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Effect of hydrogen on the ductility reduction of F82H martensitic steel after different heat treatments

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Abstract

The influence of heat treatment on hydrogen embrittlement (HE) susceptibility of F82H martensitic steel, a candidate material for the first wall and blanket of the DEMO reactor, was investigated by means of low strain rate tests conducted at room temperature on notched cylindrical specimens, pre-charged with hydrogen. Three types of thermal treatment were compared, which produced quite different material hardness and strengths. As a general rule, F82H steel exhibited a strong susceptibility to HE in all conditions, with transitions between ductile and brittle behaviour ranging from 0.5 to 1.0 wppm H. The comparison of results produced by the different heat treatments confirmed for F82H also the tendency toward an increase of the susceptibility with material hardness. Moreover, the examined steel appeared to be more sensible to HE than another martensitic steel (MANET II), which was also proposed as a candidate material for fusion reactor first wall application.

1. Introduction

Since about 30 years, ferritic/martensitic (FM) steels have been proposed as structural materials for the first wall and blanket structures of fusion reactors, instead of austenitic steels. This was mainly based on irradiation experience in fast reactors, which indicated FM steels to be more resistant to swelling phenomena [1]. Moreover, these steels also show a higher thermal conductivity and a lower thermal expansion coefficient, which appeared as favourable properties in order to reduce thermal stresses in operating conditions. Finally, compatibility studies with potential breeders and coolants, like lithium or the lithium–lead eutectic alloy, showed that FM steels corrode at a remarkably lower rate [1,2].

Therefore, in the last 15 years research programs have been organized in Europe, USA and Japan, aimed at developing low-activation FM steels, in which some elements like molybdenum and niobium were substituted (e.g. by tungsten, tantalum, etc.) in order to obtain a rapid decay of irradiation-induced radioactivity. Preliminary results indicated that martensitic steels containing 7–9% Cr are preferable to steels having a higher Cr content, both for first wall and blanket applications, due to the total absence of δ-ferrite and to the consequent increase in toughness and impact strength [1,3].

Among low-activation martensitic steels, F82H has been proposed and characterised in the last few years with encouraging results, particularly as far as tensile and impact properties are concerned and including also the effects of irradiation. Nevertheless, a few additional properties need to be investigated, among which hydrogen embrittlement (HE) susceptibility must surely be included. Indeed, preliminary analysis indicated that, in nuclear fusion reactors, the content of hydrogen isotopes in structural materials might reach rather high values, at least under some operating conditions. Hydrogen production rate in first wall steels, due to (n, p) transmutation reaction, is predicted to be about 0.9 wppm/dpa under 14 MeV neutrons irradiation [4,5]. Moreover, additional external hydrogen sources (e.g. aqueous corrosion, radiolysis of cooling water, hydrogen added to cooling or purging gas) must be taken into account [6].

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Due to such sources and to the low-diffusivity and high-trapping site concentration, the hydrogen content in structural materials may be quite high for low-temperature operating conditions [7]. As an example, at 100°C the steady state hydrogen concentration in the first wall can reach several tens wppm [4].

For temperatures above 300°C, the increasing diffusivity together with a strong reduction of the natural trapping sites should induce a strong reduction of hydrogen content, to a few wppm under steady state conditions. However, irradiation-induced hydrogen traps may remarkably increase the hydrogen content even at high temperature. Accounting for such mechanisms, an estimate of about 35 wppm of hydrogen was obtained, for the first wall of DEMO WCLL blanket, under steady state conditions [8].

In the present paper, the influence of heat treatment on HE susceptibility of F82H martensitic steel was investigated. The study was based on low strain rate tests conducted at room temperature using notched cylindrical specimens. Before the tests, specimens were electrochemically charged with hydrogen. The behaviour of the material after three distinct thermal treatments and with different hydrogen contents was investigated. The 'as-received' material was in the normalised (austenitisation and quenching) state; in addition, two different tempering treatments were applied, having different tempering temperatures at 625°C and 750°C. It is worth noting that the 'as-received' material status is not actually suitable for structural applications, due to its intrinsically low ductility. However, it was included in the present study for comparison and because it can be considered representative of conditions possibly occurring in localised regions of actual components, such as welded joints not subjected to adequate Post Weld Heat Treatments.

After a detailed explanation of the test procedures, the results are discussed, highlighting the HE susceptibility of F82H steel and the influence of heat treatments and hardness. Moreover, a comparison with MANET II (DIN 1.4914) martensitic steel, which was previously characterised [9], is also included.

