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Journal of Nuclear Materials 307–311 (2002) 1221–1226

journal of nuclear materials

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The influences of irradiation temperature and helium production on the dimensional stability of silicon carbide

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Abstract

Isotropic volume expansion, or swelling, is a well-known irradiation-induced phenomenon for silicon carbide (SiC), as observed after neutron irradiation. In this work, the influences of irradiation temperature and helium production on fluence-dependent swelling behavior in cubic SiC were characterized following the establishment of an experimental technique to determine ion-irradiation-induced swelling within an accuracy of <0.02%. Saturation swelling behavior was confirmed at temperature >200 °C. Measured swelling values yielded approximately at the lower edge of neutron-induced swelling data band at $T \sim< 600$ °C. A fusion-relevant helium-to-dpa condition significantly enhanced swelling at $400 <\sim T <\sim 800$ °C. The temperature dependence of saturated swelling both with and without helium co-implantation suggested a transient in defect behavior between 800 and 1000 °C. The surviving defect production efficiency in heavy-ion irradiated SiC at 333 K was very roughly estimated to be 20% from low-dose swelling data.

1. Introduction

Silicon carbide (SiC) and its composites are promising structural and functional materials for fusion power reactors, owing to the superior high-temperature properties, thermo-chemical stability, irradiation tolerance, inherent low-activation and low-after heat properties [1]. Recent development of high performance radiation-resistant SiC fiber-reinforced SiC matrix composites (SiC/SiC composites) is further enhancing the attractiveness of SiC-based materials for fusion and other advanced energy system applications [2,3].

Technical issues for SiC relevant to nuclear environments include dimensional instability, thermal and electrical transport property changes, and hardness and fracture toughness changes [4]. Volume expansion, or swelling, is a well-known irradiation-induced phenomenon for SiC, as observed after neutron irradiation [5]. Swelling potentially determines the low-temperature limit (point defect swelling) and high-temperature life time (cavity swelling) of the design window of SiC-based blanket structures [6]. However, existing neutron-induced swelling data suffer from significant scatter, in addition to the fact that they do not provide sufficient information on fluence-dependent swelling development [5]. As for the high-temperature swelling in SiC, reported data are inconsistent and of extremely limited quantity. Moreover, the influence of the transmutant helium production in anticipated fusion conditions on swelling behavior remains almost unexplored.

In this work, an attempt was made to characterize the influences of irradiation temperature within a broad temperature range and transmutant helium production on dose-dependent swelling behavior of SiC by taking advantage of the excellent irradiation condition controllability in an ion-bombardment technique.

2. Experimental

The materials used were high purity (>99.9995%) polycrystalline 3C(beta)–SiC produced through chemical

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PII: S0022-3115(02)01062-0

vapor deposition (CVD) process (Rohm and Haas Co., Woburn, USA) [7]. This material has a mass density of 3.21 Mg/m^3, which is exactly equal to the theoretical density of 3C–SiC.

The ion beam irradiation was carried out mostly at the DuET Facility, Institute of Advanced Energy, Kyoto University [8]. 5.1 MeV Si^{2+} ions or 4.0 MeV Ni^{3+} ions were used to introduce displacement damage in a 'single-beam' experiment, while an additional beam of energy-degraded 1.0 MeV He$^+$ ions were simultaneously implanted in a 'dual-beam' experiment for the helium effects study. Rates of displacement damage production and helium implantation were calculated using TRIM-92 code assuming a sublattice-averaged displacement threshold energy of 35 eV [9]. The damage level was 0.01–3.0 dpa in most cases, and the irradiation temperature was 333–1673 K. The displacement damage rate was in a range of $0.4 \sim 1.0 \times 10^{-3}$ dpa/s. The accuracy of ion fluence determination is believed to be within 5% considering factors that might contribute to it. The helium-to-dpa ratio was kept constant at 60 appm He/dpa in all the dual-beam experiments. The irradiation temperature was maintained typically within ±10 K of the nominal ones.

