Laboratory measurement of radioactivity purification for ^{212}Pb in liquid scintillator *

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Abstract: Liquid scintillator (LS) has been widely used in past and running neutrino experiments, and is expected also to be used in future experiments. Requirements on LS radio-purity have become higher and higher. Water extraction is a powerful method to remove soluble radioactive nuclei, and a mini-extraction station has been constructed. To evaluate the extraction efficiency and optimize the operation parameters, a setup to load radioactivity to LS and a laboratory scale setup to measure radioactivity using the ^{212}Bi-^{212}Po-^{208}Pb cascade decay have been developed. Experience from this laboratory study will be useful for the design of large scale water extraction plants and the optimization of working conditions in the future.

Keywords: liquid scintillator, radioactive load, radioactive measurement, cascade decay, water extraction

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1 Introduction

Liquid scintillator (LS) plays a very important 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 is filled with LS of 20 ktons fiducial mass. To suppress the accidental background, as well as to achieve the potential goal of solar neutrino studies, the basic radioactivity contamination requirement for the JUNO LS is 10^{-15} g/g (in this paper, g/g means gram of ^{232}Th or ^{238}U per gram of LS) for both ^{238}U and ^{232}Th.

The general methods to remove radioactive contamination are water extraction, nitrogen stripping and distillation, which are sensitive to soluble nuclei, Rn and insoluble nuclei, respectively. Before the mass production of purified LS, each method should have a prototype and optimized operation parameters. For previous experiments such as Borexino (a solar neutrino experiment, using trimethylbenzene as the LS solvent), which holds the record for lowest radioactive contamination with ^{232}Th/^{238}U of 10^{-18}g/g [1], the parameters of large scale purification plants and their prototypes are consistent [2, 3].

Though these purification methods are efficient for the Borexino LS, they need to be carefully studied for JUNO's linear alkyl benzene (LAB) -based LS. A water extraction prototype has been constructed at the Institute of High Energy Physics (IHEP), Beijing, to validate the prototype and optimize operation parameters. LS radioactivity before and after purification should be measured. However, the typical U and Th contaminations in LS are 10^{-13} to 10^{-14} g/g, and it is impossible to measure such low radioactivity in the laboratory. A general method is to load radioactive nuclei, such as ^{222}Rn or ^{220}Rn, to the LS, and purify the LS with the prototype, then measure the purified and un-purified LS with a clean detector.

In this paper, the Rn loading technology, the radioactivity measurement setup, and the water extraction efficiencies, as well as the optimized operation parameters are reported. The efficiency has reached the world average level, indicating the prototype is successfully working, and the optimized parameters are useful to future middle-scale and mass production plants.

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2 Radioactivity measurement setups

2.1 ^{220}Rn loading

The limit of radioactivity measurement in the laboratory is 10^{-9} g/g. The natural contamination of LS is 10^{-13} to 10^{-14} g/g, however, so it is impossible to measure such low radioactivity in the laboratory. In order to study the effect of LS purification laboratory experiments, the only solution is an artificial pollution of the samples with radioactivity. Since powder radio-sources do not dissolve in LS and the solubility of liquid radio-sources in LS is not high, the general method is to load radon to LS. Because radon is a non-polar gas, the solubility of radon in LS is high, about 13 times the radon concentration in air at room temperature [4]. Therefore, it is effective to load radioactivity in LS by bubbling radon into the LS.

The commonly used radon is ^{222}Rn, which is from the ^{238}U decay chain and has a 3.8 day half-life. For example, in the Borexino experiment, ^{222}Rn was loaded to LS and the contamination of its daughter ^{210}Po was measured. The disadvantage is that it requires months for ^{210}Po to accumulate to a measurable amount, since its mother ^{210}Pb's half-life time is 22.3 years [5]. Besides, the long half-life time of ^{210}Pb would pollute the experiment setup.

Compared with ^{222}Rn, a better candidate is ^{220}Rn, which is from the ^{232}Th decay chain as shown in Fig. 1, and has a 55 s half-life. After loading, ^{220}Rn quickly decays to ^{212}Pb with a 10.6 hour half-life. The decay of ^{212}Pb's daughters ^{212}Bi and ^{212}Po are well-known cascade decays (β-α cascade decay), since the half-life of ^{212}Po is only about 300 ns. The cascade decay supplies a pair of time correlated signals in our experiments, with high efficiency and extra low background. With ^{220}Rn loading, the water extraction efficiency is estimated with the nucleus ^{212}Pb. The 10.6 hours half-life of ^{212}Pb is long enough to do extraction and measurement, and it will not cause any contamination to the experiment setup. Meanwhile, a large amount of ^{220}Rn in LS will decay to ^{212}Pb in a very short period of time, leading to high radioactivity loading efficiency.

