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IMPACT BEHAVIOR OF 9-Cr AND 12-Cr FERRITIC STEELS AFTER LOW-TEMPERATURE IRRADIATION*

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Abstract

Miniature Charpy impact specimens of 9Cr-1MoVNb and 12Cr-1MoVW steels and these steels with 1 and 2% Ni were irradiated in the High-Flux Isotope Reactor (HFIR) at 50°C to displacement damage levels of up to 9 dpa. Nickel was added to study the effect of transmutation helium. Irradiation caused an increase in the ductile-brittle transition temperature (DBTT). The 9Cr-1MoVNb steels, with and without nickel, showed a larger shift than the 12Cr-1MoVW steels, with and without nickel. The results indicated that helium also increased the DBTT. The same steels were previously irradiated at higher temperatures. From the present and past tests, the effect of irradiation temperature on the DBTT behavior can be evaluated. For the 9Cr-1MoVNb steel, there is a continuous decrease in the magnitude of the DBTT increase up to an irradiation temperature of about 400°C, after which the shift drops rapidly to zero at about 450°C. The DBTT of the 12Cr-1MoVW steel shows a maximum increase at an irradiation temperature of about 400°C and less of an increase at either higher or lower irradiation temperatures.

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1. Introduction

A steel in a fusion reactor first wall will experience displacement damage from the high-energy neutrons from the fusion reaction. Displacement damage can lead to changes in the impact behavior as measured by Charpy impact tests [1-5]. In addition to displacement damage, large amounts of transmutation helium will also form in the first-wall material of a fusion reactor. To study the effect of simultaneously produced displacement damage and transmutation helium, nickel has been added to the 9-Cr and 12-Cr steels [6]. When these nickel-doped steels are irradiated in a mixed-spectrum reactor, such as the High Flux Isotope Reactor (HFIR), displacement damage is produced by the fast neutrons in the spectrum, and helium is produced by a two-step transmutation reaction of ^{58}Ni with the thermal neutrons in the spectrum. One objective of this work was the determination of the effect of helium on the impact properties.

2. Experimental Procedure

Electroslag-remelted heats of standard 9Cr-1MoVNb (0.1% Ni) and 12Cr-1MoVW (0.5% Ni) steels were prepared. These compositions with 1 and 2% Ni, designated 9Cr-1MoVNb-2Ni, 12Cr-1MoVW-1Ni, and 12Cr-1MoVW-2Ni, were also prepared. Specimens were obtained from normalized-and-tempered plates. The normalizing treatment for the 9-Cr steels was 0.5 h at 1040°C and for the 12-Cr steels 0.5 h at 1050°C, after which they were air cooled. The 9Cr-1MoVNb steel was tempered 1 h at 760°C; the 12Cr-1MoVW and 12Cr-1MoVW-1Ni steels were tempered 2.5 h at 780°C.

The 9Cr-1MoVNb-2Ni and 12Cr-1MoVW-2Ni steels were tempered 5 h at 700°C. Tempered martensite microstructures were obtained by such heat treatments. Details on chemical composition, heat treatment, and microstructure have been published [6].

Miniature Charpy V-notch (C_V) specimens were machined from the heat-treated plate in the longitudinal (LT) orientation. The subsize specimens measured 5 by 5 by 25.4 mm and contained a 0.76-mm-deep 30° V notch with a 0.05- to -0.08-mm-root radius. In the irradiated conditions, such miniature specimens show impact behavior similar to that found in full-sized C_V specimens [7,8].

The specimens were irradiated in three capsules in HFIR at the reactor-coolant temperature of ~50°C as follows: Specimens in the capsule designated HFIR-CTR-T3 were irradiated in the peripheral target region of the reactor to displacement-damage levels of 5.7 to 9.1 dpa. Capsules designated HFIR-CTR-RB1 and HFIR-CTR-RB2 were irradiated in the beryllium-reflector region to displacement-damage levels of 2.7 to 4.5 dpa and 4.3 to 9.2 dpa, respectively. The displacement-damage levels and helium concentrations of the specimens depended on the position of the specimen in the capsule; the helium concentration also depended on the amount of nickel in the material.

