EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the fission cross-section of ^{243}Am at EAR-1 and EAR-2 of the CERN n_TOF facility

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Abstract: We propose to measure the neutron-induced fission cross-section of ^{243}Am in EAR-1 and EAR-2 of the n_TOF facility using Micromegas detectors. Recent attention has lead to the investigation of the effect of americium isotopes as burnable actinides on a variety of nuclear reactors. Among americium isotopes, ^{243}Am, with a half-life of 7364 years, is the one with the least studied neutron-induced fission cross-section in the thermal and resonance region. The aim of this experiment is to produce a single accurate data-set covering the energy range from thermal up to a few hundreds of MeV neutron energy. Due to the lack of experimental data in the thermal and resonance region, the available evaluated libraries disagree on the existence of the first two resonances of ^{243}Am. The combination of the two available experimental areas as well as the use of a "hybrid" target configuration consisting of thick and thin ^{243}Am samples, is deemed crucial to acquire data with satisfactory statistical uncertainty in a wide energy range. The part of the measurement that will be performed in EAR-1 will cover the energy region from

the fission threshold up to a few hundreds of MeV, by taking advantage of the thick ^{243}Am samples. In EAR-2, the provided high neutron flux will result in satisfactory statistics in the thermal and resonance region, using the thick samples, whereas the sub-threshold and MeV region, will become available as an overlapping region between EAR-1 and EAR-2 using the thinner samples.

Requested protons: 6x10^{18} (3x10^{18} in EAR-1 , 3x10^{18} in EAR-2)
Experimental Area: EAR-1 and EAR-2

1 Introduction

1.1 Motivation

The design, feasibility and sensitivity studies on the new generation of nuclear reactors, such as Accelerator Driven Systems-ADS [1] and Generation IV Fast Neutron Reactors [2], require high-accuracy data for a variety of neutron-induced reactions, at energies ranging from thermal up to several tens of MeV. In this scope, recent attention has lead to the investigation of americium isotopes, classified as high-level nuclear waste from conventional reactors, that could be used as a burnable minor actinide on future nuclear reactors [3]. Among these long-lived minor actinides, the ^{243}Am isotope ($T_{1/2}$ = 7364 y), which is the longest-lived of the americium isotopes, plays an important role in the radiotoxicity of the nuclear waste, as it contributes to the long-term production of ^{239}Pu with a half-live of 24110 years, via α- and subsequent β- decay.

Data for the ^{243}Am(n,f) reaction are urgently needed, since the NEA (Nuclear Energy Agency) has classified this reaction in the "General Request List" (GRL) [4]. The needs for nuclear data relevant for advanced nuclear systems summarized in the OECD/NEA WPEC Subgroup 26 Final Report [5], highlight that, for this specific fission cross-section reaction, target accuracies of $\sim 2\%$ are needed in the MeV region.

Present uncertainties and target requirements for the neutron-induced fission cross-reaction of ^{243}Am relevant to Accelerator-Driven Minor Actinide Burners (ADMAB) and Sodium-cooled Fast Reactors (SFR) are summarized in Table 1.

Table 1: Initial and Target Uncertainties of the $^{243}Am(n,f)$ cross-section [5]

	Uncertainty (%)		
Energy Range	**Existing**	**Target for ADMAB**	**Target for SFR**
6.07 - 2.23 MeV	11.0	2.3	8.2
2.23 - 1.35 MeV	6.0	1.9	-
1.35 - 0.498 MeV	9.2	1.6	7.2

1.2 Present status of data

The published fission cross-section data in the thermal and resolved resonance regions (Fig. 1), are not enough both in quantity and quality. In the resonance region, the data of Kobayashi et al. [6] remain the only available data in the range from 50 meV to 15 keV. Unfortunately, for this data-set the energy resolution is very coarse. Additionally, below 50 meV only four data points are available and present a huge discrepancy [6, 7, 8, 9]. Moreover, evaluated libraries, in particular ENDF/B-VIII.0 [10], JEFF-3.3 [11] and TENDL-2019 [12], show major inconsistencies below the eV region due to the lack of experimental data.

