



Dose dependence of helium bubble formation in nano-engineered SiC at 700 °C



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ARTICLE INFO

Article history:

Received 28 August 2015

Received in revised form

26 January 2016

Accepted 29 January 2016

Available online 3 February 2016

Keywords:

SiC

Helium

High temperature

Irradiation effects

TEM

ERDA

ABSTRACT

Knowledge of radiation-induced helium bubble nucleation and growth in SiC is essential for applications in fusion and fission environments. Here we report the evolution of microstructure in nano-engineered (NE) 3C SiC, pre-implanted with helium, under heavy ion irradiation at 700 °C up to doses of 30 displacements per atom (dpa). Elastic recoil detection analysis confirms that the as-implanted helium depth profile does not change under irradiation to 30 dpa at 700 °C. While the helium bubble size distribution becomes narrower with increasing dose, the average size of bubbles remains unchanged and the density of bubbles increases somewhat with dose. These results are consistent with a long helium bubble incubation process under continued irradiation at 700 °C up to 30 dpa, similar to that reported under dual and triple beam irradiation at much higher temperatures. The formation of bubbles at this low temperature is enhanced by the nano-layered stacking fault structure in the NE SiC, which enhances point defect mobility parallel to the stacking faults. This stacking fault structure is stable at 700 °C up to 30 dpa and suppresses the formation of dislocation loops normally observed under these irradiation conditions.

Published by Elsevier B.V.

1. Introduction

Because of its low neutron capture cross-section, outstanding chemical and thermal stability, silicon carbide (SiC) is considered as a promising candidate for structural component and cladding material in the next generation fusion and fission nuclear reactors [1–3]. For advanced nuclear applications, SiC will be exposed to high temperatures, intense fluxes of fast neutrons, and the production of helium and hydrogen gas atoms from nuclear reactions. In advanced fission reactors, the helium generation rate is reported to be 2.5 appm He/dpa [4]. In a fusion reactor, SiC will be exposed to not only a high irradiation dose (up to 200 dpa), but also high

helium generation rates (70 appm/dpa in the reactor, 130 appm/dpa for the first wall) [5–7]. The damage accumulation from high temperature irradiation may lead to different types of defects that will induce hardening effects, microstructure evolution and volume swelling [8]. In order to enhance the high temperature radiation tolerance, the response of different types of SiC to radiation damage has been investigated both computationally [9–14] and experimentally [7,8,15–17]. Ion irradiation is widely employed in experimental studies since it provides an effective approach [18–20] to investigate the irradiation response of SiC to high irradiation dose and high helium accumulation at controlled temperatures. Some promising paths for improving radiation resistance, such as grain refinement [21], fiber-bonded reinforced composites [22,23], and increase volume fraction of grain boundaries [24,25], have been reported. Composites made with the advanced fibers of Nicalon Type S and the UBE Tyranno SA retain dimensional stability and have negligible change in strength up to 10 dpa at 800 °C [5]. SiC composites also show great irradiation resistance under neutron irradiation to doses of 30–40 dpa at 300–800 °C [26]. However, the

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irradiation performance of the composite is strongly affected by the degradation of interfacial shear properties [27]. Recent investigations have shown that nano-engineered (NE) SiC, which contains high-densities of stacking faults (SFs) with nm spacing formed within a nano-sized grain structure, can confine three-dimensional random point defect migration to a two-dimensional like movement parallel to the SFs [24,28]. As illustrated in Fig. 1, the irradiation-induced defects encounter higher migration barriers across the SFs; thus point defects can either migrate two-dimensionally parallel to the SFs or recombine within a short distance. As a result, this nano-engineered structure enhances the rate of point defect annihilation, leading to improved radiation tolerance of SiC.

Helium pre-implanted NE SiC is employed in this study to investigate subsequent heavy ion irradiation on bubble formation. Although two-dimensional diffusion is supported by density functional theory calculations [25], the nucleation and growth of helium bubbles in NE SiC, as well as the irradiation response and stability of this novel structure at high temperature, are not well understood. Also, the high-density of SFs with average spacing of 1–2 nm could result in significant defect migration and bubble coarsening along grain boundaries, especially under high dose heavy ion irradiation. Such bubble growth at grain boundaries may lead to degradation of mechanical properties. In our previous study [29], helium bubble formation was not observed in NE SiC following helium implantation at 277 °C nor after thermal annealing at 700 °C for 1 h. Helium bubbles of measurable size (up to 5 nm) were only observed after Au ion irradiation to 10 dpa at 700 °C. Consequently, we in the present work investigate the evolution of helium bubbles in NE SiC at much higher doses under prolonged irradiation (up to 30 dpa) at 700 °C in order to evaluate helium bubble growth and microstructural evolution, and to gain insight on the performance of NE SiC under extreme irradiation environments.

2. Experimental methods

NE SiC with a high density of <111>-type SFs (spacing of 1–2 nm) within columnar grains of 100–300 nm size were grown along the <111> direction on (1 0 0) silicon substrates using low-pressure chemical vapor deposition. Processing techniques and fabrication of these NE SiC thin films are described elsewhere [29]. The average thickness of the NE SiC films is 530 nm. In this study, the corresponding implanted helium concentration and dose in displacements per atom (dpa) were calculated using Stopping and Range of Ions in Matter (SRIM) full-cascade simulations (version 2012) [30,31]. The parameters used in the simulations are threshold displacement energies of 20 and 35 eV for the C and Si atoms, respectively [32], and a sample density of 3.21 g cm^{-3} .

