A Comparative Study on the Effect of Aging on Mechanical Properties of LLDPE–Glass Fiber, Mica, and Wood Fiber Composites

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Synopsis

Linear low density polyethylene (LLDPE) was reinforced with wood fiber, glass fiber, and mica. The effect of aging on mechanical properties of the composites was examined under different conditions: (i) exposure at 105°C for 7 days; (ii) immersion in boiling water for 4 h. Samples containing glass fibers showed by far the best results with regard to tensile strength, elongation, and fracture energy. LLDPE filled with mica produced poor results compared to wood fiber composites. Dimensional stability of LLDPE–wood fiber composites, after boiling water treatment, was inferior to mica and glass fiber composites.

INTRODUCTION

Recently, there has been a growing interest in fiber reinforced composites due to their great versatility and high performance.[1] The choice of a plastic for high temperature performance is one of the most important and complex tasks facing the designer and materials engineer. Virtually all properties of plastics are affected by a raise in temperature, but in different ways and to different degrees. Therefore, an independent approach in each case is required from the standpoint of understanding the behavior of plastics and their application to material selection.

All polymers absorb moisture in a humid atmosphere and when they are immersed in water. The sorption of water by nonpolar polymers containing a filler depends mainly on the nature of the filler. For hydrophilic fillers such as cellulosic fibers, an increase in water sorption may be expected. Kinetics and equilibria of water sorption in PE–cellulose composites were studied by Sapieha et al.[2] The use of water absorption kinetic data to predict laminate property changes in unsaturated polyester–glass fiber composites was discussed by Pritchard and Speak.[3]

Many studies have been published concerning the processing conditions and properties of thermoplastics with wood fiber,[4-7] glass fiber,[8,9] mica,[10] and $CaCo_3$.[11] But there is not much literature available on the aging studies of thermoplastics filled with cellulosic fillers. This study investigates the effect of aging on mechanical properties of linear low density polyethylene (LLDPE) filled with glass fiber, mica, and wood fiber. The composites were subjected to aging at 105°C for 7 days or immersed in boiling water for 4 h.

Journal of Applied Polymer Science, Vol. 40, 645–655 (1990)
 CCC 0021-8995/90/5-60645-11$04.00

EXPERIMENTAL

Materials

Linear low density polyethylene (LLGR-0534-A) was supplied by Novacor Ltd. (melt index = 5.0 g/10 min; density = 0.934 g/cc). The chemithermomechanical pulp (CTMP) of aspen was prepared in a Sund defibrator.[12] The average fiber aspect ratio (L/D) was 11.9. Glass fiber 731 BA 1/32 (0.8 mm, silane coated) was supplied by Fiber Glass of Canada. Mica 200-NP-Suzorite (200 mesh, silane coated) was supplied by Marietta Co., Montreal.

Preparation of Composites

The CTMP aspen was dried at 55°C in an air-circulating oven for 24 h before being mixed with the polymer. Compounding of polymer and filler was performed in a Roll Mill (C. W. Brabender Laboratory Prep. Mill No. 065). Usually about 40–50 g of polymer was mixed with fibers at 150°C. The concentration of the fiber varied from 0 to 40% by weight of fiber. In the case of wood fiber, an adhesion promoting agent (polymethylenepolyphenyl isocyanate, 3% by weight of the polymer) was added during the mixing process. Glass fiber and mica were used as supplied by the manufacturer.

The resulting mixture was collected, remixed 5–10 times, allowed to cool to room temperature, and then ground to mesh size 20. The above mixture was compression-molded at 155°C (pressure 3.2 MPa) into dog-bone-shaped tensile specimens.

Aging Studies

The composites were subjected to different environmental conditions:

(a) Conditioned at room temperature (not subjected to any treatment).
(b) Samples were kept at 105°C in an air-circulating oven for 7 days and then slowly cooled to room temperature. The above samples were conditioned at room temperature before testing.
(c) Samples were immersed in boiling water for 4 h and then dried between two sheets of filter paper. The above samples were conditioned at room temperature before testing.

