Temperature dependence of the light yield of the LAB-based and mesitylene-based liquid scintillators[*]

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Abstract: We studied the temperature dependence of the light yield of linear alkyl benzene (LAB)-based and mesitylene-based liquid scintillators. The light yield increases by 23% for both liquid scintillators when the temperature is lowered from 26 ℃ to −40 ℃, correcting for the temperature response of the photomultiplier tube. The measurements help to understand the energy response of liquid scintillator detectors. Especially, the next generation reactor neutrino experiments for neutrino mass hierarchy, such as the Jiangmen Underground Neutrino Observatory (JUNO), require very high energy resolution. As no apparent degradation on the liquid scintillator transparency was observed, lowering the operation temperature of the detector to ∼4 ℃ will increase the photoelectron yield of the detector by 13%, combining the light yield increase of the liquid scintillator and the quantum efficiency increase of the photomultiplier tubes.

Key words: liquid scintillator, reactor neutrino, linear alkyl benzene, mesitylene

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1 Introduction

Organic liquid scintillators (LS) are widely used to detect reactor neutrinos [1–5] due to their high light yield and high hydrogen fraction. A liquid scintillator is made of a solvent and a small amount of fluor, and often has an additional tiny amount of wavelength shifter. For example, the Daya Bay undoped liquid scintillator consists of linear alkyl benzene (LAB) as the solvent, 3 g/L 2,5-diphenyloxazole (PPO) as the fluor, and 15 mg/L p-bis-(o-methylstyryl)-benzene (bis-MSB) as the wavelength shifter, while the gadolinium-doped LS has the same recipe but mixed with a Gd complex with 0.1% Gd in mass [6, 7].

The energy response of the liquid scintillator detector need be well understood for precision measurements in a reactor neutrino experiment. The light yield of the liquid scintillator, to which the visible energy of an event in the detector is proportional, is, however, temperature dependent because of the thermal quenching effects. Ex-

cited solvent molecules by ionization may undergo non-radiation transition when colliding with other molecules. Normally the light yield will increase at lower temperature when the viscosity of the solvent rises, thus collisions reduce. If a quencher is present in the solution, then the situation will be complex since the collisions may also impede the energy transfer from the exited solvent molecule to the quencher.

The temperature dependence of the light yield has been studied for some liquid scintillators [8] but is still scanty in the literature. In this study, we will study the temperature effects of the Daya Bay LS, which is based on the relative new solvent LAB, over a range from −40 ℃ to 26 ℃. As a comparison, LS with the same solute fractions but another solvent, mesitylene, is also measured.

Such studies are of particular interests for the design of the next generation reactor neutrino experiments, such as the Jiangmen Underground Neutrino Observatory (JUNO). To determine the neutrino mass hierarchy

by precisely measuring the energy spectrum of the reactor neutrinos, the JUNO detector requires a very high energy resolution of $3\%/\sqrt{E(\text{MeV})}$ [9]. Previous experiments reached $(5–6)\%/\sqrt{E}$ [1, 5]. This unprecedented energy resolution requirement is a major challenge for JUNO. The increasing light yield of the LS at lower temperatures provides an option to operate the detector at low temperature, for example, at ~ 4 ℃, right above the ice point of the buffer water shielding of the neutrino detector. For this purpose, we also studied the light transmittance of the LS at low temperature.

In this paper, the experimental setup, light yield measurement, and correction for the temperature effects of the photomultiplier tube (PMT) are described in Section 2. The transmittance is studied in Section 3, followed by a conclusion and discussion.

2   Temperature dependence of the light yield

2.1   Experimental setup

The light yield of the LS is measured via Compton scattering of γ rays from a radioactive source. To improve the precision of the measurement, we tag the scattered γs at a fixed direction. The coincidence of the recoil electron in the LS and the scattered γ selects the events of known deposited energy in the LS, and thus reduces the uncertainty of the light yield measurement to sub-percent level from $\sim 5\%$ of the common method by fitting the Compton edge.

