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Division I

RADIATION EMBRITTLEMENT OF VVER-1000 REACTOR PRESSURE VESSEL MATERIALS

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INTRODUCTION

At present, justification of possible extension of service life of operating NPP Units with VVER-type reactors up to 60 years and more is a main problem, solution of which is required for sucessfull power supply in Russia for the near 30 years.

One of main problems when supporting long term operation of NPP with VVER is justification of the strength of reactor pressure vessel (RPV) as nonreplaceable equipment. Mechanical properties changes of RPV metal under influence of operating factors limits its life, which is determined by pressure vessel service life during which its possible brittle fracture is ruled out under any conditions including acidents.

During operation the RPV metal is subject to simultaneous effect of fast neutron flux and high temperature that corresponds to coolant temperature.

VVER-1000 reactor pressure vessels are manufactured of ferritic steel of grade 15Kh2NMFA(-A), the welds are made using welding wire of grade SV-10KhGNMAA or similar to it. The VVER-1000 RPV materials are characterized by a low content of such impurity elements as phosphorus and copper and by rather high content of nickel and manganese.

As a result of investigations of changes in the properties of VVER-1000 RPV materials under irradiation it is shown that there are dependences of irradiation embrittlement rates on nickel and manganese contet in metal [Kryukov et al. (2002), Erak et al. (2008), Margolin et al. (2012), Miller et al. (2009), Alekseenko et al. (1997), Odette et al. (1986), Hawthorne (1976), Giannet et al. (1982), Kryukov et al. (1997), Erak et al. (1994), Amaev et al. (1999), Nikolaev (2007)]. This effect shows most speakingly in VVER-1000 RPV weld metal wherein nickel and manganese content is considerably more than those in base metal.

The paper deals with the investigstion of behavior of the VVER-1000 RPV weld metal and base metal materials with different content of nickel and manganese after accelerated irradiation (20-400 times higher than the RPV wall) and during irradiation within the framework of surveilance-specimen programs wherein the irradiation rate approximately coincides with the reactor vessel wall.

The surveillance specimens investigation database is limited by the value of fast neutron fluence, and this makes impossible to make an advanced long term predication concerning change in reactor vessel metal properties that corresponds to the long term of operation. For long term predication a joint analysis of databases of surveillance specimens and research programs wherein the results on radiation embrittlement are available for high irradiation doses. An effect of fast neutron flux on the rate of transition temperature shift has been investigated for adequate prediction of radiation embrittlement of vessel material.

The comparative analysis of radiation embrittlement of VVER-1000 RPV materials after accelerated irradiation as well as of surveillance data has been carried out.

As the reactor pressure vessel material is subject to thermal ageing [Nikolaev (2007)] the degradation mechanisms realized both under neutron irradiation and under thermal effect were taken into account in analysis of change in metal properties. The influence of irradiation rate on the embrittlement level of material was taken into consideration. The objective of present work was to develop dependences for long term prediction of radiation embrittlement of the VVER-1000 RPV materials with regard for flux effect and thermal ageing.

RADIATION EMBRITTLEMENT AND THERMAL AGEING OF VVER-1000 RPV MATERIALS

It is known that radiation embrittlement and thermal ageing of VVER-1000 RPV materials are realized by the so-called hardening and non- hardening mechanisms [Gurovich et al. (1997)].

In case of radiation embrittlement formation of radiation-induced phases (carbides and precipitates) and dislocation loops belongs to the hardening mechanisms, and formation of phosphorus segregation on the grain boundaries or precipitate-matrix interface [Gurovich et al. (2000), Gurovich et al. (2009)] – to the non-hardening mechanisms. Moreover, in case of long term irradiations the damage induced by both mechanisms is realized, and in case of accelerated irradiations the damages are realized basically by the hardening mechanism, as segregation accumulation at the different boundaries under 290-300°C demands a long time and/or high doses of irradiation.

In case of thermal ageing the material damage can also be realized by both mechanisms. Although in this case there is the following feature in behavior of the reactor pressure vessel materials. Phosphorus segregation along the grain boundaries at temperature T=290-320°C occurs sufficiently slow and begins to perceptibly contribute to change in properties only after long term exposures (~200 000 hours and more). A fraction of intergranular fracture for surveillance specimens of the VVER-1000 PRV materials being during such time in the reactor reaches 25-30%

Change in the properties of the VVER-1000 reactor pressure vessel materials by the hardening mechanism with a temperature exposure at T≈300-350°C and with times up to ~40000 hours is poorly studied, but in compliance with a model approximation [Erak et al. (2008), Margolin et al. (2012)] used as a normative guide in Russia, has a dependence with extremum. After this, a transition temperature shift reaches a stable level and then, after at t>100000 hours, can start rising again due to a contribution connected with grain boundary segregation of phosphorus.

