

Creep Resistance of Wood-filled Polystyrene/High-Density Polyethylene Blends

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ABSTRACT: Creep, the deformation over time of a material under stress, is one characteristic of wood-filled polymer composites that has resulted in poor performance in certain applications. This project was undertaken to investigate the advantages of blending a plastic of lower-creep polystyrene (PS) with high-density polyethylene (HDPE) at ratios of 100:0, 75:25, 50:50, 25:75, and 0:100. These various PS–HDPE blends were then melt blended with a short fiber-length wood flour (WF). Extruded bars of each blend were examined to measure modulus of elasticity and ultimate stress. Increasing the ratio of WF increased modulus of elasticity in all composites, except between 30 and 40% WF, whereas the effect of WF on ultimate stress was variable, depending on the composite. Scanning electron microscopic images and thermal analysis indicated that the wood particles interacted with the PS phase, although the interactions were weak. Finally, creep speed was calculated by using a three-point bending geometry with a load of 50% of the ultimate stress. Creep decreased only slightly with increasing WF content but more significantly with increasing PS content, except at pure PS. The WF/75PS–25HDPE blend showed the least creep. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 79: 418–425, 2001

Key words: polyethylene; polystyrene; wood fiber; composite; creep

INTRODUCTION

Wood-filled plastic composites have received much research interest in recent years and are beginning to find their way into the marketplace.¹ One such product, “plastic lumber,” an alternative to solid wood as a building material, consists of extruded plastics (often recycled) in the shape of dimension lumber. Several factors encourage the popularity of this new material: the increasing demand for building materials, the low maintenance of plastic lumber, the decreasing avail-

ability of quality timber, and the increasing need for new technology to encourage recycling. Because the lack of a suitable end market for mixed recycled plastics is regarded as one of the most important factors hampering plastics recycling,² “plastic lumber” holds great promise for recycling efforts.

However, plastic lumber, which is typically manufactured from high-density polyethylene (HDPE), has poor mechanical properties compared with solid wood: its tensile strength and stiffness are typically one-fourth or less than that of solid wood. In addition, the poor creep properties of plastic lumber—that is, its tendency to slowly deform over time—have led to the need for some in-field replacements.³ The mechanical

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properties (strength and stiffness) of the plastic blends can be improved by adding reinforcing fillers. Wood fiber and wood flour have been widely used as reinforcing materials in plastic composites, including some plastic lumber products.³

Polystyrene (PS) and HDPE are two of the most widely used plastics in the world.⁴ PS typically exhibits less creep than HDPE does and has superior strength and stiffness. However, because it is more brittle than HDPE (as well as less widely available and not always recycled), PS has not been as widely used for plastic lumber. Mixing the two holds the potential for a composite stronger than HDPE alone, but not as brittle as PS. Decreasing the creep of these materials might also increase the market for products incorporating recycled plastics, especially HDPE.

The morphology and mechanical properties of large-dimension, extruded PS-HDPE blends have been studied by Li et al.^{5,6} In the composites studied, Li et al. observed a component distribution gradient and evaluated the mechanical properties in relationship to the hierarchical morphology. Other researchers found that the addition of PS in an injection molding process increased the stiffness of the blend and suggested the best matrix composition for commercial applications was 35PS-65HDPE.^{7,*} Previously in our laboratory, we studied the interaction between wood flour (WF) pretreated with phenol-formaldehyde resins and PS-HDPE blends in compression-molded WF/PS-HDPE composites.⁸

Creep in thermoplastics is a complex phenomenon, which depends both on material properties (molecular orientation, crystallinity, etc.⁹) and external parameters (applied stress, temperature, and humidity). The presence of wood fibers introduces several additional parameters that affect the mechanical and creep behavior of the composites. These parameters include the fiber-volume fraction, the fiber-aspect ratio, the orientation of fibers as a result of processing, and the mechanical properties of the fibers.

The objective of this study was to investigate the feasibility of decreasing the creep of HDPE blends through the addition of PS and WF, and by using extrusion techniques. The mechanical properties and creep response of various WF/PS-HDPE composites were studied. Scanning electron microscopy (SEM) and differential scanning

calorimetry (DSC) were used to characterize the resulting composites.

