DELAMINATION FRACTURE STUDY OF GRAPHITE/EPOXY COMPOSITE MATERIALS

by

Laurie M. Veeder

Mechanical Engineering

Submitted in Partial Fulfillment of the Requirements
of the University Undergraduate Fellows Program

1983-1984

Approved by:

Walter L. Bradley

Dr. Walter L. Bradley, Faculty Advisor

April 1984

ABSTRACT

Delamination Fracture Study of Graphite/Epoxy Composite Materials

Critical energy release rates have been determined for two composite systems. The effect of resin thickness on fracture toughness in brittle fiber - ductile resin systems has been investigated. Material resin content has been varied through manufacturing for one composite. Two different specimen geometries and the use of epoxy resin with and without fibers has produced a variation in resin thickness in the other material. Electron microscopy has allowed for detailed examination of the fracture surfaces. Composites with a thicker region of ductile resin exhibit higher energy release rates than do those with less resin in the crack growth region.

ACKNOWLEDGEMENTS

I would like to express my sincere thanks to Dr. Walter Bradley for his continuous support and encouragement during the course of this study.

I would also like to thank Carl Fredericksen for his infinite patience and expert help and advice in specimen preparation and testing, and also Bob Harbert for his advice and assistance in using laboratory equipment.

A special thanks goes to Liz Chakachery and Bill Jordan for their assistance in using the scanning electron microscope and preparing photographs for this report.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

INTRODUCTION

Graphite/epoxy composites have received increased attention from designers in recent years, particularly for aerospace applications. Most of this interest stems from the low density and high strength and stiffness of these materials. Due to their anisotropic nature, fiber composites are subject to delamination, with present limiting design strains being primarily dependent on failure by compressive buckling. Increased resistance to delamination could increase design strains by as much as fifty percent (1).

Early graphite/epoxy composite systems were designed to maximize resin stiffness and glass transition temperature. While material characteristics for in-plane stresses are dependent on the properties of the graphite fibers, the behavior of the composite subjected to out-of-plane loading will be determined by the resin matrix. The original high strength resins proved to be brittle and susceptible to delamination. Consequently, buckling and out-of-plane loads caused rapid failure. More recently developed resins

---

This thesis follows the style of the Journal of Composite Materials.

are tougher, and research continues as attempts are made to increase toughness without sacrificing stiffness and glass transition temperature.

When fibers are added to a ductile resin matrix, the brittle fibers decrease the zone of plastic deformation ahead of a crack propagating through the resin. Because such deformation is the primary mechanism of energy absorption in a brittle fiber - ductile matrix system, the presence of fibers decreases the overall delamination toughness (1). Varying the resin thickness between fibers should, consequently, change the fracture toughness for a given graphite/epoxy system with this combination of components.

Two composite systems, Hexcel T6C190/F155 and Hexcel T6T145/F185, have been investigated in an effort to quantify delamination fracture toughness for varying resin thicknesses. Testing has included both microscopic and macroscopic analysis, with the descriptions of such testing and the results presented here.

REVIEW OF LITERATURE

Investigation of delamination fracture toughness has involved the use of various specimen geometries. One such specimen is the double cantilever beam, shown in Figure 1. This was utilized by Vanderkley (2) and by Cohen (3) to calculate energy release rates for graphite/epoxy systems as a method of quantifying fracture toughness. Linear elastic fracture mechanics (3) is the method of data analysis implemented here in determining these energy release rates.

Study of transverse cracking in composites has been undertaken using compact tension specimens, seen in Figure 2. This type of specimen was also used previously by Vanderkley (2) and Cohen (3). Data analysis in this study has involved the use of fracture mechanics, including data relating compliance and crack length as determined by Saxena and Hudak (4). In addition, non-linear elastic J-integral calculations have been performed to determine energy release rates for the compact tension specimens based on ASTM-E813.

Previous observation of composite failure using a scanning electron microscope has revealed mechanisms responsible for energy release in a brittle fiber -

