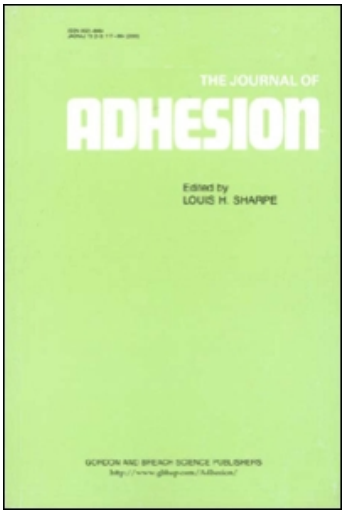

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A New Single Fiber/Resin Interface Test for Highly Cross-Linked Resin Systems

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A new single fiber/resin interface test technique has been developed to test fibers in highly cross-linked resins that were difficult to characterize before. The new approach was based on the fracture mechanics considerations to allow brittle thin resin film with embedded single fiber to undergo large strain elongation. This was achieved by sandwiching the resin between high elongation epoxy doublers. Several systems with graphite fibers in highly cross-linked epoxy and bismaleimide resins were successfully characterized for strains up to 6%. Because of the thin resin film used, even matrix materials with limited transparency in bulk form could easily be tested. At sufficiently high strains, the fiber/resin systems showed interface-related behavior consistent with their edge delamination performance previously studied.

KEY WORDS Bismaleimide resin; epoxies; fracture mechanics; graphite fibers; interface-related behavior; laminated composites.

INTRODUCTION

Laminated composites are designed to utilize the desired properties of fibers. The composite performance, however, is often limited by weak links other than the fibers in the materials. For example, matrix-controlled failure can easily be induced at load levels well before the fibers are broken. In addition, the micro-mechanical response of composites involves the load transfer between fiber and matrix. Insufficient strength at or close to the fiber/matrix interface will also lead to premature failure of the composites.

The exact role of the fiber/matrix interface in controlling composite failure is not fully understood. The interfacial strength is, in the first place, difficult to quantify. A certain degree of success has, however, been reached in tests using a single filament for interfacial shear strength measurement. The fiber pullout method[1–3] and the embedded single fiber test[4–6] are two most representative techniques of this kind. Although both tests offer reliable measurements related to the interface, difficulties in conducting tests and interpreting results still exist.

The embedded single fiber test determines the "critical length" of the fiber in a neat resin specimen under tension. Such information can then be related to the

interfacial shear strength and fiber strength. As each specimen can yield many fiber segments, a relatively small number of specimens is needed for each evaluation. However, as the specimen has to be loaded to a fairly large strain level (above 3%), only relatively flexible resins have been used in the past. For example, most of the work in this area involving thermoset resins has been based on high elongation diglcycidyl ether of Bisphenol A (DGEBA) epoxy ststems.[4-6] For multi-functional epoxies and bismaleimide (BMI) with relatively high glass transition temperature, T_g, for aerospace applications, their relatively low elongation to failure has so far prevented them from being characterized directly using this technique. Besides, the necessity of optically observing the fiber precludes the use of matrices with insufficient optical transparency.

The method developed by Jacques and Favre[7] used a single fiber coated with brittle resin and then embedded in bulk ductile resin for a fragmentation test. Although this method could allow fibers in brittle resins to be evaluated, certain difficulties exist in specimen preparation. Many thermoset resins, especially in modified forms, have relatively high viscosity in the processing temperature range. Coating fibers with such resins was difficult, if not impossible, without also damaging the fibers. Dissolving resins in solvents may solve the coating problem but is not always desirable for the morphological stability of the formulated resins.

In this paper, attempts have been made to apply the embedded single fiber test to characterize the interface-related shear strength between graphite fibers and relatively brittle resin systems. A new technique has been developed to allow highly cross-linked resins, in readily available film form, to be used as the matrices for the fibers. The approach also made it possible to work with resins that had limited transparency in bulk form. Several fiber/matrix systems were evaluated using this new method. The test results were compared with other interface-sensitive composite properties.

