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Neutron-induced swelling and embrittlement of pure iron and pure nickel irradiated in the BN-350 and BOR-60 fast reactors

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Abstract

Pure iron and nickel were irradiated in the range of 2–15 × 10^{-7} dpa/s at 345–650 °C to very high neutron exposures in two fast reactors, BOR-60 and BN-350, to study void swelling and changes in mechanical properties of these two metals. Both nickel and iron swell in this temperature range with the maximum swelling rate at ~500 °C in nickel, but possibly at ⩽350 °C for iron. It also appears that the swelling rate in nickel and possibly in iron may be dependent on the dpa rate, increasing with decreasing dpa rate. The evolution of mechanical properties of the two metals is quite different. The differences reflect the fact that b.c.c. iron is subject to a low-temperature embrittlement arising from a shift in ductile–brittle transition temperature, while f.c.c. nickel is not. Nickel, however, exhibits high temperature embrittlement, thought to arise from the collection of transmutant helium gas at the grain boundaries. Iron is not strongly affected by transmutation since it generates much less helium during equivalent irradiation.

1. 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. Similar challenges exist in the modeling of mechanical properties as they evolve in response to radiation. The construction of theoretical models and computer simulations involving multi-scale modeling is facilitated by benchmark 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 significant void swelling at much lower doses than do even the simplest of binary model alloys, and therefore Ni and Fe were addressed in this study.

While pure nickel has been irradiated a number of times to rather high neutron exposures [1–7] to obtain swelling information, data on pure iron is in general more limited in dpa levels 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 simple metals at high neutron exposure levels.

This study focuses on the results of high dose (22–70 dpa) irradiations of pure iron and nickel in two fast

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reactors over a wide range of temperature, 345–650 °C. Both swelling and mechanical property data were collected.

2. 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 tensile specimens, which were 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 types were measured to determine their density using hydrostatic weighing in CCl_4, the second type was also 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 min at 850 °C and then water-quenched. Some of the cylinder specimens were irradiated in the core of the BN-350 fast reactor in contact with flowing sodium at 500 °C, reaching 50 dpa at 1.5×10^{-6} dpa/s. 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 and 70 dpa. 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 limited check on the reproducibility of the swelling process.

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 and 3.7×10^{-7} dpa/s. While most canisters were filled with cylindrical specimens, nine of the iron tensile specimens were irradiated at 345 °C to 58 dpa, and six of the nickel tensile specimens were irradiated at 355 °C to 70 dpa. In most cases iron and nickel were irradiated at identical dpa rates at a given temperature. At some irradiation temperatures there were sometimes two specimens reaching different dpa levels in different canisters. However, both the dpa rate and time in reactor were varied to reach the two final exposures.

Archive specimens were used to measure the density and mechanical properties of unirradiated material. Tensile tests were conducted under argon gas at 1 mm/min at test temperatures ranging from 20 to 800 °C.

3. Experimental results

Fig. 1 presents the swelling observed in pure nickel, showing an apparent peak swelling temperature at ~500 °C. Fig. 2a presents the majority of the BOR-60 swelling data plotted vs. dpa level, and shows that the temperature dependence of swelling is expressed primarily in the average swelling rate, as best as can be judged by ‘curves’ defined by only one and sometimes two data points. The maximum swelling rate appears to lie at ~500 °C. The BN-350 datum at 500 °C appears to confirm this conclusion even though swelling was developed at a much higher dpa rate in BN-350.

