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ABSTRACT

The work presents the description and comparison with calculations of the integral 14-MeV neutron experiments for leakage neutron spectra, total neutron leakages from U, Th, Be, Pb and Bi spherical samples, and for reaction rates in U and Th samples, carried out in Kurchatov Institute of Atomic Energy (KIAE), RF.

A detailed description of the used measurement methods, experimental samples of multiplying materials and experimental results are given. The calculations were done by the BLANK code with the neutron data prepared from ENDL-75 and ENDF/B-IV libraries and by MCNP/4A code with FENDL-1 data.

It was shown that the calculation with ENDF/B-IV data describes the shape of the neutron spectrum better than ENDL-75 for all considered materials. The total neutron multiplication for the U sample is reproduced more accurately by the calculation with ENDL-75 data, and for Th, and Pb assemblies, by the calculation with ENDF/B-IV data.

The calculation with ENDF/B-IV data well reproduces the neutron leakage from lead samples while the calculation with ENDL-75 data slightly underestimates the neutron multiplication on lead. The calculation with ENDF/B-IV data describes the neutron multiplication for beryllium sufficiently well. The calculated total neutron leakage from Be samples with ENDL-75 data noticeably exceeds the experimental one. The calculations with FENDL-1 data perfectly (within experimental error) predict total multiplication in lead and beryllium spheres. For both bismuth spheres there is underprediction of total multiplication in calculations with FENDL-1 ~8%.

The calculations with FENDL-1 data in a whole predict neuron spectra for beryllium better than with ENDF/B-IV data but still there is overestimation of neutron spectra in the energy range 0.7-3 MeV.

1. Introduction

The problem of improving the constants for fusion neutronics analyses still has high priority. A new generation of neutron data for fusion neutronics calculations is characterised by a substantially higher level of requirements to the accuracy of cross-sections (1-3% for total and 10-20% for double differential ones). This is especially true in regard to the breeder and coolant materials in the blanket as well as for shielding materials. The natural criterion of the data quality is an integral experiment which enables to obtain information on the advantages and disadvantages of a file and to determine the level of confidence in the calculation results.

In this work a review of integral experiments and corresponding analyses on testing the neutron data for neutron multiplying materials, such as U, Th, Be, Pb and Bi, performed in the I.V. Kurchatov Institute of Atomic Energy[1] , is given.

2. General aspects of total neutron leakage measurement

The number of neutrons leaking from the sample, normalised to 14-MeV source neutron, may be approximately presented by the expression:

$$M = T + (1 - T)\,\mu, \qquad (1)$$

where $T = e^{-\Sigma_{in} l}$ is the transmission of 14-MeV neutrons by a sample of thickness l; Σ_{in} is a macroscopic cross-section of the inelastic interaction; μ is a neutron yield in a single inelastic interation event.

In expression (1) we made the assumptions that:

- elastic scattering does not change the direction of movement and the energy of the neutrons emitting by the source,
- secondary neutrons from inelastic interaction do not produce multiplication,
- neutrons are not absorbed in the sample.

The first assumption is justified by well known anisotropy of 14-MeV neutrons scattering by heavy nuclei mainly in a forward direction. Neglecting of secondary neutron multiplication is valid for assemblies made of non-fissile elements (lead, bismuth and a number of others) of any thickness, since the threshold of (n,2n)-reaction for these materials is usually higher than the maximum energy of the secondary neutron spectrum of (n,2n)-reaction. In the case of fissile materials (uranium, thorium), it is necessary to supplement the expression (1) with additional factor, but for small thickness this factor is close to unity.

In principle, the measurement of the value of M is similar to measurement of radionuclide neutron sources intensity, and for that reason the same experimental methods can be applied.

As early as in the 1950s, when (n,2n)-cross-section determining by measurement of 14-MeV neutron multiplication in spherical samples was introduced, the use was made of all-wave neutron counters (see, e.g., Ref. [1]). The so-called "long counter" of Hanson and

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[1] The neutron leakage spectra measurements were done jointly with IPPE (Obninsk).

McKibben [2] whose efficiency depended only slightly on neutron energy was widely used. In later studies (e.g., Ref. [3]) it was shown that the neutron response of the long counter is not constant in the 0.002-1 MeV and 5-14 MeV energy ranges. This, in turn, for accurate calibration of the counter requires the knowledge of the spectrum of emitted neutrons, which inevitably affects the accuracy of neutron yield measurement.

With the development of various methods of neutron spectrometry and, in particular, of the time-of-flight method, the significance of "long counters" diminished considerably and they were displaced almost entirely from precise neutron-physics experiments. Down to the present, however, one of the varieties of all-wave counters - total absorption detectors (manganese bath, boron tank, water tank, etc.) - are used for the calibration of sources. The principle of the method is that the source under study is placed in an absorbing medium of sufficiently large size, in which neutrons are slowed down and then captured. The source intensity is determined by integration of the absorption rate over the moderator volume. Since the neutrons are captured in the thermal energy region, knowledge of the source neutron spectrum is not necessary, and integration over a 4π solid angle makes unnecessary taking into account their angular distribution.

