Integral experiments with minor actinides at the BFS critical facilities: state-of-the-art survey, re-evaluation and application

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Abstract. The paper presents the results of a computational and experimental analysis of a systematized and revised series of experiments carried out between 1990-2013 on measurements of absolute fission rate of minor actinides (from ^{237}Np to ^{245}Cm) in different neutron spectra at the BFS-1,2 facilities. A total of 25 critical configurations, i.e., reactor core models with different fuels and coolants were examined. The earlier experimental data have been revised according to more accurate data processing methods with account of permanent chamber deformations and introduced corrections to the efficiency of detecting fragments (fission events in the chambers). The computational models of assemblies presented in the international handbooks were supplemented with evaluated fission rate ratio models using non-analog algorithms. The resulting consistent set of experimental data and computational models can be used in solving various applied and fundamental problems. A generalization and re-evaluation of a series of integral experiments at the BFS facilities can serve as an information base for the verification and refinement of evaluated minor actinides neutron data. The analysis of all the available set of data on minor actinides fission rate ratio measurements can be used for supporting rationale and planning of research programs for critical assemblies.

Key Words: minor actinides, BFS critical assemblies; reaction rate ratios, integral experiments

1. Introduction

The feasibility and performance evaluation of ways involving minor actinides (MA) in the nuclear fuel cycle of nuclear power to reduce the radiation exposure of nuclear waste requires reliable information on nuclear data characterizing neutron interaction with MA. However, due to the lack of reliable experimental information as well as the accompanying problem of large uncertainties in MA nuclear data, it becomes impossible to achieve the required accuracy in calculations of reactor characteristics and spent nuclear fuel composition, which complicates the justification of reactor designs for MA transmutation and requires efforts to improve the accuracy of MA nuclear data.

There are several possible approaches to solving this problem, i.e., neutron data improvement based on more accurately measured characteristics of nuclear interactions as well as accounting and re-evaluation of the existing experimental information. So far as the capabilities of experimental techniques are limited and conducting new experiments requires considerable time and material expenditures, the most realistic way to reduce uncertainties in nuclear data is to use in calculations the neutron data obtained with account of all data sets (including previously unaccounted) on integral and differential experiments.

Up to date, there is a sufficient amount of differential experimental data on MA, i.e., direct measurements of neutron cross-sections at different energies. However, the use of only the differential measurement results without regard to the integral measurement information for the preparation of evaluated nuclear data leads to serious errors in the calculated values of reactor characteristics. Integral experiments make it possible to directly determine the accuracy of estimated reactor characteristics, i.e., energy integrals of neutron fluxes by neutron cross-sections, whereas differential experiments are a source of detailed information about the energy dependence of neutron cross-sections. In contrast to the differential ones, the integral experiments allow the nuclide characteristics to be reflected with due account for the neutronics of a multiplying system. The possibility of taking measurements in a critical system where a neutron spectrum is formed close to that of the reactor under study increases the practical significance of integral experiments.

The integral experiments designed to reduce uncertainties in MA cross-sections were carried out in different years at various research reactors and facilities, e.g., FR-5, BFS (JSC "SSC RF – IPPE"), MASURKA, MINERVE (CEA, France), FCA (JAEA, Japan), VENUS (SCK-CEN, Belgium, ZPPR, FLATTOP (LANL, USA), ZEBRA (UKAEA, England), etc. These experiments showed that to improve the accuracy of nuclear data and reduce the computational uncertainties in the reactor characteristics is possible by jointly using integral and differential experiments [1-4].

A generalization and re-evaluation of a series of integral experiments at the BFS facilities which were supplemented with precision calculation models can serve as an information base for the verification and refinement of evaluated MA neutron data. The analysis of all the available set of data on MA fission rate ratio measurements carried out at the BFS facilities can be used for supporting rationale and planning of research programs for critical assemblies.

