The Mechanical Properties of Woodfiber-Plastic Composites: Theoretical vs. Experimental

John Simonsen

Abstract

The use of wood as a reinforcing filler in thermoplastics is discussed. The rule of mixtures for longitudinal and transverse mechanical properties for fiber-reinforced composites are briefly reviewed. Using an estimate for the stiffness of the wood fiber, a reinforcement efficiency factor is deduced from experimental data on wood-polymer composites through the use of a curve-fitting algorithm. Various systems are compared. Theory suggests that an improvement in stiffness of a factor of approximately two to four may be possible, depending upon the system. Strength data from various wood-polymer systems are compared. The simple theory considered here is not helpful in comparing strength values among different systems. The use of styrene-maleic anhydride (SMA) copolymers improved the strength of wood-reinforced SMA composites, in contrast to styrene-based systems. Accelerated aging of samples showed serious degradation in properties, suggesting caution and additional testing before recommending composites such as these for outdoor use.

Introduction

Conventional wisdom holds that although wood fibers have been used as fillers in plastics for a long time, they do not act as reinforcing fillers. This means that while certain properties (e.g., stiffness) increase with filler content, and others (e.g., cost) decrease, the ultimate strength of a composite of wood fibers in a thermoplastic matrix decreases with increasing filler content. This paper explores the theoretical understanding of composites, which comes primarily from mechanical engineering, and what that can tell us about wood fillers in thermoplastics. We will briefly review a simple model for fiber-reinforced composites and use it to compare experimental data. This view of wood-polymer composites can help us in two ways. It gives us a basis from which to make comparisons of composite performance. It also gives us some idea of where we stand compared to what is possible based upon these simple models and the inherent mechanical properties of the components. Results with other fillers and accelerated aging studies will also be presented.

Rule of mixtures

The simplest theory describing the mechanical properties of composite materials is the rule of mixtures. This states that the modulus of a composite is the volume-weighted average of the moduli of the components (5):

$$M_c = \sum V_i M_i \qquad [1]$$

where:

M_c = composite modulus
V_i = volume fraction of component i
M_i = modulus of component i

In our case, we will simplify the number of phases to two: the plastic matrix phase, and the fiber (wood or other filler) phase. We also confine our attention

The author is Assistant Professor, Dept. of Forest Products, Oregon State Univ., Corvallis, Oreg.

to only two moduli: stiffness and strength. Thus we have for stiffness:

$$E_L = V_m E_m + V_f E_f \quad [2]$$

and for strength:

$$\sigma_L = V_m \sigma_m + V_f \sigma_f \quad [3]$$

where:

L = composite modulus in the longitudinal direction (along the fiber axis)
m = matrix
f = fiber

Since there are only two phases:

$$V_m + V_f = 1 \quad [4]$$

Equations [2] and [3] may then be written as:

$$E_L = E_m + V_f(E_f - E_m) \quad [5]$$

$$\sigma_L = \sigma_m + V_f(\sigma_f - \sigma_m) \quad [6]$$

Equations [5] and [6] may be "reduced" by dividing by the matrix modulus to give:

$$\frac{E_L}{E_m} = 1 + V_f\left(\frac{E_f}{E_m} - 1\right) \quad [7]$$

$$\frac{\sigma_L}{\sigma_m} = 1 + V_f\left(\frac{\sigma_f}{\sigma_m} - 1\right) \quad [8]$$

Plots of reduced composite modulus versus V_f will then have an intercept of 1 and a slope proportional to the inherent properties of the components.

The derivation of the rule of mixtures is widely available and not repeated here (5,9,10). However, the major assumptions used are of interest:

1. The strain exhibited by the composite in response to an applied stress is equal in all phases of the composite.
2. Each phase is perfectly elastic.
3. There is perfect bonding between the phases.
4. The fibers are of infinite length (end effects are ignored), parallel, and regularly spaced so that the composite properties are homogeneous everywhere.
5. Component properties are homogeneous within each component.

