



IAEA

International Atomic Energy Agency

INDC(NDS)-0689
Distr. AD,AL,BN,J,SD

INDC International Nuclear Data Committee

High resolution measurements of aggregate delayed neutron spectra in different time intervals from thermal neutron induced fission of ^{235}U

V.M. Piksaikin, A.S. Egorov, A.A. Goverdovski,
D.E. Gremyachkin, K.V. Mitrofanov

State Scientific Center of the Russian Federation
Institute of Physics and Power Engineering,
Obninsk, Russia

September 2015

IAEA Nuclear Data Section
Vienna International Centre, P.O. Box 100, 1400 Vienna, Austria

Selected INDC documents may be downloaded in electronic form from
<http://www-nds.iaea.org/publications>
or sent as an e-mail attachment.

Requests for hardcopy or e-mail transmittal should be directed to
NDS.Contact-Point@iaea.org

or to:

Nuclear Data Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna
Austria

Printed by the IAEA in Austria

September 2015

High resolution measurements of aggregate delayed neutron spectra in different time intervals from thermal neutron induced fission of ^{235}U

V.M. Piksaikin, A.S. Egorov, A.A. Goverdovski,
D.E. Gremyachkin, K.V. Mitrofanov

Abstract

Composite delayed energy spectra from epithermal neutron induced fission of ^{235}U have been measured for twelve delay-time intervals extending from 0.12 to 150 s. These data provide a comprehensive set of composite spectra with enhanced sensitivity to neutrons emitted in less than 1 s following fission. Extensive comparisons between these measurements and the composite spectra obtained by summation are presented and discussed herein.

September 2015

Contents

1. Introduction	7
2. Experimental method	8
3. Processing of experimental data.....	13
4. Results	14
5. Equilibrium delayed neutron spectrum	21
6. Conclusions	24
References	25

1. Introduction

The phenomenon of emission of delayed neutrons (DN) that accompanies the process of fission of heavy nuclei is of considerable interest for understanding many fundamental aspects of nuclear physics, astrophysics, and the process of nuclear fission. Studying the process of delayed neutron emission provides unique insight into the form of the strength function describing beta decay, the nuclear level density, the competition between neutron and gamma emission and other properties of fission fragments that are far from the valley of beta stability. The energy spectrum of delayed neutrons emitted from fission of heavy nuclei is of great importance for the calculation of the effective delayed neutron fraction. At present there are two sources of information on the delayed neutron spectra – experimental data on the aggregate delayed neutron spectrum and experimental and theoretical data on delayed neutron spectra of individual precursors. On the basis of the summation method, the delayed neutron spectra from individual precursors are used to calculate the group and composite spectra by summing the contributions from individual precursors. Some of the delayed neutron spectra for important precursors are not available and therefore the experimental data have to be supplemented by nuclear model calculations. This approach was the basis of the delayed neutron spectra database incorporated in the ENDF/B VII.1 library.

However, to date there is no theoretical model capable of reproducing even the gross features of the experimental data. A comparison of the measured spectral data of individual precursors with the results of calculations obtained from different theoretical models (one-parameter evaporation model, Maxwell distribution, BETA code model, “gross theory” beta-delayed model) reveals that all of these approaches suffer from one shortcoming or another. Particularly the simplified one-parameter evaporation model and the more sophisticated BETA code which is based on statistical model calculations with appropriate nuclear level information for the precursor, its daughter, and granddaughter, fail to describe correctly both the overall form and the structure of the low-energy part of the delayed neutron spectrum [1]. Recently Möller and Kawano [2] developed a more microscopic technique to calculate the delayed-neutron energy spectra for fission products. The calculated delayed-neutron spectra, which are purely theoretical predictions, reasonably agree with those evaluations that are based on experimental data.

The Nuclear Data Section of the International Atomic Energy Agency (IAEA) has initiated a Coordinated Research Project on the Development of a Reference Database for beta-delayed neutron emission [3], with the aim of generating a dedicated database containing a compilation of microscopic and macroscopic (aggregate) DN data, evaluations and benchmarks of the microscopic data against measured aggregate data. One of the tasks defined within this project is to estimate the consistency of the different approaches that are or have been used to generate the microscopic databases of DN data for individual precursors by comparing with aggregate delayed neutron spectra that include contributions from a variety of individual precursors. To achieve this task, we have measured composite delayed energy spectra in different time intervals after the end of irradiation of ^{235}U sample by epithermal neutrons, and compared them with the results of existing microscopic DN databases using the summation method.

2. Experimental method

The measurements of the energy spectra of delayed neutrons emitted in the fission of ^{235}U nuclei by epithermal neutrons were carried out at the facility of the electrostatic accelerator KG-2.5 IPPE. The main components of the set-up are shown in Fig. 1.

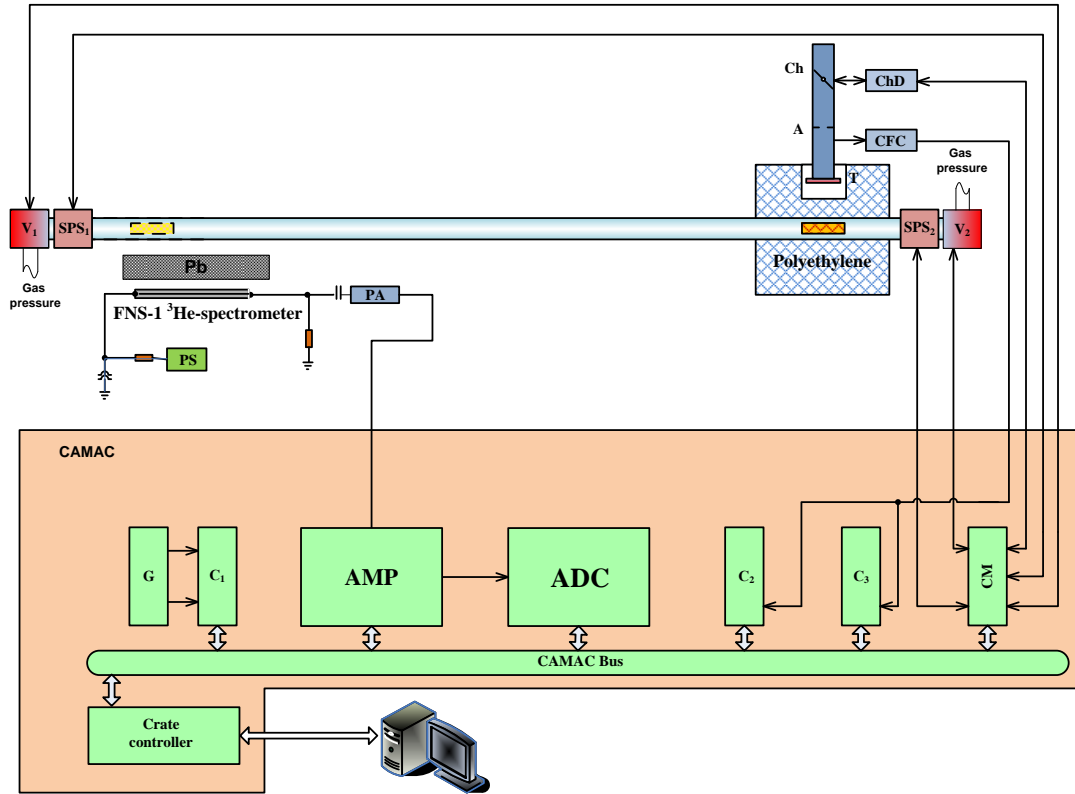


Fig. 1. Block diagram of the experimental setup: (PAD) preamplifier, amplifier, and discriminator; (S) summator; (PA) preamplifier; (V) electromagnetic valve; (SPS) sample position sensor; (CM) control unit; (CFC) current-to-frequency converter; (ADC) analog-to-digital converter; (PSc) preset-scaler; (G) quartz generator of pulses; (PS) power source; (Ch) chopper; (ChD) magnetic chopper drive; (A) ion guide aperture; (T) accelerator target; (C₁) counter with a preset exposure time; (C₂) counter of total counts from the CFC; (C₃) counter of the CFC counts within preset time intervals.

Measurements have been performed by the method of cyclic irradiation of ^{235}U sample by epithermal neutrons followed by delayed neutron counting with an ^3He -spectrometer. The neutron source was based on utilization of the $\text{T}(p,n)^3\text{He}$ reaction on the solid tritium target of the IPPE electrostatic accelerator CG-2.5. For production of the epithermal neutron flux the accelerator target was surrounded by a polyethylene cube of 27 cm. The ^{235}U metallic sample was placed in the horizontal hole of the cube. During irradiation the sample was located at about 3.2 cm from the neutron target. Transport of the sample from the irradiation position to the ^3He -spectrometer was accomplished by a fast pneumatic system.

The sample transport tube used was a thin-walled stainless steel tube with an outer diameter 10 and a wall thickness of 0.3 mm. Two solenoid valve controls the supply of compressed air in the guide tube. The position of the sample in the neutron detector was fixed with a plug with a hole in the center for

controlling the discharge of excess pressure in front of a moving sample and mitigating the impact of the sample. Information on the status of the sample was obtained from the two photodiodes and light sources mounted on the guide tube in position in the center of irradiation and neutron spectrometer. On the average the sample delivery time was 120 ms which is short enough to get information related to the shortest precursors groups.

The data acquisition and processing system makes it possible to measure the following parameters: time dependencies of the neutron flux intensity from the target and the ion current on the target of the accelerator, sample transportation time, the pulse height distribution of delayed neutrons coming from the neutron spectrometer. The PC computer serves as a central processor controlling the irradiation and counting time, the number and the width of time channels for the delayed neutron spectra measurements.

