Conf-9305135--3

DEVELOPMENT OF NONDESTRUCTIVE EVALUATION METHODS AND PREDICTION OF EFFECTS OF FLAWS ON THE FRACTURE BEHAVIOR OF STRUCTURAL CERAMICS*

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March 1993

Submitted for inclusion in the proceedings of the 7th Annual Conference on Fossil Energy Materials, Oak Ridge, TN, on May 14-16, 1993

*Work supported by the U.S. Department of Energy, Office of Fossil Energy, Advanced Research and Technology (AR&TD) Materials Program, under Contract W-31-109-Eng-38.

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ABSTRACT

Characterization of ceramic matrix composites by nondestructive evaluation (NDE) methods and an understanding of fracture behavior, together with correlation of fracture and NDE data, may provide insight into the prediction of component performance and the development of process technology. Work in this program has emphasized continuous-fiber ceramic matrix composites (CFCCs) with two-dimensional (2-D) lay-ups composed of chemical-vapor-infiltrated (CVI) SiC/SiC materials, primarily those made of Nicalon plain weave with 16 x 16 tows/in. However, one sample that was examined comprised a three-dimensional (3-D) SiC/SiC braid made by Techniweave. All studied samples were provided by Oak Ridge National Laboratory or Ceramic Composites Inc. and consisted of 100 layers/in. or ≈40-45 vol.% fibers. Specimens with cloth lay-up orientations of 0/30/60, 0/90, and 0/45 were examined by 3-D X-ray microtomography to characterize in-plane fiber orientations. Current information suggests that fiber misalignment affects mechanical properties differently for differing materials.

A near-term goal has been to establish detection capability for angular orientation. By using a new 1024 x 1024 x 14 bit detector, images from 3-D X-ray CT data with pixel sizes of <140 μm and a special 2-D fast-Fourier transform (FFT) image processing analysis, we have shown that fiber orientations can be measured to ±2-1/2°. We have also begun to explore 3-D FFT analysis to establish detection of 3-D braid/weave fiber spacing. Another NDE method, multinuclear (^{1}H, ^{13}C, and ^{29}Si) NMR spectroscopy, is being studied to determine the chemical state of the fiber surface and its potential impact on fiber pullout. Surface chemistry of fibers and the chemistry of the interfacial regions in composites are included in the study. We are also conducting initial studies to investigate the bulk composition of the matrix materials (α, β, amorphous phase,

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silica, and oxynitride concentration) and the surface chemistry of Si_3N_4 and SiC fibers. The first ^{29}Si NMR experiments have focused on enhancing the signal from the surface by selectively reducing surface relaxation. A third NDE approach being explored is a low-frequency (0.5 MHz) acousto-ultrasonic method that measures time-of-flight of P-waves propagating in the composite. This method is being studied as a means to characterize, in a bulk sense, the fiber/matrix interfaces in CFCCs.

Fracture mechanics work to correlate with NDE data focused on strength distribution studies of as-fabricated Nicalon fibers obtained from bundle tests. In addition, strength distribution of fractured Nicalon fibers in composites was assessed by measuring fracture mirror radii. Scanning electron microscopy was conducted to determine the distribution of fiber pullout length distribution for fibers in composites, in order to accurately estimate their strength distribution. From the strength distribution plots, scale parameters were determined to be 3.45 GPa for as-fabricated fibers and 1.31 GPa for fibers in processed composites. However, the Weibull moduli for the two distributions were similar. The above-mentioned reduction in strength of the fibers in processed composites is believed to be due to surface flaws and defects, probably introduced during high-temperature processing and handling, as confirmed by a detailed fractographic evaluation.

Effects of fiber misorientation on mechanical properties of NDE-tested CVI continuous-fiber composites are currently being investigated in an effort to correlate NDE results (fiber orientation and density distribution) with fracture properties for continuous-fiber-reinforced ceramic matrix composites.

