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Division I

STUDY OF VVER-440 REACTOR PRESSURE VESSEL WELD METAL AFTER 45 YEARS OF OPERATION

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ABSTRACT

There are presented the results of a chemical composition study, mechanical characteristics and parameters of brittle fracture (ductile-to-brittle transition temperature and T_0) of metal samples cut from the irradiated weld No. 4 of the VVER-440/179 reactor vessel after 45 years of operation. The values of ductile-to-brittle transition temperature distribution over the wall thickness of VVER-440 reactor pressure vessel irradiated weld (140 mm) is obtained.

INTRODUCTION

The reactor pressure vessel (RPV) is one of the main barriers separating the external environment from the products of nuclear fuel fission. Monitoring the integrity of the reactor vessel is one of the most important tasks of ensuring the safe operation of a reactor plant .

In common practice, the control of changes of the reactor vessel metal properties is carried out with the help of surveillance specimens (SS), which are made of the same materials as the reactor vessel. Specimens are installed in the reactor, periodically unloaded, tested, which makes it possible to assess the process of the reactor vessel metal properties degradation during operation and predict property changes.

For VVER-440 reactor plant of the first generation (V-179), which was created in the 60s of the last century, a program of surveillance specimens was not provided, the actual values of the ductile-to-brittle transition temperature in the initial state were not determined. The phosphorus and copper content in the weld metal was not regulated and was not determined at the manufacturing stage. The absence of this information caused uncertainty in the assessment of radiation embrittlement and the need to anneal the reactor vessel to continue further operation.

Power unit No. 3 of the Novoronezh NPP (NVNPP-3) has been in operation since 1971. In 1987, the irradiated weld joint of the NVNPP-3 reactor vessel was subjected to recovery annealing at a temperature of 430 °C, and again in 1991 – at a temperature of 475 °C. After that, the NVNPP-3 reactor plant was operated for 25 years, and was shut down in 2016. In 2017, through samples (trepans) were cut from the reactor vessel of the NVNPP-3 to study metal properties after long-term impact of operational factors.

The absence of anticorrosive surfacing on the inner surface of the reactor vessel made it possible to control the reactor vessel metal properties after annealing by periodically cutting special samples from the inner surface of the reactor vessel (templets) [1-3]. The thickness of the templets allowed the production of only sub-sized specimens (3×4×27 mm for the base metal (BM) and 5×5×27.5 mm for the weld metal (WM)). The correctness of the use of sub-sized specimens was justified at the Kurchatov Institute [2, 4, 5].

In 2017, after the shutdown of the NVNPP-3 power unit, through-hole samples were cut out of several elements of the reactor vessel, in order to study the metal of the irradiated (core shell, weld No. 4) and non-irradiated (upper nozzle shell, lower nozzle shell, weld No. 6) elements of the reactor vessel (Figure 1).

