Determination of Group Neutron Cross-Sections and Their Integral Characteristics for Minor Actinides by GRUCON Code Based on Estimated Data of ENDFB, JENDL, JEFF, BNAB

Yu.V. Grigoriev[1,2], E.A. Koptelov[1], O.N. Libanova[1],
Zh.V. Mezentseva[3], A.V. Novikov-Borodin[1], V.V. Sinitsa[4]

[1]*Institute for Nuclear Research RAS, Moscow, Russia*
[2]*Joint Institute for Nuclear Research, Dubna, Russia*
[3]*Institute of Physics and Power Engineering, Obninsk, Russia*
[4]*National Research Centre "Kurchatov Institute", Moscow, Russia*

Abstract. Utilization of radioactive wastes of nuclear power engineering is one of the urgent tasks, because nowadays there are hundreds of tons of long-living fission fragments and minor actinides, which need to be kept in special radioactive waste storages or to be transmuted into short-living isotopes. In this connection it is necessary to elaborate the waste transmutation techniques and to solve complicated technical problems, in particular to create a database of necessary scientific and technical information of neutron and other nuclear-physics values. As for neutron constants, there are insufficiently known the neutron cross-sections of radiation capture of fission fragments and cross-sections of fission and capture of the lower minor actinides: isotopes of neptunium, americium, curium, thorium, plutonium and uranium. In this paper we suggest to measure the cross-sections of the isotopes by means of TOF and neutron time slowing-down technique in the energy range from 1 eV to 200 keV by using the new fast fission chambers. To estimate the efficiency of measurement techniques the group neutron cross-sections of isotopes mentioned above by using GRUCON code based on estimated data of ENDFB, JENDL, JEFF, BNAB have been calculated. The calculated group cross-sections point out that there are large errors from 10 to 30 % in cross-section values in resonance range.

Introduction

Estimation of the efficiency of measurement techniques by analyzing the group neutron cross-sections of lower minor actinides: isotopes of neptunium, americium, curium, thorium, plutonium and uranium with help of GRUCON code by using data of ENDFB, JENDL, JEFF, BNAB shows that there are large errors from 10 to 30 % in cross-section values in resonance range. Knowing the neutron cross-sections of radiation capture of fission fragments and cross-sections of fission and capture of the lower minor actinides is important for utilization of radioactive wastes of nuclear power engineering. It is one of the urgent tasks, because nowadays there are hundreds of tons of long-living fission fragments and minor actinides, which need to be kept in special radioactive waste storages or to be transmuted into short-living isotopes. In this connection it is necessary to elaborate the waste transmutation techniques and to solve complicated technical problems, in particular to create a database of necessary scientific and technical information of neutron and other nuclear-physics values.

There is proposed in this paper to measure the cross-sections of the isotopes by means of TOF and neutron time slowing-down technique in the energy range from 1 eV to 200 keV by using the new fast fission chambers. Both of these techniques may be realized on LSDS-100 neutron spectrometer (Ref. [1–3]) of Experimental complex of Institute for Nuclear Research RAS.

1. **The GRUCON code**

The GRUCON code is a system of modules for evaluated nuclear data processing for production of detailed and multi-group working libraries for transport calculations in reactor physics and radiation shielding applications. The package has an original architecture and command language.

The GRUCON project was started in the Laboratory of Constants of Institute for Physics and Power Engineering (IPPE) at the beginning of the 1970's, Ref. [4–7] from the development of formats for storing the data from microscopic experiments and also evaluated nuclear data aimed at generating computer libraries. The evaluated data should be non-contradicting, unambiguous and complete (in view of the application they are intended for). In order to satisfy these requirements, the measured data undergo an evaluation procedure that includes the analysis of contradictions between the data from various experiments and the selection of parameterization methods that ensure an unambiguous restoration of selected data. Where the evaluation procedure is impossible (due to the lack of measured data), the parameters are calculated on the basis of theoretical models, or determined from systematics.

The GRUCON project was resetted in NRC "Kurchatov Institute" in the year 2012 to create a program alternative to US data processing program NJOY, which de facto became a standard data processing tool adopted everywhere in the world. The reset was caused by realizing the shortcomings of program NJOY and the necessity to update the computer libraries of constants to be used in mathematical simulation of physical processes of nuclear plants in order to substantiate their nuclear safety.

