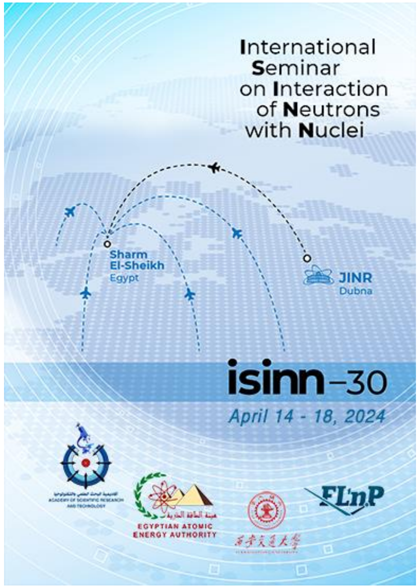
The measurement of the ^{6}Li(n, t)^{4}He reaction cross-section in the energy range of 4.25–7.50 MeV



P.S. Prusachenko[1], T.L. Bobrovskiy[2]*

1 — Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research
2 — Leypunsky named Institute for Physics and Power Engineering

*email: prusachenko@jinr.ru

Outline

- Motivation
- Status of cross-section data on $^{6}Li(n, t)^{4}He$ reaction
- Current problems
- Experimental method and setup
- Data analysis
- Results
- Conclusions

Motivation

The $^{6}Li(n,t)^{4}He$ reaction is of great interest for:

- Thermonuclear industry as a tritium breeder
- Monitoring neutron flux in various nuclear physics experiment (nTOF, LANL etc)
- Neutron shielding

Some open problem:

- The existing experimental datasets on the $^{6}Li(n,t)^{4}He$ reaction are quite limited and contradictory for an energy range above 4.0 MeV
- There is significant difference (20-50%) in evaluated cross-sections
- New experimental data are extremely contradictory

Methodical features

- The Frish gridded ionization chambers are most often used for $^{6}Li(n,t)^{4}He$ reaction cross-section measurements for the neutron energies less than 4 MeV
- It’s difficult to use the ionization chambers at higher energies due to the high Q-value of the reaction
- The scintillation method is usually used in this energy range

The previous experiments

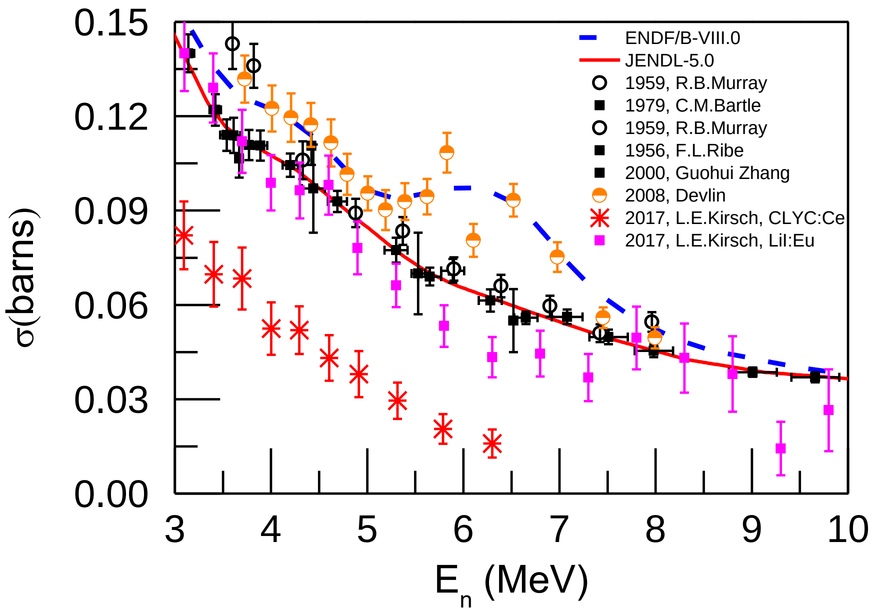


Fig.1. Experimental data and theoretical evaluations on cross-section of $^{6}Li(n,t)^{4}He$ reaction above 3 MeV

- Some new experiments were performed
- M. Devlin's experiment was performed using a set of silicon semiconductor detectors. There was a resonant structure contradictory to all previous experiments. There was no detailed description of experimental procedure, the set of angles was limited.
- Leo E. Kirsch et all performed new measurements relative to the ^{252}Cf prompt fission neutron spectrum. Two different scintillators – LiI:Eu and Cs_2LiYCl_6:Ce – were used. There were a great difference between the results obtained using different scintillators.