The rule of mixtures is derived by considering a stress applied in the fiber (longitudinal) direction and assuming a constant strain in both phases. A second model may be developed by considering a stress applied transverse (perpendicular) to the fiber direction and assuming that the stress is equal in both phases. In this case, the fiber and matrix will exhibit different strains. The other assumptions required for the derivation of the rule of mixtures remain the same. The equation derived from these assumptions is generally considered to be a lower bound on the composite properties (9). In the case of stiffness:

$$\frac{1}{E_T} = \sum\left(\frac{V_i}{E_i}\right) \quad [9]$$

where:

E_T = transverse elastic modulus

For the two component system we are considering here, this reduces to (4):

$$E_T = \frac{E_m E_f}{V_m E_f + V_f E_m} \quad [10]$$

Since wood fiber fillers are classified as short fiber composites, we must relax assumption 4. A rigorous derivation is not available for this situation, but a useful approximate solution for the stiffness in a short fiber composite in which the fibers are randomly aligned in three dimensions (E_{c3D}) is (6):

$$E_{c3D} = \left(\frac{1}{5}\right)E_L + \left(\frac{4}{5}\right)E_T \quad [11]$$

where:

E_L = composite stiffness measured in the fiber (or longitudinal) direction (Eq. [5])
E_T = composite stiffness measured in the transverse direction (Eq. [10])

One conclusion this theory provides is the probable improvement in modulus with fiber alignment. E_L from Equation [5] was derived under the assumption of infinitely long, regularly spaced fibers. This is the E we should see if all the fibers were perfectly aligned. If we expand Equation [10] and substitute values for the parameters, we obtain: E_f = 20 GPa, E_m = 1.16 GPa (the value for polyethylene), and V_f = 0.4, E_L/E_{c3D} = 2.58. This value compares well with the industrial rule of thumb that fiber alignment increases the modulus of the composite by approximately two (2).

The rule of mixtures is usually considered to provide an upper bound to what is possible in wood-polymer composites. While this is generally observed to be the case, (in fact, the rule of mixtures typically overpredicts performance), it is not necessarily always true. In the cases where chemical reactions alter the properties of the matrix or fiber or both (e.g., by cross-linking), the rule of mixtures does not apply since the components no longer possess their original properties. In this case it is possible for a composite to possess properties which exceed those predicted by the rule of mixtures.

The rule of mixtures theoretically applies to both the strength and stiffness of the composite. Note that in order to use this equation, we require values for the stiffness and strength of the wood fiber used in the plastic. We will use representative values measured from tests on single fibers: E_f = 20 GPa (2) and σ_f = 750 MPa (11).

The predictability of Equation [8] is typically less than Equation [7]. This is because the ultimate strength often involves the fracture mechanics of the composite, which are extremely complex. Interfacial adhesion, fiber alignment, and stress concentration at fiber ends may significantly effect the ultimate strength (8). Additional complications arise from the failure behavior of the sample. Samples may break cleanly (brittle behavior), or they may "yield" before breaking, meaning that strain increases with no increase in stress, or stress and strain may both increase to the limit of travel of the testing apparatus. Moduli are calculated differently in each case, complicating comparisons of the composites.

We can, however, use this theory to compare different systems with regard to their stiffness (elastic modulus). To do this we modify the parameter for the stiffness of the fiber by substituting ϕE_f for E_f in equations [11]. ϕ is called the efficiency factor and can be used to compare different systems. The equation now becomes:

$$\frac{E_{c3D}}{E_m} = \frac{1}{5}\left(1 + V_f\left(\frac{\phi E_f}{E_m} - 1\right)\right) + \frac{4}{5}\left(\frac{\phi E_f}{V_m \phi E_f + V_f E_m}\right) \quad [12]$$

We plot the reduced modulus as a function of fiber volume fraction using the Marquardt-Levenberg algorithm to fit the data using ϕ as the fit variable (7). Comparing ϕ values for different systems is a means of evaluating the efficiency of the reinforcement provided by the fiber.

Exposure to the elements may also effect the material properties of these composites. The results of an accelerated aging study are presented below.

Materials and methods

Plastics

The polystyrene was contributed by Dow Chemical Company as product Styron 685D. The high-density polyethylene (HDPE) was contributed by Phillips Chemical Company as Marlex EHM 6007. Polypropylene was contributed by Amoco Chemicals as product 7634. Its melt index was 23. Dylark 232 and 332 were contributed by Arco Chemical Co. Dylark 232 is a styrene-maleic anhydride (SMA) copolymer containing about 7 percent maleic anhydride by weight. Dylark 332 contains about 14 percent maleic anhydride.

