GAMMA-RAY SPECTROMETRY OF ULTRA LOW LEVELS OF RADIOACTIVITY WITHIN THE MATERIAL SCREENING PROGRAM FOR THE GERDA EXPERIMENT

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Abstract

In present and future experiments in the field of rare events physics a background index of 10^{-3} counts/(keV kg a) or better in the region of interest is envisaged. A thorough material screening is mandatory in order to achieve this goal. The results of a systematic study of radioactive trace impurities in selected materials using ultra low-level gamma-ray spectrometry in the framework of the GERDA experiment are reported.

Keywords: double beta decay, natural radioactivity, ultra low-level gamma-ray spectroscopy

1 Introduction

GERDA, the GErmanium Detector Array, is a calorimetric experiment using enriched germanium (the active isotope being ^{76}Ge, enriched to 86%) (Abt et al., 2004). It is crucial to keep strict control of the radioactive background. For this a background index b is defined as the number of counts per unit detector mass, unit exposure time and unit energy interval. Considering that the neutrinoless double beta decay of ^{76}Ge has a monochromatic line in the summed electron energy spectrum at a very precisely known energy, $Q_{\beta\beta}$=2039 keV. The condition that the background has to be below b must be satisfied only in the signal region within a width of approximately 10 keV, which is determined by the detector energy resolution. In particular, germanium used as detector and source contemporaneously, has an excellent energy resolution, and can be produced industrially with an extreme radiopurity.

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GERDA is currently under construction in the underground laboratories of the Laboratori Nazionali del Gran Sasso (LNGS). The GERDA experiment will use bare Ge crystals immersed in liquid Ar. The experimental set-up is described for example in (Bettini, 2007). In its phase-I GERDA will use the enriched Ge-diodes employed in the previous double beta experiments HdM (Klapdor-Kleingrothaus et al., 2004) and IGEX (Aalseth et al., 2002). Their total mass is about 18 kg. In phase-II about 20 kg of either new custom-made segmented true coaxial n-type Ge-detectors or unsegmented broad energy p-type Ge-detectors will be deployed.

In phase-I, the background will be mainly dominated by the intrinsic cosmogenic ^{60}Co of the existing Ge-diodes which will contribute with about 10^{-2} counts/(keV kg a). However, this level should be sufficient to confirm or refute the evidence of neutrinoless double beta decay in ^{76}Ge reported in (Klapdor-Kleingrothaus et al., 2004) with an exposure of one year.

In order to reach the envisaged background index $b=10^{-3}$ counts/(keV kg a), no component of the background budget must be larger than $b=10^{-4}$ counts/(keV kg a). For this reason GERDA has set up a radiopurity screening program for all materials and components (e.g. suspension system, front-end electronics, cabling), measuring samples at different sites, mainly by γ-spectroscopy, by radon emanation measurements and by inductively coupled plasma mass spectrometry. Extremely high radiopurity standards must be met. Moreover, analytical methods or Monte Carlo simulations estimate all background sources in every component of the apparatus. For this reason the MAJORANA and GERDA Collaborations, where MAJORANA represents a similar experiment as GERDA located in the U.S.A., have developed together the MaGe frame and database (Bauer et al., 2006; Chan et al., 2008), that provide the allowed specific activity for each component.

In this paper the analytical procedures and methods applied for gamma-ray spectroscopy by the GERDA collaboration will be specifically described. After a discussion of the radiopurity requirements for GERDA, the screening facilities involved will be introduced briefly and then the results obtained with low-level germanium gamma-ray spectrometry will be presented.

2 Radiopurity requirements

The most important sources of contamination for GERDA can be classified according to their origin:

- primordial radionuclides (^{238}U, ^{232}Th (and their decay chains) and ^{40}K);

- cosmogenic radionuclides, such as e.g. ^{60}Co, ^{68}Ge;
- anthropogenic radionuclides, such as ^{60}Co, ^{137}Cs.

Due to their environmental occurrence and activity, the primordial radioisotopes are generally the most important contaminants.

