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Low Background CdWO₄ Scintillation Detector

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Low background scintillation setup has been developed that exhibits 3 orders of magnitude lower counting rate of a large volume (2.1 kg) CdWO₄ detector in the energy region 0.5–2.6 MeV, and one order of magnitude above 3 MeV. The background reduction was achieved by application of radiopure passive shield, active plasticscintillation muon veto placed above the setup and pulse-shape discrimination. Construction of additional plastic scintillation counters is in progress to reduce the residual cosmic muons background. The setup can be applied to develop radiopure scintillators, measure radioactive contamination of materials, carry out small scale low counting experiments.

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1. Introduction

Low counting experiments play an important role in astroparticle and nuclear physics as a way to test the Standard Model of particles, study properties of neutrino and weak interaction, search for dark matter and investigate rare nuclear decays. As low as possible radioactive background is requested by the experiments. Therefore, suppression of background is one of the most important issue to be addressed in the investigations of rare decays [1, 2]. Low background scintillation detectors based on inorganic crystal scintillators are widely used to study double β decay [3–5] search for dark matter [6, 7] investigate rare β [8] and α [9] decays. Moreover, recently developed cryogenic scintillating bolometers are very promising detectors for the next generation experiments to search for double β decay [10–15], and dark matter [16].

Basement background scintillation low setup (BALOO) was constructed for the purposes of radiopure scintillators R&D, screening of materials for low counting experiments, small scale experiments (search for double β decay, investigation of rare α and β decay, search for hypothetical decays and processes). The measurements described in the present study have been performed with a cadmium tungstate $(CdWO_4)$ crystal scintillator. CdWO₄ is up-to-date the most promising detector to investigate double β decay of cadmium. The material is proposed as one of the four detectors for the large scale double β decay project CUPID [17]. CdWO₄ crystals possess low levels of radioactive contamination [18], good optical and scintillation properties, particle discrimination capability [19]. The crystals can be grown of large volume (up to 20 kg) [20], technology of crystals production from enriched materials is already developed [21]. Double processes in tungsten isotopes can be investigated with the detector, too.

2. Experimental setup

Figure 1 shows a schematic view of the BALOO setup. A large volume CdWO₄ scintillation crystal with dimensions of $\emptyset7\times7$ cm² and mass 2.128 kg was produced by the low-thermal-gradient Czochralski technique [22]. The scintillator is viewed by a 5 inch low background photomultiplier tube (PMT, EMI D724KFL) through a $\emptyset100\times162$ mm² high purity quartz light guide (to reduce contribution of the PMT radioactive contamination). The detector is shielded by 6–12 cm layer of oxygen free high conductivity (OFHC) copper, and by 15 cm of old lead (produced more than 40 y ago). The passive shield is sealed by silicone compound with an aim to remove radon by nitrogen flux.

A muon veto counter (MVC) of the BALOO consists of four plastic scintillators $(50 \times 50 \times 12 \text{ cm}^3)$ covered by aluminized polyethylene terephthalate film. Each plastic scintillator is viewed by $\emptyset 17.5$ cm low radioactive PMT (FEU-125nf). The MVC is mounted above the lead shield. The construction of BALOO allows easy access to the inner volume of the detector by shift aside the top part of the lead shield with the MVC.

The BALOO setup is located in an especially designed laboratory on the basement floor of the Institute for Nuclear Research (Kyiv, Ukraine). The laboratory is equipped by temperature stabilization (the temperature in the laboratory is 22 ± 0.5 °C since August 2016) and air filtration systems.

The energy scale of the detector was measured with 60 Co, 137 Cs, 207 Bi, and 232 Th γ sources. The energy spectra accumulated with 60 Co and 232 Th γ sources are presented in Fig. 2. The energy resolution of the detector (full width at half maximum, FWHM) depends on energy of γ quanta (E_{γ}) as following:

FWHM [keV] = $\sqrt{12.3 \times E_{\gamma}}$, (2.1) where E_{γ} is in keV.

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Fig. 1. Schematic view of the BALOO setup.