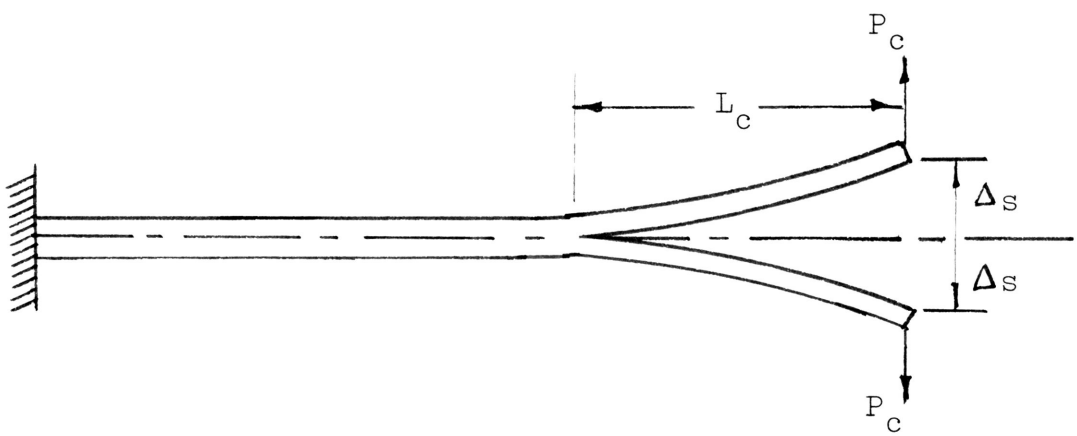


Figure 1. Double cantilever beam specimen subjected to mode I loading

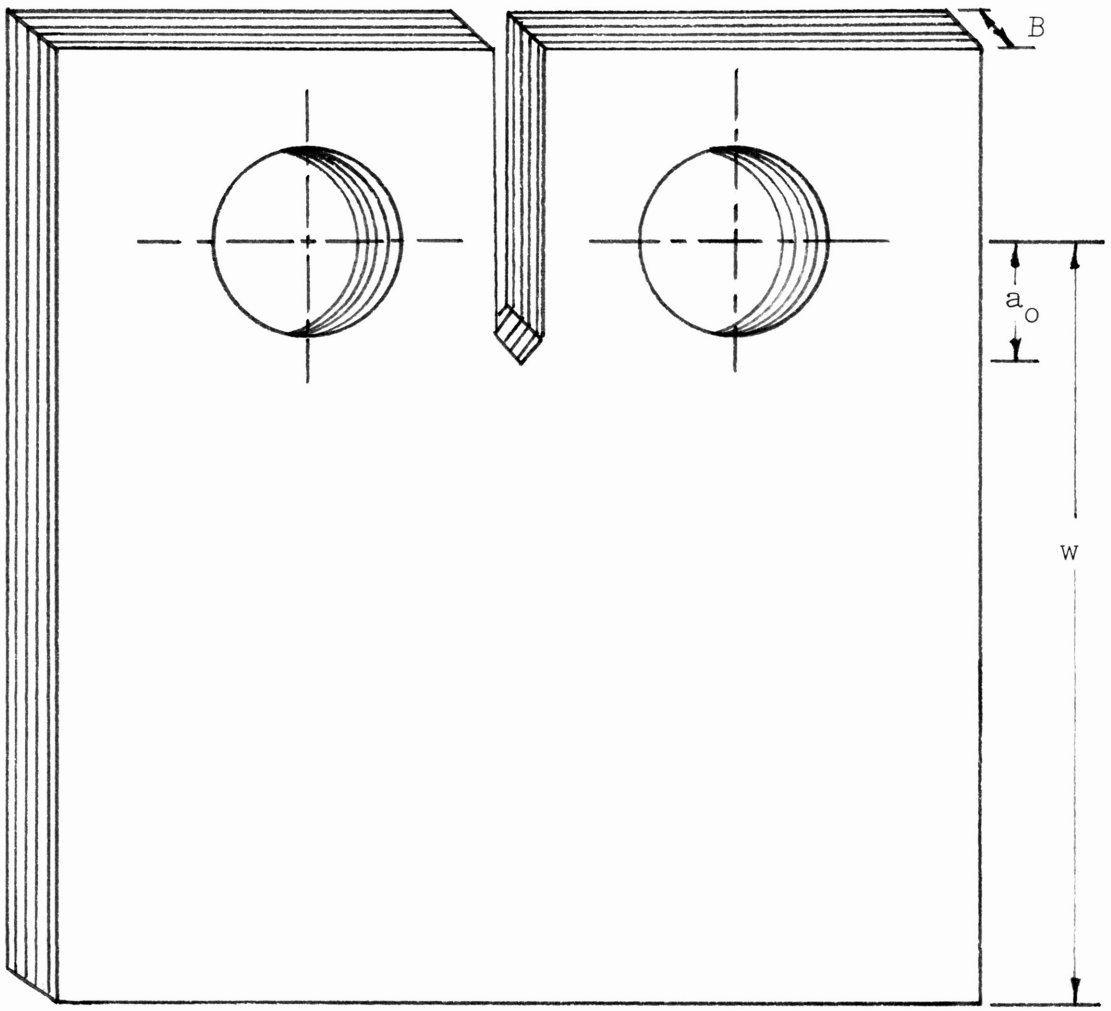


Figure 2. Compact tension specimen with critical dimensions shown.

ductile matrix system (1). Examination of specimen surfaces after fracture has allowed for identification of artifacts of these mechanisms. This has made possible a more complete understanding of the micro-mechanistic basis for the observed critical energy release rates obtained through data analysis.

Fatigue precracking of the compact tension specimens was attempted as a method of introducing a sharp crack. The approach used by earlier investigators, namely, driving a razor blade into the blunt machined-in notch in the specimen, was considered to give a crack of uncertain acuity. A fatigue crack is very sharp and allows the measurement of a true minimum G_{1c} value.* No references to fatigue precracking of composite materials were found in the literature reviewed in preparation for this study.

*It should be noted that "G_{1c}" values measured using specimens with insufficiently sharp notches are not valid G_{1c} values, even though they may be reported to be.

ANALYTICAL PROCEDURE

In order to measure and compare the ability of materials to resist crack propagation, a consistent method must be adopted. For this study, energy release rates were calculated for both composite systems. For elastic materials, the critical energy release rate, G_c, is a measure of the crack tip work required to cause crack extension per unit area of new surface created (3). For materials which experience both elastic and plastic deformation, the energy release rate J_c may be used for comparison purposes. These values are calculated in different ways but have equivalent definitions and are indications of the same material behavior.

Double Cantilever Beam Analysis

The double cantilever beam (DCB) specimens were subjected to mode I loading, with symmetrical tensile loads applied to both sides of the beam. This is a "worst case" model of delamination, and allows easy determination of G_{1c} values (2).

The equation for the release rate is as follows:

$$G_{1c} = \frac{P_c^2 L_c^2}{BEI}$$

where P_c is critical load, L_c is crack length, B is specimen width, E is modulus, and I is the moment of inertia. Due to the fact that the modulus may not be known exactly, it may be calculated using the formula

$$EI = \frac{2PL_c^3}{3\Delta_s}$$

where Δ_s is the total beam tip displacement at the load line. All these values are obtained from the load-displacement data generated during testing.

This analysis assumes linear elastic material behavior, which is a good approximation for a unidirectional composite with load-bearing linear elastic graphite fibers (3). Large rotations of the beam centerline, corresponding to large beam tip displacements, may result in significant geometric nonlinearities, resulting in serious discrepancies between linear and nonlinear analysis. Linear analysis has been shown to be appropriate (5) when

$$\frac{\Delta_s}{2L_c} > 0.4$$

The linear theory was found to be appropriate for all DCB speciment of both composite systems tested in this study.

Compact Tension Specimen Analysis

In order to take into account any nonlinear behavior of the material due to viscoelastic properties or plastic deformation, J-integral analysis was used for the compact tension specimens according to ASTM-E813. The equation for critical energy release rate is as follows:

$$J_{1c} = \frac{2.2 \int Pd\delta}{B(w-a_o)}$$

where $\int Pd\delta$ is the area under the load-displacement curve to the point where the crack begins to grow, a_o is the original crack length, and w is the specimen width.

Because it was difficult to ascertain the exact moment of crack growth, an upper and lower bound for J_{1c} were calculated for each compact tension specimen. The load for the lower bound was determined using a 5% secant offset to find a point with no more than 2% crack extension. The load for the upper bound was chosen to be the maximum load on the test data plot. These boundaries ensured that the actual moment of crack extension was bracketed and so the J_{1c} value for the material was also bounded.

EXPERIMENTAL PROCEDURE

This study involved the use of two Hexcel composite systems: T6C190/F155 and T6T145/F185. Two approaches for investigating effects of resin content on fracture toughness were taken. The first approach involved variation of resin content through manufacturing in the F155 system. The second approach, used with the F185 material, examined three degrees of constraint on resin deformation produced by varying the thickness of the resin rich-region through which the crack propagated. This variation was accomplished using two specimen geometries and using neat resin and fiber reinforced resin.

Preparation and Testing of F155

The initial step in preparing the F155 specimens was fabrication of the composite panels. Preimpregnated tape was supplied by the manufacturer in 12 in. wide rolls. Sheets of 1 ft^2 area were cut and layered so that the fibers were unidirectional. Sixteen sheets were layered for each of two composite panels.

Variation in manufacturing, resulting in variation of resin content in the specimens, involved a process

known as bleeding. One F155 panel was bled by placing glass cloth on either side of the material during curing. In this way, excess resin was absorbed, leaving an approximately 10% higher relative fiber volume in the sample.

Included along one side of both composite panels was a 1 in. wide strip of teflon. This was inserted between the eighth and ninth plies, on the side of the specimen to which the fibers ran perpendicular. This teflon insert produced a sharp crack from which the delamination of the DCB specimens could be initiated.

The cure cycle for the F155 was the one recommended by Hexcel for this material, as seen in Figure 3. Curing was done in an air cavity press with the temperature, pressure, and vacuum controlled by a Micrion microprocessor.

Following curing, the panels were cut into DCB specimens. These consisted of four bled and four unbled strips, 1 in. wide and 9 in. long. In addition, six 1 in. by $\frac{1}{2}$ in. samples were cut for examination by SEM. All cutting was done using a diamond saw.

