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A review of some effects of helium on charpy impact properties of ferritic/martensitic steels

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Abstract

To evaluate the effect of helium on Charpy impact properties of ferritic/martensitic steels, two approaches are reviewed: quantification of results of earlier tests performed by other researchers on specimens irradiated in reactors with very different neutron spectra, and evaluation of isotopic tailoring experiments. Data analysis can show that if the differences in reactor response are indeed due to helium effects, then irradiation in a fusion machine at 400°C to 100 dpa and 1000 appm He will result in a ductile-to-brittle transition temperature (DBTT) shift of over 500°C. However, it can be shown that the response as a function of dose and helium level is unlikely to be simply due to helium based on physical reasoning. Shear punch tests and microstructural examinations support this conclusion based on irradiated samples of a series of alloys made by adding various isotopes of nickel in order to vary the production of helium during irradiation in High Flux Isotope Reactor (HFIR). The addition of nickel at any isotopic balance to the Fe–12Cr base alloy significantly increased the shear yield and maximum strengths of the alloys. However, helium itself, up to 75 appm at over 7 dpa appears to have little effect on the mechanical properties of the alloys. This behavior is instead understood to result from complex precipitation response. The database for effects of helium on embrittlement based on nickel additions is therefore probably misleading and experiments should be redesigned to avoid nickel precipitation.

1. Introduction

Ferritic/martensitic steels are being considered for use as structural materials in fusion power systems. In that application, they must not only withstand radiation damage, but also accommodate helium, a transmutation product, to levels as high as several thousand parts per million by end of life. The issue of helium accumulation on properties has been an ongoing concern, but lacking a high energy neutron source capable of high dose, design of unambiguous experiments to study such effects is very difficult.

Many review papers in the recent proceedings of these conferences have concluded that helium appears to play an important role on embrittlement of ferritic/martensitic steels [1–5]. Recently, results on helium effects have been obtained from an isotopic tailoring experiment wherein alloys were prepared containing 1.5% nickel additions of different nickel isotopes to an Fe–12Cr base alloy so that following irradiation in a mixed spectrum reactor such as the High Flux Isotope Reactor (HFIR) different levels of helium could be achieved by the two step nuclear reaction. Results included both mechanical properties based on shear punch testing and microstructural characterization [6]. Also, the data base that best appeared to demonstrate an effect of helium on Charpy impact testing has been quantified to establish the observed behavior as a function of helium content, and the results are difficult to understand based on a helium mechanism controlling deformation [7]. This paper is intended to review those results for the fusion materials development community.

2. Quantitative description of the effect of helium on charpy impact response

The database for the effect of irradiation on shift in ductile-to-brittle transition temperature (DBTT) for

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miniature Charpy V notch specimens is extensive including data for 9Cr–1MoVNb (T91), 12Cr–1MoVW (HT9), and an alloy with extra nickel additions, 12Cr–1MoVW–2Ni (HT9 + 2Ni) following irradiation in the HFIR and the Experimental Breeder Reactor (EBR) – II [7]. The database includes estimates of the helium levels generated in each of the specimen conditions. If these data are considered as a quantitative basis for a helium effect, and data on 9Cr–1MoVNb–2Ni (T91 + 2Ni) and 12Cr–1MoVW–1Ni (HT9 + 1Ni) are added, plots can be generated for relevant temperature ranges of interest as shown in Fig. 1 [7]. Fig. 1(a)–(c) show the shift in DBTT in °C as a function of the specimen helium content for the temperature ranges 55–60°C, 300°C, and 390–400°C, respectively. From these figures, the following can be deduced. At 60°C, large scatter exists in any trend of shift in DBTT as a function of helium content. At 300°C, scatter is still large, but a trend can be identified that shows the shift in DBTT to be approximately linear with increase in helium content, with an inherent shift that can be estimated at about 120°C for low helium levels, and may be sensitive to alloy composition. At 400°C, this trend is more clearly defined such that linear response is predicted, with an inherent shift of about 200°C. From these plots, it can be anticipated that helium does not have a major influence at 60°C, whereas at 300°C and 400°C, increasing helium levels contribute to large increases in shift in DBTT.

