



Effect of triple ion beams in ferritic/martensitic steel on swelling behavior

E. Wakai ^{a,*}, T. Sawai ^a, K. Furuya ^a, A. Naito ^a, T. Aruga ^a, K. Kikuchi ^a,
S. Yamashita ^b, S. Ohnuki ^b, S. Yamamoto ^c, H. Naramoto ^c, S. Jistukawa ^a

^a Department of Materials Science, Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken 319-1195, Japan

^b Hokkaido University, Kita-ku, Sapporo 060-8628, Japan

^c Japan Atomic Energy Research Institute, Takasaki, Gunma 370-1290, Japan

Abstract

The synergistic effects of displacement damage and atomic hydrogen and helium on swelling of the ferritic/martensitic steel, F82H, has been investigated. The irradiation was performed at temperatures between 470 and 600 °C to 50 dpa (displacement per atoms) under conditions of simultaneous ion beams consisting of Fe³⁺, He⁺ and H⁺ ions or Fe³⁺ and He⁺ ions. The swelling of F82H steel under triple beams with 18 appm He/dpa and 70 appm H/dpa was larger than that under dual beams with 18 appm He/dpa. The swelling in F82H under triple beams increased with decreasing irradiation temperature from 0.1% to 3.2%, while swelling under dual beams was between 0.04% and 0.08%. On the other hand, in the case of triple beam irradiation with a high ratio of gas/dpa, the swelling tended to increase with irradiation temperature. The swelling in ferritic/martensitic steels is significantly enhanced by the synergistic effect of displacement damage, hydrogen and helium atoms.

© 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

Helium and hydrogen accumulations due to transmutation and implantation in ferritic/martensitic steels have been considered as a potential cause for irradiation-induced helium-embrittlement [1–5] and -hydrogen-embrittlement [6,7] and irradiation-induced-swelling [8,9]. The swelling of F82H and other 7–9Cr low-activation ferritic steels irradiated at 430 °C to 67 dpa (displacement per atoms) in FFTF was reported by Morimura et al. [10,11]; the smallest swelling was in F82H (0.1%) compared to the other steels (0.1–0.7%). Recently, the synergistic effect of displacement damage and helium production on swelling in F82H doped with ¹⁰B has been examined [8]. The swelling of F82H steel

irradiated to 51 dpa was 0.6–1.2%, depending on helium concentration, suggesting that the swelling of ferritic/martensitic steels was enhanced by helium. The purpose of this study is to investigate the synergistic effect of displacement damage, helium and hydrogen production on swelling in F82H ferritic/martensitic steel under triple/dual ion beams.

2. Experimental procedure

The material used in this study was ferritic/martensitic steel, IEA-heat F82H (Fe–8Cr–2W–0.2V–0.04Ta–0.1C). Irradiations were performed under triple or dual ion beams in the Takasaki ion accelerators for advanced radiation application (TIARA) facility at JAERI. The specimens were irradiated with simultaneous triple or dual ions, consisting of either 10.5 MeV Fe³⁺ ions, 1.05 MeV He⁺ ions and 0.38 MeV H⁺ ions or Fe³⁺ and He⁺ ions, to 50 dpa at 1 μm depth. The Fe³⁺, He⁺ and H⁺ ion beams were generated by a Tandem accelerator,

* Corresponding author. Tel.: +81-29 282 6563; fax: +81-29 282 5922.

E-mail address: wakai@realab01.tokai.jaeri.go.jp (E. Wakai).

Single-ended accelerator and ion implanter, respectively. The damage peak of the Fe^{3+} ions was about 150 dpa and the depth was about 1.75 μm . The irradiations by helium and hydrogen atoms were controlled to implant over depth ranges from 0.84 to 1.32 μm and 0.88 to 1.39 μm , respectively. As seen in Fig. 1, these were achieved by using two aluminum foil energy degraders, based on calculations with SRIM97 code. The ratios of hydrogen and helium concentrations to dpa (appm/dpa) were adjusted to simulate fusion reactor and spallation neutron source (SNS) target conditions, about 70 appm H/dpa and 18 appm He/dpa, and about 1700 appm H/dpa and 180 appm He/dpa, respectively. In the dual ion beams, it was 18 appm He/dpa. The irradiation temperatures were 470, 510 and 600 $^{\circ}\text{C}$, and the damage rate was about 1.6×10^{-3} dpa/s.

