***In Situ* Transmission Electron Microscopy He⁺ Implantation and Thermal Aging of Nanocrystalline Iron**

Brittany Muntifer^{a,b}, Youwu Fang^b, Asher C. Leff^c, Aaron Dunn^{a,d}, Jianmin Qu^{b,e},
Mitra L. Taheri^c, Remi Dingreville^a, Khalid Hattar^a

^aSandia National Laboratories, Albuquerque, NM 87185, US

^bDepartment of Civil and Environmental Engineering, Northwestern University, Evanston, IL 60208, USA

^cDepartment of Materials Science & Engineering, Drexel University, Philadelphia, PA, USA

^dGeorge W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology, UMI 2958 Georgia Tech CNRS, 57070 Metz, France

^eSchool of Engineering, Tufts University, Medford, MA 02155, USA

Keywords: Nanocrystalline iron, *In situ* TEM, Helium implantation, Annealing

Abstract

The high density of interfaces in nanostructured materials are hypothesized to improve radiation tolerance compared to coarse-grained materials. In order to investigate the roles of vacancies, self-interstitials, and helium, both room temperature *in situ* TEM He⁺ implantation and annealing, as well as high temperature He⁺ implantation was performed on nanocrystalline iron. Dislocation loops formed by the accumulation of mobile point defects rather than by displacement cascades at intermediate temperatures. Around 600 °C, loops disappeared through gradual shrinking, which is hypothesized to correspond to the annihilation of self-interstitial atoms by mobile vacancies that also resulted in cavity formation. The room temperature implantation resulted in cavities evenly distributed throughout the grain after annealing, whereas cavities were predominately observed at grain boundaries for the elevated temperature implantation. This difference is associated with the formation of stable helium-vacancy complexes in the grains during room temperature implantation, which is not present during high temperature implantation.

1 Introduction

Materials used in future nuclear reactors will have to withstand elevated temperatures and simultaneous high energy neutron irradiation, which results in point defects, displacement cascades and transmutation reactions [1, 2]. The combination of these effects may lead to severe degradation of mechanical properties and limit the lifetime of the material. Studies have shown that certain grain and phase boundaries may act as preferential trapping sites for irradiation induced defects [3-5] and promote recombination of interstitials and vacancies [6-8], which may reduce the degradation of mechanical properties. Due to the high density of grain boundaries and interfaces, a number of nanostructured materials have displayed a higher tolerance to radiation damage compared to their coarse-grained counterparts. For example, reduced defect density and increased radiation tolerance in nanocrystalline metals has been reported for ion irradiated Fe [3, 9], Pd [10], Cu [11], W [12], and Ni [13, 14].

Among the radiation-induced defects, helium is a common transmutation product which results in increased swelling and blistering of materials and contributes to the degradation of other mechanical properties [15, 16]. In this study, nanocrystalline iron was chosen as a model material because iron is the basis of more complex steels, which may be used as structural materials for internals in future reactors. Numerous studies involving helium implantation into coarse grained iron have been performed to characterize irradiation-induced defect evolution with helium concentration and temperature; however, only limited irradiation studies have been performed on nanocrystalline iron [3]. Notably, defect evolution in coarse grained iron has been characterized as a function of helium implantation temperature [17, 18] as well as post-implantation annealing [17, 19, 20]. These two different thermal pathways appear to lead to similar defect structures at temperature in coarse-grained iron films. A systematic comparison

between thermal pathways is still required for a complete understanding of radiation damage evolution in nanocrystalline iron, where grain boundaries serve as primary helium and defect sinks.

To elucidate mechanisms involved in radiation defect evolution as a function of temperature and helium content in nanocrystalline iron, two types of experiments were performed: 1) helium implantation at room temperature followed by annealing to 600 °C and 2) helium implantation at elevated temperatures up to 600 °C. These two thermal conditions were chosen to gain insight into the role of the sequence of irradiation and annealing on damage accumulation, which is important when designing experiments intended to mimic reactor conditions. It was observed that the dislocation loop behavior was similar at each temperature, whether implanted directly at that temperature or implanted at room temperature and annealed, but the final cavity structure depended strongly on the thermal pathway. In comparison to helium implantation studies performed in coarse grained iron, the dislocation loop growth with temperature was significantly limited, and the loops completely anneal out above 600 °C.

