CUORE crystal validation runs: results on radioactive contamination and extrapolation to CUORE background

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Abstract

The CUORE Crystal Validation Runs (CCVRs) have been carried out since the end of 2008 at the Gran Sasso National Laboratories, in order to test the performances and the radiopurity of the TeO₂ crystals produced at SICCAS (Shanghai Institute of Ceramics, Chinese Academy of Sciences) for the CUORE experiment. In this work the results of the first 5 validation runs are presented. Results have been obtained for bulk contaminations and surface contaminations from several nuclides. An extrapolation to the CUORE background has been performed.
1. Introduction

The production of the CUORE [1] crystals was appointed to SICCAS (Shanghai Institute of Ceramics, Chinese Academy of Sciences) and began in March 2008, with the synthesis of the TeO₂ powder. Given the goal of CUORE in terms of background (<0.01 counts/keV/kg/y at the Q-value [2]), the radio-purity of the TeO₂ crystals is a crucial issue.

Radioactive contaminations may come from long-lived, naturally occurring isotopes, such as ²³⁸U and ²³²Th and their daughters and from cosmogenic activation of the detector materials after their production. To minimize the influence of long-lived nuclei, great care is devoted to the selection of all materials and ancillaries used for the preparation of the detector. Seal level transport and underground storage of prepared crystals are necessary in order to minimize their cosmogenic activation.

A dedicated protocol [3] is defined for the radio-purity related quality control of the crystal production process, starting from metallic tellurium synthesis to the final processing of ready-to-use TeO₂ crystals. Radio-purity certification procedures, involving ICP-MS (Inductively Coupled Plasma Mass Spectrometry) measurements, γ spectroscopy with HPGe detectors and α spectroscopy with Surface Barriers Detectors (SBD), are applied in each production phase to test the above mentioned materials.

At the same time, cryogenic tests are designed to test the ready-to-use TeO₂ crystals upon their arrival at LNGS. The crystal validation is performed through experimental runs, each called CCVR (CUORE Crystal Validation Run), in which 4 crystals randomly chosen from a batch coming from SICCAS, are mounted in an setup similar to a CUORE single module and operated at cryogenic temperatures for a time period of several weeks in order to test the bolometric performance and the compliance of the crystals to the contract limits in terms of radio-purity (see Table 1).

| Isotope | Allowed Contamination |
|---------|-----------------------|
| ²³⁸U    | < 3 · 10⁻¹³ g/g      |
| ²³²Th   | < 3 · 10⁻¹³ g/g      |
| ²¹⁰Pb   | < 1 · 10⁻⁸ Bq/kg     |
| ²¹⁰Po   | < 0.1 Bq/kg          |

Table 1: Contamination limits for the ready-to-use TeO₂ crystals [3].

In this work the results of the first 5 validation runs, carried out from the end of 2008 to the middle of 2010, are presented. A summary of all CCVRs detector operation is reported in Table 2. The 4 crystals tested in CCVR1 (two of them were again tested in CCVR2) were sent to LNGS by plane. This was necessary in order to ensure a fast response on the radioactivity level of the crystals. All the other crystals were transported by ship.

| Run     | Duration          | Livetime [d] |
|---------|-------------------|--------------|
| CCVR1   | Dec 20, 2008 - Mar 9, 2009 | 59.9         |
| CCVR2   | Jun 6, 2009 - Jun 30, 2009   | 19.4         |
| CCVR3   | Nov 11, 2009 - Jan 4, 2010  | 43.05        |
| CCVR4   | Mar 31, 2010 - May 17, 2010 | 25.8         |
| CCVR5   | Aug 11, 2010 - Oct 1, 2010  | 30.3         |

Table 2: Summary of CCVR data taking period and live time.

Radioactivity study was performed on the high energy region of the spectrum (above 4000 keV), where the contribution of the α lines from uranium and thorium decay chains is expected. Given the short range of α particles, their signature is a clear indication of a radioactive contamination within the crystals or on their surface. Results (mainly upper limits) are obtained for both bulk contaminations (Sec.5) and surface contaminations (Sec.7). For ²¹⁰Pb, for which the study of the α lines was not possible, the lower energy portion of the spectrum has been used (Sec.5).

An extrapolation to the CUORE background is performed in Sec.8.

2. Experimental setup

Each CCVR setup consists of an array of four crystals, arranged in a single floor which in a first approximation represents the single CUORE module. The four 5 × 5 × 5 cm³ crystals are enclosed in a pair of Oxygen Free High Conductivity (OFHC) copper frames which serve both as mechanical support and thermal bath. The two frames are connected to each other by four small columns, also made of copper. Both frames and copper are wrapped with several layers of polyethylene. The crystals are connected to the copper frames by small Teflon supports that represent the weak thermal conductance versus the heat sink.

Each crystal is provided with a Neutron Transmutation Doped Ge thermistor (NTD), which converts the phonon signal into a detectable voltage pulse [4]. Some of the CCVR crystals are provided with two NTD thermistors. In these cases both channels are analyzed and the best performing channel (from the point of view of the energy resolution) is used for the final results.

The CCVR experimental setup is hosted in a dilution refrigerator placed in the Hall C of National Laboratory of INFN at Gran Sasso and operated at ~ 10 mK. A complete description can be found in [5] and references therein for what concerns the electronics and the DAQ and in [6] for what concerns the cryogenic setup and the shields.

3. Data analysis

CCVR data processing, from raw data to the final spectra, follows the procedure described in details in [7].