The method based on the principle of data normalization is used in the development of program GRUCON. In the great number of data used in a certain application sphere, there is distinguished a set of structures, which are elementary enough to make the task of plotting the algorithms that will unite these structures easier for a programmer, at the same time, these structures are universal enough to combine the variety of methods for representing every type of the data and to make it easy for user to control the computational process.

Each structure is assigned in a uniform manner – in the form of a standard representation consisting of a heading and a data array. The heading includes the information necessary for searching and exchange of representations between different parts of the memory. In the data array, the structure of the data is put in univocal correspondence with the heading of the representation.

The normalization of the data structures together with the data representations allows consolidating the algorithms of data search and data exchange in a small set of modules of the system support which significantly simplify the work of the programmer of functional modules by providing an interface between them. The simplicity of module interface based on normalized data allows transferring the function for composing computational procedures and organization of computational process to the user and providing him with important tool – language defining the problem to be solved

The strong point of normalized data is that they guarantee a minimal dependence on exterior formats of data representation. The linkage with exterior "non-standard" structures is ensured by a set of modules-converters (designed for input/output of data) to be developed separately from the main program based on data processing algorithms.

Program package GRUCON consists of a set of modules based on cross-section data conversion algorithms. These data are important for generating the working files of cross-sections for computer programs from the libraries of evaluated nuclear data. The set of modules were created on the basis of data structures derived using the methods of normalizing the data used both in the libraries of evaluated data, or the working libraries of detailed and multi-group types.

For the purpose of operational storage of standard data structures, a disc memory is used, where numerated files of direct access are stored. These files form a working library of the program (BSP). Entering the data into the library, as well as search of data in the operational memory and exchange of data with the operational memory are performed using the modules of system support. Library content and addresses of the data to be transmitted to a current module are stored in the operational memory as a library's catalogue and the module's registers.

For the purpose of describing the order in which modules are called and data flow is controlled in the process of data processing ("processing scenario"), a special language is used – the language of program GRUCON. This language consists of the commands which affect the successive execution of the functional modules and the data allocation address at each at each successive processing step.

Functional modules, system support and the programming languages created on the basis of standard data structures allow presenting program GRUCON, as «the package of applied programs with normalized functional content», herein called as «package GRUCON». As the program package develops, the set of standard structures and functional modules can be extended and amended. Meanwhile, the language structures and system content of the package stay, practically, unchanged. This allows considering package GRUCON as the program capable to be extended.

The flow diagram of GRUCON package is shown on Figure 1. The run of data processing program begins with the reading in of set of commands (scenario) by interpreter and entering them to the operational memory. Then, a successive interpretation and execution of each command is carried out.

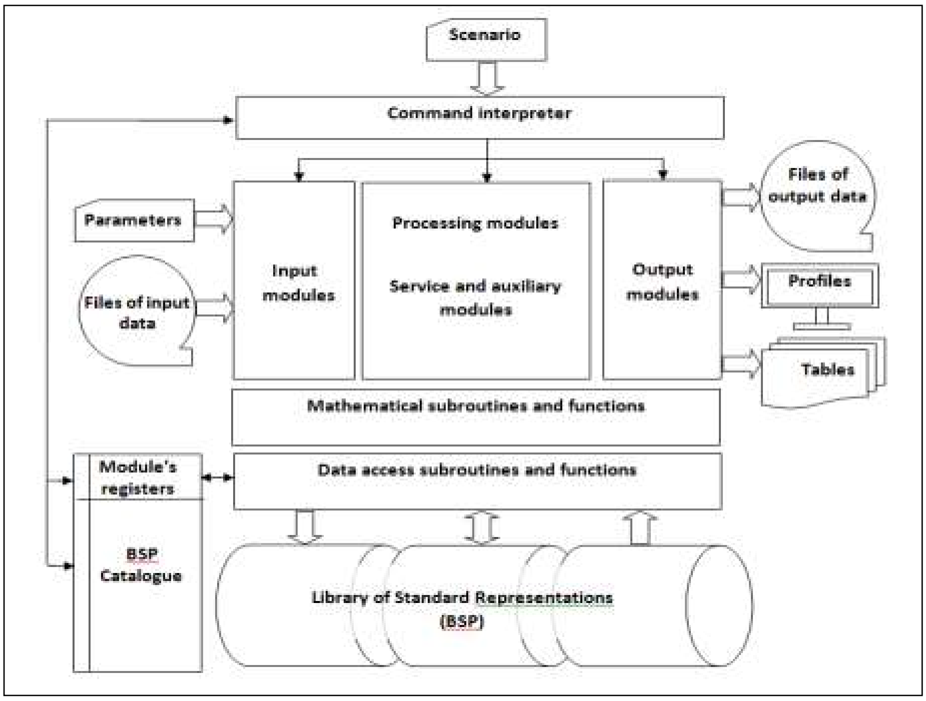

Figure 1. The flow diagram of the GRUCON package.

