Development of ultra-pure NaI(Tl) detector for COSINE-200 experiment

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The annual modulation signal observed by the DAMA experiment is a long standing question in the community of dark matter direct detection. This necessitates an independent verification for its origin using the same detection technique. The COSINE-100 experiment has been operating with 106 kg of low-background NaI(Tl) detectors providing interesting checks for the DAMA signal. However, due to higher background in the NaI(Tl) crystals used in COSINE-100 relative to those used for DAMA, it was difficult to reach final conclusions. Since the start of COSINE-100 data taking in 2016, we also have initiated a program to develop ultra-pure NaI(Tl) crystals for COSINE-200, the next phase of the experiment. The program includes efforts of raw powder purification, ultra-pure NaI(Tl) crystal growth, and detector assembly. With extensive research and development of NaI(Tl) crystal growth, we have successfully grown a few Ti-doped crystals with high radio-purity. A high light yield has been achieved with our detector assembly technique. Here we report the ultra-pure NaI(Tl) detector developments at the Institute for Basic Science, Korea. The technique developed here will be applied to the production of ultra-pure NaI(Tl) detectors for the COSINE-200 experiment.

I. INTRODUCTION

Although numerous astronomical observations support the conclusion that most of the matter in the universe is invisible dark matter, an understanding of its nature and interactions remains elusive [1, 2]. Dark matter phenomenon can be explained by new particles, such as weakly interacting massive particles (WIMPs) [3, 4]. Even though tremendous efforts to search for evidence of WIMP dark matter by directly detecting nuclei recoiling from WIMP-nucleus interactions are being pursued, no definitive signal has been observed [5, 6]. One exception is the DAMA experiment that, using an array of NaI(Tl) detectors [7, 8], observes an annual modulation of event rates that can be interpreted as resulting from WIMP-nuclei interactions [9, 10]. However, this result has been the subject of a continuing debate because the WIMP-nucleon cross sections inferred from the DAMA modulation are in conflict with limits from other experiments [11, 12].

An unambiguous verification of the DAMA signal by independent experiments using the same NaI(Tl) crystal target material is mandatory. Experimental efforts by several groups using the same NaI(Tl) target medium are currently underway [23–29]. Even though interesting physics results from COSINE-100 [30, 31] and ANAIS-112 [32] have been reported, none of these efforts have yet achieved a background level similar or lower than that of the DAMA experiment. This large background is a strong impediment to the establishment of any unambiguous conclusion of the DAMA’s observation.

With the COSINE-100 experience, we have identified the crystal growth as the key component for reducing the overall background, of which radioactive 40K and 210Pb contents within a crystal are the most troublesome contributors [28, 33, 34]. An advanced detector design such as an active veto system using liquid scintillator can effectively reduce the impact of 40K impurities [25, 35, 36] but 210Pb has to be eliminated at the crystal production stage.

As part of an effort to upgrade the on-going COSINE-100 experiment for the next phase COSINE-200 experiment, we have conducted extensive R&D on low-background NaI(Tl) crystal growth. An initial chemical purification stage based on a recrystallization method demonstrated sufficient reduction of K and Pb in the powder [37]. A mass production facility for the powder purification has been assembled in our institute [38]. Two custom-made Kyropoulos machines were established at the same site for both the demonstration of low-background crystal growth and the timely production of 200 kg of NaI(Tl) crystals [39]. In the meantime, we developed detector assembly techniques for low-background NaI(Tl) detectors. In this article, our progress in the development of low-background NaI(Tl) detectors and the prospects of the COSINE-200 experiment is described.

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FIG. 1. Two dedicated Kyropoulos growers for the NaI(Tl) crystals. (a) A small grower for low-background NaI(Tl) R&D and (b) a full-size grower for 120 kg size NaI(Tl) crystal production. Both growers have an accompanying setup to facilitate hot crystal annealing.

