# IMPROVED GAMMA SPECTROMETRY OF VERY LOW LEVEL RADIOACTIVE SAMPLES

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## ABSTRACT

Today, many laboratories face the need to perform measurements of very low level activities using gamma spectroscopy. The techniques in use are identical to those applicable for higher levels of activities, but there is a need to use better adapted materials and modify the measurement conditions to minimize the background noise around the area.

CEA's (SPR/SARCLAY) design of a very low level activity laboratory has addressed the laboratory itself, the measuring chamber and the detector.

The lab is constructed underground using specially selected materials of construction. The lab atmosphere is filtered and recycled with frequent changeovers. The rate of make-up fresh air is reduced and is sampled high above ground and filtered.

The chamber volume, purposefully reduced, is constructed using a 2 centimeter thick lead shell of very low activity ($10^{-3}$ dps/gramme) plus a secondary shell 13 centimeters thick of low activity lead($6 \times 10^{-2}$ dps/gamme). The internal coverage of copper and cadmium is chosen to minimize backscattered radiation as well as the X-ray fluorescence of lead.

The detector of high relative efficiency (63%) is mounted in a cryostat with angled design for the cold finger. Only the detector itself is inside the cell. All construction materials are preselected. Lead and aluminum parts are specially designed for very low background contributions.

All the efforts in reducing background have resulted in an improvement on the detection capabilities as:

- Disappearance of parasitic radiation from K40, Cs137, Co60 and Ra226. The only peaks left, barely discernible, are the 511 Kev annihilation gamma and the 351 and 609 Kev peaks of Bi214.
- Improved detection limits for Co57, Cs137 and Co60: 14, 25 and 22 dps/cubic meter respectively.
- Better use of lab time, since we can obtain regular levels of detection using acquisition times shorter by a factor of 16.

## INTRODUCTION

"Low level" measurements using gamma spectroscopy are those which require detection of activities below $10^{-2}$ Bequerels (dps) per cubic centimeter or per gram.

The need for those measurements has increased control on the environment, the nuclear facilities, radioactive fallout, food-stuffs, natural radiation and especially migration of radionuclides from waste sites.

Several laboratories are involved in developing methods capable of detecting such low levels, especially by reducing the background to its lowest possible levels. (Fig. 1)

In order to reach this goal, the CEA (French Atomic Energy Commission) has improved on:

- the laboratory facilities
- the measurement chamber
- the detector

While at the same time:

- Installing the equipment at the Saclay site, close to the facilities being surveyed.
- Associating the measuring chamber with an automatic changer using a standard sample geometry.
- Respecting the budget constraints on price of similar equipment.

## RADIATION SOURCES

In order to minimize the background, we need to address:

- Cosmic radiation effects
- Radiation from natural radionuclides
- Radiation from artificial radionuclides

### Cosmic Radiation

We know that cosmic radiation includes:

- Primary components (protons, neutrons, alpha, heavy ions, etc.) absorbed progressively by the atmosphere.
- Secondary components resulting from the interaction of the primary components with nuclei of the atmosphere atoms. Those are nucleons, electrons and mesons with energies between 1 and 10 Mev and with an average flux of 1 particle per minute per square centimeter.
- Radioactive products created by the secondary

Fig. 1. Photo of the "Low Level" Laaboratory of CEN/SACLAY (SPR/SRSE).

components (Tritium, Beryllium 7, Carbon 14 and Sodium 22).

**Natural Radioisotopes**

The four elements mentioned above (Tritium, Beryllium 7, Carbon 14 and Sodium 22) are part of the natural background but their activity is insignificant in gamma spectroscopy.

The significant part of the natural background is the soil. The remaining radioisotopes in the soil are those with long half lifes. Other initially present but shorter lived have already completely decayed.

**Artificial Radioisotopes**

Several radioisotopes are in use in non-nuclear facilities. Additionally, experimental nuclear explosions, nuclear plant accidents and other mishaps have generated contamination of the environment with fission products, activation products and fissile materials aerosols.

The most frequently found isotopes in gamma spectrometry are Cesium 137 and Cobalt 60.

