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Embrittlement behaviour of low-activation alloys with reduced boron content after neutron irradiation

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Abstract

Ferritic/martensitic steels for fusion applications have been irradiated up to 2.4 dpa in the Petten high flux reactor (HFR); their embrittlement behaviour was investigated by instrumented Charpy-V tests with subsize specimens. The aim of this mid-dose range programme was a comparison of low-activation alloys subjected to different heat treatments and with reduced B contents (down to 2 wt ppm). In the present report, the results of different OPTIFER alloys (Ia, II, IV, V, VI), as obtained in Phases IA and IB of the HFR-irradiation programme (2.4 dpa, at 250–450 °C), are analysed and assessed in comparison to the results of the former MANITU programme. The evaluation clearly shows the eliminated embrittlement problem for the advanced European reduced-activation alloys in comparison to international reference steels. This improvement can be clearly correlated to the reduction of the boron content. Furthermore, the influence of different heat treatments on the impact properties is presented.

1. Introduction

Within the former MANITU irradiation programme [1,2], promising low-activation ferritic/martensitic steels were investigated by subsize Charpy tests. Irradiation temperatures of 250/300/350/400/450 °C at doses up to 2.4 dpa were covered. Particular emphasis was put on the behaviour of the different alloys in comparison to each other, especially at the critical low irradiation temperatures. These data now have been complemented by another two 2.4 dpa irradiation programmes covering the same temperature range: high flux reactor (HFR) Phases IA and IB.

The aim of the Phase IA programme was to define the influence of the alloys' heat treatments before irradiation. Promising low-activation alloys (LAA) from MANITU, until then only irradiated and examined in the as-delivered state (i.e. austenitization temperatures above 1040 °C, cf. Table 1), were subjected to a heat treatment at lower austenitization temperature, leading to smaller grain sizes and better impact properties before irradiation. In addition, the Japanese F82H mod. steel was included. In the Phase IB programme, OPTIFER alloys with different heat treatments and strongly reduced boron contents were irradiated to study the influence of both parameters on the embrittlement behaviour. The OPTIFER-V alloy corresponds to OPTIFER-Ia and is very similar to EUROFER 97, which became available after the irradiations (see Table 2).

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, see Fig. 1. The same type had already been used in previous investigations to enable a direct comparison of the results. For the same reason, all tests were carried out with the same instrumented facility which is installed

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Table 1
Heat treatment and selected properties of unirradiated materials

	Heat treatment	Grain size (μm)	USE (J)	DBTT (°C)	Dynamic yield stress at 26 °C (MPa)
OPTIFER-Ia*	1075 °C/0.5 h + 780 °C/2 h	50	10.1	−80	543
OPTIFER-Ia	900 °C/0.5 h + 780 °C/2 h	10	10.6	−85	500
OPTIFER-V*	1040 °C/0.5 h + 750 °C/2 h	50	9.5	−80	605
OPTIFER-V	950 °C/0.5 h + 780 °C/2 h	12	9.7	−100	592
OPTIFER-II*	950 °C/0.5 h + 780 °C/2 h	55	9.2	−70	495
OPTIFER-II	900 °C/0.5 h + 780° C/2h	–	9.7[a]	−75[a]	470
OPTIFER-VI*	1040 °C/0.5 h + 750 °C/2 h	35	9.7	−56	578
OPTIFER-VI	950 °C/0.5 h + 780 °C/2 h	10	9.1	−70	540
OPTIFER-IV	900 °C/0.5 h + 750 °C/2 h	10	9.3[a]	−80[a]	519
F82H	1040 °C/0.5 h + 750 °C/2 h	35	10.7	−70	550
F82H mod.	1040 °C/0.6 h + 750 °C/1 h	55	9.8	−40	555
ORNL 3971	1050 °C/0.5 h + 750 °C/1 h	25	9.2	−80	599

[a] Values estimated from 10 × 10 mm Charpy-V results.