The degree of swelling was determined by means of a precision surface profilometry following irradiation through molybdenum meshes. The measured linear swelling was assumed to be identical with a volumetric swelling, based on the fact that the irradiated volumes are as thin as a few microns on the specimen surfaces. Dual-beam-induced swelling was calculated as a sum of swelling in a single-beam case at the same dose and the additional volume expansion in the helium-co-implanted volume. The surface height change as a consequence of irradiation-induced volume expansion was measured with the Micromap™ interferometric optical surface profiling system (Micromap Inc., Tucson, USA). The practical resolution in the profiling is determined by the surface smoothness of the object. The standard experimental procedure is described elsewhere [10].

Fig. 1 shows the calculated depth-profiles of displacement damage and stopped ions in SiC for the case of 4 MeV Ni irradiation. A nominal damage level was defined as the dpa averaged over the damage range. According to the calculation in Fig. 1, the estimated contribution of the ion deposition to the surface height increase is 0.45 nm/dpa, assuming unchanged atomic density due to the chemical effect of implantation. On the other hand, the estimated sputtering rate of the irradiated surface is ~0.1 nm/dpa. Therefore, because of the potential phase instability and atomic density change associated with ion implantation, the ion deposition is the primary factor that imposes potential uncertainty to the swelling determination in high-dose cases.

3. Results and discussion

The fluence dependence of swelling within a fluence range of 0.01–3 dpa at 333–1673 K is plotted in Fig. 2 for both single- and dual-beam irradiation cases. After irradiation at 333 K, a step height corresponding to about 0.02% of volume change was the detection limit that was determined by surface roughness. Therefore,

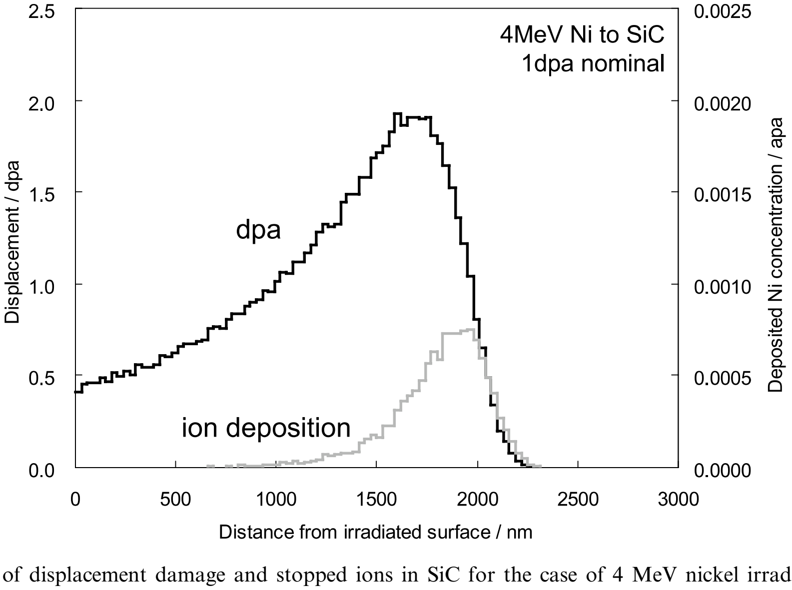

Fig. 1. Depth-profiles of displacement damage and stopped ions in SiC for the case of 4 MeV nickel irradiation. See text for the calculation condition.

