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Radiation swelling of SiC under neutron irradiation

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Abstract

Ceramic materials produced on the basis of SiC and SiC/SiC composites are considered due to their high temperature strength, pseudo-ductile fracture behavior and low-induced radioactivity as candidate materials for fusion reactors. The radiation resistance of ceramic materials under neutron irradiation is the key problem which determines the use of these materials in fusion reactor environment. In the present paper the general physical mechanisms of radiation swelling of SiC are investigated. Recent experimental results concerning the effect of neutron and charged particle irradiation on radiation swelling of SiC are presented. A new theoretical model is suggested for the description of radiation swelling in ceramic materials. Point defects in ceramic materials can have an effective charge (e.g., an F+ center, vacancy with a single trapped electron). The theoretical model is based on kinetic consideration of charged point defect accumulation and kinetic growth of dislocation loops in the matrix taking into account the effect of internal electric field formed under irradiation in the matrix on diffusion processes of charged point defects. The theoretical results for radiation swelling are compared with the existing experimental data for irradiated SiC material.

1. Introduction

Radiation resistance of ceramic materials produced on the basis of SiC and SiC/SiC composites in fusion reactor environment is determined by radiation damage accumulation under high energy neutrons and ions from the fusion plasma. The possibility of use of these materials in fusion applications is based on their high temperature strength, pseudo-ductile fracture behavior and low-induced radioactivity. The key point in investigations of degradation of mechanical properties under irradiation like creep, fatigue, shape stability and thermal properties is the understanding of physical mechanisms of point defect accumulation and the investigation of such phenomena as radiation creep and swelling. Point defects (vacancies and interstitials) in ceramic materials can have an effective charge which can change due to traps of electrons (e.g., F+, F centers: vacancies with a single or two trapped electrons). Due to the accumulation of charged point defects in the matrix an internal effective electrical field is formed which affects the diffusion process of charged point defects. The growth kinetics of voids and dislocation loops in such materials have completely different mechanisms comparing with metals.

Many results on radiation effects in ceramic materials have been published in the literature during the last years (see good reviews [1,2]). In the present paper the physical mechanisms of radiation swelling of fusion ceramic materials are investigated. For this aim a theoretical model of radiation swelling in ceramic materials is suggested taking into account the charge state of point defects and the effect of internal electric field formed under irradiation on diffusion processes of point defects in ceramic materials. The obtained theoretical results for radiation swelling are compared with recent experimental data for radiation swelling in SiC [3,4].

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2. Physical model for low-temperature radiation swelling

Let us consider the radiation swelling mechanisms in irradiated ceramic two-component materials (like SiC, MgO) using our previous model [5]. Irradiation of ceramic materials by fast particles (ions, electrons and neutrons) produces in them point defects (interstitials and vacancies). These point defects 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 charged point defects can recombine, accumulate in the matrix or can be captured by existing dislocations or point defect clusters (dislocation loops, voids). Each point defect creates in the crystal lattice an elastic strain field, these strains lead to the overall volume increase of irradiated material by $e_\alpha\omega$ [6], where e_α is the dilatation of point defect type α ($\alpha = \mathrm{I}$ for interstitials and $\alpha = \mathrm{V}$ for vacancies) and ω is the atomic volume. The absorption of point defect type α on a sink of type s (dislocation loop, void et al.) results in the change of sample volume by the value $e_{\alpha s}\omega$. Thus, if the total number of point defects of the type α absorbed by sinks of the type s in a unit volume is equal to $n_{\alpha s}$ and the concentrations of free point defects are equal to $C_{\alpha 1}$ and $C_{\alpha 2}$ for the components: $k = 1$ and $k = 2$ respectively ($k = 1 = \mathrm{Si}$, $k = 2 = C$) in two-component material (SiC), then the total radiation swelling (S_{tot}) at some irradiation dose can be written in the following form:

$$S_{\mathrm{tot}} = \sum_{K}^{2} C_{\mathrm{I}K}e_{\mathrm{I}K} + \sum_{K}^{2}(C_{\mathrm{V}K} - C_{\mathrm{V}K}^{0})e_{\mathrm{V}K} + \omega\sum_{S,K}(n_{\mathrm{I}K}^{\mathrm{S}}e_{\mathrm{I}K} + n_{\mathrm{V}K}^{\mathrm{S}}e_{\mathrm{V}K}). \tag{1}$$

