PICOLON dark matter search project by large-volume NaI(Tl) scintillator

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PICOLON (Pure Inorganic Crystal Observatory for LOw-energy Neutr(al)ino) project is developing high-purity NaI(Tl) crystals to search for WIMPs (dark matter). The XENON experiment has excluded a large region of WIMPs. However, the annual modulation of the signal reported by the DAMA/LIBRA group has not been rejected using the same target, NaI(Tl). Recently, COSINE-100 reported an annually modulating signal of the opposite phase from the data taken using a 106 kg NaI(Tl) DM detector. We plan to build a radiopure large-volume NaI(Tl) detectors to resolve the inconsistency amoung the existing observations. We have developed and confirmed methods to remove radioactive impurities in NaI(Tl) crystals. The concentration of natural potassium is less than 20 ppb, and the U, Th series are less than a few ppt. Using large detectors effectively reduces background events originating from outside of the crystal. The possibility of background reduction and prospects for cosmic dark matter searches with large NaI(Tl) crystals using ultra-high purification techniques will be presented.

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1. Introduction

The search for cosmic dark matter is one of the most essential tasks in particle physics, cosmic ray physics, and astrophysics. Many groups worldwide are working hard to develop high-sensitive detectors for dark matter search. The XENON group is currently the most sensitive search experiment using a two-phase liquid xenon detector. The XENONnT experiment uses a large volume of 5.9 tons of high-purity liquid xenon. They developed a time-projection chamber (TPC) to precisely detect events' locations and eliminate the background from external sources. As a result, the world's lowest background of 4.3×10^{-4} $\mathrm{keV_{ee}^{-1}day^{-1}kg^{-1}}$ for electron scattering events was achieved [1]. However, no event that could be considered cosmic dark matter has been found in the experiment so far.

The search for cosmic dark matter with NaI(Tl) detectors needs to be clarified. Since 1998, the DAMA/LIBRA group has been conducting cosmic dark matter search experiments using a 250 kg NaI(Tl) detector and has reported annual modulation between 2 $\mathrm{keV_{ee}}$ and 6 $\mathrm{keV_{ee}}$ [2], where $\mathrm{keV_{ee}}$ is the energy scale calibrated by electron energy. They used the purest NaI(Tl) crystals and reported a background of 1 $\mathrm{keV_{ee}^{-1}day^{-1}kg^{-1}}$. No group has yet performed experiments with lower background levels than the DAMA/LIBRA group using the NaI(Tl) detector. Experiments with other nuclei have contradicted the results of the DAMA/LIBRA group, but the annual modulation results have yet to be available. The possibility that this is an essential physical phenomenon cannot be ruled out since the results are not due to the detector itself or environmental factors in the surrounding environment. Therefore, it is essential and urgent to carry out the verification experiment using the NaI(Tl) detector with better sensitivity.

The COSINE-100 group reported an annual modulation in the background count rate [3]. They reported that an annual modulation was observed in the low-energy region of the NaI(Tl) detector, but its phase is opposite to that of the DAMA/LIBRA results. This confusion is due to the high background count rate of NaI(Tl), and if low-background measurements are made in this study, everything will be resolved, and the search for cosmic dark matter will become more attractive.

We propose a high-precision verification experiment and a promising cosmic dark matter search using a giant NaI(Tl) crystal. The purity of NaI(Tl) crystals has been maintained at the world's highest level through previous research [4]. By increasing the size, we aim to reduce the background caused by external factors and achieve a low background equivalent to that of the Xenon group. In the following chapters, we will explain the verification of NaI(Tl) enlargement and the effect of background reduction by simulation.

2. Purification of NaI(Tl) crystal and material selection

The XENON group achieved the lowest background level reported at 10^{-4} $\mathrm{keV_{ee}^{-1}day^{-1}kg^{-1}}$[1]. The background level using NaI(Tl) is worse than four orders. We proceed following tasks to obtain the low background for sufficient sensitivity.

- Purification of the target crystal.
- Purification of the surrounding materials.
- Eent selection to remove the background events.

The primary origin of background events is radioactive impurities (RIs) in NaI(Tl) crystal. Producing a high-purity crystal is one of the most critical tasks in searching for dark matter with sufficient sensitivity. We successfully reduced the concentration of RIs by optimizing two purification methods; re-crystallization and resin [4]. The re-crystallization is effective for water-soluble RIs, for example, ^{40}K. We prepared a saturated water solution of NaI at the temperature of 100 °C. We get pure NaI sediment to cool the saturated solution to room temperature slowly. We expected the iteration of re-crystallization to get a purer deposit. However, the concentration of ^{40}K was not reduced expectedly for more than three times iterations. We decided to perform the re-crystallization twice.

