EFFECTS OF OXIDATION ON THE STRENGTH OF C/C COMPOSITES FOR GT-MHR CONTROL RODS

J. P. Strizak

Oak Ridge National Laboratory
Post Office Box 2008
Oak Ridge, Tennessee 37831-6088, U.S.A.

INTRODUCTION

Carbon-carbon (C/C) composite materials are being considered for a high temperature control rod design for the Gas-Turbine Modular Helium Reactor (GT-MHR). Control rod materials are exposed to high temperature, neutron irradiation, and helium with its impurities (O_2 + H_2O + CO + CO_2). During certain operating events the peak temperature in particular areas of the reactor core can exceed 1300°C, sufficiently high to preclude the insertion of the reference metallic control rods into those areas. Thus the desire to develop high temperature control rods to increase the operating margin of the GT-MHR.

A review [1] of the properties of carbon materials suitable for high-temperature control rods identified several potential graphites. On the other hand, C/C composites offer higher strength and stiffness and perhaps, most significantly, increased fracture toughness.

A conceptual C/C control rod has been designed [2] and prototypic components have been manufactured using existing carbon composite fabrication technology methods. The composite materials employed for the prototype were selected to maximize tolerance to neutron damage and to minimize irradiation-induced dimensional changes. However, no data for the selected materials were available on the effects of oxidation nor the potential loss in strength due to oxidation. Preliminary data on the effects of oxidation on strength (apparent interlaminar shear strength and flexural strength) of the candidate C/C composite materials for high-temperature control rods have recently been obtained.

EXPERIMENTAL

The reference GT-MHR control rod design consists of a number of tubular elements (metal canisters) strung on a central metallic spine. Each canister contains neutron absorbing boronated graphite for controlling reactor activity. The conceptual high temperature control rod in Fig. 1 is comprised of three components, all manufactured from C/C composites. The tube elements have a multilayer braided architecture with an integral partial tube closure forming a smooth, spherical radius, and local reinforcement with radial fiber bundles at the open end of the tube for thread strength. The swivel and connector were machined from a block of orthogonal three-directional (3D) C/C composite. To optimize the tolerance of the control rod components to neutron damage and minimize irradiation-induced dimensional changes, pitch fibers and pitch impregnation were selected.

Short beams were machined from the 3D orthogonal C/C block to determine the apparent interlaminar shear strength. Radial sections were machined from the central portion of a control rod tube. The thickness of the beams was equivalent to the thickness of the tube wall and the beam length was parallel to the longitudinal axis of the tube element. Two beam lengths were tested to determine apparent interlaminar shear and flexure strengths. To evaluate the effects of oxidation, the test specimens were exposed to water vapor in helium at 800°C to induce mass losses up to ~5 wt %.

DISCUSSION

Above 10% burnoff the activation energy for the oxidation process in the 3D orthogonal composite was much higher. The associated decrease in oxidation rate was attributed to differences in reactivity between fibers and impregnant. Optical analyses indicated that the pitch fibers were much less reactive than the pitch impregnant. Below 10% burnoff oxidation of the 3D orthogonal composite appeared to correspond to the selective removal of the impregnant. The decrease in density of the composite closely followed the percentage burnoff, and there was no noticeable decrease in geometric volume, suggesting that oxidation through the interior of the test specimens was uniform.

The two control rod composite materials behaved somewhat differently. The impregnant of the braided tube appeared to be less accessible to oxidize than that in the more open 3D orthogonal architecture. Oxidation rates were somewhat lower for the braided architecture. Further, the activation energy after 5% burnoff was nearly equal to that for the 3D orthogonal composite after 10% burnoff.

The deleterious effects of air oxidation on the mechanical properties of graphites [3-5] as well as chopped fiber C/C composites [6] has been demonstrated. Although fibers dominate the mechanical properties of C/C composites, the role of the matrix (impregnant) is not insignificant. The matrix can affect efficient load transfer between fibers. In this study, the selective oxidation of the impregnant resulted in a decrease in the apparent interlaminar shear strength of the orthogonal 3D composite (Fig. 2). Oxidation to 5% burnoff resulted in an ~13% reduction in shear strength.

Multilayer braiding readily lent itself to producing the tubular structure for the prototype control rod. The braided architecture incorporated axially aligned fibers in each layer, producing a pseudo 3D product. The circumferential-to-axial fiber distribution was high, 90%. The tube construction did not contain through-thickness fiber bundles to counteract interlaminar weakness (except in the open end of the tube). Oxidation of the pitch matrix further decreased the apparent interlaminar shear strength (Fig. 3). The flexural strength, on the other hand, was strongly influenced by the architecture. Oxidation of the pitch impregnant appeared to have little effect on flexure strength (Fig. 3). (Data scatter was attributed to severed axial fibers when the control rod tube was machined to the required outside diameter.)