1. M. Devlin et al, Differential Cross Section Measurements for the $^{6}Li(n,t)^{4}He$ Reaction in the Few MeV Region, AIP Conf. Proc. 1090, 215–219 (2009); https://doi.org/10.1063/1.3087015

2. Leo E. Kirsch et al, A new measurement of the Li(n,α)t cross section at MeV energies using a ^{252}Cf fission chamber and Li scintillators, Nucl. Instrum. Methods Phys. Res. A 874, 57-65 (2017); https://doi.org/10.1016/j.nima.2017.08.046

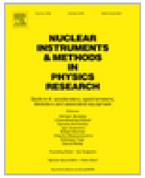
The Leo E. Kirsch's experiment

- There is a good description of experiment
- ^{252}Cf as a neutron source, start from fission fragments
- $LiI{:}Eu$ and $Cs_2LiYCl_6{:}Ce$ (CLYC) scintillators
- Short flight path in different experimental runs – 65 and 125 cm
- Digital acquisition system based on waveform digitizer (500 MS per second and 14 bit ADC resolution)
- Digital signal processing – **constant fraction discriminator for timestamps determination**, pulse shape discrimination for CLYC
- **The time scale was calibrated using the position of prompt γ-rays peak**

Our analysis of the Leo E. Kirsch's experiment

ELSEVIER

Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment



Volume 1056, November 2023, 168582

Full Length Article

Features of using Cs_2LiYCl_6:Ce based scintillation detector for time-of-flight application

P.S. Prusachenko, T.L. Bobrovskiy

Show more

+ Add to Mendeley Share Cite

https://doi.org/10.1016/j.nima.2023.168582

Our analysis of the Leo E. Kirsch's experiment

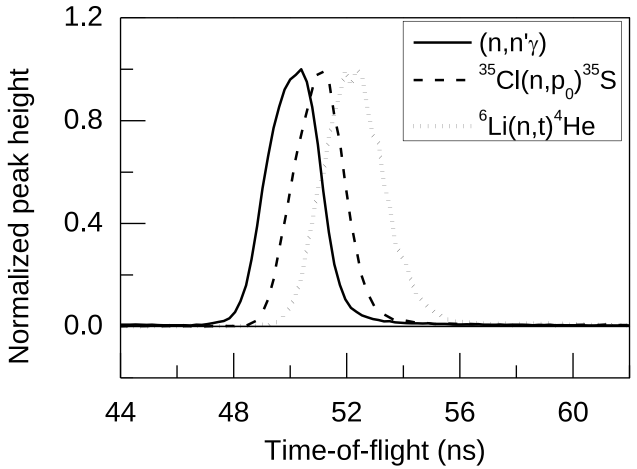


Fig. 2. Normalized TOF spectra corresponding to different types of particles. TOF scale calibrated by prompt γ-rays peak.

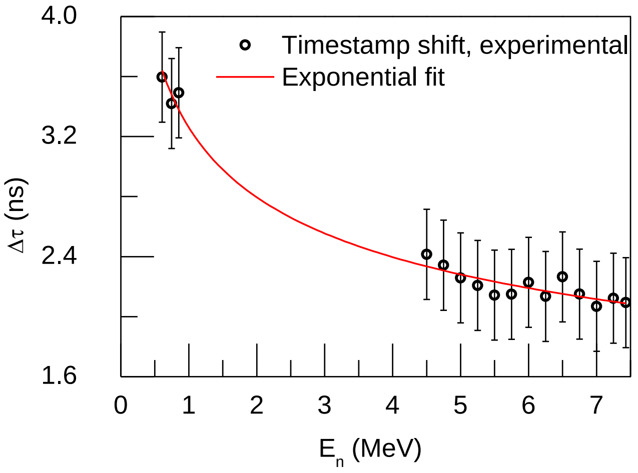


Fig. 3. Timestamp shift for CLYC detector

- There are a significant difference in the rise times for the signals corresponding to the different types of particles (γ-rays, protons, α-particles)
- Systematic shift of timestamps between "neutron" and "gamma" events occurs when the constant fraction algorithm is used
- The position of $^{6}Li(n,t)^{4}He$ peak is shifted when the γ-rays peak uses for calibration