It is clear that during irradiation of VVER-1000 reactor pressure vessel materials at $T_{irr.}$~290-300°C all damage mechanisms that correspond to both neutron and temperature effect on material are realized simultaneously. In this case the integral shift of transition temperature in some approximation can be presented as a sum of "radiation" and "thermal" components.

So, in papers [Erak et al. (2008), Margolin et al. (2012)] an assumption was advanced that transition temperature of materials under irradiation may be represented as a result of two additive processes: radiation embrittlement concerned with formation of radiation-induced precipitates and dislocation loops and with thermal embrittlement to be realized with exposure times up to 100 000 hours in the form a function with the extremum and kept then a constant value.

In this case the transition temperature shift observed on surveillance specimens can be calculated as the sum of radiation ΔT_F and thermal ΔT_T embrittlement (Figure 1a):

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

where ΔT_T is described by the formula:

$$\Delta T_T(t)=\left(\Delta T_t^{\inf}+b_T\exp\left(\frac{t_T-t}{t_{OT}}\right)\right)\cdot th\left(\frac{t}{t_{OT}}\right) \qquad (2)$$

and ΔT_F is described by the formula:

$$\Delta T_F=A_F\cdot\left(\frac{F}{F_0}\right)^m,\ (F_0=1.0\times10^{22}\ \text{neutron/m}^2) \qquad (3)$$

where $\Delta T_t^{\inf}$, b_T, t_{OT}, t_T, A_F, m are parameters that correspond to the results of VVER-1000 surveillance specimens study [Margolin (2012)]. A schematic diagram of these dependencies is given in Figure 1.

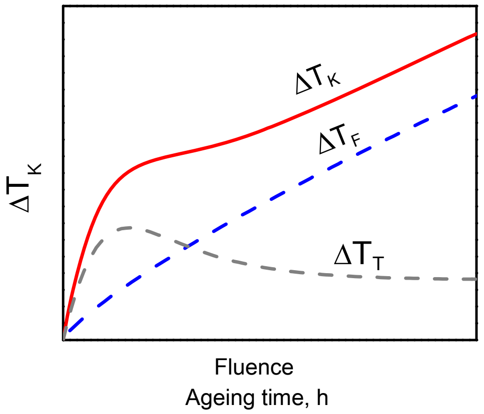


Figure 1. Schematic diagram of transition temperature shift of VVER-1000 reactor pressure vessel materials taking into account radiation and thermal effects onto material.

In formula (2) for weld metal with C_{Ni}>1.5% $\Delta T_t^{\inf}$=18 °C with the exposure time more than 100 000 hours, and for base metal and weld metal with C_{Ni}<1.3% $\Delta T_t^{\inf}$=2 °C.

It is to be noted that reaching the constant value of the thermal component of transition temperature shift $\Delta T_t^{\inf}$ with times more than 100 000 hours is an assumption which requires further investigations of microstructure and strength properties of the VVER-1000 reactor pressure vessel materials after thermal ageing at working temperatures. So, with the exposure times of 200 000 hours and more a significant increase in phosphorus content at the grain boundaries is probable that can result in increasing in material embrittlement value but not in keeping the constant value of thermal component.

When materials with different fast neutron flux are irradiated, the so-called flux effect can be observed.

For the materials of VVER-1000 reactor pressure vessel welds the flux effect is caused by different change in material structure at nanolevel after irradiation with various rate to the same level of damaging dose of neutrons. These changes can occur both by the “hardening” and by the “non-hardening” mechanism; this is to be specified more detailed by the results of microstructure investigations. But when analyzing of irradiation-caused changes in material properties a possible effect of fast neutron flux on the result should be taken into account. In this connection, if it is necessary to predict changes of RPV material properties according to accelerated irradiation results, several aspects appear that need to be taken into account:

- firstly, it is necessary to take into account the flux effect in radiation component of transition temperature shift (ΔT_F);

- secondly, in order to predict RPV material state corresponding to long operation period it is necessary to take into account a contribution of thermal ageing effects that are realized in RPV material during operation and are not realized in specimen materials under accelerated irradiation.

