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ABSTRACT

The purpose of this work is to extend the energy range of the data on the relative abundances and the periods of delayed neutrons from fission of ^{235}U beyond the threshold of the ^{235}U(n,n'f) reaction. The measurements of the relative abundances and half-lives of precursors for delayed neutrons emitted from neutron-induced fission of ^{235}U in the energy range 0.4 – 8 MeV have been performed with a modified experimental setup at IPPE, Obninsk. The obtained data have been used to verify the energy dependence of the ^{235}U fission product yields obtained from the GEF code using the summation method and the IAEA microscopic delayed-neutron database. The delayed-neutron relative abundances and half-lives data for neutron-induced fission of ^{235}U in the energy range 0.4 – 8 MeV are available from the IAEA reference database https://www-nds.iaea.org/beta-delayed-neutron/database.html.

July 2023

Contents

1. INTRODUCTION

The energy dependence of delayed neutron (DN) macroscopic data in the energy range above 5 MeV needs to be carefully investigated. The data on the relative abundances and the half-life of delayed neutron precursors from fission of ^{235}U in this energy range are presented only in one paper by Maksyutenko [1]. These data are not consistent with the energy dependence of the average half-life $<T_{1/2}(E_n)>$ of DN precursors which can be estimated with the help of the following expression

$$< T_{1/2}(A, E_n) > = \frac{\sigma_{n,f1} \cdot <T_{1/2}(A,E_n)> + \sigma_{n,n'f} \cdot <T_{1/2}(A-1,E_{n1})>}{\sigma_f(E_n)}, \quad (1)$$

where $\sigma_f(E_n) = \sigma_{n,f1} + \sigma_{n,n'f}$, $\sigma_{n,f1}$, $\sigma_{n,n'f}$, – the fission cross section, the first and the second chance fission cross sections, respectively; A – the atomic number of target nuclide.

Calculations for fission of ^{235}U have been made using fission cross sections from ENDF/B-VII.I and average half-life $<T_{1/2}(A-1)>$ for ^{234}U from the systematics [2]. The results show that the $<T_{1/2}>$ value at the neutron energy 8 MeV is equal to 9.5 s whereas the corresponding $<T_{1/2}>$ value in Maksyutenko's paper is 12.5 s. This means that Maksyutenko's value was overestimated by about 3 s.

The purpose of this work is to extend the energy range of data on the relative abundances and the periods of DN from fission of ^{235}U beyond the threshold of the ^{235}U(n,n'f) reaction. These data are important for reactor kinetic calculations as well as for the verification of the fission yield data [3] and the chance fission analyses [4]. The measurements of the DN relative abundances and the half-lives of their precursors from neutron-induced fission of ^{235}U in the energy range 0.4 – 8 MeV have been made with a modified experimental set-up at IPPE. The data have been used to verify the energy dependence of the fission product yields for ^{235}U obtained from the GEF code [5].

2. EXPERIMENTAL SET-UP

A fully renovated experimental setup was installed at the ion beam line of the electrostatic accelerator at IPPE. The main components of the setup are shown in Fig.1.