EXPERIMENTAL DETAILS

Background

The single fiber technique normally uses a specimen of dog-bone-shaped neat resin in which a single fiber is embedded, as shown in Figure 1. The specimen is then subjected to tensile loading to result in the breaking of the embedded fiber into fragments. As the load increases, the fragments will reach a critical length l_c which is too short to transfer stress needed to break the fiber. l_c can be related to the shear strength τ_c at resin/fiber boundary by [4-6]

$$\tau_c = \frac{\sigma_f d}{2 l_c} \tag{1}$$

where σ_f is the fiber tensile strength and d the fiber diameter.

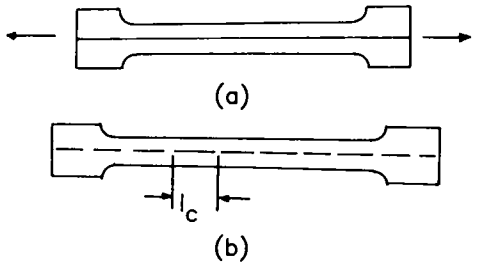

FIGURE 1 (a) Dog-bone-shaped specimen under tensile loading to induce (b) fiber fragments of critical length l_c.

It should be mentioned that, as pointed out by Bascom and Jensen,[6] τ_c cannot be strictly interpreted as the fiber/matrix interfacial shear strength. Direct evidence of the interface controlling the type of failure involved in the test has not been conclusive. In fact, there was an indication of the failure being related to the shear yield strength of the resin. As the fiber/matrix stress transfer may or may not be limited by the exact interface, τ_c will be simply defined here as the interface-related shear strength.

New test specimen design

The new technique developed here basically placed a single fiber between two uncured thin resin films (~0.0038 cm thick each) which were then sandwiched between two thick doublers (0.25 cm thick each), as shown in Figure 2. The doublers were made of high elongation epoxy DGEBA cured with poly-

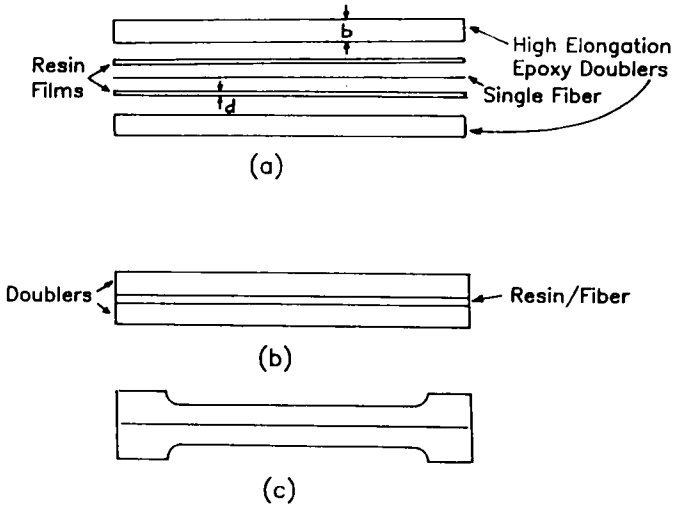

FIGURE 2 Schematics of (a) a sandwich of doublers, resin films and single fiber to form the specimen ((b) side view and (c) top view) after assembly.

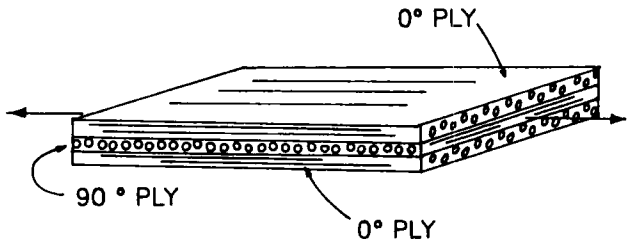

FIGURE 3 Schematic of a $[0°/90°]_s$ laminate subjected to tension in the 0° direction.

oxypropylene diamine (Jefferson Chemical Co., D230). The thin resin films could be any matrix systems of interest (*e.g.* epoxies and BMI) and quite often were taken directly from the hot-melt coated resin films for prepregging purposes. In this way, resin materials closest to their matrix form in composites were evaluated.