Table 2
Chemical composition of the different alloys in wt%

	OPTIFER low-activation alloys					Other low-activation alloys[a]			For compar. EUROFER 97
	OPTIFER-Ia	OPTIFER-V	OPTIFER-II	OPTIFER-VI	OPTIFER-IV	F82H	F82H mod.	ORNL 3971	
Cr	9.3	9.48	9.5	9.35	8.5	7.73	7.7	8.9	8.91
W	0.965	0.985	0.006	0.005	1.16	2.06	1.94	2.01	1.08
Mn	0.5	0.39	0.49	0.61	0.6	0.083	0.16	0.44	0.48
N	0.015	0.0225	0.0159	0.025	0.06	0.0027	0.006	0.0215	0.02
Ta	0.066	0.061	0.018	0.083	0.15	0.018	0.02	0.06	0.14
C	0.1	0.115	0.125	0.125	0.11	0.092	0.09	0.11	0.12
V	0.26	0.245	0.28	0.275	0.23	0.189	0.16	0.23	0.2
B	0.006	0.0002	0.0059	0.0002	0.003	0.003	0.0002	<0.001	0.001
Ti	0.007	0.007	0.006	0.007	<0.02	0.0104	0.01	<0.01	0.006
Fe	Balance	Balance	Balance	Balance	Balance	Balance	Balance	Balance	Balance

[a] Also included in HFR-irradiations MANITU, Phases IA and IB.

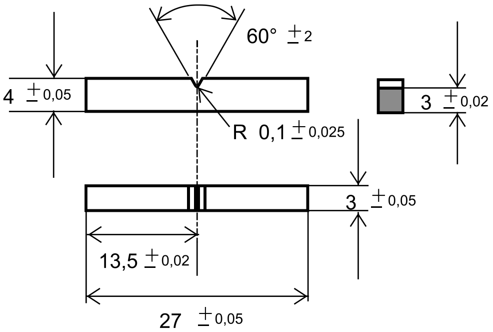

Fig. 1. Subsize Charpy specimen (all dimensions in mm).

in the hot cells. The test and evaluation procedures were also identical with those employed in previous investigations [1–4]: 15 J pendulum impact hammer; striker radius 2 mm, distance between supports 22 mm, impact velocity 3.85 m/s; strain gauges applied in striker, PC-controlled test execution and recording, sampling rate 1 MHz; semiautomatic specimen cooling, heating and loading system (−180 to 600 °C).

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

Dynamic yield stress was derived as stated in [3] from the original force vs. deflection curve at the onset of plastic deformation. In contrast to previous investiga-

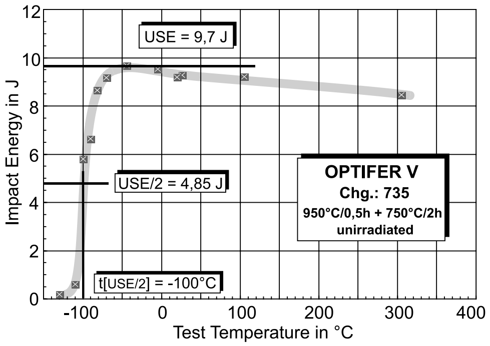

Fig. 2. Determination of USE and DBTT (example: OPTIFER-V).

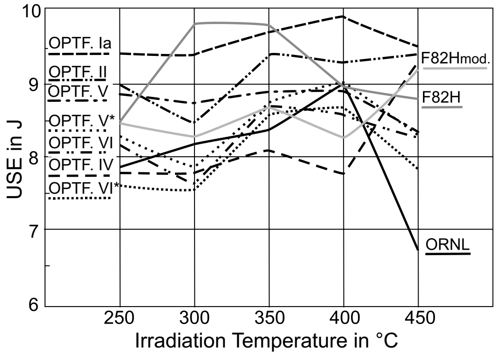

Fig. 3. Upper shelf energy vs. irradiation temperature (parameter: materials).

