NEUTRON-INDUCED SWELLING AND EMBRITTLEMENT OF PURE IRON AND PURE NICKEL IRRADIATED IN THE BN-350 AND BOR-60 FAST REACTORS - N. I. Budylkin, E**.** G. Mironova and V. M. Chernov Bochvar Institute of Nonorganic Chemistry, Moscow, Russia, V. A. Krasnoselov Research Institute of Atomic Reactors, Dimitrovgrad, Russia, S. I. Porollo Institute of Physics and Power Engineering, Obninsk, Russia, and F. A. Garner (Pacific Northwest National Laboratory)*

OBJECTIVE

The purpose of this effort is explore the differences in swelling and tensile properties that arise between two simple fcc and bcc metals when subjected to high fluence irradiation over a wide range of irradiation temperatures.

SUMMARY

Pure iron and nickel were irradiated to very high exposures in two fast reactors, BOR-60 and BN-350. It appears that both nickel and iron exhibit a transient-dominated swelling behavior in the range of 2 to 15×10^{-7} dpa/sec, with the shortest transient at ~500°C in nickel, but at <350°C for iron. It also appears that the duration of the transient regime may be dependent on the dpa rate.

When the two metals are irradiated at 345-355°C, it is possible to obtain essentially the same swelling level, but the evolution of mechanical properties is quite different. The differences reflect the fact that iron is subject to a low-temperature embrittlement arising from a shift in ductile-brittle transition temperature, while nickel is not. Nickel, however, exhibits high temperature embrittlement, thought to arise from the collection of helium gas at the grain boundaries. Iron generates much less helium during equivalent irradiation.

INTRODUCTION

Development of a comprehensive theory of the irradiation-induced nucleation and growth of voids in multi-component austenitic or ferritic-martensitic alloys is a challenging task, even after more than three decades following the first observation of void swelling. The construction of theoretical models is facilitated by experimental data developed from very simple systems without the complexity associated with segregation, precipitation or phase changes. Therefore data on the basic components of iron-base and nickel-base alloys such as pure metals (Fe, Ni) and simple binary Fe-Cr or ternary Fe-Cr-Ni model alloys are particularly useful. Pure metals in general exhibit void swelling at much lower doses than do even the simplest of model alloys, and therefore were addressed in this study.

While pure nickel has been irradiated a number of times to rather high neutron exposures [1-7], the data on pure iron is in general more limited in dpa level attained [8-14] and most of the data for both metals often cover rather narrow ranges of irradiation temperature. With respect to radiation-induced changes in mechanical properties, however, there are almost no data available for these metals.

This study focuses on the results of high dose (22-70 dpa) irradiations of pure iron and nickel in two fast reactors over a wide range of temperature, 345 to 650°C. Both swelling and mechanical property data were collected.

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Experimental Details

Two types of specimens were employed. The first were right circular cylinders of 6 mm in diameter and 28 mm length. The second were miniature tensiles also 28 mm long with gauge length of 15 mm and 3 mm diameter, with the grip ends also 6 mm in diameter. While both sets were measured to determine their density using hydrostatic weighing in CCl_4, the second set was subjected to tensile testing. The accuracy of density measurements before and after irradiation was ±0.2% and ±0.5%, respectively, reflecting the greater difficulty in remote measurement of radioactive specimens.

The nickel (99.95%) and iron (99.95%) specimens were annealed in sealed ampoules filled with pure argon for 60 minutes at 850°C and then water quenched. Some of the cylinder specimens were irradiated in the core of the BN-350 fast reactor and were in contact with flowing sodium at 500°C, reaching 50 dpa at 1.5×10^{-6} dpa/sec. The standard NRT model was used to calculate the displacement levels.

Another group of specimens were irradiated in contact with sodium coolant at temperatures ranging from 345-650°C and doses between 22-70 dpa. The irradiations were conducted in separate canisters in either the outer row of the BOR-60 reactor core or the first row of the reflector at dpa rates between $2.2\text{-}3.7 \times 10^{-7}$ dpa/sec. While most canisters were filled with cylindrical specimens, the iron tensile specimens were irradiated at 345°C to 58 dpa, and the nickel tensile specimens were irradiated at 355°C to 70 dpa. In most conditions iron and nickel were irradiated at identical dpa rates at a given temperature.