The doublers were already cured and the resin films uncured when the sandwich was assembled. The bonding surfaces of the doublers before assembly were lightly sand-blasted and thoroughly cleaned with isopropyl alcohol. After the assembly, the specimen edges were sealed by Mylar tape to prevent resin from flowing out. The specimen was under spring clamp pressure and heated in an oven to cure the resin. A dog-bone configuration of 0.8 cm × 6 cm in the gage section was machined to the specimen after completion of cure.

The central idea of the new specimen design was to allow the specimen to undergo the necessary elongation without premature failure of the resin. The DGEBA-based doubler materials have been shown to sustain large deformation (*e.g.* see references[4–6]). The thin resin layer in which the fiber was embedded was normally not expected to have the needed strain capability in its bulk form. However, when it was sandwiched between the doublers, the strain to failure of the resin was greatly increased. This was analogous to the thickness dependence[8–10] of failure strain of a 90° ply sandwiched between 0° plies in a $[0°/90°]_s$ type of composite as shown in Figure 3. The theoretical basis of this approach is given below.

Theoretical basis for the new specimen design

Although the theoretical basis for the 90° ply failure problem was established by Bailey *et al.*[8–10] for composites, the same principles can be applied here for the resin/fiber assembly shown in Figure 2. Their approach was based on the fracture energy balance that had to be satisfied to induce cracks in a weak (or brittle) material (such as thin resin or 90° ply) sandwiched between a strong (or ductile) material. The strain to failure ε of the inner material can be expressed in terms of the thickness, elastic properties and fracture toughness of the materials in the assembly. The detailed derivation can be found elsewhere[9] and is not repeated here. The major results, however, are given below after some rearrangement.

For the sandwich assembly shown in Figure 2, the minimum failure strain ε of the thin resin is given by

$$\varepsilon = \left[\frac{2G_{IC}}{dE_2}\left(\frac{bE_1G_2}{(E_2)(bE_1 + dE_2)}\right)^{1/2}\right]^{1/2} \tag{2}$$

where b and d are the thicknesses (Figure 2), E_1 and E_2 are the moduli of the doubler and fim materials, respectively; G_2 and G_{IC} are the shear modulus and the fracture toughness of the fim material. From Eq. (2) it is clear that ε increases with decreasing film thickness d. For the new specimen design with $b = 0.254$ cm, $d = 0.0038$ cm, $E_1 = 2.76$ GPa (DGEBA epoxy), $E_2 = 4.14$ GPa (multifunctional epoxy), $G_2 = 1.59$ GPa, $G_{IC} = 100$ J/m^2 (worst case), ε was estimated from Eq. (2) to be 2.9%. Such an ε value was considered adequate, especially when many resins of interest had higher G_{IC} and thus larger ε.

Test fixture

To conduct the single fiber test, a fixture was constructed (as shown in Figure 4) to load the specimen in tension through a hand-driven mechanism. The tester was placed under an optical microscope for direct observation. The specimen was viewed between cross polarizers to enhance the birefringent image of the fiber segments. A strain gage was also mounted on the gage section of the specimen but outside the viewing area. The strain gage was conditioned using a Vishay 2300 Signal Conditioning Amplifier (a Measurement Group product). The strain of the specimen was constantly monitored during the test.

