CALCULATION AND EXPERIMENTAL STUDIES ON MINOR ACTINIDES SAMPLES IRRADIATION IN FAST REACTORS

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Abstract

Review of the experimental work on the 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 programmes 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 and BOR-60 reactor adjusted to conditions of specially developed benchmark models.

Introduction

Within sequence of years a great number of physical experiments was carried out in BN-350 and BOR-60 reactor. 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 experimental data analysis and compilation are currently carried out in order to develop the database supporting the International programmes on MA transmutation and for calculation code and nuclear date verification as well.

Measurements in BN-350 reactor

Method of the "needle" detectors irradiated in the inter-element space of the fuel assembly 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 1.2 mm diameter have been fabricated and consecutively loaded into capillary of 1.6 mm 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 Figure 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. The other type is long-term 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.

Figure 1. **The scheme of samples location into assembly**

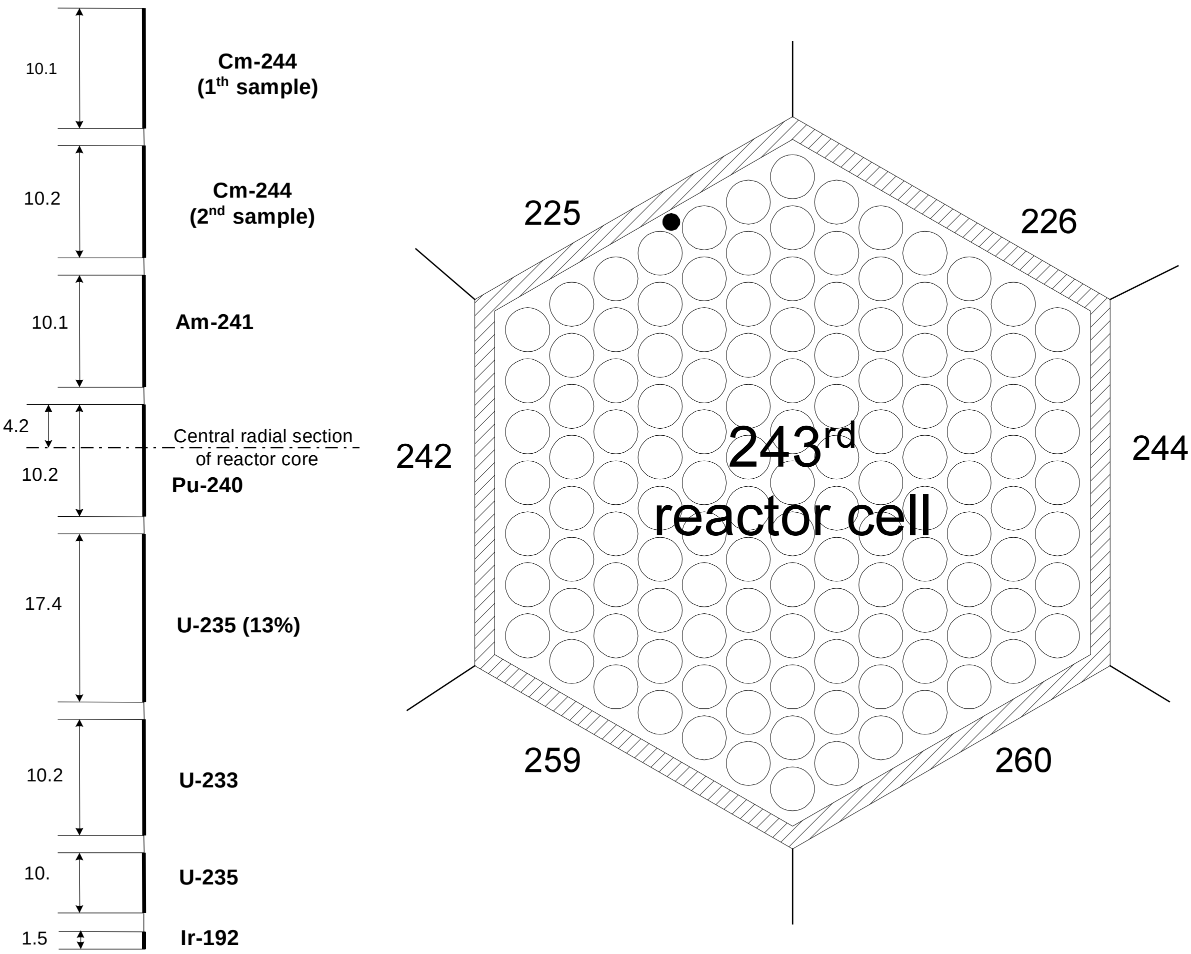

Table 1. **The main BN-350 reactor parameters**

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	96×2	
Hexagonal lattice pitch, mm	98	
Core subassembly fuel pin diameter, mm	6.9×0.4	
The number of fuel pins in core subassembly	127	
Fuel column height, mm	1 000	
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	14.2×0.4	
The number of fuel pins in radial blanket subassembly	37	
Fuel column height in radial blanket, mm	1 680	
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	1 106	
The number of subassemblies in the in-vessel storage	109	
Number of control rods, Including:	12	
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