EXPERIMENTAL

Materials

HDPE was contributed by Phillips Petrochemical Company (Houston, TX) as Marlex EHM 6007. The manufacturer's specifications gave the molecular weight as 120,000 (by gel permeation chromatography), the density as 0.964 g/cm³, and the melt index as 0.65 g/10 min (190°C/2 kg). PS was contributed by Dow Chemical Corporation (Midland, MI) as Dow 685D. The weight-average molecular weight was approximately 300,000, the density was 1.04 g/cm³, and the melt index was 1.5 g/10 min (200°C/5 kg).

The wood flour (60-80 mesh) was contributed by Natural Fiber Composites, Inc. (Baraboo, WI). The aspect ratio of the WF was analyzed by using a microscope with an Image Analysis System (NIH Image version 1.6). More than 300 randomly selected fibers were measured with a resulting aspect ratio (length/diameter) of 3.0, with a standard deviation of 1.6. The WF was dried overnight in a vacuum oven at 60°C before processing.

Preparation of WF/PS-HDPE Composites

The composites were prepared in two steps. Initial mixing was performed in a Banbury mixer preheated to 135°C. In this first step, 160 g of WF was blended for 4 min with 260 g of PS-HDPE at a ratio of 100:0, 75:25, 50:50, 25:75, or 0:100. This gave a 40% (by weight) WF stock composite (40% WF/60% plastic composite). After being ground and passed through a 6-mm mesh screen, the stock samples were dried overnight in a vacuum oven at 60°C before further processing.

Second, the 40% WF/60% plastic mixture was diluted with PS-HDPE at the same ratio as in step one, 100:0, 75:25, 50:50, 25:75, or 0:100, to yield the various WF/plastic mixtures (10, 20, 30, and 40% WF, by weight). This mixture was then extruded through a 19-mm (3/4-in.) single-screw Brabender extruder attached to a Brabender Plasticorder drive unit. The extruder was operated at 40 rpm, with the barrel temperature set at 170°C for all heating zones. The melt temperature in the die region was 180°C. A rectangular die (2 × 12 mm) was attached to the ex-

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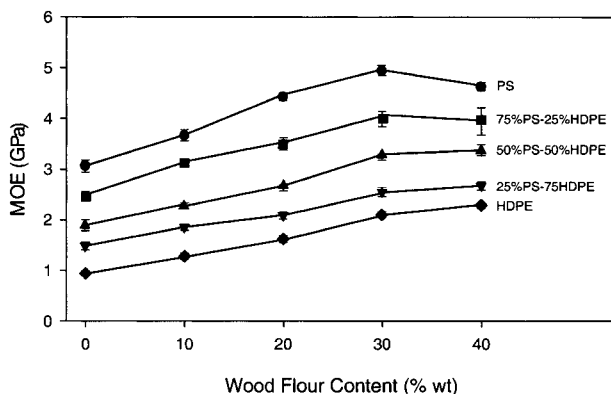


Figure 1 MOE vs filler content for PS–HDPE blends.

truder. The melt pressures measured at the entrance to the die were in the range of 1 to 3.5 MP, depending on the material extruded, with a typical extrusion rate of 1.2 m/min. To compensate for the die swell, a shaping die was placed 5–8 cm downstream from the extrusion die. Water was used to cool and solidify the extrudate at the shaping die. A belt puller was placed downstream from the shaping die and manually synchronized with the extruder to minimize the sag in the melt extrudate as it exited the rectangular die. The extruded samples were dried for about 4 h in a vacuum oven at 60°C. Test bars (1.9 × 11 × 60 mm) were cut from the cooled extrudate and chosen randomly for testing.

