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Swelling behavior of F82H steel irradiated by triple/dual ion beams

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Abstract

Irradiations for spallation target vessels and structural materials of fusion reactors were simulated using simultaneous triple/dual ion beams consisting of Fe^{3+}, He^{+} and H^{+} ions or Fe^{3+} and He^{+} ions at temperatures between 470 and 600 °C to 50 dpa. The swelling of F82H (Fe–8Cr–2W–0.2V–0.04Ta–0.1C) martensitic steel was enhanced by a synergistic effect of displacement damage and the implantation of helium and hydrogen. The maximum swelling of F82H steel was 3.2% at 470 °C under a simulation of structural materials of fusion reactors, and was higher than 1.2%, which applied to a simulation of spallation target vessel. The swelling under a simulation of fusion reactor decreased with increasing irradiation temperature, however the swelling under a simulation of spallation target vessel was again increased at 600 °C by the high helium concentration. From the microstructural analysis of taking account of cavity growth process, the cause of the enhancement of swelling under a simulation of fusion reactor is thought to be gas pressure of hydrogen and helium in cavities during irradiation. The effects of 50% cold-working and carbon implantation on swelling behavior were also examined. The swelling was reduced from 3.2% to 1.4% by 50% cold-working, and to 0.5% by carbon implantation.

1. Introduction

Helium and hydrogen accumulations due to transmutation and/or implantation in systems of ferritic/martensitic steels have been considered as a potential cause for irradiation-induced helium embrittlement [1–5], 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 in fast flux test facility (FFTF) was reported [10,11]; the smallest swelling of 0.12% was found in F82H (Fe–8Cr–2W–0.2V–0.04Ta–0.1C in mass%), compared to the other steels (0.21–0.74%). The synergistic effect of displacement damage and helium production on swelling in F82H doped with ^{10}B was examined [8]. The swelling of F82H irradiated to 51 dpa was 0.6–1.2%, depending on helium concentration, suggesting that swelling of ferritic/martensitic steels may be enhanced by helium atoms. Swelling behavior of F82H irradiated by triple/dual beams under simulation irradiations of fusion reactors has reported [12]. The temperature of beam window depends on the system design of Pb–Bi flow and the power of proton beams. There is a possibiltiy that a beam window in target vessel will be used at temperature higher than 500 °C. In case of fusion system, the temperature estimated from a heat transfer calculation in blanket structure using F82H steel will be between about 300 and 580 °C. The purpose of this study is firstly to

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compare the swelling behavior of the spallation target vessels and the structural materials of fusion reactors under the simulation ion irradiation and secondly to study behaviors of the mechanism of swelling in F82H steel using triple/dual ion beams.

2. Experimental procedure

The material used in this study was ferritic/martensitic steel, IEA-heat F82H steel (Fe–8Cr–2W–0.2V–0.04Ta–0.1C in mass%). Irradiations were performed with triple or dual ion beams in the Takasaki ion accelerators for advanced radiation application (TIARA) facility of JAERI. The specimens were irradiated with simultaneous triple ion beams of 10.5 MeV Fe^{3+} ions, 1.05 MeV He^{+} ions and 0.38 MeV H^{+} ions or dual ion beams of Fe^{3+} and He^{+} ions to 50 dpa (displacement per atoms) at 1 μm depth. The Fe^{3+}, He^{+} and H^{+} ion beams were generated by a Tandem accelerator, single-ended accelerator and ion implanter, respectively. As shown in Fig. 1, the damage peak of the Fe^{3+} ions was about 1.75 μm, and the irradiation by helium and hydrogen atoms was controlled to implant over a depth range from about 0.85 to 1.30 μm. This was achieved by using two aluminum foil energy, based on calculations with SRIM2000 code [13–15]. The ratios of hydrogen and helium concentrations to dpa (appm/dpa) for simulation irradiations of structural materials of fusion reactors or spallation neutron source (SNS) target vessel were about 70 appmH/dpa and 18 appmHe/dpa, and about 1700 appmH/dpa and 180 appmHe/dpa, respectively. In the dual beams, it was 18 appmHe/dpa. In order to examine the influence of helium and hydrogen atoms for swelling behavior at high temperature, the irradiations were performed at 470, 510 and 600 °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 and equipped with micropick up system. the microstructures were examined by transmission electron microscopy.

