Journal of Nuclear Materials 386–388 (2009) 199–202

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Capture efficiency for clustering reaction between charged defects in β-SiC

Y. Watanabe *, K. Morishita, A. Kohyama

Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan

ABSTRACT

In order to investigate the formation kinetics of interstitial loops in β-SiC during irradiation, a capture efficiency of a charged point defect to a charged sink was derived taking into account electric interaction between the defects. The derived capture efficiency depends much on effective charges of defects. For defect accumulation in β-SiC under high-energy electron irradiation, the numerical analysis using the rate theory based model with the capture efficiency was performed, and the calculation results indicate that an interstitial loop grows while keeping its charge to be as neutral as possible, leading to the suppression of the growth of interstitial loops. Roughly speaking, the calculated concentration of clustered interstitials with effective charges approaches to the experimental data. It implies that the capture efficiency derived in the present work may play an important role for better reproducing of the experimental result.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

SiC/SiC composite is a candidate for blanket structural materials of nuclear fusion reactor because of good mechanical strength and low activation properties [1]. Point defects and defect clusters such as interstitial loops and voids are produced and accumulated in a material under irradiation, which changes the material microstructure, leading to the degradation of material properties such as mechanical properties, thermal properties and dimensional stabilities. Irradiation effects on SiC materials have been widely studied by a variety of experiments and numerical analysis [2–6]. However, the defect accumulation process including the formation mechanism of defect clusters in SiC is not well understood yet. This is because SiC materials have an ionic nature in addition to a covalent one. It indicates that point defects and defect clusters can have effective electric charges which will influence the diffusion and growth of defects. Ryazanov et al. [7] proposed a model for formation of interstitial loops in β-SiC under irradiation, considering charge state of defects, where an interstitial loop grows and maintains its charge to be neutral, in other words, an interstitial loop grows under the condition of stoichiometric constraint. Interstitial loops treaded in the model are relatively large loops as so-called 'perfect loops'.

In the present work, an applied model for formation of interstitial loops is developed in order to investigate the formation kinetics of interstitial loops in β-SiC. Notice that interstitial loops treated here are relatively small interstitial clusters as so-called 'black spot defects', and such small interstitial clusters in β-SiC are observed in variety of experiments using the high voltage electron microscopy (HVEM), ion-accelerators and nuclear fission reactors [5,8]. At the first, the capture efficiency of a charged point defect to a charged sink was derived taking into account electric interaction between the defects. And then, numerical analyses were conducted, where the defect accumulation in β-SiC during high-energy electron irradiation was calculated using the kinetic rate theory based model with the derived capture efficiency. Finally, the calculation results were compared with that of an HVEM observation experiment.

2. Procedure

2.1. Derivation of the capture efficiency of a charged point defect to a charged sink

As represented in Fig. 1, consider a charged point defect i migrating in the electric field which is created by a charged spherical sink j, where the sink is also a point defect or a defect cluster. The migrating defect i will diffuse depending on the electrical potential gradient in addition to the defect concentration gradient; and then, the diffusion flux of the defect i can be expressed as [9]

$$j = \frac{1}{\Omega}\left(-D_i \nabla C_i - \frac{e q_i D_i C_i}{k_B T}\nabla V\right), \quad (1)$$

where V is the electrical potential in eV at the location of the defect i, e is the elementary electric charge, q_i is the effective electric charge of the defect i in dimensionless unit, D_i and C_i are the diffusion coefficient and concentration of defect i in the matrix, respectively, Ω is atomic volume of the defect i, k_B is the Boltzmann

* Corresponding author. Tel.: +81 774 38 3463; fax: +81 774 38 3467.
E-mail address: y-watanabe@iae.kyoto-u.ac.jp (Y. Watanabe).

