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## Preparation of drop deposited sources in $4\pi$ absorbers for total decay energy spectrometry

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**Abstract** Total decay energy spectrometry ( $Q$  spectrometry) with cryogenic detectors is a promising technique for analysis of  $\alpha$ -emitting actinides. However, this technique is very sensitive to the quality of the source preparation due to the absorption of the nuclear recoil energy in the source material. Drop deposition of the radioactive solution without care produces spectra with unpredictable peak shapes. In order to keep the simplicity of the drop deposition technique, we study the deposition of solution on latex nanoparticles or in nanoporous gold (NPAu) to improve the source quality.  $4\pi$  absorbers were fabricated and tested using these two techniques. The absorbers contained a mixture of Pu isotopes. From the best  $Q$  spectrum, the composition of Pu isotopes and other actinides was measured and it is in very good agreement with the reference values given by alpha and mass spectrometry.

**Keywords** Total decay energy spectrometry •  $Q$  spectrometry • Metallic magnetic calorimeter • Source preparation

### 1 Introduction

The spectrometry of the total decay energy (or  $Q$  spectrometry) of  $\alpha$ -emitting nuclides consists in measuring the  $Q$  values of the radionuclides enclosed in a  $4\pi$  absorber. The absorber is sized to absorb all the particles emitted during the radioactive decay, so the summed energy of the particles is equal to the  $Q$ -value. Compared with the complex spectra from  $\alpha$  spectrometry, the corresponding  $Q$  spectrum becomes very simple. For a

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mixed radionuclide spectrum, only one peak appears per radionuclide and these are generally spaced by at least a few tens of keV. Cryogenic detectors are suitable for  $Q$  spectrometry because they are able to provide very high-energy resolution and it is convenient to enclose sources in their absorbers for a high detection efficiency [1].

Usually, the source is deposited on the surface of one half of the metal absorber, and enclosed by diffusion welding with the other half. The nature of the source deposit in the absorber can broaden the full width at the half-maximum (FWHM) energy resolution. Indeed, absorbers with electro-precipitated or self-deposited sources have demonstrated FWHM resolutions of about 1 keV at about a few MeV, whereas drop and dried radioactive solutions in the absorber produce spectra with deformed and split  $Q$ -peaks with much larger FWHM [1-4]. Numerical simulations show that the recoil nuclei are absorbed in a few tens of nm of material [3] and considering that the dried radioactive drops on gold form a  $\mu\text{m}$ -scale deposit, it is clear that the recoil energy is mainly absorbed in the deposit. This energy is therefore only partially released as heat to the metal absorber, and part of it is not even converted to heat but rather expended in the creation of defects like dislocations; this may explain the distorted  $Q$  peak [3].

Despite these difficulties, the use of drop deposition has some advantages: it is a very simple procedure to prepare sources, the deposited activity is traceable through the weight, and unlike electro-precipitation there is no dependence of the deposition yield on the element. Thus, to keep the drop deposition attractive meaning to have a high-energy resolution, we investigated two techniques of source deposition in the nm scale to minimize the nuclei energy absorption in the deposit. The techniques consist in dropping and drying the radioactive solution, for the first one on a latex pad, and for the second one on a nanoporous gold layer. The mother solution used to prepare the sources is a 3N nitric acid ( $\text{HNO}_3$ ) with 4 kBq/g of  $^{239}\text{Pu}$ . For each technique, the obtained source was embedded between two gold foils to form the absorber, which was then glued on a metallic magnetic sensor with Stycast 1266. The metallic magnetic calorimeters (MMCs) were cooled down to about 12 mK to measure the total decay energy spectrum. The pulse shapes and the  $Q$  peaks were analyzed to characterize the source quality.

