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

Measurement of the ^{244}Cm and ^{246}Cm neutron capture cross sections at the n_TOF facility

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ABSTRACT

The neutron capture reactions in ^{244}Cm and ^{246}Cm isotopes open the path of the formation of heavier Cm isotopes and heavier elements such as Bk and Cf. In addition, both isotopes are two of the minor actinides with a large contribution to the decay heat and to the neutron emission in irradiated fuels proposed for the transmutation of the nuclear waste and fast critical reactors. The available experimental data for both isotopes is very scarce due to the difficulties in performing the measurements: high intrinsic activity of the samples and the limited facilities capable of providing isotopically enriched samples. We propose to perform a neutron capture cross section measurement with isotopically enriched samples of ^{244}Cm and ^{246}Cm provided by JAEA. The measurement will cover the range from 1 eV to 550 eV and will be performed at the n_TOF Experimental Area 2 (EAR-2). In addition, a normalization measurement with the ^{244}Cm sample will be performed at Experimental Area 1 (EAR-1) with the Total Absorption Calorimeter.

Requested protons: 8.5×10^{18} protons on target
Experimental area: EAR2 (8.0×10^{18} protons) and EAR1 (0.5×10^{18} protons).

1 INTRODUCTION AND MOTIVATION

Accurate neutron capture cross section data for minor actinides (MAs) are required to estimate the production and transmutation rates of MAs in LWR reactors with a high burnup, critical fast reactors like Gen-IV systems and other innovative reactor systems such as accelerator driven systems (ADS) [1]. The ^{244}Cm ($T_{1/2}$=18.1 years) and ^{246}Cm ($T_{1/2}$=4730 years) isotopes are among the most important MAs due to the difficulties in their transmutation (by fission) and their contribution to the radiotoxicity of the irradiated nuclear fuels. In particular, ^{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. Furthermore, both ^{244}Cm and ^{246}Cm are two of the most important neutron emitters in the irradiated nuclear fuel, making the need of accurate data associated to their production especially important. In addition, both of them are in the path of the creation of any heavier Cm isotopes and heavier elements like Bk and Cf.

The first and only data available until 2012 on the ^{244}Cm and ^{246}Cm neutron capture cross sections came from an experiment [2] which used the neutrons produced in an under-ground nuclear explosion. The ^{244}Cm(n,γ) cross section data range from 20 eV to 1 keV and the ^{246}Cm(n,γ) data from 20 eV to 400 eV. Recently, a second measurement of both (n,γ) cross sections has been performed with a large coverage Ge array in the Accurate Neutron Nucleus Reaction Measurement Instrument ANNRI at J-PARC [3], in the energy range of 1 eV to 300 eV. The scarcity of the available data and the many experimental challenges involved in the two previous measurements makes highly desirable to perform an additional measurement under different conditions i.e., in a different facility, with different detectors and monitors and a different methodology.

We propose to measure both ^{244}Cm and ^{246}Cm neutron capture cross sections at the n_TOF EAR-2 flight path with the same samples used at J-PARC. The combination of two times higher instantaneous fluence at n_TOF EAR-2 with respect to the one used at J-PARC during the Cm measurement (120 kW of beam power, upgraded in 2015 to 500 kW), the use of a high performance digital acquisition system (12 bits resolution, 1 Gsample/s) and the total energy detector technique (which allows to reach accuracies at the level of a few %) based on C_6D_6 detectors [4] will allow a good control of the systematic uncertainties in the neutron energy range between 1 eV to 250 eV. We expect as well to be able to obtain results in the range from 250 eV to 550 eV with lower accuracy.

In the neutron energy range above 100 eV, the measurement with germanium detectors performed at J-PARC required important dead time corrections (up to 90%) due to the high counting rates and the long decay times of the germanium pulses (> 4 μs). The use of faster organic C_6D_6 scintillators (τ_{fast}=2.8ns) for the higher counting rates expected at EAR-2 will lead to a pileup probabilities (i.e. dead time corrections) below a few percent. In addition, the detector signals will be digitized with 12 bit resolution flash ADCs and stored on disk, then processed with sophisticated pulse shape analysis techniques [5] and the final data will corrected with sophisticated pileup correction methods [6][7]. Such a methodology guarantees an excellent control of the systematic uncertainties associated to the counting of signals.

