APPLICATION OF CYCLOTRONS IN MATERIALS SCIENCE

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ABSTRACT

Particle accelerators have been used for some time in materials science to study the effects of irradiation damage and to implant impurity atoms to well defined concentrations. In the past the majority of this work has been of a fairly fundamental nature but recently important technological problems in connection with fast reactor development, have been highlighted. This paper will review the use of cyclotrons to study a variety of materials problems and in particular the Harwell Variable Energy Cyclotron to study the phenomenon of void formation in irradiated materials. It will illustrate how simulation experiments can help to provide an understanding of an important technological problem and furthermore provide data which can be used together with that from fast neutron irradiation to select the most suitable materials for the design of fast reactors.

INTRODUCTION

Particle accelerators have been used to study radiation effects in materials for many years. In general two basic phenomenon have been studied, namely radiation damage and the effects of implanted impurities. The particular advantage of cyclotrons in these studies is their ability to produce very energetic beams of heavy ions at substantial beam currents, e.g. 5uA of 50 MeV Ni^{6+} ions.

Early work was concerned with studies of a fairly fundamental nature, and was primarily directed at an understanding of the nature of radiation damage and defects in metals, e.g. Eggleston[1] and Cooper et al.[2] However, cyclotrons are currently being used to provide data for technologically important problems in fast reactor materials. In this paper we will briefly review the results of the more technologically oriented studies, emphasising those areas where the use of cyclotrons have made the largest impact.

ION IMPLANTATION

The inert gases which are produced in materials as a consequence of transmutation in a nuclear reactor, have many important consequences in the ultimate behaviour of reactor components. For instance, in nuclear fuels, the fission product gases Kr and Xe agglomerate to form bubbles which give rise to an overall swelling of the material. Furthermore, the inert gas He, which is produced in steels in both thermal and fast reactors, plays an important role in determining the ultimate mechanical properties of irradiated components.

It is often difficult to separate the effects of He and of radiation damage in reactor steels which have been exposed to high neutron doses. However, by implanting He into unirradiated material using, for instance, a cyclotron, it has been possible to isolate the significance of the gas. In order to obtain meaningful results, it is a necessary requirement to implant the He to an almost uniform concentration through foils of about 0.01" thickness. The implanted foils are then cut to produce conventional tensile samples and subjected to a variety of thermal treatments prior to testing.

The technique of producing uniform implantations has been discussed previously by Worth[3] and by King[4]. In this paper it will suffice simply to point out that such uniform implantations can easily be achieved by the use of a programmed energy degrader, either in the form of a tapered wedge or a tapered wheel. From our knowledge of the stopping power of He ions in solids we can simply compute the rate at which the energy degrader should be moved in order to produce a uniform concentration of implanted atoms.

As an example of this type of experiment, we will discuss the effect of He implantation on the ductility of 304 stainless steel at high temperatures, carried out on the Oak Ridge Isochronous Cyclotron by King[4]. Tensile specimens of 304 steel were uniformly implanted with He to an atomic concentration of 0.83×10^{-6} and annealed for 1 h at 929°C. These specimens were then subjected to standard tensile tests at temperatures from 500°C to 900°C, together with control specimens, at a strain rate of 0.026 min^{-1}. No significant changes were found in the yield strength, ultimate tensile strength, however the effects of He on the total strain of 304 steel are illustrated in Fig. 1. Within the limits of the experimental uncertainty, the results demonstrate that although 0.83×10^{-6} at. con. He has essentially no effect on the tensile ductility at 500 and 600°C, severe reductions of ductility occur over the 700°-900°C test temperature range. Experiments on similar samples irradiated in a thermal reactor to produce comparable He concentrations show qualitatively similar reductions in ductility. However, in the reactor case substantial radiation damage is produced within the specimens, and we are therefore led to conclude that the loss in ductility can be ascribed to the presence of He.

