3.4 HIGH DOSE IRRADIATION STUDY OF NEWER GENERATION NUCLEAR GRADE SiC COMPOSITES: IRRADIATION EXPERIMENTS - T. Koyanagi, N.O. Cetiner, Y. Katoh (Oak Ridge National Laboratory), T. Nozawa and K. Ozawa (Japan Atomic Energy Agency)

OBJECTIVE

This report describes the technical planning and progress of high dose irradiation experiment on newer generation nuclear grade SiC composites for fusion structural applications.

SUMMARY

A study of high dose neutron irradiation effects on newer generation nuclear grade SiC fiber reinforced SiC matrix composites has started, employing a series of new rabbit capsules. The activity is part of the JAEA-USDOE collaboration, with an irradiation matrix designed to achieve a maximum neutron fluence beyond 100 dpa-SiC for a range of irradiation temperature from ~300 to ~1000°C. Twelve capsules containing three types of SiC composites and high-purity monolithic SiC were prepared. Irradiations for all the rabbit capsules will start in Cycle 457 or 458 at the High Flux Isotope Reactor. Properties to be evaluated include dimensional stability, mechanical, thermal, and electrical properties, and microstructures.

PROGRESS AND STATUS

Introduction

Excellent irradiation stability up to ~40 dpa has been proven for early nuclear grade high-crystalline and near-stoichiometric Hi-Nicalon Type S SiC fiber reinforced SiC composite (SiC/SiC) with SiC-pyrolytic carbon (PyC) multilayer interphase in previous ORNL-JAEA collaborations on fusion materials; no obvious change was observed after the saturation of changes in properties such as swelling, thermal conductivity, and elastic modulus at low neutron fluence [1]. The recent work in this collaboration has accomplished neutron irradiations to greater than 70 dpa in HFIR, and provided an extended understanding of effects of high does neutron irradiation on thermomechanical properties of this SiC/SiC. One of the important findings was irradiation-temperature dependent degradation of ultimate flexural strength of this composite following irradiation to ~70 dpa, which was attributed primarily to fiber damage [2, 3]. In addition, damage to the multilayer interphase was also indicated at low irradiation temperatures.

Based on these results, additional high dose irradiation experiments on newer generation nuclear grade SiC/SiC with different SiC fibers and the same interphase is planned to seek SiC/SiC irradiation tolerant beyond 100 dpa. The SiC/SiC with high-crystallinity and near-stoichiometric Tyranno SA3 SiC fiber is expected to be more irradiation tolerant compared to Hi-Nicalon Type S SiC/SiC due to the difference of grain size and impurity phases, based on a high dose ion-irradiation experiment [4].

This report presents capsule design, the current status of HFIR irradiation, and plans for the post-irradiation evaluations.

Experimental Procedure

<u>Materials</u>

Three types of SiC/SiC were prepared for this irradiation study. All the matrices were fabricated through chemical vapor infiltration (CVI) methods by Rolls-Royce High Temperature Composites Inc. (Customer P.O. Number: 4000124452, Rolls-Royce HTC, Inc Job Number: 13C-529). The first type was a composite with two-dimensional (2D) satin-weave Hi-Nicalon Type S fiber reinforcement in a 0°/90° stacking configuration. The second was a composite with 2D satin-weave Tyranno SA3 fiber reinforcement in a 0°/90° stacking configuration. The third was a composite with 0°/90° cross-ply laminated Ultra SCS fiber reinforcement. The fiber/matrix interphase was PyC for all composites. The nominal PyC thickness was 125 nm for the SiC/SiC with Type S and SA3 fibers, and 400 nm for the SiC/SiC with SCS fiber. The nominal porosities for all the SiC/SiC were less than 15%. Note that

fiber/matrix interphase used in this work was different from that used for the previous series of high dose irradiation experiments so that discussion of effect of the interphase type on the high dose irradiation stability will be possible using two types of Type S SiC/SiC. The SiC/SiC plates were machined to bar specimens with dimensions 25×2.8×1 mm^3. The appearances of the bar specimens are shown in Figs. 1 and 2. The SiC fibers were typically exposed on the surfaces of all the composites.

