Aging of LAB-based liquid scintillator in stainless steel containers *

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Abstract: Types 316 and 304 stainless steel are two candidates for the storage vessels and piping systems of LAB-based liquid scintillator (LS) in the JUNO experiment. LS aging experiments are carried out at temperatures of 40 ℃ and 25 ℃. After 192 days aging at 40 ℃, the attenuation length of LS was reduced by 6% in a glass container, 12% in a type 304 stainless steel tank, and 10% in a type 316 stainless steel tank. At 25 ℃ in 304 and 316 stainless steel tanks, the attenuation length was reduced by 6% after 307 days. The light yield and the absorption spectrum were practically the same as that of the unaged sample. The concentration of element Fe in the LAB-based LS did not show a clear change. Type 316 and 304 stainless steel can be used as vessels and transportation pipeline material for LAB-based LS.

Key words: JUNO, liquid scintillator, aging, light yield, absorption spectrum, attenuation length

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1 Introduction

The Jiangmen Underground Neutrino Observatory (JUNO), which aims to determine the mass hierarchy at a confidence level of $\Delta x^2_{\rm MH} \sim (10\div 12)(3\div 3.5\sigma)$ [1], is designed with a target-detector filled with 20 kton liquid scintillator (LS) of $3\%/\sqrt{E(\rm MeV)}$ energy resolution. To measure the energy spectrum of the reactor neutrinos precisely, optimum transparency of the LS is needed, with requirements on the attenuation length of 22 m, light yield of 1200 pe/MeV, and radiation background $<10^{-15}$ g/g. Aging is recognized as one of the main degradation mechanisms affecting the properties of LS.

The same composition as the Daya Bay LS is used in this study: 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 [2].

Prolonged contact with oxygen likely causes LS oxidation effects [3–5]. In addition, high-energy rays or incident particles on the radiation field may also modify the composition and properties of the LS. The LS must therefore be stored in an oxygen-free and low-radiation environment.

In addition to considering oxidation and radiation effects, the erosion and impurity quenching effects caused by storage vessels should be taken into consideration. Stainless steel is the material used for the LS storage vessels and piping systems in the JUNO experiment. Type 316 and 304 stainless steel are widely used and highly resistant to liquid organics, but this quality depends on the composition and concentration of the liquid organics and the surrounding temperature [6, 7]. The properties of LS, therefore, have to be tested after long-time contact with 316/304 stainless steel.

In this study, the aging effects are identified by evaluating the light yield, attenuation length, the absorption spectrum and the change in Fe content.

The heating method plays an important role in accelerating the reaction rate of these experiments. LS aging experiments are carried out at a temperature of 40 ℃, with tests done at a temperature of about 25 ℃ as a comparison.

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2 Experimental procedure

2.1 Experimental setup

An LS aging experimental system with two different heat-treatment conditions (40 ℃ and 25 ℃) was set up. A schematic drawing of the experimental setup is shown in Fig. 1. Two incubators ((a) and (b)) were used to control the temperature with a variation of ±1 ℃. Four conical glass flasks were put in incubator (a). The volume of each flask was 2 L, and the flasks were sealed well to avoid against oxidation effects. Type 316 and 304 cylindrical stainless steel tanks were placed inside cuboid incubator (b). Each tank had a volume of 15 L and a diameter of 21 cm, with 10 L of LS inside. In addition, 600 mL/min nitrogen was pumped into each tank and finally into a bubble beaker to help avoid oxidation effects. To increase the contact area between LS and stainless steel, in order to accelerate the aging speed, a 180 mm×430 mm sheet of 316 and 304 stainless steel was placed into each corresponding tank.

In order to check the temperature of each LS container, a thermocouple temperature sensor was placed on one of the conical glass flasks and on the 316 stainless steel tank. The LS in the four flasks stabilized at 38.75–40.50 ℃, and the LS in the 316 and 304 tanks stabilized at 39.75–40.50 ℃.

