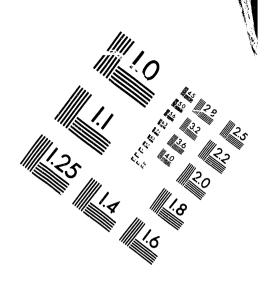
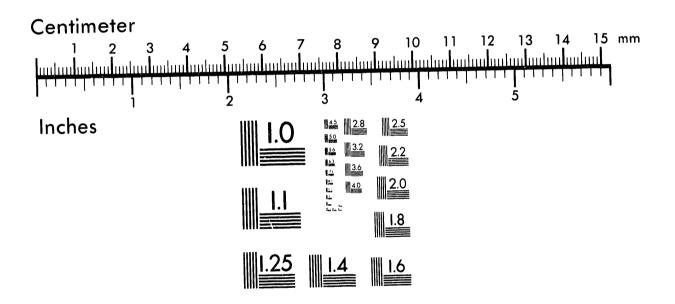


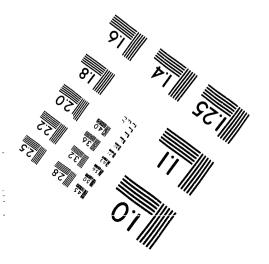


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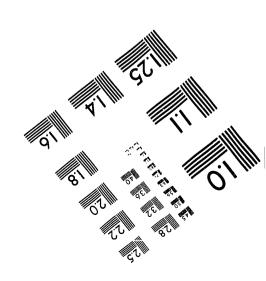
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EVOLUTION OF MICROSTRUCTURE IN FACE CENTERED CUBIC METALS DURING IRRADIATION: A REVIEW

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### Abstract

When fcc metals and alloys are irradiated at elevated temperatures, they tend to evolve toward saturation microstructures that are independent of the starting state of the metal and the early details of irradiation history. This leads to property changes and rates of dimensional change that also eventually become independent of the starting state. The evolution of microstructure in complex alloys, especially during the transient regime, is usually determined by the complex interaction of many microstructural and microchemical processes. The more complex the alloy, the more difficult it is to identify and define the separate influence of each participating mechanism. The use of irradiation studies conducted on simple metals or model alloys assists in understanding the behavior of alloys of engineering relevance. A review of such studies shows that a number of prevailing perceptions of radiation-induced microstructural evolution are not universally correct.

Key words: irradiation, charged particles, neutrons, saturation, microstructure, swelling, irradiation creep, mechanical properties, face centered cubic metals and alloys.

### Introduction

Radiation-induced changes in dimensional stability or mechanical properties of metals have been shown in many studies to be directly related to concomitant changes in microstructure. Many of these studies have been conducted on relatively complex alloys developed for engineering applications. In most cases, however, the microstructural evolution in these alloys is determined by the outcome of a competition between a large number of microstructural and microchemical mechanisms, subsets of which often work in opposition to each other. The outcome can change in favor of one mechanism or subset of mechanisms over others in response to variations in environmental or materials variables. In some cases, the dominant variable may be one that is relatively uncontrolled and even unrecognized by the experimenter as exerting an influence on the experiment.

If one desires to isolate and study the action of one particular mechanism, it is best to use a simple metal or model alloy, thereby reducing the number of competing mechanisms. This paper reviews the results of a variety of irradiation studies conducted on relatively simple fcc metals and model alloys, and then relates the insights derived from these studies to the explanation of phenomena that have been observed in more complex alloys of engineering relevance. Two themes will recur throughout this paper. First, a number of generally accepted statements of conventional wisdom that were derived from complex alloys have been found to be only partially correct and often are not universally applicable to all fcc metals and alloys. Second, there is a tendency in each metal or alloy to evolve toward a saturation or quasi-steady state microstructure defined primarily by the irradiation

temperature and displacement rate but not by the starting condition of the metal or alloy. Since void growth is often the dominant feature of microstructural evolution in fcc metals, this paper will focus first on void swelling and its various consequences.

# Influence of Starting State on Void Swelling

There is a very general perception, developed primarily from research on stainless steels, that cold-working a metal prior to irradiation always reduces swelling, exerting a monotonic but diminishing influence 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. Studies of relatively simple metals and alloys show, however, that the role of starting dislocation density and dislocation arrangements, as well as their interactions with variables such as helium generation rate, are much more complex than previously assumed.

Figure 1 shows that the swelling of well-annealed pure nickel exhibits a strong dependence on irradiation temperature at both low and high helium generation rates[1]. This strong dependence on temperature has been observed in other studies[2], most of which employed only annealed material. In one recent study, however, it was shown that 30% cold-working actually increased swelling, especially at higher temperatures, and almost erased the temperature dependence of swelling[3,4], a trend also observed in a few other studies[2]. Note in figure 2, however, that aging at 650°C for 10 hrs after cold-working further increased swelling, rather than producing some intermediate behavior

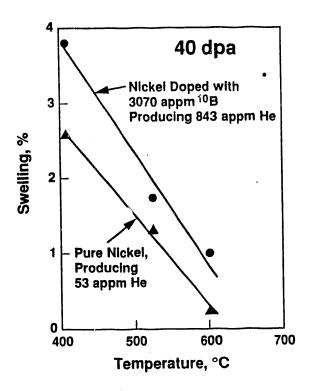


Figure 1. Swelling observed in well-annealed pure nickel and boron-doped nickel irradiated in FFTF/MOTA to 40 dpa[1].

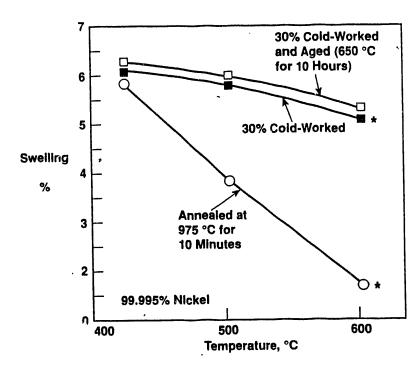


Figure 2. Swelling of pure nickel in various starting states observed after irradiation to 12-14 dpa in EBR-II. The higher level of swelling of annealed nickel compared to that of Fig. 1 is a result of a higher starting density of dislocations in the EBR-II study[3].

between that of the annealed and cold-worked states. Stubbins and Garner[3] showed that cold-working, especially when followed by aging, leads to a relatively stable dislocation cell structure that resists the eventual collapse of the dislocation density that always occurs in nickel[2]. When collapse occurs, swelling tends to saturate, as shown clearly in figure 3. Collapse occurs more quickly at higher temperatures. At relatively low irradiation temperatures, the higher microstructural densities produced during irradiation prolong the period before collapse occurs and thus cold-working has little or no effect on swelling. Figure 4 presents an example of such behavior[5]. Note that the swelling again appears to be independent of temperature over the range studied and that impurities tend to delay the onset of swelling. Harbottle and Silvent also showed that a reduction in the purity led to a lower initial swelling rate at 350°C in the SILOE reactor, which they demonstrated to result from a higher dislocation density arising from enhanced loop nucleation [6].

The tendency of cell structures developed by cold-work and aging to enhance swelling was also demonstrated by Horsewell and Singh[7], who showed that the presence of grain boundary dislocation networks, and especially subgrain walls, tend to significantly accelerate the onset of swelling in neutron-irradiated aluminum at 120°C (figure 5). The acceleration effect was found to be maximized at an intermediate cell size. Risbet and Levy also showed that swelling of neutron irradiated aluminum at 55°C and low fluence levels increased with cold-work and was maximized at 27.5% cold-work[8].

In a higher fluence experiment conducted at 50°C, van Witzenburg and Mastenbroek[9] showed that the onset of swelling in pure aluminum was not very sensitive to cold-work level and cell size compared to that observed at 120°C. Swelling eventually tended to saturate at a level that was strongly dependent on cold-work level, however, first increasing and then decreasing swelling

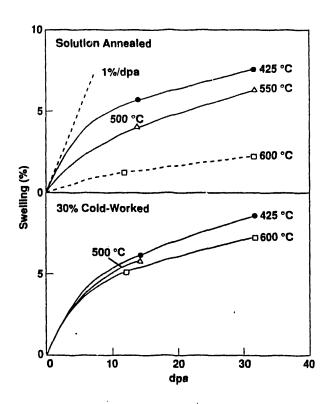


Figure 3. Tendency toward saturation of swelling in pure nickel during irradiation in the EBR-II experiment shown in Figure 2[4].

