

Tensile Behavior of Glass Fiber-Reinforced Acetal Polymer

G. F. HARDY and H. L. WAGNER,*
Celanese Research Company, Summit, New Jersey 07901

Synopsis

The tensile stress-strain behavior of glass fiber-reinforced polyacetal resin was investigated for various fiber concentrations, fiber length distributions, and finishing agents. The polyacetal fiber blends change considerably in strength and elongation at break when treated with ammonium chloride, but otherwise similar specimens still follow a common stress-strain curve to a point shortly before failure. As the mean fiber length decreases, the modulus and tensile strength fall, but the elongation at break remains almost unchanged. The observed tensile behavior is discussed in terms of a simplified model, which assigns the fibers to two categories: a fraction α parallel to the applied load, and the remainder distributed in a plane perpendicular to the load axis. By fitting this model to the stress-strain curves, two other constants of each system are derived: a length-dependent efficiency factor β for parallel fibers, whose magnitude agrees with the predictions of Rosen and his co-workers, and a factor γ which expresses the constraint of the matrix resin by the "transverse" fibers. The behavior of γ is consistent with Tsai's theory of the transverse modulus of laminates, if a reasonable amount of fiber-fiber contact is assumed. In terms of this model, possible interpretations of the behavior under repeated loading and the mechanism of tensile failure are presented.

INTRODUCTION

Reinforcement of polymers by strong, rigid fibers can lead to substantial increases in tensile strength, modulus, and resistance to creep at elevated temperatures.¹ For laminates based on thermosetting resins, in which the fiber geometry is usually simple and well controlled, the theory of these effects is quite highly developed.^{2,3} On the other hand, the properties of fiber-reinforced thermoplastics are not yet amenable to detailed interpretation, in spite of rapidly increasing interest in these materials.^{4,5} This is in part because strong fiber-matrix adhesion is difficult to achieve for some thermoplastics,^{6,7} but there are other difficulties. The usual methods of melt fabrication result in products which contain rather short fibers, more or less randomly dispersed in a resin matrix which typically has rather complex viscoelastic properties. These characteristics cannot be directly handled by the customary linear elastic theories of unidirectional composites.

* Present address: National Bureau of Standards, Washington, D. C. 20234.

In this communication, we will describe some features of the observed stress-strain behavior of acetal polymers containing glass fibers. A recently discovered chemical treatment^{8,9} has made it possible to control the tensile strength and elongation to break of polyacetal-glass fiber blends, without grossly changing the fiber geometry, fiber finish, or the mechanical characteristics of the resin matrix. Using a simple mechanical model, we will attempt to interpret and predict the tensile properties in terms of concepts already established in the theory of unidirectional composites.

EXPERIMENTAL

Materials

The resin matrix used in preparing most of the test specimens was an acetal copolymer containing less than 5 wt-% of oxyethylene units. It was stabilized by depolymerization of unstable chain ends and contained approximately 0.5 wt-% of a conventional antioxidant-acid acceptor stabilizer system.¹⁰ The melt flow rate of this polymer at 190°C (ASTM D1238, condition E) was 9.0 g/10 min; its inherent viscosity was 1.25, measured in 98% *p*-chlorophenol-2% α -pinene at 60°C and a concentration of 0.1 g/100 ml.

Two representative types of commercial E-glass fiber were used, each in the form of 1/4-in. chopped strand. According to information supplied by the manufacturer, one of these contained a poly(vinyl acetate) binder and vinylsilane coupling agent; the other was finished with partially cured epoxy resin and a glycidoxysilane coupling agent.

For comparative purposes, a commercial glass fiber-reinforced nylon 66 was also examined. This product contained 33 wt-% randomly dispersed glass fiber, of about 0.4 mm average length. The composition of the fiber finish is not known. Unfilled nylon resin from the same source was also tested, to determine the mechanical properties of the matrix.

Fabrication

Uniformly dispersed blends of glass fiber and acetal copolymer were prepared by two methods. (1) The polymer was melted on a two-roll mill at about 190°C. Chopped glass fiber was added in small portions over a 5-min period, and the blend was homogenized for another 5 min. Final average fiber lengths ranged from about 0.2 to 0.7 mm, depending on the amount of glass fiber used. (2) A mixture of chopped glass fiber and resin was blended in a Banbury mixer for 15 min at about 180°C and 230 rpm. The final average fiber length was about 0.05 mm.

In either case, the hot product was pressed flat and cut into 1/4-in. chips for injection molding.

The test moldings were ASTM Type I tensile bars, 1/8 in. thick, with moderately open gating located at one end. The fiber orientation was kept as constant as possible by carefully standardizing molding conditions. A

ram injection molding machine was used at a pressure of 25000 psig, with a 60-sec cycle and a 25-sec dwell time. For acetal-based materials, the nozzle and cylinder were set at 205–225°C and the die temperature was 110–120°C. When molding the nylon resins, the nozzle and cylinder were set at 275°C and 290–315°C, respectively.

