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Annealing effects on mechanical properties and microstructure of F82H irradiated at ⩽60 °C with 800 MeV protons

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Abstract

Martensitic steel F82H was irradiated with 800 MeV protons up to 7.8 dpa at low temperatures ⩽60 °C. Tensile tests have been performed at 22, 160, 250, 350 and 400 °C. The microstructure of specimens with doses of 6–7.8 dpa after annealing 20 min and 2 h at 160, 250, 350 and 400 °C has been studied. The tensile results show that the yield of irradiated specimens decreases gradually with increasing test temperature. The ductility starts to recover at about 300 °C and is substantially recovered at 400 °C. The recovery depends on irradiation dose. The TEM results indicate that the number density of dislocation loops induced by irradiation decreases with annealing temperature, while the mean size of loops increases only slightly at 400 °C. A part of dislocation loops transferred into network dislocations and resulted in increase of network dislocation density. However, the total dislocation density (including loops) decreases gradually with annealing temperature.

1. Introduction

The irradiation induced changes in microstructure and mechanical properties of materials irradiated at low temperatures can be partly or even completely recovered when they are annealed at higher temperatures. Although a lot of annealing experiments have been done on materials irradiated with neutrons in fission reactors, similar investigations on materials irradiated with high-energy protons and neutrons in spallation targets are still very limited, especially for studies on microstructure.

In the case of the irradiation in SINQ targets (STIP), the irradiation temperature and dose depend strongly on the position in the target due to relatively small size of the proton beam [1]. The number of specimens with exactly same irradiation condition is very limited. The strong position dependence of irradiation condition makes the comparison between specimens and materials more difficult. Considering the operation temperatures at the beam windows of liquid metal target containers in different spallation sources are mostly close to 200 and 250 °C, a common test temperature of 250 °C was selected by the STIP partners [2–4]. However, for the sake of developing the liquid lead–bismuth target for the megawatt pilot target experiment (MEGAPIE) [5],

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it is also interesting to investigate the behaviour of irradiated materials at around 350 °C which is higher than the irradiation temperature of most STIP-I specimens. It is of essential interest to study annealing effects on the microstructure and mechanical properties of materials irradiated at low temperatures. The F82H specimens irradiated in the acceleration production of tritium (APT) program [6] are ideal for the purpose since they were irradiated at 60 °C and below. In the present work, a series of tensile tests were performed at 22, 250, 350 and 400 °C for investigating the changes in tensile properties, and meanwhile, transmission electron microscopy (TEM) observations were performed for studying the microstructural changes after corresponding tensile tests and additional annealing.

2. Experimental

Material of F82H was obtained from the fusion materials program [7]. The specimens were EDM cut from a 15 mm thick plate (IEA Heat 9741). The nominal composition is: Fe+ 7.7Cr, 0.16Mn, 0.16V, 1.95W, 0.02Ta, 0.11Si and 0.09C in wt%. The plate was normalized at 1040 °C for 38 min and tempered at 750 °C for 1 h.

Miniature type tensile specimens of 0.25 mm thick, 5 mm long and 1.2 mm wide in gauge area were irradiated with other specimens of the APT materials irradiation program at the Los Alamos Neutron Science Center (LANSCE) [5] to doses up to 7.8 dpa at 60 °C and below. More detailed information about the irradiation of these tensile specimens can be found in Ref. [6].

Tensile tests were performed on a 2 kN MTS mechanical test machine equipped with a video-extensometer so that the displacement was measured directly from the gauge area. The tests were performed at 22, 160, 250, 350 and 400 °C with a strain rate of about $10^{-3}\ s^{-1}$. Each tensile test was run until the specimen failed. Tests at temperatures above room temperature (22 °C) were performed 15–20 min after the temperature stabilized. At each temperature, three specimens of doses about 0.8, 1.7 and 6–7.8 dpa were tested.

Four discs of 1.0 mm diameter were punched from the grip sections of each tensile specimen for transmission electron microscopy (TEM) observation. To investigate annealing time effects, two of the four discs were again heated up to the tensile test temperature and aged for 2 h. The TEM investigation on the microstructure was performed with a JEOL 2010 type microscope equipped with an EDX analysis system. The most often used image conditions were bright field (BF) and weak beam dark field (WBDF) at $(\mathbf{g}, 4\mathbf{g})$ or $(\mathbf{g}, 6\mathbf{g})$, $\mathbf{g} = \mathbf{110}$. For all specimens, only the micrographs of $\mathbf{g}(5\mathbf{g})$ $\mathbf{g} = \mathbf{110}$, $\mathbf{z} = \mathbf{111}$ were used for quantifying the size and number density of defect clusters. The thickness of a thin foil was deduced from the number of fringes, which had an uncertainty of about ±15%.

