Effect of Oxide Particles and Pre-Implanted Helium on Defect Evolution during Electron Irradiation

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In-situ observations of ferritic/martensitic steels by electron irradiation with a 1.25 MeV high voltage microscope at 573 K were carried out to study damage evolution in the steels. The development of interstitial type loops and cavities in both of the two steels, F82H-IEA and F82H-ODS, showed smaller and more numerous defects in the ODS steel. The cavities were formed preferentially at the interface between oxide particles and matrix. The results suggest that ODS particles may function to suppress the nucleation and growth of loops and cavities arising from irradiation. The effect of pre-implanted helium was also studied. The pre-implanted helium led to a homogenous distribution of black dots and cavities in the steels, and these may act as sinks for point defects arising from irradiation, causing a suppression of the subsequent growth of loops and cavities. The hardening corresponding to the microstructural evolution was estimated by assuming parameters extracted from the ion irradiation. [doi:10.2320/matertrans.MD201318]

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1. Introduction

Oxide dispersion strengthened (ODS) ferritic/martensitic steel is a candidate for structural components in fusion reactors because of its superior radiation resistance and elevated temperature strength. Previous studies have shown that mechanically-alloyed ODS alloys exhibit high densities ($\sim 10^{23} \text{ m}^{-3}$) of nanoscale (2–5 nm diameter) particles with Y, Ti, and O.^{1,2)} It has been reported that the presence of a fine dispersion of oxide particles enhances the thermal stability and improves the mechanical properties at high temperatures.^{3–6)} A further important factor to consider in structural materials for fusion reactors is that high energy neutrons at 14 MeV energy may cause transmuted helium by reactions with the atoms making up the materials, and this degrades the mechanical properties of the materials, through swelling, embrittlement, and hardening of the irradiated components. The interface between the high density oxide and matrix is considered to act as a sink for such helium atoms, as well as vacancies, to suppress nucleation and growth of cavities.^{5,7–9)} However, further investigation is required for a more complete understanding of the suppression mechanism of the oxide particles.

The aim of this study is to investigate the role of oxide particles in the nucleation and growth of loops and cavities, and the effect of pre-injected helium on defect clustering and cavity growth.

2. Experimental Procedure

Two steels, F82H-IEA and F82H-ODS (both provided by the Japan Atom Energy Agency), were fabricated into 3 mm diameter disks with a thickness of 0.12 mm, followed by electrochemical polishing in an electrolyte of acetic acid and perchloric acid (19 : 1). The chemical compositions and heat treatment conditions of the specimens of the two steels are shown in Table 1.^{10,11)}

Table 1 Chemical compositions and heat treatment conditions of the RAFM and ODS steel (mass%) used here.

	C	Si	Cr	W	V	Ta	Ti	Y ₂ O ₃	Ex. O
F82H-IEA* ¹	0.090	0.11	7.71	1.95	0.16	0.02	—	—	—
F82H-ODS* ²	0.16	0.013	7.85	1.9	0.18	0.10	0.19	0.368	0.092

*¹ normalized at 1040 C for 40 min and tempered at 750 C for 1 h.

*² normalized at 1050 C for 2 h and tempered at 750 C for 1 h.

Fig. 1 SRIM calculations for the He implantation, with the concentration 0.2 at.% and displacement damage 0.07 dpa.

Helium ions at 70 and 140 keV were injected into the electro-polished samples at room temperature. Figure 1 shows the Stopping and Range of Ions in Matter (SRIM) calculations of the profile of the implanted helium and displacement damage.

The subsequent electron irradiation experiment was performed in a high voltage electron microscope (JEOL HRHVEM 1300) operated at 1.25 MeV at a temperature of 573 K. The dose rate was $3.9\text{--}4.8 \times 10^{-3} \text{ dpa/s}$.

3. Results

Martensitic steel F82H-IEA has prior austenite grain boundaries and martensite packets composed of laths. The F82H-ODS was developed based on F82H-IEA and

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contains fine oxide particles ((3.8 ± 1.3) nm diameter and $(1.1 \pm 0.1) \times 10^{23} \text{ m}^{-3}$). Figure 2 shows bright field images of the as-implanted F82H-IEA and F82H-ODS steels. Black dots and cavities with a homogeneous diameter distribution of around 1.7 nm can be observed in the matrix, formed by the helium pre-implantation.

