Brittle fracture resistance of reactor pressure vessel steels in the initial state*

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Abstract

The authors investigate the influence of chemical and structural inhomogeneity on the brittle fracture resistance (BFR) of VVER vessel materials in the initial state (without irradiation). It is proposed to replace the brittle fracture resistance assessment using the critical brittleness temperature $T_C$ for the BFR assessment using the brittle-viscous transition temperature $T_T$. Consideration was given to calibration charts used for studying the $T_T$ dependence on the grain size and heat treatment.

A comparison of the $T_C$ and $T_T$ values in the experimental industrial 15H2NMFA-A steel billets shows that the $T_C$ values are significantly lower than the $T_T$ values:

– at the lower level of conservatism, the difference between $T_C$ and $T_T$ is 22 °C;
– at the upper level of conservatism, this difference is 24 °C.

The array data on the critical brittleness temperature and the ductile-to-brittle transition temperature of impact test samples of 15H2NMFA (for VVER-1000) and 15H2NMFA grade 1 (for VVER-1200) steels were statistically processed. The industrial shell samples were manufactured at the “Energomashspetsstal” plant (Kramatorsk, Ukraine).

It was found that, in the metal of VVER-1000 vessel surveillance specimens with the copper content

– less than 0.06%, heat treatment has a significant effect on the $T_C$ value, which changes from −99 to −28°C;
– from 0.07 to 0.12%, heat treatment has a significant effect on the $T_T$ value, which changes from −60 to −40°C.

Keywords

Brittle fracture resistance assessment; critical brittleness temperature; ductile-to-brittle transition temperature; standard deviation; conservative estimates of the VVER-1000 vessel life.

Radiation embrittlement of VVER vessel materials is determined by standard shifts in the critical brittleness temperature $\Delta T_c$ (Tab. 1) (see PNAE G-7-008-89 (1987) with refinements in RD EO 1.1.2.09.0789-2012 (2012) and RD EO 1.1.2.99.0920-2013 (2013)).

The normative conservative margin in defining $\Delta T_c$ ($\delta T_c = 38 \, ^\circ C$) for the base metal of 15H2NMFAA and 15H2NMFA grade 1 steels at a fluence up to $100 \times 10^{22} m^{-2}$ is several times higher than the difference of normative $\Delta T_c$ for these steels not exceeding 10 °C (see Fig. 1), which does not correspond to the assumption that the metal chemical inhomogeneity has a significant effect on $\delta T_c$ (Vishkarev et al. (1980a)).

The BFR of vessel materials, $T_c = T_{c0} + \Delta T_c$, is determined by the non-additive summation of normative guaranteed shifts in the critical brittleness temperature with the normative guaranteed critical brittleness temperature of the metal in the initial state, $T_{c0}$ (TU 0893-013-00212179-2003 (2003)) of 15H2NMFA grade 1 steel and characterized by an overestimated conservative margin, $\delta T_c$ (Dub et al. 2016).

In determining $T_{c0}$ in the control sets of surveillance specimens of the VVER-1000 base metal, corresponding to 15H2NMFA grade 1 steel (TU 0893-013-00212179-2003 (2003)), the upper conservative value $T_{c0}^{\text{max}} = –45 \, ^\circ C$ at $\delta T_c = 38 \, ^\circ C$; the lower conservative value $T_{c0}^{\text{min}} = –83 \, ^\circ C$ at $\delta T_c = 0 \, ^\circ C$.

Therefore, with additive summation, the normative $T_{c0} = –83 \, ^\circ C$ corresponds to the actual value.

### Determining the normative critical brittleness temperature in 15H2NMFAA steel by methods


A comparison of the $T_c$ and $T_{c0}$ values of the experimental material of industrial 15H2NMFAA steel billets shows that the $T_c$ values are significantly lower than the $T_{c0}$ values. The critical brittleness temperature, $T_c$, is a normative criterion for determining the BFR which is widely used in calculations; therefore, the application of the $T_c$ criterion is of considerable interest.

