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Critical issues in achieving desirable mechanical properties for short fiber composites

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Abstract - Short fiber reinforced polymers enjoy the advantage of providing stiffness levels achievable with continuous fibers while at the same time being moldable into complex shapes. This paper summarizes the understanding and prediction of stiffness, dimensional stability, strength, and fracture toughness and identifies areas in which further work is needed. The control of fiber orientation is discussed both from processing and data base points of view. Finally, the nature of time-dependent behavior and its implications are discussed briefly.

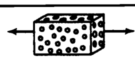
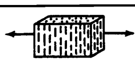
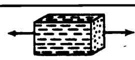
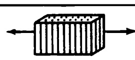
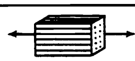
INTRODUCTION

Short fiber reinforced polymers were developed largely to fill the property gap between continuous fiber laminates used as primary structures by the aircraft and aerospace industry and unreinforced polymers used largely in non-load bearing applications. In some respects the short fiber systems couple advantages from each of these property-bounding engineering materials. If the fibers are sufficiently long, stiffness levels approaching those for continuous fiber systems at the same fiber loading can be achieved, while the ability of the unreinforced polymer to be molded into complex shapes is at least partially retained in the short fiber systems. Thus, short fiber reinforced polymers have found their way into lightly loaded secondary structures, in which stiffness dominates the design, but in which there must also be a notable increase in strength over the unreinforced polymer.

Table 1 illustrates the wide ranges of mechanical properties imparted to composites by fiber geometry and orientation for a glass fiber/epoxy system. Both the modulus and strength data are bounded on the lower side by the particulate (sphere)-reinforced material while the upper bound in both cases is given by the continuous fiber-reinforced systems. Between these two bounds lie the discontinuous fiber-reinforced systems on which this paper is focused. It is these systems, wherein fiber loading, aspect ratio, and orientation distribution all can vary, which present very challenging problems in property prediction and utilization.

This paper will review some currently critical issues which underly the efficient use of short fiber-reinforced systems. The understanding and prediction of stiffness, dimensional stability, strength, and fracture toughness will be examined. From a processing point of view, the control of fiber orientation distribution will be discussed. Finally, the nature of time-dependent behavior and its issues will be probed briefly.

TABLE 1. Experimental Stress-Strain Data for a Variety of Glass/Epoxy Systems.

	System (stress direction)	Filler Shape and Orientation	Strength, σ $\times 10^{-3}$ psi	Stiffness, E $\times 10^{-6}$ psi	Ultimate Strain, ϵ, %	Volume Fraction Filler, V_f
1	unfilled resin		10-12	0.3 - 0.4	4-5	0
2	bead filled		9-10.5	1.5-1.7	2.0-2.5	0.50
3	short fibers (transverse)		5.5	1.4	0.4-0.5	0.50
4	short fibers (longitudinal)		40	4.5	0.6-1.0	0.50
5	continuous fibers (transverse)		4-6	1.8-2.1	0.4	0.60
6	continuous fibers (longitudinal)		130-160	6.3-6.8	2.0	0.60

* abnormally low data due to anisotropic, low-strength tapes

GENERAL APPROACHES TO PROPERTY PREDICTION

There are two major approaches to mechanical property prediction for short fiber reinforced plastics. One is the "laminate analogy" which combines the micromechanics of combining different phases with the macromechanics of lamination theory. The success of the laminate approximation is strongly dependent upon the assumption of physical volume averaging in real natural systems, combined with an ability to estimate the properties of the individual plies, each of which contains uniaxially oriented fibers. Figure 1 schematically illustrates the equivalence between a random-in-a-plane short fiber reinforced composite and its laminate analogue. Only the top half of the laminate (above the midplane) is cut away to show the (0, ±45, 90°) orientation of the plies. As is implied in the figure, this approach has been used successfully to predict modulus (1), dimensional stability (1), strength (2), stress-strain behavior (2), and to a very limited degree, fracture toughness (3). The realities of distributions of both aspect ratio and orientation can be accommodated.

The other approach to property prediction has thus far been limited to the prediction of stiffness and dimensional stability. McCullough and coworkers (4,5) and Christensen (6) have used alternative approaches to arrive at upper and lower bounds for the stiffness problem. Both of these latter approaches can accommodate the general random 3-dimensional problem from a bounding viewpoint, but neither can explicitly accommodate the distributions of aspect ratio and fiber orientation.

