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A study of CaMoO₄ crystals for the AMoRE Experiment

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1. Introduction

The resent observations of neutrino oscillations are conclusive evidence that the neutrino has a non-zero mass, and provides motivation for neutrinoless double beta decay (0ν DBD) searches. Searches for 0ν DBD are challenging experimental approaches aimed at establishing the neutrino's nature (Dirac or Majorana) and absolute mass scale [1-3].

A scintillating crystal that contains a candidate isotope for DBD provides a promising technique for the highdetection-efficiency and good energy resolution. The AMoRE (Advanced Molybdenum based Rare process Experiment) collaboration is studying the potential of CaMoO₄ scintillation crystals for searches for 100 Mo 0 ν DBD [4, 5]. ¹⁰⁰Mo is one of the best candidate isotopes for 0ν DBD searches because of its high Q-value (3,034 keV) and a reasonable natural abundance (9.63%) that results in manageable costs for enrichment. However, natural CaMoO₄ crystal contains an unavoidable background from the 2ν DBD of ⁴⁸Ca (0.187% of natural abundance). In order to reduce this background, we have developed ⁴⁸Ca depleted, ¹⁰⁰Mo enriched ⁴⁰Ca¹⁰⁰MoO₄ scintillation crystals: (SB28, SB29, and S35) [6]. Measurements of scintillation properties of and internal backgrounds in these crystals were carried out at the YangYang underground Laboratory (Y2L), which was originally established for dark matter searches in Korea [7].

Here, we report on the development of $CaMoO_4$ crystals, results of scintillation property measurements, and estimates of the internal backgrounds in the enriched crystals.

2. General properties of CaMoO₄ crystals

We have 3 enriched ${}^{40}Ca^{100}MoO_4$ scintillation crystals (SB28, SB29, and S35) that were produced at JSC

FOMOS-Materials in Russia [6] and 1 natural CaMoO₄ crystal (CARAT 1) produced by the CARAT Co. in Ukraine for ¹⁰⁰Mo 0 ν DBD searches.

The enriched crystal S35 was grown directly from the raw ⁴⁰Ca¹⁰⁰MoO₄ in pallet form. The as-grown crystal was slightly blue and after 40 hours annealing in the air became transparent. In a complementary approach, crystals SB28 and SB29 were grown from a recrystallized initial charge to reduce the internal contamination from the raw material. The as-grown crystals SB28 and SB29 exhibited a strong blue color due to the oxygen and Mo⁶⁺ ions deficiencies. In order to reduce internal contamination from the raw material, there was no additional charging of MoO₃ during the SB28 and SB29 crystal growing [6]. As is evident in Fig. 1, S35 is much clearer than SB28 and SB29. The CARAT1 crystal was grown from two natural raw crystals that were produced from raw CaMoO₄ in powder form. These crystals were grown by the Czochralski method (detailed processes are described in Ref [6]). The shape of each crystal is roughly an elliptic cylinder; the dimensions and mass are as follows (quoted are the major and minor axes of an ellipse, heights of the crystal and its mass): (Table 1. and Fig 1.)

	SB28	SB29	S35	CARAT#1
Height (mm)	25.5	51.0	40.0	50.0
Major axis (mm)	49.5	50.0	45.0	53.0
Minor axis (mm)	40.5	42.5	40.0	44.0
Mass (g)	196	390	256	411

Table 1. The dimensions and masses of CMO crystals.



Fig 1. Pictures of CaMoO₄ scintillation crystals for AMoRE.

3. Scintillation properties of CaMoO₄ crystals

The emission spectra of SB28, S35, and CARAT1 samples that were cut from the same ingots, were measured using X-ray irradiation with a QE65000 spectrometer (Ocean Optics Co.). The dimensions of SB28 and S35 were same $(10\times10\times10 \text{ mm}^3)$ with a cubic shape, however, the sample of CARAT1 was thinner and different shape (ϕ 10 mm x 1 mm). All of these samples appeared transparent and colorless.



Fig 2. Emission spectra of $CaMoO_4$ crystal samples under the X-ray irradiation.

Fig 2 shows the X-ray emission spectra of the three samples. The peaks all occur at the same peak wavelength value 520 nm. This means that the enrichment does not have any effect on the scintillation processes and we can expect high quantum efficiency with a green enhanced PMT [8].

The relative light output, energy resolution and decay time were measured using a ¹³⁷Cs γ -ray source. 3 inch of green enhanced PMT was used for higher quantum efficiency than bi-alkali PMT at the 520 nm of emission wavelength. The PMT signal was fed into the home-made preamplifier (x 10). The amplified signal was digitized using 400-MHz flash analog to digital convertor (FADC), which was produced by Notice-korea Co. The data acquisition process was controlled by Linux based ROOT package.

SB29 and SB28 produced less light output than S35 and CARAT1, because of the shorter attenuation length caused by higher oxygen and Mo⁶⁺ deficiency, mentioned previously. The energy resolution of S35 and CARAT1 were much better than SB28 and SB29, because of the higher light output. However, the decay times of the crystals were measured to be quite similar

as 17μ s at room temperature. Table 2. shows the scintillation properties of the CMO crystals. The absolute light yields was obtained to comparing with a reference CsI(Tl) crystal.