We took out 2 L LS samples from the conical glass and type 316 and 304 stainless steel tanks after 53, 94, 144 and 192 days, respectively. A 316 and a 304 tank of the same geometry, with 10 L of LS but without the stainless steel sheets inside, were kept at 25 ℃. The tanks were sealed well to avoid oxidation effects. The LS stabilized at (25±2) ℃. 2 L of LS was taken out for comparison on the 307th day.

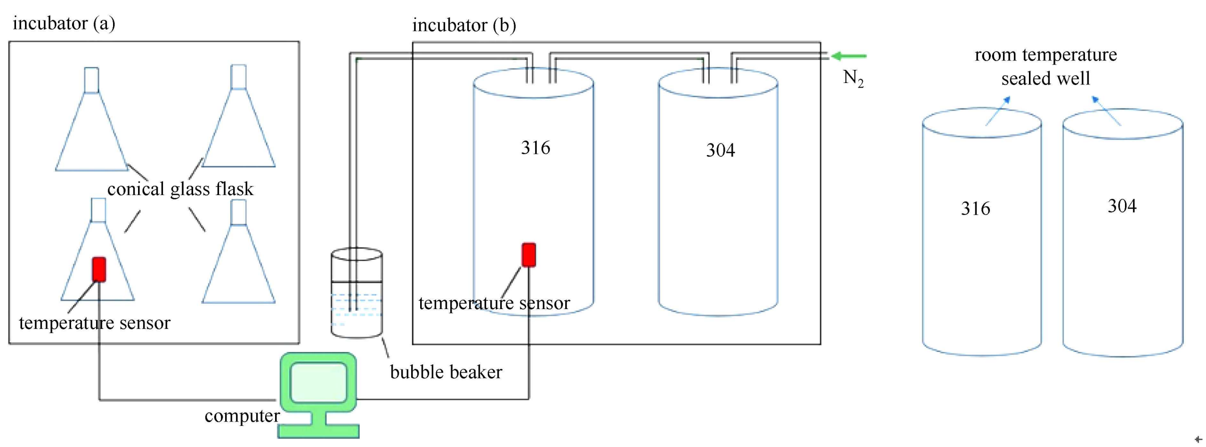

Fig. 1. LS aging system.

2.2 Aging time conversion for stainless steel tanks

The LS aging experiments were carried out in stainless steel tanks at room temperature and 40 ℃, so the different LS samples in the steel tanks correspond to different reaction rates.

According to Van't Hoff [8], for homogeneous thermochemical reactions, if the reaction temperature rises 10 ℃, the reaction rate becomes 2–4 times higher, which can be expressed as

$$k=Q^{\frac{T_2-T_1}{10}}, \tag{1}$$

where k is the chemical reaction rate; T_2, T_1 are temperatures; and Q is a constant, in the range 2 to 4.

Comparing the 40 ℃ and 25 ℃ aging experiments, adding stainless steel sheets to the 40 ℃ tanks increases the contact area between stainless steel and LS by up to about 1.5 times, so a correction of the reaction rate needs to be done. A factor for the increased contact area is added (σ=1.5), and the Q value is taken as 3 (the median value). So,

$$t_{\text{eq.}}=\sigma t_{\text{ag.}}3^{\frac{T_2-T_1}{10}}, \tag{2}$$

which means that at time $t_{\text{ag.}}$ at temperature T_2 in the stainless steel tank, the equivalent time will be $t_{\text{eq.}}$ at 25 ℃.

According to Eq. (2), at 40 ℃, 53 days aging time of LS is equivalent to 1.13 years at 25 ℃, 94 days is equiv-

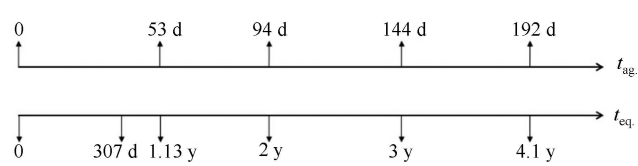

Fig. 2. Time conversion.

alent to 2 years, 144 days is equivalent to 3 years, and 192 days is equivalent to 4.1 years. The time conversion is illustrated in Fig. 2.

