DIMENSIONAL AND MICROSTRUCTURAL STABILITY OF 3C-SiC IN HIGH TEMPERATURE IRRADIATION CONDITIONS—S. Kondo, Y. Katoh, L. L. Snead (Oak Ridge National Laboratory)

OBJECTIVE

The cavity swelling and progressive dislocation evolution are critical because they may cause unstable volume expansion and irradiation creep deformation, respectively. The objective of this work is to reveal the temperature and fluence dependence of the cavity swelling and dislocation development in 3C-SiC. Of importance is that both the irradiation fluence and temperature were better characterized in this experiment than in most earlier work.

SUMMARY

Microstructural analysis of 3C-SiC following neutron-irradiation at high temperatures provided quantitatively reliable data of temperature and fluence dependence of cavity swelling and dislocation evolution. Post-irradiation annealing effects on microstructural defects were also studied.

Cavities were observed for all irradiation conditions (1050-1460˚C, up to 9.6 dpa) except for 1050˚C, 1.4 dpa. However, the magnitude of cavity swelling was very small below ~1300˚C: the largest cavity swelling observed below that temperature was only ~0.01% at 1300˚C, 9.3dpa. The temperature dependent increase in the cavity swelling was observed clearly above ~1400˚C. Fluence dependent swelling became plainly visible above ~1400˚C, and the maximum value observed in this work was 0.25% at 1460˚C, 9.6 dpa.

The dominating dislocation structure in the high temperature regime is Frank faulted loop. Minor black spots and small loop populations, which dominated the defect microstructure in lower temperature regime, were still coexisted with the Frank loops up to ~1150˚C. Defect number density decreased and the size increased with increasing temperature. Significant decrease in the number density and increase in the size was observed at 1300-1460˚C.

PROGRESS AND STATUS

Introduction

SiC and its composites attract a lot of interest for the nuclear application due to the low activation properties and remarkable mechanical properties at very high temperature. In recent years, many efforts have been made to evaluate irradiation effects such as dimensional instability, modification of thermal and electrical transport properties, and mechanical property changes [1]. This work focused on cavity swelling and dislocation development in the very high temperature regime, which will be primary information for clarifying the underlying mechanisms of most irradiation effects.

There are limited studies of microstructural changes in 3C-SiC at low and intermediate irradiation temperatures (<~800˚C) [2-8]. Black spot defects and small loops, which are aggregates of displaced Si and C atoms, are the dominating microstructures in the temperature regime. The negative temperature dependence of saturated swelling values in 3C-SiC has been reported up to 1000˚C [9], which was primarily attributed to the lattice dilation near the point defects and/or their clusters. In the high temperature regime (<~1000˚C), recent self-ion- and neutron-irradiation experiments revealed that the dominant defects were interstitial-type Frank loops and cavities [10]. Cavity formation resulting from super saturated vacancy migration is a common phenomenon in most irradiated materials at elevated temperature (roughly 0.3-0.5 T_m). Recent densitometry experiments on irradiated 3C-SiC showed that the volume expansion increased with increasing temperature and fluence above ~1100˚C [11,12].

Table 1 Irradiation conditions and microstructural parameters for neutron-irradiated 3C-SiC.