Test Methods

The tensile tests were carried out according to ASTM D638 at a crosshead speed of 0.2 in./min. Reported elongation was based on crosshead motion and a nominal gage length of 4.5 in. Matrix resins were also tested at higher speeds, up to 0.5 in./min. The polyacetal specimens were conditioned for 48 hr at 23°C and 50% RH; the nylon specimens were tested dry, as molded, after aging at least 48 hr in a closed container. Reported tensile data are averages of at least five independent determinations. Several materials were also tested after first loading at 0.2 in./min to produce a predetermined strain, then unloading at 0.2 in./min and allowing to relax for at least ten times the loading period.

Approximate average fiber lengths in the test specimens were determined by microscopic examination at 50–100 \times . A small chip cut from the molded specimen was first treated with nitric acid (polyacetal) or formic acid (nylon) to remove the matrix resin. The resulting glass fiber mat could then be dispersed for study by gentle agitation.

Ammonium Chloride Treatment

The reinforcing efficiency of glass fiber in polyacetal resin was varied by adding traces of ammonium chloride, as described in recent patents.⁸ Because of the small amount required, it was most convenient to do this by dry blending the ammonium chloride (0.01–0.02 wt-%) with the powdered polymer before adding the glass fiber. At this concentration, in the absence of glass fiber, ammonium chloride was found to have no detectable effect on the mechanical properties of properly fabricated polyacetal.

Tensile Properties

Using epoxy resin-finished glass fiber, the effect of ammonium chloride treatment was examined at four levels of fiber content, ranging from 10 to 40 wt-%. The results of these experiments are summarized in Table I and Figure 1 (solid curves). The most striking feature to be observed is that all the stress-strain curves for a given glass content seem to lie largely on a common envelope, irrespective of the efficiency of reinforcement, as judged by breaking strength. In keeping with this, the initial tensile moduli at a given glass fiber content were almost independent of strength, within the accuracy with which the initial tangents to these curves could be drawn. Only at fairly high elongations, close to the final break, do the observed stresses fall below the common envelope. The increased strength produced by ammonium chloride treatment is, thus, associated with increased elongation to break, for a given glass fiber content.

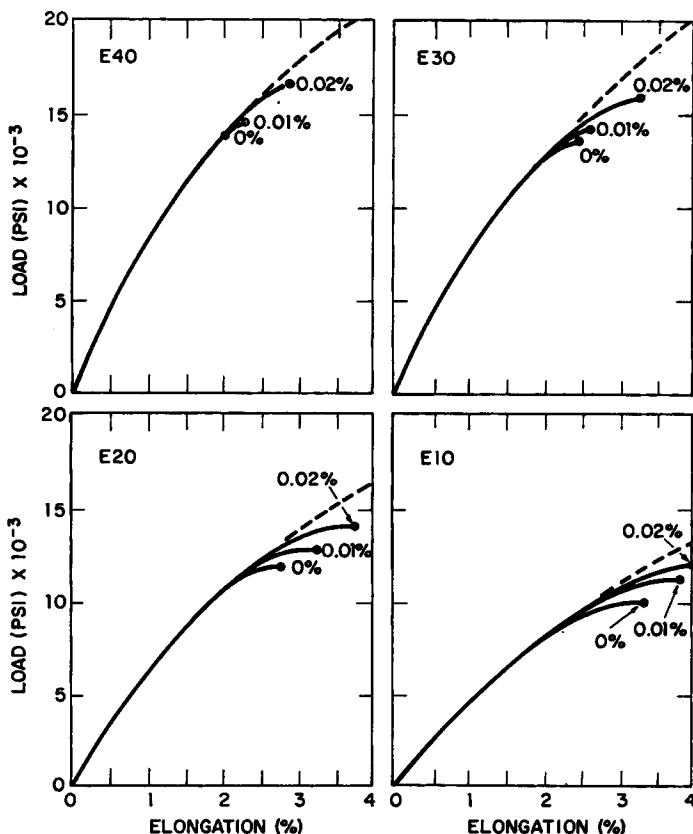


Fig. 1. Observed stress-strain curves and fitted eq. (1) for polyacetal containing 10–40 wt-% glass fiber finished with epoxy resin. The wt-% NH_4Cl added is indicated for each experimental curve.

The effects of increased glass fiber content, which may be seen by comparing the various envelopes, are about as would be expected.^{2,3} The initial modulus rises, as does the level of tensile strength which can be achieved. For a given ammonium chloride concentration, the elongation to break falls fairly rapidly as the fiber content increases.

A second experiment was performed with the use of glass fiber finished with poly(vinyl acetate). Some of the results are shown in Figure 2. All of these specimens contained 40 wt-% glass fiber, but the distribution of fiber lengths was varied by roll mill blending of various proportions of material originally prepared by the roll mill and Banbury methods. This was done at two levels of ammonium chloride concentration.

