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Synergistic influence of displacement rate and helium/dpa ratio on swelling of Fe–(9, 12)Cr binary alloys in FFTF at ∼400 °C

F.A. Garner [a,*], D.S. Gelles [a], L.R. Greenwood [a], T. Okita [b], N. Sekimura [b], W.G. Wolfer [c]

[a] *Pacific Northwest National Laboratory, Structural Materials Res., P.O. Box 999, M/S P8-15, Richland, WA 99352, USA*
[b] *University of Tokyo, Tokyo, Japan*
[c] *Lawrence Livermore National Laboratory, Livermore, Japan*

Abstract

Contrary to the behavior of swelling of model fcc Fe–Cr–Ni alloys irradiated in the same FFTF-MOTA experiment, model bcc Fe–Cr alloys do not exhibit a strong dependence of swelling on dpa rate at ∼400 °C. This is surprising in that an apparent flux sensitivity was observed in an earlier comparative irradiation of the same Fe–Cr binaries conducted in EBR-II and FFTF. The difference in behavior between the two experiments is ascribed to the higher helium generation rates of Fe–Cr alloys in EBR-II compared to that of FFTF, and also the fact that lower dpa rates in FFTF are accompanied by progressively lower helium generation rates.

1. Introduction

In a previous publication it was shown that simple binary Fe–Cr alloys irradiated in the in-core region of FFTF at ∼400 °C required much higher dpa levels to approach the steady-state swelling condition than that required to reach this condition when the same alloys were irradiated in in-core regions of EBR-II, as shown in Fig. 1 [1]. Based on comparisons with various austenitic alloys, the different swelling behavior in the two reactors was interpreted to be a consequence of the large difference in swelling rates in the two reactors. For instance, simple Fe–Cr–Ni model austenitic alloys irradiated in FFTF over a wide range of dpa rates in a later experiment were shown to exhibit progressively longer transient regimes of swelling as the displacement rate increased [2].

It must be recognized, however, that comparative irradiations of simple Fe–Cr binary alloys conducted in two different reactors are potentially influenced by differences in other important spectral or operating variables, and the observed difference in transient behavior cannot confidently be ascribed simply to differences in dpa rate without eliminating the possible role of these other variables.

Since the transient regime of Fe–Cr–Ni austenitic alloys irradiated in FFTF showed such a clear dependence on dpa rate, it was natural to turn to another experiment included in the same FFTF irradiation series to assess the effect of dpa rate on ferritic alloys. Fortunately, the simple austenitic and ferritic alloys were both irradiated in the same comprehensive experiment.

2. Experimental details

Relatively pure Fe–9Cr and Fe–12Cr (at.%) with no added solute were prepared by arc melting from high purity Fe and Cr. The major measured impurity was

* Corresponding author. Tel.: +1-509 376 4136; fax: +1-509 376 0418.
E-mail address: frank.garner@pnl.gov (F.A. Garner).

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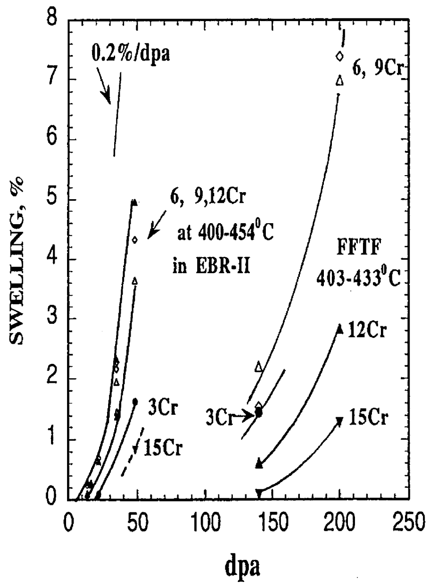

Fig. 1. Comparison of swelling of Fe–Cr binary alloys at ~400 °C in in-core regions of EBR-II and FFTF, as observed by Garner et al. [1].

carbon at ~180 wppm. The binary alloys were rolled to sheets of 0.25 mm thickness, cut into 3 mm disks, wrapped with Zr foil and annealed for 60 min at 900 °C in high vacuum.

