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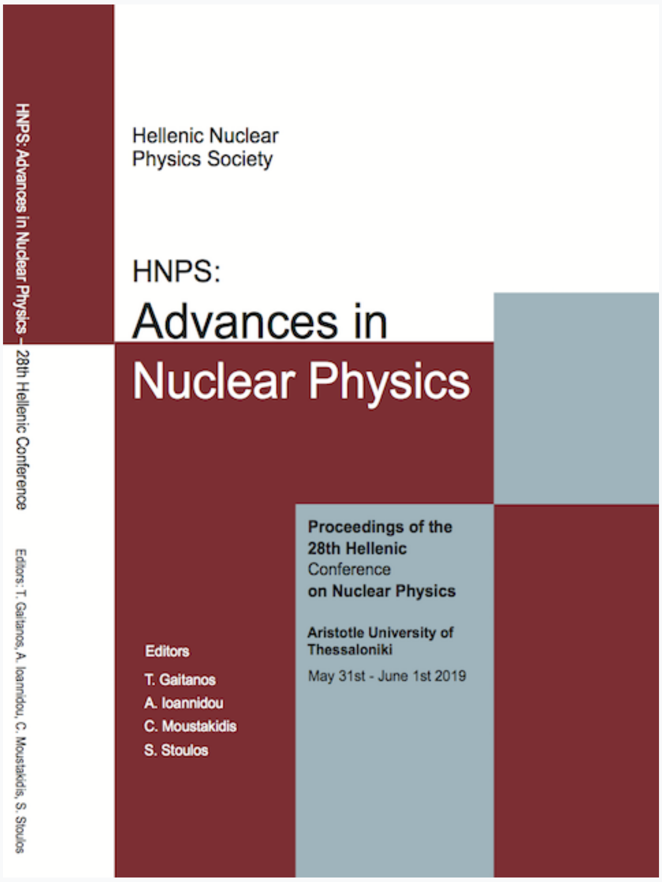

Cross section measurement of 241Am(n,f) reaction at the Experimental Area 2 of the n_TOF facility at CERN: First Results

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Cross section measurement of ^{241}Am(n,f) reaction at the Experimental Area 2 of the n_TOF facility at CERN: First Results

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Abstract *The condition for the safe design and operation of fast neutron reactors and energy boosters (Generation-IV reactors, ADS systems [1]) is the accuracy of nuclear data. The ^{241}Am isotope ($T_{1/2}$ = 433 years) is highly present in nuclear waste, accounting for about 1.8% of the actinide mass in PWR UOx nuclear reactors' waste [2]. In addition, the ^{241}Am isotope is further produced by the β decay of the ^{241}Pu isotope ($T_{1/2}$ = 14.3 years). Given the high production rate of ^{241}Am isotope, its incineration with concurrent energy production is considered to be of utmost importance for the design and implementation of the recycling of existing nuclear waste. Sensitivity studies of the proposed systems for energy production showed that high-precision measurements of the cross section of the ^{241}Am(n,f) reaction are required. In the present work, the ^{241}Am(n,f) reaction cross section was measured in the Second Experimental Area of the n_TOF facility at CERN, using an array of Micromegas detectors. For the measurement, six targets of ^{241}Am with average activity of 17 MBq per sample were coupled with an equal number of detectors in a common chamber. Additionally two ^{235}U and two ^{238}U samples were coupled with Micromegas detectors utilizing the neutron flux determination. Within this work, an overview of the experimental set-up and the adopted data analysis technique is presented along with preliminary results.*

Keywords *Americium-241, Fission, Time-Of-Flight technique, Micromegas detectors, n_TOF-CERN*

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INTRODUCTION

Minor actinides are responsible for a large fraction of the long-term radiotoxicity of the waste from conventional nuclear reactors. They are mainly produced by successive neutron capture reactions and alpha decays derived from 238U nuclei that are present in the nuclear fuel. It is commonly accepted by the scientific community that the incineration/transmutation of the existing actinides in high-level waste from PWR UOx fuel [3] can be addressed through the development of fast neutron reactors and energy boosters (Generation-IV Reactors, ADS systems). A needful condition for the safe design and operation of these systems is the accuracy of nuclear data.

