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Embrittlement of reduced-activation ferritic/martensitic steels irradiated in HFIR at 300°C and 400°C

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Abstract

Miniature tensile and Charpy specimens of four ferritic/martensitic steels were irradiated at 300°C and 400°C in the high flux isotope reactor (HFIR) to a maximum dose of ≈12 dpa. The steels were standard F82H (F82H-Std), a modified F82H (F82H-Mod), ORNL 9Cr–2WVTa, and 9Cr–2WVTa–2Ni, the 9Cr–2WVTa containing 2% Ni to produce helium by (n,α) reactions with thermal neutrons. More helium was produced in the F82H-Std than the F82H-Mod because of the presence of boron. Irradiation embrittlement in the form of an increase in the ductile–brittle transition temperature (ΔDBTT) and a decrease in the upper-shelf energy (USE) occurred for all the steels. The two F82H steels had similar ΔDBTTs after irradiation at 300°C, but after irradiation at 400°C, the ΔDBTT for F82H-Std was less than for F82H-Mod. Under these irradiation conditions, little effect of the extra helium in the F82H-Std could be discerned. Less embrittlement was observed for 9Cr–2WVTa steel irradiated at 400°C than for the two F82H steels. The 9Cr–2WVTa–2Ni steel with ≈115 appm He had a larger ΔDBTT than the 9Cr–2WVTa with ≈5 appm He, indicating a possible helium effect.

1. Introduction

Neutron irradiation in a fusion reactor will cause displacement damage and transmutation helium formation in the reactor structural materials. The irradiation effect of most concern for ferritic/martensitic steels is embrittlement below 400–450°C, exhibited as an increase in the ductile–brittle transition temperature (DBTT) and a decrease in the upper-shelf energy (USE) in a Charpy impact test. There is also evidence that transmutation helium in the steel can affect the Charpy impact behavior [1–5].

There have been several experiments to study the embrittlement of the reduced-activation steels irradiated to high fluences in fast reactors [6–8] and to low fluences in mixed-spectrum reactors [2,3,5] and research reactors [4,8,9]. The effect of helium on properties is important because of the helium to be generated by transmutation reactions in the first wall of a fusion plant. The thermal fluence in a mixed-spectrum reactor offers the possibility of generating transmutation helium by (n,α) reactions in the irradiated material if it contains ^{58}Ni or ^{10}B [1,2].

In this experiment, tensile and Charpy specimens of three reduced-activation martensitic steels were irradiated in the mixed-spectrum high flux isotope reactor (HFIR). To examine helium effects, a nickel-doped steel was also irradiated, and one of the steels contained boron.

2. Experimental

The nominal compositions (in wt%) of the steels irradiated are: (1) standard F82H (F82H-Std), Fe–7.5Cr–2W–0.2V–0.04Ta–0.0034B–0.1C; (2) modified F82H (F82H-Mod), Fe–7.5Cr–2W–0.15V–0.02Ta–0.1C; (3) ORNL 9Cr–2WVTa, Fe–9Cr–2W–0.25V–0.07Ta–0.1C; (4) 9Cr–2WVTa–2Ni, the 9Cr–2WVTa containing 2% Ni. The F82H-Std was from a 5000 kg vacuum-induction heat obtained by the Japan Atomic Energy Research Institute (JAERI) from NKK Corporation,

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Kawasaki, Japan. The F82H-Mod was from a 5 ton vacuum-induction heat also produced by NKK Corporation for collaborative research coordinated by an International Energy Agency (IEA) committee. The 9Cr–2WVTa and 9Cr–2WVTa–2Ni steels were from small 400-g vacuum arc-melted button heats produced at Oak Ridge National Laboratory.

Miniature Charpy specimens of F82H-Std and F82H-Mod were obtained from 15 and 7.5 mm plates, respectively. The button melts of 9Cr–2WVTa and 9Cr–2WVTa–2Ni were rolled to 6.4-mm thickness, from which the Charpy specimens were machined. Pieces of the plate for each steel were rolled to 0.76-mm thicknesses from which tensile specimens were obtained.

The steels were irradiated in the normalized-and-tempered condition. For the two F82H steels, austenitization was at 1040°C followed by tempering 1 h at 750°C. The 9Cr–2WVTa and 9Cr–2WVTa–2Ni steels were austenitized at 1050°C followed by tempering 1 h at 750°C.

