EVALUATION OF THE ACTIVATION AND BURN-UP EXPERIMENTS CARRIED OUT IN BN-350 REACTOR

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ABSTRACT

Review of the experimental work on the ratio measurement of cross sections of different nuclides included into the nuclear fuel composition and structure materials and material composition variations of different types of fuel and actinide samples under the influence of fast reactor neutron irradiation is presented in the report. Brief characteristics of the experimental methods applied, experimental programs performed, fuel and actinide samples researched are presented. Statement of problem is given as well as some results of the project on the estimated data bank development maximally accounting the whole set of experimental results obtained in BN-350 reactor adjusted to conditions of specially developed benchmark models.

1. INTRODUCTION

Within sequence of years a great number of physical experiments was carried out in BN-350 reactor. Part of these experiments was devoted to the BN-350 reactor reliable and safe operation. However, the main part of experiments had fundamental character and their results were of unique importance [1-3]. A series of burn-up experiments were carried out with pure actinide samples (Th, Pa, U, Np, Pu, Am, Cm), fuel samples and structure elements irradiated in the reactor core and blanket for the purpose of the further radiochemical research. The other set of experiments was connected with the study of BN-350 reactor nuclear and physical characteristics. The experimental data analysis and compilation are currently carried out in order to develop the database supporting the International programs on MA transmutation and weapon-grade plutonium utilization and for calculation code and nuclear date verification as well.

2. EXPERIMENTAL METHODS

Method of activation disc detectors with the use of measuring fuel assemblies. Historically it was the first method being applied in BN-350. Special fuel assemblies with the central removable container have been manufactured for the measuring. The sets of U-235, U-238, Pu-239, etc. disc detectors were put into container. Detectors were irradiated at the minimal controlled power level (about 1MW) during several hours (5-10 hours). After that the

indicators were removed from the fuel assembly and the induced activity analysis was carried out with the help of scintillation detectors first and then with semi-conducting ones based on Ge(Li). The measurement results were the ratios between neutron reaction cross sections and reaction rate distributions in radial and axial directions.

Method of the "needle" detectors irradiated in the inter-element space of the fuel assembly. This unique method has been developed for the neutron field investigation in standard fuel assemblies of BN-350 reactor with the minimal disturbances. It has been found out that thin stainless steel capillaries can be inserted into the inter-element space of fuel assembly without noticeable deterioration of thermal exchange conditions. That's why the sets of experimental samples of 1.2mm diameter have been fabricated and consecutively loaded into capillary of 1.6mm diameter. The capillary was inserted into the inter-element gap and fastened in the head part of the fuel assembly for irradiation period. As a rule a sample of high-enriched uranium-235 (90% enrichment) was loaded into capillary and was used as a neutron fluence monitor. In order to determine the control sample location in the fuel assembly the iridium-192 radioactive nuclide was loaded into the bottom part of the pipe. The actinide sample arrangement in the fuel assembly irradiated in the 243th cell of reactor is shown on the fig.1 as an example. This method has become a basic one in the research at BN-350 in the experiments of two types. The first one is the activation measurements by the sample irradiation at low power level described above with the further radiochemical analysis of the induced activity. This type of measurements has been described above. The other type is continuous irradiation of fuel and actinide samples at standard power level with the further radiochemical analysis of the sample nuclide composition variation. These measurements allow investigation of such neutron reactions which are not determined by the activation method, for example, radiation capture reaction on uranium-235, plutonium isotopes, americium-241, etc.