This review is not the place to discuss the possible

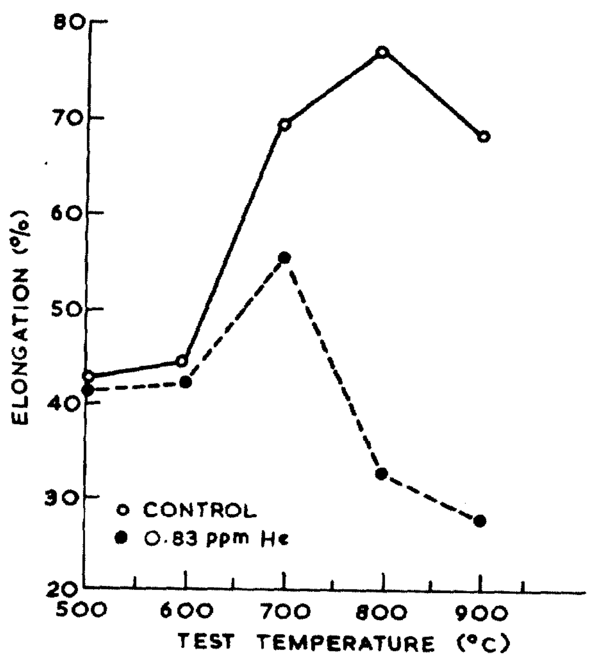

Fig. 1 Ductility of 304 steel annealed 1 h at 925°C and tensile tested at 0.026 mm^{-1} strain rate. (From King[4])

mechanisms of the embrittlement of steels and the above experiment will suffice as an example of how cyclotrons have helped our understanding of the problems.

RADIATION CREEP

The creep of solids at elevated temperatures under stress is a well known phenomenon. However, it has been realised for some time that during reactor irradiations, creep rates are enhanced over and above those expected for purely thermal conditions. Such enhancement is generally ascribed, through one mechanism or another, to the increased defect concentrations which are generated during the irradiation.

The phenomenon of radiation enhanced creep can conveniently be studied during cyclotron irradiation. Under suitable conditions the radiation damage rate produced within a test specimen during cyclotron irradiation can be made essentially uniform throughout its thickness. Given such a situation, and providing the specimen temperature can be adequately controlled, the enhancement of creep during such an irradiation can readily be measured using more or less standard techniques. Work of this nature is currently being carried out at Argonne, Harwell and Karlsruhe. However, in this instance we will choose to illustrate the experiments from the work of Harkness et al[5] using the cyclotron operated by the Chemistry Division of Argonne National Laboratory.

Deuteron beams at currents up to 10uA cm^{-2} at 22.4 MeV were passed through a rotating energy degrader such that the damage throughout a 0.007 in. test specimen of annealed 304 stainless steel was approximately uniform. The sample was mounted in a specially designed rig such that the load was transmitted through the sample via a hydraulic load train. Strain measurement was achieved using a linear variable differential transducer such that deflections of 50u in. could be reliably measured. The deuteron beam from the cyclotron impinged onto the sample which was maintained at constant temperature. Figure 2 shows a typical result of the enhancement of creep in 304 steel at 420°C at a stress of 51,500 psi during a deuteron flux of 4.3uA on sample. The creep rate was clearly enhanced during the bombardment and the results have been used to shed light on the strain rate sensitivity and temperature dependence of irradiation creep.

