Highly Sensitive Gamma-Spectrometers of GERDA for Material Screening: Part 2

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September 20, 2007

Abstract

The previous article about material screening for GERDA points out the importance of strict material screening and selection for radioimpurities as a key to meet the aspired background levels of the GERDA experiment. This is directly done using low-level gamma-spectroscopy. In order to provide sufficient selective power in the mBq/kg range and below, the employed gamma-spectrometers themselves have to meet strict material requirements, and make use of an elaborate shielding system.

This article gives an account of the setup of two such spectrometers. CORRADO is located in a depth of 15 m w.e. at the MPI-K in Heidelberg (Germany), GEMPI III is situated at the Gran-Sasso underground laboratory at 3500 m w.e. (Italy). The latter one aims at detecting sample activities of the order $\sim$10 μBq/kg, which is the current state-of-the-art level. The applied techniques to meet the respective needs are discussed and demonstrated by experimental results.

1 Introduction

The previous article [1] points out the relevance of various material screening procedures employed for the material selection in scope of the GERDA experiment [2]. The most direct technique for the detection of gamma-active radioimpurities in the construction and shielding materials is the low-level gamma-spectroscopy. Like GERDA it is based upon the operation of high purity germanium detectors (HPGe)[3], and hence is sensitive to the same sources of background, out of which gamma-active isotopes with Q-values above $Q_{\beta\beta}$ (2039 keV for ^{76}Ge) are of special concern.

In order to meet the aspired background index of $<10^{-2}$cts/(keV·kg·y) at $Q_{\beta\beta}$ in Phase I, and $<10^{-3}$cts/(keV·kg·y) in Phase II, sensitivities in material screening ranging from mBq/kg down to some 10 μBq/kg are required. The latter is the current state-of-the-art level accomplished by the GEMPI-spectrometer [4, 5] located in the Low Level Research Facility (LLRF) in the Gran Sasso underground laboratory at 3500 m w.e.

Apart from reaching the highest possible sensitivities of the γ-spectrometers, it is necessary to provide sufficient screening capacity to cope with the amount of samples to be screened. Since the measurement time for best sensitivity with GEMPI can reach up to several months for one sample, we seek to build new γ-spectrometers in different depths and sensitivities.

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The CORRADO-spectrometer has been set up in the Low Level Laboratory (LLL) at 15 m w.e. at the MPI-K Heidelberg (Germany) as a follow-up to DARIO [6, 8]. It can reach a sensitivity of ∼1 mBq/kg corresponding to 10^{-10} g/g for uranium and thorium, and is designed for large samples, partly for their preselection. GEMPI III is successor of GEMPI at LLRF, aiming for a sensitivity of 10 μBq/kg corresponding to 10^{-12} g/g for uranium and thorium. Both spectrometers' design has been improved with respect to their precurser instruments. Their shielding systems suit the demands of their depth, and are described in the subsequent sections. While CORRADO is operating and first data can be presented, GEMPI III suffers a ^{207}Bi contamination, which is discussed in section 4.2.

2 Basic Design Criteria

The central task in the construction of a low-level γ-spectrometer is to minimize the lowest detectable specific activity A_{min} in a certain counting time t, as a measure of it's sensitivity:

$$A_{min} = \frac{\sqrt{B}}{M \cdot \epsilon \cdot t} \tag{1}$$

This equation holds for the simple case that only one γ-line of a given isotope is taken into account for the evaluation, and the background from the line is negligible. B denotes the background counts in the continuum, M the sample mass and ϵ the full energy peak efficiency associated with the sample.

Minimizing A_{min} means to maximize the potential signal count rate of the sample, represented by the product $M \cdot \epsilon$. Therefore a large sample mass M in a geometry preferably encasing the detector for best efficiency is needed. A large crystal also leads to a higher efficiency. However, the product $M \cdot \epsilon$ only increases until self-absorption within the sample becomes dominant. An optimal sample size can be estimated from the 1/e thickness for different high density sample material and the highest energy of interest, in this case the 2615 keV line of ^{208}Tl (see [4] for more detail). This results in the choice of a Marinelli type geometry for the sample chamber with a volume of about 18 l.

