

## Radial Heat Flux – Irradiation Synergism in SiC ATF Cladding

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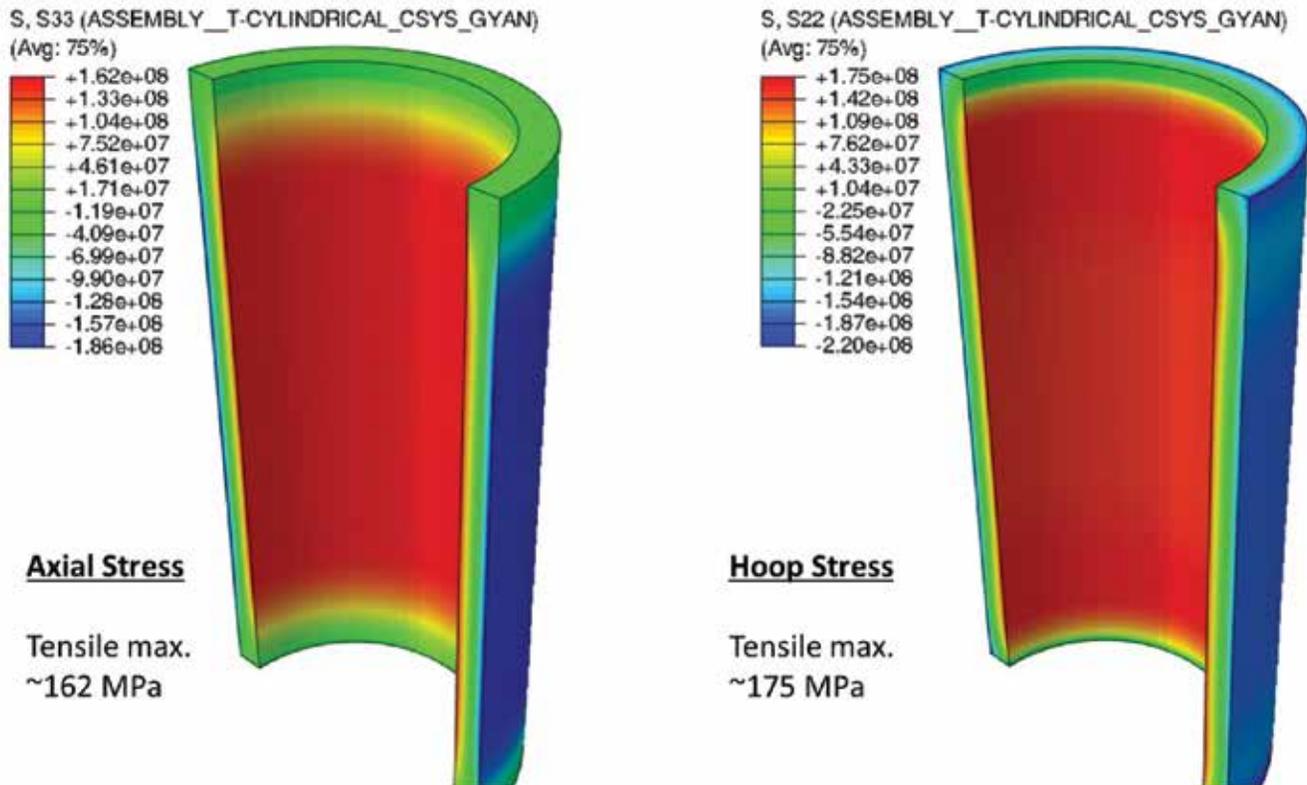


Figure 1. The “inverse thermal stress” predicted by multiphysics finite element thermomechanical analysis to develop in SiC/SiC tube as the synergistic effect of radial heat flux and neutron irradiation. Assumed conditions include radial heat flux 0.6 MW/m<sup>2</sup> and outer surface temperature 573K.

Silicon carbide fiber-reinforced silicon carbide matrix (SiC/SiC) composite is an enabling materials technology for the ultimate accident-tolerant fuels for light-water reactors (LWRs) and the core structures for advanced high-temperature reactors of various concepts. This ceramic composite combines the intrinsic benefits of SiC (e.g., outstanding irradiation

tolerance and steam-oxidation resistance) and the engineered benefits of fiber composites (e.g., damage tolerance and design flexibility).

