STP484-EB/Jan. 1970

AGENDA DISCUSSION SESSION

How Do We Solve the Void Problem?

T. T. Claudson[1]—The effect of metal swelling on the design, operation, and safety of fast reactors has only recently become apparent through the design efforts associated with the Fast Flux Test Facility (FFTF) and LMFBR demonstration plants. Extrapolation of current swelling data to high burnup indicates that swelling of cladding and structural materials will limit the reactor core performance. Core performance limitations due to materials behavior during the early stages of the development of a new reactor concept is not new. In the past metallurgical solutions to these problems have been possible, such as the development of corrosion-resistant zirconium alloys for water reactor systems. There are data at present which indicate again that there may be metallurgical solutions to the problems associated with metal swelling.

Swelling has been observed in several metals including aluminum, molybdenum, nickel, as well as in stainless steel. Through the comparison of the response of different metals and alloys, it is possible to gain some indication of several approaches to the solution of the problem. In the case of nickel it has been determined that impurity content and cold work are important. Figure 1 shows the change in volume as a function of fluence for nickel 200 and nickel 270 irradiated in the EBR-II. The response to swelling of Inconel 600 is also shown for comparison. One can see that annealed nickel 200 swells less than annealed nickel 270. Furthermore 50 percent cold-worked nickel 200 swells less than the same material in the annealed material. Inconel 600, a high nickel content alloy, did not swell although irradiated under the same conditions. The major difference between nickel 270 and nickel 200 was impurity content, the nickel 200 having about 1600 ppm of silicon.

Further indication of the effect of alloy content on swelling response is shown in Fig. 2. Specimens of AISI 316 and 348 were irradiated under identical conditions in the EBR-II. As temperature and fluence were increased the swelling response of the two alloys varied. Whereas the 348

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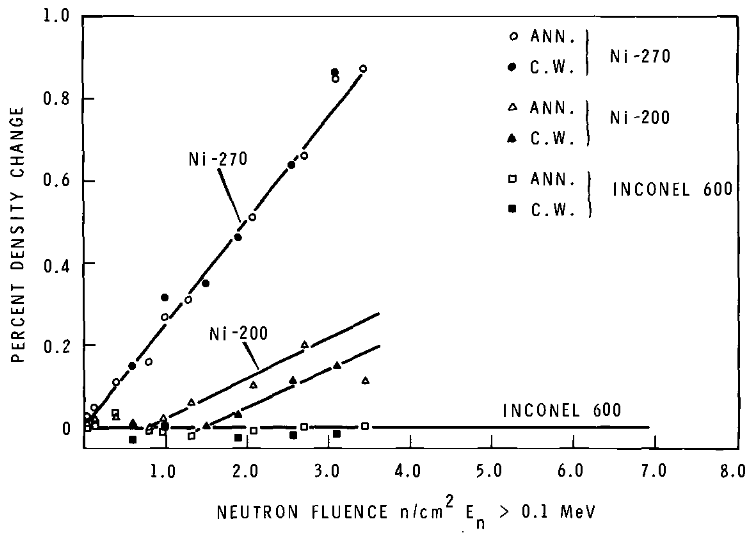

