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▶ To cite this version:

B. Laurent, J. Taieb, G. Bélier, Paola Marini, P. Morfouace. New developments of a fission chamber for very high radioactivity samples. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2021, 990, pp.164966. 10.1016/j.nima.2020.164966 . hal-03368961

HAL Id: hal-03368961 https://hal.science/hal-03368961

Submitted on 7 Oct 2021

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New developments of a fission chamber for very high radioactivity samples

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6 Abstract

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A new fission chamber was developed and built for prompt-fission-neutron spectra (PFNS) measurements in neutron-induced fission of actinides, and is described in J. Taieb *et al.*, Nucl. Instr. and Meth. A 833 (2016) 1-7. It allowed us to minimize incident- and outgoing-neutron scattering, and optimize both time resolution and alpha to fission fragment discrimination. The fission chamber was validated and used for 238 U and 252 Cf PFNS measurements. To measure PFNS of samples with a natural alpha activity of the order of 10 MBq, further improvements to the detector were done and are presented in this work. Tests of the improved fission chamber lead to a fission fragment detection efficiency of 91% for a 14.41 MBq 240 Pu deposit, and 99% for a 247 kBq 242 Pu deposit. The fission chamber was successfully used for 239 Pu PFNS measurements at the Weapons Neutron Research facility of the Los Alamos Neutron Science Center (WNR@LANSCE).

7 Keywords: Fission chamber, high activity samples, efficiency, segmented

s anode

• 1. Introduction

In recent publications [1, 2], a new type of light-weighted fission chamber developed for Prompt-Fission-Neutron Spectra (PFNS) measurements was presented. Actinide mass, time resolution and the amount of structural material were optimized to improve both alpha-fission fragment discrimination and timeof-flight measurements resolution, as well as to reduce the amount of neutrons

November 30, 2020

Preparint explored to the Elsevier

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scattered from the detector. These chambers were successfully used in PFNS
experiments at different facilities: LICORNE at Institut de Physique Nucléaire
Orsay (IPNO), Weapon Nuclear Research (WNR) at Los Alamos, 4MV Van
de Graaff at Bruyères-le-Chatel. Some results on ²³⁸U(n,f) reactions were published [2, 3].

The next step was to use the fission chamber in experiments using high 20 alpha-activity actinides such as ²³⁹Pu. While the main principle of the detector 21 remains the same (gas type and pressure, gap between anode and cathode), 22 improvements are needed to deal with the high activity of the sample in terms 23 of radioprotection on one hand, and to avoid alpha-signals pile-up, which de-24 grades the alpha-fission fragment discrimination, on the other. By improving 25 this discrimination, cuts on angular and energy distributions of detected fis-26 sion fragment are reduced and the induced bias on extracted PFNS and others 27 observables (angular distribution, mean neutron energy, number of neutrons 28 emitted per fission...) become negligible. In the following, details of the de-20 tector developed for ²³⁹Pu, but well suited for any other high alpha-activity 30 sample, will be presented. 31

³² 2. New design of the chamber

33 2.1. Mechanical aspects

A schematic view of the fission chamber is shown in Fig. 1. As for the 34 chamber described in [1], the material budget was minimized, to ensure low 35 neutron-scattering rate, thus reducing the neutron background generated dur-36 ing the experiment. Following the same idea, all used materials are almost 37 hydrogen-free. However, because of the high activity of the ²³⁹Pu deposits, han-38 dling and assembling the targets in the chamber had to be performed in gloves 39 box. Therefore, the external case of the ²³⁹Pu detector had to be redesigned to 40 take into account this specificity. Moreover, the dimensions of the detector were 41 increased so that structural material does not intercept the neutron flight path. 42

When in use, the fission chamber is filled with flowing tetrafluoromethane (CF₄) gas at a fixed absolute pressure typically 100 mbar above atmospheric pressure.

