

PICOLON dark matter search

~ Development of highly redio-pure NaI(Tl) scintillator~

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Abstract. The PICOLON project is developing a high purity NaI(Tl) crystal by optimizing the purification methods. The reduction method for ⁴⁰K was established and we got sufficiently pure NaI(Tl) crystal for ⁴⁰K. On the other hand, the concentration of ²¹⁰Pb is still higher than our goal. The investigation is now in progress to fix the origin of the contamination.

The construction of the second phase detector system of PICOLON detector will be started in 2020 spring. The total mass of the second phase is about 54 kg. The background level by the test measurement using a single module is reported.

1. WIMPs search by NaI(Tl) scintillator

The sensitivity to the WIMPs (Weakly Interacting Massive Particles) has been increased in recent decades. The experimental limit on the normalized cross section of spin-independent interaction has already reached to less than 10^{-46} cm² [1]. Although no significant signal for



WIMPs by using many target nuclei, only NaI(Tl) detects a significant annually modulating signal which can be interpreted to be WIMPs. Only one group, DAMA/LIBRA reported a significant annually modulating signal by applying a large volume NaI(Tl) scintillator [2].

No significant verification by using the same target nuclei has been performed yet because of the large background due to lack of the purity of NaI(Tl) crystal. Many groups are trying to develop the NaI(Tl) detector with the highest sensitivity to WIMPs. The requirements for the NaI(Tl) detector to verify the DAMA/LIBRA result and to find a new signature for dark matter are listed below.

- The smallest radioactive impurities (RIs) in the NaI(Tl) crystal.
- The smallest RIs in surrounding materials.
- The lowest energy threshold at least down to 1 keV_{ee} , where keV_{ee} stands for the electron equivalent energy.
- The large detector more than one hundred kg.

Our group, PICOLON (Pure Inorganic Crystal Observatory for LOW-energy Neutr(al)ino) is developing a NaI(Tl) detector suitable for WIMPs search. COSINE in Korea, ANAIS in Spain, SABRE in Italy are developing the NaI(Tl) detectors independently. We will report on the present status and prospects of the PICOLON project.

2. PICOLON dark matter search project

2.1. Outline of the PICOLON project

The aim of the PICOLON project is the investigation of fundamental processes in nuclear and particle physics. Cosmic dark matter and neutrino properties are present our main subjects. Our present main aim is to verify the annual modulating signal which is claimed by DAMA/LIBRA by developing the highly radio-pure NaI(Tl) detector. We started purification work from 2012 and successfully reduced the main origins of the background, U-chain, Th-chain, and ^{40}K .

PICOLON project consists of four phases. (Phase-I) The development of highly radio-pure NaI(Tl) crystal, (Phase-II) The search for annual modulating signal by a small scale (54 kg) NaI(Tl) array, (Phase-III) The search for annual modulating signal and verify the DAMA/LIBRA's result by a large scale (250 kg) NaI(Tl) array, (Phase-IV) The search for dark matter by a huge and extremely log background scintillator. A conceptual diagram is shown in Figure 1.

