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Embrittlement of irradiated F82H in the absence of irradiation hardening

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ABSTRACT

Neutron irradiation of 7–12% Cr ferritic/martensitic steels below 425–450 °C produces microstructural defects and precipitation that cause an increase in yield stress. This irradiation hardening causes embrittlement, which is observed in a Charpy impact or fracture toughness test as an increase in the ductile-brittle transition temperature. Based on observations that show little change in strength in steels irradiated above 425–450 °C, the general conclusion has been that no embrittlement occurs above these temperatures. In a recent study of F82H steel, significant embrittlement was observed after irradiation at 500 °C, but no hardening occurred. This embrittlement is apparently due to irradiation-accelerated Laves-phase precipitation. Observations of the embrittlement of F82H in the absence of irradiation hardening have been examined and analyzed with thermal-aging studies and computational thermodynamics calculations to illuminate and understand the embrittlement during irradiation.

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1. Introduction

The effects of neutron irradiation on the mechanical properties of commercial and reduced-activation ferritic/martensitic steels have been studied extensively [1–9]. Below 425–450 °C, irradiation damage hardens the steels, causing an increase in yield stress and ultimate tensile strength and a decrease in ductility [1–5]. Irradiation hardening affects toughness, and the effect is observed qualitatively in a Charpy impact test as an increase in the ductile–brittle transition temperature (DBTT) and a decrease in upper-shelf energy (USE) [6–11]. The magnitude of the shift varies inversely with irradiation temperature, similar to the variation in hardening.

For irradiation above 425–450 °C, tensile properties are generally unchanged, although enhanced softening may occur over that caused by thermal-aging alone, depending on fluence and temperature [3–5]. In this paper, embrittlement of F82H at temperatures above the irradiation-hardening regime will be examined and discussed.

2. Experimental

The reduced-activation steel F82H-IEA heat (nominally Fe–7.5Cr–2.0W–0.15V–0.02Ta–0.09C) was irradiated in the High Flux Isotope Reactor (HFIR) in the normalized-and-tempered condition [12,13]. Normalization involved austenitizing at 1040 °C to transform the steel to austenite, after which it was air cooled to form 100% martensite. Tempering was at 750 °C for 1 h.

One-third-size pre-cracked Charpy (PCVN), Charpy V-notch (CVN), and 0.18 T disk-compact fracture toughness [DC(T)] specimens were irradiated in HFIR to ≈3–5 and 20 dpa at 300–500 °C [12,13]. The low-dose irradiation was in two capsules in reactor removable beryllium positions, and the 20 dpa irradiation was in a capsule in the target position [13]. Because of the different neutron fluxes in the two reactor positions, the time the capsules were in the reactor were similar (≈5000 h).

The PCVN, CVN, and DC(T) specimens were tested to determine the transition temperature before and after irradiation. Tensile specimens were irradiated to 4.7 and 20 dpa at 300 and 500 °C and tested at −100 °C, room-temperature, and at the irradiation temperature [12].

3. Results and analysis

3.1. Experimental observations

A large increase in yield stress was observed after irradiation in HFIR at 300 °C to 4.7 and 20 dpa, with most of the change occurring for the 4.7 dpa irradiation [12,13]. There was only a slight further increase after 20 dpa (Fig. 1(a)). Irradiation at 500 °C to 4.8 and 20 dpa had no effect on yield stress [12].

Fracture toughness transition temperature shifts were evaluated with the master curve methodology [12], and the shifts showed a pronounced effect of irradiation temperature (Fig. 1(b)). The greatest effect occurred for the lowest temperature irradiations, with the shift decreasing with increasing temperature. Unexpectedly, there was a shift of 33 °C for the 5 dpa CVN specimen

