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Phase Development and Swelling in Fe-Mn and Fe-Cr-Mn Alloys During Neutron Irradiation

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NOTICE

PHASE DEVELOPMENT AND SWELLING IN Fe-Mn AND Fe-Cr-Mn ALLOYS DURING NEUTRON IRRADIATION

F. A. Garner, D. S. Gelles, H. R. Brager, and J. M. McCarthy, (Hanford Engineering Development Laboratory)

1.0 Objective

The object of this effort is to determine those factors which control the swelling of alloy systems which have the potential for reduced long-term activation.

2.0 Summary

Whereas second phases do not form in Fe-Cr-Ni ternary alloys during neutron irradiation in the range 400-600°C, iron-rich ferrite precipitates form in simple Fe-Mn and Fe-Cr-Mn alloys at 520 and 600°C at 14 dpa. The cause of the large densification (2.2%) earlier observed in Fe-35Mn at 9 dpa and 420°C does not appear to be related to precipitation, however, and is thought to be the result of compositional segregation on a rather fine scale.

3.0 Program

Title: Irradiation Effects Analysis (AKJ)
Principal Investigator: D. G. Doran
Affiliation: Hanford Engineering Development Laboratory

4.0 Relevant DAFS Program Plan Task/Subtask

Subtask II.C.1 Effects of Material Parameters on Microstructure

5.0 Accomplishments and Status

5.1 Introduction

In previous reports(1-3) it was shown that the swelling of simple Fe-Mn binary and Fe-Cr-Mn ternary alloys in FFTF-MOTA at 520 and 600°C at 14 dpa was remarkably insensitive to the chromium content and only weakly dependent on the manganese level, as shown in Figures 1 and 2. It was also shown that the swelling of these alloys at 420°C and 9 dpa was likewise rather insensitive to composition but that there was an increasing tendency to densify with increasing manganese content, approaching 2.2% densification at Fe-35Mn, as shown in Figure 2.

This densification was also thought to account for the slight dependence of density change on manganese content observed at 520 and 600°C at 16 dpa. In effect, the observed swelling as measured by density change was the sum of relatively composition-independent void swelling and composition-sensitive phase instabilities.

In the most recent report it was shown that there was some basis for assuming that there was a compositional dependence of precipitation at 520°C.(3) In Fe-10Cr-XMn alloys, there was an increase found in the formation of large precipitates (unidentified at that time) when the manganese level fell from 30 to 20%.

There was also a concomitant increase in swelling, however, showing that the increase in precipitation was somehow related to the onset of swelling and was not in itself the sole cause of the slight dependence of density change on composition.

In this report a number of the binary and ternary alloys have been examined using transmission election microscopy and x-ray analysis. In some cases the examination was only cursory and all specimens continue to be examined.

5.2 Examination of Fe-5Cr-15Mn Irradiated at 600°C to 14 dpa

As shown in Figure 3, there were two exceptions to the general trend established by the swelling data of all other alloys at 520 and 600°C. These exceptions both involved Fe-5Cr-15Mn. At both 520°C and 600°C the swelling was significantly below that expected from the trends observed at higher chromium levels. The 520°C specimen has not yet been examined but the 600°C specimen has been examined. As shown in Figures 4a-4b the matrix has decomposed into large elongated ferrite precipitates, on the order of 2-10 microns in size) and a matrix of retained austenite. The relative volume of the two phases is difficult to determine because the austenite is easily attacked and destroyed in the polishing, leaving the ferrite particles protruding far from the surfaces and edge of the foil.

An even more interesting observation is that voids were found but they appeared to be coated with a shell that also resisted electropolishing. Note in Figure 4b that some coated voids are also suspended from precipitates hanging over the edge of the foil. At this point it is assumed that the void shells are ferritic in nature, but this assumption has not yet been tested.

5.3 Examination of Specimens Irradiated at 520°C to 14 dpa

To date we have examined Fe-35Mn, Fe-10Cr-30Mn, Fe-15Cr-20Mn and Fe-10Cr-20Mn.

