STRENGTH TESTING OF MONOLITHIC AND DUPLEX SILICON CARBIDE CYLINDERS IN SUPPORT OF USE AS NUCLEAR FUEL CLADDING

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ABSTRACT

An important aspect of nuclear fuel rod cladding is its ability to withstand, throughout its lifetime, internal pressure loads during steady-state operation, transients, and accidents. Consideration is now being given to the use of silicon carbide duplex tubes as a replacement for the existing use of zirconium alloys. This project investigated the mechanical response of various silicon carbide tubular structures when subjected to internal pressurization using an expanding internal plug method. According to this test method, an incompressible polyurethane plug, fitted inside the test specimen, is subjected to axial compressive loading, which results in the internal pressurization of the test specimen.

Monolithic tubes manufactured were tested along with a series of fiber-reinforced composite tubular test specimens and duplex tubes comprised of a monolithic tube that is surrounded by a fiber-reinforced composite. The test results showed that:

- The monolith test specimens failed at about 0.2% tangential strain and the values of Young's modulus obtained were consistent with those published in the literature.
- The duplex test specimens also failed at about the same strain when the inner monolithic SiC failed. However, they exhibited a “graceful” mode of failure (no brittle fracture of the duplex test specimen) with the composite layer holding the structure in one piece.
- One set of composite-only test specimens exhibited a load-carrying capability lower than that of the monolithic SiC tubes, and tended to fracture in several pieces at low strain values.

INTRODUCTION

One important aspect of fuel rod cladding is its ability to withstand internal loads. Internal loads can arise from a pressure gradient between the inside of the cladding and the external environment. Internal pressure sources include the pre-fill gas pressure (Helium) and the accumulation of fission gases over the life of the fuel rod. Swelling of the fuel over long periods of operation may also provide a source of pressure loads on the cladding. External pressure, at power operation, is usually higher than the internal pressure. However, during certain transient and accident scenarios, there may be a positive gradient from the inside to the outside, which results in the tensile tangential loading of the cladding.

Another potential load arises from the reactivity-initiated accident, wherein the fuel pellet expands quickly due to the deposition of energy from the accident. The expansion could include both thermal expansion as a solid fuel pellet and expansion due to melting of some of the fuel. These various loads are postulated in the safety review of the fuel design [1], and thus it is necessary to know the cladding response.

Alloys of zirconium have been the cladding material of choice for most of the water-cooled reactors for almost half a century. These alloys have some common and desirable attributes, including:

- relatively inexpensive;
- generally compatible with operating environment;
- compatible with uranium dioxide
- strong;
- abundant.
- Relatively low neutron cross-section

However, as reactors proceed to ever-higher burnup levels, some problems with Zr have become apparent. The prolonged use in water reactor environment has produced surface corrosion to the extent that zirconium alloys interact less favorably during the postulated loss of coolant accident [2]. Further, test results are now showing that the consequences of the postulated reactivity insertion accident may be less favorable than previously thought, for these prolonged operating exposures. Although these negative factors may not absolutely preclude the use of Zr alloys for very high fuel burnup scenarios, there is nonetheless some interest in alternate materials for fuel cladding. For this reason, there are efforts to explore the use of various forms of Silicon Carbide (SiC) as a replacement for Zr tubing. SiC is an ideal candidate material for these applications because of its excellent mechanical, chemical and thermal properties, its ability to retain strength at elevated temperatures and most importantly its low neutron absorption cross-section.

One important aspect of programs to develop and qualify cladding materials is focused on determining their strength, which is the subject that concerns this report. The mechanical testing of the various SiC test specimens described in this report constitutes the first exploratory step in the direction of qualifying SiC for use as nuclear fuel cladding in a fission reactor. Several types of SiC tubes were fabricated for the purpose of testing the structural response to internal pressure. The test geometries include:

- Monolithic cylinders of Silicon Carbide
- Duplex test specimens, consisting of an inner monolith cylinder and a continuous SiC fiber-reinforced SiC matrix composite overlay.
- A continuous fiber-reinforced composite structure alone, with no inner monolithic cylinder

The general purpose of this test program was to determine the stress-strain relationships when the tubular structures are subjected to internal pressurization. Of interest was not only the stress and strain at failure up to the failure of the monolith (and the failure strain is quite small for the monolith) but also the behavior of the duplex test specimens after the initial failure of the interior monolith cylinder. The next phases of this study will include material properties at elevated temperatures, and the effects of radiation, and exposure to PWR water chemistry, on material properties. This paper will discuss some of the test results; a full description of the test specimens and test results is contained in reference 3.