After the triple ion irradiation, the hydrogen depth profiles in F82H steel irradiated with triple ion beams were measured at room temperature by using the $^{1}H(^{15}N, \alpha\gamma)^{12}C$ nuclear resonance reaction occurred at 6.385 MeV [16], after the triple ion irradiation. $^{15}N^{3+}$ or $^{15}N^{4+}$ ions were accelerated from 5.8 to 13.2 MeV by a Tandem accelerator as shown in Fig. 2. The yield from the characteristic γ-rays of the resonant nuclear reactions was measured as a function of ^{15}N ion energy with a 0.075 m × 0.075 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

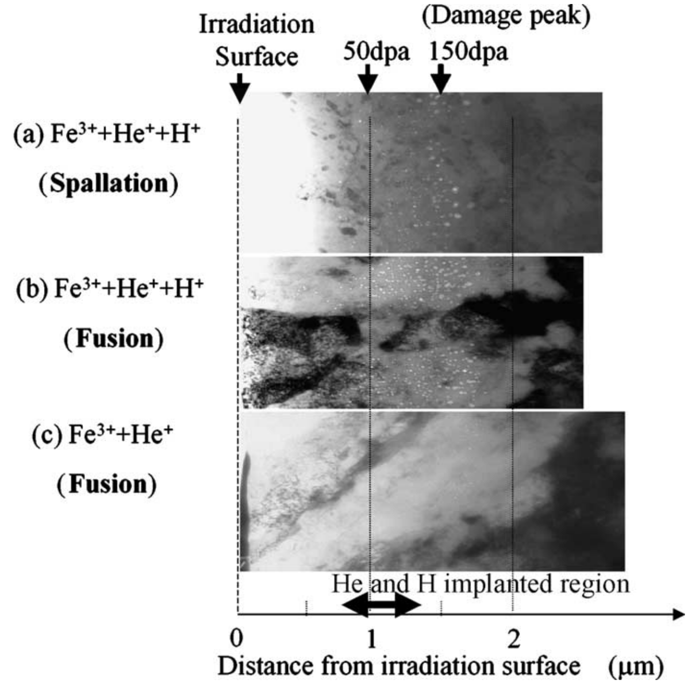

Fig. 1. Bright-field images, of F82H steels irradiated at 470 °C under simulation irradiations of spallation target vessel by triple ion beams and structural materials of fusion reactor by triple ion beams and dual ion beams, respectively, taken by a transmission electron microscope. The micrographs are cross-section images from the irradiation surface to about 2.5 μm in depth.

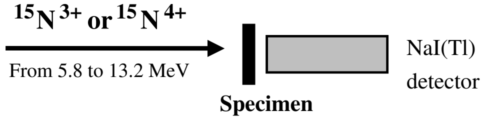

Fig. 2. Schematic configuration of hydrogen depth profile analysis using $^{1}H(^{15}N, \alpha\gamma)^{12}C$ resonance nuclear reaction after triple ion beams of a simulation irradiation of spallation target vessel. The resonance occurred at 6.385 MeV.

hydrogen concentration was determined by referencing to an amorphous Si (14 at.% H) film on a Si substrate. The details are given in elsewhere [17].

