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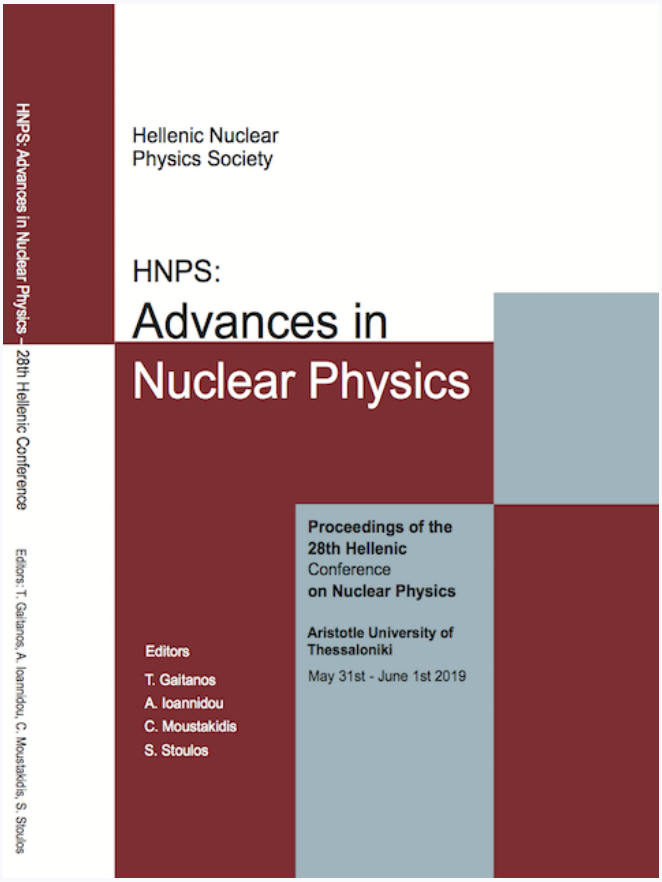

Measurement of the 234U(n,f) cross section in the energy range between 14.8 and 17.8 MeV using Micromegas detectors

Antigoni Kalamara, S. Chasapogloou, V. Michalopoulou, A. Stamatopoulos, Z. Eleme, M. Kokkoris, A. Lagoyannis, N. Patronis, R. Vlastou

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Measurement of the ^{234}U(n,f) cross section in the energy range between 14.8 and 17.8 MeV using Micromegas detectors

A. Kalamara[1], S. Chasapoglou[1], V. Michalopoulou[1,2], A. Stamatopoulos[1], Z. Eleme[3], M. Kokkoris[1], A. Lagoyannis[4], N. Patronis[3], R. Vlastou[1]

[1] *Department of Physics, National Technical University of Athens, Zografou campus, 15780 Athens, Greece*
[2] *European Organization of Nuclear Research (CERN), Geneva, Switzerland*
[3] *Department of Physics, University of Ioannina, 45110 Ioannina, Greece*
[4] *Tandem Accelerator Laboratory, Institute of Nuclear Physics, N.C.S.R. "Demokritos", Aghia Paraskevi, 15310 Athens, Greece*

Abstract *The ^{234}U(n,f) cross section has been measured by using a Micromegas detector assembly, at incident neutron energies of 14.8, 16.5 and 17.8 MeV, where only a few available discrepant data exist in literature leading to poor evaluations. The ^{235}U(n,f) and ^{238}U(n,f) reactions were used as reference ones. The actinide targets and the Micromegas detectors were placed in a chamber filled with a Ar:CO_2 gas mixture at atmospheric pressure. The quasi- monoenergetic neutron beams were produced at the 5.5 MV Tandem Accelerator Laboratory at the National Center for Scientific Research "Demokritos" implementing the ^{3}H(d,n)^{4}He reaction. In addition, α-spectroscopy measurements were performed for the determination of the samples' active masses and the corresponding impurities. Due to the fact that the monochromaticity of the neutron beams was affected by parasitic reactions with the materials of the experimental area, as well as from the beam line itself, a detailed study of the neutron spectra was carried out by coupling both NeuSDesc and MCNP5 codes. Monte-Carlo simulations were also performed by using FLUKA and GEF codes for the estimation of the fission fragments detection efficiency. Finally, a methodology is introduced for the compensation of the parasitic neutron contribution to the experimental fission yield, which can be applied in facilities without time-of-flight capabilities.*

