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Amorphization resistance of nano-engineered SiC under heavy ion irradiation

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ABSTRACT

Silicon carbide (SiC) with a high-density of planar defects (hereafter, 'nano-engineered SiC') and epitaxially-grown single-crystalline 3C-SiC were simultaneously irradiated with Au ions at room temperature, in order to compare their relative resistance to radiation-induced amorphization. It was found that the local threshold dose for amorphization is comparable for both samples under 2 MeV Au ion irradiation; whereas, nano-engineered SiC exhibits slightly greater radiation tolerance than single crystalline SiC under 10 MeV Au irradiation. Under 10 MeV Au ion irradiation, the dose for amorphization increased by about a factor of two in both nano-engineered and single crystal SiC due to the local increase in electronic energy loss that enhanced dynamic recovery.

1. Introduction

Silicon carbide (SiC) and its composite materials are proposed for utilization as accident-tolerant nuclear fuel cladding [1] and structural components in fusion reactors [2,3]. In these applications, SiC will be exposed to severe radiation environments, and the mechanical properties of SiC and composites materials will be significantly degraded due to radiation damage accumulation. The enhancement of radiation tolerance is of technological importance to avoid serious accidents, and much effort has been devoted to develop radiation tolerant materials [4–8]. In the case of SiC-SiC composites, the current generation of 3C-SiC fibers [9,10], as well as the matrix [11], can contain high densities of stacking faults, which can affect the irradiation response of composites. We have recently synthesized nano-engineered silicon carbide (NE-SiC), which is basically 3C-SiC with columnar grains (20–100 nm in diameter) and high densities (nm-spacing) of stacking faults and twins, similar to that found in SiC-SiC composites, and investigated its amorphization resistance under Si ion irradiation [12,13]. The results of this study revealed that NE-SiC is more radiation resistant than single-crystalline SiC at room temperature. On the other hand, it has been reported that NE-SiC exhibits a lower radiation resistance than conventional SiC with micron-sized grains under *in situ* TEM irradiation with 1 MeV Kr ions [14]. However, there is a very subtle difference in the threshold dose for amorphization between NE-SiC and conventional SiC, and it seems to be difficult to distinguish them. For direct comparison, it is necessary to compare specimens irradiated under the same irradiation conditions, such as species, temperature, fluence, and flux. In the present study, we perform Au ion irradiations of NE-SiC and single-crystalline SiC at the same time and compare their amorphization resistance.

2. Experimental procedures

NE-SiC thin films, approximately 530 nm thick with nm-sized columnar grain through the thickness of the films, were deposited on a Si substrate by low-pressure chemical vapour deposition, and the details of the sample preparation techniques were described elsewhere [12]. For a reference material, single-crystalline 3C-SiC thin films (hereafter referred to as SC-SiC),

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approximately 1 μm thick and epitaxially-grown on a Si substrate, were obtained from GT Crystal Systems Inc. (Salem, MA). Both samples were irradiated with Au ions at room temperature using the capabilities of the Ion Beam Materials Laboratory at the University of Tennessee [15]. To examine the effects of irradiation energy on amorphization, 2 MeV and 10 MeV Au ion irradiations at ion fluxes of 9.4×10^{11} and 3.2×10^{11} Au/cm^{2}s, respectively, were carried out in the present study. To maintain the identical irradiation conditions, both NE-SiC and single crystalline 3C-SiC were mounted side-by-side and irradiated under raster scanning beam simultaneously. Cross-sectional specimens for transmission electron microscopy (TEM) observations were prepared by a combination of mechanical polishing and ion thinning. The specimens were characterized by a JEOL JEM-3000F (operated at 200 kV) at the Kyushu Institute of Technology.

3. Results

The irradiation-induced structural changes from 2 MeV Au ions in epitaxially-grown single-crystalline 3C-SiC (SC-SiC) are shown in Fig. 1(a–c) as a function of ion fluence, while those in NE-SiC are shown in Fig. 1(d–f). The dark-field TEM images were taken by using the 002 reflection of the 3C-SiC. The cross-sectional images of the specimens irradiated to a fluence of 5×10^{13} Au/cm^2 reveal that a highly damaged layer is formed in SC-SiC at the depth of 220–450 nm from the surface (Fig. 1(a)), while no remarkable damage layer is observed in NE-SiC (Fig. 1(d)). We have recently proposed that the strain field near stacking faults gives an impact on point defects migration and damage recovery processes [13]. The nanostructures associated with planar defects are maintained in Fig. 1(d), which is attributed to the difference between SC-SiC and NE-SiC in damage accumulation. It should be noted that the nano-columns of NE-SiC are rotated around the [111] axis as reported previously [16]. Therefore, only the columns satisfying the Bragg condition become bright, and the dark contrast of Fig. 1(d) is not due to amorphization. Indeed, no halo rings are observed in the electron diffraction pattern obtained from the damaged region of the NE-SiC.