2 Experimental Procedure

2.1 Nanocrystalline Iron Thin Film Production

Nanocrystalline free-standing iron thin films approximately 50 nm in thickness were deposited on optical quality, polycrystalline NaCl substrate at room temperature by pulse laser deposition (PLD) using a KrF excimer laser from a 99.9985% (metal basis) iron target (Alfa Aesar). For further information on the deposition process, see [21]. The salt substrate was then dissolved in a deionized water bath, and the free-standing iron film was floated onto a copper clamshell Transmission Electron Microscopy (TEM) grid. To establish a through-thickness grain

structure, the films were annealed to a final temperature of 600 °C at an average rate of 50 °C/min and held at the maximum temperature for 5 minutes using a 2.3 mm Hummingbird heating stage in a JEOL 2100 TEM operated at 200 kV.

2.2 Helium Implantation Parameters

In situ helium implantation of the iron films was carried out utilizing a JEOL 2100 TEM and a 10 kV Colutron ion accelerator that are part of the *In situ* Ion Irradiation TEM (I³TEM) facility at Sandia National Laboratories [22]. The implantations occurred in either a Hummingbird heating stage or a Gatan 628 stage tilted 30° towards the ion beam line in order to provide real time nanoscale observations during implantation. The helium beam had an average flux of approximately 10^{14} He⁺/cm²s and each sample was implanted to a total dose of approximately 1.5×10^{16} He⁺/cm². Two thermal pathways for defect evolution were investigated: (1) Helium implantation at room temperature followed by annealing to 600 °C, and (2) Helium implantation at elevated temperatures up to 600 °C.

Based on Stopping and Range of Ions in Matter (SRIM) simulations [23], the 10 keV helium concentration profile curve was bell shaped, with an average penetration depth of 30 nm and a maximum depth of approximately 80 nm, further than the film thickness of 50 nm. Approximately 80% of incident helium ions were expected to implant in the film, with the remainder being transmitted. Irradiation to 1.5×10^{16} He⁺/cm² resulted in an average damage level of approximately 2 displacement per atom (dpa) and nearly 3.75 at.% helium (assuming all implanted helium remained in the sample and did not escape through the free surfaces). The dpa measurement and penetration depth were estimated using the SRIM recoil simulations under the “detailed calculation with full damage cascade mode” [23] at the tilted geometry used in the

experiment. SRIM simulations show that the damage profile is not uniform; instead, the damage is heavily concentrated at the entrance side of the 50 nm-thick film and drops rapidly throughout the film. The small grain size made tilting to optimal defect imaging conditions (two-beam, down-zone, or weak-beam) difficult; therefore, all imaging was done in bright-field mode.

3 Results

Helium implantation into nanocrystalline iron resulted in two types of visible radiation damage: dislocation loops and cavities. It is assumed here that visible black spots observed during in-focus imaging are small dislocation loops. The nature of dislocation loops (vacancy or interstitial) is difficult to characterize experimentally, due to the small size of the grains [24]. The dislocation loops are therefore assumed to be interstitial in nature, as has been seen in previous helium implantation experiments in coarse-grained iron [17-19]. In addition, damage that appears as circular features that are black in over-focus, white in under-focus, and invisible in-focus (Fresnel imaging) are assumed to be small cavities [24]. In this work, the term cavities is used, since the He^+ concentration was not determined to distinguish if the cavities were over-pressurized He^+ bubbles or voids dominated by agglomerated vacancies.

Section 3.1 discusses dislocation loops, which form as a result of room temperature helium implantation, and their associated response to post-implantation annealing to 600 °C. Several types of dynamic behavior of dislocation loops observed during annealing are highlighted in Section 3.2. Section 3.3 discusses dislocation loop accumulation as a function of implantation temperature, and compares this behavior with that observed in the case of room temperature implantation and annealing. Section 3.4 compares the final cavity structure for the two different thermal conditions.

3.1 Dislocation loop accumulation with annealing temperature

Figure 1 illustrates the observations of room temperature helium implantation to a total dose of $1.5 \times 10^{16} \text{ He}^+/\text{cm}^2$, which results in approximately 2 dpa and 3.75 at% He^+ , followed by post irradiation annealing. The pre-implantation and post-implantation images at room temperature in Figs. 1 (a) and (b), show the same area of the sample. After irradiation, it was difficult to discern many radiation induced defects; however, a small number of dislocation loops on the order of 2 nm in size were visible in a few grains in which the defects were in optimal electron imaging conditions. Fresnel imaging did not reveal any cavity structures.

The helium implanted sample was immediately annealed and Figs. 1 (c)–(f) show images at annealing temperatures up to 600 °C. The elevated temperature images do not show the same areas of the film. Note that defects are present in all grains; however, diffraction conditions are vary from grain to grain such that defects are not visible in all grains in a single image. The annealing process can be broken down in four stages: (1) Between 200 °C and 300 °C, a high density of small dislocation loops became visible in grains in which the dislocation loops are well oriented for imaging. (2) By 400 °C the dislocation loops had grown in size to an average of nearly 4 nm. (3) By 500 °C the loops were over 5 nm on average, with some as large as 15 nm. (4) By 600 °C, all dislocation observable loops had annealed out of the film.