As the first step, helium ion implantations were performed using the 200 kV ion-implanter at Los Alamos National Laboratory (LANL) [33]. The SiC thin films were implanted with 65 keV helium ions to fluences from 1×10^{15} to $1 \times 10^{16} \text{ ions cm}^{-2}$ at a

temperature of 277 °C, which is above the critical temperature for amorphization [34]. The helium concentration peak was located at 320 nm from the surface, as illustrated in Fig. 2. After the helium ions were implanted, subsequent heavy ion irradiations were carried out in a multi-purpose target chamber using the 3.0 MV tandem accelerator facilities in the Ion Beam Materials Laboratory at the University of Tennessee [35].

The ion irradiations were conducted using 9 MeV Au^{3+} ions at 700 °C to ion fluences of 1.5×10^{16} (average dose of 20 dpa) and $2.3 \times 10^{16} \text{ ions cm}^{-2}$ (30 dpa) to produce a relatively flat-damage profile at the helium peak region (depth of 270–390 nm) under

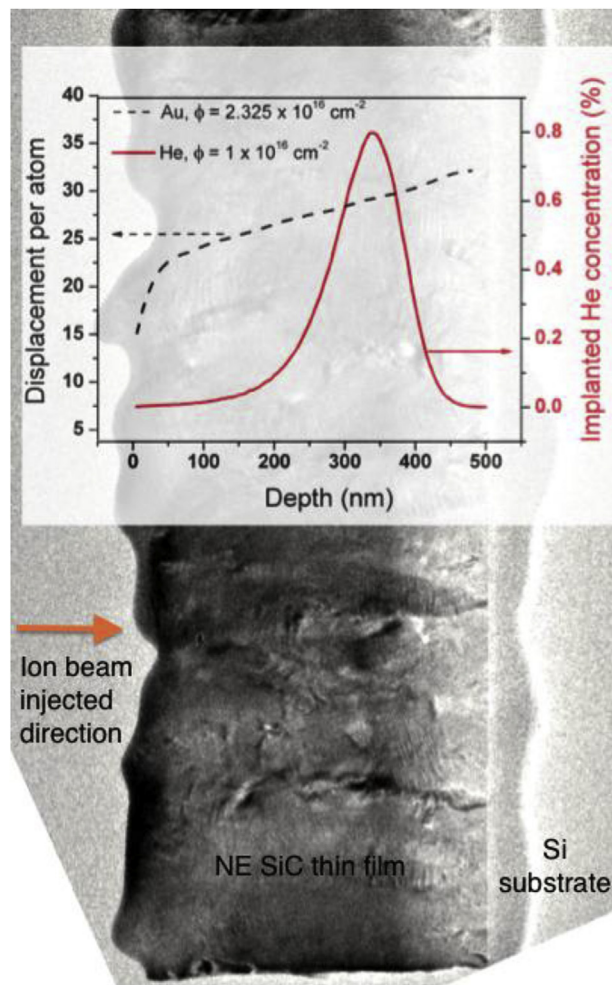


Fig. 2. SRIM 2012 simulation of the irradiation damage prediction and implanted helium concentration in NE SiC. The depth profile of SiC film on Si substrate is shown in background TEM image.

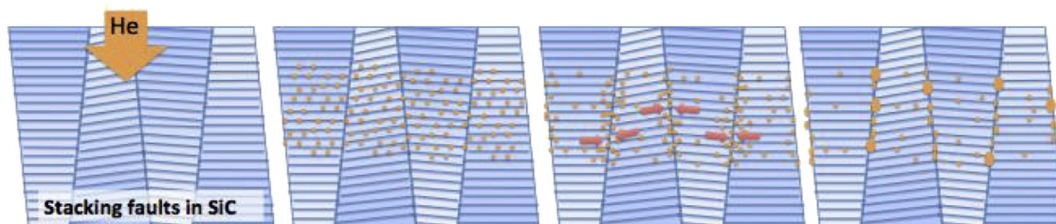


Fig. 1. Defect diffusion schematics. Migration occurs mainly between SFs, following a two-dimensional process in NE SiC. Since SFs prevent defect migrating across layer parallel to surface, helium atoms may tend to diffuse to grain boundaries.

high vacuum conditions (9.8×10^{-8} torr). The energy of the ion beam was chosen such that the damage energy deposited into the NE SiC film has a smooth gradient, especially over the depth range of the helium concentration peak, while minimizing the Au concentration in the SiC film. The ion flux was kept constant ($2.3 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$) during the Au irradiation, and the ion beam was rastered over the irradiated area of $10 \times 10 \text{ mm}^2$ to ensure a uniform irradiation (at scanning frequencies of 517 and 64 Hz for horizontal and vertical direction, respectively, and a current density of 11.1 nA mm^{-2}). The average fluence-to-dpa conversion factor was 0.13 dpa per 10^{14} ions cm^{-2} at the helium peak regime (i.e. 270–390 nm). A glass scintillator and a CCD camera were used to accurately locate the ion beam. The temperature was controlled using a HRN (LPS-800-1) heater controller from Thermionics Northwest Inc. [36], details have been discussed elsewhere [29].