II. CRYSTAL GROWTH

We have built two dedicated Kyropoulos growers for NaI(Tl) crystals. A small R&D grower is used for proof of principle tests for low-background NaI(Tl) crystal as shown in Fig. 1(a). This grower is specifically designed for fast growth of pilot NaI(Tl) crystals. Two advantages of the fast growth with low impurity contamination were considered in the design: a short period of crystal growth minimizes radon contamination from emanations from the grower’s materials or from the ambient air; fast growth requires special conditions such as a thin interface that enhances the purification of the impurities during crystallization.

For the small grower, we use a 12 cm diameter, 10 cm high quartz crucible that is surrounded by heaters and refractories inside the chamber. The chamber has a cylindrical shape with dimensions of 34 cm diameter and 50 cm height. A same-size annealing machine is included as shown in Fig. 1(a). Figure 2 shows the top view of the growing setup before starting the growth process.

The quartz crucible can contain up to 1.7 kg of the NaI powder and can produce a 1.1 kg crystal ingot (typical dimensions of the ingot: diameter 60–80 mm height 50–80 mm), shown in Fig. 3.

After an extensive experience of crystal growth with the small grower, we successfully produced several thallium doped NaI(Tl) crystals with low-background NaI powder. Based on the success of the small grower, we constructed a full-size grower that can accommodate a 61 cm diameter and 44 cm height quartz crucible for the growth of an approximately 120 kg crystal ingot (see Fig. 1(b)). The full-size grower is similar to that of the smaller one, which minimizes differences in growth conditions.

FIG. 2. A top view of the grower with NaI powder loaded in the middle crucible, with the chamber cupola removed. A hollow-disk-shape quartz lid covers the top of the refractories to prevent fumes from coming out when heated.

FIG. 3. Four NaI(Tl) crystal ingots produced by the small grower : (a) NaI-025, (b) NaI-034, (c) NaI-035, and (d) NaI-036.

All thallium-doped low-background crystals shown in Fig. 3 start with low-background NaI powder from the Sigma-Aldrich (SA) company named “Astro-Grade”. Previous studies show that this grade powder contains low potassium (less than 10 ppb level) and lead (a few ppb level). Typically, NaI powder does not contain significant amounts of uranium and thorium.

When the crystal growth is finished, sample pieces of the crystal ingot are measured with inductively coupled plasma mass spectrometry (ICP-MS) to study impurity contaminations. The first grown crystal, NaI-025, was found to be highly contaminated with both K and Pb, for which the main cause was later identified as originating from fumes produced by the refractories as shown in Table I. Replacing the refractories with one fabricated from relatively radiopure materials resulted in significantly reduced impurities in the fumes. In addition, a quartz cover that blocks the top of the refractories prevented further diffusion of the fumes into the NaI crucible. Subsequently grown crystals, NaI-034, NaI-035, and NaI-036, had significantly reduced K and Pb contaminations, as can be seen in Table I.

We tested different types and amounts of thallium iodide (TlI) from different vendors as the dopant in the NaI(Tl) crystals. Initially, we used 99.999% purity TlI powder from SA. However, this powder created black dust in the melt that were traced to carbon impurities in the powder. This dust makes it difficult to optimize crystal growth conditions and the produced ingots contained black speckles throughout their entire bulk. We
TABLE I. ICP-MS results for: initial materials; fumes produced during the growing process; and the grown crystals.

