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Compositional and temperature dependence of void swelling in model Fe–Cr base alloys irradiated in the EBR-II fast reactor

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Abstract

A series of annealed and aged Fe–*x*Cr, Fe–12Cr–*y*C and Fe–12Cr–0.1C–*z*Mo model alloys were irradiated in EBR-II at eight temperatures between 400°C and 650°C and dose levels ranging from 35 to 131 dpa. Swelling-induced density changes observed in the binary alloys generally peaked at mid-chromium levels, with the chromium and temperature dependence expressed primarily in the duration of the transient regime. The steady-state swelling rate at the lower irradiation temperatures was much higher than previous estimates, reaching ~0.2%/dpa and possibly still climbing at higher neutron exposures. The dependence of swelling on molybdenum and carbon was more complex, depending on whether the temperature was relatively low or high. At temperatures of 482°C and above, the effect of carbon additions was very pronounced with swelling of Fe–12Cr jumping dramatically from near zero at 0.002% C to 6–10% at 0.1% C. This indicates that the major determinant of the composition and temperature dependence probably lies in the duration of the nucleation-dominated transient regime of swelling and not primarily in the steady-state swelling rate as previously envisioned. This raises the possibility that significant swelling may occur earlier in fusion and spallation neutron spectra where high gas generation rates may assist void nucleation.

1. Introduction

Until recently, it was thought that the steady-state swelling rate of iron-based bcc alloys was not only strongly dependent on composition but was also very temperature-dependent. In all cases, however, the steady-state swelling rate was thought to be lower by an order of magnitude compared to that of iron-based fcc alloys, which exhibit a steady-state swelling rate of ~1%/dpa, independent of composition and temperature in the range studied. The origin of these perceptions originally arose from irradiation in EBR-II of simple Fe–Cr–Ni ternaries examined by Garner and coworkers [1–5] and simple Fe–Cr binaries examined by Gelles [6–8], with both alloy series irradiated side-by-side in the very same packets. Gelles and coworkers carried this work further, irradiating these alloys to much higher exposures in the FFTF fast reactor [9–11], but in general preserving his previous EBR-II based conclusions.

In a recent review article by Garner et al. [12], however, it was shown that an error in dose assignments for bcc specimens in EBR-II at lower irradiation temperatures was responsible for the misperception of very low and variable swelling rates for bcc alloys. When the dose assignments were corrected, the bcc binary alloys were found to exhibit the same general dependence on temperature and composition exhibited by the fcc ternary alloys in the same packets. The steady-state swelling rate of bcc alloys were observed to reach at least ~0.2%/dpa. In the review, other evidence was shown that suggested that the swelling rate might still be climbing to higher values.

Additional conclusions reached in the review article were that the transient regimes of bcc alloys were in general much longer than those of fcc alloys, especially at higher irradiation temperatures and higher dpa rates.

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In particular, it was shown that even complex structural alloy such as HT9 and 9Cr–1Mo would eventually overcome nucleation difficulties and swell at higher rates [12,13]. A summary of many of these results is shown in Fig. 1.

Since these conclusions represent a significant departure from the previous perception of bcc behavior and have some potentially important implications for fusion applications of ferritic alloys, it is important to separately confirm these conclusions. It is also important to see if they extend to higher temperatures and to compositions more representative of structural bcc alloys. Therefore, similar but previously unexamined sets of specimens irradiated in the same EBR-II series were measured to determine their change in density.

2. Experimental details

Three series of Fe–xCr ($x = 3$, 6, 9, 12, 15, 18 wt%), Fe–12Cr–yC ($y = 0.002$, 0.1, 0.2 wt%) and Fe–12Cr–0.1C–zMo ($z = 0$, 1, 2, 3 wt%) model alloys were prepared in the form of 0.3 mm thick, 3.0 mm diameter microscopy disks in the annealed and aged (1040°C/1 h/AC + 760°C/2 h/AC) condition. These alloys were irradiated in stainless steel 'weeper' packets in contact with coolant sodium in EBR-II at eight temperatures between 400°C (±10°C) and 650°C (±20°C). Gelles and coworkers [6–8] have published more details of alloy preparation and irradiation technique.

