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Embrittlement behaviour of different international low activation alloys after neutron irradiation

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Abstract

The embrittlement behaviour of ferritic/martensitic steels, after irradiation in the Petten High Flux Reactor (HFR), was investigated by Charpy-V tests with subsize specimens. The main objective, apart from studying the effects of particularly low doses, was to compare the irradiation response of low activation alloys from various countries with different levels of Cr, minor alloying elements and impurities. Specimens were irradiated at temperatures ranging between 250°C and 450°C to dose levels between 0.2 and 2.4 dpa. The evaluation clearly showed a tendency to a much reduced embrittlement problem for the advanced reduced-activation alloys; it could not, however, be attributed to lower Cr-contents. Instead, the role of helium, already suspected in our earlier work, could be correlated with the characteristic burn-up of different boron levels in the steels. This correlation translates for a fusion reactor environment into a non-saturating deterioration of alloys even without any boron, due to helium-yielding high energy neutron reactions with the base elements. Though the effect seems to be less detrimental for the more homogeneous distribution of the helium in the matrix.

1. Introduction

The MANITU irradiation and fracture-toughness testing programme on promising low-activation ferritic/martensitic steels as reported at the preceding ICFRM-7 [1] has been completed. And impact test data for doses of 0.2/0.8/2.4 dpa and at temperatures of 250/300/350/400/450°C are now available. Particular emphasis was to be laid on the further evolution of the impact properties with increasing dose at the different irradiation temperatures, and on the behaviour of the different alloys in comparison to each other, especially at the critical lower irradiation temperatures. The previous investigation [1] had shown for irradiation parameters of 250°C/0.8 dpa that the US-heat, referenced as 'ORNL', had exhibited the lowest ductile-to-brittle transition temperatures (DBTT). Although its chromium content, distinctly above that of the Japanese F82H and not far from various FZK-alloys (see Table 1), would have suggested a different behaviour [2]. In addition to examine the dose dependency, it is also interesting to look for other factors which might have had an influence on fracture toughness deterioration.

2. Experimental

The Charpy specimens were produced parallel to the rolling direction (L-T) of the material plates and according to the European standard for subsize specimens. The same specimen type was used in our previous investigations to enable a direct comparison of the results. For the same reason, all tests have been carried out with the same instrumented facility which is installed in the Hot Cells. The test and evaluation procedure was also identical as those in our previous investigations [1,2].

For each experiment the force-vs.-deflection curve was recorded and the impact energy was determined by integration. As usual this quantity was plotted against the test temperature and from this the characteristic values upper shelf energy (USE) and ductile-to-brittle transition temperature (DBTT, i.e. temperature at USE/2) were derived.

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The irradiations of the MANITU programme were all carried out in the HFR, Petten. The target levels of 0.2, 0.8, and 2.4 dpa were reached at least within ±20% depending on the core position of the specimens. The irradiation temperatures of 250°C, 300°C, 350°C, 400°C, and 450°C were maintained within ±5% by a proper balance between n,γ-heating and compartment cooling with different He–Ne mixtures.

A minimum of five specimens for each material, dose level, and irradiation temperature ensured a sufficient number of measurement points in order to connect and group them to curves with the irradiation temperature as abscissa and the materials and dose levels as parameter.

3. Results and interpretation

The results including those for 0.8 dpa [1] are shown in Fig. 1. It can be seen that for a given irradiation temperature DBTT is shifted from the 'unirradiated' value, which is also indicated, to higher temperatures with increasing dose. A few exceptions may be attributed to statistical scatter rather than to systematic effects. A comparison of the different materials repeats what has already been noticed previously in [1], namely that the more advanced Low Activation show improvements both of the absolute values of DBTT and their impairment by the irradiation (ΔDBTT) compared to the MANET steels. As demonstrated in Fig. 2, USE shows a more or less complementary behaviour, i.e. the higher the dose the lower is USE, especially when the irradiation temperature was low. Dynamic yield, derived from measurements at 100°C, has also been evaluated, but the results are not given in this paper because of space limitations.