A schematic diagram of the experimental arrangement is shown in Fig.2. A neutron energy spectrum calculated using the Monte Carlo program is presented in Fig.3. The average energy of slowing down neutrons averaged over the sample was 2.85 eV.

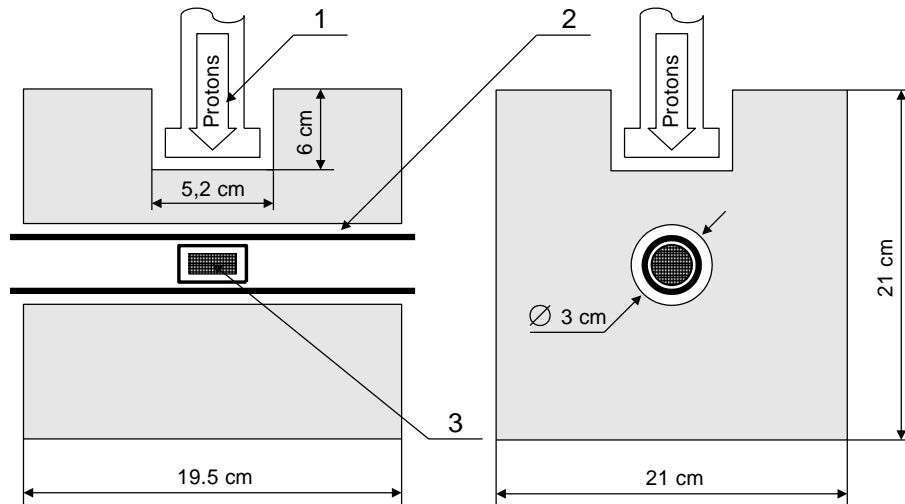


Fig. 2. Neutron source (accelerator target) and sample position for low energy experiment. 1 -target assembly, 2 -tube of sample transfer system, 3 - ^{235}U sample.

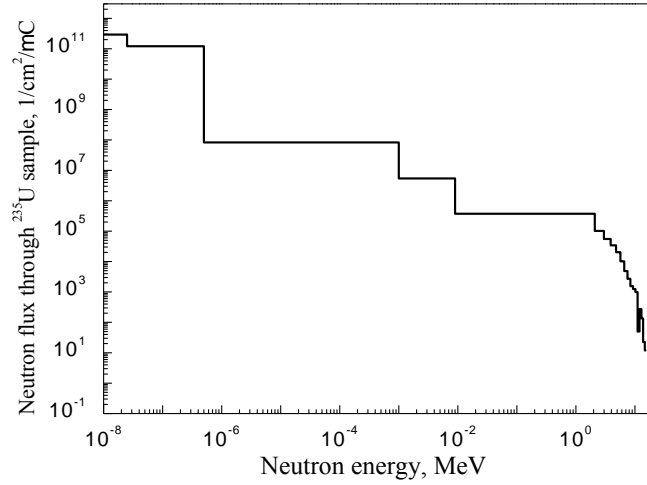


Fig. 3. Neutron energy distribution averaged over the sample under investigation. Average energy of slowing down neutrons is 2.85 eV.

A high resolution ^3He -spectrometer of FNS-1 type was employed for the measurements of delayed neutron spectra. The ^3He -spectrometer is a cylindrical gridded ion chamber with active volume dimensions of 5 cm diameter and 15 cm length. It is filled with ^3He at a partial pressure of 6 atm, Ar of 3 atm, and CH_4 of 0.5 atm. This value of gas pressure of the ^3He -spectrometer was enough to enable full ranges of tritons and protons from the $^3\text{He}(n,p)^3\text{H}$ reaction in active volume of the spectrometer for primary neutrons with energy up to 2.5 MeV. The lead shielding of 6.1 cm thick placed between the ^{235}U sample and ^3He -specrometer were used to suppress gamma-background from irradiated sample and to decrease the pile-up effects.

Measurements with different irradiation and counting time intervals were made to emphasize particular delayed neutron precursors. Two sets of measurements have been performed. The first one was made with irradiation time of 120 s which was followed by measurements of delayed neutron spectra in the following sequence of time intervals after the end of irradiation: 0-2, 2-12, 12-22, 22-32, and 32-152 s. The second set of measurements was made with irradiation time of 20 s and measurements of delayed neutron spectra in the time intervals: 0-1, 1-2, 2-3, 3-4, and 4-44 s. The neutron background was measured in the same conditions as in the experiments but instead of uranium samples we used empty container.

The raw experimental data compiled in these two types of experiments are presented in Fig. 4 and Fig. 5.

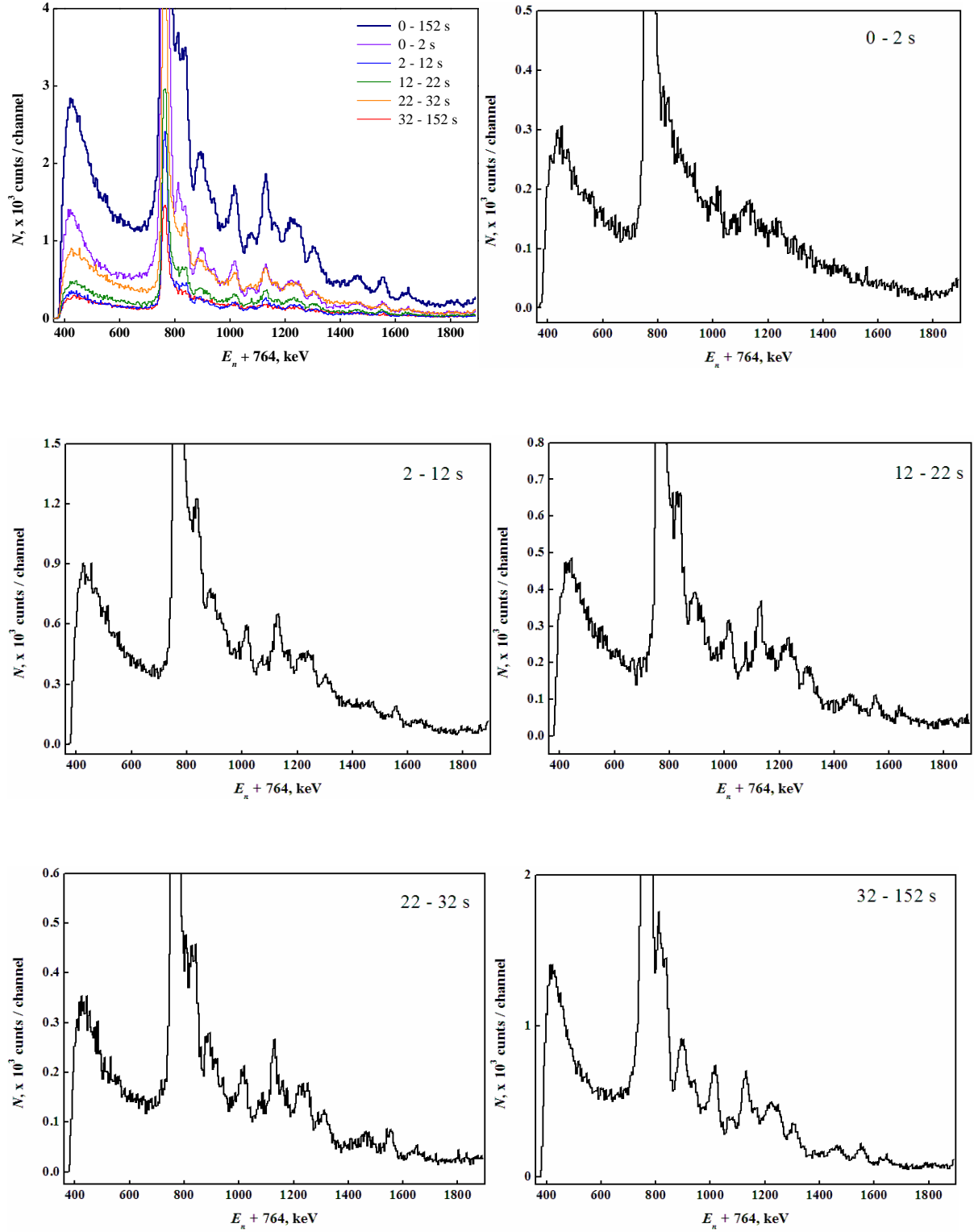


Fig. 4. The pulse height distribution of delayed neutrons from thermal neutron induced fission of ^{235}U registered by FNS-1 type ^3He -spectrometer. The irradiation time is 120 s. Measurements of delayed neutron spectrum were made in the following time sequence after the end of irradiation: 0-2 s, 2-12 s, 12-22 s, 22-32 s, and 32-152 s (see inserts in figures).