INTRODUCTION

Intelligent processing of advanced ceramic matrix composite materials using data obtained from nondestructive evaluation (NDE) methods and correlating these data to process related fracture behavior are necessary for process development. The purpose of the work reported for this activity is to develop NDE methods for intelligent processing, examine fracture behavior of composites, and correlate the NDE data with fracture behavior as an input to process understanding. At present, NDE is being conducted on chemical vapor infiltrated (CVI) SiC/SiC continuous fiber composites either produced at Oak Ridge National Laboratory (ORNL) or by others. The CVI composite specimens studied to date are of two-dimensional plane-weave specimens made of Nicalon multifiber tows with a cloth architecture of 16 x 16 tows per inch and laid up with 100 cloth layers per inch (approximate 40 vol.% fibers). A 3-D braid architecture specimen used SiC/SiC as well with Nicalon fibers. Of current interest is the nondestructive measurement of axial and radial density gradients, as well as measurement of fiber orientation. To map density variations and determine fiber orientation we are working on 3-D microfocus X-ray computed tomography (XCT) and advanced image processing technology. Of interest also is the detailed chemistry of the fiber surface, as well as that of the fiber/matrix interface after infiltration. In this case, we are developing multi-nuclear solids nuclear magnetic resonance technology. For the evaluation of fracture behavior, effort has concentrated on continuous fiber reinforced ceramic matrix composites (CFCCs). High strength of fibers and weak interfaces are requisites for "tough" ceramic composites.[1,2] Strength and toughness of composites are greatly influenced by the strength properties of the reinforcements. These reinforcements are susceptible to thermal degradation and surface defects or to damage introduced during composite fabrication, thereby contributing to strength degradation. Therefore, it is important to establish factors that lead to strength degradation during fabrication of the composites. Specifically, in the present study, we have evaluated the effects of processing on flaw generation and resulting strength degradation of Nicalon fibers in continuous SiC-fiber–reinforced SiC matrix composites.

CHARACTERIZATION OF CVI-PRODUCED SiC/SiC CONTINUOUS FIBER COMPOSITES BY X-RAY MICROFOCUS CT

Detection of Fiber Orientation

One parameter of interest for CFCCs with multidirectional layups is the relative angle of the fiber tow between lay-ups. This is important because mechanical properties are known to be affected by misoriented fiber tows. At present, little theoretical information is available about the effects of fiber orientation on mechanical properties, but current information suggests that ±5° will have an impact.

Recent theoretical work by Buesking[3] has shown that the impact of fiber orientation on modulus is significantly dependent upon the origin of the fibers, even if the same infiltration process method (CVI) is used. Fig. 1 shows one theoretical predicted effect of fiber orientation on elastic modulus. One sees that for one fiber type, a 5° off axis orientation can change elastic modulus by ≈10%. For the second fiber type, there is essentially no effect.

We have previously reported 4 on the development of X-ray computed tomographic imaging together with image processing to nondestructively measure in-plane fiber orientations. The prior work was limited to 2D work. We have extended this work to 3-D analysis. Now all work this year has however used simulated data. Figure 2 shows a schematic diagram of a 3-D weave architecture being developed for tube type applications and also shows the 3-D weave geometry we have simulated. We have generated the 3-D FFT image shown in Fig. 3.