Effect of external moderator on neutron multiplication in a sample

An example of the use of the total absorption method in studies on the multiplication of 14-MeV neutrons in fusion reactor blanket may be found in the work of Basu et al. [4], dealing with the measurement of neutron leakage from beryllium assemblies. A 14-MeV neutron source was placed at the centre of a rectangular beryllium sample which, in turn, was positioned at the centre of a large beryllium cube. The moderation of the fast neutrons and the capture of the thermal neutrons took place by the hydrogen nuclei. The thermal neutron flux was measured by boron counters. Along with the main experimental results, the authors mention a negligibly small effect of the moderator on the yield of neutrons from the assemblies. However, no quantitative evaluations in support of this fact were presented.

Obviously, the main problem arising in the attempt to use the total absorption method for measuring the neutrons multiplication in a sample is an effect of the external layer of the moderator on the yield of neutrons from the multiplying sample. As a result of scattering in the external moderator, the neutrons leaking from the sample are able to return to the sample and be absorbed in it or to give further multiplication. The flow of neutrons into the sample depends on the geometry of the experiment as well as on the type of moderator and multiplier.

A schematic representation of the measurement system is shown in Fig.1, where the spherical test sample with a central point-source of 14-MeV neutrons is placed inside a moderator of fairly large size. The experiment may be optimised by varying the dimensions of the inner cavity in the moderator. By increasing its radius one may reduce the angle of visibility of the sample from the internal surface of the moderator and thus reduce the probability for scattered neutrons to come back to the sample. However, the possibility of such regulation is limited by increasing of the moderator volume and, as a result, complicating experimental arrangement. In the choice of a moderator, preference

may be given to a water or some hydrogenous substances, such as paraffin or polyethylene. A water, having high moderating capacity, causes rapid drop in the energy of the neutrons, bringing them below the threshold of the multiplication reactions on the material of the sample. The use of an aqueous moderator makes the assembly compact which is important from the point of view of experimental technique.

The most widespread realisations of the total absorption methods involve the use of the magnesium bath, the boron tank or the water tank. Since in all the mentioned techniques the neutrons are moderated in scattering on hydrogen, the difference in thermal neutron current into the sample will be determined only by absorption of the neutrons in the moderator. The maximal macroscopic absorption cross-section corresponds to an aqueous solution of boric acid, that determined our choice of this type of the moderator.

The neutronics characteristics of the typical model of the sample placed in a boron tank were studied in special neutronics analysis [7]. The arrangement of the TAD is similar to that shown in Fig.1. The diameter of the internal cavity is 40 cm. A cadmium filter 1 mm thick is located at the inner surface of the cavity. The thickness of the moderating layer is 45 cm. The nuclear density of the isotope ^{10}B in a water, $7.4 \cdot 10^{19}$ cm^{-3}, corresponds to the maximum solubility of boric acid of natural isotopic composition in water at a temperature of 20^0C. The calculations were performed with the Monte Carlo code BLANK [6]. Sixty-five group constants were prepared on a basis of ENDL library in the 0.1- 15 MeV range and 21-group constants for calculations in a P_1-approximation below 0.1 MeV. Tables 1-3 present the neutron leakage data and the main reaction rates in the tested samples, surrounded by water and without it.

Table 1. The rates of main processes in beryllium assemblies with and without water

Reaction rate	Layer thickness of the sample, cm					
	1.5*		3		8	
	without water	with water	without water	with water	without water	with water
(n,2n)	0.1073	0.1071	0.2150	0.2149	0.6017	0.6072
(n,α)	0.0037	0.0037	0.0675	0.0676	0.0147	0.0148
Leakage	1.121	1.120	1.223	1.224	1.524	1.553

*) The radius of internal cavity in beryllium assemblies is 4.5 cm.

Table 2. The rates of main processes in lead assemblies with and without water

Reaction rate	Layer thickness of the sample, cm			
	3(1)		6(2)	
	without water	with water	without water	with water
(n,2n)	0.1761	0.1759	0.3271	0.3302
Leakage	1.193	1.203	1.347	1.357

*) The dimensions of the assemblies were as follows:

(1) δ = 3 cm; r_{int} = 9 cm; R_{ext} = 12 cm;

(2) δ = 6 cm; r_{int} = 3 cm; R_{ext} = 12 cm.

Table 3. The rates of main reactions in uranium samples with and without water (isotopic composition of uranium: ^{238}U - 99.6 %, ^{235}U - 0.4 %)

Reaction rate	Layer thickness of the sample, cm					
	1(1)		4(2)		8(3)	
	without water	with water	without water	with water	without water	with water
^{238}U(n,f)	0.068	0.066	0.2853	0.2892	0.523	0.524
^{235}U(n,f)	0.715-3	1.106-3	0.492-2	0.727-3	0.016	0.020
Fission source	0.283	0.289	1.137	1.157	2.002	2.016
^{235}U(n,2n)+ ^{238}U(n,2n)	0.052	0.055	0.1372	0.1395	0.229	0.226
^{235}U(n,3n)+ ^{238}U(n,3n)	0.023	0.023	0.369	0.414	0.804	0.871
Absorption	0.072	0.083	0.369	0.414	0.804	0.871
Leakage	1.308	1.325	2.064	2.059	2.681	2.645

*) The dimensions of samples were as follows:

(1) δ = 1 cm; r_{int} = 10 cm; R_{ext} = 11 cm;

(2) δ = 4 cm; r_{int} = 8 cm; R_{ext} = 12 cm;

(3) δ = 8 cm; r_{int} = 4 cm; R_{ext} = 12 cm;

The calculational error of functionals is estimated as 2-5 %. The effect of water exceeds the error limits only in the case of U-235 fission rate and of absorption rate in uranium samples, i.e. the parameters sensitive to thermal neutrons from the moderator. However, the variation in total neutron leakage does not exceed 2 %. Thus, to within the error limit of the calculation, the influence of water on multiplication in samples of beryllium, lead, uranium could be neglected. Calculations for a total absorption detector without cadmium filter has shown that in the uranium spheres the U-235 fission rate increased by a factor of 4 and highly influences the neutrons leakage from the samples.

