Morphology and Properties of Wood-Fiber Reinforced Blends of Recycled Polystyrene and Polyethylene

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ABSTRACT: Material properties of composites produced from recycled plastics and recycled wood fiber were compared. A blend of high-density polyethylene and polystyrene was used as a simulated mixed plastic. Stiffness was generally improved by the addition of fiber, as expected, but brittleness also increased. Pre-treatment of the wood filler with phenol-formaldehyde resins did not significantly affect material properties. Differential scanning calorimetry indicated no interaction between the polyethylene phase and the other phases present in the composite. Glass transition temperatures for the various combinations of components indicated a possible interaction between the polystyrene phase and untreated wood filler. This was supported by scanning electron micrographs, which indicated a less-coalesced morphology for samples filled with treated wood flour compared to those with untreated wood flour.

KEY WORDS: wood/polymer composite, polystyrene, polyethylene, recycled, plastics.

INTRODUCTION

PLASTICS RECYCLING IS currently hampered by a lack of markets for products made from recycled plastics. One potential use for recycled, mixed plastics is in the production of extruded plastic boards, commonly called "plastic lumber" [1]. In comparison to wood, plastic lumber has greater resistance to rot and greater dimensional stability. However, it lacks strength and stiffness; its modulus of elasticity (MOE) values are typically four or five times lower than wood [2]. It also exhibits excessive creep, and it is expensive. Wood-filled plastic

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lumber has recently appeared in the marketplace [3,4]. Wood-fiber fillers for plastics have been utilized since the dawn of the plastics age, primarily to reduce product cost. While wood-plastic composites are often stiffer than plastics alone, the full potential of the wood fiber has not yet been utilized due to the fundamental incompatibility of the components. Wood is hydrophilic, and most synthetic polymers are hydrophobic. Because this hydrophilic-hydrophobic incompatibility prevents the wetting and mixing required for a good dispersion of wood in synthetic polymers [5], the material properties of wood/plastic composites are limited. Furthermore, in the absence of chemical bonding between the two components, the strength of these composites is determined by the degree of mechanical adhesion between the plastic and wood [6]. Chemical additives have been developed to overcome this hydrophilic-hydrophobic incompatibility [7–9]. Compatibilizers for polypropylene have entered the marketplace [10]. The development of technology to improve the material properties of blends of recycled plastics, while a challenging research problem, offers the promise of significantly expanded and badly needed markets for recycled plastics.

It has been reported that the addition of polystyrene (PS) to high-density polyethylene (PE) significantly improves strength and stiffness, with a polystyrene content of 35% showing the best results [11]. This study utilized a polyethylene/polystyrene blend from recycled sources as a model for a blended "mixed plastics" system.

The objective of this study was to gain some preliminary information on filled blends of plastics and to determine whether pretreatment of the wood phase affected composite material properties.

MATERIALS

Plastics

Recycled high-density polyethylene (PE) and polystyrene (PS) were used. The PE, primarily from recycled milk jugs, was obtained from Partek Industries (Vancouver, Washington) as pellets. The polystyrene (PS) was obtained from packing noodles ground in a Hammermill. The two plastics were tested separately and also in blended form, in a 65% PE/35% PS (wt/wt) mixture. No other blend compositions were investigated.

Wood

Disc-refined flour from scrap pallets and plywood trimmings was obtained from the Evanite Corporation (Corvallis, Oregon) by screening the fiber from the slurry immediately downstream from disc refining. The wood flour was dried overnight at 105°C, then stored in an environmentally controlled hot, dry room (32°C, 30% relative humidity).

Wood Treatment

Treated and untreated wood flour was used. The untreated portion was ground to pass 16 mesh in a Wiley mill and used without further modification. An optical measuring system manufactured by Micro Motion Systems, Inc. was used to analyze the wood flour. The average length was found to be 0.48 mm with a standard deviation of 0.3 mm. The average aspect ratio was 5.8 mm with a standard deviation of 3.0 mm. The untreated wood flour contained dust, which was not included in the dimension measurements.

The remaining wood flour was oven-dried overnight at 105°C, then impregnated with phenol formaldehyde resin by overnight soaking in a phenol-formaldehyde resin-forming solution. Different resin loading levels were prepared by varying the resin concentration in the soaking solution to obtain a series of percent weight gains (PWGs) of the resin in the wood flour. The resin solutions used are presented in Table 1. The phenol/formaldehyde ratios are similar for each. The resins were cured by tightly wrapping the treated wood flour samples in aluminum foil and heating them at 105°C overnight, then exposing the samples to air and oven drying overnight, again at 105°C. The cured samples were then ground in a Wiley mill to pass 16 mesh. The treated wood flour was also analyzed optically. The average length was found to be 0.40 mm with a standard deviation of 0.2 mm. The average aspect ratio was 8.1 mm with a standard deviation of 4.3 mm. The treated wood flour also contained dust, which was not included in the dimension measurements.

