PROGRESS IN THE DEVELOPMENT OF A SiC_f/SiC CREEP TEST- M. L. Hamilton, C. A. Lewinsohn, R. H. Jones, G. E. Youngblood, F. A. Garner (Pacific Northwest National Laboratory)[a], and S. L. Hecht (Westinghouse Hanford Company)

OBJECTIVE

The objective of this work is to determine the irradiation creep behavior of SiC_f/SiC composite materials. This requires the development of an appropriate creep specimen since pre-existing creep specimen geometries are not adequate.

SUMMARY

An effort is now underway to design an experiment that will allow the irradiation creep behavior of SiC_f/SiC composites to be quantified. Numerous difficulties must be overcome to achieve this goal, including determining an appropriate specimen geometry that will fit in the irradiation volumes available and developing a fabrication procedure for such a specimen. A specimen design has been selected, and development of fabrication methods is proceeding. Thermal and stress analyses are being performed to evaluate the viability of the specimen and to assist with determining the design parameters. A possible alternate type of creep test is also being considered. Progress in each of these areas is described in this report.

INTRODUCTION

An earlier report demonstrated that monolithic SiC exhibited irradiation creep at 800-1000°C that was very similar to that observed in metals,[(1)] indicating that there is a need to understand both the mechanism and the technical implications of irradiation creep in SiC. It is anticipated that a creep strain of 0.1% will result in a composite component after only three years of operation at a stress of 100 MPa; this is a significant strain for a composite such as SiC_f/SiC. Lower creep rates are possible with lower stresses but this would be a severe performance limitation for SiC_f/SiC.

It is therefore considered necessary to quantify the irradiation creep behavior of SiC_f/SiC composites in this temperature regime. The pressurized tube geometry that has come to be considered standard for irradiation creep studies of metallic specimens is not appropriate for composites since the material is not fully dense and since welded closure of the tube is impossible. Thus development of an appropriate specimen is a necessary prerequisite for determining irradiation creep in composites. The majority of the effort to date has been in this direction.

The difference in the relative creep rates of monolithic SiC and SiC fibers suggests that creep of the composite will be dominated by creep of the fibers, so a parallel effort has also been initiated to determine the irradiation creep properties of SiC fibers. The results of the latter effort are also documented elsewhere in this progress report and will not be discussed further here.[(2)] Because obtaining irradiation creep data on SiC_f/SiC requires development of a novel test specimen to utilize the available irradiation facilities in the U.S., a parallel effort has been initiated to determine whether such data might be generated using another irradiation facility. Progress in this area is also described below.

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PROGRESS

Specimen Development

After considering various possible specimen geometries, a geometry similar to that of the standard pressurized tube was selected for development. The geometry that was selected comprises an open-ended SiC_f/SiC tube with a pressurized metallic bladder placed inside the tube (see Figure 1). A stress state that is nominally biaxial (but which has yet to be fully analyzed) is produced in the tube when the assembly is heated and intimate contact is obtained between the outer composite sleeve and the inner pressurized bladder. A hoop stress is produced in the sleeve via the pressurized bladder, while an axial stress is expected from the friction between the two. The ratio between the hoop and axial stresses, however, is expected to be higher than the 2:1 ratio found in a standard pressurized tube specimen, and therefore the effective stress will be even closer to the hoop stress than in a standard specimen, where the effective stress is $0.866\sigma_\theta$.