It can be seen, first of all, that the stress-strain curve for the material wholly prepared on the roll mill is very similar to that observed for 40% epoxy finished glass fiber (compare with Fig. 1). Numerical details are given in Table I. However, the most striking feature of these data is the observed fact that the replacement of fairly long fibers by very short fibers

TABLE I
Experimental Materials and Constants of Fitted Stress-Strain Curves

| Series | Glass content, wt-% | | Mean fiber length, L/d | $\alpha = 0.4$ | |
|--------------------|------------------------|----------------|-----------------------------|---------------------|---------------------|
| | | | | β | γ |
| N ^a | 33 | | 40 | 0.54 | 1.60 |
| E10 ^b | 10 | | 60 | 0.51 | 1.15 |
| E20 | 20 | | 35 | 0.42 | 1.35 |
| E30 | 30 | | 30 | 0.38 | 1.65 |
| E40 | 40 | | 20 | 0.29 | 2.05 |
| V40/0 ^c | 40 ^d | 0 ^e | 25 | 0.31 | 2.05 |
| V30/10 | 30 | 10 | 20 ^f | 0.28 | 2.05 |
| V20/20 | 20 | 20 | 15 ^f | 0.23 | 2.05 |
| V10/30 | 10 | 30 | 10 ^f | 0.15 | 2.05 |
| V0/40 | 0 | 40 | 5 | (0.05) ^g | (2.05) ^g |

^a Commercial glass fiber-nylon blend.

^b Epoxy-coated glass fiber in polyacetal.

^c Vinyl acetate-coated glass fiber in polyacetal.

^d Fiber derived from roll mill blend.

^e Fiber derived from Banbury blend.

^f Calculated from values for V40/0 and V0/40.

^g Extrapolated.

has very little effect on the elongation to break, while it drastically reduces the attainable tensile strength. The entire stress-strain envelope is lowered in proportion to the amount of short fiber material used; thus, in this case, the initial modulus is reduced as well. Here again, the effect of increased ammonium chloride concentration is simply to increase the stress and strain at which the curve deviates from a common envelope. The approximate constancy of elongation to break over the entire range of blends holds for either level of ammonium chloride treatment.

The behavior of two of these same materials (100% roll mill fiber, at two levels of ammonium chloride treatment) was also investigated after the specimens had been subjected to varying amounts of prestrain and then unloaded. This experiment was undertaken to examine the possibility that the resin may pull away progressively from the fiber ends as the loading increases.¹¹ The initial tensile modulus found in each trial is plotted in Figure 3 as a function of the prestrain. Fairly substantial changes in modulus were found, but it will be shown below that they were most likely to be due to differences in the properties of the resin matrix, caused by the strain history.

The stress-strain behavior of the glass-reinforced nylon composition, which was tested for comparison with the polyacetals, is shown in Figure 4.

DISCUSSION

Model for Tensile Behavior

We will attempt to rationalize the shapes of the stress-strain envelopes in Figures 1, 2, and 4, assuming perfect adhesion between fibers and matrix

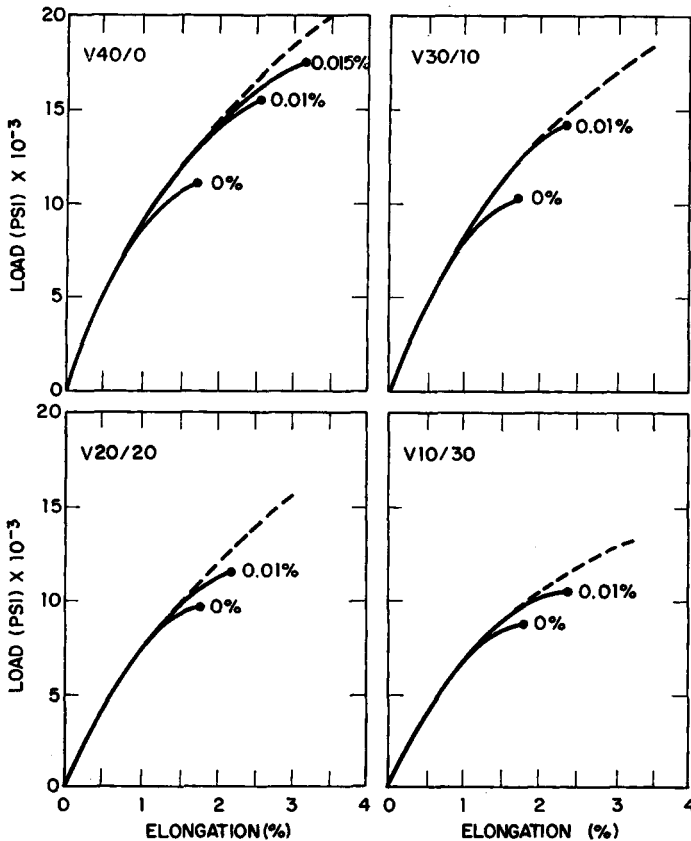


Fig. 2. Observed stress-strain curves and fitted eq. (1) for polyacetal containing 40 wt-% glass fiber finished with poly(vinyl acetate). Various proportions of longer and shorter fibers were used, at the indicated NH_4Cl contents.

resin. The negative deviations which precede failure will thus be ascribed to breakdown of the adhesive bond, or perhaps to some other form of structural deterioration such as fiber fracture.

