DOSE RATE DEPENDENCE OF THE AMORPHIZATION OF SILICON CARBIDE –

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OBJECTIVE

The obejective of this work is to provide information on the dose rate dependence for amorphization of SiC in order to determine under which environments this phenomenon will be an issue for fusion applications.

SUMMARY

Single crystal silicon carbide (SiC) has been 2 MeV silicon ion irradiated in various irradiation temperature and ion flux ranges to measure the effect of these parameters on the critical dose for amorphization. The temperature and flux range for which amorphization was observed ranged from 80 to 400 K and 0.066 to 3 x 10^{-4} dpa/s, respectively. The critical dose, D_{crit}, was found by locating the depth of the boundary between partially crystalline and fully amorphous material using dark field TEM from samples prepared in cross section. This depth was compared to the damage profile as calculated using the TRIM-96 code. The temperature dependence of D_{crit} is found to agree well with previously reported values, though new evidence suggests a defect species becoming mobile in the 250-300 K range. Also of significance is that D_{crit} was dependent on flux at 340 K, ranging from 0.79 displacements per atom at the lowest ion flux to ~0.6 dpa at the highest flux level. The dose rate dependence of D_{crit} is compared with a chemical rate theory model previously described by the authors. It is seen that the dose rate dependence is substantially weaker than theorized. An extrapolation of the measured dose rate dependence is also compared with recent data on fast neutron amorphized SiC.

PROGRESS AND STATUS

1. Introduction

The amorphization of SiC has been widely studied in support of SiC electronic applications. The bulk of the work utilized high-energy ions and electrons to cause atomic displacement-driven amorphization. Numerous studies [1-10] have shown that SiC becomes amorphous during ion-beam irradiation at temperatures between 77 K and room temperature for damage levels of ~0.1 to 0.5 displacements per atom (dpa.) Over the past several years there has been substantial effort to understand the role of irradiation temperature on D_{crit} [9,11-17]. From this work the onset of dynamic recovery stages have been identified for which D_{crit} increase with increasing temperature. The absolute value of D_{crit} and the onset of these dynamic recovery stages appear to be dependent not only on the irradiation temperature but also on the irradiating species. For example, D_{crit} for electrons has been shown [15] to be ~0.7 dpa at ~50 K, increases to a plateau region of ~ 1 dpa in the 80 to 200 K range and then to increase rapidly with the crystal becoming non-amorphizable above ~300 K. This is compared with a heavy Xe ion irradiation [18] where D_{crit} was approximately ~0.19 dpa at 40 K, increases to ~ 0.24 dpa at 100K and then increases rapidly with temperature at ~500 K. It is noted that the reported doses for these two examples have been recalculated assuming sublattice-averaged displacement energy of 40 eV.

Other variables such as crystal structure (α vs β) [10,14,15] and the effect of implanted species [17] on the amorphization of SiC have been studied. However, the effect of dose rate on the amorphizability of SiC has not been investigated to this point, though it is known that displacement dose rate has a strong influence on the amorphizability of the silicon crystal [19]. This paper presents evidence for dose rate effects in the amorphization of SiC under silicon ion irradiation. The data are then compared to a chemical rate theory model previously used to describe the dose rate and temperature effects on the amorphization of SiC. Finally, the dose rate data is compared with some limited recent work demonstrating that SiC becomes amorphous by very low dose rate fission-neutron irradiation [20].

2. Experimental Procedure

Single crystal 6H alpha silicon carbide was procured as on-axis n-type wafers from Cree Systems, Inc. The doping level was approximately 4×10^{17} nitrogen atoms/cm^3. Discs, 3 mm in diameter were cut from the 0.36 mm thick wafers and mounted with either Aquadag™ or silver paint to a copper target block. Wafers were received with both sides polished and no further surface preparation was performed prior to irradiation. Two MeV Si ion implantation was carried out with the Tandem Van de Graaff accelerator in the Surface Modification and Characterization (SMAC) user facility of the Solid State Division of ORNL. Temperature was measured by a thermocouple embedded in the target block and controlled by a combination of resistive heating, forced air and liquid nitrogen cooling within ±3°C during irradiation.

