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A Study of Radioactive Contamination of $^{40}Ca^{100}MoO_4$ Crystals for the AMoRE Experiment

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***Abstract*—A calcium molybdate ($CaMoO_4$) crystal scintillator, with molybdenum enriched in ^{100}Mo and calcium depleted in ^{48}Ca ($^{40}Ca^{100}MoO_4$), was developed by the Advanced Molybdenum based Rare process Experiment (AMoRE) collaboration to search for a neutrinoless double beta ($0\nu\beta\beta$) decay of ^{100}Mo. We are planning to use about 10 kg of $^{40}Ca^{100}MoO_4$ crystals as cryogenic bolometers for the first phase of the experiment (AMoRE-I) at the Yang Yang underground laboratory (Y2L) in Korea. This experiment calls for an extremely low level of radioactive contamination in detectors, particularly by thorium, uranium, and radium decay chains. We measured scintillation properties and radioactive contamination of $CaMoO_4$ and $^{40}Ca^{100}MoO_4$ crystals at the Y2L. We also estimated the acceptable level of internal radioactive background using Monte Carlo simulation for the AMoRE-I.**

***Index Terms*—$CaMoO_4$, double beta decay, radioactive contamination, time-amplitude analysis.**

I. Introduction

NEUTRINOLESS double beta ($0\nu\beta\beta$) decay is forbidden in the Standard Model; however, neutrino oscillation results established the existence of nonzero neutrino mass [1], [2], and the $0\nu\beta\beta$ decay is the most promising avenue for learning more about the nature of neutrinos. The $0\nu\beta\beta$ process can only occur if the neutrinos are Majorana type, i.e., particle and antiparticle are identical [3]. The Advanced Molybdenum based Rare process Experiment (AMoRE) collaboration is searching for the $0\nu\beta\beta$ decay of ^{100}Mo [4]. We chose $CaMoO_4$ scintillating crystals as both the ^{100}Mo source and the $0\nu\beta\beta$ detector because it has the highest scintillation efficiency at cryogenic

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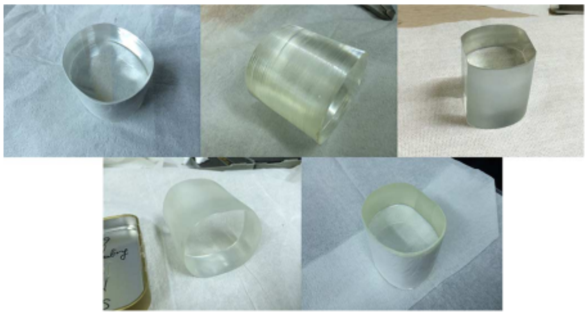

Fig. 1. $CaMoO_4$ crystals. Clockwise from top left: SS68, S35, SE1, CARAT, and NSB29.

temperature among the Mo-contained scintillators [5], [6]. We have developed $CaMoO_4$ crystals from molybdenum enriched in ^{100}Mo and calcium depleted in ^{48}Ca to increase the experimental sensitivity and decrease the $2\nu\beta\beta$ background from ^{48}Ca. The simultaneous usage of $^{40}Ca^{100}MoO_4$ crystals as the $0\nu\beta\beta$ decay source and the detector maximizes the experimental efficiency [7]. We are planning to use an approximately 10 kg array of $^{40}Ca^{100}MoO_4$ crystals as cryogenic scintillating bolometers for the first phase of the experiment (AMoRE-I).

The $0\nu\beta\beta$ decay process is very rare; the NEMO-3 experiment, which has studied the $0\nu\beta\beta$ decay of ^{100}Mo, reported a half-life limit of $T_{1/2}(0\nu\beta\beta) > 1.1 \times 10^{24}$ years at the 90% confidence level [8]. The sensitivity of the experiment is inversely proportional to square root from background rate [9]. Therefore, reduction of background is important feature of any $0\nu\beta\beta$ experiment. The $^{40}Ca^{100}MoO_4$ crystals were produced by the JSC FOMOS-Materials Co., Moscow, Russia, which also carried out materials purifications [10], and the radio purity of several samples has been already tested [11]. We studied the influence of background in the AMoRE-I experiment using a Monte Carlo simulation. The required contamination levels for ^{226}Ra (^{238}U chain), ^{228}Th (^{232}Th chain) and ^{227}Ac (^{235}U chain) are ≤ 100 µBq/kg, 50 µBq/kg and 500 µBq/kg, respectively.

