1. Introduction

An extensive irradiation programme was performed at the VVER 2 of the Rheinsberg nuclear power plant between 1983 and 1990. Almost 2000 specimens made from 37 various heats of preferentially VVER pressure vessel steels were irradiated in different irradiation rigs during one reactor cycle each.

The programme should considerably enhance the data base for irradiated materials. Mainly, the influence of detrimental elements, such like phosphorus or copper, and the metallurgical treatment should be analyzed. The final goal was to contribute to the validation of the predictive formula describing the neutron embrittlement as they are formulated in the Russian guides.

The prototype reactor VVER 2 is especially suitable as irradiation facility because the neutron fields and the temperature condition are comparable with the VVER 440. Moreover, the large cross section of the high flux channels allows to irradiate large and numerous specimens.

The programme comprises two parts. A first part is a German-Russian cooperation project. In this part Materialprüfanstalt Stuttgart and Forschungszentrum KFA Jülich were also involved. A second part was designed and prepared by Zentralinstitut für Kernforschung Rossendorf which was the predecessor for Forschungszentrum Rossendorf (FZR).

Testing of the specimens was delayed as the FZR’s hot cell laboratories had been reconstructed. Since 1998 the FZR has had new hot cell laboratories with facilities for preparation and testing of irradiated specimens. In these laboratories the specimens of the first part of the irradiation programme were tested. Some results of the tests are presented in the following.

2. Experimental

The material tested by FZR [1] consists of

- 4 heats from VVER 440-type base metal 15Kh2MFA (15CrMoV 2) (code: R1, R2, R3, D25)
- 2 heats from VVER 1000-type base metal 15Kh2NMFAA (15CrNiMoV2) (code: R16, R17) and
- 1 heat from VVER1000-type weld metal 10KhGNMAA) (10CrMnNiMo 1) (code: R19) [1].

Most of the heats have a rather low content of the harmful elements copper and phosphorus (Tab. 1). Hence the irradiation sensitivity should be low. From each heat, Charpy V-notch standard and precracked specimens were available in the unirradiated, the irradiated and partly the post-irradiation annealed state.
The specimens were taken from the 1/4 to 3/4 thickness position and in L-S, T-S, L-T and T-S orientation. The specimens were placed in open irradiation rigs in the high flux irradiation positions (target channels) with direct contact to the coolant (255 °C inlet temperature). The mean neutron flux rate was 2.6 \(10^{12}\) cm\(^{-2}\) s\(^{-1}\) [E>1MeV]. The fluences of different specimens of one set vary up to a factor of 2. Therefore, the results of the tests were corrected to the same mean fluences. Both the details of the irradiation and the correction procedure are given in [2].

The Charpy impact tests were performed with an instrumented impact pendulum in the temperature range of -150 °C to +300 °C. Testing of the precracked specimens is based on the master curve concept according to ASTM E 1921-97 using the multiple temperature method [3]. The specimens were loaded by three-point bending with a servo-hydraulic test system „MTS 810-Test Star“ in a nitrogen cooled environmental chamber.

3. Results

The two VVER 440 base metal heats (R1 and R3) excepted, in general all investigated materials exhibit a high toughness in the unirradiated state. Particularly the Charpy toughness parameters of the VVER 1000 base metal are excellent. As usual the toughness of the weld metal is lower but sufficient according to the Russian specification. Definitely, irradiation within the investigated fluence range from 23.2 to 138 \(\cdot 10^{18}\) /cm\(^2\) [E > 0.5 MeV] affects a clear degradation of the toughness. Irradiation shifts the ductile-brittle transition temperature to higher temperatures, lowers the upper shelf energy and extends the transition range. Figs. 1 and 2 illustrate this phenomenon for heats of VVER 440 base metal and of VVER 1000 weld metal.

Using the Russian procedure [1] for the evaluation of the irradiation sensitivity

\[
\Delta TT = A_F \cdot 3 \sqrt{F}
\]

an irradiation embrittlement coefficient \(A_F\) can be estimated from the fluence \(F\) (in \(10^{18}\)/cm\(^2\) [E > 0.5 MeV]) and the measured transition temperature shift \(\Delta TT = TT_F - TT_0\). Tab. 1 summarizes the calculated values for \(A_F\). The values range from 3.4 for a 15Kh2MFA heat (R1) to 47.5 for the weld metal (R19). This means that for typical end of life fluences \(\geq 10^{20}\) /cm\(^2\) [E>0.5 MeV] the transition temperature shifts range between 20 and \(\geq220\) °C. Whereas a shift of 20 °C proves an excellent irradiation resistance, a shift of more than 250 °C is not acceptable even if the transition temperature is very low in the initial state.