Transmission electron microscopy (TEM) samples were prepared by bonding the irradiated wafer face to an identical, non-irradiated sample, followed by diamond slicing of the sandwich. Specimens were mechanically thinned and ion milled with argon ions at 6 keV and 15° using a liquid nitrogen cooled stage during milling. A final mill was performed at 3 keV, 11° for 10 minutes at liquid nitrogen temperature to reduce near-surface amorphization associated with argon implantation. A Phillips CM-30 microscope was used for the TEM. Data on amorphization was found by measuring the depth of the transition from partially crystalline to completely amorphous using weak beam dark field (WBDF) TEM imaging in a (g,3g), g=0006 diffracting condition. This is a departure from our earlier work [9,17] which used the transition range found by TEM bright field imaging. Data on the amorphization threshold, D_{crit}, are found by comparing the measured transition depth with a TRIM-96 [21] calculation. Figure 1 shows the calculated dpa and ion concentration levels for a representative irradiation. Amorphization depth data were only obtained at < 1 nm depths in order to avoid the implanted region.

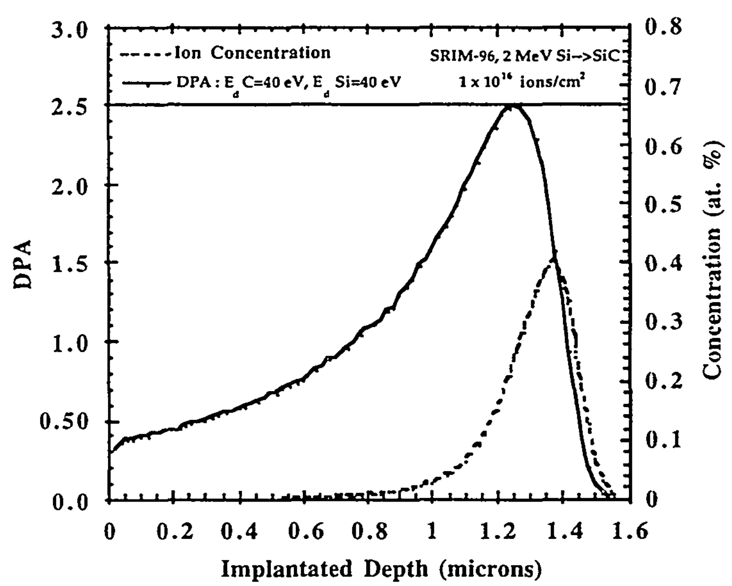

Figure 1

TRIM-96 Simulation of SiC implanted with 2 MeV Si to an ion fluence of 1×10^{16} ions/cm^2 assuming average displacement energy of 40 eV on both the silicon and carbon sublattices.

A cross sectional WBDF image for a sample which has been amorphized by a 2 MeV Si ion irradiation is shown in Figure 2A. Inset in the figure are the diffuse rings of the mid-range diffraction patterns and diffraction pattern depicting the 6H diffracting conditions used for the WBDF image. Figure 2B is a higher magnification WBDF image showing the transition from partially crystalline to fully amorphous structure at ~340 nm.