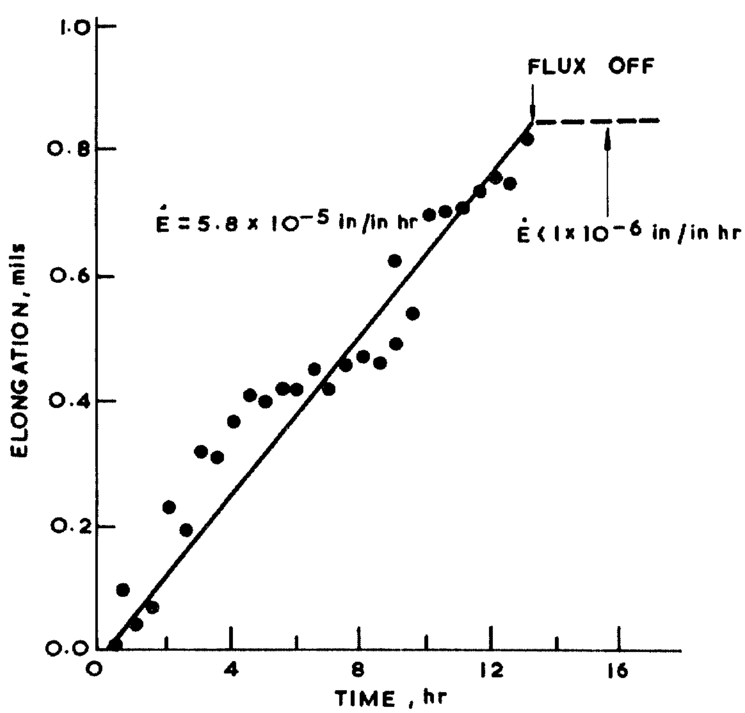

Fig. 2 Test results showing enhanced creep due to deuteron bombardment. (From Harkness et al[5])

VOID FORMATION

The observation by Cawthorne and Fulton in 1966[6] that fast reactor steels subjected to high neutron exposures, exhibit prolific void formation, has been the inspiration for wealth of experimental and theoretical studies[7]. The acquisition of data relevant to fast reactor design problems by fast neutron irradiation is, by its nature, a tedious and time consuming business. An attempt to obtain void swelling data over a shorter timescale has been made using charged particle accelerators such as the Variable Energy Cyclotron (VEC) at Harwell. Due to their larger mass and stronger interaction with the atoms of a solid, a beam of energetic charged particles can produce irradiation damage at a rate many thousand times faster than can neutrons in a fast reactor. So in principle, the damage density produced during many years irradiation with a reactor can be simulated in a few hours using ion beams.

The basic requirement is to produce a region of uniform damage within a metal specimen whilst the specimen is maintained at an elevated temperature. Furthermore, the bombarding ion species must be chosen such that it does not produce adverse chemical or physical effects within the sample which might influence the formation of voids. Also the damage must be sufficiently removed from the surface such that the results are representative of the bulk material. Finally, the presence of He - which is created within the reactor as a result of (n,α) reactions - has led to speculation as to its possible influence on void formation. To this end an attempt has been made to simulate the reactor condition by implanting He uniformly throughout the sample prior to irradiation, as outlined above. In practice it would be more realistic to introduce the He during the damaging irradiation, but this introduces experimental difficulties. A compromise situation is to introduce He and to carry out the damage irradiation alternatively in an attempt to build up the He and damage concurrently.

It is generally assumed that it is the total number of displaced atoms which is important for the formation and growth of voids, a fundamental requirement for the comparison between accelerator and reactor irradiations is the normalization of results via the total number of displaced atoms. The theoretical considerations behind such a normalization have been discussed in detail previously[7]. In this instance it will be sufficient to point out that the most difficult part of the normalization is in calculating the damage produced during fast neutron irradiation. However, the best estimates to date suggest that the damage produced by a total neutron dose of 4×10^{22} neutrons/cm^{-2} at the centre of the Dounreay Fast Reactor (DFR) is ~20 displacements/atom.

The choice of ion species and ion energy are inter-related and to some extent conflicting requirements necessitate compromise. Ideally one would like the particles to pass right through the sample, for example high energy protons; however, because of their low scattering cross-section, the time taken to accumulate sufficient damage is generally prohibitive. On the other hand, if we have no alternative but to stop the ions in the specimen we must avoid using those gaseous ions which are insoluble in the specimen, particularly the inert gases and other gaseous elements which are thought to influence the nucleation of voids. Similarly, any other ion that is likely to cause precipitation within the sample during irradiation should be avoided. The most obvious ion species would therefore be "self ions", i.e. in the case of a nickel specimen, Ni ions. Up until recently, the availability of self ions comparable with structural steels and nickel based alloys has been rather limited, and a large amount of data has been collected using ions such as carbon; carbon is already present in steels and the addition of extra atoms is thought only to present problems at high doses. However, recent work using Ni ions has in general borne out the results previously obtained using carbon.