Two sets of identical specimens are placed in sealed, helium-filled packets at each of seven different capsule positions of the materials open test assembly (MOTA), ranging from below the core to above the core of the fast flux test facility (FFTF). The packets in general contained four identical specimens of each of the two alloys. In many cases the Fe–Cr–Ni and Fe–Cr alloys were located side-by-side in the same packet. Two or more identically-loaded packets were placed in each capsule, with the dpa rate dependent on the axial location in MOTA.

With the exception of the below-core canister, the temperatures in MOTA capsules are actively controlled to ±5 °C of the nominal target, although the nominal target temperatures varied a little from capsule to capsule.

The first irradiation sequence occurred in cycle 11 of MOTA-2A for 2.58×10^7 s, and a subset of specimen packets was then removed. Other identical specimen packets continued in cycle 12 of MOTA-2B for 1.71×10^7 s. The dose rates in the various capsules ranged from 1.4×10^{-8} to 1.7×10^{-6} dpa/s. The dose levels attained by the specimens varied from 0.37 to 43.8 dpa in cycle 11 and an additional 0.24–24.0 dpa in cycle 12. Table 1 summarizes the irradiation conditions.

The starting and post-irradiation densities were measured using an immersion density technique known to be accurate to ±0.2% change in density. In some cases it was not possible to clearly identify and retrieve all four specimens, but in general there were at least two identical specimens measured in each capsule. Determination of microstructural evolution in these specimens using a transmission electron microscope has not yet been initiated.

3. Results

Fig. 2 shows the measured changes in density observed in the irradiation series. Surprisingly, the results are quite different than expected. There is no obvious effect of dpa rate for either of the two alloys. There is some small range of data scatter among nominally identical specimens, but with only a few exceptions, the individual data points from separate specimens are

Table 1
Irradiation conditions for Fe–Cr alloys irradiated in FFTF cycles 11 and 12 (MOTA-2A and MOTA-2B)

Dose rate, dpa/s		Dose, dpa		Temperature, °C	
#11	#12	#11	#11 and #12	#11	#11 and #12
1.7×10^{-6}	1.4×10^{-6}	43.8	67.8	427	408
7.7×10^{-7}[a]	9.6×10^{-6}	20.0[a]	34.5	390	387
8.2×10^{-7}[b]	8.4×10^{-7}	21.1[b]	28.8	430	424
3.2×10^{-7}[c]	3.5×10^{-7}	8.22[c]	13.1	373	373
1.5×10^{-7}	1.3×10^{-7}	3.87	6.12	430	431
4.6×10^{-7}	4.2×10^{-8}	1.18	1.91	434	437
1.4×10^{-8}	1.4×10^{-8}	0.37	0.61	436	444

Note that in three of the seven cases the specimens irradiated in both cycles did not experience completely identical conditions with single cycle packages.

[a] 6.8×10^{-7} dpa/s and 17.7 dpa in #11 for 2 cycle irradiation specimens.
[b] 5.4×10^{-7} dpa/s and 14.0 dpa in #11 for 2 cycle irradiation specimens.
[c] 2.7×10^{-7} dpa/s and 6.90 dpa in #11 for 2 cycle irradiation specimens.

