

Melt Blending of Linear Low-Density Polyethylene and Polystyrene in a Haake Internal Mixer. I. Compatibilization and Morphology Development

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SYNOPSIS

Morphology development was investigated during blending of linear low-density polyethylene (LLDPE) and polystyrene (PS) (the minor phase) in a Haake internal mixer. In addition, the blending was studied using 5 wt % Kraton styrene-ethylene/propylene (SEP) diblock or styrene-ethylene/butylene-styrene (SEBS) triblock copolymers as compatibilizing agents. Scanning electron microscopy (SEM) examinations of blends show a bimodal particle size distribution of the minor phase during the initial stage of morphological development. Morphology observations suggest that the main function of the compatibilizer during blending lies in reducing the interfacial tension between two immiscible polymer phases. The addition of the copolymer reduces the average domain size from a micron to a submicron range. Compatibilization mechanisms of the copolymers with the immiscible blends are proposed. The superiority of SEBS triblock over SEP diblock as a stabilizer of the morphology was shown. © 1995 John Wiley & Sons, Inc.

INTRODUCTION

Polymer blends play an important role in the modern polymer industry not only for the development of new materials but also for practical recycling. Most polymer blends are incompatible, and the final performance of a polymer blend is determined by the polymer compatibility and the phase morphology. Many workers have shown the beneficial effects of a commercial block copolymer, Shell Chemical's Kraton G, on the morphology and mechanical behavior of blends of polyolefin and polystyrene.^{1,2} The extent of improvement was found to be dependent on the molecular weight, the composition, and the molecular structure of the copolymer. Fayt et al.^{3,4} have pointed out that the molecular structure is a key criterion when designing the most efficient emulsifier.

In the past, it has been well documented how the final blend morphology influences the performance

of the blend.^{5,6} Studies of the way that phase morphology evolves during blending, however, are quite limited. In 1991, Scott and Macosko⁷ first reported the results of model experiments which investigated morphology development during the initial stages of mixing. The authors concluded that the primary mode of morphological development at short mixing times appeared to be the shearing of the phases into ribbon or sheetlike structures followed by shear and interfacial tension-driven breakup of these structures. Most reduction in the dispersed phase size was demonstrated to occur in the very early stages of mixing.

The objective of this study is to better understand and achieve control of the development of phase morphology during blending. Two means were used to control the morphology: (1) introduction of compatibilizers and (2) variation of blending parameters. In particular, it should be noted that the morphology being referred to is usually of a dynamic character (size, shape of the minor phase), as a result of the equilibrium between the breakup and coalescence of the dispersed/deformed domains in the flow field.

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This study consists of two parts. In the present part (Part I), the morphology development during both noncompatibilized and compatibilized blending of polystyrene (PS) and linear low-density polyethylene (LLDPE) was investigated. In order to acquire a more detailed understanding about the physics of the morphology development during polymer blending, both the reduction in the average domain size and the complexity of the domain-size distribution were studied. A styrene-ethylene/propylene (SEP) diblock or a styrene-ethylene/butylene-styrene (SEBS) triblock copolymer was used as the compatibilizer for the immiscible PS-LLDPE blends. Copolymers SEP and SEBS have the same proportion of the styrene block to the rubber portion (30/70). The diblock as well as the triblock were chosen to demonstrate the dependence of compatibilization upon the molecular structures of the copolymers. Through the investigation of the evolution of the blend morphology, an attempt was made to understand the compatibilization mechanisms by which these copolymers function when incorporated in a blend of incompatible polymers. It is also of interest to examine the influence of the mixing sequence on the resulting blend morphology.

The detailed study of the morphology-processing relationship during blending is reported in Part II of this study.⁸

EXPERIMENTAL

Materials

The LLDPE used in this study is an injection-molding grade (SCLAIR 2107), supplied by DuPont of Canada. The PS, Styron 680, was obtained from the Dow Chemical Company. Two Kraton elastomers used as the compatibilizers are SEBS low-molecular-weight triblock (G1652) and SEP high-molecular-weight diblock (G1702) copolymers, supplied by the Shell Company. The molecular weights of the polymers are listed in Table I.

Blending

Three blend compositions were investigated. One was 80% LLDPE/20% PS (in wt %), the other two were 76% LLDPE/5% SEP/19% PS and 76% LLDPE/5% SEBS/19% PS. All of the blends were prepared using the Haake melt blender. The temperature was set at 180°C for the three heating sections. Roller blades were operated at 50 rpm. The total mass of material charged to the mixer was 45.5

Table I Molecular Weight of Polymers

Polymer	M_n^a
LLDPE	69,800
PS	200,000
S-EP	45,000/115,000
S-EB-S	7,000/37,500/7,000

^a Provided by the suppliers.

g, which corresponded to 70% of the mixer capacity. This mixer loading has been shown to produce the most uniform mixture.⁹

Pellets (or powders) of the blend components were premixed before introduction into the mixer. At the start of the mixing process (zero time), the dry mixture was introduced through a chute into the preheated mixing chamber. After a specified mixing time the mixer was stopped, a sample was taken from the top of the mixer between the two blades and immediately immersed in ice water to freeze the morphology.