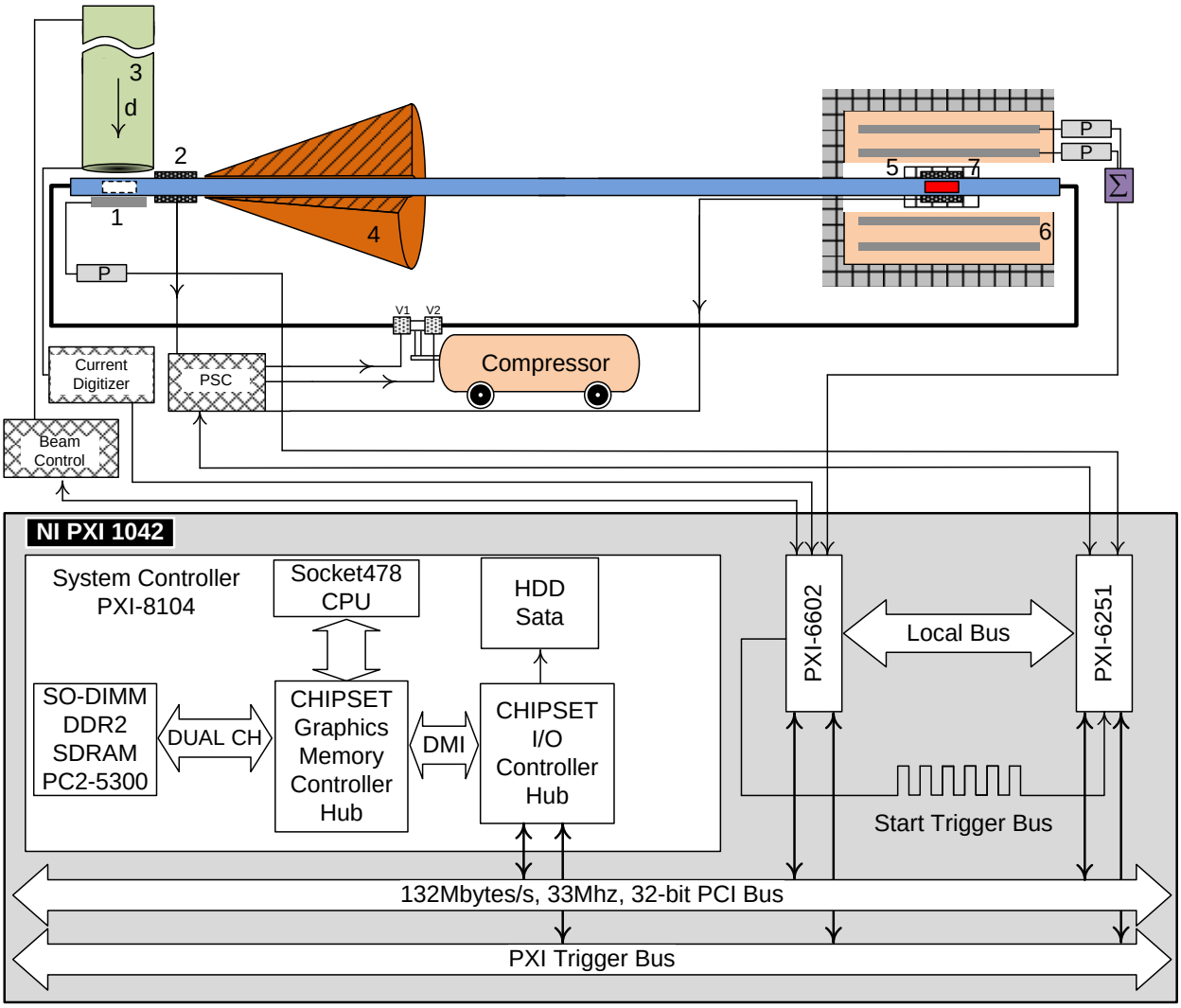

FIG. 1. Experimental set-up for investigation of delayed neutron characteristics.

P – preamplifier, amplifier and discriminator unit, Σ – pulse summation module, V1 and V2 – electromagnetic valves, PSC – pneumatic system control module, 1 – fission chamber, 2, 5 – sample position detectors, 6 – neutron detector, 4 – pneumatic transfer system, 4, 7 – shielding. The electronic system for acquisition, processing, and visual control of the measuring process of accumulating experimental data is based on National Instruments (NI) modules described in further detail below.

A short description of some of the components of the set-up is given in the following.

2.1 4π-neutron detector

A cylindrical polyethylene block with a central channel for installing a pneumatic transport system and Pb shield was used as a moderator for the registered neutrons. Twenty-one proportional ^{3}He counters were placed in the moderating matrix parallel to the central channel along two concentric circles. The main criterion for choosing this configuration of ^{3}He counters in the matrix was the maximum neutron detection efficiency and the minimum variation with energy of the efficiency in the energy range that corresponds to the energy spectrum of delayed neutrons.

The optimization of the placement of ^{3}He counters in the detector moderator was carried out by simulating various configurations with the Monte Carlo method. The variation in the radii of the two concentric rings of the ^{3}He counters R_1 and R_2 showed that when using a moderator in the form of a cylinder with the diameter of 50.5 cm and the generatrix equal to 64 cm, the maximum detector efficiency with the minimum energy dependence is achieved at values R_1 = 10.9 cm and R_2 = 14.9 cm. The efficiency of the neutron detector has a flat energy dependence in the energy range of delayed neutrons and its value in this region is close to 40%. The value of the efficiency weighted by the prompt neutron spectra of ^{252}Cf is $\langle\varepsilon_n\rangle = 34.96 \pm 0.81\%$.

2.2 Data acquisition system

The electronic system used for data acquisition and processing, as well as visual control of the measurement process is based on National Instruments (NI) modules. The system includes a PXI-8104 controller, a PXI-6602 timer/counter, and a PXI-6251 multifunctional module. All modules are installed in a PXI-1042 rack (see Fig. 1) equipped with a PXI/PCI bus, which allows the controller processor and individual modules to be integrated into a single platform. The signals from the neutron detector were fed sequentially to preamplifiers, amplifiers, and discriminators. The registration of the number of pulses coming from the detectors to the data collection and processing system was carried out continuously, including the time of irradiation of the sample and counting the activity of delayed neutrons after the interruption of the ion beam.

