Irradiation Tolerance of Amorphous SiOC/Crystalline Fe Composite

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Abstract

We examined the radiation tolerance of amorphous silicon oxycarbide (SiOC) and crystalline Fe composite by using ion irradiation and transmission electron microscopy (TEM). The Fe/SiOC multilayer thin films were grown via magnetron sputtering with controlled length scale on a surface oxidized Si (100) substrate. These alloys were irradiated by 120 keV He⁺ ions to a damage level of ~1.3 displacements per atom (dpa) at room temperature. TEM characterization shows no sign of point defect clusters in Fe layer, nor an indication of crystallization or new phase formation in SiOC layer. Our findings suggest that the crystalline/amorphous interface and Fe grain boundary can help to annihilate point defects generated during irradiation, and therefore enhance radiation tolerance properties.

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

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1. Introduction

Development of radiation tolerant materials is critical for longer and safer life cycles in nuclear reactors..[1, 2] Different kinds of interfaces and grain boundaries have been introduced as defect sinks to remove radiation damage and suppress radiation induced dimensional and property changes. For example, interfaces between the metal matrix and nanoscale oxides in oxide dispersion strengthened (ODS) steels systems prove to benefit swelling resistance and

creep resistance.[3, 4] Nanocrystalline metals displays enhanced radiation resistance due to grain boundary assisted defect annihilation.[5] Nanoscale metallic interfaces including Cu/Nb,[6] Cu/V,[7] Fe/W[8] etc., have shown strong defect sink strength and the ability to suppress He bubble formation. Molecular dynamics (MD) simulations reveal that the sink strength is proportional to the amount of free volume created by incoherent interfaces.[9] Compared to crystalline solids, amorphous materials are believed to have a higher amount of free volume. Amorphous/crystalline interface, therefore, can be another type of efficient sink to accumulate and recombine radiation induced vacancies and interstitials for enhanced radiation tolerance property.

In this study, to examine the roles of interfaces in amorphous-ceramic and crystalline-metal composites, α -Fe and amorphous SiOC were chosen as the amorphous ceramic constituent. We selected α -Fe since it represents ferritic-martensitic alloys interesting for reactor applications.[10] The selection of SiOC considers its higher thermal and radiation stabilities. In a previous study, we reported that amorphous SiOC is a new class of superior radiation tolerant materials.[11] It has been demonstrated that SiOC thin films are extremely stable, remaining amorphous even after irradiation up to 18 dpa at room temperature and up to 20 dpa at 600°C. The observations suggested that the atomic displacing processes and subsequent structural changes did not lead to crystallization, segregation, or void formation. In addition, it has been shown that amorphous SiOC had good oxidation and creep resistance,[12-15] making it a promising candidate for a structural material.

2. Experimental

In this paper, Fe/SiOC multilayer films with controlled individual layer thicknesses were fabricated by magnetron sputtering. Also, pure nanocrystalline Fe films were prepared for comparison. Prior to depositions, the base pressure was 9.8×10^{-6} Pa and the Ar partial pressure during deposition was ~ 0.65 Pa. Amorphous SiOC layers were synthesized by radio frequency (RF) co-sputtering from SiO₂ and SiC targets at room temperature, while nanocrystalline Fe layers were obtained by using DC magnetron sputtering.

The Fe/SiOC multilayers and pure Fe films were irradiated together at room temperature using 120 keV He ions. A total fluence of 4×10^{20} ions/m² was used to create an averaged ~ 1.3 dpa of damage in these films. The cross-sectional microstructures of Fe/SiOC multilayers and

pure Fe films before and after irradiation were characterized by TEM. The cross-sectional TEM specimen was prepared by conventional dimple and grinding followed by ion-milling. Low energy (3.5 keV) and low angle (5°) were selected to reduce the ion milling damage. A FEI Tecnai G2 F20 TEM was employed to examine the microstructure of these films before and after irradiation. The typical TEM operation voltage was 200 kV.

