Measurement of the ^{244}Cm capture cross sections at both CERN n_TOF experimental areas

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Abstract.
Accurate neutron capture cross section data for minor actinides (MAs) are required to estimate the production and transmutation rates of MAs in light water reactors with a high burnup, critical fast reactors like Gen-IV systems and other innovative reactor systems such as accelerator driven systems (ADS). Capture reactions of ^{244}Cm open the path for the formation of heavier Cm isotopes and of heavier elements such as Bk and Cf. In addition, ^{244}Cm shares nearly 50% of the total actinide decay heat in irradiated reactor fuels with a high burnup, even after three years of cooling.
Experimental data for this isotope are very scarce due to the difficulties of providing isotopically enriched samples and because the high intrinsic activity of the samples requires the use of neutron facilities with high instantaneous flux. The only two previous experimental data sets for this neutron capture cross section have been obtained in 1969 using a nuclear explosion and, more recently, at J-PARC in 2010.
The neutron capture cross sections have been measured at n_TOF with the same samples that the previous experiments in J-PARC. The samples were measured at n_TOF Experimental Area 2 (EAR-2) with three C_6D_6 detectors and also in Experimental Area 1 (EAR-1) with the Total Absorption Calorimeter (TAC). Preliminary results assessing the quality and limitations of these new experimental datasets are presented for the experiments in both areas. Preliminary yields of both measurements will be compared with evaluated libraries for the first time.

1 Introduction

Neutron capture cross section data for minor actinides (MAs) are required to calculate the transmutation and production rates of MAs in light-water reactors (LWR) with a high burnup, critical fast reactors like Gen-IV systems and accelerator driven systems (ADS) [1]. Accurate measurements of these cross sections, however, are very difficult due to the high radioactivity of MAs and the difficulty to find appropriate samples. Therefore, data with high accuracy are not available, in particular ^{244}Cm ($T_{1/2}$ =18.1 years) is one of the most important MAs due to his contribution to the radiotoxicity of the irradiated nuclear fuels and the difficulty of its transmutation. There are only two previous measurements. The first, done in 1969, used the neutrons produced in underground nuclear explosion. The capture cross section was measured from 20 eV to 1 keV [2]. The second measurement was done in 2010 by Kimura *et al.* at J-PARC [3]. The resonance analysis was done up to 30 eV.

2 The experiment

The ^{244}Cm cross section has been measured at the n_TOF spallation neutron-time-of-flight facility at CERN. This facility have a nominal intensity of 7×10^{12} protons per pulse and a time spread of 7 ns (rms). The facility has two experimental areas, one at 185 m [4] (EAR-1), the other at 19 m [5] (EAR-2). The instantaneous flux is up to 40 times higher in EAR-2 while the energy resolution is better in EAR-1.

Table 1. Istopic composition of the ^{244}Cm sample

Isotope	mole fraction (%)
^{240}Pu	30.9 ±0.6
^{244}Cm	60.1 ±1.1
^{245}Cm	2.5 ±0.3
^{246}Cm	6.6 ±0.3

The yield has been obtained doing experiments in both areas. The measurement in EAR-2 has been done with three BICRON C_6D_6 detectors and the Total Energy Detection (TED) [6] technique. The measurement in EAR-1 has been done with the Total Absorption Calorimeter (TAC) [7]. The main measurement is using the C_6D_6, in which much more statistics was achieved. The one with the TAC was carried out to have an alternative normalization and a more complete information of the capture cascades.

Two of the samples already measured at J-PARC [3], with ~0.4 mg ^{240}Pu and ~0.8 mg ^{244}Cm in total, have been used for this measurement. The isotopic abundances of the measured sample are given in Table 1.

2.1 The measurement in EAR-1 with the TAC

For the calculation of the detection efficiency a very detailed description of the experimental set-up [10] has been implemented in the Geant4 toolkit [11]. Validation tests

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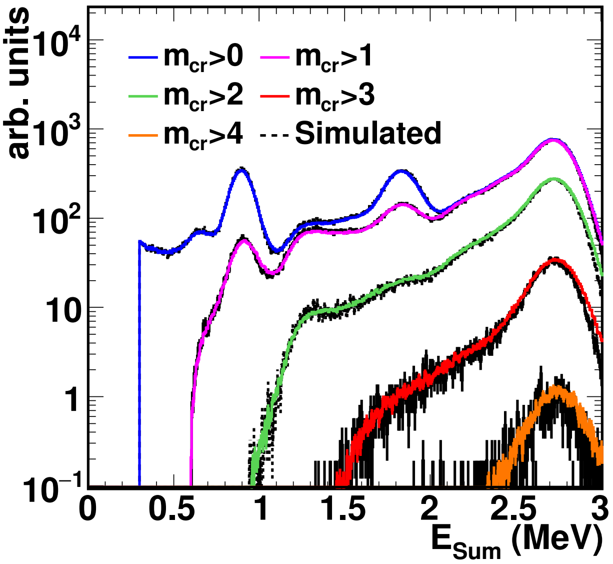

Figure 1. Comparison between simulated and experimental response function to an ^{88}Y source with the TAC.

