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An optimal solvent of liquid scintillator for fast-neutron imaging applications

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ABSTRACT

For fast neutron detection and imaging applications in the inertial confinement fusion (ICF) experiments, a scintillator with high reflective index, good transparency in the wavelength range of the fluorescence emission spectra, fast decay component with a few nanoseconds decay time is required. In this work, when 2,5-diphenyloxazol and p-bis(o-methylatyryl)-benzene were used as the fluors of a liquid scintillator, three kinds of solvents including PX (p-xylene), PXE (l-phenyl-l-xylylethane) and DIN (di-isopropylnaphthalene) were selected to prepare a liquid scintillator. The wavelength-dependent attenuation length and the fluorescence emission spectra of liquid scintillator mixtures were measured by an UV–vis spectrometer and a FL spectrofluorimeter, respectively. The decay time of liquid scintillator was tested employing the time-correlated single photon counting (TCSPC) technique featured in high dynamic range of several orders of magnitude in light intensity. The experimental results indicated that the optimal solvent was PX from the point of energy transfer efficiency and optical properties.

1. Introduction

As a sustainable, clean, and safe energy, fusion energy plays an important role in solving the global energy crisis [1,2]. Some famous global research institutions, such as the Institute of Laser Engineering (ILE) at the Osaka University, the OMEGA-EP at the Laboratory for Laser Energetics (LLE) at the Rochester University [3,4] and the Lawrence Livermore National Laboratory (LLNL) [5], have already focused the research on the Fast Ignition to obtain the fusion energy. In the fast ignition experiments, quasimonoenergetic neutron spectra with energy about 14.1 MeV are produced in the fusion capsule filled with a deuterium–tritium (DT) or deuterium–deuterium (DD) mixture [6,7]. Therefore, how to detect the neutron spectra is a key issue to understand the dynamics of the imploded fusion plasma. Using a well characterized organic scintillation detector and the time-of-flight (TOF) technique to diagnose the neutron signals is desirable [8]. Particularly, liquid scintillator with good optical properties plays a key role in the scintillation detector.

A scintillator is always made up of an aromatic solvent and one or more fluors (primary fluor and wavelength-shifter). The solvent acts as a vehicle for energy transfer of solution and interact with neutron to produce scattering electrons. Therefore, good transparency in the wavelength-dependent fluorescence emission spectra, high refractive index, long-term stability and high flash-point [9] is required for an optimal solvent. PC (pseudocumene) and LAB (linear-alkyl benzene) are the traditional solvents of liquid scintillator. However, PC can withstand large pulse amplitude but shows the shortages of high corrosion and low flash-point. LAB has such good transmittance in the interesting region that it is the best candidate for large-scale detectors, but the low reflective index makes it unsuitable to be applied to neutron diagnostics. With the increasing improvement of the neutron diagnostics technique, the conventional solvents could not meet the demand of neutron diagnostics in the fast ingition experiments any longer. Pursuing an appropriate solvent in liquid sicntillator for neutron diagnostics is paid much more attention.

In this paper, 2,5-diphenyloxazol (PPO) and p-bis(o-methylatyryl)-benzene (bis-MSB) were selected as fluors of liquid sicntillator due to their high fluorescence quantum yield φ_F, short

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lifetime τ_F, large Stokes shift, high solubility and low cost. PX (p-xylene), PXE (l-phenyl-l-xylylethane) and DIN (di-isopropylnaphthalene) were used as solvents. The absorption spectra, the emission spectra and the decay time of the solution were measured. According to the experimental results, p-xylene was chosen to be an optimal solvent suitable for PPO and bis-MSB.

2. Scintillation mechanisms

Both solvent and fluor in a liquid scintillator, are aromatic hydrocarbons featured in benzenoid and heterocyclic ring structures [10,11]. The interaction among atoms in the ring structure is accomplished via localized σ-electrons or delocalized π-electrons. The π-electrons are vitally significant for the scintillator, because when the kinetic energy of energetic particles crossing the liquid scintillator is deposited mainly in the solvent molecules, the electrons (especially the π-electrons) of benzene-ring structures of the solvent are instantly excited, immediately the excited electrons de-excites to the ground states. However, the de-excitation processes are very complicated and influenced by various factors [10,11]. Hence, the primary mechanisms are briefly discussed. It is possible for the unsteadily excited molecular to decay to the ground states by radiative transitions or non-radiative transitions. Radiative transitions contain spin-forbidden transitions between triplet states and singlet states which would emit phosphorescence (up to ms) and spin-allowed ones just between singlet states sending fluorescence (a few ns). On the other hand, non-radiative transitions include internal conversion, intersystem crossing and vibrational relaxation. Finally the de-excitation of the π-electron in the fluors molecule emits visible or near ultraviolet light originating from the transitions between the first excited spin-singlet states and one of the vibrational levels of the ground states.

