Production and characterization of $^{228}$Th calibration sources with low neutron emission for GERDA

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ABSTRACT: The GERDA experiment at the Laboratori Nazionali del Gran Sasso (LNGS) searches for the neutrinoless double beta decay of $^{76}$Ge. In view of the GERDA Phase II data collection, four new $^{228}$Th radioactive sources for the calibration of the germanium detectors enriched in $^{76}$Ge have been produced with a new technique, leading to a reduced neutron emission rate from $(\alpha, n)$ reactions.

The gamma activities of the sources were determined with a total uncertainty of $\sim 4\%$ using an ultra-low background HPGe detector operated underground at LNGS. The neutron emission rate was determined using a low background LiI(Eu) detector and a $^3$He counter at LNGS. In both cases, the measured neutron activity is $\sim 10^{-6}$ n/(s·Bq), with a reduction of about one order of magnitude with respect to commercially available $^{228}$Th sources. Additionally, a specific leak test with a sensitivity to leaks down to $\sim 10$ mBq was developed to investigate the tightness of the stainless steel capsules housing the sources after their use in cryogenic environment.

KEYWORDS: Instrumentation for neutron sources; Neutron detectors (cold, thermal, fast neutrons); Radiation monitoring; Detection of defects

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1 Introduction

The GERmanium Detector Array (GERDA) [1] is an experiment searching for neutrinoless double beta decay ($0\nu\beta\beta$) in $^{76}$Ge at the Gran Sasso National Laboratory of INFN (Italy). In GERDA, the germanium crystals, enriched to $\sim$ 86\% in $^{76}$Ge, are simultaneously operated as source and detectors. They are immersed in 64 m$^3$ of liquid argon (LAr), acting as cooling medium and shield against external radiation. The expected $0\nu\beta\beta$ decay signature is a peak at the Q-value of the reaction ($Q_{\beta\beta}$), 2039 keV for $^{76}$Ge [2]. The GERDA physics program is divided in two stages: the first physics run, denoted as Phase I, took place between November 2011 and June 2013 with about 15 kg of $^{76}$Ge and a background index (BI) at $Q_{\beta\beta}$ of $10^{-2}$ events/(keV·kg·yr). In the second data collection period, Phase II, the active mass will be doubled and a BI of $10^{-3}$ events/(keV·kg·yr) is aimed for [3]. This is achievable with the use of materials with higher radio-purity, the application of pulse shape discrimination (PSD) and the implementation of an active anti-coincidence veto in LAr.

The energy calibration of the germanium detectors is performed on a weekly basis. Up to three calibration sources can be lowered in LAr in the vicinity of the detectors through a dedicated calibration system [4–6] developed and built at the University of Zurich. In order to avoid radon contamination of LAr, the sources cannot be removed from the experimental setup during physics runs. They are therefore parked in gaseous argon on the top of the LAr cryostat, at the maximum possible distance from the detectors. In addition, a tungsten absorber is mounted below the sources to further shield the detectors from gamma radiation.

The choice of the isotope for the sources is based on several requirements. First, a substantial number of gamma lines has to be available for the calibration of the energy scale up to $Q_{\beta\beta}$. Second, the half life has to be at least of the same order of the experiment’s live time, i.e. a few years. Finally,
the PSD methods for germanium detectors are usually tuned with double escape events, which have very similar signal properties to the expected $0\nu\beta\beta$ decay events [7]. For this reason, a double escape peak (DEP) with sufficient statistic has to be available. The candidate which optimally fulfills these requirements is $^{228}$Th, with a 1.9 yr half life, a dozen of high statistic lines between 500 keV and 2.6 MeV and a DEP at 1592.5 keV [6].