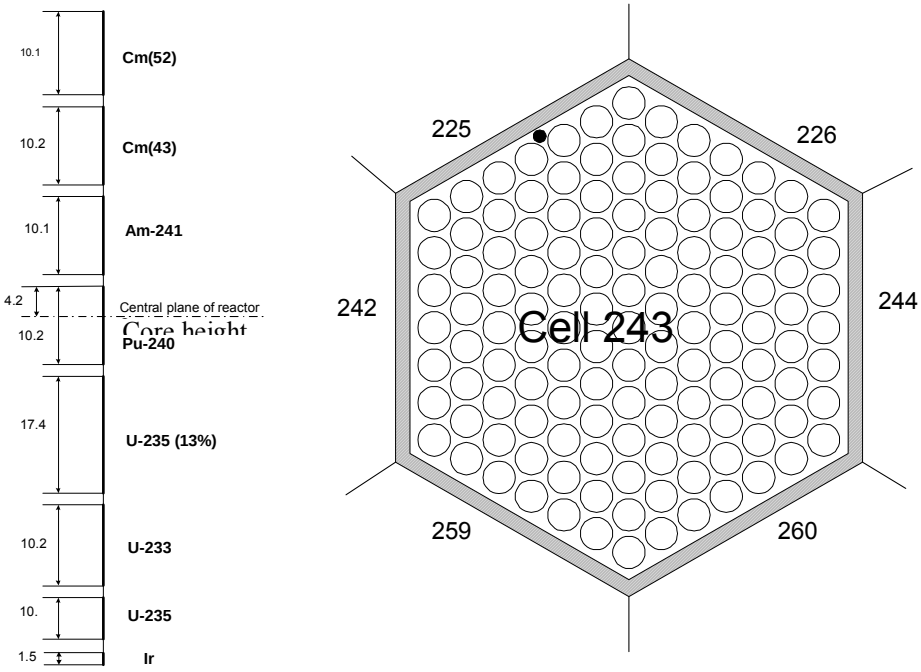

Fig.1. Samples layout in capillary and a fuel assembly irradiated in the 243th cell of BN-350 reactor.

Integrated gravichemical method of irradiated sample nuclide composition study. This method is the basic one during the research of the sample nuclide composition after long-term irradiation during one or several micro-runs (operation periods). It proposes the recovery of weight amounts of strictly stochoimetric compositions of uranium and plutonium and purification from impurities. Separate isotope amounts are determined by a combination of weighing (the use of the most precise method, if possible), radiometric and mass-spectrometric methods. The methods of alpha-spectrometric (actinides) and gamma-spectrometric (fission fragments) analysis are used. As the irradiated samples and their solutions are highly active the special specimens are prepared for the measurements by small amount sampling and further considerable dilution. Characteristic errors of α- and γ-measurements are presented in the Table 1. Mass-spectrometric measurements give the most precise results in relative concentration of one or another isotopes (parts of percent). Sometimes these measurements were combined with the method of the isotope dilution.

Table 1. Characteristic errors of α- and γ-spectrometric measurements and their main components.

Isotope	Error components, %					Total error %
	Statistics	Dilution	Sample preparation	Irradiation yield	Reference	
^{241}Am + ^{238}Pu (α)	1.0	1.8	2.0	0.5	< 0.3	2.9
^{241}Am (γ)	0.4	1.0	2.0	-	2.0	3.0
^{238}Pu (γ)	3.0	0.4	2.0	1.1	3.0	4.8
^{243}Cm (γ)	1.5	1.0	2.0	2.8	3.0	4.9
^{137}Cs (γ)	0.9	1.0	2.0	-	2.0	3.1

3. EXPERIMENTAL PROGRAM CHARACTERISTICS

Study of BN-350 start loading. BN-350 reactor start-up was accompanied by great set of measurements. For our opinion the measurements of spectral indexes, reaction rate distributions by the method of activation detectors and the research of the uranium fuel nuclide composition variation are of fundamental importance beyond the frames of the usefulness for BN-350 only. Activation measurements have been carried out with the use of special measuring fuel assemblies. During the data analysis the neutron field disturbances caused by these assemblies should be taken into account.

The fuel directly from fuel elements has been subjected to radiochemical research, as a rule, by dilution of one of this element as a whole. In this program the largest amount of samples has been tested, that was connected with great work on the experimental method mastering. However, due to this reason many results possess a methodic character and some of them are not reliable enough. Nevertheless, in this cycle direct information on alpha value for uranium-235 has been succeeded in obtaining for the first time as well as possible reproduction parameters in the core and blanket.

Study of the core with the second type of loading. This phase is connected with the research of BN-350 reactor characteristics with new type of fuel assemblies. It is logical continuation of the previous one. However, it is characterized by improved measurement quality. At that time the method of the “needle” control samples irradiated in the inter-element space of fuel assembly was first applied. The samples of plutonium-239 and power plutonium recovered from WWER reactors were studied for the first time. These experiments allow obtaining

alpha value of plutonium-239 as well as a number of other cross sections on plutonium isotopes and even americium-241.

Study of U-236 and Np-237 samples in order to determine Pu-236 accumulation and (n, 2n) reaction cross section on Np-237. This work has been initiated in connection with great urgency of determination of radiation dangerous uranium-232 nuclide accumulation, which main channel of generation is the mentioned reaction. In one's time this experiment indicated the excessive value of reaction cross section in ENDF/B-V file.

Study of mixed uranium-plutonium fuel in module of 7 fuel assemblies with MOX fuel. This cycle of research is one of the most laborious and productive. The research of fuel of both fuel elements and control samples has been carried out. Different nuclide accumulation characteristics, integral in fuel elements, have been measured as well as their altitude distribution. It is important to mention that irradiation has been carried out in plutonium fuel insertion where the typical fast breeder reactor spectrum is generated. Measurement results possess reliable information on plutonium breeder reproduction characteristics and plutonium isotope composition variation under irradiation. This program is supplemented with the program of the fuel composition variation research in the mode of uranium and plutonium recycle. Separation and recovery of uranium and plutonium fractions after irradiation, preparation of new samples from fractions recovered and repeated irradiation have been carried out in this program. The program of recycle research in its experimental part has not been completed yet.

