Radiopurity of an archeological Roman Lead cryogenic detector

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Abstract

Archeological Roman Pb is known to be a suitable material for shielding the experimental apparatus in rare event searches. In the last years the intrinsic radiopurity of this material was investigated using different technologies. In this work we applied the latest advancements of the cryogenic technique to study the internal radiopurity of a 1 cm\textsuperscript{3} sample of archeological Roman Pb. We report the lowest measured limit on \(^{210}\text{Pb}\) content inside the sample, with a concentration lower than 603 \(\mu\text{Bq/kg}\), better than any other reported measurement. Furthermore we also studied \(^{238}\text{U}\) and \(^{232}\text{Th}\) impurity concentrations. Our values concur with independent measurements reported in literature.

1 Introduction

In the Earth crust a temperature gradient of 0.5 K/km is observed \cite{1}. This variation is produced by \(\beta\) decays of long-lived isotopes, which are naturally present in the Earth interior. The most abundant radionuclides responsible for such heat release are \(^{238}\text{U}\) and \(^{232}\text{Th}\), and their decay products, and \(^{40}\text{K}\). These can be found practically in all materials on Earth, also because some of the nuclides of the decay chains are gaseous, such as \(^{222}\text{Rn}\).

The successful realization of ultra-low background experiments searching for elusive natural processes, such as neutrinoless double-beta decay \((0\nu\beta\beta)\) \cite{2}, dark matter-nucleus \cite{3} interactions or neutrino-nucleus interactions \cite{4} is made particularly difficult by environmental radioactivity. Unavoidably, natural radioactivity is included in many materials employed for the detector construction, making difficult to achieve ultra-low background conditions. As an example, if massive shielding surrounding the detector are not made from radiopure materials, these can become themselves the main background source, giving a larger contribution with respect to the external component (i.e. cosmic rays or high energy \(\gamma\)s). For these reasons material assay is of paramount importance for low counting rate experiments.

The first precaution adopted by ultra-low background experiments is to install their detectors in deep underground laboratories, where they can profit from the overburden rock which acts as a 4\(\pi\) shielding. To suppress the cosmic ray induced component of the background. However the environmental radioactivity can still limit the detector sensitivity. For this reason, further shields are installed around the experimental setup and, depending on the size of the detector, different types of materials are employed. Large volume detectors, such as Borexino \cite{5}, Gerda \cite{6}, DarkSide \cite{7}, take advantage of the availability of highly-pure liquids (e.g. water or Ar) for shielding the detectors. At the same time they instrument the shield to function as veto detector. On the other hand, small volume detectors, such as CUORE \cite{8}, CUPID \cite{9}, CRESST \cite{10}, which require a cryogenic facility, are designed to be compact and to be shielded with an overall reduced occupancy, given the technical limitations of the cryogenic systems. For this reason in these experiments Pb is used as primary shield and it is installed in proximity of the detector’s sensitive components.

The search for highly-pure Pb is a topic which is active since many years in the field of low background experiments. While the purification of liquids or gases is a well established technology \cite{11}. The Purification of Pb is a process in which the community of low background
scientists have not put too much effort due to the complexity of purifying materials which are not liquid at standard pressure and temperature. Furthermore, the availability of radiopure Pb is not always ensured and it requires thorough screening of its internal contaminations.

The most abundant contaminant measurable in Pb samples is $^{210}\text{Pb}$, produced by the $^{238}\text{U}$ decay chain. The difficulties in mitigating the concentration of $^{210}\text{Pb}$ in Pb samples are hampered by the chemical affinity of this nuclide with metal Pb and by its long half-life of 22.3 y. Whenever a $^{210}\text{Pb}$ contamination of a Pb sample occurs, most probably this could only be reduced by a cool-down on the time scale of the half-life of the nuclide. Therefore, it can be easily understood that ancient Pb samples, produced more than 100 y ago (i.e. more than 5 times the $^{210}\text{Pb}$ half-life), may feature an extremely low $^{210}\text{Pb}$ concentration.

In this work we review the status of different measurements on the radioactivity of ancient Roman Lead samples and we present the results on the excellent radiopurity of a pure Pb cryogenic detector made from archeological Roman Pb.

2 The $^{210}\text{Pb}$ Nuclide

Archaeological $^{210}\text{Pb}$ is a superior raw material for shielding realization. Its application is hampered by the presence of high internal contamination of $^{210}\text{Pb}$.