The SEM samples were mounted in a room-temperature-set epoxy, polished with 0.3 micron alumina paste,

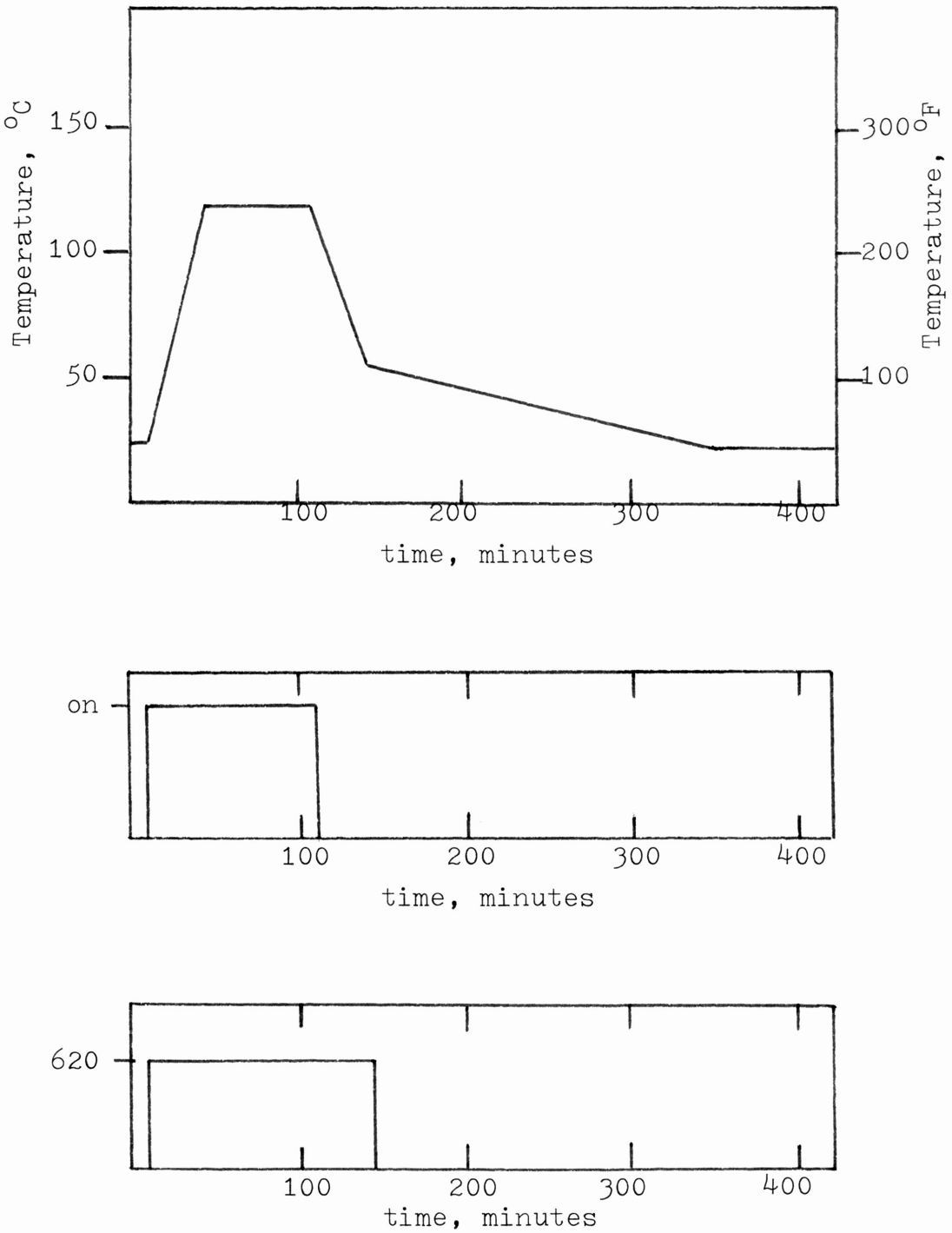


Figure 3. Cure cycle for Hexcel T6C190/F155 graphite/epoxy composite

and coated with a 200Å thick layer of gold-palladium. This coating is necessary to conduct unreflected electrons from the surface and prevent charging of the specimen. Cross sections of both the bled and unbled panels, showing relative fiber and resin distribution, were photographed in the SEM. These photos are shown in Figures 4 and 5, where the fibers and resin-rich regions between plies are clearly evident. As was expected, the bled specimen showed a thinner resin-rich region and less resin within the plies.

Delamination of the DCB specimens was done using an MTS servo-hydraulic test machine. Figure 6 shows the fittings which were epoxied to the ends of the specimens. These allowed for mode I loading of the composite beams with minimal rotation of the specimen as the crack grew.

As mentioned previously, teflon strips were inserted in the composite panels before curing to introduce a crack. From this beginning flaw, the crack was extended by hand to a point where the initial crack length was measured; this point was slightly beyond the ends of the fittings on the specimen.

