Use of active scintillating targets in nuclear physics experiments - Measurement of spontaneous fission

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Abstract. A novel detector has been used, in order to perform measurements of spontaneous fission to α-decay ratios for ^{240}Pu, ^{242}Pu and ^{252}Cf isotopes. The detectors are based on the well-known technique of liquid scintillating counting. The principle and advantages of the use of such detectors in nuclear physics is discussed. The application to the characterization of spontaneous fission is described and it is demonstrated that highly precise measurements are possible, and that the main limit is due to the isotopic content knowledge of the measured samples.

1 Introduction

Recently [1] a new kind of active target has been proposed in the context of nuclear physics experiments. It is based on the well-known technique of the liquid scintillating counting. This target can be used in the context of highly precise nuclear data measurements. Measurements of fission cross sections, spontaneous fission half-lives, prompt particle emission in fission, (n,xn) reactions or radiative capture cross sections can be envisaged. It can also be used in nuclear physics experiments whenever a veto or trigger for fission events is needed. For example it can be interesting in spectroscopy studies of fission fragments, since a fission trigger can suppress the large amount of γ-rays originating from radiative capture events. One of the advantages of such kind of active target is that fission fragments are stopped within a short time.

The liquid scintillation counting technique has been widely used for environmental assays of α and β emitters for a long time [2,3], and is still in development [4]. Nevertheless, very little is known on fission fragment detection by this technique [5,6]. In reference [5] it is claimed that the signal for fission fragments is similar to electron signals in toluene based scintillators, while in reference [6] it is claimed it is intermediate between electron and α-particle. In reference [1] we have shown that it is in fact intermediate between electrons and α-particle in a DIN based scintillator. We have also studied toluene based scintillators which exhibits the same behaviour. The response of organic liquid scintillators to fission fragments still need to be understood, because it is well known [7]

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that for light charged particles, the higher the mass the higher the slow fluorescence proportion. However for fission fragment this proportion is unexpectedly small, and this might be connected to the low light yield observed for fission fragment. References [1,5,6] agree that this yield is about a factor 80 lower for fission fragments compared to light yields for electrons. Hence contrarily to fission chambers which use a charge separation between α-particles and fission fragments, pulse shape discrimination is very important in order to separate fission events from electrons and mainly from proton recoils in fast neutron induced reaction studies.

In reference [1] it was shown that fission count losses are extremely small (about 10^{-4} %) and very small for α-decays (about 0.5 %). This feature allows very precise counts for both kinds of events. This unique capability is associated with the usual advantages of organic liquid scintillators: Pulse Shape Discrimination (PSD) and fast fluorescence that provides good time resolution. Finally the last but not the least advantage is that the liquid-liquid extraction, used in order to load the scintillator with an actinide, is a fast and convenient process (about 10 minutes).

We focus here on the use of this active target for spontaneous fission probability measurements relative to α-decay for the ^{240}Pu, ^{242}Pu and ^{252}Cf isotopes. They are motivated by the need for high precision measurements on neutron induced cross sections for the plutonium isotopes[12], since a good knowledge of spontaneous fission can be used to determine the fission detection efficiency. Californium 252 was foreseen as a test of this new method.

2 Experimental setup

2.1 Scintillator preparation

The ^{240}Pu and ^{242}Pu samples were provided and characterized by the JRC.G.2 laboratory at Geel, Belgium [8]. The original Pu powders were delivered in the form of dried nitrate solutions. In order to extract the isotopes into liquid scintillators the powders were first dissolved into 2 mL of HNO_3 1M solution. Then the plutonium was extracted into 1.2 mL of EJ309 scintillator, that contained 0.05 M of the extracting molecule HDEHP. For ^{252}Cf the mother solution (0.1M HNO_3) was purchased from the Eckert-Ziegler company. Contrarily to the plutonium samples, ^{252}Cf was extracted into a DIN based scintillator. Table 1 summarizes the main characteristics of the 3 prepared scintillator solutions. The isotope half-lives are taken from the NNDC database [9].

Table 1 Summary of the final test tube characteristics.

Isotope	Scintillator	Activity (Bq)	Fission rate (fission.s^{-1})	Counting duration (days)	Half-life (days)
^{240}Pu	EJ309	70×10^{3}	4×10^{-3}	60	6561(7)
^{242}Pu	EJ309	1.2×10^{3}	0.06	3	$3.73(3)\times10^{5}$
^{252}Cf	20	7.3	0.23	73.5	2.645(8)