The mechanical behavior will be approximated by that of a group of fibers parallel to the applied load, surrounded by a matrix consisting of the polymer plus other fibers randomly arranged in a plane perpendicular to the loading direction. For brevity, these will be referred to as the "parallel" and "transverse" fibers, respectively. The sum of the volume fractions of these two hypothetical groups of fibers will be taken to be equal to the true volume fraction of added fibers (V_f).

The fraction of the fiber content assumed parallel to the applied load will be denoted by α . We have not carried out a detailed investigation of the true orientation distribution of the fibers (a matter of some difficulty), and will not attempt to predict the values of the components to be assigned to a fiber of given orientation. Instead, α will be regarded as a measure of the effective orientation distribution, to be derived from the tensile data.

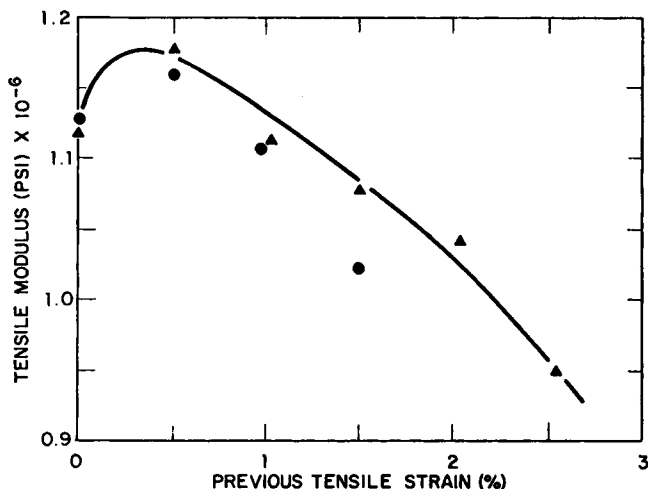


Fig. 3. Observed values of initial tensile modulus of polyacetal containing 40 wt-% glass fiber after the indicated amount of previous strain and relaxation: (●) no NH_4Cl ; (▲) 0.015 wt-% NH_4Cl . Solid curve predicted by eq. (1), using constant α , β , γ , and resin properties obtained after a suitable previous strain.

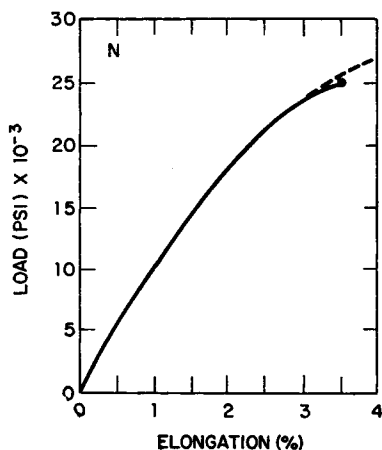


Fig. 4. Observed stress-strain curve and fitted eq. (1) for nylon containing 33 wt-% glass fiber.

Following the usual "law of mixtures,"² we can then divide up the total stress σ on an element of the composite into contributions from the "parallel" fibers (assumed linearly elastic) and the matrix:

$$\sigma = \alpha\beta V_f E_f \epsilon + (1 - \alpha V_f) \sigma_{m, \epsilon}$$

In this equation, $\sigma_{m, \epsilon}$ is the stress in the matrix (consisting of resin and "transverse" fibers), E_f is the Young's modulus of the fibers, and β is a mechanical efficiency factor for the "parallel" fibers, due to their finite

length (see below). For fibers which are very long, β becomes equal to unity.

It is now necessary to evaluate $\sigma_{m,\epsilon}$ for the composite strain ϵ . Here we encounter two difficulties: the matrix is itself a composite material, and the polymer component is highly nonlinear in its elastic behavior, even before the onset of plastic flow. We make the following assumption: the effect of the "transverse" fibers is simply to make the true elongation of the resin component equal to the overall strain ϵ , multiplied by some constant factor γ . Thus, $\sigma_{m,\epsilon}$ at the composite elongation ϵ is taken to be equal to the resin tensile stress $\sigma_{r,\gamma\epsilon}$ as measured at an elongation of $\gamma\epsilon$.

There is considerable precedent for an assumption of this sort. Nielsen^{12,13} has proposed that the stress-strain behavior of a polymer containing rigid spherical particles can be represented by

$$\epsilon(\text{resin})/\epsilon(\text{composite}) = \gamma = (1 - V_f^{1/3})^{-1}$$

where γ is again the ratio of true strain in the resin to the observed strain for the filled specimen. Kies² and Schulz¹⁴ have investigated the strain concentration produced in a matrix containing parallel, uniformly spaced fibers, when loaded in a direction perpendicular to the fiber axis; they find that the matrix between fairly closely spaced fibers may be forced to sustain an elongation as much as 5-20 times that of the composite as a whole. The factor γ is thus to be regarded as an average strain concentration factor for resin containing the transverse fibers.