Straw

Straw was obtained locally in the Willamette Valley, dried and stored outdoors under cover. It was ground to pass a 16-mesh screen in a Wiley mill and dried overnight at 105°C before use. The dried straw flour was stored over a silica gel desiccant. The straw flour was analyzed with an optical measuring system manufactured by Micro Motion Systems, Inc. The average fiber length was found to be 0.7 mm with a standard deviation of 0.6 mm. The ground straw was dusty. The dust was excluded from the particle measurements. The average aspect ratio of the fibers was 4.5 with a standard deviation of 4.7.

Wood fiber

Thermomechanical pulp (TMP) was obtained from the Smurfit Newsprint Corporation mill at Newburg, Oregon. The wood species are primarily hemlock and true firs, with a small amount of spruce. The fiber was obtained from the slurry stream prior to bleaching, air-dried, ground in a Wiley mill to pass a 16-mesh screen, dried in a vacuum at 60°C overnight and stored over a silica gel desiccant. The TMP was analyzed optically. The average length was found to be 1.0 mm with a standard deviation of 0.6 mm. The average aspect ratio was 22 with a standard deviation of 14. The pulp also contained dust, although not as much as the straw. The dust was not included in the dimension measurements.

Wood flour made from Douglas-fir (*Pseudotsuga menziesii* (Mirb.) Franco) ground to 80 to 100 mesh was obtained from the Menasha Wood Corporation, Olympia, Washington as product T-14. The T-14 was dried in a vacuum at 60°C overnight and stored over a silica gel desiccant. This filler was analyzed optically and found to have an average length of 0.9 mm with a standard deviation of 0.6 mm. The average aspect ratio was 9 with a standard deviation of 5.

E-glass fiber

E-glass fiber was contributed by Owens Corning as product 731-ED, 1/8-inch milled fiber.

Sample preparation

The plastics and fillers were blended in a Brabender Plasticorder set at 30 rpm and 350°F. The plastic

was added first and blended for 3 to 5 minutes to ensure complete melting. The filler (wood flour, TMP, straw, or E-glass) was then added and blended. The Plasticorder contained a torque rheometer which indirectly measured the melt viscosity. Typically the melt viscosity rose when filler was added to the plastic melt. The viscosity then fell as the filler blended into the plastic. When the filler was completely blended, the melt viscosity plateaued at a value higher than that of the plastic alone. When the melt viscosity plateaued, as determined by the torque rheometer, the blending was continued for an additional 2 minutes. It was assumed that the dispersion was maximized at that point. The melt was then removed from the Plasticorder, cooled, and stored for grinding.

The prepared blends were ground in a Wiley mill to a particle size of approximately 3 mm (0.1 in.). These powders were then compression-molded in a thermostatted Carver laboratory press. The press conditions were 180°C (360°F) and 6.9 MPa (1,000 psi) for 10 minutes. The sample size was approximately 2 by 13 by 55 mm (0.08 by 0.5 by 2 in.).

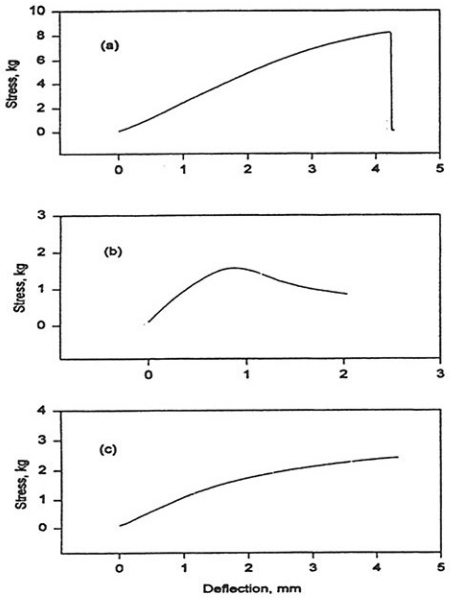

FIGURE 1.—*Examples of stress-strain curves for plastics: a) MOR (brittle fracture); b) yield strength (yielding behavior); 3) offset yield strength (ductile behavior).*

Material properties

The modulus of elasticity (MOE) was determined on samples tested in flexure using a three-point bending apparatus in accordance with ASTM D 790-86. Five samples were tested for each determination of MOE wherever possible. In some cases fewer samples were used due to material limitations.