FIG. 1—*Swelling in nickel irradiated in the EBR-II at 480 C.*

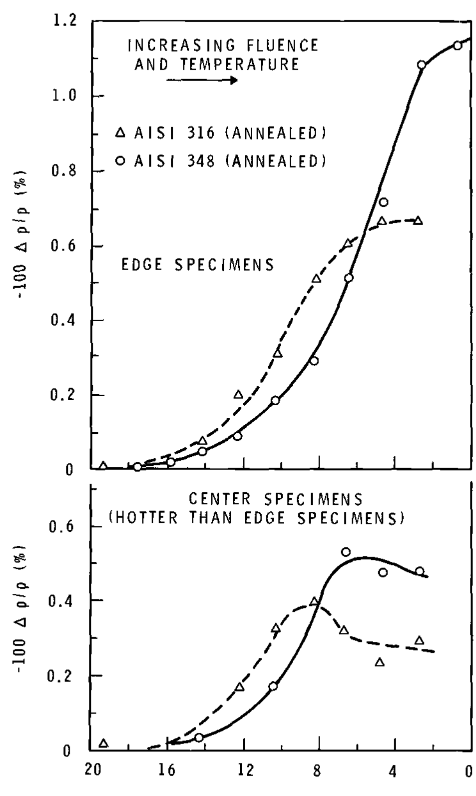

FIG. 2—*Effect of temperature and fluence on swelling of AISI 316 and 347.*

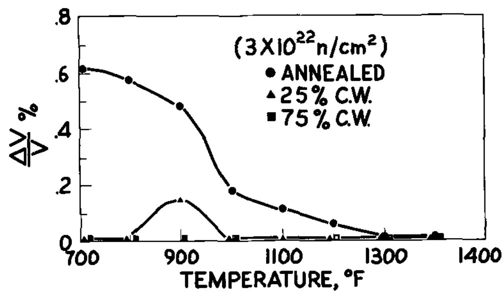

FIG. 3—*Effect of cold work on the swelling response of austenitic stainless steel.*

alloy had superior properties (lower swelling) at lower temperature and fluences, the 316 alloy was better as temperature and fluence were increased.

Metallurgical structure appears to have a significant effect on the swelling of metals. It has already been mentioned that cold-worked nickel swells less than the same material in the annealed condition. Figure 3 shows the effect of cold-working AISI 316 on swelling, at low temperatures the effect is marked. As irradiation temperatures increase the benefit of cold work becomes less apparent because of recovery.

From the above discussion it appears that swelling in engineering alloys may be affected by several metallurgical variables including structure and composition. This indicates that the solution to the problem may involve alloy composition modifications, heat treatment, mechanical working, or a combination of them, all of which are within present commercial capability.

The role of the designer and other related metallurgical phenomena must not be forgotten. While swelling will promote high bending stresses in core components such as ducts, it is anticipated that irradiation induced creep will aid in the relaxation of these stresses. Also, as designers become more acquainted with the problem and better means of predicting the actual swelling values as a function of stress, temperature, and fluence become available, improved designs can be effected. No doubt the true solution will involve a combination of these factors.

H. W. Wiedersich[2]—With regard to the differences in the various theories on void formation, I would like to make some comments. The theories are still in a state of flux. Even though the basic models used are similar and the gross features of swelling emerge from any one of the theories, their quantitative predictions do vary significantly. Swelling is a very complicated phenomenon, and, therefore, approximations which simplify the complexity must be made. The differences in theories and, hence, in

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results lie mainly in the differences between the approximations used. With improvements in the understanding of the subtleties of the phenomenon, a more realistic assessment of the consequences of the various simplifying assumptions will be possible and appropriate modifications will be made. On the request of Dr. Bullough, I give just one example of the differences.

The assumptions that voids grow during the early stages in a point defect environment essentially determined by dislocation sinks and recombination and that there exists *no* concentration gradient of defects in the vicinity of a void yield a void volume growth proportional to the cube of time, t^3, as suggested by Bullough and co-workers. If, however, the concentration gradient resulting from the defect loss to the void is assumed not to be negligible, one obtains a 3/2 power dependence in time for otherwise unchanged conditions. The latter assumption is contained in the treatment of Harkness and Li. Of course, their result on the time dependence is more complicated, because they allow the point defect environment to change as nucleation and growth proceeds.

Gases which dissolve in metals and have reasonably high diffusion coefficients at temperatures of void formation, such as hydrogen and nitrogen, may ease the nucleation of voids by virtue of lowering of the surface energy by absorption. However, they will not reduce the barrier to nucleation to any significant extent by the internal pressure in the nuclei in equilibrium with the dissolved gas. The internal pressure may build up to a sufficiently large magnitude only when gases with very low solubilities and, hence, very high equilibrium pressures are trapped and low diffusion coefficients prevent their escape to surfaces in the temperature range of interest. Aside from noble gases, notably helium, so-called "residual hydrogen" could be of importance to void nucleation. As H. H. Podgurski [*1*][3] has shown, hydrogen can be trapped in steel in the form of CH_4, which is rather insoluble and immobile.

