ANALYZING IRRADIATION-INDUCED CREEP OF SILICON CARBIDE—Y. Katoh, L. L. Snead, and S. Golubov (Oak Ridge National Laboratory)

OBJECTIVE

The objectives of this work were to develop simple models of transient irradiation creep and swelling of silicon carbide, to examine adequacy of the models, and to help defining specific priority objectives of future experiments, in support of resolving critical design issues for silicon carbide-based materials for flow channel insert and other fusion applications.

SUMMARY

Irradiation creep, which is among the major lifetime-limiting mechanisms for nuclear structural materials, is stress-driven anisotropic plastic deformation occurring in excess of thermal creep deformation in radiation environments. In this work, experimental irradiation creep data for beta-phase silicon carbide (SiC) irradiated at intermediate temperatures is analyzed using a rate theory model with an assumed linear-coupling of creep strain rate with the rate of self interstitial atom (SIA) absorption at SIA clusters. The model reasonably explains the experimentally observed time-dependent creep rate of ion-irradiated SiC and swelling evolution of ion- and neutron-irradiated SiC. Bend stress relaxation behavior during irradiation was then simulated using the developed model to confirm agreement with the experimental data obtained by neutron irradiation experiments. Recommended directions of future experiment are provided to further verify and improve the models and assumptions in this work.

PROGRESS AND STATUS

Introduction

Creep deformation and rupture are major lifetime-limiting mechanisms for high temperature materials for structural applications. In nuclear environments, irradiation-induced creep is additive to thermally activated creep deformation [1]. Irradiation creep is anisotropic plastic deformation driven by external or internal stresses, and is induced or enhanced by irradiation in excess of thermal creep deformation. In many cases, irradiation creep is caused by the preferred nucleation of planar defect clusters and/or preferred absorption of supersaturated point defects at edge dislocations [2]. Because irradiation creep is typically not very sensitive to temperature, irradiation creep tends to dominate at lower temperatures. For structural ceramics and ceramic composites for nuclear applications, irradiation creep is one of the most critical unresolved issues [3]. In this work, limited experimental irradiation creep data for beta-phase silicon carbide (SiC) is analyzed using a rate theory model.

Model Description

In mean-field rate theory of defect evolution in irradiated materials, Frenkel defect concentrations are expressed by following equation.

$$\dot{C}_{i,v} = G_{i,v} - \mu_R D_i C_i C_v - D_{i,v} C_{i,v} \sum S^{i,v} \qquad (1)$$

where G is the generation rate, μ_R is the recombination coefficient, D is the diffusivity, S is the sink strength of various extended defects, and subscripts i and v denote self interstitial atom (SIA) and vacancy, respectively. Recombination coefficient is given by $\mu_R = 4\pi r_R/\Omega$, where r_R is the recombination radius and Ω is the atomic volume. Equation (1) assumes $D_i >> D_v$. In SiC, vacancies are practically immobile below ~800ºC, as reported by various fundamental experiments [4,5]. Recent ab-initio calculation studies report minimum migration energies of ~3.5 eV and ~2.4 eV for C and Si vacancies, supporting the extremely limited vacancy mobility at <~800ºC [6]. For intermediate temperature, at which only SIAs are practically mobile, Eq. (1) can be rewritten as

$$\dot{C}_i = G - \mu_R D_i C_i C_v - D_i C_i \sum S_i \tag{2}$$

$$\dot{C}_v = G - \mu_R D_i C_i C_v \tag{3}$$

where $G = G_i = G_v$.

Irradiation-produced defects observed in SiC by transmission electron microscopy after irradiation at <~1000ºC are tiny unidentified defect clusters and dislocation loops. Both types of clusters are believed to be of interstitial-type, because of the very limited mobility of vacancies. For simplicity, all the defect clusters are treated as Frank dislocation loops of interstitial-type here, and the loop number density is assumed to be the temperature-dependent constant. Now, because thermal dissociation of SIA clusters may be inhibited due to very high SIA formation energy (>~6 eV [7]) in SiC, the derivative of the average loop radius rl is determined only by SIA influx and is given by:

$$\dot{r}_l = D_i C_i / b \tag{4}$$

where b is the magnitude of Burgers vector. Additionally, microstructural sinks other than SIA clusters are ignored, because typical mean inter-spacing of SIA clusters are <~10 nm, which is more than one order smaller than typical subgrain sizes in chemically vapor-deposited SiC. Thus, total sink strength for freely migrating SIAs is given as:

$$\sum S_i \approx S_l^i = 2\pi Z_l^i r_l N_l \tag{5}$$

where Z^i_l, taken as 1 in this work, and N_l are loop-interstitial bias factor and loop number density, respectively.