The boron tank method

Let us consider in more details the measurement of neutron leakage by the boron tank method. The neutrons emitted by the sample are fully absorbed in physically infinite aqueous solution of some boron compound, e.g. boric acid. If N_B, N_H and N_O are the nuclear densities of boron, hydrogen and oxygen, then the number of neutrons with the starting energy E emitted by the sample will be equal to the number of neutrons absorbed in the tank:

$$Q(E) = \int_0^\infty \int_{E'} N_B \sigma_B(E') \cdot (1 + \frac{N_H \sigma_H(E')}{N_B \sigma_B(E')} + \frac{N_O \sigma_O(E')}{N_B \sigma_B(E')}) \cdot 4\pi^2 \Phi(r, E, E') dr dE'. \quad (2)$$

If at point r of the moderator there is a boron counter with a ^{10}B nuclear density, N_Z, and a volume, V_Z, then, neglecting the variation in flux over the volume of the counter, we can write the following expression for the count rate:

$$Z(r, E) = N_z V_z \int_{E'} \sigma_B(E') \Phi(r, E, E') dE . \quad (3)$$

It is assumed here that the efficiency of the counter is unity. Using the notation for $Z(r,E)$, we represent $Q(E)$ in the form:

$$Q(E) = \frac{4\pi N_B}{N_Z V_Z} \int_0^\infty \int_{E'} (1 + \frac{N_H \sigma_H(E')}{N_B \sigma_B(E')} + \frac{N_O \sigma_O(E')}{N_E \sigma_E(E')}) \cdot r^2 Z(r, E') dr dE'. \quad (4)$$

Varying the value of N_{H2O}/N_B, i.e. changing the concentration of boric acid in the aqueous solution, one can reduce the capture of thermal neutrons by hydrogen (the absorption of fast neutrons by hydrogen is small). For high concentrations of boron, when $N_H \sigma_H \ll N_B \sigma_B$, expression (4) takes the form:

$$Q(E) = \frac{4\pi N_B}{N_Z V_Z} \int_0^\infty r^2 Z(r, E) dr. \quad (5)$$

The total source intensity will be :

$$Q_0 = \int Q(E)dE. \tag{6}$$

Neutron response of the TAD

We designate the neutron response in the TAD as the value:

$$\varepsilon(E) = \frac{\frac{4\pi N_B}{N_Z V_Z}\int_{R_1}^{R_2} r^2 Z(r,E)dr}{Q(E)}, \tag{7}$$

where R_1 and R_2 are the internal and external radii of the moderator. The efficiency $\varepsilon(E)$ is always less than unity, for the following reasons:

(1) Some of the fast neutrons can be captured by oxygen in the $^{16}O(n,p)^{16}N$ and the $^{16}O(n,\alpha)^{13}C$ reactions with the thresholds at 10 MeV and 3 MeV, respectively;

(2) With increasing energy there is an increasing probability of neutron leakage from the volume of a TAD of finite dimensions;

(3) Some of the thermal neutrons are captured by hydrogen and by structural components of the TAD (including the cadmium filter).

For the arrangement shown in Fig.1, the energy dependence of the TAD neutron response was calculated. The dimensions of the total absorption detector were the same as those in the calculation referred to above. At the centre of the TAD was located a point-wise, isotopic and monoenergetic neutron source. The neutron energy varied in the range of 0.01-14 MeV. The dependence of $\varepsilon(E)$, calculated with the BLANK code, is shown in Fig.2. The figure shows that the recording efficiency is about 88% right up to 5-6 MeV (approximately 6% of the neutrons are captured by hydrogen, 4% by cadmium, and the remainder leak out of the TAD). Above 6 MeV, neutron leakage from the TAD and the capture of fast neutrons by the oxygen of the water starts to make a definite effect.

Characteristics of neutron leakage measurement by the boron tank method

Let us consider the energy distribution of the neutrons leaking from the samples with a central 14-MeV neutron source. On Fig.3 the neutron leakage spectrum from uranium sphere with a layer thickness of 8 cm calculated with the BLANK code is shown. It may be approximated in the form of two components: 14-MeV source neutrons and secondary neutrons from inelastic interactions, including (n',n), (n,2n), (n,3n) and (n,f) reactions. Because of the insignificant energy loss, the neutrons from elastic scattering can be united with the source neutrons. The relationship between the scattered and unscattered parts of the spectrum will be determined by the thickness of the multiplier. The calculated neutrons leakage spectra from lead and uranium samples of various thickness show that for all sizes of considered samples made of heavy elements, the representation of the

leakage spectrum in the from of the sum of two components is fully justified; the boundary of the non-elastically scattered neutron spectrum can be assumed to be 6 MeV.

