"EFFECTS OF NEUTRON IRRADIATION ON THE FRACTURE OF SILICON CARBIDE"

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ABSTRACT

The fracture of two phase siliconized SiC containing 8-10 wt% excess silicon was determined after neutron irradiation to a fluence of $1.2X10^{25}n/m^2$ (E>1MeV) with sample temperature $\lesssim$473°K (200°C) during irradiation. High temperature (1473°K) fracture of samples with one as fired surface in tension exhibit no decrease in fracture strength within experimental error. The Weibull modulus giving an indication of strength scatter is not significantly affected by irradiation. The mode of fracture for both irradiated and unirradiated material is primarily transgranular as determined by scanning electron microscopy. Radiation-induced swelling and thermal conductivity changes saturate after a fluence of $\approx 4X10^{24}n/m^2$ and most of the annealing of electrical resistivity occurs in the 500-1100°K temperature range. Results on SiC irradiated at a temperature of 1373°K (1100°C) are also included in this paper.

KEYWORDS

Neutron irradiated silicon carbide; fracture strength; swelling; electrical resistivity; thermal conductivity.

INTRODUCTION

Ceramic materials such as silicon carbide (SiC) are being considered in various applications as an alternative to metals for possible use in the first-wall, limiters and blanket of nuclear fusion reactors. These structures will be subjected to a severe environment of high temperature and intense radiation fields including ions, 14-MeV neutrons, X-rays and electrons. One promising candidate is reaction-bonded SiC, which possesses excellent high temperature properties, low oxidation at high temperature, low induced nuclear activation, and low gamma heating. (Rovner and Hopkins 1976.) Reaction-bonded material (NC-430, Norton Co., Worcester, Mass.) is characterized by large grains(50-100μm) of α-phase SiC an hexagonal close packed structure (HCP) embedded in a matrix of β-phase a face centered cubic structure (FCC), and free silicon, the latter comprising some 8-10% of the total weight.

Among material parameters, the high temperature fracture strength behavior as a function of neutron fluence and irradiation temperature are extremely important. Some fracture work has been done with neutron-irradiated reaction-bonded material (Matthews 1974, Matheny et.al. 1978). The radiation response data of Matthews 1974 on

Refel SiC exhibit somewhat different results than those of Matheny et. al. 1978, and the results of the present study which could be due to differences in the manufacturing processes of the starting material. Matheny et.al. used NC-430 SiC in their experiments. Along with fracture strength; irradiation-induced volume, density, electrical resistivity, microstructural property and thermal conductivity changes were investigated and results on the radiation response of these properties form the basis for this paper.

EXPERIMENTAL DETAILS

Samples of reaction-bonded SiC were irradiated in position V-15 of the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory, details about the samples including irradiation temperature, displacements per atom, fluence, fracture strength and surface conditions are given in Table I. All sample dimensions were measured before and after irradiation, using a micrometer with a reproducibility of 0.005 mm. The nominal sample dimensions were 2.54X2.54X31.8 mm^3.

TABLE 1 Details of Sample Surfaces, Irradiation Conditions, Fluences and Fracture Strength

No of Samples	Fluence 10^{24}n/cm^{2}E>1MeV	Displacement per atom dpa	Irradiation Temperature °C	Fracture Strength MPa	Surfaces
14	0	0	-	257 ± 20	all machined
15	4	0.87	∿1100°C	201 ± 14	" "
12*	0	0	-	231 ± 17	one as fired
14	4	0.87	<200°C	208 ± 21	" " "
14	12	2.6	$\lesssim$200°C	229 ± 16	" " "

*G. Trantina, General Electric Company, private communication.

Fracture strengths were determined using a three-point bend rig with SiC arms having a span of 2.29 cm, the loading rate used was 7.48 kg/min. All samples were equilibrated for 10 minutes at 1200 ± 5°C before loading. The fracture rig was placed in an electric furnace with an air environment. Samples having an as-fired surface, had this surface placed in tension during loading.

A liquid displacement technique was used to determine densities. By weighing the sample before and during immersion in tribromomethane (ρ = 2.869 g/cm^3), using a digital scale with 0.0005 gm precision, the density could be determined using the expression

$$\rho_{23°C} = (W_a/W_a-W_t) \text{ X } 2.869 \text{ g/cm}^3 \qquad 1$$

where W_a = weight in air, and W_t = weight in tribromomethane.

The isochronal annealing of electrical resistivity was obtained using a standard four-probe technique (Valdes 1954). The post irradiation heat treatments were carried out with samples subjected to annealing times of ten minutes at all temperatures.

