

Oxygen quenching in a LAB based liquid scintillator and the nitrogen bubbling model^{*}

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Abstract The oxygen quenching effect in a Linear Alkyl Benzene (LAB) based liquid scintillator (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 λ -shifter) is studied by measuring the light yield as a function of the nitrogen bubbling time. It is shown that the light yield of the fully purged liquid scintillator is increased by 11% at room temperature and the room atmospheric pressure. A simple nitrogen bubbling model is proposed to describe the relationship between the relative light yield (oxygen quenching factor) and the bubbling time.

Key words Linear Alkyl Benzene, oxygen quenching, nitrogen bubbling, liquid scintillator

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1 Introduction

It has been shown experimentally and theoretically that the presence of oxygen in a liquid scintillator (LS) can lower the light yield, modify the fluorescence pulse shape, shorten the attenuation length and decrease the positron annihilation lifetime [1–3]. For most aromatic molecules, the quenching of the electronically singlet state (S_1) leads to the formation of a triplet state (T_1). The oxygen molecule is somewhat special. Its ground state is a triplet and the next state is a singlet lying about 0.98 eV over the ground state [4]; oxygen molecules in aromatic molecules can absorb the energy of the singlet state of aromatic molecules and make a spin allowed transition to the triplet state. This decreases the fluorescence. Such a transition only occurs in aromatic molecules which have an energy gap of S_1 - T_1 greater than 0.98 eV. Most aromatic molecules meet this requirement. In most experiments, the dissolution of oxygen into the LS is undesirable, since this brings uncertainties to the experiments. Hence, the oxygen quenching effect should be well studied and determined.

Usually, there are three ways to eliminate the dissolved oxygen from solutions: 1) the ultrasonic [5]; 2) the vacuum distillation; 3) the nitrogen(or argon)

bubbling [2]. In neutrino experiments, a large quantity of liquid scintillators is required. The most economical and practicable way to eliminate oxygen in LS is the nitrogen (or argon) bubbling.

Linear alkyl benzene (LAB), which is composed of a linear alkyl chain of 10–13 carbon atoms attached to a benzene ring, is a low cost product of the petrochemical industry and is often used as the material of detergent. Its aromatic structure makes it useful as a scintillator solvent. It has many appealing properties, including the high flash point (130 °C), the low toxicity, the high light yield and excellent transparency [6]. A LAB based liquid scintillator will serve as the antineutrino target in the Daya Bay neutrino experiment [7].

In this work we measured the effect of oxygen on the light yield of the LAB LS, and built a nitrogen bubbling model to describe the relationship between the relative light yield (the oxygen quenching factor) and the nitrogen bubbling time. Parameters in the model were determined by our experimental data.

2 The nitrogen bubbling model

When an LS is exposed to the air, oxygen molecules dissolved in the LS exchange with those

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in the air. This process is in dynamical equilibrium. It is reasonable to assume that the oxygen dissolved in the un-bubbled LS is saturated due to its long time contact with the air. This means that the number of oxygen molecules dissolving into the LS is equal to that escaping from the LS. When the LS is flushed with nitrogen, the oxygen partial pressure in the nitrogen bubbles present in the LS, can be thought to be zero. Therefore, the oxygen molecules will escape from the LS and enter the nitrogen bubbles. The dissolution of oxygen molecules into the LS can be ignored at the interface of nitrogen and the LS. Since the diffusion rate of oxygen molecules into the LS is much higher than the oxygen escaping rate, which denotes that oxygen molecules are uniformly distributed in the LS, it is reasonable to assume that the oxygen escaping rate is proportional to the contact area of the bubbles with the LS and the oxygen partial pressure in the LS. The latter is proportional to the oxygen concentration. Then, the equation describing the variation of the oxygen molecule number in the LS, dN/dt , is given by

$$\frac{dN}{dt} = -k_e[Q]S, \quad (1)$$

where $[Q]$ is the oxygen concentration dissolved in the LS, k_e and S are the oxygen escaping rate and the nitrogen-LS contact area, respectively. The oxygen concentration can be expressed as the following:

$$[Q] = \frac{N}{V_s}, \quad (2)$$

where N and V_s are the oxygen molecule number in the LS and the LS volume, respectively. Hence, Eq. (1) can be rewritten as

$$\frac{dN}{dt} = -\frac{k_e N}{V_s} S. \quad (3)$$

Then, the variation of the oxygen molecule number in the LS is

$$\frac{dN}{N} = -\frac{k_e S}{V_s} dt. \quad (4)$$

Eq. (4) can be written in the integration form:

$$N = N_0 \exp\left(-\frac{k_e S}{V_s} t\right), \quad (5)$$

where N_0 is the oxygen molecule number in LS without bubbling.