In this study, we analyzed the internal radioactive contamination of $CaMoO_4$ crystals using a time-amplitude analysis. In order to decrease the effect of external background, especially induced by cosmic rays, these measurements were carried out in the 700-m-deep Yang Yang (Y2L) underground laboratory at the Yang Yang Stored Water Power Plant in Korea.

II. $CaMoO_4$ Crystals

A. Features of $CaMoO_4$ Crystals

We have tested five $CaMoO_4$ crystals, which are shown in Fig. 1. Four crystals were grown by the JSC FOMOS-Materials Co. These crystals are enriched in ^{100}Mo and depleted in ^{48}Ca ($^{100}\text{Mo} - 96.1\%$, $^{48}\text{Ca} < 0.001\%$). One $CaMoO_4$ crystal from natural molybdenum was produced by the CARAT Co., Lviv, Ukraine. Their masses are listed in Table I.

TABLE I
Parameters of $CaMoO_4$ Crystals

Name	Weight (g)	Note
SS68	350	Nb doped (300 ppm)
NSB29	390	Thermal annealing
S35	256	
SE1	353	
CARAT	411	Natural

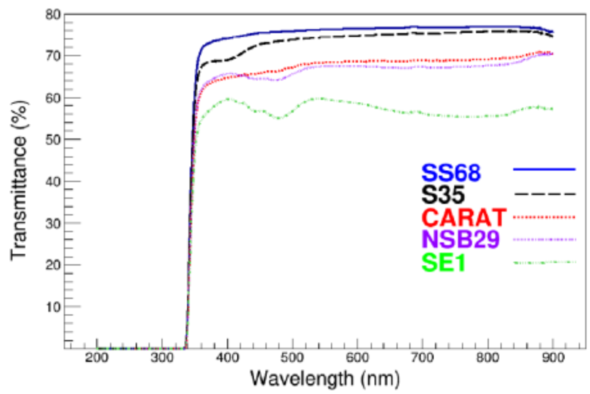

Fig. 2. Optical transmittances of $CaMoO_4$ crystals (SS68: 4 cm, S35: 4 cm, CARAT: 5 cm, NSB29: 5 cm, SE1: 5 cm).

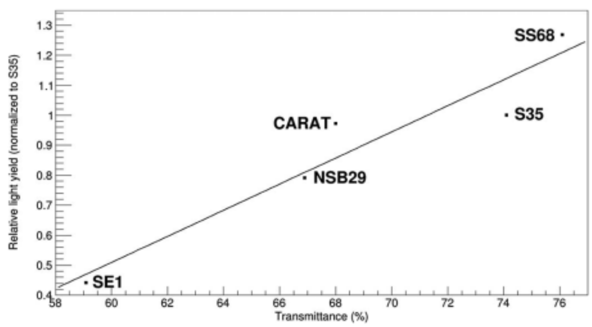

Fig. 3. Relative light yield versus 520-nm transmittance of $CaMoO_4$ crystals. The straight line indicates the observed positive correlation.

B. Transmittance Versus Light Yield Correlation

Fig. 2 shows the transmittances of $CaMoO_4$ crystals measured with a JASCO V-650 spectrometer. A positive correlation between the light yield and transmittance of the crystals is observed. Fig. 3 shows this correlation for the transmittance at 520 nm (peak emission wavelength of $CaMoO_4$ [11]). The relative light yield is defined as the ratio between the positions of the full absorption peak in the tested samples and S35.

