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The conflicting roles of boron on the radiation response of precipitate-forming austenitic alloys at ~400 °C

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A B S T R A C T

The behavior of void swelling at ~400 °C of model f.c.c. alloy Fe–15Cr–16Ti–0.25Ti–0.05 C doped with boron was examined in the FFTF–MOTA. Boron additions modify the neutron-induced swelling of Fe–15Cr–16Ni–0.25Ti–0.05 C somewhat, but the changes appear to arise primarily from the influence of boron as a chemical species rather than as a source of helium. Boron additions initially depress swelling strongly, but the effect saturates by <100 appm. The reduction in swelling is thought to arise from boron's influence on distribution and precipitation of carbon. As the boron level is raised to significantly larger levels swelling begins to increase, but at a slower rate per boron atom. This subsequent increase is thought to reflect the higher He/dpa ratio generated by the boron, overwhelming the helium produced by (n, α) reactions with nickel.

1. Introduction

In an earlier report it was shown that two simple, annealed austenitic alloys, ternary Fe–15Cr–16Ni and quaternary Fe–15Cr–16Ni–0.25Ti, when irradiated in FFTF–MOTA at ~400 °C over a wide range of dpa rates, exhibited a very strong influence of dpa rate on void swelling [1]. While the steady state swelling rate of ~1%/dpa was unaffected by dpa rate, the transient regime was strongly affected, with a progressive shortening of the transient duration as the dpa rate decreased. A later paper showed that carbon additions to the quaternary alloy resulted not only in a reduction of swelling but also in a total loss of the influence on dpa rate [2].

It was also shown in the earlier study that the addition of 500 appm of natural boron to the ternary and quaternary alloys had very little effect on swelling [1]. However, this latter conclusion was not as confidently demonstrated at all irradiation conditions.

The ambiguity arose from the manner in which the specimens were stacked in the experimental packets which contained as many as 100 specimens each. All specimens were standard 0.3 mm thick microscopy disks stacked in stainless steel packets. In some packets, the boron-containing alloys were separated by ~1.5 cm distance from the boron-free alloys. In packets which were irradiated in-core with minimal gradient in dpa rate the conclusion was very clear in that there was no obvious influence of boron on the swelling. In packets lying across the core boundary or away from the core boundary, however, it appeared that swelling could be either increased or decreased by boron. However, not only was the effect of dpa rate gradients across the packet dominating the results but there was no record of how a given packet was oriented in the gradient, potentially giving rise to either increases or decreases in apparent swelling.

Also contained in the FFTF–MOTA experiment was Fe–15Cr–16Ti–0.25Ti–0.05 C, also in the annealed condition. Boron-doped variants of Fe–15Cr–16Ni–0.25Ti–0.05 C were also included in other packets. As shown in Fig. 1, however, there was no significant separation of the boron-free and boron-containing specimens, allowing a more confident determination of the role of boron on swelling, even in packets that exist in dpa gradients.

2. Experimental details

Relatively pure Fe–15Cr–16Ni–0.25Ti–0.05 C (at.%) was prepared by arc melting high purity Fe, Ni, Cr, Ti and C. The alloy was rolled to sheets of 0.25 mm thickness, cut into 3 mm disks and annealed for 30 min at 1050 °C in high vacuum. Also included in some sets of specimens were Fe–15Cr–16Ni–500 appm natural boron, and/or Fe–15Cr–16Ni–0.25Ti-0.05 C with natural boron levels of 100, 500 and 2500 appm, all prepared using the same procedures. The specimens were stacked in the same order in each nominally identical packet, but there was no control placed or record kept of which end of the packet was oriented toward the top or bottom.

