Measurement of mass yield of the reaction ^{241}Am(2n,f) at the Lohengrin Spectrometer

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1.1 Motivations

The fission process is the splitting of an excited heavy nucleus into at least two lighter nuclei [1]. Fission is characterized by different phases (see fig.1.1). First, there is the formation of the compound nucleus resulting from the capture of a neutron; then the nucleus evolved towards a deformed shape. This deformation phase is followed by scission, defined as the configuration where the two daughter nuclei properties (mass, nuclear charge, kinetic energy,...) are determined. he produced nuclei are called primary fragments. As they are neutron-rich and very excited, they emit neutrons, called prompt neutrons (around 2.5 on average for actinides) and γ. The resulting nuclei are called secondary fragments. As they are still neutron-rich, they follow a β-decay chain, typically three decays, to reach the stability valley. Some nuclei (around 200) emit also neutrons via β-n reactions. These are called delayed neutrons and are produced up to some seconds after fission. The probability of producing a given nucleus per fission is called fission yield. Because of the time scale of the process, only secondary fragments (not primary fragments) can be measured directly.

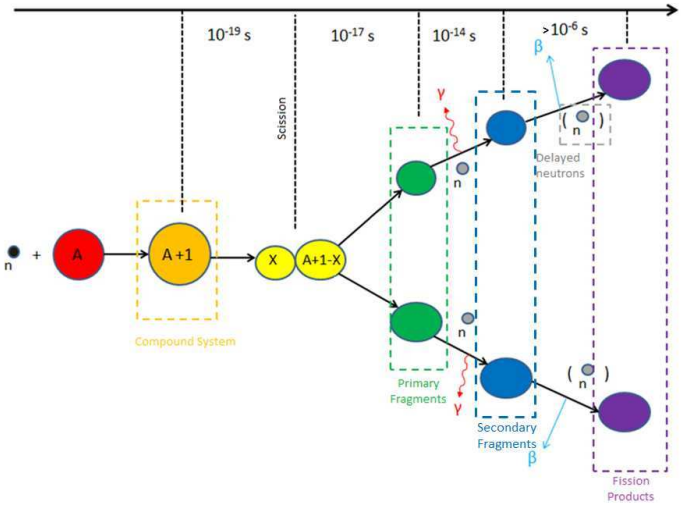

FIG. 1.1 – Schematic view of the time scale of the neutron-induced fission process.

Figure 1.2 shows typical fission fragments mass yields. We can clearly distinguish two peaks, showing that the most probable fission is asymmetric. This asymmetry is due to nuclear structure, especially the double magic nucleus ^{132}Sn which stabilizes the heavy peak around $A = 140$, the light peak is centered on the complementary nucleus(*i.e.* $A = 90$ for Uranium and $A = 100$ for Americium). In the case of the thermal neutron-induced fission, the symmetric splitting is three orders of magnitude less probable than the asymmetric one.

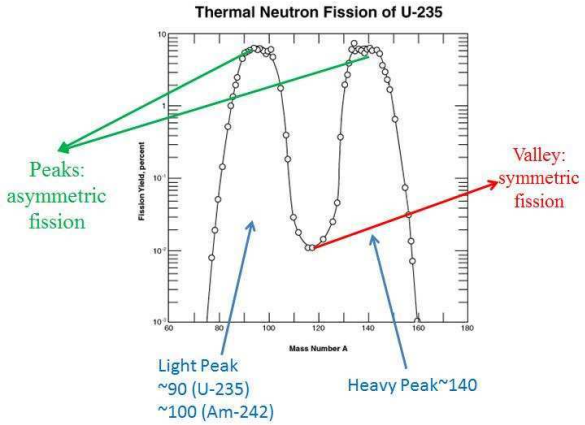

FIG. 1.2 – Mass yields of ^{235}U in thermal neutron-induced fission extracted from nuclear data library.

The fission of the ^{242}Am nucleus is interesting for both fundamental studies and applications. It 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 [2, 3]. This reveals that the entrance channel of the reaction is affected by the spin; therefore one of the remaining questions is the influence of the spin on the final state (*i.e.* fission yields, kinetic energy,...).

Moreover, ^{241}Am is the main responsible of the radiotoxicity of nuclear wastes between 200 and 1000 years if plutonium is extracted from spent fuel, a process followed in France. The best way to reduce its radiotoxicity is to transmute [1] it via neutron irradiation either thermal or fast which leads to the fission of ^{242}Am. Fission fragments being responsible for the production of β-delayed neutrons, poisons and heating after the shutdown of the reactor, their yields are of prime importance for studying fuel containing minor actinides.

[1] The transmutation is the conversion of one isotope into another.