After irradiation, the specimens were thinned using a Hitachi FB-2000A focused ion beam (FIB) with a Ga ion gun operated at 30 kV equipped with micro-pick up

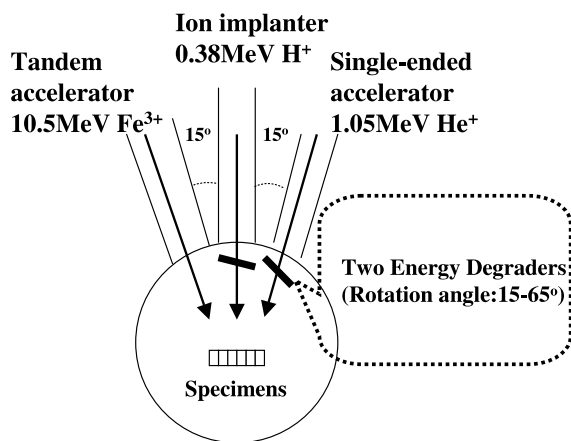


Fig. 1. Schematic configurations of three beam lines, two energy degraders and specimens.

system [12]. The microstructures were examined by transmission electron microscopy.

Following irradiation, the hydrogen depth profiles in F82H steel irradiated with triple ion beams were measured at room temperature by using the $^1\text{H}(^{15}\text{N}, \alpha\gamma)^{12}\text{C}$ nuclear resonance reaction at 6.385 MeV [13], after the triple ion irradiation. $^{15}\text{N}^{3+}$ or $^{15}\text{N}^{4+}$ ions were accelerated from 5.8 to 13.2 MeV by a Tandem accelerator. The yield from the characteristic gamma rays of the resonant nuclear reactions was measured as a function of ^{15}N ion energy with a $0.075 \text{ m} \times 0.075 \text{ m}$ NaI(Tl) detector placed just behind the samples. The beam size was about 3 mm in diameter and the beam current was about 30 nA. The absolute hydrogen concentration was determined by referencing to an amorphous Si (14 at.% H) film on a Si substrate. The details are given elsewhere [14].

3. Results

Fig. 2(a) and (b) show cavities formed in F82H steel irradiated at 470 $^{\circ}\text{C}$ with triple ion beams of Fe^{3+} ions, He^+ ions and H^+ ions under 18 appm He/dpa and 70 appm H/dpa and dual ion beams of Fe^{3+} ions and He^+ ions under 18 appm He/dpa, respectively. The size distributions for cavities that formed at depths from 0.9 to 1.1 μm in F82H steel irradiated to 50 dpa with the triple and dual ion beams are shown in Fig. 3(a) and (b), respectively. Many cavities with small sizes less than about 6 nm and with larger sizes were observed on dislocations, and the size distributions were bi-modal in triple and dual beams. The swelling of F82H irradiated with triple and dual ion beams was about 3.2% and 0.08%, respectively. The average swelling in F82H steel was significantly enhanced by the triple ion irradiation.

In Fig. 4, cavities formed at 510 $^{\circ}\text{C}$ in the F82H steels irradiated with triple and dual beams are seen. Cavities were also formed on dislocations. In the dual beams,