In addition to the one-step mixing procedure mentioned, two additional blends were made using a two-step mixing procedure on ternary blends of 76% LLDPE/5% SEP/19% PS at the same operation conditions. One blend was prepared by adding SEP into the mixer after 1.5 min of mixing of LLDPE and PS. In the other blend, the time for addition of SEP was 5 min after mixing of LLDPE and PS.

Characterizations of Phase Morphology

A scanning electron microscope (SEM), Jeol 5400, was used for examining the phase morphology. Fracture surfaces of samples were prepared in liquid nitrogen. In order to increase the contrast and observe the matrix and the dispersed phases, toluene was used as a dissolution etchant to remove the PS phase from the LLDPE phase of the blend. These samples were then coated with a gold/palladium alloy before being examined in the SEM.

MORPHOLOGICAL OBSERVATIONS

Noncompatibilized Blends of 80% LLDPE/20% PS

The mixing times investigated were 1.0, 2.0, 2.5, 3, 4, 5, 10, and 20 min. By definition, zero time corresponds to the time of introduction of pellets to the mixer chamber.

The SEM photographs of the blend morphologies after various mixing times are shown in Figure 1. The final morphology consists of the minor phase, PS, dispersed as droplets in a matrix of LLDPE. During mixing LLDPE melts first. After 1 min of mixing, large PS regions are still present within the PE matrix. After 2–3 min of mixing, the structure contains many highly elongated particles, as well as some nearly spherical particles. After a mixing time of 3 to 4 min, there exists a bimodal size distribution of PS particles; one of about 200 μm , and the other of about 2 μm . The number of 200- μm PS particles decreases with additional mixing. After 5 min, nearly all the particles are of a size of the order of 2 μm . Even with further mixing time, no more significant breakup is achieved. After 20 min of mixing, the roller blades were stopped and the mixer chamber removed. Samples from three mixer locations—between two blades (or pool), behind the blade (wing), and the gap between the mixer wall and the blade—were investigated. The SEM examination of the blend morphology indicates that there are no differences in PS particle size associated with sampling location in the mixer.

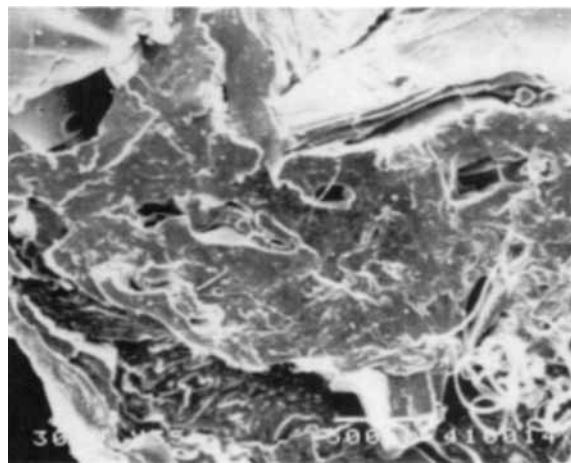
Compatibilized Blends of 76% LLDPE/ 5% SEP/19% PS

The examination of the phase morphology showed that the final morphology during blending 20% PS and 80% LLDPE develops within 5 min of mixing when blending at 180°C and 50 rpm. In addition to reporting the effect of adding compatibilizers at the beginning of the mixing upon morphology development, this section also examines the effect of adding compatibilizer at different stages of the mixing process upon morphology development.

Case 1. Adding SEP at the Same Time with the Blend Components ($T = 0$)

The morphology was examined after 1.5, 3, 5, 10, and 20 min of mixing. The SEM micrographs shown in Figure 2 indicate that the blend morphology was modified through the addition of SEP. A higher degree of dispersion was observed upon the addition of SEP into the blend.

For blends containing compatibilizer, we observe small PS particles of 200 μm as well as larger PS particles at a mixing time of 1.5 min [see Fig. 2(a)]. A small quantity of particles about 2 μm in diameter was observed in this blend. When mixing time was increased to 2–4 min, a bimodal size distribution of



a

Figure 1 Morphology development of 20% PS/80% LLDPE blends at the different mixing time in the Haake mixer. (a) Mixing of 1 min, (b, c) mixing of 2 min. A bimodal size distribution of the dispersed PS droplets is shown on the micrographs of the two magnifications. (d, e) Mixing of 5 min. The reduction in the number of the droplets is observed.

the PS droplets with a smaller average diameter was observed. The primary mode of the morphology development (or droplet breakup) in the PS/SEP/LLDPE blend, seemed to be qualitatively the same as that for the PS/LLDPE noncompatibilized blend. The phase morphology observed for blends mixed between 5 and 20 min was very similar. The final morphology was essentially developed within 5 min of mixing with the average domain size of the dispersed phase in the submicron range.

