EPJ Web of Conferences **146**, 04035 (2017) DOI: 10.1051/epjconf/201714604035
ND2016

Neutron-induced fission cross-section measurement of ^{234}U with quasi-monoenergetic beams in the keV and MeV range using micromegas detectors

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Abstract. Accurate data on neutron-induced fission cross-sections of actinides are essential for the design of advanced nuclear reactors based either on fast neutron spectra or alternative fuel cycles, as well as for the reduction of safety margins of existing and future conventional facilities. The fission cross-section of ^{234}U was measured at incident neutron energies of 560 and 660 keV and 7.5 MeV with a setup based on 'microbulk' Micromegas detectors and the same samples previously used for the measurement performed at the CERN n_TOF facility (Karadimos et al., 2014). The ^{235}U fission cross-section was used as reference. The (quasi-)monoenergetic neutron beams were produced via the ^{7}Li(p,n) and the ^{2}H(d,n) reactions at the neutron beam facility of the Institute of Nuclear and Particle Physics at the 'Demokritos' National Centre for Scientific Research. A detailed study of the neutron spectra produced in the targets and intercepted by the samples was performed coupling the `NeuSDesc` and `MCNPX` codes, taking into account the energy spread, energy loss and angular straggling of the beam ions in the target assemblies, as well as contributions from competing reactions and neutron scattering in the experimental setup. Auxiliary Monte-Carlo simulations were performed with the `FLUKA` code to study the behaviour of the detectors, focusing particularly on the reproduction of the pulse height spectra of α-particles and fission fragments (using distributions produced with the `GEF` code) for the evaluation of the detector efficiency. An overview of the developed methodology and preliminary results are presented.

1. Introduction

Feasibility, design and sensitivity studies on new generation reactors require high-accuracy cross-section data for a variety of neutron-induced reactions from thermal energies to several tens of MeV. Capture and fission cross-section of isotopes involved in the Th/U fuel cycle, long-lived Pu, Np, Am and Cm isotopes, long-lived fission fragments relevant for transmutation projects or isotopes considered as structural materials for advanced reactors are among the nuclear data for which there is pressing need in the context of new reactor designs and of waste transmutation applications.

Although ^{234}U in particular plays a limited role in the ^{235}U cycle, it is more important in the context of the thorium cycle, where it builds up due to neutron capture in ^{233}U and acts like ^{240}Pu in the conventional uranium cycle. There are a number of previous measurements of this cross-section in the energy ranges studied in this work, most of them performed several decades ago. More recently, three time-of-flight measurements have been performed by Paradela et al. [1] and Karadimos et al. [2] at the CERN n_TOF facility and by Tovesson et al. [3] at LANSCE (Los Alamos). These three measurements are in overall good agreement with each other, although the data by Karadimos are systematically higher in the energy range between 0.5–1.0 MeV.

Preliminary results are presented here for three energies (560, 660 keV and 7.5 MeV). Data from irradiations at 460 keV, 6.5, 8.7 and 10.0 MeV are being analysed and will be complemented by further irradiations in the 4-6 MeV range in the near future.

2. Experimental

2.1. Detectors and data acquisition

The measurements for this work were performed with Micromegas (Micro-MEsh GAseous Structure) detectors [4–6] of the 'microbulk' variant [7,8]. In the Micromegas, which is a micro-pattern gas detector (MPGD), the gas volume between the anode and cathode is separated into two regions by a micromesh. The micromesh is a thin (~5 μm) conductive layer with 35 μm diameter holes on its surface at a distance of 50 μm from each other, although these values may slightly vary.

The electrical field in the drift region, where the primary ionisation of the gas by the particles emitted by the samples takes place, was set to ~0.9kV/cm. The drift gap was 8 mm. In the amplification region, which is 50 μm wide and where the charge multiplication occurs, a much higher field of ~48 kV/cm was applied. In these

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conditions, where the ratio of the two fields is very high (>50 in this case) the configuration of the electrical field lines becomes such that the micromesh is 100% transparent to electrons.