Testing and Characterization

In a previous study, we describe the mechanical testing, creep experiments, rheological testing, SEM, and DSC.¹⁰

RESULTS AND DISCUSSION

Mechanical Properties

All samples showed increasing stiffness or modulus of elasticity (MOE) with increasing PS content, as expected. For all composites, the MOE increased linearly with increasing WF content, and increasing PS content, to about 30% WF/70% plastic (Fig. 1). There appeared to be a leveling-off of the MOE between 30 and 40% WF content, which was not expected. This effect may be related to inadequate shear in the mixer and extruder in the composites with higher WF content. A similar phenomenon was observed by Yam et al.¹¹ The effect was more pronounced for higher

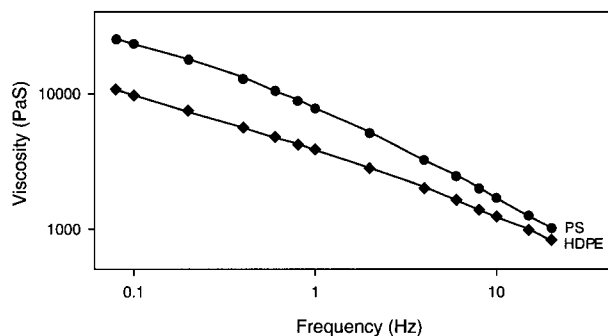


Figure 2 Dynamic viscosity of PS and HDPE at 180°C.

PS contents, which may reflect the higher melt viscosity of PS compared with HDPE at the extrusion temperature of 180°C (Fig. 2).

Depending on the fracture mode of the sample, the ultimate stress is appropriately reported as modulus of rupture, yield strength, or offset yield strength. The ultimate stress of WF/100PS decreased with increasing WF content, whereas the ultimate stress of WF/100HDPE composites increased with increasing WF content (Fig. 3). This trend has been reported previously for uncompatibilized PS composites.¹² The 100% HDPE-content samples with less than 30% WF showed offset yield strength fractures. The remaining composites showed either yielding fracture behavior or brittle fracture.

Pure extruded PS did not show brittle fracture. This increased ductility of PS has been discussed in our previous study,¹⁰ and also has been observed by other researchers.¹³ Its ductility may be attributed to elongation induced by the force of

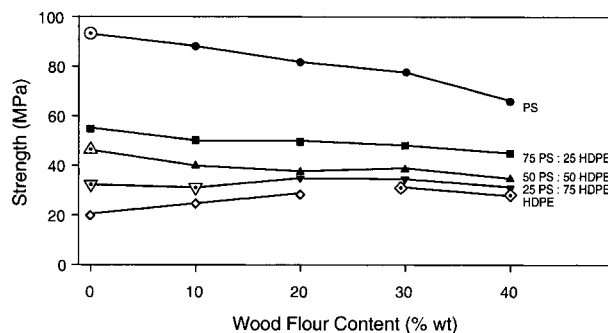


Figure 3 The ultimate stress (as strength, in MPa) of WF/PS–HDPE composites. Filled symbols indicate brittle fracture (or MOR), cross-haired symbols indicate yield strength (or YS), and unfilled symbols indicate ductile behavior (or YS).

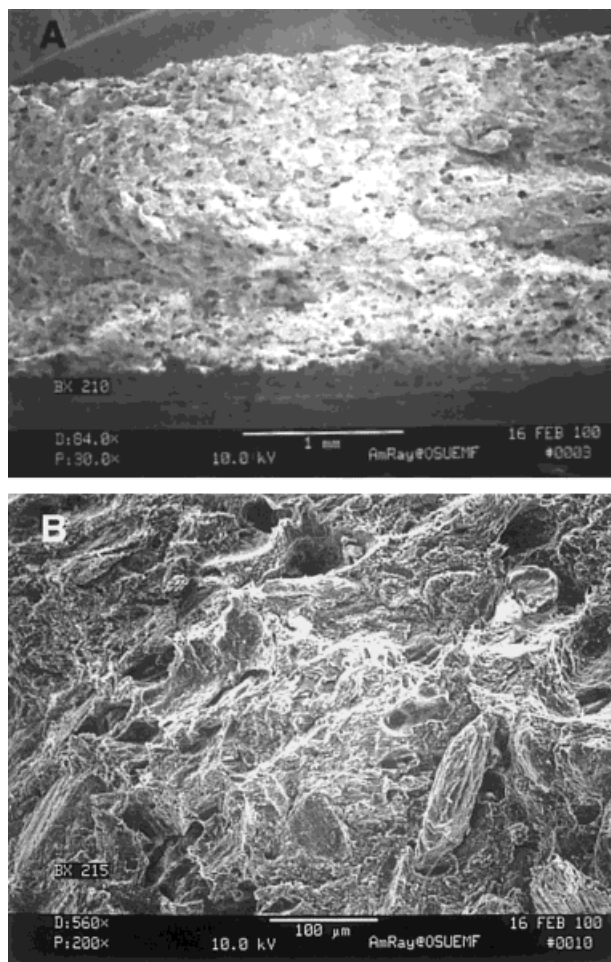


Figure 4 SEM image of (A) 40% WF/60% (75PS-25HDPE), at 30 \times original magnification; (B) 40% WF/60% (50PS-50HDPE), at 200 \times original magnification.