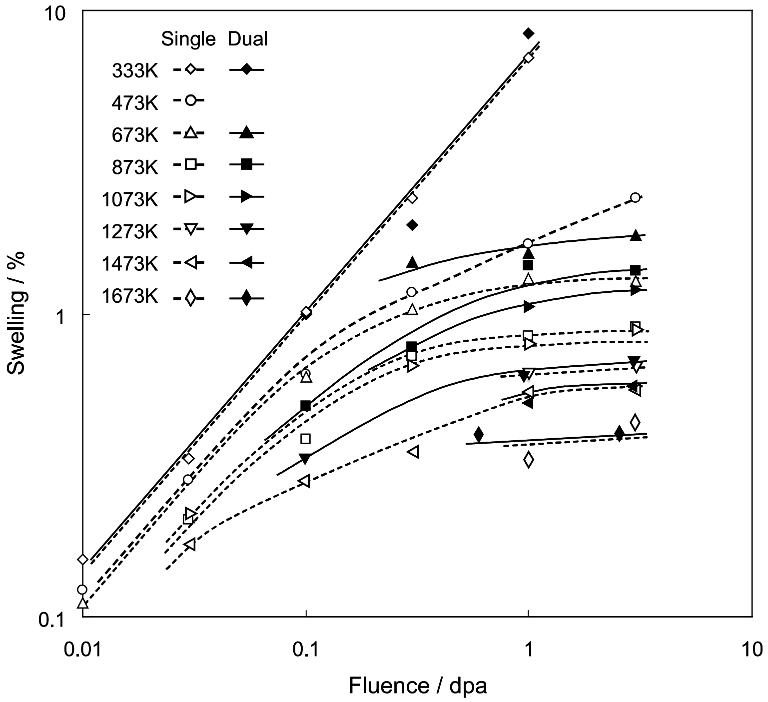

Fig. 2. Fluence-dependence of swelling in SiC in a low-fluence regime under single- and dual-beam ion irradiation.

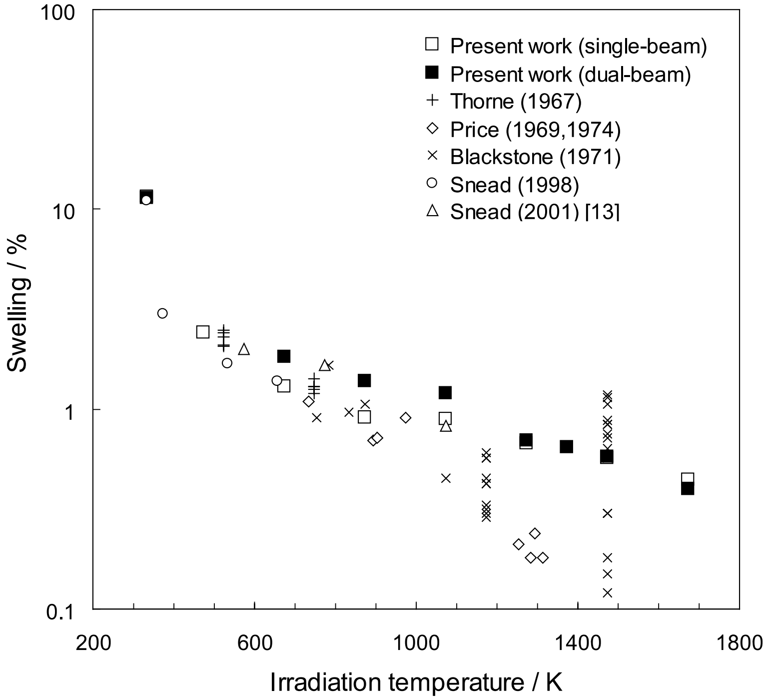

Fig. 3. Swelling in SiC at 3 dpa plotted against irradiation temperature. Neutron data were taken from compilation in reference [12] unless otherwise noted.

relative errors in swelling quantification are approximately 20% and 2% for swelling values of 0.1% and 1%, respectively, at that temperature. The irradiation-induced surface roughening appeared somewhat enhanced at elevated temperatures and approximately doubled at 1673 K compared to that at 333 K.

Swelling rates, and consequently swelling, monotonically decreased with the increasing irradiation temperature. The volume expansion exhibited a tendency toward saturation at temperatures over 673 K. Helium co-implantation notably enhanced swelling at 673–1073 K, while it caused only very slight swelling increase at

