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THE MICROSTRUCTURE OF "TRIPLE-BEAM" ION IRRADIATED Fe and Fe-Cr ALLOYS*

L. L. Horton,[1] J. Bentley,[2] and W. A. Jesser[1]

[1]Department of Material Science, University of Virginia, Charlottesville, VA 22901
[2]REMAG, Oak Ridge National Laboratory, Oak Ridge, TN 37830

MASTER

The development of defect structures in Fe—10% Cr, "triple-beam" (He$^+$, D_2^+ and 4 MeV Fe^{++}) ion irradiated to 10 dpa at temperatures from 725 to 950 K has been studied. Limited cavity formation was observed. Peak swelling of ~0.02% occurred at irradiation temperatures of ~850 K. Similar irradiations at this temperature led to ~0.22% swelling in iron, while no cavities were observed in Fe—5% Cr. Calculations have shown that cavities formed at 875 K and above are more bubble-like than those at lower temperatures. Dislocation structures varied from interstitial dislocation loops with $\underline{b} = a\langle 100\rangle$ at low temperatures to a coarse dislocation network at 850 K. Large dislocation loops had a convoluted shape which suggested that loop growth had occurred preferentially in <110> directions. The results are discussed and related to current swelling suppression mechanisms for ferritic steels.

1. INTRODUCTION

The potential of ferritic steels for application as first-wall and blanket structural materials for proposed fusion reactors is currently being evaluated. Detailed analyses of the defect structures which result from fusion-environment irradiation is an important aspect of this evaluation. It is particularly difficult to evaluate radiation damage that will occur in a fusion environment as no prototype reactors currently exist. One technique commonly used to study the microstructural development of defect structures utilizes transmission electron microscopy (TEM) examinations of specimens bombarded by energetic ions. In the "triple-beam" ion irradiation procedure developed at ORNL, energetic heavy ions, helium ions and deuterium ions simultaneously bombard the specimen [1–3]. The helium and deuterium ions are injected so as to come to rest within the damaged region created by the heavy ion bombardment and simulate transmutation produced helium and hydrogen.

While the "triple-beam" and other ion irradiation techniques, as well as a variety of neutron irradiation techniques, have been used extensively in producing specimens for TEM characterization of the defect structures in many candidate structural materials, relatively little data is available concerning the defect structures in ferritic materials. The lack of available microstructural data may be partially explained by the relatively recent interest in ferritics for fusion applications and by the difficulty of TEM examination of ferromagnetic materials. These difficulties are further enhanced by the usually complex microstructures of unirradiated ferritic steels.

The purpose of this investigation is to study the development of defect structures in "triple-beam" ion-irradiated pure iron and simple iron-chromium alloys using TEM. These materials were selected because of their importance as the basis of the more complex steels. Since the chromium content of many of the ferritic stainless steels being considered for fusion applications is ~10%, emphasis has been placed on the analysis of the defect structures in Fe—10% Cr. These results should provide a foundation for expanded analyses of radiation damage in ferritic steels. In this paper, the effects of irradiation temperature and chromium content on the damage microstructures are reported.

2. EXPERIMENTAL PROCEDURE

2.1 Specimen Material

The Fe—5% Cr and Fe—10% Cr alloys used in this study were fabricated at ORNL from MARZ grade iron (99.99+%) and IOCHROME chromium (99.996 %) obtained from Materials Research Corporation. A full description of the alloy preparation procedure is given elsewhere [4]. The iron underwent the same preparation procedure as the alloys. Rods (3-mm-dia) of each material were annealed in flowing dry hydrogen for 24 h at 1400 K, furnace cooled to 1000 K, and held at this temperature for 2 h, followed by furnace cooling. Interstitial impurity concentrations for each material are given in Table 1. Disk specimens were cut from the rods and prepared for ion bombardment [4].

Table I. Interstitial impurity concentration in specimen materials (wt ppm)

Alloy	C	H	N	O
Fe	34	2	<1	6
Fe—5% Cr	24	8	17	<1
Fe—10% Cr	34	2	4	6

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2.2 Triple-Beam Irradiation

The specimens were bombarded with He^+, D_2^+, and 4 MeV Fe^{++} ions in the dual Van de Graaff accelerator facility at ORNL [1–3]. The energy of the He^+ and D_2^+ beam was ramped sinusoidally at 2.5×10^{-2} Hz between 0.2 to 0.4 MeV. A discussion of the damage profiles and implanted helium and deuterium profiles for these irradiation conditions can be found elsewhere in these proceedings [5]. The specimens were irradiated to ~10 dpa with 100 at. ppm He and 410 at. ppm D. These are the He/dpa and H(D)/dpa concentration ratios expected for a ferritic steel first wall [6,7]. The nominal irradiation temperatures were 725, 775, 800, 850, 900, and 950 K. Actual irradiation temperatures, measured with an infrared pyrometer, for the individual specimens examined in this investigation are given in Table 2. The displacements per atom (dpa) indicated in Table 2 are calculated from the ion beam flux measured for each specimen during the irradiation.