Figure 1. (a) Film before room temperature implantation (b) same area of film after room temperature implantation to $1.5 \times 10^{16} \text{ He}^+/\text{cm}^2$. The sample was immediately annealed in-situ after implantation, although the same area of the film was not imaged at elevated temperatures. (c) Film at 300 °C, a high density of nanometer sized dislocation loops is visible. The loops continue to grow in size with temperature. (d) Film at 400 °C, the loops have an average size of nearly 4 nm . (e) Film at 500 °C, the loops reached an average size of over 5 nm with some as large as 15 nm. (f) The film at 600 °C, the dislocation loops have completely annealed out of the film.

Figure 2 shows a series of images taken from video recordings during post-implantation annealing. The central grain remains in similar diffraction conditions during the annealing process. Very few loops can be seen in the imaging conditions of the grain in Fig. 2 until around 300 °C. A high density of small loops, around 3 nm in size, was present above 300 °C. Upon further annealing, the loops size and density increased. After 400 °C, the loop density dropped rapidly and the loops continued to increase in size, reaching a maximum size around 550 °C. By

600 °C the loops had annealed out of the sample through several mechanisms discussed in section 3.2.

Figure 2. Post implantation annealing showing development of defects in one grain that maintained good imaging conditions during annealing. (a) At 300 °C, a high density of small loops with an average size of over 3 nm was present.(b)-(c) loops continue to grow in size while apparent density decreases(d) At 550 °C, a significant decrease in density of loops was observed. The loops reached a maximum size with some loops surpassing 15 nm diameter.

3.2 Dynamic Dislocation Loop Observations

In situ TEM allowed for dynamic observations from video recordings during implantation and annealing, which provided insight into the mechanisms responsible for the size and density

trends of loops demonstrated in Fig. 2. Three phenomena in particular are discussed here: 1) the gradual appearance of small loops during room temperature irradiation, 2) the gradual growth of loops and decrease of loop density between 400 °C and 500 °C, and 3) the rapid decrease in density and disappearance of loops between 550 °C and 600 °C.

During room temperature implantation, a low density of small loops gradually became visible in grains well oriented for dislocation loop imaging. This gradual appearance of the loops, taking 1–2 seconds to reach full size, suggests a different mechanism for dislocation loop formation than in the case of heavy-ion irradiation, in which dislocation loops can form directly in displacement cascades [25]. Instead, these loops appeared to grow by accumulation of small defects (such as individual self-interstitial atoms (SIAs) and small SIA clusters), which are beyond the resolution of TEM. Supplemental Video 1 demonstrates this mechanism.

Dislocation loops were observed to slowly grow during the annealing process up to approximately 550 °C. Most of this growth was also apparently due to the accumulation of smaller defects, which were beyond the resolution of the TEM. Around 500 °C, a second mechanism of loop growth was observed. At this temperature, a small fraction of TEM visible loops became mobile. Figure 3 shows two loops, pointed out inside the white circle of Fig. 3(a), coalescing into one larger loop over the course of approximately half a second. The full dynamics can be seen in Supplemental Video 2. This mechanism is likely also responsible for some of the loop density decrease apparent in Fig. 2.

Figure 3 Images from video recordings during post-implantation annealing around 500 °C which show (a) two dislocation loops inside the white circle, (b) loops approaching each other and (c) loops coalescing into one loop over the course of 0.54 s.

A second mechanism that contributes to the decrease in dislocation loop density during annealing is the apparent escape of mobile loops at the free surface. Figure 4 shows a loop, circled in white in Fig. 4(a), disappearing over two video frames, corresponding to 0.13s. This can also be viewed in Supplemental Video 3. Because there was no change in global contrast in this time period, it is thought that the loop should still be in adequate imaging conditions. This disappearance may result from either the rapid dissolution of point defects into the matrix or, more likely, the migration of the loop to the nearest sink, which in this case is the free surface. Figures 3 and 4 demonstrate two mechanisms competing to decrease the density of dislocation loops which have also been observed in coarse grained irradiated iron [25-27] Both of these mechanisms rely on loop mobility and are limited to only a small fraction of visible loops which were seen to be mobile.

Figure4 Dislocation loop disappearing around 525 °C. Assumed to escape the surface.

In contrast to the previously mentioned mechanisms involving dislocation loop mobility, between 550 °C and 600 °C most loops were immobile and appeared to shrink in size until no longer visible, as in Fig. 1(f). The mechanism responsible for this observed behavior is likely annihilation with vacancies dissociating from vacancy and helium-vacancy clusters, as discussed in Section 4.