Figure 1 shows the scheme of the experimental setup. The LS sample is contained in a cylindric quartz glass vessel of 5 cm in diameter and 5 cm in height. The vessel is wrapped with an Enhanced Specular Reflector (ESR) to increase the photon collection efficiency, and is coupled to a CR135 PMT. The scattered γ is tagged by a coincidence detector, which is a plastic scintillator (PS) detector located about 10 cm from the LS at an angle of about 30°. The LS is irradiated by γ-rays from a ^{137}Cs (15 μCi) source. The source, the LS vessel, the CR135 PMT, and the coincidence detector are put in an enclosed thermostat with temperature adjustable from -70 ℃ to 155 ℃. The signals from the CR135 PMT and the coincidence detector are recorded by a CAEN DT5751 ADC unit with a self-trigger function. The relative light yield of the LS is determined by comparing the peak values measured by the ADC at different temperatures.

To separate the temperature effects of the PMT from that of the LS, a temperature-resistant optical fibre is directly coupled to the photocathode of the CR135 PMT, transmitting light from an LED driven by a pulse generator. The LED is located in an incubator box operating at 25 ℃, emitting light at 430 nm wavelength. The pulse generator is at room temperature, which is almost

a constant during the measurement. The LED flashes at a frequency of 10 kHz during the LS light yield measurement and the data is triggered by the coincidence with the PS detector (noise). The probability of the LED signal overlapping with a Compton signal is found to be small enough.
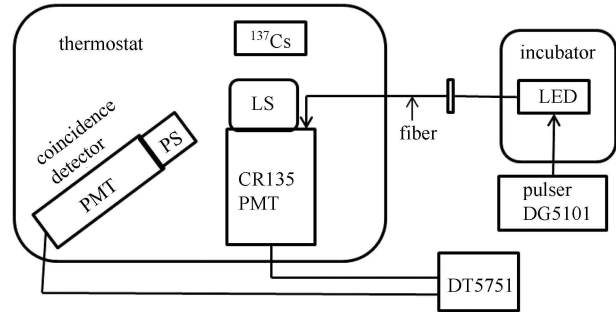

Fig. 1.   Experimental setup for measuring the temperature dependence of the LS light yield.

The LS sample is bubbled with nitrogen from the bottom of the vessel before measurement to remove oxygen and water in the LS. Oxygen is a quencher of the LS. The presence of oxygen reduces the light yield of the LAB-based Daya Bay LS by up to 11% [10], shortens the time constant of the scintillation [11], and may change the temperature effects of the LS. Normally, the LAB-based LS contains tens of ppm water. To avoid possible impacts on the light transmittance of the LS at low temperature, water is also removed by bubbling nitrogen. The LS sample is covered with nitrogen during the measurement. A temperature sensor is mounted on the LS vessel for monitoring. Data are taken only after the temperature has reached stability for more than 30 minutes.
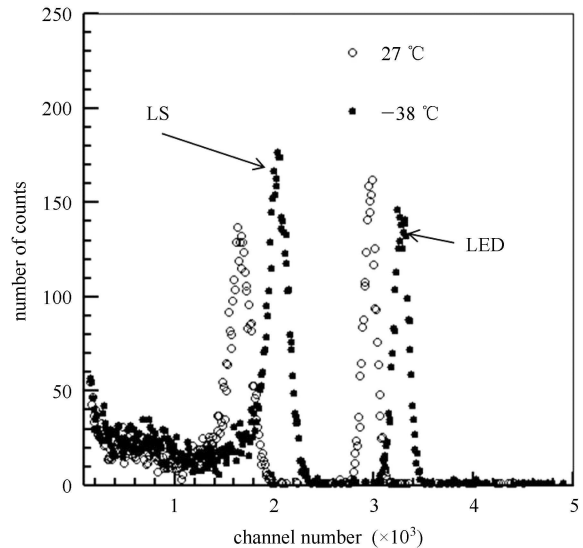

Fig. 2.   ADC channel distribution of the LS events and the LED monitoring signal.

2.2   Relative light yield of the LS

Figure 2 shows an example of the ADC distribution of the measurement. The light yield of the LS and the light intensity of the LED are measured by the ADC peaks, which are fitted with Gaussian functions. The LED intensity is stable to 1% at constant temperature. The observed shift at different temperatures shows that the CR135 PMT response suffers from the temperature variation.

