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Simulating Void Formation in Fast Reactor Steels

DK 669.1:621.039.5

By Drs. J. A. Hudson, D. J. Mazey and R. S. Nelson

Bei den seit einigen Jahren in Betrieb stehenden Prototypanlagen von schnellen Brutreaktoren wurde festgestellt, dass infolge der gegenüber herkömmlichen Kernreaktoren erheblich höheren Neutronendichten im Kern die Werkstoffe gewissen Strukturänderungen unterworfen sind. Die Langzeitbestrahlung der Reaktorwandungen mit schnellen Neutronen (über 10^5 eV) bewirkt unter anderem die Bildung von Hohlräumen im Werkstoff, die die Materialdichte verändern. Es wurden Wege gesucht, dieses Materialverhalten

The Harwell Variable Energy Cyclotron has proved to be very useful in determining void formation rates in metals likely to be used for components in fast breeder reactors where fast neutron doses will be much higher than those experienced in today's reactors.

With the advent of the fast reactor, certain components will be subjected to fast neutron doses far exceeding those hitherto achieved. Even at this early stage of development it is well established that after *prolonged irradiation* at temperatures about 0.4 of the absolute melting temperature stainless steel components undergo density changes resulting from radiation-induced porosity. This porosity has been identified to result from the formation and growth of small voids a few hundred Å in diameter, within the material [1].

The voids are thought to nucleate as small gas bubbles from either the helium which is formed continuously by transmutation during the irradiation or from dissolved gas already present in the material. The bubbles grow into voids by the condensation of irradiation-produced vacancies. Vacancies and interstitials are produced in equal numbers during irradiation but the net flux of vacancies to voids is thought to occur because dislocations in the metal have a slight preferential attraction for interstitials.

The elastic scattering cross-section for charged particle collisions is, in general, several orders of magnitude higher than that for neutron-atom collisions. For this reason the damage build-up which takes years in a reactor can, in principle, be simulated within hours in a particle accelerator. The limitation is that whereas fast neutrons produce uniform radiation damage throughout a large specimen, ion bombardment can only result in damage to a depth roughly equal to the ion's range in the solid, which is relatively small. Ion bombarded solids can thus only be investigated for void formation by techniques suitable for small volumes, and in practice transmission electron microscopy is used in most cases.

Although it is a relatively simple matter to form voids during ion bombardment at the appropriate temperature, several special conditions must be stipulated for a useful quantitative simulation of the neutron case. In the first place

unter kontrollierten Laboratoriumsbedingungen innert kurzer Zeit zu reproduzieren, um es untersuchen zu können. Mit der Methode der Simulation der Neutronenbestrahlung im Zyklotron mit variabler Energie der UK Atomic Energy Authority (UKAEA) in Harwell wurden zufriedenstellende Ergebnisse erzielt; in einigen Materialien wurde die Hohlräumbildung in Abhängigkeit von der Bestrahlungsintensität und von der Neutronendichte ermittelt. Die Versuchsdaten sowie Ergebnisse werden im folgenden Beitrag beschrieben.

the radiation damage must be formed effectively in bulk material away from surfaces which influence the point defect and dislocation content of the solid; both these quantities are of paramount importance in determining the kinetics of void growth. Second, the quantitative comparison between the radiation damage produced by fast neutrons and that produced by the ion bombardment must be known. Third, it is desirable to produce the ion bombardment damage uniformly within a given volume.

Formation Experiments

These considerations have led to the choice of the Harwell Variable Energy Cyclotron (VEC) for void formation experiments. A comparison of this with other accelerators from the point of view of void formation has been made previously [3]. Until recently the bombarding species in the experiments has been 20 MeV C^{++} ions. From examination of specimens extracted at different depths below

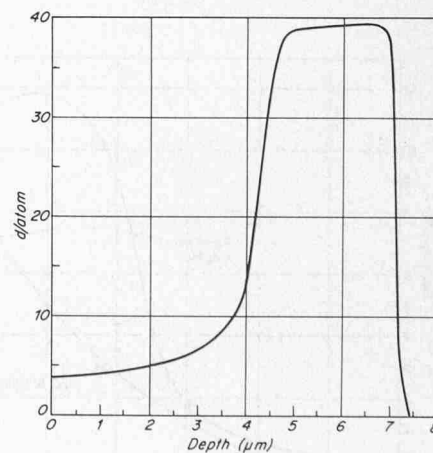


Figure 1. Damage density in displacements/atom (d/atom) as a function of depth for a 20 MeV C^{++} irradiation of nickel or metal of similar density

