Procedure of accelerated irradiation results usage for prediction of the material state corresponding to long-term operation of the reactor pressure vessel

D. Erak [a], B. Gurovich [b], E. Kuleshova [c], Ya. Shtrombakh, O. Zabusov [d], D. Zhurko [e]

National Research Centre “Kurchatov Institute”, Kurchatov sq.1, Moscow 123182, Russia

[a] erak@mail.ru, [b] ba_gurovich@irtm.kiae.ru, [c] evgenia-orm@yandex.ru, [d] zabusov@rtm.kiae.ru, [e] zhurko_d@mail.ru

Keywords: reactor pressure vessel, radiation embrittlement, flux effect, thermal ageing, accelerated irradiation, long-term prediction.

Introduction

At present, validation of lifetime extension for operating Nuclear power plant (NPP) units with VVER-type reactors is one of the main strategic directions of works in the development of nuclear power engineering in Russia for the years ahead. Possibility of lifetime extension up to 60 years and more is under consideration for VVER-1000 reactors.

An overriding problem at long operation of NPP with VVER is verification of reactor pressure vessel (RPV) safety (as nonreplaceable equipment) for entire period of extension. Assurance of reliable work of the RPVs is one of the most important problems of NPP safety. During operation there is a change in mechanical properties of RPV metals, that results in radiation embrittlement. This limits the RPV safe radiation lifetime during which its brittle failure is impossible, including emergencies.

During operation the RPV metals undergo the simultaneous effect of operational factors (fast neutron flux and high temperature, corresponding to the temperature of the NPP coolant).

VVER-1000 reactor pressure vessels are manufactured of ferritic steel 15Kh2NMFA (-A), the welds are made using weld wire SV-10KhGNMAA or similar one. The materials of VVER-1000 RPV are with low content of phosphorus and copper and rather high content of nickel and manganese.

Some studies of radiation embrittlement of VVER-1000 RPV materials performed both within the research programs with specimens accelerated irradiation and within surveillance programs with irradiation in operating conditions of the RPV wall show strong dependence of irradiation embrittlement on nickel and manganese content in the metal. The limiting elements in view of radiation embrittlement for VVER-1000 RPVs are the welds where content of nickel and manganese is considerably more than in base metal.

Hence, prediction of radiation embrittlement for RPV welds is especially urgent task for validation of RPVs long operation.

Behavior of the materials of VVER-1000 RPV welds with different nickel and manganese content after accelerated irradiation (100-400 times higher than the RPV wall) was studied in this work. Effect of fast neutron flux on increase rate of transition temperature was also studied for adequate prediction of radiation embrittlement of the RPV materials.

The comparative analysis of radiation embrittlement of VVER-1000 welds after accelerated irradiation and surveillance data was carried out.

The influence of irradiation rate and thermal ageing on the embrittlement level is taken into consideration. Elaboration of a procedure of taking the flux effect and thermal ageing into consideration when using the results of accelerated irradiation for long-term prediction of VVER-1000 RPV materials radiation embrittlement is the goal of this work.

Radiation embrittlement and thermal ageing of VVER-1000 RPV materials

It is known that radiation embrittlement and thermal ageing of VVER-1000 RPV materials is realized by so-called strengthening and non-strengthening mechanisms [1].

In case of radiation embrittlement, the strengthening mechanisms, realizes due to formation of radiation-induced phases (carbides and precipitates) and dislocation loops, and non-strengthening mechanisms – due to formation of phosphorus segregation on the grain boundaries or precipitate-matrix interface [2, 3]. With this, in case of prolonged irradiations the damages induced by the both mechanisms are realized, and in case of accelerated irradiation the damages are realized basically by the strengthening mechanism, because segregation accumulation at the different boundaries under 290-300°C demands a long period of time and/or high doses of irradiation. This fact can be indirectly proved by absence of intergranular component in fractures of the tested Charpy specimens of VVER-1000 weld metal irradiated to fluence of 5×10^{23} neutron/m^2 (E>0,5 MeV) during ~ 7000 hours. Meanwhile, in fractures of Charpy surveillance specimens of VVER-1000 RPV materials irradiated to fluence of 5×10^{23} neutron/m^2 (E>0,5 MeV) during 100000 hours the portion of brittle intergranular fracture reaches 30-35%. Nevertheless, for the specimens irradiated in accelerated way to fluence 10×10^{23} neutron/m^2 (E>0,5 MeV) during ~ 7000 hours a share of intergranular component in fractures of tested Charpy specimens of VVER-1000 weld metal is 25-30% that indicates a formation of radiation-induced segregation of phosphorous under accelerated irradiation that becomes significantly apparent with high values of fast neutron fluence.