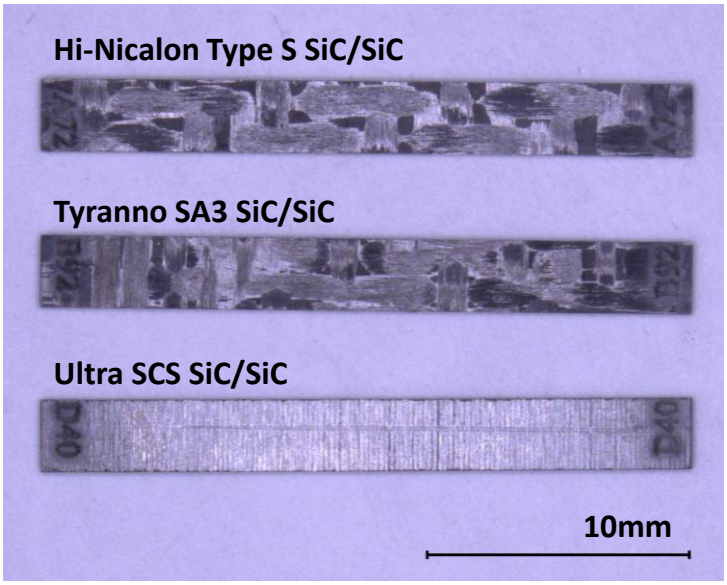


Figure 1. Appearance of SiC/SiC bend bar specimens used for this irradiation study.