The considered leakage spectrum characteristics allow to assign each component its own detector response.

Let us write the response of the TAD to the value of m in the following way:

$$m = \varepsilon_{14} \cdot T + \langle\varepsilon\rangle (1-T) \cdot \mu \tag{8}$$

where ε_{14} and $\langle\varepsilon\rangle$ are the responses for 14-MeV neutrons and neutrons from inelastic interactions. If the multiplication of secondary neutrons in the sample is not neglected, the value m can be written in the form:

$$m = \varepsilon_{14} \cdot T + \langle\varepsilon\rangle N_{sec} \tag{9}$$

where N_{sec} is a number of secondary neutrons leaking from the sample, normalized on one 14-MeV source neutron.

The efficiency ε_{14} can be determined in a simultaneous measurement in the TAD of the integral

$$\int_{R_1}^{R_2} 4\pi r^2 Z(r,E)dr$$

and in measurements of the intensity of the 14-MeV neutron source by the method of associated α-particles. To determine $\langle\varepsilon\rangle$ it is sufficient to use any radionuclide neutron source of known intensity with the spectrum within the 0-6 MeV energy range. For instance, as shown in the calculation, the neutron counting efficiency for a ^{252}Cf source amounts to approximately 87%, which coincides with the value of ε in the mentioned range. Photoneutron sources, neutrons from the (d,d) reaction with 2.5 MeV energy and a number of other sources are suitable for purposes of calibration.

The experimentally measured value of m, by itself, does not yet allow determining M, the neutron leakage from the sample, since in the general case $\varepsilon_{14} \neq \langle\varepsilon\rangle$. For this reason it is necessary to determine individually the terms in the expression for M: transmission, T, and the number of secondary neutrons, $1-T$ or N_{sec}. The transmission, T, can be found from a supplementary measurement, for example, with threshold detectors. If the value of T is known, we can determine N_{sec}:

$$N_{sec} = \frac{m - \varepsilon_{14} T}{\langle\varepsilon\rangle}. \tag{10}$$

After this, we find M, which is equal to:

$$M = T + N_{sec} \ . \tag{11}$$

The predicted error in measurement of M is 5-6%, which includes:

- the error in measurement of ε_{14} (the error of absolute neutron yield ≈ 3%),
- the error in measurement of $<\varepsilon>$ (the californium source intensity is known to 2-3%),
- the error in measurement of T (in relative measurement an error of 1-2% can be achieved),
- the error in measurement of m with normalization to one 14-MeV neutron amounts to 3%.

In conclusion, emphasis should be placed on the advantages of measuring M by the total absorption method. First, in contrast to the spectrometric methods, M is measured in the entire energy range of the neutrons emitted and the spectrum is not to be extrapolated below the neutron response threshold of the spectrometer. Second, owing to the high neutron response of the TAD, the measurements can be performed with low intensity (10^8-10^9 n/s) sources, there being no special requirements whatever with respect to neutron generators (e.g. there is no need for a pulsed operating regime of a neutron generator with short pulses, which is a prerequisite for time-of-flight spectrometers). Lastly, the measurement technique in the total absorption method is fairly simple and can be carried out easily.

Experimental facility and samples

An experimental facility consists of a modernised neutron generator NG-150m (E=150 KeV, I =3 mA) and the TAD. A titanium-tritium target 28.5 mm in diameter with the activity of about 5-10 Bq was placed at the center of a spherical sample. The deuteron beam diameter at the target surface does not exceed 10 mm, the current is about 1 mA. A surface-barrier counter for recording of associated α-particles due to (d,t) reaction is mounted at an angle of 178° to the deuteron beam, at a distance of 830 mm from the target.

The total absorption detector of neutrons is a spherical layer filled with a boric acid solution in a distilled water. Boron in boric acid is enriched to 88.6 % in the ^{10}B isotope. The ^{10}B nuclear density is $7.95 \cdot 10$ cm^{-3}. The TAD outer spherical shell 1320 mm in diameter is made of 2 mm thick brass sheet. A 2 mm thick inner shell is made of stainless steel and is 600 mm in diameter. It is mounted on a cylindrical support rated for a 750 kg load.

In a vertical cross section through the TAD center there are radial channels housing a boron chamber. The channels are placed at angles of 0°, 40°, 80°, 120°, and 140° to the deuteron beam. The boron neutron absorption rate is measured by the KNT-10 chamber having the external diameter 7 mm and the total length 70 mm. The length of an active boron layer in a chamber is 5 mm.

A lead spherical sample 500 mm in diameter has an internal spherical cavity 50 mm in diameter. The sample has an axial channel 43 mm in diameter to house a target in the center. The characteristics of the samples are given in Tables 4, 5.

