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A Low Background Counting Facility at Laboratori Nazionali del Gran Sasso

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A low background counting facility with gamma spectroscopy is working in the underground laboratory at LNGS. The main features and some measurements are described here. Copyright © 1996 Elsevier Science Ltd

Introduction

At Laboratori Nazionali del Gran Sasso a low background counting facility is in operation and its measurements are directed to looking for very weak and rare signals. Examples are: searches for double-beta decay, dark matter candidates, solar neutrinos, nucleon decay.

Such experiments have to fight against background signals whose sources have different origins: cosmic rays, natural or induced radioactivity, and electronic noise. Therefore experimental equipment has to be shielded against cosmic radiation, this is achieved by installing it in underground laboratories, and against particles emitted in nuclear decays, which is usually achieved by means of high Z and density shielding materials (lead, copper, mercury). Neutrons are produced in fission and (alpha, n)-reactions in the building materials and in the rocks of the mountain, which contain uranium and thorium. They can be thermalized with hydrogen-containing moderators (water, paraffin, polyethylene etc.) and captured in the moderator itself (where ^{6}Li, ^{10}B can be added) or by adding cadmium foils, for example (however p, alpha, and in particular gamma rays will be produced and should be further shielded).

In very low background experiments, the materials used in contructing the detector are themselves a source of background: each material contains radioactive contaminants at a level (in most of the cases) ranging from 10^{-6} to 10^{-12} g per g of material.

It is a standard procedure to measure the contents of radionuclides in materials before their use.

One of the simplest methods is to measure the gamma ray activity of the samples by means of germanium diodes. Large germanium detectors with good energy resolution are now available and a careful selection of low activity materials has been performed at Laboratori Nazionali del Gran Sasso to obtain detectors with a low background counting rate (Arpesella *et al.*, 1992a).

Radioactivity at the Laboratori Nazionali del Gran Sasso

The Laboratori Nazionali del Gran Sasso is located at a depth of about 3500 hg cm^{-2} of standard rock, the intensity of charged cosmic rays is reduced by a factor of about 10^{6} with respect to the flux at the surface. The measured rate is ~1 muon m^{-2} h^{-1}, the energy threshold being 2 TeV (Ahlen *et al.*, 1990).

The reduction of neutrons produced mainly by spontaneous fisssion in the rocks is also considerable. In Gran Sasso Laboratory the radioactivity of the rock is rather low (Bellotti *et al.*, 1986).

Neutron flux has been measured by several groups using different techniques. Recent measurements were made by Belli *et al.* (1989) in hall A for several months. They report a neutron flux of $\sim 3 \times 10^{-6}$ cm^{-2} s^{-1} for the thermal and $< 0.3 \times 10^{-6}$ cm^{-2} s^{-1} for the fast component (Bodini *et al.*, 1996). The neutron flux is therefore at least three orders of magnitude smaller than in any low background laboratory at the surface.

In contrast, the background due to gamma rays is similar or even higher than in a surface laboratory due to the radioactivity of the rocks and concrete which cover the halls.

Figure 1 shows a gamma ray spectrum registered at the Gran Sasso Laboratory with a Ge(Li) detector with appropriate shielding, compared to the spectrum obtained by a Ge(HP) detector of same dimensions in Milano, with a similar shielding. Two orders of magnitude are gained on the background level by installing the detector underground.

Figure 2 illustrates a gamma energy spectrum from natural radioactivity, as measured in hall C with a Ge(HP) detector.