3. Results

3.1. Microstructures of F82H steel irradiated by triple/dual beams

Fig. 3(a)–(c) show the bright-field images of F82H steels irradiated at 470 °C under a simulation of spallation target vessel by triple ion beams and under simulations of structural material of fusion reactors by triple ion beams and dual ion beams, respectively. The micrographs are cross-section images from the irradiation surface to about 2.5 μm in depth. Cavities are observed in F82H steel irradiated in these irradiation conditions, and larger size cavities are formed at around the damage peak in these specimens. The gas atoms implanted area was magnified in Fig. 3, where the depth is about 1.0 μm and the displacement damage is 50 dpa. The largest cavities in these micrographs were observed in the simulation irradiation of fusion reactors under triple beams. Many cavities with small sizes less than 6 nm and with large sizes were seen in matrix and on dislocations, and the size distributions were bi-modal. The size of cavity formed by the triple beams of fusion condition was remarkably larger than that of dual beams of fusion-like condition. The number density of cavities in the simulation of spallation target vessel was higher than the others. The shape of cavity with smaller size than about 6 nm was sphere in three irradiation conditions. The shapes of larger size cavity formed in the simulation irradiations were close to spherical shape but had facets, respectively. The swellings in Fig. 3(a)–(c) is 1.2%, 3.2% and 0.1%, respectively. The swelling in F82H irradiated with triple ion was the most among the irradiation conditions. The number density and root mean diameter of cavities formed in F82H steel irradiated in three beam conditions was summarized in Fig. 4. The dependence of irradiation temperature on swelling in these conditions is shown as in Fig. 5, and the swelling in the simulation irradiation of fusion reactors decreased with increasing irradiation temperature. The dependence of swelling on irradiation temperature under the simulation of spallation target vessel was a different from the swelling behavior of simulation of fusion reactor, and it increased at 600 °C. The increase of swelling may be due to the increment of gas pressure caused by high helium concentration in cavities. In Fig. 6, dislocation features are shown for the F82H steels irradiated at 470 and 600 °C to 50 dpa in these irradiation conditions. The density of dislocations is given in Fig. 7. The densities of dislocations decreased with the irradiation temperature and were nearly equal at each temperature.

3.2. Hydrogen distribution in F82H steel irradiated by triple ion beams

In order to examine the diffusion of hydrogen atoms in F82H steel irradiated at 80 °C to 30 dpa and 470 °C to 43 dpa in the simulation of spallation target vessel, the depth profiles of hydrogen concentration were measured by the $^{1}H(^{15}N, \alpha\gamma)^{12}C$ resonance nuclear reaction

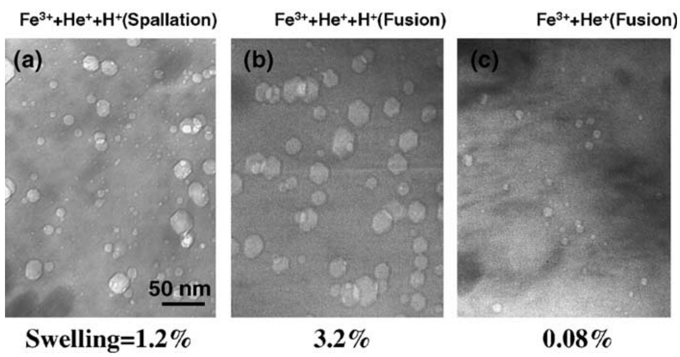

Fig. 3. Cavities formed in F82H steel irradiated at 470 °C to 50 dpa and the area is at the depth of around 1 μm. (a) A simulation irradiation of spallation target vessel (triple beams), (b) a simulation irradiation of structural materials of fusion reactor (triple beams) and (c) a simulation irradiation of fusion reactor (dual beams).

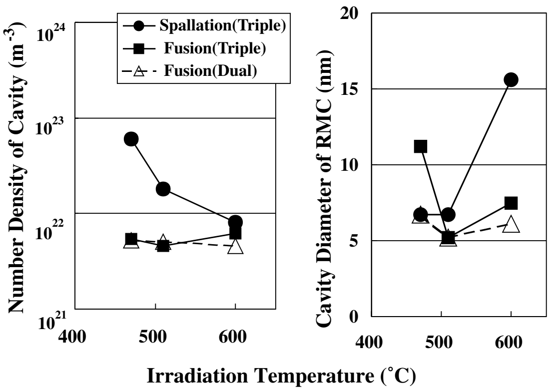

Fig. 4. The number density and diameter of root mean cube of cavities formed in F82H steel irradiated by three beam conditions.