Table 4. Chemical contents of Pb and Bi samples

Pb (Contents of the basic material is not less than 99.4%)

Element	Al	Cu	Ag	Zn	Fe	Sb	Mn
Weight fraction, %	0.01	0.02	0.09	0.1	0.01	0.3	0.03

Bi (Contents of the basic material is not less than 96.4%)

Element	Cu	Ag	Fe	Mn	Pb
Weight fraction, %	0.05	0.1	0.01	0.24	3.5

Isotopic contents of the TUD lead sample

Isotope	204	206	207	208
Contents, %	1.36±0.05	24.9±0.1	21.4±0.1	52.3±0.1

Table 5. Parameters of the spherical shells

Element	Nuclear density, $10^{22}cm^{-3}$	Shell thickness, cm	Inner radius, cm	Outer radius, cm	Channel diameter, cm
Pb	3.30	3 9 22.5	9 3 2.5	12 12 25	5.0 5.0 5.0
Bi	2.82	3 9	9 3	12 12	5.0 5.0
Th	2.93	3 7 10	3 6 3	6 13 13	5.0 5.0 5.0
U	^{238}U - 4.460 ^{235}U - 0.019	1 2 8	10 10 4	11 12 12	5.0 5.0 5.0

Measurement of leakage neutron total flux

The procedure of measuring $<\varepsilon>$, ε_{14} and M are identical and consist in the following. The boron chamber was installed in one of the TAD radial channels. The KNT-10 counting rate normalized to one source neutron was measured at a fixed point. The chamber travel along the channel length allowed the spatial distribution of a neutron absorption rate to be measured. The chamber travel step varied from 10 to 50 mm depending on the point of measurement. The obtained radial distributions of the KNT-10 chamber counting rate were numerically integrated over the TAD volume.

In measurements of the efficiency $<\varepsilon>$ the ^{252}Cf source was used with the intensity of 5-10 n/s. The error in determining the neutron yield is 1.5 % at a confidence level 0.68. The average intensity of a 14 MeV neutron source when measuring ε_{14} and M was 3-10 n/s (the measurement error was about 2 %).

2. Measurements of total neutron leakage from spherical shells made of U, Th, Be, Pb and Bi by the total absorption method

The results of the measurement of total neutron leakage from U, Th, Be, Pb, and Bi spherical shells of various thickness are presented in [7,8]. The experimental values here and later on were compared with corresponding calculations by the BLANK code with the data from ENDL-75 and ENDF/B-IV libraries and by MCNP/4A with FENDL-1 data. Table 6 lists the experimental and calculated values for a total neutron leakage from the samples. Experimental errors are given with a confidence level of 68%. The statistical error of calculations for functionals did not exceed 1%, for the groups fluxes, 4%.

It is seen from Table 6 that the calculation with ENDL-75 data satisfactorily describes neutron leakage from uranium samples; and with ENDF/B-IV data the calculated leakage are slightly less than the experimental one. For the thickest sample the difference amounts to 6%, at the experimental error at 3%. The measured neutron leakage from thorium samples are higher than the calculated ones with ENDL-75 data as well as with ENDF/B-IV data. The difference for the sample with 10-cm shell thickness at the experimental error of 3% comes up to 11% with ENDL-75 data, and to 9% with ENDF/B-IV data. The calculation with ENDF/B-IV data well reproduces the neutron leakage from lead samples. The calculation with ENDL-75 data slightly underestimates the neutron multiplication on lead. As a result, a 5.5% excess in the experimental neutron leakage over the one calculated with ENDL-75 data is observed for the sample with 9-cm shell thickness. The calculation with ENDF/B-IV data describes the neutron multiplication for beryllium sufficiently well. The calculated (with ENDL-75 data) total neutron leakage from Be samples noticeably exceeds the experimental one. The difference for Be assemblies with a shell thickness of 8 cm with an experimental error of 3% amounts to 14%. The calculations with FENDL-1 data perfectly (within experimental error) predict total multiplication in lead and beryllium spheres. For both bismuth spheres underprediction of multiplication in calculations with FENDL-1 makes 8%.

Table 6 Experimental and calculated neutron leakage from the samples

Material	thickness (cm)	Experiment	BLANK calculation				MCNP/4A	
			ENDL-75	C/E	ENDF/B-IV	C/E	FENDL-1	C/E
U	1 (r=10...11)	1.33±0.04	1.31	0.985	1.29	0.970		
	2 (r=10...12)	1.57±0.05	1.60	1.019	1.54	0.981		
	8 (r= 4...12)	2.67±0.08	2.68	1.004	2.52	0.943		
Th	7 (r= 6...13)	1.63±0.05	1.52	0.932	1.52	0.932		
	10 (r= 3...13)	1.82±0.06	1.64	0.901	1.67	0.917		
Be	5 (r= 6...11)	1.36±0.04	1.49	1.096	1.39	1.022	1.33	0.978
	8 (r= 3...11)	1.53±0.05	1.76	1.150	1.60	1.046	1.51	0.986
Pb	3 (r= 9...12)	1.26±0.04	1.19	0.944	1.20	0.952	1.22	0.968
	9 (r= 3...12)	1.53±0.05	1.45	0.948	1.48	0.967	1.54	1.006
	22.5 (r=2.5...25)	1.86±0.08			1.796	0.966	1.81	0.973
Bi	3 (r= 9...12)	1.26±0.04					1.166	0.926
	9 (r= 3...12)	1.56±0.05					1.431	0.917

3. TOF measurements of the neutron leakage spectra from U, Th, Pb and Be spherical samples

Measurements of the neutron leakage spectra from U, Th, Pb and Be spherical samples with layer thickness of 8, 7, 9 and 5 cm, respectively, were carried out by the time-of- flight technique with the spectrometer based on the KG-0,3 cascade generator [9]. The generator is operated under pulsed conditions with a pulse duration of 2.5 ns, frequency 1.25 MHz and accelerating voltage 250 kV. The neutron generator target through a special channel of 50 mm was placed in the center of spherical samples. The neutron leakage spectra were measured with the 3.5 m flight base in the energy range of 0.35-15 MeV by a detector made of a stilbene crystal of 63 mm in diameter and 50 mm in height. A detailed description of the experiment is given in [10].