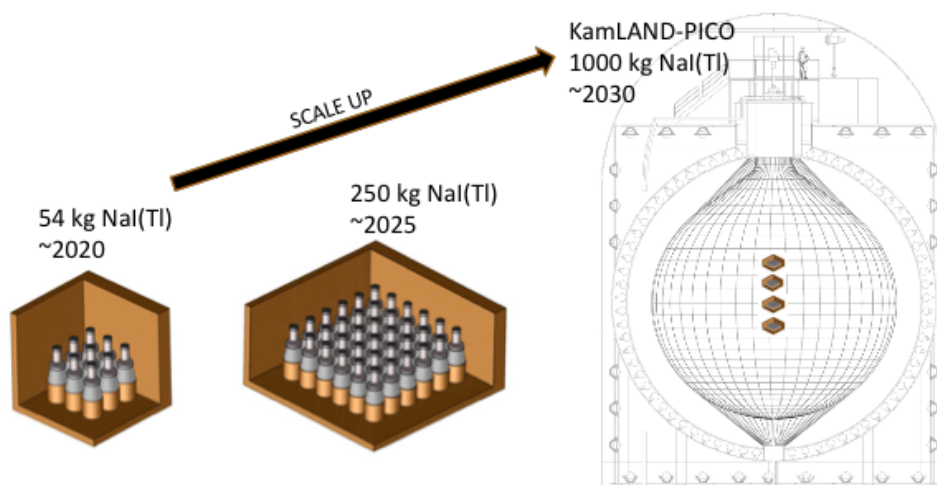


Figure 1. A conceptual drawing of the PICOLON project from Phase-II.

2.2. Present status of the NaI(Tl) purification

The radioactive impurities (RI) in NaI(Tl) crystal were removed individually because the chemical and physical behaviors are different from each other elements.

The ^{40}K was effectively removed by applying the re-crystallization method [3, 4]. The RIs belong to U-chain are not in secular equilibrium because half-lives of some isotopes are too long. It consists of five subchains which start and end at long-lived isotopes. The RIs belong to Th-chain, on the contrary, becomes quickly in secular equilibrium, because the half-lives are relatively short for almost all the RIs.

2.3. U-chain

U-chain is divided into five subchains; (1) $^{238}\text{U}\sim^{234}\text{U}$ ($T_{1/2,^{234}\text{U}} = 2.45 \times 10^5$ y), (2) $^{234}\text{U}\sim^{230}\text{Th}$ ($T_{1/2,^{230}\text{Th}} = 8.0 \times 10^4$ y), (3) $^{230}\text{Th}\sim^{226}\text{Ra}$ ($T_{1/2,^{226}\text{Ra}} = 1.60 \times 10^3$ y), (4) $^{226}\text{Ra}\sim^{210}\text{Pb}$ ($T_{1/2,^{210}\text{Pb}} = 22.3$ y), (5) $^{210}\text{Pb}\sim^{206}\text{Pb}$ (stable).

The subchains (1) to (4) were effectively reduced by selecting surrounding materials during crystallization. The subchain (5) is not in secular equilibrium with other sub-chains because ^{210}Pb is provided not only from the raw material of NaI but also from ^{222}Rn in air and water.

We tried to remove Pb ion in the water solution of NaI by using ion-exchange resin. Proper selection and appropriate usage of resin resulted effective removal of ^{210}Pb [5]. The lowest concentration in NaI(Tl) crystal was 30 ± 7 $\mu\text{Bq/kg}$, however, recent results showed worse values around 1000 $\mu\text{Bq/kg}$. To keep stable reproducibility of the ^{210}Pb concentration is the present issue.

2.4. ^{40}K

The contamination of natural potassium results in a serious background in low energy regions due to beta-rays and X-rays since the natural abundance of ^{40}K in potassium is 0.0117 %. The concentration of natural potassium in NaI(Tl) should be less than 20 ppb to verify the DAMA/LIBRA's result.

The potassium was effectively removed from NaI powder by the re-crystallization method. The solubility of potassium iodide in 100 g water is 144 g at 25 °C and the contamination of potassium in NaI is less than a few ppm. Small NaI crystals were deposited from saturated NaI solution when it was cooled from 100 °C to room temperature. Potassium iodide remains in the water and removed from NaI powder by filtration. This method was effectively removed potassium ion and the concentration of potassium is less than 20 ppb which corresponds to about 600 $\mu\text{Bq/kg}$.