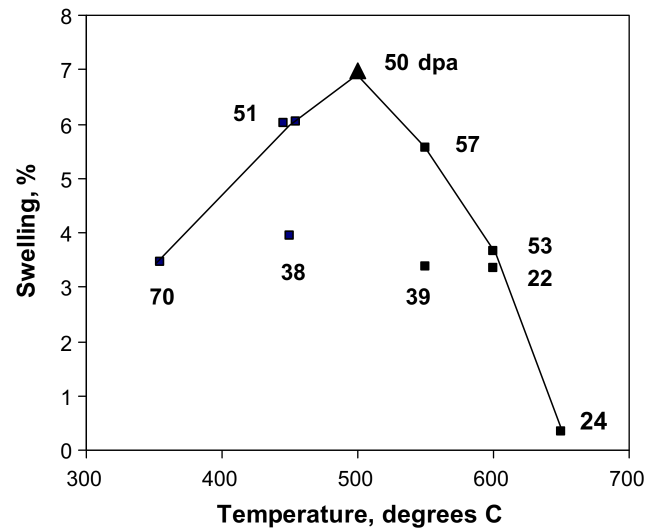

Fig. 1. Swelling observed in pure nickel irradiated in BOR-60 (squares) and BN-350 (triangle at 500 °C only) vs. irradiation temperature. Dpa levels are indicated for each data point. The two ‘artificially separated’ data points at 51 dpa and 450 °C indicate that the swelling of identical specimens in a single canister is rather reproducible.

Irradiation at each temperature proceeds at a different dpa rate in each capsule. Thus, there is the possibility that the apparent temperature dependence may actually result from the combined influence of two variables, temperature and dpa rate. This is especially obvious, as shown in Fig. 2b where swelling at 600 °C was developed at 2.7 and 3.7×10^{-7} dpa/s to reach 22 and 53 dpa, respectively. Swelling at 600 °C clearly proceeds faster at the lower dpa rate. If the rate effect were to be ignored then one might reach the erroneous conclusion that swelling had essentially saturated at 22 dpa.

The effect of dpa rate at 450 and 550 °C in this experiment is not as pronounced. This situation arises because the difference in dpa rates is not very large, with radiation proceeding at 3.7×10^{-7} and 3.3×10^{-7} dpa/s. Additionally, the lower dose level was reached at the higher dpa rate, as shown in Table 1. Note, however, that the swelling rate observed at both temperatures at the lower dpa rate is marginally higher than that at the higher dpa rate, confirming the generality of the rate effect in this experiment.

Table 2 presents the irradiation conditions and swelling data for pure iron. Fig. 3 shows a comparison of the higher fluence swelling data for Ni with the data for pure iron. Comparison of the BN-350 data at 500 °C for Fe and Ni at an identical dpa rate of 1.5×10^{-6} dpa/s clearly shows that at 500 °C Fe swells much less than Ni.

At ~350 °C the swelling of pure Fe at 58 dpa is essentially equal to that of Ni at 70 dpa. Note that at all higher temperatures iron swells less than nickel in the BOR-60 irradiation series. The swelling rate appears to decrease

