Quality Study of Purified Liquid Scintillator

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Abstract. We have been distilling the KamLAND liquid scintillator (LS) for the low energy solar neutrino observation. The distillation removes radioactive impurities from LS efficiently. We developed two types of high sensitivity radon detectors to monitor $^{222}$Rn contamination which causes a primary background source $^{210}$Pb. Their required sensitivity is several mBq/m$^3$. The features and the measurement results of these detectors are presented. We also report the study of liquid scintillator properties after the distillation: attenuation length, light output and PPO density.

1. MiniLAND : a high sensitivity Rn detector

MiniLAND was developed as a high sensitivity radon detector to monitor $^{222}$Rn concentration in LS after the distillation. The detector achieved very low background environment by internal and external shields and optically-isolated cell structure to reduce accidental backgrounds drastically. NaI solution as an internal gamma ray shield has potential applications for some other experiments.

The detector consists of 4 acrylic boxes, 16 cells (100x20x20 cm$^3$) and contains ~0.36 m$^3$ LS. Total 32 PMTs (low background type) are set on cells and detect a prompt and delayed signal in a certain time window to tag $^{222}$Rn daughter coincidence events ($^{214}$Bi/$^{214}$Po). All cells are optically separated from each other with reflectors (sandwiching air and a tyvek sheet between two acrylic boards), which lead to reduction of accidental backgrounds and give better light yield to PMTs by total reflection.

Low background environment is essential for a high sensitivity detector. The miniLAND detector is surrounded by 3 layers of shield which are a copper box, low impurity lead bricks and boron-containing polyethylene bricks. Very low background materials are selected at the construction of these shields based on our studies of contamination and background reduction impacts. For example, $^{210}$Po activity from our lead is ~15 % (25 Bq/kg) of one from usual lead and a lead shield reduces gamma rays above 300 keV by 10$^{-4}$ from MC simulation.

External gamma rays are the dominant backgrounds to monitor radon, but gamma rays from a PMT are not negligible. NaI solution between acrylic boxes and PMTs reduces gamma rays from PMTs above 300 keV by 10$^{-1}$ from MC simulation. NaI solution consists of 62 wt% NaI + 0.1 wt% Na$_2$S$_2$O$_3$ and both of these are inexpensive. A gamma ray shield for PMT backgrounds must be transparent, high atomic number and high density material. NaI satisfies these requirements: Z=53, 64.1 wt% high solubility (at 20 degrees) and high transmission with Na$_2$S$_2$O$_3$. NaI powder of our choice is very low impurity product: $^{238}$U < 9.2, $^{232}$Th < 17 and $^{40}$K < 41 mBq/m$^3$.

These above features allow a detector to have a high sensitivity to measure several mBq/m$^3$ radon. All the results of $^{222}$Rn measurements in purified LS were less than several mBq/m$^3$. MiniLAND has been used to qualify the distilled LS before it is supplied to KamLAND.
2. High sensitivity Rn detector by concentration method
A sensitive radon (Rn) detector was newly developed by means of combining conventional concentration in a cold trap with an electrostatic collection of charged decay products. Although various methods had been examined to measure Rn dissolved in liquid scintillator (LS), small and portable types of detectors with the sensitivity of an order of mBq/m³ have not been realized.

Rn dissolved in LS is extracted by bubbling and then trapped in the copper wool under liquid nitrogen temperature (cold trap). The carrier gas, high purity nitrogen (N₂) produced by newly constructed facility for KamLAND solar phase, is circulated to reduce background from contamination in N₂ itself. Concentrated Rn in the cold trap is released under room temperature, then pushed out of the trap to the detector vessel to count decay products of Rn. Among the progenies of ²²²Rn, polonium (²¹⁸Po and ²¹⁴Po) is positively charged under less humidity. Charged Po is collected by electrostatic field onto the PIN-photo diode to detect α particles emitted by Po decays. There are two kinds of filters through the path (moderate cold trap and molecular sieves) to reduce vaporized organic components and moisture in processing carrier gas, which make the collection efficiency of Po worse due to neutralization. We evaluated the total detection efficiency using a commercial Rn counter (Model1027, Sun Nuclear Corporation) calibrated in advance. The result obtained was 0.191 ± 0.013.

As a result, the detection sensitivity for Rn dissolved in LS was found to be 7mBq/m³ using ~55 litters of measuring LS. Further, the developed method is applicable to measure Rn density in gases whose condensation points are comparable or less than liquid N₂ temperature (sensitivity ~26 μ Bq/m³), and Rn emanation rate from materials (~8 atoms/day). We realized, in the present study, high sensitivity Rn measurement using the proposed method. The developed detector can provide useful information for LS purity during purification period for KamLAND solar phase.

3. Properties of the KamLAND liquid scintillator before and after the purification
We have investigated various methods to find the most effective way to remove the radioactive background sources from the KamLAND LS for the solar neutrino observation. As a result, it turned out that the distillation and nitrogen purge are effective to remove radioactive metals and gases from LS, respectively. There was a concern that properties of LS may change after the distillation because LS is heated in the distillation process. Hence we evaluated attenuation length, light output and PPO density of LS before and after the distillation because these parameters are directly linked to the photon yield.

Attenuation length is measured with a vertically aligned tube monitoring transmission of LED light through various depth of sampled LS. It became slightly shorter after the distillation.

Light output is estimated with a measurement of ⁶⁰Co Compton. It was almost same as before.

PPO density is measured with a gas chromatography spectroscopy analysis and was consistent with a preset value at the distillation system.

These results showed that properties of distilled LS were not significantly degraded by the distillation.