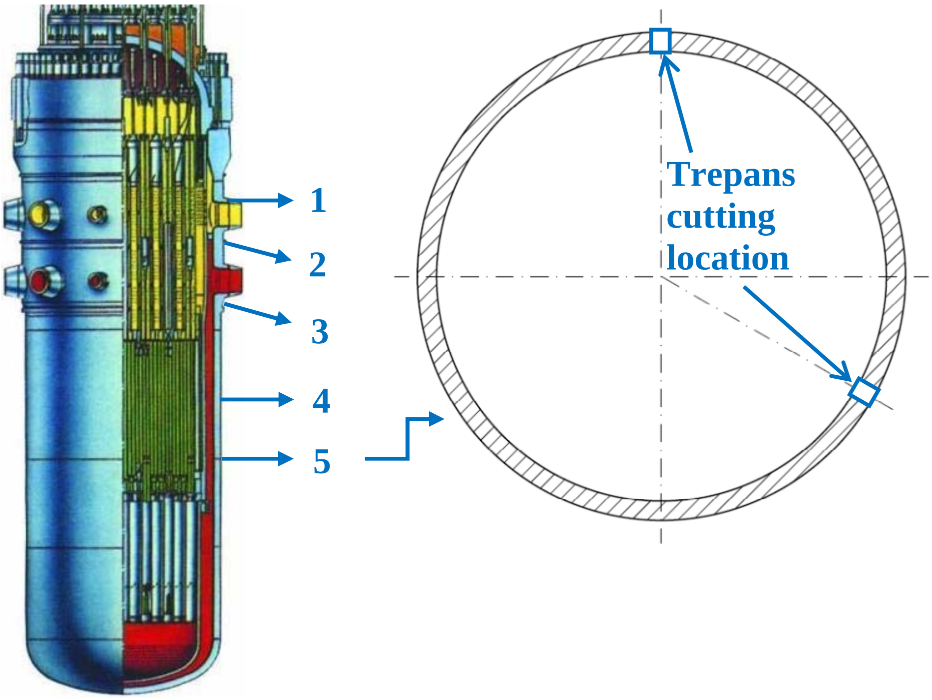

Figure 1. Diagram of the places where trepans are taken from the VVER–440 RPV and the cross section of the weld No. 4. 1 - upper nozzle shell, 2 - weld No. 6, 3 - lower nozzle shell, 4 - core shell, 5 - weld No. 4.

The metal of weld No. 4 of the VVER-440 reactor vessel connects the upper and lower shells of the core and is exposed to neutron irradiation. Due to the increased tendency to radiation embrittlement, the irradiated weld joint is the most critical element of the reactor vessel.

This paper presents the results of the study of metal trepans from irradiated weld no.4. An assessment was made of the correctness of determining the state of the RPV metal obtained during the study of the metal of templets cut directly from the inner surface of the reactor vessel VVER-440

MATERIALS AND RESEARCH METHODS

The weld No. 4 is made by automatic welding using Sv-10KHMFT welding wire under a layer of AN-42 flux. The root of the seam is welded with Sv-08A welding wire under the AN-42 flux. The condition of the weld metal under study is characterized by the following sequence: Irradiation (17 years) + Annealing 430°C/150 hours + Irradiation (3 years) + Annealing 475°C/150 hours + Irradiation (25 years). The irradiation temperature is 270 °C. Figure 2 shows information on the temperature regime of operation, including annealing of the irradiated weld joint of the NVNPP-3 RPV.

Figure 2 shows the scheme of changing the properties of the inner surface of the weld No. 4 metal during neutron irradiation and annealing.

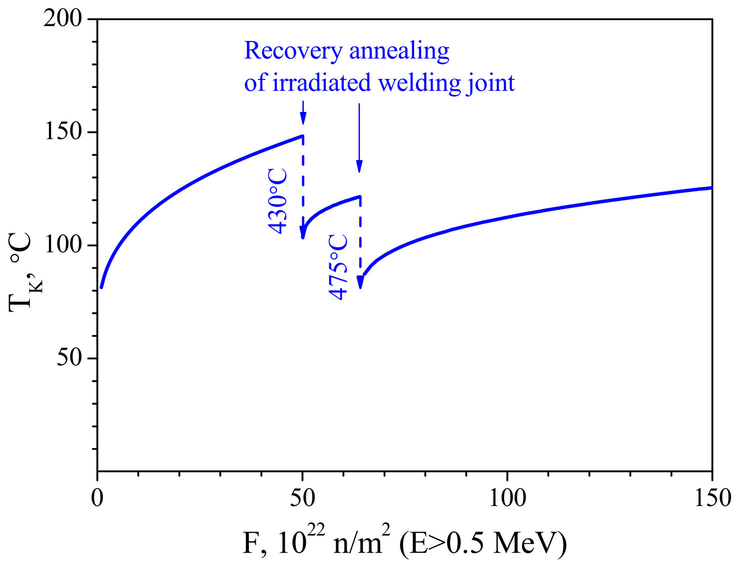

Figure 2. Diagram of radiation embrittlement of the NVNPP-3 RPV's weld No. 4 metal during operation and recovery annealing

Cylindrical trepans (∅110 mm, ~140 mm long) were cut from two places along the circumference of the weld No. 4 of the VVER-440 reactor vessel. The layout of the trepan selection sites is shown in Figure 1.

The chemical composition of the weld metal was determined on a longitudinal plate cut off from the trepan along the entire thickness of the weld. The detection of the weld boundaries was carried out in a 2% solution of nitric acid.

The content of alloying elements was determined by photoelectric spectral method on an optical emission analyzer PMI-MASTER UVR from the surface of the plate. After that, samples were cut off from the plate to determine the content of phosphorus and copper along the entire thickness of the weld in increments of 10 mm. The copper content was determined by inductively coupled plasma mass spectrometry using ICP MS "Elan DRC-e" PerkinElmer Instruments equipment, and the phosphorus content was determined by photometric method on a UV-2100UV/VIS RayLeigh spectrophotometer. The results of determination of phosphorus and copper content are presented in detail in the article [6].