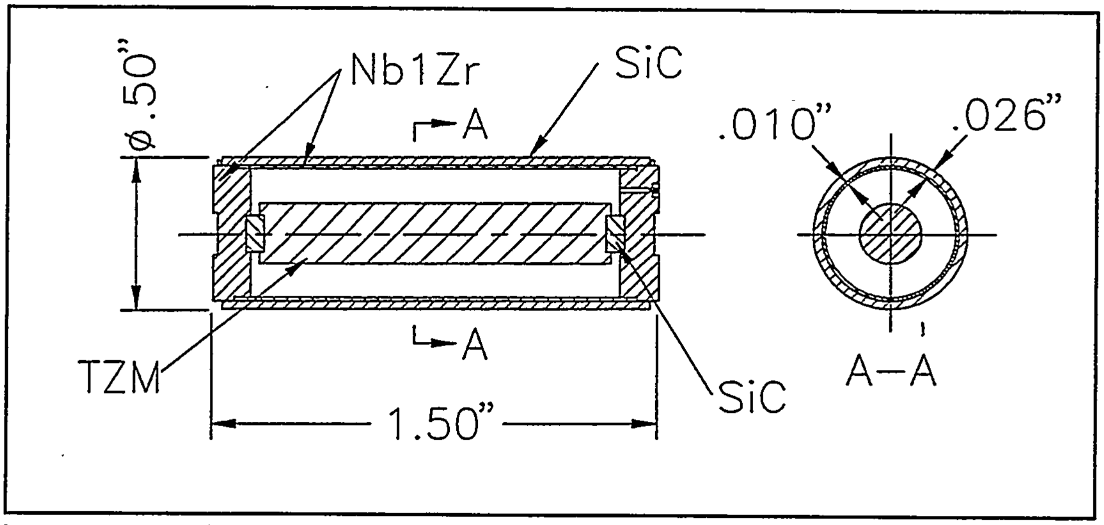

Figure 1. Schematic of the composite creep specimen design.

A number of significant fabrication and irradiation issues surfaced as development of such a specimen proceeded. Specimen volume is constrained by the space available in the proposed irradiation facility, HFIR. The choice of materials from which the bladder can be constructed is constrained by the likely effect of transmutations anticipated in HFIR, by the paucity of irradiation creep data in the temperature range of interest, and by the possible chemical interactions between the composite tube and the bladder. Additionally, fabrication of the composite in a tubular geometry is not trivial, although it has recently been demonstrated that such tubes can probably be made successfully.[3]

The material selected for the bladder is Nb-1Zr. Irradiation creep data were generated for this alloy in FFTF at temperatures ranging from about 1000-1200°C. Regressions were fit to these data, which were extrapolated to the temperatures of interest to allow the thermal and stress analyses described below to be performed for the proposed irradiations. Thermodynamic calculations indicated that the likelihood of chemical interaction between the sleeve and the bladder was high at the temperatures of interest. Some experimental effort has therefore been devoted to determining the magnitude of this interaction and to developing a coating for the bladder to prevent it. Performance predictions were made to determine the structural behavior of the proposed creep specimen to determine if the concept of using an internal bladder to drive creep of the surrounding specimen was viable. When the feasibility of this type of test had been demonstrated, parametric studies were performed to improve the design and to assess sensitivities. These issues are discussed in more detail in the following paragraphs.

Investigation of reactivity between β-SiC and Nb-1Zr

Data on the chemical compatibility between Nb-1Zr and SiC are not available in the literature. The reactivity between Nb-1Zr and SiC was therefore investigated using a computer program (CHEMSAGE) capable of calculating the Gibbs free energy of formation and rate of reaction for all compounds within the program database that can exist for a given thermodynamic state (pressure, temperature, number of moles). The actual reaction that will occur, however, will be limited by the kinetics of the transport mechanisms and the chemical reactions. After initial calculations indicated that the likelihood of reaction was high, a series of diffusion couple experiments was initiated to gather information on the kinetic aspects of the reactivity between Nb-1Zr and SiC.

Initially, the thermodynamic stability of Nb-1Zr and β-SiC (the phase present in the matrix of the chemical vapor infiltrated composites) was calculated at the lower end of the range of temperature of interest: at 800°C, with 1.01 bar total pressure, in pure helium. At this temperature, a niobium carbide (Nb_8C_7) and a niobium silicide ($NbSi_2$) had a negative Gibbs free energy of formation, as did zirconium carbide (ZrC_4), indicating that a reaction that produces these phases is energetically favorable. This does not imply that the reaction will proceed, however, since kinetic limitations must also be considered. Note that the thermodynamic calculations do not indicate metastable, or intermediate, phases that may also form.

