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Study of high-purity NaI(Tl) crystals using the PICOLON purification method

K. Kotera[a,*] **and PICOLON Collaboration**

[a] *Graduate School of Sciences and Technology for Innovation, Tokushima University, 2-1 Minami Josanjimacho, Tokushima, Tokushima, 770-8506, Japan*

E-mail: c612344002@tokushima-u.ac.jp

Dark Matter Detection is an important issue in both cosmology and particle physics. WIMPs (Weakly Interacting Massive Particles) are one of the most promising candidates for dark matter. The XENON group has the most sensitive detector in the world, however, they reported no significant signal. On the other hand, the DAMA/LIBRA group reports the annual modulation by NaI(Tl), however, their sensitivity is less than the one reported by XENON group. From the perspective of the scientific control, the verification of the annual modulation of the DAMA/LIBRA group is essential to use NaI(Tl) crystals with backgrounds comparable to those of them. PICOLON (Pure Inorganic Crystal Observatory for Low-energy Neut(ra)lino) aims to search for dark matter using ultra-pure NaI(Tl) crystals and to verify the annual modulation reported by the DAMA/LIBRA group. We report the background of a new PICOLON crystal (Ingot #94) developed using the Ingot #85 purification method.

*Speaker

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1. Status of dark matter search

Dark matter is a sort of particle which cannot be observed by the optical method and accounts for 26.4% in the total energy density of the universe. The WIMPs (Weakly Interacting Massive Particles) are one of the most promising candidates in the dark matter. Detection of WIMPs requires a detector sensitivity with WIMP-nucleon SI (spin-independent) cross section of 10^{-40} cm^2 at 50 GeV/c^2 WIMPs mass. However, the DAMA/LIBRA experiment reported an annual modulation signal of 13.7σ in the SI cross section $\sigma \sim O(10^{-42})$ cm^2 for 50–80 GeV WIMPs mass on I nucleus, $\sigma \sim O(10^{-40})$ cm^2 for 10–20 GeV WIMPs mass on Na nucleus [1].

ANAIS, COSINE, and SABRE are trying to verify the result of the DAMA/LIBRA experiment using NaI(Tl) detectors. A result of ANAIS experiment reports no annual modulation, while the COSINE experiment claims negative annual modulation using the DAMA/LIBRA-like analysis method [2][3].

PICOLON (Pure Inorganic Crystal Observatory for LOw-energy Neutr(al)ino) aims to research dark matter using ultra high-purity NaI(Tl) detectors and to verify the annual modulation. The high-purity NaI(Tl) crystals are essential to verify the annual modulation by the DAMA/LIBRA experiment. We developed a previous ultra high-purity NaI(Tl) crystal named Ingot #85 in 2020 with a background comparable to that of DAMA/LIBRA [4]. We report the analyzed background for 2–6 keV$_{ee}$ in the Ingot #94 using the Ingot #85 purification method. keV$_{ee}$ is the energy scale calibrated by electron equivalent energy.

2. Experimental setup and background analysis

Ingot #94 detector installed in Kamland site in Kamioka Underground Laboratory (2700 meter water equivalent), Hida-city, Gifu, Japan. The Ingot #94 shield consists of 5 cm thick oxygen-free copper blocks and 15 cm thick low-activity lead blocks. Pure nitorogen gas is supplied to purge radon. The NaI(Tl) crystal of detector has a mass of 1.344 kg, a diameter of about 76.2 mm, and a length of about 76.2 mm. Signal waveform is record by 8-bit Flash ADC in MoGURA electric board using 2 μs trigger gate signal [5]. We closed the shield after energy calibration and stated the measurement of background.

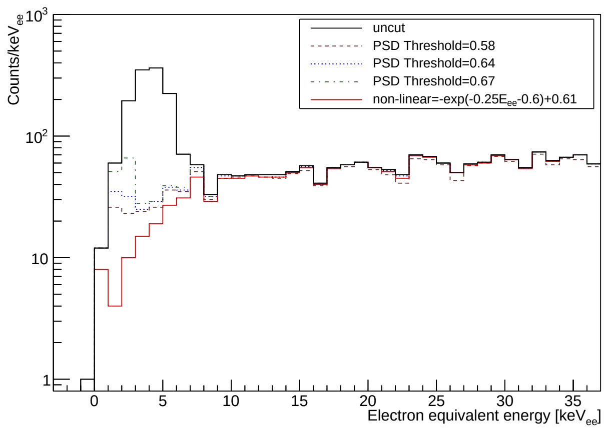

Figure 1: Effectiveness of PSD cut. The ^{60}Co energy spectrum has a flat event rate in low energies range below 40 keV.

