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CARVEL experiment with $^{48}CaWO_4$ crystal scintillators for the double β decay study of ^{48}Ca

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This paper is dedicated to memory of Yuri Georgievich Zdesenko, Doctor of Physical and Mathematical Sciences, Professor, Corresponding Member of the Ukrainian National Academy of Sciences, Physicist, Man and Teacher, who made during his life important contributions to studies of double beta decay and other rare nuclear processes

Abstract

The CARVEL (CAlcium Research for VEry Low neutrino mass) experiment to search for the double β decay of ^{48}Ca with the help of enriched $^{48}CaWO_4$ crystal scintillators has been considered. Scintillation properties (energy resolution, α/β ratio, pulse-shape discrimination ability) and radiopurity of $CaWO_4$ scintillators were studied. Despite rather high radioactive contaminations, the background rate of the $CaWO_4$ detector in the energy region 3.6–5.4 MeV (energy window of the ^{48}Ca neutrinoless 2β decay) was reduced down to 0.07 counts/(yr keV kg). With ≈100 kg array of the $^{48}CaWO_4$ crystals the sensitivity of the CARVEL experiment (in terms of the half-life limit for the 0ν2β decay) is estimated as $T_{1/2}^{0\nu} > 10^{27}$ yr. This value corresponds to the neutrino mass constraint $m_\nu < (0.04$–$0.09)$ eV.

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1. Introduction

The great interest to the neutrinoless (0ν) double beta decay [1–5] have arisen from the recent observations of neutrino oscillations [6–9], strongly suggesting that neutrino have nonzero mass (m_ν). Indeed, the 0ν2β decay—forbidden in the Standard Model (SM) since it violates lepton number (L) conservation—requires neutrinos to be massive Majorana particles [10]. At the same time, many extensions of the SM incorporate L violating interactions and, thus, could lead to this process, which, if observed, will be a clear evidence for a new physics beyond the SM and an unique confirmation of the Majorana nature of the neutrino. While oscillation experiments are sensitive to the neutrino mass difference, only the measured 0ν2β decay rate can give the absolute scale of the effective Majorana neutrino mass, which could allow one to test different neutrino mixing models.

Despite numerous efforts, the 0ν2β decay still remains unobserved (see latest reviews [1–5,11]) and only half-life limits for 0ν mode were set in direct measurements with several nuclides: $T_{1/2}^{0\nu} \geqslant 10^{22}$ yr for ^{48}Ca [12]; $T_{1/2}^{0\nu} \geqslant 10^{23}$ yr for ^{82}Se, ^{100}Mo [13], ^{116}Cd [14], ^{128}Te, ^{130}Te [15], ^{136}Xe [16], and $T_{1/2}^{0\nu} \geqslant 10^{25}$ yr for ^{76}Ge [17,18]. The best of them are given in Table 1. These bounds have already brought the most stringent restrictions on the values of the Majorana neutrino mass ($m_\nu \leqslant 0.3$–2 eV), the right-handed admixture in the weak interaction ($\eta \approx 10^{-8}$, $\lambda \approx 10^{-6}$), the neutrino-Majoron coupling constant ($g_M \approx 10^{-4}$), and the R-parity violating parameter of minimal supersymmetric standard model ($\approx 10^{-4}$) [1–3,11].

Moreover, nowadays the 2β decay research is entering new era, when *discovery* of the 0ν2β decay has become realistic. As it is suggested by oscillation data, this goal could be reached if the present level of the experimental sensitivity will be enhanced to $m_\nu \approx 0.01$ eV–0.05 eV [21–23]. However, at the end of 2001 the discovery of the neutrinoless 2β decay of ^{76}Ge with half-life of 1.5×10^{25} yr ($m_\nu = 0.39$ eV) has been claimed [24], which then was strongly criticized in Refs. [25,26,20]. Nevertheless, recently Heidelberg group has claimed again their observation of the effect, but now with 4σ confidence level and with the best fit corresponding to neutrino mass of ≈0.4 eV [27]. It is stated by authors that improved statistical significance is due to slightly increased exposure (additional ≈17 kg × yr) and mostly due to careful re-analysis of their previous data (≈55 kg × yr). Hence, in order to check this intriguing claim, it would be very important now to realize another experiment with ^{76}Ge as well as experiments with other nuclides, which can be constructed during reasonable time and will be really able to observe the 0ν2β decay rate corresponding to neutrino mass $m_\nu \approx 0.1$ eV.

Table 1
The best reported $T_{1/2}^{0\nu}$ and m_ν limits from direct 2β decay experiments

Nuclide	Experimental limit $T_{1/2}^{0\nu}$ (yr)		Reference	Limit on m_ν (eV) on the basis of [19][a]	
	68% C.L.	90% C.L.		68% C.L.	90% C.L.
^{76}Ge	3.1×10^{25}	1.9×10^{25}	[17]	0.27	0.35
	–	1.6×10^{25}	[18]	–	0.38
	4.2×10^{25}[b]	2.5×10^{25}[b]	[20]	0.24	0.31
^{82}Se	–	1.4×10^{23}	[13]	–	2.1
^{100}Mo	–	3.1×10^{23}	[13]	–	2.1
^{116}Cd	2.6×10^{23}	1.7×10^{23}	[14]	1.4	1.7
^{130}Te	–	5.5×10^{23}	[15]	–	0.94
^{136}Xe	–	4.4×10^{23}	[16]	–	2.2

[a] The NME calculations of Ref. [19] were chosen because of the most extensive list of 2β nuclei calculated in that work, allowing one to compare the sensitivity of different experiments to the m_ν bound within the same scale.

[b] Results were established [20] by analyzing the cumulative data sets of the Heidelberg-Moscow [17] and IGEX [18] experiments.

1.1. Sensitivity of the 2β decay experiments

In order to determine factors, on which sensitivity of 2β decay experiments depends on, let us consider carefully Table 1 with the best results. One can note that most of them were obtained by using so-called "active" source technique, in which a detector, containing 2β candidate nuclei, serves simultaneously as source. This technique has the following advantages: (a) 2β source is measured in 4π geometry and self-absorption of the electrons in a source is absent, (b) the energy resolution of the detector does not depend on the angular and energy distribution of the electrons emitted in 2β decay. The points (a) help to increase the detection efficiency (which is typically close to 100%), while (b) is useful to discriminate the effect from background.

There exists another class of 2β decay experiments with "passive" source, where the latter is placed (e.g., in form of foil) between two detectors. The advantage of this approach is the possibility to obtain (with the help of proper detectors) full information about 2β decay event: time coincidence, tracks and vertex of the emitted electrons, their energies and angular distribution. However, self-absorption of emitted electrons in the source decreases detection efficiency and causes the broadening and shifting of the $0\nu2\beta$ decay peak to the lower energies, which make it difficult to discriminate the effect from background.

It is also interesting to stress that experiments, other than those with ^{76}Ge, offer m_ν bounds in the range of $\approx$1–2 eV, which is not so drastically weaker, especially taking into account that, e.g., ^{116}Cd result was obtained with the small detector ($^{116}CdWO_4$ crystal scintillators with $\approx$0.1 kg of ^{116}Cd) [14] in contrast with those used for ^{76}Ge studies ($\approx$10 kg of enriched HP ^{76}Ge detectors [17,18]). This fact could be explained mainly due to the larger energy release available for 2β decay of ^{116}Cd ($Q_{\beta\beta}$ = 2809 keV) as compared with that of ^{76}Ge ($Q_{\beta\beta}$ = 2039 keV). First, it is because that the $0\nu2\beta$ decay probability strongly depends on $Q_{\beta\beta}$ value, roughly as $Q^5_{\beta\beta}$ [28]. Secondly, the larger the 2β decay energy, the simpler it is—from an experimental point of view—to overcome background problems. Let us remember that the background from natural radioactivity drops sharply above 2615 keV, which is the energy of γ's from ^{208}Tl decay (^{232}Th family).

