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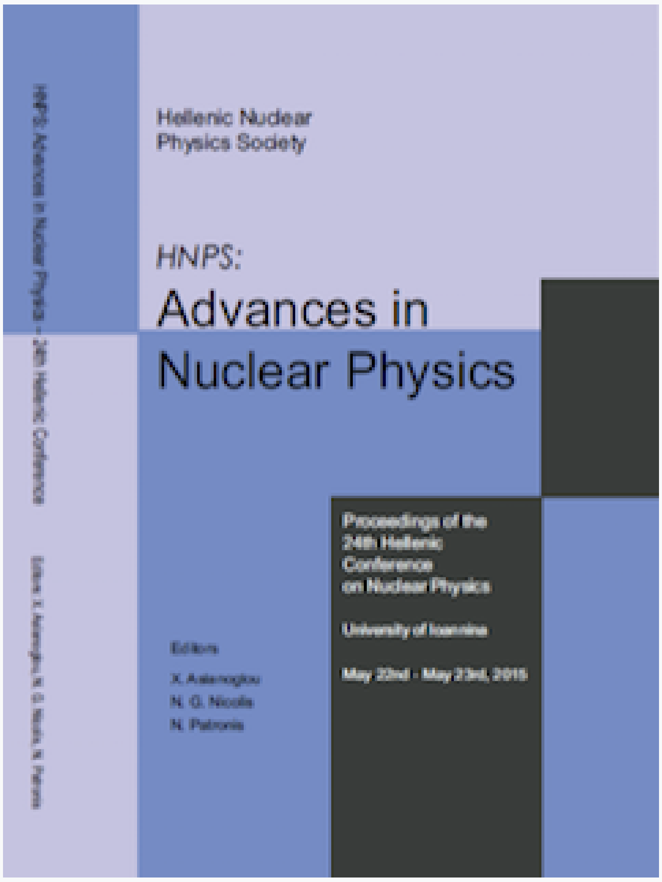
HNPS Advances in Nuclear Physics

Vol 23 (2015)

HNPS2015



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doi: 10.12681/hnps.1902

To cite this article:

Tsinganis, A., Stamatopoulos, A., Colonna, N., Vlastou, R., Schillebeeckx, P., Plompen, A., Heyse, J., Kokkoris, M., Barbagallo, M., Berthoumieux, E., Calviani, M., Chiaveri, E., & n_TOF Collaboration, and the. (2019). Neutron induced fission cross-section of 240Pu(n,f): first results from n_TOF (CERN) Experimental Area II. *HNPS Advances in Nuclear Physics*, *23*, 22–27. https://doi.org/10.12681/hnps.1902

Proceedings of the 24th Syposium of the HNPS

Neutron induced fission cross-section of ^{240}Pu(n,f): first results from n_TOF (CERN) Experimental Area II

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Abstract The accurate knowledge of neutron cross-sections of a variety of plutonium isotopes and other minor actinides, such as neptunium, americium and curium, is crucial for feasibility and performance studies of advanced nuclear systems (Generation-IV reactors, Accelerator Driven Systems). In this context, the ^{240}Pu(n,f) cross-section was measured with the time-of-flight technique at the CERN n_TOF facility at incident neutron energies ranging from thermal to several MeV. The present measurement is the first to have been performed at n_TOF's newly commissioned Experimental Area II (EAR-2), which is located at the end of an 18m neutron beam-line and features a neutron fluence that is 25-30 times higher with respect to the existing 185m flight-path (EAR-1), as well as stronger suppression of sample-induced backgrounds, due to the shorter times-of-flight involved. Preliminary results are presented.

