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Radioactive contamination of $ZnWO_4$ crystal scintillators

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ABSTRACT

The radioactive contamination of $ZnWO_4$ crystal scintillators has been measured deep underground at the Gran Sasso National Laboratory (LNGS) of the INFN in Italy with a total exposure 3197 kg h. Monte Carlo simulation, time–amplitude and pulse–shape analyses of the data have been applied to estimate the radioactive contamination of the $ZnWO_4$ samples. One of the $ZnWO_4$ crystals has also been tested by ultra-low background γ spectrometry. The radioactive contaminations of the best $ZnWO_4$ samples are estimated to be less than 0.002 mBq/kg (^{228}Th and ^{226}Ra), the total α activity is 0.18 mBq/kg. The β active ^{65}Zn and the α active ^{180}W have been detected in $ZnWO_4$ crystals. The effect of the re-crystallization on the radiopurity of the $ZnWO_4$ crystal has been studied. The radioactive contamination of the ceramic components of the set-ups used in the crystals growth has been checked by low background γ spectrometry. Some ideas for future improvement of the radiopurity level of $ZnWO_4$ crystal scintillators are briefly discussed.

1. Introduction

The luminescence of zinc tungstate ($ZnWO_4$) was studied sixty years ago [1]. Large volume $ZnWO_4$ single crystals of comparatively high quality were grown [2] and studied as scintillators in the eighties [3]. The main characteristics of the $ZnWO_4$ scintillators are given in Table 1. The material is non-hygroscopic and chemically resistant. The use of $ZnWO_4$ scintillators was proposed to search for double beta decay in Ref. [4] for the first time. The first low background measurement with a small $ZnWO_4$ sample (mass of 4.5 g) was performed in the Solotvina Underground Laboratory (Ukraine) at a depth of $\approx$ 1000 m of water equivalent (mwe) in order to study its radioactive contamination, and to search for double beta decay of zinc and tungsten isotopes [5]; the possibilities to use $ZnWO_4$ crystals in the field of dark matter were also discussed. The luminescence of $ZnWO_4$ down to helium temperature was studied in Ref. [6] and subsequently investigations of $ZnWO_4$ crystals as scintillating bolometers have recently been performed [7–9].

The radioactive contamination of a 119 g $ZnWO_4$ scintillator was measured to be in the mBq/kg level, in the Solotvina Underground Laboratory [16,9]. Long-time low background scintillation measurements using several $ZnWO_4$ crystal scintillators (with mass in the range 0.1 – 0.7 kg) have been performed at the LNGS with the aim to search for double β processes in zinc and tungsten isotopes [17–19]. The data collected with different $ZnWO_4$ crystals in the same set-up also allow the estimation of the level of the radioactive contamination of the material. One sample has also been tested by ultra-low background HP Ge γ spectrometry. The effect of the re-crystallization procedure on the radioactive contamination of this material has also been investigated. Moreover, a few samples of ceramics, the most contaminated components in the set-ups used in the crystals growth, have also been measured by a low background HP Ge detector.

2. $ZnWO_4$ crystal scintillators

Four clear, slightly colored $ZnWO_4$ crystal scintillators have been used in the present study. All the samples are listed in Table 2.

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Table 1
Properties of the $ZnWO_4$ crystal scintillators.

Density (g/cm^3)	7.87 [2]
Melting point (°C)	1200 [2]
Structural type	Wolframite [10–13]
Cleavage plane	Marked (010) [14]
Hardness (Mohs)	4–4.5 [15]
Wavelength of emission maximum (nm)	480 [1,2,15]
Refractive index	2.1–2.2 [15]
Effective average decay time [a] (μs)	24 [5]

[a] For γ rays, at room temperature.

The samples ZWO-1 and ZWO-2 were produced in the Institute for Scintillation Materials (ISMA, Kharkiv, Ukraine) from crystal ingots grown in platinum crucibles by the Czochralski method [16,20]. The $ZnWO_4$ compounds used to grow the crystals were synthesized from two batches of zinc oxide from different producers. The crystal ZWO-3 was obtained by re-crystallization from the sample ZWO-2 at the ISMA. The sample ZWO-4 was produced in the Nikolaev Institute of Inorganic Chemistry (Novosibirsk, Russia) by the low-thermal gradient Czochralski technique in a platinum crucible [21].

3. Measurements

3.1. Low background scintillation measurements

The measurements (see Table 2) have been carried out in the DAMA/R&D set-up [17,18,22] at the LNGS having average overburden of about 3600 mwe. In each measurement the $ZnWO_4$ crystal was fixed inside a cavity of $\varnothing 49 \times 59$ mm in the central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length. The cavity was filled up with high purity silicone oil. The light-guide was optically connected, on the opposite sides, to two low radioactive EMI9265–B53/FL 3 in. photomultipliers (PMT). The light-guide was wrapped by PTFE reflection tape. The detector was surrounded by Cu bricks and sealed in a low radioactive air-tight Cu box continuously flushed with high purity nitrogen gas (stored deeply underground for a long time) to avoid the presence of residual environmental radon. The Cu box was surrounded by a passive shield made of 10 cm of high purity Cu, 15 cm of low radioactive lead, 1.5 mm of cadmium and 4/10 cm polyethylene/paraffin to reduce the external background. The whole shield has been closed inside a Plexiglas box, also continuously flushed by high purity nitrogen gas.

In order to suppress the background caused by γ rays from the PMTs, two polished high purity quartz light-guides ($\varnothing 66 \times 100$ mm) were optically connected to the opposite sides of the polystyrene light-guide during the Run 3 and Run 5.

