LOW BACKGROUND SCINTILLATION SETUP TO INVESTIGATE RADIOPURITY OF MATERIALS

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Background of 2.128 kg $CdWO_4$ scintillation detector was reduced by 3 orders of magnitude in the energy region 0.5…2.6 MeV, and one order of magnitude above 3 MeV in the low background scintillation setup located in the basement floor of a laboratory building at the Institute for Nuclear Research (Kyiv, Ukraine). It was achieved by application of radiopure passive shield, plastic scintillators to veto cosmic rays, and pulse-shape discrimination to reject fast Cherenkov signals in the quartz light-guide of the detector. Sensitivity of the setup to radioactive contamination of different materials were estimated. For instance, for a 3.36 kg titanium sample over 30 days of measurements detection limits are on the level of 2.9 mBq/kg (^{40}K); 0.6 mBq/kg (^{137}Cs); 2 mBq/kg (^{226}Ra), and 0.5 mBq/kg (^{228}Th).

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INTRODUCTION

Low counting experiments are required to investigate rare nuclear processes, the most interesting of which is neutrinoless double beta (0ν2β) decay. The investigations of 0ν2β decay are considered as a way to test the lepton number conservation, the neutrino nature (Dirac or Majorana particle?), to determine an absolute scale and the neutrino mass scheme. Apart of the most natural light neutrino exchange mechanism, the decay could be mediated by presence of right-handed currents in weak interaction, by majoron emission, as well as by many other effects beyond the Standard Model of particle physics. Detection of rare processes, particularly of the 0ν2β decay, requires substantial reduction of radioactive background [1, 2].

Inorganic crystal scintillators are widely used to study 2β decay [3–5], rare β [6], and α [7] decays, and to search for dark matter [8, 9]. Moreover, crystal scintillators can be used as cryogenic scintillating bolometers, that is a very promising detector technique for the next generation 2β [10–15], and dark matter experiments [16].

BALOO (Basement Low background scintillation setup) was developed and constructed in the Lepton Physics Department of the Institute for Nuclear Research (Kyiv, Ukraine) to R&D of radiopure scintillators, select radiopure materials for low counting experiments, and to carry out small scale rare decay experiments.

The measurements described in the present paper have been performed with a cadmium tungstate ($CdWO_4$) crystal scintillator. $CdWO_4$ is one of the most promising detector materials to investigate 2β decay of cadmium thanks to its low level of radioactive contamination [17], high scintillation properties, particle discrimination capability [18]. The single crystals of a large volume can be grown (up to 20 kg) [19], technology of crystals production from enriched materials is also well established [20]. $CdWO_4$ is foreseen to be utilized in the large scale 2β decay experiment CUPID [21].

BALOO SETUP

A schematic view of the BALOO set-up is presented on Fig. 1. A 2.128 kg $CdWO_4$ crystal scintillator with dimensions ∅7×7 cm was produced by the low-thermal-gradient Czochralski technique [22]. The scintillator is viewed by a low background photomultiplier tube (PMT, EMI D724KFL, ∅ 5″) through a ∅10×16.2 cm high-purity quartz light guide to reduce contribution of the PMT radioactive contamination. A 6…12 cm layer of oxygen-free high conductivity (OFHC) copper, and 15 cm of old lead (produced more than 40 y ago) were applied as passive shield of the detector. The slots in the passive shield were filled by silicone grease with an aim to remove radon by nitrogen flow.