Measurements of the module with plutonium fuel assembly impact on neutron field are also of great interest. The measurements have been carried out by the method of activation detectors in three phases: without plutonium, with fresh MOX fuel and depleted MOX fuel.

Study of metallic uranium fuel. In this specific experiment the neutron and physical peculiarities of fuel assembly with metallic fuel and the isotopic kinetics have been studied. However, such methodic fact as the other radiochemical research method application is important from the point of view of the method error analysis.

Study to substantiate thorium fuel cycle. Vast enough program on this matter has been carried out in BN-350. One of the main purposes was the research of the possibility of uranium-233 production with small concentration of uranium-232 isotope and more precise definition of U-232 sources. In various irradiation versions from parts of ppm up to 700 ppm of uranium-232 has been accumulated. Cross sections have been determined on thorium and protactinium isotopes by the activation method. These measurements have been partly used in Russian system BNAB for Th-232(n, γ) (n,f), Pa-231(n,γ), Pa-233(n, γ) cross sections testing.

Study to support the minor actinide transmutation program. This phase of research is currently carried out in part of radiochemical measurements and analysis. By present the samples presented in the Table 1 have been irradiated in BN-350 reactor core. During irradiation up to 35% of initial amount of actinides had time either to burn up or turn into daughter nuclides. This program will allow obtaining reliable direct information on MA transmutation real rate in fast reactor spectrum and competition of burning and secondary activity accumulation.

Study of neutron spectra, spectral indexes and hydrogen and helium generation cross sections on structure materials. In the course of this program the "needle" activation

detectors of wide set of materials (see, for example, the Table 3 below) have been prepared, which have been irradiated at small power level. Induced activity measurements allow the ratios definition of neutron reaction cross sections. In this case many cross sections have peculiarities: different energy thresholds or dominating resonance. That's why according to the set of indexes the neutron energy spectrum can be determined by its restoration with the help of one of known algorithms. Good determination of neutron spectra allows the threshold reaction cross section determination and testing leading to the reaction gaseous products accumulation and damage of structure materials.

Brief characteristics of actinide samples irradiated in BN-350 reactor are presented in the Table 2. The Table is supplemented with the figure 2 where the neutron reaction cross sections measured are also marked. It can be seen from the figure that practically all main channels of reactions responsible for the nuclide conversion in Th-U, U-Pu fuel cycle and fuel cycle with MA have been researched.

Table 2. The list of actinide samples researched.

№	Irradiation date	Number of samples	Composition	Radiation zone	Measurement results	Cross section ratio derived
1. Study of uranium oxide fuel of fuel elements of the first loading						
1.1	1973 – 1976	9	Enriched (26%) **U-235**	HEZ	Pu,FP accumulation U, Pu isotopic composition	α^5 $\sigma_c(8)/\sigma_f(5)$ $\sigma_c(39)/\sigma_f(5)$ $\sigma_{n,2n}(39)/\sigma_f(5)$
1.2	1973 – 1976	7	Enriched (17%) **U-235**	LEZ	Pu,FP accumulation U, Pu isotopic composition	
1.3	1973 – 1976	22	Depleted uranium	HEZ, LEZ, blanket	Pu,FP accumulation U, Pu isotopic composition	
2. Study of uranium oxide fuel of fuel elements of the second loading and control samples irradiated in fuel assemblies of the second loading.						
2.1	1978-80	3	**U-235** (21%)	LEZ	Pu,FP accumulation U, Pu isotopic composition	-
2.2	1981	1	**U-235 (**88%)	LEZ	Pu,FP accumulation U, Pu isotopic composition	α^5
2.3	1981	1	**Pu-239** (95%)	LEZ	Am, Cm, FP accumulation Pu isotopic composition	α^9 $\sigma_c(40)/\sigma_f(39)$ $\sigma_c(41)/\sigma_f(39)$ $\sigma_c(Am241)/\sigma_f(39)$ $\sigma_{n,2n}(39)/\sigma_f(39)$ $\omega(Am\text{-}242^m)$
2.4	1981	2	MOX, reactor (WWER) **Pu (74% Pu-239**)	LEZ	Am, Cm, FP accumulation Pu isotopic composition	
3. U-236 and Np-237 sample research in order to determine Pu-236 accumulation and (n,2n) reaction cross sections onPu-239.						
3.1	1977-78	2	**U-236**(99.66%)	HEZ, blanket	Pu-236/Pu-238	$\sigma_{n,2n}/\sigma_c(Np^{237})$
3.2	1977-78	4	**Np-237**(100%)	HEZ, LEZ, blanket	Pu-236/Pu-238	$\sigma_{n,2n}/\sigma_c(Np^{237})$
4. Study of mixed uranium-plutonium fuel in the module of 7 fuel assemblies with MOX fuel						
4.1	28.09.82 16.06.83	9	MOX fuel: **21% Pu**	7Pu (LEZ)	Am, Cm, FP accumulation	α^9 $\sigma_c(8)/\sigma_f(39)$