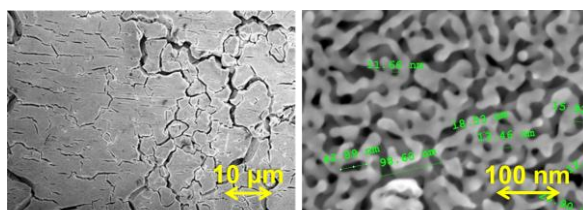
## **2 Gold surface preparations for drop deposition**

The first surface preparation is commonly used in metrology of ionizing radiation for preparing sources by drop deposition of radioactive solution [5]. It consists in electro-spraying an ethanol solution containing latex particles of  $\sim 70$  nm diameter onto a metallic surface using a potential of a

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few thousand volts. In our case, the metallic surface at the ground potential is a gold foil with a thickness of 25  $\mu\text{m}$  and a diameter of 12 mm. Therefore, a hydrophilic latex pad covers the gold area. The drops of radioactive solution uniformly wet the latex pad, and after drying the activity is homogeneously distributed over the area. It avoids the production of aggregates and coarse grains of salt. A 28  $\mu\text{L}$  drop of the radioactive solution was deposited with a micropipette on the latex pad. To make the  $4\pi$  absorber, a square of 1  $\text{mm}^2$  of the gold disk was cut and enclosed between two 25  $\mu\text{m}$  thick gold foils by diffusion welding. The corresponding activity is about 1 Bq.

The other studied method consists in dropping and drying the solution in nanoporous gold (NPAu) [6]. We expect that the solution fills the NPAu cavities and the solution will dry on very large surface, minimizing the deposit thickness once the source is dried.



**Fig. 1** SEM images with two magnifications of the fabricated NPAu used in the  $4\pi$  absorbers. (Color figure online).

The NPAu is fabricated by dealloying  $\text{Ag}_{81}\text{Au}_{19}$  (at%) in concentrated  $\text{HNO}_3$ . A AgAu foil with a thickness varying between 8 and 25  $\mu\text{m}$  is laminated from a AgAu nugget, and the foil is diffusion welded onto the surface of half of the gold absorber in order to strengthen the NPAu since NPAu is very fragile. The sample is then dipped into  $\text{HNO}_3$  until the silver is completely etched and dissolved. The remaining gold forms a nanoporous structure. The size of the pores in the gold depends on the etching time, the temperature, the concentration of  $\text{HNO}_3$  and the stoichiometry of the AuAg alloy [7]. Using concentrated (68%)  $\text{HNO}_3$ , by varying the time and the temperature, we found that an etching time of 3 h at 20°C provides pores of about a few tens of nm while the silver is removed. The composition of the nanoporous gold was checked by energy dispersive X-ray spectrometry (EDX), and the size of the pores was obtained from SEM images (Fig. 1). Some deposition tests with drops of non-active 3N  $\text{HNO}_3$  have shown that the NPAu acts as a foam: it absorbs the solution while allowing it to evaporate easily, 0.3  $\mu\text{L}$  of 3N  $\text{HNO}_3$  evaporates in 10 min on NPAu of few

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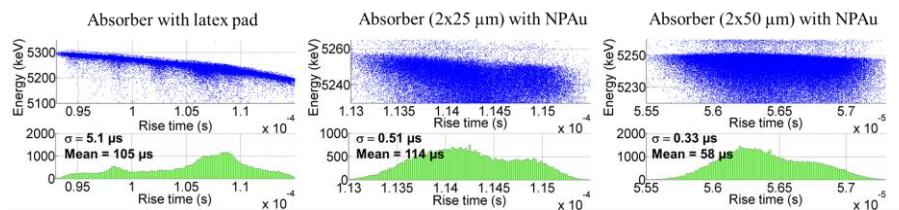
mm<sup>2</sup>. About 0.3 μL of the radioactive solution was deposited on the NPAu, corresponding to 1.2 Bq. Two absorbers were fabricated by diffusion welding, one with NPAu enclosed between two 25 μm thick gold foils and the other one between 50 μm gold foils.

