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Radiopurity of $ZnWO_4$ Crystal Scintillators

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Recently $ZnWO_4$ was proposed as perspective material for low-counting experiments to search for rare processes. Such experiments demand high radiopurity of $ZnWO_4$ crystal scintillators. With this aim radioactive contamination of large volume (0.1–0.7 kg) $ZnWO_4$ crystal scintillators were measured in the underground Laboratori Nazionali del Gran Sasso of the INFN at the depth of ≈ 3600 m water equivalent.

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1. Introduction

Zinc tungstate ($ZnWO_4$) is a very promising scintillator material. It has been proposed for application in searches for double beta decay in Ref. [1]. First low--background measurement with the small $ZnWO_4$ sample (mass of 4.5 g) was performed in the Solotvina Underground Laboratory (SUL, Ukraine; average overburden of ≈ 1000 m water equivalent, m w.e.) in order to study its radioactive contamination, and to search for double beta decay of zinc and tungsten isotopes [2]. Recently, radiopurity of large volume $ZnWO_4$ scintillator (mass of 119 g) has also been tested in the SUL [3]; other detectors (with masses of 0.1–0.7 kg) have been measured at Laboratori Nazionali del Gran Sasso (LNGS) [4, 5]. Possibilities to apply $ZnWO_4$ crystals for dark matter search were discussed in Ref. [2, 6–9].

In the present work, radiopurity of $ZnWO_4$ crystal scintillators for the next generation experiments was studied. Some of the results were previously published in [4, 5].

2. Experimental — detector and low-background measurements

In our studies at the LNGS three clear, slightly pink colored $ZnWO_4$ crystal scintillators were used. All crystals used for measurements are listed in Table I. The samples ZWO-1 and ZWO-2 were produced from monocrystal grown by the Czochralski method [3]. The crystal ZWO-3 was grown by the low-thermal gradient Czochralski technique [10]. The sample of $ZnWO_4$ crystal was fixed inside a cavity ($\oslash 49 \times 59$ mm) in the central part of a polystyrene light-guide ($\oslash 66 \times 312$ mm). The cavity was filled up with high purity silicone oil. The light-guide was optically connected on opposite sides by optical couplant to two low radioactivity EMI9265–B53/FL 3" photomultipliers (PMT)[1]. The light-guide was wrapped by PTFE reflection tape.

TABLE I

Samples of $ZnWO_4$ crystal scintillators used in the study.

ID	Size [mm]	Mass [g]	run	Lifetime [h]	Manufacturer
ZWO-1	$20 \times 19 \times 40$	117	2	2906	ISM Kharkiv[a]
ZWO-2	$\oslash 44 \times 55$	699	3	2130	ISM Kharkiv[a]
ZWO-3	$\oslash 41 \times 27$	239	5, 7	835, 4305	NIIC Novosibirsk[b]

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The detector has been installed deep underground ($\simeq 3600$ m w.e.) in the low background DAMA/R&D setup at the LNGS of the INFN (Italy). It was surrounded by Cu bricks and sealed in a low radioactive air-tight Cu box continuously flushed with high purity nitrogen gas to avoid presence of residual environmental Radon. The Cu box has been 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.

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[1] In order to suppress external background, the detector on stage run 7 was slightly modified: in addition two polished quartz light guides ($\oslash 66 \times 100$ mm) were optically connected on opposite sides of polystyrene light-guide.

The whole shield has been closed inside a Plexiglas box, also continuously flushed by high purity nitrogen gas.

An event-by-event data acquisition system accumulates the amplitude, the arrival time, and pulse shape of the events by a 1 GS/s 8 bit transient digitizer (DC270 Acqiris) with the sampling frequency of 20 MS/s over a time window of 100 μs.

The energy interval of data taking was chosen as 0.05–4 MeV. The energy scale and resolution of the $ZnWO_4$ detectors for γ quanta have been measured by means of ^{22}Na, ^{60}Co, ^{133}Ba, ^{137}Cs, and ^{228}Th sources. The energy dependence of the energy resolution in each run can be fitted by the function: $\mathrm{FWHM}_\gamma(\mathrm{keV}) = \sqrt{a + bE_\gamma}$, where E_γ is the energy of γ quanta in keV. For instance, the values of parameters a and b for the $ZnWO_4$ detector on stage run 7 are $a = 2398(570)$ keV^2 and $b = 7.96(72)$ keV.