Irradiation temperature		Fluence	Cavity size	Cavity density	Cavity volume fraction	Loop radius	Loop density
˚C		dpa	nm	m^{-3}	%	nm	m^{-3}
1050	3C-SiC	1.4	Not detected	-	-	1.2	1.1×10^{23}
1050	3C-SiC	4.9	1.1	1.0×10^{20}	3.1×10^{-6}	1.5	1.4×10^{23}
1130	3C-SiC	1.5	1.1	1.8×10^{20}	4.3×10^{-6}	2.2	9.1×10^{22}
1130	3C-SiC	5.1	1.1	3.0×10^{20}	5.0×10^{-5}	3.3	9.1×10^{22}
1130	3C-SiC	7.7	1.1	1.7×10^{21}	6.2×10^{-5}	4.1	9.2×10^{22}
1220	3C-SiC	1.7	1.1	2.5×10^{21}	2.0×10^{-4}	2.2	1.2×10^{23}
1300	3C-SiC	1.9	1.8	5.5×10^{22}	5.9×10^{-3}	3.8	6.5×10^{22}
1300	3C-SiC	5.3	1.2	2.7×10^{23}	9.6×10^{-3}	6.3	5.4×10^{22}
1300	3C-SiC	9.3	1.7	2.0×10^{23}	1.4×10^{-2}	8.5	2.7×10^{22}
1400	3C-SiC	5.6	1.7	4.7×10^{23}	3.3×10^{-2}	18	4.8×10^{21}
1400	3C-SiC	9.4	2.1	4.4×10^{23}	5.6×10^{-2}	27	2.9×10^{21}
1460	3C-SiC	1.9	3.0	3.6×10^{22}	1.3×10^{-2}	16	4.0×10^{21}
1460	3C-SiC	5.8	4.5	6.9×10^{22}	1.1×10^{-1}	28	1.2×10^{21}
1460	3C-SiC	9.6	5.7	6.6×10^{22}	2.4×10^{-1}	43	3.7×10^{20}

Experimental Procedure

The material used for this work was polycrystalline 3C-SiC, which was produced by chemical vapor deposition (CVD) by Rohm and Haas Advanced Materials (Woburn, Massachusetts). The CVD material is extremely pure, with typical total impurity concentration of less than 5 wppm. The grain size is between 5 and 10 µm in the plane parallel to the deposition substrate, with the grains elongated in the <111> growth direction perpendicular to the substrate. The material is typically free of microcracks or other large flaws, but atomic layer stacking faults on the {111} planes are common. There is no porosity in CVD SiC, and the material is generally considered to be of theoretical density (approximately 3.21 g/cm^3).

Three fixed-core capsules, each containing 10 sub-cupsles, were irradiated in the High Flux Isotope Reactor, at Oak Ridge National Laboratory. The highest fluence for specimens studied here was 9.6×10^{25} n/m^2 (>0.1 MeV), corresponding to 9.6 dpa-SiC. Irradiation temperatures ranged from 1050 to 1460˚C, determined by the post-irradiation inspection of melt wires inserted in both ends of each sub-capsule. Irradiation conditions and results of defect microstructures for each specimen are given in Table. 1.

Results and Discussion

Cavity microstructure in irradiated 3C-SiC are shown in Fig. 1. Very sparse (<1×10^{21} m^{-3}) cavities near 1 nm in diameter were observed only in grain boundaries at 1050˚C, 5.0 dpa. No cavities was detected at lower fluence of 1.4 dpa at 1050˚C. The image shown does not represent general microstructures in the specimens but show the selected area which contains cavities. A high density (2.7×10^{23} m^{-3} at 9.3 dpa, 1300˚C, and 4.4×10^{23} m^{-3} at 9.4 dpa, 1400˚C) of cavity formation becomes general above ~1300˚C. Most of these cavities appeared to be spherical, although the cavity size is comparable to the TEM resolution limit in this work. Faceted voids are observed at 1460˚C as shown in Fig. 1, which were confirmed to be near-tetrahedron, bounded by {111}