A fine dispersion of precipitates may reduce the swelling by providing additional sinks for point defects. Precipitates in the matrix are expected to act as self-compensating sinks for interstitials and vacancies, because any excess annihilation of interstitials will produce a compressive strain field around the precipitate which attracts vacancies and repels interstitials. Excess vacancy annihilation will reverse the situation. Grain boundaries, precipitates in boundaries, and dislocation networks are not self-compensating sinks. The precipitates have to be highly dispersed to be effective; their number density should be higher than typical observed void densities, that is, greater than 10^{15} cm^{-3}.

J. D. Elen[4]—Concerning the regime for void formation in body centered cubic metals, I would like to show the influence of chemical interstitial

[3] Italic numbers in brackets refer to the list of references at the end of this paper.
[4] PETTEN, Reactor Centrum Nederland, Netherlands.

impurity on the damage structure in vanadium. A series of three purities of recrystallized vanadium was irradiated in the high flux reactor at Petten to a fast neutron dose of 1.6×10^{20}n/cm^2 in a flux of 1.1×10^{14} n/cm^2-s, $E>0.1$ MeV. The carbon, oxygen, and nitrogen contamination is given below.

Chemical Impurity, ppm			Interstitial Damage, μm		Voids		
N	C	0	l	d	N, cm^{-3}	$\bar{a}$, Å	V, deg/00
310	355	255	0.2	0.4	9.4×10^{14}	68	0.3
610	330	2030	0.4	0.8	5.0×10^{14}	110	0.9
340	385	700	0.6	1.1	3.5×10^{13}	283	1.4

The as-irradiated damage structure is essentially different from the small cluster type observed by Elen [2] and Rau et al [3] in pure vanadium after similar irradiations. The irradiation capsule here, not being instrumented, might have been different in temperature. From postirradiation annealing the irradiation temperature was estimated to be about 200 C. A uniform distribution is observed of large dislocation tangles (Fig. 1), centered at second-phase particles. The original impurity determined structure is swept out. The tangles consist of large interstitial {111} 1/2**a** ⟨111⟩ dislocation loops.

The vacancy damage shows up in multiple-beam condition as a population of perfect cubic voids (Fig. 2). Considering the various purities, there seems to be a relation between the number N, the average size $\bar{a}$,

