Radioactive contamination of $CaWO_4$, $ZnWO_4$, $CdWO_4$, and Gd_2SiO_5:Ce crystal scintillators

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Abstract.
Radioactive contaminations of $CaWO_4$, $ZnWO_4$, $CdWO_4$, and Gd_2SiO_5:Ce crystal scintillators were measured in the Solotvina Underground Laboratory. The Monte Carlo simulation of contributing radioactive sources, the time-amplitude and pulse-shape analyses of the experimental data were applied. $CaWO_4$ and GSO scintillators are considerably polluted by uranium and thorium, while the total α activity in $ZnWO_4$ and $CdWO_4$ scintillators does not exceed the level of a few mBq/kg. Particular radioactivity was observed in $CaWO_4$ (α active ^{180}W), $CdWO_4$ (β active ^{113}Cd, $2\nu2\beta$ decay of ^{116}Cd, ^{180}W), and GSO (α active ^{152}Gd) detectors. The radioactive contamination of the studied scintillation crystals is compared with that of commonly used detectors.

Keywords: Scintillation detectors; Radioactive contamination
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INTRODUCTION

There is class of experiments on searches for rare events (as for instance, low-level gamma-, beta-, alpha- spectrometry, double beta decay (2β) and dark matter particles search, measurements of solar neutrino flux) which demand ultra-low background of detector. After the main sources of background (such as environmental radioactivity, cosmic rays, radioactivity of shield) are suppressed, internal radioactive contamination of detector begins to be the most important source of background.

Many experiments to search for rare α [1, 2, 3] and β decay [4, 5, 9], 2β decay [6, 8, 10, 11, 12, 13, 14, 15] and dark matter particles [16, 17, 10, 18] were performed using crystal scintillators. Some of scintillation crystals are also used as low-background scintillating bolometers [19, 20, 21, 22]. Here we analyze number of low-background measurements to estimate radioactive contamination of calcium, zinc, cadmium tungstate ($CaWO_4$, $ZnWO_4$, $CdWO_4$), and cerium-doped gadolinium orthosilicate (Gd_2SiO_5:Ce, GSO) crystal scintillators as promising detectors for rare events experiments.

EXPERIMENTAL METHODS

$CaWO_4$, $ZnWO_4$, $CdWO_4$, and GSO scintillation crystals studied in the present work were grown by Czochralski method. The main properties of the scintillators are presented in Table 1. All these crystals are non-hygroscopic and chemically resistant.

[1] Deceased.

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TABLE 1. Properties of $CaWO_4$, $ZnWO_4$, $CdWO_4$, and GSO crystal scintillators

	$CaWO_4$	$ZnWO_4$	$CdWO_4$	GSO
Density (g/cm^3)	6.1	7.8	8.0	6.71
Melting point ($^\circ$ C)	$1570-1650$	1200	1325	2173
Hardness (Mohs)	$4.5-5$	$4-4.5$	$4-4.5$	5.7
Wavelength of emission maximum (nm)	$420-440$	480	480	440
Refractive index	1.94	$2.1-2.2$	$2.2-2.3$	1.85
Effective average decay time (μs)*	8	24	13	0.05
Photoelectron yield [% of NaI(Tl)]*	18%	13%	20%	20%

* For γ rays at 20° C.

Radioactive contaminations of the crystals were measured in the Solotvina Underground Laboratory built in a salt mine 430 m underground ($\simeq$1000 m w. e., cosmic muon flux 1.7×10^{-6} cm^{-2} s^{-1}, neutron flux $\leq 2.7\times10^{-6}$ cm^{-2} s^{-1}, radon concentration in air <30 Bq m^{-3}) [23].

To estimate radiopurity of cadmium tungstate scintillators, the data of the ^{116}Cd and ^{106}Cd double beta decay experiments [24, 8, 25, 12] were used. The radioactive contamination of GSO scintillator was determined on the basis of the experiment [26, 11].

The $CaWO_4$ ($40\times34\times23$ mm) was measured during 1734 h in the low background set-up. The $CaWO_4$ scintillator was viewed by the special low radioactive 5" PMT (EMI D724KFL) through the high pure quartz light-guide 10 cm in diameter and 33 cm long. The $CdWO_4$ crystal ($\oslash59\times21$ mm) was measured during 37 h in the same set-up. The $ZnWO_4$ scintillation crystal ($\oslash14\times4$ mm, 429 h) was viewed by the 3" PMT (FEU-137) through the high pure quartz light-guide 4.9 cm in diameter and 25 cm long. The detectors were surrounded by an passive shield made of teflon ($3-5$ cm), plexiglass ($6-13$ cm), high purity copper (thickness $3-6$ cm), lead (15 cm) and polyethylene (8 cm). Two plastic scintillators ($120\times130\times3$ cm) were installed above the passive shield to generate a cosmic muons veto. For each event in the detector the amplitude of a signal and its arrival time were recorded. In addition, scintillation pulse shape of the $CaWO_4$ and $CdWO_4$ scintillators were digitized with a 20 MHz sampling frequency.