Figure 3(a) shows the relative light yield of the LAB-based LS before correcting for the PMT temperature effects. The measurements have been done three times by different people and with slightly different hardware. The data labeled "Third" corresponds to this measurement and the other two were done before. The three measurements are in good agreement. The temperature

response of the CR135 PMT monitored by the LED is shown in Fig. 3(b).

A similar measurement was done for the mesitylene-based LS. After correcting for the measured temperature response of the PMT shown in Fig. 3(b), the temperature dependence of the LS light yield is shown in Fig. 4. The light yield increases by 23% as the temperature decreases from 26 ℃ to −40 ℃ for both liquid scintillators.

Fig. 4.   The relative light yield of the LAB-based LS (LLS) and the mesitylene-based LS (MLS) after correcting for the PMT temperature effects, normalized at 26 ℃.

The variation of the PMT response at difference temperature might be a combination of the variation of the PTM gain and the quantum efficiency changes of the PMT photocathode. The PMT gain decreases as the temperature increases because of the negative temperature coefficient of the dynodes [12]. Quantum efficiency of the PMT is typically 25% at 430 nm at room temperature for the CR135, which uses a bialkali (SB-K-Cs) photocathode. The quantum efficiency of the bialkali has almost a constant temperature coefficient of −0.2%/℃ for photons of wavelength between 200 nm and 550 nm [13]. This will increase by 13% when the temperature is lowered from 26 ℃ to −40 ℃. Therefore, the quantum efficiency increase dominates the PMT temperature effects that we have measured. In this measurement, the temperature stability of the temperature-resistant optical fibre is estimated to be 0.5% and the stability of the LED intensity is about 1%.

3   Temperature dependence of the light transmittance

3.1   Experimental setup
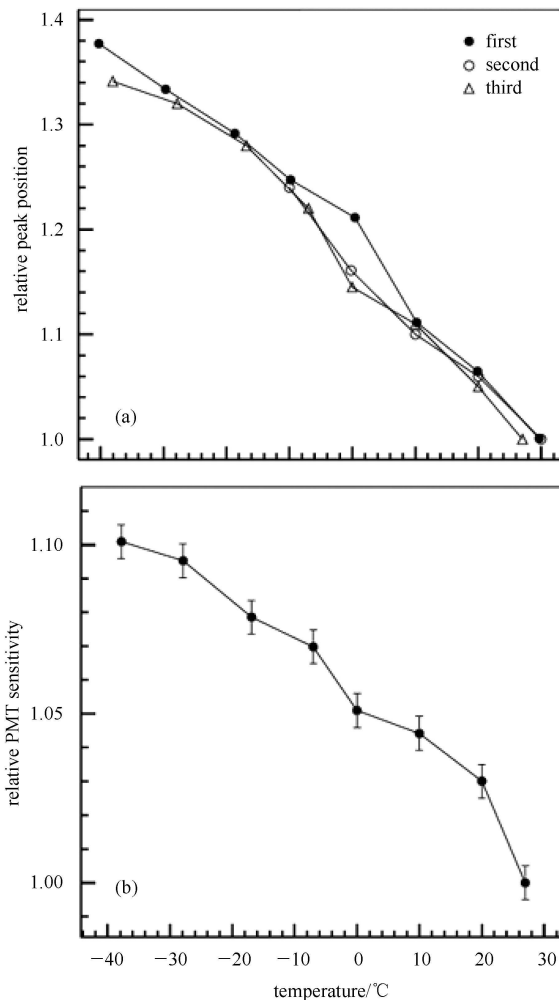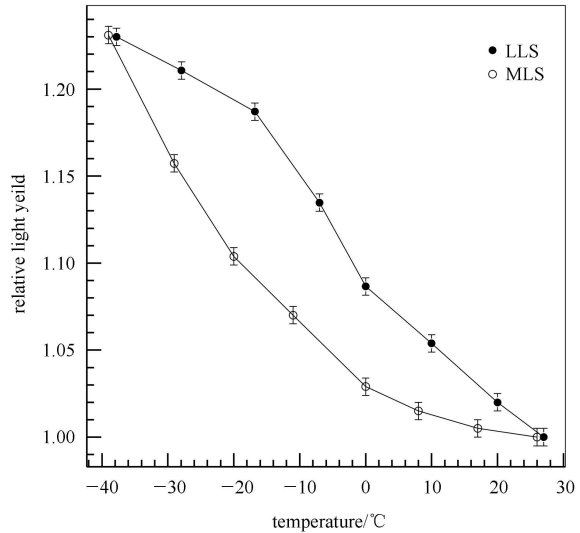
For JUNO's large detector of ∼38 m diameter the

