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Dose dependence of defect accumulation in neutron irradiated copper and iron

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Abstract

In order to investigate the difference in defect accumulation between fcc Cu and bcc Fe, tensile specimens were neutron irradiated at ≃70 °C in the HFIR reactor at Oak Ridge National Laboratory to fluences in the range of 4.5×10^{20}–4.7×10^{24} n/m^2 ($E > 1$ MeV) corresponding to displacement dose levels in the range of about 0.0001–0.8 dpa. Irradiated specimens were characterized using positron annihilation spectroscopy, transmission electron microscopy and electrical conductivity measurements. A limited number of iron specimens were also tensile tested. At 0.0001 dpa, a low density of very small vacancy clusters (1–3 vacancies) were detected in iron, while bigger three-dimensional cavities were observed at higher doses. Both their density and average size increased with increasing dose level. In contrast, no such cavities were observed in copper. Irradiation led to an increase in yield stress and a decrease in the uniform elongation for iron.

1. Introduction

The problem of defect accumulation in the form of loops, tetrahedra and voids in irradiated metals has been studied both in fcc and bcc metals and alloys over a long period of time. In a review of the available experimental results Singh and Evans [1] concluded that there exist rather large differences in defect accumulation behaviour between fcc and bcc metals, particularly when irradiated under cascade damage conditions. One of the significant differences in the defect accumulation behaviour between fcc and bcc metals is found to be in the area of void nucleation. Transmission electron microscopy (TEM) results clearly demonstrate, for example, that void nucleation in the studied bcc metals occurs already at temperatures just above the recovery stage III (see Table 2 in [1] and Fig. 1 in [2]) whereas in fcc metals, voids have not been observed to nucleate much below the recovery stage V [1].

In a recent work, it has been shown that indeed there exists a significant difference in void nucleation between Fe and Cu neutron irradiated to a dose of 0.2–0.3 dpa [3]. In Cu, void nucleation took place only at temperatures above 200 °C (i.e. close to the recovery stage V) while in Fe, voids were formed even at ≃50 °C (i.e. close to the recovery stage III).

An interesting question arose regarding the lowest dose at which voids may begin to nucleate and as to how the void population may evolve as a function of dose in iron irradiated at around 50 °C. In the present work we have therefore investigated the effect of irradiation dose on the defect accumulation in iron at an irradiation temperature of 50–70 °C. For comparison, a parallel study on copper has also been carried out.

In the following section (Section 2) details of materials and experimental procedures used in the present investigations are described. The results of PAS, TEM investigation, electrical conductivity measurements and tensile tests are described in Section 3. The main

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conclusions, particularly related to the differences in the defect accumulation behaviour between bcc iron and fcc copper are summarised in Section 4.

2. Materials and experimental procedure

The specimens used for the investigations were tensile specimens, 17 mm long with a gauge length of 8 mm and gauge width and thickness of 1.5 and 0.25 mm, respectively. The materials were Cu of 99.999 wt% purity and Fe of 99.995 wt% purity.

After machining, the specimens were recrystallized by annealing in vacuum approaching 10^{-5} Pa, 1/2 h at 450 °C for the copper, and 600 °C for the iron. These temperatures were selected to give a minimum of ten grains across the thickness of the specimens in order to ensure that during tensile testing the specimens would display bulk, polycrystalline properties. Mean grain sizes were measured to be 33 μm for the iron and 18 μm for the copper.

For irradiation, the specimens were vacuum sealed in tightly fitting aluminium envelopes to prevent corrosion in the coolant water and to ensure minimum thermal gradients between the specimens and the flowing water.

Irradiations were conducted in the hydraulic tube facility of the High Flux Isotope Reactor (HFIR) at ORNL. The neutron fluxes at the capsule positions used in the present work are about 4×10^{18} n/m^2/s ($E > 1$ MeV). The temperature of the specimens during irradiation was estimated to be about 70 °C. Specimens were irradiated to fluences in the range from 4.5×10^{20} to 4.7×10^{24} n/m^2 ($E > 1$ MeV) equivalent to a displacement damage in a range from about 10^{-4}–0.8 displacements per atom (dpa NRT). Unfortunately, the Cu specimens were accidentally deformed during handling with the result that only TEM investigations and qualitative PAS measurements could be made for copper, while no electrical conductivity nor tensile tests could be carried out.

