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

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Résumé. 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. This paper presents the results of a measurement at the Lohengrin mass spectrometer (ILL, France) on the reaction ^{241}Am($2n_{th}$,f): a total of 41 mass yields in the light and the heavy peaks have been measured and compared with the fission process simulation code GEF. Modus operandi and first results of a second experiment performed in May 2013 on the same reaction but with the goal of extracting the isotopic yields are presented as well: 8 mass yields were re-measured and 18 isotopic yields have been investigated and are being analyzed. Results concerning the kinetic energy and its comparison with the GEF Code are also presented in this paper.

1. Motivations

Among the actinides presenting an interest for fundamental research and applications, ^{242}Am is a very good example of an high radiotoxicity odd-odd short-lived nucleus. ^{242}Am possesses also two long-lived states: a high-spin isomer (5^-) with a half-life of 141 years and the ground state (1^-) with a half-life of 16 hours. The measured fission cross sections are (5972 ± 173) barn for the isomer and (1751 ± 55) barn for the ground state [1]. This reveals that the entrance channel of the fission 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, ...). No data are nowadays available for the comparison of the final state issued from the fission of a nucleus in its (spin) isomeric state and in fundamental state. In a phenomenon where odd-even effects are present it is also interesting to notice that the most studied nuclei are nuclei with an even nuclear charge (^{235}U, ^{239}Pu, ^{233}U, ^{252}Cf) whereas

$Z_{Am} = 95$, the only other odd charge nucleus where thermal neutron induced fission had been studied in detail before was ^{237}Np.

Nuclear waste management is another motivation for the studies on the fission of ^{242}Am. ^{242}Am is produced by radiative capture on ^{241}Am, which is the main responsible of the radiotoxicity of Plutonium-separated nuclear waste from PWR reactors between 200 and 1000 years after irradiation. The best way to reduce its radiotoxicity is to transmute it, which consequently leads to the fission of ^{242}Am. Fission of ^{242}Am, like of any other actinide, produces fragments with large neutron cross-sections that are considered as neutron poisons for reactor operation as well as delayed neutrons, which play a role in a reactor control. A precise estimation of their quantity is important to design dedicated transmutation facilities where the fraction of minor actinides is not negligible any more.

2. Experiment

The double capture reaction was used to overcome the impossibility of using a ^{242}Am target and to produce ^{242}Am in both its isomeric ($T_{1/2} = 141$ y) and ground state ($T_{1/2} = 16.02$ 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 ($5 \cdot 10^{14}$ n $\cdot$ cm$^{-2} \cdot$ s^{-1}) combined with a mass and energy separation of fission products thanks to the mass spectrometer Lohengrin.

The target is placed 50 cm away from the core of the reactor. It consists in a 300 μg/cm^2 thick deposit of ^{241}Am (purity $> 99\%$) on a Ti backing and is covered by a 0.25 μm thick nickel foil that prevents a too large loss of fissile material by fission fragment sputtering. The fission products fly through a beam pipe under vacuum to reach the mass spectrometer called Lohengrin [2], composed of a dipole magnet followed by an electrostatic deflector. The magnet deflects the nuclei according to their momentum over ionic charge ratio (Av/q) while the electrostatic deflector allows their selection according to the kinetic energy over ionic charge ratio (E/q). The kinetic energy considered in this selection process is the energy of the fission product, after prompt neutron emission and minus the energy lost in the target and the cover foil. Fission products are produced stripped in the fission process ($q = Z$) and capture electrons as they go through matter. At the exit of the cover foil, they generally present an ionic charge close to $q = 20$-22.

A second magnet called RED Magnet (Reverse Energy Dispersion) allows to switch between an ionization chamber in straight direction (mass measurements) or refocus into a vacuum chamber with a tape where fragments are implanted (isotopic measurements). The ionization chamber [2] measures the energy of the fission product so, thanks to the E/q and A/q selection done by the spectrometer, we know q and deduce the mass of the fission product (A). When the fragments are deflected and implanted on a tape, γ rays produced after beta decay are registered with two high-efficiency HPGe clover detectors placed around the implantation position. Knowing the γ intensity, isotopic yields can be deduced. The same set up was previously used to measure the isotopic yields of ^{239}Pu(n_{th},f) and ^{233}U(n_{th},f) [3–5].

3. Data analysis and uncertainties

As Lohengrin selects nuclei according to their given ionic charge q and energy E, to obtain the yield of a fission product with a given mass A (Y(A)) one needs to integrate the measured fragments differential yields Y(A,q,E) over charge and energy:

$$Y(A) = \int \sum_{q} Y(A,q,E) dE.$$

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%
Reproducibility	3%
Correlation between E and q	3%
Relative Normalisation (Burn-up)	∼3%
Total of the systematic error	5.5%

Assuming that there is no correlation between q and E, this integral can be rewritten as the product of the energy distribution (E-Scan) measured at a fixed ionic charge $Y(A,q_0,E)$ and the ionic charge distribution (q-scan) at a fixed energy $Y(A,q,E_0)$, divided by the differential yield at the common point $Y(A,q_0,E_0)$. The number of measurements can thus be drastically reduced to an E-scan and a q-scan. The influence of the correlation between energy and ionic charge distribution on the yield values was studied in reference [3] and estimated to add less than 3% on the relative uncertainty. Table 1 resumes the different sources of uncertainties and their contribution. As no analytical function was found to describe precisely the energy distributions 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 that have been estimated from the maximum fluctuation of the extrapolated part contribution to the total distribution area observed in the set of E-scans (around 30) available for $A = 105$.