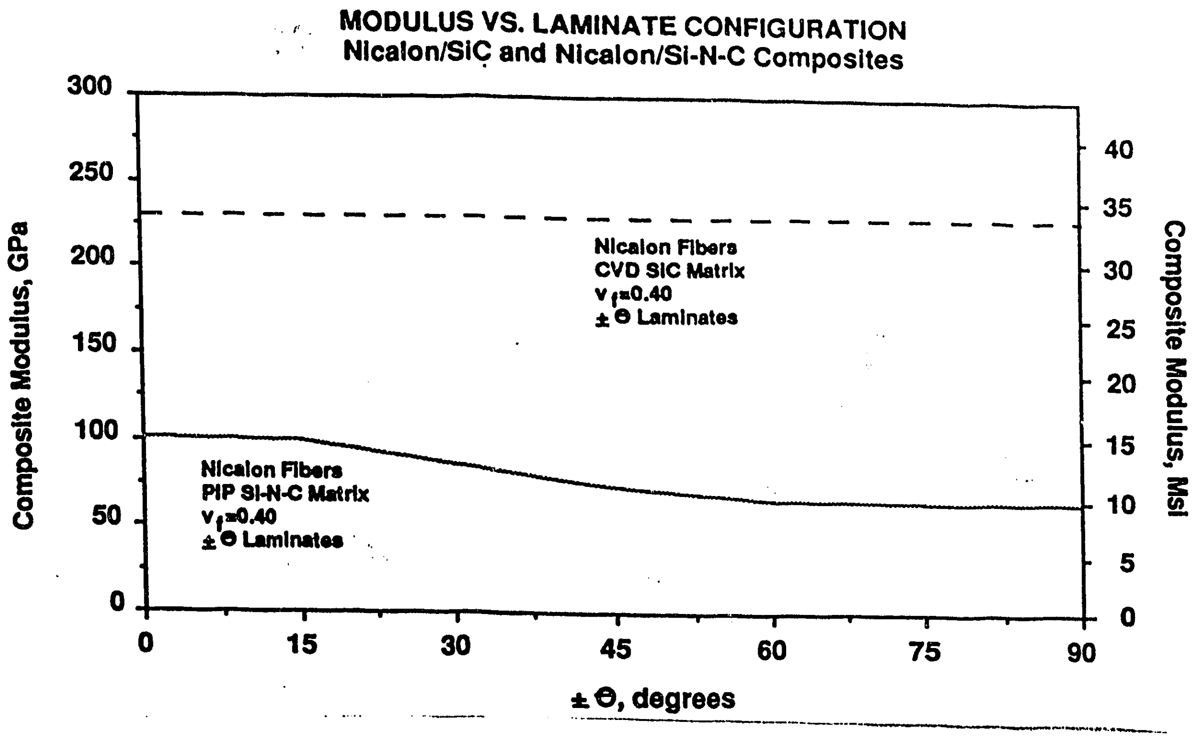


Fig. 1. Theoretically calculated effect of fiber orientation on 2D laminates of SiC/SiC. (from Buesking[3]).