Mechanisms commonly used to explain irradiation creep phenomena of metallic materials are stress-induced preferred loop nucleation or alignment (SIPN), stress-induced preferred absorption of SIAs at dislocations (SIPA), and preferred absorption-enabled glide (PAG) [2,8]. Both SIPN and SIPA require only SIA clusters and migrating SIAs to operate, hence are potential operating mechanisms in SiC. Because there are no gliding dislocations observed in SiC irradiated at relatively low temperatures, PAG can not explain irradiation creep in SiC. Models of SIPN and SIPA both predict linear stress dependence of creep rates for metallic materials, which is in accordance with most experimental observations for metals [2].

In SiC irradiated at <~1000ºC, configurations of SIA clusters have not been characterized sufficiently to discuss kinetic models of irradiation creep [9]. Moreover, because the growth rate of tiny SIA clusters are so slow (cluster sizes remain a few nanometers at 7.7 dpa and 800ºC), it is very difficult to differentiate SIPN and SIPA even if either are the sole operators. However, for either mechanism, the creep rate is believed to be proportional to the total SIA influx to SIA clusters, thus

$$\dot{\varepsilon}_{ic} / \sigma = k_{ic} D_i C_i S_l^i \tag{6}$$

assuming linear stress dependency is maintained, and k_{ic} is a coupling coefficient for creep rate and SIA absorption at SIA clusters.

Analytical Solution

Minimum migration energies for C and Si SIAs in cubic SiC were estimated to be ~0.5 and ~1.4 eV, respectively, by a recent *ab-initio* calculation [6]. Because of the expected high diffusivities (relative to evolutions of other types of defects), the SIA concentration can be treated as in pseudo-equilibrium with

vacancies and other sinks. The C and Si Frenkel defects do not have to be differentiated with this pseudo-equilibrium treatment and vacancy immobility, when antisite defects are neglected. During most of the time of interest (>~10^{-3} dpa), $\alpha C_v >> D_i S_l$ is true, then

$$C_i \approx G/\mu_R D_i C_v \tag{7}$$

$$\dot{C}_V \approx S_l^i G/\mu_R C_v \tag{8}$$

From Eqs. (4), (5), (7), and (8), the following relations are derived.

$$C_i \approx \frac{1}{D_i}\left(\frac{Gb}{9\pi N_l \mu_R t^2}\right)^{1/3} \tag{9}$$

$$C_V \approx \left(\frac{3Gt}{\mu_R}\right)^{2/3}\left(\frac{\pi N_l}{b}\right)^{1/3} \tag{10}$$

$$r_l \approx \left(\frac{3Gt}{\mu_R \pi N_l b^2}\right)^{1/3} \tag{11}$$

The approximated analytical result presented here is consistent with the general analytical solution for the kinetics of low-temperature irradiation processes in metals provided by Golubov [10]. With the analysis presented here, Eqs. (6), (9), and (11) indicate that the irradiation creep rate is proportional to the -1/3 power of time or dose. Additionally, if low temperature swelling of SiC is assumed to be accounted solely by vacancies, Eqs. (10) and (11) imply that irradiation creep rate is in inverse proportion to the square root of swelling. This also suggests that, under a constant stress, a linear compliance that explicitly couples irradiation creep strain and swelling, exists.

Computational Analysis and Comparison with Experiments

In Fig. 1, calculated evolution of SIA absorption rate at SIA clusters, $D_i C_i S^l_i$, is presented. Parameters used for this calculation are listed in Table. 1. The -2/3 power relationship is achieved at a dose of ~10^{-3} dpa, approximately when a recombination-dominant condition ($\alpha C_v >> D_i S^l_i$) is established, and continues until onset of the saturation behavior. A dose of ~10^{-3} dpa is also typical before which SIA cluster nucleation through a homogeneous nucleation process nears completion. The calculation here incorporated cascade re-solution terms for SIA cluster growth and SIA production, which enables the saturation behavior of defect accumulation that is widely observed in experiments. The effect of cascade re-solution becomes apparent between 0.1 and 1 dpa in this calculation, as shown in Fig. 1. Equations (2)-(4) were slightly modified to take the effect of stoichiometric constraint for SIA clusters into consideration. Details of computational method will be published elsewhere [11].