The trends identified in Fig. 1(b) and (c) are difficult to explain based on physical arguments limited to helium embrittlement. Such arguments would require that T91 and HT9 be extremely sensitive to low levels of helium, such that levels on the order of 2 appm He are responsible for a shift of approximately 200°C in DBTT. We know of no other element that will produce such drastic degradation in fracture toughness. Furthermore, additions of helium beyond the 2-appm helium level appear to cause linear increases in shift in DBTT, but at a much lower effective rate of increase than at the lowest helium levels. Therefore, a different mechanism is likely to be responsible.

Linear extrapolation of the behavior found in Fig. 1(c) can provide the prediction that at 1000 appm He, corresponding to a dose of about 100 dpa in a fusion environment, the shift in DBTT should be more than 500°C. If helium were responsible for the behavior shown in Fig. 1(c) at high helium contents, then it is apparent that use of martensitic steels in a fusion environment that produces helium in the 1000 appm range is unwarranted.

3. Isotopic tailoring

An experiment with fewer variables has been developed based on isotopic tailoring. A series of alloys has been made adding various isotopes of nickel in order to vary the production of helium during irradiation by the two step nuclear reaction in a mixed spectrum reactor [6]. The alloys used a base composition of Fe–12Cr with an addition of 1.5% nickel, either in the form of ^{60}Ni which produces no helium, ^{59}Ni which produces helium at a rate of about 10 appm He/dpa, or natural nickel which provides an intermediate level of helium due to delayed development of ^{59}Ni. Specimens were irradiated as transmission electron microscopy (TEM) disks in the HFIR at Oak Ridge, TN to about 10 dpa at 300°C, 400°C, 500°C and 600°C, and examined. Recently, results from that experiment following irradiation to low dose were published [6] and the results are summarized here.

Table 1 provides a summary of the experimental conditions. Note that helium production in heats E62 and R168 is very low but is in the range where large shifts in DBTT of about 170°C are predicted from Fig. 1(c), whereas R168 and R169 have sufficient helium levels to account for shifts only somewhat higher in magnitude to 210–230°C.

Results of shear punch testing are provided in Figs. 2 and 3 based on at least two tests for each specimen condition listed in Table 1. Shear punch testing is essentially a blanking operation which is common to sheet metal forming. A 1 mm diameter punch is driven at a

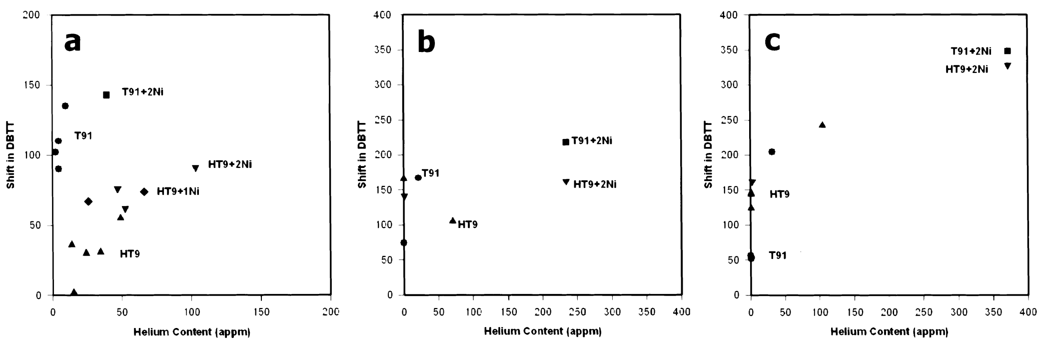

Fig. 1. Shift in DBTT as a function of helium content following irradiation at 55–60°C in (a) at 300°C in (b) and at 400°C in (c).