In Tables 7-10 the experimental and calculated integral neutron leakage spectra in various energy ranges, corresponding to a total neutron leakage in 4π are given.

From a comparison of the calculated and experimental results for the uranium sample, it is seen that the calculation with ENDF/B-IV data is significantly closer to the experiment than that with ENDL-75 data. However, within the 6-10 MeV range the reproducing of spectrum by a calculation both with ENDL-75 and ENDF/B-IV data is unsatisfactory. The region 10-15 MeV is described well by calculations with both libraries.

The neutron leakage spectrum from the Th-sample within the 2-4 MeV and 10-15 MeV energy ranges is described well by the calculation with both ENDL-75 and ENDF/B-IV data. In the 0.6-2 and 4-10 MeV ranges the calculation with both libraries significantly underestimates the neutron spectrum. The calculation with ENDF/B-IV data for the 0.6-2 MeV range is closer to the experiment; but for the 4-10 MeV range, that with ENDL-75 data is closer.

Table 7. Integral neutron leakage spectrum from uranium sample

Energy interval, MeV	Experiment	BLANK calculation			
		ENDL-75	C/E	END/B-IV	C/E
0.35- 0.8	0.79 ±0.04	0.67	0.85	0.74	0.94
0.8 - 6	0.63 ±0.03	0.89	1.41	0.74	1.10
6 - 10	0.047±0.003	0.033	0.70	0.024	0.51
10 - 15	0.30 ±0.02	0.31	1.04	0.32	1.07
0.35- 10	1.47 ±0.07	1.59	1.08	1.45	0.99

Table 8. Integral leakage neutron spectrum from thorium sample.

Energy interval, MeV	Experiment	BLANK calculation			
		ENDL-75	C/E	END/B-IV	C/E
0.35- 0.8	0.79 ± 0.04	0.67	0.85	0.74	0.94
0.6 - 2	0.53 ± 0.03	0.34	0.64	0.36	0.68
2 - 4	0.093± 0.005	0.096	1.03	0.097	1.04
4 - 10	0.13 ± 0.008	0.043	0.33	0.03	0.23
10 - 15	0.51 ± 0.03	0.52	1.02	0.52	1.02
0.6 - 15	1.20 ± 0.06	1.00	0.83	.005	0.84

Table 9. Integral neutron leakage spectrum from lead sample

Energy interval, MeV	Experiment	BLANK				MCNP/4A	
		ENDL-75	C/E	END/B-IV	C/E	FENDL-1	C/E
0.35- 1.2	0.41 ±0.02	0.43	1.04	0.44	1.06	0.421	1.027
1.2 - 3	0.37 ±0.02	0.33	0.89	0.33	0.90	0.518	1.402
3 - 5	0.066±0.003	0.065	0.98	0.059	0.89	0.0776	1.175
5 - 10	0.060±0.004	0.028	0.46	0.030	0.50	0.0361	0.602
10 - 15	0.44 ±0.02	0.47	1.07	0.46	1.04	0.377	0.856
0.35- 10	0.91 ±0.05	0.85	0.94	0.86	0.95	1.053	1.157

Table 10. Integral neutron leakage spectrum from beryllium sample

Energy interval, MeV	Experiment	BLANK				MCNP/4A	
		ENDL-75	C/E	END/B-IV	C/E	FENDL-1	C/E
0.35- 0.7	0.096±0.005	0.054	0.56	0.075	0.78	0.0.089	0.931
0.7 - 3	0.20±0.01	0.25	1.24	0.16	0.81	0.241	1.206
3 - 10	0.19±0.01	0.34	1.73	0.20	1.005	0.209	1.102
10 - 15	0.69 ±0.03	0.73	1.06	0.69	0.99	0.589	0.866
0.35- 10	0.49 ±0.02	0.64	1.32	0.43	0.88	0.540	1.102

The experimental neutron leakage spectra from lead sample within the 0.35-5 MeV range are sufficiently well described as a whole by calculations with ENDL-75 and ENDF/B-IV data. Only in the 1.2-3 MeV range are the experimental results higher by 10% than those calculated with mentioned data libraries. Both libraries underestimate neutron leakage in the 5-10 MeV range by approximately a factor of two. The 10-15 MeV region is satisfactorily described by these calculations. The calculation with FENDL-1 data demonstrates good agreement with the experiment in the lower energy range 0.35-1.2 MeV but in the range 1.2-3 MeV there is significant overestimation ~40%. Underestimation of neutron flux in the energy range 5-10 MeV is some less than in other calculations. The total neutron flux for FENDL-1 calculation in the energy range 0.35-10 MeV is overestimated by ~16% which compensates approximately the same underestimation in the range 10-15%.

