Progress Report
(March 1984-February 1985)

(NASA-CR-175998) STRUCTURAL CHARACTERISTICS OF HIGH TEMPERATURE COMPOSITES Progress Report, Mar. 1984 - Feb. 1985 (Massachusetts Inst. of Tech.) 23 p HC A02/MF A01 CSCL 11D
N85-30037
Unclas
G3/24 29890

NASA Grant NAG 3-377

Structural Characteristics of High Temperature Composites

by

J.F. Mandell

Department of Materials Science and Engineering
Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

Summary

Progress during this period is described in detail in the attached paper presented at the 9th Annual Conference on Composites and Advanced Ceramic Materials, American Ceramic Society, January 20-23, 1985, Cocoa Beach, Florida. The following is a brief summary of that paper and several additional items.

1. A tensile test method has been developed which should give tensile and simulated shear (±45°) data for fiber composites up to 1000°C.

2. Longitudinal and some transverse stress-strain data have been obtained for a glass matrix/Nicalon fiber system up to the matrix limiting temperature of 600°C. This demonstrates the functioning of the test method and the high temperature test facility which has been established on this grant.

3. Transverse and longitudinal compression tests have been run, mostly in an end loaded configuration. A more satisfactory compression test is still required, and is under development.

4. Although there are no major obstacles with this project, progress has been slower than expected. In part, this stems from inherent aspects of high temperature testing which were not anticipated, while delays have also occurred in equipment acquisition and in the supply of materials. High temperature test development is slower than expected due to difficulties in the machining of both grips and specimens. Many grip modifications could only be accomplished by specialty companies outside of M.I.T. with long, unanticipated delays at each modification. While specimens have been machined at M.I.T., the slow grinding required for the complex specimen

shape often requires one day per specimen. Similarly, the long heating and cooling cycles of the testing equipment limit testing to one or two specimens per day.

The most significant problem has been in the supply of materials. The number of samples supplied by Corning Glass Works has fallen short of original estimates by a factor of 3-6 times. The largest batch of material received could not be tested because it proved to be too thin for the details of our tensile test method. The materials supply problem may now be solved by interest generated from presentation of the attached paper. Corning Glass Works now has us scheduled for an increased supply of samples, including BMAS matrix/Nicalon samples which will allow testing to 1000°C. United Technologies Research Center has agreed to supply LAS matrix/Nicalon samples also useful to above 1000°C, and Pratt and Whitney may supply graphite fiber/glass matrix materials. Additional ceramic matrix/SiC filament (AVCO) composite may be available from NASA-Lewis.

5. Testing for off-axis, particularly transverse properties, has been a problem due to flaws introduced during molding and machining. This appears to have limited the study of off-axis properties in other studies using other material sources as well. While we will continue to attempt to measure off-axis properties on unidirectional material, we also plan to measure transverse ply cracking in 0/90 laminates and cracking in ±45° laminates, and to deduce the in-situ off-axis properties from laminate data. This may provide more representative off-axis, particularly transverse, data for application to typical laminates. We are scheduled to receive some 0/90 material from Corning Glass Works within one month.

6. Personnel and Publications: The only publication from this project is attached. Personnel have not changed since the

last report except for the addition of an undergraduate assistant. One student (Mr. B. Edwards) is expected to finish his M.S. thesis by June, 1985. The second student (Mr. Dodd Grande) should finish his doctoral thesis within a year. A new masters level student is scheduled to join the project in June, 1985.

7. Future plans: We anticipate that the project will still be somewhat behind schedule by the end of the current year (9/30/85) for the reasons already given. We do expect to accomplish most of the proposed scope of the project by that date, but with a reduced quantity of data.

TEST METHOD DEVELOPMENT FOR STRUCTURAL CHARACTERIZATION OF FIBER COMPOSITES AT HIGH TEMPERATURES

by

J.F. Mandell, D.H. Grande and B. Edwards
Department of Materials Science and Engineering
Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

Summary

Test methods used for structural characterization of polymer matrix composites can be applied to glass and ceramic matrix composites only at low temperatures. New test methods are required for tensile, compressive and shear properties of fiber composites at high temperatures. A tensile test which should be useful to at least 1000°C has been developed and used to characterize the properties of a Nicalon®/glass composite up to the matrix limiting temperature of 600°C. Longitudinal and transverse unidirectional composite data are presented and discussed.

Prepared for Ceramic Engineering and Science; paper originally presented at 9th Annual Conference on Composites and Advanced Ceramic Materials, American Ceramic Society, Jan. 20-23, 1985, Cocoa Beach, Florida.

INTRODUCTION

The early stages of development of structural glass and ceramic matrix fiber reinforced composites have been guided by feedback from one primary source of mechanical property data: flexural tests on unidirectional composites stressed in the longitudinal (fiber) direction. Flexural tests have two advantages for high temperatures: (1) loads are introduced by local compression, and (2) strains can be calculated from the displacement of the load points. However, the only meaningful data available from flexural tests are modulus (a combination of tension and compression), tensile stress and strain at first matrix cracking, shear strength if shear failures occur with short beams, and a general idea of qualitative stress-strain behavior from the shape of the curve beyond first matrix cracking. Behavior after first cracking (which may occur at 10-20% of the total failure strain) is greatly complicated by the stress gradient and shifting neutral axis of the flexural specimen. The data available from flexural tests are clearly not adequate for design and analysis, but neither are they adequate for materials research and development beyond the early stages. It is essential to consider the behavior of composites in various orientations under tensile, shear and compressive stresses, as well as combined stresses. It is also essential to study the progression of damage generation and failure in multidirectional laminates typical of potential applications; flexural tests are even less useful with multidirectional laminates due to the complications of property variations through the thickness combined with the inherent strain gradient from the test.

Test Methods for Polymer Matrix Composites

To obtain acceptable properties in various (planar) directions, advanced fiber composites are usually fabricated as laminate structures composed of individual unidirectional plies oriented in selected directions to give the desired balance of properties for particular applications. The behavior of the composite is rationalized by a set of properties for each (usually identical) ply combined with a suitable analysis to predict laminate behavior from the ply properties [1]. The minimum ply properties required for laminate design and analysis are the longitudinal and transverse tensile and compressive moduli and strengths, Poisson's ratios, and the in-plane shear modulus and strength. Thermomechanical analysis of laminates is also commonly required, so that the coefficients of thermal expansion in the longitudinal and transverse directions are included as ply properties, both for prediction of the laminate expansion coefficients in various directions and the calculation of thermal residual stresses within each ply of the laminate. Thermal residual stresses are often a significant fraction of the polymer composite ply transverse strength, particularly for high processing temperature systems.

