Nuclear Inst. and Methods in Physics Research, A 897 (2018) 47–53

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Characterization of a tin-loaded liquid scintillator for gamma spectroscopy and neutron detection

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ARTICLE INFO

Keywords:
Tin-loaded liquid scintillator
Energy resolution
Timing resolution
Neutron light output
Pulse shape discrimination

ABSTRACT

A tin-loaded liquid scintillator has been developed for gamma spectroscopy and neutron detection. The scintillator was characterized in regard to energy resolution, pulse shape discrimination, neutron light output function, and timing resolution. The loading of tin into scintillators with low effective atomic number was demonstrated to provide photopeaks with acceptable energy resolution. The scintillator was shown to have reasonable neutron/gamma discrimination capability based on the charge comparison method. The effect on the discrimination quality of the total charge integration time and the initial delay time for tail charge integration was studied. To obtain the neutron light output function, the time-of-flight technique was utilized with a ^{252}Cf source. The light output function was validated with the MCNPX-PoliMi code by comparing the measured and simulated pule height spectra. The timing resolution of the developed scintillator was also evaluated. The tin-loading was found to have negligible impact on the scintillation decay times. However, a relatively large degradation of timing resolution was observed due to the reduced light yield.

1. Introduction

Plastic and liquid scintillators are widely used in many applications such as high-energy physics, radiation environmental monitoring, and nuclear security because of the following attractive characteristics: (a) the scintillators can be manufactured in large sizes and various shapes with ease; (b) the material cost is significantly lower compared with many commonly used detector materials including NaI(Tl) (inorganic scintillator), CZT (room-temperature semiconductor), and high-purity germanium; (c) many of them are sensitive to neutrons and γ-rays and different particle types are discriminable using Pulse Shape Discrimination (PSD) techniques; (d) the scintillation decay times are on the order of nanoseconds, which is required to mitigate the adverse effect of pulse pile-up in high-rate applications; (e) the scintillators can provide timing resolution of less than one nanosecond, making them suitable for timing measurements. However, organic scintillators have rather long mean free paths for gamma rays in the energy range of interest from tens of keV to a few MeV due to the low effective atomic number. Most detected gamma-ray events are from Compton scattering interactions. This results in an absence of photopeaks in the pulse height distributions, limiting their use in gamma spectroscopy measurements. In addition, some of them are toxic, flammable, and fragile, limiting their applications in certain scenarios such as nuclear decommissioning and nuclear security.

Many efforts have been devoted to increasing the photoelectric effect probability by loading heavy materials into low-Z plastic or liquid scintillators [1–10]. Non-exhaustive examples are summarized here. Cho et al. reported the photon detection efficiency of the plastic scintillator was remarkably improved because of the tin-loading [1]. The NE-140 plastic scintillator loaded with 5% tin was able to clearly separate the photopeaks corresponding to the 42 keV and 100 keV photons. The FWHM of the 100 keV photopeak was reported to be 40%. In the search of double beta decay of ^{124}Sn, Hwang et al. synthesized liquid scintillators with loading of up to 50% tetramethyltin to increase the photopeak efficiency [2,3]. Koshimizu et al. studied the capability of bismuth-loaded plastic scintillators in high-energy X-ray detection [4]. They found the detection efficiency was significantly improved by increasing bismuth concentration. In addition to the loading with tin or bismuth, lead was also used as a metal carboxylate additive to increase the photoelectric effect probability. For example, Hamel et al. presented lead-loaded plastic scintillators for the use in high-efficiency position sensitive X-ray imaging systems [5].

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https://doi.org/10.1016/j.nima.2018.04.054
Received 2 February 2018; Received in revised form 28 March 2018; Accepted 26 April 2018
Available online 3 May 2018
0168-9002/

A tin-loaded liquid scintillator has been acquired and mixed by Nanoptics Inc. in this work with the aim to design low cost, high sensitivity gamma spectroscopy and neutron detection systems with intent to be usable as high efficiency vehicle-based mobile detection units. In the proposed design the liquid scintillator will be contained in a thin walled (0.125″) aluminum vessel with dimension of 6″ by 6″ by 12″ depth. This is because the metal container eliminates the well-known problem of gradual fogging of plastic scintillator plates with time, especially in hot and humid environments. The liquid scintillator will be made into a non-pourable gel to ensure safe recovery from a catastrophic accident. It is expected the gel will only be slightly affected by the physical and vibrational environment in a moving vehicle compared to brittle crystal detectors. The use of gel can also reduce the risk of chemical toxicity. Wavelength shifting fibers will be emerged in the gel to minimize the self-absorption of scintillation photons. The γ-ray detection efficiency of the systems will be 3–5 times greater than that of the spectroscopic scintillating plastics produced by Symetrica Ltd [11,12]. This was based on our Monte Carlo simulation results. In the simulation, the size of the tin-loaded liquid scintillator was 6″ by 6″ by 12″ and the shape/volume of the plastic scintillator were the same as those used in the spectroscopic scintillating plastic detectors developed by Symetrica Ltd. The relative detection efficiency was defined as the ratio of total interaction probabilities in the tin-loaded liquid scintillator and the spectroscopic scintillating plastic scintillator. The systems will permit two simultaneous and different types of γ-ray measurements. First, Compton edge data can be deconvolved to give spectroscopic results similar in quality to that obtained from the Symetrica spectroscopic plastic scintillators (5% at 662 keV). Secondly, photopeak data with peak-to-total ratio comparable with that from a standard 3″ diameter by 3″ height NaI(Tl) detector can be obtained. The combination of high detection sensitivity and relatively accurate peak location measurement ensures reliable identification of threat sources in a complex gamma spectrum of naturally occurring radioactive materials. The systems will also have acceptable neutron/gamma discrimination capability. In this paper the characterization of such a scintillator of 7.62 cm diameter by 7.62 cm height (3″ by 3″) is presented in regard to energy resolution, PSD performance, neutron light output function, and timing resolution.