All these facts demonstrate the importance of the development of new detectors, containing 2β decay candidate nuclides with as large $Q_{\beta\beta}$ energy release as possible. Among 35 double β^- decay candidates available in nature, there are only six (including ^{116}Cd), whose $Q_{\beta\beta}$ values exceed 2.6 MeV: ^{48}Ca (4274 keV), ^{82}Se (2995 keV), ^{96}Zr (3348 keV), ^{100}Mo (3035 keV), and ^{150}Nd (3368 keV) [29].

Since ^{48}Ca has the highest $Q_{\beta\beta}$ energy, the development of the appropriate detector with Ca nuclei would be of considerable interest for the future 2β decay experiments. However, apart from a reasonable content of candidate nuclei, detector for 2β study should satisfy certain demands to its radiopurity, because the latter restricts the level of residual background, which could be reached with the detector. Besides, the energy resolution of the detector is a very important characteristic. Foremost, with the high energy resolution it is possible to minimize the irreducible background produced by the $2\nu2\beta$ decay events. It is because for the case of a poor resolution, the events from the high energy tail of the 2ν distribution could run into the energy window of the 0ν peak and, thus, generate the background which cannot be discriminated from the $0\nu2\beta$ decay signal, even in principle. However, the better is the energy resolution, the smaller part of the 2ν tail can fall within the 0ν interval [3,30,31].

Likewise, the role of the energy resolution of the detector is even more crucial for the discovery of the $0\nu2\beta$ decay. Indeed, this process manifests itself by the peak at $Q_{\beta\beta}$ energy, whose width is determined by the energy resolution of the detector. Hence, the latter should be sufficient to discriminate this peak from background and to recognize the effect [31].

One possible detector for the 2β decay study of ^{48}Ca is the calcium tungstate ($CaWO_4$) crystal scintillator [32], whose physical properties are similar to those of $CdWO_4$ scintillators already applied in the 2β experiment with ^{116}Cd [14]. Moreover, $CaWO_4$ scintillators were considered [33,34] as promising detectors for the dark matter particle quest. [1] The reason is that these crystals

[1] Weakly interacting massive particles (WIMPs)—in particular neutralino, predicted by the Minimal Supersymmetric extension of the SM—are considered as possible component of the dark matter in the Universe. It is supposed that WIMPs interact with matter through scattering on nuclei, and hence, producing low energy nuclear recoils. At present the most sensitive experiments for WIMPs search apply different detectors: Ge semiconductor detectors, low temperature bolometers, and scintillators (see reviews [35]).

can also work as cryogenic detectors, and hence, allows one to detect phonon and scintillation light signals simultaneously. Such an approach could provide a powerful tool for discrimination of the effect (signals of recoil nuclei) from the background events caused by electrons, alpha particles, etc. Because of an extremely low counting rate expected in direct WIMPs experiments, and because of small energy of recoil nuclei, the WIMPs searches require detectors with the low energy threshold (≈10 keV), and with the low level of background, that is with the tiny radioactive contaminations.

The purpose of our work is to study the scintillation properties and radioactive contamination of the $CaWO_4$ crystal scintillators and to develop on this basis the high sensitivity experiment for the double beta decay searches.

2. Measurements and results

For our studies three clear, colorless $CaWO_4$ crystals (20 × 20 × 10 mm, 40 × 34 × 23 mm, and ∅40 × 39 mm) were grown by Czochralski method. Their scintillation properties were studied and reported in [36]. The main characteristics of the $CaWO_4$ scintillators are presented in Table 2 together with those of well known cadmium tungstate ($CdWO_4$) scintillators given for comparison. Both crystals are chemically resistant and non-hygroscopic. Radioactive contaminations of the crystals were measured in the low background set-up installed in the Solotvina Underground Laboratory (built in a salt mine 430 m underground or ≃1000 m of water equivalent) [37]. In the set-up a scintillation crystal was viewed by the special low-radioactive 5″ photomultiplier tube (EMI D724KFLB) through the high purity quartz light-guide ∅10 × 33 cm. The detector was surrounded by a passive shield made of teflon (thickness of 3–5 cm), plexiglass (6–13 cm), high purity copper (3–6 cm), lead (15 cm) and polyethylene (8 cm). Two plastic scintillators (120 × 130 × 3 cm) installed above the passive shield were used as a cosmic muons veto.

Because of rather long decay time of $CaWO_4$ scintillators, a special electronic unit [38] was used for spectrometric measurements. Event-by-event data acquisition records information on the amplitude (energy), arrival time and pulse shape of a signal. For the latter, a transient digitizer based on the fast 12 bit ADC was used with the sample frequency of 20 MS/s [39]. Pulse shapes of events in the chosen energy interval (usually, higher than ≈180 keV) were recorded in 2048 channels with 50 ns channel's width. The pulse-shape analysis, based on the optimal digital filter, ensures clear discrimination between γ rays and α particles, as well as rejection of false events, like double pulses, etc. [39,40]. Besides, the technique of the time–amplitude analysis of background data (described, e.g., in [14,41]) was used for recognition and selection of the short-living chains from ^{232}Th, ^{235}U and ^{238}U families (trace contaminations of the $CaWO_4$ crystals).

Table 2
Properties of $CaWO_4$ and $CdWO_4$ crystal scintillators

Physical parameter	$CaWO_4$	$CdWO_4$
Density (g/cm^3)	6.1	8.0
Melting point (°C)	1570–1650	1325
Structural type	Sheelite	Wolframite
Cleavage plane	(101)	(010)
Hardness (Mohs)	4.5–5	4–4.5
Wavelength of emission maximum (nm)	420–430	480
Refractive index	1.94	2.2–2.3
Effective average decay time[a]	8 μs	13 μs
Photoelectron yield relative to NaI(Tl)[a]	≈18%	≈20%

[a] For γ rays, at indoor temperature.

2.1. *Response of $CaWO_4$ scintillators to γ rays and α particles*

The $CaWO_4$ crystals, wrapped by teflon reflector tape, were viewed by the photomultiplier tube (PMT) Philips XP2412. The linearity, energy scale and resolution of the detectors were measured with γ sources ^{60}Co, ^{137}Cs, ^{207}Bi, ^{232}Th and ^{241}Am in the energy range of 60–2615 keV. As an example, the energy spectra measured by $CaWO_4$ scintillator (∅40 × 39 mm) with ^{137}Cs and ^{60}Co gamma sources are shown in Fig. 1a. Note, that the resolution FWHM = 7.8% at

