Effect of Helium and Hydrogen Production on Irradiation Hardening of F82H Steel Irradiated by Ion Beams

Eiichi Wakai¹, Masami Ando¹, Tomotsugu Sawai¹ and Somei Ohnuki²

¹Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency, Tokai-mura, Ibaraki 319-1195, Japan

²Materials Science Division, Hokkaido University, Sapporo 060-8628, Japan

Effects of helium and hydrogen production on irradiation hardening of martensitic steel F82H (Fe-8Cr-2W-0.2V-0.04Ta-0.1C) were examined by dual or triple beam experiments. The effects of tempering and cold working were also examined. The irradiations were performed at about 500°C to 50 dpa under simultaneous dual beams of 10.5 MeV Fe³⁺ and 1.05 MeV He⁺ or triple beams of those and 380 keV H⁺ ions. The value of appm-He/dpa for the dual ion beams was about 15, and the values of appm-He/dpa and appm-H/dpa for the triple ion beams were 15 and 15 (or 150), respectively. The hardness of the irradiated specimens measured at room temperature using a micro indentation after the irradiations. Irradiation softening and hardening was observed in F82H-std, F82H+20%CW and a non-tempered F82H steels irradiated at about 500°C to 18 and 50 dpa, respectively, by dual ion beams. The hardness of the specimens irradiated at about 500°C to 18 dpa under triple ion beams was harder than that under dual ion beams. [doi:10.2320/matertrans.MBW200611]

(Received October 18, 2006; Accepted March 15, 2007; Published May 25, 2007)

Keywords: irradiation hardening, irradiation softening, martensitic steel, triple ion beams, dual ion beams, micro indentation, microstructure

1. Introduction

Low-activation ferritic/martensitic steels are candidate materials for the first wall and blanket structure of fusion reactors. The high-energy neutrons produced in the D-T fusion reaction induce displacement damage and generate gas atoms from (n, p) and (n, α) reactions in the materials. The presence of these gases may lead to degradation of mechanical properties. The changes of mechanical properties are intimately related with the microstructural evolutions such as the formation and growth of defect clusters, which is easily affected by the existence of helium atoms. The effects of helium production as a function of displacement damage and heat treatment on swelling behavior of F82H steel by neutron irradiation at 300 and 400°C were reported,¹⁾ and the synergistic effects of displacement damage helium and hydrogen on swelling behavior at 470–600°C^{2–5)} and irradiation hardening⁶⁾ at 270°C and 360°C of F82H steel by ion irradiation were also reported. These results showed that the production of gaseous atoms enhanced the swelling and hardness of the F82H steel, and the non-tempered treatment for the F82H steel remarkably suppressed the swelling.

The purpose of this study is to investigate the effects of ratios of hydrogen and helium to displacement damage and of initial microstructures due to cold working and tempering on hardness of F82H steel irradiated at about 500°C by dual or triple beam conditions.

2. Experimental Procedure

The chemical compositions of the F82H-std steel (IEA-heat, rolling No. KG820-2, sampling position 42W-16, grain size number 3.5) used in this study are given in Table 1. The heat treatment for F82H steel was firstly normalized at 1040°C for 38 minutes in a vacuum followed by air-cooling. After that the specimens were tempered at 750°C for 1 hour in a vacuum followed by air-cooling.

In this study, four types of F82H specimens were prepared as shown in Table 2, and they were denoted as F82H-std, F82H (as-nor.), F82H (10 min-tempered) and F82H+

20%CW. The F82H-std was normalized at 1040°C for 0.5 h and tempered at 750°C for 1 h (a second N&T heat treatments) after the first N&T heat treatments. The F82H (as-nor.) was normalized at 1040°C for 0.5 h after the first N&T treatments. The F82H (10 min-tempered) was normalized at 1040°C for 0.5 h and tempered at 750°C for 10 minutes after the first N&T treatment. The F82H+20%CW was cold-rolled up to 20% after the first N&T treatments. The microstructures of the specimens were observed using a transmission electron microscope of Hitachi HF2000 before irradiation experiments.