TEST PROCEDURE

Tubular test specimens (monolithic, composite and duplex) were subjected to internal pressurization at ambient conditions according to the expanded plug test method [4]. The tests were carried-out using an electromechanical testing machine (MTS Model Alliance RT-50) and a test fixture consisting of a platen and two concentric pistons (Figure 1). The radial deformation of the test specimens was measured using a pair of capacitance proximity probes with a 0.8 mm range (Capacitec Mod HTP-75). Because these measurements require a conductive surface, it was necessary to apply a thin coating of chromium paint on the outer surface of the test specimens

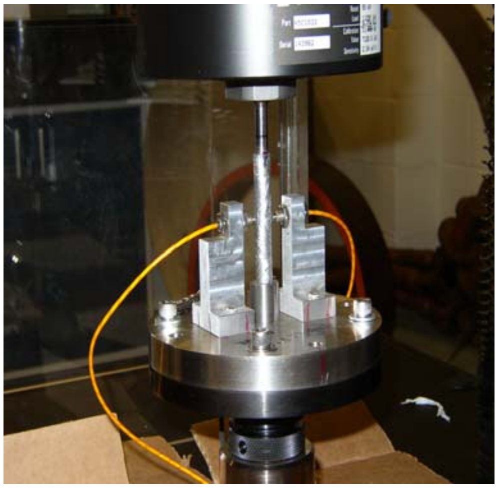

Figure 1. Test set-up showing the test specimen, the loading pistons and the proximity gages

Plugs of incompressible polyurethane with a shore hardness of A95 were machined to a length of 12.5 mm and to a diameter slightly smaller than the inner diameter of the test specimen. The plug was mounted on the base pedestal and had a dowel interface to assure alignment with the piston. An initial small load was placed on the assembly to assure that the plug and specimen were centered and seated onto the base. The tests were performed under a constant crosshead displacement rate (0.017 mm/s). Tests on monolith specimens were terminated at the time of failure; however, for the duplex specimens, the crosshead was driven further until manually stopped (usually when the expanding specimen came close to the proximity gages). The test specimens were cut to a length of 63.5 mm using an instrumented saw equipped with a diamond blade.

Several types of SiC specimens were processed and tested as described below:

- Two monolithic tubes of SiC were tested. One was sintered α-SiC made by St. Gobain[1]. The second tube was CVD β-SiC made via CVD by TREX Enterprises[2].
- Three duplex tubes, identified as Round 4, were tested. These included test specimens made

[1] Niagara Falls, NY 14303
[2] Honolulu, HI 96813

by NovaTech[3] and Ceramic Composites Inc, (CCI)[4]. These tubes consisted of a 0.30-inch thick inner monolithic cylinder of sintered α-SiC and a composite outer layer consisting of continuous Hi-Nicalon fibers with a SiC matrix densified by chemical vapor infiltration. A helical winding architecture was used, with no crossover of tows that allowed for infiltration by CVI in between winding of the two layers. This was intended to reduce voids and delaminations found in previous rounds. A second purpose was to examine the CVI infiltration of tubes made with fiber tows with extra sizing, 7% by weight, as compared with the normal sizing provided on fiber tows of about 1% by weight. The idea was to increase the spacing between individual fibers in a tow during winding, by virtue of the thicker sizing, and then, after burnoff, this might allow less resistance to flow of the CVI gases into the interior of the tube. Nippon Carbon, the producer of CG-Nicalon fibers, produced a separate batch of fiber tows with high sizing specifically for this experiment. As a first step in the infiltration in the CVI reactor, methane gas was injected to provide a thin (less than 0.5 microns) pyrolytic carbon coating on the fibers. This interface layer was provided to assure composite behavior during mechanical loading. The inside diameter of Round 4 tubes was 0.330 inches; the wall thickness was 0.060 inches; and the outside diameter was 0.450 inches. The double composite layer was 0.030 inches thick.