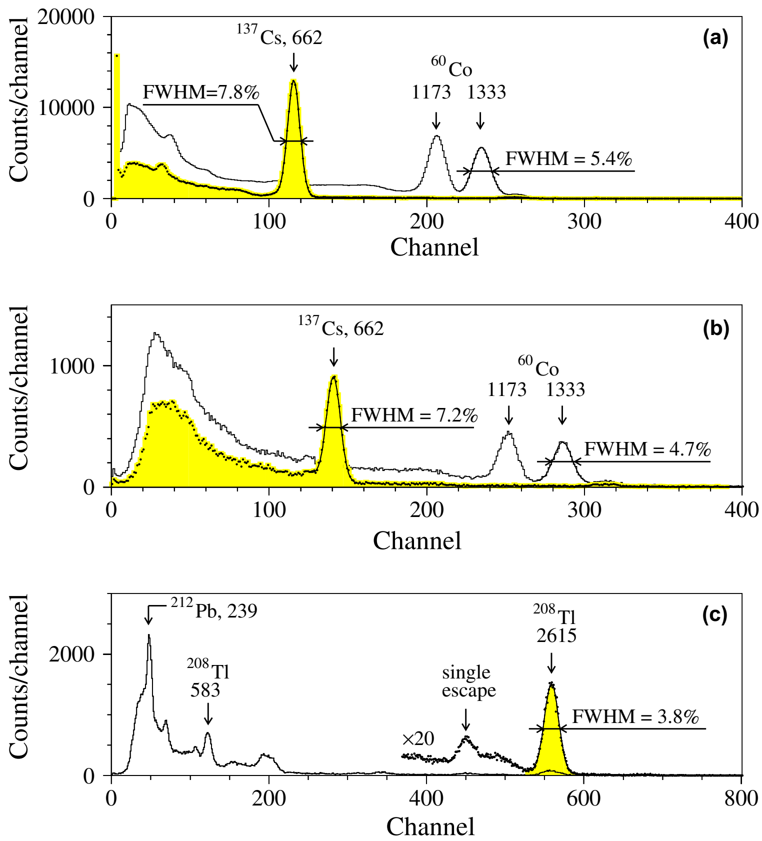

Fig. 1. Energy spectra measured by a $CaWO_4$ crystal (∅40 × 39 mm) with ^{137}Cs and ^{60}Co sources for two arrangements: (a) standard, where the crystal wrapped by teflon reflector was directly coupled to PMT with optical glue; (b) the $CaWO_4$ crystal was placed in liquid and viewed by two distant PMTs (see text). (c) The same as (b) but with ^{232}Th source. Energies of γ lines are in keV.

662 keV is similar to that obtained with NaI(Tl) scintillators.

The crystal response to α particles (in the energy range of 0.5–5.3 MeV) was measured with the help of collimated beam of α particles from a ^{241}Am source and various sets of the thin mylar film absorbers (see for details [40]). Besides, α peaks of ^{147}Sm and nuclides from the ^{232}Th, ^{235}U and ^{238}U chains, present in trace amount in the $CaWO_4$ crystal (see below), were used to extend the energy interval up to ≃8 MeV. These peaks were selected with the help of the pulse-shape and time–amplitude analysis of the data obtained in the low background measurements with the $CaWO_4$ crystal. Within the energy interval 0.5–2 MeV the measured α/β ratio [2] decreases with increasing energy: $\alpha/\beta = 0.21(2) - 2.3(14) \times 10^{-5} E_\alpha$, while it increases above 2 MeV: $\alpha/\beta = 0.129(12) + 2.1(3) \times 10^{-5} E_\alpha$, where E_α is in keV. Because quenching of the scintillation light is due to higher ionization density caused by α particles in comparison with β particles [42], the behaviour of the α/β ratio can be explained by the energy dependence of ionization density for α particles. The $CaWO_4$ crystals were irradiated in the

[2] The "α/β ratio" is defined as ratio of α peak position measured in the γ energy scale to the energy of α particles. Because γ quanta interact with detector by β particles we use more convenient term "α/β ratio".

directions perpendicular to main crystal planes with aim to check a possible dependence of the α signal on direction of irradiation. While earlier such a dependence has been found for $CdWO_4$ [40], we did not observe it for $CaWO_4$ crystals.

2.2. Pulse-shape discrimination

The pulse shape of $CaWO_4$ scintillation signals can be described by the formula: $f(t) = \sum A_i/(\tau_i - \tau_0) \times (e^{-t/\tau_i} - e^{-t/\tau_0})$, $t > 0$, where A_i are amplitudes (in %) and τ_i are decay constants for different light emission components, τ_0 is integration constant of electronics (≈0.2 μs). The following values were obtained by fitting the average of 4 thousand of individual pulses: $A_1^\alpha = 76\%$, $\tau_1^\alpha = 8.8$ μs, $A_2^\alpha = 18\%$, $\tau_2^\alpha = 3.2$ μs, $A_3^\alpha = 6\%$, $\tau_3^\alpha = 0.3$ μs for ≈4.6 MeV α particles and $A_1^\gamma = 82\%$, $\tau_1^\gamma = 9.0$ μs, $A_2^\gamma = 15\%$, $\tau_2^\gamma = 4.4$ μs, $A_3^\gamma = 3\%$, $\tau_3^\gamma = 0.3$ μs for ≈1 MeV γ quanta. This difference allows one to discriminate γ(β) events from those of α particles. For this purpose we used the method of the optimal digital filter (for the first time proposed in [43]), which previously was successfully applied with $CdWO_4$ scintillators [39]. To obtain the numerical characteristic of $CaWO_4$ signal, called the shape indicator (SI), each experimental pulse was processed with the following digital filter: $\mathrm{SI} = \sum f(t_k) \times P(t_k)/\sum f(t_k)$, where the sum is over time channels k, starting from the origin of pulse and up to 75 μs, $f(t_k)$ is the digitized amplitude (at the time t_k) of the signal. The weight function $P(t)$ is defined as: $P(t) = \{f_\alpha(t) - f_\gamma(t)\}/\{f_\alpha(t) + f_\gamma(t)\}$, where $f_\alpha(t)$ and $f_\gamma(t)$ are the reference pulse shapes for α particles and γ quanta.

The shape indicator measured by $CaWO_4$ crystals for alpha particles in the (1–5.3) MeV region depends on energy [36] and does not depend on the direction of α irradiation relative to the crystal axes. For γ quanta no dependence of the SI on the energy (from 0.1 to 2.6 MeV) was observed. The distributions of the shape indicator measured with α particles ($E_\alpha \approx 5.3$ MeV) and γ quanta (≈1.2 MeV) are depicted in the inset of Fig. 2 (the larger value of the shape indicator corresponds to the shorter decay time of the scintillation pulse). As it is seen, distinct discrimination between α particles and γ rays (β particles) was achieved. As an illustration of the PS analysis, the background data (accumulated during 171 h with $CaWO_4$ detector) is shown in Fig. 2 as scatter plot for the SI values versus energy. In this plot one can see two clearly separated populations: the α events, which belong to U/Th families, and γ(β) events.

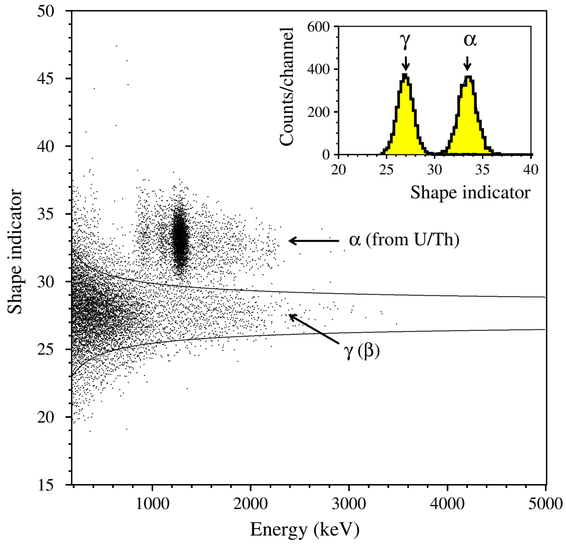

Fig. 2. Scatter plot of the shape indicator SI versus energy for 171 h background data measured with the $CaWO_4$ crystal scintillator (40 × 34 × 23 mm). Lines show ±2σ region of SI for γ(β) events. (Inset) The SI distributions measured in calibration runs with α particles (E_α = 5.3 MeV which corresponds to ≈1.2 MeV in γ scale) and γ quanta (≈1.2 MeV).

