

Application of PbWO_4 crystal scintillators in experiment to search for 2β decay of ^{116}Cd

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Abstract

Lead tungstate (PbWO_4) crystal scintillators are discussed as an active shield and light guides in ^{116}Cd double-beta decay experiment with Cadmium tungstate (CdWO_4) scintillators. Scintillation properties and radioactive contamination of PbWO_4 scintillators were investigated. Energy resolution of CdWO_4 detector, coupled to PbWO_4 light guide, was tested. Efficiency of PbWO_4 -based active shield to suppress background from the internal contamination of PbWO_4 crystals, as well as possible contribution from radioactivity of copper shield and phototubes were calculated. Using of lead tungstate crystal scintillators as high-efficiency 4π active shield could allow to build sensitive 2β experiment ($T_{1/2}^{0\nu 2\beta} \sim 10^{26}$ yr) to search for $0\nu 2\beta$ decay of ^{116}Cd with $^{116}\text{CdWO}_4$ crystal scintillators.

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1. Introduction

Studies of the ^{116}Cd 2β decay with the help of cadmium tungstate (CdWO_4) crystal scintillators enriched in ^{116}Cd to 83% have been performed in the Solotvina Underground Laboratory [1] since 1988. The results obtained in the different phases of these researches have been published earlier [2]. Beginning from 1998, the experiment was carried out in collaboration with the group from the University and INFN Firenze, Italy [3–6].

In the apparatus, which is described in detail in Refs. [3,5], four $^{116}\text{CdWO}_4$ crystals (total mass 330 g) were exploited. They are viewed by a low background 5 in.

phototube (PMT) through light guide $\varnothing 10 \times 55$ cm, which is glued of two parts: high pure quartz (25 cm) and plastic scintillator. The enriched $^{116}\text{CdWO}_4$ crystals were surrounded by an active shield made of 15 natural CdWO_4 crystals of large volume with total mass of 20.6 kg. These are viewed by a PMT through an active plastic light guide $\varnothing 17 \times 49$ cm. The whole CdWO_4 array is situated within an additional active shield made of plastic scintillator $40 \times 40 \times 95$ cm, thus, together with both active light guides, a complete 4π active shield of the $^{116}\text{CdWO}_4$ detector was provided. Due to the active and passive shields, and as a result of the time–amplitude and pulse-shape analysis of the data, the background rate of $^{116}\text{CdWO}_4$ detector in the energy region 2.5–3.2 MeV ($Q_{2\beta}$ energy of ^{116}Cd is 2805 keV) was reduced to 0.04 counts/(yr kg keV). It is one of the lowest background

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which has ever been reached with crystal scintillators. After 14183 h of data taking in the Solotvina Underground Laboratory the half-life limit on $0\nu 2\beta$ decay of ^{116}Cd was set as $T_{1/2} \geq 1.7 \times 10^{23}$ yr at 90% C.L., which corresponds to an upper bound on the effective Majorana neutrino mass $\langle m_\nu \rangle \leq 1.7$ eV [5]. This result is among the strongest world-wide restrictions (in addition to bounds obtained in experiments with ^{76}Ge [7,8], ^{82}Se and ^{100}Mo [9], ^{130}Te [10], and ^{136}Xe [11]). CdWO_4 crystals possess several unique properties required for a 2β decay experiment: good scintillation characteristics, low level of intrinsic radioactivity, and possibility of pulse-shape discrimination to reduce the background.

To enhance sensitivity of ^{116}Cd experiment to the level of neutrino mass 0.1–0.05 eV, one has to increase the measurement time and the mass of enriched $^{116}\text{CdWO}_4$, improve the energy resolution and reduce the background of the detector. As it was shown by Monte Carlo calculations, the required sensitivity could be achieved by using 150 kg of $^{116}\text{CdWO}_4$ crystals placed into a large volume of high-purity liquid (CAMEO project [12]). The project calls for the background reduction from the current 0.04 counts/(yr keV kg) to 10^{-3} – 10^{-4} counts/(yr keV kg). To decrease the background, the CAMEO project intends to use ≈ 1000 t of high-purity water or liquid scintillator ($\approx 10^{-15}$ g/g for ^{238}U and ^{232}Th) as a shield for $^{116}\text{CdWO}_4$ crystals. Due to low density of these liquids, the necessary dimensions of the shields are huge ($\approx \varnothing 11 \times 10$ m).

We propose an alternative solution for a sensitive 2β decay experiment with $^{116}\text{CdWO}_4$ by using lead tungstate (PbWO_4) crystal scintillators as high-efficiency 4π active shield. PbWO_4 crystal scintillators have been developed as heavy and fast detectors [13] for high-energy physics experiments. Scintillation characteristics of PbWO_4 have been intensively studied during the last decade [14–19]. High registration efficiency to γ quanta, very good transparency (a few meters in the region of CdWO_4 emission spectrum [20]), substantial difference of scintillation decay time in comparison with CdWO_4 , well developed tons-scale production [21] make this material very attractive to build a relatively small yet sensitive experiment to search for 2β decay of ^{116}Cd . In this paper, we study the possibility of applying PbWO_4 crystals as material for light guides and active shield in a ^{116}Cd double-beta decay experiment with CdWO_4 scintillators.