5.3.1 Examination of Fe-35Mn

As shown in Figure 5, once again we observe a decomposition into ferrite precipitates which resist electropolishing and the more easily removed retained austenite. The precipitates range from 0.2 to 1.5 μm in size. In this case the precipitates were analyzed by EDX and found to be essentially pure iron. Once again the voids appear to be coated and resistant to electropolishing. Within any one grain the swelling and phase decomposition appeared to be rather uniform, but adjacent grains often exhibited quite different levels of evolution. In some grains in which the precipitation was low, void swelling was present but at obviously lower levels.

5.3.2 Examination of Fe-10Cr-30Mn

This specimen as originally polished possessed a poorly polished surface and insufficient thin area. It was later flash-polished, yielding a thinner region for examination but also causing the ferrite precipitates to preferentially etch instead of the austenite matrix, as shown in Figure 6. Once again it was obvious that substantial ferrite formation had occurred. Since the ferrite particles did not protrude from the foil edge and could not be easily analyzed for their composition, a broad beam scan of a large area was performed. A drop in the overall iron level from 70 to 65% was observed, confirming that the lost ferrite precipitates were again rich in iron.

5.3.3 Examination of Fe-10Cr-20Mn

As shown in Figure 7 there is substantially less ferrite in this specimen, indicating that decreasing the manganese from 30 to 20% at 10% chromium affects the rate of formation of ferrite. The local swelling levels are in general larger than that of the Fe-10Cr-30Mn alloy. The dislocation evolution is easier to study in the absence of precipitation and appears to be typical of that observed in Fe-Cr-Mn alloys. The relative orientation of the ferrite precipitates were analyzed with respect to the matrix. They were found to exhibit the well-known Kurdjimov-Sachs relationship where $(111)_\gamma \parallel (011)_\alpha$ and $[\bar{1}01]_\gamma \parallel [\bar{1}\bar{1}1]_\alpha$.

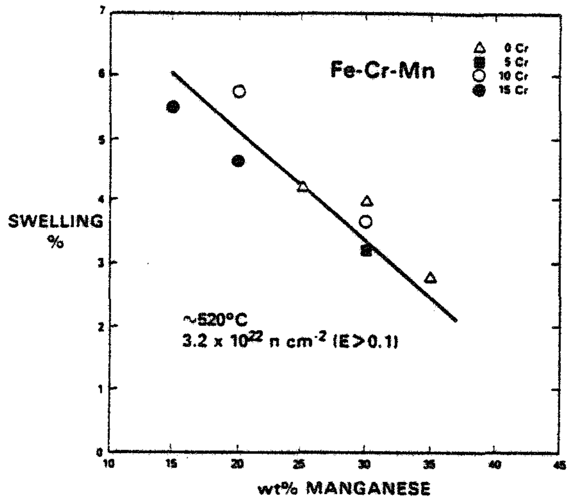

Figure 1. Neutron-induced swelling of Fe-Mn and Fe-Cr-Mn alloys in FFTF at 520°C and 14 dpa as determined by immersion density measurements.(1-3)

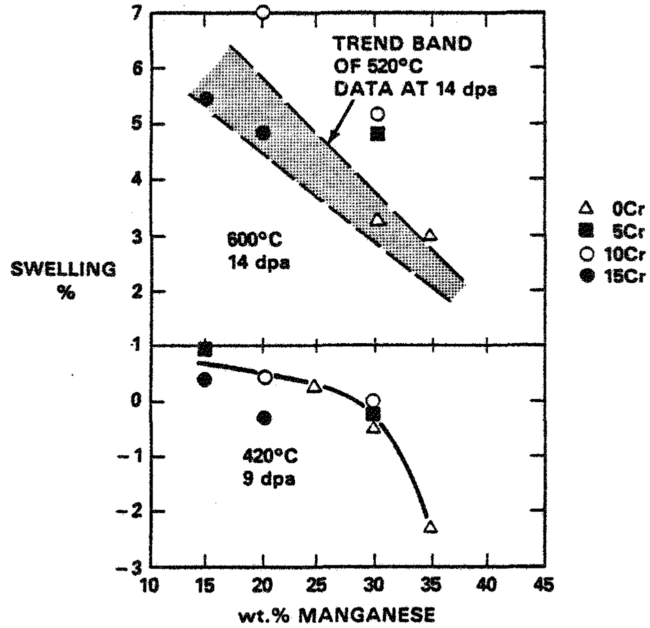

Figure 2. Neutron-induced density changes of Fe-Mn and Fe-Cr-Mn alloys at (14 dpa, 600°C) and (9 dpa, 420°C).(3)

