EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the fission cross-section of ^{241}Am

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Abstract

We propose to measure the neutron-induced fission cross-section of ^{241}Am in EAR-2 of the n_TOF facility. The isotope ^{241}Am is important for different waste transmutation and recycling scenarios. The present experimental uncertainties do not meet the requirements set by sensitivity studies of advanced nuclear systems. The aim of this experiment is to produce accurate data at the fission threshold and up to several MeV, as well as for selected resonances in the resolved resonance region. The characteristics of EAR-2, namely the stronger background suppression by about a factor of 10 and the 10-30 times higher neutron flux with respect to EAR-1 (with an additional factor of about 10 with the use of the large collimator) will allow to address the two main challenges associated with this measurement: the high α-activity of the samples and the rapidly decreasing cross-section below the fission threshold.

Requested protons: $3x10^{18}$ protons on target
Experimental Area: EAR-2

1. Introduction

1.1 Motivation

Isotopes of several transuranic elements are present in large quantities in nuclear waste. They are produced inside the reactor through a sequence of neutron capture reactions and α-decays starting mainly from the ^{238}U present in the fuel and include plutonium, neptunium, americium and curium isotopes. Actinides are responsible for the long-term radiotoxicity of nuclear waste, which remains important for over one hundred thousand years.

Feasibility, design and sensitivity studies on new generation reactors (ADS, Generation IV) that could address the issue of transmutation of nuclear waste using fuels enriched in minor actinides require high accuracy cross-section data for a variety of neutron-induced reactions from thermal energies to several tens of MeV. The NEA (Nuclear Energy Agency) "Nuclear Data High Priority Request List" [1] lists data requests from different fields, including but not limited to advanced reactor design, while the OECD/NEA WPEC Subgroup 26 Final Report [2] summarises the needs and target accuracies for nuclear data relevant for advanced nuclear systems.

The isotope ^{241}Am is present in high-level nuclear waste, representing about 1.8% of the actinide mass in spent PWR UOx fuel [3]. Its importance increases due to additional production from the beta-decay of ^{241}Pu with a half-life of 14.3 years. It is clear that both the production of ^{241}Am in conventional reactors (including its further accumulation through the decay of ^{241}Pu) and its destruction through transmutation are very important for the design of any recycling solution.

Sensitivity analyses of proposed advanced systems have shown that the accuracy requirements for the $^{241}Am(n,f)$ cross-section are very tight [1,2,4]. Present uncertainties and target uncertainty requirements for the neutron-induced fission cross-section of ^{241}Am are summarised in Table 1.

Table 1: *Initial and target uncertainties of the $^{241}Am(n,f)$ cross-section [1,2].*

	Uncertainty (%)	
Range	**Initial**	**Target**
19.6 - 6.07 MeV	12.7	5.7
6.07 - 2.23 MeV	11.7	1.7
2.23 - 1.35 MeV	9.8	1.4
1.35 - 0.498 MeV	8.3	1.2
183 - 498 keV	8.3	4.0

1.2 Present status of data

Evaluations of actinide cross-sections are often limited by the availability of experimental results, both in terms of their accuracy and their coverage of isotopes and reactions of interest [3]. In particular, measurements of the fission cross-section of ^{241}Am are challenging for two main reasons; firstly, due to its relatively short half-life of 433 y, ^{241}Am has a specific activity of 127 MBq/mg. This strong α-particle background limits the mass of the samples that can be used in a measurement. Secondly, the fission cross-section displays a steep threshold with its value decreasing rapidly below 1 MeV, reaching 100 mb already at 500 keV incident neutron energy.

For these reasons, there are still significant discrepancies among available datasets and evaluated libraries, which mostly follow the data by Dabbs et al. [5]. In Figure 1, the available experimental data between 500 keV and 20 MeV are shown, with an important spread of the data even at the fission plateau. The present uncertainties remain too high for the needs of applications, as shown in Table 1.