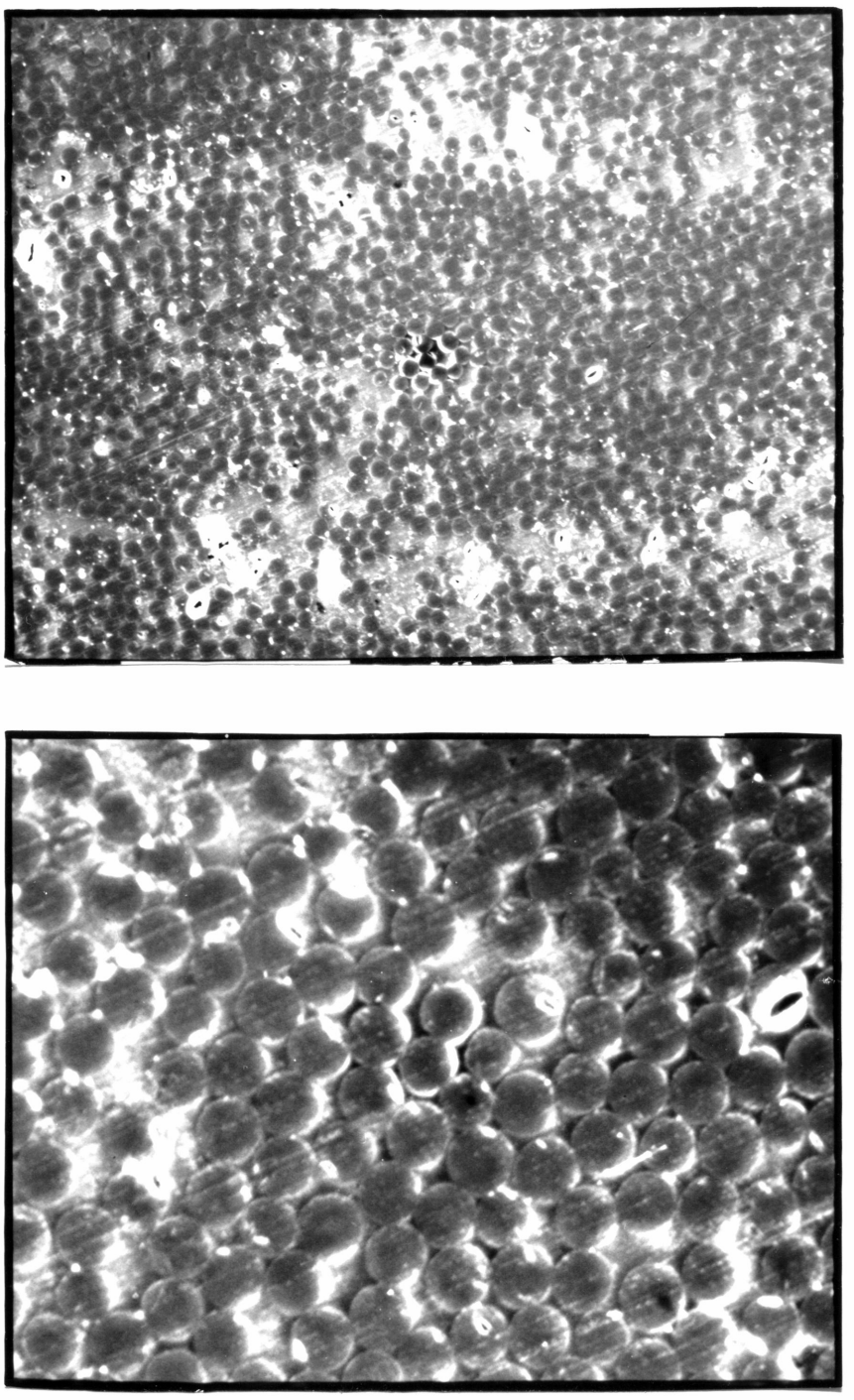


Figure 4. Bled F155 sample at 300X and 1000X

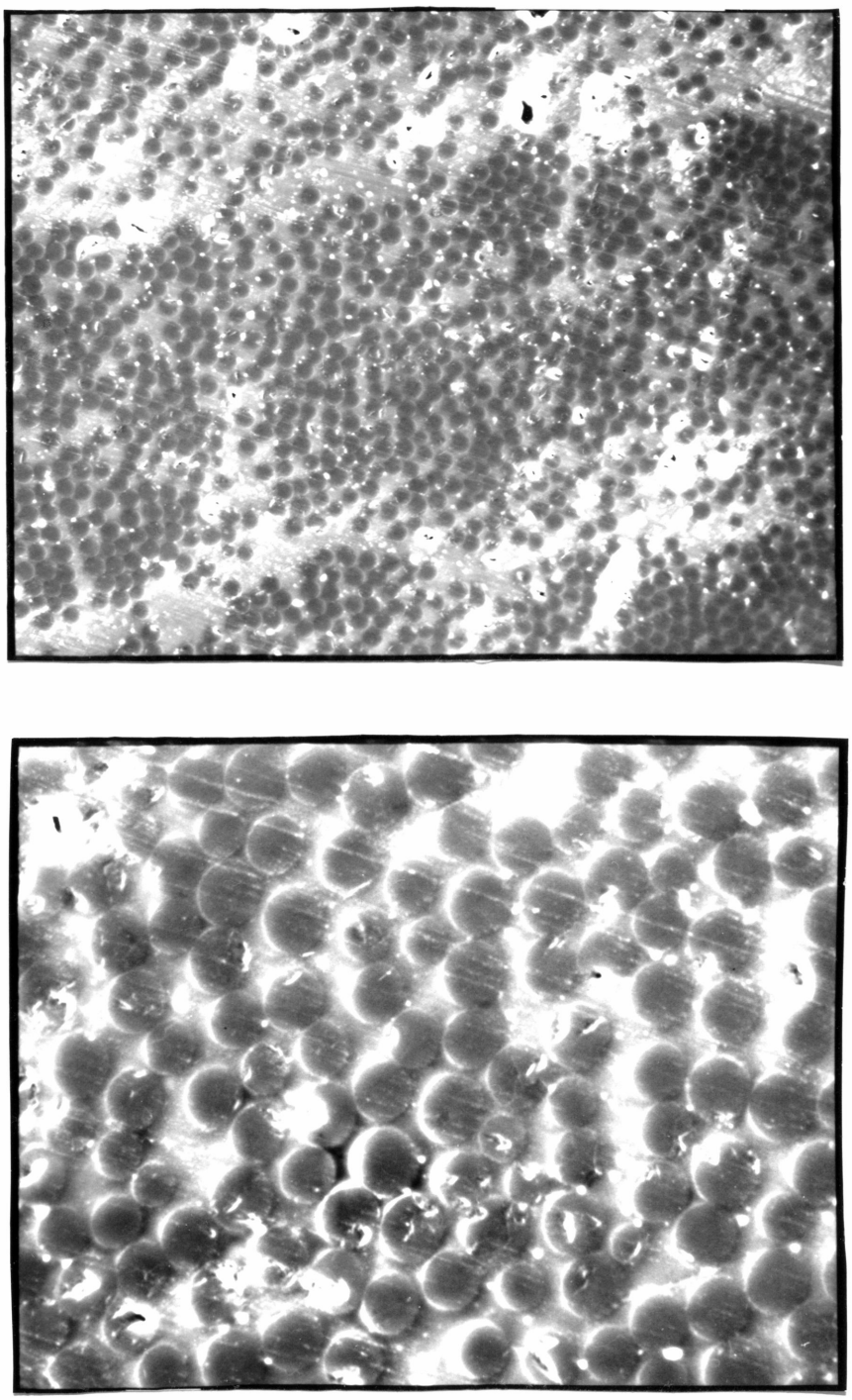


Figure 5. Unbled F155 sample at 300X and 1000X

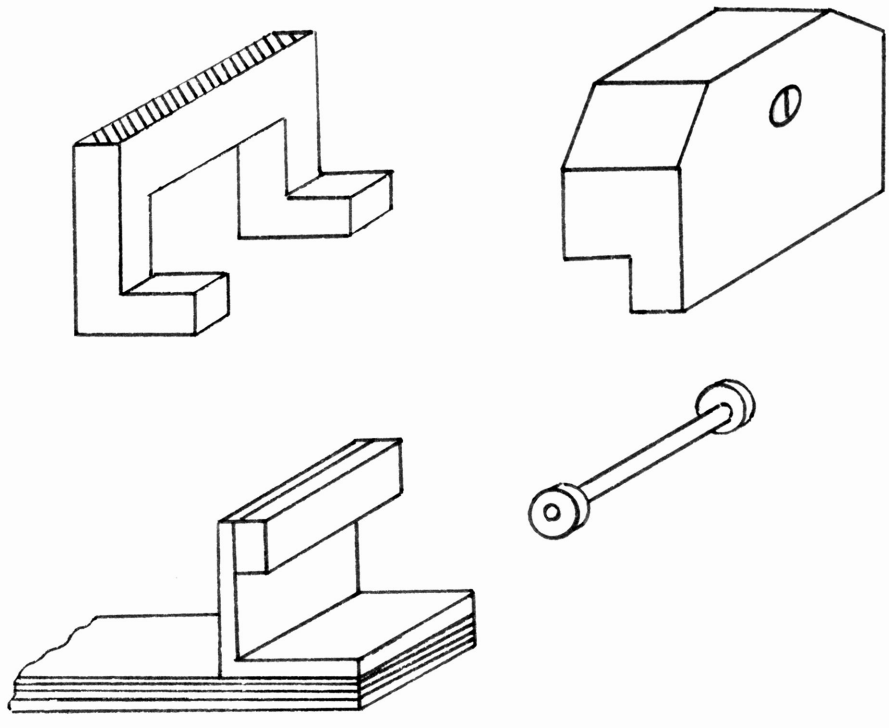


Figure 6. Exploded view of DCB fitting and attachments

During testing, the specimens were subjected to mode I loading at a uniform rate, while load and beam tip deflection were monitored. The appendix contains an example of typical DCB delamination data. Periodically, loading was reversed momentarily, giving a 10-20% unload, and crack length was measured visually on the specimen and recorded on the chart.

Preparation and Testing of F185

The F185 composite system differs from the F155 in its crosslink density and volume fraction of second phase additions. Both systems contain rubber particles as a second phase to incresse matrix deformation. The F155 material contains 6% rubber particles by volume, while the F185 contains 14% rubber particles (3).

