Round Robin Assessment Of The Single Fiber Fragmentation Test

M. J. RICH, L. T. DRZAL, D. HUNSTON, G. HOLMES AND W. MCDONOUGH

ABSTRACT

The Versailles Project on Advanced Materials and Standards (VAMAS) was established following an economic summit meeting in 1982 held at Versailles by the Heads of State of the seven leading industrial nations and representatives from the Commission of the European Communities. The objective of the VAMAS coalition is to promote world trade in high technology areas which includes the assessment and development of test methods for advanced materials. In 1998, Technical Work Area 5, Polymer Composites, established a program to measure the fiber-matrix interfacial behavior with an initial focus on the single fiber fragmentation test (SFFT).

The National Institute of Standards and Technology (NIST), in dialogue with Michigan State University, developed a set of protocols for conducting the single fiber fragmentation test which included coupon size, coupon fabrication, testing parameters, data analysis, etc. Seven laboratories participated in the round robin with each laboratory supplied with 10 coupons to be tested in compliance with the NIST protocols. The results of the round robin found that five of the laboratories produced data that were in close agreement while two of the participating laboratories had results that were 20 % variant from the population mean. A review of the coupons returned by the two laboratories with the variant data showed that one laboratory incorrectly measured the critical length and the other laboratory incorrectly measured the fiber diameter. When corrected for these errors, the outlying data are shifted to be in statistical agreement with the population mean. The results of the round robin show that the fragmentation test generates a consistent measurement of the aspect ratio when the test is executed in accordance to test protocols.

INTRODUCTION

In 1982 representatives and Heads of State from Europe, Japan, Canada, and the United States met in Versailles for the first Joint Economic Summit. One issue addressed at that inaugural meeting was the need to encourage international trade in high technology products through collaboration on establishing protocols of practice and specifications for advanced materials. This led to the establishment of VAMAS, the Versailles Project on Advanced Materials and Standards. The VAMAS Steering Committee established Technical Working Areas (TWA) addressing specific issues that were identified in need of the development of test methodologies. The focus of the TWAs is to conduct the basic research that precedes the establishment of vigorous standards. By working closely

M. J. Rich and L. T. Drzal, College of Engineering, 2100 Engineering Bldg., Michigan State University, East Lansing, MI 48824-1226

Donald Hunston, Gale Holmes, and Walter McDonough, National Institute of Standards and Technology, Gaithersburg MD, 100 Bureau Drive, Gaithersburg, MD 20899-8543

with organizations such as ASTM, the time frame for establishing standards can therefore be shortened [1].

In 1998, TWA5, Polymer Composites, established a program on measuring the mechanical properties of the fiber-matrix interface. The program chose to focus, at least initially, on the single fiber fragmentation test (SFFT). The SFFT is a micromechanical method used to evaluate the level of adhesion between a continuous reinforcing fiber and a matrix, generally a polymer matrix. In practice, a single filament, such as a carbon fiber, is axially aligned in a micro-tensile coupon as illustrated in Figure 1. The test specimen is loaded in tension which causes shear stresses to develop between the rigid fiber and the comparatively lower modulus polymer. With the application of sufficient tensile load, the shear forces will exceed the tensile strength of the encapsulated fiber and the fiber will fail within the coupon. With continued loading, the fragmentation process is repeated until the fiber is rendered to a characteristic aspect ratio. The aspect ratio is an indicator of the level of adhesion between the fiber and polymer. Fibers with low adhesion will have longer aspect ratios than fibers with greater adhesion, when all other factors are held constant.

