High Temperature Radiation Responses of Amorphous SiOC/Crystalline Fe Nanocomposite

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Abstract

The radiation tolerance of amorphous silicon oxycarbide (SiOC) and crystalline Fe nanocomposites were examined by ion irradiation and transmission electron microscopy characterization. A comparison was made between a pure Fe film and SiOC/Fe multilayers. The composites were subjected to 120 keV He⁺ ions to average damage levels from approximately 0.5 to 10.7 displacements per atom (dpa) at 600 °C. Compared to pure Fe films, the swelling resistance in the Fe layers of thick SiOC/Fe (80/60 nm) multilayer films is improved by 2.2 times and the averaged void size is reduced to half. In some instances a crystalline Fe_xSi_yO_z reaction layer formed between the Fe and SiOC components of the composite, and the interface between Fe and Fe_xSi_yO_z was observed to be incoherent. Void denuded zones were observed in the Fe layer close to the SiOC/Fe and Fe/ Fe_xSi_yO_z interfaces. For thin SiOC/Fe (14/14 nm) multilayers, layer breakdown was observed and the extent of the layer breakdown became more significant with increasing dpa values. However, there were no voids in the Fe component of the thin SiOC/Fe nanocomposites. These results suggest that the SiOC/Fe and Fe/Fe_xSi_yO_z interfaces act as efficient defect sinks which promote point defect recombination and suppress void swelling.

Keyword: Radiation tolerant materials, amorphous SiOC, nanocrystalline Fe, interface.

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1. Introduction

Next-generation nuclear energy technology requires the development of radiation tolerant materials for service in extreme irradiation environments [1], [2] and [3]. To suppress radiation damage effects, major efforts have been made to introduce defect sinks such as grain boundaries and interfaces to increase self-healing capabilities [4] and [5]. For example, oxide-dispersion-strengthened (ODS) alloys with interfaces between nanoscale oxides and ferritic/martensitic steels, have shown superior high temperature thermal stability and void swelling resistance [2] and [6]. Also, grain boundaries in austenitic stainless steels have been found to promote defect annihilation and effectively reduce the concentration of voids and void swelling [7], [8] and [9]. In addition, increasing the volume fraction of interfaces in several immiscible metallic nanolayer systems such as Cu/Nb was demonstrated to suppress radiation induced damage [10].

Compared to traditional approaches which strive solely to minimize the effect of radiation damage in crystalline solids, amorphous silicon oxycarbide (SiOC) has been developed recently and has shown very good steady-state irradiation properties [11]. Previous studies have demonstrated that amorphous SiOC alloys sustain their glassy states within a wide envelope of irradiation conditions [11] and [12]. For potential service in extreme irradiation environments, amorphous SiOC can be paired with a crystalline metal component such as Fe to form a composite for improved mechanical properties and better heat conductivity [2] and [13]. Similar to what has been observed in metallic nanolayer composites [10] and [14], the amorphous/crystalline interfaces are expected to be strong sinks for defects. This interface behavior is anticipated because the amorphous structure on the SiOC side of the interface is able to adopt numerous atomic-level conformations and add free volume to the interface, which increase its capacity to trap and relax away conventional crystal defects (such as vacancies and

interstitials) that diffuse to the interface from the crystalline Fe layers. The ability of the amorphous SiOC to conform itself to the atomic-level morphology of the adjacent Fe layer is also expected to enhance mechanical adhesion, giving rise to high interfacial fracture toughness and resilience against dislocations arriving at the interface from the Fe layer.

This amorphous SiOC/crystalline Fe composite is attractive since it exhibits promising thermal and radiation tolerant properties [15] and [13]. For example, a thermally stable intermixed layer ($\text{Fe}_x\text{Si}_y\text{O}_z$) was observed in thick SiOC/Fe multilayers after 600 °C annealing. More interestingly, the thickness of the intermixed layer remains small and does not vary as annealing time goes up, suggesting that the $\text{Fe}_x\text{Si}_y\text{O}_z$ layer acts as a barrier for further Si, Fe, O diffusion and helps to maintain morphological stability. In addition, the amorphous/crystalline interface in SiOC/Fe composites has been shown to effectively remove defects at room temperature, under both light ion (He) and heavy ion (Kr) irradiation [16] and [17].