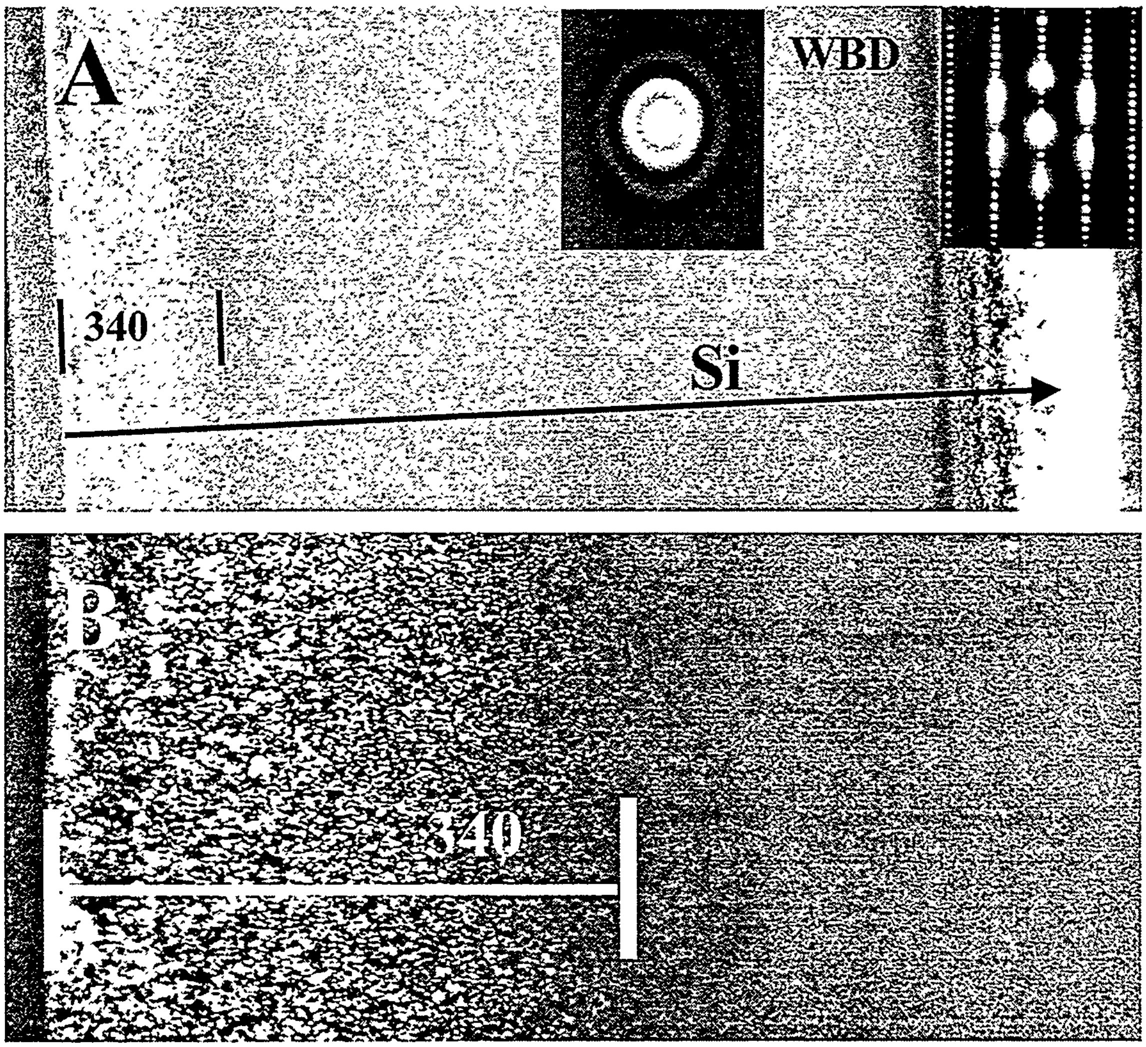

Figure 2: TEM WBDF images of 2 MeV Si ion irradiated SiC at 340 K to 1.2×10^{16} ion/cm^2.

Figure 3 shows the effect of D_{crit} as a function of irradiation temperature at a dose rate of 6.6×10^{-6} dpa/s. The 2 MeV Si ion displacement dose rate for this series varied in the range of $1\text{-}5 \times 10^{-4}$ dpa/s. In Figure 3, D_{crit} has increased from about 0.2 to ~0.25 dpa as the irradiation temperature increased from 80 to 100 K. Above this temperature an apparent plateau region exists until approximately 250 K at which point the D_{crit} begins to increase. Though the data is rather limited, it appears that the increase in D_{crit} begins to level off above 300 K and then rapidly increases above ~340 K.

Figure 4 gives the effect of dose rate on D_{crit} at an irradiation temperature of ~340 K. The figure shows the critical dose decreasing with increasing dose rate, dropping from 0.79 dpa to 0.6 dpa as the dose rate increased from $\sim 6.6 \times 10^{-6}$ dpa/s to 1.8×10^{-4} dpa/s. At the highest flux studied (2.9×10^{-4} dpa/s) the critical dose for amorphization increases slightly. The reason for this is unclear, though the trend is consistent with the increased beam heating at the elevated flux rate.