Table 1 lists the design goals for the radiopurity concentrations allowed for the major components of the detector. They have been determined either via analytical estimates or using the MaGe Monte Carlo simulation package. As can be seen, the tolerable activity concentrations depend on the position of the material and on the variation of the mass fractions of the components. For simplicity in the notation, the ^{238}U and ^{232}Th specific activities given in Table 1 assume secular equilibrium. For ^{232}Th the specific activity limits have been obtained mostly by looking exclusively at the ^{208}Tl 2615 keV gamma-line. In several cases only the limit on ^{232}Th is given, as the ^{238}U concentration is less critical, because its contribution to the region of interest around 2039 keV is smaller, due to the lower branching ratios of the "dangerous" gamma emissions with respect to the aforementioned ^{208}Tl. In general in these cases one can assume that if for ^{238}U the same specific activity limit is met, as for ^{232}Th, then the aim is reached.

Most modern methods of material processing will disturb the secular equilibrium in the decay chains if the chemistry of the elements is different as far as the processing is concerned. Thus, for the cases of the natural decay chains, information on deviations from the secular equilibrium is important. This is especially true for the cases in which in some sub-series gamma-emitting nuclides are dominant, while the gamma-intensity for the first member of the series is low. There is no experimental analytical method that provides by itself complete information for the background contribution from the entire decay chain.

3 Low-level and ultra low-level Germanium gamma-ray facilities in GERDA

Almost all solid detector materials, beginning with the concrete components for the base of the 10 m diameter SS water tank to the Cu parts used for the suspension system of the Ge-diodes have been or are planned to be screened by gamma-ray spectrometry.

Ultra low-level Ge-spectrometry can reach sensitivities to levels in the μBq range for kilogram sized samples, which corresponds to 10^{-12} g g^{-1} levels of U and Th contamination (Hult et al., 2006). This level of sensitivity is sufficient to control materials or components close to the inner detector (see Table 1). Since the detector itself consists of Ge-diodes the tolerable contamination level is defined by the gamma-activity and Ge-detectors are therefore ideal for doing the screening. A further advantage is that most measurements can be performed in a non-destructive way without a laborious sample pre-treatment.

The most abundant contaminants, ^{40}K and a large part of the U- and Th-decay chains are detectable by their emitted γ-rays. Within certain limits, deviations from secular equilibrium in the U and Th series can also be observed. Most important, this is possible in the case of the ^{232}Th-^{228}Ra-^{228}Th sub-series, where the time for noticeable changes in the activity ratios (and re-establishment of equilibrium) is well within the range of the lifetime of the GERDA experiment. This piece of information is important in the light of the fact that GERDA aims to maintain the background index at the design level for the whole duration of the experiment. For the determinations of the U and Th chain activities, the highest sensitivity is obtained using the sub-chains starting with ^{226}Ra (U) and ^{228}Th (Th) due to the high gamma-ray abundance in the decay of their daughters. For the case of solid samples, which are not very fine grained (in which case ^{222}Rn cannot diffuse out), equilibrium within the aforementioned subchains can be assumed.

Low-level and ultra low-level Ge-spectrometry for GERDA is performed at five different laboratories: the LNGS (Italy), the Institute for Reference Materials and Measurements (IRMM; HADES - High Activity Disposal Experimental Site, Belgium), the Max-Planck-Institut für Kernphysik (MPIK) in Heidelberg (Germany), the Joint Institute for

Nuclear Research (JINR) in Dubna (Russia) and the Baksan Neutrino Observatory (BNO) run by the Institute for Nuclear Research of the Russian Academy of Sciences (INR RAS) (Russia). The reason why so many laboratories are involved in the material screening for GERDA is that the measurements at such low levels of radioactivity concentrations are very time consuming (see e.g. in table 2 the measurement for PTFE and PEN). Moreover one can also do in cases, eg. when doubts may have occurred, an intercomparison between the results of two different laboratories in order to be confident that the final result obtained is correct.