2. Material and experimental procedures

The composition of F82H steel heat employed in the present work is reported in Table 1.

The 'as-received' material was in the normalised state (austenitisation at 1040°C for 1/2 h, cooling in air) and will be indicated in the following as 'not tempered' (NT). In addition, material subjected to two different tempering treatments was employed. The first treatment included 1 h at 625°C and cooling in air (T625 material) while the second treatment included 2 h at 750°C followed by cooling in air (T750 material). The latter was the typical tempering treatment for this type of steel, while the former was introduced in order to achieve intermediate values of hardness (300–330 HV30) and yield or tensile strength. This allowed to better investigate the relationship between HE susceptibility and hardness or yield strength for this type of material. The mean grain size obtained by the applied procedure was ASTM 7-8 (23–32 μm) [10].

Specimens for HE susceptibility tests were machined according to the shape shown in Fig. 1, with specimen axial direction parallel to the rolling direction.

Before the test, the specimens were charged to the required hydrogen concentration by means of an electro-chemical apparatus, in which the specimen works as cathode in a 0.05 M H_2SO_4 solution with some drops of thiourea as hydrogen promoter, while a platinum plate is the anode. Current densities ranging from 1 to 100 A/m^2 have been used to obtain different hydrogen concentrations. Specimens were maintained in the electrochemical cell for at least 16 h, so as to allow a rather uniform hydrogen concentration to be achieved in the specimen volume. Since this procedure does not allow the hydrogen content to be controlled with sufficient accuracy, the actual hydrogen concentration of each specimen was measured immediately after the test, using

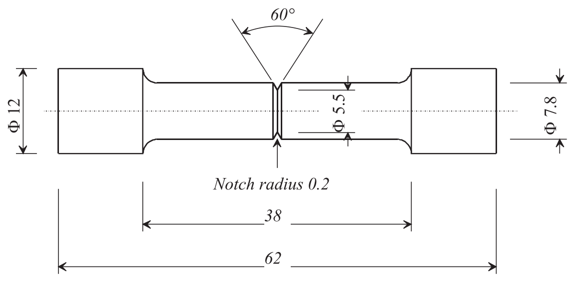

Fig. 1. Geometry of the notched specimens used for HE tests (dimensions in mm).

Table 1
Chemical composition of F82H steel (wt%)

C	Cr	Ni	Mo	V	Si	Mn	S
0.09	7.68	0.02	0.003	0.16	0.11	0.11	0.002
P	B	N	Ta	Al	Cu	W	Ti
0.002	0.0002	0.007	0.02	0.003	0.01	1.96	0.01

a mass spectrometer connected to a hydrogen desorption apparatus.

Tests were carried out making use of an electro-hydraulic MTS test frame, operated under displacement control. An extensometer having 25.4 mm gage-length was used to measure average strain in the central portion of the specimens including the notch. During the tests, the average strain rate in the calibrated cylindrical region of the specimen was set to 2.5×10^{-6} s^{-1}. Based on available results obtained on similar materials, such strain rate was assumed to be sufficiently low that further reduction would not significantly affect the material behaviour.

Due to the presence of the notch, the area reduction coefficient Z (%), appeared to be the most suitable parameter in order to quantify the effects of HE on the mechanical behaviour. This required to measure with rather high accuracy the area of the fractured surface after the test. To this end, a semi-automatic procedure was set up, which employed a video camera connected to a PC to obtain a digital image of the fracture surface and a specific elaboration program developed within the MATLAB environment to estimate the area via a subpixeling technique. Controlled illumination conditions were employed in order to increase accuracy and reproducibility. Through some qualification tests, which were performed on specifically manufactured areas, it was possible to estimate an accuracy of about 0.5% for the applied procedure.

For each heat treatment, eight tests were performed with notched specimen having different hydrogen contents, in order to achieve a complete characterisation of its effects on the mechanical behaviour of the material. In addition, standard tensile tests were performed for each heat treatment on smooth (unnotched) specimens not charged with hydrogen (virgin material), in order to characterise the tensile behaviour of the basic material. All the tests were performed at room temperature (20–25°C).

3. Results and discussion

Results for T750 F82H, tempered at 750°C, were already reported in [9]. They are, however, included in the present paper for comparison with other heat treatments.