C. Light Yield and Energy Resolution

All the $CaMoO_4$ crystal scintillators were calibrated in the with a ^{22}Na gamma source. The calibration spectra are shown

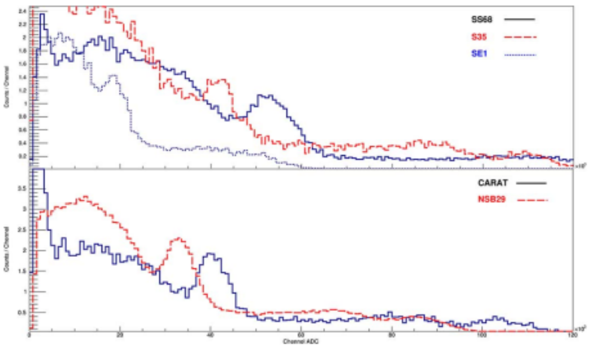

Fig. 4. Energy response spectra of $CaMoO_4$ crystals accumulated using ^{22}Na as the radioactive source. (Top) SS68 (solid line), S35 (dotted line), SE1 (dashed line). (Bottom) CARAT (solid line), NSB29 (dashed line).

TABLE II
RELATIVE LIGHT YIELD, ENERGY RESOLUTION, AND TRANSMITTANCE OF $CaMoO_4$ CRYSTALS

Name	Relative Light Yield* (Normalized to S35)	Energy Resolution** (%)	Transmittance at 520 nm (%)
SS68	1.3	18.4	76
NSB29	0.78	18.6	67
S35	1.0	15.0	74
SE1	0.44	23.6	59
CARAT	0.97	16.5	68

* S35 is the reference crystal. ** FWHM at 511 keV (^{22}Na radioactive source).

in Fig. 4. Measurement results of light yields and energy resolutions are listed in Table II.

III. LOW-BACKGROUND EXPERIMENT

A. Yang Yang Underground Laboratory

The Y2L is under 700-m minimum Earth overburden, which is equivalent to about 2000-m water depth. The cosmic-ray muon flux in the laboratory is measured to be $2.7 \times 10^{-7}/\mathrm{cm}^2/\mathrm{s}$ [12]. Presently, Y2L is also utilized by the Korea Invisible Mass Search (KIMS) dark matter search experiment using room temperature CsI(Tl) and NaI(Tl) scintillating crystals.

B. Experimental Setup

We measured the background of $CaMoO_4$ crystals in the low-background setup at the Y2L. For these measurements, the crystal under study was placed inside an active 4π CsI(Tl) detector shield that is used to veto external gamma background as well as cosmic muons. The veto counter consists of 14 CsI(Tl) crystals. Twelve 30-cm-long CsI(Tl) crystals surround the cavity where the $CaMoO_4$ detector was placed, while two shorter CsI(Tl) crystals were installed to cover the end region. The long CsI(Tl) crystals have an asymmetric geometry shape, where one end is $65\ \mathrm{mm} \times 65\ \mathrm{mm}$, and the other end has $55\ \mathrm{mm} \times 55\ \mathrm{mm}$ as shown in Fig. 5. A 3-inch PMT is coupled to the larger end, and a 2-inch PMT is attached to the

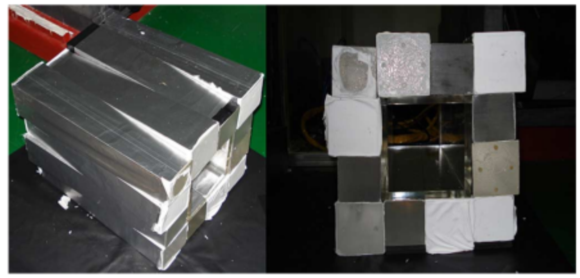

Fig. 5. Dimension and structure of long CsI(Tl) crystals.

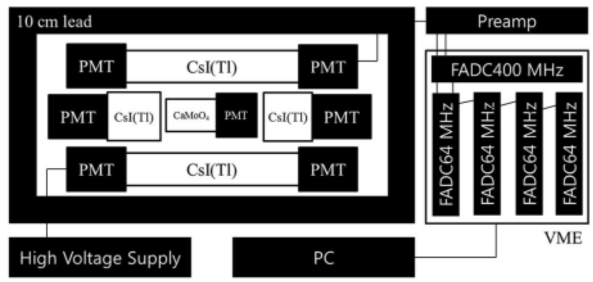

Fig. 6. Schematic of the whole detection system.

smaller end. The veto system is continuously flushed by N_2 gas to reduce the radon-induced background. It is also surrounded with a 10-cm-thick low-background lead shield to reduce the external background. The $CaMoO_4$ crystal is readout with a green-enhanced 3-inch PMT.

