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Defect structure development in a pure iron and dilute iron alloys irradiated with neutrons and electrons

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Abstract

The defect structure and mechanical property changes were observed for pure iron of 99.99% purity and a series of Fe–(0.1% and 0.4%) Cr and Fe–(0.1% and 0.4%) Mn dilute alloys irradiated with neutrons. From the comparison of the defect structures with yield strength change, a large contribution of the invisible defect clusters to the irradiation hardening was expected in the specimens irradiated in Japan Materials Test Reactor (JMTR), whereas these clusters are not found after irradiation in the Fast Flux Test Facility (FFTF). The electron irradiation experiments showed that addition of chromium and manganese to 0.1% in pure iron, development of large dislocation loops is suppressed, and frequent nucleation of small loops at the early stage of the electron irradiation is observed, similar to that in ultra-high purity iron of 99.9999% purity. The mechanisms of dislocation loop development in the early stage of irradiation for Fe–Cr and Fe–Mn are considered to be different.

1. Introduction

The authors have investigated the mechanical property changes due to neutron irradiation in terms of the defect structures [1,2]. In a previous paper [3], a correlation between the mechanical property change and defect structures in two series of iron base dilute binary alloys of Fe–Cr and Fe–Mn is reported. In these alloy series, an anomalous change in the irradiation hardening was observed at alloying concentration of 0.1% for the specimens irradiated in the Fast Flux Test Facility (FFTF), in the USA. Hardening in the specimens irradiated to 7×10^{26} n/m^2 in FFTF was much smaller than that in pure iron of 99.99% purity and alloys of much higher Cr and Mn levels, whereas in the specimens irradiated to 1.0×10^{24} n/m^2 in JMTR (Japan Materials Test Reactor) the hardening increased with increase in the alloying concentration. This suggests that a small addition of Cr and Mn brings about reduction in the irradiation hardening at higher fluences which is not observed for the fluence levels of JMTR.

In the present experiment, the origin of the difference between JMTR and FFTF irradiations as described above was considered from the viewpoint of the defect structures and the defect structure development during electron irradiation.

2. Experimental

2.1. Specimen preparation for neutron irradiation and electron irradiation

A pure iron of 99.99% purity (hereafter referred to as 4n-Fe) and two series of iron base dilute binary alloys, Fe–(0.1 and 0.4 wt%) Cr and Fe–(0.1% and 0.4%) Mn, were prepared by melting 4n-Fe with 99.9% pure chromium and manganese. The specimens for the neutron and electron irradiations, disks of 3 mm diameter(TEM disks) with a thickness of 0.2 mm for FFTF and 0.1 mm for JMTR, and miniaturized tensile specimens whose gauge section was 1.2 mm × 5 mm with a thickness of 0.25 mm for FFTF and 0.20 mm for JMTR were

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punched from cold rolled sheet followed by annealing at 1173 K for 1.8 ks in H_2. The grain size was about 40 μm. Preparation of alloy sheet was identical for each of the alloys.

For the electron irradiation experiments, a series of Fe–Mn alloys with 0.05%, 0.1% and 0.7% Mn were also prepared. As reference material for the electron irradiation experiments, thin sheet about 20 μm thick, of an ultra-high purity 99.9999% iron (referred to as 6n-Fe, provided by M. Isshiki of Tohoku University) was also prepared by cold rolling thin wire and careful annealing at 1173 K for 1.8 ks in a vacuum of 10^{-6} Torr.

2.2. Neutron irradiation

Both TEM disks and tensile specimens were concurrently irradiated to 1×10^{24} n/m^2 at 673 K in JMTR and to 7×10^{26} n/m^2 at 698 K in FFTF (both fluences are for neutrons of $E > 0.1$ MeV). Irradiation dose is almost 3 orders of magnitude higher for FFTF irradiation. In JMTR, undesired irradiation during start up and shut down of the reactor was avoided by using a temperature controllable rig equipped with an electric resistance heater [4].

2.3. Tensile testing, TEM observation and electron irradiation

The tensile tests were carried out at a constant strain rate of 5.5×10^{-4}/s at room temperature. The yield strength at 0.2% offset and the strain hardening exponent were calculated from load–elongation curves.

