Minireview

Underground Gamma-Ray Spectrometry†

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Abstract

Gamma-ray spectrometry using high purity Ge detectors has made significant advances in recent years because large crystals have become readily available and the importance of very radiopure materials in the construction of detectors has been understood. The combination of these improvements has made it possible to decrease detection limits in special low-background counting systems. Gamma-ray spectrometry systems located underground are particularly improved by the new developments. This paper deals with the current state-of-the art of underground gamma-ray spectrometry as well as providing examples of new applications of underground gamma-ray spectrometry that were made possible due to the advances in detectors and technique.

Key words: gamma-ray, spectrometry, underground, HPGe, cosmic rays, radioactivity.

Introduction

Ever since the introduction of the Ge(Li) detectors in 1963,[1] gamma-ray spectrometry using Ge-crystals has constituted the backbone of every modern up-to-date radioactivity laboratory. This is due to the ease with which quantitative estimates of many gamma emitting radionuclides in unknown samples can be obtained with little sample preparation (mostly without any chemical steps and often non-destructively). The wide range of measurable gamma-ray energies (below 10 keV to 10 MeV) and the high energy resolution (~0.1%) make this technique a true multi-nuclide/isotope determination method. There has always been a need to measure small amounts of radioactivity (so called low-level measurements) in gamma-ray spectrometry, but traditionally this was better done using NaI detectors with high counting efficiency. Improvements of Ge–detector technology have led to bigger Ge crystals (and higher counting efficiency) and have given Ge-detectors an edge over NaI-detectors in spite of their higher cost.

The main contributions to the background in gamma-ray spectrometry come from (i) ambient radioactivity in the laboratory (ii) ^{222}Rn and its progeny (iii) radioactivity in the detector and the shield and (iv) cosmogenically induced background.[2,3] The detector can be shielded from environmental radioactivity in the laboratory, e.g. walls, relatively easily by the use of high-Z materials e.g. lead. The radon contribution can be reduced by active measures like e.g. ventilation. The radioactivity in the detector and the shield are very cumbersome to reduce because of the extremely radiopure materials that must be used. If the first three contributions are taken care of the contribution from cosmogenically induced signals dominates the background spectrum. The best way to reduce this contribution is to place the detector deep underground. In recent years the number of Ge-detectors in operation underground has increased steadily and so has the number of underground laboratories. Many laboratories are located in existing underground chambers abandoned for various reasons. In e.g. the Baradello hill in Italy, a laboratory is being established at a water purification plant[4] and in south of France in a former military installation.[5] The Felsenkeller laboratory in Dresden[6] is located in a former storage place for ice and beer inside a hill with easy access. The easy access led to the decision of placing a complete radioactivity laboratory (not only gamma-ray spectrometry) there although the background reduction for other techniques e.g. liquid scintillation counting, is not as great (about a factor of 2).

The HADES (High Activity Disposal Experimental Site) underground laboratory in Mol was constructed in order to study the geological properties of the

surrounding clay layer and whether it is suitable to have the final repository of nuclear waste in such a stratum. There are many studies taking place and the laboratory is growing. IRMM (Institute for Reference Materials and Measurements) performs gamma-ray spectrometry in HADES and the measurement capacity has recently been increased. This was triggered by needs for low-level measurements in fields like reference measurements,[7] validation of other techniques,[8] isotopic fingerprinting (using trace amounts of radionuclides to study various processes in e.g. the environment[9] or for safeguards), neutron measurements[10,11] and materials testing for large scale detectors in fundamental physics research.[12]

