3.3 PROGRESS IN CHARACTERIZATION OF PRECIPITATES AND DEFECT STRUCTURES IN Mg^+ ION IMPLANTED CUBIC SILICON CARBIDE — W. Jiang, J. Zhang, Z. Zhu, T. J. Roosendaal, S. Y. Hu, C. H. Henager, Jr., R. J. Kurtz (Pacific Northwest National Laboratory), and Y. Wang (Los Alamos National Laboratory)

OBJECTIVE

This study aims at characterizing precipitates and defect structures in Mg^+ ion implanted and high-temperature annealed cubic silicon carbide (3C-SiC).

SUMMARY

This report describes the progress of our current experimental effort on Mg^+ ion implanted 3C-SiC. Following our initial study [1] that suggests possible formation of Mg_2Si and MgC_2 precipitates as well as tetrahedral voids in $^{24}Mg^+$ ion implanted 3C-SiC, we have designed specific experiments to confirm the results and examine the inclusions and defects. Relatively low fluence (5.0×10^{15} $^{24}Mg^+/cm^2$) implantation in 3C-SiC was performed to reduce defect concentrations and isolate individual defect features for characterization. In addition, $^{25}Mg^+$ isotope was implanted in 3C-SiC to the same previously applied ion fluence (9.6×10^{16} ions/cm^2) for atom probe tomography (APT) study of precipitates. Each set of the samples was annealed at 1573 K for 2, 6 and 12 h, respectively. The depth profiles of the implanted Mg were measured using secondary ion mass spectrometry (SIMS) before and after the annealing steps. The samples are currently being analyzed using transmission electron microscopy (TEM) and APT.

PROGRESS AND STATUS

Introduction

Cubic silicon carbide has been a candidate material for a number of applications in fusion reactor designs [2,3]. Upon exposure to high-energy neutrons, silicon carbide undergoes transmutation reactions with magnesium as the major metallic transmutant, as predicted by Sawan, *et al*. [4]. The impact of the transmutants and irradiation induced defects on SiC structural modifications and property degradation is currently unknown. Our previous scanning transmission electron microscopy (STEM) study [1] suggests that precipitates of cubic Mg_2Si and tetragonal MgC_2 as well as tetrahedral voids of ~5 nm in size are likely formed in 3C-SiC implanted to 9.6×10^{16} $^{24}Mg^+/cm^2$ at 673 K and subsequently annealed at 1573 K for 12 h. As the defect concentration was too high and the defect features could not be spatially resolved by TEM, the conventional under- and over-focus imaging technique could not be used to confirm the defect nature. Likewise, we cannot ascertain whether the observed bright-contrast triangular shape in the STEM image is associated with stacking fault tetrahedra (SFTs). To increase the probability of generating isolated defect features, samples were implanted to an order of magnitude lower in ion fluence (5.0×10^{15} $^{24}Mg^+/cm^2$) at 673 K. The low-dose samples have been annealed at 1573 K and are currently being examined to study defect structures, including possible tetrahedral voids, stacking fault tetrahedra, and Mg nucleation site using atomic-level resolution aberration-corrected TEM.

In addition, the precipitates of cubic Mg_2Si and tetragonal MgC_2 were not observed directly, but derived from fast Fourier transformation (FFT) of a high-resolution STEM image [1]. APT was attempted to confirm and better characterize the precipitates, but element ^{24}Mg and atomic cluster C_2 could not be resolved in the mass spectra. To solve the technical issues, ^{25}Mg isotope was obtained and $^{25}Mg^+$ ion implantation (9.6×10^{16} $^{25}Mg^+/cm^2$ at 673 K) was performed. The implanted 3C-SiC samples have been annealed at 1073 and 1573 K and are being analyzed using APT. The 1073 K annealed sample is intended for analysis without involving voids in the crystal structure. Reported here are some of the initial data, including x-ray diffraction (XRD) pole figures for the 3C-SiC film before ion implantation and the depth profiles of the implanted Mg by SIMS before and after the thermal annealing steps.