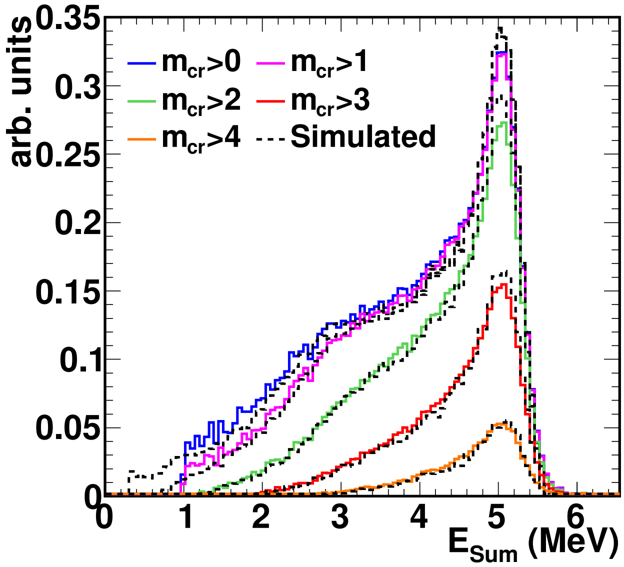

Figure 3. Comparison between simulated and experimental deposited energy spectra in the TAC by ^{240}Pu(n,γ) cascades.

of the simulation code with an ^{88}Y source are presented in figure 1. The electromagnetic cascades released after neutron capture have been obtained from the NuDEX code [12]. As in previous works, the Photon Strength Functions of the compound nuclei, ^{241}Pu and ^{245}Cm, have been modified in order to reproduce the deposited energy spectra. In this case a fitting procedure based on the differential evolution algorithm has been applied [13]. After the fitting process, an excellent reproduction of the deposited energy spectra has been achieved, as presented in figures 2 and 3.

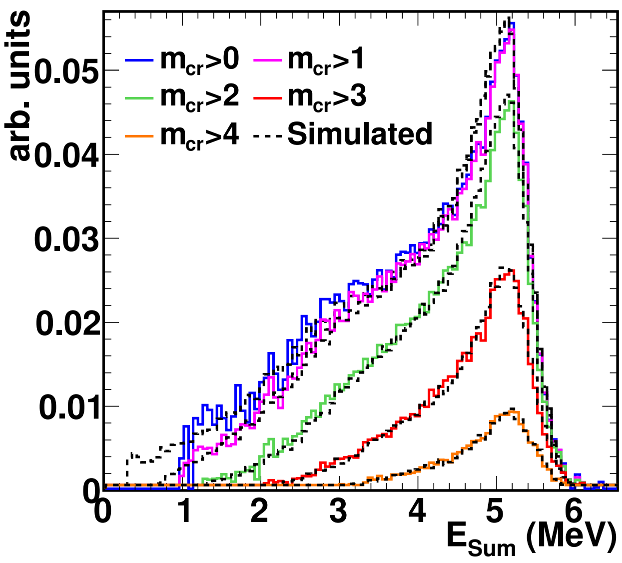

Figure 2. Comparison between simulated and experimental deposited energy spectra in the TAC by ^{244}Cm(n,γ) cascades.

A preliminary experimental yield (no background subtracted) of the first resonances is presented in figure 4. Theoretical yields calculated with the JEFF-3.3 [14] cross sections are also plotted. The yield has been normalized to the first resonance of ^{240}Pu at 1.1 eV.

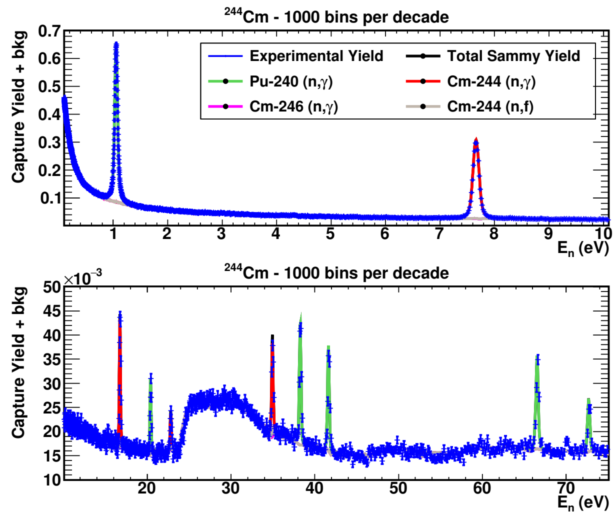

Figure 4. Preliminary experimental yields (no background subtracted) of the measured ^{244}Cm sample in EAR-1 with the TAC. Together with the experimental data points, we show an estimation of the contribution of the reactions in each isotope present in the samples. These contributions have been obtained using the SAMMY [15] computer code and JEFF-3.3 data [14]