Materials and methods

The development of Ge-detectors: The lithium-drift process necessary to produce Ge(Li)-detectors was first described by Pell in 1960.[13] In 1963 Tavendale published a paper describing a Ge(Li)-detector.[1] The improvement of the Czochralski technique to produce Ge-crystals of high purity, led to the development of high purity germanium (HPGe) detectors in 1972.[14] Ge(Li) detectors need to be kept at liquid nitrogen temperatures at all times, while HPGe detectors can be warmed up without degradation. Initially the high purity crystals were very small (at first 1 cm^3) so Ge(Li) detectors were produced in high numbers several years after the introduction of HPGe-detectors. Later developments[15] include larger crystals, thinner deadlayers, multi segmented crystals, multicrystal cryostats and low background detectors. This paper will mainly deal with the latter point. It must be pointed out that in order to obtain low detection limits, aspects other than the background including resolution, peak-to-Compton ratio and crystal size are of vital importance. Obtaining the desired parameters is not trivial as these parameters are interdependent, e.g. increasing the crystal size and thus increasing the efficiency usually means that both the resolution as well as the background increase as well. In the literature a range of more or less complex figures of merit (FoM) for gamma-ray spectrometry have been used.[16,17,18] For low level measurements it is useful to look at the expression (i), which in the first approximation is inversely proportional to the minimum detectable activity.

$$\text{(i)} \quad FoM = \frac{\varepsilon(E)}{\sqrt{R(E)B(E)}}$$

R is the resolution (i.e. FWHM in keV), B the background count rate per keV and ε the relative efficiency for a ^{60}Co point source 25 cm above the centre of the detector endcap (i.e. a commonly used parameter to summarize the efficiency performance of a Ge crystal relative to a 3" x 3" NaI crystal), all measured at the energy E.

Figure 1 shows the improvement of the FoM for the 1332 keV line of ^{60}Co (assuming no ^{60}Co in the background) as defined in Equation (i) over the past 40 years. The FoM improved a factor of 2 every 3.6 years, which is less significant but still comparable to developments in fusion, computer and accelerator technology[19] over the same time period. One can thus say that the interference free detection limit (i.e. without consideration of background produced by the sample itself and therefore the only characteristic which is exclusively relating to the measurement system) of a long-lived gamma-ray emitting radionuclide (with high emission probability, Pγ) using a Ge-detector, has decreased from a fraction of a Bq in 1963 to a fraction of a mBq today.

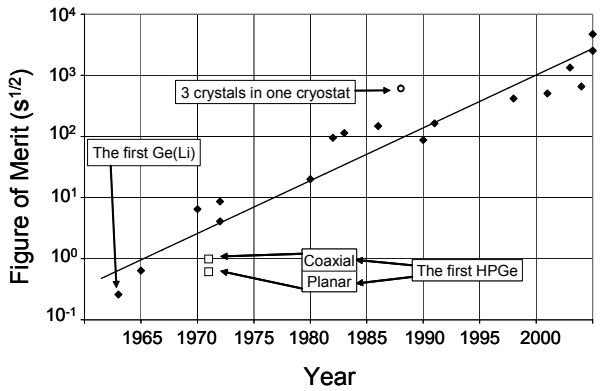

Figure 1. The FoM for Ge-detectors at 1332 keV as defined in Equation (i). The solid line is a fit to data points that were based on information found in literature as well as given by detector manufacturers. Note that the background is not necessarily the intrinsic background of the detector. This FoM does thus not solely show detector improvements but also the effect of improved passive shielding and use of underground laboratories.

The detection limit also scales with the inverse of the square root of the measurement time. In applications where extremely long measurement times (a few years) can be used, like in measurements of the double beta decay of ^{76}Ge, detection limits are in the order of a μBq.

Today it is possible to purchase low background detectors from a range of commercial manufacturers. Different manufacturers have slightly different approaches to low background detector design but some common features are: (i) pre-amplifier placed far from the crystal so that it can be placed outside the lead shield, (ii) the front-end electronics should be minimised and placed further from the detector than normal and maybe even be shielded by a cm or two of

dense material, e.g. electrolytic copper, (iii) soldering is minimised and if necessary pure tin is used, (iv) the use of glue is minimised, (v) welding is minimised and (vi) all other materials used inside the detector are such that they are known to be radiopure. The HPGe-detectors with the lowest background (e.g. the one reported by Neder, Heusser and Laubenstein[20]) have, however, been assembled using materials specially selected by the customer. As part of the joint research action IDEA (Integrated Double Beta Decay European Activities[21]) IRMM is involved in selecting materials for HPGe-detectors to be operated deep underground.