Pulse shape analysis was applied to $CaWO_4$ and $CdWO_4$ data as described in [27, 1, 28, 15]. Technique of time-amplitude analysis to recognize a presence of the short-living chains from ^{232}Th, ^{235}U and ^{238}U families is described in [24, 11]. The Monte Carlo simulation of contributing radioactive sources was performed with the help of GEANT3 and GEANT4 packages [29], and event generators DECAY0 and DECAY4 [30].

RESULTS AND DISCUSSION

The energy spectrum measured with the $ZnWO_4$, $CaWO_4$ and $CdWO_4$ crystal scintillators in the low background set-up is presented in Fig. 1 (the spectra are normalized by the measurement times and the detector masses).

The background of the $ZnWO_4$ detector is substantially lower than that of the $CaWO_4$ and is comparable with that of the $CdWO_4$ above ≈0.5 MeV. Note that below 0.5 MeV the counting rate of the $ZnWO_4$ detector is one order of magnitude lower than that of $CdWO_4$. Obviously, it is due to presence of the β active ^{113}Cd isotope (natural abundance of $\approx12\%$) in the $CdWO_4$ crystals.

Activity of thorium and uranium α active daughters in $CdWO_4$ and $CaWO_4$ crystals was determined with the help of pulse shape analysis. In addition, the pulse shape analysis allows to estimate activity of ^{228}Th and ^{226}Ra due to selection of double pulses associated with the fast sequences ^{214}Bi $\rightarrow$ ^{214}Po ($T_{1/2}=164$ μs) $\rightarrow$ ^{210}Pb and ^{212}Bi $\rightarrow$ ^{212}Po ($T_{1/2}=0.3\mu$ s)$\rightarrow$ ^{208}Pb from ^{235}U chain.

More precise data on radioactive impurities associated with the daughters of ^{232}Th, ^{235}U and ^{238}U were obtained with the help of the time-amplitude analysis. For example, the fast sequence of two α decays from the ^{232}Th family was searched for: ^{220}Rn ($Q_\alpha=6.41$ MeV, $T_{1/2}=55.6$ s) $\rightarrow$ ^{216}Po ($Q_\alpha=6.91$ MeV, $T_{1/2}=0.145$ s) $\rightarrow$ ^{212}Pb (which is in equilibrium with ^{228}Th). In the same way, the activity of ^{226}Ra (^{238}U family) in the scintillators was measured by selection of the fast sequence ^{214}Bi ($Q_\beta=3.27$ MeV, $T_{1/2}=19.9$ m) $\rightarrow$ ^{214}Po ($Q_\alpha=7.83$ MeV, $T_{1/2}=164$ μs) $\rightarrow$ ^{210}Pb. The presence of ^{227}Ac (^{235}U family) was estimated by time-amplitude analysis of ^{219}Rn ($Q_\alpha=6.95$ MeV, $T_{1/2}=3.96$ s) $\rightarrow$ ^{215}Po ($Q_\alpha=7.53$ MeV, $T_{1/2}=1.78$ ms)$\rightarrow$ ^{211}Pb from ^{235}U chain.

The background spectrum measured with the GSO crystal ($\oslash47\times57$ mm) during 13949 h [11] is depicted in Fig.

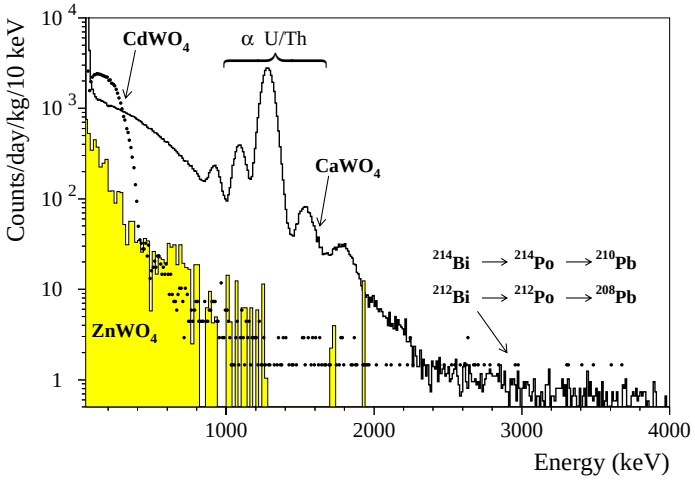

FIGURE 1. Energy spectra of $ZnWO_4$ (4.5 g, 429 h), $CaWO_4$ (189 g, 1734 h), and $CdWO_4$ (448 g, 37 h) scintillation crystals measured in the low background set-up

