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Journal of Nuclear Materials 271&272 (1999) 97–101

journal of nuclear materials

Effects of neutron irradiation on microstructure and mechanical properties of pure iron

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Abstract

Tensile specimens of pure iron were irradiated with fission neutrons (i) at 320 K to displacement dose levels of 7.5×10^{-3}, 7.5×10^{-2} and 3.75×10^{-1} dpa (NRT) and (ii) at 523 K to dose levels of 7.5×10^{-2} and 2.25×10^{-1} dpa (NRT). Both unirradiated and irradiated specimens were tensile tested at the irradiation temperatures. Microstructures of the as-irradiated and irradiated and tensile tested specimens were investigated by transmission electron microscopy. Fracture surfaces of tensile tested specimens in unirradiated and irradiated conditions were examined in a scanning electron microscope. Results of these investigations are reported and discussed particularly in terms of the role of interstitial clusters (produced directly in the cascades) and their transport via one-dimensional glide. It is suggested that the formation and interaction of 'cleared' channels may play a significant role in determining the deformation and fracture behaviour of the irradiated pure iron.

1. Introduction

Effects of neutron irradiation on physical and mechanical properties of the low activation ferritic–martensitic steels are being extensively studied internationally (e.g. Japan, USA and Europe) since these alloys are considered to be candidate materials for the blanket and first wall of fusion reactors (e.g. DEMO and commercial) [1]. These alloys are considered to have a number of more attractive properties than alternative structural materials like ferritic and austenitic steels or vanadium alloys [2]. These considerations have led to the establishment of a comprehensive R&D programme within the framework of the European Fusion Technology Programme.

Although the ferritic–martensitic class of steels is very resistant to swelling and maintains good fracture toughness at irradiation temperatures above about 673 K [3,4], they are prone to loss of ductility at lower irradiation temperatures [5,6]. This is a matter of concern from the point of view of mechanical performance and lifetime of these alloys under fusion irradiation conditions, particularly when the mechanism controlling the loss of ductility is not understood. The present work, which is a part of the European Fusion Technology Programme, was initiated to address the problem of irradiation-induced loss of ductility in these alloys. It was recognized, however, that the complexity of the microstructure in these alloys does not render them attractive candidates for mechanistic studies. It was therefore decided to focus the investigations first on pure iron. In the following the main results of the present investigations on the effect of neutron irradiation on the microstructural evolution and mechanical properties of pure iron are presented and discussed.

2. Materials and experimental procedure

Thin sheets (0.25 mm thick) of pure iron (99.9999%; C < 0.01 ppm) were purchased from Goodfellow (England) in the cold-worked state. A series of annealing experiments was carried out to determine the appropriate annealing condition to give fully recrystallized material. The annealing at 923 K for 2 h in vacuum ($\sim 10^{-6}$ Torr) was selected to be the reference annealing condition. This treatment gave an average grain size of $\sim$30 μm. The tensile specimens of pure iron were annealed at

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923 K for 2 h (in vacuum of $\sim 10^{-6}$ Torr) prior to irradiation and tensile testing in the unirradiated condition.

The tensile specimens of pure iron were irradiated with fission neutrons in the DR-3 reactor at Risø National Laboratory (a) at 320 K to neutron fluences of 5×10^{22}, 5×10^{23} and 2.5×10^{24} n/m^2 ($E > 1$ MeV) corresponding to displacement dose levels of 7.5×10^{-3}, 7.5×10^{-2} and 3.75×10^{-1} dpa (NRT), respectively and (b) at 523 K to neutron fluences of 5×10^{23} and 1.5×10^{24} n/m^2 corresponding to displacement dose levels of 7.5×10^{-2} and 2.25×10^{-1} dpa (NRT). All irradiation experiments were carried out with a neutron flux of 2.5×10^{17} n/m^{2}s corresponding to a dose rate of 3.75×10^{-8} dpa (NRT)/s. Irradiations at 523 K were carried out in a temperature controlled rig where the irradiation temperature is monitored, controlled and recorded continuously. All irradiations were performed in an atmosphere of pure helium or a mixture of pure helium and pure argon.

Both unirradiated and irradiated specimens were tensile tested in an INSTRON machine at a strain rate of 1.3×10^{-3} s^{-1}. In all cases, irradiated specimens were tensile tested at the irradiation temperature in vacuum of $\sim 10^{-5}$ Torr (Table 1).

