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IMPROVEMENT OF RADIATION EMBRITTLEMENT DEPENDENCES FOR VVER-1000 PRESSURE VESSEL MATERIALS ON SERVICE-LIFE EXTENSION

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The results received by now while investigating the sets of surveillance specimens for VVER-1000 reactor pressure vessels (RPV) are the most representative for studying and evaluation of radiation embrittlement of materials of this type of RPVs. It is primarily due to the fact that neutron irradiation of surveillance specimens (SS) of VVER-1000 RPV materials is carried out with a flux density corresponding to the inner surface of a reactor pressure vessel opposite to the core with the average lead factor for sets of surveillance specimens intended for evaluation of radiation embrittlement varying from 0.5 to 3.

On the base of SS research results, standard dependences have been developed and are currently in use for radiation embrittlement of VVER-1000 RPV materials with the operation range limited by the value of fast neutron fluence of 6.4×10^{23} neutron/m^2 with the energy above 0.5 MeV where the general transition temperature is modelled as a sum of radiation and thermal contributions and the thermal component has a form of a function with the maximum which is hypothetically caused by carbide precipitation and coagulation [1,2].

Microstructure investigations of SS materials performed at the National Research Centre "Kurchatov Institute" after different time exposures failed to prove the hypothesis of carbide hardening under temperature aging and, at the same time, unambiguously showed the existence of significant radiation- and temperature-stimulated phosphorus segregation on grain boundaries and in interphase areas [3-5]. Thereby, there arose a necessity to develop a dependence that would account for the physical processes proceeding in the material under irradiation at the reactor operation temperature.

Forecasting of the RPV material condition on service life extension up to 60 years and more requires an expansion of the validity range of the developed dependences to the maximum fast neutron fluence of ~ 8.0×10^{23} neutron/m^2. With this aim in view, the SS material was subjected to accelerated irradiation to gain higher fast neutron fluences and conservatism of the suggested dependences was verified on their extrapolation.

On using the accelerated irradiation results for verification of the forecast conservatism for the developed dependences, account was taken of the effects related to the difference in time and rate of accelerated irradiation as compared to the conditions of RPV wall irradiation.

EMBRITTLEMENT OF 15H2NMFA(A) STEEL AND ITS WELDS UNDER SIMULTANEOUS IMPACT OF NEUTRON IRRADIATION AND TEMPERATURE

Development of a physically reasonable dependence of radiation embrittlement requires understanding of the mechanisms responsible for degradation of properties in materials under irradiation.

The results received in [6-11] show that irradiation initiates the following changes in the fine structure of VVER-1000 RPV materials:

- Emergence and increase of the density of dislocation loops in the base metal (BM) and weld metal (WM) both within design values of a fluence and after achievement of the design values of a fast neutron fluence [6-8].
- Emergence and increase of the density of radiation-induced precipitates (enriched in nickel, manganese and silicon in case of WM, and in nickel and silicon in a case of BM) over the

complete range of fast neutron fluences with nearly invariable density of carbides and carbonitrides in BM and lack of their radiation-induced precipitations in WM [6-8].

Increase in the level of grain-boundary segregations of impurities (primarily phosphorus) and doping elements on high values of a fast neutrons fluence even at low levels of phosphorus concentration (below 0.006%). As a result of the investigation performed [9-11] it was found that an appreciable level of phosphorus segregation is observable on the boundaries of former austenitic grains even at low values of fast neutron fluences but with high exposure times.

Thus, the resulting effect of the transition temperature shift of the VVER-1000 RPV materials at the irradiation stages close to the design levels and higher (over 4.0×10^{23} neutron/m^2) is caused by a simultaneous action of mechanisms of radiation hardening (hardening mechanism) and the growth of impurity segregations on grain boundaries (non-hardening mechanism).

Micrographic investigations of temperature SS sets [3-5] prove that on exposure to operation temperatures for up to 200,000 hours, an embrittlement mechanism takes effect in the VVER-1000 RPV material related to phosphorus grain-boundary segregation. No structural changes which might have led to a yield strength change have been found in the material of the specimens under study.