The loading method used in the paper was bubbling ^{220}Rn into the LS sample. The corresponding setup inside a glove box is shown in Fig. 2. A ^{232}Th source releasing 1200 Bq ^{220}Rn produced by Nanhua University was used. Nitrogen went through a bubbler filled with water, then it blew through the source and took ^{220}Rn out. Finally it went through a bubbler filled with LS. According to research conducted by Nanhua University, the ^{220}Rn release rate of ^{232}Th source increases with higher environmental humidity. After bubbling ^{220}Rn into the LS for 74.2 hours, the concentration of ^{212}Pb reached equilibrium. In the following study, LS was bubbled with ^{220}Rn for 20 hours, which reached 2/3 of the ^{212}Pb concentration equilibrium [6].

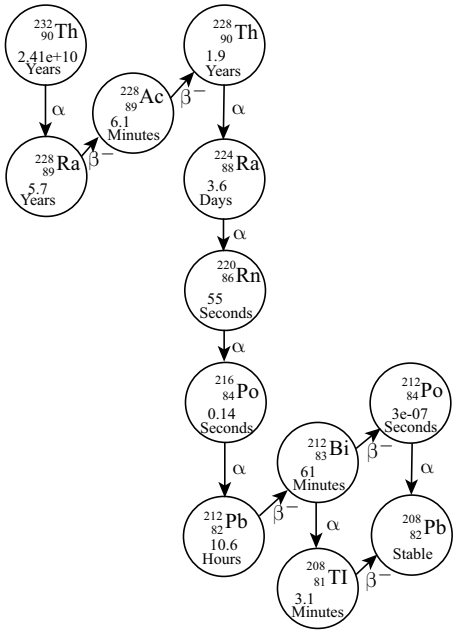

Fig. 1. (color online) The natural decay chain of ^{220}Rn.

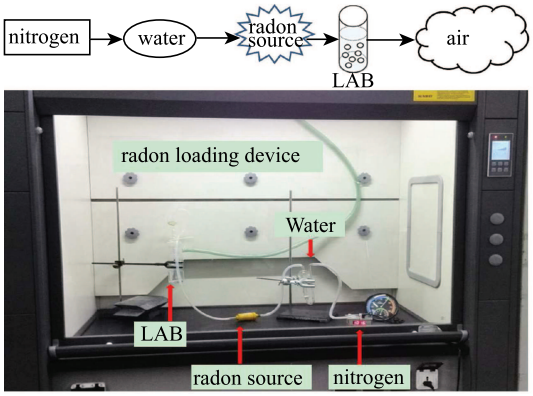

Fig. 2. (color online) Laboratory setup for radon-loading of LS samples.

The radioactivity was measured by the experimental setup depicted in Fig. 3. In a light-tight box, a pair of 2" PMTs (XP2020) was placed on both sides of an LS sample cell, to do double coincidence measurement. The coincidence measurement is to reduce the influence of fluctuations in a single PMT on the experimental data. The LS container was a cylindrical quartz glass bottle, with 5 cm diameter and 1.5 cm thickness, and a capacity of 17.1 g LS. Gamma rays from ambient radioactivity were attenuated by shielding with low-activity lead bricks [4]. A flash ADC (DT5751 made by CAEN with 1 GHz sampling frequency) was used for data acquisition. The total background event rate (including β, α, γ and cascade decay events) during measurement was 0.25 Hz.

After finishing the data acquisition for all events, β-α cascade events were picked out by offline analysis. This setup was designed as a β-α counting system.

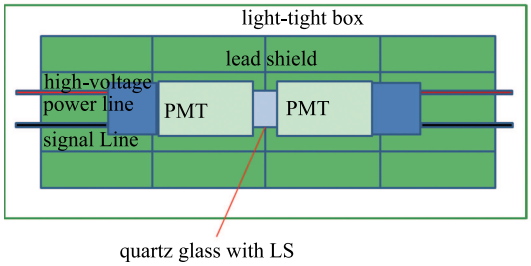

Fig. 3. (color online) Laboratory setup for measuring the efficiency of radiopurification.

