THE INFLUENCE OF HYDROGEN ON THE TOUGHNESS OF REACTOR PRESSURE VESSEL STEELS

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1. Introduction

Low-alloyed ferritic steels are, just like high-alloyed austenitic steels, susceptible to H-embrittlement. In a nuclear reactor, corrosion or radiolysis lead to the formation of H-atoms on the inner surface of the reactor pressure vessel and thus to a H-uptake during reactor operation.

In case the structural defects which arise as a result of irradiation become trapping centres for the H-atoms, safety-affected synergisms between irradiation- and hydrogen-embrittlement need to be considered.

Earlier investigations have shown that a H-content larger than 2.5 – 4 ppm effects a clear decrease in toughness and above all causes total embrittlement for high strength steels ($R_m \geq 1200$ MPa) [1]. After several years of exposure in the pressurized water of the reactor near the core, these critical concentrations are not reached [2].

Pachur [3] developed a hypothesis which postulates an immediate link between radiation embrittlement and the presence of hydrogen. Knowing that the phenomenon is not encompassed by the surveillance programmes, makes it of high relevance with regard to technical safety. Notwithstanding many findings which contradict Pachur’s hypothesis [4], the results described above should be seen as an incentive to reexamine earlier data on a more advanced experimental basis.

The paper reports about new experiments in this field and portray data about hydrogen diffusion behaviour, the solubility of hydrogen and the influence of hydrogen on mechanical properties. These data were taken from experiments on a variety of unirradiated pressure vessel steels under conditions corresponding to those present in pressurized water reactors.

2. Experimental programme

The following reactor pressure vessel steels were investigated:

- A 508 Cl. 3 (Code: JFL)
- A 533 B Cl. 1 (Code: JRQ)
- 15 Xh2NMFAA (Code: WWER) and
- 22NiMoCr 3.7 (Code: A2).

As was one weld material:

- 10XhMFT.

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The diffusion of hydrogen was evaluated from permeation experiments after electrochemical charging in two stages: $i = -0.5$ and $-5 \text{mA/cm}^2$ in $0.1 \text{n NaOH}$ with $5 \times 10^{-5} \text{ mol/l As}_2\text{O}_3$ in the $25 - 80 \degree \text{C}$ temperature range.

An assessment of the amounts of differently bound hydrogen and their dependence on the temperature and charging parameters was made by means of a hydrogen analyser (LECO-RH 402) or using a mass spectrometer after thermo-desorption at constant heating rates. Besides charging in $0.1 \text{n NaOH (+ As}_2\text{O}_3)$, charging in boric acid/KOH - corresponding to PWR-operating conditions - was carried out.

In order to characterize the influence of hydrogen on the strength and toughness of the materials, tensile tests were performed at strain rates of $10^{-6} - 10^{-4}/\text{s}$ in a temperature range of $25 - 250 \degree \text{C}$ after electrolytic H-precharging and during H-charging at a cathodic flux ranging from $-0.5$ to $-5 \text{ mA/cm}^2$ in a simulated environment representing the water in a reactor pressure vessel. Finally, H-precharged Charpy-specimens were tested in an instrumented Charpy V-notch test up to temperatures of 50 K.

### 3. Results and conclusions

The effective diffusion coefficient and the lattice diffusion coefficient are shown in an Arrhenius plot in Fig. 1. The experiments were performed in two steps with different cathodic current. In the first step the H-diffusion is ingreated by lattice defects which traps the H-atoms (effective diffusion coefficient). After saturation of the potential traps the diffusion is an order of magnitude faster and characterizes the lattice diffusion.

![Arrhenius plot](image)

- **1st stage** $i_c = 0.5 \text{ mA/cm}^2$
  - Effect of traps
- **2nd stage** $i_c = 5 \text{ mA/cm}^2$
  - Traps are saturated

**Fig. 1:** H-diffusion after electrochemical charging in 2 stages, in $0.1 \text{n NaOH}$ and $5 \times 10^{-5} \text{ mol/l As}_2\text{O}_3$. 
Table 1:  H-concentration [ppm] in the initial condition and after charging and effusion at room temperature and 250 °C (inaccuracy: ± 0.025 ppm)

<table>
<thead>
<tr>
<th>Material</th>
<th>Initial condition</th>
<th>Boric acid / KOH solved + trapped</th>
<th>Boric acid / KOH solved + trapped</th>
<th>0.1n NaOH + As2O3 solved + trapped</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>RT</td>
<td>250°C</td>
<td>RT</td>
</tr>
<tr>
<td>15Xh2NMFA</td>
<td>0.78</td>
<td>1.14</td>
<td>0.75</td>
<td>1.17</td>
</tr>
<tr>
<td>A533 - JRQ</td>
<td>0.39</td>
<td>0.64</td>
<td>0.5</td>
<td>0.81</td>
</tr>
<tr>
<td>A508 – JFL</td>
<td>0.90</td>
<td>1.47</td>
<td>0.74</td>
<td>0.81</td>
</tr>
<tr>
<td>22NiMoCr3.7</td>
<td>0.75</td>
<td>0.66</td>
<td>0.67</td>
<td>1.3</td>
</tr>
</tbody>
</table>

The JFL, JRQ and A2 steels show a diffusion behaviour which is comparable in both permeation steps. In the 15Xh2NMFAA (WWER) steel, however, the diffusion is clearly slower, in the first, as well as in the second permeation step. This may be related to a higher defect concentration and a different alloy composition (higher Cr-content) for this steel. The H-concentration after charging at room temperature and at 250 °C and the effusion at room temperature are given in table 1. Independently of the charging parameters and the material, the H-concentrations vary very little and remain well below the critical concentration of about 3 ppm.

Fig. 2: Ultimate tensile strength $R_m$, yield stress $R_{p0.2}$ and total elongation $A$ as a function of the strain rate under H-charging with $-5$ mA/cm² in 0.1 n NaOH + $5 \times 10^{-5}$ As₂O₃ for A508 Cl. 3 (JFL)

Accordingly, no or hardly any influence of H-charging on the mechanical properties can be evidenced. A small decrease of the fracture toughness with increasing H-charging can be observed, but is considered negligible. Fig. 2 reveals the dependence of the ultimate tensile strength, the yield stress and the total elongation on the strain rate and temperature for the A508 Cl. 3 steel under in-situ charging at $-5$ mA/cm².

The total elongation remains high and independent of the temperature and strain rate. Similar results were obtained also for the other materials. The temperature-dependence is influenced by strain ageing effects which are also observed in the uncharged condition. For the Charpy V-notch tests at temperatures in the ductile-to-brittle transition range, the results for uncharged and H-charged specimens lie within the material inhomogeneity scatter (Fig.3).
Fig. 3: Charpy V-notch impact-temperature-curve for un- and H-precharged A508 Cl 3 (JFL) specimens

Through the experiments prove an uptake of H under PWR-like conditions of water chemistry and operation temperature, an essential effect on the toughness cannot be observed for reactor pressure vessel steels in unirradiated condition. The H-concentrations remain far below the critical values. Even radiolytically induced H-concentrations under flowing coolant conditions are insufficient to create an effect of embrittlement.

On the basis of the above experiments, however, we cannot exclude that for an irradiated material and/or under stagnating coolant conditions, H-induced or –assisted embrittlement takes place.

An extension of the experimental programme to irradiated RPV steels was therefore be initiated.

References