A stainless steel chamber capable of holding up to 10 sample-detector modules was used to house the samples and detectors and was filled with a circulating Ar:CO_2 (85:15) mixture at atmospheric pressure. An internal support frame is in place to mount the sample holders and detectors. The cylindrical chamber is placed along the beam-line and its axis is aligned to the neutron beam. The entrance and exit windows are made of 25 μm-thick kapton and have a diameter of 15 cm. The chamber was placed a few centimetres (between 5–12 cm) from the neutron source.

The detector signals were read through ultra low-gain preamplifiers followed by standard spectroscopy amplifiers and ADCs. As shown later, this gives a reasonably good energy resolution and a clear separation between α-particles and fission fragments.

2.2. Samples

The actinide samples used were produced with the painting technique by IPPE (Obninsk) and JINR (Dubna) and were originally used at n_TOF (including in the work by Karadimos et al. on ^{234}U, mentioned earlier). In addition to ^{234}U, ^{235}U and ^{238}U samples were included as a reference. Aluminium masks of 0.5 mm thickness and a 4 cm diameter hole were placed in front of the samples in order to keep their angular acceptance with respect to the neutron source to within $\pm 5 - 10^{\circ}$, thus minimising the uncertainty in the energy of the intercepted neutrons. The effective total mass of the different isotopes was 7.5, 5.0 and 5.6 mg for ^{234,235,238}U respectively, as measured by α-counting with an uncertainty of up to 2%. As reported by Diakaki et al. [9], the homogeneity of a subset of the available actinide samples was checked by RBS (Rutherford Back-Scattering) and they were found to be laterally homogeneous within 10–15%.

2.3. Neutron beam and irradiations

The experiment was performed at the 5.5 MV Tandem van de Graaf of the 'Demokritos' National Centre for Scientific Research. Irradiations were performed with two different neutron source setups to cover energies in the hundreds of keV and in the few MeV range.

Thin LiF deposits (~55 μg/cm^2) on tantalum backings were produced by evaporation and bombarded with continuous proton beams to produce neutrons in the 400-700 keV range via the ^{7}Li(p,n)^{7}Be reaction. This reaction yields strictly monoenergetic neutrons from threshold up to about 500 keV, where the first excited state of ^{7}Be begins to contribute to the emission of a second 'family' of neutrons. This contribution increases with energy and it was accounted for in the analysis of the data presented here. The flux values achieved in the samples with this setup were between $2–5 \times 10^4$ n·cm^{-2}·s^{-1}, while the width of the nominal neutron energy peak is of the order of 15 keV.

The quasi-monoenergetic neutron beams in the MeV region were produced via the ^{2}H(d,n)^{3}He reaction by bombarding a deuterium gas target with deuteron ions.

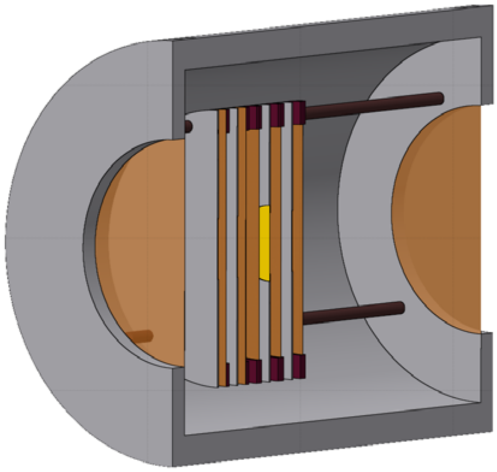

Figure 1. A 3D view of the geometry of the chamber, including samples and detectors, as simulated in MCNPX. The geometry of the appropriate neutron source was added in each case at the correct position, a few centimetres from the entrance window.

The 3.7 cm long gas target is fitted with a 5 μm molybdenum entrance foil and a 1 mm platinum beam stop and is constantly cooled with a cold air jet during irradiation to diminish the risk of damage to the Mo foil. The deuterium pressure was kept at 1000–1300 mbar and controlled remotely, while its values were also recorded at a regular interval. Using this setup, the achieved flux in the samples varied between 3×10^5-4×10^6 n·cm^{-2}·s^{-1}, with a width of the nominal neutron energy peak of about 30 keV. For the these irradiations, data was taken both with and without the deuterium gas in the gas cell in order to subtract the contribution of parasitic reactions, such as deuteron break-up and other neutron-producing reactions on various materials (molybdenum isotopes in the entrance window, the gas cell, deuterium implanted in the Pt beam-stop etc.).