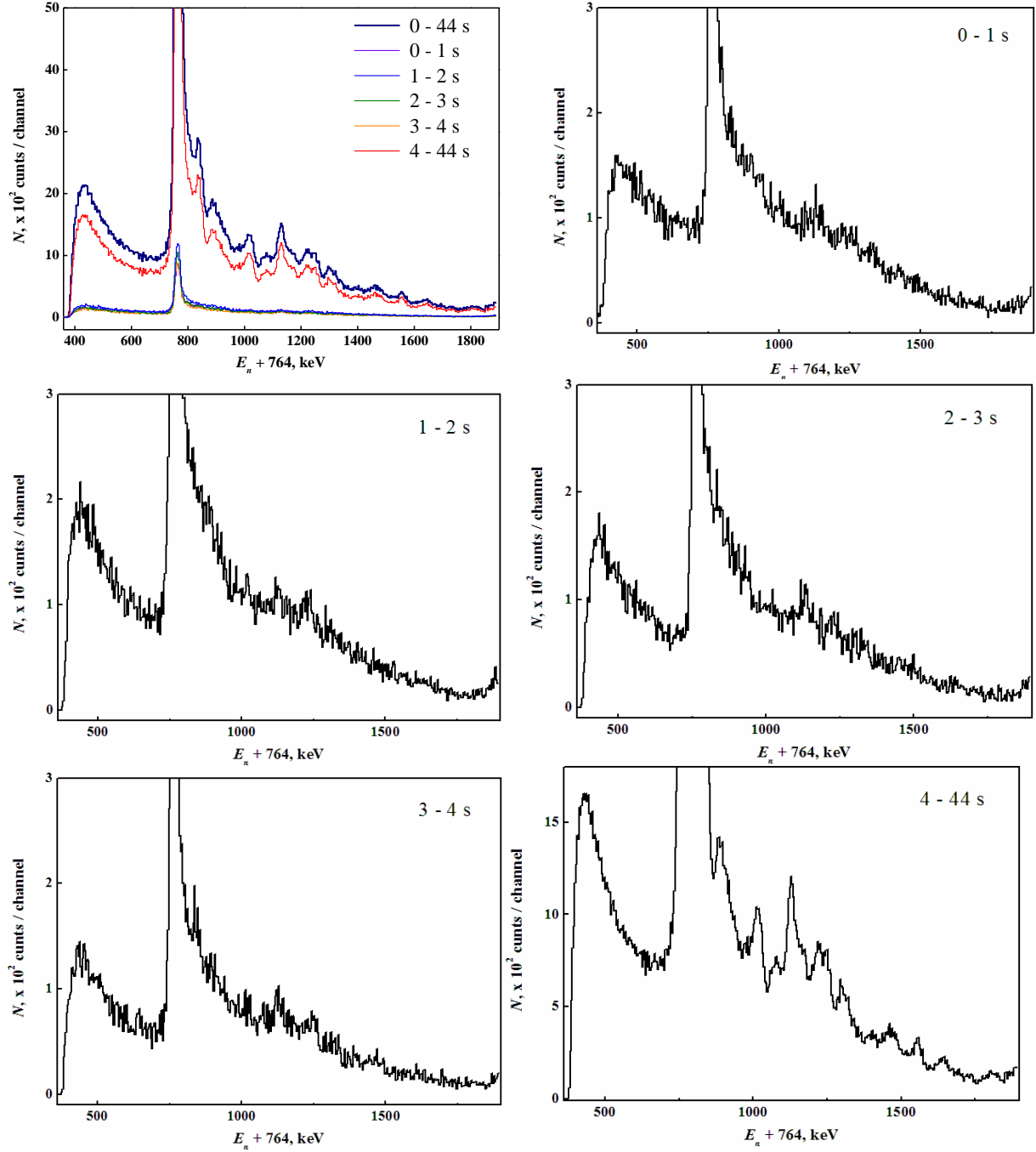


Fig. 5. The pulse height distribution of delayed neutrons from thermal neutron induced fission of ^{235}U registered by FNS type ^3He -spectrometer. The irradiation time is 20 s. Measurements of delayed neutron spectra were made in the following sequence of time intervals after the end of irradiation: 0-1 s, 1-2 s, 2-3 s, 3-4 s, and 4-44 s (see inserts in figures).

3. Processing of experimental data

It is well known that the spectral data obtained with the help of a neutron spectrometer based on the $^3\text{He}(n,p)^3\text{H}$ reaction are mainly distorted by recoil nuclei and the edge effect which create a continuum below full-energy peak. There are different approaches to account for these distortions. In the present work we use the procedures that were developed and carefully tested for the FNS-1 type ^3He -spectrometer by Ohm et al. [4]. The relative efficiency of the ^3He -spectrometer was estimated using Eq. (1) from [5] which was obtained for the FNS-1 type spectrometer in the energy range $0.019 \leq E_n \leq 2.77$ MeV by Franz et al. [6]:

$$\begin{aligned} \varepsilon = & \exp[3.75 - 0.629 \cdot \ln E_n] - 0.15 \cdot \exp\left[-1/2 \cdot \left(\frac{E_n - 120}{21}\right)^2\right] - 0.39 \cdot \exp\left[-1/2 \cdot \left(\frac{E_n - 250}{89}\right)^2\right] - \\ & - 0.13 \cdot \exp\left[-1/2 \cdot \left(\frac{E_n - 440}{127}\right)^2\right] + 0.057 - 3.06 \cdot 10^{-5} E_n \end{aligned} \quad (1)$$

Equation (1) was tested at several energy points in a separate experiment. The final data for the energy dependence of ^3He -spectrometer efficiency are presented in Fig.6.

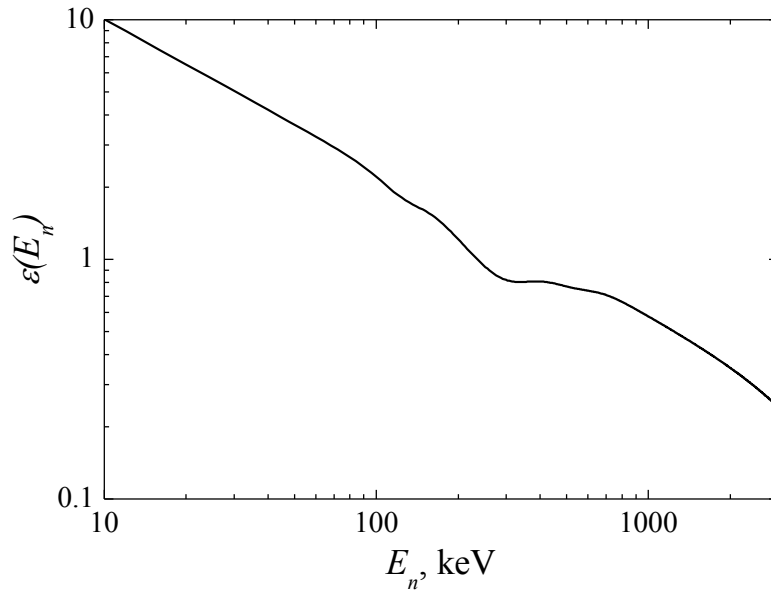


Fig. 6. Relative efficiency of FNS-1 type ^3He -spectrometer.

A correction was applied for the neutron flux attenuation effect in the Pb gamma-shield (6 cm thick) of the neutron spectrometer. The transmission correction was made on the basis of Monte-Carlo calculations and appropriate measurements which were made with the help of mono-energetic neutrons.

4. Results

The analysis of the experimental data produced two sets of spectra. The first one is the aggregate delayed neutron spectra measured in time intervals 0.12-2, 2-12, 12-22, 22-32, 32-152 s after the end of 120 s - irradiation of ^{235}U sample. The second type of data is the aggregate delayed neutron spectra measured in time intervals 0-1, 1-2, 2-3, 3-4, and 4-44 s after the end of 20 s - irradiation.

The spectra measured in different time windows are compared with spectra calculated by appropriately summing spectra from individual precursors. One set of these calculated spectra data in a definite time-window was obtained with the help of formula written for N considered precursors of delayed neutrons

$$N(E_n)dE = A \sum_{i=1}^N \left[\left(\frac{P_{ni} CY_i}{\lambda_i} \right) (1 - e^{-\lambda_i t_{irr}}) (e^{-\lambda_i t_d}) (1 - e^{-\lambda_i \Delta t_c}) \right] T_i \chi_i(E_n) dE_n, \quad (2)$$

$$T_i = \left[\frac{M}{1 - e^{-\lambda_i T}} - e^{-\lambda_i T} \frac{1 - e^{-M \lambda_i T}}{(1 - e^{-\lambda_i T})^2} \right], \quad (3)$$

where T_i term describes dependence of sample activation on number of irradiation cycles; A is the saturation activity; P_{ni} is the emission probability of the i -th precursor; CY_i is the cumulative yield of i -th precursor; $\chi_i(E_n)$ is the delayed neutron energy spectrum associated with i -th precursor; λ_i is the decay constant of i -th precursor; t_{irr} is the irradiation time in s; t_d is the delay time in s; Δt_c is the neutron counting time in s; M is the number of cycles; T is the period of one irradiation cycle (irradiation-cooling-counting).

P_{ni} data were taken from the evaluation of Blachot et al. [7] with introduction of some corrections based on the data from Rudstam et al. [8]. CY_i data were taken from the JEF 2.2 file. Only experimental data were considered for delayed neutron spectra from individual precursors. The set of individual precursors is shown in Table I.

Table I. The list of delayed neutron precursors used in the present summation calculations.

