The laboratory measurement of radioactivity purification for Pb212 in liquid scintillator (post-print)

Authors: Wei Hu, Jian Fang, Bo-Xiang Yu, Xuan Zhang, Li Zhou, Xiao Cai, Li-Jun Sun, Wan-Jin Liu, Lan Wang, Jun-Guang Lu

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Abstract

The liquid scintillator (LS) has been widely utilized in previous, current, and future neutrino experiments, with increasingly stringent requirements for LS radiopurity. Water extraction is a powerful method for removing soluble radioactive nuclei, and a mini-extraction station has been constructed. To evaluate the extraction efficiency and optimize operational parameters, a setup for loading radioactivity into LS and a laboratory-scale setup for measuring radioactivity using the Bi$^{\{212\}\text{-}Po}${212}-Pb^{208} cascade decay have been developed. Experience gained from laboratory studies will be useful for the design of large-scale water extraction plants and the optimization of future operations.

Full Text

Preamble

Wei Hu1,2;1), **Xiao Cai**1, **Jian Fang**1;2), **Bo-Xiang Yu**1, **Xuan Zhang**1,2, **Li-Jun Sun**1, **Wan-Jin Liu**1, **Lan Wang**1, **Li Zhou**1, **Jun-Guang Lu**1

1 State Key Laboratory of Particle Detection and Electronics, Institute of High Energy Physics, CAS, Beijing 100049, China
2 Institute of High Energy Physics, University of Chinese Academy of Sciences, Beijing 100049, China

Abstract

Liquid scintillator (LS) has been widely utilized in past, current, and future neutrino experiments, with increasingly stringent requirements on radio-purity. Water extraction is a powerful method for removing soluble radioactive nuclei, and a mini-extraction station has been constructed for this purpose. To evaluate

extraction efficiency and optimize operation parameters, we developed a setup to load radioactivity into LS and a laboratory-scale measurement system utilizing the 212Bi-212Po-208Pb cascade decay. The experience gained from these laboratory studies will be valuable for designing large-scale water extraction plants and optimizing their operating conditions.

Keywords: liquid scintillator, radioactive loading, radioactive measurement, cascade decay, water extraction
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Introduction

Liquid scintillator plays a crucial role in intensity frontier neutrino experiments. The Jiangmen Underground Neutrino Observatory (JUNO) is a multi-purpose neutrino experiment with the primary scientific goal of determining the neutrino mass hierarchy. The neutrino detector will be filled with 20 kilotons of LS (fiducial mass). To suppress accidental backgrounds and achieve the potential goals of solar neutrino studies, the basic radioactivity contamination requirement for JUNO LS is 10−15 g/g (in this paper, g/g refers to grams of 232Th or 238U per gram of LS) for both 238U and 232Th.

The general methods for removing radioactivity contamination are water extraction, nitrogen stripping, and distillation, which are sensitive to soluble nuclei, radon, and insoluble nuclei, respectively. Before mass production of purified LS, each method requires a prototype and optimized operation parameters. For example, the Borexino experiment (a solar neutrino experiment using trimethylbenzene as the LS solvent), which holds the world-record radioactivity contamination levels of 10−18 g/g for 232Th/238U [1], demonstrated that the parameters of large-scale purification plants are consistent with those of the prototype [2][3].

Although these purification methods are efficient for Borexino' s LS, they require careful study for JUNO' s LAB-based LS. A water extraction prototype has been constructed at IHEP, Beijing, to validate the prototype and optimize operation parameters. The radioactivity of LS before and after purification must be measured. However, typical U and Th contaminations in LS are on the order of 10−13 to 10−14 g/g, making direct measurement impossible in the laboratory. A common approach is to load radioactive nuclei such as 222Rn or 220Rn into the LS, purify it using the prototype, and then measure both purified and unpurified samples with a clean detector.

