

# Mechanical Properties and Creep Resistance in Polystyrene/Polyethylene Blends

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Received 28 December 1998; accepted 1 March 1999

**ABSTRACT:** Recycled plastics, predominantly high-density polyethylene (PE), are being processed in the shape of dimension lumber and marketed as “plastic lumber.” One drawback to these products is their low creep resistance or high creep speed. The objective of this study was to examine the feasibility of reducing the creep speed of PE-based products by blending the PE with a lower-creep plastic, in this case polystyrene (PS). Various blends of PE and PS were prepared in either a laboratory extruder or a bowl mixer and then compression-molded. The mechanical properties, creep behavior, morphology, and thermal properties of extruded and compression-molded samples were determined. The modulus of elasticity of the extruded blends could be estimated by a weighted average of PS and PE, even in the absence of a compatibilizer. Processing strongly affected the morphology and mechanical properties of the blends. For 50% PS : 50% PE blends, the stress–strain curves of the extruded samples showed PE-like behavior, whereas those from compression-molded samples were brittle, PS-like curves. Flexural strength was 50% higher in the extruded samples than in those from compression molding. The creep experiments were performed in three-point bending. Creep speed was lower in 50% PS : 50% PE and 75% PS : 25% PE blends than in pure PS. Creep speed of 75% PS : 25% PE was lowest of all the extruded blends. PE formed the continuous phase even when the PS content was as high as 50 wt %. For a 75% PS : 25% PE blend, cocontinuous phases were observed in the machine direction. A ribbonlike PS-dispersed phase was observed in the 25% PS : 75% PE and 50% PS : 50% PE samples. Blending low-creep-speed PS with high-creep-speed PE appeared to successfully improve the performance of the final composite. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 76: 1100–1108, 2000

**Key words:** polystyrene; polyethylene; blend; creep; extrusion

## INTRODUCTION

Polystyrene (PS) and polyethylene (PE) are two of the most widely used plastics in the world,<sup>1</sup> with annual production rates in the hundreds of bil-

lions of pounds. Less than 10% of this production is currently recycled, for a variety of reasons.<sup>2</sup> One use for recycled plastics, especially PE, is extrusion into shapes resembling lumber—thus the term “plastic lumber.” These products are used as building materials in a variety of applications, including decking, landscaping, signs, and outdoor furniture. One drawback to some plastic lumber products is their tendency to creep. This is especially true for those products composed primarily of PE.

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*Journal of Applied Polymer Science*, Vol. 76, 1100–1108 (2000)  
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Creep in plastic lumber products may be reduced by blending the low-creep-resistance (or high-creep-speed) PE with a lower-creep-speed plastic, such as PS. These blends may also possess mechanical properties intermediate between PS and PE; therefore, they may be tough, but not as brittle as PS alone. They also may possess good elongation, but be less ductile than pure PE. This kind of blend might also provide a useful matrix for wood/plastic composites.<sup>3</sup> The objective of this study was to investigate the feasibility of improving the performance of PE as a material for the manufacture of building materials by blending it with PS.

PS is incompatible with PE.<sup>2</sup> Blends of these compounds consist of mixtures of phases of the two components, either dispersed or continuous. Typically, these blends exhibit weak adhesion (i.e., poor stress transfer) between the PS and PE phases, which manifests as inferior mechanical properties in the final composite. Most previous studies have focused on compatibilization of the phases. Almost all compatibilizers reported have been block copolymers consisting of at least two blocks, with one block similar in structure or chemistry to PS and another similar to PE. Thus, the diblock copolymer, as it were, ties the two phases together. The effect of compatibilization on the morphology of the blend<sup>4-7</sup> and its mechanical properties<sup>8-12</sup> has been well studied. Compatibilization promotes the formation of an interlocking structure in PS/PE blends, which allows more equal sharing of imposed stresses and might therefore improve the mechanical properties of the blends.<sup>8</sup>

PS/PE blends can be processed in several ways: by compression molding,<sup>10,13</sup> by coextrusion,<sup>14,15</sup> or with a rolling mill.<sup>4,5,16</sup> Generally, the morphology of the resulting blend is related to its processing history. The morphology of the blend strongly influences mechanical properties. Usually, smaller sizes in the dispersed phase produce better mechanical properties.<sup>12</sup> The size and shape of the minor phase are critical to the impact properties.<sup>17,18</sup> A literature search found no reports on creep resistance in this blend system.