Keywords n_TOF, neutron-induced fission, plutonium

INTRODUCTION

The accurate knowledge of neutron cross-sections of a variety of plutonium isotopes and other minor actinides is crucial for feasibility and performance studies of advanced nuclear systems [1-2]. Such isotopes, that present a fission threshold at a few hundred keV, accumulate during the operation of a conventional thermal reactor, but could be effectively transmuted in reactors with a fast neutron spectrum. Improved knowledge of the neutron-induced fission cross-sections of these isotopes is not only important for the design of advanced systems, but also for the more efficient operation of existing reactors, since safety margins can be more accurately defined. In particular, the non-fissile and long-lived ^{240}Pu isotope contributes to the long-term residual activity of nuclear waste. It is included in the Nuclear Energy Agency (NEA) High Priority List [3] and the NEA WPEC Subgroup 26 Report on the accuracy of nuclear data for advanced reactor design [4]. In this

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context, an experiment to measure the $^{240}Pu(n,f)$ cross-section was executed at the CERN n_TOF (neutron time-of-flight) facility [5-8]. Preliminary results are presented in this article.

EXPERIMENTAL SETUP

The n_TOF facility and Experimental Area II (EAR-2)

Neutrons at n_TOF are spallation products created by a bunched 20GeV/c proton beam delivered by CERN's PS (Proton-Synchrotron) accelerator onto a lead target 40cm in length and 60cm in diameter. A 1 cm-thick layer of circulating water surrounds the target in order to cool it down and also act as a neutron moderator. Beyond the target and after an additional 4cm-thick layer of (borated) water, a ~185 m vacuum tube leads to the first measuring station (Experimental Area I, or EAR-1) which has been in operation since 2001.

A new experimental area (Experimental Area II or EAR-2) [9-10] was commissioned in the second half of 2014. EAR-2 is located at the end of an 18m neutron beam-line placed vertically above the spallation target. The proximity to the target yields a gain in flux of 25-30 times compared to the existing experimental area (EAR-1), while the neutrons are delivered in an approximately 10 times shorter time interval. The very high instantaneous flux and extended energy range (from thermal to over 100MeV) allow to cover the region of interest in a single experiment and mitigate the adverse effects of the strong α-background produced by the samples and the low fission cross-section below and near the fission threshold.

Both experimental areas meet the requirements to operate as Type A Work Sectors [11], meaning unsealed radioactive samples can be handled.

Samples and detectors

Three plutonium oxide (PuO_2) samples were used [12], for a total mass of approximately 2.3mg of ^{240}Pu (~0.10 mg/cm^2 per sample, 99.89% purity). The material was electro-deposited on an aluminium backing 0.25mm thick and 5cm in diameter, while the deposit itself had a diameter of 3cm. Additionally, a ^{235}U sample with a mass of ~0.6mg and a ^{238}U sample with a mass of ~0.8mg were used as reference. All samples were manufactured at IRMM (Belgium). Contaminants present in the ^{240}Pu samples (most notably ^{239}Pu) have a non-negligible contribution to the fission yield in certain energy ranges (mainly below 1keV) which was subtracted during the analysis.

The measurements were carried out with Micromegas (Micro-MEsh GAseous Structure) gas detectors [13-16]. The gas volume of the Micromegas is separated into a charge collection region (several mm, 5mm in this case) and an amplification region (typically tens of μm, 50μm in this case) by a thin "micromesh" with 35μm diameter holes on its surface. A chamber capable of holding up to 10 sample-detector modules was used for the measurement. The detectors were operated with an $Ar:CF_4:isoC_4H_{10}$ gas mixture (88:10:2) at a pressure of 1 bar. A picture of the chamber placed in the experimental area is shown in Figure 1.