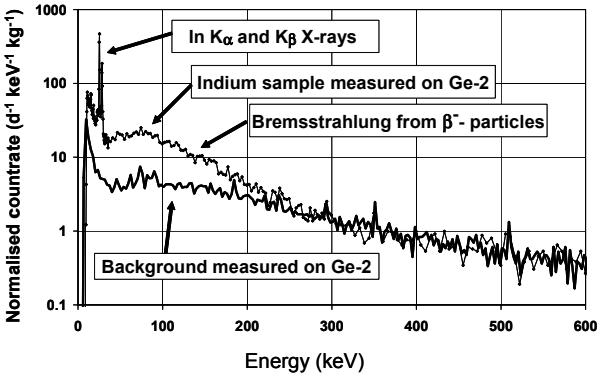

Figure 2. The gamma-ray spectra from a pure In-sample (mass 11.8 g) as well as background measured on a small (0.2 kg Ge) n-type planar HPGe detector in HADES.

Indium is an interesting example for the kind of problems one must be aware of in low-level gamma-ray spectrometry. Because of its physical properties it is commonly used in electrical contacts on Ge crystals as well as dopant. Indium consists to 95.7% of ^{115}In which is radioactive with the very long half live of about $5 \cdot 10^{14}$ years. Its background contribution is difficult to detect specifically since there is no gamma-ray associated with it. If the indium is sitting inside or near the Ge-crystal, the bremsstrahlung from the β^- -particles (with E_β-max 497 keV) will be registered and give a continuous background contribution like the one shown in Figure 2. Above ground, neutron activation of ^{115}In (thermal capture cross section of 200 barns) and the subsequent decay of ^{116}In add several gamma lines to the background spectrum. Indium should therefore be avoided in low level applications.

Underground measurements: The first underground gamma-ray spectrometric measurement has been reported to be measurements using NaI detectors in a derelict coal mine beneath Glasgow University[18] at a depth of 30 m. The first reported underground measurement using a Ge-crystal is likely to be in the paper from 1972 by Heusser and Kirsten,[22] who studied muon induced reaction in a laboratory located at the shallow depth of 15 m w.e. (m water equivalent).

Having in mind that many laboratories are located in the basement of tall buildings it is necessary to make a definition of what an underground measurement really is. Table 1 shows a proposed definition useful for gamma-ray spectrometry using Ge-crystals. Note that Table 1 is not necessarily applicable to experiments looking for rare events, where for example the cosmic rays can even not be neglected deep underground.

In this paper the acronym ULGS stands for ultra low-level gamma-ray spectrometry. This means that additional measures (such as using a muon shield or an underground laboratory) have been taken compared to low-level gamma-ray spectrometry, where a detector and shield made from selected radiopure materials are used.[23]

When performing ULGS with an active shield (or "veto counter" or "muon shield"), one gains a

Table 1. Definition of the idiom describing the depth of a laboratory and some characteristics related to gamma-ray spectrometry (not looking for rare events) in laboratories at different depths.

Depth (m w.e.)	Idiom	Characteristics
< 10	Not underground (above ground)	The soft component (e^-, e+, photons) of cosmic rays is strongly reduced and plays a minor part. Very little reduction of muon flux and neutron induced by muons. Muon shields are useful.
10 – 100	Shallow underground	The soft component of cosmic rays has vanished. The muon flux is reduced by a factor of 5-50. There is still a significant flux of neutrons produced by muons (reduction of factor 2-10). The activation of crystal and shield are still important factors. Muon shields are useful.
100 – 1000	Semi deep underground	Cosmogenic activation can be neglected. A slight improvement can still be obtained by discriminating against muons. The neutron flux is dominated by (α,n) sources rather than cosmogenic neutrons.
> 1000	Deep underground	The influence of the cosmic rays can be neglected. The only source for neutrons to consider are (α,n) reactions.

lot above ground and at shallow depth laboratories.[24] It is, however, not possible to avoid the cosmogenic activation of detector and shield so a muon shield can not completely substitute an underground location.