Table 1: Irradiation embrittlement coefficients \(A_F\)

<table>
<thead>
<tr>
<th>material</th>
<th>code</th>
<th>Cu / weight-%</th>
<th>P / weight-%</th>
<th>(\Phi_{mean}) [x (10^{18}) ncm(^{-2})]</th>
<th>(\Delta TT_{48J}) [°C]</th>
<th>(A_F)</th>
</tr>
</thead>
<tbody>
<tr>
<td>15Kh2MFA</td>
<td>R1</td>
<td>0.10</td>
<td>0.011</td>
<td>43.6</td>
<td>12</td>
<td>3.4</td>
</tr>
<tr>
<td></td>
<td>R2</td>
<td>0.12</td>
<td>0.014</td>
<td>80.7</td>
<td>32</td>
<td>7.4</td>
</tr>
<tr>
<td></td>
<td>D25</td>
<td>0.11</td>
<td>0.017</td>
<td>127.6</td>
<td>99</td>
<td>19.7</td>
</tr>
<tr>
<td></td>
<td>R3</td>
<td>0.12</td>
<td>0.024</td>
<td>47.5</td>
<td>47</td>
<td>13.1</td>
</tr>
<tr>
<td>15Kh2NMFAA</td>
<td>R16</td>
<td>0.07</td>
<td>0.012</td>
<td>46.0</td>
<td>65</td>
<td>18.1</td>
</tr>
<tr>
<td></td>
<td>R17</td>
<td>0.13</td>
<td>0.013</td>
<td>72.7</td>
<td>119</td>
<td>28.5</td>
</tr>
<tr>
<td>10KhNGMAA</td>
<td>R19</td>
<td>0.05</td>
<td>0.008</td>
<td>65.1</td>
<td>191</td>
<td>47.5</td>
</tr>
</tbody>
</table>
Fig. 1: Charpy V impact energy temperature curves of VVER-440 base metal (R3) in the as-received (U), irradiated (I) and annealed (IA) state

\[ A_F = 800 \times (\% P + 0.07 \times \% Cu) \]  \hspace{1cm} (2a)

for weld metal, WWER-440, \( T_{irr} = 270 \, ^\circ C \)

\[ A_{F_T} = A_{F \, 270} + K \left( T_{270} - T_{irr} \right) \]  \hspace{1cm} (2b)

with \( K = 0.2 \) for base metal and \( K = 0.4 \) for weld metal

maximum values for VVER 440, \( T_{irr} = 270 \, ^\circ C \)  maximum values for VVER 1000, \( T_{irr} = 290 \, ^\circ C \)

\[ A_F \leq 15 \text{ weld metal} \hspace{1cm} A_F \leq 20 \text{ weld metal} \]

\[ \leq 18 \text{ base metal} \hspace{1cm} \leq 23 \text{ base metal} \]  \hspace{1cm} (2c)
For the different materials the transition temperature shifts are calculated using equations (1) to (2a-c) and compared with the measurements. The comparison is shown in Fig. 3. On the base of the Russian guidelines always conservative predictions are obtained apart from the weld metal (R19). The very high irradiation embrittlement sensitivity of material R 19 is not correctly predicted. As the material meets the Russian specification regarding both the chemical composition and the heat treatment, this irradiation response is unexpected. Also the microstructure does not give a clue for the irregularity. R19 is a weld material. Weld metal is considered to have the highest susceptibility to embrittlement. Nevertheless, the finding cannot be explained consistently what is a proof for our lack of adequate knowledge on the phenomenon.

An annealing treatment to recover the toughness has become a must for the old VVER-440 s. There are well-tried annealing technologies but, unfortunately, their efficiency has not yet been sufficiently validated. The effect of annealing can be described by a recovery parameter R defined as relative change of the transition temperature TT or the upper shelf energy USE

\[ R = \frac{P_{\text{irr}} - P_{\text{ann}}}{P_{\text{irr}} - P_{\text{unirr}}} \times 100 \ [%] \]  

where \( P_{\text{irr}} \), \( P_{\text{ann}} \), \( P_{\text{unirr}} \) are the concerning Charpy impact test parameters (TT, USV) in the unirradiated, the irradiated or the annealed state.

After 100 h annealing at 475° C the recovery parameter R is shown in Fig. 4. Both "over-recovery" and incomplete recovery can be observed. From the safety point of view the results are favourable: the heats with the highest irradiation sensitivity reveal complete recovery. One
Fig. 4: Recovery of Charpy V parameters

Fig. 5: Fracture toughness, $K_{Jc}$ - temperature curve for heat R3 from VVER-440 steel in the unirradiated (U) and irradiated (I) state
should notice that the transition temperature shift of heat 16 is hardly recovered by annealing. Heat 16 only differs from the other heats in the small grain size and the shorter tempering at lower temperature.

Fracture mechanics parameters were determined using the master curve concept [4]. The master curve is a commonly valid fracture toughness-temperature curve. The position on the temperature axis is defined by a material-dependent reference temperature $T_o$. Fig. 5 presents the fracture toughness-temperature curve (mean curve with 5 and 95% probability boundaries) for heat R3 in the unirradiated and irradiated state. Irradiation shifts the curve to higher temperature. The shift is larger than the transition temperature shift measured by Charpy impact tests. In other cases both characteristic temperature shifts are approximately comparable with each other. Predictive formula which describe the influence of the irradiation on fracture mechanics parameters for VVER steels have not yet been available. Nevertheless, the Charpy transition temperature shift seems to provide a useful estimation for the irradiation-induced changes of the fracture mechanics response, too.

4. Conclusion

For 7 heats from VVER 440 and 1000 reactor pressure vessel steels, the change of the Charpy impact parameters and the master curve reference temperature $T_o$ due to irradiation and annealing could be determined.

As a rule, the irradiation behaviour meets the prediction of the Russian guides in a conservative manner and is not critical from the safety point of view. However, there are surprising non-conservative results after both irradiation and annealing. Such outliers are suspect and we have not yet been able to explain the effect. Microstructural investigations are planned to clarify the abnormal behaviour.

References