2. Complex of BFS Large Critical Experiment Facilities (JSC "SSC RF – IPPE")

The complex of large critical experiment facilities at the JSC "SSC RF – IPPE" includes two critical facilities, BFS-1 and BFS-2, providing a unique experimental base for a wide range of research programs in the field of reactor physics, critical safety and applied nuclear engineering problems. Over the past several decades, the BFS experimental data have been widely used to improve nuclear data and codes for calculational support of industrial and research reactors. Due to a wide range of nuclear fissile materials at the BFS facilities, it is possible to perform experimental studies of fast reactor cores designed to solve the problems of MA transmutation, uranium-233 production, plutonium utilization, etc.

At the BFS facility, integral experiments (including benchmark ones) are carried out intended to study the neutronic of research and industrial fast reactors with a capacity up to 1000 MW (t) with different fuels (uranium metal, uranium dioxide and nitride, plutonium metal, MOX fuel) and coolants (sodium, lead, lead-bismuth, hydrogen-containing coolants).

The BFS-2 facility is the world's largest critical facility; its dimensions (height – 3 m, diameter – 5 m) make possible a full-scale simulation of the cores and shields of fast reactors with a capacity up to 3000 MW as well as in-vessel shielding and storage.

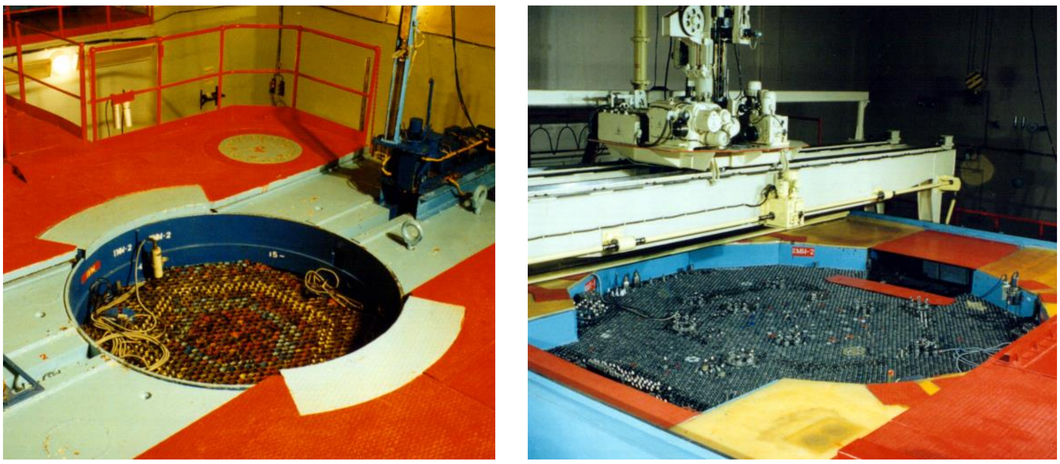

FIG.1. Complex of the BFS-1 (left) and BFS-2 (right) facilities.

Within the framework of the International Criticality Safety Benchmark Evaluation Project (ICSBEP) and International Reactor Physics Experiment Evaluation Project (IRPhEP) [5-7], a part of experiments carried out in different years at the BFS-1 and -2 facilities were thoroughly described and supplemented with precision (Monte-Carlo) calculation models. In total, eight evaluations of benchmark experiments carried out at about 30 core configurations of critical facilities were included in the IRPhEP and ICSBEP Handbooks. These experiments covered such research areas as the study of fast reactor characteristics with sodium and lead coolants as well as the study of the minimum critical mass in the MOX fuel production. The majority of benchmark experiments included in the IRPhEP and ICSBEP Handbooks contain not all but a few experimental data on MA reaction rate ratio measurements taken at the BFS facilities and the presented data were not evaluated as benchmark models, therefore they could not be used for solving applied problems.

3. A List of MA Experimental Programs Performed at the BFS Facilities

The experimental programs for the study of MA properties in the reactor conditions at the BFS facilities were performed for the following isotopes: uranium, plutonium, ^{237}Np, ^{241,243}Am, ^{244,245}Cm. The measurements were taken at the BFS facilities of different configurations similar in composition to a particular type of industrial or research fast reactor. The BFS configurations, i.e., fast reactor models, are composed of structural materials, a coolant and tubes filled with fuel pellets in the amounts corresponding to the content of these materials in the core or fertile fuel zone under study.