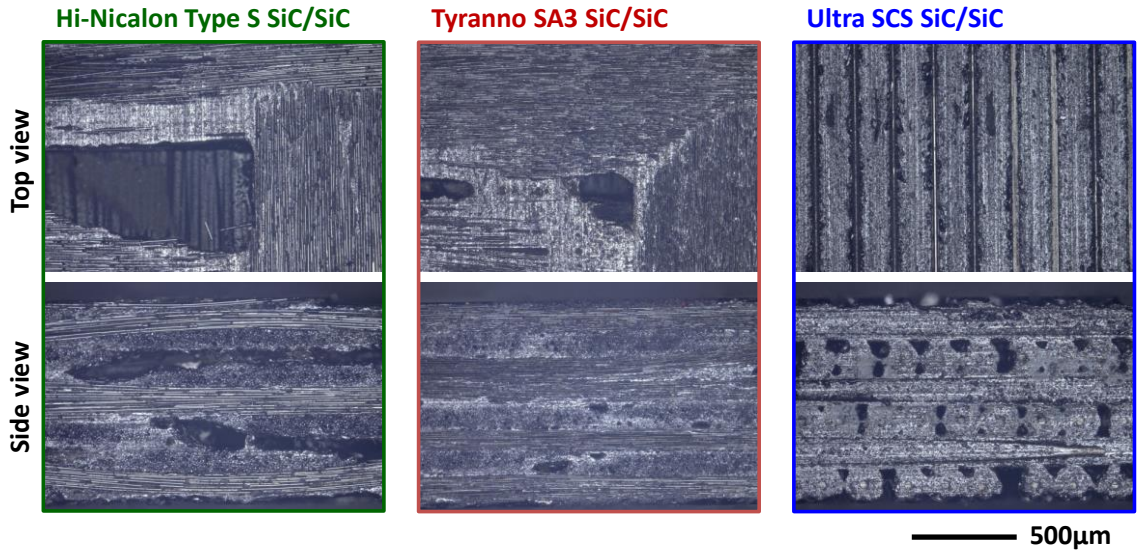


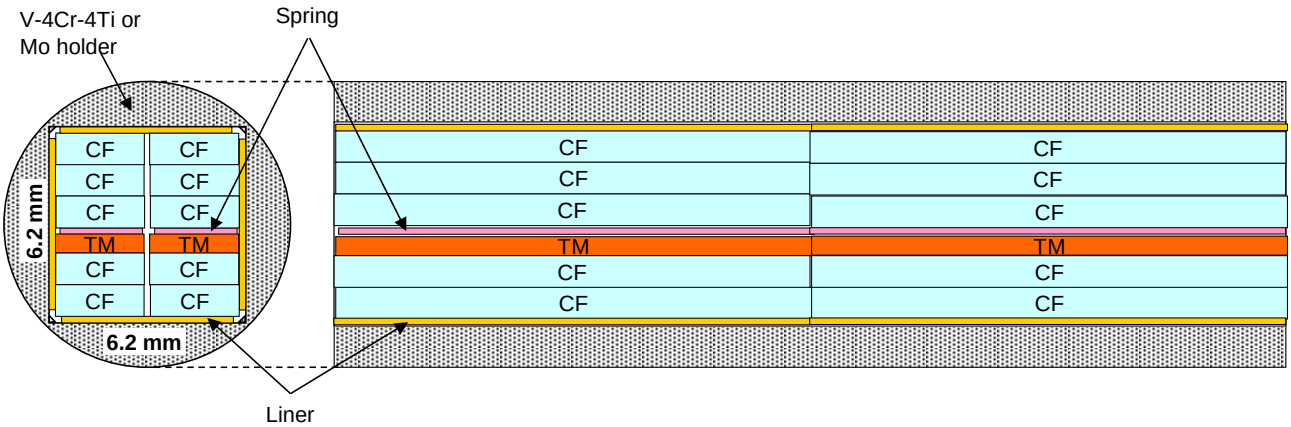
Figure 2. Top and side views of SiC/SiC bend bar specimens.

Irradiation Capsules

Schematic image of a capsule housing configuration is shown in Fig. 3. The specimen holders were made of V-4Cr-4Ti alloy or molybdenum. Each capsule contains 20 composite bar specimens (6 Type S,

7 SA3, and 7 SCS specimens), and 4 monolithic chemical vapor deposited (CVD) SiC (The Dow Chemical Company, High Resistivity Grade) specimens. Additional CVD SiC materials were located between SiC/SiC specimens and the holder to prevent any contamination of the specimens from the metallic holder during the irradiation. Springs made of CVD SiC are also used to maintain physical contact between the holder and internal SiC materials to control irradiation temperature.

Irradiation capsules were designed to control the specimen temperatures using three-dimensional thermal analysis with ANSYS software. The simulation results are shown in Fig. 4. The expected temperatures gradients in the SiC/SiC and CVD SiC specimens are less than 10% of target temperature for all irradiations. For irradiation at the target temperature of 950°C, the temperature of the SiC/SiC and CVD SiC specimens is expected to gradually decrease, depending on the dimensional change (swelling) of the molybdenum holder during irradiation [5]. The average irradiation temperature in the specimens will be ~900°C during the late stage of high does irradiation.



Type ID	Material	Dimensions[mm]	Qty	Main Purpose
CF	SiC/SiC	25x2.8x1	20	Flexure
TM	SiC	25x2.8x0.40	4	Thermometry
Spring	SiC	24 x 2.5 x 0.15	4	Spring
Liner	SiC	50x5.4x0.2	4	Separator

Figure 3. Schematic illustration of SiC/SiC specimens and SiC internal components in a metallic holder.