3. Results

3.1. Tensile tests

Fig. 1 presents the engineering tensile stress–strain curves of the specimens tested at 22, 160, 250, 350 and 400 °C. It can be seen that the yield stress shows a general trend of gradually decreasing with increasing test temperature. As illustrated in Fig. 2, the yield stress shows a linear dependence of test temperature except for that of the specimen of 1.8 dpa and tested at 400 °C. At 250 °C and below, the tensile curves of the specimens with similar doses resemble each other. The

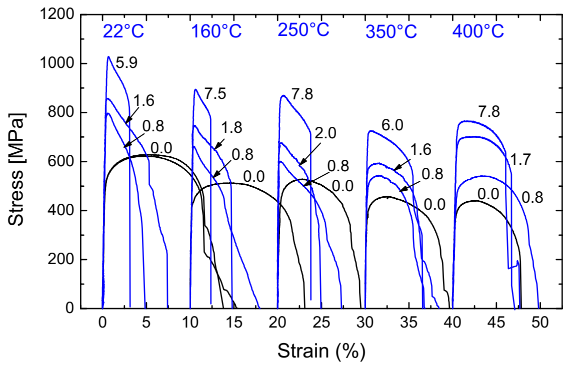

Fig. 1. The engineering tensile stress–strain curves of the irradiated and unirradiated F82H specimens tested at 22, 160, 250, 350 and 400 °C. The shift in x-axis between curves at different temperatures is 10%.

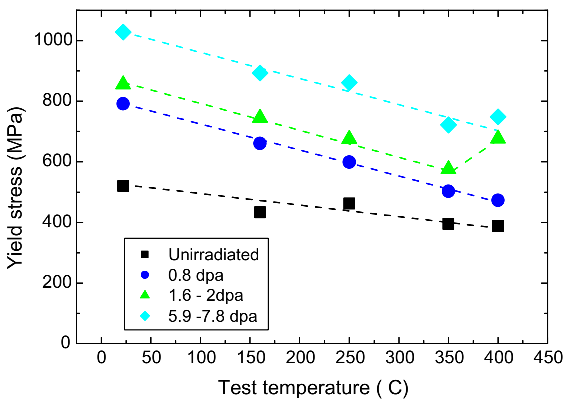

Fig. 2. The test temperature dependence of the yield stress of the irradiated and unirradiated F82H specimens.

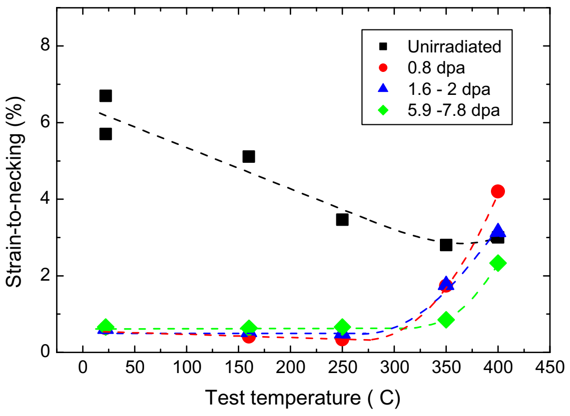

Fig. 3. The test temperature dependence of the strain-to-necking of the irradiated and unirradiated F82H specimens.

uniform elongation or strain-to-necking (STN) of the irradiated specimens is generally less than 1%. The STN starts to recover at a temperature about 300 °C, as can be seen in Fig. 3. At 350 °C, the STN recovers slightly for the specimen of 6.0 dpa, and to about 1.7% for the specimens with lower doses of 0.8 and 1.6 dpa. At 400 °C, the STN is more or less fully recovered. The values of yield stress, ultimate tensile stress, strain-to-necking and total strain of each curve are listed in Table 1. The value of the total strain is taken at the breaking point where the first large stress drop takes place.

3.2. TEM observations

The microstructure of as-irradiated specimens was investigated and reported in our previous paper [8]. The results showed that the main feature of the change in microstructure induced by radiation damage was the appearance of small defect clusters or dislocation loops. Both the mean size and density of the loops increased with irradiation dose in a range of 0.8–5.9 dpa.