In this report, ultimate strength is defined in three different ways depending upon the fracture behavior of the material. Figure 1 depicts examples of the different types.

1. Modulus of rupture. In this case the stress increased with strain until samples broke cleanly. This is brittle fracture.
2. Yield strength. Some samples typically did not break, but rather yielded under the stress. The yield strength was defined as the first point at which the stress-strain curve shows a slope of zero. This point is reported for those samples that either yielded before breaking or did not break at all (i.e., they simply continued to bend to the limits of travel of the testing machine).
3. Offset yield strength. These samples neither broke nor showed a yield point. The stress-strain curve in these cases did not show a slope of zero, but rather was a rising smooth curve to the limits of travel of the testing machine. In these cases the ultimate strength was reported as the 0.5 percent strain offset yield strength. This was calculated with the method specified in ASTM D 790-86 and described in ASTM D 638-89.

Accelerated aging

The samples were boiled for 10 minutes in distilled water, then stored in an environmental chamber maintained at 90°F and 35 percent relative humidity overnight. This cycle was repeated five times (1). The samples selected for this procedure were polystyrene and polyethylene filled with either straw or Douglas-fir wood flour.

Results and discussion

Stiffness

The use of reduced moduli allows the comparison of different systems on a more even basis. However, the drawback of using this comparison is that the slope of the reduced modulus versus filler content curve depends upon the ratio of the filler to matrix modulus (E_f/E_m). Thus a weaker matrix will show relatively greater improvements in moduli for the same filler. For this reason, comparisons are made between various fillers in the same matrix. Even so, there are various grades of the thermoplastics studied here, each with a different modulus. This does not introduce large errors in the comparison, but should be noted. The elastic moduli of polypropylene filled with a variety of fillers is shown in Figure 2. Wood-based fillers appear to perform similarly to calcium carbonate, a particulate filler. Glass and carbon fillers show superior performance to wood at equivalent filler contents.

The stiffness of a variety of fillers in polyethylene is shown in Figure 3. The use of stearic acid as a dispersing agent gave an increase in stiffness for the TMP-filled composites. The use of dibutylphthalate had no effect. Straw performed better than TMP in this matrix. This may indicate superior compatibility between the fiber and the matrix in the case of straw. E-glass was superior to the other fillers tested.

Stiffnesses for filled styrene polymers are shown in Figure 4. The elastic moduli of the unfilled SMA copolymers was similar to polystyrene (3.3 for SMA vs. 3.1 for polystyrene), so the comparisons shown here can be considered valid. Wood fillers performed similarly whether in polystyrene or SMA. Straw appeared to give slightly inferior performance at higher filler contents. Since the straw was slightly superior as a filler in HDPE, there may have been a difference in compatibility between filler and matrix for these two materials, with wood more compatible with styrene and straw more compatible with HDPE. Again, glass outperformed the other fillers in terms of stiffness increase with filler content.

Various matrices filled with E-glass are compared in Figure 5. The data were fit using the Marquardt-Levenberg algorithm. The efficiency, ϕ, was selected as the fit variable. Since HDPE has the least stiff

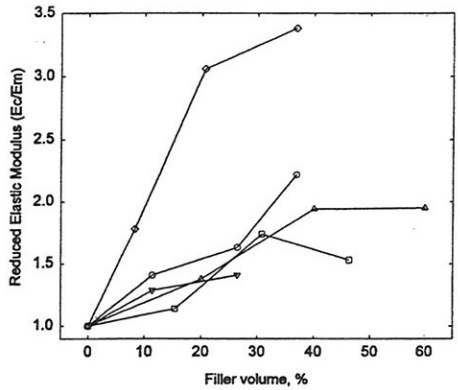

FIGURE 3.—*Elastic moduli of polyethylene filled with chopped E-glass (◇); TMP with 5 percent stearic acid (○); straw (△); TMP with no additives (□); TMP with 5 percent dibutyl phthallate (▽).*