TEM observations for the defect structures were made for the disks irradiated with neutrons. The electron irradiation experiment was carried out at a constant dose rate of 5×10^{22} e/m^2 s by using an ultra-high voltage electron microscope operated at 1 MV. Irradiation temperatures were in a range 473–773 K.

3. Results and discussion

3.1. Mechanical property change from neutron irradiation

The variation of the yield strength with Cr and Mn concentrations before and after neutron irradiation is shown in Fig. 1(a) and (b), and increment of yield strength is plotted in Fig. 1(c) and (d). In the figure, before irradiation, the yield strength of the 0.1% (Cr, Mn) alloy is a little higher than for both 4n-Fe and the

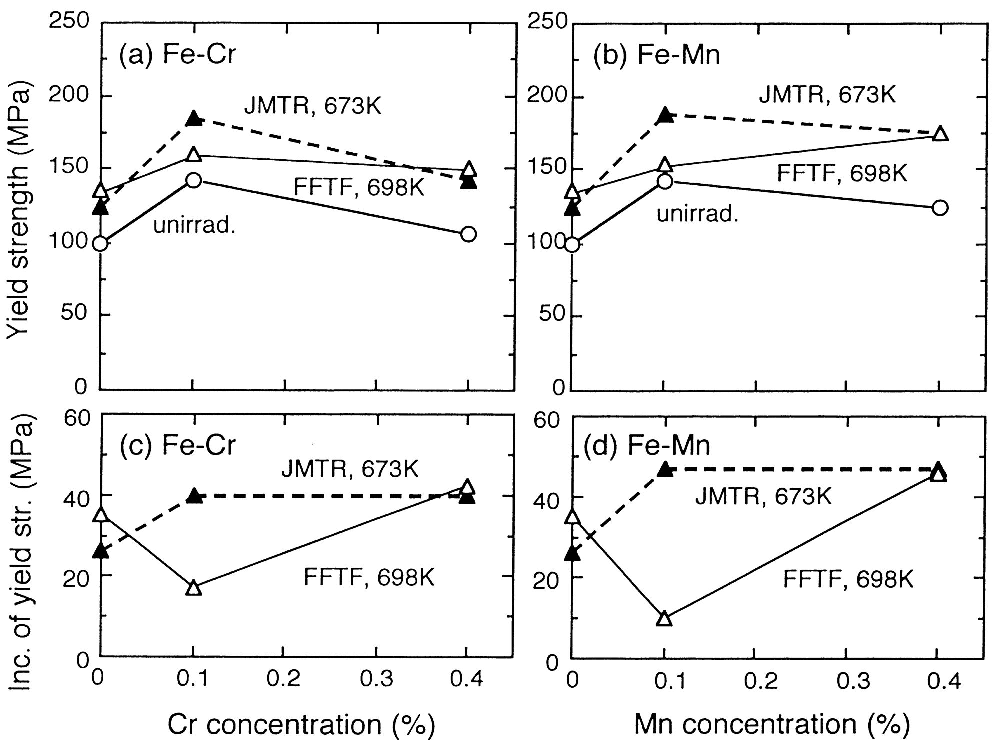

Fig. 1. Yield strength change of the Fe–Cr (a) and Fe–Mn alloys (b), increment of yield strength of the Fe–Cr (c) and Fe–Mn alloys (d) after irradiation in JMTR (closed triangles) and FFTF (open triangles), respectively.

