ELSEVIER

Journal of Nuclear Materials 307–311 (2002) 1168–1172

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Irradiation effects on thermal expansion of SiC/SiC composite materials

M. Ishihara [a,*], S. Baba [a], T. Hoshiya [a], T. Shikama [b]

[a] *Japan Atomic Energy Research Institute, Oarai, Ibaraki 311-1394, Japan*
[b] *Tohoku University, Sendai, Miyagi 980-8577, Japan*

Abstract

Irradiation-induced dimensional change and thermal expansion of two kinds of composites, self-particle reinforced SiC_p/SiC composites and a Hi-Nicalon™ SiC fiber reinforced SiC_f/SiC composite, and monolithic α-SiC were measured after irradiation at 0.2 dpa with irradiation temperatures of 573, 673 and 843 K using the JMTR. From the measurement, swelling was observed for the SiC_p/SiC composites and the monolithic α-SiC, on the contrary, the SiC_f/SiC composites showed a shrinkage. The measured thermal expansion increased with increasing the specimen temperature below the irradiation temperature, and then rapidly decreased over the irradiation temperature. The so-called 'temperature monitor effect' of the silicon carbide was clearly observed for all specimens, the monolithic α-SiC and both composites.

1. Introduction

Silicon carbide ceramics are thought to be promising materials for in-vessel structural components in a future fusion reactor concept, because the ceramics have superior thermomechanical stabilities at high temperatures as well as under high energy neutron irradiations. Focussing on the fiber-reinforced composites, SiC_f/SiC composites have been investigated extensively worldwide with the aim of improving as-fabricated qualities, irradiation characteristics and so on.

One of the developed heat-resistant SiC fibers is the so-called Hi-Nicalon™ SiC fiber, which contains excess carbon (C/Si ratio is 1.39 atomic) [1] manufactured by Nippon Carbon Co., Ltd. Recently, advanced near-stoichiometric SiC composition fiber, Hi-Nicalon™ Type S (C/Si $<$ 1.05 atomic) is developed, and material characterization research is now performing [2].

The Hi-Nicalon™ SiC fiber was successfully applied to the reinforcement of SiC-matrix composites [3]. Youngblood et al. irradiated the fiber at a damage level of 43 dpa at 1000 °C and reported the microstructural stability [4,5], furthermore, several researchers also studied tensile and fracture behaviour of Hi-Nicalon™ SiC fibers under unirradiated conditions [6,7]. Recently, mechanical properties of the SiC_f/SiC composite with the Hi-Nicalon™ fiber are studied without irradiation conditions [8,9], and moreover, there are several researches on irradiation effects of the composite [10]. Therefore, there are a few reports available for the understanding of irradiation effect on thermomechanical properties of the SiC_f/SiC composite, however they are not satisfactory up to now.

In order to characterize the thermal and mechanical properties, the authors have initiated a series of irradiation studies using monolithic SiC, self-particle reinforced SiC composites (SiC_p/SiC) and continuous SiC fiber, Hi-Nicalon™ fiber, reinforced SiC composites (SiC_f/SiC) [11]. These specimens were irradiated in the Japan Materials Testing Reactor (JMTR). In this paper the results of post-irradiation examinations on the irradiation-induced dimensional change and thermal

* Corresponding author. Tel.: +81-29 264 8734; fax: +81-29 2624 8712.
E-mail address: ishihara@popsvr.tokai.jaeri.go.jp (M. Ishihara).

0022-3115/02/$ - see front matter
PII: S0022-3115(02)01056-5

expansion are presented, and their property changes in view of lattice defects etc produced by the neutron irradiation are discussed.

2. Experimental

2.1. Materials

Two kinds of composites, continuous SiC fiber reinforced SiC_f/SiC and particle reinforced SiC_p/SiC, and one monolithic α-SiC were irradiated in this study. A 2D woven-fabric Hi-Nicalon™ fiber reinforced SiC-matrix composite was fabricated by using a polymer impregnation and pyrolysis process, manufactured by Nippon Carbon Co., Ltd. The surface of the Hi-Nicalon™ fiber was coated with 0.4 μm amorphous BN before preparation of the woven-fabric composite. The $SiC_p/SiCs$ were self-particle reinforced SiC composites with 5 and 10 wt% of 25 μm (mean particle diameter) SiC particles. Their matrices consist of the monolithic α-SiC, Hexaloy fabricated by Hitachi Chemical Engineering Co., with an average particle size of below 1 μm. These four kinds of materials were machined into pillar shaped specimens as shown in Table 1.