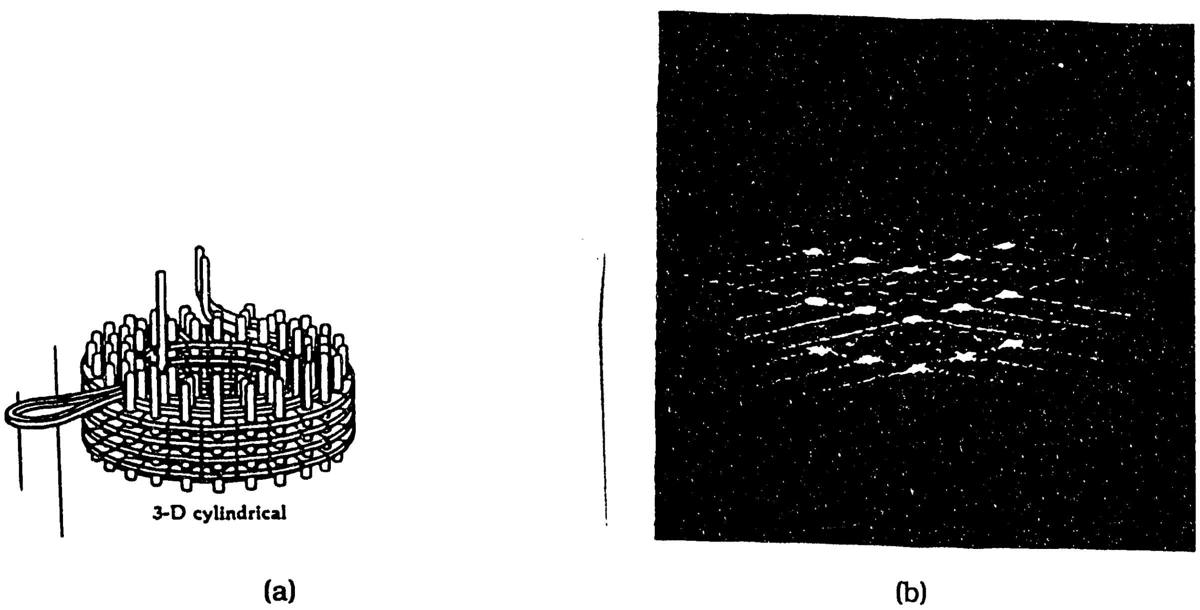


Fig. 2. 3-D weave geometries for CFCC materials. (a) schematic diagram showing 3-D weave geometry for a cylindrical component, (b) simulated data showing 3-D weave simulation for 3-D fiber orientation analysis.

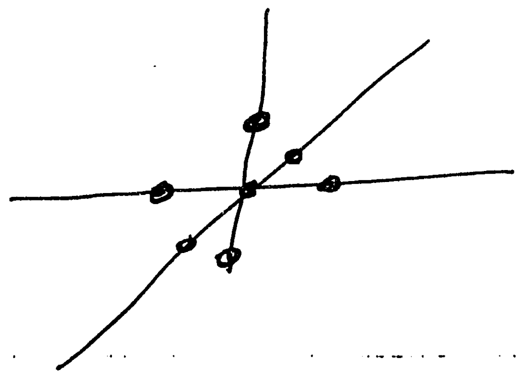


Fig. 3. 3-D FFT image of the simulated data shown in Fig. 2.

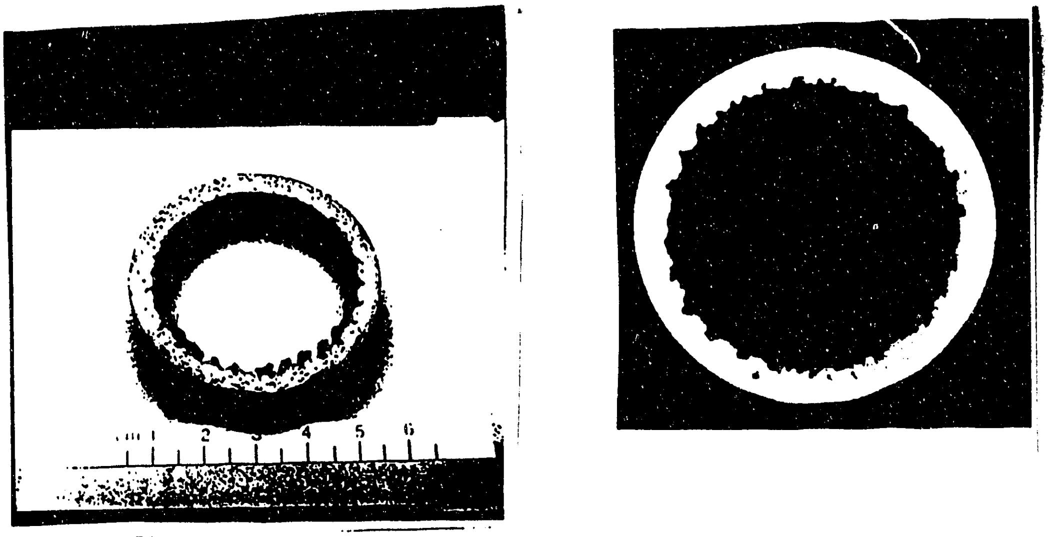
What these results show is that if the X-ray CT image data can discrimate between fiber tows and the matrix, then the 3-D dimensionality of the fiber tows can be characterized.

We have also examined actual CVI SiC/SiC 3-D weave architecture's by X-ray CT. One image is shown in Fig. 4. One can see from the optical photo that there are significant voids through the wall thickness and that there are significant variations in the inside wall. The resulting X-ray tomographic image shows many of these features to the 100 μm scale easily.