Mechanical Tests

Tensile properties of the composites were studied with the help of an Instron Model 4201. The reported composite properties were measured at peak load. A minimum of six samples were tested in each series. The results were automatically calculated by an HP86B computing system using an Instron 2412005 General Tensile Test Program. The coefficient of variation was less than 8.0%.

RESULTS AND DISCUSSION

Experimental results of LLDPE filled with wood fiber, glass fiber, and mica subjected to different environmental conditions are summarized in Figures

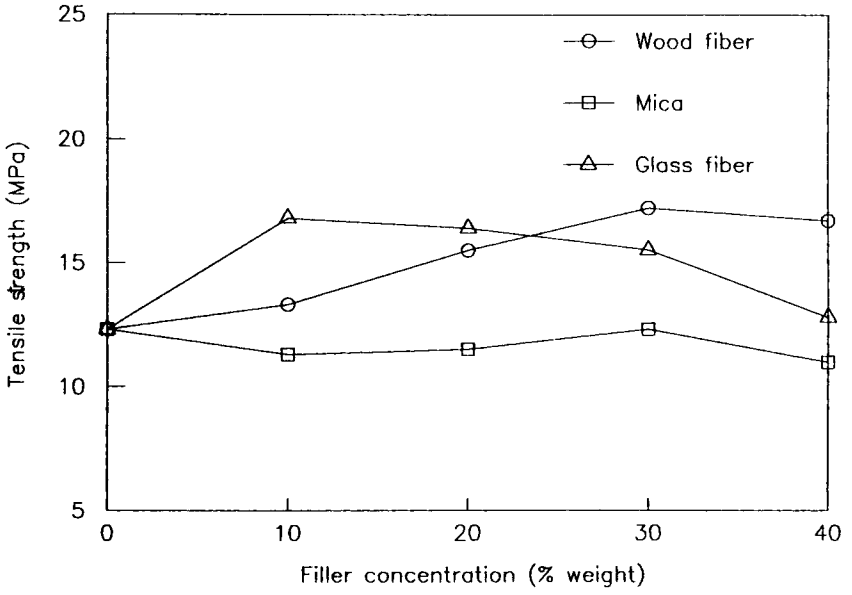

Fig. 1.

1–12. The tensile properties of the composites are presented as a function of the weight fractions of the filler.

Effect of Aging on Tensile Strength

Figures 1–3 show the tensile strength of composites, as a function of filler concentration, after being subjected to different aging treatments. Composites

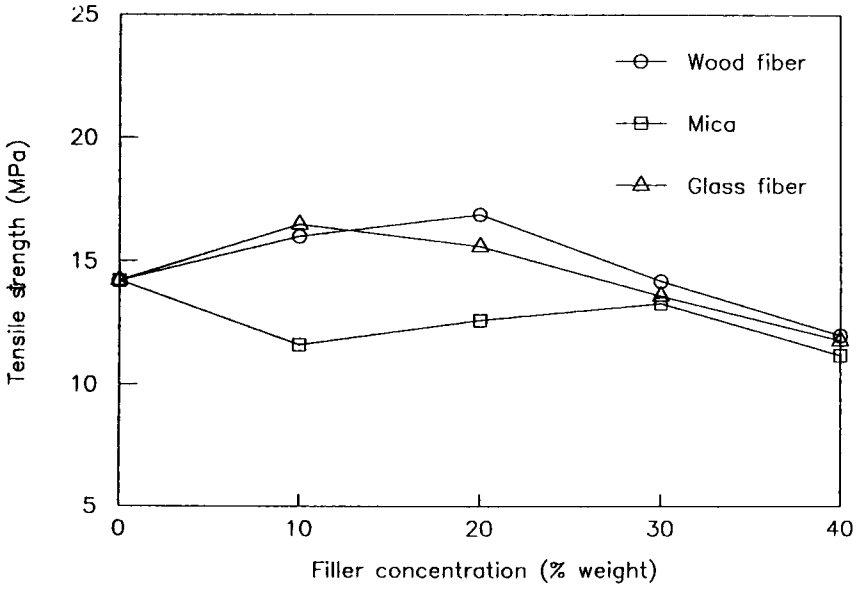

Fig. 2.

