

From $^{232}\text{Th}(n, n'\gamma)$ cross sections to level production and total neutron inelastic scattering cross sections

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Abstract. To probe the neutron inelastic scattering off ^{232}Th , an experiment took place at the EC-JRC Geel conducted with the experimental setup GRAPhEME to detect emitted γ -rays. The prompt γ -ray spectroscopy method was used and 70 experimental $^{232}\text{Th}(n, n'\gamma)$ cross sections were obtained from the experimental data. Combining these cross sections, nuclear-structure data available in databases and hypotheses to complete the latter, neutron inelastic level production cross sections in ^{232}Th and the total inelastic cross section were calculated. For the first time, the total inelastic cross section of an actinide nucleus was derived on the total neutron energy range from experimental data only. Comparisons of (n, n') cross section data with evaluated data reveal a good agreement between them all above 300 keV of neutron energy. TALYS calculations are compatible but lower than the evaluated data.

1 Introduction

In the context of the fourth generation of nuclear reactor development, new fuels are considered and investigated [1]. Thorium-232 is a nucleus involved in the $^{232}\text{Th}/^{233}\text{U}$ fuel cycle. From a neutron capture on thorium-232 followed by two β^- decays, uranium-233 is produced. This nucleus is interesting for energy purposes as it is a primary fission energy source.

For better accuracy of the simulations of all the different nuclear reactions happening inside the reactors, it is required to know, with the best precision, the cross section of each of these possible processes. Our collaboration, led by IPHC-CNRS, has therefore conducted an experiment at the EC-JRC/GELINA (GEel LINear Accelerator) facility [2, 3] to enhance the accuracy of the inelastic neutron scattering off ^{232}Th .

Using the prompt γ -ray spectroscopy method, 70 $^{232}\text{Th}(n, n'\gamma)$ cross sections have been obtained. From these cross sections and nuclear structure information, we have determined both level population cross sections and the total inelastic scattering cross section as it will be seen in section 3. The results from this experiment will then be shown in section 4 before performing, in section 5, a comparison of our experimental total $^{232}\text{Th}(n, n')$

cross section with experimental values and theoretical calculations.

2 $(n, n'\gamma)$ cross sections

The neutron beam is produced by the GELINA facility. For the study of (n, n') reaction on ^{232}Th [4], GRAPhEME (GeRmanium array for Actinides PrEcise MEasurement) [5], an experimental setup developed by the IPHC-CNRS team, has been used. At the time of this experiment on ^{232}Th , GRAPhEME was composed of 4 HPGe detectors, for the detection of the γ -rays deexcitation. The prompt γ -ray spectroscopy method combined with time-of-flight measurements have enabled the determination of the number of γ s as a function of the neutron energy. From this we can calculate the $(n, n'\gamma)$ cross section for a transition from an excited level i to a lower energy level j designed as $\sigma_{i \rightarrow j}^{(n, n'\gamma)}$. Performing this analysis, 70 $(n, n'\gamma)$ cross sections have been obtained [4].

These cross sections are then used for the calculation of both the neutron level population cross sections of any excited level k , written $\sigma_k^{(n, n')}$, and the total neutron inelastic scattering cross section $\sigma_{\text{tot}}^{(n, n')}$ using respectively the formulae (1) and (2). In these equations, E_n is the incoming neutron energy and $\alpha_{i \rightarrow j}$ represents the internal conversion coefficient of the transition from i to j . This coeffi-

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cient must be considered as the prompt γ -ray spectroscopy method does not provide information about decay through internal conversion.