3 Data analysis

3.1 The β-α cascade event selection

3.1.1 Real β-α cascade event

After ^{220}Rn loading, three hours' data was taken to determine the initial ^{212}Pb concentration. The coincidence time window for signals from the two PMTs was required to be smaller than 5 ns, due to the length difference of the cables connected to the two PMTs. 99.99% of the β-α cascade events met this requirement, with a statistical error of 9.55×10^{-8} (statistical error will not be discussed in this section since it was too small).

The time interval distribution between the β decay and α decay of the β-α cascade events is shown in Fig. 4. The distribution can be described by the formula below [7],

$$f(t)=\frac{1}{\tau}\times N_0\times \mathrm{e}^{-\frac{t}{\tau}}, \tag{1}$$

where τ is the lifetime of ^{212}Po and N_0 is a parameter related to the concentration of ^{212}Po. $T_{1/2}$ is the half-life of ^{212}Po and $T_{1/2}=\ln2*\tau$. Hence, the formula above can be written as,

$$f(t)=\frac{\ln2}{T_{1/2}}\times N_0\times 2^{-\frac{t}{T_{1/2}}}. \tag{2}$$

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

$$f(t)=\frac{1}{p_1}\times p_0\times 2^{-\frac{t}{p_1}}. \tag{3}$$

The parameter p_1 in the fitting result stands for the half-life of ^{212}Po. In theory, the half-life is 298 ns, while the experimental result was 298.4±1.2 ns. Therefore, the β-α counting setup was reliable for detecting β-α cascade events.

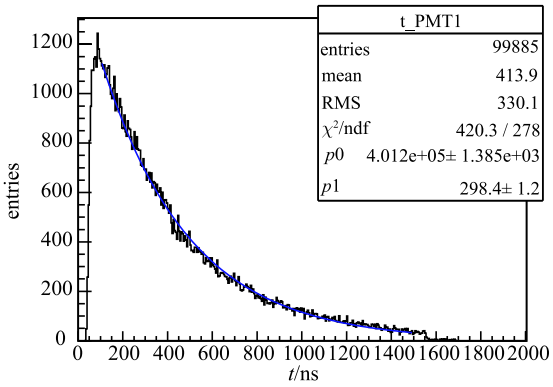

Fig. 4. (color online) The time interval distribution between the β decay and α decay.

For each β-α cascade event, there was a time interval between the β event and the α event. Theoretically, the time interval measured by the two PMTs should be the same. At least, the difference due to differences between the two PMTs or other experimental effects should be very small. Fig. 5(a) shows the difference of time interval of β-α cascade events detected by the two PMTs, which was within the coincidence time window. The number of entries before normalization was 104640. A Gaussian function was used to fit the distribution and the result was a mean value of 0.51 and σ of 0.91. The difference of the time interval was mostly within (−2 ns, 3 ns). 95.92% of the events met this requirement, which was consistent with the probability of the variable from the Gaussian distribution being within the 2 sigma range.

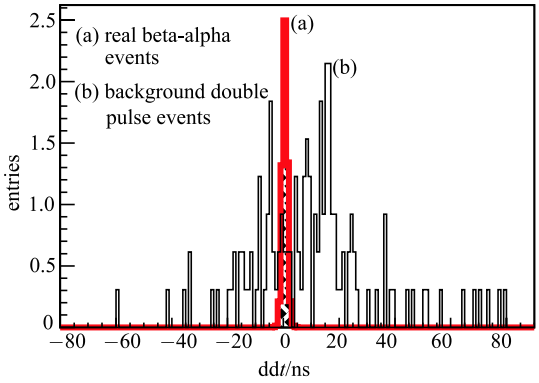

Fig. 5. (color online) Difference of time interval of β-α pulse between the two PMTs (entries are normalized).

The event energy was proportional to the total charge collected. Measurement of the total charge from the PMT was estimated by integrating the entire pulse. Fig. 6(a) shows the integral value of the α event pulse (the second pulse in a double-pulse event). Due to the mono-energy of α events, the distribution was centralized like a Gaussian distribution. 92.80% of the second pulse integral value was within (1700 FADC, 8000 FADC) after the two selection criteria discussed above.