Experimental Procedure

Single crystal (001)-oriented 3C-SiC films (~2 µm in thickness) on Si (001) substrates used in this study were characterized using XRD pole figures. Scans were performed for 3C-SiC (111) and (220) poles at 2θ = 35.597° and 59.978°, respectively. The samples were implanted 7° off the surface normal with 200 keV $^{25}Mg^{+}$ ions at 673 K to a high fluence of 9.6×10^{16} ions/cm^2. The ion fluence corresponds to maxima of ~6 at.% ^{25}Mg at 280 nm and 54 dpa at 220 nm according to SRIM simulation [5], as shown in Figure 1. The isotope ^{25}Mg was obtained from National Isotope Development Center (NIDC) at Oak Ridge and the implantation was performed at Los Alamos National Laboratory (LANL). Additional implantation was also performed at LANL using 200 keV $^{24}Mg^{+}$ ions to a lower fluence of 5×10^{15} ions/cm^2 at the same elevated temperature. The implanted sample was cleaved into smaller pieces (5 mm × 5 mm) for different thermal annealing conditions. Each set of the low and higher dose samples was thermally annealed at 1573 K for 2, 6 and 12 h in flowing Ar gas, respectively. Additional annealing of the $^{25}Mg^{+}$ ion implanted sample was also performed at 1073 K for 12 h to prevent void formation due to vacancy clustering in the structure. To minimize surface oxidation, the samples were placed inside a chimney of Ta metallic foil to reduce the oxygen partial pressure. The Mg depth profiles in the SiC samples were measured before and after the annealing steps using time-of-flight SIMS (ToF-SIMS) with 2 keV O_2^{+} ions as sputtering beam and 25 keV Bi^{+} ions as analyzing beam. The depth scale was calibrated by measuring the crater depth with a Veeco Dektak 150 stylus profilometer.

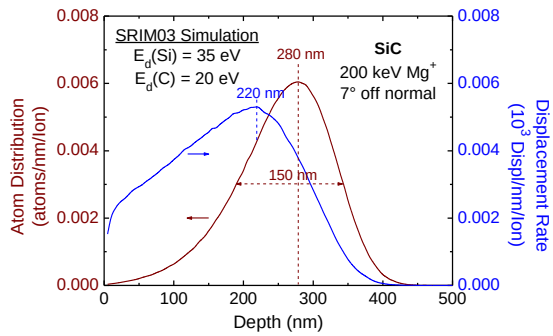

Figure 1. Depth profiles of Mg implants and atomic displacement rate in Mg^{+} ion implanted SiC from SRIM simulation.

Results and Discussion

The 3C-SiC films on Si substrates used in this study were analyzed using XRD pole figures before ion implantation. The data are shown in Figure 2, along with a stereographic display of poles from simulation. The data clearly show four symmetric 3C-SiC (111) and (220) [equivalent to (110)] poles, respectively, suggesting that the film is a single crystal with a nearly on-axis (001) cut. Additional symmetric scans of the sample (data not shown) indicate that the crystal has high monocrystalline quality without presence of any secondary phases.

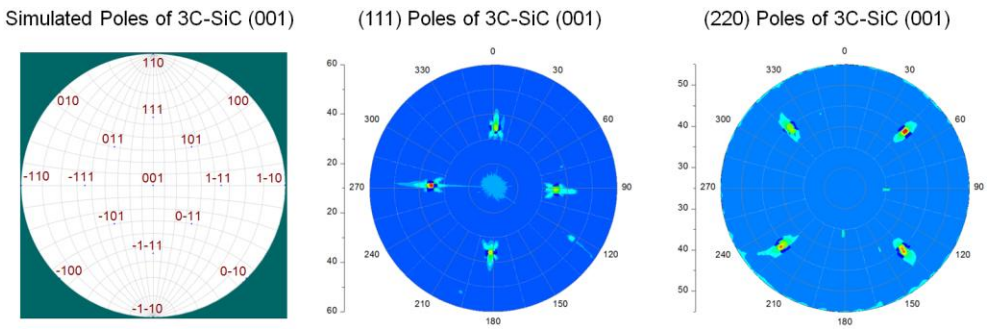

Figure 2. Comparison of simulated poles of (001) oriented 3C-SiC with the experimental data for (111) and (220) poles of an as-grown 3C-SiC film on Si (001).

Prior to annealing of the ion implanted samples, an as-grown 3C-SiC was furnace annealed for a test inside a Ta chimney at 1573 K under Ar gas flow. ToF-SIMS was used to measure the oxygen concentration for assessment of surface oxidation. The data are shown in Figure 3. Oxidation from the surface to a depth of ~10 nm was observed after the annealing for 12 h, but more extensive surface oxidation beyond the depth did not occur. This annealing condition was applied for the Mg^+ ion implanted 3C-SiC.