The helium depth distributions are investigated using Time-of-Flight Elastic Recoil Detection Analysis (ToF-ERDA) system. ToF-ERDA, complementary to RBS and conventional ERDA, can detect forward-scattered recoiling atoms from a single collision. Equipped with a Si detector and two time-of-flight detectors, ToF1 and ToF2, with a corresponding ToF distance of 647.5 mm in a forward scattering geometry [35], the velocity and energy of the individual recoil atoms from the target can be measured simultaneously in coincident mode. Both ToF1 and ToF2 contain thin carbon foils, and when a recoil atom passes through each foil, secondary electrons are ejected and detected to trigger a timing event for each detector, from which the velocity is determined. To determine the helium depth distribution, 20 MeV Ni^{8+} and 18 MeV Ti^{7+} ion beams were employed to scatter the elements in NE SiC samples (Ni beam for the heavy ion irradiated samples and Ti beam for the non-irradiated one, respectively). The schematic diagram of ToF-ERDA set-up is shown in Fig. 3. The system setup is provided elsewhere [35,37].

Post-irradiation microstructural evolution characterization was performed using transmission electron microscopy (TEM). Cross-sectional TEM specimens were prepared by mechanically polishing down to a thickness below $20 \mu\text{m}$, which was followed by ion milling in a Gatan precision ion polishing system with beam energy from 5 keV down to 3 keV. The specimens were characterized using a ZEISS Libra 200 MC transmission-electron-microscope operating at 200 kV. In this work, all images for bubble analysis were recorded in a slightly underfocused condition (with a defocus value of about 900 nm, far away from the Scherzer defocus condition). By measuring the inside edge of the first dark Fresnel fringe, the bubble can be delineated reasonably well for either sphere or faceted bubbles. Since most of the bubbles observed in this work are spherical, by calculating the square root of the product of the longest inner diameter, d_1 , and the shortest inner diameter, d_2 , an average diameter of each bubble, $(d_1 \times d_2)^{0.5}$, can be estimated. Because some bubbles are not perfectly round, several TEM images

were taken to obtain an average diameter at each condition.

TEM Specimen thickness was estimated with the Log-ratio method [38]. In this method, electron signal from the whole spectrum (I_t) is compared with the signal from the zero-loss peak (I_0) in a low loss EELS spectrum. With the Poisson statistics relation

$$t/\lambda = \ln(I_t/I_0)$$

where λ is the inelastic mean free path of electrons in the SiC (i.e. 136.43 nm for 200 keV). Instead of a precise value of thickness, the EELS measurement yields an estimated thickness. In this study, considering the uncertainty of the EELS measurement, for most of specimens, the estimated thicknesses of observed regions are between 100 and 200 nm. For all estimates of bubble densities, a 150 nm thickness is assumed.

3. Experimental results and analysis

Helium depth distributions in NE SiC before/after ion irradiation acquired from ToF-ERDA are shown in Fig. 4. The ToF-ERDA spectra indicates that the helium distribution is slightly shallower and about 10% higher than the SRIM2012 predictions, which is within experimental error and consistent with previous results obtained using conventional ERDA measurement [29]. Although the free surface and film/substrate interface acts as sinks for helium, helium atoms were only observed in a depth range of about $\pm 60 \text{ nm}$ from the helium injected peak position (i.e. 330 nm from the surface). No broadening or redistribution of helium depth distribution was observed along the ion path, even after thermal annealing or heavy-ion irradiation.

The microstructural evolution in NE SiC induced by helium implantation and subsequent heavy ion irradiation is shown in Fig. 5. All TEM micrographs were recorded at a depth of about 350 nm from surface (near the helium concentration peak), which is about 150 nm from the film/substrate interface. For each specimen, bubble size and density measurements were performed using more than three different selected areas, each with a size of $50 \times 50 \text{ nm}^2$, to reduce the statistical uncertainty. Most of the uncertainty in bubble density values is due to the thickness variation from the TEM sample.

Following the 9 MeV Au^{3+} irradiation to a dose of 20 dpa, helium bubbles could be observed in specimens with helium fluences of 1×10^{15} ions cm^{-2} (800 appm at peak) and 3×10^{15} ions cm^{-2} (2400 appm at peak), as shown in Fig. 5(a). The higher helium fluence sample (1×10^{16} ions cm^{-2}) was consumed in sample preparation. The average diameters of the helium bubbles are $1.7 \pm 0.4 \text{ nm}$ and $1.8 \pm 0.4 \text{ nm}$ for helium fluences of 1×10^{15} and 3×10^{15} ions cm^{-2} , respectively, as summarized in Table 1. Bubble nucleation and growth are driven by the continuous production of irradiation-induced defects. The number density of helium bubbles slightly decreased with increasing helium concentration, as shown in Table 2. The nucleation of helium bubbles at elevated temperature, as compared with the thermal annealing result (not shown in figure), is enhanced by the defects produced by the Au^{3+} ion irradiation.

As shown in Fig. 5(b), helium bubble formation is observed in specimens for all ranges of helium concentrations after irradiation with Au^{3+} ions to a dose of 30 dpa. The average diameters of the helium bubbles are $1.7 \pm 0.4 \text{ nm}$, $1.8 \pm 0.4 \text{ nm}$ and $2.4 \pm 0.7 \text{ nm}$, respectively, for the lowest (1×10^{15} ions cm^{-2}), medium (3×10^{15} ions cm^{-2}), and highest helium (1×10^{16} ions cm^{-2}) concentrations. Some of the larger helium bubbles are observed to be non-spherical; however, this asphericity is assumed to be an artifact from the TEM observation due to superposition of bubbles along the electron beam direction. Because the preferential helium

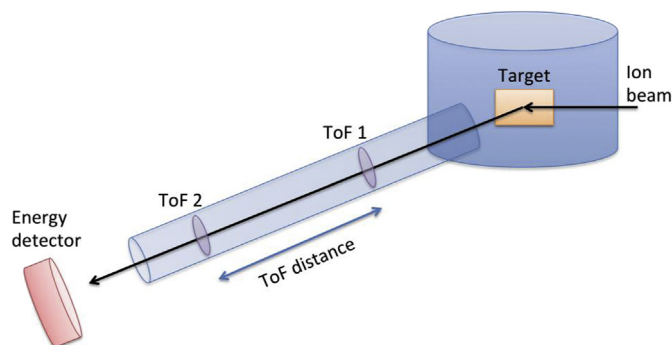


Fig. 3. Schematic diagram of ToF-ERDA set-up.