METHODS

Composite Preparation and Testing

The samples were prepared in a Haake Buchler System 40 with mixing bowl and roller blades attached. The temperature was controlled at 190°C. The components of each sample were added sequentially to the mixing bowl. The plastics component(s) was added first, followed by the wood filler, either treated or untreated. Filler loadings of 25, 50 and 75% by weight were prepared for each matrix type (PE, PS and blend). To ensure good blending, the mixing was continued for 10 minutes after addition of all the components. The mix was removed from the mixing bowl, cooled, and ground to a coarse powder before compression molding into samples at 175°C and 10 Mpa (1500 psi).

Material Properties

Ultimate stress and flexural MOE were determined using a 3-point bending apparatus in accordance with ASTM standard D790-86. Five specimens were used

Table 1. Resin solution compositions.

Treatment	Phenol, %	Formaldehyde, %	Methanol, %	Ammonia, %
12 PWG	5.0	2.4		0.09
30 PWG	9.9	4.5		0.09
75 PWG	21.4	9.5		0.09

for each measurement except where molding defects disqualified some specimens. A minimum of three specimens was used in these cases.

Ultimate stress is defined in three different ways. Samples of pure polystyrene, filled and unfilled, and samples of the blended polymers with 50 and 75% loadings broke cleanly. The modulus of rupture (MOR) was reported for these samples. The 0 and 25%-loaded blended samples did not break but rather showed a yield point, defined as the first point at which the stress-strain curve shows a slope of zero. In this case, the value for the yield point was used in the same equation used to calculate MOR. The resulting value is termed the *yield strength* (YS). Samples of pure polyethylene, filled and unfilled, neither broke nor showed a yield point. In this case, the ultimate stress was calculated using the method specified in ASTM standard D790-86 and described in ASTM standard D638-89. This value was termed the 0.5% strain *offset yield strength* (OYS).

Differential Scanning Calorimetry

Melting temperatures and enthalpies of melting were measured on a Perkin Elmer Differential Scanning Calorimeter (DSC-7). The DSC was interfaced to a model 1020 system controller.

Dynamic Mechanical Analysis (DMA)

Dynamic mechanical properties (E', E'' and tan δ) were determined on a Rheometrics, Inc., RSA-II Solids Analyzer. The samples were tested at 5°C intervals between -100 and 150°C using a three-point bending geometry and a constant frequency of 1 Hz. Typical sample dimensions for this analysis were 48 mm × 12 mm × 2 mm. Tan δ values were reported as a function of temperature.

Scanning Electron Microscopy (SEM)

Micrographs were taken on an Amray Model AMR-900 scanning electron microscope. Fracture surfaces were prepared by freezing the samples in liquid N_2, breaking them under impact, and then coating the fractured surfaces with a gold-palladium alloy.

RESULTS AND DISCUSSION

Adding wood filler both to PS and HDPE separately and to the polymer blend increased the composite's MOE as expected (see Figure 1). No significant difference in the MOE values for each matrix occurred with the use of treated vs. untreated wood. The stiffnesses in Figure 1 are compared to those predicted by the transverse rule of mixtures, usually associated with the lower bound in filled composites [13].