Ion irradiations were performed at about 500°C under a simultaneous dual ion irradiation with 10.5 MeV Fe³⁺ ions and 1.05 MeV He⁺ ions or a simultaneous triple ion irradiation with the Fe ions, the He ions and 380 keV H⁺ ions in TIARA facility of JAEA. The ion irradiations were performed to about 2.3, 18 and 50 dpa. The ratios of He (appm)/dpa and H (appm)/dpa at the depth of 1.75 μ m are 15 and 15 (or 150) for dual and triple beam irradiation. The displacement damage and ion concentrations were calculated by using SRIM2003 code, and the major component of displacement damage is due to the contribution of elastic collision of Fe ions. During the ion irradiations, the temperature of the surface in all specimens was continuously measured with Nikon's thermal vision system. After irradiation, the indentation testing was performed on these specimens, using a Berkovich diamond indenter at a maximum load to 25 mN, on an ultra micro-indentation, UMIS-2000. Micro-hardness profiles were obtained by a contact pressure evaluation method based on the procedure proposed by Oliver *et al.*⁷⁾ 30 data points were measured for each specimens and the average was calculated. The detail of the micro-indentation procedure have been explained elsewhere.⁸⁾

3. Results

3.1 Microstructures and hardness before irradiation

Figs. 1(a)–1(d) show the microstructures of F82H (as-nor.), F82H (10 min-tempered), F82H-std, and F82H+

Table 1 Chemical compositions of the F82H steel (IEA heat) used in this study.

Alloys	Cr (mass%)	C	N	P	S	Al	Si	V	Ti	Mn	Ta	W
F82H	7.84	0.09	0.007	0.003	0.001	0.001	0.07	0.19	0.004	0.1	0.04	1.98

Table 2 Four kinds of specimens used in this study.

Specimens	Normalizing	Tempering	Cold work
F82H-std	1040°C, 40 min.	750°C, 60 min.	—
F82H (as-nor.)	1040°C, 40 min.	—	—
F82H (10 min-Tempered)	1040°C, 40 min.	750°C, 10 min	—
F82H+20%CW	1040°C, 40 min.	750°C, 60 min.	20%

Fig. 1 Microstructures of (a) F82H (as-nor.), (b) F82H (10 min-tempered), (c) F82H-std, and (d) F82H-std+20%CW steels taken by a transmission electron microscope before irradiation.

20%CW specimens, respectively. Lath structure and a high density dislocation were observed in the F82H (as-nor.) specimen, and no formation of carbides were seen as given in Fig. 1(a). In the F82H (10 min-tempered) and F82H-std specimen, the densities of dislocations were lower than that of F82H (as-nor.), and the carbides were observed. The density of dislocation in the F82H+20%CW steel was slightly higher than that of F82H-std with a relatively high number density of dislocations about $(2-5) \times 10^{14} \text{ m}^{-2}$. Figure 2 shows micro-hardness of the specimens measured at room temperature using an ultra micro-indentation. The hardness of F82H (as-nor.) steel was about twice of F82H-std. The hardness of F82H+20%CW was slightly higher than that of F82H-std.

3.2 Hardness after ion irradiation

Figure 3 shows the micro-hardness of F82H-std irradiated at about 500°C to 50 dpa by dual beams of Fe^{3+} and He^+ ions

Fig. 2 Micro-hardness of F82H-std, F82H (10 min-tempering), F82H (as-nor.) and F82H-std+20%CW steels measured by a micro-indentation before irradiation.

as a function of displacement damage. The micro-hardness of F82H-std before irradiation was about 3.4 GPa at room temperature. A small of irradiation-hardening was observed in the specimen irradiated at about 500°C to 2.3 dpa, and the micro-hardness decreased somewhat at 18 dpa and then it

Fig. 3 Dependence of micro-hardness on displacement damage in the F82H-std steel irradiated at 500°C to 50 dpa under dual ion beams of Fe^{3+} and He^+ ions.