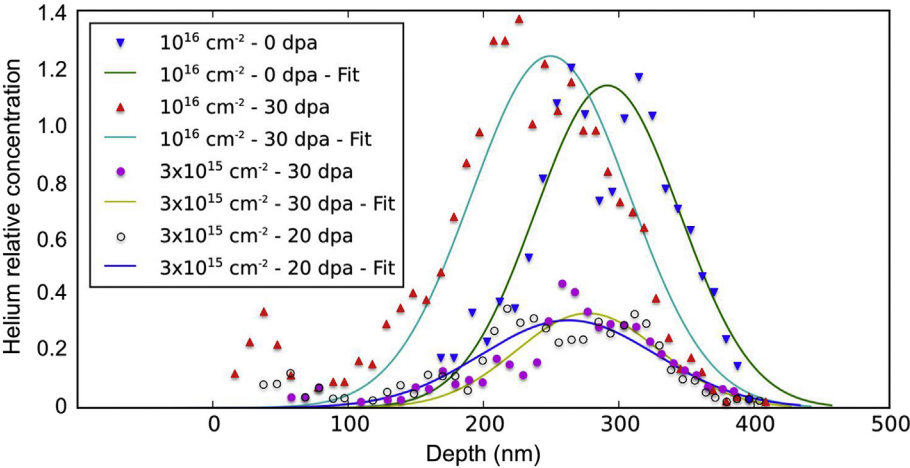


Fig. 4. Helium depth distribution and Gaussian fits (solid lines) from ToF-ERDA measurements in NE SiC before and after irradiation.

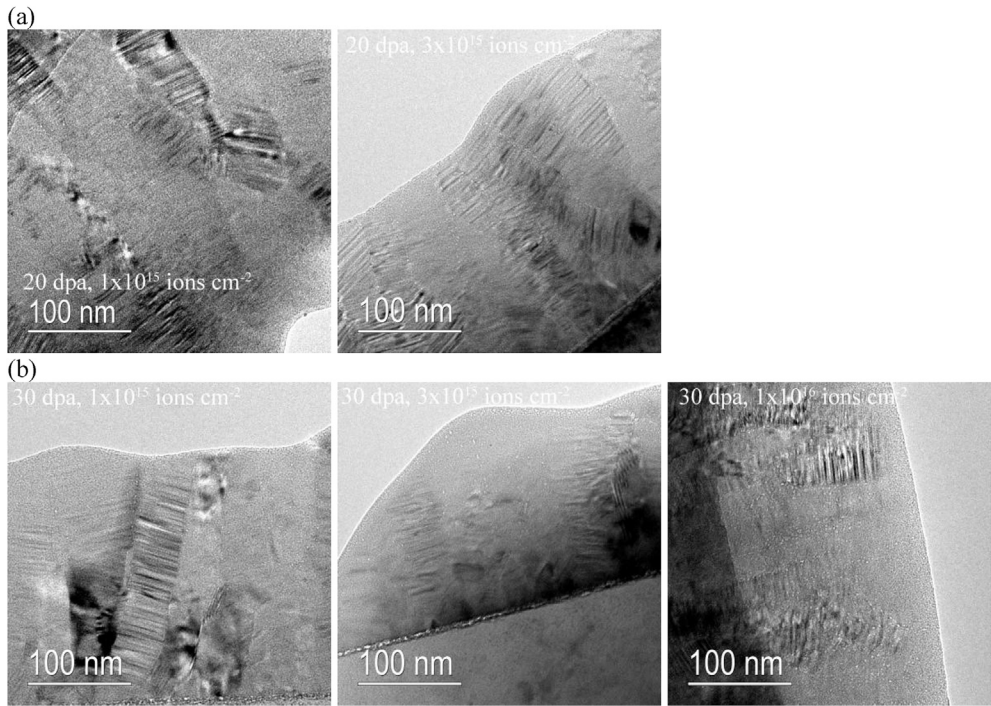


Fig. 5. Cross-sectional bright field TEM micrographs (acquired at 200 kV, 900 nm under-focus condition) for NE SiC after Au³⁺ ions irradiation to (a) 20 and (b) 30 dpa at 700 °C with He fluences from 1 × 10¹⁵ ions cm⁻² to 1 × 10¹⁶ ions cm⁻². All micrographs were taken at the depth range of 350 nm from the surface (or about 150 nm from the substrate).

Table 1
Averaged helium bubble diameters under different heavy ion irradiation doses.

