Neutrron dosimetry intercomparison run for verification of the neutron fluence

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1. Introduction

When neutron flux with energy distribution close to so called "fission spectra" passes through a solid mainly four nuclear reactions are observed:

- (n,γ), when the final nuclei is an isotope (often radionuclide) of the target nuclei, as for instance $^{54}Fe(n,\gamma)^{55}Fe$;
- n => n', non-elastic scattering – leads only to excitation of the nuclei and often to formation of isomers, as for instance $^{93}Nb(n,n')^{93m}Nb$;
- n => p, conversion of the target nuclei to a nuclei with 1 charge less and the same number of nucleons in it (isobar) as for instance $^{54}Fe(n,p)^{54}Mn$;
- n => α, conversion of the target nuclei to a nuclei with 2 charges and 3 nucleons less as for instance $^{63}Cu(n,\alpha)^{60}Co$.

The first reaction is characteristic for lower energy neutrons (so called "thermal neutrons"). For the materialization of the next three reactions neutrons need higher energies. The probability for interaction sharply increases around some characteristic for the respective reaction energy of the neutrons, called "reaction threshold". Such reactions are called in general "threshold reactions"

When thresholds are above 0.5 MeV final nuclei obtains a significant kinetic energy and the respective atoms easily leave their places in the crystal lattice of the solid they belong to. In this way the number of defects in volume or mass unit increases above the equilibrium value, typical for the solid temperature. During a prolonged irradiation this non-equilibrium can reach a significant magnitude due to which the mechanical properties of the solid body could change considerably.

In the case of the steel for reactor pressure vessel the change of the mechanical properties is manifested in so called "embrittlement" of the steel. The "embrittled" steel does not stand large mechanical stresses and in case of appearance of such stresses some cracks in the material appear and even destruction of the steel may occur. In the reactor pressure vessels such stresses may arise in case of sharp change of the temperature of the water for technological or especially accident reasons.

The importance of these processes for the safe operation of the reactors is doubtless at the moment. With the design stage conditions are created for realization of a "surveillance program" during the operation. In such a way are designed VVER 1000 reactors in Kozloduy, so with the help of these programs there is a possibility to evaluate the life time resource of the vessel. Because in VVER 440 reactors such programs are not designed a special "ex-vessel" experiments are organised or exceptional actions are undertaken such as cutting of tamplets from the inner lining of the vessel at weld 4 of unit 1 in NPP "Kozloduy".

A key quantity in evaluation of the life time resource for both types of reactors is the maximal value of "neutron fluence" with neutron energy higher than 0.5 MeV, passed through the reactor vessel. For the determination of the fluence a complicated computer calculations are carried out of neutron transport. For the verification of the calculations measurements of “neutron monitors” are carried out which are placed in the surveillance sets as well as in the ex-vessel experiments. The calculated by means of computed fluence activities of “neutron monitors” are compared to the same activities, obtained by means of experimental measurement of the monitors. The experimental measurement is a complicated process, each step of which is connected to a certain statistical error. For estimation of the reliability of experimental measurements of neutron monitors an international intercomparison runs are organized. In this work results are presented of INRNE participation in two such runs”: first, organized by SEC NRS (Scientific and Engineering Center for Nuclear and Radiation Safety, Moscow, Russia on the unit 3 of Balakovo NPP and second one, organized in the frame of TACIS program by Research Institute for Nuclear Reactors (NIIAR), Dimitrovgrad, Russia on the irradiation facility KORPUS.

The neutron monitors are present in design sets for irradiation in VVER 1000 as well as in the ex-vessel experiments, carried out on both types of reactors. The most often/widely used sets consist of:

Nb monitor. The activity of ^{93m} Nb, obtained by the reaction ^{93}Nb(n,n’)^{93m}Nb is measured. In the nature ^{93} Nb is 100 % distributed. Its testimony is considered as especially important, because the reaction threshold is 0.5 Mev and for this reason activity measured is to the highest degree proportional to the embrittlement in comparison to the other two monitors;

Fe monitor. The activity of 54 Mn obtained by the reaction ^{54}Fe(n,p)^{54}Mn is measured. In the natural Fe mixture 5.9 % are of ^{54}Fe. The reaction threshold is 2.5 *MeV*. In some cases Fe monitors are used, enriched with ^{54} Fe for higher sensitivity;

Cu monitor. The activity of 60Co, obtained by the reaction 63Cu(n, α) $^{60Co.}$ In the nature ^{63}Cu is distributed 69.17 %. The reaction threshold is 6.0 *MeV*. Again in this case enriched with ^{63}Cu monitors could be used for higher sensitivity.