| Sample                  | K (ppb) | Pb (ppb) | U (ppt) | Th (ppt) | Tl (mol %) |
|-------------------------|---------|----------|---------|----------|------------|
| NaI (Astro-grade)       | 5       | 1.3      | <10     | <10      | –          |
| TII (SA powder)         | 3400    | 500      | <10000  | <10000   | –          |
| TII (SA bead)           | 150     | 100      | <10000  | <10000   | –          |
| TII (AA powder)         | 2100    | 600      | 10000   | <10000   | –          |
| TII (AA bead)           | 4400    | 600      | 20000   | <10000   | –          |
| Fume with old refractories | 1021235 | 10407    | <10     | <10      | –          |
| Fume with new refractories | 320    | 25       | <10     | <10      | –          |
| NaI-025                 | 740     | 170      | <10     | <10      | 0.024      |
| NaI-034                 | 8       | 0.4      | <10     | <10      | 0.039      |
| NaI-035                 | 13      | 2        | <10     | <10      | 0.036      |
| NaI-036                 | 23      | 10       | <10     | <10      | 0.197      |

speculate that the carbon contamination in the TII powder may have been caused by plastic or PTFE material used in the chemical purification process. After a variety of tests for different types of TII, we found that the bead type TII does not produce any significant black dust either the melt or the ingot. To confirm the reproducibility of this, we also tested TII from Alpha-Aesar (AA) in both powder and bead forms and concluded that the bead type contains less carbon. Contamination levels of radioisotopes for various TII samples were measured with ICP-MS (see Table I).

Additionally, since the NaI melt directly touches the crucibles, we checked the impact of the crucible type by exchanging the quartz crucible with a platinum one and found no noticeable dependence on the crucible type. Specifications of the crystal growth conditions for various NaI(Tl) crystals grown in the laboratory are summarized in Table II.

III. DETECTOR ASSEMBLY

Once the crystal ingots are prepared, samples from the top and bottom sections are cut by a bandsaw for ICP-MS measurements. The remaining bulk of the crystal is machined to a cylindrical shape using a lathe in a low-humidity glovebox shown in Fig. 4. When shaping the crystal, mineral oil is continuously poured on it to prevent cracks from developing and suppress NaI(Tl) dust in the environment. Four separate ingots were assembled as detectors after the crystal surfaces were polished. For NaI-025 and NaI-036, we did not machine the barrel surfaces, while NaI-034 and NaI-035 were machined into cylindrical forms. Studies show the cylindrical shape improves light collection while not affecting the background contamination level.

The crystal is dry-polished using aluminum oxide lapping films ranging from 400 to 8,000 grits. After the polishing, several layers of soft PTFE sheets are wrapped on the barrel surface as a diffusive reflector and an optical interface (silicon rubber EJ-560) is coupled to the top and bottom surfaces. A 5 mm-thick copper casing with quartz windows at both ends is used to encapsulate the crystal airtight as shown in Fig. 4. Hamamatsu 3-inch PMTs (R12669SEL) are coupled via an optical interface to each quartz window. The PMTs are fixed with PTFE guard rings using stainless steel bolts.

All assembly procedures are performed inside a low-humidity glovebox that maintains the humidity level within a few tens of ppm (H₂O) using N₂ gas and a molecular sieve trap. All assembly parts are cleaned using diluted Citranox liquid with sonication, and baked in an oven before moving into the glovebox. Several weeks after the assembly is finished, a visual inspection shows no degradation of the crystal in terms of its transparency. For evaluation, we used an underground low-background setup for further detailed measurements.

IV. BACKGROUND MEASUREMENTS

A. The experimental setup

To evaluate the background contamination levels in the NaI(Tl) crystals, we used the same experimental apparatus that was used for previous NaI(Tl) crystal R&D at the Yangyang Underground Laboratory (Y2L) [23] [33].
TABLE II. Growth conditions of each crystal ingot. Here SA and AA indicate Sigma-Adrich and Alpha-Aesar, respectively.

| Crystal | Growth date | Quartz cover | Refractory | Crucible | TlI amount loaded | TlI powder |
|---------|-------------|--------------|------------|----------|-------------------|------------|
| NaI-025 | May/24/2018 | X old Quartz | 0.1 mol % | SA powder |
| NaI-034 | Sep/18/2019 | O new Platinum | 0.1 mol % | SA powder |
| NaI-035 | Nov/12/2019 | O new Quartz | 0.12 mol % | AA bead |
| NaI-036 | Feb/4/2020 | O new Quartz | 1 mol % | SA bead |