2.3 Neutron source

The flux of monoenergetic neutrons was generated by means of the ^{7}Li(p,n)^{7}Be and D(d,n)^{3}He nuclear reactions in the neutron energy range 0.42 – 2.92 MeV and 4 – 8 MeV, respectively. The proton and deuteron beams were provided by the IPPE electrostatic accelerator at the appropriate energy of the ion beam. Lithium targets were prepared by evaporation of LiF onto 0.5-mm copper backings. Deuterium targets were produced by absorption of deuterium on a ~1 mg/cm^2 thick Ti layer deposited on 0.5-mm copper backings. Target thickness was chosen so that the ion energy loss in the target does not exceed the neutron kinematic energy spread.

A spectrometer based on a stilbene crystal was utilized to measure the energy distributions of neutrons emitted from the D(d,n)^{3}He reaction, $d^2N(\theta, E_n)/(d\Omega \cdot dE_n)$, at an angle of 0^0 with respect to the deuteron beam. The deuteron beam had energies ranging from 1 to 5 MeV. The spectrometer was placed in the shield against neutrons scattered in the experimental hall. The

gamma background was suppressed by a filter made of lead sheet. The energy scale of the spectrometer was calibrated using the standard gamma sources, ^{137}Cs and ^{60}Co. A correction for the transmission through the lead filter was calculated using Monte Carlo simulations. The measured spectra emitted at 0^0 from the D(d,n)^{3}He reaction on a D-Ti target with Cu-backing are presented in Fig. 2.

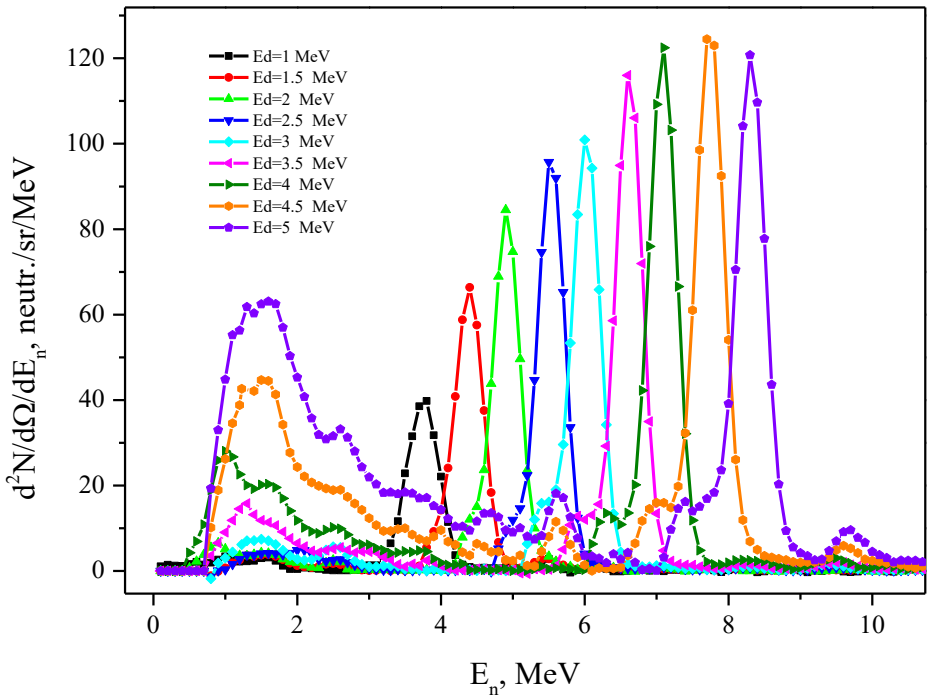

FIG. 2. Neutron energy spectra $d^2N(\theta,E_n)/(d\Omega\cdot dE_n)$ *from the D(d,n)^{3}He reaction on a D-Ti target (Cu-backing) measured at θ = 0^0 relative to the deuteron beam with the stilbene crystal spectrometer (normalized on one incident deuteron).*

It can be seen from Fig. 2 that the low-energy neutron component is observed even at the deuteron energy of 3.5 MeV. At the energy of 4.5 MeV, an additional contribution of neutrons most likely from the break-up reaction Cu(d,pn)Cu becomes comparable with the main neutron component of the reaction D(d,n)^{3}He while, at the energy of 5 MeV, it is twice as high. The integration of the spectra $\int(d^2N(\theta,E_n)/d\Omega\cdot dE_n)dE_n$ over the energy range of monoenergetic neutrons and over the energy range of break-up neutrons allows us to obtain the differential cross-sections of the main component of the D(d,n)^{3}He reaction (monoenergetic) and the break-up reaction Cu(d,pn)Cu (non-monoenergetic), respectively. The obtained data are presented in Fig. 3. The data are compared with Drosg's estimate of the differential cross-section $d\sigma(\theta)/d\Omega$ of the main reaction D(d,n)^{3}He at θ = 0^0 [6].