The effect of fast neutron flux on the rate of transition temperature shift was studied in order to take into account the flux effect. A conversion formula considering the irradiation rate was proposed in the following form:

$$\Delta T_F^{low\,flux} = \beta \ \Delta T_F^{high\,flux} = \frac{A_F^{SS}}{A_F^{RP}} \ \Delta T_F^{high\,flux} \tag{4}$$

where A_F^{SS} - the radiation embrittlement coefficient obtained by the results of surveillance-specimen investigation, and A_F^{RP} - the radiation embrittlement coefficient obtained by the results of specimen investigations within the research programs (accelerated irradiation). A difference in a value of radiation embrittlement (ΔT_F) of materials under accelerated and non-accelerated irradiation is shown schematically in Figure 2.

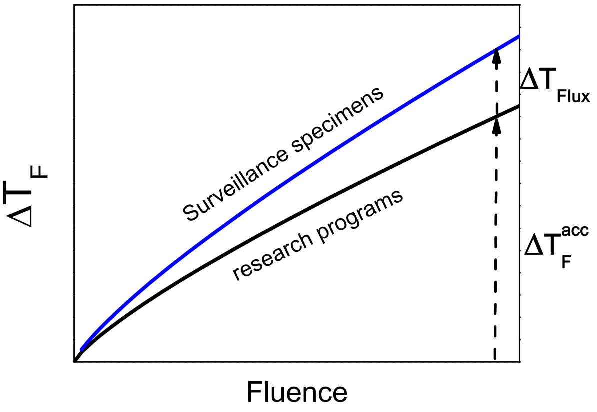


Figure 2. Schematic diagram of radiation embrittlement under accelerated and non-accelerated irrradiation.

FLUX EFFECT INVESTIGATION

The VVER-1000 RPV weld materials with different nickel and manganese content were studied.

Nickel and manganese content in weld and base metal was within a wide range covering the prescribed specifications for frame material. Copper and phosphorus content is not high and corresponds to average values for VVER-1000 RPV materials.

The specimens were irradiated with high accelerated factor in VVER-1000 power reactor (Novovoronezh NPP, Unit 5) and in NRC "Kurchatov Institute" research reactor IR-8. Comparative data on irradiation parameters are presented in Table 1. Irradiation temperature in all cases was within the range of 290ºC ± 10ºC.

It is to be taken into account simultaneous running the processes affecting additionally the transition temperature shift for correct comparison of radiation embrittlement effect on operating characteristics of materials with different fast neutron fluxes and, hence, with various exposure times.

Table 1 : Irradiation parameters of the materials under consideration

	Neutron flux E>0.5 MeV, ($\times 10^{14}$), [$m^{-2}s^{-1}$]		Spectral index $SI_{0,5/3,0}$	
	min	max	min	max
VVER-1000 surveillance specimens	2.58	24.0	5.0	7.8
VVER-1000 research assemblies	80.0	1000	6.7	9.4
IR-8 research assemblies	200 - 1600		8.6 - 11.3	
VVER-1000 reactor pressure vessel wall	~ 4.0		5.8	

Based on the model approximation of material damage with simultaneous impact of temperature and neutron flux that represented by the expressions (1-3), we single out a clear form of a dependence of radiation component of transition temperature shift for VVER-1000 RPV weld and base metal and analyze the flux effect:

$$\Delta T_F(F) = \Delta T_K(F,t) - \Delta T_T(t) \qquad (5)$$

In further analysis of databases on radiation embrittlement of VVER-1000 RPV weld and base materials it is assumed that $\Delta T_T(t)$ is described by dependence (2) for surveillance specimens [Margolin (2012)], and $\Delta T_T(t)$=0 for research programs. Then the value of radiation embrittlement is determined as follows:

$$\Delta T_F^{highflux}(F\ ) = \Delta T_K^{high\ flux} \qquad (6)$$

In dependences (5, 6) ΔT_F is described by the dependence (3),wherein a chemical factor should take into account a possible effect of impurities and some alloying elements on radiation embrittlement of the metal.

The VVER-1000 reactor pressure vessel weld metal is a very pure material as to impurities, with this, impurity variation in various reactor pressure vessels is very insignificant.