The calculation of the neutron leakage spectrum from Be sample with the ENDF/B-IV data reproduces the experimental spectrum sufficiently well, while the difference in some ranges is significant. The calculation with ENDL-75 data differs significantly from the experiment within the entire range for measured spectrum below 10 MeV. In the 3-10 MeV energy range the overestimation by the calculation reaches 70%. Calculation fith FENDL-1 in general agrees with the experiment within ~10% except the energy range 0.7-3 MeV where overestimation rishes ~20%.

4. Measurements of the (n,γ), (n,f) and (n,2n) reaction rates in uranium and thorium spherical samples

The measurement of (n,γ), (n,f) and (n,2n) reaction rates in uranium and thorium spherical shells with layer thickness of 8 and 7 cm, respectively, was carried out by the activation method [11]. The ^{238}U(n,2n) and Th(n,2n) reaction rates were determined by measurement of U and Th activities, respectively. The U(n,γ) and Th(n,γ) processes rates are determined from measuring the activity of daughter nuclei Np and Pa, respectively. Fission fragments of Ce and La were chosen as an indicators of fission rate for U, and correspondingly Ce was chosen for Th. The radial and angular distributions of reaction rates in samples were measured. The use of high-threshold detector Cu(n,2n) made it possible to separate in (n,2n) and (n,f) reaction rates the components corresponding to 14 MeV source neutrons and inelastic interaction secondary neutrons.

The experimental and calculated (n,2n), (n,γ) and (n,f) reaction rates integrated over volume of uranium and thorium samples, as well as (n,2n) and (n,f) reaction components on source neutrons and inelastic interaction secondary neutrons, are listed in Tables 11 and 12.

From Table 11 the following conclusions can be drawn.

The experimental ^{238}U(n,2n) reaction rate with an error 4.7% is higher than the one calculated using ENDL-75 constants by 12%, and with ENDF/B-IV constants, by 18%.

The calculated U(n,γ) reaction rates for both constant libraries are close to the experimental ones.

The measured U(n,f) reaction rate at an error 4.1% agrees satisfactorily with the corresponding calculated results with ENDL-75 data, but is higher by 9% than the calculated fission rate with ENDF/B-IV data. The comparison of experimental and calculated U(n,2n) and U(n,f) reaction rates corresponding to source neutrons and secondary neutrons has demonstrated:

- the calculation with both data libraries describes satisfactorily, within the limits of experimental errors, the U(n,2n) and U(n,f) reaction rates for source neutrons;
- the experimental U(n,2n) reaction rate for secondary neutrons at an error of 29% is 2.4 times higher than the corresponding calculated value with ENDL-75 data and 2.2 times higher than that with ENDF/B-IV data;
- the experimental U(n,f) reaction for secondary neutrons within the limits of experimental error of 10% is in agreement with ENDL-75 constants, but is 1.3 times in excess of the corresponding calculated value with ENDF/B-IV data.

Table 11. Integral (n,2n), (n,f) and (n, γ) reaction rates in uranium sample

Reaction type		Total reaction rate		Reaction rate for source neutrons		Reaction rate for secondary neutrons	
		ENDL-75	ENDF/B-IV	ENDL-75	ENDF/B-IV	ENDL-75	ENDF/B-IV
U(n,2n)	experim.	0.254±0.012		0.199±0.010		0.055±0.016	
	calcul.	0.227	0.216	0.204	0.191	0.023	0.025
	C/E	1.12	1.18	0.975	1.042	2.4	2.2
U(n,f)	experim.	0.487±0.020		0.250±0.013		0.237±0.024	
	calcul.	0.476	0.447	0.260	0.265	0.216	0.182
	C/E	1.023	1.09	0.96	0.94	1.097	1.3
U(n,γ)	experim.	0.295±0.012		-		-	
	calcul	0.287	0.282	-		-	
	C/E	1.028	1.046	-		-	

Table 12. Integral (n,2n), (n,f) and (n,γ) reaction rates for thorium sample.

Reaction type		Total reaction rate		Reaction rate for source neutrons		Reaction rate for secondary neutrons	
		ENDL-75	ENDF/B-IV	ENDL-75	ENDF/B-IV	ENDL-75	ENDF/B-IV
Th(n,2n)	experim.	0.233±0.010		0.198±0.009		0.035±0.013	
	calcul	0.208	0.233	0.194	0.230	0.014	0.003
	C/E	1.12	1.0	1.02	0.86	2.5	11.7
Th(n,f)	experim.	0.077±0.004		0.056±0.003		0.021±0.005	
	calcul.	0.066	0.066	0.054	0.056	0.012	0.01
	C/E	1.17	1.17	1.04	1.0	1.75	2.1
Th(n,γ)	experim.	0.051±0.002		-		-	
	calcul.	0.094	0.074	-		-	
	C/E	0.54	0.69	-		-	

From table 12 the following conclusion can be drawn.

- The experimental Th(n,2n) reaction rate at an error of 4.3% is higher than the one calculated using ENDL-75 data by 12% and agrees with the calculated one with ENDF/B-IV data.

- The experimental Th(n,γ) reaction rate at an experimental error of 4% is significantly, by 46% lower than the one calculated using ENDL-75 data and by 31% lower than that with ENDF/B-IV data.

- The experimental Th(n,f) reaction rate at an error of 5.2% is higher by 17% than the one calculated using either ENDL-75 or ENDF/B-IV data.

The comparison of experimental and calculated Th(n,2n) and Th (n,2n) and Th(n,f) reaction rates corresponding to source neutrons and secondary neutrons has indicated the following.