The unique set of benefits for SiC/SiC composite comes with a unique set of challenges. This project addresses one of the most critical challenges, this material’s inability to maintain fission-product-gas containment.

*This project is intended to experimentally verify the irradiation-induced inverse stress behavior and determine its impact of the stress on the performance of SiC/SiC as the LWR fuel cladding.*

### Project Description

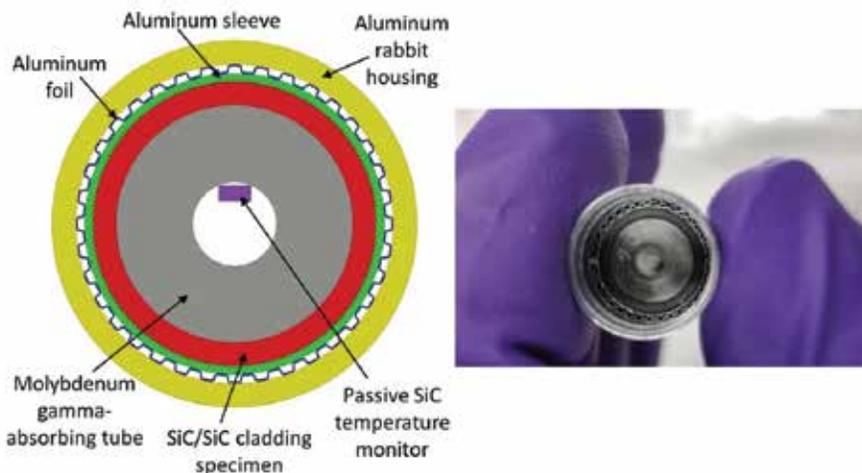
Among the unique challenges for fuel cladding made from SiC/SiC composite is internal stress arising from differential irradiation strain. SiC swells rather rapidly during the first week or two of operation as the LWR fuel cladding until the point-defect accumulation saturates. The swelling of SiC accompanies a decrease in the thermal conductivity, resulting in a steep temperature drop across the cladding-wall thickness due to high heat flux from fuel to coolant. The saturated swelling strain exhibits negative dependence on temperature. The results are the tensile stress at the inner surface and compressive stress at the outer surface of the cladding wall i.e., the opposite effect from normal thermal stress (Figure 1).

The implications of the predicted “inverse thermal stress” range from significant (such as microcracking initiating from the inner surface) to severe (crack networking leading to a loss of hermeticity and a threat to structural integrity) for the fuel cladding. This R&D project is

intended to experimentally verify the irradiation-induced inverse stress behavior and determine its impact of the stress on the performance of SiC/SiC as the LWR fuel cladding [1].

The project consists of three technical tasks: 1) design and build an irradiation vehicle that enables neutron irradiation of small tubular specimens under an LWR-relevant radial heat-flux condition, 2) experimentally verify the thermal gradient through the tube wall thickness, and 3) examine the damage in the irradiated tubes and other effects of neutron irradiation under a steep temperature gradient. Each task is first-of-a-kind development involving significant technical obstacles that have to be overcome. However, success of the project will have implications beyond its immediate objectives because this innovative approach will enable studies on irradiation effects accompanying significant dimensional instability and irradiation behavior of any material under a steep temperature gradient.

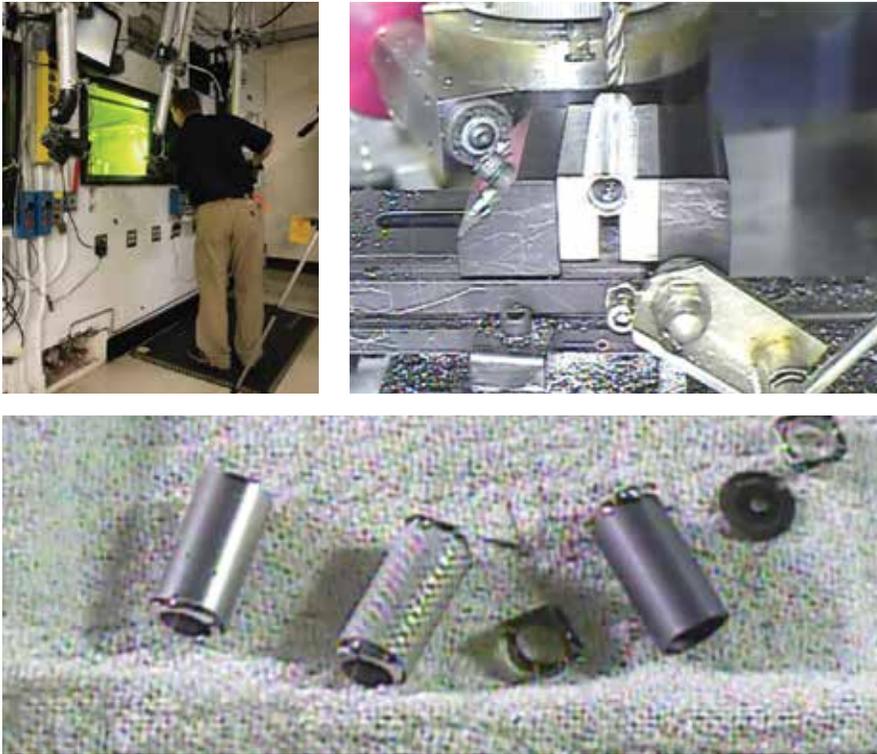
Figure 2. Design and construction of the Fire Rabbit irradiation vehicle developed in the project, enabling simultaneous high radial heat flux and neutron exposure for tubular test specimens.