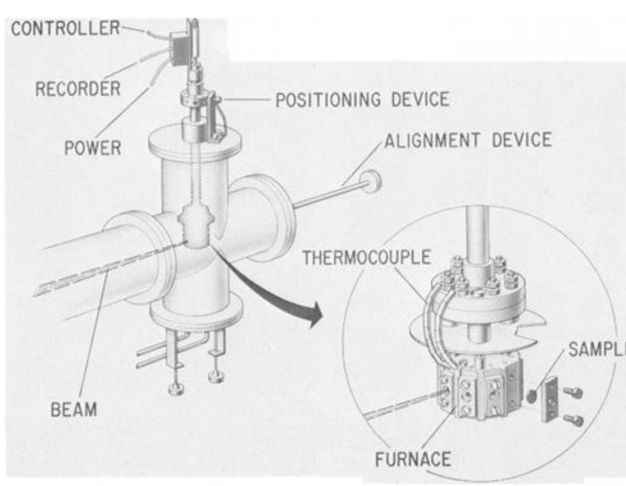

FIG. 4—*Specimen chamber for ion bombardment studies.*

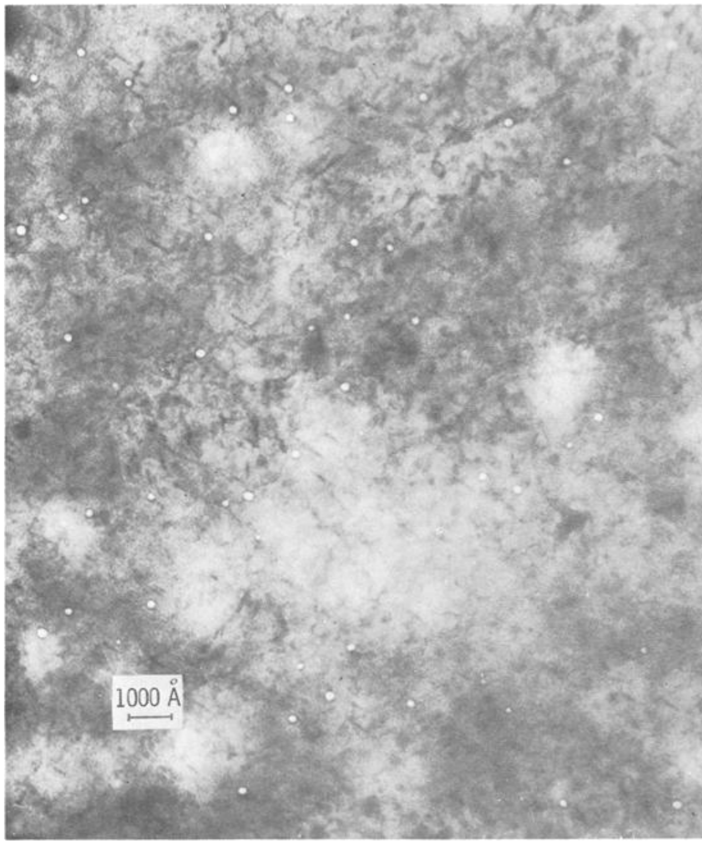

FIG. 5—*Voids in 20 percent cold-worked 316 stainless steel after irradiation with 5-MeV Ni^{++} at 565 C to 3×10^{16} ions/cm^2.*

and the total volume fraction V of the voids and the size l and interspacing d of the interstitial loop tangles. There is no systematic change with purity. The observations are tabulated for brevity. They might be considered in terms of the influence of the interstitial damage nucleation, which seems to be determined by impurity, on the supersaturation of vacancies.

G. L. Kulcinski, H. R. Brager, and J. J. Laidler[5]—The use of charged particle irradiation to produce voids in stainless steel was first demonstrated by Nelson and Mazey [*4*] in 1969. These authors initially used 100-keV protons and carbon ions, but later 22-MeV carbon and 100-keV nickel ions were used [*5*, *6*]. High-temperature bombardment with these ions resulted in a microstructure very similar to the voids and faulted loops observed after high-temperature neutron irradiation. More recently,

[5] Pacific Northwest Laboratories, Battelle Memorial Institute, Richland, Wash. 99352.