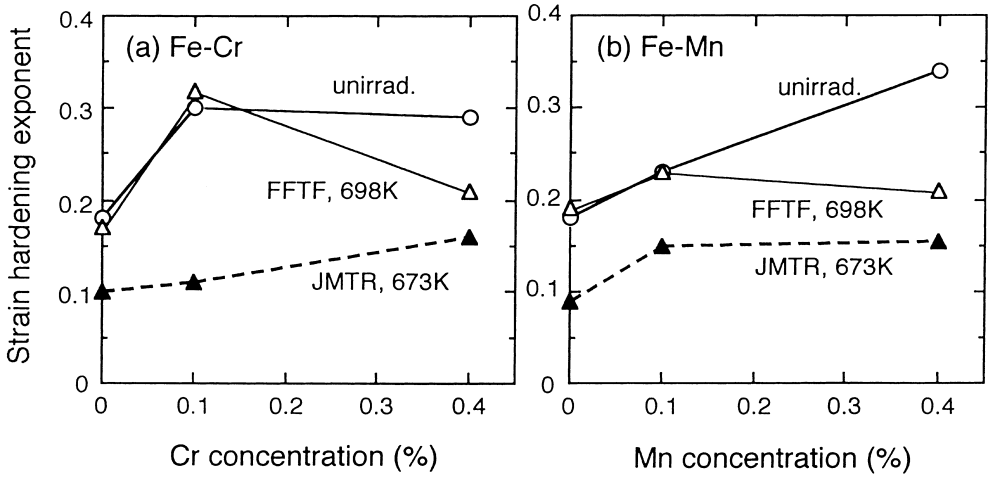

Fig. 2. Variation of the strain hardening exponent of the Fe–Cr (a) and Fe–Mn alloys (b), irradiated in JMTR (closed triangles) and FFTF (open triangles).

0.4% (Cr,Mn) alloys. After irradiation in JMTR, plots of the yield strength shifted upward by almost the same amount. While after irradiation in FFTF, the increase in the yield strength of the 4n-Fe, 0.4%Cr and 0.4%Mn is almost the same as that for the JMTR irradiation, although the defect structure is quite different in both irradiations as shown in the next section. However, the increase in the yield strength of the Fe–0.1%Cr and Fe–0.1%Mn alloys irradiated in FFTF were only 1/2 and 1/4, respectively, of that for each of alloys irradiated in JMTR as seen in Fig. 1(c) and (d). A large decrease in the strain hardening exponent can be seen for both 0.4%Cr and 0.4%Mn alloys irradiated in FFTF, however, the reduction is small compared with that for the JMTR irradiation (shown by closed triangles), while the exponent of the 4n-Fe and 0.1% (Cr,Mn) alloys does not change after irradiation in FFTF as seen in Fig. 2(a) and (b). These results show that irradiation to much higher doses than that for JMTR results in the recovery of reduced ductility due to irradiation at a critical alloying concentration depending upon the base metal.

3.2. Defect structures

3.2.1. Neutron irradiated pure Iron, Fe–Cr and Fe–Mn alloys

The dislocation loop number densities and dislocation densities of alloys irradiated in JMTR and FFTF observed by TEM are shown in Figs. 3 and 4, respectively. In Fig. 3(a), the dislocation loop number density

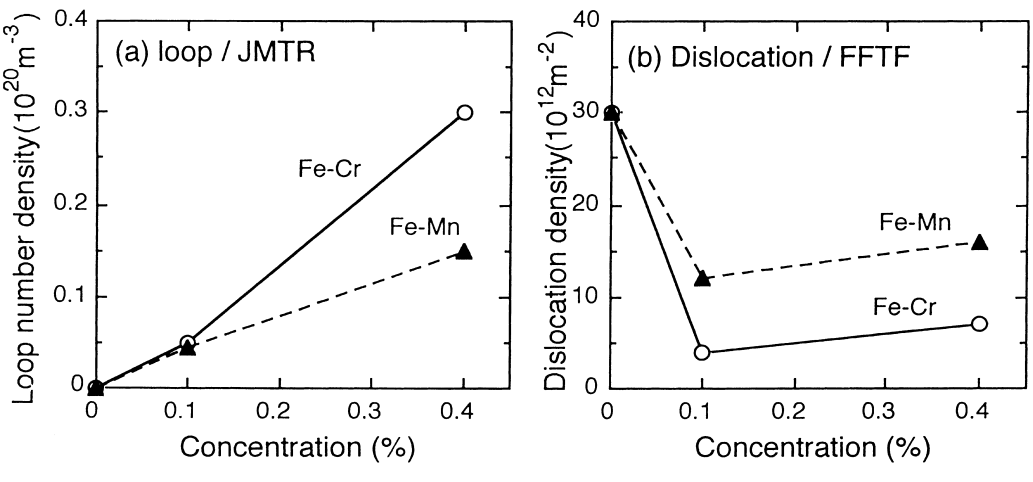

Fig. 3. Dislocation number density (a) and dislocation density (b) plotted for Fe–Cr (open circles) and Fe–Mn alloys (closed triangles), irradiated in JMTR and FFTF, respectively.