			79% depleted U		Pu isotopic composition	$\sigma_c(40)/\sigma_f(39)$ $\sigma_c(41)/\sigma_f(39)$ $\sigma_c(241)/\sigma_f(39)$ $\sigma_{n,2n}(39)/\sigma_f(39)$
5. Study of metallic uranium fuel irradiated in experimental fuel radial blanket assembly with the same fuel.						
	06.12.87 22.03.88	3	Depleted **U-238**	LEZ	Pu,FP accumulation U, Pu isotopic composition	
6. Study of thorium and uranium-233, -234 samples for thorium fuel cycle substantiation.						
6.1	1987-1988	8	Dioxide **Th**	Outer radial blanket	U, Pa-233 accumulation U isotopic composition Dependence of U-232 accumulation on their location in the blanket	σ_c(Th-232)/σ_f(5) $\sigma_{n,2n}$(Th-232)/σ_f(5) σ_c(Pa-231)/σ_f(5)
6.2	1990-1992	6	Metal **Th**	blanket	U-233,U-232, FP accumulation	
6.3	1990-1992	1	**Th**	LEZ	U-233,U-232, FP accumulation	
6.4	1990-1992	2	**U-233**	LEZ	U, FP* isotopic composition	$\sigma_c(3)/\sigma_f(3)^*$
6.5	1990-1992	1	**U-234**	LEZ	U, FP* isotopic composition	$\sigma_c(4)/\sigma_f(4)^*$
7. Study of minor actinide (MA) samples for substantiation of the possibility of their transmutation.						
7.1	1990-1992	1	**Np-237**	LEZ	Under investigation	σ_c/σ_f(Np-237)*
7.2	1990-1992	2	**Am-241**	LEZ	Under investigation	σ_c/σ_f(Am-241)*
7.3	1990-1992	1	**Pu-238**	LEZ	Under investigation	σ_c/σ_f(Pu-238)*
7.4	1990-1992	2	**Pu-240**	LEZ	Under investigation	σ_c/σ_f(Pu-240)*
7.5	1990-1992	2	**Cm-243+ Cm-244**	LEZ	Under investigation	σ_c(Cm-244)/σ_f(5)*

* Presumably.

