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Evaluation of ^{232}Th and ^{237}Np fast neutron-induced fission cross sections by simultaneous evaluation approach

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Abstract. The neutron-induced fission cross sections for ^{232}Th and ^{237}Np were evaluated from 500 keV to 200 MeV and from 70 keV to 200 MeV, respectively. The experimental fission cross sections for ^{232}Th and ^{237}Np, as well as their ratios to the ^{235,238}U, and ^{239}Pu fission cross section in the EXFOR library, were reviewed and analyzed using the least-squares method. The newly published ^{232}Th/^{237}Np fission cross-section ratios, obtained from the time-of-flight measurements at the CERN n_TOF and CSNS Back-n facilities, have been added to the EXFOR database. The new evaluation shows that the ^{232}Th fission cross section is systematically lower than that provided by the JENDL-5 library.

1 Introduction

The accurate estimation of fission cross sections is fundamental to the advancement of nuclear science and technology, particularly in the design and operation of nuclear reactors and the development of nuclear data libraries. High-fidelity cross section data are critical for minimising uncertainty in reactor simulations, optimizing reactor performance, and assuring the safe and efficient use of nuclear materials. The significance of accurate fission cross section data for isotopes like as ^{232}Th and ^{237}Np has been emphasized in recent research and play a pivotal role in the development of new reactor designs, including Accelerator Driven Subcritical Systems (ADSS) and fast reactors [1, 2]. However, notable discrepancies still exist in the available data, particularly at specific energy ranges, which can lead to considerable uncertainties in nuclear modeling and safety assessments [3]. Addressing these gaps requires the use of newly measured data and innovative evaluation techniques in order to meet the stringent requirements of current nuclear systems.

As nuclear data libraries increasingly incorporate covariance information, the Nuclear Reaction Data Centres (NRDC) are focusing more on compiling experimental uncertainty and covariance data. They improved the EXFOR format by including correlation property flags and a computer-readable format for matrices [4–6]. New experimental datasets have been gathered at time-of-flight facilities like CERN n_TOF and LANSCE. CERN n_TOF collaboration's recent data has been compiled into the EXFOR library in collaboration with the IAEA Nuclear Data Section [7]. Additionally, there has been progress in compiling benchmark field neutron spectra and cross sections in the EXFOR library for the IRDFF-II project.

The Accelerator Driven Subcritical System (ADSS) can speed up thorium utilization in power generation. In ADSS and similar fast reactors, ^{232}Th fission makes up 3% of total fissions, and at least 8% in gas-cooled reactors, impacting reactor performance significantly. These cross sections significantly contributes 2% of delayed neutrons near the fission threshold (0.7 MeV), compared to 0.25% for ^{233}U [8]. In fast reactors, having higher neutron flux than current thermal reactors, accurate fission data is crucial to reduce design uncertainties. However, existing data for the ^{237}Np(n,f) reaction show discrepancies of up to 8% around 5 MeV, indicating a need for improved accuracy [3].

Simultaneous evaluation is a technique used to assess fission cross sections for isotopes like ^{232}Th and ^{237}Np, along with their ratios to reference cross sections such as ^{232}Th/^{235}U and ^{237}Np/^{235}U. After finalizing the JENDL-5 evaluation [9, 10], we updated our experimental databases to include new data from the EXFOR library and removed a questionable dataset. This article aims to reevaluate the fission cross sections of ^{232}Th (from 500 keV to 200 MeV) and ^{237}Np (from 70 keV to 200 MeV) by including these experimental datasets newly added in the EXFOR library [11], and to check whether the evaluation considering these new measurements support the JENDL evaluation.

2 Evaluation Method

The current evaluation employed the SOK [12, 13] least-squares analysis code to refine prior estimates using experimental datasets. For a detailed explanation of the evalu-

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Table 1. ^{237}Np cross section and ^{237}Np/^{235}U fission cross section ratios included in the experimental database for the present evaluation. "Ver.", "Lab." and "Pts." give the date (N2) of the SUBENT record in EXFOR, EXFOR/CINDA abbreviation [9] of the institute where the experiment was performed, and number of data points, respectively.

EXFOR #	Ver.	First author	Year	Lab.	Pts.	Energy range (eV)		Ref.
^{237}Np								
23736.003	20210331	P.Salvador-Castineira	2017	2UK NPL	7	5.67E+05	2.40E+06	[19]
23189.002	20240717	M.Diakaki+	2013	2GRCATH	4	4.58E+06	5.32E+06	[21]
^{237}Np/^{235}U								
22742.004.1	20160329	M.Diakaki	2016	2ZZZCER	122	1.49E+05	2.00E+06	[3]
51015.002.1[a]	20240716	C.Paradela	2010	2ZZZCER	446	9.77E+02	1.00E+09	[18]
14166.002	20080315	F.Tovesson	2008	1USALAS	1460	1.01E-02	1.98E+05	[22]
41455.004	20170724	O.Shcherbakov	2002	4RUSLIN	166	5.77E+05	1.96E+08	[23]

[a]ad hoc EXFOR entry prepared from 23126.003

ation methods and procedures, readers are referred to our primary reference [14].

