



Characterization of a liquid scintillator based on linear alkyl benzene for neutron detection

G. Bentoumi^{a,*}, X. Dai^a, H. Fritzsche^c, G. Jonkmans^a, L. Li^a, G. Marleau^b, B. Sur^a

^a Atomic Energy of Canada Limited, Chalk River (Ont.), Canada

^b Institut de Génie Nucléaire, École Polytechnique de Montréal, Montréal (Qc.), Canada

^c National Research Council, Canadian Neutron Beam Centre, Chalk River (Ont.), Canada

ARTICLE INFO

Article history:

Received 3 July 2012

Received in revised form

26 September 2012

Accepted 28 October 2012

Available online 10 November 2012

Keywords:

Scintillation

Boron

Linear alkyl benzene

Neutron

Pulse shape discrimination (PSD)

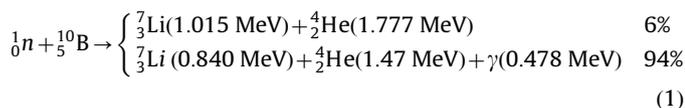
ABSTRACT

A liquid scintillator (LS) based on linear alkyl benzene (LAB) solvent has been characterized using multiple radiation sources. The results confirm that boron-loaded LAB is suitable for neutron detection in a gamma ray environment. To study indirectly the LAB pulse shape discrimination (PSD) capability between neutrons and gamma rays, a dissolved ²¹²Pb source emitting alpha and beta particles was used to emulate the conditions in a mixed radiation field for detecting neutrons in the presence of a high gamma ray background. The quenching factor depends on the alpha energy and increases from 10 to 25 as the alpha energy decreases from 10 to 1 MeV. ¹⁰B loaded LAB-based LS has been tested in a neutron beam of energy which is equal to 14.56 MeV. The observed peak at 60 keVee is attributed to the absorption of neutrons. Our results show that a boron-loaded LAB-based scintillator is a sensitive medium for neutron detection in a relatively large background of gamma rays. A neutron detector could be achieved with a figure of merit (FOM) of 1.75.

Crown Copyright © 2012 Published by Elsevier B.V. All rights reserved.

1. Introduction

There is a need in the nuclear industry for detectors that can measure neutron flux in an environment with a relatively large background of gamma rays. ³He filled ionization chambers have been extensively used for this purpose because of their good neutron-to-gamma discrimination capability. However, there is currently a global shortage of ³He. As a result, it is necessary to design new neutron detectors that rely on other mechanisms and are just as efficient as ³He detectors. The ¹⁰B capture process has long been suggested as an alternative to ³He. With an absorption cross-section of 3838 barn, an incident thermal neutron may be captured by ¹⁰B producing ⁷Li and an alpha particle as follows [1]:



The ⁷Li and the alpha particle have a short range and deposit their energies into the surrounding medium. A considerable number of optical photons are emitted following this interaction in the case of an appropriate ¹⁰B-loaded scintillator medium. These optical photons are then easily transformed to an electrical

pulse by a photomultiplier tube (PMT) providing an indication that a neutron has been detected. Therefore, this interaction can be used to detect neutrons.

Linear alkyl benzene (LAB) has been proposed as a solvent for a liquid scintillator since it has a high flash point (130 °C) and is inexpensive [4]. LAB is used in mixture with a fluor such as 2,5 diphenyl-oxazone (PPO) and a wavelength-shifter such as 1,4-Bis(2-methylstyryl)benzene (bisMSB) to improve scintillation light output and to match the PMT spectral response [2]. Development of particle identification techniques is necessary because the energy spectrum cannot be used to discriminate between neutrons and gamma rays [1]. One of the proposed methods to achieve discrimination between neutrons and gamma rays is pulse shape discrimination (PSD). It is based on the fact that the time-resolved optical emission depends on the nature of the particle initiating the signal [3].

In this paper we present results of the characterization of a LAB-based liquid scintillator for neutron detection. The experimental setup, PSD capabilities, and results with neutron beams are explained.

2. Experimental procedure

The liquid scintillator used in all the experiments is based on LAB solvent with fluor PPO and wavelength-shifter Bis-MSB, at concentrations of 3 g/L and 20 mg/L, respectively. Fig. 1 shows a

* Corresponding author.

E-mail address: bentoumig@aecl.ca (G. Bentoumi).