As said above, a transition temperature shift (owing to a simultaneous impact of irradiation and high temperatures) is expressed in the operating standard dependence by a sum of irradiation and temperature components:

$$\Delta T_K(F, t) = \Delta T_F(F) + \Delta T_T(t), \tag{1}$$

As it became clear from the result of the performed microstructure investigations, the radiation component must reflect the changes in the material properties resulting from both hardening and non-hardening embrittlement mechanisms, and the thermal one only from non-hardening mechanisms. Then, in this case it is more physically reasonable to represent the total transition temperature shift resulting from a simultaneous impact of irradiation and temperature in the form of a sum of hardening and non-hardening components:

$$\Delta T_K(F, t) = \Delta T_{hard}(F) + \Delta T_{non\text{-}hard}(t), \tag{2}$$

rather than a sum of the radiation and thermal components.

The non-hardening component in dependence (2) is a result of the processes caused both by irradiation and temperature. The final result of these processes depends on an integrated dose of fast neutrons as well as on the exposure time in the reactor. The integrated dose of fast neutrons is related to time via the flux. Because the variation of a fast neutron flux in the existing SS database is no more than ~ 3 times between the minimal and the maximal values which corresponds to a flux variation on a reactor pressure vessel wall, the allowance for the neutron effect in the non-hardening part only via time is correct. The hardening component is related to the accumulated dose of fast neutrons. Therefore, it is correct to consider the additivity of contributions from non-hardening and hardening components.

The non-hardening component in dependence (2) is related to the formation of thermally- and radiation-stimulated phosphorus segregation on grain boundaries and interphases. Ref. [12] proves that the kinetics of thermally-stimulated phosphorus accumulation in steels of this type can be adequately described within the theory of solid-phase chemical reactions of the first type in the form:

$$\Delta C_B^P(t) = B + (C - B)\cdot(1 - \exp(-Ft)) \tag{3}$$

where: $\Delta C_B^P(t)$ is phosphorus concentration of on the boundary at instant *t*;

$B = \Delta C_B^P(0)$ is phosphorus concentration on grain boundaries at instant $t = 0$;

$C = \Delta C_B^P(\infty)$ is phosphorus equilibrium concentration on the grains boundary for $t \to \infty$;

t is time in hours;

$F = \gamma \cdot \frac{D}{d^2}$, to within the accuracy of numerical multiplier, $\gamma \approx 1$,

D is a phosphorus diffusion coefficient on the grain boundary;

d is an average distance to the boundary, i.e. a magnitude of the order of grain size.

The regression data analysis with the Auger-electron spectroscopy of the material of temperature sets of VVER-1000 RPV WM SS brought the value of coefficient F in equation (3) to equal 1.1×10^{-5} s^{-1}.

For the description of a thermally stimulated change of the transition temperature via non-hardening mechanisms, bearing in mind that:

1) the increase of $\Delta T_{non\text{-}hard}$ is related to the growth of a phosphorus grain-boundary segregation with time as described by equation (3) [3,5,10,13];
2) nickel concentration in metal correlates with the transition temperature variation rate [14];
3) the analysis of equation (1.3) shows that the increase in the phosphorus initial concentration on grain boundaries leads to a decrease in the phosphorus accumulation rate on the boundaries in the course of temperature exposures, i.e. the initial state effects the embrittlement rate, and the value of T_{k0} can be taken as a measure of the initial state [13, 14],

it is suggested to use equation (4)**:**

$$\Delta T_T = A\times f\,(C_{Ni}\,\cdot, T_{k0}) \times (1\text{-}\exp\times(F\cdot t))^n \qquad (4)$$

where the following parameters are used in the capacity of explaining parameters: time (t), nickel content (C_{Ni}) and T_{k0}, A and n are adjustable coefficients, and the value of coefficient F is taken equal to 1.1×10^{-5} s^{-1} similarly to equation (3).

The phosphorus concentration is not included into explaining parameters for its content in a database of WM with the nickel concentration of more than 1.3% varies within rather narrow limits from 0.006 to 0.009%.

A regression analysis of the temperature SS database gave a dependence of transition temperature via thermal aging for the exposure temperature of 310-320 °C [13]:

$$\Delta T_T = 1.3\cdot C_{Ni}^{4}\times\exp(-0.02\cdot T_{k0})\times(1-\exp\times(-1.1\times10^{-5}\cdot t))^{0.6} \quad \sigma=9\ (^{o}C) \qquad (5)$$

where $1.08\% < C_{Ni} < 1.89\%$.