1.2 Experimental setup

Because of the life time of its ground state, a target of ^{242}Am cannot be easily produced for direct irradiation. Hence, double capture on ^{241}Am (see figure 1.3) is necessary to access the fission of ^{242}Am. Because of the reaction cross sections involved in the double capture process, a high neutron flux is required. This is one of the main reasons why the experiment has been performed at the High Flux Reactor (HFR) of the Institut Laue-Langevin (ILL) in Grenoble (France).

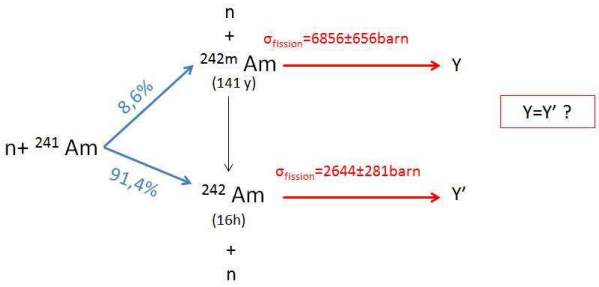

FIG. 1.3 – Schematic view of the double capture process on ^{241}Am.

The HFR (figure 1.4) produces the highest thermal neutron flux in the world (around $10^{15} n.cm^{-2}s^{-1}$). The core of the reactor is composed by highly enriched Uranium surrounded by a heavy water moderator. The neutron beam line used for the experiment is called H9. The flux in this beam line is essentially thermal (more than 95%). A target is set-up in H9 50 cm from the core. The target is covered by a nickel foil to prevent from sputtering (figure 1.5). Sputtering is a process where atoms are ejected from the target because of the crossing of very energetic fission fragments within the target material.

The fission fragments fly over the beam pipe (under vacuum) to reach a mass spectrometer called Lohengrin [4], composed of a magnet followed by a condenser (figure 1.4). The fragments 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 (which is never accessible) minus the energy lost in the target and in the nickel foil. Fission fragments are produced naked in the fission process ($q = Z$) and they capture electrons as they cross matter.

The magnet selects the nuclei according to the A/q ratio. After the magnet, a condenser selects the nucleus according to the E/q ratio. An ionization chamber is set-up at the focal plane of the spectrometer (figure 1.4) to count the fission fragments. This chamber measures the energy of the fragment so, thanks to the selection done by the spectrometer, we deduce the mass of the fragment. The mass resolution of the spectrometer is around 0.1 $a.m.u.$

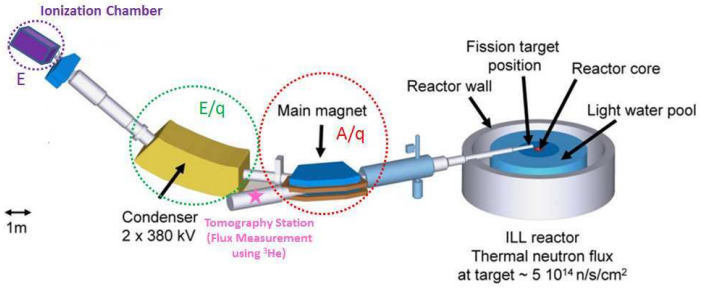

FIG. 1.4 – Schematic view of the ILL reactor and the Lohengrin spectrometer.[4]

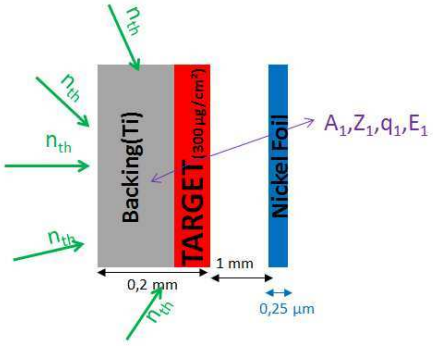

FIG. 1.5 – Sketch of the target, its backing and the nickel foil.

1.3 Data Analysis

1.3.1 Mass yields

As we only measure nuclei with a given ionic charge q and energy E ($Y(A,q,E)$), to extract the production probability of a fragment 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 there is no correlation between q and E, the number of needed points to determine correctly the integral can be dramatically reduced [5]. Two distributions are then necessary : the E-distribution at a given charge and the q-distribution at a given energy, usually the most probable one (figure 1.6).