Quantification of Feature Volumes

As part of the NDE development activities we began an ieffort to quantify the volumes of the features we detect by three-dimensional X-ray microfocus tomography. We need to quantify these feature volumes so that the data can be used in fracture mechanics models for predicting fracture behavior . We used our new SUN SPARC II/GS work station for all of this analytical work.

Determining feature size was a three-stage process that required finding the feature, determining the outer boundary of the feature to be examined (edge detection), and calculating the size from a volume whose edges are known.



(a) (b)

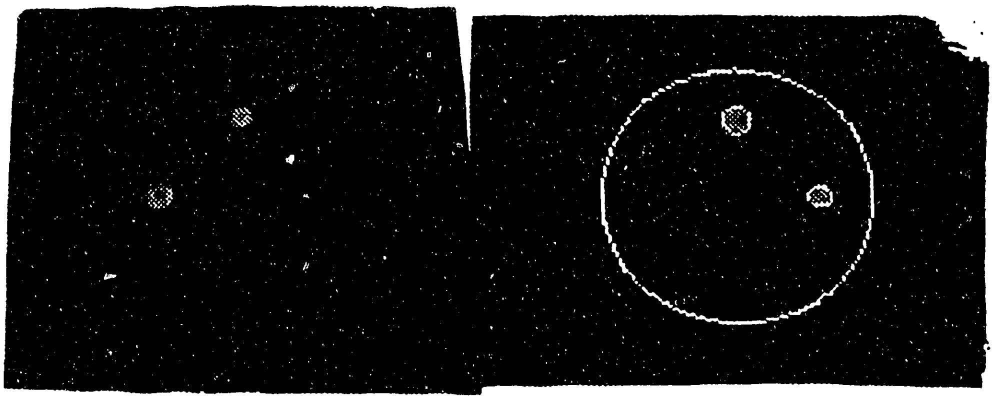
Fig. 4. 3-D weave architecture (a) photograph of section used for study b) X-ray tomographic image of one cross-section of the 3-D weave. (note voids)

To find the location of the features of interest, three approaches were considered: (1) train a neural network to learn the characteristics of the object under study and then scan the object for anomalies, (2) determine a multi-dimensional characteristic vector for the solid object and use a rule-based approach to determine features, and (3) use a gradient field of the attenuation to help locate anomalies.

Use of the gradient was chosen, because it afforded a direct approach to locating anomalies. This approach was very susceptible to a noisy system and some thought was given to methods of smoothing the data before proceeding. The smoothing techniques were not used because the computational time proved significant, while not improving the performance significantly.

The information used in this initial image processing work came from two sources: real data from XR-CT images of ceramic objects, and synthesized data.

Excellent results in tracing the outer edge of features was achieved (see Fig. 5). Individual closed areas were obtained on separate "slices" of the original data. The algorithm then combined slice information by correlating the characteristics and the location of each slice. The volumes were then obtained by summing over the correlated voxels of all slices.



(a) (b)

Fig. 5. X-R-CT-reconstructed image from synthesized data to show how gradient edge detection algorithm determines edges of two simulated voids; (a) data as observed, (b) white outlines show detected edges.

CHARACTERIZATION OF CONTINUOUS SiC FIBER/SiC MATRIX COMPOSITES BY FRACTURE STUDIES

High strength of fibers or whiskers and weak interfaces are requisites for "tough" ceramic composites.[1] The strength and toughness of composites are greatly influenced by the mechanical properties, especially the strength of reinforcements[2,5]. These reinforcements are susceptible to thermal degradation and mechanical damage introduced during fabrication of composites therby contributing to strength degradation. Therefore, it is important to establish factors that lead to strength degradation during fabrication of the composites. We have evaluated the effects of processing on flaw generation and resulting strength degradation of Nicalon fibers in a continuous SiC fiber-reinforced SiC matrix composites. Also, Si_3N_4 whisker-reinforced Si_3N_4 composites were fabricated and their mechanical properties were measured to evaluate the change in bulk flaw population during processing.