Fig. 2. Energy spectra accumulated by the CdWO_4 low background scintillation detector with $^{60}{\rm Co}$ and $^{232}{\rm Th}~\gamma$ sources.

3. Results and discussion

3.1. Background measurements

Background measurements with the CdWO₄ lowbackground detector were carried out during September 2016–April 2017 (over more than 2000 h) in various shield concepts: without shield; in the lead shield; in anticoincidence (AC) with the MVC, with the copper shield installed. The background energy spectra accumulated in the experimental conditions are presented in Fig. 3. The spectrum accumulated without shield over 17 h contains γ peaks of ⁴⁰K (1461 keV), and radionuclides of ²³²Th and ²³⁸U chains. The γ quanta produce background up to the energy 2.6 MeV (γ quanta of ²⁰⁸Tl with energy 2615 keV, daughter of ²³²Th). The background above the energy is due to cosmic rays (mainly muons).

The lead shield reduces the background by 2 orders of magnitude in the energy region up to 2.6 MeV (see



Fig. 3. Energy spectra accumulated with $CdWO_4$ detector in various shield configurations (Pb denotes lead shield, MVC: muon veto counter, Cu: copper shield, PS: pulse-shape discrimination).

Table I). However, there were remaining γ peaks in the data due to γ quanta of $^{40}{\rm K}$ and $^{232}{\rm Th}/^{238}{\rm U}$ daughters. Application of the MVC reduces the background several times, especially after the 2615 keV peak of $^{208}{\rm Tl}$.

TABLE I

Background measured with CdWO₄ detector in various shield configurations in the BALOO setup. The background counting rates without shield, and in the Solotvina underground laboratory shielded by 5 cm of OFHC copper and 20 cm of lead are given for comparison.

Shield	Index of background [counts/(keV kg d)]		
$\operatorname{configuration}$	$0.5{-}1.5 { m MeV}$	1.5–3.0 MeV	3.0–5.0 MeV
without shield	$5.13(5) \times 10^3$	$8.61(9) \times 10^2$	6.45(8)
lead shield	$5.93(6) \times 10^1$	7.09(7)	1.97(2)
muon veto (MVC)	$2.72(3) \times 10^1$	3.27(3)	$6.47(7) \times 10^{-1}$
copper shield	$1.72(2) \times 10^{1}$	1.10(1)	$4.41(5) \times 10^{-1}$
fast pile-ups rejection (PS)	6.04(6)	$7.87(8) \times 10^{-1}$	$2.93(3) \times 10^{-1}$
Solotvina	6.12(7)	$4.9(2) \times 10^{-2}$	$1.6(3) \times 10^{-3}$

Installation of the copper shield suppressed substantially the γ background due to 40 K and U/Th. Further reduction of the background (especially in the energy region 0.4–0.9 MeV) was achieved by pulse-shape discrimination of fast Cherenkov signals caused by muons in the quartz light-guide (and of pile-ups of the Cherenkov and CdWO₄ scintillation signals) with the help of home-made electronic unit by comparison of the total versus fast components of the signals (PS). The main peculiarities in the data after application of the PS are β spectrum of $^{113}\mathrm{Cd}$ $(Q_{\beta} = 322 \text{ keV})$, a weak 662 keV γ peak of ¹³⁷Cs (a consequence of the Chernobyl accident), and a broad peak with energy ≈ 940 keV. Finally, the background of the CdWO₄ detector has been reduced by 3 orders of magnitude in the energy interval 0.5-2.6 MeV and by one order above 3 MeV in comparison to the unshielded detector.

3.2. Origin of $\approx 940 \ keV \ peak$

Measurement with a transient digitizer (20 MS/s, 12 bit) was performed to investigate the 940 keV peak nature. Pulse-profiles of the CdWO₄ detector signals were recorded over 18 h and analyzed by using the optimal filter method [19]. In the analysis, for each event the socalled shape indicator (SI) was calculated as following:

$$SI = \frac{\sum_{k=1}^{n} f(t_k) \times P(t_k)}{\sum_{k=1}^{n} f(t_k)},$$
(3.1)

were $f(t_k)$ — amplitude of the signal in the time channel t_k . The weight function P(t) was defined as:

$$P(t) = \frac{f_{\alpha}(t) - f_{\gamma}(t)}{f_{\alpha}(t) + f_{\gamma}(t)}$$

$$(3.2)$$

where $f_{\alpha}(t)$, $f_{\gamma}(t)$ are the reference pulse shapes for α particles and γ quanta.