An event-by-event data acquisition system accumulates the amplitude and the arrival time of the events. The sum of the signals from the PMTs was recorded with the sampling frequency of 20 MS/s over a time window of 100 μs by a 8 bit transient digitizer (DC270 Acqiris).

The energy scale and the energy resolution of the $ZnWO_4$ detectors were measured by means of ^{22}Na, ^{60}Co, ^{133}Ba, ^{137}Cs and ^{228}Th γ sources. The energy dependence of the energy resolution can be fitted by the function: $FWHM_\gamma$ (keV) $= \sqrt{a+bE_\gamma}$, where E_γ is the energy of γ quanta in keV. For instance, the energy spectra accumulated by the detector ZWO-4 with ^{60}Co, ^{137}Cs and ^{228}Th γ sources are shown in Fig. 1. The parameters a and b were determined as $a=2398(570)$ keV^2 and $b=7.96(72)$ keV, respectively. Both the calibration and the background data were taken in the energy interval ~0.05–4 MeV.

The energy distributions accumulated over Runs 1, 2, and 4 with the $ZnWO_4$ scintillation detectors in the low background set-up are shown in Fig. 2. The background spectra accumulated over Runs 2 and 3 with $ZnWO_4$ crystals before and after the re-crystallization are depicted in Fig. 3 (Top) while the energy spectra accumulated over Runs 4 and 5 (before and after the installation of the additional quartz light-guides) are shown in Fig. 3 (Bottom). The spectra are normalized on the mass of the crystals and time of the measurements. A few peaks in the spectra can be ascribed to γ quanta of naturally occurring radionuclides ^{40}K, ^{214}Bi (^{238}U chain) and ^{208}Tl (^{232}Th) from materials of the set-up. As one can see from Fig. 3 (Bottom), the background spectrum measured over the Run 5 has also a peculiarity: a comparatively wide distribution in the energy interval $\approx 0.6-1.1$ MeV. Taking into account the α/β ratio,[2] this peak is mainly due to the radioactive contamination of the crystal ZWO-4 by α active nuclides of ^{232}Th, ^{235}U and ^{238}U families; this statement will further be proved by the pulse–shape discrimination in Section 3.2.2. The background counting rates of the $ZnWO_4$ detectors in the energy intervals 0.2–0.4, 0.8–1.0, and 2.0–2.9 MeV are given in Table 2.

3.2. Data analysis

The time–amplitude analysis, the pulse–shape discrimination between β(γ) and α particles, the pulse–shape analysis of the double pulses, and the Monte Carlo simulation of the measured energy distributions have been applied to estimate the radioactive contamination of the $ZnWO_4$ crystals.

3.2.1. Time–amplitude analysis

The technique of the time–amplitude analysis is described in details in Refs. [23,24]. The arrival time and energy of each event have been used for the selection of the following fast decay chain in the ^{232}Th family: ^{220}Rn ($Q_\alpha=6.41$ MeV, $T_{1/2}=55.6$ s)→^{216}Po ($Q_\alpha=6.91$ MeV, $T_{1/2}=0.145$ s)→ ^{212}Pb. All events within 0.5–1.75 MeV have been used as triggers, while a time interval 0.026–1.45 s (88.2% of ^{216}Po decays) and the same energy window have been set for the second events. Sixty events of the fast chain ^{224}Ra→^{220}Rn→^{216}Po→^{212}Pb were found in the data of Run 5. Taking into account the efficiency of the events selection in the time interval, one can calculate the activities of ^{228}Th in the $ZnWO_4$ crystal ZWO-4 as 18(2) μBq/kg. The search for the fast decay chain from the ^{227}Ac (^{235}U) family has also been performed in a similar way. Twelve events ^{219}Rn ($Q_\alpha=6.95$ MeV, $T_{1/2}=3.96$ s)→^{215}Po ($Q_\alpha=7.53$ MeV, $T_{1/2}=1.78$ ms)→^{211}Pb have been selected from the data of Run 5. Thus, the activity of ^{227}Ac in the crystal ZWO-4 has been calculated as 11(3) μBq/kg. The activities of ^{228}Th and ^{227}Ac in the $ZnWO_4$ crystal scintillators obtained by the time–amplitude analysis are presented in Table 3.

3.2.2. Pulse–shape discrimination (PSD) between β(γ) and α particles

As demonstrated in Ref. [5], the dependence of the pulse shapes on the type of irradiation in the $ZnWO_4$ scintillator allows one to discriminate γ(β) events from those induced by α particles. The optimal filter method proposed by E. Gatti and F. De Martini in 1962 [25] has been applied for this purpose.

For each signal $f(t)$, the numerical characteristic of its shape (shape indicator, SI) is defined as: $SI=\sum_k f(t_k)P(t_k)/\sum_k f(t_k)$, where the sum is over the time channels k, starting from the origin of signal and up to 50 μs, and $f(t_k)$ is the digitized amplitude (at the time t_k) of a given signal. The weight function $P(t)$ is defined as: $P(t)=\{\overline{f_\alpha}(t)-\overline{f_\gamma}(t)\}/\{\overline{f_\alpha}(t)+\overline{f_\gamma}(t)\}$, where $\overline{f_\alpha}(t)$ and $\overline{f_\gamma}(t)$ are the reference pulse shapes for α particles and γ quanta, respectively.

[2] The relative light yield for α particles as compared with that for γ quanta (β particles) can be expressed through α/β ratio, defined as ratio of α peak position in the energy scale measured with γ sources to the real energy of α particles (E_α). Because γ quanta interact with detector by β particles, we use more convenient term "α/β ratio". The α/β ratio for $ZnWO_4$ scintillator was taken from Ref. [5].