To determine the mechanical characteristics of the weld metal, the trepans were cut into discs using electroerosion cutting (Figure 3).

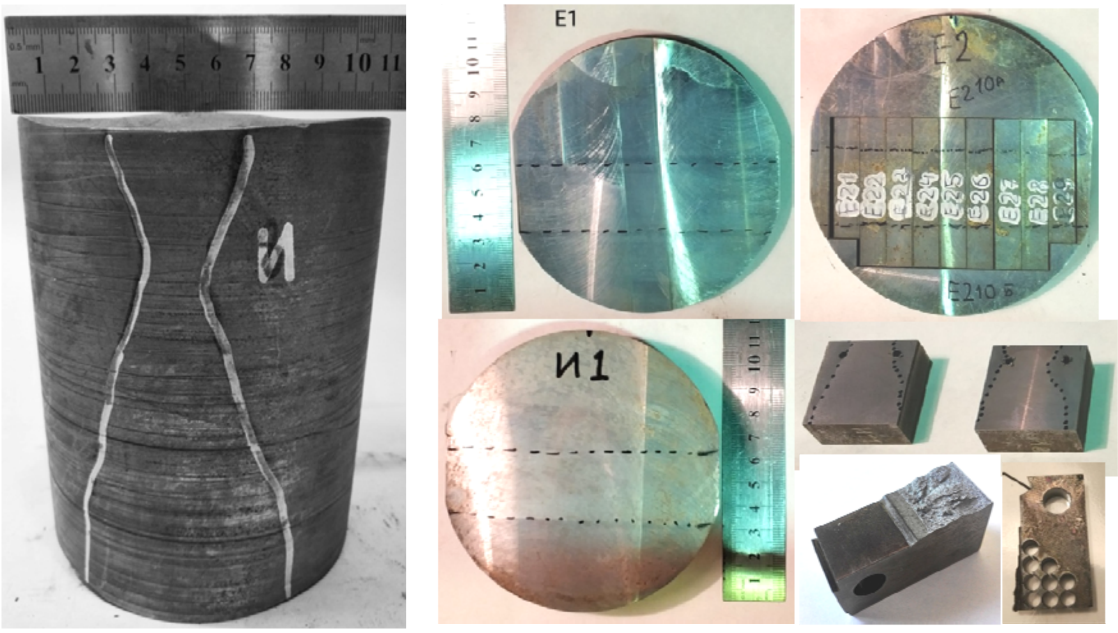

Figure 3. Photograph of trepans from weld No. 4, cut-out discs and specimens after grinding and etching. The weld boundary detected by etching is indicated by a marker.

After cutting, the weld boundary was revealed on each disk. This made it possible for each sample to place the test area directly in the weld weld metal. Specimens for impact bending of the Charpy type with dimensions of 10×10×55 mm, as well as smooth cylindrical 5-fold specimens with a diameter of the working part ∅3 mm for static tensile tests were made of trepan E from several layers in thickness. Specimens of CT-0,5 type - from the internal cutting of the weld (trepan И). Smooth cylindrical 5-fold static tensile specimens with a diameter of the working part ∅1.2 mm are made from a half of the tested sample CT-0,5 (Figure 3).

The trepan cutting scheme is shown in Figure 5. Each separate group of specimens was made from a single layer (disk) using reconstitution technology in accordance with [6]. For each group of specimens (layer), the value of the critical brittleness temperature (T_K) was determined based on the test results of 12 specimens. For the root of the weld, the results were determined based on the results of testing 9 specimens due to the limited volume of the weld metal.

Static tensile specimens are cut from three different layers of trepan E weld metal. The rectangles highlighted by solid lines in Figure 4 correspond to the place of cutting of each group of Charpy specimens. The circles indicate the cross section of the workpieces for tensile specimens. The incision on the Charpy and CT-0,5 specimens was made in the radial direction in the horizontal plane. The orientation of the specimens and incisions is the same as in the manufacture of surveillance specimens (SS) of VVER RPV.

