

Neutrino-Less Double Beta Decay Experiment Using Ca $^{100}\text{MoO}_4$ Scintillation Crystals

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Abstract—Search for neutrino-less double beta decay of ^{100}Mo is proposed using active method with Ca $^{100}\text{MoO}_4$ scintillation crystals which show the brightest scintillation light among variety of inorganic scintillation materials containing Mo. Study of X-ray luminescence and scintillation properties such as energy response, number of photoelectrons/keV, absolute light yield, decay time, pulse shape discrimination and radioactive contamination of CaMoO₄ crystals grown by the Czochralski method with different conditions are presented. Further R&D of resolution optimization, crystal quality improvement and background reduction are underway. Significant improvement of sensitivity to neutrino-less double beta decay can be achieved by using ^{100}Mo enriched Ca $^{100}\text{MoO}_4$ crystals with good energy resolution and low background. Further reduction of background induced by ^{48}Ca two neutrino double beta decay can be achieved by using ^{48}Ca depletion. We are planning to install several kilograms of Ca $^{100}\text{MoO}_4$ crystals depleted in ^{48}Ca at underground laboratory for the neutrino-less double beta decay experiment in the near future.

Index Terms—CaMoO₄, double beta decay, energy resolution, internal background, isotopic enrichment.

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I. INTRODUCTION

THE nuclear double beta ($\beta\beta$) decay is a good probe into the fundamental properties of neutrinos as well as weak interactions. The neutrino-less ($0\nu\beta\beta$) decay is forbidden in the Standard Model because it violates lepton number conservation, and the existence of $0\nu\beta\beta$ decay would indicate the existence of nonzero Majorana neutrino mass. The $\beta\beta$ decay of a quite few nuclei have been studied during the last decade. A comprehensive review of experimental and theoretical data on nuclear $\beta\beta$ decays is described in [1], [22], [23]. Many experimental and theoretical investigations show that even the half-life of the $2\nu\beta\beta$ decay process is not precisely predicted because of the strong influence of nuclear structure effects. Thus, experimental searches for $0\nu\beta\beta$ decay on several nuclei are necessary.

The $0\nu\beta\beta$ decay of ^{100}Mo has been studied by ELEGANT V [2], [24] and NEMO3 [3], [25] collaboration. The NEMO3 collaboration reported a limit of 4.6×10^{23} years using 6.9 kg of ^{100}Mo [3], [25]. However, both NEMO3 and ELEGANT V experiments use the spectroscopic technique (passive source method) with a tracking device and thin Mo foil. The detector energy resolution and efficiency of ELEGANT V and NEMO3 were reported as 7–5% and 15%, and 8–10% and 8%, respectively. No calorimetric measurement (active source method) on this isotope has been realized so far. In this letter, we present further development of an experiment to search for $0\nu\beta\beta$ decay of ^{100}Mo using CaMoO₄ crystals with enriched ^{100}Mo proposed earlier in [4]–[6].

Among the variety of inorganic scintillation materials which contain Mo with the structure of scheelite or wolframite, CaMoO₄ shows the brightest scintillation light at room temperature. This scintillation material did not get attention due to its long decay time and moderate light output. However it is not an obstacle for $0\nu\beta\beta$ decay search since it is a rare process. It is already recognized that CaMoO₄ crystal can be used for the ^{100}Mo $0\nu\beta\beta$ decay experiment [4]. Since then, extensive study has been done for the energy resolution, decay time and internal background of the CaMoO₄ crystals [5], [6]. It was shown that alpha particles and gamma rays can be separated using the different time characteristics of the crystal which is useful for the reduction of ^{208}Tl and ^{214}Bi radioactive decay background. It was also demonstrated that the intensity of scintillation light increases as the temperature decreases. Moreover, technology of the crystal growth of CaMoO₄ crystals is improved since then [6], [7].