For transmission electron microscopy (TEM) investigations, 1 mm wide and $\sim$0.1 mm thick strips were prepared from the irradiated materials and were electropolished at 20 V in a solution of 20% perchloric acid in methanol at the ambient temperature. Thin foils were examined in a 200 keV JEOL 2000FX electron microscope. To examine the deformed microstructure, TEM samples were taken from the zones as close as possible to the fracture surfaces of the tensile tested specimens. The fracture surfaces were examined in a scanning electron microscope.

3. Experimental results

3.1. Post-irradiation microstructure

The TEM examinations of the irradiated specimens of pure iron showed the presence of small defect clusters and loops at all three doses. The dislocation line density in all cases was found to be very low ($< 10^{12}$ m^{-2}). An example of the as-irradiated defect micro-structure is shown in Fig. 1. The cluster density in pure iron irradiated at 320 K was found to increase with increasing dose. The dose dependence of the cluster density is shown in Fig. 2. For comparison, the dose dependence of cluster density in pure copper neutron irradiated at similar temperatures is also shown in Fig. 2.

The cluster density in pure iron irradiated at 523 K to a dose level of 2.25×10^{-1} dpa (NRT) was found to be 2.5×10^{21} m^{-3} (Fig. 2).

3.2. Pre- and Post-irradiation tensile properties

The deformation behaviour of pure iron irradiated to different doses at 320 K and tested at 320 K is illustrated in Fig. 3 in the form of stress–strain curves. The unirradiated pure iron deforms in a fairly normal way with characteristics common to pure bcc metals showing first the yield drop and then the Lüders strain followed finally by a substantial degree of work hardening. The post-irradiation tests show three clear effects of irradiation. First, the upper yield stress increases with the dose, second, the magnitude of the yield drop increases with the dose and finally the irradiated specimens show practically no ability to work harden.

Fig. 4 shows the stress–strain curves for pure iron tested in the unirradiated and irradiated conditions. Both irradiations and tensile tests were carried out at 523 K. First of all, it should be noted that in the case of the unirradiated pure iron although the upper yield stress at 523 K is lower than that at 320 K, the amount of work hardening is considerably higher than that at 320 K. Consequently, the ultimate tensile strength, σ_{max}, of the unirradiated pure iron at 523 K is higher not only than that of irradiated specimens at 523 K, but also than that of the unirradiated pure iron tested at 320 K. As regards the effect of irradiation, it can be clearly seen that the yield stress increases with the dose (Fig. 4). It should be noted that even at 523 K, there is a clear indication of an upper yield point and a small but finite Lüders strain in the specimen irradiated to a dose level of 2.25×10^{-1} dpa.

Table 1
Tensile properties of unirradiated and irradiated pure iron

Material	Dose (dpa)	Irrad. Temp. (K)	Test Temp. (K)	σ_y^u (MPa)	$\sigma_{0.2}$ (MPa)	σ_{max} (MPa)	ε_u^p (%)	ε_t (%)
Pure iron	0	–	320	235	–	315	24.5	27.0
	7.5×10^{-3}	320	320	275	–	275	25.7	28.5
	7.5×10^{-2}	320	320	302	–	302	16.2	19.2
	3.75×10^{-1}	320	320	414	–	398	15.1	16.8
	0	–	523	–	145	409	13.1	18.0
	7.5×10^{-2}	523	523	–	190	325	10.3	15.0
	2.25×10^{-1}	523	523	227	225	360	12.1	18.0

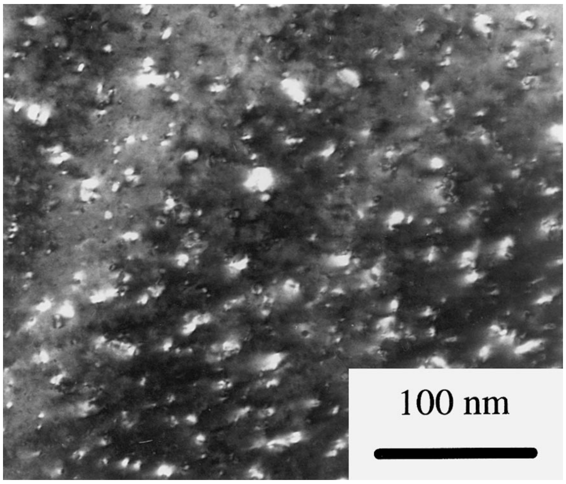

Fig. 1. Defect clusters in pure iron irradiated at 320 K to a dose level of ~3.75×10^{-1} dpa (NRT). The mean cluster size was found to be ~5 nm.