Here $C_{\mathrm{V}K}^{0}$ is the thermal vacancy concentration of components ($k = 1$ and $k = 2$) and summation is over all different sink types (dislocations, dislocation loops and voids). For the calculation of the dilatation of free point defects in the ceramic materials we have to use the computer simulation of equilibrium configurations of point defects in the crystal lattice with the real interatomic potentials for this material. In our further calculations of radiation swelling of ceramic materials we will use the values for dilatations of point defects which are close to analogous values for the metals and are equal to $e_{\mathrm{I}K} = 1.1$–2.0 and $e_{\mathrm{V}K} = -(0.1$–$0.2)$. The absorption of an interstitial atom and a vacancy by a dislocation network ($s = \mathrm{D}$) and dislocation loop ($s = \mathrm{L}$) results in the volume change on the values ω and $-\omega$, respectively, so we will use $e_{\mathrm{I}K}^{\mathrm{D}} = e_{\mathrm{I}K}^{\mathrm{L}} = 1$ and $e_{\mathrm{V}K}^{\mathrm{D}} = e_{\mathrm{V}K}^{\mathrm{L}} = -1$.

Here we will investigate the physical mechanisms of radiation swelling in ceramic materials at some irradiation doses and temperatures when the crystal lattice contains only the dislocation network with the dislocation density ρ_{d} and interstitial dislocation loops with the density N_{L} formed under irradiation, whose concentration saturates to the maximum value during the nucleation stage. These crystal defects are considered only as sinks for point defects of two components ($k = 1$ and $k = 2$). According to the experimental TEM data for irradiated SiC samples such consideration without vacancy void formation under irradiation takes place at irradiation temperatures $T \leqslant 1100$ K [7]. It should remarked that the dislocation loop formation without voids has been observed also at $T \leqslant 1070$ K in Mg–$\mathrm{Al_2O_3}$ under ion and electron irradiation [8].

Then 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,

$$C_{\mathrm{I}K} + \omega(n_{\mathrm{I}K}^{\mathrm{D}} + n_{\mathrm{I}K}^{\mathrm{L}}) = C_{\mathrm{V}K} - C_{\mathrm{V}K}^{0} + \omega(n_{\mathrm{V}K}^{\mathrm{D}} + n_{\mathrm{V}K}^{\mathrm{L}}), \tag{2}$$

we can rewrite the relation for radiation swelling (1) in the following form:

$$S_{\mathrm{tot}} = \sum_{K}^{2}(C_{\mathrm{V}K} - C_{\mathrm{V}K}^{0})(1 + e_{\mathrm{V}K}). \tag{3}$$

In Eq. (3) we also took into account that interstitial dislocation loops are formed and grow due to the absorption of the mobile interstitial atoms. Due to this process the concentration of free vacancies of component k in the matrix is higher comparing with the interstitial concentration ($C_{\mathrm{V}K} \gg C_{\mathrm{I}K}$).

From Eq. (3) we can see that radiation swelling in ceramic materials is determined by the accumulation of generated vacancies of two components ($k = 1$ and $k = 2$). Let us determine now the main parameters and dependence of radiation swelling in ceramic material (SiC) on the early stage of irradiation using the consideration of the growth kinetics of dislocation loops and the kinetics of point defect accumulations of two types in the matrix.