0022-3115/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2008.12.098

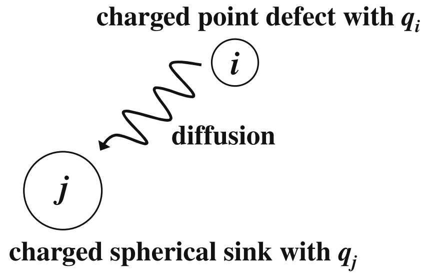

Fig. 1. Schematic representation of the diffusion of a charged point defect around a charged spherical sink.

constant and T is the temperature. For simplicity, it is assumed here that the diffusion flux of Eq. (1) is isotropic around the sink j. When the radial component of the electrical field at r shown as $V = -eq_j/(4\pi\varepsilon_0 r)$ is applied, the total current of the defect i to the sink j is written by

$$J_i = \frac{1}{\Omega} 4\pi R_j Z_{ij} \left[D_i C_i - D_i C_i^{\text{eq}} \exp\left(\frac{e^2 q_i q_j}{4\pi\varepsilon_0 R_j} \frac{1}{k_B T} \right) \right], \tag{2}$$

$$Z_{ij} = \begin{cases} \dfrac{\frac{e^2 q_i q_j}{4\pi\varepsilon_0 R_j k_B T}}{\left[\exp\left(\frac{e^2 q_i q_j}{4\pi\varepsilon_0 R_j}\frac{1}{k_B T}\right) - 1\right]} & \ldots \quad (q_i q_j \neq 0) \\ 1 & \ldots \quad (q_i q_j = 0) \end{cases}, \tag{3}$$

where, R_j and q_j are the radius and an effective charge of the sink j, respectively, C_i is the concentration of defect i in the matrix, C_i^{eq} is the concentration of the defect i around the sink j. Z_{ij} is the capture efficiency of the defect i to the sink j.

2.2. Rate theory model of defect accumulation

Numerical analysis for simulating defect accumulation process in β-SiC irradiated with a high-energy electron beam at 973 K was performed with the following assumptions:

(a) Frenkel pairs (silicon interstitials, silicon vacancies, carbon interstitials and carbon vacancies) are homogeneously produced in β-SiC by atomic displacements under high-energy electron irradiation. The production rates assumed to be 5.31×10^{-4} dpa/s for silicon Frenkel pairs and 6.48×10^{-4} dpa/s for carbon Frenkel pairs, which are approximated from the condition in the HVEM experiment [8].
(b) Migration energies of vacancies are so high [10] that vacancies can not freely migrate at this temperature.
(c) Interstitials can freely migrate with the migration energies of 1.53 eV [10] for silicon interstitials and 0.74 eV [10] for carbon interstitials.
(d) The surface of specimen is a permanent sink to defects, and the sink strength is evaluated from the specimen thickness. It is estimated to be 100 nm which is the same value as that in the HVEM experiment [8].
(e) Same type defects mutually annihilate: $I^{Si} + V^{Si} \to 0$, $I^C + V^C \to 0$. (I^{Si} for silicon interstitials, V^{Si} for silicon vacancies, I^C for carbon interstitials and V^C for carbon vacancies).
(f) Antisite defects (Si_C for silicon atom located at carbon lattice site and C_{Si} for carbon atom located at silicon lattice site) are produced by the interactions between silicon interstitial and carbon vacancy, and between carbon interstitial and silicon vacancy, respectively: $I^{Si} + V^C \to Si_C$, $I^C + V^{Si} \to C_{Si}$. It is assumed that antisite defects can not be thermally dissociated at this temperature due to the relatively high biding energies of 1.0∼8.5 eV [10,11].
(g) Two interstitials form a di-interstitial: $I^{Si} + I^{Si} \to I_2^{Si}$, $I^C + I^C \to I_2^C$, $I^{Si} + I^C \to I^{Si}I^C$. A di-interstitial is assumed to be thermally stable at this temperature, and is regarded as the nucleus of an interstitial loop. A di-interstitial grows into a larger interstitial loop by absorbing interstitials. For simplicity, the size distribution of interstitial loops is not considered. It is assumed that an interstitial loop has the effective charge that is the sum of the effective charges of interstitials in the loop.
(h) A point defect in β-SiC can have a wide range of charge state [12], and the exact value is not understand, yet. Therefore, the effective charge of a point defect is here treated as an adjustable parameter with the absolute value ranging from 0.0 to 2.0. For simplicity, it is assumed that silicon interstitial and carbon vacancy have the same value of effective charge in positive, while a carbon interstitial and a silicon vacancy have the same value of effective charge in minus.