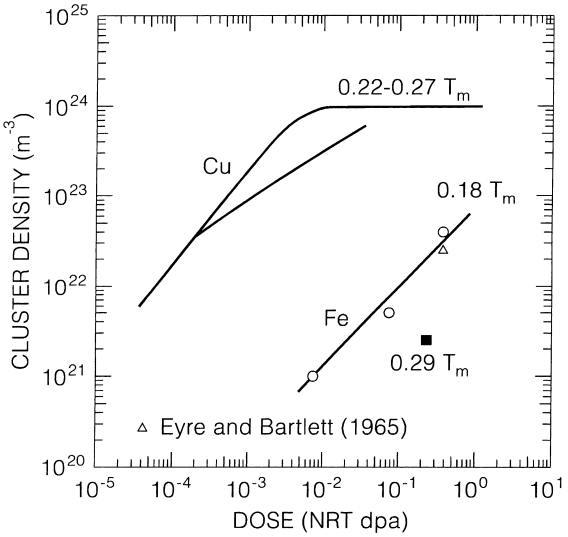

Fig. 2. Dose dependence of cluster density in pure iron irradiated at 320 K. △: From Ref. [7].

3.3. Post-deformation microstructure

The post-deformation microstructure of the unirradiated pure iron tensile tested at 320 K is dominated by the formation of cells and cell walls; some areas containing deformation bands were also observed. Since the observed microstructure is that existing at the end of the deformation process (i.e. after the fracture), it is impossible to determine how the microstructure has evolved during the tensile test. However, the observations lead to an overall impression that the plastic

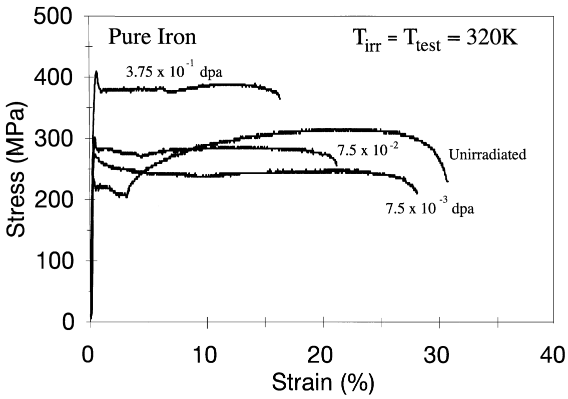

Fig. 3. Stress–strain curves for pure iron irradiated and tensile-tested at 320 K.

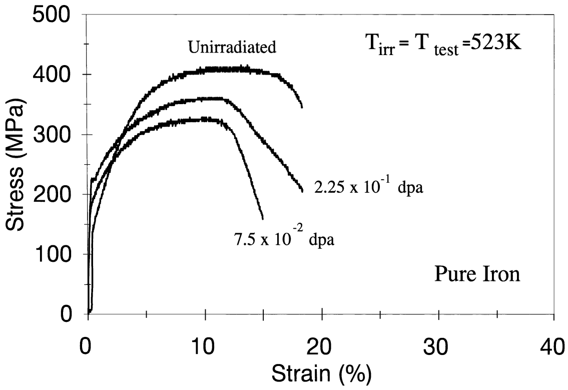

Fig. 4. Stress–strain curves for pure iron irradiated and tensile-tested at 523 K.

deformation in the unirradiated pure iron has occurred in a relatively homogeneous fashion.

The deformation behaviour of specimens irradiated to the lowest dose level of 7.5×10^{-3} dpa (NRT) is rather similar to that of the unirradiated ones, except the absence of well-defined cells. At this dose level, there is no indication of the formation of 'cleared' channels. However, already at the dose level of 7.5×10^{-2} dpa (NRT), the deformed microstructure in iron irradiated at 320 K is found to be noticeably different from that in the unirradiated pure iron. The irradiation-induced microstructure even at this low dose level seems to be sufficient to prevent the formation of cells and cell walls. Instead, the microstructure appears to be dominated by the high density of homogeneously distributed dislocations and deformation bands. Some of these deformation bands have the appearance of cleared channels.

