For the TPVs, having disperse phase morphology, the blend made in the extruder and having smaller EPDM domains, exhibits a higher modulus than the blend made in the Brabender. In SEBS blends, having predominantly co-continuous phase morphology the blends made in the Brabender exhibit the higher modulus. This must be due to more co-continuity between the two phases in the Brabender blend, which results in a higher resistance to deformation during tensile measurements. Further studies aimed at understanding the deformation behavior of the two phases and its relationship with morphology are in progress.

3. Thermal properties

3.1 Melting and crystallization

The melting temperature (T_m) was taken from the first heating curve of DSC and the crystallinity index X_c of the blends was calculated by applying the following formula:[8]

$$X_c(iPP) = \frac{100\Delta H^*(iPP)}{\Delta H^0(iPP)}$$

$$X_c(blend) = \frac{100\Delta H^*(blend)}{\Delta H^*(iPP)}$$

where $\Delta H^*(iPP)$ is the apparent enthalpy of fusion per gram of the iPP in the blend; $\Delta H^0(iPP)$ is the heat of fusion per gram of 100% crystalline iPP (202J/g)[8] and $\Delta H^*(blend)$ is the apparent enthalpy of fusion per gram of the blend. The values are reported in Table 1.

The iPP melted in presence of rubber and oil in both blends exhibits almost the same values of T_m, which is lower than the T_m obtained for pure iPP (164℃). Such a noticeable difference in T_m values is in agreement with results obtained in previous work[8] studying iPP/EPDM blends, in which the EPDM was cured with peroxide. This can be due

Table 1. Enthalpy of fusion per gram of iPP in the blend (J/g), enthalpy of fusion per gram of blend, crystallization temperature, melting temperature and crystallinity for blends made in the Brabender and extruder.

Sample		ΔH* (blend) (J/g)	X_c (blend) (%)	ΔH* (iPP) (J/g)	X_c (iPP) (%)	T_c (℃)	T_m[1] (℃)
TPV	Brabender	28.5	14	85.71	41	107	157
	Extruder	32.5	16	97.51	47	107	157
SEBS/PP/ Oil	Brabender	28.9	14	86.78	42	99	156
	Extruder	32.3	15	96.90	46	98	158

[1]Taken from 1st heating curve

to a decrease in lamellar thickness of the PP crystals and/or defects in the PP lamellae. The X_c (blend) values are close in all the blends. The X_c (iPP) values of the blends made in Brabender are higher than the blends made in the extruder. It is interesting to see that when iPP crystallizes in the presence of cured EPDM/Oil in the TPV blends, the crystallization temperature remains the same as observed in pure iPP (108℃) but when iPP crystallizes in presence of SEBS, the crystallization peak shifts to lower temperature. This indicates that the cured EPDM particles have more nucleating ability during the crystallization process compared with the SEBS. Further experiments aimed at understanding the crystallization behavior of iPP in both the blends are in progress.

4. Viscoelastic behavior

The complex viscosity for the blends is shown in figure 9 as a function of angular frequency. The curves of the ternary blends are sandwiched between the binary blends of Rubber/Oil and PP/Oil.

The viscosity of the ternary blends contains contribution from both the PP/Oil phase and the corresponding rubber/Oil phase. The curves are separated from each other at low frequencies but approach each other at high frequencies. This means that at high frequencies the viscosity is determined by the phase with low viscosity (iPP) and at low frequencies the viscosity is determined by the phase with high viscosity (rubber phase). The TPV made in the extruder shows higher viscosity at low frequencies, which must be related to its morphology. The TPV blends behave as a suspension. Taylor[9] showed long ago that this would lead to a viscosity increase. No significant differences were observed between the other blends.

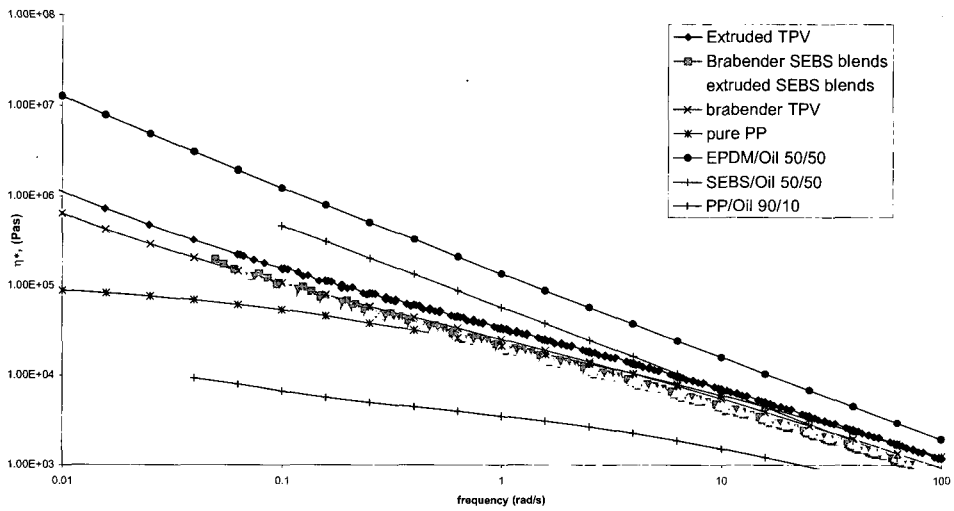


Figure 9. Variation of complex viscosity with angular frequency for TPV and SEBS blends.

Ⅳ. Conclusions

A comparative study of the blend morphology of ternary thermoplastic elastomer blends of EPDM/PP/Oil and SEBS/PP/Oil by different microscopic techniques shows that the method of RuO_4 staining and LVSEM imaging is well suited to the analysis of phase morphology.

The mechanical, thermal and rheological properties of the blends made in a Brabender internal mixer and an extruder are very much comparable. This shows that significant differences in morphology are obtained by making the blends in a Brabender or twin screw extruder. Differences exists in particle shape/size (in TPV blends) and in the degree of co-continuity of the phases (SEBS blends). These differences are small but reproducible and can be correlated to their mechanical properties.

The crystallization temperature of iPP in TPV blends remains the same as that of pure iPP but in it drops to a low value in SEBS blends. This proves that cured EPDM particles in TPV blends have more nucleating ability during crystallization as compared to the SEBS.

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