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Development of ultralow-background cryogenic calorimeters for the measurement of surface α contamination

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Abstract

Next-generation experiments searching for rare events must satisfy increasingly stringent requirements on the bulk and surface radioactive contamination of their active and structural materials. The measurement of surface contamination is particularly challenging, as no existing technology is capable of separately measuring parts of the ^{232}Th and ^{238}U decay chains that are commonly found to be out of secular equilibrium. We will present the results obtained with a detector prototype consisting of 8 silicon wafers of 150 mm diameter instrumented as bolometers and operated in a low-background dilution refrigerator at the Gran Sasso Underground Laboratory of INFN, Italy. The prototype was characterized by a baseline energy resolution of few keV and a background $<100\,\mathrm{nBq/cm^2}$ in the full range of α energies, obtained with simple procedures for cleaning of all employed materials and no specific measures to prevent recontamination. Such performance, together with the modularity of the detector design, demonstrate the possibility to realize an alpha detector capable of separately measuring all alpha emitters of the ^{232}Th and ^{238}U chains, possibly reaching a sensitivity of few nBq/cm^2.

Keywords: Material screening, Bolometric α detector, Low-radioactivity measurements
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1. Introduction

In the field of rare event searches, such as direct Dark Matter (DM) detection or neutrinoless double beta ($0\nu\beta\beta$) decay, the naturally occurring radioactive uranium and thorium contaminants are among the most worrisome background sources. In particular, ^{238}U and ^{232}Th are the progenitors of decay chains composed of 14 and 10 radioactive isotopes, respectively. Both chains are about equally divided between α and β emitters, with 1/3 of the isotopes also producing de-excitation X or γ rays in coincidence with the α or β particle. While ^{238}U and ^{232}Th have very long half-life values of $\sim 10^{10}$ y, their progeny features a variety of half-life values ranging from 300 ns to 10^5 y. Moreover, the chemical properties of the various chain members strongly affect their relative concentration in a given material, often causing the chain to be out of secular equilibrium.

The contamination can have different spatial distributions in the material and structural components used in the experiments, and can be separated into *bulk* contamination if its density is homogeneous, or *surface* contamination if it is concentrated on the surface of the considered item. While any bulk contamination is fully determined by the history of the material production, the surface contamination depends also on the subsequent machining operations, the cleaning procedure, and the exposure to ambient radon. The average concentration of uranium and thorium in the Earth crust is at the level of 10^{-6} – 10^{-5} g/g [1], a value that needs to be suppressed by several orders of magnitude to fulfill the requirements of rare event experiments. Several technologies allow measuring ultra-low bulk contamination levels: Inductively Coupled Plasma Mass Spectrometry can reach sensitivities of $\lesssim 10^{-14}$ g/g for both uranium and thorium [2, 3] corresponding to <40 and <125 nBq/kg, respectively; low-background germanium detectors operated underground allow γ spectroscopy with sensitivity down to 1 μBq/kg [1], and Neutron Activation Analysis offers an even higher sensitivity, but for a limited number of materials [4, 5]. Thus, it is common practice to select the materials based on their bulk radio-purity.

However, surface contamination is a significant – if not dominant – contributor to the background of several current and future experiments. For example, cryogenic calorimeters (or bolometers) for $0\nu\beta\beta$ decay, consisting of crystals containing the candidate $\beta\beta$ isotope operated at ~ 10 mK and read-out with thermistors or superconducting sensors, search for a signal induced by two electrons at the Q-value of the reaction (typically in the 2–3 MeV range, depending on the isotope). The background of the most sensitive current-generation experiment, CUORE, is induced predomintantly by degraded α particles emitted by the surface contamination of the copper structure supporting the crystals [6, 7, 8], with a ^{232}Th and ^{238}U activities of 5 nBq/cm^2 and 14 nBq/cm^2, respectively [6]. In the near future, the next generation experiments CUPID [9] and AMoRE [10] aim at suppressing this background by performing particle discrimination. Nevertheless, surface contamination still represents a background. In fact ^{214}Bi, which belongs to the ^{238}U chain, β-decays with an end-point enrgy of 3.3 MeV, and can induce a background as already observed in two pilot experiments [11, 12]. Therefore, a further minimization of the surface contamination is necessary.

