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Factors which control the swelling of Fe–Cr–Ni ternary austenitic alloys

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Abstract

In agreement with limited earlier studies, a comprehensive irradiation experiment conducted in both EBR-II and FFTF demonstrates that while cold-working usually decreases void swelling of ternary Fe–Cr–Ni alloys at relatively low irradiation temperatures, it in general increases swelling at higher irradiation temperatures. Aging of cold-worked specimens to produce cellular dislocation networks tends to further increase swelling, especially at higher nickel levels. The swelling of ternary alloys at lower nickel levels also appears to be sensitive to details of the preirradiation annealing treatment. The differences in the details of reactor operating conditions also exert an influence on void nucleation and thereby on the duration of the transient regime of swelling. In the current irradiation series this leads to the swelling developed in EBR-II at ~ 30 dpa being consistently larger than that in FFTF. All of these results confirm an earlier conclusion that the primary variability of void swelling of FeCrNi alloys lies in the incubation and transient regimes, rather than in the steady-state swelling rate regime. Under certain conditions, the transient regime can be made to approach 0 dpa.

1. Introduction

There is a very general and long-standing perception [1–6], developed primarily from research on stainless steels, that cold-working a metal prior to irradiation always reduces radiation-induced void swelling, exerting a monotonic but diminishing influence on swelling with increasing cold-work level. It is also generally accepted that factors which promote void nucleation, such as high helium generation rates, can at least partially overcome the suppressive effect of cold-work on void nucleation.

In several recent reviews [7,8], however, it was shown that the role of starting dislocation density and dislocation arrangements, as well as their interactions with variables such as irradiation temperature or helium generation rate, are much more complex than previously assumed. In many cases, cold-working can actually increase swelling, especially in pure metals such as Ni and Al and also in simple ternary and quaternary austenitic alloys [7]. Both cold-worked and cold-worked and aged starting conditions sometimes lead to increased swelling of some commercial stainless steels as well, especially at high irradiation temperatures [8].

Instead of delaying swelling, Garner and co-workers have recently shown that cold-working often increases swelling of relatively simple Fe–Cr–Ni model alloys, especially under conditions where void nucleation is relatively difficult, i.e., for relatively high nickel or phosphorus levels and at relatively high irradiation temperatures [9,10]. At lower nickel levels and temperatures, where void nucleation is relatively easy, cold-working usually delays and thereby reduces swelling in Fe–Cr–Ni alloys.

The current interest in cold-work's influence on swelling arises from the use of both annealed and cold-worked alloys in a series of isotopic tailoring [11,12] and spectral tailoring experiments [13,14]. These experiments were di-

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Table 1
Composition in wt% of alloys employed in the EBR-II and FFTF irradiation experiments

Alloy designation	Fe	Ni	Cr	C	O	N
E18	Bal	12.1	15.1	0.005	0.016	0.0024
E90	Bal	15.7	15.6	0.013	n.m.	n.m.
E19	Bal	19.4	14.9	0.003	0.018	0.0015
E20	Bal	24.4	14.9	0.003	0.017	0.0017
E21	Bal	29.6	15.3	0.004	0.017	0.0020
E22	Bal	34.5	15.1	0.003	0.017	0.0021
E37	Bal	35.5	7.5	0.002	0.016	0.0013
E23	Bal	45.3	15.0	0.002	0.014	0.0017

n.m. = not measured.

rected toward determining the relative influence of various important material and environmental variables both on neutron-induced void swelling and concurrent changes in mechanical properties. Of particular interest in these studies was the role of helium and its interaction with other variables. The major interactive variables studied in both of these experimental series was that of the base composition and the thermomechanical starting state. Extensive prior work had determined the effect of both major elements and minor solute elements on void swelling, but these experiments concentrated primarily on alloys irradiated only in the annealed condition [15,16]. No comprehensive study involving cold-worked conditions of such simple alloys had previously been conducted.