The main drawback of using $^{228}$Th sources is that, starting from a $^{228}$Th nucleus, 5 $\alpha$ particles with energies between 5.2 and 8.79 MeV are emitted before a stable nucleus ($^{208}$Pb) is reached [8]. If the radioactive substance is embedded in materials with low threshold for ($\alpha$, $n$) reactions, a parasitic neutron flux is obtained. This is the case for standard commercial sources, for which ceramic components, e.g. NaAlSiO$_2$, are used for practical reasons. In GERDA, neutrons could generate $^{77}$Ge and $^{77m}$Ge via $^{76}$Ge activation, both of which undergo beta decay with $>$ 2 MeV Q-value, or they could be captured in the materials surrounding the detectors and produce high energy gamma rays. The background contribution at $Q_{\beta\beta}$ prior to the application of pulse shape discrimination and liquid argon veto cuts induced by such neutrons would be $3 \times 10^{-5}$ events/(keV·kg·yr·kBq) [4]. With a total activity of $\sim$ 70 kBq, this would not fulfill the background requirements of GERDA Phase II [3]. The reduction of the neutron source strength by about one order of magnitude can be achieved by embedding $^{228}$Th in a metallic material with threshold for ($\alpha$, $n$) reactions higher than 8.8 MeV. As described in [4, 9], gold is the best candidate due to its 9.94 MeV threshold, its $< 20 \mu$m range for $^{228}$Th alphas, its ductility and ease of procurement. This paper describes the production and characterization in terms of gamma and neutron activity of four custom made $^{228}$Th sources for GERDA Phase II. The main novelties with respect to [9] are the measurement of the gamma activity in background-free conditions and the comparison with a full MC simulation of the experimental setup. Moreover the measurement of the neutron activity was performed with a low-background LiI(Eu) detector and a $^3$He counter, both of which are operated underground at LNGS.

Given the ultra-low background requirements of the GERDA experiment and the need to deploy the sources at cryogenic temperatures, it is crucial to avoid any leakage of the source active material. Thus, a new leak test procedure with a sensitivity to leaks of $\approx 10$ mBq activity was developed at the ENEA-INMRI, Italy, and tested for the first time on the GERDA Phase II sources.

In section 2 the production of low-neutron emission $^{228}$Th sources is revised. Sections 3 and 4 describe the measurements and the estimation of the sources activity and neutron strength. In section 5 the results of leak tests after use in cryogenic environment are reported.

2 Custom production of $^{228}$Th calibration sources

The production technique of $^{228}$Th sources with reduced neutron emission was originally developed by the University of Zurich (UZH) and the Paul Scherrer Institute (PSI). Several prototype sources were produced during GERDA Phase I [4]. The technique was later used and improved [9] to produce a strong calibration source of several MBq activity for Borexino. A brief summary of the technique is given below.

The radioactive substance was provided in form of ThCl$_4$ dissolved in 1 M HCl by Eckert und Ziegler Isotope Products Int., Valencia (CA), U.S.A., with a total $^{228}$Th activity of 150 ± 23 kBq. Chlorine has two stable isotopes, $^{35}$Cl and $^{37}$Cl, both of which have ($\alpha$, $n$) thresholds below 8.8 MeV.

It is therefore mandatory to completely separate chlorine from thorium prior to deposition on gold.
The thorium tetrachloride solution was evaporated in a PTFE crucible almost to dryness; it was subsequently worked-up two times with 1 ml concentrated nitric acid and dried by evaporation. Thus the ThCl$_4$ was converted in Th(NO$_3$)$_4$ using surplus concentrated HNO$_3$ through the reaction:

$$\text{3ThCl}_4 + \text{16HNO}_3 \xrightarrow{-115^\circ\text{C}} [\text{3Th(NO}_3)_4]_s + [\text{4NOCl} + \text{8H}_2\text{O} + \text{4Cl}_2]_g$$

(2.1)

where the subscripts $s$ and $g$ stand for solid and gaseous, respectively. Subsequently, the solid Th(NO$_3$)$_4$ was diluted in two molar HNO$_3$ and transferred into a gold crucible prepared out of a 2 $\times$ 2 cm gold foil of 20 $\mu$m thickness. The gold has a $\geq 99.99\%$ purity and was produced by mechanical means, lamination and hammering, with no alteration of the original purity level [10]. The HNO$_3$ was evaporated by heating the solution to $\sim 120^\circ\text{C}$. Subsequently, the gold crucible was folded and heated to $\sim 700^\circ\text{C}$ to form ThO$_2$ on the gold surface to evaporate the remaining nitrogen and oxygen through the reaction:

$$\text{3Th(NO}_3)_4 \xrightarrow{-700^\circ\text{C}} [\text{3ThO}_2]_s + [\text{12NO}_2 + \text{3O}_2]_g$$

(2.2)

Finally, the gold crucible was further folded and wrapped in a second gold foil of the same dimension in order to prevent any loss of radioactive material. The procedure was performed separately for each of the four sources. The production of the GERDA Phase II sources was performed at the Institute for Nuclear Chemistry of the University of Mainz, Germany.