This includes a 12-module array of CsI(Tl) crystals inside a shield that includes 10 cm of copper, 5 cm of polyethylene, 15 cm of lead, and 30 cm of liquid-scintillator-loaded mineral oil [41, 42]. The shielding stops external neutrons and gamma rays, and vetoes cosmic-ray muons. The availability of this already-established setup has been advantageous for a timely evaluation of new crystals. The NaI(Tl) crystals have been tested inside the CsI(Tl) detector array with an arrangement indicated schematically in Fig. 5.

B. Light yields

High light yield of the NaI(Tl) crystal is crucial for the COSINE-200 experiment because it is possible to lower the experimental threshold to 1 keV and thereby match the DAMA experiment’s value [8]. The doping concentration of thallium (Tl) is one of the most important factors in the light yield as described in Refs. [43, 44]. We originally targeted a doping level of 0.1 mol% for optimal light yield based on the above-noted references. To accomplish this, we applied approximately 0.1 mol% TlI powder into the initial mix for the NaI-025, NaI-034, and NaI-035 crystals. However, the measured doping levels in the final produced crystals were significantly reduced to 0.02−0.04 mol% (Table I). This was mainly due to the evaporation of a large fraction of the thallium during the high-temperature growth period. We measure relatively low light yields from these crystals; between 9 and 12 photoelectrons (NPEs) per keV. Because of light absorption by the black impurities inside the crystals, slightly low light yields using TlI powder (NaI-025 and NaI-034) were observed (see Table IV).

On the other hand, for NaI-036, 1 mol% of bead-type TlI from SA was added to the initial mix and a 0.2 mol% Tl concentration was measured in the ingot after growth. In this crystal, we obtain the highest light yield of 17.1±0.5 NPE/keV. Note that the COSINE-100 crystals and DAMA crystals have light yields of approximately 15 NPE/keV and 5−10 NPE/keV, respectively,
and are able to operate with a 1 keV threshold. The light yield from NaI-036 is sufficient for the COSINE-200 experiment. Figure 7 shows the light output comparison between NaI-025 and NaI-036 as determined from $^{241}$Am calibration data. The NaI-036 crystal’s larger light output is also evident in the improved energy resolution.

C. Internal natural backgrounds

To produce ultra low-background NaI(Tl) crystals, one should understand internal contamination of natural radioisotopes. Contamination levels for various radioisotopes are described for each of the four crystals below.

1. $^{40}$K background

One of the most dangerous sources of background for WIMP searches with NaI(Tl) crystals is contamination from $^{40}$K. Its natural abundance is roughly 0.012% of the total amount of potassium ($^{nat}$K). About 10% of $^{40}$K decays produce a 1460 keV $\gamma$-ray in coincidence with a 3.2 keV X-ray. If the 1460 keV $\gamma$-ray escapes the crystal and only the 3.2 keV X-ray is detected, an event is tagged by the mean time of their signals, defined as,

$$<t> = \sum_i A_t t_i A_i.$$

Here $A_t$ and $t_i$ are the charge and time of each digitized bin of a recorded event waveform. Figure 8(a) shows a scatter plot of the energy versus the mean time for the NaI-036 crystal. Alpha-induced events are clearly separated from $\beta$ or $\gamma$-induced events because of their faster decay times. Selected $\alpha$ events (red points) are used to understand the internal contamination of heavy elements such as $^{235}$U, $^{232}$Th, and $^{210}$Pb. We also analyzed the energy spectra of these $\alpha$ events (see Fig. 9(b)) in order to understand the internal contamination. Our understanding of the alpha contamination is summarized in Table III.