For each PAS measurement, two samples of approximately 5×3.5 mm^2 were cut from one tensile specimen. The Fe samples were cleaned by electropolishing and the Cu samples by chemical etching. A conventional positron lifetime spectrometer was used for the PAS measurements [4]. The recorded lifetime spectra each contained a total of $(4–10) \times 10^6$ counts. All specimens were measured several times to assure reproducibility. The measured lifetime spectra were analysed with the PATFIT programs [5].

TEM was performed using a combination of bright field and weak beam dark field imaging modes. The defect cluster density was determined from the slope of plots of the areal density vs. foil thickness. The foil thickness was measured from thickness fringes in weak beam dark field micrographs.

The electrical conductivity measurements were made on the tensile specimens and were carried out as described in [6].

Both unirradiated and irradiated Fe specimens were tensile tested at 70 °C in a vacuum of $<10^{-2}$ Pa at a strain rate of 1.2×10^{-3} s^{-1}.

3. Results

To illustrate directly the influence of dose on the obtained positron lifetime data, the spectra measured for Fe are shown in Fig. 1. Clearly, the long-lived parts of the spectra become more intense with increasing dose. This shows qualitatively that the density and/or size of three-dimensional cavities increase with dose. No PAS results are available for the iron specimens irradiated to the highest dose (0.72 dpa) because of the high activity level of these specimens.

In the quantitative analysis, the lifetime spectra for Fe could be resolved into three lifetime components for doses higher than 10^{-4} dpa. The result of this analysis is shown in Fig. 2. The two long lifetimes, τ_2 and τ_3 and their associated intensities I_2 and I_3 arise from positrons that are trapped in three-dimensional vacancy clusters. The trend of the lifetime variation is that both τ_2 and τ_3 increase with irradiation dose from a value of about 230 ps, equivalent to a cluster of just 2–3 vacancies [7] up to about 325 and 525 ps, equivalent to clusters of roughly 10 vacancies and more than 50 vacancies, respectively [4,7–9]. This association of the two resolved lifetimes

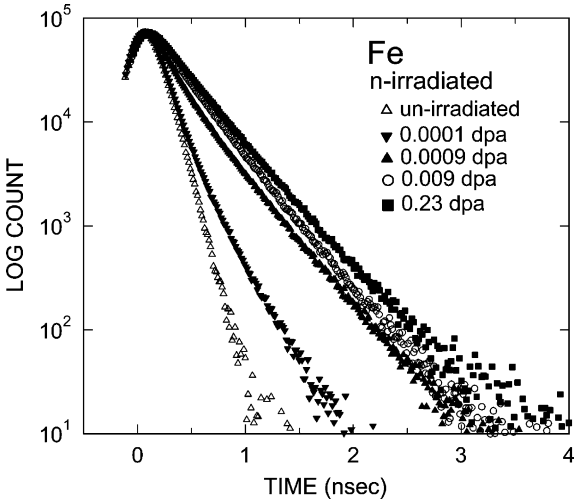

Fig. 1. Positron lifetime spectra for Fe irradiated to different doses. The spectra for the doses higher than 10^{-4} dpa show long-lived components that are due to three-dimensional vacancy clusters of different sizes. The specimen with the dose of 0.23 dpa was irradiated at ≃50 °C in DR3 at Risø.

