MECHANICAL STRENGTH OF CTP TRIPLEX SIC FUEL CLAD TUBES AFTER IRRADIATION IN MIT RESEARCH REACTOR UNDER PWR COOLANT CONDITIONS

Herbert Feinroth
Ceramic Tubular Products LLC
Rockville, Maryland USA

Matthew Ales and Eric Barringer
Ceramic Tubular Products LLC

Gordon Kohse and David Carpenter
Massachusetts Institute of Technology
Cambridge, Massachusetts USA

Roger Jaramillo
Oak Ridge National Laboratory
Oak Ridge, Tennessee USA

ABSTRACT

An experiment was conducted in the MIT Research Reactor (MITR) to irradiate triplex silicon carbide fuel cladding tubes under typical Pressurized Water Reactor conditions. Measurements were made to determine the impact of exposure on strength and swelling. The SiC clad tubes were fabricated by Ceramic Tubular Products (CTP) with dimensions typical of 15 x 15 commercial PWR reactor fuel. The triplex tubes contain 3 layers, an inner monolithic SiC layer to maintain hermeticity, a central SiC/SiC composite layer to provide a graceful failure mode in the event of an accident, and an outer SiC environmental barrier layer. Clad tubes were exposed to 300 ^{o}C pressurized water containing boric acid, lithium hydroxide, and hydrogen overpressure, typical of PWRs. Thirty nine (39) specimens of various types were exposed to coolant, some within the neutron flux region and some outside the neutron flux region. Twenty seven (27) were removed for examination and test after 4 months exposure. Following examination, twenty specimens were reinserted for additional exposure, along with 19 new specimens. The 4 month specimens were weighed and measured at MIT, and some were shipped to Oak Ridge National Laboratory (ORNL) where they were mechanically tested for hoop strength using a polyurethane plug test apparatus. Results were compared with the pre-irradiation strength and dimensions. Some specimens retained their original strength after exposure, others with a less homogeneous monolith, lost strength.

INTRODUCTION

Gamma Engineering, one of the parent companies of Ceramic Tubular Products, has been developing multilayered silicon carbide clad tubes for use in commercial light water reactors since 1999. Reference (1) summarizes the initial development work performed under a DOE small business grant. This early work confirmed that a two layer silicon carbide clad tube, combining a dense inner monolith layer with a composite outer layer, is impermeable to fission gases during normal reactor operation, and exhibits a graceful failure mode during severe accidents. This latter behavior allows the fuel clad to retain the solid uranium oxide fuel and also to maintain coolability during severe accidents.

After 2001, Gamma continued to refine and test the multilayered concept. Several different fiber architectures, fiber types and matrix infiltration methods were examined in an effort to assure radiation resistance and to provide increased strength of the multilayered tube for both normal operation and accident conditions. Reference (2) summarizes early mechanical hoop strength tests of the un-irradiated duplex tubes using the plug testing approach developed by ORNL for the Mixed Oxide Fuel test program. Details of this hoop test method are described in reference (2).

In May, 2006, with support from both the DOE and Westinghouse, an irradiation test was initiated in the MIT research reactor (MITR) to examine the effects of exposure to neutron irradiation and hot PWR coolant on the behavior of multilayered silicon carbide fuel clad tubes. Tube diameter and thickness of the test specimens were almost typical of the 15 x 15 commercial PWR fuel assemblies now in use. Test specimens were about 2 inches long. After initial exposure of 39 specimens for a period of 4 months, about half the specimens were removed for examination and testing, and replaced with new specimens for continued irradiation. The removed specimens were allowed to cool, examined for dimensional changes, and then tested for hoop strength to determine the effect of irradiation on strength. This paper describes the multilayered clad specimens, the conditions of the exposure in the MITR, and the results of the strength testing and dimensional examination. It also describes more recent measurement of dimensional changes after exposure for an additional 8 months. Additional mechanical tests (hoop strength) after the additional 8 months are planned in the future.

