Journal of Nuclear Materials 471 (2016) 149–153

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

In situ Observation of Microstructure Evolution in 4H–SiC under 3.5 keV He^+ Irradiation

Qiang Shen [a, b], Guang Ran [a, *], Jonathan Hinks [b, **], Stephen E. Donnelly [b], Lumin Wang [a], Ning Li [a]

[a] *College of Energy, Xiamen University, Xiamen, Fujian 361102, China*
[b] *School of Computing and Engineering, University of Huddersfield, Huddersfield HD1 3DH, United Kingdom*

ARTICLE INFO

Article history:
Received 3 July 2015
Received in revised form
16 January 2016
Accepted 19 January 2016
Available online 21 January 2016

Keywords:
SiC
In situ TEM
In situ irradiation
Bubble disc
Microstructure

ABSTRACT

4H–SiC was irradiated with 3.5 keV He^+ ions using the MIAMI facility at University of Huddersfield. The evolution of microstructure and gas bubbles during the irradiation at 700 °C, 800 °C and 900 °C was observed by *in situ* transmission electron microscopy. Under irradiation, isolated bubbles and bubble discs formed in the SiC matrix. Bubble discs lying on {0001} and {10-10} crystal planes were beginning to form at ion fluence above 2.3×10^{20} He^+/m^2 at 700 °C. The density of bubble discs increased with increasing irradiation fluence. However, growth rates were different at different of the implantation periods and temperature holding periods. The nucleation and growth of the bubble discs were attributed to be coalescence of the adjacent He vacancies and combination of loop punching and trap mutation, respectively.

1. Introduction

Silicon carbide is considered as a promising candidate material used in nuclear energy due to its superior irradiation tolerance, high thermal conductivity, low thermal expansion, good thermal shock resistance and chemical inertness [1,2]. One of the most important applications is that SiC can be used as a structural layer in Tri-structural isotropic (TRISO) particles which have been used as nuclear fuel in high temperature gas-cooled reactors for several decades [3,4]. At present, TRISO particle fuels are also considered to be used in fluoride salt high temperature reactors and light water reactors [2,5]. After the catastrophic events at the Fukushima Daiichi nuclear reactors, ceramic accident tolerant fuel (ATF) systems utilizing SiC-based cladding have been under development as a potential replacement for zircaloy due to significantly low exothermic reaction with steam and their ability to retain a stable core geometry to very high temperature [6,7]. In the newly designed fully ceramic micro-encapsulated fuel (FCM) system, the TRISO particles are embedded in an additional fission product barrier of SiC matrix thus providing enhanced safety and superior irradiation resistance as compared to the traditional TRISO compact as well as UO_2 fuels [5,8]. SiC has been also proposed as a first wall material in fusion reactor [9,10]. Helium is inevitably introduced into the first wall material by energetic helium bombardment from the plasma and (*D*,*T*) reactions [11]. Helium atoms gather to form bubbles or bubble discs that may hasten the degradation of SiC-based materials.

Effects of helium irradiation on SiC have been widely investigated. Zhang et al. studied the helium fluence dependence of nanoscale cavity formation in 4H–SiC [12]. Helium bubbles were formed in 4H–SiC when helium was implanted at the room temperature, 227 °C and 600 °C. Planar bubble clusters were found when these He-implanted samples were annealed at temperature above 700 °C. Hojou reported the helium bubbles were likely to be formed along the basal plane of hexagonal SiC [13]. In Chen's work, He^+ implantation of hot-pressed SiC at ambient temperature resulted in the formation of helium platelets lying on {0001} planes [14,15]. Transformation from the platelet-shaped cavities into bubble discs was reported when irradiated samples were annealed at 1227 °C. Bubble-loop complexes were formed when annealed temperature at 1427 °C. Li et al. observed bubble discs associated with dislocation loops and stacking faults inside grains after post-

---

* Corresponding author.
** Corresponding author.
E-mail addresses: gran@xmu.edu.cn (G. Ran), j.a.hinks@hud.ac.uk (J. Hinks).

http://dx.doi.org/10.1016/j.jnucmat.2016.01.017

irradiation annealing at 1000 °C [16]. Texier et al. found the large cavities lying on {0001} and {10-10} are formed and experienced on transformation when the He irradiated 4H–SiC were annealed at 1700 °C [17].

These large amount of *ex situ* experiment results of helium irradiation SiC are important for understanding of irradiation effects on SiC. However, these experiments cannot provide and display the temporally and spatially microstructure variation that are very important for the underline fundamental irradiation effect. Based on the characteristics of online facilities of accelerator and TEM, *in situ* observation was used to understand the evolution of microstructure. Pawley reported the evolution of bubbles in 4H–SiC by helium implantation at 700 °C [18]. Hojou's *in situ* experiment demonstrated that the pre-implantation of hydrogen could enhance the formation and growth of helium bubbles in SiC [19]. Those studies provided rich microstructure evolution behaviors of SiC under ion irradiation. However, there are still some questions that need to be addressed, such as, the mechanism of the nucleation and growth of bubble discs. In the present work, the microstructure evolution behaviors of 4H–SiC by helium irradiation are investigated by *in situ* TEM in the temperature range of 700°C–900 °C.

2. Experiment

4H–SiC single crystal from Cree Corporation, USA, was used in the present work. The specimen surface is 7.88° off the (0001) plane. Both sides of each specimen were polished by diamond sand paper. After thinned to 10–15 μm, specimen was glued on a molybdenum grid by G-1 epoxy glue. Then, specimen was thinned to about 100 nm thickness by Ar ion milling using a Precision Ion Polishing System.