3. Accumulation of point defects and evolution of microstructure in the matrix of irradiated SiC

Neutron and ion irradiations of SiC produce the displaced atoms (point defects in two components: Si and C) with a strong difference of generation rates [9]. So due to the strong difference of threshold energies (E_{d}) for production of displaced atoms in two subsystems: Si and C in SiC ($E_{\mathrm{d}} = 93$ eV for Si and $E_{\mathrm{d}} = 16$ eV for C) the generation rates of silicon ($G_1 = G_{\mathrm{Si}}$) and carbon ($G_2 = G_{\mathrm{C}}$) in two sublattices have also a strong difference and can vary under neutron and ion irradiation in the interval: $G_C/G_{Si} \approx$ 3–6. It is very important for the description and explanation of physical mechanisms of radiation swelling of SiC because the ceramic materials under irradiation are electrically neutral and point defects have the effective electrical charge.

As we have remarked previously we will consider here the defect microstructure evolution at early stages of irradiation when in the matrix of SiC under irradiation only dislocation loops are formed and the temperature is not so high ($T < 1100$ K) for void formation. It is the typical situation for the evolution of a defect microstructure in ceramic materials after the onset of irradiation [8,10,11]. In this case the point defect sinks are only the dislocation network and produced dislocation loops. The concentration of produced vacancies will be changed mostly due to the recombination with mobile interstitials and absorption by line dislocations and dislocation loops. The ensemble of dislocation loops is characterized by the average loop radius R_L and volume density of loops N_L. The processes of absorption of charged point defects by dislocations and dislocation loops have their special peculiarities. In our model the interstitial dislocation loops and straight edge dislocations are considered as the addition planes consisted from two types of charged atoms: anions and cations. So due to Coulomb repulsion in dielectric ionic materials there is a strong driven force for planar interstitial condensation on a dislocation loop and dislocation line to remain stoichiometric, or at least to balance anion and cation charges. So we have to require equality of the normal components of anionic and cationic interstitial and vacancy currents across the surface of dislocation core with radius r_0: $j^n_{I1} = j^n_{I2}$ and $j^n_{V1} = j^n_{V2}$ at $r = r_0$. The currents of kth component point defects for network dislocations and dislocation loops have a very small difference and consequently we will use the same value for them.

The kinetic equations describing the accumulation of charged kth component point defects under irradiation in the matrix for two-component ceramic material (SiC) can be written in the following form:

$$\frac{dC_{VK}}{dt} = G_{VK} - j_{VK}(\rho_D + \rho_L) - \alpha D_{IK} C_{IK} C_{VK} \quad (k = 1, 2), \tag{4}$$

$$\frac{dC_{IK}}{dt} = G_{IK} - j_{IK}(\rho_D + \rho_L) - \alpha D_{IK} C_{IK} C_{VK} = 0 \quad (k = 1, 2). \tag{5}$$

Here ρ_D is the network dislocation density, ρ_L is the dislocation density of dislocation loops ($\rho_L = 2\pi R_L N_L$); G_{VK}, G_{IK} are the generation rates of kth component vacancies and interstitials ($G_{V1} = G_{I1} = G_{Si}$, $G_{V2} = G_{I2} = G_C$), α is the point defect recombination coefficient ($\alpha = 4/a^2$, a is the lattice spacing).

The growth kinetics of dislocation loops and dislocation climb in ceramic materials with charged point defects have been considered separately [12] and it was shown that the total current of charged kth component point defects on the dislocation line can be written in the following form:

$$\begin{aligned} J_{IK} &= 2\pi R \cdot 2\pi r_0 j^n_{IK} \\ &= 2\pi R \frac{2\pi r_0}{\ln\left(\frac{8R}{r_0}\right)} \frac{D_{I1} C_{I1} D_{I2} C_{I2}}{D_{I1} C_{I1} + D_{I2} C_{I2}}. \end{aligned} \tag{6}$$

The growth rate of dislocation loops in ceramic materials taking into account the absorption of two types of interstitial atoms and vacancies and the remaining of two stoichiometric components in dislocation loops is given by the following relation:

$$\begin{aligned} \frac{dR_L}{dt} &= \frac{4\pi r_0}{b}(j^n_{I1} - j^n_{V1}) \\ &= \frac{4\pi}{b \ln\left(\frac{8R}{r_0}\right)} \left[\frac{D_{I1} C_{I1} D_{I2} C_{I2}}{D_{I1} C_{I1} + D_{I2} C_{I2}} \right. \\ &\quad \left. - \frac{D_{V1}(C_{V1} - C^0_{V1}) D_{V2}(C_{V2} - C^0_{V2})}{D_{V1}(C_{V1} - C^0_{V1}) + D_{V2}(C_{V2} - C^0_{V2})} \right]. \end{aligned} \tag{7}$$

Here D_{IK}, D_{VK} are the diffusion coefficients of interstitials and vacancies of kth components, b is the Burgers vector.