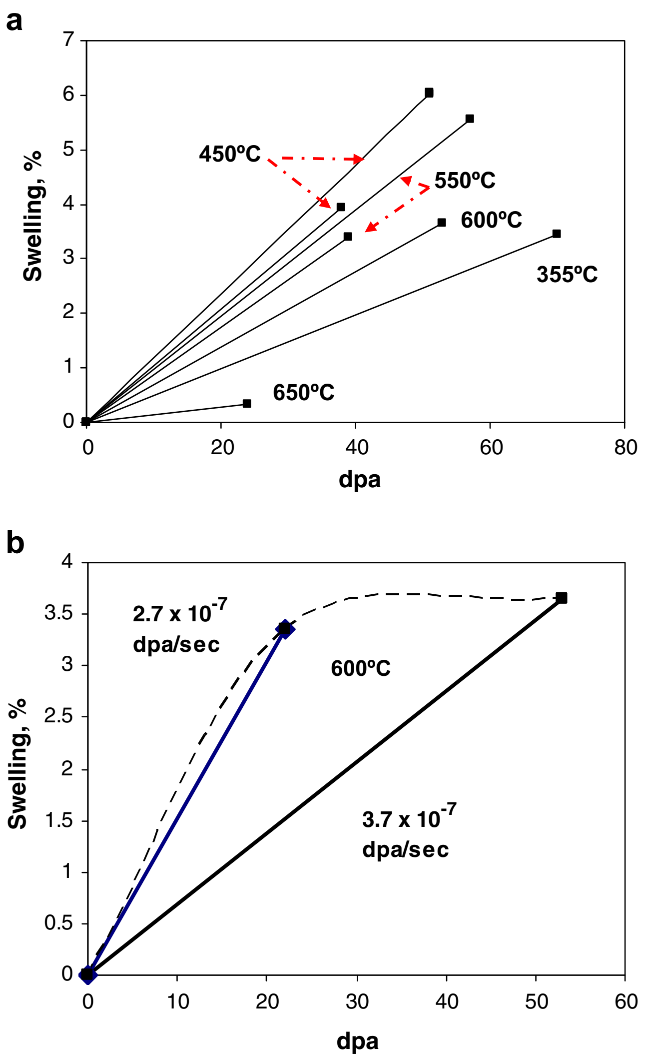

Fig. 2. (a) Swelling (minus one datum at 600 °C and 22 dpa) observed in pure nickel irradiated in BOR-60, suggesting that a peak swelling rate lies near ~500 °C. (b) Two data points at 600 °C are shown where radiation proceeded at two dpa rates. One interpretation suggests that the swelling rate increases at lower dpa rates. The second interpretation ignores the difference in dpa rate and suggests that swelling saturates with increasing dose. The latter interpretation is felt to be erroneous and misleading, however.

Table 1
Irradiation conditions and swelling for pure nickel

Temperature (°C)	Dose (dpa)	Dpa rate (dpa/s)	Swelling, $-\Delta\rho/\rho$ (%)
Multiple specimens			
450	38	3.7×10^{-7}	3.9
450	51	3.3×10^{-7}	6.0
550	39	3.7×10^{-7}	3.3
550	57	3.3×10^{-7}	5.5
600	22	2.7×10^{-7}	3.2
600	53	3.7×10^{-7}	3.6
Single specimens			
355	70	2.2×10^{-7}	3.5
500	50	1.5×10^{-6}	7.0
650	24	3.7×10^{-7}	0.3

Table 2
Irradiation conditions and swelling for pure iron

Temperature (°C)	Dose (dpa)	Dpa rate (dpa/s)	Swelling, $-\Delta\rho/\rho$ (%)
345	58	1.8×10^{-7}	3.2
430	38	3.7×10^{-7}	1.2; 1,5
450	51	3.3×10^{-7}	1.5
460	36	3.7×10^{-7}	1.0
500	50	1.5×10^{-6}	0.4
550	57	3.3×10^{-7}	0.7; 1.0
650	25	3.7×10^{-7}	0.2

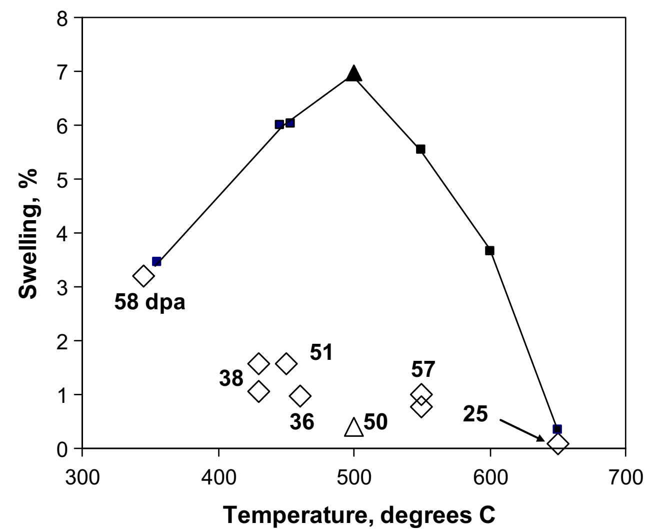

Fig. 3. Comparison of the swelling of nickel (closed symbols) and iron (open symbols) at comparable irradiation conditions in BOR-60 and BN-350, with the latter shown as triangles. Dpa levels are indicated for each data point. Data pairs at 430 and 550 °C indicate some relatively small variability of swelling for identical specimens irradiated in the same capsule.

continuously as the temperature falls from 345 to 650 °C, with the peak swelling of iron possibly occurring at a temperature ⩽350 °C as shown in Fig. 4. This is not a fully confident conclusion, however, since there are no data between 345 and 430 °C, possibly missing a peak temperature nearer 400 °C.

