DOI 10.1007/s10512-019-00556-8
Atomic Energy, Vol. 126, No. 5, September, 2019 (Russian Original Vol. 126, No. 5, May, 2019)

^{239}PU–BE-SOURCE BASED NEUTRON REFERENCE FIELDS

V. N. Peleshko,[1] E. N. Savitskaya,[1] A. V. Sannikov,[1]
M. M. Sukharev,[1] and A. G. Muzoverov[2]

UDC 539.1.074.3

New neutron reference fields, based on a ^{239}Pu–Be source, intended for increasing the accuracy of measurements of neutron dosimeters and radiometers in the IHEP accelerator complex are described. A Bonner SB-RSU-01 spectrometer was used to measure the spectra of the neutron reference fields. The BON95 code was used to reconstruct the spectra; the initial spectra for the iteration procedure were determined by parameterization or by a calculation using the FAN15 code. Good agreement was obtained between the neutron spectra and the integral characteristics of the reference fields reconstructed by the two methods.

The neutron spectra in the accelerator complex at the Institute of High-Energy Physics (IHEP) vary over a wide energy range and differ significantly from the spectrum of a calibrated source. Under such conditions, reference fields with expanded energy range of the spectra are created in order to take account of the energy dependence of the sensitivity of neutron dosimeters and radiometers. At IHEP such fields were created on the basis of ^{239}Pu–Be and ^{252}Cf sources [1]. The service life of these sources is now exhausted; in addition, it has been noted in the ^{252}Cf-based reference fields that the time-dependence of the neutron yield deviates from an exponential decay law with the recommended half-life 2.645 yr [2].

The isotopic composition in terms of californium nuclides is not given in the certificate of the ^{252}Cf source. Specifically, the content of the short-lived isotope ^{254}Cf with half-life to spontaneous fission 60.5 days is not indicated. For long decay times, the main neglected neutron source is ^{250}Cf with half-life 13.08 yr. According to measurements of the neutron yield performed over several years, the relative ^{250}Cf content in the source was about 11% initially, after which its contribution increased because of the long half-life. Moreover, the radionuclide ^{248}Cm accumulated as ^{252}Cf decayed, since the decay of ^{252}Cf occurs primarily via the emission of α-particles.

On the basis of these factors, a decision was made to eliminate the ^{252}Cf source and create new neutron reference fields IHEP OP-2017 based on ^{239}Pu–Be. Its advantage over ^{252}Cf is the ^{239}Pu half-life $\sim 2.41 \cdot 10^4$ yr, which ensures that the neutron yield remains constant during the entire operating time. The small contribution of other plutonium isotopes and the interval between calibrations can be taken into account by the procedure described in [3], where the neutron yield increases linearly in time.

In the present work, the OP-2017 reference fields are described, and a Bonner SB-RSU-01 spectrometer was used to measure the spectrum and integral characteristics of the neutron radiation [4]. The spectra were reconstructed by two methods using the BON95 code [4]. In the first method, the initial spectra for the iteration procedure were found by parameterization; the computed spectra were used in the second method.

Description of the IHEP OP-2017 neutron reference fields. In developing the new reference fields, the neutron radiation fields in [4] were used as a basis. The fields based on the ^{252}Cf source are excluded or ^{239}Pu–Be is used in the same geometry. Moreover, the standard distance is taken to be the distance 1 m between the center of the source and the measurement point, instead of the 75 cm used in [1, 4]. An exception is the scattered radiation field behind the absorbing cone, where

[1] Institute of High-Energy Physics, National Research Center Kurchatov Institute, Protvino, Russia; e-mail: sannikov@ihep.ru.
[2] Osnova Lab, Moscow, Russia.

Translated from Atomnaya Énergiya, Vol. 126, No. 5, pp. 275–280, May, 2019. Original article submitted November 23, 2018.

