IRRADIATION EFFECTS ON IMPACT TOUGHNESS OF LOW-CHROMIUM BAINITIC STEELS—

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OBJECTIVE

The objective of this work is to develop an understanding of the effect of irradiation on fracture behavior the reduced-activation ferritic/martensitic steels that are of interest for fusion applications and to use that knowledge to develop steels with improved properties.

SUMMARY

Charpy specimens of five bainitic steels were irradiated at 378-404ºC in the Experimental Breeder Reactor (EBR-II) to 26-33 dpa. The steels were experimental reduced-activation 3Cr-WV steels with additions tantalum, boron, and nickel. The steels were normalized, and specimens of the normalized steel were given two tempering treatments: 1 hr at 700ºC and 1 h at 750ºC. The Charpy tests demonstrated only minor effects of 1% W, 0.05% Ta, and 0.005 %B in the steels in the unirradiated condition. Tungsten and tantalum had a favorable effect on the irradiated properties. Nickel, on the other hand, had a favorable effect on the impact toughness of the steel before and after irradiation.

PROGRESS AND STATUS

Introduction

The 9% Cr reduced-activation ferritic/martensitic steels are being considered favorably for applications as first wall and blanket structural materials for future fusion reactors. Oak Ridge National Laboratory (ORNL) has developed reduced-activation ferritic/martensitic steels, and a martensitic 9Cr-2WVTa (nominally Fe-9Cr-2W-0.25V-0.07Ta-0.1C; all compositions are in weight percent) steel has proved to have excellent high-temperature strength and exceptional irradiation resistance [1-3].

ORNL also developed a bainitic 2¼Cr-2WV (nominally Fe-2.25Cr-2W-0.25V-0.1C) steel that had excellent strength [1], but the toughness and irradiation resistance were inferior to those of 9Cr steels [2,3]. By modifying the 2.25 Cr steel with additional chromium and tungsten additions, the strength and toughness were improved [4,5].

When these types of steel are irradiated below ≈425ºC, displacement damage by neutron irradiation hardens the steel lattice, causing an increase in strength and a decrease in toughness. The effect on impact toughness is measured in a Charpy test as an increase in the ductile-brittle transition temperature (DBTT) and a decrease in the upper-shelf energy (USE).

The possible effect of helium on hardening and embrittlement is important because large amounts of transmutation helium will form in the ferritic/martensitic steel first wall of a fusion reactor. Nickel-doped 9 and 12 Cr steels have been irradiated in a mixed-spectrum reactor such as the High Flux Isotope Reactor (HFIR) to study the effect of helium on fracture [6]. Helium is formed in a mixed-spectrum reactor by a two-step transmutation reaction between ^{58}Ni and the thermal neutrons in the mixed-neutron spectrum. This technique allows for the simultaneous production of displacement damage and helium in the steel matrix, thus simulating what will happen in a first wall. Results from such irradiation experiments at 400ºC have been interpreted to indicate an effect of helium on embrittlement [6]. This conclusion was based on the comparison of the steels with and without nickel and on the differences between the behavior of the nickel-doped steels in a mixed-spectrum reactor, where considerable helium forms, and in a fast reactor, where very little helium forms [6].

More-recent irradiation experiments of nicked-doped 9Cr reduced-activation steels have indicated that the nickel-doped steels hardened more than steels without the nickel addition [7,8]. A 9Cr-2W steel with and without 1% Ni was irradiated in the Japanese Materials Test Reactor (JMTR) to 0.15 dpa at 170ºC, and an increase in the room temperature yield stress of up to 350 MPa was observed for the nickel-containing steel, compared to a 120 MPa increase for the steel without nickel. However, no difference in the strength increases was observed for the steels irradiated at 220ºC [7]. Irradiation of these steels to 2.2 and 3.8 dpa at 270 and 348ºC, respectively, in the Advanced Test Reactor (ATR) indicated that the nickel-containing steel hardened about 20% more than the steel without nickel at 270ºC, but strengths were similar after the irradiation at 348ºC [8]. Likewise, there was a larger shift in DBTT for the nickel-containing steel than the one without nickel when irradiated at 270ºC, but not after irradiation at 348ºC. TEM analysis indicated that nickel refined the size of the defect clusters, which were more numerous in the nickel-containing steel [8].

