Materials Transactions, Vol. 43, No. 4 (2002) pp. 622 to 626
Special Issue on Radiation-Induced Effects by High Energy Particle/Solid Interaction

Effects of Helium on Radiation Behavior in Low Activation Fe–Cr–Mn Alloys

Hu Benfu[1], Hiroshi Kinoshita[2], Tamaki Shibayama[2] and Heishichiro Takahashi[2]

[1]*Department of Materials Science and Engineering, University of Science and Technology, Beijing 100083 P.R. China*
[2]*Center for Advanced Research of Energy Technology, Hokkaido University, Sapporo 060-8628, Japan*

In order to investigate the effects of helium on the microstructural evolution in Fe–Cr–Mn (W, V) alloy during three irradiation modes of electron beam, electron/He^+-ion dual-beam and electron irradiation after helium pre-injection, in situ observations were carried out using a high voltage electron microscope connected with an ion accelerator. The interstitial-type dislocation loops and small voids were formed in the early stage of irradiation. The average void size was smaller and void number density was higher under irradiation condition by electrons after helium pre-injection and by dual-beam in comparison with the case of electron irradiation. Irradiation-induced grain boundary segregation of Cr and Mn was suppressed due to existence of helium. From these results it is suggested that helium strongly influences dislocation structural development and void nucleation during irradiation. The effect of interactions between helium and point defects is discussed.

(Received December 7, 2001; Accepted February 25, 2002)

Keywords: *void, helium effect, low activation alloy, electron irradiation, dual-beam irradiation*

1. Introduction

In the new century the world will face the needs of new energy sources. Generating electricity by nuclear fusion reactor is one of the most attractive long-term energy options. As one of the materials for this fusion reactor, nonmagnetic austenitic steel with high manganese is attractive and prospective because of its lower induced radioactivity. The mechanical properties and irradiation damage resistance of higher manganese steels are, however, inferior than Fe–Cr–Ni austenitic stainless steel.[1,2] Fusion reactor materials will be exposed to fusion neutrons with energy of 14 MeV. Radiation damage of materials in a fusion reactor environment is characterized by synergistic effects of the cascade damage and nuclear transmutation products such as hydrogen and helium. The studies on development of Fe–Cr–Mn instead of Fe–Cr–Ni have been done. However, the details on irradiation damage and phase stability of Fe–Cr–Mn alloy under irradiation with helium is not clarified.

In this study, observations of irradiation damage characteristics by electron beam irradiation, electron irradiation after helium injection and dual-beam with electron-helium ion for Fe–Cr–Mn (W, V) alloy are carried out using a high voltage electron microscope connected with a high energy ion accelerator to simulate the damage behaviors under fusion condition and also to follow the effect of He as a transmutant.

2. Experimental Procedures

A Fe–Cr–Mn (W, V) alloy used has the following chemical compositions: C:0.26, Cr:12.5, Mn:16.57, Si:0.26, Ni:1.00, V:0.94, W:2.00, S:0.013, P:0.001, N:0.12. The alloy was prepared from high-purity raw materials, and it was remelted in a vacuum induction furnace to reduce the impurities such as O and H. The sample was annealed at 1323 K for 1 h in a sealed quarts tube at a vacuum of 6×10^{-6} Pa after forging, and then the disk specimens with 0.2 mm thickness and 3 mm diameter for TEM observation were punched out and electorochemically-jet polished.

Dual-beam irradiations were performed in a HVEM (JEH-ARM1300) and an ion accelerator (300 keV). The electron irradiation was carried out with a damage rate of about 2×10^{-3} dpa/s at 673 K to doses up to 10 dpa. Helium injection was done with 70 appm/dpa using ion accelerator with 100 keV accelerating voltage. The irradiation was performed by three kinds of irradiation modes; 1) single-electron beam irradiation, 2) electrons/He^+-ions dual-beam irradiation, and 3) electron irradiation after He pre-injection.

The concentration of solute elements after irradiation was analyzed with an energy dispersive x-ray spectroscopy across a grain boundary including both of irradiated and unirradiated regions.