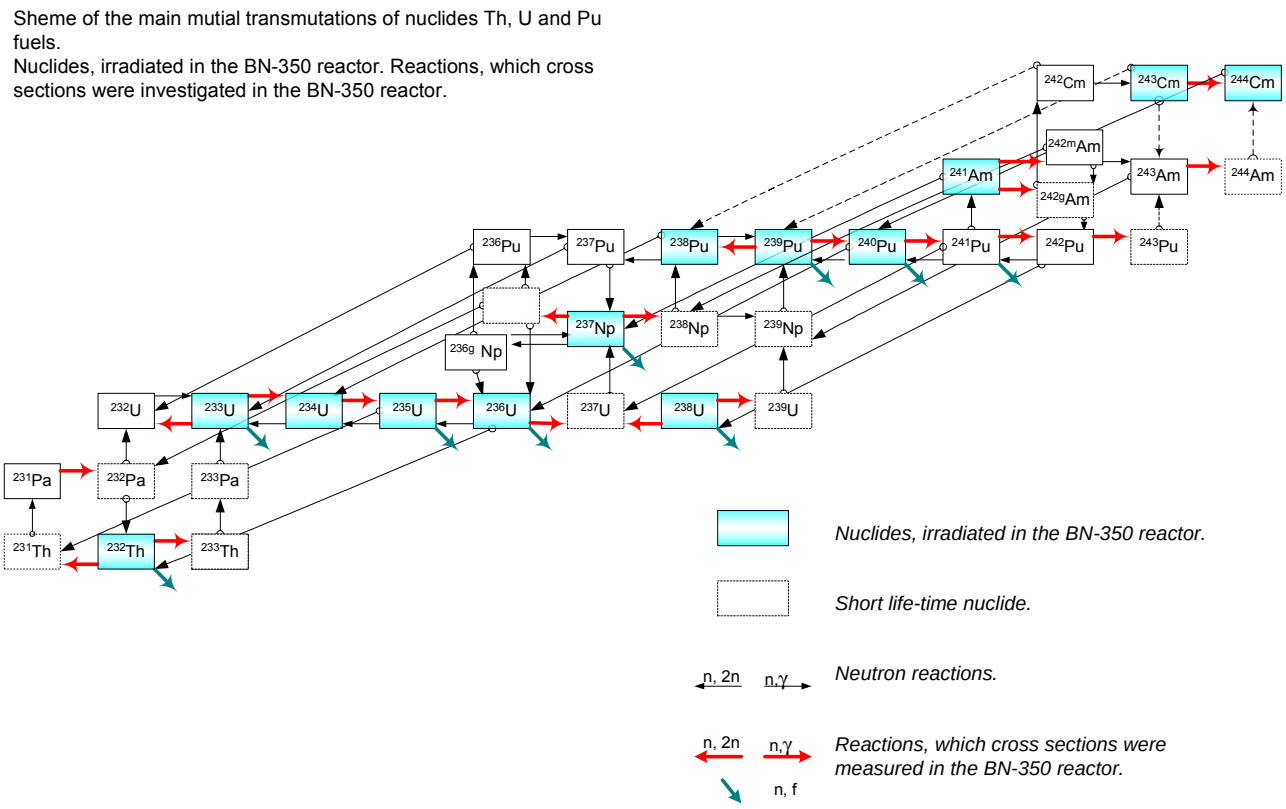

Fig.2. Radionuclides and reaction channels being studied in BN-350 reactor.

4. EXPERIMENT ANALYSIS.

At present an intense analysis takes place according to the transmutation program and the recycle problem analysis completion is planned. However, the tasks of analysis are not limited by these problems. Experimental research during the whole operation period was certainly accompanied with the calculation analysis. In particular, the measurement results of cross section ratios were compared with the same calculated data obtained on the base of BNAB constant library and promoted its improvement. Experiments also facilitated the calculation method improvement. The main drawback of the analysis performed is that the analysis results reflected the status of constant and software provision at the moment of the experiment execution. It is difficult to compare and analyze the measurement results, which are performed and analyzed in different times. It is impossible to use BN-350 experiments for verification of modern versions of nuclear data and codes.

That's why we make attempts to:

- Create the database of the whole set of experimental neutron and physical research at BN-350;
- Make an expert opinion on the information accumulated;
- Develop benchmarks based on the information, adjust experimental results to the benchmark simulation conditions;
- Carry out the calculation analysis on the single constant and code basis, analyze its consistency;

- Maintain obtained knowledge for further application by new generation of Russian and foreign specialists.

This work is under execution now. We expect and even have obtained partially the results in four data types:
- reaction cross section ratios on actinides and structure material, spectral indexes;
- reaction rate distribution in BN-350 reactor core volume;
- nuclide composition variation of different fuel compositions (UOX, MOX, Th, recycled fuel, etc.) and actinide samples during long-term irradiation in BN-350 reactor;
- neutron spectra.

As an example, figure 3 show the model of the core loading during 7 MOX subassemblies were irradiated. The main reactor parameters for this loading are given in table 3.

Table 3. The main BN-350 reactor parameters.

Thermal power, MWt	750	
Subassemblies irradiation interval, eff.days	IC	OC*
	395	456
Interval between reloadings, eff.days	79	
Number of subassemblies in the core	IC	OC
	109	115
Type of fuel	UO_2	
Fuel enrichment, %		
IC	17	
OC	26	
Core FSA hexagonal cladding across flats dimensions, mm	96 x 2	
Hexagonal lattice pitch, mm	98	
Core subassembly fuel pin diameter, mm	6.9 x 0.4	
The number of fuel pins in core subassembly	127	
Fuel column height, mm	1000	
Fuel effective density, g/cm^3	8.6	
Axial blanket material	Depleted UO_2 (0.4% ^{235}U)	
Fuel column height in upper axial blanket, mm	300	
Fuel column height in lower axial blanket, mm	380	
Effective density of depleted UO_2 in axial blanket, g/cm^3	8.6	
Radial blanket material	Depleted UO_2 (0.4% ^{235}U)	
Radial blanket fuel pin diameter, mm	14.2 x 0.4	
The number of fuel pins in radial blanket subassembly	37	
Fuel column height in radial blanket, mm	1680	
Effective density of depleted UO_2 in radial blanket, g/cm^3	9.4	
The number of radial blanket assemblies including:	356	
Inner radial blanket	101	
Outer radial l blanket	255	
Inner radial blanket subassemblies irradiation interval, eff.days	553	
Outer radial blanket subassemblies irradiation interval, eff.days	1106	
The number of subassemblies in the in-vessel storage	109	
Number of control rods,	12	

including:	
SR - shim rods	6
AR - automatic regulator	2
TC - temperature reactivity compensators	1
SA - scram assemblies	3

