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Indentation fracture toughness of neutron irradiated silicon carbide

S. Nogami [a], A. Hasegawa [a,*], L.L. Snead [b]

[a] *Department of Quantum Science and Energy Engineering, Tohoku University, Aramaki-aza-Aoba 01, Aoba-ku, Sendai, Miyagi 980-8579, Japan*

[b] *Oak Ridge National Laboratory, Oak Ridge, TN 37831 6138, USA*

Abstract

The indentation fracture toughness and other mechanical properties (elastic modulus and hardness) of high-purity stoichiometric chemical vapor deposition processed-SiC under neutron irradiation in a dose range of 10^{24}–10^{25} n/m^2 ($E_n > 0.1$ MeV) at the temperature range of 80–1150 °C were investigated. Indentation fracture toughness decreased slightly at the temperatures below 400 °C, and increased above 400 °C, for example, from 5.08 MPa m$^{-1/2}$ (non-irradiated) to 6.52 MPa m$^{-1/2}$ (800 °C neutron irradiated). Mechanical properties investigated in this study were significantly dependent on the irradiation temperature up to 1150 °C, while generally independent on the irradiation dose up to 7.7 dpa signifying low-dose saturation.

1. Introduction

Silicon carbide (SiC) has been considered as a structural material for fusion applications because of its low neutron induced radioactivation and small degradation of mechanical properties at elevated temperatures [1–4]. One of the significant issues for SiC as a structural material is its low fracture toughness. For this reason, SiC fiber reinforced SiC matrix (SiC/SiC) composites have been developed [1–3]. The change of mechanical properties due to neutron irradiation is a central issue for fusion structural materials [1–3], and has received little study. For example, the behavior of fracture toughness under irradiation has not been well defined and may be of great importance. Considering this property is indicative of crack generation and propagation in SiC/SiC composites.

The purpose of this study is to clarify the fracture toughness and other mechanical properties (elastic modulus and hardness) of high-purity stoichiometric chemical vapor deposition (CVD) processed-SiC under neutron irradiation in a dose range of 10^{24}–10^{25} n/m^2 ($E_n > 0.1$ MeV) at various temperatures.

The fracture toughness was evaluated by the technique of indentation fracture toughness, which involves measurement of crack length introduced by an indentation using diamond tip defined by Evans [5]. This is one of the best techniques available for measuring the toughness of a small and brittle material like an irradiated ceramic because it can be evaluated by a crack growth behavior in a very small region.

2. Experimental and analysis procedure

2.1. Material and neutron irradiation tests

Material used in this study was polycrystalline β-SiC, that was fabricated by CVD process by Rohm and Haas. This material has a stoichiometric composition with high purity. Bulk SiC samples were machined into 25 mm × 1 mm × 1 mm bars. At least one side of the sample was mechanically polished.

* Corresponding author. Tel.: +81-22 217 7923; fax: +81-22 217 7925.

E-mail address: akira.hasegawa@qse.tohoku.ac.jp (A. Hasegawa).

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Neutron irradiation tests were performed in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory and the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory. Samples were irradiated in HFIR up to ~6 and 7.7×10^{25} n/m^2 ($E_n > 0.1$ MeV) at 300 and 800 °C and in HFBR up to ~1.1×10^{25} n/m^2 and ~0.15×10^{25} n/m^2 ($E_n > 0.1$ MeV) at 80–90, 250–270, 385, 890–980 and 960–1150 °C. In all cases thermocouples embedded in sample holders were used to monitor irradiation temperature. Irradiation conditions in this study are summarized in Table 1, where doses were calculated using an assumption that 1×10^{25} n/m^2 ($E_n > 0.1$ MeV) corresponds to 1 displacement per atom (dpa).

2.2. Microindentation test

Microindentation test was performed using a Vickers diamond (Model Micromet™ 3, Buehler, Ltd.), and doing so, Vickers hardness H_v, indentation diagonal $2a$, and crack length c were measured. Each indentation was made at a load of 2 kg with a dwell time of 15 s. Cracks from the indentation sides and subsurface cracks were not included in the analysis. Indentations were spaced more than 0.1 mm apart to prevent an interaction between indentations and free edge. The number of the indentation test was 4–9 in each sample in this study depending on goodness of data.

2.3. Nanoindentation test

Nanoindentation test was performed using a NANO INDENTER® II (Nano-instruments, Oak Ridge, TN), which is an ultra-low load, ultra-low depth (nanometer scale) sensing machine. This machine is equipped with a Berkovich diamond tip. In this study, one indentation included three separate loading/unloading segments. Indentation depths and loading (unloading) rates were constant in each division; 50 nm and 50 μm/s, 100 nm and 125 μm/s, and 200 nm and 300 μm/s. The indenter was unloaded until 90% of the load has been removed in each unloading segment and thermal drift corrected. The number of the indentation tests was 12–15 for each sample.