The data processing commands include the information about address allocations of the data to be processed in the form of a number of BSP catalogue record which contains the registration of these data (the input data), or a number of BSP catalogue record where the data shall be registered (the results of data processing). On the basis of this information, the interpreter calculates the minimum and the maximum number of words occupied, or can be assigned to these data in BSP and operational memory, performs the input determination of the current location addresses, and enters the information into module registers.

Besides the processing commands, scenario may include the servicing commands which indicate the scribing of BSP segments, the display of the catalogue's information content, termination or end of the data processing operation.

After the setting of the addresses, on the basis of the content of the command, the executor of commands defines the name of the module for which these data are intended, and calls the module. The modules can be separated according to the designation in three main types, they are:

- input modules that read in the parameters and data from the files at the peripherals, convert them to internal representations and transform and record them in the library of standard representations;
- data processing modules and a multi-group of service modules and utility (adjustment) modules that read in the processed data and parameters from the BSP library, perform a required computational operation and record the results into the BSP library and register them in the catalogue;
- output modules that read in the data from the BSP library and convert them into tables and records for visual representation in tabulated or graphic form, or for subsequent application in the format of working libraries.

Each record in the library of standard representations is followed by entering the name and allocation address of the data to BSP catalogue – a «data registration».

The package GRUCON-D includes modules allowing to:

- reconstruct cross sections in required energy range for given temperature;
- prepare generalized subgroup parameters with regard to correlations of cross sections of different materials, reactions, temperatures, and as result of collisions, to describe the resonance effects in neutron transport problems;
- calculate the energy-angular distributions of neutrons scattered on the resonances;
- prepare group cross sections and matrices from photo-atomic interaction data library;
- prepare group cross sections from activation data library.

Programming language is Fortran-90, possibility to perform calculations with double and quadruple precision is provided. The version is tested for 32- and 64-bit computers with Windows-7 and Linux-Suse-12.3 operating systems. The verification has been performed by comparison with calculation results obtained by the Prepro-2012 processing code. The distribution package includes installation procedure for Intel, Lahey and gfortran compilers; testing procedure with 12 test problems. Code is certified (certificate of state registration No. 2014663246), its executables (32 and 64-bit) for Linux and Windows, tests and documentation, also as User's Manuals in English and Russian are available on webpage of International Atomic Energy Agency (https://www-nds.iaea.org/grucon/).

2. Estimation of nuclear reaction data

Estimation of the group neutron cross-sections of lower minor actinides: isotopes of neptunium, americium, curium, thorium, plutonium and uranium with help of GRUCON code shows that there are large errors from 10 to 30 % in cross-section values in resonance range in data from different databases: ENDFB, JENDL, JEFF, BNAB and ROSFOND. The cross-sections and group cross-sections estimated by means of GRUCON code are presented on Figure 2. One can see the large difference in group cross-sections from different databases.