2. Experimental details

To aid in the understanding of the relationships between compositional, thermomechanical and environmental variables, a series of Fe–Cr–Ni specimens irradiated in the AA-14 experiment in EBR-II was examined. Seven Fe–15Cr–XNi alloys (X = 12–45 at.%) were irradiated in 'weeper' packets in contact with the reactor sodium. In addition Fe–7.5Cr–35.5Ni was also irradiated. The specimens were in the form of 3 mm diameter microscopy disks and were irradiated at 425°C to 31 dpa and to 32 dpa at both 500 and 600°C. With only a few exceptions, one specimen of each alloy was irradiated side-by-side in each of three conditions: annealed (975°C/10 min/WQ), 30% cold-worked, and 30% cold-worked and aged (650°C/10 h/WQ). This insured that the effect of local variations in temperature or flux history did not interfere with the study of material variables. Post-irradiation measurements to determine density changes were performed using an immersion density technique known to be accurate to ±0.16% change in density.

The results of the EBR-II experiment were sufficiently interesting to warrant repeating this irradiation of the Fe–15Cr–Ni alloys under nominally similar conditions in the Fast Flux Test Facility (FFTF). Two specimens, rather than one, were used for each combination of material and thermomechanical starting condition in this second experiment, however. The irradiation conditions reached in FFTF were 12.3 dpa at 375°C, 35.8 dpa at 400°C, 36.3 dpa at 520°C and 33.2 dpa at 600°C. The major difference between the two irradiation experiments was that, while there was no active temperature control in the EBR-II experiment, the FFTF experiment was conducted in the Materials Open Test Assembly (MOTA) where the temperature is actively controlled within ±5°C.

The alloy compositions used in this study are shown in Table 1 and were identical for both the EBR-II and FFTF series of experiments.

3. Results: Swelling in EBR-II

The swelling values calculated from measured density changes induced in EBR-II are presented in Fig. 1 for Fe–15Cr–Ni alloys and in Table 2 for Fe–7.5Cr–35.5Ni. The following features of these data should be noted.

(a) In a previous study a plateau in swelling of annealed Fe–15Cr–Ni alloys was observed versus Ni concentration at low nickel levels and low irradiation temperatures, followed by a decrease in swelling with increasing nickel content thereafter [15]. At higher temperatures the plateau at low Ni concentrations often disappears. This behavior was preserved in the EBR-II portion of this experiment for all three starting conditions, but with one important exception. Note that in seven of the nine sets of data, swelling decreases somewhat at the lowest nickel levels, producing a peak in swelling versus nickel content. Such behavior has not been observed previously.

(b) The swelling at 31–32 dpa of annealed Fe–15Cr–Ni alloys at 425 and 500°C is relatively independent of temperature, but decreases significantly at 600°C, in agreement with the results of earlier studies [15]. The swelling of annealed Fe–7.5Cr–35.5Ni falls to near zero at 500°C and stays small at 600°C.

(c) Swelling of Fe–15Cr–Ni alloys at 425 and 500°C is decreased by cold-working, but at 600°C it is increased at higher nickel levels. A similar behavior was observed in Fe–7.5Cr–35.5Ni.

(d) The swelling of Fe–7.5Cr–35.5Ni was less at all temperatures and starting conditions than that of Fe–15.1Cr–34.5Ni, similar to the behavior of annealed speci-

Table 2
Swelling (%) observed in Fe–7.5Cr–35.5Ni irradiated in EBR-II

Irradiation temperature	425°C	500°C	600°C
Annealed	1.17	0.04	0.04
Cold-worked	0.05	−0.02	1.05
Cold-worked and aged	1.35	0.12	0.71