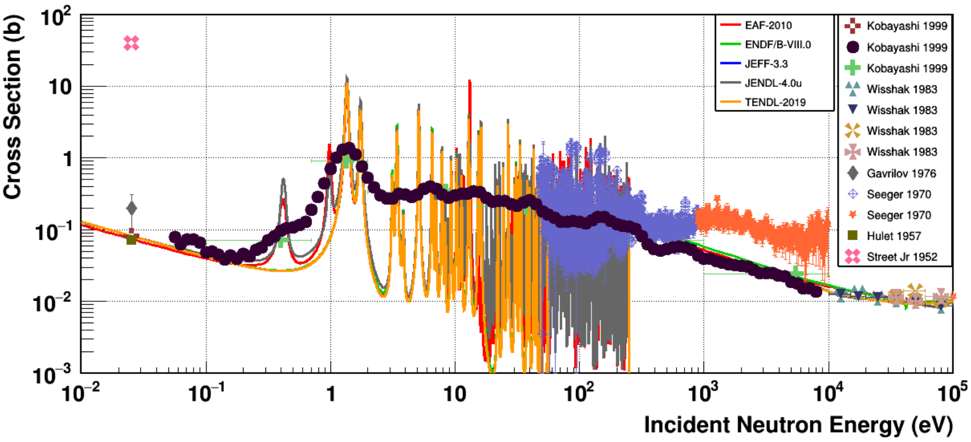

Figure 1: Previous measurements of ^{243}Am(n,f) cross-section up to 100 keV along with the available evaluated libraries (retrieved from the EXFOR database).

In the subthreshold energy region, Wisshak and Käppeler [13] provided data between 5 and 250 keV. In the high neutron energy region (Fig. 2), large deviations among the existing data are observed especially around the threshold as well as at the plateau of the fission reaction. Furthermore, above 20 MeV there is only one available set of data by Laptev et al., [14] that extends up to 200 MeV. However, these data were normalized to three energy points (1, 5 and 10 MeV) according to the ENDF/B-VII.0 library [15].

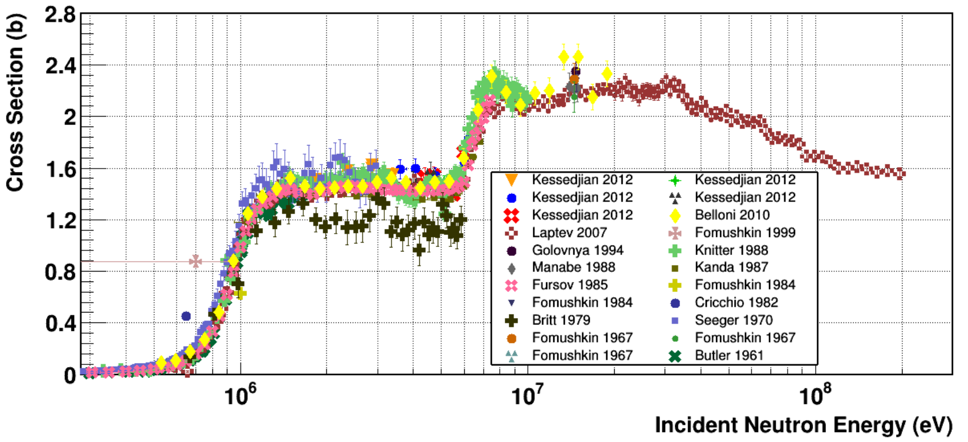

Figure 2: Previous measurements of ^{243}Am(n,f) cross-section above 300 keV (retrieved from the EXFOR database).

1.3 Previous measurement at n_TOF and prospects for the new measurement

A measurement of the ^{243}Am(n,f) cross-section was already performed at n_TOF in 2004 (EAR-1) during Phase I [16]. In this experiment, the statistical accuracy was limited due to the mass of AmO_2 samples ($\sim$ 4.8 mg) [17]. Therefore, a wide energy bin (20 bpd) was chosen in order to maintain the statistical uncertainty in each bin below 3% for the energy region from 0.5 to 20 MeV. From the same measurement, the analysis of the data at the resolved resonance region was seriously affected from the contaminants in the sample.