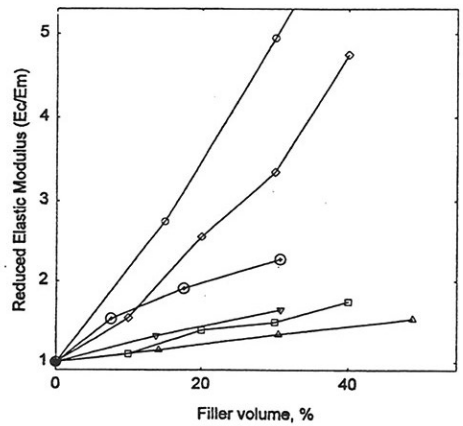

FIGURE 2.—*Elastic moduli of polypropylene filled with carbon fiber (○); commercial glass fiber (◇) (Heathcott 1994); chopped E-glass (✦); TMP (▽); calcium carbonate (□); and wood flour (△).*

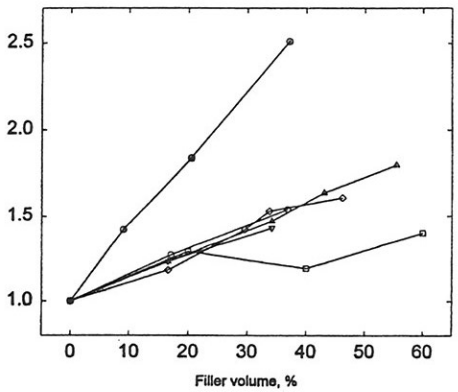

FIGURE 4.—*Elastic moduli of polystyrene filled with chopped E-glass (✦); wood flour (△); TMP (◇); straw (□); SMA copolymer with 7 percent maleic anhydride content filled with TMP (▽); and SMA copolymer with 14 percent maleic anhydride content filled with TMP carbon fiber (○).*

matrix, it showed relatively higher increases of reduced stiffness with filler content. However, the greater increase in stiffness may have also been due to superior compatibility. It is difficult to separate the factors in this case. The performance of the HDPE composite also dropped off at high filler contents. This did not appear to be the case with polystyrene. The polypropylene system showed a curvilinear behavior which may be due to incomplete dispersion of the filler in the matrix.

Figure 6 compares wood fillers in various matrices. There did not appear to be one system that excelled above the others, except that the addition of stearic acid improved the stiffness compared to HDPE containing no additives. The apparent superiority of HDPE may be due to the lower stiffness of the matrix. The curves were fit to the data using the Marquardt-Levenberg algorithm.

The efficiency factors (ϕ) determined by fitting the data in Figures 5 and 6 using Equation [12] are presented in Figure 7. HDPE filled with E-glass gave the highest value for ϕ. Glass in polypropylene and polystyrene showed similar efficiencies to wood-filled systems. The efficiency values for wood varied from 25 percent for wood flour-filled polypropylene to 63 percent for TMP-filled polystyrene. The efficiency factors for these systems vary by a factor of about two. It is apparent that for the better performing systems, an additional improvement by a factor of two would reach about 100 percent efficiency.

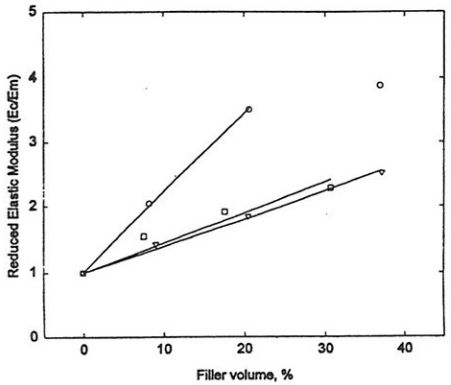

FIGURE 5.—*Reduced elastic moduli of polymers filled with chopped E-glass: polyethylene (○); polypropylene (□) (Heathcott 1994); and polystyrene (▽). Experimental data fit via the Marquardt-Levenberg algorithm.*