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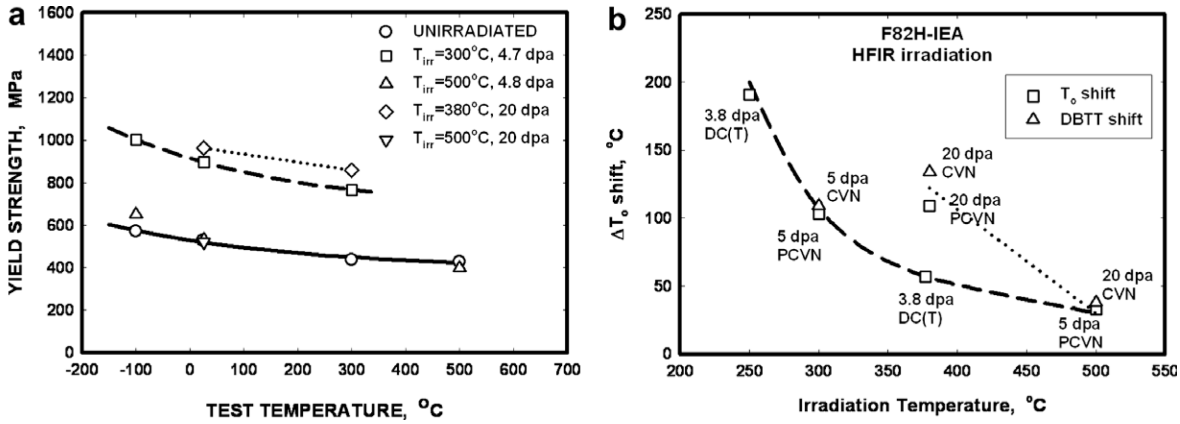

Fig. 1. The (a) yield stress as a function of test temperature and (b) transition temperature as a function of irradiation temperature for F82H irradiated in HFIR in the range 250–500 °C [13].

irradiated at 500 °C, and this was corroborated by a 38 °C shift for the 20 dpa CVN specimen [13].

3.2. Analysis

Based on thermal-aging observations of F82H discussed below, it is postulated that the increase in transition temperature in the absence of irradiation hardening can be traced to irradiation-accelerated Laves-phase [$(Fe,Cr)_2W$] precipitation and irradiation-accelerated coarsening of $M_{23}C_6$ particles. For normalized-and-tempered F82H tempered at 750 °C, the major precipitate is chromium-rich $M_{23}C_6$ with a small amount of MX, where M is vanadium and tantalum rich, and X is carbon and nitrogen. Calculations with the computational thermodynamics program JMatPro (v.3) [14] predicted that 1.9 wt% $M_{23}C_6$, 0.06 wt% MX, and 0.04% M_6C form during tempering at 750 °C. There is little change in the amount of the $M_{23}C_6$ and MX as temperature is decreased. Calculations above 400 °C indicate that the amount of Laves that forms decreases with increasing temperature with an abrupt cutoff at ≈640 °C, where 1.22% Laves is predicted, after which the amount is predicted to drop to zero by ≈650 °C (Fig. 2). However, M_6C is predicted to form below 756 °C and increase with decreasing temperature to 1.15% at 650 °C, where it then drops abruptly to zero at 639 °C.

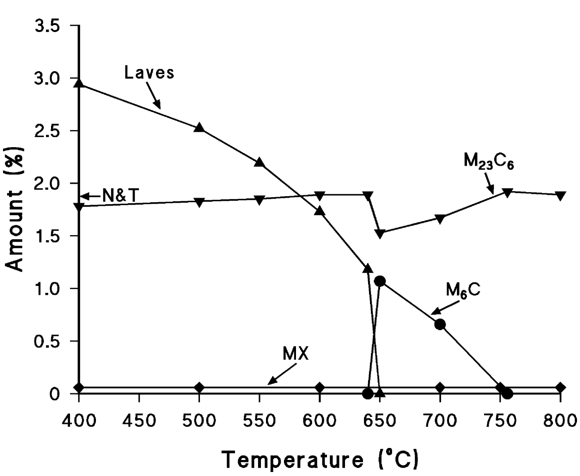

Fig. 2. Equilibrium amounts of $M_{23}C_6$, Laves-phase, M_6C, and MX precipitates calculated to form in F82H steel at 400–800 °C. Calculations were made with the computational thermodynamics program JMatPro.