Before and after correction

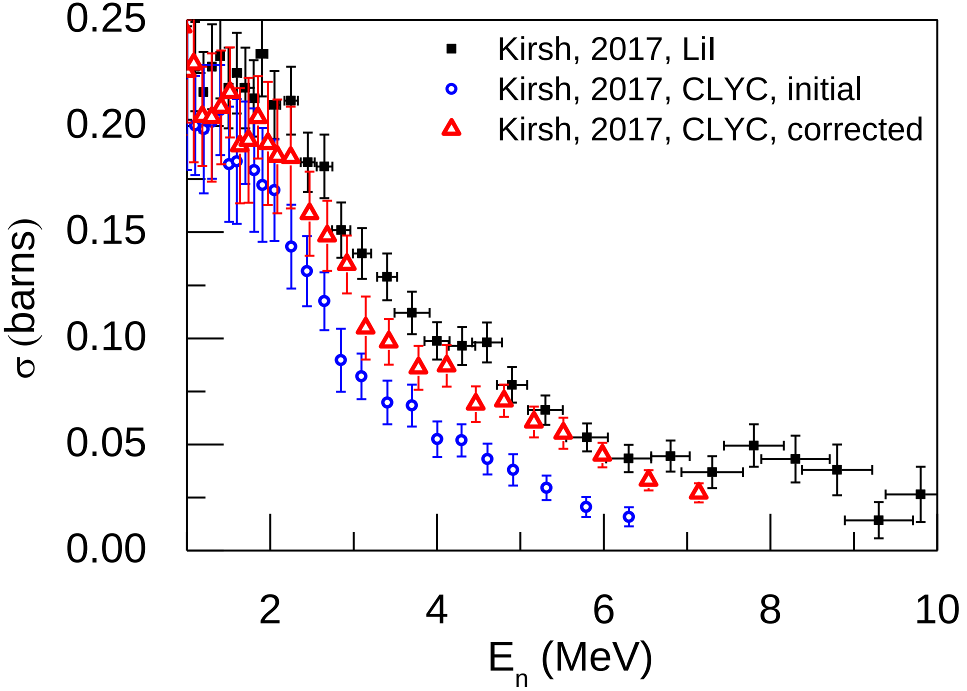


Fig. 4. The experimental data on the $^{6}Li(n,t)^{4}He$ reaction cross-section from the Leo Kirsch's work obtained by the CLYC-based detector before and after correction

- The timestamp determination error – the neutron energy determination error
- The neutron energy determination error – the error in the neutron yield choice
- ^{252}Cf spectrum has sharp dependence of neutron yield on energy – the large error
- The discrepancy observed in the Leo E. Kirsch's paper mostly vanishes after our correction

Open questions

- The exact value of constant fraction in Leo E. Kirsch’s work is unknown
- The timestamp shift for LiI:Eu?
- The γ-background correction increases the uncertainty
- There are contradictions between Leo E. Kirsch data (LiI:Eu), M. Devlin data and other results

The new experiments is needed!

The aim of the work was to obtain the new experimental data on $^{6}Li(n,t)^{4}He$ reaction cross-section taking into account the existing methodical problem

Our new measurements

Eur. Phys. J. A (2024) 60:12
https://doi.org/10.1140/epja/s10050-024-01236-3

THE EUROPEAN PHYSICAL JOURNAL A

Check for updates

Regular Article - Experimental Physics

The measurement of the ^{6}Li(n, t)^{4}He reaction cross-section in the energy range of 4.25–7.50 MeV

P. S. Prusachenko[a], **T. L. Bobrovskiy**

Institute for Physics and Power Engineering, Experimental Nuclear Physics Department, Bondarenko Sq. 1, Obninsk 249033, Russian Federation

Received: 24 October 2023 / Accepted: 26 December 2023

Communicated by Aurora Tumino.