3. Analysis

3.1. Simulations

The neutron production and propagation through the experimental setup was studied in detail for both neutron source setups. In particular, the NeuSDesc (Neutron Source Description) code [10], developed at JRC-IRMM (Geel, Belgium), was used to obtain the energy spectrum and angular distribution of the produced neutrons for each setup and for each beam energy. NeuSDesc makes use of the SRIM (Stopping and Range of Ions in Matter) software [11,12] to handle the energy loss and angular straggling of the ions in the neutron producing target (e.g. lithium, deuterium gas target etc.) and surrounding elements, such as entrance windows and beam stops, as defined by the user. These results are then convoluted with available double differential cross-sections of the neutron producing reactions to generate a realistic neutron source, including competing reactions, such as deuteron break-up and ^{7}Li(p,n$_1$). An additional useful feature of the code is the ability to export this information as an MCNPX [13] source definition. The geometry of the neutron sources and the experimental setup (chamber with samples, detectors, supports etc.) was built in MCNPX and the NeuSDesc output was used as a source. A 3D section of the simulated geometry is shown in Fig. 1.

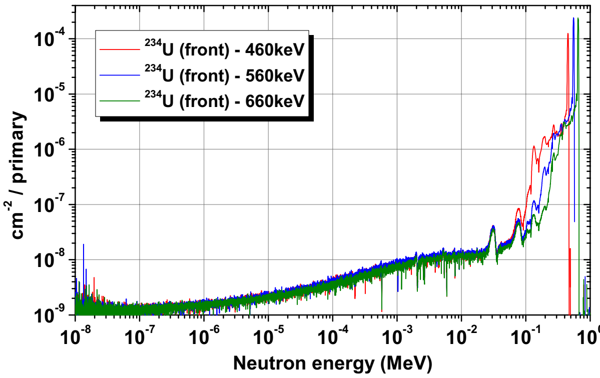

Figure 2. Simulated neutron fluence in the front ^{234}U sample for nominal neutron beam energies of 460, 560 and 660 keV. The nominal peaks can be seen, as well as the long 'tails' of low-energy neutrons.

The neutron fluence was scored within the volume of each sample, while neutrons were scored down to thermal energies in order to estimate the contribution of the neutrons with energies lower than the nominal one to the measured fission counts. An example of the results in shown in Fig. 2, where the calculated neutron fluence in the first ^{234}U sample is shown for three different nominal neutron beam energies. The main peak can be seen, along with an extension of the spectrum to very low energies. The number of fission events caused by these parasitic neutrons was estimated by convoluting the neutron fluence predicted by the `NeuSDesc-MCNPX` calculation with the evaluated cross-sections of the respective isotopes. With this method, this contribution was found to be between 10–12%, depending on the isotope. The sensitivity of this factor to the choice of a particular evaluated (or calculated, e.g. TENDL) cross-section was checked. Despite differences among these, the variation of the correction estimated with different libraries was no more than 1%.

Further refinements of the simulations are foreseen, both in the level of detail of the geometry as well as in a sensitivity study of the results to different parameters, such as the effect of possible misalignments of the chamber or an incorrect estimation of the distance of the samples from the neutron source.

Additional simulations were performed with `FLUKA` [14, 15], using fission fragment distributions generated with the `GEF` code [16], to estimate the fraction of fission fragments stopped inside the samples and possible edge effects near the rims of the masks. These contributions were found to be well below 1%. The behaviour of the detectors was also studied, focusing particularly on the reproduction of the pulse height spectra of α-particles and fission fragments for the evaluation of the fraction of fission signals rejected by the amplitude threshold applied to the pulse-height spectrum. Depending on the selected threshold, this correction varied between 3 and 7%. This method was validated in the past using the well known spontaneous fission rate of ^{242}Pu.

