BFS critical experiments for the Minor Actinides data correction

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Abstract

Starting from 90th at BFS critical assemblies the wide experimental program on MA constants investigations for different neutron spectrum was launched. The measurements of fission cross section ratios, central reactivity worths, Doppler effect were carried out, and also the experiments on direct introduction of MA in the fuel composition and investigations of its influence on neutronic parameters such as sodium void reactivity effect, control rod worths and spectrum indexes were fulfilled. The investigations of the traditional cores with oxide and metal fuel and sodium coolant have finished. The works on testing the cores with nitride fuel and Pb / Pb-Bi coolant are carried out. The study of the MA data for the neutron spectrum of molten-salt reactor and ADS system is in preparation. During the work the recommendations for ^{237}Np and ^{241}Am capture cross-sections correction in the energy diapason of neutrons for fast reactor MA burner were received. The aim of the work is the creation of the total set of evaluated integral experiments on critical assemblies for the definition of the MA data.

KEYWORDS: Fast critical assembly, minor actinides, methods of measurements, set of evaluated integral experiments

1. Introduction

The problem of refining of minor actinides (MA) fission cross-section and, especially, MA capture cross-section is rather actual, since at present their uncertainties are several times more then it is required [1]. For the last 20 years the fission and capture cross sections of the ^{235}U were investigated thoroughly, their uncertainties are estimated as 2% now. At the same time the uncertainties of the cross sections of the MA reach 15%. For the solving of the problem, together with new micro data on MA, the experiments of integral type at critical assemblies and reactors are necessary. These experiments are:

- Measurements of the ratio of fission cross-section by chambers;
- Central reactivity (worths) coefficients measurements;
- Doppler-effect measurements;
- Direct introducing of MA into fuel of the critical assembly core and study of its influence on the neutron characteristics (on sodium void reactivity effect -SVRE, control rod worths -CRW, and spectrum indexes).

Starting from 90th at BFS critical assemblies the wide experimental program the MA constants investigations at different neutron spectrum concerned is carried out.

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2. The methods of MA measurements at BFS facilities

At critical assemblies BFS, in different years were studied the following:

- The ratios of fission cross sections of MA:

${}^{i}\sigma_f / {}^{U235\ or\ Pu239}\sigma_f$ ***(i → ²³⁷Np, ²⁴¹Am, ²⁴³Am, ²⁴⁴Cm, ²⁴⁵Cm, ²³⁸Pu, ²⁴⁰Pu, ²⁴¹Pu)*** by fission chambers

- ${}^{Np237}\sigma_c / {}^{238}\sigma_c$ by activation method (foils)
- Small sample worths (^{237}***Np,*** ^{241}***Am,*** ^{240}***Pu,*** ^{241}***Pu)***
- Doppler effect for ^{237}***Np,*** ^{238}***U.***
- Adding the **Np** in the fuel composition of the assembly with aim to investigate his influence at neutronic characteristics such as SVRE, CRW, spectral indexes etc.

2.1 Fission cross-section ratios of MA

For measurements of MA fission cross section ratios at BFS critical assemblies, fission chambers of different types as well as solid state detectors are used.

2.1.1. Small size fission chambers

For the measurements of fission cross section ratios and fission rate distributions the small-size fission chambers (SSC, [2]) are used at BFS facilities . By design the fission chamber is a cylindrical stainless steel tube of 6 mm outer diameter and 0.5 mm wall thick and it acts as a cathode. Inside of this tube, another tube of 4 mm diameter is fixed by means of insulators and it acts as an anode and the layer of the fissile material is deposited on the outer surface of the anode. During the measurements the chambers with different MA materials, are placed by means of the measurement manipulator in to the inter tube gap of BFS facility on the required height. The calibration of fission chambers is carried out in the thermal neutron BFS column with well known parameters. So as the fission cross sections of the MA isotopes majority have the threshold behaviour, a known small fraction of the non-threshold U-235 or Pu-239 isotopes is introduced in the layouts. And so, for the realization of this method the precise knowledge of the impurities in layouts is required. At BFS facilities the SSC with layouts of ^{233}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu , ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm are in disposal. The main components of the uncertainties, taking into account during the measurements of the MA fission rate ratios, are the statistics errors, the errors of the material of the chamber components, the uncertainties of the thermal cross-sections and the uncertainties of the ratios of the fission rates of the non-substantive isotopes of fission chamber layouts. The total uncertainties for the chambers with non threshold isotope layout are till 1.5% and for the threshold – 2.5%.