A regression analysis of Auger-electron spectroscopy data for a metal of irradiated SS sets of VVER-1000 RPV WM made it possible to estimate the radiation-stimulated diffusion coefficient for phosphorus at 290 °C for a fast neutron flux corresponding to a flux onto a vessel wall. The value of F in equation (3) for the irradiated weld metal has been obtained which is nearly twice as large as the estimate made in the course of investigation of specimens of temperature sets of weld metal at the temperature of 310-320 °C.

As shown in ref. [15], the functional dependence of the phosphorus accumulation kinetics on thermally stimulated segregation is also applicable for an assessment in case of radiation stimulated segregation with the use of diffusion parameters obtained on the basis of experimental results. Hence, the transition temperature dependence for VVER-1000 RPV WM on account of implementation of embrittlement via the non-hardening mechanism for 290 °C on irradiation with regard for equation (5) and a change in the diffusion constant can be written as follows:

$$\Delta T_{non\text{-}hard}=1.3\cdot \mathbf{C_{Ni}}^{4}\times \exp(-0.02\cdot T_{k0})\times(1-\exp\times(-2.2\cdot 10^{-5}\cdot t))^{0.6}\ (^{o}C), \qquad (6)$$

where $1.08\% < C_{Ni} < 1.89\%$.

The hardening component of the transition temperature of steel on irradiation (ΔT_{hard}) was defined as a difference between the experimentally determined by SS testing total transition temperature of a material after long-time irradiation and the non-hardening part value corresponding to the irradiation time as calculated using equation (6).

Thus, the hardening part of the transition temperature for the irradiated SS was calculated from formula:

$$\Delta T_{hard}(F\ ) = \Delta T_{K}(F,t) - \Delta T_{non\text{-}hard}(t) \qquad (7)$$

A model depending on the Ni and Mn content in the material which is confirmed by results of microstructure investigations [2,6-9,14,16,17] was chosen as a regression model for the description of the hardening component in equation (7). The following dependence was suggested in the capacity of a simplified approximation:

$$\Delta T_{hard} = B^{OC}\cdot C_{N}i\cdot C_{Mn}\cdot F^{0.8} \qquad (8)$$

where C_{Ni} is nickel concentration in the metal, weight%, $1.08\% C_{Ni} < 1.89\%$,
C_{Mn} is manganese concentration in the metal, weight%, $0.40\% \le C_{Mn} \le 1.10\%$,
F is a fast neutron fluence with $E \ge 0.5$ MeV in terms of 10^{22} neutron/m^2,
B^{OC} is an adjustable coefficient.

Statistical processing of the database of testing results of irradiated surveillance specimens of VVER-1000 RPV WM gives the following values for regression equation (8):

$$\Delta T_{hard}=1.48\cdot C_{Ni}\cdot C_{Mn}\cdot F^{0.8} \qquad \sigma = 12.4\ ^{\circ}C \qquad (9)$$

Then the total transition temperature shift of VVER-1000 RPV WM on irradiation is described by dependence:

$$\Delta T_{K}(F,\ t)= \Delta T_{hard}\ (F)+ \Delta T_{non\text{-}hard}\ (t)\ , \qquad (10)$$

where $\Delta T_{non\text{-}hard}\ (t)=1.3\cdot C_{Ni}^{4}\times \exp(-0.02\cdot T_{k0})\times(1-\exp\times(-2{,}2\cdot 10^{-5}\cdot t))^{0.6}$
$\Delta T_{hard}\ (F)=1.48\cdot C_{Ni}\cdot C_{Mn}\cdot F^{0.8}$, $\sigma=12.4\ ^{o}C$.

The conservative estimate of the transition temperature shift of VVER-1000 RPV WM on irradiation is described by dependence:

$$\Delta T_{K}(F,\ t)= \Delta T_{hard}\ (F)+ \Delta T_{non\text{-}hard}\ (t)+2\ \sigma, \qquad (11)$$

where $\Delta T_{non\text{-}hard}\ (t)=1.3\cdot C_{Ni}^{4}\times \exp(-0{,}02\cdot T_{k0})\times(1-\exp\times(-2.2\cdot 10^{-5}\cdot t))^{0.6}$
$\Delta T_{hard}\ (F)=1.48\cdot C_{Ni}\cdot C_{Mn}\cdot F^{0.8}$, $\sigma=12.4\ ^{o}C$.