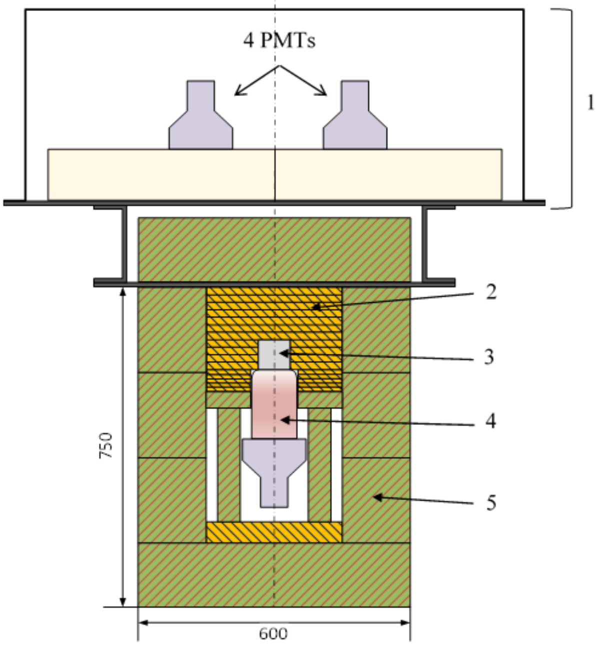

Fig. 1. Schematic view of BALOO: 1 – muon veto counter (MVC); 2 – OFHC copper; 3 – $CdWO_4$; 4 – quartz light guide; 5 – lead

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A muon veto counter (MVC) of the BALOO consists of four plastic scintillators (50×50×12 cm) covered by aluminized polyethyleneterephthalate film. Each plastic scintillator is viewed by ∅7″ low radioactive PMT (FEU-125nf). The MVC is placed above the lead shield.

The BALOO setup is installed in a specially designed laboratory room on the basement floor of the Institute for Nuclear Research (Kyiv, Ukraine).

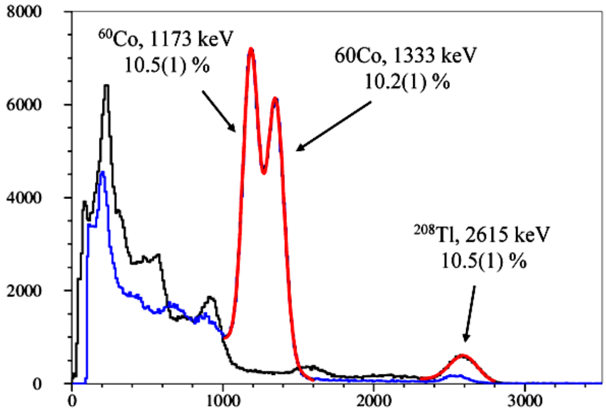

Fig. 2. Energy spectra of ^{60}Co and ^{232}Th γ-sources accumulated by the CdWO$_4$ low background scintillation detector

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 detector was calibrated by using ^{60}Co, ^{137}Cs, ^{207}Bi, and ^{232}Th γ-sources. The energy spectra accumulated with ^{60}Co and ^{232}Th γ-sources are shown in Fig. 2. The energy resolution of the detector (full width at half maximum, FWHM) depends on energy as:

$$FWHM = \sqrt{12.3 E_\gamma}\,, \qquad (1)$$

where E_γ is energy of γ-quanta in keV.

RESULTS AND DISCUSSION

BACKGROUND REDUCTION

Background measurements with the CdWO$_4$ 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 ^{40}K (1461 keV), and radionuclides of the ^{232}Th and ^{238}U chains. The γ-quanta produce background up to the energy 2.6 MeV (γ-quanta of ^{208}Tl with energy 2615 keV, daughter of ^{232}Th). The background above the energy is predominantly caused by cosmic rays (mainly muons).

Tabl. 1 presents the background counting rates in various shield configurations. Installation of the lead shield reduced the background by 2 orders of magnitude in the energy region up to 2.6 MeV in comparison with the unshielded detector. However, there are still peaks due to γ-quanta of ^{40}K and ^{232}Th/^{238}U daughters. Further several times reduction of the background, especially after the 2615 keV peak of ^{208}Tl, has been achieved by application of the MVC to reduce counts caused by cosmic rays (mainly muons).