Table 1
Main parameter values used for numerical calculations of radiation swelling in SiC

$G_1 = G_{Si}$	Point defect generation rate of Si atoms	1×10^{-3} dpa/s
$G_2 = G_C$	Point defect generation rate of C atoms	3×10^{-3} dpa/s
E^{Si}_{mV}	Silicon vacancy migration energy	0.8 eV
E^{C}_{mV}	Carbon vacancy migration energy	0.7 eV
E^{Si}_{mI}	Silicon interstitial migration energy	0.3 eV
E^{C}_{mI}	Carbon interstitial migration energy	0.2 eV
E^{Si}_{FV}	Silicon vacancy formation energy	2.5 eV
E^{C}_{FV}	Carbon vacancy formation energy	2.4 eV
ρ_D	Network dislocation density	10^{10} cm $^{-2}$
$e_{V1} = e_{V2}$	Vacancy dilatation	−0.1
a	Lattice parameter	5.14×10^{-8} cm
$D_{VK} = D^0_{VK} \exp(-E^K_{mV}/T)$,		$D^0_{V1} = D^0_{V2} = 10^{-9}$ cm^2/s

The selfconsistent solution of the system of kinetic Eqs. (3)–(7) with the following initial conditions ($t = 0$):

$$C_{IK}(t=0)=0, \quad C_{VK}(t=0)=C^0_{VK}, \quad R_L(t=0)=a, \tag{8}$$

allows one to find the main parameters of radiation swelling of ceramic material (SiC) under different types of irradiation.

The results of numerical calculations of the system Eqs. (3)–(7) with the initial conditions (8) using the microscopic values for SiC (see Table 1) and comparison with the experimental data [4] are presented in Figs. 1–3. The experimental investigations of radiation swelling of SiC have been performed on the DuET multiple-beam irradiation facility of Kyoto University (see [3,4]). So the time dependence for the average dislocation loop radius at different irradiation temperatures is given in Fig. 1. We can see that the growth rate of the average loop radius has a very strong temperature dependence, and at high temperatures the average loop radius increases very quickly up to the saturated values. The dose dependence of radiation swelling is shown in Fig. 2. This dependence at some characteristic doses has also a saturation of radiation swelling. This behavior is explained by the effect of uncompensated vacancies on radiation swelling. We can see in Fig. 2 that the characteristic doses of swelling saturation are increased with increasing generation rate and decreasing temperature. To obtain the temperature dependence of radiation swelling we do not consider here the nucleation stage for dislocation loops but for the approximation of the temperature dependence of dislocation loop density we will use the following relation which has been used previously for the approximation of the experimental temperature dependence of the dislocation loop density in ceramic materials [12]:

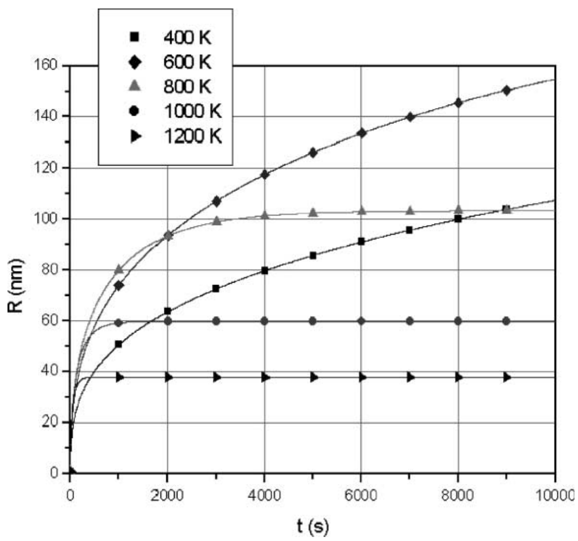

Fig. 1. Time dependence of the average radius of dislocation loops (R) on the growth stage of dislocation loops at different irradiation temperatures.