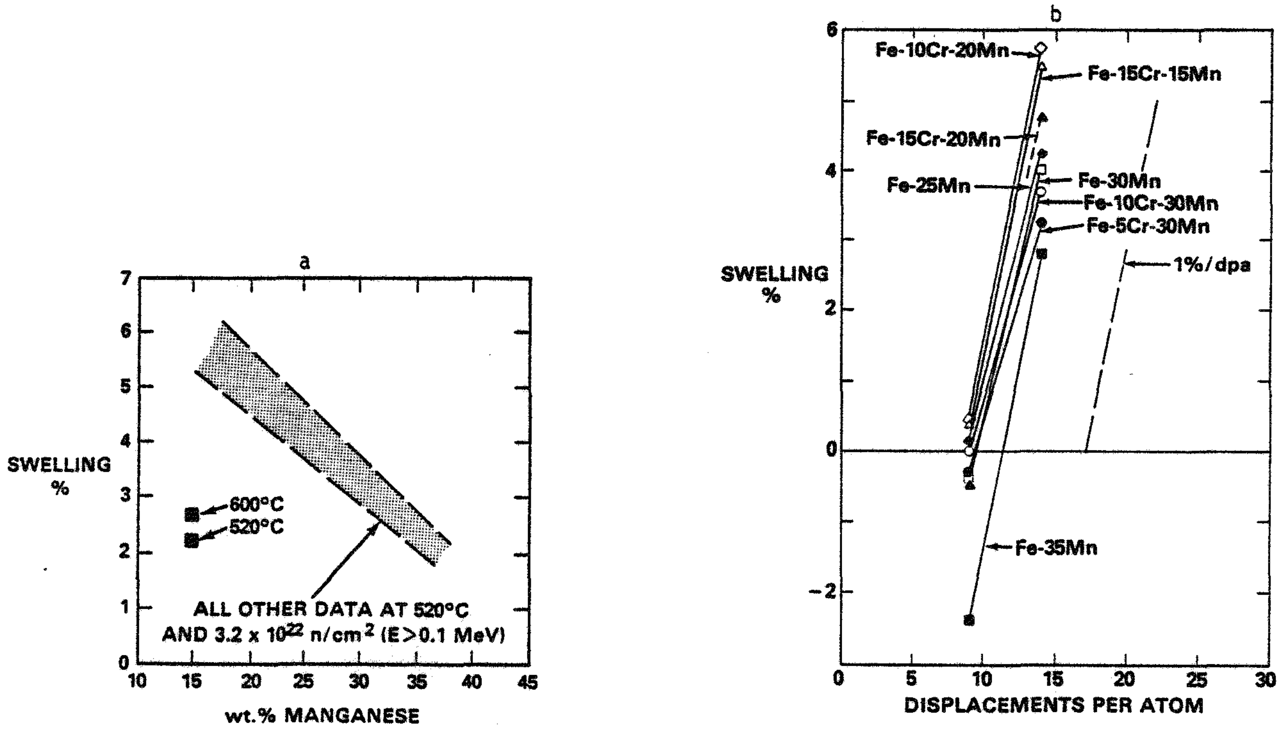

Figure 3. (a) Comparison of swelling of Fe-5Cr-15Mn at 14 dpa with the trend of other Fe-Cr-Mn alloys at 520°C and 14 dpa. (b) Plot of 420 and 520°C data ignoring temperature, with the exception of Fe-5Cr-15Mn at 520°C.