Keywords *cross section, ^{234}U(n,f), neutron beam, α-spectroscopy, Monte-Carlo simulations*

Corresponding author: A. Kalamara (a.kalamara@ipta.demokritos.gr) | *Published online:* May 1st, 2020

INTRODUCTION

The accurate knowledge of neutron induced fission cross sections of actinides leads to the optimization of the design of new generation reactors as well as Accelerator Driven Systems (ADS) [1,2]. Especially ^{234}U, is involved in the Th/U cycle (where it builds up from neutron capture in ^{233}U) which is proposed to replace the Pu/U one in ADS and Generation-IV reactors.

Concerning the experimental data for the ^{234}U(n,f) cross section, several datasets exist in literature, however, for neutron energies between 14 and 18 MeV, there are only 7 datasets [3-9] that present significant discrepancies (12-60%). Therefore, the purpose of this work was to lessen the aforementioned discrepancies and apart from that, to complete the meauserements of the ^{234}U(n,f) reaction over a wide energy range by combining the present project with two previous ones published by A. Tsinganis et al. [10] and A. Stamatopoulos et al. [11]. The final data of the present work have been submitted in the proceedings of the 2019 International Conference On Nuclear Data for Science and Technology (May 19-24 2019, Beijing, China), and they are going to be published on EPJ Web of Conferences during 2020. However, details of this work on the α-spectrosopy measuremets and on the MCNP5 simulations that are not mentioned in the aforementioned publication, will be presented in this one.

DETERMINATION OF THE SAMPLE MASSES

In every cross section measurement, the accurate determination of the number of the target nuclei of the samples that are going to be used is of great importance. In the present work, ^{234}U and ^{238}U samples produced at the IPPE (Obninsk) and JINR (Dubna) with the painting technique were used for the fission cross section measurements. They are actinide disks (5 μm thickness, ~ 5.2 cm diameter) deposited on a 100 μm Al backing [10, 11]. In the front side of the samples (which is going to be measured with the micromegas detector) aluminum masks were placed (0.6 mm thickness, 5.0 diameter) so that all the samples have the same dimensions (see Fig. 1). In the case of ^{234}U, the Al mask was also useful in order to reduce the high counting rate.

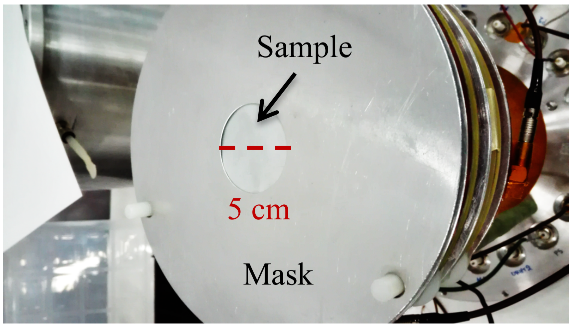

Figure 1. *An actinide sample, along with its aluminum mask.*

In order to determine the active mass of all samples along with the corresponding impurities, α-spectroscopy measurements were performed at the Nuclear Physics Laboratory of the National Technical University of Athens. The measurement was performed by using a silicon surface barrier (SSB) detector, of 6.2 cm diameter, in a vacuum chamber (~10^{-4} atm) and the experimental setup is presented in Fig. 2. A typical spectrum of the ^{234}U sample is shown in Fig. 3.

