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Fission neutron irradiation of copper containing implanted and transmutation produced helium

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High purity copper containing approximately 100 appm helium was produced in two ways In the first, helium was implanted by cyclotron at Harwell at 323 K In the second method, helium was produced as a transmutation product in 800 MeV proton irradiation at Los Alamos, also at 323 K The distributions of helium prior to fission neutron irradiation were determined by a combination of transmission electron microscopy (TEM) and positron annihilation techniques (PAT) These specimens, together with pure copper, were then irradiated with fission neutrons in a single capsule in fast flux test facility (FFTF) at $\sim 686 \pm 5$ K to a dose level of ~ 48 dpa (7.7×10^{26} nm^{-2}, $E > 0.1$ MeV) Investigation of the void and dislocation microstructures in the three specimens by TEM showed large differences between the specimens in void size and swelling The observed differences as well as the effect of the presence of other transmutation produced impurity atoms in the 800 MeV proton irradiated copper will be discussed

1. Introduction

It is well known that radiation damage at elevated temperatures can lead to, apart from void swelling, other interesting phenomenon such as void lattice formation (see ref. [1] for review). However, the problem of void lattice formation, particularly in fcc metals, is still not properly understood It is noteworthy that there exists practically no definite experimental evidence for the formation of void lattices in fcc metals under fast neutron irradiation. So far, there has been only one investigation in which the possible existence of void lattices in fcc aluminium irradiated with fast neutrons has been suggested [2]. It is not at all clear, for example, whether it is the high dose or high cavity density which is a prerequisite for the onset of the void lattice formation. In 1987 we reported the observation of void hyperlattice in high purity aluminium irradiated with fast neutrons [3]. In this experiment the lattice formation took place even when the void density was relatively low.

Thus, in order to investigate whether or not it is at all possible to obtain void lattices in fcc metals, a number of pure fcc metals were irradiated with fast neutrons in FFTF to a relatively high dose of ~ 48 dpa In order to enhance the possibility of void lattice formation, Cu specimens containing about 100 appm helium were also irradiated

There were two additional interests in carrying out these investigations. First, the role of irradiation in cavity coarsening is not fully understood [4] and it was hoped that the irradiation of helium implanted specimens would provide some interesting information. Secondly, very little is known about the effect, if any, of the transmutational impurities produced during irradiation on the swelling behaviour of materials. For this reason, Cu preirradiated with 800 MeV protons were further irradiated with fast neutrons in the FFTF.

In the following we first describe the materials, preirradiation treatments and irradiation conditions (section 2). The results of TEM investigations are given in section 3, and are discussed in section 4. Section 5 provides a list of conclusions emerging from the present work.

2. Experimental procedure

In the present investigation three types of specimens were used. All specimens were in the form of 3 mm diameter discs Thickness of the discs was ~ 0.1 mm. Type one specimens were of OFHC copper with an oxygen content of ~ 10 appm. Prior to irradiation in FFTF, these specimens were annealed at 823 K for 2 h in a vacuum of $\sim 10^{-6}$ Torr. The second type of specimens were of 99.999% Cu implanted with 100 appm of helium at ~ 323 K by cyclotron (VEC) at Harwell. The third type of specimens were also of 99.999% Cu and contained ~ 90 appm of helium, helium was produced as a transmutation product in 800 MeV proton irradiation at ~ 323 K at Los Alamos in the Meson Physics Facility (LAMPF) The oxygen contents of the last two types of Cu are not known The helium concentration of 90 appm was achieved by a proton fluence of 1.78×10^{24} p^+/m^2. A proton fluence of 3.94×10^{24} p^+/m^2 produces 201.5 appm of helium [5].

Some of the specimens implanted with 100 appm helium and irradiated at LAMPF were investigated by TEM as well as positron annihilation techniques prior to irradiation in the FFTF.