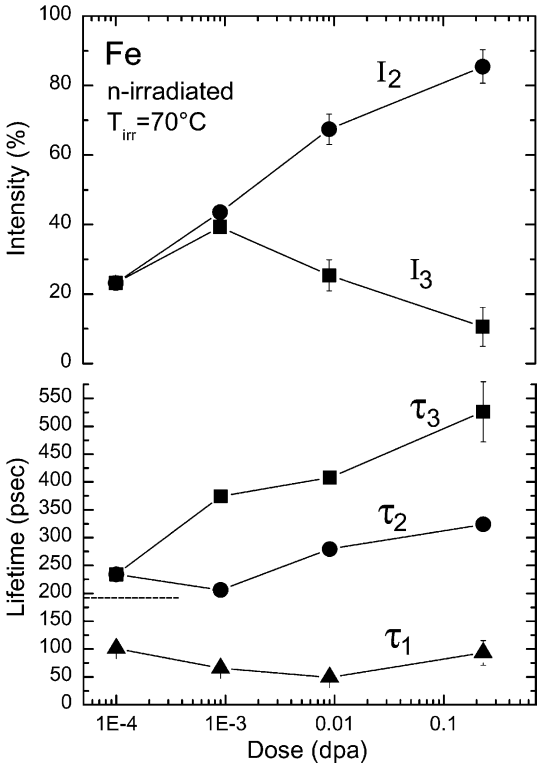

Fig. 2. Positron lifetimes and their intensities (derived by a conventional analysis of the spectra in Fig. 1) as functions of irradiation dose. The lifetimes due to vacancy clusters (τ_2 and τ_3) and their total intensity ($I_2 + I_3$) increase with increasing dose. This shows that both sizes and densities of three-dimensional vacancy clusters increase with dose. The dashed line indicates the lifetime of positrons in mono-vacancies.

with specific cluster sizes should only be considered as a two-bin representation of a wide size distribution of cavities rather than a measure of the presence of only two discrete cavity sizes.

In order to investigate this possibility further, the lifetime spectra for Fe were analysed on the assumption that they are composed of five components, four of which have fixed lifetimes: 200, 300, 400 and 500 ps, equivalent to three-dimensional vacancy clusters of sizes of about 0.35, 0.54, 0.73 and >1.0 nm in diameter, respectively. The outcome of the analysis is shown in Fig. 3, where the columns represent the intensities of the various lifetime components.

As discussed in for example Ref. [4] the intensities of the various lifetime components are measures of the densities of the different types of defects. The positron trapping rates κ_i into defects of type 'i' can be derived from the intensities and lifetimes (Fig. 3) using the so-called 'trapping model' [4]. The defect densities C_i can then be calculated from the trapping rates, using the relation:

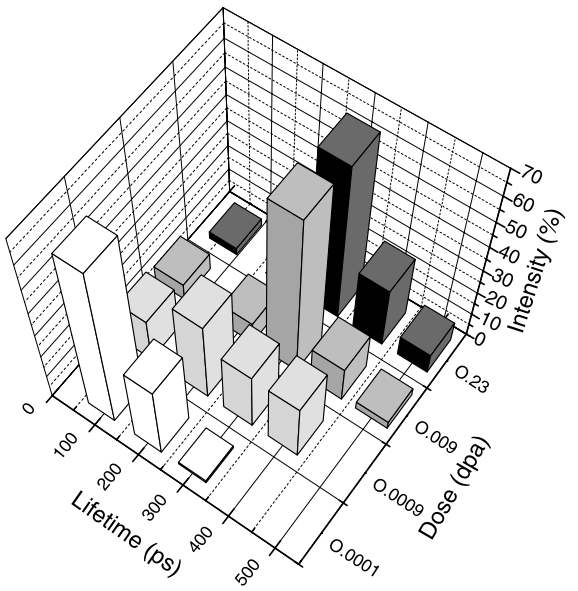

Fig. 3. Diagram showing the intensities of specific lifetime components (i.e. the lifetime distributions) in the spectra shown in Fig. 1. The columns for lifetimes $\geqslant 200$ ps show lifetime distributions for positrons trapped in vacancy clusters.

$$\kappa_i = \mu_i \times C_i, \tag{1}$$

where the so-called specific trapping rate, μ_i, depends on the type of the defect. For three-dimensional vacancy clusters, μ increases with cavity size [4]. For a given size, the uncertainty on the μ-value is roughly a factor of two [4].

