Post-Irradiation Examination of the Radiation Annealing of the Embrittlement of the Reactor Pressure Vessel Steel

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Abstract. Influence of neutron irradiation on RPV steel degradation are examined with reference to the possible reasons of the substantial experimental data scatter and furthermore – nonstandard (non-monotonous) and oscillatory embrittlement behavior. In our glance this phenomenon may be explained by presence of the wavelike component in the embrittlement kinetics.

We suppose that the main factor affecting steel anomalous embrittlement is fast neutron intensity (dose rate or flux), flux effect manifestation depends on state-of-the-art fluence level. At low fluencies radiation degradation has to exceed normative value, then approaches to normative meaning and finally became sub normative. Data on radiation damage change including through the ex-service RPVs taking into account chemical factor, fast neutron fluence and neutron flux were obtained and analyzed.

In our opinion controversy in the estimation on neutron flux on radiation degradation impact may be explained by presence of the wavelike component in the embrittlement kinetics. Therefore flux effect manifestation depends on fluence level. At low fluencies radiation degradation has to exceed normative value, then approaches to normative meaning and finally became sub normative.

Moreover as a hypothesis we suppose that at some stages of irradiation damaged metal have to be partially restored by irradiation i.e. neutron bombardment. Nascent during irradiation structure undergo occurring once or periodically transformation in a direction both degradation and recovery of the initial properties. According to our hypothesis at some stage(s) of metal structure degradation neutron bombardment became recovering factor. As a result oscillation arise that in tern lead to enhanced data scatter.

1. Introduction
As a main barrier against radioactivity outlet reactor pressure vessel (RPV) is a key and irreplaceable component in terms of safety and extended LWR plant life. For this reason, it is important for RPV materials degradation forecasting to be as accurate as possible.

The surveillance programmes call upon to predict ahead RPV materials characteristics conservatively to guarantee RPV structural integrity without any compromise. But for any length of time results of the RPV steels behavior under neutron irradiation study are characterized by high data scatter [1].

Despite the great amount of experimental results coming from surveillance programmes and research fields, the comprehension of this phenomenon still remain unanswered.
Applicability of dose-damage relations to operating reactors thoroughly analyzed [2]. The overall effect of the all the input uncertainties (in the measurement of TTS, composition and exposure conditions) on the development of the dose-damage relations and their ability to predict data is illustrated in Figure 1.

![Figure 1. Dependence of the range of the residuals for different dose-damage relations on the fast neutron fluence.](image)

2. Prerequisites to the substantiation of the possible (potential) RPV steel embrittlement data scatter sources

Among unaccounted factors dose rate (neutron flux) is the most enigmatic. For example, it was established that irradiation at low flux enhanced embrittlement of the steel with high or medium copper contamination [3]. Moreover flux effect can lead to the alteration of the standard monotonous dose-damage dependence to the non-monotonous one [4].

2.1. Background.

The changing condition of the RPV steel during service is predicted by dose-damage relationship also known as embrittlement curve. The general dependence of the temperature transition shift (TTS) on radiation embrittlement coefficient $A_F$ is as follows:

$$TTS = A_F \times F^n$$  \hspace{1cm} (1),

where $A_F$ is the radiation embrittlement coefficient and $F$ is the fast ($E \geq 0.5$ MeV) neutron fluence in units of $10^{18} \text{sm}^{-2}$ (fluence factor), $n$ – coefficient (~1/3).

For Russian RPV Cr-Mo-V base steel $A_F = A \times (P + 0.07Cu)$, where $A_F$ – coefficient (800 at 270°C), $P$ and $Cu$ – are the weight concentrations of these elements.

Along with routine investigations in Russia systematic research on actual radiation embrittlement and flux effect manifestation of the decommissioned PWR pressure vessels via through samples (trepans) has been carried out.

The earliest commercial PWR prototype unit Novovoronezh-1 (NV1) RPV after 20 years of operation was trepanned in 1987. Then Novovoronezh-2 (NV2), the oldest PWR type experimental reactor-prototype ERP and, finely, nuclear icebreaker NIB «Lenin» RPVs also were trepanned. The most interesting and unexpected data were discovered during trepans of the first nuclear ship – icebreaker «Lenin» investigation.

Chemical analyses of the icebreaker RPV material were carried out with FSQ «Baird» optical emission spectrometer (Table 1).
Table 1. Chemical composition of the icebreaker RPV materials under study (wt. %).  

<table>
<thead>
<tr>
<th>Material</th>
<th>C</th>
<th>Mn</th>
<th>Si</th>
<th>P</th>
<th>Cu</th>
<th>Mo</th>
<th>Ni</th>
<th>Cr</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weld</td>
<td>0.05</td>
<td>1.03</td>
<td>0.41</td>
<td>0.035</td>
<td>0.15</td>
<td>0.49</td>
<td>0.17</td>
<td>1.39</td>
<td>0.15</td>
</tr>
<tr>
<td>Base</td>
<td>0.17</td>
<td>0.45</td>
<td>0.28</td>
<td>0.018</td>
<td>0.09</td>
<td>0.67</td>
<td>0.35</td>
<td>2.75</td>
<td>0.09</td>
</tr>
</tbody>
</table>

Fast (E≥0.5MeV) neutron fluence evaluation was based on the specific Mn-54 and Nb-93m activities of the vessels steel and on the Nb-93m activity of the RPV cladding.