He fluence (ions cm ⁻²)	He ⁺ implanted + subsequent Au ³⁺ post irradiation 700 °C (10 dpa) [29]	He ⁺ implanted + subsequent Au ³⁺ post irradiation 700 °C (20 dpa)	He ⁺ implanted + subsequent Au ³⁺ post irradiation 700 °C (30 dpa)
	Bubble averaged size (nm)		
1 × 10 ¹⁵	1.7 ± 0.5	1.7 ± 0.4	1.7 ± 0.4
3 × 10 ¹⁵	1.7 ± 0.9	1.8 ± 0.4	1.8 ± 0.4
1 × 10 ¹⁶	2.4 ± 1.0	Unavailable	2.4 ± 0.7

bubble distribution along the grain boundaries in the NE SiC, determination of the volume swelling is not straightforward; the estimated bubble volume swelling from average bubble size and density is less than 0.05%. The average diameters and densities of

helium bubbles are summarized in Tables 1 and 2. As is shown in Tables 1 and 2, the divergence of the bubble diameter is reduced as the damage level is increased; however, the average bubble size is relatively invariant with dose, suggesting that bubble growth is

Table 2

Helium bubble number densities under different heavy ion irradiation doses.

He fluence (ions cm ⁻²)	He ⁺ implanted + subsequent Au ³⁺ post irradiation 700 °C (10 dpa) [29]	He ⁺ implanted + subsequent Au ³⁺ post irradiation 700 °C (20 dpa)	He ⁺ implanted + subsequent Au ³⁺ post irradiation 700 °C (30 dpa)
	Number density (10 ²² m ⁻³)		
1 × 10 ¹⁵	0.7	4.45 ± 1.1	7.11 ± 1.7
3 × 10 ¹⁵	0.66	3.69 ± 0.8	4.16 ± 1.0
1 × 10 ¹⁶	0.58	Unavailable	2.77 ± 0.7

inhibited over this dose range. For each implanted helium concentration, the bubble density is observed to increase with irradiation dose, which suggests that nucleation is an ongoing process at 700 °C under heavy ion-irradiation in this dose regime. For a given irradiation dose, the bubble density decreases with increasing helium concentration, except at the lowest irradiation dose (10 dpa). These results on bubble size and density suggest that nucleation processes are dominant in this dose range at 700 °C, and the size of stable bubble nuclei increases only slightly with helium concentration.

The number density of helium bubble under various irradiation conditions are summarized in Table 3. The mean helium bubble diameter in NE SiC is lower than other values reported in the literature [39–45]. It's also worth mentioning that those studies in the literature were investigating bubble formation at higher temperatures and helium doses.

It is proposed that the formation of vacancies under heavy ion irradiation can immobilize the helium. Those irradiation induced vacancy clusters can trap helium and result in the nucleation of helium bubbles. Although bubbles with larger diameters preferentially form along grain boundaries, the concentration gradient (denuded zone) does not exist in NE SiC due to the lack of defect sinks (i. e. carbon interface) at grain boundaries.

We observe that in the low dose irradiated specimen (10 dpa, 1 × 10¹⁶ helium ions cm⁻²), smaller helium bubbles with an average diameter of 1.7 nm are randomly distributed within the grain; whereas, larger bubbles with an average diameter of 2.6 nm are formed along the grain boundaries. This location-dependent size divergence is not observed after higher dose irradiations.

Continuous evolution of helium bubbles after irradiation to 30 dpa leads to a slight increase in average helium bubble size to 2.3 nm within the grains and a slightly shrinkage to 2.5 nm at grain boundaries. No obvious helium bubbles are observed outside the helium-implanted region, neither near the surface nor at the film/substrate interface, which supports that radiation-induced helium migration is limited perpendicular to the SF planes but readily occurs two-dimensionally parallel to the SFs. Furthermore, it is important to note that no dislocation loops are observed in the NE SiC irradiated at 700 °C to 30 dpa, which is in marked contrast to observations in CVD SiC irradiated at similar temperatures [29].

4. Discussion

Studies of helium behavior in SiC single crystals (4H and 6H) following room temperature implantation and annealing have indicated that bubbles, platelets or even planar clusters are observed to form only at higher temperature region (mostly higher than 800 °C) or above a certain threshold helium concentration (i.e. 600 appm at 1427 °C) [46,47]. The threshold helium concentration for bubble formation in 3C SiC has also been reported in previous studies [26] (i.e. 17000 appm at room temperature and 20000 appm at 650 °C). According to the reported threshold concentration value for bubble formation in 3C SiC, our observation of no resolvable bubble formation in the helium implanted NE SiC (2400 appm at peak), after 2 h of annealing at 700 °C without irradiation, is in good agreement with previous results [29] that indicate no gas bubbles or cavities with measurable size could be resolved in the helium-implanted specimen.

Table 3Summary of density and diameter of bubble of NE SiC and SiC_f/SiC composite irradiated from 10 dpa to 100 dpa at 700 °C–1300 °C.