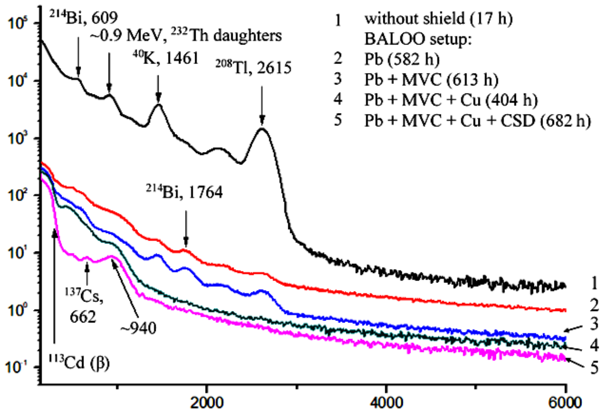

Fig. 3. Background energy spectra accumulated with CdWO$_4$ scintillation detector in different shield configurations (see text)

Table 1

Background counting rates measured with CdWO$_4$ detector in different shield configurations in the BALOO setup. Backgrounds of the CdWO$_4$ scintillator in the Solotvina underground laboratory are given for comparison

Shield configuration	Index of background counts/(keV×kg×d)		
	0.5…1.5 MeV	1.5…3.0 MeV	3.0..5.0 MeV
Without shield	5130(5)	861(9)	6.45(8)
Lead	59.3(6)	7.09(7)	1.97(2)
MVC	27.2(3)	3.27(3)	0.647(7)
Copper	17.2(2)	1.10(1)	0.441(5)
CSD	6.04(6)	0.787(8)	0.293(3)
Solotvina	6.12(7)	0.049(2)	0.0016(3)

Using of the copper shield allowed to suppress 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 rejection of fast Cherenkov signals, and their pile-ups with CdWO$_4$ signals, caused by muons in the quartz light-guide with the help of a specially designed electronic unit (CSD, Cherenkov signals discrimination). The unit compares the total and fast components of the signals. The main peculiarities in the spectra in the current shield configuration (lead + copper + MVC + CSD) are β-spectrum of ^{113}Cd (natural isotope of cadmium, Q_β = 322 keV), a weak 662 keV γ-peak of ^{137}Cs (a consequence of the Chernobyl accident), and a broad peak with maximum at ~ 940 keV. Finally, the total background reduction of the CdWO$_4$ is on the level of 3 orders of magnitude in the energy interval 0.5…2.6 MeV and one order above 3 MeV.

PULSE SHAPE DISCRIMINATION AND ORIGIN OF ~ 940 keV PEAK

A transient digitizer (20 MS/s, 12 bit) was applied to record $CdWO_4$ pulse profiles, that allowed to identify the 940 keV peak nature. The pulse shapes acquired over 18 h were analyzed by using the optimal filter method [19]. The so called shape indicator (SI) was calculated for each event as following:

$$SI = \frac{\sum_{k=1}^{n} f(t_k) \times P(t_k)}{\sum_{k=1}^{n} f(t_k)}, \qquad (2)$$

where

$$P(t) = \frac{f_\alpha(t) - f_\gamma(t)}{f_\alpha(t) + f_\gamma(t)} \qquad (3)$$

is the weight function, $f_\alpha(t)$, $f_\gamma(t)$ are the reference pulse shapes for α particles and γ-quanta. $f(t_k)$ is amplitude of the signal in the time channel t_k.