Radionuclides observed with a Germanium detector

The main gamma sources of contamination can be classified as:

(i) radionuclides belonging to natural radioactive

series (^{226}Ra, ^{214}Bi, ^{214}Pb from ^{238}U chain, ^{228}Ac, ^{224}Ra, ^{208}Tl, ^{212}Pb from the ^{232}Th chain, the actinium series which originates from ^{235}U);

(ii) long-lived natural radionuclides like ^{40}K (relative isotopic abundance 1.17×10^{-4}; 1.26×10^{9} yr; ^{87}Rb, ^{115}In, ^{133}La, ^{142}Ce, etc.);

(iii) radionuclides of cosmogenic origin (e.g. ^{3}H, ^{7}Be, ^{14}C, ^{22}Na, ^{26}Al, ^{60}Co);

(iv) radionuclides of artificial origin (long-lived fission products like ^{95}Nb($t_{1/2}$ = 35 days), ^{95}Zr ($t_{1/2}$ = 64 days), ^{144}Ce ($t_{1/2}$ = 284 days), ^{106}Ru ($t_{1/2}$ = 367 days), ^{134}Cs ($t_{1/2}$ = 2.06 yr), ^{125}Sb ($t_{1/2}$ = 2.71 yr), ^{137}Cs ($t_{1/2}$ = 30.2 yr).

Radon (^{222}Rn) and thoron (^{220}Rn) are short-lived radioactive gases which originate as daughter products from the decay chains of uranium and thorium, they can diffuse and escape from the materials, thus breaking the secular equilibrium. By gamma spectroscopy, only the parts of these chains due to radium daughters can easily be investigated. Equilibrium can be broken at this level too, during chemical processing to obtain the final material, radium being more reactive than its radioactive parents. Therefore contamination with ^{238}U and ^{232}Th is expressed assuming that the radioactive chains are in equilibrium.

Cosmogenic radionuclides differ from material to material, they are produced by cosmic ray nuclear reactions through n, p, etc. interactions (at a relatively important rate when they remain above ground).

The Facility

At Laboratori Nazionali del Gran Sasso the low background facility is installed in the underground laboratory in a prefabricated building along the by-pass in front of Hall C. There are six germanium detectors whose main characteristics are described in Table 1.

The present detectors were constructed by PGT (Europe) with Silena Detektors (Europe) and Ortec (U.S.A.). These companies permitted us to follow the assembly and mounting procedures of the detectors after a careful selection of all materials used for the cryostat and its internal fittings.

Each detector is completely shielded by 25 cm of low activity lead (< 20 Bq/kg of ^{210}Pb) and 10 cm of oxygen free, high conductivity copper for the inner shielding, both in brick form. At the bottom of the shielding, there are 5 cm of acrylic and a thick foil of cadmium, as further shielding against neutrons.

A plastic cover surrounds the brick shielding where nitrogen is flushed in to reduce the radon contamination. Figure 3 shows the factor of 2 improvement obtained with this radon removal system. Tables 2 and 3 show the counting rates for the main contaminants present as background in the detectors and shielding. For two detectors, the radon removal has been achieved with a 2 cm thick acrilic shielding with 0.01 mbar of nitrogen overpressure.

