Improved determination of fast neutron fluence onto the WWER pressure vessel metal surveillance specimens

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Abstract. The special methodology for the determination of the neutron fluences onto the surveillance specimens of the WWER-1000 reactor pressure vessel metal has developed by INR NASU specialists and is successfully applied. The methodology is based on the Monte-Carlo code that is used for neutron transport calculations to the locations of the surveillance specimens. The methodology improvement is described. The fundamentals of the technique of the calculation-experimental determination of the fast neutron fluences onto surveillance specimens and their errors are presented. It is shown a dosimetry experiment in a particular reactor is needed to obtain experimental data about irradiation conditions in the locations of the surveillance specimens.

1 Introduction

The reactor pressure vessel (RPV) metal state monitoring during the whole operating period is one of the main conditions of the reliable and safe RPV operation. An important source of information about RPV metal property change is the surveillance program. Together with this, the results of the testing of the surveillance specimens (SS) are representative only in the case if the irradiation conditions of SS in the reactor are known with the required accuracy.

The specific nature of the SS locations in WWER-1000 reactors operated at Ukrainian NPPs does not allow to experimentally determine their irradiation conditions. The solution of this problem requires application of the special methodology based on calculation of the neutron transport to the SS location in the reactor. Such methodology has been developed in INR NASU and successfully applied on Ukrainian NPPs. It includes neutron transport calculations by the code MCSS [1] and the usage of experimental data that can be obtained after unloading of the specimens for testing.

The descriptions of WWER-1000 reactor regular SS program and the developed methodology are given in [2, 3].

This work is devoted to the development of used methodology with the purpose to increase validity of determination of the fast neutron fluence values onto SS by the direct

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usage of its experimental activities. Theoretical substantiation of the technique is presented in the next chapter.

2 The fundamental principles of calculation-experimental determination of neutron fluences onto SS and their errors

It is known that the activity of a product of any activation reaction at the moment of the end of irradiation is calculated using the following formula [4]:

$$A = \lambda N \int_0^T dt \cdot e^{-\lambda(T-t)} \int_0^\infty dE_n \cdot \varphi(E_n, t) \cdot \sigma(E_n), \tag{1}$$

where λ – the decay constant of activation reaction product;

N – the number of activated isotope nuclei;

T – the total time of irradiation;

$\varphi(E_n, t)$ – the energy-time distribution of neutron flux density;

$\sigma(E_n)$ – the activation reaction cross-section for neutrons with the energy E_n.

Taking into consideration urgently weak change of fast neutron spectrum in time in the SS location the expression (1) can be written in such form:

$$A = \alpha \cdot N \cdot \Phi \cdot \sigma^{eff}, \tag{2}$$

where $\alpha = \dfrac{1 - e^{-\lambda T}}{T}$;

$\Phi = \int_0^T dt \int_E^\infty dE_n \cdot \varphi(E_n, t)$ – the fluence of neutrons with higher energy than setting conditional energy threshold *E;*

$\sigma^{eff} = \dfrac{\int_0^T dt \int_0^\infty dE_n \cdot \varphi(E_n, t) \cdot \sigma(E_n)}{\int_0^T dt \int_E^\infty dE_n \cdot \varphi(E_n, t)}$ – the effective activation reaction cross-section for setting conditional energy threshold *E*.

The equality (2) is fair both for real values and for calculated ones (further an inferior indices *r* and *c* will be used for indication of real and calculated values respectively). Therefore:

$$\frac{A_c}{A_r} = \frac{\alpha_c}{\alpha_r} \cdot \frac{N_c}{N_r} \cdot \frac{\Phi_c}{\Phi_r} \cdot \frac{\sigma_c^{eff}}{\sigma_r^{eff}}. \tag{3}$$

In mathematical statistics it is postulated that values of any quantities that are obtained through observation (this term underpins any way of value obtaining, for example, measurement or computer modelling) are evaluations of real values [5]. Taking into account that the evaluation of real value A_r is a measured value A_e, such result is obtained from (3):

$$\Phi_r = \left(\frac{A_e}{A_c}\right) \cdot \Phi_c \tag{4}$$

This ratio is fair for any spatial point. Obviously, in some spatial region *V* ratios of calculated activity values and experimentally obtained ones must be equal in a close approximation, and the difference is due to uncertainties of their distribution. The principle of this region choice will be further provided.

An average ratio of calculated activity values and experimentally obtained ones for the spatial region V is:

$$R = \frac{1}{N_V} \sum_{(x,y,z)\in V} \frac{A_e(x,y,z)}{A_c(x,y,z)} . \tag{5}$$

where N_V – the general number of detectors of the same type in the region V .