To allow for a better quantitative comparison of the irradiation behaviour of the different materials, the dose dependency of DBTT is presented in Fig. 3 for one of the more critical, i.e. lower, irradiation temperatures. DBTT-shift (ΔDBTT), rather than absolute DBTT, has been chosen in order to eliminate the differences introduced by the different Cr contents. As demonstrated by Fig. 3, ΔDBTT behaves very similarly for all steels, but the susceptibilities to dose are quite different.

Starting from some suspicion about the influence of helium on impact toughness [3], and from complementary investigations with He-implanted Chary-V specimens [4], the boron contents, as given in Table 1, seems to be a controlling factor of DBTT-shift by irradiation. The result was quite evident: the higher the boron concentration, the steeper the slopes were. But all curves showed the same tendency to saturate at rather low doses. Indeed boron, with its 20% of the isotope ^{10}B is one of the strongest thermal neutron absorbers which shows a burn-up effect already at very moderate neutron fluences and is thereby transformed to stable helium and ^{7}Li.

The burn-up constant τ, defined here as the dose D in dpa, necessary to decrease the initial ^{10}B-content down to a fraction of 1/e (i.e. ca. 37%), was derived from the neutronic calculations of HFR-Petten to be 0.34 dpa for the spectral conditions of this irradiation. A burn-up of 99.3%, corresponding to five times the burn-up constant, is reached after ca. 1.6 dpa which is in the range of the dose when saturation for irradiation hardening was found for MANET-I [5]. For a comparison with the measured embrittlement behaviour the ^{10}B-to-He transformation curve has also been drawn into Fig. 3. The dose dependency of the curve is qualitatively similar to that of ΔDBTT for the different steels. The different curves in Fig. 3 have been analysed by applying the growth function $N(\mathrm{He}) = N_0(^{10}\mathrm{B})*(1-\exp(-D/\tau))$. Under the assumption that ΔDBTT is proportional to N(He), and neglecting effects such as He bubble growth and size, this equation can be brought into the linearised form: $\ln(1-\Delta\mathrm{DBTT}/\Delta\mathrm{DBTT}_{max}) = -D/\tau$, from which $\Delta\mathrm{DBTT}_{max}$ can be estimated for the different materials by a fit to the common slope being $-1/\tau$. The values are given in Table 2 together with the corresponding total He-amounts resulting from complete ^{10}B transmutation. Indeed the ratio of ΔDBTT over He in atomic ppm (appm) is more or less the same for all materials measured, and amounts to approximately 2–3 K per appm He.

This is a significant effect that seems to override all other factors, but leaves a few questions open which shall now be discussed. One is a DBTT-shift that had been previously measured for MANET-I to be about 260 K at doses of 5 dpa and above [3]. A reasonable explanation could be that displacement damage induced hardening as such also has an effect which shows up at increased doses and probably does not saturate at medium dose levels (10–15 dpa) as supposed earlier. Another point is that a much lower DBTT-shift amounting to only about 45 K was found for He implanted specimens to about 300 appm by a high-energy cyclotron in a parallel investigation [4]. Certainly, the distribution of He atoms in the material plays a decisive role and it is general assumption that the distribution of boron in steel is quite inhomogeneous. The third question is raised by the recovery effect of property deterioration at increased irradiation temperatures, i.e. more than 350°C. Explanations for this behaviour are proposed in [6], but further investigations will be needed.

A critical aspect of the influence of helium on DBTT arises, if one translates our results to fusion reactor conditions where about 10 appm helium have been calculated to be generated per dpa by the (n, α)-reaction of fast neutrons with iron [7]. This is a non-saturating source, and tremendous DBTT-shifts could be extrapolated if one takes a target value of at least 100 dpa for structural materials into account. Fortunately the effects of helium homogeneity and recovery phenomena at

