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Size distribution of black spot defects and their contribution to swelling in irradiated SiC

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HIGHLIGHTS

- We investigated the role of black spot defects (BSD) in swelling of ion-irradiated 4H-SiC.
- Swelling by XRD is corrected for the effect of the rigid substrate.
- BSDs contributes 62% to the overall swelling and the remainder is caused by isolated point defects.
- Swelling from TEM is lower by 40% to 70% than the XRD values.

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ABSTRACT

Experimental and modeling efforts were combined to investigate the role of black spot defects (BSD) in swelling of carbon- and krypton-irradiated 4H-SiC. Samples were exposed to conditions favoring BSD formation: irradiation at temperatures 600–950 °C and damage levels of 0.4–0.8 dpa. The maximum XRD swelling values, corrected for the effect of the rigid substrate, of 0.58% for C and 0.75% for Kr-irradiation were measured at the lowest irradiation temperature of 600 °C and decreased with increasing temperature. The swelling values estimated from TEM are on the same order of magnitude, but usually 40–70% lower than those measured by XRD. The contribution of BSDs to the overall swelling is 62% and the remainder of the swelling is caused by isolated point defects. The obtained results contribute to understanding of what defect types account for swelling and how their concentration evolves with the irradiation temperature and damage level.

1. Introduction

Excellent mechanical and thermal properties as well as outstanding radiation resistance have secured silicon carbide (SiC) numerous applications especially in demanding areas like nuclear systems (TRISO, fusion blanket), space telescopes, and airplane engines [1]. In the case of light water reactors (LWRs), SiC is envisioned as an accident-tolerant cladding capable of withstanding high radiation doses and pressures. Such harsh conditions will expose the material to high strains and stresses leading to swelling and creep. The swelling of SiC at various temperatures and damage levels has been studied extensively [2] and is fairly well understood quantitatively. However, in order to develop predictive tools for swelling modeling under various reactor conditions, the underlying mechanisms have to be studied especially at LWRs operation temperatures, below 1000 °C, where swelling is dominated by point defect formation and clustering. Previous results on neutron and silicon-irradiated SiC at medium-to-high damage levels (4.4–100 dpa) and temperatures of 600–1400 °C [3,4] have shown that qualitative study of swelling via conventional transmission

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electron microscopy (TEM) is insufficient, as the observed defects account only for about 10% of estimated total value [4]. It was therefore suggested that so-called black spot defects (BSDs – small interstitial clusters), observed by some researchers as patches of dark TEM contrast, contribute to the remainder of the swelling [2].

In this work, we improve on the qualitative description of swelling by accounting for black spot defects and the rigid substrate effect. High-resolution TEM was employed to study BSD size and density in ion-irradiated 4H-SiC. Based on the TEM data we calculate contributions to swelling from the observed black spot defects and from point defects that are expected to be present at these irradiation conditions based on previous modeling studies. We compare these results to swelling estimated by X-ray diffraction (XRD) experiments. The swelling calculation from XRD is corrected by accounting for the effect of the un-irradiated substrate on the lateral confinement of the damage zone.

2. Experimental approach and modeling details

2.1. Material

{0001} single crystal, hexagonal 4H-SiC (a = 3.073 Å, c = 10.053 Å), n-doped, 4.1° off towards $[11\bar{2}0]\pm0.5°$, with low micropipe and double-side polish from Cree was chosen to study black spot defects. Single-crystal was selected for easy strain and swelling measurements by XRD and to reduce the defect density in the virgin material for TEM investigations.

2.2. Ion-irradiation

To simulate neutron irradiation, 4H-SiC was irradiated at 600 °C, 800 °C, and 950 °C by 3.15 MeV C^{2+} up to 0.4 dpa and 1 MeV Kr^{+} at 600 °C and 800 °C up to 0.4 and 0.8 dpa. Irradiations were performed at conditions at which BSDs are expected to form without introducing much strain [2].

Carbon irradiation was performed using the 1.7 MV tandem accelerator located at UW-Madison under the following conditions: 3.15 MeV C^{2+} (projected range $R_p = 2.23 \pm 0.11$ μm) up to a fluence of 5.14×10^{16} C/cm^2 which corresponds to 0.4 dpa at the depth of 1 μm (see Fig. 1), assuming threshold displacement energies of 20 eV and 35 eV for C and Si [5], respectively. For the damage level calculation, the method proposed by Stoller et al. [6] was employed.