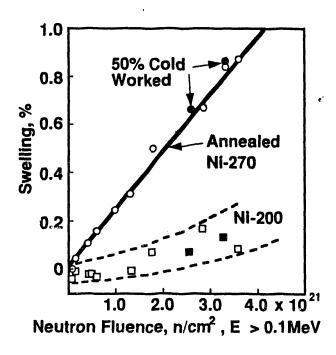


Figure 4. Lack of influence of 50% cold-work on swelling of nickel alloys irradiated at relatively low temperatures in EBR-II to  $\leq$  1.8 dpa. Ni-270 and Ni-200 are 99.98 and 99.6% pure nickel, respectively[5].

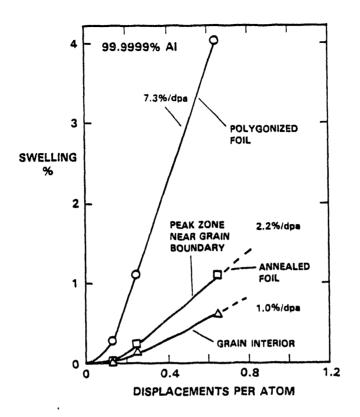


Figure 5. Influence of dislocation substructure on swelling of pure aluminum irradiated at 120°C in the DR-3 reactor[7].

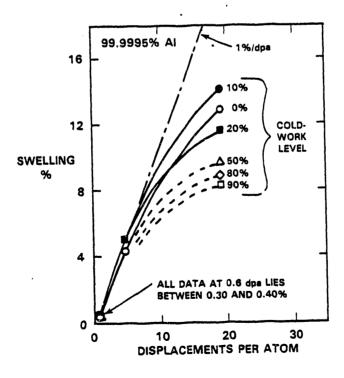


Figure 6. Influence of cold-work level on swelling of pure aluminum irradiated at 50°C in the HFR Petten reactor[9].

(figure 6). It is not clear, however, whether the lower saturation value developed at higher cold-work levels arose due to a cell size effect or to a dislocation collapse mechanism resulting from the release of strain energy stored in the cell network at higher cold-work levels.

Another possible factor contributing to saturation in aluminum at these higher fluence levels may be related to the formation of significant amounts of silicon by transmutation of aluminum, reaching ~0.4 at% Si at 2.3 x 10<sup>26</sup> n m<sup>-2</sup> (E>0.1 MeV). Silicon existing either prior to irradiation or formed by transmutation has been shown by Farrell[10] to delay or suppress, respectively, the swelling rate in aluminum irradiated at 55°C. This suppression appears to arise from the formation of solid silicon shells on yoid surfaces[11]. Silicon is insoluble in aluminum and also forms precipitates as well as yoid shells, as shown in figure 7.

The tendency for increasing levels of cold-work to first accelerate the onset of swelling and then to suppress it has also been observed by Leffers and coworkers[12] in electron-irradiated pure copper, as shown in figure 8. Garner and coworkers[13] showed that 40% cold-working somewhat delayed the onset of swelling in Cu-5Ni irradiated with fast neutrons at both 375 and 423°C, but did not change the eventual swelling rate.

Copper also transmutes strongly during neutron irradiation, forming significant amounts of nickel and zinc and a smaller amount of cobalt at high neutron fluences[14]. These elements are soluble in copper, however, and do not appear to affect its steady-state swelling rate, as shown in figure 9. Note that at 365-430°C Cu-5Ni also swells at the same rate as copper. Dislocation collapse does not occur in either metal in this temperature



Figure 7. Silicon-coated cavities observed by Farrell and coworkers in 1100 grade aluminum after irradiation at 55°C for three years in HFIR[11].

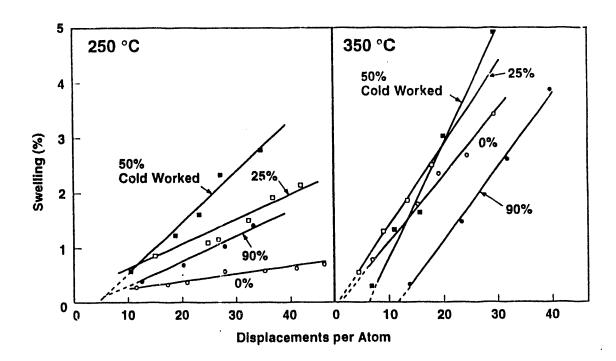


Figure 8. Effect of cold-work level on swe'ling of pure copper during 1.0 MeV electron irradiation[12].

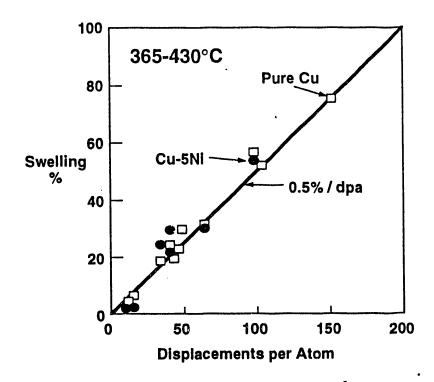


Figure 9. Swelling of pure copper and Cu-5 wt% Ni at temperatures of 365-430°C after irradiation in FFTF/MOTA[14].

range[14,15] and, therefore, swelling does not saturate or exhibit a strong dependence on temperature.

With the exception of a few early electron irradiation studies conducted on foils which were too thin[16], irradiations conducted with either charged particles or neutrons on Fe-Cr-Ni model alloys or more complex Fe-Cr-Ni commercial alloys have not led to saturation at any meaningful (<100%) swelling level[17,18]. In one proton irradiation study on 316 stainless steel, saturation appeared to develop at ~260%[19]. However, when very large and unrealistic levels of helium preinjection (1400 appm) were employed in 4 MeV Ni<sup>+</sup> ion bombardment of Fe-17Cr-16.7Ni-2.5Mo, saturation occurred at only 20% at ~170 dpa[20]. This experiment, however, is not considered to be a reactor-relevant experiment since the starting microstructure was overwhelmed with an unusually high density of very small helium bubbles.

In general, once swelling is initiated and regardless of alloy starting condition, all austenitic steels and all simple alloys based on the Fe-Cr-Ni system continue to swell to very large levels over a wide range of temperature at a maximum rate of 1%/dpa[17]. Instead of delaying swelling, Garner and coworkers have recently shown that cold-working often increases the neutron-induced swelling of simple Fe-Cr-Ni and Fe-Cr-Ni-P alloys, especially under conditions where void nucleation is relatively difficult, i.e., for relatively high nickel or phosphorus levels and high temperatures[21]. At lower nickel levels and temperatures, cold-working reduced swelling as expected from most results on typical 300 series stainless steels. More recently, however, another study by Garner and Edwards[22] demonstrated that aging of cold-worked. Fe-15Cr-Ni alloys prior to neutron irradiation led to swelling greater than

that of both cold-worked and annealed alloys for all nickel levels (12-45%) and temperatures studied, as shown in figure 10. This demonstrates once again that cold-work and associated dislocation cells formed on aging can strongly accelerate the onset of void swelling. The ~30% swelling observed in the Fe-15Cr-12Ni ternary alloy at 425°C (figure 10) is consistent with a 1%/dpa swelling rate and a transient regime of swelling that is between 0 and 1 dpa, significantly less than the ~10 dpa observed for the annealed alloy in earlier studies[17].

In more complex steels, aging can also strongly accelerate swelling, especially at relatively high cold-work levels and when high stress levels are applied. This effect is thought to arise from (a) the temperature and stress-assisted release of strain energy stored in heavily deformed dislocation networks, leading to recovery early in the irradiation[23], and (b) from the cold-work and stress-induced acceleration of the microchemical evolution, especially that associated with high temperature, thermally stable but sluggishly forming phases such as sigma; chi, and laves[24,25]. Figure 11 shows that aging not only accelerates the onset of swelling in cold-worked alloys but also in annealed alloys, thus demonstrating the separate effect of aging on microchemical evolution[26].

In complex steels, cold-working almost always delays swelling with the suppression most likely arising from the influence of the initially high dislocation density ( $\geq 10^{11}~{\rm cm}^{-2}$ ) in temporarily retarding long range solute diffusion and, therefore, the microchemical evolution. Transmutation is very low in typical Fe-Cr-Ni alloys and plays no significant role in the microstructural or microchemical evolution.