2.5. Measurement of impurities in NaI(Tl)

The results of RI reduction were estimated by making a NaI(Tl) scintillator and perform low background measurement. A cylindrical NaI(Tl) crystal with a diameter of 12.7 cm and a length of 12.7 cm was made by using a high purity graphite crucible. The NaI(Tl) crystal was encapsulated into an acrylic housing. A quartz light guide was glued at the one edge of the acrylic housing to avoid the deliquesce and to introduce scintillation photons to a photomultiplier tube (PMT). The scintillation photons were collected by a low-background PMT with 7.62 cm diameter, Hamamatsu Photonics R11065-20mod. The waveform of the current signal from the PMT was digitized to perform off-line analysis for the particle identification and the noise reduction. The detector was installed into a passive shield made of 10 cm thick copper and 20 cm thick lead placed at KamLAND area in Kamioka underground observatory in Gifu Prefecture Japan.

Low background measurement was performed for two months to get the trend of cosmogenic RIs and alpha rays from ^{210}Po . The energy spectrum in the low energy region is shown

in Figure 2. Two RIs, ^{125}I ($T_{1/2} = 59.4$ day) and ^{210}Pb were prominently observed. The ^{125}I was produced when the NaI was placed in the surface laboratory and factory. This isotope decays with relatively short half-life so that it is not serious background origin.

The concentration of ^{210}Pb was estimated from both gamma-ray due to the beta decay of ^{210}Pb and alpha-ray due to the alpha decay of ^{210}Po ($T_{1/2} = 138.4$ day). The value derived from the gamma-ray was about 1000 $\mu\text{Bq/kg}$. The event rate of alpha-ray from ^{210}Po was increasing and its trend was well fitted by exponential and constant. The final activity of ^{210}Po was calculated as 860 $\mu\text{Bq/kg}$ which was consistent with the value derived from gamma-ray.

3. Conclusion and prospect

The purification methods are listed in Table 1. In this table, all the Th-chain isotopes are assumed to be secular equilibrium, ^{238}U and ^{226}Ra are assumed to be secular equilibrium.

We have developed a highly radio-pure NaI(Tl) detector to search for cosmic dark matter. The purification methods for U-chain, Th-chain, and ^{40}K were well established and the concentrations of them were small enough to start the experiment. The last problem is the reduction of ^{210}Pb . We have investigated the behavior of lead ion in the water and the surrounding environment. The purification method is developed concurrently with the construction of the phase-II of PICOLON detector system.

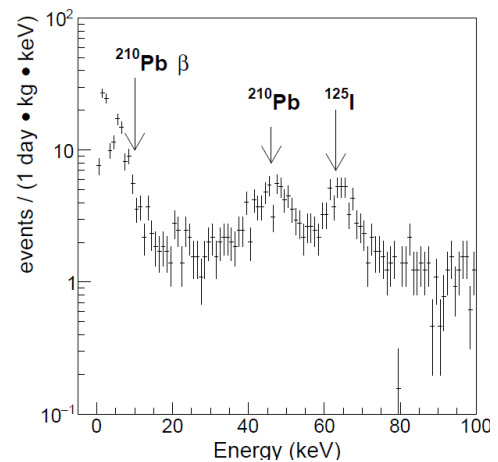


Figure 2. The energy spectrum in low energy region.

Table 1. Impurities of NaI(Tl) crystals developed by the PICOLON project.

	Ingot#26 (2015)	Ingot#37 (2016)	Ingot#71 (2018)	Ingot#76 (2019)	Goal
Size	3" ϕ \times 3"	4" ϕ \times 3"	3" ϕ \times 3"	5" ϕ \times 4"	5" ϕ \times 5"
^{nat}K (ppb)	2630	120	< 20	< 20	< 20
^{232}Th (ppt)	0.4 ± 0.5	3.7 ± 0.5	1.7 ± 0.2	–	< 4
^{238}U (ppt)	4.7 ± 0.3	5.9 ± 0.3	9.7 ± 0.8	4.4 ± 0.2	< 10
^{210}Pb ($\mu\text{Bq/kg}$)	30 ± 7	2300	1076	860	< 50
Method	Pb resin	I#26+ cation resin	Double re- cryst.	Pb resin+ dou- ble re-cryst.	

4. Acknowledgment

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