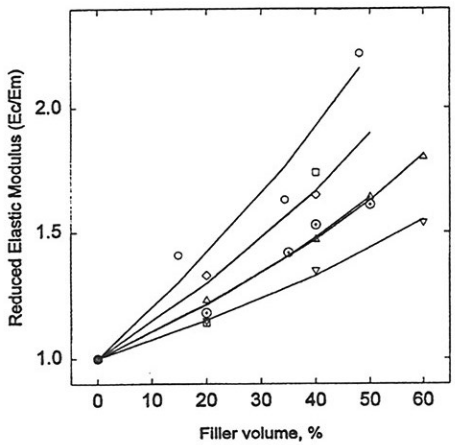

FIGURE 6.—*Reduced elastic moduli of filled polymers: polyethylene filled with TMP and 5 percent stearic acid (○); polyethylene filled with TMP, no additives (□); polypropylene filled with wood flour (▽); polypropylene filled with TMP (◇); polystyrene filled with wood flour (△); polystyrene filled with TMP (◈); experimental data fit via the Marquardt-Levenberg algorithm.*

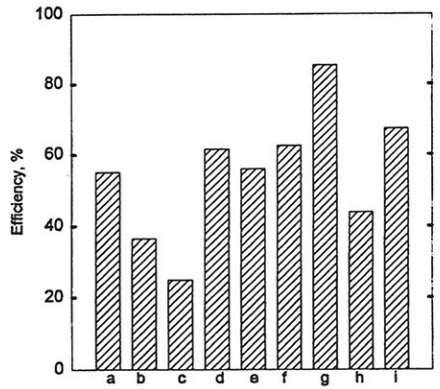

FIGURE 7.—*Comparison of efficiency factors for elastic moduli in filled plastics: polyethylene filled with TMP and 5 percent stearic acid (a); polyethylene filled with TMP, no additives (b); polypropylene filled with wood flour (c); polypropylene filled with TMP (d); polystyrene filled with wood flour (e); polystyrene filled with TMP (f); polyethylene filled with E-glass (g); polypropylene filled with E-glass (h); polystyrene filled with E-glass (i); experimental data fit via the Marquardt-Levenberg algorithm.*

This would approach the theoretical limit for these systems, assuming the value for E_f was accurate.

Strength

Comparisons of ultimate strengths among different systems is complicated by their fracture behavior (see "Materials and methods" section). Because of this, comparisons of different materials, or even the same material exhibiting different fracture behavior at different filler contents, are not valid.

Typically, the strengths of filled thermoplastics fall into two general classes. These are shown in Figure 8. Reinforcing fillers show increasing strength with increasing filler content, while particulate fillers show decreasing strength. One way to define whether a filler is reinforcing or not is from the slope of the plot of ultimate strength versus V_f. A positive slope is reinforcing. Simple theoretical models all predict reinforcement for fillers which are stronger than the matrix. The data obtained in this study, with some exceptions, showed decreasing strengths with increasing filler content. Consequently, the strength data in this study were not compared to theory. The simple theory outlined here does not help us to understand the strength behavior of these materials. We can compare the slope of the line in strength versus filler content graphs to compare the reinforcing ability of different systems, but we can only do this between systems with the same plastic matrix and showing the same type of failure behavior.

Strength data for polypropylene are shown in Figure 9. The only systems which exhibited reinforcement were from a commercially prepared glass-filled polypropylene (4) and a compatibilized recycled newsprint filler (11). The remaining systems tested all showed the behavior of particulate systems. These data support the importance of compatibilizing the filler with the matrix.

Strength data for HDPE are shown in Figure 10. Here the fracture behavior changes with filler content for wood and straw-filled composites. At low filler contents, the samples showed ductile fracture reported as offset yield strength. The higher filler contents yielded. The glass-filled samples showed offset yield strength behavior throughout the filler content levels investigated. In this case, straw appeared to perform slightly superior to TMP. This is similar to the pattern observed for stiffness. At higher filler levels, all systems seem to exhibit decreasing strength. This may be due to decreased wetting of the fiber by the matrix as the ratio of filler surface area to matrix volume reached high values.