The MA nuclear properties were studied in neutron spectra formed in the cores with different fuels (U, Pu, UO_2, UN, MOX), coolants (Na, Pb, Pb-Bi, hydrogen-containing coolants) and structural materials (stainless steel, Al, Ni, Ti, Nb, Zr, C, B_4C, Al_2O_3, etc.). The extensive research programs of MA experiments at the BFS facilities included the measurements (Table I):

- of the spectrum average MA fission rate ratio (^{238,240}Pu, ^{237}Np, ^{241,243}Am, ^{244,245}Cm) to the ^{239}Pu fission rate, (Fi/F49), by absolute fission chamber (ACs);
- of the spectrum average MA fission rate ratio ($^{238,240\text{-}242}$Pu, ^{237}Np, ^{241,243}Am, ^{244,245}Cm) to the ^{239}Pu fission rate, (Fi/F49), by small-scale fission chamber (SSCs);
- of the spectrum average MA fission rate ratio (^{238,242}Pu, ^{237}Np, ^{243}Am) to the ^{239}Pu fission rate, (Fi/F49), by triple-segment fission chambers (TSCs);
- of the spectrum average capture rate ratio of ^{237}Np and ^{241}Am to the ^{239}Pu fission rate, (Ci/F49), by foil activation;
- of the relative reactivity ratio of ^{237}Np and ^{241}Am samples to the ^{235}U sample reactivity, (Ri/R25); and
- of the Doppler effect when the heated ^{237}Np samples were moved into the core central channel.

All of these measurements were generally taken in the core central channel, i.e., a cavity formed by removing the central fuel tube. Reactivity was compensated by additional loading of fuel rods in the core periphery. Thus, each of these measurements entails a slight modification of the initial fuel loading scheme.

Data on all loadings have been digitized and compiled into the PROTVA database of calculation tasks on experiments at the BFS critical facilities for the MMK (transport code implementing the Monte Carlo method), TRIGEX (three-dimensional diffusion reactor calculation program) and FFCP (cell code implementing the first collision method with nuclear data in the sub-groups representation) codes. The PROTVA database does not include experimental data or a description of the specifics of the preformed measurements; it contains data on fuel loading schemes and tube compositions (i.e., a list of materials and their configuration in the core).

Most of the experimental data on MA integral experiments at the BFS critical facilities have been published over the years. In order to systematize the accumulated information and provide its re-evaluation and use to test and correct the evaluated nuclear data files, verify the calculation codes as well as select and create problem-oriented sets of MA evaluated nuclear data files, activities have been initiated on forming a database on MA integral experiments at the BFS critical facilities. This database is intended to supplement the aforementioned existing information and software tools for the BFS experimental programs.

The database covers the MA integral experiments (including the previously unpublished ones) carried out in different years at the BFS facilities of various configurations and compositions to simulate both reactor systems and external fuel cycle facilities/processes. The data base includes a description of the BFS configurations and experimental conditions, experimental values of reaction rate ratios measured by various methods, topical collection of publications reflecting the results of performed experiments, and a set of calculation tasks for MCNP which may reproduce the experimental conditions. To exemplify the use of the database being created, the results of calculation-to-experiment discrepancies (in measurements by ACs) in the spectrum average MA-to-^{239}Pu fission rate ratio are presented.

TABLE I: A LIST OF MA CONFIGURATIONS AND MEASUREMENTS AT THE BFS FACILITIES.

Year	**Coolant**	**F37**	**F48**	**F40**	**F51**	**F53**	**F64**	**F65**
1990	NA	☒	☒	☒	☒	☒	☒	☐
1993		☒	☒	☒	☒	☒	☒	☐
1994		☒	☒	☒	☒	☒	☐	☐
1996		☒	☒	☒	☒	☒	☒	☒
1997		☒	☐	☐	☒	☒	☐	☐
1990	Pb&PbBi	☒	☐	☐	☒	☒	☒	☐
1991		☒	☒	☐	☒	☐	☐	☐
1999		☒	☒	☒	☒	☒	☒	☐
2000		☒	☐	☐	☒	☒	☐	☐
2001		☒	☐	☐	☒	☒	☐	☐
2004	CH_2	☒	☐	☐	☒	☒	☒	☒
2008		☒	☒	☒	☒	☒	☐	☐

4. Specifics of Computational Models

A detailed description of the BFS facilities is given in the international handbook [5,6]. The critical assemblies are composed of tubes which are filled with pellets of reactor materials 46 mm in diameter (Fig.2). The thickness of the pellets ranges from fractions to tens of millimeters, which makes it possible to compose assemblies of different heterogeneity and, among other things, study the cross-section self-shielding effects.