The pulse amplitude is estimated by means of an Optimum Filter (OF) technique [8] that reduces the noise superimposed to the signal, maximizing the signal to noise ratio.
The mean detector response and the noise power spectral density, needed to build the OF transfer function, are estimated from bolometric pulses and noise samples (data samples recorded randomly and without triggered events) by a proper averaging procedure.

Gain instability corrections are performed using the 5407.5 keV $\alpha$ line from $^{210}$Po. This element is always present in recently grown crystals but decays away with a half life of 138.38 days.

The energy calibration is performed using $^{232}$Th $\gamma$ sources inserted inside the cryostat external lead shield. An example of calibration spectrum (Channel2 - CCVR1) is shown in Fig. 1. Gamma lines from the $^{232}$Th decay chain are clearly visible in the spectrum. The calibration is performed using a third-order polynomial function and the $^{210}$Po peak is used in addition to the $^{232}$Th $\gamma$ peaks.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Calibration spectrum of Channel2 - CCVR1. Gamma lines from the $^{232}$Th decay chain are visible in the spectrum.}
\end{figure}

Each CCVR sum spectrum is composed by events which survive two different type of data selection: global and event-based requirements.

Global requirements are applied following criteria decided a priori on the detector performances (an excessive noise level, ADC saturation, etc.). They identify bad time intervals that need to be discarded. These kind of cuts introduce a dead time that is accounted for by properly reducing the live time of the interested detector.

Event-based requirements comprise: pile-up rejection, pulse-shape and coincidence selection.

The presence of a pile-up prevents the OF algorithm from providing a correct evaluation of the pulse amplitude. The pile-up rejection is performed by imposing an extendable (paralyzable) dead window of 7 seconds to each event.

The pulse-shape analysis is used to reject non-physical events. The pulse shape parameters are the rise time and decay time of the OF-filtered waveform and other parameters that measure the deviation of filtered raw signal from the average detector response.

As a first step, each CCVR spectrum is corrected for the corresponding efficiency of the event-based cuts (from Table 1 and 2) and then the spectra are summed together. Four types of spectra are produced for all CCVRs:

- **Total Energy spectrum (TOT):** it contains all the general cuts and the pulse shape cuts.
- **Anti-coincidence Energy spectrum (M1):** it contains the events that caused an energy deposition in one crystal only.
- **Coincidence Energy spectrum (M2):** it contains the events that caused an energy deposition in two crystals.
- **Coincidence Sum Energy spectrum (M2sum):** it contains the sum energy of multiplicity 2 events. For instance in the case of an $\alpha$ decay on the surface of a crystal, both the energy of the $\alpha$ particle $E_1$ in the facing crystal and the corresponding nuclear recoil energy $E_2$ in the original crystal are detected. In the spectrum M2sum the variable $E_{TOT} = E_1 + E_2$ is plotted.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Energy spectra for the full CCVR statistics. Top: anti-coincidence and coincidence spectra. Bottom: Sum energy spectrum of multiplicity 2 events.}
\end{figure}

### 3.1. Efficiency of event-based cuts

Due to the high rate of $^{210}$Po events, there is a significant loss of efficiency due to pile-up rejection. The efficiency is estimated as:

$$\epsilon_{\text{pile-up}} = 1 - P_{\text{pile-up}} = e^{(-\tau T)}$$

(1)
where $P$ is the probability of a pile-up, $r$ is the counting rate of the events that passed the global cuts described above, and $T$ is the length of a time interval containing an event during which another event would be considered pile-up. The interval $T$ contains some time after the event during which a double pulse would result; some time before the event during which the event’s baseline would be spoiled by the tail of the preceding pulse. Properly, $T$ depends on the energy of the other event: the higher the energy of the other event, the longer its tail remains too large. The counting-rate is channel-dependent and sometimes also time-dependent. In the specific case of $^{210}$Po events, the counting-rate obviously decreases with time due to polonium decay (half-life: 138.38 days).

For the sake of simplicity, an average pile-up reduction efficiency for each CCVR is computed. This will apply to all channels in the full energy range and is calculated using formula (1) with $T = 7$ s and using as $r$ the global counting rate after the general cuts.

$$
\varepsilon_{\text{eff}} = \frac{r_{\text{MC}}}{r_{\text{full}}} = \frac{r_{\text{MC}}}{r_{\text{MC}} + \Delta r_{\text{MC}}}
$$

Table 3: Efficiencies of event-based cuts for each CCVR.

| Run   | $\varepsilon_{\text{pile-up}}$ | $\varepsilon_{\text{PSA}}$ | $\varepsilon_{\text{AC}}$ |
|-------|---------------------------------|------------------------------|---------------------------|
| CCVR1 | 0.84 ±0.01                       | 0.96±0.01                   | 0.987±0.003               |
| CCVR2 | 0.88 ±0.01                       | 0.98±0.01                   | 0.982±0.005               |
| CCVR3 | 0.89 ±0.01                       | 0.97±0.01                   | 0.990±0.002               |
| CCVR4 | 0.88 ±0.01                       | 0.94±0.02                   | 0.987±0.003               |
| CCVR5 | 0.89 ±0.01                       | 0.98±0.01                   | 0.990±0.003               |

The pulse shape cuts efficiency $\varepsilon_{\text{PSA}}$ is evaluated on the background peak at 2614.5 keV due to $^{208}$Tl, by a simultaneous fit on both the spectra of accepted and rejected events as detailed in [7].