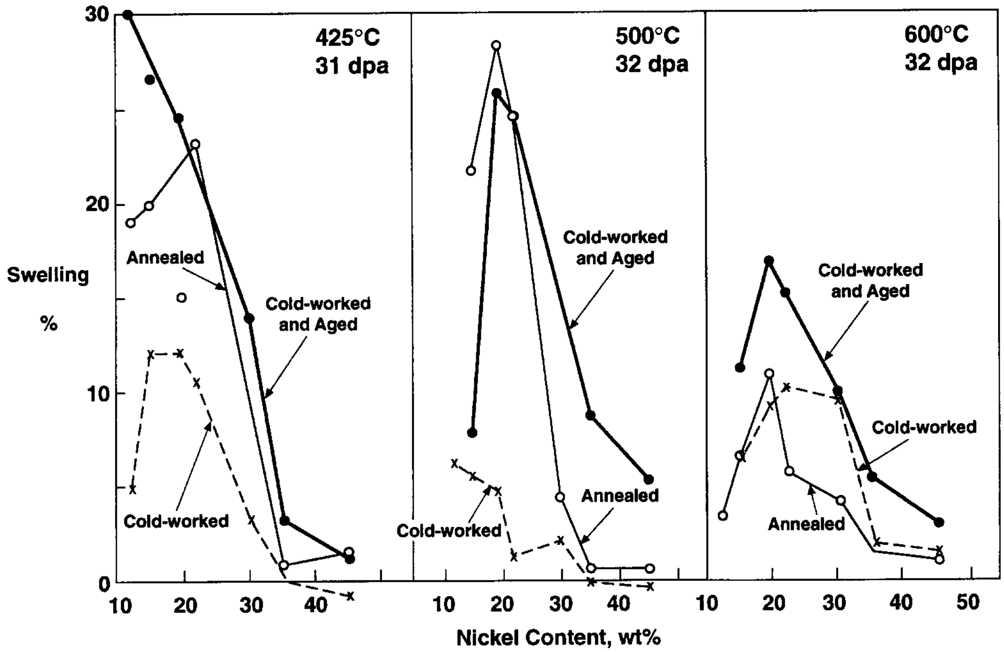

Fig. 1. Swelling observed in Fe–15Cr–Ni alloys irradiated in the EBR-II AA-14 experiment.

mens versus chromium content observed in earlier studies [15].

(e) Under some conditions, cold-working followed by aging can also significantly increase swelling, sometimes even to levels higher than that of annealed material.

4. Results: Swelling in FFTF

The swelling values of Fe–15Cr–Ni alloys calculated from measured density changes are presented in Figs. 2–5 for the FFTF irradiation experiment. Whenever the mea-

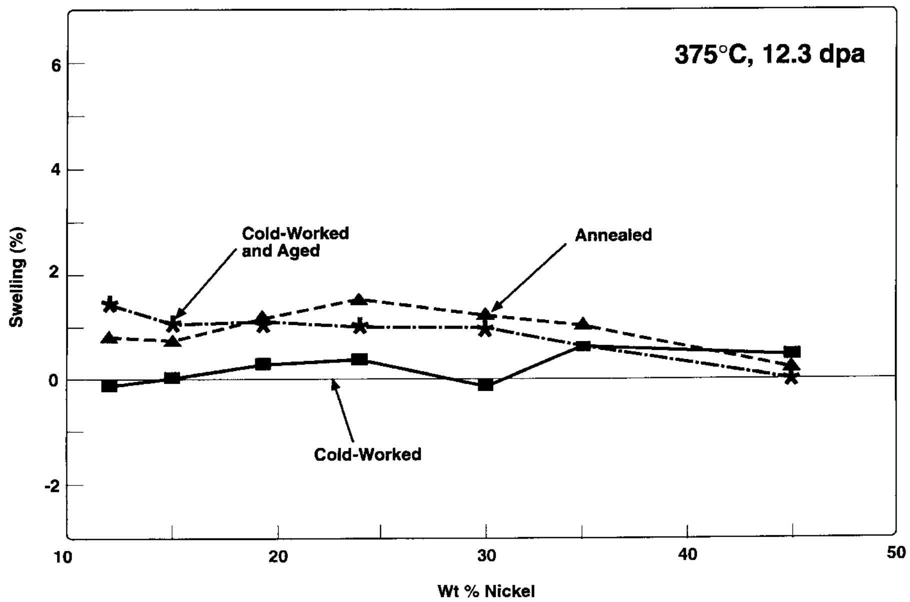

Fig. 2. Swelling observed in Fe–15Cr–Ni alloys irradiated in FFTF-MOTA to 12.3 dpa at 375°C.