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

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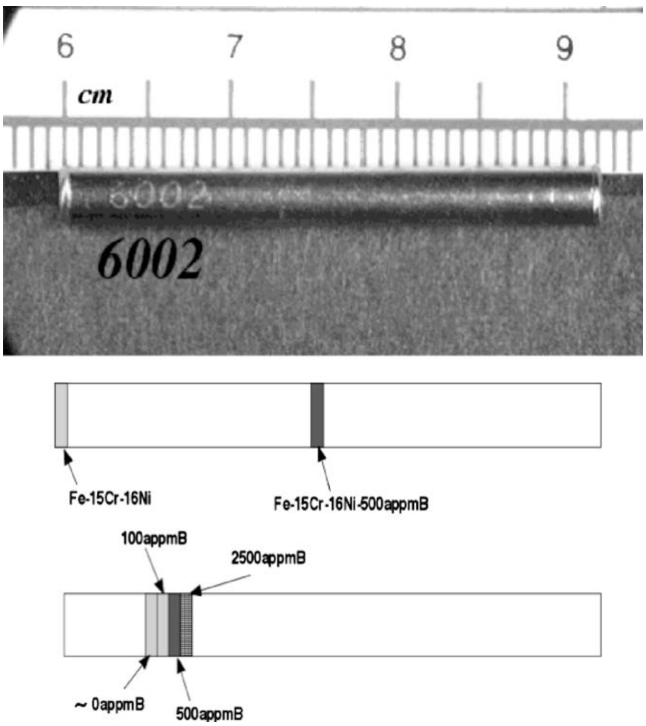

Fig. 1. Sealed stainless steel packets containing ~100 microscopy disks. Note in the original study [1] that the boron-free and boron-containing specimens were separated by ~1.5 cm such that strong gradients in dpa rate out of core could lead to significant differences in dpa. However, in the specimens examined in this paper, all boron contents including ~0 appm were in intimate contact.

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. The various 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 were actively controlled to ±5 °C of the nominal target, although the nominal target temperatures varied a little from capsule to capsule. In the below-core capsule the temperatures were largely controlled by the inlet coolant temperature and the gamma heating level, and therefore can be calculated with rather small error.

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 8.9×10^{-9} to 1.7×10^{-6} dpa/s. The dose levels attained by the specimens varied from 0.23 to 43.8 dpa in Cycle 11 and an additional 0.38–24.0 dpa in Cycle 12.

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. Microscopy was not performed on these specimens.

3. Results

As shown in Fig. 2 by Sekimura and coworkers [2], the swelling of the carbon-doped alloy at ~400 °C appears to show no obvious influence of the dpa rate. Note that in the two data sets that the different dpa levels were produced at different dpa rates at constant time.

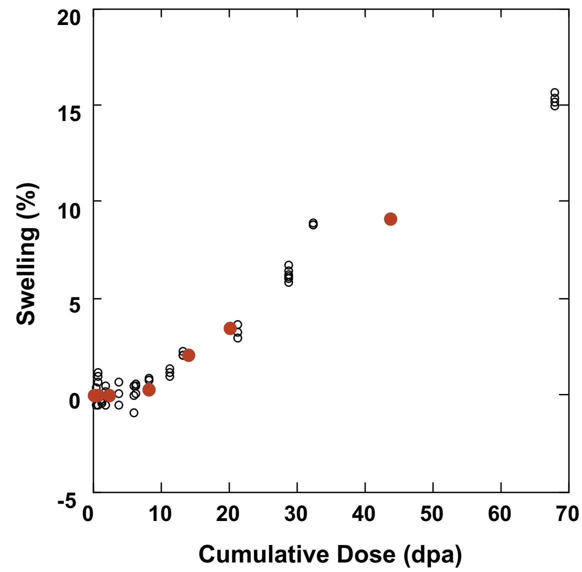

Fig. 2. Swelling of simple model f.c.c. Fe–15Cr–16Ni–0.25Ti-0.05 C in FFTF–MOTA at ~ 400 °C, showing that the swelling is relatively independent of dpa rate. The solid data were obtained from irradiation in MOTA-2A only and the open data from irradiation in both MOTA-2A and 2B.

As was observed in the two simple undoped alloys, the range of swelling between identical specimens is relatively small, indicating the reproducibility of the swelling phenomenon. Surprisingly, the swelling of the twelve data ensemble of the carbon-doped alloy appears to be following a general, lower-swelling trend somewhat characteristic of the undoped alloys at the highest dpa rate.