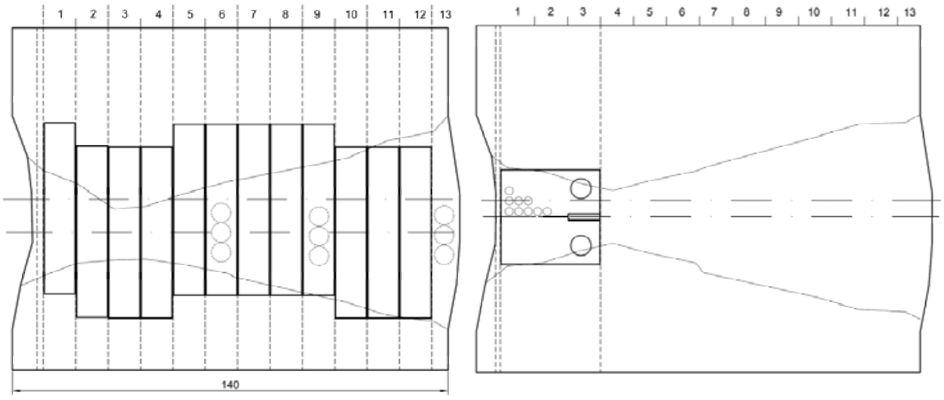

Figure 4. The scheme of specimens cutting of trepans E and И, bold lines indicate the orientation of the Charpy and CT-0,5 specimens, circles – tensile specimens.

Testing of specimens and processing of the results of impact bending tests were carried out in accordance with [7]. The specimens were tested in a materials science protective chamber on a pendulum RKP-300, which has a potential energy reserve of 300 J. The parameters of the temperature dependence of the fracture energy were estimated by the least squares method. The values of the critical temperature of fragility were determined using the criterion level of 47 J.

The tensile specimens were tested for uniaxial static stretching at room temperature on a Zwick/Roell Z030 universal testing machine with a capacity of 30 kN (Germany) at a loading speed of 2 mm/min to determine the strength characteristics ($R_{p0.2}$, R_m). Testing and processing of the results were carried out in accordance with [7, 8].

The fracture toughness tests of the specimens were carried out in accordance with [7] on a Zwick/Roell Z100 universal testing machine with a capacity of 100 kN (Germany). The initial fatigue cracks were grown on a resonant testing machine "RUMUL MICROTRON" (Switzerland) in the automatic mode of monitoring the load values, the number of cycles and the length of the crack. The stress intensity coefficient K_f did not exceed 12 MPa√m. The procedure for growing initial fatigue cracks for all specimens was carried out at room temperature.

RESULTS AND DISCUSSION

Chemical composition

Figures 5, 6 show the results of chemical analysis of the metal of trepans from the irradiated weld No. 4.

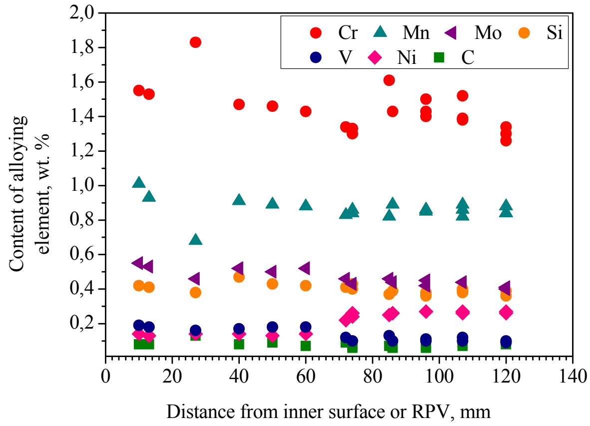

Figure 5. The content of alloying elements according to the thickness of weld No. 4

As shown in Figure 5 – the content of alloying elements is fairly uniform in the thickness of the weld. There is a local increase in the concentration of chromium and a decrease in the concentration of manganese in the area of the seam root, which, apparently, is associated with local mixing of the base metal and the seam metal during welding. This effect was also noted for unirradiated weld No. 6 [9].

Evaluation of radiation embrittlement of the metal of the NVNPP-3 reactor vessel after annealing required determination of the content of impurities: phosphorus and copper in the weld metal. And since these parameters were not determined during the manufacture of the reactor vessel, the values of phosphorus and copper obtained from the study of templets from the inner surface were determined to assess the radiation embrittlement of the metal of the reactor vessel. In Figure 6, these values are marked with a red dotted line.