The sources were then encapsulated by Eckert und Ziegler Nuclitec GmbH, Braunschweig, Germany, with a double-sealed stainless steel VZ-3474 capsule, and certified according to ISO-2919 requirements.

3 Measurement of source activity

A precise estimation of the sources activity is required for several reasons. First, a correct $0\nu\beta\beta$ decay analysis can be performed only if a proper understanding of all the background components is available. This is possible thanks to the development of a background model via Monte Carlo (MC) approach and the fit of the model to the physics data. The correctness of the MC geometry and physics implementation can be assessed by simulating the calibration measurements and comparing the simulated with the experimental spectra recorded by the germanium detectors. In GERDA Phase I, the activity of the calibration sources was known with a 15% uncertainty [4], hence the quality of the MC model could only be inferred by comparing the spectral shape of simulated and measured spectra, and not by the absolute number of events. Second, the simulation of the calibration measurement is required for the estimation of the PSD efficiency on the DEP, the Compton continuum and the full energy peaks [7]. Finally, an estimation of the LAr veto efficiency for different background source locations can be addressed by lowering the $^{228}$Th sources to different positions and comparing the measured with the simulated suppression factors.

The measurement of the Phase II $^{228}$Th source activities was performed in the Gator facility [11] at LNGS. Gator consists of a high-purity p-type coaxial germanium crystal encased in an ultra-low activity copper cryostat. The detector is placed in a $25 \times 25 \times 33$ cm$^3$ cavity where the material samples can be inserted. The integrated background of Gator is $\sim 0.16$ events/min in the 100-2700 keV range. The $\sim 30$ kBq activity of Phase II sources induces a count rate which is 4 orders of magnitude higher, hence the measurements described here can be considered background free.
A 20 min long measurement was performed for each of the four sources, placed on a PTFE holder at ∼ 120 mm distance from the top of the detector to reduce the pile-up rate. The dead time was between 16 and 25%, depending on the source activity. The spectrum is recorded in the 10-2770 keV range, thus covering the entire 228Th spectrum, with the highest energy line at 2614.5 keV.

In order to estimate the activity, a Geant4 [12] based simulation was performed. A negligible fraction of events are present in the experimental spectrum above the 2614.5 keV peak. These are pile-up events and are not accounted for in the MC. Similarly, a ∼ 25% discrepancy between data and MC is visible below ∼ 70 keV. This is attributed to a sub-optimal implementation of the 228Th decay chain in Geant4. To avoid any bias due to the two effects explained above, the analysis is performed in the 100-2617 keV range.

For the comparison of measured and simulated data the finite experimental energy resolution is applied to the simulated spectrum via energy smearing on a single event basis. The full width at half maximum (FWHM) of all visible gamma peaks present in the spectrum (see figure 1) is determined via peak fitting. The FWHM as a function of energy is extracted through a chi-square fit using the function [13]:

$$\text{FWHM}(E) = \sqrt{w_e^2 + w_p^2 \cdot E + w_c^2 \cdot E^2}$$  \hspace{1cm} (3.1)

where $w_e^2$ is the constant electronic noise contribution, $w_p^2 \cdot E$ is related to the charge production, and $w_c^2 \cdot E^2$ is connected to the charge collection. For each simulated event with energy $E$ a quantity $\Delta E$ is extracted with a random sampling from a Gaussian distribution centered at zero and with $\sigma = \text{FWHM}(E)/2.355$, and then added to $E$. Eq. 3.1 is used for the smearing of all simulated events, with the exception of those belonging to the annihilation peak and to the single escape peak. Namely, they are broader than a corresponding gamma line due to Doppler broadening. In these cases the FWHM from the peak fit, and not the fitted FWHM as function of energy, is exploited.