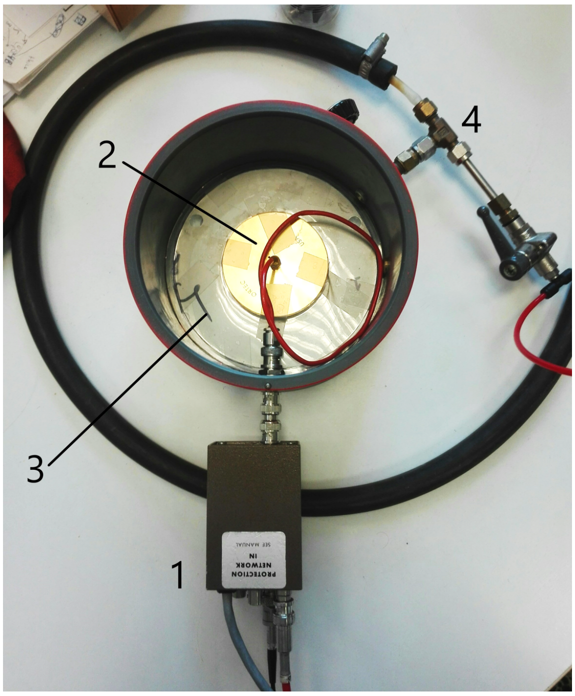

Figure 2. *The vacuum chamber in which the a-spectroscopy was performed. The pre-amplifier (1), the silicon detector (2), the actinide sample (3) and the vaccum pump (4) have been marked.*

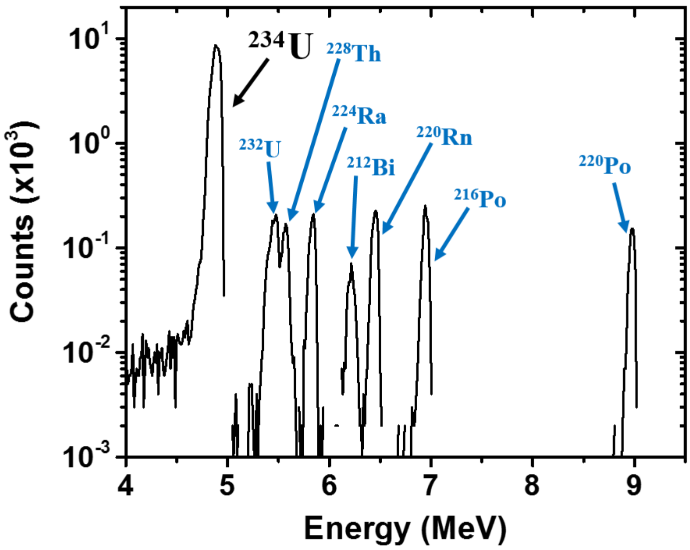

Figure 3. *A typical spectrum of the a-activity emitted from the ^{234}U sample. Apart from the main α-peak at ~4.8 MeV, some other peaks also exist that are attributed to several daughter nuclei (the origin for each one of them is marked), due to the decay chain of ^{234}U. However, these impurities were found to be much less than 1% of the total active mass.*

For each isotope, the activity is given by the following expression:

$$C = \lambda \cdot N \tag{1}$$

where λ is the decay constant of the radioactive nucleus and N is the number of the actinide nuclei in the sample.

However, in the present measurements, an extra correction was needed, which was of crucial importance for the reliability of the results. That is the correction for the solid angle of the measurements. The dimensions of both the detector and the samples, along with the distance between them ((0.10±0.05) cm), had to be accurately determined in order to estimate the solid angle Ω. Therefore, the number of the nuclei of the actinide samples were finally estimated by the following relation:

$$N = C_\Omega \cdot \frac{4\pi}{\Omega} \tag{2}$$

where C_Ω is the activity of the samples, which was determined experimentally (counts/sec) and Ω is the solid angle that was determined by means of the SACALC code [12].

NEUTRON IRRADIATIONS

The cross section measurements were performed at the 5.5 MV Tandem T11/25 Accelerator Laboratory of NCSR "Demokritos". The quasi-monoenergetic neutron beams were produced via the ^{3}H(d,n)^{4}He (D-T) reaction, using a solid Ti-T target of 373 GBq activity, which consists of a 2.1 mg/cm^2 Ti-T layer (25.4 mm in diameter) on a 1 mm thick Cu backing (28.5 mm in diameter). Due to the D-T cross section, the neutron production increases for decreasing deuteron energy. Nevertheless, the lower the deuteron energy, the lower the intensity of the deuteron beam coming from the accelerator. Therefore, a compromise was made by placing two successive Mo foils (5 μm each) so that the deuteron beam impinges on the Mo foils, it looses a part of its energy in them and then it reaches the Ti-T target with a lower energy and consequently with a higher probability to react with the target atoms.