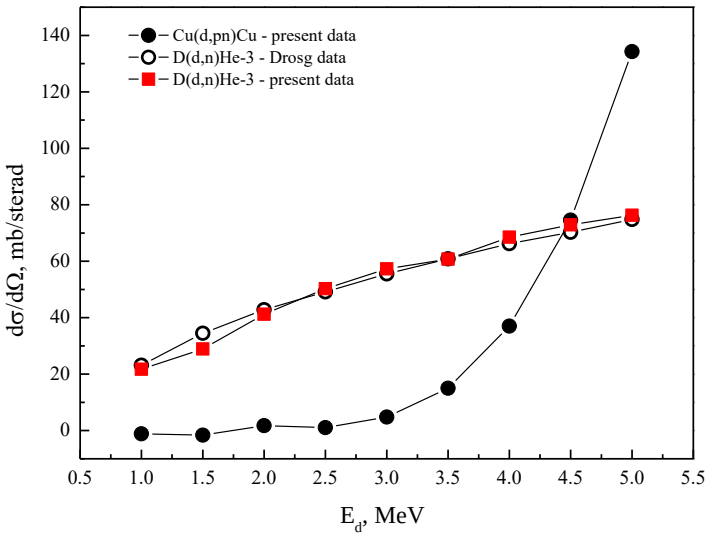

FIG.3. The differential cross-section $d\sigma(\theta)/d\Omega$ *of the main component of the D(d,n)^{3}He reaction (monoenergetic) and the differential cross-section of the break-up reaction Cu(d,pn)Cu (non-monoenergetic) at $\theta = 0^0$. The differential cross-section data of the main reaction at $\theta = 0^0$ from Drosg's estimation [6] are plotted for comparison.*

The non-monoenergetic component of the neutron spectrum was considered as a background and was accounted for in the processing of the experimental decay curves.

3. THE MEASUREMENT PROCEDURE AND PROCESSING OF THE EXPERIMENTAL DATA

The experimental method employed in these measurements is based on cyclic irradiations of the fissionable samples followed by the measurement of the time dependence of the delayed neutron activity. During irradiation time the current of the ion incident on the neutron target, the pulse spectra from the ^{239}Pu fission chamber, and the counts from the neutron detector are registered. The neutron detector stability was checked every day by counting the Pu-Be source in a standard geometry. The irradiation time was 180 and 15 s. The DN counting time was 600 s for long irradiation and 500 s for short irradiation experiments. The sample delivery time in this experiment was about 200 ms.

The collected DN decay curve data were summed up over all cycles and transformed into the time scale with a channel width of 0.01 s. Some of the decay curves measured with the D(d,n)^{3}He reaction are shown in Fig. 4.