$$\sigma_k^{(n,n')}(E_n) = \sum_{\substack{\text{transitions emitted} \\ \text{by the level } k \text{ to any level } f}} \sigma_{k \rightarrow f}^{(n,n'\gamma)}(E_n) \times (1 + \alpha_{k \rightarrow f}) - \sum_{\substack{\text{transitions received} \\ \text{by the level } k \text{ from any level } i}} \sigma_{i \rightarrow k}^{(n,n'\gamma)}(E_n) \times (1 + \alpha_{i \rightarrow k}) \quad (1)$$

$$\sigma_{\text{tot}}^{(n,n')}(E_n) = \sum_{\substack{\text{transitions from } i \\ \text{to the ground state}}} \sigma_{i \rightarrow 0}^{(n,n'\gamma)}(E_n) \times (1 + \alpha_{i \rightarrow 0}) \quad (2)$$

3 Results

For the calculation of (n, n') cross sections, as both formulae (1) and (2) indicate, all the $(n, n'\gamma)$ cross sections must have been measured and nuclear-structure data are required to complete them.

Nevertheless, as described in this section, some transitions have not been seen experimentally and not all structure information is available in databases. Thus, hypotheses have been made for the calculation of cross sections.

3.1 Completing the information

We will successively investigate the two causes of incomplete information and see how they have been overcome.

Unobserved transitions

To know if all the transitions were observed experimentally, comparisons have been made with both the nuclear-structure database ENSDF hosted by the NNDC [6, 7] and Demidov's work [8]. This has revealed that, for the levels accessible by (n, n') reactions seen experimentally, a couple of transitions were not measured during our experiment as we have identified 70 out of the 118 that are referenced in either the database or Demidov's work. This difference can be explained by the impurities within the target that increased the density of γ -rays (in the detectors), the multiplicity of γ s sources such as fission products, sample radioactivity... and the discrimination power of our detectors preventing the dissociation of γ -rays too close in energy. Moreover, for high energy γ s, as the higher their energy, the lower the detection efficiency, we have fewer counts.

Nevertheless, we do not necessarily need to directly measure a transition to obtain its cross section data. Indeed, an unmeasured $(n, n'\gamma)$ cross section can be deduced from a measured one, if they are both emitted by the same level with the knowledge of their respective intensity. Looking for intensities in ENSDF, it has been noticed that for the considered 118 $(n, n'\gamma)$ transitions, not all of them have intensities referenced and when they do, sometimes no uncertainty is provided with the value.

In the situation where the intensity is not given, no hypothesis has been made regarding its value and the (n, n') cross sections for both the initial and the final state have not been calculated. When the missing value is the uncertainty of the intensity, a percentage of 10% of the intensity has been chosen as it is the maximum percentage observed in ENSDF. Finally, 29 unmeasured transition cross sections have been deduced from our experimental set of data rising the total number of transition cross sections up to 99.

Internal conversion coefficients

All internal conversion coefficients (ICCs) must be known in order to calculate the (n, n') cross sections as required in both formulae (1) and (2). Nevertheless, out of the 118 $(n, n'\gamma)$ transitions mentioned before, only 48 of them have given ICCs. Therefore, most of them had to be determined and the ICC calculation model BrICC [9] has been used for the unreported coefficients. For the estimation of this quantity, BrICC requires for each transition the multipolarity of the emitted γ -ray. As 50 out of the 118 transitions have a tabulated multipolarity, a hypothesis had to be made for the others. The applied hypothesis has been to consider only the allowed electric and magnetic multipolarities of orders 1 or 2. This is based on the lifetime of the excited states. Finally, ICCs calculated with BrICC are at most 0.1 and in most cases below 0.02.

For the uncertainties on the ICCs, when BrICC has been used for the ICC calculation, the related uncertainty has been considered as they are also calculated by this nuclear model. Otherwise, with the ICC value being found in ENSDF, in almost all cases there is no uncertainty provided with the coefficient so a percentage of 15% of the ICC value has been chosen as it is the maximum BrICC value.

Only 4 (n, n') level population cross sections have been calculated directly from the experimental results without any assumption. With the application of the hypotheses, 23 additional (n, n') level population cross sections and the total (n, n') cross section have been obtained. Out of the 52 excited levels for which we have observed a deexcitation during the GELINA experiment, 27 level population cross sections have therefore been calculated.

