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X-RAY CHARACTERIZATION OF ATOMISTIC DEFECTS CAUSING IRRADIATION CREEP OF SIC

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ACRONYMS

ATF	accident-tolerant fuel
CVD	chemical vapor deposited
DOE	Department of Energy
dpa	displacement per atom
HFIR	High Flux Isotope Reactor
LWR	light water reactor
NSLS-II	National Synchrotron Light Source-II
PDF	pair distribution function
rpm	revolutions per minute
SiC	silicon carbide
XPD	X-ray powder diffraction
XRD	X-ray diffraction

ABSTRACT

Silicon carbide (SiC) fiber–reinforced SiC matrix composites have been widely studied for use in developing an accident-tolerant fuel (ATF) cladding for light water reactors (LWRs) in recent years because of their superior oxidation and irradiation resistance and low neutron absorption cross section. An important task is the establishment of the thermo-mechanical analysis capability to predict the in-pile performance and failure probability of the cladding. Irradiation creep is known to be a key property in modeling the stress state under irradiation. To better understand the radiation creep mechanism from an atomic point of view, this study conducted synchrotron-based X-ray diffraction (XRD) experiments on SiC neutron-irradiated with and without applied stress at the National Synchrotron Light Source-II (NSLS-II). This report presents progress of the XRD study and future plans. The important finding obtained was that the macroscopic creep strain and strain due to the lattice expansion were similar in case of neutron irradiation at 300°C to 0.1 displacement per atom (dpa), which indicates that the lattice strain is a major source of the transient creep strain.

1. INTRODUCTION

SiC-based ATF claddings for LWRs continue to undergo development worldwide because of their inherent high-temperature oxidation resistance, relatively low neutron absorption cross section, and irradiation resistance [1]. An important task in their development is the establishment of a thermomechanical analysis capability to predict the in-pile performance and failure probability of the SiC based cladding [1]. Irradiation creep is known to be a key property for modeling the stress state under irradiation. In short, and especially for SiC materials that possess very limited strain tolerance, irradiation creep provides an important stress mitigating function.

The PI's previous study evaluated neutron irradiation creep of SiC materials at 380–800°C up to 30 dpa, using a bend stress relaxation test in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory [2,3]. In addition, an in-pile instrumentation creep experiment on SiC at 300°C in the Halden reactor was conducted by the PI [4]. These previous studies [2,3] proposed that irradiation creep behavior can be modeled by summing the swelling-coupled primary creep and the secondary creep. However, the microstructures observed by conventional transmission electron microscopy could not fully support our model, potentially because of the presence of atomistic defects that could not be observed due to resolution limitations. Since the creep mechanism is not physically well understood, a previous parametric thermomechanical analysis of irradiated SiC composites did not provide an accurate result [5].

To better understand the radiation creep mechanism from an atomic point of view, we propose to conduct synchrotron-based XRD experiments on SiC that has been neutron-irradiated with and without applied stress at a beam line in the NSLS-II facility in Brookhaven National Laboratory. The world-class high brightness of the X-ray source and the high-resolution analysis capability at the requested XRD beam line enables an accurate determination of the structure of the irradiated SiC materials. This structural information is sensitive to atomistic defects [6]. XRD is an appropriate technique for evaluating irradiation creep strain because radiation-induced lattice strain, which can be evaluated by XRD, is mostly responsible for the dimensional change in SiC irradiated under a point defect swelling regime (~200–~800°C) [7]. The difference in lattice strain between SiC irradiated with and without applied stress is defined as XRD-based radiation creep strain. Therefore, precise evaluation of the lattice strain, together with identification of the strain origin, is a pathway to investigate the details of irradiation creep in SiC.

This report presents the progress of the conventional and pair distribution function XRD experiments and analysis.