3. Experimental Results

3.1 Microstructural change during irradiation

Figure 1 shows the microstructural evolution in alloy after irradiation with three irradiation modes. During single-electron beam irradiation to 2.5 dpa the fine black dots contrast were observed inside grain and along grain boundary. The black dots were partially observed as stacking fault tetrahedra and interstitial dislocation loops with increasing irradiation to 5 dpa. The microstructures by electron-irradiation after helium injection were obviously different from that of single-electron beam irradiation. Namely interstitial type dislocation loops with stacking fault were preferentially formed after 2.5 dpa. The dislocation loops grew with increasing irradiation dose. Due to further irradiation to 4.2 dpa the dislocation density increased and the voids were observed.

In the case of dual-beam irradiation the density of interstitial dislocation loops initially was very high after 2.5 dpa, and voids formation was recognized inside stacking fault loops. The stacking faults gradually transformed to perfect dislocation loops with increasing of dose. Finally a number of fine voids were formed on the dislocation loops and in the matrix.

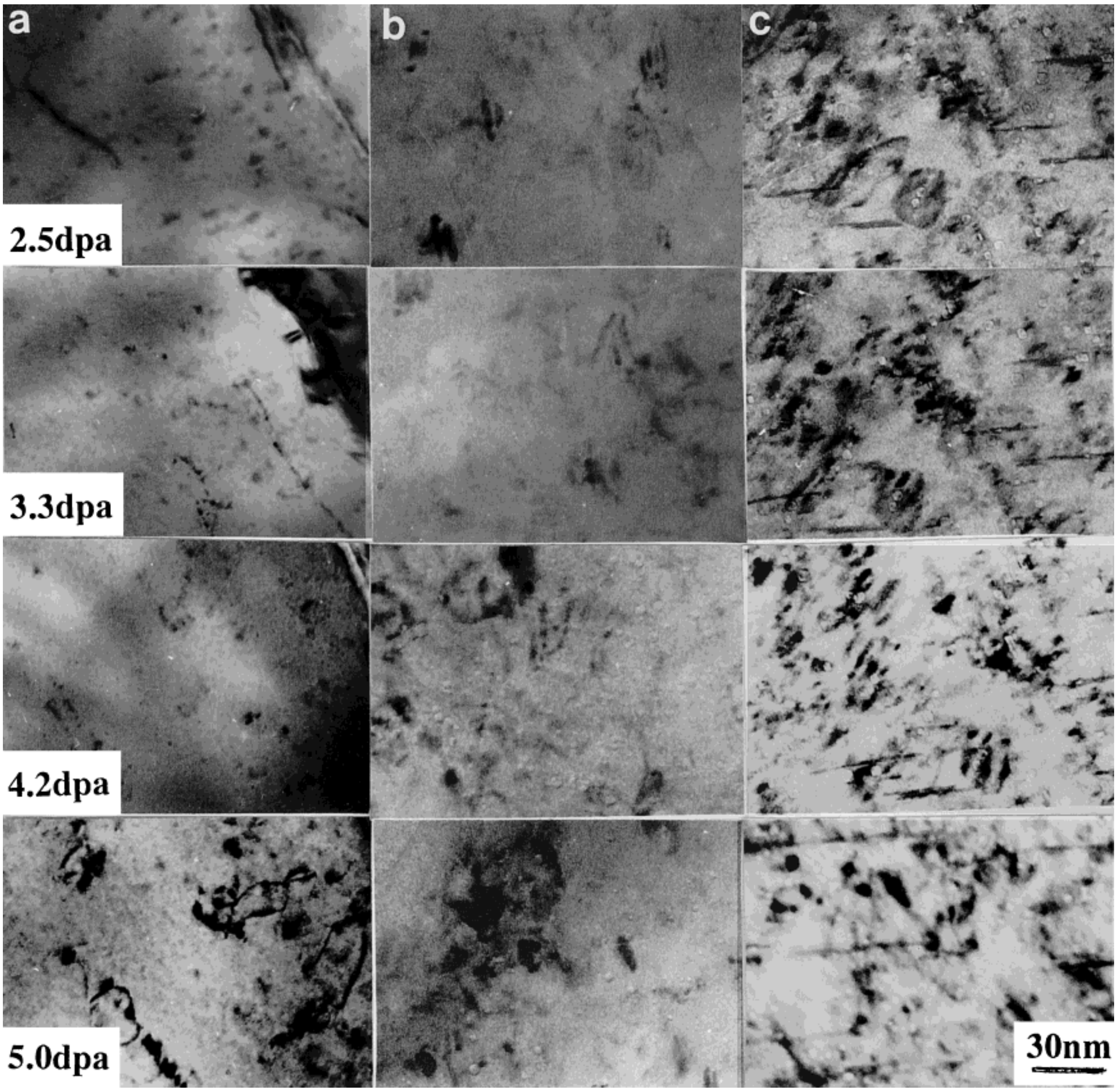

Fig. 1 Damage microstructures observed in Fe–Cr–Mn (W, V) alloy irradiated by three different modes; (a) single Electron (e^-) irradiation, (b) Electron irradiation after helium injection ($He^+ + e^-$), (c) Helium/electron dual beam irradiation ($He^+/e)^-$.

3.2 Void formation and growth

Figure 2 shows the distribution of voids after irradiation to 10 dpa by three different irradiation modes. It can be seen that under electron beam irradiation the void number density was lower and the average void size was about 2.5 nm. However, the void number density after irradiation by electrons