In this paper concentration will be placed on the laminate analogy as a format from which to discuss the critical issues in short fiber reinforced materials.

Stiffness and dimensional stability

The design format for predicting stiffness for short fiber systems is well developed for 2-dimensional structures. By employing a combination of micromechanics (Halpin-Tsai equations) and macromechanics (laminated plate theory--the laminate analogy), one can calculate the in-plane stiffnesses and the Poisson ratio (1,7,8) for any 2-dimensional fiber orientation distribution. Furthermore, one can also account for the reality of a distribution of fiber aspect ratios (9), which is a natural consequence of breakage in extrusion and injection-molding equipment.

Table 1 and Fig. 2 summarize the situation for the 2-dimensional plane-stress stiffness problem for the glass-epoxy system. Clearly the modulus depends on the fiber aspect ratio, the volume fraction of fibers, the fiber-to-matrix modulus ratio and the fiber orientation distribution. Note particularly in Fig. 2 that for a unidirectionally oriented ply, the critical aspect ratio has almost no dependence on fiber volume fraction, but does significantly depend on the fiber-to-matrix modulus ratio. However, a fictitious five-fold increase in E_F/E_m (keeping E_m constant) increases the critical aspect ratio (at which the continuous fiber modulus is approached) from about 100 to over 200 for a fiber volume fraction of 0.5. These same aspect ratio dependencies are reflected in any fiber orientation distribution including that of random-in-a-plane.

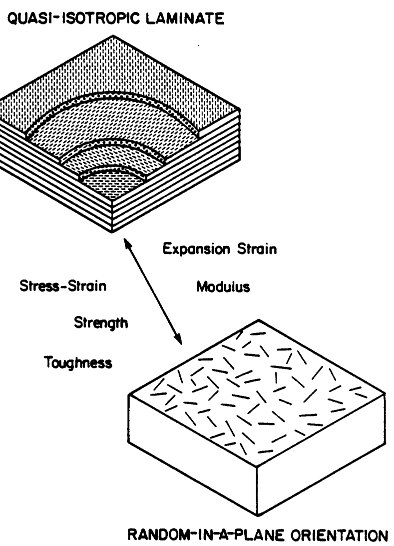


Fig. 1. Schematic of laminate analogy for predicting mechanical properties of 2-dimensional short fiber composites.

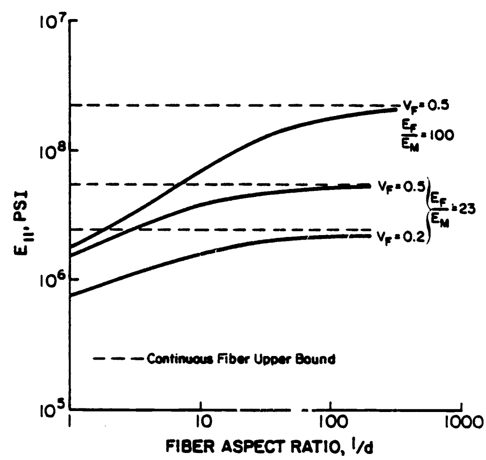


Fig. 2. Effects of fiber aspect ratio, fiber volume fraction, and fiber-to-matrix stiffness ratio on the longitudinal stiffness of unidirectionally oriented, short glass fiber/epoxy composites.

Many structural applications utilizing short fiber composites in reality have 3-dimensional fiber orientation distributions; even thick sheets may have out-of-the-plane distribution components. As yet there is no direct and efficient calculation format to handle the general 3-dimensional problem. Some bounding cases have been examined, however. Halpin, Jerina, and Whitney (9) treated the orthogonal 3-dimensional problem by modeling a plain, square woven fabric pierced by a straight yarn perpendicular to the fabric plane. Results showed that the moduli in the plane of the woven fabric in the 3-dimensional case were about 7% lower than the comparable moduli for the 2-dimensional material (plain-square weave) at 50v% loading. Thus, the 3-dimensional weave overcomes the low shear strength between layers of a 2-dimensional fabric laminate with only a small sacrifice in laminate in-plane stiffness.