Figure 2. **Reactor map of BN-350**

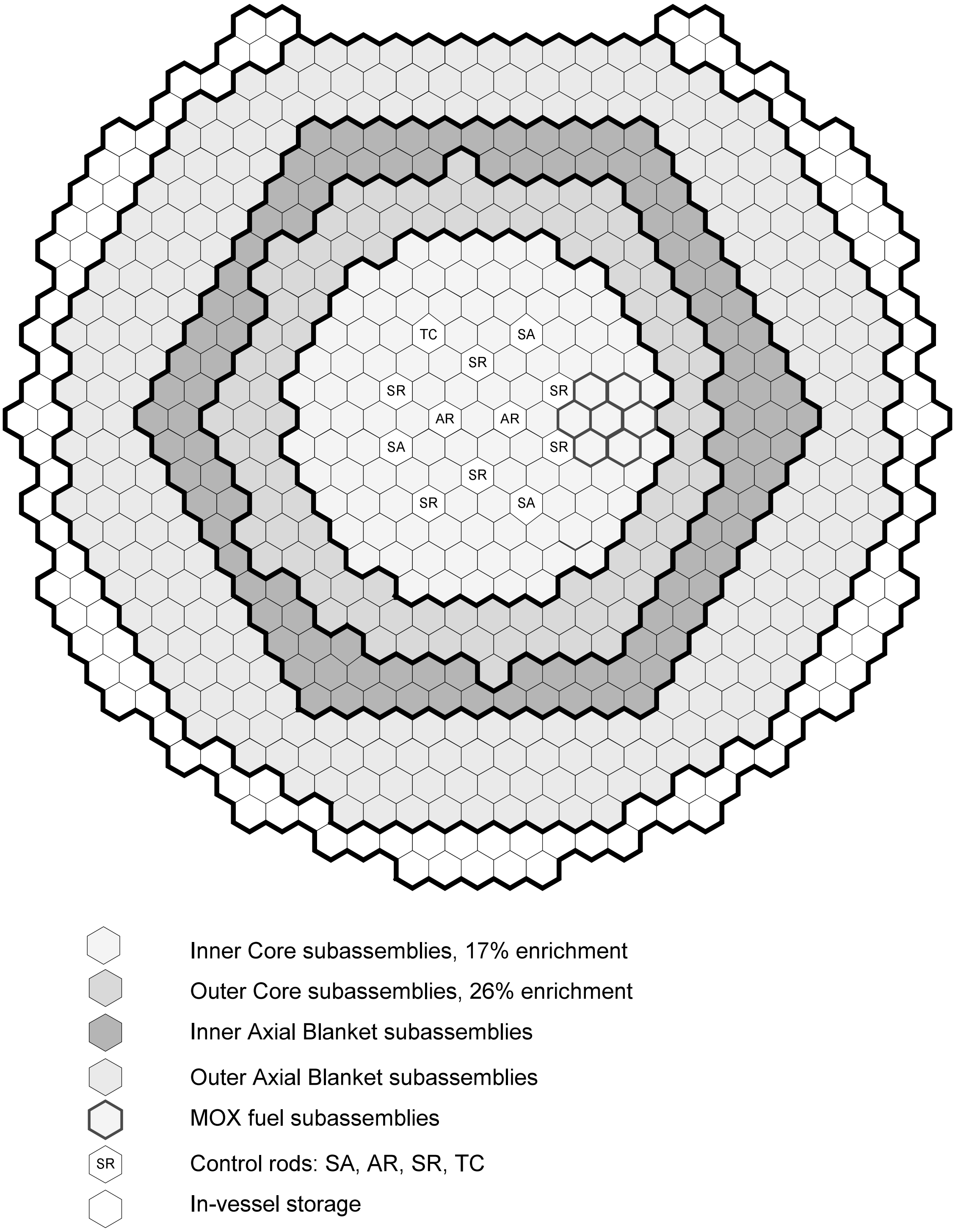

Table 2. **The list of actinide samples researched**

№	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%) ^{235}**U**	HEZ	Pu, FP accumulation U, Pu isotopic composition	α^5 $\sigma_c(8)/\sigma_f(5)$
1.2	1973-1976	7	enriched (17%) ^{235}**U**	LEZ	Pu, FP accumulation U, Pu isotopic composition	$\sigma_c(39)/\sigma_f(5)$ $\sigma_{n,2n}(39)/\sigma_f(5)$
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	1978-1980	3	^{235}**U** (21%)	LEZ	Pu, FP accumulation U, Pu isotopic composition	–
2.2	1981	1	^{235}**U (**88%)	LEZ	Pu, FP accumulation U, Pu isotopic composition	α^5
2.3	1981	1	^{239}**Pu** (95%)	LEZ	Am, Cm, FP accumulation Pu isotopic composition	α^9 $\sigma_c(40)/\sigma_f(39)$ $\sigma_c(41)/\sigma_f(39)$
2.4	1981	2	MOX, reactor (WWER) **Pu (74%** ^{239}**Pu**)	LEZ	Am, Cm, FP accumulation Pu isotopic composition	$\sigma_c(Am241)/\sigma_f(39)$ $\sigma_{n,2n}(39)/\sigma_f(39)$ $\omega(^{242m}Am)$
3. ^{236}***U and*** ^{237}***Np sample research in order to determine*** ^{236}***Pu accumulation and (n,2n) reaction cross sections*** ^{239}**Pu**						
3.1	1977-1978	2	^{236}**U(99.66%)**	HEZ, blanket	$^{236}Pu/^{238}Pu$	$\sigma_{n,2n}/\sigma_c(^{237}Np)$
3.2	1977-1978	4	^{237}**Np(100%)**	HEZ, LEZ, blanket	$^{236}Pu/^{238}Pu$	$\sigma_{n,2n}/\sigma_c(^{237}Np)$
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** **79% depleted U**	7Pu (LEZ)	Am, Cm, FP accumulation Pu isotopic composition	α^9 $\sigma_c(8)/\sigma_f(39)$ $\sigma_c(40)/\sigma_f(39)$ $\sigma_c(41)/\sigma_f(39)$ $\sigma_c(241)/\sigma_f(39)$ $\sigma_{n,2n}(39)/\sigma_f(39)$
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 ^{238}**U**	LEZ	Pu, FP accumulation U, Pu isotopic composition	

Table 2. **The list of actinide samples researched** (contd.)

