

THE HFIR 14J SiC/SiC COMPOSITE AND SiC FIBER COLLABORATION -
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OBJECTIVE

The objective of this report is to summarize the planned tests of the SiC composite and fiber specimens selected for the upcoming JUPITER 14J irradiation experiment in the HFIR reactor.

SUMMARY

A short introduction with references establishes the current status of research and development of SiC_f/SiC composites for fusion energy systems with respect to several key issues. The SiC fiber and composite specimen types selected for the JUPITER 14J irradiation experiment are presented together with the rationale for their selection.

PROGRESS AND STATUS

Introduction

The HFIR/14J neutron irradiation experiment is part of the U.S./Monbusho collaboration. This report describes the portion of the collaboration that will examine the irradiation behavior of SiC composites and SiC-based fibers. The HFIR/14J experiment will be the third in a series of such experiments [1]. The first irradiation in this series was carried out in the EBR-II reactor during 1993-94 as part of the COBRA 1A2 experiment. In COBRA 1A2, the SiC-type specimens were irradiated at 800°C to a relatively high dose of 80 dpa-SiC. The just-completed second irradiation in this series was carried out as part of the HFIR/11J-12J experiment at 300 and 500°C to a dose of ~10 dpa-SiC. PIE tests for the 11J-12J experiment will be performed during 1999. The HFIR/14J irradiation is scheduled to commence in January, 1999 and will be carried out at 300, 500 and 800°C to a dose of ~10 dpa-SiC, the same dose as for the HFIR/11J-12J experiment.

Results from the COBRA 1A2 SiC experiment are described in detail in previous semiannual progress reports [2-4]. At the time of this experiment, the most advanced continuous fiber-reinforced SiC composite (SiC_f/SiC) was made by Dupont using the isothermal chemical vapor infiltration (ICVI) method. This composite was made with ceramic grade (CG) Nicalon™ fiber coated with a 150 nm pyrocarbon (PyC) interface. A 3D surface showing a proposed strength-irradiation dose-temperature dependence derived from experimental data for this "reference" material is given in [2]. The topological features of the 3D surface suggested three degradation mechanisms for the composite, all of which were related to the thermochemical and structural instability of the Nicalon CG fiber in a neutron irradiation environment. The approximately 50% strength degradation of the Dupont SiC_f/SiC reference material irradiated at 800°C was related to a mass loss mechanism whereby the fiber oxygen constituent reacted with carbon, either within the fiber amorphous Si-C-O

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phase or as the PyC interface, and was exsolved as CO gas [3]. Together with the mass loss mechanism, Nicalon CG fibers irradiated at temperatures exceeding 800°C exhibited crystallization and grain growth, which were correlated to observed decreases in fiber tensile strength [5]. Thus overall, irradiated Nicalon CG fiber loses strength and shrinks, and it debonds from the CVI matrix in a composite made with this fiber. In other experiments at lower irradiation temperatures (<500°C), a relatively large differential SiC matrix swelling and fiber shrinkage also caused fiber/matrix debonding and degradation of the composite strength [6]. Composite made with Nicalon CG fiber generally is considered unacceptable for fusion applications. It was suggested that replacing the Nicalon CG with Hi Nicalon™ fiber, a fiber with much reduced oxygen content and temperature stability to about 1200°C, would alleviate these problems in the composite. In fact, a composite made with Hi Nicalon fiber irradiated at 385°C to 1.1 dpa-SiC exhibited only a 20% strength degradation [7]. This SiC_f/SiC composite was made with a multilayer interface which also probably had a bearing on its somewhat improved mechanical behavior.

Even better irradiation stability is expected if a composite could be fabricated using more crystalline, stoichiometric SiC fibers such as Dow Corning Sylramic™, Nippon Carbon Hi Nicalon Type S™ or Ube Tyranno SA™, in which case composite matrix and fiber properties should be closely matched so that differential swelling or shrinkage would be minimized. Preliminary fiber density [8] and length change studies [4] before and after irradiation indicated expected swelling behavior for the irradiated Sylramic fiber. Unfortunately, the Sylramic fiber tensile strength decreased 50% after irradiation at 500°C to 2 dpa-SiC. This observed decrease in strength was related to the approximately 3% TiB₂ content of the Sylramic fiber [8]. Nicalon Type S fiber does not contain boron, however at this time composites made with Type S fiber have not been available for irradiation testing.

To date, neutron irradiation experiments primarily have emphasized the examination of the stability and strength degradation of SiC_f/SiC composites and SiC fibers [9,10]. Other key issues and some preliminary performance requirements have been identified for further examination in reference [11] as well as in the proceedings of the 1st and 2nd IEA-sponsored SiC/SiC composite workshops for fusion applications [12,13]. Papers reporting on the key issues of design and specimen testing, component fabrication, joining, thermal conductivity, irradiation creep, impurities and activation effects also appear in these proceedings and elsewhere [14-19].