2.2. Testing

The irradiation test was performed at temperatures of 573, 673 and 843 K at a fluence level of $1.8 \times 10^{24}\ m^{-2}$ ($E > 1$ MeV), which corresponds to the damage level of 0.2 dpa, using the JMTR. For each temperature, two specimens were irradiated, and the temperature was measured by thermocouples. The accuracy of the temperature measurement was below 0.5 °C, and the temperature fluctuation during the irradiation test was controlled below 5 °C. After neutron irradiation, the irradiation-induced dimensional change and thermal expansion were measured. The dimensional change was measured at room temperature by a laser micrometer, manufactured by KEYENCE Co. Thermal expansion was measured by the thermomechanical analysis method from room temperature to 1673 K with a heating rate of 10 K/min in helium gas to avoid an oxidation. The temperature in the thermal expansion test was measured by thermocouples, and its accuracy was below 0.5 °C.

3. Results and discussion

3.1. Irradiation-induced dimensional change

The obtained data are plotted as a function of irradiation temperature in Fig. 1. The dimensional change of the SiC_f/SiC was negative and much lower than those of the SiC_p/SiC and the monolithic α-SiC. Several researchers investigated the irradiation-induced dimensional change of the SiC [12–16], and Price reported that the dimensional change in expansion was monotonically decreased with increasing irradiation temperature from about 1% at near room temperature irradiation to about 0.05% at 1273 K irradiation independent of α- or β-SiC

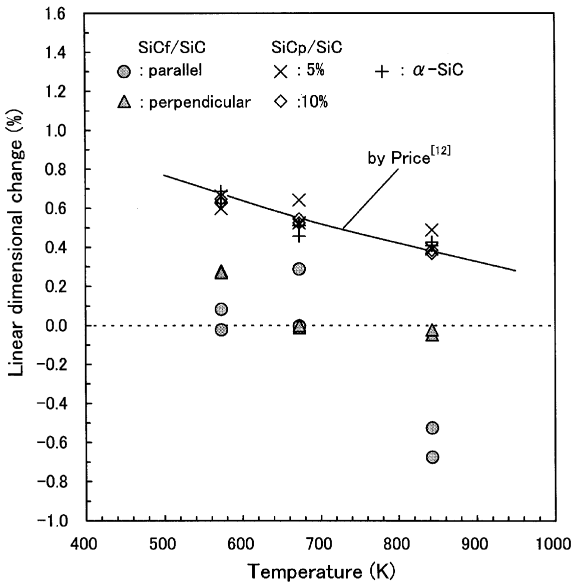

Fig. 1. Measured irradiation-induced dimensional change of SiC_f/SiC, SiC_p/SiC and monolithic α-SiC specimens at 0.5 dpa.

Table 1
Specimen geometry

Specimen		Geometry (mm)	Bulk density (g/cm^3)
Fiber reinforced SiC_f/SiC composite	Parallel to the fiber (‖)	∅4.5 × L20.0	2.12
	Perpendicular to the fiber (⊥)	∅4.7 × L20.0	
Particle reinforced SiC_p/SiC composite	5 wt%	∅4.5 × L19.0	3.06
	10 wt%	∅4.5 × L18.5	3.02
Monolithic α-SiC		∅4.5 × L18.0	3.11

∅: diameter, L: length.

as well as grain size, porosity of the material. Furthermore, previous analyses have shown that the irradiation-induced dimensional change of the SiC saturates and becomes independent of the fluence above 1–3 $\times 10^{24}$ m^{-2} [12]. Therefore, estimated line by Price [12] could be also plotted in Fig. 1. It can be seen from this figure that the obtained data fit well in the estimated line except for the SiC_f/SiC data.

It is reported that the neutron irradiation of a high-crystalline SiC results in swelling due to the generation of an equally number of interstitials and vacancies, and on the other hand, an amorphous SiC takes shrinkage due to the crystallization. Therefore, it is thought that the SiC_p/SiC and the monolithic α-SiC were high-crystalline SiCs, and that they showed almost the same swelling as the Price estimated line. On the other hand, the SiC_f/SiC would be amorphous SiC, because it shrank in the dimension.