MONTE-CARLO SIMULATIONS FOR THE NEUTRON FLUX

In order to study the neutron beam and to estimate:
- The neutron energy uncertainty,
- The solid angle effect in the neutron flux from one sample to another,
- The ratio: $\frac{(\#\, Neutrons\ per\ cm^2)_{Main\ neutron\ energy\ peak}}{(\#\, Neutrons\ per\ cm^2)_{From\ the\ whole\ energy\ range}} = f_{par}$, which is necessary in order to correct for the low energy parasitic neutrons contribution in the fission yield,
- the NeuSDesc [13] and the MCNP5 [14] codes were implemented.

For the NeuSDesc software, the following information was necessary:
- Reaction: T(d,n)4He, T/Ti target
- Ion energy (keV): 1750, 2300, 2800
  For the three irradiations at 14.8, 16.5 and 17.8 MeV neutron energy, respectively.
- Ti, Li or LiF thickn. (ug/cm2): 2123.5
- T, D/Ti ratio: 1.543
- Entrance foil: Molybdenum
- Entrance foil thickness: 10000 nm
- # Simulated ions in SRIM: 1000
- Customize sdef card settings: (x0, y0, z0) = (-0.305, 0, 0)
  (Vx, Vy, Vz) = (1, 0, 0)
  Properly adjusted in order to match with the geometry described in MCNP5.
- Settings: # Directions: 90
  # Point sources: 5
  The number of angular intervals (so called “Directions”), multiplied by the number of evenly spaced geometrical positions (so called “Point sources”) is limited in MCNP to 498, which is adequate for a good neutron source description.

The output of the NeuSDesc is a detailed description of the D-T reaction, in which the deuteron energy loss in the target and entrance foil have been taken into account. Moreover, the format of the output is compatible with the MCNP code (sdef card) and therefore, it can be used as is in the MCNP code.

Once the neutron source definition card (sdef) has been obtained using the NeuSDesc code, it can be used in the MCNP5 code, in order to propagate the neutrons over the whole irradiation setup (Cu backing, Al flange and other materials of the target assembly). The results of the MCNP simulations, that were executed for 10^8 number of simulated particles (nps), give the neutron energy distribution for each irradiation and they are presented in Figs. 4(a) – 1(c).

Concerning the estimation of the solid angle effect in the neutron flux for a reference (^{238}U) and a measured target (^{234}U), the integral of the main neutron energy peak was determined for each of the targets, by integrating the areas under the solid and dashed lines in Fig. 5, respectively. The ratio of these two integrals $\left(\frac{Neutron\ Flux\ {}^{238}U}{Neutron\ Flux\ {}^{234}U} = \frac{\Phi\, {}^{238}U}{\Phi\, {}^{234}U}\right)$ corresponds to the effect of the solid angle (see also Eq. (4) in “Analysis” section).