The critical brittleness temperature, $T_c$, is defined as the temperature of the intersection of the temperature-impact strength curve with the level of determining the transition temperature by the steel yield strength PNAE G-7-008-89 (1987).

The transition temperature, $T_c$, characterizes the steep rise position in the curve of the temperature dependence of the absorbed energy (GOST R ISO 148-1-2013 (2014)). The test conditions are as specified in (GOST R ISO 148-1-2013 (2014) and DIN EN ISO 148-1-2011 (2010)). The temperature dependence of the absorbed energy is established by constructing a smooth curve over individual points. A specific value of the absorbed energy is obtained as a percentage of the value corresponding to the upper area $K_V$, for example, 50%.
The main difference between the methods (PNAE G-7-008-89 (1987)) and (GOST R ISO 148-1-2013 (2014)) consists in the detection levels of the transition temperature, $T_T$, and the critical brittleness temperature, $T_C$, relative to the level of the upper area $KV$ and $KCV$:

- for $T_T$, the transition temperature detection level is 50% relative to the level of the upper area $KV$;
- for $T_C$, the transition temperature detection level, for example, is 26.2% relative to the level of the upper area $KCV$ at the yield point of 15H2NMFAA steel = 490 MPa, which is accompanied by a shift to lower temperatures.

### Studies of $T_C$ and $T_T$ in experimental industrial billets made of 15H2NMFAA steel

The experimental-standard VVER-1000 core shell made of experimental-industrial 15H2NMFAA steel billets was manufactured at the “Energomashspetsstal” plant by electric-arc melting with vacuum casting (forging 14308, melting Nos. 37356 and 17501); it contained 1.31% Ni, 0.005% P and 0.04% Cu; its impurity content practically corresponded to 15H2NMFA grade 1 steel (Tab. 2).

The experiments were aimed at determining the critical brittleness temperature, $T_C$, and estimating errors in determining, $T_C$, depending on the number of samples tested and the test pattern Kazantsev et al. (2015) and GOST 9454-78 (1994). To this end, a statistical analysis of 1120 results of impact-bending tests was carried out (see Tab. 2) (Dub et al. 2011a; Dub et al. 2016, Vishkarev et al. 1980). It was assumed that the lower conservative level of the experimental value is its arithmetic average $A$ and the upper one is the sum $A + s_{ST}$, where $s_{ST}$ is the standard deviation (Dub et al. (2016), Dub et al. (2011b)).

A comparison of the calculated data confirms that the main difference between the methods is in the different levels of $KV$ and $KCV$ in determining the transition temperature, $T_T$, and the critical brittleness temperature, $T_C$, with the difference $T_T - T_C$ reaching 22 °C and 24 °C at the lower and upper conservative levels, respectively (Tab. 2).

It seems statistically justified to use the transition temperature, $T_T$, to determine the brittle fracture resistance of VVER vessel materials, since $T_T$ is located at the center of the normal probability distribution $KV$, depending on the temperature of impact-bending test samples (Fig. 2).

The dependence of the absorbed energy $KV$ on the temperature of testing industrial 15H2NMFAA steel billets at the lower and upper conservative levels of $T_T$ was obtained by constructing a smooth curve completely corresponding to the normal standard distribution over separate points (Tab. 2a) with the inflection point in $T_T$. At the upper conservative level of $T_T$, the smooth curve reflects the spread of points relative to the normal standard distribution (Fig. 2b) with the inflection point in $T_T$, taking into account the standard deviation: $s_{ST} = 12$ °C.

### Study of $T_T$ in the experimental calibration metal of 15H2NMFAA steel of the basic chemical composition

The VVER-1000 and VVER-1200 core shells made of industrial 15H2NMFAA and 15H2NMFA grade 1 steel billets are subjected to the final heat treatment according to the conditions developed in the JSC RPA “CNIITMASH” (Markov (2011)).