2. Measurements and results

2.1. Scintillation properties

The main properties of PbWO_4 crystal scintillators are presented in Table 1 where characteristics of CdWO_4 are also given for comparison. Measurements were carried out with two clear, colorless, undoped PbWO_4 crystals grown by Czochralski method. One crystal ($45 \times 22 \times 22$ mm, 182 g of mass, referred below as PWO-1) was produced in the Institute for Scintillation Materials (Kharkov, Uk-

Table 1
Properties of PbWO_4 and CdWO_4 crystal scintillators

	PbWO_4	CdWO_4
Density (g/cm ³)	8.28	8.0
Melting point (°C)	1123	1325
Structural type	Sheelite	Wolframite
Cleavage plane	Weak (101)	Marked (010)
Hardness (Mohs)	3	4–4.5
Wavelength of emission maximum (nm)	420–440	480
Refractive index	2.2	2.2–2.3
Effective average decay time ^a (μs)	0.01	13

^aFor γ rays, at indoor temperature.

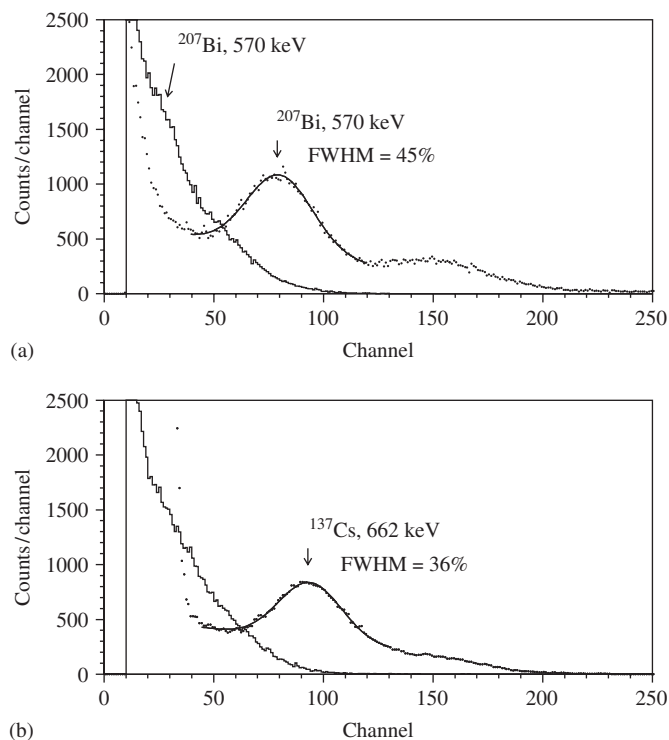


Fig. 1. Energy spectra measured by PbWO_4 scintillation crystal ($45 \times 22 \times 22$ mm) with (a) ^{207}Bi and (b) ^{137}Cs γ sources at two temperatures: $+24^\circ\text{C}$ (solid lines) and -18°C (points). For the spectra measured at -18°C , the fits of the ^{207}Bi (570 keV) and ^{137}Cs (662 keV) γ peaks by Gaussian plus exponential functions are also shown.

raine), and the second one ($32 \times 32 \times 10$ mm, 83 g of mass, PWO-2), produced in the Bogoroditsk Technical Chemical Plant (Russia) [21], was supplied by the Research Institute of Nuclear Problems (Minsk, Belarus).

Response of the PWO-1 scintillator to γ rays was measured with ^{137}Cs and ^{207}Bi γ sources at temperatures $+24$ and -18°C . The crystal was wrapped by PTFE reflector tape and optically coupled by Dow Corning Q2-3067 couplant to PMT XP2412. The measured energy spectra are presented in Fig. 1. The energy resolution FWHM = 45% and 36% was obtained at the temperature -18°C for 570 and 662 keV γ rays, respectively. The relative pulse amplitude has been increased in ≈ 3 times

with the detector cooling from +24 to −18 °C. The temperature of the detector was stabilized and measured with an accuracy of ±0.5 °C.