* IC – Inner Core, ITC – Intermediate Core, OC – Outer Core

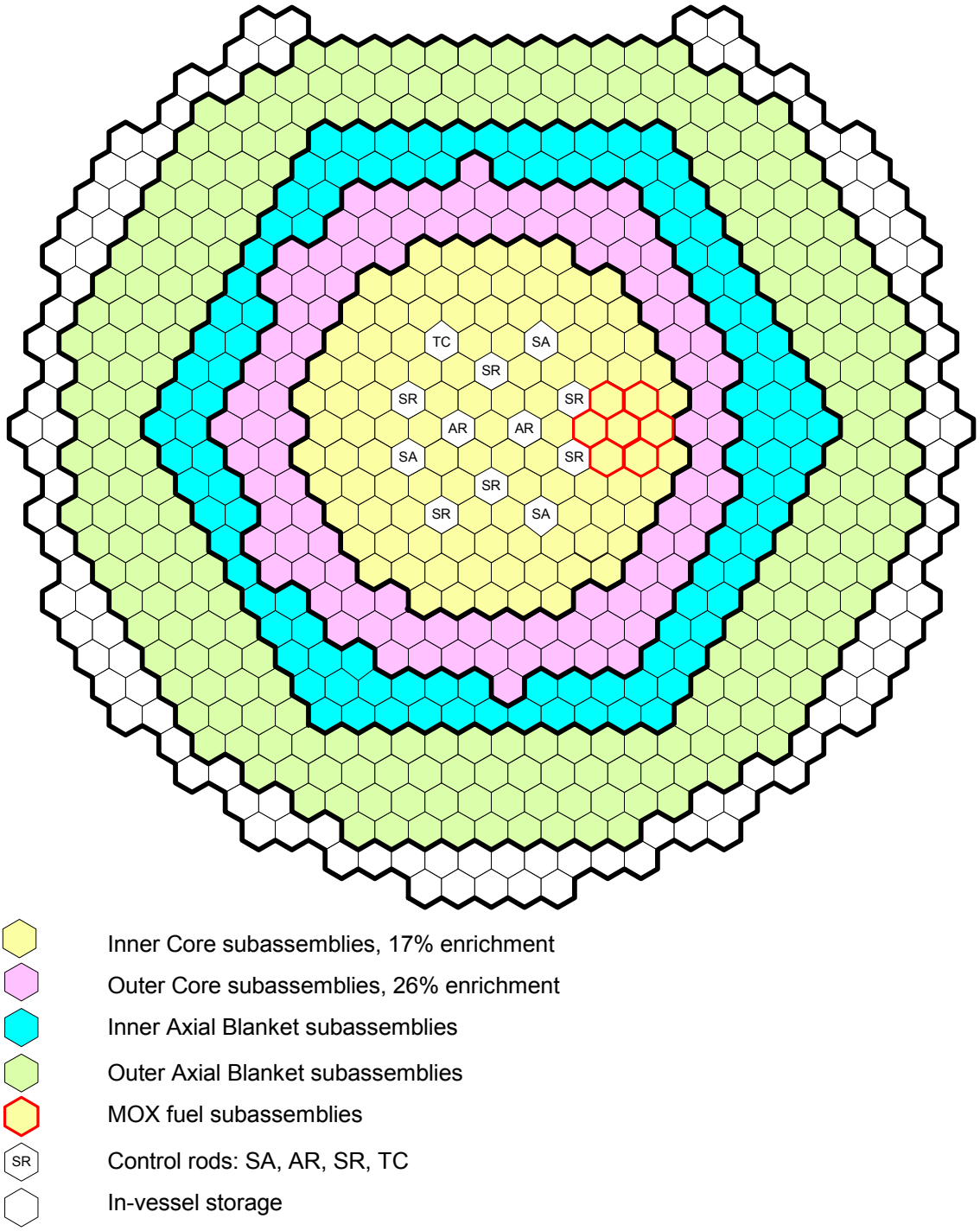

Fig. 3. Reactor map for 7 MOX subassemblies loading.

Reaction cross section ratios and spectral indexes. Discrepancy between calculation and experiment for the main cross sections on actinides is presented in the Table 4. The data have generalized character because many cross sections have been obtained from the nuclide composition analysis of irradiated fuel and actinide samples. Calculation has been carried out with the help of TRIGEX code [4] with BNAB-93 [5] constants.