Experimental method

- The CLYC crystal was used
- γ-background was rejected by the pulse shape discrimination
- Quasi-monoenergetic neutron source in combination with time-of-flight method to avoid timestamp shift effect and background neutrons
- ^{235}U fission cross-section for data normalization

Experimental setup

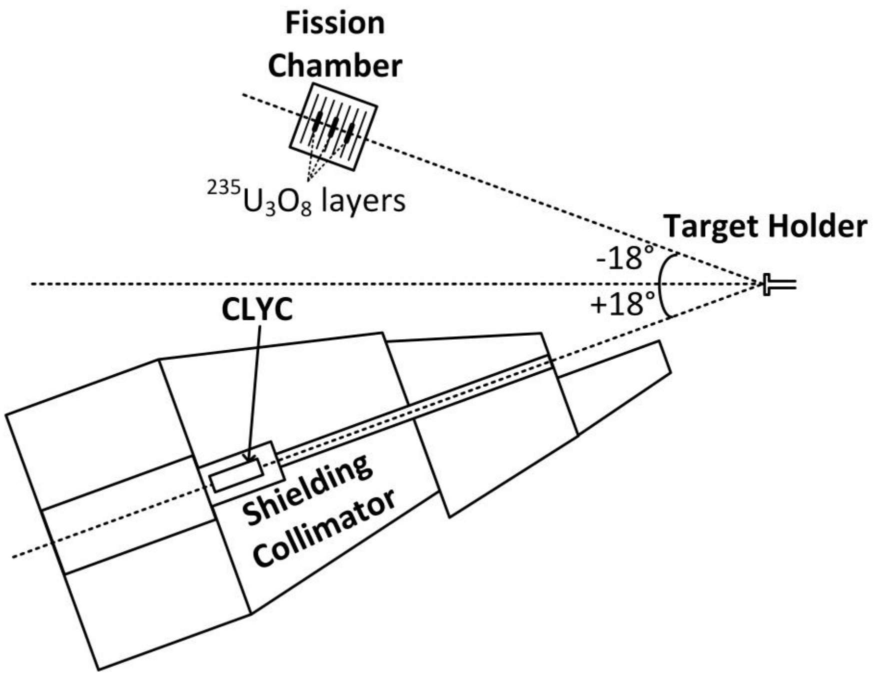


Fig. 5. Layout (not to scale) of experimental setup for the measurement of the $^{6}Li(n,t)^{4}He$ reaction cross-section

Geometry and accelerator:

- An axially symmetrical geometry of detectors
- 3MV Tandem accelerator
- Target — TiD_2 (~1 mg/cm^2) on copper backing
- Neutron energy range from 4.25 to 7.40
- Pulsed deuteron beam

CLYC:

- Crystal size of 38x20 mm
- ^{6}Li enrichment of 95%
- Flight path 186.3±0.5 cm
- Cadmium case (~0.5 mm)
- Inside the shielding collimator

Fission chamber:

- 4 double side layers of $^{235}U_3O_8$
- Total number of ^{235}U atoms is $56 \pm 1 \cdot 10^{18}$
- Efficiency is 0.91±0.01
- Flight path 46.5±0.5 cm
- Cadmium case (~0.5 mm)

Acquisition system:

- Waveform digitizer
- Sampling rate of 500 MS/s, ADC resolution of 14 bit

Digital signal processing

- Timestamps – the constant fraction algorithm emulation. The optimal constant fraction values were 20% for CLYC and 30% for fission chamber and pick-up electrode

- Pulse integrals – 3000 ns and 80 ns from the pulse start for CLYC and fission chamber respectively

- Pulse shape discrimination for CLYC – the comparison of pulse integrals in different windows – 50 and 3000 ns from pulse start

