CORRELATION BETWEEN STRUCTURE AND PROPERTIES OF CHOPPED-FIBER CARBON-CARBON COMPOSITES

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Introduction

Structure-property relationships were studied in random chopped-fiber, carbon-carbon composites. Flexural strength, strain-to-failure, and modulus were correlated with graphite crystallite size and lattice strain as affected by processing variations.

Experimental Method

Disc-shaped specimens were prepared from chopped, copolymer PAN-based fibers and 20- to 80-weight-percent high-char yield, p-polyphenylene-type, graphitizing resins, by dispersing the fibers in a solution of the resin, partially curing, attritioning, molding, and completely curing followed by pyrolysis to 1000°C (Ref. 1) and graphitization for 8 or 16 hours at 2600°C. Fibers were made from the oxidized precursor by heating to 1800°C (PAN-1800) or 2600°C (PAN-2600). Flexural properties were measured on a four-point bend device using specimens with a 10:1 length:thickness ratio strain gaged on the tension surface. Crystallite variations were determined from x-ray diffraction line profiles.

Experimental Results

Data on the effect of matrix concentration for PAN-1800 composites heat-treated 16 hours at 2600°C are plotted in Figure 1. Flexural strength reaches a maximum at an initial matrix concentration of 50 to 60% (Curve A). Similarly, strain-to-failure reaches a maximum at an initial matrix concentration of 60% (Curve B). The best combination of strength, 10,260 psi, and strain-to-failure, 0.60%, are obtained when the matrix concentration is 60%.

Discussion of Results

Flexural behavior was reconciled by analyzing x-ray diffraction-line broadening and the stresses developed by matrix contraction during carbonization. The former provides an indication of the crystallite size and strain distribution, whereas the latter provides a possible mechanism for the graphitization trends.

- X-ray line-broadening analyses of all samples show a linear relationship between matrix content and either crystallite size or lattice strain after carbonization and graphitization, as shown in Figure 2. A linear relationship exists in the range of matrix concentrations from 35 to 80%. Size and strain broadening occur simultaneously but separation was not possible with the simplified analysis used with most samples. For a limited number of samples, a more detailed calculation was made using the change in broadening with diffraction order and more precise corrections (Refs. 2 and 3). These results revealed that both factors are present, but mean crystallite size depends more upon the matrix concentration than does the strain parameter. The latter was thus disregarded.

- Crystallinity and mechanical behavior correlate well for all samples. Figure 3 shows that both the ultimate-flexural strength (F^{FU}) and strain (ϵ^{FU}) increase with crystallite size. This increase demonstrates the importance of lattice perfection for producing a composite with good mechanical properties, and together with preceding data shows that the best results are attained with an initial matrix precursor concentration of about 60 weight percent.

- Mechanisms responsible for the crystallinity changes with matrix content appear to be related to residual stresses in the matrix, induced by contraction of the matrix during carbonization. A theoretical analysis, assuming that each fiber is encased in a cylindrical sheath of matrix, shows that increasing the matrix content (sheath thickness) causes the hoop, radial, and longitudinal matrix-stresses to increase as do the hydrostatic stresses in the fiber. Microscopic evidence of matrix graphitization strongly suggests that this is where most of the observed effects are taking place. Also, since a nongraphitic, amorphous (glasslike carbon) matrix would be brittle regardless of fiber properties, improved mechanical properties would not be expected from crystalline changes in the fiber alone.

Conclusions

The flexural strength and strain-to-failure of random short-fiber, carbon-carbon composites are directly proportional to the matrix content (and are not the fiber content, as might be expected) in the range from 35 to 60 weight percent of initial precursor, and this results from stress-induced graphitization of the matrix that reduces its brittle behavior by increasing the crystallite size, by reducing lattice strains, or by both.

References

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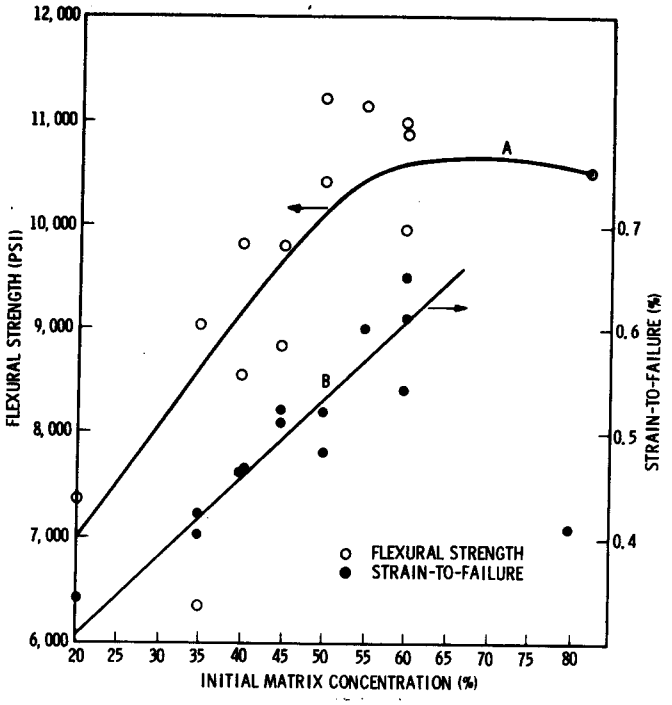

