Tensile Behavior of Unidirectional Polyethylene Fibers PMMA and Glass Fibers—PMMA Composite Laminates

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ABSTRACT: Unidirectional (UD) composite laminates based on glass fibers (GF) and high-performance polythylene fibers (PEF) were prepared with partially polymerized methyl methacrylate (MMA) at room temperature, followed by heating at 55°C (well below the softening point of PEF) for 2 h. The tensile strength, modulus of elasticity, fiber efficiency and strength efficiency of both the composite laminates, loaded parallel to the fibers, at the same volume fraction range, were investigated. All the properties were compared between the two composite laminates. It was observed that the measured tensile strength and modulus of elasticity deviated from the values calculated from the Rule of Mixture (ROM). The deviation was minimal at the lower volume fraction of fibers, and increased with the fiber volume. An interesting feature that was observed was that the efficiencies of PEF-reinforced composite was higher than that of the GF-reinforced composite at the same volume fraction of the fibers. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 76: 1489–1493, 2000

Key words: composite laminate; poly(methyl methacrylate) matrix; polyethylene fibers; glass fibers; tensile behavior

INTRODUCTION

The present trend of the polymer scientists is to prepare thermoplastics and thermosetting composites exhibiting high mechanical behavior, light weight, low cost, and covering different static and dynamic fields of application. By permutation and combination of various fibers and polymers a wide range of composites having unique properties for versatile applications, as an alternative to conventional materials like metals, wood, etc. have been prepared.

Gel-spun PEF of ultrahigh molecular weight of exceptionally good tensile (10 times that of steel), low density (0.97 g/c.c), and good abrasion, chem-

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ically inert with low dielectric value, high strength-to-weight, and high stiffness-to-weight ratios,¹ was used as one of the reinforcing fibers. Moreover, these PEF possess a relatively high energy to break compared with carbon, aramid, and GF.² Due to these unique properties, PEF have high potential for use in composite structures. A few workers have used PEF as a reinforcing fiber, but these works are mainly based on the thermoset matrix.²⁻⁶ Composites based upon thermoplastic polymeric matrices potentially offer several advantages compared to those based upon thermosetting resins.^{7,8} Thus, one could expect a composite structural material based on PEF-reinforced poly(methyl methacrylate) (PMMA), a thermoplastic polymer, as a matrix. GF, a well-known reinforcing fiber-reinforced PMMA, was also prepared at the same volume fraction of the fiber range.

The present work reports the tensile behavior of UD-PEF-reinforced PMMA laminates (poly-

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ethylene fiber-reinforced composite, PEFRC) and GF-reinforced PMMA laminates (glass fiber-reinforced composite, GFRC) at a different volume fraction of the fibers. The experimental tensile strength, modulus of elasticity, fiber efficiency, and strength efficiency have also been compared with the theoretical values, the latter being constructed using the ROM equation. All the above properties have been compared between PEFRC and GFRC at same volume fraction of the fiber range.

THEORETICAL ASPECT

The ultimate tensile strength of UD–fiber composites in the longitudinal direction has traditionally been predicted by ROM more or less successfully. The basic approach is to consider the mode of failure of the composite, evaluate each contribution of the fibers and the matrix at the point of failure, and calculate the ultimate strength of the composite as the sum of the contributions according to their relative volumetric propertions.^{9,10} The ROM stated in its most general form can be given as:

$$\sigma = \sigma_m V_m + \sigma_f V_f \tag{1}$$

where σ is the strength of the composite, σ_m the strength of the matrix, σ_f the strength of the fibers, and V_m and V_f the volumetric fractions of the matrix and the fibers, respectively. If the elongation of fibers are greater than that of the matrix, then the above equation after matrix cracking should be modified by neglecting the contribution of the matrix, and can be rewritten as:

$$\sigma = \sigma_f V_f \tag{2}$$

The ratio of the experimental tensile strength to the theoretical tensile strength is the strength efficiency of the composite.