2.1 Formalism

The SOK code models the logarithm of the cross section as a linear combination of Schmittroth's roof functions $\phi_j(E)$ that are defined by introducing n energy nodes between E_1 and E_n [10, 12, 13, 15] and is expressed as:

$$\ln \sigma(E) = \sum_{j=1}^{n} \lambda_{\text{eva},j} \phi_j(E) \qquad (1)$$

with $\phi_j(E)$, are the basis functions centered around each energy node E_j, defined by:

$$\phi_j(E) = \begin{cases} \frac{E-E_{j-1}}{E_j-E_{j-1}} & \text{if } E_{j-1} \le E \le E_j, \\ \frac{E_{j+1}-E}{E_{j+1}-E_j} & \text{if } E_j \le E \le E_{j+1}, \\ 0 & \text{otherwise.} \end{cases} \qquad (2)$$

and $\lambda_{\text{eva},j} = \ln \sigma_{\text{eva}}(E_j)$ $(j = 1, n)$ represents the logarithm of the cross section at energy E_j to be determined by fitting. It is equivalent to fitting to the first-order B-spline function where the experimental cross section at energy E_i can be expressed using interpolation between adjacent energy nodes E_j and E_{j+1} $(E_j < E_i < E_{j+1})$ as:

$$\begin{aligned} \lambda_{\text{exp},i} &= \lambda_{\text{eva},j}\ \phi_j(E_i) + \lambda_{\text{eva},j+1}\ \phi_{j+1}(E_i) + \delta_i \\ &= \lambda_{\text{eva},j}\ c_{i,j} + \lambda_{\text{eva},j+1}\ c_{i,j+1} + \delta_i \end{aligned} \qquad (3)$$

where

$$c_{i,j} = \frac{E_{j+1} - E_i}{E_{j+1} - E_j}, \quad c_{i,j+1} = \frac{E_i - E_j}{E_{j+1} - E_j}$$

are interpolating coefficients and δ_i is the residual fitting error. These coefficients ensure a smooth transition between the cross section values at the nodes E_j and E_{j+1}, effectively interpolating the cross section at any intermediate energy E_i.

After solving for the coefficients, we can compute the evaluated cross section across the entire energy range of interest using initial Equation (1).

For more detailed information on the SOK method, including the least-squares solution and handling of cross-section ratios, refer to [12, 13]. For ^{232}Th, prior (initial) estimates λ_{eva} were sourced from JENDL-5 [16] and Yavshits et al., [17] while ^{237}Np the estimates were exclusively from JENDL-5. For other nuclides, JENDL-5 cross sections were used except for ^{233}U above 20 MeV, where results from our simultaneous evaluation [10] were applied.

2.2 Experimental Database

The evaluation process for ^{232}Th, from 500 keV to 200 MeV, and ^{237}Np, from 70 keV to 200 MeV, focused on cross sections and their ratios using experimental reports from 1970 onward, and providing both statistical and systematic uncertainties. Datasets were generally excluded if they were digitized from article figures, compiled without partial uncertainties, or derived from nuclear explosions.
Revisions of EXFOR input: EXFOR entries were revised by adding missing information, removing duplicate datasets from the same measurements, and reformatting the entries. EXFOR 51015.002 [18] prepared to address compilation issues in EXFOR 2316.003. The original uncertainties were almost constant but needed refinement. Table 1 summarizes the experimental ^{237}Np and ^{237}Np/^{235}U datasets used in the current evaluation for the experimental datasets of ^{232}Th, ^{233,235,238}U and $^{239-241}$Pu used in the present evaluation.
Assignment of correlation properties: An essential part of the current evaluation is assignment of correlation properties to each partial uncretainity. We utilized the following two methods to determine the correlation properties for each partial uncertainty.
- adopting correlation coefficient received from experimentalists and compiled in EXFOR
- estimating from partial uncertainties in EXFOR

Conversion to SOK input format: The SOX code [20] converted EXFOR data into SOK input files, assuming counting statistics uncertainties are uncorrelated and other uncertainties are fully correlated within datasets and refining cross-section estimates with experimental data.

3 Result and discussion

Figures 1-4 show the newly evaluated (posterior) cross sections alongside the prior cross sections and experimental data.