Materials

Several resins used as matrices in CIBA-GEIGY composite systems were chosen for the study. The epoxy-based resins were R922, R914 and R6376. These resins, with T_g above 180°C, have mainly been used for aircraft structural applications. A toughened BMI resin R6452 which had a T_g of 280°C was also studied. All the resins were directly taken from the hot-melt coated films (0.0038 cm thick) for

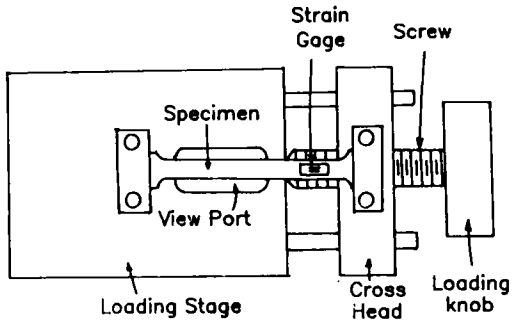

FIGURE 4 Test fixture for the single fiber/resin tests in this study.

prepregging purposes. The resins in bulk form had tensile strains to failure less than 2.5% under normal test conditions.

Two graphite fibers, T500 (AMOCO) and IM6 (Hercules) were specifically examined. These fibers gave the highest and the lowest edge delamination strengths,[11] respectively, among several fibers in both the R914 and R6376 laminates previously studied. To demonstrate the extreme case of poor fiber/matrix adhesion, some T500 fibers were treated with RAM 225 mold release agent (RAM Chemical). The fibers were immersed in a solution of 25% RAM 225 in heptane/isopropyl alcohol mixture (2:1 weight ratio) for 1 minute. The weight gain of the treated fibers after drying was about 1% corresponding to an estimated fiber surface coating 0.025 μm thick.

The doubler material (0.254 cm thick) was cast from CIBA-GEIGY Araldite GY 6010 (DGEBA) and hardener D230 (Jefferson Chemical). The doublers were cut from the cured plates, sandwiched with the resin/fiber and then subjected to the curing cycle of the resin used. The epoxy systems were cured at 177°C for 2 hours. The BMI resin (R6452) was cured at 80°C for 2 hours, 177°C for 4 hours and post-cured at 250°C for 4 hours.

Initially, several thermoplastic plates of polycarbonate and acrylics (Plexiglass) were experimented with as the possible doublers. These materials, however, could neither survive the curing process nor bond well to the resin films when under load. The epoxy-based doublers solved all these problems.

RESULTS AND DISCUSSION

Experiments conducted on a variety of fiber/resin systems successfully yielded fiber fragments as shown in Figure 5 for (a) R922/IM6, (b)R6376/IM6 (c) R6376/T500 and (d) R6452/T500. The optical microscope observations of fiber fragments were very similar to those for fibers in bulk resin casting reported in the literature. The fibers in the sandwich specimens studied here were fully embedded in the thin resin films, as a typical cross-sectional view of such a specimen in Figure 6 shows.

The resin films sandwiched between the doublers were seldom fractured before the final doubler failure. Only occasionally localized resin cracks close to the broken fiber ends occurred during loading. The cracks were confined in the resin layers and did not propagate into the doubler materials. The observations confirmed the effectiveness of the specimen design.

It is interesting to observe one particular specimen (R914/IM6) where resin cracks extended from the fiber for 1 to 3 mm in lengths, as shown in Figure 7. These cracks, however, did not appear to affect the fiber fragmentation simply because the doublers effectively transferred load throughout the specimen. All such resin cracks were observed to originate from the broken fiber ends instead of resulting in fiber breakage.

For each system, the critical fragment lengths of a large number (~140) of fiber segments were measured under the microscope. The fragment lengths l_c varied in