Simultaneous rate equations derived from the above assumption (a)–(h) were solved using the backward differentiation formula method [13].

3. Results and discussion

In Fig. 2, the calculated capture efficiency Z_{ij} is plotted as a function of $x = e^2 q_i q_j/(4\pi\varepsilon_0 R_j k_B T)$. It shows a monotonic decreasing function of x. When q_i and q_j are opposite in sign to each other, x takes negative and Z_{ij} is greater than 1, showing that a charged point defect i is strongly attractive with a charged sink j. In contrast, when q_i and q_j are the same in sign as each other, Z_{ij} is less than 1, indicating a strong repulsive interaction between the charged defects. When $q_i = 0$ or $q_j = 0$, x takes 0 and Z_{ij} is 1, where the defect i diffuses depending on only the defect concentration gradient.

As to numerical analysis by the rate theory based model for defect accumulation in β-SiC irradiated with a high-energy electron beam at 973 K, Fig. 3 shows the time dependence of concentration of interstitial loops, C_L, under the following three cases for effective charge of point defects: (I) $q_I^{Si} = q_V^C = q_I^C = q_V^{Si} = 0.0$ (no effective charge), (II) $q_I^{Si} = q_V^C = +0.5$, $q_I^C = q_V^{Si} = -0.5$ and (III) $q_I^{Si} = q_V^C = +2.0$, $q_I^C = q_V^{Si} = -2.0$. C_L increase dramatically with increasing irradiation time, and saturates after about 10^2 s. Although the saturated C_L does

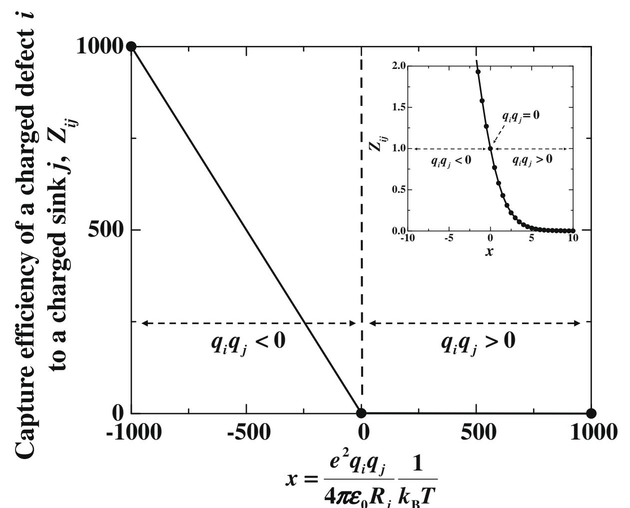

Fig. 2. Calculated capture efficiency Z_{ij} as a function of $x = e^2 q_i q_j/(4\pi\varepsilon_0 R_j k_B T)$. It shows a monotonic decreasing function of x, where $Z_{ij} > 1$ at $x < 0$ ($q_i q_j < 0$), $Z_{ij} = 1$ at $x = 0$ ($q_i q_j = 0$) and $Z_{ij} < 1$ at $x > 0$ ($q_i q_j > 0$).