The weld fusion boundary between the base metal and the weld seam material after applying the anticorrosive cladding is not subjected to the final heat treatment by...
appropriate modes, but is heat treated according to technological tempering conditions.

The obtained billets were heat treated according to the following conditions providing a certain structure and grain size of the metal (GOST 5639-82 (1982)):

- the weld seam metal and the base metal of the fusion boundary after technological tempering have a tempered cast structure with a grain size of 90–120 μm (hereinafter “isothermal annealing”);
- the metal after standard heat treatment in basic mode (normalizing, quenching, tempering) has an improved structure with a grain size of 40–60 μm (hereinafter “quenching”);
- the metal after heat treatment in conditions including quenching combined with isothermal annealing and subsequent tempering has a structure with additional perlite-type phase recrystallization, effectively grinding the inherent austenite grain to a level of 15–25 μm (hereinafter “tempering”).

The experimental sets of small-sized impact specimens were made in accordance with (GOST R ISO 148-1-2013 (2014)) from the laboratory metal by LCR-welding of industrial billets, using a non-consumable tungsten electrode with argon as a shielding gas (Surkov et al. (1979), Anosov et al. (1982), Anosov et al. (1985), Anosov et al. (1990)).

The metal of industrial shells of 15H2NMFA steel (15H2NMFA grade 1 of the basic chemical composition), melting No. 132009, containing 1.16% Ni, 0.0025% P and 0.025% Cu with given structural changes, smelted on a high-quality pure charge, was used for the billets (Dub et al. (2012), Dub et al. (2011b)).

To study the heat treatment effects on the BFR of the local continuous remelting (LCR) laboratory metal, immediately after remelting by welding, the metal is subjected to high tempering, and experimental sets of small-sized impact specimens are manufactured which are notionally named “tempering” or “zero heat treatment level” of the metal.

The final heat treatment of the VVER vessel core shells is determined by the corresponding modes:

- “quenching” for 15H2NMFAA steel of VVER-1000 vessels;
- “isothermal annealing” for 15H2NMFAA and 15H2NMFA grade 1 steels of VVER-1200 vessels.

In the course of the study, calibration curves were constructed that reflect the $T_p$ dependence on the structure (grain size) corresponding to different heat treatments. To confirm the $T_p$ dependence on the grain size for the LCR metal structures of 15H2NMFA grade 1 steel of the basic chemical composition, it is necessary to use the results of testing industrial billets of standard VVER-1200 vessel shells subjected to “isothermal annealing” and the metal of surveillance specimens of industrial VVER-1000 vessel shells subjected to “quenching” (Dub et al. (2011b), Dub et al. (2012)).

<table>
<thead>
<tr>
<th>Heat treatment conditions</th>
<th>Content of elements, %</th>
<th>GOST R ISO 148-1-2013 Conservative level: upper/lower</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>P</td>
<td>Cu</td>
</tr>
<tr>
<td>“Tempering”</td>
<td>1.16</td>
<td>0.0025</td>
</tr>
<tr>
<td>“Hardening”</td>
<td>1.16</td>
<td>0.0025</td>
</tr>
<tr>
<td>“Isothermal annealing”</td>
<td>1.16</td>
<td>0.0025</td>
</tr>
</tbody>
</table>

Study of $T_p$ in the industrial steel billets of VVER vessel shells

The standard upper and lower nozzle shells of 15X2NMFAA steel and the lower (supporting) and upper core shells of 15H2NMFA grade 1 steel of the VVER-1200 reactor, and the elongated core shell of the VVER-TOI reactor of 15H2NMFA grade 1 steel are manufactured at the “Energomashspetstal” plant. The vessel equipment components were subjected to “isothermal annealing”. Tables 4–6 provide basic information on the shells.