In this study, the mechanical properties, morphology, and creep behavior of PS/PE blends from an extrusion process were investigated. The results were compared with those from blends formed by compression molding.

## EXPERIMENTAL

### Materials

High-density PE was contributed by Phillips Petrochemical Company (Houston, TX) as Marlex EHM 6007. The molecular weight was 120,000 (by gel permeation chromatography); the density, 0.964; and the melt index, 0.65 g/10 min (190°C/2 kg). PS (Dow 685D) was contributed by Dow Chemical Corporation (Midland, MI). The weight-average molecular weight was approximately 300,000; the density, 1.04; and the melt index, 1.5 g/10 min (200°C/5 kg). Characteristic data were provided by the suppliers.

### Processing

The plastics were received as pellets and were dry-mixed before going through a 19-mm (0.75-in.) single-screw Brabender extruder. The Brabender extruder was operated at 40 rpm, with the barrel temperature set at 170°C for all heating zones. The temperature at the die region was measured as 180°C. A rectangular die (2 × 12 mm) was attached to the extruder. The pressures were in the range of 1–2.5 MPa, depending on the material extruded. A typical extrusion rate was 1.2 m/min. To compensate for the die swell, a shaping die was placed 5–8 cm downstream of the extrusion die. Water was used to cool and solidify the extrudate at the shaping die. A puller was placed downstream from the shaping die and synchronized with the extruder manually to minimize drop in the extrudate as it exited the rectangular die. Test bars (60 mm) were cut from the cooled extrudate and chosen randomly for testing.

Samples were also prepared in a Brabender Plasticorder bowl mixer with cam blades attached. The mixer speed was set at 30 rpm and 177°C. Typical mixing time was 10 min.

The samples from the bowl mixer were ground in a Wiley mill to a particle size of approximately 3 mm (0.1 in.) before compression molding. The blends were then compression-molded in a thermostated Carver laboratory press. The press conditions were 180°C and 6.9 Mpa for 10 min. The samples were then cooled to room temperature under pressure at ambient temperature. The molded samples were 2 × 13 × 55 mm.

### Testing of Mechanical Properties

Mechanical properties were determined with a three-point bending apparatus in accordance with

ASTM standard D 790-86. Whenever possible, five samples were tested for each determination of the modulus of elasticity (MOE), and the ultimate stress. The crosshead speed was 1.3 mm/min. The samples were flattened by filing. Typically it was difficult to obtain a flat molded sample with a high PE content.

Ultimate stress is defined in different ways, depending on the fracture behavior of the material:

1. Modulus of rupture (MOR): In this case, the stress increases with strain until samples break cleanly. This is brittle fracture, which was observed only for the compression-molded 100% PS samples.
2. Yield strength (YS): The yield strength is defined as the first point at which the stress-strain curve shows a slope of zero. This point is reported for those samples (the 25, 50, and 75% PS blends and the extruded 100% PS) that either yielded before breaking or did not break but continued to bend to the limits of travel of the testing machine.
3. Offset yield strength (OYS): In the cases where the samples neither broke nor showed a yield point (100% PE), the stress-strain curve did not show a slope of zero, but rather rose smoothly to the limits of travel of the testing machine. In these cases, the ultimate strength was reported as the 0.5% strain offset yield strength. This was calculated with the method specified in ASTM standard D790-86 and described in ASTM standard D638-89.

The creep experiment was done in an environmental chamber set at 19.5°C and 50% relative humidity. The specimens were  $1.71 \pm 0.01$  mm (mean  $\pm$  SD) thick. The span/thickness ratio ( $L/D$ ) was  $20.0 \pm 0.2$ . The creep tests were performed at a load of 50% of the ultimate strength, unless otherwise indicated. This rather high loading was chosen to ensure that we would see creep in most of the samples, thereby allowing valid comparisons.

