Alpha radioactivity measurement techniques

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Preample

Alpha radioactivity measurement

- Non-exhaustive presentation (far from it, sorry ☹)
- Only an overview of techniques
- Mainly based on:
  - LNHB Website : http://www.lnhb.fr ☺
  - Bergeron D., Kossert K., Collins S., Fenwick A. Realization and dissemination of activity standards for medically important alpha-emitting radionuclides, Applied Radiation and Isotopes 184, June 2022, 110161
  - Pommé S., Methods for primary standardization of activity, Metrologia 44 (2007) S17-S26
- In medical applications and studies, quantitative activity measurements to evaluate the accumulation of radioactivity in tissues are indispensable

LNHB-MA

National Metrology Institute for Activity Measurement

LNHB-MA, Bât 602, CEA Saclay

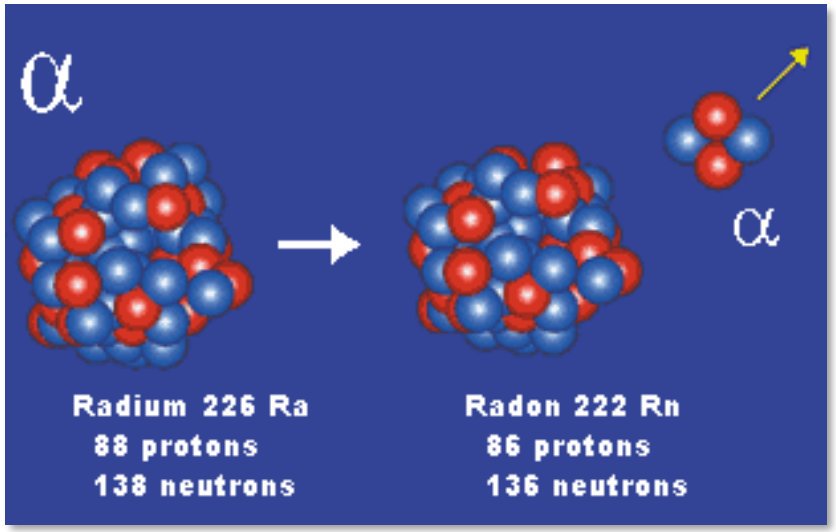
Defined Solid Angle Alpha

Radon-222 primary standard

Cryogenic detectors

Alpha radioactivity

$$^{A}_{Z}X \rightarrow {}^{A-4}_{Z-2}Y + {}^{4}_{2}He$$



- Radiation identified in 1908 by Sir Rutherford
  - Emission of an alpha particle (2 protons and 2 neutrons, He particle)
  - Atomic number Z > 84
  - When nuclei are large, they are unstable* and approach stability by expelling helium.

*Big nuclei are instable because of their overall excess of nucleons

- The energy released during alpha decay is in the form of kinetic energy shared between the alpha particle and the recoiling nucleus.
- Alpha particles carry about 98% of the energy and the recoil nucleus the rest. The energy of the alpha particle is unique for a given disintegration. It is higher than that of beta electrons and gamma rays, of the order of 4 million electron volts (MeV) or more.

Main measurement techniques

Liquid Scintillation

LSC-TDCR

LTAC-TDCR

4π β γ Measurements

4π α γ Measurements
Plastic Scintillator

Cryogenic detectors

Gridded Ionization Chamber

Defined Solid Angle

Liquid Scintillation

- Very old and very powerful technique (Kallmann, 1950 and Reynolds et al., 1950)
- First commercial detector in 1954 (Packard Instruments) and since then, many generations of counters which can be found around the world.
- Liquid scintillation devices mainly used for the activity measurements of aqueous samples in environmental monitoring, and more specifically for radionuclides decaying by pure beta transition or electronic capture, for which gamma-ray spectrometry methods are inoperative.
- Used for alpha-emitting radionuclides (Bergeron D. E. et al., ARI, Volume 184, June 2022, 110161) destructive and poor resolution to separate the various radionuclides

Tri-Carb counter, ca. 1957

Cassette P., Determination of the Wall Effect for Alpha Emitters. Ph. Cassette, Editor, LSC2010

Advances in LS Spectrometry: Proceedings of the 2010 International Conference on LS Spectrometry, Paris, France, 6-10 September 2010, Radiocarbon

Broda, P. Cassette and K. Kossert. Radionuclide metrology using liquid scintillation counting. Metrologia 44 (2007) S36-S52.

Cassette, P., 2002. Evaluation of the influence of wall effects on the liquid scintillation counting detection efficiency for the standardization of high-energy beta and alpha radionuclides. In: Mobius, S., Noakes, J.E., Schonhofer, F. (Eds.), LSC 2001, Advances in Liquid Scintillation Spectrometry. Tucson: Radiocarbon, pp. 45–55.