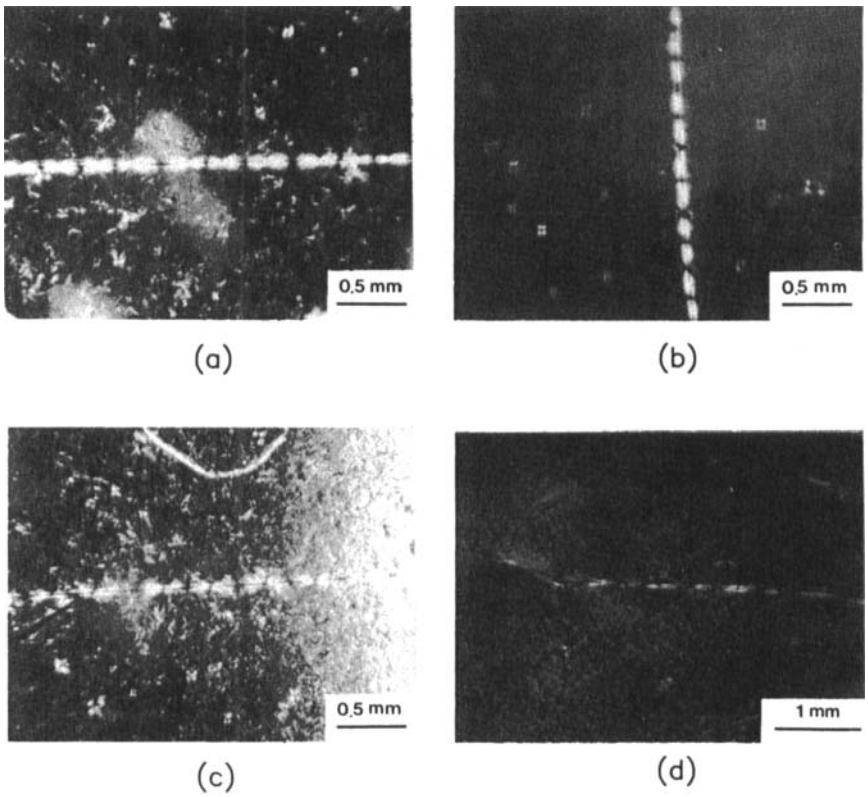

FIGURE 5 Optical micrographs of fiber fragments of (a) R922/IM6, (b) R6376/IM6, (c) R6376/T500 and (d) R6452/T500.

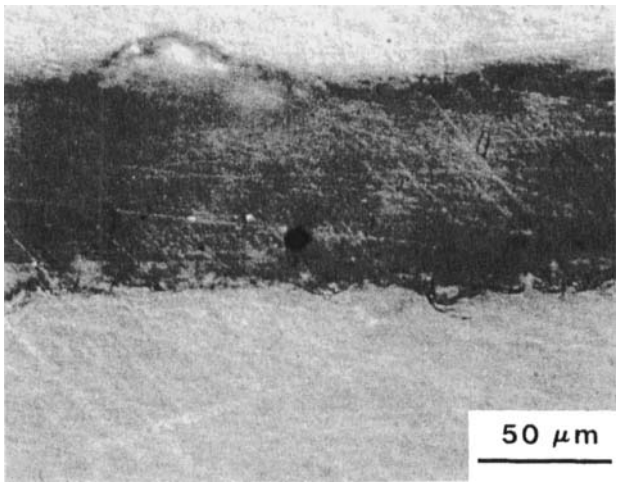

FIGURE 6 Cross-sectional view of the sandwich assembly of single fiber embedded in the resin between doublers.

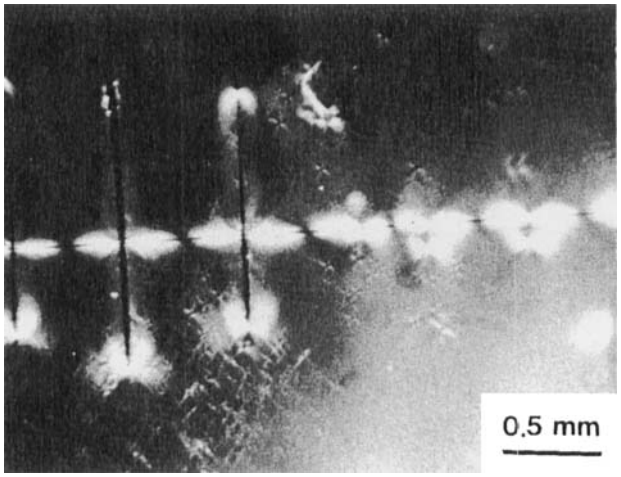

FIGURE 7 Resin cracks originating from the broken fiber ends showing little effect on the fiber fragmentation process.