Specimens for Fracture Studies

Nicalon-fiber-reinforced SiC matrix composites fabricated by chemical vapor infiltration (CVI) obtained from Oak Ridge National Laboratory were used for the evaluation of the effects of processing methods on flaw generation and resulting strength degradation in composites As-received composites were close to 90% dense. Fabrication and mechanical properties of the composite specimens are described elsewhere.[6]

To evaluate the change in bulk flaw population during densification of whisker composites, the powders of monolithic Si_3N_4 (~0.3 μm) and its composites with 15 vol.% Si_3N_4 whiskers (0.6 μm in diameter and 45 μm in length) were prepared by a conventional ceramic powder processing route.[7] A set of 60 specimens (≈12.7 mm in diameter and ≈3.2 mm thick) each of 0 and 15 vol.% Si_3N_4 whisker-reinforced Si_3N_4 matrix composite were cold-pressed and bisque-fired in our laboratory. Subsequently, half of each set of specimens were hot isostatically pressed (HIPed) at Allied-Signal Aerospace Company, Garrett Processing Division, CA.

Mechanical and Physical Properties Evaluation and their Correlation with Critical Flaws

A. Nicalon fiber-SiC Matrix Composites

Single-fiber strength distribution of as-fabricated Nicalon fibers was obtained from bundle tests.[8] Bundle tests were conducted on a universal testing system on fiber tows with gage lengths ranging from 27 to 100 mm under ambient conditions and at a loading rate of 0.5 mm/min. Weibull parameters were obtained from the load vs. strain plots following the procedure described by Chi et al.[9] Weibull distribution used to describe fiber fracture strengths was as follows:

$$F(\sigma) = 1 - \exp\left[-\frac{L}{L_o}\left(\frac{\sigma}{\sigma_o}\right)^m \right] \tag{1}$$

In this representation, F(s) is the failure probability at an applied stress s, L_o is the fiber gage length at which Weibull parameters are estimated, L is the standard gage length taken to be 10 mm, s_o is the scale parameter signifying a characteristic strength of the distribution, and m is the Weibull modulus. Results based on seven bundle tests gave an average value for Weibull modulus and scale parameter as 7.1 and 3.45 GPa, respectively at a standard gage length of 10 mm. These results were presented at the Sixth Annual Conference on Fossil Energy Materials.[4]

The strength of the fibers in composites was evaluated from characteristics fracture features of the fibers. To this end, continuous Nicalon-fiber-reinforced SiC composite specimens (2.9 x 4.2 x 51.0 mm) were fractured in a four-point-bend mode on a universal testing system. Strength of fractured fibers was determined from the measured values of fracture mirror radii as shown in Fig. 6 and an empirical relationship.[9] As reported previously[8], values of scale parameter and Weibull parameter evaluated were 2.3 GPa and 6.0, respectively. For convenience, earlier it was assumed that the gage length of fractured fibers in composites was equal to the standard gage length of 10 mm. But, gage length of fibers over which uniform stress acts in composites is expected to be much smaller, on the order of fiber pullout lengths. Therefore, for a reliable prediction of strength distribution, an accurate evaluation of gage length, L_o, is needed which can be used for better estimation of s_0.

To this effect, scanning electron microscopy (SEM) of fractured composites was used to determine fiber pullout length distribution (as shown in Fig. 7) from which the gage length of fractured fibers was determined. Fiber gage length was taken to be twice the average pullout length and was found to be 340 mm. Using this value of fiber gage length, the value of s_0 was estimated to be 1.31 GPa.