In this study, we compared four different CaMoO₄ crystals using X-ray induced luminescence and energy response irradiated by various radioactive sources. Scintillation properties



Fig. 1. Photographs of the CaMoO_4 crystals produced at different places.

such as energy resolution with different light sensors, attenuation length for long crystals, number of photoelectrons/keV, and absolute light yield of CaMoO_4 crystals are also presented. Internal background of one CaMoO_4 crystal was measured at the 750 m of minimum depth underground laboratory at YangYang Stored Water Power Plant in Korea (Y2L). We expect significant improvement of sensitivity for the $0\nu\beta\beta$ decay of ^{100}Mo can be achieved by using several kilograms of CaMoO_4 crystals, with good energy resolution and low background, enriched in ^{100}Mo and depleted in ^{48}Ca .

II. EXPERIMENTAL SETUP

A. CaMoO_4 Samples

Fig. 1 shows the photographs of CaMoO_4 crystals produced at different places. The following large CaMoO_4 crystals from the Innovation Centre of the Moscow Steel and Alloy Institute (ICMSAI), Moscow, Russia are used for the crystal characterization [7]:

- 1) CMO-L1: $2.2 \times 2.2 \times 12.5 \text{ cm}^3$ (2006)
- 2) CMO-L2: $3.0 \times 3.0 \times 20.3 \text{ cm}^3$ (2007)
- 3) CMO-L3: $3.0 \times 3.0 \times 21.3 \text{ cm}^3$ (2007)

These crystals are used for the relative light yield comparison and attenuation length measurement. After the test, the first crystal was cut into two pieces, and its internal background was measured at YangYang underground laboratory.

Following four CaMoO_4 crystals from Russia and Ukraine are used for the comparison of light output and X-ray luminescence. CMO-S1, CMO-S2 were produced by ICMSAI, CMO-S3 was produced by Institute of Materials (IM), Lviv, Ukraine and CMO-S4 was produced by Institute of Single Crystals (ISC), Kharkiv, Ukraine:

- 1) CMO-S1: $1.0 \times 1.0 \times 1.0 \text{ cm}^3$ (2004)
- 2) CMO-S2: $1.0 \times 1.0 \times 1.0 \text{ cm}^3$ (2008)
- 3) CMO-S3: $1.0 \times 1.0 \times 1.0 \text{ cm}^3$ (2008)
- 4) CMO-S4: $\varnothing 1.0 \times 1.0 \text{ cm}^3$ (2007)

Also a $\varnothing 3.8 \times 2.0 \text{ cm}^3$ of cylindrical shape crystal (CMO-M) produced by IM is used for the energy resolution optimization. The CMO-M crystal corresponds to the CMO-3 in [6].

B. X-Ray Luminescence

X-ray induced emission spectrum was recorded with an X-ray tube (DRGEM Co.) having a tungsten anode. The power setting was 100 kV and 1 mA. The sample crystal under study was mounted in the sample holder located in front of the X-ray tube. The emission spectrum was measured by using a QE65000 fiber optic spectrometer (Ocean Optics Co.).

C. Radioactive Source Test

Green extended RbCs photocathode photo-multiplier tubes (PMT) of 3 inch diameter (D726Uk from Electron tube Ltd.) are directly attached to the CMO-S or CMO-M crystals surface. The RbCs PMT enhanced the quantum efficiency (QE) of the luminescence from CaMoO_4 crystals since the luminescence peak is at 520 nm (14% QE). The cathode plane of a PMT covers the whole attached area of the crystal surface. Other surfaces are wrapped with Teflon sheet followed by black tape.

Signals from the PMT are amplified using a home-made amplifier ($\times 10$) with low noise and high slew rate. The signals from the amplifier are fed into the 400 MHz FADC. The home-made FADC module is designed to sample the pulse every 2.5 ns for duration up to $64 \mu\text{s}$ so that one can fully reconstruct each photoelectron pulse. The FADC located in a VME crate is read out by a Linux-operating PC through the VME-USB2 interface with a maximum data transfer rate of 10 Mbytes/s. The DAQ system is based on the ROOT [8] package. The 662 keV γ rays from ^{137}Cs radioactive source and 5.5 MeV α particles from ^{241}Am radioactive source were used for the crystal characterization.