Conditions				Mean size (nm)		Correspond density (10 ⁻²² m ³)	
NE SiC	8000–800 He appm/	10 dpa	700 °C	1.7–2.4		0.7–0.6	
		30 dpa		1.7–2.4		5.4–2.1	
				Matrix	Fiber	Matrix	Fiber
SiC _f /SiC composite	Dual beam	10 dpa	800 °C	1.2	–	0.85	–
	1300 He appm/*Keng et al. [43]						
	Dual beam	100 dpa	800 °C–1200 °C	9	5	0.26	0.45
	13000 He appm/*Keng et al. [43]						
	Dual beam	10 dpa	800 °C–1200 °C	3.7–21.9	0–3.8	1–6	–
	1300 He appm/*Miwa et al. [56]						
	Dual beam		1000 °C–1300 °C	4.9–23.2	–	1.61–5.92	–
	1300 He appm/						
	* Taguchi et al. [57]		800 °C–1000 °C	4.4–4.6	0–2.7	0.9–3.3	–
	Triple beam						
	1300 He appm/400H appm		900 °C–1200 °C	2.2–2.8	1.3–2.1	6.1–2.1	1.2–7.1
	* Hasegawa et al. [58]						
	Triple beam		1000 °C–1300 °C	4.6–20.9	–	2.51–7.22	–
	1300 He appm/400H appm						
	* Chen et al. [59]						
	Triple beam						
	1300 He appm/400H appm						
	* Taguchi et al. [57]						

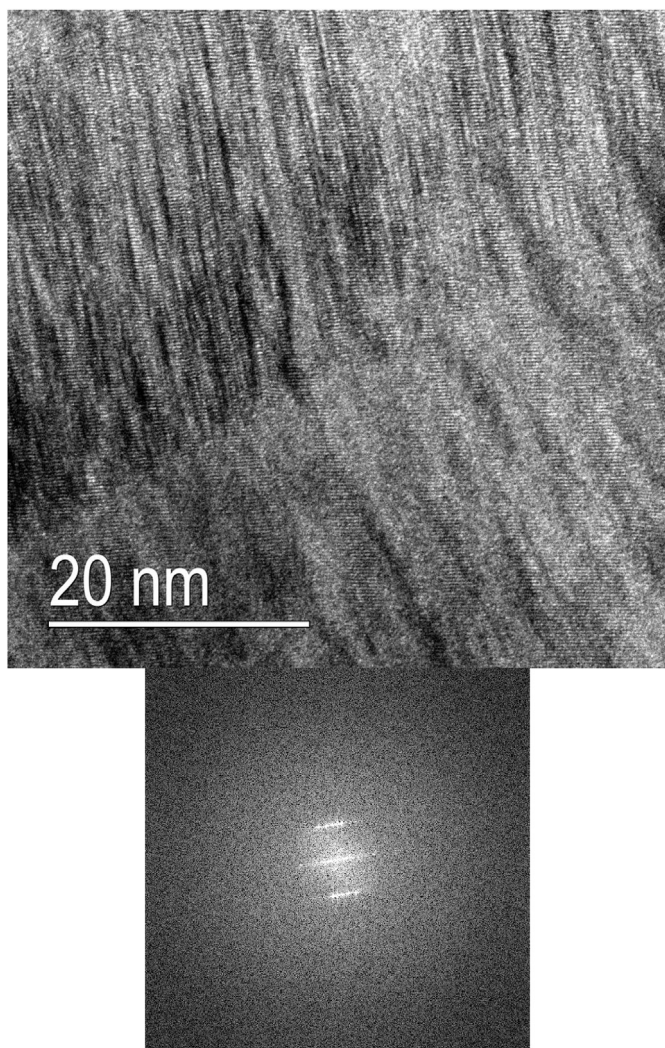


Fig. 6. Cross-sectional high resolution TEM micrograph (acquired at 200 kV) for NE SiC after Au^{3+} ions irradiation to 30 dpa at 700 °C with He fluence of 1×10^{16} ions cm^{-2} (micrograph was taken at the depth range of 350 nm from the surface).

We have investigated phase stability and helium bubble formation for an irradiation dose of 10 dpa at 700 °C previously [29] and doses of 20 dpa and 30 dpa at 700 °C in the present study. The fact that the SF structure remained after irradiation is demonstrated from the HRTEM image and FFT pattern, as illustrated in Fig. 6. This result indicates that the crystallinity and SF structure exhibit great irradiation resistance at high temperature to irradiation up to 30 dpa. For helium implanted NE SiC, helium bubble formation at 700 °C only occurs under subsequent heavy-ion irradiation. Interestingly, dislocation loops formation in NE SiC seems suppressed under irradiation at 700 °C to 30 dpa, which may inhibit bubble growth by eliminating biased sinks for interstitials.

It is worth noting that TEM examination of a specimen from a masked region, which was subject to 2 h of thermal annealing at 700 °C, revealed no evidence of bubble formation, consistent with our earlier findings [29] after 1 h of annealing at 700 °C.

4.1. Helium diffusion associated with SF confinement

A previous study [25] has shown an increase in threshold amorphization dose for NE SiC compared to single crystal SiC. The

accumulation of irradiation-induced defects can be significantly reduced due to the presence of many SFs, grain boundaries and also the grain texture. Defects created from nuclear collision cascades are confined between the SFs and annihilated at grain boundaries or by recombination within the SFs. Based on DFT calculations, the presence of SFs makes interstitial defects more mobile parallel to the SFs and decreases the binding energy of interstitial-antisite defects, both of which suppress or delay defect accumulation [25,48]. The Si interstitial annihilation at grain boundaries and the Si antisite removal phenomenon are also enhanced due to the presence of SFs, thus enabling defect migration and defect interactions in NE SiC. In our study, the ToF-ERDA results demonstrate that helium diffusion normal to the surface is negligible under irradiation at 700 °C. The increased nucleation of bubbles at grain boundaries suggests enhanced helium and defect migration parallel to the stacking faults, but not across the stacking faults (normal to the surface). Therefore, both the enhanced radiation tolerance (suppressed dislocation loop formation) and helium confinement in the NE SiC film under ion-irradiation at 700 °C to 30 dpa are attributed to the high density of SFs.