Fig. 3 shows that addition of 500 appm boron to the carbon-doped alloy reduced the swelling level somewhat, but the alloy in general retained its insensitivity to dpa rate. Since this comparison was made only at 500 appm B, it is not possible from this comparison to determine the effect of smaller amounts or observe the effects of progressively larger additions of boron. Three packets

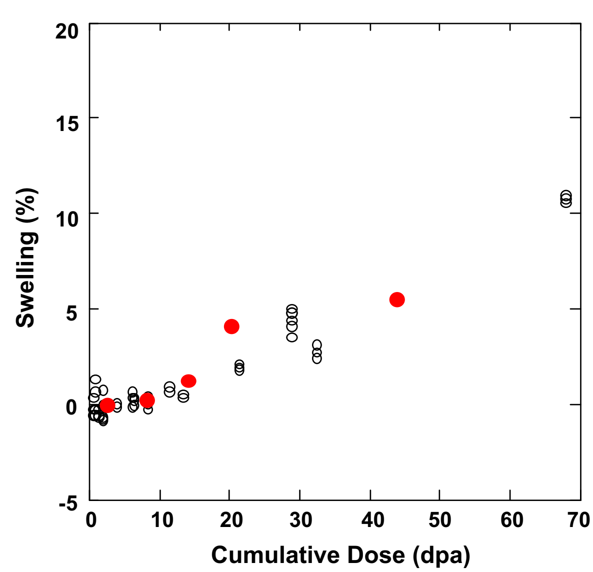

Fig. 3. Swelling of simple model f.c.c. Fe–15Cr–16Ni–0.25Ti-0.05 C–500 appm B in FFTF–MOTA at ~400°C, showing that the swelling is reduced somewhat by boron addition but remains relatively independent of dpa rate. The solid data were obtained from irradiation in MOTA-2A only and the open data from irradiation in both MOTA-2A and 2B.

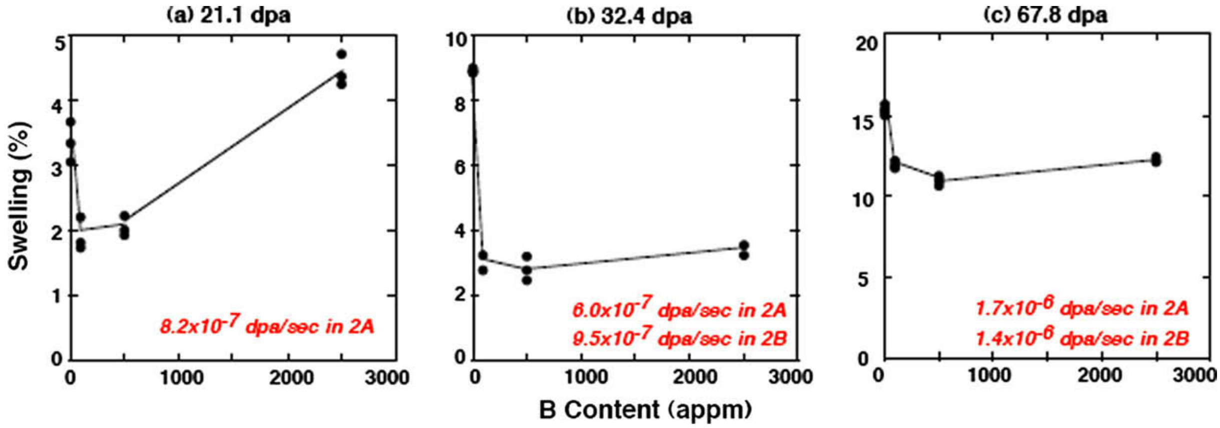

Fig. 4. Swelling of simple model f.c.c. Fe–15Cr–16Ni–0.25Ti-0.05 C with various levels of boron, as observed after irradiation in FFTF–MOTA at ~400 °C at three different dose rate ranges to reach three dose levels. Note that at 32.4 and 67.8 dpa irradiation proceeded in two successive MOTAs with slightly different dpa rates in each increment.

containing a wider variety of boron levels were therefore examined to address this issue.