2.2 The measurement in EAR-2 with the C_6D_6

In the n_TOF EAR-2, the three C_6D_6 detectors were placed 5 cm from the centre of the sample. The data analysis has been performed in this case following the TED technique, which can be applied when (i) no more than one γ-ray is detected per capture cascade, and (ii) the detection efficiency is proportional to the γ-ray energy. In order to fulfil the second condition we applied the Pulse Height Weighting Technique (PHWT), which has been validated to perform measurements at n_TOF [16]. In this technique, each count in the detectors is weighted by a factor, which is obtained from Monte Carlo simulations. For the calculation of this weighting factors, we have implemented a very detailed description of the experimental setup in Geant4 [17].

The corrections due to the counts lost below the detection threshold and the effect of detecting multiple γ-rays of the same cascade have been performed by means of Monte Carlo simulations as well. For this, the same ^{240}Pu(n,γ) and ^{244}Cm(n,γ) cascades used for normalizing the TAC measurement (figures 2 and 3) were simulated in the C_6D_6 setup. These cascades were fitted to reproduce the TAC experimental data but not the C_6D_6 data. The simulations were performed with Geant4 with the same geometry implemented for the calculation of the weighting factors. Figure 5 shows how well these cascades reproduce the response function of the C_6D_6 detectors.

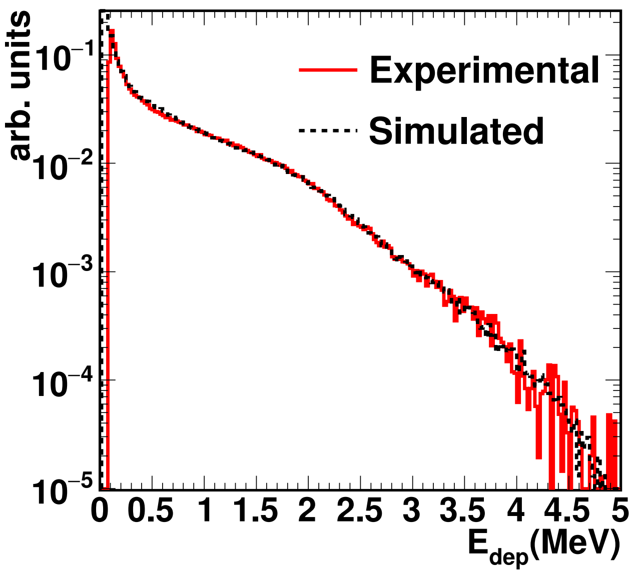

Figure 5. Comparison between simulated and experimental response functions of the C_6D_6s to ^{244}Cm(n,γ) cascades.

Other experimental effects such as the pile-up have been considered and corrected. The experimental yield has been normalized to the first resonance of ^{240}Pu at 1.1 eV. A preliminary experimental yield (no background subtracted) is presented in figure 6. As in the case of the TAC, theoretical yields calculated with the JEFF-3.3 cross sections are plotted as well. A preliminary version of the EAR-2 resolution function [18] has also been considered.

3 Results

The experimental yields obtained in both experimental areas have been compared with the yields obtained from