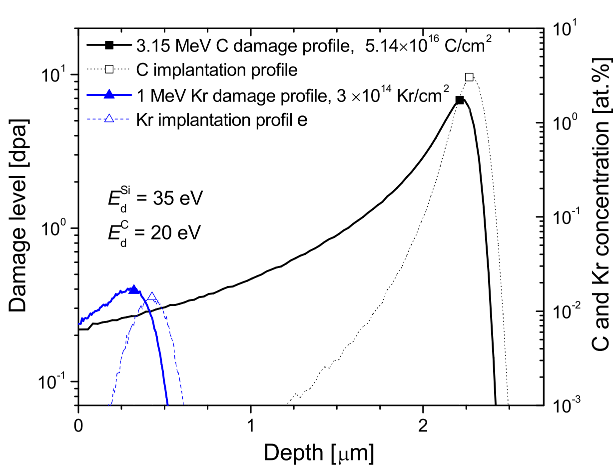

Fig. 1. Damage and C and Kr ion distributions in SiC irradiated with 3.15 MeV C and 1 MeV Kr ions to a damage level of 0.4 dpa at the depth of 1 μm and peak, respectively. Calculations were performed using SRIM-2013.00 [26], assuming the displacement threshold energies to be 20 eV for C and 35 eV for Si.

The maximum C concentration is about 3 at.% with a majority of the C interstitials located at the end of the irradiation range. The average current was around 4 μA, and the flux was kept at a level of 6.5×10^{12} C/(cm^{2}s), resulting in a damage rate of 5×10^{-5} dpa/s. The beam was rastered (64 Hz horizontally, 517 Hz vertically) over the entire irradiation area, and its uniformity was controlled by an infrared camera. Sample temperature, controlled by two thermocouples attached diagonally to the sample holder, was attained by external and beam heating with fluctuations of ±20 K. The background pressure during irradiation was kept around 10^{-6} Torr. 1 MeV Kr-irradiations (R_p=0.4 ± 0.09 μm) were performed at the University of Illinois, Urbana-Champaign, Frederick Seitz Material Research Laboratory using an HVEE van der Graaf accelerator. The irradiations were conducted up to a fluence of either 3×10^{14} Kr/cm^2 or 6×10^{14} Kr/cm^2, which corresponds to 0.4 and 0.8 dpa at the damage peak, respectively (see Fig. 1). The maximum Kr concentration was about 0.03 at.% (for 6×10^{14} Kr/cm^2), which does not alter the stoichiometry of the implanted SiC samples. The implantation spot was 6 × 6 mm^2, and the flux varied between 1.4×10^{12} Kr/(cm^{2}s) and 3×10^{12} Kr/(cm^{2}s) (current 80–170 nA) yielding a damage rate of 1.7×10^{-3} dpa/s to 4×10^{-3} dpa/s. The background pressure was around 5×10^{-7} Torr, and the sample temperature was measured by a K-type thermocouple attached to the sample holder. The temperature uncertainty was within ±5 K. The same method of damage level calculation was employed as for C-irradiation. During the irradiation, all samples were attached to the sample stage using silver paint, but only one sample was irradiated at a time while the other three were kept heated during this period. Once the desired fluence was reached, the beam was shifted to the next sample. In all cases, the beam was perpendicular to the sample surface, and ion channeling was avoided as a consequence of the 4.1° off-cut angle of the virgin material.

2.3. XRD measurements

Ion-induced strain and point defect swelling were measured by means of XRD. The measurements of single crystal 4H-SiC were conducted at room temperature with a PANalytical X'pert PRO diffractometer (45 kV, 40 mA) in Bragg (reflection) geometry utilizing $Cu_{K_{\alpha1}}$ radiation (λ = 0.154056 nm) in combination with a hybrid monochromator consisting of a closely coupled X-ray mirror and a 4-bounce Ge 220 monochromator (18 arcsec resolution). By adjusting azimuthal and polar angles, the sample was precisely oriented in order to align a given crystallographic direction exactly with the diffraction vector. The alignment was performed on the crystal planes instead of the surface, which reduced the axial divergence. In the case of the off-cut samples used in this research, the XRD curves were measured at an angle α = 4.1° from the reciprocal lattice vector corresponding to the (0004) pole with the $[11\bar{2}0]$ direction (off-cut angle direction) in the detection plane. The $2\theta - \omega$ scans were taken at the (0004) pole with 0.001° steps, 0.5 s per step. To determine whether there is any in-plane compressive stress, reciprocal space maps (RSMs) were recorded in the vicinity of the (004) and (106) reflection.

The value of the total normal strain was calculated from the equation

$$\varepsilon_N^{tot} = \frac{\Delta c}{c} = -\Delta\theta \cot\theta_B, \quad (1)$$

where $\Delta\theta$ is the difference between the diffraction angle $\theta = (2\theta)/2$ and the Bragg angle θ_B, and c = 10.053 Å.

To determine the fraction of the damage zone being probed by X-rays, we calculate the sample thickness x required to reduce the amount of transmitted X-ray intensity by half (the so-called half-