At the dose level of 3.75×10^{-1} dpa, the deformed microstructure is completely different from that of the unirradiated and deformed pure iron at 320 K. At this dose level, the plastic deformation seems to be concentrated mainly in the cleared channels (Fig. 5a). Generally, there is no indication of dislocation generation and interactions in the volumes between the cleared channels. There are clear indications, however, of dislocation activities and interactions within the cleared channels. It is rather significant to note that the formation of cleared channels at this dose level becomes so extensive that the cleared channels begin to interact with each other. This could be very important from the point of view of crack nucleation and fracture of the material.

The SEM examinations of fracture surfaces of pure iron specimens irradiated at 320 K showed that the fracture behaviour changes with increasing dose levels. The specimen irradiated to a dose level of 7.5×10^{-3} dpa (NRT) fractures in a ductile manner preceded by a substantial amount of plastic deformation. The fracture behaviour changes significantly already at the dose level of 7.5×10^{-2} dpa (NRT) where the final failure seems to occur by a combination of ductile and brittle fracture. Fig. 5(b) shows the fracture surface of the pure iron specimen irradiated to the highest dose level of 3.75×10^{-1} dpa demonstrating intergranular type of brittle fracture which results.

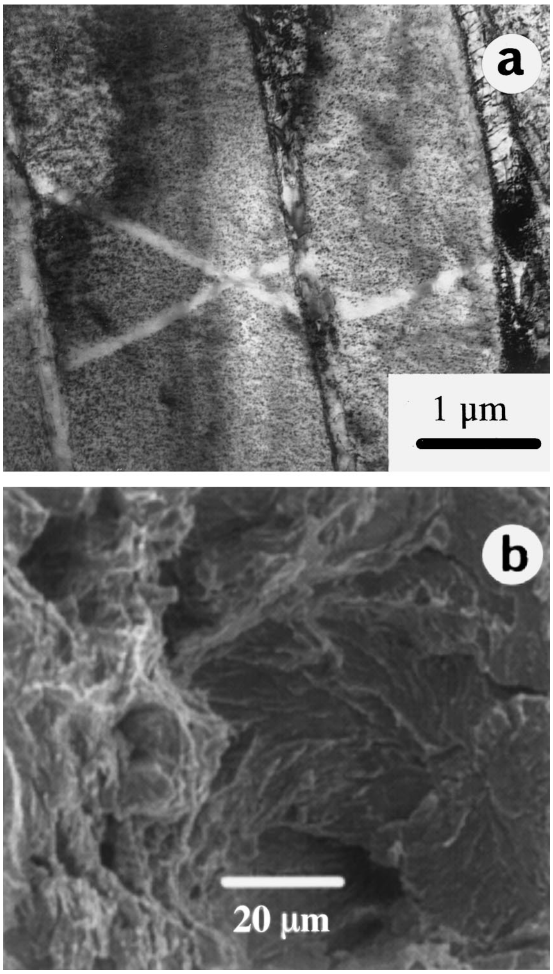

Fig. 5. Pure iron, irradiated at 320 K to 3.75×10^{-1} dpa (NRT) and tensile tested at 320 K: (a) TEM of post-deformation microstructure, (b) SEM of fracture surface.

4. Discussion

The dose dependence of cluster density measured in pure iron irradiated at 320 K (Fig. 4) clearly demonstrates that the damage accumulation in pure iron is enormously less efficient than in pure copper irradiated under similar conditions. Similar observations have been reported for pure Mo and Mo-alloys [8,9]. Possible reasons for this difference between fcc and bcc metals have been discussed in detail by Singh and Evans [8] and will not be repeated here. It is important, however, to mention the results of two different comparative studies of intracascade clustering [10] and cluster morphology and stability [11] using molecular dynamic (MD) simulations. One of the main conclusions of the work reported by Phythian et al. [10] was that both the size and number of SIA clusters formed in a cascade (at a given damage energy) are smaller in iron than that in copper. Furthermore, the recent study of Osetsky et al. [11] has demonstrated that the SIA clusters in iron are not stable in the sessile configuration, whereas they are stable in copper. In other words, practically all SIA clusters produced in cascades in bcc iron are likely to be glissile. Consequently, a number of SIA clusters produced will annihilate at all available sinks and a number of them will coalesce via glide [8]. Both of these factors would reduce the cluster density in iron.

An important implication of the possibility that practically all SIA clusters produced in iron are glissile is that the decoration of the grown-in dislocations by small SIA clusters [12] would be very efficient. This may make the "source hardening" [13] to be the dominant mechanism controlling the yielding behaviour of bcc iron.