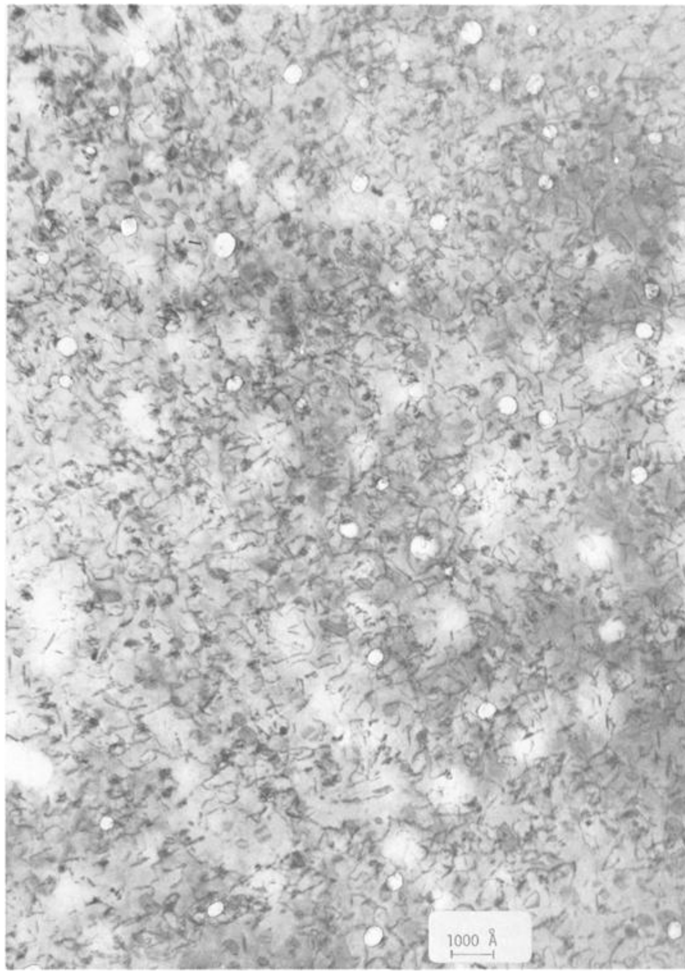

FIG. 6—*Voids in solution treated 316 stainless steel irradiated with 5-MeV* Ni^{++} *at 585 C to* 3×10^{16} *ions/cm*^{2}.

Keefer et al [7] have used 1.2-MeV protons and Kulcinski et al [8] have used 5-MeV Cu ions to produce voids in Type 316 stainless steel.

Ideally, one would like to use bombarding ions which will not substantially alter the chemistry of the metal or alloy bombarded. This requirement in stainless steels would dictate the use of iron, chromium, or nickel and in some special cases molybdenum, columbium, or titanium. Furthermore, to make meaningful correlations between ion and neutron fluences and to eliminate surface effects, the incident ion should penetrate at least a few thousand angstroms. The latter requirement means that heavy ion energies ≥ 1 MeV must be achieved.

This note describes an experiment designed to fulfill the above requirements by using 5-MeV Ni^{++} ions to produce voids in 316 stainless steel at 565 to 585 C. The void size and density in the ion bombarded material is consistent with that which would be observed after an equivalent neutron fluence of 4×10^{22} n/cm², $E>0.1$ MeV, at the temperature.

Experimentally, the specimens were irradiated at temperatures from 565 to 585 C while in intimate contact with a resistance heated stainless steel furnace (Fig. 4). Beam currents of up to 9 μA/cm² (5.4×10^{13} ions/cm²/s) were used in this study.

The specimen material was certified Type 316 stainless steel which was solution treated for 10 min in hydrogen at 1040 C. Some of this material underwent a 20 percent reduction in area after this treatment. The actual specimens were 3-mm-diameter disks 0.2 to 0.22 mm thick. The preparation of the specimens for microscopy is described elsewhere [*9*].

Typical microstructures of the solution treated and 20 percent cold-worked material after irradiation with 5-MeV Ni^{++} to a fluence of 3×10^{16} ions/cm² are shown in Figs. 5 and 6. In both materials voids and faulted loops were present. The void density is somewhat lower in the solution treated specimens (3×10^{13} cm^{-3} versus 6×10^{13} cm^{-3}) and the void size is somewhat larger (400 Å versus 250 Å) than the cold-worked specimens. The calculated volume increase is approximately 0.1 percent for the solution treated specimen and approximately 0.05 percent for the cold-worked steel.

Calculations of the equivalent neutron fluence to be expected as a result of irradiation with 3×10^{16} ions/cm² were made from the theory of Lindhard et al [*10*] in a manner described elsewhere [*8*]. The results of the calculations indicate that the equivalent neutron fluence should be in the range of 2 to 5 $\times$ 10^{22} n/cm². For the specific specimens of this study, 4×10^{22} n/cm² is a reasonable estimate.