According to the ROM, the modulus of elasticity of a well-aligned UD-fiber-reinforced composite, E, in the direction of the fiber alignment is given by¹¹:

$$E = BE_f V_f + E_m V_m \tag{3}$$

where B is the fiber efficiency factor for modulus, E_f the modulus of elasticity of the fiber, and E_m the modulus of elasticity of the matrix.

EXPERIMENTAL PROCEDURE

Fibers and other reagents used are as follows: PEF (Spectra 900, 1200 den) supplied by Allied-Signal Corporation, Petersburg, USA; GF (433 BF-225) supplied by Owens Corning Fiberglas Corporation, Ohio; MMA Supplied by Western Chemical Corporation, Calcutta, India; Benzoyl Peroxide (Bz_2O_2) supplied by Loba-Chemie Indoaustranal Corporation, Bombay, India; N,N dimethyl aniline (NDA) supplied by E. Merk Limited, Bombay, India.

MMA was purified by a standard technique^{12,13} and Bz_2O_2 was recrystallized from chloroform¹⁴ and dried in vacuum. The purification of NDA was achieved by distillation under reduced pressure before use.

The PEF used for the preparation of composites were treated with chromic acid following noted procedures.^{2,15,16} The surface of GF were already treated with standard treatment, used directly for making composites. The wetting characteristics of PMMA on treated and untreated GF and PEF have been studied by contact angle determination as noted previously.^{17–19} Improved wetting was found when the treated fibers were investigated.²⁰

The UD-plies were made in a dust-free chamber on a glass sheet using partially polymerized MMA as the resin with an amineperoxide (NDA- Bz_2O_2) initiator system in bulk at room temperature.²¹ Laminated structures were prepared by stacking these plies of PEF and GF unidirectionally in the mold, and the composites were made by using same resin at room temperature until it solidified within the mold, and shrinkage was controlled using extra resin in the mold. Finally, the composite was heated to a temperature of 55°C for 2 h to ensure the completion of MMA polymerisation. UD-laminates were prepared up to four plies for PEF (designated as S_1 to S_4 , respectively) and GF (designated as G_1 to G_4 , respectively). A detailed description of the preparation of laminates is given elsewhere.^{20,22–24}

Tensile testing of the samples were carried out at 25 ± 0.5 °C, using a dumbbell-shaped test specimen in an Instron Universal Testing machine. The specification of the dumbbell is as follows: gauge length 20 mm, width 6 mm, and thickness 1.70 mm with end tabs, and were loaded with serrated jaw wedge grips. A strain rate of 5 mm/ min was used throughout the investigation. In all cases, 12 specimens were tested, and average values are reported.



Figure 1 Stress-strain curves for GFRC (G_1 to G_4 are one ply to four-ply laminates, respectively).

RESULTS AND DISCUSSION

Figures 1 and 2 show the stress-strain behavior of the GFRC and PEFRC, respectively, and the PMMA matrix. The failure strain of GF and PEF is about 2 and 2.5 times to that of matrix (ε_m) respectively. The stress-strain curve display a point of inflection (Knee point) around ε_m , which enables the curve to be approximated by two regions—one indicating the elastic (below the point corresponding to ε_m), other the plastic region (beyond the point corresponding to ε_m). It is established that the elastic region of the composite is dependent on ε_m . The increase in fiber content leads to an increase in the first crack stress (corresponding to ε_m), tensile strength, and ultimate strain. It is observed from the stress-strain curves that the main influence of the fibers is in



Figure 2 Stress-strain curves for PEFRC (S_1 to S_4 are one- to four-ply laminates, respectively).



Figure 3 Comparison of stress–strain curves. (—) GFRC; (- - -) PEFRC.

the postcracking zone, where the contribution of the matrix is small or even negligible, because of its multiple cracking, where is the ultimate strength of the laminates is determined by the fibers.

PEFRCs have a quite different load transformation characteristics from one region to the other than that of GFRC. Figure 3 shows the comparative curves of both the composite laminates (PEFRC and GFRC) in that region. From the figure its is clear that there is a certain drop in modulus around the point A of the GFRC. But in case of PEFRC, a smooth and continuous drop in modulus has been found. This is due to the fact that at this region there is no appreciable change in modulus of GF (E remains nearly 70 GPa), but in the case of PEF, the modulus decreases steadily (E at 1.5 and 4.5% strain in 72 GPa and 45.5 GPa, respectively).