There are two populations of events on the dependence of the shape indicator on energy presented in Fig. 4a. The energy spectra of the populations are shown in Fig. 4b. A calibration measurement with 232 Th γ source has confirmed that the shape indicator values for γ quanta are similar to the observed in the background data, while the events above the γ band with energies in the energy interval 0.4–1.2 MeV are due to α particles of U/Th and daughters (taking into account the quenching of CdWO₄ scintillation efficiency to α particles [23]). We assume that the α events are mainly due to decays of ²¹⁰Po $(Q_{\alpha} = 5407 \text{ keV}, T_{1/2} = 138 \text{ d})$. The nuclide ²¹⁰Po is daughter of ²¹⁰Pb (²³⁸U chain), that is contamination of the crystal (however, the energy of β decay of ²¹⁰Pb $Q_{\beta} = 63.5$ keV is too low to be detected in our measurements due to the high counting rate caused by the β decay of ¹¹³Cd). For the moment we cannot explain the events with energies ≈ 0.2 –0.5 MeV, with the shape indicator values higher than ≈ 40 . The events can be due to surface contamination of the crystal or surroundings materials by α active U/Th nuclides, including radon deposited on the surfaces.

3.3. Efficiency of the muon veto counter

A dependence of the MVC efficiency to the cosmic muons on the MVC counting rate has been studied. For this purpose, energy spectra of the CdWO₄ detector were accumulated in coincidence (CC) with the MVC at various MVC counting rates. Then the counting rates of the $CdWO_4$ detector above 5 MeV were calculated. The events are mainly due to the cosmic muons passing both the $CdWO_4$ detector and the MVC. The dependence of the counting rate of the $CdWO_4$ detector on the counting rate of the MVC is presented in Fig. 5. One can see that above the MVC counting rate ≈ 350 counts/s the counting rate in the CdWO₄ detector remains stable (≈ 0.068 counts/s). Therefore, the MVC operating with the counting rate ≈ 350 counts/s provides a highest possible efficiency with the lowest dead time in the experimental conditions.

The efficiency of the MVC detector was estimated by measurements of high energy events by the CdWO₄ detector (well above the energy 5 MeV, where the cosmic



Cherenkov signals pileup

Fig. 4. (a) Shape indicator (SI) versus energy accumulated with the CdWO₄ detector over 18 h in the BALOO setup, (b) energy spectra of raw data and contributions of α and $\gamma(\beta)$ particles.



Fig. 5. Dependence of the counting rate of the $CdWO_4$ detector above 5 MeV in coincidence with the MVC on the counting rare of the MVC. The dashed line shows the CdWO₄ detector in CC with MVC counting rate 0.068 counts/s.

background dominates) in CC and AC with the MVC (see Fig. 6). The efficiency of MVC to reject cosmic muons (ratio of detected muons number to the total number of muons passing the MVC) can be estimated as 80% by calculation of the ratio $S_{CC}/(S_{CC}+S_{AC})$, where S_{CC} is area of the CC spectrum, S_{AC} is area of the AC spectrum.

The MVC efficiency was also evaluated considering the geometry of the setup. We have calculated probability (efficiency of the MVC) for muon to pass both the CdWO₄ detector and the MVC (ε_{MV}) as following:



Fig. 6. Energy spectra of CdWO₄ in coincidence (that is number of rejected muon signals) and anticoincidence with MVC (number of not rejected muon signals). The energy scale is given in channels since the difficulties to measure energy scale above 3 MeV. Roughly, the peak in the data is at energy ≈ 110 MeV.