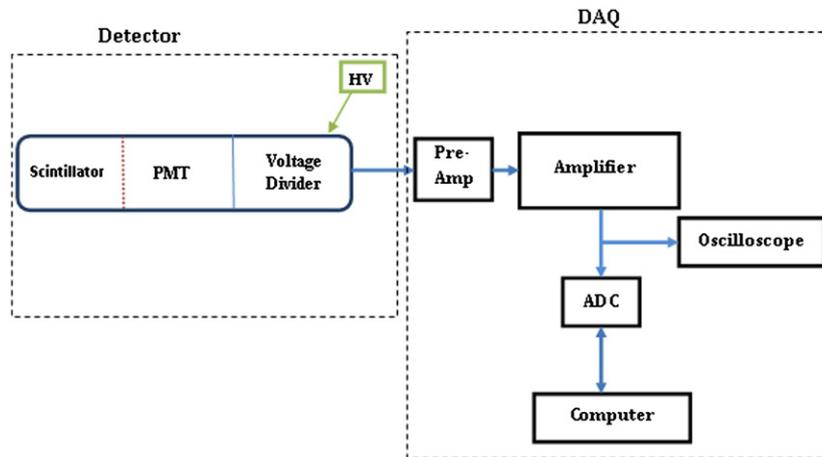


Fig. 1. Experimental setup for scintillator characterization.

scheme of the experimental setup. The LS is filled into a 1 in. diameter and 2 in. tall cylindrical glass container. To maximize the photon collection, optical grease is used in-between the container and the PMT while the remaining container's faces are wrapped with aluminum foil. Under radiation excitation, the energy released in the scintillator by the incident particle transforms to a certain number of optical photons [4,5]. The PMT then converts a fraction of the optical photons into a measurable electrical pulse. For the energy spectrum collection, the signal from the PMT is connected after amplification to a versa module Europa (VME)-based electronic setup for data acquisition (DAQ). It has the function of processing and classifying the electrical pulses according to their total charge. Its main component is the analog/digital converter (ADC) SIS3320 from Struck Innovative Systeme [6]. C++ based software has been developed to control all the operations of the data readout and recording system. When the amplitude of the electrical pulse exceeds the threshold level, a set of mathematical operations is triggered into the ADC module to calculate the area under the pulse. The result is then transferred to a computer to build-up a histogram. In all our experiments, the measured count rate is low. This implies a negligible probability of having multiple events (pile-up). After calibration, a spectrum representing the intensities versus the energy of the incident particles is obtained. Based only on this spectrum it is impossible to identify, without ambiguity, two different particles that deposit the same energy into the scintillator. One of the proposed techniques to achieve discrimination between neutrons and gamma rays is pulse shape discrimination. It requires a temporal resolution of the order of nanoseconds [3]. To explore the PSD technique, the signal from the PMT is connected to a Tektronix oscilloscope DPO 7254 that has a sampling rate of up to 10 GS/s.

3. Pb-212 measurement

The radioisotope ^{212}Pb was loaded into the LAB-based LS by bubbling a gas stream containing ^{220}Rn from a ^{228}Th column generator. ^{212}Pb and its daughters (^{212}Bi , ^{212}Po and ^{208}Tl) are alpha and beta emitters. Samples with activities of 50 Bq were regularly obtained. Fig. 2 shows the energy spectrum as collected with the DAQ system. The spectrum was energy-calibrated using ^{60}Co (1.17 and 1.33 MeV), ^{133}Ba (0.356 MeV), and ^{137}Cs (0.662 MeV) gamma sources. The spectrum is dominated by two well-defined peaks at approximately 0.6 and 0.9 MeVee (MeV electron-equivalent) and a continuum background. The two peaks were attributed to two alpha particles with energies of 6.20 and 8.95 MeV, respectively. The first one is emitted subsequent to the decay of ^{212}Bi while the other

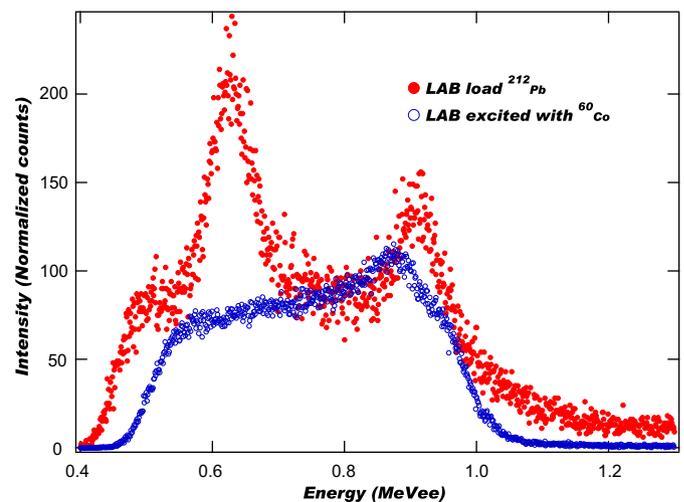


Fig. 2. ^{212}Pb Energy spectrum detected by LAB based scintillator.