One of the most important fissionable isotopes to be considered as a potential candidate for incineration/transmutation is 241Am (T1/2 = 433 years) that represents almost 1.8% of the actinide mass in spent fuel. Its production rate within spent fuels increases further due to the β- decay of 241Pu isotope (T1/2 = 14.3 years). Due to the fact that the information of the fission rate of 241Am(n,f) is needed for an extended energy range, this reaction is included in the Nuclear Energy Agency (NEA) "High Priority Request List" (HPRL) [4].

The aforementioned measurement is challenging due to the high specific activity of ^{241}Am (127 Mbq/mg) and its rapidly increasing cross section above the fission threshold. A previous measurement at n_TOF in 2003 [5] was performed in the first experimental area (EAR-1) [6] and provided results that were in good agreement with the existed ones but they were suffering from low statistics. Therefore, the results were given only above the fission threshold and in a rather coarse energy grid.

The construction of the second experimental area (EAR-2) [7] at the n_TOF facility [8] resulted in an enhanced neutron beam intensity and provided much better signal to background conditions than EAR-1. Consequently, high accuracy cross section data can be obtained even at energies below the fission threshold also for high activity samples. Within the present work the study for the fission reaction of ^{241}Am cross section was taken over from the n_TOF Collaboration in EAR-2.

EXPERIMENTAL DETAILS

The n_TOF facility

The measurement was performed at the n_TOF facility at CERN, in the second experimental area (EAR-2) which was commissioned in July 2014. The n_TOF is a Time-Of-Flight facility based on a spallation neutron source. The neutron beam is produced when a pulsed beam of 20 GeV/c protons delivered from the Proton Synchrotron accelerator (PS) of CERN, impinges on a cylindrical lead target with dimensions 40 x 60 cm^2 (length x diameter). The proton beam has a time width of 7 ns RMS with a maximum repetition rate of 0.8 Hz (1.2 sec between consecutive bunches). The proton accelerator delivers dedicated pulses with a nominal value of ~7x10^{12} protons/bunch and parasitic ones with reduced intensity by a factor of 2.

The lead target is surrounded by a 1 cm thick layer of water in constant circulation that serves as coolant and moderator. The first experimental area (EAR-1) is located at 185 m distance horizontally with respect to the lead target, while the second one (EAR-2) lies approximately 20 m above the spallation target and vertical to the direction of the proton beam. Due to its shorter flight path, EAR-2 provides up to 40 times higher neutron flux compared to EAR-1.

The enhanced neutron flux [9] of EAR-2 allows the minimization of the sample thickness while preserving high counting rates. The unique combination of high instantaneous flux and high background suppression, provided in EAR-2, allow accurate fission cross section measurements from thermal energies up to the MeV region.

Samples

The targets used in the measurement were fabricated at JRC- Geel (Belgium). Six samples of ^{241}Am (99.98% purity) were used with a total mass of 0.78 mg (~4.6 μg/cm^2 per sample) and an activity of ~0.1 GBq. Additionally, for the determination of the neutron flux, two ^{235}U (0.26 mg, 0.30 mg) and two ^{238}U (2.07 mg, 2.21 mg) samples were used as reference foils. In all cases, the sample material was electroplated in a surface 6 cm in diameter on top of an aluminum backing 0.025 mm thick. The sample-backing configuration was mechanically supported by a 2 mm thick aluminum ring 10 cm in diameter.

Detectors

The measurement was carried out using an array of Micromegas detectors (Micro-Mesh Gaseous Structure) which are parallel plate avalanche gaseous detectors that consist of two regions: the conversion (7 mm) and the narrow amplification region (50 μm) [10]. In the conversion region, which is also referred in literature as drift region, the ionization takes place and charge carriers are directed towards the amplification region where an avalanche multiplication occurs due to the high electric field applied. The two regions are separated by a thin micro-mesh foil, 9.5 cm in diameter with 60 μm holes on its surface. This is the active area of the detector which is by far larger than the sample diameter (6 cm) as to avoid efficiency losses.