Although thermal and irradiation stability at room temperature have shown in amorphous SiOC/crystalline Fe system, the composites needs to be tested in a more extreme environment such as a combination of high temperature (>500 °C) and irradiation to evaluate the potential applications in fast reactors. At such temperatures (homologous temperatures 0.3-0.5 T_M), the Fe component cannot thermodynamically support large concentrations of vacancies and irradiation induced defects are sufficiently mobile to diffuse and cluster into voids. Therefore, void swelling represents one of the most important radiation degradation phenomena [18] and [19]. However, the effect of this new amorphous/crystalline SiOC/Fe interface on void swelling and on the layered morphological stability during irradiation at elevated temperatures is largely unknown. In this work, we investigated and compared the high temperature (600 °C) radiation responses of pure Fe films and SiOC/Fe composites with different Fe layer thicknesses

and different doses for the first time. The results serve to better understand and show the role of SiOC/Fe interfaces on defect mitigation out to harsher environments.

2. Experimental

In this study, pure nanocrystalline α -Fe films and SiOC/Fe multilayer films were prepared by magnetron sputtering at room temperature. The base pressure of 9.2×10^{-6} Pa was reached prior to depositions and the typical argon partial pressure during sputtering was ~ 0.65 Pa. DC magnetron sputtering was used to deposit α -Fe layers, while radio frequency (RF) sputtering was used to prepare amorphous SiOC layers from SiO₂ and SiC targets. All multilayered films had a similar total thickness of 420 nm and the thickness of the pure α -Fe film was 180 nm. In thick Fe/SiOC multilayer films, the thickness of Fe and SiOC layers were 60 and 80 nm, respectively, while the individual layer thickness for thin Fe/SiOC multilayer films was 14 nm. A detailed microstructure characterization of as-deposited SiOC/Fe multilayers can be found in reference [16].

The SiOC/Fe multilayers and pure α -Fe films were subjected to 120 keV He ions at 600 °C. The heating ramp rate for the sample stage was 20 °C/min. The total fluences of 4×10^{16} , 2×10^{17} , 5×10^{17} and 8×10^{17} ions/cm² were used to obtain approximately 0.5, 2.7, 6.7 and 10.7 dpa of damage in SiOC/Fe multilayers, respectively. The dose rate was 3.43×10^{13} ions/second with an irradiation area about 1 cm $\times$ 1 cm. The sample temperature was monitored and kept within ± 3 Celsius during the irradiation. Transmission electron microscopy (TEM, FEI Tecnai G2 F20) with an operation voltage of 200 kV was used to examine the microstructure evolution of SiOC/Fe multilayers and pure Fe films before and after He irradiation. Cross-sectional TEM specimens were prepared using conventional dimple and grinding, followed by a low energy (3.7

keV) ion-milling procedure. e. The Stopping and Range of Ions in Matter (SRIM)-2008 was used to simulate depth-dependent damage and defect concentration profiles by choosing the ion distribution and quick calculation of damage option [20]. The displacement energies used for Fe, Si, O and C were 40, 15, 28, 28 eV, respectively [21]. The simulation of the SiOC/Fe multilayers was treated as one amorphous SiOC-Fe layer with average atomic density as 7.8×10^{22} atoms/cm³.

3. Results and discussion

In this study, we selected 120 keV He ion for irradiation because of the needs to minimize He doping and maximize displacement efficiency in the multilayer films. Fig. 1a and 1b present the simulated radiation damage (in units of displacements per atom (dpa)) and helium concentrations as a function of depth in pure Fe, and SiOC/Fe multilayer films with a dose of 2×10^{17} ions/cm², respectively. All results are obtained from the SRIM code [20]. The SRIM simulations, shown as Fig. 1a and 1b, predict that an approximate average of 1.4 dpa in the pure Fe films and 2.7 dpa in the Fe layers of the SiOC/Fe multilayer films is deposited after He⁺ irradiation with a fluence of 2×10^{17} ions/cm². From the depth profiles of the helium and damage distributions (seen as Fig. 1a and 1b), it is seen that damage dominates the nanocrystalline alpha-Fe films because the majority of He ions come to rest in the Si substrate. However, some He is predicted to be close to the film/substrate interface in irradiated SiOC/Fe multilayer films. Therefore, the surface regions were selected for examining irradiation effects because there should be a negligible amount of He in the near surface region (<200 nm).