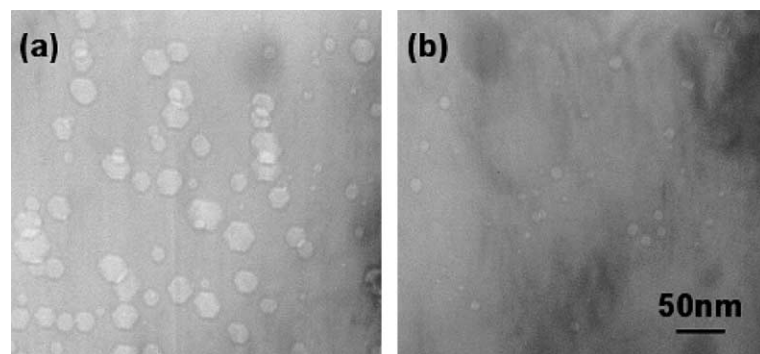


Fig. 2. Cavities formed in F82H steel irradiated at 470 $^{\circ}\text{C}$ to 50 dpa at the depth of around 1 μm under (a) triple beams of Fe^{3+} , He^+ and H^+ ions and (b) dual beams of Fe^{3+} and He^+ ions.

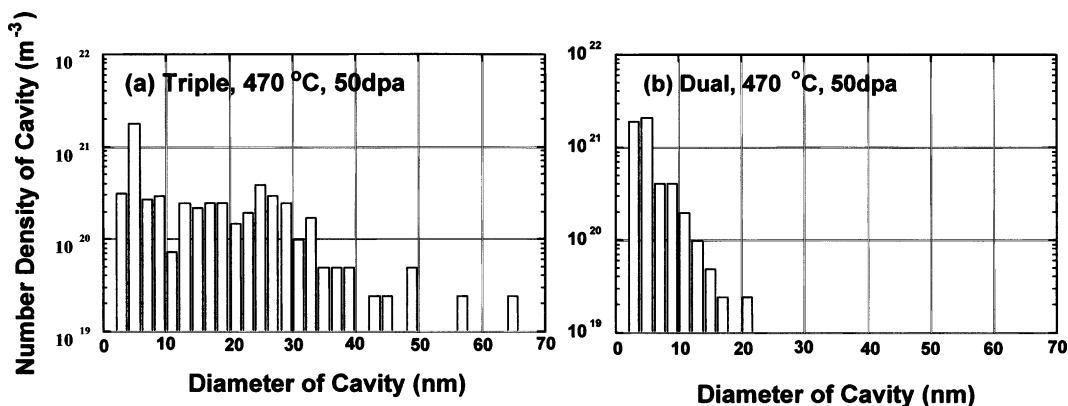


Fig. 3. Size distributions of cavities formed in F82H steel irradiated at 470 °C to 50 dpa with the triple and dual ion beams under fusion condition. It was measured at depths from 0.9 to 1.1 μm .

many small cavities with size of about 6 nm were formed in F82H steel, as shown in Fig. 3(a). In the triple beams under the fusion conditions of 18 appm He/dpa and 70 appm H/dpa, larger cavities of 20–30 nm and small cavities of about 5 nm were formed in F82H steel, as seen in Fig. 4(b). In the spallation target conditions of 180 appm He/dpa and 1700 appm H/dpa, higher density cavities with sizes from about 5 to 15 nm were formed as shown in Fig. 4(c). The average swelling in Fig. 4(a)–(c) was 0.04%, 0.15% and 0.29%, respectively. The swelling was enhanced by the spallation target condition.

In F82H steel irradiated at 600 °C to 50 dpa with the dual ion beams and triple ion beams, many cavities were formed at lath boundaries and on dislocations as given in Fig. 5. The size of cavities formed in F82H irradiated by triple ion beams at fusion conditions was similar to

that irradiated in the dual ion beams. The cavities formed under spallation target conditions were larger than those formed under fusion conditions. A summary of results for cavities formed in F82H steel irradiated from 470 to 600 °C is given in Fig. 6 and Table 1. Swelling in F82H under fusion conditions of triple ion beams increased from about 0.1% to 3.2% with decreasing irradiation temperature, while under dual ion beams it remained at low levels from about 0.04% to 0.08% at temperatures between 470 and 600 °C.