Fitzgerald, R., Forney, A.M., 2011. Determination of the Wall Effect for Alpha Emitters. Ph. Cassette, Editor, LSC2010. Advances in LS Spectrometry: Proceedings of the 2010 International Conference on LS Spectrometry, Paris, France, 6-10 September 2010, Radiocarbon. The University of Arizona, Tucson, Arizona, USA, ISBN 978-0- 9638314-7-7, pp. 331–339.

Liquid Scintillation

- Weighed aliquots of a radioactive solution are added to an LS vial that contains the liquid scintillator and – in some cases – additional ingredients to ensure sample stability and/or to vary counting efficiencies.
- The LS sources are then measured in special counters with two or three photomultiplier tubes (PMT) and coincidences, *i.e.*, events which are detected at the same time in at least two PMTs, are counted.
- Since alpha particles that are emitted as a consequence of an alpha decay have high energies of a few MeV, a considerable number of scintillation photons is created and, as a consequence, it becomes possible to achieve counting efficiencies of virtually 100%, which was confirmed by several experimental studies (see, e.g., Cassette, 2002; Kossert et al., 2009; Fitzgerald and Forney, 2011).

LS vial

Wallac counter, ca. 2016

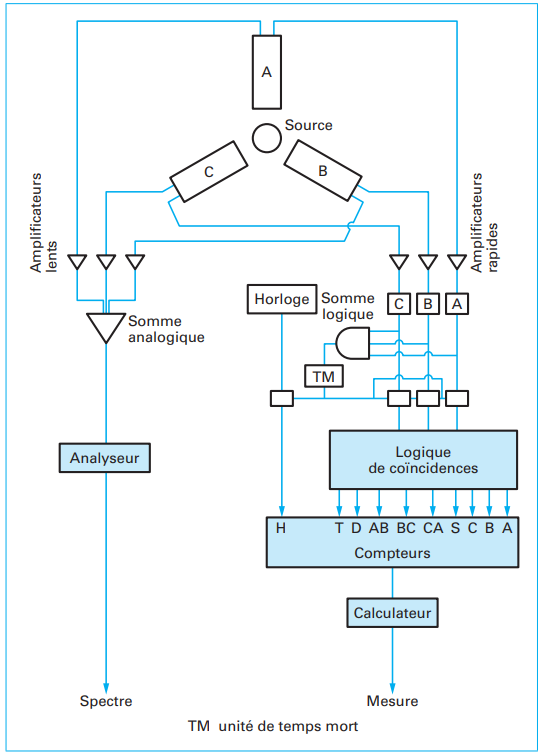
Ramebäck H., Alpha spectrometry and liquid scintillation counting for the measurement of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and age, 2020, https://doi.org/10.1016/j.apradiso.2020.109293

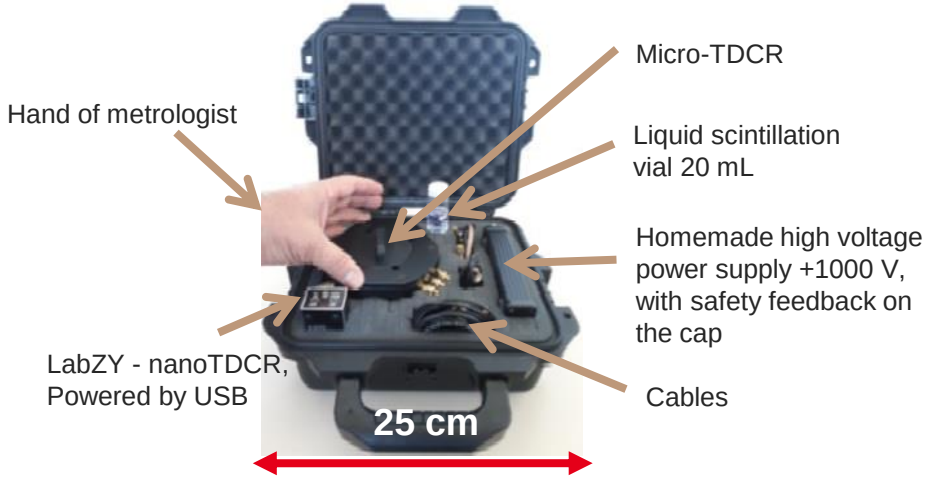
High-precision measurement of the half-life of 147Sm, Kossert K. et *al*., Applied Radiation and Isotopes, Volume 67, Issue 9, September 2009, Pages 1702-1706

Liquid Scintillation

Triple to Double Coincidence Ratio (TDCR)

- LNHB develops methods for the primary measurement of activity by liquid scintillation, enabling the production of national standards.
- Specific counters with three photomultipliers and its principle is based on the free parameter model: the ratio of the number of triple coincidences (three simultaneous photomultipliers) and double coincidences (two simultaneous photomultipliers) makes it possible to find the intrinsic light yield of the scintillator, proportional to the average number of light photons emitted by absorption in the scintillator.
  - The knowledge of this free parameter allows, in fine, to calculate the detection efficiency.
  - This method is known in the literature as the Triple to Double Coincidence Ratio (TDCR in English, RCTD in French) method.