Dose levels examined in this series depend on the temperature and ranged from 35 to 48 dpa at 400–454°C, 74 to 101 dpa at 482–510°C, and 121 to 131 dpa

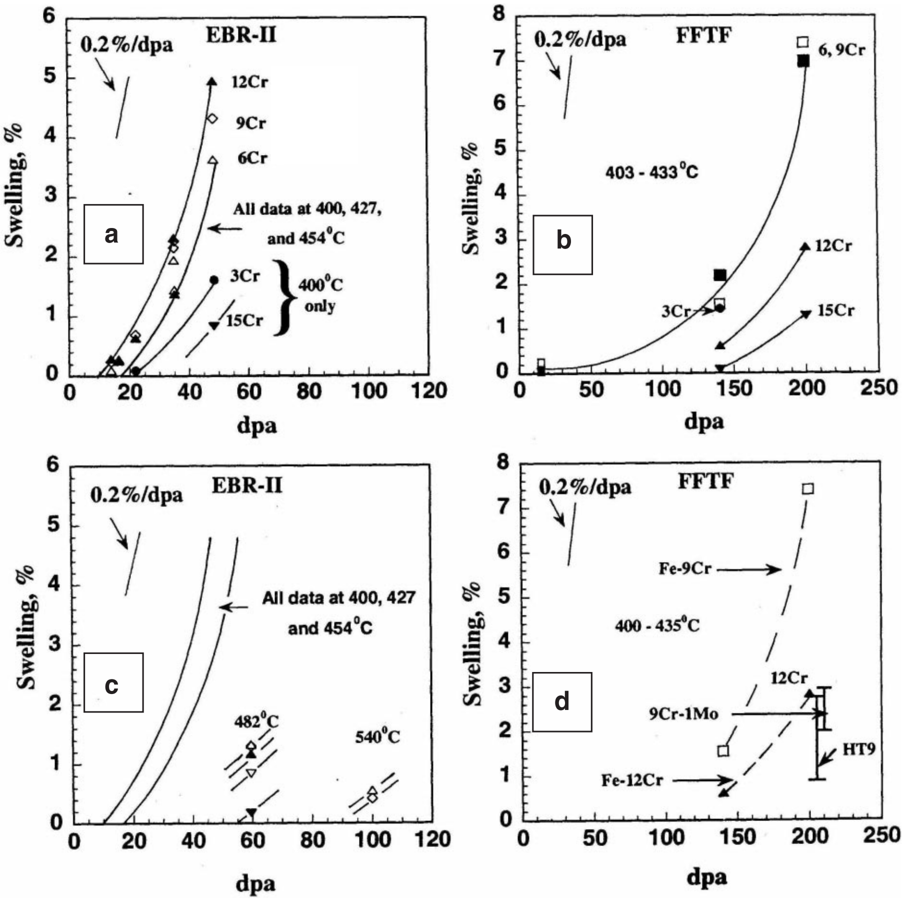

Fig. 1. (a) Compilation of swelling data of Fe–(3–15)Cr binary alloys irradiated in EBR-II (with corrected dpa levels) showing swelling rates on the order of ~0.2%/dpa, following transient regimes that are somewhat dependent on composition; (b) similar compilation in FFTF, showing swelling rates on the order of ~0.2%/dpa, following transient regimes that at low temperature are also somewhat dependent on composition, but which are very dependent on the different flux-temperature histories of the two reactors [12]; (c) at higher temperatures the transient regimes are longer in EBR-II; (d) also shown are HT9 (12% Cr) and 9Cr–1Mo irradiated in FFTF, showing that their swelling is relatively close to their binary equivalents [13]. The range of swelling values for these two alloys demonstrates the effect of stress to increase swelling, with the lowest swelling at zero stress and monotonically increasing with stress.

at 538–650°C, as shown in Fig. 2. Swelling measurements were performed using an immersion density technique known to be accurate to ±0.2% change in density.

3. Results

As shown in Fig. 2, swelling tends to peak at intermediate chromium levels, in agreement with the results of Gelles and coworkers [6–11]. Sometimes negative swelling values most likely represent densification associated with combined effects of segregation and formation of α' and σ precipitates [6–11,14], with precipitation most pronounced at higher dpa levels and higher chromium levels. Note that swelling appears to be occurring at all temperatures except 650°C. Preliminary microscopy examination of these specimens finds no voids at 650°C for any chromium level [14].