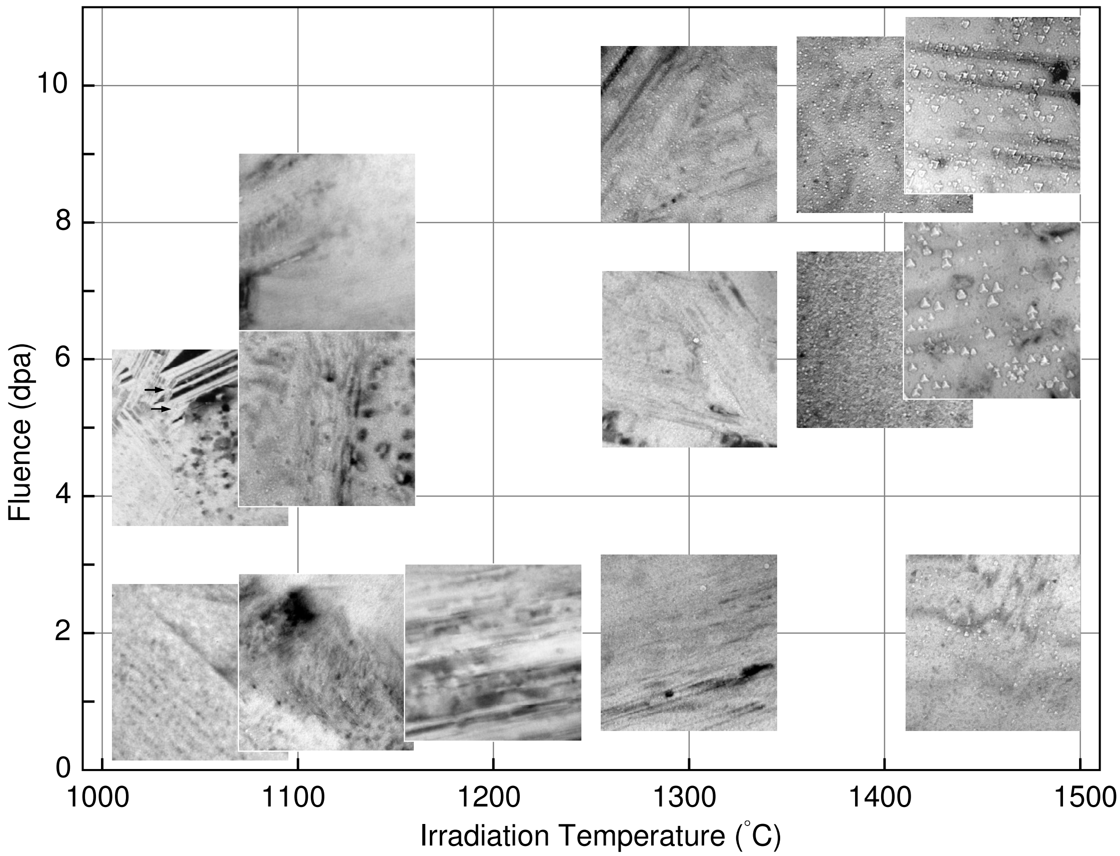

Fig. 1 Cavity microstructures in β-SiC irradiated with neutron.

surfaces, with the corners generally truncated [10]. More detailed analysis of faceted void surfaces will be published elsewhere [13].

The estimated volume fractions of cavities (V_{cavity}/V_0) are plotted with previous neutron data [10] in Fig. 2 as a function of irradiation temperature. This shows that the small spherical cavities make only insignificant contribution to the swelling below ~1300˚C (~0.01% at 1300˚C, 9.6dpa). The cavity volume fractions at ~6 and ~10 dpa increase steeply with increasing temperature. Figure 3 shows the irradiation fluence dependence of cavity swelling for three temperatures. The fluence dependence becomes notable above ~1400˚C. The swelling rates estimated from these plots assuming linear fluence dependence are approximately 1×10^{-3}, 6×10^{-3} and 0.03 %/dpa for the temperatures ~1300, 1400, and 1460˚C, respectively.

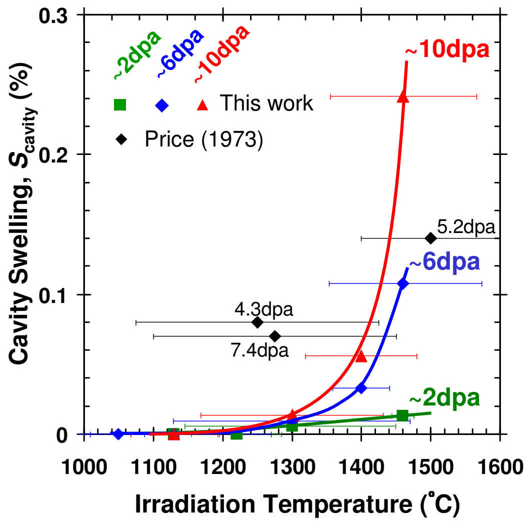

Fig.2 Irradiation temperature dependence of cavity swelling in neutron-irradiated 3C-SiC.