Fig. 3.   (a) The relative light yield of the LAB-based LS before correcting for the PMT temperature effects, normalized at 26 ℃. (b) The temperature response of the CR135 PMT, normalized at 26 ℃.

light transmittance of the LS is as equally important as the light yield. The temperature dependence of the light transmittance is measured with 430 nm wavelength light, over a range from −40 ℃ to 26 ℃.

The scheme of the experimental setup is shown in Fig. 5. The light source is a DH-2000 deuterium tungsten halogen lamp, which is followed by a monochromatic filter at 430 nm. The light transmits in a temperature-resistant fibre, passes the LS sample in a cuvette of dimension of 1 cm×1 cm×3 cm, and is received by an Ocean Optics QE65000 spectrophotometer. A temperature probe is attached to the cuvette to monitor the LS temperature. The spectrum of the light is shown in the bottom panel of Fig. 5, and was measured by the spectrophotometer. The relative transmittance can be obtained by continuously measuring the light intensity as temperature varies.
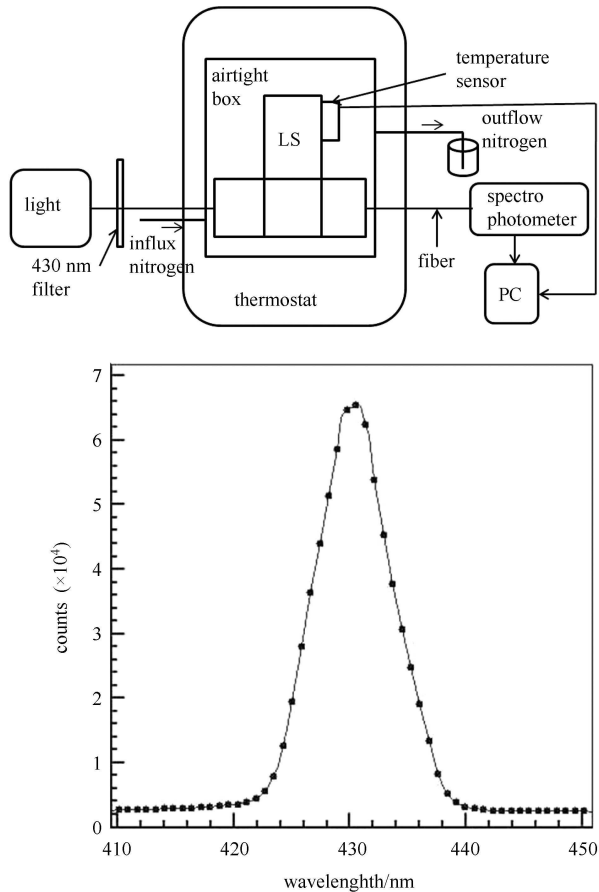

Fig. 5.   Top: Schematic view of the experimental setup for the light transmittance measurement. Bottom: The measured light spectrum after passing the monochromatic filter.

The surface of the cuvette may frost and bias the transmittance measurement when the temperature goes down. We observed such phenomenon when the trans-

mittance started to drop dramatically at a certain temperature (e.g. −7 ℃ for one of our measurements of LAB-based LS), although it is not visible on the surface of the cuvette by eye. After excluding the possible causes, such as the crystallization of water content in the LAB, precipitation of scintillation fluor, and freezing of the solvent itself, we improved the experimental setup by sealing the LS cuvette in a transparent airtight box to guard against the frost. The box is flushed with dry nitrogen before the experiment to remove the vapor in the air, and we maintained a small positive pressure with nitrogen. The nitrogen is released from the box through a bubbler to monitor the airtightness of the box.