Tensile specimens 25.4 mm long with a reduced gage section 7.62 mm long by 1.52 mm wide by 0.76 mm thick were machined from 0.76 mm sheet. Tests were conducted at the irradiation temperature in vacuum on a 120 kN Instron universal test machine at a nominal strain rate of $\approx 1 \times 10^{-3}$ s^{-1}. Charpy specimens were one-third size V-notch specimens measuring 3.3 × 3.3 × 25.4 mm^3 with a 0.51 mm deep 30° V-notch and a 0.05–0.08 mm root radius that were machined from normalized-and-tempered plate along the rolling direction with the notch transverse to the rolling direction (L–T orientation).

Two irradiation capsules with 16 specimen positions ≈25.4 mm long were irradiated in HFIR peripheral target positions with the specimens at nominal temperatures of 300°C and 400°C. At each position, either four 1/3-size Charpy specimens, four SS-3 tensile specimens, or about 100 transmission electron microscopy specimens in a 12Cr steel subcapsule were enclosed in 12Cr steel holders that were then placed inside aluminum sleeves. To control the temperature obtained from nuclear heating, the gas gap between the outer diameter of the steel specimen holder and the aluminum sleeve was adjusted to compensate for the variation in nuclear heating rate along the length of the capsule. A thermal gradient, calculated to be less than 45°C, exists from the interior to the exterior of the specimens.

Three flux monitors in each capsule were analyzed to determine fluence as a function of distance from the reactor midplane. Total maximum fluence at the midplane was 6.9×10^{26} n/m^2, with a thermal fluence of 3.2×10^{26} n/m^2 (<5 eV) and a fast fluence of 1.68×10^{26} n/m^2 (>0.1 MeV). Maximum displacement damage at the midplane was ≈12 dpa. Only specimens at the center position along the length of the capsule received the peak fluence. Therefore, displacement damage and helium concentration of a specimen varied depending on its position in the capsule relative to the reactor midplane.

Two tensile specimens and 4–6 Charpy specimens of the four steels were irradiated. The F82H steels were irradiated at 300°C and 400°C, and the 9Cr–2WVTa and 9Cr–2WVTa–2Ni were irradiated only at 400°C. Specimens irradiated at 300°C were at the ends of the capsules and received 9–10 dpa. Displacement damage in specimens irradiated at 400°C was 11–12 dpa.

3. Results

All steels hardened after irradiation (Table 1). Tensile data for the normalized-and-tempered F82H-Mod are not available from this experiment. Instead, data from another HFIR irradiation to ≈5 dpa at 300°C [10] are given in Table 1 (no data are available at 400°C). Comparison of the 300°C data shows an increase in yield stress of ≈75% for the F82H-Mod, which is somewhat greater than the 57% increase for the F82H-Std. Note that the F82H-Std was stronger than the F82H-Mod at 300°C before irradiation.

Before irradiation, the strength of the 9Cr–2WVTa was similar to that of the 9Cr–2WVTa–2Ni when tested at 400°C. Irradiation caused somewhat similar increase in the yield stress of the 9Cr–2WVTa–2Ni (41%) and the 9Cr–2WVTa (35%). Ductilities of the two steels before and after irradiation were also similar (Table 1).

Results for the Charpy tests are given in Table 2. The DBTT was obtained at half the upper-shelf value, the same procedure as was used in previous publications [1,6]. One problem is that because of the limited number of specimens irradiated, no upper-shelf was determined for the 9Cr–2WVTa–2Ni. Therefore, to estimate the DBTT for this steel, the same upper-shelf as that determined for the irradiated 9Cr–2WVTa was used as a conservative estimate (if the actual value is less, which it could well be, then the DBTT would be less). The steels had similar USE values before irradiation.

The largest shift in DBTT (ΔDBTT) occurred for the two F82H steels irradiated at 300°C. There was little difference in ΔDBTT of the two steels; the F82H-Std showed the larger decrease in USE (Table 2). At 400°C, where all four steels were irradiated, the 9Cr–2WVTa had the smallest shift (79°C), and the F82H-Mod had the largest shift (146°C), with intermediate values for the F82H-Std (117°C) and the 9Cr–2WVTa–2Ni (133°C). (Note that the ΔDBTT for F82H-Mod at 400°C was almost as large as it was for this steel at 300°C.) The percent decrease in the USE of the 9Cr–2WVTa (40%) was greater than that of the F82H-Std (21%) and F82H-Mod (23%). No USE was determined for 9Cr–2WVTa–2Ni for the reasons stated above.