The TEM discs were separated by 0.001 in. thick molybdenum spacers and were encapsulated in a sealed 20% cold worked 316 stainless steel canister (1.12 in. long, 0.144 in. o.d. with wall thickness 0.007 in.), under a room temperature pressure of 0.4 atm helium. This canister was placed in contact with the reactor sodium coolant in a weeper basket at level 2C-2 of the materials open test assembly (MOTA) and irradiated in FFTF in Richland, Washington, USA, operating at 291 MW thermal energy for 341.8 effective full power days. The temperature of the coolant in contact with the canister was 678 ± 5 K during full power operation, the specimens are calculated to have experienced approximately 8 K higher temperature (686 ± 5 K) due to gamma heating within the packet. Somewhat lower temperatures were experienced during the ascent to power stages of the various segments of the irradiation. The specimens were exposed to 7.7×10^{26} n/m^2 ($E > 0.1$ MeV) which produced ~48 dpa in pure copper.

Irradiated specimens were electropolished in a solution of 25% H_3PO_4, 25% ethylene glycol and 50% water at 11 V for about 15 s. Examinations were performed in a JEOL 2000FX electron microscope at Risø National Laboratory, Denmark.

3. Results

The defect microstructures of the specimens in the as-implanted and as-irradiated with 800 MeV protons (i.e. pre-MOTA irradiation) states are shown in fig. 1. The total cluster density in the 800 MeV proton irradiated specimen was found to be 1.1×10^{24} m^{-3} [6] of which 2.7×10^{23} m^{-3} were the stacking fault tetrahedra (SFT). Very few loops were observed in this specimen and there was no indication of either network dislocations or helium bubbles. The cluster density in the 100 appm implanted (at Harwell) specimen was found to be $\sim 6 \times 10^{22}$ m^{-3} and practically no SFTs were observed. A close examination of this specimen showed, on the other hand, the presence of small (1–2 nm) bubbles although in a relatively low concentration ($\leq 10^{21}$ m^{-3}).

The positron lifetime measurements carried out on the 100 appm implanted Cu specimens showed the presence of submicroscopic clusters of vacancy type. The helium content of the clusters (i.e. helium bubbles) was estimated to be 0.3–1 times the number of vacancies. The cluster density was estimated to be roughly 3×10^{23} m^{-3} for an average size of ~1 nm diameter.

The void and void plus dislocation microstructures of copper and copper containing 100 and 90 appm helium after FFTF/MOTA irradiation are shown in fig. 2. The void distribution in all three specimens was reasonably uniform. It is interesting, though somewhat surprising, to note that even after a displacement dose of ~48 dpa, the dislocation density in the pure copper specimen remained very low and that there was no indication of irradiation-induced dislocation network formation. In copper specimens containing 100 and 90 appm helium, on the other hand, the beginning of dislocation network formation was evident. Furthermore, unlike in pure copper, 10–20% of the clusters in these specimens were loops of sizes up to 20 nm. Fig. 3 shows the cluster densities, as seen in the TEM investigations, for the specimens that were implanted as well as for those that were irradiated with 800 MeV protons. The size distributions of voids observed in the three different conditions (i.e. no helium, helium implanted and helium produced via nuclear reactions) are shown in fig. 4. The void size, density, swelling and the cluster density values determined from TEM inves-

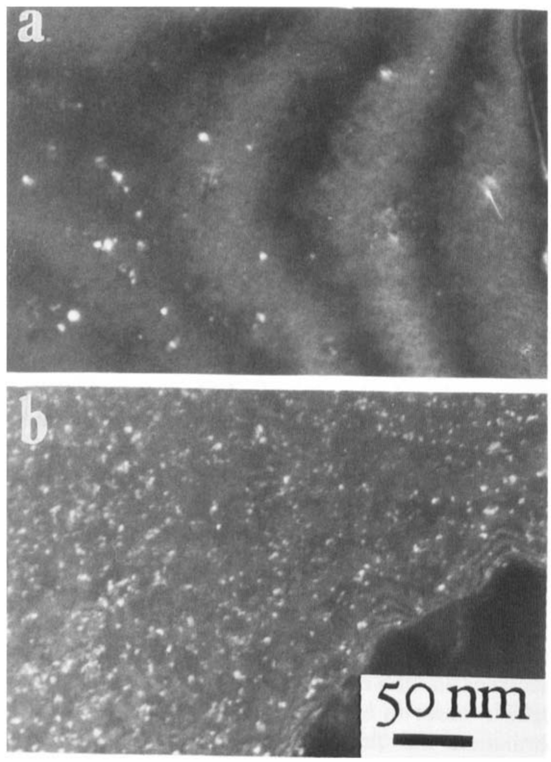

Fig. 1. Defect microstructures of copper specimens (a) in the as-implanted (with 100 appm helium) and (b) as-irradiated (with 800 MeV protons) conditions. The irradiation with 800 MeV protons is expected to have generated about 90 appm helium.