Calibration of industrial billets made of 15H2NMFA grade 1 steel of the basic chemical composition

An increase in the phosphorus content from 0.0025 to 0.0050% in the industrial billets of the VVER-1200 vessel standard shells, subjected to “isothermal annealing”, is accompanied by a rise of $T_p$ to the level corresponding to the grain size of “quenching” mode for 15H2NMFA grade 1 steel of the basic chemical composition of (Fig. 3), which confirms the negative effect of phosphorus on the properties of grain boundaries.

At the same time, the metal of the core supporting shell and elongated shell ingot top end, subjected to “isothermal annealing” and corresponding to the technical standards in (TU 0893-013-00212179-2003 (2003)), are characterized by $T_p$ above the level corresponding to the grain size of “quenching” mode for 15H2NMFA grade 1 steel of the basic chemical composition.

This metal, when the VVER-1200 vessel is assembled, is located in the upper part of the core, which is sufficiently far from the radiation source, guaranteeing safe operation.
Table 4. Results of determining $T_T$ in the VVER-1200 vessel standard nozzle shells made of 15H2NMFAA steel

<table>
<thead>
<tr>
<th>Material (melting No., shell)</th>
<th>Content of elements, %</th>
<th>GOST R ISO 148-1-2013 Conservative level: upper/lower $K_{ inv }, J/cm^2$</th>
<th>$T_T$, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>15-12003, lower, nozzle area</td>
<td>Ni: 1.34, P: 0.005, Cu: 0.04</td>
<td>327 / 336</td>
<td>–64 / –65</td>
</tr>
<tr>
<td>15-12001, upper, nozzle area</td>
<td>Ni: 1.34, P: 0.005, Cu: 0.04</td>
<td>294 / 306</td>
<td>–69 / –70</td>
</tr>
</tbody>
</table>

Table 5. Results of determining $T_T$ in the VVER-1200 vessel standard core shells made of 15H2NMFA grade 1 steel

<table>
<thead>
<tr>
<th>Material (melting No., shell)</th>
<th>Content of elements, %</th>
<th>GOST R ISO 148-1-2013 Conservative level: upper/lower $K_{ inv }, J/cm^2$</th>
<th>$T_T$, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>15-1056, Lower, Core</td>
<td>Ni: 1.08, P: 0.005, Cu: 0.02</td>
<td>263 / 291</td>
<td>–65 / –65</td>
</tr>
<tr>
<td>15-1073, Upper, Core</td>
<td>Ni: 1.10, P: 0.005, Cu: 0.02</td>
<td>317 / 321</td>
<td>–75 / –76</td>
</tr>
<tr>
<td>15-1062, Supporting, Core</td>
<td>Ni: 1.08, P: 0.005, Cu: 0.01</td>
<td>291 / 307</td>
<td>–48 / –48</td>
</tr>
</tbody>
</table>

Table 6. Results of determining $T_T$ in the ends of the VVER-1000 vessel standard core elongated shell made of 15H2NMFA grade 1 steel

<table>
<thead>
<tr>
<th>Material (melting No., shell)</th>
<th>Content of elements, %</th>
<th>GOST R ISO 148-1-2013 Conservative level upper/lower $K_{ inv }, J/cm^2$</th>
<th>$T_T$, °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>15-13247, elongated, core, ingot top end</td>
<td>Ni: 1.08, P: 0.005, Cu: 0.04</td>
<td>267 / 297</td>
<td>–47 / –47</td>
</tr>
<tr>
<td>15-13247, elongated, core, ingot bottom end</td>
<td>Ni: 1.08, P: 0.005, Cu: 0.04</td>
<td>356 / 369</td>
<td>–75 / –76</td>
</tr>
</tbody>
</table>

Figure 3. Calibration chart of $T_T$ depending on the metal structure (grain size) of the VVER-1200 vessels made of 15H2NMFA grade 1 steel, corresponding to “tempering”, “quenching” and “isothermal annealing” modes for different billets

**Calibration of the $T_T$ dependence on the metal grain size of 15H2NMFAA steel in VVER-1000 vessel shell surveillance specimens**