The external structure is a 1 mm-thick aluminium parallelepiped soldered on a 4 mm-thick aluminium flange. The structure supports glued titanium foils on each side: 100 μ m thick on rectangular sides, 50 μ m thick on entrance and exit sides. Titanium foils are glued from the inner side, to stand the gas pressure inside the chamber. The soldered flange is screwed on the flange glued to the PCB. An o-ring ensures the gas tightness between the cover and the PCB.



Figure 1: Scheme of the assembled fission chamber (titanium foils are not showned).

All the electronic connections and mechanical support were placed on the 52 bottom side of the chamber. This side consists in a printed circuit board (PCB) 53 supporting all front-end electronics components. The anodes are connected on 54 the internal side, while pre-amplifiers are connected on the external one. The 55 PCB also integrates high voltage connections for the drift field between anodes 56 and cathodes, low voltage connections for pre-amplifiers and a readout connector 57 for each anode. A 3 mm-thick aluminium flange is glued on the external face 58 of the PCB to fix the cover. Aluminium structures fixed on the internal face of the PCB supports three rods made of Torlon[®] [4] to position the electrodes. 60

61 2.2. The cathodes

Twenty-two ²³⁹Pu samples were prepared by the JRC-Geel target laboratory. 62 The sample thickness is a compromise between the amount of matter (and thus 63 the fission rate) and the energy loss of the emitted fission fragments in the 64 deposit. The lower the loss, the better the alpha-fission fragment discrimination. 65 The Plutonium material was electroplated on a 25 μ m-thick aluminium back-66 ing (see Fig. 2), glued on the cathode frame. The diameter of the deposit is 67 33 mm with a thickness of 250 $\mu g/cm^2$. Each deposit contains $\approx 2.1 \text{ mg} (5 \text{ MBq})$ 68 of ²³⁹Pu for a total of 47 mg housed in the chamber. The ground connexion of 69 the cathode to the chamber is ensure by a glued cable. 70



Figure 2: 239 Pu sample electroplated on the cathode frame, with glued cable for the ground connexion in the detector.

71 2.3. Fission chamber assembly

The mounting of the Pu deposits inside the fission chamber was performed at JRC-Geel in a dedicated gloves box. The PCB with internal supports was positioned vertically and then cathodes, spacers and anodes were threaded on the insulating rods (see picture 3(a)). Each anode collects the signal from two deposits (cathodes): one on each side. So there are 11 electronics channels corresponding to the 22 deposits.

Once the 22 cathodes and the 11 anodes are installed, the chamber is closed
by the cover (picture 3(b), right), and it can be removed from the gloves box
after contamination clean-up.



Figure 3: Pictures of the fission chamber assembly in a gloves box. The stack of anodes and cathodes (with 239 Pu) is mounted (a) before closing the detector with the cover (b).

⁸¹ 3. Dedicated segmented anodes

82 3.1. Anode principle

One anode of the fission chamber deals with 10 MBq alpha activity com-83 pared to a standard fission rate during experiments of around 15 f/s. In order to 84 improve alpha-fission fragment discrimination, a dedicated anode was designed. 85 The goal is to reduce the impact of alpha pile-up on the detection of fission 86 fragments and enhance their discrimination. As the alpha-fission fragment dis-87 crimination is based on the different charge-signal amplitudes induced by alpha 88 particles and fission fragments, the worst scenario occurs when an alpha is emit-89 ted parallel to the cathode. Indeed in this case its deposited charge is close to 90 the one deposited by a fission fragment emitted perpendicularly to the cath-91 ode. The alpha-particle track length can be longer than the radius of the anode 92 (37 mm), while the track length of the fission fragment will be only the gap 93 between the anode and the cathode (2.5 mm), as shown in Fig. 4(a). The anode 94 was therefore segmented in two concentric regions and the collected charge was 95 read only from the central zone. This allowed us to reduce the amount of alpha 96 signals with amplitudes comparable with the ones from fission fragments. The 97 external part of the anode is isolated from the central part but kept at the same 98



electrical potential, so not to distort the electric field in the collection region.

Figure 4: Principle of the segmented anode. The red zone corresponds to the charge collection zone. In case of a full anode, alphas and alphas pile-up can have greater amplitudes than fission fragments (FF) ones (a). With the segmented anode, this amplitude is reduced (b).

