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Effect of helium on dislocation loop formation and radiation swelling in SiC

A.I. Ryazanov^{a,*}, A.V. Klaptsov^a, A. Kohyama^b, Y. Katoh^b, H. Kishimoto^c

^a Russian Research Center, Kurchatov Institute, 123182 Moscow, Russia

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

^c Department of Chemical Engineering, University of California, Santa Barbara, Engineering II, Rm 3357,

Santa Barbara, CA 93106-5080, USA

Abstract

SiC and SiC/SiC composites are considered as candidate materials for fusion reactors. In a fusion reactor environment, helium atoms will be produced in SiC up to very high concentrations (15000–20000 at. ppm) and therefore it is very important to understand how helium effects radiation swelling of SiC. In this paper a theoretical model of the helium effect on radiation swelling of SiC is suggested. This model is based on considering of kinetic growth of dislocation loops in the matrix taking into account the effect an internal electric field formed near dislocation loops has on diffusion processes of charged point defects. The trapping of helium atoms by vacancies results in an enhanced growth rate of dislocation loops and finally a swelling increase. The theoretical results for radiation swelling are compared with the existing experimental data. It is shown that helium atoms increase the radiation swelling of SiC, especially at high temperatures.

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1. Introduction

Silicon carbide (SiC) and its composites are expected to be used as structural materials for fusion reactor due to their high thermo-chemical stability, radiation resistance and low-induced radioactivity [1]. The radiation resistance of ceramic materials under neutron irradiation is determined by such phenomena as radiation swelling and creep. When ceramic materials are irradiated, point defects are produced in the matrix and then they have the tendency to accumulate in defect clusters such as voids, dislocation loops, etc. The kinetics of radiation swelling and creep is related to nucleation and growth of such clusters. Point defects: vacancies and interstitial atoms in ceramic materials can have an effective charge and this charge state can change during diffusion process and due to trapping of electrons (e.g., an F^+ , F centres: vacancies with a single and two trapped electrons). The kinetics of growth and accumulation of charged point defects in defect clusters takes place under the effect of internal electric field, which is produced by the system of charged point defects and defect clusters in the matrix. This electric field affects the diffusion process of charged point defects, and the growth kinetics of voids and dislocation loops in such ceramic materials are determined by completely different mechanisms than in metals.

In a previous paper [2], the physical mechanisms of low-temperature radiation swelling of fusion ceramic materials were investigated. The suggested theoretical model for the description of radiation swelling was based on charged point defect kinetics in ceramic materials [3]. This theoretical model accounted for the charge state of point defects and the effect of an internal electric field produced under irradiation in the matrix on the diffusion processes of point defects in ceramic materials.

^{*}Corresponding author. Tel.: +7-095 196 9177; fax: +7-095 421 4598.

E-mail address: ryazanoff@comail.ru (A.I. Ryazanov).

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A very important problem for the investigation of radiation resistance of SiC is the effect of helium on radiation swelling. Many papers [4–8] have been published concerning the effect of helium on the behaviour of SiC under irradiation. In the present paper, a theoretical model of radiation swelling is developed that takes into account the effects of helium on the nucleation and growth of dislocation loops. In this model helium atoms play two roles: they can trap vacancies and due to this process interstitial dislocation loops can grow faster and helium atoms can promote the nucleation of interstitial dislocation loops.