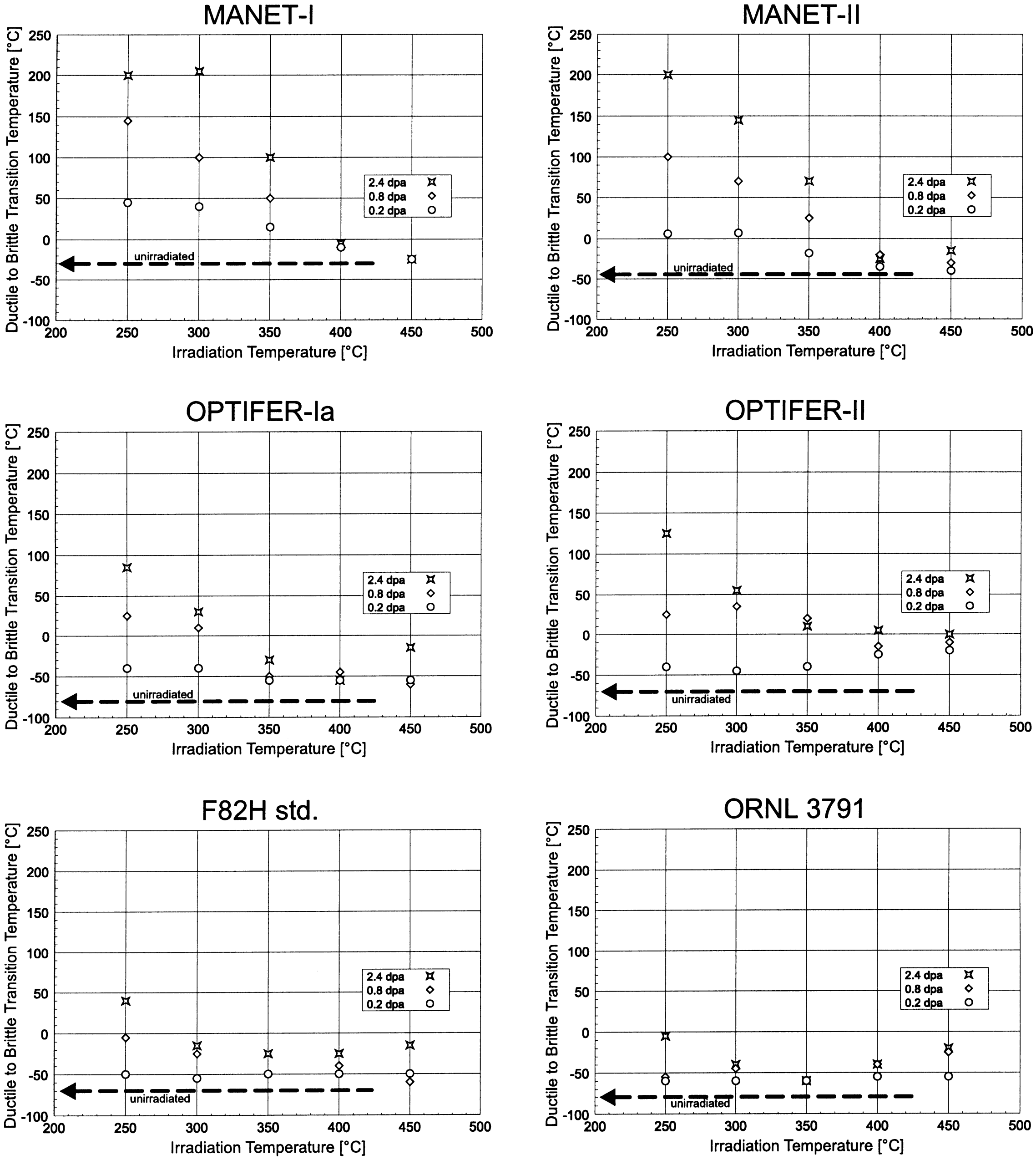

Fig. 1. Ductile-to-brittle transition temperature vs. irradiation temperature (parameter: irradiation dose).

higher temperatures, as discussed above, create a more relieved situation with respect to impact properties deterioration.