The dislocation structure induced by irradiation at 60 °C and below will change after annealing at higher temperatures. Fig. 4 shows the dislocation structure in specimens irradiated to 5.9 and 7.8 dpa at different conditions: as-irradiated, and annealed about 20 min at 250, 350 and 400 °C. The detailed quantitative results are given in Table 2, including those previously reported [8]. The annealing temperature dependence of the size and density of dislocation loops is illustrated in Fig. 5. It can be seen that the size of dislocation loops increases with annealing temperature. The density does not change much at 350 °C and below, but decreases significantly at 400 °C. Considering the experimental

Table 1
Irradiation dose, calculated He and H concentrations, yield stress ($YS_{0.2}$), ultimate tensile strength (UTS), strain-to-necking, and total strain of the F82H specimens tested at 22, 160, 250, 350 and 400 °C

Temperature (°C)	Dose (dpa)	He (appm)	H (appm)	$YS_{0.2}$ (MPa)	UTS (MPa)	Strain-to-necking (%)	Total strain (%)
22[a]	0	–	–	520	620	5.7	11.3
22[a]	0	–	–	520	627	6.7	11.5
22[a]	0.8	125	365	792	796	0.63	4.0
22[a]	1.6	235	710	855	857	0.61	5.3
22[a]	5.9	1055	2673	1028	1028	0.66	3.03
160	0	–	–	433	513	5.11	11.58
160	0.8	125	365	661	661	0.41	3.89
160	1.8	270	815	745	745	0.53	4.7
160	7.5	1380	3380	893	893	0.63	2.34
250	0	–	–	462	528	3.46	8.3
250	0.8	125	365	599	599	0.34	5.5
250	2.0	305	890	675	675	0.49	4.29
250	7.8	1440	3505	861	870	0.66	3.78
350	0	–	–	395	458	2.8	8.6
350	0.8	125	365	503	543	1.74	5.47
350	1.6	235	710	575	593	1.76	5.94
350	6.0	1055	2670	722	726	0.85	5.47
400	0	–	–	387	439	3.0	7.7
400	0.8	125	363	473	541	4.2	8.66
400	1.7	250	760	676	701	3.14	6.67
400	7.8	1440	3505	748	765	2.33	6.03

[a] Results from the previous work [8].

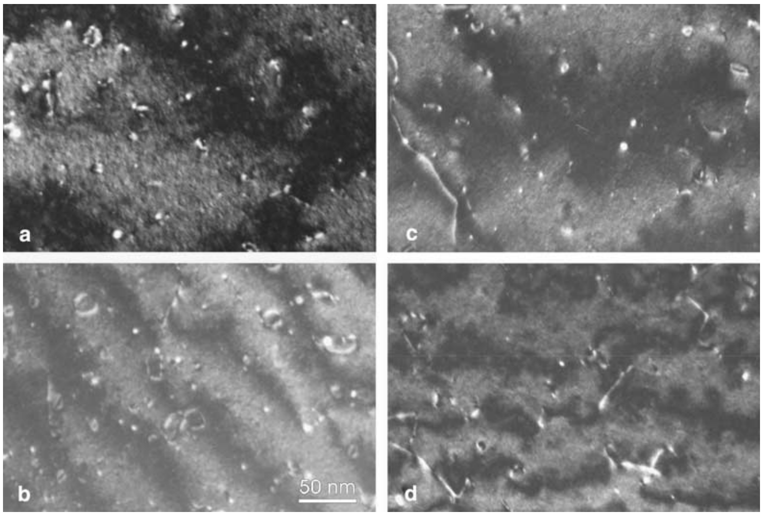

Fig. 4. The dislocation structure in irradiated specimens at different conditions: (a) 5.9 dpa as-irradiated; (b) 7.8 dpa tested at 250 °C; (c) 5.9 dpa tested at 350 °C; (d) 7.8 dpa tested at 400 °C. The holding time at each test temperature is about 20 min.

Table 2
The irradiation and annealing conditions and TEM measurement results of the F82H specimens

Specimen	Dose (dpa)	Test or annealing temperature and time	Mean loop size (nm)	Loop density (m^{-3})	Dislocation line density (m^{-2})	Total dislocation line density (m^{-2})
As-received	–	–	–	–	1.2×10^{14}	1.2×10^{14}
I01[a]	0.8	22	2.1	1.6×10^{22}	–	–
I02[a]	1.6	22	2.4	2.4×10^{22}	–	–
I03[a]	5.9	22	3.1	3.3×10^{22}	–	–
I09-1	7.5	160 °C/0.3 h	5.0	4.0×10^{22}	1.2×10^{14}	7.5×10^{14}
I13-1	7.8	250 °C/0.3 h	4.4	4.1×10^{22}	1.2×10^{14}	6.9×10^{14}
I16-1	6.0	350 °C/0.3 h	4.3	3.4×10^{22}	1.6×10^{14}	6.1×10^{14}
I20-1	7.8	400 °C/0.3 h	5.0	2.2×10^{22}	2.2×10^{14}	5.7×10^{14}
I13-2	7.8	250 °C/2 h	4.6	4.4×10^{22}	1.4×10^{14}	6.8×10^{14}
I16-2	6.0	350 °C/2 h	4.8	2.4×10^{22}	2.0×10^{14}	5.5×10^{14}
I20-2	7.8	400 °C/2 h	5.6	1.9×10^{22}	3.0×10^{14}	6.4×10^{14}

[a] Results from the previous work [8].