To examine the diffusion of hydrogen atoms in F82H, hydrogen depth profiles in F82H steels irradiated at 470 °C to 43 dpa with triple beams of Fe^{3+} , He^+ and H^+ ions under spallation target conditions were examined. The implanted hydrogen and helium concentrations in these specimens were about 8 and 0.4 at.%,

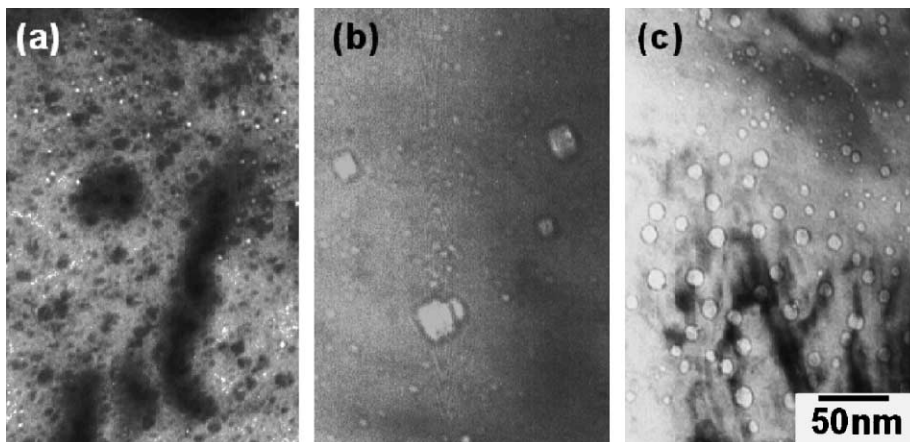


Fig. 4. Cavities (objects with white contrast) formed in F82H irradiated at 510 °C to 50 dpa under dual/triple beams. The ratios of He/dpa and H/dpa were (a) 18 appm He/dpa, 0 appm H/dpa, (b) 18 appm He/dpa, 70 appm H/dpa, and (c) 180 appm He/dpa, 1700 appm H/dpa.

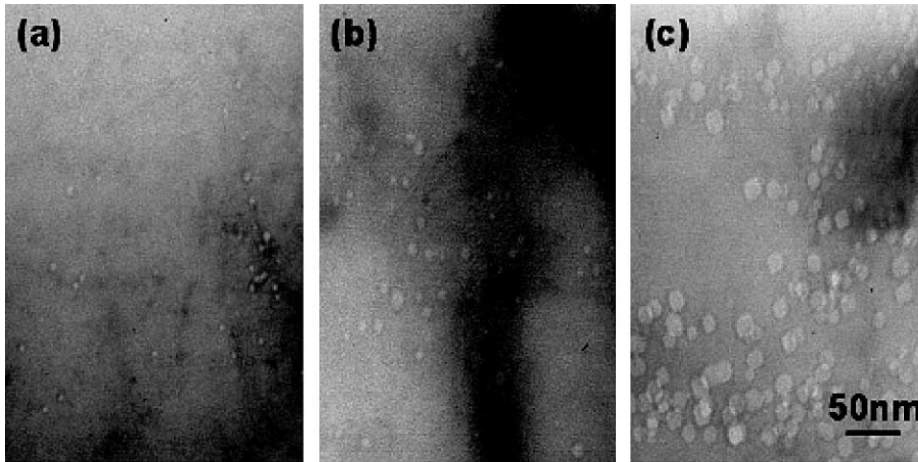


Fig. 5. Cavities (objects with white contrast) formed in F82H irradiated at 600 °C to 50 dpa under dual/triple beams. The ratios of He/dpa and H/dpa were (a) 18 appm He/dpa, 0 appm H/dpa, (b) 18 appm He/dpa, 70 appm H/dpa, and (c) 180 appm He/dpa, 1700 appm H/dpa.