2.1.2. Segment fission chambers

The segment chamber (SC) is a thin wall (0.5-0.7 mm) stainless steel cylinder of 44 mm diameter. The wall acts as one of the chamber electrodes. Internal electrodes are wall parts of one more cylinder which is coaxial with the external cylinder. Fissile material layers are deposited on central parts of the internal electrodes in form of strips with a length 46-50 mm. It is possible to place till four layouts in one chamber. The design of the chambers allows to put them in the empty BFS tubes, i.e. during the measurements at critical configuration, the fuel composition of one of the rod should be removed. The calibration of fission chambers also is carried out in the thermal neutron BFS column. At BFS facilities the SC with layouts of ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu , ^{237}Np, ^{243}Am and ^{232}Th. are in disposal. The components of the uncertainties and their values are close to the SSC ones.

2.1.3. Absolute fission chambers

Absolute fission chambers (AC) are closed to segment fission chamber by their geometrical parameters and have also a high efficiency of fission fragments registration due to thin layers (< 50 $\mu g/cm^2$) of fission materials. They also have a thin wall (0.5-0.7 mm) stainless steel cylinder of 44 mm diameter. One of electrodes of chamber is a flat bottom of the cylindrical case of chamber made of stainless steel with accurately polished inner surface. Another electrode in the kind of thin disk was located at the distance of 1 mm from the first one. The layer of fission substance under investigation is placed in the center of flat disk (electrode) with diameter of 20 mm. This electrode is separated from the case by teflon insulator. In order to measure by this method, the quantity of fission material in detector and efficiency of fission registration ε_f are determined preliminary, after this there is no necessity for additional calibration measurements in thermal neutron flux as in the case with SC and SSC. There is a large set of chambers with different layers of minor actinides and Pu and U isotopes at the BFS stand.

2.1.4 Solid state detector method

In this method the targets (aluminum disks with diameter of 6 mm, thickness of 0,7 mm) with fissile materials layouts and the glass disks, placed together in the Al containers, inserted in the inter tube gaps of the BFS investigated assembly. After irradiation the glass disks corrode by acids and the amount of tracks counted by microscope. The layers of targets were manufactured in Radium Institute (St. Petersburg) from isotopes with high enrichment on main isotope (for example ^{235}U - 99,99 weight %) by vacuum deposition technique and were calibrated. The targets with layouts ^{235}U, ^{239}Pu, ^{242}Pu, ^{237}Np, ^{241}Am, ^{243}Am are in disposal. The uncertainties of the method are 5%.

2.1.5. The comparison of the methods

As you can see from the information above, for the BFS facilities conditions the same MA fission ratio can measure by different methods. Frequently for the receiving of the reliable results, some of the MA fission ratios are measured at the critical assembly by two or more method. The wide work on comparison of all the methods presented in items 2.1.1 -2.1.4, was carried out at BFS-66 critical assembly (mock-up of BN-600 with MOX fuel) and was prolonged at BR-1 reactor, having principal another design as BFS stands, and more hard and reference neutron spectrum. The measurements with large dimension fission chambers (SC and AC) were not carried out due to geometry peculiarities of the BR-1. The results of the measurements, carried out by independent methods at BFS-66 and BR-1, agreed good in the limits of uncertainties declared.

2.2 The ratio of ^{237}Np capture cross section to the fission in ^{239}Pu ($\sigma_c^{237}/\sigma_f^{239}$)

To measure fission rate in the ^{239}Pu absolute fission chamber and simultaneously irradiate in this place the foil of ^{237}Np in the core, and then, carrying out the same measurement in the thermal flux, anyone can receive the ratio $\sigma_c^{237}/\sigma_f^{239}$ [3]. For this, using high pure Ge detector, the intensity of the γ-lines of ^{238}Np with energies 984.4 keV, 1026 keV and 1029 keV, which are the products of the capture of the neutrons by the ^{237}Np nuclei, are measured, and divided to the fission rate in the ^{239}Pu absolute fission chamber. The affectivities of the γ-quant registration are determined on the base of the ^{237}Np foils irradiation results in the thermal column, using well known thermal capture cross section of ^{237}Np. The uncertainty of the method is estimated about 2.5%.