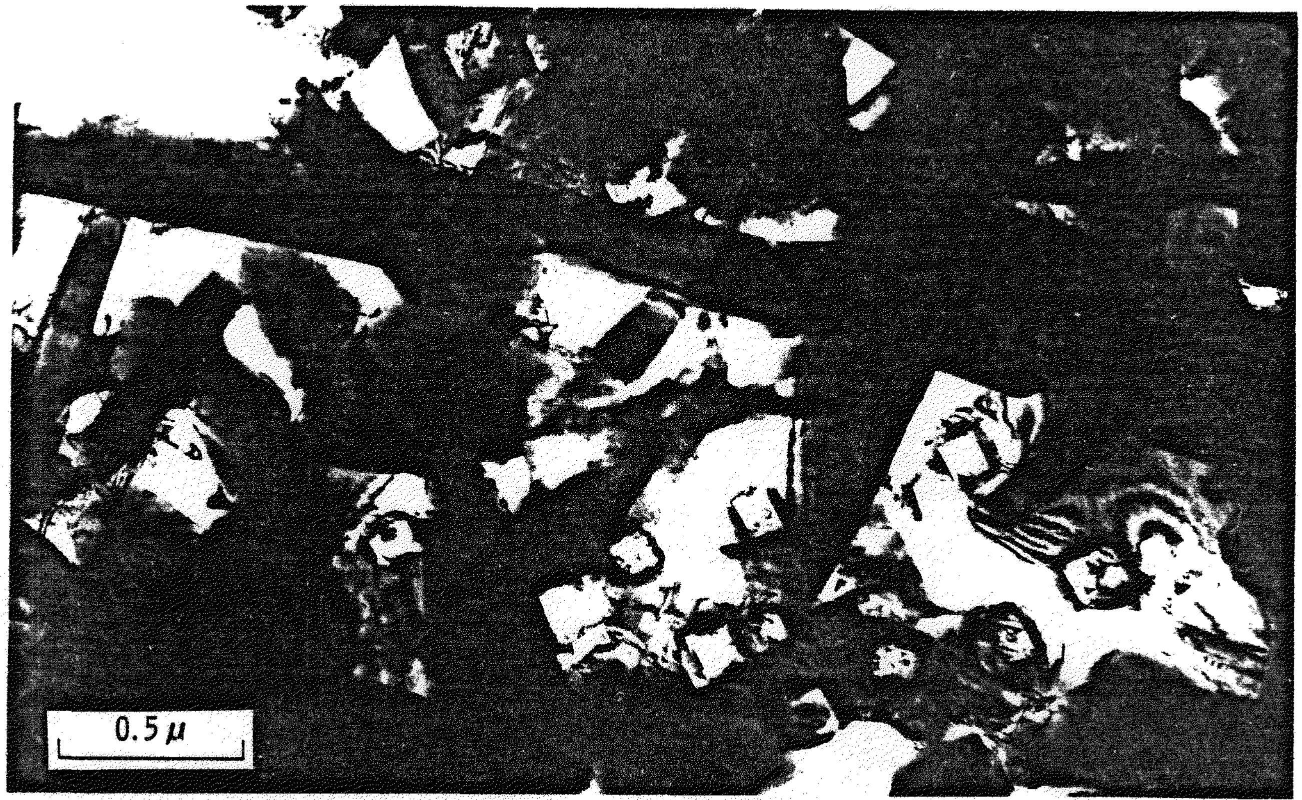

Figure 4. Micrographs showing formation of large iron-rich ferrite precipitates protuding from the strongly etched austenite matrix in Fe-5Cr-15Mn at 520°C and 14 dpa. Voids are also coated with ferrite and resist electropolishing. The arrow in (b) points to an almost unsupported void which has resisted electropolishing.

Figure 5. Decomposition of Fe-35Mn at 520°C and 14 dpa.