Table 1
Summary of the irradiation conditions in this study

Reactor (cycle, capsule, position, etc.)			Irradiation temperature/°C	Dose/dpa_SiC
HFIR	14J		300	6
			800	7.7
HFBR	SiC-1	Pos. 1	960–1150	1.1
		Pos. 2 (−2)	960–1150	1.1
		Pos. 2	960–1150	1.1
		Pos. 3	890–980	1.1
		Pos. 4	250–270	1.1
		Pos. 5	385	1.1
		Pos. 6	80–90	1.1
	V4	Pos. 1	105	0.15
		Pos. 2	160	0.15
		Pos. 3	256	0.15

Hardness H_{nano} and elastic modulus E_{nano} were evaluated using an unloading curve by a method proposed by Oliver and Pharr [6]. H_{nano} is calculated from the following formula:

$$H_{nano} = P_{max}/A(h_c), \tag{1}$$

where P_{max} is the maximum indentation load and A, which is a function of constant indentation depth h_c, is a contact area of the indenter with a sample surface. E_{nano} is defined as follow:

$$E_{nano} = (1 - v_s^2)/(1/E_r + (1 - v_i^2)/E_i), \tag{2}$$

where v_s, v_i, and E_i are a Poisson's ratio of the sample, that of the indenter (0.07 for the diamond tip), and an elastic modulus of the indenter (1141 GPa for the diamond tip), respectively. Reduced modulus E_r in Eq. (2) has the form:

$$E_r = \pi^{1/2} S/2\beta A(h_c)^{1/2}, \tag{3}$$

where S and β is a slope of the initial unloading curve and a geometrical correlation factor (1.034), respectively. The theoretical geometrical shape function for Berkovich tip $A(h_c)$ represents the same area-to-depth ratio as the Vickers diamond tip. In practice, however, the shape of the indenter is not an ideal shape, therefore, $A(h_c)$ was calibrated by procedure given by Oliver and Pharr [6].

2.4. Indentation fracture toughness

Evans–Davis model [5] was used for the evaluation of indentation fracture toughness K_c in this study. This model describes K_c as follows:

$$K_c = 0.4636(P_v/a^{3/2})(E_{nano}/H_v)^{2/5} 10^F, \tag{4}$$

$$F = -1.59 - 0.34B - 2.02B^2 + 11.23B^3 - 24.97B^4 + 16.23B^5, \tag{5}$$

$$B = \log_{10}(c/a), \tag{6}$$

where P_v is a peak load in Vickers hardness test. Modulus from the third division of the indentation curve in the nanoindentation test was used as E_{nano} in calculating K_c because the scattering of E_{nano} among 12–15 indentations was the smallest. Behavior of H_v change was different from that of H_{nano} as described later, possibly

due to the difference of the shape of indenter-tip and the calculation method between two tests. The relation between an ideal shaped tip are described as $H_v = 0.927\ H_{nano}$ in the open literature [7].

3. Results and discussion

The dependencies on the irradiation temperature of Vickers hardness H_v and indentation hardness H_{nano} of CVD-SiC irradiated up to various doses are shown in Figs. 1 and 2, respectively. The error bars denote plus or minus of one standard deviation calculated from 4 to 9 data points for H_v and 12 to 15 data points for H_{nano} at each irradiation conditions. From the results, it is clear that the hardness increased at all neutron doses and temperatures studied. The value H_{nano} of the non-irradiated sample (~37.8 GPa) was almost the same as the open literature values (~36 GPa) [8]. Furthermore, results suggest that the hardness of CVD-SiC is independent on the irradiation dose above a saturation value, while significantly dependent on the irradiation temperature. Such low-dose saturation in hardness was shown previously on identical materials to those used in this study irradiated in HFIR [8]. The hardness is shown to rapidly increase with temperature below 100 °C and with almost no change observed above 100 °C. It is noted that, for temperatures below ~180 °C, amorphization of SiC is possible leading to a decreased hardness of ~55% [9]. However, the material of this study did not undergo amorphization. The ratio of the irradiated to the non-irradiated hardness was about 119% for H_v and 110% for H_{nano}.