2.3 The methods based on measurements of reactivity of critical assemblies

For the measurements of changing of the reactivity of the critical assembly, we use the system named reactimeter. This techniques use during measurements of SVRE, control rod worths, oscillation of small samples, Doppler effect, monitoring of power and others. It includes three neutron detectors installed in the radial blanket instead of the blanket rods on the distances 300 - 400 mm from the core boundary. The detectors are ionization chambers filled with BF^3. To increase the detector efficiency, six neighboring BFS tubes are filled with a polyethylene moderator. Changing of the assembly power arising after core perturbation is recorded and processed by means of a complex equipment consisting of measurement line with detector - boron chamber, current - frequency transducer and personal computer. Experimental value of reactivity of assemblies depending on time is determined in on-line and off-line modes with the help of the program-reactimeter on the base of solution of point neutron kinetics equations (inverse kinetic – IK), where input data are tabular constants of decay of nucleuses - predecessors of delayed neutrons, calculated group parts of delayed neutrons and experimental data as pulses of current whose frequency is proportional to the assembly power. Disturbance effects are determined as difference of critical assembly states after introduction of the sample and before its introduction. On the base of numerous experiments at BFS facilities and series of calculating experiments the final uncertainty of IK technique in a range of reactivity from + 0,3 βeff up to -10 βeff is estimated as ± (2 - 7) %. Reactivity error having been increased monotonous when reactivity itself increased.

2.3.1 Central small samples worths

The technique for measurements of reactivity worth coefficients is based on a periodical perturbation of critical assembly reactivity by small size samples. Structurally such samples are stainless steel tubes containing an investigated material and closed from both sides by steel plugs. Periodical movements (oscillations) of the driving tube with the sample up and down, along a gap between BFS tubes, are carried out by an automatic manipulator or by operator. The current values of reactivity are fixed by digital reactimeter on the base of point kinetic equation, table data of decay constants and the calculated group parts of delay neutrons. The reactivity effects are determined as differences of the critical assembly reactivity values in the states with and without the samples in the core. The following samples of MA ^{237}Np, ^{241}Am, ^{240}Pu, ^{241}Pu, and also the samples of ^{235}U, ^{239}Pu as etalons are using in experiments at BFS.

2.3.2 Doppler effect measurements

For the estimation of the effect of resonance absorption of neutrons as a function of temperature the method of the small samples worths measurements (see 2.3.1 above) during changing of their temperature are used at BFS. The measurements are carried out using oscillation of the samples together with heater in cold and hot conditions, in the central BFS channel. The temperature in assembly near heater during measurements don't exceed 40^0C, at the same time the temperature of the sample and heater is 620^0C. The control of the temperature field inside the heater and in the core near heater is carried out by several thermocouples. The Doppler effect value is determined by reactimeter as difference of the critical assembly perturbations due to introduction of the sample in the hot and cold conditions. At BFS critical assemblies the investigations of Doppler-effect for the ^{237}Np, ^{238}U, ^{240}Pu samples and civil plutonium.

2.3.3 The sodium void reactivity effect

The sodium void reactivity effect (SVRE) is determined as a difference of reactivity values for the critical assembly states with and without sodium in some part of the core volume. In BFS conditions, the void is formed when sodium pellets are changed by empty boxes. The procedure of carrying out the measurements is rather simple, however changing compositions of fuel rods in the critical assembly takes a lot of time. The change of reactivity resulting from the change of sodium by the void is measured by the digital reactimeter.

2.4 The simulation of the introduction of MA placing in the core of the reactor –burner

Having of the 11 kg of the neptuniuim-237 dioxide in the form of standard BFS pellets, we can simulate the placing of this actinide in the core of fast breeders. This amount is enough to simulate the 17% of ^{237}Np concentration in the all heavy nuclides of the core of the reactor-burner. The simulation of such concentration permits to investigate the influence of the introduction of MA on safety parameters of the cores of the reactor-burners (sodium void reactivity effect, control rod worths).

3. The models of fast reactors at BFS critical assemblies for the MA studies

3.1 Fast reactors with Sodium coolant and MOX fuel

The Models of fast reactors with sodium coolant and MOX fuel investigated on critical assemblies BFS-67, BFS-69, BFS-71 and BFS-62. All of these critical assemblies, have different sizes, amount of subzones of the enrichment and differ by presence of control rod mock-ups. The specific differences of investigated critical assemblies and measured values are presented in Table 1.