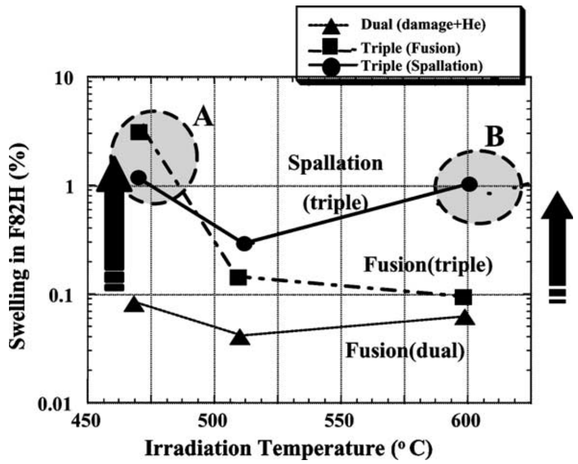

Fig. 5. The dependence of swelling of F82H steels on irradiation temperature under these conditions. In the area A the synergistic effect of displacement damage, helium and hydrogen occurred, and in the area B the enhancement of swelling was caused by the synergistic effect of displacement damage and high concentration helium.

technique after the triple ion irradiation, a few weeks later. The depth profiles of hydrogen atoms in F82H steels are shown in Fig. 8. The implanted hydrogen concentrations in the specimens irradiated at 80 and 470 °C calculated by SRIM were estimated as about 6 and 8 at.%, respectively, and the implanted helium concentrations were also calculated as 0.4 and 0.8 at.%, respectively. In this experiment, the hydrogen concentration measured was about 0.5 at.% in the F82H steel irradiated at 80 °C in a range from about 0.9 to 1.3 μm, and many hydrogen atoms was likely diffused out in the region. In the specimens irradiated at 470 °C, the hydrogen concentration could not be detected. In this experiment, the background level was about 0.13 at.% H.

4. Discussion

The main results in this study were two contents as follows: (a) the enhancement of swelling under triple ion beams at 470 °C in simulation irradiation of structural materials of fusion reactors and the increase of swelling under a simulation irradiation of spallation target vessel at 600 °C and (b) the method of swelling suppression of cold-working and carbon implantation. In the following sentence, the contents (a) and (b) are discussed.

4.1. Enhancement of swelling under triple ion beams

It is known that diffusion of hydrogen atoms in iron-based alloys is fast at 400 °C. It is easy to understand that hydrogen atoms implanted at 470 °C in F82H steel under triple ion beam conditions cannot be detected by the $^{1}H(^{15}N, \alpha\gamma)^{12}C$ nuclear resonance reaction. However, a synergistic effect of displacement damage, helium and hydrogen on swelling in F82H steel occurred remarkably at 470 °C in fusion condition. A similar result was reported in the study of pure vanadium and vanadium alloys [18]. Triple beam irradiation with He, H and Ni strongly enhanced the growth of cavities and the swelling in pure vanadium compared with those under dual beam irradiation with He and Ni or single ion beam irradiation with Ni, whereas simultaneous implantation of He without H did not affect on cavity growth and swelling. The cavities were stabilized by both helium and hydrogen, and excess interstitials were considered to enhance dislocation microstructure evolution resulting in excess vacancies for cavity growth. In V–5Cr–5Ti alloy, no cavities were detected without implantation of He. However, Ni, He and H triple beam irradiation was found to enhance swelling. The injection of helium induced the nucleation of fine cavities. Cavity nucleation and growth were further amplified when hydrogen was injected together.