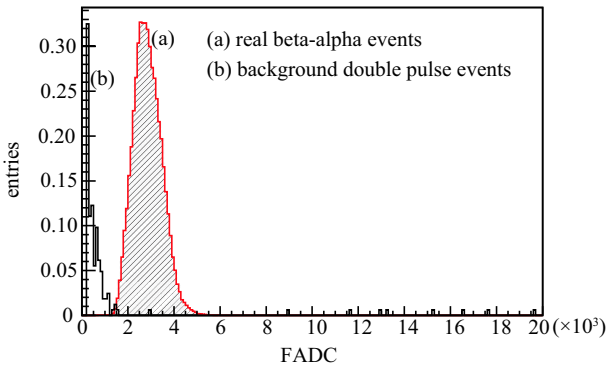

Fig. 6. (color online) The integral value of the second pulse (entries are normalized).

3.1.2 Cuts selection

Based on the analysis of real β-α events, several cut criteria were selected.

(1) The coincidence time window of the two PMTs was required to be smaller than 5 ns;

(2) The difference of time interval of β-α cascade events detected by the two PMTs was within (-2 ns, 3 ns);

(3) The integral value of α events was within (1700 FADC, 8000 FADC).

After bubbling ^{220}Rn into LS for 20 hours, there were about 2×10^4 β-α events detected in 17.10 g LS in 30 minutes' data taking after using the cut criteria discussed above.

3.1.3 Background events

Background events were taken for one day using the pure LS (17.10 g) without loading ^{220}Rn to it. 163 double pulse events were found. However, the integral value distribution of the second pulse (maybe a fake alpha event pulse) and the difference of time interval of the double pulse events detected by the two PMTs were very different from real β-α events, as shown in Fig. 5(b) and Fig. 6(b). The integral values of the second pulses were mostly less than 1700 FADC and only a small percentage of the difference of the time interval was within (−2 ns, 3 ns). After applying these cut criteria to the background events, there remained only 2 background β-α cascade events a day, while there were 163 without cut selections.

Compared with 2×10^4 real β-α cascade events detected in ^{220}Rn loaded LS in 30 minutes, the 2 background events in one day can be neglected in the following study.

3.2 Study of water extraction

Purification by water extraction relies on the polarity of water molecules to separate polarized impurities, e.g. free-state ions of radioactive metals, from the non-polar LAB and fluor molecules. Water extraction is very effective for most ionic metals such as K, Ra, and Bi, but is less effective for Po and Pb. For Po and Pb, the reduction was seen to be equally fast but less effective with an 82%−87% removal fraction in a SNO+ laboratory study [8, 9].

After the scintillator reached a good ^{212}Pb concentration, the scintillator was purified by water extraction. The purification efficiency is defined as

$$u=\frac{y-x}{y}=1-\frac{x}{y}. \tag{4}$$

Here, x means number of events after purification, y means number of events before purification and u means purification efficiency.

The statistical error of efficiency is calculated by a Clopper-Pearson parameter estimation of a binomial distribution [10]. The formula is:

$$\sigma_+=\left(1+\frac{n-\hat{s}}{\hat{s}+1}f_{1-\alpha/2}(2(n-\hat{s}),2(\hat{s}+1))\right)^{-1}-\hat{p}, \tag{5}$$

$$\sigma_-=\hat{p}-\left(1+\frac{n-\hat{s}+1}{\hat{s}}f_{\alpha/2}(2(n-\hat{s}+1),2\hat{s})\right)^{-1}. \tag{6}$$

Here, $1-\alpha$ is the confidence level; n is the total number of events; $f_{\alpha/2}$ is the upside $\alpha/2$ fractile of F-distribution; $\hat{s}$ is the passed number of events and $\hat{p}=\hat{s}/n$. In this paper, $n=y$, $\hat{s}=y-x$ and the confidence level is set as 0.683.

Water extraction was done with equal amount of 12 MΩ deionized water and liquid scintillator. A separatory funnel and a magnetic stirrer were used to mix 35 ml of water and 35 ml of scintillator. The solution was mixed and then separated. The operation was called one stage extraction. After each separation a clean separatory funnel and fresh deionized water were used to do multiple stage extraction. Scintillator samples were placed in small test tube which contained 17.1 g of LS sample. Then the β-α counting system was used to measure the radioactivity in the scintillator before and after purification.