2.3. Background and radioactive contamination of the $CaWO_4$ crystal

As mentioned earlier, the $CaWO_4$ crystal (40 × 34 × 23 mm) was measured with the help of the low background set-up, installed in the Solotvina Underground Laboratory. The energy resolution of the detector was determined with several γ sources (^{60}Co, ^{137}Cs, ^{207}Bi, ^{232}Th and ^{241}Am) and can be fitted in the energy interval 60–2615 keV by the function: $\mathrm{FWHM}_\gamma(\mathrm{keV}) = -3 + \sqrt{6.9E_\gamma}$, where E_γ is the energy of the γ quanta in keV. The routine calibrations were carried out weekly with ^{207}Bi and ^{232}Th sources.

The energy spectrum of the $CaWO_4$ detector, measured during 1734 h in the low background apparatus, is presented in Fig. 3, where the data obtained with the $CdWO_4$ scintillator in the same set-up are shown for comparison. Both spectra are normalized by their measuring time and the corresponding detector mass. It is clear from this comparison, that radioactive contamination of the $CaWO_4$ crystal is much higher than that of the $CdWO_4$ one. With the aim to recognize the origins of these contaminations, the data accumulated with the $CaWO_4$ crystal were separated into α and β spectra with the help of the pulse-shape discrimination technique.

First, the α spectrum, which is depicted in Fig. 4, was analyzed. The total internal alpha activity in the crystal is ≈0.4 Bq/kg. The intense and clear peak at the energy 1.28 MeV is attributed to intrinsic ^{210}Po (daughter of ^{210}Pb from the ^{238}U family) with activity of 0.291(5) Bq/kg. Apparently, the equilibrium of the uranium chain in the crystal was broken during crystal production, because the peak of ^{238}U (see Inset (a) in Fig. 4) corresponds to a much lower activity of 14.0(5) mBq/kg. Peaks of the uranium's daughters ^{234}U, ^{230}Th, ^{226}Ra are not resolved (their Q_α values are very close), however, the area of the total peak (at ≈1.1 MeV) is in satisfactory agreement with the activity of ^{238}U and ^{226}Ra. Another member of the family, ^{222}Rn, is not discriminated from ^{210}Po (an expected energy of α peak is ≈1.34 MeV in γ scale), while ^{218}Po peak is well resolved. The activity of ^{226}Ra determined on the basis of the ^{218}Po peak area is 5.9(8) mBq/kg.

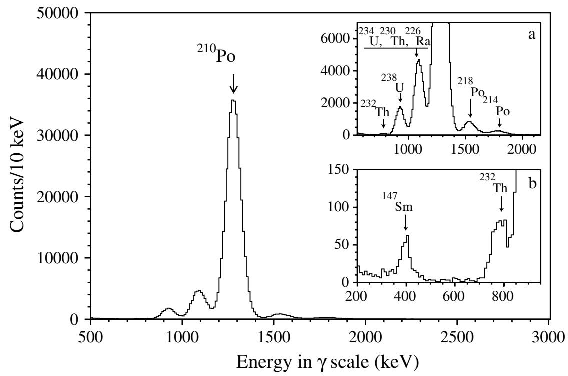

Fig. 4. Energy spectrum of α events selected by the pulse-shape analysis from background data of Fig. 3 measured with the $CaWO_4$ detector. (Inset a) The same spectrum but scaled up. It is well reproduced by the model, which includes α decays of nuclides from ^{232}Th and ^{238}U families. (Inset b) Low energy part of the α spectrum.

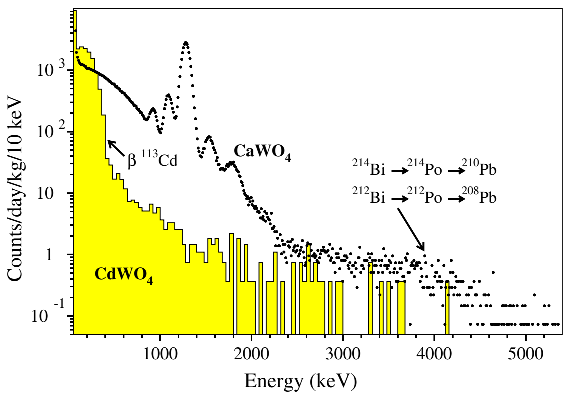

Fig. 3. Energy spectra of $CaWO_4$ (mass of 0.189 kg, during 1734 h, dots) and $CdWO_4$ (0.448 kg, 36.6 h, filled histogram) scintillation crystals measured in the low background set-up. Beta decay of ^{113}Cd (Q_β = 316 keV, $T_{1/2} = 7.7 \times 10^{15}$ yr) dominates in the low energy part of the $CdWO_4$ background. Broad distribution in the spectrum of the $CaWO_4$ detector after 3 MeV is caused by the fast sequences ^{214}Bi → ^{214}Po → ^{210}Pb and ^{212}Bi → ^{212}Po → ^{208}Pb (see text).

In the low energy part of alpha spectrum (Inset (b) in Fig. 4) the peak at the energy ≈0.8 MeV can be attributed to ^{232}Th with an activity of 0.69(10) mBq/kg. A weak alpha peak with the energy in γ scale of 395(2) keV (corresponds to the energy of α particles 2243(9) keV) can be explained by traces of ^{147}Sm (E_α = 2247 keV, $T_{1/2} = 1.06 \times 10^{11}$ yr, whose isotopic abundance is 15.0% [44]) with an activity 0.49(4) mBq/kg. The presence of ^{147}Sm in $CaWO_4$ crystal (at the level of ≈6 mBq/kg) was also observed in Ref. [34], where this crystal was operated as a low temperature bolometer with very good energy resolution for alpha particles.

Besides, the raw background data (Fig. 3) were analyzed by the time–amplitude method, when the energy and arrival time of each event were used for selection of some decay chains in ^{232}Th, ^{235}U and ^{238}U families. For instance, the following sequence of α decays from the ^{232}Th family (which is in equilibrium with ^{228}Th) was searched for and observed: ^{220}Rn (Q_α = 6.40 MeV) → ^{216}Po (Q_α = 6.91 MeV, $T_{1/2}$ = 0.145 s) → ^{212}Pb. Because the energy of α particles from ^{220}Rn decay corresponds to

$\simeq$1.6 MeV in γ scale of $CaWO_4$ detector, the events in the energy region 1.4–2.2 MeV were used as triggers. Then all events (with appropriate energies) following the triggers in the time interval 20–600 ms (containing 85% of ^{216}Po decays) were selected. The obtained α peaks (the α nature of events was confirmed by the pulse-shape analysis described above), as well as the distributions of the time intervals between events are in a good agreement with those expected for α particles of $^{220}Rn \rightarrow {}^{216}Po \rightarrow {}^{212}Pb$ chain [45] (see Fig. 5). On this basis the activity of ^{228}Th in the $CaWO_4$ crystal was calculated as 0.6(2) mBq/kg, which is in a good agreement with activity of ^{232}Th (0.69(10) mBq/kg) determined from α spectrum. The α peak with the energy $E_\alpha \approx 7.3$ MeV, which is present in the energy distribution of the second event, can be attributed to ^{215}Po from ^{235}U family. Corresponding activity of ^{227}Ac in the crystal is 1.6(3) mBq/kg.

Similarly for the analysis of the ^{226}Ra chain (^{238}U family) the following sequence of β and α decays was used: ^{214}Bi ($Q_\beta = 3.27$ MeV) $\rightarrow$ ^{214}Po ($Q_\alpha = 7.83$ MeV, $T_{1/2} = 164$ μs) $\rightarrow$ ^{210}Pb. To select the β decays of the ^{214}Bi, the lower energy threshold was set at 0.18 MeV, while for the α decay of ^{214}Po the energy window 1.6–2.4 MeV was chosen. Time interval of 90–500 μs (56% of ^{214}Po decays) was used. The obtained spectra lead to the ^{226}Ra activity in the $CaWO_4$ crystal equal to 5.6(5) mBq/kg, which value agrees with that derived on the basis of ^{218}Po peak in α spectrum—5.9(8) mBq/kg.