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, ^{233}Pa accumulation U isotopic composition dependence of ^{232}U accumulation on their location in the blanket	$\sigma_c(^{232}Th)/\sigma_f(5)$ $\sigma_{n,2n}(^{232}Th)/\sigma_f(5)$ $\sigma_c(^{231}Pa)/\sigma_f(5)$
6.2	1990-1992	6	Metal **Th**	blanket	^{233}U, ^{232}U, FP accumulation	
6.3	1990-1992	1	**Th**	LEZ	^{233}U, ^{232}U, FP accumulation	
6.4	1990-1992	2	^{233}**U**	LEZ	U, FP* isotopic composition	$\sigma_c(3)/\sigma_f(3)^*$
6.5	1990-1992	1	^{234}**U**	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	^{237}**Np**	LEZ	under investigation	$\sigma_c/\sigma_f\,(^{237}Np)^*$
7.2	1990-1992	2	^{241}**Am**	LEZ	under investigation	$\sigma_c/\sigma_f\,(^{241}Am)^*$
7.3	1990-1992	1	^{238}**Pu**	LEZ	under investigation	$\sigma_c/\sigma_f\,(^{238}Pu)^*$
7.4	1990-1992	2	^{240}**Pu**	LEZ	under investigation	$\sigma_c/\sigma_f\,(^{240}Pu)^*$
7.5	1990-1992	2	^{243}**+Cm** ^{244}**Cm**	LEZ	under investigation	$\sigma_c(^{244}Cm)/\sigma_f\,(5)^*$

* Presumably.

Measurements in BOR-60 reactor

Two pins with eight ampoules in each were irradiated in BOR-60 reactor. These pins are located into experimental assembly having 19 pins (see Figure 3). Studying actinides were set into quartz capsule having follow dimensions: outer diameter – 4.5 mm; length – 20-21 mm; thickness – 0.7-0.8 mm. The set of samples is shown in Table 3.

Figure 3. **Map of BOR-60 experimental assembly**

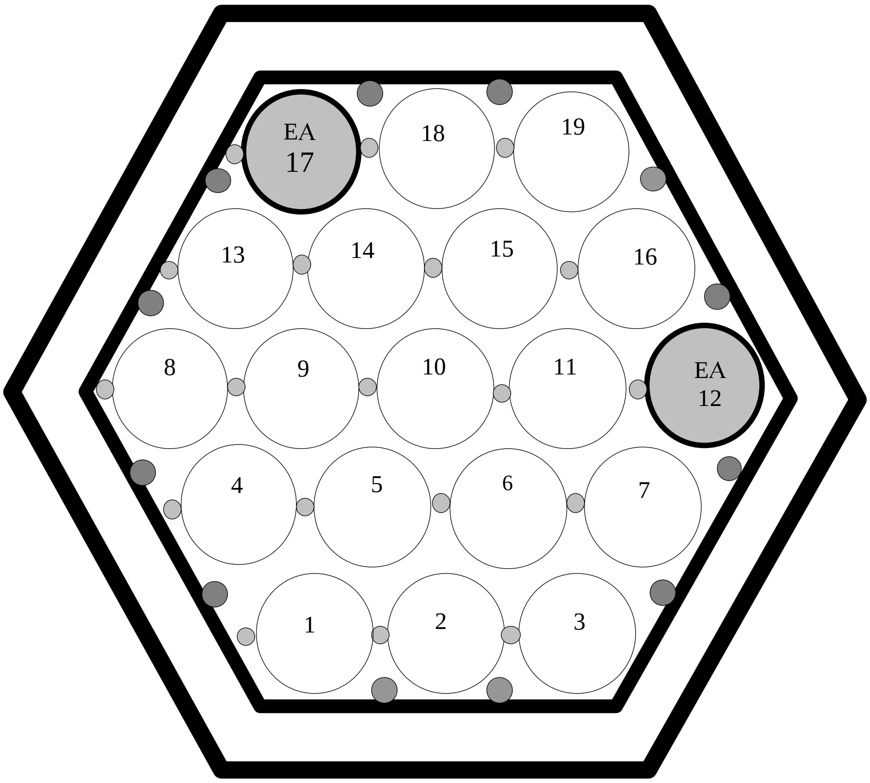

Table 3. **The initial MA isotope composition into the ampoules**

Ampoule marking	**The basic isotope**	**Mass** (mkg)	**The isotope composition** (%)
1	^{232}Th	419	^{232}Th – 100
2	^{237}Np	408	^{237}Np – 100
3	^{239}Pu	115	^{238}Pu – 0.36 ^{239}Pu – 94.80 ^{240}Pu – 4.66 ^{241}Pu – 0.18 ^{242}Pu – <0.01
4	^{240}Pu	126	^{238}Pu – 0.18 ^{239}Pu – 1.00 ^{240}Pu – 98.82
5	^{242}Pu	127	^{242}Pu – 99.54
6	^{241}Am	129	^{241}Am – 100
7	^{243}Am	113	^{241}Am – 2.37 ^{243}Am – 97.62 ^{244}Cm – <0.01
8	^{244}Cm	129	^{244}Cm – 100