2. Physical model for low-temperature radiation swelling under helium beam irradiation conditions

Let us consider the physical mechanisms of radiation swelling in SiC taking into account helium effects on this phenomenon. The experimental investigation of these processes are done under dual beam irradiation of SiC [8], when helium ions and other types of ions (Si^{2+} , C^{2+} , Ni³⁺) bombard the target. Under these irradiation conditions the point defects (interstitial atoms, vacancies and interstitial helium atoms) are generated simultaneously in the materials. The interstitial atoms and vacancies produced in both components of SiC have an effective charge, which makes them very efficient traps for either electrons or holes. Due to such traps, the effective charge and concentration of point defects can change during irradiation. The interstitial atoms and vacancies can also mutually recombine, can trap by the helium atoms, existing dislocations and point defect clusters (dislocation loops, voids). Each point defect creates in the matrix an elastic strain field. These strains lead to the overall volume increase of the irradiated material by $e_{\alpha}\omega$ [9], where e_{α} is the dilatation of point defect of type α ($\alpha = I$ for interstitial atoms, $\alpha = V$ for vacancies and $\alpha = He$ for helium atoms) and ω is the atomic volume. The absorption of the interstitial atom and the vacancy on a sink of type s (dislocation loop, void, etc.) results in the change of sample volume by the value $e_{\alpha s}\omega$. We denote the total number of the interstitial atoms and the vacancies absorbed by sinks of the type s in a unit volume as equal to n_{α}^{S} and the concentrations of free point defects equal to $C_{\alpha 1}$, $C_{\alpha 2}$ for the two components (K = 1 = Si, K = 2 = C) and C_{He} for helium atoms. Some vacancies may capture helium atoms, leading to a vacancy-helium atom compound, which changes the sample volume by the value $e_{\text{HeV}}\omega$ with the concentration of the compound equal to C_{HeV} . Thus, the total volume increase of SiC under irradiation or radiation swelling (S_{tot}) at some irradiation dose can be written in the following form:

$$S_{\text{tot}} = \sum_{K=1}^{2} C_{\text{IK}} e_{K} + \sum_{K=1}^{2} C_{\text{VK}} e_{\text{VK}} + C_{\text{He}} e_{\text{He}} + \sum_{K=1}^{2} C_{\text{HeVK}} e_{\text{HeVK}} + \omega \sum_{S,K} \left(n_{\text{IK}}^{S} e_{\text{IK}} + n_{\text{VK}}^{S} e_{\text{VK}} \right).$$
(1)

Here the summation is over all different sink types (dislocations, dislocation loops and voids). For the calculation of the dilatations of free point defects, a computer simulation should be carried out of equilibrium configurations of point defects in the crystal lattice with a realistic interatomic potential for this material. In our calculations of radiation swelling, we will use the values for dilatations of point defects which are close to analogous values for the metals and which are equal to $e_{IK} = 1.2$, $e_{VK} = -0.1$ and $e_{He} = 1.2$. The absorption of an interstitial atom or vacancy by a dislocation network (s = D) and dislocation loop (s = L) results in the volume change by the values ω and $-\omega$, respectively, so we will use $e_{IK}^{D} = e_{IK}^{L} = 1$ and $e_{VK}^{D} = e_{VK}^{L} = -1$. We should also calculate the dilatation of vacancy-helium atom compounds, which is equal to $e_{\text{HeV}} = 1.1$

We will investigate the physical mechanisms of radiation swelling at the irradiation doses and temperatures when the crystal lattice contains only the dislocation network with the dislocation density $\rho_{\rm D}$ and interstitial dislocation loops with the volume density $N_{\rm L}$, whose concentration is saturated at the maximum value under irradiation. These crystal defects are considered only as sinks for point defects of the two components (K = 1 and 2). Using the material conservation law for point defects of component K which takes into account the recombination of point defects in the matrix and accumulation of them on the dislocation network and dislocation loops when the nucleation stage is finished, we can write the following expression:

$$\sum_{K=1}^{2} \left[C_{IK} + \omega \left(n_{IK}^{D} + n_{IK}^{L} \right) \right] + C_{He}$$
$$= \sum_{K=1}^{2} \left[C_{VK} + \omega \left(n_{VK}^{D} + n_{VK}^{L} \right) \right] + Pt, \qquad (2)$$

where P is the generation rate of helium atoms. We can rewrite the relation for radiation swelling from Eq. (1) in the following form:

$$S_{\text{tot}} = Pt + \sum_{K=1}^{2} \left[C_{VK} (1 + e_{VK}) + C_{IK} (e_{IK} - 1) + C_{\text{HeVK}} e_{\text{HeV}} \right] + C_{\text{He}} (e_{\text{He}} - 1).$$
(3)

From Eq. (3) we can see that the radiation swelling under helium beam irradiation conditions is determined

by the accumulation of generated vacancies of both components and free and captured helium atoms. Let us determine now the main parameters and dependencies of radiation swelling SiC at the early stage of irradiation considering the growth kinetics of dislocation loops and the kinetics of point defect accumulations of both types in the matrix.