Fig. 3 shows that the addition of carbon to Fe–12Cr at temperatures below 454°C and doses below 48 dpa tends to monotonically decrease the swelling. At 482–650°C, however, an increase to 0.1% C leads to very large increases in swelling, although a subsequent increase to 0.2% C at 593–650°C reduces swelling.

Fig. 4 indicates that addition of molybdenum to Fe–12Cr–0.1C tends to decrease swelling strongly at low Mo levels, with some hint of smaller subsequent increase in swelling at higher Mo levels.

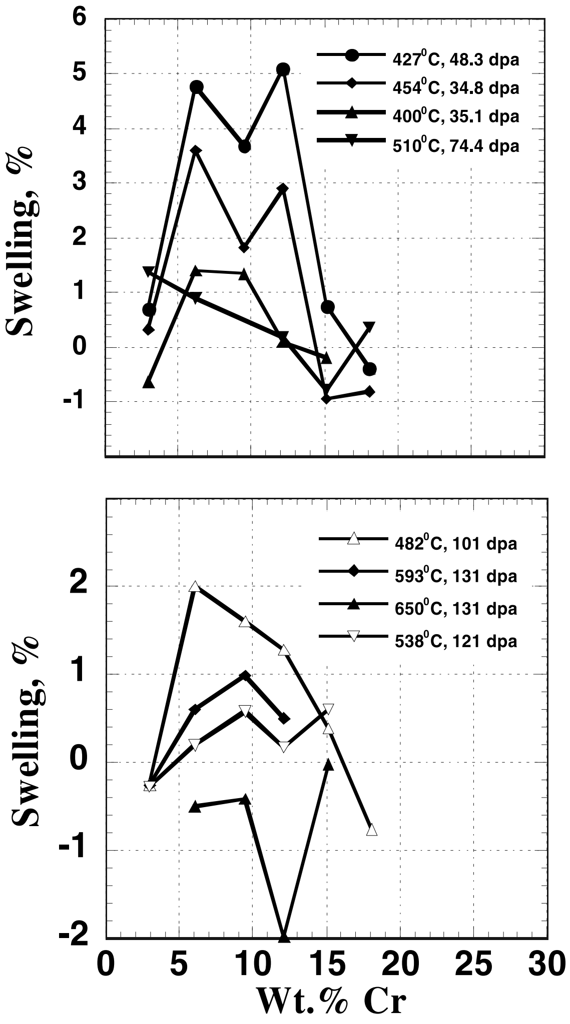

Fig. 2. Swelling of Fe–Cr binary alloys as a function of Cr level for various irradiation conditions in EBR-II.

