Purification of the NaI(Tl) crystal for dark matter search project PICOLON

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Abstract. Direct search for dark matter is one of the most important problems in astrophysics. Significant signal for dark matter will be a hint to clarify the origin of the universe. Only DAMA/LIBRA experiment with NaI(Tl) detector has ever suggested the presence of dark matter signal. Verifying the DAMA/LIBRA result by a NaI(Tl) detector is urgent and important task. We have tried to purify NaI(Tl) crystal to search for dark matter. In this presentation, the present status of purification will be discussed. The concentration of potassium is successfully reduced to desired sensitivity. The ^{210}Pb, which is difficult to reduce, has been reduced effectively. Present status of low background measurement in Kamioka observatory will be shown.

1. Introduction

Weakly interacting massive particles (WIMPs) are one of the most attractive dark matter candidates. Expected count rate of WIMPs detected by 1 ton NaI(Tl) crystals is a few events a year. Also expected energy of signal is lower than 10 keV_{ee}, where keV_{ee} stands for the observed energy calibrated by kinetic energy of electron. Therefore low back ground detector is necessary to search for WIMPs. DAMA/LIBRA is searching for WIMPs by highly radio-pure and large volume NaI(Tl) crystals for a long time. As a result, they reported that they observed annual modulation signal[1].

PICOLON aims at verifying an annual modulation signal claimed by DAMA/LIBRA by high sensitivity NaI(Tl) detector. We have been trying to purify the NaI(Tl) crystals because the impurities in a NaI(Tl) crystal become the origin of background. Our goal is to make the NaI(Tl) crystal purer than DAMA/LIBRA's one. We have successfully reduced U and Th in the NaI(Tl) crystals enough to search for dark matter. Current most important task is to reduce potassium and ^{210}Pb.

2. Recrystallization

2.1. Purification of NaI by recrystallization

We have ever applied cation exchange resins to reduce potassium, however, potassium could not be reduced effectively. We tried to purify NaI powder by recrystallization to reduce potassium. Recrystallization is the method of dissolving the crystal once and crystallizing it again. COSINE-100 reported that potassium could be effectively reduced by recrystallization[2].

There are 3 steps in the procedure of recrystallization. Firstly, NaI is dissolved in water at high temperatures. Secondly, NaI solution is cooled. NaI in saturated solution is deposited as the temperature becomes lower. On the other hand, potassium remains dissolved because the mass of it in solution is much less than the solubility of potassium in water. Finally, NaI crystals are separated from solution by suction filtration.

2.2. The concentration of potassium

The background measurement of the NaI(Tl) crystal which is purified by recrystallization was done in Kamioka Underground Observatory in Gifu Prefecture, Japan. The experimental cite is located in KamLAND area in the observatory. Figure 1 shows the background spectra taken by Ingot68 and Ingot71. Ingot68 is the NaI(Tl) crystal purified by resins for lead and for potassium. Ingot71 is the NaI(Tl) crystal purified by recrystallization. The prominent peak of ^{40}K (1462 keV_{ee}) was successfully reduced by recrystallization. The concentration of potassium was reduced to 20 ppb by recrystallization. Although a small peak is observed at 1462 keV_{ee} in Ingot71, this gamma ray came from surrounding materials of the detector because of no significant events of beta rays from ^{40}K. We successfully established the reduction method for potassium for desired sensitivity.

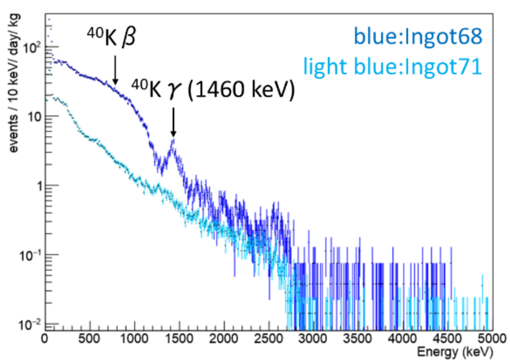

Figure 1. Background spectra of Ingot68 (blue) and Ingot71 (light blue).

3. The concentration of ^{210}Pb

Our last problem is to remove ^{210}Pb. We have used the resin to remove lead. In the case of Ingot26, we realized the ^{210}Pb concentration of 30 μBq/kg by using the resin to remove lead. However, the concentration of ^{210}Pb was high in all NaI(Tl) crystals after Ingot26. We guessed the cause may be that the cation exchange resin increased ^{210}Pb in NaI. Therefore, we decided not to use a cation exchange resin in the next purification of NaI. The latest crystal was made from the NaI purified by recrystallization and resin to remove lead. We evaluated the concentration of ^{210}Pb by two ways.

One way is evaluation by gamma rays. Figure 2 shows three peaks in low energy region. ^{210}Pb emits gamma rays whose energy is 46.5 keV (B). The other peaks come from ^{126}I (A) and ^{125}I (C). Since ^{126}I has a half-life of 13.11 days and ^{125}I has a half-life of 59.408 days, these peaks disappear after enough time has passed. However the peak of ^{210}Pb remains because of its long half-life. Therefore it is important for WIMPs search to remove ^{210}Pb. The concentration of ^{210}Pb in NaI(Tl) is calculated by assuming all the gamma rays from ^{210}Pb was emitted in the NaI(Tl) crystal and resulted as 920 μBq/kg.

Another way is evaluation by alpha rays. Assuming that 226Ra, 222Rn, 218Po and 214Po are in radioactive equilibrium, the concentration of ^{210}Po is estimated to be 870 μBq/kg. When enough time has passed ^{210}Po and ^{210}Pb is the same counts because of radioactive equilibrium. Evaluation by alpha rays is necessary to calculate the concentration of ^{210}Pb because Gamma rays are the sum of inside and outside. Alpha rays are increasing slightly now (Figure 3). Further investigation will be done from now on.

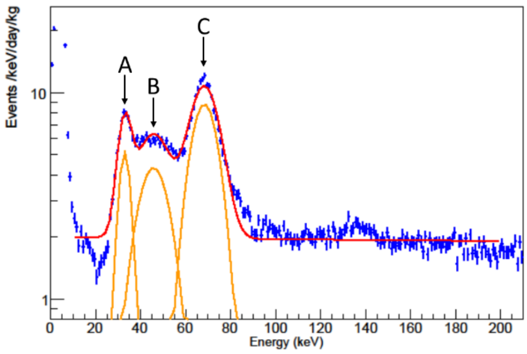

Figure 2. Peak in low energy.

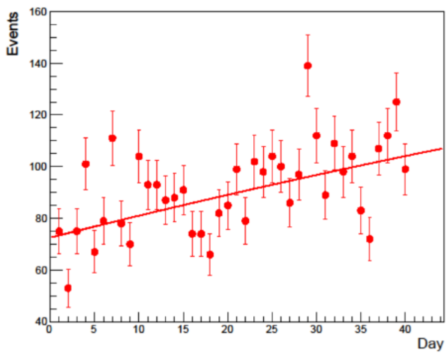

Figure 3. Alpha events trend.

4. Summary and prospects
We found that the recrystallization method was effective to reduce the concentration of potassium. We purified NaI by double recrystallization and resin to remove ^{210}Pb. According to the results of low background measurement of Ingot76 in Kamioka Underground Observatory, ^{210}Pb still remained in the NaI(Tl) crystal.

We suspect that ^{222}Rn water increases the ^{210}Pb in the NaI(Tl) crystals. We are investigating ^{210}Pb in pure water. Development of pure water is ongoing. Then large volume NaI(Tl) detector will be constructed in 2020.

5. Acknowledgment
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