The activity is estimated with a maximum likelihood analysis performed with BAT [14]. The log-likelihood is written as:

$$\ln L = \sum_{\text{bin}} \ln \left( \frac{\lambda_i^{k_i} \cdot e^{-\lambda_i}}{k_i!} \right)$$  \hspace{1cm} (3.2)

where $k_i$ is the measured number of events and $\lambda_i$ the expectation value in the bin $i$, defined as:

$$\lambda_i = \frac{A \cdot \Delta t \cdot R_i}{N_{MC}}$$  \hspace{1cm} (3.3)

$A$ is the source activity, $\Delta t$ is the live time of the measurement, $R_i$ the number of events in the $i$-th bin of the simulated spectrum and $N_{MC}$ the total number of simulated events. The only free parameters is $A$, its best value is given by the maximum of the $\ln L$, and its uncertainty is obtained from the central 68% interval.

The experimental spectrum, along with the simulated one scaled according to the best-fit result, is shown in figure 1 for one of the sources. The good agreement between data and MC at all energies in the considered range together with the high statistics of the measurement are reflected in a statistical uncertainty on the activity of ∼ 2‰. The estimated activity of the four Phase II sources is reported in table 1.
Figure 1. $^{228}$Th spectrum recorded by the Gator spectrometer with the AD9857 source (black), together with the simulated spectrum (red) normalized according to the best fit. The gamma lines used for the energy calibration in GERDA are labeled. SEP and DEP stand for single and double escape peak, respectively.

Table 1. Estimated activity of the four GERDA Phase II $^{228}$Th calibration sources. The first error is to the statistical uncertainty of the fit, while the second is the total systematic uncertainty.

| Source | Activity [kBq] |
|--------|----------------|
| AD9854 | $24.2 \pm 0.1$ (stat) $^{+0.1}_{-0.0}$ (syst) |
| AD9855 | $34.2 \pm 0.1$ (stat) $^{+1.5}_{-1.3}$ (syst) |
| AD9856 | $30.8 \pm 0.1$ (stat) $^{+1.4}_{-1.2}$ (syst) |
| AD9857 | $41.3 \pm 0.1$ (stat) $^{+1.8}_{-1.6}$ (syst) |

The major source of uncertainty for this measurement is of systematic origin. It is due to the limited knowledge of the experiment’s geometry, to an imprecise knowledge of the cross sections for the physics processes involved, and to the smearing and fitting procedures.

Regarding the implementation of the geometry in the MC simulation, the major uncertainty is induced by the distance between the source and the germanium crystal, known with $\pm 2$ mm precision. All other uncertainties connected to the geometry and thickness of the materials in between can be considered negligible. A difference of $\pm 2$ mm distance has negligible effect in the spectral shape but results in a different covered solid angle and, therefore, a different activity. The simulations and the analysis have been repeated after changing the source height by $\pm 2$ mm. This results in a $^{+2.5}_{-2.1}$% systematic error on the estimated activity.

The uncertainty due to the limited accuracy of the physics models implemented in GEANT4 is almost exclusively related to that of the cross sections for the photo-electric, Compton and pair-production processes. The accuracy of these was estimated to be at a 5% level, as described in [15]. As for the previous case, the simulations have been repeated after separately changing the cross sections of each of the three processes by $\pm 5\%$. Given the considered energy range, the largest effect on the activity is induced by the cross section for Compton scattering and is $^{2.0}_{-0.6}$%. The systematic error related to the photo-electric effect is $^{+0.4}_{-0.2}$%, while the one related to the pair-production is below 1%.
The effect of the energy smearing applied to the simulated spectra is studied increasing and decreasing the FWHM at all energies by its uncertainty $\sigma_{FWHM}(E)$. Given that the entire spectrum, and not only the peak regions, is exploited for the fit, the contribution of the energy smearing to the systematic uncertainty on the activity is only at the order of 1‰ and can be neglected.

Finally, the uncertainty given by the choice of the energy range used for the analysis has to be considered. To quantify this effect, the analysis has been repeated by increasing the minimum of the energy range from 100 to 2600 keV with 20 keV steps. The root mean square (RMS) of the resulting activities distribution has been taken as systematic error. This results in 2.5%.

The three sources of systematic uncertainties considered so far originate from independent causes. Therefore, the errors are summed in quadrature, resulting in a $^{+4.4}_{-3.9}$% uncertainty in the activity. As a comparison, the activity of the GERDA Phase I sources was only known with a ±15% uncertainty [4]. Given the high precision obtained in the measurement, a more precise validation of the GERDA Phase II MC simulation is possible. In addition, the measurement of $^{56}$Co and $^{226}$Ra radioactive sources for GERDA Phase II will be performed using the same procedure.