The energy spectra accumulated over the runs 2, 3, 5, and 7 with the $ZnWO_4$ detectors in the low-background setup are shown in Fig. 1. The spectra are normalized to 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 setup. The data of the run 2 and run 3 have already been analyzed and published in [5].

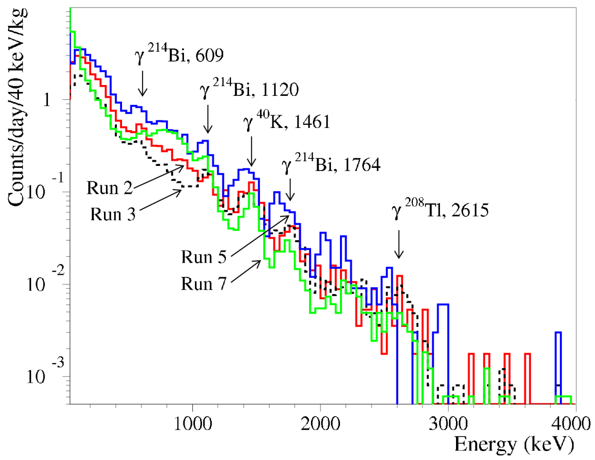

Fig. 1. (Color online) Energy spectra of $ZnWO_4$ scintillators measured in the low background setup during run 2 (solid red line), run 3 (dashed black line), run 5 (solid blue line), and run 7 (solid green line). Energies of γ lines are in keV.

3. Results and discussion

The time–amplitude analysis[2], the pulse-shape discrimination[3] between $\beta(\gamma)$ and α particles, the pulse-shape analysis of the double pulses[4], and the simulation of the measured energy spectra were used to estimate the radiopurity of the $ZnWO_4$ crystals.

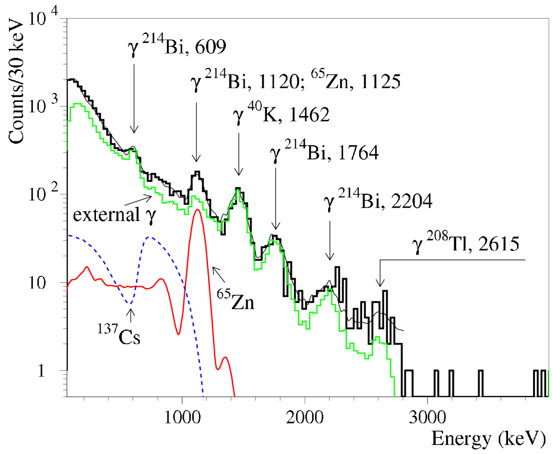

Fig. 2. (Color online) Energy spectrum of $\beta(\gamma)$ events accumulated in the low-background setup with the ZWO-3 crystal scintillator over 4305 h (run 7) together with the model of the background. The main components of the background are shown: spectra of ^{65}Zn, ^{137}Cs, and the contribution from the external γ quanta from PMTs in these experimental conditions.

The energy distributions of the possible internal contamination of the $ZnWO_4$ crystals (^{40}K, ^{60}Co, ^{87}Rb, ^{90}Sr+^{90}Y, ^{137}Cs, β active U/Th daughters) and from external γ quanta were simulated with the help of the GEANT4 package [16]. The initial kinematics of the particles emitted in the decay of nuclei was given by an event generator DECAY0 [17]. The measured background spectra were fitted by the model built from the simulated distributions. For instance, the fit of the spectrum measured with ZWO-3 sample (run 7) in the energy region 0.12–2.8 MeV and the main components of the background are shown in Fig. 2. The peak in the spectrum at the energy 1131 ± 8 keV cannot be explained by contribution from external γ rays[5]. We suppose presence of ^{65}Zn[6] ($T_{1/2} = 244.26$ d, $Q_\beta = 1351.9$ keV [18]) in the crystal to explain the peak. There are no other clear peculiarities in the spectrum which could be ascribed to the internal trace contamination by radioactive nuclides. Therefore we can obtain only limits on the activities of the β active radionuclides and U/Th daughters.