A summary of all labs and their most important properties can be found in Budjáš (2008). It has to be noted that the IRMM was supported by the ILIAS (Integrated Large Infrastructure for Astroparticle Science) project, financed by the European Commission within the 6th framework program. Part of the screening measurements has been done within the Joint Research Activity IDEA (Integrated Double-beta decay European Activities).

4 Data analysis and results

Laubenstein et al. (2004), Heusser et al. (2006) and Maneschg et al. (2008) have already published results of gamma-ray screening measurements for GERDA for some selected materials. In terms of radiopurity, they represent some of the cleanest metals and plastics ever measured.

4.1. Data evaluation

Each laboratory uses its own spectrum analyzing program, tuned to low count rates. The counting efficiency is determined from either calibrated geometry or by using Monte Carlo simulations. The efficiency determination is based on measuring activity standards. Budjáš (2007) has published an intercomparison study within the GERDA collaboration, discussing in detail the evaluation accuracy of each lab.

4.2 Sample preparation

Prior to the measurement, the samples are cleaned. The protocols used differ according to the material of the sample. Sometimes an acid treatment is mandatory in order to remove surface contamination, in other cases a plain washing with soap and thorough rinsing with ultra-pure water afterwards is sufficient. Nevertheless, the first few hours, sometimes even a few days, of a measurement are not taken into account because of the presence of ^{222}Rn and ^{220}Rn daughter nuclides that have plated out onto the components before introduction to the detector system.

4.3 Screening results

A compilation of selected results obtained by the GERDA screening laboratories is illustrated in table 2. Only data concerning materials of a more general importance or wider spread interest are reported here. The ^{226}Ra-activity concentration is usually determined using the gamma-ray lines of the ^{222}Rn daughters ^{214}Pb and ^{214}Bi. Care has to be taken that no loss of ^{222}Rn occurred and that the radon gas is in equilibrium with its daughters. The activity concentration of ^{228}Ra is determined via its daughter product ^{228}Ac, whereas that of ^{228}Th by the gamma ray lines of ^{212}Pb, ^{212}Bi and ^{208}Tl.

The quoted uncertainties are combined standard uncertainties determined according to (ISO, 1995). Due to the lack of sufficient standardization for determining an upper limit different methodologies have been chosen in the various laboratories in order to estimate the upper limit in case of a null result (result below decision threshold). In all cases a coverage factor of k=1.645 is used.

The sensitivity that has been achieved in each measurement depends not only on the efficiency and the background of the spectrometer in use but also on the measuring time and the amount (mass) of the sample. This is clearly seen in the results of the PTFE measurements. The lowest concentration for U, ^{226}Ra and Th was obtained for a 28.06 kg sample measured with GeMPI for ca. 105 days.

Of general interest are the results obtained for plastic materials that are commonly used as support for printed circuits. The best choice seem to be PTFE (see results for pure PTFE and for Cuflon®) and PEN, as they are more radiopure than Kapton® HN. This means that in the design the solution of Kapton® and Cu must be changed e.g. to PEN and Cu.

Another interesting result is the measurement of the PE (Murtfeldt), which demonstrates that this material can be used to substitute PTFE in the Ge crystal holders. Although it is ten times worse in radiopurity it is still acceptable. Having a smaller density than PTFE it would also reduce the mass of material close to the Ge-detectors.

Nomex® and steel cables have been measured in order to have an alternative to copper as material for the support strings. As can be seen Nomex® is not indicated as substitute, because it does not fulfill the requirements. Steel, instead, would be a possible candidate.

The measured coax cables are fine with the requirements. The pogo pins are too high by a factor of ten with respect to the very stringent requirement reported in table 1. In fact, the search for alternatives is still ongoing. The superinsulation foil, which is the one that was eventually used in the construction of the inner cryostat, is complying with the specifications, too.

Also the IGLIDUR plastic is fine, as it is sitting further away from the Ge-crystals close to the top of the water tank and its radiopurity is within the requirements for the stainless steel of the water tank itself.