Table 1. Parameters used in calculation

Parameter (*Symbol*)	Value	Unit	Reference
Recombination radius (r_R)	7.5×10^{-10}	m	[12]
Diffusivity pre-exponential for SIA (D^0_i)	1.58×10^{-6}	m^2/s	[12]
SIA migration energy (E^m_i)	0.5	eV	[6]
SIA loop number density (N_l)	2.2×10^{25} x exp(-0.00404 T)	m^{-3}	[9]
Displacement rate (G)	1×10^{-6}, 1×10^{-3}, 3×10^{-6} (n, Si^{2+}, D^+)	dpa/s	-
Fraction of Si displacement to total displacement (f_{Si})	0.24, 0.32, 0.44 (n, Si^{2+}, D^+)	-	[13]

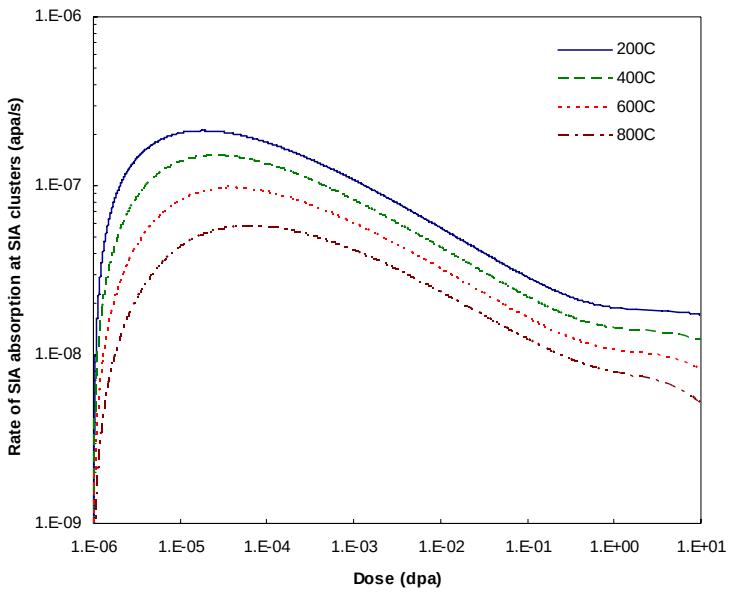

Fig. 1. Calculated dose-dependent evolution of rate of SIA absorption at SIA clusters in beta-SiC at intermediate temperatures.

In Fig. 2, swelling in high-purity, chemically vapor-deposited SiC during Si ion irradiation [14] is plotted against dose. It is seen that the magnitude of swelling approximately follows the 2/3 power law in a broad temperature range below ~800ºC until saturation occurs (<~0.3 dpa). In Fig. 3, dose dependence of swelling in the identical material during neutron irradiation in HFIR [15] is plotted. It is shown that the 2/3 power law also fits the neutron data reasonably at relatively low doses (<~0.1 dpa). These observations support adequacy of the present defect accumulation model.

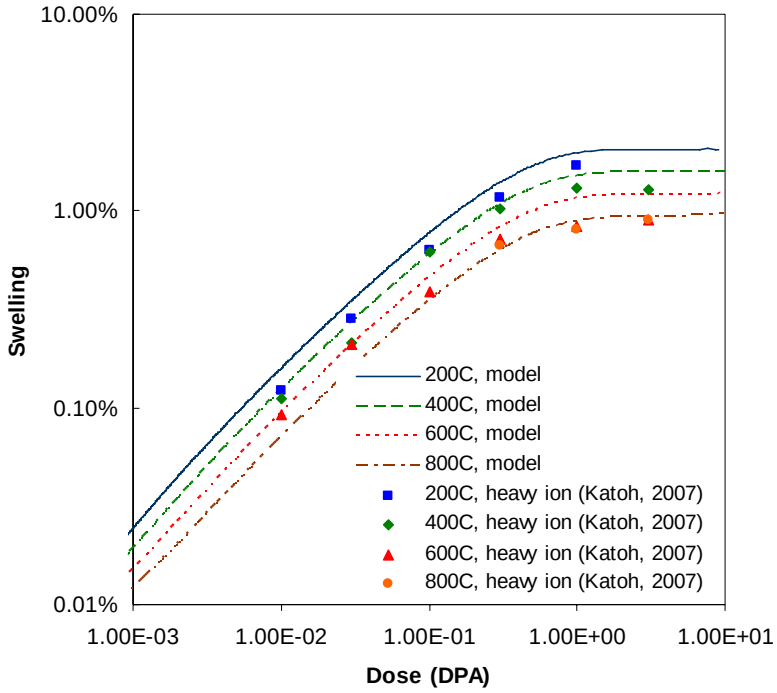

Fig. 2. Dose dependent evolution of swelling in high-purity beta-SiC during heavy ion irradiation: comparison of model and experiment. Experimental data by Katoh [14].