the puller and the rapid quenching of the molten extrudate at the shaping die. In the present study, we found the ultimate stress was the greatest for 100PS and decreased with increasing HDPE content, reflecting the weaker, but less brittle, nature of the HDPE component (Fig. 3). In addition, WF/100PS showed decreasing ultimate stress with increasing WF content. The WF (which contains dust) may provide discontinuities in the composite that serve to propagate fractures and thus reduce its ultimate stress.

Morphology

The WF was reasonably well dispersed in the plastic matrix [Fig. 4(A)]. In addition, air pockets were observed in most samples. These are presumably caused when residual moisture in the wood filler turns to steam during extrusion. All

samples were observed to contain air pockets. The volume fraction of the pockets varied from sample to sample, and may be the source of some of the variability in the test results.

The fracture surfaces of all samples showed partially immersed wood fibers, pullouts, and air pockets. A typical image is shown in Figure 4(B).

At higher magnification, the matrix was observed to change as the PS-HDPE ratio changed [Fig. 5(A-E)]. A single-phase matrix (PS) is shown in Figure 5(A). At 75PS-25HDPE, the HDPE phase is seen in a cross-sectional image as the discrete phase [Fig. 5(B)]. The size of the dispersed phase is on the order of a micron. The more ductile HDPE was stretched during fracture and can be seen extending from the fracture plane. A quite different image is observed in the machine direction [Fig. 5(C)]. This image, from the center of the extrudate, reveals co-continuous phases similar to those observed in unfilled blends.¹⁰ The co-continuous structure was observed to decrease toward the edges of the extrudate, suggesting greater flow in the center [Fig. 5(D)]. The cross-sectional image of the 50PS-50HDPE sample suggested similar co-continuous phases [Fig. 5(E)], whereas the 25PS-75HDPE sample presumably shows PS dispersed in HDPE [Fig. 5(F)]. Again, the size of the dispersed phase was observed to be on the order of a micron.

Many of these images reveal a lack of interphasal adhesion between PS and HDPE, and the degree of pullout of the WF suggests weak interfacial shear strength between the filler and the matrix. It is difficult to conclude from these images whether the filler adheres more to PS or to HDPE.

The presence of the co-continuous phases was hypothesized to affect the material properties of the composites, as discussed below.

Thermal Analysis

DSC spectra indicated that the glass transition temperature of the PS phase decreased slightly with increasing filler content (Table I), which is consistent with the results of previous research.¹⁴ This supports the hypothesis that there is a preferential adsorption of PS on the wood filler from PS-HDPE blends.

DSC spectra indicated that the specific enthalpy of the HDPE phase (determined from the integrated area of the melting peak) remained constant regardless of HDPE or WF content. This suggests that there is no significant change in the