tigations are quoted in table 1. The TEM based swelling rate in Cu (without helium) in the present experiment (table 1) is not very different from the one reported in ref. [7] (based also on the TEM results) but is noticeably lower than the swelling rate obtained from the density change measurements [8].

It should be pointed out that all three specimens were investigated for the existence of void lattices. However, TEM examinations provided no evidence of void ordering in any of the three specimens.

4. Discussion

The absence of void lattice formation in the present experiment may have resulted due to the high irradia-

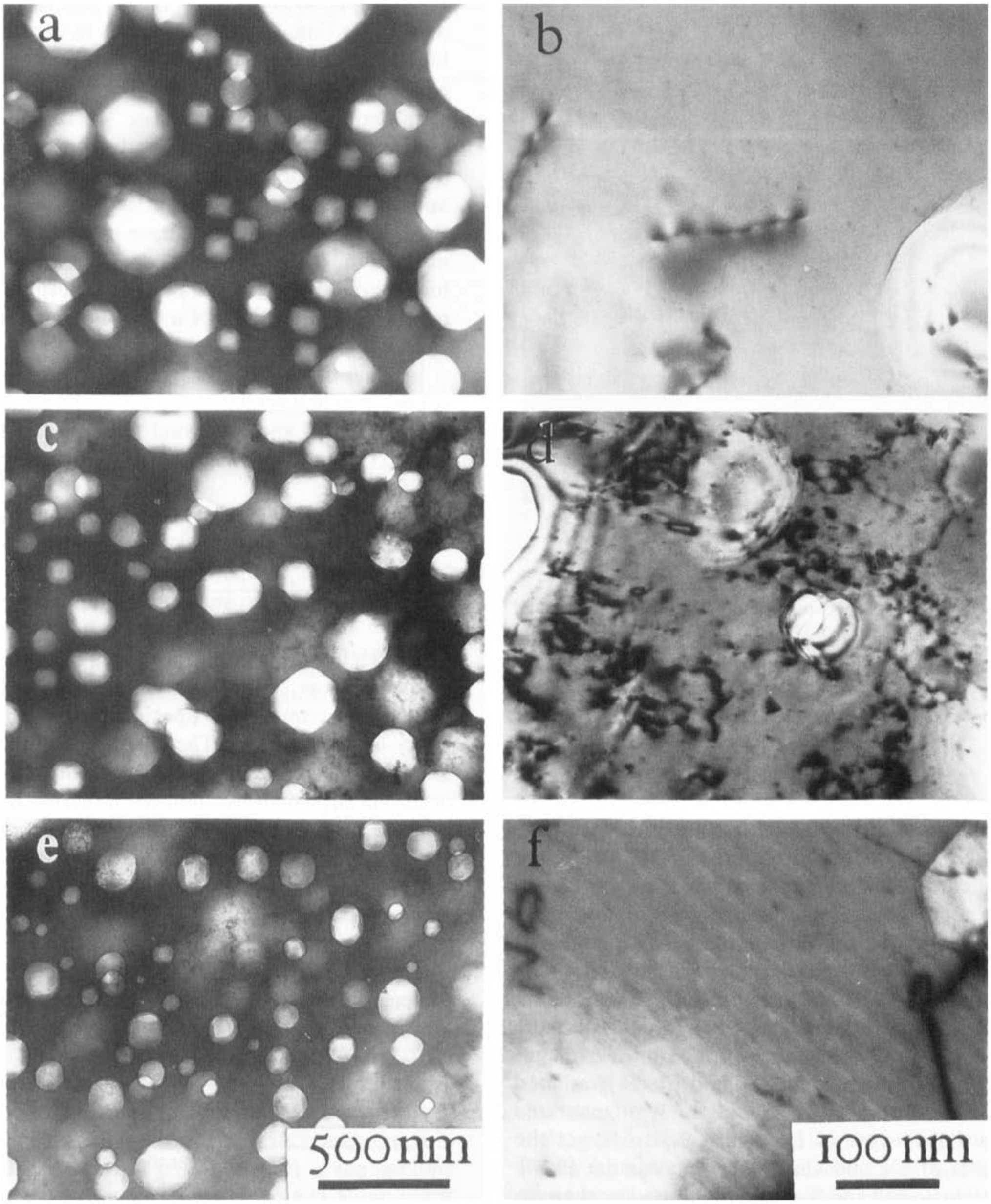

Fig 2 Void and dislocation microstructures of specimens irradiated in FFTF/MOTA at 686 K to a dose level of ~ 48 dpa (a), (b) Cu, without helium, (c), (d) Cu containing 100 appm of implanted helium and (e), (f) Cu containing about 90 appm of helium generated during irradiation with 800 MeV protons

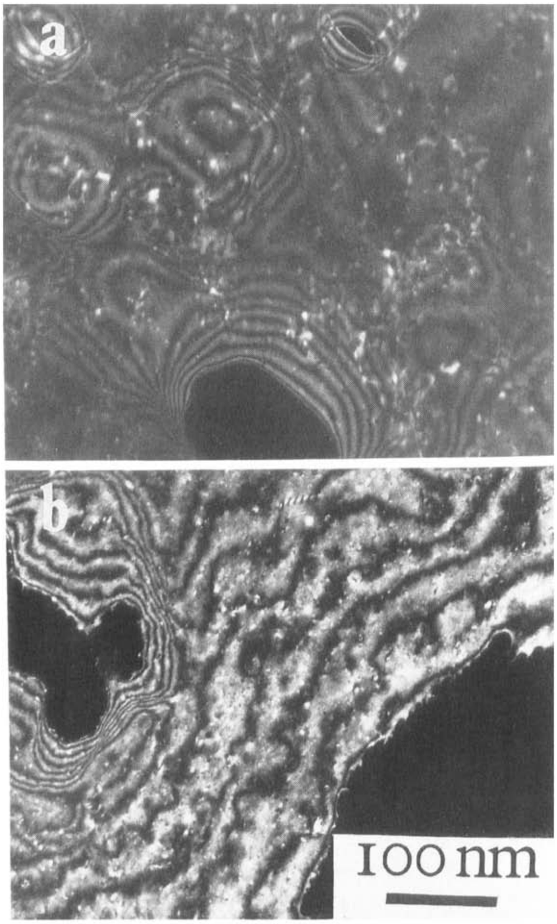

Fig 3 Defect clusters in copper containing (a) 100 appm of implanted helium and (b) 90 appm of (helium produced during 800 MeV proton irradiation) after irradiation in FFTF/MOTA at 686 K to a dose level of ~48 dpa.

tion temperature which yielded rather low void densities Even preimplantations of 100 and 90 appm of helium in copper did not cause increases in void densities.

It is somewhat surprising that the void density in all three types of copper specimens turned out to be very similar In other words, the preimplantation of helium up to 100 appm had practically no influence on the final void density. Similar observations