With this proposal, a new accurate measurement is called to address the discrepancies in the existing data and also to expand the experimental data to the thermal region, which is yet unexplored, as well as to the high energy region. To cover the needs in statistics, samples with $\sim$ 6 times higher mass will be used, while a "hybrid" target configuration consisting of thick and thin ^{243}Am samples will be implemented in order to avoid pile-up issues and maintain high reaction rates in all energy regions. Performing the first part of the measurement in EAR-2 will ensure satisfactory statistics in the thermal and resonance region, thanks to this area's higher instantaneous flux. For energies higher than $\sim$ 1 MeV, strong pile-up issues as well as extended overlapping with the prominent gamma flash pulse impose high correction factors. For these energies, the measurement at EAR-1 can provide high accuracy data and even more importantly extend the experimental information up to 200-300 MeV, a previously unexplored energy region. In this way, the combination of the measurement in EAR-1 and EAR-2 will provide a single data-set ranging from thermal up to a few hundreds of MeV neutron energy for the first time.

2 Experimental setup

2.1 Samples

For the successful realization of the proposed measurement, it is instrumental to ensure the availability of high purity actinide material that allows the production of high quality ^{243}Am samples. In this respect, the expertise, the experience and the available instrumentation as provided by the EC-JRC Geel Laboratory is a key-factor for the proposal. In detail, the JRC laboratory will take over the chemical separation of the material as to avoid possible ^{239}Pu contaminants [18] as well as the accurate characterization of the targets. The chemical separation needs to be performed just before the production of the ^{243}Am targets and consequently the characterization and the experiment should follow the soonest possible. In this way ^{239}Pu built-up will be avoided.

It is planned to use six high purity AmO_2 samples, for a total mass of 25 mg of ^{243}Am ($\sim$ 150 ug/cm^2 per sample) and five high purity AmO_2 samples, for a total mass of 0.4 mg of ^{243}Am ($\sim$ 3 ug/cm^2 per sample). By distributing the total mass of americium into several samples, the Micromegas detectors (explained in Sec. 2.2) will be protected from potential radiation damage. The material will be electroplated in a surface of 6 cm in diameter on an aluminum backing 0.025 mm thick. For the measurement, additional samples of ^{235}U, ^{238}U and ^{10}B will be prepared to be used as reference for the neutron flux determination.

2.2 The Micromegas detectors

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

Table 2: Main characteristics of the requested ^{243}Am samples.

Isotope	Quantity	Optimized for	Diameter (cm)	Mass per Target (mgr)	Total Activity (MBq)
^{243}Am	6	EAR-2/EAR-1	6	4.2	187.8
^{243}Am	5	EAR-2	6	0.085	3.15

neutrons as well as to avoid the perturbation of the neutron flux. For this reason, the microbulk design [19, 20] was developed based on the Micromegas principle (Fig. 3). This design has also been utilized at n_TOF in the past for neutron-induced fission cross-section measurements of ^{242}Pu [21], ^{240}Pu [22], ^{237}Np [23], ^{241}Am [24] and ^{230}Th [25].

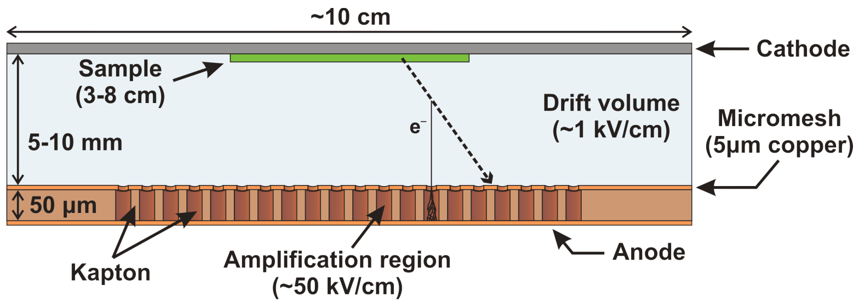

Figure 3: An illustration of the basic principle of operation of a Micromegas detector.

Two fission chambers will be used in order to house 18 sample-detectors modules. Within the chambers a continuous flow of Ar:CF_4:isoC_4H_{10} gas mixture (88:10:2) will be maintained. This particular gas mixture is commonly used at CERN for this kind of measurements and has excellent timing properties due to the relatively high electron drift velocity.