At all of these assemblies the influence of placement of all available NpO_2 (11 kg) in form of BFS pellets, on the main neutronic parameters was investigated.

At whole it should be mention the worsening of the safety parameters due to disposal of Np in the core: the decreasing of control rod worth (see Table 2) and the increasing of SVRE positive value (see Table 3).

Table 1: Critical assemblies with sodium coolant and MOX fuel.

Series	**BFS-67** [4]	**BFS -69** [4]	**BFS -71**	**BFS -62**
Modification	-1 / -2 / - 3	-1 / -2	-1 / -2	-5 / -6 /-7
Model	Super Phenix	CAPRA	Max Enrichment	BN-600 MOX
Characteristic	Benchmark	Benchmark	Benchmark	4 zones model with CR mock-up
Assembly Sizes: R (cm)	35+30 [a)]	38+14 [a)]	37	73+10+14+8
H (cm)	76	75	77	105
Fissile Enrichment (%)	19	40	57	15:18:17:21
Sizes of Np insertion: R(cm)	0 / 15/ 21	0 / 16	0 / 16	0 / 18 / 12
H(cm)	0 / 38 /38	0 / 45	0 / 46	0 / 31 /31
Amount of Np insert (%)	0 / 13 / 7	0 / 14	0 / 16	0 / 11 / 17
Measured parameters				
Spectral indexes	+	+	+	+
Cross-section ratios of MA	+	+	+	+
SVRE	+	+	+	+
Doppler-effect	-	+	-	+
Power realize	-	+	+	-
Small samples worths	+	+	+	+
CRW	+	+	-	-

[a)] – uranium driver thickness

Table 2: The absorption worth at BFS-67 series

Absorber	Absorption worth (%β_{eff})		
	BFS-67-1	**BFS-67-3**	**BFS-67-2**
B_4C (80%)	-2.16 ± 0.12	-2.13 ± 0.13	-2.06 ± 0.12
B_4C nat.	-0.879 ± 0.039	-0.825 ± 0.036	-0.787 ± 0.035

Table 3: Results of SVRE measurements at BFS-67

				SVRE, (%β_{eff})		
№	FR voided.	Cells voided.	Na (kg) voided	BFS-67-1	BFS-67-3	BFS-67-2
1	31	3+4+5+6	6.920	6.2 ± 0.3	-	18.8 ± 0.8
2	1	4+5	0.112	0.157±0.010	0.324±0.017	0,398±0.008

3.2 The Models of fast reactors with sodium coolant and metal fuel

At these critical assemblies (Table 4), only rate reactions ratios and small samples worths for the nuclear data testing were measured. There was no the introduction of NpO_2 in the cores.

Table 4: Critical assemblies with sodium coolant and metal fuel

Assembly		**BFS -55**	**BFS -73**	**BFS -75**
Model		-	KALIMER	KALIMER
Characteristic		Benchmark	Benchmark	Benchmark
Fuel		Pu	U	U
Assembly Sizes:	*R (cm)*	51	55	26+18 [a)]
	H (cm)	102	98	103
Fissile Enrichment (%)		10	19	15:20

[a)] – uranium driver thickness

In common the results of the analysis of these measurements are considered acceptable [5].

3.3 Fast reactor with sodium coolant and fuel with inert matrix

It should be mention that except previously have carried out experiments at BFS-58 critical assembly, the new series of BFS-91 critical assemblies were accomplished. At this series the Doppler effect, spectral indexes and small sample worths were measured.

The investigation on the ROX-fuel problem was carried out as on the model with fast neutron spectrum (BFS-91-1 critical assembly), so as on the model with more softer neutron spectrum – BFS-91-2 critical assembly.

3.4 Fast reactor models with Pb and Pb-Bi coolant and nitride fuel

Fast reactor models with Pb and Pb-Bi coolant and nitride fuel were studied at BFS-61, BFS-77, BFS-85 and BFS-87 critical assemblies [6]. Till now days the nitride fuel was modeling by means of the metal fuel (U or Pu) with introduction of some materials (for example C), to be available at BFS facilities, for adjusting of the neutron spectrum of the model and the design. At these assemblies (see Table 5), also only the reactions ratios rates and small samples worths of MA for the nuclear data testing were measured. The introduction of NpO_2 in the cores was not carried out.