In a similar study of F82H steel for a synergistic effect of displacement damage and hydrogen production on microstructures of ferritic/martensitic steel, the microstructure was examined in F82H steel irradiated at 250 °C to 3 dpa in HFIR, using F82H doped with ^{54}Fe (F82H(^{54}Fe)) utilizing a nuclear reaction of $^{54}Fe(n, p)^{54}Mn$ [19]. In the F82H(^{54}Fe) specimen, the amount of hydrogen production was measured to be 182 appm [20,21]. Dislocation loops formed in the normal F82H were observed on $\{1\,1\,1\}$ planes with $(\mathbf{a}/2)\langle 1\,1\,1\rangle$ Burgers vectors and some loops were arranged along dislocation lines. The number density and mean size of the loops were $1.4 \times 10^{22}\ m^{-3}$ and 7.9 nm, respectively. While, two types of dislocation loops formed in the F82H(^{54}Fe)

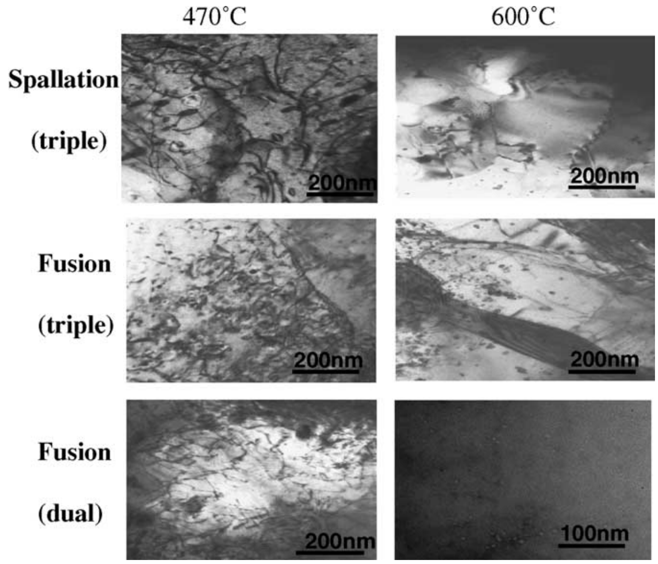

Fig. 6. Dislocation features are shown for the F82H steel irradiated at 470 and 600 °C to 50 dpa under these irradiation conditions.

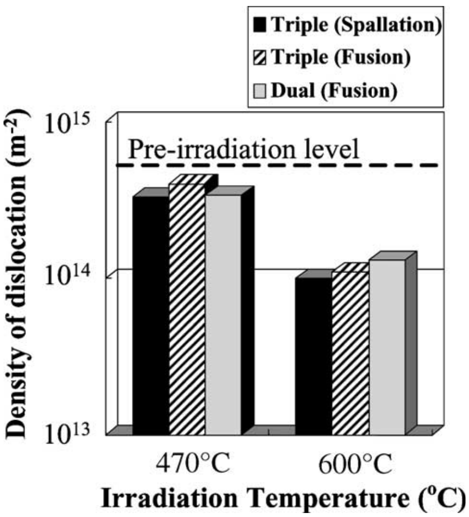

Fig. 7. The densities of dislocations and cavities decreased with the irradiation temperature and the densities of dislocations under those conditions were nearly equal at each temperature.

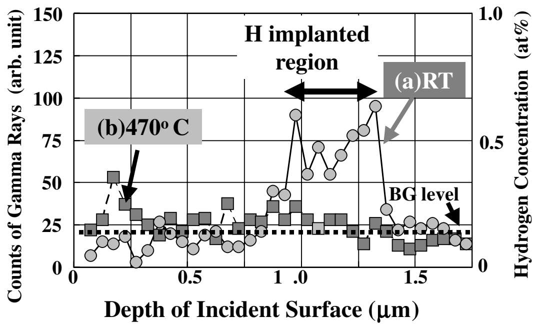

Fig. 8. Depth profiles in F82H steels irradiated at 80 °C to 30 dpa and 470 °C to 43 dpa by a simultaneous triple beams of Fe^{3+}, He^{+} and H^{+} ions in a simulation irradiation of spallation target vessel were measured by gamma ray counter. The conditions of triple ion beams are (a) RT, 6 at.% H, 0.8 at.% He, 30 dpa and (b) 470 °C, 8 at.% H, 0.4 at.% He, 43 dpa.