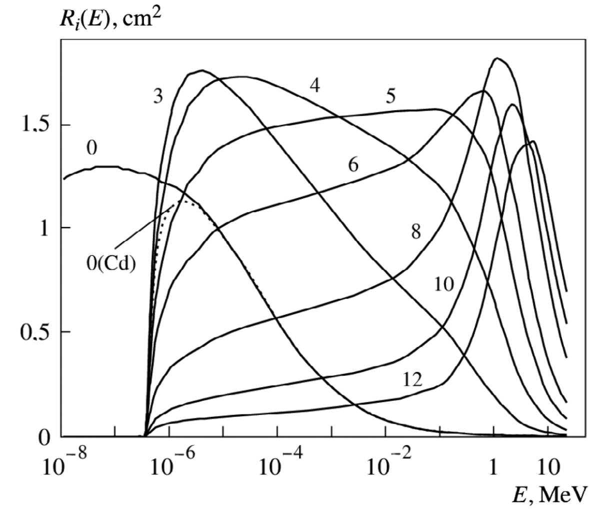

Fig. 1. Energy dependence of the sensitivity of the Bonner SB-RSU-01 spectrometer with irradiation perpendicular to the axis of the counter [4] (the numbers on the curves denote the diameter of the polyethylene sphere in inches).

the distance is equal to 1.5 m. The fields were realized in concrete enclosures with area 5.4 × 17.8 m^2 and height 4 m. The source and the measurement point were located at a height of 1.65 m above the floor.

Here is a brief description of the reference fields and the conventional notation:

1) ^{239}Pu–Be source without shielding (Pu–Be);

2) source in the UKPN-1M (UKPN) calibration-check setup [5];

3) source at the center of a 30.5 cm in diameter polyethylene sphere (CH_2);

4) source in the UKPN-1M setup with a thermal nozzle [6] (UKPN-T);

5) source behind an absorbing cone at a distance of 1.5 m (KONUS).

The radiation scattered from the floor, ceiling, and walls makes a small contribution in the UKPN and UKPN-T fields. On the other hand, the field behind the absorbing cone is almost completely due to the scattered radiation. The Pu–Be and CH_2 fields contain both components of the radiation.

The measurements were conducted with a ^{239}Pu–Be source IBN-25 No. 050 with neutron yield 10^7 nsec^{-1} according to the data in the certificate. The ambient equivalent dose rate at a distance of 1 m from the source in the UKPN-1M setup according to data from the latest check [7] is equal to 132.6 μSv/h to within 10% with confidence probability 0.95. It was used in the calibration of the SB-RSU-01 spectrometer.

Bonner SB-RSU-01 spectrometer and measurement procedure. The spectrometer was developed on the basis of a SBDN-01 detection block of the RSU-01 radiometer-dosimeter. The detector was placed at the center of polyethylene spheres with diameter 3, 4, 5, 6, 8, 10, and 12 in (1 in = 2.54 cm), covered with cadmium with thickness 0.1 cm. To eliminate in-leakage of thermal neutrons, a cadmium jacket was placed on the part of the detection block protruding from the spheres. A counter with and without cadmium cover was also used in measuring the flux density of thermal neutrons.

The SBFN-01 detection block contains a neutron scintillation detector in the form of a thin, 25 mm in diameter, disk placed on the photocathode of the photoelectron multiplier. The detector was fabricated from polymethylmethacrylate with added LiF powder, enriched with ^{6}Li, and ZnS(Ag). The neutrons are detected via the reaction $^6\mathrm{Li}(n, \alpha)^3\mathrm{H}$ followed by detection of the charged products from the reaction by means of a ZnS scintillator. A standard RSU-01 detector was used to develop the spectrometer without the use of an additional lightguide, which degrades the sensitivity of the device. The attendant cavity (photomultiplier and divider) inside Bonner spheres and the shape of the detector itself lead to sensitivity anisotropy, which depends on the diameter of the sphere and the energy of the neutrons.