The measurements of thermal conductivity, K, were performed at room temperature with the sample surrounded by an insulating material. Given the steady-state heat flow equation

$$Q = -KA\frac{dT}{dx} \qquad 2$$

where Q is the heat input, A the contact area, and $\frac{dT}{dx}$ is the spatial temperature gradient, it is possible to measure relative values of K if Q, A, and dx are kept the same in all cases. In this experiment, uncertainties in the heat reaching the sample, as well as the contact area do not allow absolute measurements, but did permit measurement of the thermal conductivity after irradiation K, relative to the thermal conductivity before irradiation K_o.

The microstructure of the fracture surfaces was examined using a 20 KeV scanning electron microscope (SEM).

RESULTS AND DISCUSSION

a) Fracture - Studies of the fracture strength of irradiated reaction-bonded SiC have been reported by Matthews 1973 and Matheny et.al. 1978. Shown in Fig. 1 are the results of the fracture tests as well as data by Matheny et.al. 1978. Note that the samples irradiated at a temperature of $\lesssim$200°C show little, if any, strength loss. The data by Matheny for samples also irradiated at this temperature indicate a slight strength loss with increasing fluence, a trend also observed for the samples irradiated at $\approx$1100°C. It is possible that the fracture behavior of a sample having an as-fired surface under tensile stress is determined to a great extent by the surface irregularities and gross flaws already present as a consequence of manufacture, with irradiation-induced flaws being less significant.

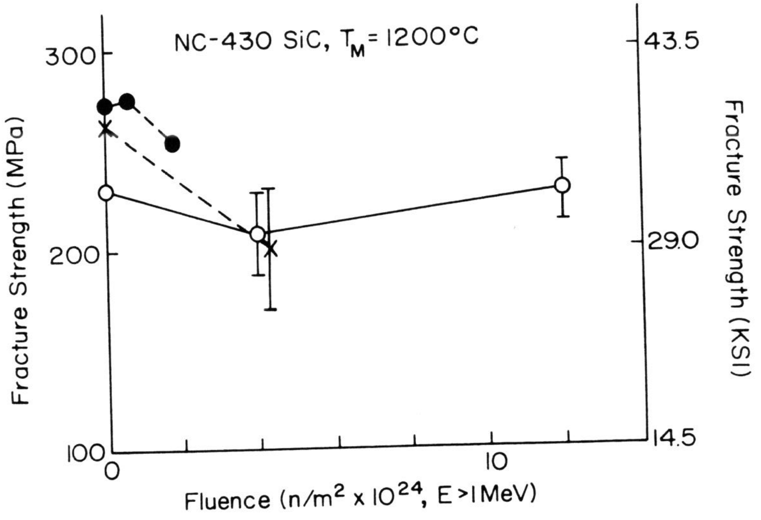

Fig.1) O, ● T_{irr} $\lesssim$200°C (O one as fired surface ● all machined surfaces); X T_{irr} $\approx$1100°C (all machined surfaces); ● Matheny et al data

The scatter in strength can be analyzed using Weibull statistics (Weibull 1951). The strength of a brittle ceramic is governed by a random distribution of flaws, with the Weibull relation giving the probability of failure as

$$P = 1-e^{-R} \qquad 3$$

where R is the risk of rupture given by

$$R = \int (\sigma/\sigma_o)^m \, dV \qquad 4$$

Here σ is the applied stress on the material, σ_o is a normalizing constant stress, and V the volume of the test specimen. The exponent m, known as the Weibull modulus, is a measure of scatter in the strength, and consequently can be used to predict the size effect when the test material is used in large structures. The Weibull modulus can be calculated for a given series of fracture tests by fitting the best straight line through a plot of ℓn ℓn (1/(1-P) vs m ℓn σ + A where A is a constant and P is the median rank given as n-0.3/N+0.4 (Johnson 1951), with N being the number of samples tested and n the order of failure. The values of the Weibull modulus for the various fracture tests are given in Fig. 2 along with results reported by Matheny et al (1978).

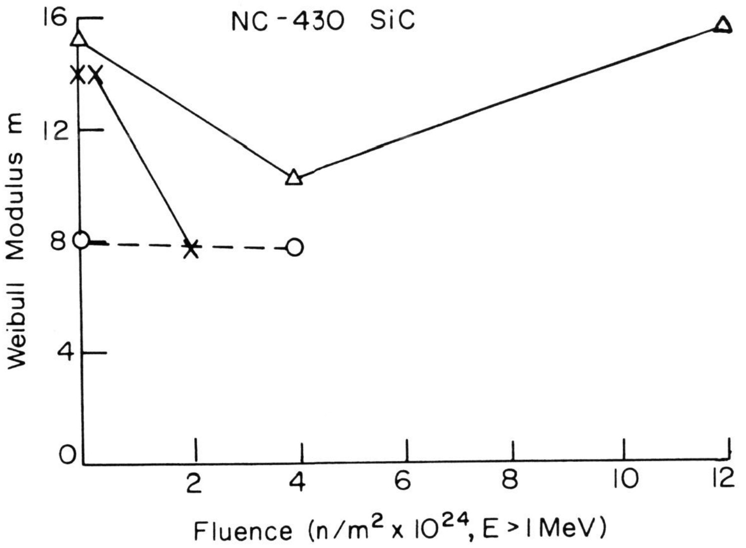

Fig. 2) X, Δ T_{irr} ≲200°C (Δ one as-fired surface X all machined surfaces); O T_{irr} ≈1100°C (all machined surfaces); X Matheny et al data