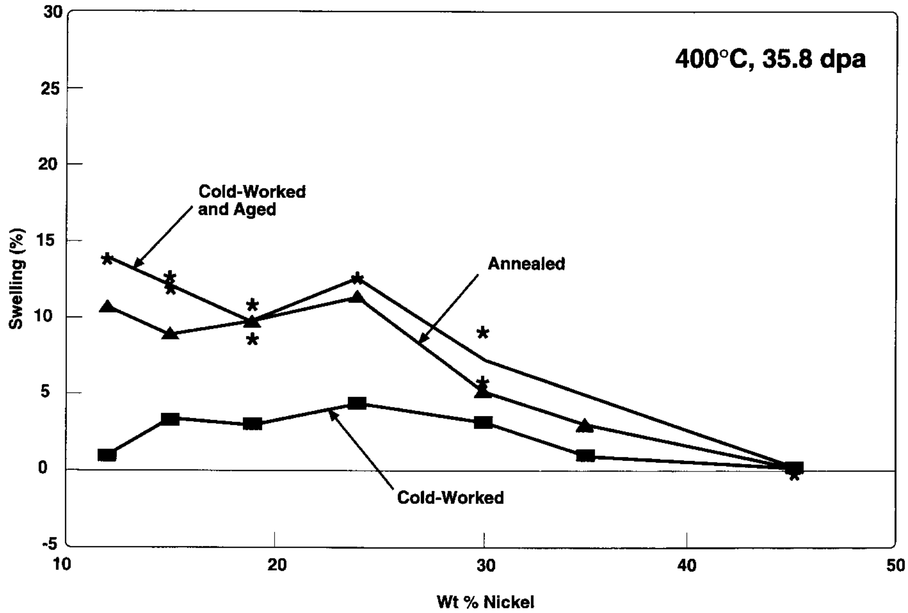

Fig. 3. Swelling observed in Fe–15Cr–Ni alloys irradiated in FFTF-MOTA to 35.8 dpa at 400°C.

sured densities of the two specimens at each combination of composition and starting condition agreed within ±0.16%, the average is plotted. Otherwise both measurements are plotted, providing some indication of the reproducibility of the swelling behavior.

The following features of these data are considered to be significant.

(a) At 375°C and 12.3 dpa, the swelling is relatively low and independent of nickel content in the annealed case. While cold-working reduces swelling to near-zero levels, aging of cold-worked material restores most of the swelling to levels comparable to that of the annealed specimens. At the two lowest nickel levels swelling is actually increased above that of annealed specimens.