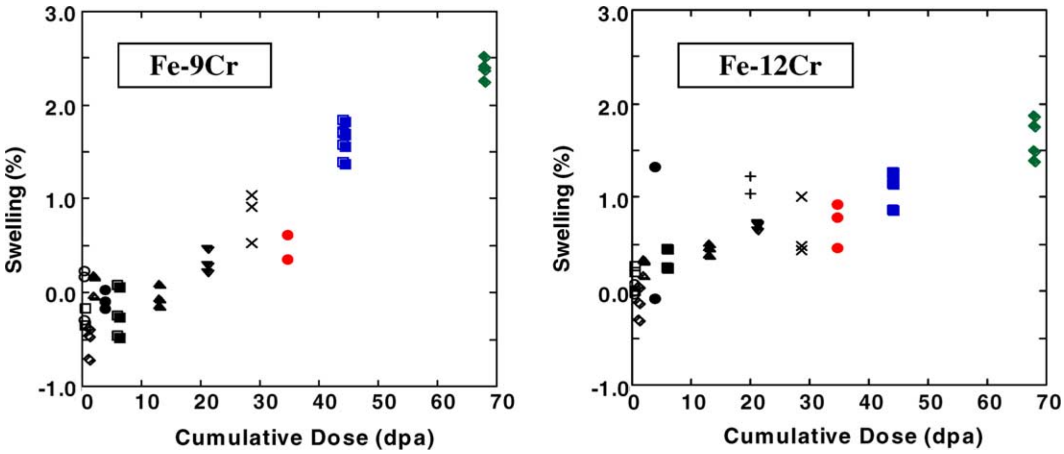

Fig. 2. Swelling of two simple model bcc Fe–Cr alloys in FFTF-MOTA at ∼400 °C, as observed in the current experiment, showing some early densification but no apparent dependence of swelling on dpa rate.

relatively consistent with each other, especially considering that voids are probably not the only contribution to the density change.

There appears to be some densification occurring in both alloys which is visible at the lowest dpa levels. Such densifications are usually the result of phase instabilities such as alpha-prime formation or carbide-induced densification [3,4], as discussed in the next section.

The swelling rate of the Fe–9Cr alloy appears to be ∼0.05%/dpa, significantly less than the ∼0.2%/dpa observed at steady-state in the earlier FFTF/EBR-II comparative experiment [1]. The swelling rate for the Fe–12Cr alloy appears to be slightly smaller. The failure to attain the anticipated swelling rate of ∼0.2%/dpa is thought to indicate that steady-state swelling has not yet been attained in this experiment. Since the Fe–Cr alloys in the earlier FFTF experiment conducted at high dpa rates required almost 200 dpa before approaching 0.2%/dpa, this is not a surprising result, but it was anticipated that at the lower dpa rates the swelling would be accelerated as observed in the austenitic alloys in the same experiment.

4. Discussion

The densification observed in this experiment probably cannot be ascribed solely to formation of the alpha-prime phase, since the densification in the 12Cr alloy is similar to or slightly less than that of the 9Cr alloy, although the formation of this phase should be at roughly twice as large at 12% Cr [3]. Another possibility is the densification in the 9Cr alloy as a consequence of an austenite to ferrite phase transformation upon cooling of the specimen during fabrication, followed by reversion to austenite during irradiation. Based on the Fe–Cr phase diagram such a transformation may not have happened to the same degree in the 12Cr alloy.

Wakai et al. has shown that Fe–9Cr alloys containing 100 wppm C exhibit very different precipitation behavior compared to that of a higher purity alloy with only 3 wppm C [4]. In particular carbon appears to suppress the formation of the alpha-prime phase. Densification might also arise from precipitation of the 160 wppm C as chromium carbides, as observed in austenitic alloys [5,6]. A similar densification at ∼454 °C was observed in an experiment conducted on Fe–12Cr irradiated in EBR-II [7].

The identification of the cause of the densification observed in awaits examination by microscopy. However, the post-densification swelling behavior is the most important aspect of this experiment.

When this series of measurements was initiated it was expected that a clear effect of dpa rate on the duration of the transient regime would be seen, since the model Fe–Cr–Ni alloys in the same FFTF packets exhibited such a behavior. One would reasonably expect the Fe–Cr alloys to exhibit a flux-dependent behavior in the current FFTF experiment as was observed in the earlier FFTF/EBR-II comparative irradiation.

This lack of expected behavior requires us to address whether it is reasonable to expect that an experiment conducted only in FFTF would yield a result similar to that conducted in EBR-II and FFTF. What other differences in variables beside dpa rate might be operating to produce such a different result?

The earlier experiment utilized specimens produced in the USA while the recent experiment utilized specimens prepared in Japan. Whereas the current Japanese experiment involved a heat treatment of 60 min at 900 °C in high vacuum, the earlier experiment utilized 1040 °C/1 h/AC + 760 °C/2 h/AC treatment for both the

EBR-II and FFTF irradiations. The latter intermediate anneal would have assured that the specimens were ferritic rather than austenitic.