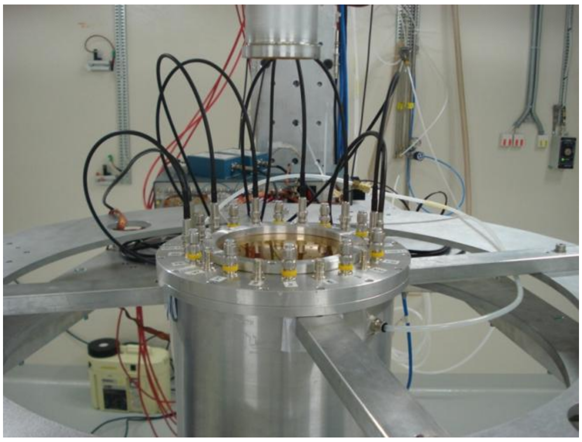

Fig. 1. A view of the chamber housing the samples and detectors, and the associated electronics placed in the neutron beam in EAR-2. The neutron beam arrives vertically from below and then continues in the vacuum tube above the chamber and onto the beam dump a few meters later. Approximately 15-20cm of air are present before and after the chamber.

The analogue detector signals were digitised with 8-bit flash-ADCs [17] with a 500 MHz sampling rate. In order to minimise the volume of data to be transferred and recorded, a zero-suppression algorithm was applied to avoid recording long sequences of noise where no useful signals are present.

THE PREVIOUS MEASUREMENT IN EAR-1

This measurement was originally attempted in EAR-1, in parallel with the measurement of the ^{242}Pu fission cross-section [18-19]. Due to the lower neutron flux, it was necessary to measure over a period of several months to collect the necessary statistics. An unexpected effect of the high α-activity of the ^{240}Pu samples (>6 MBq per sample) was encountered in the course of the measurement [20], with a steady degradation of the fission fragment amplitude distribution. After the end of the measurement, a visual inspection of the detectors used with the ^{240}Pu samples revealed a circular discolouration of the mesh whose dimension and position exactly matched those of the samples. Upon closer inspection with a microscope, it became clear that the micromesh had suffered serious damage, particularly around the rims of the holes which were evidently deformed (Figure 2). This lead to a degradation of the electrical field and therefore of the detector gain and overall performance. In time, this made the fission fragment and α-particle signals virtually indistinguishable in the obtained pulse-height spectra. It should be noted that, even when detectors were operating normally, the long α pile-up tail greatly reduced the quality of the separation, as can be seen in Figure 3 (black).

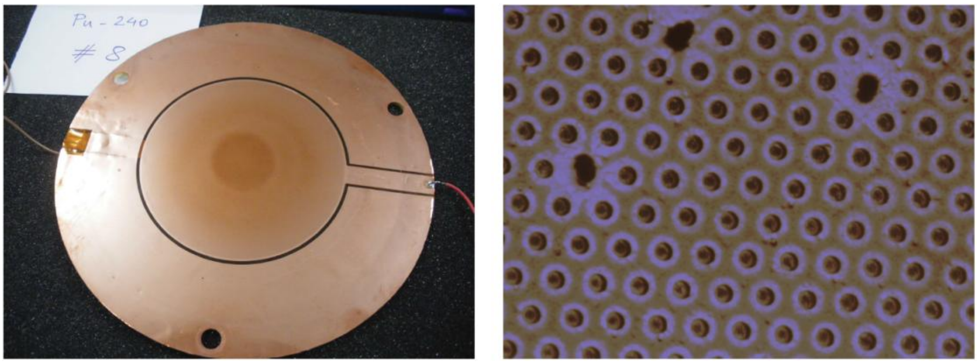

Fig. 2. Left: One of the Micromegas detectors used with a ^{240}Pu sample pictured after the end of the measurement. A 3cm diameter discolouration is visible on the micromesh. Right: Picture of the micromesh taken with an electronic microscope. Mechanical damage around the rims of the holes can be observed. This leads to a severe deterioration of the detector gain and performance.