Helium can also be generated in a steel that contains boron by an (n,α) reaction between a neutron and ^{10}B. This reaction will occur quite rapidly in both a mixed-spectrum reactor and a fast reactor.

In this report, Charpy properties are reported for a series of reduced-activation 3Cr-WV steels that contained different tungsten levels, and steels that contained small additions of tantalum, boron, and nickel after irradiation in the Experimental Breeder Reactor (EBR-II).

Experimental Procedure

Compositions and designations of the steels used in this experiment are given in Table 1. In the original Oak Ridge National Laboratory (ORNL) alloy development program for development of reduced-activation steels [1], 18-kg heats of electroslag-remelted steels ranging from 2.25 Cr to 12 Cr were prepared by Combustion Engineering Inc, Chattanooga, TN. Material from the 18-kg heat of 2.25Cr-2WV steel was used as the master alloy to prepare 450-g vacuum arc-melted button heats of 3Cr steels prepared for this work.

Table 1. Chemical composition of the steels tested

Element[a]	3Cr-2WV	3Cr-3WV	3Cr-3WV-2Ni	3Cr-3WVTa	3Cr-3WVTaB
C	0.087	0.091	0.089	0.089	0.088
Mn	0.31	0.30	0.29	0.30	0.30
Si	0.09	0.09	0.08	0.08	0.09
P	0.014	0.015	0.015	0.016	0.015
S	0.008	0.009	0.008	0.008	0.008
Cr	3.07	3.05	3.02	3.02	3.03
Mo	<0.01	<0.01	<0.01	<0.01	<0.01
W	2.10	3.01	2.86	3.25	2.95
Ni	0.02	0.02	2.01	0.02	0.02
V	0.25	0.24	0.23	0.24	0.24
Nb	<0.01	<0.01	<0.01	<0.01	<0.01
Ta	<0.01	<0.01	<0.01	0.05	0.05
N	0.014	0.015	0.013	0.015	0.013
B	<0.001	<0.001	<0.001	<0.001	0.005

[a] Balance iron

The small heats were cast as 25.4 mm x12.7mm x 152 mm ingots, after which they were rolled to 6.4-mm plate and 0.76-mm sheet. The steels were normalized by austenitizing for 0.5 h at 1050ºC in a helium atmosphere, and they were quickly cooled in flowing helium. Specimens were irradiated in two tempered conditions: 1 h at 700ºC and 1 h at 750ºC.

One-third-size Charpy specimens measuring 3.3 x 3.3 x 25.4 mm with a 0.51-mm-deep 30º V-notch and a 0.05- to 0.08-mm-root radius were machined from normalized-and-tempered 6.4-mm plates. Specimens were machined with the longitudinal axis along the rolling direction and the notch transverse to the rolling direction (L-T orientation). The absorbed energy vs. temperature values were fit with a hyperbolic tangent function to permit the USE and DBTT to be consistently evaluated. The DBTT was determined at an energy level midway between the upper- and lower-shelf energies. Details of the test procedure for the subsize Charpy specimens have been published [9-11].

Six Charpy specimens of each heat and each heat-treated condition were irradiated in the COBRA experiment in EBR-II at temperatures of 378 to 405ºC. Fluence was determined from flux monitors in the irradiation canisters. There was some variation in fluence for different specimens, depending on their position in the canisters, but the individual sets of specimens for a given steel and heat treatment were kept together in the canisters and experienced the same irradiation conditions. Specimens were irradiated to about $5.5x10^{26}$ to $6.9\ x10^{26}$ n/m^2 (E>0.1 MeV), which produced between 26 and 33 dpa. Helium concentrations were calculated to about 3 appm for the steels without nickel and boron, about 6 appm for the nickel-containing steel, and about 50 appm for the steel with boron.

Results

First, the properties of the normalized-and tempered properties will be discussed, followed by a discussion of the properties after irradiation.