It is known [Kryukov et al. (2002), Margolin et al. (2012), Miller et al. (2009), Alekseenko et al. (1997), Odette et al. (1986), Hawthorne (1976), Giannet et al. (1982), Kryukov et al. (1997), Erak et al. (1994), Amaev et al. (1999), Nikolaev (2007)] that among alloying elements nickel and manganese influence considerably on the radiation embrittlement of the VVER-1000 RPV materials. Atoms of these elements show a susceptibility to formation of dislocation barriers in the form of non-equilibrium segregats (clusters) or they are a part of the latter as one of the components [Miller et al. (2009)].

Thus, the chemical factor should take into account Ni, Mn and Si content in material. An attempt of consideration of Si effect onto radiation embrittlement of reactor pressure vessel weld materials was made in papers [Margolin et al. (2012), Nikolaev (2007)] in regression analysis of experimental data on surveillance specimens' tests. The authors of the present paper suppose that it is reasonable to consider only evident effect of Ni and Mn on radiation embrittlement of VVER-1000 RPV weld metal in order to simplify the form of functional dependence and reduction of the number of adapted coefficients having taken into consideration the limitation of databases of research programs.

Respectively, as model regression to describe the behavior of weld materials under irradiation, the following formula taken into account Ni and Mn content was chosen:

$$\Delta T_F = A_F(C_{Ni}, C_{Mn})\, F^{0,8} \qquad (7)$$

where ΔT_F is the ductile-to-brittle transition temperature shift, C_{Ni} – nickel concentration, C_{Mn} – manganese concentration, F – fast neutron fluence in 1×10^{22} neutron/m^2.

To evaluate the “flux effect” correctly the research programs database has been limited to the fast neutron fluence value of 6×10^{23} neutron/m^{-2} in accordance with the highest value of accumulated fast neutron fluence on surveillance specimens data.

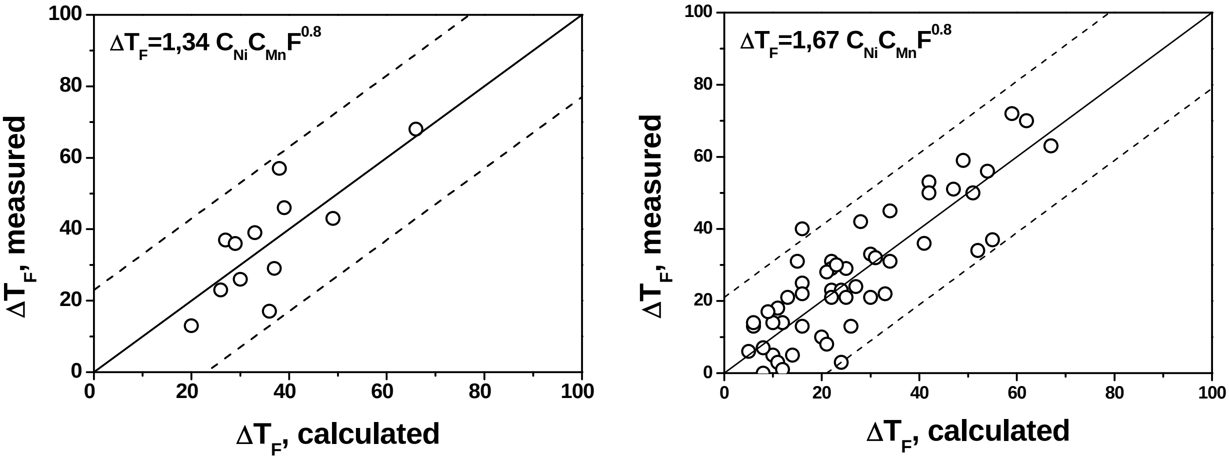
During research programs database analysis of radiation embrittlement of weld metal with high nickel content (C_{Ni}>1.3%) the following formula was obtained:

$$\Delta T_F = 1.34\ C_{Ni}\ C_{Mn}\ F^{0,8} \quad (\sigma = 10.4\ ^{\circ}C) \tag{8}$$

And for surveillance specimens database the following formula was obtained:

$$\Delta T_F = 1.67\ C_{Ni}\ C_{Mn}\ F^{0,8} \quad (\sigma = 10.6\ ^{\circ}C) \tag{9}$$

Figures 3 (a) and (b) show comparison of experimental and calculated data of radiation contribution to transition temperature shift for research programs for weld and surveillance specimens of welds, respectively. It is shown that formulae (8) and (9) describe the analyzed data block of experimental results with a probability of 95% approximately with the same value of dispersion.



a) b)

Figure 3. The comparison of experimental and calculated values of radiation component in transition temperature shift of weld for research programs (a) and for surveillance specimens (b).