Generally, dimensional changes of the fiber-reinforced composites are determined from those of the fiber and matrix as well as fiber–matrix interaction. Hollenberg et al. treated the fiber–matrix mechanical interaction, and derived simple equations considering the balance of forces within the composite [14]. The volume of the Hi-Nicalon™ fiber in the SiC_f/SiC was about twice that of the matrix in this study. In this case, dimensional change of fiber-reinforced composite is described by following equation.

$$\delta_{\text{composite}} = \frac{1}{3}\delta_{\text{matrix}} + \frac{2}{3}\delta_{\text{fiber}}, \tag{1}$$

where $\delta_{\text{composite}}$, δ_{matrix} and δ_{fiber} are the dimensional change in composite, matrix and fiber, respectively. For a fully decoupled fiber–matrix interface, the dimensional change of the composite is dominated by the matrix swelling, and in this case:

$$\delta_{\text{composite}} = \delta_{\text{matrix}}. \tag{2}$$

Several researchers reported that less crystalline SiC-based fibers shrank during irradiation due to the disordered structure of the fiber [4,16]. In Fig. 2, obtained SiC_f/SiC data, and Hollenberg et al. data [14], C-coated Nicalon fiber reinforced SiC_f/SiC manufactured by a chemical vapour infiltration (CVI) from 4.3 to 25 dpa, are plotted with Price reported curve [12]. The Hollenberg et al. data fit well in the Price estimated line [12], and it can be concluded that the fiber might decouple from the matrix under heavy irradiation due to the fiber shrinkage and matrix swelling, and hence, could not provide a compressive stress to limit the swelling of the matrix [14].

However, in our low dose irradiation case, the shrinkage of the Hi-Nicalon™ fiber was less than 10% compared with the Nicalon fiber in Fig. 2 [14]. Hence, it could be expected that the fiber was coupled well with the matrix, and expected that the fiber–matrix interaction would work.

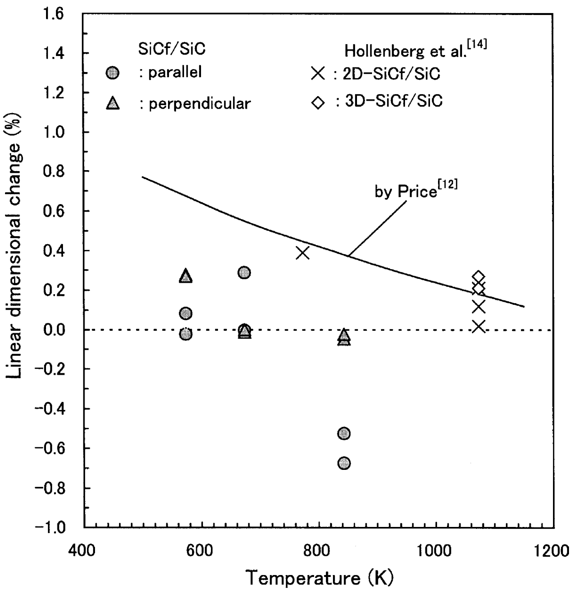

Fig. 2. Comparison between obtained data and C-coated Nicalon fiber–CVI–SiC_f/SiCs specimens.

Under the assumptions, that the fiber was coupled well with the matrix and that the matrix showed a swelling according to the Price estimated line [12], we can roughly estimate the dimensional change of the Hi-Nicalon™ fiber by Eq. (1). Fig. 3 shows the estimation

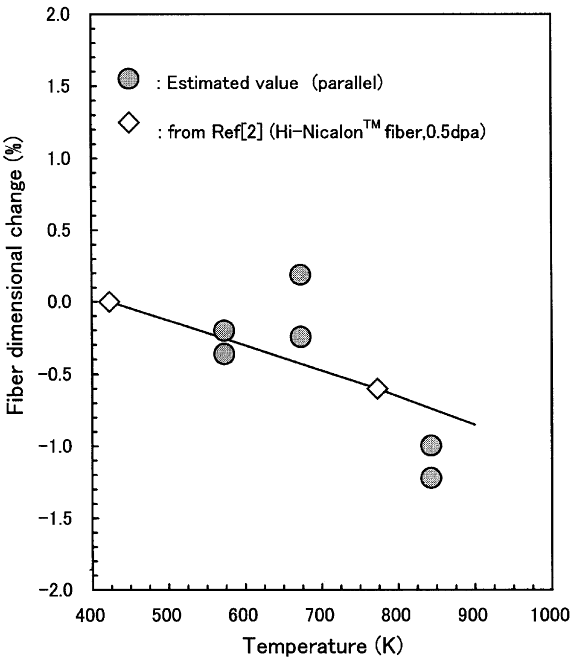

Fig. 3. Estimated irradiation-induced dimensional change of SiC-based fiber (parallel direction).

results. Obtained data by Osborne et al. dimensional change of Hi-Nicalon™ fiber at 0.5 dpa, were also plotted in this figure. It is found that the estimation results correspond well to the experimental data; namely, the SiC matrix would be almost crystalline one and showed swelling. However, since there is little knowledge on the irradiation-induced dimensional change of the fiber as well as the matrix SiC, it is necessary to obtain the experimental data to understand the irradiation-induced dimensional changes of the SiC_f/SiC composite.