Shiba thermally aged tensile and Charpy specimens of F82H-IEA for 1000, 3000, 10000, and 30000 h at 400 (aged 30000 h only), 500, 550, 600, and 650 °C. Although aging caused a reduction in room-temperature strength that was quite large at the highest aging temperatures and longest times (Fig. 3(a)), there was an adverse effect on impact properties (Fig. 3(b)) [15,16]. The largest strength decreases occurred at 600 and 650 °C; at 650 °C, a 33% decrease occurred after 30000 h.

Despite the large decrease in strength after aging at 650 °C, the largest increase in Charpy DBTT (105 °C) also occurred at this temperature after 30000 h. The DBTT also increased at the other temperatures, with the magnitude of the increase decreasing with decreasing aging temperature. The change was relatively small at 400 and 500 °C, even after 30000 h.

Precipitates were extracted from normalized-and-tempered and thermally aged specimens, and the amount extracted increased with aging time. After 10000 and 30000 h, the largest increase in the amount of precipitate occurred at 600 °C. Chemical and EDX analyses indicated that the largest elemental increases in the extracted precipitates were tungsten, iron, and chromium, all three of which are present in Laves-phase; M_6C and $M_{23}C_6$ are tungsten- and chromium-rich phases, respectively. X-ray diffraction of the precipitates verified the presence of Laves-phase at 550, 600, and 650 °C after aging for 10000 h. TEM observations indicated Laves-phase formed on $M_{23}C_6$ particles on prior-austenite grain boundaries and lath boundaries. Another effect observed was the coarsening of $M_{23}C_6$ particles.

When the total calculated amount of precipitate (the sum of the curves in Fig. 2) is compared to the measured precipitate after 30000 h, the effect of kinetics versus equilibrium is evident (Fig. 4). Equilibrium was reached by 30000 h at 650 °C [15], and it is being approached after aging 30000 h at 600 °C. Significant amounts of precipitate formed at 550 °C, but because of the reduced kinetics at 400 and 500 °C, little additional precipitate formed at these aging temperatures, even after 30000 h. Much more is to be expected with longer aging times. More details on the identification of the precipitates and a comparison of the extracted precipitates and those calculated with JMatPro have been published [17].

The 33 °C increase in transition temperature observed after irradiation of F82H to 5 dpa at 500 °C in HFIR was attributed to Laves-phase precipitation because the effect is much larger than was observed after 5000 h (the approximate time in reactor) of thermal-aging at 500 °C (Fig. 3). After thermal-aging 5000 h at 500 °C, there was no change. The increase after 5 dpa can be compared to the 28 °C increase during thermal-aging for 10000 h at

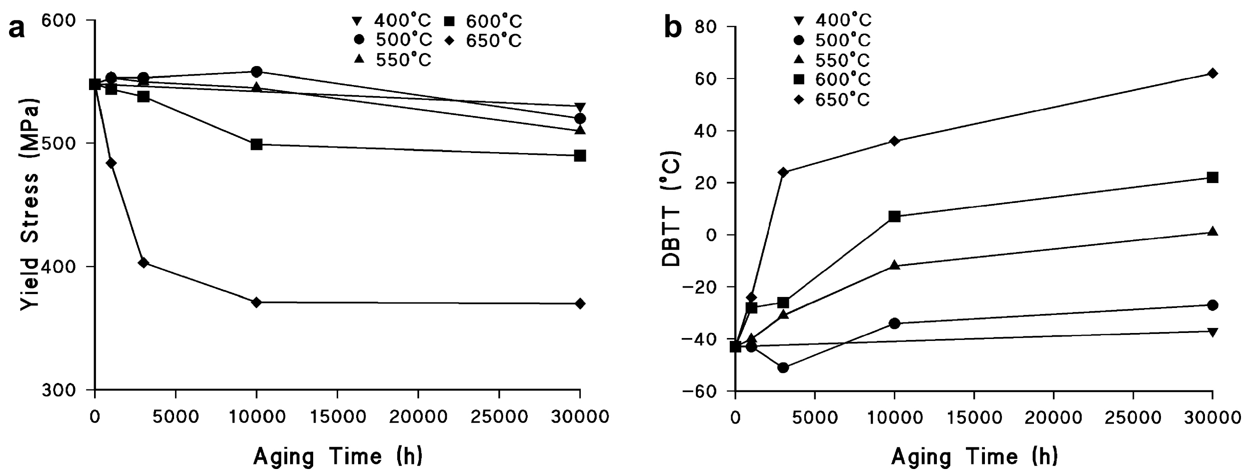

Fig. 3. The (a) yield stress and (b) ductile-brittle-transition temperature as a function of aging time at 400, 500, 550, 600, and 650 °C [15,16].