We measured the number of electron-hole (e-h) pairs and absolute light yield of the CaMoO_4 crystal with the UV sensitive windowless (630-70-73-510) large area avalanche photodiode (LAAPD) produced by Advanced Photonics Co. It has an excellent quantum efficiency (close to 80%) in the visible and near infrared. The LAAPD is calibrated with X-rays of ^{55}Fe source for the calculation of the number of e-h pairs per channel. Signals from 16 mm LAAPD are fed into a low noise preamplifier (TENNELEC TC175B) followed by a shaping amplifier (Ortec 570). The portable 25 MHz flash analog to digital converter (FADC) board is used for signal digitization. The digitized signals from the 25 MHz FADC are readout by Linux based computer through a USB2 connection.

D. YangYang Underground Laboratory

Since a large amount of budget is needed to build a new underground laboratory, therefore we have taken the opportunity to utilize our existing facility. Korea Middle land Power Co. constructed a pumped storage power plant at YangYang, Korea. The tunnel built for this power plant has earth overburden of approximately 700 m. The muon flux was measured to be $2.7 \times 10^{-7} / \text{cm}^2 / \text{s}$. It is consistent with the water equivalent depth of approximately 2000 m. The rock composition surrounding the laboratory was analyzed by the ICP mass spectrometry. The contamination of ^{238}U and ^{232}Th was reported to be at a level of $< 0.5 \text{ ppm}$ and $5.6 \pm 2.6 \text{ ppm}$, respectively. The contamination level of ^{222}Rn in the air is 1–2 pCi/liter ($40\text{--}80 \text{ Bq/m}^3$) which is comparable to that at the ground level. The neutron flux in the experimental hall is $8 \times 10^{-7} / \text{cm}^2 / \text{s}$.

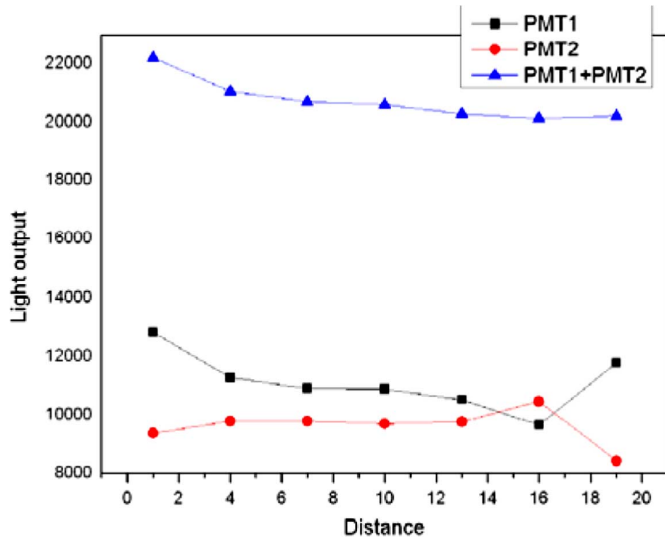


Fig. 2. Light yield difference of CMO-L3 at different position from the PMT. The distance is measured from the PMT 1. The error bars are within the size of points.

for $1.5 \text{ MeV} < E_{\text{neutron}} < 6.0 \text{ MeV}$. The laboratory is currently used for the CsI(Tl) crystal based WIMP search experiment (KIMS). The detail description of YangYang underground laboratory can be found elsewhere in [9], [26].

For the internal background measurement of the CMO-L1 crystal, a setup of 4π CsI(Tl) active shielding detector is used. The 4π CsI(Tl) active shielding detector is composed of 14 CsI(Tl) crystals: Out of these 14 CsI(Tl) crystals, 12 long CsI(Tl) crystals having dimension of $65 \times 55 \times 300 \text{ mm}^3$, while the other two endcap CsI(Tl) crystals having dimension of $90 \times 90 \times 60 \text{ mm}^3$. An acrylic case is placed inside the 4π CsI(Tl) to support the crystals. The whole detector assembly is shielded by 10 cm thick lead blocks, in order to reduce the background. The N_2 gas is flown for the reduction of ^{222}Rn background.