Figures 4 and 5 show the variations of tensile strength and modulus of elasticity (*E*) with V_f for both GFRC and PEFRC. Curves in these figures include theoretical and experimental. Theoretical values of *E* are obtained by dividing σ [calculated from eq. (1)] by corresponding strain [at 0.5% strain, σ_m is 14.5 MPa, σ_f is 350 MPa (PEF)]. Tensile strength and *E* appear to be linearly dependent on V_f . Tensile strength of the GFRC is higher than that of PEFRC at same V_f (Fig. 4). But in the case of *E*, PEFRC shows higher values



Figure 4 Tensile strength dependence of the volume (%) of fibers. (—) GFRC; (- -) PEFRC; (\bullet) theoretical points; (\bigcirc) experimental points.

than that of GFRC (Fig. 5). This behavior is due to the fact that the tensile strength of GF is higher compared with PEF, whereas E of the PEF is higher than that of GF.

The interesting feature of these studies is that the distance between corresponding theoretical and experimental curves increases with the increase of V_f , and the deviation is minimum at lower range of V_f . This is a case contrary to the observation of Mittelman and Roman²⁵ on UD– Kevlar–epoxy composites of the similar V_f range. Mittelman et al. have used the fiber-winding and impregnation technique at a V_f range of 0.26– 0.73, and this has been analyzed by considering nonhomogeneous fiber distribution at a lower V_f



Figure 5 Modulus of elasticity dependence of the volume (%) of fibers. (—) GFRC; (- -) PEFRC; (\bullet) theoretical points; (\bigcirc) experimental points.



Figure 6 Strength efficiency dependence of the volume (%) of fibers. (—) GFRC; (- - -) PEFRC.

range. In the present work, the hand lay-up technique has been used with a V_f range of 0.089-0.357. At a higher V_f , fiber interaction takes place, and either they tend to bundle up among themselves or touch each other physically, which is due to the fact that the hand lay-up technique produces more or less a random nature of fiber distribution in the matrix. Due to the above facts, proper and uniform penetration of the matrix does not takes place throughout the fiber surfaces, leaving interstitial voids. The fiber surfaces in contact with the voids are ineffective.²⁶ It is found that void content (% by volume) increases from single-ply to multiple-ply laminates (0.60 to 1.05% for GFRC and 0.30 to 0.85% for PEFRC, respectively, for single- to four-ply laminates in the present work). Thus, the degree of fiber misalignment and void content increases with the increase in V_{f} . These facts are reflected in the experiment by the deviation of the experimental curve from the theoretical one (Figs. 4 and 5).

In Figures 6 and 7 the strength efficiency



Figure 7 Fiber efficiency dependence of the volume (%) of fibers. (—) GFRC; (- - -) PEFRC.

 $(\sigma_{\text{EXPT}}/\sigma_{\text{ROM}})$ and Fiber efficiency [calculated from eq. (3), where E_f is 70 GPa (GF) and 103 GPa (PEF) and E_m is 2.9 GPa] is plotted against respective V_f . Both the efficiencies are found to decrease with the increase in V_f . One of the most interesting feature of the studies is that the efficiencies of PEFRC is always higher compared to GFRC. This may be due to the fact that brittle nature of GF that is sensitivity to abrasion with handling makes the composite weaker due to the breakage of fiber during the manufacturing process.

CONCLUSIONS

From the above studies the following conclusion maybe drawn:

- 1. The knee of the composite stress-strain curve is associated with fracture of the matrix.
- 2. Comparing same V_f , the tensile strength is higher in the case of GFRC than that of PEFRC, but when *E* is concerned, PEFRC shows a higher value compared to GFRC.
- 3. The experimental tensile strength and E for a given composite deviates from the theoretical values over the range of V_f used. Deviation becomes higher with the increase in V_f .
- 4. Fiber and strength efficiencies of the PEFRC are higher than that of GFRC at the same V_{f} .

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