Figure 1. Schematic representation of the fragmentation process

A number of models have been proposed to relate the average aspect ratio of the fragments to interface characteristics. In the most commonly used approach, the interfacial shear strength, τ, is calculated using a shear lag analysis [2] by the relationship

$$\tau = \frac{\sigma_f d}{2l_c} \qquad\qquad 1$$

where σ_f is the fiber tensile strength, d is the fiber diameter, and l_c is the experimentally determined fiber critical length. Other relationships have been reported that are derivations of Equation 1. Drzal and co-workers[3] used statistical analysis of the fragment lengths employing a two parameter Weibull analysis to describe the critical length distribution, leading to

$$\tau = \frac{\sigma_f}{2\beta}\Gamma\left(1 - \frac{1}{\alpha}\right) \qquad\qquad 2$$

where α and β are, respectively, the Weibull shape and scale parameter of the length/diameter aspect ratio distribution and Γ is the gamma function. Still other-models have been proposed for evaluation of the interfacial shear strength using the fragmentation approach, and these are discussed in a review article by Drzal and coworkers [4].

In the early 1990sa round robin program on interfacial test methods was conducted under the auspices of the Royal Aerospace Establishment (RAE) [5]. The results showed an unexpectedly large laboratory to laboratory variation in the data. This poor agreement among laboratories was blamed, in large part, on lack of standardization. In that program each participating organization was supplied with fiber and resin from master batches. Each organization was responsible for sample fabrication
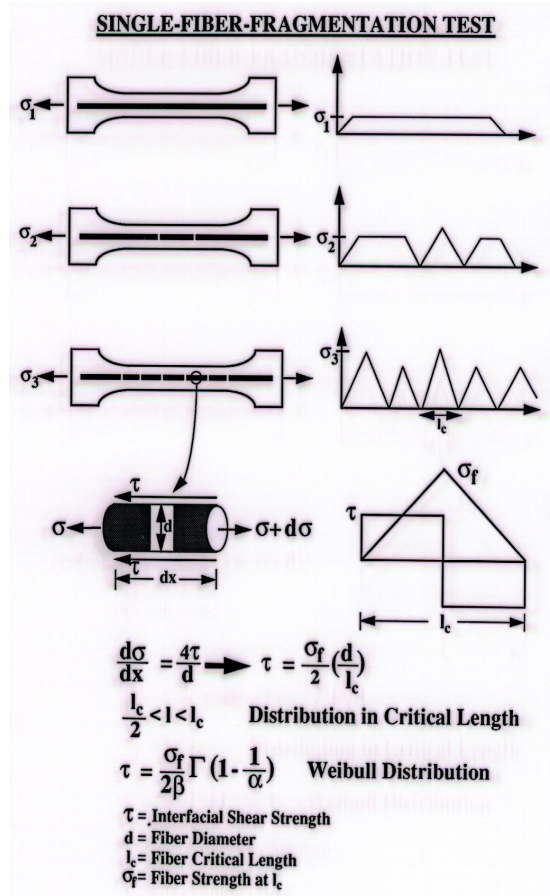
and execution of the test. The participants were given only general instructions on sample fabrication and no protocols were specified for execution of the fragmentation test. The participants were, therefore, allowed to select their own test procedures, such as rate of strain application. Additionally, there was no post-test review to evaluate the quality of the coupons or the accuracy of the measurements reported by the seven participating groups that conducted the fragmentation test.

   To address these issues, a VAMAS program was developed with three goals. First, standardized testing and sample fabrications procedures would be developed. Second, a round robin would be conducted to evaluate the procedures. Third, a database of results would be generated so researchers could evaluate various analysis procedures such as those discussed above. This is the first of several papers which will report the results for various aspects of the VAMAS activity. Here the focus is the single fiber fragmentation test, and the results of the international round robin. In contrast to the RAE program where the participants were given discretion in making the samples and selecting test procedures, the VAMAS project developed a set of detailed procedures for executing the fragmentation test and reporting the data. Another distinction of the VAMAS program is that single fiber fragmentation coupons were supplied to the participants, and the specimens were collected at the conclusion of the program to validate the authenticity of the results and to investigate the cause of any variance in data that might exist between the reporting laboratories.