Moreover, a dedicated modern front-end electronics with very short shaping times (≈ 10 ns) also reduces the signals pile-up and improve the time resolution of the setup.

This peculiar anode design, suitable for high-activity samples fission chamber, have been patented [5].

¹⁰⁵ 3.2. Validation tests with high alpha-activity ^{240,242}Pu deposits

This new anode concept was tested with high alpha-activity deposits of ^{240,242}Pu. These two isotopes are characterized by a very high alpha-activity compared to their spontaneous fission decay rate. They are then good candidates to test offline the limits of alpha-fission fragment discrimination. A



Figure 5: Segmented anode scheme.

14.41 MBq- α ²⁴⁰Pu and 0.247 MBq- α ²⁴²Pu deposits were mounted alternatively in the chamber, equipped with plain and then segmented anodes to quantify the improvements. The other characteristics of the chamber were kept constant (gas pressure, gap between anode and cathode, electric field, electronics, DAQ system). Fig. 6 and 7 show the pulse-height spectra collected for these deposits when using a plain anode.



Figure 6: Pulse height spectrum of the ²⁴⁰ Pu sample (14.41 MBq), with a fission event detection efficiency of 66%. The red dotted line show the threshold used to determine the number of detected fission fragments.

Because of the deposits thickness (0.24 mg/cm^2) , some of the fission fragments are stopped in the deposit. Monte-Carlo simulations [6] showed that



Figure 7: Pulse height spectrum of the 242 Pu sample (0.247 MBq), with a fission event detection efficiency of 97%. The red dotted line show the threshold used to determine the number of detected fission fragments.

fission fragments have an efficiency of 94% to exit the deposits with enough energy to be detected in the gas afterwards. This loss has to be taken into account in the fission detection efficiency calculation.

To improve the alpha-fission fragment discrimination, the time signal of the 121 anode was integrated over two gates and a pulse-shape discrimination (PSD) 122 technique was used. The first gate is centered on the rising part of the sig-123 nal, while the second one is centered on the maximum amplitude. By plotting 124 the correlation of these two charge signals (Fig. 8), the alpha-fission fragment 125 separation can be improved from 66% to 91% for the 14.4 MBg ²⁴⁰Pu sample. 126 Table 1 summarizes the measured efficiencies for the ^{240,242}Pu samples, in 127 different configurations: plain anode, segmented anode, segmented anode and 128 PSD analysis. These results show that using the segmented anode and a PSD 129 analysis, an efficiency as high as 91% can be obtained. For the lower activity 130 242 Pu sample (0.247 MBq), the standard fission chamber provides a very good 131 alpha-fission fragment discrimination and the improvements of the detector has 132 less impact. 13

These tests validate the new concept of anode developed for high alphaactivity actinides used in the fission chamber and new experimental measurements of ²³⁹Pu(n,f) PFNS were performed at WNR@LANSC (see section 5).

Actinide	240 Pu	242 Pu
Alpha activity (MBq)	14.41	0.247
SF branching ratio	5.7E-8	$5.5 ext{E-6}$
SF rate (Hz)	0.82	1.36
Fragment emission efficiency	0.94	0.94
Expected fission rate (Hz)	0.77	1.28
Plain anode		
Measured fission rate (Hz)	0.51	1.24
FF efficiency (%)	66	97
Segmented anode		
Measured fission rate (Hz)	0.60	1.25
FF efficiency (%)	78	98
${\bf Segmented \ anode + PSD}$		
Measured fission rate (Hz)	0.70	1.27
FF efficiency (%)	91	99

Table 1: Informations concerning $^{240,242}Pu$ tests, with the values of expected and measured fission rates for each isotope. SF stands for Spontaneous Fission.



Figure 8: Example of a 2D histrogram showing the alpha-fission fragment discrimination for the 240 Pu sample. This histogram is obtained by plotting one charge integration versus the second one (see text for details).