3. Accumulation of point defects and evolution of microstructure in the matrix of irradiated SiC by helium ion beam

Helium ion irradiation of SiC produces displaced atoms (point defects in two components: Si and C) with a strong difference of generation rates [10]. Due to the strong difference of threshold energies in SiC $(E_d = 93 \text{ eV} \text{ for Si} \text{ and } E_d = 16 \text{ eV} \text{ for C})$ [10] the generation rates of silicon $(G_1 = G_{\text{Si}})$ and carbon $(G_2 = G_{\text{C}})$ on the two sublattices are also quite different and can vary between neutron and ion irradiation in the interval: $G_{\text{C}}/G_{\text{Si}} \approx 3-6$. In order to describe the physical mechanisms of radiation swelling in SiC under irradiation, it is important to consider electric neutrality and that point defects have an effective electric charge.

As we have mentioned, we will consider defect microstructure evolution at early stages of irradiation when only dislocation loops are formed and the temperature is not high enough (T < 1100 K) for void production. This is the typical situation for the evolution of defect microstructure in ceramic materials after the onset of irradiation [11,12]. In this case the point defect sinks are only the dislocation network and the dislocation loops. The concentration of vacancies is changed mostly due to recombination with mobile interstitial atoms, absorption by line dislocations and dislocation loops, and capture of highly mobile helium atoms. The ensemble of dislocation loops is characterized by the average loop radius $R_{\rm L}$ and volume density $N_{\rm L}$. The processes of absorption of charged point defects by dislocations and dislocation loops have special peculiarities. In our model, the interstitial dislocation loops and straight edge dislocations are considered to be additional planes composed of two types of charged atoms: anions and cations. Therefore, due to Coulomb repulsion in ionic materials there is a strong driving force for interstitial condensation on a dislocation loop or dislocation line to remain stochiometric, or at least to balance anion and cation charges. Here we propose an advanced theoretical model similar to a previous one [2]. It can be shown that the total current of charged Kth component point defects to the dislocation line in the case of materials which have a stochiometric composition like MgO, can be written in the following form:

$$\begin{split} J_{11} &= 2\pi R_{\rm L} \cdot \pi r_0 j_{11}^n = 2\pi R_{\rm L} \cdot \frac{2\pi}{\ln(8R_{\rm L}/r_0)} \\ &\times \frac{D_{11} C_{11} (D_{12} C_{12} + D_{\rm V1} C_{\rm V1})}{D_{11} C_{11} + D_{12} C_{12} + D_{\rm V1} C_{\rm V1} + D_{\rm V2} C_{\rm V2}}, \\ J_{12} &= 2\pi R_{\rm L} \cdot \pi r_0 j_{12}^n = 2\pi R_{\rm L} \cdot \frac{2\pi}{\ln(8R_{\rm L}/r_0)} \\ &\times \frac{D_{12} C_{12} (D_{11} C_{11} + D_{\rm V2} C_{\rm V2})}{D_{11} C_{11} + D_{12} C_{12} + D_{\rm V1} C_{\rm V1} + D_{\rm V2} C_{\rm V2}}, \\ J_{\rm V1} &= 2\pi R_{\rm L} \cdot \pi r_0 j_{\rm V1}^n = 2\pi R_{\rm L} \cdot \frac{2\pi}{\ln(8R_{\rm L}/r_0)} \\ &\times \frac{D_{\rm V1} C_{\rm V1} (D_{\rm V2} C_{\rm V2} + D_{\rm I1} C_{\rm I1})}{D_{11} C_{\rm I1} + D_{\rm I2} C_{\rm I2} + D_{\rm V1} C_{\rm V1} + D_{\rm V2} C_{\rm V2}}, \\ J_{\rm V2} &= 2\pi R_{\rm L} \cdot \pi r_0 j_{\rm V2}^n = 2\pi R_{\rm L} \cdot \frac{2\pi}{\ln(8R_{\rm L}/r_0)} \\ &\times \frac{D_{\rm V2} C_{\rm V2} (D_{\rm V1} C_{\rm V1} + D_{\rm I2} C_{\rm I2})}{D_{\rm I1} C_{\rm I1} + D_{\rm I2} C_{\rm I2} + D_{\rm V1} C_{\rm V1} + D_{\rm V2} C_{\rm V2}}, \end{split}$$

where r_0 is the dislocation core radius, D_{IK} and D_{VK} are the diffusion coefficients of interstitial atoms and vacancies of the *K*th component, respectively.