These effects, which increase the measurement error, are minimized when the Bonner spheres are irradiated predominantly from the lateral surface side of the counter. In the standard measurement procedure [4], the axis of the counter is arranged approximately perpendicular to the direction toward the supposed maximum of the radiation intensity, which is usually well known. Anisotropy is manifested only in a small solid angle upon irradiation from the back. Estimates show that for the chosen measurement geometry the additional uncertainty owing to the sensitivity anisotropy does not exceed 5% in the worst case of an isotropic angular distribution of the radiation. Measurements in the fields 1–4 were performed with the counter arranged vertically in the polyethylene spheres. In the field 5 with scattered radiation predominating, the counter was arranged horizontally in the spheres.

In measuring the thermal neutron flux density by means of a counter without a cadmium cover, the axis of the counter was arranged in three mutually perpendicular directions and the minimum result was chosen. The indication of the cadmium-covered counter was also measured in this position in order to subtract out the contribution of the epicadmium neutrons.

The energy dependences of the sensitivity of the spectrometer $R_i^c(E)$ were calculated in [4]. Subsequently, they were refined by comparing the reconstructed spectra and the integral characteristics of the reference fields with the data in [1, 8]. The optimized sensitivity functions $R_i(E) = k_i R_i^c(E)$ with irradiation at an angle of 90° with respect to the axis of the counter, taking account of the correction factors k_i, are shown in Fig. 1. In the present work, the spectrometer was additionally calibrated using the UKPN field check data [7]: $R_i'(E) = k_{cal} R_i(E)$. The calibration coefficient was determined from the condition that the ambient equivalent neutron dose rate, determined according to the reconstructed spectrum in the UKPN field, must be equal to 132.6 μSv/h.

Reconstruction of the neutron spectra. The neutron spectra were reconstructed from the SB-RSU-01 measurements with the aid of the BON95 code, which is based on parameterization and iterations. The code implements the solution of the system of equations

$$M_i = \sum_{j=1}^{n} A_i(E_j)\varphi(E_j) + \Delta M_i, \quad i = 1, ..., m,$$

where M_i denotes the indications of the detector (counting rate) in different Bonner spheres; ΔM_i are uncertainties, including the measurement error ΔM_i^{exp} and the sensitivity functions $A_i(E_j) = R_i'(E_j)\Delta E_j$; $\varphi(E_j)$ is the desired neutron spectrum in the group representation.

The method of parameterization is used to search for the initial spectrum, which is refined by means of the iteration procedure of the directional divergence method [9]. In the present variant of the code, the low-energy spectra where parameterized by a linear superposition of a maxwellian peak of thermal neutrons $F_{th}(E)$, the $1/E$ spectrum of the intermediate neutrons $F_{int}(E)$, and a quasi-maxwellian peak of fast neutrons with the measured temperature and width $F_f(E)$:

$$E\varphi_p(E) = a_1 F_{th}(E) + a_2 F_{int}(E) + a_3 F_f(E), \tag{1}$$

where

$$\begin{cases} F_{th} = X_{th}^{3/2}\exp(-X_{th}), \quad X_{th} = E/T_{th}; \\ F_{int} = [1-\exp(-X_{th})]\exp(-X_f); \\ F_f = X_f^{3/2}\exp(-X_f), \quad X_f = (E/T_f)^c. \end{cases}$$

The temperature of the thermal neutron peak was chosen to be T_{th} = 0.035 eV, which corresponds to the spectrum behind the hydrogen-containing shielding. The exponential factors in the expression for F_{int} are included in order to suppress this component in the region of the thermal and fast neutrons. The free parameters c and T_f describing the width and temperature of the fast neutron peak are varied step-by-step in the prescribed grid in order to search for the optimal solution that minimizes χ^2 in the least-squares method:

$$\chi^2 = \frac{1}{m}\sum_{i=1}^{m}\left(\frac{M_i - M_i^p}{\Delta M_i^{exp}}\right)^2 = \min, \tag{2}$$

where $M_i^p = \sum_{j=1}^{n} A_i(E_j)\varphi_p(E_j)$.