Finally, let us analyze the energy spectrum of $\beta(\gamma)$ events selected with the help of the pulse-shape technique and presented in Fig. 6. The background in the very low energy part of the β spectrum (see Inset in Fig. 6) is mainly due to β decay of ^{210}Pb ($Q_\beta = 64$ keV), whose measured activity is consistent with that determined from the α peak of ^{210}Po. The counting rate for the $\beta(\gamma)$ spectrum above the energy threshold of 0.2 MeV is $\approx$0.45 counts/(s kg). The contribution of the external γ rays to this background rate was estimated as small as $\approx$2%, by using results of measurements with $CdWO_4$ crystal (mass of 0.448 kg) installed in the same low background set-up (see Fig. 3). Therefore, the remaining $\beta(\gamma)$ events are caused by the intrinsic contaminations of the $CaWO_4$ crystal. The major part of this β activity can be ascribed to: ^{210}Bi (daughter of ^{210}Pb)—$\approx$0.3 Bq/kg; ^{234m}Pa–14 mBq/kg; ^{214}Pb and ^{214}Bi (^{238}U family)—$\approx$9 mBq/kg; ^{211}Pb (^{235}U family)—$\approx$2 mBq/kg. The residual ($\approx$0.13 counts/

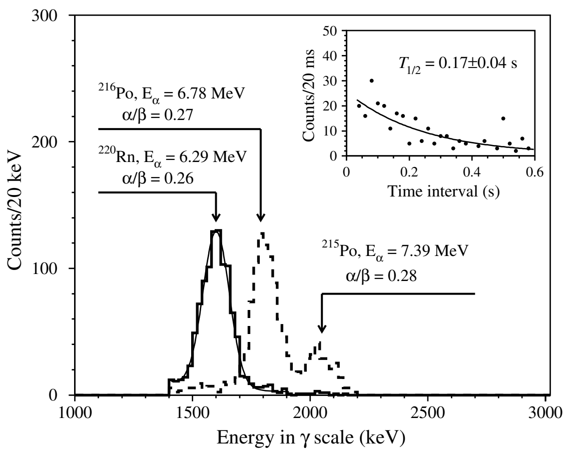

Fig. 5. Alpha peaks of ^{220}Rn and ^{216}Po selected by the time–amplitude analysis from the data accumulated with the $CaWO_4$ detector. (Inset) The distribution of the time intervals between the first and second events (dots) together with the exponential fit (line). Obtained half-life of ^{216}Po (0.17 ± 0.04 s) is in an agreement with the table value: 0.145(2) s [45].

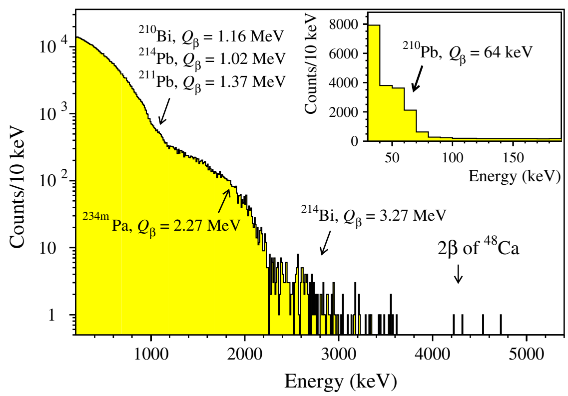

Fig. 6. Energy spectrum of $\beta(\gamma)$ events selected by the pulse-shape analysis from background data of Fig. 3 measured with the $CaWO_4$ detector. It is described by β spectra of ^{210}Bi ($Q_\beta = 1.16$ MeV), ^{214}Pb ($Q_\beta = 1.02$ MeV), ^{211}Pb ($Q_\beta = 1.37$ MeV), ^{234m}Pa ($Q_\beta = 2.27$ MeV), and ^{214}Bi ($Q_\beta = 3.27$ MeV). (Inset) In the low energy region the background (measured during 15.8 h) is caused mainly by β decay of ^{210}Pb ($Q_\beta = 64$ keV).

(s kg)) could be explained by broken equilibrium in ^{210}Bi → ^{210}Po chain, and/or by other β active impurities (^{40}K, ^{90}Sr in equilibrium with ^{90}Y, ^{137}Cs, etc.) probably present in the crystal.

The summary of the measured radioactive contamination of the $CaWO_4$ crystal scintillator (or limits on their activities) is given in Table 3, again in comparison with the $CdWO_4$ detectors [14,38,46]. From this comparison one can see that radioactive impurities in the $CaWO_4$ crystals (available at present) are much higher (by factor of 10–10^3) than those of the $CdWO_4$ scintillators. To understand the origin of the radioactive contamination of the $CaWO_4$ scintillator, row materials for $CaWO_4$ crystal growing (calcium carbonate and tungsten oxide) were measured in the low background set-up, based on $CdWO_4$ scintillator of large mass (≈1 kg), in the Solotvina Underground Laboratory. It is clear from these measurements, than the large U/Th contamination of the $CaWO_4$ crystal (in contrast with $CdWO_4$) can be explained by radioactive contamination of the calcium carbonate compound used for the crystal growing.

Nevertheless, it is remarkable that in the β(γ) spectrum of the $CaWO_4$ detector (Fig. 6) measured during 1734 h there are only 5 background counts in the energy region 3.6–5.4 MeV, that is, the background rate equals 0.07 counts/(yr kg keV). At the same time, in that energy interval of the initial spectrum (Fig. 3) there exists the broad distribution. It can be attributed to: (i) the β → α decays of ^{214}Bi → ^{214}Po → ^{208}Pb (^{238}U family)—these events were tagged by the time–amplitude and pulse-shape discrimination techniques as described above; (ii) the fast β → α sequence of ^{212}Bi ($Q_β$ = 2254 keV) → ^{212}Po ($E_α$ = 8784 keV, $T_{1/2}$ = 0.299 μs) → ^{208}Pb (^{232}Th family). Since two decays in the last (and fast) chain can not be time-resolved in the $CaWO_4$ scintillator (with decay time ≃8 μs), they will result in one event registered by the detector. However, because the shape indicator of such an event is different than those for pure α and β pulses, this component of the detector background was effectively rejected by the pulse-shape analysis. Five counts, remaining in the 3.6–5.4 MeV interval, most probably belong to β decay of ^{208}Tl.

Table 3
Radioactive contaminations in $CaWO_4$ and $CdWO_4$ scintillators

Chain	Nuclide	Activity (mBq/kg)	
		$CaWO_4$	$CdWO_4$
^{232}Th	^{232}Th	0.69(10)	0.053(5)
	^{228}Th	0.6(2)	0.039(2)[a]
^{235}U	^{227}Ac	1.6(3)	0.0014(9)
^{238}U	^{238}U	14.0(5)	⩽0.2
	^{226}Ra	5.6(5)	⩽0.004
	^{210}Pb		⩽0.4
	^{210}Po	291(5)	⩽0.4
	^{40}K	⩽12	0.3(1)
	^{90}Sr (^{90}Y)	⩽70	⩽0.2
	^{137}Cs	⩽0.8	0.43(6)[a]
	^{147}Sm	0.49(4)	⩽0.04

[a] In some $CdWO_4$ crystals only limits on corresponding activities were found: ^{228}Th—⩽0.003 mBq/kg, ^{137}Cs—⩽0.3 mBq/kg.

3. Discussion

3.1. $CaWO_4$ crystals for 2β decay search

Calcium tungstate crystals contain several potentially 2β decaying nuclides, including ^{48}Ca (see Table 4). As mentioned in Section 1, due to the highest $Q_{ββ}$ energy of ^{48}Ca this nuclide would be of utmost interest for the future 2β experiments. At the same time, $CaWO_4$ scintillator is a promising detector for 0ν2β decay search of ^{48}Ca because of its good energy resolution and excellent pulse-shape discrimination ability, which allow one to reduce background in the energy region of interest effectively.