Ultimate strengths for styrene systems are shown in Figure 11. All the fillers tested in polystyrene, including E-glass, showed particulate behavior, despite the fact that some of them were fibers. In the case of styrene, a brittle polymer, all the strength data are reported as modulus of rupture. The SMA copolymers showed markedly different behavior to polystyrene with TMP filler. In this case the strength increased with increasing filler up to about 17 percent filler for both 7 and 14 percent maleic anhydride contents and strength increased again at 37 percent filler content for the 14 percent maleic anhydride sample. This behavior is similar to that

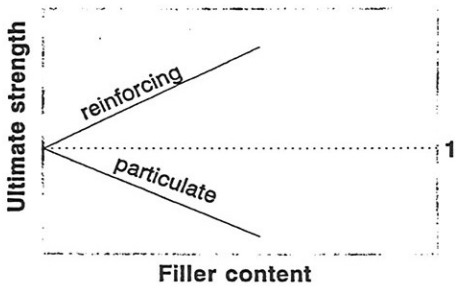

FIGURE 8.—*Typical behavior of samples filled with reinforcing fibers and particulate (nonreinforcing) fillers.*

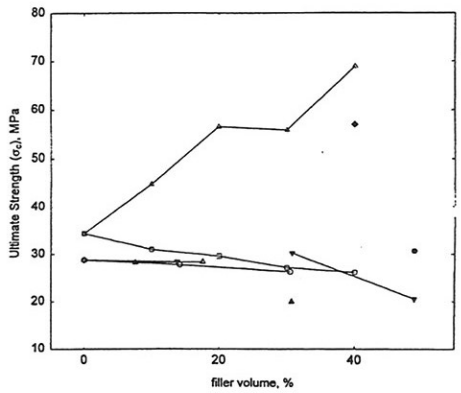

FIGURE 9.—*Ultimate strengths of polypropylene filled with: compatibilized glass (Heathcott 1994) (△); TMP, offset yield strength (▽); TMP, yield strength (▼); chopped E-glass (◇); calcium carbonate (Heathcott 1994) (□); wood flour, offset yield strength (●); recycled newspaper (Sanadi 1994) (◆).*

observed in polypropylene and maleic anhydride-modified polypropylene (5). While we have no strong evidence that a similar mechanism is active in both plastics, the coincidental behavior is apparent.

Accelerated aging

HDPE filled with straw and wood flour were subjected to an accelerated aging procedure. All samples tested showed a degradation in properties upon accelerated aging (Figs. 12 and 13). The drop was especially severe in stiffness. Strength appeared to be reduced, but the reduction was not significant.

Similar data for polystyrene are shown in Figures 14 and 15. These samples all showed large decreases in both strength and stiffness. The reduction in stiffness was especially pronounced.

These data indicate that composites of wood or other biofillers in thermoplastics may not be impervious to the effects of outdoor exposure. Before recommending these composites for outdoor use, careful testing should be performed.

Conclusions

In the case of stiffness, the rule of mixtures appears to be a reasonable model for wood-filled thermoplastics. Since wood filler stiffness efficiencies varied from about 25 percent to about 63 percent, improvements in stiffness through better bonding between matrix and filler will probably be limited to a factor of about two to perhaps four, depending upon the system. An additional factor of approximately two may be gained in the fiber direction by orienting the fibers.

Strength properties do not conform to simple models and generally decreased with increasing filler content for the systems studied here, with the exceptions of glass-filled polypropylene and HDPE and wood-filled SMA.

Accelerated aging studies show a serious degradation in mechanical properties upon exposure. These results indicate that caution should be used before recommending that wood-filled thermoplastics be used in outdoor environments.

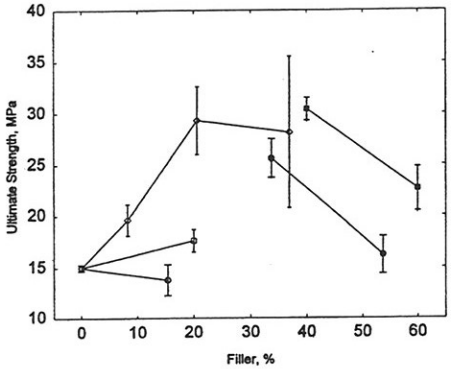

FIGURE 10.—*Ultimate strengths of polyethylene filled with: chopped E-glass, offset yield strength (◇); TMP, offset yield strength (○); TMP, yield strength (●); straw, offset yield strength (□); straw, yield strength (■).*

