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Abstract.
Center for Underground Physics (CUP) at Institute for Basic Science (IBS) has been operating the COSINE-100 for WIMP search and the AMoRE (Advanced Mo based Rare process Experiment) for neutrinoless double beta decay search. Both experiments are using ultra-pure scintillating crystals at the underground facility to avoid the cosmic muon background.
In order to grow the ultra-pure crystals, the raw materials are purified and their radioactive levels are measured using ICP-MS and HPGe. A clean-room facility has been constructed to avoid external contaminations during the crystal treatment procedure after the growth.
We present the current status of the crystal growth facility and discuss further about next levels for the ultra-pure crystal production in near future.

1. Introduction

Scintillating crystal detector is a promising technique for a rare process searching experiments. DAMA has been reporting a positive annual modulation signal which might be from weakly interacting massive particle (WIMP) as a strong candidate of dark matter using NaI(Tl) scintillating crystal detectors [1]. There are also number of proposals searching for neutrinoless double beta decays using a scintillating bolometric technique which can provide a high energy resolution and excellent particle identification by simultaneous measurement of heat and light at milli-kelvin temperatures [2, 3]. These experiments require an ultra-pure scintillating crystal to minimize the radioactive background in the detector itself.

CUP has been established since 2013 and is operating two major experiments at an underground facility located in Yangyang(Y2L), Korea. The COSINE-100, an experiment searching for a WIMP using ultra-pure NaI(Tl) crystals which is the same as the DAMA, has been running successfully to confirm or refute the DAMA's result. Recently a physics result using ~106 kg of ultra-pure NaI(Tl) crystals was published [4]. We are planning to increase the crystal mass by more than 100 kg of the same NaI(Tl) crystals in near future. The AMoRE is searching for neutrinoless double beta decays of ^{100}Mo using the scintillating bolometric technique [5]. Recently the pilot stage has finished using ~1.8 kg of $CaMoO_4$ scintillating crystals which is ^{48}Ca depleted to avoid the internal background from $2\nu2\beta$ decay of ^{48}Ca and ^{100}Mo is enriched up to 96% to increase the ^{100}Mo fraction in the crystals. The final stage is planning to use ~200 kg of Mo based scintillating crystals to have ~100 kg of ^{100}Mo.

Both experiments strongly depend on crystal growth conditions which must require highly purified raw materials and be handled under a radioactive-background-free facility. The raw materials are purified well by ourselves as reported in Ref. [6, 7]. Even more, the grown crystals are much more purified than the original raw materials by a segregation effect. They are about to satisfy the requirements for both experiments.

Here, we report the current status of the ultra-pure growths of NaI(Tl) and Mo-based crystals (XMO) at CUP and how the crystals are going to be handled to prevent the extra contaminations during the treatment processes of the grown crystals.

2. Growth of Mo based scintillation crystals for the AMoRE

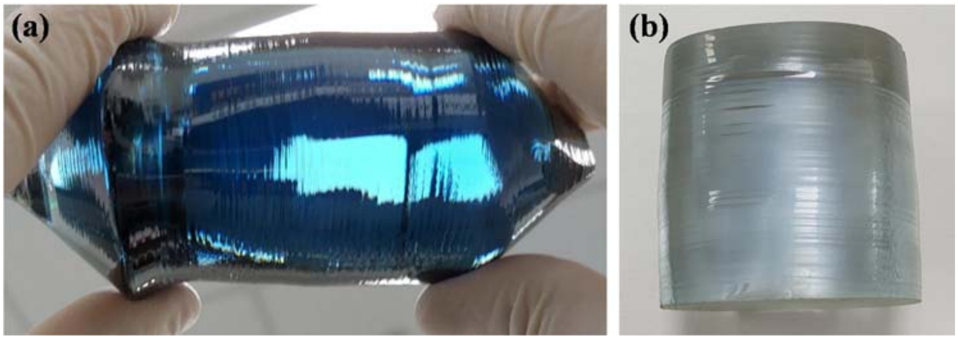

Figure 1. (a) An as-grown CaMoO4 crystal ingot, (b) an annealed CaMoO4 crystal

The XMOs have been grown by a Czochralski method which adopts the inductive heating method to melt the raw material in a metallic crucible. $CaMoO_4$(CMO) and Li_2MoO_4(LMO) are strongly motivated for the AMoRE experiment because of their low internal radioactive background levels. In order to prevent the external contamination during the crystal growth, the growers are installed in the clean facility to be controlled under class 10,000 environment. Each crystal is grown by a dedicated grower to avoid any cross contaminations by each other. The crucibles for each type of crystals are made of different metals because of the different melting points of the raw materials. An iridium crucible has been used for CMO and a platinum for LMO crystals. The growing process can be controlled automatically by a high-resolution load cell which can measure the weight of the crystal ingot during the crystal growth precisely. The crucibles are surrounded by a ceramic thermal insulator which can be an extra contamination source. A proper fused alumina insulator has been confirmed by measuring its radioactivity at the Y2L (Table 1). We are planning to use the high purity alumina for the insulator.

Table 1. HPGe measurement results of refractory materials. Unit : mBq/kg

	Term (days)	Mass (kg)	^{238}U	^{40}K	^{228}Ac	^{228}Th
Fused alumina	5.28	0.43	11	640	10	7
High purity alumina	15.2	0.164	6	44	<5	<4.4

Highly purified raw materials have been used to synthesize with a purified MoO_3 by CUP. We could obtain CMO and LMO ingots successfully in dimensions of Ø 50 mm × H 70 mm which is a proper volume to be used in the AMoRE detector setup [8].