Presursor	$^{87}_{35}\text{Br}$	$^{141}_{55}\text{Cs}$	$^{137}_{53}\text{I}$	$^{136}_{52}\text{Te}$	$^{88}_{35}\text{Br}$	$^{72}_{29}\text{Cu}$	$^{146}_{57}\text{La}$	$^{138}_{53}\text{I}$	$^{93}_{37}\text{Rb}$
$T_{1/2}$, s	55.65	24.84	24.5	17.63	16.29	6.49	6.27	6.23	5.84
Presursor	$^{92}_{37}\text{Rb}$	$^{89}_{35}\text{Br}$	$^{79}_{31}\text{Ga}$	$^{94}_{37}\text{Rb}$	$^{139}_{53}\text{I}$	$^{119}_{47}\text{Ag}$	$^{85}_{33}\text{As}$	$^{90}_{35}\text{Br}$	$^{114}_{45}\text{Rh}$
$T_{1/2}$, s	4.49	4.40	2.85	2.70	2.28	2.1	2.02	1.91	1.85
Presursor	$^{143}_{55}\text{Cs}$	$^{119}_{46}\text{Pd}$	$^{142}_{55}\text{Cs}$	$^{80}_{31}\text{Ga}$	$^{135}_{51}\text{Sb}$	$^{103}_{41}\text{Nb}$	$^{148}_{57}\text{La}$	$^{129}_{49}\text{In}$	$^{120}_{47}\text{Ag}$
$T_{1/2}$, s	1.79	1.76	1.684	1.68	1.68	1.5	1.26	1.23	1.23
Presursor	$^{81}_{31}\text{Ga}$	$^{134}_{50}\text{Sn}$	$^{144}_{55}\text{Cs}$	$^{147}_{56}\text{Ba}$	$^{140}_{53}\text{I}$	$^{113}_{44}\text{Ru}$	$^{129m}_{49}\text{In}$	$^{148}_{56}\text{Ba}$	$^{145}_{55}\text{Cs}$
$T_{1/2}$, s	1.22	1.05	0.99	0.89	0.86	0.8	0.61	0.61	0.59
Presursor	$^{91}_{35}\text{Br}$	$^{120}_{46}\text{Pd}$	$^{141}_{53}\text{I}$	$^{95}_{37}\text{Rb}$	$^{92}_{35}\text{Br}$	$^{146}_{55}\text{Cs}$	$^{130}_{49}\text{In}$	$^{147}_{55}\text{Cs}$	$^{96}_{37}\text{Rb}$
$T_{1/2}$, s	0.54	0.5	0.43	0.38	0.34	0.32	0.29	0.23	0.2
Presursor	$^{97}_{37}\text{Rb}$	$^{98}_{37}\text{Rb}$							
$T_{1/2}$, s	0.17	0.11							

The second set of calculated aggregate spectra for the time intervals was obtained on the basis of the 8-group delayed neutron spectra included in the JEF 2.2 library and the 6-group spectra included in the ENDF/B-VI library, which in turn were derived by applying the summation techniques on a database of individual precursors developed with the help of both experimental and theoretical delayed neutron spectra [1]. The aggregate data were obtained by

$$N(E_n)dE = A \sum_{i=1}^N \left[\left(\frac{a_i}{\lambda_i} \right) (1 - e^{-\lambda_i t_{irr}}) (e^{-\lambda_i t_d}) (1 - e^{-\lambda_i \Delta t_c}) \right] T_i \chi_i(E_n) dE_n \quad (4)$$

$$T_i = \left[\frac{M}{1 - e^{-\lambda_i T}} - e^{-\lambda_i T} \frac{1 - e^{-M \lambda_i T}}{(1 - e^{-\lambda_i T})^2} \right], \quad (5)$$

where T_i term describes dependence of sample activation on number of irradiation cycles, A is the saturation activity; a_i is the relative abundances of the i -th delayed neutron group taken from JEF 2.2 or ENDF/B-VI files; $\chi_i(E_n)$ is the energy spectrum associated with i -th delayed neutron group; λ_i is the decay constant of i -th delayed neutron group; t_{irr} is the irradiation time in s; t_d – is the delay time in s; Δt_c is the delayed neutron counting in s; M is the number of cycles; T is the period of one irradiation cycle (irradiation-cooling-counting).

Comparison of measured data with calculated spectra in the appropriate time intervals in experiments with irradiation time 120 s is made in Figs.7-11.

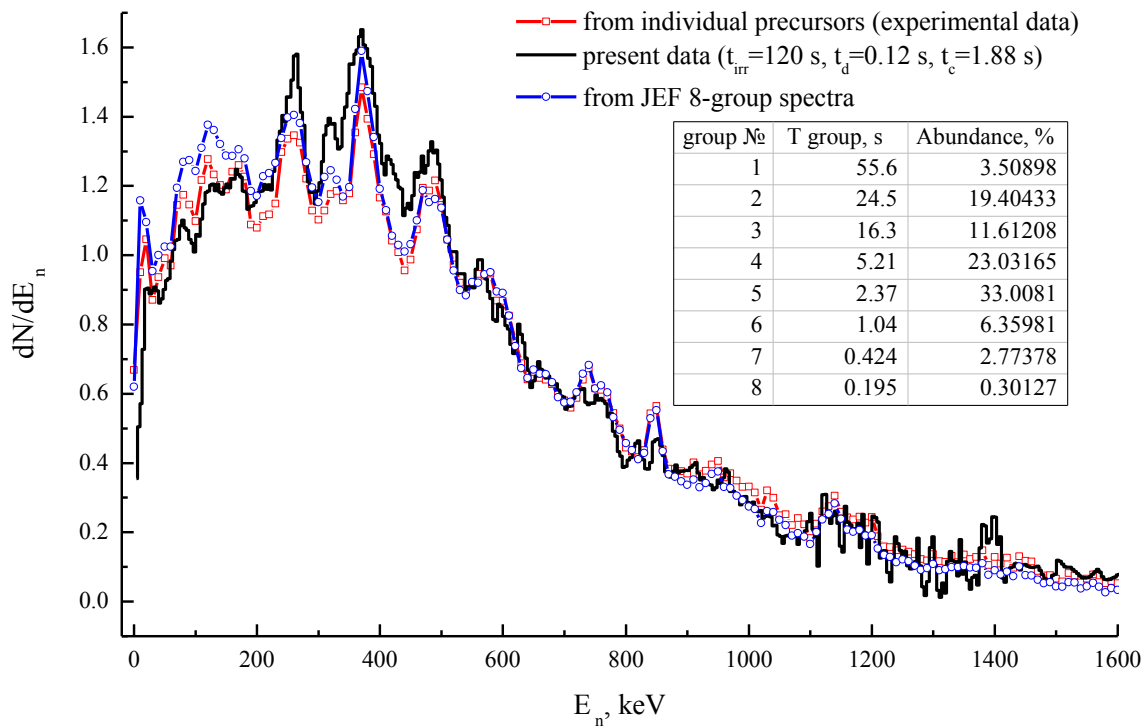


Fig. 7. Composite spectrum of delayed neutron in the time interval 0.12-2 s from neutron induced fission of ^{235}U (irradiation time – 120 s).

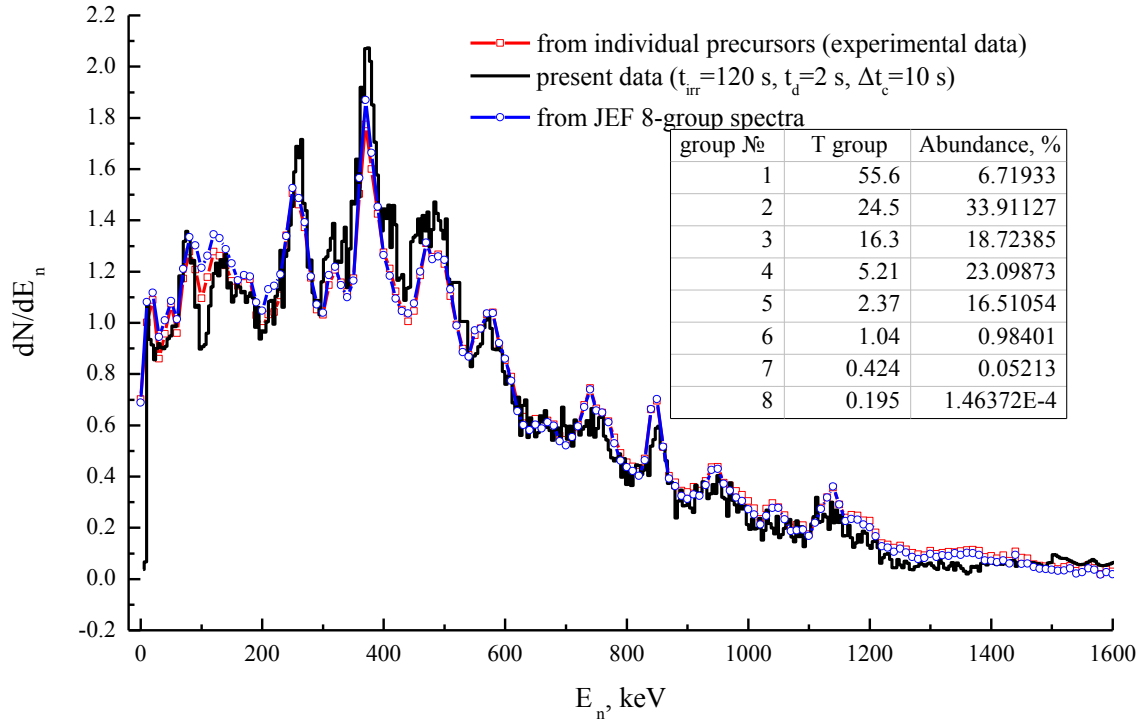


Fig. 8. Composite spectrum of delayed neutron in the time interval 2-12 s from neutron induced fission of ^{235}U (irradiation time – 120 s).