The output signals from the PMTs, coupled to the $CaMoO_4$ and CsI(Tl) crystals are amplified by a home-made low-noise amplifier with a gain of 10, and transmitted to Flash Analog-to-Digital Convertors (FADCs) made by NOTICE Co. [13]. It is comprised of one 400-MHz FADC and four 64-MHz FADCs. The 400-MHz FADC is used for the $CaMoO_4$ signals and provides a trigger to the 64-MHz FADC modules as shown in Fig. 6. The FADC400 has an 82 μs window and digitizes the signal every 2.5 ns. The FADCs are located in a VME crate and are read out by a Linux computer via a USB2 connection. The recorded data are then analyzed with a data analysis program based on ROOT package [14].

IV. DATA ANALYSIS AND RESULTS

Alpha particles emitted by alpha active U/Th daughters appear in the gamma energy region, below 3 MeV, due to the quenching of scintillation efficiency. There are fast subchains in the ^{232}Th, ^{235}U, and ^{238}U families, which can be selected by time-amplitude analysis [15]. Using the time-amplitude analysis, isotopes having a relatively short half-life could be selected out by their characteristic energy and decay-time distributions. In the ^{235}U family: $^{219}\mathrm{Rn}$ ($Q = 6.95$ MeV, $T_{1/2} = 3.96$ s) $\rightarrow$ $^{215}\mathrm{Po}$ ($Q = 7.53$ MeV, $T_{1/2} = 1.78$ ms) $\rightarrow$ $^{211}\mathrm{Pb}$ can be selected. We selected all pairs of events, prompt, and delayed, in a time interval of 1–6 ms (corresponding to 58% events selection efficiency for ^{215}Po decay). We selected the delayed events

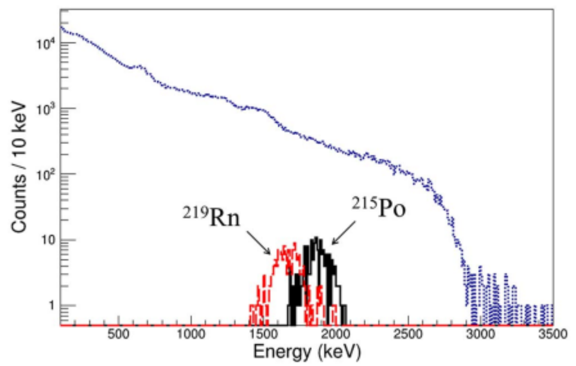

Fig. 7. Energy spectra of ^{215}Po (solid spectrum) and ^{219}Rn (dashed spectrum) events selected with the time-amplitude analysis from the data accumulated with the SS68 detector over 36.5 days (dotted histogram).

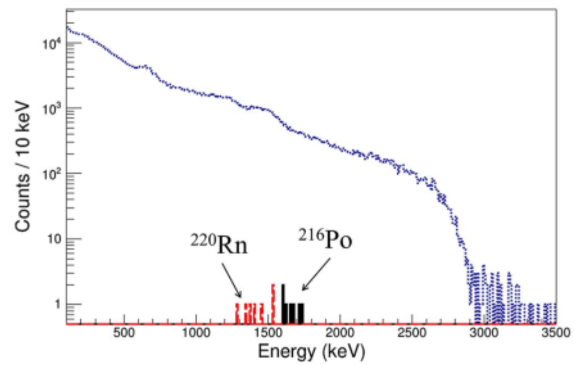

Fig. 9. Energy spectra of ^{216}Po (solid spectrum) and ^{220}Rn (dashed spectrum) events selected with time-amplitude analysis from the data accumulated with the SS68 detector over 36.5 days (dotted histogram).