have been reported for neutron-irradiated Cu and Cu–B irradiated at 673 K, although in Cu–B helium was generated continuously at a rate of 1.5×10^{18} at./m^3/s, yet the void density in Cu and Cu–B was very similar [9,10]. Recently, Horsewell and Singh (unpublished work) have investigated copper irradiated with 600 MeV protons at 673 K, the production rate of helium in this experiment was about 100 appm per dpa Here too, the high helium generation rate did not cause any increase in void density

The fact that the void densities in pure copper and copper containing helium are found to be about the same, does not mean that the same nucleation mechanism has been operative in the two cases. In pure Cu, for instance, the dominating driving force for the nucleation must have been the available vacancy supersaturation and the residual gas atoms during very early stages of irradiation. The void nucleation in Cu under neutron irradiation conditions may terminate already, for example, at about 10^{-2} dpa at 523 K [11]. Since the nucleation time/dose is expected to be even lower at higher temperatures [12], in the present experiment at 686 K, the void nucleation in pure Cu might have saturated at a very low dose It should be noted, however, that the neutron irradiation of pure Cu to a dose of ~48 dpa is likely to have produced about 15 appm of helium [13] Thus, it is possible that some of these helium atoms might have caused additional void nucleation. This may be one of the reasons for the higher void density in the present experiment than that reported for Cu [9] and Cu–B [10] irradiated at 673 K to a dose of only 1 dpa.