The coefficients a_k in Eq. (1) are also found by the least-squares method for each combination of three parameters. Owing to linearization of the spectra Eq. (2) becomes a system of linear equations for the unknowns a_k

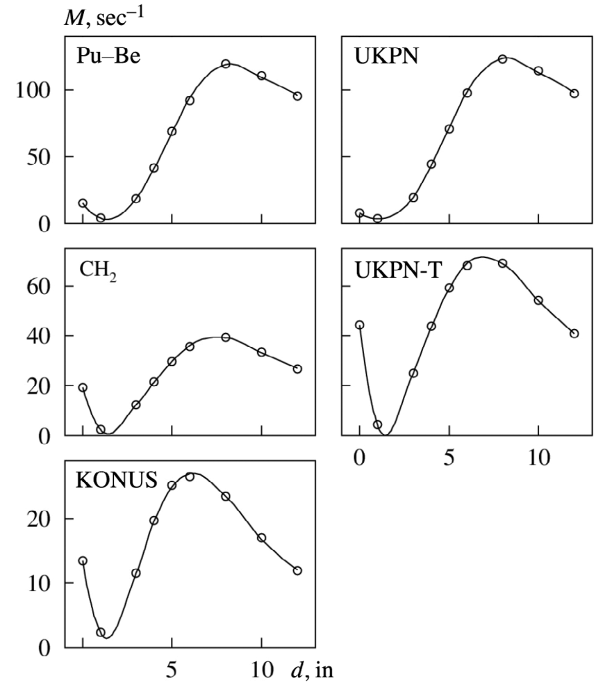

Fig. 2. Measured (○) and computed, according to the neutron spectrum reconstructed by the parameterization method, SB-RSU-01 counting rate (——) versus the diameter of the sphere in the OP-2017 reference fields.

$$\frac{\partial\chi^2}{\partial a_k} = \sum_{i=1}^{m}(B_{ik} - C_{ik}a_k) = 0,$$

which possesses a unique solution, since the components of the parameterised spectrum are linearly independent. The optimal spectrum $\varphi_p(E_j)$ corresponding to the minimum of χ^2 is then used as the initial spectrum in the iteration procedure with the termination criterion $\chi^2 < 1$. The obtained spectrum $\varphi_0(E_j)$ is used to calculate the total error

$$\Delta M_i = \sqrt{(\Delta M_i^{\exp})^2 + \left[\sum_{j=1}^{n}\Delta A_i(E_j)\varphi_0(E_j)\right]^2}. \quad (3)$$

The described procedure is performed for N different combinations of the spectrometer indications: $M_i' = M_i + \xi\Delta M_i$, where ξ is a random number with a normal distribution. As a result, we obtain N different neutron spectra (the standard $N = 25$), according to which the average values and the uncertainty of the spectrum and various functionals are determined.

Measurements and reconstruction of the neutron spectra by the parameterization method. The measurements as a function of the diameter of the spherical moderator in five reference fields are presented in Fig. 2. The data at diameter 1 in correspond to a cadmium-covered counter. In measurements with each sphere, the results were determined as an average over three exposures with duration 100 sec. The statistical errors (root-mean-square deviation) were determined using the relation $\Delta M_i^{\exp} = 1.7\sqrt{(M_i/t)}$, where t is the measurement time, sec, obtained from an analysis of the experimental data. They are small and in all cases fit within the size of the experimental points. The main contribution in the total error ΔM_i (3) is contributed by the error of the sensitivity functions $\delta A_i(E_j)$, equal to 10%, with the exception of the counter without a cadmium cover and a cadmium-covered counter, for which the errors were equal to 15%.

The neutron spectra of the OP-2017 reference fields, as reconstructed by the parametrization method, with errors are shown in Fig. 3. The number of energy groups in the reconstruction of the spectra was equal to five per decimal order of the neutron energy. The calibration coefficient obtained in the reconstruction of the spectrum in the UKPN field is equal to 0.89.