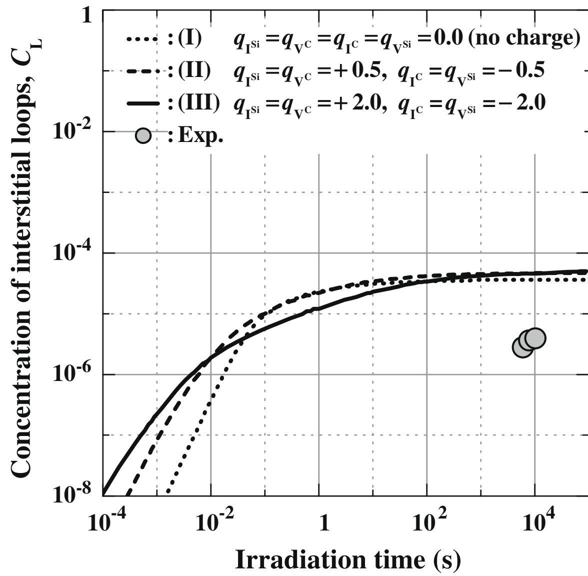

Fig. 3. Time dependence of concentration of interstitial loops, C_L, under the following three cases for effective charge of point defects: (I) $q_I^{Si} = q_V^C = q_I^C = q_V^{Si} = 0.0$ (no effective charge), (II) $q_I^{Si} = q_V^C = +0.5$, $q_I^C = q_V^{Si} = -0.5$ and (III) $q_I^{Si} = q_V^C = +2.0$, $q_I^C = q_V^{Si} = -2.0$.

not depend much on effective charges, the nucleation of interstitial loops in the cases of (II) and (III) causes in much earlier time than those in the case of (I). This is because the production of di-interstitial as the nucleus of an interstitial loop is enhanced, where the most of produced di-interstitial is $I^{Si}I^C$ which is mainly formed by the electric attractive interaction between I^{Si} and I^C. Fig. 4 represents the time dependence of concentration of clustered interstitials, C_{LI}, which is defined by the total number of interstitials in all loops. Notice that the fraction, C_{LI}/C_I, means the average number of interstitials in a single interstitial loop. As shown in the figure, C_{LI} greatly depends on effective charge and decrease with increasing the magnitude of charges, leading to that the average loop size (C_{LI}/C_I) becomes smaller. This is because an interstitial loop grows while keeping its charge to be as neutral as possible, resulting in the suppression of the growth of interstitial loop. In fact, as shown in Fig. 5,

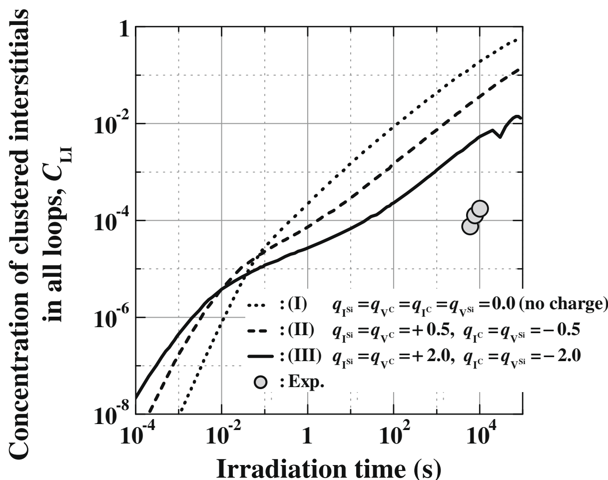

Fig. 4. Time dependence of concentration of clustered interstitials in all loops, C_{LI}, under the following three cases for effective charge of point defects: (I) $q_I^{Si} = q_V^C = q_I^C = q_V^{Si} = 0.0$ (no effective charge), (II) $q_I^{Si} = q_V^C = +0.5$, $q_I^C = q_V^{Si} = -0.5$ and (III) $q_I^{Si} = q_V^C = +2.0$, $q_I^C = q_V^{Si} = -2.0$.