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. Typical errors of α- and γ-measurements are presented in the Table 4. 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 4. **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

Analysis of experimental results

Calculation analysis of experiments is based on:

- ABBN-93 constant system; [1]
- three-dimension hexagonal geometry code TRIGEX [2] in diffusion approximation for neutron field calculation;
- CARE [3] code for calculation of nuclide composition of the spent fuel and samples.

The following problems are being solved while analysis:

- neutron field precise calculations according to detail description of changing in core configuration in course of fuel reloading and burn-up;
- calculation of nuclide composition of fuel pins or samples;
- comparison of calculation and experimental data on nuclide composition;
- adjustment of neutron fluences according to results of analysis of monitor samples;
- neutron reactions cross sections rates decision according to results of nuclide composition analysis;

- definition of benchmark model of experiments, directed on neutron data precision;
- reducing results of separate measurements to benchmark model conditions and analysis of consistency;
- creation of data base of experiments;
- neutron data precision and adjustment.

Results of comparison of series of cross section rates, obtained in BN-350 experiments are given in Table 5. Tables 6 and 7 show preliminary results of expected changing in nuclide composition of actinide samples, being analysed at present.

Table 5. **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**
^{232}Th	Fiss Capt	-1 ± 4 5 ± 6	3 ± 7 -8 ± 7	5 ± 6 0 ± 7
^{231}Pa	Capt	2 ± 5	-	–
^{235}U	α	-2 ± 3	10 ± 4	–
^{236}U	Fiss Capt	4 ± 5 5 ± 5	1 ± 5 –	0 ± 6 –
^{238}U	Fiss capt n,2n	3 ± 3 -1 ± 3 -5 ± 11	3 ± 5 1 ± 3 10 ± 10	2 ± 4 0 ± 4 –
^{237}Np	fiss capt n,2n	4 ± 4 – 4 ± 6	-2 ± 5 -3 ± 6 2 ± 6	3 ± 5 – –
^{239}Pu	fiss α n,2n	1 ± 3 2 ± 4 –	0 ± 3 15 ± 6 -	0 ± 4 1 ± 3 -6 ± 7
^{240}Pu	fiss capt	3 ± 5 0 ± 5	4 ± 5 –	– 9 ± 6
^{241}Pu	α	-8 ± 11	–	-6 ± 6
^{241}Am	capt	0 ± 8	–	-11 ± 5

* Low enrichment zone

** High enrichment zone

Table 6. **The calculation estimations of characteristics MA samples, irradiated in BN-350**

Isotope	**Reactor cell**	**Irradiated time, days**	**Fluence 10^{23} $cm^{-2}s^{-1}$**	**Basic isotope burn-up** (%)	**Secondary actinide generation** (%)	**Total burn-up** (%)
^{237}Np	89	781	2.0	35	25	10
^{238}Pu	89	781	1.8	31	8	23
^{240}Pu	110	781	1.7	11	0.5	10.5
^{241}Am	89	781	1.9	35	25.5	9.5
^{241}Am	243	797	2.0	35	25.7	9.3
^{244}Cm	243	797	1.5	25	11	14
^{244}Cm	243	797	1.7	31	14	17

Conclusions

- In the BN-350 reactor there have been performed wide investigations of nuclide composition changing of fuel samples and several separate isotopes, which allow to increase accuracy and reliability of nuclide data for basic isotopes, such as U, Pu and some minor actinides.
- Series of experiments has got visible discrepancies between calculation and experimental data, for example, for such isotopes as ^{240}Pu, ^{241}Am. As a rule, analysis of U or Pu samples shows the larger accumulation of Cm isotopes in experiment. In the whole, the minor actinides data are still insufficient. At the present time the work on analysis of minor actinides samples, irradiated in BN-530 and BOR-60, is carried out and it is going to give the necessary experimental information.
- Another unsolved problem is that analysis of various experiments was carried out at the different time using different approaches and different nuclear data, that does not allow to carry out complete analysis of obtained data. It is necessary to perform:
- evaluation of experiments carried out in BN-350 and BOR-60;
- creation of benchmarks on the base of these experiments for testing nuclear data and calculation codes and storing it in the unified data base;
- calculation analysis of benchmarks.