The grown CMO ingot shows a slightly bluish color due to its oxygen deficiency [9] because of high growing temperature (Fig. 1. a). In order to recover the deficiency, the ingots were annealed in a high temperature furnace. CMO and LMO ingots were annealed at 1250 ℃ for a month and at 500 ℃ for 100 hours under the air atmosphere, respectively. Fig. 1 shows the bluish CMO ingot (a) and 1 month annealed one (b).

The ingots were cut using a thin diamond band saw (~ 0.5 mm thickness) with a mineral oil coolant to minimize the stress and roughness on the crystal surface. The surface roughness of crystal must be optimized for the gold film evaporation on the crystal surface to be used as a phonon collector in the

AMoRE detector setup. So the cut surfaces are lapped to remove the kerf and improve the flatness using a grinding machine. Polishing process must be done in a glove box which can control the moisture level below 10 ppm because of the hygroscopic property of LMO crystals and oleic acid and SiC powder mixture have been used as an abrasive during the polishing process to avoid the moisture content. The sizes of applied SiC powders are decreasing (from 3 μm to 1 μm) for a fine surface. In order to make a proper surface for gold film evaporation, a 0.05 μm of colloidal silica is used for the final step of the polishing process.

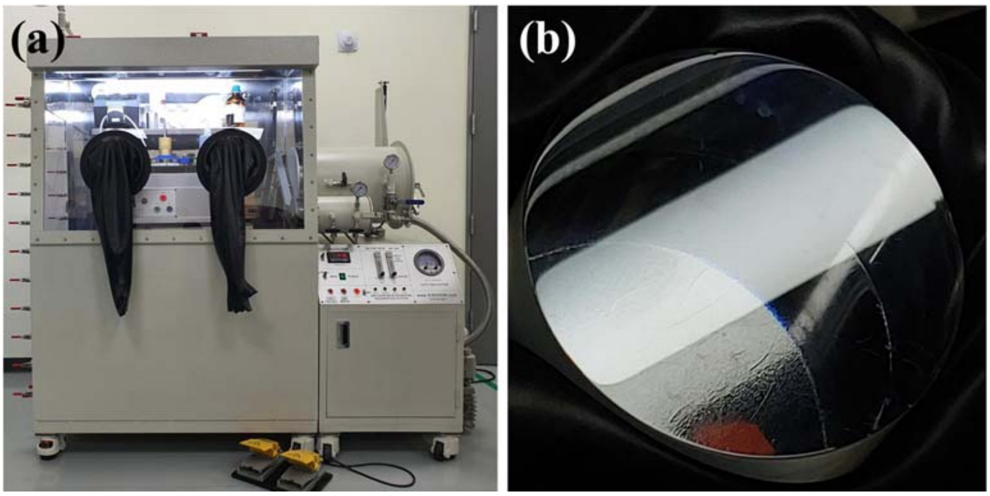

Figure 2. (a) Polishing machine in a glove box, (b) a Li_2MoO_4 crystal's polished surface.

3. Growth of NaI(Tl) crystal for the COSINE experiment

The NaI(Tl) crystals have been grown by a modified Kyropoulos method. Two growers with different volumes are installed at a clean facility, but in a separated place from the Czochralski growers. A ventilation system and a dust collector are installed in the growing room because of the vaporization of NaI and Tl during the growth.

In order to optimize the growing conditions, especially for concentration of Tl, smaller size crystals with dimensions of Ø 60 mm × H 70 mm have been grown using a smaller grower for an R&D study so far [8]. Large size crystals with dimensions of Ø 450 mm × H 250 mm (~115kg) are going to be grown in a bigger grower under the optimized conditions. A commercial quartz crucible was used for the NaI(Tl) crystal growth in the R&D grower, and a synthetic quartz crucible is going to be used for the larger growers because of its smaller contamination. The high purity alumina in Table 1 is also being considered to be used as the thermal insulator for its smaller contamination.

A commercial NaI (Astro grade, Sigma Aldrich) and TlI (99.999 %, Sigma Aldrich) powders are used and Tl concentration is adjusted to be 0.1 mol% for the test growing.

The most important issue for the NaI(Tl) growth is a moisture-free environment because of its strong hygroscopic property. In this reason, we built the annealing furnace placed quite close by the grower to keep the high temperature (~450 ℃) during the transferring. It spends in the furnace under 450 ℃ temperature with air atmosphere for a week. The NaI(Tl) has been treated similarly with the LMO crystal, but we need further considerations for the large volume crystal under moisture-free environment.

4. Summary

The test growth of CMO, LMO, and NaI(Tl) crystals for rare process experiments have been successful by CUP. Highly purified powders were used as the raw materials for low radioactive-background crystals and further purification process is under preparation by ourselves. We also searched clean parts in the grower to minimize the contamination during the crystal growth. The polishing process has been modified for the proper thin gold film evaporation on crystals. Moreover, there were several modifications to avoid the moisture for the hygroscopic crystals (LMO and NaI(Tl)), such as moisture-free glove box and modified annealing furnace. Further improvements are under planning for the ultra-pure scintillating crystals to satisfy both purposes.

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