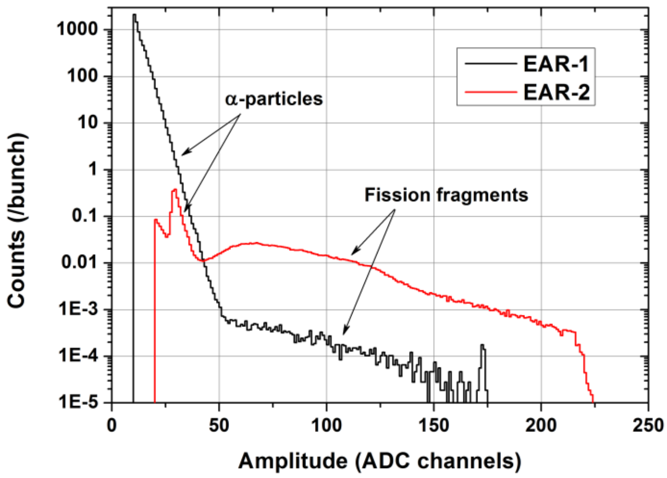

Fig. 3. Pulse-height spectra obtained from a ^{240}Pu sample during the measurements in EAR-1 (black) and EAR-2 (red). Counts are normalised per beam bunch for direct comparison. The significant suppression of the sample-induced α-background in EAR-2 is evident, as is the much higher rate of fission events.

DATA ANALYSIS AND RESULTS

The digitised raw data from each detector are analysed off-line by means of a pulse recognition routine that determines the amplitude and position in time of the detected signals, as well as the signal baseline, among other quantities [21]. Throughout the measurement, beam-off data were taken in order to record the α- and spontaneous fission background produced by the samples. The behaviour of the detectors is studied by means of Monte Carlo simulations performed with the FLUKA code [22-23], focusing particularly on the reproduction of the pulse height spectra of α-particles and fission fragments for the evaluation of the detector efficiency and the correction associated to the signal amplitude threshold.

The interactions of the proton beam with the spallation target lead to a significant production of prompt γ-rays and other relativistic particles that reach the experimental area at (nearly) the speed of light and constitute what is commonly termed the “γ-flash”. This causes an initial signal lasting a few hundred ns, followed by a baseline oscillation that lasts for several μs or, in terms of neutron energy, down to less than 1 MeV. In order to remove this oscillation, an average shape is obtained from at least several hundred signals and then subtracted from each raw data “movie” before processing the data.

In Figure 3, the pulse-height spectrum obtained from one of the ^{240}Pu samples (red) is compared to a spectrum recorded with the same sample during the measurement in EAR-1 (black). It can be clearly observed that the increased neutron flux leads to a considerably higher number of recorded fission events and that there is a much stronger suppression of the sample-induced α-particle background, resulting in a considerably clearer separation from the fission fragments.

Figure 4 shows the fission counts as a function of incident neutron energy recorded in all three ^{240}Pu samples between 10 eV and 30 keV, after applying an appropriate signal amplitude threshold to reject the background and subtracting the contribution of contaminants present in the sample. Several resonance clusters can be observed, attributable to the coupling of Class-I and Class-II states, and resonances are visible up to a few tens of keV. Above 100 keV, data has been obtained up to at least several MeV, with statistical uncertainties below 2-3% and even below 1% in the range between 0.5-5 MeV.