At low concentration, the quenching of fluorescence by a quencher in solution can be described by the well-known Stern-Volmer relationship,

$$\frac{I_0}{I} = 1 + k_Q[Q], \quad (6)$$

where I_0 is the intensity or rate of fluorescence with-

out a quencher present, I is the intensity or the rate of fluorescence with a quencher, $[Q]$ is the quencher concentration dissolved in the LS, and k_Q is the quenching constant. Hence, the relative light yield as a function of the bubbling time is given by

$$\frac{I_0}{I} = 1 + \frac{k_Q N_0}{V_s} \exp\left(-\frac{k_e S}{V_s} t\right). \quad (7)$$

Note that N_0/V_s is the saturated concentration of oxygen which depends on the temperature and atmospheric pressure. At fixed temperature and atmospheric pressure, N_0/V_s is a constant. Denote the $k_Q N_0/V_s$ item by a constant A . Then Eq. (7) goes over into

$$\frac{I_0}{I} = 1 + A \exp\left(-\frac{k_e S}{V_s} t\right). \quad (8)$$

The oxygen quenching factor f_Q is defined as the light yield of the LS with oxygen to that without oxygen, i.e. I/I_0 . Eq. (8) can then be expressed in the form of f_Q ,

$$f_Q = 1 / \left(1 + A \exp\left(-\frac{k_e S}{V_s} t\right)\right). \quad (9)$$

The bubbling setup can be illustrated by the simplified plot in Fig. 1. The nitrogen-LS contact area consists of two parts: the area of the nitrogen bubble surface and the contact surface area at the liquid level, i.e.

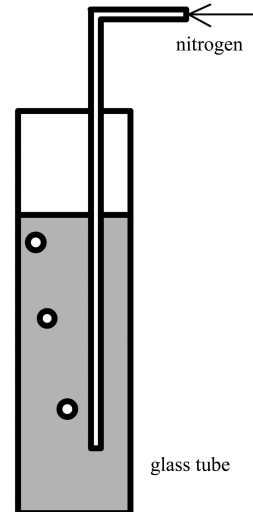


Fig. 1. Bubbling setup.

$$S = nS_b + S_l, \quad (10)$$

where n is the average number of nitrogen bubbles present in the LS. S_b and S_l are the average surface area of the bubbles and the contact surface area at the liquid level, respectively. Then, Eq. (9) can be rewritten as

$$f_Q = 1 / (1 + A \exp(-k_e(nS_b + S_l)t/V_s)). \quad (11)$$

The parameters A and k_e are constants depending on the temperature and pressure for a specific LS. In the following sections we will evaluate A and k_e for a LAB LS at room temperature and atmospheric pressure experimentally.

3 Experiment

In order to observe the light output variation due to the oxygen quenching, sets of LS samples (LAB as the solvent, 3 g/L PPO as the fluor and 15 mg/L bis-MSB as the wavelength shifter) were bubbled with different times. Fig. 1 shows the bubbling setup. Six sets of LS samples (50 ml for each sample) were used. One set of samples was not bubbled and the other five were bubbled with high purity nitrogen for 5 min (200 ml), 12.5 min (500 ml), 18.7 min (750 ml), 25 min (1000 ml) and 31.25 min (1250 ml), respectively (The numbers in the brackets are the nitrogen volumes). The nitrogen flow rate was precisely controlled by a flowmeter to have a constant value of 40 ml/min. The bubbles present in the LS can be thought to be spherical. The bubble number appearing in the tube can be easily counted and the diameters of the bubbles can be measured with rulers. Under our conditions, 4 bubbles, with 4 mm in diameter, were present in the LS. The diameter of the tube was 23 mm. Hence, the LS–nitrogen contact area was 616.5 mm².

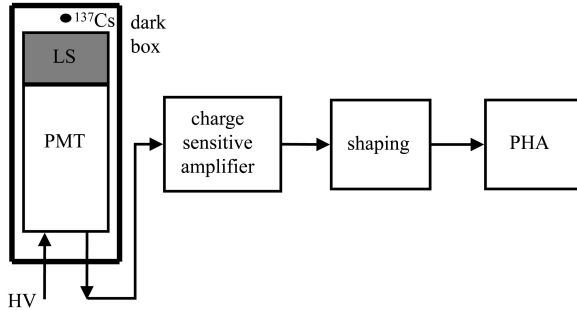


Fig. 2. Schematic view of the experimental setup.

