

Measurement of mass yields from the $^{241}\text{Am}(2n_{\text{th}},f)$ reaction at the Lohengrin Spectrometer

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Abstract. The study of fission yields has a major impact on the characterization and understanding of the fission process and is mandatory for reactor applications. While the yields are known for the major actinides (^{235}U , ^{239}Pu) in the thermal neutron-induced fission, only few measurements have been performed on ^{242}Am . The interest of ^{242}Am concerns the reduction of radiotoxicity of ^{241}Am in nuclear wastes using transmutation reactions. This paper presents the measurement of the fission mass yields from the reaction $^{241}\text{Am}(2n_{\text{th}},f)$ performed at the Lohengrin mass spectrometer (ILL, France) for both the light and the heavy peaks: a total of 41 mass yields have been measured. The experiment was also meant to determine whether there is a difference in mass yields between the isomeric state and the ground state as it exists in fission and capture cross sections. The method used to address this question is based on a repeated measurement of a set of fission mass yields as a function of the ratio between the ^{242g}Am and the ^{242m}Am fission rates. The presented experiment is also a first step towards the measurement of the isotopic fission yields of ^{242}Am .

1 Motivations

The fission of the ^{242}Am nucleus is interesting for both applications and fundamental studies. It is produced by radiative capture on ^{241}Am , which is the main responsible of the radiotoxicity of Plutonium-separated nuclear wastes between 200 and 1000 years after shut-down of a PWR reactor. The best way to reduce its radiotoxicity is to transmute it either in thermal or fast reactors, which consequently leads to the fission of ^{242}Am . While yields are known for major actinides (^{235}U , ^{239}Pu) in thermal neutron-induced fission, only few measurements have been performed on ^{242}Am [1-3]. Moreover, the two main data libraries (JEFF-3.1.1 and ENDF/B-VII.0) do not agree among each other on the light peak. In addition, ^{242}Am has a spin-isomer which lives 141 years while the ground state lives only 16 hours. The measured fission cross sections are 6856 ± 656 barn for the isomer and 2644 ± 281 barn for the ground state [4-5]. This reveals that the entrance channel of the reaction is affected by the spin; one of the remaining questions is consequently the influence of the spin on the final state (i.e. fission yields, kinetic energy of the fission products,...).

2 The Experiment

The double capture reaction was used to overcome the impossibility of using an ^{242}Am target and to produce ^{242}Am in both its isomeric ($T_{1/2}=141\text{y}$) and ground state ($T_{1/2}=16.02\text{h}$). Because of the rather low reaction rate in the double capture process, a high neutron flux is required. The experiment was performed at the High Flux Reactor (RHF) of the Institut Laue-Langevin (ILL) in Grenoble (France) which provides the highest thermal neutron flux in the world for on-line fission studies ($5 \cdot 10^{14} \text{ n.cm}^{-2} \cdot \text{s}^{-1}$) combined with a mass and energy separation of fission products thanks to the mass spectrometer Lohengrin.

A target, made by a Ti backing, a layer of ^{241}Am ($300\mu\text{g/cm}^2$ and more than 99% in concentration) and a nickel foil ($0.25\mu\text{m}$) set after the active matter to prevent sputtering, is placed in H9 (a beam line of RHF) 50 cm away from the core. The fission products fly through the beam pipe under vacuum to reach a mass spectrometer called Lohengrin [6], composed of a magnet followed by an electrostatic deflector. The fission products reach the magnet with a given mass A , kinetic energy E and ionic charge q . One has to note that the energy at this point is the energy at fission after prompt neutron emission (which is never directly accessible) minus the energy lost in the target and cover foil.

Fission products are produced stripped in the fission process ($q=Z$) and they capture electrons as they go through matter. The magnet selects nuclei according to their momentum over ionic charge ratio (Av/q). After the magnet, an electrostatic deflector selects nuclei according to the kinetic energy over ionic charge ratio (E/q). An ionization chamber [6] is set-up at the focal plane of the spectrometer to count the fission products. This chamber measures the energy of the fission product so, thanks to the selection done by the spectrometer, we deduce the mass of the fission product (A).

3 Data analysis and uncertainties

As Lohengrin only selects nuclei according to their given ionic charge q and energy E ($Y(A,q,E)$), to obtain the production probability of a fission product with a given mass A ($Y(A)$) we need to integrate over charge and energy: $Y(A) = \int \sum_q Y(A,q,E) dE$.

Assuming that there is no correlation between q and E , the number of points needed to determine correctly the integral can be dramatically reduced. Two distributions are then sufficient: the E -distribution at a given charge (E -Scan) and the q -distribution at a given energy (q -Scan), usually the most probable one. The product of the integral of these two distributions divided by the value at the common point gives $Y(A)$. In reference [7] the influence of the correlation between energy and ionic charge on the yield values was studied and estimated to be less than 3%.