$$\varepsilon_{MV} = \frac{\int_{\Omega_1} I(\theta) \,\mathrm{d}\Omega}{\int_{\Omega_2} I(\theta) \,\mathrm{d}\Omega} \tag{3.3}$$

where

$$I(\theta) = I \cos^n \theta, \tag{3.4}$$

where $n \simeq 2$ [24], I_0 — vertical muon fluence, θ zenith angle. Ω_1 is solid angle covered by MVC over the horizon (the vertex of angle is on the bottom of the CdWO₄ crystal), and $\Omega_2 = 2\pi$. The calculated part of the muons that pass both the CdWO₄ detector and the MVC $\varepsilon_{MV} \simeq 80\%$, that is in agreement with the measured value. Construction of additional plastic scintillation counters is in progress to reduce the residual cosmic muons background. The additional muon counters should be placed aside the passive shield to cover the CdWO₄ detector completely ($\Omega_1 = 2\pi$).

4. Conclusions

The radioactive background of CdWO₄ scintillation detector was studied over about 2000 h. The level of background was reduced by 3 orders of magnitude in the energy interval 0.5–2.6 MeV (energy region where γ quanta of environmental radioactivity dominate), and by one order of magnitude above 3 MeV (cosmic rays background) by application of passive shield, muon veto, and pulse-shape discrimination of the Cherenkov signals in the quartz light-guide. A peak with energy ≈ 940 keV in the spectra in the last shield concept can be attributed mainly to ²¹⁰Dî (α active daughter of ²¹⁰Pb from ²³⁸U chain). The efficiency of muon veto counter is 80%. An R&D of additional plastic scintillation counters is in progress to reduce the residual cosmic muon background.

Further improvement of the background can be achieved by pulse shape discrimination and timeamplitude analysis (to identify fast decay chains in the 232 Th, 235 U and 238 U daughters, that can be presented in the CdWO₄ crystal as trace contamination), by cleaning of the detector and the copper shield details (particularly, to remove the 137 Cs surface contamination), and by nitrogen flux to remove radon.

The BALOO setup is high low-background performance scintillation spectrometer, that can be applied to investigate radioactive contamination of scintillators and materials, and to carry out small scale low counting experiments.

References

- R.L. Brodzinski, J.H. Reeves, F.T. Avignone III, H.S. Miley, J. Radioanal. Nucl. Chem. 124, 513 (1988).
- [2] G. Heusser, Ann. Rev. Nucl. Part. Sci. 45, 543 (1995).
- [3] F.A. Danevich, A.Sh. Georgadze, V.V. Kobychev, B.N. Kropivyansky, A.S. Nikolaiko, O.A. Ponkratenko, V.I. Tretyak, S.Yu. Zdesenko, Yu.G. Zdesenko, *Phys. Rev. C* 68, 035501 (2003).
- [4] P. Belli, R. Bernabei, V.B. Brudanin, F. Capella, V. Caracciolo, R. Cerulli, D.M. Chernyak, F.A. Danevich, S. d'Angelo, A. Di Marco, A. Incicchitti, M. Laubenstein, V.M. Mokina, D.V. Poda, O.G. Polischuk, V.I. Tretyak, I.A. Tupitsyna, *Phys. Rev. C* 93, 045502 (2016).
- [5] S. Umehara, T. Kishimoto, I. Ogawa, R. Hazama, H. Miyawaki, S. Yoshida, K. Matsuoka, K. Kishimoto, A. Katsuki, H. Sakai, D. Yokoyama, K. Mukaida, S. Tomii, Y. Tatewaki, T. Kobayashi, A. Yanagisawa, *Phys. Rev. C* 78, 058501 (2008).
- [6] R. Bernabei, P. Belli, F. Capella, R. Cerulli, C.J. Dai, A. d'Angelo, H.L. He, A. Incicchitti, H.H. Kuang, X.H. Ma, F. Montecchia, F. Nozzoli, D. Prosperi, X.D. Sheng, R.G. Wang, Z.P. Ye, *Eur. Phys. J. C* 67, 39 (2010).
- [7] S.C. Kim, H. Bhang, J.H. Choi, W.G. Kang, B.H. Kim, H.J. Kim, K.W. Kim, S.K. Kim, Y.D. Kim, J. Lee, J.H. Lee, J.K. Lee, M.J. Lee, S.J. Lee, J. Li, J. Li, X.R. Li, Y.J. Li, S.S. Myung, S.L. Olsen, S. Ryu, I.S. Seong, J.H. So, Q. Yue, *Phys. Rev. Lett.* 108, 181301 (2012).
- [8] P. Belli, R. Bernabei, N. Bukilic, F. Capella, R. Cerulli, C.J. Dai, F.A. Danevich, J.R. de Läter, A. Incicchitti, V.V. Kobychev, S.S. Nagorny, S. Nisi, F. Nozzoli, D.V. Poda, D. Prosperi, V.I. Tretyak, S.S. Yurchenko, *Phys. Rev. C* **76**, 064603 (2007).
- [9] F.A. Danevich, A.Sh. Georgadze, V.V. Kobychev, S.S. Nagorny, A.S. Nikolaiko, O.A. Ponkratenko, V.I. Tretyak, S.Yu. Zdesenko, Yu.G. Zdesenko, *Phys. Rev. C* 67, 014310 (2003).
- [10] C. Arnaboldi, J.W. Beeman, O. Cremonesi, L. Gironi, M. Pavan, G. Pessina, S. Pirro, E. Previtali, Astropart. Phys. 34, 143 (2010).
- [11] J.W. Beeman, F.A. Danevich, V.Ya. Degoda, E.N. Galashov, A. Giuliani, V.V. Kobychev, M. Mancuso, S. Marnieros, C. Nones, E. Olivieri, G. Pessina, C. Rusconi, V.N. Shlegel, V.I. Tretyak, Ya.V. Vasiliev, *Phys. Lett. B* **710**, 318 (2012).
- [12] H. Bhang et al., J. Phys. Conf. Ser. 375, 042023 (2012).