a broad range. Such data could in principle be treated statistically. Although Weibull distribution has often been used for this purpose, there were uncertainties in using such a distribution.[6] Besides, fiber strength σ_f in Eq. (1) needed to be first determined for its Weibull distribution. This had to be done for fiber with length at or near l_c which was difficult to achieve. There was, however, evidence[6] showing that average fiber strength correlated well with the normal mean of (l_c/d). For the purposes of verifying the technique and determining the relative performance between different fibers, this simple mean value approach was used in this study.

To obtain meaningful l_c results, the specimens had to be subjected to high enough elongation for l_c to reach asymptotic values. The test fixture as designed at present could yield strains as high as 6%. However, several systems were loaded only to 3% of strain before the specimens were fractured. Further modification of the test setup or different doubler materials will, undoubtedly, allow higher elongation of the specimens.

The fragment lengths l of different systems are given in Figure 8 as a function of strain. The results indicated that the fragment lengths began to level off to reach the critical l_c values for strain above 4%. Although not all the fiber systems were fragmented to yield l_c, the effectiveness of the test technique was largely demonstrated.

Based on the fragment length results, the interface-related shear strengths τ were calculated from Eq. (1) using the σ_f and d values given in Table I for different fibers. It should be noted that τ was different from τ_c unless the true l_c was reached. However, τ values so calculated as a function of strain in Figure 9 showed the relative interface-related performance among different systems.

For strain at 3%, the relative performance between IM6 and T500 fibers varied with the resins. While IM6 had higher τ than T500 for R922 and R6376 resins, the

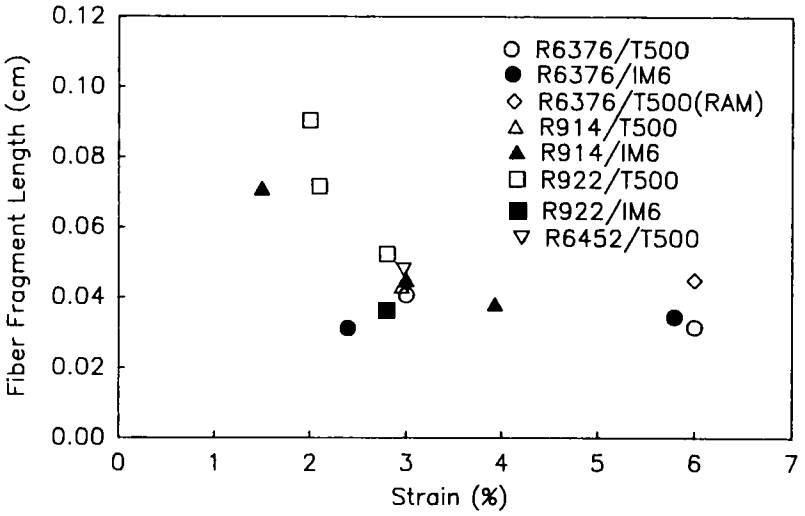

FIGURE 8 Fiber fragment length plotted against strain.