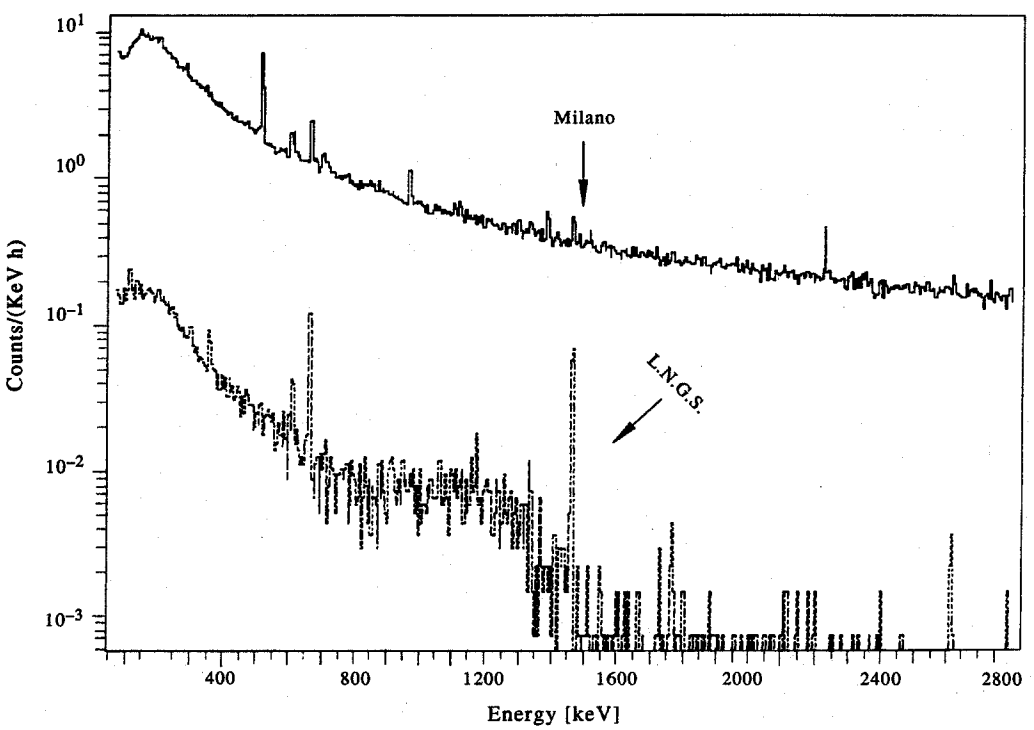

Fig. 1. Background gamma spectra with Ge detctor obtained in Milano and LNGS with proper shielding.

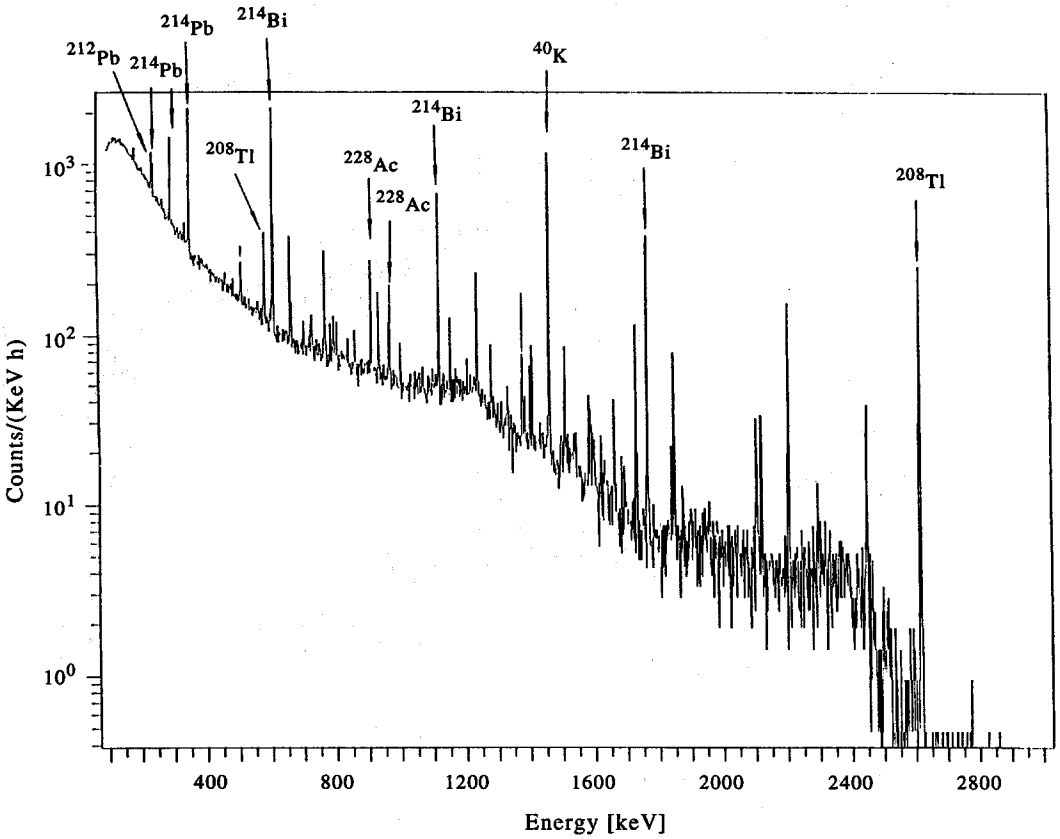

Fig. 2. Gamma energy spectrum from natural environmental radioactivity measured with a GeHP detector in hall C at LNGS.