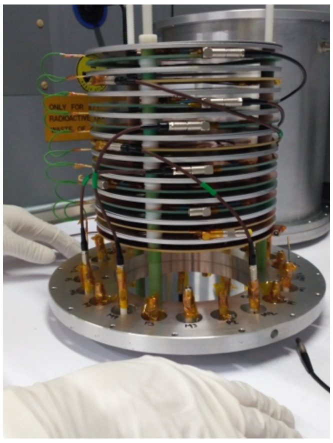

Figure 1. *The stack of samples and detectors before mounting inside the fission chamber.*

In total ten detectors were used for the measurement: six for the americium and four for the uranium reference samples. The sample-detector modules (Fig.1) were housed in a cylindrical aluminum fission chamber that was filled with a gas mixture of Ar:CH_4:isoC_4H_{10} (88:10:2) at atmospheric pressure. During the measurement, the gas flow and pressure were monitored and controlled with a dedicated flow-regulator system. In this way, the gas pressure was kept constant ensuring stable gain conditions during the whole data-taking period.

Data Analysis

At n_TOF the detector signals are stored as waveforms and digitized. In this way, the characteristics of interest, such as the amplitude, the timing information, the risetime and the full width could be extracted. The first step in the data analysis procedure is the processing of raw data of the digitized pulses ("movies"). In this stage, a pulse shape analysis routine was used [11] where the basic characteristics of each pulse are recorded in an event-by-event mode. In this way, the parameters such as the time-of-flight, the amplitude, the rising time, the full-width-at-half-maximum as well as other parameters (33 in total) for each recorded pulse are saved in highly compressed files to be used in the next steps of the data analysis procedure.

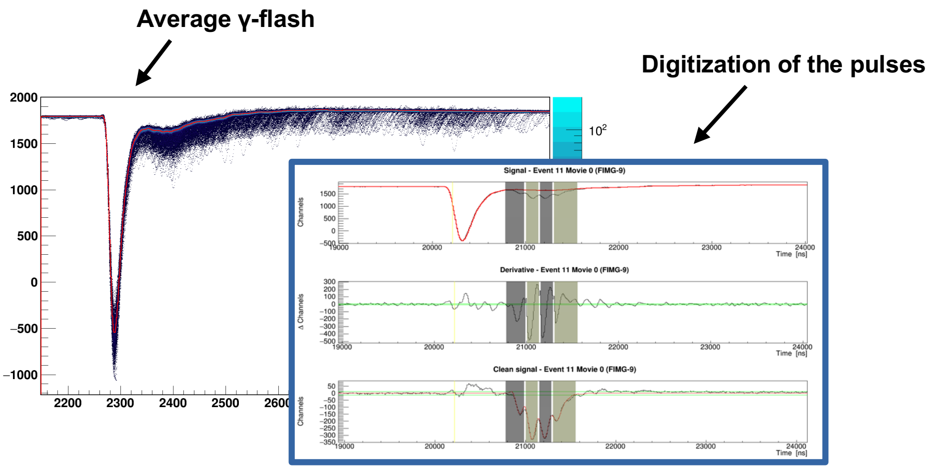

Figure 2. *Subtracted average γ-flash and digitization of the pulses*

The interaction of the proton beam with the spallation lead target results in the generation of γ−rays and other relativistic particles that reach the experimental areas prior to the arrival of the neutrons. This is commonly referred as "γ−flash"and causes the creation of a high amplitude signal with a width of a few hundreds of ns. The γ−flash is accompanied by small oscillations that are diminished after the first μs. In energy terms this corresponds to 1-2 MeV. For this reason, a special routine was developed [12] where the average γ−flash shape for each detector is extracted and then subtracted from the raw data. In this way, pulses generated from fission fragment events could be reconstructed for neutron energies up to a few MeV.

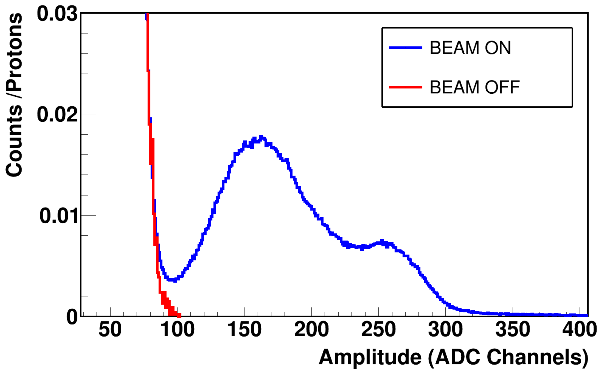

Figure 3. *An example of beam-on (blue) and beam-off (red) pulse height spectrum for one of the ^{241}Am targets. As can be seen, the alpha-decay particles are well separated from the fission fragments but still an amplitude threshold has to be applied in the analysis along with the corresponding correction factor.*