The extreme strength anisotropy and planar (sheet) form of most unidirectional fiber composites have complicated the development of appropriate test methods for the required ply properties. Dumbbell-shaped specimens are often used for tensile properties of polymers and multidirectional composites [2], but unidirectional longitudinal composites split parallel to the fibers at the specimen shoulder, shifting the failure to within the grip area where the complex stress state renders the results invalid. The dumbbell shape can often be used for transverse tensile properties. Most ply properties for advanced polymer matrix composites are now obtained with straight-sided strips of material to which tapered end tabs are bonded with a suitable adhesive; typical test methods are described in Refs. 3 and 4. Gage-section tensile failures can be obtained in this manner with most unidirectional composites.

Compression tests present greater problems due to the variety of possible failure modes [5], which include elastic specimen buckling, successive delamination and buckling of surface plies, end brooming and axial splitting, as well as what might be termed material compressive failures which may involve microscale modes of shear, fiber buckling, or fiber failure. The mode of failure often changes when the specimen size or test method is changed, as from compressive end loading to shear loading through tabs, and depending on whether constraint against buckling is applied along the gage section [4]. Analogous mode and srength variations may be expected in applications, depending on geometry, buckling constraint and load introduction. Thus, a single compression strength value may have little general meaning in composites. Currently, the most important compression tests [4] for establishing ply properties in polymer composites introduce the load through shear using bonded tabs much like tabbed tensile coupons; lateral buckling constraint is usually provided.

In-plane unidirectional shear properties and interlaminar shear properties are also difficult to determine, and a number of test methods are available [4,6]. Since most composites are not readily available as rods or tubes, direct torsion tests are usually not practical. Other tests, whether off-axis tension, rail shear or Iosepescu [6] provide reliable values of shear modulus, but differing levels of uncertainty for the complete shear stress-strain curve to failure, which is usually nonlinear.

The shear strength for unidirectional material and laminates is frequently determined with the short beam shear test [4]; when failure is in the proper mode, this test gives results useful for materials development and for comparisons between materials, but not for design.

Other test methods such as three- and four-point flexure [4], impact [7], and fracture toughness [8] are also used with polymer matrix composites, but not for the purpose of obtaining basic data for design and analysis. Fracture toughness in particular has not been a generally useful test method or property for continuous fiber polymer matrix composites. Many composites are not notch sensitive even for notches of one cm or more in length; of those that are notch sensitive, some of the more brittle may be analyzed using linear elastic fracture mechanics for certain stress directions and ply arrangements [8]. However, the fracture toughness and degree of notch sensitivity are not a simple consequence of unidirectional ply properties, but depend primarily on the details of ply configuration (orientation of different plies and ply stacking sequence) for each particular laminate. In most cases the notch sensitivity for catastrophic failure is a consequence of the extent of the stable matrix/interface cracking (damage zone) which forms in areas of high stress concentration; the larger the stable cracking damage zone, the lower the overall notch sensitivity [8]. Since failure in most applications of composites does not occur by a single dominant crack, fracture mechanics has not been of great interest even though it may be applicable for certain narrow ranges of structure. Localized cracks parallel to the fibers of individual plies, and delaminations between plies are limited areas where fracture mechanics has been of greater interest [9].

Adaptation of Test Methods to High Temperatures

The most successful tensile, compressive and shear test methods for unidirectional polymer matrix composites are those with straight sided coupons loaded through adhesively bonded tabs. Strains are measured in the longitudinal and transverse directions with adhesively bonded strain gages. Unfortunately, none of these methods appear to be useful at the high temperatures of interest with glass/ceramic or other non-polymeric matrices. For example, at 1000°C there are no strain gages or suitable (tough) adhesives for bonding tabs or gages. Standard tabbed, strain gaged specimens did work properly at low temperatures in this study, and were used for some of the room temperature data. Furthermore, the usual serrated faces used to grip specimen or tab surfaces do not work well with the glass matrix system used in the present study due to matrix and interface fragmentation which makes load introduction a problem even with untabbed specimens (Fig. 1). Thus, it appears that an entirely new set of test methods is needed to characterize the properties of fiber composites at high temperatures.

This paper describes the development of a tensile specimen intended for use up to 1000°C. The specimen has been used to obtain longitudinal and transverse tensile properties up to 600°C, but should also be applicable to higher temperatures and other orientations as well as for multidirectional laminates, including simulated shear tests. This work is part of a continuing effort to: (1) develop high temperature test methods, (2) characterize the properties of available composite systems, (3) study modes of failure, and (4) adapt and develop models for the properties of high temperature composites.

EXPERIMENTAL METHODS

Materials

The materials used to date have been unidirectional Nicalon fiber reinforced glass matrix (Code 1723) supplied by Corning Glass Works. These were developmental materials from various batches having an average fiber volume fraction of approximately 0.35. The neat matrix has a coefficient of thermal expansion of approximately $45.8 \times 10^{-7}/°C$, and Young's modulus is 86 GPa at room conditions [10]. Nicalon single fibers are variously reported to have approximate values of $\alpha = 31 \times 10^{-7}/°C$, E = 180-200 GPa and a tensile strength of

2.1 GPa. The composites were machined wet with diamond edged wheels and blades.

Equipment

The test system used in this program is an Instron Mod. 1331 servohydraulic testing machine fitted with an Instron Mod. 3117 high temperature furnace system. The furnace system includes a three zone split furnace, microprocessor controller, water cooled alignment rings, reverse stress pull rods, and (external) axial and transverse quartz rod extensometers. The system is designed for testing to 1000°C. As part of the project, special grips for tension and compression are being developed, again for use to 1000°C. The tension grips to be discussed later were adapted from Applied Test Systems Mod. 4053A wedge couplings. All tests were run in air with a heat-up time of 1 to 2 hours and displacement rates which produced failure in several minutes (strain rates on the order of $10^{-4}/s$).