Lavengood and Goettler (10) used an approximate averaging technique to generate a rule-of-thumb expression for the modulus of a structure having 3-dimensionally random fiber orientation,

$$\bar{E}_1 = \frac{1}{5} E_{11} + \frac{4}{5} E_{22} , \tag{1}$$

where E_{11} and E_{22} are the longitudinal and transverse engineering stiffnesses for a unidirectionally oriented ply. At 30v% loading, Equation 1 predicts a value about 20% lower than the in-plane stiffness for random 2-dimensional orientation. A rule-of-thumb expression for the latter is,

$$E_1 = \frac{3}{8} E_{11} + \frac{5}{8} E_{22} \tag{2}$$

Thus, it is clear that more work needs to be done to provide a general format for handling any 3-dimensional orientation distribution.

Dimensional stability (i.e., expansion coefficients) during temperature excursions or exposure to swelling environments is a stiffness-dominated phenomenon which depends on the same parameters as stiffness as well as the dimensional stabilities of the fiber and matrix. Like stiffness, two-dimensional geometries are well understood and describable (8). However, the 3-dimensional situation is more complicated and reliable design formats, even for the bounds, are not yet available.

For both stiffness and dimensional stability, essentially continuous fiber property levels can be achieved if the fiber aspect ratio is high enough (see Fig. 2). In many short fiber systems, however, the important ratio is not the individual fiber aspect ratio, but a fiber bundle aspect ratio. For processes involving molding compounds which are basically encapsulated fiber bundles, it is rare to find well dispersed individual fibers in the final part. The degree of adhesion at the interface does not affect stiffness and dimensional stability (11). All that is required of a good system is that there be good material contact, i.e., no voids at the interface.

Strength

One may also analyze the short fiber strength problem in terms of the laminate analogue model described above. Again the problem breaks down into describing the behavior of a single unidirectionally oriented ply and then viewing the laminate as a combination of unidirectionally oriented plies. The longitudinal strength of a unidirectionally oriented short fiber ply depends, in addition to those factors mentioned above for stiffness, on the strength of the interface, the strength of the fiber and, in ways different from the stiffness, on the fiber (or bundle) aspect ratio. Unlike for stiffness, continuous fiber composite strengths cannot be attained in discontinuous fiber systems, even at extremely high aspect ratios (12). For unidirectionally aligned systems, the plateau strength values in the fiber direction for very high aspect ratios rarely exceeds 70% of the strength of continuous fiber systems at the same fiber content. Furthermore, the critical aspect ratio, at which the maximum strength is achieved in short fiber systems, is usually much higher than that needed to achieve the maximum (continuous fiber) stiffness for the same fiber volume loading in the same system. One can appreciate this point by comparing Figs. 2 and 3 at a fiber volume fraction of 0.5. In the case of strength, the plateau value is attained at an aspect ratio of about 500 whereas the critical aspect ratio for stiffness occurs at about 100. This difference in critical aspect ratios between strength and stiffness depends on the interface condition and the fiber-to-matrix stiffness ratio, as well as the fiber volume fraction.

The strength of a uniaxially aligned fiber ply may be predicted utilizing the following set of equations (13):

$$G = \frac{[SRF] - [SRF]_o}{[SRF]_\infty - [SRF]_o} = 1 - 0.97 \exp\,[-0.42\beta] \tag{3}$$