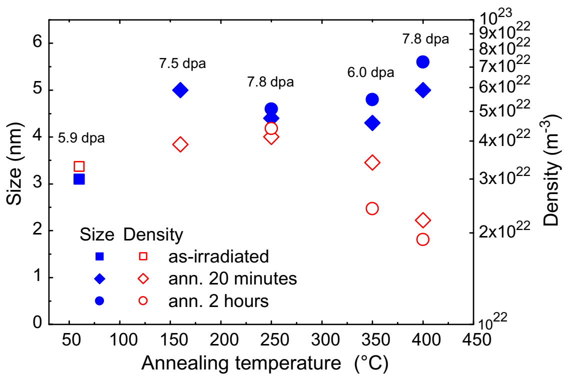

Fig. 5. The annealing temperature dependence of the size and density of small dislocation loops in the F82H specimens irradiated to 6 and 7.8 dpa at ⩽60 °C.

uncertainty of about ±15%, the additional annealing up to 2 h made only slight change in the dislocation structure.

The line density of the network dislocations is counted from the number of dislocation line per unit area. The results indicate that the line density of the network dislocations increased after annealing, especially after annealing 350 and 400 °C, as can seen in Fig. 6. It is interesting to note that the total dislocation line density is nearly the same after annealing for 20 min and 2 h. The total dislocation line density is the network dislocation line density plus the total line length converted from the small dislocation loops.

In all the specimens no bubbles were observed, although the helium concentration is about 1400 appm in specimens of 7.8 dpa.

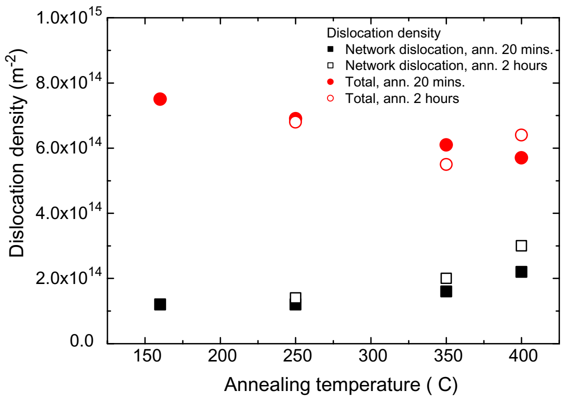

Fig. 6. The annealing temperature dependence of network dislocation density and total dislocation density including the contribution of small dislocation loops in the F82H specimens irradiated to 6 and 7.8 dpa at ⩽60 °C.

4. Discussion

The temperature dependence of the yield strength of unirradiated martensitic steels is well established. It indicates that the yield stress decreases gradually with increasing temperature up to about 400 °C and more steeply above 450 °C [9]. The present results of unirradiated specimens agree with this. For irradiated specimens, the yield stress shows a stronger dependence on test temperature as can be seen in Fig. 2. The quicker decrease of yield stress of the irradiated specimens should be attributed to the decrease of the total dislocation density caused by the annealing-out of small dislocation loops. It is interesting to note that the slopes for specimens of different doses are essentially the same, at least for those tested ⩽350 °C. This is not understood yet due to the lack of TEM observations on the specimens of lower doses. For the same reason, it is not clear why the specimen of 1.7 dpa tested at 400 °C shows a higher yield stress as compared to the specimen of 1.6 dpa tested at 350 °C. The explanation needs further TEM observations on specimens of lower doses.

Fig. 7 shows the microstructure of F82H specimens irradiated in STIP at higher temperatures to higher doses of 10–12 dpa [10]. Comparing Fig. 2 with Fig. 7 it can be seen that the features of the dislocation microstructure of specimens irradiated at ⩽60 °C after annealing at higher temperatures are very similar to those irradiated at the corresponding temperatures. However, in the specimen irradiated at 360 ± 30 °C, high-density helium bubbles of about 1.6 nm were observed, while no bubbles were seen in the specimens annealed at 350 and 400 °C for 2 h. This is believed to be due to the low annealing temperature. Systematic annealing experiments on helium implanted martensitic steel specimens by Kimura et al. demonstrated that helium bubbles or voids could not be observed after annealing below 450 °C [11].