HADES: IRMM performs ULGS in the underground laboratory HADES, which is located at a depth of 223 m (500 m w.e.) at the premises of the Belgian nuclear research centre SCK•CEN (Studiecentrum voor Kernenergie, Centre d'Etude de l'Energie Nucléaire) in Mol, Belgium. It is the Economic interest grouping EURIDICE (European Underground Research Infrastructure for Disposal of Nuclear Waste in Clay Environment) that is responsible for HADES operation.[25] HADES is obviously a suitable name for an underground location but it is also the acronym for High Activity Disposal Experimental Site since it was constructed in order to study the possible location for the final repository of the Belgian nuclear waste. The clay layer has several advantages as a medium for storage of nuclear waste like high plasticity and not being permeable for water.

The ULGS work started in 1992 in small scale with one HPGe-detector[26] as an exploratory research project. The usefulness of the technique led to an increase of the capacity. For the moment there are 6 HPGe-detectors in HADES a 7th is under construction. The increased capacity was made possible as the available space in HADES has grown with the extension of new galleries. Figure 3 shows a schematic drawing of the present situation regarding shafts and galleries in HADES.

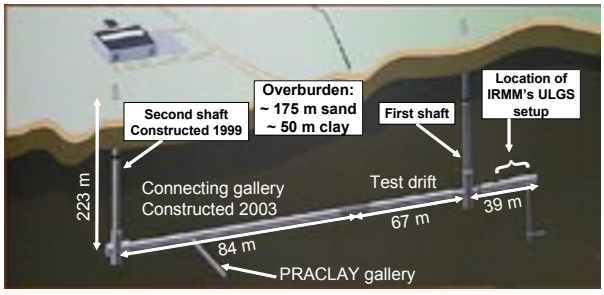

Figure 3. A schematic drawing of the HADES underground research facility in Mol, Belgium, at the premises of the Belgian nuclear centre, SCK•CEN.

The mission of IRMM's ULGS work is to support EU-policies and European Commission work in fields related to harmonisation of measurements, industry, nuclear safety and environment. This means that a wide range of measurements are necessary. This has led to the decision of equipping the laboratory with general purpose detectors rather than detectors specialised for a specific type of measurements. The non routine character of the work makes calculation of the detection efficiency for many types of samples a requirement. Although reference samples are used whenever possible, the general rule is that the efficiency is calculated using the Monte Carlo code EGS4.[27]

Applications

Several interesting developments in the use of underground gamma-ray spectrometry have arisen in recent years. The first major achievements were in the field of fundamental physics and double beta decay in particular. As the benefits of the technique became clearer to a wider community the number of applications has grown. The examples that follow shall illustrate the versatility of the technique.

Small samples: For normal environmental monitoring as well as for special investigation, a great number of samples are needed. If during a sampling campaign the amount of materials (in kg or m^3) can be reduced by a factor 100-1000, a lot can be gained. At the IAEA-MEL in Monaco an underground laboratory was recently inaugurated[25] in order to be able to reduce the size of marine samples collected during sampling campaign at sea. In a project carried out at IRMM it was shown that it is possible to measure ^{210}Pb distribution within individual bones (*in vitro*) of persons that were exposed to only normal radon doses. For this only very small samples can be used.[9] In collaboration with VKTA (Verein für Kernverfahrenstechnik und Analytik) in Rossendorf, IRMM measured freeze-dried lung tissue autopsy from former uranium-miners in the Wismut company. The ^{210}Pb activity in such samples could provide important information helping to quantify the effect of ^{222}Rn on the human health. Although there are many thousands of samples available each sample has only a small mass. Table 2 gives the results of 3 typical samples that were measured in HADES. The HADES measurements showed that it is possible to measure the ^{210}Pb activity in these samples using only a small n-type Ge-crystal (40 cm^3) and the not very labour intensive ULGS technique. The drawback is the long measurement time. These samples were measured for about one month, which was necessary in order to reach detection limits of 1 mBq/g (corresponding to 1.5 mBq total activity) in these samples.