E. Large Crystal Characterization

Two PMTs are attached to each face of the CMO-L1, CMO-L2 and CMO-L3 crystals. The relative light yields of these crystals are compared with CMO-S1 crystal irradiated with 662 keV γ -ray source. After comparison, the measured relative light yields of CMO-L1, CMO-L2 and CMO-L3 are 90%, 63% and 75%, respectively. For the cross checking, ^{241}Am α -source is used for the relative light output comparison. Attenuation length of the CMO-L1, CMO-L2 and CMO-L3 crystals are measured with ^{241}Am α -source since it is easy to identify the peak and the interaction point in the crystal is well defined. We made holes in the Teflon wrapping at every 2 cm for the CMO-L1 crystal and 3 cm distance of the CMO-L2 and CMO-L3 crystals for α -source irradiation. As shown in Fig. 2, total light yield with different position from the PMT shows light yield variation within 10%. However, the end point of each side of PMT shows abnormal behavior since that point is only 1 cm distance from the each PMT. If we remove first and last points as they are very close to the PMT, the attenuation length is measured to be bigger than 50 cm. The attenuation length and characteristics of the CMO-L1 and CMO-L2 are similar to that of CMO-L3.

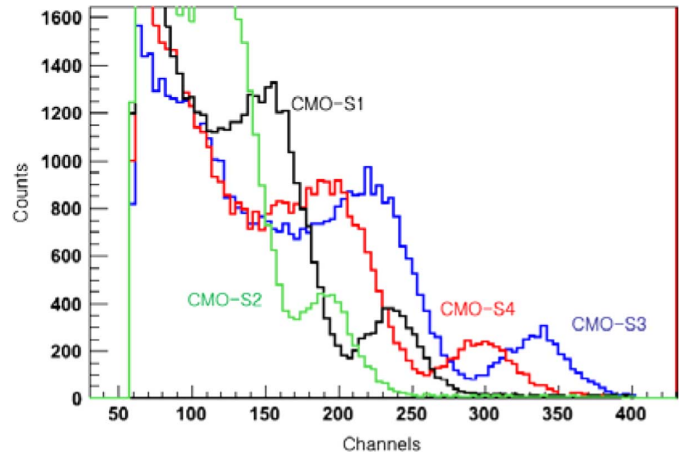


Fig. 3. Light yield comparison between four different CaMoO₄ crystals with 662 keV γ -rays.

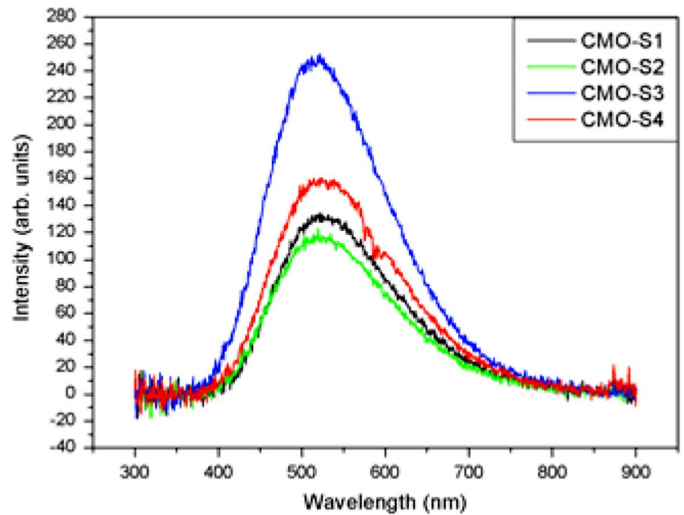


Fig. 4. X-ray luminescence spectra of CaMoO₄ crystals.