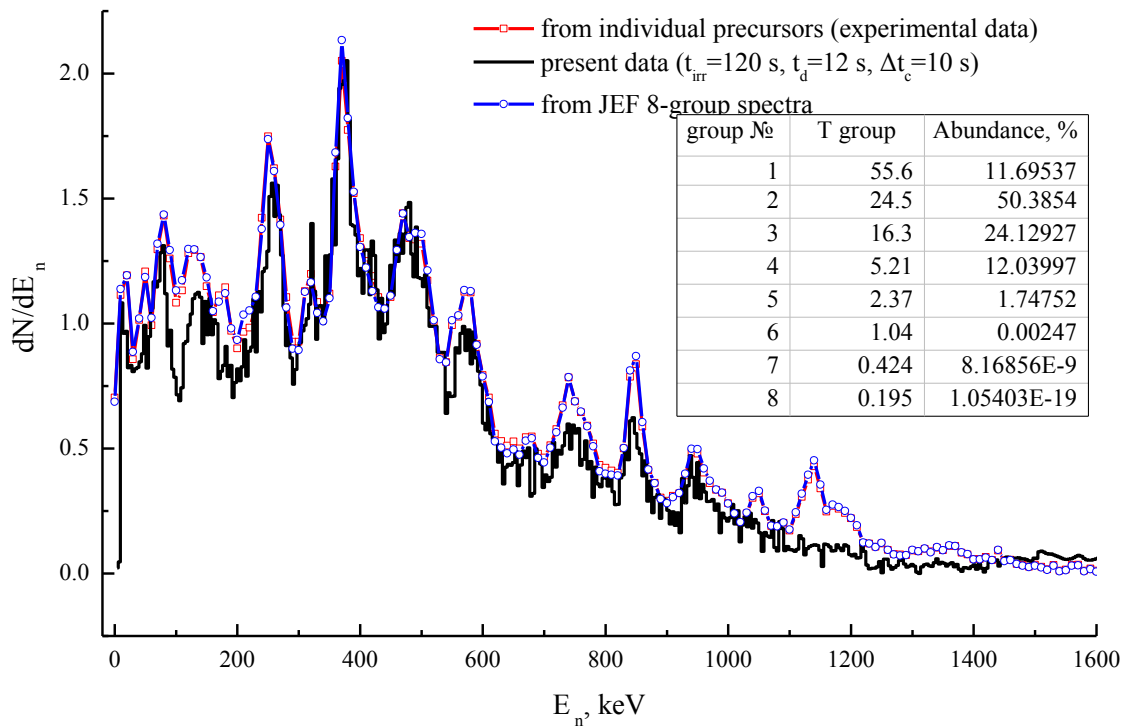


Fig. 9. Composite spectrum of delayed neutron in the time interval 12-22 s from neutron induced fission of ^{235}U (irradiation time – 120 s).

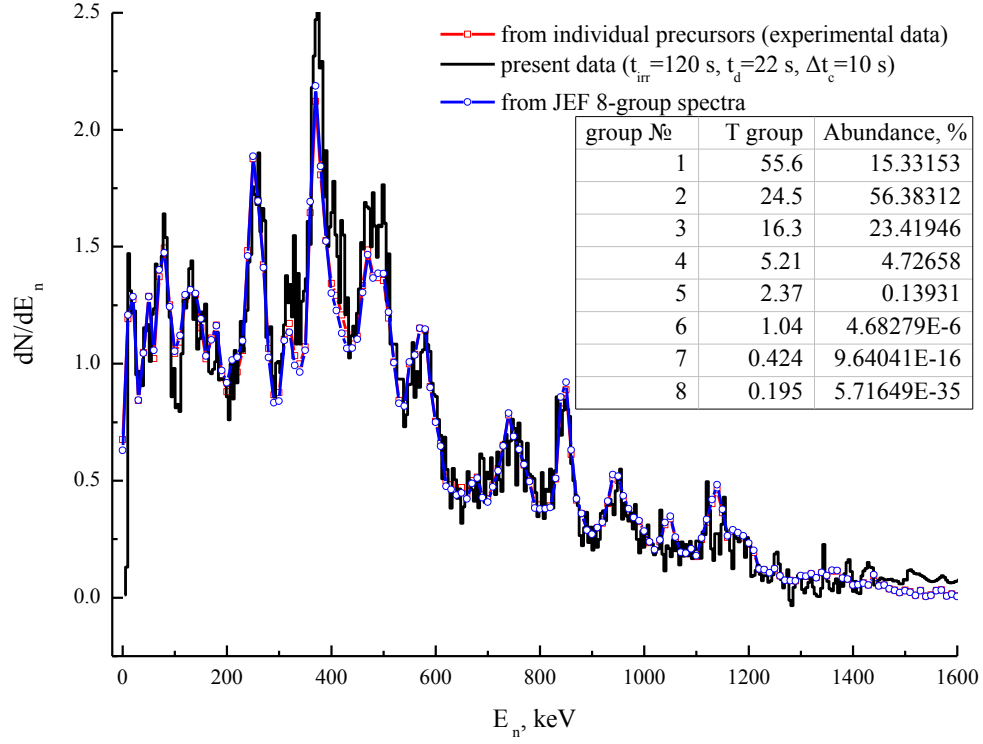


Fig. 10. Composite spectrum of delayed neutron in the time interval 22-32 s from neutron induced fission of ^{235}U (irradiation time – 120 s).

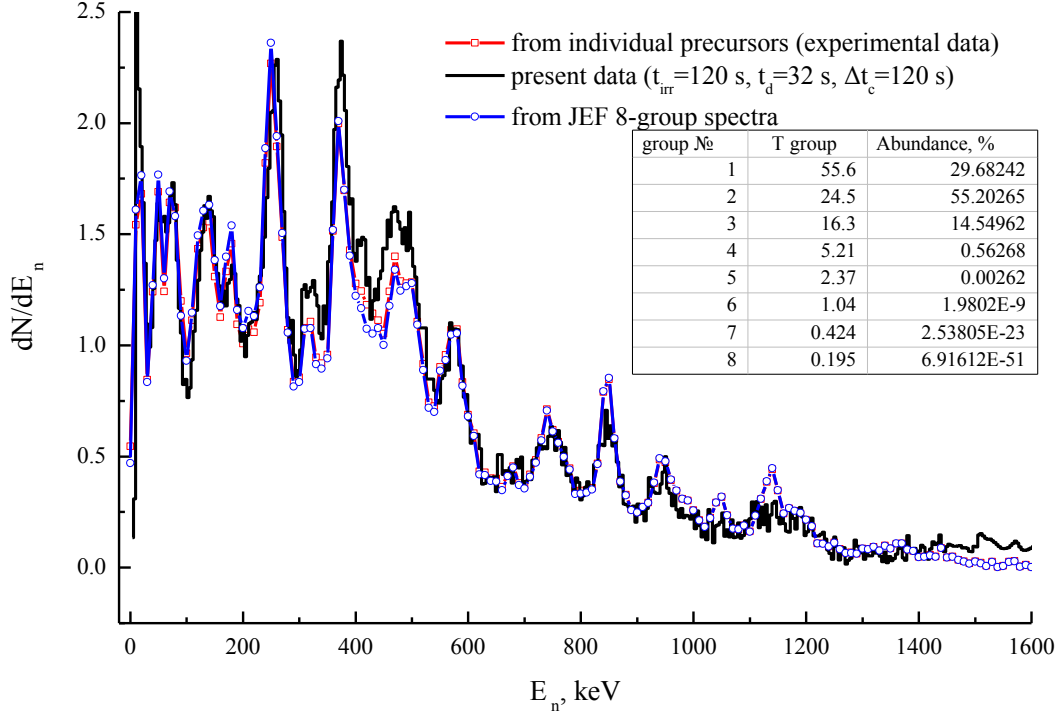


Fig. 11. Composite spectrum of delayed neutron in the time interval 32-152 s from neutron induced fission of ^{235}U (irradiation time – 120 s).

It is seen from Figs. 7-11 that DN spectra calculated in the time windows 2-12, 22-32 and 32-152 s in general agree very well with our experimental data. The main difference between the spectra is the following: both types of calculated spectra mentioned previously have lower peaks at neutron energies 250, 320, 370, 400 keV in the time window 2-12 s. Also, in the calculated spectra, the group of peaks at energies 400 keV is not resolved. In time interval 12-22 s, peaks at neutron energies 570, 740, and 850 and the group of peaks below 220 keV at neutron energy 130 keV are more intense in the calculated spectra than in the measured one. In the time windows 0.12-2 we observe that agreement between measured and calculated spectra is better for the calculated spectrum obtained from experimental precursor data.

From the comparison of the experimental and calculated spectra of delayed neutrons one can draw the following conclusions. A good agreement between the measured and calculated spectra especially in the time interval 32-152 s (see Fig.11) on the one side indicates that the experimental procedure used in the present measurements does not suffer from any systematic errors. On the other side, it also confirms that the existing experimental data base for individual precursors describes the aggregate delayed neutron spectrum rather well in long delay-time intervals. Differences in the calculated and measured aggregate spectrum observed in shorter delay-time intervals are most likely related to the rather unreliable spectral data available for individual precursors with short half-lives. In the energy range below 200 keV, there is a notable disagreement between the spectrum calculated using the database for individual precursors and that obtained from the 8-group parameterization in the JEF 2.2 library. This difference is probably a result of the inadequate theoretical approach used for the unmeasured precursors when building the 8-group parameterization combined with the procedure of redistributing individual precursor spectra between temporary groups that was implemented.

Comparison of the composite spectra measured in the experiment with irradiation time 20 s in different time windows with calculated spectra is made in Figs.12-16.