MATERIALS [1]

   Single fiber fragmentation coupons were made at Michigan State University from master batches of carbon fiber and epoxy resin. Coupon dimensions are provided in Figure 2. The fiber was AS4 fiber lot D1682-3L (Hexcel, Salt Lake City, UT), a surface treated carbon fiber which has a reported tensile modulus of 228 GPa (33.1 Msi), tensile strength of 4.278 GPa (620 ksi), and failure strain of 1.87 % based on tow properties. Marketing reports on the Hexcel internet site report that the average fiber diameter of AS4 is 7.1 μm [6]. The matrix was diglycidyl ether of bisphenol A, Epon 828 (formerly Shell Chemical, now Resolution Performance Products, Houston ,TX) cured with the stoichiometric amount (14.5 g/100 g resin) of meta-phenylene diamine (Sigma-Aldrich Corp., St. Louis, MO). A schedule of 2 h at 75 $^{\circ}$C followed by 2 h at 125 $^{\circ}$C was used to process the single fiber fragmentation coupons. All specimens were processed in a dedicated, clean oven to preclude contamination. Each coupon was examined using transmitted optical microscopy to verify fiber alignment and overall quality. If the fiber was wavy, or bow-shaped within the coupon the specimen was discarded. After fabrication, the fiber diameters were measured using a video caliper at Michigan State University. The single fiber fragmentation coupons were then delivered to NIST where they were cataloged and singularly sealed in a hermetic metallic envelope. Ten fragmentation coupons were provided to each participant for evaluation.

---

[1] Certain materials and equipment identified in this manuscript are solely for specifying the experimental procedures and do not imply endorsement by NIST or that they are necessarily the best for these purposes.
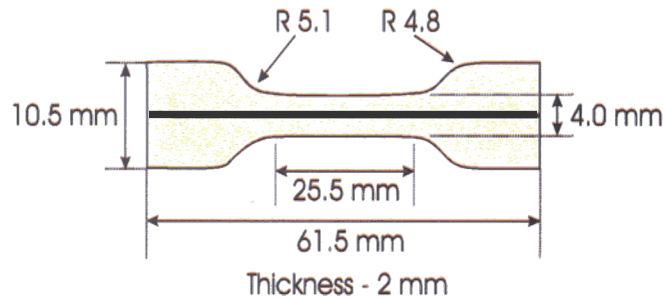
Figure 2. Dimensional details of the fragmentation coupon.

FRAGMENTATION TEST PROTOCOLS

The participants of the fragmentation round robin were provided with a 10 page booklet that delineated the precise procedures to be used to execute the test [7].   The booklet described the procedures and potential difficulties in making measurements of the fiber diameter, detection of breaks, and measurement of fragment lengths. The instructional booklet also provided a template for reporting the entire data set.

In the VAMAS round robin, the application of strain increments was explicitly detailed. The coupons were to be loaded with a series of cycles consisting of a step strain followed by a holding period where the strain is held fixed. Each step strain was to be limited to 0.2 % and held for 8 min after which the gage length was to be scanned and the number of fiber fractures was to be counted. Ten minutes after the previous strain increment, the next step strain was to be applied to the specimen. The small applications of step strain and the ensuing holding period ensured that the coupons were tested in a static condition which eliminates the influences of different loading rates that might otherwise occur among the participants. Using these protocols, each coupon required approximately 3 to 4 h to obtain saturation where the encapsulated fiber was rendered to its characteristic aspect ratio. Saturation was defined as no increase in the number of fiber fractures after three successive step strain cycles. At saturation the fragment lengths were to be measured and reported in sequential order as they occurred in the gage area. Each participant was to report the fiber diameter, number of breaks as a function of coupon strain, and the individual fragment lengths for each coupon. The samples were to be returned to NIST after testing was completed.

The primary steps to conduct the fragmentation test were:
1. Measure the fiber diameter at 5 separate locations in gage area to accuracy of +/- 1 µm
2. Place fiduciary marks on coupon to be used for coupon strain measurement
3. Increment coupon strain by 0.2 %, hold for 8 min before counting fiber breaks
4. After 10 min from conclusion of previous strain increment, apply next step strain cycle. Repeat to saturation.
5. Photodocument the fiber under polarized light when at saturation
6. Measure fragment lengths before load is released from coupon
7. Report results using form provided and return specimens to NIST

A total of seven laboratories from five countries participated in the VAMAS international fragmentation round robin as listed in alphabetical order in Table 1.