Table 1
Tensile results for reduced-activation Cr–W steels

Steel	Temperature (°C)	Irradiation		Strength (MPa)		Elongation (%)	
		dpa	appm He	Yield	Tensile	Uniform	Total
F82H-Std	300			522	603	3.0	12.0
(8Cr–2WVTaB)	300	9	39	822	832	0.2	6.7
	400			464	532	2.8	11.6
	400	12	41	646	677	0.9	7.2
F82H-Mod	300			438	498	0.5	3.2
(8Cr–2WVTa)	300	5	2	765[a]	777	0.5	6.1
	400					0.8	3.5
	400					0.9	5.0
9Cr–2WVTa	400			715	817	1.6	4.5
	400	11	5	963	983	0.6	5.8
9Cr–2WVTa–2Ni	400			733	824	1.6	4.3
	400	11	114	1034	1075	0.6	5.7

[a] Data taken from another HFIR irradiation experiment.

Table 2
Charpy impact properties of reduced-activation Cr–W steels

Steel	Irradiation temperature (°C)	Irradiation		DBTT (°C)	ΔDBTT (°C)	USE (J)
		dpa	appm He			
F82H-Std	Unirradiated			−103		12.3
(8Cr–2WVTaB)	300	10	40	56	159	7.9
	400	12	41	14	117	9.7
F82H-Mod	Unirradiated			−82		10.8
(8Cr–2WVTa)	300	9	4	70	152	8.3
	400	11	4	64	146	8.3
9Cr–2WVTa	Unirradiated			−94		10.8
	400	11	5	−15	79	6.5
9Cr–2WVTa–2Ni	Unirradiated			−113		10.8
	400	11	115	21	133	n/m

Selected specimens from the lower- and upper-shelf regions of each steel at each irradiation temperature were examined by scanning electron microscopy (SEM). Lower-shelf fractures were typical cleavage fractures and the upper-shelf fractures were typical of ductile tearing, similar to the fracture mode of the steels in an unirradiated condition.

4. Discussion

One objective of this experiment was to determine the effect of helium on Charpy properties by comparing nickel- and boron-containing steels with steels of similar composition but without those elements. As F82H-Std contained ≈34 appm B, it contained ≈40 appm He, compared to ≈5 appm He in the F82H-Mod. Essentially the only difference between the 9Cr–2WVTa and 9Cr–2WVTa–2Ni is the 2% Ni in the latter steel, resulting in ≈115 appm He, compared to ≈5 appm He in the 9Cr–2WVTa.

No helium effects could be delineated in the F82H steels. The ΔDBTT of the F82H-Std (40 appm He) was similar to and less than that for the F82H-Mod (5 appm He) at 300°C and 400°C, respectively. The F82H-Mod had a larger change in yield stress at 300°C. Therefore, if helium plays a role in the embrittlement of F82H-Std, something else (e.g., more hardening, chemical composition differences, microstructural differences, etc.) in F82H-Mod causes a similar (at 300°C) or larger (at 400°C) embrittling effect.

One major difference between the F82H-Std and the F82H-Mod was the prior austenite grain size estimated to be 22 (ASTM No. 8) and 125 μm (ASTM No. 3), respectively. This could result in a lower DBTT for the F82H-Std before irradiation, as observed, but it is not known how this might affect the ΔDBTT.

Previous work on 9Cr–2WVTa indicated it had excellent resistance to embrittlement (as determined by the ΔDBTT) when irradiated in the fast flux test facility (FFTF) [6] and HFR [2], and this was attributed to tantalum in solution either raising the fracture stress or changing the flow behavior [11]. However, it was observed that the ΔDBTT increased with increasing fluence, which is opposite to most other steels, which show a saturation of ΔDBTT with fluence [6,11]. Further, for a given fluence at different temperatures, the ΔDBTT increased with increasing temperature, which is also opposite to what is observed for most steels. These contrary observations were attributed to a loss of tantalum from solution by precipitation during irradiation [11]. Recently, tantalum-containing precipitates were found in a 9Cr–2WVTa steel irradiated in FFTF at 460°C but not in one irradiated at 390°C [12], which is in agreement with the postulated effect [11]. It might be speculated that the reason for the larger ΔDBTT for the F82H-Mod at 400°C is that this steel, which has ≈0.02% Ta compared to ≈0.04% Ta for the F82H-Std loses a critical amount of its tantalum at the higher temperature before the F82H-Std does. Based on the previous results [2,6], this would explain the similar ΔDBTTs for the F82H-Mod at 300°C and 400°C.

Besides differences in boron and tantalum concentrations in the two F82H steels, there was also a slightly higher manganese concentration in the F82H-Std (0.49% vs 0.17%). Manganese can contribute to solid solution strengthening, but it is not clear how this might affect the irradiation behavior.

Thus, the greater irradiation hardening, the larger prior austenite grain size, and the lower tantalum and manganese concentrations in F82H-Mod relative to the F82H-Std may all increase the ΔDBTT of the F82H-Mod relative to the F82H-Std, whose properties might be affected by the higher helium concentration. However, since these effects cannot be quantified, it must be concluded that the effect of the helium is unclear.