Tests were carried out in a pendulum-type impact machine specially modified to accommodate the subsize specimens. Detailed information on the test equipment has been published [8]. To obtain the ductile-brittle transition temperature (DBTT) and upper-shelf energy (USE), impact energy-temperature curves were generated by fitting the data with a hyperbolic tangent function. Because of the limited number of specimens available, the USE could not always be determined accurately. The DBTT was chosen at a fixed energy level of 9.2 J, which is analogous to the frequently used criterion of 68 J in a full-size specimen.

3. Results

The results are given in Table 1. There are difficulties with these types of tests. First, reactor space limitations restrict the number of specimens that can be irradiated per capsule. For RB1, there were six specimens for each of the two 9-Cr steels, four for 12Cr-1MoVW, five for 12Cr-1MoVW-1Ni, and only three for 12Cr-1MoVW-2Ni. In RB2, only four specimens were irradiated for each material except 9Cr-1MoVNb-2Ni, for which there were only two specimens. No data are given for 9Cr-1MoVNb-2Ni irradiated in RB2. The T3 capsule contained more specimens (seven or eight for each steel) but for only two steels: 12Cr-1MoVW and 12Cr-1MoVW-2Ni. A second problem with these types of irradiation experiments results from the variation in flux along the length of the capsule. This variation leads to different fluences for the different specimens of the same steel. Therefore, specimens with different displacement-damage levels and helium concentrations were used to produce a given Charpy impact curve.

The variations in displacement-damage level and helium concentration for each set of specimens are given in Table 1. Note that, although the 12Cr-1MoVW and 12Cr-1MoVW-2Ni steel specimens irradiated in RB2 had displacement-damage levels similar to those irradiated in

T3, these specimens contained more helium than those irradiated in RB2. This was caused by a higher thermal flux in the reflector region of the reactor where the RB2 capsules were located in the reactor. Transmutation helium is generated from nickel by thermal neutrons.

Table 1 gives the DBTT as measured at the 9.2 J level, the DBTT shift, (ΔDBTT), and the USE. Note that the data obtained in the T3 and RB2 experiments on the 12Cr-1MoVW and 12Cr-1MoVW-2Ni steels are in good agreement. This adds confidence to the results obtained from sets of specimens with only 3 or 4 specimens because 7 and 8 specimens were irradiated and tested for the two materials in the T3 experiment and only four specimens of each in the RB2 experiment.

4. Discussion

Previous work has shown that the nickel additions have no significant effect on the microstructures and the precipitates that form in the 9Cr-1MoVNb and 12Cr-1MoVW steels [9]. The differences in the Charpy behavior of the unirradiated nickel-doped and undoped steels were previously discussed [7]. A tempering treatment of 700°C was used for the steels containing 2% Ni because nickel raises the A_{C_1} temperature. Although the 760 and 700°C tempering temperatures for the 9Cr-1MoVNb and 9Cr-1MoVNb-2Ni steels, respectively, resulted in somewhat similar hardnesses, the strength of the 9Cr-1MoVNb was less than that for 9Cr-1MoVNb-2Ni [6]. Generally, the steel tempered to the lowest strength is expected to have the lowest DBTT, just the opposite to what was observed (Table 1).

There are indications that nickel decreases the DBTT and the USE in pearlitic steels [10]. Chromium, molybdenum, and tungsten have the opposite effect. Assuming the same effect occurs in a martensitic structure, these alloying effects would explain the observations on the 9-Cr steels. Such large effects were not observed for the 12-Cr steel.

As expected, the results on the irradiated specimens showed that neutron irradiation caused an increase in DBTT. A relatively large decrease in USE accompanied the increase in DBTT for the 9-Cr steels, but the 12-Cr steels showed relatively small changes in USE. The decrease in USE for the 9Cr-1MoVNb was progressively larger with increasing displacement-damage level. The 9Cr-1MoVNb-2Ni steel also showed a large decrease in USE following irradiation.