results from the decay of ^{212}Po . We notice a quenching factor of approximately 10 between electron and alpha particles in this energy range. This observation is in good agreement with reported values [7,8]. The background is attributed to the different beta particles released by the ^{212}Pb decay chain.

Because the alpha particle at 8.95 MeV has twice the probability of being emitted than the alpha at 6.20 MeV, it is expected that the 0.9 MeVee peak would be twice as intense as that at 0.6 MeVee. However, and as shown in Fig. 2, we observe experimentally a very different trend. This unexpected result is attributed to the response function of the electronic system. The alpha particle at 8.95 MeV always follows a beta emission. It has a large probability to be emitted within a period of 299 ns (half life of ^{212}Po) after the emission of the beta particle. When a beta particle triggers the DAQ system, a dead time of the order of microseconds is required for the electronic system to recover for the registration of a new event. Consequently any alpha particle emitted during the dead time is not registered and will not contribute to the histogram in Fig. 2.

4. Pulse shape discrimination

The pulse shape discrimination (PSD) technique was used to discriminate between neutrons and gamma rays. According to Eq. (1), using a ^{10}B loaded scintillator, the detection of the thermal neutrons is equivalent to detecting alpha particles

whereas the detection of gamma rays is based on the detection of Compton-scattered electrons similar to beta particles [1]. Therefore, the isotope ^{212}Pb can be used to emulate the same environment a neutron detector would have to face in a mixed radiation field, i.e. detecting neutrons in the presence of an intense gamma ray background. In the decay chain of ^{212}Pb successive alpha and beta particles are chosen that are separated by a half life of 299 ns. The oscilloscope was therefore configured in the A-B trigger mode to record only two successive pulses within a window of 500 ns and pulse height greater than 8 mV for beta particles and to 100 mV for alpha particles. As shown in Fig. 3, the generated electrical pulse can be well described by a convolution of two exponential functions with the time resolution of the measuring system:

$$I(t) = \left(\sum_{i=1}^2 A_i \cdot \exp(t-t_0)/\tau_i \right) \otimes R(t) \quad (2)$$

where τ_i is the decay time constant of the exponential decay function i and A_i is the contribution of this component such that $A_1 + A_2 = 1$; t_0 is the time at which the exponential decay starts. $R(t)$ is the time resolution function of the system and can be approximated by a Gaussian curve with a full width at half maximum (FWHM) of less than 2 ns.

As it has been shown by Marrodán et al. [2,11] the two decay time constants depend strongly on the concentration of PPO and the oxygen impurity in the scintillator. We have found on the average decay time constants of $\tau_1 = 4$ and $\tau_2 = 13$ ns; in good agreement with the values reported in the literature for such PPO concentration in LAB [2]. The second decay exponential ($\tau_2 = 13$ ns) is called the tail of the pulse. Its relative amplitude (A_2/A_1) tends to increase when the scintillator is excited by a neutron or an alpha particle. This means that an alpha pulse lasts longer than a beta pulse. For quantification of this phenomenon we have defined a ratio R between the area under the tail (S_{tail}) calculated from t_Q to t_{end} and the total area under the pulse (S_{total}) calculated from $t=0$ to t_{end} :

$$R = \frac{S_{\text{tail}}(t_Q \rightarrow t_{\text{end}})}{S_{\text{total}}(t=0 \text{ ns} \rightarrow t_{\text{end}})} \quad (3)$$

Fig. 4 shows the raw data for the ratio R versus the total area under the detected pulses created by the alpha and beta particles. The time t_Q was chosen so that there is minimum contribution of the first decay exponential to the area under the tail. t_Q was fixed