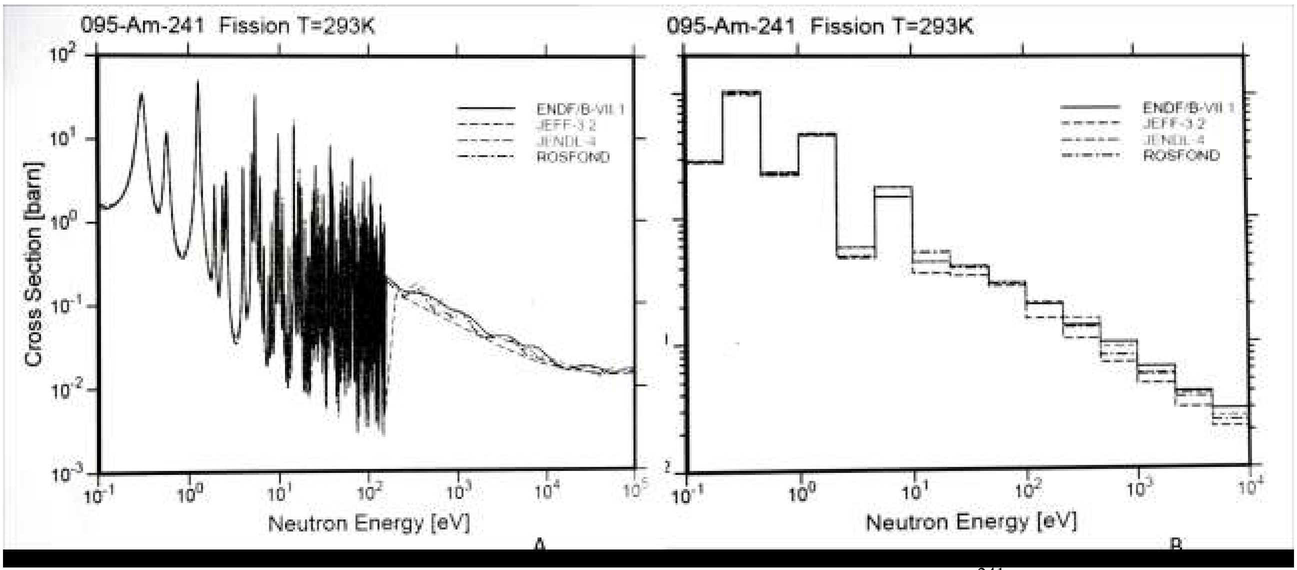

Figure 2. The cross-sections (A) and group cross-sections (B) of ^{241}Am fission.

Group cross-sections correspond to observed values during experiments. Experimental (Ref. [8]) and group cross-sections of ^{232}Th and ^{237}Np calculated by GRUCON code from ENDF and JENDL databases are presented on Figure 3. Some data from databases exceed the experimental errors.

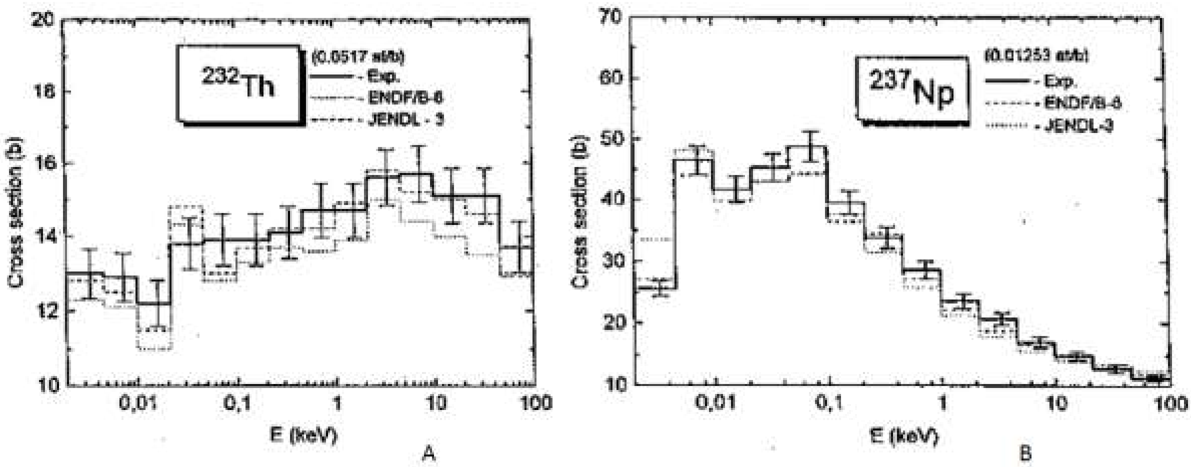

Figure 3. Experimental and calculated observed total cross-sections of ^{232}Th (A) and ^{237}Np (B).

To know precise values of the neutron cross-sections of radiation capture of fission fragments and cross-sections of fission and capture of the lower minor actinides is important for utilization of radioactive wastes of nuclear power engineering. It is one of the urgent tasks, because nowadays there are hundreds of tons of long-living fission fragments and minor actinides, which need to be kept in special radioactive waste storages or to be transmuted into short-living isotopes. In this connection it is necessary to elaborate the waste transmutation techniques and to solve complicated technical problems, in particular to create a database of necessary scientific and technical information of neutron and other nuclear-physics values.