The radioactive contamination of different materials can be studied and identified by measuring samples in the form of small cylinders which can be accomodated just above the cryostat in Marinelli beakers that closely fit over the endcap of the detector. Their volume is of the order of 1 L.

The contamination from U and Th is calculated on the hypothesis of secular equilibrium in the radioactive chains, where only gamma emitters are identified with this technique.

Read-out

The electronic system of these four detectors is similar: the pulses from the detector are collected by a charge preamplifier with the input JFET cooled at liquid-nitrogen temperature. The signals are amplified with a standard (Silena or Ortec) spectroscopic amplifier and processed by an analog to digital converter and a 4096 channel analyser (MCA-Silena model or Memory Buffer by Ortec).

Conversion gain is set at about 0.7 keV/channel in order to explore a gamma energy range up to ~3 MeV, while retaining a very good energy resolution.

The power supply of the spectrometer electronic chain is connected to the mains through a power stabilizer with back-up batteries; this reduces the electronic noise and protects against power failures of up to 2 h.

Off-line analysis

Detector efficiencies for samples of different geometry and different materials are determined by means of a Monte Carlo code (EGS4). This

Table 1. The main characteristics for the germanium detectors. Efficiency is relative to a 3″ × 3″ NaI crystal

Type	Volume (cm³)	Relative efficiency	FWHM (keV) at 1332 KeV	Peak:Compton ratio
Ge(HP) PGT	314	74%	2.2	67.5:1
Ge(HP)n-type Ortec	235	56%	2.0	63.2:1
Ge(HP) Ortec	423	85.5%	1.9	89.2:1
Ge(HP) Ortec II	414	96%	1.9	87.9:1
Ge(HP) Ortec III	363	91.4%	1.8	96:1
Ge(HP) Ortec IV	518	113%	2.0	88.3:1

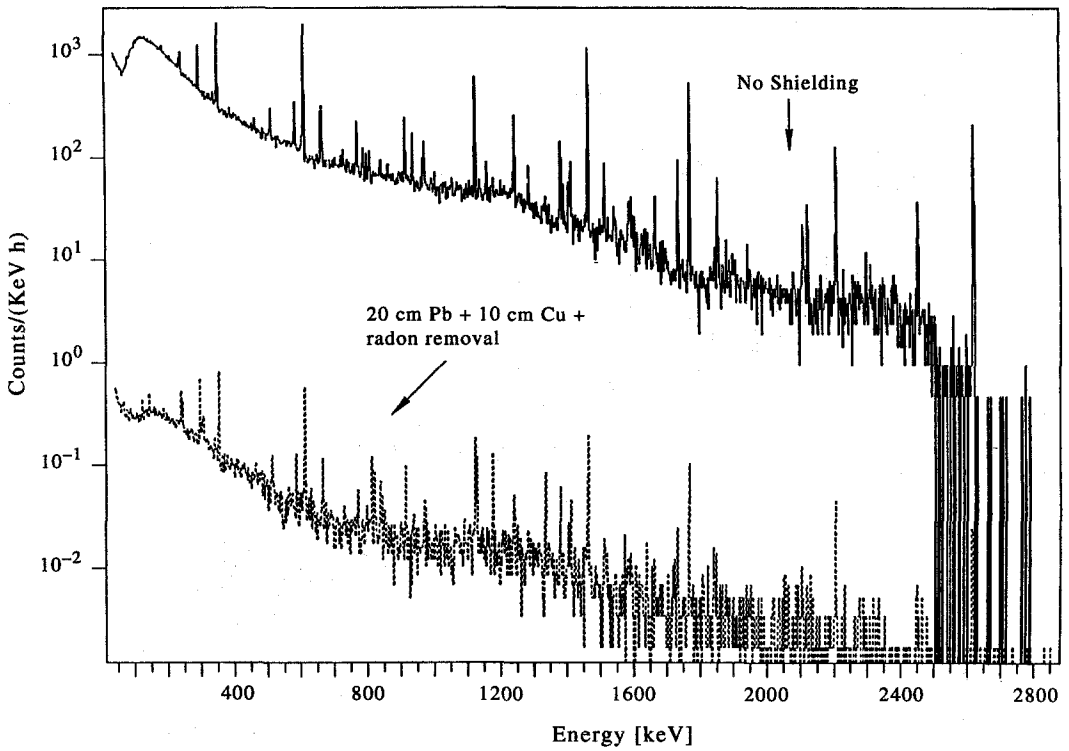

Fig. 3. Background gamma spectra with a GeHP detector in air and with proper shielding.

