

N. D. Khanbekov¹, V. V. Alenkov², A. A. Burenkov¹, O. A. Buzanov², V. N. Kornoukhov¹

¹Institute for Theoretical and Experimental Physics, Moscow, Russia

²JSC Fomos-Materials, Moscow, Russia

DATA ANALYSIS OF THE INTERNAL BACKGROUND MEASUREMENTS OF ⁴⁰Ca¹⁰⁰MoO₄ SCINTILLATION CRYSTALS

The sensitivity of neutrinoless double beta (0ν2β) decay experiments is mainly dependent on the internal background of a detector which, in its turn, is defined by the purity of material and possibility for selection of background events. The AMoRE (Advanced Mo based Rare process Experiment) collaboration plans to use ⁴⁰Ca¹⁰⁰MoO₄ scintillation crystals as a detector for search of 0ν2β decay of ¹⁰⁰Mo isotope. A purpose of this paper is further investigation of internal background of ⁴⁰Ca¹⁰⁰MoO₄ scintillation elements with a low background setup at YangYang underground laboratory. We present new approaches for selection of background events from analyzing data and the latest updated values of background index of ⁴⁰Ca¹⁰⁰MoO₄ crystals as a result of the new technique application.

Keywords: neutrinoless double beta decay, data analysis, scintillators, calcium molybdate, low-background physics, time-amplitude analysis, radioactive background.

Introduction

Discovery of the neutrino oscillation means that neutrinos have non-zero mass [1]. It's become a new impetus for further searches of physics beyond the Standard Model. Detection of 0ν2β decay would give an opportunity to determine the effective mass of neutrino $\langle m_\nu \rangle$ and would confirm that neutrino is a Majorana particle, i.e. neutrino is identical to anti-neutrino.

One part of the Heidelberg-Moscow collaboration claimed to observe 0ν2β decay of ⁷⁶Ge isotope [2]. Nowadays the GERDA experiment sets a goal to confirm or disprove this result [3]. Several groups are carrying on experiments with other isotopes. EXO-200 [4] and KamLand-Zen [5] experiments search for the 0ν2β decay of ¹³⁶Xe isotope.

Next generation 0ν2β experiments need the high energy resolution, low background of a detector and many tens or hundreds kilograms of the working isotope with the high transition energy $Q_{\beta\beta}$. ¹⁰⁰Mo isotope is one of the best candidates for 0ν2β experiments because of one of the highest values of $Q_{\beta\beta} = 3034$ keV and possibility for production of this isotope in a big amount by centrifugation method [6, 7].

Appropriate detector for the experiment with ¹⁰⁰Mo isotope which was chosen by the AMoRE collaboration [8] is a scintillation bolometer based on ⁴⁰Ca¹⁰⁰MoO₄ single crystals. The fractional mass of the molybdenum element in the crystal is relatively high (about 50 %). The maximum energy of γ -background from natural long-lived isotopes is 2615 keV (²⁰⁸Tl from ²³²Th-chain). Thus, on the condition that the resolution of a detector is good, external γ -background doesn't contribute to the ROI (the Range of Interest) for ¹⁰⁰Mo.

On the contrary the two-neutrino double beta decay of ⁴⁸Ca ($Q_{\beta\beta} = 4271$ keV) will give unavoidable

background, limiting the sensitivity of the experiment with enriched ⁴⁰Ca¹⁰⁰MoO₄. Since natural calcium contains 0.187 % of ⁴⁸Ca we need to use calcium depleted in ⁴⁸Ca isotope [6]. Another problem is the internal background due to the presence of dangerous radionuclides inside the detector. There are two isotopes with decay energy higher than 3034 keV: ²⁰⁸Tl ($Q_{\beta} = 5001$ keV) from ²³²Th-chain and ²¹⁴Bi ($Q_{\beta} = 3272$ keV) from ²³⁸U-chain. Two supplementary approaches are used to reduce this background: purification of material to remove radioactive impurities and selection of background events by sophisticated data analysis.