The stress–strain curves obtained in smooth specimen tests conducted on virgin material are reported in Fig. 2 for the three different heat treatments, while related main mechanical properties, including hardness (HV10), yield stress (YS), ultimate tensile stress (UTS), total elongation (El) and area reduction (Z) at failure are summarised in Table 2. Observed values for the most commonly used status, i.e. T750, appear in satisfactory agreement with those typical for this material.

A significant increase of hardness and of yield and tensile strength, accompanied by lowering of the ductility is observed when passing from the NT material to material tempered at 625°C and, to a greater extent, at 750°C.

Fracture surfaces of virgin NT F82H showed a mixed failure mechanism, with zones of ductile fracture with microvoids and zones of brittle transgranular fracture (Fig. 3). These brittle zones were not observed in tempered specimens, which showed a completely ductile fracture surface.

The results obtained in HE tests conducted on notched specimens are reported in Table 3, while in Figs. 4–6 the area reduction coefficient Z (%), is plotted vs. hydrogen content for NT, T625 and T750 materials, respectively.

From Table 3 and Figs. 4–6, an abrupt reduction in material ductility can be clearly observed, as soon as the hydrogen content exceeds a rather narrow threshold

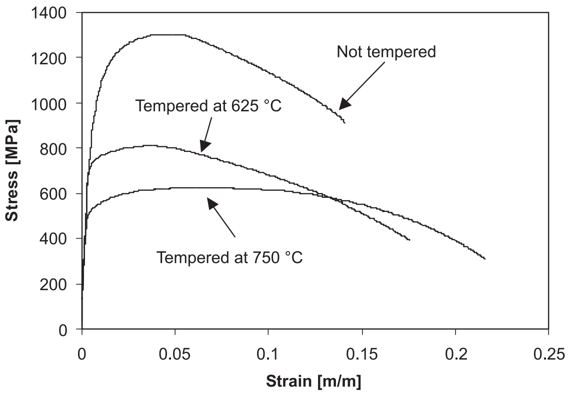

Fig. 2. Stress–strain curves for F82H steel after different heat treatments.

Table 2
RT mechanical properties of F82H steel after different heat treatments

Material	Hardness (HV30)	YS (MPa)	UTS (MPa)	El (%)	Z (%)
Not tempered	400–410	960	1304	14.1	50.3
Tempered at 625°C (T625)	300–330	729	810	17.1	76.5
Tempered at 750°C (T750) [8]	200–210	515	625	21.6	79.1

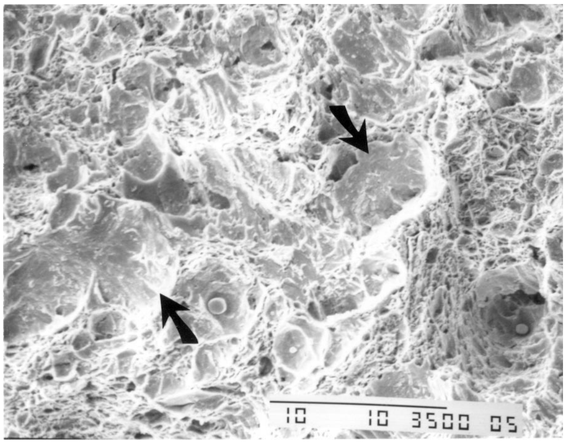

Fig. 3. SEM image of the fracture surface for virgin NT material showing typical micro-dimples and, in the zones indicated by the arrows, isolated transgranular pseudo-cleavage facets. The bar corresponds to 10 μm.

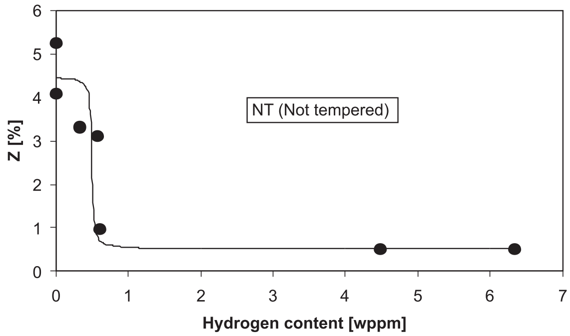

Fig. 4. Area reduction coefficient Z vs. hydrogen content for NT material.