In case of temperature ageing, damage of material can be also realized by the both mechanisms. Though, in this case, there is a following peculiarity in RPV material behavior. A phosphorus segregation on the grain boundaries under T=290-320°C occurs rather slowly and makes an appreciable contribution into properties change only after prolonged holding (~200000 hours and more). The portion of brittle intergranular fracture for surveillance specimens of VVER-1000 RPV materials that have remained in the reactor during such period of time reaches 25-30%.

Change of transition temperature shift of VVER-1000 reactor pressure vessel materials by the strengthening mechanism with holding under T≈300-350°C for up to ~40000 hours has a dependence with extremum and it is usually associated with precipitation and coagulation of carbides. After this a transition temperature shift comes to a stable level and then, after t>100000 hours, it can start rising again due to contribution connected with phosphorus segregation on the grain boundaries. At the same time some microstructure investigations recently have been in NRC “Kurchatov institute” not show any carbide precipitations in thermal aged materials.

It is clear that in case of VVER-1000 reactor pressure vessel material irradiation under $T_{irr.}$~290-300°C all damage mechanisms related to both neutron and temperature impact are realized. With this, the integral shift of transition temperaturecan be presented approximately as a sum of “radiation” and “thermal” components.

So, in the works [1, 2] the idea was proposed that transition temperature shift of materials under irradiation is a result of two additive processes: natural radiation embrittlement concerned with formation of radiation-induced nickel-manganese precipitates and dislocation loops and thermal embrittlement caused by long time exposure – there is phosphorus segregation on the grain boundaries of the irradiated material.

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 (Fig. 1a)

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

Where ΔT_T is described by the formula

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

and ΔT_F is described by the formula

$$\Delta T_F = A_F \cdot \left(\frac{F}{F_0} \right)^m, \ (F_0 = 1{,}0 \times 10^{22}\ \text{neutron/m}^2) \tag{3}$$

$\Delta T_t^{\inf}$, b_T, t_{OT}, t_T, A_F, m – parameters, according to the results of VVER-1000 surveillance specimens studies [2]. A schematic diagram of these dependencies are given in Fig.1a.

In the formula (2) $\Delta T_t^{\inf}$ =18 ºC with exposure time more than 100 000 hours.

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

For RPV weld materials the flux effect probably manifests itself in reduced precipitate density in the material irradiated in accelerated way in IR-8 in comparison with precipitate density in the material of irradiated surveillance specimens. In this connection, if it is necessary to predict change of RPV material properties according to the 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 temperature ageing effects that are realized in RPV material during operation and are not realized in specimen materials under accelerated irradiation.

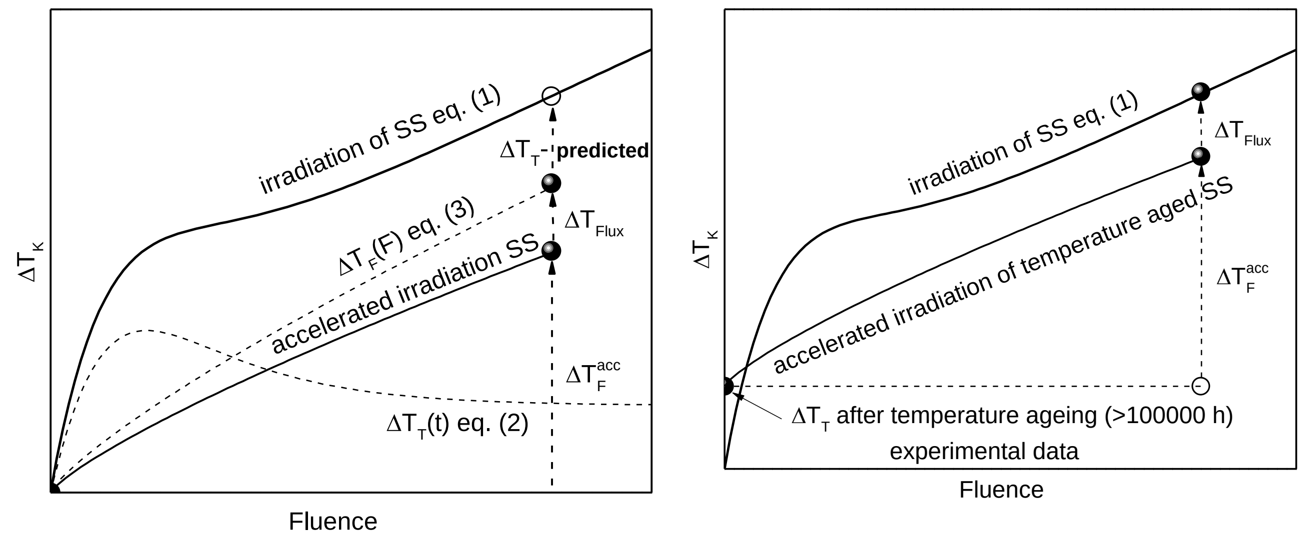
The effect of fast neutron flux density on the rate of transition temperature shift was studied. A conversion formula considering the effect of irradiation rate was proposed in the following form:

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

Temperature ageing can be taken into account by two ways.

The first way is the following: the obtained result of accelerated irradiation corrected with account of flux effect is added with a value corresponding to the predicted time by existing dependence of transition temperature shift change due to temperature ageing, but in this case it is necessary to know the dependence of change $\Delta \mathrm{T_T}(t)$ (Fig. 1a).

The second way seems to be more acceptable and consists of accelerated irradiation of specimens of temperature sets of surveillance specimens exposed to operating temperature for a significant period of time (>100000 hours) without irradiation (Fig. 1b).



a b

Fig. 1. Two ways of obtained predictive values of the vessel materials radiation embrittlement with the use of accelerated irradiation

In case of realization of accelerated irradiation by any of suggested ways a question of taking the flux effect into account remains unsolved. Value of this effect for VVER-1000 reactor pressure vessel weld materials with nickel content Ni>1.5% is stated in this work.

Materials and the methods of studies

The materials of the welds of VVER-1000 RPVs with different nickel and manganese content were studied. The chemical composition of these materials is presented in Table 1.

Table 1. Chemical composition of the materials studied

Material	Chemical composition, [%]								
	C	Si	Mn	Cr	Ni	Mo	S	P	Cu
Weld 1	0.110	0.14	0.73	1.88	1.74	0.55	0.008	0.010	0.08
Weld 2	0.060	0.40	0.94	1.70	1.70	0.67	0.012	0.007	0.04
Weld 3	0.035	0.35	0.92	1.60	1.60	0.55	0.011	0.007	0.06
Weld 4	0.040	0.24	0.97	1.60	1.28	0.60	0.006	0.011	0.06
Weld 5	0.075	0.30	0.92	1.80	2.45	0.65	0.007	0.006	0.05
Weld 6	0.070	0.31	1.10	1.72	1.88	0.68	0.010	0.009	0.03
Weld 7	0.040	0.28	0.98	1.71	1.76	0.66	0.006	0.010	0.04

It should be noted that nickel content in weld metal is in a wide range, that covers the specified values in technical specifications for the material. Copper and phosphorus content is not high and corresponds to average values for VVER-1000 reactor materials.

The accelerated irradiation of specimens was performed in the VVER-1000 reactor (Unit No. 5, Novovoronezh NPP) and in the research reactor IR-8 at NRC "Kurchatov institute".

Comparative data concerning parameters of irradiation are presented in Table 2.

Table 2. 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	72.0	1500	6.7	9.4
IR-8 research assemblies	200 - 1760		8.6 - 11.3	
VVER-1000 reactor pressure vessel wall	4.07		5.8	

The studies were carried out using method of Charpy specimen of $10\times10\times55$ mm^3 and subsize Charpy specimens of $5\times5\times27,5$ mm^3 testing.

In all cases the irradiation temperature was in the range of 290ºC ± 10 ºC.

Study of irradiation embrittlement of VVER-1000 RPV materials under accelerated irradiation

The results of transition temperature evaluation after the accelerated irradiation were analyzed. A clear dependence of radiation embrittlement of RPV materials on nickel content was obtained as it was in previous works of various researchers [6-13]. The data for the materials with similar Mn content ~ 0,92 – 0,97% and various Ni content 1,28 – 2,45% are presented in Fig.2.