While the F155 samples were manufactured from supplied pre-preg material, the F185 composite panels used in this study were received in a cured state from Hexcel. In addition, cast blocks of the neat resin without fibers were made available. These panels and blocks were machined into DCB specimens (composite material only) and compact tension specimens (composite and neat resin). The delamination specimens were

10 in. long and 1 in. wide; the compact tension specimens were 2.25 in. long, 2.4 in. wide, and were 0.53 in. thick for the composite and 0.47 in. thick for the neat resin.

The testing assembly and procedure for the F185 DCB specimens was the same as was used for the F155 material. Teflon had been inserted in the samples before curing, so the crack initiation process was the same for both composite systems. The same fixtures for mode I loading were attached to the specimens, and the collected data measured the same parameters. The purpose of the delamination specimens was to measure the energy release rate for the material as a crack propagated through the resin-rich region between plies.

As mentioned previously, for the F185 system, three degrees of constraint on the resin were tested and energy release rates compared. The DCB delamination tests represented one condition; the compact tension tests on specimens with and without fibers respesented the other two. Testing performed on the composite CT specimen represented a "worst case," with transverse crack propagation occurring within plies

through a region of minimum resin. The CT specimen made of neat epoxy exemplified the other extreme, with no fibers to constrain the resin and limit deformation during cracking.

In order to measure the crack initiation and crack propagation energies, a sharp crack was needed in the compact tension specimens. A blunt notch instead of a sharp crack would give an overestimate of resistance to crack extension. A sharp crack was introduced through fatigue precracking. The neat material was subjected to tensile-tensile loading at 3 Hz, while the composite underwent 10 Hz cycling under tensile-tensile loads. These fatigue cracks were allowed to grow until $a/w = 0.5$, where w is the width of the specimen from the load line to the far edge, and a is the length of the crack, also measured from the load line. Once a sharp crack was present, the compact tension specimens were broken by applying a load at a constaint rate and measuring load vs. crack tip opening. The appendix contains a sample of the data collected. An LVDT attached to the surface of the specimen monitored crack tip opening, and its distance from the load line was noted in order to

correct for that variation in opening size during data analysis.

Following fracture of the neat resin and composite specimens, the samples were sectioned and inspected in the SEM. Both the fracture surfaces were examined, and the sides of the neat material along the crack were viewed. These photographs and their significance in light of caluclated energy release rates will be discussed later in this report.

RESULTS

Data Analysis for F155

Using the equations cited previously, critical energy release rates were calculated for the DCB specimens tested. For each specimen, an average value of G_{1c} was found. These values, along with an overall average G_{1c} for the bled and unbled samples, are shown in Table 1. All data met the criterion for linear elasticity such that $\Delta_s/(2L_c) < 0.4$ (5).

TABLE 1

Bled Composite	Unbled Composite
Sample 1: G_{1c} = 561 J/m^2	Sample 1: G_{1c} = 1008 J/m^2
Sample 2: G_{1c} = 546 J/m^2	Sample 2: G_{1c} = 1008 J/m^2
Sample 3: G_{1c} = 599 J/m^2	Sample 3: G_{1c} = 947 J/m^2
Sample 4: G_{1c} = 557 J/m^2	Sample 4: G_{1c} = 1010 J/m^2
Average G_{1c} = 565.75 J/m^2	Average G_{1c} = 993.25 J/m^2

Data Analysis for F185

The delamination data for the DCB specimens was analyzed in the same manner as for the F155 and critical energy release rates were calculated. This data also met the linear elasticity criterion (5).

Values for critical energy release rates, J_{1c}, for the compact tension specimens were calculated as

noted earlier. The integration term in the J_{1c} equation was evaluated by graphical integration to determine the area beneath the load-displacement curves for the composite and the neat resin specimens. The original crack length values were determined through examination of the fracture surface following failure and measuring the extent of the fatigue precracking. All crack lengths and displacements are measured relative to the load line. Recall that the upper and lower bounds for J_{1c} represent an interval in which the crack began to propagate. Table 2 summarizes the DCB and compact tension results.

TABLE 2

	J_{1c},lower	J_{1c},upper
Composite CT	1628.1 J/m^2	3651.04 J/m^2
Neat Resin CT	4582.5 J/m^2	8134.65 J/m^2

Mode I Delamination

Sample 1: G_{1c} = 1835.73 J/m^2

Sample 2: G_{1c} = 1939.68 J/m^2

Sample 3: G_{1c} = 1875.11 J/m^2

Sample 4: G_{1c} = 1932.06 J/m^2

Average G_{1c} = 1895.65 J/m^2

DISCUSSION

Both the results of the F155 and F185 testing support the hypothesis that in a brittle fiber - ductile resin system, deformation of the matrix is a primary energy absorption mechanism during crack propagation. A composite with more resin should, therefore, exhibit a higher energy release rate and a correspondingly high fracture toughness, and this is what the results indicate.

For the F155 system, the bled delamination specimens absorbed less energy per unit crack extension than did the unbled samples. Not only does this show that the thickness of the resin-rich region between plies is significant, but also that manufacturing variations may produce dramatic changes in material properties.

The three degrees of constraint in the F185 system also demonstrated the significance of the amount of ductile resin with relation to fracture toughness. The neat epoxy, free of any constraint imposed by graphite fibers, had the largest energy release rate. The double cantilever beam specimens, in which the crack was propagated through the resin-rich region

between plies, had a significantly smaller energy release rate. However, the value of J_{1c} for the composite compact tension specimen was smaller yet, where the crack was grown in a region between fibers with the least amount of resin available to deform and dissipate energy. It should be noted that the wide range between upper and lower J_{1c} values for the compact tension specimens is probably misleading; in all likelihood, the lower values are the better approximations of the actual critical energy release rates for the materials.

Photographs taken in the SEM reveal evidence to support the analytical results of testing for the F185 material. Figure 7 shows fibers on the fracture surface of the transverse delamination specimen. Although the F185 matrix is composed of a ductile epoxy resin, the fiber surfaces are coated with resin that appears to have fractured in a brittle fashion. The constraint imposed on the thin resin layer between fibers was sufficient to eliminate most plastic deformation, resulting in a low fracture toughness measurement.

Compare this behavior to that illustrated in Figure 8, which shows a region of crack growth in the

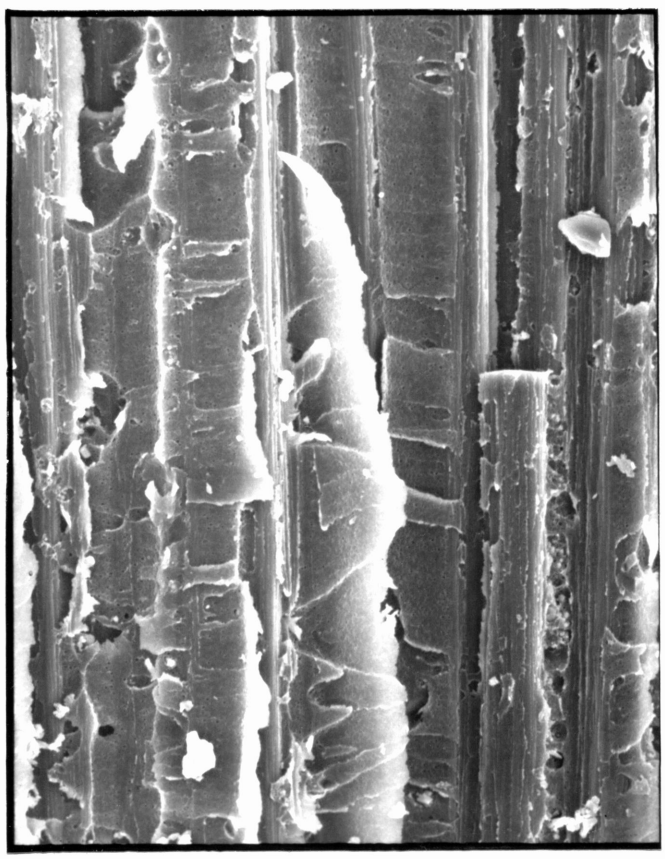


Figure 7. F185 composite showing fracture surface of transverse delamination specimen at 1000X