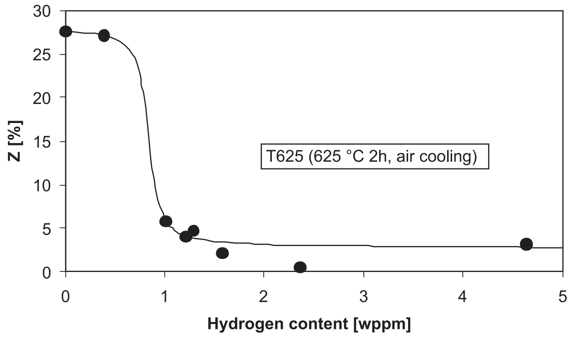

Fig. 5. Area reduction coefficient Z vs. hydrogen content for T625 material.

range. Beyond this threshold, the area reduction drops to very low values, usually less than 10% of the value typical of the virgin material.

The macroscopically brittle behaviour obtained in these conditions was clearly confirmed by microscopical observations of the fracture surfaces. See for instance Fig. 7, showing a detail of the fracture surface obtained for NT F82H and a 0.61 wppm H content, where a clear intergranular fracture with the presence of secondary cracks can be observed.

The transition between ductile and brittle behaviour was observed to occur for increasing hydrogen content, passing from NT to T625 and to T750 material. However, for all the analysed heat treatments, the negative effects of hydrogen could be noted even for very low concentrations, i.e. less than 1–2 wppm H.

These results could appear to contrast with those obtained by Hara et al. [12], who did not observe any significant hydrogen effect for F82H steel. However, it is worth noting that tests reported in [12] were actually conducted with very low-hydrogen contents (less than 0.5 wppm) and with unnotched specimens.

Since stress triaxiality due to notches is known to produce an enhancement of the local hydrogen content as compared with the bulk value [13,14] (an increase up to 6.9 times can be theoretically estimated under favourable conditions), it can be reasonably argued that, immediately before fracture, actual notch region concentrations in the present tests were probably significantly higher than the bulk values reported in Table 3.

Table 3
Results of tensile tests on notched specimens (room temperature)

NT		T625		T750	
H content (wppm)	Z (%)	H content (wppm)	Z (%)	H content (wppm)	Z (%)
0.0	4.1	0.0	27.7	0.0	43.4
0.0	5.2	0.4	27.2	1.1	16.2
0.3	3.3	1.0	5.8	1.4	17.4
0.6	3.1	1.2	4.1	1.7	17.7
0.6	1.0	1.3	4.7	2.1	13.9
4.5	<0.5	1.6	2.1	2.7	7.0
6.3	<0.5	2.4	<0.5	3.1	7.2
		4.6	3.2		

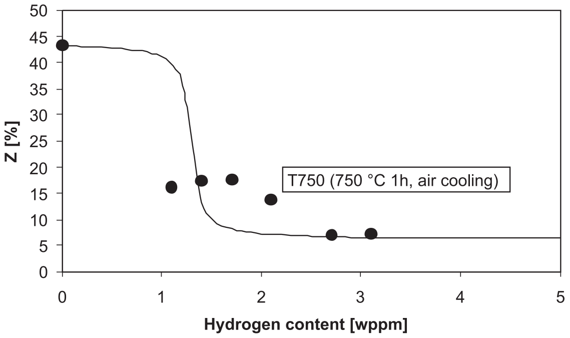

Fig. 6. Area reduction coefficient Z vs. hydrogen content for T750 material [8].

Therefore, the results reported in [12] are not surprising, as applied hydrogen contents were simply too low to produce observable effects.

Based on the present results, the susceptibility of F82H steel to HE must be considered high, at least at low temperatures, even if the enhancing effect of notch stress field triaxiality is considered. Further studies are therefore required for better evaluating the suitability of this steel for fusion reactor first wall applications, taking into account the operating conditions, stress states and the hydrogen content of the actual components.

In order to achieve a more independent and quantitative comparison of the HE susceptibility of the three heat treatments, it was decided to obtain an analytical representation of the Z (%) dependence on the hydrogen content. To this end, the following function was found to be particularly suitable:

$$Z(\mathrm{H}) = a + b\,\mathrm{arctg}\left(\frac{\mathrm{H} - c}{d}\right), \qquad (1)$$