The comparison of formulae (8) and (9) for accelerated irradiated specimens and for surveillance specimens, respectively, allows to calculate a coefficient taking into account the flux effect and it is equal to 1.25.

Then the value of ΔT_F for RPV wall can be obtained by multiplication of ΔT_F value under accelerated irradiation for this material by the coefficient β=1.25.

A simple model independent on chemical elements content in material is plotted as a regression model for base and weld metal with nickel concentration C_{Ni}<1.3 % :

$$\Delta T_F = 1.45\, F^{0,8} \qquad \sigma = 18.3\ ^{o}C \tag{10}$$

As a result of processing the database for research programs a model is obtained:

$$\Delta T_F = 1.44\, F^{0,8} \qquad \sigma = 19.7\ ^{o}C \tag{11}$$

A ratio of radiation embrittlement coefficients in models (10) and (11) is equal practically to 1.0, this means no need for consideration of flux effect for radiation embrittlement of VVER-1000 RPV materials with low content of nickel.

Thus, the value of the radiation component of transition temperature shift can be obtained based on the results on accelerated irradiation using the following dependence:

$$\Delta T_F^{low\ flux} = \beta \times \Delta T_F^{high\ flux}, \tag{12}$$

where $\beta = 1.25$ for weld metal with nickel concentration $C_{Ni} > 1.3\%$ and $\beta = 1.0$ for base metal and weld metal with nickel concentration $C_{Ni} < 1.3\%$.

PREDICTION OF RADIATION EMBRITTLEMENT FOR VVER-1000 RPV MATERIALS DURING LONG TERM-OPERATION

The obtained ratios connecting the results of accelerated and non-accelerated irradiations make possible to add available database of testing the irradiated surveillance specimens of VVER-1000 reactor pressure vessels to the results of research programs and create hereby an extended database to establish a model of transition temperature shift within the range of the fluence corresponding to long term operation.

As the results of thermal ageing investigations of base metal at 300-320°C and under short (up to 150 000 hours) exposures that are made lately in NRC "Kurchatov Institute" [Chernobaeva et al. (2013)], do not confirm a form of functional dependence with maximum (2), available array of experimental values of ΔT_K may be considered without this assumption. Then we obtain the following regression dependence for conservative evaluation of transition temperature shift of VVER-1000 RPV base metal during long term-operation (Figure 4):

$$\Delta T_K = 8.2F^{0.4} + \Delta T_T + 37, \tag{13}$$

where ΔT_T is accepted to be equal to 0°C according to an array of experiment results now available, and in future, when obtaining additional results on thermal ageing of VVER-1000 RPV base metal can be changed to the corresponding functional dependence on time and phosphorus content in the material.

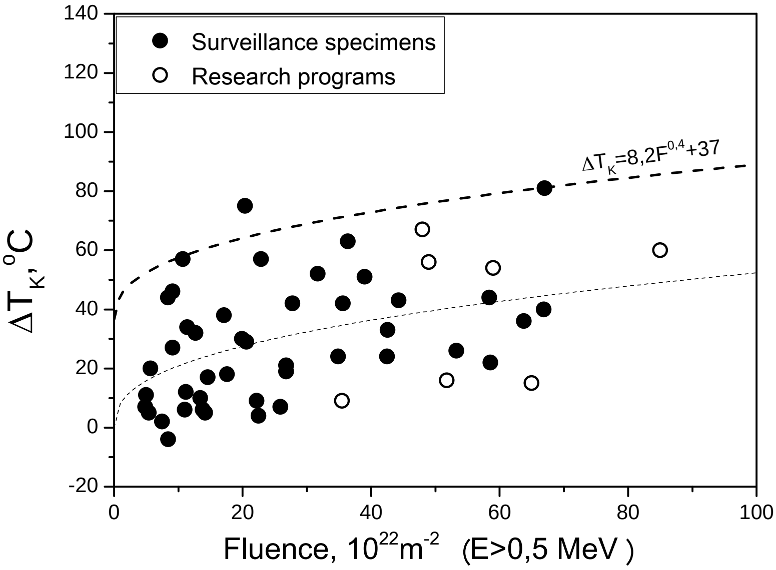


Figure 4. Comparison of predictive dependence (13) with the experimental results of VVER-1000 RPV base metal obtained during investigation of surveillance specimens and research programs.