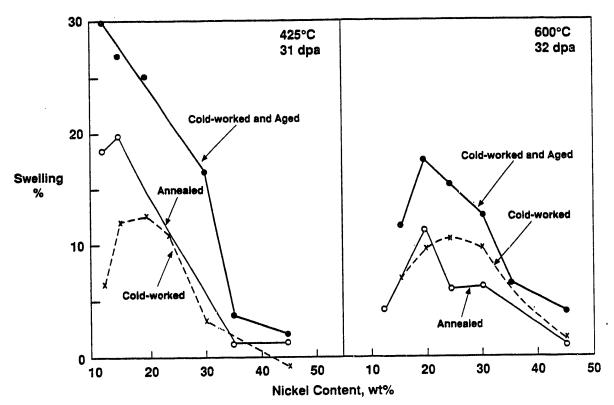


Figure 10. Influence of nickel content and starting state on swelling of Fe-15Cr-Ni alloys irradiated in EBR-II[22]. Annealed at 975°C for 10 min, aged at 650°C for 10 hrs after 30% cold-working.

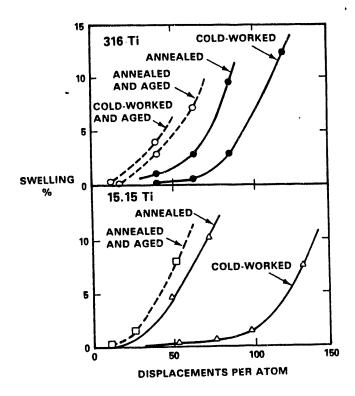


Figure 11. Effect of thermomechanical starting state on void swelling of two stainless steels in PHENIX at 500°C[26].

### Dislocation Evolution During Irradiation

The starting dislocation microstructure is not maintained during irradiation. A common feature of the preceding studies is the tendency for the dislocation microstructure to evolve toward a saturation or quasi-steady state level that is independent of the starting state. In relatively soft materials, especially at higher irradiation temperatures where Trank loops nucleate at low densities, it is often difficult to develop a stable dislocation network starting from a fully annealed state. In soft alloys with relatively large stacking fault energies, such as nickel and aluminum, even established dislocation networks will collapse to very low levels, with dislocations falling into the voids themselves and causing a saturation of swelling[3].

In model Fe-Cr-Ni alloys[27-29] and various 300 series stainless steels[30,31], the stacking fault energy is relatively low and the saturation density of network dislocations does not collapse and has been shown to be  $6\pm3\times10^{10}$  cm<sup>-2</sup>, relatively independent of starting state, temperature, displacement rate, He/dpa ratio and most other important variables. (The saturation density is slightly dependent on solute content and precipitation, however, with softer Fe-Cr-Ni alloys lying toward the lower end of the  $6\pm3\times10^{10}$  band.) This saturation process involves an order of magnitude reduction in the dislocation density of typical cold-worked steels and a comparable or larger increase in the density of annealed steels[32,33], as shown in figures 12 and 13. The bulk-averaged dislocation densities derived from the data presented in figure 13 exhibit much less scattering[33] than do measurements acquired by microscopy and therefore make it easier to observe the dependence of dislocation density on other variables. Typical densities determined by microscopy[32,34,35] are shown in

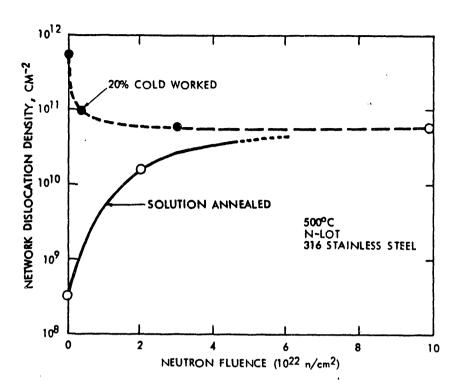


Figure 12. Saturation of network dislocation density in both 20% cold-worked and annealed AISI 316 after irradiation in EBR-II at 500°C[32].

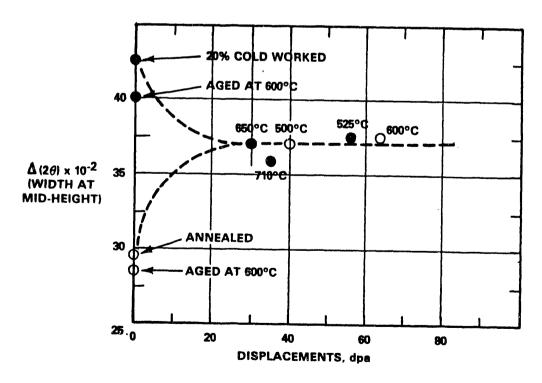


Figure 13. Independence of dislocation density in AISI 316L on starting condition and temperature after irradiation in the RAPSODIE reactor as measured by x-ray time broadening of the (311) austenite reflection[33].

figures 14-16. The scatter in these data represents both the local heterogeneity of dislocation structure in cold-worked materials and the difficulty of measuring them by microscopy.

There is a general perception, however, that the neutron-induced saturation density of network dislocations is moderately or even strongly dependent on temperature[36,37]. This perception arose primarily from the tendency of early papers from the United Kingdom to ambiguously report the total dislocation density, without specifying it as such. These data included both network and loop line length. When the first subsets of the data shown in figure 16 were published[38] they were designated only as the "line dislocation density." Another contribution to the perception of a strong temperature dependence of line length arose from the early publication of relatively low fluence data for solution-annealed steel[39]. The annealed steel had not yet approached the saturation state, especially at higher temperatures.

The loop density and its associated line length are strongly dependent on irradiation temperature[32] as shown in figure 17, and therefore it is the loop line length that accounts for the apparent strong temperature dependence usually attributed to the network dislocation component.

Another factor that contributes to the perception of a temperature-dependent saturation density for network dislocations is the fact that charged particle experiments experience strong temperature-dependent losses of dislocations due to surface proximity[30,31,33,40]. These losses lead to saturation densities that are moderately sensitive to temperature and displacement rate, as shown in figure 18. For a given displacement rate and temperature, however, the

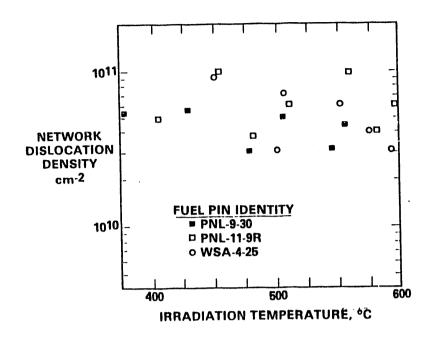


Figure 14. Network dislocation densities measured by microscopy in cladding from three 20% cold-worked AISI 316 fuel pins at doses ranging from 20 to 50 dpa (NRT) after irradiation in EBR-II[32].

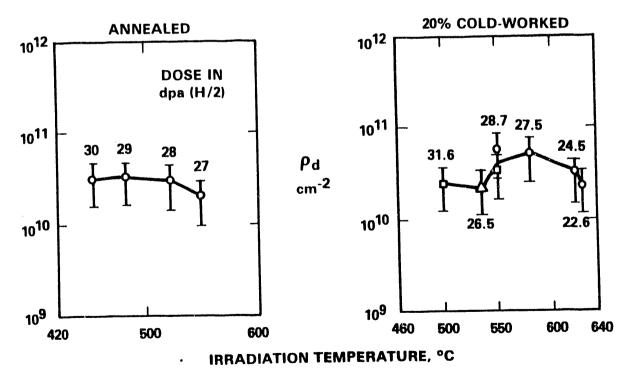


Figure 15. Network dislocation densities measured by microscopy in M316 cladding from two fuel pins irradiated in DFR[34].

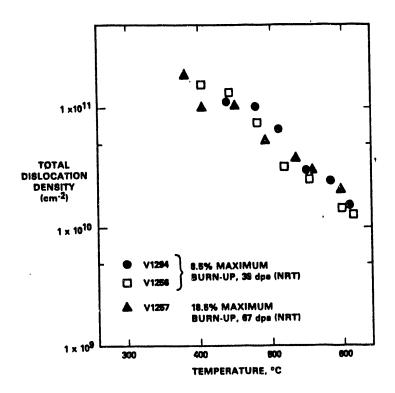


Figure 16. Total network and loop dislocation density observed in M316 cladding from three fuel pins irradiated in DFR[35].

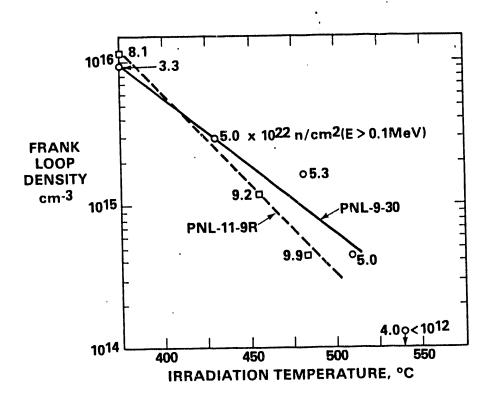


Figure 17. Frank loop densities observed in 20% cold-worked AISI 316 cladding from two fuel pins irradiated in EBR-II[32].