An increase in the nickel content from 1.00 to 1.35%, P from 0.0025 to 0.0100%, and Cu from 0.025 to 0.12% in the VVER-1000 vessel witness sample billets, subjected to “quenching” mode, is accompanied (see Fig. 3) by:

Figure 4. Effects of the copper content on $T_T$ in the VVER-1000 vessel surveillance specimens of 15H2NMFAA steel subjected to “quenching” mode and corresponding to “tempering, “quenching” and “isothermal annealing” modes of the LCR metal of 15H2NMFA grade 1 steel of the basic chemical composition at the conservative level: a) upper; b) lower
− a rise of $T_a$ above the level corresponding to “tempering” mode for 15H2NMFA grade 1 steel of the basic chemical composition;
− a fall of $T_a$ below the level corresponding to “quenching” mode for 15H2NMFA grade 1 steel of the basic chemical composition;

The effects of the copper content on $T_a$ is determined when the metal structure of the VVER-1000 vessel surveillance specimens (“tempering, quenching” and “isothermal annealing” modes) corresponds to the LCR metal structure of 15H2NMFA grade 1 steel of the basic chemical composition (Fig. 4).

In the metal of VVER-1000 vessel surveillance specimens with the copper content
− less than 0.06%, heat treatment has a significant effect on the $T_a$ value, which changes from −99 to −28°C;
− from 0.07 to 1.2%, heat treatment has a significant effect on the $T_a$ value, which changes from −60 to −40°C.
− the $T_a$ value is practically independent of the level of conservatism (see Fig. 4).

Conclusion

1. A comparison of the $T_a$ and $T_a$ values in the experimental metal of industrial 15H2NMFAA steel billets (see Tab. 2) shows that the $T_a$ values are significantly lower than the $T_a$ values:
   − at the lower level of conservatism, the difference between $T_a$ and $T_a$ is 22 °C;
   − at the upper level of conservatism, this difference is 24 °C.

2. Significant differences in the results are related to the determination of $T_a$ directly at the inflection point of the normal standard probability distribution and $T_a$ at the point located much lower; therefore, it seems statistically reasonable to use the transition temperature, $T_a$, in determining the brittle fracture resistance of VVER vessel materials.

3. The calibration chart reflecting the $T_a$ dependence on the structure (grain size) of the LCR metal of 15H2NMFA grade 1 steel of the basic chemical composition, corresponding to “tempering”, “quenching” and “isothermal annealing” modes, should be used for confirming the results of testing standard VVER vessel shells.

4. An increase in the phosphorus content from 0.0025 to 0.0050% in the metal of industrial billets of VVER-1200 vessel standard shells, subjected to “isothermal annealing”, is accompanied by a rise of $T_a$ to the level corresponding to the grain size of “quenching” mode of the LCR metal of 15H2NMFA grade 1 steel of the basic chemical composition (Fig. 3), which confirms the negative effect of phosphorus on the properties of grain boundaries.

5. An increase in the nickel content from 1.00 to 1.35, phosphorus from 0.0025 to 0.0100%, and copper from 0.025 to 0.12% in the VVER-1000 vessel surveillance specimen billets, subjected to “quenching”, is accompanied (see Fig. 3) by:
   − a rise of $T_a$ above the level corresponding to “tempering” mode for 15H2NMFA grade 1 steel of the basic chemical composition;
   − a fall of $T_a$ below the level corresponding to “quenching” mode for 15H2NMFA grade 1 steel of the basic chemical composition;

6. In the metal of VVER-1000 vessel surveillance specimens with the copper content
   − less than 0.06%, heat treatment has a significant effect on the $T_a$ value, which changes from −99 to −28 °C;
   − from 0.07 to 1.2 %, heat treatment has a significant effect on the $T_a$ value, which changes from −60 to −40 °C.
   − the $T_a$ value is practically independent of the level of conservatism (see Fig. 4).

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