Previously the AMoRE collaboration, in order to estimate the radioactive contamination, used a time-amplitude analysis [9], which exploits the energies and time difference between primary and secondary signals to select specific fast sequences of decays in the U/Th chains. This study reports a new technique (and its application) of data analysis of scintillation signals, which allows us to solve a problem of pileup events from the ²¹²Bi-²¹²Po and ²¹⁴Bi-²¹⁴Po decays [9, 10] and improve a background index of S35 and SB29 crystals.

Production of ⁴⁰Ca¹⁰⁰MoO₄ crystals and scintillation elements

Enriched molybdenum (with 96.1 % of enrichment by ¹⁰⁰Mo isotope) is produced by the JSC Production Association Electrochemical plant (Zelenogorsk, Krasnoyarsk region, Russia [11]) and supplied in the form of molybdenum oxide ¹⁰⁰MoO₃. The results of ICP-MS measurements show that the enriched material is very pure. Concentrations of ²³⁸U and ²³²Th in the oxide do not exceed 0.07 and 0.1 ppb, respectively.

Calcium carbonate $^{40}\text{CaCO}_3$ enriched in ^{40}Ca (99.964 %) and depleted in ^{48}Ca (content is ≤ 0.001 %) is produced by the FSUE Electrochim-pribor (Lesnoy, Sverdlovsk region, Russia [12]). The concentration of ^{238}U and ^{232}Th in the enriched powder measured by ICP-MS is below 0.2 and 0.8 ppb, respectively. However, HPGe measurements at the Baksan Neutrino Observatory showed that the activity of ^{226}Ra and its daughter isotopes in the decay chain was on a level of hundreds mBq/kg [13] (which corresponds to concentration equal ~ 20 ppb). For that reason the raw materials were subjected to additional purification. A new technique of purification of calcium carbonate in the form of calcium formate $\text{Ca}(\text{HCOO})_2$ [14] allowed to reduce a content of ^{40}K , ^{208}Tl , ^{228}Ac , ^{226}Ra (^{214}Bi) in 20, 8, 160 and 5 times respectively in comparison with the standard procedure of purification on Electrochim-pribor plant.

$^{40}\text{Ca}^{100}\text{MoO}_4$ crystals were grown by Czochralski technique by JSC Fomos-Materials [15]. The specific process, which is named recrystallization, consisted of two steps. On the first step two single crystals (boules) were pulled out of mixture in platinum crucible. Then, on the second step, the grown boules were used as a raw material for pulling of a final single crystal. The recrystallization process allows to reduce radioactive contamination in the crystal [14]. Thus ^{226}Ra content was decreased from 260 mBq/kg in initial calcium carbonate $^{40}\text{CaCO}_3$ to 6.4 mBq/kg in a $^{40}\text{Ca}^{100}\text{MoO}_4$ crystal (the reduction factor of 40). Crystals were bluish just after growing. To decolorize crystals they were annealed in oxidative (oxygen) atmosphere [6, 14].

Conic parts of crystals were cut and then their ends were optically polished to produce scintillation elements. Thus, the studied scintillation elements have cylindrical shape with elliptic base, unpolished side surface and optical polished butt-ends. In the future $^{40}\text{Ca}^{100}\text{MoO}_4$ single crystals, produced by this

technique from more pure raw materials, will be used for production of scintillation elements of cryogenic scintillation detector of AMoRE Collaboration.

Two $^{40}\text{Ca}^{100}\text{MoO}_4$ scintillation elements were used for our measurements: SB29 and S35. The SB29 crystal was produced with recrystallization process, but S35 was grown directly from $^{40}\text{Ca}^{100}\text{MoO}_4$ charge (raw material). Dimensions and masses of the crystals are presented in the Table.