The addition of the SEP copolymer not only contributes to a higher degree of dispersion, but also modifies the interface between the PS particles and LLDPE matrix. This improved adhesion between the two immiscible phases is substantiated by the more ductile fracture surface shown in the SEM micrograph [Fig. 2(e)]. Compared with the compatibilized blends, the interfacial boundary between the particles and the matrix in the immiscible blends is very distinct. Although a direct morphology examination of the location of SEP in the PS/LLDPE blends was not attempted in this study, Fayt et al.¹⁰ have demonstrated that, in melt blends of 20% PS and 80% LDPE with the poly(hydrogenated butadiene-*b*-styrene) copolymers (HPB-*b*-PS), the copolymer forms a continuous layer around the dispersed particles of PS. Thus, the brighter layer around the PS particle

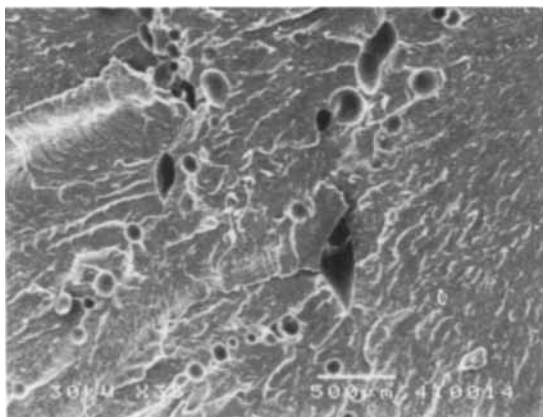


Fig. 1b in low magnification

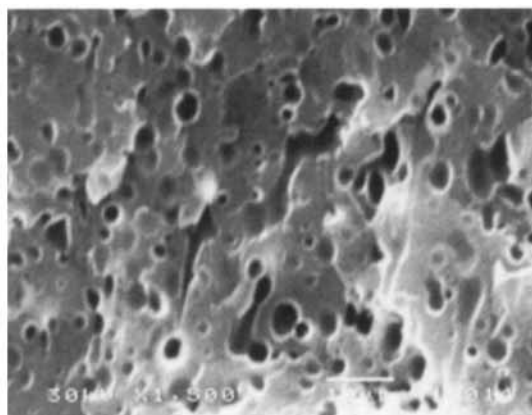


Fig. 1c in high magnification

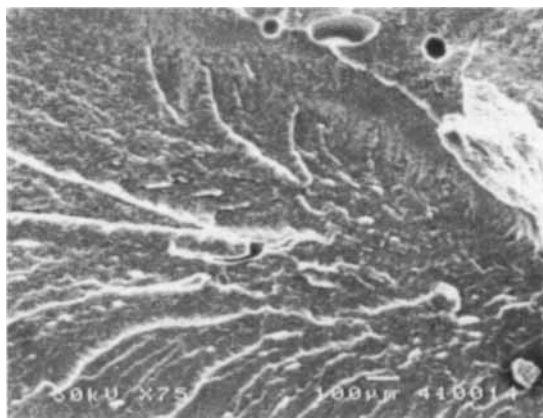


Fig. 1d in low magnification

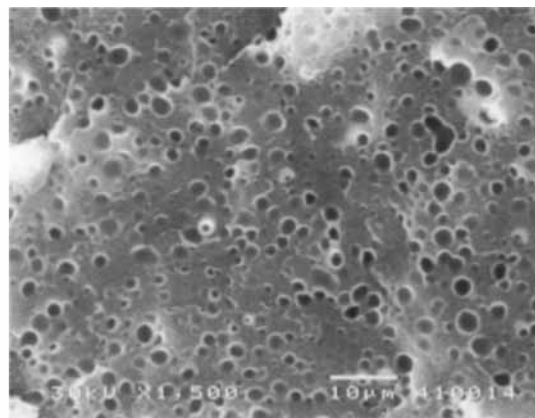


Fig. 1e in high magnification

Figure 1 (Continued from the previous page)

shown on Figure 2(f) may be an indication of the presence of a third phase.

Case 2. Adding SEP During the Period of Morphology Evolution ($T = 1.5$ min)

SEP was introduced into the mixer chamber after 1.5 min of mixing of LLDPE and PS. The morphology was investigated after 2.5, 4.5, 6.5, and 11.5 min of mixing. After 2.5 min of mixing, there was a greater breakup of PS when SEP copolymer was present [see Fig. 3(b)]. More elongated droplets were observed when copolymers were not present [Fig. 3(a)]. The addition of SEP increases the extent of dispersion of PS in LLDPE with a uniform

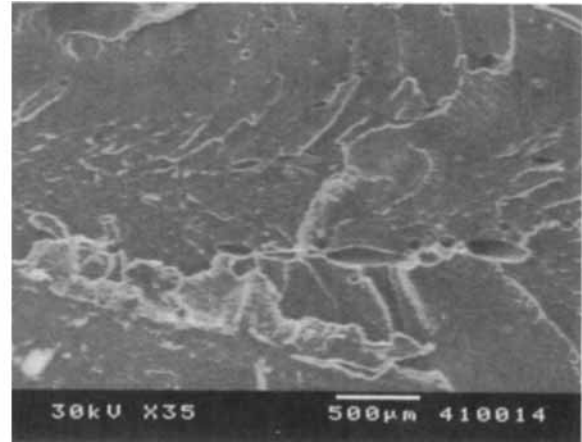
dispersion of the PS particles achieved in 6.5 min of mixing. No further breakup of the PS phase was observed at 11.5 min of mixing.