Fig. 4 also suggests that the swelling rate of pure iron might be sensitive to the dpa rate. Note that the BN-350 datum at 500 °C, 50 dpa and 1.5×10^{-6} dpa/s lies significantly lower than the level expected from the lower flux BOR-60 data.

Fig. 5 presents 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 during irradiation. Most importantly, the elongation falls to zero in irradiated nickel when tested above ~500 °C.

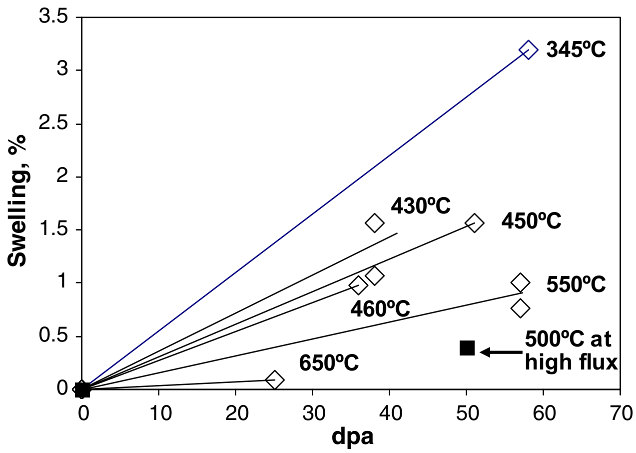

Fig. 4. Swelling observed in pure iron in BOR-60 (open diamonds) plotted vs. dpa, showing that the peak swelling rates lies at ⩽345 °C. Note that at 430 and 550 °C some small variation was observed in swelling of identical specimens. Also shown is the 500 °C datum generated at higher dpa rate in BN-350.

Fig. 6 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.

Fig. 6 also 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. The reason for the rise in elongation at higher test temperatures is not understood, but obviously signals a change in failure mechanism. 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.

4. Discussion

The temperature dependence of void swelling in both iron and nickel appears to arise primarily from the temperature and flux dependence of the steady-state swelling rate. Above ~350 °C nickel always swells more than iron at a given set of dpa rate and temperature conditions, but it cannot be stated with certainty that this statement is also true below 350 °C.

It is thought to be quite likely that the peak swelling temperature of pure iron at high dpa rates has never been clearly observed because of the high inlet coolant temperatures (>365 °C) associated with most fast reactors. BOR-60 and BN-350, however, have inlet temperatures on the order of 320 °C.

Data from other experiments at lower exposures [10,11] indicates that the peak swelling temperature of pure iron was in the range 400–425 °C, but the higher exposure data, especially at 345 °C, from the current experiment implies that the peak swelling temperature may depend on the combined influence of dpa rate and temperature on the swelling rate.

The temperature regime of void swelling is thought to be determined by the competition between three processes, namely increasing vacancy mobility with increasing temperature, void nucleation in response to vacancy supersaturation, and vacancy emission from void surfaces. Vacancy supersaturation increases with increasing dpa rate and vacancy emission increases with increasing temperature. In pure metals such as iron and nickel this competition is thought to produce a temperature-dependent swelling rate that peaks in the middle of the swelling regime. The swelling regime shifts with changes in dpa rate, moving to higher temperatures with increasing dpa rate [15]. In most pure metals the incubation dose is very small, essentially zero dpa, and the dose rate dependence is reflected primarily by an increase in swelling as the swelling regime moves