### Density

The density of samples was measured by a buoyancy method with a Denver Instrument XE-310 balance. The samples were submerged in water

with fine copper wire. The density of the water was assumed to be 1.00 g/cm<sup>3</sup>, so the weight of the displaced water was equal to the volume of the submerged sample and wire. The density was calculated as

$$d = \frac{W}{V_{\text{total}} - V_{\text{cu}}} \quad (1)$$

where  $d$  = density (g/cm<sup>3</sup>),  $W$  = sample weight (g),  $V_{\text{total}}$  = weight of displaced water and wire (g), and  $V_{\text{cu}}$  = weight of the submerged wire (g). The standard deviation of the measurements was 0.002 g/cm<sup>3</sup>.

### Microscopy

Phase morphology was examined with an AmRay 1000A scanning electron microscope (SEM). The samples were dipped in liquid nitrogen and then fractured. To increase the contrast between the matrix and the dispersed phases, all samples of PS/PE blends were soaked in toluene at room temperature to dissolve the PS phase. All samples were coated with a Au-Pd film (8–10 nm) before imaging. The SEM was operated at 10 kV on Polaroid-type 55 positive-negative film.

### Rheological Testing

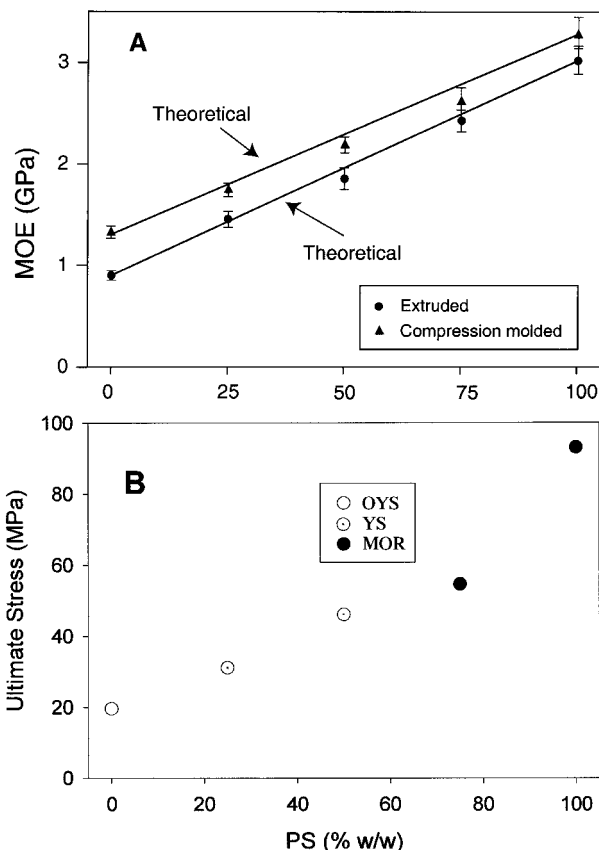
The relaxation times of polymers and blends were measured with a Bohlin CS-50 rheometer. The samples were molded to discs of 25 mm diameter  $\times$  1 mm thickness. The molding temperatures were 160°C for PE, 175°C for 50% PS : 50% PE, and 195°C for PS. The samples were held at the molding temperature for 30 min. Oscillation tests were performed for these samples to obtain elastic modulus ( $G'$ ) and viscous modulus ( $G''$ ) at various oscillation frequencies ( $f$ ). The relaxation time ( $\tau$ ) was calculated as<sup>21</sup>

$$\tau = \frac{1}{f_c} \quad (2)$$

where  $f_c$  = the frequency where  $G'$  and  $G''$  intersect.

### Differential Scanning Calorimetry (DSC)

The thermal behavior of the samples was characterized with a Shimadzu differential scanning calorimeter with a TA50 software package. The



**Figure 1** (A) Modulus of elasticity (MOE) for extruded and compression-molded PS/PE blends; (B) flexural strength of extruded blends.

temperature was ramped at 10°C/min in a range of at least 70–160°C. If a second run was performed, the samples were cooled at ambient conditions below 100°C.