Case 3. Adding SEP after the Final Morphology Developed ($T = 5$ min)

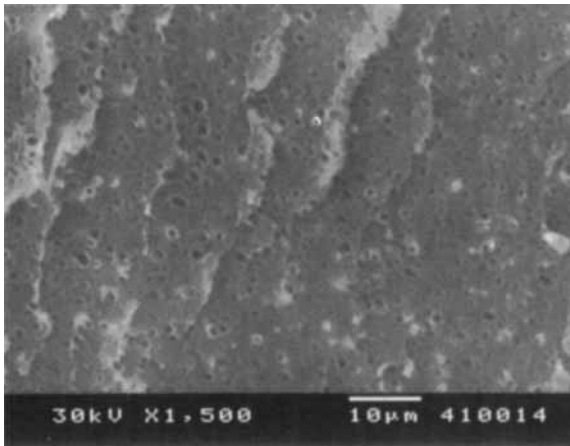
SEP was fed into the mixer after 5 min of mixing of LLDPE and PS. The morphology was investigated at 6, 8, 10, and 15 min. It is believed that addition of copolymer strongly modified the interfacial adhesion between the phases [see Fig. 4(a)]. In fact, at 8 min, the morphology consists of an interlocked structure [Fig. 4(b)] as well as the dispersed phase behavior. At 10 min, the dispersed



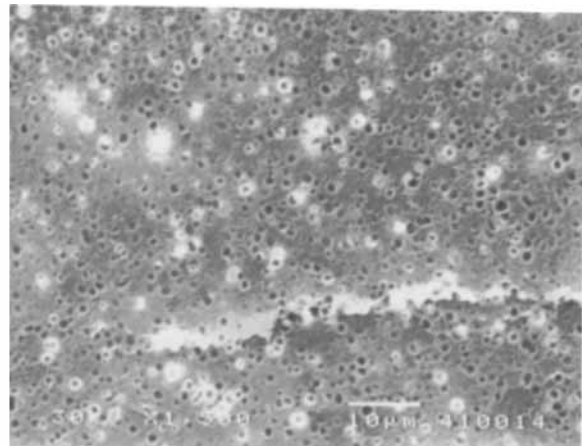
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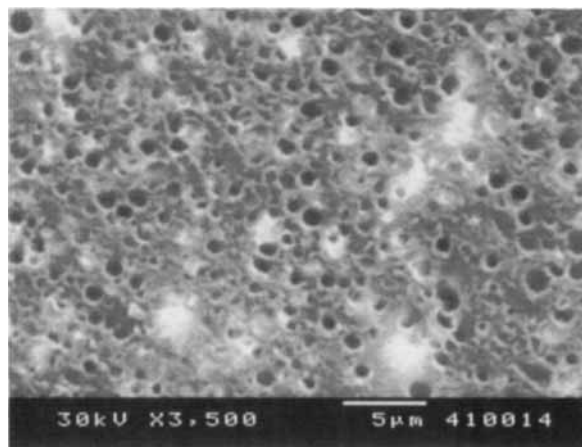
Figure 2 Morphology development of the compatibilized blends of 19% PS/76% LLDPE with 5% SEP. SEP was added into the blend at $t = 0$: (a) mixing of 1.5 min, (b, c) mixing of 3 min, (d) mixing of 5 min, (e) mixing of 20 min, (f) mixing of 1.5 min. The brighter layer is the indication of the SEP phase.

phase contained uniform size particles. Again, no further size reduction of the PS phase was observed after 10 min of mixing.

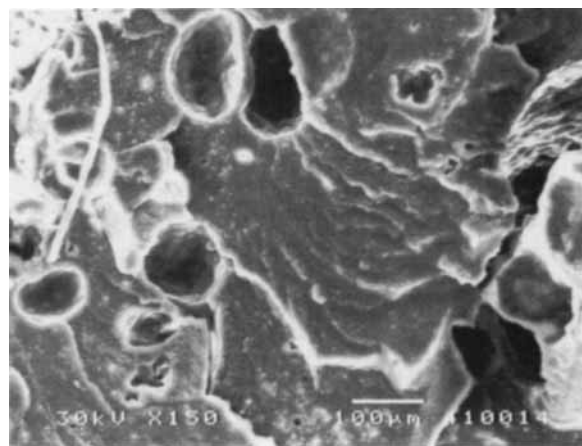
Compatibilized Blends of 76% LLDPE/ 5% SEBS/19% PS

The morphology was investigated after 1, 2.5, 4.5, 7, and 10 min of mixing time. The SEM photographs

of the blend morphology are shown in Figure 5. Breakup of PS into small particles of $200 \mu\text{m}$ was observed immediately after the mixing process began (time = 1 min). A bimodal particle size distribution of PS developed at the initial mixing stage was reduced to the submicron unimodal range during 4.5 min of mixing. The high compatibility of PS/LLDPE blends on addition of SEBS is indicated by the dramatic reduction of the dispersed phase scale,



e



f

Figure 2 (Continued from the previous page)

as well as the diffused interfacial boundary between the two blend components. The addition of SEBS triblock copolymer to PS/LLDPE blends stabilizes the dispersed droplet size to a finer scale than that for the SEP diblock copolymer.

The observed morphology indicates that SEBS tends to interlock the dispersed phase with the matrix and aids in forming the phase structure of a co-continuous interlocking, or interpenetrating, network, i.e., one in which a continuous phase of one

polymer fills the voids in a continuous phase of the second polymer.