5 Conclusions

The needs of the GERDA double beta decay experiment require the knowledge of ultra-low levels of radioactivity. In this paper, we presented the results of specific measurements on a number of materials of importance not only to the GERDA experiment but also to other experiments in rare event research. In addition we described our own techniques and instrumental developments. Here we summarize the main points. We operate a number of underground Ge-detectors, whose backgrounds are among the lowest achieved so far. Such detectors allowed the measurement from 10^{-9} to 10^{-12} g g^{-1} of ^{238}U, ^{232}Th, ^{226}Ra and few 10^{-9} g g^{-1} of K as lowest radioactive contents in various construction materials.

The techniques and results reported in this paper fulfill the needs of GERDA. We believe that the measured activity levels of the materials listed in the tables will also be useful for the design of future activities in rare events physics and maybe to other fields where extremely low background is needed.

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Table 1: The maximum allowable activities for some natural and man-made radionuclides of the major GERDA detector components that guarantee for each single part listed a certain background index for phase-2 with liquid Ar. For the parts listed in the rows 2 to 6 the maximum allowable activity corresponds to a background index of $b=10^{-4}$ counts kg^{-1} keV^{-1} a^{-1}, for the part listed in row 7 to $b=10^{-5}$ counts kg^{-1} keV^{-1} a^{-1}. and for all the others to $b=10^{-3}$ counts kg^{-1} keV^{-1} a^{-1}. The numbers have been obtained by using both the MaGe simulation tool and analytical estimates. The maximum activities given for ^{40}K and ^{137}Cs are calculated assuming that the contribution in the region of interest is < 1% of the contribution due to the double beta decay with neutrino emission.

Part	mass [kg]	^{238}U [$\mu Bq\ kg^{-1}$]	^{232}Th [$\mu Bq\ kg^{-1}$]	^{40}K [$\mu Bq\ kg^{-1}$]	^{137}Cs [$\mu Bq\ kg^{-1}$]
water (external shield)	622×10^3		$< 4 \times 10^4$		
cryostat stainless steel	30×10^3		$< 5 \times 10^2$		
cryostat inner Cu liner	14×10^3		< 100		
cryostat superinsulation	80		$< 5 \times 10^5$		
signal and HV cables, Kapton and Cu	2.79		$< 1.9 \times 10^3$		
pogo pins	0.02058		$< 1.7 \times 10^3$		
Argon	88.6×10^3	< 1.8	< 0.4		
Ge-detector electronics	0.7	$< 8 \times 10^3$	< 460	$< 5 \times 10^3$	$< 5.4 \times 10^5$
Ge detector holders Cu	0.651	< 430	< 200	< 930	
Ge detector holders PTFE	0.147	$< 1.9 \times 10^3$	$< 1.4 \times 10^3$	$< 4 \times 10^3$	< 580
cable 1 on Ge detector holder, Kapton and Cu	0.0191	$< 1.8 \times 10^3$	$< 2.0 \times 10^3$	$< 4.0 \times 10^4$	$< 1.5 \times 10^4$
cable 2 on Ge detector holder, Kapton and Cu	6.8×10^{-3}	$< 2.0 \times 10^3$	$< 3.5 \times 10^3$	$< 5.5 \times 10^4$	$< 2.6 \times 10^4$
cable 3 on Ge detector holder, Kapton and Cu	1.3×10^{-3}	$< 9.4 \times 10^3$	$< 1.3 \times 10^4$	$< 2.6 \times 10^5$	$< 1.3 \times 10^5$
bond pad, Ni part	8.4×10^{-4}	$< 1.8 \times 10^4$	$< 4.7 \times 10^4$		
bond pad, Au part	1.2×10^{-5}	$< 3.3 \times 10^5$	$< 1.7 \times 10^6$		
bond wire, Al	1.7×10^{-6}	$< 2 \times 10^7$	$< 5 \times 10^7$		
Cu strings	0.140	$< 1 \times 10^4$	$< 1 \times 10^4$		
Ge crystals	44.2	< 0.4	< 1.9		

Table 2

A compilation of selected results obtained by the GERDA screening laboratories. The quoted uncertainties are combined standard uncertainties (ISO, 1995), the upper limits are given with a coverage factor of k=1.645 .