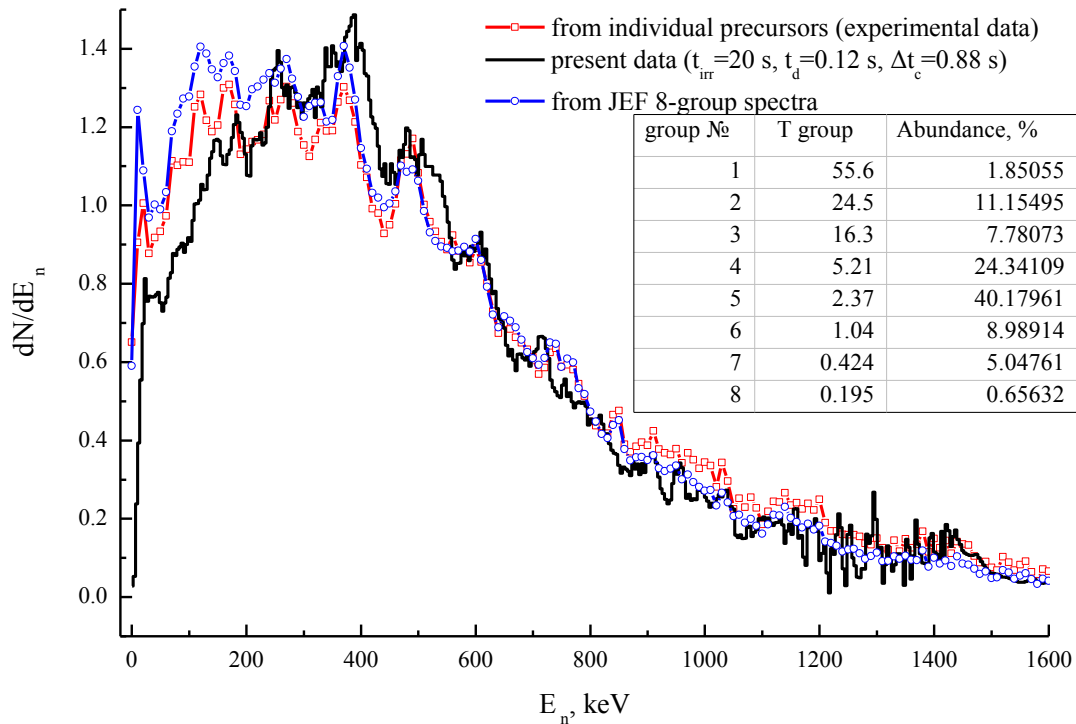


Fig. 12. Composite spectrum of delayed neutron in the time interval 0.12-1 s from neutron induced fission of ^{235}U (irradiation time – 20 s).

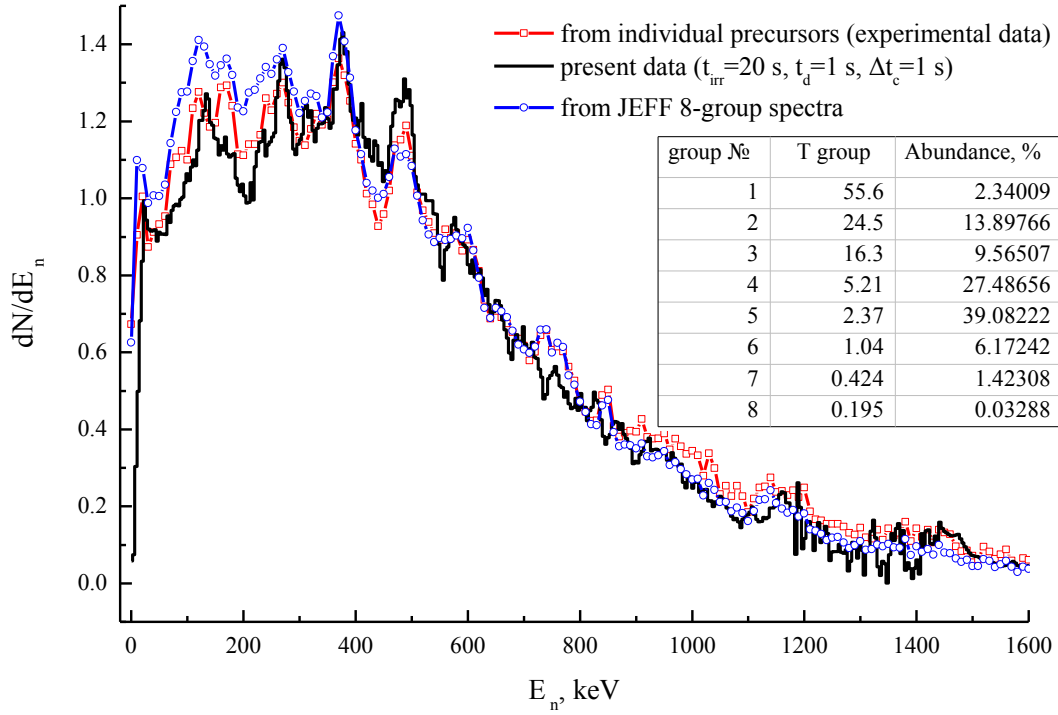


Fig. 13. Composite spectrum of delayed neutron in the time interval 1-2 s from neutron induced fission of ^{235}U (irradiation time – 20 s).

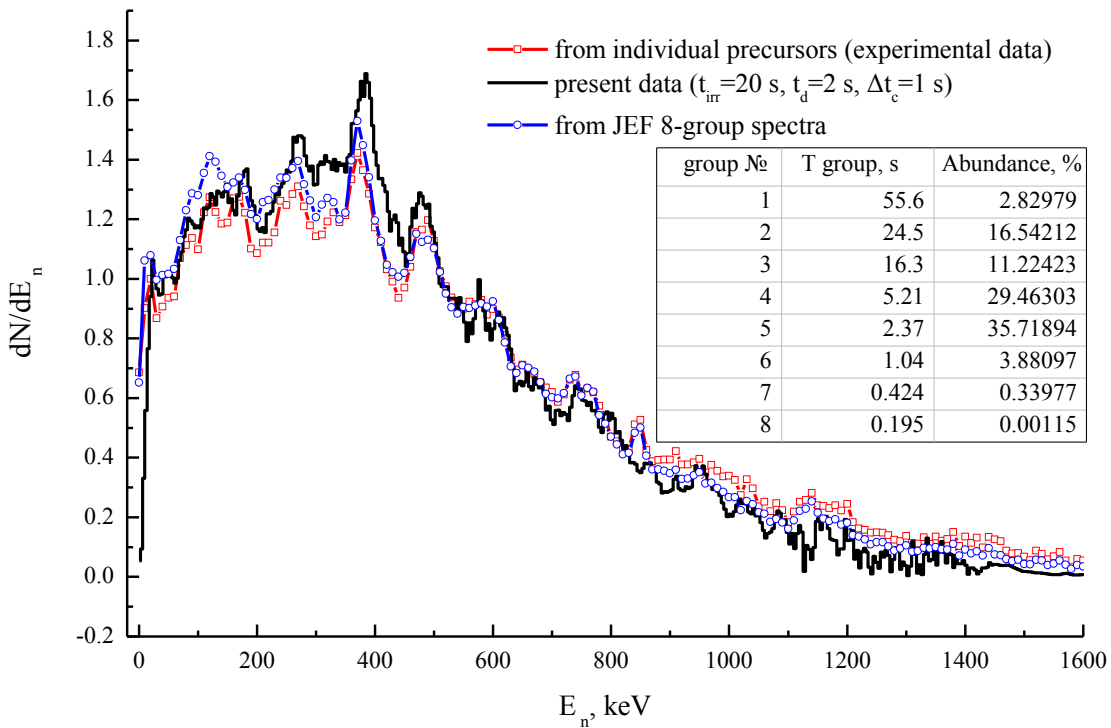


Fig. 14. Composite spectrum of delayed neutron in the time interval 2-3 s from neutron induced fission of ^{235}U (irradiation time – 20 s).

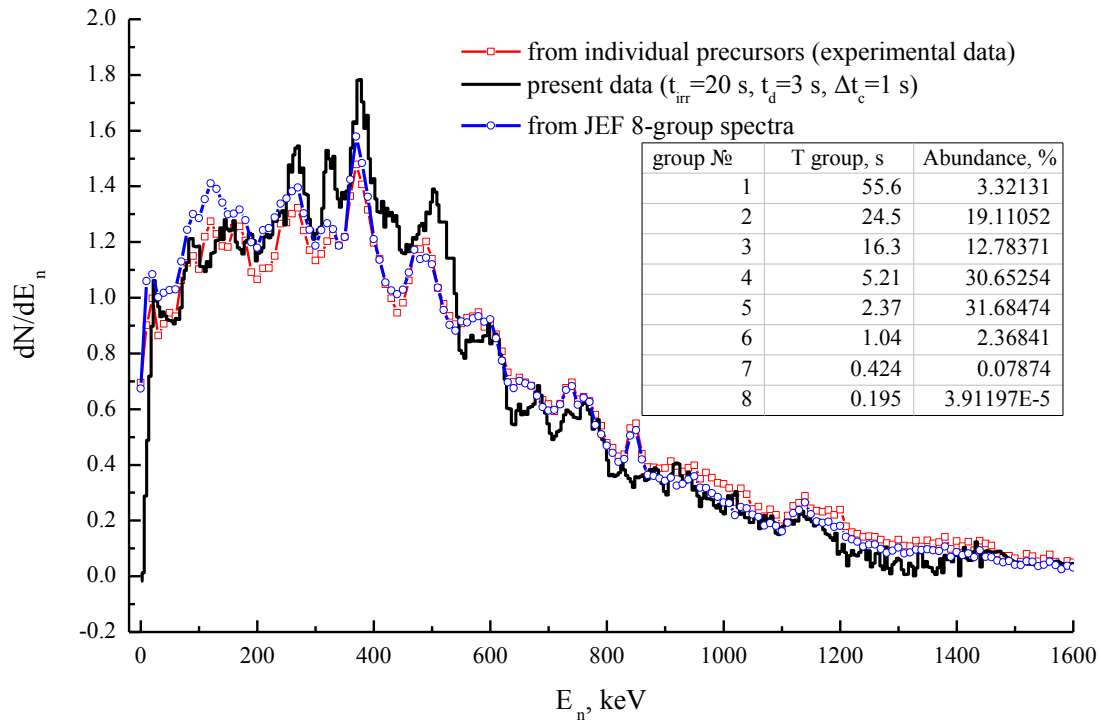


Fig. 15. Composite spectrum of delayed neutron in the time interval 3-4 s from neutron induced fission of ^{235}U (irradiation time – 20 s).