Table 1
Isotopic tailoring experimental details. All impurities not listed are ⩽ 0.01 w/o

Heat #	Nominal	Cr	Ni	C	Other	Irradiation conditions [a]	Code
E62	Fe–12Cr	11.6	na [b]	0.002	$0.026O_2$	300°C/6.5 dpa/2.1He	6A5M
						400°C/7.3 dpa/2.4 He	6A5N
						500°C/7.9 dpa/2.6He	6A5O
						600°C/8.2 dpa/2.7He	6A5P
R168	Fe–12Cr–1.5^{60}Ni	11.7	1.32	0.004	0.02Si	300°C/6.4 dpa/2.1He	715M
					0.02Mn	400°C/7.2 dpa/2.3He	715N
						500°C/7.8 dpa/2.5He	715O
						600°C/8.1 dpa/2.6He	715P
R169	Fe–12Cr–1.5^{59}Ni	na	na	na	na	300°C/6.6 dpa/70He	735M
						400°C/7.5 dpa/76He	735N
						500°C/8.0 dpa/82He	735O
						600°C/8.3 dpa/86He	735P
R170	Fe–12Cr–1.5^{Nat}Ni	11.5	1.54	0.004	0.02Si	300°C/6.5 dpa/41He	745M
					0.02Mn	400°C/7.4 dpa/46He	745N
						500°C/7.9 dpa/50He	745O
						600°C/8.2 dpa/51He	745P

[a] Irradiation temperature/dose/helium production in appm.
[b] na: not available.

constant rate of 0.127 mm/min (0.005 in./min) through a TEM-sized disk (nominally 0.25 mm thick and 2.8 mm in diameter) and the load on the punch is measured as a function of punch travel, providing an effective shear yield strength (τ_{sy}) and an effective maximum shear strength (τ_{sm}). Reproducibility was good: effective shear strength typically varied by no more than 30 MPa between duplicate specimens. Fig. 2 shows a summary of τ_{sy} as a function of helium content. Fig. 3 shows a similar plot for τ_{sm}. In each case, data for the unirradiated