The same procedure is applied for the evaluation of the anticoincidence cut efficiency $\varepsilon_{\text{AC}}$. In this case, instead of the 2614.5 keV line (which is usually in coincidence with other $\gamma$ lines), the photopeak at 1460.8 keV due to $^{40}$K is used. Results are summarized in Table 3.

### 3.2. Monte Carlo simulations

In order to extract meaningful information on the activity or contamination of a given nuclide in CUORE crystals from CCVRs data, it is necessary to rely on Monte Carlo simulations, capable of reproducing the main features of the detector geometry and response.

CCVRs simulations are performed with the GEANT4-based code developed by the CUORICINO ad CUORE collaboration and described in [9] and [10]. The simulation takes into account the energy resolution and the threshold of each detector. Properly, the events that passed the global cuts described above, and $T$ contains some time after the event during which a double pulse would result; some time before the event during which the event’s baseline would be spoiled by the tail of the preceding pulse. The counting-rate is channel-dependent and sometimes also time-dependent. In the specific case of $^{210}$Po events, the counting-rate obviously decreases with time due to polonium decay (half-life: 138.38 days).

The pulse shape cuts efficiency $\varepsilon_{\text{PSA}}$ is evaluated on the background peak at 2614.5 keV due to $^{208}$Tl, by a simultaneous fit on both the spectra of accepted and rejected events as detailed in [7].

The same procedure is applied for the evaluation of the anticoincidence cut efficiency $\varepsilon_{\text{AC}}$. In this case, instead of the 2614.5 keV line (which is usually in coincidence with other $\gamma$ lines), the photopeak at 1460.8 keV due to $^{40}$K is used. Results are summarized in Table 3.

| Run   | $\varepsilon_{\text{pile-up}}$ | $\varepsilon_{\text{PSA}}$ | $\varepsilon_{\text{AC}}$ |
|-------|---------------------------------|------------------------------|---------------------------|
| CCVR1 | 0.84 ±0.01                       | 0.96±0.01                   | 0.987±0.003               |
| CCVR2 | 0.88 ±0.01                       | 0.98±0.01                   | 0.982±0.005               |
| CCVR3 | 0.89 ±0.01                       | 0.97±0.01                   | 0.990±0.002               |
| CCVR4 | 0.88 ±0.01                       | 0.94±0.02                   | 0.987±0.003               |
| CCVR5 | 0.89 ±0.01                       | 0.98±0.01                   | 0.990±0.003               |

Table 3: Efficiencies of event-based cuts for each CCVR.

### 4. Results on background rates of CUORE crystals

From the energy spectra of all CCVRs the background rates in various energy regions can be calculated.

Six energy regions of interest are identified in the spectra and the corresponding count rates for anticoincidence (M1) and coincidence (M2) spectra are calculated. Results for the global spectra are reported in Table 4 (errors are statistical only).

| Energy Region | Count Rate (counts/kg/keV) |
|---------------|-----------------------------|
| (2700, 3200) keV | $^{100}$Pt (3200, 3400) | $^{208}$Po (3400, 3900) |
| M1             | 0.19 ±0.02                  | 0.38 ±0.04                  | 0.09 ±0.01                  |
| M1-PoSub       | 0.13 ±0.02                  | 0.34 ±0.04                  | 0.06 ±0.01                  |
| M2             | 0.05 ±0.01                  | 0.02 ±0.01                  | 0.02 ±0.01                  |
| M2-PoSub       | 0.02 ±0.01                  | 0.01 ±0.01                  | 0.008 ±0.008                |
| U/Th (4000, 5000) keV | $^{208}$Po (5000, 6000) | $^{208}$Po (6000, 8000) |
| M1             | 0.19 ±0.01                  | -                           | 0.057 ±0.004                |
| M1-PoSub       | 0.13 ±0.01                  | -                           | 0.057 ±0.004                |
| M2             | 0.04 ±0.01                  | -                           | 0.014 ±0.002                |
| M2-PoSub       | 0.014 ±0.007                | -                           | 0.014 ±0.002                |

Table 4: Count rates measured in (counts/kg/keV). Errors are statistical.

The continuum region (2700, 3200) keV is of great interest since it is the region immediately above the Q-value of the neutrinoless double beta decay of $^{130}$Te [7].

In the region (3200, 3400) keV a contribution of the $\alpha$ line from $^{100}$Pt is expected. This contamination is almost unavoidable for TeO$_2$ crystals, as explained in Sec. 5.

From 4000 to 8000 keV the contribution of the various $\alpha$ lines from U and Th decay chains is expected.

In between, there is the region (5000, 6000) keV, which is affected by the $^{210}$Po contamination (see Sec. 5.1). This produces not only a peak at the energy of the Po $\alpha$ line 5407.5 keV but also a broad background over the entire region due to mis-identified pile-up events (above the peak energy) or to the escape of the $\alpha$ that releases part of its energy in an inert material (below the peak energy). An indication of the rate in this region is of no particular interest, also because of the relative short...
half-life of $^{210}$Po (138.38 days) that guarantees a huge reduction of this count rate when CUORE will start the data taking.