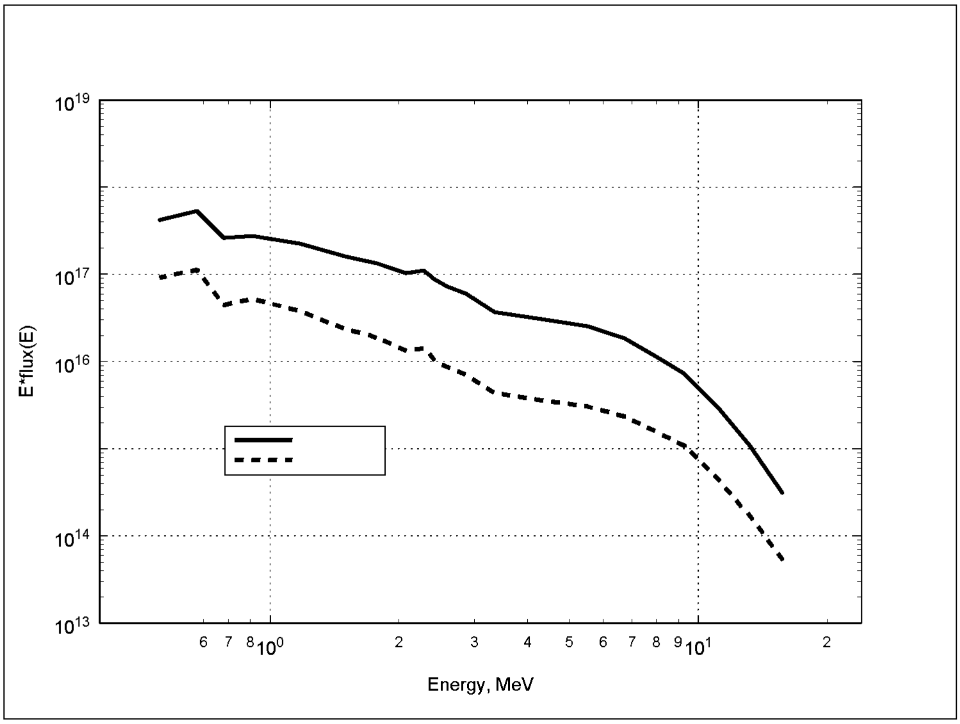

Fig. 1 Energy distribution of the neutrons in “ex-vessel” position

On fig. 1 a spectra is presented from which it can be seen that the most often used monitors practically cover the whole energy range of neutrons in "ex-vessel" position.

2. International Intercomparison Run Balakovo

The organized by SEC NRS intercomparison run was executed in two stages. The first consisted of irradiation on unit 3 of Balakovo NPP of sets of neutron dosimeters (monitors), committed by the participants and determination of the activities in their laboratories. The considerable differences in the niobium results necessitated the execution of the second stage – measurement of the activity of the same Nb foils by different participants involving additional experts. INRNE participated in the second stage. The two Nb foil received by INRNE were irradiated for one cycle on unit 3 of Balakovo NPP. The results of the second stage were published in [1] and reported at 10th ISRD'99 meeting in Osaka, Japan.

The results of the analysis of both Nb foils are presented in table 1.

Tabl. 1.

Foil # B2

Particip.	method	EOIA Bq/g	Uncert. %	EOIA, Bq/ncl	Uncert %	RR	Uncert. %
VTT (Fin)	solution	50289	3,0 (1s)	7.755E-18		3,930E-16	4%(1s)
FZR (Ger)	solution	48645	3,8 (2s)	7,501E-18		3,865E-16	
ECN (Hol)	solution	47940	2,3 (?)	7,392E-18			
SKODA (Czech)	intact	52075		8,030E-18	4,9 (?)		
INRNE (Bul)	solution	45100	4,0 (1s)	6,955E-18	4,0 (1s)	3,540E-16	4,7 (1s)
Mean				**7,527E-18**	**5,4**		

Foil # B3

Particip.	method	EOIA Bq/g	Uncert. %	EOIA, Bq/ncl	Uncet. %	RR	Uncert. %
VTT (Fin)	solution	25391	3,0 (1s)	3.915E-18		1,984E-16	4%(1s)
FZR (Ger)	solution	30912	3,8 (2s)	4,767E-18		2,456E-16	
INRNE (Bul)	solution	22700	3,0 (1s)	3,500E-18	3,0 (1s)	1,782E-16	3,9 (1s)
Mean				**4,061E-18**	**15,9**		

where:

EOIA, Bq/g – is the specific activity of ^{93m}Nb at the end of irradiation;
EOIA, Bq/ncl – is the specific activity for one nucleus, obtained by deleting by the quantity 6.485E+21 nucl/g, characteristic for Nb.
RR – reaction rate

For the foil # B2 our result is lower than average by 7.6 %, for the foil #b3 – by 13.8 %, or an average shift – 10.7 % with standard uncertainty about 3,1 %, which coincide with the declared by us standard uncertainty of our results. It can be seen from the table that the results of other participants also are not homogeneous – the standard uncertainty of the average reaches 16 %. This probably is due to the problems with non-homogeneity of the flux through the foil, which is connected to the foil dimensions – this effect is still not clarified. The results also show that Nb calibration is a problem for all participants. So it is obvious that problems must be solved and such international intercomparison runs must be carried out in the future.