When the results on F82H-Std are compared with other data for this steel [2,9], the present irradiation produced a much larger ΔDBTT. This steel had a shift of ≈50 and 40°C after 2.4 dpa in the mixed-spectrum HFR at 300°C and 400°C, respectively [2]. In both the present HFIR and previous HFR irradiations, essentially all ^{10}B was transmuted to helium by <2 dpa [2]. On the other hand, when notched Charpy specimens were irradiated to ≈67 dpa at 430°C in the FFTF – a fast reactor where much less helium is produced – essentially no embrittlement was observed, indicating that 430°C is above the temperature where irradiation hardening ceases [7]. Although not directly comparable because pre-cracked Charpy specimens were used, a smaller ΔDBTT (25–30°C) was observed after irradiation in FFTF to 15 dpa at 370°C [7]. Previous irradiations of F82H-Std at 230–350°C in the Japan Materials Test Reactor to 0.2–0.6 dpa indicated a helium effect by comparing the Charpy behavior of the F82H-Std with this composition containing 100 appm He obtained by adding ^{10}B [4]. Without the ^{10}B addition, a ΔDBTT of ≈50°C was observed. With the ^{10}B addition, the DBTT was well above room temperature (the highest temperature tested was 20°C), because the fracture energy remained on the lower-shelf at 20°C [4].

The results of the present tests, especially the comparison between the mixed-spectrum and fast reactor results for F82H-Std, could be interpreted to show a helium effect. However, the difference between the F82H-Std and F82H-Mod steels apparently contradicts such an interpretation, unless, as discussed above, other differences in F82H-Std and F82H-Mod exist.

Contrary to the F82H steels, which showed a much larger ΔDBTT after irradiation in HFIR in the present experiment than when irradiated in FFTF [7], the 9Cr–2WVTa steel showed a similar behavior in the HFIR irradiation and a previous irradiation experiment on a different heat of 9Cr–2WVTa steel irradiated to ≈14 dpa in FFTF [6]. In that experiment, the ΔDBTT at ≈400°C was 43°C, compared to 79°C for the heat of steel irradiated to ≈11 dpa in this HFIR experiment.

The 9Cr–2WVTa–2Ni steel was irradiated to determine if there was an effect of ≈115 appm He produced from the ^{58}Ni in the steel. The ΔDBTT of the 9Cr–2WVTa–2Ni (133°C) was 68% greater than for 9Cr–2WVTa (79°C). Irradiation hardening of the steels was similar. The yield stress of the 9Cr–2WVTa and 9Cr–2WVTa–2Ni increased 35% and 41%, respectively. The question is: Is the difference in ΔDBTT a helium effect? Similar observations for nickel-doped modified 9Cr–1Mo and Sandvik HT9 steels were taken as evidence of a helium effect [1,13].

The results are tentatively taken to be a helium effect. However, in previous work on the nickel-doped modified 9Cr–1Mo and Sandvik HT9, there were indications of possible intergranular fracture on specimens tested on the lower-shelf, and this led to the proposal that helium affected the fracture behavior of the steels by causing a change in fracture mode [13]. No indication of intergranular fracture was observed on the 9Cr–2WVTa–2Ni steel.

5. Summary and conclusions

Tensile and Charpy specimens of two 7.5Cr–2WVTa steels (F82H-Std and F82H-Mod), a 9Cr–2WVTa steel, and a 9Cr–2WVTa–2Ni steel were irradiated in HFIR to 9–12 dpa. Irradiation of the F82H-Std and F82H-Mod was at 300°C and 400°C, and the 9Cr–2WVTa and

9Cr–2WVTa–2Ni was at 400°C. Irradiation of the two F82H steels at 300°C produced similar relatively large increases in DBTT (>150°C). After irradiation of the four steels at 400°C, the 9Cr–2WVTa developed the smallest shift in DBTT (79°C), followed by the F82H-Std (117°C), the 9Cr–2WVTa–2Ni (133°C) and the F82H-Mod (146°C).

A primary objective of the experiments was to determine if helium affected the impact properties of the steels. The F82H-Std contained ≈40 appm He after irradiation compared to 4–5 appm in the F82H-Mod. No helium effect could be detected on the F82H steels, because despite the helium, it still had properties as good or better than those of the F82H-Mod. The 9Cr–2WVTa and 9Cr–2WVTa–2Ni steels had similar tensile properties before and after irradiation. However, the shift in DBTT for the 9Cr–2WVTa–2Ni was 68% higher than for the 9Cr–2WVTa. One possible explanation for this is the 115 appm He in the nickel-containing steel compared to ≈5 appm He in the 9Cr–2WVTa.

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