A stability study of the β-α counting system was conducted by measuring the purification efficiency at three different times with the same LS sample. The efficiencies were consistent with each other, being $84.3^{+1.2}_{-1.3}$%, $82.7^{+1.5}_{-1.6}$% and $83.3^{+1.8}_{-1.9}$%. Therefore, it was reliable to optimize purification parameters by using the β-α counting system.

Figure 7 shows the relationship between extraction efficiency and stirring time. The stirring speed was 600 r/min, while the stirring times were 1 min, 2 min, 4 min, 8 min, 16 min and 32 min. The efficiency increased slowly after extraction for 8 min. When extracted for 32 min, the radioactivity of LS decreased $86.7^{+0.5}_{-0.5}$% with 737 β-α cascade events in LS samples. Before purification there were 5529 β-α cascade events. The error in Fig. 7 to Fig. 9 is statistical error.

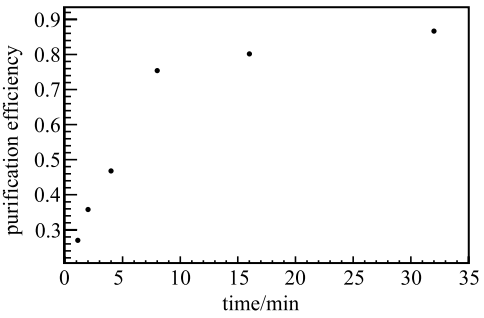

Fig. 7. Purification efficiency vs. extraction time, 600 r/min stirring speed.

Figure 8 shows the relationship between extraction efficiency and the extraction stage. The extraction time was 3 min and the stirring speed was set at 1200 r/min. After extraction for 5 stages, the purification efficiency became almost stable, reaching a not very high efficiency of $92.1^{+0.3}_{-0.4}\%$. Since Pb is a very polar atom, it was expected that its affinity to water would be higher than that to LS by several orders of magnitude. The most likely explanation is that a fraction of the Pb was bound in a nonpolar configuration which reduced the partitioning coefficient and thus the purification efficiency [8].

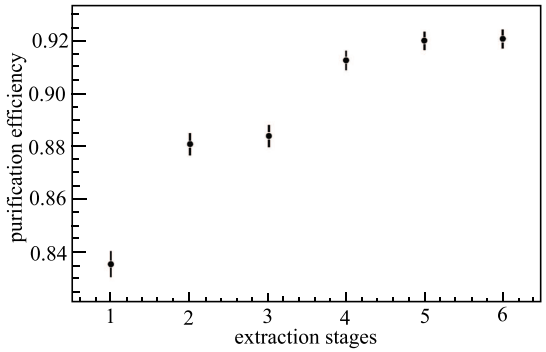

Fig. 8. Purification efficiency vs. extraction stages, 3 min extraction time and 1200 r/min stirring speed.

Figure 9 shows the relationship between extraction efficiency and the volume proportion of LS to water. In laboratory measurement, the purification efficiency decreased slowly when the proportion of LS to water was larger than 6. Then the efficiency decreased sharply as soon as the volume proportion reached 6. According to this result, the volume proportion of LS to water used in JUNO purification can be set at 5.

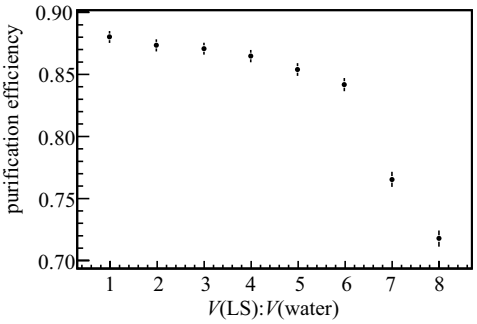

Fig. 9. Purification efficiency vs. volume proportion of LS and water, 10 min extraction time and 800 r/min stirring speed.

The water extraction efficiency for ^{212}Pb can reach more than 84% with laboratory scale purification setup. The extraction stage and volume ratio of LS to water can be set at 5 in future large scale purification plant design and operation.

4 Conclusions

To study the water extraction in the future JUNO LS purification plants, an extraction prototype has been constructed, and a background free efficiency measurement had been achieved with ^{220}Rn-loaded LS. The measured water extraction efficiency for ^{212}Pb was about 84%, reaching the world average level, and optimized operation parameters have been obtained. Now, a medium scale water extraction tower has been built based on the laboratory study results. The radioactivity loading setup and β-α counting system will be useful for the investigation of the parameters involved in large scale purification plants in the future.

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