Fig. 4 shows that most of the decrease in swelling observed at 500 appm boron was already attained at the 100 appm level. Beyond 500 appm there was a tendency to increase swelling, with the amount of the increase dependent on the dpa rate.

4. Discussion

It is clear that addition of boron at 500 appm decreases the swelling somewhat of Fe–15Cr–16Ni–0.25Ti–0.05 C but does not appear to change the insensitivity to dpa rate that occurred when carbon was added. Note also in both Figs. 2 and 3 that the steady-state swelling rate of ~1%/dpa observed in the carbon-free alloys [1,2] was not attained in the carbon-added alloys. This behavior is not understood and may be related to the loss of the flux dependence of swelling.

Previous studies on the chemical effect of boron has shown that boron interacts with the precipitation of carbides and therefore may exert its influence on void swelling both directly or indirectly through its influence on carbon [3]. With respect to boron's contribution it must be recognized that boron can play several roles, however, first as a chemical species and second as a source of transmutants helium and lithium produced by the $^{10}B(n, \alpha)\ ^{7}Li$ reaction with thermal and epithermal neutrons.

Note, however, that ^{10}B comprises only 20% of natural boron. As shown in reference [4] some helium in these specimens is produced by various (n, α) reactions, primarily with nickel in these alloys, such that the boron-generated helium is additive to that generated in the base alloy. Additionally, the burn-up of boron in a fast reactor is very sensitive to minor differences in the epithermal portion of the neutron spectra. This effect causes the He/dpa ratio due to boron to increase strongly below the core and decrease above the core [4].

In the core center positions from which the data shown in Fig. 4 were attained, the impact of 100 appm boron is only a 30–40% increase in the helium generation rate which is already rather large. For example, at 67.8 dpa the boron contribution increased the helium from 15.5 appm generated by the base alloy to only 20 appm arising from the 100 appm boron addition.

Since the primary impact of boron was attained by 100 appm boron and the boron-induced increase in helium was not large, it is concluded that the primary effect of boron is probably chemical in nature and most likely arises from its influence on the distribution of carbon and its possible precipitation in the alloy. However, in Fe–15Cr–16Ni–0.25Ti–0.05 C irradiated in this experimental series most of the carbon appeared to be retained with very little precipitation at ~400 °C [2]. This suggests that perhaps TiC complexes were responsible for producing the observed effects on swelling and that Ti–C–B complexes are perhaps even more effective. Since microscopy was not performed on the current specimens, however, it can not be confidently stated that precipitation did or did not occur.

5. Conclusion

Boron additions depress the neutron-induced swelling of Fe–15Cr–16Ni–0.25Ti-0.05 C somewhat at ~400 °C, but the changes appear to arise primarily from the influence of boron as a chemical species rather than as a source of helium. Boron additions initially depress swelling strongly but the effect saturates by <100 appm. The depression is thought to arise from boron's influence on distribution and precipitation of carbon.

As the boron level is raised to significantly larger levels, then swelling increases but at a slower rate per boron atom. This increase is thought to reflect the higher He/dpa ratio generated by the boron overwhelming the helium produced by (n, α) reactions with nickel.

Acknowledgements

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References

[1] T. Okita, W.G. Wolfer, F.A. Garner, N. Sekimura, J. Nucl. Mater. 329–333 (2004) 1013.
[2] N. Sekimura, T. Okita, F.A. Garner, J. Nucl. Mater. 367–370 (2007) 897.
[3] F.A. Garner, Irradiation Performance of Cladding and Structural Steels in Liquid Metal Reactors of Materials Science and Technology A Comprehensive Treatment, vol. 10A, Springer, Berlin, 1988. Chapter 6.
[4] L.R. Greenwood, F.A. Garner, J. Nucl. Mater. 329–333 (2004) 1147.