***In situ* study of heavy ion irradiation response of immiscible Cu/Fe multilayers**

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Abstract

Recent studies show that immiscible metallic multilayers with incoherent interfaces can effectively reduce defect density in ion irradiated metals by providing active defect sinks that capture and annihilate radiation induced defect clusters. Although it is anticipated that defect density within the layers should vary as a function of distance to the layer interface, there is, to date, little *in situ* TEM evidence to validate this hypothesis. In this study monolithic Cu films and Cu/Fe multilayers with individual layer thickness, h , of 100 and 5 nm were subjected to *in situ* Cu ion irradiation at room temperature to nominally 1 displacement-per-atom inside a transmission electron microscope. Rapid formation and propagation of defect clusters were observed in monolithic Cu, whereas fewer defects with smaller dimensions were generated in Cu/Fe multilayers with smaller h . Furthermore *in situ* video shows that the cumulative defect

density in Cu/Fe 100 nm multilayers indeed varies, as a function of distance to the layer interfaces, supporting a long postulated hypothesis.

Keywords: *in situ* ion irradiation; heavy ion irradiation; immiscible interfaces; Cu/Fe multilayers; size effect

Introduction

High energy neutrons and heavy ions can induce significant densities of interstitials, vacancies, and defect clusters in irradiated metallic materials, and correspondingly lead to degradation of their performance in nuclear reactors. For instance neutron irradiation to a dose of several to tens of displacements-per-atom (dpa) may lead to void swelling, radiation embrittlement, and radiation creep in a variety of metallic materials with face-centered cubic (FCC) structures (1-4). Meanwhile advanced nuclear energy systems call for extraordinary materials that can sustain neutron irradiation damage up to 200 dpa (5-9). Such a daunting challenge drives extensive materials research towards the design and discovery of advanced radiation-tolerant materials.

In general, there are several approaches to enhance radiation tolerance in materials (5). First, the inherent radiation tolerance of metallic materials depends on intrinsic properties, specifically bonding and crystal structure. Stronger atomic bonding in metals corresponds to higher threshold displacement energy, resulting in less radiation damage (10). Crystal structure is another key factor in terms of radiation damage. For instance the visible density of defect clusters is greater in FCC Cu than in body-centered cubic (BCC) Fe subjected to the same dose of radiation (11, 12). Molecular dynamics (MD) simulations suggest that although the total

defect densities in Cu and Fe are very similar at the same dose, cluster size in Cu at the same dose is significantly larger than that in Fe, due to the formation of large vacancy clusters in the cascade core in FCC Cu, and the greater number of free vacancies in BCC Fe (13). Bacon *et al* also suggested that defect clusters in irradiated BCC Fe are relatively small and more finely dispersed, which enhances recombination during radiation (14). Similarly, ferritic/martensitic steels with primarily BCC structures are frequently shown to possess better void swelling resistance than austenitic stainless steels with FCC crystal structures (2). Second, an intensively investigated approach is to design materials with high-density defect sinks, including grain boundaries (GBs) (15-19), twin boundaries (20-23), and phase boundaries (24-31). A majority of these boundaries, acting as unbiased defect sinks, can absorb point defects and their clusters, and promote their recombination (32, 33). For instance nanocrystalline (nc) metals with a high volume ratio of GBs showed significantly reduced defect density during radiation (16, 34-36), and recently direct evidence on defect cluster absorption by high angle GBs in nc Ni was reported via *in situ* radiation experiments inside a transmission electron microscope (TEM) (37). In nanotwinned Ag with a high density of twin boundaries, the density of irradiation induced defects decreased at smaller average twin spacing (20, 21), due to a twin boundary affected zone of radiation induced defect density (23). Furthermore Zhang's group demonstrated that by deliberately introducing voids into nanotwinned Cu, incoherent twin boundaries (ITBs) can act as diffusion channels to deliver radiation induced defects into voids, significantly mitigating radiation damage (22).