Irradiation to a higher fluence induces amorphization in both the specimens. The cross-sectional views of Fig. 1(b) and 1(e) reveal that amorphization occurs partially in the specimens irradiated to a fluence of 2×10^{14} Au/cm^2, and complete amorphization from the surface to the *end-of-range* of the projectiles is observed in the specimens irradiated to a fluence of 5×10^{14} Au/cm^2 (Fig. 1(c) and 1(f)). The resistance of NE- and SC-SiC to radiation-induced amorphization can be examined by comparing the results in Fig. 1(b) and 1(e). The locations of amorphous/crystalline interface (160–540 nm for SC-SiC and 190–570 nm for NE-SiC, marked with lines) are almost the same for these materials. The graph overlapped on the dark-field image is damage and Au distributions estimated by Monte Carlo simulations based on SRIM code [17]. Our recent study of Au ion irradiated SiC demonstrated a significant overestimation of electronic stopping powers in the energy region up to 10 MeV predicted by the SRIM code, which leads to large errors in predicting the damage profile and ion range in SiC [18]. The discrepancy between experiments and simulations can be compensated by reducing the atomic density of the target for the calculation, though there is no physical meaning. The assumed sample density in the SRIM calculations is 2.41 g/cm^3 (75% of the theoretical density (3.21 g/cm^3)), with threshold displacement energies of 20 and 35 eV for the C and Si sublattices, respectively [19]. The Au concentration is ~0.01 at%, and therefore the impurity effects on amorphization are negligible. The SRIM result reveals that the near-surface amorphous/crystalline interface corresponding to 0.53 and 0.57 dpa, respectively, for SC-SiC and NE-SiC, which suggest that the radiation tolerance of NE- and SC-SiC is comparable under these conditions. (Because of large volume swelling associated with amorphization, the damage dose at the interface is not discussed here.) On the other hand, *in situ* TEM irradiation with 1 MeV Kr ions revealed that NE-SiC possesses a lower radiation resistance than microcrystalline SiC [14]. The Kr irradiations were performed parallel to the stacking faults and twins rather than perpendicular to the faults as we have done, and therefore the discrepancy between the present and previous studies is attributed to the difference in irradiation conditions.

To examine the effects of ion energy, we performed 10 MeV Au ion irradiation. Fig. 2 shows cross-sectional dark-field images of (a) SC-SiC and (b) NE-SiC irradiated to a fluence of 1×10^{15} Au/cm^2. These pictures include the full view (~3.75 μm thick) of the irradiated region. The dark (A) and bright (B) contrasts correspond to irradiated SiC thin film and Si substrate, respectively, while the black region (C) is the unirradiated Si substrate. The damage and Au distributions calculated by SRIM are overlapped on the images. For the calculation, the atomic density was set to 2.41 g/cm^3 for SiC (the same as that of Fig. 1) and 1.86 g/cm^3 for Si (80% of the theoretical density (2.33 g/cm^3)). The thickness of SiC layer is 1100 nm for Fig. 2(a) and 530 nm for Fig. 2(b), respectively. Note that the damage distribution estimated by the calculations is shallower than that of the experiments, because volume swelling associated with amorphization is not considered. The *end-of-range* in Fig. 2(b) is slightly deeper than that in Fig. 2(a), which is mainly attributed to the difference in thickness of the more dense SiC thin films. The damage distributions in the SiC thin films are nearly identical between Fig.1(b) and 1(e), suggesting the atomic density of NE-SiC is the same as that of SC-SiC. Since the SC-SiC is thicker than NE-SiC, there is more energy loss by the Au ions in penetrating the SC-SiC before entering the Si substrate. As a consequence, the Au ion range in Si is shorter with increasing thickness of the SiC film. The results in Fig. 2(a) and 2(b) support this interpretation.

The electron diffraction experiments reveal the existence of both the crystalline and amorphous phases in the SiC thin films (see the inset of Fig. 2(c) and 2(d)). To determine the amorphous/crystalline interface, the dark-field TEM images obtained near the surface of SC-SiC and NE-SiC are shown in Fig. 2(c) and 2(d), respectively. The bright contrast corresponding to the crystalline phase decreases in intensity with increasing depth, and finally disappears due to amorphization at depths of 270 nm (1.1 dpa) and 480 nm (1.2 dpa) for SC-SiC and NE-SiC, respectively. The contrast due to the existing crystalline phase are clearly observed in the deeper region in Fig. 2(d) than (c), indicating that the local threshold dose for amorphization of NE-SiC is higher than that for SC-SiC under these conditions. To confirm the spatial distribution of the residual crystallinity quantitatively, the contrast intensity profile was examined. Fig. 2(e) shows the line-profile of the contrast intensity as a function of depth. For NE-SiC, the intensity profile was obtained from a single grain. The scale of the longitudinal axis is the same as that of the dark-field image and the broken line denotes the background intensity. These results confirm that the contrast well above the background intensity level is observed to a deeper region in NE-SiC than SC-SiC.

The present study indicates that the amorphization resistance is comparable between NE-SiC and SC-SiC under 2 MeV Au ion irradiation at room temperature, whereas the former exhibits slightly greater radiation tolerance than the latter under 10 MeV Au irradiation. We have recently examined the threshold dose for amorphization of NE-SiC under 2 MeV Si ion irradiation, and found that NE-SiC is six times more radiation resistant than single-crystalline SiC at room temperature [13], which is much larger than observed in the present study under Au ion irradiation. Jamison et al. [20] examined structural changes of NE-SiC under