Measurement of the ²⁴²Pu(n,f) cross section at n_TOF

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Abstract. Knowledge of neutron cross sections of various plutonium isotopes and other minor actinides is crucial for the design of advanced nuclear systems. The 242 Pu(n,f) cross sections were measured at the CERN n_TOF facility, taking advantage of the wide energy range (from thermal to GeV) and the high instantaneous flux of the neutron beam. In this work, preliminary results are presented along with a theoretical cross section calculation performed with the EMPIRE code.

1 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]. In particular, the non-fissile and long-lived ²⁴²Pu 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 context, the ²⁴²Pu(n,f) cross section was measured at n_TOF relative to the well-known ²³⁵U(n,f) cross section. Preliminary results are presented in this work.

2 EXPERIMENTAL SETUP

The measurements were performed at the CERN n_TOF facility [5], where neutrons from thermal and up to over 10 GeV are produced through spallation induced by a bunched proton beam impinging on a massive lead target and travel along an approximately 185 m path to reach the experimental area. These characteristics, along with the high instantaneous flux, 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.

Four plutonium oxide (PuO₂) samples were used [6], for a total mass of 3.6 mg of 242 Pu (~0.13 mg/cm², 99.97% purity). The material was electro-deposited on an aluminium backing 0.25 mm thick

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Figure 1. Top panel: The beginning (first few μ s) of the recorded signals during the same proton bunch from two adjacent detectors. The γ -flash signal and the baseline oscillations are clearly visible. Bottom panel: the residual signal after the subtraction of the two signals above. The oscillation is almost entirely suppressed.

and 5 cm in diameter, while the deposit itself had a diameter of 3 cm. Additionally, a 235 U sample (UF₄) with a mass of 3.3 mg deposited on a 0.2 mm thick aluminium backing was used as reference (~0.47 mg/cm²).

The measurements were carried out with Micromegas (Micro-MEsh GAseous Structure) gas detectors [7]. The gas volume of the Micromegas is separated into a charge collection region (several mm) and an amplification region (typically tens of μ m) by a thin "micromesh" with 35 μ m diameter holes on its surface. A chamber capable of holding up to 10 sample-detector modules was constructed and used for the measurement. The detector was operated with an Ar:CF4:isoC4H₁₀ gas mixture (88:10:2). The shielding of the pre-amplifier module was improved to mitigate the baseline oscillation observed following the prompt γ -flash.

3 ANALYSIS AND RESULTS

3.1 Data analysis

The raw data from each detector, digitally recorded at a sampling rate of 100 MHz with 8-bit Acqiris flash-ADCs, are analysed by means of a pulse recognition routine that determines the amplitude and position in time of the detected signals, among other quantities. The signal baseline is determined by analysing the pre-trigger and post-acquisition window data. Since the Pu samples are in the same chamber as the ²³⁵U it can be assumed that they receive the same neutron flux, while the fission count rates are sufficiently low to ignore pile-up effects. 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 was studied by means of Monte Carlo simulations performed with the FLUKA code [9, 10], 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 quality of the peak-search routine.

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 μ s or, in terms of neutron energy, down to 1-2 MeV (Fig. 1, top panel). This problem can be mitigated by applying a software "compensation" technique [8] to the digitally recorded data. This method is based on the observation that the oscillations recorded in adjacent detectors for the same proton bunch are almost identical, as can be seen in Fig. 1. The subtraction of the two signals causes the oscillations to largely cancel each other out, leaving a residual signal that consists of pulses attributable either to fission fragments or

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Figure 2. The first ²⁴²Pu resonance at 2.7 eV (top left panel) and resolved resonances between 700 and 800 eV (top right) and around 1800 eV (bottom left). Data above the fission threshold (bottom right). The experimental data are normalized to the ENDF cross-section for comparison.

 α -particles (Fig. 1, bottom panel). This signal is then analysed with the peak search routine used for the lower energy region, thus extracting the desired pulse height spectra.

3.2 Results

The spontaneous fission background dominates the low energy region and remains visible up to about 1 keV. Still, several resonances can be observed above this background. The first 242 Pu resonance at ~2.7 eV can be seen in the top left panel of Fig. 2, after subtraction of the spontaneous fission background, as determined with a fit of the beam-off data, and normalization to the ENDF cross section. The top right and bottom left panels panel show resolved resonances up to approximately 1900 eV, including one at ~780 eV and one at 1830 eV not present in the evaluated libraries and, at a preliminary analysis, not attributable to any of the stated sample impurities. Data above 1 keV are shown in the bottom right panel of Fig. 2. The data displayed are combined from the two analysis methods; the conventional "straightforward" analysis, which fails above about 1 MeV due to the baseline oscillations, and the high-energy analysis described in section 3.1. The analysis of the high energy region will be extended at least up to 100 MeV. The data presented here correspond to approximately 60% of the total accumulated statistics.

4 THEORETICAL CALCULATIONS

A theoretical calculation of the 242 Pu(n,f) cross section was performed with the EMPIRE nuclear reaction model code [11] (version 3.1). The level densities of the nuclei involved in the calculations

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Figure 3. Theoretical calculation of the 242 Pu(n,f) cross section performed with the EMPIRE code, with experimental data retrieved from the EXFOR database.

were treated within the framework of the Enhanced Generalised Superfluid Model (EGSM). Initial values for the fission barrier parameters (barrier height and width) were retrieved from the RIPL-3 library [12] and subsequently adjusted to better reproduce the experimental data. Preliminary results can be seen in Fig. 3.

5 CONCLUSIONS

Preliminary results from the ²⁴²Pu(n,f) experiment performed at the CERN n_TOF facility are presented. The experimental setup and analysis method is described, including auxiliary Monte-Carlo simulations and an off-line technique to recover high-neutron energy data affected by the prompt γ -flash. Preliminary theoretical calculations performed with the EMPIRE code are also presented.

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