A general feature of neutron and ion irradiated metals is the existence of zones denuded of voids on either side of the grain boundaries. Such zones are thought to be a direct consequence of the role of grain boundaries as a defect sink. Similar denudation effects are to be expected at free surfaces. Furthermore, free surfaces tend to modify the radiation induced dislocation structures in their vicinity especially during irradiation at elevated temperature. Such dislocations are thought to be crucial in determining the rate of void growth, and quantitative data should if possible, therefore, be obtained from regions of the sample at least ~0.5um below the surface. In addition, in order to simulate more precisely the neutron case and to facilitate examination, it is desirable to produce a uniformly damaged region about 1-3um behind this 0.5um surface layer. The damage distributions for 20 MeV C^{++} ions and 48 MeV Ni^{6+} ions from the VEC produce rather sharp peaks below the surface, see Fig. 3. However, such distributions can be used together with a programmed rocking of the target to produce a uniform damage zone over any required depth below the surface.[8] Typical rocked damage distributions using the above ions from the VEC are also shown in Fig. 3.

As previously mentioned the acquisition of quantitative data depends on relating the total damage produced during ion bombardment to that produced during neutron irradiation. It is therefore of some importance to check that irradiations to the same calculated damage dose in the reactor and accelerator do in fact correspond. We must, of course, be careful to choose a system such that any dose rate effects can be neglected, or at least accounted for. In this context it is possible to choose corresponding temperatures for nickel, which at the peak swelling temperatures show little or no effect of dose rate on the magnitude of the swelling. In order to eliminate the variability which might result from different helium concentrations, it was decided to perform these check irradiations with nickel samples, taken from the same parent material, each of which having been previously annealed to the same

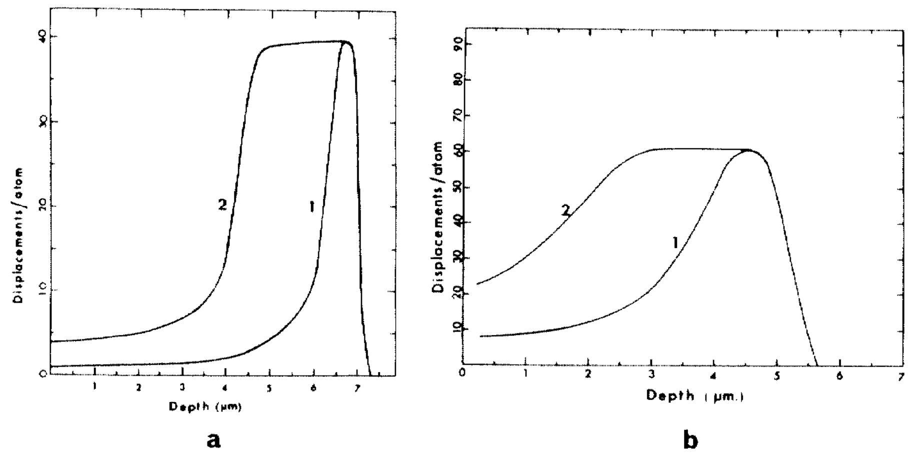

Fig. 3 Depth distributions of damage: 1 - unrocked, 2 - rocked.
(a) 20 MeV C^{++} (b) 48 MeV Ni^{6+}.

temperature, and implanted uniformly with identical concentrations of He (10^{-5} atom/atom). A selection of samples were then irradiated in either DFR or the VEC at corresponding temperatures to equal total damage doses. Figure 4 shows two electron micrographs of