3.3 Dislocation loop behavior at elevated implantation temperature

In order to investigate the difference in defect behavior between room temperature implantation followed by annealing and implantation directly at elevated temperature, a second set of implantation experiments were performed in which helium was implanted directly at elevated temperatures. Figure 5 illustrates bright field TEM images of high temperature helium implanted nanocrystalline iron thin films to $10^{16} \text{ He}^+/\text{cm}^2$, where a different film was implanted at each temperature. Implantation at 300 °C, Fig. 5(a), resulted in small nanometer sized dislocation loops in grains in which the defects were well oriented for imaging. Implantation at

400 °C and 500 °C, Figs. 5 (b) and (c), resulted in a high density of dislocation loops with an average size of approximately 3–4 nm and a maximum size of over 10 nm. In contrast, implantation at 600 °C, Fig. 5(d), resulted in no visible loops.

Figure 5 High temperature helium implantation into nanocrystalline iron to a total of 10^{16} He⁺/cm². (a) Implantation at 300 °C resulted in nanometer size dislocation loops, (b) implantation at 400 °C and (c) 500 °C resulted in dislocation loops with maximum diameter over 10 nm and (d) implantation at 600 °C did not result in visible dislocation loops.

In both the room temperature implantation/post-implantation annealing case, as well as the high temperature implantation case, dislocation loop sizes increased with temperature and the loops were not present above approximately 600 °C. Further analysis is needed to characterize loop size and density in grains at the same diffraction conditions for the two thermal cases, but it

appears as though the two thermal conditions result in loops of similar average size and the same trends in density are observed.

3.4 Cavity Formation

For both room temperature helium implantation followed by annealing (Fig. 1) as well as high temperature helium implantation (Fig. 5), through-focus imaging did not reveal the presence of cavities until approximately 600 °C. Figure 6 shows under-focus images of cavities at 600 °C for both the annealed film, Fig. 6(a), as well as the high temperature implanted film, Fig. 6(b). Several examples of cavities are highlighted with black arrows in each.

For the room temperature implantation followed by annealing case, Fig. 6(a), the cavities appear to be evenly distributed throughout the image, with no obvious preference for grain boundaries or various grain orientations. The observed behavior is drastically different in the high temperature implantation case, Fig. 6(b), where the cavities are larger and appear to demonstrate a higher density along certain grain boundaries.

Figure 6 Under-focus image revealing cavity structure at 600 °C in (a) room temperature implanted then annealed and (b) high temperature implanted nanocrystalline iron thin films.

4 Discussion

In situ helium implantation TEM experiments were performed on nanocrystalline iron thin films at room temperature, which was followed by annealing, as well as at elevated temperatures. Dislocation loops were created during helium implantation to $1.5 \times 10^{16} \text{ He}^+/\text{cm}^2$, corresponding to approximately 2 dpa, at implantation temperatures below 600 °C. Video recordings during irradiation at room temperature revealed one mechanism of loop growth, seen in Supplemental Video 1. In this video, a loop appears and grows over approximately one second during helium implantation. This is likely the result of clusters of defects, which are too small to be visible by TEM, coalescing into larger observable defects. A similar effect was seen during annealing, in which many loops slowly appeared and continued to grow through the accumulation of small defects, such as SIAs and small SIA clusters, which were beyond the resolution of the TEM in these conditions.

Previous experiments have shown that both interstitial and vacancy loops may form in α -iron as a result of radiation damage. For example, TEM experiments have suggested that both interstitial [28-30] and vacancy [30, 31] loops exist in neutron irradiated iron. Self-ion irradiation also results in interstitial [26, 32-34] and vacancy [35] loops. Heavy-ion irradiation resulted in vacancy loops [26, 36-38]; in contrast, light ion irradiation, such as helium [17-19] and hydrogen [27], as well as electron [17, 19, 27, 39] resulted in interstitial loops. It has been proposed that the dislocation loop nature may be dependent on the size of defects created during irradiation. Large cascades may collapse into metastable vacancy loops [36]. In contrast, irradiation with species that do not cause large cascades have only been seen to create interstitial loops, and the vacancies should take the form of voids, which are more energetically favorable than vacancy loops in α -iron [40]. Since 10 keV helium ions do not result in dense cascades in iron [23], it is unlikely that the loops are vacancy in nature. The loops are therefore assumed to be interstitial loops in this discussion, as has been seen in previous helium implantation experiments in coarse-grained iron [17-19].