In most cases there was only one specimen per irradiation condition, but in three cases there were two specimens irradiated side-by-side, allowing a check on the reproducibility of the swelling process.

Tensile tests were conducted under argon gas at 1 mm/min at test temperatures ranging from 20-800°C.

Experimental Results

Figure 1 presents the swelling observed in pure nickel, showing an apparent peak swelling temperature at ~500°C. Figure 2a presents the majority of the swelling data plotted vs. dpa level, and shows that the temperature dependence of swelling is expressed primarily in the transient regime with a minimum transient at ~500°C. The temperature dependence of void swelling in nickel appears to arise primarily from the dependence of the transient regime of swelling on temperature, with the steady state swelling rate tending to increase to a value that is not strongly dependent on temperature.

Note that in both figures that irradiation at each temperature proceeds at a different dpa rate. Thus, there is the possibility that the apparent temperature dependence may actually result from the combined influence of two variables, especially at higher irradiation temperatures, as shown in Figure 2b.

Figure 3 presents a comparison of the higher fluence nickel data with the swelling data for pure iron. Note that with the possible exceptions of swelling at 345-355 and 650 °C, iron swells less than nickel. Even more importantly, however, is the observation that the peak swelling of iron occurs at some temperature below 350°C. Once again the temperature-flux dependence of swelling is exhibited in the duration of the transient regime of swelling, as shown in Figure 4.

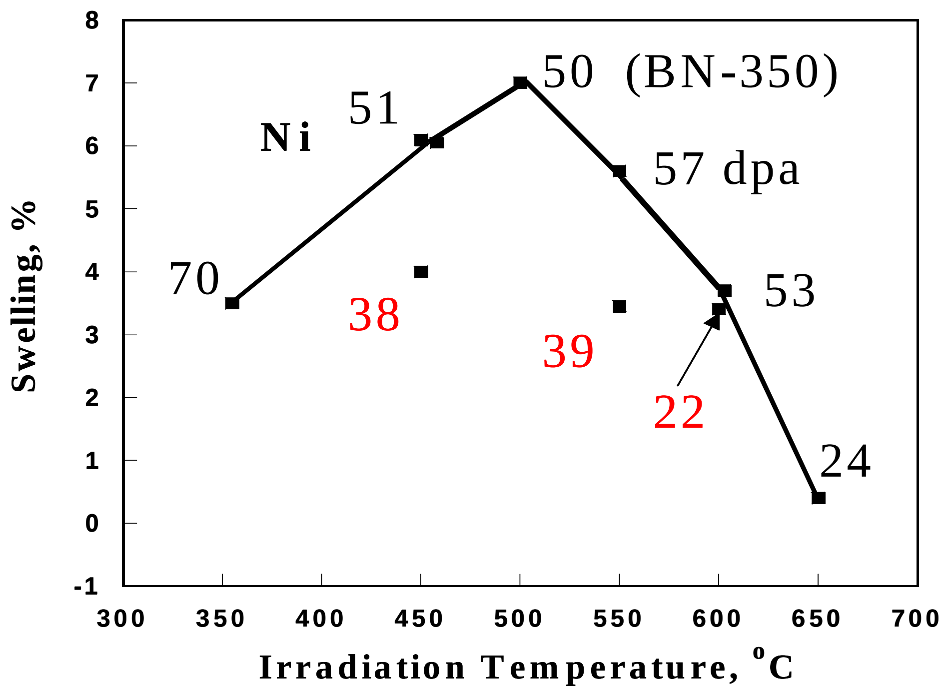

Figure 1. Swelling observed in pure nickel irradiated in BN-350 (500°C only) and BOR-60, plotted vs. irradiation temperature. Dpa levels are indicated for each data point. The two data at 51 dpa and 450°C indicate that the swelling is rather reproducible.

Figures 5a and 5b present the strength and ductility characteristics of pure nickel, both before and after irradiation to 70 dpa at 355°C. Before irradiation, annealed nickel is rather soft and ductile, with both strength and elongation decreasing with test temperature. Irradiation raises the strength and lowers the elongation in agreement with the behavior usually observed in irradiated metals.

Also as typically observed, the yield strength approaches the ultimate strength. Most importantly, the elongation falls to zero in the irradiated nickel when tested above ~500°C.