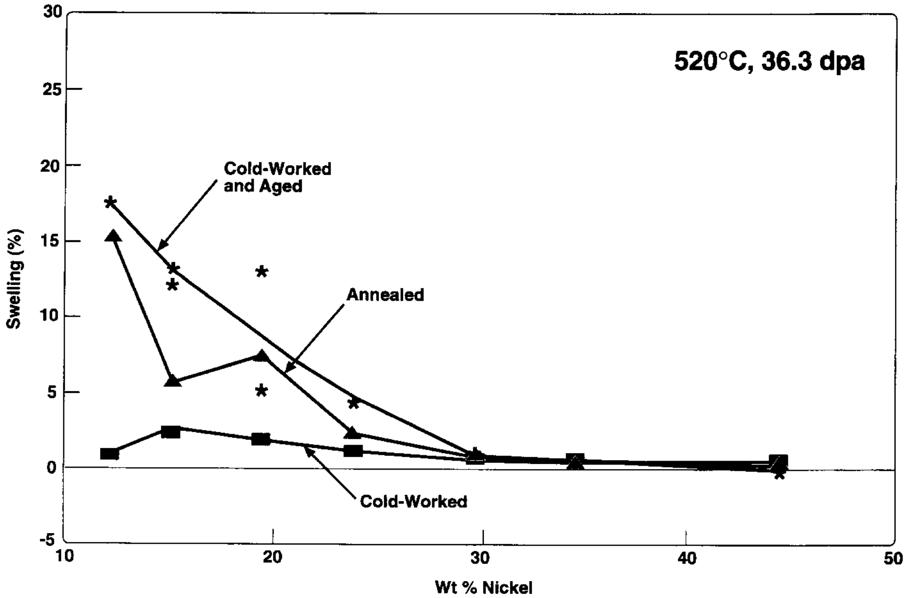

Fig. 4. Swelling observed in Fe–15Cr–Ni alloys irradiated in FFTF-MOTA to 36.3 dpa at 520°C.

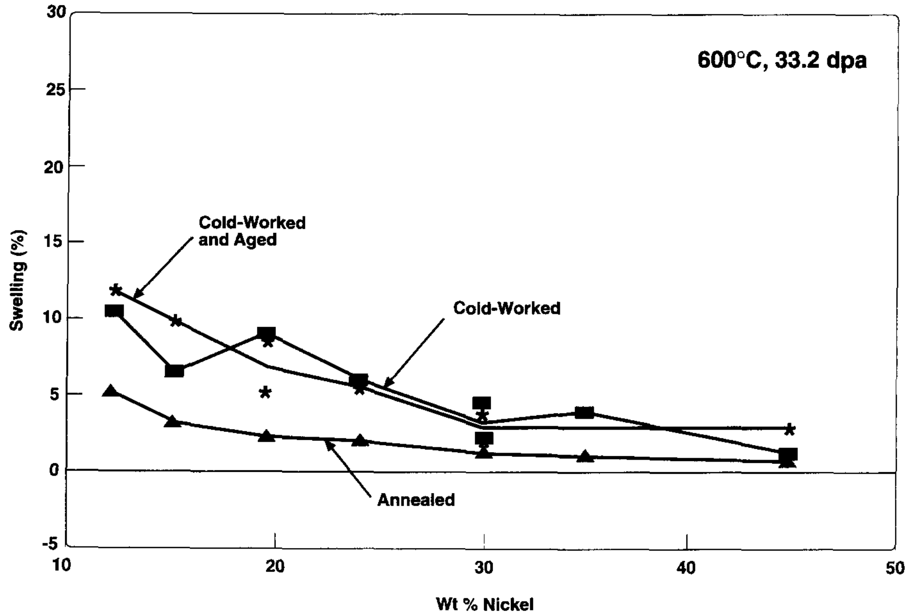

Fig. 5. Swelling observed in Fe–15Cr–Ni alloys irradiated in FFTF-MOTA to 33.2 dpa at 600°C.

(b) At 400°C and 35.8 dpa the expected dependence of swelling on nickel content is observed, with a plateau at low nickel content followed by decreasing swelling with increasing nickel level. Cold-working reduced swelling at all nickel levels, and aging restores the swelling to levels comparable to or slightly higher than that of the annealed specimens.

(c) At 520°C and 36.3 dpa, the dependence of swelling on nickel content is stronger as would be expected from the results of earlier studies. Cold-working decreases swelling but aging restores and even increases swelling when compared with the swelling of annealed material.

(d) At 600°C and 33.2 dpa, however, cold-working leads to higher swelling than that of annealed material at all nickel levels. Aging appears to cause no significant further increase in swelling.

(e) When compared to the swelling behavior exhibited in EBR-II, the swelling in FFTF at comparable dpa levels is in general much lower.

(f) For all solution-annealed and cold-worked specimens, the swelling of the two specimens for each material agreed within the accuracy limit of the measurement technique. This indicates that the swelling phenomenon itself is very reproducible for a given starting state and set of irradiation conditions. For reasons that are not currently understood, however, significant variations in swelling were often observed in the two specimens that were cold-worked and aged. This variability may reflect the onset of intermetallic or ferritic phase instability, which may be sensitive to grain boundary diffusion during aging and to minor differences in aging-induced stress relaxation that can occur in each specimen.