Analysis of CLYC spectra

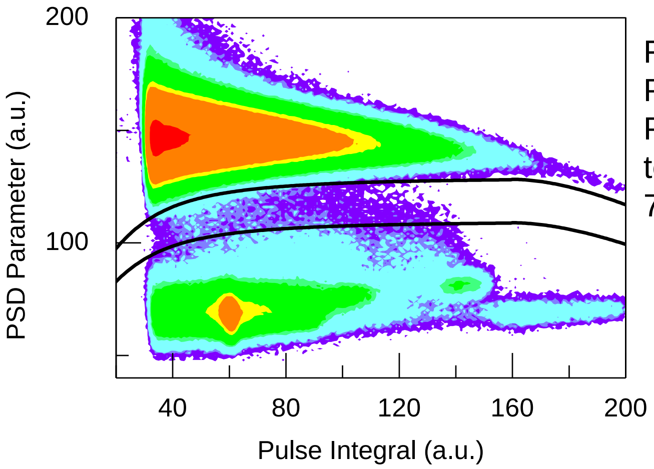


Fig. 6. 2-d spectrum, Pulse Integral – PSD Parameter corresponding to the neutron energy of 7.0 MeV

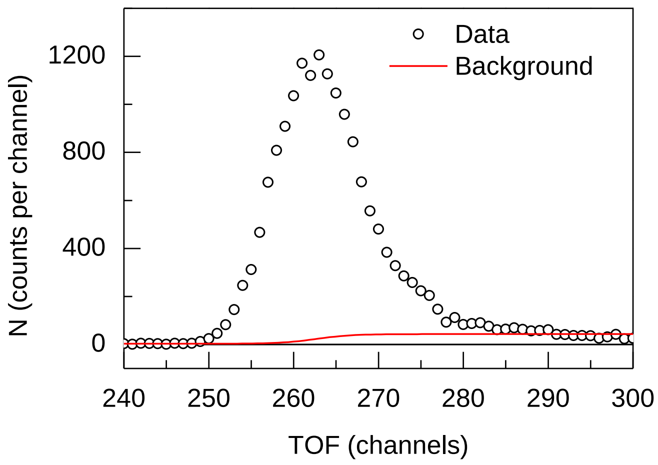


Fig. 8. 1-d TOF spectrum corresponding to $^{6}Li(n,t)^{4}He$ events

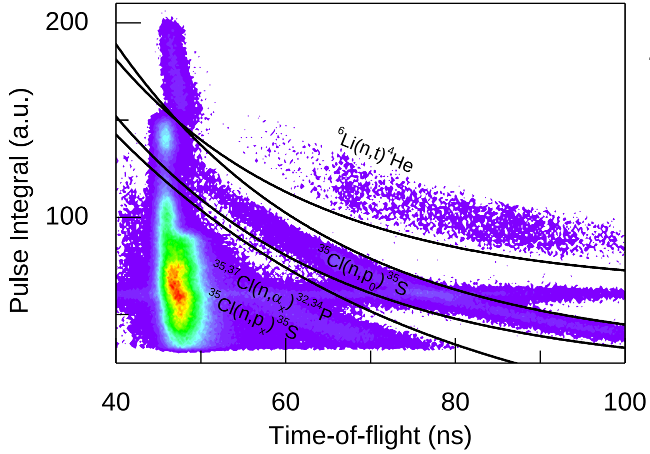


Fig. 7. 2-d spectrum, TOF -Pulse Integral, corresponding to the neutron energy of 7.0 MeV

- Rejection of the γ-rays background – analysis of the “Pulse Integral – PSD” spectra
- Separation of the events corresponding to $^{6}Li(n,t)^{4}He$ reaction – “TOF – Pulse Integral” spectra analysis
- The one-dimensional TOF spectra corresponding to $^{6}Li(n,t)^{4}He$ reaction were built
- The areas of $^{6}Li(n,t)^{4}He$ peaks were obtained after removing the background substrate