3.1 Evaluated fission cross sections for ^{232}Th

Figure 1 shows the ^{232}Th/^{235}U cross-section ratio below 1 MeV. In this energy range, three experimental datasets are available. Fursov et al. [24] and Behrens et al. [25] align

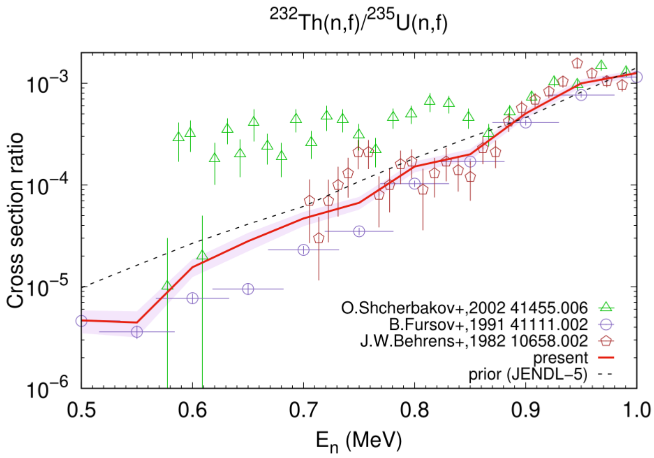

Figure 1. Simultaneous fitting for ^{232}Th/^{235}U with ^{233,235,238}U and $^{239-241}$Pu fission cross section below 1 MeV.

with the present evaluation, while Shcherbakov et al. [23] shows discrepancies below 0.85 MeV. The current evaluation indicates that the cross sections are slightly higher than those in JENDL-5 below 0.7 MeV, which could be due to Shcherbakov et al's data. However, between 0.85

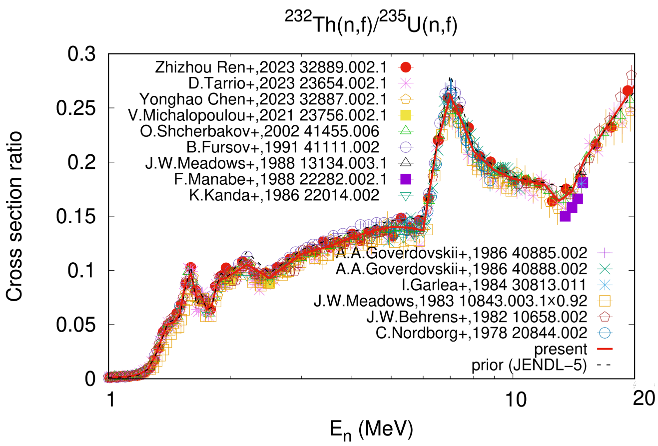

Figure 2. Simultaneous fitting for ^{232}Th/^{235}U with ^{233,235,238}U and $^{239-241}$Pu fission cross section above 1 MeV.

and 1 MeV, all three datasets converge and show better consistency with the JENDL-5 and present evaluations.
Figure 2 illustrates the ^{232}Th/^{235}U cross-section ratio above 1 MeV. The ratio initially obtained was consistently higher than JENDL-5, not accurately reflecting the experimental data. This discrepancy was resolved by treating Meadows' ratio as a shape ratio and adjusting its normalization factor. The adjusted fitting yielded a normalization factor of 0.922 ± 0.010, aligning better with experimental results. Excluding Meadows' ratio from the fitting produced similar outcomes. The revised ratio is systematically lower than JENDL-5 between 2 to 6 MeV and 7 to 15 MeV. In contrast to JENDL-5, which aligns with Fursov et al.'s higher ratio in the 2 to 6 MeV range, the new evaluation does not support this.

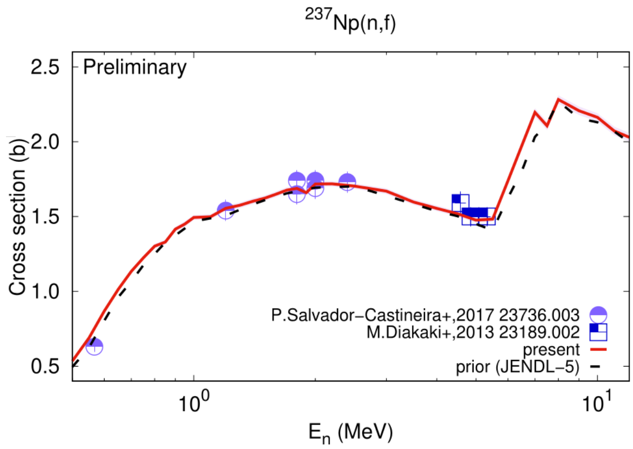

Figure 3. Simultaneous fitting for ^{237}Np with ^{233,235,238}U and $^{239-241}$Pu fission cross section.