Eqs. (4) were obtained using the condition of electric neutrality for charged point defects in the matrix. Meanwhile, this condition should follow from the kinetic equations used here. In our previous papers [2,13], we did not take into account exactly the effect of an internal electric field on the diffusivity of charged point defects in the matrix far from the grain boundary. Here we will take into account this effect on the diffusion accumulation of charge point defects in the matrix. The kinetic equations for the concentrations of point defects in the presence of an internal electric field in this case have the following form

$$\frac{\partial C_{\alpha}}{\partial t} = F_{\alpha}(\{C_{\alpha}\}) - \operatorname{div} \mathbf{j}_{\alpha},\tag{5}$$

$$\mathbf{j}_{\alpha} = -D_{\alpha}\nabla C_{\alpha} - \frac{q_{\alpha}}{kT} D_{\alpha}C_{\alpha}\nabla\varphi, \tag{6}$$

where q_{α} , D_{α} and C_{α} are the charge, diffusion coefficient and concentration of α -type's point defect, respectively; $F_{\alpha}(\{C_{\alpha}\})$ is the term describing the different processes such as a point defect generation, annihilation and so on; \mathbf{j}_{α} is the current of point defects in the matrix; φ is the electric potential in the matrix. The electric potential in the matrix φ can be found from the following Poisson equation:

$$\Delta \varphi = -\frac{4\pi}{\varepsilon \omega} \rho, \tag{7}$$

where ρ is the total density of charged point defects, ω is the atomic volume, ε is the dielectric permeability. In general Eqs. (5)–(7) should solve with the boundary conditions and equations describing the nucleation and growth of interstitial dislocation loops. However, if the size of sample is much larger than the denuded zone size, we can see that the concentrations of charged point defects do not have space dependencies in the matrix, but the electric potential φ according Eq. (7) has the space dependence. In this case Eq. (5), which takes into account (6), can be written as

$$\frac{\mathrm{d}C_{\alpha}}{\mathrm{d}t} = F_{\alpha}(\{C_{\alpha}\}) - \Phi_{\alpha}D_{\alpha}C_{\alpha}, \quad \Phi_{\alpha} = \frac{4\pi q_{\alpha}}{\varepsilon\omega kT}\rho.$$
(8)

Thus the kinetic equations describing the nucleation and growth of dislocation loops and the accumulation of charged point defects under irradiation in the matrix for two components (K = 1, 2) of ceramic material (SiC) can be written in the following form:

$$\frac{\mathrm{d}C_{\mathrm{V}K}}{\mathrm{d}t} = G_{\mathrm{V}K} - j_{\mathrm{V}K}(\rho_{\mathrm{D}} + \rho_{\mathrm{L}}) - \alpha D_{\mathrm{I}K}C_{\mathrm{I}K}C_{\mathrm{V}K} - \nu D_{\mathrm{He}}C_{\mathrm{He}}C_{\mathrm{V}K} - \Phi_{\mathrm{V}K}D_{\mathrm{V}K}C_{\mathrm{V}K}, \qquad (9)$$

$$\frac{dC_{IK}}{dt} = G_{IK} - j_{IK}(\rho_{\rm D} + \rho_{\rm L}) - \alpha D_{IK}C_{IK}C_{VK} - \mu(D_{I1} + D_{I2})C_{I1}C_{I2} - \gamma(D_{IK} + D_{\rm He})C_{IK}C_{\rm He} - \Phi_{IK}D_{IK}C_{IK},$$
(10)