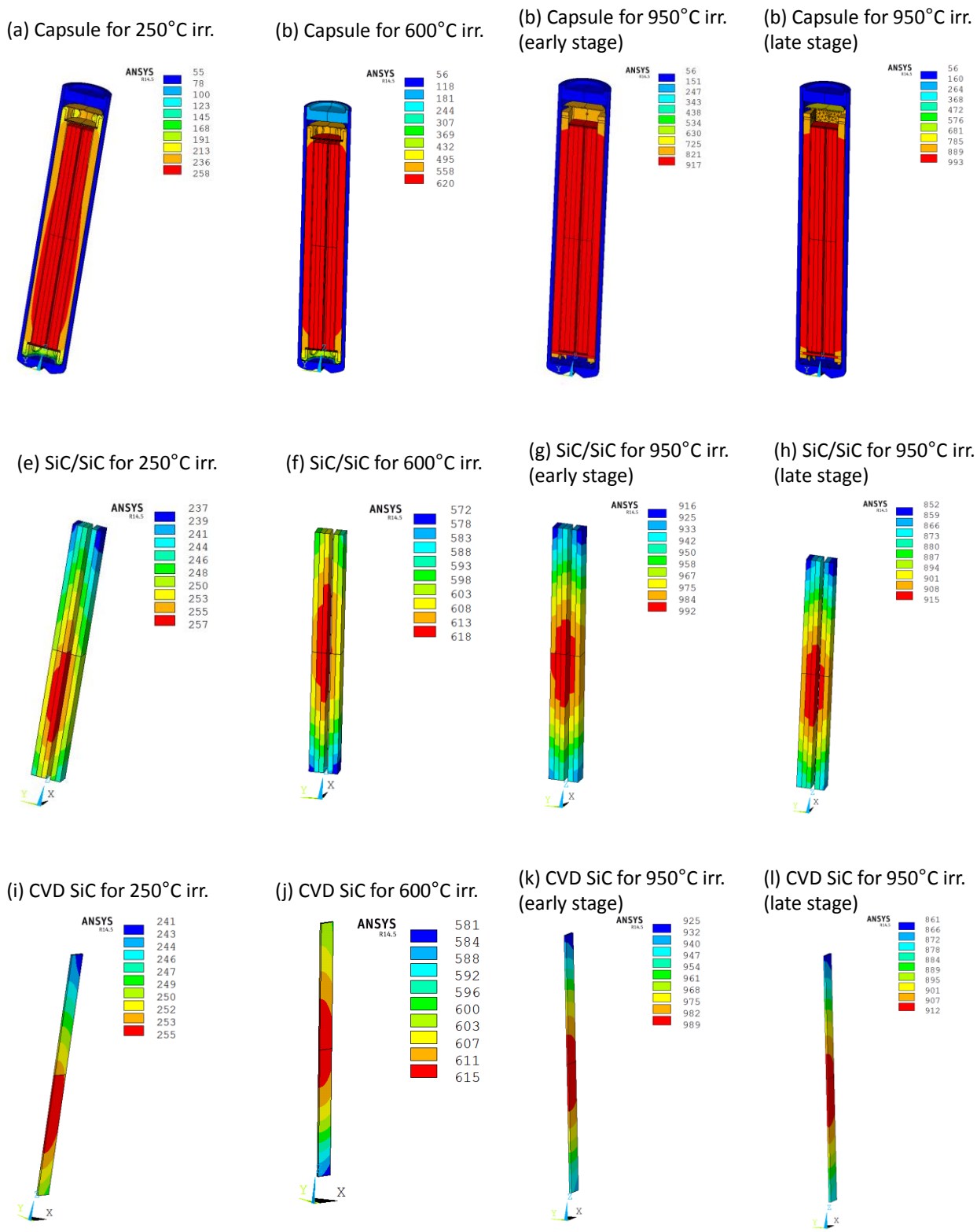


Figure 4. Simulation of temperatures in rabbit capsules, SiC/SiC specimens, and monolithic CVD SiC specimens designed for irradiation at ~250°C (a, e, f), ~600°C (b, f, j), ~950°C in the earlier stage (c, g, k), and ~950°C in the late stage (d, h, l).

Irradiation Matrix and the Schedule

This series of high dose irradiation experiments consists of irradiation of 12 rabbit capsules. All rabbits had the same capsule design, shown in Fig. 3. The planned target fluences are up to ~200 dpa , assuming the equivalence of 1 dpa with 1 x 10^{25} n/m^2 (E > 0.1 MeV). The target irradiation temperatures are 250, 600, and 950°C. The specimens are irradiated under inert gas atmospheres such as helium for 250°C irradiation, neon for 600°C, and argon for 950°C. The V-4Cr-4Ti holder is used for irradiation at 250 and 600°C. The molybdenum holder is used at 950°C. The irradiation matrix is summarized in Table 1.

The SCF1-9 rabbits will be irradiated starting in HFIR Cycle 457 (11/18/2014~), and the irradiations for SCF10-12 rabbits will start in Cycle 458 (1/13/2015~) [6].

Table 1. Irradiation conditions planned for high dose irradiation experiments.