Properties of $^{40}\text{Ca}^{100}\text{MoO}_4$ scintillation elements

	SB29	S35
Height, mm	48/51	42
D big, mm	50	44
D small, mm	42.5	39.5
Mass, g	390	259
Annealing, h	240	48
Color after annealing	bluish	colorless

Low-background experimental setup

Background measurements of $^{40}\text{Ca}^{100}\text{MoO}_4$ scintillation elements (below in the text we will use word “crystals”) were carried out in the YangYang underground laboratory (Y2L) on depth of 700 m (approximately 2000 m of water equivalent). Information about laboratory radioactive background was given in [16, 17]. In order to decrease external background the 4π gamma veto system was used [18, 19]. The system consisted of 14 CsI(Tl) crystals (12 long crystals for sides and 2 short crystals for front-end caps) which were able to register the internal and external gammas. The veto-system rejected the coincidence events from $^{40}\text{Ca}^{100}\text{MoO}_4$ and one or several CsI(Tl) crystals. The system was shielded by lead (10 cm thick, Fig. 1). During the measurements, the whole internal cavity of system was continuously flushed by nitrogen (at rate of 4 l/min) to prevent the contamination by ^{222}Rn present in air.

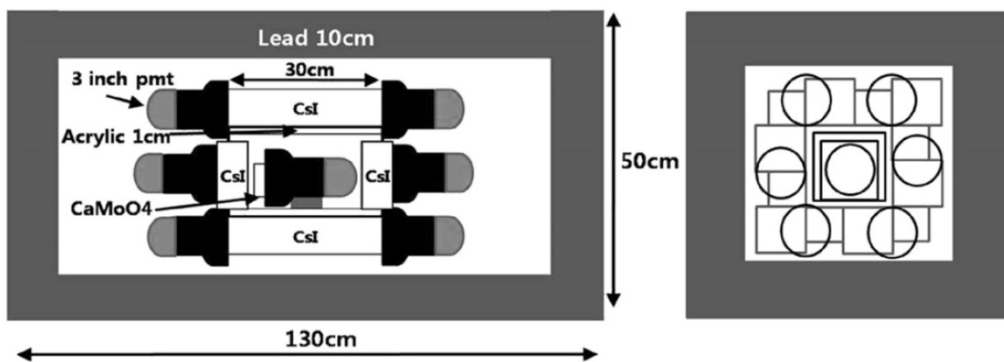


Fig. 1. Scheme of the setup for background measurements of $^{40}\text{Ca}^{100}\text{MoO}_4$ crystals [20].

The scintillation light from $^{40}\text{Ca}^{100}\text{MoO}_4$ crystals was collected by 7.62 cm diameter green enhanced

PMT (Electron tube Ltd.). Crystals were wrapped by Teflon tape and connected to PMT using optical

grease. Signals from $^{40}\text{Ca}^{100}\text{MoO}_4$ and from CsI(Tl) veto-system were amplified 10 times and digitized by 400 MHz FADC and 64 MHz FADC respectively. The recorded time-window from $^{40}\text{Ca}^{100}\text{MoO}_4$ was 82 μs (the decay constant of CaMoO_4 is 16.4 μs). Veto-system data were recorded simultaneously with $^{40}\text{Ca}^{100}\text{MoO}_4$ signals [9].

Selection procedure for SB29 and S35 scintillation elements

We used background data which were collected during 50 days for S35 crystal and 90 days for SB29 crystal. First of all we worked out ^{214}Bi and ^{208}Tl (^{220}Rn) problem by time-amplitude analysis [6, 10]. In order to select corresponding events from raw background data, we used the following decay chains: ^{214}Bi ($Q_\beta = 3.27$ MeV, $T_{1/2} = 19.9$ min) \rightarrow ^{214}Po ($Q_\alpha = 7.83$ MeV, $T_{1/2} = 164$ μs) \rightarrow ^{210}Pb and ^{220}Rn ($Q_\alpha = 6.41$ MeV, $T_{1/2} = 55.6$ s) \rightarrow ^{216}Po ($Q_\alpha = 6.91$ MeV, $T_{1/2} = 145$ ms) \rightarrow ^{212}Pb . To find ^{214}Bi events we checked signals in the time window after each event which equals to $5 \cdot T_{1/2} \approx 0.8$ ms (there $T_{1/2}$ stands for the half life of ^{214}Po isotope). If the event with the energy release which corresponds to ^{214}Po in the electron equivalent energy is found we conclude that previous event was from ^{214}Bi decay. Thus we are able to find up to 85 % of ^{214}Bi decays [6]. Last results of measurement of ^{214}Bi (1.74 mBq/kg) and ^{208}Tl (0.26 mBq/kg) content for S35 crystal were reported in [9]. However at that time we were not able to count pileup signals from two consecutive events in the same 82 μs window.