Fig. 2a shows a typical cross-sectional TEM micrograph of the as-deposited thick SiOC/Fe (80/60 nm) multilayer film, in which alternating Fe and SiOC nanolayers with clear

interfaces are observed. The Fe layers exhibit a typical columnar structure that is associated with sputter deposition with an average grain size of ~15 nm. Compared to the as-deposited thick SiOC/Fe films, significant Fe grain growth and a discontinuous intermixed layer is observed between the Fe and SiOC layers after 600 °C irradiation with a damage level of 2.7 dpa (Fig 2b). The intermixed layer, which consisted of Fe, Si and O, was also observed in samples exposed to pure thermal annealing [22] or irradiation at room temperature [13]. Fig. 2c shows a typical high resolution image of the discontinuous intermixed layer where no intermixed and intermixed ($\text{Fe}_x\text{Si}_y\text{O}_z$) regions are highlighted. It is interesting to observe that the intermixed layer is crystalline and that interface between the Fe layer and the intermixed $\text{Fe}_x\text{Si}_y\text{O}_z$ layer is incoherent (seen as Fig. 2d). In addition to the formation of an intermixed layer and significant Fe grain growth, high temperature irradiation also resulted in void formation in the Fe component of the SiOC/Fe nanocomposite (Fig. 2c). Similar to what is observed in the Fe component of the SiOC/Fe nanocomposite, void formation was also observed in the pure Fe films after irradiation. Fig. 2e and 2f present typical over- and under-focus images from the pure Fe films after 600 °C irradiation. Fresnel contrast is used to confirm the formation of voids which are revealed as bright dots surrounded by a dark fringe in the under-focus condition and dark dots surrounded by a bright fringe for the over-focus condition.

To examine the high temperature irradiation stability of the SiOC/Fe nanocomposite, He^+ irradiations with four exemplary doses (4×10^{16} , 2×10^{17} , 5×10^{17} and 8×10^{17} ions/cm²) were conducted. Fig. 3a and 3c show the typical cross-sectional TEM images from the thick SiOC/Fe multilayer films after 600 °C irradiations with fluences of 2×10^{17} ion/cm² and 8×10^{17} ions/cm², respectively. Only the top SiOC/Fe layers are highlighted in this study where the emphasis is on irradiation damage rather than helium implantation effects. To have a fair comparison with

irradiated thick SiOC/Fe films (Fig. 3a and 3c), the structural evolution of pure Fe films with the same dose is also presented (Fig. 3b and 3d). Although the void density in the pure Fe films is slightly bigger than that in the top Fe layer of the thick SiOC/Fe multilayer sample, the average void size in pure Fe films is two times larger than that in the Fe layer of the thick SiOC/Fe multilayer sample under the same irradiation damage level. In general, there are two characteristic regimes of void swelling: an initial low-swelling transient regime during which void nucleation and initial growth phase exist, and a steady-state swelling regime where the volumetric swelling increases linearly with irradiation dose [19], [23] and [24]. Void swelling examined in this work is mainly in the initial low-swelling transient regime.