The stress-strain relation for the total fiber reinforced material will then be approximated by the sum of (1) a linear term due to the parallel fibers and (2) a nonlinear term derived from the stress-strain relation of the resin itself by compressing the strain axis:

$$\sigma = \alpha\beta V_f E_f \epsilon + (1 - \alpha V_f) \sigma_{r,\gamma\epsilon} \quad (1)$$

With three disposable constants, it is evident that at least a fair fit to the observed stress-strain envelopes will always be possible. The justification for this approach must then rest on the plausibility of the values found for α , β , and γ . However, it should be noted that there are certain restraints, imposed by the physical meaning assigned to each constant. Thus, it seems reasonable to require that a single value of α be assigned to all the data presented herein, since the molding conditions were kept constant, so that the distribution of fiber orientations probably did not vary greatly. For a given set of fiber and resin properties, β should vary only with fiber length and concentration; γ should be ≥ 1 , and be controlled by the total fiber content and the value of α .

Application to Experimental Results

The stress-strain envelopes shown in Figures 1, 2, and 4 (dotted lines) were obtained by fitting eq. (1) to the initial portions of the observed tensile loading curves. For any given value of α , γ was chosen to make the differ-

ence $\sigma = (1 - \alpha V_f) \sigma_{r,\gamma\epsilon}$ linear in ϵ up to the greatest possible elongation. The values of $\sigma_{r,\gamma\epsilon}$ were read off the measured stress-strain curve for pure resin at an ordinate $\gamma\epsilon$, as explained above. Since the factor γ implies that the actual strain rate in the resin was 0.2γ in./min, it was necessary to interpolate between values obtained at several strain rates, but this correction was appreciable only for the highest fiber contents.

Equation (1) could be fitted very closely to all the observed stress-strain curves. Up to a point just before the specimens broke, the fitted curve was always considerably closer to the averaged data than were most of the individual curves on which the averages were based. The values obtained for the constants α , β , and γ are listed in Table I. At this point, there is little limitation on the value of α , since it appears only in the product $\alpha\beta$ and in the factor $1 - \alpha V_f$ which is always close to unity. The value chosen for α will be justified in the next section.

Fibers Parallel to the Applied Load

The values obtained for α and β , reflecting the behavior of that fraction of the fibers assumed parallel to the applied load, may be compared with the results of a theoretical analysis described by Rosen.^{15,16} It is assumed that the "parallel" fibers take up a share of the load applied to the composite, by stress transfer from the surrounding matrix. Near the middle of a long fiber, the simple "law of mixtures" condition applies, the fiber and matrix elongate equally, and each bears a fraction of the load determined by its modulus and cross-sectional area. The tensile stress in a fiber is taken to be zero at an end; it builds up toward the central portion by shear between the fiber and the matrix. The following relations are found¹⁶ for the fiber tensile stress (σ_f) and interfacial shear stress (τ) at a distance x from the end of a fiber of length L and diameter d :

$$\sigma_f/\epsilon E_f = 1 - \cosh \eta x + \tanh \eta L/2 \cdot \sinh \eta x \quad (2)$$

$$\tau/\epsilon E_f = \eta d/4 (\tanh \eta L/2 \cdot \cosh \eta x - \sinh \eta x) \quad (3)$$

$$\eta = (G_b/E_f)^{1/2} [\alpha^{1/2} V_f^{1/2} / (1 - \alpha^{1/2} V_f^{1/2})]^{1/2} (2/d) \quad (4)$$

In these equations, G_b is the shear modulus of the matrix, which is assumed to be a linear elastic material. We can integrate eq. (2) over the length of a fiber, and obtain an expression for the average fiber stress, which is the same as the product $\beta E_f \epsilon$ in eq. (1). Thus,

$$\beta = 1 - (\tanh N/N) \quad (5)$$

$$N = (G_b/E_f)^{1/2} [\alpha^{1/2} V_f^{1/2} / (1 - \alpha^{1/2} V_f^{1/2})]^{1/2} (L/d) \quad (6)$$

To use these relations, it is necessary to obtain an approximate value of G_b for the elastically nonlinear environment of the parallel fibers. Due to the strain concentration effect, the matrix modulus must be γ times the modulus of the resin. The latter quantity was approximated by the secant modulus of the polymer over the range of interest; that is, up to an

elongation of γ times the observed elongation to break of the strongest specimens. (This turned out to be close to the yield maximum of unfilled polymer.) To convert tensile to shear modulus, we have assumed that Poisson's ratio ν is 0.4 for both polyacetal and nylon and used the relation $G = E/2(1 + \nu)$. The values of resin tensile modulus actually used in the computation were 1.5×10^5 psi for the polyacetal and 1.9×10^5 psi for nylon. The tensile modulus and Poisson's ratio of the glass fiber were taken to be 10.6×10^6 psi and 0.22, respectively.¹⁷

The curve in Figure 5 shows the relation between β and N which is predicted by eq. (5). The value of α used in Table I was chosen to give the best fit of all the experimental β to this theoretical curve. The points in Figure 5 are these experimental values of β , plotted against values of N , computed from known properties of each material by using eq. (6). Considering the many approximations made, the agreement which can be obtained is surprisingly good.