2.3 Electronics and data acquisition

A setup based on existing electronics from previous fission measurements, consisting of custom-made pre-amplifiers (INFN-Bari), will be used for fast signal shaping. Incremental improvements have been made over the past few years in the design of the pre-amplifiers, resulting in a significant reduction of baseline oscillations and in the enhancement of the signal-to-noise ratio. The output of the pre-amplifiers will be directed to the standard n_TOF Data Acquisition System based on flash-ADCs (12- or 14-bit).

2.4 Beam request

By using the large collimator in both experimental areas, i.e. 6 cm diameter in EAR-2 and 8 cm diameter in EAR-1, we can profit from increased statistics. As can be seen in Fig. 4, by assuming 3x10^{18} protons for each experimental area and by adopting six times higher americium sample mass than in the previous measurement, we can achieve satisfactory statistics from thermal up to a few hundreds of MeV neutron energy. In this way, and by taking advantage of the thick and thin ^{243}Am samples, we can establish an extended overlapping region so as to provide a unified data-set covering the whole energy spectrum. For the reaction rate estimations, shown in Fig. 4, the n_TOF-Phase III neutron flux was considered for both experimental areas. For EAR-1 we do not expect the neutron flux to change in Phase IV, while for EAR-2 the flux will increase above 700 keV, however the resulting counting rates are still expected to be low enough for the thin samples in order to have minimal pile-up corrections.

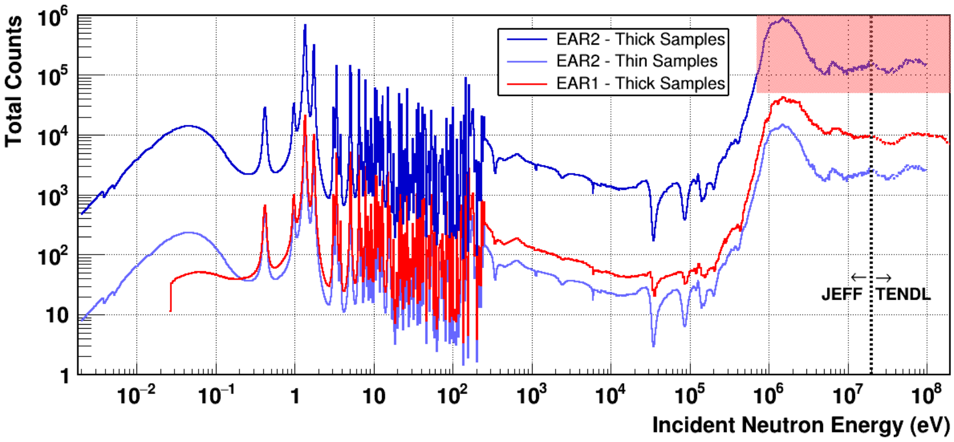

Figure 4: Total expected counts for the ^{243}Am(n,f) reaction using the evaluated libraries JEFF-3.3 [11] (continuous line) and TENDL-2019 [12] (dashed line), taking into account $3x10^{18}$ protons for each experimental area at 100 bins per energy decade. In the extreme high energy region the only available evaluation is from TENDL-2019. The red shaded rectangular shows the region where the strong pulse pile-up, hinders the measurement in EAR-2. As can be seen, for this energy region (700 keV and above) high accuracy cross-section data can only be obtained from the EAR-1 measurement.

2.5 Summary

We aim to collect accurate data on the neutron-induced fission cross-section of ^{243}Am for the first time with a single data-set from thermal neutrons up to a few hundreds of MeV. The production and accurate characterization of the ^{243}Am targets provided by EC-JRC Geel will allow for the collection of data with satisfactory statistics and without significant contamination issues. The realization of the experiment in EAR-2 will profit from an improved signal-to-background ratio, while the data collected from the EAR-1 measurement will allow for the extension of the cross-section data in the high energy region. With this proposed measurement, the previous n_TOF data will be significantly extended in neutron energy but also improved in terms of energy resolution, as a finer binning is expected to be implemented. Discrepancies among previous measurements and evaluations will be addressed and a cross-section measurement with a unified single data-set that covers 10 orders of magnitude in neutron energy from thermal up to $\sim$ 300 MeV will be realized.

Summary of requested protons: $6x10^{18}$ protons on target in total
$3x10^{18}$ protons (EAR-1) and $3x10^{18}$ protons (EAR-2)

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