Table 4
Double β decay candidates present in $CaWO_4$ crystals

Transition (decay channel)	Mass difference (keV) [29]	Isotopic abundance (%) [44]
^{40}Ca → ^{40}Ar (2ε)	193.62(0.21)	96.941(0.156)
^{46}Ca → ^{46}Ti (2β$^-$)	988.3(2.2)	0.004(0.003)
^{48}Ca → ^{48}Ti (2β$^-$)	4274(4)	0.187(0.021)
^{180}W → ^{180}Hf (2ε)	144(4)	0.12(0.01)
^{186}W → ^{186}Os (2β$^-$)	489.9(1.4)	28.43(0.19)

To prove this statement, let us estimate the half-life limit on the neutrinoless 2β decay of ^{48}Ca, which could be derived on the basis of our measurements with the $CaWO_4$ detector, whose background spectrum is depicted in Fig. 6. As it is described in Section 1, in the low background set-up the resolution of the $CaWO_4$ detector ($40 \times 34 \times 23$ mm) was measured with γ sources in the energy range 60–2615 keV. In particular, at 2.6 MeV (γ line of ^{208}Tl) the FWHM = 132 keV (5.1%). By extrapolating the measured energy dependence, we get the FWHM = 170 keV (4%) value at 4.27 MeV. Therefore, the 0ν2β decay of ^{48}Ca, if it occurred, should be seen as the sharp peak centred at the energy 4.27 MeV. To derive the half-life limit, we use the known formula: $\lim T_{1/2} = \ln 2 \cdot N \cdot \eta \cdot t / \lim S$, where $N = 7.4 \times 10^{20}$ is number of ^{48}Ca nuclei, $t = 1734$ h is the measuring time, $\eta = 0.87$ is the detection efficiency, and $\lim S$ is the number of events in the peak which can be excluded with a given confidence level on the basis of experimental spectrum. In the area of the 0ν2β peak there are only two counts, while the background expected in the same energy region was calculated as 2.9 counts (taking into account the intrinsic contamination of the $CaWO_4$ crystal and its simulated response function for ^{232}Th decay chain). Thus, in accordance with the Particle Data Group recommendations [47] we can accept $\lim S \approx 1.5$ counts at 68% C.L., which gives the half-life limit: $T^{0\nu}_{1/2} > 6 \times 10^{19}$ yr. Obviously, this pilot limit (obtained with the small *non-enriched* crystal) is modest as compared with those achieved in previous experiments ($\approx 10^{21}$–10^{22} yr), where enriched ^{48}Ca sources [48,49] or CaF_2(Eu) crystals of large volume (≈ 2300 cm^3) [12] were used.

On this basis, a CARVEL (CAlcium Research for VEry Low neutrino mass) project with ≈100 kg array of enriched $^{48}CaWO_4$ crystal scintillators is proposed. The detector array includes 50 modules, each of them consists of the cylindrical $^{48}CaWO_4$ crystal (∅7.5×7.5 cm), which is viewed from opposite sides by two ultra-low background 5″ PMT-s through two light-guides 10 cm in diameter and 70 cm long. Each light-guide is glued of two parts: high purity quartz (40 cm) and plastic scintillator (30 cm). The whole crystal array is surrounded by an active shield made of plastic scintillator, thus, together with active light-guides, a complete 4π active shield of main detectors is provided. The outer passive shield is made of copper (5 cm), lead (30 cm) and polyethylene (16 cm).

The sensitivity of the experiment depends on the source mass (≈16 kg of ^{48}Ca) and detector background in the 0ν2β decay window of ^{48}Ca. The possible sources of background are as follows: (i) radioactive contamination of $^{48}CaWO_4$ crystals by ^{228}Th and ^{226}Ra; (ii) cosmogenic activities in $^{48}CaWO_4$; (iii) external background; (iv) beta decay and two neutrino 2β decay of ^{48}Ca.

For our background calculation the contamination criterion for ^{228}Th has been accepted as ≈5 μBq/kg, whose achievement is a realistic task—we recall that the actual radiopurity of some samples of $CdWO_4$ crystals is even better (see Table 3). Besides, the ^{226}Ra activity in $CaWO_4$ crystals should be reduced to the level ≈20 μBq/kg. It will allow one to reject background from ^{208}Tl in the crystals by tagging α event from the preceding ^{212}Bi decay. The ^{228}Th activity of 100 kg detector corresponds to $\approx 1.6 \times 10^5$ decays during 10 yr measuring period. According to our Monte Carlo simulation, the calculated background contribution to the energy window 4.17–4.38 MeV from the ^{228}Th activity is ≈700 counts. However, applying the time–amplitude analysis and pulse-shape discrimination technique, this background rate can be reduced to ≈1.7 events for 10 yr exposure. It should be noted, that required radiopurity of the $^{48}CaWO_4$ detectors is less severe than that for the other high sensitivity 2β decay projects with ^{76}Ge [50–52], ^{100}Mo [53], ^{130}Te [54] and ^{136}Xe [55].

While the background associated with the secondary cosmic rays can be reduced to the necessary level by the proper deep underground site for a 2β decay experiment, the cosmogenic activities, produced in the $^{48}CaWO_4$ detectors during their stay on the Earth's surface, could be dangerous. For our case these activities were calculated with the program COSMO [56], assuming one month exposure period on the Earth's surface and a deactivation time of 1 yr underground. Fortunately, it was found that among a total number of 158 nuclides with $T_{1/2} > 25$ d produced in $^{48}CaWO_4$ crystals there are none, which can contribute to background in the energy window of the ^{48}Ca neutrinoless 2β decay. Apparently, it is due to the high $Q_{\beta\beta}$ energy of ^{48}Ca. We recall that, on the contrary, the cosmogenic activation of the detectors would be a serious problem for the future 2β projects with other nuclides, whose $Q_{\beta\beta}$ energies are smaller (^{76}Ge [50–52], ^{100}Mo [53], ^{116}Cd [57], ^{130}Te [54]). For example, cosmogenic activation of one ton array of HP Ge detectors enriched in ^{76}Ge to 86% were calculated in [50,51], where it was shown that remaining activities of ^{22}Na, ^{60}Co and ^{68}Ga (10–100 μBq) would create noticeable background near the energy 2039 keV ($Q_{\beta\beta}$ value of ^{76}Ge).

Similarly, the large $Q_{\beta\beta}$ value of ^{48}Ca makes requirements to the radiopurity of the materials for the detector mount and shield less stringent than those for the other experiments. For instance, in the project GENIUS [51] it is foreseen to shield HP ^{76}Ge semiconductor detectors with the help of ≈1000 t of liquid nitrogen, whose radiopurity must be as low as $\approx 10^{-15}$ g/g for ^{40}K, ^{238}U and ^{232}Th. The project CAMEO [57] intends to use ≈1000 t of super high purity water or liquid scintillator ($\approx 10^{-15}$ g/g for ^{238}U and ^{232}Th) as a shield for $^{116}CdWO_4$ crystals. Because of a low density (⩽1 g/cm^3) of these liquids (nitrogen, water, scintillator) the necessary dimensions of the shields are huge ($\approx \varnothing 12 \times 12$ m) [51,57]. On the contrary, in our ^{48}Ca project the "conventional" high purity (10^{-10}–10^{-12} g/g for ^{238}U and ^{232}Th) constructional and shielding materials (plastic, copper, steel, lead) can be used. It was proved by our calculations, that with such "conventional" passive and active shields, e.g., similar to those used for 2β study of ^{116}Cd with $^{116}CdWO_4$ scintillators [14], the background contributions from the radioactive impurities in the materials of the set-up, as well as from the surroundings (γ and neutron fluxes) would be negligible. For example, 10^{-10} g/g for ^{232}Th and ^{238}U contaminations of 4 t copper and 30 t lead shield correspond to rather high activities: ^{232}Th—14 Bq (Pb) and 1.6 Bq (Cu); ^{238}U—42 Bq (Pb) and 4.8 Bq (Cu). However, only events with simultaneous registration in the $CaWO_4$ detectors of two γ quanta are dangerous (there are no single γ rays with the energy more than 3.2 MeV in the ^{232}Th and ^{238}U chains). The probability of random coincidence of two events (from activities of ≈50 Bq) in the time window 20 μs is $\approx 5 \times 10^{-2}$. This number should be multiplied on probability that both γ quanta will be not absorbed in Cu and Pb shield, then, the efficiency of registration by $CaWO_4$ detectors must be taken into account too. In accordance with our calculation, the number of events detected during 10 yr from 2.6 MeV γ rays originating in Cu shield is $\approx 10^{-3}$, that is completely negligible (it is even less for the Pb shield located farther of detectors).