In addition to the slow accumulation of SIAs to form visible loops and continued loop growth during annealing, a second mechanism for loop growth observed in this work is the coalescence of larger SIA loops, which are visible in the TEM, at temperatures above 500 °C (Fig. 3). Interstitial loops in iron have been seen to be highly mobile and undergo one-dimensional (1-D) diffusion even in the absence of stress [25, 41, 42]; however, the presence of helium has been seen to significantly decrease mobility of loops [20, 43]. It should be noted that in this experiment, only a small fraction of the loops were mobile, and 1-D loop hopping was not observed at all. The mechanisms of loop growth observed in Fig. 3 did not appear to be the dominant mechanism of growth. Instead, most loops were immobile and grew in size from 300

°C to 500 °C through further accumulation of small defects which were beyond the resolution in these imaging conditions.

During annealing, some loops were seen to escape the surface, as shown in Fig. 4; however, most loops were not mobile and instead shrank in size above 550 °C. The shrinking and disappearance of dislocation loops corresponds to the appearance of cavities (Fig. 6). The results shown in Figs. 1 and 6 suggest that in room temperature implanted films, vacancies are trapped in stable vacancy or helium-vacancy clusters that are too small to be visible in the TEM. Some of these clusters become unstable and emit vacancies above approximately 550 °C, at which point they annihilate with interstitial loops, causing them to shrink and disappear. The remaining helium and vacancies accumulate into small cavities which are visible as white spots in the under-focused image in Fig. 6. Due to the fact that the vacancy and helium-vacancy clusters initially present in the material before annealing are evenly distributed throughout the grains, the remaining cavities that appear after annealing are also evenly distributed throughout the grains. As the binding energy of helium to small helium–vacancy clusters is very high [44], helium does not dissociate from these clusters even at high temperatures and therefore cannot travel to grain boundaries. By contrast, during high temperature implantation, defects are much more mobile [45] and likely to reach grain boundaries or other sinks before stable helium–vacancy nuclei are able to form. Helium and vacancies therefore diffuse to grain boundaries and free surfaces, resulting in larger cavities with a high density at grain boundaries.

Helium implantation experiments have been performed on coarse-grained iron samples by other groups [17, 19, 20]. In these experiments, helium implantation at room temperature to similar concentrations as performed here resulted in 1–3 nm sized dislocation loops [17, 19, 20],

and post irradiation annealing experiments revealed that the loops slowly grew several nanometers in size between room temperature and 400 °C [19, 20], similar to the results presented in nanocrystalline iron in Fig. 1. Elevated temperature helium implantations to similar concentrations reached in this experiment have also been performed in coarse-grained iron by other groups. During helium implantation in large grained iron at temperatures from 200 °C to 400 °C [17, 18], small dislocation loops were formed of similar size as seen in nanocrystalline iron (Fig. 6). For both room-temperature helium implantation followed by annealing, as well as elevated temperature implantation, similar dislocation loop behavior is observed in coarse-grained and nanocrystalline materials at temperatures up to around 400 °C.

A significant difference in loop behavior in nanocrystalline iron and the large grained iron is evident above 500 °C. Between 500 °C and 600 °C, loops in large grained iron continued to grow during annealing and became large dislocation loops with size around 50 nm, and by 700–800 °C these formed forests of large line dislocations [19, 20]. High temperature implantation at 500 °C in coarse-grained iron also resulted in large dislocation loops on the order of 100 nm in size [17]. In contrast, in the temperature range where loops in large grained materials grew into large dislocation forests, the loops in nanocrystalline iron reached a maximum size of only around 15 nm, and completely annealed out by 600 °C for post-irradiation annealing (Fig. 1) and never formed for the case of high-temperature implantation (Fig. 6). It should be noted that a similar grain size effect on dislocation loops was observed in nanocrystalline iron irradiated with Kr ions [9], where loop growth was stunted in grains below approximately 100 nm diameter. In this case, the author cited the mobility of small nanometer sized dislocation loops, and the associated loss to surfaces and grain boundaries, as the primary limiting factor for loop growth. However, in the current experiment, loop mobility was limited

and loss to grain boundaries was not observed at all. Instead, the loops appear to slowly decrease in size upon annealing and disappear by 600 °C, or never form at all in the case of implantation at 600 °C.

Cavity evolution trends with temperature varied significantly in the helium-implanted coarse-grained iron studies, making comparison to the results seen here difficult, particularly because no attention was paid to cavity structures in the grain core compared to grain boundaries. It does appear that, in general, larger cavities were formed in coarse-grained material than nanocrystalline material. For example, after helium implantation at room temperature in coarse-grained iron to similar concentrations as performed here, visible cavities were present at room temperature which ranged in size from around a nanometer [17] to 10 nm [20] which grew to over 20 nm by around 600 °C [20]. Implantation at elevated temperatures resulted in 2–4 nm cavities during implantation at 400 °C, and 12–20 nm cavities during implantation at 600 °C [18]. In contrast, no cavities were visible in this study until around 600 °C for either thermal condition, and reached sizes of only several nanometers at that temperature. The decrease in cavity size in nanocrystalline iron could be due to the high density of grain boundaries, which can absorb and annihilate Frenkel pairs and act as fast diffusion pathways for helium.