Note the apparent lack of decrease in strength scatter in the as-fired samples irradiated to a fluence of $12X10^{24} n/m^2$ as opposed to an increase in scatter with increasing fluence in the results reported by Matheny et al who used all-machined surfaces. This trend seems to lend credence to the possibility that irradiation does not significantly affect the strength-determining flaw distribution of as-fired samples. Additionally, variation among unirradiated samples cut from different batches of NC-430 material (with all machined surfaces) is seen to be rather large. For the samples having all machined surfaces irradiated at ≈1100°C it seems that although there was a loss of strength, the scatter was not greatly affected.

b) Swelling ($\frac{\Delta V}{V_o}$), Density ($\frac{\Delta\delta}{\delta_o}$), and Thermal conductivity (K/K_o).

The irradiation-induced volume, density, and thermal conductivity changes (K, K_o are the post and pre-irradiation values of thermal conductivity) are shown in Figs. 3,4, and 5 respectively. The volume and density results corroborate each other, with the swelling of the material saturating after a fluence of ∿$4X10^{24} n/m^2$. The

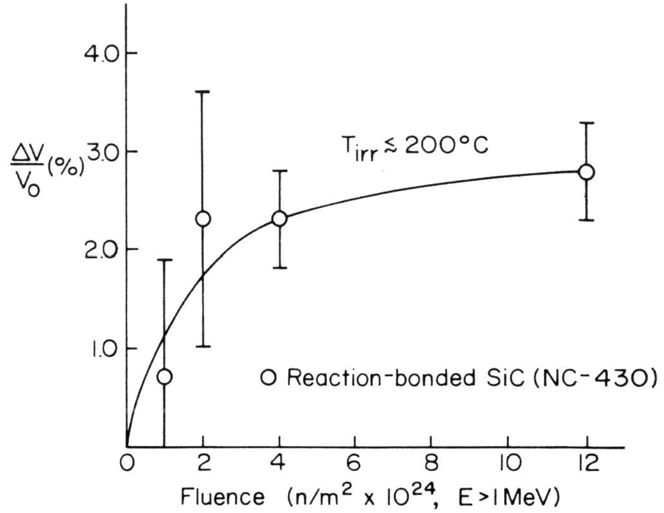

Fig.3) Swelling $\frac{\Delta V}{V_o}$(%) vs fluence for samples irradiated at ≲200°C before heat treatment

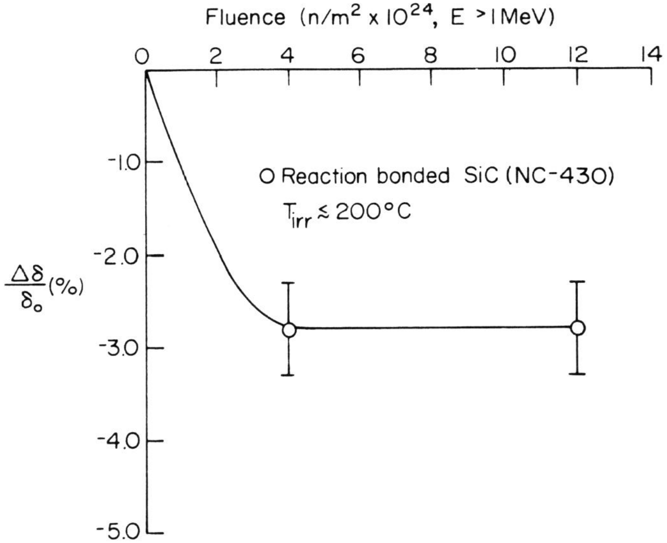

Fig.4) Density decrease $\frac{\Delta\delta}{\delta_o}$(%) vs fluence for samples irradiated at ≲200°C before heat treatment