As it is shown in previous section for the results on radiation embrittlement of VVER-1000 RPV weld metal obtained during accelerated irradiation it is not required to make a special correction for consideration of flux effect for harmonization of these data with surveillance specimens investigation results with nickel content < 1.3% and such a correction is required if nickel content is $C_{Ni} \geq 1.3\%$. Taking into account the fact we extend the array of experimental results to the high values of fast neutron fluence we obtain the following dependence for the radiation component of transition temperature shift for VVER-1000 RPV weld metal in regression analysis of the created database.

$$\Delta T_F = 1.68\ C_{Ni}\ C_{Mn}\ F^{0,8} \quad (\sigma = 13.5\ ^{o}C) \tag{14}$$

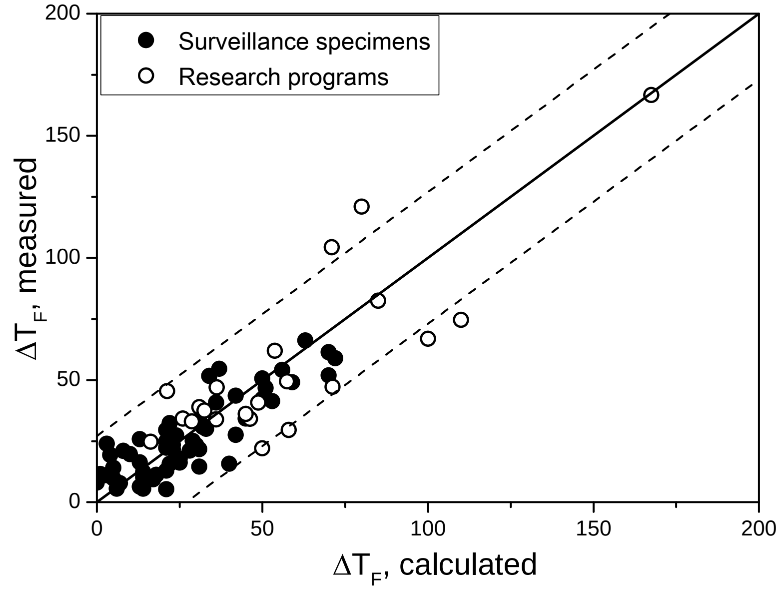


Figure 5. Comparison of experimental values and model-calculated values (14) of radiation embrittlement of VVER-1000 RPV weld metal obtained in frame of surveillance specimens and research programs.

Then with regard for dependence for material thermal ageing we obtain the following model for conservative prediction of change in VVER-1000 RPV weld metal properties during long term operation.

$$\Delta T_K = 1.68\ C_{Ni}\ C_{Mn}\ F^{0.8} + \Delta T_T + 27, \tag{15}$$

where ΔT_T value is determined by a normative dependence (2), and in future when obtaining additional experimental data it can be changed due to replacement of the constant value of material thermal ageing during long term exposure times by the component depending on phosphorus concentration in material and exposition time of reactor pressure vessel operation.

CONCLUSIONS

The data available on radiation embrittlement of VVER-1000 RPV materials obtained under non-accelerated (according to surveillance specimens programs) and accelerated irradiation conditions has been analyzed.

With model assumption that transition temperature shift can be presented as a sum of radiation component ΔT_F and thermal component ΔT_t in the form of $\Delta T_k = \Delta T_t + \Delta T_F$, the flux effect has been evaluated for VVER-1000 RPV materials. The value of radiation component of transition temperature shift can be obtained from accelerated irradiation results using the following dependence:

$$\Delta T_F^{low\ flux} = \beta \times \Delta T_F^{high\ flux},$$

where $\beta = 1.25$ for weld metal with nickel concentration $C_{Ni} \geq 1.3\%$ and $\beta = 1.0$ for base metal and weld metal with nickel concentration $C_{Ni} < 1.3\%$.

Databases on radiation embrittlement of VVER-1000 RPV materials are extended with regard for this ratio.

The dependences which may be used for long term prediction of transition temperature shift of VVER-1000 RPV base metal (13) and of weld metal (15) have been obtained, where the value of ΔT_T for base metal is accepted to be equal to 0°C, and it is determined for weld metal in compliance with Russian normative guide (2). In future, when obtaining additional experimental data the value of ΔT_T for long term operation may be changed by replacement of the constant value of metal thermal ageing by the component dependent on phosphorus concentration in material and on time of reactor pressure vessel operation.

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