4.1. Subtraction of $^{210}$Po induced rate

Because of the presence of $^{210}$Po, an excess of count rate could arise in the M1 and M2 spectra below the energy of the 5407.5 keV $\alpha$ line, if the contamination of $^{210}$Po is close enough to the surface for the $\alpha$ to escape and release part of its energy in an inert material (M1 spectrum) or in a nearby detector (M2 spectrum). This contribution from the M2 spectrum can be estimated, calculating for each energy region the rate of M2 events in which the total energy $E_{\text{TOT}}$ lies in the interval (5407.5±50) keV. The M2 count rate subtracted for this contribution is defined as M2-PoSub and reported in Table 4 for comparison.

In a similar way the contribution of surface $^{210}$Po in the M1 spectrum is evaluated. Because of the geometry of the CCVR setup, our coincidence analysis is sensitive only to 8 faces over 24. So the anti-coincidence count rate due to $^{210}$Po of the remaining 16 faces is assumed to be two times the coincidence rate vs. time behaviour (see Sec. 5.1). The notation (2700, 3900) refers to the combination of both the continuum region (2700, 3200) keV and (3400, 3900) keV, excluding the $^{190}$Pt energy region of (3200, 3400) keV. It can be inferred that:

- in the region (2700, 3900) keV the CCVRs anti-coincidence rate is compatible within 1.8 $\sigma$ with the corresponding TTT value.
- in the region (4000, 5000) keV, as already measured in the TTT run, a reduction in the count rate with respect to Cuoricino is observed.
- in the region (5000, 6000) keV a comparison is not possible since CCVR is affected by the high rate of $^{210}$Po. This is due to the fact that in CCVRs only recently grown crystals are measured, unlike TTT or Cuoricino.
- in the region (6000, 8000) keV, as already measured in the TTT run, a reduction in the count rate with respect to Cuoricino is present. However, the CCVR count rate is greater than the TTT value, probably because of the presence of mis-identified pile-up $(M1)$ or coincidences $(M2)$ with $^{210}$Po events, extending above 6000 keV. This contribution should decay away with $^{210}$Po.

A more detailed evaluation of the comparison can be found in [11].

4.2. Background rates comparison with previous TeO$_2$ detectors

It is interesting to compare CCVRs rates with the ones obtained with previous detectors, like TTT [11] or Cuoricino [7]. This comparison is shown in Table 5 where the M1 and M2 count rates for CCVRs are after polonium subtraction, as described in [11].

|                | Continuum (2700, 3900) [keV] | U/Th (4000, 5000) [keV] |
|----------------|---------------------------|------------------------|
|                | M1                        | M2                     | M1                         | M2                     |
| CCVR           | 0.09±0.02                 | 0.13±0.01              | 0.015±0.007                | 0.014±0.003 |
| TTT            | 0.052±0.008               | 0.28±0.02              | 0.009±0.003                | 0.0018±0.0005 |
| Cuoricino      | 0.104±0.002               | 0.522±0.003            | 0.009±0.001                | 0.084±0.001 |

Table 5: Count rate comparison with previous detectors, measured in [counts/keV/kg/y]. Here CCVR values are after polonium subtraction, as described in [11]. Errors are statistical.

5. Results on bulk contaminations of CUORE crystals

Bulk contaminations in CUORE crystals are expected from:

- $^{210}$Po, as a result of the chemical affinity between polonium and tellurium;
- natural contaminants, like $^{238}$U and $^{232}$Th with their radioactive chains;
- $^{210}$Pb, as a result of the deposition of lead nuclei produced by $^{222}$Rn decays, during the crystal handling in free atmosphere;
- $^{190}$Pt, due to the fact that platinum is used in several phases of the crystal production cycle. Platinum crucibles are used for the calcination of TeO$_2$ powder used for the crystal growth and the growth crucibles are made of platinum foil. The central part of the as-grown crystal ingot is selected for the CUORE crystals in order to avoid the risk of platinum contamination on the surface of the crystal due to possible diffusion during the growth process. Possible Pt contaminations are therefore in the bulk of the CUORE crystals.

For $^{210}$Po the activity is determined from a fit to the anti-coincidence rate vs. time behaviour (see Sec. 5.1). For $^{238}$U and $^{232}$Th (see Sec. 5.2), the limit on the level of contamination is determined from the intensities of the $\alpha$ peaks in the anti-coincidence spectrum of all crystals, or from integrals centered at the peak position if the peaks are not visible.
This is because an α decay from bulk contamination releases the entire Q-value of the reaction (α energy + nuclear recoil) in a single crystal.

For 210Pb, the limit on bulk contamination is determined from a fit in the energy region (40, 60) keV in a subset of CCVRs data with high statistics and low threshold (see Sec. 6).

5.1. 210Po bulk activity

The activity of 210Po can be measured from the intensity of 5407.5 keV α line in the anti-coincidence spectrum.

The plot in Fig. 3 shows the global rate of 210Po events over time for CCVR1. For each channel the 210Po events are selected in a ± 20 keV window around the energy of the α line.

![Figure 3: Global rate of 210Po events over time for each CCVR1. The fit result is overlayed.

The units on the x-axis are days since the start of the first background measurement. Each point represents a group of measurements whose livetime is at least 5 days.

The horizontal error bars indicate the beginning and the end of each group of runs, they are for visualization only.

Each point is corrected with the corresponding rate-based efficiency, calculated with the equation (1). A larger dead time window (9 seconds instead of 7 seconds) compared to the rest of the analysis is used, to be more conservative in the removal of pile-up pulses.