An α equal to 0.4 might seem to imply only a moderate bias of the fiber orientation toward the loading direction, but there is theoretical evidence that the fiber efficiency falls off very rapidly as the angle between the fiber axis and the loading direction increases from zero.^{2,3} This will tend to reduce the values of α computed by the present method, as compared to the true orientation distribution. A qualitative microscopic examination of typical sections actually revealed a rather strong tendency for fibers to be aligned at small angles to the loading direction.

Equations (2) and (3) can be used to estimate two other important properties of each system, the maximum interfacial shear stress (at the fiber ends) and the maximum fiber tensile stress (at the middle). The results of this computation are summarized in Table II, using, in each case,

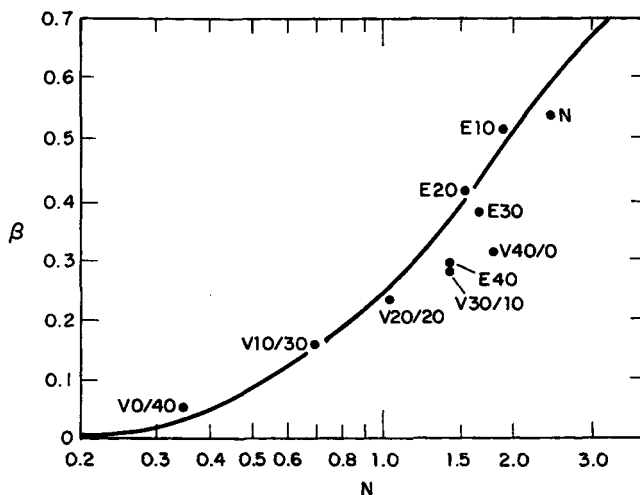


Fig. 5. Fiber efficiency β plotted against the structural constant N defined by eq. (6). The solid curve is the relation predicted by eq. (5).

TABLE II
Predicted Stress-Strain Behavior of Parallel Fibers

| Series ^a | N^b | β^c | Maximum elongation at break, % | τ_{\max} , psi ^d | $\sigma_{\max} \times 10^{-3}$, psi ^e |
|---------------------|-------|-----------|--------------------------------|----------------------------------|---|
| N | 2.46 | 0.60 | 3.5 | 11,200 | 308 |
| E10 | 1.91 | 0.50 | 4.0 | 6,500 | 301 |
| E20 | 1.52 | 0.40 | 3.7 | 7,700 | 228 |
| E30 | 1.68 | 0.44 | 3.3 | 9,100 | 224 |
| E40 | 1.41 | 0.37 | 2.8 | 9,300 | 160 |
| V40/0 | 1.77 | 0.47 | 3.1 | 11,000 | 220 |
| V30/10 | 1.41 | 0.37 | 2.3 | 7,600 | 131 |
| V20/20 | 1.06 | 0.26 | 2.2 | 6,500 | 89 |
| V10/30 | 0.71 | 0.14 | 2.4 | 5,500 | 53 |
| V0/40 | 0.35 | 0.04 | (2.4) ^f | 3,000 | 15 |

^a See Table I.

^b From eq. (6).

^c Predicted by eq. (5).

^d Maximum shear stress at average fiber end, from eq. (3).

^e Maximum tensile stress at middle of average fiber, from eq. (2).

^f Extrapolated.

the maximum elongation actually observed. The values obtained for the maximum shear stress are unlikely to be very accurate, since the geometry of the fiber ends would have a great effect on the stress distribution.¹⁸ They do, however, suggest that localized plastic flow would be appreciable in certain cases in which the computed shear stress is very high. As pointed out by Rosen,¹⁶ plastic regions (near the ends) would tend to reduce the effective fiber length compared to the fully elastic case. It thus seems significant that the computed interfacial shear is greatest for precisely those materials whose values of β fall below the predicted fiber efficiency curve in Figure 5. On the other hand, a model in which the major part of the resin-fiber interface was undergoing plastic deformation would clearly be inappropriate in most cases.

The estimated maximum fiber tensile stresses indicate that complete stress transfer to the fibers was not attained in any system, due to insufficient fiber length (σ_{\max} is always less than $E_f \epsilon$). The maximum tensile stresses calculated are quite reasonable, in the light of reported strengths of short E-glass fibers.^{2,16,17}

Fibers Transverse to Load

The constant γ reflects the extent to which the mechanical properties of the resin phase are modified by the presence of the transverse fibers. The data in Table I indicate that γ rises significantly, but fairly slowly, with increasing fiber content; it does not seem to be greatly affected by fiber length, fiber finish, or the nature of the matrix resin.