2.2. α/β ratio

The α/β ratio was measured with the PWO-1 crystal using a collimated ^{241}Am α source and thin ($\approx 0.65 \text{ mg/cm}^2$) mylar absorbers to obtain α particles in the energy range 2.1–4.6 MeV. The energies of α particles were determined with the help of a surface-barrier detector. In addition, α peak of ^{210}Po ($E_\alpha = 5.30 \text{ MeV}$) from internal contamination of PbWO_4 crystal by ^{210}Pb (see Subsection 2.3) was used. The dependence of the α/β ratio on energy is depicted in Fig. 2 where the α spectrum measured with the ^{241}Am α source is shown too. The α/β ratio increases above 3 MeV: $\alpha/\beta = 0.08(2) + 0.025(5)E_\alpha$, while it decreases as $\alpha/\beta = 0.23(4) - 0.024(14)E_\alpha$ at lower energies, where E_α is in MeV. The same behaviour of the α/β ratio was observed for CdWO_4 [4], calcium tungstate (CaWO_4) [22], and zinc tungstate (ZnWO_4) [23] crystal scintillators. The energy resolution for ^{210}Po α peak was measured as 39%, which is comparable with the energy resolution obtained with γ sources.

2.3. Radioactive contamination of PbWO_4 crystal scintillators

To estimate radioactive contamination, the PWO-1 crystal was measured in the Solotvina Underground Laboratory built in a salt mine 430 m underground ($\approx 1000 \text{ m}$ of water equivalent) [1]. The crystal was wrapped by PTFE tape and optically coupled to low radioactive PMT FEU-110. The detector was cooled to −18 °C in a temperature-controlled chamber. The temperature in the chamber was stabilized and measured with an accuracy of ±0.5 °C. The shaping time of the spectroscopy amplifier was set to 0.8 μs . Amplitude (energy) and arrival time of signals have been recorded by the event-by-event data acquisition system. The energy scale was calibrated with ^{207}Bi γ source. The energy spectrum accumulated during 2.15 h is shown in Fig. 3. The intense peak at the energy $\approx 1.2 \text{ MeV}$ (in γ scale) can be attributed to intrinsic ^{210}Po (daughter of ^{210}Pb from the ^{238}U family) with activity of 53(1) Bq/kg. The major part of events up to the energy $\approx 1 \text{ MeV}$ can be ascribed to β active ^{210}Bi (daughter of ^{210}Pb). The ^{210}Pb contamination of the PWO-2 crystal was measured as 79(3) Bq/kg.

Besides, the raw background data accumulated with the PWO-1 were analyzed by the time–amplitude method (described in detail in Ref. [24]), when the energy and arrival time of each event were used for find the fast sequence of β and α decays: ^{214}Bi ($Q_\beta = 3.27 \text{ MeV}$) \rightarrow ^{214}Po ($E_\alpha = 7.69 \text{ MeV}$, $T_{1/2} = 164 \mu\text{s}$) \rightarrow ^{210}Pb (^{238}U family). To select the β decays of ^{214}Bi , the energy threshold was set at 0.3 MeV (interval from 0.3 MeV to the end of the ^{214}Bi β spectrum contains 76% of the ^{214}Bi β events). For

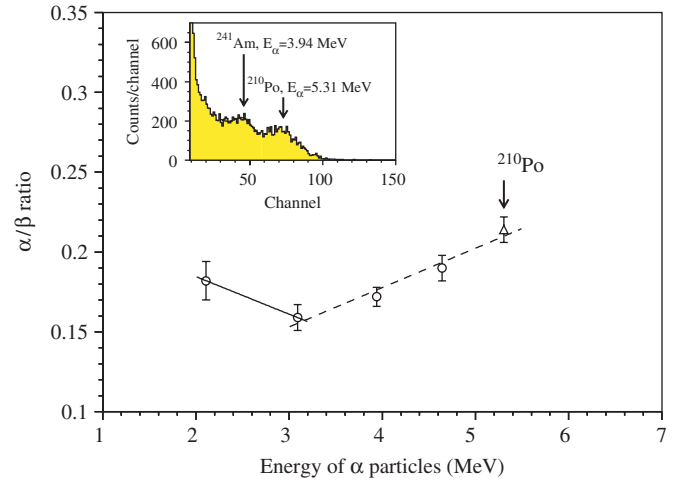


Fig. 2. Dependence of the α/β ratio on energy measured with the PbWO_4 scintillator $45 \times 22 \times 22 \text{ mm}$. The crystal was irradiated by α particles from ^{241}Am source through absorbers to obtain energies in 2.1–4.6 MeV range (circles). Triangle corresponds to α particles of ^{210}Po . (Inset) The α spectrum of ^{241}Am source measured with mylar absorber to obtain energy 3.94 MeV. Second peak at ≈ 73 channel is caused by α decay of ^{210}Po (daughter of ^{210}Pb) inside the scintillator.