2.3 TEM Specimen Preparation

The first step in preparing specimens for TEM was the controlled removal or sectioning of the damaged region to a predetermined depth of 0.9 µm, followed by back-thinning. The sectioning depth was selected based on the damage-depth profile for iron irradiated at 850 K [5]. The specimens were sectioned electrolytically using the vertical jetting apparatus developed by Lee [8]. Back-thinning was performed in a Tenupol electropolishing apparatus. The specimens were examined in a JEM 120C with a special objective lens pole-piece for the observation of magnetic materials [9].

3. RESULTS

Ion bombardment of Fe–10% Cr resulted in the development of both dislocation and cavity microstructures. No radiation-induced precipitation was observed. The series of micrographs in Figs. 1 and 2 summarize these defect structures. Quantitative data measured for these microstructures are shown in Table 2.

A comparison of the dislocation densities in Table 2 shows a general coarsening of the structure with increasing temperature. At irradiation temperatures between 723 and 803 K, the dislocation structure consisted primarily of loops. As indicated in Table 2, except for the specimen irradiated at 765 K, the loop diameter, d_L, increased and the loop concentration, C_L, decreased with increasing temperature. The smaller than expected loop diameter found at 765 K is probably related to the lower damage in this specimen as compared to the other specimens. At all temperatures, loops were observed to intersect the foil surfaces without gliding out of the foil. For beam directions near <001>, the majority of the loops were observed to be near edge and on {100} planes. Complete analyses of the geometry and nature of similar loops in iron triple-beam ion-irradiated at 850 K [5] and in Fe–10% Cr irradiated at 850 K to doses <10 dpa [4] have shown that the loops are interstitial, near-edge, with b = a<100>. Loops that were not observed to be on {100} are probably interstitial loops with b = a/2<111>, as a few loops with these Burgers vector were found in the complete analyses.

At the low temperatures, the loops varied in shape from round to rectilinear, with the sides aligned in <100> directions. For irradiation temperatures above 765 K, many of the dislocation loops had an irregular convoluted shape. One such loop with b = a[001] is shown in Fig. 3, a high magnification micrograph of the specimen irradiated at 803 K. This micrograph was taken with multiple-beam diffracting conditions at the (012) pole in order to view the a[010] loops and on-edge profiles of a[100] loops as well as the full perimeter profile of the a[001]

Table 2. Irradiation parameters and quantitative experimental data[a]

Alloy	T_I (K)	T_N (K)	dpa	Λ (m/m^3)	C_L (m^{-3})	d_L (nm)	C_c (m^{-3})	$\bar{d}_c$ (nm)	CVF (%)
Fe–10% Cr	723	725	10.7	3.9×10^{14}	5.7×10^{21}	18			
Fe–10% Cr	729	725	10.9	3.6×10^{14}	6.1×10^{21}	18			
Fe–10% Cr	765	775	8.3	4.2×10^{14}	7.[illegible]	15			
Fe–10% Cr	781	775	9.0	2.3×10^{14}	2.5×10^{21}	24			
Fe–10% Cr	785	800	9.9	2.2×10^{14}	1.4×10^{21}	45	2.4×10^{20}	7	4.3×10^{-3}
Fe–10% Cr	803	800	10.4	1.1×10^{14}	3.2×10^{20}	60			
Fe–10% Cr	849	850	10.1	1.1×10^{14}			2.6×10^{19}	23	1.7×10^{-2}
Fe–10% Cr	855	850	9.6	5.7×10^{13}			3.2×10^{19}	21	1.6×10^{-2}
Fe–10% Cr	875	900	9.5	2.4×10^{12}			9.5×10^{19}	5	6.2×10^{-4}
Fe–10% Cr	954	950	9.8	3.0×10^{12}			6.2×10^{19}	5	4.1×10^{-4}
Fe–10% Cr	971	900	9.5	2.2×10^{12}			9.0×10^{19}	5	5.9×10^{-4}
Fe	850	850	10.6	5×10^{13}			1.9×10^{21}	13	2.2×10^{-1}
Fe–5% Cr	852	850	10.6	1.3×10^{14}					

[a] Notation: T_I, actual irradiation temperature; T_N, nominal irradiation temperature; Λ dislocation density; C_L, loop concentration; d_L, loop diameter; C_c, cavity concentration; $\bar{d}_c$ volume average cavity diameter; and CVF, cavity volume fraction or swelling.