after helium injection became very high and the average void size was about 1.3 nm. The void distribution was heterogeneous. Under the dual-beam irradiation the void number density became rather lower than that of irradiation by electron after helium injection, and the average void size became larger.

Figure 3 shows the dose dependence of void size and void number density for the three different irradiation modes. With increasing the dose, the void size and the number density at a given dose up to 6 dpa increased and at higher dose the void size tended to saturate for all of irradiation modes. Especially the void number density nucleated due to electron irradiation after helium pre-injection was higher than other irradiation modes. Evidently the injected helium influences the void nucleation in the early stage of irradiation.[3)]

3.3 Radiation-induced solute distribution near grain boundary

Figure 4 shows the Fe, Cr, Mn solute distribution in the vicinity of the grain boundary after electron irradiation to 10 dpa at 673 K. Both Cr and Mn concentrations were slightly depleted at the grain boundary. Cr, and Mn approached to average concentration of matrix away from grain boundary.

Figure 5 shows the Fe, Cr and Mn concentrations after irradiation to 10 dpa in the presence of helium. Figure 5(a) is the case of electron irradiation after He injection and (b) is the result of electrons/He^+-ion dual-beam irradiation. Fe, Cr and Mn concentrations at grain boundary were not changed in detectable magnitude.

3.4 Void swelling

Figure 6 shows the dose dependence of sink strength related to void number density. The sink strength decreased according to the following order of the electron irradiation after helium injection, electrons/He^+ ions dual beam irradiation and electron irradiation. These results show that pre-injection