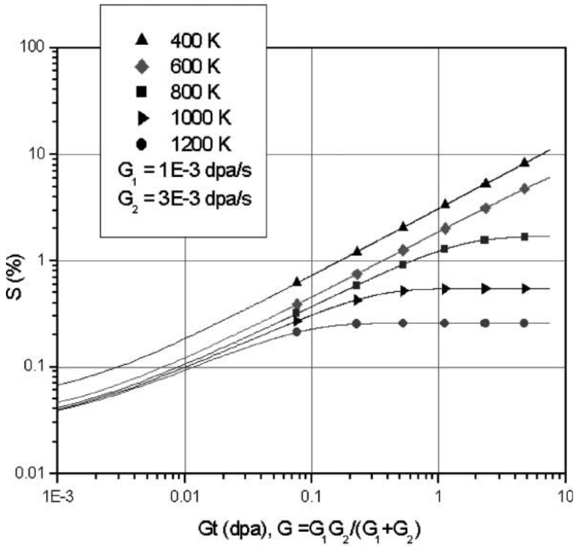

Fig. 2. Radiation swelling of SiC as a function of the displacements per atoms at different irradiation temperatures.

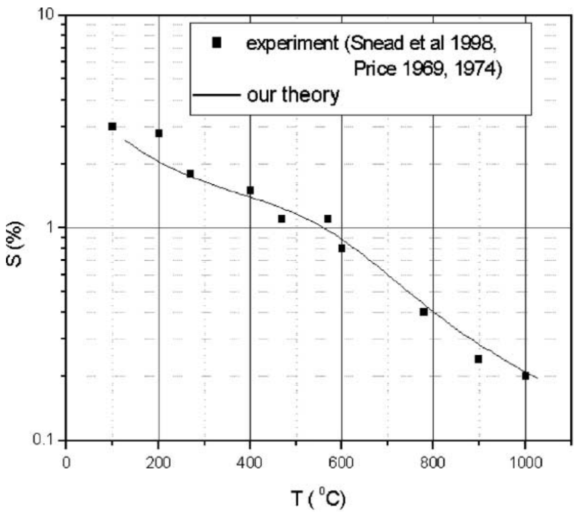

Fig. 3. Comparison of the experimental and theoretical temperature dependence of radiation swelling of SiC.

$$N_L = N^0_L[\exp(E^1_{mI}/T) + \exp(E^2_{mI}/T)]^{1/2}. \tag{9}$$

For SiC the value $N^0_L = 8.0 \times 10^{13}$ cm^{-3} gives the reasonable experimental data for the dislocation loop density. The microscopical values for the energy migration of charged point defects used in the numerical calculations which are comparable with the same values for other ceramic materials and give a good fit to swelling data for SiC are presented in Table 1. The comparison of the experimental data [3,4] with theoretical calculations for the temperature dependence of radiation swelling of SiC is given in Fig. 3. We can see that the temperature dependence strongly decreases with increasing temperature.

4. Conclusions

Based on the obtained recent experimental data and theoretical results we can make the main following final conclusions concerning the radiation swelling behavior in SiC:

- Low-temperature swelling in SiC due to point defect accumulation in the matrix may exceed 1% at temperatures below 673 K.
- Low-temperature swelling in SiC saturates in helium free conditions. The saturation behavior is highly predictable using the suggested theoretical model. Saturation of radiation swelling in SiC is determined by the growth rate saturation of interstitial dislocation loops.
- The recent experimental data clearly demonstrate the strong monotonous decrease of radiation swelling in SiC up to 1200 °C under neutron irradiation. The theoretical calculations give the same temperature dependence for the decrease of radiation swelling in SiC with temperature increase.

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