The spectrum average fission rate ratios are calculated using the formula (1). For calculations, it is required to integrate with respect to energy the flux product averaged over the geometrical region of interest (calculated volume) for the corresponding reaction cross-sections.

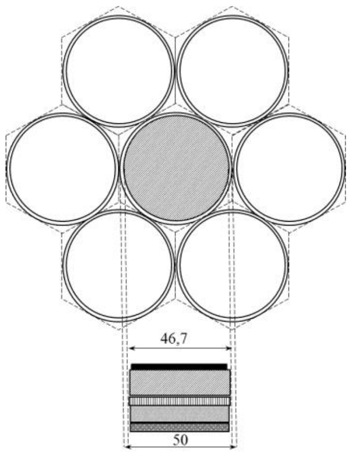

FIG.2. Arrangement of pellets in the BFS tubes.

A computational model of detection region measurements taken using an AC is shown in Figure 3. To improve statistics in the calculations by MCNP, the AC real detection region geometry (fissile material layer 20 mm in diameter) is substituted for a dummy region and a detection region is introduced (shaded in Fig. 3). The results of the calculations compared in the detection region real and dummy geometries coincide within the computational accuracy; therefore, the systematic error of this transformation can be neglected [8-11]. On this basis, detailed calculation models were drawn for the MCNP code which were partially included into the databases of reactor experiments of the IRPhEP and ICSBEP Handbooks. The computational analysis presented below was carried out using MCNP and the ENDF/B-VI.8 Library.

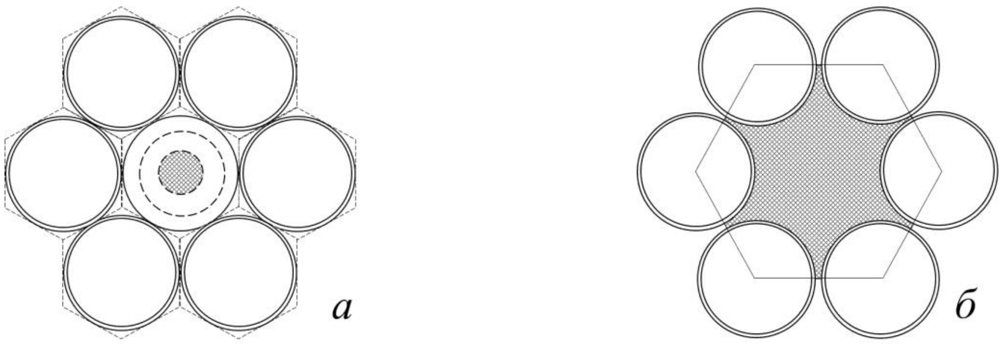

FIG.3. Detection area in measurements (a) and calculations (b).

To perform serial calculations, considerable computer time expenditures are required. The computational analysis showed that in order to achieve the required accuracy for calculated characteristics, it is necessary to use non-analogous calculation methods. An optimal variance reduction algorithm was determined to perform calculations using a single-core processor and it was shown that the obtained calculation time values could be reduced by orders of magnitude using multi-core processors. The results were checked as for the absence of discrepancies in series of direct calculations with large statistics (on a supercomputer for the MCNP parallelized version) for different random-number sequences. The calculation results obtained using variance reduction methods were compared with the direct calculation results with an estimated accuracy of less than 1.5%; the differences in the results of direct and indirect calculations did not exceed 1%. The results of the analysis have shown that the combination of spatial-energy splitting in the method of weight windows leads to a significant increase in the calculation efficiency and a reduction of computer time from a few days to a few hours at a statistical error in the reaction rate ratio of less than 2%.