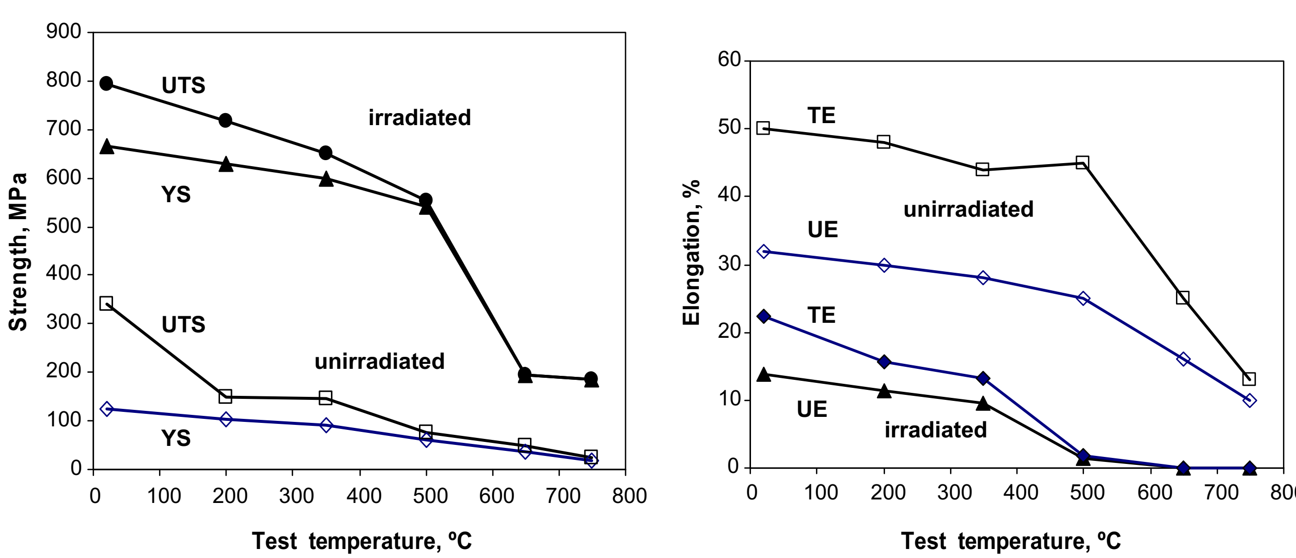

Fig. 5. Strength and elongation characteristics vs. test temperature for both unirradiated and irradiated nickel (70 dpa, 355 °C).