The value of the scale parameter obtained above was incorporated in Eq. 1 to determine the fiber strength distribution of Nicalon fibers in composites and compared with as-fabricated fiber strength distribution shown in Fig. 8. Although the Weibull modulus in the two cases remained same (7.1 for the as-fabricated and 6.0 for the fibers in composites), the strength of the fibers incorporated in the composite was approximately 50% lower than that of the as–fabricated fibers. This reduction in fiber strength may have been caused either by the increase in the severity of pre–existing flaws or by the introduction of new flaws during processing. This loss of strength is attributed to microstructural and stochiometric changes that occur in the fibers at elevated temperatures.[11]

B. Si_3N_4(w)/Si_3N_4 Composites

Average density of green (cold pressed and bisque–fired) monolithic and composite disk specimens were 1.52 and 1.60 g/cm^3, respectively. Monolithic and composite bar specimens received after HIPing had a density of 3.14 and 3.16 g/cm^3, respectively, measured using Archimedes method. Phase content of HIPed monolithic specimens determined by X–ray diffraction indicated close to 70% of b phase in both monolithic and composite specimens. Modulus of elasticity was 309 and 303 GPa for monolithic and composite specimens, respectively.

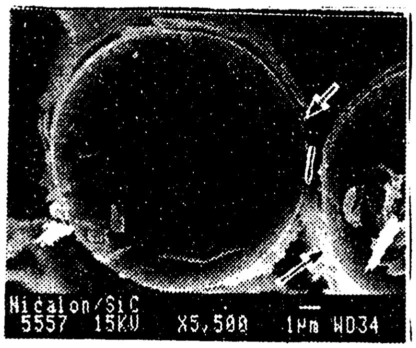


Fig. 6. Micrograph of Fractured Nicalon Fibers in SiC Matrix Composite Showing Characteristic Fracture Features.

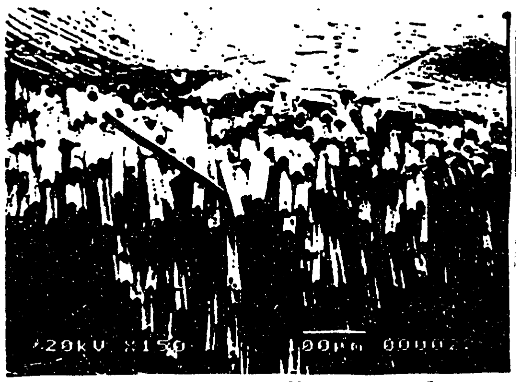


Fig. 7. Fiber Pullout Length Distribution for Nicalon Fiber-Reinforced SiC Matrix Composites.

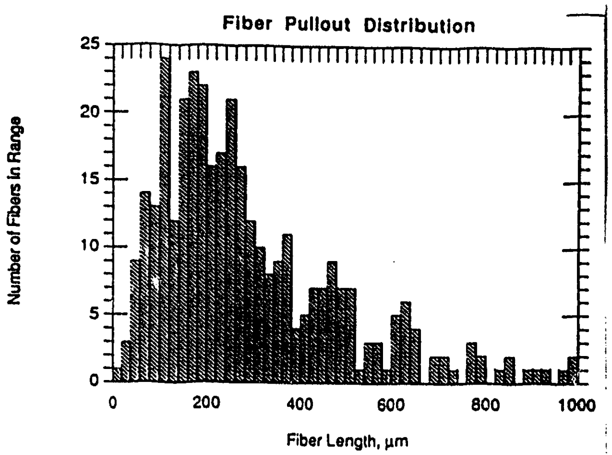


Fig. 8. Weibull strength distribution of Nicalon fibers in as-fabricated state (bundle Ttst) and after processing (mirror size evaluation).

The tensile strength of green and HIPed disk specimens were measured in a diametral compression mode. The HIPed specimens were tested in as-fabricated condition at a crosshead speed of 1.27 mm/min in ambient conditions. The Weibull strength distribution plots were constructed for both monolithic and composites in green and densified states. The Weibull moduli were evaluated from the slopes of the strength versus probability of failure plots on a logarithmic scale. The average strengths obtained from diametral testing of green monolithic Si_3N_4 and its composites disks were 1.8 MPa and 2.8 MPa, respectively. The corresponding values for Weibull moduli were 4.4 and 8.1. The average strengths of the HIPed disk monolithic and composite specimens were 304 MPa and 272 MPa and the Weibull moduli were 4.0 and 4.9, respectively.