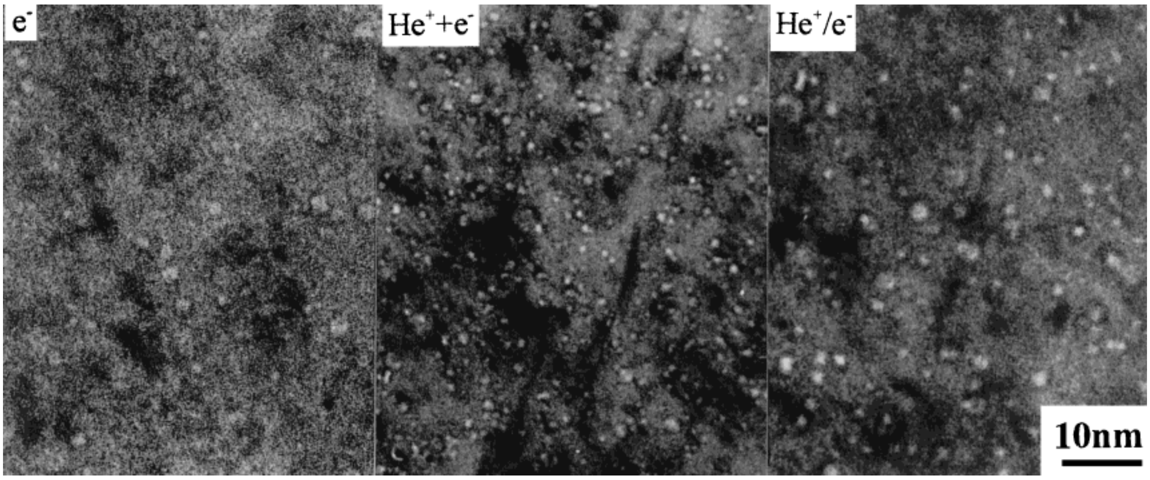

Fig. 2 Void structures observed in Fe–Cr–Mn (W, V) alloy irradiated at 673 K to 10 dpa by three different irradiation modes.

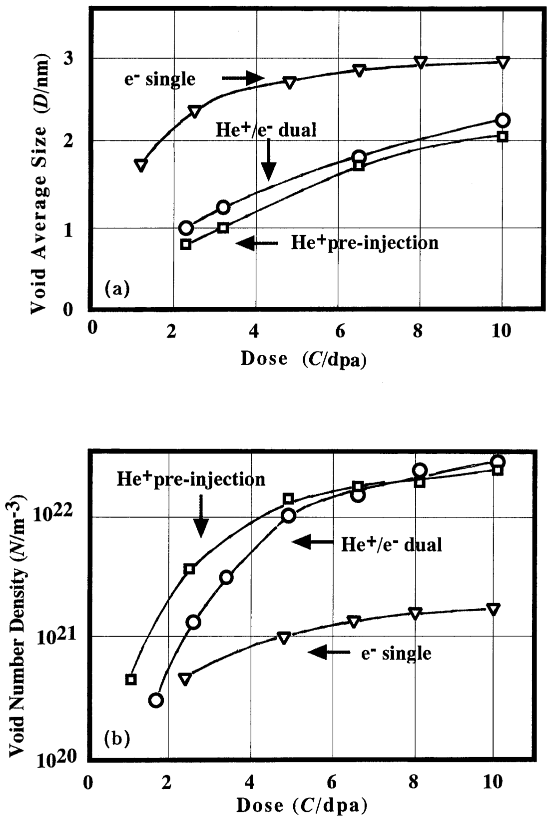

Fig. 3 Dose dependence of the average size and the number density of voids in Fe–Cr–Mn (W, V) alloy.

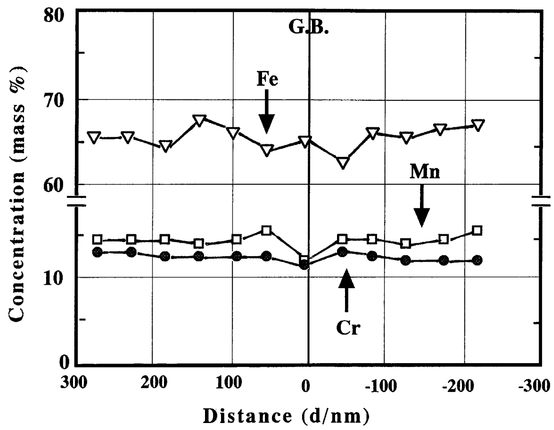

Fig. 4 Solute distribution in the vicinity of grain boundary in Fe–Cr–Mn (W, V) alloy by electron irradiation to 10 dpa at 673 K.

of helium enhances void nucleation as a result of interaction between helium and point defects, mainly with vacancies.

The results of void swelling are shown in Fig. 7. The void swellings of Fe–Cr–Mn alloy irradiated by three different modes were very low as a whole. In the case of helium injection the void swelling gradually increased, and the swelling after dual-beam irradiation to 10 dpa was about 0.03%. Comparing with the swelling of single-electron irradiation (0.009% at 10 dpa), it was about three times higher. It is also clarified that the void swelling rate is influenced by the existence of helium. That is, the swelling rates during irradiation with helium were higher than that of simple electron irradiation.