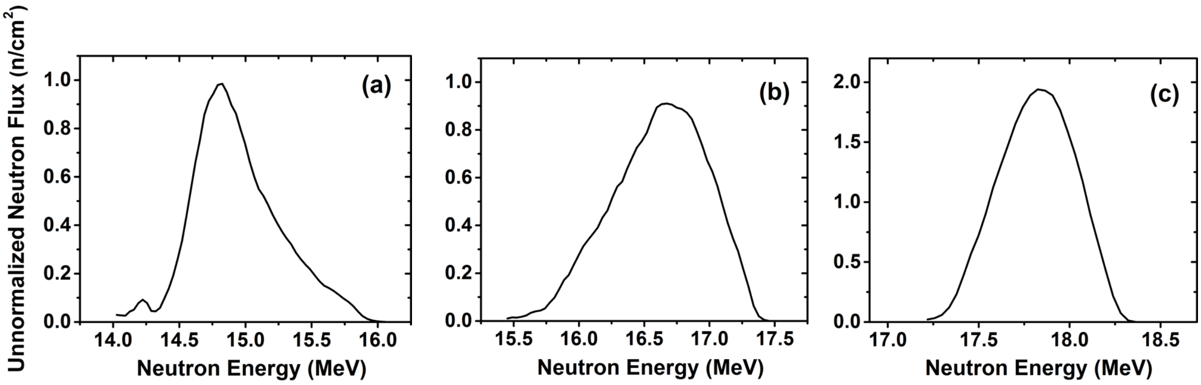

Figure 4. *Unnormalized neutron flux for the three irradiations at (a) (14.8 ± 0.3), (b) (16.5 ± 0.3), and (c) (17.8 ± 0.2) MeV, as obtained by coupling the NeuSDesc and MCNP5 codes. The figures are focused on the main neutron energy region, to highlight the FWHM of each neutron peak from which the neutron energy uncertainty was estimated.*

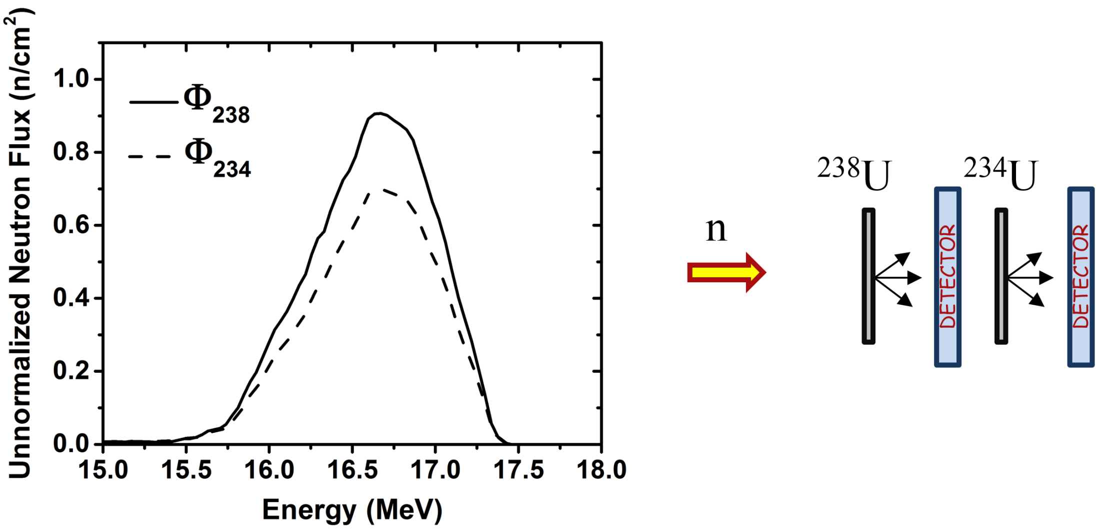

Figure 5. (Left) *The results of the MCNP5 simulation for the neutron flux with respect to the neutron energy in both the reference (^{238}U) and measured (^{234}U) targets, for the irradiation at (16.5 ± 0.3) MeV. As shown in the schematic representation (Right), the ^{238}U target was placed in front of the ^{234}U one, with respect to the neutron beam.*

During the irradiations, apart from the main energy neutrons, also some low energy ones exist in the neutron spectrum. Such neutrons, can also be called "parasitic" and may stem from break-up reactions, such as ^{3}H(d,np), ^{3}H(d,2n), ^{3}H(d,nd) etc, from reactions with ^{12}C nuclei (^{12}C(d,n)) that are present due to the carbon built up process, from reactions with the materials of the beam pipes (i. e ^{16}O(d,n), due to oxidization processes) and also from scattering in the materials of the whole experimental area. In order to determine the ratio: $\frac{(\#\, Neutrons\ per\ cm^2)_{Main\ neutron\ energy\ peak}}{(\#\, Neutrons\ per\ cm^2)_{From\ the\ whole\ energy\ range}} = f_{par}$, which is necessary in order to correct for the low energy parasitic neutrons contribution in the fission yield, the simulated flux over the whole energy range was needed. The simulated neutron flux with respect to the neutron energy is presented in Fig. 6.