## METHODS OF PROTECTING FROM THE BACKGROUND

**Cosmic Radiation**

There are two possible methods to minimize the background due to cosmic rays:

- Shields of the right material and thickness.
- Elimination of the peaks in the spectrum.

TABLE I

Naturally Occurring Isotopes in Soil

| Isotope | Half Life ($10^9$ years) | Activity |
|---|---|---|
| Uranium 238 | 4.5 | $3x10^{-4}$ to $6x10^{-2}$ |
| Uranium 235 | 0.7 | $1.4x10^{-5}$ to $2.8x10^{-3}$ |
| Thorium 232 | 14.1 | $7x10^{-3}$ to $8x10^{-2}$ |
| Potassium 40 | 1.3 | $8x10^{-2}$ to $10x10^{-2}$ |

**Natural Background**

The means differ depending on whether we deal with radioactive solids in the materials or with gaseous products including aerosols.

**Solids Included in Construction Materials**

Radionuclides are present:

- In the building materials
- In the shield
- In the materials used for the detector construction

Rigorous selection of all those materials for radioactivity content is imperative, especially for the shielding screens.

**Gases**

Rn220 and Rn222 come from U238 and Th232. They are released from the soil, water and building materials. They decay by alphas and produce daughters which are themselves alpha, beta and gamma emitters.

Shortly after formation, the new ions are absorbed in the particles present in the atmospheric air and in aerosols. The quantities of those gases are dependent on the soil and building characteristics, meteorological conditions and height. Their effects vary greatly with the speed of the ventilation air.

## LABORATORY CONSTRUCTION

In order to build the "low activity measuring lab" for the "Protective Services against radiation of CEN-SACLAY", the following constraints were followed: (Fig. 2.)

- The building was built underground, with regular materials screened for radioactive contents.
- A major part of the laboratory atmosphere is recycled after proper filtration.
- The small make up fresh air is obtained high above ground.

**Location**

The location was chosen away from the nuclear laboratories which could increase the ambient background, but within a reasonable distance for those installations which it serves.

The lab laboratory is in an L shape. Working area is 84 square meters (900 square feet) and is approximately 10 feet underground. Floors, walls and ceilings are concrete. Their thicknesses are 15 and 34 centimeters (6" and 14") respectively. They are the first barrier against cosmic radiation. (Fig. 3)

All the materials used (like sand, stone and cement) have been preselected for their gamma background levels. Sand and cement are materials which contain Potassium 40, Radium 226 and Thorium 232.

TABLE II

Activity in dps/gram

| | K40 | Ra226 | Ac228 | Cs137 |
|---|---|---|---|---|
| Sand | 0.2 | 0.07 | $6x10^{-3}$ | -- |
| Cement | 0.2 | 0.2 | $6x10^{-2}$ | -- |

**Ventilation**

The decay products of Rn220 and Rn220 are solids. They are generated in the atmosphere and attach to dust particles and later can be deposited on the detector and/or measurement cell and/or laboratory walls.

Fig. 2. Automatic Changer for Low Activity Samples.

They can increase the background noise of the detector. Proper filtration of the in/out air into the measuring areas can minimize this effect.

**Air Filtration**

There are two filters in series one filter upstream of the fan/compressor, two filters downstream (paper/glass fibers).

The air is blown in by eight ducts. There are twenty air changes per hour.

**Fresh Air Intake**

Air intake is at the roof level, 20 meters above the ground. It is known that Rn220/Rn222 concentration decrease by a factor of 10 each 10 meters above ground.

**Leak Tightness of the Laboratory**

The lab environment is kept at a slight overpressure with respect to the exterior, assuring leak tightness. This is in addition to the construction safeguards. All walls and floors are covered by epoxy resin print.