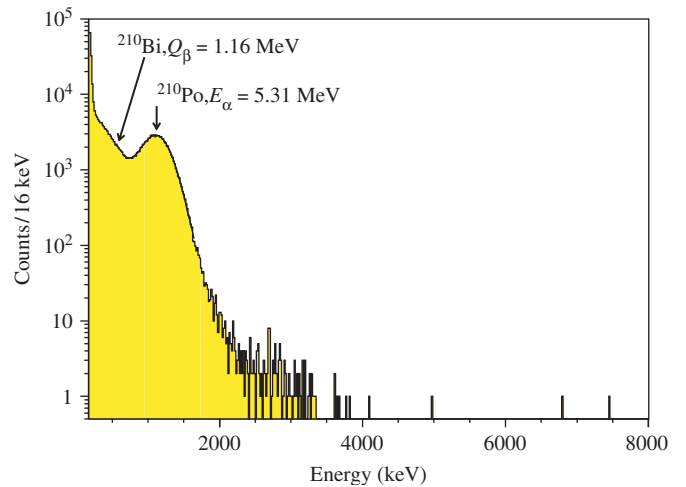


Fig. 3. Energy spectrum of PbWO_4 scintillation crystal (PWO-1) measured in the Solotvina Underground Laboratory at the temperature −18 °C during 2.15 h. Peak at the energy $\approx 1.2 \text{ MeV}$ (in γ scale) can be attributed to α decay of ^{210}Po from internal contamination of the crystal by ^{210}Pb . Broad distribution up to $\approx 1 \text{ MeV}$ corresponds to β spectrum of ^{210}Bi ($Q_\beta = 1.16 \text{ MeV}$).

the α decay of ^{214}Po , the energy window 1.6–2.6 MeV (94% of α events) and time interval of 90–1000 μs (67% of ^{214}Po decays) were chosen. There are no peculiarities in the obtained spectra which could be attributed to the sequence of decays searched for. The limit on the activity of ^{226}Ra in the PbWO_4 crystal $\leq 10 \text{ mBq/kg}$ was set. Comparing this value with the ^{210}Po activity, we can conclude that equilibrium of the uranium chain in the crystal is strongly broken.

Because the shaping time of the spectroscopy amplifier (0.8 μs) exceeds the half-life of ^{212}Po , and taking into

Table 2
Radioactive contaminations in PbWO₄ and CdWO₄ crystal scintillators

Chain	Source	Activity (mBq/kg)	
		PbWO ₄	CdWO ₄ [25–27,5]
²³² Th	²²⁸ Th	≤13	≤0.004–0.039(2)
²³⁸ U	²²⁶ Ra	≤10	≤0.004
	²¹⁰ Pb	(53–79) × 10 ³	≤0.4

account the α/β ratio, events from the fast sequence of ²¹²Bi β decay ($Q_\beta = 2.25$ MeV) and ²¹²Po α decay ($E_\alpha = 8.78$ MeV, $T_{1/2} = 0.3$ μ s) can result in one event registered in the detector with energy from 2.5 to 5 MeV. In the energy region 3.4–5 MeV (where $\approx 60\%$ of events from the sequence are expected), there are 7 events, which gives the limit on the activity of ²²⁸Th (²³²Th family) in the PWO-1 crystal ≤ 13 mBq/kg.

The summary of the measured radioactive contamination of the PbWO₄ crystal scintillators (or limits on their activities) is given in Table 2 in comparison with CdWO₄ scintillators.

2.4. PbWO₄ crystal as light-guide for CdWO₄ scintillator

A possibility to use PbWO₄ crystal as a light guide for CdWO₄ scintillation detector has been tested in measurements. With this aim, the energy resolution and relative pulse amplitude were measured with a CdWO₄ crystal in two conditions. First, the CdWO₄ crystal (10 × 10 × 10 mm, produced in the Institute for Scintillation Materials, Kharkov), wrapped by PTFE reflector tape, was optically coupled to PMT XP2412. The shaping time of the spectroscopy amplifier was set to 16 μ s. The energy resolution was measured with ¹³⁷Cs, ²⁰⁷Bi, and ²³²Th γ sources. In particular, the energy resolution (FWHM) 7.1%, 5.8% and 3.6% were obtained for 662, 1064 and 2615 keV γ lines, respectively. It should be stressed that the energy resolution of 3.6% at the energy 2615 keV was never reported for CdWO₄ scintillator. The energy spectrum measured with ²³²Th γ source is presented in Fig. 4(a). Then, the CdWO₄ crystal was viewed by the PMT through the PbWO₄ crystal PWO-1 (wrapped by mylar). The crystals and the PMT were optically coupled by Dow Corning Q2-3067 couplant. The energy resolution of 3.9% (for 2615 keV γ line of ²³²Th) and 86% of relative pulse amplitude were obtained (see Fig. 4(b)).

3. Background simulation

The different sources of background in the 2β experiment with ¹¹⁶CdWO₄ crystal scintillators were considered in Ref. [12]. Here we focus our attention on radioactive contamination of PbWO₄ crystals, photomultipliers and copper shield by ²³²Th and ²³⁸U. In addition, we consider background due to cosmogenic activation of PbWO₄.