Meanwhile there are increasing studies on the role of layer interfaces in radiation damage in multilayers (24-26, 38, 39) and various types of metallic multilayer systems have been investigated, including those with FCC/BCC interfaces (Cu/Nb (25, 26, 29, 40, 41), Cu/V (42-

44), Cu/Mo (45), Cu/W (46), Al/Nb (47), Ag/V (48, 49)), FCC/FCC interfaces (e.g. Cu/Ni (50), Cu/Co (51) Ag/Ni (52, 53)), BCC/BCC interfaces (Fe/W (54)), and BCC/amorphous interfaces (Fe/Fe₂Zr (55), Fe/a-Y₂O₃ (31), Fe/TiO₂ (30)). In addition, it is worth mentioning that coherent interfaces can also mitigate radiation damage (51, 56). The study of He ion irradiated Cu/Fe nanolayers shows that swelling is mitigated in FCC/FCC Cu/Fe with layer thickness of 1 nm, resulting from the suppression of bubble size by coherent layer interfaces (56). Among them, layer interfaces in immiscible systems are more effective (than those in miscible counterparts) for mitigating radiation damage as they inherently recover from radiation induced intermixing (24, 38). High-density misfit dislocations at incoherent interfaces would serve as defect sinks for radiation-induced point defects and catalysts for efficient Frenkel pair recombination (41, 57). As many layer interfaces are effective defect sinks, one anticipates that, similar to the existence of GB denuded zones near high angle GBs, interface affected zones should exist near layer interfaces in immiscible multilayers. Han *et al.* reported void-denuded zones in Cu/Nb nanolayered structure by *ex situ* radiation experiments (58). However, there is little *in situ* evidence to validate if the defect density is indeed lower close to the layer interfaces.

In situ irradiation experiments inside a TEM provide a unique opportunity to investigate defect generation, migration kinetics, and interactions between radiation-induced defect clusters and defect sinks. Although multilayer systems have been extensively studied by *ex situ* irradiation techniques (19, 24-26, 38), there are limited *in situ* evidence that show the influence of layer thickness on radiation tolerance in FCC/BCC multilayer systems. Here we use Cu/Fe multilayers as a model system to investigate, via an *in situ* radiation technique, thickness dependent response to radiation damage under heavy ion irradiation. Cu and Fe were selected as both are classical model systems for FCC and BCC crystal structures, and they have drastically

different responses to radiation damage. This study shows that there is a size dependent variation of defect density in multilayers, and the concentration of defects indeed varies as a function of distance to layer interfaces, manifesting the existence of an interface affected zone.

Experimental

Cu/Fe multilayer films with individual layer thickness, h , of 100 and 5 nm (referred to as Cu/Fe 100 nm and Cu/Fe 5 nm hereafter) were magnetron sputtered at room temperature on HF-etched Si (110) substrates. The chamber was evacuated to a base pressure less than 8×10^{-8} torr prior to deposition. Before the deposition of Cu/Fe multilayers, a 100 nm-thick Cu seed layer was deposited. Details of the deposition parameters were reported previously (59). Cross-sectional TEM (XTEM) samples were prepared by dimpling and low angle (5°) low energy (3.5 keV) Ar ion milling and subsequent ion polishing (2 keV).

In situ ion irradiation experiments were conducted at room temperature in a JEOL JEM-2100 TEM operated at 200 kV in the Ion Beam Lab at Sandia National Laboratories (60). **The defect evolution in the coarse-grained Cu specimen was evaluated by using (111) two-beam imaging condition.** A 6 MV EN Tandem ion accelerator was used to generate a 3 MeV Cu^{3+} beam. **The ion beam entering the microscope forms an angle of 90° to the vertical transmitted electron beam. The TEM thin foils were tilted by $20 - 30^\circ$ relative to the vertical axis during the *in situ* radiation studies.** Special attention was paid to ensure the layer interfaces were edge-on during *in situ* radiation experiments in TEM. Depth-dependent damage and defect concentration profiles were calculated by the Stopping and Range of Ions in Matter software package (SRIM-2008) using full damage cascades mode (61, 62). The Cu/Fe multilayers with equal layer thickness were simplified as $\text{Cu}_{50}\text{Fe}_{50}$ alloys for simulation. SRIM simulations show that most Cu ions at 3 MeV pass entirely through the thin specimen, leaving behind only displacement