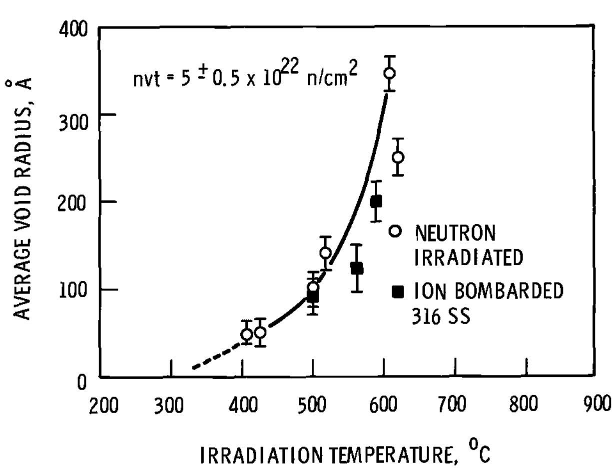

FIG. 7—*Effect of irradiation temperature on the void size in austenitic stainless steel.*

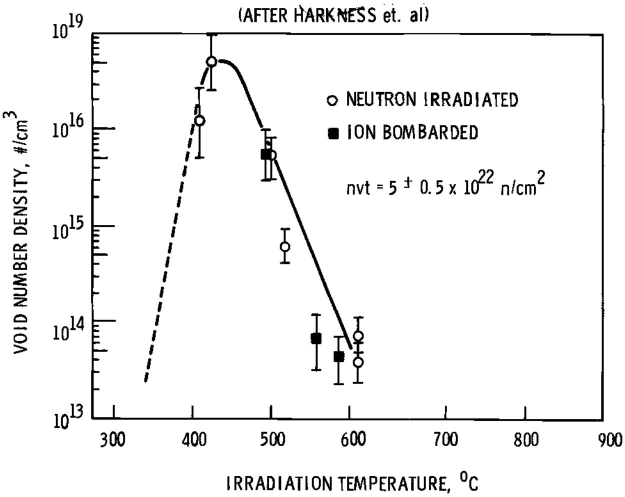

FIG. 8—*Effect of irradiation temperature on void number density in austenitic stainless steel.*

Figures 7 and 8, taken from the data of Harkness et al [*11*], show how the void size and density (open circles) vary with temperature at a constant neutron fluence of approximately $5 \times 10^{22} n/cm^2$. The closed circles are the data from this study and one datum point from a previous experiment [*8*] using 5-MeV copper ions. It is obvious that the agreement between the neutron irradiated and ion bombarded specimens is quite good, at least in this temperature and fluence range. These results also point out the insensitivity of the resulting microstructure to the rate at which the atoms are displaced. For a typical neutron irradiation in EBR-II the displacement rate is $2 \times 10^{-6} s^{-1}$, whereas in the present experiments it is in the neighborhood of $10^{-2} s^{-1}$.

References

[*1*] Podgurski, H. H., *Transactions of the American Institute of Mining, Metallurgical and Petroleum Engineers*, TAIMA, Vol. 221, 1961, p. 389.

[*2*] Elen, J. D., "Direct Observation of Damage in Neutron Irradiated Vanadium," RCN-96, Reactor Centrum Nederland, 1967.

[*3*] Rau, R. C. and Ladd, R. L., *Journal of Nuclear Materials*, JNUMA, Vol. 30, 1969, p. 297.

[*4*] Nelson, R. A. and Mazey, D. J., *Symposium on Radiation Damage in Reactor Materials*, Vol. I, International Atomic Energy Agency, Vienna, 1969, p. 157.

[*5*] Nelson, R. S., AERE-R-6151, Atomic Energy Research Establishment, Harwell, England, 1969.

[*6*] Mazey, D. J., *Journal of Nuclear Materials*, JNUMA, Vol. 35, 1970, p. 55.

[*7*] Keefer, D. W., Neely, H. H., Robinson, J. C., and Kramer, D., "Void Formation in Stainless Steel by Proton Irradiation," presented at this symposium, see pp. 332–346.

[*8*] Kulcinski, G. L., Laidler, J. J., and Doran, D. G., to be published.

[*9*] Brager, H. R., Kissinger, H. E., and Kulcinski, G. L., to be published.

[*10*] Lindhard, J., Scharff, M., and Schioott, H. E., *Matematiks-Fysiske Meddelser*, Danske Videnskabernes Selskab, MFYMA, Vol. 33, No. 14, 1963.

[*11*] Harkness, S. D. and Ki, C. Y., to be published.