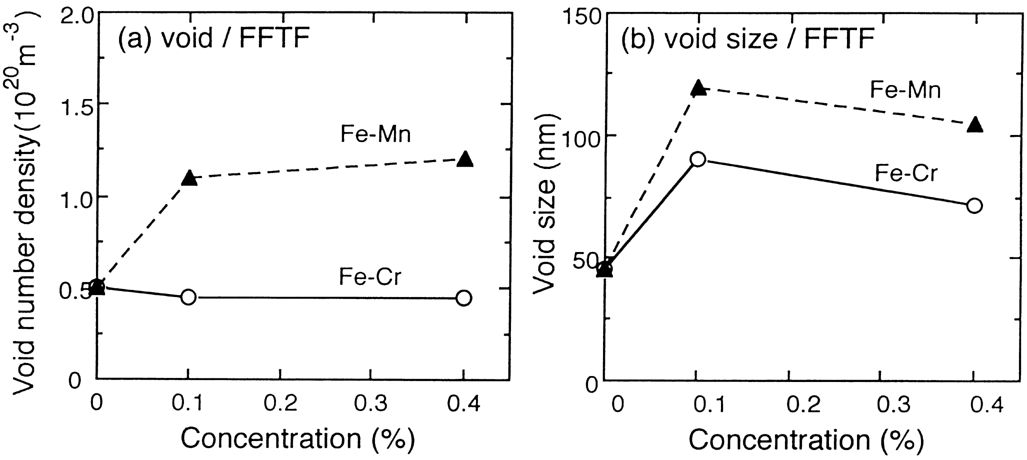

Fig. 4. Void number density (a) and void size (b) plotted for the Fe–Cr (open circles) and Fe–Mn alloy (closed triangles), irradiated in FFTF.

is much smaller for the 4n-Fe, 0.1% (Cr,Mn) alloys than that of the 0.4% (Cr,Mn) alloys irradiated in JMTR. Therefore, if the irradiation hardening in these alloys is caused by the dislocation loops, the 0.4% (Cr,Mn) alloys must show much larger increment in the yield strength than the others. However, this expectation differs from the experiment, viz., the increment of yield strength of the 0.1% and 0.4% (Cr,Mn) alloys are almost equal. In the Fe–0.1%Mn alloy, voids smaller than 25 nm could be observed, but whose number density was not large enough to explain this discrepancy [3]. Such an unexpected result differing from the defect structure observation suggests a larger contribution from invisible defect clusters, such as micro-voids [5] to hardening of the 4n-Fe and 0.1% (Cr,Mn) alloys than the 0.4% (Cr,Mn) alloys [3].

For the irradiation in FFTF, the dislocation densities of both 0.1%Cr, 0.4%Cr and 0.4%Mn alloys are much smaller than of 4n-Fe as shown in Fig. 3(b), and the void number density variation with chromium and manganese concentration cannot be seen between 0.1% and 0.4% as seen in Fig. 4. The void size is maximum at 0.1%Cr and 0.1%Mn, and voids are much larger than for 4n-Fe. It should be noted that although the void size is maximum at 0.1%Cr and 0.1%Mn, with similar dislocation densities for the 0.4%Cr and 0.4%Mn alloys, the increase in yield strength is minimum.

Dislocation densities of the Fe–(Cr,Mn) alloys irradiated in JMTR increase with increases in concentration. While in the specimens irradiated in FFTF, the defect structures can be considered to have developed after the growth stage arising from the growth of dislocation loops later than that of JMTR irradiation, and the dislocation densities of the 0.1% and 0.4% (Cr,Mn) alloys are rather much smaller than that for 4n-Fe. This must be because the dislocations remain from the coalescence of loops. We can conclude from the neutron irradiation experiment that manganese atoms enhance both void nucleation (Fig. 4(a)) and voids growth (Fig. 4(b)) at fluences as high as those from FFTF, and the former tendency is consistent with previous work, where we observed the evolution of tiny voids in the Fe–0.1%Mn irradiated in JMTR [3]. While chromium atoms enhance dislocation loop nucleation (Fig. 3(a)) and growth (Fig. 3(b)), as mentioned above, a smaller dislocation density is due to dislocation loop coalescence. Both manganese and chromium atoms in critical concentration (about 0.1%) reduce the defect evolution.