automatic program has been tested with measurements taken with calibrated radioactive sources, which provide many different gamma-ray energies (^{226}Ra, ^{152}Eu to evaluate cascade effects but also single nuclides). These sources were placed in different positions on the top face and sides of the detectors, they include point-, and diffuse-sources. The agreement with Monte Carlo calculations is within 10% in the energy range of natural gamma radioactivity.

From the above mentioned efficiency and low background, the sensitivity level of these detectors for gamma ray spectro-scopy reaches 10^{-3} Bq/kg (corresponding to a concentration of the order of 10^{-10} g/g for ^{238}U and ^{232}Th if the chain is in equilibrium). A long measurement (700 h) has been performed with 26 kg of low radioactive lead in a Marinelli beaker-shape around one large germanium detector. In the final spectrum, the characteristic X-rays of ^{210}Pb were present and indicate a contamination of the order of 20 Bq/kg, while no peaks from ^{238}U and ^{232}Th were found above background: an upper limit on their contamination of 100 mBq/kg was computed.

The spectra from the MCA are recorded on VAX computer and analysed with the TASSO code, which has been developed by Professor Fiorini's group from the Marmot code (Bodini *et al.*, 1986). It allows

Table 2. Counting rate of the main gamma lines for the various Germanium detectors. 1 s errors are indicated

Isotope	Energy (keV)	Ge(HP)PGT counts (h)	Ge(HP)n-type counts (h)	Ge(HP) Ortec counts (h)
^{238}U	295.2	0.10 ± 0.05	0.18 ± 0.04	0.380 ± 0.051
	351.9	0.2 ± 0.06	0.22 ± 0.04	0.622 ± 0.047
	609.3	0.17 ± 0.02	0.21 ± 0.05	0.539 ± 0.042
	1764	0.04 ± 0.02	0.03 ± 0.02	0.136 ± 0.017
^{232}Th	238.6	0.12 ± 0.06	0.38 ± 0.08	0.567 ± 0.055
	583.1	0.05 ± 0.03	0.20 ± 0.04	0.252 ± 0.031
	2614.7	0.02 ± 0.01	0.03 ± 0.02	0.142 ± 0.018
^{40}K	1460.7	0.17 ± 0.05	0.27 ± 0.09	0.619 ± 0.039
^{137}Cs	661.6	0.16 ± 0.02	0.20 ± 0.08	0.219 ± 0.028
^{60}Co	1173.2	0.56 ± 0.06	0.35 ± 0.10	0.163 ± 0.022
	1332.5	0.45 ± 0.05	0.32 ± 0.10	0.185 ± 0.022
^{125}Sb	427.9	1.19 ± 0.09	1.19 ± 0.09	
	600.6	0.62 ± 0.05	0.62 ± 0.05	
	635.9	0.36 ± 0.04	0.36 ± 0.04	
^{106}Ru	621.8	0.23 ± 0.03	0.23 ± 0.03	
	1050.1	0.06 ± 0.03	0.06 ± 0.03	

Table 3. Counting rate of the main gamma lines for the various Germanium detectors. 1 s errors are indicated