damage in the irradiated TEM foil. Subsequent *ex situ* TEM characterization was carried out on an FEI Tecnai G2 F20 Super Twin microscope operated at 200 kV. The average thickness of TEM thin foils was determined to be ~ 100 nm, by using a convergent beam electron diffraction (CBED) technique (63). The dimension of individual defects was measured along their short axis. The statistics on the number of defects per unit area was collected from a large number of snapshots (images) captured during *in situ* radiation experiments. The average defect density was calculated by using the number of defects per unit area and the measured average TEM foil thickness. The error bar was determined from standard deviations among numerous measurements from various micrographs. To obtain defect statistics in Kr ion irradiated Cu/Fe 100 nm and Cu/Fe 5 nm multilayers, multiple TEM image sizes were used with dimensions of $\sim 350 \times 350$ nm and 120×120 nm, respectively.

Results

Microstructures of monolithic Cu, Cu/Fe 100 nm and Cu/Fe 5 nm multilayers irradiated by Cu ions at 3 MeV are compared in TEM micrographs in Fig. 1. In monolithic Cu irradiated to a nominal dose of 0.5 dpa (2.5×10^{14} ions/cm²), high-density defect clusters were generated (Figs. 1a). In comparison, irradiation to 1 dpa (5×10^{14} ions/cm²) led to a moderate increase in loop density in the Cu layers of Cu/Fe 100 nm samples (Figs. 1b). Columnar grains with an average grain size of 16 nm in Fe layers remained after irradiation. Few radiation-induced defect clusters were detected in irradiated Cu/Fe 5 nm nanolayers (Figs. 1c).

Several TEM micrographs in Fig. 2 recorded during *in situ* radiation (up to 1 dpa) of Cu/Fe 100 nm multilayers revealed microstructural evolution. As shown in Fig. 2a, the as-deposited multilayer had a low defect density. This limited number of defects might have been

produced during growth or by the Ar ion milling. At a radiation dose of 0.1 dpa, a few defects appeared in Cu layers. While many of these defects have round geometry suggestive of dislocation loops, some defects with triangular geometry were also frequently identified suggestive of stacking-fault tetrahedra (SFT). The exact nature of these defects could not be determined due to the dynamic nature of the *in situ* TEM experiment, however both of these defect structures are often observed in irradiated Cu (64). With the increase of radiation dose to 0.5 and 1 dpa, the dimensions of the defect clusters increased monotonically without significant increase in defect density in the Cu layers. Few visible defect clusters appeared in the Fe layers throughout the entire radiation experiment. In contrast defects were readily visible and appeared much more frequently in the Cu layers during *in situ* radiation, permitting us to collect reliable statistical data for defect clusters in the irradiated Cu layers. In addition, no significant indication of intermixing along layer interfaces was observed in the irradiated multilayers.

The Cu/Fe 5 nm multilayer showed the accumulation of significantly less displacement damage defects under identical radiation environments. As marked in Fig. 3, several pre-existing growth defects (marked as 1-4 in blue arrows) in the layers appeared to be stable during the *in situ* radiation experiment and, as such, were used as fiducial marks. The density of newly generated defect clusters (labeled as 5 and 6 by red arrows) during radiation is low compared to that of pre-existing defects. Supplementary video 1 is provided to demonstrate the dynamics of Cu/Fe 5 multilayers exposed to 3 MeV Cu irradiation.