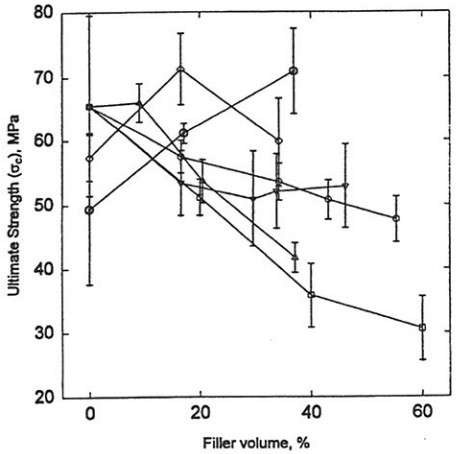

FIGURE 11.—*Ultimate strengths (MOR) for styrenic polymers: SMA with 14 percent maleic anhydride content (◈); SMA with 7 percent maleic anhydride content, filled with TMP (◇); polystyrene filled with chopped E-glass (△); polystyrene filled with wood flour (○); polystyrene filled with TMP (▽); polystyrene filled with straw (□).*

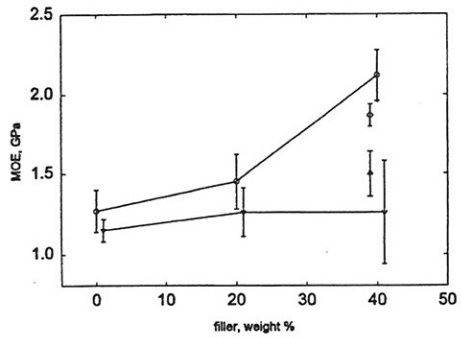

FIGURE 12.—*Comparison of elastic moduli for polyethylene filled with: straw, before accelerated aging (○); straw, after accelerated aging (▽); wood flour, before accelerated aging (◇); wood flour, after accelerated aging (△).*

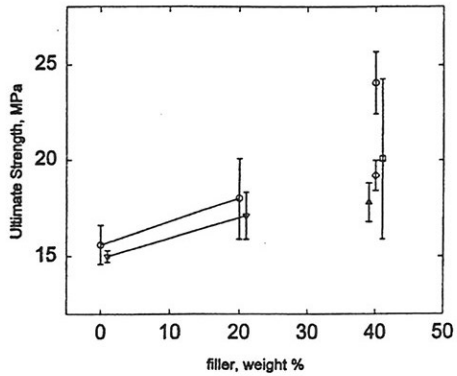

FIGURE 13.—*Comparison of ultimate strengths for polyethylene filled with: straw, offset yield strength before accelerated aging (○); straw, offset yield strength after accelerated aging (▽); straw, yield strength before accelerated aging (◈); straw, yield strength after accelerated aging (□); wood flour, offset yield strength before accelerated aging (◇); wood flour, offset yield strength after accelerated aging (△).*

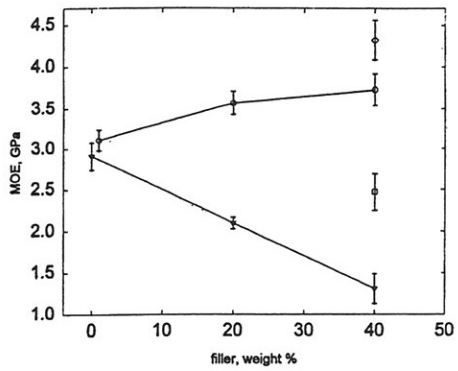

FIGURE 14.—*Comparison of elastic moduli for polystyrene filled with: straw, before accelerated aging (○); straw, after accelerated aging (▽); wood flour, before accelerated aging (◇); wood flour, after accelerated aging (□).*

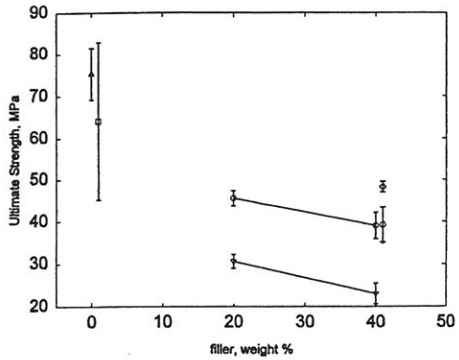

FIGURE 15.—*Comparison of ultimate strengths for polystyrene filled with: straw, MOR before accelerated aging (△); straw, MOR after accelerated aging (□); straw, yield strength before accelerated aging (○); straw, yield strength after accelerated aging (▽); wood flour, yield strength before accelerated aging (◇); wood flour, yield strength after accelerated aging (◈).*

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