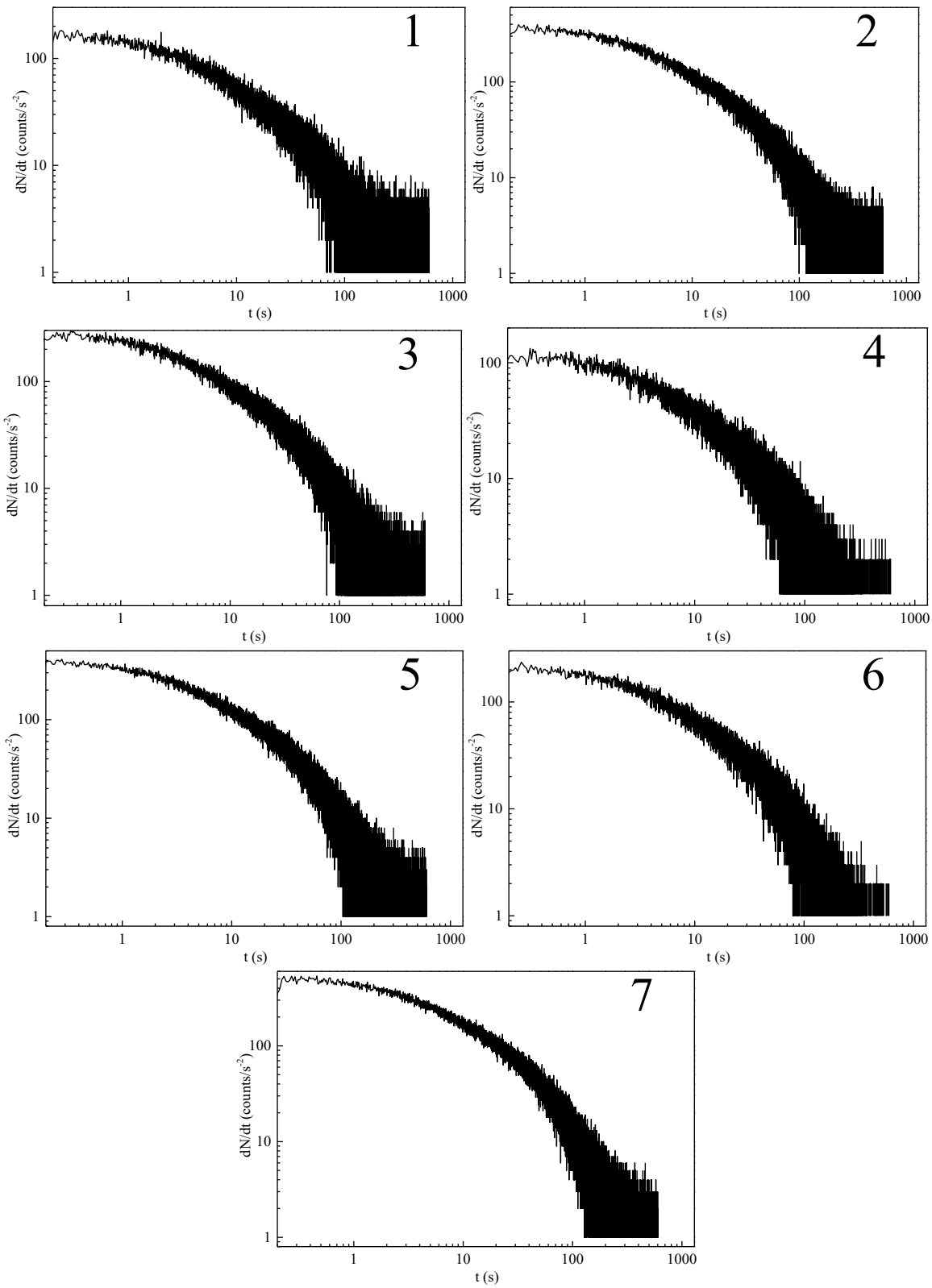

FIG.4. Decay curves of delayed neutrons from fission of ^{235}U by neutrons in the energy range 4.15-8.25 MeV in uniform scale with the time channel width of 0.01 s. 1 – energy of incident deuterons E_d=1 MeV, neutron energy E_n = 4.15 MeV, 82 cycles of irradiation; 2 – E_d = 2 MeV, E_n = 5.25 MeV, 74 cycles; 3 – E_d = 3 MeV, E_n = 6.27 MeV, 35 cycles; 4 – E_d = 3.5 MeV, E_n = 6,77 MeV, 9 cycles; 5 - E_d = 4MeV, E_n = 7.27 MeV, 22 cycles; 6 – E_d = 4.5 MeV, E_n = 7.76 MeV, 5 cycles; 7 – E_d =5 MeV, E_n = 8.25 MeV, 12 cycles.

For the analysis of the DN decay curves to estimate the relative abundances and periods of delayed neutrons we used iterative least-squares method (LSM) described in [7]. The model equation for cyclic irradiation used in the LSM is given by the following expression

$$N(t_k) = A \cdot \sum_{i=1}^{m} T_i \cdot \frac{a_i}{\lambda_i} \cdot (1 - e^{-\lambda_i \cdot \Delta t_k}) \cdot e^{-\lambda_i \cdot t_k} + B \cdot \Delta t_k \,, \qquad (2)$$

$$T_i = (1 - e^{-\lambda_i \cdot t_{irr}}) \cdot \left(\frac{n}{1 - e^{-\lambda_i \cdot T}} - e^{-\lambda_i \cdot T} \cdot \left(\frac{1 - e^{-n \cdot \lambda_i \cdot T}}{((1 - e^{-\lambda_i \cdot T})^2} \right) \right), \; A = \varepsilon_n \, \sigma_f \, \varphi \, N_f \, \nu_d \,,$$

where $N(t_k)$ – the number of counts registered by the neutron detector in the time-channel t_k with time-channel width Δt_k, ν_d – the total delayed neutron yield per one fission, B – the intensity of the neutron background, λ_i and a_i – the decay constant and the relative abundance of i-th group of DN, n – the number of cycles, m – the number of delayed neutron groups, T – the duration of one cycle of measurements, which includes the irradiation and the delayed neutron counting time, t_{ir} - irradiation time, ε_n – the efficiency of the neutron detector, φ - the neutron flux, σ_f – the fission cross section, N_f – the number of atoms in the sample under investigation.

The correction for the break-up neutrons was introduced using the following equation

$$N(t_k) = N_{dn}(t_k) + N_{dpn}(t_k) \quad ,$$

where $N_{dn}(t_k)$ – the decay curve related to incident neutrons from the main reaction, $N_{dpn}(t_k)$ – the decay curve generated by break-up neutrons. The values $N_{dpn}(t_k)$ were estimated with the help of Eq. (2) using the average energy from the N_{dpn} spectra (see Fig.2) and the corresponding DN parameters (a_i, T_i) measured at this neutron energy with the $^7Li(p,n)^7Be$ reaction.