3. Precise measurements of neutron cross-sections

To reach high precisions in measurements of the neutron cross- sections of radiation capture of fission fragments and cross-sections of fission and capture of the lower minor actinides, the time-of-flight method (TOF) is proposed to be used in the lead neutron slowing-down spectrometer (LSDS-100), Ref. [1,3], at Experimental complex of Institute for Nuclear Research (INR RAS). The energy resolution of LSDS is low (30-45%) and is being determined experimentally, but the aperture ratio luminosity of the neutron flux detector is 10^2-10^4 times greater than in the case of a time-of-flight method. It allows to make experiments with small amount of substance and small cross-sections of its interaction with the slowed moderated neutrons. In order to study the neutron fission cross-sections of minor actinides, the new fast ionized fission chambers of IPPE (Institute of Physics and Power Engineering) with thin layers of minor actinides are supposed to use for measurements at the LSDS-100 by the transmutation program.

3.1 TOF technique for LSDS-100

A relation between an energy of lead slowing-down moderated neutrons E (eV) and the time delay of t (μs) is described by the expression: $E(t)=K/(t+t_0)^2$, where values of K=170.5 (keV×μs^2), t_0=0.3 (μs) in the case of the LSDS-100 have been measured in the previous experiment, Ref. [3]. The energy resolution of LSDS is low (30–45%) and is being determined experimentally, but the aperture ratio luminosity of the neutron flux detector is 10^2-10^4 times greater than in the case of a time-of-flight technique. It allows making experiments with small amount of substance and small cross-sections of its interaction with the slowed moderated neutrons.

To expand the experimental possibilities of LSDS-100 with total weight of 100 tons, which is mounted in the form of a parallelepiped of 3348×1620×1728 mm^3 dimensions from lead prisms of 1 and 0.25 ton weights and of 99.996 % natural lead purity, it is proposed to install the polyethylene moderator in the form of a plate of 100×100×30 mm^3 dimensions or of a disk with 100 mm diameter and a thickness of 30 mm in one of available vertical channel-wells over a lead target. Such moderator forms a wide spectrum of neutrons from thermal to fast 200 MeV neutrons. If the moderator is cooled up to the temperature of liquid nitrogen, then the thermal part of neutrons will be shifted to the range of cold neutrons. To improve background conditions and to provide the neutron beam forming, it is necessary to mount inside the vertical channel-well over the moderator the neutron guide, which is the vacuum pipe of 90 mm diameter and of 1–10 m length with the collimators nearby the moderator and at the end of the neutron guide in front of the detector. It is supposed to use as detectors the fast counters of neutrons, γ-rays and the fission chambers. Certainly, due to the moderator the mounting of the proposed equipment of the TOF-spectrometer in the lead body of the LSDS-100 will slightly worsen its energy resolution, but in fact the spectrometer geometry is not changed. The parameters of the LSDS-100 combined with the TOF-spectrometer will be defined experimentally. If the mutual influence of spectrometers is negligible, it is possible to make measurements simultaneously on both facilities. Otherwise the moderator should be removed from the body of the LSDS-100. The TOF-spectrometer may be used also when the LSDS-100 is running. In that case it is necessary to install the good detector shielding from the background neutrons leaking from a body of a lead cube.

With using the TOF-technique, the energy $E(t)$ of neutrons, registered by the detector, is defined by the equation: $E(t) = mv^2/2 = (72.3 \cdot L/t)^2$, where t (μs) is a time of flight of a neutron of distance L (m) from the moderator to the detector. The energy resolution is defined from the following ratio: $\Delta E/E = 2v/L = 0.0276E^{1/2}\Delta t/L$, where v is a speed of a neutron with an energy E, ΔE is an uncertainty of energy, Δt is an uncertainty of time.

The values of the energy resolution of the time-of-flight spectrometer at various energies and flying bases at Δt=2 μs are presented in Table 1.

Table 1. The resolution *ΔE/E* (%) versus the energy of neutrons and the length of the flying base *L*.