Application of the computed neutron spectra for reconstruction of the OP-2017 spectra. The OP-2017 neutron spectra are calculated using the FAN15 code [10]. The spectrum of an 80 g ^{239}Pu–Be source, measured with the aid of nuclear

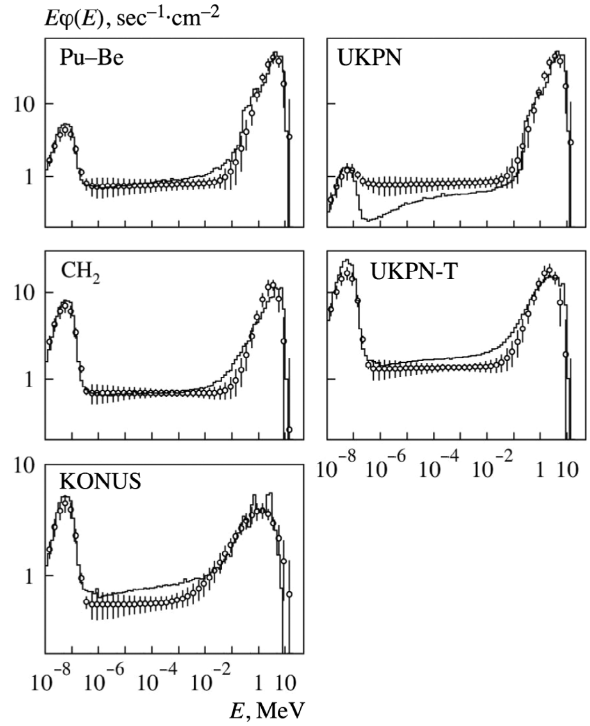

Fig. 3. OP-2-17 neutron Spectra (histograms) reconstructed by the parametrization method (○) and calculated using the FAN15 code.

photoemulsion (<1 MeV) and a stilbene spectrometer (>1 MeV) in [11], were used as the primary source of neutrons. The average energy of the spectrum is equal to 4 MeV and the maximum energy 11 MeV. The transport of neutrons in the UKPN-1M and UKPN-1M setups with a thermal nozzle and in a polyethylene sphere and absorbing cone as well as the moderation and scattering of neutrons in the concrete floor, ceiling, and walls were taken into account in the calculations. The geometry of the UKPN-1M and UKPN-1M with a thermal nozzle was set according to the data of [5, 6].

The computational results are displayed in Fig. 3 in comparison with the spectra reconstructed by the parameterization method using SB-RSU-01 data. On the whole, satisfactory agreement obtains between the computed and experimental spectra. The discrepancies can be explained by irregularities in the spectrum of ^{239}Pu–Be source, which are not described by parametrization (1), as well as inaccurate setting of the composition of the concrete and the designs of the UKPN-1M and UKPN-1M setups with a thermal nozzle.

The computed spectra were used as the initial approximation in the iteration procedure. The reconstructed neutron spectra are displayed in Fig. 4 in comparison with the spectra reconstructed by the parametrization method. With the exception of the UKPN field in the range 10^{-7}–10^{-6} MeV and the irregularities in the KONUS field, which are associated with the energy dependence of the neutron scattering cross-sections on the nuclei of the concrete, the spectra reconstructed by the two methods are in agreement with one another to within the limits of error.

Integral characteristics of the neutron spectra of the OP-2017 reference fields. The integral characteristics of the OP-2017 neutron reference fields, which were calculated according to the spectra reconstructed by two methods, are presented in Table 1. The individual equivalent dose rate $\dot{H}_P(10)$ is presented only in fields with a small contribution of scattered radiation, since this functional depends on the angular distribution of the radiation. With the exception of the average energy of the spectrum, all other quantities agree with one another for the different methods of reconstruction. The difference between them is significantly less than the errors, for which the root-mean-square deviation is presented.