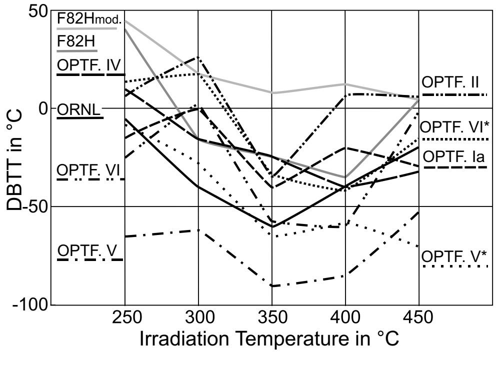

Fig. 4. Ductile-to-brittle transition temperature vs. irradiation temperature (parameter: materials).

tions, dynamic yield stress could not be determined at 100 °C. The materials' DBTT and, thus, the test temperatures were significantly lower. As a consequence, the dynamic yield stress vs. temperature curve ended up far below 100 °C. This led to a comparison of recent and former results at a temperature of 26 °C.

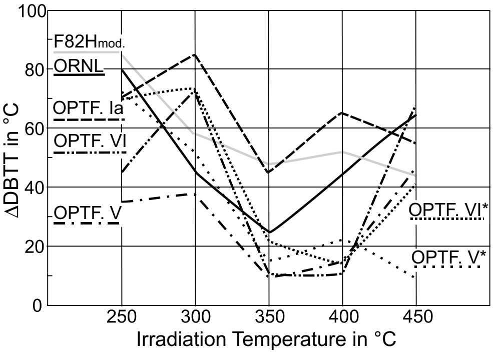

Fig. 5. Irradiation-induced shifts of ductile-to-brittle transition temperature vs. irradiation temperature (parameter: materials).

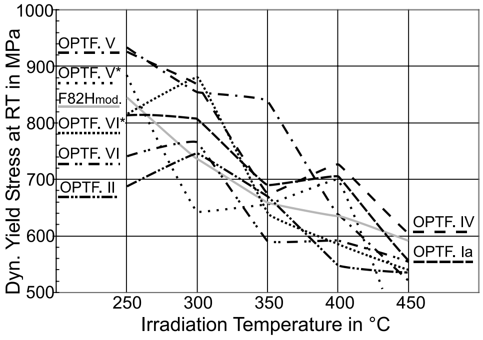

Fig. 6. Dynamic yield stress measured at 26 °C vs. irradiation temperature (parameter: materials).

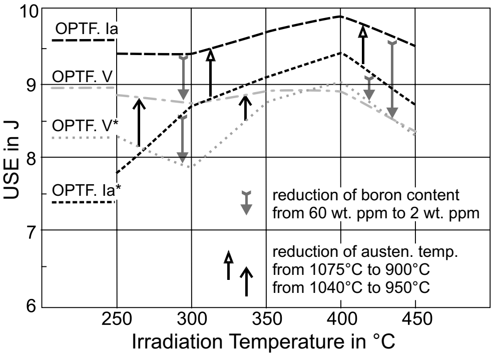

Fig. 7. Influence of heat treatment and boron content for OPTIFER-Ia/OPTIFER-V.

All irradiations were carried out in the HFR, Petten. The target value of 2.4 dpa was reached within a range of +4 and +22% at least, depending on irradiation temperature and, thus, on the vertical core position of the specimens. The irradiation temperatures of 250, 300, 350, 400, and 450 °C were maintained within ±6% by a proper balance between n, γ-heating and compartment cooling with different He–Ne mixtures.

Between 5 and 8 specimens of each material and each irradiation temperature ensured a sufficient number of measurement points for drawing the Charpy energy vs. test temperature curve. The USE and the DBTT were derived from those in the diagrams shown in Fig. 2. These results were combined and grouped in curves with the irradiation temperature as abscissa and the materials as parameter (Figs. 3–7).

3. Results and discussion

The results are presented in Figs. 3–7, including those for some selected materials from [2], where it appears meaningful for comparison. Fig. 3 shows the USE as a function of irradiation temperature. As already assessed in [3], the LAA generally maintain a high impact toughness in the whole temperature range: With 9.4–9.9 J, the OPTIFER-Ia steel has the highest impact toughness of all investigated alloys at all temperatures, and this seems to be less affected by the low irradiation temperatures than other LAAs, where a slight tendency to higher impact toughness for higher irradiation temperatures is recognisable. In the middle temperature range, F82H mod. shows a 0.5 J lower impact energy than the previously investigated F82H [3], but for 250 and 450 °C, the impact energy is higher by 0.6 J. However, all LAAs are found to reach energy values above 7.6 J. According to former results, the ORNL 3791 steel was found in top position, but compared with the new results, this steel ranks in the middle of the LAAs. Especially OPTIFER-Ia, -II and -V are significantly better than the average.