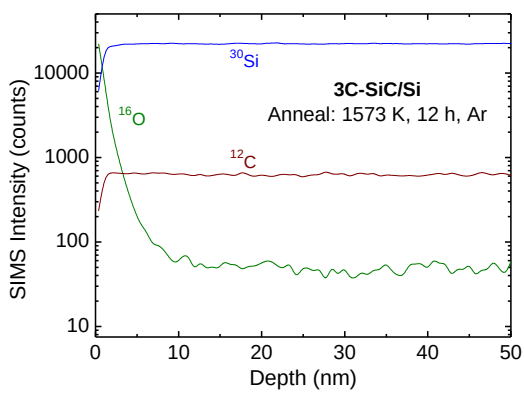

Figure 3. ToF-SIMS depth profiles of ^{16}O, ^{30}Si and ^{12}C in an as-grown single crystal 3C-SiC film on Si annealed at 1573 K in Ar gas.

The normalized SIMS depth profiles of the implanted Mg in 3C-SiC are shown in Figure 4. The normalization follows $I'_{Mg}(i) = 100I_{Mg}(i)/[w\Sigma I_{Mg}(j)]$, where $I_{Mg}(i)$ is the Mg intensity at channel number i and w is the channel width. This normalization procedure does not alter full width at half maximum (FWHM) of the peak, but allows for all the ToF-SIMS data measured under different conditions (dwelling time of sputtering beam, beam current, etc.) to be plotted in one figure for comparison. In Figure 4(a), the peak maximum of the ^{24}Mg profile in the as-implanted 3C-SiC is located at ~240 nm with FWHM of ~145 nm, which is consistent with our previous observation [1], but slightly shallower than the SRIM prediction (Figure 1). Similarly, the profile peak for ^{25}Mg in Figure 4(b) is also at ~240 nm with a slightly larger FWHM of ~160 nm. There is no significant difference in the ^{24}Mg and ^{25}Mg profiles, as expected. According to Figure 4, annealing at 1573 K for 2, 6 and 12 h does not lead to a peak position shift or a noticeable change in the shape of the Mg depth profiles, indicating that there is no significant Mg diffusion during the thermal annealing. The small deviation shown for the $^{25}Mg^+$ ion implanted and 6 h annealed

sample may be due to the uncertainty in the depth calibration. It should be noted that this non-diffusion behavior is not the same as that observed in a previous study [1], where Mg diffused into the bulk of a 3C-SiC single crystal after annealing at 1573 K for 12 h. Possible reasons include some difference in the

actual sample temperature during the thermal annealing in the two cases because significant Mg diffusion in single crystal SiC starts to occur between 1473 and 1573 K [1].

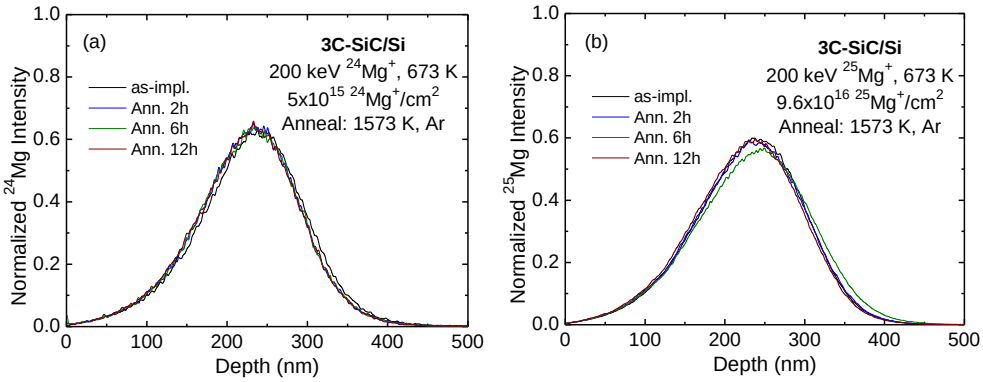

Figure 4. ToF-SIMS depth profiles of (a) ^{24}Mg and (b) ^{25}Mg in single crystal 3C-SiC films on Si before and after furnace annealing at 1573 K for 2, 6 and 12 h, respectively, in Ar gas.

At 1573 K, implantation induced vacancies in 3C-SiC are mobile and vacancy clustering is expected to occur during the thermal annealing. Although long-range diffusion of the implanted Mg in 3C-SiC does not occur, short-range migration of Mg is possible, which can lead to precipitation of secondary phases. In fact, preliminary APT data (not shown) have suggested formation of precipitates that contain ^{25}Mg in the 12 h annealed sample. This study is focused on the formation and growth of possible voids and precipitates as a function of annealing duration at 1573 K as well as the dependence of the precipitate characteristics on annealing temperature. We will report the results when the data are available.

Acknowledgements

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