Fig. 4 Micro-hardness of F82H (as-nor.), F82H-std and F82H-std+20%CW steels irradiated at 500°C under dual ion beams of Fe^{3+} and He^+ ions.

was changed to increase at 50 dpa. The micro-hardness of F82H-std, F82H+20%CW and F82H (as-nor.) specimens irradiated by the dual ion beams at about 500°C to 50 dpa is given in Fig. 4, and the irradiation softening and hardening were observed at 18 and 50 dpa, respectively, in these specimens. The largest irradiation softening occurred in the specimen with highest dislocation density.

The effect of hydrogen concentration to displacement damage in the F82H-std and F82H+20%CW steels was examined by dual and triple ion irradiations as shown in Fig. 5. In both specimens, the increase of micro-hardness due to irradiation in triple beam condition was larger than that in dual beam condition. The co-implantation of hydrogen and helium atoms enhanced the micro-hardness, but the large increment of hydrogen concentration for the co-implantation on the micro-hardness was relatively small.

4. Discussion

As shown in Figs. 3 and 4, the micro-hardness irradiated at 500°C to 50 dpa by dual ion beams depended on displacement damage, *i.e.*, small increment of micro-hardness occurred at the initial damage stage of 2.3 dpa, however after that, the micro-hardness was changed to decrease between 2.3 and 18 dpa and then it increased between 18 and 50 dpa. The change of micro-hardness during irradiation would be related to microstructural evolution of defect clusters such as dislocations, dislocation loops and cavities. It is well known that the formation of dislocation network and disappearing dislocations during irradiation decreases the hardness. As shown in a reference of irradiation softening in a

Fig. 5 Effect of triple and dual ion beams on micro-hardness of F82H-std and F82H-std+20%CW steels irradiated at 500°C to 18 dpa.

Fig. 6 Explanation of irradiation softening and hardening of F82H steel during dual beam irradiations. The process A is mainly due to the process of disappearing dislocations, and the process B is mainly due the process of the formation and growth of cavities.

9Cr-2W martensitic steel,⁹⁾ the reduction of strength was induced by neutron irradiation of FFTF/MOTA up to 36–61 dpa at 410°C. F82H steel has similar microstructures with the 9Cr-2W martensitic steel, and the irradiation softening of F82H steel would be easily expected during irradiation at over 400°C. However, the change from softening to hardening by irradiation has been reported. In order to explain this process from irradiation softening to irradiation hardening, we have to think the other factors except for the disappearing process of dislocations. It should here point out a difference of helium production amount during irradiation between present experiment data and the other experimental ones. The production amount of helium atoms in the reference⁹⁾ is a few appm-He level and that of present study is about 750 appm-He at 50 dpa. According to our previous data of swelling behavior,¹⁾ it was found that the increase of helium atom production enhanced the nucleation of cavities in F82H steel. Accordingly, the increment of number density of cavities due to helium implantation can be expected in present study. It is very known that defect clusters such as bubbles and voids have barrier strength for deformation,¹⁰⁾ and therefore the formation and growth of cavities could contribute on irradiation hardening, and the micro-hardness could increase somewhat with increasing the size and number density of cavities. In Fig. 6, the schematic explanation of this process is given, and there are two processes of dislocation disappearing and cavity formation and growth. The process A is mainly due to the process of disappearing dislocations,

and the process B is mainly due the process of the formation and growth of cavities.

As shown in Fig. 4, the enhancement of irradiation hardening due to co-implantation of hydrogen and helium atoms was found. In previous studies^{2,3)} of swelling behaviors by triple and dual ion beams to 50 dpa, the dislocation densities were similar to each other, and the sizes and number densities of cavities depended on the ratios of He/dpa and H/dpa for ion irradiation conditions. Hydrogen atoms enhanced the growth of cavities during triple ion irradiations. Therefore, the enhancement of micro-hardness by the triple ion irradiation in present study might be explained by the formation of larger cavities. The similar effects of enhancement of irradiation hardening¹¹⁾ and swelling^{12,13)} due to triple ion beams in the other martensitic steel,¹⁰⁾ vanadium and vanadium alloys¹²⁾ and SiC/SiC composites¹³⁾ were reported.