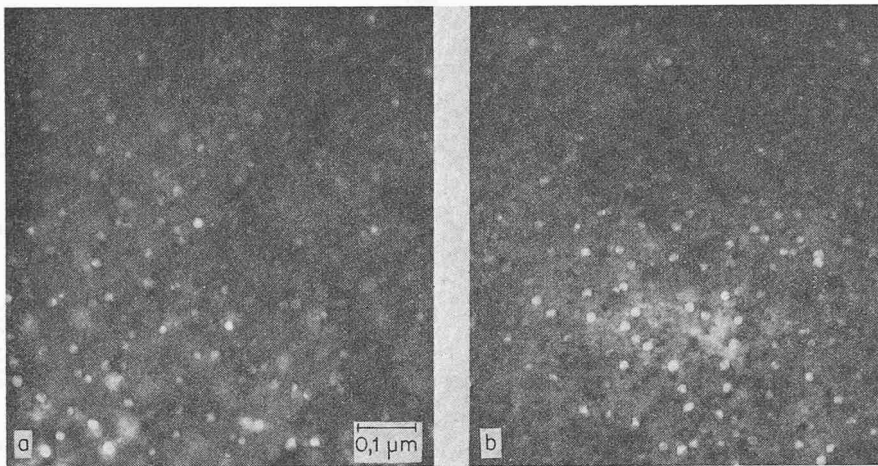


Figure 2. Comparison of voids produced in nickel by neutron and ion irradiation. a DFR (Dounreay Fast Reactor), b VEC (Variable Energy Cyclotron)

the bombarded surface of nickel foils it has been possible to calibrate the damage density versus depth profile for the C^{++} irradiations [3].

Basis of the calibration is a comparison of the volume fraction of voids formed at different depths for different irradiation doses. At all doses the damage density in the early stages of the C^{++} track can be calculated since here the elastic collisions are in the regime of the Rutherford scattering cross-section. Figure 1 shows the damage density in displacements/atom (d/atom) as a function of depth for a 20 MeV C^{++} irradiation of nickel (or metal of similar density). In this case the foils have been mounted on a "rocking target holder". This apparatus rocks the foils in a controlled manner during irradiation to widen the damage peak of the C^{++} ions.

The control function was devised from the previously determined calibration curve for normal incidence irradiation during which 40 d/atom would be created at the peak by 3×10^{17} ions/cm². As the Figure shows, the rocking holder produces an almost uniform damage density from 5 μm down to 7 μm below the surface. A 2 μm uniform depth of damage has been chosen as the best compromise between the desire to obtain uniform damage and the restriction of irradiation times in the cyclotron.

More recently, irradiations have been performed with 48 MeV Ni^{6+} ions. In this case one has the advantage of damage produced quickly and away from the surface, and in addition the introduction of impurities such as carbon is avoided.

In order to simulate the helium production by (n,α) reactions in the reactor, helium is uniformly implanted prior to irradiation to a concentration of 10^{-5}

In order to relate the results of VEC irradiations to the reactor case, the damage density for a particular fast neutron spectrum must be known. Calculations have now been made for the spectra in the Dounreay Fast Reactor (DFR) [4]. Figure 2 shows voids in two nickel specimens, one irradiated in the VEC with 20 MeV C^{++} ions and the other irradiated in the DFR. The ion bombardment was arranged to produce the same number of displaced atoms as that calculated to have accumulated during the reactor irradiation. The similarity in the void populations and particularly the values of swelling is readily apparent. In direct comparisons of VEC and reactor irradiations the increased dose rate (about 1000 times) of the VEC must be considered. In general this larger dose rate increases the temperature of maximum void growth by about 100 °C [2].