Two approaches were used to solve this problem. The first one, which is also named ‘‘a tail to total method’’ [21], is based on possibility to select normal events by waveform analysis. If we make partial charge integration within a small time window inside the signal (it is better to take a region near the end of the signal), for signals without overlapping it must be a roughly linear dependence of the integrated energy on the range of integration. But pileup signals fall out of this dependence. This fact gives us an instrument for selection of pileup events beyond linear dependence (Fig. 2).

Using this method we identified as pileups 60.5 % of background events in the ROI for $0\nu 2\beta$ -decay ($2935\text{-keV} < E < 3135\text{-keV}$, taking into account the energy resolution of S35 crystal).

In the following we will discuss the second approach for identification of pileup events.

As it was already shown scintillation light from $^{40}\text{Ca}^{100}\text{MoO}_4$ crystals consists of several light-emission components with different decay constants. Shapes of these components and values of decay constants are different for α -events and $\gamma(\beta)$ -events

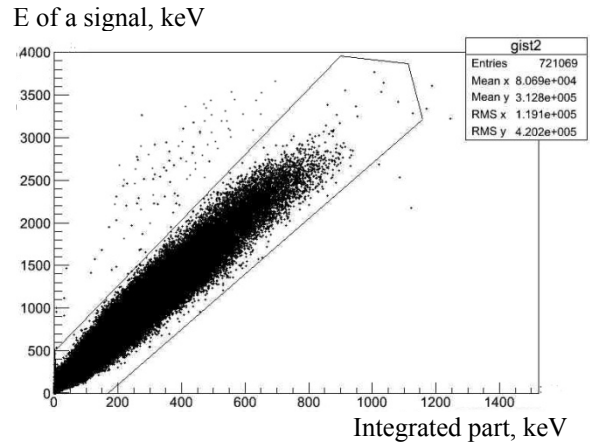


Fig. 2. Plot of energy of a signal versus energy of the selected part inside the signal for SB29 crystal.

[6, 22]. Thus an average decay time can be used for selection of pileup events in the data.

The average decay time of scintillation signal was calculated as

$$\langle t \rangle = \sum E_n \cdot n / E,$$

where E_n is the energy in one bin recorded by FADC in the signal histogram, n is a number of bins from the beginning of the signal and E is the total energy of the signal. Dependence of the total energy versus average decay time for S35 crystal is shown in Fig. 3. Colour gradient is determined by the density of points in the plot region (by ROOT analysis). Elliptical circles indicate the events from α -decay of ^{210}Po and ^{214}Po isotopes. As we can see, centers of these concentrations are shifted to the left from the center of a main distribution (which consists of gamma events).

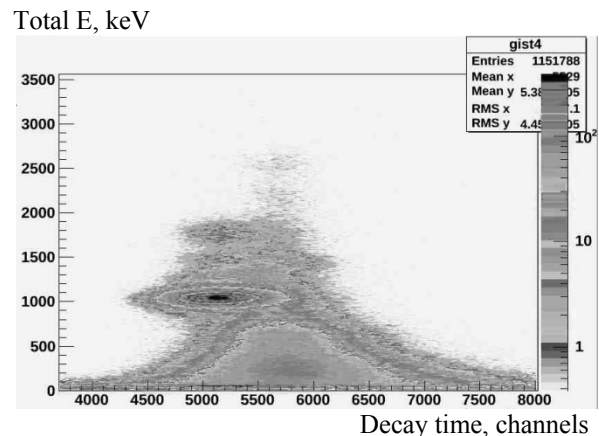


Fig. 3. Dependence of the total energy versus average decay time (in channels) for S35 crystal. Colour gradient is determined by the point density in the given region of the plot.