The calculated size distributions for the three-dimensional vacancy clusters in Fe are shown in Fig. 4. The size (for a cluster of N_v vacancies) is given by an equivalent diameter, i.e. the diameter of a sphere with a volume equal to N_v vacancies. Clearly, it has been possible to derive an estimate of the cavity size distribution for different doses. With increasing dose, both the density and the average size increase. The total density of the three-dimensional vacancy clusters as a function of dose is shown in Fig. 5. The calculated swelling, S, is also plotted in Fig. 5. For doses lower than 0.01 dpa, the swelling increases roughly linearly with dose, with a swelling rate of (4 ± 2)%/dpa. At higher dose the swelling levels off.

All the positron lifetime spectra for Cu could be resolved into only two components, the longer lifetime, τ_2, being equal to (182 ± 2) ps for all doses. This lifetime can be associated with defects of the size of vacancies. Thus, the spectra for Cu showed no long-lived components that can be associated with three-dimensional vacancy clusters. The intensity of the defect component, I_2, varies with dose, but it is not possible to separate the contributions from irradiation and those due to handling dislocations in the specimens.

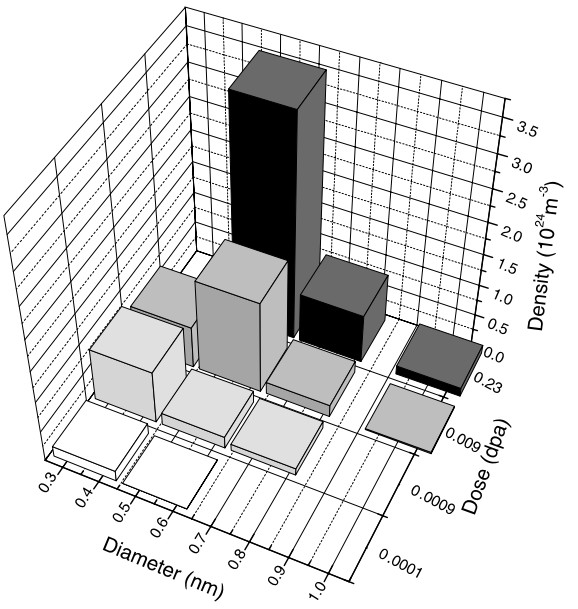

Fig. 4. Size distributions for the three-dimensional vacancy clusters in Fe at different doses derived from the PAS data in Fig. 3. The size (for a cluster of N_v vacancies) is given by an equivalent diameter, i.e. the diameter of a sphere with a volume equal to N_v vacancies.

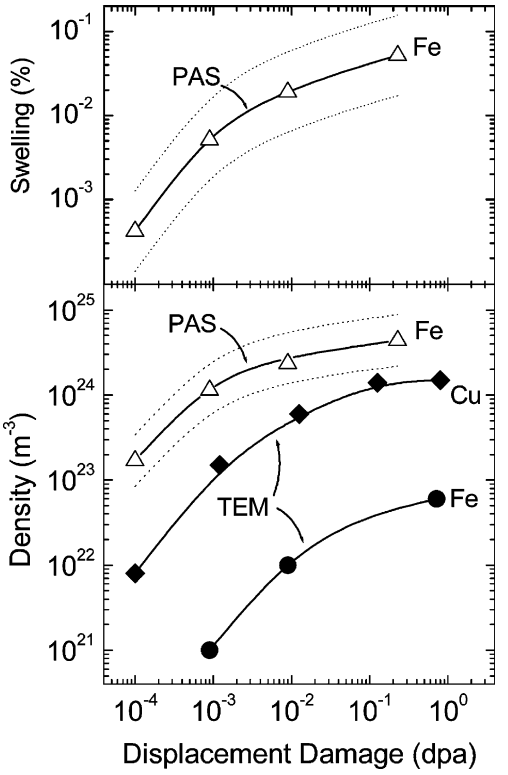

Fig. 5. Dose dependence of defect cluster density in neutron irradiated Cu and Fe. In Cu (◆) the clusters are mainly SFTs. In Fe the TEM results (●) are mainly densities of interstitial clusters, while the PAS results (△) are the total density of three-dimensional vacancy clusters obtained from Fig. 4. At the top of the figure, the dose dependence of the swelling in Fe is shown. The dotted curves indicate the estimated uncertainty bands for both density and swelling derived from PAS data.