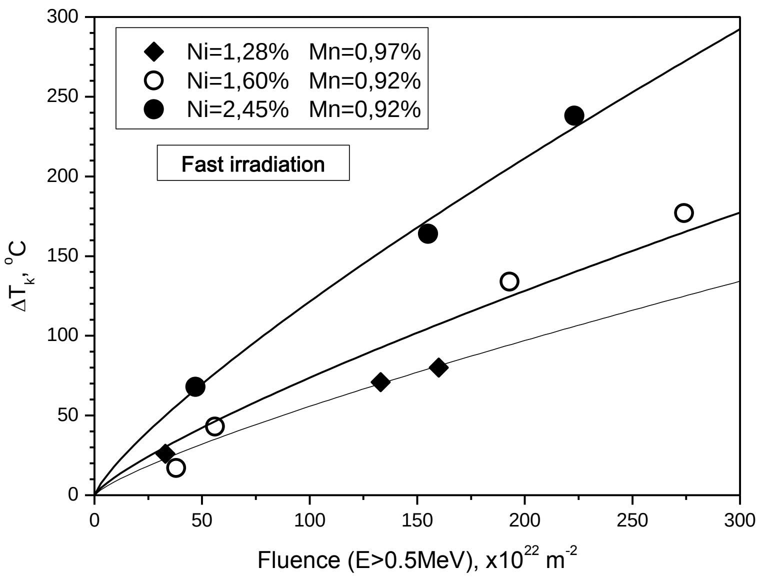


Fig. 2. Clear dependence of radiation embrittlement of RPV materials on nickel content

The data show obvious dependence of irradiation embrittlement on nickel content in the material. It should be noted that as in some previous studies [10, 11] the continuous linear (or close to it) dependence of transition temperature on fast neutron fluence is observed under the accelerated irradiation.

As it is known [6-13] not only nickel, but also manganese influences considerably on the radiation embrittlement of RPV materials. Therefore the regression analysis of the database was carried out for the functional dependence where nickel and manganese content in the material were the parameters. The results of microstructure studies of the VVER-1000 irradiated materials show formation of Ni-Mn- Si precipitates under the irradiation.

Taking into account the results of microstructure studies, a model depending on Ni and Mn content in the material is chosen as a regression for describing the behavior of weld materials under irradiation:

$$\Delta T_F = A_F(C_{Ni}, C_{Mn})\, F^{0,8} \qquad (5)$$

where ΔT_F is the transition temperature shift, C_{Ni} is nickel content and C_{Mn} is manganese content, F is fast neutron fluence in 1×10^{22} neutron/m^2.

During processing the database of weld metal with high nickel content (C_{Ni}>1.5%) the radiation embrittlement is described in the following way for research programs:

$$\Delta T_F = 1{,}34\ C_{Ni}\ C_{Mn}\ F^{0,8} \quad \sigma = 10{,}4\ ºC, \qquad (6)$$

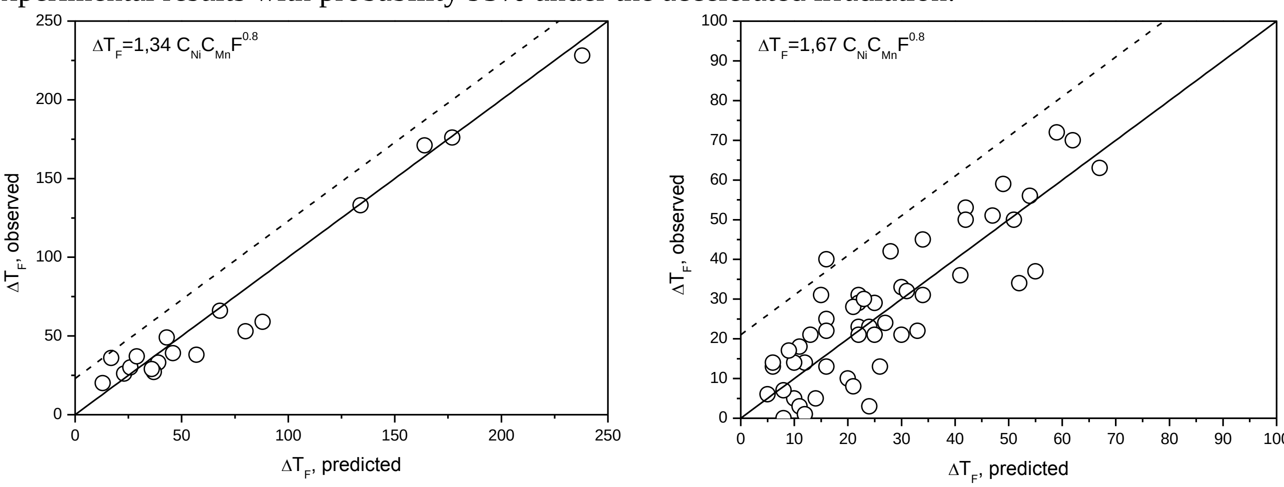
For the correct comparison of radiation embrittlement at different fast neutron fluxes, and, therefore, under different holds at operational characteristics of the materials, it is necessary to consider the possibility of simultaneous processes influencing additively on the transition temperature shift of the materials.