The results indicate a difference between the 9-Cr and 12-Cr steels. For the 9Cr-1MoVNb steel, there was a large increase in DBTT after irradiation to 2.3 to 3.4 dpa, with essentially no change for the higher dpa (4.3-5.7 dpa). The shift in DBTT for the 12-Cr steels was substantially lower: after ~4 dpa, only a small increase occurred. However, this curve was obtained with only four specimens, and one of the data points did not follow the expected sequence. (Note in Table 1 that this Charpy curve was the only one that had a large decrease in the USE, which could be a further indication that this curve is inaccurate.) Because of the displacement of this one point, the DBTT calculated by the curve-fitting procedure is probably inaccurate, and the USE is higher than that given in Table 1. However, there was good agreement in the results for the 12Cr-1MoVW steel specimens irradiated to ~9 dpa in RB2 and T3. These curves showed a ΔDBTT of 31 and 36°C, respectively, with little change in USE. Thus, the increase in DBTT for the 9Cr-1MoVNb steel (102–110°C) was nearly three times that for the 12Cr-1MoVW steel (31–36°C).

Gelles et al. [11,12] tested pre-cracked Charpy specimens of 9Cr-1MoVNb and 12Cr-1MoVW that were irradiated in the RB1 and RB2 capsules. Although different heats of steel, slightly different heat treatments, and different orientations were irradiated, similar changes in DBTT were obtained: The DBTT of the 9Cr-1MoVNb and 12Cr-1MoVW steels increased 90 and 30°C, respectively [11], in RB1 and 130 and 62°C, respectively [12], in RB2.

The majority of the previous results were for specimens irradiated at elevated temperatures [1-5]. Irradiation of 12Cr-1MoVW steel in EBR-II to displacement-damage levels of 26 dpa (little helium) at 390°C gave rise to an increase in DBTT of 144°C [3], a considerably larger increase than observed for irradiation at 50°C. Similarly, specimens irradiated 4 to 9 dpa in HFIR at 300 and 400°C had ΔDBTTs of 164 and 217°C, respectively [4]. As the irradiation temperature was increased above 400°C, ΔDBTT decreased; it was 50°C at 550°C [3,5].

All of the data are shown in Fig. 1(a), which is updated from when it was previously presented [4]. The figure indicates that the increase in DBTT for 12Cr-1MoVW steel goes through a maximum around 400°C. This is in contrast with the data for the 9Cr-1MoVNb steel [Fig. 1(b)]. After irradiation at 390°C in EBR-II, the ΔDBTT for 9Cr-1MoVNb is only about 50°C [3], compared with 90 to 110°C after the

50°C irradiation; after irradiation at 450°C and above, no change in DBTT was observed [3]. Thus, the ΔDBTT-temperature curve for the 9Cr-1MoVNb steel shows decreases as the temperature increases from 50 to 400°C, after which it drops rapidly to zero.

The behavior observed for the 9Cr-1MoVNb steel is the expected behavior. An increase in DBTT is expected to result from the hardening caused by the displacement damage, and as the temperature is increased, some of the hardening is offset by recovery effects. Gelles et al. found black-dot damage in both the 9Cr-1MoVNb and 12Cr-1MoVW steels irradiated at 50°C, with no apparent difference between the two steels [11]. In tensile tests after irradiation at 50°C, similar amounts of hardening were observed for 9Cr-1MoVNb and 12Cr-1MoVW steels [13]. With an increase in the irradiation temperature, defects and some of the dislocations of the structure can anneal out, although some additional hardening can occur because of irradiation-induced precipitation [9,14]. Tensile tests on 9Cr-1MoVNb and 12Cr-1MoVW steels have shown that the strength was increased by irradiation at 400°C, but the strength increase was less than that after irradiation at 50°C [15,16]. After irradiation at 450°C and above, there was little change in strength compared to the unirradiated steel. These tensile results show a change in properties with irradiation temperature similar to that observed for the impact properties of the 9Cr-1MoVNb steel [Fig. 1(b)].