2. EXPERIMENTS

Polycrystalline, chemical vapor deposited (CVD) 3C-SiC (high resistivity grade, 99.9995% pure purchased from Dow Chemical Co.) samples were irradiated in the HFIR or Halden reactor. A bulk specimen rather than a powder specimen was used for the XRD experiments. The initial average grain size was a few microns with no strong grain texture. The irradiation temperature and neutron fluence ranged from 300 to 800 °C and 0.1 to 30×10^{25} n/m^2 (E >0.1 MeV), respectively. These fluences correspond with nominal fluence of 0.1 to 30 dpa in SiC. Several of the specimens were irradiated with applied flexural and tensile stresses in the HFIR and Halden reactors, respectively. The bend creep specimens for the XRD experiment were prepared from uniaxially tensile-stressed areas. The bent specimens were thinned to ~30 μm from the compressive side using mechanical polishing. For the tensile creep specimens, the gauge section of the irradiated tensile bars was machined into disc specimens. The X-ray beam direction was parallel to the specimen thickness direction. Five and seven specimens from the Halden irradiation were evaluated for irradiation with applied stresses of 100 and 0–5 MPa, respectively. Details of the irradiation experiments can be found elsewhere [2–4].

XRD and PDF measurements were performed at the NSLS-II using the high-energy X-rays available at the X-ray Powder Diffraction (XPD) beamline. All measurements were performed in transmission mode with an amorphous silicon-based flat panel detector (Perken-Elmer) mounted orthogonal to and centered on the beam path. Figure 1 shows appearance of the XPD beamline. The sample-to-detector distances and tilts of the detector relative to the beam were refined via Rietveld refinement of a diffraction pattern of a LaB6 powder standard (NIST standard reference material 660c) using a BRUKER TOPAS software. The wavelength of the incident X-rays was 0.2370 Å (52.3149 keV). The sample-to-detector distance was calculated to be 1351.94 and 199.29mm for the XRD and PDF measurements, respectively. The samples position in the beamline was reproducible to within a few microns.

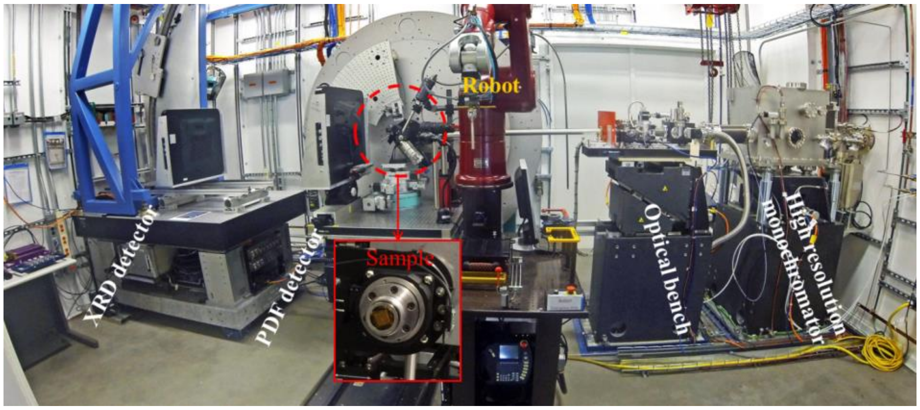

Figure 1 XPD beamline endstation at NSLS-II.

The tensile creep samples were rotated during the collection to limit the effect of any preferred orientation and to give powder like diffraction rings. During the rotation, the sample-to-detector distance was kept constant. The typical rotation speed was ~30 rpm. Multiple patterns were collected to avoid saturation of the detector with typical count times of 0.5-1 second, depending on the sample. All raw two-dimensional patterns were background corrected by subtracting the dark current image and any air scattering within Igor Pro (Wavemetrics). Noticeable artifact regions of the detector (like the beam stop, dead pixels) were masked. The corrected and masked two-dimensional detector images were then radially integrated to obtain the one-dimensional powder diffraction patterns and then the background subtracted XRD patterns were fit using Rietveld refinement with the BRUKER TOPAS software. The peak profiles were modeled by a modified pseudo-Voigt function. The instrument contribution to the broadening of the measured profiles was quantified by fitting a LaB_6 NIST powder standard, with known crystalline-domain size and negligible strain contribution. Typical uncertainty of lattice constant obtained by Rietveld refinement was 0.005%. . PDFgetx3 was used to convert the reduced 1D XRD patterns to G(r) over a range of 0.7 -23 $Å^{-1}$. PDFGui was used to refine the PDFs. The lattice parameter, Debye Waller factor and scale parameters could vary. The coherent grain size was initially allowed to vary but returned very large values (and no difference compared to the unirradiated samples) and was thus not included as a fitting parameter.