$^{210}\text{Pb}$ can induce different type of signatures in a detector, its decay scheme is shown in Fig. 1. It $\beta^-$ decays with an energy transition of 63 keV; it may populate an excited level at 46 keV which would then produce a 46 keV $\gamma$ before reaching the ground state of $^{210}\text{Bi}$. This nuclide has a short half-life (5 d), and it $\beta^-$ decays releasing 1.16 MeV of energy shared among the decay products. The energetic electron can produce bremsstrahlung radiation but also high-energy X-rays (up to 90 keV), while crossing the Pb sample.

After its decay, $^{210}\text{Bi}$ populates the ground state of $^{210}\text{Po}$ which, and in most of the cases (99.99% [12]) $\alpha$-decays on the stable nuclide $^{206}\text{Pb}$. The energy of the $\alpha$ is about 5.3 MeV, and it is considered a relevant source of background for $0\nu\beta\beta$ experiments [13]. In order to mitigate this possible background source, Pb is never directly facing cryogenic detectors but a thick layer of highly-pure Cu is always interleaved between the two.

3 Archeological Pb

While studying the radiopurity of modern Pb samples and Pb-based compounds, high level of $^{210}\text{Pb}$ concentration can be measured [14,15] at the level of 1000 of mBq/kg. This is due to the presence of $^{238}\text{U}$ decay chain products in Pb ores.

While refining raw metal Pb, as a matter of fact a $^{210}\text{Pb}$ concentration process is encountered. In fact, while other radioactive nuclides which are chemically not affine to Pb are segregated from the slag, $^{210}\text{Pb}$ is concentrated. This process directly results in high purity Pb, where a strong concentration of $^{210}\text{Pb}$ is present. As already discussed, a cool-down time sufficiently long would allow to reduce this residual radioactivity, making Pb an excellent highly-pure material for low background physics. Within the Roman Empire age, Pb was refined for various purposes [16] providing nowadays an important unintentional source of low radioactivity Pb.

The radiopurity of ancient Roman Pb was widely investigated and very low level of intrinsic contaminations were measured. The current best value for $^{210}\text{Pb}$ is a limit of 4 mBq/kg [17], which was obtained using an absorber of pure Pb operated as cryogenic detector. This innovative technique was proposed after the limited sensitivity of previous measurements. The standard technique used up to the publication time of [17] was $\gamma$-spectrometry with HP-Ge detectors, which provided limits at the level of hundreds of mBq/kg [19]. The HP-Ge (High-Purity Germanium) technique is not as sensitive as the cryogenic technique, because of the extremely low detection efficiency due both to geometrical reasons (the detector and the sample are separated) and to self-absorption in the sample. In fact, the most common approach is to detect the 46 keV $\gamma$ which has a very small branching ratio of 4% (see Fig. 1) and thus it is mostly absorbed by the sample. Instead, the cryogenic technique studies $^{210}\text{Po}$, the decay product $^{210}\text{Pb}$. The signal induced by $^{210}\text{Po}$ is in a favourable
Table 1 Summary of the most sensitive measurements on the concentration of $^{210}\text{Pb}$ in ancient Pb samples using different techniques.

| Detector | $^{210}\text{Pb}$ | Sample mass | Ref. |
|----------|------------------|-------------|------|
|          | [mBq/kg]         | [kg]        |      |
| HP-Ge    | $\rm{< 900}$     | 4.5         | [19] |
| HP-Ge    | 30000            | –           | [20] |
| Planar Si | 100              | 0.01        | [21] |
| Bolometer | $\rm{< 20}$     | 0.09        | [22] |
| Bolometer | $\rm{< 4}$      | 0.01        | [17] |

region of the energy spectrum with an excellent signal-to-background ratio (at 5.4 MeV), and it also has a branching ratio of almost 100%.

On the other hand, $\gamma$-spectrometry with HP-Ge detectors is extremely effective for investigating the concentration of $^{238}\text{U}$ and $^{232}\text{Th}$ in Pb samples. This is possible thanks to the high energy $\gamma$'s produced by these nuclides, (up to 2.6 MeV) which reduce the possibility of self-absorption in the sample, therefore enhancing the detection efficiency. Currently the best limits on the concentration of these isotopes are: $<46 \mu\text{Bq/kg}$ and $<45 \mu\text{Bq/kg}$, respectively [18].

In Tab. 1 a summary of the most relevant measurements on low-activity Pb samples is shown.