The DN decay data with the uniform scale were transformed into a time scale suitable for estimating DN parameters with the least-squares method (LSM): 0.02 s – 200 channels; 0.04 s – 200 channels; 0.1 s – 200 channels; 1 s – 200 channels; 10 s – 37 channels. The results of the estimation of the relative abundances and periods of DN by means of the LSM are presented in Fig. 5 where the black points are experimental data in an uneven time scale and the smooth red line is the result of the LSM estimation.

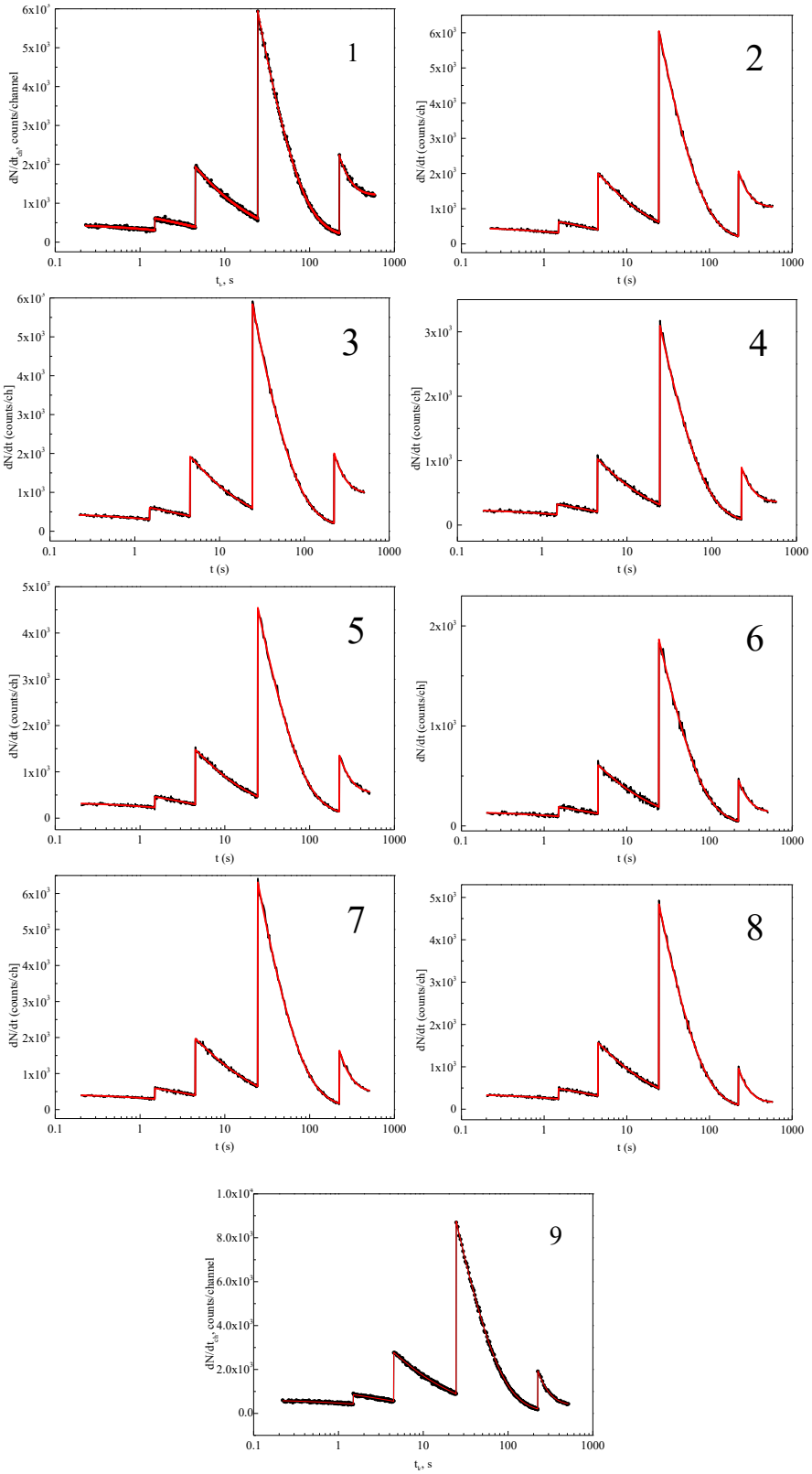

FIG. 5. Decay curves of delayed neutrons from fission of ^{235}U by neutrons in an uneven time scale. 1 - energy of incident deuterons Ed = 1 MeV, energy of neutrons generated in the target En = 4.15 MeV, 99 cycles of irradiation; 2 – Ed = 1.5 MeV, En = 4.70 MeV, 84 cycles; 3 - Ed = 2 MeV, En = 5.25 MeV, 74 cycles; 4 – Ed = 2.5 MeV, En = 5.76 MeV, 27 cycles; 5 – Ed = 3 MeV, En = 6.27 MeV, 35 cycles; 6 – Ed = 3.5 MeV, En = 6.77 MeV, 9 cycles; 7 – Ed = 4 MeV, En = 7.26 MeV, 22 cycles; 8 – Ed = 4.5 MeV, En = 7.76 MeV, 7 cycles, 9 – Ed = 5 MeV, En = 8.25 MeV, 11 cycles. Black curves are experimental data. Smooth red curves are the results of the LSM.