The He irradiation processes in Fe results in primarily producing isolated defects (vacancies and interstitials) and small dilute cascades [25]. The irradiation defect population can be reduced by either recombination of interstitials and vacancies or diffusion to defect sinks such as dislocation, grain boundaries, interface, etc. [26]. Void formation and swelling occurs when there is preferential absorption of interstitials at different sinks [27] and [28]. The smaller void size observed in the SiOC/Fe multilayer system suggests that there is a lower concentration of supersaturated vacancies in SiOC/Fe multilayers compared to that in the pure Fe layer during irradiation. This implies that the amorphous SiOC/crystalline Fe interfaces and the new Fe/Fe_xSi_yO_z incoherent interfaces are acting as efficient defects sinks to promote recombination of interstitials and vacancies, thereby extending the low-swelling transient regime of the system. This idea is further supported by the presence of void denuded zones in the Fe layer close to SiOC/Fe and Fe/ Fe_xSi_yO_z interfaces [29] and [30]. Fig. 3e and 3f present scanning transmission electron microscopy (STEM) images of the thick SiOC/Fe multilayer samples after 600 °C irradiation with a fluence of 2×10^{17} ion/cm² and 8×10^{17} ions/cm², respectively. One important

feature of the 600 °C irradiation is that the thickness of the intermixed $\text{Fe}_x\text{Si}_y\text{O}_z$ layer almost remains constant as the irradiation dose and annealing time go up [15], indicating that the intermixed layer acts as a barrier for further Si, Fe, O diffusion.

To summarize the differences discussed above, a statistical analysis of the void size and void swelling in pure Fe films and Fe layers from thick SiOC/Fe multilayer composites as function of damage after 600 °C irradiation has been performed. Figs. 4a and 4b schematically show the regions where the void analysis were carried out on the two different samples. Figs. 4c and 4d show the results of this analysis. Both the void size and void swelling go up monotonic as the radiation dose increases. As shown in Fig. 4c, as the damage level varies from 1 to 8 dpa, the averaged void size in the pure Fe films is approximately twice that as in Fe layers in the composite. Consequently, the calculated void swelling rate in pure Fe films is 0.22%/dpa, compared to ~0.1% in the composite Fe layers.

One key question in this study is whether interfaces in the irradiated SiOC/Fe composite can suppress void swelling. Two interfaces exist in the irradiated composite, amorphous-SiOC/Fe and crystalline- $\text{Fe}_x\text{Si}_y\text{O}_z$ /Fe, where the interface between $\text{Fe}_x\text{Si}_y\text{O}_z$ and Fe is incoherent. For the composite system, void size and density in the Fe layer are smaller than that observed in the pure Fe film. In addition, void denuded zones with widths of ~10 nm are observed in the Fe layer near the interface in the composite (Fig. 3a and 3c). These findings suggest that the concentration of supersaturated vacancies close to the composite interfaces is too low to nucleate and form voids during 600°C irradiation. These data suggest that the SiOC/Fe and Fe/ $\text{Fe}_x\text{Si}_y\text{O}_z$ interfaces are playing a crucial role in promoting the recombination of radiation induced interstitials and vacancies, therefore, extending the low-swelling transient regime in the Fe component.

A typical cross-sectional TEM image of the as-deposited thin SiOC/Fe (14/14 nm) multilayer sample is shown in Fig. 5a. In contrast to the irradiation behavior of the thick SiOC/Fe multilayer samples after 600 °C irradiation which maintain their layered structure, thin SiOC/Fe multilayer samples undergo layer breakdown after 0.5 dpa irradiation at 600 °C; pinch-off is observed in the Fe layers and grain spheroidization occurs (Fig. 5b). The layer breakdown becomes more severe as the irradiation dose increases (Figs. 5c and 5d). The corresponding SAD patterns show extra diffraction rings and diffraction spots (highlight in Fig. 5e), indicating the formation of a new phase. Typical intermixed regions are highlighted in Fig. 5c and 5d. These results are similar to the pure thermal annealing result [15]. Although layer breakdown occurs in thin SiOC/Fe multilayer films, one important observation is that no void formation occurs in the Fe component of the composite even after ~10.7 dpa irradiation at 600 °C. The results indicate that the interfaces in the thin composite continue to act as efficient defect sinks even after layer break down.