3. Results and discussion

Fig. 1 presents typical cross-sectional TEM micrographs of as-deposited Fe/SiOC multilayers films. An alternating Fe and SiOC nanolayer structure with clear interfaces is observed. In thick Fe/SiOC multilayer films (Fig. 1a), Fe and SiOC layers are 60, 80 nm in thickness, respectively. The individual layer thickness for thin Fe/SiOC multilayer films (Fig. 1b) is 14 nm. Both films had a similar total thickness of ~ 420 nm. In addition, all Fe layers exhibit a typical columnar growth with average grain size of ~ 20 nm while all SiOC layers show uniform contrast. The high quality structures suggest that a tunable Fe/SiOC interface density can be achieved by refining the layer length scale. The inset of Fig. 1a and 1b are the corresponding selective area diffraction (SAD) patterns. Both SAD patterns exhibit a diffuse halo around the central beam and ring diffraction patterns with similar interval spacing. These data confirm a pure body-centered cubic (BCC) structure for the Fe layers and the amorphous nature of the SiOC layer for both thick and thin Fe/SiOC multilayer films.

To maximize displacement efficiency and minimize He doping in the films, 120 keV He ions were selected for the irradiation. Fig. 2a and 2b show the depth profiles of dpa values and helium concentrations in the Fe and Fe/SiOC films, respectively, obtained by using the Stopping and Range of Ions in Matter (SRIM) simulation.[16] The He^+ irradiation results in ~ 1.1 dpa in the pure Fe films and ~ 1.3 dpa in the Fe/SiOC composite films. In irradiated pure Fe samples, the majority of He atoms came to rest in the Si substrate. While for Fe/SiOC composite films, there was a negligible amount of He in near surface region (< 200 nm). Therefore, these regions were selected for examining irradiation-induced defect formation.

The cross-sectional TEM images of the pure Fe films before and after irradiation are shown in Fig. 3a and 3b, respectively. The as-prepared Fe film exhibit a columnar structure with an average grain size of ~ 18 nm. The grain size distribution and the crystal structure of pure Fe film is in good agreement with that of the Fe layers in the composite films. The corresponding

SAD pattern shown in the inset of Fig. 3a reveals a BCC structure. After ~1.1 dpa irradiation, there are no indications of dislocation loops in the nanocrystalline Fe films **and grain boundaries structure remain stable**, as shown in Fig. 3b. Also, the SAD pattern of Fig. 3b indicates that it retains its BCC structure after irradiation. Small defects clusters have been observed by others in bulk Fe after 0.01 dpa irradiation.[17, 18] The suppression of defect cluster formation in nanocrystalline Fe films was attributed to the presence of a nanocrystalline grain structure with a large density of grain boundaries. Recent simulation work shows that point defects created by collision cascades can efficiently annihilate at grain boundaries in several metal systems including alpha-Fe[5] and Cu[19]. This process reduces the number of retained point defects in nanocrystalline Fe films, and therefore, suppresses defect clusters formation.

Fig. 4a and 4b show TEM micrographs obtained from thick and thin Fe/SiOC films after irradiation to ~1.3 dpa at room temperature. After irradiation, no defect clusters are observed in the Fe layer and the SiOC layers maintain their amorphous state. In addition, clean Fe/SiOC interfaces are clearly visible, suggesting no secondary phase formation or irradiation-induced mixing or diffusion between the Fe layers and the amorphous SiOC layers. These results demonstrate good **stability of crystalline Fe/ amorphous SiOC interface and** room temperature radiation stability in the Fe/SiOC composite system. Besides grain boundaries in the Fe layers, the crystalline/amorphous interfaces in the Fe/SiOC composite films are anticipated to be regions of high free volume and are able to adopt numerous atomic-level configurations.[9, 20, 21] Therefore, these crystalline/amorphous interfaces can act as highly efficient defect sinks to enhance point-defect recombination, as observed in the present study.

4. Conclusions

We examined the radiation stability of the Fe/SiOC composite system after irradiation to ~1.3 dpa at room temperature. After irradiation, no point defect clusters are observed in the Fe layers and the SiOC layers remain amorphous. The observations suggest that Fe/SiOC crystalline/amorphous interfaces and Fe grain boundaries act as efficient defects sinks, promoting interstitials and vacancies recombination.

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Figures and Figure Captions

Fig. 1. The cross-sectional TEM micrographs of as-deposited (a) thick, (b) thin Fe/SiOC multilayers.

Fig. 2. The depth profile of radiation damage and helium concentration which is simulated by TRIM (dose of 4×10^{20} ion/m²).

Fig. 3. The cross-sectional TEM micrographs of pure Fe films (a) before and (b) after RT irradiation.

Fig. 4. The cross-sectional TEM images of (a) thick, (b) thin Fe/SiOC multilayers after RT irradiation.