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Journal of Nuclear Materials 295 (2001) 16–20

journal of nuclear materials

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Embrittlement behaviour of different international low activation alloys after neutron irradiation

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Received 12 October 2000; accepted 17 February 2001

Abstract

The embrittlement behaviour of ferritic/martensitic steels after irradiation in the Petten high flux reactor (HFR) was investigated by instrumented Charpy-V tests with subsize specimens. The main objective, apart from studying effects of particularly low doses, was a comparison of low activation alloys (LAA) from various countries with different Cr contents and different types and concentrations of minor alloying elements and impurities. In the present report, the results of another three materials (OPTIMAR, OPTIFER-IV, GA3X) obtained within the second phase of the MANITU programme (0.8 dpa, at 250–450°C) were analysed and assessed in comparison to the results of the first irradiation up to 0.8 dpa. The evaluation clearly showed a reduced embrittlement problem for the advanced reduced-activation alloys. Of the examined alloys, the GA3X steel shows the very best embrittlement behaviour after neutron irradiation. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

The MANITU irradiation and fracture toughness testing programme on promising low activation ferritic/martensitic steels as reported at the preceding ICFRM-8 [1] has been complemented by data of another three alloys. Their impact test data cover irradiation temperatures of 250°C/300°C/350°C/400°C/450°C at a dose of 0.8 dpa. Particular emphasis was put on the behaviour of the different alloys in comparison to each other, especially at the critical low irradiation temperatures. The previous investigation at irradiation parameters of 250°C/0.8 dpa [2] had shown that the US-heat, referenced as 'ORNL', had yielded 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 Tables 1 and 2), would have suggested a different behaviour [3].

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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 was used in the previous investigations already 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 in the Hot Cells. The test and evaluation procedure were also identical with those employed in previous investigations [1–3]: 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 warming, cooling, and loading system (180–600°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 DBTT (temperature at USE/2) were derived, see Fig. 2.

PII: S0022-3115(01)00508-6

Table 1
Heat treatment and selected properties of unirradiated materials

	Heat treatment	Hardness (HV2)	Grain size (μm)	USE (J)	DBTT (°C)	Dynamic yield stress at 100°C (MPa)
OPTIFER-IV	1000°C/0.5 h + 730°C/2 h	238	13 ± 2	8.9	−86	570
GA3X	1000°C/1 h + 700°C/2 h	256	55 ± 5	9.4	−62	650
OPTIMAR	965°C/2 h + 075°C/0.5 h + 700°C/2 h	308	25 ± 5	6.4	−21	730
OPTIFER-Ia	1075°C/0.5 h + 780°C/2 h	199	50 ± 5	10.0	−80	470
F82H	1040°C/0.5 h + 750°C/2 h	227	35 ± 5	10.7	−70	540
ORNL 3971	1050°C/0.5 h + 750°C/1 h	245	25 ± 5	9.2	−80	550
MANET-I	980°C/2 h + 1075°C/0.5 h + 750°C/2 h	279	30 ± 5	6.6	−30	670

Table 2
Chemical composition of the different alloys in wt%

	LAA		10–11% Cr–NiMoVNb steel	LAA[a]			10–11% Cr–NiMoVNb steel[a]
	OPTIFER-IV	GA3X	OPTIMAR	OPTIFER-Ia	F82H	ORNL 3971	MANET-I
Cr	8.5	9.17	10.5	9.3	7.73	8.9	10.8
W	1.16	2.12		0.965	2.06	2.01	
Mn	0.6	0.042	1.22	0.5	0.083	0.44	0.76
N	0.06	0.0018	0.033	0.015	0.0027	0.0215	0.02
Ta	0.15	0.011		0.066	0.018	0.06	
C	0.11	0.159	0.11	0.1	0.092	0.11	0.14
P	0.004		0.004	0.0047	0.0031	0.015	0.005
S	0.004		0.003	0.005	0.0031	0.008	0.004
V	0.23	0.314	0.21	0.26	0.189	0.23	0.2
B	0.003	0	0.0072	0.006	0.003	<0.001	0.0085
Si	0		0.31	0.06	0.091	0.21	0.37
Ni		0.021	0.60	0.005	0.032	<0.01	0.92
Mo		0.0077	0.63	0.005	0.0053	0.01	0.77
Al		0.015	0.014	0.008	0.01	0.017	0.054
Co		0.003	0.003		0.0024	0.012	0.01
Cu		0.0017		0.035	0.0059	0.03	0.015
Nb		0.011	0.21	0.009	0.0057		0.16
Zr						<0.001	0.059
Ti	<0.02	0.001		0.007	0.0104	<0.01	
Fe	Balance	Balance	Balance	Balance	Balance	Balance	Balance

[a] Alloys from former investigations.