One should note also that dense shielding materials (copper, steel, lead) reduce dimensions of the set-up substantially and, hence, would make realization of the project much simpler and less expensive. Besides, the problem of the omnipresent ^{222}Rn gas (which is the "nightmare" of any super-low background experiments in the field of solar neutrinos detection, 2β decay and dark matter searches, etc. [58]) can be also neglected because of very high $Q_{\beta\beta}$ value of ^{48}Ca. It is because among all daughters of ^{222}Rn, beta active ^{214}Bi has the largest Q_β value (3.27 MeV). In principle, a fast sequence of β decay of ^{214}Bi and α decay of ^{214}Po could produce background events up to 5.5 MeV in $CaWO_4$ scintillator. However, such a background would be strongly suppressed with the help of the pulse-shape and time–amplitude analysis.

The irreducible background could be also caused by the β decay of ^{48}Ca, which is energetically allowed ($Q_\beta = 278$ keV), followed by the β decay of ^{48}Sc ($Q_\beta = 3994$ keV, $T_{1/2} \approx 44$ h) [45]. However, because of a small available energy and large change in spin, the β decay of ^{48}Ca is strongly suppressed (up-to-date only half-life limit of the order of 10^{20} yr was set experimentally for this transition [59]). Due to this, and taking into account the time and energy distributions of both (^{48}Ca and ^{48}Sc) β spectra, the contribution of the sequence of two β decays ($^{48}\text{Ca} \to {}^{48}\text{Sc} \to {}^{48}\text{Ti}$) to background in the energy window of the 0ν2β decay of ^{48}Ca is negligible.

Finally, let us consider the irreducible background, which could be produced by the 2ν2β decay events with the energies close to the $Q_{\beta\beta}$ value of ^{48}Ca. Because the better the energy resolution of the detector, the lower is the contribution of such events, the energy resolution is crucial for the success of the proposed experiment. Thus, we have to justify the feasibility to achieve the proper energy resolution with the $CaWO_4$ crystal scintillator.

The relative energy resolution of an ideal scintillation detector can be expressed by the formula [42]: $\mathrm{FWHM} = 2.36/\sqrt{N_{pe}}$, where N_{pe} is the mean number of photoelectrons produced by PMT photocathode. The latter can be written as a product: $N_{pe} = N_{ph} \times E \times \mathrm{LC} \times \mathrm{QE}$, where N_{ph} is a mean number of photons created in a scintillator per 1 MeV of the energy deposit E, LC is the part of photons arrived to the photocathode of the PMT, and QE is quantum efficiency of the photocathode to photons emitted by the scintillator. Because the value of the photon yield (N_{ph}) of the $CaWO_4$ scintillator is not available in the literature, we have estimated it in the relative measurements with two crystals: $CaWO_4$ ($\varnothing 40 \times 39$ mm) and NaI(Tl) $\varnothing 40 \times 40$ mm. The light yield of the NaI(Tl) is $(3.8–4.3) \times 10^4$ photons per 1 MeV [60]. Both crystals were coupled to the same PMT XP2412 with the bialkali photocathode, hence, the QE values were calculated as the convolution of the emission spectrum of the corresponding scintillator (taken for $CaWO_4$ from [61]) and spectral sensitivity of the XP2412 photocathode: QE(NaI(Tl)) = 0.24; QE($CaWO_4$) = 0.22. The relative (to NaI(Tl)) pulse amplitude of the $CaWO_4$ scintillator for γ lines of ^{137}Cs (661 keV) and ^{207}Bi (570 and 1064 keV) has been measured as 0.18. The light collection efficiencies for the NaI(Tl) and $CaWO_4$ detectors were calculated with the help of GEANT4 code [62] as 65% and 52%, respectively. On this basis we can estimate photon yield of $CaWO_4$ scintillators as $(0.8–1.2) \times 10^4$ photons per 1 MeV, that is $\approx 4 \times 10^4$ photons for 4.27 MeV. Therefore, with the slightly increased to 65–70% light collection the 0ν2β decay peak of ^{48}Ca could be measured with the energy resolution of FWHM ≈ 2.8%.

This optimistic theoretical estimate has been proved in the measurements. Because the high refractive index of the $CaWO_4$ crystal is the main cause which reduces the total collection of a scintillation light, we have tried to improve it by placing the $CaWO_4$ crystal in liquid (with the $CdWO_4$ crystal a ≈40% increase of the light collection was obtained [57]). With this aim, a cylindrical $CaWO_4$ crystal ($\varnothing 40 \times 39$ mm) was fixed in the centre of a teflon container with inner diameter 70 mm. The latter was coupled on opposite sides with two PMTs Philips XP2412, so that the distance from each flat surface of the crystal to the corresponding PMT's photocathode was 25 mm. The container was filled up with the pure and transparent silicon oil (refractive index ≈1.5). The results of measurements with ^{60}Co, ^{137}Cs and ^{232}Th γ sources are depicted in Fig. 1, where spectra obtained with the standard detector arrangement ($CaWO_4$ crystal wrapped by teflon reflector and directly coupled to the PMT photocathode with optical glue) are also shown. By comparing these spectra, one can find that ≈20% increase of light collection was reached with the $CaWO_4$ crystal placed in the liquid. It resulted in improvement of the detector energy resolution in the whole energy region—for example, at 2.6 MeV the measured FWHM resolution is equal to 3.8% (see Fig. 1c). A fit of the data measured with various γ sources gives FWHM ≈2.9% at 4.27 MeV. On this basis we assume for the CARVEL project a modest improvement of the energy resolution of $CaWO_4$ scintillators at $Q_{\beta\beta}$ energy up to FWHM ≈ 2.5%. Such an advancement of the energy resolution would be achieved as a result of further purification of the crystals, which will improve their optical transmission (our Monte Carlo calculations show that increase of the $CaWO_4$ absorption length from current ≈30 cm to 70–90 cm would enlarge a total light collection to 75–80%).

The response functions of the set-up with 100 kg $^{48}CaWO_4$ crystals (and 10 yr measuring period) for 2ν2β decay of ^{48}Ca ($T^{2\nu}_{1/2} = 4 \times 10^{19}$ yr) and 0ν2β decay with $T^{0\nu}_{1/2} = 10^{26}$ yr are shown in Fig. 7. One can see that due to a good energy resolution the contribution of the 2ν tail to the energy window of the 0ν peak is rather small. On the other hand, it is obvious from Fig. 7 that neutrinoless 2β decay of ^{48}Ca with half-life of $\approx 10^{26}$ yr would be clearly distinguished from the 2ν2β decay distribution.