3 Results

3.1 Relative light yield measurements

The light yield of the LS is generally measured via Compton scattering of γ rays. When colliding with γ rays, excited LS molecules will emit definite wavelengths of ultraviolet or visible light. Fig. 3(a) shows the pulse amplitude spectrum without coincidence detection, showing all the features of a Compton spectrum.

To reduce the fit error, the measurement is designed to use Compton scattering of known energy to produce mono-energetic electrons in the LS by tagging the scattered photons at certain angles. A coincidence detector is added, which consists of a $LaBr_3$ crystal and a photomultiplier tube. With this coincidence system, only fixed angles of scattering γ photons are received. They deposit a small fixed range of energy in the LS, giving a Gaussian peak. Fig. 3(b) shows the pulse amplitude spectrum with coincidence detection; the peak can be fitted with a Gaussian function.

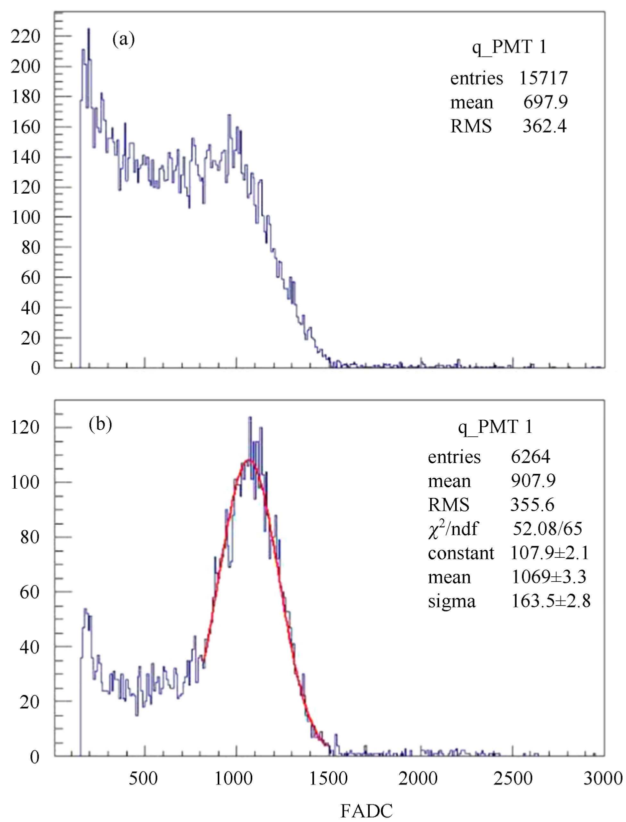

Fig. 3. LS energy spectrum of non-coincidence detection (a) and coincidence detection (b).

As shown in Fig. 4, the LS samples were irradiated by a radioactive source ^{137}Cs (15 μCi). Each LS sample was quantitatively weighed to be 130 g. The solid angle subtended by the LS from the $LaBr_3$ crystal was about 0.03 sr. All LS measurements were made in a dark room.

The measured result shows that the light yield reduces by a maximum of 2% after 192 days aging at 40 ℃ (Fig. 5). In consideration of the statistical error, also 2%, the light yield is practically the same as that of the unaged LS. With long-time contact with the 316 and 304 stainless steel, the light yield of LAB-based LS reduces very little.

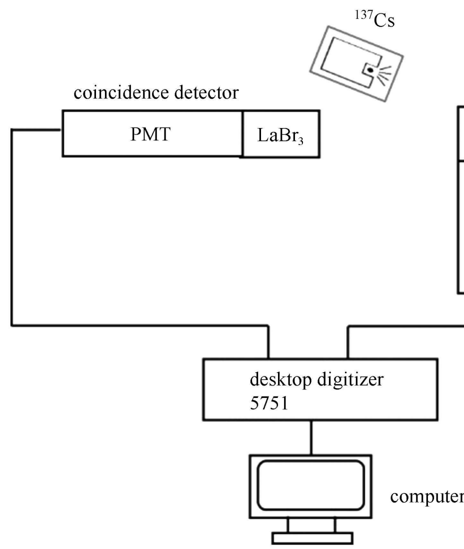

Fig. 4. Experimental setup for measuring the LS light yield.