The regression analysis of the available database of the VVER-1000 surveillance tests performed with taking into account simultaneous radiation and thermal embrittlement of the material (described by the formulas 1-3) led to the following functional dependence of the radiation part in transition temperature shift:

$$\Delta T_F = 1{,}67\ C_{Ni}\ C_{Mn}\ F^{0,8} \quad \sigma = 10{,}6\ ºC, \qquad (7)$$

The parameters of regression were not improved by the attempt to take silicon into account.

The comparison of experimental and calculated data of radiation part in transition temperature shift of the weld research programs and of the weld surveillance specimens is presented in Fig.3 (a) and (b) respectively. It is shown that the formula describes the analyzed data block of experimental results with probability 95% under the accelerated irradiation.



a b

Fig. 3. The comparison of experimental and calculated data of radiation part in transition temperature shift of a) the weld research programs and of b) the weld surveillance specimens

To evaluate the "flux effect" correctly, the database of the research programs has been limited to the fast neutron fluence value of 60×10^{22} neutron/m^{-2} in accordance with the data obtained from surveillance specimens.

In order to show the results of processing at one diagram we normalize the value of ΔT_F to the factor $C_{Ni} \cdot C_{Mn}$ (Fig. 4).

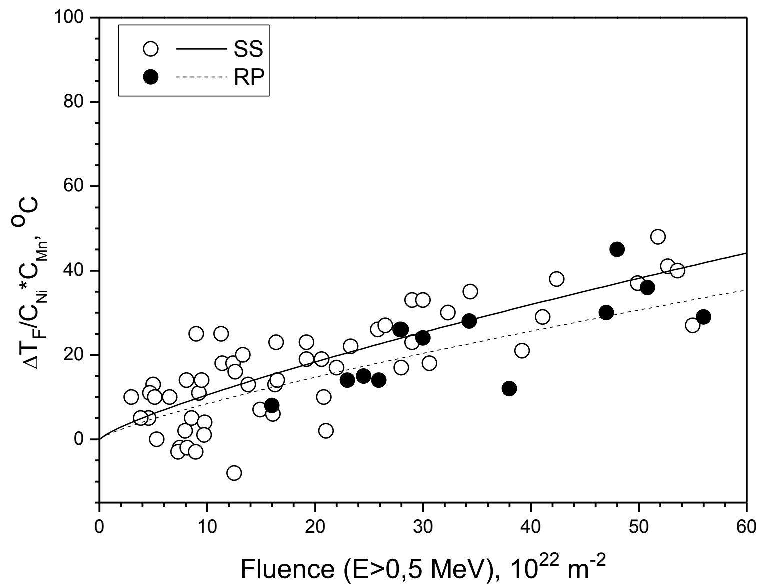


Fig. 4. Radiation embrittlement of weld metal (C_{Ni} >1.5%) for surveillance specimens and research programs

The comparison of expressions (5) and (6) for specimens irradiated by accelerated way and surveillance specimens, respectively, allows to calculate the conversion factor, which accounts the flux effect.

Then ΔT_F for the RPV wall can be obtained by the multiplication of ΔT_F value under accelerated irradiation for this material by the coefficient of (dependence 4) β =1,25 within the fluxes ranges limited in table 2.

Conclusions

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

With model assumption that the transition temperature shift can be presented as a sum of radiation part ΔT_F and thermal part ΔT_t in $\Delta T_k = \Delta T_t + \Delta T_F$, a procedure of accelerated irradiation results application for prediction of change of properties of VVER-1000 reactor pressure vessel weld materials corresponding to long-term operation was suggested. This procedure is described by the formula $\Delta T_k = \Delta T_t + \Delta T_F^{acc} + \Delta T_{Flux}$,

where ΔT_T - transition temperature shift due to thermal ageing determined for the period of time > 100 000 hours according to existing dependencies or on surveillance specimens,

ΔT_F^{acc} – transition temperature shift after accelerated irradiation.