Table 4. Comparison of calculated (C) and experimental (E) data on cross section ratios on actinides, (C – E)/E, %

Isotope	σ_x/σ_{f5}	LEZ	HEZ	MOX Sub-zone
Th-232	fiss	-1 ± 4	3 ± 7	5 ± 6
	capt	5 ± 6	-8 ± 7	0 ± 7
Pa-231	capt	2 ± 5	-	-
U-235	α	-2 ± 3	10 ± 4	-
U-236	fiss	4 ± 5	1 ± 5	0 ± 6
	capt	5 ± 5	-	-
U-238	fiss	3 ± 3	3 ± 5	2 ± 4
	capt	-1 ± 3	1 ± 3	0 ± 4
	n,2n	-5 ± 11	10 ± 10	-
Np-237	fiss	4 ± 4	-2 ± 5	3 ± 5
	capt	-	-3 ± 6	-
	n,2n	4 ± 6	2 ± 6	-
Pu-239	fiss	1 ± 3	0 ± 3	0 ± 4
	α	2 ± 4	15 ± 6	1 ± 3
	n,2n	-	-	-6 ± 7
Pu-240	fiss	3 ± 5	4 ± 5	-
	capt	0 ± 5	-	9 ± 6
Pu-241	α	-8 ± 11	-	-6 ± 6
Am-241	capt	0 ± 8	-	-11 ± 5

On the whole, the main cross sections, as it is seen from the Table, are in good enough agreement. The result of alpha value measurement for uranium-235 and plutonium-239 in HEZ has not been confirmed by the other measurements. Apparently, either an experimental mistake takes place or the experimental conditions have not been precisely reproduced by the calculation. At present the efforts are made to define them more precisely. As for americium-241, evidently, data also need further precise definition. The analysis of "pure" americium-241 samples currently takes place and it will provide more reliable data. Data on some other isotopes: curium-244, -245, uranium-234, are also expected to be obtained as well as more precise definition of a probability of americium-242m metastable isotope generation.
Comparison results of several spectral indexes and cross sections on structure materials and europium-153 are presented in the Table 5. It can be seen that there are noticeable discrepancies in some reactions, which can be used for more precise definition of corresponding cross sections.

Table 5. Spectral indexes and ratios of reaction cross sections on structure materials and uranium-235 fission cross section.

Reaction	High enrichment zone (HEZ)		Low enrichment zone (LEZ)	
	Experiment	C/E	Experiment	C/E
^{46}Ti(n,p)			(7.0±0.3)-4	0.94
^{47}Ti(n,p)			(1.30±0.06)-3	1.09

^{48}Ti(n,p)			(1.71±0.08)-5	0.80
^{54}Fe(n,p)	(6.75±0.60)-3	1.06	(5.4±0.3)-3	1.03
^{56}Fe(n,p)	(7.1±0.6)-5	1.16	(6.2±0.2)-5	1.02
^{58}Ni(n,p)	(9.3±0.6)-3	1.02	(7.2±0.3)-3	1.02
^{60}Ni(n,p)			(1.36±0.07)-4	0.89
^{54}Fe(n,α)	(4.73±0.33)-5	0.29	(4.3±0.2)-5	0.25
^{59}Co(n,α)			(8.9±0.6)-6	0.95
^{92}Mo(n,α)			(4.2±0.4)-6	2.81
^{50}Cr(n,γ)			(1.63±0.08)-2	1.10
^{55}Mn(n,γ)	(1.5±0.1)-2	1.07	(2.33±0.12)-2	0.98
^{58}Fe(n,γ)			(5.3±0.2)-3	1.12
^{59}Co(n,γ)	(1.74±0.13)-2	0.82	(2.04±0.10)-2	0.91
^{64}Ni(n,γ)			(2.86±0.15)-3	1.47
^{94}Zr(n,γ)			(9.7±0.5)-3	1.29
^{98}Mo(n,γ)			(4.1±0.2)-2	1.11
^{153}Eu(n,γ)			1.14±0.05	1.12

Reaction rate distribution. Calculation results of Pu-239 fission rate radial distribution being compared with the experimental data are presented on the fig.4. Results are presented for three reactor states: for uranium zone, zone with plutonium insertion of 7 fuel assemblies and with the insertion burnt up. The energy release raise in 30-50cm area is provided by plutonium impact. On the whole, sufficient data agreement has been obtained.