Statistical studies on evolution of the density and dimensions of defect clusters were performed on the irradiated monolithic Cu sample, the Cu layers of the Cu/Fe 100 nm, and Cu/Fe 5 nm multilayer samples. As shown in Fig. 4a, defect density in irradiated monolithic Cu increased rapidly with dose and did not reach saturation by 0.5 dpa (65). In contrast, defect density in the Cu layers of irradiated Cu/Fe 100 nm multilayer samples increased much more slowly than that

in monolithic Cu, and by 0.5 dpa appeared to saturate at a level approximately 1/5 of that in monolithic Cu. In the ion irradiated Cu/Fe 5 nm multilayer sample, few defect clusters were generated during radiation, and the overall defect density barely changed as shown by a horizontal dash line in Fig. 4a. Meanwhile the pre-existing defects (growth defects) were stable during radiation.

Significantly lower defect density was observed in the Cu/Fe 5 nm multilayer sample than that in the Cu/Fe 100 nm multilayer. Another interesting observation is the average defect size, shown in Fig.4b, in monolithic Cu was 7 ± 2 nm, similar to that in the Cu layers in Cu/Fe 100 nm multilayers: 9 ± 3 nm. In contrast, the defect size in irradiated Cu/Fe 5 nm was 3 ± 1 nm on average, with none exceeding the 5 nm layer thickness.

Discussion

The influence of Cu/Fe layer interfaces on absorption of radiation induced defects

Incoherent layer interfaces in immiscible multilayer systems have been proven as defect sinks in irradiated multilayers (24-26, 41). The Cu/Fe interfaces in irradiated Cu/Fe multilayers also appear effective in alleviating radiation-induced damage. This *in situ* irradiation study shows a clear layer thickness dependence of defect density in irradiated Cu/Fe multilayers: multilayers with smaller h show much lower radiation induced defect density than those in irradiated monolithic Cu. Meanwhile, the smaller layer thickness ($h = 5$ nm) effectively restrain the growth of defect clusters in Cu layers, evidenced by the fact that the average defect size in the irradiated Cu/Fe 5 nm multilayer (approximately 3 nm) was much smaller than that in monolithic Cu and the Cu/Fe 100 nm multilayer (approximately 7 and 9 nm, respectively). In the Cu/Fe 100 nm multilayers, the average grain size of Cu is ~ 50 nm, less than the individual layer

thickness of 100 nm. Subsequently, the grain boundaries may also have acted as defect sinks during *in situ* radiation.

A size effect on alleviation of radiation induced defect cluster density has been observed in several immiscible metallic multilayer systems. For instance radiation damage in terms of the density of He bubbles in He ion irradiated Cu/V, Cu/Nb, Ag/Ni and Ag/V multilayers decreases monotonically with decreasing h (38, 42, 48, 52). Recently we reported that the density of He bubbles in He ion irradiated Cu/Fe multilayers first decreased with decreasing h to approximately 5 nm, and then increased at smaller h , however, void swelling decreases monotonically with decreasing h (56).

The size effects on layer interface-alleviated radiation damage in numerous immiscible metallic multilayers seem to suggest that there should be a layer interface affected denuded zone, wherein the density of defect clusters decreases with proximity to the layer interface in comparison to the layer interior. It is challenging to directly observe interface affected (or defect denuded) zone by *ex situ* radiation experiments (58). There is indeed some *in situ* evidence that captured the absorption of mobile defect clusters by layer interface in immiscible Ag/Ni multilayers (53). However the post radiation analysis or comparisons of individual images captured at different doses during radiation did not provide the *in situ* experimental existence of a layer interface affected zone. To address this question, we performed accumulative dose dependent studies in Cu/Fe multilayers.

A statistical study of cumulative defect density in Cu layers in Cu/Fe 100 nm multilayers irradiated over a dose of 0.25 - 0.31 dpa in 160 s is provided in Fig. 5 (See the 4-times accelerated Supplementary Video 2 for details of the dynamics). A maximum defect density was

identified in the middle of the Cu layer, whereas defect density quickly diminished on both sides of the centerline. The density of defect clusters in Cu reached a minimum level adjacent to the Cu/Fe layer interfaces. Although there are still defects in the regions immediately adjacent to the layer interface, such a distribution of defect density clearly supports the prior hypothesis of the existence of a layer interface affected zone.