2 EXPERIMENTAL SETUP

The measurements will be performed with two ^{244}Cm samples and one ^{246}Cm sample, all of them provided by JAEA. Each ^{244}Cm sample is made with 0.68 mg of CmO_2 and the ^{246}Cm sample is made with 2.1 mg of CmO_2. The total activities of the two ^{244}Cm samples and the ^{246}Cm sample are 2.5 GBq and 1.15 GBq, respectively. In the three cases the curium oxide is mixed with 25 mg of aluminum powder, and pressed to a disk of 5 mm diameter and 0.3 mm thick. The disk is placed inside an aluminum capsule with 9 mm diameter and 1.2 mm thick. The isotopic composition of the samples is given in Table 1.

Due to the low mass of the samples the measurement have to be performed at the n_TOF EAR-2. We will use a set of four C_6D_6 detectors for detecting the capture γ-rays. These detectors have been used successfully in 2015 for the ^{171}Tm(n,γ) [8] and ^{147}Pm(n,γ) [9] cross section measurements. The samples will be placed in the sample exchanger, in air, with the minimum amount of dead material intercepting the neutron beam along the beam line. The neutron beam intensity will be monitored with neutron transparent silicon flux monitor [10].

Isotope	^{244}Cm sample	^{246}Cm sample
^{240}Pu	184 µg	168 µg
^{244}Cm	412.6 µg	380 µg
^{245}Cm	14.3 µg	19.7 µg
^{246}Cm	38.3 µg	1098 µg
^{247}Cm	0.98 µg	54.0 µg
^{248}Cm	0.72 µg	172 µg

Table 1 Estimated masses of the actinides present in one ^{244}Cm sample and in the ^{246}Cm sample.

3 OBJECTIVES AND BEAM TIME REQUEST

The number of capture signals and the magnitude of the different background components detected in the C_6D_6 detectors has been carefully estimated, including the capture, elastic and fission reactions of the isotopes listed in Table 1, the background due to elastic and capture reactions in the aluminum powder and canning, the γ-ray background due to the ^{244}Cm spontaneous fission (SF), and the EAR-2 background related to the neutron beam (with no sample in place), determined experimentally from the ^{171}Tm(n,γ) and ^{147}Pm(n,γ) measurements done in 2015. The estimated time of flight (TOF) spectra for the ^{244}Cm(n,γ) and ^{246}Cm(n,γ) measurements are shown in Figure 1. The resonance at 1 eV corresponds to the ^{240}Pu isotope, which will be used to normalize the ^{244}Cm and ^{246}Cm capture cross sections.