In order to obtain the fission yields, the number of fission products measured at a given mass should also be normalized to the number of fissions that occur in the target during the measurement. As this number cannot be directly measured at Lohengrin, the chosen procedure is to normalize all the measurements to the fission yield of a given mass (here $A=105$), typically the most produced one, which is measured every 8 or 12h. As a consequence, only relative fission yields are obtained.

The Table 1 resumes the different sources of uncertainties and their contribution. As no analytical function was found to describe precisely the energy distribution (obtained for a given mass and a given ionic charge) with a reasonable number of parameters, a quadratic interpolation between the data points and a linear extrapolation on the edges were used. These extrapolations lead to systematic uncertainties. They have been estimated as the maximum fluctuation of the extrapolated part contribution to the total distribution area observed in the set of energy distributions available for $A=105$. Systematic uncertainty was evaluated from the dispersion of the values measured for the same (q,E) in the q -scan and E -scan. This point has the lowest statistical error (<1%). Such a comparison was performed systematically as a function of time for three masses ($A=105$, $A=98$, $A=136$) and measured once for all the measured masses. Even if no common bias in the mean value

can be observed, the standard deviation of 3% cannot be explained only by statistics and is consequently considered as systematic uncertainty. Another possible source of uncertainty is a change in the neutron flux that can occur between two repeated measurements of a given mass every 8 or 12h. This flux was monitored by ^3He detectors and no variation above the statistical ones was observed over a mass measurement cycle.

Table 1. Sources of relative uncertainties and their respective contributions.

Source	Contribution
Statistical	~1 %
Extrapolation of the low part of the energy distribution	1.5 %
Extrapolation of the high part of the energy distribution	1%
Discrepancies between the two measurements of the common point	3%
Total of the systematic error (preliminary)	3.5%

4 Physics Results

4.1 Kinetic energy distributions

Due to a rather large energy variation (up to 7 MeV) during the experiment which we believe is a consequence of a degradation of the target and its cover foil, only relative kinetic energies could be obtained. Since this distribution is not Gaussian, the most probable value is taken as kinetic energy. The associated uncertainty is determined by the following two different effects. A first uncertainty of 0.6 MeV is due to the determination of the kinetic energy from the measured energy distribution. Secondly, an error of 0.6 MeV is due to the correlation between the energy and the ionic charge. This error has been estimated from the data obtained in references [3,8].

We used the kinetic energy value of the mass 105 measured by U. Güttler at the Lohengrin Spectrometer in 1991 [3] to shift our data. The results are plotted on Figure 1 showing a very good agreement with the past measurement.

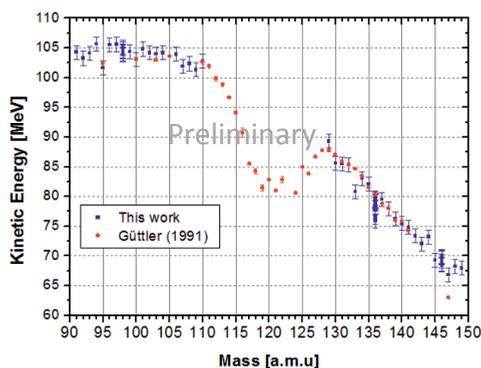


Fig. 1: Kinetic energy of the fission products as a function of their mass. The energies obtained in this work are normalized to the kinetic energy of the mass 105 measured by Güttler [3].

4.2 Fission Yields

The preliminary result of our measured fission mass yields is shown on Figure 2. Each mass yield is normalized to the yield of the mass 105 ($Y_{105}=6.5\%$, according to JEFF-3.1.1). The experimental

results have been compared with the most commonly used nuclear data libraries, JEFF-3.1.1 and ENDF/B-VII.0 as well as with the experimental data obtained by Wofberg in 1971 [1] (on which the evaluations are mainly based). As we see on Figure 2, the heavy mass region shows the largest discrepancies. Concerning the light peak, our measurement is closer to the ENDF/B-VII library. Finally, our results are in good agreement with the yields measured by Wofberg et al. for the fission of ^{242m}Am .

It should be noted that the mass 95 seems to present too large yields.