The RPV materials radiation degradation (embrittlement) was determined by finding the ductile-to-brittle transition temperature shift (TTS). RKP-300 impact pendulum test machine for standard Charpy specimens testing was used.

2.2. Test results

The unexpected results of the icebreaker RPV weld and base metal studying are given in Figure 2, where one can recognize that the actual (measured) radiation embrittlement coefficients of the trepan materials for the periphery (remote) zone of the vessel are significantly higher that for the inner part. One can see also that hardness measurement and Charpy impact testing results agree.

![Figure 2. Radiation embrittlement coefficient $A_F$ and hardness value HB through the icebreaker RPV wall distribution. TT – transition temperature, TT$_0$ – transition temperature in initial state. Note: HB=252[189kgf/mm$^2$] - hardness values at as-received condition|after annealing 650°C/2h.](image)

3. Data analysis and discussion

First impression from foregoing decommissioned PWR pressure vessel material properties study – enhanced degradation rate at low neutron fluxes. Examples for icebreaker base metal and weld metal are demonstrated in Figures 3,4. Registered facts denote that known as «flux effect» factor was in action. Unexpected circumstance however is reduced embrittlement zone appearance that follows after of previous part of enhanced embrittlement. It is seen by means of TTS on neutron flux and fluence dependencies observation.
In support of the unusual data gained, we started «flux effect» purposeful study using surveillance specimens from WWER-440/213 RPV. Taking into account the flux level that is the irradiation position of the selected standard Charpy-type surveillance specimens additional sub size specimens were manufactured and tested. Analysis shows that 3 fold difference in flux level may lead to evident distinction in terms of TTS:~60°C at neutron fluence of $4 \times 10^{19}$ cm$^{-2}$ and copper content 0.1% mass.[5].

Flux effect (FE) depends on Cu content in the steel: the lower Cu concentration the lower FE. However super cleaning of the metal up to less than 0.05 wt.% Cu diminishes FE but does not reduce data scatter [6].

It is seen from Figures 3,4 that alteration of the standard monotonous dose-damage dependence to the non-monotonous one is possible. Search of similar far from trivial effect brings to example where it characterized by authors as «quite atypical» [7]. For the sake of correctness, it is necessary to underline that the first mention concerning distinction between test reactors and low-lead-factor (surveillance) data had appeared as early as 1980 [8].

Careful surveillance programs database analysis shows that damage-dose relationship can contain oscillating component. Figure 5 shows TTS-fluence dependence for RPV materials with $A_F=15-24°C$:

![Figure 3. Comparison of the TTSs between «remote» and «inside» icebreaker RPV base metal specimens.](image1)

![Figure 4. Comparison of the TTSs between «remote» and «inside» icebreaker RPV weld metal specimens.](image2)

![Figure 5. TTS-fluence dependence for RPV steels with $A_F=15-24°C$:](image3)

- ● – weld metal (flux $4 \times 10^{11}$ sm$^{-2}$ s$^{-1}$);
- ○ – base metal (flux $30 \times 10^{11}$ sm$^{-2}$ s$^{-1}$);
- - - - Regulatory guide.
With surprising we have seen the serrated-type curve and independence on neutron flux and material origin (base or weld metal). It is necessary to remember that there is an example when contrary to the famous radiation embrittlement in metals neutron irradiation at some range of fast neutron doses improves both the strength and ductility of steel [9].

The very wonderful fact is the effect of the temporary weakening of the embrittlement appearance. Possible comprehensible explanation is as follows: the radiation-induced copper-rich precipitates nature (dimensions and concentration) alteration. Evidently, we have fixed phenomenon when neutron irradiation in some range of low doses improves the mechanical properties of the steel. In this case we have to consider irradiation temporarily as a recovery factor.

**Conclusion**

Influence of neutron irradiation on RPV steel degradation are examined with reference to the possible reasons of the substantial experimental data scatter and furthermore – nonstandard (non-monotonous) and oscillatory embrittlement behavior. In our glance this phenomenon may be explained by presence of the wavelike component in the embrittlement kinetics. As a result of dose rate effect manifestation peripheral RPV’s zones in some range of fluencies have to be damaged to a large extent than situated closely to core. This finding recently was confirmed [10].

As a hypothesis we suppose that at some stages of irradiation damaged metal have to be partially restored by irradiation i.e. neutron bombardment. In this case we have to consider that provisionally irradiation became a recovering factor. As a result oscillation arises that in turn lead to enhanced data scatter.

**References**