3.2   Relative transmittance of the LS

A liquid scintillator that is exposed to air normally contains water of tens of ppm. Water crystals could be formed in the LS and degrade the transmittance at low temperature. Water can be removed from the LS thoroughly by bubbling with enough dry nitrogen. The nitrogen bubbling is also necessary to purge the oxygen in the LS. The water content in the LS can be measured to below 1 ppm with a 831KF Coulometric Moisture Analyzer. Table 1 shows the relationship between the remaining water content and the volume of nitrogen flushed. Three litres of nitrogen is needed to purge all of the water in the cuvette.

Table 1.   Relationship between the water content in the LS samples and the volume of nitrogen flushed.

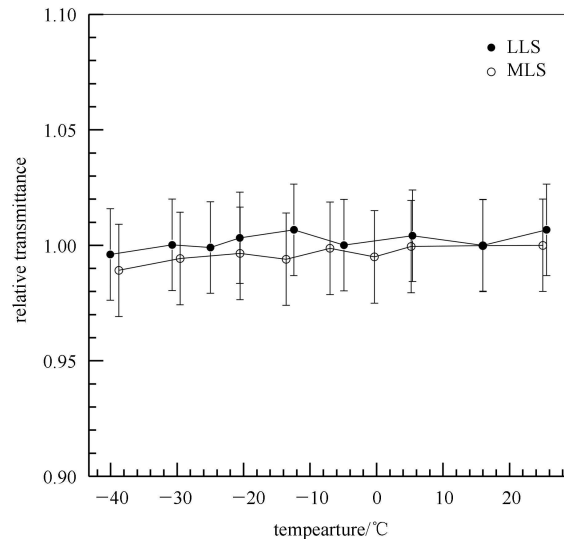| LS	0 L	1 L	3 L
LLS	∼27 ppm	∼16 ppm	∼0 ppm
MLS	∼140 ppm	∼70 ppm	∼0 ppm

Fig. 6.   Relative transmittance of the LAB-based LS (LLS) and the mysitylene-based LS (MLS) for light of 430 nm wavelength.

Figure 6 presents the result of the relative transmittance for the LAB-based and the mysitylene-based LS. We use the temperature instability of the system without LS in the cuvette as the systematic error of the measurement. For both the LAB-based and the mysitylene-based LS, transmittance stays stable when the temperature decreases from 26 ℃ to −40 ℃. The melting point of the LAB we used is below −60 ℃ and that of the mesitylene is −44 ℃. Lowering the temperature to −40 ℃ will not cause a phase transition of the LS.

4   Conclusion and discussion

The temperature dependence of the light yield and the transmittance of two liquid scintillators, LAB-based LS and mysitylene-based LS, have been measured. For both liquid scintillators, when the temperature is lowered from 26 ℃ to −40 ℃, the light yield increases by 23%, with the PMT effects corrected, and the transmittance remains stable. Frosting on the sample vessel at low temperature is observed to have significant impacts on the measurements. This is avoided by coupling the PMT to the sample vessel with silicone oil for the light yield measurement, or by putting the vessel in a nitrogen purged transparent box for the transmittance measurement.

The light yield increase at low temperature provides an option for the next generation reactor neutrino experiments for neutrino mass hierarchy, such as JUNO, which requires very high energy resolution. When using water as an outer buffer, the operation temperature of the detector could be lowered to ~ 4 ℃. As no apparent degradation on the liquid scintillator transparency was observed, lowering the operation temperature to ~ 4 ℃ from 26 ℃ will increase the photoelectron yield of the detector by 13%, in which 9% comes from the light yield of the LAB-based liquid scintillator and $\sim 4\%$ comes from the quantum efficiency of the PMT with bialkali photocathode.

Operating at even lower temperatures, such −40 ℃, will increase the photoelectron yield by 32%. But this is more difficult to realize and requires an oil buffer instead of a water buffer.

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