3.2 Neutron inelastic scattering cross sections

Level production cross section

Using the formula (1) and the previously exposed hypotheses, level population cross sections can be calculated. Figure 1 represents the (n, n') population cross section for the level at 556.9 keV in ^{232}Th as a function of the incoming-neutron energy.

In this Figure are displayed the $(n, n'\gamma)$ cross sections used for the calculation in various colours and shapes. The one representing a transition emitted by the level at 556.9 keV is in a filled line. The others, depicting transitions that are received by the considered energy level,

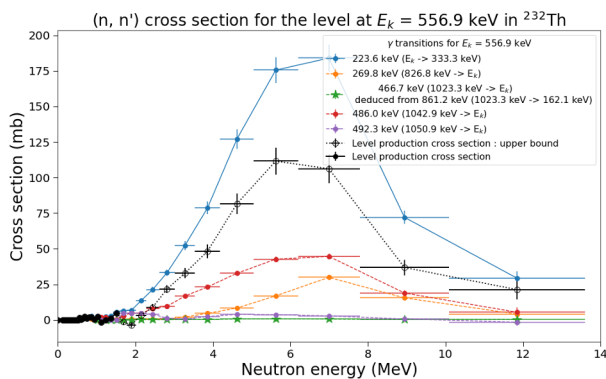


Figure 1. Neutron level production cross section in ^{232}Th for the 556.9 keV level (black-continuous curve) and the upper bound (black-dotted curve) and the $(n, n'\gamma)$ transitions cross sections involving this level (other colours).

are in dashed lines. Among these ones, the 466.7 keV γ -transition, emitted by the 1023.3 keV level, has been deduced from the 861.2 keV γ -transition emitted by this same energy level and is featured with stars in Figure 1.

The level production cross section in black is depicted with filled black dots up to 1.647 MeV. Above this energy, as the structure of the ^{232}Th has not been probed in our experiment, we lack information and the cross section data are an upper bound. This "upper bound" description means that the cross section may be lower considering both higher energy levels and the continuum can feed this level but it will not be higher since all the cross sections of deexcitation transitions have been summed according to formula (1). To underline this difference when analysing the graph, data are shown as unfilled dots joined by a dotted line.

With both the density level increasing at relatively low excitation energy and the efficiency of the detectors preventing the identification of high energy γ s, we get an upper bound of the cross section at low neutron energy.

Total inelastic cross section

The total (n, n') cross section, reported in black in Figure 2, is calculated using the equation (2) which is a sum of all measured $(n, n'\gamma)$ cross sections that feed the ground state.

From threshold reaction up to neutron energy at 1.578 MeV (^{232}Th maximum excitation energy level, probed in our experiment, with a transition downwards the ground state): we have measured $(n, n'\gamma)$ cross sections for all the γ -transitions that decay to the ground state and come from levels at excitation energy up to 1.578 MeV. The calculated-summed total cross section is thus "exact" and is depicted in the Figure with filled black dots.

For neutron energies higher than 1.578 MeV: the nucleus reaches excitation energies higher than 1.578 MeV. We have not measured the 6 γ -transitions (referenced in ENSDF) coming from these high energy levels that can decay directly to the ground state. The summed total cross section is thus lower than the real one, we call it a "lower bound" and it is presented as open black dots in Figure (2).