The main room for improvement now remains in the reduction of the background, which enters (1) as $\sqrt{B}$. Apart from the continuos background B, which can be determined from the sample spectrum itself, the line background has to be derived from a separate background spectrum. Its effective rate must be quantified with regard to the absorption and radon displacement caused by the sample, which may lead to additional uncertainty [9]. Thus it is essential to have low line background rates and a good control over the chambers radon content. For a detailed discussion of background sources and suppression techniques see [10] and [11].

3 The CORRADO-Spectrometer

The CORRADO-spectrometer is situated in the Low Level Laboratory at MPI-K at the shallow depth of 15 m w.e. At this depth the nucleonic component of the cosmic rays virtually vanishes and the muon flux is reduced by a factor of ∼3 [12]. The construction was completed in March 2007, and testing and background measurements have been done. The detector's shielding system is shown in figure 1.

The spectrometer is placed on a solid steel table with the detector's dewar underneath. The HPGe crystal (coaxial, p-type, mass 930 g[1]) is mounted in a cryostat made out of carefully selected material. It has an aluminum end cap, and it's elbow shaped copper neck is designed to avoid direct line of sight to external gamma rays. In order to improve the Monte Carlo

[1] corresponding to a relative efficiency of 37%

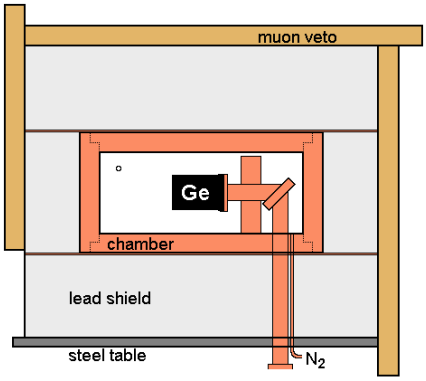

Figure 1: Cross-section of the CORRADO-spectrometer.

model of the cryostat and crystal several x-ray images were taken before the insertion into the shielding.

The sample chamber is made out of 5 cm thick electrolyte copper. It allows for samples in Marinelli geometry with the dimensions 25×25×33 cm^3. The chamber can be evacuated and flushed with nitrogen. During operation it is constantly flushed by the boil-off nitrogen from a dewar to ensure a low and stable ^{222}Rn level in the chamber. Outside the chamber there is an additional passive shield made of 15 to 20 cm low activity lead. The innermost layer partly consits of lead from the Polish company Plombum with a ^{210}Pb contamination of 5 Bq/kg [5]. A thicker shield is not reasonable since it would lead to a higher neutron production due to cosmic rays [11]. All lead bricks and copper parts of the shielding went through a thorough etching procedure to remove surface contamination. The full passive shield and flushing system supresses the background due to environmental and airborne radioactivity by a factor ∼100 – see figure 4. The shielding can be opened from the frontside to insert samples.

CORRADO is equipped with five multiwire proportional chambers operated in anticoincidence with the Ge-diode as a muon veto. With a dead time of 50 μs/signal it is set to reject background from muon induced bremsstrahlung and short lived muon-activated isotopes. A suppression factor of ∼10 (88%) has been determined from the difference of the vetoed and the unvetoed spectra (figure 4) [13]. This is a preliminary result and we seek to approach the high suppression (>93%) of DARIO [6]. Alltogether a background suppression of three orders of magnitude compared to the unshielded detector in the LLL has been achieved.

The background spectrum with full shield reveals one background line from ^{40}K with (1.45± 0.80) cts/day. All other isotopes lie below the detection limit – results are given in table 1. A long-term measurement with several weeks of counting time is scheduled as soon as the ongoing renovation of the LLL is completed. The aspired sensitivity of ∼1 mBq/kg has been achieved for ^{228}Th with a Makrolon®[2] sample of 13 kg and only 2.5 days of counting time – see table 2.