RESULTS AND DISCUSSION

The first test method investigated in this program was uniaxial tension. Tensile data are most important for most materials, and tension tests can be adapted to provide simulated shear loading as well as flaw sensitivity studies. The goal was to develop a test method useful to 1000°C which would provide gage-section failures for unidirectional specimens in various directions; tests which work for unidirectional specimens usually also work for multidirectional laminates. Exploratory work indicated three limitations:

1. Specimen surfaces could not be loaded directly with serrated grip faces.

2. Straight coupons with adhesively bonded tabs were impractical due to the lack of tough high temperature adhesives.

3. Standard dumbbell shaped specimens split at the shoulders for longitudinal specimens; splits also initiate at very low stresses at holes.

Figures 2, 3, and 4 illustrate the tensile test method which overcomes these limitations. The specimen shape has a constant width but a reduced thickness in the gage section produced by wet grinding in small increments. Figure 4 gives the dimensions which were found to yield satisfactory results for 3.2 mm thick unidirectional Nicalon/1723 glass; the width was a constant 6.4 mm in all cases. The thickness is reduced in two stages, first following a 5° taper, then a 76 mm radius with a smooth transition to the gage section. Smooth metal wedge blocks are fit to the straight and 5° taper sections as shown in Figure 3; the metal used depends on the test temperature. When a displacement is applied to the grip, load is introduced through the 5° taper section, which is also compressed normal to the surface to suppress splitting at the shoulder. (A similar type of load introduction has been used with acoustic emission studies on width tapered specimens [11].) The 76 mm transition outside the wedge blocks insures that failure will not initiate from the stress concentration at the edge of the wedge block. The 76 mm radius has proven sufficient to prevent shoulder splitting in this region for the very slight thickness reduction between the wedge blocks and the gage section. The resulting overall specimen length of 76 mm for a 19 mm gage length is short enough for most small samples available in this study. A larger transition radius such as 150 mm is recommended if larger samples are available. Most longitudinal and transverse specimens failed in the gage section, usually near the end of the shoulder, as shown in Figure 2 (all longitudinal specimens). Some of the failures spread into the grips after initiating in the gage section, including those shown for 20° and 600°C. Multidirectional laminates have not been tested with this specimen shape, but no problems are anticipated beyond providing sacrificial plies to be ground away in the reduced thickness section.

Figure 5 gives a typical longitudinal stress-strain curve at 20°C with loading-unloading loops. The general features of the curve are as expected [12,13] for brittle matrix materials: an initially linear portion with an elastic modulus close to the rule of mixtures value; matrix cracking at strains in the 0.1 to 0.2% range which produce softening; a reduced elastic modulus at high strains which is close to that for the fibers alone ($E_f V_f$); and an ultimate strength and strain close to the fiber values, again assuming no matrix contribution. Figure 6 gives typical stress-strain curves for different temperatures. Variations in the curve shapes appear to derive in part from material variability in these early samples, rather than entirely from the effects of temperature. The data given here are presented as a demonstration of the test method and the general property trends; larger batches of consistent material are required to firmly establish the properties.

The dashed portions of the curves in Figure 6 are indicative of an inherent limitation on the use of quartz rod extensometers. The extensometer must be removed prior to specimen failure to prevent damage to the quartz rods (one rod was damaged in these tests). As the tests are run, both the ram displacement and the extensometer output are recorded simultaneously, and the strain is estimated from the stress-stroke curve after removal of the extensometer. There is also a problem with attachment of the extensometer to the specimen surface. The only reliable method of several attempted involved machining small indentation points approximately 0.1 to 0.15 mm deep with a carbide drill. Larger indentation points caused splitting at low stresses which did not affect the longitudinal strength, but did cause inaccuracy in the strain measurement. The shallow indentations provided an adequate extensometer anchor without affecting the strain measurements or the specimen strength.

Figure 7 gives the longitudinal ultimate tensile strength as a function of temperature; all specimens were taken from a single plate sample. The data show little strength loss except at 600°C. The increase in strength at elevated temperatures reported for flexural tests on some systems [13-15] is not observed in these tests.

Figure 8 shows some typical features of failed longitudinal specimens, as well as a typical cross-section (d). At temperatures below 600°C, failures were generally similar to Fig. 8(a), with pullout lengths on the order of 10-30 fiber diameters. Matrix cracks evident in (a) and (c) were spaced on the order of 0.1 mm apart at failure; individual matrix cracks typically extend across most of the specimen width. At 600°C some specimens failed in the same manner as at lower temperatures, but others showed extensive fiber pullout (b); in these cases the stress-strain curve did not show the usual sudden fracture, but rather an extended tail, gradually approaching zero stress as the fibers were pulled out. The extensive pullout behavior is typical of conditions where the matrix does not tend to shrink relative to the fibers, as with low coefficient of expansion matrices such as LAS [15].

Additional tests have been run in tension in the transverse direction as well as in compression in the transverse and longitudinal directions. Due to limitations on the amount and consistency of available materials, these have only been exploratory results. The tension test appears to function properly for off-axis as well as longitudinal specimens. However, the Nicalon/glass system is very brittle and difficult to handle without damage for other than longitudinal specimens, and many specimens were broken during machining and gripping operations. There may also have been some isolated regions of porosity which contributed to this problem in these early batches of material. Table I gives the average elastic constants at 23°C for the longitudinal and transverse directions. These properties appear reasonable based on the reported fiber and matrix properties given earlier. Table II gives strength results of invididual transverse tension tests for two batches of material, illustrating the problems with testing for this property. The low strength values represent local porosity or handling damage, and are not meaningful properties; a typical strip of material several cm long was usually found to have one or two weak spots when flexed with the fingers. The higher strength values appear to be representative of the material away from the weak spots, and may be indicative of typical transverse tensile properties, with strengths around 25 to 30 MPa and failure strain around .03 to .04%. Stress-strain curves were essentially linear to failure.