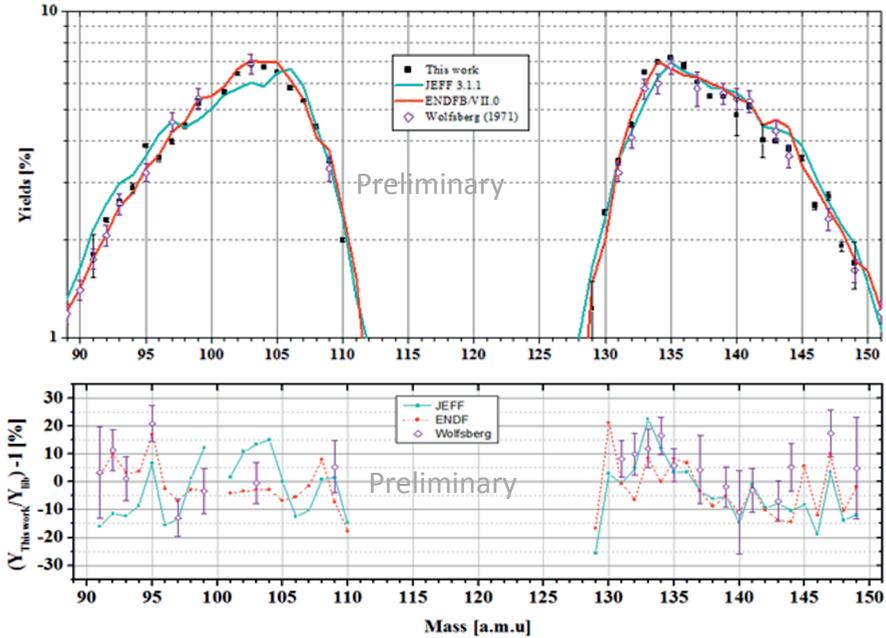


Fig. 2: (Top) Fission mass yields of ^{242}Am compared with JEFF-3.1.1 and ENDF/B-VII.0 libraries as well as the experimental data from Wofberg [1]. (Bottom) Relative difference between the measured yields and the ones given by JEFF-3.1.1, ENDF/B-VII.0 and Wofberg. Only the statistical error for our points is reported here.

4.3 Comparison between the fission yields of the ground state and the isomer

As mentioned before, this measurement was partly meant to determine whether there is a difference in mass yields between the fission of the isomeric state and the ground state of ^{242}Am as it exists in fission and capture cross sections. The method used to address this question is based on the repeated measurement of a fixed set of fission mass yields as a function of time. This allows being sensitive to the variation of the ratio between the $^{242g}\text{Am}(n,f)$ and the $^{242m}\text{Am}(n,f)$ fission rates as shown on Figure 3. The ratio between the yields does not seem to be influenced by the evolution of the ratio between the fission rates. Two sets of points do not follow the flat systematic: the first set of points, noted A on Figure 3, can be explained by a very quick variation of the properties of the target and the second set, noted B, by an observed vacuum problem in the beam-line. Given the small fluctuations (less than 3%) of these ratios ($Y(A)/Y(A')$) and the precision of around 4% for the relative fission yields, we have deduced an upper limit following a procedure not detailed in this paper. As a consequence, we can only conclude that the fission mass yields generated by the neutron-induced fission of ^{242g}Am and those due to $^{242m}\text{Am}(n,f)$ do not differ by more than 45%. This limit can be reduced to 25% for the masses for which measured fission yields are available in the literature for the $^{242m}\text{Am}(n,f)$ fission reaction [1].

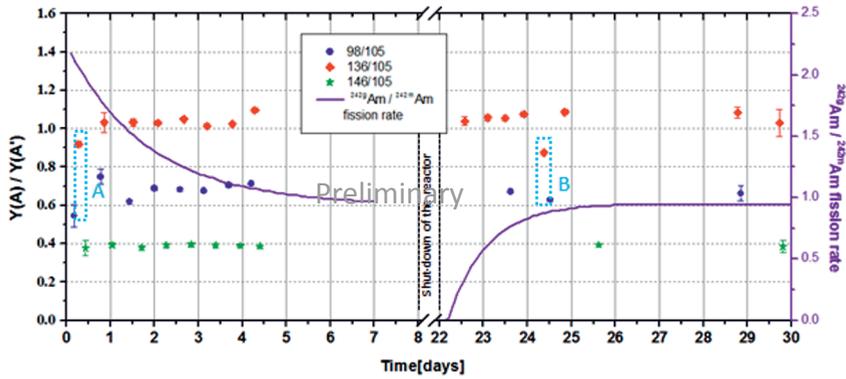


Fig. 3: Fission mass yield ratios as a function of time (left scale) and corresponding evolution of the ratio between the $^{242g}\text{Am}(n,f)$ and the $^{242m}\text{Am}(n,f)$ fission rates (right scale).

5 Conclusion & Perspectives

The presented measurement of the $^{241}\text{Am}(2n_{th},f)$ fission yields shows that our results largely improve the experimental data on the mass yields by enlarging the range of measured mass and by reducing the experimental uncertainties. These results are in better agreement with the ENDF/B-VII library in the light mass region and are consistent with JEFF-3.1.1 and ENDF/B-VII in the heavy one. This preliminary analysis also led us to establish an upper limit of 25 % in the difference between the mass yields coming from the fission of the two nuclear states of ^{242}Am . Even if the mass fission yields are of great interest on themselves, they are also a step towards the measurement of isotopic yields. These are needed for applications as well as for theory. The experimental set-up used to measure the isotopic yields will be realized by coupling two HPGe clover detectors to the spectrometer in order to perform gamma spectrometry of mass-separated beams. Such a method has already been applied successfully to measure isotopic yields of heavy fission products in $^{239}\text{Pu}(n_{th},f)$ [7-8].

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