Differences in impurity and gas content in the US and Japan preparation have not yet been defined. This possibility of preparation differences contributing to the different swelling response is now being explored in more depth.

The details of temperature history might have had a strong effect. The rise to power and the temperature history are somewhat different between EBR-II and FFTF. This possibility has been explored previously and shown to sometimes exert a strong impact on microstructural evolution in austenitic alloys [8]. However, consideration so far indicates that the difference cannot be ascribed to small details of temperature history.

However, the authors suspect that another, more subtle difference may be operating in the two sets of irradiation experiments. In earlier work on these alloys it was proposed that the flux and temperature sensitivity of swelling in Fe–Cr binaries reflected a greater difficulty of void nucleation in the bcc Fe–Cr system compared to that of the fcc Fe–Cr–Ni system [1,7]. It was further speculated that such a difficulty in void nucleation might reflect a potentially greater role for helium to stabilize voids, especially under fusion or spallation neutron conditions. In fact, the difference in transient duration observed in the original FFTF and EBR-II irradiations might reflect more the differences in helium generation rate than the sole impact of differences in dpa rate.

In the current experiment, differences in dpa rates are accompanied by differences in helium generation rate, with lower dpa rates accompanied by lower helium generation rates as shown in Fig. 3. Might the potential effects of lower dpa rate to shorten the transient duration be cancelled by the associated lower helium generation rates?

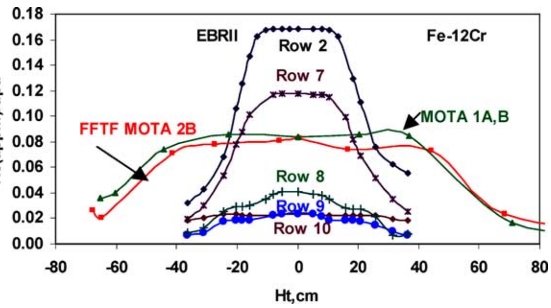

Fig. 3. Calculated He/dpa ratios for Fe–12Cr irradiated in EBR-II and FFTF. The earlier EBR-II experiment was conducted in-core in row 2 at ~0.18 appm He/dpa while the current FFTF experiment was conducted both in and out-of-core, with He/dpa ratios falling from 0.08 to 0.02 appm/dpa.

In every case, the helium generation rates in Fe–Cr alloys in the current experiment are lower than those in EBR-II. All of the EBR-II data were generated at He/dpa ratios on the order of ~0.17 appm/dpa, while the FFTF irradiations proceeded at ~0.02–0.08 appm He/dpa. Note that the lowest dpa rates in FFTF are coincident with progressively lower helium generation rates.

It is proposed that model austenitic alloys irradiated in the same FFTF packets of the current experiment are able to exhibit their potential flux sensitivity much easier, since nucleation is not only easier in the fcc system, but the helium generation rates in FFTF are larger in Fe–15Cr–16Ni due to the much larger contribution of nickel to helium generation.

If void nucleation in bcc Fe–Cr alloys is indeed dependent on helium availability, then one cannot expect EBR-II results from the previous experiment to be comparable to FFTF results, even at the same dpa rate. These results and conclusions require further study and some indication of the reasons for the observed behavior may be obtained after microstructural examination is completed.

5. Conclusions

An experiment conducted in FFTF-MOTA to determine the effect of dpa rate on void swelling of Fe–9Cr and Fe–12Cr model binary alloys appears to indicate that there is no effect of dpa rate on the evolution of swelling, which is contrary to the result observed in model Fe–Cr–Ni alloys irradiated in the same experiment. This result is also contrary to that of an earlier comparative irradiation experiment on nominally similar Fe–Cr alloys irradiated in EBR-II and FFTF.

While differences in alloy preparation or operational history may have contributed to this surprising result, the authors favor an interpretation based on the strong differences in helium generation rate in EBR-II and FFTF, especially in the out-of-core FFTF capsules, and the possible sensitivity of void nucleation to helium availability.

Acknowledgements

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