## RESULTS AND DISCUSSION

### Mechanical Properties

#### MOE

The MOE of the blends varied as the weighted average of the PE and PS components [Fig. 1(A)], as expected. The compression-molded samples had a higher MOE than did the extruded samples, with the MOE of compression-molded 50% PS : 50% PE ( $2.26 \pm 0.05$  GPa) about 25% higher than that of extruded samples ( $1.87 \pm 0.11$  GPa). (Results are expressed as mean  $\pm$  SD.) A similar increase in ductility in extruded blends was also observed by Stell,<sup>22</sup> but without interpretation.

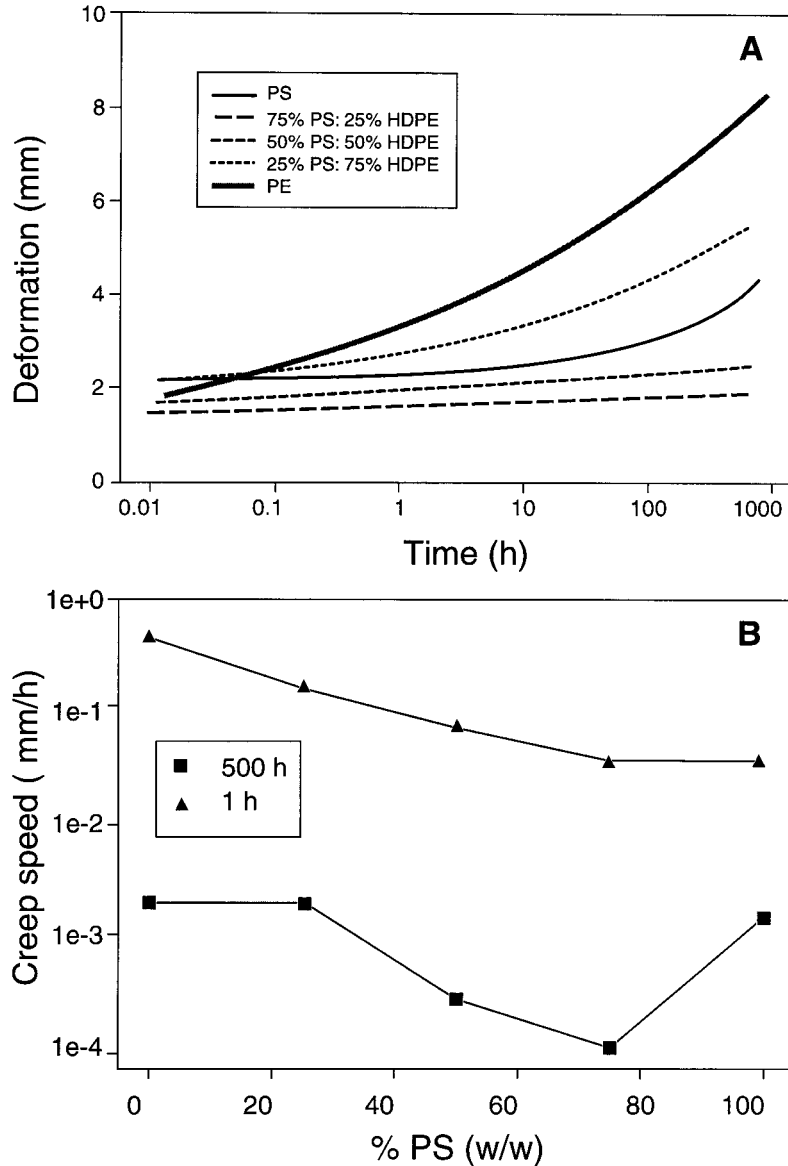
### Strength

The flexural strength of PS/PE blends increased with increasing PS content, but was lower than that predicted by a simple weighted average of the components [Fig. 1(B)] (which would be a straight line connecting 100% PE and 100% PS). This is typical of blended systems with poor interfacial adhesion.<sup>10,23</sup> Interpretation of the data was complicated by the varying fracture modes of the samples. PS is brittle and PE is ductile. As the PS content increased, the appropriate strength measure changed from offset yield strength (OYS; 0.5% strain was chosen) for 100% PE, to yield strength (YS) for 25 and 50% PS. Because the 75 and 100% PS extruded samples showed brittle fracture, the strength was reported as the MOR. Li<sup>17</sup> reported a similar result. Processing greatly influenced the strength of the blends. At 50% PS : 50% PE, the extruded sample was about 50% stronger ( $46.1 \pm 0.5$  MPa) than the compression-molded sample ( $29.8 \pm 1.5$  MPa).