3.2.2. Electron irradiated pure iron, Fe–Mn and Fe–Cr alloys

The defect structures developed after electron irradiation for 150 s at 573 K are shown Fig. 5(a)–(d). Small dislocation loops at high density with irregularly curved dislocation lines are seen in the 6n-Fe as shown in Fig. 5(a). In the 4n-Fe, a disk shaped loop in which curved parallel lines can be seen (shown by an arrow A), and several rod like parallel images can be seen as Fig. 5(b). The rod images are the side views of the disk shaped loops.

In the Fe–0.1%Mn as shown in Fig. 5(c), observed from near a [1 0 0] direction, the number and size of dislocation loops is much smaller compared to that for 4n-Fe. Dark spotty images show side views of the loops and this indicates that loop growth was limited in this alloy. In the Fe–0.7%Mn, very small dislocation loops of irregular shape and segments of dotted lines arranging parallel to one of the ⟨1 0 0⟩ directions crossing each other at a right angle can be seen in Fig. 5(d). This feature is typical for alloys with concentrations higher than several tenths percent.

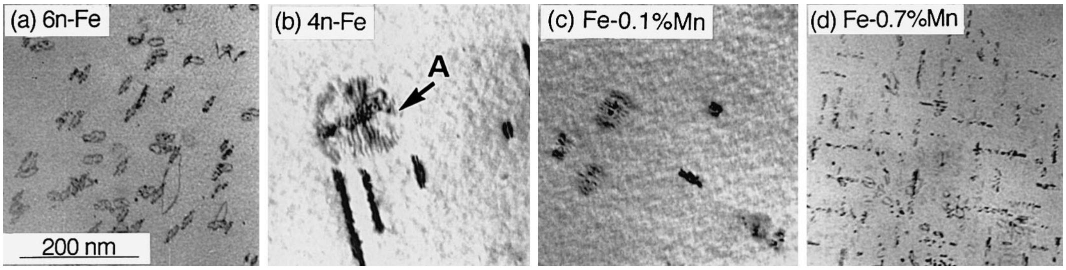

Fig. 5. Defect structures after irradiated with electrons for 150 s at 573 K. Ultra-high purity iron (6n-Fe) of 99.9999% purity (a), pure iron (4n-Fe) of 99.99% purity (b), Fe–0.1%Mn (c) and Fe–0.7%Mn (d). The foil planes of (b) to (d) are close to (1 0 0), and (a) is close to (1 1 1), respectively.

After irradiation for 150 s at 623 K, very small loops are observed for the 6n-Fe as show in Fig. 6(a). Reduction of the number density and size is observed by comparing with that observed for the irradiation at 573 K as was shown in Fig. 5(a). Dislocation loops in 4n-Fe are larger at increased number density as seen in Fig. 6(b). Fig. 6(c) shows loops developed in Fe–0.1%Mn. This defect structure seems to be similar to that of the 6n-Fe.

Fig. 6(d) shows two kinds of images; structures of dislocation loops whose dislocation lines are multiplely folded to form a maze pattern, and irregularly shaped large curved loops. The side view of such loops are seen as bold straight line segments crossing each other at right angles. This shows each of dislocation loop is lying close to {1 0 0} planes.

Fig. 7(a) and (b) shows the development of loops in the Fe–0.1%Mn irradiated for 150 and 200 s, respectively. While in the Fe–0.1%Cr as shown in Fig. 7(c), after a very short irradiation time (20 s), small dislocation loops are generated. Much faster growth of the loops than that for the Fe–Mn alloys is observed, viz., after continuing irradiation for a shorter time (less than 100 s), dislocation loops grow much larger, as shown in Fig. 7(d).