Fig. 1 Flexure Strength and Strain-to-Failure Versus Matrix Concentration for Pan 1800 Heat-Treated to 2600°C for 16 Hours

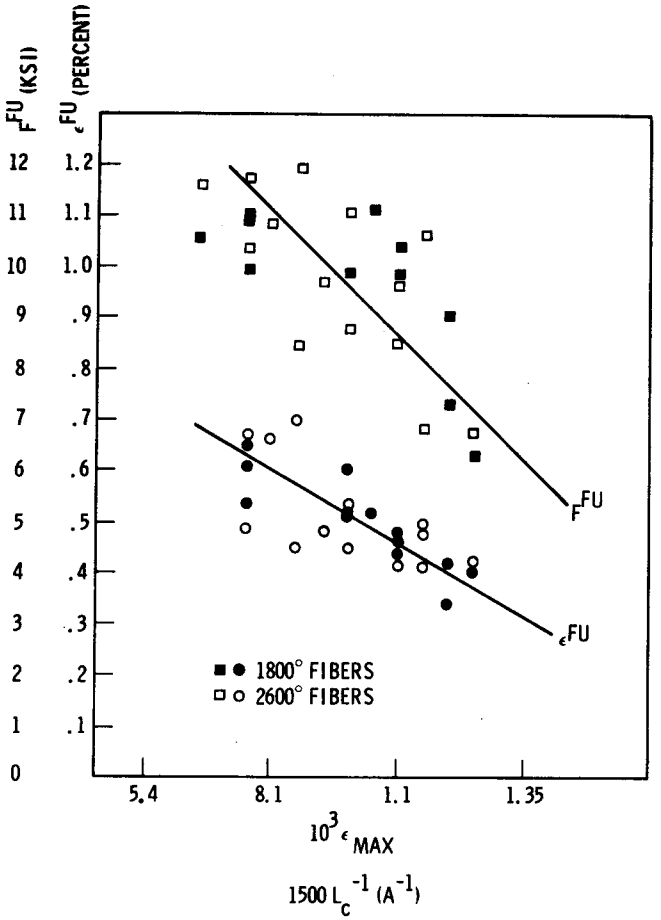

Fig. 3 Cumulative Plot of Flexural Strength (F^{FU}) and Strain-to-Failure (ϵ^{FU}) Versus Maximum Lattice Strain and/or Reciprocal Crystallite Size for Carbon-Carbon Random Fiber Composites With Initial Matrix Content Ranging From 30 to 60 Weight Percent

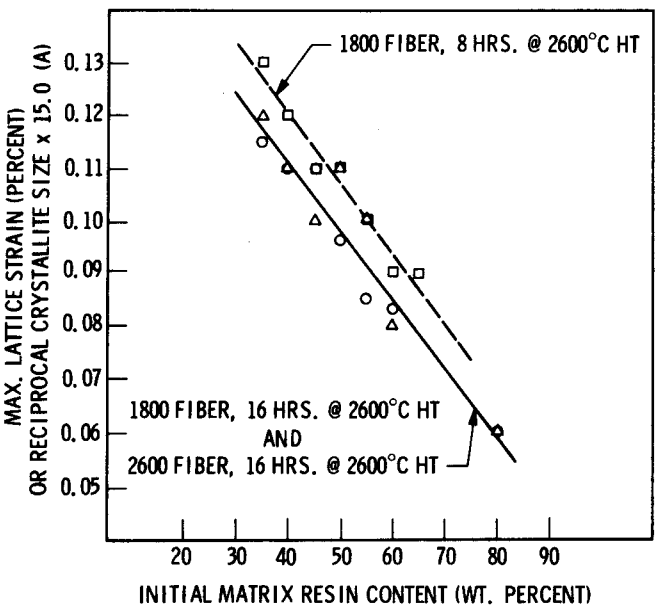

Fig. 2 Crystallographic Lattice Strain Normal to Basal Planes and/or Mean Reciprocal Crystallite Size in That Direction for Carbon-Carbon Composites With Initial Matrix Resin Content in Range From 30 to 80 Weight Percent