The fit function is a pure exponential:

\[ r(t) = r_0 e^{-\frac{t}{T_{1/2}}} \]  

(3)

where \( r_0 \) is the rate at the beginning of the measurement and \( T_{1/2} \) is the 210Po half-life. The half-life of the exponential decay has been evaluated for all CCVRs and is shown in Tab 6. It is in good agreement (1σ) with the half-life of 210Po (138.38 d). This indicates that the 210Po contamination is out of equilibrium and it is not being fed by 210Pb.

From the value of \( r_0 \) returned by the fit for each CCVR and for each crystal, the 210Po activity at the beginning of the measurement is extracted as follows:

\[ A[\text{Bq/kg}] = \frac{r_0}{86400[\text{s/day}]} m[\text{kg}] \]  

(4)

where \( m \) is the crystal mass.

Knowing the time elapsed since the “crystal birth date” (growth completed, before the cut and shape) and the start of the measurement, the 210Po activity at production is computed.

Table 6: 210Po half-life for all CCVRs. The activity has been fitted using an exponential function. All the values are consistent (within 1σ) with 138.38 days.

| CCVR  | Crystal | 210Po activity [Bq/kg] |
|-------|---------|------------------------|
| CCVR1 | 041     | 0.0257 ± 0.00001       |
|       | 011     | 0.0510 ± 0.0005        |
|       | 039     | 0.0229 ± 0.0001        |
|       | 007     | 0.0414 ± 0.0004        |
| CCVR2 | 076     | 0.021 ± 0.004          |
|       | 011     | 0.07 ± 0.02            |
|       | 096     | 0.055 ± 0.006          |
|       | 007     | 0.047 ± 0.015          |
| CCVR3 | 190     | 0.0078 ± 0.0005        |
|       | 236     | 0.0159 ± 0.0005        |
|       | 180     | 0.0203 ± 0.0008        |
|       | 229     | 0.0283 ± 0.0008        |
| CCVR4 | 340     | 0.032 ± 0.004          |
|       | 313     | 0.005 ± 0.001          |
|       | 354     | 0.039 ± 0.004          |
|       | 380     | 0.040 ± 0.004          |
| CCVR5 | 455     | 0.019 ± 0.002          |
|       | 416     | 0.024 ± 0.003          |
|       | 436     | 0.032 ± 0.003          |
|       | 421     | 0.020 ± 0.003          |

Table 7: 210Po activity at production time for all CCVRs crystals.

5.2. U/Th bulk contaminations

238U and 232Th bulk contaminations are evaluated from the anti-coincidence sum spectrum of all CCVRs. For each of the peaks reported in Table 8 the number of counts (corrected by the efficiency of the event-based cuts from Table 3) within an energy window of ±6 σ around the α Q-value is estimated. A σ of 2.2 keV, corresponding to the average between the values of the 210Po peak σs for each CCVR, weighted by the corresponding lifetime of that run (see Table 9 for details) is used. Using the Bayesian approach, the upper limits \( N_u \) at 90% C.L.
Table 8: 90% C.L. limits on the number of events ascribed to several nuclides from uranium and thorium decay chain. For each nuclide is also shown the Q-value and the half-life of the α decay.

Table 9: Summary of CCVRs energy resolutions, evaluated with a gaussian fit.

Table 10: Upper limits at 90% C.L. on the activity and on the bulk contamination of uranium and thorium decay chains in the hypothesis of secular equilibrium.

| Run       | Livetime [days] | σ [keV] |
|-----------|-----------------|---------|
| CCVR1     | 59.9            | 1.6     |
| CCVR2     | 19.4            | 1.4     |
| CCVR3     | 43.03           | 2.2     |
| CCVR4     | 25.8            | 3.5     |
| CCVR5     | 30.3            | 2.8     |

where \( \epsilon_{MC} \) is the Monte Carlo detection efficiency, \( T \) the CCVRs livetime, \( m \) the crystal mass and \( \Gamma \) the branching ratio of the nuclide. In this analysis a total contamination of the anti-coinccide events in the crystals is assumed \( (\epsilon_{MC} = 1) \). Results are shown in Table 10.

The upper limit on the activity for each nuclide is calculated using the following formula:

\[
A_u [\text{Bq/kg}] = \frac{N_u}{\epsilon_{MC} T [\text{s}] m [\text{kg}] \Gamma} \tag{5}
\]

where \( \epsilon_{MC} \) is the Monte Carlo detection efficiency, \( T \) the CCVRs livetime, \( m \) the crystal mass and \( \Gamma \) the branching ratio of the nuclide. In this analysis a total contamination of the anti-coinccide events in the crystals is assumed \( (\epsilon_{MC} = 1) \). Results are shown in Table 10.

6. 210Pb activity

During the production of CUORE crystals great care is devoted in order to minimize the exposure of the crystals to free atmosphere, to avoid contamination of radon and its daughters. Nevertheless, a small 210Pb contamination (bulk or surface) might occur. This contamination can be very dangerous because, even if the soft beta (16.96 keV and 63.5 keV) and γ (46.5 keV) radiation from 210Pb is generally self absorbed, the induced bremsstrahlung of the daughter 210Bi can give rise to a continuum up to 1.16 MeV. Moreover, the α decay of the daughter 210Bi can contribute to the continuum background in the double beta decay energy region.

In CCVRs crystals the 210Pb contamination cannot be estimated from 210Po, because this contamination is out of equilibrium (see Sec. 5.1). The only available signature is a combination of a beta spectrum (end point 16.96 keV) with a de-excitation energy of 46.5 keV (most of the cases through a conversion electron). This produces a broad signature in the energy region (40, 60) keV, whose shape depends on the location of the Pb contamination.