RABBIT ID	TARGET TEMP. (°C)	HFIR POSITION		FAST FLUX (n/cm^2 s)	# OF CYCLES	FAST FLUENCE PER CYCLE (n/cm^2)	TOTAL FLUENCE (n/cm^2)	TARGET DPA
SCF1	~250	TRRH	6	9.50E+14	1	1.97E+21	1.97E+21	2
SCF2	~250	TRRH	6	9.50E+14	6	1.97E+21	1.18E+22	10
SCF3	~250	TRRH	5	1.04E+15	14	2.16E+21	3.02E+22	30
SCF4	~250	TRRH	4	1.08E+15	45	2.24E+21	1.01E+23	100
SCF5	~250	TRRH	4	1.08E+15	90	2.24E+21	2.02E+23	200
SCF6	~600	TRRH	6	9.50E+14	6	1.97E+21	1.18E+22	10
SCF7	~600	TRRH	5	1.04E+15	14	2.16E+21	3.02E+22	30
SCF8	~600	TRRH	4	1.08E+15	45	2.24E+21	1.01E+23	100
SCF9	~600	TRRH	4	1.08E+15	90	2.24E+21	2.02E+23	200
SCF10	~950	TRRH	5	1.04E+15	5	2.16E+21	1.08E+22	10
SCF11	~950	TRRH	5	1.04E+15	14	2.16E+21	3.02E+22	30
SCF12	~950	TRRH	4	1.08E+15	45	2.24E+21	1.01E+23	100

Plan of Post-Irradiation Experiments

The post-irradiation examination is planned to include evaluation of flexural strength, Young's modulus, swelling, thermal diffusivity at room temperature, electrical conductivity up to the irradiation temperature for the composite specimens, and microstructures. The flexural tests will be conducted in accordance with the guideline of ASTM standard C1341-13. Dynamic Young's modulus will be investigated by impulse excitation of vibration according to ASTM C1259-14. Swelling will be determined by apparent density measurement before and after irradiation. Thermal diffusivity will be measured using a flash diffusivity technique, per ASTM E1461-13. Electrical conductivity will be measured in a four-probe configuration for the in-plane orientation and in a two-probe configuration for the through-thickness orientation [7]. The microstructures will be investigated using optical and electrical microscopes. Properties of the monolithic CVD SiC TM specimens in Fig. 3 will be also investigated to understand the irradiation stabilities of the matrix of CVI SiC/SiC. The swelling will be evaluated by the density-gradient technique based on ASTEM D1505-10. Room temperature thermal diffusivity will be also measured. The actual irradiation temperatures will be determined by the post-irradiation annealing of the CVD SiC specimens [8].

References

[1] Y. Katoh, T. Nozawa, L.L. Snead, K. Ozawa, H. Tanigawa, J. Nucl. Mater., 417 (2011) 400-405.
[2] A.G. Perez-Bergquist, T. Nozawa, C. Shih, K.J. Leonard, L.L. Snead, Y. Katoh, J. Nucl. Mater. (2014), doi:10.1016/j.jnucmat.2014.06.038
[3] Y. Katoh, T. Nozawa, C. Shih, K. Ozawa, T. Koyanagi, W. Porter, L. L. Snead, "High Dose Neutron Irradiation of Hi-Nicalon Type S Silicon Carbide Composites, Part 2: Mechanical and Physical Properties", submitted to Journal of Nuclear Materials.
[4] S. Kondo, T. Hinoki, M. Nonaka, K. Ozawa, Acta Materialia, 83 (2015) 1-9.
[5] B.V. Cockeram, R.W. Smith, N. Hashimoto, L.L. Snead, J. Nucl. Mater., 418 (2011) 121-136.
[6] http://neutrons.ornl.gov/facilities/HFIR/pdf/OperatingForecast.pdf
[7] Y. Katoh, S. Kondo, L.L. Snead, DC Electrical Conductivity of Silicon Carbide Ceramics and Composites for Flow Channel Insert Applications, J. Nucl. Mater., 386-388 (2009) 639-642.
[8] L.L. Snead, A.M. Williams, A.L. Qualls, Revisiting the Use of SiC as a Post Irradiation Temperature Monitor, in: M.L. Grossbeck, T.D. Allen, R.G. Lot, A.S. Kumar (Eds.) The Effects of Radiation on Materials: 21st International Symposium, ASTM STP 1447, ASTM International, West Conshohocken, PA, 2003.