	SB28	SB29	S35	CARAT#1
Emission (nm)	520	-	520	520
Light yield (Photon/MeV)	2,970	1,970	6,150	6,110
Resolution on 662 keV (%)	28.6	35.6	15.6	16.3
Decay time (µs)	17.3	-	17.0	-

Table 2. Scintillation properties of CMO crystals.

We confirmed that the enrichment does not affect to the scintillation properties between S35 and CARAT#1. However, several steps of re-crystallization process with no additional charging of MoO_3 would not be expected a better scintillation property.

4. Internal background of CaMoO₄ crystals

We accumulated background data with S35 for 90 days, SB28 for 47 days, and SB29 for 78 days and used it to estimate the internal background from the ²³⁸U/²³²Th decay chains. The analysis of the SB29 background data was not successful because of its poor energy resolution. The background data were accumulated with a 4π gamma veto system, which was previously used to search for 0ν EC/ β^+ decay of ⁹²Mo and ⁸⁴Sr [9, 10], to isolate internal background events only, and time amplitude analysis [11] and previous event cuts were used for the selection of specific fast sequences in the ²³⁸U/²³²Th chains [4, 8, 9, 11]. The selected α events were verified using pulse shape discrimination (PSD).

The internal background level of S35 was much higher than SB28. In particular, there was a high event rate in the ²¹⁰Po α -decay energy region of the S35 spectrum. In order to determine the origin of the internal background in S35, another experimental setup is being prepared that will measure coincidence events between S35 and a HPGe detector. Fig 3. shows the measured energy distributions of internal background of S35 (a) and SB28 (b) with 4π gamma veto system. As shown in Fig. 3, ²¹⁰Po α -decay rate of SB28 was lower than that of S35. It could be explained that the internal background level can be reduced by re-crystallization process, even it performed worth scintillation properties for higher oxygen and Mo⁶⁺ deficiency.



Fig 3. The internal background spectra of S35 (a) and SB 28 (b). (Red : 214 Po α -decay, Blue : 216 Po α -decay, Green : 220 Rn α -decay)

The internal background of CARAT#1 also has been measured with HPGe detector. The consistence of HPGe coincidence measurement with 4π gamma veto system was confirmed using S35 measurement. More than 50 days of accumulated data was used to analyze the internal background of CARAT#1. External background was rejected by selection of anticoincidence signal between crystal and HPGe detector. The time amplitude analysis and previous event cuts were used to select out α -decay. The measured internal background levels of those CMO crystals are shown in Table 3.

(mBq/kg)	SB28	SB29	S35	CARAT#1
²¹⁰ Po	-	-	57	181
²¹⁴ Po (²³⁸ U)	0.08	-	1.74	2.83
²¹⁶ Po (²³² Th)	0.07	-	0.26	0.04

Table 3. The internal background levels of the CMO crystals.

In case of CARAT#1, the background level of ²¹⁴Po was higher than S35. Otherwise, the background level of ²¹⁶Po was lower than SB28. It means that there was a possibility to be contaminated by ²²⁴Ra during the experiment such as surface contamination by radon, which has about 3 days of half-life, in air.

The selected α events, which energy were much accurate than external α radioactive sources, were used to calculate the α/β ratio of those CMO crystals. The calculated a/b ratio of S35 (a) and CARAT#1 (b) are shown in Fig 4.



Fig 4. The calculated $\alpha \beta$ ratio of S35 (a) and CARAT#1 (b) using measured internal α -decay events.

The α/β ratio differences between S35 and CARAT#1 is less than 10%. We need further study about the enriched CMO crystals, not only for internal background but also characteristics for the future experiment.

5. Conclusions & Discussions

The characteristics of CaMoO₄ scintillation crystals (especially ${}^{40}Ca^{100}MoO_4$ crystal) have been studied to evaluate their applicability for a ${}^{100}Mo 0\nu$ DBD search by the AMoRE collaboration. Isotopically enriched material is a powerful choice not only for 0ν DBD experiment but also any type of rare decay searches in particle physics. The ${}^{48}Ca$ -depleted, ${}^{100}Mo$ -enriched ${}^{40}Ca^{100}MoO_4$ scintillation crystal is a promising detector with low background (from 2ν DBD of ${}^{48}Ca$) and containing a large effective mass of ${}^{100}Mo$. However, further studies are needed, such as improve the scintillation properties (SB series CMO crystal), reduce the internal background (S series CMO crystal), and small difference of a/b ratio between enriched CMO crystal and natural CMO crystal.

Additionally, we have been developing a Metallic Magnetic Calorimeter (MMC), which is one of

cryogenic detector with extremely good energy resolution (required 15 keV of energy resolution on 3 MeV) [5]. The scintillation properties will not be critical issues to cryogenic system. It means that SB series CMO crystals also will be good materials for the future experiment.

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