This paper reports on the radon loading technology, the radioactivity measurement setup, water extraction efficiencies, and optimized operation parameters. The achieved efficiency reaches the world average level, indicating successful prototype operation, and the optimized parameters will be useful for future medium-scale and mass production plants.

2 Radioactivity Measurement Setups

2.1 220Rn Loading

The limit of radioactivity measurement in the laboratory is 10−9 g/g, while the natural contamination of LS is 10−13 to 10−14 g/g. Therefore, direct measurement of such low radioactivity is impossible in the laboratory. To study the purification effect on LS in laboratory experiments, the only solution is artificial contamination of samples with radioactivity. Since powdered radioactive sources do not dissolve in LS and the solubility of liquid radioactive sources in LS is not high, the general method is to load radon into LS. Because radon is a non-polar gas, its solubility in LS is high—approximately 13 times the radon concentration in air at room temperature [4]. Therefore, bubbling radon through LS is an effective method for loading radioactivity.

The commonly used radon isotope is 222Rn from the 238U decay chain, which has a half-life of 3.8 days. For example, in the Borexino experiment, 222Rn was loaded into LS and the contamination of its daughter 210Po was measured. The disadvantage of this approach is that it requires months for 210Po to accumulate to a measurable amount, since its parent 210Pb has a half-life of 22.3 years [5]. Additionally, the long half-life of 210Pb would contaminate the experimental apparatus.

A better candidate is 220Rn from the 232Th decay chain, as shown in Fig. 1 [Figure 1: see original paper], which has a half-life of 55 seconds. After loading, 220Rn quickly decays to 212Pb with a half-life of 10.6 hours. The decay of 212Pb' s daughters, 212Bi and 212Po, produces a famous cascade decay (- cascade) because the half-life of 212Po is only about 300 ns. This cascade decay provides a pair of time-correlated signals in our experiments with high efficiency and extremely low background. With 220Rn loading, the water extraction efficiency is estimated using the 212Pb nucleus. The 10.6-hour half-life of 212Pb is long enough for extraction and measurement, yet short enough to avoid contaminating experimental setups. Meanwhile, a large amount of 220Rn in LS decays to 212Pb in a very short period, leading to high radioactivity loading efficiency.

A 1200 Bq 220Rn source produced by Nanhua University was used. Nitrogen passed through a bubbler filled with water, then flowed through the source to carry 220Rn out, and finally passed through a bubbler filled with LS. Research conducted by Nanhua University showed that the 220Rn release rate from the 232Th source increased with higher environmental humidity. After bubbling 220Rn into LS for 74.2 hours, the concentration of 212Pb reached equilibrium. In the following studies, LS was bubbled with 220Rn for 20 hours, which achieved 2/3 of the equilibrium 212Pb concentration [6].

The laboratory setup for radon loading of LS samples is shown in Fig. 2 [Figure 2: see original paper]. The loading method used in this paper involved bubbling 220Rn into the LS sample, with the corresponding setup housed inside a glove

box.

The radioactivity was measured using the experimental setup depicted in Fig. 3 [Figure 3: see original paper]. In a light-tight box, a pair of PMTs (XP2020) was placed on opposite sides of an LS sample cell to perform double coincidence measurements. Coincidence measurement reduces the influence of single PMT fluctuations on experimental data. The LS container was a cylindrical quartz glass bottle with 5 cm diameter, 1.5 cm thickness, and a capacity of 17.1 g of LS. Gamma rays from ambient radioactivity were attenuated by shielding with low-activity lead bricks [4]. A flash ADC (DT5751 by CAEN with 1 GHz sampling frequency) was used for data acquisition. The total background event rate (including , , , and cascade decay events) during measurement was 0.25 Hz. After acquiring all events, - cascade events were selected through offline analysis. This setup was designed as a - counting system.