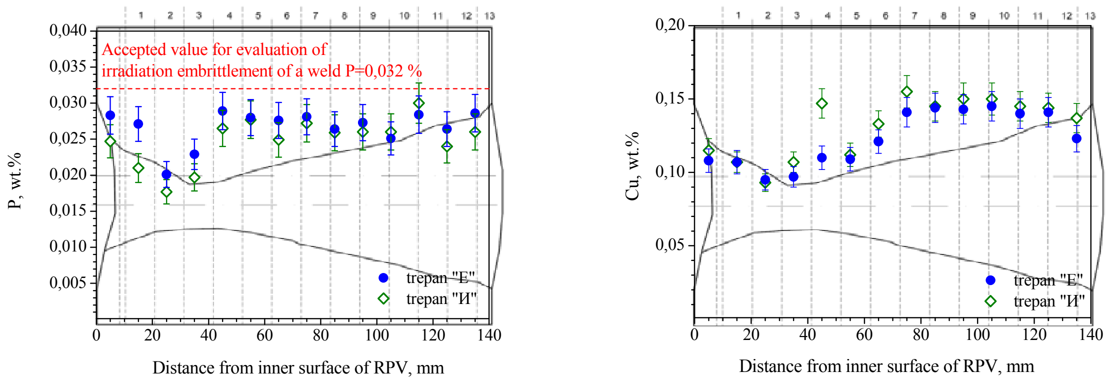

Figure 6. Copper and phosphorus content by weld No. 4 thickness.

As can be seen from Figure 6, the phosphorus and copper content are not heterogeneous in the thickness of the weld.

The copper content varies from 0.09 to 0.15% by weight. The copper content in the external cutting exceeds the value taken to assess the radiation embrittlement of WM No. 4 of the NVNPP-3 reactor vessel. However, after annealing, differences in copper content do not significantly affect the rate of radiation embrittlement for the following reason. During irradiation, copper is released from a solid solution with the formation of copper-enriched precipitates. During the reduction annealing, the entire volume of the weld metal is maintained at a temperature of 475 ± 15 °C for 150 hours. As a result of annealing, the equilibrium concentration of copper in a solid solution, which is determined by the heating temperature of the metal, becomes the same throughout the thickness of the weld. As shown in [14-17], for this temperature, the equilibrium concentration of copper in a solid solution does not exceed ~0.10%

by weight. Thus, during repeated irradiation after annealing, the factor that varies in the thickness of the weld and significantly affects radiation embrittlement is the phosphorus content in the weld metal.

The regularity of the distribution of phosphorus content over the thickness of the weld has a complex shape [6]. The phosphorus content is minimal at the root of the weld and increases with distance from it. The inner cutting (thickness ~25 mm) is shorter than the outer (100100 mm). In this regard, the maximum phosphorus content in the weld corresponds to the external cutting (at a distance of ~90 mm from the root of the seam P = 0.030% wt.). The phosphorus content in the weld metal No. 4 of the NVNPP-3 reactor vessel is in the range of ~0.018 to 0.030% by weight. A similar form of phosphorus distribution along the seam thickness, with a maximum on the outer cutting, was obtained by studying the metal of trepans from the welds of the first-generation VVER-440 reactor housings [6, 10-13].

According to the results of the study of the metal of the SS No. 4 templets from the inner surface of the reactor vessel, the phosphorus content P = 0.032% by weight was determined, adopted to predict the radiation embrittlement of the seam metal. The phosphorus values do not exceed P = 0.032% by weight over the entire thickness of the weld No. 4. Thus, the assessment of the phosphorus content based on the results of the templet study is conservative.

Evaluation of radiation embrittlement after annealing of the VVER-440 RPV's irradiated weld metal

The predicted value of the T_{KR} of the weld metal during repeated irradiation after annealing is determined by the dependence (1) in accordance with [15].

$$T_{\kappa R} = T_{\kappa A} + \left[646 \cdot (C_P - 0{,}02) \cdot e^{(1 - 0{,}01 \cdot T_{\kappa A})}\right] \cdot \left(\frac{F_R}{F_0}\right)^{0{,}36}, °C \qquad (1)$$

where T_{KA} is the value of the ductile-to-brittle transition temperature after recovery annealing, °C; C_P – phosphorus content in the weld metal, wt. %; F_R is the fluence of fast neutrons (E > 0,5 MeV) accumulated during re-irradiation after annealing, neutron/m^2; F_0 is a coefficient equal to 10^{22}, neutron/m^2.