Sabot B. , et al., 2022, Performance of portable TDCR systems developed at LNE-LNHB, NIMA 1034

4π β-γ Measurements

Coincidences technique

- Widely used in the field of radioactivity metrology, the 4πβ-γ coincidence method (Campion, 1959) is a direct method developed for measuring the activity of radionuclides whose decay is accompanied by at least two distinct types of ionizing radiation.
- Its use is not limited to the measurement of β-γ emitter activity, it also applies to radionuclides whose α is followed by the emission of γ photons.

LNHB device

- The detection device and counting electronics consist of two separate and independent channels (known as β and γ), each of which is exclusively sensitive to a single type of radiation. The peculiarity of the coincidence method applied with classical instrumentation lies in the addition of an extra channel (called coincidence channel) whose counting is incremented each time a decay is detected simultaneously in the two channels β (4π proportional counter) and γ (NaI(Tl) scintillator detectors).
- The activity of a radioactive source is determined by calculating the ratio between the product of the individual channel count rates (β and γ) and the coincidence count rate. The particularity of the method is that this calculation does not require knowledge of the detection efficiency of the detectors used

Bobin, C., Thiam, C., Bouchard, J., Jaubert, F., 2010. Application of a stochastic TDCR model based on Geant4 for Cherenkov primary measurements. Appl. Radiat. Isot. 68, 2366–2371.

Bobin, C., 2007. Primary standardization of activity using the coincidence method based on analogue instrumentation. Metrologia 44, S27-S31.

4π α-γ measurements

- Activity measurement (e.g. of Ac-225) by the 4πα–γ anti-coincidence spectroscopy method using a ultrathin plastic scintillator sheets and a Ge-detector.
- Example: the detection of α-particle from Po-213 and interference of β-particle from Bi-213 was successfully observed.
- The present method is expected to be applicable to other complex decaying α/β decay chains.

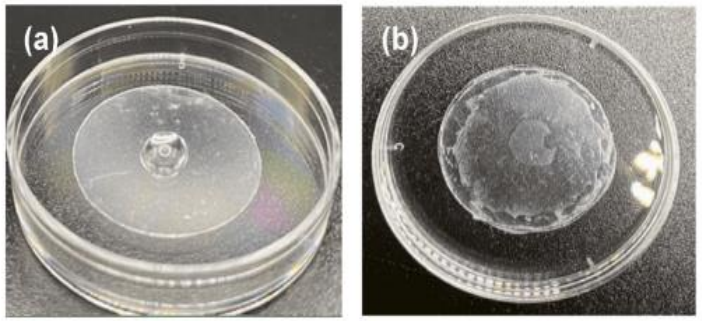


Fig. 2. Source sandwiched between two plastic scintillator sheets. (a) Droplet of ^{225}Ac solution deposited on a plastic scintillator sheet (b) Finished source.

K.-i. Mori et *al*.

- Quite complicated: approximately 95% of α-detection efficiencies for Po-213 and Fr-221 was experimentally obtained with few interferences of β-particle.

https://doi.org/10.1016/j.apradiso.2023.111061 Ken-ichi Mori et *al*.

K.-i. Mori et al.