RESULTS AND DISCUSSIONS

Experimental observations indicate that melt dispersion begins at a very early stage in the mixing process. In pellet-pellet mixing, because of the poor thermal conductivity of polymers, the initial melting of pellets occurs first at the heated surface. At the same time the solid pellets are subjected to a very intense electric field. Under these conditions dispersion seems initially to take place by an abrasive process involving local stretching of small domains at the heated surface to the point where they rupture and thus become particles. In the particular blending system used with PS and LLDPE, it is believed that very early dispersion is also associated with the abrasion of the edges of brittle PS pellets against the mixer wall.

On the basis of the morphologies that were observed as mixing time increases, a mechanism of morphology development during blending PS and LLDPE at short mixing times (illustrated in Fig. 6) is proposed. Brittle PS solid pellets are initially abraded against the mixer wall while LLDPE melts. Melting associated with abrasion of PS resulted in a bimodal size distribution of PS particles during the initial stage of mixing. Later in the mixing process, deformable PS particles as well as the melt become stretched into many strips near the mixer wall. Due to interfacial and shearing forces, the strips are reduced into irregularly shaped particles and finally become nearly spherical particles. For blends of 20% PS and 80% LLDPE at 180°C and 50 rpm, the final equilibrium morphology is reached within 5 min of mixing.

The complex cyclic shear in the Haake mixer causes breakup of PS into microdispersed particles from 4000 to 2 μm in LLDPE matrix at the very beginning of mixing. Once the final equilibrium droplet size is reached, no more significant breakup occurs during further mixing. One possible reason for this behavior is that the PS droplets appear to be less deformable than equivalent viscous Newtonian droplets, and this resistance to shear breakdown makes intensive shear mixers (e.g., Haake) ineffective after a critical mixing time. Moreover, when the difference in elasticity between the blend components is large and the dispersed phase forms droplets, neither low shear nor high shear mixing is satisfactory, inasmuch as the droplets resist deformation. In such cases it is better to alter the motion of

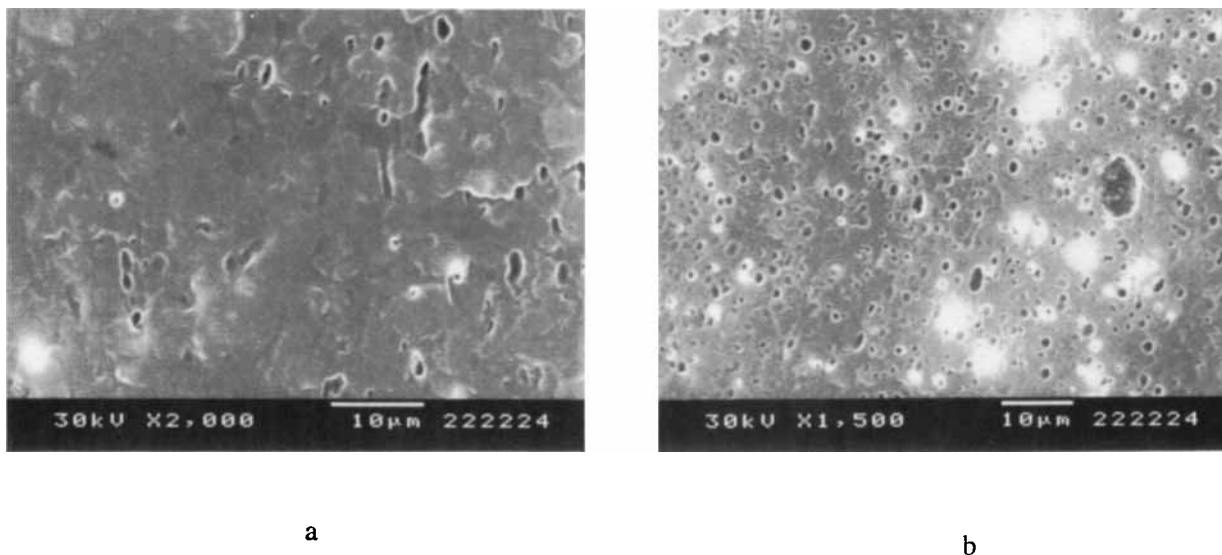


Figure 3 Morphology development of the compatibilized blends of 19% PS/76% LLDPE with 5% SEP. SEP was added into the blend at $t = 1.5$ min: (a, b) mixing of 2.5 min.

the matrix rather than increase the shear forces. A liquid droplet may still be drawn down into a fiber by an elongational type of flow, and subsequent shearing may cause fiber breakup.

It is demonstrated that the primary mode of morphological development (or droplet breakup) for the compatibilized LLDPE/SEP or SEBS/PS blend is the same as that for the noncompatibilized LLDPE/PS blend. Scott and Macosko¹¹ investigated morphological development during nonreactive and reactive polymer blending. These authors concluded

that the initial morphological development during blending of nonreactive PS/nylon (PA) is qualitatively the same as that for the PS-Ox/PA reactive blends.