Analysis of spectra from fission chamber

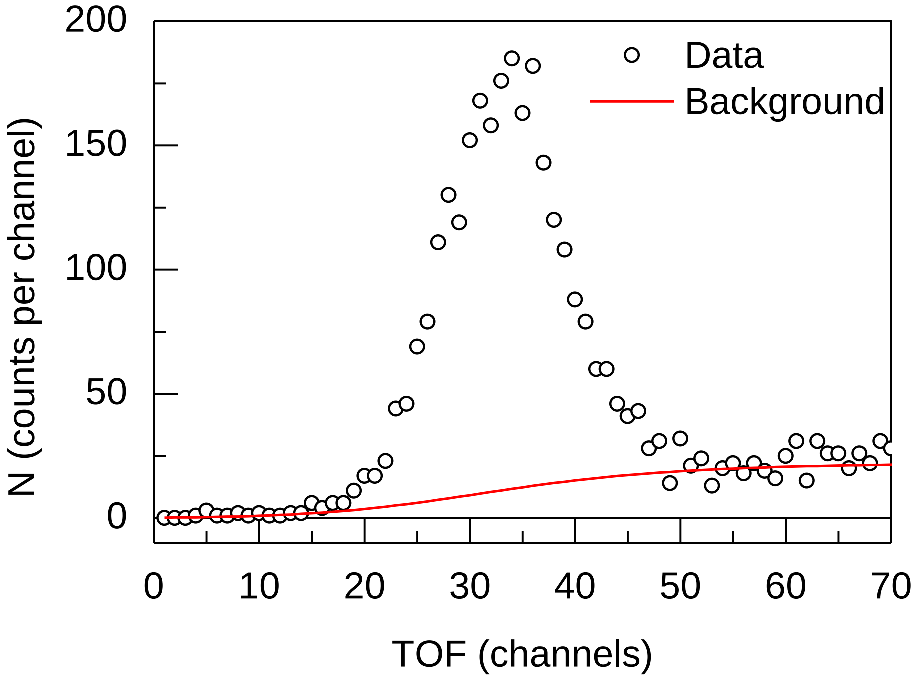


Fig. 9. 1-d TOF spectrum corresponding to the fission chamber events after α-particles rejection

- Rejection of α-particles
- Building the TOF spectra for each value of neutron energy
- Determination of peak areas after background removing

Cross-section determination

$$\sigma(E) = \frac{N_{clyc}\alpha_{clyc}\sigma_{ff}(E)n_u\varepsilon_{ff}}{N_{ff}\alpha_{ff}n_{clyc}\beta}\frac{R_{chamb}^2}{R_{clyc}^2}$$

- N_{clyc} and N_{ff} – area of peaks corresponded to ^{6}Li(n, t)^{4}He reaction and monitor chamber respectively
- α_{clyc} and α_{ff} – multiple neutron scattering correction for CLYC and monitor chamber
- n_u and n_{clyc} – number of ^{235}U and ^{6}Li atoms
- σ(ff) – ^{235}U fission cross-section
- ε(ff) – efficiency of fission fragments detection
- β – wall effect correction factor for CLYC detector
- R_{chamb} and R_{clyc} – the flight paths to the monitor chamber and CLYC detector

Corrections

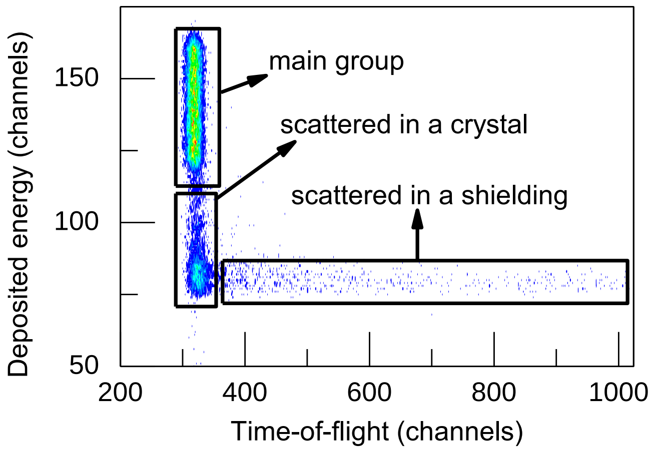


Fig. 10. The example of simulated 2-d spectrum, "TOF - Dep. Energy“, for the CLYC crystal.