4 Neutron strength measurement

The measurement of the neutron source strength of the four GERDA Phase II sources was performed with a LiI(Eu) detector and a $^3$He counter underground at LNGS in a low background environment. As both detectors are suitable for the detection of thermal neutrons, a moderator had to be used in order to thermalize the neutrons emitted by the $^{228}$Th sources. Three measurements were performed: first, the total detection efficiency was determined using an AmBe neutron source of known neutron strength, keeping the same geometrical configuration as for the $^{228}$Th sources screening. In a second step, the background spectrum was acquired for a period long enough to make its influence on the neutron source strength uncertainty smaller than the systematic error. Finally, the four GERDA Phase II sources were measured.

4.1 Measurement with LiI(Eu) detector

The first measurement of the neutron source strength was performed with a LiI(Eu) detector. The physical process exploited for the detection of thermal neutrons is:

$$^6\text{Li} + n \rightarrow ^7\text{Li}^* \rightarrow ^3\text{H} + ^4\text{He} + 4.78 \text{ MeV}$$  \hspace{1cm} (4.1)

The experimental signature is a peak at the Q-value of the reaction, i.e. 4.78 MeV. Given the different light yield induced by electrons with respect to alphas and tritons [16], the thermal neutron peak is recorded at ~ 4.1 MeV in electron-equivalent energy scale [17]. Since no environmental $\gamma$-ray is commonly present with an energy higher than 2615 keV (full energy peak of $^{208}$Tl), this assures a very good $\gamma$-n discrimination. A major concern for the detection of small thermal neutron fluxes is the presence of environmental thermal neutrons, as well as radioactive contaminants of the LiI(Eu) crystal bulk or its surface, emitting $\alpha$ or $\beta$ particles in the 3.5-4.5 MeV range.

The employed experimental setup consists of a LiI(Eu) cylindrical crystal with 25.4 mm diameter and 3 mm height enriched to 96% in $^6$Li provided by Scionix, Bunnik. The crystal is coupled to a 1” square R8250 low background photomultiplier tube (PMT) by Hamamatsu
Photonics. In order to minimize the background, the system is encased in a custom made copper holder (figure 2(a)). The detector is surrounded by 20 cm thick borated polyethylene (PE) panels acting as a shielding for external neutrons. A 2 cm thick lead brick and a 5 cm thick PE moderator were present in front of the detector during all measurements (figure 2(b)). The detector was designed in the past to measure the neutron emission rates of the $^{228}$Th prototype sources produced during Gerda Phase I. A detailed description can be found in [4].

The estimation of the total efficiency $\varepsilon$ was performed using an AmBe source with a neutron strength of $160 \pm 4 \text{ n/s}$ [18]. For this measurement only, the setup was installed at the surface laboratory at LNGS. The AmBe source was measured for 12.56 h, and a 5 days long background run was taken under the same conditions. The collected spectra are shown in figure 3. The analysis was performed via a maximum likelihood analysis in the 7000-9500 bin range, with $\varepsilon$ as the parameter of interest:

$$\ln L = \ln \left( \frac{k_s^{\lambda_s}}{\lambda_s^s k_s!} e^{-\lambda_s} \right) + \ln \left( \frac{k_b^{\lambda_b}}{\lambda_b^b k_b!} e^{-\lambda_b} \right) - \frac{(S - \mu_S)^2}{2 \cdot \sigma_S^2}$$

(4.2)

where $k_s$ and $k_b$ are the measured number of counts in the fit range for the AmBe and the background measurement, respectively, while $\lambda_s$ and $\lambda_b$ are the corresponding expectation values, and $S$ is the known AmBe neutron emission rate. A Gaussian prior is assigned to $S$, with $\mu_S = 160 \text{ n/s}$ and $\sigma_S = 4 \text{ n/s}$. The expectation values are defined as:

$$\lambda_s = \Delta t_s \cdot (\varepsilon \cdot S + B)$$

(4.3)

$$\lambda_b = \Delta t_b \cdot B$$

(4.4)
Figure 3. LiI spectra recorded with and without the AmBe source. The peak due to thermal neutrons is clearly visible around channel 8000, corresponding to about 4.1 MeV in electron-equivalent energy scale. For these measurements only, the LiI(Eu) detector was operated at the surface laboratory at LNGS.