If we consider the q distribution, two different cases may occur. As explained before, the ionic charge is determined by the crossing of ions through matter (mean charge at equilibrium $< q >= 21,22$); so we expect a Gaussian distribution (figure 1.7). Sometimes the nucleus can reach an isomeric state. It crosses the matter and de-excites by emitting electrons (internal conversion or Auger cascade) after crossing the nickel foil but upstream the magnet. The charge selected by the spectrometer is consequently higher ($q = 24 - 28$). The q distribution in this case has to be fitted by two Gaussians with the same width (figure 1.8), one centered on 21 and one on the average charge of the isomer af-

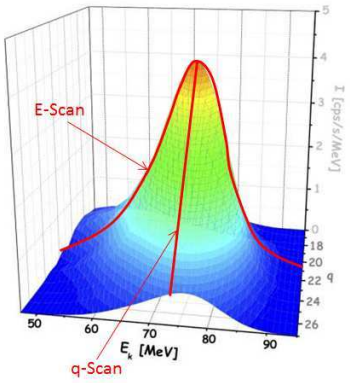

FIG. 1.6 – Correlation between the energy and ionic charge distributions of a fission fragment.

ter de-excitation. These isomers are called nano-second isomers since they have a short life time, typically $10\ ps - 1\ ns$ (these times are determined by the distance between the target and the nickel foil). In order to be detected they have to be produced in a reasonable quantity and de-excite via electron emission (typically coming from energy transitions of the order of $100\ keV$).

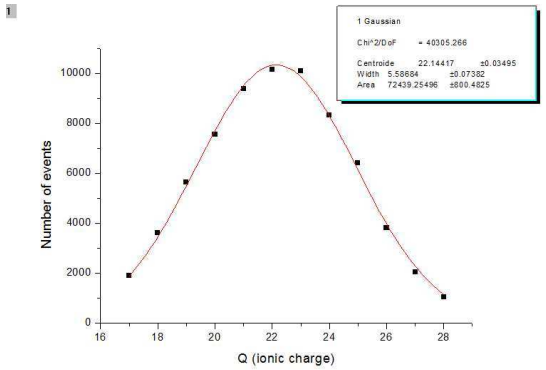

FIG. 1.7 – Ionic charge distribution of mass 136 at $E = 70\ MeV$

The E distribution is one of the most complex part of the analysis. This distribution is mainly Gaussian when the fragment is produced by fission. However, the crossing of matter (target and nickel foil) affects this distribution and this effect is dependent on the target and its evolution in time (burn up, sputerring,..). We try to fit this distribution by two Gaussians as it was done in the past [5]. As can be seen on figure 1.9, this fit function is not sufficient to correctly describe the energy distribution. We are presently studying two possibilities : trying to define a function based on physical considerations and able to fit our data or to simply make an analytical approach : interpolating between the points and extrapolating at low energy.

In order to obtain the fission yields, the number of fragments measured at a given mass should 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 procedure used to measure the burn up is the repeated measurement of a given mass, typically the most produced one, every 8 or 12h. Therefore, only relative fission yields are obtained. For fissile nuclei, when the burn up is only determinated by the fission process, the time evolution is a exponential (figure 1.10, left). Sometimes, the sputtering is important and changes sometimes dramatically the fission rate evolution (figure 1.10, right).

For double capture, as in our case, the expected burn up is more complicated (see figure 1.11).

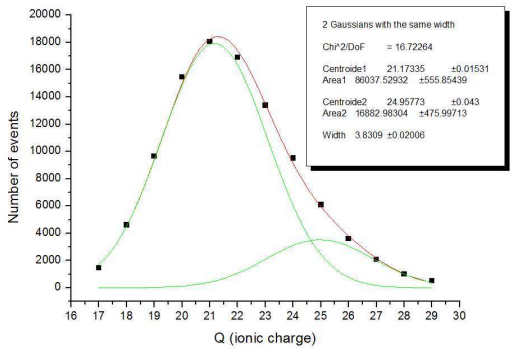

FIG. 1.8 – Ionic charge distribution of mass 105at $E = 96\ MeV$

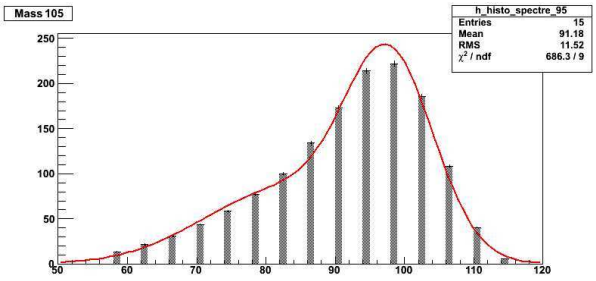

FIG. 1.9 – Energy distribution for $A = 105$ and $q = 21$.The fit function is the sum of 2 Gaussians. The error bars are only statistical.