SAMPLE PREPARATION

Because funding was limited, post irradiation hoop strength testing was performed on only five tube specimens. Each specimen was originally 1.9 inch in length during irradiation, and about 1.4 inch in length during post irradiation mechanical testing. The specimens were selected for this irradiation exposure and post irradiation examination on the basis of having a variety of different monolith characteristics, fiber suppliers and outer environmental barrier coating, in order to learn which of these characteristics offered the best performance during exposure.

During irradiation, the specimens were 1.9 inches long, and were exposed to coolant both inside and out, and at the cut ends, although coolant flow velocities were much lower on the inner and exposed end surfaces. After exposure, a ¼ inch ring was removed from each end for corrosion studies. Hence, the post irradiation mechanical testing was performed on 1.4 inch long specimens. Except for specimen H2-5, all specimens were exposed to neutron irradiation. Specimen H2-5 was exposed to coolant but not to in-core neutron flux.

Characteristics of the five exposed and tested specimens are presented in Table I:

Table I – Test Specimen Characteristics

Specimen Identity	Monolith	Fiber	Barrier coating	Core Location	% of peak fluence
H2-1	Coorstek	Sylram iBN	CVI cap	Tier 3	95
H2-5	Coorstek	Sylram iBN	CVI cap	Out of core	0
F1-1	Coorstek	HiNic-S	CVI cap	Tier 3	95
D1-2	TREX	Sylram iBN	Machine + TREX cvd	Tier 4	100
B1-2	TREX	HiNic-S	CVI cap	Tier 2	90

Common characteristics – All specimens incorporated a beta phase chemical vapor deposition (CVD) monolith of about 15 mils thickness and 0.350 – 0.354 inch ID, a fiber wound architecture developed by Ceramic Tubular Products using a unique winding device, a carbon interface coating on the fibers, and a composite layer using chemical vapor infiltration (CVI). As noted in Table 1, four of the specimens incorporated an outer environmental barrier coating (EBC) which was merely an extension of the CVI composite layer after the infiltration “capped off”, whereas the fifth specimen, D1-2, had a unique EBC in which the “capped off” CVI layer was machined smooth, and an additional SiC layer was deposited by TREX using CVD. Both types of fibers are stoichiometric beta phase fibers. The Sylramic iBN fiber also has a thin interface layer of in-situ diffused boron carbide.

IRRADIATION CONDITIONS IN THE MIT TEST REACTOR

Although the 5 MW MIT research reactor operates at near atmospheric pressure and temperature, a special titanium autoclave inserted in the reactor, and connected to a closed loop outside the reactor, provides inlet water at 300 ^oC and 1500 psi at a mass flow of 0.25 kg/sec. Oxygen is removed from the circulating water by bubbling hydrogen through the coolant makeup tank and a catalytic recombiner. The heated portion of the loop is insulated by a CO_2 gas gap, assuring a temperature drop of not more than 20 ^oC over the length of the in-core section. Thus, this closed loop simulates the actual coolant conditions in a typical PWR commercial reactor.

The design of the titanium autoclave also allows for a portion of the test specimens to operate in the PWR coolant but outside of the high neutron flux core area. This allows separating the effects of coolant corrosion only, from the combined effects of coolant exposure and neutron irradiation.

Figure 1 presents the neutron flux profile for the MIT reactor. The MIT staff has estimated an average neutron fluence (> 1 MeV) during the 4 month test of 0.5 x 10^{21} n/cm^2 based on neutron activation of the titanium loop materials. Based on the flux profile in Figure 1, we calculate that the central tiers of this experiment (tiers 2, 3, 4 and 5) received about 0.63 x 10^{21} neutrons/cm^2.