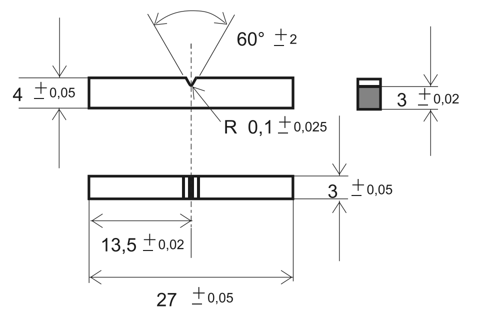

Fig. 1. Miniaturised KLST Charpy specimen (all dimensions in mm).

Dynamic yield stress was derived as stated in [2] from the original force vs. deflection curve at the onset of plastic deformation. Dynamic yield stress was determined at 100°C for all materials, except for the GA3X steel: because of its low DBTT, test temperatures and, thus, the dynamic yield stress vs. temperature curve ended up at 26°C. If suggestive, this curve was extrapolated up to values of 100°C.

All irradiations of the MANITU programme were carried out in the HFR, Petten. The target value of 0.8 dpa was reached at least within a range −2% to +15%, depending on irradiation temperature and, thus, on the vertical core position of the specimens. The irradiation

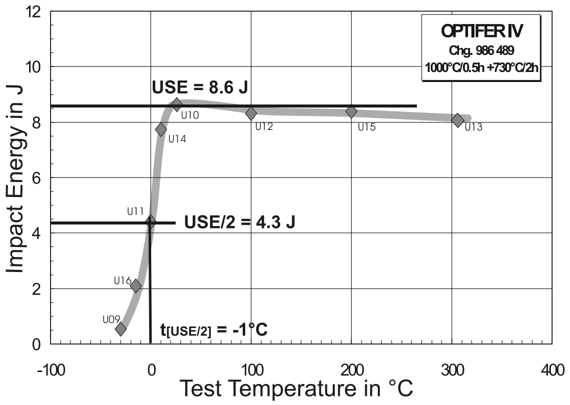

Fig. 2. Determination of USE and DBTT (example: OPTIFER-IV irradiated at 300°C).

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.

Between five and eight specimens for 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 diagrams shown in Fig. 2. These results were combined and grouped in curves with the irradiation temperature as abscissa and the materials as parameter.

3. Results and discussion

The results, including those for some selected materials from [2], are presented in Figs. 3–7.

Fig. 3 shows the USE as a function of irradiation temperature. As already assessed in [2], the low activation alloys (LAA) generally maintain a high impact

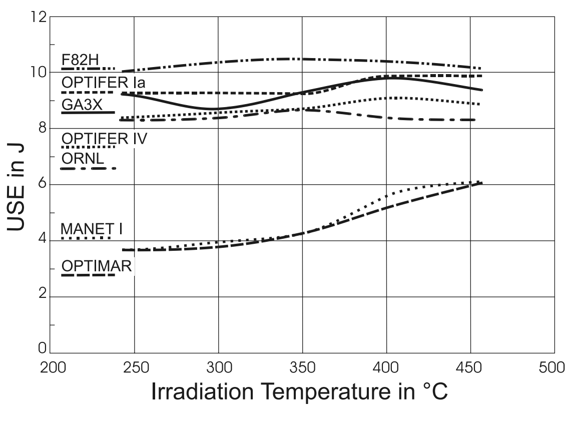

Fig. 3. USE vs. irradiation temperature (parameter: materials).

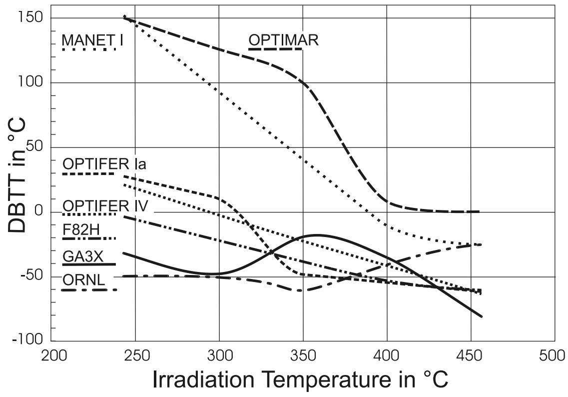

Fig. 4. DBTT vs. irradiation temperature (parameter: materials).