4. RESULTS AND DISCUSSION

The data on the energy dependence of the relative abundances of DN and the half-lives of their precursors for neutron-induced fission of ^{235}U are presented in the 6-group model. The data that were obtained from several runs at a given energy of primary neutrons were averaged considering the correlation matrices calculated at each run [8]. The present DN temporary data are compared with the corresponding data measured by other authors in Fig. 6 by means of the average half-life of delayed neutron precursors which is obtained from the expression $< T_{1/2} >= \sum_{i=1}^{6} T_i \cdot a_i$.

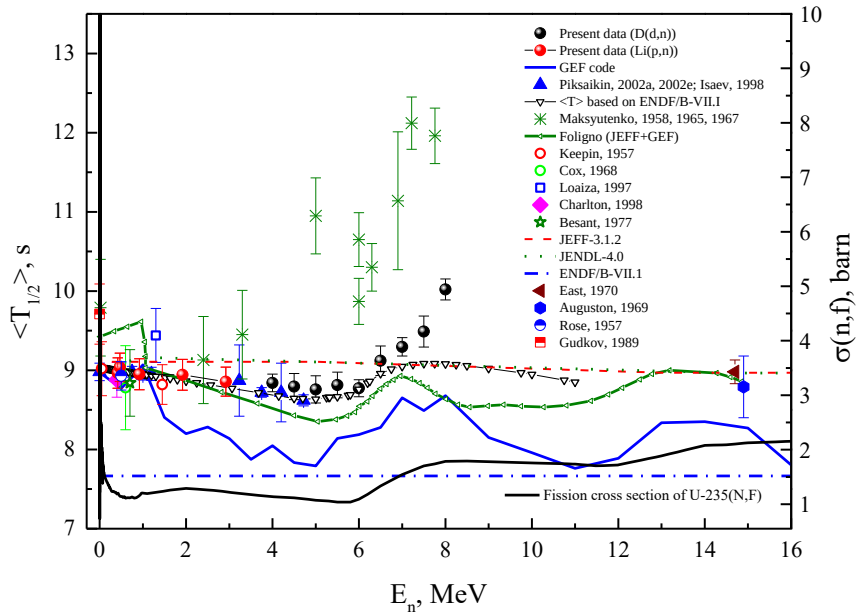

FIG. 6. The energy dependence of the average half-life of DN precursors from neutron-induced fission of ^{235}U. References can be found in [9]. The data by Foligno (JEFF+GEF) are discussed in [10].

In Fig. 6, in addition to the experimental data, we also compare summation calculations of $<T_{1/2}>$ obtained from the following formula:

$$< T_{1/2}(E_n) >= \sum_i P_{ni} \cdot CY_i(E_n) \cdot T_{1/2,i} \,/\, \sum_i P_{ni} \cdot CY_i(E_n) \qquad (3)$$

where $CY_i(E_n)$ – the cumulative yield of i-th DN precursor, P_{ni} and $T_{1/2,i}$ – the DN emission probability and the half-life of the i-th precursor, respectively. The cumulative yields of delayed neutron precursors $CY_i(E_n)$ were obtained from the GEF code [6]. The summation was made overall all precursors which are included in the new microscopic beta-delayed neutron database generated by the IAEA CRP [11].

As can be seen in Fig. 6, the present results on the energy dependence of $<T_{1/2}(E_n)>$ agree within the uncertainties with the energy dependence observed in earlier studies in the energy range from thermal to 5 MeV. As for the disagreement with Maksyutenko's data that was discussed in the introduction, the most probable reason is that those data were affected by the

"blocking" effect [12] in the neutron detector that was placed in the vicinity of the neutron source. At the time, in the absence of a rabbit system for the transportation of the sample, the neutron detector had to be placed as close as possible to the neutron source.

The most important features of the present data are the smooth decrease of $<T_{1/2}(E_n)>$ in the energy range from thermal to about 6 MeV and a more steeper increase in the energy range above the threshold of the second-chance fission cross section $\sigma_{n,n'}$. The $<T_{1/2}(E_n)>$ data obtained with the fission yields $CY_i(E_n)$ from the GEF code agree with the present data in the energy range from thermal to 1 MeV. It should be noted however, that in this energy range there are many experimental data on fission fragment yields that provide a good basis for a proper choice of model parameters in the GEF code. In the region above 1 MeV, the GEF-based $<T_{1/2}(E_n)>$ data reproduce the general trend observed in the present data, but they systematically underestimate them ($\Delta<T_{1/2}> \approx 1$ s). The most probable reason for this discrepancy is an underestimation of the cumulative yields in the energy range above 1 MeV by the GEF code considering that the microscopic DN data (P_{ni}, $T_{1/2,i}$) used for the calculation of $<T_{1/2}(E_n)>$ are the same in the whole energy range. The rather low GEF fission product yields could be due to the inadequate estimation of the most probable charge in the isobaric beta – decay chains.