4. Conclusions

We examined the irradiation stability of the SiOC/Fe composite system after increasing damage levels (approximately 0.5, 2.7, 6.7 and 10.7 dpa,) at 600 °C. No point defect clusters were observed in the SiOC layers and these layers remained amorphous. Irradiation induced void formation was observed in both pure Fe layers and thick SiOC/Fe multilayers (80/60 nm) after irradiation. The average void size was smaller and void swelling was less in SiOC/Fe multilayers than that in pure Fe films, indicating that the interfaces in the composite are effective defect sinks. The strength of this sink is demonstrated by the presence of void denuded zones in the Fe layer of the composite. For thin SiOC/Fe multilayers (14/14 nm), the layered structure breaks

down after 600 °C irradiations but no voids were observed in the Fe grains. All the findings strongly support the idea that crystalline/amorphous SiOC/Fe interfaces and Fe/Fe_xSi_yO_z incoherent interfaces act as efficient defects sinks, extending the low-swelling transient regime and suppressing void swelling during irradiation at 600 °C.

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References

- [1] S.J. Zinkle, G.S. Was. *Acta Materialia* 61 (2013) 735-758.
- [2] G.R. Odette, M.J. Alinger, B.D. Wirth. *Annual Review of Materials Research* 38 (2008) 471-503.
- [3] G.S. Was. *Journal of Nuclear Materials* 367–370, Part A (2007) 11-20.
- [4] C. Sun, S. Zheng, C.C. Wei, Y. Wu, L. Shao, Y. Yang, K.T. Hartwig, S.A. Maloy, S.J. Zinkle, T.R. Allen, H. Wang, X. Zhang. *Scientific Reports* 5 (2015).
- [5] M.J. Demkowicz, R.G. Hoagland, J.P. Hirth. *Physical Review Letters* 100 (2008).
- [6] G.R. Odette, D.T. Hoelzer. *Jom* 62 (2010) 84-92.
- [7] D. Chen, J. Wang, T.Y. Chen, L. Shao. *Scientific Reports* 3 (2013) 1450.
- [8] B.N. Singh. *Philosophical Magazine* 29 (1974) 25-42.
- [9] X.M. Bai, A.F. Voter, R.G. Hoagland, M. Nastasi, B.P. Uberuaga. *Science* 327 (2010) 1631-1634.
- [10] A. Misra, M.J. Demkowicz, X. Zhang, R.G. Hoagland. *Jom* 59 (2007) 62-65.

- [11] M. Nastasi, Q. Su, L. Price, J.A. Colón Santana, T. Chen, R. Balerio, L. Shao. *Journal of Nuclear Materials* 461 (2015) 200-205.
- [12] Q. Su, B. Cui, M.A. Kirk, M. Nastasi. *Philosophical Magazine Letters* 96 (2016) 60-66.
- [13] Q. Su, L. Price, L. Shao, M. Nastasi. *Materials Research Letters* 4 (2016) 48-54.
- [14] W.Z. Han, M.J. Demkowicz, N.A. Mara, E.G. Fu, S. Sinha, A.D. Rollett, Y.Q. Wang, J.S. Carpenter, I.J. Beyerlein, A. Misra. *Advanced Materials* 25 (2013) 6975-6979.
- [15] Q. Su, J. Jian, H.Y. Wang, M. Nastasi. *Philosophical Magazine* 95 (2015) 3876-3887.
- [16] Q. Su, L. Price, J.A. Colon Santana, L. Shao, M. Nastasi. *Materials Letters* 155 (2015) 138-141.
- [17] Q. Su, B. Cui, M.A. Kirk, M. Nastasi. *Scripta Materialia* 113 (2016) 79-83.
- [18] L.K. Mansur. *Journal of Nuclear Materials* 216 (1994) 97-123.
- [19] F.A. Garner, M.B. Toloczko, B.H. Sencer. *Journal of Nuclear Materials* 276 (2000) 123-142.
- [20] J.B.B. J.F. Ziegler, U. Littmark. *The Stopping and Range of Ions in Solids*, Pergamon Press, New York (1985).
- [21] R.E. Stoller, G.R. Odette, B.D. Wirth. *Journal of Nuclear Materials* 251 (1997) 49-60.
- [22] Q. Su, J. Jian, H. Wang, M. Nastasi. *Philosophical Magazine* 95 (2015) 3876-3887.
- [23] W.G. Wolfer. *Journal of Nuclear Materials* 122 (1984) 367-378.
- [24] S.J. Zinkle, C. Kinoshita. *Journal of Nuclear Materials* 251 (1997) 200-217.
- [25] M. Nastasi, J.W. Mayer. *Materials Science & Engineering R-Reports* 12 (1994) 1-52.
- [26] I.J. Beyerlein, M.J. Demkowicz, A. Misra, B.P. Uberuaga. *Progress in Materials Science* 74 (2015) 125-210.
- [27] L.K. Mansur. *Nuclear Technology* 40 (1978) 5-34.
- [28] B.N. Singh, S.J. Zinkle. *Journal of Nuclear Materials* 206 (1993) 212-229.
- [29] B.N. Singh, M. Eldrup, S.J. Zinkle, S.I. Golubov. *Philosophical Magazine a-Physics of Condensed Matter Structure Defects and Mechanical Properties* 82 (2002) 1137-1158.
- [30] S.J. Zinkle, K. Farrell. *Journal of Nuclear Materials* 168 (1989) 262-267.