The state of the metal of the irradiated weld during the operation of the reactor vessel is determined by the simultaneous influence of several factors: the accumulated fluence of fast neutrons, the phosphorus content and the initial value of T_K. Each of these parameters varies according to the thickness of the weld. The distribution of phosphorus over the thickness of the weld is described above and is shown in Figure 6. The distribution of the actual values of T_{K0} in the initial state is unknown.

Figure 7 show the distribution of experimental values of T_{KR} and yield strength over the thickness of weld No. 4 obtained in this work.

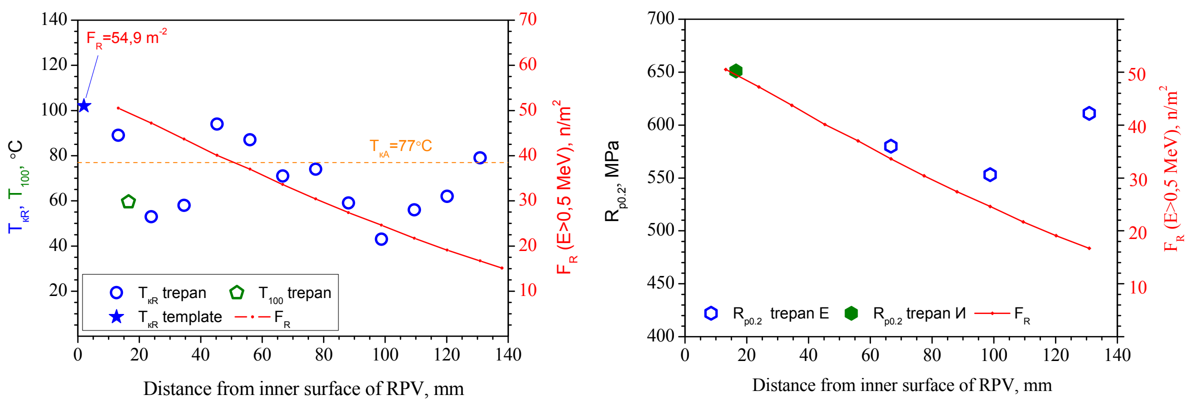

Figure 7 –Values of T_{KR} and T_{100} of weld metal No. 4 according to the results of the trepan metal study. T_{KA} is the value of the ductile-to-brittle transition temperature of metal after annealing. Distribution of yield strength values ($R_{p0.2}$) over the irradiated weld No. 4 thickness

As shown in Figure 7 accumulated fluence of fast neutrons varies naturally along the thickness of the weld, decreasing from inner to outer surface of RPV.

A solid line in Figure 7 shows the distribution of fast neutron fluence values over the thickness of the irradiated weld of the VVER-440 reactor vessel accumulated over 25 years of repeated irradiation after annealing. Fluence values are determined by computational and experimental method. The asterisk in Figure 7 shows the T_K value for the weld metal from the templet from the inner surface of the reactor vessel with a close value of accumulated fluence (the templet metal was tested after irradiation in the hull and additional accelerated additional radiation in the channels of the VVER-440 SS to F_R =54.9 n/m2). The maximum value of the critical brittleness temperature corresponds to the inner surface of the reactor vessel according to the results of the study of the templet metal after irradiation. When comparing the T_K results, one should take into account the fact that the fluence for a group of specimens from the templet is ~10% higher than the fluence for the group of specimens from trepan closest to the inner surface of the reactor vessel.

The study of the weld metal from trepans showed the following. The yield strength value is in the range of 553-651 MPa. T_{KR} values vary in thickness in the range from 43 to 94 ° C. The minimum value of T_{KR} (43 °C) is observed on the external cutting. In the area of the seam root, the T_{KR} value is 53 °C. The internal cutting corresponds to T_{KR} = 89 ° C. The maximum values of the critical temperature of brittleness are noted in the external cutting, at a distance of 20-30 mm from the root of the seam T_{KR} = 94 and 87 ° C. According to the test results of specimens of CT-0.5 from the internal cutting of trepan, the value T_{100} = 59 °C (F_R = 49.1 n/m^2) was determined, marked with a pentagon in Figure 7. The T_0 value is 30°C lower than the T_{KR} value for this seam area. The difference in neutron fluence values for groups of Charpy specimens from the inner seam layer and CT-0.5 specimens does not exceed 5%.