Table 1. List of participating organizations.

Organization	Contact Person
Bundesanstalt für Materialforschung und - prüfung (BAM), Berlin, Germany	Andreas H. Hampe
Centro de Investigacion Cientifica, Mérida, México	Pedro J. Herrera-Franco
Research Center for Advanced Science and Technology, The University of Tokyo	Nobuo Takeda
Michigan State University, East Lansing MI USA	L. Drzal and M. Rich
National Institute of Standards and Technology (NIST), Gaithersburg, MD USA	D. Hunston, G. Holmes, and W. McDonough
National Physical Laboratory (NPL), Middlesex, UK	J. Lodeiro and G. Sims
University of Sheffield, Sheffield, UK	Frank R. Jones

RESULTS AND DISCUSSION

   The experimental results are presented anonymously without association to any of the participating laboratories.

The Fragmentation Process

   Each participating group was instructed to determine the number of fiber fractures as a function of coupon extension. Figure 3 presents a summary chart of the number of fiber fractures versus strain for a set of 10 fragmentation coupons evaluated by one of the test laboratories. In general, it can be seen that the fracturing process began at ≈1.5 % strain and increased linearly to ≈2.5 % strain. Saturation was attained at 2.5 % to 3.0 % coupon strain. Similar trends were found for the other reporting agencies.   For calculation of the interfacial shear strength using shear-lag analysis of fragmentation data, the relationship between the number of breaks to coupon strain is not relevant. The valid application of Equation 1 requires that the encapsulated fiber be rendered to its critical length where no additional fractures can be induced with the application of additional strain. The
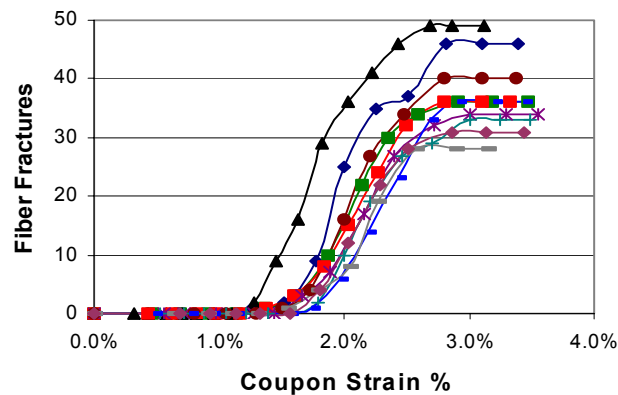

Figure 3. Number of fiber fractures as a function of
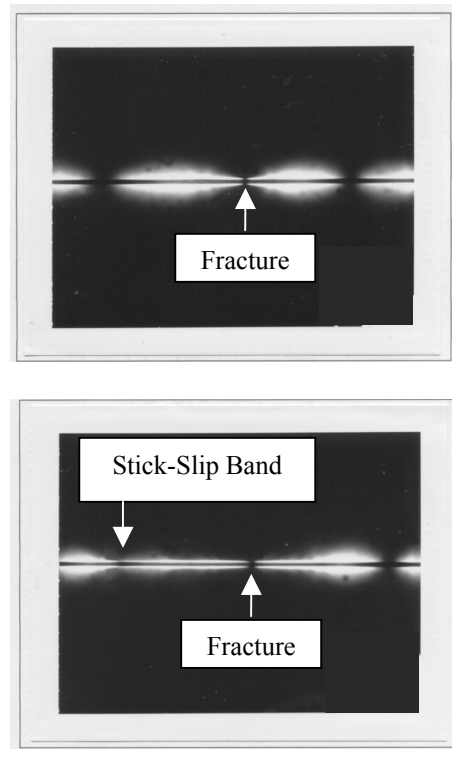coupon strain for a set of 10 specimens.

Figure 4. AS4 carbon fiber in Epon 828-
MPDA matrix at critical length.

behavior of the sample set in Figure 3 shows that the critical length was achieved in all ten coupons at 3 % strain or less (i.e. additional strain produced no further fractures).