Next-generation experiments require extensive campaigns of surface contamination reduction and screening, only through which they can test and validate their techniques for cleaning or coating the surfaces of their components. The most common approach is using α detectors. An ideal detector should allow measuring surface contamination of few $\mathrm{nBq/cm^2}$ in a short time – one month or less. To reach such a goal, the system should be capable of measuring material samples with a total area of at least $1\,\mathrm{m}^2$, and have a background $\lesssim 10\,\mathrm{nBq/cm^2}$ in the entire α region. Moreover, the capability of distinguishing α events from different nuclei of the ^{232}Th and ^{238}U chain and identifying possible breaks of secular equilibrium, can be granted only through an energy resolution better than $\sim 20\,\mathrm{keV}$ FWHM.

2. The detector prototype

The requirement listed above could be satisfied with an array of large-area bolometers interleaved with material samples. In fact, bolometers are very sensitive to α particles because of the lack of dead-layers, present in e.g. silicon detectors. This feature grants a high detection efficiency and prevents any signal deformation, allowing in principle to measure the depth profile of surface α contamination, and to distinguish α peaks induced by different components of the ^{232}Th and ^{238}U chains, allowing to separately measure those parts of the chains that are out of secular equilibrium. Moreover, bolometers can be fabricated with a variety of absorbers, including high-resistivity intrinsic float-zone silicon and sapphire, which are commercially available in the form of wafers with up to 20 cm in diameter.

To investigate the feasibility of a bolometric α spectrometer for the measurement of surface radioactive contamination, we have realized and tested a first detector prototype consisting of 8 silicon wafers of 15 cm diameter and 1 mm thickness mounted on 4 copper frames, with 2 wafers per frame (Fig. 1). The wafers have a resistivity $\sim 20\,\mathrm{k\Omega{\cdot}cm}$ and were purchased from TopSil [13]. Each wafer is instrumented with a Neutron Transmutation Doped (NTD) [14] thermistor glued on it. NTDs guarantee a large energy dynamic range, that can reach 4 orders of magnitude in sensitivity to temperature variation. Thus, they allow to measure events from $\sim 1\,\mathrm{keV}$ up to 10 MeV. The NTDs' response is temperature-dependent, hence we instrumented each wafer with a small resistor (denoted as heater) and used it to inject artificial heat pulses for an offline temperature stabilization. Such a technique is commonly applied to bolometers [15, 16]. We employed the same NTDs used for the CUPID-Mo demonstrator [17], and leftover heaters from the CUORE production batch. We performed two sets of measurements at a base temperature of $\sim$15 mK, for a total live time of 6.5 d, in a R&D cryostat of the CUPID group at the underground laboratory of LNGS.

For energy calibration, we realized a custom α source by smearing a nitric acid solution containing ^{147}Sm on a nylon foil, and placed the foil around the detector frames. ^{147}Sm emits exclusively one α particle with the energy of 2.248 MeV, thus allowing to calibrate the detector without spoiling the 3–10 MeV energy region, where all α's from the ^{232}Th and ^{238}U chains are expected. The drawback of ^{147}Sm is its long half life of $1.1{\cdot}10^{11}$ y: in order to achieve an activity of 0.1 Bq, sufficient to calibrate the detectors, a relatively large amount of samarium was needed, resulting in a layer that was much thicker than the $\lesssim$ 10 nm required for obtaining high-resolution α peaks. In addition, the nylon porosity favored the samarium penetration in the foil to the extend that the ^{147}Sm α spectral shape could only give a rudimentary estimate of the energy scale.