***E* (keV) / *L* (m)**	**0.9**	**1.6**	**2.5**	**49**
1	1.67	2.01	2.74	3.66
2	0.83	1.01	1.37	1.66
4	0.41	0.50	0.69	0.96
8	0.21	0.26	0.34	0.48
50	0.033	0.040	0.054	0.060

It is seen from the Table 1 that the energy resolution of the TOF- spectrometer even on the shortest base of 1 m at the energies low than 50 keV is better than of the LSDS-100. As soon as the integral output of the fast neutrons in the lead target of LSDS-100 is approximately 10^{14}n/(cm^2×s) at the average proton current of 5 μA, the integral neutron flow on the neutron absorbing detector surface with square of 100 cm^2 at the distance of 1 m from the pulsed neutron source will be approximately 10^{10} n/(cm^2×s) and at the distance of 10 m will be 10^8 n/(cm^2×s), which is 100 times higher the flow density at the middle of each of five experimental channels of LSDS-100, Ref. [3]. It is worth noting that besides traditional researches on the fundamental and applied nuclear physics the proposed combined spectrometer based on LSDS-100 and TOF-spectrometer may also be used for modeling the different types of targets and to investigate the isomeric states excited by protons in wide range of energies. As soon as there are five experimental cylindrical channels with 65-80 mm diameter, two wells with 100×100 mm^2 cross section square and the thermal graphite prism in the body of LSDS-100, it allows to make various experiments on development of techniques of a transmutation of a long-living waste of atomic power engineering.

3.2 The detection of velocities of the fission reactions

In order to study the neutron fission cross-sections of minor actinides, the new fast ionized fission chambers of IPPE (Institute of Physics and Power Engineering) with thin layers of minor actinides are supposed to use for measurements at the LSDS-100 by the transmutation program.

The technique of determination of the ratio of the average fission cross-sections with help of the absolute fission chambers with known number of nuclei in the layer and known efficiency of the registration of the fission fragments is suitable for determination of the velocities of the fission reactions of the most of trans-actinides, having threshold character, and also for determination of absolute fission velocities of ^{239}Pu and ^{252}Cf.

The scheme of the absolute fission chamber is presented on Figure 4.

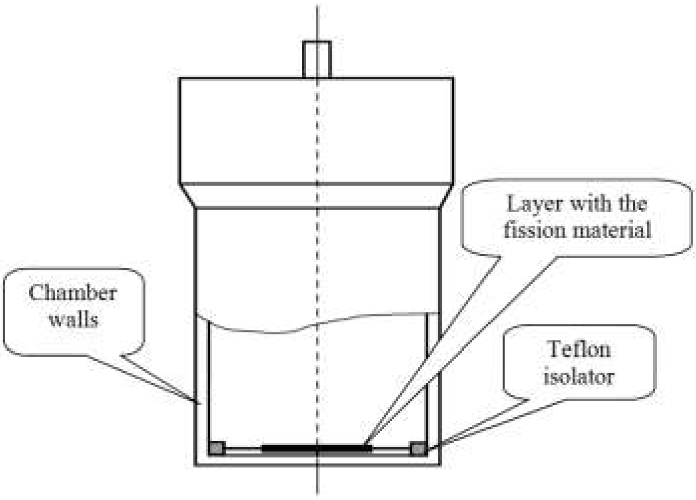

Figure 4. The scheme of the absolute fission chamber.

To calculate the ratio of the average isotope fission cross-section for the technique, where the absolute plane fission chambers of the test stand are used during measurements, there was used the expression:

$$\frac{\sigma_f^i}{\sigma_f^j} = \sigma_f^{i/j} = \frac{N^i \varepsilon^j m^j}{N^j \varepsilon^i m^i} - \sum_k^n \left(a^{k/j} \cdot \sigma_f^{k/j}\right),$$

where

N^i, N^j are the count velocity of the fission chamber with the layer of the *i*-th nuclide and the count velocity of the fission chamber with the layer of the *j*-th nuclide in the active zone;

ε^i, ε^j are the efficiencies of the fission fragments registration by the fission chamber with the layer of *i*-th nuclide and by the fission chamber with the layer of *j*-th nuclide correspondingly;

m^i, m^j are the number of nuclei in the layer of *i*-th nuclide and *j*-th nuclide correspondingly;

i, j are the atomic number of being measured and reference nuclide correspondingly;

k are the atomic numbers of doped nuclides, containing in the active layer of the measured (*i*-th) nuclide;

$a^{k/j}$ is the ratio of the number of nuclei of doped nuclides to the number of nuclei of the basic nuclide (being measured), $\sigma_f^{k/j}$ is the ratio of the average fission cross-sections of doped nuclides to the number of nuclei of basic nuclide. It is supposed here for simplicity that the contaminants in the layer of the basic nuclide may be neglected (see Ref. [1] for details).