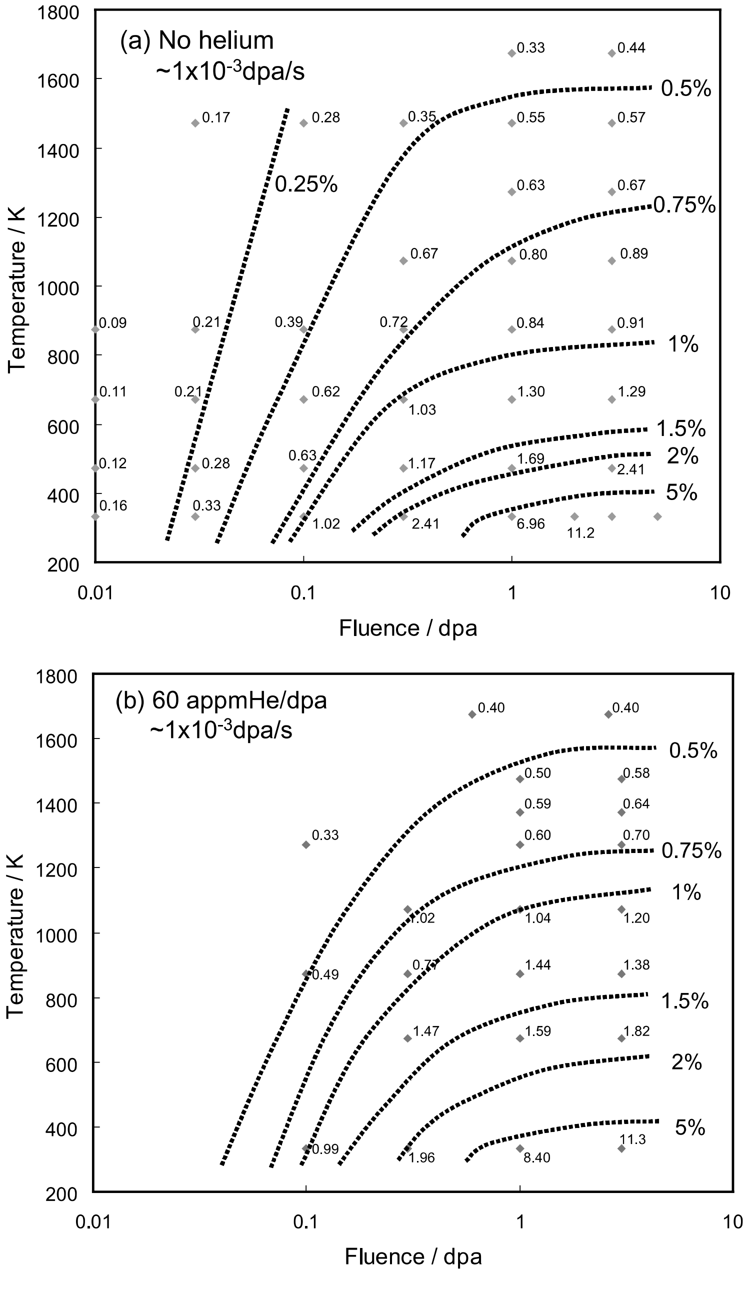

Fig. 4. (a) Contour plots of swelling in SiC by single-beam (a) and dual-beam (b) irradiation as a function of fluence and temperature. Labels for individual data points indicate measured swelling in percent. Note that high-dose rates in the ion irradiation experiment may have resulted in the increased swelling.

higher temperatures. The swelling behavior at 333 K was not apparently influenced by helium.

In Fig. 3, the swelling at 3 dpa is plotted as a function of irradiation temperature, along with reported neutron irradiation data [11,12]. It should be reasonable to regard swelling values at 3 dpa as saturated ones from Fig. 2. The ion irradiation data points for below 673 K fall close to the lower edge of the neutron data band. Considering the outstanding irradiation condition controllability in the current ion irradiation experiment, we suspect that majority of neutron data have been affected by irradiation temperature uncertainty and/or unintentional transient low-temperature neutron exposure that is peculiar to the irradiation in fission reactor cores [13]. At temperatures over 873 K, the ion data start to deviate from the low edge of the neutron data band. This is probably due to the higher equilibrium defect concentration arising from the approximately three orders higher damage rates in the ion irradiation.