The difference in mechanical properties caused by processing was hypothesized to be the result of an elongation of the polymer by force exerted on the extrudate by the puller and the rapid cooling of the extrudate at the shaper die. The birefringence of all samples containing PS under polarized light gave further evidence of elongation and indicated some degree of alignment in the PS phase. Annealing an extruded PS sample at 125°C for 24 h removed the birefringence in the PS and shrank the sample length by 65 to 75%. Thus, the processing appeared to induce some degree of elongation into the extrudate.

### Creep

All the samples showed some creep [Fig. 2(A)]. A linear transition of decreasing creep with increasing PS content was not observed. The 50% PS : 50% PE and 75% PS : 25% PE blends showed lower creep than 100% PS, with 75% PS : 25% PE exhibiting the least creep of all the samples tested. The different samples also showed different initial deflections. This is because they were loaded at 50% of estimated ultimate stress, which was a different load for each sample type. Since the stiffnesses also varied, but not linearly with the strengths, the samples were loaded at different points on the stress–strain curve; thus, initial deflections were different for different sample types.



**Figure 2** (A) Creep deflections for various PS/PE blends in three-point bending at 50% loading; (B) creep speed ( $V_c$ ) of PS/PE blends.

Creep was further characterized with a mathematical model, expressed as the empirical power function equation<sup>24</sup>

$$\varepsilon = \varepsilon_0 + at^b \quad (3)$$

where  $\varepsilon$  is the specimen deformation at time  $t$ ,  $\varepsilon_0$  is the initial deformation, and  $a$  and  $b$  are the time-independent and time-dependent fitting parameters, respectively. This equation was used to fit the deformation versus time data and gave values of  $R^2 > 0.85$  (Table I). The creep speed

was defined by taking the time derivative of eq. (3):

$$V_c = abt^{b-1} \quad (4)$$

where  $V_c$  is the creep speed.

$V_c$  is a complicated, nonlinear function of the PS content [Fig. 2(B)]. At 1 h, the creep speed decreases rapidly with increasing PS content up to 75% PS. Within experimental error, 100% PS exhibited the same creep speed as the 75% PS : 25% PE sample. However, the creep speed is also

**Table I Parameters of the Equation Modeling Creep in PS/PE Blends [Eq. (4)]<sup>a</sup>**

Sample	$R^2$	$\varepsilon_0$	$a$	$b$
100% PS	0.95	2.24 (0.05)	0.09 (0.02)	0.48 (0.03)
75% PS : 50% PE	0.87	1.12 (0.26)	0.54 (0.26)	0.07 (0.03)
50% PS : 50% PE	0.96	1.32 (0.12)	0.55 (0.12)	0.13 (0.01)
25% PS : 75% PE	0.86	2.25 (0.22)	1.05 (0.16)	0.2 (0.04)
100% PE	0.98	0.13 (0.4)	3.21 (0.40)	0.14 (0.01)

<sup>a</sup> Values are expressed as mean  $\pm$  SE.

a function of time. At  $t = 500$  h, the creep speed was remarkably less in the 50 and 75% PS content blends than in the other samples. The 100% PS sample showed a creep speed similar to 100% PE at 500 h.

In Figure (2A), the 100% PS sample is showing accelerating creep speed, usually a prelude to fracture, whereas the 50 and 75% PS blends appear stable, with low creep speeds. This anomalous behavior may perhaps be explained by the processing conditions. The blends were all extruded at the same temperature and at similar pressures, with PE appearing to be the continuous phase in the PS/PE blends. Thus, the viscosity of the continuous phase in the blends would be lower than that of the 100% PS sample. These differing viscosities may have given rise to differing amounts of elongation of the PS. The number of variables that control the elongation is large: melt viscosity, flow profile of the melt through the die, rate of cooling, and others.<sup>25</sup> Thus, we cannot predict the elongation for these different systems; yet they may affect the elongation, and thus the moduli and creep speed, of the samples.