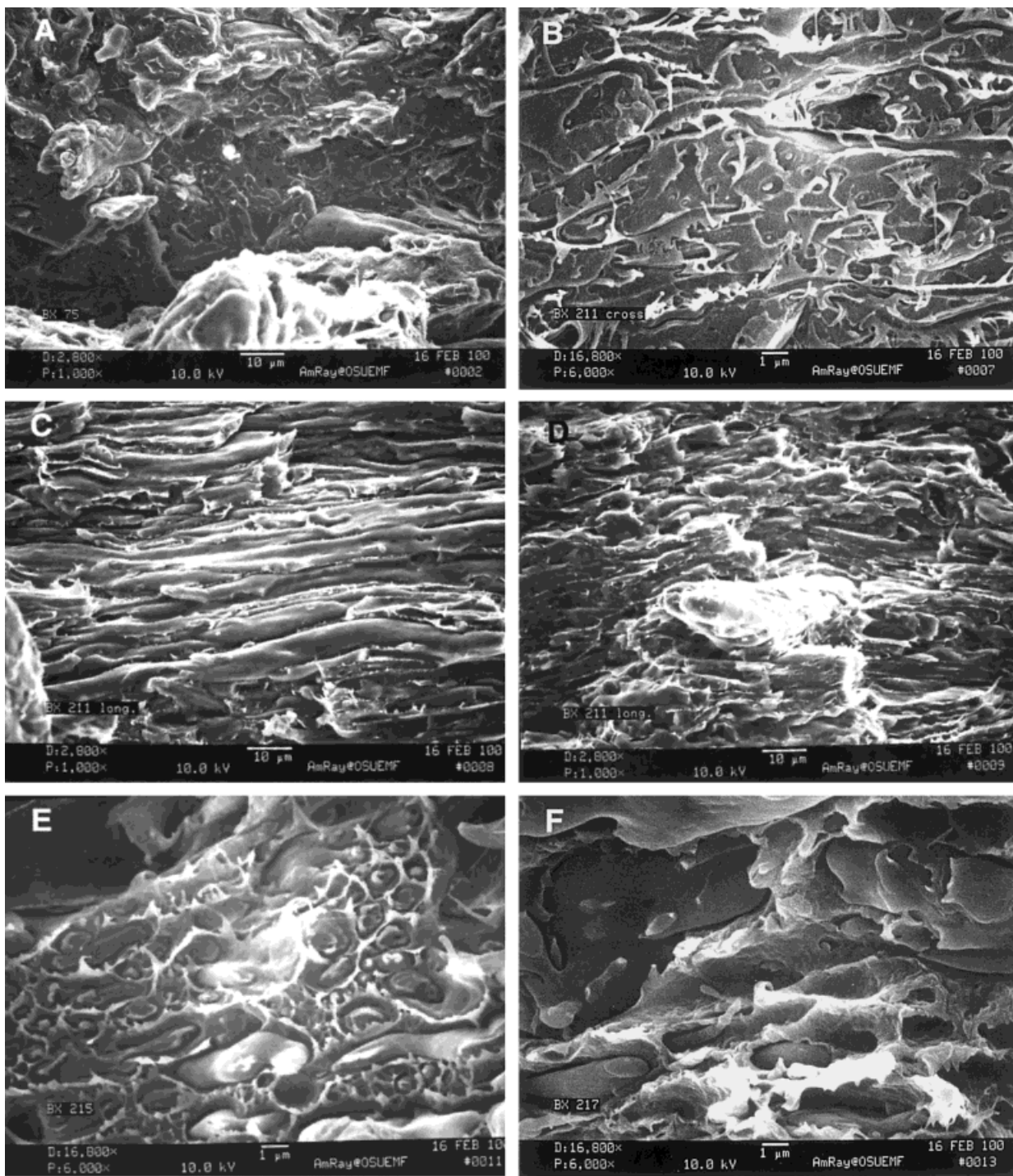


Figure 5 SEM image of (A) 40% WF/60% PS, at 1000 \times original magnification, cross-section; (B) 20% WF/60% (75PS-25HDPE), at 6000 \times original magnification, cross-section; (C) 20% WF/60% (75PS-25HDPE), at 1000 \times original magnification, where the image is from the center of the extrudate and in the machine direction; (D) 20% WF/60% (75PS-25HDPE), at 1000 \times original magnification, where the image is from the edge of the extrudate and in the machine direction; (E) 40% WF/60% (50PS-50HDPE), at 6000 \times original magnification, cross-section; (F) 40% WF/60% (25PS-75HDPE), at 6000 \times original magnification, cross-section.

Table I Glass Transition Temperature (T_g) of PS in WF/PS Composites

Wood Flour Content	T_g (°C)
0	114.2
10	113.6
20	113.4
30	112.4
40	112.2

crystallinity of HDPE in the WF/PS-HDPE composite system. We concluded that there were no interactions between WF and the HDPE phase. Similar results were obtained in previous work.^{8,10}

Creep

Following Findley et al.,¹⁵ a three-parameter empirical power equation was used to model the creep response of the samples [Fig. 6(A-C)]:

$$\epsilon = \epsilon_0 + at^b \tag{1}$$

where ϵ is the creep deformation, ϵ_0 , a , and b are model parameters, and t is the creep time. The correlation coefficients for the curve fit are typically >95%, and the standard deviations of the individual a and b parameters are approximately 10% (Table II).