Pileup signals must have longer decay time and are located more to the right from the mean $\beta(\gamma)$ -time (Fig. 4). It’s worth to notice that most of

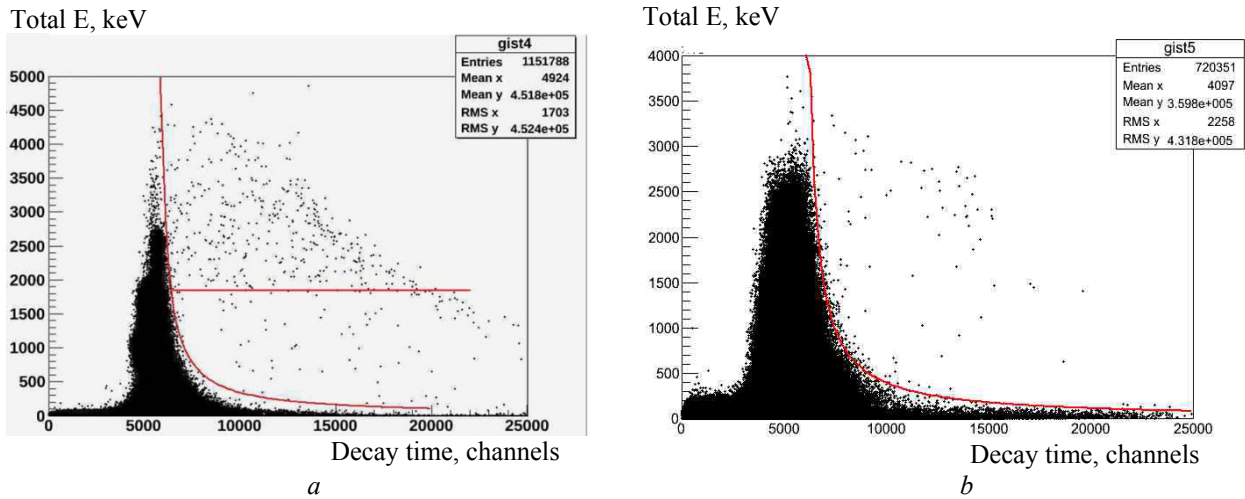


Fig. 4. Dependence of the total energy versus the average decay time for S35 (a) and SB29 (b) crystals with hyperbolic fitting of the edge of the distribution.

events on the right part of the picture are above the level of energy of ^{214}Po events (1.93 MeV in γ -scale, straight line in Fig. 4, a). It also indicates that the found events are pileups. Using standard ROOT function we fitted the edge of the distribution on the plot by hyperbola in order to select pileup events.

Using this method we selected 64.2 % events from the ROI of S35 crystal. Due to small internal background of SB29 there was no difference for results of the both approaches for this crystal.

The background index of SB29 scintillation element

The measured FWHM energy resolution of SB29 is 26.5 % with 662 keV γ -rays (^{137}Cs source) at room temperature. An energy spectrum is presented in Fig. 5.

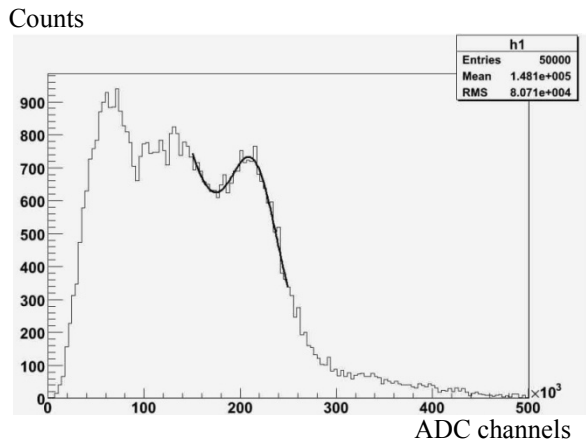


Fig. 5. Energy spectrum (^{137}Cs source) measured with SB29 crystal.

The background spectrum of SB29 crystal is presented in Fig. 6. An analysis of such background spectrum for CaMoO_4 can be found in [6].