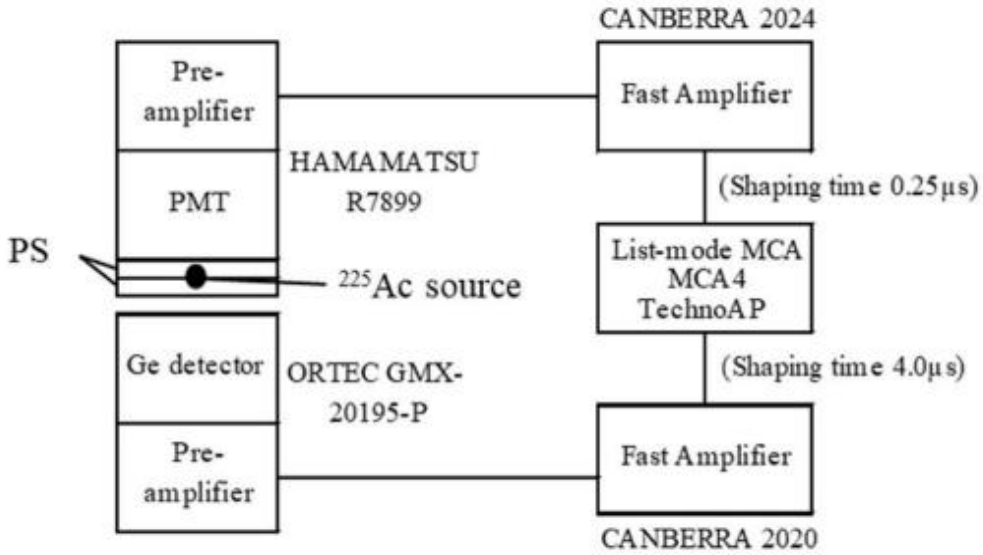
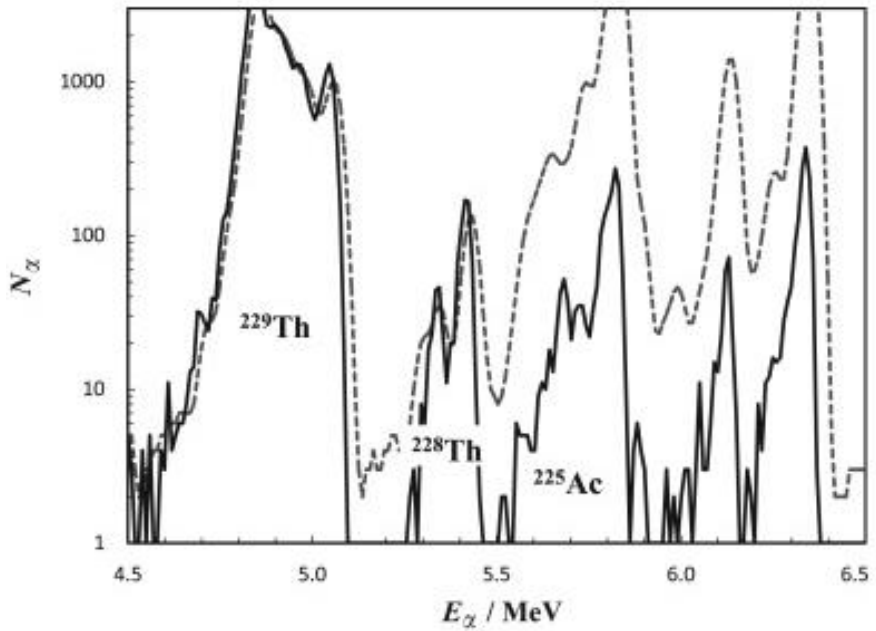


Fig. 3. Schematic of the 4πα–γ anti-coincidence spectroscopy system.

LTAC-TDCR

- This method is coming from the NIST: a new primary standardization of the alpha-emitter Ra-223 using Live-timed Anticoincidence Counting (LTAC) and the Triple-to-Double Coincidence Ratio Method (TDCR).
- The LTAC is based on 4πβ–γ anticoincidence counting.
- e.g. Silicon surface barrier detector alpha spectrum of normalized counting rates Nα. Dashed line is before Th-229 purification and solid line is after purification. After purification, it was possible to resolve the Th-228 from the Ac-225 ingrowth:



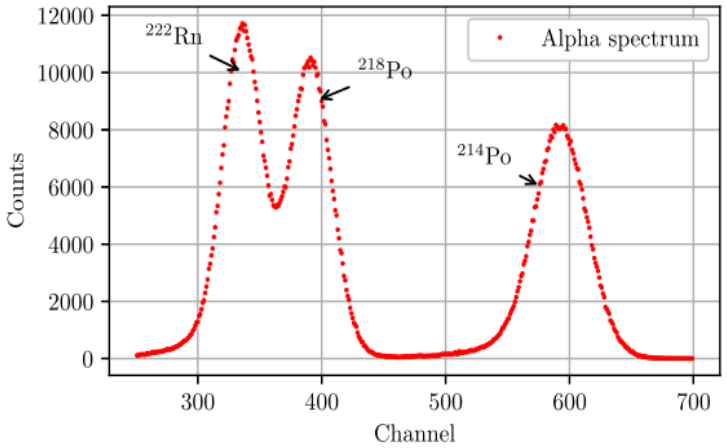
Fitzgerald et al., 2010

A new primary standarsization 229^{Th}, Fitzgerazld et al., 2010, doi:10.1016/j.apradiso.2010.01.007

Fitzgerald, R., Schultz, M.K., 2008. Liquid-scintillation-based anticoincidence counting of Co-60 and Pb-210. Appl. Radiat. Isot. 66, 937–940, doi:10.1016/j.apradiso.2008.02.037

Plastic scintillators

- Plastic scintillators discriminating alpha rays from beta rays emitted by a radioactive medium and method for discriminating alpha rays from beta rays using these scintillators are presented in: Matthieu Hamel, Romuald Woo, Benoît Sabot, Krasimir Mitev, Patent: WO2023166261A1
- Evaluation of radon absorption and detection properties of a plastic scintillator developed for PSD measurements V. Todorov, S. Georgiev, M. Hamel, C. Dutsov, B. Sabot, I. Dimitrova, K. Mitev. H ttps://doi.org/10.1016/j.measurement.2024.114554 Received 12 January 2024.