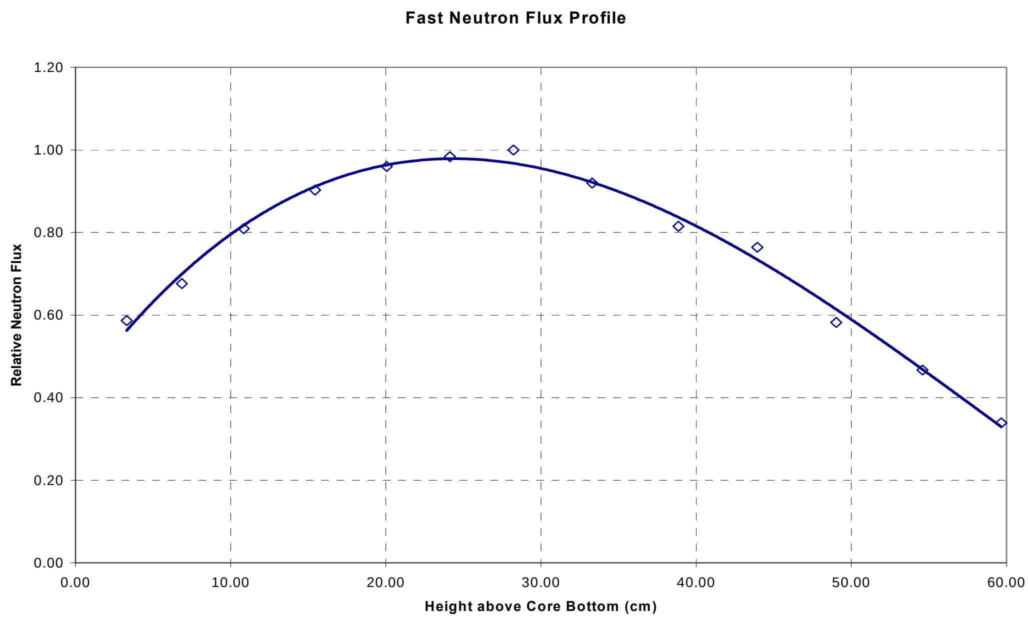


Figure 1 - Neutron Flux Profile in MITR-II Reactor

With respect to fast neutron damage, one displacement per atom (dpa) for silicon carbide is achieved with a fast neutron fluence (> 1 MeV) of about 1 x 10^{21}. Thus during this experiment, the four irradiated specimens that were mechanically tested received about 0.63 dpa exposure. For comparison purposes, the clad in a fully burned LWR fuel rod (50 gwd/t) receives about 10 dpa fast neutron exposure.

MECHANICAL TESTING AT ORNL

ORNL successfully completed the room-temperature hoop strength tests for the five 1.40” specimens supplied by MIT. Due to the high levels of contamination for these specimens, testing was performed in a hot cell located within Building 3525.

Figure 2 shows the load vs. ram displacement data for the five specimens. The specimen load is the ram force minus the force required to compress the plug without a specimen. The x axis has been normalized such that the origin corresponds with the onset of specimen loading.

This figure indicates that H2-1 and H2-5 had the largest load bearing capability of 850 pounds, and F1-1 had a bit lower load capability of about 700 pounds. Specimens D1-2, and B1-2 both had about half the load capability of the H series, 400 to 450 pounds. As noted below, we ascribe this decrease in capability to structural defects (laminations and porosity) in the monolith layer.

The general failure behavior for all of the specimens was similar to that previously observed for as-fabricated specimens. That is, the specimens were loaded until the monolith layer failed, at which point all load was transferred to the composite/EBC layers. For each specimen, load was maintained as ram displacement was continued.