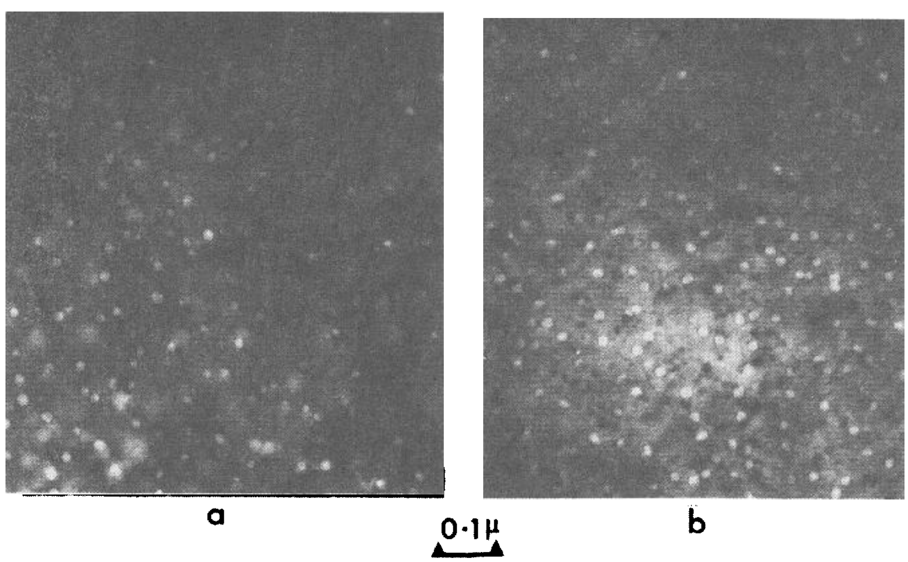

Fig. 4 Comparison between DFR and VEC irradiated Ni containing 10^{-5} atomic conc. of helium. Both samples were irradiated to the same calculated dose of 5 displacements/atom.

voids which illustrate the results of this experiment.* A computation of swelling from a knowledge of the void density, the void size and the foil thickness in the two cases gives answers which, within the expected experimental error, agree remarkably well. This result, therefore gives us confidence both in the simulative technique and in the model for calculating damage.

Due to the restricted depth of the irradiated volume during ion bombardment we are limited to the use of the transmission electron microscope for the examination of samples. The volume swelling ($\Delta V/V$) is calculated from the void size and density. In this review we will just outline some of the more important results. Figure 5 shows a sequence of electron micrographs showing the change in void microstructure in 316 steel as a function of displacement dose at 525°C. Figure 6 shows this same data in graphical form whereas Fig. 7 shows the temperature dependence of void swelling at 40 displacements/atom (equivalent to 8×10^{22} neutrons/cm).

Data such as illustrated in Figs. 5, 6 and 7 above together with that from reactor irradiations can be used to assess the relevance of void swelling in fast reactor design. For instance, due to the non-uniform damage rates throughout a reactor, some key components are expected to suffer distortion as a consequence of differential swelling. Such distortions must be minimised either by operational or engineering modifications or by finding alternative materials. The choice of other materials is restricted to those which are both compatible with liquid sodium at temperatures up to about 700°C, and at the same time, exhibit the correct mechanical behaviour. For some time scientists within the UKAEA have been studying a selection of nickel based alloys, and in particular a nimonic alloy called PE 16. After a suitable heat treatment this material contains within its structure a fine dispersion of very small precipitates, called gamma prime precipitates (γ'). For instance, after heating to 750°C for four hours, the γ' precipitates grow to about 100 Å diameter and have an average separation of just over 500 Å. It was decided to perform a series of accelerator irradiations on this material. The results turned out to be extremely encouraging. For irradiation at 525°C to 200 displacements/atom (about 5 years in DFR) the total swelling was less than about 0.4%. Figure 8 shows the swelling plotted as a function of dose.

The main role of the accelerator studies has been to produce advance data to damage doses which will not be achieved in the reactor for many years. However, the final tests will, of course, have to be made using reactors.