In Figure 7, the dashed line marks the value of the critical temperature of brittleness after recovery annealing T_{KA} = 77 ° C, obtained from the results of the study of the templet metal from the inner surface of the RPV. It can be seen that even after 45 years of operation, most of the T_{KR} values in the weld are below T_{KA}. This suggests that the initial values of T_K for different layers of weld metal before irradiation and after annealing are significantly different. Differences in the initial values of T_{K0} and differences in phosphorus content for different layers of the weld are likely to be the reasons for the increase in the values of T_{KR} in the metal of the external cutting of the weld, at a distance of 110-140 mm from the inner surface. As shown in [9], the heterogeneity of the properties along the thickness of the weld is determined by the complex form of the distribution of phosphorus contents along the thickness of the weld. This is an additional confirmation that it is the phosphorus content, all other things being equal, that is the most significant factor determining the properties of the metal of VVER-440 welds after reduction annealing. Thus, the distribution of T_{KR} values over the thickness of the weld after 45 years of operation has a complex shape. The reasons for this distribution of T_{KR} values is the simultaneous influence of several factors varying in the thickness of the seam. In this regard, the correlation between the values of T_{KR} and each individual factor (fast neutron fluence, phosphorus content) is insignificant.

Figure 8 shows the values of T_{KR} depending on the fluence accumulated after annealing for templet and trepan metal. A solid line shows the normative dependence used to assess the radiation embrittlement of the weld of the VVER-440 reactor vessel during repeated irradiation after annealing.

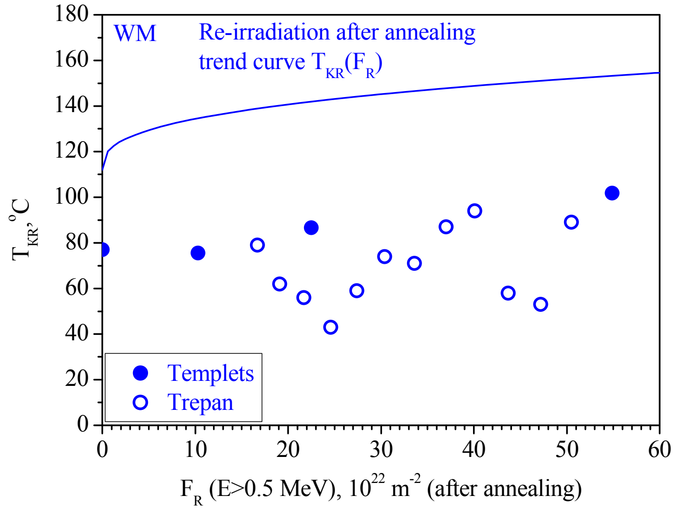

Figure 8. T_{KR} values for the metal of the VVER-440 RPV's irradiated weld, depending on the fluence after annealing (according to the test results of sub-sized specimens from templets and standard specimens from trepan)

For trepans cut out after 45 years of operation, the value of T_{KR} was determined by the results of testing standard specimens. For templets cut at different times from the inner surface of the RPV, the value of T_{KR} was determined by the results of testing sub-sized specimens. It can be seen that all the values of the T_{KR} of the weld metal obtained on trepans do not exceed the values obtained on templets. Thus, the assessment of the state of the metal after annealing, based on the results of sub–sized specimens from templets, conservatively and adequately describes the real state of the RPV metal.

The normative dependence used to assess the radiation embrittlement of the weld of the VVER-440 reactor vessel during repeated irradiation after annealing conservatively describes the actual state of the RPV metal.

CONCLUSION

A study of the weld metal No. 4 of the NVNPP-3 reactor vessel was performed after 45 years of operation. The content of chemical elements was determined by the thickness of the weld, including the content of phosphorus and copper. The values of T_K are determined over the entire thickness of the weld, the values of the yield strength and the crack resistance parameter T_0 on the inner surface of the irradiated weld are presented.

It is shown that the heterogeneity of properties along the thickness of the weld is largely determined by the complex shape of the distribution of phosphorus contents along the thickness of the weld, as well as by the accumulated fluence and the distribution of initial properties along the thickness. It is the phosphorus content, all other things being equal, that is the most significant factor determining the properties of the metal of VVER-440 welds under irradiation after reductive annealing.

It is shown that the system for monitoring the properties of the reactor vessel metal using specimens from samples taken from the inner surface of the reactor vessel is conservative and adequately describes the real state of the RPV metal.

It is shown that the normative dependence used to assess the radiation embrittlement of the weld of the VVER-440 reactor vessel during repeated irradiation after annealing conservatively describes the actual state of the RPV metal.

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