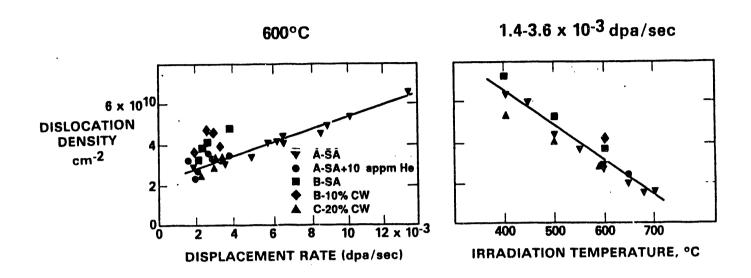


Figure 18. Surface influence on dislocation density in three heats of AISI 316L irradiated with 0.5 MeV Ni<sup>+</sup> ions, as reflected in the influence of temperature and displacement rate[?3]. The three heats of steel (A,B,C) are nominally identical with the largest variations in their nickel (12.8-13.7 wt%) content. SA = solution annealed, CW = cold-work.

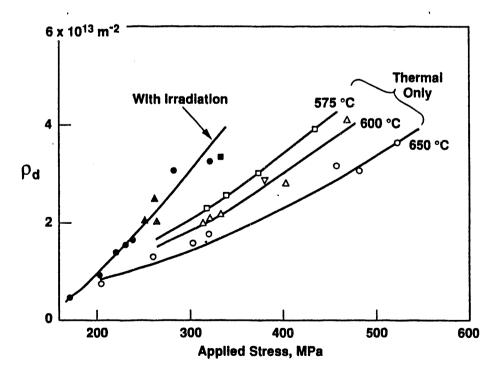


Figure 19. Effect of irradiation and stress level on saturation dislocation density during thermal creep of annealed 03Kh20N45M4BCh[43]. The irradiations were conducted in SM2 at  $575-600^{\circ}C$  and in RBT at  $650^{\circ}C$ , both at stress levels above the yield strength.

saturation state exhibits the same type of behavior observed in neutron irradiation experiments[30,31]. Note in figure 18 that the saturation density is not very sensitive to heat-to-heat variations, helium preinjection or coldwork level.

Ion-bombarded foils which are swelling also experience a stress state that encourages dislocation losses to the surface[41,42]. The effect of applied and internal stresses on dislocation and loop evolution and void swelling is also quite pronounced, but will not be covered in detail in this paper. It is sufficient to point out that imposed stress states alter the distribution of Burger's vector's of both loops and network dislocations. Stress also increases the density of dislocation loops and accelerates the onset of void nucleation. A detailed summary of stress effects on microstructural evolution is presented elsewhere[42].

It is important to note, however, that most stress effects studies are conducted at stress levels below the yield strength, such that network dislocations and dislocation loops interact primarily via their competition for point defects. When such experiments are conducted above the yield strength, however, glide occurs and there is a strong physical interaction between dislocations and loops that changes the nature of the saturation states of both components. Figure 19 shows that the saturation density of network components in such experiments is lower than the ~6x10<sup>10</sup> cm<sup>-2</sup> value observed at lower stresses, but the saturation state is now strongly stress dependent. The independence of temperature and displacement rate is still preserved, however [43]. Another facet of such experiments is that polygonized cell structures develop [43,44]. The dislocation velocities are increased by stress and tend to interact with and annihilate loop nuclei, thus leading to a reduction in loop density [44], as shown in Figure 20. This

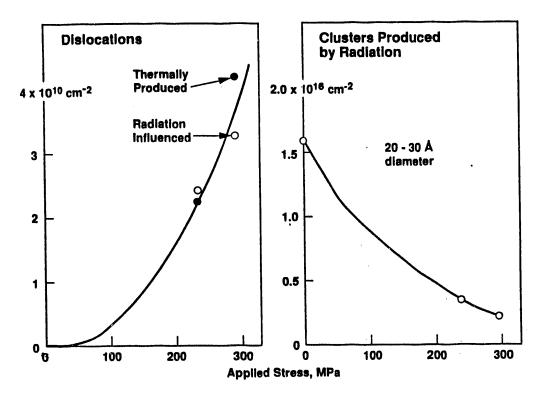


Figure 20. Influence of irradiation and stress levels above the yield strength on dislocation and loop microstructure of annealed 0Kh16N15M3B at  $5.8x10^{20}$  n cm<sup>-2</sup> and  $530^{\circ}C[44]$ .

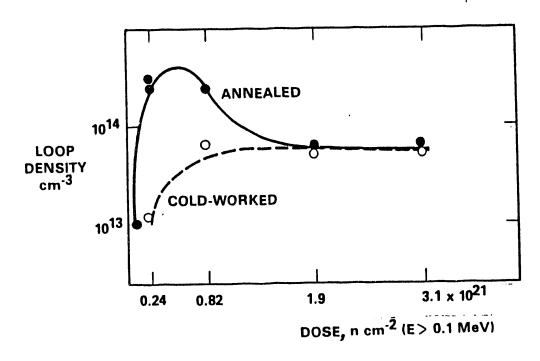


Figure 21. Frank loop evolution in pure aluminum irradiated at 55°C in the SILOE reactor[8].

reduction is in contrast to the increase in loop density with increasing ctress observed for experiments conducted below the yield strength [42].

The data shown in figures 16 and 17 demonstrate that Frank loops also approach a saturation or quasi-steady state condition since there is no significant change in loop density or size distribution after the network dislocation density reaches its final state. It is often observed, however, that the loop density can approach the equilibrium state from either above or below its final value. As shown in figure 21, annealed pure aluminum overshoots the saturation level, reflecting the sessile and therefore relatively unreactive nature of Frank loops, which must grow large enough to interact to form mobile dislocations[45], which in turn interact with each other and thereby reduce their number. In cold-worked materials all loops are continuously subject to unfaulting interactions with mobile dislocations and therefore the saturation level is usually approached from below. Aluminum appears to be typical of all fcc metals in this respect.

# Void Evolution During Irradiation

The difference between mobile and immobile microstructural components becomes much more important for voids, which are completely sessile. The saturation level of voids is, to the first order, a very strong and relatively well-defined function of irradiation temperature and displacement rate under typical neutron irradiation conditions, but it is possible that atypical levels of variables which strongly affect void nucleation (helium, oxygen, non-isothermal temperature history) can produce a metastable state than can be quite persistent. If, however, the metastable void density is far above the optimum level at a given temperature and displacement rate, convergence to the optimum level will eventually occur[46-50], as shown in figure 22. In this

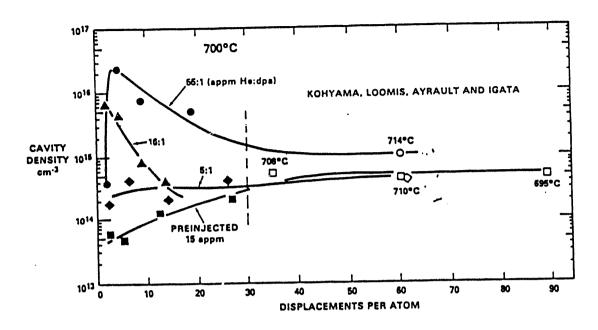


Figure 22. Void densities observed in annealed Fe-20Ni-15Cr irradiated with 3 MeV Ni<sup>+</sup> ions at ~700°C using a wide range of helium injection conditions[46-48].

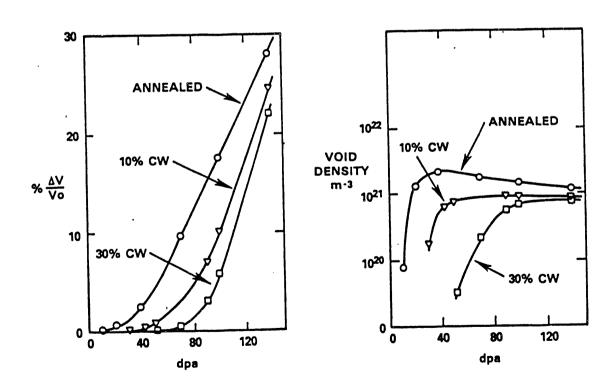


Figure 23. Effect of cold-work on swelling and dislocation density of Fe-15Cr-14.6Ni-2.6Mo-0.8Nb-0.22Si during 1.0 MeV  $\rm Cr^+$  irradiation at 650°C[49].

case, the reduction in void density occurs via coalescence, a much slower mechanism compared to that involving mobile microstructural components.