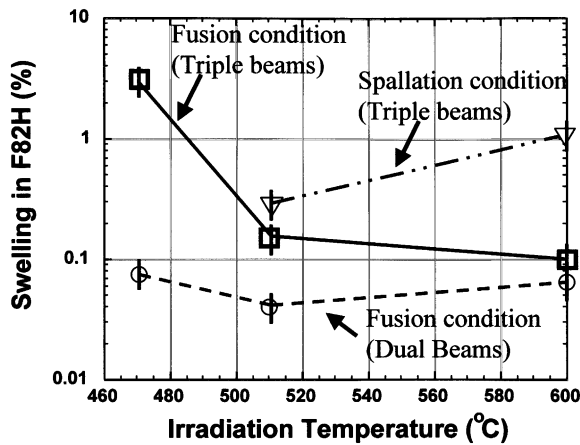


Fig. 6. Temperature dependence of the average swelling in F82H steel irradiated at 470–600 °C to 50 dpa with dual and triple ion beams.

respectively. After the irradiation, the hydrogen concentration was measured, but it was less than the detection level (~ 0.2 at.%) in the specimen.

4. Discussion

Diffusion of hydrogen atoms in iron-based alloys is very fast, and hydrogen atoms implanted at 470 °C in F82H steel under triple ion beam conditions could be detected using the $^1\text{H}(^{15}\text{N}, \alpha\gamma)^{12}\text{C}$ nuclear resonance reaction. However, the synergistic effect of displacement damage, helium and hydrogen on swelling in F82H steel occurred at temperature lower than 510 °C. According to a study on a vanadium alloy [15], where enhanced swelling by the synergistic effect of triple ion beams was reported, the cause was related to the development of dislocation loops. The synergistic effect of displacement

Table 1

A summary of results for cavities formed in F82H steel irradiated at 470–600 °C under the triple and dual beams

Irradiation temperature (°C)	Dual or triple beams	Cavity number density (m^{-3})	Cavity diameter of RMC (nm)	Swelling (%)
470	Dual	5.2×10^{21}	6.7	0.08
	Triple (fusion)	5.4×10^{21}	11.2	3.2
510	Dual	5.1×10^{21}	5.2	0.04
	Triple (fusion)	4.6×10^{21}	8.6	0.15
	Triple (spallation)	1.8×10^{22}	6.7	0.29
600	Dual	4.6×10^{21}	6.1	0.06
	Triple (fusion)	6.2×10^{21}	7.5	0.09
	Triple (spallation)	8.1×10^{21}	15.6	1.0

For fusion condition, the ratios of appm He/dpa and appm H/dpa were 18 and 70, respectively. For the spallation target condition, these were 180 appm He/dpa and 1700 appm H/dpa, respectively. RMC means the root mean cube of cavity radius.

damage and hydrogen production on microstructures in F82H steel irradiated at 250 °C to 3 dpa in HFIR, using F82H doped with ^{54}Fe (F82H(^{54}Fe)) utilizing the nuclear reaction $^{54}\text{Fe}(n, p)^{54}\text{Mn}$, has been investigated in Ref. [16]. In the study, the amount of hydrogen production was estimated to be 182 appm by Greenwood et al. [17]. In the doped F82H(^{54}Fe) specimen, cavities were formed, but were not formed in normal F82H specimen. The structure and number density of dislocation loops was different in the F82H(^{54}Fe) and normal F82H. It is suggested that the nucleation and growth of cavities and dislocation loops may be influenced by the presence of hydrogen atoms. The synergistic effect of displacement damage and helium production on swelling in F82H steel was examined [8,10,11] in HFIR and FFTF, and the swelling was 0.1–1.2%, depending on helium production, suggesting that swelling of F82H steel was enhanced by helium. In present study, the swelling in F82H steel is significantly enhanced by the synergistic effect of displacement damage, helium and hydrogen atoms, and the presence of hydrogen atoms is very important for swelling behavior under triple ion beams. The details of the swelling mechanism will be reported in another study [18].

The sink balance for point defects between dislocations and cavities is very important for swelling behavior in the theory of cavity growth [19–21]. The ferritic/martensitic steel F82H has a high density dislocation before irradiation, and the introduction of additional dislocations in F82H steel is useful for the reduction of swelling. The relation between the largest cavity size and dislocation density in F82H steel irradiated with dual ion beams at 510 °C to 50 dpa has been reported [22]. The dislocation density was changed by adjusting the tempering time and temperature and cold-working. The result shows that cavity size decreases with increasing dislocation density. The method may be useful for exploring the reduction of swelling under triple beams, and will be the subject of further study [18].