TABLE I
Fiber diameters d and strengths σ_f

Fiber Type	d (μm)	σ_f (MPa)
T500	7.0	3.72
IM6	5.4	4.38

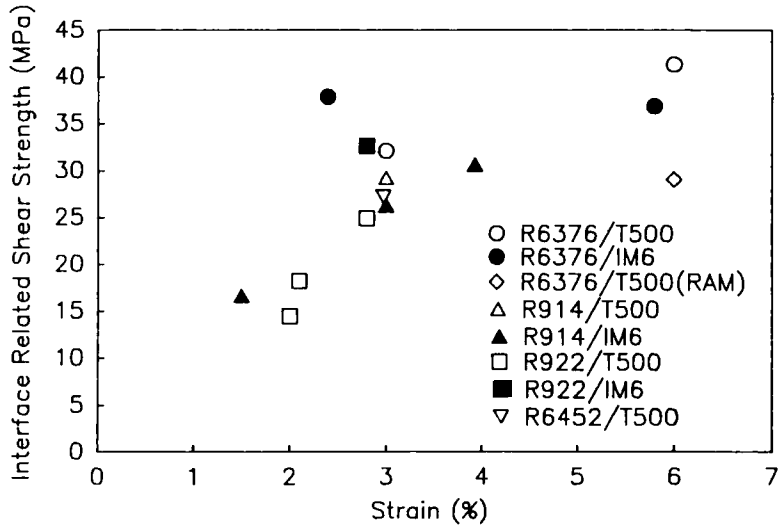

FIGURE 9 Interface-related shear strength as a function of strain.

TABLE II
Edge delamination fracture toughness G_c and strengths σ_c of $[\pm 30/\pm 30/90°/90°]_s$ laminates[11] and interface related shear strengths τ_c

Laminate system	G_c (kJ/m^2)	σ_c (MPa)	τ_c (MPa)
R6376/T500	0.54	343	41.3
R6376/IM6	0.22	232	36.8
R6376/T500 (RAM-Treated)	0.20	216	29.0

reverse was the case for R914. The lack of a definitive trend perhaps indicated that 3% strain was not sufficient to induce the critical length l_c.

At 6% strain level, however, l_c values of several fiber/matrix systems appeared to be reached to yield meaningful τ_c for comparison. The interface-related shear strengths of R6376/T500, R6376/IM6 and R6376/T500 treated with RAM 225 were in decreasing order as shown in Figure 9. These results were consistent with the observed interface-sensitive edge delamination performance of these systems.[11] The edge delamination fracture toughness G_c and strength σ_c values as given in Table II of $[\pm 30°/\pm 30°/90°/\overline{90°}]_s$ type of specimens were in the same order.

It is interesting to point out that the BMI R6452 was dark brown and had limited transparency in its bulk form. However, under the microscope the definition of fiber fragments was observed (Figure 5(d)) because of the relatively thin layer of resin used. After the relatively high temperature cure and post cure, the doublers were still largely transparent without indication of degradation. No debonding occurred between the doublers and BMI resin throughout the strain range tested.

The fiber fragment length and interface-related shear strength values of R6452/T500 were comparable with those of the epoxy systems as shown in Figure 8 and 9. Because l_c and τ_c of this system may not have been reached at 3% strain, its performance relative to the other systems could not be exactly determined. However, the applicability of the technique to highly cross-linked systems such as BMI was demonstrated.

CONCLUSIONS

A technique of single fiber test for interface-related shear strength measurement was successfully developed to test fibers in highly cross-linked resins with limited elongation. The central idea of the new approach was to sandwich a thin resin film, in which the fiber was embedded, between epoxy doublers with large elongation. When the specimens were loaded in tension, the constraint of the doublers delayed the resin failure to allow the fiber fragments to develop fully.

The new technique was shown to be able to yield large strain for critical interface-related shear strength calculation. Although many specimens failed to

develop high enough strain levels for such calculations, the limiting factors there were the loading fixture alignment and, to a lesser degree, the doubler materials used. The sandwiched thin resin film of interest largely sustained high strains allowed by the doublers without disturbing the fiber fragmentation process.

Several fiber/resin systems were characterized using the new method for strain up to 6%. At sufficiently high strain, T500 fiber was found to have higher interface-related shear strength than IM6 and release-agent-treated T500 in the R6376 resin matrix. This behavior was consistent with the interface-sensitive edge delamination of these systems studied previously. Overall, the new method successfully evaluated systems as brittle as BMI, thus removing the previous restrictions related to the original single fiber test.

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