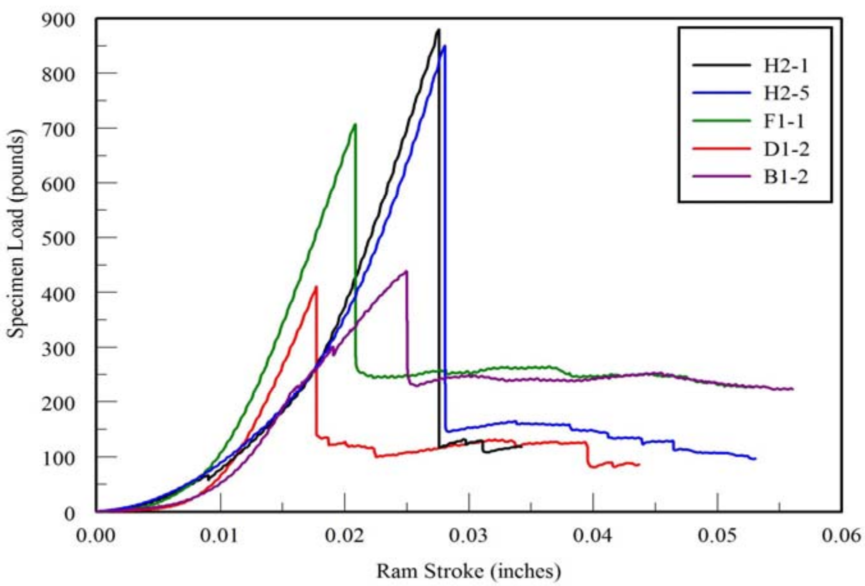


Figure 2. Load vs. Ram displacement for specimens exposed to PWR coolant in the MIT reactor.

The data in Figure 2 show that four of the five specimens had similar strain behavior during the initial loading period. That is, the effective elastic modulus (related to slope) was similar. However, B1-2 was observed to have a somewhat lower effective modulus. Moreover, there appeared to be some initial “yielding” of the specimen prior to failure of the monolith layer (see curvature of plot in Figure 2). This behavior is inconsistent with that observed for the other SiC specimens, including the as-fabricated specimens. The reasons for this difference in strain behavior are not known at this time.

COMPARISON WITH AS-FABRICATED SPECIMEN BEHAVIOR

The room-temperature hoop strength data for the five exposed specimens (peak load at failure) are compared to test results obtained for as-fabricated specimens in Figure 3. The hoop strength for F1-1, H2-1 and H2-5 where within the range measured for the as-fabricated specimens. The as fabricated test results comprised a larger data base and the average strength and standard deviation are as follows:

- F-Series Specimens – 4 data points; average strength = 808 lbs; SD = 85 lbs
- H-Series Specimens – 3 data points; average strength = 881 lbs; SD = 8 lbs

There appears to be a substantial reduction in strength during irradiation for the two specimens (B1-2 and D1-2) fabricated with the TREX monolith. It should be noted that the number of specimens fabricated with the TREX monolith that have been tested is somewhat limited, and caution must therefore be exercised when interpreting these results.