The second step in the analysis procedure is the accurate determination and implementation of the needed correction factors to the fission yields, in order to accurately estimate the ^{241}Am fission cross section in an extended energy range. The most important correction factor is the one corresponding to the amplitude cut, applied to the pulse height spectrum so as to isolate the fission events from the alpha particles, attributed to the decay of ^{241}Am. In this respect, extensive Geant4 [13–15] simulations will be performed to characterize the experimental pulse height spectrum and correct for the applied amplitude cut. To estimate the amplitude threshold, beam-off data were collected as well (Fig.3) during the data-taking period. From these measurements the gain-stability of the detectors was also confirmed.

RESULTS AND DISCUSSION

The excitation function of the ^{241}Am(n,f) reaction was determined using the ^{235}U(n,f) and ^{238}U(n,f) reference reactions for the neutron flux estimation. Preliminary results of the experimental cross section as derived from the weighted average cross section between the six targets of americium can be seen in Fig. 4 for the whole energy range covered within the present work.

In Fig. 5 preliminary cross section at near threshold energies is presented along with the available data libraries. Given that the calculation of the needed correction factors is still in progress the data were normalized to the aforementioned data libraries at 1.2 MeV neutron energy. Overall, the trend of the excitation function is in good agreement with the aforementioned data bases and the achieved statistical uncertainty is low enough for fulfilling the accuracy requests for this energy region according to the OECD/NEA WPEC Subgroup 26 Final Report [16].

CONCLUSIONS

The ^{241}Am(n,f) cross section was measured in the second experimental area EAR-2 of the n_TOF facility at CERN with Micromegas detectors. Data are obtained for an extended energy range from thermal up to the MeV region. Preliminary results at near threshold energy region show a good agreement with the available evaluated libraries. The upcoming steps of the data analysis are the accurate determination of the correction factors (amplitude cut, dead-time, possible impurities, etc) so as to calculate the cross section fulfilling the precision requirements for future developments of nuclear energy applications.

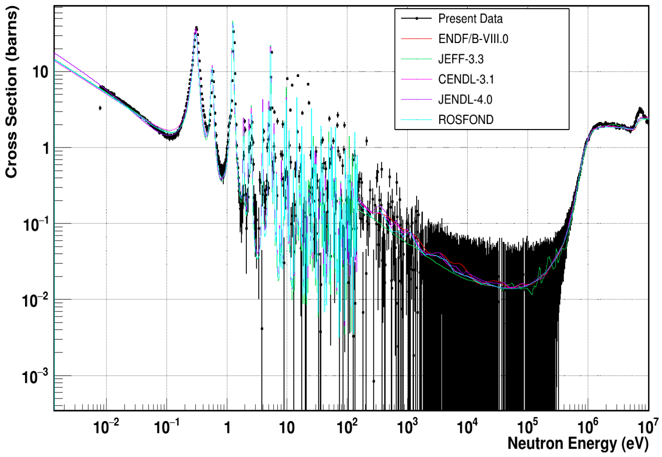

Figure 4. *Experimental Data of the present work for the $^{241}Am(n,f)$ reaction covering the energy range from a few meV up to the MeV region for 60% of the full statistics.*

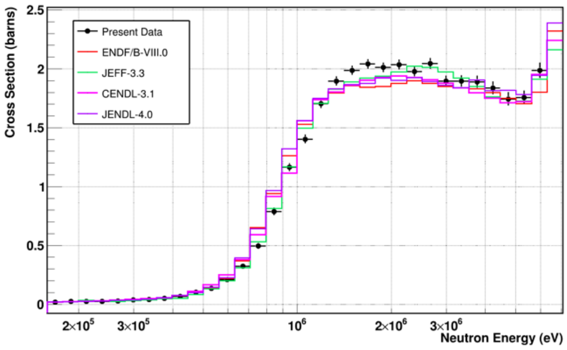

Figure 5. *Preliminary $^{241}Am(n,f)$ cross section derived from the weighted average from the americium samples. The error bars represent the statistical uncertainty. The experimental data are normalized to the evaluated libraries at 1.2 MeV for comparison.*

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