material	mass [kg]	measuring live time [d]	^{238}U [Bq kg^{-1}]	^{226}Ra [Bq kg^{-1}]	^{228}Ra [Bq kg^{-1}]	^{228}Th [Bq kg^{-1}]	^{40}K [Bq kg^{-1}]	other radionuclides [Bq kg^{-1}]
coax cable (Cu/PTFE, Habia)(a)	3.1583	6.87	$< 5.9\times10^{-2}$	$< 1.4\times10^{-3}$	$< 2.5\times10^{-3}$	$(1.1 \pm 0.5)\times10^{-3}$	$(4.0 \pm 0.4)\times10^{-1}$	^{110m}Ag: $(1.3 \pm 0.3)\times10^{-3}$ ^{108m}Ag: $(7.8 \pm 2.4)\times10^{-4}$
Nomex® 464 yarn (DuPont™) (b)	0.167	21.8	–	$< 2.5\times10^{-2}$	$(8.7 \pm 2.7)\times10^{-3}$	$(11.1 \pm 1.1)\times10^{-2}$	$(9.5 \pm 0.4)\times10^{-1}$	^{137}Cs: $(4.3 \pm 1.3)\times10^{-3}$
steel cable for suspension(b)	2.29	12.75	–	$(3.1 \pm 1.5)\times10^{-3}$	$(4.4 \pm 1.9)\times10^{-3}$	$(11.8 \pm 1.1)\times10^{-3}$	$<6\times10^{-3}$	^{60}Co: $(19.2 \pm 1.0)\times10^{-2}$
pogo pins(b)	0.019	33.6	–	$(5.8 \pm 1.5)\times10^{-2}$	$(2.2 \pm 0.5)\times10^{-2}$	$(4.0 \pm 1.0)\times10^{-2}$	$(3.8 \pm 0.3)\times10^{-1}$	^{210}Pb: (1.4 ± 0.1)
SI foil (Jehier Insulray IR 305) (c)	0.50	12.68	–	$(2.3 \pm 0.3)\times10^{-1}$	$< 9.7\times10^{-2}$	$< 5.2\times10^{-2}$	(1.94 ± 0.95)	–
PEN foil (TEONEX®, DuPont™)(d)	2.05	29.79	–	$< 2.0\times10^{-3}$	$< 1.4\times10^{-3}$	$< 1.4\times10^{-3}$	$< 3.6\times10^{-3}$	–
Murtfeldt UHMW-PE (a)	6.99	24.45	$< 3.4\times10^{-3}$	$< 1.8\times10^{-4}$	$< 1.4\times10^{-4}$	$(1.4 \pm 0.8)\times10^{-4}$	$(3.0 \pm 0.7)\times10^{-3}$	–
PTFE (Dyneon™ TF 1620) (a)*	28.06	105.26	$< 1.2\times10^{-3}$	$(2.5 \pm 0.9)\times10^{-5}$	$< 2.8\times10^{-5}$	$(3.1 \pm 1.4)\times10^{-5}$	$(6.0 \pm 1.1)\times10^{-4}$	–
Kapton® HN (DuPont™) (a)	0.851	19.90	$< 2.7\times10^{-2}$	$(14.6 \pm 1.3)\times10^{-3}$	$< 1.3\times10^{-3}$	$< 1.1\times10^{-3}$	$< 5.5\times10^{-3}$	–
Cuflon® (Crane Polyflon) (a)	0.9616	15.53	$< 1.3\times10^{-1}$	$< 8.5\times10^{-4}$	$< 3.0\times10^{-3}$	$< 1.9\times10^{-3}$	$(4.8 \pm 1.5)\times10^{-2}$	–
IGLIDUR® (c)	1	12	–	$< 1.1\times10^{-2}$	$< 5\times10^{-3}$	$< 6\times10^{-3}$	$(3.3 \pm 1.1)\times10^{-2}$	–

(a) measured at LNGS; (b) measured at IRMM; (c) measured at MPIK; (d) measured at BNO; * Sintered on special request in a clean room by ElringKlinger Kunstofftechnik GmbH;