Figure 4. Background of the LiI(Eu) detector operated underground at LNGS (red) and spectrum recorded with the four GERDA Phase II $^{228}\text{Th}$ sources (black). While the $\gamma$ contribution is only present up to channel $\sim 6000$, a thermal neutron signal is present around channel 8000. The faint peak in the background spectrum around channel 7000 is likely due to an internal $\alpha$ contamination of the crystal.

where $B$ is the background index in events/s in the fit range and $\Delta t_s\ (\Delta t_b)$ is the live time of the AmBe (background) measurement. The measured efficiency is:

$$\epsilon = \left(5.32^{+0.18}_{-0.15} \pm 0.27\right) \cdot 10^{-4} \ (4.5)$$

where the first error is the statistical uncertainty of the fit and the second is the systematic related to the choice of the energy range.


The four GERDA Phase II sources were measured together underground for 11 days, and a background measurement was carried out for 142 days. All the values reported here refer to the average values of the four sources. Figure 4 shows the two recorded spectra. The estimation of the neutron source strength was performed using the same method as for the efficiency. The log-likelihood contains a Gaussian prior for the total efficiency and the total activity $A = 130.4 \pm 5.5$ kBq of the four sources:

$$
\ln L = \ln \left( \frac{\lambda_s^k}{k_s!} e^{-\lambda_s} \right) + \ln \left( \frac{\lambda_b^k}{k_b!} e^{-\lambda_b} \right) - \frac{(\varepsilon - \mu_{\varepsilon})^2}{2 \cdot \sigma_{\varepsilon}^2} - \frac{(A - \mu_A)^2}{2 \cdot \sigma_A^2} \tag{4.6}
$$

where the subscripts $s$ and $b$ refer to the measurement with the sources and the background, respectively. The expectation values are given by:

$$
\lambda_s = \Delta t_s \cdot (\varepsilon \cdot A \cdot S + B) \tag{4.7}
$$

$$
\lambda_b = \Delta t_b \cdot B \tag{4.8}
$$

The resulting neutron source strength (in units of neutrons per second per Bq of gamma activity) is:

$$
S = \left(8.2^{+1.7}_{-1.3} \pm 1.1\right) \cdot 10^{-7} \, \text{n/(sec \cdot Bq)} \tag{4.9}
$$

The first uncertainty is the statistical error from the fit, while the second is the systematic uncertainty related to the choice of the energy range. This in particular includes the effect of the background peak present around channel 7000 (see figure 4).

The estimated neutron source strength is based on the assumption that the AmBe and $^{228}$Th emitted neutron spectra are the same. As mentioned in [4, 9], while the first has a mean energy at $\sim 4$ MeV, gold-encapsulated $^{228}$Th yields a mean neutron energy of 2.58 MeV. The use of the same total efficiency for the AmBe and $^{228}$Th measurements thus induces a systematic error, which was computed to be 12% in [9]. As a result, the neutron source strength for the GERDA Phase II calibration sources is:

$$
S = \left(8.2^{+1.7}_{-1.3} \text{(stat)} \pm 1.1(\text{fit range}) \pm 1.0(\text{eff})\right) \cdot 10^{-7} \, \text{n/(sec \cdot Bq)} \tag{4.10}
$$

For comparison, the same measurement and analysis was performed for a commercial $^{228}$Th source (i.e., with ceramic substrate) with 19.5 $\pm$ 2.9 kBq activity. The neutron source strength in this case is $7.5^{+2.5}_{-1.3} \cdot 10^{-6}$ n/(sec\cdot Bq). Thus, the use of gold as a support material for $^{228}$Th yields a reduction in the neutron emission rate by one order of magnitude.

### 4.2 Measurement with a $^3$He counter

A second measurement of the neutron source strength was performed with a $^3$He counter. The exploited reaction is:

$$
^3\text{He} + n \rightarrow ^1\text{H} + p + 746 \, \text{keV} \tag{4.11}
$$

The expected signal induced by thermal neutrons is therefore a peak at 746 keV. In spite of this low Q-value, $\gamma$ radiation does not represent a background for the measurement because of the gaseous state of the detector, with a mean energy deposit for electrons of about 2 keV/cm. The discrimination between neutrons and gammas or electrons is thus performed based on the deposited energy alone.
Figure 5. Background of the $^3$He counter operated underground at LNGS (red) and spectrum recorded with the four GERDA Phase II $^{228}$Th sources (black). The thermal neutron peak is visible around channel 2900, while the broad continuum below channel 2000 is due to γ background.