3.2. Thermal expansion

In the post irradiation examination, thermal expansion of the SiC_f/SiC, SiC_p/SiC and monolithic α-SiC increased with increasing specimen temperature below the irradiation temperature and then rapidly decreased above the irradiation temperature. Fig. 4 shows the thermal expansion of the SiC_f/SiC specimens irradiated at 843 K. In the second measurement using the same specimens, the thermal expansion was fully recovered, i.e. a monotonic increase with increasing temperature was observed, and showed almost the same values obtained in the unirradiated specimens. The so-called 'temperature monitor effect' of the silicon carbide was clearly observed for all specimens, the monolithic α-SiC and the SiC_p/SiC and SiC_f/SiC composites.

It is said that thermally activated processes facilitate the migration of point defects and accelerate the rearrangement of the interstitials, vacancies as well as point defect clusters from the lattice during post irradiation annealing, and the annealing processes would not be significant below the irradiation temperature because during irradiation equilibrium states were reached between the irradiation-induced production of defects and the migration processes. For the temperature monitoring use, changes in macroscopic length, thermal conductivity, electrical resistivity or lattice parameters were investigated with isochronal heat-treatment in about one hour for each measuring steps [5,17,18].

It is thought that the mitigation and rearrangement of defects in thermally activated process during post irradiation annealing decreased thermal expansion measured under higher temperatures than the irradiation temperature. Therefore, the inflection point of the thermal expansion might indicate the irradiation temperature of the specimen. Fig. 5 shows the inflection point of thermal expansion coefficient obtained by the first measurement specimens irradiated at 843 K. From this figure, the irradiation temperature was estimated about 100 K higher than the irradiation temperature; the estimated temperature would be higher than the irradiation temperature, because the point defect clusters etc. might not be fully recovered by the post irradiation annealing at a heating rate of 10 K/min. However, it can be said that the thermal expansion measurement with continuous heating after irradiation has a potential to predict the irradiated temperature as a simple manner, not the isochronal heating. It is expected that if the thermal expansion is measured with the isochronal heat-treatment in about 1 h for each measuring steps, the

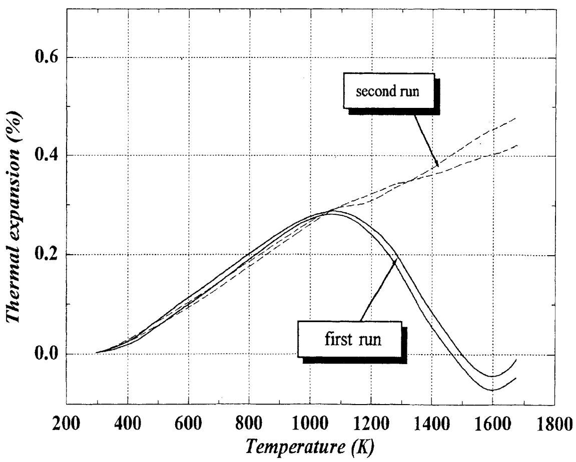

Fig. 4. Thermal expansion of SiC_f/SiC after neutron irradiation at 843 K up to 0.2 dpa.

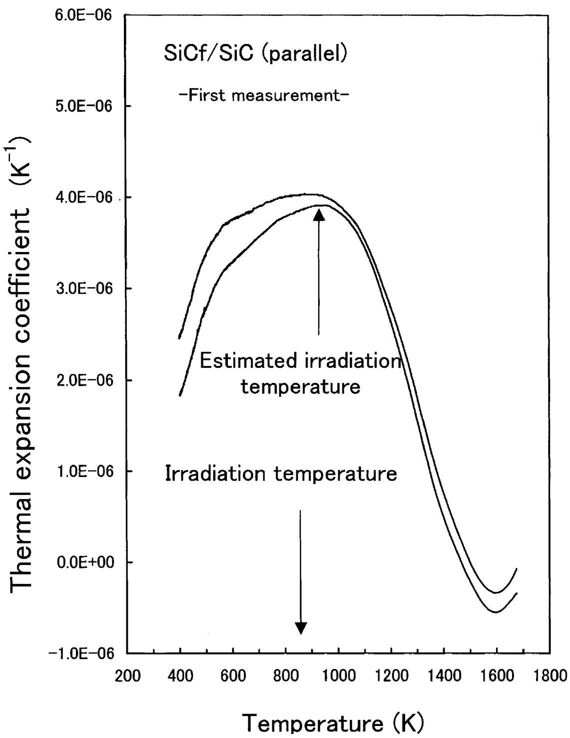

Fig. 5. Estimated irradiation temperature using thermal expansion coefficient after neutron irradiation at 843 K up to 0.2 dpa.

irradiation temperature could be determined more precisely from the thermal expansion coefficient.