The cavity density at grain boundaries in Fig. 6(b) appears to vary significantly between boundaries. Further work is under way to characterize grain boundary type in relation to cavity density. Dual beam experiments, in which helium is implanted concurrently with heavy-ion irradiation, causing cascade damage, will also be used to further investigate helium interactions with varying defect types.

5 Conclusions

Nanostructured materials are of interest for applications in future nuclear reactors, and developing an understanding of the differences between defect accumulation in nano-grained and bulk materials is critical for designing and developing future materials. During room temperature helium implantation in nanocrystalline iron thin films to approximately 2 dpa, dislocation loops formed by the slow accumulation of point defects. In contrast to previously reported experiments in bulk metals, in which loops continued to grow into large forests of dislocations, the high density of grains limited loop size and resulted in complete disappearance of loops around 600 °C, which also corresponded to the appearance of cavities. The dislocation loops were seen to have limited mobility, and annealed out predominately through shrinking and the annihilation of interstitial loops. A significant difference in final cavity structure was observed depending on thermal conditions. In the case of room temperature implantation followed by annealing, stable bubble nuclei formed evenly distributed throughout the grain structure, which grew to bubbles upon annealing. For high temperature implantation, the increase mobility of defects does not allow for stable nuclei to form; instead, helium and vacancies migrate to sinks such as grain boundaries where they form large cavities.

Acknowledgments

The authors thank Dan Bufford, Dan Buller, Cajer Gong, Osman El-Atwani, James Nathaniel, Mike Marshal, and Sarah Blair for their assistance and insightful discussion. This work is primarily supported by the US Department of Energy's Nuclear Energy University Program (DE-NE0000678). MLT and ACL gratefully acknowledge funding from the United States Department of Energy Basic Energy Sciences (DOE/BES) under the Early Career program

through contract DE-SC0008274. Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

References

- [1] Y. Guerin, G. S. Was, and S. J. Zinkle, "Materials Challenges for Advanced Nuclear Energy Systems," *Mrs Bulletin*, vol. 34, pp. 10-14, Jan 2009.
- [2] G. R. Odette, M. J. Alinger, and B. D. Wirth, "Recent developments in irradiation-resistant steels," *Annual Review of Materials Research*, vol. 38, pp. 471-503, 2008 2008.
- [3] K. Y. Yu, Y. Liu, C. Sun, H. Wang, L. Shao, E. G. Fu, *et al.*, "Radiation damage in helium ion irradiated nanocrystalline Fe," *Journal of Nuclear Materials*, vol. 425, pp. 140-146, Jun 2012.
- [4] I. J. Beyerlein, A. Caro, M. J. Demkowicz, N. A. Mara, A. Misra, and B. P. Uberuaga, "Radiation damage tolerant nanomaterials," *Materials Today*, vol. 16, pp. 443-449, Nov 2013.
- [5] X. Zhang, E. G. Fu, N. Li, A. Misra, Y. Q. Wang, L. Shao, *et al.*, "Design of Radiation Tolerant Nanostructured Metallic Multilayers," *Journal of Engineering Materials and Technology-Transactions of the Asme*, vol. 134, Oct 2012.
- [6] X.-M. Bai, A. F. Voter, R. G. Hoagland, M. Nastasi, and B. P. Uberuaga, "Efficient Annealing of Radiation Damage Near Grain Boundaries via Interstitial Emission," *Science*, vol. 327, pp. 1631-1634, Mar 26 2010.
- [7] J. W. Corbett, R. B. Smith, and R. M. Walker, "RECOVERY OF ELECTRON-IRRADIATED COPPER .2. INTERSTITIAL MIGRATION," *Physical Review*, vol. 114, pp. 1460-1472, 1959.
- [8] C. J. Ortiz and M. J. Caturla, "Simulation of defect evolution in irradiated materials: Role of intracascade clustering and correlated recombination," *Physical Review B*, vol. 75, May 2007.
- [9] G. A. Vetterick, "Radiation Damage in Nanocrystalline Iron," PhD, Drexel University, 2014.
- [10] M. Rose, A. G. Balogh, and H. Hahn, "Instability of irradiation induced defects in nanostructured materials," *Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms*, vol. 127, pp. 119-122, May 1997.
- [11] W. Han, E. G. Fu, M. J. Demkowicz, Y. Wang, and A. Misra, "Irradiation damage of single crystal, coarse-grained, and nanograined copper under helium bombardment at 450 degrees C," *Journal of Materials Research*, vol. 28, pp. 2763-2770, Oct 2013.
- [12] O. El-Atwani, J. A. Hinks, G. Greaves, S. Gonderman, T. Qiu, M. Efe, *et al.*, "In-situ TEM observation of the response of ultrafine- and nanocrystalline-grained tungsten to extreme irradiation environments," *Scientific Reports*, vol. 4, May 6 2014.