In Figure 6(A-C), all the composites having 75PS-25HDPE and 50PS-50HDPE matrices showed lower creep than all other composites, even lower than the 100% PS matrix.

Creep speed (V_c) was derived from eq. (1) by taking the derivative with respect to time:

$$V_c = abt^{b-1} \tag{2}$$

Then ab from eq. (2) was defined as the “relative creep speed” at unit creep time.

Creep speed decreased with increasing PS content, except for pure PS (Fig. 7). One exception to this trend was the 40% WF/60% (75PS-25HDPE) composite. We speculate that this anomaly may be attributed to incomplete blending at the high filler content, but this result requires further study. The effect of WF content on creep speed did not appear to be as large as that of the PS content.

In addition, the pure PS sample showed a higher creep speed than 50PS-50HDPE and 75PS-25HDPE samples. In another study, a sim-

ilar trend was observed in unfilled PS-HDPE blends.¹⁰ It was concluded in that study that the morphology of the PS phase was elongated during processing. The filled samples in this study showed similar behavior and morphology. This suggests that the same elongation phenomenon may be responsible for the creep behavior in this study.

We conclude that the most effective creep reduction strategy is stiffening the matrix (by incorporating PS in this case) rather than adding filler.

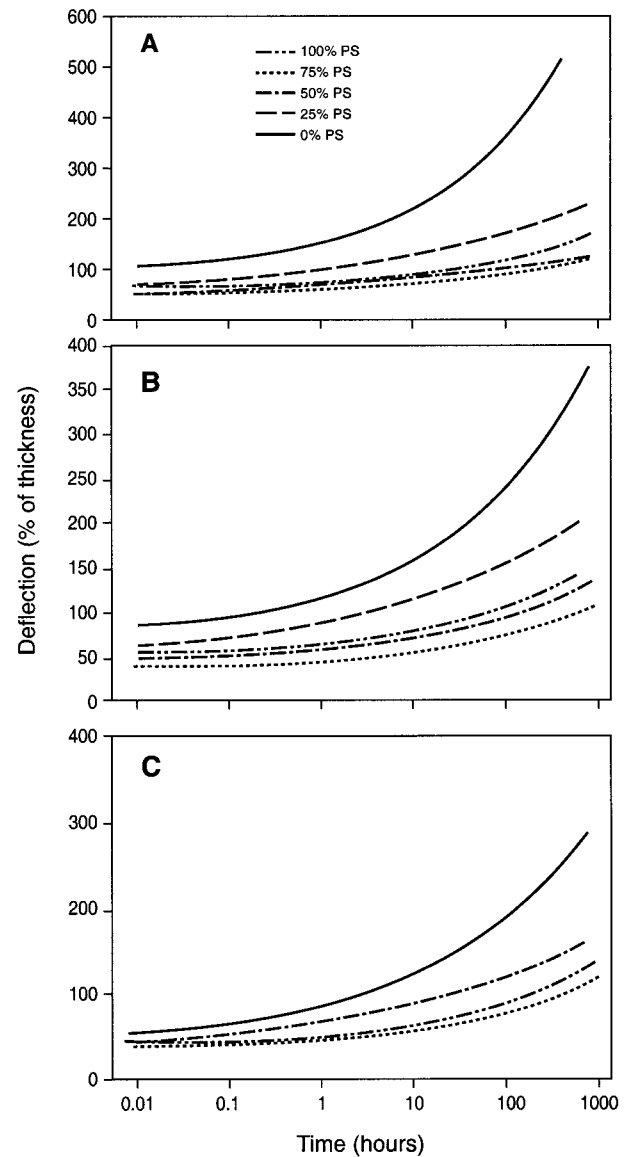


Figure 6 Creep response of (A) 20% WF/80% polymer blends and various blend ratios, (B) 30% WF/70% polymer blends and various blend ratios, and (C) 40% WF/60% polymer blends and various blend ratios.