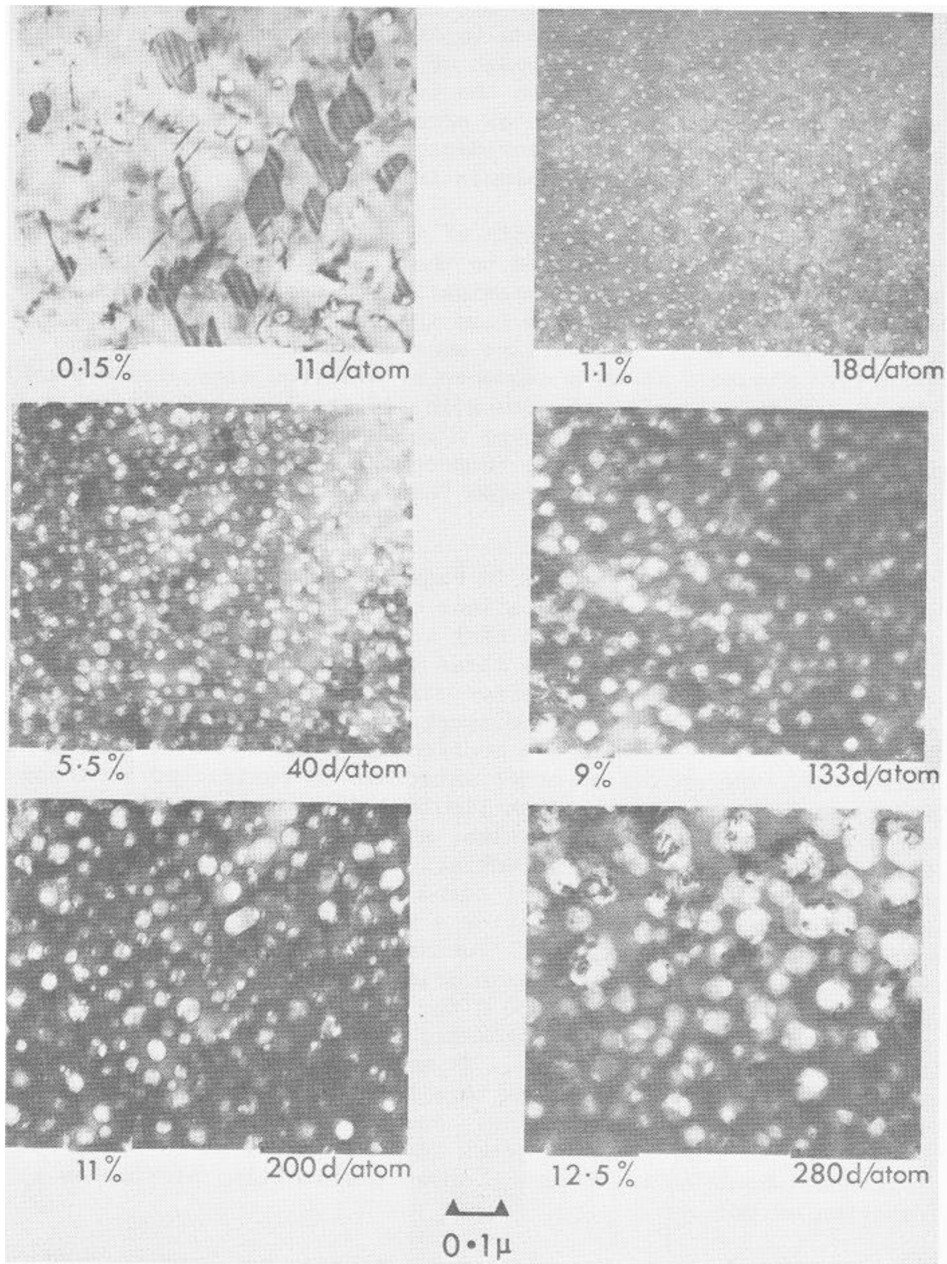

Fig. 5 Micrographs showing the increase in voidage with dose – 20 MeV C^{++} bombardment of 316 steel at 525°C.