[2] a polycarbonate from Bayer MaterialScience

Energy	Isotope	Counts per day
100-2700 keV		4609 ± 17
609 keV	^{214}Bi	<1.9
1333 keV	^{60}Co	<2.1
1461 keV	^{40}K	1.45 ± 0.80
2615 keV	^{208}Tl	<1.3

Table 1: CORRADO-background countrates based on 16.7 days of counting. Upper limits are given with 90% confidence level.

Long lived isotope	Specific activity [mBq/kg]
^{226}Ra	<2.38
^{228}Ra	<4.78
^{40}K	<24.99
^{228}Th	<1.14

Table 2: Makrolon® sample of 13 kg after 2.5 days counting in CORRADO. Upper limits are given with 90% confidence level.

4 The GeMPI III-Spectrometer

4.1 The Design

GeMPI III is located in the LLRF at the LNGS with an overburden of 3500 m w.e. At this depth the muon flux is reduced by six orders of magnitude to approximately 1 μ/m^2h. This makes an active muon shield dispensable. The assembly of the shielding was completed in January 2007, but a subsequently discovered ^{207}Bi contamination postpones it's operation until today. Figure 2 shows a view of the spectrometer's shielding system with the detector inside.

The cryostat is custom made from electro-refined copper and other carefully selected and screened material. All materials were chosen to minimize bulk contamination and to avoid high neutron activation cross sections. It was stored underground between fabrication and cleaning processes to reduce activation by cosmic rays. The detector is a coaxial, p-type HPGe-crystal with a mass of 2.3 kg. The conception and assembly of the cryostat system was undertaken in close collaboration with Canberra Semiconductors N.V., Olen, Belgium.

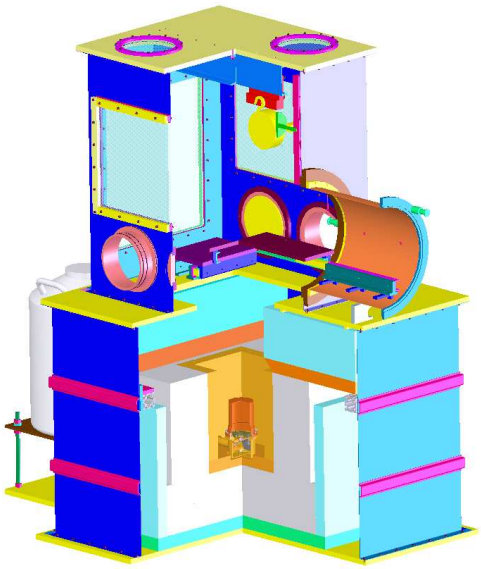

Figure 2: Cutaway view of the GeMPI III shielding and radon protection system.

The sample chamber consists of 5 cm of NOSV copper [7] with the inner dimensions 25×25×30 cm^3. It is surrounded by 20 cm of low activity lead. The innermost 5 to 10 cm consist of lead from the Polish company Plombom (see section 1). The shielding on top sits on rollers and may be moved aside to have access to the chamber from above. Like for CORRADO all lead and copper parts went through an etching procedure to remove surface contamination. The shield is completed by 5 cm thick slabs of borated (7%) polyethylene from the bottom and the four sides.

The setup is enclosed in a two-part air-tight stainless steel casing for radon protection, which is constantly flushed at slight overpressure with boil-off nitrogen. The bottom part envelopes the shielding, whereas there is a sample storage and handling compartment on top. It has acrylic windows, glove openings from two sides, an airlock system for the insertion of samples, and a movable table to transfer heavy loads. Therewith it is possible to permanently operate the spectrometer without exposing the inner volume to air. A space in the upper compartment is destined for the storage of samples prior to their measurement, to allow for the decay of ^{222}Rn/^{220}Rn and their progenies. The entire assembly of the GeMPI III-setup was done under cleanroom conditions.