Plans

Information on the capsule design for 14J is presented in a report by Grossbeck, et al., in this issue. The actual SiC specimen matrix includes 102 bend bars (described in Tables 1A-1C), 22 fiber tubes (described in Tables 2A-2B), 20 thermal diffusivity discs (described in Table 3), 40 TEM discs (described in Table 4) and 6 fiber creep BSR fixtures (described in Table 5). Also given in these tables is the primary focus of the study for each of the selected specimen types.

The new "reference" SiC_f/SiC was made by Dupont Lanxide Corp. using a conventional isothermal chemical vapor infiltration (ICVI) process. The fabric preform made with Hi Nicalon fiber was CVD-coated with a 150 nm thick PyC interface. The original plate was 2.3 mm thick, contained six plies of 2D 0-90 weave fabric and had a bulk density of 2.60 ± 0.04 g/cc. The plate was cut into several flexure bars and some thermal diffusivity discs. Room

Table 1A. Bend bars* (30 x 6.0 x 2.2 mm³) to be irradiated at 800°C

Composite Type	No	Supplier (Process)	Primary Focus
Hi Nic/150nm PyC/0-90	5	Dupont (ICVI)	"Reference" Composite
Hi Nic/porous SiC/0-90	4	ORNL (FCVI)	porous SiC interface
Hi Nic/multilayer/0-90	5	Hypertherm (ICVI)	multilayer interface
Sylramic/multilayer/0-90	5	Hypertherm (ICVI)	Stoichiometric fiber/multilayer
MER CVR/CVR/0-90	4	MER (CVR)	Improved thermal conductivity
MER CVR/Ceraset/0-90	4	MER (CVR-CVI)	Improved thermal conductivity
Nic CG/PyC/Guipeux 3D	5	SEP (ICVI)	3D weave architecture
Ti ₃ SiC ₂ (monolithic)	5	Drexel (sintered)	irradiation stability
CVD-B SiC (monolithic)	5	Mitsui (CVD)	Dense monolithic SiC reference
Nic S/PyC/0-90	5	NRIM (ICVI)	Stoichiometric SiC fiber
Nic S/PyC/0-90	5	ORNL (FCVI)	Stoichiometric SiC fiber
Nic S (chopped)/PyC	5	ORNL (FCVI)	Dense, improved T. Cond.
Nic S/PyC/0-90	5	Ube (PIP)	Stoich. fiber/ stability PIP matrix
Adv. PCS/multilayer/3D	5	? (Hot press)	Dense, improved T. Cond.
Other	5	? (?)	?

* 37/35 bend bars supplied by U.S./Japan, respectively.

Table 1B. Bend bars* (30 x 6.0 x 2.2 mm³) to be irradiated at 500°C

Composite Type	No	Supplier/Process	Primary Focus
Hi Nic/150 PyC/0-90	3	Dupont (ICVI)	"Reference" Composite
Nic S/PyC/0-90	5	ORNL (FCVI)	Stoichiometric SiC fiber
Nic S (chopped)/PyC	5	ORNL (FCVI)	Isotropic/hermetic
Nic S/PyC/0-90	5	Ube (PIP)	Stoich. fiber/ stability PIP matrix

* 3/15 bend bars supplied by U.S./ Japan, respectively.

Table 1C. Bend bars* (30 x 6.0 x 2.2 mm³) to be irradiated at 300°C

Composite Type	No	Supplier/Process	Primary Focus
Hi Nic/150 PyC/0-90	3	Dupont (ICVI)	"Reference" Composite
Nic S/PyC/0-90	5	ORNL (FCVI)	Stoichiometric SiC fiber
Nic S (chopped)/PyC	5	ORNL (FCVI)	Dense, improved T. Cond.
Nic S/PyC/0-90	5	Ube (PIP)	Stoich. fiber/ stability PIP matrix

* 3/9 bend bars supplied by U.S./ Japan, respectively.

Table 2A. Fiber tubes* (2.0 mm dia x 60 mm) to be irradiated at 800°C

Fiber Type	No	Supplier	Density (g/cc)	XRD-gs (nm)
Hi Nicalon	2	Nippon	2.69	4.4
Hi Nicalon (annealed at 1500°C/1hr)	2	Nippon	2.81	?
Hi Nicalon S	2	Nippon	3.08	11
Tyranno SA	2	Tyranno	?	?
Sylramic	2	Dow Corning	3.0-3.1	80
MER CVR	2	MER	2.8	<5
Other	3			

* 15 fiber tubes supplied by U.S. Each fiber bundle is 52.0 mm long.