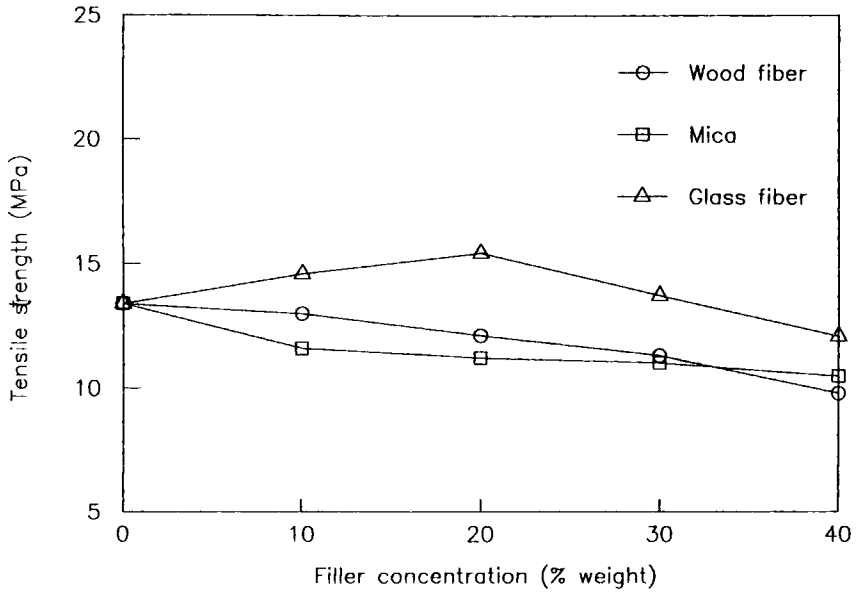

Fig. 3.

which are not subjected to any aging showed an increase in strength, at lower filler concentrations, in LLDPE–glass fiber composites (Fig. 1). But at higher filler content (30 and 40%), samples containing wood fiber produced slightly higher strength values compared to glass fiber and mica composites. This may be possibly due to the better dispersion of wood fibers in the matrix. Wood fibers are relatively short and coarse, which is an advantage for higher loading of the filler in the polymer. Synthetic fibers are relatively straight and smooth-surfaced, whereas the wood fiber is twisted and kinked, with an irregular surface. Thus the wood fiber is likely to offer greater resistance to withdrawal from the matrix than synthetic fibers.[13]

In the case of composites exposed at 105°C for 7 days, LLDPE–glass fiber composites showed a better retention of strength compared to wood fiber composites (Fig. 2). After aging, LLDPE–glass fiber composites retained 89.9% of the strength compared to 81.3% of wood fiber composites at 30% filler concentration. The deterioration of strength is affected by a number of factors which are interrelated: (a) thermal degradation of the matrix, (b) the loss of strength of fiber, and (c) poor filler–matrix adhesion.

Decrease in strength was also observed in the case of samples immersed in boiling water for 4 h (Fig. 3). There was not much difference in the strength values of LLDPE–glass fiber composites with the increase in filler concentration, while, in wood fiber composites, the strength decreased steadily as the concentration of the filler increased in the samples. At 30% filler concentration, the loss in strength was 35.4% in wood fiber composites compared to 11.2% in glass fiber composites. The mica composites performed poorly compared to wood fiber composites. In glass fiber composites, since the fibers do not absorb water and the absorbed water is concentrated within the matrix phase, the loss of strength is due to the degradation of bonding at the