Results of the initial thermodynamic calculation indicate that a diffusion barrier coating must be placed between the Nb-1Zr and the β-SiC to prevent a detrimental reaction from occurring during the creep experiments. Additional thermodynamic calculations were therefore performed to identify a candidate coating material. A survey of in-house coating specialists and commercial coating vendors was conducted to identify materials and fabrication methods that would be suitable for the experiment of interest. Zirconia (ZrO_2), alumina (Al_2O_3), and a proprietary aluminide coating (aluminum oxide-based with minor amounts of chromium, iron, and titanium) were selected as candidate materials. The additional calculations indicate that the most suitable coating material appears to be alumina (Al_2O_3). Alumina is stable with respect to both β-SiC and Nb up to 1000°C in pure helium (1.01 bar total pressure), but it will react with Zr at 800°C in pure helium (1.01 bar total pressure) to form zirconia (ZrO_2). Nevertheless, an alumina coating offers the best balance between ease and availability of fabrication, chemical compatibility, and ability to obtain the small coating thickness needed to minimize disruption of stress transfer from the bladder to the composite sample.

An experimental approach is being used to determine the kinetic stability of an alumina coating with Nb-1Zr and β-SiC. Since the Zr is in solid solution with the Nb, the reaction rate is expected to be slow. Fixture design and analytical methods have been verified for these experiments. Samples of Nb-1Zr and β-SiC (fabricated by chemical vapor deposition by Morton Advanced Materials, Woburn, MA), were placed in a U-shaped Mo fixture and held in flowing, gettered argon (<0.1 ppm O_2) at 1000°C for 50 hours. The fixture was designed so that the Nb-1Zr and β-SiC specimens were held together by thermal expansion at pressures comparable to those anticipated in a creep test. Although the pieces were not bonded after cooling to room temperature, energy dispersive x-ray analysis of a reaction layer on the Nb piece, performed with an analytical electron microprobe, indicated the presence of Si as much as 3 μm from the surface. These experimental results confirm the initial thermodynamic calculations and emphasize the need for a coating material.

Based on the results of the thermodynamic calculations and experiments described above, additional experiments will be performed to investigate the suitability of using alumina as a diffusion barrier coating. Alumina coatings will be deposited onto Nb-1Zr samples via sputter deposition at PNNL. These samples will be tested in the fixtures described above and analyzed by optical microscopy, scanning electron microscopy, and an analytical electron microprobe. The minimum thickness required to prevent reaction between Nb-1Zr and β-SiC will be determined. If alumina does not prevent Nb-1Zr and β-SiC from reacting, then the proprietary aluminide coating will be evaluated further.

Performance Predictions

Finite element analysis of the proposed experiment design modeled the pressurized Nb-1Zr thin-walled bladder, the composite specimen tube, and the interaction between the two tubes. A two-dimensional axisymmetric model using axisymmetric shell elements for the tubes and gap elements for the interaction were employed. The model considered internal pressure loading on the bladder tube, thermal expansion of both tubes, irradiation creep of both tubes, and irradiation swelling of the Nb-1Zr tube. Because irradiation creep and swelling correlations did not exist for the Nb-1Zr material at the temperatures of interest, regression equations were developed from data given in the space power program literature.

Analysis cases were run for variations in Nb-1Zr tube thickness, irradiation creep and swelling to account for uncertainties. Both 800°C and 1000°C full power temperatures were analyzed for all variations in other parameters. Neutron fluxes were assumed to be those for the HFIR reactor RB* location. Results showed that the experiment is indeed feasible and that the hoop stress in the composite tube varied less than 5% over a period of 300 effective full power days in the HFIR reactor. For the specimen geometries that were assumed, the calculated permanent diametral change in the specimen at the end of the test was on the order of 5 μm, a value large enough to be measurable using existing equipment. Uncertainties in the Nb-1Zr material properties did not have a significant effect on the experiment parameters.

Test Capsule Design and Thermal Analyses

Two locations in the HFIR reactor are being considered for the composite creep test, the RB* and the PTP locations. Each test location has a distinct capsule design. A scoping, finite element thermal analysis was made for each design. These analyses were 2-dimensional and axisymmetric, and considered internal heat generation due to gamma heating, conduction within the specimen/bladder assembly and capsule, thermal radiation across gas gaps, and convection to the reactor coolant.