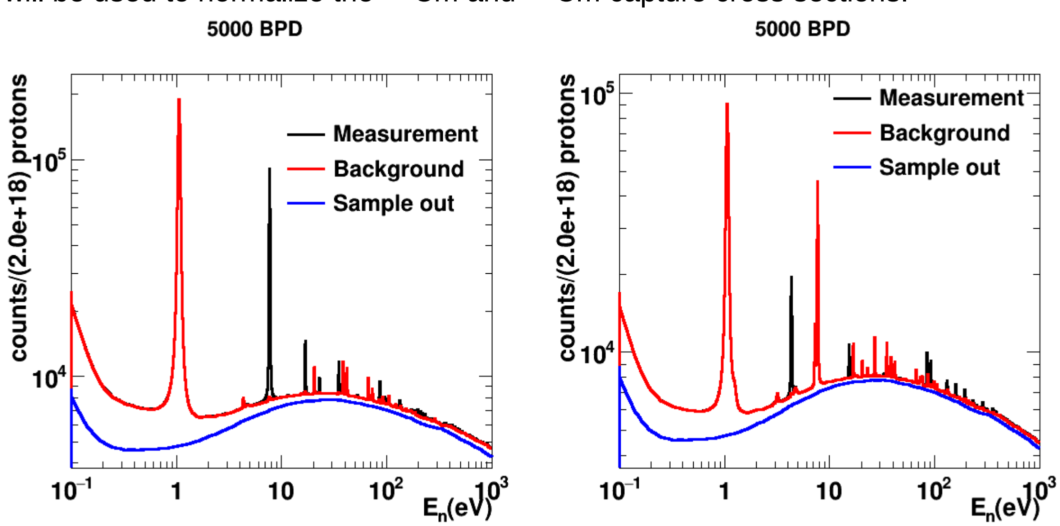

Figure 1 Expected number of counts in the ^{244}Cm (left) and ^{246}Cm (right) measurements for a total of 2×10^{18} protons (each), using 5000 bins per decade (BPD). In black, all counts registered in the four C_6D_6 detectors. In red, all counts not coming from $^{244/246}$Cm(n,γ) reaction. In blue, all the counts coming from the background when no sample is placed in the beam.

The (n,γ) cross sections will be obtained by analyzing the individual resonances seen in the corresponding TOF spectra. The accuracy in the cross section will be limited, to a lower extent, by the statistical uncertainties, which will depend on the number of capture reactions detected (i.e. area of the measured resonances), and mostly by the systematic uncertainties, which will depend on how accurate is our knowledge of all the other parameters needed to determine the cross section: sample mass, background corrections, methodology of the pulse height weighting technique, determination of the background or the shape of the resolution function

The estimated overall systematic uncertainty will amount to ~ 7-8%, where the determination of the sample mass from the Pu/Cm mass ratio (4.5%) made by α-spectrometry [3], the pulse height weighting technique (1-2%) and the effect of the background (1-2%) will be the largest components. Thus, in order to determine the ^{244}Cm(n,γ) and ^{246}Cm(n,γ) cross sections with an accuracy better than 10% it will be necessary to keep the statistical uncertainties well below 5%.

Figure 2 show the statistical uncertainties as a function of the resonance energy E_{n},. Three different zones have been marked to highlight the problems in the analysis of the data: between 1 eV and 250 eV (zone 1), where the measurement will allow to get resonance integrals with an overall statistical uncertainty below 5%; between 250 eV and 550 eV (zone 2), where the results obtained in this region will depend largely on the real value of the (n,γ) cross sections and the real background in the measurement; and above 550 eV (zone 3), where with the information available at present it seems impossible to obtain a cross section value in that region with the available sample mass.

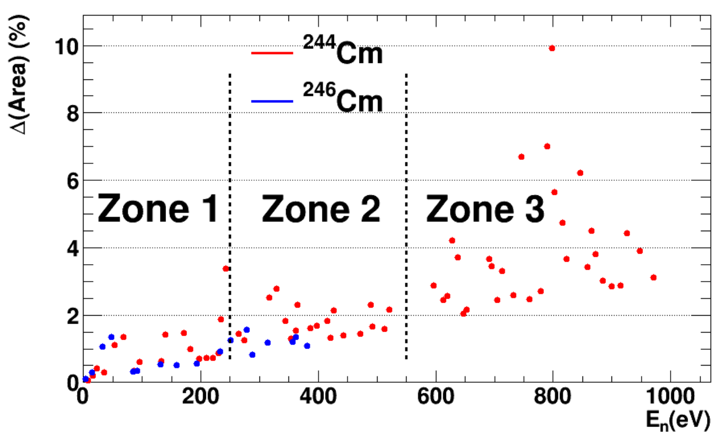

Figure 2 Estimated statistical uncertainties in the area of the resonances with energies E_{n}, for a measurement at EAR-2 performed with 2x10^{18} protons. Red points: known resonances in ^{244}Cm+n reactions [11]. Blue points: known resonances in ^{246}Cm+n reactions.

At the view of these results, 2x10^{18} protons are requested for the ^{244}Cm(n,γ) measurement. Such a value will allow to determine the resonance integrals with an uncertainty better than 5% up to 250 eV and to provide data up to 550 eV (i.e. Zone2) with an uncertainty in the range of 10% - 20% for an uncertainty in the normalization of the background better than 1%. The high statistics in Zone 1 is required in order to have a control over the dominant sources of systematic uncertainty: the normalization of the background, the effect of the resolution function and the use of the pulse height weighting technique necessary to correct the C_6D_6 data. The results in Zone 2 will depend largely on the real experimental conditions to be found during the experiment and the real cross section value.