2.2 Countings

After the liquid-liquid extraction the scintillator was sealed inside a test tube with an epoxy glue and a silicon plug. The test tube was placed vertically into a tube holder. The inside

part of this holder has a spherical shape and was painted in order to reflect the scintillating light towards a Hamamatsu R6231-100 PMT. The PMT was optically coupled thanks to an optical grade silicon grease, to a quartz window glued to the holder. The space between the test tube, the window and the light reflector was filled with an optical grade silicon oil, in order to optimize the optical coupling between the scintillator and the PMT. The anode signal was digitized with a FASTER data acquisition system, developed by the LPC Caen laboratory [10]. It is based on a 12 bit 500 MS/s flash ADC, and a numerical module implemented into a FPGA. The numerical module is a QDC program that allows signal filtering, base line restoration, constant fraction discrimination and charge integration. Up to 4 time windows can be defined for the charge integration. Finally charges, counters, and time stamps are sent to the acquisition computer through a 1 GigaByte/s network link. The counting time for each isotope is given in Table 1. The threshold was always kept low enough in order to store all decay events, and especially all the α-decay events. Even if this is not mandatory from a statistical point of view, tests were performed with higher thresholds and triggering rate fluctuations beyond the targeted accuracies were observed. Hence for the ^{240}Pu sample, more than 12 To of data were accumulated, but this guarantee that we could compute the fission/α-decay rates ratio without bias related to threshold instability. For each event an identification ratio is defined as the ratio of the slow over the fast charges. Fig. 1 presents a 2-dimension plot obtained by histogramming the total charge on the x-axis, and the identification ratio on the y-axis.

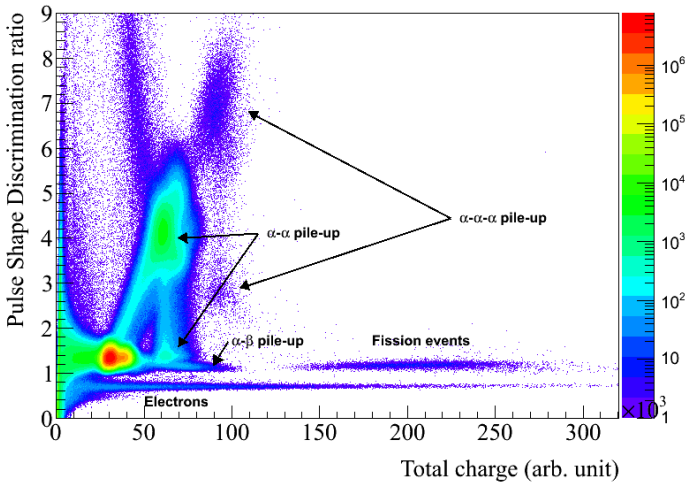

Fig. 1 Identification histogram for the ^{242}Pu sample.

In this 2-dimensional histogram, one can identify a secondary electrons line at the lowest ratios, an α-particle decay peak with high counting rates, fission events at high charges with ratio slightly smaller compared to α-particles and finally different kind of piled events.

2.3 Analysis

As mentioned previously the detection efficiencies for fission events are very close to 100 %. Hence the number of spontaneous events is obtained easily by counting the number of fission events into a graphical cut placed around the observed events. The determination of the number of α-particle is complicated by the presence of piled events (double and triple for ^{242}Pu). This is even more complicated for ^{240}Pu because its specific activity is 56.85 times higher compared to ^{242}Pu. Fig. 2 shows the identification histogram obtained for the

^{240}Pu sample. There we can observe events with up to 6 α-particles piled-up. These events can be easily identified by inspecting the total charge of the signal. For twice the α-decay energy, we observe 2 peaks. The first one with an identification ratio identical to α-particles corresponds to 2 piled α-particles which are emitted within a very small time interval. The peak for higher ratios corresponds to a second α-particle that piles during the beginning of the slow decay of the first event. Intermediate situations corresponding to intermediate time intervals, or higher time intervals are observed with intermediate ratio or/and total charges. For higher pile order (≥3) one can perform the same identification. For small time intervals the probability is smaller and smaller, and more combinations appear. For example for 3 piled α-particles, one can observe 2 decays within a short time, and a third one later, or a first decay with 2 decays that pile on the slow component. In total 3 peaks associated to these 3 combinations appear.

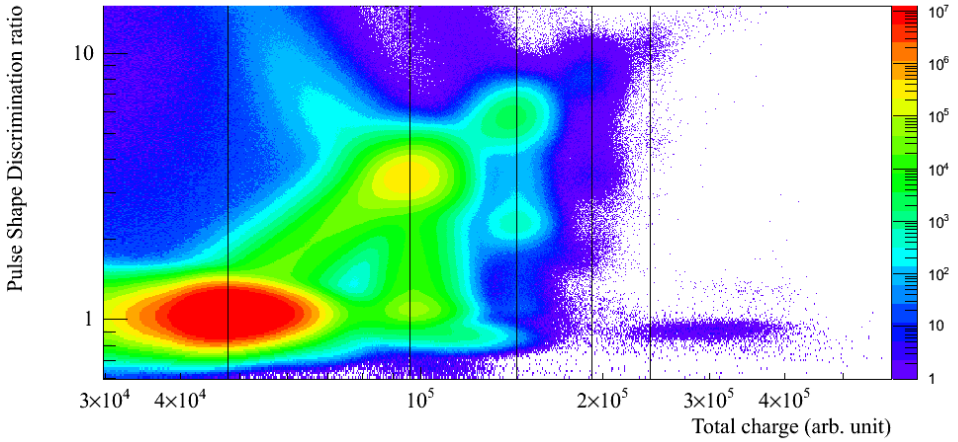

Fig. 2 Identification histogram for the ^{240}Pu sample. The vertical lines locate multiple integers of the α-decay energy.