In spite of worse energy resolution SB29 crystal is an example of $^{40}\text{Ca}^{100}\text{MoO}_4$ crystal with high puri-

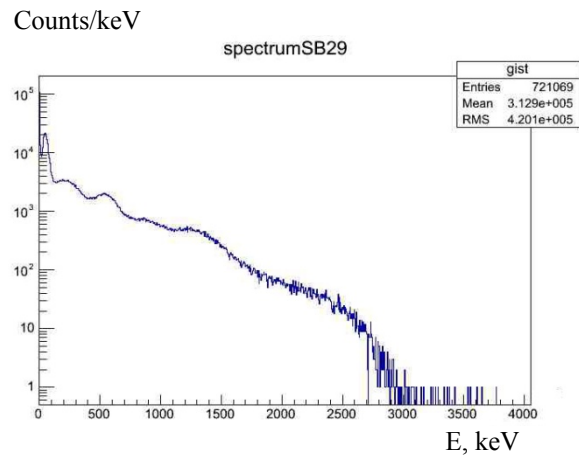


Fig. 6. Background spectrum of the SB29 crystal after 90 days of measurements.

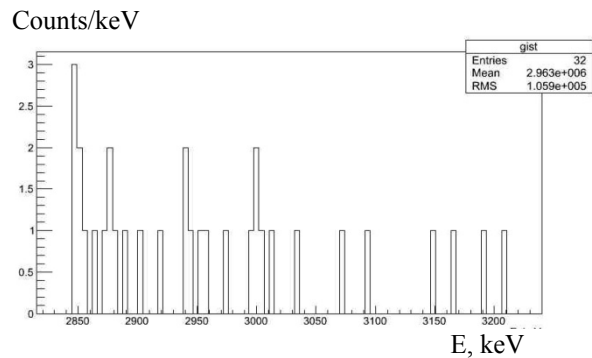


Fig. 7. Background events of SB29 crystal in the range of interest (2847 keV < E < 3223 keV) after 90 days measurements.

ty and, as a consequence, low internal background. After 90 days of measurements we registered only 32 events in the ROI (2847 keV < E < 3223 keV) and 6 events among them were identified as pileups events by methods we described in previous section (Fig. 7). Thus, the background index was defined as $0.7 \text{ counts keV}^{-1} \text{ kg}^{-1} \text{ year}^{-1}$, which gives the sensitivity level for $0\nu 2\beta$ -decay of ^{100}Mo equal to

$1.8 \cdot 10^{22}$ years at confidence level of 90 % with time of measurements 90 days using one SB29 crystal.

Conclusions

$^{40}\text{Ca}^{100}\text{MoO}_4$ scintillation elements based on enriched ^{100}Mo and Ca depleted in ^{48}Ca were produced at the first time.

Internal background of scintillation elements was studied. Waveform analysis or checking average decay time of a signal can be used to solving a problem of pileup events.

In case of S35 crystal a number of background events in the energy range of interest was decreased by 64.2 %. The S35 background index is 3.3 counts $\text{keV}^{-1} \text{kg}^{-1} \text{year}^{-1}$. It gives a new value of sensitivity of $0\nu 2\beta$ -decay experiment at level of $6.8 \cdot 10^{21}$ years in comparison with $4.0 \cdot 10^{21}$ years without excluding pileups [9].

The analysis of background events of SB29 crystal was done. Application of the method which was proposed in this paper allowed to select 6 pileup events of total 32 in the ROI ($2847 \text{ keV} < E < 3223 \text{ keV}$). Thus, the background index of SB29 is 0.7 counts $\text{keV}^{-1} \text{kg}^{-1} \text{year}^{-1}$.

For a cryogenic experiment with 5 kg of $^{40}\text{Ca}^{100}\text{MoO}_4$ scintillation elements and 5 years of measurements the limit for $0\nu 2\beta$ -decay half-life is $2.9 \cdot 10^{23}$ years with 90 % confidence level at the condition that the background index of the detector will be identical to the one for SB29 crystal.