In Fig. 4, calculated irradiation creep strains in SiC during 14 MeV deuteron irradiation are presented as a function of dose, along with experimental data replotted from the torsional irradiation creep data for SCS-6 CVD SiC fibers by Scholz [16]. The creep strain appears to be proportional to approximately 2/3 power of dose, which is in agreement with the analytical model prediction. In this calculation, the coupling coefficient of creep rate with SIA absorption rate at SIA clusters, kic in Eq. (6), was taken to be $3x10^{-3}$ MPa^{-1} to fit the experimental data, when diffusivity Di is normalized to dpa. Fig. 4 also shows that the relative strains at different irradiation temperatures are in good agreement between experiment and calculation, which further support the model. The discrepancy between experiment and calculation at 800ºC may be due to non-linear stress dependence or slightly anomalous creep behavior recorded only at this temperature in Scholz work.

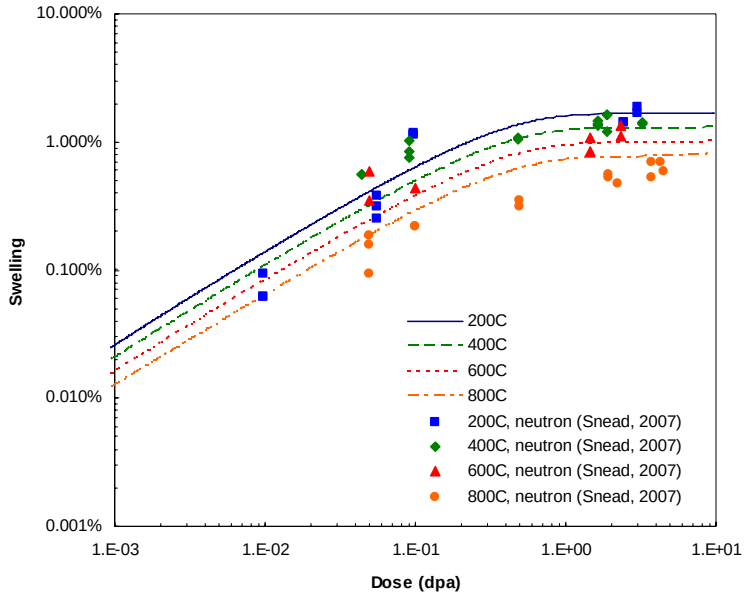

Fig. 3. Dose dependence of swelling in high-purity beta-SiC during neutron irradiation: comparison of model and experiment. Experimental data by Snead [15].

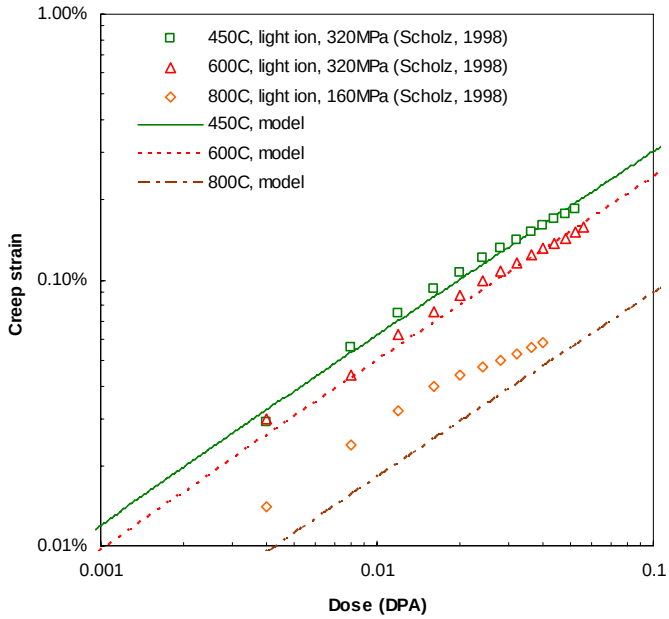

Fig. 4. Dose dependence of creep strain in beta-SiC during light ion irradiation: comparison of model and experiment. Experimental data by Scholz [16].