The extruded samples of 50% PS : 50% PE showed a larger initial creep speed ( $V_c$ ) than did compression-molded samples. This is because their actual load was greater, as a result of their greater strength, but their MOE values were smaller than those for compression-molded samples. Thus, we would expect a greater initial de-

formation; however, they showed similar  $V_c$  values:  $0.08 \pm 0.02$  for the extruded samples and  $0.07 \pm 0.04$  for the compression-molded samples. The different morphologies did not result in different creep speeds within our experimental error. Unfortunately, a compression-molded 75% PS : 25% PE sample was not tested for creep.

### Density

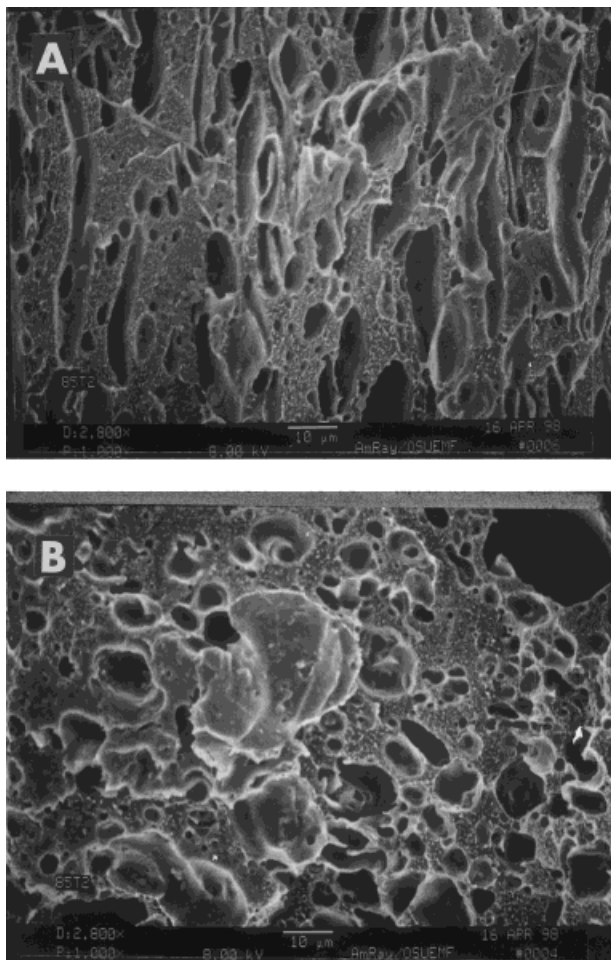
To investigate the differences between extruded and compression-molded samples further, we measured the density of the samples. The density for the 50% PS : 50% PE blend from compression molding ( $0.970 \text{ g/cm}^3$ ) was higher than that from extrusion ( $0.957 \text{ g/cm}^3$ ). Since DSC measurements showed that the crystallinity of the 50% PS : 50% PE blends from different processes was almost equivalent (about 51%), these data suggest that there are voids inside the extruded blends. Lahrouni<sup>26</sup> also has attributed the difference between the measured and the calculated density to the presence of voids in PS/PE blends. Shrinkage during PE crystallization is about 10%.<sup>18</sup> Thus, the difference in MOE may be the result of voids that arise from different thermal expansion coefficients for the different phases in the blended samples. This morphological difference probably arises, in turn, from processing methods.

### Morphology

PE was the continuous phase in 100% PE, 25% PS : 75% PE, and 50% PS : 50% PE [Fig. 3(A) and (B)]. The dispersed PS was elongated in the machine direction [Fig. 3(A)] and was uniformly distributed, except at the surface skin region, where PE predominated, possibly because of its low viscosity [Fig. 3(B)]. This "boundary layer" (not shown) appeared to be only about 100  $\mu\text{m}$  thick.

**Table II Glass Transition Temperature ( $T_g$ ) of 50% PS : 50% PE Blends**

Sample	$T_g$ ( $^{\circ}\text{C}$ )
Extruded, first run	114.3
Extruded, second run	112.8
Compression-molded	112.7



**Figure 3** SEM of 50% PS : 50% PE blend: (A) machine direction, showing elongated PS phase; (B) transverse direction.

The morphology of the 25% PS : 75% PE blend (images not shown) was similar to that of the 50% PS : 50% PE.