2. $^{210}$Pb background

In the COSINE-100 experiment, the main source of the alpha contamination was due to decays of $^{210}$Po ($E_\alpha =$
5.4 MeV) nuclei that probably originated from a $^{222}$Rn contamination in NaI-036, we observe similar levels of $^{222}$Rn exposure occurs, it will grow with a characteristic time of $\tau_{210}$, which is determined using a 210Pb level. Thus, the total alpha rate over time can provide information about the date of the $^{222}$Rn exposure and the amount of the $^{210}$Pb contamination. We can determine the total number of alphas from the mean time distribution (Fig. 9). Figure 10 shows the total alpha rates in the crystals as a function of data-taking time. In NaI-025, one can see that the alpha rate from $^{210}$Po increases. The $^{210}$Po level in NaI-025 is $\pm 0.26$ mBq/kg, which is consistent with the 46.5 keV $\gamma$ peak measurement. However, we only have upper limits for NaI-034 & NaI-035 because there are no observed alphas from $^{210}$Po. Due to an $^{220}$Ra contamination in NaI-036, we observe similar levels of $^{210}$Pb and $^{226}$Ra contamination at the beginning of the crystal growth period.

4. $^{232}$Th background

Contaminations from the $^{232}$Th decay chain can be identified by studying three-alpha delayed time coincidences of $^{224}$Ra, $^{220}$Rn and $^{216}$Po, where the half-lives are 3.66 day, 55.6 s, and 0.145 s, respectively. Due to the short half-life of $^{216}$Po ($t_{1/2}=0.145$ s), identification of two $\alpha$ particles from $^{220}$Rn and $^{216}$Po is relatively straightforward without any background as can be seen in Fig. 11(a). With the selection of two consecutive $\alpha$ signals, $^{226}$Ra can also be identified. No significant $\alpha$ events from $^{232}$Th in NaI-025 and NaI-036 are measured. However, $\alpha$-$\alpha$ delayed coincidence events from NaI-034 and NaI-035 are identified (see Fig. 11(a) for NaI-034) with measured activity levels of $35 \pm 4 \mu$ Bq/kg and $7 \pm 2 \mu$ Bq/kg for NaI-034 and NaI-035, respectively. Assuming equilibrium of the $^{232}$Th chain, this is similar to $2.5 \pm 0.8 \mu$Bq/kg of the COSINE-100 C6 and 2–31 $\mu$Bq/kg level in the DAMA crystals.

5. $^{238}$U background

$^{238}$U is one of the most common radioisotopes in nature primarily because of its long decay time. A study of its decay products allows us to understand its contamination level. Delayed Coincidence $\alpha$-$\alpha$ events with $t_{1/2} = 3.10$ min time difference from $^{222}$Rn-$^{218}$Po can be used to infer the $^{238}$U contamination levels shown in Fig. 11(b). The measured rate for $^{224}$Ra-$^{220}$Rn ($t_{1/2}=55.6$ s) is extracted from the $^{232}$Th level, which is determined using $^{220}$Rn-$^{216}$Po decay. In this way, we infer $^{218}$Po levels of $64 \pm 11 \mu$ Bq/kg and $8 \pm 6 \mu$ Bq/kg for NaI-034 and NaI-035, respectively. Because of the high total $\alpha$ rates in NaI-025 and NaI-036, we cannot measure 3 min half-life $\alpha$-$\alpha$ coincidence events.

Another method for studying the $^{238}$U chain exploits the 164.3 $\mu$s half-life of $^{214}$Po $\alpha$-decay, which follows its production via the $\beta$-decay of $^{214}$Bi. Due to the 200 $\mu$s dead time of the trigger system, only events in the higher end of the exponentially falling distribution are acquired but the number of these events is sufficient to measure the $^{238}$U contamination (see in Fig. 11(c)). The measured activity calculation includes a correction for the effect of the inefficiency caused by the 200 $\mu$s dead time. NaI-036 shows a relatively large $^{210}$Po contamination of $451 \pm 48 \mu$ Bq/kg, while NaI-025, NaI-034, and NaI-036 show contaminations of $26 \pm 7 \mu$ Bq/kg, $46 \pm 7 \mu$ Bq/kg, and $13 \pm 6 \mu$ Bq/kg, respectively. The contamination in NaI-036 is considerably larger than those of the DAMA crystals (8.7–124 $\mu$Bq/kg) but the other crystal contamination levels are similar to those of DAMA.