Normalized-and-Tempered Properties

Optical microstructures for the normalized-and-tempered (1 h at 700ºC) steels indicate they are 100% bainite (Fig. 1). The major difference in the steels is the prior-austenite grain size (Table 2). For the

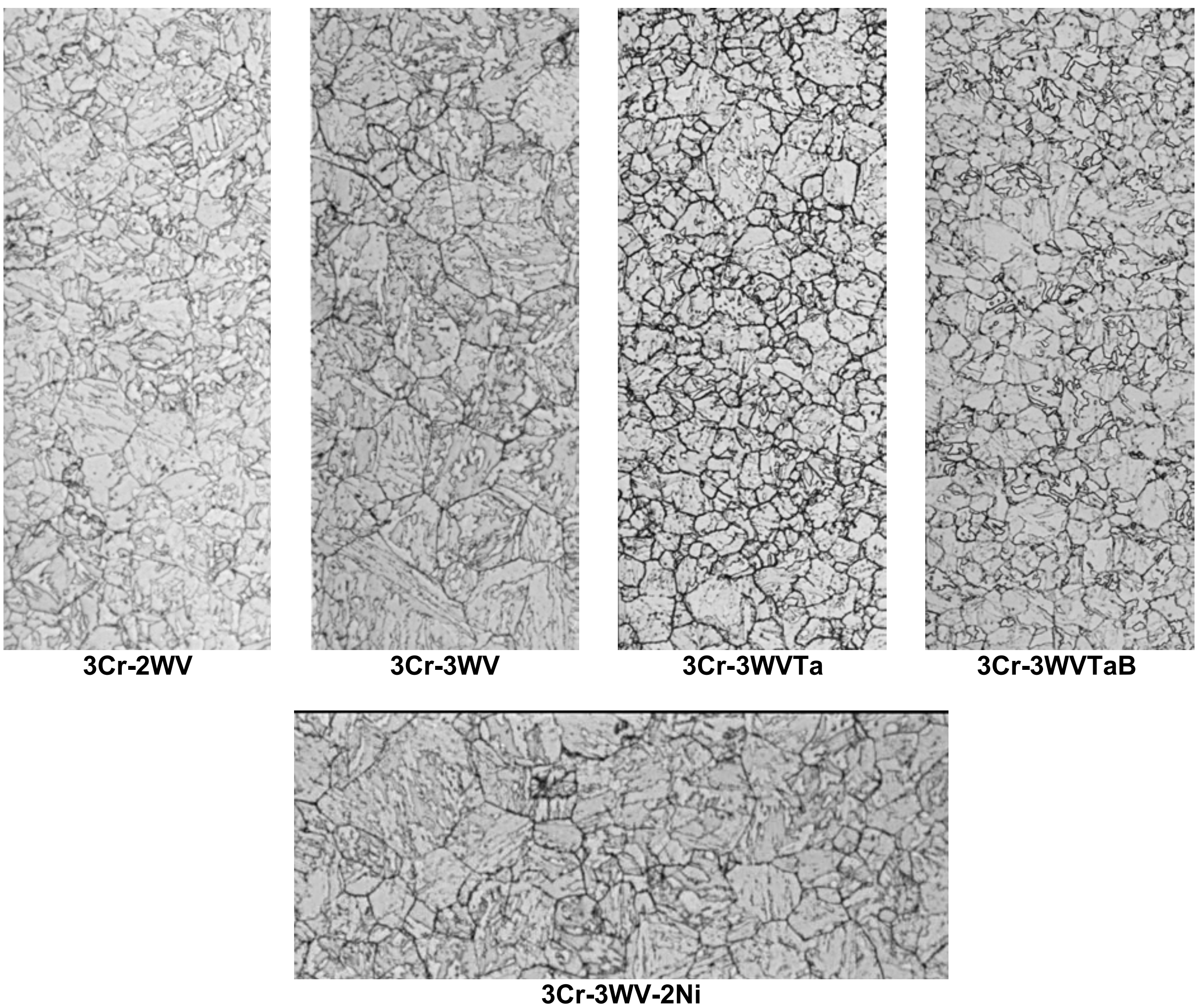

Figure 1. Bainitic microstructure of the 3Cr steels.

Table 2. Prior austenite grain size of steels

Steel	Prior-Austenite Grain Size (μm)
3Cr-2WV	20
3Cr-3WV	38
3Cr-3WVTa	31
3Cr-3WVTaB	16
3Cr-3WV-2Ni	15

steels without tantalum, the grain sizes of the 3Cr-3WV and 3Cr-3WV-2Ni are larger than those of the 3Cr-2WV, and the grain size of the steel containing the nickel is slightly smaller than for the steel without nickel. The addition of the tantalum refines the grain size, and the prior-austenite grain sizes of the 3Cr-3WVTa and 3Cr-3WVTaB, which are similar, are much smaller than for the steels without tantalum.