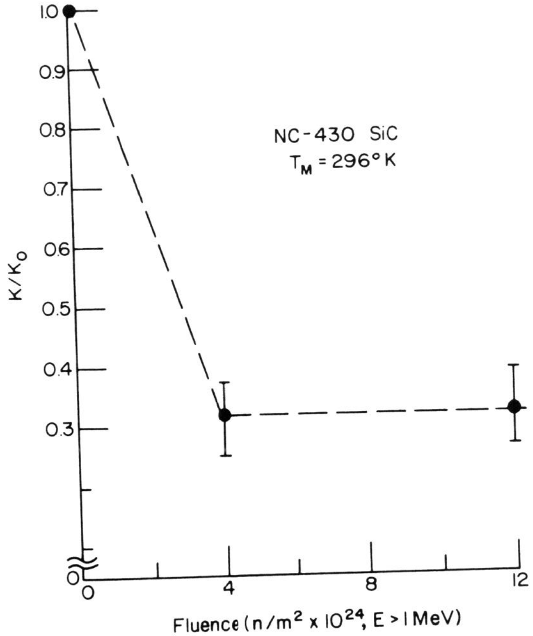

Fig. 5) Relative thermal conductivity vs fluence

saturation fluence and magnitude of the swelling are in agreement with results obtained by others (Price 1977). The decrease in thermal conductivity saturates at a fluence level similar to that at which saturation of swelling occurs. In the thermal conductivity measurements the samples had all been exposed to 1200°C for 10 min, hence it is likely that some of the lattice damage had annealed out and the original K/K_o may be even smaller. The above results clearly indicate that the lattice damage resulting in changes in the above properties saturates for the fluences considered in this study.

Swelling, density, and thermal conductivity measurements were not carried out with the samples irradiated at $\sim$1100°C.

c) Resistivity - The isochronal annealing behavior of electrical resistivity for samples irradiated at $\lesssim$200°C is shown in Fig. 6 along with data by Matheny et al (1978). The values ρ_o and ρ are the pre-and post-irradiation resistivity values measured at 296°K after 10 minutes at each indicated temperature. The recovery (lowering) of the resistivity is seen to begin at $\sim$500°K for the samples irradiated at $\lesssim$493°K. This annealing behavior of neutron-irradiated SiC has been utilized as an irradiation temperature monitor (Price 1973). The increase in resistivity due to irradiation saturates at a level $\sim10^4$-10^5 times the unirradiated value for all the low irradiation temperature cases. Significant damage is apparently present in the lattice if the irradiation temperature is low. The samples irradiated at $\sim$1100°C show only a factor 2 increase in resistivity and no annealing stage, indicative of significant recovery during irradiation.

It appears to be advantageous to operate ceramics at high temperatures if the lattice damage determined as an increase in electrical resistivity is at all an indicator of effects on mechanical strength. To investigate this it would be necessary to perform fracture strength vs annealing temperature tests, thus involving a large number of samples and costly reactor irradiations.

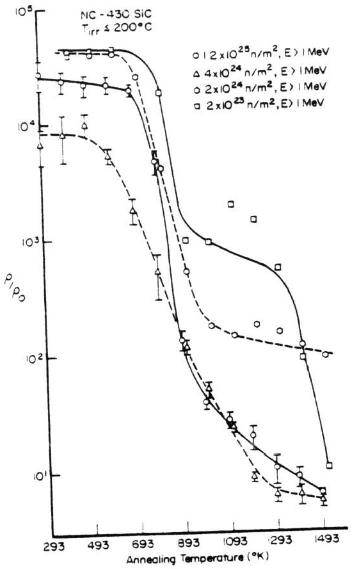

Fig. 6) Isochronal annealing of electrical resistivity

Inspection of SEM photos of fracture surfaces shows little change in the predominantly transgranular mode of fracture in all cases, although some increase in the size of the α grains was apparent in the samples irradiated at $\sim$1100°C.

Although one normally observes transformation of β-phase to α-phase SiC at high temperature (2000°C), we have observed the β→α transformation at 1100°C using transmission electron microscopy analysis of neutron-irradiated SiC. The neutron energy transfer events apparently enhance the β→α transformation, causing it to occur at 1100°C, rather than at the normal temperature for unirradiated SiC. The SEM photos also revealed the presences of small voids 1-5μm in size for both irradiated and unirradiated material which can be attributed to manufacturing processes. Microcrack lines were not observed on the fracture surfaces of either the irradiated or unirradiated samples in the SEM photos.

SUMMARY AND CONCLUSIONS

1) High temperature 1473°K (1200°C) fracture strength of reaction bonded SiC with one as-fired surface in tension exhibits no decrease in fracture strength within experimental error after irradiation to a fluence of 1.2X10^{25}n/m^2 with sample temperature during irradiation kept at $\lesssim$200°C. The mode of failure is transgranular for both irradiated and unirradiated material.

2) The Weibull modulus is not significantly effected by irradiation.

3) Radiation-induced swelling and thermal conductivity changes saturate after a fluence of $\cong$4X10^{20}n/m^2.

4) Annealing of the electrical resistivity occurs mainly in the temperature range 500-1100°K irrespective of the fluence used in the present experiments. Samples irradiated at $\gtrsim$1373°K exhibit a much smaller increase in electrical resistivity

(factor ∿2) and no discernable annealing stage indicating that significant annealing is occuring during irradiation.

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