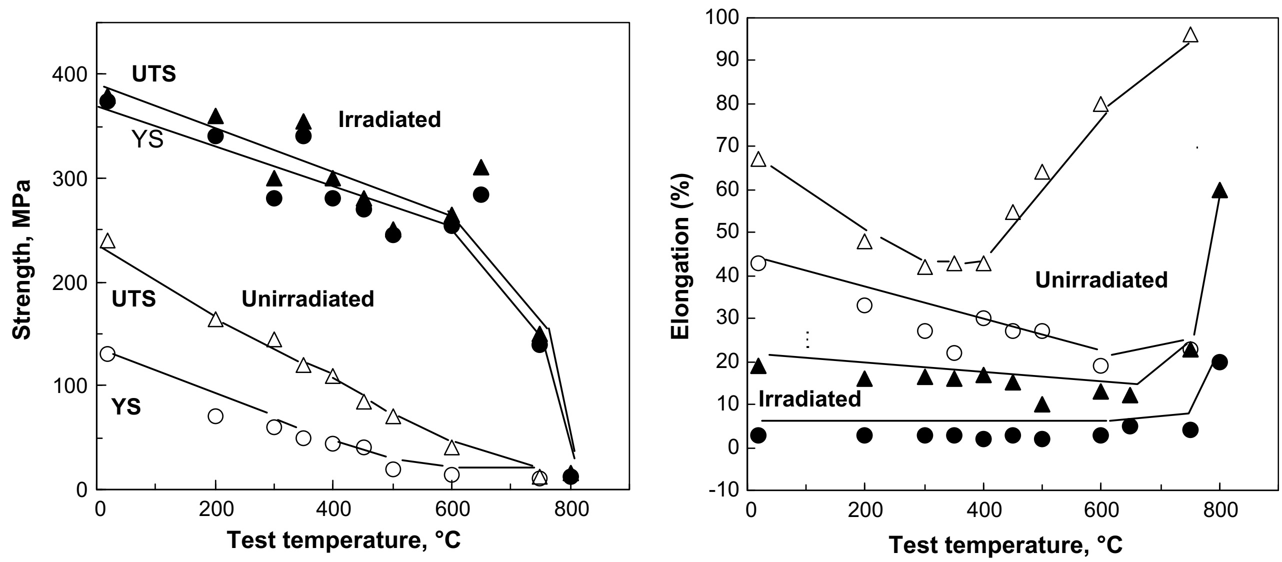

Fig. 6. Strength and elongation characteristics vs. test temperature for both unirradiated and irradiated iron (58 dpa, 345 °C).

down in temperature with decreasing dpa rate. Such behavior was observed in nickel irradiated in this study.

In order to explain the difference in peak swelling temperature for pure iron and nickel, however, it is necessary to demonstrate that differences in metal properties will affect the balance of these three processes. Odette has compiled a list of factors thought to affect the swelling of alpha iron and ferritic alloys [16]. He notes that alpha iron has much higher self-diffusion rates than gamma iron, Fe–Cr–Ni alloys and pure nickel. The self-diffusion coefficient of alpha iron is ~30 times greater than that of gamma iron in the temperature regime of swelling. In effect, the homologous temperature of alpha iron is significantly higher than that of pure nickel at a given real temperature. This factor alone would tend to shift the swelling regime of iron to lower real temperatures.

Additionally, as discussed later, helium generation is much lower in iron than in nickel, making it more difficult to stabilize voids against dissolution, especially at higher temperatures. Such an influence would also tend to push the peak swelling rate toward lower temperatures.

The maximum swelling rate of pure nickel observed in this experiment appears to be on the order of ~0.15%/dpa. This value is well below the ~1%/dpa observed in other experiments [4,17,18]. It is significant to note that higher swelling rates were observed in those experiments that operated at lower displacement rates.

In this experiment nickel did not reach or exceed the saturation swelling level of 8–10% as identified earlier as characteristic of pure nickel by Garner [19]. Saturation of swelling in nickel has been observed to result from collapse of the dislocation network into the voids [5,6,18], leading to a sharp drop in the dislocation density. The higher dpa rates of this experiment most likely retard swelling sufficiently to preclude reaching saturation until doses >50 dpa are attained. At lower dpa rates reached in the BR-10 fast reactor the swelling rate is indeed increased significantly [18].

The maximum swelling rate observed in pure iron in this experiment is on the order of ~0.1%/dpa. Recent experimental estimates of the maximum steady-state swelling rate of pure iron and Fe–Cr binary model alloys range from 0.2%/dpa to 0.5%/dpa [9,12,20,21], considerably higher than earlier estimates. Interestingly, Sniegowski and Wolfer predicted theoretically that alpha iron would exhibit a maximum swelling rate of ~0.2% per dpa compared to ~1%/dpa for gamma iron and pure nickel [22].

While not shown directly in this paper, it also appears from earlier studies that the steady-state swelling rate of Fe also increases as the dpa rate decreases [9,12].

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. This behavior is consistent with the well-known ductile–brittle transition temperature (DBTT) phenomenon and its shift with radiation exposure.

Nickel, however, exhibited embrittlement at high temperature, 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 [23,24]. 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.

5. Conclusions

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

It appears that both nickel and iron develop swelling in the range of $2–15 \times 10^{-7}$ dpa/s, with the maximum swelling rate at ~500 °C in nickel, but possibly at <350 °C for iron. It also appears that the swelling rate may be dependent on the dpa rate, increasing as the dpa rate decreases.

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 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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