Helium ions were implanted into the 4H–SiC specimen at 700 °C, 800 °C and 900 °C using the MIAMI facility at University of Huddersfield, UK [20]. The microstructure and gas bubble evolution was monitored *in situ* during helium ion irradiation using a 200 keV electron beam. The ion flux and total ion fluence were 2.5×10^{17} $He^+/(m^2 \cdot s)$ and 9.1×10^{20} He^+/m^2, respectively. The incident angle of helium ion was 30° off the normal direction of specimen surface. The ion energy was designed as 3.5 keV by Monte Carlo calculation using the Stopping and Range of Ions in Matter (SRIM) software. The displacement energy of Si and C was assumed to be 35 eV and 20 eV, respectively. The depth of helium ion concentration peak and displacement damage peak are about 35 nm and 15 nm, respectively. The implantation helium concentration peak is about 26 at% and peak displacement damage is 15.5 dpa after a fluence of 9.1×10^{20} He^+/m^2.

3. Results and discussion

Fig. 1 shows under-focused TEM BF images of the 4H–SiC specimens irradiated by 6.1×10^{20} He^+/m^2 fluence at (a) 700 °C, (b) 800 °C, and (c) 900 °C. Specimens are viewed along the [0001] crystal direction. It can be seen that lots of isolated bubbles distribute in the SiC matrix at 700 °C, 800 °C and 900 °C. However, a high density of bubble discs are observed at 800 °C and 900 °C with few at 700 °C. Fringes with bright and dark contrast surrounding the bubble discs can be seen in the images. All TEM images in this work were taken at the same under-focused condition of 3.0 μm.

The result is in good agreement with Zhang's ex situ experiment, where bubbles discs were formed when the specimens were annealed above 700 °C with a helium fluence of 2.5×10^{20} He^+/m^2 [21]. It is speculated that a temperature threshold around 700 °C may be the essential for the formation of bubble discs in 4H–SiC. Chen's post-implantation annealing experiments detected the formation of He platelet-shaped cavities at room temperature in polycrystalline SiC. Then helium platelets transformed to bubble discs after the sample had been annealed up to 1227 °C [15]. However, in the present work, the transformation phenomenon was not observed. This may be due to the instantaneous transformation and the subtle contrast change between He platelets and bubble discs. The nucleation of platelet-shaped cavities at the early stage of the implantation is thought to be necessary for the formation of bubble discs. Both isolated bubbles and bubble discs are formed in the irradiated areas as shown in Fig. 1. Thus, The isolated bubbles are in favor to be formed rather than platelet-shaped bubble discs if no platelets have been formed. Similar evolution behavior has been reported in He-implanted Mo although detailed mechanism remains in questions [22].

With increasing He^+ ion fluence, the bubble discs undergo significant growth at 800 °C as shown in Fig. 2. Three bubble discs as indicated by white rectangles i Fig. 2(a) were monitored during the ion irradiation. The average length of bubble discs increases from 10 nm to 30 nm when ion fluence increases from 2.3×10^{20} ions/m^2 to 6.1×10^{20} ions/m^2. However, the bubble discs almost stop growing after ion fluence up to 6.1×10^{20} ions/m^2. Even though SiC is further irradiated to 9.1×10^{20} ions/m^2 fluence, the average length of bubble discs still remain at 30 nm. The relationship between the size of bubble discs and ion fluence is shown in Fig. 3. The length and width of bubble discs increase significantly from 2.3×10^{20} ions/m^2 to 4.6×10^{20} ions/m^2, and then slow down. The average width is about 3.3 nm after irradiation with 6.1×10^{20} ions/m^2 fluence. During the experiment helium irradiation had been paused at a fluence of 3.1×10^{20} ions/m^2, 3.8×10^{20} ions/m^2 and 4.6×10^{20} ions/m^2 for 6 min, 16 min and 15 min, respectively. The bubble discs still keep growing even though the helium irradiation is stopped. The length of #3 bubble disc increases from 7.5 nm to 17.5 nm at only 6 min pause after irradiation with 3.1×10^{20} ions/m^2 as shown in Fig. 4(a). Correspondingly, the width of #3 bubble disc increases from 1.8 nm to 2.7 nm as shown in Fig. 4(b). The similar growth of other two bubble discs during the implantation and temperature holding periods are observed as shown in Fig. 3 and Fig. 4.

The pressures inside the isolated bubbles and the bubble discs should be balanced during helium irradiation in consideration of the co-existence of the isolated bubbles and the bubble discs in the range of 700–900 °C. Based on the profile of SRIM calculation, almost all implanted helium remain in the specimen. According to Pawley's work [18], less than 10% of implanted helium atoms form bubbles and bubble discs after irradiation with 4.6×10^{20} ions/m^2. Thus, other parts of helium atoms exist as interstitial atoms and invisible helium clusters in SiC matrix. The interstitial helium atoms have high mobility and the vacancies have low mobility in SiC in the range of 700–900 °C [14,15]. Pramono indicated that helium atoms were most likely to diffuse by dissociative/interstitial mechanism in this temperature range [23]. The growth rates of the bubble discs are calculated as shown in Fig. 5. It can be seen that #2 and #3 bubble discs show biggest growth rates under the annealing procedure after irradiation with 3.1×10^{20} ions/m^2. The high ion flux causes serious damage at the beginning of stage. The heat effect provides the recombination of Frankel pairs of Si/C interstitials and Si/C vacancies and the diffusion of helium interstitials. This leads to the high growth rate of bubble discs during the temperature holding time between fluences of 3.1×10^{20} ions/m^2 and 3.8×10^{20} ions/m^2. When the sample was irradiated with a fluence more than 3.8×10^{20} ions/m^2, the growth rates during the implantation periods become relatively bigger than those during the temperature holding periods. The implantation introduced more helium atoms into the irradiated area so that the bubble discs continued to grow.