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REFERENCES

1. *Araki T. et al.* Measurement of Neutrino Oscillation with KamLAND: Evidence of Spectral Distortion // *Phys. Rev. Lett.* - 2005. - Vol. 94. - P. 081801.
2. *Klapdor-Kleingrothaus H.V., Dietz A., Harney H.L., Krivosheina I.V.* Evidence for neutrinoless double beta decay // *Modern Physics Letters.* - 2001. - Vol. A16 (37). - P. 2409 - 2420.
3. *Schonert S., Abt I., Altmann M. et al.* The GERmanium Detector Array (Gerda) for the search of neutrinoless $\beta\beta$ decays of ^{76}Ge at LNGS // *Progress in Particle and Nuclear Physics.* - 2006. - Vol. 57(1). - P. 241 - 250.
4. *Ackerman N. et al.* Observation on Two-Neutrino Double-Beta Decay in ^{136}Xe with the EXO-200 Detector // *Phys. Rev. Lett.* - 2011. - Vol. 107. - P. 212501.
5. *Gando A. et al.* Limits on Majoron-emitting double-beta decays of Xe-136 in the KamLAND-Zen experiment // *Phys. Rev.* - 2012. - Vol. C86. - P. 021601.
6. *Annenkov A.N. et al.* Development of CaMoO_4 crystal scintillators for a double beta decay experiment with ^{100}Mo // *Nucl. Instrum. Methods.* - 2008. - Vol. A584. - P. 334 - 346.
7. *Belogurov S.G. et al.* CaMoO_4 scintillation crystal for the search of ^{100}Mo double beta decay // *IEEE Trans. Nucl. Sci.* - 2005. - Vol. 52. - P. 1131 - 1135.
8. *Bhang H. et al.* AMoRE experiment: a search for neutrinoless double beta decay of ^{100}Mo isotope with $^{40}\text{Ca}^{100}\text{MoO}_4$ cryogenic scintillation detector // *J. Phys.: Conf. Ser.* - 2012. - Vol. 375. - P. 042023.
9. *So J.H. et al.* Scintillation Properties and Internal Background Study of $^{40}\text{Ca}^{100}\text{MoO}_4$ Crystal Scintillators for Neutrino-Less Double Beta Decay Search // *IEEE Trans. Nucl. Sci.* - 2012. - Vol. 59. - P. 2214 - 2218.
10. *Danevich F.A. et al.* Search for 2β decay of cadmium and tungsten isotopes: Final results of the Solotvina experiment // *Phys. Rev.* - 2003. - Vol. C68. - P. 035501.
11. *Electrochemical plant* <http://www.ecp.ru>
12. *FSUE Electrochimpribor* <http://www.ehp-atom.ru/>
13. *Barabanov I.R., Buzanov O.A., Veresnikova A.V. et al.* Content of radioactive isotopes in the original materials and ready-made scintillation CaMoO_4 crystal according to underground low-background semiconductor spectrometer data, Preprint INR of RAS, № 1237/2009 Moscow (in Russian).
14. *Alenkov V. et al.* Growth and characterization of isotopically enriched $^{40}\text{Ca}^{100}\text{MoO}_4$ single crystals for rare event search experiments // *Cryst. Res. Technol.* - 2011. - Vol. 46. - P. 1223 - 1228.
15. *JSC Fomos-Materials* <http://newpiezo.com/index.html>
16. *Lee H.S. et al.* Limits on interactions between weakly interacting massive particles and nucleons obtained with CsI(Tl) crystal detectors // *Phys. Rev. Lett.* - 2007. - Vol. 99. - P. 091301.
17. *Kim H.J. et al.* Neutrino-Less Double Beta Decay Experiment Using $^{40}\text{Ca}^{100}\text{MoO}_4$ Scintillation Crystals // *IEEE Trans. Nucl. Sci.* - 2010. - Vol. 57. - P. 1457 - 1480.
18. *So J.H. et al.* 4π CsI(Tl) detector for EC/ β^+ decay searching in ^{84}Sr // *NSS-MIC Conf. Record.* - 2008. - Vol. 55. - P. 3259 - 3261.
19. *Lee J.I. et al.* Experimental study on neutrinoless double beta decay of ^{92}Mo // *Nucl. Instrum. Meth.* - 2011. - Vol. A654. - P. 157 - 161.
20. *Lee J.I. et al.* Experimental study on neutrinoless double beta decay of ^{92}Mo // Reprinted from *Nucl. Instrum. Meth.* - 2011. - Vol. A654. - P. 157 - 161, with permission from Elsevier.
21. *Back H.O. et al.*, Pulse-Shape discrimination with the Counting Test Facility // *Nucl. Instr. Meth.* - 2008. - Vol. A584. - P. 98 - 113.
22. *Lee H.S. et al.* Development of low background CsI(Tl) crystals for WIMP search // *Nucl. Instrum. Meth.* - 2007. - Vol. A571. - P. 644 - 650.