Table 5: Critical assemblies with Pb and Pb-Bi coolant and nitride fuel

Assembly		**BFS -61**	**BFS -77**	**BFS -87-1**
Model		-	BREST-300	-
Characteristic		Benchmark	Benchmark	Benchmark
Fuel		**U-Pu-C**	**U-Pu-C**	**U-Pu-C**
Assembly Sizes:	***R (cm)***	45	40+13 [a)]	37
	H (cm)	86	110	66
Fissile Enrichment (%)		15	11	25

a) – uranium driver thickness

Except of these experiments, at the series from two critical assemblies BFS -95-1 and -2, which as BFS-77 were BREST-300 reactor mock-up, but with another heterogeneity of the core cell, the influence of the plutonium composition (^{240}Pu content) on fission cross section ratios and small samples worths of MA were investigated.

3.5 Molten Salt Reactor (MSR) with graphite reflector (the program is in preparation)

The analysis of MSR physics shows that for the calculations of this reactor the knowledge of data of wide actinides spectrum is demand. Not only data of Np, Am and Pu isotopes, but Cm, Bk and Cf also cause the significant input, and that is should be tested in critical experiments.

The possibilities of the BFS stands permit to assemble the model close to the spectrum parameters to MSR, to investigate the physical aspects of such reactor and the data of MA by methods presented above.

3.6 Accelerated Driven Systems, ADS (the program is in preparation)

The BFS facilities have the needed set of the materials for the simulation of the reactor part of Accelerated Driven Systems: the plutonium fuel with different vector, the uranium dioxide depleted and enriched, lead and lead-bismuth (about 200t) and the necessary structural materials. One of the advantages of ADS are their possibility to burn the long-lived fragments of fission and MA.

Now days in Dubna (Russia) the design of the facility with Subcritical Assembly (SAD) with combined neutron spectra driven by proton accelerator at proton's energy 660 MeV for experiments on long lived fission products and Minor Actinides transmutation is under investigations. For the solving of this task at BFS-1 stand the assembly simulated the composition and the sizes of the reactor part of SAD will by created. The aim of this joint calculation-experimental work, carried out by specialists from several Russian institutions (JINR, NIKIET, IPPE, ITEP), is the experimental testing and choosing of the methods, will be utilize for the basing of the nuclear safety during SAD facility start-up and the definition of the operational parameters (first of all the methods of subcritical level control in the wide diapason with taking into account of the peculiarities of the neutronic parameters of the SAD, method of effective part of the delayed neutron measurements - βeff – the experimental reactivity scale). The experimental checking of the main neutron-physical parameters of the SAD core planed to be carried out also (the dependence of the reactivity from the sizes and the compositions of subzones, power-fission rates- distributions, the fission cross sections for ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm and ^{245}Cm) for the verification of the calculational design tools used. The results of the analysis of the carried out experiments will be used for licensing of the SAD design and during working out of the physical and power start-up program .

The program of the experiments at BFS facilities will be include two phases: in critical state , when measuring the main reactor parameters and working through the methods of the deep negative subcritical level measurements and in subcritical state, under different Keff, when comparing different method choosing the better for further utilization in SAD facility. For all phases of this program the measurements of fission cross sections and small samples worths of MA by methods presented above planed to be carried out for the verification of the constants on the neutron spectrum typical for ADS.

4. Conclusion

1. The studying of MA for traditional cores with oxide fuel and sodium coolant is finished. During the work on this Program the recommendation on cross section correction for Np-237 and Am-241 capture in the energy diapason of neutrons for the fast reactor MA burner already have been received.
2. The work on investigations of cores with Pb and Pb-Bi coolant is in process now.
3. The investigation of MSR and SAD is under preparation now.
4. After this it is seems that experimental data base will be fulfilled. The analysis of the experiments carried out are planed in the frames of the international joint projects (ISTC, IRPhEP etc.) and will include :
   - the calculational analysis of the measured values and their sensitivity to the nuclear parameters of the minor actinides
   - the providing of the demand precision of the calculational simulation of the accelerated driven systems and the other MA burners
   - the working out and the improvement of the computational tools, used for the analysis of the ADS and others MA burners
   - the working out of the files of the estimated MA nuclear data, based upon the results of the experiments carried out.

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