- Three duplex tubes, identified as Round 5, made by NovaTech and CCI, were also tested. These tubes had a 0.030-inch thick inner monolithic cylinder of sintered α-SiC and a composite outer layer consisting of continuous Hi-Nicalon fibers with two different sizings, and a SiC matrix densified by chemical vapor infiltration. However, these tubes differed from the Round 4 tubes in that they used the "bamboo" fiber architecture, in which fibers are interwoven around the tube at 45° angles with tow crossover during winding. This winding architecture was chosen to increase the structural integrity, and resistance to delamination, of the composite layer. There was no separate CVI infiltration between layers, as there was with Round 4 tubes. However, the carbon interface layer was provided prior to CVI as in the round 4 tubes. These Round 5 duplex tubes had an inside diameter 0.355 inches; total wall thickness 0.040 inches, and outside diameter 0.435 inches. The composite layer was 0.010 inches.
- Starfire Systems[5] and NovaTech produced ten all-composite tubes for Gamma under funding provided by Westinghouse Electric Co. The steps involved in processing these tubes were as follows:

    - Coat 14-inch long graphite mandrels with release agent.
    - Dry wind the mandrels with bamboo pattern using Tyranno-SA fiber tows
    - Apply PyC fiber coating and SiC fiber polymer coating
    - Infiltrate matrix with Starfire polymer, cure and pyrolize at 1200°C for 8 hours. Repeat PIP cycles until no further weight is gained (about 8 cycles)

  The finished Starfire tubes measured 0.371 inch ID, 0.420 inch OD, with a tube thickness of about 0.025 inches.

[3] Lynchburg, Virginia 24501
[4] Millersville, MD 21108
[5] Saratoga Technology + Energy Park, 10 Hermes Road, Suite 100, Malta, NY 12020

RESULTS FOR MONOLITHIC CYLINDERS

The data obtained during the mechanical tests consists of measurements of radial displacement of the outermost surface of the cylindrical test specimens, the axial compressive force recorded by the load cell and the piston displacement. From the radial displacement a tangential "strain" value was calculated according to:

$$\varepsilon_\theta = \frac{\Delta r}{r}$$

For data reduction and comparison with conventional material properties, the force on the piston[6] was converted to pressure, and the pressure then converted to stress, using the thin-cylinder approximation, i.e.-

$$\sigma_\theta = \frac{P}{t} r$$

where P is the internal pressure, r is the inner radius of the test specimen and t the wall thickness. For the linear portion of the initial deformation, values of Young's modulus were calculated. In general, the values obtained were consistent with those reported in the literature.

Results for two different types of monolith test specimens are shown in Figure 2. The test specimens failed in a brittle fashion, producing many small fragments during failure. For the TREX test specimen, the peak strain was 0.202% and the peak stress was 80 ksi. For the St Gobain test specimen, the peak strain was 0.224 % and the failure stress was 37.8 ksi.

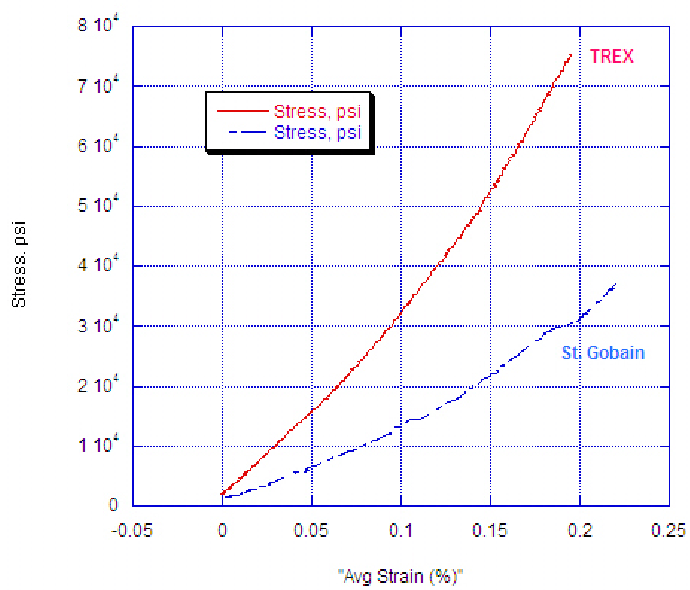

Figure 2. Stress-strain curves for two monolith cylinders

6 The small load carried by the plug itself was generally negligible in reference to the failure load, and was not accounted for in the calculations.

RESULTS FOR DUPLEX CYLINDERS

The conversion from applied load (or pressure) to tangential stress for the duplex test specimens requires an assumption about the load-carrying properties of the duplex geometry. One assumption is that the inner and outer layers (that is, the monolith and the composite) react as a tightly coupled pair of annular cylinders, and follow an elastic deformation curve at the onset of loading. This somewhat arbitrary assumption is convenient for purposes of comparing one test specimen with another. However, it is tantamount to assuming that the modulus of the monolith (i.e., CVD material) is the same as the composite, and this is not the case. Work is underway to quantify the load sharing between monolith and composite during the initial loading (that is, before failure of the monolith).