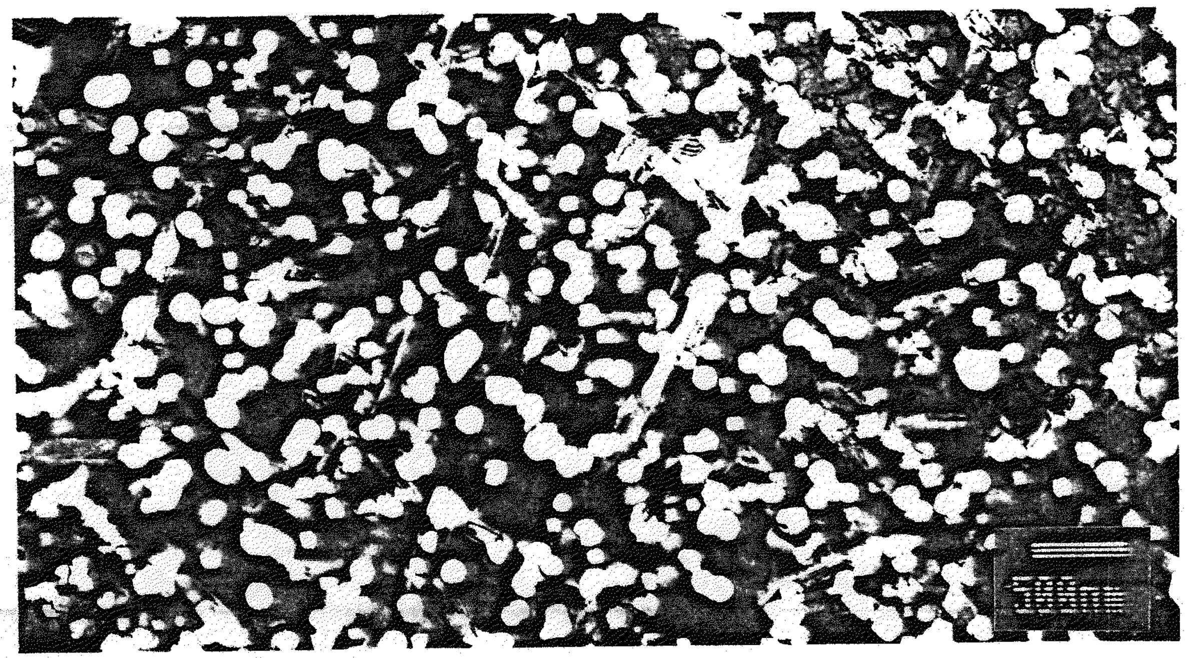

Figure 6. Preferential attack of ferrite particles during flash-polishing of Fe-10Cr-30Mn (520°C, 14 dpa).