Several simulations are performed both for a bulk contamination and for a surface contamination with exponential density profile \( [9] \) and contamination depth varying from 0.01 µm to 10 µm. Even if, for the sake of simplicity, a common energy threshold of 50 keV is set for all CCVRs, there are subset of data where the threshold can be set to a lower value in order to look for this signature. CCVR1 data are used because they struck a balance between high livetime (CCVR1 has the highest statistic) and good bolometer performances. Only for CCVR1 data, the analysis is repeated with an energy threshold of 40 keV. The anti-coinccide spectrum between 40 and 60 keV is fitted with an exponentially decreasing background and allowing the presence of a 210Pb spectrum with shape taken from Monte Carlo simulation. The free parameters of the fit are the two parameters of the exponential background and the total number

Each nuclide are entirely due to a bulk contamination of that nuclide. This is a conservative hypothesis since there is not a clear indication (for example a line) that such a bulk contamination actually exists and that the observed counts are not due to background of some other origin.
of counts from $^{210}$Pb. The fit is repeated for each Monte Carlo signature. The number of $^{210}$Pb events is compatible with zero within the error for all signatures. An example of Monte Carlo spectrum for a $^{210}$Pb bulk contamination and the corresponding fit to the experimental spectrum is shown in Fig. 4. The upper limit at 90% C.L. on the number of $^{210}$Pb counts is $N_e = 1.644 \sigma$, where $\sigma$ is the error on the number of counts returned from the fit. The upper limit on bulk contamination is computed using Eq. 5. The upper limit on surface contamination is extracted using the following formula:

$$A_n[\text{Bq/cm}^2] = \frac{N_e}{\epsilon_{MC} T[S S][\text{cm}^2]}$$

(6)

where $\epsilon_{MC}$ is the Monte Carlo detection efficiency, $T$ is the live time, and $S$ is the surface of a crystal (150 cm$^2$). Results are shown in Table 11. The upper limit on the activity for bulk contamination is below the contract limit of $10^{-5}$ Bq/kg (see Table 12).

7. Results on surface contaminations of CUORE crystals

Surface contaminations of CUORE crystals are expected to come from the same nuclides listed in Sec. 5 except for $^{190}$Pt, which is all in the bulk. Uranium and thorium surface contaminations can be investigated by means of coincident events in two facing crystals: surface contamination of an $\alpha$-decaying nuclide should appear in the total energy spectrum (M2sum in Fig. 2) as a peak at the Q-value of the decay because the total energy ($\alpha + \text{nuclear recoil}$) is collected by the two facing crystals. At the same time, if the surface contamination is deep enough, the $\alpha$ particle can be absorbed by the crystal, giving rise to the same signature of a bulk event.

| Isotope | Energy [keV] | Energy + $5407.5$ [keV] |
|---------|--------------|--------------------------|
| $^{127}$Te | 88.3 | 5495.8 |
| $^{129}$Te | 105.5 | 5513 |
| $^{125}$Te | 144.8 | 5552.3 |
| $^{123}$Te | 247.5 | 5655 |
| $^{212}$Bi | 294 | 5701.5 |

Table 12: Energy of the gamma lines from Tellurium metastable isotopes and corresponding sum energy when in coincidence with a $^{210}$Po event.

Because of the presence of these coincidences, only nuclides with a Q-value lower than 5407.5 keV (see Table 13) are used for surface contamination analysis. $^{210}$Po and $^{212}$Bi are therefore discarded. The scatter plot of M2 events is shown in Fig. 6. The shadowed region contains the events with a total energy within 4 and 5 MeV.

For each nuclide listed in Table 13, the number of counts is computed from the M2sum energy spectrum in an energy window of $\pm 6 \sigma$ around the Q-value and divided by $\epsilon^2$ (where $\epsilon$ is the efficiency of the event-based cuts of Table 5), since two coincident events have independent probabilities of passing the cuts. In Table 13 are shown the corresponding upper limits at 90% C.L., computed using the Bayesian approach with 0 expected background counts and a flat prior for the signal. Monte Carlo spectra for $^{238}$U and $^{232}$Th contamination on the crystal surface are generated with exponential profile and various penetration lengths. For each contamination depth, the containment efficiency is calculated in a $\pm 6 \sigma$ interval around the Q-value of each nuclide both for the M2sum spectrum and for the M1 spectrum. Results are shown in Fig. 7. For penetration length of 0.01 and 0.1 $\mu$m the higher containment efficiency comes from the M2sum spectrum, whereas for depths of 1, 5 and 10 $\mu$m this arises from the M1 spectrum.
Figure 5: Top: M2sum spectrum in the region (5300, 6000) keV. Peaks originated by the coincidence of the 5407.5 keV α line and a low energy γ line from Te metastable isotopes are clearly visible. Bottom: total and anti-coincidence spectra in the region 0-500 keV: low energy lines from Te metastable isotopes are clearly visible.

Figure 6: Scatter plot of M2 events. The shadowed region contains events with total energy between 4 and 5 MeV, used for surface analysis.