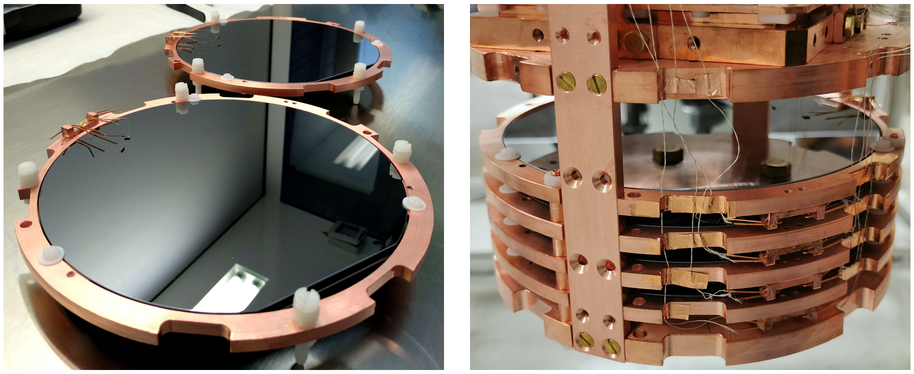

Figure 1: Left: two detector prototypes after fabrication. The NTD and heater, wire-bonded to custom pin connectors made of capillary copper tubes, are visible on the left of each detector. Right: the detector prototypes mounted in the cryostat. The copper structure at the top is the mounting of a different detector. Constantan wires connect the copper pins to the readout electronics.

3. Detector performance

Out of the 8 prototypes, one detector never functioned, one was affected by high noise because of a malfunctioning electronic board, and two detectors saw α events but no heater pulses, as a consequence of broken gold wires or failing wire bonds. The two heaterless detectors were the ones on the top and bottom of the array. The same detectors were also affected by a higher α background because they were facing two copper plates that are part of the detector structure. Therefore, we considered the data of only the 4 detectors that showed the best performance.

The detector signals were characterized by a rise time, defined as the 10-90% part of the leading edge, between 30 and 60 ms (Fig. 2, left), and a decay time between 50 and 200 ms. The reason for such variation in the signal time profile is still unclear, but might be due to either different thermalization of the detectors to the heat sink, or to the different working points of the NTDs. The event rate in all detectors was of few events per minute, with the great majority of events

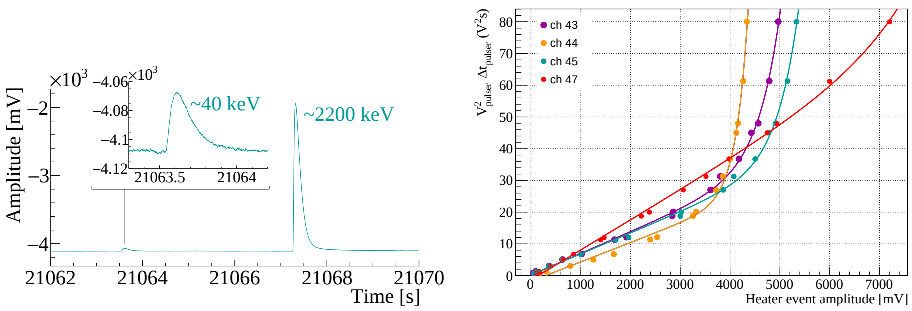

Figure 2: Left: example of data stream from one of the prototype Si detectors, showing a ^{147}Sm event at ∼2200 keV, and a smaller event (enlarged in the inset) with an energy of ∼40 keV. Right: power injected into the heater as a function of the corresponding event amplitude (in mV). The detector gain was chosen so that 1 mV corresponds to ∼1 keV of deposited energy.

below 1 MeV. The event rate in the α range (above 2.5 MeV) was of the order of few per day. All detectors see ^{147}Sm events with a rate of ∼10 events/h (Fig. 3).