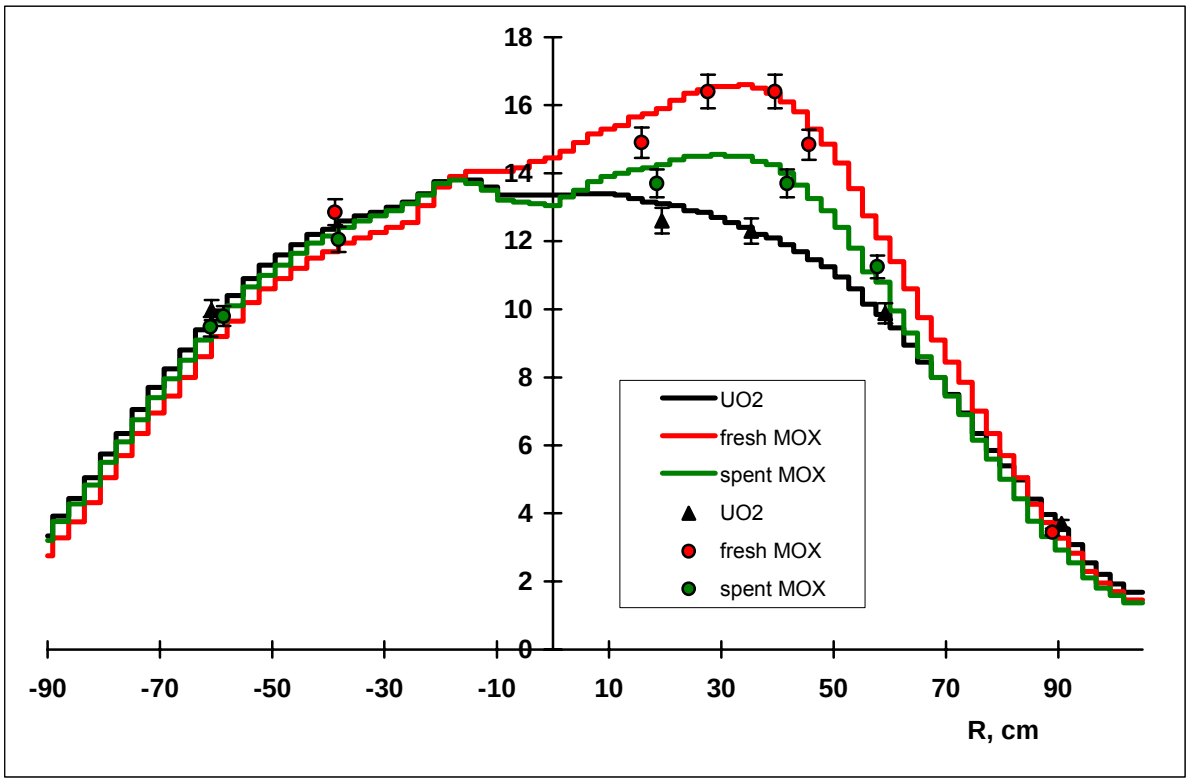

Fig. 4. Pu-239 fission rate radial distribution.

Nuclide composition of spent fuel. Comparison of calculation and experiments on the measurement of irradiated MOX fuel nuclide composition is presented in the Table 6. The results are rather preliminary because data processing is still carried out. It is also carried out for the other fuel types (UOX, Th). The largest discrepancy is observed in trans-plutonium element accumulation. This indicates that the integrated consideration of the fuel and actinide sample analysis results is especially urgent. Currently radiochemical analysis of Np-237, Pu-238, Pu-240, Am-241 and Cm-244 samples is carried out. The authors consider that simultaneous analysis of the whole set of data will allow the mentioned discrepancy reduction.

Table 6. Discrepancy between calculated and measured nuclide compositions of MOX fuel irradiated in plutonium insertion of BN-350 reactor.

№	Parameter	C/E-1, %
1	Uranium isotopic composition:	
	U-235	-2±2
	U-236	-19±12
	U-238	-0±2
2	Pu isotopic composition:	
	Pu-236	+6±15
	Pu-238	-7±2
	Pu-239	0±1
	Pu-240	-2±1
	Pu-241	+3±2
3	Ratio of Pu and U amounts	0±2
4	Trans-plutonium element accumulation:	
	Am-241	-40±1
	Cm-242	-16±3
	Cm-244	-60±9
5	Burn-up	0±3

CONCLUSION

The purpose of this report is the work demonstration on the results adjusting of the neutron and physical experiments performed in BN-350 reactor during the years of its operation to a form suitable for application for verification of fast reactor nuclear data and neutron calculation codes taking into account continuously varying nuclide composition. The result obtained possible scope of application is the use of MOX fuel in fast reactors, study of transmutation problems, thorium cycle, etc.

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