$$[SRF]_{\infty} = 0.5 + (E_F/E_m)^{-0.87}$$

$$[SRF]_o = \frac{\sigma_m E_c (1-V_F)^{1/3}}{\sigma_F V_F E_m}$$

$$[SRF[= \bar{\sigma}_c/\sigma_F V_F$$

$$\beta = (\ell/d)/(\sigma_F/\tau_m)$$

SRF stands for strength reduction factor; σ_F, σ_m and $\bar{\sigma}_c$ are the strengths of the fiber, matrix and continuous fiber composite, respectively; V_F is the fiber volume fraction; ℓ/d is the fiber (bundle) aspect ratio; E_F, E_m, and E_c are the tensile moduli of the fiber, matrix, and sphere-filled (ℓ/d = 1) composite, respectively; τ_m is the matrix shear strength or the interface shear strength, whichever is lower. The strength reduction factor [SRF] is defined as the uniaxially aligned, short fiber system strength divided by the strength of an aligned continuous fiber system having the same volume fraction fibers. As the aspect ratio approaches unity, the [SRF] approaches that for a sphere-filled system, namely $[SRF]_o$. $[SRF]_\infty$ is the value for the [SRF] at large fiber aspect ratios where the plateau behavior is observed; it is a weak function of fiber volume fraction (13). Figure 4 shows the normalized master curve (G vs. log β) from which the strength reduction factor may be calculated for any perfectly aligned short fiber composite system. Thus, knowing the input parameters of equations (3) allows one to calculate the strength in the fiber direction of a uniaxially aligned short fiber ply or composite.

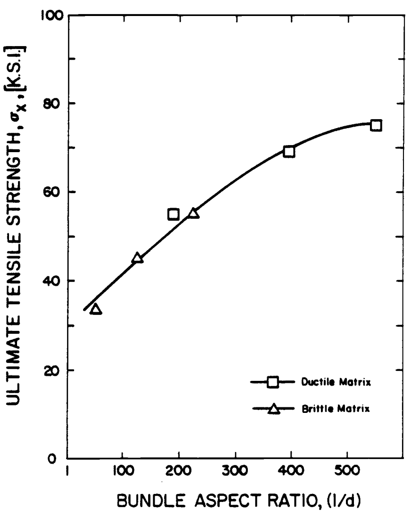


Fig. 3. Dependence of longitudinal tensile strength on the bundle aspect ratio, ℓ/d, for ductile and brittle epoxy matrix systems containing 50v% aligned glass fiber bundles.

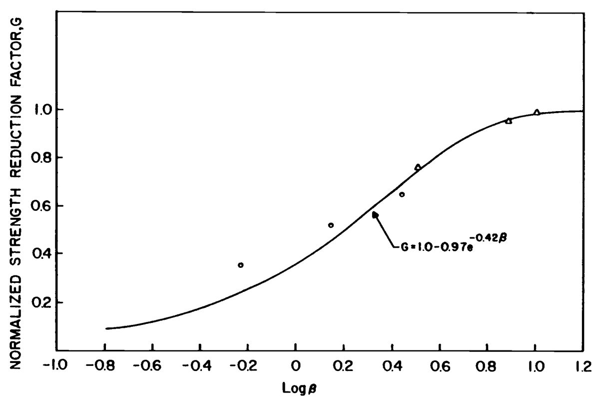


Fig. 4. Comparison of theory (solid line) with experimental data for E-glass/ epoxy systems; Δ-ductile matrix, O-brittle matrix.

The second portion of the strength calculational format requires the choice of a failure criterion. One must decide what phenomenon governs the failure of the individual plies in the laminate as it is stressed and strained. From among a number of possible choices, the maximum strain criterion seems to adequately describe glass fiber/epoxy results (2).

In order to calculate the strength for a random-in-a-plane short fiber composite, one utilizes the strength reduction factor along with the ply moduli to calculate failure strains for each ply in the laminate. As the laminate is strained, ply failure stress levels are noted, the laminate moduli are recalculated after each ply failure and the strength is the sum of the increments of stress the laminate went through until the last ply failed.

Figure 5 shows the predicted strength for a random-in-a-plane fiber orientation, along with experimental data from both brittle and ductile matrix, glass/epoxy systems. The prediction is for the brittle matrix system and provides a reasonably good (and conservative) engineering estimate of the strength. Reasonable predictions using the above approach have also been achieved for non-random orientations (14).