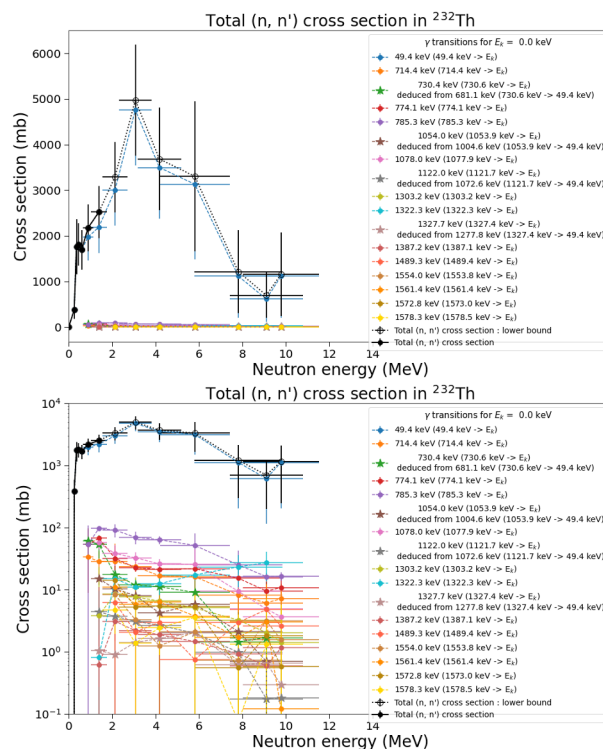


Figure 2. Total neutron inelastic scattering off ^{232}Th , in black continuous for the exact value, the lower bound starting at 1.647 MeV in a dotted line, plotted in a linear scale (top) and in a logarithmic one (bottom). $(n, n'\gamma)$ transition cross sections, downward the ground state, are displayed in various colours.

Additionally to the total (n, n') cross section the 17 $(n, n'\gamma)$ cross sections used for its calculation are also shown, in various colours, in Figure 2. One can notice on the top figure that the 49.4 keV-transition which deexcites the first energy level 2^+ is predominant. Indeed the $^{232}\text{Th}_{90}$ is an even-even nucleus and the deexcitations go mainly through its first state.

It can also be noted that this 49.4 keV-transition is highly converted with an internal conversion coefficient $\alpha = N_e/N_\gamma = 332$ [7], where N_e is the number of emitted electrons and N_γ the number of emitted photons. As GRAPHEME measures the γ s only, we detect a small portion of the decay explaining the large uncertainties on this transition cross section and consequently on the total (n, n') cross section.

4 Comparisons

Our experimental data have been compared with other experimental data as well as with evaluations of the (n, n') cross section, including with the IAEA coordinated evaluation [1, 10, 11] which is adopted by ENDF/B-VII.1 [12], ENDF/B-VIII.0 [13] and JEFF-3.3 [14] evaluated-nuclear-data libraries. Figure 3 is the comparison graph of all the data considered. The experimental cross section data are represented with dots. Those presented in this work are in blue while previous measurement data are in different colours.

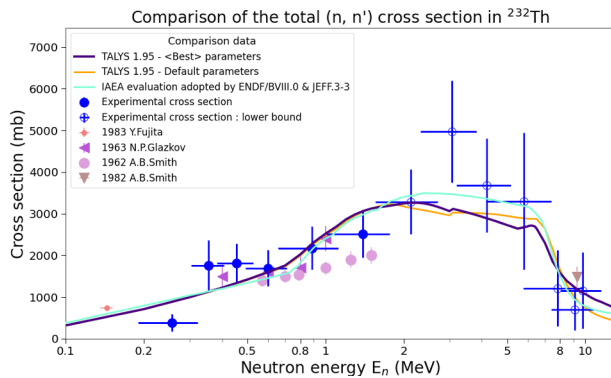


Figure 3. Total neutron inelastic scattering off ^{232}Th . The experimental data are represented with dots, in blue are those presented in this work, filled up to 1.647 MeV and unfilled above for a lower limit of the cross section. Other experimental data are represented in various colours. Theoretical models are displayed with curves as TALYS calculations using the <best> parameters and the default ones are in orange and purple respectively while the IAEA evaluation adopted by both ENDF/B-VIII.0 and JEFF-3.3 is in light blue.

Two calculations from the nuclear model structure code TALYS 1.95 [15] have been considered: one has been done using the default input parameters and another one has been performed using the so-called "best" parameters for the ^{232}Th which has a more accurate depiction of its fission cross section. The nuclear data libraries ENDF/B-VIII.0 and JEFF-3.3 have both adopted the IAEA evaluation which is also shown in Figure 3. Both TALYS calculations and the IAEA coordinated evaluation are represented with curves.