- The calculation using ENDL-75 data satisfactorily describes, in the limits of experimental errors, the reaction rates Th (n,2n) and Th(n,f) for source neutrons and Th(n,f) is satisfactorily described using ENDF/B-IV data.

- The experimental Th(n,2n) reaction rate for source neutrons at an error of 4.5% is less than that calculated with ENDF/B-IV data by 14%.

- The experimental Th(n,2n) reaction rate for secondary neutrons at an error of 37 % is 2.5 times higher than that calculated using ENDL-75 data and is more than one order of magnitude higher than that calculated with ENDF/B-IV data,

- The experimental Th(n,f) reaction rate for secondary neutrons at an error of 24% is 1.6 times higher than the corresponding calculated value with ENDL-75 data and 2.1 times higher than that with ENDF/B-IV data.

4. Conclusions

Considering the results of calculations and experiments as a whole one can conclude:

- though the calculation with ENDE/B-IV data gives better description of the neutron spectrum shape for uranium, the total neutron multiplication for uranium sample is reproduced more accurately by the calculation with ENDL-75 data;

- inaccurate calculations with both data libraries of the secondary neutron spectrum may be the main reasons for underestimation in calculation of neutron multiplication in Th sample;

- though the calculations of lead samples with ENDL-75 and ENDF/B-IV data in the neutron spectrum shape are correlated in like manner with the experiment, nevertheless, the total neutron multiplication is described better by the calculation with ENDF/B-IV data. The total neutron flux for FENDL-1 calculation in the energy range 0.35-10 MeV is overestimated by ~16% which compensates approximately the same underestimation in the range 10-15%.;

- the discrepancy between the experimental and calculated neutron spectrum for the beryllium assemblies with ENDF/B-IV data does not lead to a difference in neutron multiplication;

- the larger neutron multiplication for Be in calculations with ENDL-75 data is explained by significant overestimation in the calculated spectrum of the neutron flux in the range over the Be(n,2n) reaction threshold;

- calculation of beryllium assembly with FENDL-1 data in general agrees with the experiment within ~10% except the energy range 0.7-3 MeV where overestimation riches ~20%.

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FIGURES

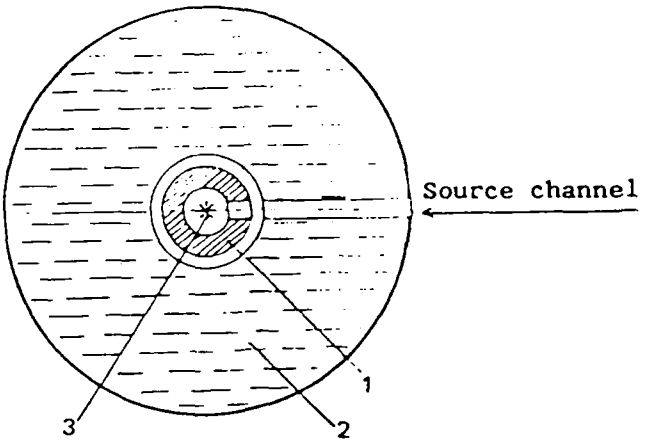

Fig. 1. Schematic representation of the experimental system for measuring the multiplication factor of 14 MeV neutrons: 1. Multiplier; 2. External moderator; 3. Source of 14 MeV neutrons.

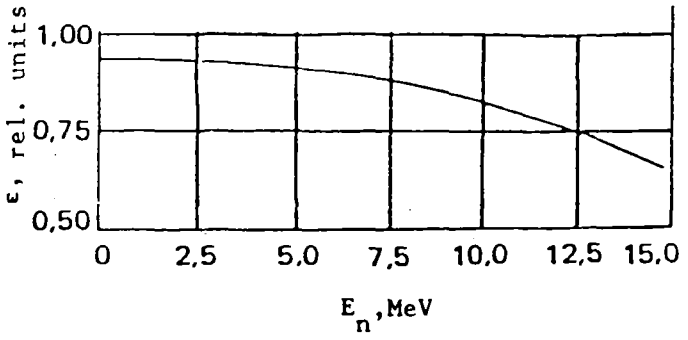

Fig. 2. Energy dependence of neutron recording efficiency of a total absorption detector.

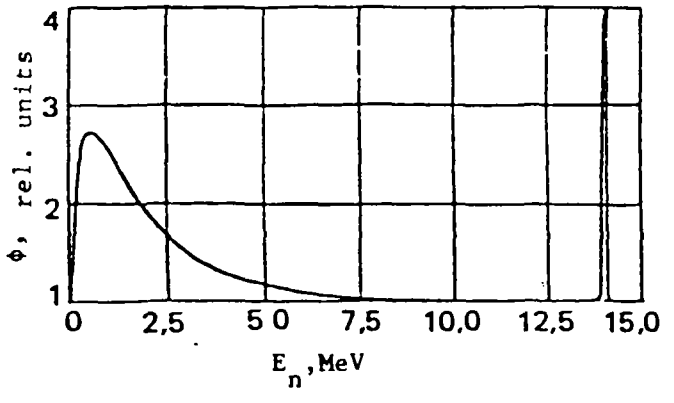

Fig. 3. Spectrum of neutron leaking from a uranium assembly with 8 cm wall thickness.

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