The reproducibility uncertainty was evaluated from the dispersion of the values measured for the same $Y(A,q_0,E_0)$ during 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 is observed, a standard deviation of 3% cannot be explained only by statistics and is consequently considered as systematic uncertainty.

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, relative fission yields are obtained. The mass used for the relative normalization is affected like all the other masses by the same uncertainties as mentioned before.

Another possible source of uncertainty is a change in the neutron flux that can occur between two normalization measurements. This possibility was evaluated by monitoring the neutron flux with ^{3}He detectors. No variation above the statistical ones was observed over a mass measurement cycle.

4. Physics results

4.1 Kinetic energy distributions

Due to a rather large variation of the kinetic energy during the experiment (a shift of up to 7 MeV was observed on the energy distribution made at $A = 105$) which we believe is a consequence of a

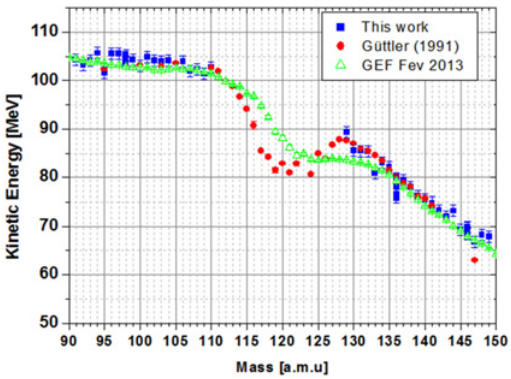

Figure 1. Kinetic energy of the fission products as a function of their mass. The energies obtained in this work are normalized to the kinetic energy of the mass 105 measured by Güttler and compared to the GEF predictions.

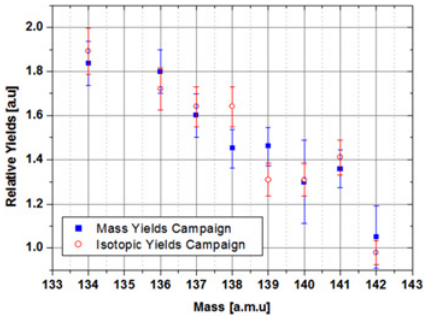

Figure 2. Fission Mass Yields obtained in the two measurement campaigns. The normalization of each set was performed on the sum from A = 136 to A = 142.

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 originates from the following two different effects. A first uncertainty of 0.6 MeV is due to the determination method of the most probable value 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, 6].

We used the kinetic energy mean value of the mass 105 measured by U. Güttler at the Lohengrin spectrometer in 1991 [6] to shift our data. The results are plotted on Figure 1 showing a very good agreement with the past measurement.

The two measurements are also compared with the GEF Code prediction [7]. While the GEF predictions agree with the experiments on the asymmetric mass region it presents a shift in the symmetric region. The same behaviour was observed for ^{235}U but not on ^{239}Pu neither on ^{233}U.

4.2 Mass fission yields

During the isotopic yield measurement campaign, eight mass yields were re-measured on the heavy peak. The comparison with the values obtained in the mass yield campaign (Fig. 2) shows that the new measurements agree with the previous ones, proving the reproducibility of the measurement.

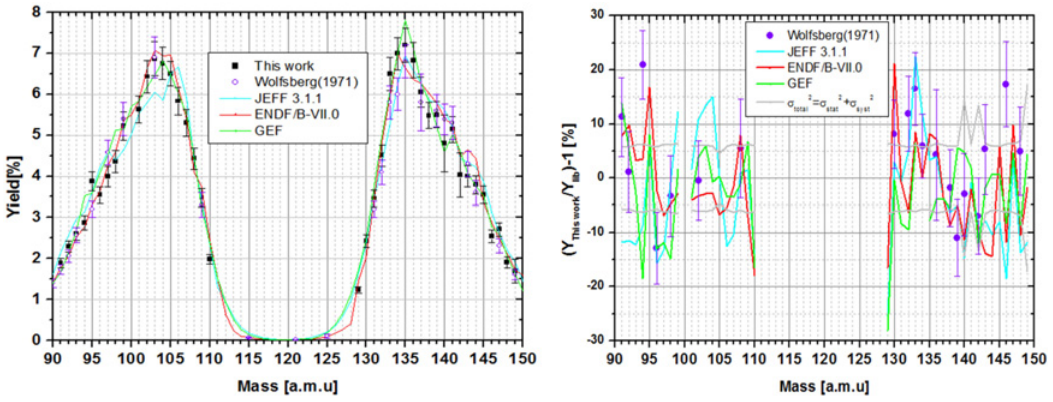

Figure 3. (Left) fission mass yields of ^{242}Am compared with JEFF-3.1.1 and ENDF/B-VII.0 libraries as well as the experimental data from Wolfsberg [7] and the GEF model. (Right) relative difference between the measured yields and the ones given by JEFF-3.1.1, ENDF/B-VII.0 and Wolfsberg.