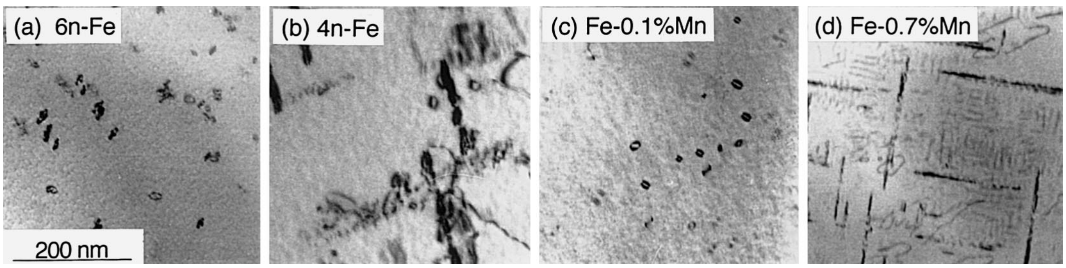

Fig. 6. Defect structures after irradiated with electrons for 150 s at 623 K. Ultra-high purity iron (6n-Fe) of 99.9999% purity (a), pure iron (4n-Fe) of 99.99% purity (b), Fe–0.1%Mn (c) and Fe–0.7%Mn (d). The foil planes are close to (1 1 1) for (a), (1 0 0) for (b) (1 1 3) for (c), respectively.

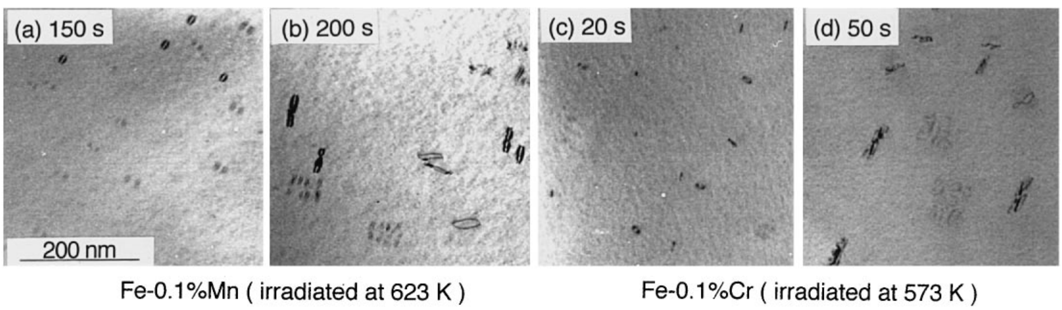

Fig. 7. Dislocation loops of Fe–0.1%Mn after irradiation for 150 s (a) and 200 s (b) at 623 K, and Fe–0.1%Cr irradiated for 20 s (c) and 50 s (d) at 573 K. The foil planes are close to (1 1 0) for (a) and (b), and (1 1 1) for (c) and (d), respectively.

Manganese and chromium additions at about 0.1% to the 4n-Fe suppress the dislocation loop development, although both chromium and manganese additions reduced the number density of the dislocation loops compared to that for 6n-Fe.

4. Conclusions

Addition of a small amount (0.1%) of chromium and manganese to pure iron (purity 99.99%) brings about an increase in the irradiation hardening after irradiation to $\sim 10^{24}$ n/m^2 (such as obtained in JMTR), whereas, for higher fluences ($\sim 10^{26}$ n/m^2) as from FFTF, the increment of the yield strength of the Fe–(Cr,Mn) is much smaller than that for pure iron. For lower neutron fluences as obtained by irradiation in JMTR, small amounts of chromium and manganese enhance the irradiation hardening to form defect clusters that are difficult to observe by electron microscopy. Large drop in yield strength for 0.1% (Cr,Mn) alloys after irradiation to higher fluences suggest that the disappearance of these defect clusters occurs along with the development of large voids. Voids as large as 100 nm do not cause yield strength increases, but instead probably decrease the yield strength.

The electron irradiation experiment supports the conclusions derived from neutron irradiation. Rapid loop growth is observed for the Fe–0.1%Cr. The faster growth of dislocation loops in the Fe–Cr alloys provides more sites for vacancy sinks, and this reduces the likelihood for void nucleation and growth.

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