The use of polarized light microscopy is often used to qualitatively assess the failure mode of the encapsulated fiber. The samples made of AS4 carbon fibers in Epon 828-MPDA epoxy exhibited an intense birefringent stress pattern at the locus of failure as reported by one of the participants in Figure 4. The top photograph of Figure 4 shows a symmetrical stress pattern typical of most breaks in the VAMAS fragmentation coupons. Breaks, as presented in the bottom photograph of Figure 4, were common, where dark sections occurred within the stress pattern which are indicative of a stick-slip crack front.

Critical Length Measurements

The critical length normalized to its fiber diameter can be used to evaluate the variation in performance of the single fiber fragmentation test. The VAMAS fragmentation test protocols directed that the fiber diameter be measured 5 times in the gage area to an accuracy of +/- 1 μm. The use of video calipers to make the diameter measurements was encouraged, however, each participating group was given discretion to select the method to make the diameter measurements. Considerable variation in mean fiber diameter was reported as can be seen in Table 2. Four of the participating agencies reported a mean fiber diameter of 7.0 μm to 7.1 μm, which is in good agreement with the product data sheet value of 7.1 μm. The technical support staff at Hexcel report a lower diameter of AS4 at 6.8 μm with a variation of less than 5 % [8]. Therefore, the diameter range within the first standard deviation is 6.5 μm to 7.1 μm based on a mean of 6.8 μm. Three of the laboratories reported a mean diameter greater than 7.6 μm, which is at variance with the manufacturer's data and indicates that the data are incorrect. It should be noted that the precision of the measurements among the 7 participants, as indicated by the coefficient of variation in Table 2,

Table 2. Fragmentation data reported by participating laboratories.

Laboratory	Fiber Diameter			Aspect Ratio		
	Diameter µm	Standard Deviation µm	Coefficient of Variation %	l/d Aspect Ratio	Standard Deviation	Coefficient of Variation %
1	7.0	0.4	5.3	76.2	25.0	32.8
2	7.0	0.2	3.4	74.2	18.8	25.4
3	7.6	0.3	3.3	64.7	10.5	16.2
4	7.1	0.2	3.1	82.4	23.0	27.9
5	7.1	0.3	4.8	66.9	8.0	12.0
6	7.7	0.4	5.2	65.1	11.4	17.5
7	8.1	0.2	2.1	50.5	14.7	29.1
HEXCEL[9]	6.8	--	<5%			

was less than 5.3 %, a value that is in general agreement with the reported variation by Hexcel. The diameter data suggests that the precision in making the measurements among the seven participants is acceptable although the accuracy for three of the laboratories may be in error.

The aspect ratios reported from the seven agencies are provided in the chart of Figure 5. The error bars on each data point represent the standard deviation among that sample set of 10 coupons. The green horizontal bar is the population average of the sample set, and may be thought of as the "average of the average" with a value of 68.6. The red bar is the first standard deviation of the population mean, 19.2. The coefficient of variation for the population was 28 %. The variation within a sample set can be very large, ranging from a low of 12 % for Laboratory 1 to 32.8 % for Laboratory 6, as can be seen in Table 2. Two sets of data reside outside the bound of the first standard deviation. The data set from laboratory 4 contained one coupon with a very large aspect ratio, 128, which was independently confirmed upon inspection of the returned samples. That particular specimen exhibited the highest aspect ratio in the entire VAMAS data set and is outside the third standard deviation of the population mean. When this deviant coupon is removed from the sample set, the data set is shifted to within the first standard deviation as indicated by the data positioned immediately to the right for laboratory 4.
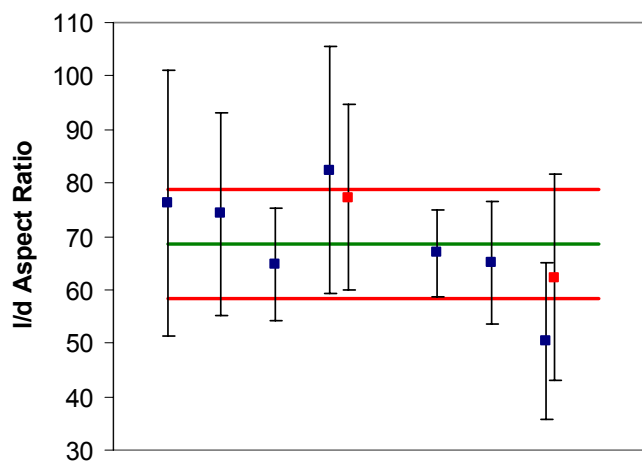