4.2. Helium bubble formation

It has been reported that helium migration can be significantly enhanced as temperature increases due to the increased mobility of defects [42,49,50], especially when vacancies become mobile. In the temperature range for interstitial helium migration, from 750 to 1060 °C, the helium diffusion coefficient is reported to be given by $D \text{ (cm}^2/\text{s)} = 1.38 \times 10^{-10} \exp\{-0.91 \pm 0.07 \text{ (eV/atom)}/kT\}$ [51]. Thus, increasing the temperature from 750 °C to 1000 °C results in an order of magnitude increase in helium diffusion. However, once the vacancies become mobile at high temperatures, helium release in single crystal SiC occurs, resulting in up to 95% helium release at 1300 °C [49].

In this present study, the incident helium ions are deposited in the nano-grains and SF layers, and produce some defects within the low-energy recoil cascades. Although the implanted helium atoms can be thermally activated to migrate, it can only occur at very high temperature (above 1000 °C, [52]). At 700 °C, any residual defects and implanted helium atoms have limited diffusion, resulting in no observable bubbles. It was reported by Duh et al. [42] and Miro et al. [53], that only a small portion of helium-vacancy pairs (vacancy trapped helium atom) can become mobile as temperature exceeds 800–1100 °C. This suggests that, at the temperature of our experiments (700 °C), helium diffusion in NE SiC is assumed to be dominated by interstitial diffusion.

During the high temperature heavy ion irradiation, the collision cascades provide not only kinetic energy transfer to helium atoms, both on the lattice and within bubbles, but also create irradiation-induced defects. It has been suggested that the formation of gas bubbles in SiC is strongly associated with defects and dislocations [47,48]. In the present study, the density of helium bubbles is found to increase with irradiation dose (Table 2); however, the helium bubble size increases only slightly with implanted helium concentration, and is independent of irradiation dose for a given helium concentration, as shown in Table 1. Hence, it is found that defect and helium migrations and interaction dynamics are dominated by nucleation processes [19].

During irradiation, both the interstitials and helium atoms are mobile at this temperature. Thus, more visible helium bubbles are observed within the grains as the irradiation dose increases. Although the irradiation dose is as high as 30 dpa, the helium bubbles did not grow to any noticeable extent. On the other hand, the helium bubble density increased significantly, from 4 to 10 times, as the irradiation dose increased (i.e. see Tables 1 and 2). This

suggests that the energy barrier for defect migration is not altered within the grains or at the SFs after high heavy ion doses. The lack of significant bubble growth with dose up to 30 dpa suggests bubbles are undergoing continuous growth and shrinkage during the nucleation process that leads to a long incubation dose prior to the onset of bubble growth. This may be due to the high mobility of Si and C interstitials and the lack of dislocation loop formation. Significant bubble growth and swelling may in fact be suppressed in this structure because of the lack of dislocation loops to provide a biased sink for interstitials.

4.3. Bubble sizes and densities

A comparison of bubble size and density with literature data is summarized in Table 3. As reported in previous studies, hydrogen enhances bubble nucleation and inhibits bubble growth in SiC, which is validated when comparing with dual and triple ion beam irradiation results. At 700 °C, bubbles were found to have a smaller average size in NE SiC than in SiC composites, although the number densities are comparable. At 800 °C for SiC composites, as damage increases from 10 to 100 dpa, bubble growth can be observed. Dramatic bubble coalescing into larger size bubbles can only be found once the irradiation temperature exceeds 1000 °C, which can also lead to a decrease in the number density.

Normalized bubble size distributions in the NE SiC after irradiation are determined and summarized in Fig. 7. Bubble sizes below 1 nm are included as a dashed line due to the limitation and uncertainty of the TEM observations. For the specimens with low helium fluence (1×10^{15} ions cm^{-2}), the density of helium bubbles increased significantly as the irradiation dose increased from 10 to 30 dpa; however, the helium bubbles exhibit similar size

distributions but with decreased divergence in size with increasing dose. For the highest implanted helium concentration (1×10^{16} ions cm^{-2}), a clear transition in the bubble size distribution is observed with increasing irradiation dose. A bimodal size distribution is clearly observed for irradiation to 10 dpa, similar to that observed in Be-doped SiC irradiated with triple ion beams [54]. However, increasing the dose to 30 dpa results in a decrease in the divergence of the bubble size distribution, and the bimodal distribution transforms into a Gaussian distribution with a similar average bubble size. This may be due in part to the shrinkage of some larger bubbles with increasing dose, due to helium ejection from the bubbles by kinetic energy transfers from recoils, which is consistent with a previous study by Pawley et al. [55] who found that bubble growth can be inhibited by displacive irradiation in 4H SiC. Compared to the helium-implanted sample with thermal ramp alone, the present results show evidence that the bubble growth rate is indeed inhibited, which can be attributed to the displacement of helium out of the bubbles by the collision cascades from incident Au ions.

For the specimens implanted to the medium helium concentration (3×10^{15} ions cm^{-2}), the larger bubble sizes at 10 dpa disappear, and there is a similar size distribution at 20 and 30 dpa, with an average bubble size of about 1.7 nm. There is a consistent loss of larger bubble sizes with increasing dose for each implanted helium concentration, suggesting that an equilibrium size distribution eventually forms at higher doses.