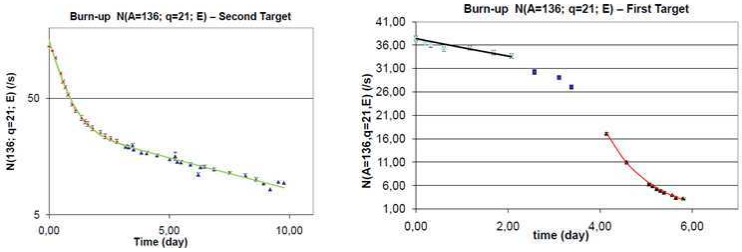

FIG. 1.10 – Example of "burn-up" on a fissile nucleus (^{233}U), with a small sputtering (on the left) and huge sputtering (on the right) [6].

The preliminary results are shown on figure 1.12.

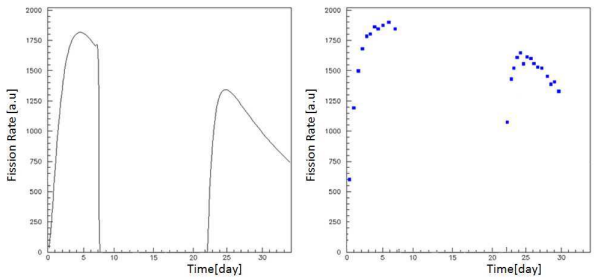

FIG. 1.11 – The solid line (on the left) represents the expected burn-up (calculated without sputtering) of the ^{241}Am target and the dots (on the right) are our measurements. Calculations and data are not normalized.

Each mass yield is normalized to the yield of the mass 105. The experimental results have been compared with the most commonly used nuclear data librairies, JEFF and ENDF. As we see in the figure, the heavy peak shows the largest discrepancies, as expected since very few measurements were performed in this mass region. However, our measurement is closer to the ENDF B-VII library in the light peak.

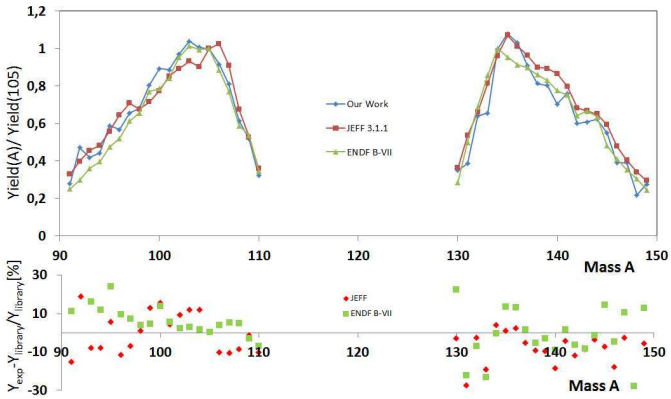

FIG. 1.12 – Comparison between the mass yields extracted from our data with the nuclear data libraries ENDF B-VII and JEFF 3.1.1.

1.3.2 ^{242g}Am and ^{242m}Am

As mentioned before, the use of the reaction ^{241}Am(2n,f) is the only way to obtain the fission yields of ^{242g}Am and ^{242m}Am separately (figure 1.3), which was one of the goal of the experiment.

If we measure the yield $Y_A(t)$ at two different times t_1 and t_2 and we are able to calculate the relative fission rate of ^{242g}Am ($\alpha(t)$) and the one of ^{242m}Am ($\beta(t)$), we can extract $Y_A(^{242g}Am)$ and $Y_A(^{242m}Am)$ by solving the following system of equations :

$$Y_A(t_1) = \alpha(t_1) * Y_A(^{242g}Am) + \beta(t_1) * Y_A(^{242m}Am) \quad (1)$$
$$Y_A(t_2) = \alpha(t_2) * Y_A(^{242g}Am) + \beta(t_2) * Y_A(^{242m}Am)$$

The figure 1.13 shows the calculated ratio $\alpha(t)/\beta(t)$ as a function of time during our experiment. During the first days, the ground state is responsible of the majority of the fission events. It is 10 times more produced than the isomer by capture on ^{241}Am and its fission cross section is only around 3 times smaller.

As the ground state quantity decreases very quickly ($T_{1/2}$= 16h), the fission rate of the ground state and the one of isomer, which has a higher fission cross section and a higher life time ($T_{1/2}(^{242m}Am)/T_{1/2}(^{242g}Am) =$ 7.7 10^4), become of the same order of magnitude.

During the shutdown of the reactor (15 days) all the ^{242g}Am in the target decays into ^{242}Cm. Then, when the measurement restarts, the target is principally made of ^{242m}Am and ^{241}Am. The fission rate is dominated by the isomer during the first hours and then it reaches the same order of the magnitude of the ground state.