Figure 6a shows that pure iron at 58 dpa and 345°C exhibits trends in strength with temperature and irradiation similar to those of nickel, but the gap between unirradiated and irradiated properties is maintained until higher test temperatures.

Figure 6b shows quite a different behavior in the elongation of iron when compared to that of nickel. Although the elongation initially decreases with test temperature, above 400°C the elongation of unirradiated specimens increases strongly with test temperature. A similar behavior is seen in the irradiated specimens but only after the test temperature exceeds ~650°C. It is significant, however, that unlike the behavior of nickel the total elongation of irradiated iron remains at acceptable levels of 10-20% throughout the 20-800°C range.

Discussion

The temperature dependence of void swelling in both iron and nickel appears to arise primarily from the temperature and flux dependence of the transient regime of swelling, with the steady state swelling rate tending to increase to a value that is not as strongly dependent on temperature.

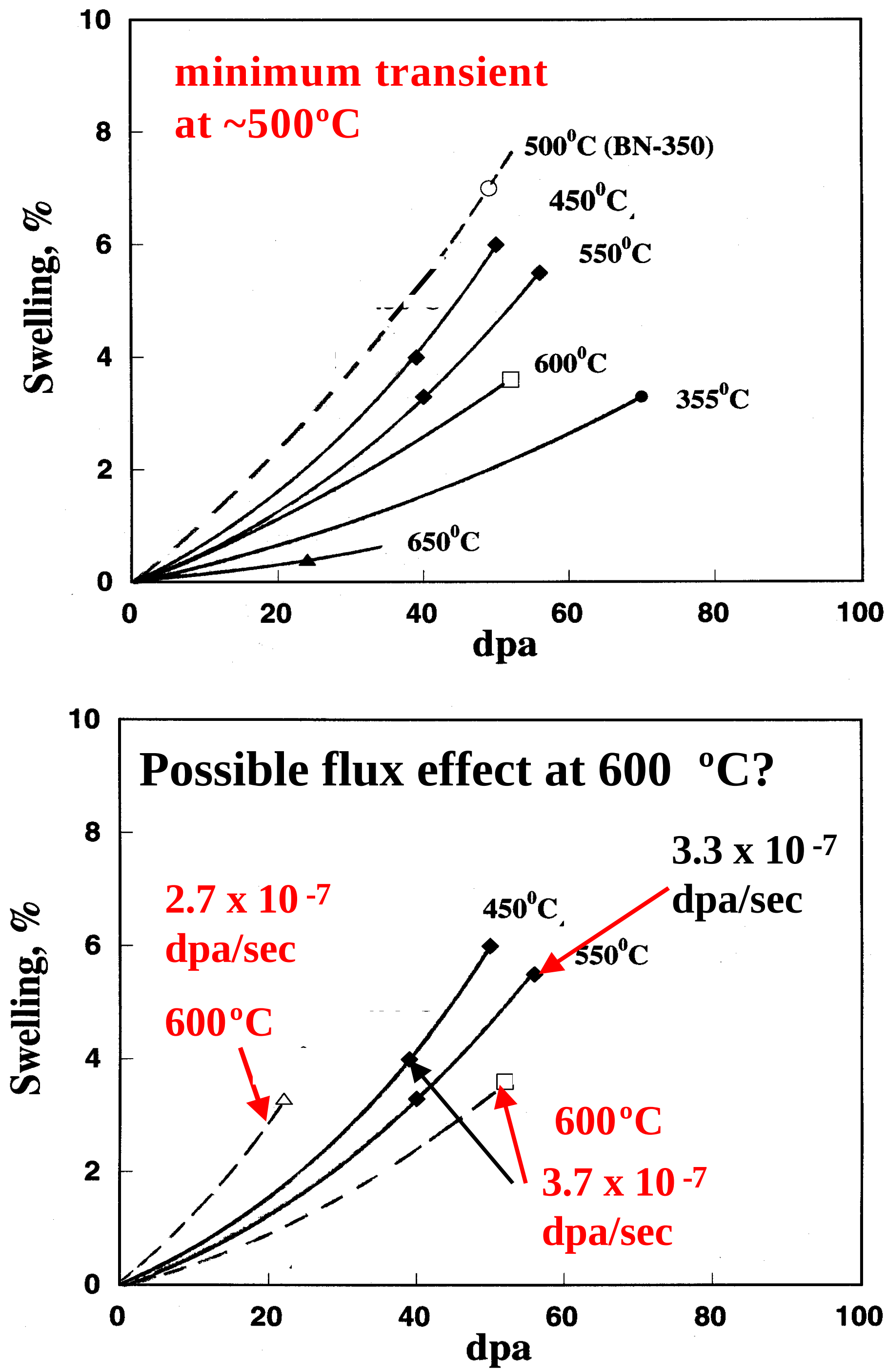

Figure 2. (top) Swelling observed in pure nickel, plotted vs. dpa for various temperatures. 2 (bottom) Swelling of a subset of irradiation conditions, indicating a possible influence of dpa rate on the duration of the transient regime, especially at 600°C.

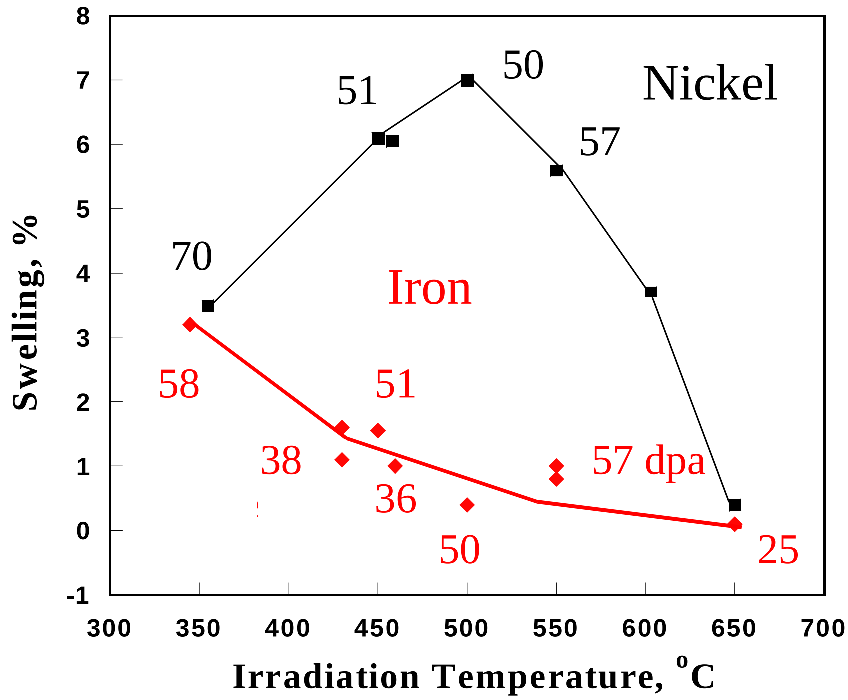

Figure 3. Comparison of the swelling of nickel and iron at comparable irradiation conditions. Dpa levels are indicated for each data point. Data pairs at 430 and 550°C indicate the reproducibility of swelling in this experiment.