The energy spectrum of $\alpha$ events in Fig. 11(b) spec-
a 0.45 mBq/kg contamination of $^{210}$Pb contamination significantly. The total alpha rate is relatively stable and is consistent with $^{210}$Po decay events from the $^{210}$Pb contamination coming from $^{226}$Ra decays.

especially, the SA powder (initial loading 0.1 mol% of NaI-034) contribute consistent contaminations of 0.8 mBq/kg of the $^{210}$Pb contaminations present in NaI-034 or NaI-035 during the crystal growth. (d) Due to $^{226}$Ra contamination in NaI-036, a relatively large number of alpha events is observed. The total alpha rate is relatively stable and is consistent with a 0.45 mBq/kg contamination of $^{210}$Pb originating from $^{226}$Ra decays.

If this is the case, we expect that the use of TlI crystals. If this is the case, we expect that the use of TlI powder can be explained by impurities contained in the TlI powder. Especially, the SA powder (initial loading 0.1 mol% of NaI-034) and SA beads (initial loading 1 mol% of NaI-036) contribute consistent contaminations of $^{226}$Ra to the crystals. If this is the case, we expect that the use of TlI beads from Alpha Aser (NaI-035) could reduce the $^{226}$Ra contamination significantly.

6. Summary of internal background

Table XIV shows a summary of the internal background measurements compared with crystal C6 of COSINE-100 and the DAMA crystals. The $\alpha - \alpha$ coincidence measurements from $^{220}$Rn $\rightarrow$ $^{216}$Ra are used to establish the $^{232}$Th contamination level with the assumption of chain equilibrium. The $^{226}$Ra measurements are average values of $\alpha - \alpha$ and $\beta - \alpha$ studies in this chain. Due to the small size of the tested crystals and the brief, one-month long measurement period, there are large uncertainties in the $^{40}$K measurements. ICP-MS measurements provide better precision and show consistent results.

D. External Background

In the COSINE-100 experiment, the external background contributions in the region of interest (ROI) are negligible because of the active veto from the 2,200 L liquid scintillator. However, the R&D setup contains a significant level of external background that is more than 10 times for the same size crystal. Because the PMTs are the main contributor to the external background, a small crystal will have relatively much large background from external sources. Since we use the same type of PMTs in the COSINE-100 experiment, we can estimate the external background contribution using the...
 GEANT4 simulation based on previous studies [34, 45].

E. Cosmogenic radionuclides

The cosmogenic production of radioactive isotopes in an NaI(Tl) crystal contributes a non-negligible amount of background in the signal region of the COSINE-100 experiment [34, 49]. This is mainly due to long-lived nuclides such as $^3$H and $^{22}$Na. All crystals of the COSINE-100 experiment were produced at the Alpha Spectra company located at Grand Junction, Colorado, USA. Due to its high altitude of about 1,400 m above sea level as well as the long delivery time to Korea, the crystals were exposed to a significant flux of cosmic-ray muons and their spallation products and, as a result, we measure a relatively high level of cosmogenic radioactive isotopes. However, the crystals that are studied here are all grown in Daejeon, Korea (70 m in altitude) and delivered underground in a short time period (typically within a few weeks). Based on our previous study in Ref. [49], one month exposure time near sea level can produce 0.004 mBq/kg of $^3$H and 0.05 mBq/kg of $^{22}$Na. These activities are an order of magnitude lower than measured activities in COSINE-100 C6, which are 0.11 mBq/kg and 0.73 mBq/kg [34] of $^3$H and $^{22}$Na, respectively. Thus, we expect that the background contributions from these long-lived cosmogenic radioisotopes are sufficiently low for the COSINE-200 experiment.