$$\frac{1}{E_c} = \frac{V_f}{E_f} + \frac{V_m}{E_m}$$

where E is modulus and V is volume, the subscript c = composite, f = filler,

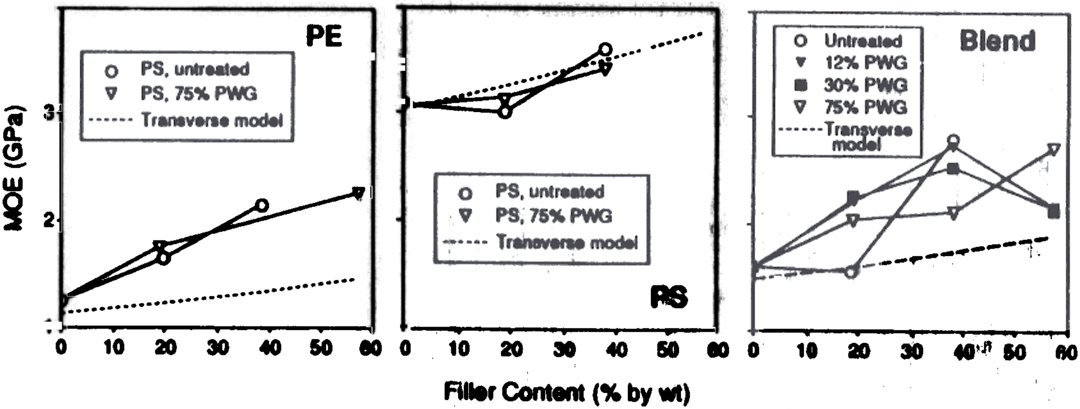


Figure 1. Moduli of elasticity for PE, PS and a 65/35 PE/PS blend filled with treated and untreated wood fiber compared to the transverse rule of mixtures model (lower bound).

and m = matrix. Since there are only two phases, $V_m + V_f = 1$. The values used to calculate E_c in Figure 1 are given in Table 2. This equation provides only a rough approximation to the lower bound expected in these composites. The PS matrix samples follow the transverse model, while the PE and blended samples rise slightly above it. While the PS samples show relatively poorer performance in comparison to this theoretical model, their measured stiffness is higher, reflecting the higher stiffness of the PS matrix. The values of blend stiffnesses are between the PE and PS stiffnesses. The scatter of the data is higher with the blend sample, possibly because of inhomogeneities in the PE/PS blends.

Ultimate stresses increased with increasing filler content for PE filled with untreated wood flour (see Figure 2). The use of treated filler showed an improvement over untreated for the 25% (w/w) loading. The higher loadings of the treated filler showed yielding behavior as opposed to offset yield strength behavior for the untreated samples, indicating different types of filler-matrix interactions. The PS samples showed decreasing ultimate stresses with increasing filler contents. This behavior is typical of thermoplastics containing non-reinforcing fillers [13]. In the blend samples, the untreated filler behaved similarly to untreated filler in PE. The blend samples with treated filler showed generally decreasing ultimate stresses with increasing filler content. Increasing the PWG of the treatment had no effect on ultimate stress, except for the 75 PWG at the highest filler content. This result requires more study.

The role of polyethylene in this system was investigated using differential scanning calorimetry. Figure 3 presents the enthalpy of melting of the polystyrene phase in all the different sample compositions. The enthalpy appears to be a function of polyethylene content only regardless of PE/PS ratio, amount of filler, or pretreatment of filler. This indicates little interaction between the PE phase and either the PS phase (if present) or the filler, either treated or untreated.

In an effort to better understand how the various treatments affect phase behavior, the materials were characterized using dynamic mechanical analysis because of its sensitivity to molecular relaxation processes.

Figure 4 compares the tan δ response (a measure of molecular mobility) of the unfilled PE/PS blend and the untreated wood/50% (w/w) filled system from −100°C to 150°C. Two common relaxations are apparent for these materials at higher temperatures. They are the polystyrene glass transition (T_g) at about

Table 2. Composite component values.

Component	*E*, GPa
	1.16
	3.1
	1.8 (weighted average of 65/35 blend)
	20

[1] Determined in this study.

[2] This is a typical value for wood [12]. It was also used as an approximate value for the treated wood, since the actual value is not known.

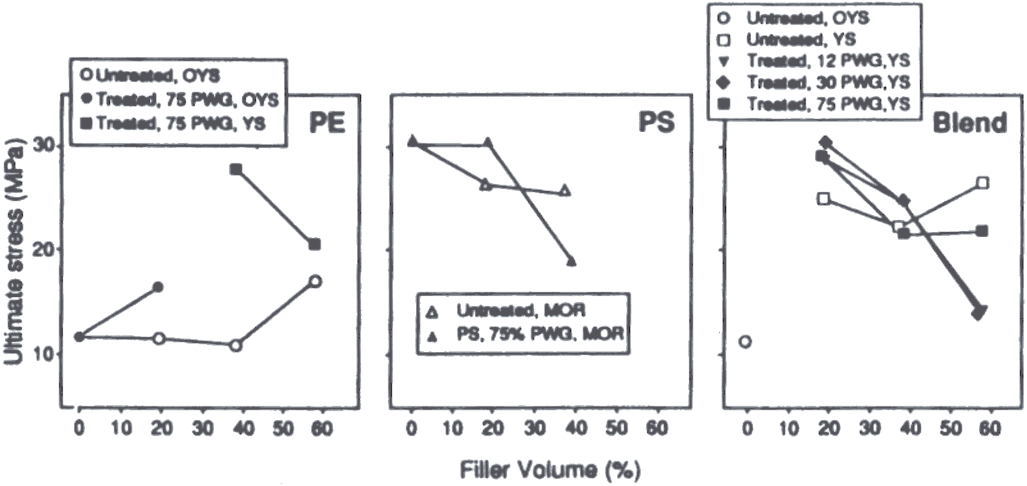


Figure 2. Ultimate stress for PE, PS and blend filled with treated and untreated wood fiber.