In other work relating the morphology of elongated, dispersed PS to the shear stress,<sup>14</sup> elongated (ribbon-type) PS was observed at the outer part of the samples, and relatively round (droplet-type) PS was found in the interior. This morphology gradient was attributed to competition between the relaxation rate of melt-flow morphology and the cooling rate in the mold. In this study, however, PS was elongated even at the sample center, where no shear stress exists. This suggests that shear stress was not the only factor causing elongation.

Since the die was about 1.5 cm long, and the typical extrusion velocity was about 2 cm/s, the polymers had about 0.8 s to relax. However, rheo-

logical testing at 180°C showed that the relaxation times for pure PE, 50% PS : 50% PE, and PS were 0.7, 1.0, and 2.5 s respectively. This suggests that the steady-state flow conditions for the PS phase of the plastic blend were not fully developed at the outlet of the die. This factor may have contributed to the pervasive ribbonlike PS distribution.

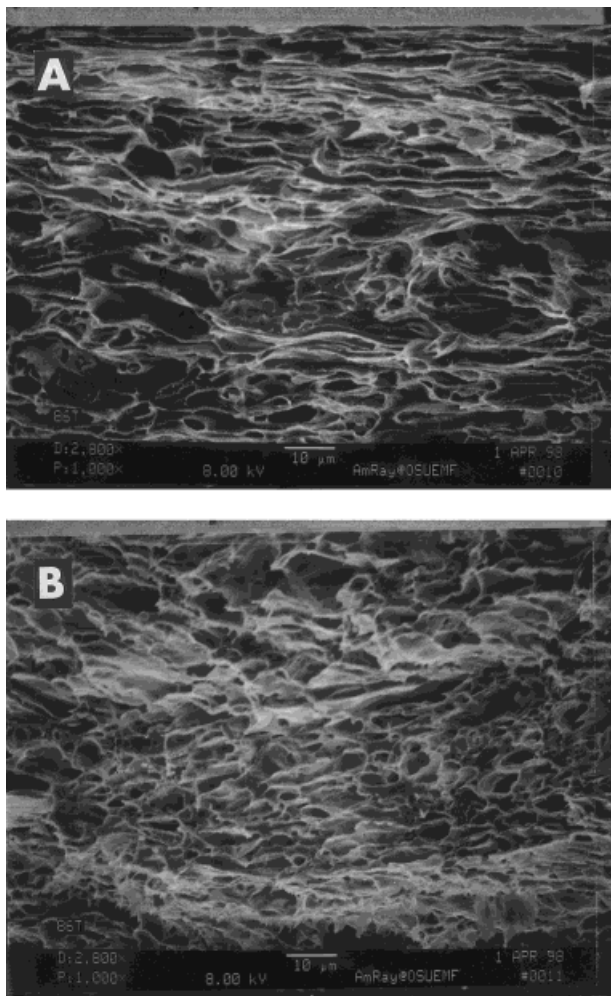
Pull force was also important in determining the morphology of the blends. The puller was operated so as to maintain a constant tension on the extrudate, which elongated the extrudate and thus gave an elongated, dispersed PS phase. The dispersed PS was distributed in both droplet and ribbon shapes. The ribbon-type domain was usually about 5–10 μm in width and varied in length. The droplet-type domain was much smaller and was mixed with the ribbon-type PS (Fig. 3).

The 75% PS : 25% PE blend also showed elongated phases in the machine direction [Fig. 4(A)]. The cross-sectional view showed significant intertwining of PS and PE [Fig. 4(B)].

The ribbon-type morphology was absent from the 50% PE : 50% PS compression-molded samples (Fig. 5). The different processes gave samples that differed in mechanical properties. The yield strength of the extruded 50% PS : 50% PE ( $46.1 \pm 0.5$  MPa) was higher than that of the compression-molded 50% PS : 50% PE blend ( $29.8 \pm 1.5$  MPa). However, the stiffness was reduced in the extruded sample, with an MOE of  $1.87 \pm 0.11$  GPa for extrusion and  $2.26 \pm 0.05$  GPa for the compression-molded sample. The poorer mechanical interlocking in the compression-molded samples relative to that in the extruded samples supports the contention that mechanical interlocking is at least partially responsible for the higher strength observed in the extruded samples. The cocontinuous phases in the 75% PS : 25% PE may also be responsible for the equivalent or lower  $V_c$ 's of 75% PS : 25% PE compared with pure PS [Fig. 2(B)].