### Accomplishments

The irradiation engineering challenge in this project is to develop a low-cost “Fire Rabbit” capsule that accommodates SiC/SiC tube specimens, enabling a radial heat flux relevant to LWR fuel claddings, and maintaining the specimen temperature constant [2]. The heat flux was produced by placing a concentric gamma-absorbing tube as a heat source inside the specimen (Figure 2). The need to maintain the specimen outer-surface temperature is the most significant challenge when SiC swells linearly by ~0.7% while a gap of a few microns causes unacceptable temperature deviations due to high heat flux. Moreover, any excessive mechanical constraint on the specimen has to be avoided.

Our team came up with the idea of using an embossed metallic foil sleeve surrounding the specimen, so that a constant heat conduction from the specimen to the capsule housing is maintained during and after the specimen swelling. Thus, an expandable thermal homogenizer sleeve was inserted between the foil and the specimen to minimize radial perturbation of temperature. The embossing pattern was optimized through a series of ex-pile experiments involving the measurement of heat conduction as a function of the compressive deformation of the foil layer. The actual radial build of the irradiation capsule is shown in Figure 2.

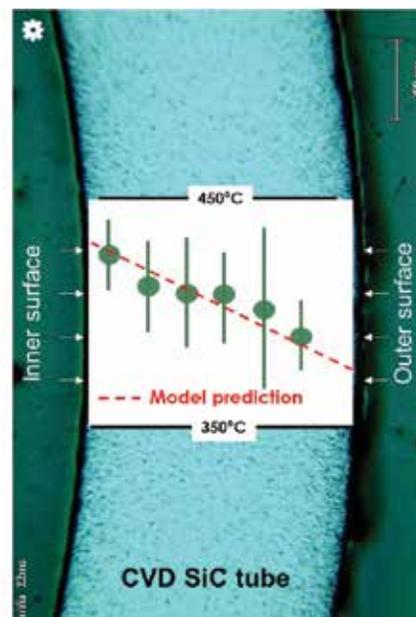
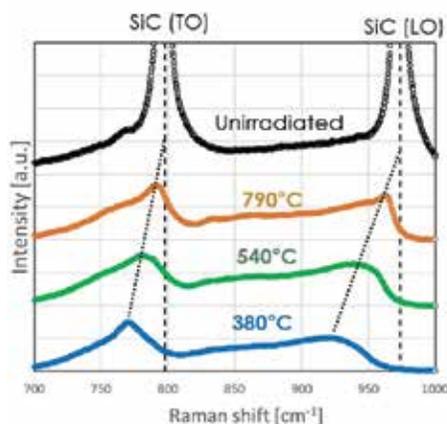


*Figure 3. Disassembly of Fire Rabbit at the Irradiated Materials Examination and Testing Laboratory following neutron irradiation in the High Flux Isotope Reactor. Top left: a hot-cell operator opening the capsule; top right: capsule housing being carefully milled to retrieve the delicate samples; bottom: retrieved specimens with aluminum thermal homogenizer jacket (left), SiC/SiC composite tube with centering thimbles (middle), and monolithic SiC tube (right).*