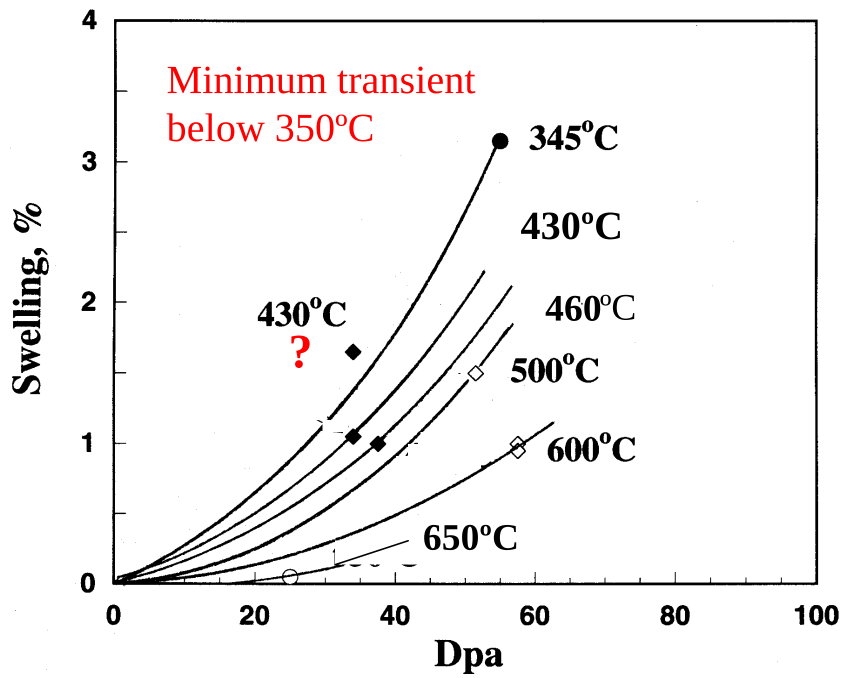

Figure 4. Swelling observed in pure iron, plotted vs. dpa for various temperatures.

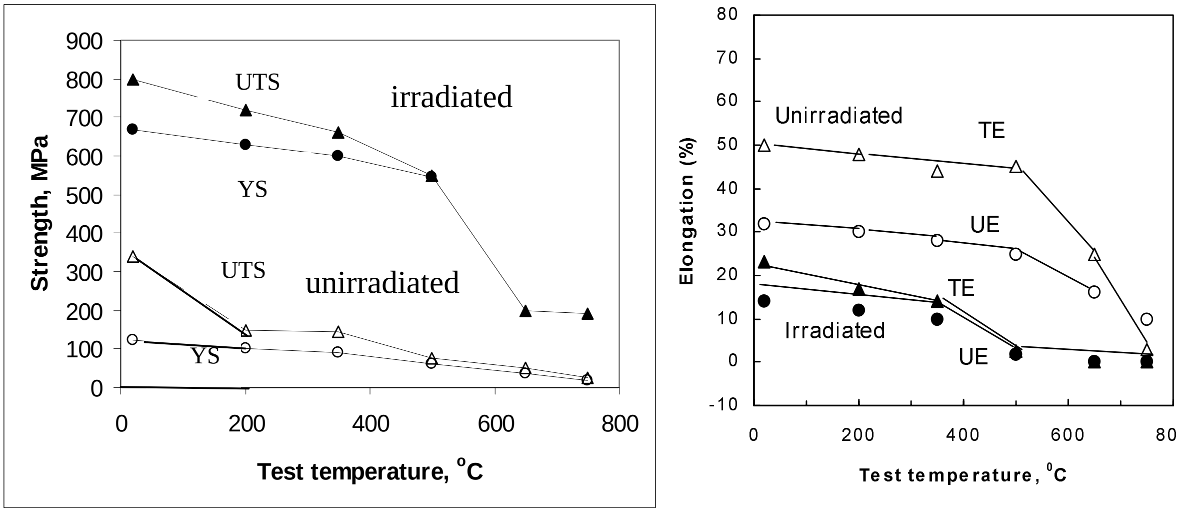

Figure 5. Strength and elongation characteristics vs. test temperature for both unirradiated and irradiated nickel (70 dpa, 355°C).