550 °C. The further increase from 33 to 38 °C is quite small considering the expected scatter in the data. However, note that in Fig. 3, the slope of the curve for 550 °C between 10000 and 30000 h is quite small, and the increase in transition temperature after 30000 h is ≈42 °C. The increase of 38 °C after irradiation at 500 °C is again much greater than that for thermal-aging for 5000 h at 500 °C. Therefore, it is concluded that irradiation at 500 °C accelerates the precipitation and coarsening kinetics, thus causing embrittlement even in the absence of irradiation hardening.

4. Discussion

The observations of moderate embrittlement of F82H in the absence of irradiation hardening were attributed to Laves-phase precipitation and $M_{23}C_6$ coarsening that occurred during irradiation. Fracture in steels is generally initiated at precipitate particles and/or inclusions with the critical stress to propagate a crack being inversely proportional to crack length [18,19]. If it is assumed fracture initiation occurs at a Laves-phase or $M_{23}C_6$ particle and the crack length at initiation equals the diameter of a particle, then fracture stress will decrease with increasing precipitate size.

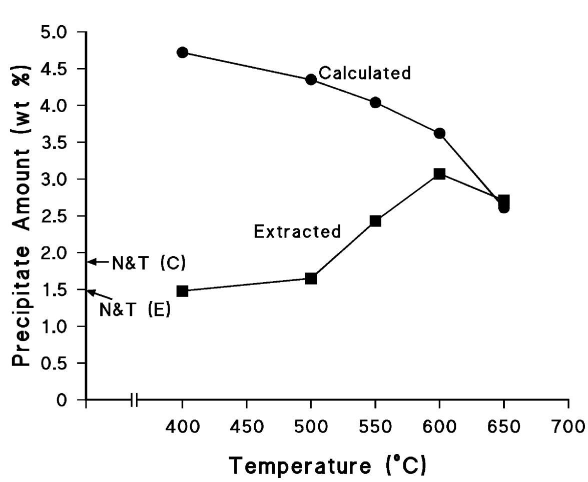

Fig. 4. Amount of extracted precipitate in normalized-and-tempered F82H steel thermally aged 30000 h at 400, 500, 550, 600, and 650 °C compared to calculated equilibrium amount of $M_{23}C_6$, Laves, M_6C, and MX phases. Amounts extracted and calculated for steel tempered at 750 °C are indicated by N&T (E) and N&T (C), respectively.

In the HFIR experiments containing the F82H-IEA heat that was irradiated to 5 and 20 dpa at 500 °C, the F82H-IEA heat with a different heat treatment (F82H-HT2), ORNL 9Cr-2WVTa steel (Fe–9.0Cr–2.0W–0.25V–0.06Ta–0.11C), and JLF-1 steel (Fe–9.0Cr–2.0W–0.20V–0.07Ta–0.10C) were also irradiated at 500 °C (Table 1) [20]. The latter two steels have compositions only slightly different from F82H. All of these steels had a positive ΔDBTT, but they all had a smaller ΔDBTT after the 5 dpa irradiation at 500 °C than the F82H-IEA heat. Laves-phase in an amount similar to that in F82H is calculated to form at 500 °C in all of these tungsten-containing steels.