Н. Д. Ханбеков, В. В. Аленков, А. А. Буренков, О. А. Бузанов, В. Н. Корноухов

**АНАЛІЗ ДАНИХ ВИМІРЮВАНЬ ВНУТРІШНЬОГО ФОНУ
СЦИНТИЛЯЦІЙНИХ КРИСТАЛІВ $^{40}\text{Ca}^{100}\text{MoO}_4$**

Чутливість експериментів по безнейтринному подвійному бета-розпаду ($0\nu 2\beta$) в основному залежить від внутрішнього фону детектора, який, у свою чергу, визначається чистотою матеріалу й можливістю відбору фонових подій. Колаборація АМоРЕ (Advanced Mo based Rare process Experiment) планує використовувати сцинтиляційні кристали $^{40}\text{Ca}^{100}\text{MoO}_4$ в якості детектора для пошуку $0\nu 2\beta$ розпаду ізотопу ^{100}Mo . Мета цієї статті – подальше дослідження внутрішнього фону сцинтиляційних елементів $^{40}\text{Ca}^{100}\text{MoO}_4$ в низькофоновій установці в підземній лабораторії Янг-Янг. Ми представляємо нові підходи до відбору фонових подій з аналізованих даних й останні уточнені значення індексу фону кристалів $^{40}\text{Ca}^{100}\text{MoO}_4$ як приклад застосування нової методики.

Ключові слова: безнейтринний подвійний бета-розпад, аналіз даних, сцинтилятори, молибдат кальцію, низькофонові фізика, час-амплітудний аналіз, радіоактивний фон.

Н. Д. Ханбеков, В. В. Аленков, А. А. Буренков, О. А. Бузанов, В. Н. Корноухов

**АНАЛИЗ ДАННЫХ ИЗМЕРЕНИЙ ВНУТРЕННЕГО ФОНА
СЦИНТИЛЛЯЦИОННЫХ КРИСТАЛЛОВ $^{40}\text{Ca}^{100}\text{MoO}_4$**

Чувствительность экспериментов по безнейтринному двойному бета распаду ($0\nu 2\beta$) в основном зависит от внутреннего фона детектора, который, в свою очередь, определяется чистотой материала и возможностью отбора фоновых событий. Коллаборация АМоРЕ (Advanced Mo based Rare process Experiment) планирует использовать сцинтилляционные кристаллы $^{40}\text{Ca}^{100}\text{MoO}_4$ в качестве детектора для поиска $0\nu 2\beta$ распада изотопа ^{100}Mo . Цель этой статьи – дальнейшее исследование внутреннего фона сцинтилляционных элементов $^{40}\text{Ca}^{100}\text{MoO}_4$ в низькофоновой установке в подземной лаборатории Янг-Янг. Мы представляем новые подходы к отбору фоновых событий из анализируемых данных и последние уточненные значения индекса фона кристаллов $^{40}\text{Ca}^{100}\text{MoO}_4$ как пример применения новой методики.

Ключевые слова: безнейтринный двойной бета-распад, анализ данных, сцинтилляторы, молибдат кальция, низькофоновая физика, время-амплитудный анализ, радиоактивный фон.

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