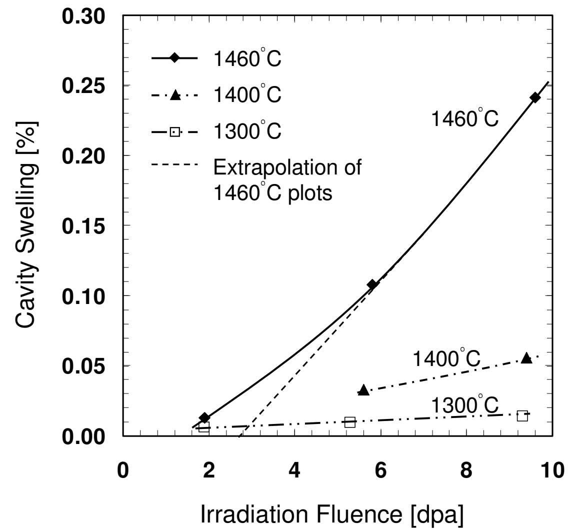

Fig.3 Irradiation fluence dependence of cavity swelling in neutron-irradiated 3C-SiC at three temperatures.

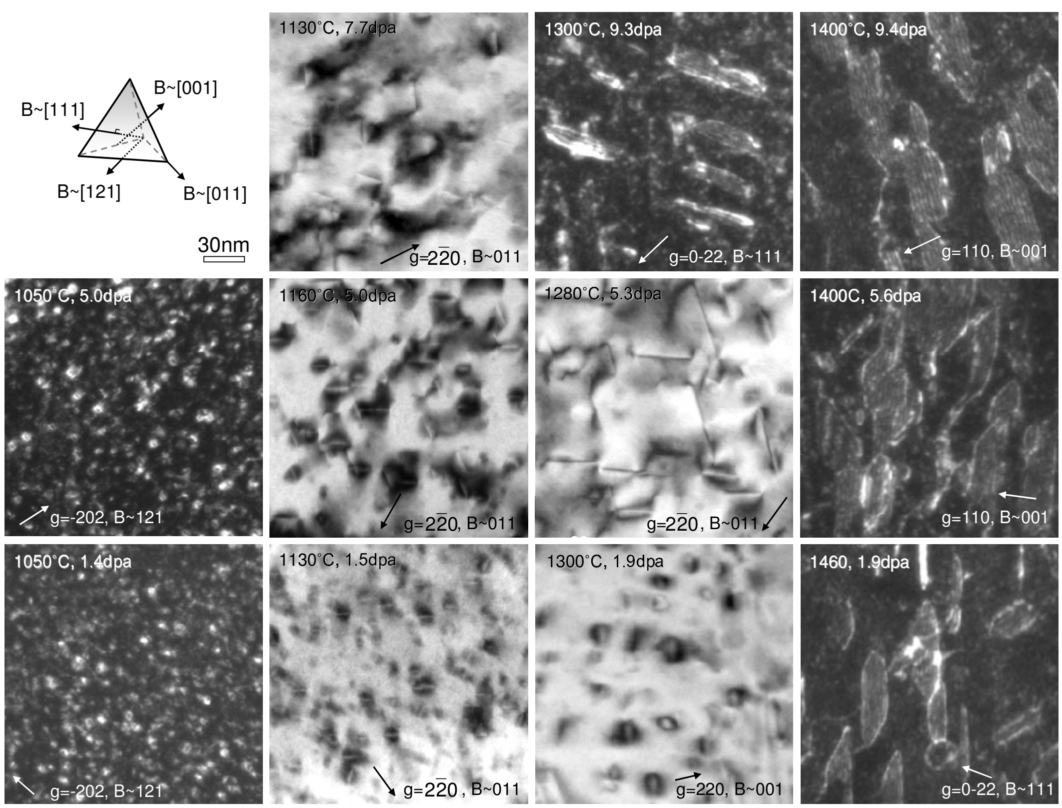

Fig. 4 Dislocation microstructures in 3C-SiC irradiated. Beam direction and g vectors are indicated in each image.