Fig. 3 indicates a potential transient of swelling behavior between 1073 and 1273 K. A rather steep decrease in swelling at 1273 K compared to that at 1073 K is also apparent in Fig. 2. At the same time, helium effect on swelling becomes almost negligible at over 1073 K. The former observation can be explained by a mobilization of vacancies in this temperature range, as reported from a variety of thermal recovery experiments [14]. The latter observation indicates that vacancies are not effectively trapped by helium in SiC. Since thermal release of implanted helium occurs at over 1073 K [15], helium-vacancy-cluster (or isolated helium) and vacancy should have similar migration activation barriers. This is also supported by the fact that cavity formation starts at ~1273 K under dual-beam irradiation [16]. The mechanism of swelling enhancement by helium at intermediate temperatures can not be clarified by this work. However, a reduced Frankel pair recombination rate and/or SIA-cluster stabilization by helium are most likely, because probably only isolated SIAs are readily mobile at these temperatures and irregular-shaped SIA-clusters are the only irradiation-produced microstructural features that have been observed by transmission electron microscopy [17].

In Fig. 4(a) and (b), contour plots of swelling values are provided, by single- and dual-beam experiment, respectively, as a function of fluence and irradiation temperature. These figures clearly depicts that, in a fusion-relevant helium/dpa condition, low-temperature application limit of SiC-based materials may have to be raised significantly from that estimated based on fission neutron data, depending on design-specific swelling tolerance.

The fluence dependence of swelling in SiC by irradiation with 4 MeV Ni^{3+} at 333 K is provided in more details in Fig. 5. The volume increase was apparently proportional to the 0.81 power of the fluence until it approached the amorphization threshold dose. Physical significance of such dependency may not be able to be clarified, since complex thermal recovery processes are operating in addition to an uneven damage profile in the

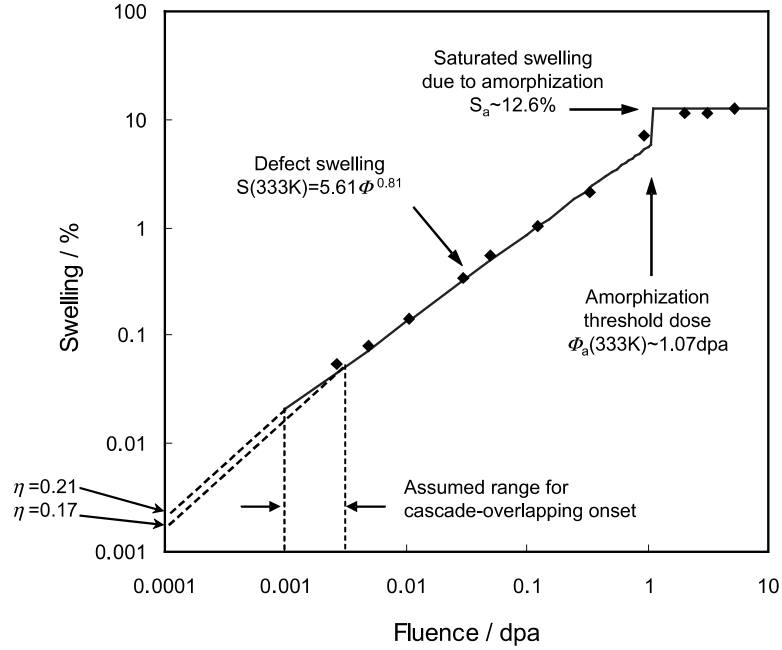

Fig. 5. Fluence-dependence of point-defect swelling in SiC under irradiation with 4 MeV Ni^{3+} at 333 K.

irradiated volume. If one assumes a linear defect accumulation up to the onset of cascade-overlapping at 1–3 mdpa without any dynamic recovery and that each surviving Frenkel pair leaves a volume expansion corresponding to one atomic volume for perfect SiC, as a very rough estimation, a surviving defect fraction to NRT-dpa can be derived to be 17–21%.

A rather rapid volume expansion occurred at slightly over 1 dpa at 333 K, indicating a progressive crystalline-to-amorphous phase transition that initiated at the damage peak range. The swelling due to amorphization was determined to be 12.6% at 3 dpa, assuming a total amorphization of the irradiated volume. This result is in good agreement with the neutron data, where the amorphization-induced swelling in originally full-dense SiC is reported to be $\sim$15% [18]. It may not be determined from the current result whether or not amorphization occurs at 473 K at a damage rate of 1×10^{-3} dpa/s.