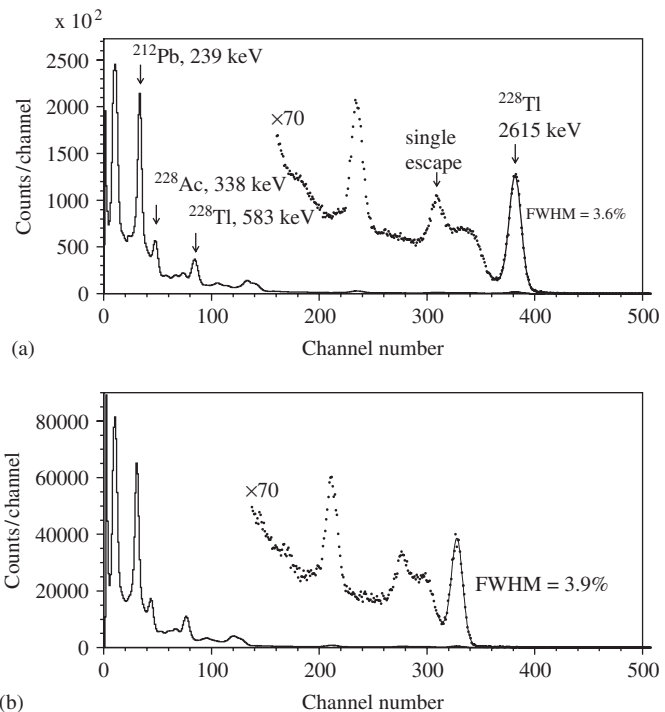


Fig. 4. Energy spectra measured by CdWO₄ scintillation crystal (10 × 10 × 10 mm) with ²³²Th γ source in two detector arrangements: (a) the crystal wrapped by PTFE reflector tape optically coupled to PMT; (b) the CdWO₄ crystal viewed by the PMT through the PbWO₄ crystal 45 × 22 × 22 mm as light guide.

Processes with β , α particles and γ rays were simulated with the help of the GEANT4 package [28] and the event generator DECAY0 [29].

3.1. Radioactive contamination of PbWO₄

The following conditions were accepted for the calculations: the CdWO₄ crystal (\varnothing 5 × 5 cm) with the energy resolution (FWHM) 4% at 2.8 MeV is placed in the center of PbWO₄ scintillation detector (\varnothing 45 × 45 cm), contaminated by ²³²Th and ²³⁸U at the level of 10⁻¹² g/g. 7.61 × 10⁶ decays of ²⁰⁸Tl inside the PbWO₄ detector were simulated. It corresponds to exposure of ≈ 250 kg × yr with the CdWO₄ detector. The calculated energy spectrum of the CdWO₄ detector, if no coincidence would be taken into account (PbWO₄ works as a passive shield), is shown in Fig. 5(a). The anticoincidence energy spectrum (the energy threshold of PbWO₄ detector was taken to be equal 0.5 MeV) is presented in Fig. 5(b). There are only two events in the energy interval of 0v2 β peak of ¹¹⁶Cd (2.7–2.9 MeV), which corresponds to the background counting rate 4 × 10⁻⁵ counts/(yr keV kg) from the ²³²Th contamination in PbWO₄.

The contamination of PbWO₄ crystals by ²²⁶Ra is even less dangerous. The Monte Carlo calculations show that no events above the energy of 2 MeV will be registered in the CdWO₄ detector during ≈ 250 kg × yr of exposure.