Consequently, the formula (4) for this region has such form:

$$\Phi_r = R \cdot \Phi_c . \tag{6}$$

Using the methods of interval estimation instead of point one it is possible to obtain the following formula for calculation the relative errors of real neutron fluence within the spatial region V:

$$\frac{\Delta\Phi_r}{\Phi_r} = \sqrt{\left(\frac{\Delta\Phi_c}{\Phi_c}\right)^2 + \left(\frac{\Delta A_e}{A_e}\right)^2 + \left(\frac{\Delta R}{R}\right)^2 + \left(\frac{\Delta\sigma^{eff}}{\sigma^{eff}}\right)^2} . \tag{7}$$

Additive components are going to be analysed separately.

The first ratio $\frac{\Delta\Phi_c}{\Phi_c}$ is the relative statistical calculation error, its value is one for all spatial points in the region V.

The second ratio $\frac{\Delta A_e}{A_e}$ is the relative measurement error. All its components have been described in the research [6]. Although this relationship value is generally individual for each irradiated neutron activation detector (NAD), the differences are small for detectors of the same type, that allows to use the averaged value.

In the third ratio the quantity ΔR is calculated for the spatial region V by a standard formula:

$$\Delta R = \sqrt{\frac{1}{N_V - 1} \sum_{(x,y,z)\in V} \left(\frac{A_e(x,y,z)}{A_c(x,y,z)} - R\right)^2} . \tag{8}$$

For finding an effective cross-section σ^{eff} the results of the neutron transport calculation into the region V are used. This region is chosen in such a way that the differences from the normalized calculation spectrum would be relatively small. Therefore, the applied method can be observed as a modification of the method based on the minimization of the effective cross-section variation for arbitrary small variation of a setting spectrum [7]. However, in this case the averaged spectrum in the region V plays the role of the setting spectrum. From a statistical point of view, the set of this spectra and the averaged spectrum can be respectively interpreted as a random sample and as a sample average that makes obvious the calculations of both the effective cross-section itself and its error $\Delta\sigma^{eff}$.

It is obvious from the point of general physics and proved by mathematical modelling that the last component in the formula (7) has the strongest impact on the real fluence error value. It grows rapidly if the setting energy threshold of activation reaction is moved away from the effective value (see Fig. 1).

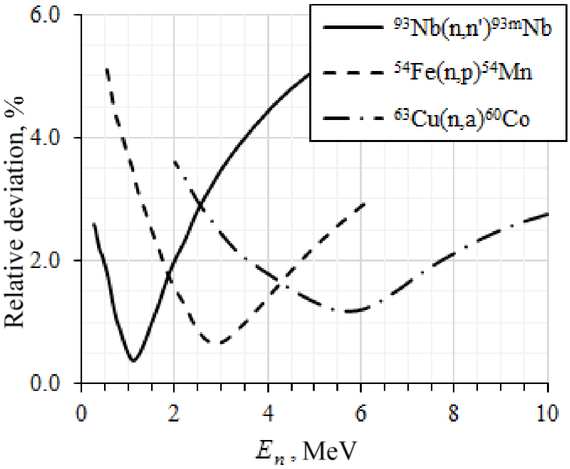

Fig. 1. Dependence of the relative deviation of the effective cross-section ($\Delta\sigma^{eff}/\sigma^{eff}$) from the specified energy threshold for various reactions used in the dosimetry SS of the WWER RPV metal.

According to the demands of the regulatory documents the knowledge of fluences with $E_n > 0.5$ MeV onto SS is required.

Dosimetry maintenance of the SS program realized on reactors WWER-440 and WWER-1000 of the Ukrainian NPPs includes irradiation of NAD from niobium, iron and copper. As it is observed from Fig. 1 only usage of activation reaction $^{93}\mathrm{Nb}(\mathrm{n,n'})^{93\mathrm{m}}\mathrm{Nb}$ for the determination of the real values of the neutron fluence with $E_n > 0.5$ MeV and of their errors may give an appropriate result.

Theoretical conclusions and developed technique usage possibility were proved during determining of the neutron fluences onto the SS of WWER-440 RPV metal. The obtained results are presented in the next chapter.

3 Determination of neutron fluences onto SS irradiated in the reactor WWER-440

Within an WWER-440 RPV metal SS additional program realized on RNPP-1 [8] chains with specimens were located opposite core in channels for SS on external surface of barrel.