5. Conclusions

(1) The swelling of F82H steel under triple ion beams was clearly larger than that under dual beams at temperatures lower than 510 °C, and the swelling was strongly influenced by the synergistic effect of displacement damage, helium and hydrogen.

(2) The swelling in F82H under fusion conditions of triple ion beams increased with decreasing irradiation temperature from about 0.1% to 3.2%, while the values under dual ion beams were between 0.04% and 0.08%.

(3) In triple ion irradiation under spallation target conditions with a high ratio of gas/dpa, the swelling tended to increase with irradiation temperature.

(4) The implanted hydrogen concentration could not be detected in the F82H steels implanted to 8 at.% hydrogen at 470 °C.

Acknowledgements

The authors are grateful to members of JAERI TI-ARA facility for the operation of the accelerators during this work, and also thank to Drs H. Tanigawa and M. Ando of JAERI for helpful discussions.

References

- [1] B. Van der Schaaf et al., *J. Nucl. Mater.* 283–287 (2000) 52.
- [2] R.L. Klueh, M.A. Sokolov, K. Shiba, Y. Miwa, J.P. Robertson, *J. Nucl. Mater.* 283–287 (2000) 478.
- [3] K. Shiba, A. Hishinuma, *J. Nucl. Mater.* 283–287 (2000) 474.
- [4] N. Yamamoto, J. Nagakawa, K. Shiba, *J. Nucl. Mater.* 283–287 (2000) 400.
- [5] E.I. Materna-Morris, M. Rieth, K. Ehrlich, *STP 1366* (2000) 597.
- [6] Y. Dai, S.A. Maloy, G.S. Bauer, W.F. Sommer, *J. Nucl. Mater.* 283–287 (2000) 513.
- [7] N. Baluc, R. Schaublin, C. Bailat, F. Paschoud, M. Victoria, *J. Nucl. Mater.* 283–287 (2000) 731.
- [8] E. Wakai et al., *J. Nucl. Mater.* 283–287 (2000) 799.
- [9] A. Kimura, M. Narui, H. Kayano, *J. Nucl. Mater.* 191–194 (1992) 879.
- [10] T. Morimura, A. Kimura, H. Matsui, *J. Nucl. Mater.* 239 (1996) 118.
- [11] A. Kimura, H. Matsui, *J. Nucl. Mater.* 212–215 (1994) 701.
- [12] H. Tanigawa et al., *J. Nucl. Mater.* 297 (3) (2001) 279.
- [13] W.A. Lanford, *Nucl. Instrum. and Meth. B* 66 (1992) 65.
- [14] P. Goppelt-Langer, S. Yamamoto, Y. Aoki, H. Takeshita, H. Naramoto, *Nucl. Instrum. and Meth. B* 118 (1996) 7.
- [15] N. Sekimura et al., *J. Nucl. Mater.* 283–287 (2000) 224.
- [16] E. Wakai, Y. Miwa, N. Hashimoto, J.P. Robertson, R.L. Klueh, S. Jistukawa, these Proceedings.
- [17] L.R. Greenwood et al., *J. Nucl. Mater.* 283–287 (2000) 1438.
- [18] E. Wakai et al., *J. Nucl. Mater.*, to be presented in Fifth International Workshop of Spallation Material Technologies, Charleston, S.C., USA, 2002.
- [19] L.K. Mansur, W.A. Coghlan, *J. Nucl. Mater.* 119 (1983) 1.
- [20] N.H. Packan, K. Farrel, *J. Nucl. Mater.* 85–86 (1979) 677.
- [21] L.L. Horton, L.K. Mansur, in: F.A. Garner, J.S. Perrin (Eds.), *Effects of Irradiation on Materials: 12th International Symposium, ASTM STP 870*, American Society for Testing and Materials, Philadelphia, 1985, p. 344.
- [22] T. Sawai, E. Wakai, K. Tomita, A. Naito, S. Jistukawa, these Proceedings.