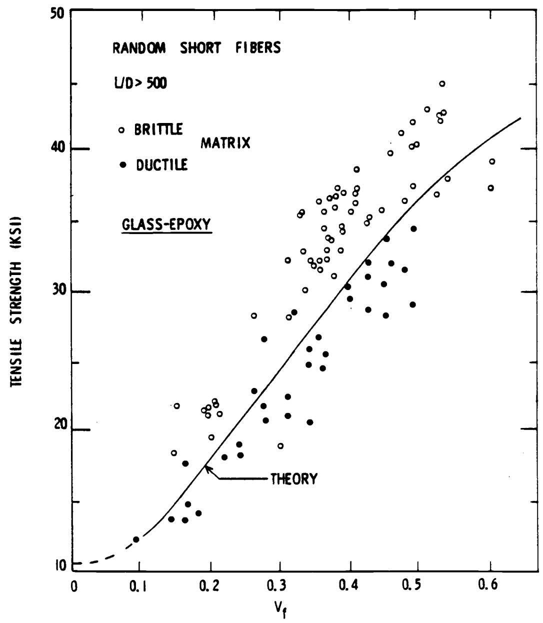


Fig. 5. Theory utilizing the laminate analogy and maximum strain failure criterion (solid line) for 2-dimensional random orientation. Note difference between brittle and ductile matrix data.

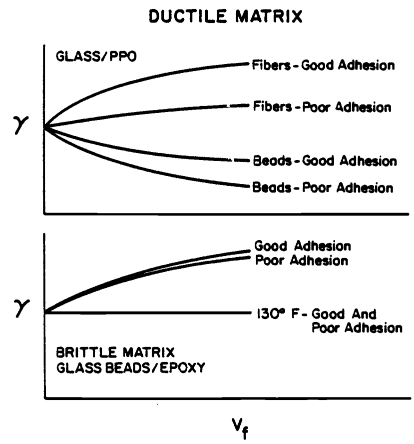


Fig. 6. Qualitative effects of reinforcement geometry, matrix ductility, degree of adhesion, and volume loading on the fracture toughness of glass reinforced plastics (15-17).

There are a number of important issues which emanate from the above approach. The degree of adhesion is extremely important and is reflected in the interface strength term, τ_m. This term is not predictable and is extremely difficult to measure experimentally. The use of single fiber pull-out tests may be misleading because these results do not account for the very important fiber-fiber interactions in the composite. The strength of flaw-sensitive fibers such as glass (σ_F in the above format) is dependent on fiber length and the intrinsic strength of the actual short fiber must be used in any predictive format. The 3-dimensional short fiber strength problem has barely been touched and will certainly become a more important issue as these systems head toward uses as primary structural materials.

Fracture toughness

The fracture toughness of polymeric composites is probably one of the least understood of all the mechanical responses. For most composites, including short fiber systems, a sometimes espoused rule of thumb is that as the strength increases, the toughness decreases. Thus, it might be implied that as the degree of adhesion increases, the toughness should decrease. While this is true generally for continuous fiber reinforced brittle matrices, it is not the case for bead-filled systems, nor for short fiber reinforced ductile matrices.

Figure 6 qualitatively summarizes some of the results obtained by DiBenedetto and coworkers (15-17). Improving the adhesion in a short glass fiber/polyphenylene oxide system actually increases the fracture toughness as measured in a double-edge-notched tensile test. The same trend is clear in the glass bead/PPO system. Thus, the reinforcement geometry and the matrix ductility are important fracture toughness considerations.

Although various attempts have been made to increase toughness by adding a ductile third-phase material either dispersed in the matrix or selectively located at the interface, there is still no good format for predicting, a priori, the toughness of a composite system. A start toward this goal has recently been made by Tsarnas and Kardos (3). Utilizing the general micro-macromechanics, laminate analogy described above, they expressed the fracture toughness, measured in a double-edge-notched tensile test, as a function of the elastic moduli, strength, and stress intensity factors of unidirectionally aligned, short fiber plies. For a random-in-a-plane fiber orientation in a sheet under plane stress, the final design equation is

$$\gamma_R = \frac{2(\gamma_1+\gamma_2)}{\pi} \qquad (4)$$

where γ_1 and γ_2 are the fracture toughnesses for a single unidirectionally oriented ply with the notches oriented perpendicular and parallel to the fiber direction, respectively.

Figure 7 shows the predicted toughness values (solid lines) for two different fiber volume fractions for random-in-a-plane fiber orientation in a glass/epoxy system. Also shown is the experimental data which contains considerable scatter. The predictions indicate a weak critical aspect ratio effect although the experimental scatter makes it difficult to corroborate this prediction. Clearly there is an urgent need for more theoretical and experimental work on the toughness of short fiber composites.