Isotope	Energy (keV)	Ge(HP) Ortec II counts (h)	Ge(HP) Ortec III counts (h)	Ge(HP) Ortec IV counts (h)
^{238}U	295.2	0.145 ± 0.048	0.225 ± 0.033	0.086 ± 0.022
	351.9	0.281 ± 0.069	0.445 ± 0.039	0.117 ± 0.038
	609.3	0.193 ± 0.052	0.317 ± 0.025	0.125 ± 0.019
	1764	0.053 ± 0.023	0.091 ± 0.013	0.029 ± 0.009
^{232}Th	238.6	0.320 ± 0.056	0.270 ± 0.030	0.116 ± 0.031
	583.1	0.134 ± 0.042	0.158 ± 0.021	0.077 ± 0.025
	2614.7	0.065 ± 0.026	0.067 ± 0.012	0.031 ± 0.008
^{40}K	1460.7	0.399 ± 0.058	0.278 ± 0.023	0.260 ± 0.041
^{137}Cs	661.6	0.180 ± 0.050	0.044 ± 0.012	0.031 ± 0.009
^{60}Co	1173.2	0.065 ± 0.026	0.281 ± 0.025	0.116 ± 0.018
	1332.5	0.088 ± 0.033	0.272 ± 0.021	0.132 ± 0.017

automatic peak search and fits with background substraction. Graphic output is analysed on a video-terminal and directed to printers.

Some Results

In the past a large number of samples have been measured. Table 4 reports a few examples.

Further Improvements

A few improvements are possible in order to obtain a better sensitivity:

(i) cleaner detectors. We are in contact with the Ortec company trying to build a cleaner germanium detector, by selecting and testing all materials which form the cryostat. We intend to put again a very low radioactive lead disc between the crystal and the JFET and electronics, inside the holder;

(ii) cleaner environment. To reduce the radon contamination in the laboratory where the detectors are situated, the ventilation system could be improved, by replacing rich-radon old air with fresh and radon-free air with the help of proper filters.

On the sample side, concentration procedures of the samples are necessary to improve sensitivity.

Measurements of Direct Physics Interest

With these germanium detectors, the following different types of measurement have been performed.

Roman lead (Alessandrello et al., 1991)

Gamma spectroscopy has been used to analyse samples from ingots of lead carried by a Roman ship sunk near Sardinia, more than 2000 years ago. These samples were compared to modern lead, modern electrolytic and specially prepared lead (Johnson and Matthey) and a sample of Dutch lead which is about 500 years old. While contaminations yielding high energy gamma rays result to be negligible in all samples, the analysis at low energies shows that

Table 4. Radioactive contamination in Bq/kg for some samples as measured with germanium detectors (limits are given at 95% C.L., errors at 1 s). The acquisition time and the mass of the samples are indicated too

	Measuring time (h)	Weight (g)	^{238}U	^{232}Th	^{40}K	^{60}Co
LNGS rock hall A			116 ± 12	12 ± 0.4	307 ± 8	
LNGS rock hall B			7.1 ± 1.6	0.34 ± 0.11	7 ± 1.7	
LNGS rock hall C			11 ± 2.3	0.37 ± 0.13	4 ± 1.9	
Concrete unicem 425	21.6	1206	52 ± 4	40.6 ± 0.3	303 ± 4.2	
Concrete 525 ptl	23.1	1095	26.1 ± 2	15 ± 0.2	164 ± 0.3	
Concrete 425 ars	23.63	1335	16.6 ± 1.8	8.8 ± 0.10	182 ± 2.7	
Concrete 425 sacci	65.74	1308	69.4 ± 1.4	15.4 ± 0.9	260 ± 2.0	
Deionized LNGS water	97.5	942	< 0.02	< 0.01	< 0.2	< 0.01
Varnish			3 ± 0.1	0.2 ± 0.03	3 ± 0.3	
Acrylic	111.7	125	< 0.02	< 0.01	< 0.2	< 0.01
Teflon HFPA44	90.1	292	< 0.02	< 0.01	< 0.2	< 0.01
Iron	48.1	844	< 0.07	< 0.02	< 0.7	< 0.02
Stainless steel 304L	96.2	414	< 0.06	< 0.07	< 1.0	0.2
s.s. AISI 304	68.6	257	0.1	< 0.04	< 1.0	0.07
s.s. CSM Roma	53.6	587	< 0.04	< 0.04	< 1.0	0.05
s.s. Russia (LVD)	77	770	< 0.08	< 0.02	< 0.9	< 0.02
Carbon steel			< 0.02	< 0.02	< 0.1	< 0.004
Titanium	162.48	65	< 0.05	< 0.04	< 0.5	< 0.02
Schott glass	118.3	442	0.8 ± 0.05	0.1 ± 0.04	1 ± 0.3	
Commercial low radioactive glass	114.7	101.4	1.7 ± 0.11	0.8 ± 0.01	10 ± 1.3	
Electrical component of phototube	68.7	65	20 ± 0.4	7 ± 0.3	9 ± 1	
Dynods	63.3	98	0.4 ± 0.07	< 0.2	< 3	< 0.05
Low bkg NaI crystal	218.1	48	< 0.02	< 0.06	< 0.2	< 0.05
Liquid scintillator NE235	698	1430	< 0.0023	< 0.0012	< 0.007	
$GaCl_3$ solution	554	3755	< 0.0018	< 0.001	< 0.010	
Roman lead	264.4	5000	< 0.01	< 0.009	< 0.3	< 0.007
J and M lead	335.3	5000	< 0.01	< 0.007	< 0.2	< 0.007
Modern lead (Samin)	189.9	5000	< 0.03	< 0.02	< 0.3	