### Thermal Analysis

The  $T_g$  of PS was higher in the extruded samples of 50% PS : 50% PE than in the compression-molded samples (Fig. 6). The  $T_g$  depression in the DSC curve was also widened in the extruded sample. These differences diminished after a second DSC run of the extruded sample. We conclude that annealing took place during the first DSC run, which reduced the difference between com-



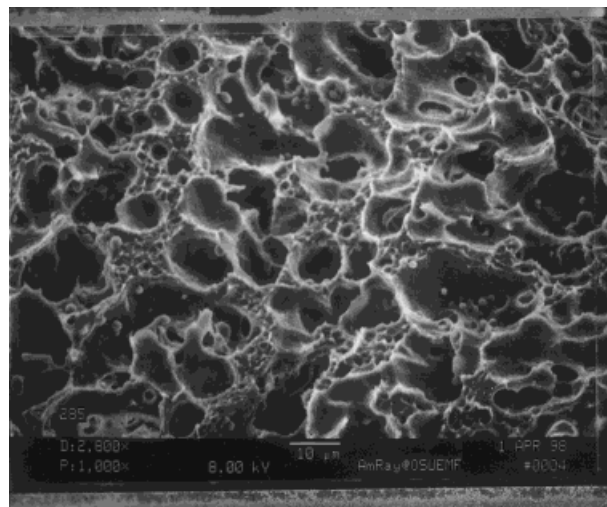
**Figure 4** SEM of 75% PS : 25% PE blend: (A) machine direction, showing elongated and intertwined phases; (B) transverse direction, showing cocontinuous phases.

pression-molded and extruded samples. These data suggest that the PS was stressed in the extruded samples, presumably from the elongation induced by the processing.

DSC spectra were analyzed for the specific enthalpy of the HDPE phase. Regardless of the PS content of the blend, the specific enthalpy of the HDPE remained constant. We concluded there was no effect of the blending or processing of the composites on HDPE crystallinity.

**CONCLUSIONS**

The MOEs of the PE/PS blends were related to the weighted average of the MOE values of the

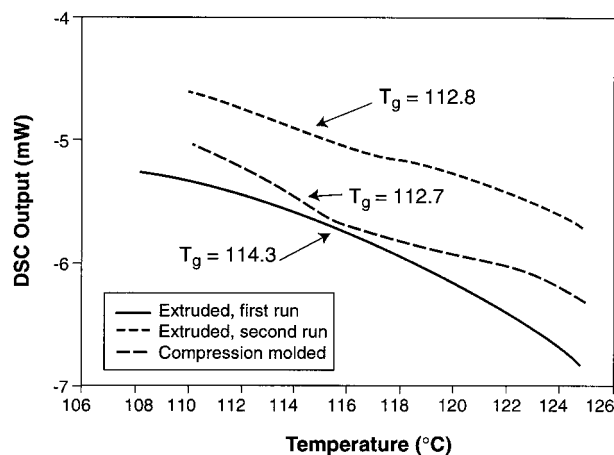


**Figure 5** SEM of compression-molded 50% PS : 50% PE blend.

components. The strength values of the blends were generally below values expected from a weighted average. Density data suggested that voids might have been present in the blends, which may account for the lower strengths. Processing can considerably influence the strength of the blend. Elongation of the extrudate, in this case, increased the strength of the blend by 50% over that of compression-molded samples.

Blending PS with PE reduced the creep speed from that observed in pure PE; the reduction was nonlinear.

Dispersed PS ribbons and droplets were observed in the 25% PS-content blend. Intertwined ribbons and droplets were observed in the ex-



**Figure 6**



truded samples containing 50 or 75% PS. The 75% PS blend showed a cocontinuous phase morphology that had the same creep speed as 100% PS.

The  $T_g$  of the PS phase was increased by extrusion, but returned to that of the compression-molded sample after annealing.

The use of blended PS/PE in "plastic lumber" products may improve their mechanical properties and reduce their creep.

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