- [13] N. Nita, R. Schaeublin, and M. Victoria, "Impact of irradiation on the microstructure of nanocrystalline materials," *Journal of Nuclear Materials*, vol. 329, pp. 953-957, Aug 1 2004.
- [14] C. Sun, M. Song, K. Y. Yu, Y. Chen, M. Kirk, M. Li, *et al.*, "In situ Evidence of Defect Cluster Absorption by Grain Boundaries in Kr Ion Irradiated Nanocrystalline Ni," *Metallurgical and Materials Transactions a-Physical Metallurgy and Materials Science*, vol. 44A, pp. 1966-1974, Apr 2013.
- [15] H. Trinkaus and B. N. Singh, "Helium accumulation in metals during irradiation - where do we stand?," *Journal of Nuclear Materials*, vol. 323, pp. 229-242, Dec 1 2003.
- [16] H. Ullmaier, "THE INFLUENCE OF HELIUM ON THE BULK PROPERTIES OF FUSION-REACTOR STRUCTURAL-MATERIALS," *Nuclear Fusion*, vol. 24, pp. 1039-1083, 1984 1984.
- [17] K. Arakawa, R. Imamura, K. Ohota, and K. Ono, "Evolution of point defect clusters in pure iron under low-energy He⁺ irradiation," *Journal of Applied Physics*, vol. 89, pp. 4752-4757, May 2001.
- [18] K. Ono, K. Arakawa, and K. Hojou, "Formation and migration of helium bubbles in Fe and Fe-9Cr ferritic alloy," *Journal of Nuclear Materials*, vol. 307, pp. 1507-1512, Dec 2002.
- [19] R. Sugano, K. Morishita, A. Kimura, H. Iwakiri, and N. Yoshida, "Microstructural evolution in Fe and Fe-Cr model alloys after He⁺ ion irradiations," *Journal of Nuclear Materials*, vol. 329, pp. 942-946, Aug 2004.
- [20] S. Moll, T. Jourdan, and H. Lefaix-Jeuland, "Direct Observation of Interstitial Dislocation Loop Coarsening in alpha-Iron," *Physical Review Letters*, vol. 111, Jul 2013.
- [21] K. Hattar, D. M. Follstaedt, J. A. Knapp, and I. M. Robertson, "Defect structures created during abnormal grain growth in pulsed-laser deposited nickel," *Acta Materialia*, vol. 56, pp. 794-801, Feb 2008.
- [22] K. Hattar, D. C. Bufford, and D. L. Buller, "Concurrent in situ ion irradiation transmission electron microscope," *Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms*, vol. 338, pp. 56-65, Nov 1 2014.
- [23] J. F. Ziegler, M. D. Ziegler, and J. P. Biersack, "SRIM - The stopping and range of ions in matter (2010)," *Nuclear Instruments & Methods in Physics Research Section B-Beam Interactions with Materials and Atoms*, vol. 268, pp. 1818-1823, Jun 2010.
- [24] M. L. Jenkins and M. A. Kirk, *Characterization of radiation damage by transmission electron microscopy*: CRC Press, 2000.
- [25] Z. Yao, M. Hernandez-Mayoral, M. L. Jenkins, and M. A. Kirk, "Heavy-ion irradiations of Fe and Fe-Cr model alloys Part 1: Damage evolution in thin-foils at lower doses," *Philosophical Magazine*, vol. 88, pp. 2851-2880, 2008.
- [26] Z. Yao, M. L. Jenkins, M. Hernandez-Mayoral, and M. A. Kirk, "The temperature dependence of heavy-ion damage in iron: A microstructural transition at elevated temperatures," *Philosophical Magazine*, vol. 90, pp. 4623-4634, 2010.
- [27] K. Arakawa, H. Mori, and K. Ono, "Formation process of dislocation loops in iron under irradiations with low-energy helium, hydrogen ions or high-energy electrons," *Journal of Nuclear Materials*, vol. 307, pp. 272-277, Dec 2002.
- [28] J. S. Bryner, "A STUDY OF NEUTRON IRRADIATION DAMAGE IN IRON BY ELECTRON-TRANSMISSION MICROSCOPY," *Acta Metallurgica*, vol. 14, pp. 323-&, 1966.

