Measurement of the 244Cm 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$_6$D$_6$ 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 $7 \times 10^{12}$ 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.

The yield has been obtained doing experiments in both areas. The measurement in EAR-2 has been done with three BICRON C$_6$D$_6$ 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.

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

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mole fraction (%)</th>
</tr>
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<tbody>
<tr>
<td>$^{240}$Pu</td>
<td>30.9 ±0.6</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>60.1 ±1.1</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>2.5 ±0.3</td>
</tr>
<tr>
<td>$^{248}$Cm</td>
<td>6.6 ±0.3</td>
</tr>
</tbody>
</table>

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
Accurate neutron capture cross section data for minor actinides (MAs) are required to estimate the production rates of MAs in light-water reactors (LWR) with a high radioactivity of MAs and the difficulties of providing isotopically enriched samples. Therefore, data with high accuracy have been obtained in 1969 using a nuclear explosion and, more recently, at J-PARC in 2010. 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 two previous measurements. The first, done in 1969, used the neutrons produced in underground nuclear explosion. The second measurement was done in 2010 by the neutrons produced in underground nuclear explosion. In both areas, Preliminary yields of both measurements will be compared with evaluated libraries for the first results assessing the quality and limitations of these new experimental datasets are presented for the experiments.

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

Figure 2. Comparison between simulated and experimental deposited energy spectra in the TAC by $^{240}$Pu(n,$\gamma$) cascades.

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

Figure 4. Preliminary experimental yields (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.

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.

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 fulfill 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 239Pu(n,γ) and 244Cm(n,γ) cascades used for normalizing the TAC measurement (figures 2 and 3) were simulated in the C6D6 setup. These cascades were fitted to reproduce the TAC experimental data but not the C6D6 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 C6D6 detectors.

![Figure 5. Comparison between simulated and experimental response functions of the C6D6 to 244Cm(n,γ) cascades.](image)

Other experimental effects such as the pile-up have been considered and corrected. The experimental yield has been normalized to the first resonance of 240Pu 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 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 239Pu and 244Cm resonances are presented, since the statistical uncertainties of this measurement are significantly larger than for the C6D6 measurement.

From the results of this preliminary comparison it follows that the 239Pu(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 244Cm. We note a 4% difference between both measurements in the value of the first and strongest 244Cm 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 244Cm 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 239Pu and ~0.8 mg 244Cm in total have been used for the measurement. In
Table 2. Preliminary comparison between the integrals over a resonance of the experimental yields ($I_{\text{Exp}}$) and JEFF-3.3 ($I_{\text{JEFF-3.3}}$). The values of the table correspond to $100\cdot(1-I_{\text{Exp}}/I_{\text{JEFF-3.3}})$. The yields have been normalized to the strongest resonance of 240Pu at 1.1 eV. Presented uncertainties are due to counting statistics only.

<table>
<thead>
<tr>
<th>Resonance</th>
<th>TAC in EAR-1</th>
<th>$C_{\text{D}_0}$ in EAR-2</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1 eV - 240Pu</td>
<td>0.0 ± 0.2</td>
<td>0.0 ± 0.1</td>
</tr>
<tr>
<td>20.4 eV - 240Pu</td>
<td>-</td>
<td>3.6 ± 4.1</td>
</tr>
<tr>
<td>38.3 eV - 240Pu</td>
<td>-</td>
<td>-1.9 ± 3.2</td>
</tr>
<tr>
<td>41.7 eV - 240Pu</td>
<td>-</td>
<td>1.9 ± 3.6</td>
</tr>
<tr>
<td>66.6 eV - 240Pu</td>
<td>-</td>
<td>7.2 ± 4.1</td>
</tr>
<tr>
<td>72.8 eV - 240Pu</td>
<td>-</td>
<td>-5.1 ± 7.9</td>
</tr>
<tr>
<td>90.8 eV - 240Pu</td>
<td>-</td>
<td>16.4 ± 13.3</td>
</tr>
<tr>
<td>105.0 eV - 240Pu</td>
<td>-</td>
<td>-2.4 ± 11.2</td>
</tr>
<tr>
<td>135.3 eV - 240Pu</td>
<td>-</td>
<td>24.7 ± 19.0</td>
</tr>
<tr>
<td>7.7 eV - 244Cm</td>
<td>2.3 ± 0.4</td>
<td>6.5 ± 0.2</td>
</tr>
<tr>
<td>16.8 eV - 244Cm</td>
<td>-</td>
<td>8.9 ± 3.3</td>
</tr>
<tr>
<td>23.0 eV - 244Cm</td>
<td>-</td>
<td>-17.5 ± 6.9</td>
</tr>
<tr>
<td>35.0 eV - 244Cm</td>
<td>-</td>
<td>14.1 ± 3.1</td>
</tr>
<tr>
<td>86.0 eV - 244Cm</td>
<td>-</td>
<td>1.3 ± 4.2</td>
</tr>
<tr>
<td>96.1 eV - 244Cm</td>
<td>-</td>
<td>20.8 ± 12.5</td>
</tr>
<tr>
<td>132.8 eV - 244Cm</td>
<td>-</td>
<td>12.1 ± 11.4</td>
</tr>
<tr>
<td>197.4 eV - 244Cm</td>
<td>-</td>
<td>30.4 ± 14.1</td>
</tr>
</tbody>
</table>

In the EAR-2 the γ-rays from neutron capture have been detected with three C6D6 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 240Pu and 244Cm have been simulated with the NuDEX code and a very good reproduction of the experimental deposited energy spectra in both the TAC and the C6D6 has been achieved. Preliminary capture yields have been obtained, showing that the measured 240Pu capture cross section is compatible with the one in JEFF-3.3, and that a difference of only 4% has been obtained between both 244Cm measurements of the first strong resonance. Further analysis is in progress.

References

[13] E. Mendoza et al., Study of photon strength functions of 231Pu and 245Cm from neutron capture measurements, in 2019 International Conference on Nuclear Data for Science and Technology (submitted)