- [13] J.W. Beeman, F. Bellini, L. Cardani, N. Casali, I. Dafinei, S. Di Domizio, F. Ferroni, L. Gironi, A. Giuliani, S. Nagorny, F. Orio, L. Pattavina, G. Pessina, G. Piperno, S. Pirro, E. Previtali, C. Rusconi, C. Tomei, M. Vignati, *JINST* 8, P05021 (2013).
- [14] D.R. Artusa et al., *Eur. Phys. J.* C 74, 3096 (2014).
- [15] T.B. Bekker, N. Coron, F.A. Danevich, V.Ya. Degoda, A. Giuliani, V.D. Grigorieva, N.V. Ivannikova, M. Mancuso, P. de Marcillac, I.M. Moroz, C. Nones, E. Olivieri, G. Pessina, D.V. Poda, V.N. Shlegel, V.I. Tretyak, M. Velazquez, *Astropart. Phys.* **72**, 38 (2016).
- [16] G. Angloher et al., *Eur. Phys. J. C* 72, 1971 (2012).
- [17] G. Wang et al., arXiv:1504.03599, [physics.ins-det] (2015).
- [18] D.V. Poda, et al., *Rad. Measur.* 56, 66 (2013).

- [19] T. Fazzini, P.G. Bizzeti, P.R. Maurenzig, C. Stramaccioni, F.A. Danevich, V.V. Kobychev, V.I. Tretyak, Yu.G. Zdesenko, *Nucl. Instrum. Methods Phys. Res. A* 410, 213 (1998).
- [20] E.N. Galashov, V.A. Gusev, V.N. Shlegel, Ya.V. Vasiliev, *Crystallogr. Rep.* 54, 689 (2009).
- [21] P. Belli et al., Nucl. Instrum. Methods Phys. Res. A 615, 301 (2010).
- [22] A.A. Pavlyuk, Ya.V. Vasiliev, L.Yu. Kharchenko, F.A. Kuznetsov, in: Proc. APSAM-92, Asia Pacific Society for Advanced Materials, Shanghai, 1992, Institute of Materials Research, Tohoku University, Sendai 1993, p. 164.
- [23] V.I. Tretyak, Astropart. Phys. **33**, 40 (2010).
- [24] M. Bektasoglu, H. Arslan, Pramana J. Phys. 80, 837 (2013).