A more complete set of test results will be generated as material becomes available. While difficult to establish, the transverse properties are likely to be critical in many applications, since it is difficult to forsee structural use of high matrix modulus materials beyond the point where cracking initiates. As expected from micromechanics, and from the apparently low interface strength, the strain to failure in the transverse direction is below the matrix cracking strain for loading parallel to the fibers. Thus, transverse property data should be considered during materials development efforts as well as in characterizing established materials. Since specimen preparation is difficult, transverse flexural data could also provide useful information at early stages of development; there should be fewer difficulties with flexural testing in the transverse direction, since failures are essentially brittle. The in-situ transverse properties for multidirectional laminates are also of great interest since they include lamination residual stresses and avoid the size and processing effects inherent in preparing relatively thick unidirectional transverse specimens.

Exploratory compression tests using end loading indicate that the transverse compression strength may be an order of magnitude higher than the best tensile values. Current efforts are in the areas of developing an appropriate compression test as well as extending the tensile results to higher temperatures using glass/ceramic matrix materials.

CONCLUSIONS

Current mechanical test methods for fiber composites are limited to low temperature use, while it is important to fully characterize the properties of evolving high temperature materials. A tensile test method has been developed which should provide adequate tensile (and probably shear) data up to at least 1000°C for unidirectional composites at various orientations, and for multidirectional laminates.

ACKNOWLEDGEMENTS

This work was supported by NASA-Lewis Research Center; the high temperature furnace and extensometer system was provided by Instron Corporation under a cooperative research program, and materials were supplied by Corning Glass Works. The authors are grateful for the generous support and assistance provided in each case.

REFERENCES

1. S.W. Tsai and H.T. Hahn, Introduction to Composite Materials, Technomic, Westport, CT (1980).

2. ASTM Test Method D638-82a, Type I (1982).

3. ASTM Test Method D3039-76 (1976).

4. J.M Whitney, I.M. Daniel and R.B. Pipes, Experimental Mechanics of Fiber Reinforced Composite Materials, The Society for Experimental Stress Analysis, Brookfield Ctr., CT, Ch. 4 (1982).

5. H.T. Hahn and J.G. Williams, "Compression Failure Mechanisms in Unidirectional Composites," NASA TM85834, NASA-Langley Research Center (1984).

6. D.E. Walrath and D.F. Adams, "The Iosipescu Shear Test as Applied to Composite Materials," Experimental Mechanics 23, 105 (1983).

7. J.G. Williams and M.D. Rhodes, "Effect of Resin on Impact Damage Tolerance of Graphite/Epoxy Laminates," in Composite Materials, Testing and Design (Sixth Conference) STP 787, I.M. Daniel, ed., ASTM, p. 450 (1982).

8. F.J. McGarry, J.F. Mandell and S.S. Wang, "Fracture of Fiber Reinforced Composites," Polymer Engineering and Science 16, 609 (1976).

9. D.J. Wilkins,d J.R. Eisenmann, R.A. Camin, W.S. Margolis and R.A. Benson, "Characterizing Delamination Growth in Graphite/Epoxy," in Damage in Composite Materials, STP 775, K.L. Reifsnider, ed., ASTM, p. 168 (1982).

10. K. Chyung, Corning Glass Works, Corning, N.Y., Personal Communication (1984).

11. J.M. Hale and J.N. Ashton, "A Novel Tensile Specimen for Sheet Material," Composites 15, 67 (1984).

12. J. Aveston, G.A. Cooper and A. Kelly, "Single and Multiple Fracture," in The Properties of Fibre Composites, Proceedings, National Physical Laboratory, IPC Science and Technology Press, Ltd., p. 15 (1971).

13. K.M. Prewo, "A Compliant, High Failure Strain, Fibre-Reinforced Glass-Matrix Composite," J. Mater. Sci. 17, 3549 (1982).

14. K.M. Prewo and J.J. Brennan, "High-Strength Silicon Carbide Fibre-Reinforced Glass Matrix Composites," J. Mater. Sci. 15, 463 (1980).

15. J.J. Brennan and K.M. Prewo, "Silicon Fibre Reinforced Glass-Ceramic Matrix Composites Exhibiting High Strength and Toughness," J. Mater. Sci. 17, 2371 (1982).

TABLE I

TENSILE ELASTIC PROPERTIES

(23°C, UNIDIRECTIONAL, V_f = .35-.40)

LOAD DIRECTION:	LONGITUDINAL	TRANSVERSE
YOUNG'S MOD.(GPa)	128	101
POISSON'S RATIO	0.24	0.19

TABLE II

TRANSVERSE TENSILE STRENGTH

(23°C, V_f = .35-.40)

BATCH II
2.7
1.3
AVE = 2.0 MPa

BATCH III
30.0 } Ave. strain to
23.0 } Failure = .035%
2.3
AVE = 18.0 MPa

FIGURE CAPTIONS

Figure	Caption
1	Surface Damage from Serrated Grip Face
2	Typical Broken Specimens from New Tensile Test Method
3	Tensile Specimen in Grip
4	Tensile Specimen Geometry
5	Cyclic Tensile Stress-Strain Curve of Unidirectional Nicalon/Glass Composite at 20°C
6	Typical Tensile Stress-Strain Curves of Unidirectional Nicalon/Glass Composite
7	Tensile Strength of Unidirectional Nicalon/Glass Composite
8	(A) Typical Fracture Profile (400°C) (B) Extensive Fiber Pullout for One Specimen only at 600°C (C) Matrix Cracks Away from Fracture Surface (20°C) (D) Typical Cross-Section (Fiber Diameter = 10-15 μm)