The behavior of γ can be related to that of the transverse modulus of a

unidirectional composite. Several theoretical treatments of this problem are available;¹⁹ that of Tsai²⁰ seems most appropriate for our present purposes. His formulation includes a contiguity factor C , to allow for the effects of fiber-fiber contacts in a randomly packed unidirectional composite; such contacts might be expected to be quite frequent in the multidirectional systems under study. When C is zero, all the fibers are isolated by resin; when C is unity, all the fibers are in contact—that is, the composite is regarded as a glass sponge whose interstices are filled with resin.

The relation derived by Tsai is:

$$E_{\text{trans}} = \gamma E_r = 2[1 - \nu_t + (\nu_t - \nu_r)V_r] \times \left[(1 - C) \frac{K_t(2K_r + G_r) - G_r(K_t - K_r)V_r}{(2K_r + G_r) + 2(K_t - K_r)V_r} + C \frac{K_t(2K_r + G_t) + G_t(K_r - K_t)V_r}{(2K_r + G_t) - 2(K_r - K_t)V_r} \right] \quad (7)$$

where V_r is the volume fraction of resin and ν is Poisson's ratio. The volume of "parallel" fibers must clearly be neglected in computing V_r , since we are attempting to estimate the properties of a mixture of resin and transverse fibers. K is $E/2(1 - \nu)$ and G is $E/2(1 + \nu)$. The properties of glass fiber, polyacetal and nylon to be used in the computation were discussed in the preceding section. Table III gives the values of C corresponding to γ for each system studied, as calculated by eq. (7). It can be concluded that the behavior of γ is in rather good accord with the theory of Tsai, taking C as 0.25 to 0.3, with a slight upward trend as the fiber content increases. The only data available for comparison are for filament wound unidirectional specimens of much higher fiber content (50–70 vol-%); values of C ranging up to 0.5 have been reported for such systems.^{20,21}

TABLE III
Stress-Strain Behavior of Resin and Transverse Fibers

| Series ^a | C ^b | Computed strain in resin at failure, % | | | |
|---------------------|----------------|--|-----------------------------|------------------------------|-----------------------------|
| | | 0% NH ₄ Cl | 0.01% NH ₄ Cl | 0.015% NH ₄ Cl | 0.02% NH ₄ Cl |
| E10 | 0.26 | 3.8 | 4.4 | | 4.6 |
| E20 | 0.25 | 3.6 | 4.3 | | 5.0 |
| E30 | 0.27 | 4.0 | 4.3 | | 5.4 |
| E40 | 0.28 | 4.1 | 4.5 | | 5.7 |
| V40/0 | 0.28 | 3.3 | 5.1 | 6.4 | |
| V30/10 | 0.28 | 3.3 | 4.7 | | |
| V20/20 | 0.28 | 3.7 | 4.5 | | |
| V10/30 | 0.28 | 3.7 | 4.9 | | |
| N | 0.31 | 5.6 | | | |

^a See Table I.

^b From eq. (7).

Mechanisms of Failure

The model used has been based on the assumption that resin-fiber adhesion is complete up to the point at which the true stress-strain curve begins to fall below the computed envelope. If this is so, the tensile behavior should not be greatly affected by a previous loading less than that necessary to cause deviation from the envelope.

Two sets of specimens were tested after varying amounts of previous loading, to examine this feature of the model. It was found at once that the behavior of the resin itself was not completely reversible under these conditions. It was thus necessary to compare the results obtained after a prestrain ϵ with that computed by eq. (1) from the properties of resin which had undergone a comparable prestrain $\gamma\epsilon$. This was carried through only for the initial tensile modulus, as shown in Figure 3. The predicted behavior (using the original α , β , and γ) is shown as a solid curve; the points are data obtained after the indicated prestrain. For the composition containing NH_4Cl , which broke at about the deviation strain of 2.5%, the agreement was good throughout. For the other, which contained no NH_4Cl , the experimental modulus fell below the computed value after prestraining to about the deviation point (1% strain). There is thus no indication that the structural constants α , β , and γ are affected by a strain history which is insufficient to cause the original stress-strain curve to depart from the envelope. On closer examination, however, one might expect to find changes due to fiber buckling² and localized plastic flow in the resin.

There are several possible modes of structural failure at the point of deviation from the envelope. (1) The parallel fibers may break near the middle, where the tensile stress is highest. This is unlikely to be true in all cases, as the computed stresses in Table II show, and cannot account for the effects of varying ammonium chloride treatment. (2) Resin may peel back from the ends of "parallel" fibers.^{11,22} Again, one would have to explain why this occurred at different interfacial stresses in different systems, as listed in Table II. (3) Resin may pull loose from the transverse fibers. Table III shows that the computed resin strain at failure is a fairly constant function of ammonium chloride content, independent of the amount of fiber, for the epoxy finished series *E*. This tends to support such a failure mode, as does the relatively small influence of fiber length on the resin strain at failure in the *V* series. A similar type of failure initiation by separation at transverse fibers has been observed in thermoset laminates containing alternate layers of unidirectional fibers arranged parallel and transverse to the applied load.²³

The values of computed resin strain in Table III also indicate that the poly(vinyl acetate) finish does not adhere to the resin as well as the epoxy finish, unless ammonium chloride has been added. However, with ammonium chloride treatment, the interphase adhesion seems to improve to a greater extent for the *V* series than for the *E* series. The maximum resin

strain computed for the glass-reinforced nylon specimen is similar to that found with the stronger polyacetal compositions.