During the subsequent electron irradiation, loops were formed, and continued to grow without changes in the density, as shown in Figs. 3 and 4. The development of a typical interstitial loop is marked with white arrows in the two figures, respectively. Figure 5 shows a dose dependence of the loop size and the number density. The data shows that the loop size increases with increases in the electron dose. The loop size is smaller and the number density is higher in

F82H-ODS than in F82H-IEA. Further, the growth rate of the loops in F82H-ODS is slower than in F82H-IEA, as shown by the flatter slopes in Fig. 5(a). These results suggest that the oxide particles in F82H-ODS could play a role to suppress the growth of loops and increase the number of nucleation sites of defects in electron irradiation.

The specimens with pre-implanted He have a larger loop size but a lower growth rate than the specimens without He. The increases in the loop size occur because the black dots were already present in the samples before the electron irradiation. The loop number density in the pre-implanted specimens is higher than in the specimens without He pre-implantation. Overall the results suggest that the pre-existing black dots and cavities may be considered to act as sinks for point defects in electron irradiation.

Figure 6 shows the evolution of cavities with electron irradiation of F82H-IEA, F82H-ODS, with and without He, respectively. Cavities were formed in all the four samples during the irradiation. In F82H-IEA, the cavities grow quickly and merge with increases in the irradiation dose. Compared to cavities in F82H-IEA, the cavity size in the other three samples is rather smaller, and the cavities grow more slowly and present a more homogeneous distribution. The cavities locating at the interface between oxide particles and matrix are marked with black arrows. The preferential

Fig. 2 TEM images of He-implanted F82H-IEA and F82H-ODS.

Fig. 3 I-loop evolution in He-implanted F82H-IEA after electron irradiation at 573 K up to 10 dpa. The white arrows indicate the development of a typical I-loop in F82H-IEA with He.

Fig. 4 I-loop evolution in He-implanted F82H-ODS after electron irradiation at 573 K up to 12 dpa. The white arrows indicate the development of a typical I-loop in F82H-ODS with He.

Fig. 5 Dose dependence of (a) loop mean size and (b) number density in F82H-IEA and F82H-ODS.

formation of cavities at these positions at the interface may be a cause of the homogeneous distribution of cavities in these samples.

Figure 7 plots the dose dependence of the cavity size and number density in the specimens. The cavity size increases with increases in the irradiation dose. The cavities in F82H-

ODS are smaller, have a lower growth rate and the cavities display a lower number density than that in F82H-IEA, which suggests that the oxide particles could play a role to suppress the cavity development in F82H-ODS. Further, the cavity size is larger and the number density is lower in the as-received samples than in the helium implanted samples, and the cavities in the as-received samples grow faster than in the helium implanted samples, especially with F82H-IEA.

Figure 8 shows the cavity distribution in the different samples. In F82H-IEA, cavities are preferentially formed around the lath boundary, while in the helium pre-implanted samples, the cavities distribute homogenously in the martensitic matrix. This suggests that the pre-injected helium suppresses the growth of cavities in F82H-IEA during irradiation. The reason for this may be hypothesized to be that the pre-existing high number density of cavities and black dots could act as sinks for radiation-induced point defects. In F82H-ODS, there is also a homogenous distribution of cavities, suggesting that the high number density of fine oxide particles could also act as a sink for point defects arising from the irradiation. Similar suppression effects of helium on the formation of loops and cavities has been reported in Si-modified 316 stainless steel.¹²⁾

4. Discussion

Electron irradiation may give rise to much higher concentrations of point defects than the thermal equilibrium concentration, therefore excess point defects diffuse to sink sites such as free surfaces, grain boundaries, and other interfaces present in metals, or they nucleate on defect clusters.¹³⁾ The F82H-ODS steel contains a high number density of nano-scale particles which provide interfaces between oxide particles and matrix as sink sites for point defects. Pre-implantation of helium introduces black dots and cavities that could also act as sink sites. The high number density of sink sites could provide an avenue to enable homogenous absorption of point defects and remove vacancies and interstitial defects by interstitials re-emitted at the interface. Nanostructured components that include a large fraction of interfaces can be expected to benefit from the ability to remove radiation-induced point defects and control helium bubble nucleation and growth to prevent embrittlement.

Fig. 6 Cavity growth under electron irradiation at 573 K in the samples investigated here. The black arrows indicate cavities located at the interface between oxide particles and matrix.

Fig. 7 Dose dependence of (a) the cavity mean size and (b) number density in F82H-IEA and F82H-ODS with and without He-implantation.

Fig. 8 Cavity distributions in F82H-IEA, helium implanted F82H-IEA, and F82H-ODS.

Fig. 9 Estimated hardening as a function of electron irradiation dose for F82H-IEA and F82H-ODS.