4. Discussions

4.1 Effects of helium on the secondary defects behaviors in the early stage of irradiation

It is well known that due to collision of electrons having high energy and high speed with lattice atoms in crystal the interstitials and vacancies are introduced, but about 10 percent (10%) of these defects disappear by mutual recombination, and others are remained in the crystals as free defects.[4] However, behaviors of these point defects are obviously influenced by helium existence. Helium atoms injected may exist in interstitial sites or substitutional sites in alloy. But the solubility of helium is very low, and also the activation energy for migration of helium is about 0.2 eV.[5,6] These facts suggest that helium atoms are relatively mobile. On the other hand, when helium occupy substitutional sites and form He–V pairs, these pairs act as void nucleation site so that promote void formation.[7]

When the alloy is electron-irradiated, point defects of interstitials and vacancies are introduced and the helium atoms will preferentially interact with vacancies rather than interstitials. The vacancies trapped by helium are less mobile and act as void nucleation site by absorbing freely migrating vacan-

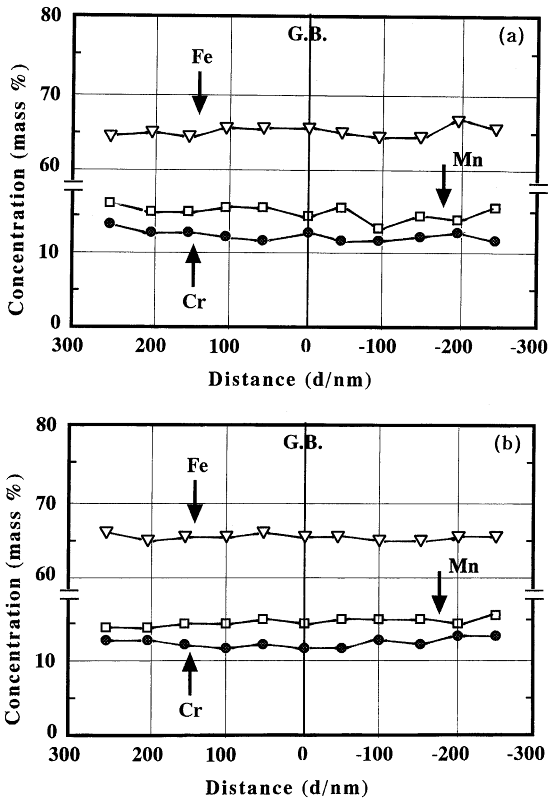

Fig. 5 Solule distribution in the vicinity of grain boundary in Fe–Cr–Mn (W, V) alloy irradiated to 10 dpa at 673 K. (a) electron irradiation after He injection, (b) electrons/He^+-ion dual-beam irradiation.

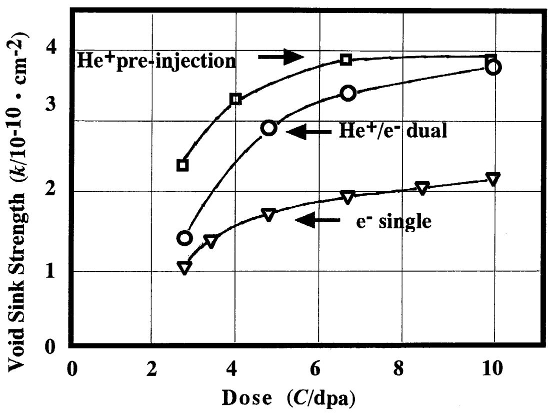

Fig. 6 Dose dependence of void sink strength in the Fe–Cr–Mn (W, V) alloy.