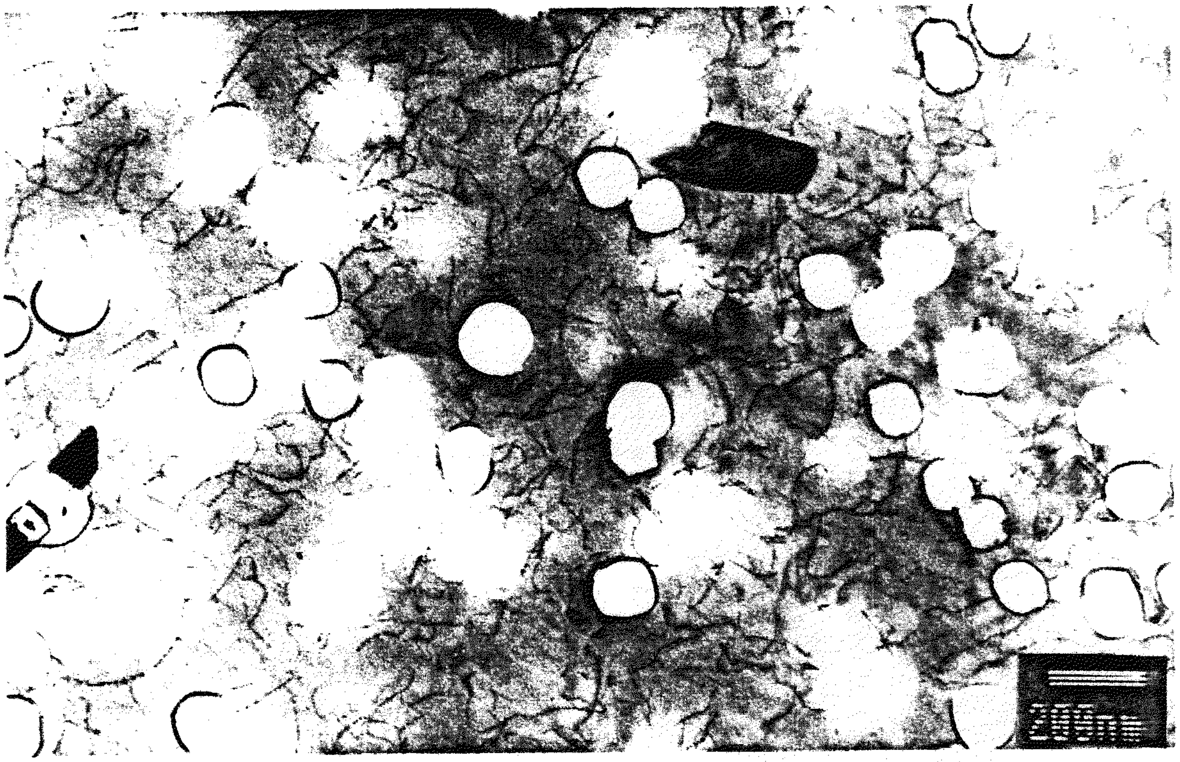

Figure 7. Micrograph of Fe-10Cr-20Mn after irradiation at 520°C to 14 dpa.

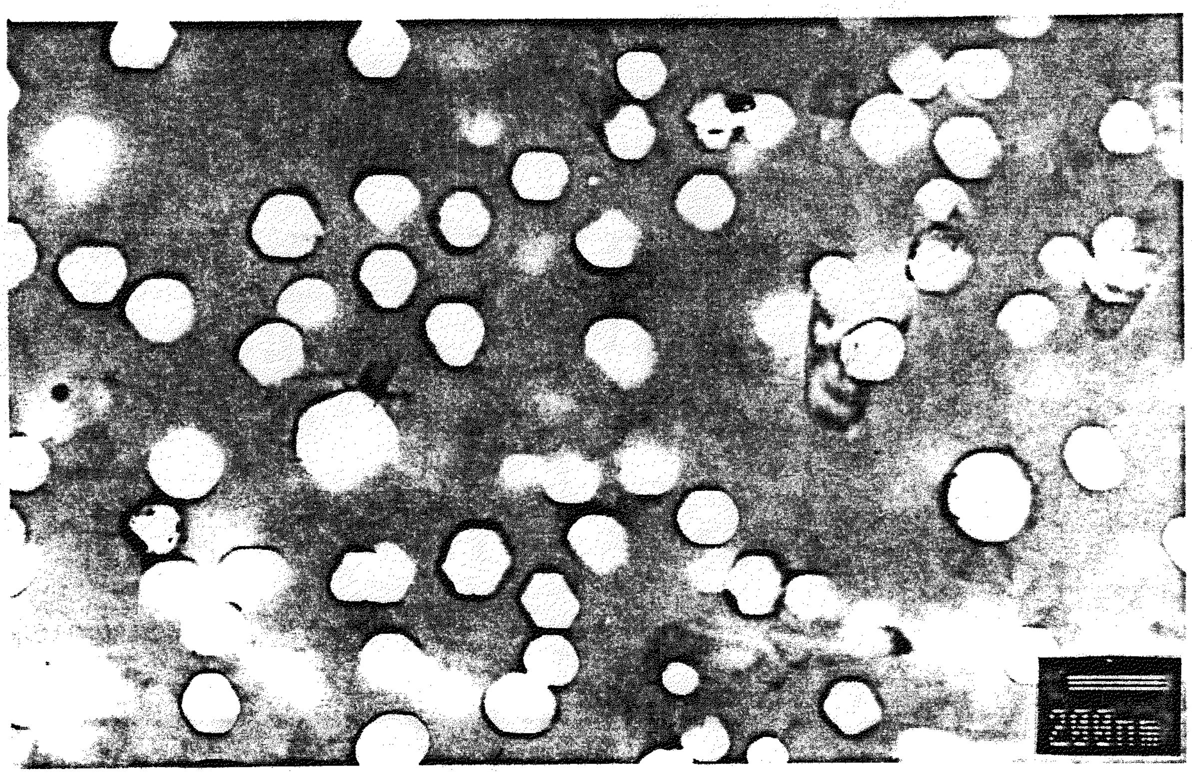

Figure 8. Micrograph of Fe-15Cr-20Mn after irradiation at 520°C to 14 dpa.

5.3.4 Examination of Fe-15Cr-20Mn

As shown in Figure 8 the ferrite density seems to have decreased somewhat relative to that of Fe-10Cr-20Mn but the swelling is comparable. The precipitates were found to average 75-80% iron.