Table 13: Upper limits at 90% C.L. on the number of counts ascribed to several nuclides from uranium and thorium decay chain from the M2sum spectrum. For each nuclide, the half-life of the α decay is also shown.

| Chain | Nuclide | Half-life [y] | Energy range [keV] | \(N_u\) 90% C.L. |
|-------|---------|--------------|--------------------|-------------------|
| 238U  | 238U    | 4.47E+09     | 4257 - 4283        | 4.6               |
| 234U  | 2.45E+05 | 4845 - 4871  | 4.6                |
| 230Th | 7.54E+04 | 4757 - 4783  | 4.6                |
| 228Ra | 1599    | 4857 - 4883  | 10.5               |
| 228Th | 1.4E+10 | 4069 - 4095  | 2.3                |

Table 14: Upper limits at 90% C.L. on the number of counts ascribed to several nuclides from uranium and thorium decay chain from the M2sum spectrum. For each nuclide, the half-life of the α decay is also shown.

Figure 7: For each penetration length, the containment efficiency of surface events is computed for both M1 and M2sum Monte Carlo spectra.

containment value for 0.2 µm depth is very similar both for M1 and M2sum spectrum.

The drop in M1 containment efficiency for very thin layers of contaminations is due to the fact that, of the about 50% of decays where the alpha is emitted toward the bulk of the crystals and the recoil is emitted towards outside, in most of the cases the recoil escapes undetected, thus the full Q-value of the alpha decay is not recorded.

The upper limits at 90% C.L. for the surface activity of each nuclide are evaluated using Eq. 6 where \(N_u\) is the 90% C.L. upper limit on the number of observed events from the M2sum or the M1 spectrum (see Table 13 for the M2sum counts and Table 8 for the M1 counts) and \(\epsilon_{MC}\) is the Monte Carlo average efficiency defined in eq. 2 for the corresponding spectrum (M2sum or M1) and for the given signature. The confidence intervals for surface activity contaminations are calculated for both the M2sum and M1 spectra, normalizing each signature with the corresponding Monte Carlo efficiency. The signature giving the most stringent result is taken into account.

For surface contaminations of 0.01, 0.1 and 0.2 µm depths, the most stringent limits come from the M2sum spectrum. For the remaining depths (1, 5 and 10 µm) the surface activity reduces practically to a bulk activity and the M1 signature produces the most stringent limits (see Table 8 for details).

For the 238U chain, the surface contamination for 2 peaks is evaluated:

- 238U, the chain parent;
- 226Ra, the most active line both in M1 and M2sum spectra.

The contribution from 210Pb is treated separately (see Sec. 6).

For the 232Th chain there is only one useable line for this analysis, that is the one from 232Th. This means that there is no way of testing the portion of the chain below 220Rn and take into account a possible non-equilibrium of the chain, as done for the bulk contamination.

The results for surface contaminations are shown in Table 14. As explained in Sec. 5.2 the above confidence intervals are calculated under the hypothesis that the observed counts for each
nuclide are entirely due to a surface contamination of that nuclide in the corresponding depth. Again, this is a very conservative hypothesis.

| Depth | Nuclide | Upper limit 90% C.L. |
|-------|---------|----------------------|
|       |         | [Bq/cm²]             |
| 0.01µm| ²⁵⁹U    | 3.1E-09              |
|       | ²²⁶Ra   | 6.3E-09              |
|       | ²³²Th   | 1.6E-09              |
| 0.2µm | ²⁵⁹U    | 3.8E-09              |
|       | ²²⁶Ra   | 7.6E-09              |
|       | ²³²Th   | 2.0E-09              |
| 1µm   | ²⁵⁹U    | 3.7E-09              |
|       | ²²⁶Ra   | 8.9E-09              |
|       | ²³²Th   | 1.9E-09              |
| 5µm   | ²⁵⁹U    | 2.0E-09              |
|       | ²²⁶Ra   | 5.4E-09              |
|       | ²³²Th   | 1.0E-09              |
| 10µm  | ²⁵⁹U    | 1.7E-09              |
|       | ²²⁶Ra   | 4.4E-09              |
|       | ²³²Th   | 8.3E-10              |

Table 14: Upper limits at 90% C.L. for surface contamination, for different penetration length values. See text for details on the calculation of confidence intervals.

8. Extrapolation to CUORE background

In order to evaluate the contribution to the CUORE background arising from crystal contamination, a simulation both for bulk and surface contamination reproducing the CUORE geometry and studying the contribution to the double beta decay (DBD) energy region is performed.

Being interested in a conservative upper limit to the CUORE background, in this extrapolation it has been assumed that CUORE crystals will have the same activity as crystals tested in CCVRs. However, when CUORE will start the data taking, most of the ²¹⁰Po (and other short lived states) will have decayed and the efficiency for pile-up rejection will be increased.

8.1. Background from bulk contamination

The CUORE geometrical efficiency for a uniform bulk contamination of crystals in ²¹⁰Pb, ²³⁸U and ²³²Th is estimated via Monte Carlo, and the corresponding CUORE background at the DBD energy region (Q-value = 30 keV) is calculated using the formula:

\[ \text{bkg}_{\text{CUORE}} = \frac{A_{\text{CCVR--bulk}} \cdot \varepsilon_{\text{CUORE--bulk}}}{\Delta E} \cdot M_{\text{CUORE}} \]

(7)

where \(A_{\text{CCVR--bulk}}\) are the values of ²¹⁰Pb, ²³⁸U and ²³²Th activities from Tables [10] and [11] \(\varepsilon_{\text{MC}}\) is evaluated from Monte Carlo simulation and \(\Delta E = 60 \text{ keV}\).