Under this assumption, the results for duplex test specimens 4BP-1 and 5P-2 are shown in Figure 3. The peak stress that was calculated for both 4BP-1 and 5P-2 was 23.6 ksi. The first-failure strain was 0.083% for 4BP-1, and about 0.17% for 5P-2. The term first-failure refers to the unique behavior of the duplex test specimens that was seen when the inner monolith failed. During one time step, at the failure point, the load being carried dropped by about 60%. This corresponds to the cracking of the monolith. From that point on, the composite structure carried the entire load. The composite layer exhibited tough behavior and a graceful mode of failure, which are desirable attributes of this material for fuel cladding applications.

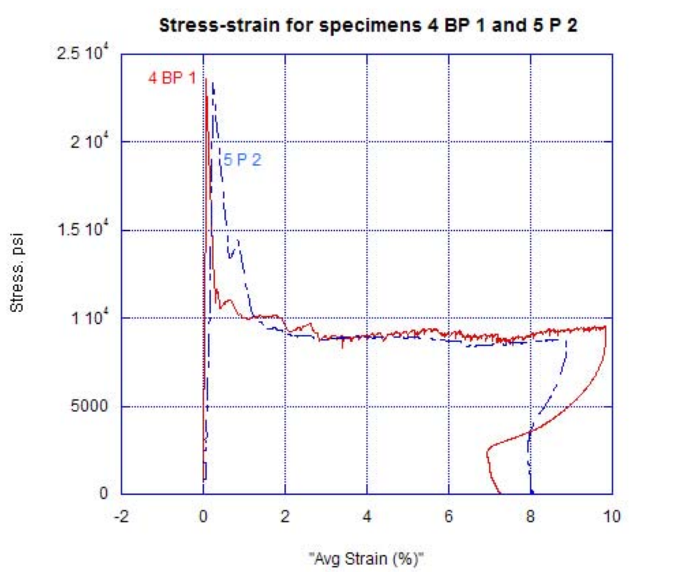

Figure 3. Stress-strain for test specimens 4 BP-1 and 5P-2

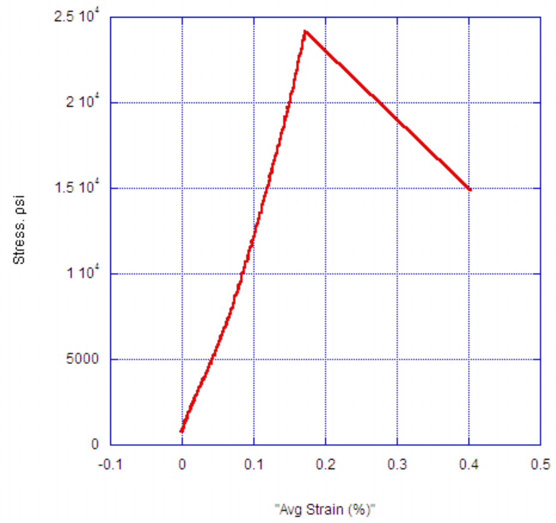

Figure 4. Stress-strain for test specimen 5P-1

The stress-strain relationship for specimen 5P-1 is illustrated in Figure 4 for the early part of the loading. The strain at first-failure was 0.172%. Although these three test specimens are considered identical, these differences in behavior illustrate the need to perform a large number of repetitions to obtain an appropriate statistical distribution of test results. This is particularly true for monolithic test specimens, which exhibit significant variability in strength

RESULTS FOR COMPOSITE-ONLY CYLINDERS

Starfire test specimen SN-010-1 failed in a brittle fashion, at a peak stress of only 6.9 ksi and at a strain of 0.5%. This failure mode was related to the inability of the fibers to debond and pullout. This behavior is illustrated in the SEM micrograph shown in Figure 6. The test specimen failed in a brittle manner and there is no evidence of fiber pullout, which would be associated with fiber debonding and fiber sliding. Fiber debonding and fiber sliding are the two main (and highly desirable) mechanisms responsible for the tough behavior exhibited by fiber-reinforced ceramic matrix composites. These mechanisms can be activated by tailoring the interface between the fibers and the matrix so that cracks that propagate through the matrix would be deflected at these interfaces. When this happens, the fibers can bridge the wake of the crack, preventing catastrophic failure. Interfaces in ceramic matrix composites are usually tailored by applying a thin, complain, coating to the fibers (e.g.- graphite, boron nitride) but it is clear from the scanning electron micrographs that such a coating was absent in this composite.