As regards the effect of irradiation on the deformation behaviour of iron, the following features need to be considered:

(a) The increase in the upper yield stress with increasing dose level;
(b) The increase in the magnitude of yield drop with increasing dose level;
(c) The loss of work hardening due to irradiation;
(d) The loss of ductility and its apparent correlation with the efficiency of cleared channel formation and the lack of dislocation generation in the volume between the channels.

As discussed in great detail in Refs. [12,13], the items (a) and (b) could be understood in terms of dislocation decoration and the stress necessary to activate the grown-in dislocations as dislocation sources. The reason for the item (c) is not quite clear, however, and it may be related to the problem of homogeneous deformation in the matrix and the localized and extensive deformation in the cleared channels. The dose dependence of item (d) would suggest that the decoration of dislocations with gliding SIA clusters becomes more effective with increasing dose. As a result, dislocations could be generated only at the points of singularities with a high stress concentration factor. Once these sources are activated they lead to the formation of cleared channels and most of the plastic deformation occurs in a very localized fashion in these channels. Meeting of these channels at grain boundaries, surfaces or other channels may lead to crack nucleation at these sites and may be responsible for causing brittle fracture. SEM observations of surface cracks localized at grain boundaries in pure iron which had been irradiated with 590 MeV protons has been reported [14]. This may explain the experimental observation that the concentration of cleared channels increases with dose and that the fracture mode changes from ductile to brittle with increasing dose.

5. Conclusions

On the basis of the experimental results presented here, the following conclusions regarding the effect of neutron irradiation on damage accumulation and deformation behaviour of pure iron can be reached:

(a) The damage accumulation during irradiation at 320 K in the form of clusters/loops increases with dose in the range 7.5×10^{-3}–3.75×10^{-1} dpa (NRT). However, the density of clusters/loops is found to be substantially lower than that in pure copper irradiated under similar conditions.

(b) It is suggested that the low cluster density in pure iron may result due to (i) a lower efficiency (than in copper) of SIA cluster generation in the cascades and (ii) the configurational instability of sessile SIA clusters/loops. The latter implies that practically all SIA clusters produced in iron are glissile.

(c) The gliding clusters may not only cause the coalescence of clusters and decrease in the cluster density but may also lead to decoration of dislocations by small clusters/loops.

(d) The enhanced dislocation decoration may, according to the source hardening mechanism, be responsible for the increase in the upper yield stress, the increase in the magnitude of the yield drop and the increase in the density of cleared channels with the increasing dose level.

(e) The decrease in the observed ductility with increasing dose appears to be related to the formation of the cleared channels and the localized deformation in these cleared channels.

Acknowledgements

This work was partly funded by the European Fusion Technology Programme.

References

[1] R.L. Klueh, K. Ehrlich and F. Abe, J. Nucl. Mater. 191–194 (1992) 116.
[2] K. Ehrlich, K. Anderko, J. Nucl. Mater. 171 (1990) 139.
[3] D.S. Gelles, J. Nucl. Mater. 212–215 (1994) 714.
[4] D.S. Gelles, J. Nucl. Mater. 233–237 (1996) 293.
[5] J.M. Vitek, W.R. Corwin, R.L. Klueh, J.R. Hawthorne, J. Nucl. Mater. 141–143 (1986) 948.
[6] V.S. Khabarov, A.M. Dvoriashin, S.I. Porollo, J. Nucl. Mater. 233–237 (1996) 236.
[7] B.L. Eyre, A.F. Bartlett, Philos. Mag. 12 (1965) 261.
[8] B.N. Singh, J.H. Evans, J. Nucl. Mater. 226 (1995) 277.
[9] B.N. Singh, J.H. Evans, A. Horsewell, P. Toft, G.V. Müller, J. Nucl. Mater. 258–263 (1998) 865.
[10] W.J. Phythian, R.E. Stoller, A.J.E. Foreman, A.F. Calder, D.J. Bacon, J. Nucl. Mater. 223 (1995) 245.
[11] Yu. N. Osetsky, V. Priego, A. Serra, B.N. Singh, S.I. Golubov, Philos. Mag. (1999), in press.
[12] H. Trinkaus, B.N. Singh, A.J.E. Foreman, J. Nucl. Mater. 249 (1997) 91.
[13] B.N. Singh, A.J.E. Foreman, H. Trinkaus, J. Nucl. Mater. 249 (1997) 103.
[14] Y. Chen, P. Spätig, M. Victoria, J. Nucl. Mater., these proceedings.