In cases where void nucleation is delayed by cold-work or solute addition, the void saturation level is usually relatively independent of these variables, and saturation can be approached from both above and below, as shown in figures 23 and 24.

## Macroscopic Consequences of Saturation

When the various components of the microstructure reach their respective saturation states, the metal under irradiation exhibits a quasi-steady state behavior. When heavily cold-worked nickel is irradiated with deuterons while under stress, for example, the strain rate quickly falls as the dislocation density declines. Then the strain rate reaches a constant value (figure 25) as the dislocation saturation microstructure is established[51]. Later, when void swelling begins, the creep rate will increase in proportion to the instantaneous swelling rate. This subject is covered in more detail elsewhere[52-54]. Since the parametric dependencies of irradiation creep are only the stress and the swelling rate, irradiation creep need not be covered separately in this paper.

As shown in figure 26, the neutron-induced swelling rate of different stainless steels eventually approaches the 1%/dpa swelling rate observed in model ternary Fe-Cr-Ni alloys[17,55], independent of composition and starting condition, as well as irradiation temperature[18,56]. As shown in figures 27 and 28, the yield strength of the alloy also reflects the attainment of saturation independent of starting state, both at temperatures where voids do not develop[57], and at temperatures where voids form relatively easily[58].

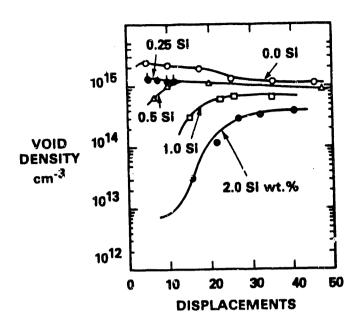


Figure 24. Influence of silicon level on void densities induced by electron irradiation in Fe-15Cr-13Ni-0.9Mn at 700°C[50].

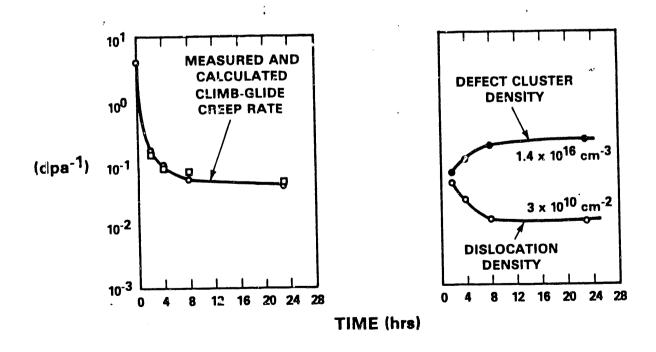


Figure 25. Relationship of creep rate to microstructure in 95% cold-worked nickel irradiated with 22 MeV D' ions at 242°C with a uniaxial stress of 345 MPa[51].

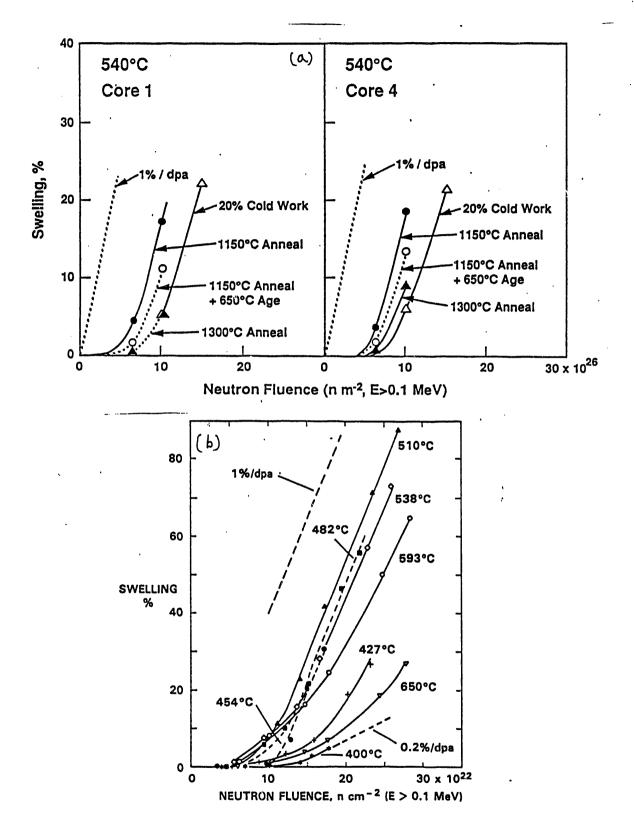


Figure 26. Swelling observed in a) various thermomechanical variations of two nominally similar heats of 20% cold-worked FFTF steels irradiated at 540°C[56] and in b) another 20% cold-worked heat irradiated at various temperatures[18], both in EBR-II.

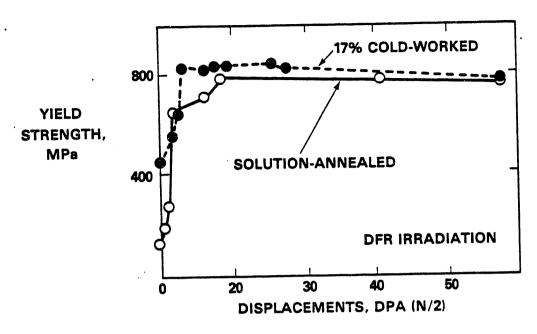


Figure 27. Evolution of yield strength in M316 stainless steel irradiated at 300°C in DFR[57]. Voids do not form easily at this temperature.

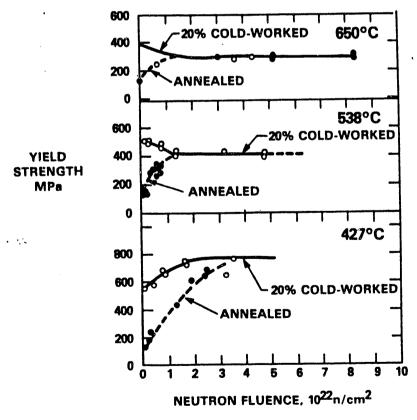


Figure 28. Evolution of yield strength in AISI 316 stainless steel irradiated in EBR-II at temperatures within the void swelling regime[58].

Note that at higher irradiation temperatures, the reduction of dislocations induced by cold-work is not completely offset by the addition of loops, precipitates, and voids. Thus the saturation state is approached from above and the cold-worked material softens.

There appears to be one important exception to the saturation concept presented to this point. If the irradiation temperature is low enough, such that dislocation mobility and the associated self-annihilation are strongly restricted, then annealed and cold-worked structures may not approach the same saturation level [59]. An example is shown in figure 29.

The evolution of swelling and yield strength changes in complex steels has also been shown to involve a strongly temperature-dependent, and sometimes flux-dependent, microchemical evolution during the transient regime, involving a wide variety of precipitates[17,60-62]. While the precipitates further harden the matrix, they also significantly alter the matrix composition, an often very sluggish process in cold-worked material, but one which strongly influences void nucleation[17,63]. Therefore, it becomes increasingly difficult to separate the relative roles of the precipitates themselves and the associated microchemical evolution of the matrix. The elements whose relative roles in solution and in precipitates are most in question in typical stainless steels appear to be nickel, silicon, phosphorus, titanium, and sometimes molybdenum[64-66]. Although many studies focus only on the direct effect of precipitates themselves, a recent series of papers on simple solutemodified Fe-Cr-Ni alloys has shown that three of these elements exert a strong and sometimes non-monotonic influence on void swelling while in solution, in addition to any role it plays upon precipitate formation[21,64,66]. Some elements, such as phosphorus, not only participate in precipitation but interact with both interstitials and vacancies, as well as exerting influence on dislocation mobility and interaction.

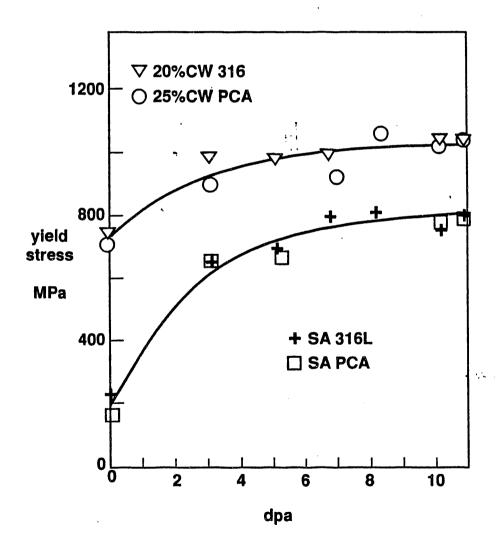


Figure 29. Hardening observed in two stainless steels irradiated in the HFIR, HFR, and R2 reactors at 250°C. The observed behavior is independent of helium/dpa rates in the range of 10 to 35 appm/dpa [59].