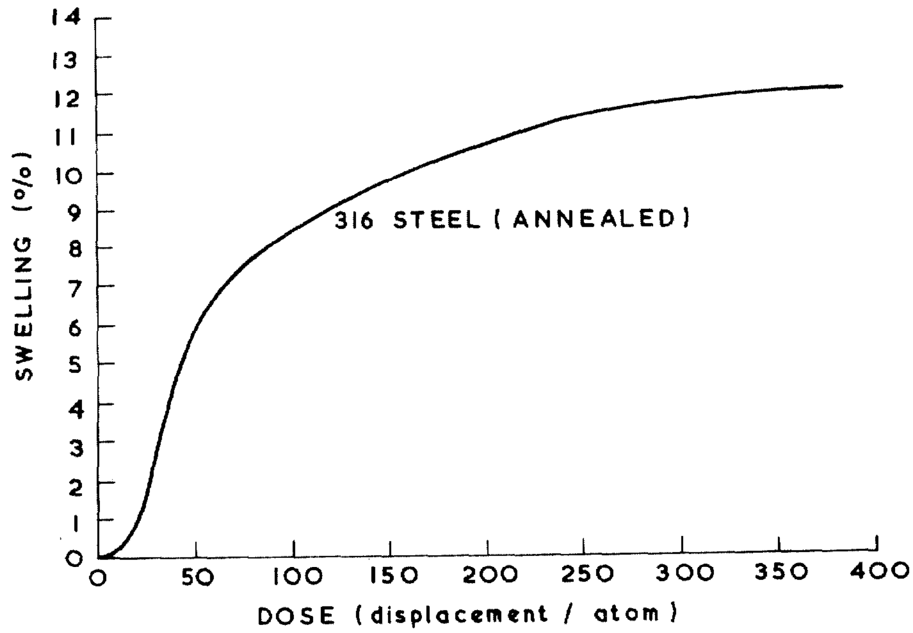

Fig. 6 Graph of the dose dependence of swelling in 316 steel at 525°C.

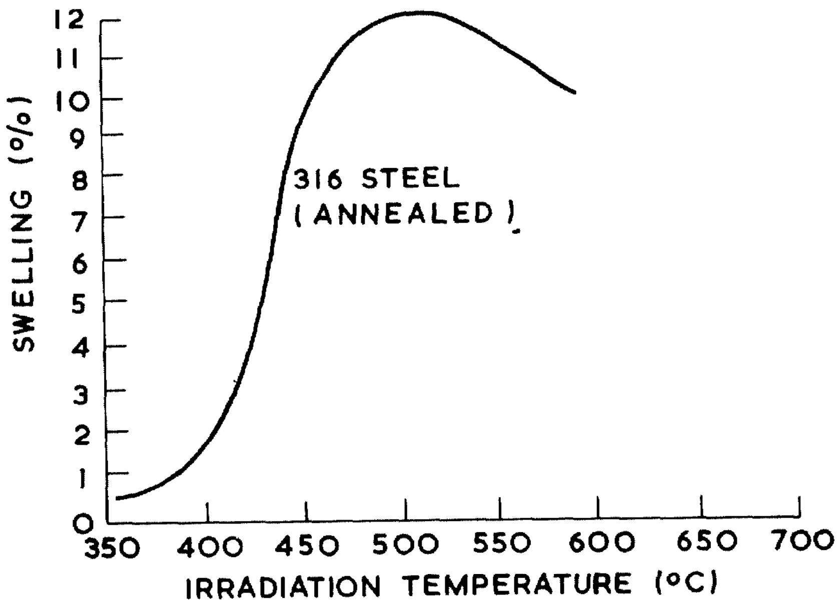

Fig. 7 Temperature dependence of void swelling in 316 steel at 40 displacements/atom. (The curve has been moved by 100°C to lower temperatures to account for the increased dose rate compared with reactor irradiation.)