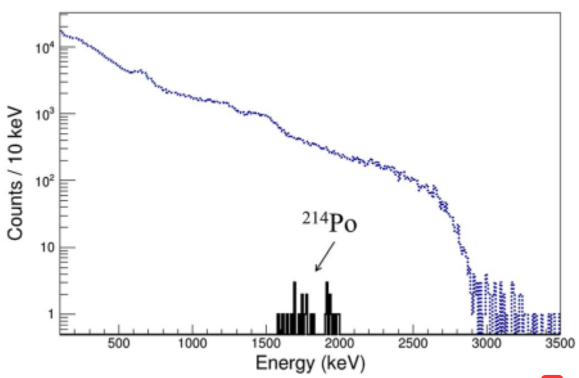

Fig. 8. Energy spectra of ^{214}Po (solid spectrum) events selected with time-amplitude analysis from the data accumulated with the SS68 detector over 36.5 days (dotted histogram).

TABLE III
RADIOACTIVE CONTAMINATION OF $CaMoO_4$ CRYSTALS

Sample	Measurement time	^{227}Ac (^{235}U family)	^{226}Ra (^{238}U family)	^{228}Th (^{232}Th family)
	days		µBq/kg	
SS68	36.5	200 ± 14	60 ± 8	30 ± 5
NSB29	59.8	700 ± 26	200 ± 14	80 ± 9
S35	19.9	1200 ± 35	4400 ± 66	500 ± 22
CARAT	45.5	90 ± 10	1500 ± 39	230 ± 15
SE1	31.9	60 ± 8	40 ± 12	50 ± 15

in the energy intervals within 3-sigma around the expected alpha peak of ^{215}Po. We plotted the prompt events' energies of their corresponding delayed events. The selected events are presented in Fig. 7. The energies of the peaks correspond to ^{219}Rn and ^{215}Po. The activity of the subchain progenitor ^{227}Ac (^{235}U family) is determined to be 200 ± 14 µBq/kg.

To estimate activity of ^{226}Ra (^{238}U family), the subchain—^{214}Bi ($Q = 3.27$ MeV, $T_{1/2} = 19.9$ min) $\to$ ^{214}Po ($Q = 7.83$ MeV, $T_{1/2} = 164$ µs) $\to$ ^{210}Pb—has been analyzed in the time interval of 100–600 µs (corresponding to 57.6% of selection efficiency for ^{214}Po decay). The detection efficiency of the ^{214}Bi beta events in the energy interval of 100–1000 keV was determined to be 66.2% by Monte Carlo simulation. The activity of ^{226}Ra is determined to be 60 ± 8 µBq/kg. The selected ^{214}Po events are shown in Fig. 8.

In the ^{232}Th family, the subchain ^{220}Rn ($Q = 6.40$ MeV, $T_{1/2} = 55.6$ s) $\to$ ^{216}Po ($Q = 6.91$ MeV, $T_{1/2} = 0.145$ s) $\to$ ^{212}Pb was analyzed to estimate activity of ^{228}Th in the crystal. However, selection of alpha events from the chain is difficult due to the relatively long half-life of ^{216}Po. To avoid selection of random background events, a pulse shape discrimination (PSD) was used to separate α signals from β/γ backgrounds [16]. Because of the similar Q-values, we have used the selected ^{219}Rn events as reference samples to determine the mean time for α signals. The activity of ^{228}Th in the crystal is determined to be 30 ± 5 µBq/kg. The selected ^{220}Rn and ^{216}Po peaks are shown in Fig. 9. The data on radioactive contamination of all five $CaMoO_4$ crystals are summarized in Table III.

V. CONCLUSIONS

We studied transmittance, relative light yield, energy resolution, and radioactive contamination of $CaMoO_4$ crystals. A positive correlation between the transmittances and relative light yield was observed for all five tested crystals. Using the time-amplitude analysis, we have identified the fast decay subchains, from ^{235}U, ^{238}U, and ^{232}Th families. Pulse shape discrimination between beta (gamma) and alpha events was applied to select events of ^{220}Rn and ^{216}Po alpha decays from the ^{232}Th chain. The radioactive contamination level of a recently produced $^{40}Ca^{100}MoO_4$ crystal (SE1) satisfies the requirements of the AMoRE-I experiment. We have installed five $^{40}Ca^{100}MoO_4$ crystals (1.5 kg) in cryogenic setup for the AMoRE-pilot experiment at the Y2L.

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