The previous results give no reason to expect a difference in behavior between 9Cr-1MoVNb and 12Cr-1MoVW. However, some significant differences have been observed in the precipitation in the 12Cr-1MoVW and 9Cr-1MoVNb during irradiation [9]. Maziasz et al. [9] observed that after irradiation in HFIR at 300 to 500°C, some of the $M_{23}C_6$ in the as-tempered microstructure of the 9Cr-1MoVNb steel had dissolved and the subgrain structure had coarsened. This contrasted with the $M_{23}C_6$ and subgrain structure in the 12Cr-1MoVW steel, which remained stable. Irradiation of the 9Cr-1MoVNb steel at 300 to 500°C also caused a dissolution of some of the finer MC particles and a coarsening of the larger particles. Some coarsening of MC also occurred in the 12Cr-1MoVW steel, but large amounts of irradiation-produced M_6C also formed.

The observed changes in the microstructure of the 9Cr-1MoVNb steel between 300 and 500°C could affect the DBTT. The coarsening of the subgrain structure should lower the DBTT. Thus, as the irradiation temperature increases, these recovery effects offset some of the hardening caused by the displacement damage. For the 12Cr-1MoVW steel, no softening due to subgrain coarsening occurs and the hardening effect due to irradiation-produced M_6C is superimposed on the production of displacement damage. Another way of looking at these results is that the 9Cr-1MoVNb steel behaves as expected, assuming that radiation hardening causes the DBTT shift. If the precipitation reactions did not occur in the 12Cr-1MoVW steel and it behaved as expected, the DBTT shift for this steel would be less than that for 9Cr-1MoVNb at all temperatures. However, the curve in Fig. 1(a) for the 12Cr-1MoVW steel consists of the expected curve [similar to Fig. 1(b), but with a lower shift at 50°C] with the effect of precipitation superimposed on it.

Helium appears to affect the impact properties. This is most clearly shown by the results for the nickel-doped and undoped 12-Cr steels (Table 1). The small amounts of data make definite conclusions difficult. However, for a given dpa (and helium concentration) the ΔDBTT values increase as the helium concentration (nickel concentration) is increased. Also, the differences in the results for the tests for experiments RB2 and T3 for the 12Cr-1MoVW-2Ni are noteworthy. The ΔDBTT for the tests from RB2 was 91°C and from T3 was 76°C. Both sets of specimens were irradiated to similar displacement-damage levels, but the specimens in T3 contained 36 to 58 appm He, while those from RB2 contained 93 to 114 appm. Furthermore, the ΔDBTT for the 12Cr-1MoVW-2Ni irradiated in T3 was similar to that for the 12Cr-1MoVW-1Ni irradiated in RB2. Similar displacement-damage levels and helium concentrations were achieved for both materials under these test conditions.

An interpretation of the difference in the results for the 9Cr-1MoVNb and 9Cr-1MoVNb-2Ni steels is difficult because of the different heat treatments used for these two steels. The 9Cr-1MoVNb-2Ni has a much lower DBTT prior to irradiation than the 9Cr-1MoVNb. Nevertheless, the 9Cr-1MoVNb-2Ni steel shows a larger shift in DBTT, which may be attributable to helium.

Because of the low irradiation temperatures, the mechanism by which helium affects the properties would be expected to be caused by helium in solution. The possibility of a saturation of ΔDBTT due to displacement-damage effects has been discussed [4,12]. The present results indicate a helium effect is superimposed on the displacement-damage effect. Although the helium effect would also be expected to saturate, a much larger ΔDBTT might be expected than the 144 to 166°C shift observed for 12Cr-1MoVW steel irradiated at 390°C in EBR-II [3,5]. More tests of the type discussed in this paper will be required to determine the maximum effect.

5. Summary and Conclusions

Charpy impact specimens of standard 9 Cr-1MoVNb and 12 Cr-1MoVW steels and these steels doped with up to 2% Ni were irradiated at 50°C in HFIR to displacement-damage levels of up to 9 dpa. Because of the nickel present in the steels, irradiation in HFIR produced up to 115 appm He.

Irradiation caused an increase in the DBTT of all steels. The increase was larger for the 9-Cr steels than the 12-Cr steels. The results from these and other studies indicate that the change in ΔDBTT with temperature for the 12Cr-1MoVW has a peak at ~400°C and decreases at higher or lower temperatures. The ΔDBTT for 9Cr-1MoVNb decreases gradually as the irradiation temperature increases from 50 to 400°C, after which it quickly decreases to zero. The data indicate that helium caused an increase in the DBTT of the 9-Cr and 12-Cr steels in addition to that caused by displacement damage.