3. RESULTS AND DISCUSSIONS

Diffraction spectra for conventional XRD and PDF analysis were successfully obtained for the SiC specimens nonirradiated as well as with and without applied stress neutron-irradiated. This report focuses on the XRD results of the tensile creep specimens irradiated in the Halden reactor. Figure 2 shows XRD patterns averaged among the specimens for each test condition. All the specimens exhibit a β-SiC phase with an F-43m crystal structure. A small shoulder peak at ~5.1 degrees is also a common feature, which indicates the presence of stacking faults [8]. The XRD patterns show that neutron irradiation results in a clear shift of the peaks to lower two-theta angles. In addition, the applied stress during irradiation causes a further peak shift to lower angles as shown in the enlarged figure, although the peak shift is small. These peak shifts to lower two-theta angles are indications of a lattice expansion due to the inverse relationship between the two values. The lattice constant, coherent grain size, and anisotropic micro-strain determined by Rietveld refinement together with macroscopic swelling and creep results are summarized in Table 1. In this study, irradiation creep strain (ε_{IC}) is given by

$$\varepsilon_{IC} = \varepsilon_{100MPa} - \varepsilon_{0MPa}, \quad (1)$$

where ε_{100MPa} and ε_{0MPa} are the total strains of the specimens with and without100 MPa applied tensile stress during irradiation, respectively. Irradiation-induced lattice expansion was 0.43% without applied stress. According to Eq. (1), the irradiation creep strain due to lattice expansion was 0.028±0.014%, which was the difference of lattice strain with and without 100MPa tensile stress. This value was consistent with the macroscopic creep strain of 0.06% obtained from the out-of-pile macroscopic measurements using a digital microscope. The effects of irradiation on the coherent grain size and micro-strain were limited regardless of the applied stress level.

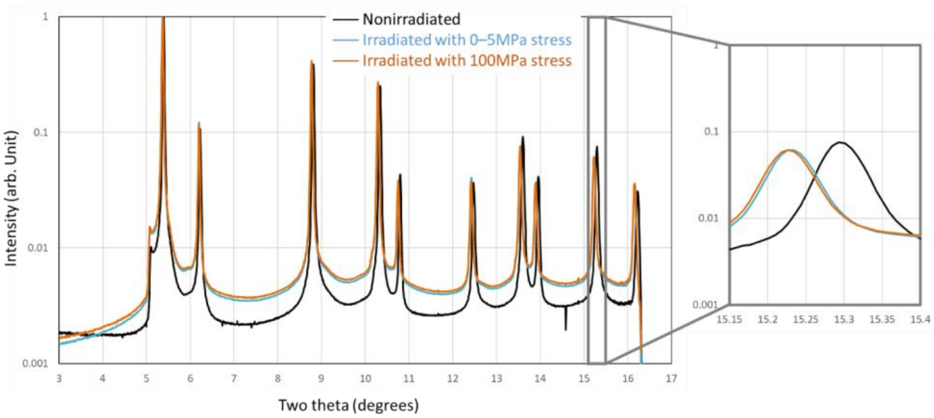

Figure 2 Averaged XRD patterns of SiC unirradiated and irradiated at 300°C to 0.095 dpa with applied stress of ~0 and 100 MPa.