The objective of the different heat treatment for F82H was to obtain a smaller prior-austenite grain size than the large grain size of F82H-IEA. After austenitizing at 920 °C instead of 1050 °C, the estimated ASTM grain size number increased from 3.3 to 6.5, corresponding to average grain sizes of ≈114 and ≈38 μm, respectively. The ORNL 9Cr-2WVTa and JLF-1 steels had ASTM grain size number 6, corresponding to ≈45 μm. They had smaller ΔDBTTs than the F82H-IEA, although they were somewhat larger than F82H-HT2.

Laves-phase and $M_{23}C_6$ form preferentially on prior-austenite grain boundaries, but the amounts formed do not depend on grain size. Therefore, if the ΔDBTT at 500 °C is caused by Laves-phase and $M_{23}C_6$ particles, the difference in the F82H with different heat treatments could be the result of the different grain sizes, assuming all else remains the same. If most of the Laves and $M_{23}C_6$ forms on prior-austenite grain boundaries, then a smaller grain size would provide a larger area for heterogeneous nucleation. This would be expected to result in a larger number of smaller precipitates, which could explain the observations based on the proposed crack-nucleation mechanism.

Thermodynamics calculations indicate that the amount of Laves-phase in reduced-activation steels at 500 °C depends mainly on tungsten content, and since ORNL 9Cr-2WVTa and JLF-1 contain 2% W like F82H, similar amounts of Laves are predicted for these

Table 1
Shift in transition temperature of steels irradiated in HFIR at 500 °C.

Material	Dose (dpa)	DBTT (°C)	ΔDBTT (°C)	Grain size No.
F82H-IEA	5	−54	30	3.3
	20	−46	38	
F82H (HT #2)	5	−92	9	6.5
ORNL 9Cr-2WVTa	5	−78	16	6
JLF-1	5	−66	19	6

steels as for F82H. The ORNL 9Cr-2WVTa and JLF-1 have relatively similar compositions and similar prior-austenite grain sizes, which are smaller than that of F82H-IEA. The ΔDBTTs for the two steels at 500 °C are similar and somewhat higher than for F82H-HT2 and about half that of F82H-IEA (Table 1). With time at temperature or for higher irradiation doses, the Laves-phase precipitation will be completed, and the particles will coarsen. It is expected that this will have a major effect on toughness, but the exact effect needs to be determined by long-time thermal-aging and/or higher-dose irradiation experiments.

The calculations indicated that the amount of Laves-phase depends on tungsten concentration, which implies embrittlement should increase for a steel with a higher tungsten concentration. This was verified by Sakasegawa et al., who tested Charpy specimens of JLF-1 with 2% and 3% W thermally aged at 550 and 650 °C [21]. Therefore, both experiment and calculations imply that the reduced-activation EUROFER 97 steel with 1% W should not be embrittled to the same extent observed for reduced-activation steels with 2% W.

Odette and co-workers [22,23] discussed fracture under similar circumstances to those presented in this paper, which they labeled non-hardening embrittlement (NHE). To describe the behavior, a multiscale model was proposed, which according to the authors is [23] 'based on the observation that cleavage occurs by the propagation of microcracks emanating from brittle trigger-particles, like large grain boundary carbides...'. They write that 'Local stress–strain concentrations due to incompatible matrix particle deformation cause some of the brittle ceramic trigger-particles to crack'.

5. Summary and conclusions

In the past, irradiation-effects studies in ferritic/martensitic steels have focused on temperatures where irradiation hardening occurs (<425–450 °C) that is accompanied by embrittlement (measured as an increase in ductile–brittle transition temperature) caused by a reduction in toughness. Embrittlement due to irradiation-enhanced precipitation in the absence of irradiation hardening has received relatively little attention.

In this paper, and increase in the ductile-brittle transition temperature of F82H steel increased when irradiated at 500 °C where no hardening occurred in a tensile test. The increase in transition temperature was attributed to Laves-phase precipitation and $M_{23}C_6$ coarsening. It occurred for a relatively low-dose (≈5 dpa) and a relatively low temperature (500 °C). Because of the low-dose (relatively short thermal exposure time) and relatively low temperature for extensive precipitation, the ultimate effect of the precipitates on properties has yet to be determined. Therefore, need exists for high-dose irradiations at higher temperatures (500–600 °C) than most previous irradiation experiments.

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