- The effect of multiple neutron scattering was evaluated using GEANT4 framework
- Two different simulations for each incident neutron energy – taking into account the full geometry of experimental setup (opt. 1) and without one (opt. 2).
- The correction factor for CLYC was no more 0.995
- The correction factor for FC after background removing was no more 0.99 in the same conditions
- The influence of wall effect in CLYC was negligible

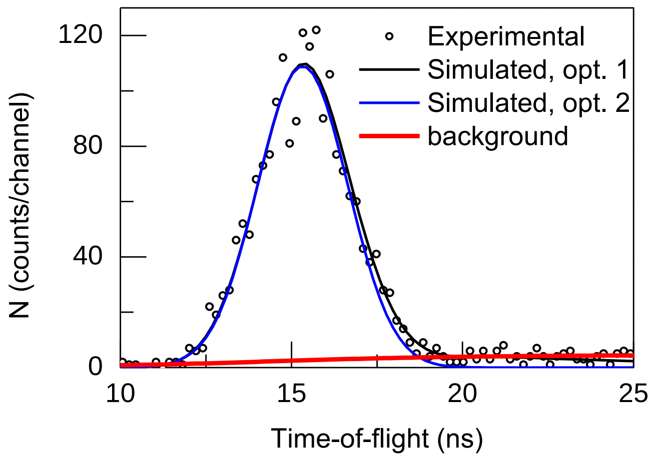


Fig. 11. The example of simulated 1-d TOF spectrum vs experimental one for fission chamber, E_n = 4.75 MeV

Targets characterization

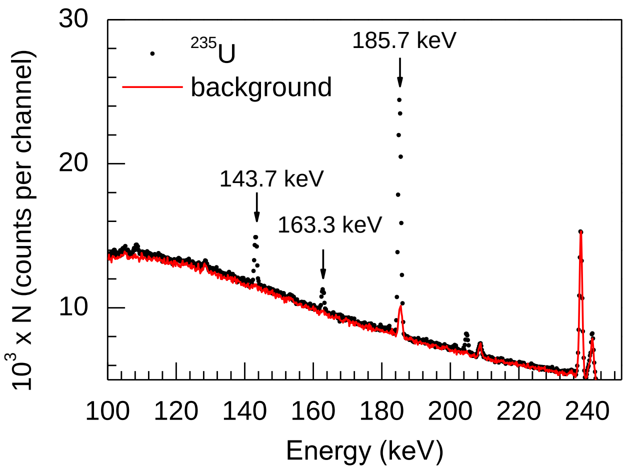


Fig. 12. Measured γ-ray spectra from radioactive decay of ^{235}U vs background.

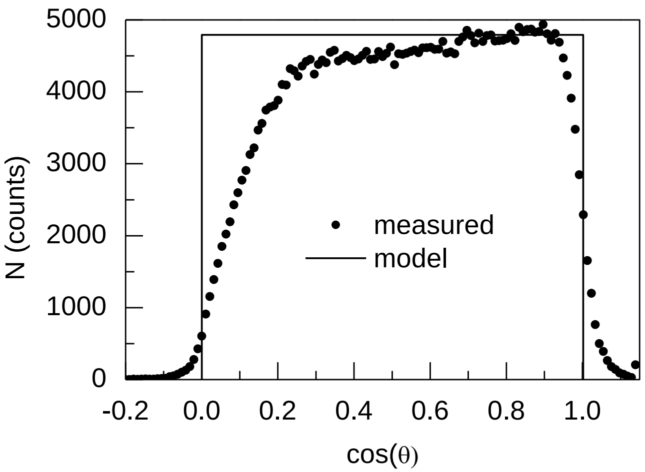


Fig. 13. Measured angular distribution of fission fragments from fission of ^{235}U by thermal neutrons vs model

- The number of ^{6}Li atoms in the CLYC was calculated based on the scintillator stoichiometry, enrichment and the crystal size given in its specification
- To verify this value the additional measurements in the neutron energy range 0.5 – 0.9 MeV were performed
- The number of ^{235}U atoms was obtained by measuring the γ-rays from the radioactive decay of ^{235}U
- The efficiency of the fission fragments detection ε_{ff} was obtained according to the method proposed by C. Budtz-Jorgensen[1]

1. C. Budtz-Jorgensen et al, Assaying of targets for nuclear measurements with a gridded ionization chamber, Nucl. Instrum. Methods Phys. Res., Sect. A **236**, 630 (1985); URL: https://doi.org/10.1016/0168-9002(85)90972-6.