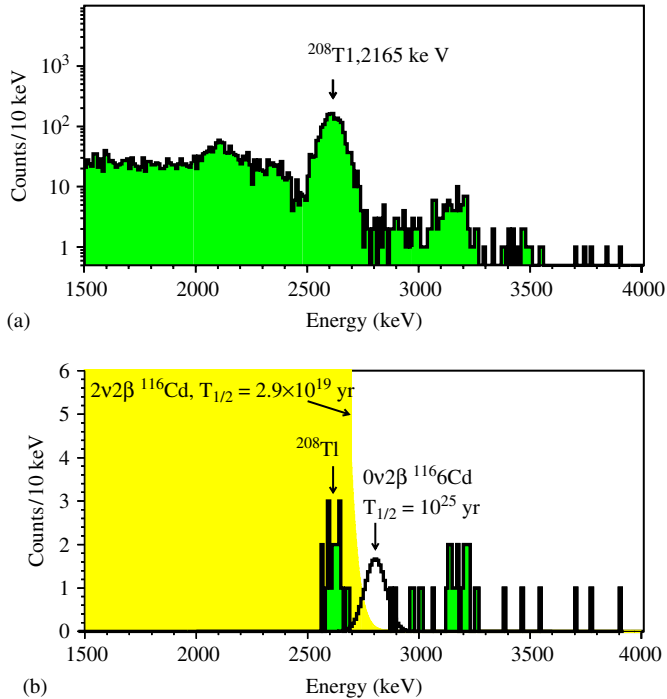


Fig. 5. The Monte Carlo simulated (a) response function of ¹¹⁶CdWO₄ detector (250 kg × yr of exposure) to decays of ²⁰⁸Tl inside shielding PbWO₄ crystals (see text). (b) The same as (a) but in anticoincidence with the PbWO₄ detector. Also the 0ν2β peak of ¹¹⁶Cd with $T_{1/2} = 10^{25}$ yr, and two neutrino 2β distribution ($T_{1/2} = 2.9 \times 10^{19}$ yr) are shown.

3.2. External γ rays from copper and PMTs

Only ²³²Th contaminations in copper shield and PMT's were taken into account (the contribution from ²³⁸U was found to be negligible) to estimate background in the vicinity of an expected ¹¹⁶Cd 2β decay peak. The following conditions were taken for the calculations: 32 CdWO₄ crystals ($\varnothing 5 \times 5$ cm) are surrounded by PbWO₄ scintillators ($70 \times 70 \times 70$ cm). A copper shield of 5 cm thick surrounds the PWO assembly. The copper is contaminated by ²³²Th at the level of 10^{-11} g/g. It should be noted such a level of radiopurity was reported in Ref. [30]. The CdWO₄ crystals are viewed through PbWO₄ light guides of 33 cm length by PMT with thorium contamination at the level of 2.5×10^{-7} g/g (PMT made of low background glass, see Ref. [31]), which corresponds to ²³²Th activity ≈ 0.2 Bq/PMT. The Monte Carlo simulation by GEANT4 gives 13 counts in the 2615 keV peak from copper and 3 from PMT during ≈ 10 yr of experiment. It results in ≈ 0.5 counts in the expected peak (2.7–2.9 MeV) from 0ν2β decay of ¹¹⁶Cd.

3.3. Cosmogenic activation of PbWO₄ crystals

The cosmogenic activation of lead tungstate was calculated with the help of the COSMO code [32]. PbWO₄ crystals was supposed to be produced during 30 days on the ground level and stored underground during 1 yr. Isotopes were selected from the full list of 175 radio-

nuclides by their decay rate after cooling (more than 10^{-3} decays/day/kg) and by their energy release (near 3 MeV). The most dangerous cosmogenic isotopes are listed in Table 3.

The background of CdWO₄ detector caused by cosmogenic activation of PbWO₄ crystals was calculated with the help of the GEANT4 code. We suppose 0.5 MeV energy threshold of PbWO₄ active shield, like for estimation of thorium- and uranium-induced background. It gives ≈ 0.16 counts from ⁶⁸Ga, ≈ 0.5 from ⁸⁸Y and ⁸⁸Zr, ≈ 0.26 from ^{106m}Ag in 25 kg CdWO₄ detector during ≈ 10 yr of measurements in the energy region of interest (2.7–2.9 MeV). The contribution from ¹⁰⁶Ru is less than 0.06 counts. Total cosmogenic background from PbWO₄ is expected to be only ≈ 0.9 counts in the energy interval of ¹¹⁶Cd 0ν2β decay peak.

4. Discussion

The response functions of a detector with enriched ¹¹⁶CdWO₄ crystals (≈ 250 kg × yr of exposure) for two neutrino ($T_{1/2} = 2.9 \times 10^{19}$ yr [5]) and neutrinoless 2β decay of ¹¹⁶Cd with half-life 10^{25} yr are presented in Fig. 5(b). Sensitivity of the experiment to neutrinoless 2β decay of ¹¹⁶Cd is at the level of $\lim T_{1/2} \approx 10^{26}$ yr (which corresponds to the limit on neutrino mass of ≈ 0.07 eV [33]). It is evident that the 0ν2β decay with $T_{1/2} \approx 10^{25}$ yr (neutrino mass ≈ 0.2 eV) would be certainly observed at this level of sensitivity.

The size of PbWO₄ active shield¹ in a setup could be equal approximately to $70 \times 70 \times 70$ cm. Thirty two enriched ¹¹⁶CdWO₄ crystals $\varnothing 5 \times 5$ cm are viewed by 3 in. PMT through logarithmic-spiral PbWO₄ crystals of 33 cm length. PbWO₄ as light guide has an advantage in comparison with plastic or quartz. Because of the high index of refraction (2.2), a logarithmic-spiral-type light guide can be made ≈ 60 mm in diameter, that allows to use 3 in. PMT (which typically have lower mass, better energy resolution and lower noise) instead of 5 in. PMT. Assuming ≈ 5 cm of passive copper, ≈ 50 cm of lead, and ≈ 50 cm polyethylene shield, dimensions of the setup are much more compact ($2.8 \times 2.8 \times 3.1$ m) if to compare with “water shield” apparatus ($\approx \varnothing 11 \times 10$ m) proposed in Ref. [12].