The principal function of the compatibilizer lies in reducing the interfacial tension between the two polymer phases. As an interfacial agent locates at the interface, the stress is more readily transferred from the continuous phase to the droplets, which results in increased dispersion. The observation of small PS particles of $200 \mu\text{m}$ at a very early stage

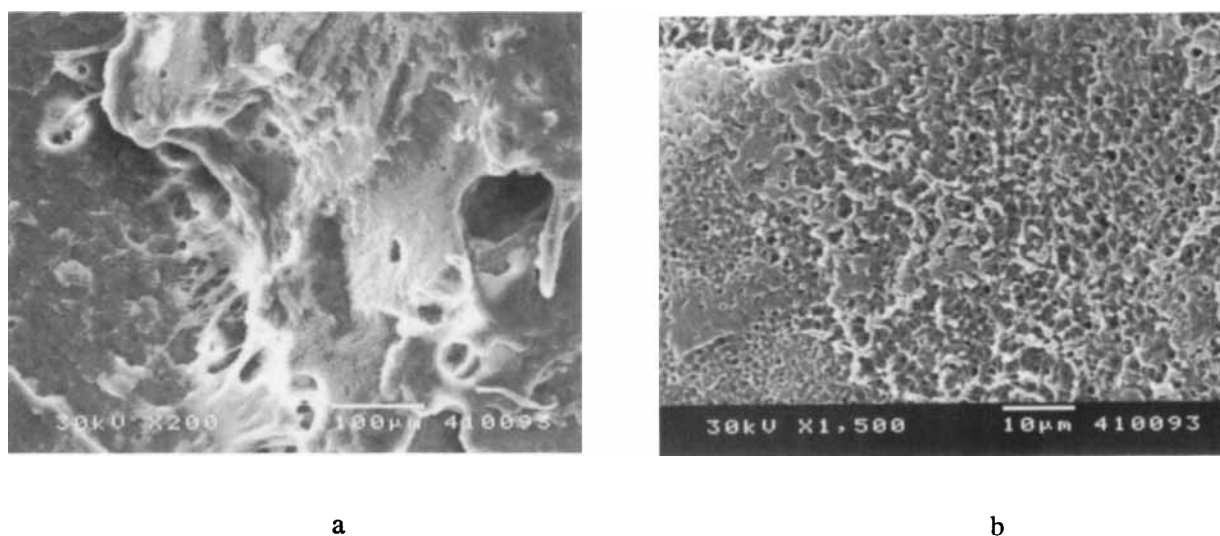
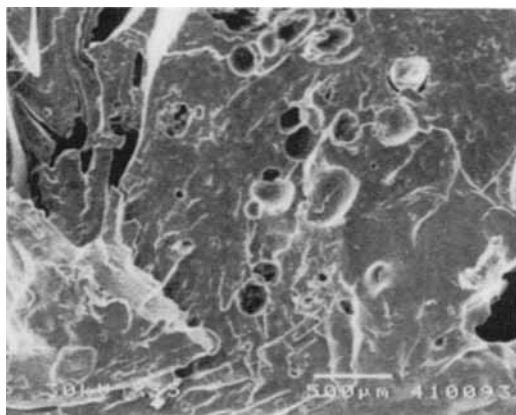
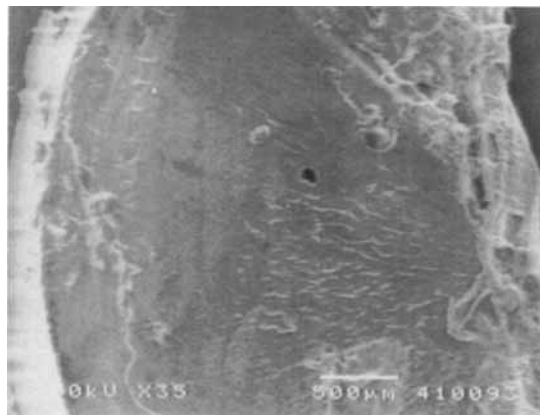


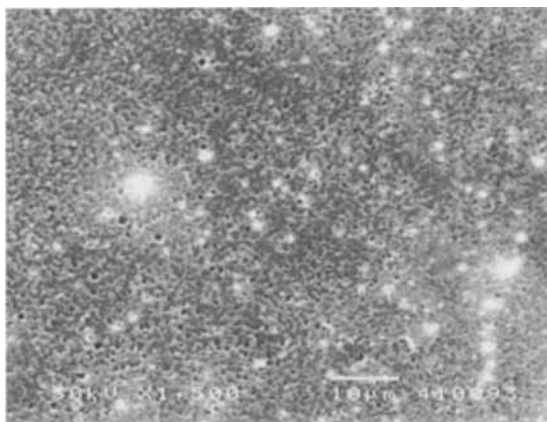
Figure 4 Morphology development of the compatibilized blends of 19% PS/76% LLDPE with 5% SEP. SEP was added into the blend at $t = 5$ min: (a, b) mixing of 6 and 8 min.



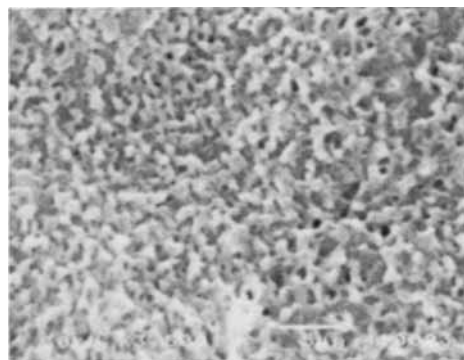
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Figure 5 Morphology development of the compatibilized blends of PS/LLDPE with SEBS in the Haake blender. SEBS was added into the blend at time = 0: (a) mixing of 1 min, (b, c) mixing of 4.5 min, (d) the interlocked structure.

in the mixing process (time = 1 min) indicated that the rate of the droplet breakup increased.