The existence of layer interface affected zones also implies that it is more difficult for defect clusters to nucleate near layer interfaces, as point defect density is lower approaching a layer interface, consistent with the reaction-diffusion model proposed by Demkowicz *et al.* (66). The defect density gradient leads to defect diffusion from grain interiors to layer interfaces. Therefore, the capability of multilayers to mitigate radiation damage by absorbing or eliminating radiation induced defects is enhanced when h is reduced to a dimension comparable to that of the fast diffusion zone suggested by the reaction-diffusion model (66).

We hypothesize that when h decreases to be comparable to the cascade size, long-range transport of interstitials away from the initial PKA due to focusing and channeling (67) is suppressed as a result of lattice and chemical discontinuity across layer interfaces. Therefore, the separation distances between radiation induced vacancies and interstitials are reduced at smaller h , leading to enhanced vacancy-interstitial recombination. Meanwhile, defects generated near interfaces could also be absorbed by interfaces directly. Here we can estimate the average cascade size as follows. The average recoil energy under 1 MeV heavy ion irradiation is ~ 5 keV, although such an estimation has slight dependence on the mass of projectiles (67). The individual cascade diameter from a recoil of that energy is approximately 5 nm in Cu according to Averback's studies (68, 69). Therefore, in our experiments, the 3 MeV Cu ions may generate slightly larger cascades, comparable to h in the Cu/Fe 5 nm multilayers. Such a crude estimation

is consistent with fewer radiation induced defects and superior stability in irradiated Cu/Fe 5 nm multilayers.

The formation of stacking fault tetrahedra in Cu in irradiated Cu/Fe 100 nm multilayers

SFTs and dislocation loops are two major types of defect clusters in irradiated FCC metals. The defect types could be identified by various methods, including weak-beam dark field (WBDF) TEM (70), high-resolution TEM (HRTEM) (21) and high-resolution scanning TEM (71). In Cu with a low stacking fault energy (SFE), $\sim 55 \text{ mJ/m}^2$ (72), SFTs are frequently observed experimentally (73). Singh and Zinkle pointed out that half of the defect clusters in irradiated Cu are SFTs (64) and a previous study on proton irradiated Cu shows that 90% of defect clusters are SFTs (74). Our recent study shows that Kr ion irradiation also induced plenty of SFTs in Cu (75). Several theoretical studies have been performed to describe the formation mechanism of SFT as a result of irradiation (76, 77). Meanwhile, there are several pieces of *in situ* evidence on the formation of SFT, including the studies on Ni based superalloy (Inconel X-750) (78), Au (79-81), and Cu (82). Here, we show bright-field TEM images of a SFT formation. As shown in Fig. 6, a small defect, a vacancy (Frank) loop emerged in the Cu layer next to the Cu/Fe layer interface during *in situ* ion irradiation at 0 s. The Frank loop continued to expand within the next 0.6 s. By 1.5 s, a defect stabilized into a triangular structure as a SFT (See Supplementary Video S3 for details). A SFT is a highly stable configuration of vacancy clusters in Cu, and can be retained in FCC metals during radiation. Due to the dynamic nature of the *in situ* experiment, the nature of the Frank loop and SFT could not be confirmed using advanced TEM defect characterization techniques.

Conclusion

We studied monolithic Cu films and Cu/Fe multilayers with h of 100 and 5 nm, subjected to *in situ* Cu ion irradiation at room temperature to approximately 1 dpa. A clear size effect in defect density and size was observed: in monolithic Cu, rapid propagation of defect clusters was observed, whereas fewer defects (less than 1/5 of defects in Cu) were generated in Cu/Fe 100 nm multilayers and radiation induced defects in Cu/Fe 5 nm multilayers were difficult to identify. A statistical study in 100 nm-thick Cu layers via *in situ* video reveals the existence of a layer interface affected zone in which the cumulative defect density declines near the Cu/Fe interfaces. Meanwhile, the defect growth in 5 nm-thick Cu layers was suppressed (approximately 3 nm) by Cu/Fe interfaces while the defect sizes in monolithic Cu (approximately 7 nm) and Cu layers in Cu/Fe 100 nm multilayer (approximately 9 nm) are similar.