F. Comparison of Different CaMoO₄ Crystals

Relative light yield of CMO-S1, -S2, -S3 and -S4 crystals are compared at 662 keV γ -rays from a ^{137}Cs radioactive source. The CMO-S2 shows lowest light yield and the CMO-S1, CMO-S3 and CMO-S4 shows 1.24, 1.76 and 1.57 times higher light yields than that of the CMO-S2 respectively, as shown in Fig. 3. The large light output difference between the CMO-S2 and the CMO-S3 is under investigation.

The luminescence spectra of the four crystals are studied under X-ray excitation at room temperature as shown in Fig. 4. The emission spectra show broad emission bands in the wavelength range from 400 nm to 700 nm with peak emission at 520 nm. This result is in agreement with [10]. Even if pulse heights are different, the shapes of the spectra are very similar to one another.

G. Number of Photoelectron Measurement

Single Photoelectron (SPE) signal can be identified at low energy since 400 MHz FADC is used for the signal digitization. The identification of SPE can reduce the effect coming from the noise and improve the energy resolution. A clustering algorithm

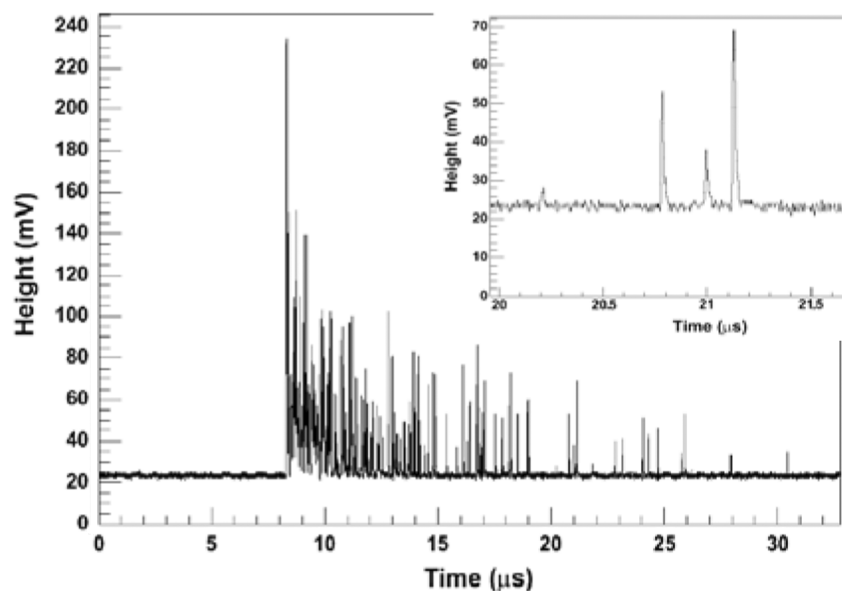


Fig. 5. Typical pulse spectra of the CaMoO_4 crystal by 400 MHz FADC and SPEs are also shown.