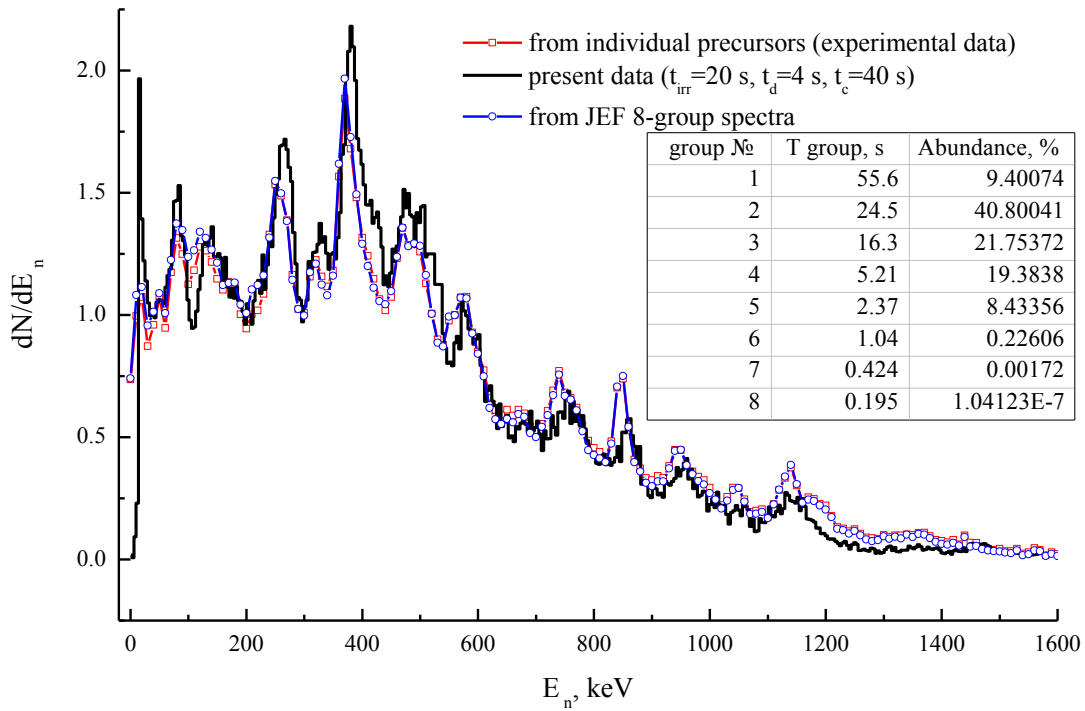


Fig. 16. Composite spectrum of delayed neutron in the time interval 4-44 s from neutron induced fission of ^{235}U (irradiation time – 20 s).

The experimental spectra in the time window 2-3, 3-4, 4-44 s with 20 s irradiation time show in general good agreement with appropriate calculated data. But as in the case of long irradiation time the experimental data have more pronounced peak structure with more intense peaks at neutron energies 270, 370, 480 keV. Peaks at neutron energies 740, 850 keV is less intense than calculated ones in the time window 4-44 s. Besides that in the time window 3-4 s calculated spectra do not reproduce peaks at 320 and 420 keV that are observed in the measured data.

It is worth noting that the composite spectra calculated on the basis of experimental data for individual precursors agree better with the measured spectra than the spectra calculated with the help of the 8-group spectra. As a rule the spectra based on the group data overestimate the intensity of delayed neutrons in the region below 200 keV especially in the case of data measured after short delays after the end of irradiation. It is most probable that the theoretical model used to calculate the spectra of possible short-lived precursors- that have not been measured- cannot reproduce the real spectra.

5. Equilibrium delayed neutron spectrum

Equilibrium delayed neutron energy spectrum in N-group model can be presented by the following expression

$$N(E_n)dE = A \sum_{i=1}^N a_i c_i(E_n) dE_n, \quad (6)$$

where A is the saturation activity; a_i is the relative abundance of i -th delayed neutron group; $\chi_i(E_n)$ is the delayed neutron spectrum of i -th group; N is the number of delayed neutron groups. Composite delayed neutron spectrum measured in different time intervals can be expressed by

$$N(E_n)dE = A \sum_{i=1}^N \left[\left(\frac{a_i}{\lambda_i} \right) (1 - e^{-\lambda_i t_{irr}}) (e^{-\lambda_i t_d}) (1 - e^{-\lambda_i \Delta t_c}) \right] T_i \chi_i(E_n) dE_n, \quad (7)$$

$$T_i = \left[\frac{M}{1 - e^{-\lambda_i T}} - e^{-\lambda_i T} \frac{1 - e^{-M \lambda_i T}}{(1 - e^{-\lambda_i T})^2} \right]. \quad (8)$$

It is seen from the above formula that the relative abundances of delayed neutron groups in composite spectrum (the term in square brackets multiplied by T_i) depend on the time constants and number of irradiation cycles used in measurements. Results obtained according to this formula are presented in the insets of all figures that show composite spectrum for each time intervals. Comparison of these data with relative abundances of the 8-group model for ^{235}U [9] shows that composite spectra measured in the first time windows (0.12-1 and 0.12-2 s) after the end of irradiation in both experiments are the closest to the equilibrium spectrum and can be considered as near-equilibrium or quasi-equilibrium. It should be noted, that composite spectrum in 0.12-1 s time-window is closer to the equilibrium spectrum than spectrum in 0.12-2 s window obtained in experiment with irradiation time – 120 s.

These data are presented in Figs. 17 and 18. In Fig.17 they are compared with spectra measured by other authors, which were calculated with the help of the group spectra obtained by decomposition of aggregate spectra. In Fig.18 comparison is made with the equilibrium spectra calculated with the help of the group spectra from JEF 2.2 and ENDF/B-VI library as well as with experimental delayed neutron spectra from individual precursors. Rudstam's equilibrium spectrum [8] presented in Fig.18 are based on early measured P_n and $\chi_i(E_n)$ data.

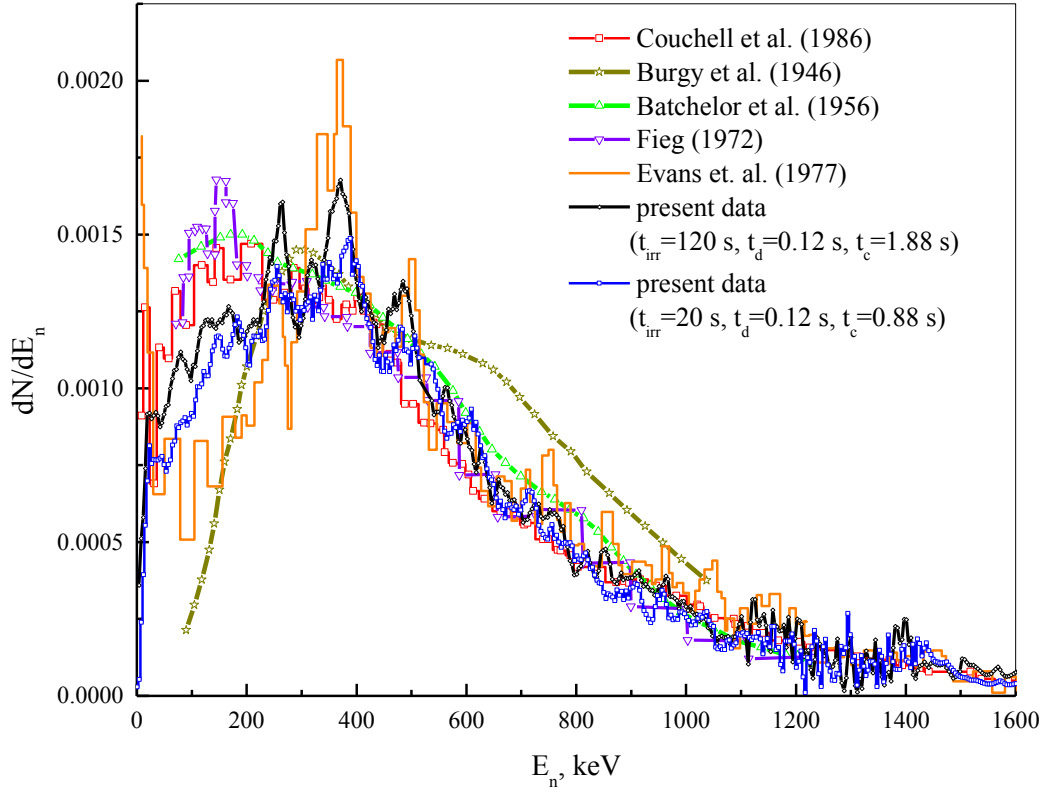


Fig. 17. Equilibrium spectrum of delayed neutrons from fission of ^{235}U by thermal neutrons. The present spectra measured in the range of 0.12-2 s after sample irradiation of 120 s and in the range of 0.12-1 after irradiation of 20 s can be considered as a near equilibrium spectrum. Equilibrium spectra by other authors were calculated with the help of the group spectra obtained by decomposition of appropriate aggregate spectra: Couchell et al.(1986) [10], Burgy et al. (1946) [11], Batchelor et al. (1956) [12], Fieg (1972) [13], Evans et al. (1977) [14].