Figure 5. Aspect ratios of AS4 carbon fibers in Epon 828-MPDA matrix reported by seven laboratories..
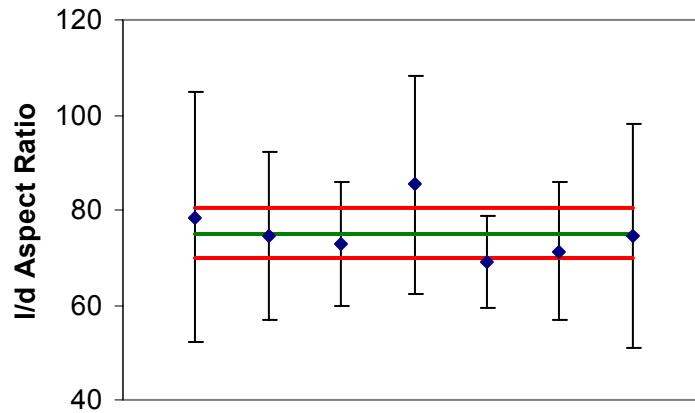
120

100

l/d Aspect Ratio

80

60

40

Figure 6. Projected l/d aspect ratios using manufacturer's diameter value of 6.8 μm.

 Laboratory 7 reported an average aspect ratio of 50.5, significantly lower than the population mean of 68.6.   Post-test measurement of the fragmentation coupons found a fragmentation length approximately 20 % greater than the value reported by laboratory 7.   Laboratory 7 ascribed the disparity to a calibration error.   Laboratory 7 employed an automated image analysis method to measure the fragment length. In general the data from the image analysis system were congruent with data manually collected using a video calipers. However, the image analysis system made incorrect measurements in about 1 % of the fibers. It is suspected that the stick slip bands, shown in Figure 4, may cause corrupt readings by the optical system. The application of new calibration factor shifted the data of laboratory 7 to within the range of the first standard deviation of the population as noted by the red data point on the right hand side for this laboratory's results in the chart of Figure 5. Laboratory 7 also reported the largest fiber mean diameter of 8.1 μm.

   None of the 7 reporting laboratories matched the diameter reported by the manufacturer, 6.8 μm, although four laboratories were within 0.3 μm on average. More accurate and rigorous procedures for measurement of the fiber diameter are required to improve the accuracy of the fragmentation test and to reduce the scatter. An example of the possible improvements that may be achieved when using more accurate diameter values is presented in Figure 6.   In this chart, the aspect ratios of all fragmentation coupons were recalculated using an average of 6.8 μm. The scatter in the data is reduced, with only a 7 % coefficient of variation for the population. The variation within a sample set of 10 coupons is largely unchanged when an average diameter of 6.8 μm is used to normalize the fragment lengths. The coefficient of variation for each sample set using 6.8 μm diameters is nearly the same as for the data determined for each participant. This scatter reflects the actual variation in interfacial shear strength for these lots of AS4 carbon fiber in Epon 828-MPDA epoxy. The typical variation within a sample set for these lots of materials has a coefficient of variation of 25 % for a set of 10 fragmentation coupons. Previous work on other batches of AS4 carbon fibers in Epon 828-MPDA reported lower values of mean aspect ratio with a lower variation in fragment lengths [9].

CONCLUSIONS

   Under the sponsorship of VAMAS, an interlaboratory round robin on the fragmentation test was completed with the participation of seven laboratories.   Each participant was supplied with 10 fragmentation coupons for determination of fiber diameter and the critical l/d aspect ratio. A set of procedural protocols to conduct the test were provided in order to minimize operator influences on the test.