4. Conclusions

The dose dependency of impact properties for different advanced ferritic/martensitic alloys at low irradiation temperatures has been revealed to be in the low-dose range mainly an effect of helium generated by different levels of boron, the ^{10}B isotope of which is burnt up with nearly the same characteristic time constant as DBTT increases towards saturation. At irradiation temperatures above 350°C the DBTT-shift is significantly lower. In addition, a more homogeneous distribution of He in the matrix may show a smaller effect on DBTT. These findings are of utmost importance for

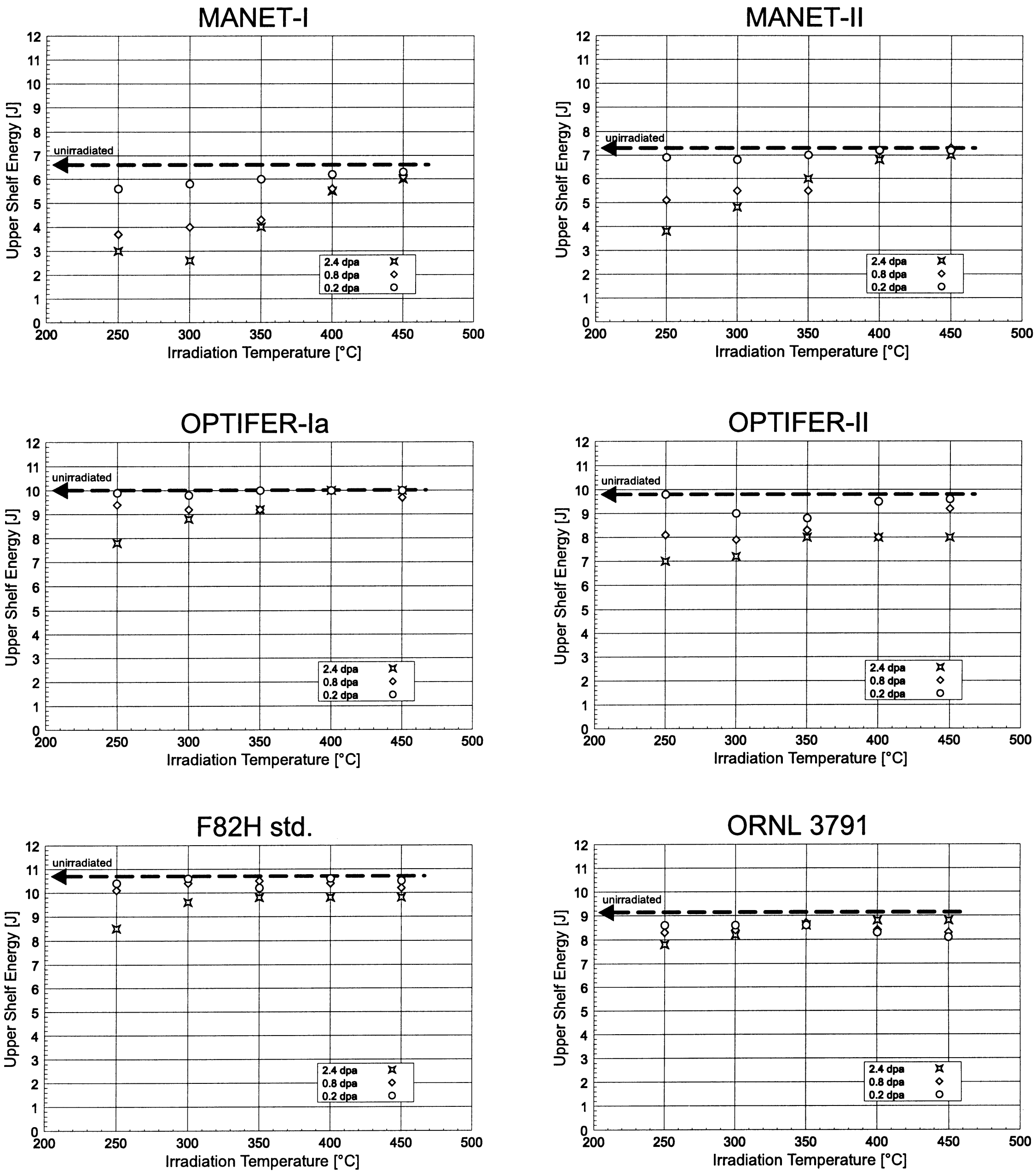

Fig. 2. Upper shelf energy vs. irradiation temperature (parameter: irradiation dose).

fusion since in a fusion reactor environment there will be an inexhaustible source for helium from fast neutron reactions with iron. Validation tests to proof and further quantify our results are indispensable. As long as no powerful fusion or accelerator-based neutron source is available, also a continuation of high-energy helium implantation tests by means of an available cyclotron should seriously be reconsidered.