The Charpy data for the 3Cr steels are summarized in Table 3, and in Figs. 2-4, the ductile-brittle transition temperature (DBTT), shift in DBTT (ΔDBTT) and upper-shelf energy (USE) are compared for the five steels. The base composition for this discussion is the 3Cr-2WV steel, which was the first modification from the 2¼Cr-2WV steel [5].

Table 3. Charpy data for unirradiated and irradiated steels

Steel	Temper	Irrd Temp	Dose	Uirrd DBTT	Irrd DBTT	DBTT Shift	Uirrd USE	Irrd USE
3Cr-2WV	700ºC	378ºC	27 dpa	-92ºC	76ºC	168ºC	10.7 J	4.8 J
	750ºC	379ºC	26 dpa	-150ºC	51ºC	201ºC	11.4 J	7.0 J
3Cr-3WV	700ºC	378ºC	27 dpa	-73ºC	78ºC	151ºC	9.0 J	5.6 J
	750ºC	383ºC	31 dpa	-116ºC	9ºC	125ºC	10.4 J	7.5 J
3Cr-3WVTa	700ºC	392ºC	33 dpa	-66ºC	52ºC	118ºC	9.4 J	7.5 J
	750ºC	386ºC	32 dpa	-103ºC	-15ºC	88ºC	11.8 J	8.4 J
3Cr-3WVTaB	700ºC	404ºC	26 dpa	-74ºC	62ºC	123ºC	7.8 J	5.8 J
	750ºC	386ºC	32 dpa	-111ºC	12ºC	136ºC	10.4 J	6.0 J
3Cr-3WV-2Ni	700ºC	392ºC	33 dpa	-125ºC	-104ºC	21ºC	10.0 J	8.4 J
	750ºC	389ºC	33 dpa	-148ºC	-125ºC	23ºC	11.2 J	10.2 J

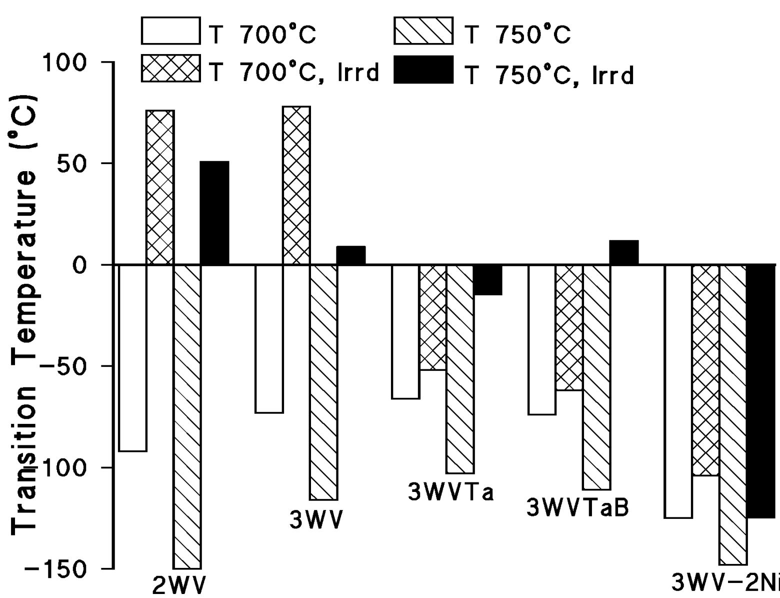

Figure 2. Ductile-brittle transition temperature for 3Cr steel before and after irradiation.

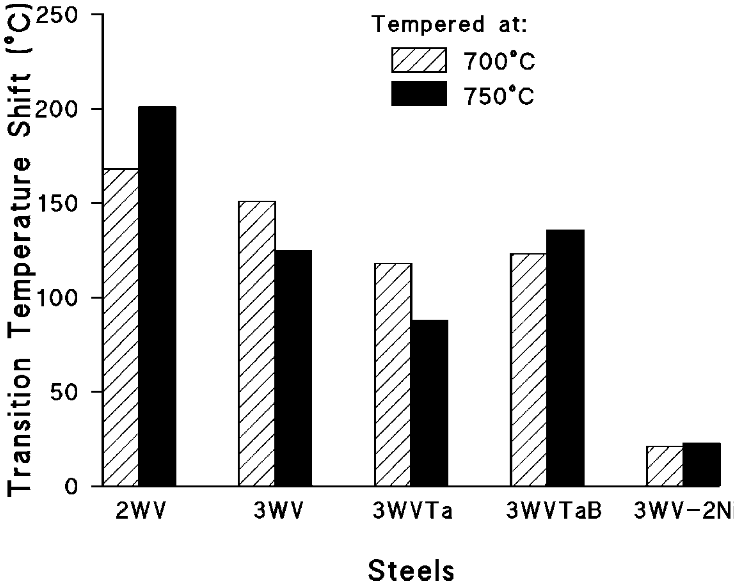

Figure 3. Shift in ductile-brittle transition temperature for irradiated 3Cr steels.