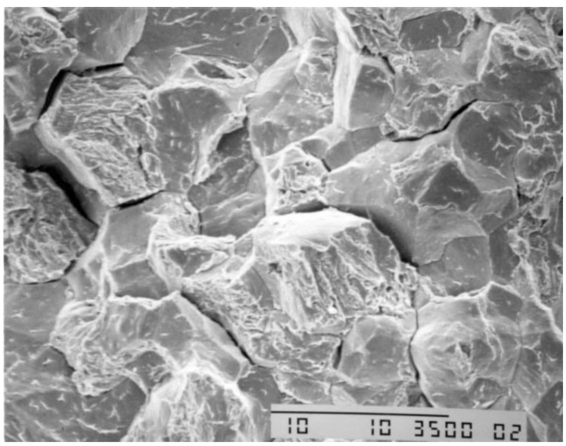

Fig. 7. SEM image of the fracture surface for NT material (0.61 wppm H content), showing typical intergranular H-fracture with several secondary cracks. The bar corresponds to 10 μm.

where H is the hydrogen content and a, b, c and d are coefficients to be obtained by fitting the experimental results. These coefficients are reported in Table 4 for the three types of materials, while related curves according to Eq. (1) are superimposed to experimental data in Figs. 4–6.

The comparison of such curves (Fig. 8) confirmed the observation on the relatively high HE susceptibility of F82H steel and its increase with increasing material yield strength and hardness. Coefficient c in Eq. (1), which represents the hydrogen content corresponding to the flex point in the curves, was assumed to be an estimate of the threshold for HE effects. This threshold is represented in Fig. 9 as a function of the yield strength of the material, showing a fairly linear decreasing trend.

Table 4
Coefficients for Eq. (1), H in wppm

	a	b	c	d
NT	0.92	0.43	0.313	0.011
T625	15.51	8.19	0.84	0.069
T750	25.1	12.1	1.3	0.065

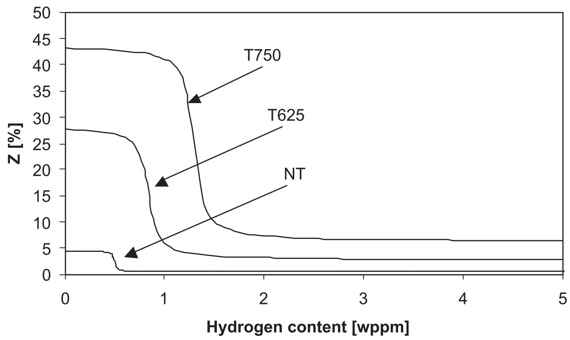

Fig. 8. Comparison of best fit curves for the three materials.

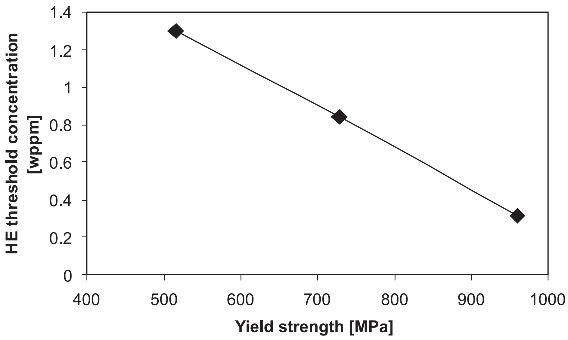

Fig. 9. Threshold concentration for HE phenomena, estimated by 'c' coefficient in Eq. (1), vs. yield strength for the three materials.

The comparison of the present results with those obtained for MANET II steel, for which the threshold for HE in the case of notched cylindrical specimens was found to be about 5 wppm [9,12], clearly indicates the higher sensitivity of F82H. As a consequence, a great caution in proposing this type of material as a candidate for first wall and blanket structural application seems to be required.

4. Conclusions

Low strain rate tests have been performed on hydrogen charged notched cylindrical specimens of F82H, a low-activation martensitic steel proposed for fusion reactor first wall applications. Three types of thermal treatment were compared: as-received (NT) material, which was in the normalised (austenitisation and quenching) state and material tempered at two different starting temperatures of 625°C and 750°C.

The analysis of the results confirmed the tendency toward an increase of HE susceptibility with increasing hardness and yield strength, as typical for many other materials. However, the results highlighted that F82H steel, in both NT and tempered forms, shows a great susceptibility to HE, with threshold concentrations for the transition from ductile to completely brittle behaviour ranging from 0.5 wppm for the NT material to 1.5 wppm for the material tempered at 750°C.

These values, which are rather low also in comparison with those shown by other candidate materials, such as MANET II, in similar conditions, appear to indicate that some caution and further investigations are required before this type of martensitic steels can be proposed for the first wall or the blanket of a fusion reactors. This is even more true, if account is taken of the many factors typical of reactor operating conditions, such as stress triaxiality, stress gradient and temperature [11], which may affect the material behaviour.

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