An existing database of VVER-1000 WM surveillance specimens was used to derive dependence (11); this database is limited with regard to a fast neutron fluence and exposure time of specimens under irradiation.

To test applicability of dependence (11) to the values of fast neutron fluence of ~8.0×10^{23} neutron/m^2, the specimens were subjected to accelerated irradiation (50-500 times faster than VVER-1000 reactor pressure vessel wall irradiation) in the IR-8 research reactor from the initial state and after long-term exposure as part of temperature sets of SS.

The data array analysis by the results of accelerated radiation of specimens from the initial state and after temperature exposure as part of temperature sets of SS has shown that this array can be described by a uniform dependence of the form $\Delta T_K^{accel}=B^{accel}\, C_{Ni}\, C_{Mn}\, F^{0.8}$ [18].

In the fast neutron fluence ranges under consideration and times of accelerated irradiation at 290 °C it is possible to neglect the contribution from the non-hardening mechanism to the transition temperature which was confirmed by microstructure investigations [14,19]. Then the procedure of using the results of accelerated irradiations for expandability of the application range of dependence (11) can be presented in the form [18]:

$$\Delta T_K^{RPV}(F,t)=\Delta T_K^{SS}(F,t)=\beta\cdot\Delta T_K^{accel}(F)+\Delta T_K^{non-hard}(t) \qquad (12)$$

where β is a correction factor taking into account an underestimation of the transition temperature “hardening” component received on accelerated irradiation.

When carrying out the statistical processing of databases on embrittlement of WM SS and research programs, the coefficient received is β =1.04.

A comparison of values calculated with dependence (10) and experimental values of the VVER-1000 RPV WM database with additional data of accelerated irradiation adjusted with formula (12) is given in fig. 1.

The comparison of experimental data and dependence (10) brings us to a conclusion of applicability of dependence (11) for conservative estimation of variation of VVER-1000 RPV WM properties on irradiation and a feasibility of spreading its action to the values of a fast neutron fluence of 8.0×10^{23} neutron/m^2 at a confidence level of 95%.

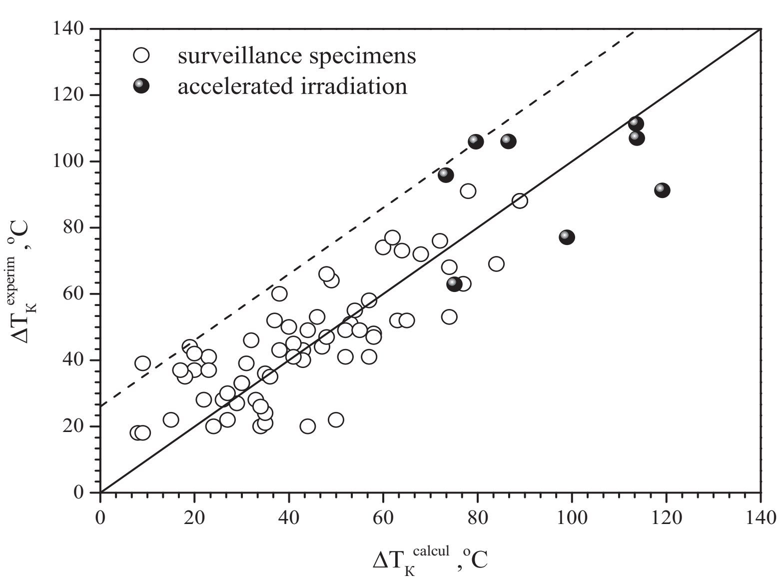

Figure 1. Experimental values of the transition temperature *vs.* values calculated from dependence (10)

No temperature aging effects have been so far found for VVER-1000 RPV BM at 320 °C with exposure time up to 200,000 hours [3-5]. Therefore, on further processing of the available irradiated SS arrayed data it was assumed that $\Delta T_{non\text{-}hard}(t)=0$.

The result of the database analysis of irradiated RPV VVER-1000 BM SS is an embrittlement dependence which conservatively describes the experimental results (see fig. 2):

$$\Delta T_K=7.24\ F^{0.43}+38\ (°C) \qquad (13)$$

where F is a fast neutron fluence in terms of 10^{22} neutron/m^2 ($E \geq 0.5$ MeV).