3.2. Data analysis

A pulse-height spectrum obtained from a ^{234}U sample is shown in Fig. 3. Despite the long tail caused by multiple α-particle pile-up, the separation between α-particles and fission fragments remains clear, even more so in the case

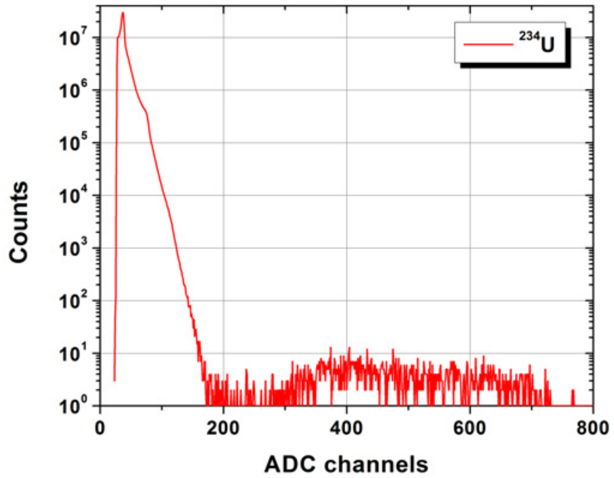

Figure 3. Pulse-height spectrum obtained from one of the ^{234}U samples.

of ^{235}U, whose activity is of the order of a few hundred Bq, compared to over 1 MBq in the case of ^{234}U. The amplitude threshold value was chosen based on the beam-off spectra so as to completely reject the α-particle background.

For each incident neutron energy, the cross-section was calculated using the formula:

$$\sigma = \frac{N}{N_{ref}} \cdot \frac{n_{ref}}{n} \cdot \frac{\epsilon_{ref}}{\epsilon} \cdot \frac{f_{ref}}{f} \cdot \sigma_{ref} \tag{1}$$

where N are the integrated fission counts corrected for the low-energy neutron contribution, n are the areal densities of the samples, ϵ is the detector efficiency, understood as the fraction of fission fragments that exit the sample deposit and enter the active volume of the detector and f is the correction factor for the amplitude threshold applied to the data (i.e. the fraction of detected fission fragments above the threshold). The subscript 'ref' refers to the respective quantities corresponding to the reference isotope, in this case ^{235}U. The neutron fluence value obtained from ^{235}U was cross-checked by γ-spectroscopy of the activated LiF foils, using the 478 keV line from the ^{7}Be decay.

4. Results and discussion

The results of the cross-section calculations can be seen in Figs. 4 (560 and 660 keV) and 5 (7.5 MeV) together with data from previous measurements and evaluations. The obtained cross-section value at 560 keV seems a few percent higher than Paradela and Tovesson, although the results are compatible within uncertainties. The significantly higher values reported by Karadimos at this energy are not confirmed. Considering that the samples used in this measurement are the same, this would seem to rule out any systematic uncertainties relating to the samples themselves (e.g. the presence of undeclared contaminants). The same can be said of the value obtained at 660 keV, where there is very good agreement with Paradela and Tovesson. At 7.5 MeV, the measured cross-section is in very good agreement with the recent measurements.

The agreement of the obtained results with the recent time-of-flight measurements provides a first strong

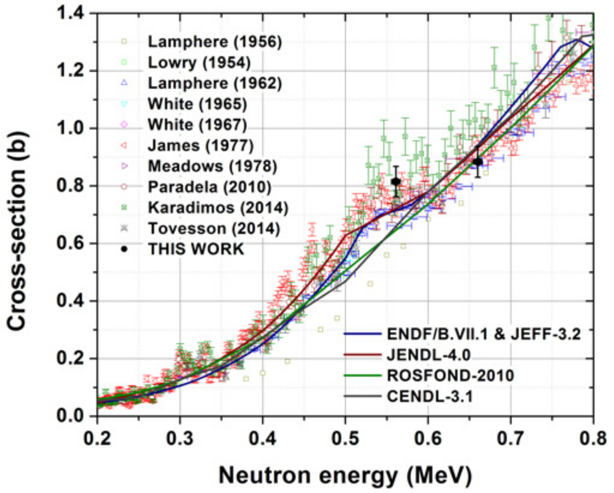

Figure 4. Results in the keV region compared with previous datasets and major evaluated libraries.