Alpha spectra of Rn-222 and its progeny in the PSDplastic.

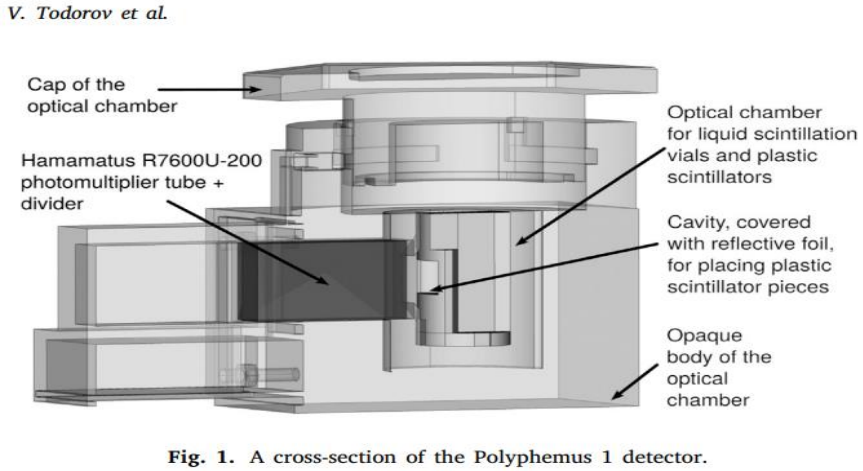
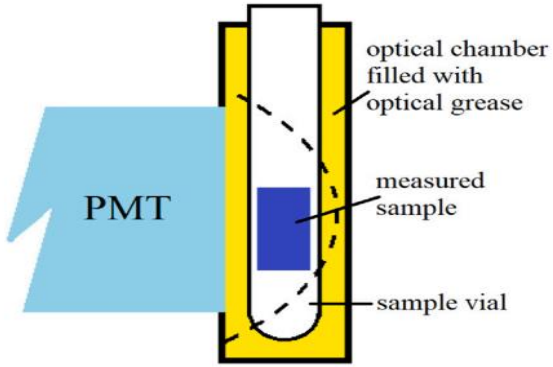


Fig. 1. A cross-section of the Polyphemus 1 detector.