3 Measurement Results and Analysis

3.1 - Counting System Performance

3.1.1 Real - Cascade Events After 220Rn loading, three hours of data were taken to determine the initial 212Pb concentration. The coincidence time window for signals from the two PMTs was required to be smaller than 5 ns, accounting for cable length differences. 99.99% of - cascade events met this requirement, with a statistical error of 9.55×10−8 (statistical errors will not be discussed further in this section as they were negligible).

The time interval distribution between the decay and decay of - cascade events is shown in Fig. 4 [Figure 4: see original paper]. The distribution can be described by the formula [7]:

$$f(t) = \times N_0 \times e^{-\frac{t}{\tau}}$$

where is the lifetime of 212Po and N0 is a parameter related to the concentration of 212Po. T1/2 is the half-life of 212Po and T1/2 = ln2 • . Therefore, the formula can be written as:

$$f(t) = \times N_0 \times 2^{-\frac{t}{T_{1/2}}}$$

The following function was used to fit the time interval distribution:

$$f(t) = \times p_0 \times 2^{-\frac{t}{p_1}}$$

The parameter p1 in the fitting result represents the half-life of 212Po. The theoretical half-life is 298 ns, while the experimental result is 298.4±1.2 ns, confirming that the - counting setup reliably detected - cascade events.

For each - cascade event, there is a time interval between the event and the event, which should theoretically be the same when measured by the two PMTs. At minimum, any difference must be very small due to PMT variations or other experimental effects. Fig. 5 Figure 5: see original paper shows the difference in time intervals of - cascade events detected by the two PMTs, which fell within the coincidence time window. The number of entries before normalization was 104640. A Gaussian function was used to fit the distribution, yielding a mean value of 0.51 and of 0.91. The difference in time interval was almost entirely within (-2 ns, 3 ns), with 95.92% of events meeting this requirement—consistent with the probability of a Gaussian variable falling within the 2 range.

The event energy was proportional to the total charge collected. The total charge from the PMT was estimated by integrating the entire pulse. Fig. 6 Figure 6: see original paper shows the integral value of the event pulse (the second pulse in a double-pulse event). Due to the mono-energetic nature of events, the distribution was concentrated like a Gaussian. After applying the two selection criteria discussed above, 92.80% of the second pulse integral values fell within (1700 FADC, 8000 FADC).

3.1.2 Cut Selection Criteria Based on the analysis of real - events, several cut criteria were established: (1) The coincidence time window of the two PMTs was required to be smaller than 5 ns; (2) The difference in time intervals of - cascade events detected by the two PMTs was within (-2 ns, 3 ns); (3) The integral value of events was within (1700 FADC, 8000 FADC).

After bubbling 220Rn into LS for 20 hours, approximately 2×104 - events were detected in 17.10 g of LS during 30 minutes of data acquisition using these cut criteria.

3.1.3 Background Events Background events were measured for one day using pure LS (17.10 g) without 220Rn loading. 163 double-pulse events were found, but the integral value distribution of the second pulse (potentially a fake event pulse) and the difference in time intervals of double-pulse events detected by the two PMTs differed significantly from real - events, as shown in Fig. 5(b) [Figure 5: see original paper] and Fig. 6(b) [Figure 6: see original paper]. The integral values of the second pulses were almost all less than 1700 FADC, and only a small percentage of time interval differences fell within (-2 ns, 3 ns). After applying these cut criteria to background events, only 2 background - cascade events remained per day, compared to 163 without cuts.

Compared with 2×104 real - cascade events detected in 220Rn-loaded LS within 30 minutes, the 2 background events per day can be neglected in subsequent studies.

3.2 Water Extraction Studies

Purification by water extraction relies on the polarity of water molecules to separate polarized impurities (e.g., free-state ions of radioactive metals) from non-polar LAB and fluor molecules. Water extraction is highly effective for most ionic metals such as K, Ra, and Bi, but less effective for Po and Pb. For Po and Pb, the reduction was equally fast but less effective, with an 82-87% removal fraction observed in SNO+ laboratory studies [9][10].