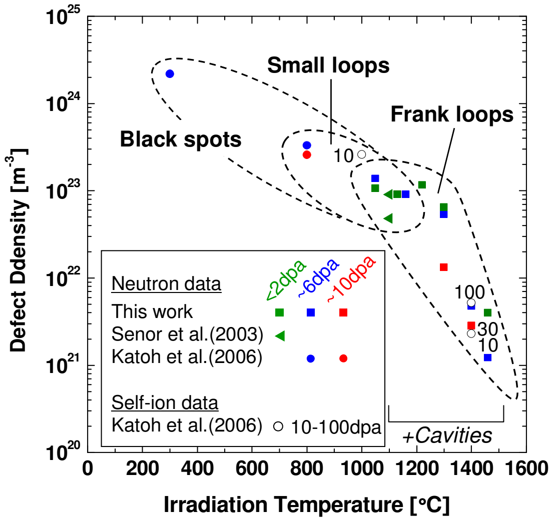

Fig. 5 Irradiation temperature dependence of defect number density in neutron-irradiated 3C-SiC. Displacement damage levels are indicated for the reported self-ion data.

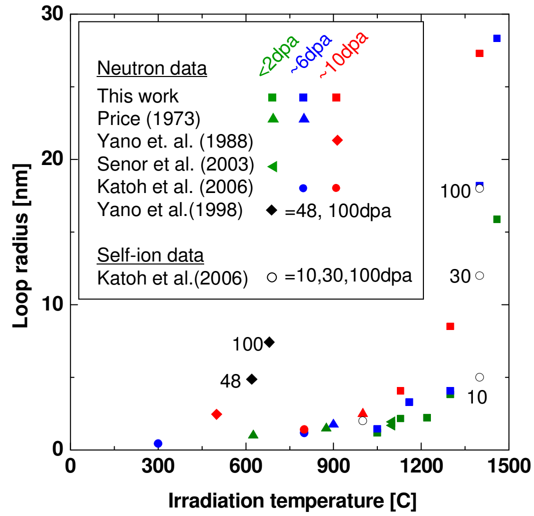

Fig. 6 Irradiation temperature dependence of defect size in neutron-irradiated 3C-SiC.

Swelling-temperature curves generally show a peak (or two peaks), because the void size is positively and void density is negatively depended on the irradiation temperature. There is no such a peak in Fig. 2 below 1460˚C. In many irradiated materials, the cavity swelling commences gradually with irradiation fluence accumulation, and the swelling rate finally reaches a steady-state level at a given temperature. The extrapolation of the fluence dependent swelling in Fig. 3 indicates an incubation fluence above ~2 dpa at 1460˚C, which is likely related to continuous void nucleation and/or an immature dislocation microstructure. The negligible cavity swelling rates in SiC observed below 1300˚C may be attributed to the limited vacancy mobility and the relatively stable dislocation microstructures, with high sink density as discussed below. The discrepancy in the value of cavity swelling from the earlier reports of Price might be primarily caused by the uncertainty of the irradiation conditions and/or impurity in the older material used by Price.

Fig. 4 shows examples of the dislocation microstructure in 3C-SiC irradiated at 1050, 1300, and 1460˚C. Various defects including small defect clusters (so-called black spots) with unidentified configuration, small loops with various Burgers vectors, and Frank loops are formed at 1050˚C as shown in Fig. 4(a). Many loops were identified to have formed on {111} planes by imaging from the {111} satellite streak near 111 spots. Most loops formed at 1300˚C were identified as Frank loops by the imaging from {111} satellite streak and/or g·b contrast analysis. Above 1400˚C, larger Frank loops with b = <111> were observed. Peripheries of the larger loops are predominantly hexagonal in shape, with sides parallel to <011> directions as shown in Fig. 4(c).