5. Discussion

Both irradiation series examined in this study have shown that under some conditions, cold-working can increase void swelling compared to that of annealed material. Depending on the irradiation and material conditions, aging after cold-working can also restore swelling to higher values. While the general trends of the two experiments with respect to nickel content, temperature and starting conditions are similar to each other and to the results of earlier studies, there are still significant differences in behavior that suggest the participation of other unrecognized variables in the initiation of the swelling process.

The peak in swelling observed at 12–15% nickel after EBR-II irradiation is inconsistent with the behavior observed in earlier EBR-II studies [15], where swelling either increased monotonically with decreasing nickel content, or exhibited a plateau at low nickel levels and relatively low temperatures. It is felt that this difference may arise from the fact that the specimens in the earlier study were better annealed, 1030°C for 0.5 h and air-cooled, as opposed to the 975°C for 10 min and water-quenched treatment used in the current study. One possible consequence of this difference is the fact that unirradiated specimens appeared to be increasingly magnetic at lower nickel levels, as qualitatively indicated by their interactions with the electron beam during microscopy examination, as shown in Table 3. The cold-worked and aged specimen at 12% Ni did not exhibit magnetic behavior in the unirradiated condition, while the annealed and the cold-worked specimens were found to be very magnetic. As shown in Table 3, there is an apparent correlation of magnetic behavior with the peaked swelling behavior versus nickel content observed at 425°C.

Another clue to the effect of cold-work at the lower nickel level can be seen in Fig. 6. Note that while the preirradiation density of both annealed and the cold-worked and aged materials exhibit the expected monotonic dependence on nickel content, the cold-worked material possessed significantly lower density, but only for 12% Ni alloy. Both of these observations suggest the onset of strain-induced ferritic instability in the 12% Ni alloy.

In an earlier review [7], it was shown that the role of cold-work in increasing swelling for relatively soft metals and alloys arose from the establishment of stable dislocation networks at higher temperatures where such networks are relatively unstable and slow to develop from radiation alone. The influence of preirradiation aging in increasing swelling was thought to lie in the development of stable polygonal cellular dislocation networks, with relatively dislocation-free internal areas allowing void nucleation to occur somewhat earlier within the cells. Preliminary results of microscopy examination confirm that this interpretation is valid for the Fe–Cr–Ni alloys irradiated in the AA-14 experiment. Each of the several aged Fe–Cr–Ni alloys examined was found to have retained a relatively stable dislocation cell structure that would be expected to remain stable during irradiation at 400–600°C, at least during the void incubation stage.

While it is known from other studies conducted in EBR-II that Fe–Cr–Ni ternary alloys tend to eventually swell at 1%/dpa in the range 400–600°C, the transient regime proceeding the 1%/dpa regime is known to be dependent on the nickel level and temperature [4]. The smallest transient regime observed at low nickel levels and low irradiation temperatures in previous EBR-II studies was on the order of ~ 10 dpa. From the current EBR-II experiment it appears that the annealing temperature and other details of thermomechanical treatment may also affect the duration of the transient regime.

The current EBR-II irradiation experiment at 425°C would be expected to yield a maximum swelling on the order of 21% at 31 dpa if the incubation period was ~ 10 dpa, which is in rough agreement with the 19 to 24% observed in the annealed condition in the 12–25% nickel range. For the cold-worked and aged specimens at 400°C, however, 30% swelling was reached at 12% Ni, implying that the transient regime was on the order of 1 dpa or less. The 25–28% swelling observed at 500°C in both the annealed and the aged specimens at 37 dpa also implies that the incubation period (3–6 dpa) appears to have been significantly reduced from the ~ 10 dpa observed in the earlier EBR-II studies.

Table 3
Qualitative magnetic response of unirradiated Fe–15Cr–XNi alloys as observed during microscopy

	wt% nickel			
	12	15	30	35
Non-magnetic	CWA	CW CWA	CW CWA	n.o.
Magnetic	SA CW	SA	n.o.	CW CWA

n.o. = annealed specimens were not observed.