5.4 Examination of Specimens Irradiated at 420°C and 9 dpa

Two specimens have been examined thus far, Fe-35Mn and Fe-30Mn. Both exhibited a high density of contrast features typical of low temperature irradiation. These were found to be small dislocation loops, largely unfaulted, (<500 Å) and small voids (<100 Å). No precipitates were observed. Small streaks observed in the diffraction pattern were found to be associated with an oxide-hydroxide of iron and manganese on the surface of the foil. This oxidation reflects the lack of chromium in these alloys.

5.5 Discussion of Results

In comparing the evolution of Fe-35Mn at (420°C, 9 dpa) with that at (520°C, 14 dpa) it is obvious that the phase stability at this composition is quite sensitive to temperature. We could possibly ascribe the difference to the higher dose as well, but there is no hint of precipitation at 420°C and 9 dpa. Otherwise the precipitation response must occur rather abruptly. This lack of precipitation also leaves us with no explanation of the large densification (2.2%) previously observed at 420°C. The amount of densification at 420°C is comparable to the relative decrease in swelling of this alloy at 520°C, which leads us in turn to suspect that substantial microsegregation has occurred at 420°C that has not yet caused precipitation to occur.

The formation of ferrite appears to be sensitive to manganese content at 10% chromium but appears to be most sensitive to chromium content at all levels of manganese investigated thus far. It remains to be determined that the shells around the voids are actually iron-rich ferrite but their resistance to electropolishing leads us to suspect that they are ferritic in nature. It is also known that iron is the slowest diffusing species compared to manganese and chromium in both the iron-based fcc and bcc crystal structures. Based on the observed behavior in the Fe-Cr-Ni system the slowest diffusing species is expected to segregate at the void surface via the Inverse-Kirkendall mechanism.

If we look at the equilibrium phase diagram for Fe-Mn the boundary between the two phase ($\alpha + \gamma$) region lies at ∿14% Mn at 600°C, ∿18% Mn at 520°C and ∿23% Mn at 420°C. The α ferrite phase is predicted to contain no more than 3% manganese, which is consistent with the finding of essentially pure iron precipitates in Fe-35Mn at 520°C. Chromium additions change the phase boundaries only a little. Therefore all but the 15Mn alloys should be stable at the conditions of irradiation, unless the inverse-Kirkendall mechanism or some other process is operating. Even though the phase boundary is closest for most alloys at 420°C, the Inverse Kirkandall mechanism is known to operate most strongly at higher temperatures. This may account for the lack of ferrite observed in the 30-35Mn alloys at 420°C.

Void swelling and ferrite formation appear to be connected but the exact relationship has not yet been determined. Part of the density change may arise from a lattice parameter change associated with ferrite formation, part with the voidage, and another part associated with micro-segregation of iron and manganese in the fcc phase. In an earlier report it was shown that the dependence of density on manganese content is very strong in the fcc phase. The inconsistent swelling behavior of Fe-5Cr-15Mn at 500 and 600°C may reflect a situation where the precipitate formed at such quantities prior to the onset of swelling that the volume of austenite is reduced, leading to less swelling overall.

5.6 Conclusions

The density changes observed in simple Fe-Mn and Fe-Mn-Cr alloys during neutron irradiation involve not only swelling, but also ferrite formation and segregation-induced changes in lattice parameter. The Fe-Cr-Mn system is prone to phase instabilities not observed in comparable Fe-Cr-Ni alloys during neutron irradiation.

6.0 References

1. H. R. Brager and F. A. Garner, "Swelling of Fe-Cr-Mn Ternary Alloys in FFTF," DAFS Quarterly Progress Report, DOE/ER-0046/19, November 1984, pp. 31-33.

2. F. A. Garner and H. R. Brager, "Neutron-Induced Swelling of Fe-Cr-Mn Ternary Alloys," DAFS Quarterly Progress Report, DOE/ER-0046/21, May 1985, pp. 41-45.

3. F. A. Garner and H. R. Brager, "Neutron-Induced Swelling and Microstructural Development of Simple Fe-Mn and Fe-Cr-Mn Alloys in FFTF," DAFS Quarterly Progress Report, DOE/ER-0046/23, November 1985, pp. 18-23.

7.0 Future Work

Microcopy at the 9 and 16 dpa levels will continue. Density data at (35 dpa, 600°C), (45 dpa, 420°C) and (45 dpa, 500°C) will be obtained.

8.0 Publications

None