3. International Intercomparison Run KORPUS

The KORPUS facility is mounted on RBT-6 reactor in NIIAR, Dimitrovgrad, Russia for irradiation of material samples in conditions, close to the real irradiation conditions. During the experiment in 1999 4 series of irradiation's were carried out at power close to the nominal 6 MWt, where in 4th seria were irradiated monitors of NIIAR as well as sets of INRNE - Bulgaria and FZR – Germany. The activity of all monitors was measured in NIIAR (only Fe monitor was measured from Bulgarian set). Afterwards sets were send to the participants for their own measurements. The results are presented in [2] and shown in tables 2 and 3.

Табле 2. Characteristics of the monitors, used in seria 4

	monitor	dimension mm	mass, mg	reaction	mass. % of the target	nucleus/mg target
INRNE	Cu	granules	292.3	^{63}Cu(n,α)^{60}Co	100	6.548E+18
	Nb	5x5x0.13	27.0	^{93}Nb(n,n')^{93m}Nb	100	6.482E+18
	Fe	granules	95.1	^{54}Fe(n,p)^{54}Mn	100	6.257E+18
FZR	Cu	Ø4x5	563.68	^{63}Cu(n,α)^{60}Co	100	6.548E+18
	Nb	Ø4x0.018	2.039	^{93}Nb(n,n')^{93m}Nb ^{93}Nb(n,γ)^{94}Nb	100	6.482E+18
	Fe	Ø4x1	59.94	^{54}Fe(n,p)^{54}Mn	2.12	1.327E+17
НИИАР	Nb11	Ø 10x0.1	62.83	^{93}Nb(n,n')^{93m}Nb ^{93}Nb(n,γ)^{94}Nb	100	6.482E+18
	Cu	Ø 10x0.5	352.41	^{63}Cu(n,α)^{60}Co	^{63}Cu 99.6	9.52E+18
	Fe	Ø 10x0.3	170.87	^{54}Fe(n,p)^{54}Mn	100	6.257E+18

Table 4. Results of the measurements in seria 4

set	monitor	RIIAR			FZR			INRNE		
		A_0,Bq	RR	σ,%	A_0,Bq	RR	σ,%	A_0,Bq	RR	σ,%
FZR	Cu	2.37 E+02	8.92 E-18	3.08	2.09E +02	7.87 E-18	3			
	Fe	3.16 E+01	9.15 E-16	8.3	3.00 E+01	8.71 E-16	3			
	Nb	1.12 E+02	3.60 E-15	4.2	1.06E +02	3.43 E-15	5			
INRNE	Cu*	2.01 E+02	8.36 E_18	2.1				1.15E +02	8.38 E-18	3.5
	Nb*	3.12 E+03	3.26 E-15	3.9				1.36E +03	3.31 E-15	4.4
	Fe*	2.47 E+03	9.56 E-16	1.8				2.48E +03	9.598 E-16	3.9

Where:
A_0 – activity of the monitor at the end of irradiation, Bq,
RR – reaction rate
σ – uncertainty of RR for confidence interval 0.68, %.
* - for copper, niobium and ferrum monitors results are presented from the measurements of RIIAR monitors which were irradiated in the same set as INRNE monitors (same position and fluence)

4. Conclusion

The participation in the international intercomparison runs showed that in order to more precisely verify the calculated values of the neutron fluence more intercomparison exercises are necessary. Due to such exercises our results improved after calibration of Nb performed and are in a very good agreement with RIIAR results in spite of the different approaches in the determination of its activity.

References

1. Borodkin, G.I. et all, **“Balakovo-3 Ex-vessel Exercise: Interomparison of Results”,** *Reactor Dosimetry, ASTM STP 1398*, John G. Williams, David W. Vehar, Frank H. Ruddy and David M. Gilliam, Eds., American Society for Testing and Materials, West Conshohoken, PA, 2000.
2. Technical Report SRR 295/GD/03/01 of AO “Atomenergoexport”, RNC “Kurchatov Institute” on “Development of modernized methods of evaluation of radiation embrittlement of reactor vessel”.