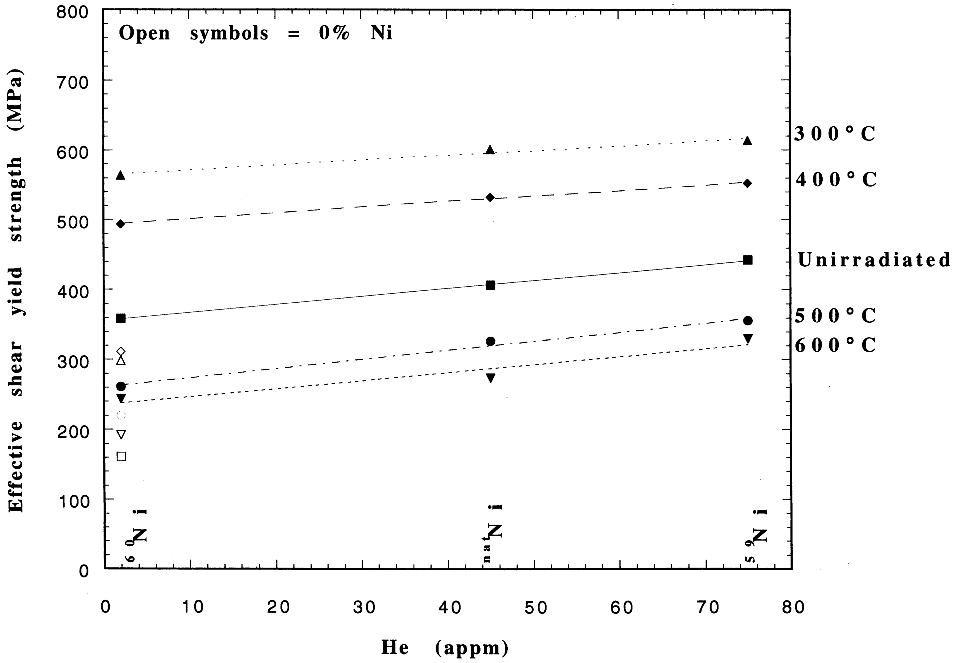

Fig. 2. Effective shear yield strengths (τ_{sy}) in Fe–12Cr–1.5Ni as a function of helium content (an open symbol signifies the control alloy [Fe–12Cr] at the same condition as the corresponding filled symbol).

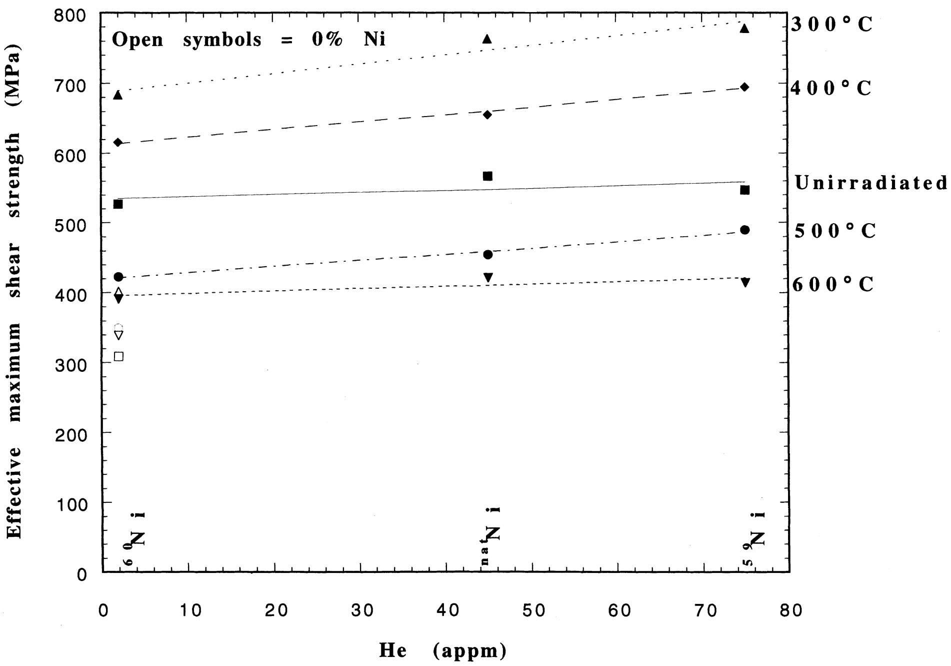

Fig. 3. Effective maximum shear strengths (τ_{sm}) in Fe–12Cr–1.5Ni as a function of helium content (an open symbol signifies the control alloy [Fe–12Cr] at the same condition as the corresponding full symbol).

material is included for comparison and alloy conditions containing no intentional additions of nickel are indicated by open symbols. From the plots of τ_{sy} and τ_{sm}, it can be seen that the addition of nickel to the Fe–12Cr base alloy significantly increases the strength of both the unirradiated and the irradiated alloys, especially for irradiation temperatures of 300°C and 400°C. The yield and maximum shear strengths are increased by about 100% compared to the unirradiated condition, a result which is independent of the nickel isotope balance used. In general, the strength of all alloys decreased with increasing irradiation temperature so that the highest strength was observed for alloys irradiated at 300°C. Alloys irradiated at 500°C and 600°C actually experienced an overall decrease in strength when compared to the unirradiated condition. However, for the Fe–12Cr material with no nickel added, strengths were similar following irradiation at 300°C and 400°C. A small increase in both τ_{sy} and τ_{sm} can be seen with increasing helium content in the irradiated alloys. Since the same trend is echoed in the data for the unirradiated material, however, it cannot be attributed to the helium level, but rather to another factor inherent to the alloys.