Once all kind of events are identified, graphical cuts can be defined in order to count the number N_i of events with i piled α-particles. This allows to compute the total number of α-decays through:

$$N_\alpha = \sum_i i\, N_i \qquad i \geq 1 \tag{1}$$

Prior to this summation N_1 is corrected for α-particles losses. In order to determine the α-particle detection efficiency, the α spectra is computed by projecting on the x-axis the previous histogram for all single α-particle events including the tail under the α peak. This tail is fitted with a linear function in order to extrapolate the tail to the lowest charges where the electron-α separation is no more possible.

Finally in order to compute the probability of spontaneous fission relative to the α-decay, one has to correct for sample impurities. This is performed from the isotopic contents of the sample given at a reference date, and by calculating the decay rates for α-decays and spontaneous fissions for every isotope at the measurement date. For long measurements the cooling calculation is performed for each run. The run duration never exceeds 7 days, hence it is completely justified to do this correction for an entire run. The final result is the ratio:

$$r = \frac{N_\alpha^{iso}}{N_f^{iso}}$$

where the upper-script iso refers to the measured isotope, obtained after the isotopic content correction. This ratio equals the ratio of the spontaneous fission half-life, over the α-decay half-life:

$$r = \frac{T_{SF}}{T_\alpha} \tag{2}$$

3 Results and discussion

The relation between the isotope half-life $T_{1/2}$, and half-lives T_α for α-decay and T_{SF} for spontaneous fission is:

$$\frac{1}{T_{1/2}} = \frac{1}{T_\alpha} + \frac{1}{T_{SF}} \tag{3}$$

One can easily deduce from relation (2) and (3) that the spontaneous fission half-life is given by:

$$T_{SF} = (1 + r)\ T_{1/2} \tag{4}$$

Table 2 gives the results obtained in this work (second and third column). For comparison we also give the last evaluated values performed by N.E. Holden [11], and the most precise experimental value (reference [12] for the ^{240,242}Pu isotopes and reference [13] for the ^{252}Cf isotope). The values listed in the second column do not include the isotope half-life uncertainty, while in the third column it is included. The spontaneous fission half-lives were obtained by using the isotope half-lives given in Table 1. It can be concluded that for ^{242}Pu and ^{252}Cf the uncertainty on the isotope half-life is the limiting term in the spontaneous fission half-life, but that the relative fission probability is much more precise, and especially for ^{252}Cf.

Table 2. Spontaneous fission half-lives.

Isotope	This work		Holden	Experimental ref
^{240}Pu	1.132(8)×10^{11}	1.132(8)×10^{11}	1.140(10)×10^{11}	1.165(13)×10^{11}
^{242}Pu	6.77(5)×10^{10}	6.77(7)×10^{10}	6.77(6)×10^{10}	6.74(9)×10^{10}
^{252}Cf	85.245(75)	85.24(27)	86(1)	85.54(22)

In order to discuss the method with respect to the obtained uncertainties on the fission probability, we give the uncertainty budget for the measurement on the ^{240}Pu sample, which again is the most complicated in this study.

Table 3 Uncertainty budget for the ^{240}Pu sample

α detection efficiency	0.01 %
α count	0.04 %
α-α pile-up	0.04 %
Activity content	0.12 %
Fission statistic	0.67 %
Total	0.68 %

The α-detection efficiency has been determined by calculating the difference of efficiencies obtained for an energy-linear dependence of the α tail and a constant α tail. The 2 next

terms were obtained by counting the number of unpiled α-decay and the number of doubly α piled events by varying the graphical cuts in a reasonable range. The activity content uncertainty was obtained by propagating the uncertainties of the isotopic content at the reference date, and the half-life uncertainties. Due to the small cooling times in this experiment, these last one were negligible. The main uncertainty is due to the fission statistics. Nevertheless it is not an absolute limitation since we could have count the sample for a longer time. If so the next most limiting uncertainty is coming from the activity sample knowledge at the time of the measurement that limits here the precision to 0.12 %. Hence one can conclude that the method that has been used in this measurement can be very precise, and that the method itself gives very small uncertainties on these half-lives. The results obtained here are already a factor of about 2 better than the best values obtained for the plutonium samples.

4 Conclusion

From a general point of view we have demonstrated that with an active scintillating target very precise counts can be obtained for fission events and α-decay events, even for high count rates since the PSD technique can be used in order to correct for count losses due to pile-up. This is clearly very interesting for studies of spontaneous fission, but such active targets can be of great interest for neutron induced fission if one thinks of using it as a small neutron detector. Thus it can be used to measure the incident neutron flux, providing very precise fission cross sections measurements.

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