- [29] I. M. Robertson, M. L. Jenkins, and C. A. English, "LOW-DOSE NEUTRON-IRRADIATION DAMAGE IN ALPHA-IRON," *Journal of Nuclear Materials*, vol. 108, pp. 209-221, 1982.
- [30] B. L. Eyre and A. F. Bartlett, "AN ELECTRON MICROSCOPE STUDY OF NEUTRON IRRADIATION DAMAGE IN ALPHA-IRON," *Philosophical Magazine*, vol. 12, pp. 261-&, 1965.
- [31] H. Kayano, H. Yoshinaga, K. Abe, and S. Morozumi, "EFFECTS OF NEUTRON-IRRADIATION ON MECHANICAL-PROPERTIES OF IRON-NITROGEN ALLOYS," *Journal of Nuclear Science and Technology*, vol. 15, pp. 200-212, 1978.
- [32] L. L. Horton, J. Bentley, and W. A. Jesser, "THE MICROSTRUCTURE OF TRIPLE-BEAM ION IRRADIATED FE AND FE-CR ALLOYS," *Journal of Nuclear Materials*, vol. 103, pp. 1085-1089, 1982.
- [33] L. L. Horton, J. Bentley, and W. A. Jesser, "THE DEPTH DISTRIBUTION OF DISPLACEMENT DAMAGE IN ALPHA-IRON UNDER TRIPLE BEAM ION IRRADIATION," *Journal of Nuclear Materials*, vol. 103, pp. 1343-1347, 1982.
- [34] B. C. Masters, "DISLOCATION LOOPS IN IRRADIATED IRON," *Nature*, vol. 200, pp. 254-&, 1963.
- [35] I. M. Robertson, M. A. Kirk, and W. E. King, "FORMATION OF DISLOCATION LOOPS IN IRON BY SELF-ION IRRADIATIONS AT K-40," *Scripta Metallurgica*, vol. 18, pp. 317-320, 1984.
- [36] M. L. Jenkins, C. A. English, and B. L. Eyre, "HEAVY-ION IRRADIATION OF ALPHA-IRON," *Philosophical Magazine a-Physics of Condensed Matter Structure Defects and Mechanical Properties*, vol. 38, pp. 97-114, 1978.
- [37] T. M. Robinson, "XENON ION IRRADIATION OF ALPHA-FE," *Physica Status Solidi a-Applied Research*, vol. 75, pp. 243-249, 1983.
- [38] M. R. Gilbert, Z. Yao, M. A. Kirk, M. L. Jenkins, and S. L. Dudarev, "Vacancy defects in Fe: Comparison between simulation and experiment," *Journal of Nuclear Materials*, vol. 386-88, pp. 36-40, Apr 2009.
- [39] N. Yoshida, M. Kiritani, and F. E. Fujita, "ELECTRON RADIATION-DAMAGE OF IRON IN HIGH-VOLTAGE ELECTRON-MICROSCOPE," *Journal of the Physical Society of Japan*, vol. 39, pp. 170-179, 1975.
- [40] M. R. Gilbert, S. L. Dudarev, P. M. Derlet, and D. G. Pettifor, "Structure and metastability of mesoscopic vacancy and interstitial loop defects in iron and tungsten," *Journal of Physics-Condensed Matter*, vol. 20, Aug 27 2008.
- [41] K. Arakawa, K. Ono, M. Isshiki, K. Mimura, M. Uchikoshi, and H. Mori, "Observation of the one-dimensional diffusion of nanometer-sized dislocation loops," *Science*, vol. 318, pp. 956-959, Nov 2007.
- [42] K. Arakawa, M. Hatanaka, E. Kuramoto, K. Ono, and H. Mori, "Changes in the burgers vector of perfect dislocation loops without contact with the external dislocations," *Physical Review Letters*, vol. 96, Mar 2006.
- [43] D. Brimbal, B. Decamps, J. Henry, E. Meslin, and A. Barbu, "Single- and dual-beam in situ irradiations of high-purity iron in a transmission electron microscope: Effects of heavy ion irradiation and helium injection," *Acta Materialia*, vol. 64, pp. 391-401, Feb 2014.
- [44] C. J. Ortiz, M. J. Caturla, C. C. Fu, and F. Willaime, "He diffusion in irradiated alpha-Fe: An ab-initio-based rate theory model," *Physical Review B*, vol. 75, Mar 2007.

- [45] D. Terentyev, N. Juslin, K. Nordlund, and N. Sandberg, "Fast three dimensional migration of He clusters in bcc Fe and Fe-Cr alloys," *Journal of Applied Physics*, vol. 105, May 15 2009.