A quantitative analysis of the cavity nucleation in copper specimens containing 100 or 90 appm of helium is complicated and is beyond the scope of the present paper. Qualitatively, however, it is worthwhile to note the following:

(i) prior to FFTF/MOTA irradiation, the sink density in these specimens is very high. Consequently, the buildup of the vacancy supersaturation is likely to be slow and hence the nucleation might have been delayed.

(ii) Helium atoms in both types of specimens must be present (as estimated from by the PAT measurements in the implanted specimen) in the form of vacancy–helium clusters. The coarsening of these clusters leading to the nucleation and growth of cavities may occur via Brownian-like motion, radiation resolution by cascade impingement and thermal resolution. At an

Table 1
Void swelling data for copper and copper containing 100 and 90 appm of helium irradiated in MOTA at 686 K to a dose level of ~48 dpa

Material	Void			Cluster density $[m^{-3}]$
	Size [nm]	Density $[m^{-3}]$	Swelling [%]	
Cu	182 8	$3 3\times10^{19}$	10.6	$2 5\times10^{21}$
Cu + 100 appm He	142 0	$4 6\times10^{19}$	6 8	$4 4\times10^{21}$
Cu + 90 appm He	137 5	2.9×10^{19}	3 9	$1 8\times10^{21}$

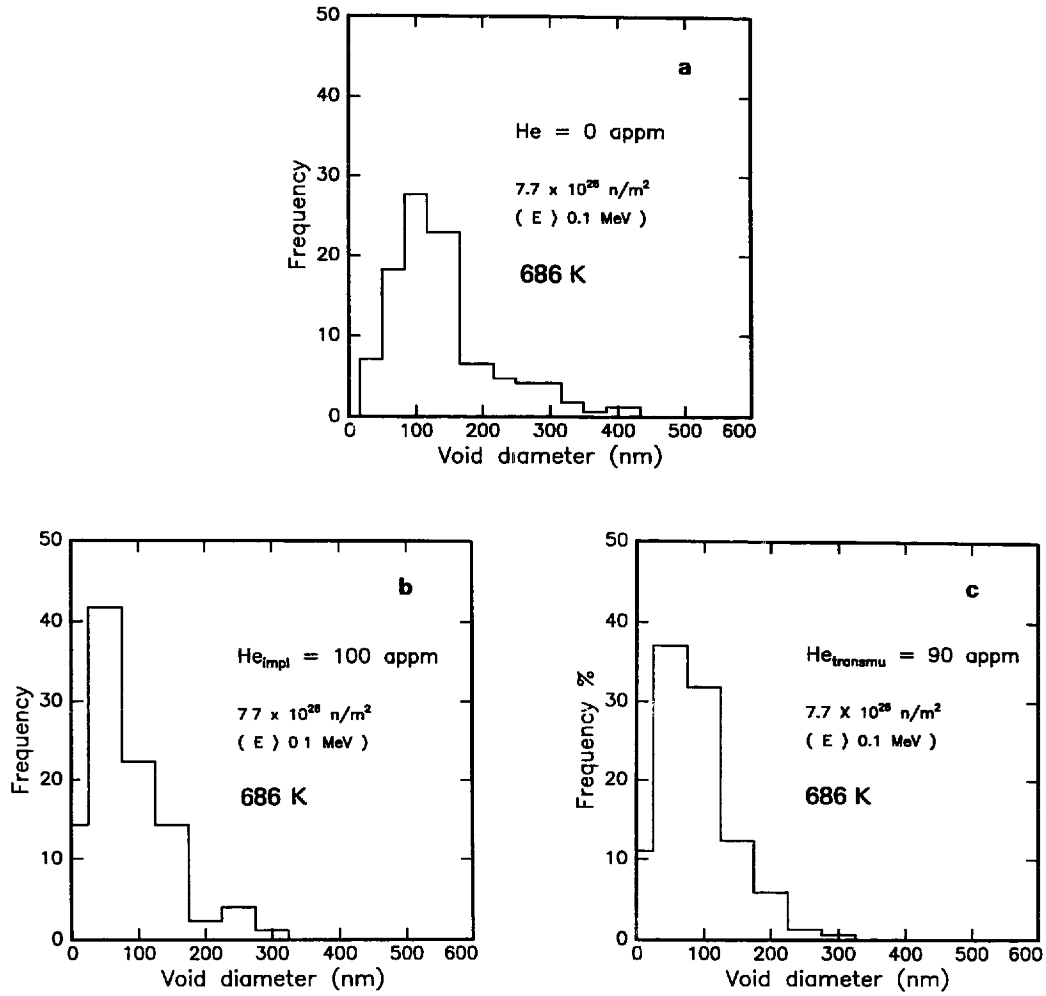

Fig 4 Void size distribution for (a) copper without helium, (b) copper with 100 appm of helium and (c) with 90 appm of helium following irradiation in FFTF/MOTA at 686 K to a dose level of ~ 48 dpa

appropriate vacancy supersaturation level, the bubbles would transform to voids that will continue to grow.

(iii) The presence of small bubbles in these specimens even after a dose of ~ 48 dpa would suggest that a secondary nucleation of bubbles must be taking place primarily via radiation resolution of helium atoms.

It is interesting to note here that the present results showing practically no effect of implanted or irradiation-induced helium on the final density of voids, are contrary to results obtained by 1 MeV electron irradiation of helium-implanted copper [14] In these experiments, preimplantation with helium increases the void density, reduces the void growth as well as the swelling rate at irradiation temperatures up to 623 K; at 723 K the swelling rate is not affected. These differences between the results of neutron and electron irradiations cannot be rationalized easily.