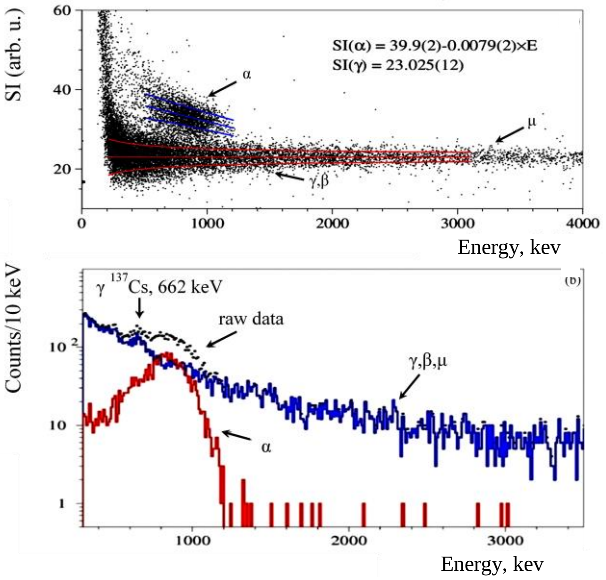

Fig. 4. Dependence of shape indicator (SI, see text) on energy of signals (in γ scale) (a). Energy spectra of γ(β,μ) and α events slected by pulse-shape discrimination (b)

Two populations of events are due to α and γ(β,μ) particles (Fig. 4,a). The energy spectra of the populations, selected using the optimal filter method, are shown in Fig. 4,b. A measurement with ^{232}Th γ-source confirmed the nature of the γ(β,μ) population, while the events above the γ band with energies in the 0.4…1.2 MeV interval are caused by α particles of the U/Th radioactive chains (taking into account the quenching of $CdWO_4$ scintillation efficiency to α particles [23]). One can assume that the α events are mainly due to decays of ^{210}Po (Q_α = 5407 keV, $T_{1/2}$ = 138 d), daughter nuclide of ^{210}Pb (^{238}U chain), that is contamination of the crystal (however, the energy of β decay of ^{210}Pb Q_β = 63.5 keV is too low to be detected in our measurements). For the moment, we cannot explain the events with energies below 0.5 MeV and shape indicator values > 30. The events can be due to surface contamination of the crystal or surroundings materials by U/Th.

Table 2

Values of full absorption peak detection efficiencies (simulated using GEANT4)

Nuclide E_γ, keV	ε, %			
	poly-styrene	quartz	titanium	copper
^{137}Cs, 662	8.48(3)	7.59(3)	6.20(2)	4.47(2)
^{228}Ac, 911	1.60(1)	1.78(1)	1.54(1)	1.16(1)
^{234m}Pa, 1001	0.055(2)	0.060(2)	0.054(2)	0.042(2)
^{40}K, 1461	0.755(9)	0.693(8)	0.595(8)	0.464(7)
^{214}Bi, 1764	0.96(1)	0.899(9)	0.791(9)	0.634(8)
^{208}Tl, 2615	3.27(2)	3.09(2)	2.79(2)	2.38(2)

SENSITIVITY TO RADIOACTIVE CONTAMINATION OF MATERIALS AND SCINTILLATORS

A potential of an experimental setup to measure low level radioactivity depends on its background, detection efficiency, energy resolution, capability to select the effect searched for. A detection limit (L_D) can be used to estimate the setup sensitivity to radioactive contamination of materials and scintillators [24, 25].

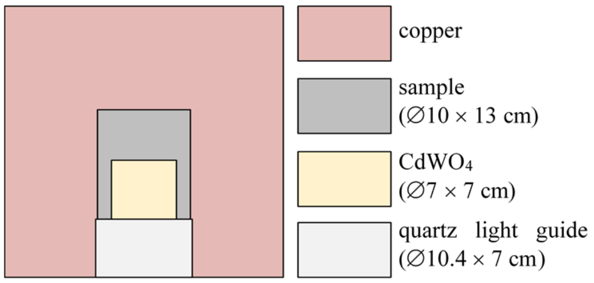

Fig. 5. Schematic view of samples in BALOO used for Monte-Carlo simulation

L_D is an activity that can not be detected with a confidence level P, over the measurement time t, in a sample with mass m. It can be obtained in an assumption that the counting rate of the effect should be on the level of p standard deviations of background counting rate n_{bg} in the region of interest ($E_\gamma \pm 1.2\sigma$ has been used as an optimal interval for γ-peaks). Assume that a standard deviations of background counting rate is equal to the square root of background events we obtain the following formula:

$$L_D \cdot m \cdot t \cdot \varepsilon \cdot \eta = p\sqrt{n_{bg} \cdot t}\,, \qquad (4)$$

where factor $p = 1$ for $P \approx 68\%$, 2 for $\approx 95.5\%$, 3 for $\approx 99.7\%$; η is quantum yield of γ-quanta per one decay; ε is full absorption peak detection efficiency. Therefore, the detection limits (for 68% confidence level (C.L.)) can be calculated as:

$$L_D = \frac{1}{m \cdot \varepsilon \cdot \eta} \sqrt{\frac{n_{bg}}{t}} \,. \qquad (5)$$

Estimates of L_D were obtained for titanium (3.36 kg), copper (6.6 kg), polystyrene (0.81 kg), and quartz (1.63 kg) samples with the same dimension in the Marinelli geometry (Fig. 5).

Table 3

Detection limits (68% C.L.) for some γ-radioactive nuclides in samples of different materials

Nuclide, (γ line)	L_D (68% C.L.), mBq/kg			
	poly-styrene	quartz	titanium	copper
^{137}Cs	1.7	0.2	0.6	1.6
^{228}Ra (^{228}Ac)	10	1.1	2.5	6.9
^{238}U (^{234m}Pa)	277	31	68	182
^{40}K	9.6	1.3	2.9	7.7
^{226}Ra (^{214}Bi)	6.7	0.9	2.0	5.0
^{228}Th (^{208}Tl)	1.9	0.3	0.5	1.3

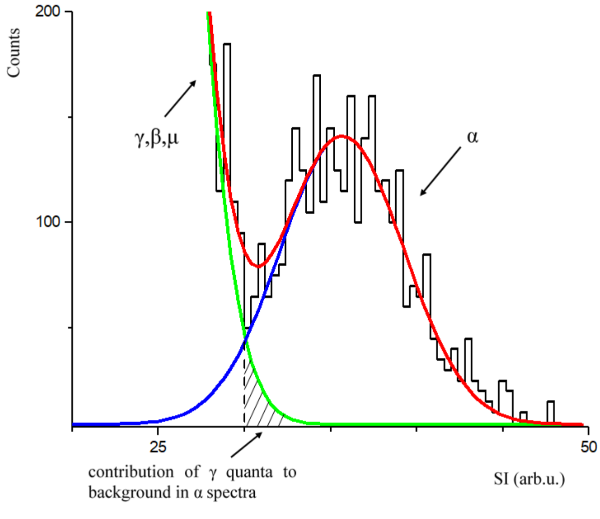

Fig. 6. Experimental distribution of shape indicator (SI, see text) for the background events in the energy interval 500…700 keV. The distribution of γ-quanta (β, μ particles) that cannot be discriminated is shown by dashed area

Response functions of the CdWO$_4$ detector to ^{40}K, ^{137}Cs, ^{208}Tl, ^{214}Bi (daughter of ^{226}Ra from the ^{238}U family), ^{228}Ac (daughter of ^{228}Ra from the ^{232}Th family), and ^{234m}Pa (^{238}U daughter) decays in the samples were Monte-Carlo simulated using the GEANT4 simulation package. The full absorption peak detection efficiencies ε for γ-quanta were obtained as a ratio of the full absorption peak area in the simulated spectra to the number of generated events (Tabl. 2).

Detection limits for ^{40}K, ^{137}Cs, ^{214}Bi, ^{228}Ac, ^{208}Tl, and ^{234m}Pa in the samples of materials were estimated using (5) over 30 d of measurements at 68% C.L. The results are presented in Tabl. 3.