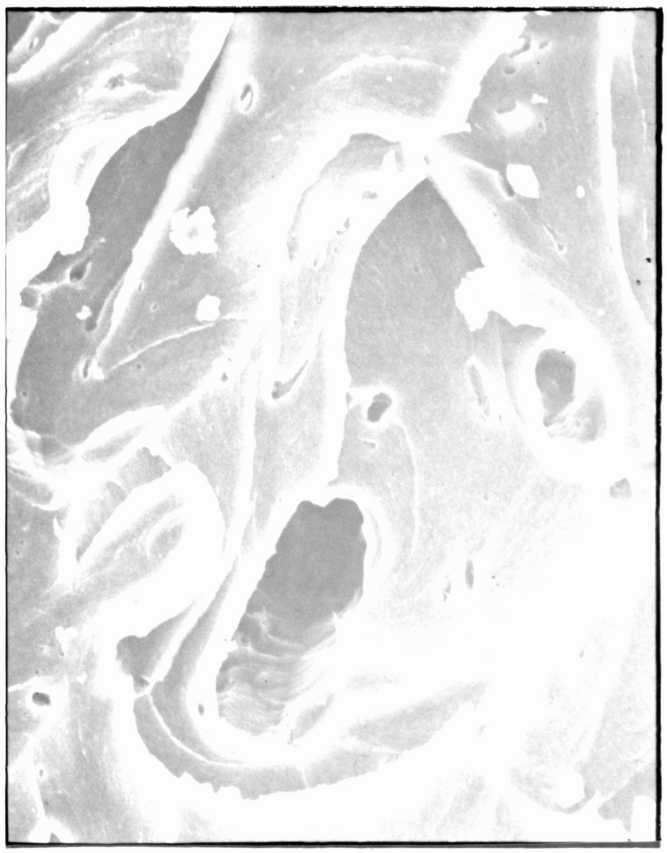


Figure 8. F185 neat resin at 1000X showing plastic deformation produced during ductile fracture

neat resin specimen. Note the extreme plastic deformation which resulted in the high energy release rate values for this material. The holes in the surface correspond to regions of deformation around rubber particles. This may be confirmed by examining Figure 9, a portion of the neat specimen broken in a brittle manner at a very low temperature. The absence of holes in the surface indicates that these are not voids due to manufacturing, but were produced during fracture.

Using fatigue precracking to initiate a sharp crack in the compact tension specimens produced the question of whether striations would appear on the fracture surface. Figure 10 shows fatigue regions of both the composite sample and the neat resin. The neat material shows no indication of striations; however, the composite surface bears marks that resemble striations. Based on the frequency of fatigue loading and the length of time the material was fatigued, there are 3.7-5.5 fewer marks per micron on the fracture surface that cycles per micron of crack growth. However, since the crack does not necessarily advance for every cycle, fewer marks than cycles does not rule out the possibility that these are striations.

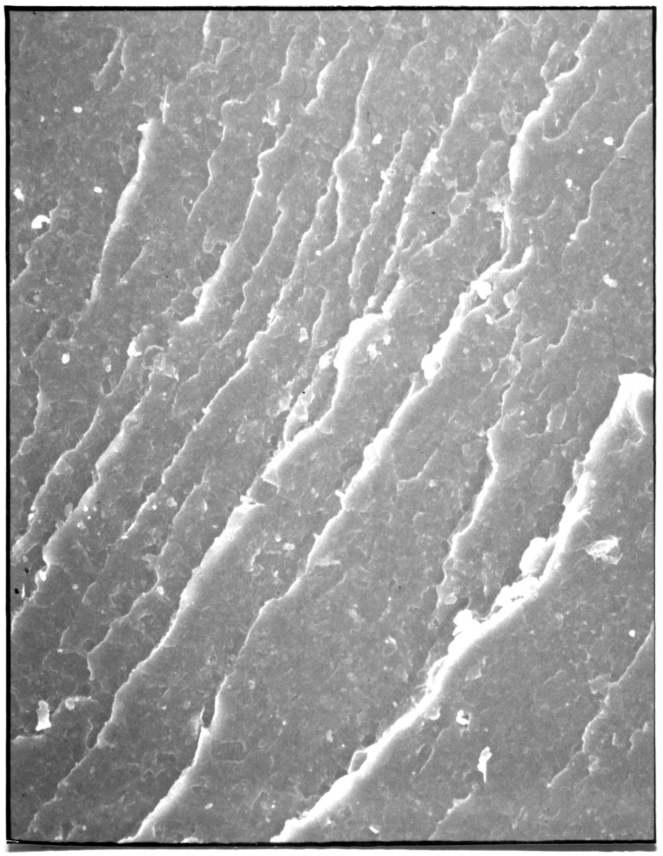


Figure 9. F185 resin at 1000X showing absence of voids in brittle fracture region