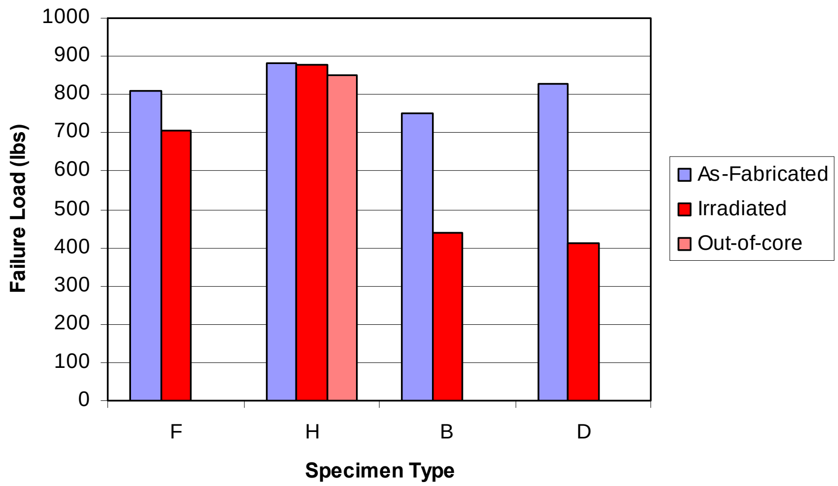


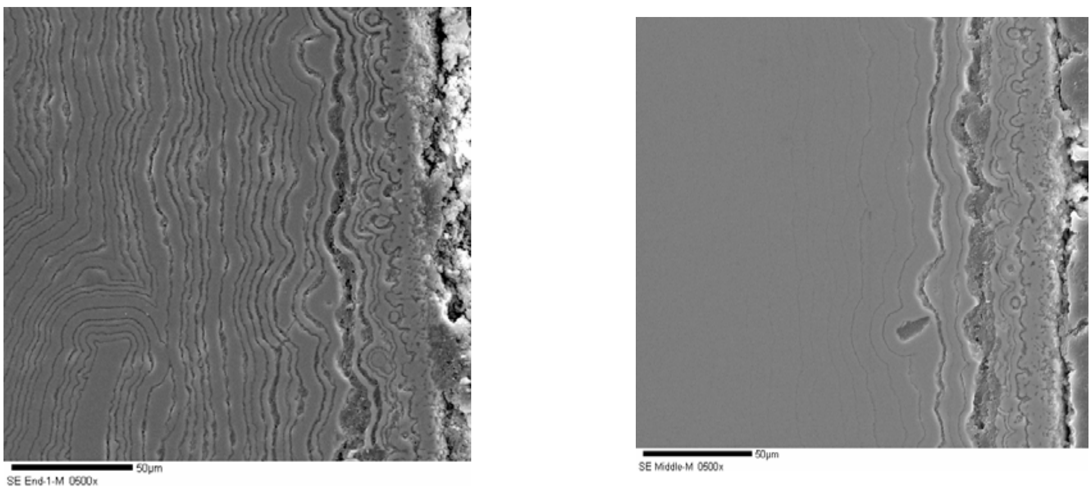
Figure 3. Comparison of room-temperature hoop strength for as-fabricated specimens and for specimens exposed to PWR coolant in the MIT Research Reactor.

RESULTS OF SEM EXAMINATIONS

Test specimens were examined visually and by SEM after 4, 8 and 12 months exposure. Figure 4 depicts an F series specimen after 12 months exposure and prior to removal of the ¼ inch end ring for final weight change measurements. The small triangular piece of composite missing at the top end is thought to have resulted from a combination of the mechanical fixture used to hold the specimens in place, and possible corrosion of the central layer starting from the exposed cut ends. The figure also illustrates the winding pattern, or fiber architecture used in CTPs triplex cladding design. Although not visible on the photograph, there is a 5 mil overcoat of SiC that follows the pattern of the winding, and that provides protection from coolant corrosion effects.

Figure 4 - Specimen F1-4 after 12 months exposure in MIT reactor

To provide some insight into the strength test results, SEM examination was performed on small ring sections of several of these specimens that were cut from the ends by MIT prior to conducting the hoop strength tests at ORNL. Figure 5 shows a high magnification (1,000 x) image of the inner surface of the TREX monolith on a B series specimen. The section is an axial cut of the ¼ inch ring cut from the end of the specimen, with the right side of the image being the inner surface of the monolith tube exposed to coolant. Examination of the polished cross section showed that the TREX monolith consists of numerous thin layers. The layered structure was more prominent on the exposed end of the sample (see Figure 5 a). Although the layers were less prominent ¼" from the exposed end (i.e., the cut end of the sample), the layers were clearly evident on the inner surface of the monolith (Figure 5 b). In fact, crack-like layering on the ID extended the whole ¼" length of the sample. This suggests that coolant could have penetrated the inner surface of the TREX monolith.



(a)Near exposed end of B1-2 (b) Near cut end of B1-2

Figure 5 SEM's of Monolith Portion of Sample B1-2 (TREX monolith) after 4 month exposure

It is speculated that the reason for the lower strength of the B and D specimens after irradiation is at least partly due to the porosity and laminations in the monolith and the possible effects of irradiation on the behavior of the duplex structure. Further investigation revealed that the supplier used a "research" type reactor in preparing the CVD material, with time variant deposition of higher carbon non-stoichiometric SiC. The layers of non-stoichiometric higher carbon SiC had accelerated corrosion leading to the observed reduction in strength. For later development work, the supplier utilized an improved deposition reactor, and conventional CVD process parameters, leading to more homogeneous, stoichiometric, dense material.