V. BACKGROUND MODELING AND PROSPECTS OF COSINE-200

For a quantitative understanding of background in these crystals recently grown, we use the GEANT4-based simulation that was developed for the background modeling of the COSINE-100 NaI(Tl) crystals [34, 45]. We take values of the contamination levels discussed in Section IV as input values. Because to the low statistics of the underground $^{40}$K measurements, we use the ICP-MS results for $^{40}$K from Table I.

We use a log-likelihood method to fit the data. The fitting range is 3 keV–3 MeV and we perform a simultaneous fit of four-channels: single-hit low-energy (3–60 keV) and high energy (60 keV–3 MeV), multiple-hit low-energy and high energy. Here, multiple hit means that there is one or more coincident hits recorded in any of the surrounding CsI(Tl) crystals. The internal backgrounds are constrained to be within 20% of their measured values. We do not consider surface $^{210}$Pb components in the crystals because the crystal surface is polished in a nitrogen gas environment and we do not see any indication of increases in energy spectra in the low-energy region that are characteristic of surface $^{210}$Pb contamination [50]. The PMT background is constrained to be within 50% of the measured values from previous studies [34, 45]. The long-lived cosmogenic radioisotopes are constrained to be within 50% of its maximum production values that are estimated in Section IV-E while other short lived cosmogenic components are freely floated. Additional external backgrounds from the CsI(Tl) PMTs and copper surface are also included as free parameters but the final results are cross-checked with the previous study in the same setup [45].

Figure 12 shows the fitted results for NaI-035 on all of the simulated background components plotted as lines with different colors. The overall energy spectra are well matched to the data for both single-hit events and multi-hit events. The small size of the crystal as well as the non-existence of an active veto detector in the R&D setup results in the external background being the dominant contributor to these spectra. However, this contribution will be significantly reduced with the large, COSINE-100-sized, crystals and the liquid scintillator veto system that will be used in the upcoming COSINE-200 setup.

To estimate the expected background in the COSINE-200 crystal, we generate simulated background spectra assuming the same internal and cosmogenic radioisotopes internal to the crystals with NaI-035, but the size of the crystal is enlarged to that of C6 of COSINE-100 (12.5 kg) and installed inside a COSINE-100-like shield. The COSINE-100 shield consists with 2,200 L liquid scintillator, 3 cm thick copper, 20 cm thick lead, and an array of plastic counters for muon tagging [51] as described in Refs. [25, 52]. We, therefore, expect levels of external background that are similar to those seen in the COSINE-100 detector [34]. Figure 13(a) shows the expected background spectrum for the COSINE-200 detector assuming the internal background as NaI-035. By taking advantage of the 2,200 L active veto detector as well as the en-

TABLE IV. Measured results of the internal contaminations from this study compared with C6 of COSINE-100 [34] and DAMA crystal results [17, 38]. Here, the total alphas are calculated for the case of the saturated $^{210}$Po decays.

| Crystal       | Mass (kg) | $^{40}$K (ppb) | $^{210}$Pb (mBq/kg) | $^{226}$Ra (mBq/kg) | $^{232}$Th (mBq/kg) | LY (NPE/keV) |
|---------------|-----------|----------------|---------------------|---------------------|---------------------|--------------|
| NaI-025       | 0.65      | 684±29         | 3.8±0.3             | 26±7                | <6                  | 10.4±1.56    |
| NaI-034       | 0.67      | <62            | 0.05±0.02           | 51±7                | 35±4                | 9.5±1.12     |
| NaI-035       | 0.61      | <42            | <0.01               | 11±4                | 7±2                 | 11.8±1.78    |
| NaI-036       | 0.78      | 44±20          | 0.42±0.27           | 45±48               | <20                 | 17.1±0.54    |
| C6 (COSINE-100)| 12.5      | 17±3           | 1.87±0.09           | <0.25               | 2.5±0.8             | 14.6±1.5     |
| DAMA          | 9.7       | <20            | 0.01–0.03           | 8.7–124             | 2–31                | 5–10         |