Table II Parameters Derived from Curve Fitting Eq. (1) to Creep Data

Samples					
WF/Plastic Composite Ratio (%)	PS/HDPE Ratio (%)	R^2	ε_0	a	b
20:80	100:0	0.94	1.26 (0.07)	0.22 (0.05)	0.33 (0.03)
	75:25	0.98	0.99 (0.03)	0.22 (0.02)	0.27 (0.01)
	50:50	0.88	0.62 (0.31)	0.75 (0.31)	0.13 (0.04)
	25:75	0.98	0.95 (0.14)	0.99 (0.13)	0.19 (0.01)
30:70	0:100	0.98	1.80 (0.22)	1.13 (0.20)	0.32 (0.03)
	100:0	0.97	1.01 (0.06)	0.26 (0.04)	0.30 (0.02)
	75:25	0.97	0.73 (0.03)	0.18 (0.02)	0.30 (0.02)
	50:50	0.97	0.86 (0.06)	0.28 (0.05)	0.27 (0.02)
	25:75	0.99	0.89 (0.09)	0.82 (0.09)	0.20 (0.01)
40:60	0:100	0.98	1.42 (0.14)	0.82 (0.10)	0.30 (0.02)
	100:0	0.99	0.78 (0.02)	0.21 (0.01)	0.32 (0.01)
	75:25	0.99	0.69 (0.02)	0.18 (0.02)	0.32 (0.01)
	50:50	0.98	0.77 (0.04)	0.20 (0.03)	0.34 (0.02)
	25:75	0.99	0.51 (0.10)	0.76 (0.09)	0.19 (0.02)
	0:100	0.94	0.75 (0.20)	0.88 (0.17)	0.26 (0.03)

Values are expressed as mean (standard error).

CONCLUSIONS

The WF/PS–HDPE composites can be blended to produce a controllable range of stiffness and fracture behaviors. The MOE generally increased with increasing PS and WF content. Similarly, in measures of the blends' fracture behavior, the

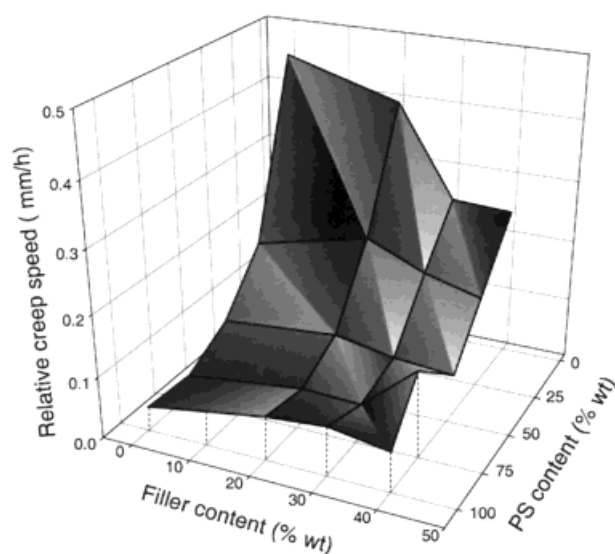


Figure 7 Creep speed of filled and unfilled PS–HDPE blends at unit creep time (1 h).

ultimate stress increased with an increasing PS ratio. The effect of WF on ultimate stress was variable, however, depending on the blend. For example, increasing the WF ratio decreased ultimate stress in WF/100PS, whereas it increased ultimate stress in WF/100HDPE. In 75PS–25HDPE, 50PS–50HDPE, and 25PS–75HDPE, the same increase in the WF ratio showed less clear effects on ultimate stress.

SEM images indicated that all samples contained a variable number of air pockets, which were presumably caused by steam generated from moisture in the filler. Co-continuous phases were observed in the center of the 50PS–50HDPE and 75PS–25HDPE filled blends, and may be a factor in the lower creep rates of these composites.

The creep of this system can be modeled successfully with an empirical three-parameter power model. The creep speed decreased with increasing PS content, and, to a lesser extent, with increasing WF content. The WF/PS–HDPE composites with plastic matrices of 50PS–50HDPE and 75PS–25HDPE showed lower creep speeds than the pure PS matrix, which may be attributed to the changes in polymer elongation induced by processing. The WF/75PS–25HDPE blend showed the least creep.

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