The three absorbers (i.e. source on latex pad, source on NPAu between 2x25 μm of gold, source on NPAu between 2x50 μm of gold) were glued on the magnetic sensors of MMCs and cooled down to about 12 mK in a wet dilution cryostat. The three absorbers were characterized by analyzing the pulse shapes and the  $Q$  spectra.

### 3 $Q$ spectra and results

#### 3.1 Absorber with source on latex pad

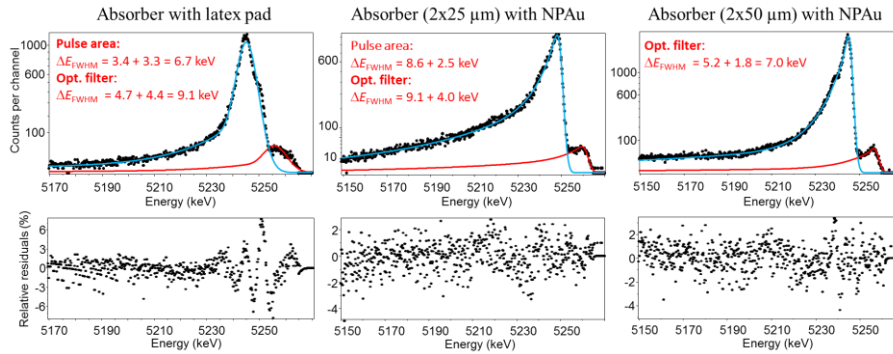
The pulses recorded with the source on latex pad exhibit a distribution of rise times (Fig. 2) with a large standard deviation of 5.1 μs. This may be due to the fact that the latex buffer remaining in the absorber has poor thermal properties at low temperatures. Moreover, the pulse heights clearly depend on these rise times (Fig. 2). After a correction of this dependence by a quadratic fit of the pulse height as a function of the rise time, the energy resolution is improved. The spectrum after optimal filtering shows a worse FWHM energy resolution (9.1 keV) than the spectrum from the pulse areas (6.7 keV) because the condition of having identical pulse shapes for optimal filter is not respected [8]. The obtained  $Q$  spectrum determined by the pulse areas is shown in Fig. 3. The  $Q$  peak of <sup>239</sup>Pu has a relatively small left tail and is nearly symmetric: the left width at half maximum (LWHM) is 3.4 keV, close to the right width at half maximum (RWHM) of 3.3 keV. The relative residuals in Fig. 3 show that the  $Q$  peak shape deviates from a Bortels function (a Gaussian with two left tails), disabling a quantitative isotopic analysis from this spectrum [9, 10].



**Fig. 2** Top row, pulse energy as a function of rise time (10% - 90%). Left and middle: the energies are obtained using the pulse area, right: they are

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obtained after an optimal filter. *Bottom row*, distribution of the rise times. The standard deviations and the mean value of the distributions are given. (Color figure online)



**Fig. 3**  $Q$  peaks of  $^{239}\text{Pu}$  (blue line) and  $^{240}\text{Pu}$  (red line) for the three absorbers fitted with Bortels functions. Left and middle: the spectra are obtained using the pulse area. Right: it is obtained after an optimal filter. The  $\Delta E_{\text{FWHM}}$  is the sum of the LWHL (left term) and the RWHL (right term). The relative residuals between the spectrum and the fit are shown. (Color figure online)

### 3.2 Absorbers with source in NPAu

The pulses from the  $2 \times 25 \mu\text{m}$  thick absorber also exhibit a slight dependence of the pulse height on the rise time (Fig. 2), but the rise time distribution has a smaller dispersion ( $\sigma = 0.51 \mu\text{s}$ ) than the absorber with latex pad. Again, in this case, a better energy resolution is obtained using the pulse areas to determine the pulse energies than with optimal filtering, indicating that the pulse shape is still affecting the resolution. The  $Q$  peaks of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are shown in Fig. 3 and are properly fitted with Bortels functions with two tails. The Bortels parameters are imposed to be identical between the  $Q$  peaks, excepted for the peak position and the peak height. The amount of each isotope corresponds to the integrated area of the Bortels function.