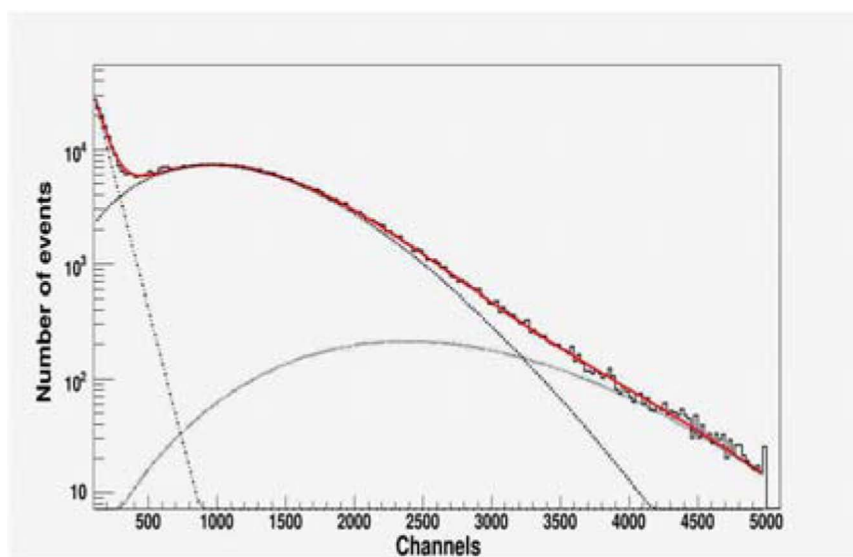


Fig. 6. SER pulse height spectrum was fitted with one exponential (dashed-dot curve), one photoelectron (dashed curve) and two photoelectron (dotted curve).

is developed to identify SPEs [9], [26]. We investigated scintillation characteristics of the crystal using single photoelectron (SPE) counting method in $24 \mu\text{s}$ window since the trigger point is set to be $8 \mu\text{s}$ at room temperature as shown in Fig. 5. In SPE counting method, it needs to identify SPE signal to reduce the effect coming from the noise to improve energy resolution because the information for the scintillation light from the detector is included only in the SPE signal region. We used a clustering algorithm to identify SPEs. If the decay time of a scintillator is long, the SPE counting method is useful for the characterization of the scintillator.

In order to measure the number of photoelectrons per keV the CMO-M crystal was cooled to 6°C . In this case, the FADC window is set to be $82 \mu\text{s}$ with the trigger point of $20 \mu\text{s}$ since the decay time becomes longer when the crystal is cooled down. The number of photoelectrons per keV can be calculated with single electron response (SER) using an LED pulse as shown in

Fig. 6. The fitted function was taken from [9], [26]. The fitted functions were overlaid in the figure. The ratio of the two photoelectron contribution was obtained to be $\sim 4\%$. The number of photoelectrons were measured by comparing the SER with the full peak of 662 keV produced by ^{137}Cs radioactive source. We obtained 1.16 ± 0.12 photoelectrons per keV. If only photoelectron statistics are considered, the energy resolution of 4% can be obtained at 3 MeV.

H. Energy Resolution

The energy resolution of the CaMoO_4 crystal (CMO-M) was studied at different temperatures. This involved the coupling of the crystals to the PMT and irradiated with 662 keV γ -rays from a ^{137}Cs source. From the recorded photo peaks, the energy resolutions were calculated by a Gaussian fitting. The energy resolution at 662 keV was obtained to be 11% (FWHM) at 6°C as

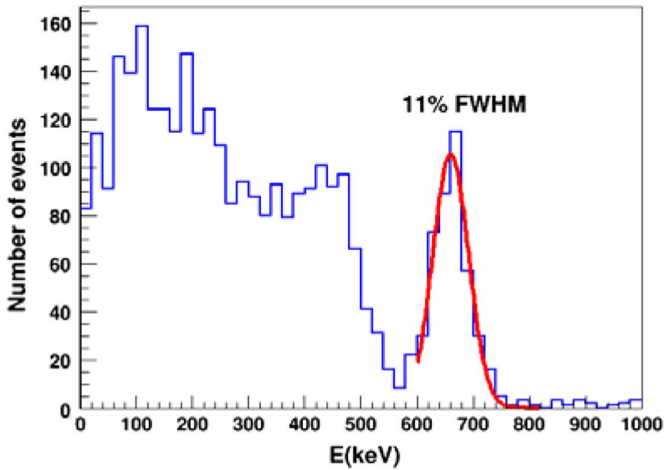


Fig. 7. Energy resolution of CMO-M crystal irradiated with 662 keV γ -rays.

shown in Fig. 7. This energy resolution is better than 11.9% at room temperature with the same crystal from [6].

Also energy resolution study of the CaMoO₄ crystal (CMO-L1) is done with the LAAPD at low temperature. Due to the long decay time at low temperature as shown in [6], preamp output signals are digitized directly and used for the analysis. At -159 °C, the energy resolution is measured to be 11% at 662 keV γ -rays.