$\Delta T_{Flux} = 0{,}25 \Delta T_F^{acc}$ –addition to flux effect for weld material with nickel content more than 1.5%.

For RPVs being under operation more than 30 years it is proposed to obtain predictive values of the vessel materials radiation embrittlement with the use of accelerated irradiation of surveillance specimens of temperature sets with exposure time > 100000 hours.

The work was carried out within the framework of the Federal Agency lot on science and innovations, State Contract 02.523.12.3012 and financial support of ISTC fund (Project № 3420).

References

[1] B.A.Gurovich, E.A.Kuleshova, Yu.A.Nikolaev, Ya.I.Shtrombakh. *Assessment of relative contributions from different mechanisms to radiation embrittlement of reactor pressure vessel steels.* J. of Nucl.Mat., 246 (1997), p.91-120

[2] B.A.Gurovich, E.A.Kuleshova, Ya.I.Shtrombakh, O.O. Zabusov, E.A. Krasikov. *Intergranular and intragranular phosphorus segregation in russian pressure vessel steels due to neutron irradiation*. J. of Nucl.Mat., v. 279, Issues 2–3, (2000), p.259-272

[3] B.A. Gurovich, E.A. Kuleshova, D.Yu. Erak, A.A. Chernobaeva, O.O. Zabusov. *Fine structure behaviour of VVER-1000 RPV materials under irradiation*. J. Nucl. Mat., 389 (2009), pp. 490–496.

[4] D.Yu.Erak, B.Z.Margolin, Yu.R.Kevorkyan, D.A.Zhurko, V.A.Nikolaev, *The analysis of radiation embrittlement of VVER-1000 reactor pressure vessel materials,* Collection of abstracts of the tenth international conference on material issues in design, manufacturing and operation of nuclear power plants equipment, St. Petersburg, October 7–9 (2008), p. 41.

[5] B.Margolin, V.Nikolaev, E.Yurchenko, Yu.Nikolaev, D.Erak, A.Nikolaeva. *Analysis of embrittlement of WWER 1000 RPV materials*. Int. J. Pres. Ves. & Piping, 89 (2012), pp. 178-186.

[6] N.N.Alekseenko, A.D.Amaev, I.V.Gorynin, V.A.Nikolaev. *Radiation damage of nuclear power plant pressure vessel steels*. Illinois USA: La Grange Park, (1997).

[7] G.R. Odette and G.E. Lucas, *Radiation Embrittlement of Nuclear Reactor Pressure Vessel Steels*, ed. L.E. Steele, ASTM-STP 909 (American Society for Testing and Materials, Philadelphia, (1986) , p. 206.

[8] J.R. Hawthorne. *Further observations of A-533 steel plate tailored for improved radiation embrittlement resistance*. J.Pressure Vessel Techn. (ASME), (1976), pp. 111-117.

[9] A.M. Kryukov, Yu.A. Nikolaev, T. Planman, P.A. Platonov. *Basic results of the Russian WWER-1000 surveillance program*. Nuclear Engineering and Design 173 (1997), pp. 333-339.

[10] D.Erak, A.Kryukov, O.Vishkarev *Nickel content effect on radiation embrittlement of VVER-1000 vessel steel*. Proceedings. Third International Conference on Material Science Problems in NPP Equipment Production and Operation, CRISM “PROMETEY” St.Petersburg, Russia, vol. 3, (1994), pp. 846-849.

[11] A.D.Amaev, D.Yu.Erak, A.M.Kryukov. *Radiation Embrittlement of WWER-1000 Pressure Vessel Materials* – Irradiation Embrittlement and Mitigation. Proceedings of the IAEA Specialists Meeting, Madrid, Spain, (1999), pp. 374 - 385.

[12] Yu. Nikolaev *Radiation Embrittlement of Cr-Ni-Mo and Cr-Mo RPV steels*, Journal of ASTM International, Vol.4, №8, paper ID JAI 100695, (2007).

[13] A. Kryukov, D. Erak et al., *Extended analysis of VVER-1000 surveillance data*, International Journal of Pressure Vessels and Piping 79, (2002), pp.661-664.