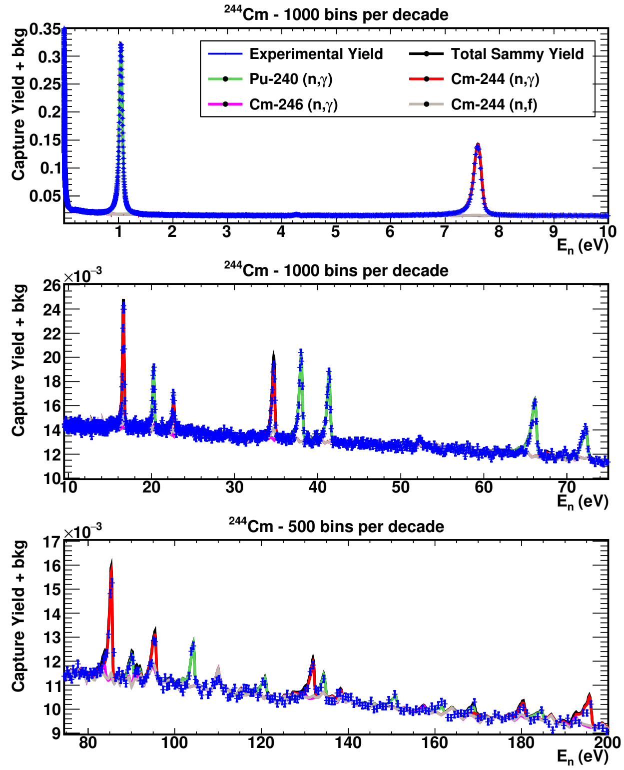

Figure 6. Preliminary experimental yields (no background subtracted) of the measured ^{244}Cm sample in EAR-2 with the C_6D_6. Together with the experimental data points, we show an estimation of the contribution of the reactions in each isotope present in the samples. These contributions have been obtained using the SAMMY computer code and JEFF-3.3 data [14].

the JEFF-3.3 cross sections using a (preliminary) n_TOF EAR-2 resolution function. The resolution function in the EAR-1 has also been considered but it has a very small effect. The results of this preliminary comparison are provided in table 2. For the TAC, only data concerning the strongest ^{240}Pu and ^{244}Cm resonances are presented, since the statistical uncertainties of this measurement are significantly larger than for the C_6D_6 measurement.

From the results of the this preliminary comparison it follows that the ^{240}Pu(n,γ) cross section measured at the n_TOF EAR-2 is compatible with the one in JEFF-3.3, within uncertainties, whereas some discrepancies are observed for ^{244}Cm. We note a 4% difference between both measurements in the value of the first and strongest ^{244}Cm resonance at 7.7 eV. Here it should be noted that the measurements have been made with the same sample, but in different experimental areas, with different detectors, and with different measurement techniques.These preliminary observations may evolve as the analysis progresses further.

4 Summary and conclusions

The neutron capture cross section of ^{244}Cm has been measured at both n_TOF experimental areas, using different detectors and measurement techniques. Two samples already measured at J-PARC with ~0.4 mg ^{240}Pu and ~0.8 mg ^{244}Cm in total have been used for the measurement. In

Table 2. Preliminary comparison between the integrals over a resonance of the experimental yields (I_{Exp}) and JEFF-3.3 ($I_{JEFF-3.3}$). The values of the table correspond to $100 \cdot (1 - I_{Exp}/I_{JEFF-3.3})$. The yields have been normalized to the strongest resonance of ^{240}Pu at 1.1 eV. Presented uncertainties are due to counting statistics only.

Resonance	TAC in EAR-1	C_6D_6 in EAR-2
1.1 eV- ^{240}Pu	0.0 ± 0.2	0.0 ± 0.1
20.4 eV- ^{240}Pu	-	3.6 ± 4.1
38.3 eV- ^{240}Pu	-	-1.9 ± 3.2
41.7 eV- ^{240}Pu	-	1.9 ± 3.6
66.6 eV- ^{240}Pu	-	7.2 ± 4.1
72.8 eV- ^{240}Pu	-	-5.1 ± 7.9
90.8 eV- ^{240}Pu	-	16.4 ± 13.3
105.0 eV- ^{240}Pu	-	-2.4 ± 11.2
135.3 eV- ^{240}Pu	-	24.7 ± 19.0
7.7 eV- ^{244}Cm	2.3 ± 0.4	6.5 ± 0.2
16.8 eV- ^{244}Cm	-	8.9 ± 3.3
23.0 eV- ^{244}Cm	-	-17.5 ± 6.9
35.0 eV- ^{244}Cm	-	14.1 ± 3.1
86.0 eV- ^{244}Cm	-	1.3 ± 4.2
96.1 eV- ^{244}Cm	-	20.8 ± 12.5
132.8 eV- ^{244}Cm	-	12.1 ± 11.4
197.4 eV- ^{244}Cm	-	30.4 ± 14.1

the EAR-2 the γ-rays from neutron capture have been detected with three C_6D_6 detectors, and in the EAR-1 with the n_TOF TAC. A very detailed description of the geometry of both experimental setups has been implemented in Geant4, in order to calculate the detection efficiency and to apply corrections to the TED technique. The capture cascades of ^{240}Pu and ^{244}Cm have been simulated with the NuDEX code and a very good reproduction of the experimental deposited energy spectra in both the TAC and the C_6D_6 has been achieved. Preliminary capture yields have been obtained, showing that the measured ^{240}Pu capture cross section is compatible with the one in JEFF-3.3, and that a difference of only 4% has been obtained between both ^{244}Cm measurements of the first strong resonance. Further analysis is in progress.

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