3. Experimental set-up

The decay time curves clearly reflect the lifetime and the percentage of all the components in the scintillator mixtures through a setup inspired from the time-correlated single photon counting technique (TCSPC), initially presented by Bollinger, Thomas [12] and Koechlin [13,14], as diagrammatically shown in Fig. 1. In this apparatus, the liquid scintillator mixtures were filled in the cylindrical quartz container (Φ40 mm × 50 mm) with stainless steel lid. The enhanced specular reflector was coated on the inner surface of the lid and the lateral of the container, and a small aperture was left on the side of the container to emit the single photoelectron. The measurement was performed by applying two XP2020 photomultiplier tubes. One phototube (PMT1) is tightly coupled to the bottom of the quartz container to detect all the occurring events. The other one (PMT2) is placed about 30 cm away from the small aperture of the quartz container. The distance is adjusted to ensure that the probability of detecting a single photoelectron from a Compton event in the PMT2 is less than 3% [15].

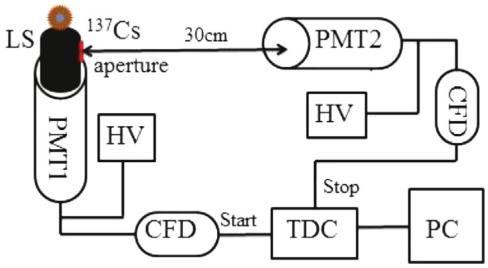

Fig. 1. Schematic diagram of the experimental setup known as Time-Correlated Single Photon Counting (TCSPC) for measuring the decay time of luminescence intensity. LS: liquid scintillator, PMT: photomultiplier tube, HV: high voltage, CFD: constant fraction discriminator, and TDC: time-digital converter.

The setup is put in a darkroom. The signals from PMT1 are attached to the constant fraction discriminator (CFD) and used as the start signals of the time-digital converter (TDC). The signals from PMT2 are linked to CFD and regarded as the stop signals of the TDC. The dynamic range of TDC is 213 ns, but the delay time of TDC is 64 ns, thus the time after 64 ns is meaningful. In the latter 149 ns, the time difference between the start signal by the liquid scintillator (LS) and the stop signal from PMT2 was recorded in the data acquisition program implemented through the Labview software in the personal computer (PC).

The time resolution of the system has been tested by a slight modification of the setup. A Cherenkov radiator is put in front of PMT2. A ^{60}Co source replacing ^{137}Cs is placed between the quartz container and the Cherenkov radiator. The ^{60}Co emits two coincident gammas in opposite directions, one colliding with the scintillator and the other with the Cherenkov radiator. When both of the collisions appear simultaneously, the coincident events are stored in PC. Therefore, the time resolution of the system is determined by the time difference between the signals in the scintillator and in the Cherenkov radiator. The key factor is the single-photoelectron transit time jitter of the PMT in the time resolution of the system. Furthermore, the error of the determination of the start time of a pulse and the time jitter of the electronic readout chain lead to the loss in time resolution. Fig. 2 displays the time resolution of the system. The Gaussian curve is well depicted by a double-Gaussian function, but there are small contributions sometimes below or above the Gaussian curve.

4. Results and discussion

4.1. Light absorption and attenuation length

It is necessary to research optical transparency markedly dependent on the wavelength to reduce self-absorption and improve light yield. Absorption spectra are tested employing a PerkinElmer UV/vis spectrophotomer (Lambda 950). Then, the Lambert Beer empirical formula has been utilized to reveal the relationship between the absorption value and attenuation length when it passes a cell path length [16,17]. The attenuation length (Λ) is obtained from the equation $\Lambda = 0.4343L/A$, Where L is the cell path length (usually a

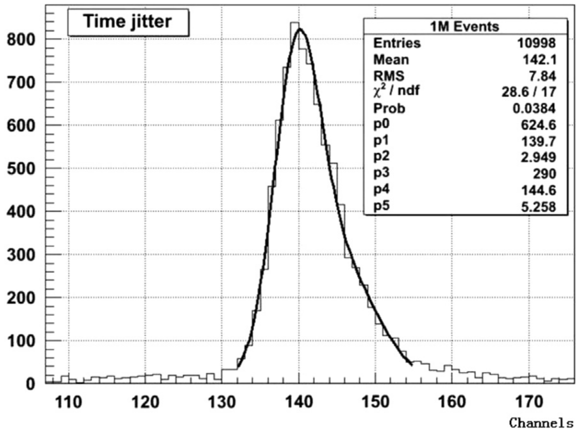

Fig. 2. Distribution of the time differences between the start signal in the scintillator and the arrival of the Cherenkov photons. The time resolution of the system is well described by a double-Gaussian function. For the first Gaussian peak, $\sigma = p^2 = 2.94 \times 90.2$ ns/channel = 0.59 ns, FWHM = 1.39 ns. The second Gaussian peak is responsible for the reflection of Cherenkov light.