I. Absolute Light Output

We measured the number of e-h pairs and absolute light yield of the CMO-L3 at room temperature using the LAAPD [10], [11]. The sample crystal is coupled to the UV-enhanced LAAPD with 10 μ s shaping time constant of spectroscopic amplifier. The crystal is irradiated with the 5.5 MeV α particles from a ²⁴¹Am source for clear peak identification and α/β ratio of 0.20 is used for the correction. After calibration with ⁵⁵Fe source, the number of e-h pairs of the crystal is determined to be $3\,500 \pm 350$ e-h/MeV. The absolute light yield is obtained to be $4\,900 \pm 590$ photons/MeV after correction for light collection efficiency of the LAAPD ($80 \pm 3\%$) and Teflon reflection ($90 \pm 5\%$).

J. Background

CMO-L1 was cut into two pieces and coupled with green extended PMT. The internal background of the crystals are measured at the Y2L underground laboratory with 4π CsI(Tl) active shielding as described in Section II-D.

Timing-correlation analysis is performed to the ²³⁸U series where the ²¹⁴Bi-²¹⁴Po decay sequence with time-correlated $\beta - \alpha$ signatures are selected as ²¹⁴Bi ($Q_\beta = 3.27$ MeV) \rightarrow ²¹⁴Po ($Q_\alpha = 7.83$ MeV, $T_{1/2} = 163.7$ μ s) \rightarrow ²¹⁰Pb. The background level of the crystal is measured to be 0.32 ± 0.01 mBq/kg, which is one order of magnitude lower than that of CMO-M produced by CARAT Co. [6]. Similar analysis is applied to the ²³²Th series where the ²²⁰Rn-²¹⁶Po decay sequence with time-correlated $\alpha - \alpha$ signatures are selected as ²²⁰Rn ($Q_\alpha = 6.41$ MeV) \rightarrow ²¹⁶Po ($Q_\alpha = 6.91$ MeV, $T_{1/2} = 0.145$ s) \rightarrow ²¹²Pb. The background level of the crystal is measured to be 0.22 ± 0.05 mBq/kg, which is consistent with that of CMO-M produced by CARAT Co. in [6].

III. DISCUSSION AND PROSPECT

We plan to place 6 kg of ¹⁰⁰Mo enriched and ⁴⁸Ca depleted CaMoO₄ crystals inside 6 cm thick CsI(Tl) Compton veto detectors, and the outer shielding setup can be similar to the KIMS experiment [9], [26]. A FADC with 400 MHz sampling rate with 10 bit being developed for the KIMS experiment can be used for the data taking. One drawback of CaMoO₄ is the background from $2\nu\beta\beta$ decay of ⁴⁸Ca whose natural abundance is 0.187%. The Q-value of $2\nu\beta\beta$ is 4272 keV and the half-life is 4.2×10^{19} years [12], [13]. Since one of serious background of CaMoO₄ crystal is expected from $2\nu\beta\beta$ decay of ⁴⁸Ca, we plan to use the CaCO₃ powder with ⁴⁸Ca depletion for producing the CaMoO₄.

Four main types of background should be considered for the proposed experiment.

- 1) $2\nu\beta\beta$ decay of ¹⁰⁰Mo;
- 2) $2\nu\beta\beta$ decay of ⁴⁸Ca;
- 3) Radioactive background from ²⁰⁸Tl and ²¹⁴Bi decay chains;
- 4) Cosmic ray induced background.

Due to 30 cm of liquid scintillator 4π active shielding, cosmic ray background can be easily eliminated. Furthermore, 50 μ s veto after cosmic muon tagging can eliminate the inelastic scattering background induced by cosmic muons.