High voltage electron microscope is useful for *in-situ* observations, but it has not been used in estimations of mechanical property changes in irradiation damage studies. However, the mechanical properties could be estimated with experimental factors in ion irradiation. The effect of irradiation damage on the tensile yield stress is interpreted by the Orowan model:¹⁴⁾

$$\Delta\sigma_y = M\alpha\mu b\sqrt{Nd}.$$

Here, M is the Taylor factor, α is the barrier strength of obstacles, μ is the shear modulus of the material matrix, b is the magnitude of burgers vector of moving dislocations, N is the number density of obstacles, and d is the mean diameter of obstacles. Subsequently, the hardening effect is estimated by $\Delta H[\text{GPa}] = \Delta\sigma_y[\text{MPa}]/270$.¹⁵⁾ The tentative calculations of the loop and cavity hardening in F82H-IEA and F82H-ODS were performed by assuming $M = 3.06$, $\mu = 77 \text{ GPa}$, $b = 0.3308 \text{ nm}$, $\alpha = 0.25$ ¹⁶⁾ for loops and $\alpha = 0.5$ ¹⁷⁾ for cavities. The estimated hardening from defects as a function of the dose is shown in Fig. 9. By this estimate, the result showed that the hardening of F82H-ODS to be lower than F82H-IEA.

5. Conclusions

The irradiation induced point defects occur at the interface between nano-scale oxide particles and matrix. The oxide particles in F82H-ODS suppressed the formation and growth of loops and cavities, resulting in a homogenous distribution

of defects. Pre-injected helium induced a homogenous distribution of black dots and cavities, which suppressed the subsequent growth of loops and cavities. Oxide particles in F82H-ODS and the defects induced by the pre-implanted helium may be assumed to act as sinks for point defects arising with electron irradiation.

By a simple application of the Orowan equation and considering experimental conditions, it was possible to make a qualitative estimate of the level of irradiation hardening resulting from electron irradiation.

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REFERENCES

- 1) R. L. Klueh, J. P. Shingledecker, R. W. Swindeman and D. T. Hoelzer: *J. Nucl. Mater.* **341** (2005) 103–114.
- 2) D. J. Larson, P. J. Maziasz, I.-S. Kim and K. Miyahara: *Scr. Mater.* **44** (2001) 359–364.
- 3) G. R. Romanoski, L. L. Snead, R. L. Klueh and D. T. Hoelzer: *J. Nucl. Mater.* **283–287** (2000) 642–646.
- 4) S. Ukai and M. Fujiwara: *J. Nucl. Mater.* **307–311** (2002) 749–757.
- 5) V. de Castro, E. A. Marquis, S. Lozano-Perez, R. Pareja and M. L. Jenkins: *Acta Mater.* **59** (2011) 3927–3936.
- 6) M. K. Miller, E. A. Kenik, K. F. Russell, L. Heatherly, D. T. Hoelzer and P. J. Maziasz: *Mater. Sci. Eng. A* **353** (2003) 140–145.
- 7) G. R. Odette, M. J. Alinger and B. D. Wirth: *Annu. Rev. Mater. Res.* **38** (2008) 471–503.
- 8) I.-S. Kim, J. D. Hunn, N. Hashimoto, D. L. Larson, P. J. Maziasz, K. Miyahara and E. H. Lee: *J. Nucl. Mater.* **280** (2000) 264–274.
- 9) K. Yutani, H. Kishimoto, R. Kasada and A. Kimura: *J. Nucl. Mater.* **367–370** (2007) 423–427.
- 10) H. Tanigawa, A. Sawahata, M. A. Sokolov, M. Enomoto, R. L. Klueh and A. Kohyama: *Mater. Trans.* **48** (2007) 570–573.
- 11) K. Shinozuka, H. Esaka, M. Tamura and H. Tanigawa: *J. Nucl. Mater.* **417** (2011) 233–236.
- 12) T. Takeyama, S. Ohnuki, H. Takahashi and M. Uchiyama: *J. Nucl. Mater.* **122** (1984) 547–551.
- 13) H. Takahashi, Y. Hidaka and S. Ohnuki: *Micron* **27** (1996) 239–246.
- 14) E. Orowan: *Symposium on Internal Stresses in Metals and Alloys*, (Institute of Metals, London, 1948) pp. 451–453.
- 15) P. M. Rice and R. E. Stoller: *MRS Proceedings* **649** (2000) Q7–11.
- 16) G. E. Lucas: *J. Nucl. Mater.* **206** (1993) 287–305.
- 17) M. Ando, Y. Katoh, H. Tanigawa, A. Kohyama and T. Iwai: *J. Nucl. Mater.* **283–287** (2000) 423–427.