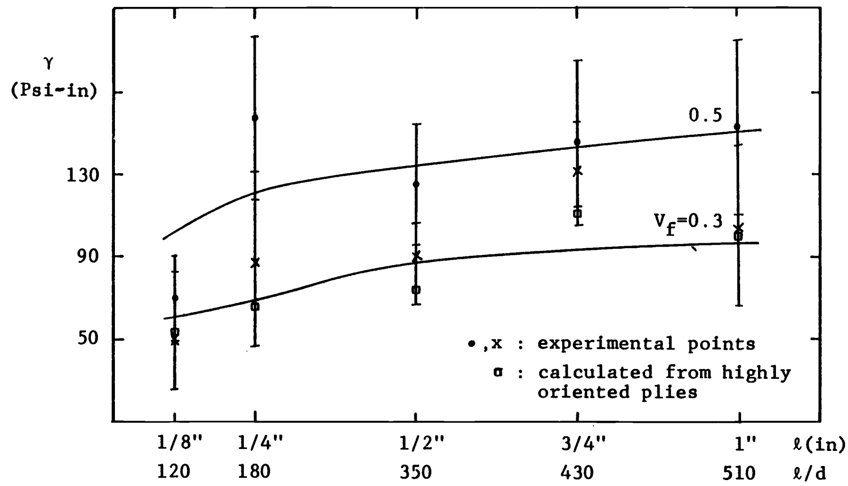


Fig. 7. Fracture toughness vs. fiber bundle aspect ratio (length) for isotropic (random-in-a-plane fiber orientation).

Orientation distributions

It is clear from the above discussion that fiber orientation distribution is one of the most crucial variables which determine the mechanical properties of short fiber composites. And yet, the technical literature abounds with a) data comparisons between samples which have been processed differently but assumed to have the same fiber orientation, or b) data compilations for systems for which the orientation distribution has not even been measured. One reason this occurs is because it is often difficult to measure the fiber orientation distribution. If the matrix is amorphous and the fibers crystalline, wide-angle pole figure X-ray analysis may be utilized to provide the distribution (18). If the matrix is transparent, Fraunhofer diffraction may be used (19), or a small fraction of the glass fibers may be prestained with an optically opaque dye and the distribution obtained from image analysis (14). Three-dimensional orientations may be characterized by sectioning the sample along orthogonal planes and analyzing the fiber images on each of these planes (20,21).

Control of fiber orientation during processing is probably the most important factor underlying the efficient use of short fibers in composites. Goettler has shown (20,22) that fiber-matrix interaction, gate geometry, fiber length, flow rate (maximum shear rate), temperature, and fiber volume loading are among the very important parameters which control the final fiber orientation in extrusion as well as injection and transfer molding. He has utilized this basic knowledge to design an expanding mandrel tube extrusion die which aligns short fibers in the circumferential direction of the extrudate (23). Advani and Tucker (24) have described 3-dimensional orientation distributions in terms of 4th-order tensors and attempted to relate these tensors to the rheological properties of fiber suspensions, thereby building a bridge between processing parameters and final performance properties. This area of research represents one of the most difficult, but most important barriers to the advancement of short fiber composite technology.

Time dependent behavior

For cases of very lightly loaded structures, creep and fatigue are not normally a problem; however, it is extremely difficult to know at what precise stress levels time dependency begins to become important. Historically, the temptation has been to unknowingly push short fiber reinforced thermoplastics into performance load ranges in which time dependency produces dimensional instability as well as eventual failure. Exactly how time-independent fibers perturb the time dependence of neat polymer is a crucial question.

Generally it is observed that a moderately reinforced polymer (up to 40 weight % randomly oriented short glass fibers) will retain the time- and temperature-dependent characteristics of the matrix polymer, except that a) the modulus at a given strain level is proportionately higher, and b) the maximum extent of the plastic deformation that can be sustained before fracture will diminish (25). Scaling rules have been established for both glassy and crystalline polymers by Matsuoka (26,27), which allow the prediction of creep and stress relaxation behavior from scaled stress-strain data. Jerina et al. (28) and Nicolais et al. (29) have superimposed composite micromechanics upon a time-dependent matrix to predict time-dependent composite behavior. Of the myriad issues facing the advancement of short fiber reinforced polymer technology, the characterization and prediction of time-dependent behavior may eventually be the most crucial.

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