The summary of the measured radioactive contamination of the $ZnWO_4$ scintillators (or limits on their activities) is given in Table II. One can see that the levels of

[2] The technique of the time–amplitude analysis is described in detail in [11, 12].

[3] The optimal filter method proposed by Gatti and De Martini in 1962 [13] was used.

[4] The technique of the analysis is described in [14, 15].

[5] The 1120 keV γ line of ^{214}Bi is not enough intense to provide the whole peak area.

[6] It can be produced from ^{64}Zn by thermal neutrons (the cross section of ^{64}Zn for thermal neutrons is 0.76 barn [18]) or/and by cosmogenic activation.

TABLE II

Radioactive contamination of $ZnWO_4$ and $CdWO_4$ scintillators.

Nuclide	Activity [mBq/kg]			
	ZWO-1	ZWO-2	ZWO-3	$CdWO_4$ [14,19,20]
^{232}Th	$\leq 0.11^a$	$\leq 0.1^a$	$\leq 0.25^a$	$\leq$ 0.026–0.053(9)
^{228}Ra	$\leq 0.2^b$	$\leq 0.05^b$	$\leq 0.1^b$	≤ 0.004
^{228}Th	$0.005(3)^c$	$0.002(1)^c$	$0.018(2)^c$	$\leq$ 0.004–0.039(2)
^{227}Ac	$\leq 0.007^c$	$\leq 0.003^c$	$0.011(3)^c$	0.014(9)
^{238}U+^{234}U	$\leq 0.1^a$	$\leq 0.08^a$	$\leq 0.12^a$	$\leq$ 0.004–0.04
^{230}Th	$\leq 0.13^a$	$\leq 0.07^a$	$\leq 0.16^a$	$\leq$ 0.18–0.5
^{226}Ra	$\leq 0.006^a$	$0.002(1)^a$	$0.025(6)^a$	$\leq$ 0.004–0.04
^{210}Po	$\leq 0.2^a$	$\leq 0.06^a$	$\leq 0.64^a$	$\leq$ 0.063–0.4
Total α	$0.38(5)^a$	$0.18(3)^a$	$1.3(1)^a$	0.26(4)–2.3(3)
^{40} K	$\leq 1^b$	$\leq 0.4^b$	$\leq 0.01^b$	0.3(1)
^{60}Co	$\leq 0.05^b$	$\leq 0.1^b$	$\leq 0.004^b$	≤ 0.4
^{65}Zn	$\leq 0.8^b$	$0.5(1)^b$	$0.7(1)^b$	–
^{87}Rb	$\leq 2.6^b$	$\leq 2.3^b$	$\leq 4.1^b$	≤ 2.3
^{90}Sr+^{90}Y	$\leq 0.6^b$	$\leq 0.4^b$	$\leq 0.01^b$	≤ 0.2
^{137}Cs	$\leq 0.3^b$	$\leq 0.05^b$	$\leq 0.2^b$	$\leq$ 0.3–0.43(6)
^{147}Sm	$\leq 0.01^a$	$\leq 0.01^a$	$\leq 0.05^a$	$\leq$ 0.01–0.04

[a] Pulse-shape discrimination.
[b] Fit of background spectra.
[c] Time–amplitude analysis.

radioactive impurities in the $ZnWO_4$ crystals are comparable (and slightly lower) than those in the $CdWO_4$.

4. Conclusions

The radiopurity of the $ZnWO_4$ crystal produced in the NIIC (Novosibirsk) is worse than that of the scintillators produced in the ISM (Kharkiv). Thus, summarizing the results of measurements of the radioactive contaminations of the $ZnWO_4$ samples, we can conclude that the typical radiopurity of medium-size $ZnWO_4$ crystal scintillators is very promising for further applications.

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