The setup shown in Fig. 2 was used to measure the light output. The LS was encapsulated in a cylindrical teflon cell (5 cm in diameter and 2.5 cm in height). The end of the cell was terminated with an UV glass which was coupled to a 2-inch high energy resolution photomultiplier tube (PMT). The cell and the PMT were placed in a dark steel box. The cell was exposed to a ¹³⁷Cs γ -ray source.

The charge of the PMT output pulse was converted to an amplitude by a charge sensitive amplifier and then shaped by a shaping filter. Finally, the shaped signal amplitude was analyzed by a pulse

height analyzer. The system stability has been measured by means of an LED driven by a pulser generator. Fig. 3 shows the system stability. The system approached stability after 6 hours burning. The system burned in our experiment about 12 hours before the data acquisition.

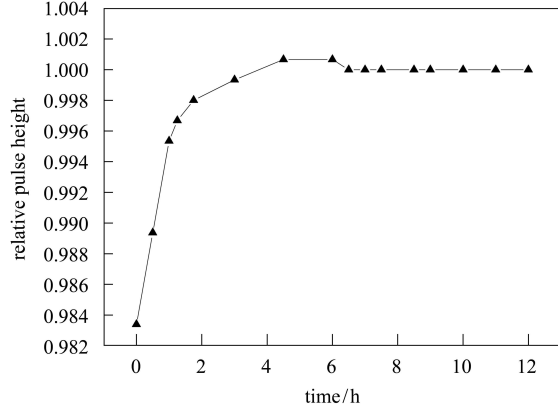


Fig. 3. System stability during 12 hours. The stability was tested by a LED driven by a pulse generator.

4 Data analysis and results

The energy spectrum $N(E)$ of the Compton scattering electrons is generated by means of the Monte Carlo code GRESP [8]. Fig. 4 shows the simulation result. It should be noted that the resolution smearing is not considered in the GRESP Monte Carlo code. We treated the resolution smearing similar to the way described in Ref. [9]. The “realistic” Monte Carlo spectrum can be obtained from the convolution of the simulation spectrum with the system response function,

$$N^{\text{MC}}(H) = \int R(H, L) N_L(L) dL, \quad (12)$$

where $R(H, L)$ is the response function, H is the ADC channel and $N_L(L)$ is the spectrum of the light output. For electrons (above about 50 keV), the light output L , the energy emitted as fluorescence, is proportional to the energy E deposited in the LS [10], i.e.

$$L = SE, \quad (13)$$

where S is the absolute scintillation efficiency. The light output L can be defined to be in units such that it is equal to the electron energy E [11], i.e. $S = 1$ and

$$L(E) = E. \quad (14)$$

If oxygen is present in the LS, the light yield will decrease. Let the quenching factor, i.e. the ratio of the

light yield with the oxygen quenching to that without oxygen quenching, I/I_0 , be f_Q . Then the light output can be rewritten as

$$L = f_Q E. \quad (15)$$

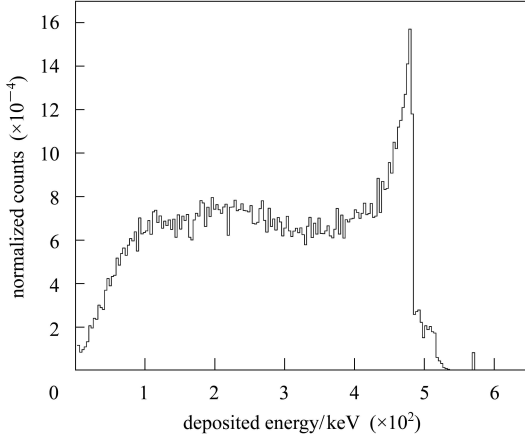


Fig. 4. Energy response of the LS to the ^{137}Cs γ -ray source simulated by GRESP. The counts are normalized to the number of γ -rays emitted from the ^{137}Cs source.