Four of the participating laboratories measured a mean fiber diameter marginally greater than the diameter reported by the manufacturer. Three laboratories reported an average fiber diameter significantly greater than the manufacturer's specification. The variance in fiber diameter calls for more rigorous procedures to accurately measure the fine dimensions of carbon fiber diameters. The precision in diameter measurements reported by the VAMAS participants was in very good agreement with the manufacturer's reported value for coefficient of variation of nominal 5%.

Two laboratories reported a mean l/d aspect ratio that was greater than one standard deviation unit from the population average. In one case, the variance was attributed to the true variation of the data where one specimen exhibited an aspect ratio more than three standard deviation units from the population average. The other laboratory that was at variance with the population statistics made a systematic error in calibration of their measurement equipment.

The aspect ratio for the material lots used in the VAMAS program was greater than previously reported work on AS4/Epon 828-MPDA system. The variation within a sample set of 10 coupons ranged from 16 % to 33 %, with an average variation of approximately 25 %. This variation in the VAMAS samples is reflective of the true variation in interfacial shear strength for this set of materials. Projected improvements in the measurement of fiber diameter suggests that the coefficient of variation in aspect ratio measured by multiple laboratories will be approximately 7 %.

Future work will report the results of an effort to derive an algorithm to be consistently applied for the calculation of interfacial shear strength from the single fiber fragmentation test.

ACKNOWLEDGEMENTS

REFERENCES

1   Don Hunston and Herzl Chai, "VAMAS-A Forum for International Cooperation on the Development of Composite Standards", Proceedings of the Fourth Annual Conference on Advanced Composites, Dearborn, Michigan, 13-15 Sept 1988.

2   A. Kelly and W. R Tyson, "Tensile Properties of Fibre-Reinforced Metals: Copper/Tungsten and Copper/Molybdenum", *J. Mech. Phys. Solids*, V13, 1965, pp 329-350

3   L. Drzal, M. J. Rich, J. D. Camping, and W. J. Park, Interfacial Shear Strength and Failure Mechanisms in Graphite Fiber Composites, 35th Annual Reinforced Plastics/Composites Conference, Paper 20C 1980.

4   L. T. Drzal, P. Herrera-Franco and H. Ho, "Fiber-Matrix Interface Tests" in Comprehensive Composite Materials, V5, A. Kelley and C. Zweben, ed Elsevier Science, 2000

5   M. J. Pitkethly, J. P. Favre, U. Gaur, J. Jakubowski, S. F. Mudrich, D. L. Caldwell, L. Drzal, M. Nardin, H. D. Wagner, L. Di Landro, A. Hampe, J. P. Arimstead, M. Desaeger, and I. Verpoest, "A Round-Robin Programme on Interfacial Test Methods", *Composites Science and Technology*, 48, 1993, 205-214

6   Hexcel internet location http://www.hexcelfibers.com/markets/products/continuous/_productlist.htm, June 2002.

7   D. Hunston, W. McDonough, G. Holmes, R. Parnas, L. Drzal, and M. Rich, Test Protocol for Single-Fiber Fragmentation Test International Round Robin, available from the authors.

8   Personal communication Hexcel Compostites, Salt Lake City, UT, 2 July 2002

9   L. T. Drzal, M. J. Rich, M. F. Koenig, and P. F. Lloyd, "Adhesion of Graphite Fibers to Epoxy Matrices: II. The Effect of Fiber Finish", *J. Adhesion*, 16, 1983, 133-152

PROCEEDINGS OF THE AMERICAN SOCIETY FOR COMPOSITES

SEVENTEENTH TECHNICAL CONFERENCE

Edited by
C. T. SUN
HYONNY KIM

Hosted by
PURDUE UNIVERSITY SCHOOL OF AERONAUTICS AND ASTRONAUTICS

Co-Sponsored by
ASTM COMMITTEE D30 ON COMPOSITE MATERIALS

October 21–23, 2002
Purdue University
West Lafayette, Indiana

Click this page to open