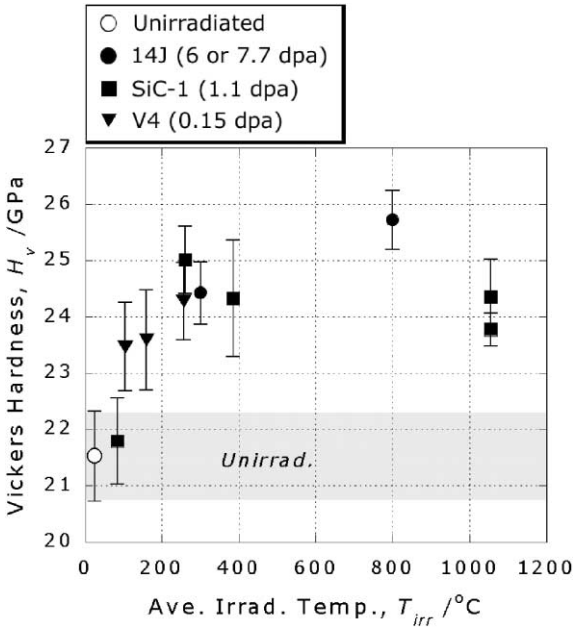

Fig. 1. Dependence of the Vickers hardness H_v on the irradiation temperature and dose.

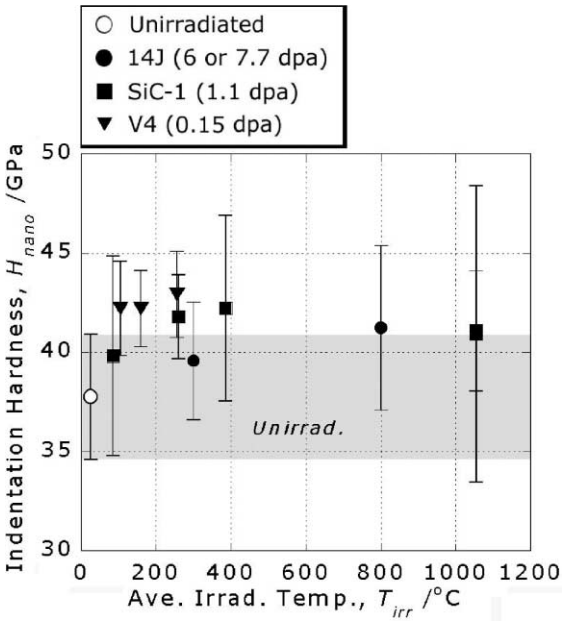

Fig. 2. Dependence of the indentation hardness H_{nano} on the irradiation temperature and dose, which are calculated using the method proposed by Oliver and Pharr [6].

The elastic modulus also exhibits a significant dependence on the irradiation temperature and independence on the irradiation dose shown in Fig. 3. The error bars denote the same meaning as in H_{nano}. The result show the rapid reduction of E_{nano} with the irradiation temperature below 100 °C from ~490 GPa for the non-irradiated one to ~400 GPa for the irradiated one. Elastic modulus E_{nano} of the irradiated samples, however, gradually recovered and showed almost the same value as the non-irradiated value when irradiated above 1000 °C.

The observed trends in the elastic modulus can be related with the swelling summarized in the open literature [10]. The physical expansion for this decrease in as-irradiated elastic modulus is based on the accumulation of point defects (vacancies) in the SiC crystal. It is well known [11] that neutron-irradiation-induced defects are more stable during low-temperature irradiation and tend to self-anneal with increased irradiation temperature until the vacancies' density responsible for lattice strain is so low at 1000 °C that essentially no swelling occur. The elastic modulus is considered to be physically dependent on the interatomic bond strength, atomic spacing and bond density, therefore, the modulus considered to decrease as irradiation-induced strain is reduced at high irradiation temperatures.

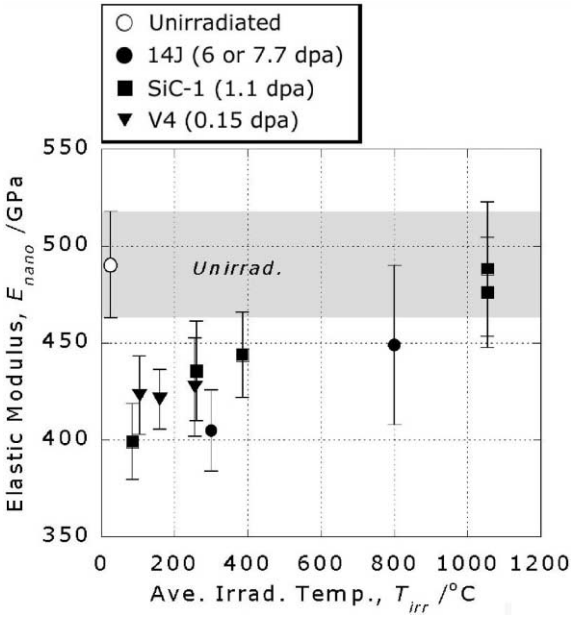

Fig. 3. Dependence of the elastic modulus E_{nano} on the irradiation temperature and dose, which are calculated using the method proposed by Oliver and Pharr [6].