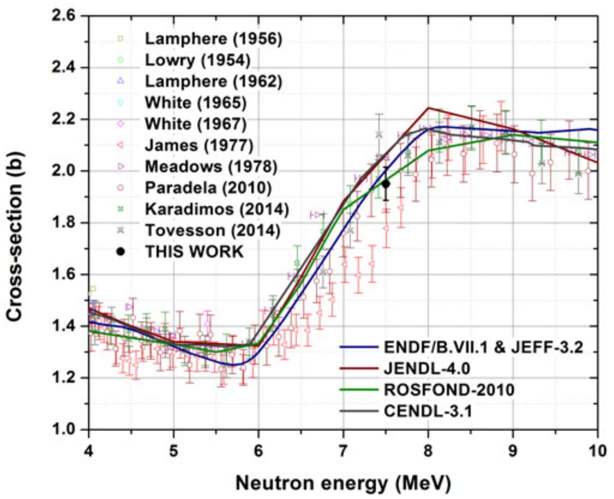

Figure 5. Results in the MeV region compared with previous datasets and major evaluated libraries.

validation of the adopted analysis methodology, including the correction for the contribution of parasitic low-energy neutrons obtained from the calculations and simulations performed with NeuSDesc and MCNPX. This methodology will be extended to energies up to 10 MeV using the deuterium-based setup and in the range around 20 MeV, where neutrons are produced through the ^{3}H(d,n)^{4}He reaction. This comprehensive validation should allow this method to be confidently applied to the measurement of less well-known cross-sections in the future.

5. Summary

The neutron induced fission cross-section of ^{234}U has been measured with monoenergetic beams at incident energies of 560 and 660 keV and 7.5 MeV. Further irradiations between 4 and 6 MeV neutron energy are planned for the near future. The results obtained are in very good agreement with recent time-of-flight measurements performed at n_TOF (CERN) and LANSCE (Los Alamos). The higher values reported by Karadimos et al. in the 0.5–1.0 MeV range are not confirmed. These results provide a first validation of the analysis methodology, including the estimation of the contribution from low-energy neutrons to the measured fission yields.

References

[1] C. Paradela et al., Phys. Rev. C **82**, 034601 (2010)
[2] D. Karadimos et al., Phys. Rev. C **89**, 044606 (2014)
[3] F. Tovesson, Nucl. Sci. Eng. **178**, 57 (2014)
[4] Y. Giomataris et al., Nucl. Instrum. Meth. A **376**, 29 (1996)
[5] Y. Giomataris, Nucl. Instrum. Meth. A **419**, 239 (1998)
[6] I. Giomataris, ICFA Instrum. Bul. **19** (1999)
[7] S. Andriamonje et al., J. Instrum. **5**, P02001 (2010)
[8] S. Andriamonje et al., J. Korean Phys. Soc. **59**, 1597 (2011)
[9] M. Diakaki et al., Eur. Phys. J. A **49**, 1 (2013)
[10] E. Birgersson, G. Lovestam, Tech. Rep. EUR 23794 EN, JRC-IRMM (2009)
[11] J. Ziegler, Nucl. Instrum. Meth. B **219220**, 1027 (2004)
[12] J. Ziegler, *Stopping and Range of Ions in Matter, SRIM 2013*, www.srim.org
[13] L. Waters et al., in *AIP Conference Proceeding* (AIP, Fermilab, USA, 2007) **896**, 81–90
[14] A. Ferrari, P. Sala, A. Fass, J. Ranft, *FLUKA: A multi-particle transport code (program version 2005)* (CERN, Geneva, 2005)
[15] G. Battistoni et al., in *AIP Conference Proceeding* (AIP, Fermilab, USA, 2007) **896**, 31–49
[16] *GEF code website*, www.cenbg.in2p3.fr/-GEF-