As was shown in ref. [18], there is no need in taking into account the flux density on radiation embrittlement of VVER-1000 BM after the accelerated irradiation.

A comparison of the VVER-1000 RPV BM database with additional data for VVER-1000 RPV BM subjected to accelerated irradiation with dependence (13) is shown in figure 2.

The comparison of experimental data and dependence (13) brings us to a conclusion of conservatism of the newly deduced dependence for BM and a feasibility of spreading its action to the values of a fast neutron fluence of 8.0×10^{23} neutron/m^2 at a confidence level of 95%.

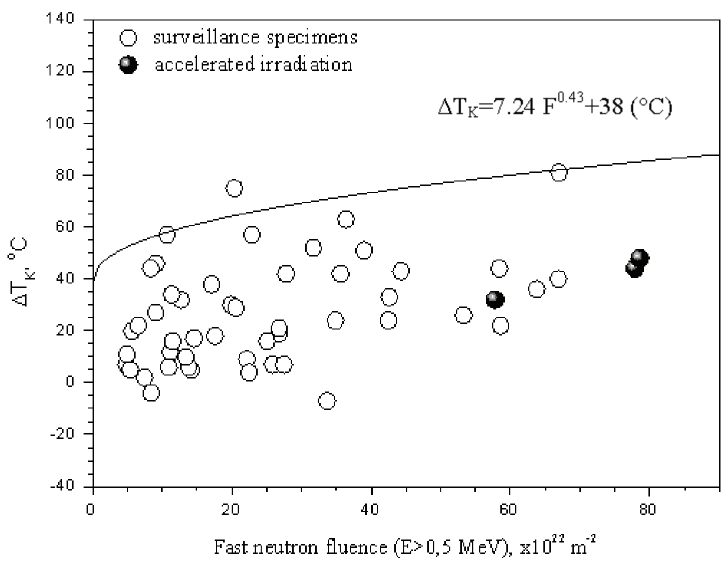

Figure 1.2. Extended database of VVER-1000 base metal *vs.* dependence (13)

Dependences from the Regularly Document [1] are conservative with regard to the existing databases of BM and WM surveillance specimens' radiation embrittlement in VVER-1000 reactor pressure and can be used for PTS analysis of reactor pressure vessels to a neutron fluence of $6.4\cdot10^{23}$ neutron/m^2 with energy above 0.5 MeV. When neutron fluence values exceed the said ones, the dependence from the Regularly Document [1] for weld metal can give a non-conservative estimate of transition temperature due to underestimation of phosphorus grain-boundary segregation processes which increases with the exposure time in a reactor under operation temperature and irradiation levels.

CONCLUSIONS

Physically reasonable dependences have been developed for conservative estimation of VVER-1000 RPV WM and BM that take into consideration the degradation of the RPV metal following the hardening and non-hardening mechanisms under the influence of operational factors:

1. The conservative estimate of a change of transition temperature of 15H2NMFA(A) steel welds due to a simultaneous impact of neutron irradiation to a fast neutron fluence of 8.0×10^{23} neutron/m^2 and temperatures of 290±10 ^{o}C obeys dependence:

$$\Delta T_K=1.48\cdot C_{Ni}C_{Mn}F^{\cdot 0.8}+1.3\cdot C_{Ni}{}^4\cdot \exp(-0.02\cdot T_{k0})\times(1-\exp\times(-2.2\times10^{-5}\cdot t))^{0.6}+25\ (^oC),$$

where $1.00\%\leq C_{Ni}\leq1.90\%$, $0.40\%\leq C_{Mn}\leq1.10\%$, $C_P\leq0.012\%$, and F is a fast neutron fluence in terms of 10^{22} neutron/m^2 ($E\geq0.5$ MeV).

2. The conservative assessment of a change of transition temperature of 15H2NMFA(A) steel owing to a simultaneous impact of neutron irradiation to a fast neutron fluence of 8.0×10^{23} neutron/m^2 and temperatures of 290±10 ^{o}C obeys dependence:

$$\Delta T_K=7.24\ F^{0.43}+38\ (°C)$$

where F is a fast neutron fluence in terms of 10^{22} neutron/m^2 ($E\geq0.5$ MeV).

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