The Haake mixer experimental torque values during blending for different mixing times are shown in Figure 7. Both PS and LLDPE solid pellets were transformed to deformable solid pellets in a very short time during initial mixing (about 45 s). As the pellets became more deformed, the torque increases to a maximum value, as the solid was transferred into the liquid. Once polymer melt is present in the mixing chamber, there is a significant decrease

in torque, since the molten materials appear to lubricate each other. Based on the torque data, as well as the morphologies observed at different mixing times, two mixing actions are believed to take place in the Haake mixer. The dispersive mixing of PS in LLDPE proceeds first, associated with the circular shear flow, especially in the high shear zone of the narrow gap between the roller wings and the mixer wall. After a critical mixing period, the torque required for blending passed the maximum value and then decreased to a stable value. During this period,

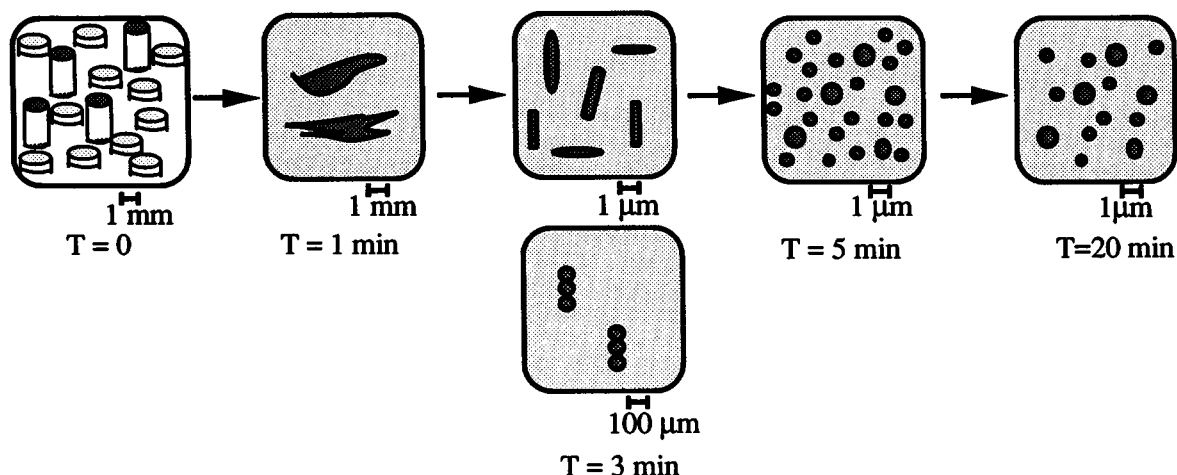


Figure 6 Schematic of morphological development during blending of 20% PS and 80% LLDPE.

the temperature of the mixture remained constant. In this experiment, it was observed that the distributive mixing was achieved in the mixer through transfer flow in the region between the two roller blades and through back axial flow.

It has been shown in the literature¹² that the presence of the compatibilizer would increase the friction or adhesion between the incompatible polymers, which should increase the torque value. The limited amount of study carried out thus far, consistent with previous investigations by Shih et al.,¹³

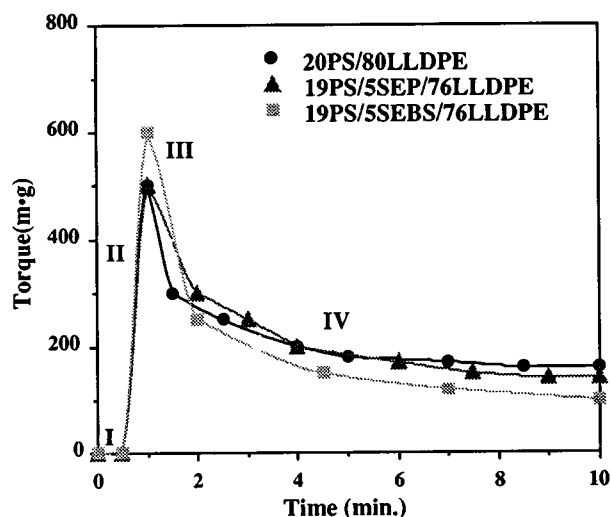


Figure 7 Torque data recorded during blending in the Haake mixer.

does not bear this out. Further investigation is necessary in order to reach a more definitive conclusion.

Both SEP and SEBS were found to be good compatibilizers for blends of LLDPE and PS at 180°C. The addition of SEP or SEBS not only reduces the dispersed-phase size, but also forms the interlock structure which represents the ideal morphology for an immiscible blend, since it allows for more equal sharing of imposed stresses by the components, and might also improve the mechanical properties of the blends.