## MEASURING CELL

The cell needs to protect the detector against the background created by the building materials and samples on

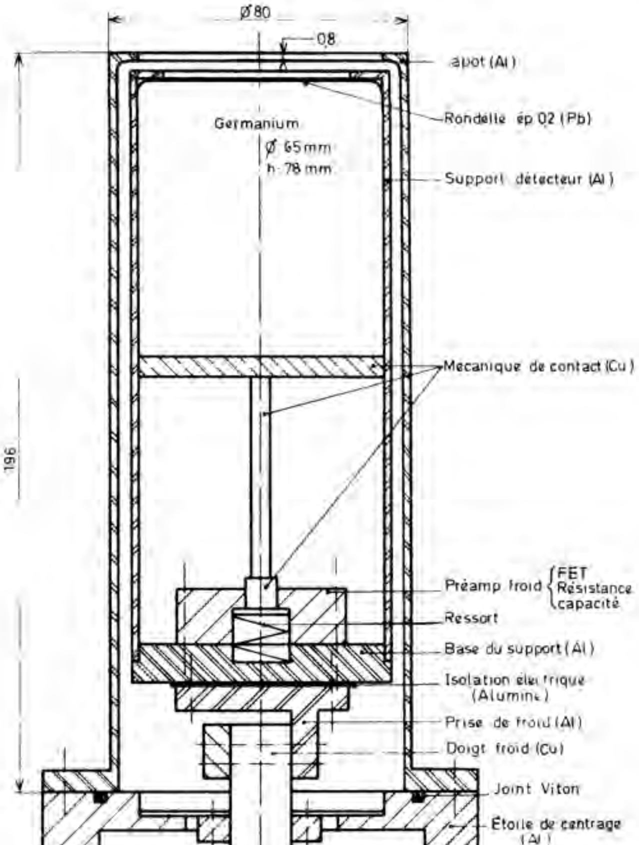

Fig. 3. Scheme

standby. The shield is made of a dense material but of very low activity.

**Protective Material Lead**

Radioactivity of lead varies in a ratio of 1 to 100. This activity is due to artificial radioactive impurities, but most importantly due to intrinsic impurities from the Uranium and Thorium decay chains.

If Uranium and Thorium are separated correctly during the lead refining process the only isotope present will be Pb210 with a half life of 22 years. We therefore will like to use old lead in order to minimize the amount of radioisotopes in it. We use the denomination TFA when the mass activity of lead is below 9x$10^{-3}$ dps/gram.

The gamma rays from Pb210 are of low energy: 12 and 46 KeV. This energy is not relevant for gamma spectroscopy. *For in installation* of very low levels we require only the internal wall of the enclosure to be made of TFA lead.

**Sample Changer**

ATEA CMSA-10 (measuring cell for automatic spectroscopy with 10 samples). The samples can have various geometrics and include Marinelli Beakers. The usable dimensions of the cell are:

- 290mm diameter ( 12")
- 270mm height ( 11")

The volume is minimized to reduce the background factor due to the air and the mass of lead required for a given wall thickness.

The outside shielding is made of:

- 2cm ( ~ 1")of lead TFA ($10^{-3}$ dps/gram)
- 13cm ( ~ 15") of lead FA (6x$10^{-2}$ dps/gram)

The internal lining of the cell minimizes the backscattered radiation, the lead radiation and the lead x-rays induced by gamma radiation. It is made of a few millimeters of copper and cadmium. Those are high purity materials having less than 3x$10^{-4}$ dps/gram (Uranium and Thorium) of very low energy x-rays.

TABLE III
X-Rays of Cell Shielding Materials

| | k alpha 2 | k alpha 1 | k beta 1 | k beta 2 |
|---|---|---|---|---|
| Head | 73 | 75 | 85 | 87 |
| Cadmium | 23 | 23 | 26 | 26 |
| Copper | 8 | 8 | 9 | - |

The cell is leak tight with the possibility to circulate an inert ???? with low radon content.

## THE DETECTOR

In order to detect very low activities, we need the highest efficiency detector possible but most important, with the best signal to noise ratio. Choice of the detector is then based on efficiency, energy resolution and sensitivity to background radiation.

Sensitivity to parasitic radiation is dependent on the detector design as well as of the materials used in the construction.

The gamma detector, manufactured by Intertechnique, has the following characteristics:

- High relative efficiency: 63% with a usable volume of 255 cubic centimeters.
- Energy resolution of 2Kev at 1.33 Mev 0.96Kev at 122 Kev
- Peak/Comton ratio of 66

The detector has an angled concept to allow only the detector itself in the measuring cell and avoid the potential radioactive sources in the liquid Nitrogen reservoir, preamplifier and molecular sieve.