Analyzing Neutron Irradiation Creep Data

In a companion paper, bend stress relaxation (BSR) irradiation creep experiment was performed using thin strip specimens of high purity CVD SiC [17]. Detailed descriptions of experimental technique development and results are found elsewhere [18]. In Table 2, BSR irradiation creep data for CVD SiC are summarized. Additionally, result from an experiment by Price [19] was interpreted and included. These data are plotted in Fig. 5, in which calculated bend stress relaxation behavior of SiC based on the model described by Eqs. (2)-(6). The coupling coefficient kic was taken to be 3.5×10^{-4} MPa^{-1} to fit the experimental data for the intermediate temperatures (below 800ºC). The fitted coupling coefficient was approximately one order smaller for the neutron-irradiated high purity CVD SiC than for the deuteron-irradiated SCS-6. This difference may be due to the effect of displacement cascade for neutron irradiation, difference in loading mode (flexural in the neutron irradiation vs. torsional shear in the deuteron irradiation), difference in absolute stress levels coupled with potentially non-linear stress dependence of creep rate, and/or difference in material quality. On the other hand, a constant irradiation creep compliance reasonably explain the experimental result at high temperatures (>1030ºC). Obviously, more comprehensive sets of experimental data will be necessary to further examine adequacy of the models and various assumptions involved.

Table 2. Summary of neutron-irradiation creep data

Material	**T_{irr} °C**	**Fluence dpa[1]**	**Reactor**	**Initial / final bend stress[2], MPa**	**Creep strain x10^{-4}**	**BSR ratio[3] *m***	**Ref.**
	640	3.7		87 / 36	1.12	0.42 (+0.03/-0.04)	
Rohm&Haas	700	0.7	HFIR[4]	102 / 72	0.63	0.72 (+0.04/-0.06)	[18]
CVD SiC	1030	0.7		86 / 61	0.52	0.73 (+0.04/-0.06)	
	1080	4.2		101 / 8	2.10	0.08 (+0.05/-0.06)	
	400	0.6		82 / 60	0.41	0.77 (+0.11/-0.19)	
Rohm&Haas	600	0.2	JMTR[5]	81 / 57	0.49	0.73 (+0.10/-0.14)	[18]
CVD SiC	600	0.6		81 / 46	0.75	0.58 (+0.10/-0.15)	
	750	0.6		80 / 55	0.53	0.71 (+0.11/-0.15)	
	780	7.7				0.36 (±0.03) [8]	
GA CVD SiC	950	7.7	ETR[6]	n/a[7]	n/a	0.73 (+0.04/-0.06) [8]	[19]
	1130	7.7				0.08 (+0.05/-0.08) [8]	

[1]Displacement per atom. [2]Maximum stress in a flexurally deformed sample. [3]Ratio of final-to-initial stresses. [4]High Flux Isotope Reactor, Oak Ridge National Laboratory. [5]Japan Materials Test Reactor, Japan Atomic Energy Agency. [6]Engineering Test Reactor, Oak Ridge National Laboratory. [7]Not available. [8]Calculated from linear-averaged creep compliance data.

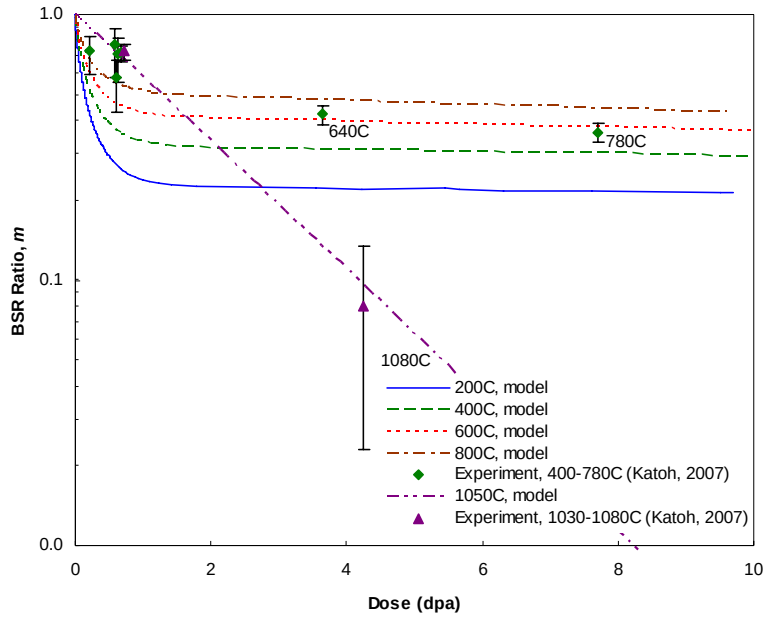

Fig. 5. Dose-dependent evolution of irradiation creep BSR ratio, m, of beta-SiC during neutron irradiation: comparison of model and experiment. Experimental data by Katoh [18] and Price [19].