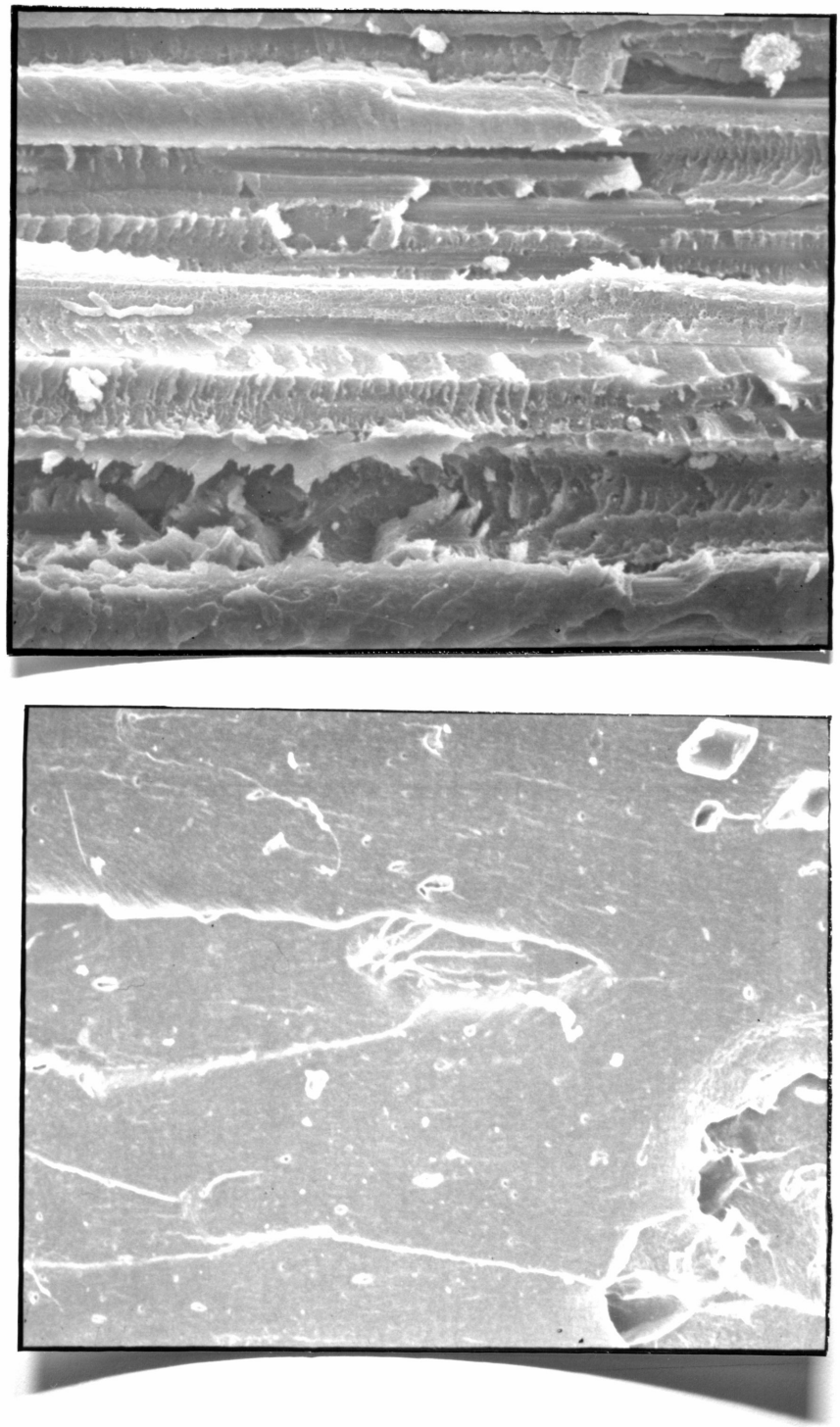


Figure 10. F185 composite and neat resin specimens showing fatigue pre-cracked region at 1000X

The neat resin sample turned lighter in color in the crack tip region during fracture. In an effort to explain this, the side of the specimen was examined. Figure 11 shows the edge of the sample below the fracture surface. A thin scale, produced when the liquid epoxy was poured into the mold and quickly cooled during casting, has cracked and fallen away in places. It was noted that this cracking extended only a short was down the specimen, accounting for the change in surface appearance in the crack region.

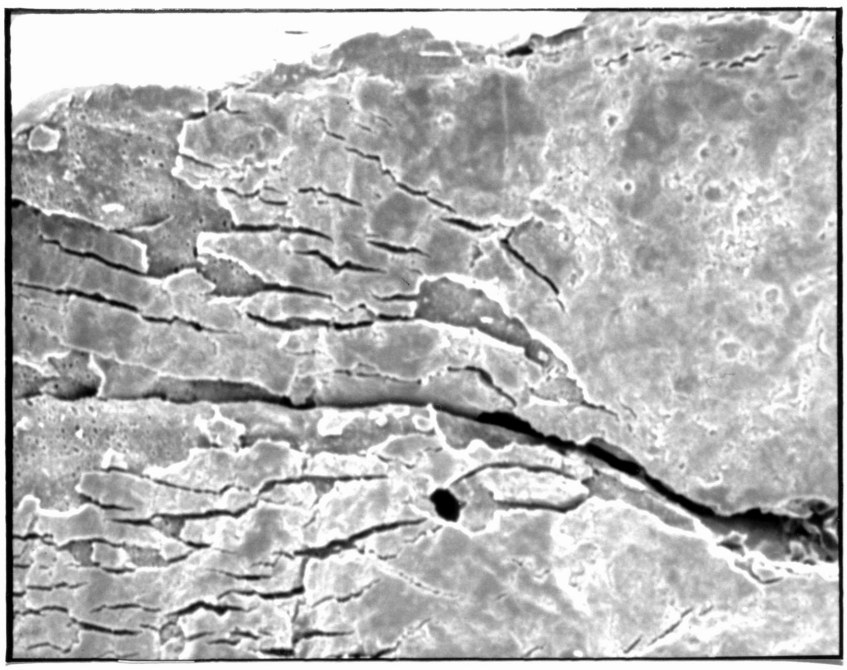


Figure 11. Edge of F185 neat resin specimen at 1000X, showing scale cracking below fracture surface

CONCLUSIONS

It may be concluded that energy absorption during crack propagation is directly related to the amount of resin in the crack tip region for a brittle fiber - ductile matrix composite material. Whether the amount of resin between fibers was controlled through manufacturing or by varying the geometry of the test specimen, samples with a larger resin-rich region between plies exhibited higher G_{1c} or J_{1c} values. The constraint imposed by the fibers reduces the size of the plastic zone ahead of the crack tip and limits the energy absorbed during fracture.

RECOMMENDATIONS

Two difficulties arose as a result of the imprecise monitoring of crack growth in the CT specimens: not being able to determine exact J_{1c} values, and not being able to distinguish noncontinuous crack growth during fatigue loading. A method of monitoring exact crack position is available in the form of a Krak gauge. This thin metal gauge is bonded to the surface of the specimen and cracks as the specimen fractures. The resistance of the Krak gauge varies linearly with the length of the crack. Using one of these, the exact moment of crack growth could be noted to aid in J_{1c} calculations. Also, the presence of striations could be confirmed by noting whether crack growth during fatigue loading is continuous or noncontinuous.

The presence of scale on the surface of the neat resin specimen prevented close examination of the behavior of the resin near the crack tip. It is desirable to investigate the size of the plastic zone ahead of the crack tip, a task which proves impossible with the brittle scale concealing the fracture artifacts. The surface of subsequent neat resin samples should be ground in order to remove the scale and improve visibility.

REFERENCES

1. Bradley, W.L. and Cohen, R.N., "Matrix Deformation and Fracture in Graphite Reinforced Epoxies," presented at ASTM Symposium on Delamination and Debonding of Materials, Pittsburgh, Pennsylvania, November 1983.