The optimal filter method allows to separate α events produced by internal (surface) contamination of the crystal scintillator. In this case the background in the α spectrum can be estimated as a number of γ(β,μ) events that cannot be rejected (Fig. 6). We estimate L_D for total α activity of U/Th with daughters in a 1 kg CdWO$_4$ crystal scintillator as 0.7 mBq/kg.

Time-amplitude analysis (t-A) can be used to select events of decays in the following fast sub-chains of the ^{232}Th and ^{238}U chains:

$$({}^{228}_{90}Th)\,{}^{220}_{86}Rn_{(\alpha,\,55.6\,\mathrm{s})} \rightarrow {}^{216}_{84}Po_{(\alpha,\,0.145\,\mathrm{s})} \rightarrow {}^{212}_{82}Pb\,, \quad (6a)$$

$$({}^{226}_{88}Ra)\,{}^{214}_{83}Bi_{(\beta,\,19\,\mathrm{m})} \rightarrow {}^{214}_{84}Po_{(\alpha,\,164\,\mu\mathrm{s})} \rightarrow {}^{210}_{82}Pb\,. \quad (6b)$$

It should be stressed that ^{220}Rn and ^{216}Po are in equilibrium with ^{228}Th, while ^{214}Bi and ^{214}Po are in equilibrium with ^{226}Ra. Therefore, L_D for ^{228}Th and ^{226}Ra can be evaluated using time-amplitude analysis of (6a) and (6b) decay chains. Background events in the t-A analysis may occur due to the random coincidences of events. The background counting rate (n) can be evaluated as [26]:

$$n = (n_1 n_2 + A\eta_1\varepsilon_1 n_2 + A\eta_2\varepsilon_2 n_1)\Delta t\,, \qquad (7)$$

where n_1, n_2 are the counting rates in the energy intervals of the first and second events selection, respectively; $\Delta t = t_2 - t_1$ is the time window used for events selection (the value of Δt should be chosen taking into account the half-lives of the fast decay nuclides in the sub-chains); A is the activity of the parent nuclide in the detector; ε_1 and ε_2 are the detection efficiencies of the first and second events, respectively. The L_D in this case can be evaluated as following (using a condition $A << n_i$, 68% C.L.):

$$L_D = \frac{1}{\eta_1\eta_2\varepsilon_1\varepsilon_2\lambda} \sqrt{\frac{n_1 n_2 \Delta t}{t}}\,, \qquad (8)$$

where λ is the probability for second event from the sequential decays to be registered in a time window t_2 - t_1:

$$\lambda = e^{\left(-\frac{t_1}{T_{1/2}}\ln 2\right)} - e^{\left(-\frac{t_2}{T_{1/2}}\ln 2\right)}\,, \qquad (9)$$

where t_1 is limited by the time resolution of the detector, and an optimal value for Δt is ~ $1.8T_{1/2}$ [26]. A summary of the detector sensitivities (L_D) to ^{226}Ra, ^{228}Th and total alpha activity of U/Th nuclides in a 1 kg scintillator over 30 d of measurement is given in Tabl. 4.

Table 4

Detection limits (68% C.L.) for ^{226}Ra, ^{228}Th and total α activity of U/Th in 1 kg scintillator over 30 d of measurement

Nuclide	Method	L_D, mBq/kg
^{226}Ra	*t-A*	0.3
^{228}Th	*t-A*	0.02
α active U and Th daughters	pulse shape discrimination	0.7