4. Conclusions

Irradiation-induced dimensional change and thermal expansion of two kinds of composites, self-particle reinforced SiC_p/SiC composites and a Hi-Nicalon™ fiber reinforced SiC_f/SiC composite, and monolithic α-SiC were measured after irradiation at 0.2 dpa with irradiation temperatures of 573, 673 and 843K using the JMTR.

Obtained results are summarized as follows:

(1) The SiC_p/SiC composites and monolithic α-SiC showed swelling, on the contrary, the SiC_f/SiC composites showed shrinkage due to the shrinkage of the Hi-Nicalon™ fiber.
(2) The shrinkage of the SiC_f/SiC is understandable taking into account of the fiber–matrix interaction considering the balance of force within the composite.
(3) The measured thermal expansion increased with temperature below irradiation temperature and then rapidly decreased over irradiation temperature. The so-called 'temperature monitor effect' of the SiC was clearly observed for all specimens.
(4) The estimated irradiation temperature from a thermal expansion coefficient was about 100 K higher than the irradiation temperature. It is thought that the point defect clusters etc might not be fully recovered. A more precise measurement is expected with isochronal heating.

Acknowledgements

Mr T. Arai organized this work primary, and Dr K. Hayashi arranged irradiation and post irradiation experiments with some of the authors. The authors would like to thank for their contribution on the work.

References

[1] M. Takeda, Y. Imai, H. Ichikawa, T. Ishikawa, N. Kasai, T. Seguchi, K. Okamura, Ceram. Eng. Sci. Proc. 14 (7&8) (1993) 540.
[2] M.C. Osborne, C.R. Hubbard, L.L. Snead, D. Steiner, J. Nucl. Mater. 253 (1998) 67.
[3] M. Takeda, Y. Imai, H. Ichikawa, Y. Kagawa, H. Kakisawa, H. Iba, Ceram. Eng. Sci. Proc. 18 (3) (1998) 779.
[4] G.E. Youngblood, D.J. Senor, G.W. Hollenberg, DOE/ER-0313/18, 1995.
[5] G.E. Youngblood, R.H. Jones, A. Hasegawa, DOE/ER-0313/19, 1995.
[6] G.E. Youngblood, C. Lewinsohn, R.H. Jones, A. Kohyama, J. Nucl. Mater. 289 (2001) 1.
[7] C.H. Henager, G.E. Youngblood, D.J. Senor, G.A. Newsome, J.J. Woods, J. Nucl. Mater. 253 (1998) 60.
[8] T. Hinoki, W. Yang, T. Nozawa, T. Shibayama, Y. Katoh, A. Kohyama, J. Nucl. Mater. 289 (2001) 23.
[9] S.P. Lee, Y. Katoh, J.S. Park, S. Dong, A. Kohyama, S. Suyama, H.K. Yoon, J. Nucl. Mater. 289 (2001) 30.
[10] L.L. Snead, M.C. Osborne, R.A. Lowden, J. Strizak, R.J. Shinavski, K.L. More, W.S. Eatherly, J. Bailey, A.M. Williams, J. Nucl. Mater. 253 (1998) 20.
[11] M. Ishihara, S. Baba, T. Takahashi, T. Arai, K. Hayashi, Fus. Eng. Des. 51&52 (2000) 117.
[12] R.J. Price, J. Nucl. Mater. 33 (1969) 17.
[13] T. Suzuki, T. Yano, T. Mori, H. Miyazaki, T. Iseki, Fus. Technol. 27 (1995) 314.
[14] G.W. Hollenberg, C.H. Henager, G.E. Youngblood, D.J. Trimble, S.A. Simonson, G.A. Mewsome, E. Lewis, J. Nucl. Mater. 219 (1995) 70.
[15] H. Huang, N. Ghoniem, J. Nucl. Mater. 250 (1997) 192.
[16] L.L. Snead, M. Osborn, K.L. More, J. Mater. Res. 10 (3) (1995) 736.
[17] R.J. Price, Nucl. Technol. 16 (1972) 536.
[18] T. Yano, K. Sasaki, T. Maruyama, M. Ito, S. Onose, Nucl. Technol. 93 (1991) 412.