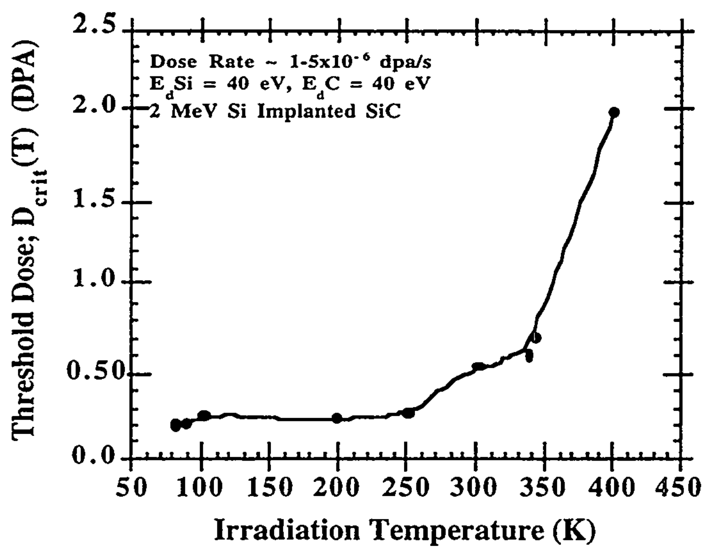

Figure 3: Threshold dose for amorphization as a function of irradiation temperature for Si ion irradiated single crystal alpha SiC.

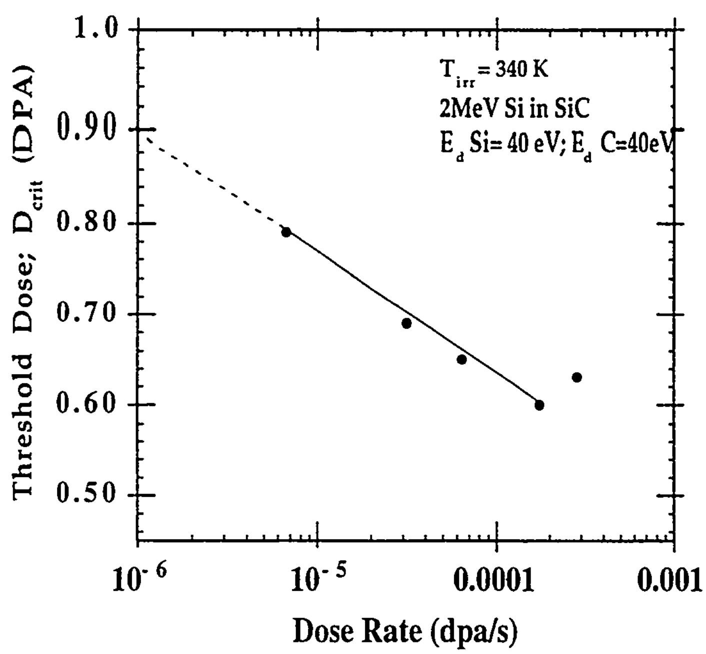

Figure 4: Threshold dose for amorphization as a function of dose rate for Si ion irradiated single crystal alpha SiC.

3. DISCUSSION

As was originally observed by Inui[14] for 2 MeV electron irradiation and by Weber and Wang[18] for 1.5 MeV xenon ion irradiation, the Si ion irradiations of this study show a measurable jump in the critical dose for amorphization above an irradiation temperature of ~90 K. It is noted that such a jump was not seen by Weber [22] for Ne and Ar results, which have closer atomic masses to Si, and therefore more similar cascade events, as compared with Xe. The increase in dose for amorphization (fig 3) demonstrates an increase in dynamic recovery for the crystal possibly due to the onset of carbon interstitial migration. The absolute value of D_{crit} for the Si ion irradiations of this paper increases from ~0.2 to 0.25 dpa which is in good agreement with earlier xenon and argon ion irradiation results [8] but substantially lower than the electron irradiation (~0.4->0.6 dpa) [14] results. This difference is likely due to the difference between the crystal lattice damage produced by high energy electrons, which is limited to the production simple point defects, as compared with the complex defects which can form within and contiguous to heavy-ion induced cascades. The amount of chemical disordering on a dpa-normalized basis is substantially higher for heavy ions as compared with high-energy electron irradiation. If chemical disordering assists the amorphization of SiC (by increasing the lattice free energy), this may explain the lower D_{crit} for ion irradiation as compared with electron irradiations and may also explain the increased temperature for which amorphization is possible with heavy ions. This assumes that in-cascade amorphization is not occurring in SiC, which has not been adequately demonstrated. Results from molecular dynamics simulations[23] and high-resolution TEM observations[10,15] argue against such in-cascade amorphization of SiC.