10

$$\frac{dC_{\text{He}}}{dt} = P - \nu D_{\text{He}} C_{\text{He}} C_{\text{V1}} - \nu D_{\text{He}} C_{\text{He}} C_{\text{V2}} - \gamma (D_{\text{I1}} + D_{\text{He}}) C_{\text{I1}} C_{\text{He}} - \gamma (D_{\text{I2}} + D_{\text{He}}) C_{\text{I2}} C_{\text{He}},$$
(11)

$$\omega \frac{dN_{\rm L}}{dt} = \mu (D_{11} + D_{12})C_{11}C_{12} + \gamma (D_{11} + D_{\rm He})C_{11}C_{\rm He}
+ \gamma (D_{12} + D_{\rm He})C_{12}C_{\rm He},
\Phi_{11} = -\Phi_{12} = -\Phi_{\rm V1} = \Phi_{\rm V2}
= \frac{4\pi q^2}{\varepsilon \omega kT} (C_{11} - C_{12} - C_{\rm V1} + C_{\rm V2}) + (\rho_{\rm D} + \rho_{\rm L})
\times \frac{2\pi}{\ln(8R/r_0)} \left[\frac{D_{11}C_{11} - D_{12}C_{12} - D_{\rm V1}C_{\rm V1} + D_{\rm V2}C_{\rm V2}}{D_{11}C_{11} + D_{12}C_{12} + D_{\rm V1}C_{\rm V1} + D_{\rm V2}C_{\rm V2}}
- \frac{C_{11} - C_{12} - C_{\rm V1} + C_{\rm V2}}{C_{\rm U1} + C_{12} + C_{\rm V1} + C_{\rm V2}} \right].$$
(12)

Here $\rho_{\rm D}$ is the network dislocation density, $\rho_{\rm L}$ is the dislocation density of dislocation loops ($\rho_{\rm L} = 2\pi R_{\rm L} N_{\rm L}$); $G_{\rm VK}$, $G_{\rm IK}$, $G_{\rm He}$ are the generation rates of vacancies and interstitial atoms *K*th components ($G_{\rm V1} = G_{\rm I1} = G_{\rm Si}$, $G_{\rm V2} = G_{\rm I2} = G_{\rm C}$) and helium atoms respectively, α is the point defect recombination coefficient ($\alpha = 3/a^2$, *a* is the lattice spacing), ν is the coefficient of helium atom capture by vacancies, γ is the coefficient of loop formation due to coupling of helium and interstitial atoms ($\gamma = 3/a^2$), and *b* is the Burger's vector module.

The growth rate of a dislocation loop taking into account the absorption of two types of interstitial atoms

and vacancies and maintaining the stoichiometry of the dislocation loop is given by the following relation:

$$\frac{dR_{\rm L}}{dt} = \frac{\pi r_0}{b} \sum_{K} (j_{\rm IK}^n - j_{\rm VK}^n)
= \frac{4\pi}{b \ln(8R/r_0)} \frac{D_{\rm II} C_{\rm I1} D_{\rm I2} C_{\rm I2} - D_{\rm V1} C_{\rm V1} D_{\rm V2} C_{\rm V2}}{D_{\rm I1} C_{\rm I1} + D_{\rm I2} C_{\rm I2} + D_{\rm V1} C_{\rm V1} + D_{\rm V2} C_{\rm V2}},$$
(13)

where b is the Burger's vector module.

The self-consistent solution of system of kinetic equations (3) and (4) and (9)–(13) with the following initial conditions (at t = 0):

$$C_{IK}(t=0) = 0, \quad C_{VK}(t=0) = 0,$$

 $R_L(t=0) = a, \quad C_{He}(t=0) = 0$
(14)

permits the main parameters of radiation swelling to be found under different types of irradiation taking into account effect of helium. The results of numerical calculations of the system equations (3) and (4) and (9)– (13) with the initial conditions (14) using the material parameters for SiC, which were fitted the experimental data [8], (see Table 1) are presented in Figs. 1–4. In our numerical calculations we have used the following expressions for diffusion coefficients of charged point defects: $D_{\alpha K} = D_{\alpha K}^{O} \exp(-E_{m\alpha}^{K}/kT)$, (where $D_{\alpha K}^{O} = 10^{-4}$ cm²/s). The concentration of vacancy-helium atom compounds has been obtained by solving the following equation:

Table 1

Main values used for numerical calculations of radiation-induced swelling in SiC [10,14]