All the materials have been preselected. The mass activity requirements are:

| | |
|---|---|
| U + Th | < 1.7x$10^{-4}$ dps/gram |
| Cs137 | < 1.7x$10^{-5}$ dps/gram |
| Co60 | < 1.7x$10^{-5}$ dps/gram |

This measurement has been conducted in the underground laboratories in MODANE, in collaboration with CEN-BG (M. Hubent and Professor M. Menraht) (Fig. 4)

## RESULTS

### Activity of the Laboratory Atmosphere

This activity is determined by sampling using atmospheric filters and measurement of the Rn222 daughters. The results obtained showed the important role of the filtration system since the activity of Bi214 detected was 100 times lower than the ground level activity.

In contrast, the epoxy resin cover of the walls have not brought the expected improvement.

### Background Gamma Spectrum

Three different background measurements, each 18 hours long, have been made using the high efficiency (63%) detector.

- The energy range was 30-2000Kev over 4096 channels.
- The detector is in the filtered environment.
- Those spectra show the effect of the shielding.
- The first, without shielding, gives the environment activity.
- The second, with 10cm of lead FA, using a large cell which included detector, coolant reservoir and preamplifier.
- The third is in the sample changer cell, with a 15cm lead shielding which included lead of TFA quality.

The Table IV presents the results: (64,800 seconds)

TABLE IV
Background Spectra

| | Shield Thickness (cm) Quality | 0 (N/A) | 10 (FA) | 15 (TFA) |
|---|---|---|---|---|
| Radioisotopes | Energy (Kev) | Area Under the Peak (counts) | | |
| Ra226 | 186 | $1.1 \times 10^4$ | 205 | |
| Bi214 | 351 | $4.9 \times 10^4$ | 407 | 77 |
| | 609 | $5.2 \times 10^4$ | 357 | 53 |
| | 17860 | $1.4 \times 10^4$ | 85 | |
| Ac228 | 911 | $3.1 \times 10^4$ | 90 | |
| K40 | 1460 | $1.2 \times 10^5$ | 455 | |
| Cs137 | 662 | $1.8 \times 10^3$ | 171 | |
| Co60 | 1332 | | 63 | |
| Annihilation | 511 | $1.9 \times 10^5$ | 2004 | 1475 |
| Compton Noise | 120 | $3.5 \times 10^4$ | 145 | 35 |
| counts/ | 364 | $4.0 \times 10^3$ | 55 | 28 |
| channel | 662 | $1.4 \times 10^3$ | 25 | 15 |
| | 1330 | 340 | 9 | 4.5 |

The first spectra shows a complex radioactive environment made of a high compton background with numerous