$T_I = 723$ K; $g = \langle 0\bar{1}1\rangle$; $z \sim [011]$ $T_I = 765$ K; $g = \langle 0\bar{1}1\rangle$; $z \sim [011]$ $T_I = 785$ K; $g = \langle 0\bar{1}1\rangle$; $z \sim [0\bar{1}1]$

$T_I = 803$ K; $g = \langle 1\bar{1}0\rangle$; $z \sim [115]$ $T_I = 849$ K; $g = \langle 0\bar{1}1\rangle$; $z \sim [133]$ $T_I = 875$ K; $g = \langle 0\bar{1}1\rangle$; $z \sim [011]$

Fig. 1. Dislocation microstructures observed in Fe—10% Cr. Arrows denote direction of the diffracting vector, g. Length of arrows equals 200 nm. Beam directions, z, and irradition temperatures as shown.

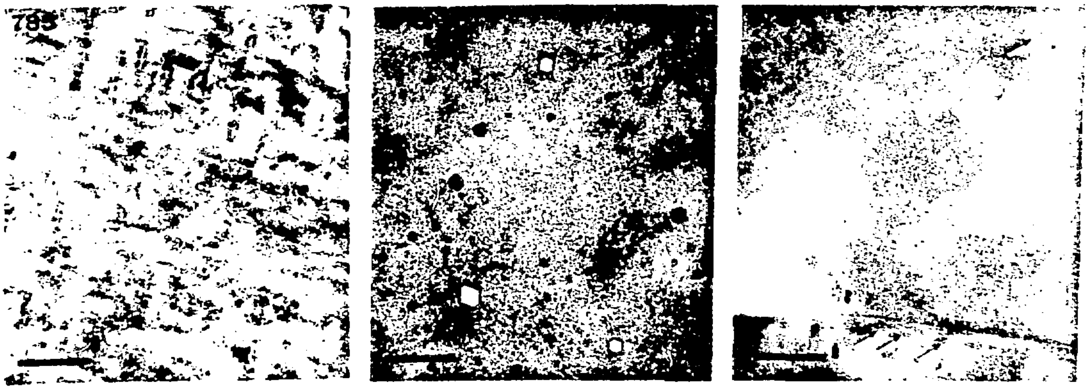

Fig. 2. Cavity microstructures observed in Fe—10% Cr. Scale markers equal 100 nm.

Fig. 3. Convoluted interstitial dislocation loop with $\underline{b} = a[001]$. $T_I = 803$ K, z ~ [012].

loops. The loops appear to have grown preferentially in ⟨110⟩ directions.

At irradiation temperatures of ~850 K, a coarse distribution of network segments was observed. At higher irradiation temperatures (>875 K), the dislocation structure appeared to be little changed from that existing in the unirradiated specimens.

Cavities were observed in specimens irradiated at temperatures of 785 K and above, except at 803 K. Apparently at 780–800 K, 10 dpa is near the threshold damage level for visible cavity formation. Therefore, a higher dose experiment is required to determine whether there are actually two swelling peaks, one near 780 K and another at ~850 K with lower swelling at ~800 K. The larger cavities had a definite crystallographic shape, as can be seen in Fig. 2. The cavity diameter was measured from a circular projection with the same area as the observed crystallographic shape.

The maximum cavity volume fraction (CVF) or swelling of 0.02% occurred for irradiation temperatures of ~ 850 K. For specimens irradiated at temperatures above and below 850 K, a higher concentration of smaller cavities than those found in the specimens irradiated at ~850 K was observed. However, for irradiation temperatures above 850 K, the cavity distribution was less homogeneous than at the lower temperatures.

As illustrated in Fig. 4, a zone denuded of cavities was observed near some grain boundaries. While the grain boundaries were also denuded of cavities for the lower temperatures, cavities were found on the boundaries in specimens irradiated at ~950 K. At this temperature, as shown in Fig. 5, cavities were also observed at the intersection of grain boundaries and chromium-rich precipitates. For irradiation temperatures of 785 and 850 K, cavities were observed along dislocations.

A comparison of the damage microstructure observed in iron, Fe–5% Cr, and Fe–10% Cr irradiated at 850 K is shown in Table 2. The swelling was an order of magnitude higher in iron (0.22%) than in Fe–10% Cr (~0.02%). No cavity formation was observed in the Fe–5% Cr specimen.