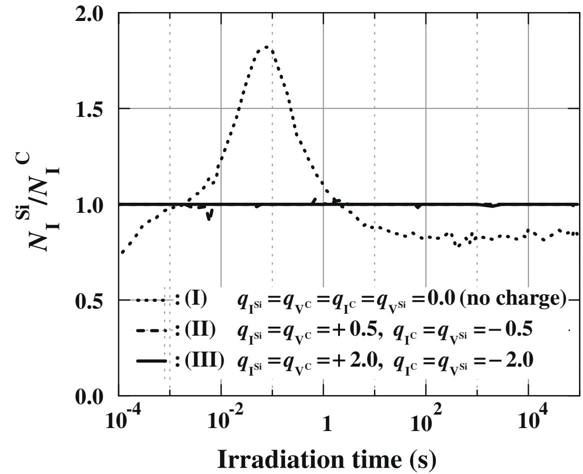

Fig. 5. Time dependence of the fraction of the numbers of silicon interstitials and carbon interstitial in an interstitial loop, N_I^{Si}/N_I^C, under the following three cases for effective charge of point defects: (I) $q_I^{Si} = q_V^C = q_I^C = q_V^{Si} = 0.0$ (no effective charge), (II) $q_I^{Si} = q_V^C = +0.5$, $q_I^C = q_V^{Si} = -0.5$ and (III) $q_I^{Si} = q_V^C = +2.0$, $q_I^C = q_V^{Si} = -2.0$.

the fraction of the numbers of silicon interstitials and carbon interstitial in an interstitial loop, N_I^{Si}/N_I^C, is always about 1 when defects have effective charges. It indicates that the stoichiometric constraint drastically retards the growth of interstitial loops.

Also Figs. 3 and 4 show the experimental data from the HVEM observation [8], in which the number and size of larger interstitial loops than those with about 1 nm in diameter are measured because of the resolution limit of the microscopy. Therefore, the absolute value of the experimental data is quantitatively different from the calculation data; nevertheless, roughly speaking, the calculated concentration of clustered interstitials with effective charges approaches to the experimental data. It implies that the capture efficiency derived in the present work may play an important role for better reproducing of the experimental result.

4. Summary

In order to investigate the formation kinetics of interstitial loops in β-SiC during irradiation, a capture efficiency of a charged point defect to a charged sink was derived taking into account electric interaction between the defects. The derived capture efficiency depends much on effective charges of defects. For defect accumulation in β-SiC under high-energy electron irradiation, the numerical analysis using the rate theory based model with the capture efficiency was performed, and the calculation results indicate that an interstitial loop grows while keeping its charge to be as neutral as possible, leading to the suppression of the growth of interstitial loops. Roughly speaking, the calculated concentration of clustered interstitials with effective charges approaches to the experimental data. It implies that the capture efficiency derived in the present work may play an important role for better reproducing of the experimental result.

References

[1] M.A. Capano, R.J. Trew, MRS Bull. 22 (1997) 19.
[2] L.L. Snead, T. Nozawa, Y. Katoh, T. Byun, S. Kondo, D.A. Pett, J. Nucl. Mater. 371 (2007) 329.
[3] Y. Katoh, N. Hashimoto, S. Kondo, L.L. Snead, A. Kohyama, J. Nucl. Mater. 351 (2006) 228.
[4] L.L. Snead, R. Scholz, A. Hassegawa, A.F. Rebelo, J. Nucl. Mater. 307–311 (2002) 1141.
[5] T. Yano, T. Iseki, Phil. Mag. A. 62–64 (1990) 421.

[6] S.J. Zinkle, C. Kinoshita, J. Nucl. Mater. 251 (1997) 200.
[7] A.I. Ryazanov, A.V. Klaptsov, A. Kohyama, H. Kishimoto, J. Nucl. Mater. 307–311 (2002) 1107.
[8] S. Kondo, Irradiation effects on microstructural evolution in SiC, PhD thesis, Kyoto University, Japanese, 2006.
[9] P.G. Shewmon, Diffusion In Solids, McGraw-Hill Book Company, 1963.
[10] F. Gao, W.J. Weber, M. Posselt, V. Belko, Phys. Rev. B 69 (2004) 245205.
[11] F. Gao, E.J. Bylaska, W.J. Weber, L.R. Corrales, Phys. Rev. B 64 (2001) 245208.
[12] M. Bockstedte, M. Heid, O. Pankratov, Phys. Rev. B 67 (2003) 193102.
[13] S.D. Cohen, A.C. Hindmarsh, Computer. Phys. 10 (1996) 138.