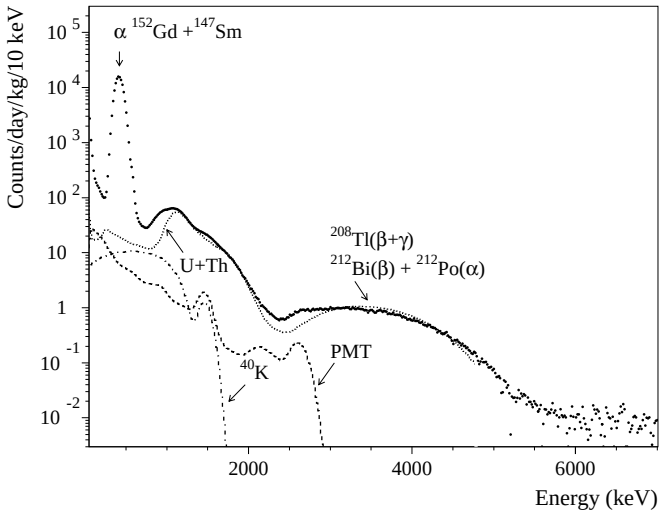

FIGURE 2. The background spectrum of the GSO crystal for 370.5 yr kg of exposure and the model of background.

2, where the following peculiarities exist: the clear peak at the energy 0.42 MeV, the comparatively wide peak at the energy around 1.05 MeV and two broad distributions dropped down at the energies 2.4 and 5.5 MeV. The first peak is attributed to α particles of ^{152}Gd ($T_{1/2} = 1.08 \times 10^{14}$ yr; $E_\alpha = 2140$ keV; abundance $\delta = 0.20\%$) and ^{147}Sm ($T_{1/2} = 1.06 \times 10^{11}$ yr; $E_\alpha = 2233$ keV; $\delta = 15\%$; samarium can be present as impurity of the GSO crystal at the level of $\approx$8 ppm). The peak near 1050 keV as well as the broad distribution up to the energy 2.4 MeV is mainly due to the radioactive contamination of the crystal by the nuclides from the ^{232}Th, ^{235}U and ^{238}U families. The distribution up to the energy 5.5 MeV is caused by decays of ^{232}Th daughter isotopes: a) β decay of ^{208}Tl ($Q_\beta = 5.00$ MeV); b) β decay of ^{212}Bi ($Q_\beta = 2.25$ MeV) followed by fast α decay of its daughter ^{212}Po ($T_{1/2} = 0.3$ μs; $E_\alpha = 8.78$ MeV or $\approx$2.7 MeV in β scale). The estimation of radioactive contamination of the GSO scintillator was performed by using the time-amplitude analysis and by analysis of the energy spectrum shape. The model of background was built with the help of Monte Carlo simulation. It should be stressed that no surface concentration of radionuclides was observed for the GSO crystal.

The summary of the measured radioactive contamination of the $CaWO_4$, $ZnWO_4$, $CdWO_4$ and GSO crystal scintillators (or limits on their activities) is given in Table 2.

As one can see from Table 2, the secular equilibrium in uranium chain is substantially broken in $CaWO_4$ crystal.

TABLE 2. Measured radioactive contaminations in $ZnWO_4$, $CaWO_4$, $CdWO_4$, and GSO crystal scintillators (mBq/kg).

Chain	Source	$CaWO_4$ *	$ZnWO_4$ †	$CdWO_4$ **	GSO ‡
^{232}Th	^{232}Th	0.69(10)	≤ 3.3	0.053(5)	≤ 6.5
	^{228}Ra				≤ 9
	^{228}Th	0.6(2)	≤ 0.2	$\leq 0.004-0.039(2)$	2.287(13)
^{235}U	^{227}Ac	1.6(3)	≤ 0.2	0.0014(9)	0.948(9)
^{238}U	^{238}U	14.0(5)	≤ 3.2	≤ 0.6	≤ 2
	^{230}Th		≤ 4.5	≤ 0.5	≤ 9
	^{226}Ra	5.6(5)	≤ 0.4	≤ 0.004	0.271(4)
	^{210}Pb	≤ 430	≤ 1	≤ 0.4	≤ 0.8
	^{210}Po	291(5)			
	^{40}K	≤ 12	≤ 12	0.3(1)	≤ 14
	^{90}Sr	≤ 70	≤ 1.2	≤ 0.2	
	^{113}Cd			580(20) §	
	^{113m}Cd			1-30	
	^{116}Cd			1.0(1) ¶	
	^{137}Cs	≤ 0.8	≤ 20	$\leq 0.3-0.43(6)$	
	^{138}La				≤ 55
	^{147}Sm	0.49(4)	≤ 1.8	≤ 0.04	700-1100
	^{152}Gd			≤ 0.04	1200
	^{180}W	0.05(2)		0.04(2)	