DIMENSIONAL CHANGES AND RADIATION INDUCED SWELLING

After both the 4 month exposure, and the additional 8 month exposure, post-irradiation dimensions were measured for some triplex tube specimens. The data are provided in Table II. The OD measurements have high variability because of the fiber architecture, and therefore are not a reliable measure of radial swelling. Axial growth measurements are more accurate.

Table II. Dimensional changes for irradiated triplex tube specimens

Sample	Exposure Time (mo)	Pre-irradiation		Post-Irradiation		Irrad Δlength		Irrad ΔOD
		Length (in)	Avg OD (in)	Length (in)	Avg OD (in)	(inches)	(percent)	(inches)
F4-1	8	1.904	0.414	1.915	0.416	0.011	0.58%	0.002
F2-3	8	1.904	0.411	1.913	0.416	0.009	0.47%	0.005
H1-4	8	1.896	0.421	1.907	0.424	0.011	0.58%	0.004
H1-2	8	1.906	0.421	1.918	0.424	0.012	0.63%	0.003
B1-4	12	1.909	0.419	1.920	0.421	0.011	0.58%	0.002
H3-4	12	1.893	0.419	1.907	0.421	0.014	0.74%	0.002
F1-4	12	1.871	0.410	1.883	0.412	0.012	0.64%	0.002

The average length growth for the specimens irradiated for 8 months (average exposure of about 1.3 dpa) and 12 months (average exposure of about 1.9 dpa) are summarized in Table III.

Table III – Average length growth of SiC triplex cladding after MITR irradiation

Exposure	Average	Std. Deviation
8 Month	0.57%	0.066%
12 Month	0.65%	0.082%

These results compare well with the data reported by Snead, et al., showing saturated volumetric swelling of about 2% (equivalent to 0.67% axial swelling) at about 300 ^{o}C. The Snead compilation (redrawn from reference 3) is shown in figure 6.

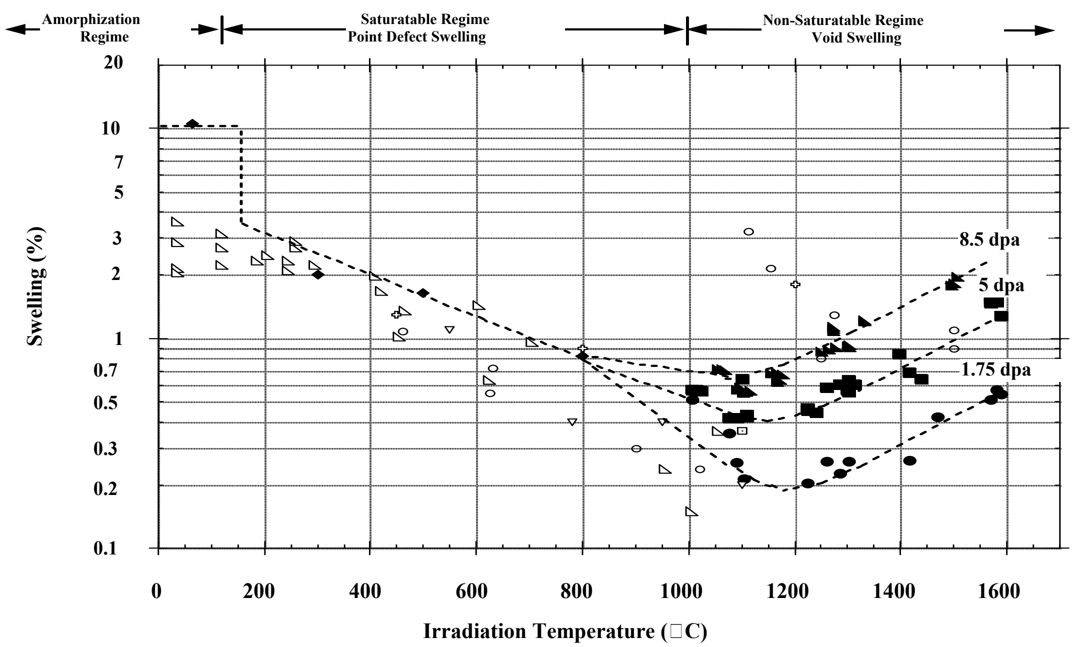


Figure 6 - Volumetric Swelling for SiC (redrawn from reference 3)

