Pawley, C J, Beaufort, M F, Oliviero, E, Hinks, J. A., Barbot, J F and Donnelly, S. E.

An in-situ TEM investigation of He bubble evolution in SiC

An *in-situ* TEM investigation of He bubble evolution in SiC

C J Pawley†[1,2], M F Beaufort[3], E Oliviero[4], J A Hinks[2], J F Barbot[3] and S E Donnelly[2]

[1] Centre for Materials and Physics, School of Computing, Science and Engineering, University of Salford, Salford M5 4WT, UK.

[2] Electron Microscopy and Materials Analysis Group, School of Computing and Engineering, University of Huddersfield, Huddersfield HD1 3DH, UK.

[3] Institut Pprime, Université de Poitiers, 86962 Futuroscope-Chasseneuil, France.

[4] Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, CNRS-IN2P3, Université Paris-Sud, Orsay Campus, France.

†Email: c.pawley@edu.salford.ac.uk

Abstract This paper presents work using the capabilities of two TEM with *in-situ* ion irradiation facilities: Microscope and Ion Accelerator for Materials Investigation (MIAMI) at the University of Huddersfield and Joint Accelerators for Nano-science and Nuclear Simulation JANNuS at Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse (CSNSM), Orsay, France, to study the nucleation and growth of He bubbles in silicon carbide (SiC) and to carry out an investigation into bubble behaviour at high temperatures and under displacing irradiation. Preliminary results on bubble nucleation and growth during He irradiation of SiC are presented together with results from a simultaneous anneal and high-energy heavy-ion irradiation of samples containing He bubbles. The displacing irradiation is observed to impede He bubble growth resulting in smaller bubbles than those obtained from an anneal alone. A tentative interpretation of these observations is presented.

1. Introduction

Silicon carbide (SiC) is a candidate material for fusion reactor applications [1]. Current designs use SiC in plasma-facing components in fusion reactors because of its high-temperature stability and radiation hardness. Additionally it is attractive because it has the potential to be cost-effective and is suitable for structural applications. Other advantages include resistance to corrosion and to thermal fatigue and limited saturation swelling.

Expected conditions within a fusion reactor will give rise to significant material challenges. Materials within the reactor will be subject to irradiation by light elements through transmutation reactions, whilst being subjected to a flux of energetic neutrons which will cause significant displacements to the atomic structure of the SiC. Data from Zinkle [2] suggest that reactor lifetime concentrations of He will be of order 1 atomic-% with maximum neutron-caused damage levels of 150 displacements per atom (dpa). Operating temperatures are expected to be between 550 and 1000ºC.

Whilst a significant amount of scientific research has focused on the performance of SiC in nuclear environments, there are still some critical questions (outlined by Riccardi [3]) that remain unanswered. We aim to address one of these in the current project – specifically the question of material stability under the combined effects of high temperature, He accumulation and displacing irradiation.

2. Experimental

Silicon carbide wafers of the 4H polytype were acquired from Cree Inc. The wafers were 3″ in diameter and were grown 8º off-axis from the [0001] direction. Samples were cut from the wafer using a diamond-wire saw and prepared for transmission electron microscopy (TEM) using the tripod method [4] and ion-beam thinning using a Gatan Model 691 PIPS with 4 keV Ar^+ ions incident at 4° to the sample surface. Samples were then irradiated within the Microscope and Ion Accelerator for Materials Investigations (MIAMI) facility at the University of Salford, UK (now at the University of Huddersfield), the details of which are documented elsewhere [5]. Samples were irradiated by 3.5 keV He^+ ions with a flux of approximately 5×10^{13} ions/cm^2/s to a final fluence of between 8×10^{16} ions/cm^2 and 10^{17} ions/cm^2. Calculations using the Monte-Carlo code SRIM [6] indicate that 3.5 keV He^+ ions have an average range of 25 nm in SiC – approximately half the thickness of the samples. The angle between the ion and electron beams was 25°. In order to improve bubble image quality, samples were tilted slightly to avoid aligning low-index directions with the electron beam, thus avoiding strong Bragg reflections. Bright-field off-zone-axis TEM micrographs were then taken using a JEOL JEM-2000FX operated at 200keV after each irradiation step to record the changes occurring within the sample. Irradiation and microscopy were performed at a constant temperature of 700ºC.