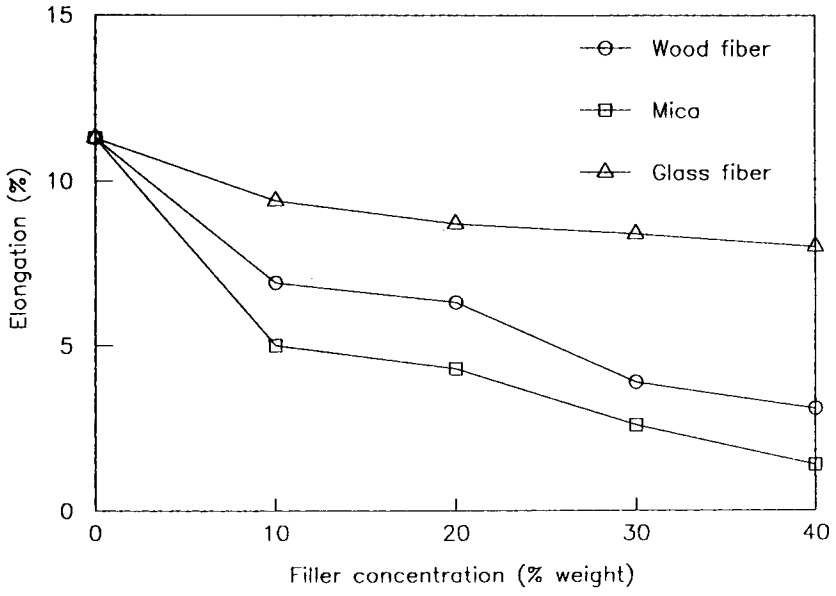

Fig. 4.

interface, whereas in wood fiber composites the situation is more complicated. In addition to the matrix, the fiber also absorbs moisture. The water uptake of the cellulose fiber depends on the concentration of the filler and temperature.[2] When the matrix–filler interface is accessible to the moisture from the environment, the wood fibers tend to swell. This results in the development of shear stresses at the interface, which leads to the ultimate debonding of the fiber.

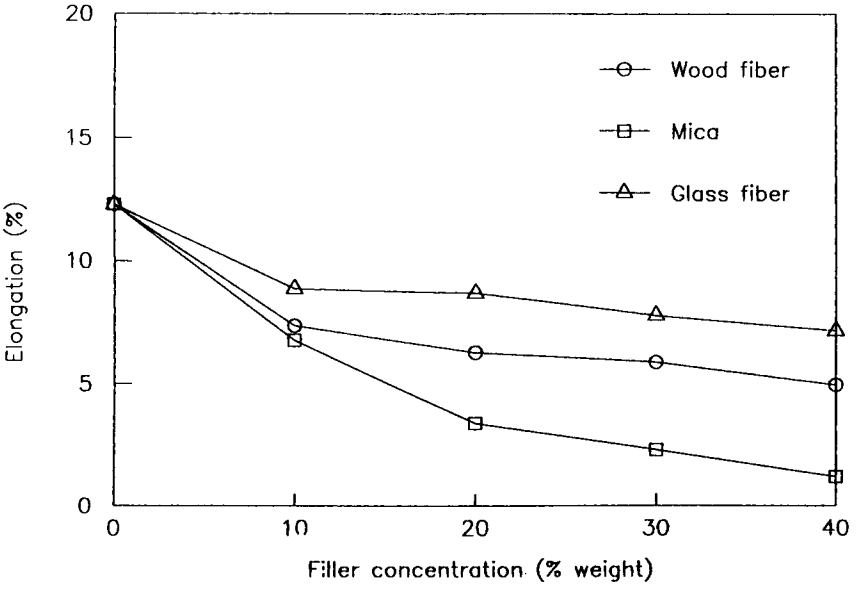

Fig. 5.