modern lead contains a considerable amount of ^{210}Pb (~180 Bq/kg), but not Roman lead (< 2.6 Bq/kg).

This Roman metal is an excellent shielding material for low radioactivity experiments.

Argon 42 (Arpesella et al., 1992b)

Natural argon could contain ^{42}Ar, which is a β^- emitter (it decays to ^{42}K, which β^- decays with a Q-value of 3.52 MeV), with a half-life of 33 yr. An upper limit on the concentration of ^{42}Ar in natural liquid argon has been measured with the Ge(Li) detector. The sample (2 L) was put in a dewar of the form of a Marinelli beaker and a limit of $\leq 10^{-18}$ g ^{42}Ar/g ^{40}Ar at 90% C.L. was obtained. This measurement could be improved by an order of magnitude with an air-tight refilling system of liquid argon. In fact, during the measurement, the radon counting rate increases because of trapping of this gas from the room in the liquid argon.

Zirconium 96 and Neodymium 150

A double-beta decay search was performed with a powder sample (18 g) of ZrO_2 enriched in ^{96}Zr (57%) with the Ge(HP) PGT detector. ^{96}Zr is a double-beta candidate which could decay into excited ^{96}Mo states at 778.2 (2^+), 1147.9 keV (0^+), 1497.7 (2^+), 1625.8 (2^+), with transition energies of 778.2, 778.2 and 369.7, 778.2 and 719.5, 778.2 and 847.6 keV. With the present statistics, a lower limit on the lifetime of 2–4 10^{19} yr (at 90% C.L.) is obtained. In addition, the single beta decay of ^{96}Nb has been studied, providing a half-life for this process longer than 3.8 10^{19} yr (90% C.L.).

Another measurement has been performed with a natural sample of metallic Nd, looking for the double-beta decay of ^{150}Nd to excited states of ^{150}Sm. By analysing the spectra (6500 h of measurement) gamma lines from natural radioactive series and other radionuclides have been observed in the sample and contaminations have been computed from their intensities. Many checks were performed in order to test the computed detection efficiency, by studying the sum peaks, single and double escape peaks respect to the full energy peaks and the results were in good agreement. No gamma lines from the double-beta decay to excited states were found: a lower limit of 10^{20} yr (90% C.L.) for the half-life for this process was calculated.

Conclusions

A low background facility is available at the Laboratori Nazionali del Gran Sasso which consists, up to now, of six germanium detectors of low intrinsic radioactivity. They have been mounted with proper shielding in order to measure the intrinsic radioactivity of various materials of different low background experiments. In the same time, some measurements of physical interest—like double-beta decay search—have been started with the same apparatus. In the future, improvements can be made to achieve better sensivity and lower background by more care for the air-ventilation system of the laboratory, with the use of lower radioactive shielding, like Roman lead.

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