ANALYSIS AND EVALUATION OF MECHANICAL TEST RESULTS

CVD beta phase silicon carbide is reported by the suppliers (Coorstek and TREX) to have a flexural strength of 450 to 468 MPa (66,000 to 69,000 psi.) The use of flexural strength as the failure strength for hoop loading was validated by testing samples of monolith. Using the thin walled cylinder formula (stress = pressure x radius/thickness), we calculated what the stress in the monolith layer would be (in the five specimens tested) if all of the hoop load was taken by the monolith layer, with none of the hoop load being shared with the composite layer or EBC prior to failure of the monolith. The results are shown in Table IV.

Table IV Internal Pressure and Stress in Monolith at Failure Assuming No Load Sharing

	Type	Failure load lb	Internal pressure psi	Stress at failure psi	Possible Composite/EBC load share
F1-1	Coorstek HiNicalon	708	7516	86,760	21.6%
H2-1	Coorstek Sylramic	880	9359	107,940	37%
H2-5	Coorstek Sylramic	851	9009	104,144	34.7%
B1-2	TREX HiNicalon	439	4542	53,111	NA
D1-2	TREX Syl w/ovct	411	4235	50,675	NA

The results show that the three specimens with Coorstek monolith exhibited an apparent strength which is 21.6 to 37% higher than the reported flexural strength of the monolith alone. Since we know that the actual stress in the monolith will not exceed the basic material capability of about 68,000 psi, the results of these tests show that the internal pressure load was actually shared amongst the three layers prior to monolith failure. What is revealed is that the combination of the tightly wound composite layer, and the outer environmental barrier layer, have reinforced the monolith layer, and these two added layers together share from 21.6 to 37% of the total hoop load applied in the test.

This result is quite important to the application of this cladding to PWR cladding. In effect, it demonstrates the capability of the cladding to take 3 to 4 times more fission gas pressure during reactor operation, as compared to zircaloy cladding which because of its creep behavior, is limited to a maximum pressure of about 2000 psi. It is this ability to take high internal pressure that exemplifies the value of this ceramic cladding to extending the burnup and lifetime of light water reactor fuel.

With regard to the behavior of the two specimens having TREX monoliths, we cannot determine whether there was load sharing in these triplex tubes because, as stated previously, the presence of high porosity and laminations in these tubes seems to have decreased strength overall, and it is not possible to separate the effects of reduced material properties from the possible effects of load sharing.

SUMMARY AND CONCLUSIONS

Irradiation testing of triplex cladding specimens is continuing in the MIT reactor, and is planned to begin with contained fuel in the HFIR reactor in 2009. The results of initial post irradiation tests described in this report indicate that:

- Triplex clad specimens made with good quality CVD monoliths have the same hoop strength as un-irradiated samples within 2 standard deviations, after exposure to typical PWR operating conditions including fast neutron irradiation averaging about 0.63 dpa.

- Average length growth due to irradiation induced swelling of triplex cladding specimens exposed to about 1.9 dpa fast neutron exposure was 0.65% and is consistent with the values in previous experiments.

- The addition of an outer composite layer with a unique fiber architecture, and an outer environmental barrier layer, to a monolith tube, can provide reinforcement to the monolith layer on the order of 21.6 to 37%, thus providing additional pressure retention capability to triplex tubes during reactor operation. This increases the capability for SiC triplex cladding (as compared to zircaloy cladding) to safely allow higher fission gas release, as required for high burnup, high power rated, nuclear fuel.

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