Table 2B. Fiber tubes* (2.0 mm dia x 60 mm) to be irradiated at 500 and 300°C

Fiber Type	No	Supplier	Density (g/cc)	XRD-gs (nm)
Hi Nicalon	1	Nippon	2.69	4.4
Hi Nicalon S	1	Nippon	3.08	11
Sylramic	1	Dow Corning	3.0-3.1	80
Tyranno SA (300°C only)	1	Ube	?	?

* 7 fiber tubes supplied by U.S. Each fiber bundle is 52.0 mm long.

Table 3. Diffusivity discs* (10.0 mm dia x 2.0-2.5 mm) irradiated at 800, 500 and 300°C

Capsule /Type	Morton B-CVD	MER/Hybrid	MER/CVR	Nic S-2D (FCVI)	Nic S-chopped (FCVI)	Nic S-2D (PIP)	Total
800°C	2	2	2	3	3	2	14
500°C	2			1			3
300°C	2			1			3

* 12/8 discs supplied by U.S./Japan, respectively.

Table 4. TEM discs* (3.0 mm dia x 0.20 mm) to be irradiated a 800, 500 and 300°C

Capsule	Morton B-CVD	Primary Focus
800°C	20	Examine irradiation defect structure
500°C	10	plus effects of post-irradiation isochronal
300°C	10	annealing on this structure

* 40 discs supplied by U.S.

Table 5. Fiber Creep by BSR.* (Two tubes 44.5 mm dia x 11.5 mm) irradiated at 800°C

Fiber Type	Characteristic Temperature for Thermal Creep (1 Hr.)	Characteristic Temperature for Thermal Creep (100 Hr.)
Hi Nicalon S	1450°C	1260°C
Sylramic	1420°C	1260°C
Tyranno SA	?	?
Hi Nicalon	1230°C	1080°C
Hi Nicalon (annealed)	?	1230°C
MER CVR	1550°C	?

* 2 creep tubes each loaded with 3 different fiber types supplied by U.S.

temperature 4-pt bend strength, strain to failure and thermal conductivity values were measured to be 628 ± 22 MPa, 0.90 ± 0.03 % and ≈ 13 W/mK, respectively. These values represent a significant improvement over values of similarly processed unirradiated SiC_f/SiC made with Nicalon CG fiber. As part of this collaborative effort, samples of this material also will be irradiated in the JMTR reactor at 800 and 1000°C to 0.5 dpa and in the HFR/Petten reactor at 750°C to 1.2, 2.5 and 5.3 dpa. Other samples will be used in helium implantation studies [18]. Properties of this new reference SiC_f/SiC before and after irradiation will be compared to the properties of similarly fabricated SiC_f/SiC except with a thicker PyC interface (1000 nm), which currently is being irradiated in HFIR as part of the JUPITER 12J experiment.

In general, the overall dimensional and structural stability after irradiation will be examined for all the other composite variations. Four point flexure tests on the standard 30 x 6.0 x 2.2 mm³ bars will be carried out at the irradiation temperature to characterize and compare mechanical property behaviors. Changes in the composite stability and mechanical properties then will be related to the variations of fiber type, interface, architecture and matrix fabrication among the selected composites. Two monolithic SiC-based materials are included in the bend bar matrix. The Ti₃SiC₂ material has a graphitic-like structure [20], and potentially could be used as a CVD-applied fiber coating. The monolithic CVD β-SiC material is included to simulate the radiation performance of a composite matrix-like material.

Since the performance of the fiber has such a strong bearing on the overall radiation performance of the composite, bare fiber tows of each of the fiber types used in the composites will be irradiated and tested following procedures discussed in [21]. For irradiation, each fiber type is inserted into a separate protective tube (2.0 mm od by 1.0 mm id x 60 mm) made from Hexoloy™ sintered SiC. The fiber bundles themselves are carefully cut to a 52.0 mm length which provides sufficient material for length change, tensile strength, density and XRD analyses.

For several fusion component design options, the thermal conductivity should exceed 10-30 W/mK during irradiation. Therefore, a concerted effort has been carried out to optimize the thermal conductivity of some types of SiC composites while maintaining acceptable mechanical properties [12,13, 22]. For these specific types of materials, thermal diffusivity discs will be included in the matrix. To further analyze the fundamental behavior of phonon scattering by irradiation induced defects, several TEM discs made from Morton β-SiC will be

inserted at all three of the irradiation temperatures. Post-irradiation annealing of the TEM discs will allow analysis of the resulting defect structures.

Finally, irradiation enhanced creep will be examined for the most advanced SiC fiber types using a bend stress relaxation (BSR) method [23]. These data will be compared to the irradiation creep data obtained by Scholz using a dynamic torsion method [11,12].

FUTURE WORK

The characterization of the properties for the unirradiated SiC composite and fiber specimens will be continued through 1999.

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