were observed: those on $\{111\}$ planes with $(\mathbf{a}/2)\langle 111\rangle$ Burgers vectors and those on $\{100\}$ planes with $\mathbf{a}\langle 100\rangle$ ones. The concentration of $\langle 111\rangle$ type to all loops was about 73%. The total number densities and mean size of dislocation loops were 2.1×10^{22} m^{-3} and 6.6 nm, respectively. Small cavities were observed in F82H(^{54}Fe), and the number density of cavity, root mean cube of cavity radius, and swelling were 5×10^{19} m^{-3}, 1.9 nm and 0.0001%, respectively. In the 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. The results suggest that the processes of nucleation and growth of cavities and dislocation loops may be slightly influenced by the presence of hydrogen atoms. In this study, the densities of dislocations were similar to each other in three simulation irradiation conditions, and this discrepancy is related to the difference of irradiation temperature. While, the densities of cavity formed by triple ion beams was nearly equal to that formed by dual ion beams, and the size of cavities in triple beams was remarkably larger than that in dual beams. On the measurement of hydrogen concentration after the irradiation at 470 °C showed that the migration of hydrogen atoms was very fast at 470 °C. It is suggested that hydrogen atoms could not stay inside cavities for long time. The densities of dislocations and cavities are very important for the swelling behavior. In order to explain the swelling behavior, it needs to examine the equation of cavity growth process as described below;

$$\mathrm{d}S/\mathrm{d}t = P\delta Z_c Q/(QZ_d + Z_c)^2 - 4\pi r_c N_c Z_c C_v^0 \times \exp[-(P_g - 2\gamma/r)\Omega/kT], \quad (1)$$

where P, Z_c and Z_d are the production rate of point defects, capture site number of point defect for cavity, capture site number of point defect for dislocation, respectively; $\delta = Z(i)_d - Z(v)_d$, $Q = \rho/(4\pi r_c N_c)$; ρ, N_c, r_c are the dislocation density, number density of cavity, mean cube of total cavity volume, respectively; C_v^0, P_g, γ, Ω are thermal vacancy concentration, pressure of gas atoms in cavity, surface energy of cavity, mean volume of cavity, respectively. From the Section 3, it can be expected that the fist term of Eq. (1) will be nearly same with each other in initial stage of irradiation for fusion conditions of dual and triple beams, because densities of cavities and dislocations were very similar to each other. Accordingly, the difference of cavity growth rate between the triple and dual beams under the fusion simulation irradiation may be due to gas pressure in cavity. The increase of gas pressure will be caused by the short time trapping of hydrogen atoms, which is induced by the contribution of the second term of Eq. (1). In case of irradiation at 600 °C, the swelling in simulation irradiation of spallation target vessel was larger than that in simulation irradiation of fusion reactors. The increase of swelling in the simulation irradiation of spallation target may be caused by higher gas pressure in cavity with higher concentration of helium.