The compatibilization mechanisms are dependent on the molecular structures. The possible compatibilization mechanisms of the copolymers with the immiscible blends are proposed in Figure 8. The SEM examinations showed that the blend morphology of LLDPE/PS with the SEP diblock copolymer is mostly controlled by the dispersed type of phase structure. Since SEP diblock copolymer has a simple molecular architecture, the penetration of copolymer segments into the respective homopolymer phases takes place and the copolymer tends to locate at the interface, thus separating unlike homopolymer species to reduce the number of unfavorable PS/LLDPE contacts and the interfacial tension between LLDPE and PS. For blends containing the triblock SEBS copolymer, the interlocked type of phase structure is dominant. From Figure 8, there are three possible conformations for SEBS in the PS/LLDPE blends. However, none of them seems to be thermodynamically favorable. The final morphology is stabilized with the interlocked

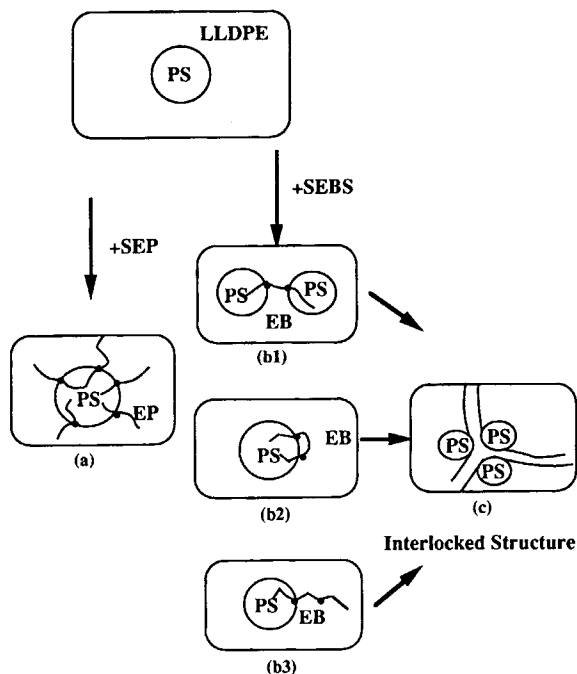


Figure 8 Proposed possible compatibilization mechanisms of the Kraton copolymer within the PS/LLDPE blends.

structure, which may be due to the low molecular weight rather than a more closely matched viscosity of SEBS with PS and LLDPE at 180°C. Another possibility is that the higher yield stress of SEBS help to hold the interlocked structure during processing without distortion.¹⁴

It has been shown that the better the interdispersion of PS and LLDPE in the block copolymer network, the better the chance for formation of interlocking networks.¹⁵ From experimental observations, it seems that the better the dispersion of PS in LLDPE, the greater the probability for formation of an interlocked phase structure. In case 3, PS was dispersed in LLDPE for 5 min before the addition of SEP. This mixing sequences was found to be beneficial for the formation of the interlocked phases in the resulting blends.

The experimental results shows that the SEBS low-molecular-weight triblock copolymer had a greater effect on compatibilizing PS and LLDPE than high-molecular-weight SEP diblock copolymer. It is known that the efficiency of block copolymers as compatibilizing agents depends on the number and arrangement of the constituent blocks. For example, the probability for individual blocks penetrating the corresponding homopolymer phase is likely to be higher for diblock copolymers than for triblock, multiblock, or star-shaped copolymers. In

addition, early studies focusing on solutions of immiscible polymers and solvent-cast blends concluded that block copolymers act as efficient emulsifiers only when the molecular weight of each segment is comparable to or higher than the molecular weight of the corresponding homopolymer.¹⁶ The results of this melt blending study do not seem to agree with those previous observations. The superiority of the triblock over the diblock is observed. Even short-block copolymers behave much more efficiently than longer ones. However, because of the structure similarity between ethylene/butylene (EB) blocks in SEBS and the butene branch in LLDPE, the chemical structure might be the key criteria in determining the activity of polymeric emulsifiers. In addition, Aggarwal et al.¹⁷ have shown that when the molecular weight of the added homopolymer is larger than that of the corresponding block copolymer, the blocks in the interfacial region between the homopolymer and the block polymer are preferentially anchored in the homopolymer phase and thus help to form a stable interface. The penetration of SEP into the interfacial region between phases of LLDPE and PS can be seriously hampered by its high viscosity (i.e., high molecular weight).

CONCLUSIONS

In this study morphological development during the blending of LLDPE and PS and the resulting mixture shows a bimodal particle size distribution of the minor phase at the initial mixing stage where the most significant changes in phase morphology occur. The reduction mechanism of droplet sizes is primarily effective in reducing the number of the largest droplets rather than decreasing the diameters of all the droplets. It is concluded that during blending the rate of the droplet breakup is increased by adding a compatibilizer. Due to the reduction of the interfacial tension between phases, a smaller particle size of the minor phase is achieved earlier in the blending process for the compatibilized blend.

The addition of SEP or SEBS modifies the dispersive morphology of blends of LLDPE and PS at 180°C. The short-triblock SEBS copolymer appears to be more efficient than the longer SEP diblock copolymer in the emulsification of LLDPE/PS blends. The addition of SEP or SEBS not only reduces the dispersed-phase size, but also results in an interlocking structure. The tendency toward forming the interlocked structure is determined by the interdispersion and the molecular structure of the copolymers. Adding SEP at the different stages

of morphological development affects the tendency toward forming the interlocked structure. The experimental results suggested that the better the interdispersion of the copolymer with PS and LLDPE, the greater the probability for forming the interlocked structure.

Qualitatively, it is concluded that the final equilibrium morphologies of the 20% PS/80% LLDPE blend and the related polyalloy are achieved in 5 min of mixing at 180°C and 50 rpm. A quantitative study of the effects of mixing time and mixing sequence upon the morphological development during polymer blending will be presented in a subsequent work.

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