Although the free surface and film/substrate interface can act as sinks for helium, in this study, helium bubbles with measurable size are only observed in a depth range from 270 to 390 nm from the surface, which is consistent with the helium implantation profile with the helium concentration peak at 320 nm. This suggests that

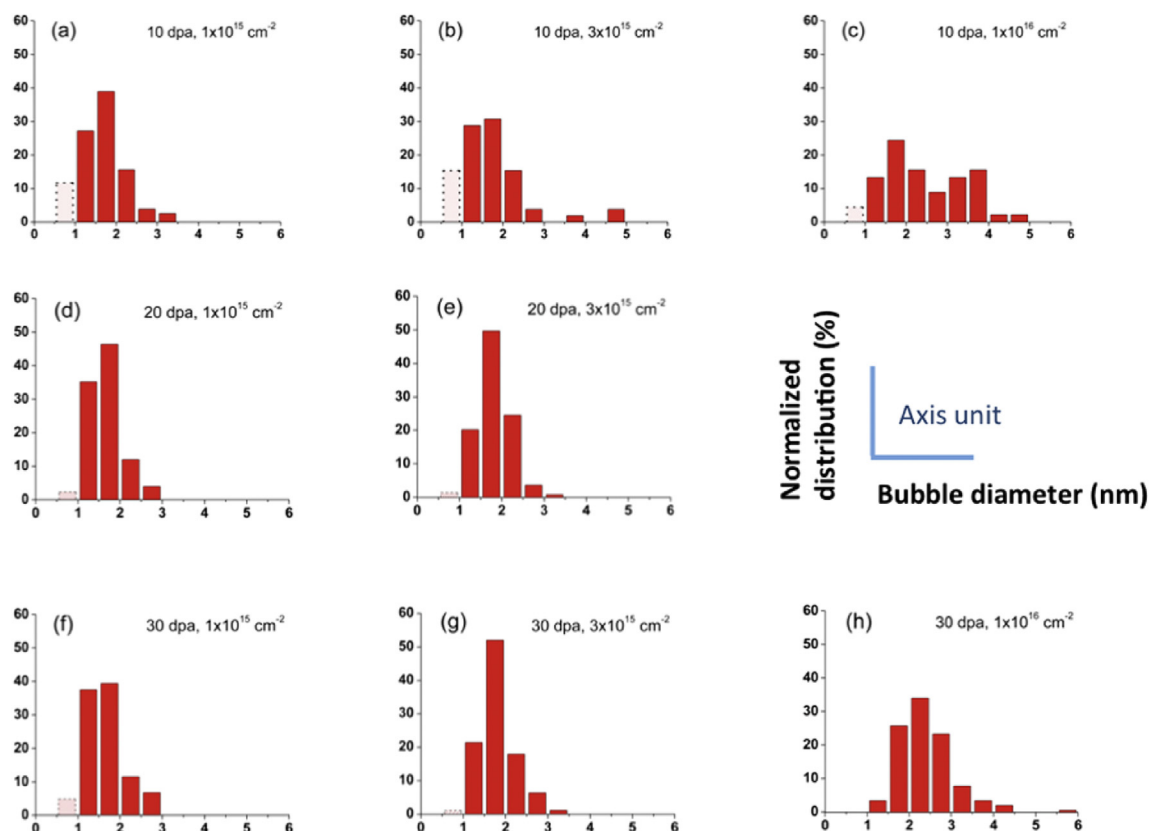


Fig. 7. Normalized bubble size distribution in NE SiC after irradiation from 10 to 30 dpa at 700 °C with helium fluence of 1×10^{15} ions cm^{-2} , 3×10^{15} ions cm^{-2} , and 1×10^{16} ions cm^{-2} , respectively.

helium atom diffusion primarily occurs two-dimensionally and over a short range in the NE SiC under the current irradiation temperature. The SFs exhibit significant radiation tolerance by retaining their self-layered interface structure after heavy ion irradiation to 30 dpa at 700 °C. The confinement of the helium in the NE SiC film under ion-irradiation to 30 dpa may be attributed to the high density SFs, which modified the migration pathways for the helium atoms into 2D-like interlayer diffusion. Since significant bubble growth is not observed outside the predicted helium implantation profile, further studies of higher helium concentrations, higher radiation doses and higher temperatures are needed for deep understanding the role of defect concentration in helium migration.

5. Conclusion

Helium bubble formation is observed in a well-defined depth region around the helium concentration peak after Au³⁺ ion irradiation to dose from 10 to 30 dpa. This subsequent irradiation of the helium implanted samples with Au ions revealed preferential formation of bubbles at grain boundaries, which indicates that helium migration is limited by two-dimensional diffusion. The inhibited helium migration perpendicular to the stacking fault direction [111] is validated from the ToF-ERDA measurement. A long bubble nucleation period with increasing dose is observed, where the bubble size remains in dynamic equilibrium and below the critical bubble size for growth, which results in a large incubation dose. Significant bubble growth and swelling may be inhibited by the suppression of dislocation loop formation in the NE SiC. The main features of the microstructures can be understood in terms of radiation-induced defects and helium migration associated with SFs confinement in this temperature region. The microstructural observations also reveal that the nano-layered SF structure in the NE SiC is highly radiation tolerant and stable under irradiation to a dose of 30 dpa at 700 °C.

Acknowledgments

This work was supported by the U.S. Department of Energy, Nuclear Energy University Programs and the University of Tennessee Governor's Chair program. Heavy ion irradiation and ion beam analysis were performed at the University of Tennessee–Oak Ridge National Laboratory Ion Beam Materials Laboratory (IBML) located at the campus of the University of Tennessee, Knoxville.

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