2. Experimental methods

The selection of individual components of the liquid scintillator is based on consideration of several factors which include light output, cost, flammability, auto-ignition, and optical transmission. A further factor was the requirement that the liquid could be made into a non-pourable gel. This factor ensures safe deployment of the scintillating material despite a breach of its container. The measurements reported in this paper were made with the liquid tin-loaded scintillator as the process of transforming the liquid into the gel state was achieved subsequent to these measurements. Preliminary results using the gel material show negligible changes in the scintillating properties. The liquid scintillator solution contains the following components: (a) 69% wt./wt. Diisopropylnaphthalene (DIN); (b) 30% wt./wt. tetra-*n*-butyltin (Bu_4Sn); (c) 0.99% wt./wt. 2,5-diphenyloxazole (PPO); (d) 0.01% wt./wt. 1,4-bis(2-methylstyryl)benzene (bis-MSB). The tin weight was calculated to be around 10.1 wt%. The scintillator was contained in a 3″ by 3″ regular cylindrical aluminum container with inner surface coated by white reflective paint and optically coupled to a 3″ diameter ET Enterprises bialkali PMT (model number: ETEL 9821KB) using silicone grease. The container was a standard commercial housing unit capable of accommodating for possible dilatation of the liquid scintillator. Oxygen in the liquid scintillator was rejected by the use of nitrogen (approximately 30 min) to reduce its impact on the detector performance such as light yield, neutron/gamma discrimination, and timing resolution. The PMT high voltage provided by a CAEN VME power supply V6533N was set at negative 1700 V.

In the measurements for studying the energy resolution, PSD, and neutron light output function, a high-speed Struck Innovative Systeme digitizer SIS3316 was employed as the data acquisition system. The digitizer has sampling rate of 250 MHz and resolution of 14 bits. Its input dynamic range was programmed to be 0–2 V to match the voltage levels of the input pulses. The PMT anode pulses were directly fed into the digitizer via a shielded coaxial cable of 6 ft length without being processing by any preamplifier or amplifier devices. The impact of cable length on PSD performance observed by Pawełczak et al. was not studied here [13]. The waveform of each pulse was acquired in an 800 ns time window with a pre-trigger time of 80 ns. The digitized data was transmitted through an Ethernet cable to a host computer for off-line processing.

Energy calibration of the tin-loaded detector was performed with 1 μCi ^{137}Cs and 1 μCi ^{22}Na sources. The sources were positioned approximately 5 cm from the front surface of the detector. The pulse height spectrum measured with the ^{137}Cs source was utilized to determine the energy resolution. Neutron/gamma discrimination performance was evaluated using a ~100 μCi ^{252}Cf source and the charge comparison based PSD technique [14]. The distance between the ^{252}Cf source and the detector was approximately 30 cm such that the possibility of pulse pile-up was negligible while maintaining a reasonable input count rate. For the neutron light output function, the response of the detector to neutrons were first obtained from the Time-of-Flight (ToF) measurements with the ^{252}Cf source. Then, these pulse spectra were used to derive the light output function. The ToF start time was recorded by a 3″ by 3″ EJ-309 detector located around 5 cm from the source. The distance between the tin-loaded detector and the source was approximately 116 cm. The experimental setup was kept as far as possible away from surrounding environments such as concrete walls to minimize the background from indirect scattering neutrons. The total ToF measurement time was 192 h.

In the timing resolution measurements, the digitizer SIS3316 was replaced by a CAEN digitizer DT5730 because the higher sampling rate available from the DT5730 compared with the SIS3316 (i.e. 500 MHz vs 250 MHz) is more suitable for timing measurements. The DT5730 has the same vertical resolution and input dynamic range as the SIS3316. The Constant Fraction Discrimination (CFD) method was employed to obtain the timestamp of each event. The CFD delay time and fraction were 6 ns and 50%, respectively. The source used in the timing measurements was the 1 μCi ^{22}Na. It was located around 10 cm from each detector. First, the timing resolution of the 3″ by 3″ EJ-309 detector was determined with two theoretically identical EJ-309 detectors. The detectors were assumed to have the same timing resolution. The PMT voltages provided to the detectors were negative 1700 V and 1710 V respectively, at which the measured ^{137}Cs pulse height spectra were overlapped with each other. The difference in the voltages is reasonable considering that the PMTs of the same model exhibit minor differences. The spectra match would reduce the effect of variation of pulse amplitude on timing resolution and therefore minimize the difference in the timing resolution of the EJ-309 detectors. One of the EJ-309 detectors then served as a reference detector in the timing measurement for the tin-loaded detector.

3. Results and discussion

3.1. Energy resolution

Energy information of each pulse was obtained with the charge integration method. The integration started at 40 ns before the peak maximum and ended at 260 ns after the maximum with total integration time of 300 ns. In the charge integration method, the pulse was not utilized when the pulse amplitude exceeded the digitizer dynamic range or it was followed by the next pulse within the charge integration time. Fig. 1(a) presents the pulse height spectrum acquired with the ^{137}Cs source. In addition to the Compton edge normally seen from a spectrum measured with low-Z liquid or plastic scintillators, Fig. 1 shows that a photopeak is observed because of the interaction of photons with