Using annealing technique to extend the lifetime of fission reactors has been applied to the pressure vessels of a number of nuclear power plants, which had doses <1 dpa and have been annealed in a temperature range

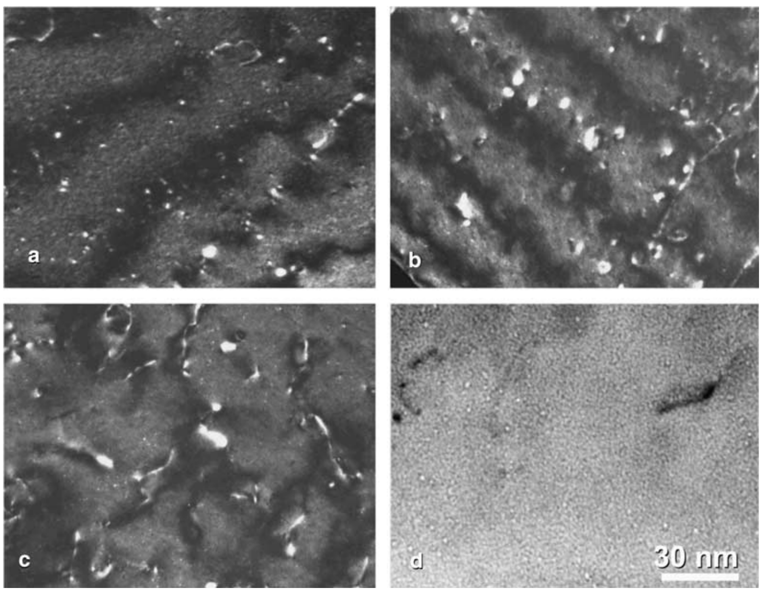

Fig. 7. The dislocation and helium bubble structures in the F82H steel irradiated in a SINQ target: (a) 10.0 dpa irradiated at 140 ± 13 °C; (b) 10.3 dpa irradiated at 255 ± 22 °C; (c) and (d) 12.2 dpa irradiated at 360 ± 30 °C [10].

of 430–475 °C [12,13]. Khabarov et al. [14] performed Charpy impact tests on steel, 13Cr2MoNbVB, irradiated in fast reactors to 4–85 dpa and at 280–350 °C. They reported that the upper-shelf energy started to recover after annealing 1 h at temperatures 450–470 °C and fully recovered after annealing 1 h at 550 °C. The present tensile tests show that the ductility begin to recover at about 300 °C, and fully recover at 400 °C for specimens of doses ⩽2 dpa, while that of the specimen of 7.8 dpa is nearly fully recovered. The results demonstrate clearly that the recovery of ductility and irradiation hardening depend not only on annealing temperature but also on irradiation dose. Specimens of higher doses need a higher annealing temperature or a longer annealing time for a complete recovery. Chen et al. performed similar annealing tests on DIN1.4926 martensitic steel irradiated at ⩽220 °C at LANSCE to 6 dpa [15]. The results of tensile tests at room temperature indicate that the ductility significantly recovered after annealing at 400 °C and nearly fully recovered after annealing at 700 °C. The recovery degree is somewhat lower than that shown in the present work. This indicates that the recovery process might be also irradiation temperature dependent. The irradiation induced microstructural changes at relative higher temperatures may also need at much higher temperatures to anneal out. However, the annealing temperature should be kept as low as possible in order to avoid inducing additional harmful microstructural changes such as formation of new precipitates, large helium bubbles or voids, and also technical difficulties for high temperature annealing of large components.

5. Conclusions

Martensitic steel, F82H was irradiated with 800 MeV protons up to 7.8 dpa at low temperatures ⩽60 °C. Tensile tests have been performed at 22, 160, 250, 350 and 400 °C. The microstructure of specimens with doses of 6–7.8 dpa after annealing for 20 min and 2 h at 160, 250, 350 and 400 °C has been studied. The results demonstrate that:

(1) The yield stress of irradiated specimens decreases gradually with increasing test temperature, while the ductility starts to recover at about 300 °C and is fully recovered at 400 °C for specimens of doses <2 dpa and almost fully recovered for specimen of 7.8 dpa. The degree of recovery depends both on irradiation dose and irradiation temperature.

(2) The number density of dislocation loops decreases with annealing temperature, while the mean size of loops increases only slightly at 400 °C. A part of dislocation loops have transferred into network dislocations and result in the increase of network dislocation density. However, the total dislocation density (including loops) decreases gradually with annealing temperature.

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