$G_1 = G_{\rm Si}$	Point defect generation	0.4×10^{-3} dpa/s
	rate for Si atoms	
$G_2 = G_C$	Point defect generation	1.0×10^{-3} dpa/s
	rate for C atoms	
Р	Helium atom generation	6.0×10^{-8} He/s
	rate	
$E_{\rm mV}^{\rm Si}$	Silicon vacancy migration	1.8 eV
	energy	
$E_{\rm mV}^{\rm C}$	Carbon vacancy migration	1.7 eV
	energy	
$E_{\mathrm{mI}}^{\mathrm{Si}}$	Silicon interstitial migra-	0.3 eV
	tion energy	
$E_{\rm mI}^{\rm C}$	Carbon interstitial migra-	0.2 eV
	tion energy	
$E_{\rm m}^{\rm He}$	Helium atom migration	0.3 eV
III	energy	
$\rho_{\rm D}$	Network dislocation den-	$10^{10} { m cm}^{-2}$
	sity	
а	Lattice parameter	3.0×10^{-8} cm
q	Effective charge of point	2e
-	states	
е	Charge of electron	4.8×10^{-10} esu
	0	



Fig. 1. A comparison of experimental data taken from Ref. [8] and theoretical calculations of radiation swelling without helium implantation P = 0.0 He/s (single beam) and with helium implantation $P = 6.0 \times 10^{-8}$ He/s (dual beam).



Fig. 2. The temperature dependence of dislocation loop radius at 1.0 dpa without helium implantation P = 0.0 He/s and with helium implantation $P = 6.0 \times 10^{-8}$ He/s at different values of the coefficient of helium atom capture by vacancy *v*.

$$\frac{dC_{\rm HeV1}}{dt} = vD_{\rm He}C_{\rm He}C_{\rm V1}, \quad \frac{dC_{\rm HeV2}}{dt} = vD_{\rm He}C_{\rm He}C_{\rm V2}.$$
(15)

The values for the migration energy of charged point defects used in the numerical calculations are presented in Table 1. A comparison between the experimental data [8] and theoretical calculations both in the case of presence of helium implantation and in the case of absence one for the fluence dependence of radiation swelling of SiC is given in Fig. 1. Since we did not have the theoretical evaluations of the coefficient of helium atom capture by vacancy v, we used this parameter as a free parameter. A comparison between theoretical calculations for the temperature dependencies of loop radiation R, the density of dislocation loops $N_{\rm L}$ and radiation



Fig. 3. The temperature dependence of dislocation loop density at 1.0 dpa without helium implantation P = 0.0 He/s and with helium implantation $P = 6.0 \times 10^{-8}$ He/s at different values of the coefficient of helium atom capture by vacancy v.



Fig. 4. The temperature dependence of swelling at 1.0 dpa without helium implantation P = 0.0 He/s and with helium implantation $P = 6.0 \times 10^{-8}$ He/s at different values of the coefficient of helium atom capture by vacancy v.

swelling of SiC for single beam (without helium implantation) and dual beam (with helium implantation) at 1 dpa are given in Figs. 2–4. It is seen that the presence of helium atoms in the matrix increases the radiation swelling in temperature region from 400 to 900 K at small values of the coefficient of helium atom capture by vacancy $v \sim 10^{-6}/a^2$. However, helium atoms have slight influence on radiation swelling of SiC at lower and at higher temperatures. As it is seen from the calculations, the additional term $\Phi_{\alpha K}$ in Eqs. (9) and (10) does not affect strongly on final results. So at least if the migration energies of interstitials and vacancies are comparable for two types of components (K = 1, 2) the effect of an internal electric field on the diffusivity of charged point defects in the matrix is small.

4. Conclusions

Based on the recently obtained experimental data and theoretical results, we can make the following conclusions concerning the radiation-induced swelling behaviour of SiC taking into account the effect of helium:

- Helium atoms in the temperature interval from 400 to 900 K increase the nucleation of interstitial dislocation loops and decrease their average radius.
- The presence of helium atoms in the matrix in the temperature range from 400 to 900 K increases the radiation swelling.
- The theoretical calculations are located in a good agreement with the existed experimental data at low fluence.

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