5. Conclusions

Synergistic effects of helium and hydrogen implantations on irradiation hardening of F82H irradiated by dual or triple beam condition were investigated by micro hardness measurements. The irradiation was performed at 500°C to 18 or 50 dpa under simultaneous dual beams of 10.5 MeV Fe³⁺ and 1.05 MeV He⁺ or the Fe, the He and 380 keV H⁺ ions. The ratios of He (appm)/dpa and H (appm)/dpa were 15 and 15 (or 150) for dual and triple ion beams. The results obtained as described below;

- (1) Irradiation softening and hardening under dual beams was observed in F82H steel irradiated at 500°C to 18 and 50 dpa, respectively.
- (2) The micro-hardness in F82H-std, F82H+20%CW and F82H (as-nor.) steels irradiated at 500°C to 18 dpa under triple beam irradiations was larger than that under dual beam irradiation.

- (3) The cause of the irradiation softening and hardening may be related with microstructural evolutions due to the processes of disappearing dislocations and the formation and growth of cavities during irradiation.

Acknowledgement

The authors would like to thank members of TIARA facility in JAEA for utilization of ion beam experiment.

REFERENCES

- 1) E. Wakai, N. Hashimoto, J. P. Robertson, R. L. Klueh, K. Shiba and S. Jitsukawa: *J. Nucl. Mater.* **283–287** (2000) 799–805.
- 2) E. Wakai, T. Sawai, K. Furuya, A. Naito, T. Aruga, K. Kikuchi, S. Yamashita, S. Ohnuki, S. Yamamoto and S. Jitsukawa: *J. Nucl. Mater.* **307–311** (2002) 278–282.
- 3) E. Wakai, K. Kikuchi, S. Yamamoto, T. Aruga, M. Ando, H. Tanigawa, T. Taguchi, K. Oka and S. Ohnuki: *J. Nucl. Mater.* **318** (2003) 267–273.
- 4) E. Wakai, T. Sawai, A. Naito and S. Jitsukawa: *J. Electron Microscopy* **51** (2002) S239–S243.
- 5) T. Sawai, E. Wakai, T. Tomita, A. Naito and S. Jitsukawa: *J. Nucl. Mater.* **307–311** (2002) 312–316.
- 6) M. Ando, E. Wakai, T. Sawai, H. Tanigawa, K. Furuya, S. Jitsukawa, H. Takeuchi, K. Oka, S. Ohnuki and A. Kohyama: *J. Nucl. Mater.* **329–333** (2004) 1137–1141.
- 7) W. C. Oliver and G. M. Pharr: *J. Mater. Res.* **7** (1992) 1564.
- 8) M. Ando, H. Tanigawa, S. Jitsukawa, T. Sawai, Y. Katoh, A. Kohyama, K. Nakamura and H. Takeuchi: *J. Nucl. Mater.* **307–311** (2002) 260–265.
- 9) A. Kohyama, Y. Kohno, K. Asakura and H. Kayano: *J. Nucl. Mater.* **212–215** (1994) 684–689.
- 10) G. E. Lucas: *J. Nucl. Mater.* **206** (1993) 287–305.
- 11) E. H. Lee, J. D. Hunn, G. R. Rao, R. L. Klueh and L. K. Mansur: *J. Nucl. Mater.* **271&272** (1999) 385–390.
- 12) N. Sekimura, T. Iwai, Y. Arai, S. Yonamine, A. Naito, Y. Miwa and S. Hamada: *J. Nucl. Mater.* **283–287** (2000) 224–228.
- 13) T. Taguchi, N. Igawa, S. Miwa, E. Wakai, S. Jitsukawa, L. L. Snead and A. Hasegawa: *J. Nucl. Mater.* **335** (2004) 508–514.