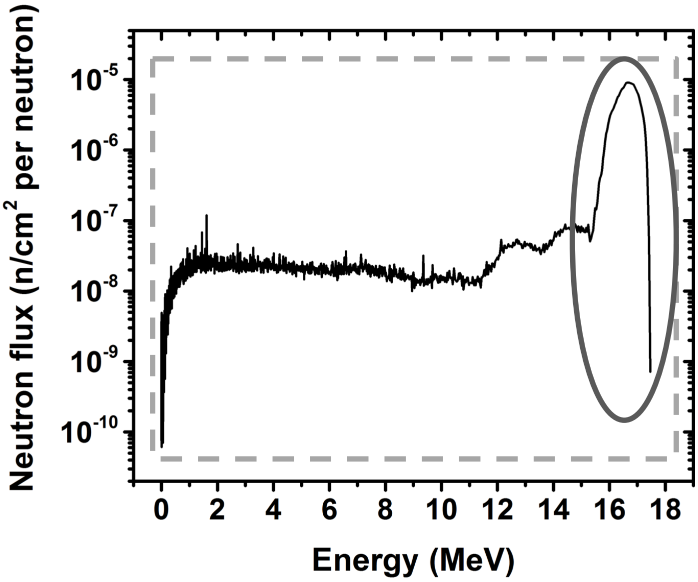

Figure 6. *The results of the MCNP5 simulation for the neutron flux with respect to the neutron energy over the whole energy range, for the irradiation at (16.5 ± 0.3) MeV. The ratio of the integrals of the two indicated regions (dark grey solid circle/light grey dashed square) is the aforementioned f_{par}, which is necessary for the cross section determination, see Eq. (4). The f_{par} factor was of the order of ~0.8 and ~0.9, for the ^{234}U and ^{238}U samples, respectively.*

ANALYSIS

The cross sections were determined according to the following relation:

$$\sigma_{^{234}U} = \sigma_{^{238}U} \cdot \frac{\Phi_{^{238}U}}{\Phi_{^{234}U}} \cdot \frac{N_{t\,^{238}U}}{N_{t\,^{234}U}} \cdot \frac{(N_{ff} \cdot f_{DT} \cdot f_{abs} \cdot f_{cut} \cdot f_{par} \cdot f_{line})_{^{234}U}}{(N_{ff} \cdot f_{DT} \cdot f_{abs} \cdot f_{cut} \cdot f_{par} \cdot f_{line})_{^{238}U}} \qquad (4)$$

where $\sigma_{^{238}U}$ is the reference reaction cross section, Φ is the neutron flux and N_t is the number of the target nuclei. In addition, N_{ff} is the number of the recorded fission fragments (ff) in the spectrum, while the following five factors f_{DT}, f_{abs}, f_{cut}, f_{par}, f_{line} are used to correct the ff integral (N_{ff}).

The reference reaction cross section for the ^{238}U(n,f) reaction ($\sigma_{^{238}U}$) was adopted from the ENDF/B-VIII.0 library [15]. The number of the target nuclei (N_t) was determined by means of the α-spectroscopy measurements that were described in the first section above. As it was mentioned in the previous section, the neutron flux ratio $\Phi_{^{238}U}/\Phi_{^{234}U}$ was determined through Monte-Carlo simulations. The N_{ff} was determined by the integral of the ff peak in the experimental spectrum obtained using the Micromegas detector (see Fig. 7). In order to correct for the dead time of the read-out system, the f_{DT} factor was used, which is given by the expression:

$$f_{DT} = \frac{Real\ Time}{Live\ Time} \qquad (5)$$

where the real and live times correspond to the experimental measurement. In addition, the next two correction factors, f_{abs} and f_{cut}, were both estimated though Monte Carlo simulations, by coupling the GEF [16] and FLUKA [17] codes. The f_{abs} was used to correct for the absorption of ff in the targets, while the f_{cut} in order to correct for the rejected ff due to the introduction of the amplitude cut in the analysis (see Fig. 7). This correction is necessary, since as shown in Fig. 7, in channels that lie lower than the amplitude cut, apart from the α-particles, probably some ff have been recorded. If it wasn't for f_{cut}, these low-channel ff would be neglected. Moreover, both f_{par} and f_{line} factors were used to account for the contribution of low energy parasitic neutrons in the fission yields. The former was determined via Monte Carlo simulations by coupling the NeuSDesc and MCNP5 codes, as it was mentioned in the previous section, while the latter was determined experimentally. More specifically, each irradiation was repeated with the Ti-T target replaced by a Cu foil and therefore, all the recorded ff attributed to parasitic neutrons from the beam line were estimated by normalizing to the deuteron current of the main beam. The f_{line} factor was defined by the following expressions:

$$f_{line} = \frac{(N_{ff})_{Tritium}}{(N_{ff})_{Total}} \tag{6}$$

$$(N_{ff})_{Total} = (N_{ff})_{Tritium} + (N_{ff})_{Parasitic} \tag{7}$$

$$(N_{ff})_{Parasitic} = (N_{ff})_{Cu} \cdot \frac{Q_{Tritium}}{Q_{Cu}} \tag{8}$$

where $Q_{Tritium}$ and Q_{Cu} are the total deuteron currents during the irradiations with the tritium and Cu targets, respectively. The f_{line} found to be ~ 1, except for the case of the ^{234}U target in the irradiation at (17.8 ± 0.2) MeV, in which it was 0.93.

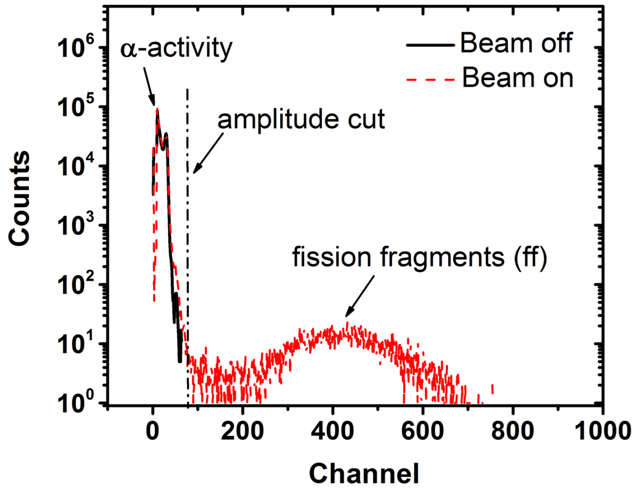

Figure 7. *Experimental spectrum of the ^{238}U target obtained using a Micromegas detector. The black solid line corresponds to the α-activity of the target (Beam off), while the red dashed one (Beam on) is the spectrum that was obtained during the irradiation at (16.5 ± 0.3) MeV. On the latter, apart from the peak of the α-background activity, also the peak of the fission fragments is present.*

RESULTS AND DISCUSSION

The neutron induced fission cross section was measured at 14.8, 16.5 and 17.8 MeV by using Micromegas detectors. In order to correct for the contribution of low energy parasitic neutrons in the fission yield, Monte-Carlo simulations were performed by coupling the NeuSDesc and MCNP5 codes. This correction (f_{par}) was crucial for the accurate determination of the cross section and therefore, the results are presented before and after this correction in Fig. 8, along with previously exisiting data [18] and evaluation libraries [15].

The final values (red points) at 14.8 and 17.8 MeV are in excellent agreement within their errors with previously existing experimental data and seem also to agree with the data by Karadimos et al. [4]. The latter dataset present a special interest, since in that project the same samples with those used in the present project had been used. Concerning the new data point at 16.5 MeV, it lies slightly highwer than the other data points and evaluation libraries. However, it reveals an increasing trend of the cross section curve around 16 MeV, which is also indicated by ENDF/B-VIII.0, JEFF-3.3 and TENDL-2017 evaluation curves.