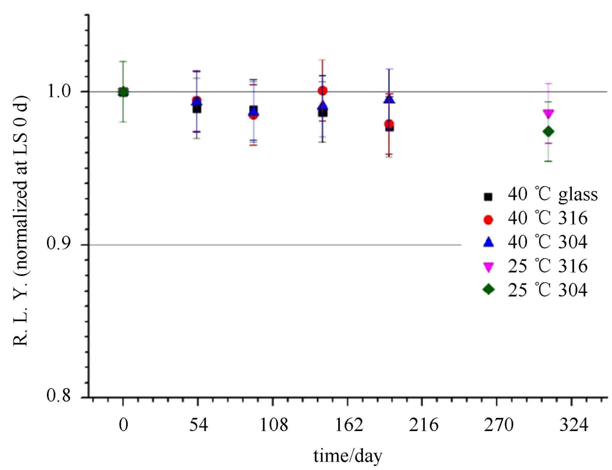

Fig. 5. The result of relative light yield (R. L. Y) measurement.

3.2 Attenuation length measurements [9]

The attenuation length L is defined in the formula $I(\lambda,x)=I_0(\lambda)\mathrm{e}^{-\frac{x}{L}}$, where λ represents the wavelength of the light beam, $I_0(\lambda)$ represents the initial monochromatic light intensity, and x represents the length of the LS. Thus, the light intensity $I(\lambda,x)$ that reaches the photomultiplier changes with x.

The attenuation length measurement apparatus is illustrated in Fig. 6. A pulse generator is used to drive the light emitting diode (LED). An LED of wavelength (430±5) nm is used as the light source, as 430 nm is an intermediate value of the emission spectrum (410–450 nm) of the LS. The light beam emitted by the LED enters the LS via a fiber, lens, filter (430 nm), and an aperture diaphragm (5 mm diameter). The length and diameter of the sample container are 1.5 m and 4 cm, respectively.

The stability of the measurement system is measured to be about 200 minutes. During the time the system is stable, the attenuation length of the LS is obtained by measuring at least 7 (x, $I(\lambda,x)$) points.

The results (Table 1) shows that after 192 days aging

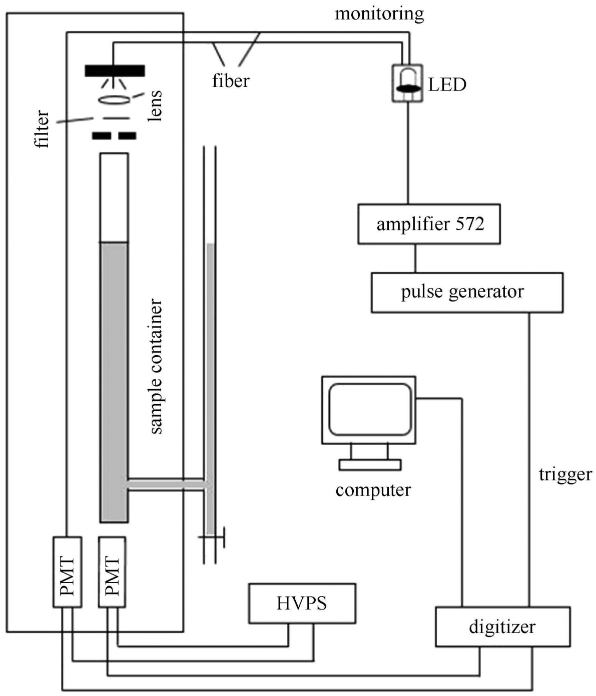

Fig. 6. Attenuation length measurement device.

Table 1. Specific data of aging LS attenuation length.

aging time/day	attenuation length of LS/m glass	316	304
0	14.84±0.53	14.84±0.53	14.84±0.53
temperature 40 ℃			
53	14.72±0.69	14.18±0.75	14.05±0.73
94	14.15±0.85	13.92±0.80	13.65±0.79
144	14.45±0.68	14.28±0.78	14.10±0.78
192	13.99±0.87	13.39±0.77	13.02±0.68
temperature 25 ℃			
307		14.07±0.68	13.89±0.71

at 40 ℃, the attenuation length of LS reduced by 6% in the glass container, 12% in the 304 stainless steel tank and 10% in the 316 stainless steel tank. In the 304 and 316 stainless steel tanks at 25 ℃, the attenuation length reduced by 6% after 307 days. Type 316 stainless steel has a relatively lower impact on the LS attenuation length than type 304 (Fig. 7).