Table contents:
- Crystal: NaI-025, NaI-034, NaI-035, NaI-036, C6 (COSINE-100), DAMA
- Mass (kg): 0.65, 0.67, 0.61, 0.78, 12.5, 9.7
- $^{40}$K (ppb): 684±29, <62, <42, 44±20, 17±3, <20
- $^{210}$Pb (mBq/kg): 3.8±0.3, 0.05±0.02, <0.01, 0.42±0.27, 1.87±0.09, 0.01–0.03
- $^{226}$Ra (mBq/kg): 26±7, 51±7, <1, 45±48, <0.25, 8.7–124
- $^{232}$Th (mBq/kg): <6, 35±4, 7±2, <20, <0.25, 2–31
- LY (NPE/keV): 10.4±1.56, 9.5±1.12, 11.8±1.78, 17.1±0.54, 14.6±1.5, 5–10
FIG. 12. Measured single-hit and multiple-hit background spectra of NaI-035 (black points) are fitted with all simulated background using a simultaneous fit of four-channels. The external component (blue-dashed filled) is the dominant contributor but this will be reduced with full-size detectors situated in the liquid scintillator veto system.

FIG. 13. (a) The expected background spectrum of a COSINE-200 crystal for low-energy single-hit events assuming the same internal and cosmogenic radioisotopes as NaI-035 but, installed in the COSINE-100 shield with an enlarged crystal size (12.5 kg). (b) The time evolution of background level due to decays of short lived cosmogenic radioisotopes after 0 days (blue-dashed-dotted line), 3 months (red-dotted line), 1 year (purple-dashed line), and 2 years (black-solid line). (c) The expected background spectra of the COSINE-200 detectors assuming background levels obtained from NaI-034 (blue-dashed-dotted line), NaI-035 (black-solid line), and NaI-036 (red-dotted line) after being underground for two years.
larged detector size, a significantly reduced background in low-energy single-hit events is expected. In the region of interest between 1 keV and 6 keV, the expected background level is approximately 0.34 counts/kg/day/keV and below that in the DAMA experiment. As time goes on at underground, short-lived cosmogenic radioisotopes will decay away as shown in Fig. 13(b). After being placed at underground for two years, the background is expected to decrease to about 0.14 counts/kg/day/keV. If we consider different background levels obtained from NaI-034 and NaI-036, the expected background from the COSINE-200 detectors would be slightly increased as can be seen in Fig. 13(c). However, it is still much less than 1 counts/kg/day/keV that is target background for the COSINE-200 experiment [33]. Using these low background crystals, COSINE-200 can test the annual modulation signal observed by DAMA without any of the ambiguities that qualify previous studies [25, 33].

VI. CONCLUSION

In the quest for dark matter direct detection, the origin of the DAMA modulation signal remains the subject of a long-standing debate. To expand the currently ongoing COSINE-100 experiment and reach a model-independent conclusion, we propose the COSINE-200 experiment with 200 kg ultra-pure NaI(Tl) crystals. As a part of this in a program to reduce background levels, we have grown four NaI(Tl) crystals in the laboratory. In this article, we present results from performance studies of these R&D devices that are suitable for the COSINE-200 experiment in terms of both background and light yield. Assuming similar quality full-size crystals installed in the COSINE-100-like shield, our study shows an expected background level reaches to 0.34 counts/kg/day/keV, which is much lower than the background levels of the DAMA crystals. With these high quality crystals, COSINE-200 will be able to produce a definitive and unambiguous test of the DAMA experiment’s annual modulation signal.

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