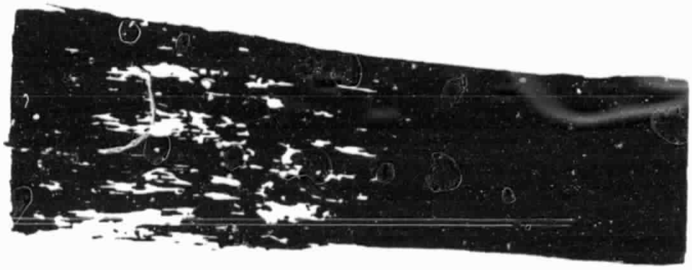


FIG. 1. SURFACE DAMAGE FROM SERRATED GRIP FACE

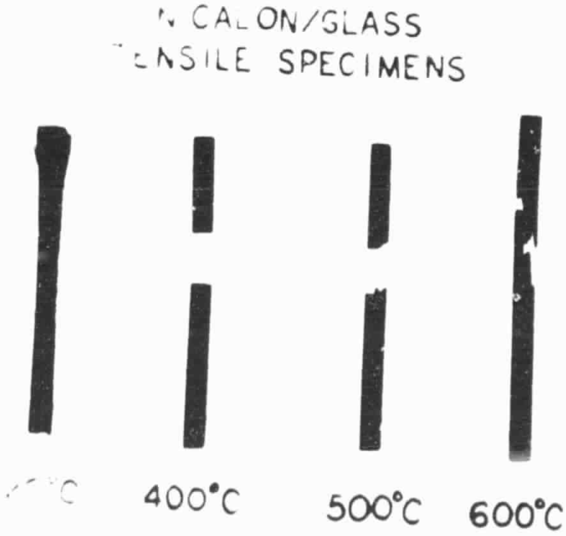


FIG. 2. TYPICAL BROKEN SPECIMENS FROM NEW TENSILE TEST METHOD

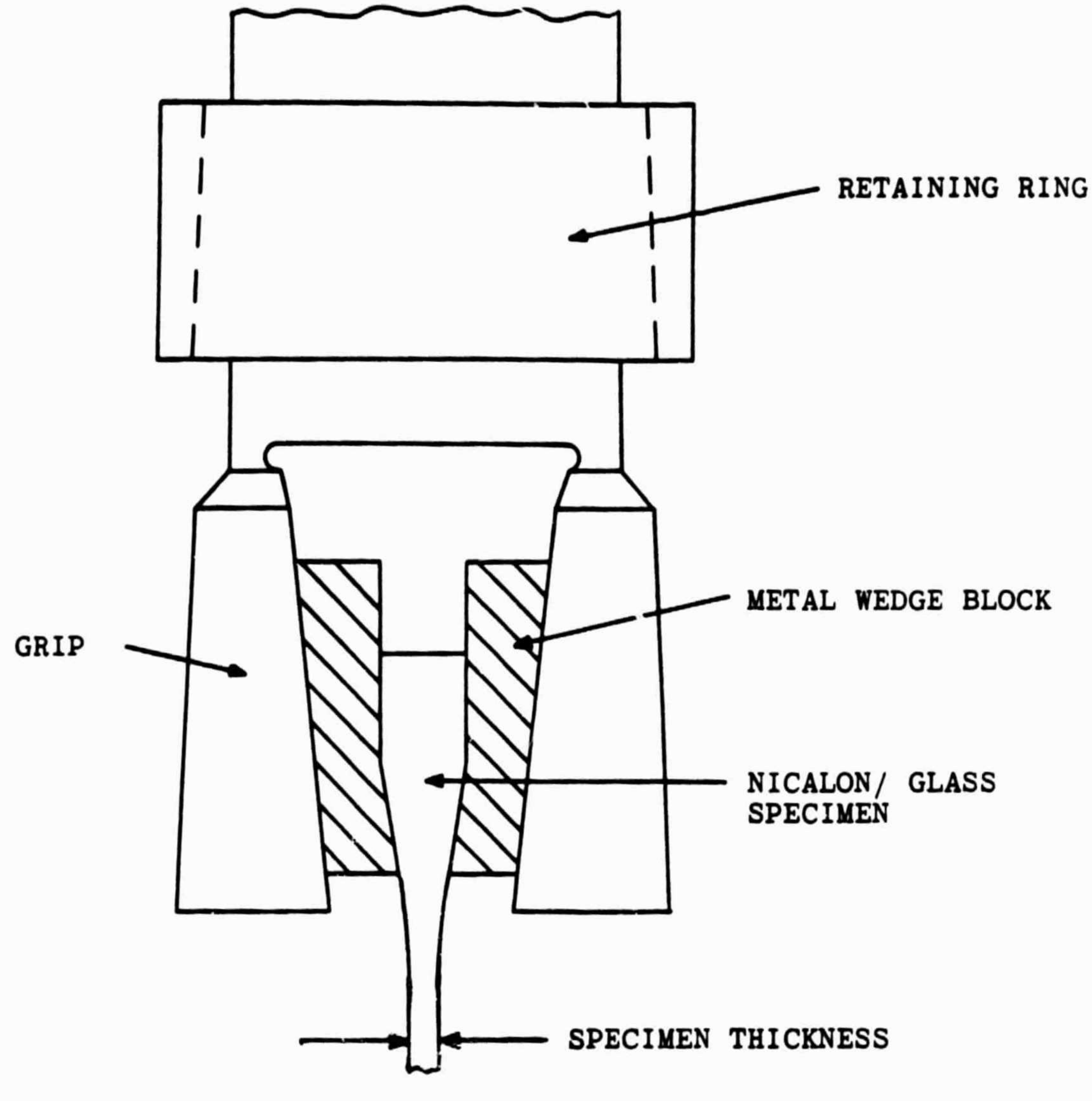


FIG. 3.