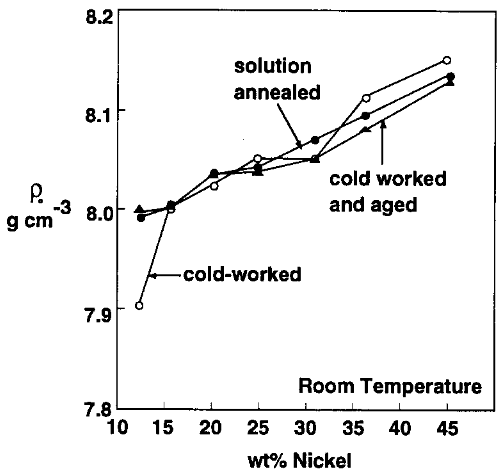

Fig. 6. Density at room temperature of unirradiated Fe–15Cr–Ni alloys as a function of nickel content and starting condition.

Since it appears possible to reduce the incubation period to near-zero by introducing a stable dislocation structure, this implies that development of a stable dislocation structure is the major prerequisite for attaining the 1%/dpa swelling rate in simple Fe–Cr–Ni alloys that do not contain solutes such as Si or P. Any other factors that influence void incubation, such as irradiation temperature and nickel level, must therefore operate as secondary determinants of the duration of the incubation period. The dependence of void nucleation on nickel content has been shown to probably arise not only as a consequence of the influence of nickel concentration on vacancy diffusivity, a dependence that is very similar to a change in temperature [15,17], but also from the concurrent dependence of dislocation bias on nickel content [18,19]. Both of these variables exert their strongest influence on void nucleation.

In the current FFTF irradiation series, however, the maximum swelling of annealed material at 425 and 520°C never exceeds 12 or 15%, respectively, at ~ 36 dpa. Aging increases the swelling only a little. This implies that the transient regimes in FFTF were larger than 10 dpa, approaching 15–24 dpa. The incubation periods at 600°C in FFTF were also obviously larger than those in EBR-II.

This suggests that the void nucleation process in FFTF must be more restricted. Although it is not possible at this time to specify the exact nature of the restriction, the answer may be in the very different temperature/flux histories of the two experiments, especially in their approach to full power and their shut-down history. A recent group of studies has shown that the details of temperature and flux histories in these periods can have a large impact on the nucleation and the maintenance of void and dislocation loop microstructure in typical irradiation experiments [20]. Also, whereas the temperature history in FFTF is quite constant during the major portion of the irradiation, the temperatures in EBR-II are not only more variable but tended to fall slowly throughout the irradiation, a condition that probably makes it slightly easier to nucleate voids.

6. Conclusions

Contrary to the previous conventional wisdom [1–6], it appears that cold-working does not always decrease void swelling, but may actually increase it in conditions where void nucleation is relatively difficult. In relatively soft, solute-free Fe–Cr–Ni alloys, for example, void nucleation is most difficult at higher irradiation temperatures and higher nickel levels. Cold-working provides a dislocation network that otherwise would be difficult to form in a fully annealed structure by irradiation alone under these conditions. When aging of cold-worked microstructures produces a relatively stable dislocation cell structure, it appears to be possible to nucleate and grow voids even more easily. Under some conditions, the transient regime can be reduced by cold-working and aging to almost 0 dpa.

Superimposed on the influence of the dislocation network are other factors which influence void nucleation. These are irradiation temperature, nickel content, annealing conditions and the details of flux–temperature history. All of these factors combine in solute-free Fe–Cr–Ni alloys to determine the duration of the transient regime of void swelling. As shown in all earlier studies by the lead author, the major variability of swelling lies in the duration of the void incubation and transient regimes, and not in the steady-state swelling rate. Applying that observation to the current results, it appears that cold-working and subsequent aging under some circumstances significantly shorten the incubation period of swelling.

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