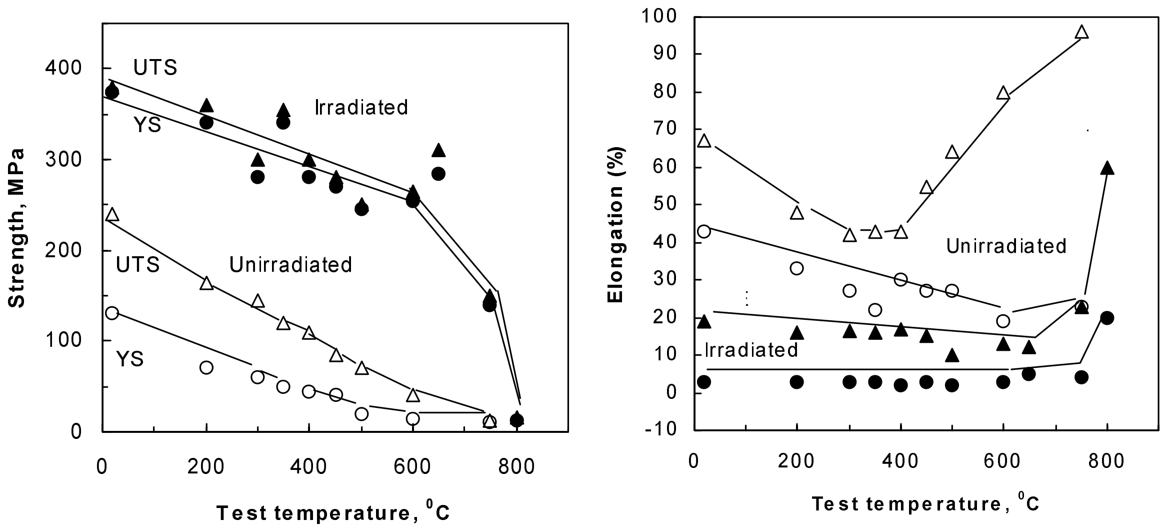

Figure 6. a) Strength and elongation characteristics vs. test temperature for both unirradiated and irradiated iron (58 dpa, 345°C).

It appears that above ~350°C nickel always swells more than iron at a given set of dpa rate and temperature, but it can not be stated with certainty that this statement is also true below 350°C.

The transient regime of swelling in nickel is clearly becoming longer with decreasing temperature below 500°C, while that of iron is clearly decreasing. It is thought to be quite likely that the peak swelling temperature (minimum transient) of iron has never been clearly observed because of the high inlet temperatures associated with fast reactors.

Data from other experiments at lower exposures has suggested that the peak swelling temperature of pure iron was in the range 400-425°C, but the higher exposure data from this experiment implies that such a conclusion reflected the combined influence of dpa rate and temperature on the duration of the transient regime.

The maximum swelling rate of pure nickel observed in this experiment appears to be on the order of ~0.2%/dpa but is still increasing with dpa. This value is well below the ~1%/dpa observed in other experiments [4,15,16]. It is significant to note that higher swelling rates were observed in experiments operating at lower displacement rates, producing significantly shorter transient regimes of swelling. Another paper addresses the effect of lower dpa rate to decrease the duration of the transient regime of swelling [17].

The maximum swelling rate observed in pure iron in this experiment is on the order of ~0.1%/dpa and still appears to be increasing with exposure. Recent estimates of the steady state swelling rate of pure iron and Fe-Cr binary model alloys range from 0.2%/dpa to 0.5%/dpa [9,12,18,19], considerably higher than earlier estimates.

The evolution of mechanical properties during irradiation is known to arise from all microstructural components, including voids, dislocations, loops and gas content, as well as the crystal structure of the metal. Although iron and nickel reached essentially the same swelling level at 345-355°C, their post-irradiation ductility behaved quite differently.

Iron exhibited significant hardening at 345°C, losing most of its uniform elongation, but still retaining 10-20% total elongation over the test temperature range. At higher test temperatures there was a partial recovery of ductility, however.

Nickel, however, exhibited high temperature embrittlement, having lost all ductility for test temperatures above 500°C. While nickel has a f.c.c. crystal structure and is therefore not prone to a DBTT shift, nickel does generate significantly more helium and hydrogen gas via transmutation, especially when compared to iron [20-21]. It is thought that collection of these gases, especially helium, at grain boundaries is facilitated at higher test temperatures, leading to grain boundary separation during tensile deformation. Unfortunately, it was not possible to examine the failure surfaces in this experiment, and this speculation cannot be confirmed.

Conclusions

When pure iron and nickel are irradiated side-by-side at a given set of dpa rate and irradiation temperature, it is possible to draw conclusions concerning their relative behavior with respect to void swelling and embrittlement.

It appears that both nickel and iron exhibit a transient-dominated swelling behavior in the range of 2 to 15×10^{-7} dpa/sec, with the shortest transient at ~500°C in nickel, but at <350°C for iron. It also appears that the duration of the transient regime may be dependent on the dpa rate.

When the two metals are irradiated at 345-355°C, it is possible to obtain essentially the same swelling level, but the evolution of mechanical properties is somewhat different. Both metals exhibit radiation hardening but the ductility losses are very different, especially at higher test temperatures, with partial recovery of ductility possible in iron. Nickel, however, exhibits high temperature embrittlement, thought to arise from the collection of helium gas at the grain boundaries. Iron generates much less helium during equivalent irradiation.

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