The capsules were irradiated in the Flux Trap facility of the High Flux Isotope Reactor for one full cycle following approval of the entirely new capsule design for the reactor. During this irradiation, the SiC sample accumulated the damage level of  $\sim 2$  displacement per atom, achieving the swelling-saturation dose. The irradiated capsules were then carefully opened using a precision milling tool in the Irradiated Materials Examination and Testing Laboratory. The capsule-opening operation, including the capsule being milled while gently clamped in fixture, and the specimens being successfully retrieved are shown in Figure 3

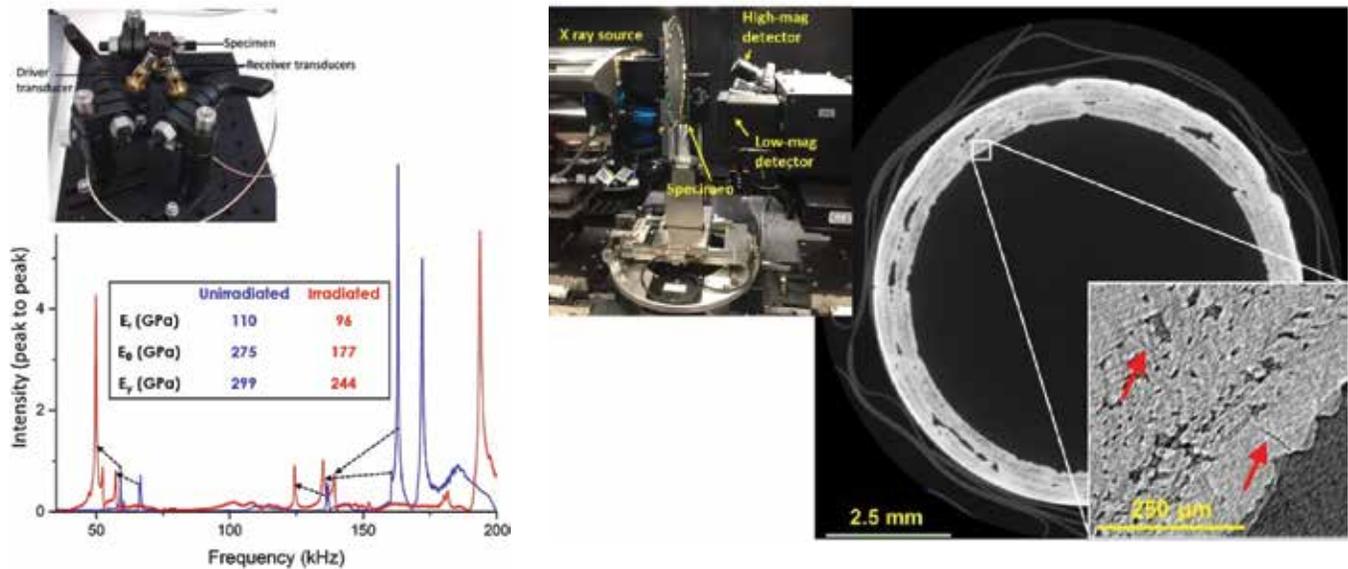
Verifying the actual temperature of irradiation is essential for this experiment because the temperature profile defines stress distribution within each specimen. Sources of potential deviations from design temperature include poor thermal contact at any interface between the specimen and the housing, poor specimen quality, and unexpected axial heat loss from the heating tube. Passive temperature monitors were used to verify the temperature of the heating tube, and the average irradiation temperatures of individual specimens were confirmed by swelling measurements.

Figure 4. Development of a laser micro-Raman technology to map the irradiation temperature of SiC by post-irradiation thermometry (left), and application of the technology in verifying the temperature gradient across tube-wall thickness during neutron irradiation of SiC samples in Fire Rabbit (right).



Moreover, a novel experimental technique for mapping irradiation temperature in specimens was developed. In this technique, the one-to-one correlation of certain Raman peaks and the lattice swelling of irradiated SiC was established [3,4] then successfully applied to determine the radial temperature profile within a thin-walled tube. Despite its limited accuracy due, partly, to the small laser spot size used for the mapping, the developed approach was able to prove the presence of a temperature slope consistent with the predicted for a CVD SiC specimen as shown in Figure 4.