The $^3$He counter is a LND-2531 produced by LND inc., New York. The tube has an effective diameter of 47.75 mm and a height of 203.2 mm. It is filled with $^3$He at a pressure of 6078 mbar and is operated at 1950 V. The counter is inserted in a 6.5 cm thick $4\pi$ PE moderator, on which a borehole is present for source insertion. The distance between the tube and the bottom of the borehole is 1.1 cm. The system is shielded by a $^{113}$Cd foil and a $4\pi$ castle of borated paraffin with 10 cm thickness for the absorption of environmental neutrons.

The four sources were screened for 17.5 hours, while the background spectrum was acquired for 10 days. The total detection efficiency, determined using an AmBe source in a previous measurement, is $(7.13 \pm 0.85) \cdot 10^{-2}$ [19]. Following the same analysis procedure used for the LiI(Eu) measurements, the neutron source strength of the GERDA Phase II sources results in:

$$S = \left(9.4^{+2.0}_{-0.8} \text{(stat)} \pm 0.4 \text{(fit range)} \pm 1.1 \text{(eff)}\right) \cdot 10^{-7} \text{n/(sec} \cdot \text{Bq)} \quad (4.12)$$

where the first uncertainty is statistical, the second is the systematic due to the choice of the energy range, and the third is the efficiency related systematic.

4.3 Interpretation of results

The neutron source strengths measured with the LiI(Eu) and the $^3$He detector agree within the errors. This demonstrates the reliability of the LiI(Eu) as a low-flux neutron detector. For both measurements, the total uncertainty of $\sim 20\%$ is mostly due to the limited statistics. A higher precision could be obtained with a longer screening, but this was not possible due to time constraints related to the preparation for GERDA Phase II.

Previous measurements performed with two custom $^{228}$Th sources for GERDA Phase I lead to similar results [4]. The measured neutron source strengths vary between $7.59 \cdot 10^{-7}$ n/(sec·Bq) and $1.50 \cdot 10^{-6}$ n/(sec·Bq), depending on the source and detector. In this case the uncertainties
were $\sim 30\%$, which might explain the spread of the values by up to a factor two. A more precise measurement was performed in [9] on a $^{228}\text{Th}$ source with gold substrate and the same encapsulation of the Gerda sources. The measured neutron source strength of $(1.22 \pm 0.17) \cdot 10^{-6} \text{n/(sec-Bq)}$ is also in good agreement with the results presented here.

The residual neutron activity is attributed to $(\alpha, n)$ reactions in $^{17}\text{O}$ and $^{18}\text{O}$ isotopes bound to $^{228}\text{Th}$ after the evaporation on gold [4, 9]. Both the isotopes have a threshold for $(\alpha, n)$ reactions below 1 MeV. Other possible candidates are isotopes of zirconium, which is present in small amount ($\sim 10 \mu\text{g}$) in the original HCl solution [9]. Additional contributions might be given by other contaminants of the active material or the used chemicals, but their presence can be hardly assessed.

The agreement of independent measurements on the gold-encapsulated $^{228}\text{Th}$ sources confirms the reliability of the production technique. The reproducibility of $^{228}\text{Th}$ sources with neutron emission at the level of $10^{-6} \text{n/(sec-Bq)}$ can thus be guaranteed to present and future low background experiments. In the case of Gerda Phase II, the neutrons emitted by the calibration sources induce a background index at $Q_{\beta\beta}$ of $\sim 2 \cdot 10^{-4} \text{events/(keV-kg-yr)}$ prior to the application of PSD and LAr veto. This is a factor 5 lower than the total expected background and is sufficient to fulfill the Phase II requirements.

5 Leak test for calibration sources in cryogenic systems

A new set of leakage tests intended to check the tightness of $^{228}\text{Th}$ radioactive sources after deployment at cryogenic temperatures has been developed at the Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti of ENEA (ENEA-INMRI). It consists of a series of source insertions in acetone and liquid nitrogen, each one followed by a search for possible leaks. A blank source, ideally identical to the Gerda $^{228}\text{Th}$ sources but with no radioactive content, was used for a cross-check.