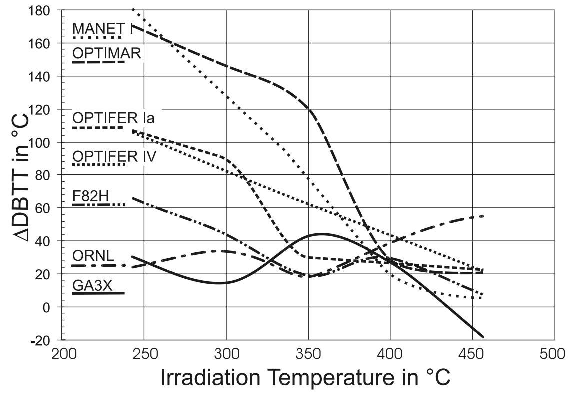

Fig. 5. Irradiation-induced shifts of DBTT vs. irradiation temperature (parameter: materials).

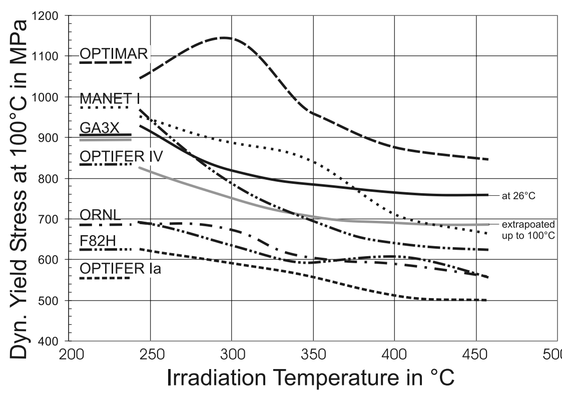

Fig. 6. Dynamic yield stress measured at 100°C (26°C) vs. irradiation temperature (parameter: materials).

toughness in the whole temperature range. The OPTIFER-IV and GA3X steels are found to rank in the middle of those steels, especially GA3X does not show

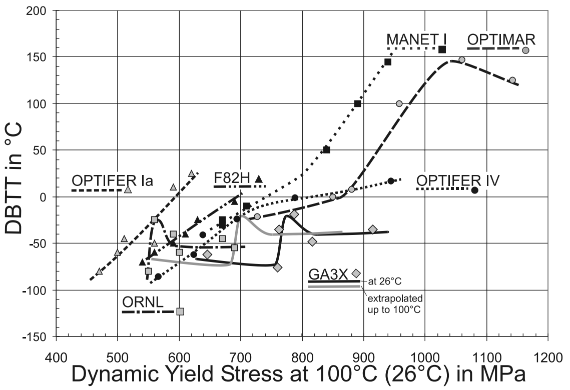

Fig. 7. Embrittlement vs. hardening (parameter: materials).

any clear dependence on the irradiation temperature. In contrast to this, the OPTIMAR steel behaves – similar to the MANET steels – more poorly, but shows some recovery above 350°C. The same picture could be conveyed from a presentation of incremental values, ΔUSE, normalised to the USE before irradiation. The OPTIFER-IV and GA3X steels show the lowest decrease of USE in comparison to the other LAA, and even a slight increase of USE can be determined in the upper temperature range. The OPTIMAR steel suffers from a significant reduction of USE in the order of 40% at irradiation temperatures of 250°C and 300°C.

Having a look at the DBTT as a function of the irradiation temperature (Fig. 4), it is again found that the OPTIMAR steel behaves poorly at the low irradiation temperatures, similar to the MANET steels. The OPTIFER-IV and GA3X steels are significantly better, they are in the range of the formerly investigated LAA.

The shift in DBTT as shown in Fig. 5 reveals essentially the same behaviour. The GA3X steel shows an increase of the DBTT, which is as small as that of the best qualified materials in this respect [2]. At an irradiation temperature of 450°C, the DBTT is even lower than that for the unirradiated material. In view of this result, it should be kept in mind that the materials were not subjected to an optimised heat treatment for Charpy tests.

Dynamic yield stress determined at 100°C, as shown in Fig. 6, exhibits a similar tendency as DBTT. However, the OPTIFER-IV and GA3X steels show a stronger hardening than the other LAA, despite their equal or smaller tendency to embrittlement.

This leads to an interesting correlation between DBTT and σDy, revealing an almost straight line of similar slope for some materials (Fig. 7): OPTIFER-Ia, MANET-I, F82H, OPTIMAR in the middle range of irradiation effects, and OPTIFER-IV for lower irradiation effects. This line was also found for other alloys (OPTIFER-II, MANET-II) in prior investigations [2]. The GA3X and ORNL alloys show more complex curves. These steels and the OPTIFER-IV steel for stronger irradiation effects have more depressed lines.