High temporal resolution: Closely connected to the possibility of collecting small samples is the possibility of studying certain processes with high temporal resolution. At Kanazawa University in Japan it has been shown to be possible to measure ^{210}Pb and ^{7}Be in only 10 m^3 of air corresponding to 10 minutes of sampling by a high volume sampler.[28] Routine sampling every day was started instead of every week earlier.

Table 2. Results from various measurements performed in HADES. All uncertainties are given as the combined standard uncertainty following the Guide to the Expression of Uncertainty in Measurements. The decision thresholds are calculated following the ISO guide 11929-3, using $\alpha = 0.05$.

Type of project	Sample	Sample mass (g)	Massic activity (dry weight) of detected radionuclides (mBq/g)	Decision threshold for selected radionuclides (mBq/g)
Environmental radioactivity	Freeze dried lung tissue	1.38	^{210}Pb[a]: 2.6±0.7	
Environmental radioactivity	Freeze dried lung tissue	1.54	^{210}Pb[a]: 15.8±1.9	
Environmental radioactivity	Freeze dried lung tissue	1.43	^{210}Pb[a]: 1.8±0.5	
Reference measurement	Ashed human faeces	2.91	^{226}Ra: 16.2±1.4; ^{228}Ra: 21±4; ^{40}K: 4000±600; ^{137}Cs: 76±11; ^{210}Pb: 30±9; ^{228}Th: 12.1±1.5;	
Reference measurement	Ashed human faeces	2.98	^{226}Ra: 9.2±0.8; ^{228}Ra: 8.6±1.7; ^{40}K: 2700±400; ^{137}Cs: 7.1±1.1; ^{210}Pb: 20±6; ^{228}Th: 4.9±0.7;	
Reference measurement	Ashed human faeces	3.04	^{226}Ra: 13.3±1.3; ^{228}Ra: 17±4; ^{40}K: 5600±600; ^{137}Cs: 6.8±0.3 ^{210}Pb: 34±10; ^{228}Th: 7.3±1.5;	
Radiopurity study	Pb from the 13th century	705	^{210}Pb: 0.95±0.18; ^{226}Ra: 1.0±0.3; ^{40}K: $(42\pm1)\cdot10^{-3}$; ^{137}Cs: $(1.1\pm0.2)\cdot10^{-3}$	^{228}Ra: $0.5\cdot10^{-3}$, ^{228}Th: $0.3\cdot10^{-3}$
Radiopurity study	Stainless steel screws (M3)	75	^{60}Co: $(11\pm2)\cdot10^{-3}$; ^{54}Mn: $(1.3\pm0.3)\cdot10^{-3}$	^{40}K: $25\cdot10^{-3}$, ^{228}Th: $3\cdot10^{-3}$, ^{228}Ra: $4\cdot10^{-3}$, ^{226}Ra: $4\cdot10^{-3}$, ^{238}U: $20\cdot10^{-3}$

[a] Other radionuclides were detected but not quantified since it was outside the scope of the investigation

Method validation: For IRMM as well as for other reference institutes it is essential to have alternative analytical techniques in order to resolve issues or disputes relating to measurements. In order to understand discrepancies with determining trace amounts of Zn in GaAs wafers using GDMS (Glow Discharge Mass Spectrometry) and ERS (Electron Raman Scattering) IRMM used a combination of neutron activation, chemical separation and ULGS.[8] This technique gave a detection limit of about 20 pg/g, which was 2 orders of magnitude lower than GDMS. IRMM could confirm that in this special case GDMS gave correct results and encountered problems were due to the Zn concentration for some samples being below decision threshold for GDMS and ERS.