Radioactive impurity in CaMoO₄ crystal can also be a source of background in the vicinity of expected $0\nu\beta\beta$ decay peak. Specially, ²⁰⁸Tl to ²⁰⁸Pb beta decay in the ²³²Th decay chain has the highest Q_β value. Also ²¹⁴Bi beta decay in the ²³⁸U decay chain can reach the $Q_{\beta\beta}$ value of ¹⁰⁰Mo. It is known that a high level of chemical purification of molybdenum was obtained by NEMO group [14], while a satisfactory radiopurity of scintillators was reached by the Kyiv group with CdWO₄ crystals [15]. Background from impurities are assumed to be small, purification of both ²³⁸U and ²³²Th decay chain background level to 0.05 mBq/kg, Compton veto and time correlation rejection with pulse-shape discrimination could provide an internal background due to radio-isotopes in ²³⁸U and ²³²Th decay series less than 0.01 counts/keV/kg/y.

With the development of high purity CaMoO₄ and good uniformity, one would expect to achieve an energy resolution as good as 5% FWHM at 3 MeV [6]. With these assumptions, the sensitivity to the half-life of ¹⁰⁰Mo $0\nu\beta\beta$ decay with 6 kg CaMoO₄ crystals 95% enriched in ¹⁰⁰Mo and depleted in ⁴⁸Ca for three years data taking is 6×10^{24} years with 90% confidence level which corresponds to a 0.2–0.7 eV neutrino mass using several nuclear matrix element (NME) calculations of ¹⁰⁰Mo [16]–[18]. Fig. 8 shows various backgrounds and signal with GEANT4 simulation [19]; the major background is from ²⁰⁸Tl decay, ²¹⁴Bi decay and $2\nu\beta\beta$ decay of ¹⁰⁰Mo. We could achieve the sensitivity of 7×10^{26} years (corresponding to mass of neutrino 20–70 meV) for five years of data taking with 100 kg of ¹⁰⁰Mo with ⁴⁸Ca depletion. The MOON (Majorana/Mo Observatory Of Neutrinos) project aims at study of neutrino mass of 100 meV and 30 meV with the ¹⁰⁰Mo target mass-year of 0.5 and 2 ton-year, respectively [20].

Since cryogenic detection technique guarantees high energy resolution as well as low energy threshold, a cryogenic detector with metallic magnetic calorimeter (MMC) method was employed to measure the energy resolution of CaMoO₄ crystal. The preliminary result with the CaMoO₄ crystal of

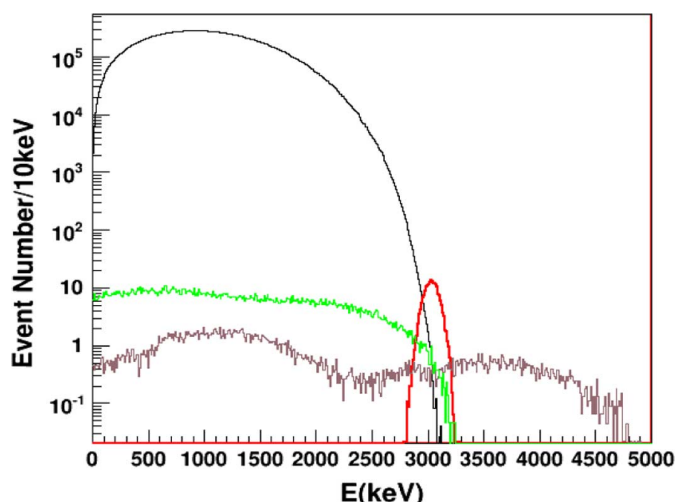


Fig. 8. Various backgrounds and signal distribution by GEANT4 simulation with 5% FWHM resolution, 6 kg of CaMoO_4 crystals with 3 years data taking.

size of $1\text{ cm} \times 1\text{ cm} \times 0.7\text{ cm}$ showed the energy resolution of 11.8 keV for 5.5 MeV alpha particles [21] and it could further be improved by factor three with better control of radioactive source. With the same condition of room temperature except assuming energy resolution of 15 keV at 3 MeV, the sensitivity to the half-life of $0\nu\beta\beta$ decay with 6 kg CaMoO_4 could be improved to 2×10^{25} years which corresponds to a 0.1–0.4 eV neutrino mass.

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