It is significant that the LAA generally suffer from less irradiation hardening and considerably less irradiation-induced embrittlement compared to MANET/OPTIMAR steels. The OPTIFER-IV steel exhibits the same order of embrittlement as OPTIFER-Ia, but hardening is much stronger. Another promising result is the similar behaviour of the GA3X and the ORNL steels. They do not show any significant change in low temperature embrittlement.

To allow for a better quantitative comparison of the different materials, the dose dependence of DBTT was plotted for $T_{irr} = 300°C$ in [1]. 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 results obtained for the OPTIMAR, OPTIFER-IV, and GA3X steels irradiated up to 0.8 dpa, see Fig. 8.

In [4], an influence of helium on impact toughness was suspected. It was described in detail in [1]: 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 transforms to helium and lithium already at moderate neutron fluxes. Helium is found as bubbles in the crack surfaces of irradiated materials. Complementary investigations using He-implanted Charpy-V specimens [5] showed an embrittlement effect of a higher helium content that by far exceeded the irradiation effect of the original material: at 0.2 dpa, the shift in DBTT increased from 18°C to 42°C.

In [1], a model for calculation of the irradiation-induced ^{10}B burn-up was presented. An exponential growth function was applied and the neutron calculations of HFR Petten were used to determine the constants:

$$N(\mathrm{He}) = N_0(^{10}\mathrm{B})(1 - \exp(-D/\tau)),$$

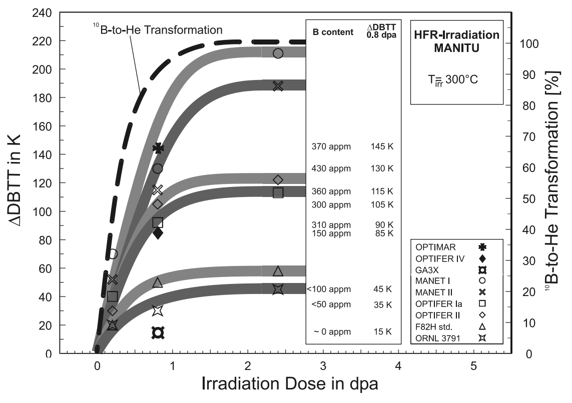

Fig. 8. Irradiation-induced shifts of DBTT and ^{10}B-to-He transformation vs. irradiation dose (parameter: materials).

where τ represents the burn-up constant for a decrease to 37% of ^{10}B (0.34 dpa) and D the irradiation dose. After about 1.6 dpa, 99.3% of the burn-up is reached.

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 was 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 [3]. The shift of DBTT for the three newly investigated materials nicely fits to these former results and can be related to the boron content.

4. Conclusions

The results of preceding investigations [1–3] have been confirmed for the three newly examined materials. All materials show irradiation hardening which decreases with higher irradiation temperatures. The dynamic yield stress and the irradiation-induced shift of the OPTIMAR steel are much higher at low irradiation temperatures compared to the low activation alloys.

The GA3X steel shows the very best embrittlement behaviour after neutron irradiation. At irradiation temperatures of 250°C and 300°C, the worsening in DBTT is the smallest one of all materials investigated.

Though the low neutron flux of this irradiation experiment does not yet permit to draw any general conclusions, it can be stated that all low activation materials examined clearly provide better impact properties than the corresponding MANET/OPTIMAR alloys. In view of the favourable irradiation embrittlement behaviour of the GA3X and ORNL steels, ferritic–martensitic Cr–WVTa alloys should be considered as potential materials for fusion applications. Especially the results for the OPTIFER-IV steel raise hope for the newly developed steels, e.g., EUROFER 97, concerning their mechanical properties (strength and ductility), embrittlement behaviour (lower loss of ductility after irradiation), and producibility (e.g., producibility in industrial scale, processability) at the same time.

Further irradiation experiments will have to verify these encouraging results obtained for LAA at considerably higher and more fusion-relevant dose levels. Besides, the complexity of the temperature dependence will probably be reduced at higher fluxes.

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 to further research and taken into consideration by minimising the boron content in material development.

Acknowledgements

The Charpy impact tests were conducted at the Central Experimental Engineering Department/Hot Cells by the Post-irradiation Examination Group II headed by O. Romer. Here, the authors would like to thank him and his staff members, W. Kunisch and A. Drasl, for their co-operation in running the tests and their support in setting up the test facilities. We also would like to thank M. Rieth for preparing and managing the MANITU programme. Last but not the least, our thanks go to D.S. Gelles for his support of the work regarding the GA3X steel. This work has been performed within the framework of the Nuclear Fusion Project of the Forschungszentrum Karlsruhe and is supported by the European Union within the European Fusion Technology Programme.

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