The water-insoluble impurities, for example, ^{210}Pb and many progenies of uranium and thorium series, should be removed by optimized resins. We selected the best resins by measuring the reduction factor of lead ions in NaI water solution. The reduction factor was calculated by the concentration of lead ions before and after the purification process. The purification by the selected resin was performed after re-crystallization.

Table 1: The concentration of RIs in present NaI(Tl) crystals by the groups using NaI(Tl) [5–7].

Conc. [μBq/kg]	DAMA/LIBRA	COSINE	ANAIS	PICOLON
^{40}K	< 600	515 ~ 1900	540 ~ 1200	< 480
^{232}Th	2 ~ 31	2.5 ~ 35	0.4 ~ 4	4.6 ± 1.2
^{226}Ra	8.7 ~ 124	11 ~ 451	2.7 ~ 10	8.7 ± 1.5
^{210}Pb	5 ~ 30	50 ~ 3800	740 ~ 3150	28 ± 5

We measured the concentration of RIs after making a NaI(Tl) detector. We produced a cylindrical NaI(Tl) crystal with a dimension of 7.62 cm in diameter and 7.62 cm in length. The low-background measurement was performed in the Kamioka underground laboratory, 2700 meters of water equivalent. We accumulated the data for one month and obtained the alpha-ray energy spectrum. The concentrations of RIs in the present NaI(Tl) are listed in Table 1.

We should take care of the RIs contained in surrounding materials. We measured the concentration of RIs in the nearest materials, such as the reflector, housing, and light guide. Their contamination was measured before we used them[4]. The contribution from the nearest materials is sufficiently low.

On the other hand, the RIs in the PMT circuit are significantly high to perform the low background measurement below 1 $\mathrm{keV_{ee}^{-1}day^{-1}kg^{-1}}$. We selected resistors and capacitors by measuring their RIs. We will apply the selected parts for PMT circuits.

3. Large-volume NaI(Tl) crystals for dark matter search

Present NaI(Tl) detector system for dark matter search consists of many modules of the order of 10 kg crystal mass. With this method, even if the purity of the crystal is high, the background due to surrounding materials makes it difficult to conduct experiments with a sufficiently low background. The main origins of background events are photomultiplier tubes (PMTs), housing, and light guides. The complex of small modules consists of many dirty surrounding materials around the pure NaI(Tl) crystals.

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XENONnT successfully reduced the background caused by the central region of the detector to the utmost limit by making a large single detector module. They used time-projection chamber (TPC) technology to locate the background events and remove the event from the surface of the detector. The outer region of the liquid xenon acts as an active shield against the events comes from the surrounding materials [1].

We propose a large-volume NaI(Tl) crystal with a mass of 360 kg: it is cylindrically shaped with a diameter of 50 cm and length of 50 cm. The cylindrical NaI(Tl) crystal was covered with smaller pieces of NaI(Tl) crystals, which act as active shields.

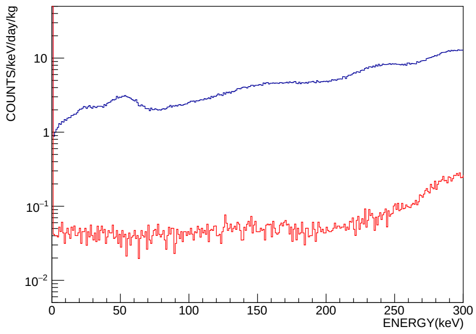

Figure 1: Blue: Background spectrum without veto. Red: Background with veto.

The background events were generated at the assembly circuit of the PMT. The gamma rays from the parts of a PMT circuit are reduced by event selection with position analysis. The position information is derived from the ratio of corrected photons by PMTs. The ratio of scintillation photons measured by PMTs depends on the position of the scintillation source. We distinguish the events from PMTs since outer modules absorb the gamma rays from the out of the detector. The effectiveness of the background reduction was estimated by Monte Carlo simulation, Geant4.10.

The simulated isotopes were ^{214}Pb, ^{214}Bi, ^{210}Pb, ^{210}Bi, and ^{208}Tl, which are the serious background origins from outside of the detector. The simulated energy spectra with and without veto by the outer modules are shown in Figure 1. In the simulation, we used the measured contamination values in the parts of a circuit. The blue energy spectrum is the observed events without any selection. The red one is the selected events observed only in the central crystal. We found about twenty times reduction in the background events.

4. Discussion

We have developed highly radiopure NaI(Tl) crystals to search for dark matter. While the current NaI(Tl) is sufficiently pure, it is insufficiently shielded against externally caused backgrounds. In this project, we propose to cover the perimeter of a giant NaI(Tl) crystal with an active shield. This method eliminates the external background and enables high-sensitivity measurements comparable to the XENON experiment.

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