We have the Ingot #94 exposure data of 28.3 d×1.3 kg. Ingot #94 crystal is pure equivalent the DAMA/LIBRA crystals, radio impurity has 4.6 ± 1.2 μBq/kg, 7.9 ± 4.4 μBq/kg, and 19 ± 6 μBq/kg for ^{232}Th , ^{226}Ra and ^{210}Po which are equivalent to those of the DAMA/LIBRA crystals [6].

A low-energies region less than 100 keV, the noise events mixed in scintillation events from PMT and VME bus line. We applied two noise reduction method: (1) A single noise rejection method separates single pulse noise from scintillation pulses. The single pulse noise waveform has only one pulse over 200 ns. In contrast, the scintillation waveform has many pulses over the scintillation decay time. Therefore, the scintillation events are discriminated using the time difference ΔT between two single pulses after the first pulse. We applied the scintillation events threshold with $\Delta T > 200$ ns; (2) A noise reduction using PSD (Pulse Shape Discrimination) used to remove noise events below 10 $\mathrm{keV_{ee}}$ from all events. We investigated noise events from VME bus traffic and PMT signal cable lines cause the baseline instability. PSD parameter R_{PSD} is defined by

$$R_{\mathrm{PSD}} \equiv \frac{Q_{\mathrm{Part}}}{Q_{\mathrm{Total}}} = \frac{\int_{200\ \mathrm{ns}}^{1200\ \mathrm{ns}} I(t)dt}{\int_{0\ \mathrm{ns}}^{1200\ \mathrm{ns}} I(t)dt}, \tag{1}$$

where $I(t)$ is the current pulse, Q_{Total} and Q_{Part} are the charges over two different time intervals since pulse start. Figure 1 shows a effectiveness of PSD analysis. The ideal ^{60}Co energy spectrum has a flat event rate in the low energy region. We applied a simple noise threshold to be $R_{\mathrm{PSD}} > 0.64$ as noise events on the PSD scatter plot in Figure 1.

3. Result and summary

Figure 2 shows the background energy spectrum for the live time of 5.6 day × 1.3 kg after 36.68 days from data acquisition started on the real time. We found faint peaks of ^{40}K (1462 keV) and ^{208}Tl (2662 keV). These γ-rays come from the surrounding materials such as the PMT circuit. Some distinct peaks below 100 $\mathrm{keV_{ee}}$ are due to impurities in NaI(Tl) crystal. By further analysis, we found cosmogenic isotopes, ^{125}I ($T_{1/2,\ ^{125}\mathrm{I}} = 59.4$ d), and ^{126}I ($T_{1/2,\ ^{126}\mathrm{I}} = 12.5$ d). They quickly decay during the low background measurement. The remaining background is due to ^{210}Pb .

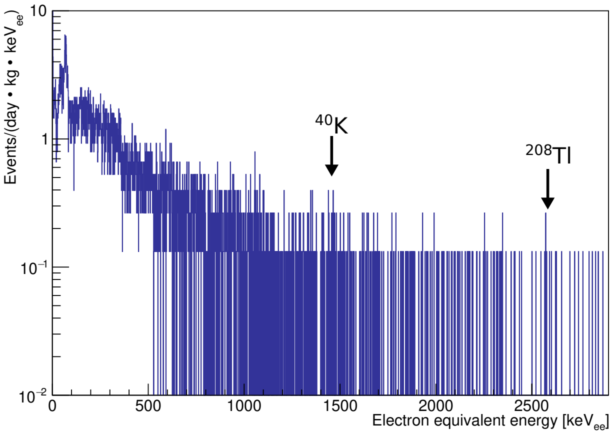

Figure 2: back ground energy spectrum for the live time of 5.6 day × 1.3 kg after 36.68 days on the real time.

The contamination derived from this peak is 480 μBq/kg, which is inconsistent with the obtained value from α-ray intensity of ^{210}Pb [6]. Table 1 show the radio-activity in NaI(Tl) detectors for the other groups. From the energy spectrum below 5 keV, the Ingot #94 detector background rate is estimated to be about 1.5–3 $\mathrm{day^{-1}kg^{-1}keV^{-1}}$. We conclude the main origin of the 46.5 keV peak is an outer source, such as a surface of the shield and PMT circuit. We are trying to further improve of background by cleaning the surrounding materials and additional shielding.

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Table 1: Radio-activity in detectors for the other groups. The unit of background rate is $\mathrm{day^{-1}kg^{-1}keV^{-1}}$.

	DAMA/LIBRA [7]	COSINE [8]	ANAIS-112 [9]	SABRE [10]	PICOLON (This work)
Background rate	1	3	3.605±0.003	1.39±0.02	1.5–3

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