Results

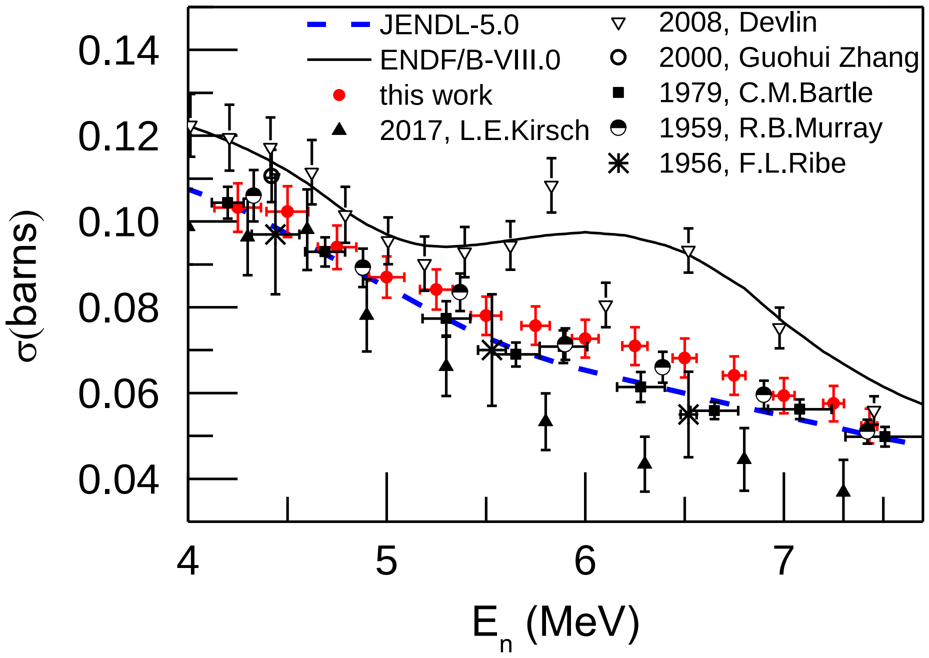


Fig. 14. The experimental data obtained in this work (red dots) compared with the other experimental results and the cross-section evaluations

Uncertainty source	Contribution (%)
Statistical (1σ)	2.0 - 3.7
Background subtraction	1.0 - 3.5
Number of ^{235}U atoms	2.2
Fission chamber efficiency	1.1
Solid angle	1.8
Number of ^{6}Li atoms	1.6
Multiple scattering correction	2.1
Detectors angle	1.3 – 3.8

- The data from this work are in agreement within uncertainties with other experimental data excepting the Devlin's and Leo Kirsch's data
- No resonant structures predicted by the R-matrix analysis of the experimental data measured by Devlin are observed
- The difference with JENDL-5.0 evaluation is on average 8%

Conclusions

- The total cross section of the $^{6}Li(n,t)^{4}He$ reaction was measured in the energy range 4.25-7.50 MeV
- The ^{235}U fission cross-section was used to normalize the data
- A Cs_2LiYCl_6:Ce based scintillation detector was used as a lithium-containing target
- The total systematic measurement error was 4.6-7.0% with a statistical error of 2.0-3.7%.
- The data obtained in the work are in agreement with old experimental results within uncertainties
- The data obtained do not support the evaluated cross section of the $^{6}Li(n,t)^{4}He$ reaction from the ENDF-B/VIII.0 library.
- At the same time, the average difference between the JENDL-5 evaluation and the our data also slightly exceeds the total systematic measurement uncertainty.

Thank you for your attention!

Pavel Prusachenko,

senior researcher of Frank Laboratory of Neutron Physics,
Joint Institute for Nuclear Research

prusachenko@jinr.ru