The defect number density and loop radius were plotted along with previous neutron and self-ion data as a function of irradiation temperature in Fig. 5, and Fig. 6, respectively. Defect density decreased and the size gradually increased with increasing temperature below ~1300˚C. Both rapid density decrease and size increase were observed above ~1300˚C. The black spots, which are dominant defects below ~00˚C, still contribute to the defect density with small loops and Frank loops in the temperature range ~800-1150˚C. The very high sink density related to the small defect clusters likely retard the significant loop growth at these lower temperatures. At 1300-1460˚C, the irradiation fluence significantly affects both the defect density and size, where loops grow rapidly and develop into larger Frank loops (>~20 nm in radius). According to conventional thought,

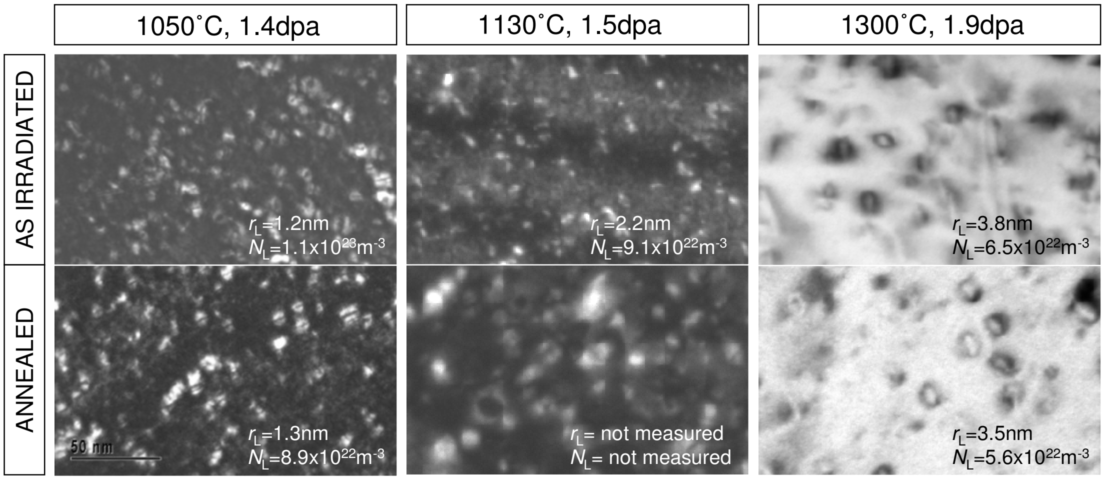

Fig. 7 Defect microstructures before and after a post irradiation anneal at 1500˚C for 1 hour.

reduced sink strength of the thermally unstable defects including small loops and cavities should allow the rapid loop growth in the very high temperature regime. The formation of dislocation networks resulting from continuous unfaulting of the Frank loops, which were not observed in the present work, might start at higher fluence as demonstrated in the self-ion irradiated SiC [8].

Figure 7 shows the defect microstructures before and after post irradiation annealing at 1500˚C for 1 hour. Measured loop radius and number density are shown in each image. Both the radius and density were not significantly changed after annealing, except that the number density of smaller loops decreased slightly. Generally, smaller loops can be removed easily because of the more excess free energy (dislocation line tension) per interstitial. For a similar reason, smaller voids may be removed by annealing. Senor et al. reported the absence of cavities in 3C-SiC irradiated to 0.9×10^{25} n/m^2 at 1100℃, whereas cavities were observed following subsequent annealing at 1500˚C for 1hour [7]. In contrast, annihilation of voids by annealing for specimen irradiated at 1130˚C was observed. Further studies are required to clarify the annealing mechanisms of irradiated SiC.

CONCLUSIONS

The temperature and fluence dependence of cavity swelling and dislocation development in CVD SiC irradiated with fast neutrons at high temperatures (1050-1460˚C, up to 9.6 dpa) were evaluated using transmission electron microscopy. The cavity swelling was very limited below 1300˚C (<0.01% at 1300˚C, 9.3 dpa). Temperature and fluence dependent swelling became visible above ~1400˚C. The maximum value of the cavity swelling was 0.25% at 1460˚C, 9.6 dpa, but this appeared to be below the peak swelling temperature. Frank loops were the dominant dislocation structure in this temperature regime, and the number density decreased and the size increased with increasing irradiation temperature. The loop microstructures depended less significantly on both the irradiation temperature and fluence below 1200˚C. A significant decrease in the number density and increase in the size were observed at 1300-1460˚C.

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