In the case of nickel the swelling versus temperature curve shows a flat region of about 150 °C over which maximum swelling occurs. This is not the case in stainless steel, as indicated at Figure 3 which shows results of 20 MeV C^{++} irradiations of solution treated and 20% cold-worked 316 stainless steel over the temperature range between 400 and 700 °C. The increased swelling resistance of 20% cold-worked steel is also indicated at Figure 3. The effect of cold working on swelling in stainless steel has been described previously [5,6].

Void swelling as a function of dose in pure nickel, solution treated 316 stainless steel, and Nimonic PE16 is shown in Figure 4. The results were obtained from VEC irradiations at 525 °C of foils containing 10^{-5} He with 20 MeV C^{++} ions. Consistent with reactor data, voids in nickel were first observed after relatively low radiation damage doses – about 0.1 d/atom. Over most of the dose range, swelling in nickel varied as a linear function of dose up to 6% at 40 d/atom. The swelling rate then fell to give 15% swelling at 350 d/atom.

The 316 stainless steel showed marked differences in swelling behaviour from nickel, as indicated at Figure 4. No voids were visible in specimens irradiated to less than 2 d/atom. Above this dose the swelling increased approximately as (dose)² up to about 40 d/atom. Above this dose too, where the swelling was about 5%, the swelling rate decreased and, as shown at Figure 4, the swelling curve merged with that for nickel to give about 15% swelling after about 2 d/atom at a swelling of about 0.1%.

Included in Figure 4 is a result from a 4 MeV proton irradiation which gave 0.01% swelling after 0.75 d/atom.

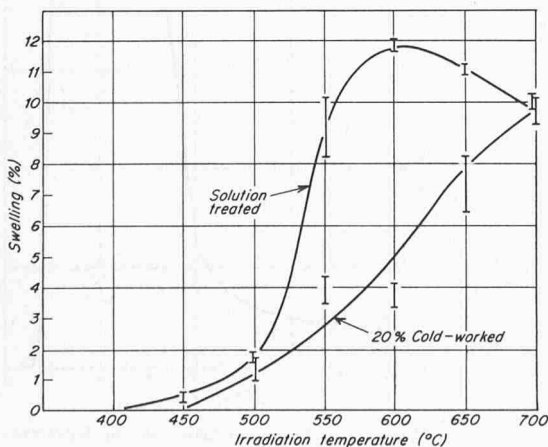


Figure 3. Temperature dependence of swelling in A151 316 steel

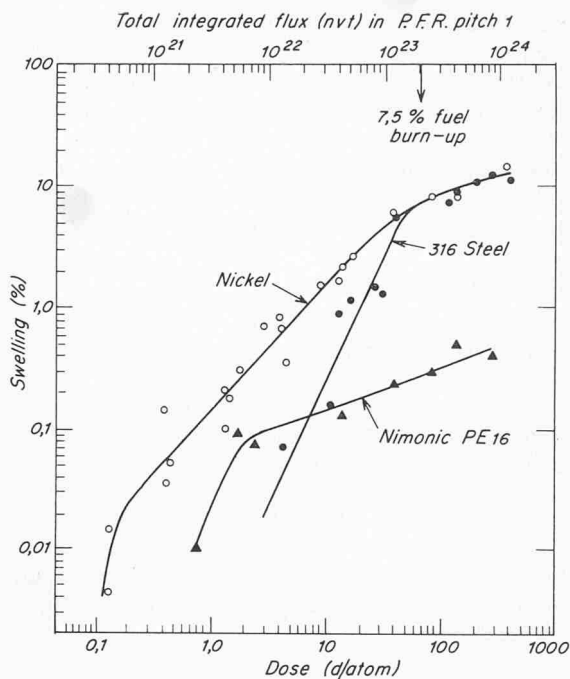


Figure 4. Dose dependence of void swelling in nickel, 316 steel and nimonic PE 16 irradiated at 525 °C with 20 MeV carbon ions in the VEC

Above 2 d/atom the swelling increased slowly [approximately as $(\text{dose})^{1/3}$] to reach about 0.5% after 300 d/atom. Those results are for PE 16 which was solution treated and aged to give γ' precipitates [$\text{Ni}_3(\text{Ti,Al})$] about 100 Å in diameter.