The energy resolution (FWHM) of the heater-injected pulser events is in the 0.5–4 keV range, depending on the specific detector. Unfortunately, no spectral feature was visible in the energy spectrum of physical events, apart from the ^{147}Sm peak that was affected by a strong low-energy tail caused by the already mentioned porosity of the nylon foil and the thickness of the active source layer. However, the exceptional resolution on the pulser events clearly indicates that a resolution <10 keV on α peaks is possible, and that these detectors could be a good candidate for high-precision α spectroscopy.

The energy threshold, obtained with a derivative trigger, was also at the level of 1–6 keV, and reflects the pulser resolution. The use of a more sophisticated triggering algorithm such as the optimum trigger [18] could reduce the threshold by up to a factor ∼5 [7], and bring it well below 1 keV. In future measurements, the detector irradiation with high-activity γ sources could allow to detect the X-rays from copper (0.9 keV and 8 keV), and silicon (1.7 keV), providing extremely useful information on the detector performance on physical events.

During the data collection, we noticed a strong non-linearity of the energy scale that was likely caused by an overbias of the NTD sensors (Fig. 2, right). Because of the limited time availability of the cryostat, we decided to keep the same NTD bias, with the goal of maximizing the data useful for the evaluation of the α background. To characterize the non-linearity of the detector response, we collected ∼2 h of data injecting heater events with 10 amplitudes ranging from an equivalent energy of ∼50 keV to ∼6 MeV. Figure 2 (right) shows the power injected to the heater as a function of the reconstructed peak amplitude. The detector gain was set so that 1 keV of deposited energy would provide a signal of ∼1 mV amplitude. As visible from Fig. 2 (right), for three detectors the energy scale was linear up to 3-4 MeV, and strongly non-linear at higher energies.

4. Results and perspectives

The integrated number of counts in the full α region above 2.5 MeV yields an α background of $<100\,\mathrm{nBq/cm^2}$. This is the lowest background ever achieved by any α detector, despite the fact that the detectors were not assembled or mounted in a clean room or in a radon-free environment. Nevertheless, the strong non-linearity of the detector response above $\sim$3 MeV prevents, at the current stage, a precise energy determination and the identification of α decaying radionuclide.

In the near future, we intend to implement a correction for such non-linearity to achieve a reliable energy evaluation up to 10 MeV. Moreover, we plan to perform additional measurements to determine the optimal NTD working points to preserve the detector response linearity, characterize the energy resolution using X-ray and α sources, measure the background with an array of at least 10 detectors, and disentangle the backgrounds originating from silicon contamination from that originating from copper by reconstructing the events in coincidence between different detectors.

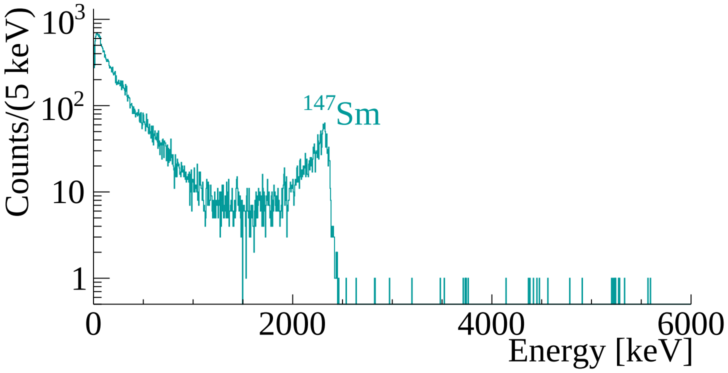

Figure 3: Sum spectrum of the 4 best-performing prototype detectors. The ^{147}Sm source yields an α peak at 2.248 keV with a low-energy tail due to the thickness of the samarium layer on the nylon foil. The β/γ continuum below 1.5 MeV is due to Compton events induced by γ sources used to calibrate other detectors operated in parallel in the same cryostat. The energy of events above $\sim$3 MeV is not reliable due to the presence of a strong detector non-linearity.

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