3.3 Absorption spectrum measurements

An L650 UV-Vis spectrophotometer was used to measure the LS absorption spectra corresponding to the wavelength range of 190–900 nm. As shown in Fig. 8, the absorption spectra of LS samples in the wavelength range of 410–510 nm are very similar. So, the absorption spectra of aged LS samples show no clear change.

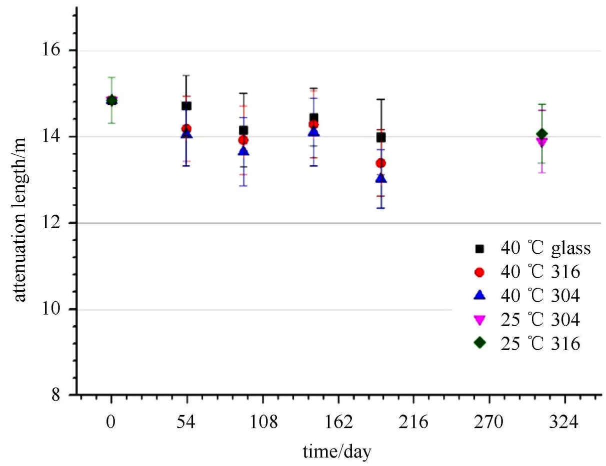

Fig. 7. Attenuation length of aging LS.

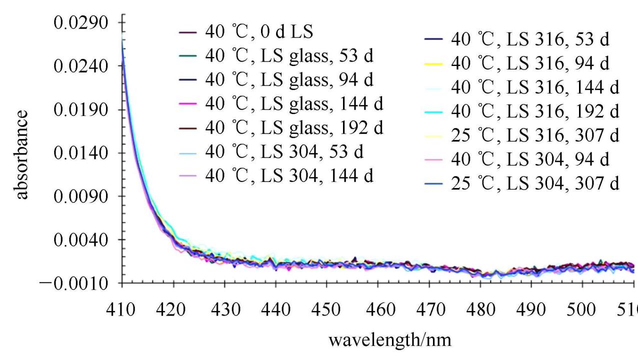

Fig. 8. Absorption spectrum measurement results.

3.4 Fe concentration measurements

The stainless steel may affect the properties of LS by impurity quenching effects. We analyze impurities that are released to the LS by examining the Fe impurity concentration. 6 mol/L nitric acid was used to extract Fe from the organic phase into the aqueous phase. The

Table 2. Results of Fe concentration test.

samples of aged LS	Fe concentration/ppm
0 d	1.84±0.13
316type, 25 ℃, 307 d	1.69±0.11
316type, 40 ℃, 192 d	1.90±0.35
304type, 25 ℃, 307 d	1.82±0.19
304type, 40 ℃, 192 d	1.69±0.11

aqueous phase was then diluted and measured by ICP-MS. The results are given in Table 2. The concentration of Fe in LS does not show a clear change from contact with type 316/304 stainless steel.

4 Conclusions

A set of aging experiments has been conducted and a suitable LS aging time calculation method has been applied, with LS aging experiments for a period of 53, 94, 144 and 192 days at 40 ℃ equivalent to 1.13, 2, 3 and 4.1 years respectively at a temperature of 25 ℃.

After 192 days aging at 40 ℃, the attenuation length of LS is reduced by 6% in glass container, 12% in 304 stainless steel tank, and 10% in 316 stainless steel tank. At 25 ℃, it is reduced by 6% in 304 and 316 stainless steel tanks after 307 days. The light yields and the absorption spectra of the aged samples are almost the same as those of the unaged one. The concentration of the element Fe in LS does not show a clear change.

The results reveal that types 316 and 304 stainless steel have only a small effect on LS, and both can be used as vessels and transportation pipeline material for LAB-based LS.

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