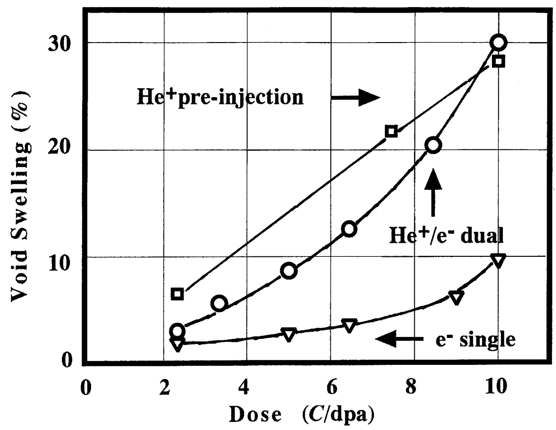

Fig. 7 Dose dependence of void swelling in Fe–Cr–Mn (W, V) alloy to 10 dpa at 673 K.

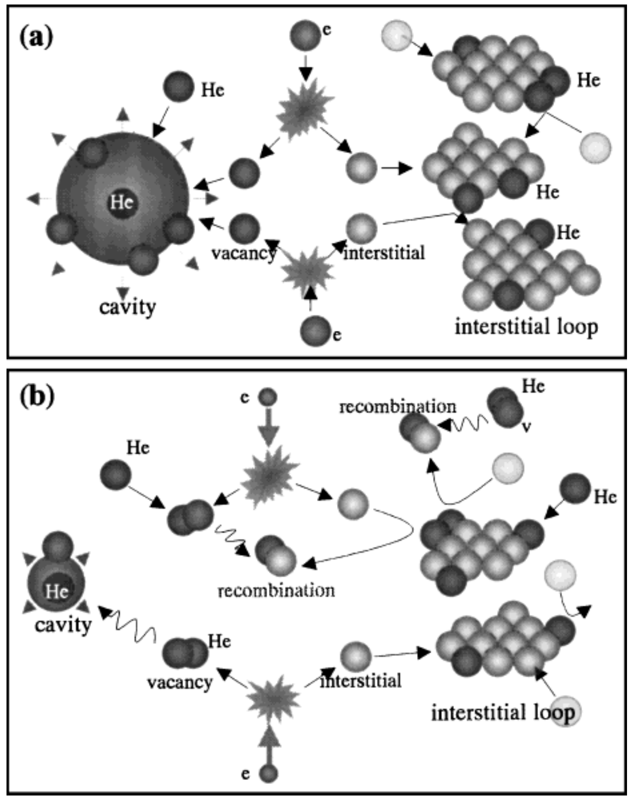

Fig. 8 Schematic drawing of secondary defects, (a) electron irradiation after He injection, (b) electrons/He^+-ion dual-beam irradiation.

cies introduced during electron irradiation, while more mobile interstitials agglomerate to form Frank type loops on the {111} plane.[8] Thus, in the case of electron irradiation after helium injection, voids are easily nucleated with higher number density in the early stage of irradiation. The scheme of interaction of point defects and secondary defects with helium under electron irradiation is shown in Fig. 8(a).

On the other hand, in the case of dual-beam irradiation, since helium and point defects are introduced simultaneously, the introduced vacancies will immediately interact with helium atoms without migration long distance. Also interstitials can easily migrate so that the interstitials meet with vacancies trapped with helium, namely the trapped vacancies and migrating interstitials will recombine frequently. Consequently the nucleation and growth of interstitial type faulted dislocation loops are retarded. Therefore the density of interstitial type faulted loops becomes lower in comparison with electron irradiation after helium injection. The scheme of formation of secondary defects by dual-beam irradiation is shown in Fig. 8(b).

4.2 Effect of helium on the void swelling

It has been observed that extrinsic Frank loops with stacking faults are first nucleated as a result of agglomeration of interstitials on the {111} planes in alloy with lower stacking fault energy. In growth process of faulted loops which were initially nucleated as the intrinsic vacancy type, small loops are newly nucleated in the faulted loops. When the stacking faults convert to perfect crystal by the motion of partial dislocation with $a/6\langle 112\rangle$ Burgers vector (a: lattice parameter), the small dislocation loops convert to stacking fault tetrahedra.[3,8] These tetrahedra formed during irradiation are also nucleated by agglomeration of vacancies.[9] The vacancy type stacking fault tetrahedra act as void nucleation sites. Especially helium atoms interact with the stacking fault tetrahedra and then stabilize it so that stacking fault tetrahedra including helium are attributed to void nucleation.[6] Therefore, when voids are formed with higher number density, void growth is retained and swelling is suppressed.