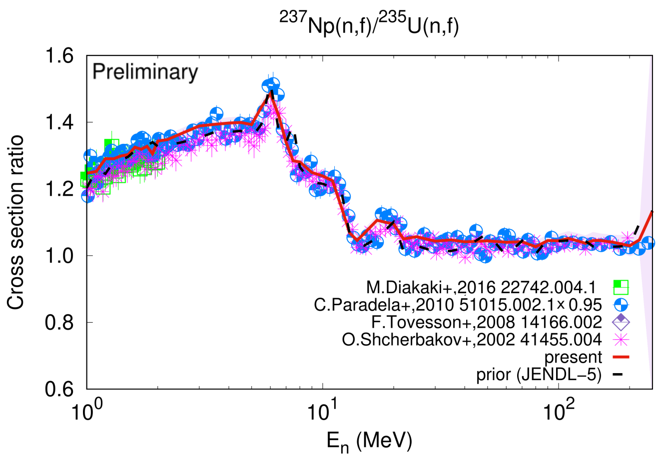

Figure 4. Simultaneous fitting for ^{237}Np/^{235}U with ^{233,235,238}U and $^{239-241}$Pu fission cross section.

3.2 Evaluated fission cross sections for ^{237}Np

Figure 3 illustrates a preliminary result of the ^{237}Np cross section from the current evaluation, compared with two datasets published after 2000. The evaluated cross-section is greater than both JENDL-5 and the experimental data from 0.4 to 1 MeV. However, between 1 and 1.5 MeV, the two experimental datasets come closer and show better

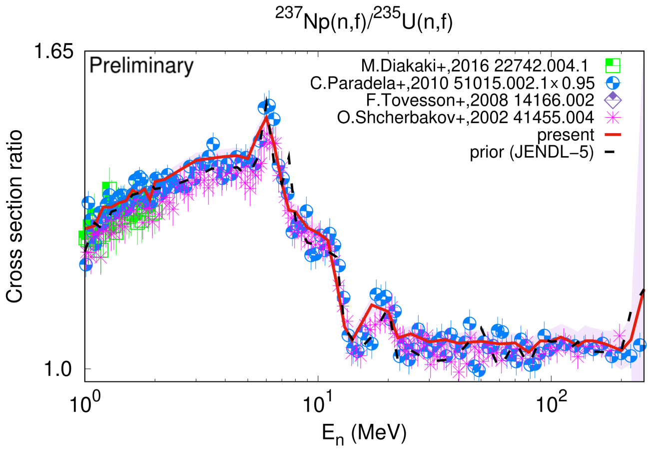

Figure 5. Simultaneous fitting for ^{237}Np/^{235}U with ^{233,235,238}U and $^{239-241}$Pu fission cross section.

consistency with the JENDL-5 and the current evaluation. Figures 4 and 5 display the ^{237}Np/^{235}U fission cross-section ratio for energies above 1 MeV, using linear and logarithmic scales respectively. The ratio initially obtained was consistently higher than JENDL-5 and did not match the experimental data well. This discrepancy was addressed by treating Paradela [18] as a shape ratio and using its overall normalization factor as an additional fitting parameter. This treatment provides a more reasonable fitting, with a normalization factor of 0.95 applied to the original Paradelas' ratio. The figures show that the present ratio is higher than the JENDL-5 ratio in the energy ranges of 1.2 to 1.8 MeV and 30 to 100 MeV. It also aligns well with Paradela's experimental datasets. The present ratio shows consistent behavior between 1.9 and 30 MeV with JENDL-5 and other experimental datasets.

4 Summary

We conducted a new evaluation of the neutron-induced fission cross sections for ^{232}Th (from 500 keV to 200 MeV) and ^{237}Np (from 70 keV to 200 MeV) using the least-squares method. The direct use of EXFOR source files motivated Nuclear Reaction Data Centers to include covariance-related information in the EXFOR library. This simultaneous evaluation includes not only ^{232}Th and ^{237}Np but also the fission cross sections of ^{233,235,238}U and $^{239-241}$Pu. It incorporates the experimental fission cross-section ratios of ^{232}Th/^{235}U, ^{237}Np/^{235}U and ^{232}Th/^{238}U, ^{237}Np/^{238}U into the analysis. The newly evaluated ^{232}Th neutron fission cross sections are systematically lower than the JENDL-5 cross sections.
In the future, it will be interesting to study the simultaneous evaluation of ^{237}Np, ^{233,235,238}U and $^{239-241}$Pu fission cross sections using ^{237}Np datasets published before the year 2000.

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