The available information for ^{246}Cm is scarcer than for ^{244}Cm. The reported resonance region is only known up to 400 eV. However, the statistical model of this nucleus allows to conclude that its structure must be, on average, analogous to ^{244}Cm, with similar level density parameters and average neutron resonance parameters. Thus, the same number of protons should be allocated for the ^{246}Cm measurement.

The background in the data will be high in comparison to the signals from the (n,γ) reactions, mainly due to the very low masses of the samples (~1 mg). In addition, the peak to background will be degraded significantly due to the resolution function above 100 eV. In order to minimize the effect of the background subtraction (i.e. fitting of the resonances on top of the smooth background distribution) it is necessary to determine the background with good statistics. For this reason, 10^{18} protons are requested to determine experimentally each component of the background with statistical fluctuations comparable to the background present in the capture TOF spectra.

In conclusion, we request 2.0×10^{18} protons to measure each of the ^{244}Cm and ^{246}Cm samples and 10^{18} protons for each dedicated background measurement: "dummy sample" measurement (a sample in place similar than the one with the Cm but without the Cm) and "sample out" (same measurement but without any sample in place, to determine the background not related with the sample). The response of the C_6D_6 detectors to the large intrinsic activity in absence of beam will also be measured. In addition, as a cross check we will monitor the integral neutron fluence impinging on the samples with the 4.9 eV saturated resonance in the ^{197}Au(n,γ) cross section. We will also determine experimentally the neutron sensitivity of the setup (graphite sample) coming from the spallation target. A summary of the different measurements and number of protons requested for the experiment at EAR-2 is presented in Table 2.

4 Additional measurement at EAR-1

We propose as well to perform a short measurement with the TAC [14] in EAR-1 of the strongest resonances of ^{240}Pu and ^{244}Cm, in order to have an additional normalization of the ^{244}Cm capture cross section with respect to the ^{240}Pu capture cross section with an independent analysis technique. In addition, we will obtain spectroscopic information about the γ-ray cascades following the ^{244}Cm(n,γ) and ^{240}Pu(n,γ) reactions, which is of great value for the determination of the statistical properties of the two compound nuclei and will serve to tune the electromagnetic de-excitation models used during the analysis to estimate different types of corrections: loss of counts due to the energy threshold, conversion electrons, the presence of isomers…

The number of protons required to perform this measurement has been calculated in order to obtain the deposited energy spectra in the TAC of the ^{244}Cm(n,γ) and ^{240}Pu(n,γ) cascades with a reasonable accuracy (100 bins with an averaged statistical uncertainty of 3%). The estimated number of protons amounts to 4.0×10^{17} protons to measure the ^{244}Cm sample and 1.0×10^{17} protons for measuring the background. A similar measurement with the ^{246}Cm sample seems to be unrealistic due to the weaker ^{246}Cm largest resonance. The summary of the number of protons requested for the measurement in EAR-1 is given in Table 3.

5 SUMMARY OF REQUESTED PROTONS

Sample	Purpose	Protons
^{244}Cm(n,γ)	capture cross section measurement	2.0×10^{18}
^{246}Cm(n,γ)	capture cross section measurement	2.0×10^{18}
Dummy – ^{244}Cm	Canning related background	1.0×10^{18}
Dummy – ^{246}Cm	Canning related background	1.0×10^{18}
Sample out	Beam related background	1.0×10^{18}
Graphite	Neutron Sensitivity	0.5×10^{18}
^{197}Au(n,γ)	Validation of the capture measurement	0.5×10^{18}
^{244}Cm/^{246}Cm/-	No beam related background	0
Total		**8.0×10^{18}**

Table 2 Number of protons requested for each measurement in EAR-2.

Sample	Purpose	Protons
^{244}Cm(n,γ)	Capture cascades of the strongest ^{240}Pu and ^{244}Cm resonances	0.4×10^{18}
Dummy – ^{244}Cm	Beam related background	0.1×10^{18}
^{244}Cm	No beam related background	0
Total		**0.5×10^{18}**

Table 3 Number of protons requested for each measurement in EAR-1.

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