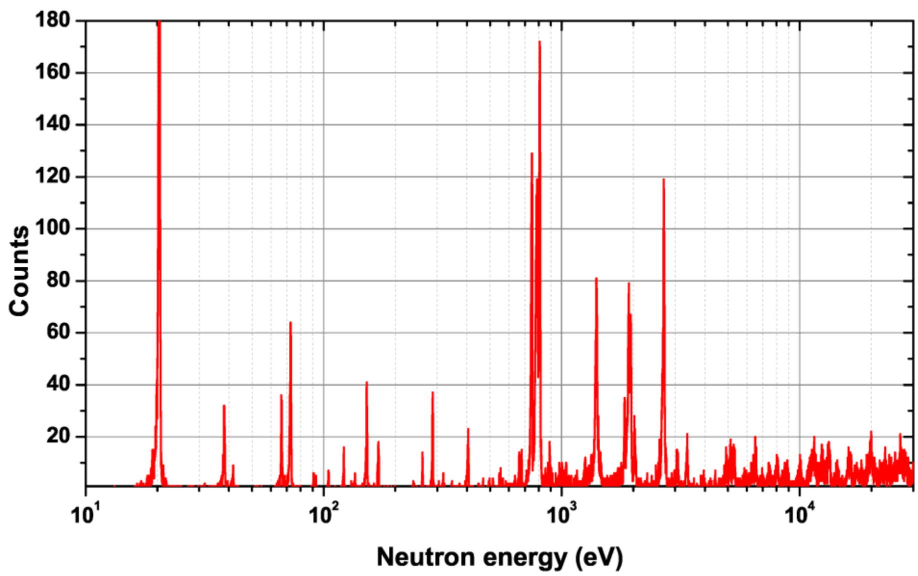

Fig. 4. Resonances observed in the measured fission yields from the three ^{240}Pu samples, after subtraction of contributions from contaminants. Several resonance clusters, attributable to the coupling between Class-I and Class-II states, are visible, while resonances can be observed up to a few tens of keV. Data are shown with a binning of 2000 bins per energy decade.

SUMMARY

The measurement of the ^{240}Pu fission cross-section is the first measurement to be performed at the newly commissioned Experimental Area II of the CERN n_TOF facility. Data were collected from thermal energies up to at least several MeV. Most notably, data showing clear resonant structures have been obtained even in the sub-threshold region (up to a few tens of keV) where the cross-section is lowest and where evaluations show a smooth behaviour of the cross-section. The success of this measurement is largely due to the favourable characteristics of EAR-2, in particular the increased neutron flux and stronger background suppression compared to EAR-1, where the measurement was not feasible. These features will allow n_TOF to expand its measurement capabilities to even more short-lived and rare isotopes, such as ^{230}Th, ^{232}U, ^{238,241}Pu and ^{244}Cm.

References

[1] Generation-IV International Forum, www.gen-4.org
[2] International Framework for Nuclear Energy Cooperation, www.ifnec.org
[3] NEA Nuclear Data High Priority Request List, www.nea.fr/html/dbdata/hprl
[4] OECD/NEA WPEC Subgroup 26 Final Report, www.nea.fr/html/science/wpec/volume26/volume26.pdf
[5] U. Abbondanno et al., CERN-SL-2002-053 ECT
[6] C. Guerrero et al., Eur. Phys. J. A 49:2, 1-15 (2013
[7] E. Berthoumieux et al., CERN-n_TOF-PUB-2013-001
[8] M. Barbagallo et al., Eur. Phys. J. A 49:12, 1-11 (2013)
[9] E. Chiaveri et al., CERN-INTC-2012-029. INTC-O-015
[10] C. Weiss et al., Nucl. Instrum. Meth. A 799, 90-98 (2015)
[11] V. Vlachoudis, 2012, CERN EDMS No. 934369
[12] G. Sibbens et al., J. Radioanal. Nucl. Ch. 299, 1-6 (2013)
[13] Y. Giomataris et al., Nucl. Instrum. Meth. A 376(1), 29-35 (1996)
[14] Y. Giomataris, Nucl. Instrum. Meth. A 419, 239-250 (1998)
[15] S. Andriamonje et al., J. Instrum. 5:02 (2010)
[16] S. Andriamonje et al., J. Korean Phys. Soc. 59:23, 1597 (2011)
[17] U. Abbondanno et al., Nucl. Instrum. Meth. A 538, 692-702 (2005)
[18] A. Tsinganis et al., Nucl. Data Sheets 119, 58-60 (2014)
[19] A. Tsinganis et al., EPJ Web of Conf. 66, 03088 (2014)
[20] Varii, European Commission, Joint Research Centre, Belgium (2013), DOI: 10.2787/81004 (Chapter 3)
[21] P. Zugec et al., Nucl. Instrum. Meth. A 812, 134-144 (2016)
[22] A. Ferrari et al., CERN, Geneva (2005), DOI: 10.5170/CERN-2005-010
[23] G. Battistoni et al., AIP, Fermilab, USA (2007), vol. 896, (pp. 31–49)