To get such an impressive result, the problem of low-radioactive PbWO₄ crystals production should be solved. In particular, content of ²¹⁰Pb has to be decreased: high counting rate (and, thus, big number of random coincidences) in ²¹⁰Pb–²¹⁰Bi–²¹⁰Po decays, observed in the current measurements, creates a problem for performing the time–amplitude analysis of events to search for the specific decay chains. It is well known that the freshly smelted lead is contaminated by ²¹⁰Pb at the level of hundreds Bq/kg [34], while its contamination by uranium and thorium is substantially less [35]. As the first step, we

¹It should be stressed that CdWO₄ crystals, successfully applied in the Solotvina experiment [5], can be also used as active shield detector.

Table 3
Cosmogenic activity in PbWO₄ scintillators calculated with the help of the COSMO code (see text)

Initial isotope and reaction	$T_{1/2}$	Energy release (keV)	Initial decay rate after 1 yr storing underground (decays/day/kg)	Number of decays during next 1 yr (decays/yr/kg)
$^{68}\text{Ge} \rightarrow ^{68}\text{Ga} \rightarrow ^{68}\text{Zn}$	271 d/68 m	106/2921	1.7×10^{-3}	0.4
$^{88}\text{Y} \rightarrow ^{88}\text{Sr}$	107 d	3623	5.4×10^{-3}	0.76
$^{88}\text{Zr} \rightarrow ^{88}\text{Y} \rightarrow ^{88}\text{Sr}$	83 d/107 d	673/3623	2×10^{-3}	0.23
$^{106}\text{Ru} \rightarrow ^{106}\text{Rh} \rightarrow ^{106}\text{Pd}$	374 d/30 s	39/3541	4.7×10^{-3}	1.2
$^{110m}\text{Ag} \rightarrow ^{110}\text{Cd}$	250 d	2892	1.6×10^{-2}	3.7

intend to grow PbWO₄ crystals from archaeological lead aiming to obtain PbWO₄ crystals less contaminated by ^{210}Pb .² As the next step, we foresee to estimate radioactive contamination of PbWO₄ crystals in low-background measurements by using the time–amplitude and pulse-shape (to select fast sequence of β and α decays from the ^{212}Bi – ^{212}Po chain) analyses. As it was demonstrated in the experiments with CdWO₄ scintillators, the sensitivities are at the level of a few $\mu\text{Bq/kg}$ for ^{228}Th , ^{226}Ra , and ^{227}Ac [5].

The energy threshold of the shielding PbWO₄ detector of 0.5 MeV can be achieved even with undoped scintillators at room temperature (see Fig. 1). However, as it was shown in Refs. [15,19], dopants like molybdenum and terbium can improve light yield of this scintillator, that allows to decrease the energy threshold of PbWO₄-based active shield.

5. Conclusions

Scintillation properties of PbWO₄ crystal scintillators were studied. The energy resolution FWHM = 36% was obtained for the 662 keV γ line of ^{137}Cs at -18°C . The α/β ratio was measured in the energy interval 2–5.3 MeV. The dependence of the α/β ratio on energy of α particles was observed. Radioactive contamination of two PbWO₄ crystals was measured in the Solotvina Underground Laboratory. Both crystals are considerably polluted by ^{210}Po at the level of 50–80 Bq/kg. For ^{228}Th (^{232}Th family) and ^{226}Ra (^{238}U) activities only upper limits were set at the level of 13 and 10 mBq/kg, respectively.

The excellent energy resolution of FWHM = 3.6% was obtained for 2615 keV γ line of ^{208}Tl with high-quality CdWO₄ crystal scintillator $10 \times 10 \times 10$ mm. The energy resolution of 3.9% (2615 keV γ line) and 86% of relative pulse amplitude was obtained for CdWO₄ scintillator viewed through PbWO₄ crystal as light guide. We expect an improvement of the light collection and the energy resolution of CdWO₄ detector by using the logarithmic-spiral PbWO₄ light guide.

Monte Carlo simulation and measurements demonstrate good abilities of PbWO₄ crystals to build 4π active shield

²For instance, only limit ≤ 4 mBq/kg on ^{210}Pb contamination in lead tungstate crystal produced from Roman lead was reported in Ref. [36].

for a sensitive ^{116}Cd double-beta decay experiment with CdWO₄ scintillators. Furthermore, PbWO₄ crystals, if their radiopurity will be proved, can be used as light guide and active shield in low counting experiments with scintillation, cryogenic and semiconductor detectors.