REFERENCES

[1] A.S. Seregin (1983), *Annotation of TRIGEX Code Intended for Low-group Neutron and Physical Reactor Calculation in Hexagonal Geometry*. Problems of Atomic Science and Engineering. Series Nuclear Reactor Physics and Engineering. Issue 4(33).

[2] G.N. Manturov, M.N. Nikolaev, A.M. Tsiboulia (1996), BNAB-93 Group Constant System. Part 1. *Nuclear Constants for Neutron and Photon Radiation Field Calculation*. Problems of Atomic Science and Engineering. Series Nuclear Constants 1, p. 59.

[3] A.L. Kochetkov (1995), Preprint IPPE-2431, Obninsk.

Table 7. **The calculation estimations of characteristics MA samples, irradiated in BOR-60**

Sample	**Isotope**	**Initial isotope composition,** (mkg)	**Isotope composition, after irradiation** (mkg)	**Total burn-up** (%)
№ 1	**$^{232}\mathrm{Th}$**	419	410.6	**0.26**
	$^{231}\mathrm{Pa}$		0.051	
	$^{233}\mathrm{Pa}$		0.70	
	$^{233}\mathrm{U}$		6.5	
	$^{234}\mathrm{U}$		0.045	
№ 2	$^{234}\mathrm{U}$		0.26	**4.7**
	$^{237}\mathrm{Np}$	408	360.2	
	$^{238}\mathrm{Pu}$		27.6	
	$^{239}\mathrm{Pu}$		0.39	
№ 3	$^{238}\mathrm{Pu}$	0.414	0.36	**13.8**
	$^{239}\mathrm{Pu}$	109.02	90.8	
	$^{240}\mathrm{Pu}$	5.35	7.3	
	$^{241}\mathrm{Pu}$	0.207	0.57	
	$^{242}\mathrm{Pu}$	0.0115	0.019	
	$^{241}\mathrm{Am}$		0.042	
№ 4	$^{238}\mathrm{Pu}$	0.227	0.19	**5.8**
	$^{239}\mathrm{Pu}$	1.26	1.05	
	$^{240}\mathrm{Pu}$	124.51	109.0	
	$^{241}\mathrm{Pu}$		7.9	
	$^{242}\mathrm{Pu}$		0.098	
	$^{241}\mathrm{Am}$		0.42	
№ 5	**$^{242}\mathrm{Pu}$**	126.42	117.2	**3.8**
	$^{243}\mathrm{Am}$		4.3	
	$^{243}\mathrm{Cm}^{x}$		0.13	
№ 6	$^{234}\mathrm{U}$		0.049	**4.7**
	$^{237}\mathrm{Np}$		0.50	
	$^{238}\mathrm{Pu}$		5.7	
	$^{239}\mathrm{Pu}$		0.08	
	$^{242}\mathrm{Pu}$		1.6	
	$^{241}\mathrm{Am}$	129	110.5	
	$^{242m}\mathrm{Am}$		1.7	
	$^{243}\mathrm{Am}$		0.05	
	$^{242}\mathrm{Cm}$		2.6	
№ 7	$^{238}\mathrm{Pu}$		0.11	**3.3**
	$^{241}\mathrm{Am}$	2.678	2.32	
	$^{243}\mathrm{Am}$	110.31	100.9	
	$^{244}\mathrm{Cm}$	0.0113	5.48	
	$^{245}\mathrm{Cm}$		0.084	
№ 8	**$^{244}\mathrm{Cm}$**	129	107.6	**4.5**
	$^{245}\mathrm{Cm}$		2.9	
	$^{246}\mathrm{Cm}$		0.025	