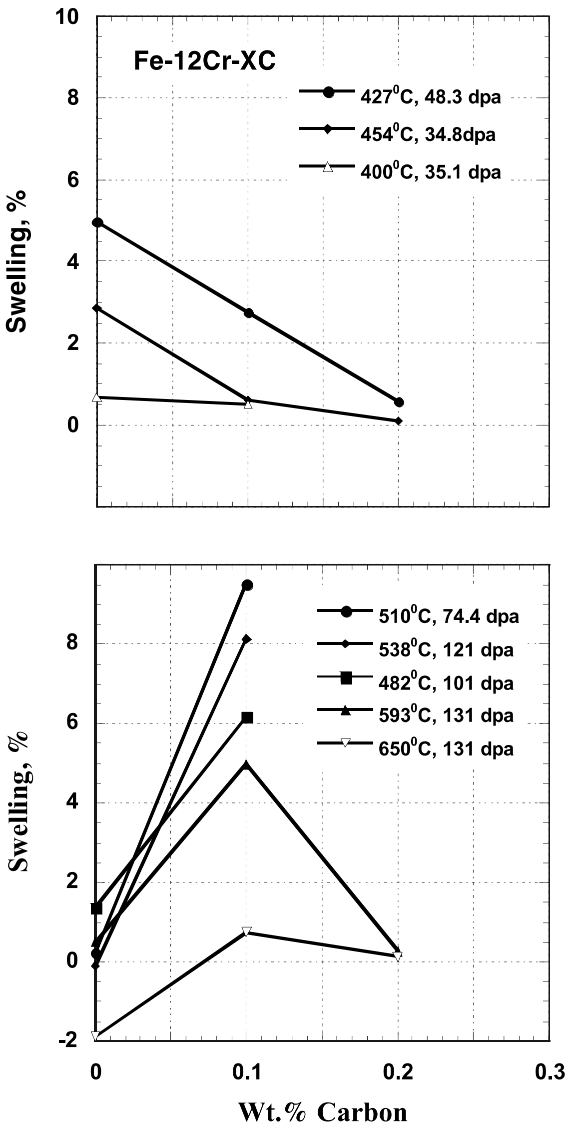

Fig. 3. Effect of carbon additions on swelling of Fe–12Cr.

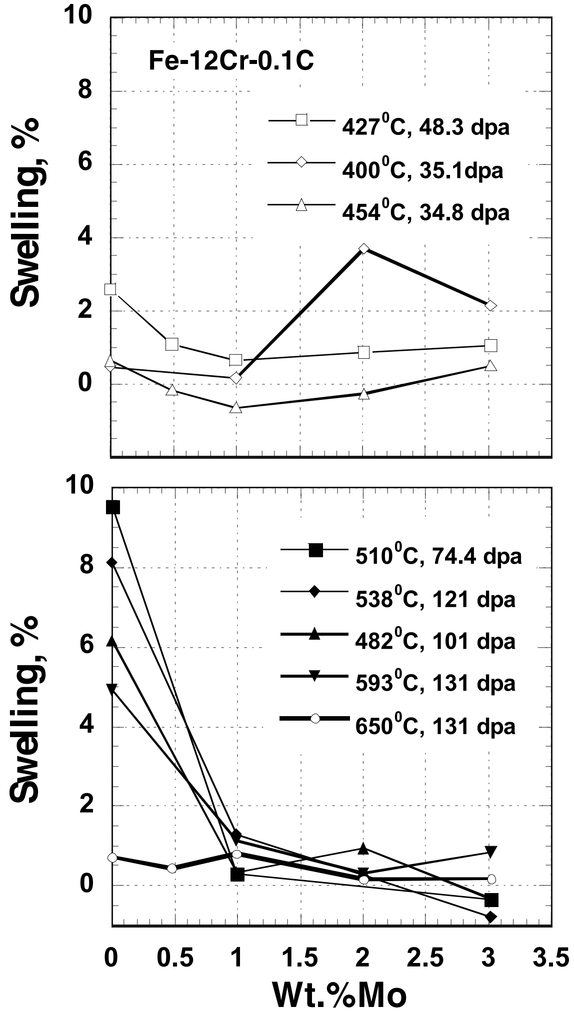

Fig. 4. Effect of molybdenum additions on swelling of Fe–12Cr–0.1C.

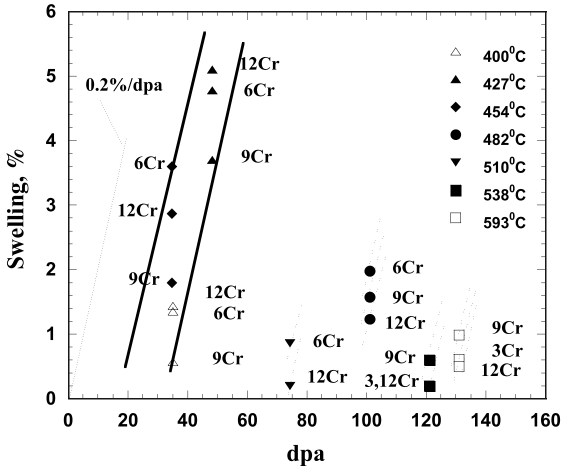

Fig. 5. Swelling of Fe–Cr binary alloys in EBR-II as a function of temperature and dpa level, showing similar trends to those in Figs. 1(a) and (b), but with swelling extending to much higher temperatures. The dotted lines at higher temperatures are used only for emphasis to separate the various data sets, with no explicit swelling rate implied.