The peaks are strongly asymmetric with a wide left tail, the LWHL is 3.4 times larger than the RWHL. However, the RWHL is smaller compared with the absorber having the latex pad, enabling to distinguish the two Pu isotopes, but not to quantify them precisely. In order to understand the origin of the tail and the rise time distribution, the absorber was characterized with

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an external alpha source. An alpha particle deposits its energy in the gold bulk part of the absorber while the  $Q$  pulses correspond to an energy deposition into the NPAu. We observed the same rise time distributions and the same peak shapes for  $\alpha$  and  $Q$  pulses. A possible explanation would be a lack of thermal conductivity of the entire absorber; this is why for the third absorber, the NPAu was enclosed in twice-thicker gold foils to increase the thermal conductivity.

Fig. 2 shows that the pulse height dependence on rise time is nearly suppressed. Compared to the previous NPAu absorber, there are several improvements:

- 1- the dispersion of the rise time is smaller ( $\sigma = 0.33 \mu\text{s}$  against  $0.51 \mu\text{s}$ ), but still larger than for an embedded self-deposited  $^{210}\text{Po}$  source ( $0.15 \mu\text{s}$ ) [1],
- 2- the best spectrum was obtained with optimal filter,
- 3- the left tail is reduced, the LWHM is 5.2 keV against 8.6 keV previously,
- 4- the RWHM is 1.8 keV, improved by the use of an optimal filter. The narrower RWHM enables to separate and quantify the  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  using Bortels functions.

A shoulder is visible at 5239 keV, it is not related to the presence of any other radionuclide, but probably rather an artefact from the recoil energy absorption in the deposit as it was observed in [4]. This shoulder has a small effect on the residuals and on the quantification of the isotopes.

Table 1 shows the isotopic ratio and the actinide composition obtained from this measurement; they are compared to the reference values. The latter ones are measured by mass spectrometry ( $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ) and by alpha spectrometry ( $^{241}\text{Am}$ ). The values are in very good agreement except for  $^{241}\text{Am}$ . The measurement by  $Q$  spectrometry reveals that the  $^{241}\text{Am}$  detected by alpha spectrometry clearly contains a contribution from  $^{238}\text{Pu}$ . In fact,  $^{241}\text{Am}$  and  $^{238}\text{Pu}$  were not distinguishable by alpha spectrometry.

**Table 1** Actinide composition of the reference values compared with those obtained by  $Q$  spectrometry. Only the statistical uncertainty is taken into account in the measured values by  $Q$  spectrometry.

X	Reference values		Measured values	
	$X/(^{239}\text{Pu} + ^{240}\text{Pu})$ (Bq/Bq)	Rel. unc. (%)	$X/(^{239}\text{Pu} + ^{240}\text{Pu})$ (Bq/Bq)	Rel. unc. (%)
$^{239}\text{Pu}$	0.9683 (40)	0.42	0.9688 (38)	0.39
$^{240}\text{Pu}$	0.03166 (42)	1.3	0.03117 (50)	1.6

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<sup>241</sup> Am	0.00793 (24)	3.1	0.00445 (19)	4.2
<sup>238</sup> Pu	-	-	0.00479 (19)	4.0

This latter measurement demonstrates the capabilities of the technique of using drop deposition in NPAu. The peak shape is well fitted by a Bortels function and it is possible to determine precisely the composition of actinides in a radioactive material without radiochemical separation. Nevertheless, some anomaly must be understood for future improvements to decrease the detection limits and improve the quantification of actinides: the FWHM energy resolution is larger than the FWHM baseline resolution (about 0.6 keV for the presented measurements) and larger than measurements with electro-precipitated sources. Moreover, the left tails are still large, indicating that the recoil nuclei lose a large fraction of their energy in the source material.

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