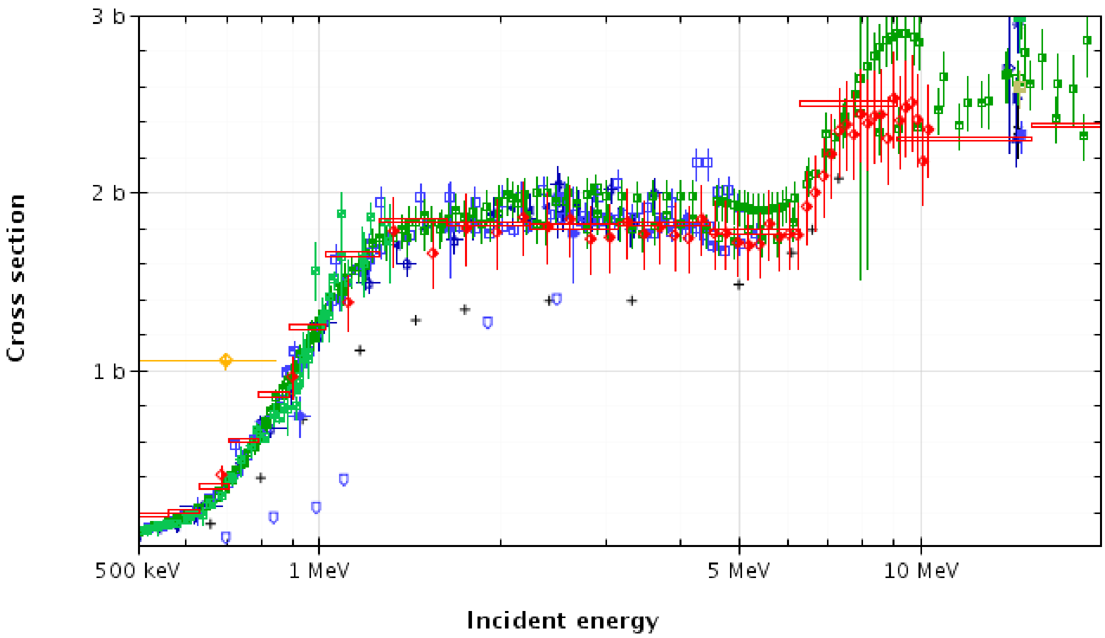

Figure 1: *Previous measurements of the ^{241}Am(n,f) cross-section above 500 keV (retrieved from the EXFOR database).*

1.3 Previous measurement at n_TOF and the case for EAR-2

A measurement of the ^{241}Am(n,f) cross-section was already performed at n_TOF in 2003 (EAR-1) [6] (proposal INTC-2003-021 [7]). The statistics of that experiment were limited due to the mass used (2.3 mg), the high pulse-height threshold that had to be set in order to reject the sample-induced α-background and the low cross-section near the fission threshold. A variable energy bin width was chosen so as to maintain the statistical uncertainty in each bin below 2%, thus leading to a very coarse energy resolution, essentially providing an average cross-section value. Using an even lower mass of material in EAR-2, the signal-to-background ratio can be improved by about a factor of 10, while the available neutron flux is 30 times higher. These favourable conditions will allow obtaining data with significantly better energy resolution and down to lower energies below the fission threshold, while mitigating the effects of the α-background.

2. Experimental setup

2.1 The Micromegas detectors

For neutron measurements, it is of particular importance to minimise the amount of material present in the beam in order to reduce background related to scattered neutrons,

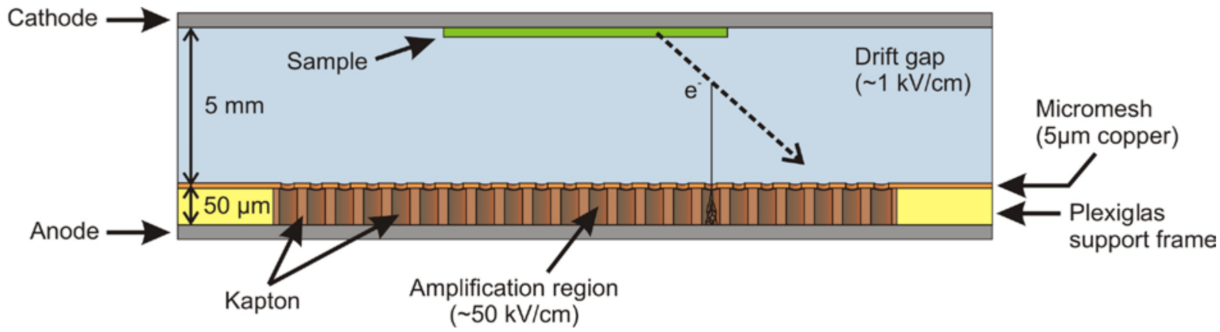

Figure 2: *An illustration of the basic principle of operation of a Micromegas detector. An ionising particle emitted from a sample ionises the gas. The ionisation electrons drift towards the micromesh and are multiplied inside the amplification region before being finally collected on the anode. Indicative values are given for the electrical field strength values and dimensions of the two regions.*

as well as to avoid perturbing the neutron flux. To this end, the *microbulk* design [8,9] was developed, based on the *Micromegas* principle (Fig. 2). This design has been utilised in past measurements performed at n_TOF. Microbulk detectors are composed of very thin layers of material, thus minimising neutron scattering. A chamber that can house up to 10 sample-detector modules is available. The chamber is filled with an Ar:CF_4:isoC_4H_{10} gas mixture (88:10:2), which is commonly used at CERN and has excellent timing properties due to the relatively high electron drift velocity. The drift gap between the sample (cathode) and the detector (anode) will be reduced from 5 mm used in previous measurements to 3 mm (without significant deterioration of the alpha – fission fragment separation) in order to reduce the signal width and improve the timing resolution.