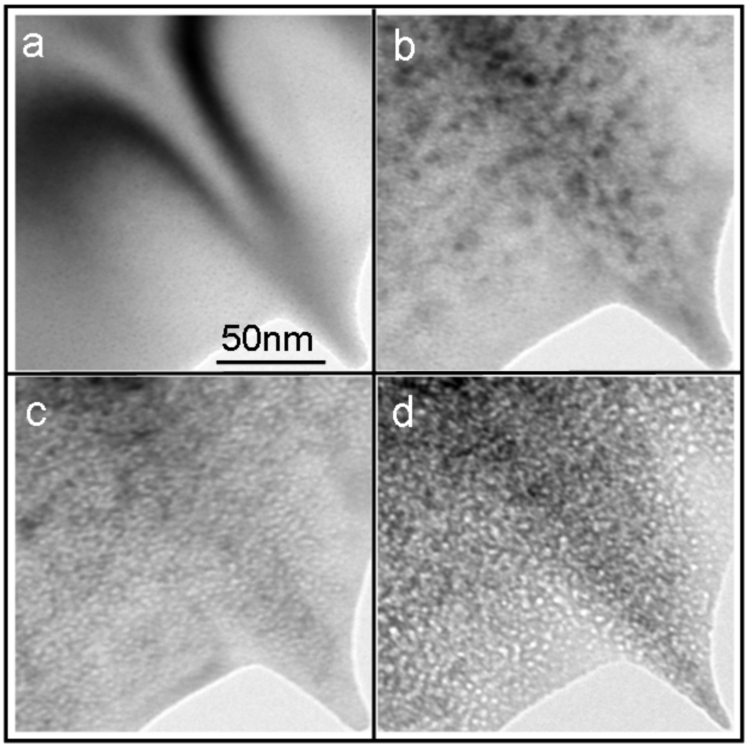

Figure 1. TEM off-zone-axis bright-field micrographs, 800 nm underfocus, of SiC irradiated with 5×10^{13} He ions/cm^2/s at 700°C, showing bubble distributions at fluences of: (a) 0; (b) 1.1×10^{16}; (c) 3.4×10^{16}; and (d) 10^{17} ions/cm^2. Scale marker applies to all panels.

Bubble-containing samples were irradiated during *in-situ* experiments at the Joint Accelerators for Nano-science and Nuclear Simulation (JANNuS) [7] facility in CSNSM, Orsay, France using 4 MeV Au^{2+} at a flux of 2.5×10^{11} ions/cm^2/s to a fluence of 2×10^{15} ions/cm^2. Samples were subjected to a temperature ramp from 700 to 1300ºC at the same time as the irradiation. The angle between the ion and electron beams was 68º. Underfocussed bright-field TEM micrographs were recorded using a Technai G^2 20 operated at 200keV. At both MIAMI and JANNuS images were captured using 4 megapixel Gatan Orius SC200 digital cameras.

3. Results and Discussion

3.1. Bubble nucleation and growth during 3.5 keV He irradiation

Figure 1 shows a region of the sample before irradiation (Figure 1a) and following irradiation at 700°C to three increasingly high fluences. The dark mottled contrast in Figure 1b is typical of that due to interstitial clusters and the figure also shows contrast typical of small nanometer-sized bubbles (small white areas in this underfocused image). At the higher fluence in Figure 1c, the bubbles are now clearly visible and the interstitial contrast is less evident. Finally, Figure 1d shows a bubble distribution with a wider range of sizes.

Analysis of the images sequenced into a video provide a strong indication that, once nucleated, bubbles remain on fixed sites (no diffusive motion is observed) and continue to grow by accumulation of injected He and vacancies from the concomitant radiation damage. They eventually grow to a size where contact with a neighboring bubble may occur – at which point coalescence is observed followed by a change in the resulting elongated bubble to an assumed spherical shape (circular in projection). The coalescence gives rise to the wider range of bubble sizes visible in Figure 1d.

3.2. Effects of displacing irradiation and high temperature on He bubble distribution

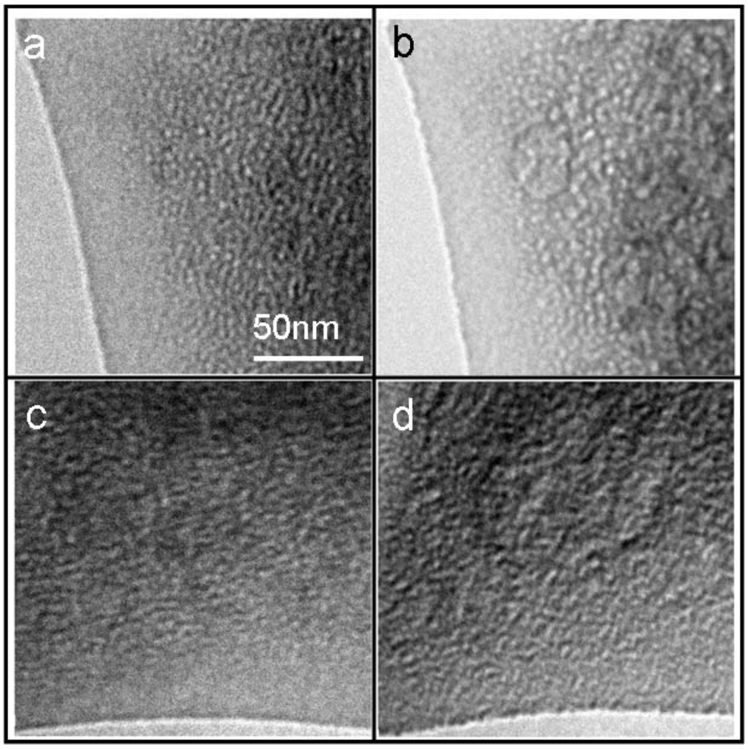

Figure 2. Bright-field micrographs of sample subjected to temperature ramp only (a and b) and subjected to identical temperature ramp and 4 MeV Au ion irradiation (flux: 2.5×10^{11} ions/cm^2/s. Images recorded at (a) 810 ºC; (b) 1045 ºC; (c) 770ºC/1.75×10^{12} ions/cm^2 and (d) 1050ºC/9.5×10^{14} ions/cm^2. Scale marker applies to all panels.