Above the 90 K dynamic recovery stage a rather broad plateau region (figure 3) exists for which no significant new recovery processes occur. At approximately 250 K the critical dose for amorphization begins to increase indicating the beginning of a second dynamic recovery stage. The trend for increasing amorphization dose for temperatures greater than 300 K was seen for the previous ion irradiations [22]. From figure 3 it appears that the temperature increase begins at a lower temperature than previously seen and may reached a second plateau region before rapidly increasing above 340 K. While additional data points are needed for confirmation, this data suggests a new, intermediate, dynamic recovery stage may be operating.

From the data of figure 4 it is apparent that the rate at which atoms are displaced in SiC does affect the critical dose required to drive the crystal amorphous. Specifically, the dose rate was increased by a factor of almost 50 leading to a reduction in the critical dose for amorphization of about 25%. The temperature at which this dose rate study was performed was 340 K, chosen based on the assumption that this temperature was well within the upper dynamic recovery stage. For this reason a reduction in D_{crit} with increasing dose rate is intuitive if one considers the amorphization occurring as a competition between irradiation-induced atomic displacements and thermally driven replacement of point defects.

In a previous paper [17] the authors modified the chemical rate theory model of Motta and Olander [24] to explain the temperature and dose rate dependent amorphization in SiC. In this model the dose rate (ϕ) dependence scales as the inverse square root of the damage dose rate. By manipulating the data of figure 3, a range in interstitial migration energies for the upper dynamic recovery was determined. [17] With this information the model was applied for the dose rate variation in this study. Analysis revealed that the dose rate dependence measured here is substantially weaker than predicted yielding a dose rate dependence proportional to $\phi^{-1/8}$ as compared with $\phi^{-1/2}$ for the model. One possible explanation for this discrepency may be that at the chosen irradiation temperature, ~340 K, was not sufficiently high for both carbon and silicon interstitials to be mobile. If in fact the evidence for a second plateau region in the range of ~300-350 K is accurate, both Si and C atoms are not fully mobile and the model should not be applied.

Furthermore, the dose rate dependence would be expected to increase from zero at the plateau to the predicted $\phi^{-1/2}$ dependence well above the plateau where full mobility is achieved. Clearly, this model needs to be reexamined regarding its dose rate dependence. Further evidence for the discrepancy between the model and empirical data is the existence of data [20,25] on neutron irradiated Cree β-SiC which fully amorphized at a dose rate of ~8 × 10^{-7} dpa/s. In this case the (above critical) dose was 2.6 dpa at an irradiation temperature of ~332 K, which may also reside on the second plateau. Applying the chemical rate theory model ($\phi^{-1/2}$) would have estimated a threshold dose of more than 10 dpa for amorphization. Furthermore, analysis of a cylindrical sample which was partially amorphized in the same neutron irradiation described above indicates amorphization occurred at approximately 0.9 dpa, which is in good agreement with the empirical $\phi^{-1/8}$ dependence depicted as the solid and dotted line in figure 4.

CONCLUSIONS

(1) A dynamic recovery stage was observed near 90 K for 2 MeV Si ion amorphized alpha SiC in good agreement with previous high-energy electron and xenon irradiations. Evidence indicated that the upper dynamic recovery stage previously identified at ~350 to 400 K may be a combination of at least two independent stages.

(2) A weak dose rate dependence on the amorphization of single crystal alpha-SiC exists at 340 K. As the dose rate is increased, the damage level required to drive the crystal amorphous is decreased. The dose rate dependence is empirically shown to follow a $\phi^{-1/8}$ dependence at 340 K compared with the $\phi^{-1/2}$ of the model previously discussed by the authors. It is speculated that this weaker than predicted dependence relates to a second, previously unidentified annealing stage. The calculated critical dose for amorphization of a fast neutron irradiation induced amorphous SiC sample is consistent with the extrapolated dose rate data from this study.

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