Comparison shows that our near-equilibrium spectrum has more pronounced peak structure compared to the corresponding experimental data obtained by other authors. The peak structure presented in Evans et al. data [14] is very close to the peak structure of the present data but the overall form of the spectrum is strongly deformed that can be explained by possible errors in the determination of the efficiency of the ^3He -specrometer. Another important feature of the present near-equilibrium spectrum is the lower intensity of delayed neutrons below 200 keV compared to the earlier experiments with the exception of the data of Evans et al. [14]. It is interesting to analyze the equilibrium spectrum data obtained by the summation techniques presented in Fig.18.

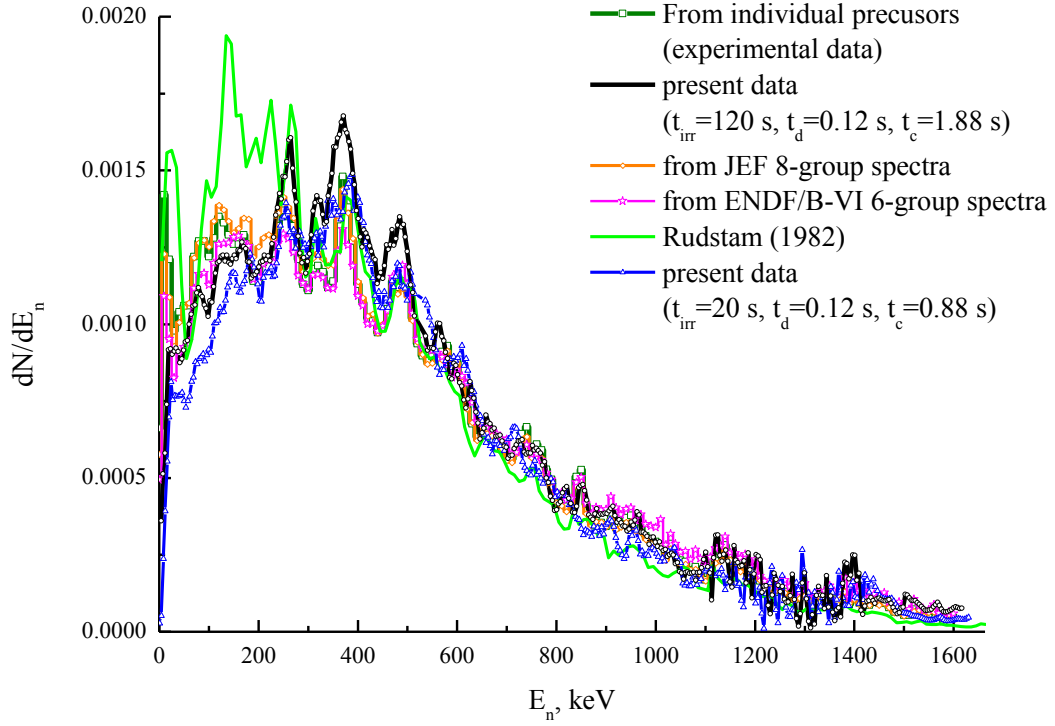


Fig. 18. Equilibrium spectrum of delayed neutrons from fission of ^{235}U by thermal neutrons. The present spectra measured in the range of 0.12-2 s after sample irradiation of 120 s and in the range of 0.12-1 after irradiation of 20 s can be considered as a near equilibrium spectrum. Also shown are the spectra calculated with the help of group spectra from JEF 2.2 and ENDF/B-VI library, and experimental delayed neutron spectra from individual precursors. Rudstam's spectra are based on early measured P_n and $\chi_i(E_n)$ data. Rudstam (1982) [8].

It is seen from Fig.18 that the equilibrium spectrum of delayed neutrons presented in JEF 2.2 and ENDF/B-VI libraries as well as the spectrum based on experimental data for individual precursors are in reasonable agreement with present data, especially in the energy range of 200-1600 keV. However, the calculated spectra do not reproduce the peaks at 350 and 550 keV and have a narrower peak at energy 380 keV. Most likely there are unresolved peaks or group of peaks at energies just above this peak in the present data. Below 200 keV the present spectrum has the same peak structure as the calculated one but the intensity of delayed neutrons in our spectrum in this energy region is lower. It is worth noting that the equilibrium spectrum calculated using only experimental precursor data is closer to our spectrum in the energy region below 200 keV than the spectra calculated with both experimental and theoretical data.

6. Conclusions

Composite delayed energy spectra from epithermal neutron induced fission of ^{235}U have been measured for twelve delay-time intervals extending from 0.12 to 150 s. These data provide a comprehensive set of composite spectra having enhanced sensitivity to neutrons emitted in less than 1 s following fission. These are the first composite delayed neutron measurements that show good overall agreement with appropriate composite spectra obtained by summation calculations based on an up-to-date individual precursor database especially for the relatively long delay-time intervals. The observed agreement can be considered as a proof of the consistency of microscopic and macroscopic approaches in developing a database of delayed neutron energy spectra. Nevertheless the calculated composite spectra have a poorly resolved peak structure in comparison to the present data. This can be attributed partly to the utilization of individual precursor spectra obtained from theoretical models that have no peak structure in the former case.

The near-equilibrium spectrum of delayed neutrons obtained in the present work has definitely, in contrast to earlier experiments, an established peak structure which agrees with results obtained from individual precursor data. However, the present equilibrium spectrum displays less structure and lower intensity of delayed neutrons in the energy range below 200 keV. Besides that, some of the energy peaks are not reproduced at all in the equilibrium spectra obtained from individual precursor data.

It was found that both equilibrium spectrum and composite spectra in different time windows, calculated using only experimental precursor data, are closer to our measured spectra in the energy region below 200 keV than spectra calculated using both experimental and theory-based precursor data.

The composite spectra of delayed neutrons for ^{235}U obtained in the present work can be used as benchmark data for testing the spectral delayed-neutron precursor database. As a by-product of this work, a close similarity was found among composite spectra calculated on the basis of group data and individual precursor data. The unfolding procedure will be applied to the present set of composite spectra with the purpose of improving the 8-group model spectra of delayed neutrons.

Acknowledgments

The authors are grateful to Paraskevi Dimitriou from the Nuclear Data Section (IAEA) for a number of helpful suggestions in preparation of this report.

References

- [1] M.C. Brady, T. R. England., Delayed Neutron Data and Group Parameters for 43 Fissioning Systems, Nucl. Sci. and Eng. **103** (1989) 129-149.
M.C. Brady, Evaluation and Application of Delayed Neutron Precursor Data, PhD Dissertation, Texas A&M University, with research performed at the Los Alamos National Laboratory (Dec. 1988).
- [2] T. Kawano, P. Möller, W. B. Wilson, Phys. Rev C **78**, 054601 (2008) 1-8.
- [3] I. Dillmann, P. Dimitriou, and B. Singh, in: Summary Report of the 1st RCM of the CRP ‘Development of Reference Database for beta-delayed neutron emission, 26-30 August 2013, Vienna, Austria, <http://www-nds.iaea.org/publications/indc/indc-nds-0643/>; Summary Report of the 2nd RCM of the CRP ‘Development of Reference Database for beta-delayed neutron emission, 23-27 March 2015, Vienna, Austria, <http://www-nds.iaea.org/publications/indc/indc-nds-0683/>
- [4] H. Ohm, K.-L. Kratz, S.G. Prussin, The analysis of delayed neutron energy spectra recorded with ³He ionization chambers, Nucl. Instrum. and Methods in Phys. Res. A **256** (1987) 76-90.
- [5] K-L Kratz, G. Hermann, Systematics of neutron emission probabilities from delayed neutron precursors, Z. Physik **263** (1973) 435-442.
- [6] H. Franz, W. Rudolph, H. Ohm, K.-L. Kratz, G. Herrmann, F.M. Nuh, D.R. Slaughter and S.G. Prussin, Nucl. Instr. and Meth. **144** (1977) 253.
- [7] J. Blachot, C. Chung, F. Storrer, JEF-2 delayed neutron yields for 39 fissioning systems, Ann. Nucl. Energy **24** (6) (1997) 489-504.
- [8] G. Rudstam, Six group representation of the energy spectra of delayed neutrons from fission, Nucl. Sci. and Eng. **80**, (1982) 238-255.
- [9] Spriggs G.D., Campbell J.M., Piksaikin V.M., Progress in Nuclear Energy **41**/1-4 (2002) 223-251.
- [10] G.P. Couchell, D.J. Pullen, W.A. Schier, R.S. Tanczyn, L.V. Fisteag, M.H. Haghighi, Q. Sharafuddin, Composite delayed neutron spectra from ²³⁵U, Proc. Int. Conf. Nucl. Data for Basic and Applied Science, Santa Fe, May 1985, vol. p. 707, Gordon and Breach Sci. Publ. (1986); W.A. Schier, Q. Sharafuddin, G.P. Couchell, L.V. Fisteag, M.H. Haghighi, D.J. Pullen, R.S. Tanczyn, Search for energy dependence among composite delayed neutron spectra of ²³⁵U, *ibid.*, vol. 1, p. 751.
- [11] M. Burgy, L.A. Pardue, H.B. Willard, E.O. Wolland, Phys. Rev. **70** (1946) 104.
- [12] R. Batchelor, H.R. Mck. Hyder, J. Nucl. Energy **3** (1956) 7.
- [13] G. Fieg, J. Nucl. Energy **26** (1972) 585.
- [14] A. E. Evans and M.S. Krick, Nucl. Sci. and Eng. **62** (1977) 652.

Nuclear Data Section
International Atomic Energy Agency
P.O. Box 100
A-1400 Vienna
Austria

e-mail: NDS.Contact-Point@iaea.org
fax: (43-1) 26007
telephone: (43-1) 2600-21725
Web: <http://www-nds.iaea.org>