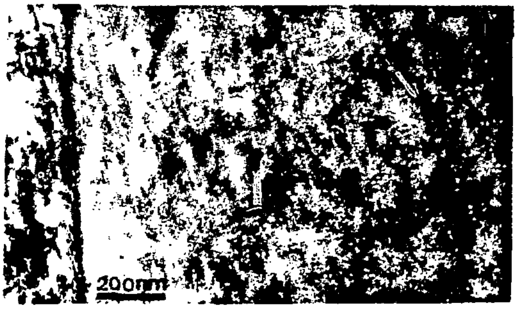

Fig. 4. A cavity denuded zone typical of those found at grain boundaries. $T_I = 785$ K.

Fig. 5. Cavities observed along grain boundaries and at the intersections of grain boundaries and Cr-rich precipitates. $T_I \sim 950$ K.

4. DISCUSSION

Several mechanisms [10–12] have been proposed for the low swelling behavior observed in ferritic materials. In the rate theory model presented by Hayns and Williams [10], incorporation of point defect trapping by solute atoms yielded swelling characteristics qualitatively similar to those observed in 1 MeV electron irradiated FV 607 ferritic steel. The mechanisms proposed by Little [11] considered the effects of the interactions of the interstitial and substitutional solutes with both point defects and dislocations. Little proposed that the formation of a solute cloud around a dislocation can lead to enhanced vacancy-interstitial recombination by reducing the bias for interstitials and by inhibiting the dislocation climb rate. It was also suggested that swelling is further reduced through preferential vacancy trapping by both interstitial and substitutional solutes. Bullough et al. [12] have evaluated the role of a high preirradiation dislocation ($\underline{b} = a/2\langle 111\rangle$) density in the swelling suppression. This mechanism is not believed to apply to ferritic materials without a high initial dislocation density and is, therefore, not applicable to the current investigation.

The present observation of loops intersecting the foil surfaces could be the result of dislocation pinning by impurity atmospheres, as proposed in Little's mechanism. However, this observation could also be interpreted as an indication of the sessile nature of a<100> loops [13, 14]. Calculations using a high-density equation-of-state for helium [15] indicates only 10% of the implanted helium is required to form equilibrium bubbles at irradiation temperatures of 875 K and above. This calculation, together with the observation of a decrease in cavity diameter compared to the lower temperatures, correlates well with the two orders of magnitude lower dislocation density at the higher temperatures if the dislocations are assumed to be biased sinks for interstitials. At the lower temperatures of 785 and 850 K, the cavities are more void-like due to the presence of an excess of vacancies (bias-driven growth). At the higher temperatures, more interstitials are available for recombination with the vacancies, constraining the cavities to grow as bubbles (gas-driven growth). The bubble-like character of the cavities is also demonstrated by the observation of cavities on grain boundaries only at the high temperatures.

In considering swelling suppression solely due to point defect trapping at solute atoms, Hayns and Williams [10] have suggested that the usual reactor-accelerator shift in the peak swelling temperature would be minimized (~ 50K). For Fe–10% Cr neutron irradiated to 30 dpa, the peak swelling was observed at ~700 K [16], 150 K lower than the peak swelling temperature reported in this study. While this larger shift may be partially due to the higher concentration of helium in the ion irradiation [17], it further suggests that the swelling suppression is not entirely due to trapping at solute atoms.

In neutron irradiation experiments Little and Stow [16] have observed a relationship between chromium content and swelling similar to that reported here. They stated that the increase in the swelling in Fe–10% Cr as compared to that in Fe–5% Cr was related to α' precipitation and the corresponding nonequilibrium segregation of the chromium in Fe–10% Cr. No α' precipitates were observed in the current investigation, however, suggesting that the increase in swelling is possibly based on a different mechanism.

The observation of rectilinear a<100> interstitial dislocation loops is common in irradiated iron and ferritic steels [13,14,18]. Kiritani et al. [18] have observed flower-like a<100> loops in electron irradiated iron. Their explanation for this shape is based on accelerated arrival of interstitials at the corners of the rectilinear loops compared with the straight parts due to dilation of the strain field at the corners. Such a mechanism is believed to be responsible for the shape of the loops observed in the specimens in this investigation.

The development of cavity and dislocation structures at 850 K with increasing damage levels in the range 0.3 to 100 dpa is currently under investigation. When complete, these studies should add to the understanding of the mechanisms of radiation damage in ferritic materials.

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