One should remember that f_Q is 1 for the LS without a quencher. Considering that the spectrum of the deposited energy is $N(E)$, the spectrum of L , $N_L(L) = N(L/f_Q S)/f_Q$. We assume that the response $R(H, L)$ of the detector for the fixed light output is Gaussian,

$$R(H, L) = B \exp\left(-\frac{(H - cL)^2}{2\sigma_{cL}^2}\right), \quad (16)$$

where c is the light output to the ADC channel conversion factor, and B is the normalization factor between the “realistic” Monte Carlo spectrum and the experimental spectrum. σ_{cL} can be expressed in the form of the system resolution,

$$\sigma_{cL} = \frac{cL}{2\sqrt{2\ln 2}} \rho, \quad (17)$$

where $\rho = \Delta(cL)/cL$ is the detector resolution, i.e. FWHM. The background can be thought to be exponential [6],

$$N^{\text{BG}}(H) = c_1 \exp(-c_2 H + c_3), \quad (18)$$

where c_1, c_2, c_3 are the parameters of the background. Then, the expected experimental spectrum can be written as

$$N^{\text{MC}}(H) = B \int \exp\left(-\frac{(H - cf_Q E)^2}{2\sigma_E^2}\right) N(E) dE + c_1 \exp(c_2 H + c_3), \quad (19)$$

with

$$\sigma_E = \frac{cf_Q E \rho}{2\sqrt{2\ln 2}}. \quad (20)$$

The expected experimental spectrum is determined by B, c_1, c_2, c_3, c, f_Q and the detector resolution ρ . c depends on the optical properties of the cell, PMT and electronics. It is independent of quenching and can be treated as a constant in our experiment. Hence the item $f_Q c$, which is proportional to the quenching factor, can be taken as one free parameter. To evaluate the free parameters we used the ROOT package (CERN data analysis package) to fit the experimental spectrum with Eq. (19). Fig. 5 shows the fit result of the background, the realistic spectrum, and the expected spectrum for the experimental spectrum. The items $f_Q c$ for different LS samples can be obtained by fitting their experimental spectra with Eq. (19).

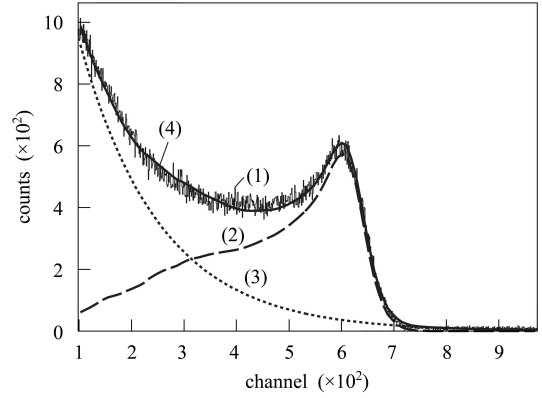


Fig. 5. (1) Experimental spectrum, (2) “realistic” Monte Carlo spectrum, (3) Background defined in Eq. (18), (4) expected experimental spectrum defined in Eq. (19) for ^{137}Cs .

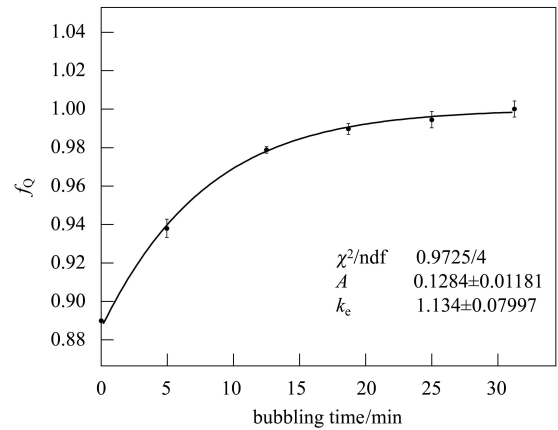


Fig. 6. f_Q as a function of the bubbling time. The solid curve shows the fits with Eq. (9).

With the increase of the nitrogen bubbling time, the light output increased. We assume that oxygen was fully bubbled, namely no oxygen quenching effect remains in the LS, when the light yield changes little. It should be reminded that the quenching factor, I/I_0 , for the full bubbled LS is 1. c was established from the fit result of the full bubbled LS. Then the values of f_Q for different LS samples were determined. Fig. 6 shows f_Q for six LS samples. The solid line in Fig. 6 shows the fits with Eq. (9). From the experimental result we know that the light yield increases by about 11% (20 °C) by means of removing the oxygen from the LS.

5 Conclusions

The oxygen quenching in a LAB liquid scintillator and the degassing model have been studied. From the experiment we know that the LAB LS light yield is increased by 11% by fully removing the oxygen at 20 °C. Moreover, we proposed a model to determine the relationship between the light yield and the bubbling time in this paper. The parameters of the model have been fixed experimentally.

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