The SS set IA-ЛК-3 that includes two container garlands 3-1 and 3-2 was loaded into the reactor after annealing its pressure vessel amidst the 28th fuel cycle. It removed and delivered to INR NASU for experiments after the end of 32th cycle.

With the help of the special methodology that is similar to one that is used for container assemblies in the reactor WWER-1000 [9] the orientation of each container towards the core was determined. The obtained data was taken into account in the calculating model of the reactor WWER-440 of RNPP-1.

For dosimetry maintenance in several containers with SS the capsules with NAD were installed. There were 36 capsules, each of which included the NAD of niobium, iron and copper. After removal the SS set IA-ЛК-3 from the reactor and container breaching spectrometric measurements of irradiated NADs were performed.

The results of comparison experimentally obtained values of activation reaction product activities with calculated ones $R(V)$ and their errors $\Delta R(V)$ (see formulas (5) and (8)) are presented in Table 1, and obtained values of fluence relative errors $\frac{\Delta\Phi_r^{0.5}}{\Phi_r^{0.5}}$ (see formula (7)) are presented in Table 2.

Table 1. The average ratios of experimentally obtained and calculated specific activities of the products of NAD activation reactions that accompanied the irradiation of the SS in the RNPP-1 reactor, and their standard deviations.

№ chains with specimens	**Activation reaction**		
	$^{93}\mathrm{Nb}(\mathrm{n,n'})^{93\mathrm{m}}\mathrm{Nb}$	$^{54}\mathrm{Fe}(\mathrm{n,p})^{54}\mathrm{Mn}$	$^{63}\mathrm{Cu}(\mathrm{n},\alpha)^{60}\mathrm{Co}$
1-1*	1.06 ± 0.13	0.96 ± 0.13	0.99 ± 0.10
3-1	1.01 ± 0.03	0.94 ± 0.04	1.02 ± 0.04
3-2	1.01 ± 0.02	0.94 ± 0.05	1.02 ± 0.05

* – experimental data were obtained by specialists of NRC "Kurchatov Institute" and presented in [10].

Table 2. Relative errors (%) of real neutron fluence, that were obtained with using the results of measurements of different types of NAD, which accompanied the irradiation of the SS in the RNPP-1 reactor.

№ chains with specimens	**Activation reaction**		
	$^{93}\mathrm{Nb}(\mathrm{n,n'})^{93\mathrm{m}}\mathrm{Nb}$	$^{54}\mathrm{Fe}(\mathrm{n,p})^{54}\mathrm{Mn}$	$^{63}\mathrm{Cu}(\mathrm{n},\alpha)^{60}\mathrm{Co}$
1-1*	15.3	22.3	22.4
3-1	9.7	18.2	20.3
3-2	9.3	18.4	20.5

* – experimental data were obtained by specialists of NRC "Kurchatov Institute" and presented in [10].

Obviously larger dispersion of ratios of experimental and calculated data for the chain 1-1 is caused by the fact that the specialists of NRC "Kurchatov Institute" could not orient containers with SS. As a result, it leads to the visibly bigger value of the real fluence error.

As it is clear from Table 2 the error value of fluence determination with the usage of activation reaction $^{93}\mathrm{Nb}(\mathrm{n,n'})^{93\mathrm{m}}\mathrm{Nb}$ product measurement results is much smaller than with the usage of activation reaction $^{54}\mathrm{Fe}(\mathrm{n,p})^{54}\mathrm{Mn}$ or $^{63}\mathrm{Cu}(\mathrm{n},\alpha)^{60}\mathrm{Co}$ product measurement results that proves the conclusions from the previous chapter.

4 The determination of neutron fluence onto SS irradiated in the reactor WWER-1000

Dosimetry maintenance of the operating SS program realized on reactors WWER-1000 of the NPPs in Ukraine has the series of essential disadvantages. First of all, this is a failed position of capsules with NAD away from SS center in round double-floor container assemblies, detectors are completely absent in single-floor assemblies. After removal the first assemblies from the reactor it appeared that niobium detectors were destroyed and became practically useless for spectrometric measurements. In fact, it means that reliable experimental data can be obtained only from spectrometric measurements of irradiated SS.

It is practically possible to measure only specific activity of the reaction $^{54}\mathrm{Fe}(\mathrm{n,p})^{54}\mathrm{Mn}$ product in SS. However, as it has been described before, the direct usage of such measurement results for obtaining the real values of the neutron fluence with $E_\mathrm{n} > 0.5$ MeV by formula (6) leads to a big error by formula (7).