The recent work on simple solute-modified model alloys has also shown that the prevailing perception that each of these elements exerts only a monotonic influence on swelling, even when precipitates do not form, is not generally correct. (See figures 30 and 31). It has also been shown that once the multiple roles of such elements in simple alloys are defined, the non-monotonic influence of these elements on swelling can also be observed and explained in more complex alloys. An example is shown in figure 32 for phosphorus-modified 316 stainless steels[67]. Similar behavior has been observed for molybdenum in both model and commercial alloys[66,68].

There also appears to be a linkage between the microstructural and microchemical evolution. Not only can the saturation matrix composition be reached by a variety of precipitation paths, but some microstructural components such as Frank loops serve as nucleation sites for  $\gamma'$  and other radiation-induced phases[60].

# Influence of Other Variables on the Saturation State

Helium is often invoked to play a strong role in microstructural evolution, especially in fusion neutron environments. Until recently, it has been impossible to conduct a one-variable experiment to study the effect of helium generation rate on neutron-induced microstructural evolution and the associated changes in mechanical properties. A new technique involving isotopic doping allows a variation in helium generation rate without introducing changes in neutron spectrum, displacement rate, or chemical composition[69,70].

Using the reactor-produced isotope <sup>59</sup>Ni and applying this technique to three simple Fe-Cr-Ni alloys in both the annealed and cold-worked conditions, it has

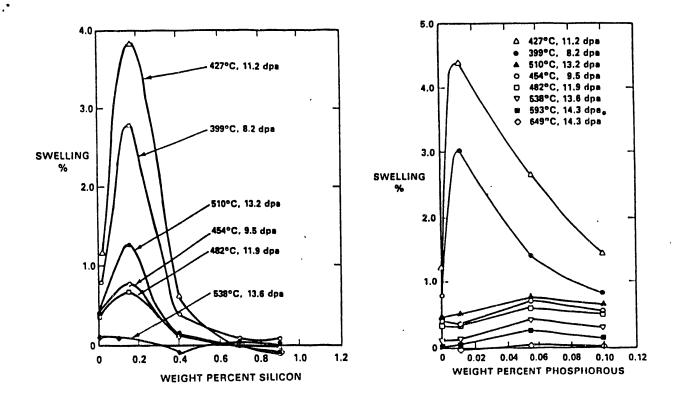


Figure 30. Non-monotonic swelling of phosphorus or silicon-modified Fe-15Cr-25Ni alloys after irradiation in EBR-II[64,65].

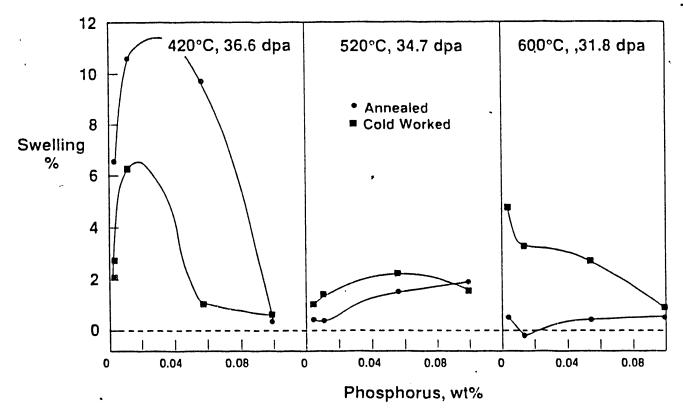


Figure 31. Non-monotonic swelling of phosphorus-modified Fe-15Cr-25Ni alloys after irradiation in FFTF[21,66].

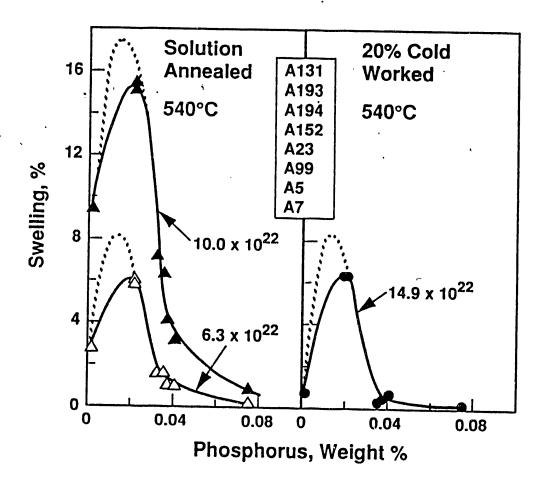


Figure 32. Swelling of Fe-16.2Cr-13.8Ni-2.5Mo-2.0Mn-1.5Si-0.2Ti-0.04C alloys with varying phosphorus levels after irradiation in EBR-II[67].

been shown that helium/dpa ratios at fusion-relevant levels alter the microstructural evolution somewhat but do not significantly affect the saturation levels of the yield strength or elongation[71,72]. An example is shown in figure 33. A similar conclusion has recently been reached for more complex steels from analysis of strength data from irradiation in various reactors[37,59].

In a companion paper in this conference it has been shown that variations in temperature history can have a pronounced effect on the early details of microstructural evolution, with large consequences on the transient regime of microstructural and microchemical evolution in particular[73]. In another portion of the <sup>59</sup>Ni experiment involving a comparison of microstructural evolution during both isothermal and non-isothermal irradiation, it was shown that detours in temperature were quickly forgiven, with the same saturation state eventually reached at a given final temperature for both types of temperature sequence[71,72].

When the microstructure becomes dominated by very large (>10%) levels of swelling, the saturation dislocation and precipitate states developed at lower swelling levels no longer dominate the response to radiation. This situation involves (1) the strong feedback effect that swelling exerts on irradiation creep via its influence on stress-affected dislocation evolution, (2) the onset of shear instabilities during mechanical deformation resulting from stress concentrations between voids, and (3) the impact of segregation at void surfaces on matrix composition and thereby on stability against martensite formation. This subject will be covered in detail in another paper[74].

## Conclusions

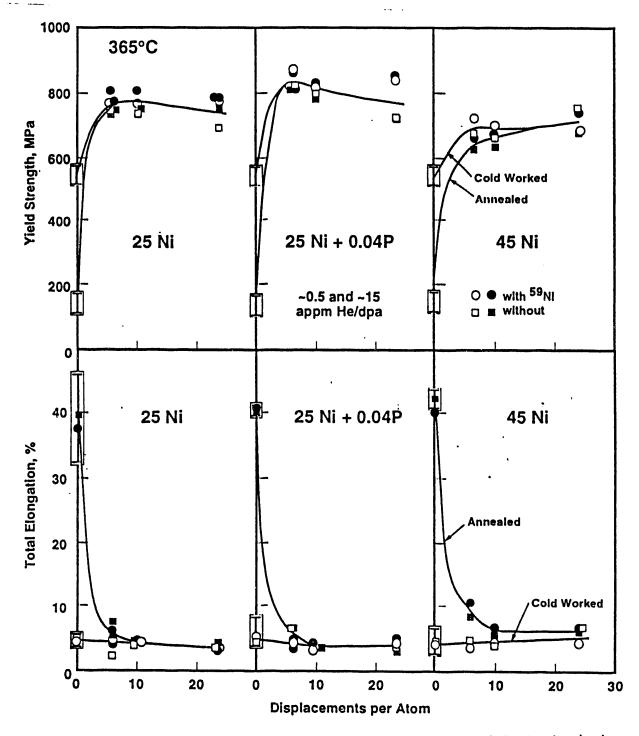


Figure 33. Influence of thermomechanical starting state and isotopic doping on yield strength and elongation of three Fe-15Cr-Ni alloys irradiated below the FFTF core at  $365^{\circ}C[71,72]$ . Filled and open symbols denote annealed and coldworked specimens, respectively. The average helium generation rates are  $\sim 0.5$  appm/dpa for undoped and  $\sim 15$  appm/dpa for doped specimens. The error bars at 0 dpa define the variation in properties observed prior to irradiation.

The evolution of microstructure in irradiated fcc metals and alloys is dominated by a tendency for each microstructural component to evolve toward a saturation level that is relatively dependent on irradiation conditions but not on the starting state of the material. This, in turn, leads to rates of change in swelling and creep deformation and also in the mechanical property evolution that reflect the existence of the saturation state. It appears that the saturation state is dependent primarily on recent irradiation history and that previous detours in temperature (and possibly other variables) are easily forgotten. These detours may have a pronounced effect on the duration of the transient regime of the evolution, however, especially when microchemical effects are dominant.