The primary results from the TEM observation were as follows: There were no visible defect clusters in the lowest dose Fe specimen (10^{-4} dpa). The defect cluster size and density increased with increasing dose between 10^{-3} and 0.72 dpa (Fig. 5). These observations suggest that defect cluster formation in fission-neutron irradiated Fe occurs via point defect nucleation and growth, as opposed to in-cascade formation of visible (>1 nm diameter) defect clusters. Dislocation loop alignment ('rafts') was observed in iron specimens irradiated to 0.72 dpa as shown in Fig. 6. A detailed dislocation loop analysis is in progress. Small cavities (maximum diameter of 1.5 nm) with a density of the order of 10^{24} m^{-3} were observed in the high dose Fe specimen in agreement with the distributions (shown in Fig. 4) obtained from positron annihilation measurements.

The dominant microstructural feature in the irradiated Cu specimens at all doses was the formation of small defect clusters (about 90% of which could be resolved as stacking fault tetrahedra). These clusters were visible at all damage levels (10^{-4}–0.8 dpa), indicating that visible (>1 nm diameter) vacancy cluster formation occurs directly in displacement cascades in Cu. The mean size of the defect clusters was ≃2.5 nm at all doses. The defect cluster density increase was proportional to dose at low doses, and reached a constant (saturation) density of $\simeq 1.3 \times 10^{24}$ m^{-3} at 0.1–0.8 dpa (Fig. 5). Cavity formation was not observed in the irradiated copper specimens, in agreement with the PAS results and with previous studies that found a minimum temperature for cavity formation in irradiated copper of ≃200 °C [1,3]. The resolvable (diameter >5 nm) dislocation loop density in the irradiated copper specimens reached its maximum value at a relatively low damage level of ≃0.01 dpa. At higher doses, the loops interacted with one another to form a moderate density ($<10^{13}$/m^2) of network dislocations. The density of interstitial-type dislocation loops in the irradiated Cu specimens was at least two orders of magnitude smaller than that for the stacking fault tetrahedra at all doses.

The results of the electrical conductivity measurements are shown in Fig. 7. Irradiation induced defects as well as transmutation products [6] lead to the decrease in conductivity with increasing dose. The low temperature data for specimens irradiated in HFIR and in DR3 are in good agreement, while the conductivities for the 100 °C samples are clearly higher. This difference is associated with trapping of defects by impurity carbon

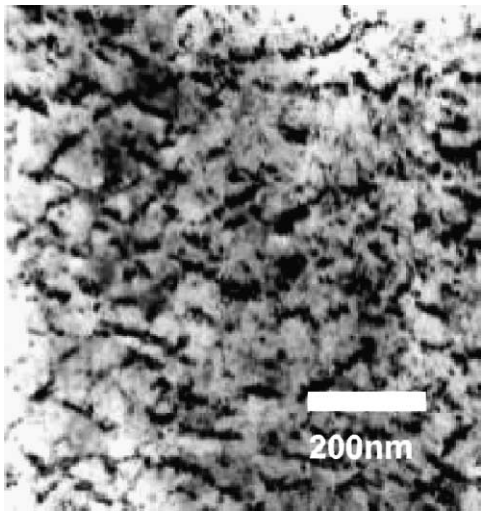

Fig. 6. Formation of rafts of loops in Fe irradiated to a dose of 0.72 dpa.

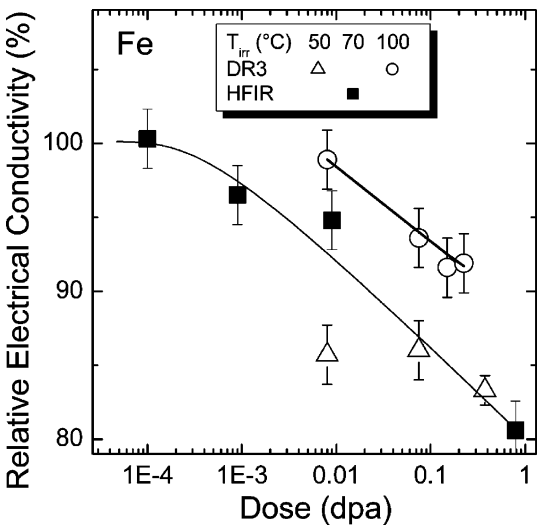

Fig. 7. Electrical conductivity, relative to that for un-irradiated Fe, as a function of dose for Fe irradiated at about 70 °C in HFIR (Oak Ridge) and 50 and 100 °C in DR3 (Risø).

atoms below the migration temperature for carbon (>70 °C).