TENSILE SPECIMEN IN GRIP

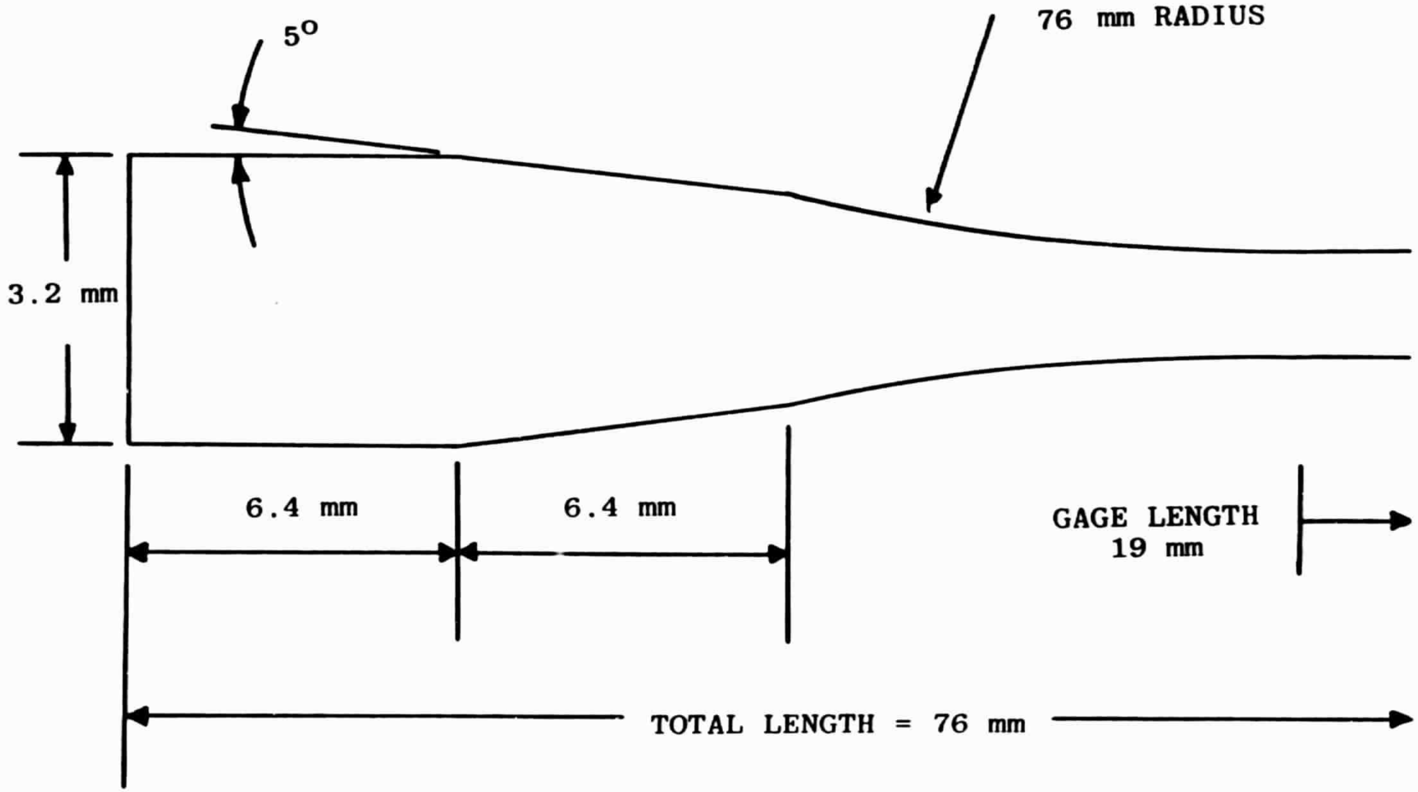


FIG. 4.

TENSILE SPECIMEN GEOMETRY

FIG. 5.
CYCLIC TENSILE STRESS-STRAIN CURVES OF UNIDIRECTIONAL NICALON/ GLASS COMPOSITE.

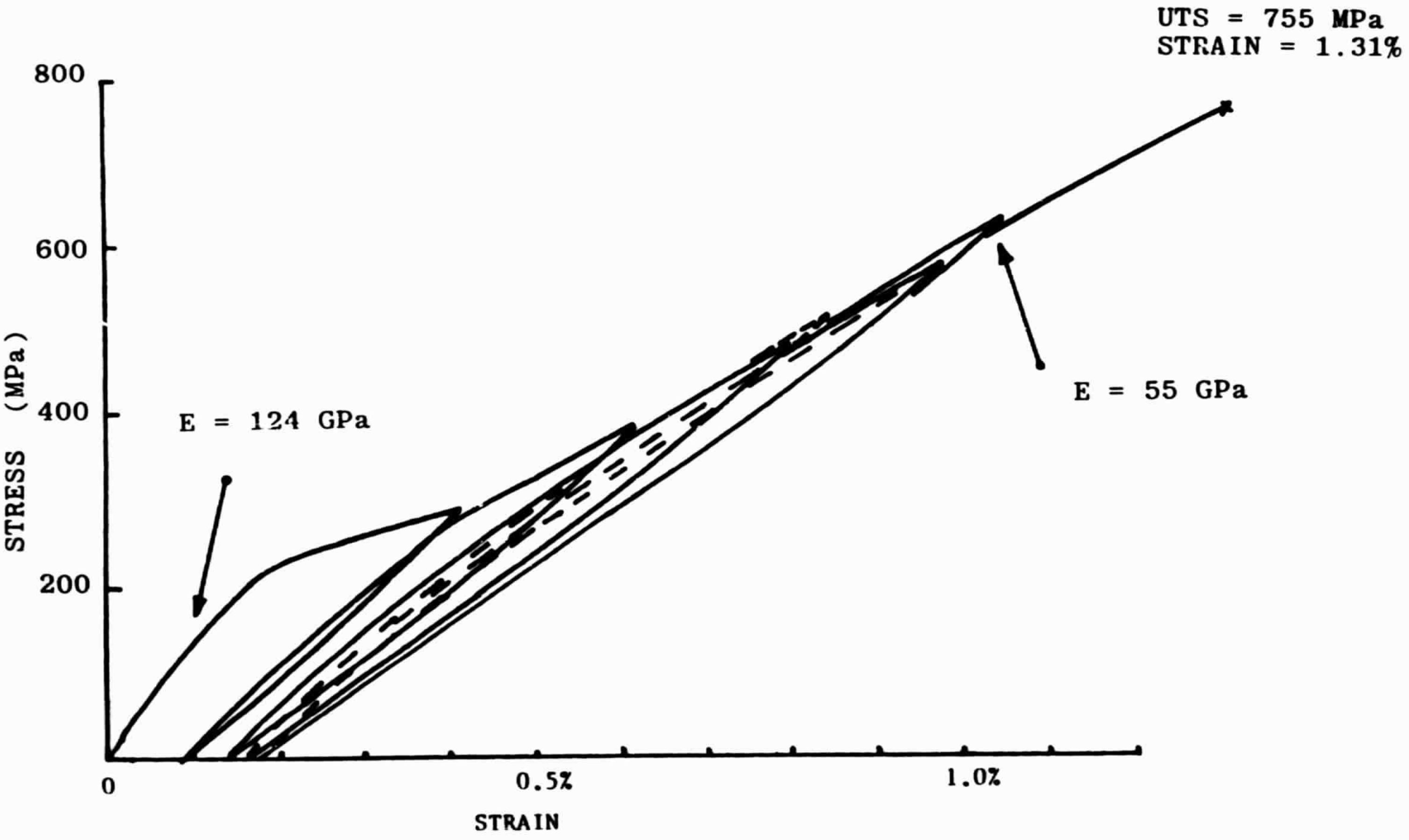


FIG. 6.