4. Conclusions

An experimental technique to determine ion-irradiation-induced swelling within an accuracy of $<0.02\%$ was established. Irradiation-induced surface roughening appeared to be the primary reliability-limiting factor in surface profilometry.

By displacement damage alone, saturated swelling behavior was confirmed at temperatures $>\sim 673$ K. The Measured swelling values yielded approximately at the lower edge of the neutron-induced swelling data band at $T \sim < 873$ K, suggesting potential transient irradiation effects in reactor irradiations. Fusion-relevant helium production significantly enhanced swelling at $673 < \sim T < \sim 1073$ K but did not impose a strong effect at >1273 K. The effect of helium on swelling was not detectable at 333 K. Our results imply that swelling of 1% $\sim$ will have to be allowed for SiC in a fusion blanket, although the amount of swelling needs to be further assessed from a viewpoint of the dose rate effect. A transient in dynamic defect recovery behavior is likely between 1073 and 1273 K, regardless of helium co-implantation. The surviving defect fraction in 4 MeV Ni-irradiated SiC at 333 K was very roughly estimated to be 20%.

Acknowledgements

This work was partly supported by Core Research for Evolutional Science and Technology (CREST), operated by Japan Science and Technology Corporation (JST) for Japanese Ministry of Education, Culture, Sports, Science and Technology.

References

[1] A.R. Raffray et al., Fus. Eng. Des. 55 (2001) 55.
[2] A. Hasegawa, A. Kohyama, R.H. Jones, L.L. Snead, B. Riccardi, P. Fenici, J. Nucl. Mater. 283–287 (2000) 128.
[3] T. Hinoki, L.L. Snead, Y. Katoh, A. Kohyama, The Effect of High Dose/High Temperature Irradiation on High Purity Fibers and Their Silicon Carbide Composites, these Proceedings.
[4] R.H. Jones, L.L. Snead, A. Kohyama, P. Fenici, Fusion Eng. Des. 41 (1998) 15.
[5] L.L. Snead, R.H. Jones, A. Kohyama, P. Fenici, J. Nucl. Mater. 258–263 (1998) 7.
[6] S.J. Zinkle, N.M. Ghoniem, Fus. Eng. Des. 51&52 (2000) 55.
[7] CVD Silicon Carbide, available from <http://www.cvdmaterials.com/silicon.htm>.
[8] A. Kohyama, Y. Katoh, M. Ando, K. Jimbo, Fus. Eng. Des. 51&52 (2000) 789.
[9] J.F. Ziegler, Ion Beam Interactions with Matter, available from <http://www.research.ibm.com/ionbeams/>.
[10] Y. Katoh, H. Kishimoto, A. Kohyama, Mater. Trans. 43 (2002) 612.
[11] S.J.Zinkle, L.L.Snead, report DOE/ER-0313/24 (1998) 93.
[12] L.L. Snead, unpublished data.
[13] M. Kiritani, J. Nucl. Mater. 160 (1988) 135.
[14] H. Itoh, M. Yoshikawa, I. Nashiyama, H. Okamura, S. Misawa, S. Yoshida, J. Appl. Phys. 77 (1995) 837.
[15] A. Hasegawa, M. Saito, S. Nogami, K. Abe, R.H. Jones, H. Takahashi, J. Nucl. Mater. 264 (1999) 355.
[16] H. Kishimoto, Y. Katoh, K.-H. Park, A. Kohyama, Microstructural Stability of SiC and SiC/SiC Composites under High Temperature Irradiation Environment, these Proceedings.
[17] Y. Katoh, A. Kohyama, T. Hinoki, Proceedings of the Eighth European Conference on Composite Materials, vol. 4 (1998) 351.
[18] C.J. McHargue, J.M. Williams, Nucl. Instrum. and Meth. B 80&81 (1993) 889.