After achieving a good 212Pb concentration in the scintillator, purification was performed by water extraction. The purification efficiency was defined as:

$$y - x = 1 - x$$

where x represents the event number after purification, y represents the event number before purification, and u represents purification efficiency.

The statistical error of efficiency was calculated using Clopper-Pearson parameter estimation of the Binomial Distribution [8]:

$$\sigma^{+} = \left(1 + \frac{n - \hat{s}}{\hat{s} + 1} f_{\alpha/2}(2(n - \hat{s}), 2(\hat{s} + 1))\right)^{-1} - \hat{p}$$

$$\sigma^{-} = \hat{p} - \left(1 + \frac{n - \hat{s}}{\hat{s} + 1} f_{1-\alpha/2}(2(n - \hat{s} + 1), 2\hat{s})\right)^{-1}$$

where 1- is the confidence level, n is the total number of events, f /2 is the upper /2 fractile of the F-distribution, $\hat{s}$ is the number of passed events, and $\hat{p} = \hat{s}/n$. In this paper, n = y, $\hat{s}$ = y - x, and the confidence level is set at 0.683.

Water extraction was performed using equal amounts of 12 M deionized water and liquid scintillator. A separatory funnel and magnetic stirrer were used to mix 35 ml of water with 35 ml of scintillator. The solution was mixed and then separated in a process called one-stage extraction. After each separation, a clean separatory funnel and fresh deionized water were used for multi-stage extraction. Scintillator samples were placed in small test tubes containing 17.1 g of LS, and the - counting system was used to measure radioactivity before and after purification.

The stability of the - counting system was studied by measuring purification efficiency at three different times using the same LS sample. The efficiencies were consistent: 84.3+1.2-1.3%, 82.7+1.5-1.9%, and 83.3+1.8-1.6%, confirming that the system reliably optimized purification parameters.

Fig. 7 [Figure 7: see original paper] shows the relationship between extraction efficiency and stirring time. The stirring speed was 600 r/min, with stirring times of 1, 2, 4, 8, 16, and 32 minutes. Efficiency increased slowly after 8

minutes of extraction. After 32 minutes of extraction, the radioactivity of LS decreased by 86.7+0.5-0.5% with 737 - cascade events in LS samples, compared to 5529 events before purification. The errors in Fig. 7 through Fig. 9 represent statistical errors.

Fig. 8 [Figure 8: see original paper] shows the relationship between extraction efficiency and extraction stage. The extraction time was 3 minutes and the stirring speed was 1200 r/min. After 5 extraction stages, the purification efficiency became nearly stable, reaching a modest efficiency of 92.1+0.3-0.4%. Since Pb is a highly polar atom, its affinity for water was expected to be several orders of magnitude higher than for LS. The most likely explanation for the limited efficiency is that a fraction of Pb was bound in a non-polar configuration, reducing the partitioning coefficient and thus the purification efficiency [9].

Fig. 9 [Figure 9: see original paper] shows the relationship between extraction efficiency and the volume ratio of LS to water. In laboratory measurements, purification efficiency decreased slowly when the LS-to-water ratio exceeded 6, then decreased sharply once the ratio reached 6. Based on these results, the LS-to-water volume ratio for JUNO purification can be set at 5.

The water extraction efficiency for 212Pb can exceed 84% using the laboratory-scale purification setup. Both the extraction stage and LS-to-water volume ratio can be set at 5 for future large-scale purification plant design and operation.

4 Conclusions

To study water extraction for future JUNO LS purification plants, an extraction prototype was constructed, and background-free efficiency measurement was achieved using 220Rn-loaded LS. The measured water extraction efficiency for 212Pb was no less than 84%, reaching the world average level, and optimized operation parameters were obtained. Based on these laboratory results, a medium-scale water extraction tower has been built. The radioactivity loading setup and - counting system will be valuable for investigating parameters relevant to large-scale purification plants in the future.

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Note: Figure translations are in progress. See original paper for figures.