Microstructural examinations provided an explanation for the observed behavior. Microstructural damage resulting from irradiation in HFIR was only on a fine scale. At low magnification, the scale of the damage was seen as a fine background mottle. However, the mottling in specimens irradiated at 400°C was coarser than that from 300°C, and the alloy without nickel at 400°C contained mottling that was equiaxed whereas the mottling in the alloys containing nickel was non-equiaxed. Examples are provided in Fig. 4 allowing comparison of each of the conditions irradiated at 300°C and 400°C shown in bright field contrast.

In order to further emphasize that nickel additions lead to precipitation Fig. 5 has been prepared showing precipitate dark field images in specimens 715N with ^{60}Ni and 735N with ^{59}Ni, both irradiated at 400°C. In both cases, the precipitate dark field image was taken for an orientation near $(\bar{2}\,1\,0)$ for $\vec{g} \approx \frac{2}{3}(4\,2\,0)$ and the bright field image was without further tilt. The areas chosen for publication include larger precipitate particles located on subgrain boundaries, in order to emphasize that particle growth was accelerated at boundaries. Although particle sizes for these two conditions appear to be similar based on the dark field images, the bright field images emphasize the greater complexity that appears to arise from the added helium. Certainly, it must be concluded that nickel additions cause Fe–12Cr to develop complex microstructures due to precipitation.

4. Discussion

The experimental results from the isotopic tailoring experiment can be summarized as follows:

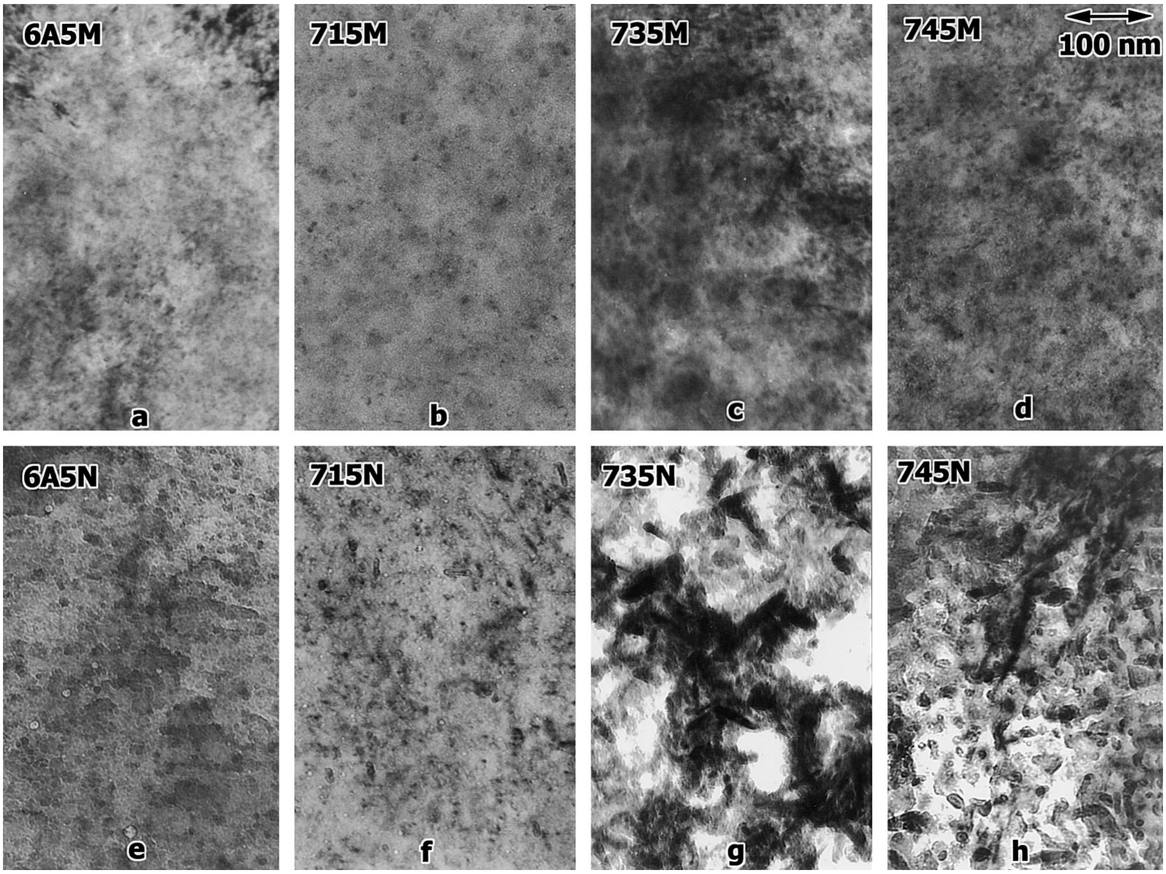

Fig. 4. Void Contrast Images for Fe–12Cr specimen 6A5M (a), Fe–12Cr–1.5^{60}Ni specimen 715M (b), Fe–12Cr–1.5^{59}Ni specimen 735M (c), and Fe–12Cr–1.5^{Nat}Ni specimen 745M (d) irradiated at 300°C, and Fe–12Cr specimen 6A5N (e), Fe–12Cr–1.5^{60}Ni specimen 715N (f), Fe–12Cr–1.5^{59}Ni specimen 735N (g), and Fe–12Cr–1.5^{Nat}Ni specimen 745N (h) irradiated at 400°C.

Nickel additions promote strength increases and precipitation in all alloys, but the strength increases are larger following irradiation at 300°C than at 400°C, whereas when no nickel is added, strengths are similar for the two irradiation temperatures. However, the cause of the additional strength for the 300°C conditions could not be elucidated by microstructural examination, probably because the features were too small to be resolved.

Helium bubble development for high helium generation conditions appeared to be very different at 300°C and 400°C. At 300°C, it appeared that high densities of bubbles formed whereas at 400°C, bubbles could not be identified, possibly because of the complexity of the microstructure, but more likely because helium accumulated at precipitate interfaces. However, the bubble-like features found following irradiation at 300°C may have been associated with precipitates; no through focus imaging was attempted.

These results indicate that the addition of nickel isotopes to ferritic/martensitic steels in order to provide understanding of helium effects adds the complicating factor of precipitate formation in interpretation of response. However, the isotopic tailoring concept can be employed without reliance on nickel additions. Greenwood and coauthors [8] have pointed out that certain iron isotopes can also provide enhanced helium production. Suzuki and coworkers [9] have initiated a similar experiment intended to investigate effects of hydrogen production using the same approach. Therefore, it appears feasible to design an isotopic tailoring experiment based on fabrication of alloys using iron isotopes to investigate not only effects of helium production during irradiation, but also effects of hydrogen production.

5. Conclusions

Analysis of available charpy impact data generated on specimens irradiated in different reactors has been used to quantify response as a function of helium