When attempting to separate the influence of the many microstructural and microchemical mechanisms that are involved in the radiation-induced evolution of microstructure, it is best to study relatively simple metals and alloys. From a review of such studies, it appears that some prevailing perceptions developed from irradiation of relatively complex alloys are either partially incorrect or generally inapplicable to all fcc metals. This is particularly true for the roles of preexisting dislocation microstructures and various microchemically important solutes. Once the various mechanisms are defined in detail using simple metals and alloys, however, it becomes easier to apply these insights to the study of more complex alloys.

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## References

- 1. T. Muroga and Y. Yoshida, Journal of Nuclear Materials 191-194 (1992) 1254-1258.
- 2. F. A. Garner, in: Fusion Reactor Materials Semiannual Progress Report DOE/ER-0313/8 (1990) pp. 125-135.
- J. F. Stubbins and F. A. Garner, Journal of Nuclear Materials 191-194
   (1992) 1295-1299.
- 4. F. A. Garner, in: Fusion Reactor Materials Semiannual Progress Report DOE/ER-0313/13 (1992) in press.
- 5. J. J. Holmes, Transactions ANS, 12 (1969) 701.
- 6. J. E. Harbottle and A. Silvent, in: Proceedings of Conference on Irradiation Behavior of Metallic Materials for Fast Reactor Core Components, Ajaccio, Corsica, 1979, pp. 391-397.
- 7. A. Horsewell and B. N. Singh, in: Radiation-Induced Changes in Microstructure: 13th International Symposium, ASTM STP 955, F. A. Garner, N. H. Packan and A. S. Kumar, Eds., American Society for Testing and Materials, Philadelphia, 1987, pp. 220-229.
- 8. A. Risbet and V. Levy, Journal of Nuclear Materials 46 (1973) 341-352.

- 9. W. van Witzenburg and A. Mastenbroek, Journal of Nuclear Materials 133 & 134 (1985) 553-557.
- 10. K. Farrell, in: Proceedings of Conference on Dimensional Stability and Mechanical Behavior of Irradiated Metals and Alloys, British Nuclear Energy Society, London, 1983, pp. 73-76.
- 11. K. Farrell, J. Bentley, and D. N. Braski, Scripta Met. 11 (1977) 243-248.
- 12. T. Leffers, B. N. Singh, and P. Barlow, RISØ National Laboratory Report RISØ-M-1937, May 1977.
- 13. F. A. Garner, D. J. Edwards, B. N. Singh, and H. Watanabe, in: Fusion Reactor Materials Semiannual Report, DOE/ER-0313/13 (1992) in press.
- 14. F. A. Garner, M. L. Hamilton, T. Shikama, D. J. Edwards, and J. W. Newkirk, Journal of Nuclear Materials 191-194 (1992) 386-390.
- 15. H. R. Brager, Journal of Nuclear Materials 141-143 (1986) 163-168.
- 16. J. J. Laidler and B. Mastel, in: Proceedings Thirtieth Annual Meeting of the Electron Microscopy Society of America, Claitors, 1972, pp. 680-681.
- 17. F. A. Garner, Journal of Nuclear Materials 122 & 123 (1984) 459-471.
- 18. F. A. Garner and D. S. Gelles, in: Effects of Radiation on Materials: 14th International Symposium (Vol. II) ASTM STP 1046, N. H. Packan, R.

- E. Stoller, and A. S. Kumar, Eds., American Society for Testing and Materials, Philadelphia, 1990, pp. 673-683.
- 19. A. S. Kumar and F. A. Garner, Journal of Nuclear Materials 117 (1983) 234-238.
- 20. N. H. Packan and K. Farrell, in: Effects of Radiation on Materials:

  Eleventh Conference, ASTM STP 782, H. R. Brager and J. S. Perrin, Eds.,

  American Society for Testing and Materials, 1982, pp. 885-894.
- 21. F. A. Garner, K. Miyahara, J. W. Newkirk, and H. Kinoshita, Journal of Nuclear Materials, (1992), 199, (1993) 132-142.
- 22. F. A. Garner and D. J. Edwards, "Factors Which Control the Swelling of Fe-Cr-Ni Ternary Austenitic Alloys", in: Fusion Reactor Materials Semiannual Progress Report DOE/ER-0313/14 (1993) in press.
- 23. F. A. Garner, M. L. Hamilton, C. R. Eiholzer, M. B. Toloczko, and A. S. Kumar, in: 16th International Symposium on Effects of Radiation on Materials, ASTM STP 1075, 1993, in press.
- 24. H. R. Brager and F. A. Garner, in: Effects of Radiation on Structural Materials, ASTM STP 683, J. A. Sprague and D. Kramer, Eds., American Society for Testing and Materials, 1979, pp. 207-232.
- 25. R. J. Puigh, A. J. Lovell, and F. A. Garner, Journal of Nuclear Materials 122 & 123 (1984) 242-245.

27. N. Sekimura, F. A. Garner, and R. D. Griffin, Journal of Nuclear Materials 191-194 (1992) 1234-1238.

and the second second second second

- 28. J. F. Stubbins and F. A. Garner, Journal of Nuclear Materials 179-181 (1991) 523-525.
- 29. J. F Stubbins and F. A. Garner, Journal of Nuclear Materials 191-194 (1992) 1300-1304.
- 30. F. A. Garner and W. G. Wolfer, in: Effects of Radiation on Materials:

  Eleventh Conference, ASTM STP 782, H. R. Brager and J. S. Perrin, Eds.,

  American Society for Testing and Materials, 1982, pp. 1073-1087.
- 31. F. A. Garner and W. G. Wolfer, University of Wisconsin Fusion Materials Report UWFDM-484, September 1982.
- 32. H. R. Brager, F. A. Garner, E. R. Gilbert, J. E. Flinn, and W. G. Wolfer, in: Radiation Effects in Breeder Reactor Structural Materials,
  M. L. Bleiberg and J. W. Bennett, Eds., The Metallurgical Society of AIME, 1977, pp. 727-755.
- 33. N. Azam, J. DeLaplace, and L. LeNaour, in: Proceedings of Conference on Irradiation Behavior of Metallic Materials for Fast Reactor Core Components, Ajaccio, Corsica, 1979, pp. 177-182.

- 34. C. Brown and G. Linekar, UKAEA TRG-Memo Report 6571, 1974.
- 35. C. Brown and E. J. Fulton in: U.S./U.K. Seminar on Cladding and Duct Materials, CONF-7910113, Richland, WA, Oct. 30 Nov. 2, 1979, pp. 11-29; also C. Brown, R. M. Sharpe, E. J. Fulton, and C. Cawthorne, in Dimensional Stability and Mechanical Behavior of Irradiated Metals and Alloys, British Nuclear Energy Society, London, 1983, pp. 63-67.
- 36. R. E. Stoller, Microstructural Evolution in Fast Neutron-Irradiated
  Austenitic Stainless Steels, Oak Ridge National Laboratory Report ORNL6430, December 1987. (See figure 2.7).
- 37. G. E. Lucas, in Proceedings of Montreux Conference on Time Dependence of Radiation Damage Accumulation and It's Impact on Materials Properties, Nov. 1992, Journal of Nuclear Materials, in press.
- 38. J. I. Bramman, C. Brown, J. S. Watkin, C. Cawthorne, E. J. Fulton, P. J. Barton, and E. A. Little, in: Radiation Effects in Breeder Reactor Structural Materials, M. L. Bleiberg and J. W. Bennett, Eds., The Metallurgical Society of AIME, 1977, pp. 479-507.
- 39. P. J. Barton, B. L. Eyre, and D. A. Stow, Journal of Nuclear Materials, (1977) 181-197.
- 40. N. Igata, A. Kohyama and S. Nomura, in: Effects of Radiation on Materials: Tenth Conference, ASTM STP 725, D. Kramer, H. R. Brager and J. S. Perrin, Eds., American Society for Testing and Materials, 1981, pp. 627-641.