In diametral-compression tests, large volume of the disk specimen is under tensile stresses, thereby promoting failure from internal or bulk flaws. This was confirmed by the observation of critical flaws in the specimen bulk. Failure was predominantly found to occur at inclusions, voids, agglomerates, etc. The strength data for disk specimens show some interesting trends. First, Weibull modulus of the green composite specimens is almost twice of that of monolithic specimens. This implies that bulk flaws in the green state are controlled by the presence of whiskers in the composites. It is possible that in green composites presence of whiskers cause toughening by such mechanisms as crack pinning and deflection. These whisker-toughening effects narrow the strength-controlling flaw size distribution thereby decreasing the variability in strengths and increasing Weibull modulus and flaw tolerance of composites.

Second, after HIPing, monolithic and composite disk specimens show similar strength distributions. This suggests that after HIPing the strength controlling bulk flaws are similar in the two cases. This is further confirmed by similar average values of the fracture strengths of densified monolithic and composite specimens. However, reduction in the Weibull modulus from 8.1 in green state to 4.9 in dense composites indicates the absence of the toughening effects present in green state. This is probably due to the development of flaws resulting from the difficulties associated with uniform distribution of whiskers during processing of composites, whisker degradation, and degradation of whisker/matrix interface.

REFERENCES

1. A. G. Evans and D. B. Marshall, "The Mechanical Behavior of Ceramic Matrix Composites," Overview No. 85, Acta Metall., **37** [10] 2567–2583 (1989).

2. T. Mah, M. G. Mendiratta, A. P. Katz, R. Ruh, and K. S. Mazdiyasni, "Room Temperature Mechanical Behavior of Fiber-Reinforced Ceramic Matrix Composites," J. Am. Ceram. Soc., **68** [1] C-27 – C-30 (1985).

3. K. W. Buesking, "Effects of Weave Geometry on Properties of Nicalon Reinforced Ceramic Matrix Composites," presented at 1993 Am. Cer: Soc. Mtg. Cocoa, Beach, FL 19993. (in press)

4. W. A. Ellingson, J. P. Singh, D. L. Holloway, S. L. Dieckman, D. Singh, E. A. Sivers, S. H. Sheen, and M. J. Wheeler, "Development of Nondestructive Evaluations Methods and Prediction of Effects of Flaws on the Fracture Behavior of Structural Ceramics", Proc. of the Sixth Annual Conf. on Fossil Energy Materials, Fossil Energy AR&TD Materials Program, ORNL/FMP-92/1, pp. 119-130 (1992).

5. S. M. Smith, R. O. Scattergood, J. P. Singh, and K. Karasek, "Effect of Silica and Processing Environment on the Toughness of Alumina Composites," J. Am. Ceram. Soc., **72** [7] 1252–55 (1989).

6. D. P. Stinton, A. J. Caputo, and R. A. Lowden, "Synthesis of Fiber-Reinforced SiC Composites by Chemical Vapor Infiltration," Am. Ceram. Bull., **65** [2] 347–350 (1986).

7. C.Y. Chu and J.P. Singh, "Mechanical Properties and Microstructure of Si3N4–Whisker-Reinforced Matrix Composites," Proceedings of 14th Annual Conference on Composites and Advanced Ceramics, Cocoa Beach, FL, Jan. 14–17 (1990).

8. P. W. Manders and T. W. Chou, "Variability of Carbon and Glass Fibers, and the Strength of Aligned Composites," J. Reinf. Plastic Composites, **2** 43–59 (1983).

9. Z. Chi, T-Wei Chou, and G. Shen, "Determination of Single Fiber Strength Distribution From Fiber Bundle Testings," J. Mater. Sci., **19** [10] 3319–3324 (1984).

10. H. P. Kirchner and R. M. Gruver, "Fracture Mirror in Alumina Ceramics," Phil. Mag. **27** 1433–1446 (1973).

11. T. J. Clark, R. M. Arons, and J. B. Stamatoff. " Thermal Degradation of Nicalon SiC Fibers," Ceram. Eng. Sci. and Proc., **6** [7–8] 576–588 (1985).

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