The project plans an extensive set of post-irradiation characterization, starting with non-destructive evaluations (NDE) and followed by destructive evaluations such as cross-sectional microscopy and stress state-characterization involving elevated-temperature annealing. NDEs are focused on detecting and characterizing microcracking behavior and the possible resultant changes in gas permeability. Because microcracking due to irradiation-heat-flux synergy is anticipated at or near the inner surface of the tubes, where stress is primarily in tension, sonic or X-ray-probe NDEs are particularly useful.



The technique of ultrasonic-resonance spectroscopy (RUS) was utilized to evaluate the collective effects of microcracks on the elastic properties of the tubes [5]. Without changes in crack microstructures, ~6% reduction in Young's modulus is anticipated in SiC due to neutron irradiation alone. However, as shown in Figure 5 (left), more significant and anisotropic decreases in elastic constants are observed after irradiation in the Fire Rabbit, implying more-

extensive microcracking across the circumferential and axial orientations than radial. High-resolution X-ray computed tomography (XCT) examination revealed the presence of radial microcracks, which were not found in unirradiated tubes. The microcrack shown in Figure 5 (right) appears to be slightly open at the inner surface and extends to the mid-plane of the tube wall, where the inverse thermal stress is anticipated to be neutral.

*Figure 5. Observation and characterization of microcracks present in the SiC/SiC composite tubes following neutron irradiation in Fire Rabbit: Resonant ultrasonic spectrometry showing significant decreases in circumferential and axial elastic moduli after irradiation (left) and X-ray computed tomography identifying radial microcracks near the inner surface of an irradiated tube (right).*

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*The results obtained by NDE characterizations are consistent with those predicted by computational modeling incorporating the known effects of irradiation in SiC/SiC composites.*

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### **Future Activities**

The results obtained by NDE characterizations are consistent with those predicted by computational modeling incorporating the known effects of irradiation in SiC/SiC composites. Within the present project, additional experimental data, including thermal conductivity, stress state, and helium permeation will be generated to further validate the multiphysics model and computation. The study will contribute toward fully establishing predictive capability regarding failure probability and behavior of SiC-based fuel cladding and core structures during the normal and off-normal operation of nuclear reactors.

### **Publications**

- [1.] Y. Katoh, K.A. Terrani, T. Koyanagi, C.M. Petrie, G. Singh, L.L. Snead, C. Deck, Irradiation – High Heat Flux Synergism in Silicon Carbide-Based Fuel Claddings for Light Water Reactors, in: LWR Fuel Perform. Meet. TopFuel 2016, Boise, Idaho, USA, 2016: pp. 823–831.
- [2.] C.M. Petrie, T. Koyanagi, J.L. McDuffee, C.P. Deck, Y. Katoh, K.A. Terrani, Experimental Design and Analysis for Irradiation of SiC/SiC Composite Tubes under a Prototypic High Heat Flux, J. Nucl. Mater. 491 (2017) 94–104.
- [3.] T. Koyanagi, M.J. Lance, Y. Katoh, Quantification of irradiation defects in beta-silicon carbide using Raman spectroscopy, Scr. Mater. 125 (2016) 58–62. doi:10.1016/j.scriptamat.2016.08.004.
- [4.] T. Koyanagi, Y. Katoh, M.J. Lance, Raman spectroscopy of neutron irradiated silicon carbide: Correlation among Raman spectra, swelling, and irradiation temperature, J. Raman Spectrosc. (2018) 1686–1692. doi:10.1002/jrs.5425.
- [5.] G. Singh, T. Koyanagi, C. Petrie, K. Terrani, Y. Katoh, Evaluating the irradiation effects on the elastic properties of miniature monolithic SiC tubular specimens, J. Nucl. Mater. 499 (2018) 107–110. doi:10.1016/j.jnucmat.2017.10.060.

Distributed Partnership at a Glance	
NSUF and Partners	Facilities and Capabilities
Oak Ridge National Laboratory	High Flux Isotope Reactor (HFIR), Irradiated Materials Examination and Testing Facility (IMET) Hot Cells, Low Activation Materials Design and Analysis Laboratory (LAMDA)
Collaborators	
General Atomics	Christian Deck (collaborator), Christina Back (collaborator)
Oak Ridge National Laboratory	Chris Petrie (collaborator), Gyanender Singh (collaborator), Hsin Wang (collaborator), Joel McDuffee (collaborator), Kurt Terrani (collaborator), Takaaki Koyanagi (collaborator), Xunxiang Hu (collaborator), Yutai Katoh (principal investigator)