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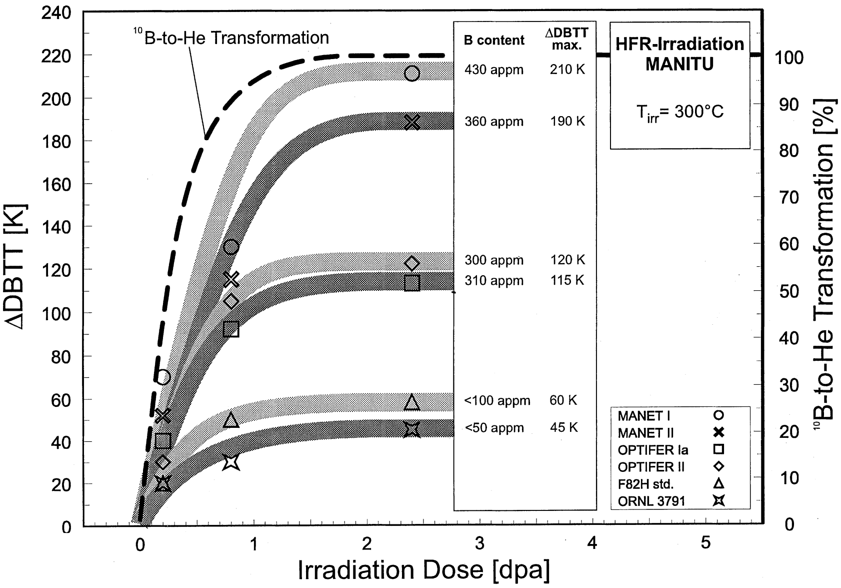

Fig. 3. Irradiation induced shifts of ductile-to-brittle transition temperature and ^{10}B-to-He transformation vs. irradiation dose (parameter: materials).

Table 1
Chemical composition of the different alloys in wt%

	10–11% Cr-NiMoVNb steels		Low activation alloys			
	MANET-I	MANET-II	OPTIFER-Ia	OPTIFER-II	F82H std.	ORNL 3791
Cr	10.8	9.94	9.3	9.43	7.73	8.9
W			0.965	0.005	2.06	2.01
Ge				1.1		
Mn	0.76	0.79	0.5	0.5	0.083	0.44
N	0.02	0.023	0.015	0.016	0.0027	0.0215
Ta			0.066	0.02	0.018	0.06
C	0.14	0.1	0.1	0.125	0.092	0.11
P	0.005	<0.006	0.0047	0.004	0.003	0.015
S	0.004	<0.007	0.005	0.002	0.003	0.008
V	0.2	0.22	0.26	0.28	0.189	0.23
B	0.0085	0.007	0.0061	0.0059	<0.002	<0.001
Si	0.37	0.14	0.06	0.038	0.09	0.21
Ni	0.92	0.66	0.005	0.005	0.032	<0.01
Mo	0.77	0.59	0.005	0.005	0.0053	0.01
Al	0.054	<0.02	0.008	0.008	0.01	0.017
Co	0.01	<0.02			0.0024	0.012
Cu	0.015	<0.01	0.035	0.007	0.0059	0.03
Nb	0.16	0.14	0.009	0.009	0.0057	
Zr	0.059	0.034				<0.001
Ce			<0.001	<0.001		
Ti			0.007	0.007	0.0104	<0.01
Fe	Balance	Balance	Balance	Balance	Balance	Balance

Table 2
Extrapolated DBTT-shift and helium content after complete ^{10}B burn-up

	MANET-I	MANET-II	OPTIFER-Ia	OPTIFER-II	F82H std.	ORNL
$\Delta DBTT_{max}$	210 K	190 K	115 K	120 K	60 K	45 K
max He	85 appm	70 appm	60 appm	60 appm	<20 appm	<10 appm

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