Table 1 Summary of results of post-irradiation macroscopic strain measurement and XRD analysis

Test condition	Applied stress during irradiation	XRD analysis			Irradiation-induced strain [%]
		Lattice constant [Å]	Coherent grain Size [nm]	Anisotropic micro strain	
Unirradiated	Not available (NA)	4.361537 (0.000581)	139.9 (56.0)	0.0353 (0.0035)	NA
Irradiated at 300°C to ~0.1dpa	~0 MPa	4.380248 (0.000529)	110.7 (6.2)	0.0428 (0.0089)	**Swelling** Out-of-pile macro: 0.42 (0.04*) Lattice (XRD): 0.429 (0.012)
	100 MPa	4.381455 (0.000601)	99.1 (16.1)	0.0420 (0.0122)	**Creep** Out-of-pile macro: 0.06 (0.04*) Lattice (XRD): 0.028 (0.014)

Note: Parentheses indicate one standard deviation unless otherwise indicated. Definition of irradiation creep strain is expressed by Eq. (1). An asterisk mark indicates uncertainty of dimensional inspection using optical microscope

Regarding the underlying mechanism of irradiation creep of SiC up to ~0.1dpa where transient creep is dominant [3], we demonstrated that the lattice strain was of the same order of magnitude as the macroscopic creep strain shown in **Table *1***. In addition, the XRD analysis found that macroscopic swelling and lattice expansion were comparable. Therefore, lattice expansion effectively contributes to the dimensional stability of CVD SiC. It is known that the magnitude of swelling of SiC is almost identical to the magnitude of the lattice expansion in the case of irradiation at intermediate temperatures (300–800°C) to a relatively low neutron dose (<~10 dpa) [6]. Therefore, this study provides additional confirmation of the relationship between swelling and lattice expansion. Moreover, this study provides the first data showing evidence of lattice strain contributing to neutron irradiation transient creep strain, based on XRD analysis. A previous study correlated radiation creep strain with swelling and provided a mechanistic creep model assuming anisotropic swelling due to applied stress [3]. A strong relationship between swelling and irradiation creep in a transient regime up to ~1 dpa is the current understanding regarding the creep mechanism [3,9]. However, experimental evidence had not been provided until this study, albeit only to 0.1 dpa. Therefore, the findings of this study represent an important addition to the knowledge of the mechanism of irradiation creep of SiC.

Although we found that additional lattice expansion due to stress is a source of creep strain based on out-of-pile experiments, it is still not clear what kind of radiation defect causes the lattice extension. A previous study found that decreasing coherent grain size and increasing anisotropic strain are indications of growth of dislocation loops in SiC [6]. This study found an insensitivity of the coherent grain size and the anisotropic strain to applied stress during irradiation, indicating that a stress of 100 MPa had a limited effect on the microstructural evolution.

To elucidate irradiation creep mechanism of SiC, on-going work includes detailed analysis of PDF spectra of the creep specimens. In addition, small angle x-ray diffraction experiment will be conducted at the NSLS-II beamline to evaluate the effects of applied stress during irradiation on microstructural evolution of SiC; analysis of scattering intensity profile will give the size, number densities, and shapes of the scattering features (e.g. irradiation-induced defect clusters). Analysis of the flexural creep specimens is underway.

4. SUMMARY

This report presents the status of the study on X-ray characterization of atomistic defects causing irradiation creep of SiC. XRD and PDF experiments of CVD SiC neutron-irradiated with and without applied stress were conducted at the NSLS-II using high-energy X-rays. Accurate lattice constant, coherent grain size, and anisotropic micro strain were obtained by Rietveld refinement. This study found that the macroscopic creep strain and strain due to the lattice expansion were similar following irradiation at 300°C to 0.1dpa, indicating that the lattice strain is a major source of the transient creep strain. This finding is critically important to elucidate creep mechanism and to develop a mechanistic creep model. In addition, the insensitivity of the coherent grain size and the anisotropic strain to applied stress during irradiation was found. This result indicates that applied stress during irradiation had a limited effect on the microstructural evolution. Further experiments using a small angle x-ray scattering technique and the analysis will be conducted in fiscal year 2019.

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