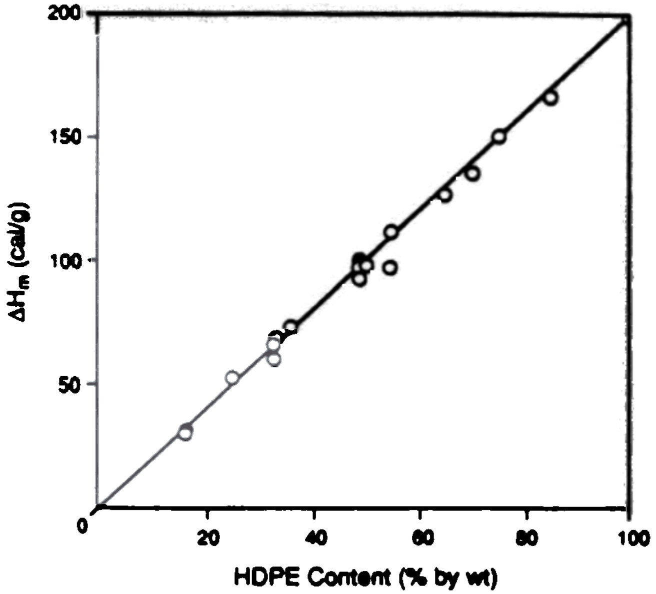


Figure 3. Enthalpy of melting for various samples regardless of plastic/filler ratio or plastic type vs. polyethylene content.

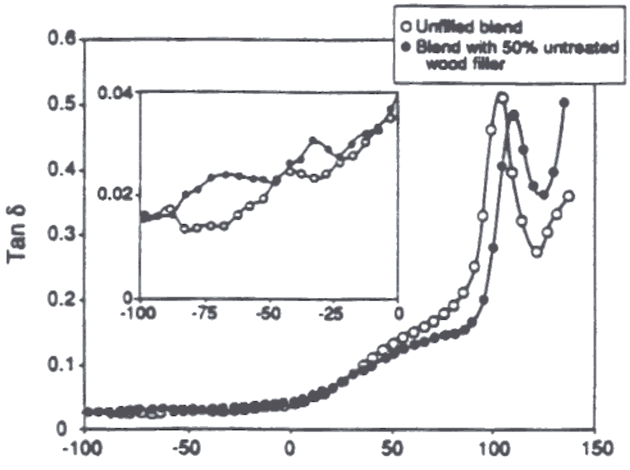


Figure 4. Variation in tan δ with temperature for the unfilled 65/35 PE/PS blend and the same blend with 50% (w/w) untreated wood fiber content.

100°C and the α transition of polyethylene centered at 55°C. Interestingly, the polystyrene T_g is shifted to about 10 degrees higher temperature when untreated wood fiber is incorporated into the blend. This suggests that mobility is restricted, relative to the unfilled blend, and may be due to interaction with the wood fiber. From these observations, it appears that there is a preferential association between the wood fiber and the polystyrene component of the plastic blend.

In comparing the composites in this study, the untreated wood fiber appeared to affect the PS component differently than did the treated fiber. Data from DSC measurements (Figure 5) support this observation. In general, the T_g decreased with increased treatment levels and filler contents. The decrease in T_g with increased treatment level indicates that wood treatment reduced the assumed association between polystyrene and wood. The behavior of the T_g with respect to filler content was complicated and appeared to pass through a maximum. Further experimentation is required in order to elucidate the cause of this maximum. These results suggest that there is a preferential association between the PS and untreated wood. When the blend is filled with a PF-treated fiber, this association is disrupted. This observation is supported by scanning electron microscopy (Figure 6). The composite prepared with the untreated wood fiber [Figure 6(a)] shows considerable domain formation in the plastic matrix. The treated fiber