5. Analysis of Calculation-to-Experiment Discrepancies

Since the considered set of experiments represents a combination of different studies performed in different years at the BFS facilities, it is necessary to select an index for their classification. This index can be the ^{238}U-to-^{239}Pu fission rate ratio, (F28/F49). It is convenient to use this ratio as a measuring scale (or a classifier) because its values characterize the neutron energy spectrum in an assembly. The higher the energy of neutrons causing fission is, the lower the values of F28/F49 are. In the considered series of measurements performed at the BFS facilities of different compositions and types, the values of F28/F49 vary in a wide range of 0.01–0.06.

Table 2 provides information on the MA fission cross-section ratio measurements. The upper values in the columns show the number of experiments, the lower one is the averaged over all assemblies C/E values. Based on Table 2, it can be concluded that the calculation and experimental values of the fission cross-section ratio of ^{237}Np, ^{238,240}Pu, ^{241}Am to the ^{239}Pu fission cross-section are consistent within the measurement error (±1σ). The discrepancies in the ^{243}Am, ^{244,245}Cm to ^{239}Pu are twice as high but still they do not exceed the boundaries determining experimental values (±3σ).

TABLE II: SUMMARY OF THE NUMBER OF MA EXPERIMENTS (*N*), AVERAGE VALUES OF C/E DISCREPANCIES (Δ), ACCURACY REQUIREMENTS.

Coolant type	**Data**	**F*i*/F49**					
		^{240}Pu	**^{237}Np**	**^{241}Am**	**^{243}Am**	**^{244}Cm**	**^{245}Cm**
Na	*N*	13	14	14	14	12	3
	±Δ,%	3,7	2,2	5,2	9,5	9,2	9,4
Pb (Pb-Bi)	*N*	5	9	7	8	4	1
	±Δ,%	2,6	2,6	4,4	6,3	11	12
All	*N*	20	27	25	26	18	6
	±Δ,%	4,4	2,7	4,6	7,9	12,1	8,3
Measurement error	±1σ,%	3	2,5	3	3,5	4	5
One-group average fission cross-section [4]							
Uncertainty related to nuclear data,%		5,8	7	10	11	50	47
Target accuracy,%		1,8	1,5	2	2,5	5	7

The maximum negative C/E discrepancy for the fission rate measurements of ^{237}Np и ^{241,243}Am is 4 and 12%, respectively, for assemblies with neptunium dioxide in the core composition. The maximum positive discrepancies, up to 4.5% for ^{237}Np are observed in assemblies with a hydrogen-containing diluent. Large positive discrepancies for ^{241,243}Am are typical for assemblies with MOX fuel. The calculated and experimental values of the ^{245}Cm fission rate are consistent within the measurement accuracy for systems with oxide fuel but inconsistent (up to 14%) for systems with other fuels. The largest discrepancy (~ 30%) for ^{244}Cm was revealed at the BFS-97-3 assembly with a hydrogen-containing diluent (polyethylene). The assemblies of the BFS-97 series are not typical for the BFS critical facilities: the core composition contained a large amount of polyethylene (polyethylene pellets in the core cells and polyethylene follower in the intertubular space). The assemblies of this series are designed to simulate processes at the technological stages of the MOX fuel production and transportation. During the calculation support of the BFS experiments, it was noted that the assemblies with hydrogen-containing materials tend to have larger C/E discrepancies.

6. Conclusion

Improvements of MA nuclear data to increase the accuracy of reactor characteristics require, on a par with the further supplementation of experimental databases with new neutron cross-section differential measurements, a wider use and accumulation of information about the integral measurements. The feasibility of creating neutron spectra similar to those of the reactors under study increases the practical value of integral experiments since it makes it possible to reflect the MA properties, which are characteristic of a given spectrum and thus reduces the calculation uncertainty of reactor performance caused by the MA nuclear data uncertainty. The formed database on MA measurements at the BFS facilities can be used to test and adjust evaluated nuclear data files, verify calculation codes. The results of the analysis of all the MA integral experiments can be a starting point for planning further BFS experimental programs on the study of MA characteristics in fast reactors.

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