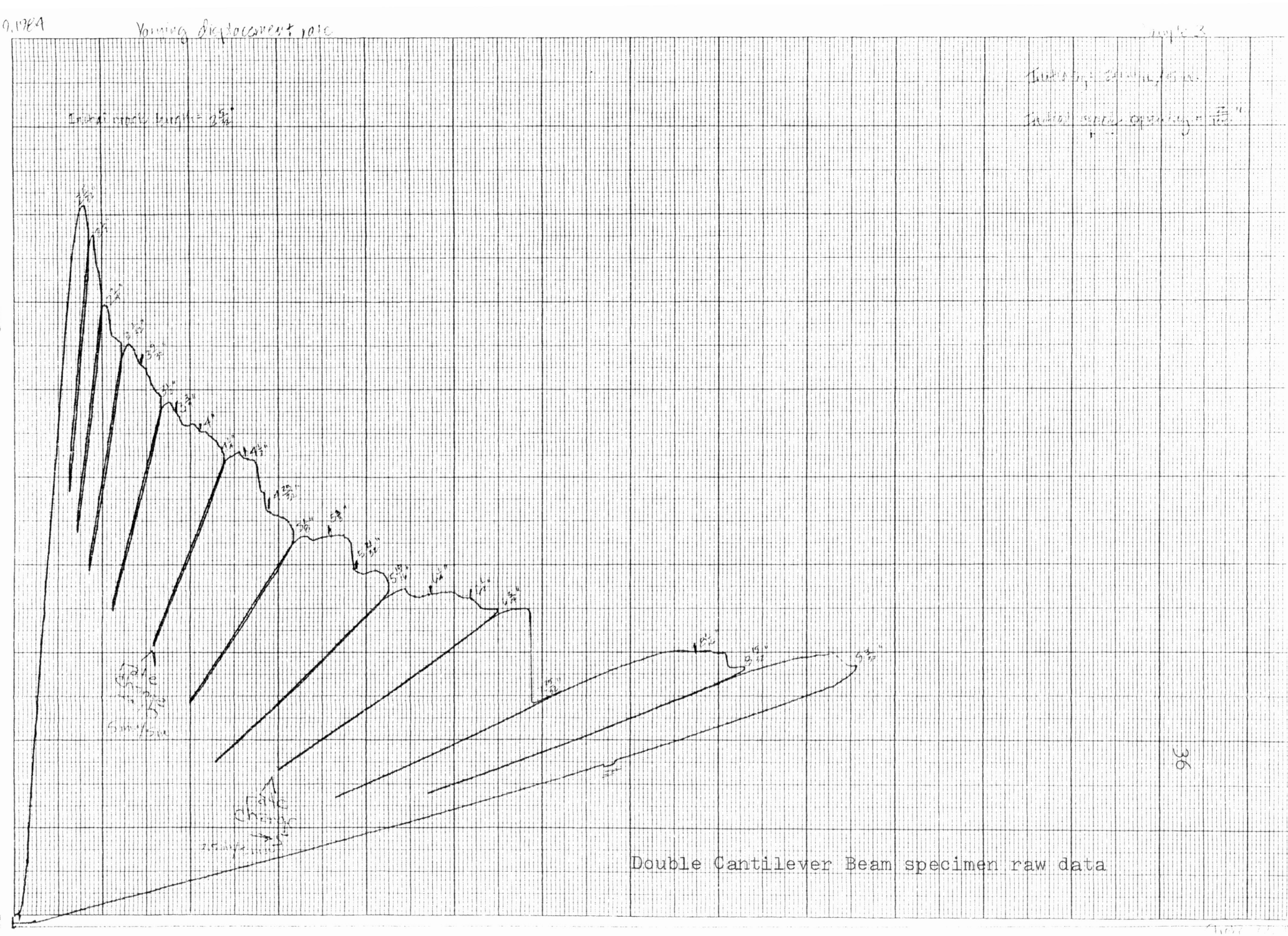
2. Vanderkley, Peter S., "Mode I-Mode II Delamination Fracture Toughness of a Unidirectional Graphite/Epoxy Composite," Master's Thesis, Texas A&M University, College Station, Texas, December 1981.

3. Cohen, Ronald N., "Effect of Resin Toughness on Fracture Behavior of Graphite/Epoxy Composites," Master's Thesis, Texas A&M University, College Station, Texas, December 1982.

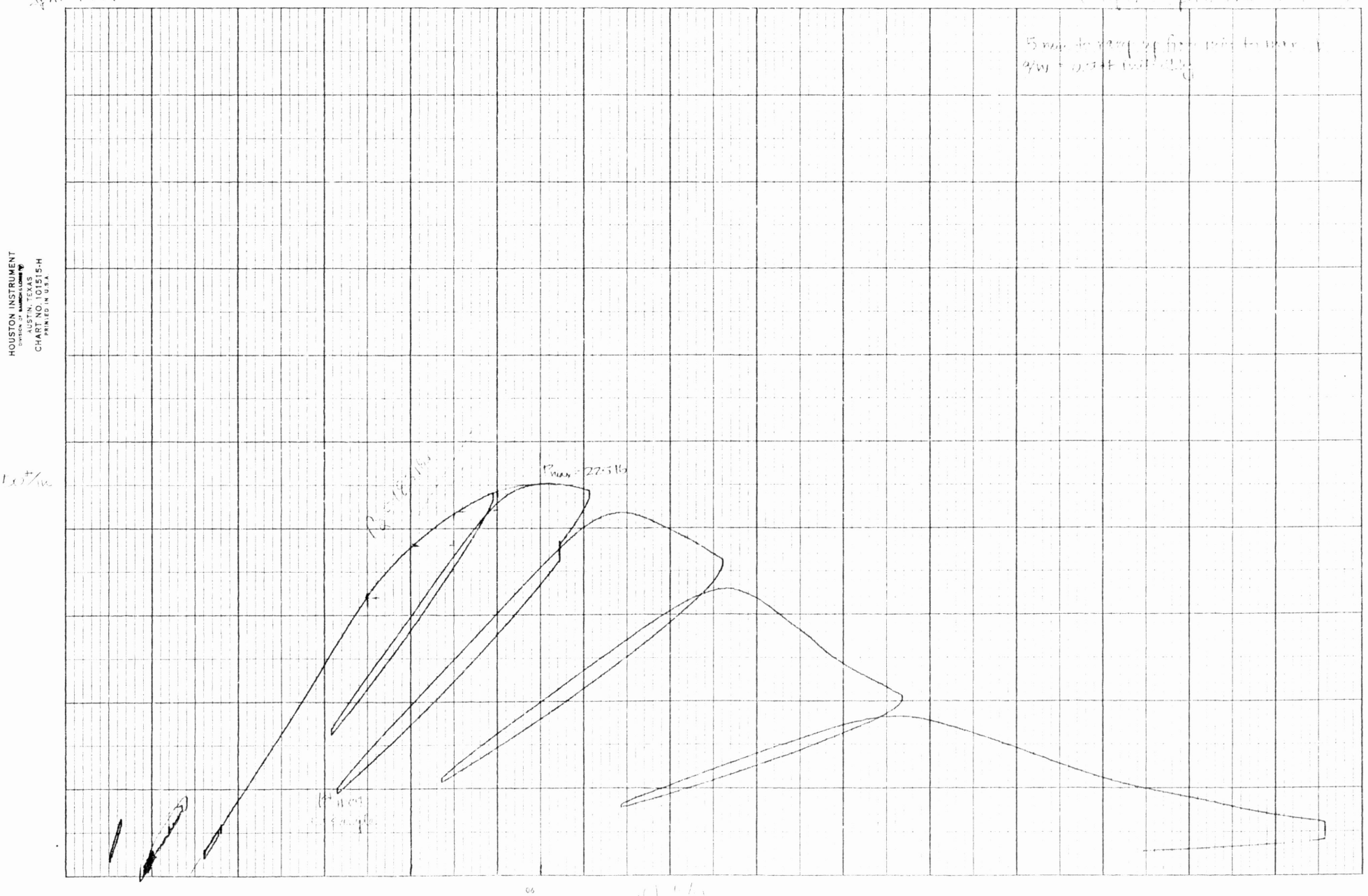
4. Saxena, Ashok and Hudak, S.J., Jr., "Review and Extension of Compliance Information for Common Crack Growth Specimens," Scientific Paper 77-9E7-AFCGR-P1, Westinghouse R&D Center, Pittsburgh, Pennsylvania, May 1977.

5. Devitt, D.F., Schapery, R.A., and Bradley, W.L., "A Method for Determining the Mode I Delamination Fracture Toughness of Elastic and Viscoelastic Composite Materials," Texas A&M University Report MM 3724-80-6, June 1980.

APPENDIX



Double Cantilever Beam specimen raw data



Compact tension specimen raw data