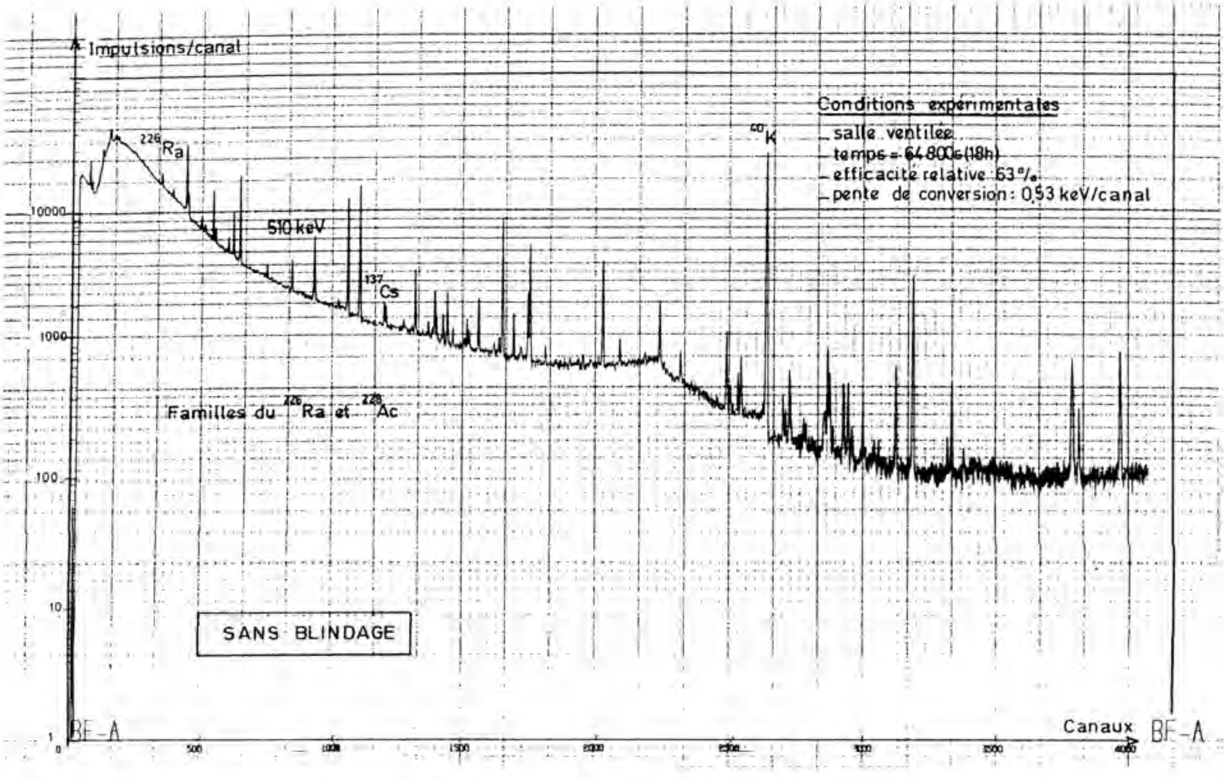

Fig. 4. Conditioned Experiments.

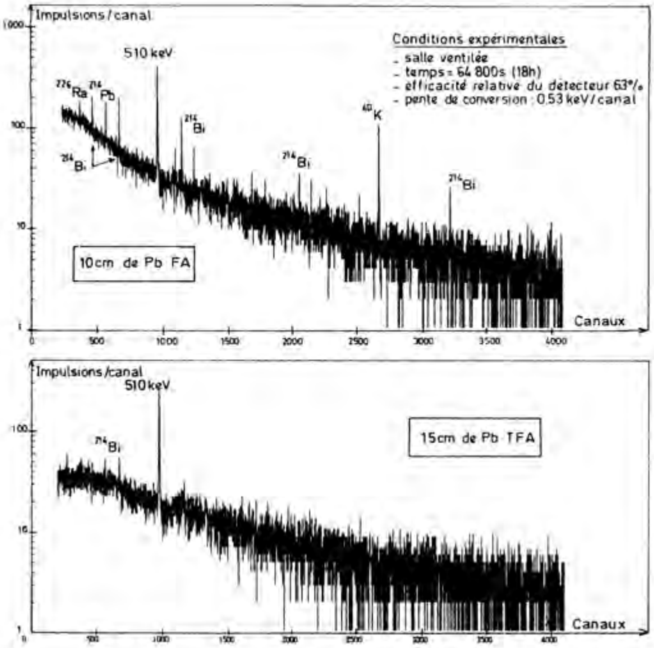

Fig. 5. Experiment Results.