The average energy of the neutron spectrum calculated by the parametrization method is systematically less than the value based on the computed spectra as the initial approximation. The KONUS field is an exception. On the whole, the results obtained with the aid of the computed spectra are more reliable. This is associated with the fact that unlike smooth

TABLE 1. Integral Characteristics of the OP-2017 Neutron Reference Fields with ^{239}Pu–Be Source IBN-25 No. 050 with Spectrometer Calibration According to $\dot{H}^{*}(10)$ in the UKPN Field According to the Check Data in [7]

Field	Initial spectrum	$E_{>0.4\ eV}$, MeV*	$\dot{H}^{*}(10)$, μSv/h	$\dot{H}_P(10)$, μSv/h	$\varphi_{>0.4\ eV}$, $sec^{-1}\cdot cm^{-2}$**	$\varphi_{<0.4\ eV}$, $sec^{-1}\cdot cm^{-2}$**
Pu–Be	Parameterization	3.39 ± 0.69	129.6 ± 11.1	–	98.6 ± 7.2	9.5 ± 1.6
	Calculation	3.47 ± 0.28	129.6 ± 8.9	–	100.2 ± 5.6	9.5 ± 1.3
UKPN	Parameterization	3.25 ± 0.68	132.6 ± 11.2	139 ± 12	101.4 ± 7.4	3.34 ± 0.62
	Calculation	3.36 ± 0.28	132.6 ± 9.1	139 ± 9.6	102.9 ± 5.8	3.45 ± 0.50
CH_2	Parameterization	2.19 ± 0.50	37.4 ± 2.8	–	34.4 ± 2.2	14.6 ± 2.4
	Calculation	2.46 ± 0.26	37.9 ± 2.7	–	35.2 ± 1.8	14.6 ± 2
UKPN-T	Parameterization	1.52 ± 0.39	60.5 ± 3.8	63 ± 4.1	60.1 ± 3.7	34.6 ± 5.7
	Calculation	1.75 ± 0.22	60.7 ± 4.4	63.3 ± 4.6	61.3 ± 2.9	34.5 ± 4.8
KONUS	Parameterization	1.34 ± 0.37	18.5 ± 1.1	–	23.1 ± 1.3	9.5 ± 1.6
	Calculation	0.99 ± 0.14	18.0 ± 1.4	–	22.6 ± 1.0	9.5 ± 1.3

* The average energy of the spectrum of epicadmium neutrons. ** The flux density of epicadmium and thermal neutrons, respectively.

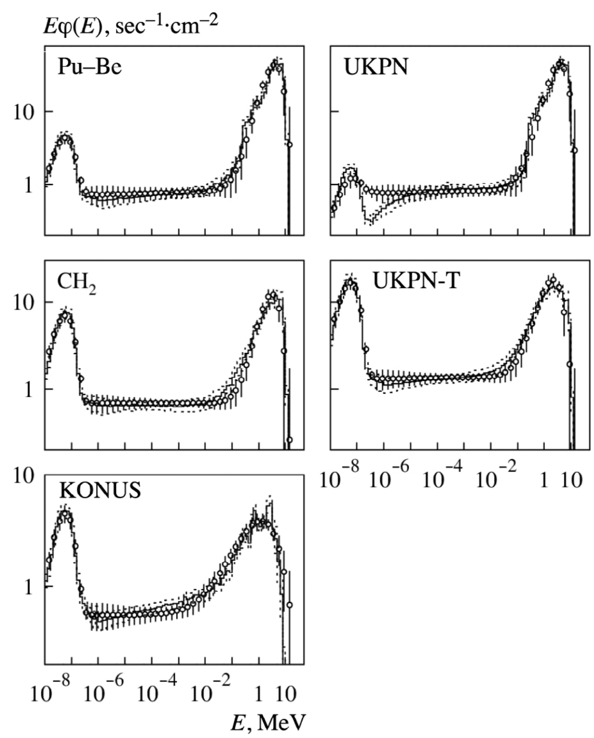

Fig. 4. Neutron spectra reconstructed using as the initial approximation in an iteration procedure the parameterized (○) and computed spectra (histograms with error corridor).

parameterized spectra with maximum energy 20 MeV the computed spectra are based on the measured ^{239}Pu–Be spectra with irregular structure and maximum energy 11 MeV.