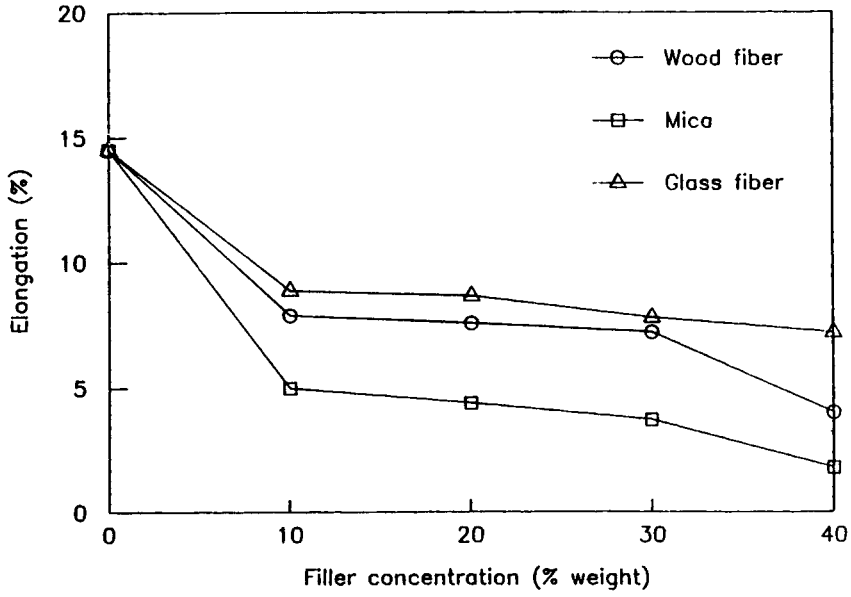

Fig. 6.

Effect of Aging on Elongation

The effect of aging and boiling water treatment on elongation of the composites is illustrated in Figures 4–6. The elongation generally decreased with the increase in filler concentration (Fig. 4). This is mainly due to the lower elongation values of the filler than polymer. Also, since the addition of filler increases the stiffness of the matrix, a loss in ductility of the composite is expected. LLDPE–glass fiber composites showed higher elongation values than wood fiber composites. But the samples filled with mica were brittle and the elongation dropped rapidly as the filler concentration increased. This is possibly due to the physical nature of the filler. The elongation was not much affected by aging, at 105°C for 7 days, as can be seen from Fig. 5.

But the situation was different in the case of composites subjected to boiling water treatment. Elongation decreased rapidly in all the cases, irrespective of the nature of the filler, as the concentration of filler increased (Fig. 6). The samples containing glass fibers lost more than 48% of elongation at 40% filler concentration compared to unfilled polymer.

Effect of Aging on Fracture Energy

Fracture energy (area under the stress–strain curve) increased significantly, at higher filler concentrations, in LLDPE–glass fiber composites (Fig. 7). The lower fracture energy values of the wood fiber composites are possibly due to the relative shortness of the wood fiber, which reduces the effective load bearing ability of the fiber in the matrix. In mica composites, the fracture energy decreased drastically with the increase in filler concentration. This is because the flakes or platelets have poorer efficiency of stress transfer than fibers. After exposure at 105°C for 7 days, the fracture energy decreased

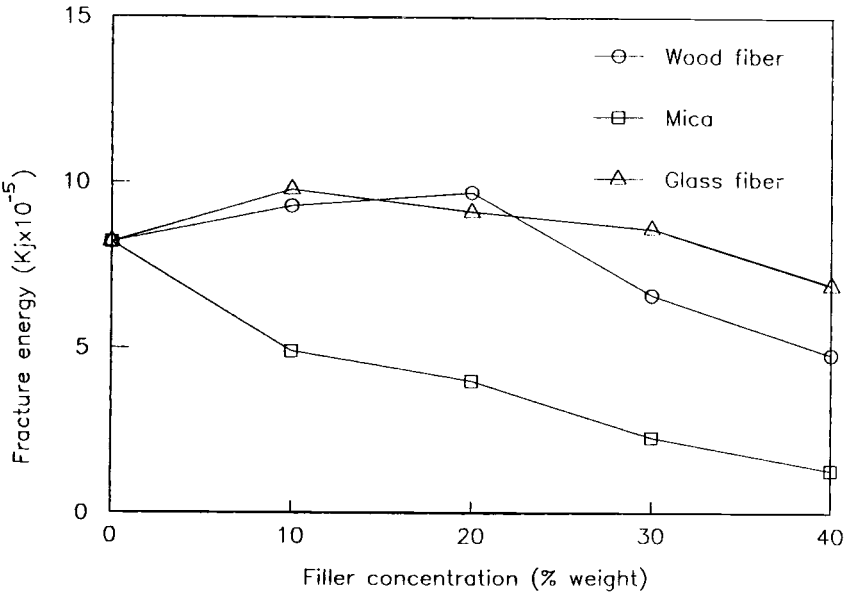

Fig. 7.

steadily in LLDPE-wood fiber composites (Fig. 8). The mica composites showed poor fracture energy absorption compared to glass fiber and wood fiber composites.