One can also estimate the energy dependence of $<T_{1/2}(E_n)>$ on the basis of the chance structure of the fission cross section of ^{235}U with the help of Eq. (1) from the introduction. The fission cross sections of the first and second chance fission were taken from ENDF/B-VII.I. The first chance fission cross sections $\sigma_{n,f1}(E_n)$ (n+^{235}U) were combined with the present data $<T_{1/2}(E_n)>$. The values of $<T_{1/2}(E_n)>$ combined with the second chance fission $\sigma_{n,n'}(E_n)$ (n+^{234}U) were taken from the systematics [2]. It can be seen in Fig. 5 that the resulting $<T_{1/2}(E_n)>$ changes slope at the same energy value where the present measured $<T_{1/2}(E_n)>$ changes its slope (from monotonic decrease to monotonic increase).

Numerical data on the measured delayed-neutron relative abundances and half-livesfor neutron-induced fission of ^{235}U in the energy range 0.4 - 8 MeV are included in the IAEA reference database (http://www-nds.iaea.org/beta-delayed-neutron/database.html).

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References:

[1] B.P. Maksyutenko, et al., Relative yields of delayed neutrons in fission of ^{238}U, ^{235}U and ^{232}Th by fast neutrons, J. Exptl. Theor. Phys. (USSR) **35** (1958) 815.

[2] V.M. Piksaikin, S.G. Isaev, A.A. Goverdovski, Characteristics of Delayed Neutrons: Systematics and Correlation Properties. Prog. Nucl. Energy **41**, Issues 1–4 (2002) 361-384.

[3] D.E. Gremyachkin, V.M. Piksaikin, K.V. Mitrofanov, A.S. Egorov, Verification of the evaluated fission product yields data from the neutron induced fission of ^{235}U, ^{238}U and ^{239}Pu based on the delayed neutron characteristics, Prog. Nucl. Energy **83** (2015) 13-25.

[4] D.E. Gremyachkin, V.M. Piksaikin, A.S. Egorov, V.F. Mitrofanov, K.V. Mitrofanov, Investigation of the chance structure of the fission cross section of ^{238}U by neutrons in the energy range from 14 to 18 MeV using the delayed neutrons, Prog. Nucl. Energy **118** (2020) 103068.

[5] GEF – A General description of the Fission process. URL: https://www.lp2ib.in2p3.fr/nucleaire/nex/gef/

[6] DROSG-2000: Neutron Source Reactions. Data files with Computer codes for 60 accelerator-based neutron source reactions prepared by M. Drosg, Faculty of Physics, University of Vienna, Austria, Version 12.01 (March 2017). URL: https://www-nds.iaea.org/public/libraries/drosg2000/.

[7] V.M. Piksaikin, L.E. Kazakov, S.G. Isaev, M.Z. Tarasko, V.A. Roshchenko, R.G. Tertytchnyi, G.D. Spriggs, J.M.Campbell, Energy dependence of relative abundances and periods of delayed neutrons from neutron-induced fission of ^{235}U, ^{238}U, ^{239}Pu in 6- and 8-group model representation, Prog. Nucl. Energy Vol. **41,** Issues 1-4 (2002) 203-222.

[8] V.M. Piksaikin, A.S. Egorov, D.E. Gremyachkin, K.V. Mitrofanov, V.F. Mitrofanov, Generation of correlation and covariance matrices for the recommended temporal DN parameters in 6- and 8-group models, EPJ Nuclear Sci. Technol. **6** (2020) 54. https://doi.org/10.1051/epjn/2020013.

[9] G.D. Spiggs, J.M. Campbell, A Summary of measured delayed neutron group parameters, Prog. Nucl. Energy **41**, Issues 1-4 (2002) 145-201.

[10] D. Foligno, P. Leconte, Energy Dependence of Delayed Neutron Data, EPJ Web of Conferences 211, 01002 (2019), WONDER-2018, https://doi.org/10.1051/epjconf/201921101002.

[11] P. Dimitriou, I. Dillmann, B. Singh, et al., Development of a Reference Database for Beta-Delayed Neutron Emission, Nucl. Data Sheets **173** (2021) 144–238. Reference Database for Beta-Delayed Neutron Emission: https://www-nds.iaea.org/beta-delayed-neutron/database.html

[12] V.M. Piksaikin, V.A. Roshchenko, G.G. Korolev, Relative yield of delayed neutrons and half-life of their precursor nuclei from ^{238}U fission by 14.2–17.9 MeV neutrons, At. Energy **102** (2007) 151-159 .

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