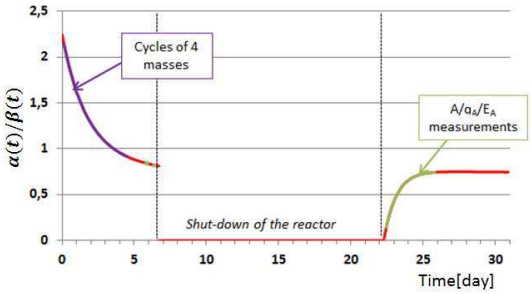

FIG. 1.13 – Ratio between ^{242g}Am and ^{242m}Am fission rates.

In order to calculate the ratio between the ^{242g}Am and the ^{242m}Am fission rates, we measured the neutron flux during the experiment. This flux monitoring allows evaluating the systematic uncertainty on the measurement due to the fluctuation of the flux during the experiment. This measurement was done with ^{3}He detectors placed inside a tomography station connected to the H9 beam line (figure 1.4).

Our neutron measurements have been compared to two fission chambers (PIL1 and PIL2) placed out of the reactor core. Our measurements were normalized to the mean value between PIL1 and PIL2 to see which fission chamber represented the best our measurement. The figure 1.14 shows that PIL1 and PIL2 have a rather different behavior before the shut down (upper plot). Our data are more consistent with PIL2 measurement which is the chamber closest to H9 beam line.

The data taken after the shutdown (lower plot) show a large discrepancy between our measurement and the fission chamber measurement during the first two days. A collaboration with the Simulations and Innovative Projects Service at ILL has begun to establish if this difference can be explained by the quick movement of the control rod during the first two days of the cycle or a movement of PIL1 and PIl2. We hope that this simulation will help to precise the actual flux value (the nominal flux : $5.10^{14} n.cm^{-2}s^{-1}$, is only a rough estimation).

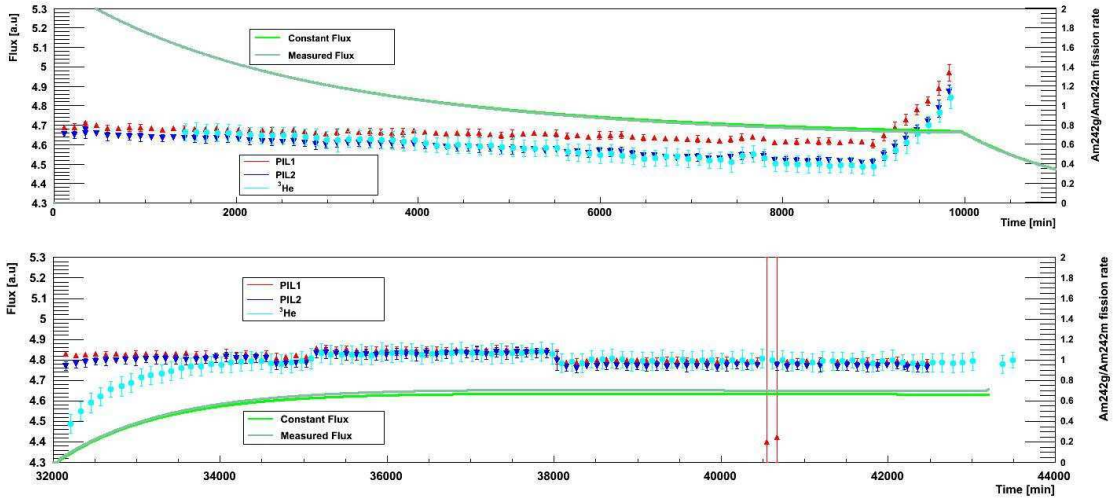

FIG. 1.14 – Time evolution of the measured neutron flux in the tomography station (^{3}He setup) and in the out-of-core fission chambers (PIL1 and PIL2). The upper plot refers to the irradiation before the shut down and the lower after the shut down. The solid lines represents the calculated fission rate ratio using a constant flux and the measured flux evolution.

1.4 Conclusion and perspectives

The analysis of ^{241}Am(2n,f) fission yields shows that our results are closer to the ENDF B-VII librairy in the light mass peak and are consistent in the heavy peak. The analysis should also lead us to understand if the two nuclear state of ^{242}Am have different fission yields. Even if the mass fission yields are of great interest on themselves, they are only a step towards the measurement of isotopic yields. These are needed for theory as well as for applications. The experimental set-up used to measure the isotopic yields will be realized by coupling two HPGe 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 fragments in ^{239}Pu(n$_{th}$,f) [5]. The experiment is planned in fall 2012.

Références

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