The fracture energy decreased rapidly as the filler concentration increased in the samples (Fig. 9). LLDPE filled with wood fibers lost 46% of the fracture energy at 30% filler level. Several different interactions have been identified as

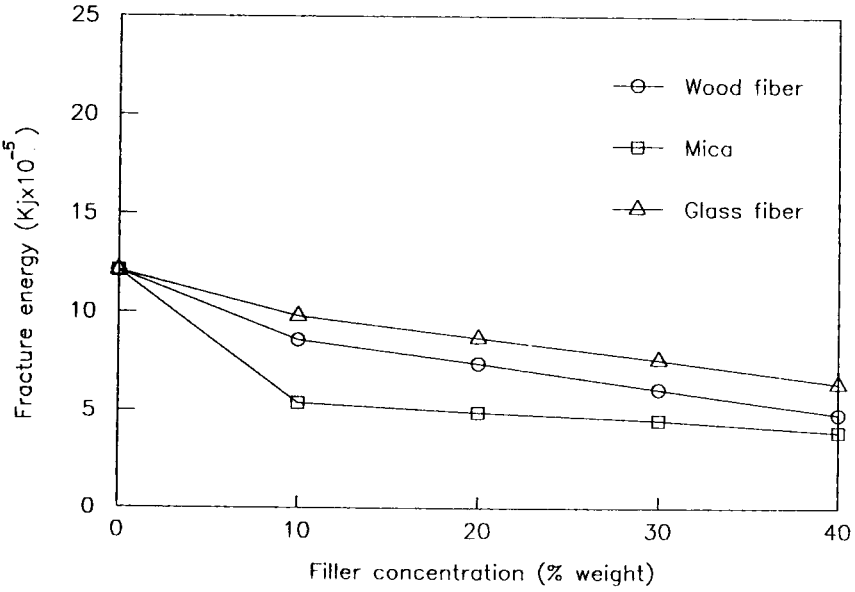

Fig. 8.

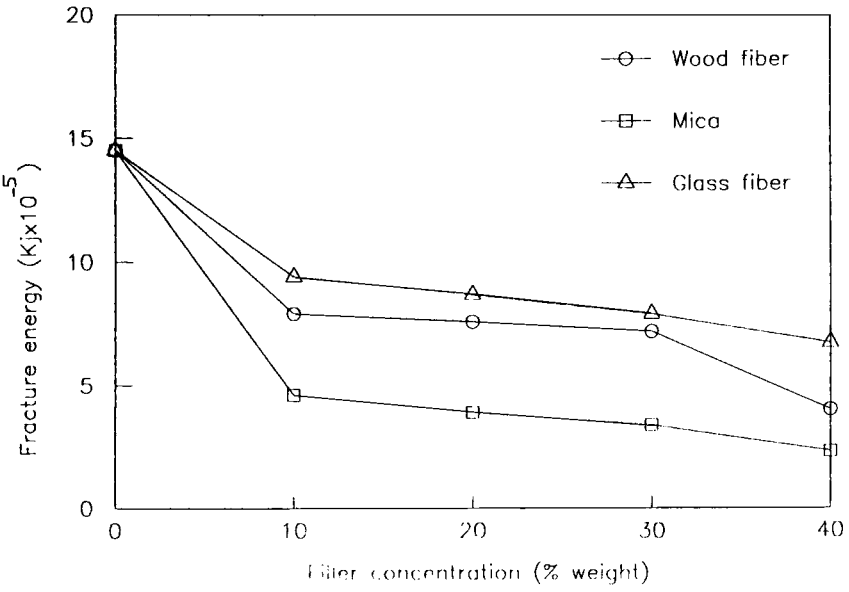

Fig. 9.

contributors to fracture: ductilities of the fiber and the matrix, interfacial fracture energy and concentration of stress at the matrix–filler interface.[14] The lower fracture energy values of the composites, after boiling water treatment, may be due to several factors: amount of water absorbed at the interface, loss in fiber strength, and the degradation of bonding at the fiber–matrix interface.