CONCLUSIONS

Tensile test data, obtained on specimens of glass fiber-reinforced polyacetal and nylon resins, over a fairly wide range of composition, seem to be consistent with a relatively simple mechanical model. By replacing the true fiber orientation distribution with two hypothetical groups of fibers arranged parallel and transverse to the applied load, the tensile stress-strain curve can be described in terms of three characteristic constants.

The effective fraction of fibers parallel to the load (α) should, in principle, be derivable from a study of the orientation distribution plus a theory of the action of oblique fibers. In practice, it seems more convenient to obtain α empirically, since its value apparently remains constant for a given set of molding conditions.

The effect of those fibers assumed perpendicular to the load axis can be expressed by an increase in the effective strain in the resin by a factor γ . The variation in γ with glass fiber content agrees with expected values for the transverse modulus ratio of a unidirectional composite, if a moderate degree of fiber contiguity is assumed.

The load bearing ability of those fibers assumed parallel to the load is reduced by a factor β , due to their finite length. The magnitude of this factor can be computed from known properties of the system, by the method of Rosen.

For a given degree of resin-fiber adhesion, there are indications that systems of varying fiber content fracture at almost the same value of true resin strain, perhaps by pulling away of the resin phase from transverse fibers.

These results suggest that it may be possible to predict the tensile behavior of fiber-reinforced thermoplastics, with sufficient accuracy for many purposes, on the basis of a limited amount of experimental data, obtained under representative conditions. The suggested model may also be of use in defining the structural changes which are likely to be of the greatest value in improving the strength of existing materials. On the other hand, much more work on specimens of differing orientation distribution will be necessary to clarify the significance of the parameter α and to explore the variation of γ with fiber orientation.

References

1. F. J. McGarry, *Exp. Mech.*, **6**, 331 (1966).
2. H. T. Corten, in *Engineering Design for Plastics*, E. Baer, Ed., Reinhold, New York, 1964, pp. 869-994.
3. J. M. Whitney, *Text. Res. J.*, **37**, 1056 (1967).
4. J. R. Best and R. P. Wood, *Plastics Technol.*, **13** No. 11, 30 (1967).
5. G. Roos, *Kunststoffe*, **57**, 421 (1967).
6. S. Sterman and J. G. Marsden, *Mod. Plastics*, **44**, No. 6, 91 (1967).

7. R. V. Viventi, H. T. Plant and R. T. Maher, *Mod. Plastics*, **45**, No. 5, 129 (1968).
8. H. L. Wagner and G. F. Hardy (to Celanese Corp.), Belg. Pat. 681,490 (1966); Netherl. Pat. 6,607,041 (1966).
9. H. L. Wagner, G. F. Hardy, and R. A. Daniel, paper presented at 22nd Annual Technical Meeting of the SPE, Montreal, March, 1966.
10. F. M. Berardinelli, R. J. Kray, and T. J. Dolce (to Celanese Corp.), U.S. Pat. 3,313,767 (1967).
11. R. A. Isaksen, S. Newman, and Q. A. Trementozzi, *Polymer Eng. Sci.*, **7**, 94 (1967).
12. L. E. Nielsen, *J. Appl. Polym. Sci.*, **10**, 97 (1966).
13. L. E. Nielsen, *J. Compos. Mater.*, **1**, 100 (1967).
14. J. C. Schulz, paper presented at 18th Annual Meeting of the Reinforced Plastics Division of SPI, Chicago, February 1963.
15. B. W. Rosen, *AIAA J.*, **2**, 1985 (1964).
16. B. W. Rosen in *Fiber Composite Materials*, American Society for Metals, Metals Park, Ohio, 1965, pp. 37-75.
17. Owens-Corning Fiberglas Corp., *Textile Fiber Materials for Industry*, 1962.
18. T. F. MacLaughlin, *Exp. Mech.*, **6**, 481 (1966).
19. J. M. Whitney, *Text. Res. J.*, **37**, 1008 (1967).
20. S. W. Tsai, *Structural Behavior of Composite Materials*, NASA Contractor Report CR-71, July, 1964.
21. B. W. Abbott and L. J. Broutman, paper presented at 21st Annual Meeting of the Reinforced Plastics Division of SPI, Chicago, February 1966.
22. J. O. Outwater, *Mod. Plastics*, **33**, No. 7, 156 (1956).
23. V. G. Grinius, paper presented at 22nd Annual Meeting of the Reinforced Plastics Division of SPI, Washington, February, 1967.

Received September 6, 1968