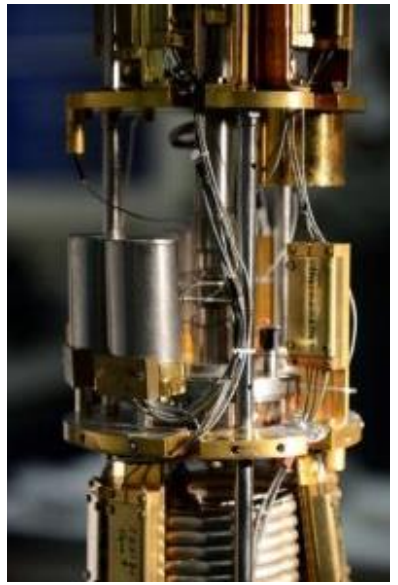


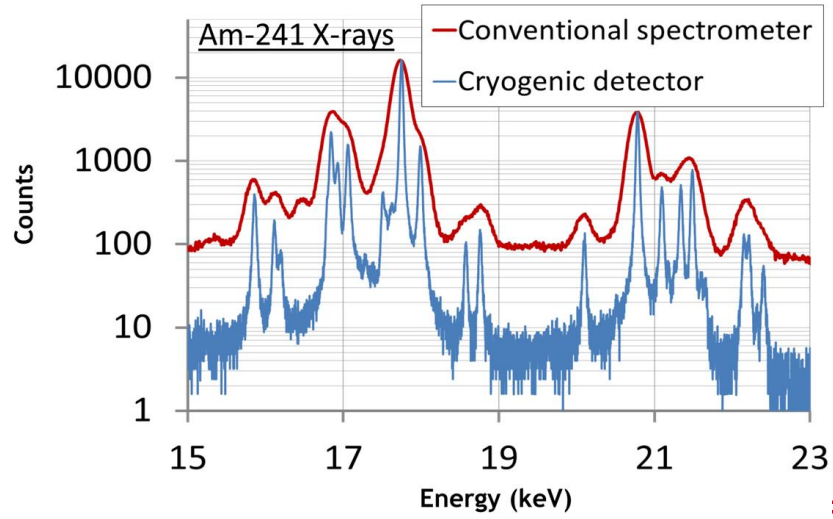
Cryogenic detectors

- A cryogenic detector is a thermal detector that measures the energy of each absorbed particle as a temperature rise through an extremely sensitive and fast thermometer.
- The thermometers used are magnetic thermometers which convert a temperature variation into a magnetization variation.
- A low-noise SQUID (“Superconducting Quantum Interference Device”) converts the magnetization variation to a voltage change and the dedicated electronics amplifies the signal.
- High sensitivity can only be achieved by operating the detector at a very low temperature (approximately 20 mK above absolute zero, i.e. -273.13 °C) in order to reduce thermal noise sources and increase the amplitude of the measured signal. These extreme temperatures are achieved by means of a He-3 / He-4 dilution refrigerator. Cryogenic detectors then offer performance that is inaccessible to conventional semiconductor or scintillator detectors in terms of energy resolution and detection threshold.

Determination of absolute L X-ray emission intensities from 241Am decay using a metallic magnetic calorimeters, Matias Rodrigues et al 2023 Metrologia 60 025005 https://doi.org/10.1088/1681-7575/acb99f

3 pictures from LNHB device





Gridded Ionisation Chamber

- When an alpha particle travels through a medium (ex argon mixture), it loses most of its energy by ionising the atoms in this medium.
- An electric field is placed across the chamber to separate and collect the electrons and the ions.
- Another grid is placed on an intermediate voltage dividing the chamber volume into two sections: the interaction region (cathode to grid) and the collection region (grid to anode). The particle track is fully ionised inside the volume of the interaction region. Only the electrons then travel through the collection region.
  - A mixed α-particle source (Np-237, Am-241, and Cm-244) gave an excellent energy resolution of 59 keV (1.0%) in FWHM (for 5.81 MeV Cm-244) which is enough to identify major sources of alpha radioisotopes (U-238, U-235, Np-237, Pu-239, Pu-238, Am-241, Cm-244) to be found near nuclear power plants.

ALPHA PARTICLE SPECTROSCOPY BY GRIDDED IONIZATION CHAMBER, Nuclear Instruments and Methods in Physics Research 223 (1984) 285-289

α-particle spectrometer based on xenon gas ionization chamber using coplanar electrodes, N. Hasebe a,b et al., NIM A, vol 925, 1 May 2019, Pages 123-127, https://doi.org/10.1016/j.nima.2019.01.079

A. Hartmann et al. / Nuclear Instruments and Methods in Physics Research A 814 (2016) 12-18:

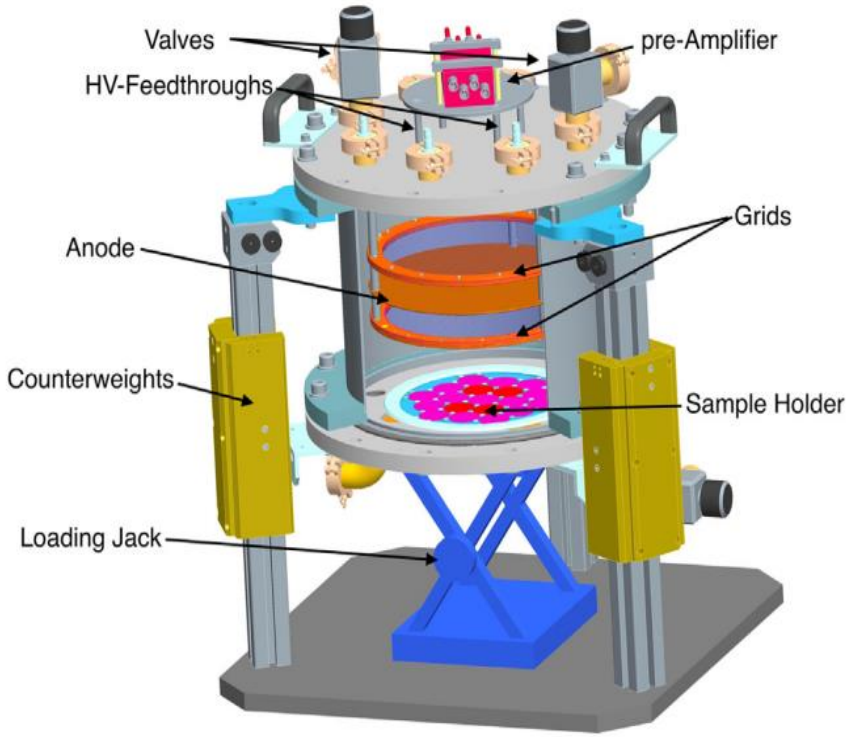
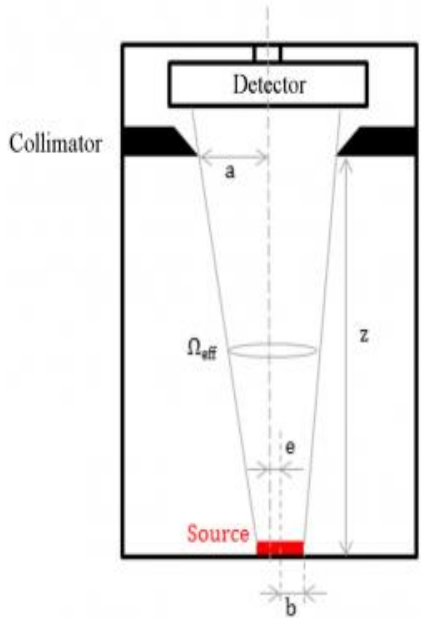