CONCLUSIONS AND INSIGHTS

The interpretation of the test results required, as stated earlier, some assumptions on load coupling and response for the duplex test specimens. This was further explored by comparing the load-strain results for a monolith test specimen and a duplex test specimen, which had an identical monolith inside. The results are shown in Figure 7. Here it is seen that the two response functions are almost identical, up to the first failure. This would seem to indicate that out to the strain at first failure (that is, about 0.2%) the responses are essentially the same, and thus the composite was not carrying much load at this point. However, after the failure of the monolith, the composite structure carries the entire load.

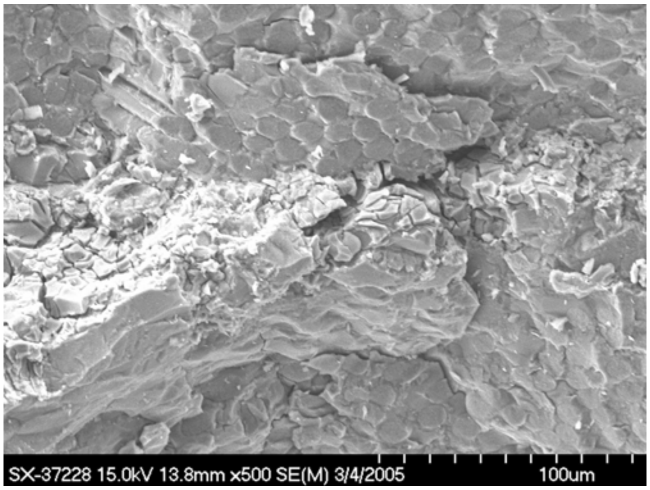

Figure 6. SEM of fracture surface of Starfire test specimen.

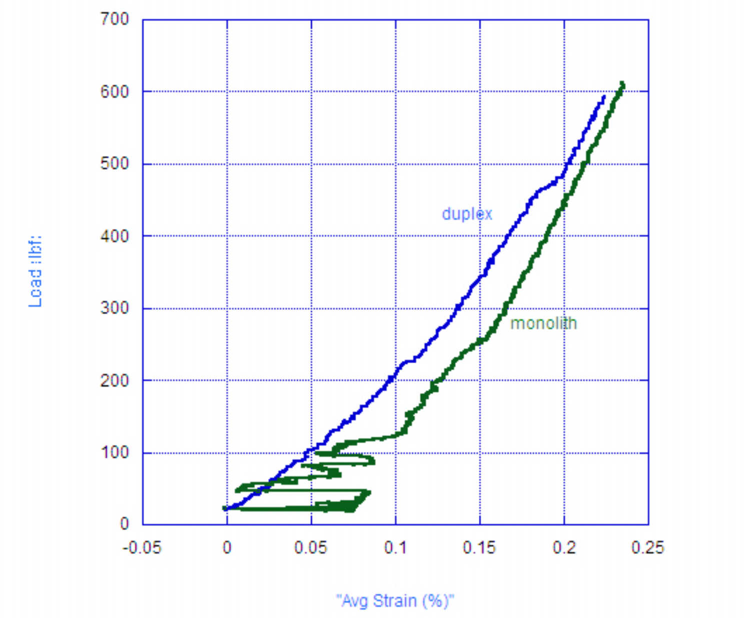

Figure 7. Comparison of load versus strain response of monolith and duplex geometry test specimens at low strains.

Some insights that have been gained from the strength testing thus far are:

- Monoliths tested alone will fail at low strains in a brittle fashion.
- Some types of duplex test specimens exhibit tough behavior and can experience large amounts of deformation and damage tolerance (graceful failure).
- The composite-only test specimens that were evaluated did exhibit poor performance because of lack of fiber debonding and sliding as a result of lack of an interfacial coating
- Although the number of tests have been limited, the data are reasonably consistent within a given test specimen geometry

Much more work needs to be done to make a more compelling case for using SiC structures as nuclear fuel cladding. Tests scheduled for calendar year 2006 include construction and initial utilization of an environmental test of SiC cladding materials in a combined radiation and reactor water chemistry environment. This is scheduled to be done at the reactor at MIT. Tests on the strength of SiC test specimens at elevated temperature are scheduled at ORNL.

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