Taking into account number of ^{48}Ca nuclei ($\approx 2 \times 10^{26}$) and measuring time of about 10 yr, we can estimate sensitivity of the experiment in

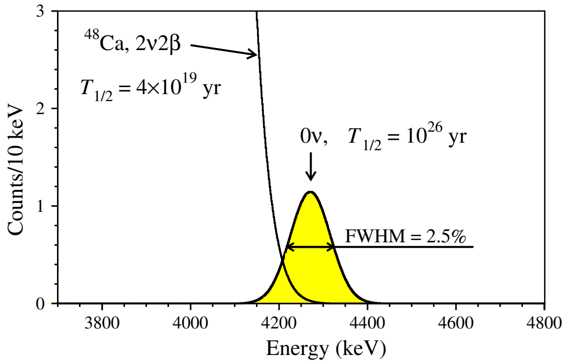

Fig. 7. The response functions of the set-up with 100 kg $^{48}CaWO_4$ crystals (10 yr measuring time) for 2β decay of ^{48}Ca: $T^{2\nu}_{1/2} = 4 \times 10^{19}$ yr and $T^{0\nu}_{1/2} = 10^{26}$ yr.

terms of the half-life limit for 0ν2β decay of ^{48}Ca as $\approx 10^{27}$ yr. The latter translates to the constraints on the Majorana neutrino mass in the range (0.04–0.09) eV, depending on the nuclear matrix elements calculations used (the following $T^{0\nu}_{1/2} \cdot \langle m_\nu \rangle^2$ values (in yr eV^2) were obtained with the shell model calculations: 3.2×10^{24} [63], 1.7×10^{25} [64], 5.3×10^{24} [65], $(6.4–7.8) \times 10^{24}$ [66], 8.8×10^{24} [67], while QRPA resulted in: 1.5×10^{24} [64], 7.1×10^{23} [65], $(1.1–3.3) \times 10^{24}$ [68], 2.5×10^{24}–2.8×10^{25} [69], 2.3×10^{24} [70]). The proposed technique with $^{48}CaWO_4$ crystal scintillators is simple and reliable, thus, such an experiment can run stably for decades.

Obviously, to get such an impressive result, which would be of great interest for modern physics, the problems of a large mass production and purification of enriched ^{48}Ca should be solved, which is actively considered now. For example, a new project MCIRI [71] for the large mass production of ^{48}Ca and some other isotopes by the method of ion cyclotron resonance heating in plasma is under development by the Kurchatov Institute (Moscow, Russia) and JINR (Dubna). Besides, the AVLIS (Atomic Vapor Laser Isotope Separation) technique is planned to be used for the same purpose in the USA.

There exists another ^{48}Ca project CANDLES [72] which intends to use non-enriched CaF_2 crystal scintillators as active source. The crystals would be immersed in the liquid scintillator, which serves as an active and passive shielding against radiation. Because CaF_2 crystals emit UV light, which can not be registered with the help of usual PMT-s, it is supposed to put each crystal in the transparent container filled with the liquid wavelength shifter. Then the usual light from these containers, propagating in the liquid scintillator, would be collected by the large number of distant PMT-s. The CANDLES sensitivity estimates for the two final stages are as follows: $m_\nu \leqslant 0.5$ eV (with 200 kg of crystals) and 0.15 eV (with 3.2 t of crystals).

It should be mentioned encouraging results obtained with CaF_2 crystal with Eu doping as a scintillating bolometer [73]. This allows separate effectively the electrons emitted in double beta decay from α particles. In addition, thermal detectors provide excellent energy resolution. A disadvantage of these detectors often is instability of operation during long time measurements, which is important for double beta decay experiments.

3.2. Dark matter search

As it was mentioned in Section 1, the calcium tungstate crystals are promising low temperature detectors for the dark matter particles quest. It is because these crystals allow one the simultaneous registration of the heat and light signals, which, as it was already demonstrated, is a powerful tool for background rejection [33,34]. Therefore, the $CaWO_4$ detectors could provide an interesting possibility to search for spin-dependent inelastic scattering of WIMPs with excitation of ^{183}W (spin 1/2$^-$) low-energy nuclear level (46.5 keV). Identification of such "mixed" (nuclear recoil plus γ quanta) events can be possible due to registration of heat and light signals, which ratio is different from those of "pure" nuclear recoil or γ(β) events. The natural isotopic abundance of ^{183}W is 14.3%, thus non-enriched $CaWO_4$ crystals can be utilized.

However, the radioactive contamination of the $CaWO_4$ detectors available now (e.g., studied in the present work and in Refs. [33,34]) must be substantially improved to make them competitive for the dark matter and 2β decay searches. Aiming to obtain $CaWO_4$ crystals less contaminated, we are going to grow them from raw materials with the preliminary check of their radioactive contamination. The method of low-background measurements (developed in the present work on the basis of the pulse-shape and time–amplitude analysis) provides simple and rather fast way to control radioactive contamination of $CaWO_4$ crystals. The sensitivities are: (i) some tens of μBq/kg for ^{228}Th, ^{226}Ra, and ^{227}Ac; (ii) few hundred of μBq/kg for ^{147}Sm and long-living α active daughters of ^{232}Th and ^{238}U; (iii) a few mBq/kg for ^{40}K, ^{137}Cs, ^{90}Sr–^{90}Y and beta active U–Th daughters.

Recently the $CaWO_4$ crystal (∅6 × 12 cm) with mass of 2 kg has been grown from the purified materials with the reduced radioactive contamination. The preliminary background measurement performed on the Earth surface gave us an encouraging result: the total internal alpha activity of 2 kg $CaWO_4$ crystal is ≈20 mBq/kg, that is ≈20 times lower than that reported above.

4. Conclusions

Scintillation properties of $CaWO_4$ crystal scintillators were studied. The energy resolution of $CaWO_4$ detectors is similar to that of NaI(Tl) scintillators (e.g., FWHM ≈ 7% for the 662 keV γ line of ^{137}Cs, and FWHM = 3.8% for the 2615 keV γ line of ^{208}Tl). Due to the difference of $CaWO_4$ scintillation pulse shapes for α particles and γ quanta (β particles), clear discrimination between them was achieved. Radioactive contaminations of the available $CaWO_4$ crystals are higher by factors from 10 to 10^3 (e.g., activity of ^{210}Po is ≈0.3 Bq/kg) than those of the $CdWO_4$ scintillators. Recently the $CaWO_4$ crystal with mass of 2 kg has been grown from the radiopure materials. As a result its total internal alpha activity was reduced to ≈20 mBq/kg, that is by factor ≈20.

By applying the developed methods of the pulse-shape discrimination and time–amplitude analysis of data, the background of the ≈0.19 kg $CaWO_4$ detector in the energy region 3.6–5.4 MeV has been reduced down to 0.07 counts/(yr keV kg). It is one of the lowest background rates which has been reached in 2β decay experiments. Due to this the lower half-life limit on 0ν2β decay of ^{48}Ca has been set as $T_{1/2}^{0\nu} > 6 \times 10^{19}$ yr at 68% C.L. by using a *non-enriched* and rather small $CaWO_4$ crystal. It demonstrates good abilities of $CaWO_4$ detectors for 2β decay study of ^{48}Ca, whose highest $Q_{\beta\beta}$ energy is the great advantage of this 2β candidate as compared with other nuclides.

On this basis, a CARVEL (CAlcium Research for VEry Low neutrino mass) project of the experiment with ≈100 kg array of enriched $^{48}CaWO_4$ crystal scintillators (≈16 kg of ^{48}Ca) has been proposed. The sensitivity of such an experiment (in terms of the half-life limit) is estimated as $T_{1/2}^{0\nu} > 10^{27}$ yr, which corresponds to the neutrino mass constraint $m_\nu < (0.04–0.09)$ eV.

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