4 Archeological Pb based detectors

Many works can be found in literature where ancient Pb is embedded in cryogenic particle detectors. We can identify two classes of detectors: Pb-based scintillators, such as PbWO$_4$ [23] or PbMoO$_4$ [24], and pure-Pb absorbers.

The possibility to produce detectors, namely scintillating crystals, from radiopure materials is of paramount importance. In fact a detector, with excellent energy resolution and with the ability to perform particle identification, can enhance the physics potential of an experiment. Lead-based crystals are known to be excellent scintillators, but unfortunately they suffer from an excess of radioactivity that makes them not suitable for rare event investigations. The reason is the high concentration of $^{210}\text{Pb}$ which enters in the crystal during its production/growth. For the growth of commercial Pb-based crystals, commercial Pb is used, known to feature a high $^{210}\text{Pb}$ concentration at the level of Bq/kg. Furthermore, such commercial crystals may also exhibit high concentrations of $^{238}\text{U}$ and $^{232}\text{Th}$ decay chain products.

Recently, this problem was overcome by the use of ancient Roman Pb for the production of scintillating crystals such as PbWO$_4$ and PbMoO$_4$. Regrettably, these compounds still show a high concentration of $^{210}\text{Pb}$, $^{238}\text{U}$ and $^{232}\text{Th}$ at the level of hundreds of mBq/kg, due to a not complete control of the crystal growth process.

In 1998, pure Pb detectors were also successfully operated [17] and featured excellent radiopurity levels, thanks to the absence of any production process. The samples were directly cut from the bulk of an ancient Roman Pb brick as the one shown in Fig. 2.

The samples (10 g of mass each) where equipped with a Ge Neutron Transmutation Doped (Ge-NTD) thermal sensor and operated as cryogenic detectors at about 10 mK. The achieved energy resolution was in the range of 100 keV FWHM at 1.3 MeV, and, while investigating the $^{210}\text{Pb}$ content in the detector, no $^{210}\text{Po}$ signal above the background level was observed. A limit on the $^{210}\text{Pb}$ concentration in the detector was set $<4$ mBq/kg [17].

The difficulties in running such detectors are highlighted by the poor energy resolution. In fact operating a cryogenic detector which is a metal and it is in a superconducting state$^1$ is not trivial while using a thermal sensor sensitive to thermal phonons, like a Ge-NTD. Furthermore the Debye temperature of the detector is about 80 K, which makes its heat capacity at low temperatures not favourable for low energy studies.

The performance of such detectors might be enhanced while operating Pb with other class of thermal sensors, suitable to read-out superconducting absorber.

5 Pure-Pb cryogenic detector

The detector used for the measurement described in this work is one of the two samples already employed in [17]. The absorber has a volume of 1 cm$^3$ and a mass

\footnote{Pb has a critical temperature of 7.2 K.}
of 10 g. This was extracted from a 40 kg Roman Pb ingot, without the need of any further process.

The absorber was equipped with a Ge-NTD thermistor similar to the one used in [25], but with reduced dimensions: 3.0 × 1.0 × 0.3 mm³. The choice for a miniatuized thermal sensor is driven by the possibility to enhance the detector performance, namely the signal amplitude, while reducing the overall heat capacity of the system: absorber plus thermal sensor. The detector was also instrumented with a Si heater which was operated as Joule resistor, meant for the monitoring and the correction of temperature drifts of the absorber during the data taking.

The absorber was installed in a Cu housing and fixed to the support structure by means of epoxy resin. The detector was surrounded by a plastic light reflector (3M Vikuiti™) only for background reasons. In fact, the purity and the thickness of the reflector [25] make it suitable as shield against possible α decays occurring on the Cu surfaces facing the Pb absorber. Our region of interest lies around 5.4 MeV (Q-value of ²¹⁰Po), thus a possible background source can be ascribed to surface α decays as discussed in [26]. A picture of the complete detector is shown in Fig. 3.

The detector was installed in the low background CUPID R&D cryogenic infrastructure located at the Laboratori Nazionali del Gran Sasso of INFN (Italy).

The thermal sensor was biased with a constant voltage through 5 GΩ + 5 GΩ load resistors, which ensure a constant current flow of 2.9 nA through the thermistor. Its working resistance was 2.7 MΩ, thus much lower than the load resistors in series. The acquired signals are amplified by custom-made low noise electronics [27] and then filtered by means of a 6-pole Bessel filter with cutting frequency of 500 Hz. Finally, the signals are fed into a NI-PXI-6284 18-bits ADC. The sampling frequency was 1 kHz and the waveforms were recorded when a software derivative trigger fired.