Fig. 1. Schematic drawing of the chamber. The grids and anode can be seen in the centre of the chamber. The first part of the signal manipulation can be seen on the top of the chamber, starting with the preamplifier. The valves are connected to the P10 canister for flushing. The jack and the counterweights are designed to allow for simple sample loading. The sample holder is interchangeable to fit different sample types and geometries.

Defined Solid Angle Method

- An alpha particle detector with 100% detection efficiency is placed at a precisely known distance from a discreet source such that the detector subtends a known solid angle (Pommé, 2007).
- The flux of alpha particles sampled over this solid angle can then be used to calculate the total emission rate for an isotropic source, which is combined with the decay data to calculate the source activity.



Schematic of DSA

Defined solid angle counter with variable geometry, Garcia-Torano E, et *al*., ARI 66 (2008) 881-885

DSA at CIEMAT: DOI: 10.1016/j.apradiso.2008.02.040

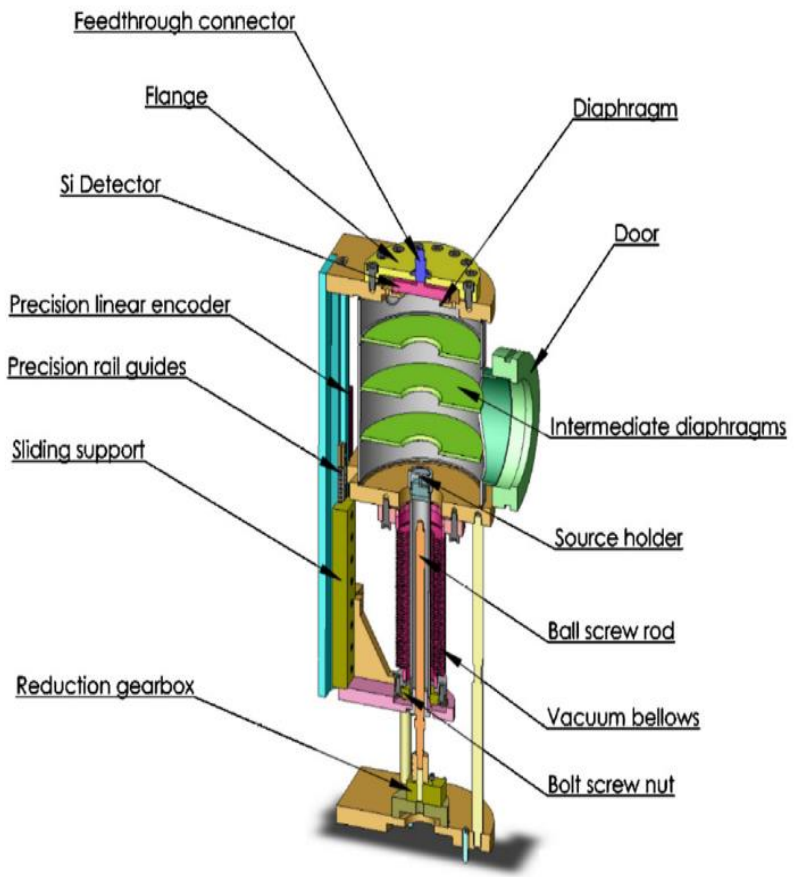


Fig. 1. Diagram of the counter showing the main components. Alpha particles are detected with a 2000 mm^2 Si detector. Diameter of the main diaphragm, source-to-detector distance and activity distribution of the source define the geometry of the measurement. Source to detector distance can be continuously varied between 80 and 190 mm.