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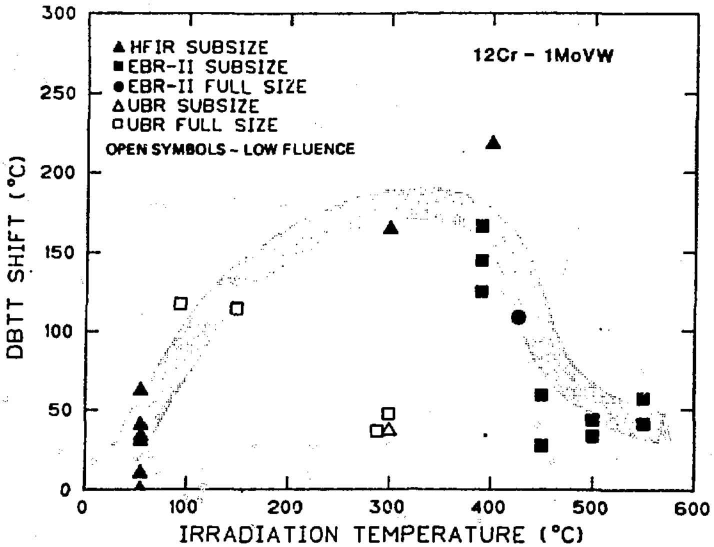
Table 1. Charpy impact properties of unirradiated and irradiated Cr-Mo steels

Steel	Experiment	Irradiation Effects		DBTT[a] (°C)	ΔDBTT (°C)	USE (J)
		dpa	(appm He)			
9Cr-1MoVNb	C[b]	Unirradiated		−40	--	26
	RB1	2.7–3.4	2.5–3	62	102	20
	RB2	4.4–5.8	4–6	70	110	17
9Cr-1MoVNb-2Ni	C	Unirradiated		-90	--	24
	RB1	2.8–3.4	35-43	53	143	11
12Cr-1MoVW	C	Unirradiated		-35	--	26
	RB1	4.3	15	-33	2	20
	RB2	7.2–9.2	30–39	-2	31	25
	T3	6.0–9.0	11-17	3	36	24
12Cr-1MoVW-1Ni	C	Unirradiated		-37	--	23
	RB1	3.9	26	38	75	20
	RB2	8.4	66	33	70	23
12Cr-1MoVW-2Ni	C	Unirradiated		-25	--	17
	RB1	4.0–4.5	50-55	37	62	11
	RB2	6.1–7.4	93-114	66	91	13
	T3	5.7–9.1	36-58	51	76	15

[a]Measured at 9.2 J.

[b]Unirradiated control specimens.

List of Figures



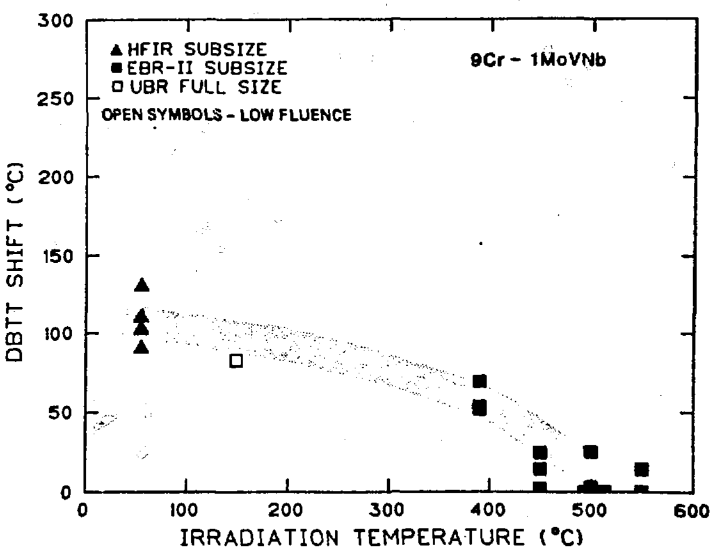


Fig. 1. Increase in DBTT as a function of irradiation temperature for (a) 12Cr-1MoVW and (b) 9Cr-1MoVNb steel.