Effect of Aging on Modulus

Tensile modulus as a function of filler concentration is presented in Figure 10. A steady increase in the modulus was observed as the filler concentration increased in the composites. The best increase in modulus was observed in LLDPE–mica composites, 1.2 GPa at 40% filler concentration compared to 0.34 GPa of unfilled LLDPE. Since the modulus of the matrix is constant, the relative packing efficiencies of the filler is an important factor in determining the modulus of the composite.[13] Samples containing mica and wood fibers after exposure to 105°C for 7 days showed a slight increase in modulus as seen from Figure 11. However, the heat aging had little influence on the modulus of LLDPE–glass fiber composites.

But the modulus dropped rapidly after boiling water treatment (Fig. 12). LLDPE filled with wood fibers suffered a 31% loss in modulus at 40% filler concentration. The loss in modulus in LLDPE–wood fiber composites was expected. Because of its hydrophilic nature, the wood fiber absorbs more water than inorganic fillers. At higher water contents the stiffness of the cellulose fiber drops considerably. This has been explained as being due to softening of the disordered zones of the cellulose microfibrils,[15] whereas the situation is different in glass fiber and mica composites. In this case, since the filler does not absorb water, the degree of water mobility at the interface is

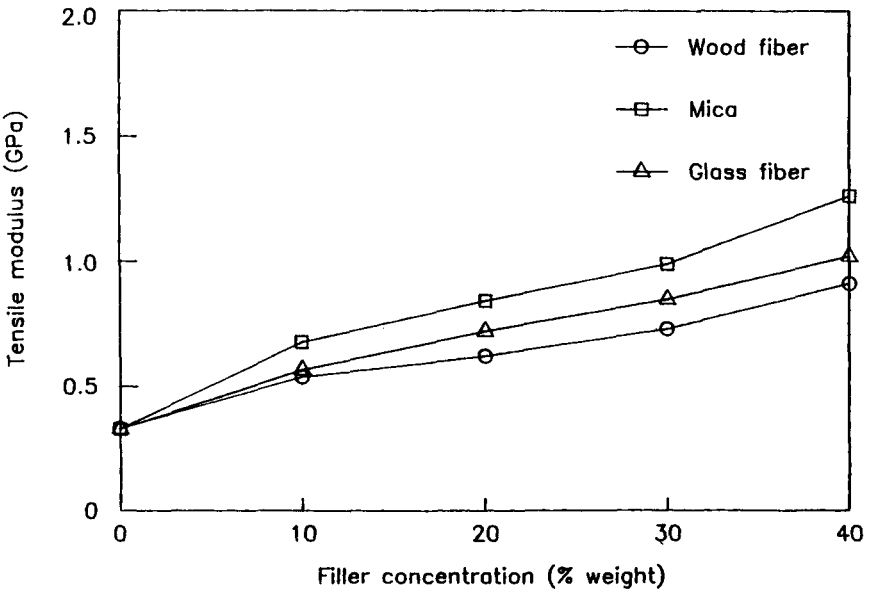

Fig. 10.

greatly reduced. So the loss in modulus is mainly due to the degradation of matrix phase.

Table I compares the dimensional stability of LLDPE reinforced with different fibers after 4 h immersion in boiling water. The variation in the cross-sectional area of wood fiber filled LLDPE was 3.87% at 40% filler concentration compared to 2.79% for glass fiber composites. Since cellulosic