6 Results

The detector was operated for more than 250 h, during which both background and calibration data were collected. During calibration runs the detector was exposed to a ²³²Th source deployed next to the experimental set-up, but outside the cryogenic system. Characteristic monochromatic γ emissions from the source were used for the calibration of the energy response of the detector, as well as for a monitoring of detector stability in time. Furthermore, a low intensity ²³²Th source was also installed nearby the detector, to study the detector response at lower energies.

The detector response to α interactions was not investigated in order not to spoil the detector background. In fact, this would have required the installation of a permanent α decay source directly facing the detector. This type of investigation was already performed with this very same detector in previous studies, in which the detector response was assumed to be the same for both α and β/γ interactions.

Fig. 4 Total background energy spectrum of a 1 cm³ pure Pb cryogenic detector acquired over 229 h. The peaks are labeled as follows: (1) ²¹²Pb, (2) ²²⁸Ac, and (3) ²⁰⁸Tl.
the most intense high energy monochromatic γ-line in the acquired energy spectrum.

The results of the data selection analysis are shown in Fig. 4, reporting the total background energy spectrum.

The energy spectrum features few monochromatic γ lines, ascribed to the near $^{232}$Th calibration source. The FWHM resolution for the most prominent line, from $^{212}$Pb, at 238 keV, is 22±1 keV. At this energy scale the detector response is clearly Gaussian, as shown in Fig. 5.

![Fig. 5 Detector energy response at 238 keV. This signature is induced by a γ interaction from $^{212}$Pb. The data are fitted using a convolution of a Gaussian function with a linear background.](image)

For the detector energy calibration and the study of the detector response at higher energies, an intense $^{232}$Th source was deployed next to the cryogenic system. The detector response at the highest energy γ line (2.6 MeV) is shown in Fig. 6. Different line shapes were studied, but the simplest that reproduced the detector response was a double-Gaussian. The reason for such shape may rely on the non-uniformity of the detector response. In fact the detector is not a single crystal and possible position dependence effects may influence the response at high energies, due to multi-Compton interactions in the absorber. This behaviour was also observed in [8]. The FWHM resolution at 2.6 MeV is computed to be 103.6±9.0 keV.

As shown in Fig. 4, the detector did not feature any clear signature around 5.4 MeV, which is exactly the Q-value of $^{210}$Po. As first approximation we can assume that the detector energy resolution scales linearly with energy [9], for this reason we would expect the energy resolution at 5.4 MeV to be 200±17 keV. In Fig. 7, a zoom in around the region of interest is shown.

![Fig. 6 Detector energy response at 2.6 MeV. This signature is induced by γ interaction from $^{208}$Tl. The data are fitted using a double-Gaussian function.](image)

We compute the minimum detectable activity [28] of $^{210}$Pb in our detector to be <603 μBq/kg at a 90%

C.L. The result achieved with this set-up is about one order of magnitude better than any previous established limit. The detector counting rate in this region is extremely small, demonstrating the excellent radiopurity level of archeological Roman Pb.

Using the same approach previously described, we are able to set upper limits on the concentration of $^{232}$Th and $^{238}$U decay chains, which are computed to be <0.28 mBq/kg and <0.38 mBq/kg, respectively. These values are not competitive with the ones reported in [13], but it is a further confirmation of the purity of such valuable material.

7 Conclusions

In this work we operated a 1 cm$^3$ cryogenic detector made of pure archeological Roman Pb, the very same absorber that was used in [17]. The low-background environment in which we run the detector allowed us
to study the internal radiopurity of the absorber. The most stringent limit on the concentration of $^{210}\text{Pb}$ was established at the level of $<603 \mu\text{Bq/kg} \left( <2 \times 10^{-19} \text{g/g} \right)$. The absorber features an overall excellent radiopurity and no excess of $^{232}\text{Th}$ and $^{238}\text{U}$ decay chain products were observed; a limit on their concentration was also set $<0.28 \text{mBq/kg}$ and $<0.38 \text{mBq/kg}$, respectively.

The excellent radiopurity level and the preliminary performance as cryogenic detector make this material an interesting candidate for neutrino physics applications such as coherent neutrino-nucleus elastic scattering, but especially for the detection of supernova neutrinos, as discussed in [29]. The performance as a cryogenic detector can be further optimized by the growth of a single-crystal absorber (e.g. melting and re-crystallization of ancient Pb) and by the development of suitable thermal sensors (e.g. Ge-NTDs or Transition Edge Sensors).

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