The successful development of high temperature C/C composite control rods for the GT-MHR will require an integrated approach to the selection of precursor and matrix materials to optimize irradiation resistance, tailoring composite architectures to meet mechanical and thermal property requirements, determining the effects of oxidation, and the selection of coatings to control oxidation resistance.

REFERENCES

1. T. D. Burchell, A Review of the Properties of Carbon Materials Suitable for MHTGR High-Temperature Control Rods, ORNL/NPR-90/14, June 1991.

2. T. D. Burchell and J. P. Strizak, Technical Plan: Development of Carbon-Carbon Composite Materials for High-Temperature NP-MHTGR Control Rods, ORNL/NPR-92/6, February 1992.

3. J. L. Wood, J. X. Zhoa, R. C. Bradt, and P. L. Walker, Jr., "The Effect of Oxidation on the Flexural Strength of Graphite," CARBON, Vol. 19, 1981, pp. 61-62.

4. P. A. Thrower, J. C. Bognet, and G. K. Matthew, "The Influence of Oxidation on the Structure of Graphite-I," CARBON, Vol. 20, 1982, pp. 457-464.

5. P. A. Thrower, G. K. Matthew, and N. J. McGinnis, "The Influence of Oxidation on the Structure of Graphite-II," CARBON, Vol. 20, 1982, pp. 465-471.

6. J. X. Zhao, R. C. Bradt, and P. L. Walker, Jr. "Effect of Air Oxidation at 873 K on the Mechanical Properties of a Carbon-Carbon Composite," CARBON, Vol. 23, No. 1, pp. 9-13, 1985.

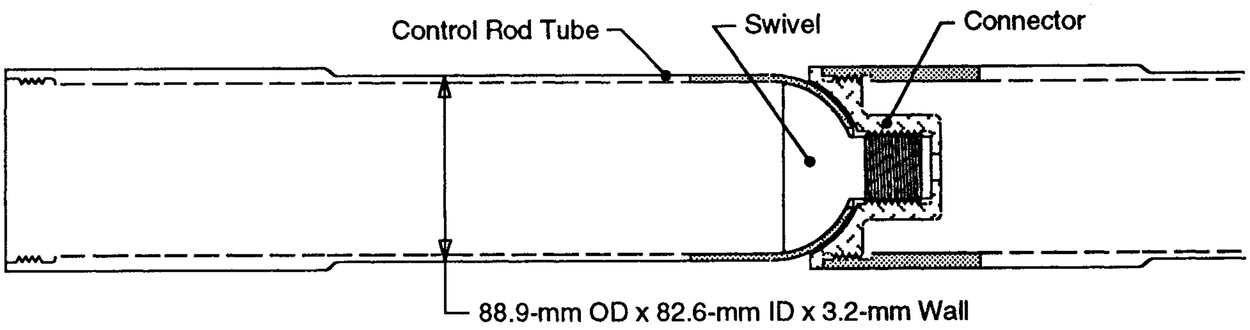

Fig. 1. Prototype C/C control rod configuration.

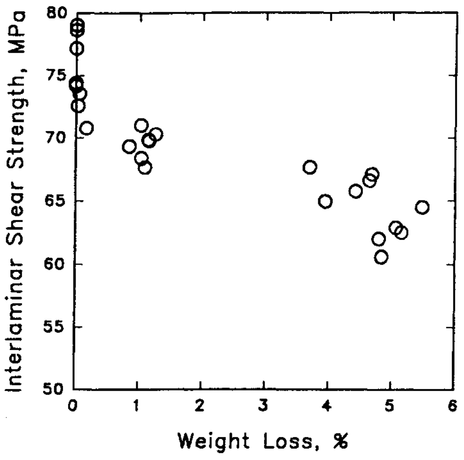

Fig. 2. Strength of 3D orthogonal composite.

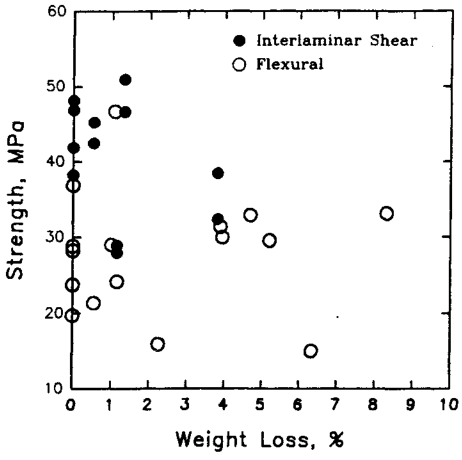

Fig. 3. Strength of braided composite tube.