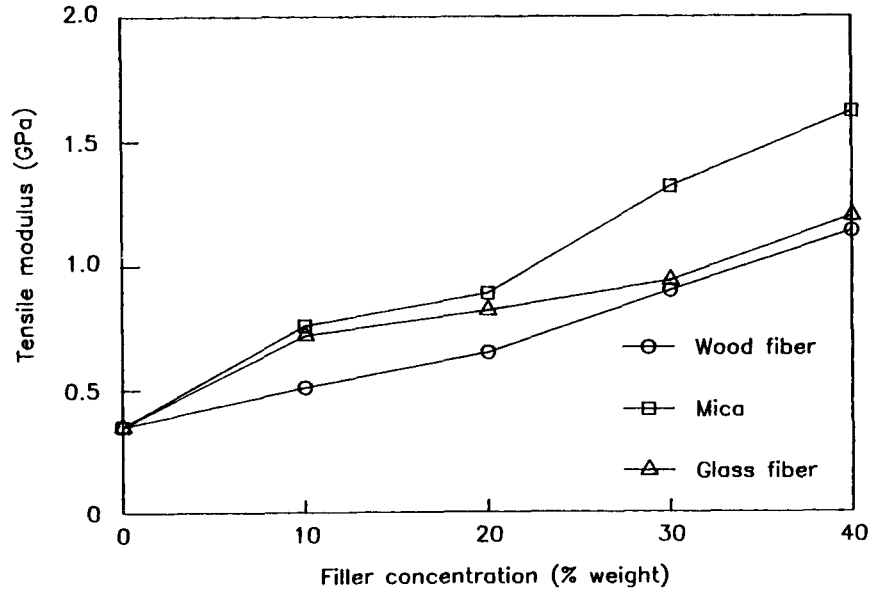

Fig. 11.

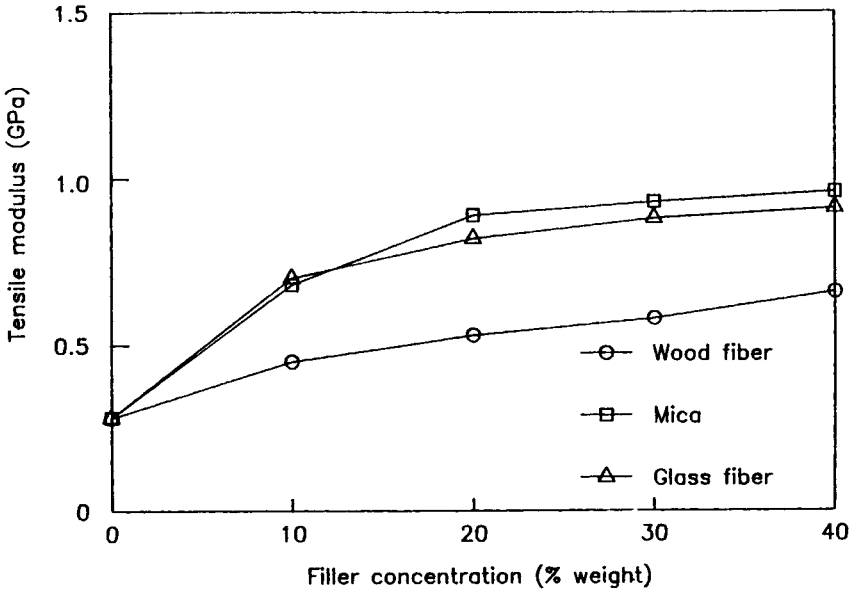

Fig. 12.

TABLE I
Effect of Weight Fraction of Fiber on Dimensional Stability
(after 4 h in Boiling Water)

Composite fiber weight (%)	Cross-sectional area increase (%)				
	0	10.0	20.0	30.0	40.0
LLDPE (unfilled)	0.78	—	—	—	—
LLDPE + wood fiber (3.0% PMPPIC treated)	—	1.23	1.71	2.92	3.87
LLDPE + mica	—	0.83	0.94	2.47	2.79
LLDPE + glass fiber	—	0.81	1.43	2.08	2.64

fillers absorb more water, one would expect a higher increase in the variation in cross-sectional area. But Kokta et al. observed that with a suitable fiber treatment the dimensional stability of the wood filled composites can be improved.[16]

CONCLUSIONS

Composites of LLDPE filled with glass fiber or wood fiber showed better retention of tensile strength and modulus after aging at 105°C for 7 days. Boiling water treatment resulted in a drastic drop in the tensile properties of wood fiber and mica composites. After immersion in boiling water for 4 h, the dimensional stability of the samples containing wood fiber was inferior to mica and glass fiber composites.

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Received April 10, 1989
Accepted July 13, 1989