Conclusions

Simple models of irradiation-induced dimensional instability of silicon carbide at intermediate temperatures were developed based on assumptions of linear-coupling between creep rate and SIA absorption rate at SIA clusters, as well as between swelling and matrix vacancy concentration. The model predicted that both swelling rate and creep compliance are proportional to the -1/3 power of dose under constant temperature and dose rate, until saturation swelling takes place. These predictions are consistent with published experimental data for beta-phase CVD SiC on swelling under neutron and self ion irradiation and on creep under light ion irradiation. Moreover, BSR irradiation creep behavior was simulated using the developed model to confirm a reasonable agreement with the experimental data obtained by neutron irradiation experiments.

To further confirm adequacy of the models and assumptions, and to narrow the ranges of uncertain parameters, it is essential to add credible experimental data on both irradiation creep and swelling, particularly at different temperatures and dose levels within an intermediate temperature (~200 - ~800ºC) / low (~1 dpa)-to-intermediate (~10 dpa) dose regime. It is also necessary that the stress dependence of irradiation creep rate is determined. Moreover, to understand the physical processes of irradiation creep in SiC, configurations of defect clusters and the influence of stress on microstructural development need to be further clarified. Result of this analysis will be utilized to design experimental matrices for future irradiation creep study on silicon carbide presently being planned in US and international fusion advanced materials programs.

References

[1] K. Ehrlich, J. Nucl. Mater. 100 (1981) 149–166.
[2] J. R. Matthews and M. W. Finnis, J. Nucl. Mater. 159 (1988) 257–285.
[3] Y. Katoh, L. L. Snead, C. H. Henager, A. Hasegawa, A. Kohyama, B. Riccardi, and J. B. J. Hegeman, Journal of Nuclear Materials (accepted for publication).
[4] H. Itoh, N. Hayakawa, I. Nashiyama, and E. Sakuma, J. Appl. Phys. 66 (1989) 4529–4531.
[5] A. Kawasuso, H. Itoh, N. Morishita, T. Ohshima, I. Nashiyama, S. Okada, H. Okumura, and S. Yoshida,

Applied Physics A 67 (1998) 209–212.
[6] M. Bockstedte, A. Mattausch, and O. Pankratov, Phys. Rev. B 68 (2003) 205201-1-17.
[7] F. Gao, E. J. Bylaska, W. J. Weber, and L. R. Corrales, Phys. Rev. B 64 (2001) 245208-1-7.
[8] L. K. Mansur, Philos. Mag. A 39 (1979) 497–506.
[9] Y. Katoh, N. Hashimoto, S. Kondo, L. L. Snead, and A. Kohyama, J. Nucl. Mater. 351 (2006) 228–240.
[10] S. I. Golubov, Phys. Met. Metall. 60 (1985) 7–13.
[11] Y. Katoh (to be submitted).
[12] R. A. Johnson, J. Nucl. Mater. 75 (1978) 77–84.
[13] F. Gao and W. J. Weber, Phys. Rev. B 63 (2000) 054101-1-7.
[14] Y. Katoh, H. Kishimoto, and A. Kohyama, J. Nucl. Mater. 307 (2002) 1221–1226.
[15] L. L. Snead, W. Cuddy, D. Peters, Y. Katoh, G. A. Newsome, and A. M. Williams, J. Nucl. Mater. (submitted).
[16] R. Scholz, J. Nucl. Mater. 258–263 (1998) 1533–1539.
[17] Y. Katoh and L. L. Snead, Ceram. Eng. Sci. Proc. 26 (2005) 265–272.
[18] Y. Katoh, L. L. Snead, T. Hinoki, S. Kondo, and A. Kohyama, Journal of Nuclear Materials (accepted for publication).
[19] R. J. Price, Nucl. Tech. 35 (1977) 320–336.