The images in the two left panels of Figure 2 show SiC containing He bubbles following irradiation to a fluence of 8×10^{16} ions/cm^2 as detailed in Section 2. Figure 2b shows the bubble morphology following a temperature ramp of 10°C/minute to a temperature of 1045°C. Figure 2d shows the bubble morphology following an identical temperature ramp to 1050°C with simultaneous irradiation with 4 MeV Au^{2+} ions. Inspection of the image reveals a mean bubble size significantly smaller than in Figure 2b. Specifically, the mean bubble radius in Figure 2d is 1.2 nm (1 nm at 700°C) compared with 1.5 nm (0.9 nm at 700°C) in Figure 2b. The mean radius of bubbles subjected to Au^{2+} irradiation during the temperature ramp grew by only 19% whereas those subjected to the temperature ramp alone exhibited a 60% increase in radius. In previous work on heavy-ion irradiation of He bubbles in Al

Birtcher *et al* [8] concluded that the relatively-dilute displacement cascades that occurred in the low-Z substrate resulted in a gradual displacement of He from the bubbles back into the lattice where it was able to diffuse and escape. The continuous production of Frenkel defects during irradiation allowed the bubbles to re-equilibrate by loss of vacancies resulting in a net reduction in mean bubble diameter. The reported shrinkage rates were 0.02 to 0.05 nm per dpa. In the current work, we measured a slightly lower mean shrinkage rate of 0.012 nm per dpa. Detailed modelling of this process is underway, but our tentative conclusion is that a similar process to that observed by Birtcher *et al* is operating in the He/SiC system in which similar dilute cascades occur to those in Al.

In the present work, measurements of bubble size distributions were rendered difficult at high Au fluences and high substrate temperatures by surface restructuring that occurred under these conditions. Future runs are scheduled at JANNuS in which similar experiments will be carried out on a population of larger, more clearly delineated bubbles and under conditions in which surface restructuring will be minimised.

4. Conclusions

4H-SiC TEM samples have been irradiated by 3.5 keV He^+ ions at a temperature of 700ºC in order to investigate He bubble nucleation and growth. Aided by the *in-situ* nature of the experiments, we are able to conclude that bubble motion is minimal – final positions of bubbles are determined by the locations on which bubbles nucleate (or at least above the size at which they become resolvable by TEM). Bubbles subsequently grow until they overlap and coalesce. The mean bubble radius following irradiation to a fluence of 10^{17} ions/cm^2 was approximately 0.9 nm.

He bubbles in SiC were then subjected to a simultaneous temperature ramp and displacing irradiation in the form of 4 MeV Au^{2+} ions in a second set of *in-situ* experiments. Bubble growth was observed to be inhibited by the displacing irradiation as evidenced by reduced enlargement compared to the thermal ramp alone. The reduced growth rate is attributed to the displacement of He out of the bubbles by the collision cascades induced by the incident Au ions.

Acknowledgments

The authors wish to acknowledge the support of the Engineering and Physical Sciences Research Council (EPSRC) for providing the funding to construct the MIAMI facility (grant EP/E017266/1). This work is supported by the French network EMIR. The work was facilitated by a Royal Society / Centre National de la Recherche Scientifique (CNRS) International Joint Project grant between the Institut Pprime (Poitiers) and Centre for Materials and Physics (Salford). C.J.Pawley wishes to thank the University of Salford for partial funding of this research under the Graduate Teaching scheme.

References

[1] Najmabadi F, Conn R W, Bathka C G, Bromberg L and Cheng E T 1991 *Fusion Technol* **19** 783–790

[2] Zinkle S J 2005 *Phys Plasmas* **12** 058101-1 – 058101-8

[3] Riccardi B, Giancarli L, Hasegawa A, Katoh Y, Kohyama A, Jones R H and Snead L L 2004 *J Nucl Mater* **329-333(Part 1)** 56–65

[4] Klepeis S J, Benedict J P and Anderson R M 1987 *Specimen preparation for transmission electron microscopy: Proc. Mater Res Soc symp* vol 115 ed J C Bravman, R M Anderson and M L McDonald (Cambridge, UK: Cambridge Uni Press) pp179–184

[5] Hinks J A, van den Berg J A and Donnelly S E 2011 *J Vac Sci Technol A* **29(2)** 021003–6

[6] Ziegler J F 2004 *Nucl Instrum Meth B* **219-220** 1027–1036

[7] Serruys Y, Ruault M O, Trocellier P, Henry S, Katasov O and Trouslard P 2005 *Nucl Instrum Meth B* **240(1-2)** 124–7

[8] Birtcher R C, Donnelly S E, Templier C 1994 *Phys Rev B* **50** 764–9