While below 300 keV our value is not in agreement with both TALYS models and the evaluation; above 300 keV, all our data - except the 3.1 MeV one - are in agreement with both TALYS calculations, the evaluation and the other experimental data.

The total (n, n') cross section is dominated by the 49.4 keV γ -ray and this low γ energy is in a region with a lot of background which complicates the extraction of the number of counts. The experimental results presented in this work are not final as we are currently working on the 49.4 keV-signal discrimination.

5 Conclusion

Using GRAPhEME and the prompt γ -ray spectroscopy method for the experiment at the GELINA facility, 70 $^{232}\text{Th}(n, n'\gamma)$ cross sections have been extracted.

From these transition cross sections and by completing them with both referenced information found in the nuclear database ENSDF and in Demidov's work and reasonable assumptions, the calculation of 27 level population cross sections and the total neutron inelastic scattering cross section has been possible. The exact total inelastic cross section has been derived from experimental data below 1.578 MeV for the first time.

The comparison performed with other experimental data, TALYS calculations and the IAEA coordinated eval-

uation adopted both in ENDF/B-VIII.0 and JEFF-3.3 libraries has revealed a good agreement between our experimental data and that evaluation.

This study of the $^{232}\text{Th}(n, n')$ cross section has shown that an important lack of structure data prevents exploring fully all experimental results and the hypotheses have increased in a significant way the number of outcomes. Nevertheless, a few pieces of data had to be left aside as a few structure data can not be evaluated. The work on the cross section determination is still in progress with an investigation of the discrepancies observed for a publication of all these final level population and total (n, n') cross section results in a near future.

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References

- [1] INTERNATIONAL ATOMIC ENERGY AGENCY, *Evaluated Nuclear Data for Nuclides within the Thorium-Uranium Fuel Cycle, Non-serial Publications*, IAEA, Vienna (2010).
- [2] D. Tronc, *et al.* *Nucl. Instrum. Methods Phys. Res. A* **228**, 217 (1985).
- [3] D. Ene, *et al.* *Nucl. Instrum. Methods Phys. Res. A* **618**, 54 (2010).
- [4] E. Party. *Etude des réactions (n, xn) pour les noyaux fertiles/fissiles du cycle du combustible innovant au Thorium*. PhD thesis. Strasbourg University. (2019).
- [5] M. Kerveno, *et al.* *Eur. Phys. J. A* **51**, 167 (2015).
- [6] M. R. Bhat. *Nuclear Data for Science and Technology*, Springer, Berlin, Heidelberg, 817-821 (1992).
- [7] *Evaluated Nuclear Structure Data File, National Nuclear Data File (NNDC)*.
- [8] A. M. Demidov, *et al.* *Phys. At. Nucl.* **71**, 1839-1847 (2008).
- [9] T. Kibédi, *et al.* *Nucl. Instr. and Meth. A* **589**, 202-229 (2008).
- [10] A. Trkov, *et al.* *Nucl. Data Sheets* **112**(12), 3098-3119 (2011).
- [11] M. Sin, *et al.* *Phys. Rev. C*, **74**(1), 014608 (2006).
- [12] M. B. Chadwick, *et al.* *Nucl. Data Sheets* **112**(12), 2887-2996 (2011).
- [13] D. A. Brown, *et al.* *Nucl. Data Sheets* **148**, 1-142 (2018).
- [14] A. Plompen, *et al.* *Eur. Phys. J. A* **56**, 181 (2020).
- [15] A. J. Koning, S. Hilaire and M. C. Duijvestijn, "TALYS-1.0", *Proceedings of the International Conference on Nuclear Data for Science and Technology*, April 22-27, 2007, Nice, France, editors O. Bersillon, F. Gunsing, E. Bauge, R. Jacqmin, and S. Leray, *EDP Sciences*, p. 211-214 (2008).