- 41. F. A. Garner, G. L. Wire, and E. R. Gilbert, in: Radiation Effects and Tritium Technology for Fusion Reactors, CONF-750989, Vol. I., Gatlinburg, TN, 1975, pp. 474-490.
- 42. F. A. Garner and D. S. Gelles, Journal of Nuclear Materials 159 (1988) 286-309.
- 43. R. B. Grabova, Yu. D. Goncharenko, V. K. Gorbatov, V. M. Kosenkov, N. P. Losev, A. Ya. Rogozyanov, and B. V. Samsonov, Fiz. Metal. Metalloved.
  61, No. 6 (1986) 1164-1169.
- 44. S. I. Porollo, V. I. Scherbak, V. D. Dmitriyev, A. S. Kruglov, and Yu. M. Pevchikh, Fiz. Metal. Metalloved. 49, No. 6 (1980) 1302-1304.
- 45. N. Azam, L. LeNaour, and J. Delaplace, Journal of Nuclear Materials 49 (1973/74), 197-208.
- 46. F. A. Garner, H. R. Brager, M. L. Hamilton, R. A. Dodd, and D. L. Porter, Radiation Effects 101 (1986) 37-53.
- 47. S. C. Agarwal, G. Ayrault, D. I. Potter, A. Taylor, and F. V. Nolfi, Jr., Journal of Nuclear Materials, 85 & 86 (1979) 653-657.
- 48. A. Kohyama, B. A. Loomis, G. Ayrault, and N. Igata, in: Effects of Radiation on Materials: Twelfth Conference, ASTM STP 870, F. A. Garner and J. S. Perrin, Eds., American Society for Testing and Materials, 1981, pp. 277-296.

- 49. V. N. Voyevodin, B. V. Matvienko, B. N. Singh, and T. Leffers in :Proceedings: Dimensional Stability and Mechanical Behavior of Irradiated Metals and Alloys, Vol. 1, British Nuclear Energy Society, 1983, pp. 33-36.
- 50. B. N. Singh, T. Leffers, M. J. Makin, G. P. Walters, and A. J. E. Foreman, Journal of Nuclear Materials 103 & 104 (1981) 1041-1046.
- 51. D. J. Michel, P. L. Hendrick, and A. G. Pieper, Journal of Nuclear Materials 75 (1978) 1-6.
- 52. D. L. Porter, F. A. Garner, and G. D. Hudman, Journal of Nuclear Materials 179-181 (1991) 581-586.
- 53. F. A. Garner and D. L. Porter, Journal of Nuclear Materials 155-157 (1988) 1006-1009.
- 54. M. B. Toloczko, F. A. Garner, and C. R. Eiholzer, Journal of Nuclear Materials 191-194 (1992) 803-807.
- 55. F. A. Garner and H. R. Brager, in: Effects of Radiation on Materials:

  Twelfth International Symposium ASTM STP 870, F. A. Garner and J. S.

  Perrin, Eds., American Society for Testing and Materials, 1985, pp. 187-201.
- 56. F. A. Garner, J. F. Bates, and M. A. Mitchell, Journal of Nuclear Materials 189 (1992) 201-209.

- 57. K. Q. Bagley, J. W. Barnaby, and A. S. Fraser, in: Proceedings,
  Conference on Irradiation Embrittlement and Creep of Fuel Cladding and
  Core Components, British Nuclear Energy Society, 1972, pp. 143-153.
- 58. F. A. Garner, M. L. Hamilton, N. F. Panayotou, and G. D. Johnson, Journal of Nuclear Materials, 103 & 104 (1981) 803-808.
- 59. J. D. Elen, and P. Fenici, Journal of Nuclear Materials, 191-194 (1992)
- 60. F. A. Garner, in: Phase Stability During Irradiation, J. R. Holland, L. K. Mansur and D. I. Potter, Eds., The Metallurgical Society of AIME, Warrendale, PA, 1981, pp. 165-189.
- 61. E. H. Lee, P. J. Maziasz, and A. F. Rowcliffe, ibid, pp. 191-218.
- 62. W. J. S. Yang, H. R. Brager, and F. A. Garner, ibid, pp. 257-269.
- 63. F. A. Garner and W. G. Wolfer, Journal of Nuclear Materials 122 & 123 (1984) 201-206.
- 64. F. A. Garner and A. S. Kumar, in: Proceedings 13th International Symposium on Radiation-Induced Changes in Microstructure, Part I, ASTM STP 955, F. A. Garner, N. H. Packan, and A. S. Kumar, Eds., American Society for Testing and Materials, Philadelphia, 1989, pp. 289-314.
- 65. T. Muroga, F. A. Garner, and J. M. McCarthy, Journal of Nuclear Materials 168 (1989) 109-120.

- 66. F. A. Garner, M. A. Mitchell, and T. Lauritzen, in: Effects of Radiation on Materials: 16th International Conference, ASTM STP, 1175, 1993 (in press).
- 67. F. A. Garner and M. A. Mitchell, Journal of Nuclear Materials 187 (1992) 223-229.
- 68. F. A. Garner and H. R. Brager, Journal of Nuclear Materials 155-157 (1988) 833-837.
- 69. R. L. Simons, H. R. Brager, and W. Y. Matsumoto, Journal of Nuclear Materials 141-143 (1986) 1057-1060.
- 70. G. R. Odette, Journal of Nuclear Materials 141-143 (1986) 1011-1017.
- 71. M. L. Hamilton and F. A. Garner, Journal of Nuclear Materials 191-194 (1992) 1239-1243.
- 72. F. A. Garner, M. L. Hamilton, L. R. Greenwood, J. F. Stubbins, and B. M. Oliver, in: Effects of Radiation on Materials: 16th International Symposium, ASTM STP 1175, 1993, in press.
- 73. F. A. Garner, N. Sekimura, M. L. Grossbeck, A. M. Ermi, J. W. Newkirk, H. Watanabe, and M. Kiritani, in this conference.
- 74. F. A. Garner and M. B. Toloczko, Journal of Nuclear Materials,
  Proceedings of Montreux Conference on Time Dependence of Radiation
  Damage Accumulation and It's Impact on Materials Properties, Nov. 1992,
  in press.

- 66. F. A. Garner, M. A. Mitchell, and T. Lauritzen, in: Effects of Radiation on Materials: 16th International Conference, ASTM STP, 1175, 1993 (in press).
- 67. F. A. Garner and M. A. Mitchell, Journal of Nuclear Materials 187 (1992) 223-229.
- 68. F. A. Garner and H. R. Brager, Journal of Nuclear Materials 155-157 (1988) 833-837.
- 69. R. L. Simons, H. R. Brager, and W. Y. Matsumoto, Journal of Nuclear Materials 141-143 (1986) 1057-1060.
- 70. G. R. Odette, Journal of Nuclear Materials 141-143 (1986) 1011-1017.
- 71. M. L. Hamilton and F. A. Garner, Journal of Nuclear Materials 191-194 (1992) 1239-1243.
- 72. F. A. Garner, M. L. Hamilton, L. R. Greenwood, J. F. Stubbins, and B. M. Oliver, in: Effects of Radiation on Materials: 16th International Symposium, ASTM STP 1175, 1993, in press.
- 73. F. A. Garner, N. Sekimura, M. L. Grossbeck, A. M. Ermi, J. W. Newkirk, H. Watanabe, and M. Kiritani, in this conference.
- 74. F. A. Garner and M. B. Toloczko, Journal of Nuclear Materials,
  Proceedings of Montreux Conference on Time Dependence of Radiation

  Damage Accumulation and It's Impact on Materials Properties, Nov. 1992,
  in press.

- 57. K. Q. Bagley, J. W. Barnaby, and A. S. Fraser, in: Proceedings,

  Conference on Irradiation Embrittlement and Creep of Fuel Cladding and

  Core Components, British Nuclear Energy Society, 1972, pp. 143-153.
- 58. F. A. Garner, M. L. Hamilton, N. F. Panayotou, and G. D. Johnson, Journal of Nuclear Materials, 103 & 104 (1981) 803-808.
- 59. J. D. Elen, and P. Fenici, Journal of Nuclear Materials, 191-194 (1992) 766-770.
- 60. F. A. Garner, in: Phase Stability During Irradiation, J. R. Holland, L. K. Mansur and D. I. Potter, Eds., The Metallurgical Society of AIME, Warrendale, PA, 1981, pp. 165-189.
- 61. E. H. Lee, P. J. Maziasz, and A. F. Rowcliffe, ibid, pp. 191-218.
- 62. W. J. S. Yang, H. R. Brager, and F. A. Garner, ibid, pp. 257-269.
- 63. F. A. Garner and W. G. Wolfer, Journal of Nuclear Materials 122 & 123 (1984) 201-206.
- 64. F. A. Garner and A. S. Kumar, in: Proceedings 13th International Symposium on Radiation-Induced Changes in Microstructure, Part I, ASTM STP 955, F. A. Garner, N. H. Packan, and A. S. Kumar, Eds., American Society for Testing and Materials, Philadelphia, 1989, pp. 289-314.
- 65. T. Muroga, F. A. Garner, and J. M. McCarthy, Journal of Nuclear Materials 168 (1989) 109-120.

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