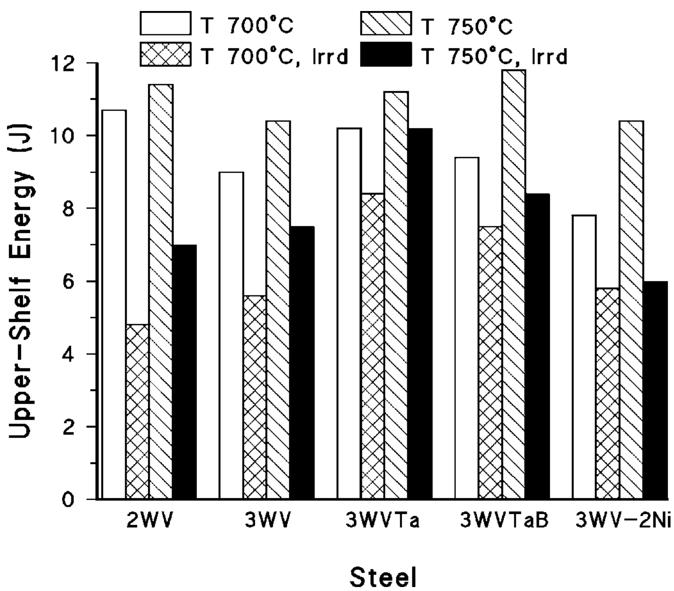

Figure 4. Upper-shelf energy for 3Cr steels before and after irradiation.

Before irradiation, the 3Cr-2WVsteel has excellent impact toughness—low transition temperature and high USE—after both tempering treatments (Figs. 2 and 3). The DBTT and USE for the steels given the 700ºC temper were always higher and lower, respectively, than those for the steels tempered at 750ºC. The addition of another 1% W to the 3Cr-2WV to produce the 3Cr-3WV caused a slight decrease in the DBTT and USE. The addition of 0.05 Ta to the 3Cr-3WV (3Cr-3WVTa) had essentially no effect on the properties relative to 3Cr-3WV. Likewise, the addition of 0.005B to the 3Cr-3WVTa (3Cr-3WVTaB) did not change the DBTT, although the USE of the 3Cr-3WVTaB appeared to be slightly less than that of the 3Cr-3WVTa. The addition of nickel to the steels had the largest effect on the impact properties relative to the properties of the 3Cr-3WV steel, especially after the 700ºC temper. The 3Cr-3WV-2Ni had properties similar to those of the 3Cr-2WV.

Irradiated Properties

After irradiation, the DBTT of the steels tempered at 700ºC were higher than for those steels given the 750ºC temper (Table 3 and Fig. 2). This did not always translate into a larger ΔDBTT (Fig. 3) for the steel given the 700ºC temper. For the 3Cr-2WV, the DBTT after the 700ºC temper showed the largest difference for a 700ºC-tempered specimen. Although there was a slightly lower ΔDBTT for the 3Cr-3WVTaB and 3Cr-3WV-2Ni steels tempered at 700ºC than 750ºC, the differences were so small that the steels can be considered as having the same ΔDBTT for both tempering conditions.

With the exception of the 3Cr-3WV-2Ni, the results show fairly large DBTT values after irradiation for the 3Cr-2WV steel after both tempering conditions and the other steels after the 700ºC temper. After tempering at 750ºC, the DBTT values after irradiation indicated that adding 1% W to the 3Cr-2WV to get 3Cr-3WV and adding 0.05% Ta to 3Cr-3WV to get 3Cr-3WVTa were beneficial (DBTT values of -15 and 9ºC, respectively). The 3Cr-3WVTaB steel also had a fairly low DBTT (12ºC) after the 750ºC temper, although it was not as low as that of the steel without boron. It should be noted that the 3Cr-3WVTaB contained over 50 appm He generated from the ^{10}B, but there was no indication that this helium affected the Charpy properties.