In Table 2 are also included measurement results from analysis of human freeze-dried faeces. The measurements were conducted for the German Federal Office of Radiation Protection with the aim of validating a method based on radiochemical separation and mass spectrometry to determine ^{226}Ra and other NORMs (naturally occurring radioactive materials) in human urine and faeces. The necessary detection limits of 5 mBq/g were difficult to obtain with gamma-ray spectrometry above ground.

Radiopurity studies: In order to benefit from the reduction of the background induced by cosmic rays in underground laboratories, it is essential that all materials used for constructing a detector and its shield are radiopure. IRMM has contributed to radiopurity studies for large scale detectors for rare events like the BOREXINO neutrino detector and the GERDA detector for double beta decay. In Table 2 are presented some data for materials that were measured in HADES in order to determine if they could be used in HPGe-detectors for underground use. It is important to note that the radioactivity levels of common construction materials can vary from one batch to another so one can not generalise the numbers given in Table 2.

Activation: There are good reasons to say that activated samples are most suitable samples for measurement using ULGS. The reason for this is that very often there is no interference from other radionuclides that increase the detection limit for the

radionuclide of interest. This fortunate situation is either due to the fact that the activated material was selectively chosen beforehand, like in the case of flux monitors in nuclear reactors, or afterwards like in the case of environmental sampling after an incident involving neutron irradiation outside a controlled area.[10]

A recent example of ULGS measurements of activated samples involve solving discrepancies between model calculations and measurements of activation induced by the A-bomb in 1945. Komura et al. could solve the problem with ^{152}Eu in granite by using the Ogoya underground laboratory[28] and Hult et al. showed that older measurements of ^{60}Co in steel could be significantly too high.[29]

Conclusions

In order to obtain low detection limits in deep and semi-deep underground gamma-ray spectrometry (and thus benefit from the reduced cosmic ray flux) it is necessary to have radiopure detectors and shields. By careful selection of materials it is possible to lower the background count rate almost by a factor of 10 in deep underground laboratories compared to using "standard" ultra low background HPGe-detectors. It is likely that some of the new development including using underground production and storage of Ge-crystals will drive the technology further forward, which will be beneficial for deep underground laboratories. For special applications it is possible to use special coincidence techniques but for the HPGe-detectors acting as all-purpose work horses it is important to keep the system simple in order to minimise the labour cost for analyses. With the large crystals available today, the main cost is the raw material (the Ge-crystal). Although the cost of a large Ge-detector is high it should be possible to operate it for many years (10-25 years) with proper maintenance. It makes good sense to keep an underground detector in operation as long as possible since the background should decrease with time as activation products including ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn will decay significantly and even the decay of ^{210}Pb ($t_{½}$=22.2 y) should be noticed provided plate-out of ^{222}Rn-daughters on the detector can be kept low enough. It has been seen in the past 10 years that new interesting applications making use of ULGS turn up. It is likely that this trend will continue. The future of the technique is very much linked to the price of Ge as well as the easy of finding suitable underground laboratories.

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Povzetek

Spektrometrija gama na osnovi germanijevih detektorjev visoke čistosti je v zadnjih letih pomembno napredovala zaradi razpoložljivosti velikih kristalov in razumevanja pomena uporabe radiološko čistih snovi pri izdelavi polprevodniških detektorjev. Kombinacija omenjenih izboljšav je omogočila znižanje meja detekcije v posebnih števcih za meritve z nizkim ozadjem. To se še posebej odraža pri sistemih za spektrometrijo gama postavljenih pod zemeljsko površino. Ta prispevek predstavlja najnovejšo tehnologijo za podzemno spektrometrijo gama in podaja primere novih aplikacij te tehnike.