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Figure captions

Fig. 1. A comparative overview of radiation damage in a monolithic Cu film, Cu/Fe 100 nm, and Cu/Fe 5 nm multilayers irradiated by 3 MeV Cu ions to 0.5-1 displacements-per-atom (dpa) at room temperature. (a) In the irradiated monolithic Cu subjected to a dose of 0.5 dpa, high-density defect clusters were generated. (b) Irradiation to 1 dpa led to a moderate increase in defect density in the Cu/Fe 100 nm multilayers. Nanoscale columnar grains in Fe layers remained after irradiation. Defect clusters were sporadically distributed primarily in the Cu layers. (c) Few defect clusters were detected in the irradiated Cu/Fe 5 nm nanolayers.

Fig. 2. In situ TEM observation of radiation-induced microstructural evolution in the Cu/Fe 100 nm multilayers irradiated up to 1 dpa. (a) The Cu in as-deposited multilayers had a low defect density. (b) At 0.1 dpa, defect clusters appeared in Cu layers, as shown by the arrows. (c) When the dose increased to 0.5 dpa, the density of defect clusters also increased. (d) No significant increase of defect density was observed up to 1 dpa. Few discernable defect clusters were

observed in the Fe layers throughout the entire radiation experiment. Layer interfaces remained essentially unchanged after irradiation.

Fig. 3. In situ TEM snapshots of defect evolutions in Cu/Fe 5 nm-thick multilayer irradiated to 1 dpa. Few defects (marked as 5-7 with red arrows) were generated in irradiated nanolayers. Several pre-existing (growth) defect clusters (marked as 1-4 by blue arrows) were identified as references and these pre-existing defect clusters only changed slightly during radiation. (The dynamics of defect generation and evolution is captured in Supplementary Video 1)

Fig. 4. A statistical study of identifiable defect cluster density and size in Cu ion irradiated monolithic Cu, 100 nm-thick Cu layers in Cu/Fe 100 nm, and Cu/Fe 5 nm multilayers. (a) In irradiated Cu, the defect cluster density increased rapidly with dose. In contrast, the defect density in Cu layers in Cu/Fe 100 nm multilayer was approximately 1/5 of that in irradiated Cu. Much fewer defects were generated in irradiated Cu/Fe 5 nm multilayer and the dash line indicates no obvious increase in defect density. The solid lines are visual guides that illustrate the trend of defect density evolution. (b) The average detectable defect size in irradiated monolithic Cu is 7 ± 2 nm, compared to 9 ± 3 nm for Cu/Fe 100 nm and 3 ± 1 nm for irradiated Cu/Fe 5 nm multilayers.

Fig. 5. A statistical study on the accumulative number of identifiable defect clusters (normalized in unit of frequency by the total number of defect clusters, ~ 90) generated in Cu layers in Cu/Fe 100 nm multilayer over a dose range of 0.25-0.31 dpa in 160 s (see Supplementary Video 2). Each data point was obtained by counting defects in the measured regions (stripes) with dimension of $\sim 10 \times 350$ nm in each Cu layer. It is evident that fewer defects are identified near the Cu/Fe interfaces during radiation; in contrast, an increase in defect-generation frequency is observed further away from the layer interfaces and the defect-generation frequency reaches a maximum in the middle of the Cu layers. The dash line is a visual guide that delineates the defect-generation profile.

Fig. 6. In situ video snapshots capturing the evolution of a radiation induced defect cluster in a Cu layer in the Cu/Fe 100 nm multilayer irradiated to ~ 0.5 dpa. (a) At $t = 0$ s, a small defect cluster (likely to be a Frank loop) appeared in the Cu layer adjacent to the Cu/Fe layer interface. (b-c) Within 0.6 s, this defect cluster continued to evolve during radiation. (d) By 1.5 s, the

defect cluster transformed to a triangular defect, which may be a stacking fault tetrahedron (SFT)
(The dynamics of this evolution process is provided in Supplementary Video 3).

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6