Figures and figure captions

Fig. 1. The depth profile of radiation damage and helium concentration for (a) nanocrystalline Fe (b) SiOC/Fe composite which are simulated by TRIM (dose of 2×10^{17} ion/cm²).

Fig. 2. Typical cross-sectional TEM images of thick SiOC/Fe multilayers (a) before and (b) after 600 °C irradiation (dose of 2×10^{17} ion/cm²). (c) Typical cross-sectional TEM image of the Fe/SiOC composite after 600 °C irradiation (dose of 2×10^{17} ion/cm²) showing formation of discontinuous intermixed ($\text{Fe}_x\text{Si}_y\text{O}_z$) interfacial layer. (d) A high resolution TEM image of the Fe/ $\text{Fe}_x\text{Si}_y\text{O}_z$ interface exhibiting incoherent lattice matching. (e) Under-focus, (f) over-focus cross-sectional TEM images of Fe films after 600 °C irradiation (dose of 2×10^{17} ion/cm²).

Fig. 3. Typical cross-sectional TEM images from (a) thick SiOC/Fe multilayers, and (b) a pure Fe film after 600 °C irradiation (dose of 2×10^{17} ion/cm²). Microstructure evolution of the thick SiOC/Fe multilayer and the Fe film after 600 °C irradiation with a dose of 8×10^{17} ion/cm² is shown in (c) and (d), respectively. The scanning transmission electron microscopy (STEM) images from thick SiOC/Fe multilayers after 600 °C irradiation with a dose of (e) 2×10^{17} ion/cm² and (f) 8×10^{17} ion/cm².

Fig. 4. A schematic illustration of the void measurement window in (a) SiOC/Fe multilayer sample and (b) pure Fe films after 600 °C irradiation. Statistical analysis of (c) void size and (d) void swelling in pure Fe films and Fe layers from thick SiOC/Fe multilayer composites as function of damage levels.

(a) As-deposited

(b) 0.5 dpa

(c) 2.7 dpa

(d) 10.7 dpa

(e) 10.7 dpa

Fig. 5. Typical cross-sectional TEM images of thin SiOC/Fe multilayers (a) before irradiation and after 600 °C irradiation with a dose of (b) 4×10^{16} ion/cm², (c) 2×10^{17} ion/cm², and (d) 8×10^{17} ion/cm². The inset of each image is the corresponding selective area diffraction pattern. (e) The selective area diffraction pattern of SiOC/Fe multilayers after 600 °C irradiation with a dose of 8×10^{17} ion/cm². An extra diffraction ring and several diffraction spots are observed, indicating the formation of an additional phase.

Graphical abstract

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Fig. 1. Typical cross-sectional TEM images from (a) thick Fe/SiOC multilayers, and (b) a pure Fe film after 600 °C irradiation (dose of 8×10^{21} ion/m²). Statistical analysis of (c) void size and (d) void swelling in pure Fe films and Fe layers from thick Fe/SiOC multilayer composites as function of damage levels. Compared to pure Fe films, the swelling resistance in the Fe layers in thick Fe/SiOC multilayer composite is improved by 2.2 times and the averaged void size is reduced to half.