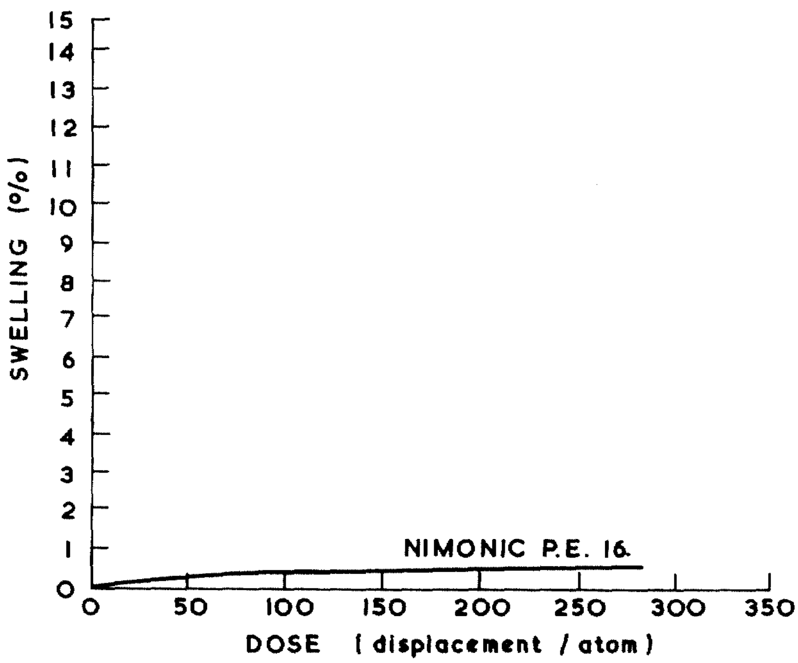

Fig. 8 Dose dependence of swelling in Nimonic PE 16 at 525°C.

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DISCUSSION

HENDRY: With regard to that very interesting slide you showed with the 280 displacement per atom, could you comment on what the beam current, the ion you used in the beam, what the current density was and what the length of the irradiation?

NELSON: That particular thing was done with 20 MeV C^{++} in the cyclotron, and the beam current was of the order of 5 or so μA. One of the problems is getting the heat out of the sample that is being put in, because we have to control the temperature very accurately. A lot of these things have all been repeated with 48 MeV N^{6+} where the current is then of the order of 5 or 6 μA. You can work out the time.

FLOOD: I wonder if you have done any work with single crystal growth or metamorphosis under irradiation or neutron bombardment like alumina to sapphire, or anything like that?

NELSON: We haven't actually done anything on the cyclotron on single crystals.

WARREN: Have you done any work, or have you any information concerning the mechánism by which holes are formed in foils by beam bombardment even though the temperature is kept down?

NELSON: What do you mean by 'down', room temperature?

WARREN: Yes.

NELSON: I think the theory of void formation that I have described is pretty well established. This in the case of steel would be between 300 and 700. In the case of copper it would be lower; in the case of aluminium it occurs at room temperature. There are some rather sophisticated computer calculations done now by people in England--by Buller and people at Harwell--and by people in America--Harkness at Argonne--and people in other places. The theory is fairly well established as to why it occurs, but whether the theory is sufficiently quantitative is another matter.

RAINWATER: The figure you had up at first for creep: seems to me that you were talking about something different than I usually think of as creep. I think of creep as where you have a material under tension and it just slowly enlarges, but if you don't have it, you seem to be referring to something where there is no indication of any particular stressing.

NELSON: The second slide was a creep where the thing was under stress

RAINWATER: But no indication of the kind of figures of material. This was under a condition where when you stopped irradiating it stopped moving.

NELSON: When I stopped it that was done with 304 steel about 450°C which is where the thermal creep at such temperature is very, very slow. But when I stopped to irradiate, there was some creep but it was very, very small compared to that which was enhanced by the irradiation.

RAINWATER: But the effect that you showed was not just the volume increase.

NELSON: No, that was a normal creep experiment.

MICHAELIS: Have you any information on effects of this kind caused by, let's say, protons of higher energy--100 MeV or above?

NELSON: One hasn't done specific experiments with protons at higher energy, but I would have thought that one could calculate the radiation damage production very well with protons at higher energy because the collision is purely a Rutherford-type collision. One can then have fairly low primary recall spectra, and one could calculate the radiation damage production quite well. I would have thought relating that to what we know about the swelling at different damage rates, one could get really quite a good guess. But we haven't done anything specifically.