2.2 Samples and sample holders

We plan to use six AmO_2 samples, for a total mass of 0.85 mg of ^{241}Am (~5 µg/cm^2 per sample). The distribution of the mass over six samples will allow reducing the individual activity. The new samples will be prepared at IRMM, Geel. The material will be electro-deposited on an aluminium backing 25 µm thick and 10 cm in diameter, while the deposit itself will have a diameter of 6 cm. Information on possible impurities in the Am samples that could contribute to the measured fission events, such as ^{242m}Am and ^{243}Am, is not yet available but will be provided by IRMM as soon as the batch of material to be used is identified. Additionally, samples of ^{10}B (~5 µg/cm^2), ^{235}U (~10 µg/cm^2) and ^{238}U (~70 µg/cm^2) will be used as reference.

The samples need to be adapted to and mounted on the interior frame of the detector chamber. Furthermore, radio-protection requirements specifically preclude any use of glue or adhesive tape in contact with the sample backing. To meet these requirements, an appropriate sample holder design has been prepared [9] and previously used, where samples are kept in place by mechanical pressure only. The entire procedure is conducted in a Class-A laboratory under constant Radio-Protection supervision. However, in the particular case of ^{241}Am, should it be requested by the CERN Radioprotection, it can be foreseen to mount the samples inside the fission chamber at IRMM before transport to CERN.

2.3 Electronics and data acquisition

A setup based on existing electronics from previous fission measurements, consisting of custom-made preamplifiers will be used for fast signal shaping. Incremental improvements have been made over the past few years, adding electronic protection to the pre-amplifier channels to prevent breakage in case of sparks and shielding the preamplifier module to reduce the baseline oscillation observed following the prompt *γ-flash*. The analogue detector signals will be input from the preamplifier module into the standard n_TOF Data Acquisition System based on flash-ADCs (12- or 14-bit).

3. Beam request

Using the large collimator (6 cm Ø) in EAR-2, the mass of the samples can be minimised. This is a very significant advantage in the case of ^{241}Am with its high specific activity. With the proposed characteristics (6 cm diameter, 5 μg/cm^2) each sample will have a mass of 0.14 mg and an activity of ~18 MBq. In total, the mass and activity will be 0.85 mg and 118 MBq respectively for the six samples. This is more than 2.5 times less than the material previously used in EAR-1.

With the allocation of $3x10^{18}$ protons on target, it should be possible to obtain data with a statistical uncertainty of less than 1% above 600 keV and less than 3% above 250 keV at 20 bins per energy decade. In addition to the data at and above the fission threshold, data on several resonances from 0.6 to 100 eV will be obtained with integrals of a few hundred to a few thousand counts. The expected counts and corresponding statistical uncertainties above 100 keV are shown in Figure 3.

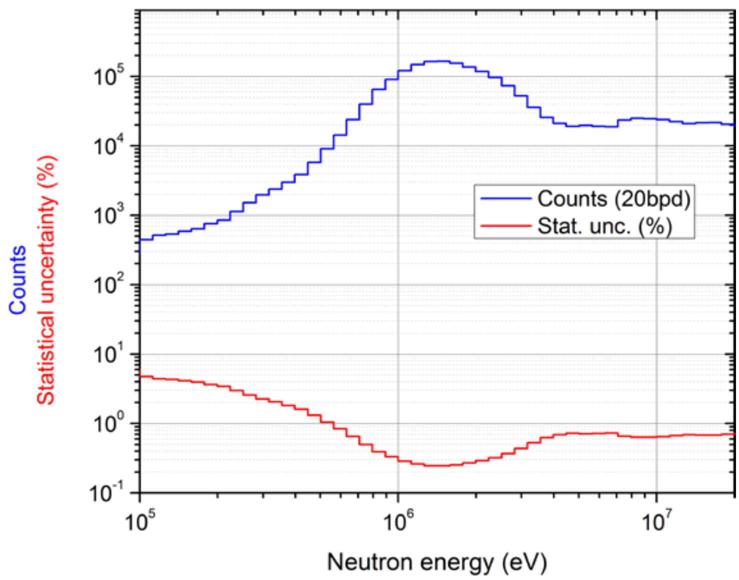

Figure 3: *Total expected counts above 100 keV incident neutron energy from ^{241}Am at 20 bins per energy decade along with the corresponding statistical uncertainty.*

4. Summary

Accurate data on the fission-induced cross-section of ^{241}Am are essential for the design of innovative nuclear systems that could address the question of high-level radioactive waste. The high specific activity of this isotope, as well as its rapidly decreasing cross-section below the fission threshold make this a challenging measurement. Both aspects can be addressed at n_TOF's Experimental Area II (EAR-2), where the high instantaneous flux can more effectively suppress the sample-induced background and allow the measurement at and above the fission threshold to be performed under significantly improved conditions with respect to the measurement performed in EAR-1 in 2003.

Summary of requested protons:

3x10^{18} protons on target (EAR-2)

References:

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