The results are shown in Table [15]

In the most conservative approach, considering the most active line (²¹²Bi), the following upper limit to the CUORE background at the DBD energy due to bulk contamination of crystals is set: \(1.1 \cdot 10^{-4}\) counts/keV/kg/y.

| chain   | Nuclide  | Upper Limit 90% C.L. |
|---------|----------|----------------------|
|         |          | [counts/keV/kg/y]    |
| ²¹⁰Pb   |          | 2.6E-05              |
| ²³⁸U    |          | 8.1E-07              |
| ²³⁴U    |          | 1.4E-06              |
| ²³⁰Th   |          | 1.7E-06              |
| ²²⁶Ra   |          | 2.1E-06              |
| ²¹⁹Po   |          | 5.1E-07              |
| ²³²Th   |          | 1.7E-05              |
| ²¹²Bi   |          | 1.1E-04              |

Table 15: Extrapolation to CUORE background from CCVRs bulk contamination limits from Tabs. [10] and [11]

8.2. Background from surface contamination

In a similar way, the CUORE geometrical efficiency for a surface contamination for several depths is estimated. The corresponding CUORE background is extrapolated using the formula:

\[ \text{bkg}_{\text{CUORE}} = \frac{A_{\text{CCVR--surf}} \cdot \varepsilon_{\text{CUORE--surf}}}{\Delta E} \cdot M_{\text{CUORE}} \]

(8)

where \(A_{\text{CCVR--surf}}\) are the surface contamination values from Tables [11] and [14] \(\varepsilon_{\text{MC}}\) is estimated via Monte Carlo, \(S\) is the surface of the CUORE crystals, \(\Delta E = 60 \text{ keV}\) and \(M_{\text{CUORE}} = 0.75 \cdot 988 \text{ kg}\).

| Depth | Nuclide  | Upper limit 90% C.L. |
|-------|----------|----------------------|
|       |          | [counts/keV/kg/y]    |
| 0.01µm| ²¹⁰Pb    | 1.6E-05              |
|       | ²³⁸U     | 8.9E-05              |
|       | ²²⁶Ra    | 1.8E-04              |
|       | ²³²Th    | 1.8E-05              |
| 0.2µm | ²¹⁰Pb    | 2.3E-04              |
|       | ²³⁸U     | 3.9E-04              |
|       | ²²⁶Ra    | 7.7E-04              |
|       | ²³²Th    | 1.5E-04              |
| 1µm   | ²¹⁰Pb    | 3.8E-04              |
|       | ²³⁸U     | 1.2E-03              |
|       | ²²⁶Ra    | 2.9E-03              |
|       | ²³²Th    | 4.4E-04              |
| 5µm   | ²¹⁰Pb    | 4.3E-04              |
|       | ²³⁸U     | 1.3E-03              |
|       | ²²⁶Ra    | 3.4E-03              |
|       | ²³²Th    | 4.7E-04              |
| 10µm  | ²¹⁰Pb    | 2.9E-04              |
|       | ²³⁸U     | 7.1E-04              |
|       | ²²⁶Ra    | 1.9E-03              |
|       | ²³²Th    | 3.0E-04              |

Table 16: Extrapolation to CUORE background from CCVRs surface contamination limits from Tables [11] and [14]
For each penetration depth, the upper limits at 90\% C.L. for the count rate at the DBD energy region are plotted. The values are computed on $^{226}$Ra line, and are due only to surface crystals contaminations.

$5.5 \times 10^{-3}$ counts/keV/kg/y. This value corresponds to the worst case: a contamination in $^{238}$U and $^{232}$Th at 5\,\mu m and a contamination in $^{210}$Pb at 0.01\,\mu m. This result is strictly dependent on the density profile assumed for the contaminants distribution, as can be seen in Fig. 8 where the upper limit at the 90\% C.L. for the rate in the DBD energy region (computed on $^{226}$Ra line) is plotted in function of contamination depth.

**Conclusions**

The CUORE Crystal Validation Runs (CCVRs) have been carried out since the end of 2008 at the Gran Sasso National Laboratories, in order to test the performances and the radio-purity of the TeO$_2$ crystals produced at SICCAS (Shanghai Institute of Ceramics, Chinese Academy of Sciences) for the CUORE experiment.

In this work the results of the first 5 validation runs, are presented. The items under analysis are the background rate, the bulk and surface contamination from several nuclides and the extrapolation to the CUORE background.

The CCVRs background rate shows a reduction with respect to the one measured in CUORICINO in all the energy regions considered. The bulk activity of $^{210}$Po is measured to be within the limit specified in the contract with the crystals producer. No indication of a bulk contamination from uranium and thorium decay chains, as well as from $^{210}$Pb (out of equilibrium), is found. The upper limits are calculated to be within the contract specification. No indication of a surface contamination from uranium and thorium decay chains, as well as from $^{210}$Pb (out of equilibrium), is found. Upper limits at 90\% C.L. are calculated for surface contamination from several nuclides and for different contamination depths.

An extrapolation to CUORE background from both bulk and surface contaminations is performed, in the most conservative assumption that the CCVRs observed background is entirely due to the bulk and the surface contamination respectively. Considering only the contribution from bulk and surface crystals contaminations, the following upper limits to the CUORE background index in the energy region around the Q-value of the neutrinoless double beta decay of $^{130}$Te are calculated:

$1.1 \times 10^{-4}$ counts/keV/kg/y and $5.5 \times 10^{-3}$ counts/keV/kg/y respectively.

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