* [28, 31]
† [15]
** [25, 32, 8, 12, 1]
‡ [26, 11]
§ In crystals produced from natural cadmium
¶ In crystals produced from Cd enriched in ^{116}Cd

The equilibrium in thorium family is broken in $CdWO_4$ scintillators too. More likely it is a rule for different materials. On the other hand, measurement of particular sub-chains with comparable precision is quite complicate problem. For that reason radioactive contamination by uranium and thorium is more properly characterized by the value of the total α activity inside a detector.

Particular radioactivity, i.e. associated with elemental composition of a detector, was observed in $CaWO_4$ (α active ^{180}W), $CdWO_4$ (β active ^{113}Cd, $2\nu2\beta$ decay of ^{116}Cd, ^{180}W), and GSO (α active ^{152}Gd) detectors.

Let us mention the effect of U/Th traces concentration on surface layer of the $CdWO_4$ crystal grown from enriched ^{116}Cd. Contribution from U/Th α active daughters was decreased in ≈ 50 times after removing of surface layer (≈ 1 mm) of the crystal [24].

In Table 3 the radioactive contamination of the studied scintillation crystals is compared with that of commonly used detectors.

CONCLUSIONS

We present the radio-purities of $CaWO_4$, $ZnWO_4$, $CdWO_4$ and GSO crystals and compare them with other detectors. $CaWO_4$ and GSO scintillators are considerably polluted by uranium and thorium (particularly, $CaWO_4$ by ^{210}Po at the level of 0.3 Bq/kg, GSO by U/Th at the level of 0.04 Bq/kg). Radiopurity of $ZnWO_4$ and $CdWO_4$ crystals is significantly better. The total alpha activity of U/Th daughters in $ZnWO_4$ and $CdWO_4$ scintillators does not exceed the level of a few mBq/kg. The contaminations of $CdWO_4$, enriched in ^{116}Cd, by ^{232}Th and ^{226}Ra are at the level of 53 μBq/kg and 4 μBq/kg, respectively. Particular, i.e. associated with elemental composition of the detectors, radioactivity was observed in $CaWO_4$ (α active ^{180}W), $CdWO_4$ (β active ^{113}Cd and ^{113m}Cd, $2\nu2\beta$ decay of ^{116}Cd,

TABLE 3. Radioactive contamination of different detectors (mBq/kg).

Detector	Total α activity U + Th	^{228}Th	^{226}Ra	^{40}K	Particular contamination	Reference
$CaWO_4$	$20-400$	0.6	5.6	≤ 12		[28, 31, 22]
$ZnWO_4$	≤ 20	≤ 0.2	≤ 0.4	≤ 12		[15]
$CdWO_4$	$\leq 0.7-2.3(3)$	≤ 0.003	≤ 0.004	0.3	580 (^{113}Cd)	[25, 32, 8, 12, 1]
GSO	40	$2.3-107$	0.3	≤ 14	1200 (^{152}Gd)	[26, 11, 33]
NaI(Tl)	≤ 0.2	$0.0004-0.014$	$0.005-0.39$	1.5		[34, 35, 36]
CsI(Tl)		≤ 0.3	≤ 0.3	≤ 20		[37, 38]
BGO	≤ 0.4	≤ 1.2			$(0.5-3)\times 10^3$ (^{207}Bi)	[39, 2]
CaF_2(Eu)		$0.04-40$	$0.05-75$	5		[14, 10]
CeF_3	3400	1100	≤ 60	≤ 330		[13]
BaF_2		400	1400			[40]
Liquid scintillator	10^{-6}	1.2×10^{-6}	6.3×10^{-6}		0.3 (^{14}C)	[41, 42]
HPGe		$\leq 2\times 10^{-5}$	$\leq 2\times 10^{-5}$			[43, 44]

^{180}W), and GSO (α active ^{152}Gd) detectors. It was found that secular equilibrium in uranium chain is substantially broken in $CaWO_4$ crystals. Uranium and thorium traces were concentrated mainly in the surface layer of the $CdWO_4$ crystals enriched by ^{116}Cd.

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