The same picture is obtained from a presentation of incremental values, ΔUSE, normalised to the USE before irradiation. The OPTIFER-Ia, -V, and F82H mod. steels show the lowest decrease of USE (not more than 12% even for the low temperature range) in comparison to the other LAA, which cover a range between 0 and −20% depending on the irradiation temperatures.

Having a look at the DBTT as a function of the irradiation temperature (Fig. 4), it is clearly found that the F82H mod. steel behaves poorly compared to the other LAAs – even the F82H investigated in MANITU has a DBTT that is lower by about 50 °C in the middle temperature range. The OPTIFER-Ia, -IV, and -VI steels are significantly better. They are in the range of the best LAA (ORNL 3791) formerly investigated. The OPTIFER-V alloy exhibits the lowest DBTT values of all investigated materials for both heat treatments.

The shift in DBTT, as indicated in Fig. 5, essentially reveals the same behaviour. It is between 35 and 80 °C for the low irradiation temperatures and between 10 and 70 °C for temperatures of 350 °C and above. OPTIFER-V again shows less irradiation-induced embrittlement than the other alloys. Around 350 °C, there seems to be a minimum shift of DBTT for most materials.

Dynamic yield stress determined at 26 °C, as shown in Fig. 6, increases with decreasing irradiation temperature. In former investigations [3], the increase of DBTT and yield stress could be correlated in a simple manner, as the proportion of both parameters' increase was similar for all tested materials. Now, irradiation hardening still is in the same order of magnitude, as it seems not to be affected by the reduction of the boron content. But the DBTT increase is significantly reduced and exhibits a minimum at 350 °C. The proportion of both increases is not constant any longer for the different irradiation temperatures, thus, irradiation embrittlement and hardening do not correlate in a simple manner for the examined alloys. However, the yield stress values for OPTIFER-Ia/V and OPTIFER-II/VI alloys are in the same range for comparable heat treatments.

OPTIFER-Ia and -V mainly differ by their boron content (0.006 vs. 0.0002 wt%). As they are both irradiated in two different heat treatments of low and high austenitization temperature, the effects of these two parameters can be studied in Fig. 7. A similar picture could also be drawn for OPTIFER-II and -VI steels. A lower austenitization temperature leads to smaller grain sizes and to higher impact energies. This is true for both the boron contents before and after irradiation. The result found in [5,6] could also be confirmed: influence of the heat treatment is not affected by irradiation – both treatments show approximately the same shift in USE and DBTT for the low temperature range (cf. Figs. 5 and 8). The influence of the lowered boron content can be seen clearly when looking at the DBTT: it is about 50 °C lower for both cases and the shift in DBTT after irradiation is reduced to 30%. The USE before and after irradiation of OPTIFER-V and -VI is not as high as for OPTIFER-Ia and -II, a tendency which could be observed formerly for other steels with a low boron content.

To allow for a better quantitative comparison of the different materials, the dose dependence of DBTT was plotted for $T_{\mathrm{irr}} = 300$ °C in [2]. It turned out that the DBTT shift (ΔDBTT) behaved very similarly for all steels, but with quite different slopes. Now, this diagram has been completed by the recently obtained results at 2.4 dpa, see Fig. 8.