4.2. Method of swelling suppression

The relation between the largest cavity size and dislocation density in F82H irradiated with dual ion beams at 510 °C to 50 dpa has been reported in [22]. The dislocation density was changed by adjusting the tempering time and temperature and cold-working. Cavity size decreased with increasing dislocation density. The method may be thought to be useful for exploring the reduction of swelling under triple beams. In present study, the effect of swelling suppression by cold working or carbon implantation has been examined. The carbon implantation was performed with energies from 1.0 to 2.5 MeV with a step of 0.3 MeV at 350 °C in order to reduce the formation of carbides and dislocation loops. Carbides are relatively stable at below 400 °C, and the formation of dislocation loops gradually decreases at above 300 °C. The implantation range for carbon atoms was corresponds to a range from 0.7 to 1.3 μm, which is nearly same with the helium and hydrogen implanted regions. The concentration of the implanted carbon atoms was estimated as about 90 appm in peak concentration, and the displacement damage was evaluated as 0.001 dpa in damage peak. Fig. 9(a), (b), (b′) and (c) show the cavities formed in F82H-std, F82H + 50%CW (area 1), F82H + 50%CW (area 2) and F82H + 90appmC, respectively, by triple ion beams under a simulation irradiation of fusion reactors. The swelling of F82H + 50%CW and F82H + 90appmC was about 1.4% and 0.5%, respectively, and it was successfully reduced by the methods. The cause of reduction of swelling by the carbon implantation may be due to the decrease of mobilities for point defects and the increase of recombination between vacancies and interstitials. The effect of cold-working on swelling may be mainly due to the decrease of vacancy concentration owing to the disappearance of vacancies and interstitials on high density dislocations.

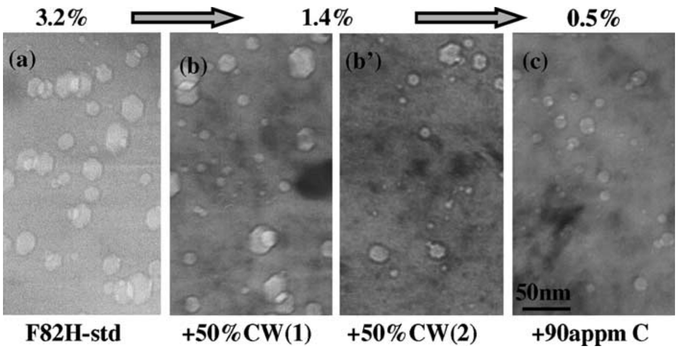

Fig. 9. Cavities formed at 470 °C to 50 dpa, (a) F82H-std, (b) F82H + 50% cold-worked (1), (b′) F82H + 50% cold-worked (2); (2) is a different area from (1), (c) F82H implanted with 90 appmC at 350 °C for about 100 s, 0.001 dpa, before triple beams.

5. Conclusions

Simulation irradiations of spallation target vessels and structural materials of fusion reactors were performed using simultaneous triple/dual ion beams at temperatures between 470 and 600 °C to 50 dpa.

1. The swelling was enhanced by a synergistic effect of displacement damage, helium and hydrogen.
2. The swelling of F82H steel under a simulation irradiation of fusion reactors was 3.2% at 470 °C in maximum, and it was larger than that in a simulation one of spallation target vessel, 1.2%.
3. The swelling under a simulation irradiation of structural materials of fusion reactors decreased with increasing temperature, however the swelling under a simulation of spallation target vessel again increased at 600 °C due to the high helium concentration.
4. Cavity density under the simulation of spallation target vessel was higher than that of the others.
5. The densities of cavity and dislocation decreased with the irradiation temperature and the densities of dislocations under those conditions were nearly equal at each temperature.
6. The main reason for the difference of swelling behavior between simulation irradiations of the spallation target vessel and structural material of fusion reactor is related to the size and density for cavity, and the difference between the triple and dual beams under a simulation of fusion reactor is related to the increase of gas pressure owing to the hydrogen atoms.
7. The swelling of F82H steel under the simulation irradiation of fusion reactor was reduced by 50% cold-working and carbon implantation, from 3.2% to 1.4% and 0.5%, respectively.
8. The hydrogen concentration measured by $^{1}H(^{15}N, \alpha\gamma)^{12}C$ resonance nuclear reaction in the F82H irradiated under a simulation of spallation target vessel at 80 or 470 °C was detected in only F82H irradiated at 80 °C in the hydrogen implanted region.

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