4.2 Investigation on a ^{207}Bi contamination

After comissioning of the spectrometer it turned out that it suffers a ^{207}Bi contamination. The two main lines are visible with (37.8±1.6) cts/d for 570 keV and (21.2±1.2) cts/d for 1064 keV, as well as the summation peak with (3.0±0.5) cts/d – see figure 3. The occurrence of the latter indicates a close proximity of the contamination to the crystal. This is confirmed by measurements with additional shielding within the sample chamber, leaving the denoted countrates unchanged. The line ratio of the count rates for the 570 keV and 1064 keV line allows to conclude that only minor attenuation takes place between source and detector. The absence of conversion electrons in the spectrum excludes a contamination inside the crystal's borehole. Further investigation employing a summation peak analysis via MC simulations to determine the distance towards the crystal is pursued [14]. The activity was determined to be $A(^{207}Bi) = (3.74 \pm 0.39)$ mBq.

^{207}Bi has no natural abundance, so possible candidates for the contamination are cross-contamination with a source and a ^{207}Bi-production via (p,n)-reaction on ^{207}Pb. The latter was ruled out by the above analysis and a cross check by screening the spare lead parts. After a long forensic struggle the origin of the contamination was identfied as a cross contamination from a hydrochloric acid solution containing ^{207}Bi. It was transferred via tweezers from an allegedly clean toolbox during the cryostat-mounting, transfusing approximately 4 μg of solution equivalent. The origin of the ^{207}Bi-source lies outside of Canberra's and our reference.

A similar ^{207}Bi contamination has been discovered on the GeMPI IV-cryostat in the course of this investigation. However, it is lower by a factor of ten. A thorough etching attempt for the cryostat and methanol baths for the crystal were performed and tested at the end of July 2007, with the results still to come.

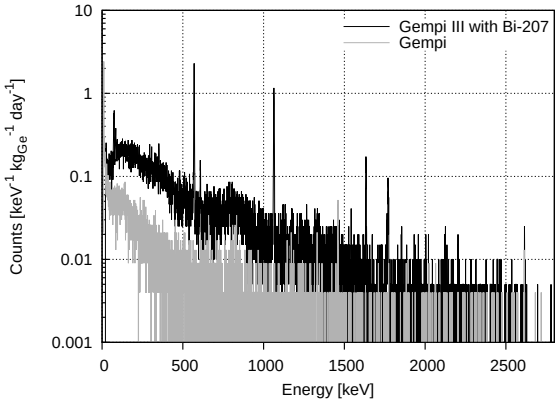

Figure 3: Background spectrum of GeMPI III (black) with ^{207}Bi-contamination and provisional nitrogen flushing in comparison to the GeMPI-spectrometer.

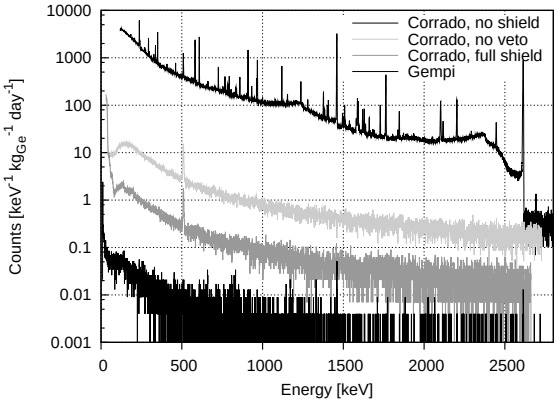

Figure 4: Background spectra of CORRADO at MPI-K Heidelberg with different shielding configurations in comparison to GeMPI at Gran Sasso.

5 Conclusion

The CORRADO-spectrometer shows, that a background supression by a factor ∼1000 is feasible in shallow depth. Apart from a radiopure cryostat and passive shielding, an active muon veto and radon protection of the sample chamber by constant nitrogen flushing is needed. Further improvement of the muon veto is intended. The spectrometer is designed for large samples and has succeeded to meet the aspired sensitivity of ∼1 mBq/kg for ^{228}Th. At this level it is suitable for material screening within GERDA.