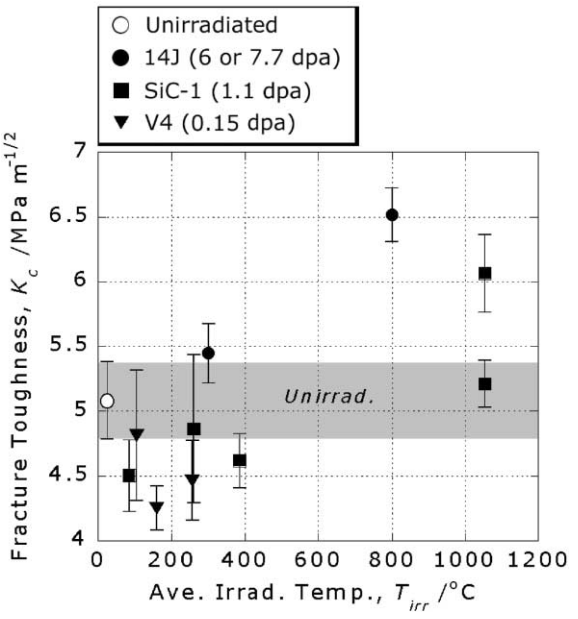

Fig. 4. Dependence of the indentation fracture toughness K_c on the irradiation temperature and dose, which are calculated by Evans–Davis model [5].

Fig. 4 shows the dependence of the indentation fracture toughness K_c of CVD-SiC irradiated in various doses on the irradiation temperature. K_c was calculated using Evans–Davis model [5] as described before, in which the average data of E_{nano} in each irradiation conditions was used as the elastic modulus, and thus, the number of the data points of K_c in each irradiation conditions corresponds to that of Vickers hardness test (4–9 points). The error bars, therefore, only include the scatter of Vickers hardness H_v, indentation diagonal $2a$, and crack length c. The value K_c of the non-irradiated CVD-SiC in this study (5.08 MPa m$^{-1/2}$) was about twice larger than that in the open literature [8]. It is noted that, while this technique is quantitative, the measurement of crack length is somewhat subjective, hence data should be regarded as qualitative in nature. A small reduction of K_c below ~400 °C, and an increase in it with increased irradiation temperature to ~1000 °C was observed. The value K_c for CVD-SiC irradiated to 7.7 dpa at 800 °C in HFIR increased by about 30% compared with the non-irradiated value.

Indentation fracture toughness of SiC is considered to be strongly affected by the plastic deformation, e.g., dislocation motion, under compression introduced by indentation tests and crack growth behavior. The previous work suggested that carbon interstitials in SiC are considered to be mobile below about 800 °C according to the molecular dynamics simulations [12], and they are considered to form a dislocation-loop. The previous work by Clinard et al. suggested that the increase of the fracture toughness of Al_2O_3 and MgO is attributed to crack pinning and deflection due to the defect clusters [13]. The increase of K_c for CVD-SiC with temperature in this study might be due to the increase of the mobility of a dislocation and crack pinning and deflection.

Mechanical properties of CVD-SiC are significantly changed by neutron irradiation as described here, however, in order to clarify the mechanism of the mechanical properties change, the detail of the microstructural development due to neutron irradiation must be investigated, for example by transmission electron microscopy, in future work. Additionally, data of the mechanical properties at temperature range of 400–800 °C and above 1100 °C are also required to complete the data systems of the neutron irradiated SiC.

4. Summary

The indentation fracture toughness of CVD-SiC irradiated up to 10^{24}–10^{25} n/m^2 ($E_n > 0.1$ MeV) at the temperature of 80–1150 °C in HFIR and HFBR was investigated and was compared with other mechanical properties (hardness and elastic modulus). The following results were obtained:

(1) The hardness (Vickers hardness H_v and indentation hardness H_{nano}) showed a similar irradiation-induced behavior. They rapidly increased with the irradiation

temperature below 100 °C and showed almost no change above 100 °C.

(2) The result showed the rapid reduction of the elastic modulus E_{nano} with the irradiation temperature below 100 °C. Elastic modulus of the irradiated samples, however, gradually recovered and showed almost the same value as the non-irradiated value when irradiated above 1000 °C.

(3) A small reduction in fracture toughness K_c below about 400 °C was seen, with a significant increase as temperature is increased up to about 1000 °C.

(4) Mechanical properties investigated in this study were significantly dependent on the irradiation temperature, while generally independent on the irradiation dose.

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