Table. 2. List of mass chain nuclei measured. The green symbols indicate the nuclei from which we can obtain an independent isotopic yield and the red crosses the ones we cannot. (Preliminary results).

A\Z	Sb	Te	I	Xe	Cs	Ba	La
134	✓(m) X(gs)	✓	✓	X	X	X	X
136	X	✓	✓	X	X	X	X
137	X	X	✓	✓	X	X	X
138	X	X	✓	✓	✓	X	X
139	X	X	X	✓	✓	X	X
140	X	X	X	✓	✓	X	X
141	X	X	X	X	✓	✓	X
142	X	X	X	X	✓	✓	~

The preliminary results of our measured mass yields are shown on Figure 3. 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 Wofsberg in 1971 [8] on which the evaluations are mainly based. As shown on Figure 3, the largest discrepancies occur in the heavy mass region. Concerning the light peak, our measurement is closer to the ENDF/B-VII.0 library. It has to be noticed the good agreement within the quoted uncertainties with the GEF code. Finally, our results agree with the yields measured by Wolfsberg et al. for the fission of ^{242m}Am. It should be noted that the mass 95 seems to present a large yield which is not compatible with the evaluated data.

4.3 Isotopic fission yields

The isotopic yields were measured for 8 masses: 134, 136, 137, 138, 139, 140, 141 and 142. According to a preliminary analysis, we will be able to extract yields for the nuclei listed in Table 2. The fission yields of the very light tail were measured in [9] and show evidence of an odd-even effect. However according to previous measurement on ^{241}Am(n,f) and ^{243}Am(n,f) this effect is not expected for the mass peak regions [10]. A previous experiment using the Lohengrin spectrometer with an absorber [11] studied the odd-even effect for proton and neutron on the light peak. An odd-even effect was observed on neutron but not on proton. The study of the heavy mass region was performed in this experiment.

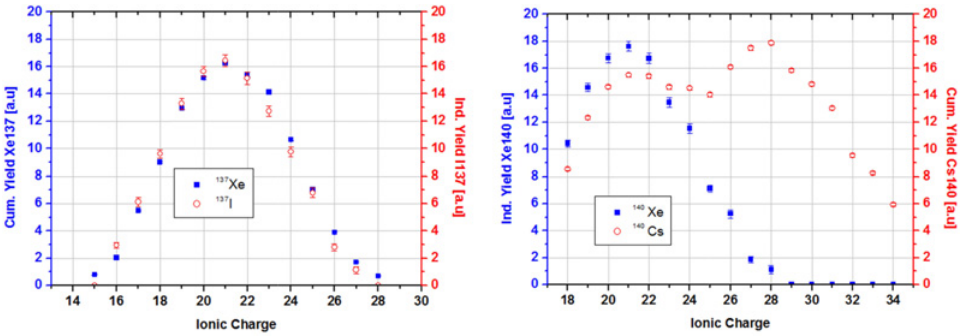

Figure 4. Ionic charge distribution for different isotopes. ^{137}Xe and ^{137}I (left part) and ^{140}Xe and ^{140}Cs (right part).

In contrast to the procedure followed in most of the past experiments at Lohengrin (including the one described in [11]), during the present measurement campaign, the whole ionic charge distribution was measured. It is indeed known that the presence of nanosecond isomers disturbs the ionic charge distribution [12] and that it can affect strongly the estimation of the fission yields if this effect is not taken into account. For example, it was clearly the case for the Güttler experiment [6]. Since isomeric states are more found in odd-odd nuclei, it has an impact on the so-called odd-even effect observed in the fission yield. The impact (up to 80%) of the presence of nanosecond isomers is shown for A = 140 in Figure 4 (right); as a comparison a "normal" distribution for A = 137 is given in Figure 4 (left). We can see that, in the case of mass 137, the Xe and I present a Gaussian distribution while, for mass 140, Xe shows a Gaussian distribution but Cs is affected by a nanosecond isomers, characterized by a second Gaussian distribution with a mean charge of 28.

5. Conclusion & perspectives

The presented measurement of the ^{241}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.0 library in the light mass region and are consistent with JEFF-3.1.1 and ENDF/B-VII.0 in the heavy one. A good agreement was also found with the results of the GEF model. 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 application purposes as well as for the improvement and validation of fission models. A first campaign of measurement of the isotopic yields was performed at Lohengrin. According to the preliminary analysis, 18 isotopic yields are expected. Such a method has already been applied successfully to measure isotopic yields of heavy fission products in ^{239}Pu(n_{th}, f) [3, 4].

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