The GeMPI III-spectrometer failed to reach the sensitivity of GeMPI, which is the most sensitive γ-spectrometer available for routine material screening. This is due to a ^{207}Bi contamination of (3.74±0.39) mBq within the cryostat of the detector. A variety of approaches are taken to confine the possible locations, and a cleaning operation has been carried out and tested on the GeMPI IV-detector.

Acknowledgements

The technical staff of the MPI workshops is thanked for the skillfull construction of the GeMPI III-cryostat and their enduring commitment in realising the shielding systems of both spectrometers. U. Schwan is thanked for her perpetual support and counsel in surface etching and the contamination issue. We greatly acknowledge the very good cooperation with Canberra Semiconductors N.V., Olen, Belgium in the design and assembly of the GeMPI III-cryostat.

This work was partly supported by INTAS (Project Nr. 05-1000008-7996) and by the DFG within the SFB Transregio 27 "Neutrinos and Beyond".

References

[1] D. Budjáš et al., Highly Sensitive Gamma-Spectrometers of GERDA for Material Screening: Part 1, *these proceedings*

[2] GERDA Collaboration, I. Abt et al., GERDA: The GERmanium Detector Array for the search of neutrinoless $\beta\beta$ decays of ^{76}Ge at LNGS, *Proposal (2004), http://www.mpi-hd.mpg.de/GERDA/proposal.pdf*

[3] M. Hult, Low-level gamma-ray spectrometry using Ge-detectors, *Metrologia 44 (2007) S87S94*

[4] G. Heusser, M. Laubenstein, H. Neder, Low-level germanium γ-ray spectrometry at the Bq/kg level and future developments towards higher sensitivity, *Proc. of Intern. Conf. Isotop. Environm. Studies Aquatic Forum 2004, 25-29 October 2004, Monte-Carlo, Monaco (http://edoc.mpg.de/220964).*

[5] H. Neder, G. Heusser, M. Laubenstein, Low-level germanium-spectrometer to measure very low primordial radionuclide concentrations, *Appl. Rad. Isot. 53 (2000) 191-195*

[6] G. Heusser, Studies of γ-ray background with a low level germanium spectrometer, *Nucl. Instr. an Meth. B 58 (1991) 79.*

[7] Grade NOSV Copper, provided by Norddeutsche Affinerie AG, *http://www.norddeutsche-affinerie.de/NA_en/rohstoffe/kupferprodukte/stranggussformate/roh_werkstoffe*

[8] G. Heusser, *Proc. 14th Europhys. Conf. on Nuclear Physics, Bratislava, 1990, Rare Nuclear Processes, ed. P. Povinec (World Scientific, 1992) 247.*

[9] M. Heisel, Low-Level γ-Spektrometrie an verschiedenen Materialien für GERDA (german language), *Diploma thesis, University of Heidelberg (2006)*

[10] G. Heusser, Low-Radioactivity Background Techniques, *Annu. Rev. Nucl. Part. Sci. 1995. 45:543-90.*

[11] G. Heusser, Background in Ionizing Radiation Detection – Illustrated by Ge-Spectrometry, *Proc. 3rd Int. Summer School on Low-Level Measurements of Radioactivity in the Environment: Techniques and Applications, Huelva, Spain 1993.*

[12] G. Heusser, Cosmic ray-induced background in Ge-spectrometry, *Nucl. Instr. and Meth. B 83 (1993) 223-228.*

[13] W. Maneschg, Low-Level-Gammaspektroskopie im Rahmen des GERDA-Experiments zur Suche nach dem neutrinolosen Doppel-Betazerfall in ^{76}Ge (german language), *Diploma thesis, University of Heidelberg (2007).*

[14] O. Chkvorets, S. Schönert, Localization of radioactive contaminations in low-background HP-germanium detectors, *in preparation*