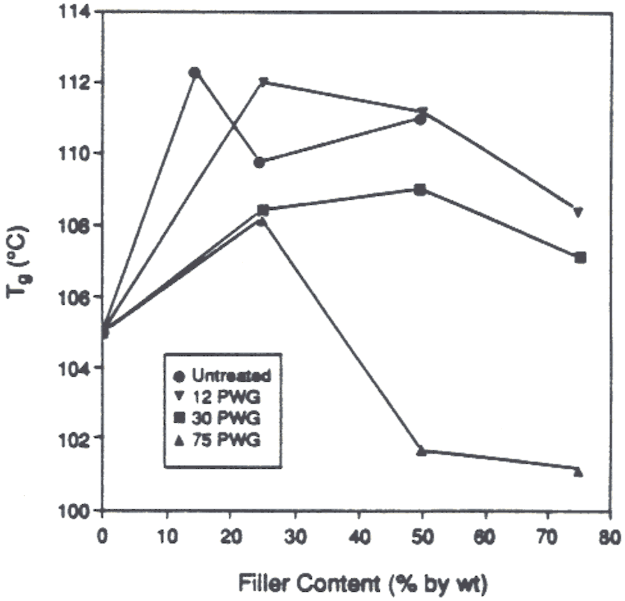
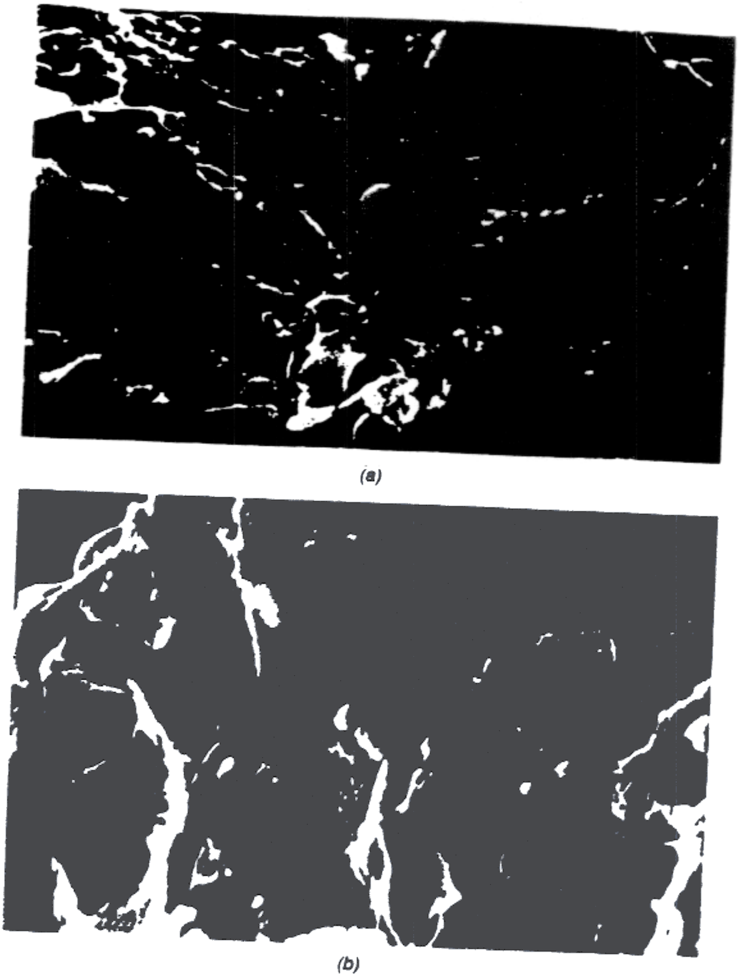


Figure 5. Glass transition temperatures for 65/35 PE/PS blend as a function of filler content and PWG of treated filler.



(a)

(b)

Figure 6. SEM photomicrograph of 65/35 PE/PS blend with (a) untreated wood and (b) treated wood filler.

composite [Figure 6(b)], however, has a much more highly coalesced morphology with much larger phases.

CONCLUSIONS

The addition of wood filler, either treated or untreated, to recycled PE, PS, and a 65/35 PE/PS blend increased the stiffness. Ultimate stresses improved for treated fillers at low loadings in PE. Filled PS showed decreasing ultimate stress with increasing filler content. Treatment of the filler had no effect. The blended polymer samples showed generally decreasing ultimate stresses with increasing filler content, with the exception of the 75 PWG at high loadings.

The differences between the T_g of polystyrene for treated wood and untreated wood samples suggest a possible association between the polystyrene phase and the wood filler phase.

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