To revise the full and partial neutron cross-sections with errors ≤ 5% and their integral characteristics (the resonance parameters, the factors of the resonance interlock and the Doppler's coefficients with errors ≤ 15%) of the constructive reactor materials Ti, Mn, Cr, Fe, Ni, Ta, Mo, the necessary experiments by the time-of-flight technique at the neutron source RADEX and LSDS-100 with TOF of INR RAS have been made. It is expected to make measurements of the time-of-flight spectrums for the samples of filters and radiators of thickness 40 mm, 20 mm, 5 mm, 2 mm, 1 mm on the path length of 50 m of the neutron pulsed source RADEX on the experimental installation (the resonance cross-section of transmission) with the fast 8-sectional liquid-based (n,γ)-detector and the high efficient neutron He-3 detector. The experimental time-of-flight spectrums will be stored in four independent measuring modules with different durations of the time channels τ=0,20 μs, 1 μs, 2 μs, 4 μs in the group energetic intervals of the constant system BNAB in the energy range from the thermal neutrons 0.1 eV up to the fast ones 200 keV.

To achieve the required accuracy of the being obtained nuclear-physics values of the mentioned above materials, it is necessary to have the stable operation of the proton accelerator at the experimental complex during 14 operating shifts with the proton energy 209 MeV, the pulse current 16 mA, the time duration of the neutron flashes 5–20 μs and the frequency of the neutron flashes 50 Hz.

In case of the considerable losses during the beam transport, it is possible to use the usual 7 mA beam current by turning off of the first and the second bunches.

There have been determined from the experimental spectrums the cross-sections, transmissions, self-indications and other integral characteristics, for example, the factors of the resonance interlock with the error less than 5%. Besides of that, the deep interference dips will let to estimate the energy dependence of the cross-section of the (*n,e*) scattering.

Acknowledgements

Dedicated to memory of Prof.Yu.V. Grigoriev, who was a leader and an inspirer of this project.

References

1. Alekseev A.A., Grigoriev Yu.V., Dulin V.A., Libanova O.N., Novikov-Borodin A.V., Matushko V.L., Mezentseva Zh.V., Ryabov Yu.V. The TOF method for the LSDS-100 spectrometer. – Proc. Int. Seminar ISINN-23, Dubna, JINR, 2016.
2. Popov Yu.P. Spectrometry of lead neutron slowing-down spectrometer. – PEPAN, **26**, pp.1503–1523, 1995 (in Russian).
3. Alekseev A.A., Bergman A.A., Berlev A.I., Koptelov E.A. Lead neutron slowing-down spectrometer (LSDS-100). / Neutron complex of INR RAS. – Preprint INR RAS 1258, 2010 (in Russian).
4. Nikolaev M.N. Nuclear Data for the Calculation of Fast Reactors. – Moscow: Atomizdat, Problems in Atomic Science and Technology, ser. Nuclear Constants 8(1), 1972 (in Russian).
5. Kolesov V.E., Nikolaev M.N. Format of the Recommended Data Library for Reactor Calculations. – Moscow: Atomizdat, Problems in Atomic Science and Technology, ser. Nuclear Constants 8(4), 1972 (in Russian).
6. Abagyan L.P., Bazazyants N.O., Bondarenko I.I., Nikolaev M.N. Group Constants for Reactor Calculations – Moscow: Atomizdat, 1964, 1–139 (in Russian); New York, Consultant Bureau, 1964 (in English).
7. Sinitsa V.V., Abagyan L.P., Bazazyants N.O., et al. GRUKON – Library of Computer Programs for Calculation of the Group Constants. – Moscow: Atomizdat, Nuclear Physics Research in the USSR, Computer Program Descriptions, vol.**27**., 1979 (in Russian).
8. Grigoriev Yu.V., Sinitsa V.V., Gundorin N.A., Popov Yu.P. Investigations of the Resonance Structure of Neutron Cross-Sections for Thorium-232 and Neptunium-237 in the 2 eV–100 keV Energy Region. – VANT, Nucl. Data, **1**, p.9, 1998.