137 4. Simulations of the neutrons scattering

The NPTool framework [7, 8] using the Geant4 simulation package was 138 used to quantify the neutron scattering generated by the structural material 139 of the fission chamber. In this work, we used the high precision transport model 140 G4NeutronHP using the default G4NDL nuclear data. During experiments, the 141 fission chamber is placed at the center of an array of 54 liquid-scintillator cells 142 (Chi-Nu cells) as it can be seen in Fig. 9. Both incoming and outgoing neutrons 143 may scatter with the air or with the detector materials, inducing neutron back-144 ground in the cells and a distortion of the neutron energy spectra. The detailed 14 5 geometry and material of the fission chamber was carefully included in the sim-146 ulation (anode, cathode, Ti walls, Al frame, PCB...) In order to quantify the 147 effect of the fission chamber, two different simulations have been performed: one 148 only with air and the second one with the fission chamber included (see Fig. 9). 149 Fig. 10 shows the fraction of incoming neutrons scattered either by the air or 150 by the fission chamber. The blue points correspond to the first simulation only 151 with air while the open red circles show the effect of the added fission chamber. 152 One can see that the fission chamber materials add a negligible background to 153 the prompt-fission neutrons. Moreover, these prompt neutrons of interest are 154 detected in coincidence with a fission, then all scattered neutrons outside the 155

- coincidence gate do not affect the measurement. As a consequence the fraction
- ¹⁵⁷ of scattered neutrons shown in Fig. 10 corresponds to a higher limit which is reduced by the coincidence gate.



Figure 9: GEANT4 geometry of the fission chamber used in the NPTool simulations. 54 cells surrounding the fission chamber are reproduced. The incoming neutron beam is represented by the blue trajectories.



Figure 10: Normalized incoming neutrons scattered on air (blue full circles) and air+fission chamber (red open circles). A maximum of 0.6% at very low energy of the incoming neutrons are scattered and detected in a Chi-Nu cell, and the effect of the fission chamber is negligible.

159 5. Experimental uses

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²³⁹Pu Prompt-Fission-Neutron Spectra (PFNS) were measured placing the
fission chamber, containing 47 mg of actinide in the WNR neutron beam to

induce fission of the Pu deposits. The detector was surrounded by the 54 liquid 162 scintillator cells of the Chi-Nu array [9]. The incoming-neutron energy is deter-163 mined by time-of-flight measurement between the beam pulsation and the event 164 detected in the chamber. Neutrons and gammas emitted by neutron-induced 165 fission reaction were detected and identified in the scintillator cells, and neu-166 trons energies measured via time-of-flight technique. Results of this experiment 167 were presented in [10]. As an example, Fig. 11 shows the pulse-height spectra of 168 239 Pu fission chamber with and without beam. The fission detection efficiency 169 was estimated to be close to 95% for 10 MBq alpha activity per channel, thanks 170 to some improvements on the design of the front-end electronics boards. 171

This good fission event identification probability, combined the very low neutron detection threshold ($\approx 200 \text{ keV}$) and the granularity of the Chi-Nu detector, allowing angular emission corrections, as well as the high recorded statistics, lead to very precise PFNS measurements. Moreover, thanks to these characteristics, the number of neutrons emitted per fission ($\overline{\nu}$) as a function of incoming energy was extracted from the measurement with an unprecedented accuracy [11].



Figure 11: Pulse height spectra of ²³⁹Pu fission chamber obtained with (red) and without (blue) beam (extracted from [10]).

179 6. Conclusions

The fission chamber developed for PFNS measurements was improved to be used with high alpha-activity samples. The housing was modified to allow the assembly of the detector in a gloves box. For fission detection, a new concept of segmented anode was developed. Thanks to the use of these segmented anodes and modern front-end electronics, a fission detection efficiency of 95% was reached for alpha activities as high as 10 MBq, to be compared to a fission rate of 15 f/s.

Thanks to its very high alpha-fission fragment discrimination capability, this fission chamber can be further used in fission cross-section as well as PFNS measurements of very active actinides, such as ^{238,240,241,242}Pu.

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 submitted to Phys. Rev. Lett. (Nov. 2020).