In [5], an influence of helium on the impact toughness was suspected. It was described in detail in [1,2]: the boron content seems to be a controlling factor for the shift of DBTT by irradiation. The ^{10}B isotope (20% of the natural composition) is a strong thermal neutron absorber and it transforms to helium and lithium already at moderate neutron fluxes. Helium is found as bubbles in the crack surfaces of irradiated materials. In the preceding report, a model for calculating the irra-

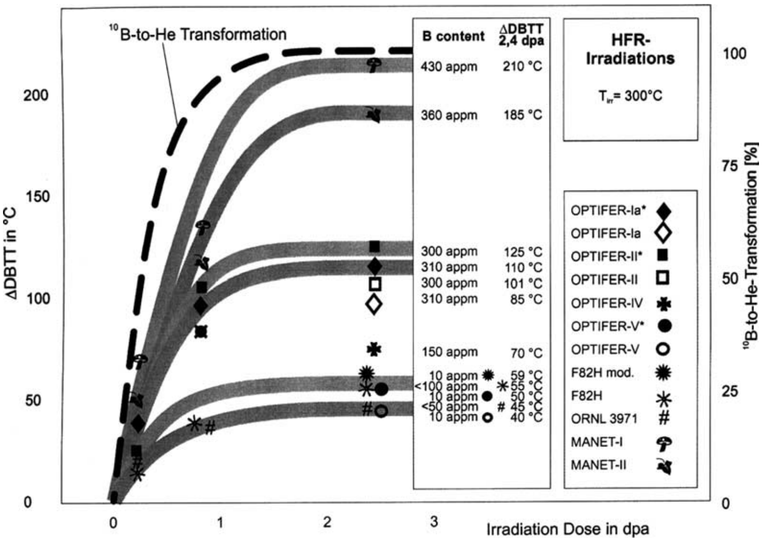

Fig. 8. Irradiation-induced shifts of ductile-to-brittle transition temperature and ^{10}B-to-He transformation vs. irradiation dose (parameter: materials).

diation-induced ^{10}B burn-up was described. It is based on an exponential growth function, using the neutron calculations of HFR Petten to determine the constants. For details, see [1].

The boron to helium transformation curve is plotted vs. the irradiation dose in Fig. 8 and can be compared to the embrittlement lines of the irradiated materials. The boron contents, as given in Table 1, were related to the slopes. The result is quite evident: the higher the boron concentration, the steeper were the slopes. This effect overrode all other factors, e.g. the variation in the Cr content, which shows up under unirradiated conditions [4]. The shift of DBTT for the recently investigated materials, especially for the boron-reduced OPTIFER alloys, nicely fits to the former results and confirms the suspected role of the boron content.

4. Conclusions

The results of preceding investigations [1–3] have been confirmed for the recently examined materials. All materials show irradiation hardening which decreases with decreasing irradiation temperatures.

The OPTIFER steels with reduced boron content show a much better embrittlement behaviour after neutron irradiation than the former alloys. Both the irradiation-induced shift of DBTT and the absolute value of DBTT are in the range of or even below those found for the ORNL and F82H steels. At irradiation temperatures of 250 and 300 °C, the worsening in DBTT is significantly reduced. The transition temperatures of the OPTIFER-V steel are the smallest ones of all materials investigated in the FZK irradiation programmes.

The influence of the boron content on the embrittlement behaviour is confirmed by the present investigation. In future irradiation programmes, this result will be subject of further research.

The influence of the pre-irradiation heat treatment is not affected by irradiation – different treatments on identical material show a similar shift in USE and DBTT after irradiation but keep their difference in the absolute values.

Though the low neutron flux of this irradiation experiment does not yet permit to draw any general conclusions, it can be stated that all examined low-activation materials with reduced boron contents provide significantly better impact properties than the corresponding alloys with higher boron contents. Especially the results of the OPTIFER-V steel (similar to EUROFER 97) raise hope for the newly developed steels as far as their mechanical properties (strength and ductility), embrittlement behaviour (lower loss of ductility after irradiation), and producibility (e.g. producibility on the industrial scale, processability) are concerned. In view of the favourable irradiation embrittlement behaviour of those steels, ferritic–martensitic alloys with reduced boron content should be considered potential materials for fusion applications.

Further irradiation experiments have to verify these encouraging results obtained for LAA at considerably higher and more fusion-relevant dose levels, as it is now done in the SPICE irradiation programme (15 dpa, 250–450 °C). Besides, the complexity of the temperature dependence will probably be reduced at higher fluxes.

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