Defined Solid Angle Method

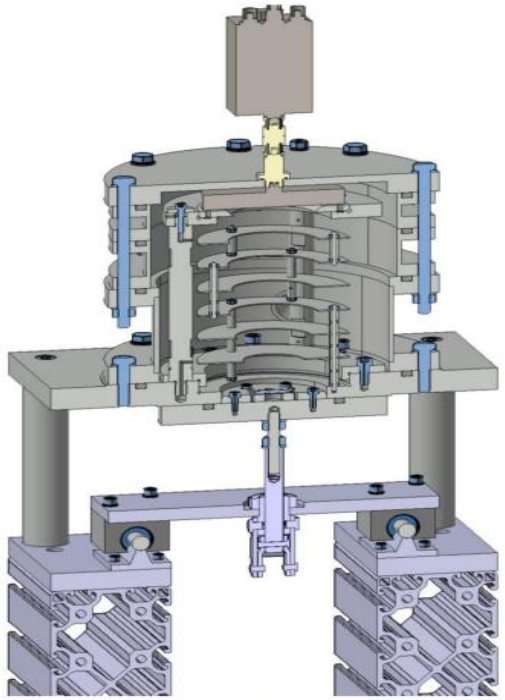


Fig. 2. CAD representation of the DSA counting system showing the retractable positioning platform with the source holder, the anti-scattering baffles, diaphragm, detector, preamplifier and the outer shell of the chamber.

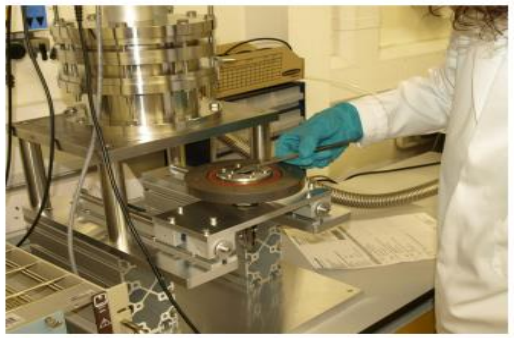


Fig. 3. Picture of the DSA counter with the source holder down.

DSA at NPL:
DOI:
10.1016/j.apradiso.2015.11.073

- While to date there are no examples of DSA being used to measure the activity of a medically important alpha emitting radionuclide, Marouli et al. (2019) discuss measurements of Ac-227 in equilibrium with its progeny, including Th-227 and Ra-223.
- The contamination risks posed by diffusion of radon progeny and high energy recoils that Marouli et al. address will be common to the decay chains of interest for medical applications. For activity measurements “contamination” means lost counts, as do the common measures taken to avoid contamination, e.g., covering dried sources with thin protective films.

Marouli, M., et al., 2019. Measurement of absolute γ-ray emission probabilities in the decay of 227Ac in equilibrium with its progeny. Appl. Radiat. Isot. 144, 34–46.

Defined Solid Angle Method

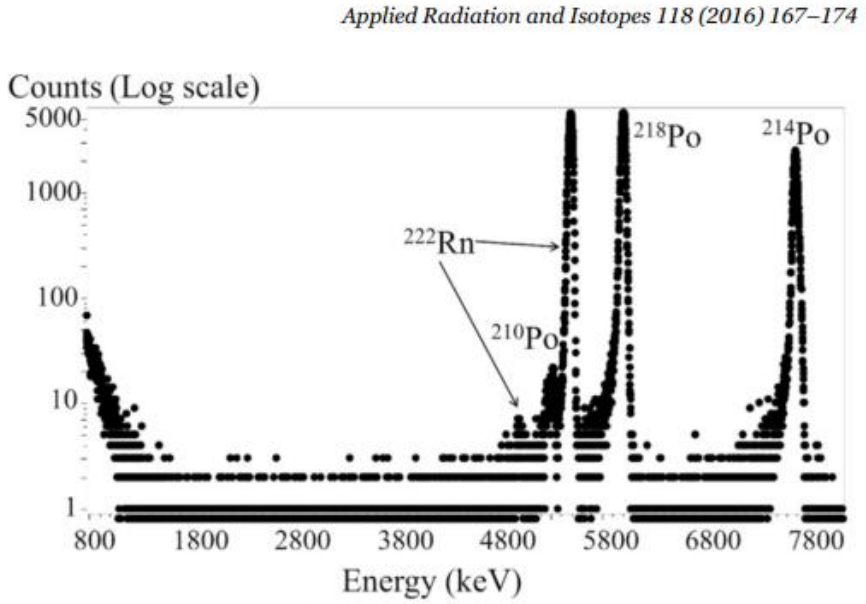


Fig. 3. Typical alpha spectrum of the frozen radon source.

An absolute radon 222 activity measurement system at LNE-LNHB, Sabot B., ARI 118 (2016) 167-174

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Thank you

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