CONCLUSIONS

The radioactive background of 2.128 kg $CdWO_4$ scintillation detector was investigated in different shield concepts over ~ 2000 h. The background counting rate was reduced by one order of magnitude above 3 MeV, and by 3 orders in the energy interval 0.5…2.6 MeV. The reduction was achieved by application of passive shield (15 cm of old lead, 6…12 cm of OFHC copper), plastic scintillation counter to veto cosmic muon signals, and pulse shape discrimination of fast signals (pile-ups) due to the Cherenkov radiation of muons in the quartz light guide. A peak with energy ~ 940 keV in the spectrum of the $CdWO_4$ detector can be explained by ^{210}Po (α active daughter nuclide of ^{210}Pb from the ^{238}U chain). Further background suppression can be achieved by development of additional plastic scintillation counters of the muon veto counter (in progress); by cleaning of the detector and copper shield details to remove surface radioactive contaminations (particularly, technogenic ^{137}Cs); by applying nitrogen flow through the setup to displace radioactive radon; and by pulse shape discrimination and time-amplitude analysis (to remove fast decay sub-chain and alpha events from internal and surface contamination of the crystal scintillator by ^{232}Th, ^{238}U, ^{235}U, and their daughters).

Sensitivity of the setup to radioactive contamination of several materials (polystyrene, quartz, titanium and copper) was estimated. For instance, the detection limit is on the level of 2.9 mBq/kg (^{40}K); 0.6 mBq/kg (^{137}Cs); 2 mBq/kg (^{226}Ra), and 0.5 mBq/kg (^{228}Th) for a 3.36 kg titanium sample over 30 days of measurements with 68% C.L. Sensitivity to radioactive contamination in scintillators is on the level of 0.3 mBq/kg (^{226}Ra); 0.02 mBq/kg (^{228}Th), and 0.7 mBq/kg for α radioactive daughters of the U and Th families (in 1 kg scintillator over 30 d of measurements with 68% C.L.). The sensitivity level is comparable to those achieved with underground low background HPGe detectors, that makes the BALOO setup a high-performance low-background scintillation spectrometer to R&D of radiopure scintillators and materials, and to realize low scale low counting experiments.

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НИЗКОФОНОВАЯ СЦИНТИЛЛЯЦИОННАЯ УСТАНОВКА ДЛЯ ИССЛЕДОВАНИЯ РАДИОАКТИВНОЙ ЧИСТОТЫ МАТЕРИАЛОВ

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Снижение фона низкофоновой сцинтилляционной установки со сцинтилляционным кристаллом $CdWO_4$ массой 2,128 кг составило три порядка в области энергий 0,5…2,6 МэВ и один порядок при энергиях больше 3 МэВ. Подавление фона было достигнуто путем установки радиоактивно чистой пассивной защиты, пластиковых сцинтилляторов для отбрасывания сигналов от космических частиц и разделением сигналов от черенковского свечения в световоде детектора по форме. Оценена чувствительность установки к радиоактивным загрязнениям разных материалов (с доверительной вероятностью 68%), например, на уровне 2,9 мБк/кг (^{40}K); 0,6 мБк/кг (^{137}Cs); 2 мБк/кг (^{226}Ra) и 0,5 мБк/кг (^{228}Th) для образца титана массой 3,36 кг за время измерения 30 сут.

НИЗЬКОФОНОВА СЦИНТИЛЯЦІЙНА УСТАНОВКА ДЛЯ ДОСЛІДЖЕННЯ РАДІОАКТИВНОЇ ЧИСТОТИ МАТЕРІАЛІВ

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Зниження фону низькофонової сцинтиляційної установки із сцинтиляційним кристалом $CdWO_4$ масою 2,128 кг складає три порядки в області енергій 0,5…2,6 МеВ і один порядок для енергій більше 3 МеВ. Придушення фону було досягнуто встановленням радіоактивно чистого пасивного захисту, пластикових сцинтиляторів для відкидання сигналів від космічних частинок і розділенням сигналів від черенковського свічення у світловоді детектора за формою. Оцінено чутливість установки до радіоактивних забруднень різних матеріалів (з довірчою ймовірністю 68%), наприклад, на рівні 2,9 мБк/кг (^{40}K); 0,6 мБк/кг (^{137}Cs); 2 мБк/кг (^{226}Ra) і 0,5 мБк/кг (^{228}Th) для зразку титану масою 3,36 кг протягом 30 діб вимірювання.