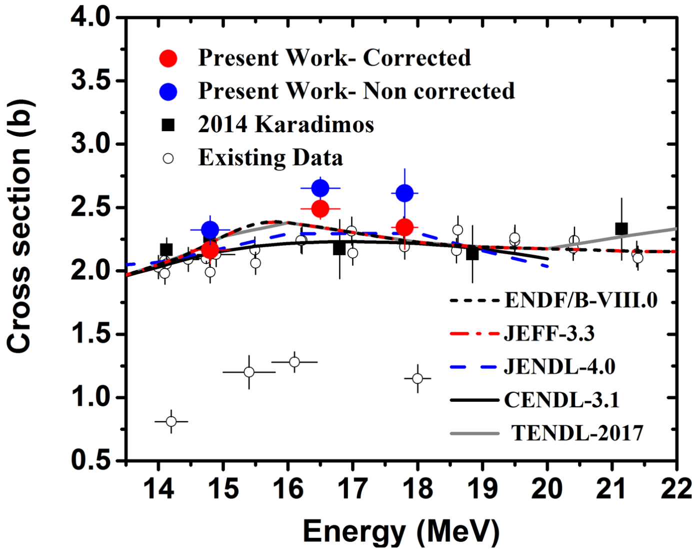

Figure 8. *Cross section results for the $^{234}U(n,f)$ reaction, with and without the f_{par} correction.*

Acknowledgments

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References

[1] NEA, Accelerator-driven Systems (ADS) and Fast Reactors (FR) in advanced nuclear fuel Cycles, Technical Report (Nuclear Energy Agency of the OECD, NEA, 2002).
[2] https://www.gen-4.org, Gen-IV International Forum.
[3] F. Tovesson, A. Laptev and T. S. Hill, Nuclear Science and Engineering 178, 57-65 (2014), doi: 10.13182/NSE13-56
[4] D. Karadimos and the n_TOF Collaboration, Phys. Rev. C 89, 044606 (2014), doi: 10.1103/PhysRevC.89.044606
[5] C. Paradela and the n_TOF Collaboration, Phys. Rev. C 82, 034601 (2010), doi: 10.1103/PhysRevC.82.034601
[6] Manabe et al.,Technology Reports of the Tohoku University 52, 97-126 (1988).
[7] J. W. Meadows, Annals of Nuclear Energy 15, 421-429 (1988), doi: 10.1016/0306-4549(88)90038-2
[8] P. H. White and G. P. Warner, Journal of Nuclear Energy 21, 671-679 (1967), doi: 10.1016/0022-3107(67)90082-2
[9] R. Babcock, Technical Report, Bettis Atomic Power Lab., Westinghouse, Pittsburgh, PA, USA (1961).
[10] A. Tsinganis et al., EPJ Web of Conferences 146, 04035 (2017), doi: 10.1051/epjconf/201714604035
[11] A. Stamatopoulos et al., Eur. Phys. J. A 54, 7 (2018), doi: 10.1140/epja/i2018-12429-2
[12] R. Whitcher, SACALC3 (2012), URL: https://sites.google.com/site/averagesolidangle2/dow/sacalc3
[13] E. Birgerssone and G. Lovestam , JRC Scientific and Technical Reports (2007).
[14] MCNP-A General Monte Carlo N-ParticleTransport Code, version 5, X-5 Monte Carlo team (2003), LA-UR-03-1987, LA-CP-03-0245 and LA-CP-03-0284.
[15] ENDF, URL: 10.1016/j.nds. 2011.11.002
[16] K.H. Schmidt et al., Nucl. Data Sheets 131, 107 (2016), doi: 10.1016/j.nds.2015.12.009
[17] A. Ferrari et al., FLUKA: A multi-particle transport code (program version 2005) (CERN, Geneva, 2005) cds.cern.ch/record/898301.
[18] EXFOR, URL: https://www-nds.iaea.org/exfor/