Table 1 shows that the void swelling in Cu containing 100 and 90 appm helium is noticeably reduced and that this reduction occurs exclusively by a substantial reduction in the void size (figs. 2 and 4). This reduction in the void size may arise due to the following reason The preirradiation microstructure of the helium containing specimens contains a very high density of very small helium bubbles. These bubbles will coarsen during MOTA irradiation and the growth rate of the cavities is likely to be very low because of the very high sink density The growth rate would remain low until the bubbles have coarsened and grown into voids and the cavity sink density has reached a certain low value This means that the voids in the specimens containing helium, must have had shorter time period (smaller dose) for full growth and hence the smaller size.

In Cu containing 90 appm of helium introduced by

800 MeV proton irradiation a further decrease in the void swelling is noted. This decrease in swelling (with respect to the swelling in the Cu containing 100 appm of implanted helium), however, seems to be caused primarily by a reduction in the void density. It is possible that the production of additional impurity atoms, particularly nickel, via mutational reactions during irradiation with 800 MeV protons might be responsible for this reduction This would be consistent with the recent observations that the presence of Ni drastically reduces the void density in Cu [7].

5. Conclusions

No void lattices were observed in the neutron irradiated copper and copper containing up to 100 appm of helium to a dose level of ~ 48 dpa.

The void densities in pure copper and in copper containing helium up to 100 appm were found to be very similar after neutron irradiation in FFTF/MOTA to a displacement dose level of ~ 48 dpa. The nucleation in pure copper may be controlled by the available vacancy supersaturation and the concentration of residual gas atoms. In the helium containing copper, on the other hand, the cavity nucleation might occur via coalescence of clusters of gas atoms, thermal resolution and radiation resolution.

The additional reduction in swelling in copper irradiated with 800 MeV protons (prior to irradiation in FFTF/MOTA) may occur via nucleation poisoning by transmutational impurities, particularly nickel.

The effects of preimplanted helium on the final void density in copper irradiated with 1 MeV electrons are quite different from what has been observed in the present irradiation experiments with fission neutrons. This difference is not fully understood and needs further study.

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