4. Discussion

Garner and coworkers earlier showed that over a range of Cr (12–22%) and Ni (12–24%) levels and a range of relatively low temperatures, the annealed fcc Fe–Cr–Ni ternary alloys irradiated in this same EBR-II experimental series could be considered to swell in a narrow band as if they were almost independent of temperature and composition [1–3]. Changes in composition or temperature outside these ranges yielded strong increases in the transient regime of swelling but no change in the steady-state swelling rate. It was later shown that the swelling data of Gelles for bcc Fe–Cr alloys could be treated in the same way [12].

As shown in Fig. 5, the binary alloys examined in this experiment indeed appear to confirm this conclusion and once again support a steady-state swelling rate on the order of ~0.2%/dpa. A surprising result is that if one is prepared to wait long enough, then swelling in these fast reactors will eventually accelerate in bcc alloys at very high temperatures where swelling was never before anticipated. Only at 650°C has swelling not yet started.

Note, however, that the transient regime does not appear to increase monotonically with increasing temperature in Fig. 5. This is an illusion resulting from the differences in dpa rate at each temperature and arises from the large gradients in dpa rate across the relatively small EBR-II core. Garner and coworkers showed earlier that higher dpa rates and more stable temperatures found in FFTF strongly extend the transient regime of swelling [4,12]. When compared at comparable dpa rates, the transient duration of Fe–Cr in EBR-II indeed increases monotonically with temperature.

Fig. 3 shows that addition of 0.1% C to Fe–12Cr causes a dramatic increase in swelling at high temperatures compared with that of the binary alloy which strongly resists swelling. Even though the data are relatively sparce in this figure, it is difficult to visualize this increase except as a reduction in transient duration. Unfortunately, Gelles did not examine irradiated Fe–Cr alloys at such high temperatures, although his data at lower temperatures confirm the carbon-induced suppression of swelling for alloys observed in the low-temperature portion of our study [8].

The earlier low-temperature and lower-dpa results of Gelles also confirm the suppressive effect of molybdenum on swelling [6,8]. It is important to remember, however, that HT9 and 9Cr–1Mo both eventually began to swell at accelerated rates in FFTF at low temperature

[13], as shown in Fig. 1(d). Even more importantly, significant precipitation accompanies swelling in these alloys, removing and redistributing elements such as C and Mo [8,13]. The relatively strong effect of stress on swelling of HT9 and 9Cr–1Mo alloys also suggests a transient-dominated behavior [15,16].

The realization that the swelling resistance of bcc alloys resides primarily in delay of void nucleation, rather than an inherently low steady-state swelling rate has some strong potential implications for their future application to fusion devices and especially accelerator-driven spallation neutron devices. Irradiation testing of these alloys has proceeded only in neutron spectra that produce very low levels of helium and moderately low levels of hydrogen. The He/dpa ratios in fusion spectra are expected to be on the order of 5–15 and $\sim$150 appm/dpa in high-energy (>100 MeV) proton beams. The hydrogen generation rates are an order of magnitude greater in each case.

Given the demonstrated strong sensitivity of the transient duration of these binary alloys to the differences in flux and temperature history between EBR-II and FFTF (both low gas generation devices) shown in Fig. 1, it is prudent to anticipate that the temperature, flux and gas history experienced in unexplored high gas generation devices may lead to swelling behavior significantly different from that obtained in any currently used fission spectrum. Based on the known effects of gaseous transmutants, the onset of swelling will most likely be accelerated in fusion and spallation neutron environments.

5. Conclusions

The neutron-induced swelling of Fe–Cr binary alloys in fast reactors is characterized first by a transient regime whose duration depends on temperature, composition and dpa rate, and second a steady-state swelling rate that is at least $\sim$0.2%/dpa and possibly still climbing to higher levels as swelling continues. The relatively long transient duration at high temperatures is abruptly shortened, however, by the addition of carbon. Subsequent addition of molybdenum tends to return the transient to higher dose levels. Since the relative swelling resistance of bcc steels is now known to arise primarily from nucleation-dominated transient behavior, it appears that irradiation in spectra with high gaseous transmutation rates might lead to significantly higher than expected swelling levels in both model and structural alloys.

Acknowledgements

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