A Chief Parameter

Although swelling is manifest as voids it is of course physically caused by the equivalent interstitial content in the metal. This is present as radiation-produced dislocation loops, lines and networks in addition to those interstitials which reach the free surface. Current theories of void growth [2] show that dislocation concentration is one of the chief parameters which determine the kinetics of void growth. The experimental determination of dislocation content is thus important in testing the predictions of theoretical models.

As stated earlier it is necessary in experiments aimed at simulating fast-neutron-induced voidage to produce the damage sufficiently far away from surfaces that the dislocation and point defect concentrations are unaltered by the proximity of the surfaces. In all three face centred cubic metals studied, the first appearance of voids was accompanied by the first appearance of small ($< 50 \text{ \AA}$) dislocation loops some of which could be positively identified as faulted interstitial loops lying on [111] planes. In nickel the loops soon unfaulted as the dose increased and grew to form a network which increased to maximum density of about $5 \times 10^{10} \text{ cm/cm}^3$ at about 40 d/atom. Above this dose, where large voids were produced by coalescence, the dislocation density decreased. In steel the dislocation density increased sharply to reach 10^{11} cm/cm^3 at 10 d/atom, then levelled off to reach 2×10^{11} at 350 d/atom but did not decrease as in nickel. In PE16 the dislocation density again increased sharply and was greater than 10^{11} cm/cm^3 at all doses above 2 d/atom.

The interpretation of the swelling behaviour of nickel and stainless steel with respect to the observed dislocation densities and current theories of void growth have been given elsewhere [2,7]. Two possible causes of the suppression of void growth in PE16 have been given. First, the coherent interface around the γ' precipitates promotes recombination by constraining point defects to move in the interface without loss of identity. Second, the radiation-produced dislocation array quickly becomes very dense, being locked by the precipitates, and greatly enhances recombination.

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Verzeichnis der Hochbauforschungsstellen 1972/73

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Vorwort

Im Jahre 1972 erreichte das gesamte Bauvolumen der Schweiz 24,6 Mrd. Fr., was rund 21% des Bruttosozialprodukts entspricht – ungefähr die Hälfte der baulichen Umwelt ist während der letzten 25 Jahr errichtet worden. Allein diese zwei Angaben sollten die heutige Bedeutung des Bauens genügend unterstreichen. Verbesserungen und Fortschritte können zum Teil nur durch Erschliessung neuer Erkenntnisse, das heisst Forschung, erarbeitet werden. Daher wird der Bauforschung heute einige Bedeutung zugemessen. Sie verlangt jedoch, dass bewusst koordiniert vorgegangen wird, um aus den gegebenen Mitteln das Maximum herauszuholen.

Drei Stellen haben sich zusammengetan, um ihre parallel geführte Informationssammlung über die Hochbauforschungsstelle der Schweiz zusammenzutragen und einer breiteren Öffentlichkeit zugänglich zu machen:

– GFB, Schweizerische Gesellschaft für Bauforschung.

- FIB, SIA-Fachgruppe für industrielles Bauen im Hoch- und Tiefbau
- HBF, Institut für Hochbauforschung der ETH Zürich

Sachbearbeiter für diese Erhebung ist R. Kehrli, HBF; Mitarbeiter sind J. Piller, GFB, C. Vezin, HBF, sowie die FIB.

In den nachfolgenden Zusammenstellungen sind die wichtigsten Ergebnisse, nämlich die Anschriften der ermittelten Stellen, die sich mit Hochbauforschung befassen, enthalten. Im separat herausgegebenen Gesamtverzeichnis¹⁾ sind die vollständigen Antworten auf die Umfrage zusammen getragen.

Gegenstand, Ziel und Umfang der Erhebung

Gegenstand der Erhebung war die Erfassung der an der Hochbauforschung beteiligten Organisationen bzw. Institutio-

¹⁾ Verzeichnis der Hochbauforschungsstellen der Schweiz. Umfang 260 Seiten A4, Preis 90 Fr. Zu beziehen beim SIA-Generalsekretariat, Postfach, 8031 Zürich, Tel. 01 / 36 15 70.