4.3 Irradiation dose, void size and void number density

It can be seen from Fig. 3 that the average void size was larger and the number density was lower, while they grew very slowly with increasing dose under electron irradiation. In the condition of helium existence the size and number density of voids increased rapidly with increasing of dose. The void size and number density under electron irradiation after helium injection were smaller and higher at the same dose, respectively than dual-beam irradiation because of different amount of injected helium at the same dose. When the irradiation dose reached to 10 dpa, the void size and the void number density was the same values due to electron irradiation after helium injection and dual-beam irradiation. This fact suggests when the void number reached to a given number density, both point defects of nterstitials and vacancies introduced by irradiation are absorbed at the voids.

4.4 Effects of helium on the irradiation induced solute concentration change near grain boundaries

From Figs. 4 and 5 it was observed that the concentration of solute elements near grain boundaries was effectively suppressed by the irradiation with helium. According to size effect on segregation,[10] the Cr, Mn which are oversize in Fe-based alloys preferentially interact with vacancies, and diffuse via vacancy diffusion mechanism.[11] Consequently, the Cr and Mn concentration near the grain boundaries are depleted. The amount of segregation depends on flux of point defects toward grain boundary. Therefore when the diffusion flux is reduced, the segregation related to the defects flow is retarded. Thus,when helium atoms are existing and trap the vacancies, therefore segregation of over sized elements are suppressed. These processes might be operative in the present Fe–Cr–Mn alloy under electron irradiation after helium injection and dual beam irradiation.

5. Conclusions

The results obtained by three kinds of irradiation modes on the irradiation damage microstructures of Fe–Cr–Mn (W, V) alloy are as follows:

(1) Helium enhanced void nucleation, and in the condition of helium existence the sink strength of voids is decreased in order of electron beam irradiation after helium injection, electrons/He^+-ions dual-beam irradiation and single-electron beam irradiation.

(2) The void swelling of Fe–Cr–Mn (W, V) alloy is enhanced by helium existence.

(3) The segregation of solute elements such as Cr, Mn near the grain boundaries was suppressed by existence of helium.

Acknowledgements

Irradiation experiments of this work were performed at the HVEM of the Research Laboratory of the Center for Advanced Research of Energy Technology, Hokkaido University, Japan.

REFERENCES

1) H. Takahashi, Y. Shindo and H. Kinoshita: J. Nucl. Mater. **258–263** (1988) 1644–1650.
2) Hu Benfu and H. Takahashi: Chinese J. Nucl. Sci. and Eng. **17** (1997) 143–149.
3) H. Takahashi, Y. Hidaka and S. Ohnuki: Interaction of point Defect with Solutes. Micron, Vol. 27, No. 3–4, (1996) 239–246.
4) W. Schilling: *Point Defects and Defect Interaction in Metals*, (University of Tokyo Press, 1982) pp. 303–308.
5) W. D. William and R. A. Jonson: *Proceedings of the International Conference on Interatomic Potentials and Simulation of Lattice Defects*, ed. by Pierre C. Gehlen, Joe R. Beeler, Jr. and Robert I. Jaffee (Plenum Press, New York, 1972) 375.
6) Y. Hidaka, S. Ohnuki, H. Takahashi and S. Watanabe: J. Nucl. Mater. **212–215** (1994) 330–335.
7) M. Kiritani, N. Yoshida and H. Takata: J. Phys. Soc. Jpn. **38** (1975) 1677.
8) K. Urban and M. Wilkens: Phys. Status Solidi (a) **6** (1971) 173–185.
9) S. Kojima, Y. Satoh, H. Taoka and M. Kiritani: Philos. Mag., **A59** (1989) 519–532.
10) P. R. Okamoto and L. E. Rehn: J. Nucl. Mater., **83** (1979) 2–7.
11) T. R. Anthony: *In Radiation-Induced Void in Metals*, Corbett. J. W. and Ianniello, L. C. (eds.) (1972) pp. 630–646.