By far, the steel displaying the best irradiation resistance was the 3Cr-3WV-2Ni. This steel had the lowest DBTT values before irradiation, and after irradiation, shifts of only 21 and 23ºC were observed for the specimens tempered at 700 and 750ºC, respectively. As a result of these small shifts, the measured DBTT values remained below -100ºC after irradiation for both tempering conditions.

Discussion

In the unirradiated condition, several observations can be made. First, the addition of 1% W to the 3Cr-2WV did not improve the impact properties. The properties of the 3Cr-3WV were slightly below those of the 3Cr-2WV, which may reflect the relative difference in prior-austenite grain size of the two steels. Adding 0.05% Ta to the 3Cr-3WV had little effect on the impact properties, even though there was a significant reduction in the prior-austenite grain size for the tantalum-containing steel, nor did the addition of 0.005% B to the 3Cr-3WVTa steel have any noticeable effect on the tantalum-containing steel.

Nickel has long been known to favorably affect the impact properties of steels, and the results for the unirradiated properties for the 3Cr steels show that the 2% Ni addition to the 3Cr-3WV composition resulted in a significant improvement of the impact properties over the steel without the nickel addition. For the steels tempered at 700ºC, the nickel-containing steel had a transition temperature that was better than that for all the steels but the 3Cr-2WV after the other steels were tempered either at 700 or 750ºC. After tempering the 3Cr-3WV-2Ni at 750ºC, the transition temperature was similar to that of the 3Cr-2WV steel and superior to the other steels. Likewise, the USE of the steel with nickel was always about as high or higher than for the other steels for the respective tempering conditions.

After irradiation, the effects of composition are somewhat different than in the unirradiated condition. For all five steels, the DBTT values for the steels irradiated at 750ºC were less than those for the respective steels tempered at 700ºC, although the shifts in DBTT were not always more for the steel with the 750ºC temper. If the steels tempered at 750ºC are compared, it appears that increasing the tungsten from 2 to 3% improves the irradiation resistance. Likewise, adding the 0.05% Ta to the 3Cr-3WV had a favorable effect. The boron addition did not appear to favorably affect the 3Cr-3WVTa composition.

By far the most impressive irradiation resistance was exhibited by the 3Cr-3WV-2Ni steel. In this case, the ΔDBTT for the steel tempered at 700 and 750ºC were similar and very low. The steel tempered at 750ºC had the lowest DBTT after irradiation because it had a considerably lower value prior to irradiation. However, the DBTT for the steel after both tempering conditions was <-100ºC. The other interesting observation was that the USE of the 3Cr-3WV-2Ni tempered at 700ºC was higher than that of any of the steels except the 3Cr-3WVTa after they were tempered at either 700 or 750ºC. It had the same USE as the 3Cr-3WVTa steel that was tempered at 750ºC. The DBTT and USE values for the 3Cr-3WV-2Ni after either temper were better than most steels for similar irradiation conditions, including the 9Cr-2WVTa [2] , which in the past has been shown to have excellent properties for most irradiation conditions.

Nickel was originally added to the steel in this experiment because the 3Cr-3WV and 3Cr-3WV-2Ni steels were to be irradiated in the HFIR, where an (n,α) reaction between ^{58}Ni and the thermal neutrons in the mixed-neutron spectrum of HFIR would produce helium, thus allowing for a determination of the effect of helium on impact properties. Irradiation of the steels in the fast-neutron spectrum of EBR-II, where very little helium forms, would thus provide a comparison of the properties of 3Cr-3WV-2Ni with and without the high helium concentration. Unfortunately, the HFIR experiments were cancelled. Nevertheless, the observations on the nickel-containing steel provide some insight into the effect of nickel on irradiation resistance.

As discussed in the Introduction, there have been some apparent contradictory observations on the effect of nickel [7,8] on irradiated steels. At temperatures below ≈300ºC, small irradiation doses were shown to cause excess hardening and an increase in DBTT of a 9Cr-2W steel that contained 1-2% Ni compared to the steel without the nickel addition [7], whereas at higher temperatures and higher doses, no indication of such hardening due to nickel in 9 and 12% Cr-Mo steels was observed [8,9]. However, in no instance in these latter studies was there any indication of an improvement in properties of the type displayed in the present experiment for the 3Cr-3WV-2Ni.

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