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References

- [1] Yu.G. Zdesenko, et al., in: Proceedings of the Second International Symposium on Underground Physics, Baksan Valley, USSR, August 17–19, 1987, Moscow, Nauka, 1988, p. 291.
- [2] F.A. Danevich, et al., Pis'ma Zh. Eksp. Teor. Fiz. 49 (1989) 417; F.A. Danevich, et al., JETP Lett. 49 (1989) 476; Yu.G. Zdesenko, J. Phys. G: Nucl. Part. Phys. 17 (1991) s243; F.A. Danevich, et al., Phys. Lett. B 344 (1995) 72; A.Sh. Georgadze, et al., Phys. Atom. Nucl. 58 (1995) 1093; F.A. Danevich, et al., Nucl. Phys. A 643 (1998) 317.
- [3] F.A. Danevich, et al., Phys. Rev. C 62 (2000) 045501; F.A. Danevich, et al., Nucl. Phys. A 717 (2003) 129.
- [4] F.A. Danevich, et al., Phys. Rev. C 67 (2003) 014310.
- [5] F.A. Danevich, et al., Phys. Rev. C 68 (2003) 035501.
- [6] F.A. Danevich, et al., Nucl. Phys. B (Proc. Suppl.) 138 (2005) 230.
- [7] H.V. Klapdor-Kleingrothaus, et al., Eur. Phys. J. A 12 (2001) 147.
- [8] C.E. Aalseth, et al., Phys. Rev. C 59 (1999) 2108; C.E. Aalseth, et al., Phys. Rev. D 65 (2002) 092007.
- [9] R. Arnold, et al., Pis'ma Zh. Eksp. Teor. Fiz. 80 (2004) 429; R. Arnold, et al., JETP Lett. 80 (2004) 377.
- [10] C. Arnaboldi, et al., Phys. Lett. B 557 (2003) 167; C. Arnaboldi, et al., Phys. Lett. B 587 (2004) 260.
- [11] R. Luescher, et al., Phys. Lett. B 434 (1998) 407; R. Bernabei, et al., Phys. Lett. B 546 (2002) 23.
- [12] G. Bellini, et al., Phys. Lett. B 493 (2000) 216; G. Bellini, et al., Eur. Phys. J. C 19 (2001) 43.
- [13] V.G. Barishevsky, et al., Nucl. Instr. and Meth. A 322 (1992) 231.
- [14] M. Kobayashi, et al., Nucl. Instr. and Meth. A 399 (1997) 261.
- [15] A. Annenkov, et al., Nucl. Instr. and Meth. A 450 (2000) 71.
- [16] M. Kobayashi, et al., Nucl. Instr. and Meth. A 465 (2001) 428.
- [17] M. Kobayashi, et al., Nucl. Instr. and Meth. A 484 (2002) 140.
- [18] A. Borisevich, et al., Nucl. Instr. and Meth. A 537 (2005) 101.
- [19] M. Kobayashi, et al., Nucl. Instr. and Meth. A 540 (2005) 381.
- [20] S. Baccaro, et al., Nucl. Instr. and Meth. A 385 (1997) 209.

- [21] A. Annenkov, et al., Nucl. Instr. and Meth. A 537 (2005) 173.
- [22] Yu.G. Zdesenko, et al., Nucl. Instr. and Meth. A 538 (2005) 657.
- [23] F.A. Danevich, et al., Nucl. Instr. and Meth. A 544 (2005) 553.
- [24] F.A. Danevich, et al., Nucl. Phys. A 694 (2001) 375.
- [25] A.Sh. Georgadze, et al., Instr. Exp. Techn. 39 (1996) 191.
- [26] S.Ph. Burachas, et al., Nucl. Instr. and Meth. A 369 (1996) 164.
- [27] F.A. Danevich, et al., Z. Phys. A 355 (1996) 433.
- [28] S. Agostinelli, (GEANT4 Collaboration), et al., Nucl. Instr. and Meth. A 506 (2003) 250; <http://geant4.web.cern.ch/geant4/>.
- [29] O.A. Ponkratenko, et al., Yad. Fiz. 63 (2000) 1355; O.A. Ponkratenko, et al., Phys. Atom. Nucl. 63 (2000) 1282.
- [30] C. Dorr, H.V. Klapdor-Kleingrothaus, Nucl. Instr. and Meth. A 513 (2003) 596.
- [31] Electron Tubes Limited, <http://www.electron-tubes.co.uk>
- [32] C.J. Martoff, P.D. Lewin, Comp. Phys. Comm. 72 (1992) 96.
- [33] A. Staudt, et al., Europhys. Lett. 13 (1990) 31.
- [34] A. Da Silva, et al., Nucl. Instr. and Meth. A 364 (1995) 578.
- [35] R.L. Brodzinski, et al., Nucl. Instr. and Meth. A 239 (1985) 207.
- [36] A. Alessandrello, et al., Nucl. Instr. and Meth. A 409 (1998) 451.