TYPICAL TENSILE STRESS-STRAIN CURVES OF UNIDIRECTIONAL NICALON/ GLASS COMPOSITE.

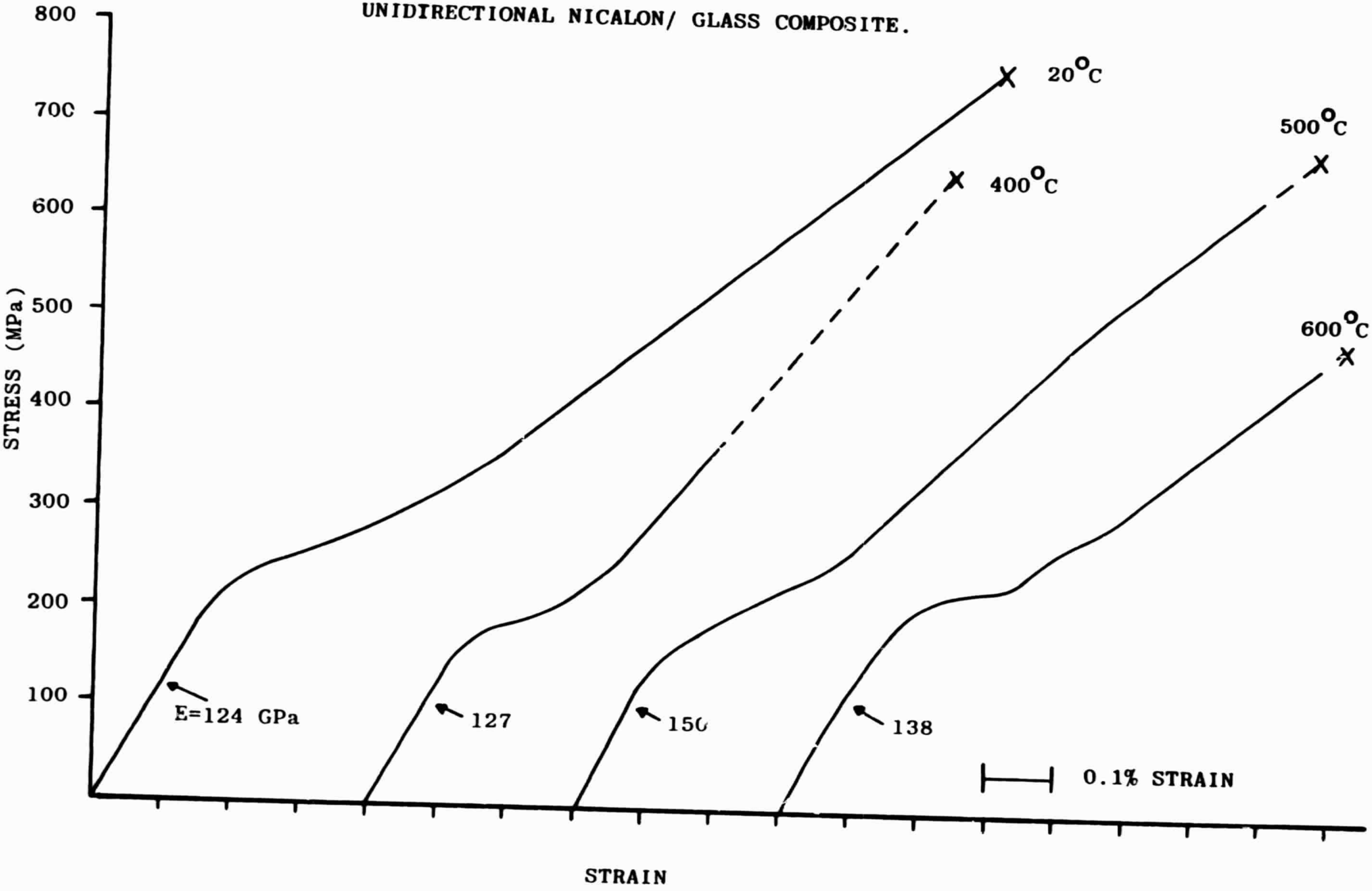
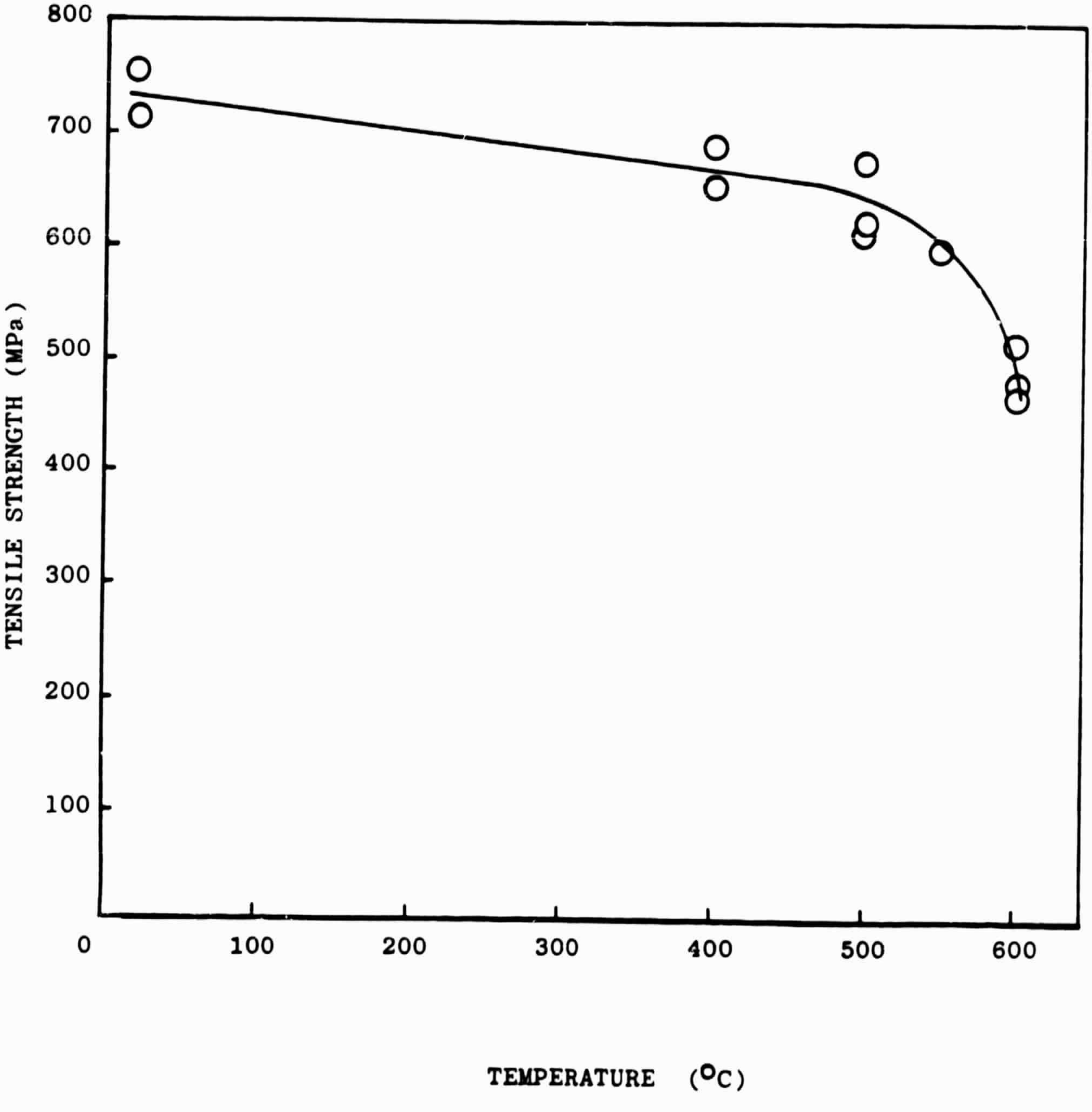
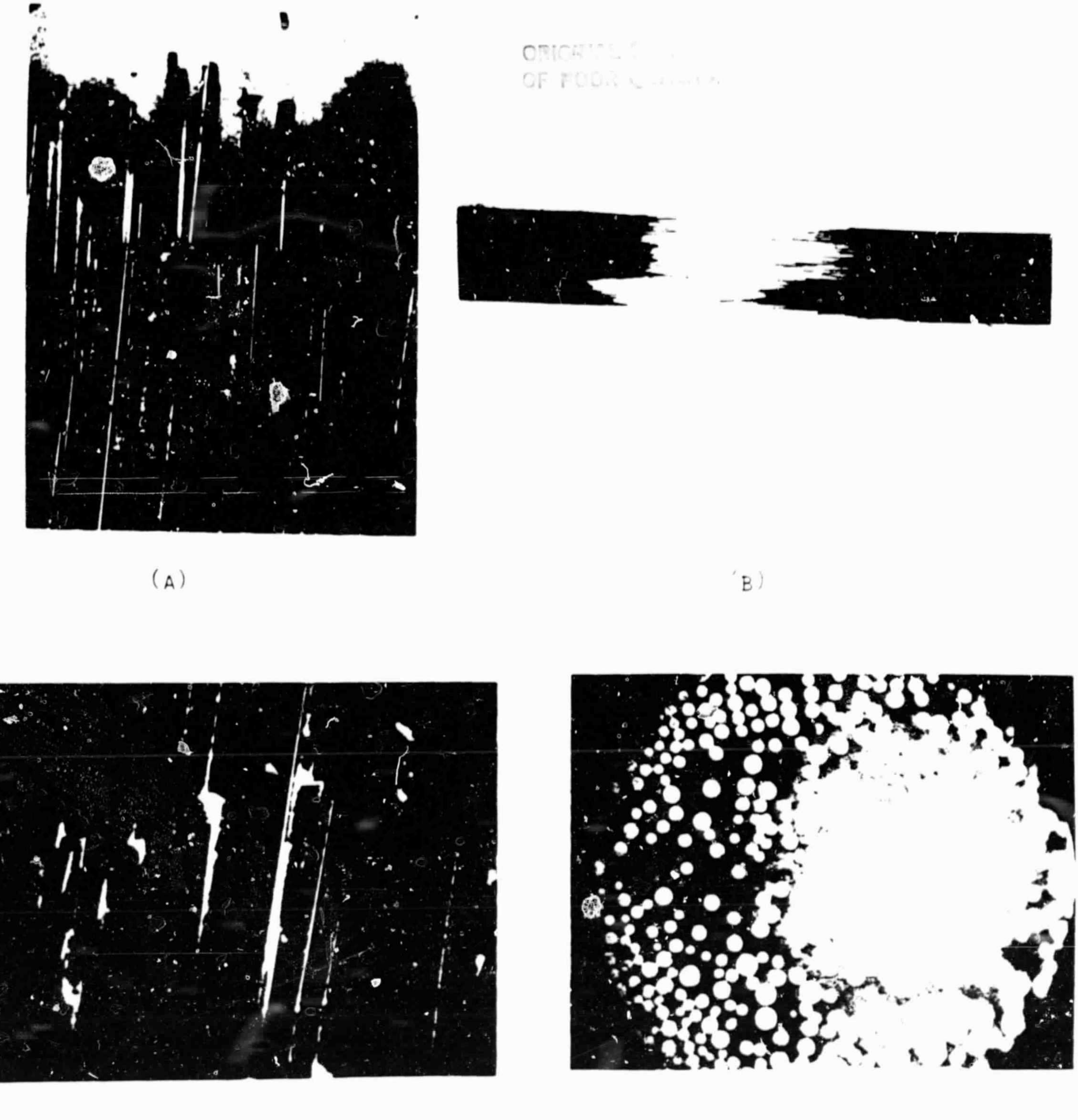


FIG. 7.

TENSILE STRENGTH OF UNIDIRECTIONAL NICALON/ GLASS COMPOSITE.





(A) (B)

(C) (D)

FIGURE 8. (A) TYPICAL FRACTURE PROFILE (400°C); (B) EXTENSIVE FIBER PULLOUT FOR ONE SPECIMEN ONLY AT 600°C; (C) MATRIX CRACKS AWAY FROM FRACTURE SURFACE (20°C); (D) TYPICAL CROSS-SECTION (FIBER DIAMETER = 10-15 μm)