For the RB* location, a configuration using two subcapsules, an inner and an outer one, was envisioned. The inner subcapsule would contain three creep specimens as well as other composite specimens such as bend bars and fibers. It is desired that the contents of the subcapsule be at approximately the same test temperature; analyses were run for irradiation temperatures of 500, 800 or 1000°C. The subcapsule was assumed to be filled with helium for maximum heat transfer and uniformity in temperature. The subcapsule wall (either stainless steel or TZM depending on the temperature) would provide much of the gamma heating to the subcapsule contents. The inner subcapsule would also provide structural axial constraint to the creep tube so that the Nb-1Zr pressurized bladder would not fail from creep rupture in the axial direction. The outer subcapsule would isolate the heated inner subcapsule from the reactor coolant. An annular gas gap between the two (to be filled with an argon-helium mixture) will be used for temperature control. At this time both passive (fixed gas mixture) and active (variable gas mixture) temperature control systems are being evaluated.

The initial thermal analysis focused on the passive design. The results of the analysis showed that by varying the thickness of the subcapsule wall, the annular gas gap and the gas mixture, the desired temperatures within the inner subcapsule could be obtained. It is assumed for analytical purposes that if a passive temperature control system is successful, an active system would also be successful and would provide the additional control needed to compensate for uncertainties in quantities such as gamma heating rates and emissivities.

The PTP capsule design comprises a single creep tube specimen in a subcapsule at each axial location. Most of the internal heat to the specimen would be provided by a TZM ballast located within the pressurized Nb-1Zr bladder. The outer subcapsule, which is to be in contact with reactor coolant water, will provide structural axial constraint for the Nb-1Zr tube. Argon gas is used as an insulator between the specimen system and the outer subcapsule.

Thermal analysis for the PTP test capsule showed that acceptable temperatures could be obtained in the specimens, although an axial gradient would exist. It was also demonstrated that the desired temperature could be obtained by varying the ballast size.

Alternate Irradiation Facility

One of the difficulties involved in designing irradiation creep experiments on SiC_f/SiC composites arises from the high gamma heating levels associated with the mixed spectra reactors currently available in the U.S. Most importantly, such high heating rates severely limit the allowable size and geometry of the creep specimen, in addition to imposing the constraint of symmetry. The much lower heating rates available in fast reactors allow larger specimens, larger deformations and non-symmetrical geometries. Unfortunately, fast reactors in the U.S. have been shut down. The possibility of using a reactor like the BR-10 fast reactor located at the Institute of Physics and Power Engineering in Obninsk, Russia, is therefore being considered and appears to hold promise.

The staff are eager for the work and are quite competent to perform tests on specimens similar to those currently used in the U.S. to generate thermal creep data on composites. Creep experiments in which measurements are made in-situ have been conducted for many years in BR-10 at temperatures as high as 600°C for extended periods on stainless steels and on a V-Cr-Ti alloy. An irradiation temperature of 800°C can be reached with supplementary electrical heating, which would require a small amount of modification to existing facilities. An on-site visit and review of the specimen design, available creep data, required strain rates and other considerations led to the conclusion that in-situ tests could be easily conducted on composite materials in BR-10.

Two types of creep tests can be performed in this facility. The most convenient test is torsion loaded, but this test mode is not representative of anticipated service conditions for SiC_f/SiC. In a torsion test mode, the noise arising from minor thermal fluctuations is much smaller than in tension, and the specimens are best configured as tubes to provide a relatively constant stress state throughout the specimen. Tension tests may be more suitable for SiC_f/SiC, but extra precautions are required to reduce transient noise arising from thermal fluctuations. Either flat or round tensile specimens are acceptable for such tests.

CONCLUSIONS

A specimen geometry has been selected for an irradiation creep test on composite materials, and model calculations and initial experiments confirmed the feasibility of the specimen design as well as the experimental design. An alternate creep test is also being considered in the Russian BR-10 reactor.

FUTURE WORK

Development of the creep specimen will continue and thermal control tests will be run on prototype control specimens to demonstrate the specimen viability. Design of the irradiation test will also continue.

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