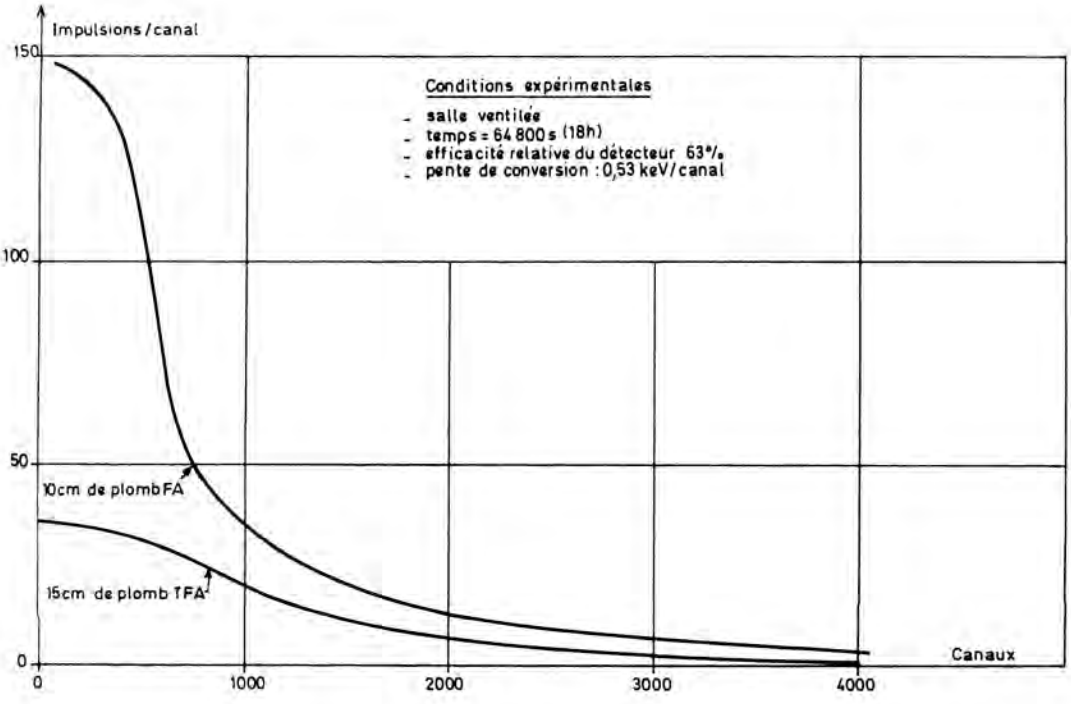

Fig. 6. Experiment Results

Tn222 gammas and daughters, Ac225, K40 and Cs137. The second spectrum demonstrates that 10 cm of lead diminishes the background but the gamma peaks are still present. They could come from the environment but also from the coolant reservoir and preamplifier. The third spectrum shows a reduction of the compton background by a factor of 4 at 120Iev, 2 at 1330Kev and the paronte peaks for Ra226, K40, Cs137 and Co60 have disappeared.

**Calculated Minimum Detection Limits**

The MDL's are calculated using the 18 hour spectra for Co57, I-181, Cs137 and Co60. Those isotopes are chosen for their appearance in the spectra and their "energy" position (low, mid, high-energy).

The MDL, in dps/cubic meter assume a Marinelli Beaker of 3 liters (0.003 cubic meters) using the following relation:

$$MDL = \frac{8.8 * SQRT(R * B)}{t * \Gamma * \varepsilon * V}$$

where MDL = Minimum Detection Level

R = Detector Resolution at the given energy (Kev)

B = Compton background at the given energy (counts*Kev)

t = Analysis time

Γ = Gamma emission percentage of the radioisotope

ε = Gamma efficiency at given energy and experimental conditions

V = Analyzed liquid volume in cubic meters

The results are tabulated in Table V

TABLE V
MDL's
GAMMA SPECTROSCOPY

| | | CLASSIC | WITHOUT BACK-GROUND | |
|---|---|---|---|---|
| Relative efficiency of detector | | 20% | 20% | 63% |
| Shielding thickness | | 5 cm | 10 cm | 15 cm |
| Lab ventilation | | no | yes | yes |
| Isotopes | Energy (Kev) | Minimum Level of Detection dps/cubic meter | | |
| Co57 | 122 | 88 | 77 | 14 |
| I131 | 364 | 113 | 79 | 24 |
| Cs137 | 662 | 112 | 80 | 25 |
| Co60 | 1332 | 110 | 80 | 22 |

We show a definite gain of sensitivity:factor of ~5 for Co57 and ~3 for I131, Cs137 and Co60.

## CONCLUSION

The results obtained in the new installation for low level gamma spectroscopy indicate a substantial improvement when the following criteria are followed

- A high efficiency, angled detector
- Careful selection of building materials (detector and cell)
- Proper ventilation
- 4 Pi geometry (Marinelli Beaker).

Application of the concepts above show:

- Disappearance of the parasitic peaks of K40, Cs137, Co60, Na226 in an 18 hour long spectrum
- Minimization of the 56Kev contribution due to cosmic rays.
- Marqued improvement of the MDLs:

14 dps/cubic meter for Co57

25 dps/cubic meter for Cs137

22 dps/cubic meter for Co60

Count time reduction by a factor of 15 to obtain same levels as regular low background techniques (column 2, table above).

For more information call:

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