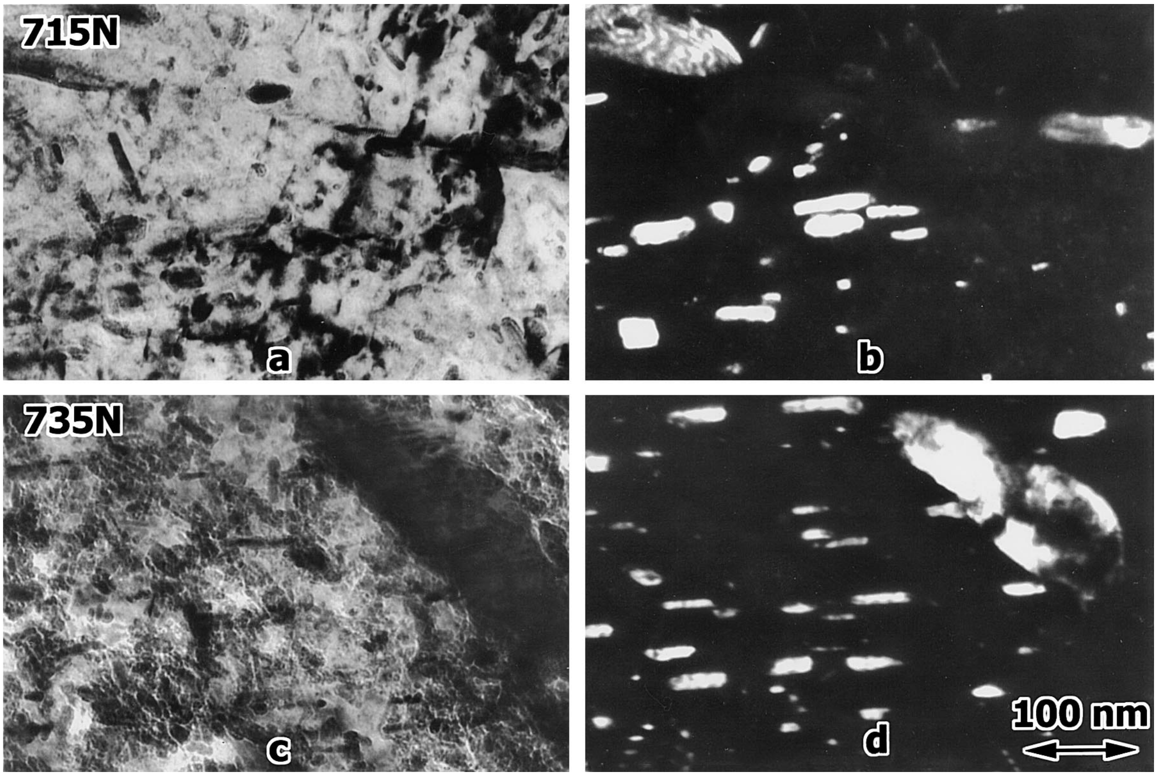

Fig. 5. Bright and dark field images of precipitation in Fe–12Cr–1.5^{60}Ni specimen 715N and Fe–12Cr–1.5^{59}Ni specimen 735N irradiated at 400°C to 7 dpa.

content. Analysis indicates that no trend applies following irradiation at 55–60°C, but at 300°C and 400°C, it is found that shift in DBTT increases rapidly at low helium levels and then continues to increase linearly at a lower rate providing the prediction that at 1000 appm He, the shift in DBTT may be as large as 500°C. However, it is difficult to understand why ferritic/martensitic steels should be so sensitive to helium at low levels or why the rate of increase should decrease and become linear at higher levels.

Isotopic tailoring by additions of 1.5% nickel of ^{60}Ni, ^{59}Ni, and ^{Nat}Ni to a base alloy Fe–12Cr have produced ferritic/martensitic alloys with a range of helium levels following irradiation in the HFIR to 7 dpa at 300–600°C. Shear punch tests showed that helium levels up to 75 appm have little, if any, effect on the effective shear yield and maximum shear strengths. The strengthening effect of nickel was evident prior to irradiation and the strength of the irradiated Fe–12Cr–1.5Ni ferritic alloys shows a strong dependence on irradiation temperature, decreasing with increasing irradiation temperature. This response is confirmed by microstructural examinations that revealed that nickel additions promote precipitation in all alloys. Therefore, future studies on helium effects in ferritic/martensitic steels should not be based on nickel additions because large microstructural changes arise from nickel precipitation making comparison with typical ferritic/martensitic steels unrealistic. Instead isotopic tailoring using iron should be considered.

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