The engineering stress–strain curves for the unirradiated and irradiated iron specimens are shown in Fig. 8. It can be clearly seen that the yield strength increases and the uniform elongation decreases with increasing dose level. It should be noted that the specimen irradiated to 0.72 dpa exhibits a strong yield drop and the loss of ability to work harden.

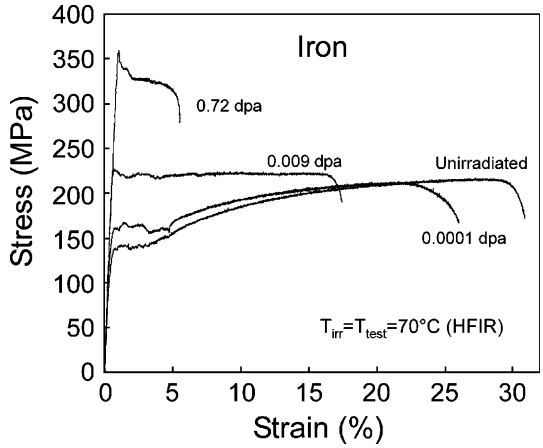

Fig. 8. Dose dependence of radiation hardening for a strain rate of 1.2×10^{-3} s^{-1}.

4. Summary and conclusion

The results clearly show the difference in defect accumulation between copper and iron during neutron irradiation in a wide dose range.

In Cu, no void nucleation is observed.

In Fe, three-dimensional cavities are observed at doses of 0.0009 dpa and above. Both the density and the average size increase with dose. At 10^{-4} dpa a low density of very small vacancy clusters (1–3 vacancies) are detected by PAS in iron. Voids become visible in TEM in the Fe specimen irradiated to 0.72 dpa with a density of the order of 10^{24} m^{-3} in the size range below 1.5 nm.

The data suggest that the swelling rate in Fe is constant ((4 ± 2)%/dpa) for doses below 0.01 dpa, but decreases at higher doses. It is interesting to note that this behaviour is similar to that observed in copper irradiated at 250 °C [10]. TEM investigations show that the density of SIA clusters both in Fe and Cu first increases with dose. At doses higher than $\simeq$0.01 dpa, the clusters begin to segregate and coalesce leading to formation of rafts of SIA clusters. The formation of the raft-like structure is significantly more efficient in bcc iron than in fcc copper.

The present work demonstrates that positron annihilation spectroscopy can provide valuable information about the microstructure of irradiated metals, in particular about the presence of small cavities that escape detection by TEM.

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References

[1] B.N. Singh, J.H. Evans, J. Nucl. Mater. 226 (1995) 277.
[2] S.I. Golubov, B.N. Singh, H. Trinkaus, J. Nucl. Mater. 276 (2000) 78.
[3] M. Eldrup, B.N. Singh, Mater. Sci. Forum 363–365 (2001) 79.
[4] M. Eldrup, B.N. Singh, J. Nucl. Mater. 251 (1997) 132; J. Nucl. Mater. 276 (2000) 269.
[5] P. Kirkegaard, N.J. Pedersen, M. Eldrup, Risø Report, Risø-M-2740, 1989.
[6] M. Eldrup, B.N. Singh, J. Nucl. Mater. 258–263 (1998) 1022.
[7] M. Puska, R.M. Nieminen, J. Phys. F: Metal Physics 13 (1983) 333.
[8] K.O. Jensen, R.M. Nieminen, Phys. Rev. B 36 (1987) 8219.
[9] M. Eldrup, Mater. Sci. Forum 105–110 (1992) 229.
[10] B.N. Singh, A.J.E. Foreman, Philos. Mag. A 66 (1992) 975.