Conclusion. The neutron spectra and integral characteristics of the OP-2017 reference fields based on ^{239}Pu–Be source IBN-25 No. 050 were measured. The range of the average energy of epicadmium neutrons of the measured spectra is equal to 1–3.5 MeV, and the contribution of the thermal neutrons in the total flux density is 3.2–36%. In addition, the angular distribution of the neutrons differs – from directed in the UKPN and UKPN-T fields to quasi-isotropic scattered radiation in the KONUS field. Comparing the two methods of reconstruction of the neutron spectra, in spite of the closeness of the results, attests in favor of using the computed spectra as the initial approximation in the iteration procedure used to reconstruct the spectra. The new reference fields will find application in establishing the energy dependence of the sensitivity of dosimetric devices used in the IHEP accelerator complex.

This work was performed by the support of the Ministry of Education and Science of the Russian Federation as part of Subsidy Agreement No. 14.607.21.0193, September 26, 2017; Agreement ID RFMEFI60717X0193.

REFERENCES

1. G. I. Britvich, V. S. Volkov, Yu. I. Kolevatov, et al., *Spectra and Integral Characteristics of Neutron Reference Fields Based on Radionuclide Neutron Sources*, IHEP Preprint 90–48 (1990).
2. G. Audi, O. Bersillon, J. Blachot, and A. H. Wapstra, "The NUBASE evaluation of nuclear and decay properties," *Nucl. Phys. A*, **729**, 3–128 (2003).
3. I. A. Kharitonov, "Correction of the flux of plutonium-beryllium neutron sources in the verification interval," *At. Energ.*, **51**, No. 3, 124–125 (1981).
4. A. V. Sannikov, V. N. Peleshko, E. N. Savitskaya, et al., "A multi-sphere neutron spectrometer based on the serial device RSU-01," *ANRI*, No. 1 (56), 62–69 (2009).
5. S. N. Balakhnichev, M. F. Yudin, and A. P. Yanovskii, "Development and research of the UKPN-1 setup for verification and calibration of neutron devices in a collimated beam," in: *Proc. Metrological Institutes of the USSR*, No. 124 (184), Energiya, Leningrad (1970), pp. 107–129.
6. S. N. Balakhnichev, S. I. Slepyshkov, M. F. Yudin, and A. P. Yanovskii, "Investigation of the thermal neutron flux of the UKPN-1 facility," *ibid.*, pp. 137–146.
7. Check Certificate No. 4.410-3002-16, "Neutron radiation testing setup UKPN-1M," VNIIFTRI, Moscow, Dec. 23, 2016.
8. G. I. Krupnyi, Y. N. Rastsvetalov, E. N. Savitskaya, and A. V. Sannikov, "A multi-sphere neutron spectrometer with an activation thermal neutron detector based on the reaction $^{115}In(n, \gamma)^{116m}In$," in: *Abstr. 9th Russ. Sci. Conf. on Radiation Protection and Radiation Safety in Nuclear Technologies* (2006), pp. 121–123.
9. M. Z. Tarasco, *On a Method for Solving Linear Problems with Stochastic Matrices*, Preprint FEI-156 (1969).
10. E. N. Savitskaya and A. V. Sannikov, "FAN15 code for calculating the transfer of low-energy photons and neutrons in arbitrary media," *At. Energ.*, **122**, 40–45 (2017).
11. M. Anderson and R. Neff, "Neutron energy spectra of different size ^{239}Pu–Be(α, n) sources," *Nucl. Instrum. Meth.*, **99**, 231–235 (1972).