Each of the four radioactive sources and the blank source was enveloped in circular aluminum foil of 24 cm$^2$ surface, and dipped in acetone (A1) for 1 hour. After acetone evaporation, the $\alpha$ contamination of the aluminum foil was measured with a Berthold LB 770 $\alpha$-\$\beta$ low level counter. The operation was repeated using liquid nitrogen (LN) and then one more time with acetone (A2). The same procedure, without the second immersion in acetone, was applied without using any radioactive or blank source for background determination. While the background measurement is needed to estimate the intrinsic contamination of aluminum, the comparison between the $\alpha$ count rates obtained with the real and blank sources allow to disentangle between real $^{228}\text{Th}$ leaks and possible removable surface contamination. Since the blank and the background count rates are not distinguishable, they have been considered as belonging to the same population (BB).

The distribution of A1, LN and A2 count rates for each of the four sources was compared with that of BB. The test conditions and the results of the analysis are reported in table 2. Following [21], the decision threshold at 95% confidence level was calculated based on the standard deviation of the BB readings. The experimental standard deviation of the BB includes components of variability due to counting statistics, long term system stability and homogeneity of used materials. If the sample reading of the sources exceeds the decision threshold, a $^{228}\text{Th}$ leak has been detected.

Out of all the 24 LN readings only 2 (8.3%) exceed the decision threshold, while they are on average lower than the A1 and A2 readings. In addition, the A1 counting rates are always slightly higher than those of A2. This means that the higher LN counting rates is not induced by a $^{228}\text{Th}$ leak,
Table 2. Experimental conditions and results for the leak test performed at ENEA-INMRI.

|                              |         |
|------------------------------|---------|
| Number of BB readings        | 108     |
| Number of A1, LN, A2 readings for each source | 6       |
| Counting time [s]            | 6000    |
| Mean BB count rate [s^{-1}]  | 0.00250 |
| Experimental standard deviation of BB count rate [s^{-1}] | 0.00087 |
| Expected Poisson standard deviation [s^{-1}] | 0.00064 |
| Decision threshold for the net individual count rate [s^{-1}] | 0.00202 |

Table 3. Activity found after each source treatment, with reported 2σ uncertainties.

| Source   | A1 [mBq] | LN [mBq] | A2 [mBq] |
|----------|----------|----------|----------|
| AD9854   | 7.5 ± 3.5| 1.0 ± 6.3| 4.4 ± 3.8|
| AD9855   | 4.2 ± 4.6| 1.0 ± 6.1| 4.0 ± 3.0|
| AD9856   | 1.0 ± 2.9| -0.8 ± 2.7| -0.2 ± 3.6|
| AD9857   | 3.2 ± 3.8| 2.6 ± 6.4| 0.8 ± 3.9|

but from a surface contamination of the source capsules, which is removed by the two insertions in acetone.

The activity removed from each of the sources after the A1, LN and A2 immersions is reported in table 3. The efficiency of the counter was measured with a standard 241Am source with known activity (555.64 Bq) and is 0.3968. For all sources, no leak is detected after the insertion in LN and the second insertion in acetone. Hence the sources are suited to be used in GERDA Phase II.

6 Summary

Four low neutron emission 228Th source were produced for the calibration of the GERDA Phase II experiment. With a threshold of 9.94 MeV for (α, n) reactions, the deposition of the radioactive material on gold allows to minimize the parasitic neutron flux and allows to reduce the neutron source strength by one order of magnitude. The neutron emission rate was measured with a low background LiI(Eu) detector and a 3He counter, both located underground at LNGS. The neutron source strengths measured with the LiI(Eu) and the 3He detectors are 8.2 ± 2.3 n/(sec·Bq) and 9.4 ± 2.3 n/(sec·Bq), respectively.

The γ activity of the each source was measured with the Gator facility at LNGS. The high signal-to-background ratio, together with a detailed knowledge of the detector geometry, allowed to obtain a very good agreement between measured and simulated spectra, with a total uncertainty of ~ 4%.

A new, dedicated procedure was developed for the investigation of the source capsule tightness after the deployment in cryogenic environment, with a sensitivity to leaks of ≲ 10 mBq activity. The application of this technique to the GERDA Phase II calibration sources excluded any leaks and guarantees the tightness of the source capsules after the insertion in GERDA.
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