To solve the problem the recalculation coefficient k obtained on the basis of measured specific activities of $^{93\mathrm{m}}\mathrm{Nb}$ and $^{54}\mathrm{Mn}$ in NADs located in the same reactor region V can be used. In this case:

$$k = \frac{1}{N_\mathrm{NAD}} \sum_{(x,y,z)\in V} \frac{\left(\dfrac{A_e^\mathrm{Nb}(x,y,z)}{A_c^\mathrm{Nb}(x,y,z)}\right)_\mathrm{NAD}}{\left(\dfrac{A_e^\mathrm{Fe}(x,y,z)}{A_c^\mathrm{Fe}(x,y,z)}\right)_\mathrm{NAD}},$$

where $\left(\dfrac{A_e^\mathrm{Nb}(x,y,z)}{A_c^\mathrm{Nb}(x,y,z)}\right)_\mathrm{NAD}$ and $\left(\dfrac{A_e^\mathrm{Fe}(x,y,z)}{A_c^\mathrm{Fe}(x,y,z)}\right)_\mathrm{NAD}$ – is the ratio of experimentally obtained and calculated specific activities of the products of the activation reaction $^{93}\mathrm{Nb}(\mathrm{n,n'})^{93\mathrm{m}}\mathrm{Nb}$ and $^{54}\mathrm{Fe}(\mathrm{n,p})^{54}\mathrm{Mn}$ respectively;

N_NAD – the number of pairs of NADs from niobium and iron in the region V .

Taking into account the presented information, formula (5) and (8) can be rewritten in such form:

$$\Phi_r^{0.5}(x,y,z) = k \cdot R^\mathrm{Fe} \cdot \Phi_c^{0.5}(x,y,z),$$

$$\frac{\Delta\Phi_r^{0.5}}{\Phi_r^{0.5}} = \sqrt{\left(\frac{\Delta\Phi_c^{0.5}}{\Phi_c^{0.5}}\right)^2 + \left(\frac{\Delta A_e^\mathrm{Fe}}{A_e^\mathrm{Fe}}\right)^2 + \left(\frac{\Delta R^\mathrm{Fe}}{R^\mathrm{Fe}}\right)^2 + \left(\frac{\Delta\sigma^{eff}}{\sigma^{eff}}\right)^2 + \left(\frac{\Delta k}{k}\right)^2}.$$

Therefore, for determination of real value of neutron fluence onto SS of WWER-1000 pressure vessel metal and its error, the knowledge of the average ratio value of experimentally obtained and calculated specific activities of said activation reaction products is required.

To provide such information obtaining the dosimetry experiment, in other words, a complex of researches for experimental determination of neutron field characteristics in SS locations is required. Such experiments have already been performed on the SUNPP-1, RNPP-3 and ZNPP-4.

As an example, in Table 3 values k and Δk were provided for different floors of various container assemblies irradiated during dosimetry experiment on the RNPP-3 reactor.

Table 3. The average values of recalculation coefficients and their standard deviations for different floors of various container assemblies irradiated during dosimetry experiment on the RNPP-3 reactor.

Floor	Index of container assembly		
	M1	M2	M3
Upper	1.24 ± 0.08	1.15 ± 0.04	1.13 ± 0.06
Lower	1.19 ± 0.05	1.20 ± 0.07	1.12 ± 0.06

The Table 3 shows that in case calculations are performed by the program MCSS [1] all the container assemblies can be included in the region V and instead of different recalculation coefficients only the average value 1.17 ± 0.07 is acceptable to use. In accordance with our evaluations, in this case the error value of the real fluence determination onto SS of the WWER-1000 pressure vessel metal amounts approximately 11-12%.

Together with this it should be noticed that the problem of recalculation coefficient equality for different assemblies and different reactors stays uncompleted and demands additional research by the way of dosimetry experiments performance.

5 Conclusions

1. Technique of calculating-experimental determination of the real values of neutron fluences with $E_n > 0.5$ MeV onto SS of WWER-1000 pressure vessel metal and their errors has been established and tested.

2. In the case the direct usage of activation reaction product measurement results for determination of real fluence values and their errors only the reaction $^{93}\mathrm{Nb}(\mathrm{n},\mathrm{n}')^{93\mathrm{m}}\mathrm{Nb}$ can provide the appropriate result.

3. In the case the measurement of the niobium is impossible it is offered to use the results of $^{54}\mathrm{Fe}(\mathrm{n},\mathrm{p})^{54}\mathrm{Mn}$ activation reaction product measurements directly from SS as well as recalculation coefficient obtained within the dosimetry experiment.

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