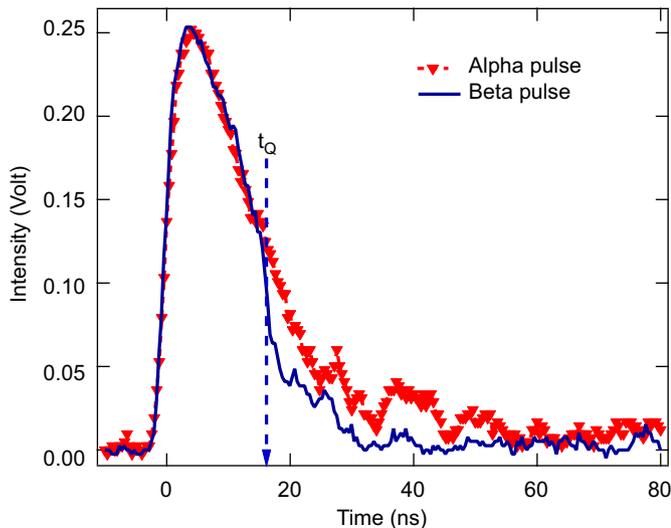


Fig. 3. Alpha and beta pulses captured by the DPO7254 oscilloscope. For comparison the beta pulse has been normalized to the maximum of the alpha pulse.

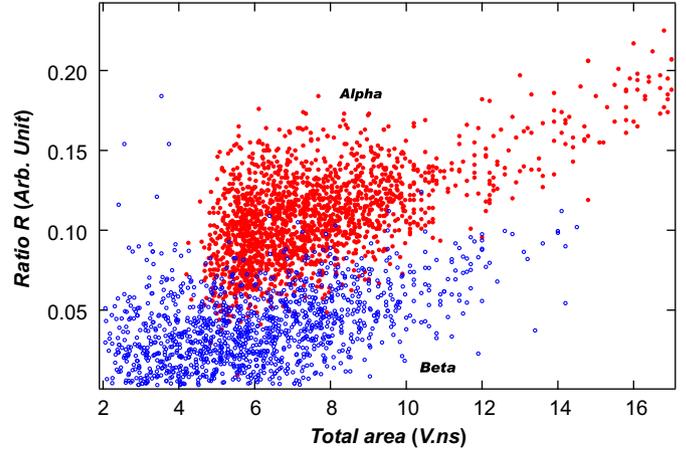


Fig. 4. Ratio R for alpha and beta pulses.

to nearly 4 times τ_1 . On the whole, the alpha pulse has a larger ratio R . Published results for other type of scintillators [3] observed the same effect and attribute it to the increase of the scintillator phosphorescence. Indeed, it has been shown that the contribution from the tail part to the total pulse area is much more important when the scintillator is excited by an alpha particle [2].

To show this result more clearly, a statistical averaging was performed on the raw data. First, the data in Fig. 4 were classified into 140 equal-width groups according to total area. Then, for each group average values of R were found for alpha and beta particles, and plotted at the center of the group range of total area. This process yielded the plot in Fig. 5, which clearly shows a separation between alpha and beta pulses. In the region of interest, where most of the alpha pulses have their total areas ($5 < S_{\text{total}} < 10$), the ratio R for alpha and beta particles scatters in a range ± 0.01 . Overall, the value of R for alpha particles is 0.07 greater than that for beta particles. To quantify these results we have defined the figure of merit (FOM) as follows:

$$\text{FOM} = \frac{(\text{Alpha}-\text{Beta})_{\text{separation}}}{\text{Bandwidth}(R_{\text{alpha}}) + \text{Bandwidth}(R_{\text{beta}})} \quad (4)$$

This leads to a FOM factor of 1.75 for the LAB-based scintillator. The reported FOM values for commercial scintillators extend from 0.8 to over 2.2 [12,13]. Improvements to the LAB-based scintillator could be made by increasing the phosphorescence part in the optical emission. As suggested by Marrodán [2] it is possible to tune the optical time constants and the phosphorescence amplitude (A_2) by varying the PPO and Bis-MsB concentrations. Furthermore, bubbling nitrogen into the scintillator, to remove oxygen impurity, for at least 2 h prior to the measurement is a good practice that has to be considered.

5. Neutron measurements

In order to test the performance of the LAB-based liquid scintillator in a neutron beam, 5% of *o*-carborane ($\text{C}_2\text{H}_{12}\text{B}_{10}$) containing natural boron was loaded into the LS. The tests were performed on the D3 reflectometer at the National Research Universal (NRU) reactor in Chalk River, Canada. We used a focusing pyrolytic graphite (PG) monochromator at a neutron wavelength of 2.37 Å ($E=14.56$ meV), along with a PG filter to reduce the higher order contributions $\lambda/2$ and $\lambda/3$. The same system, as sketched in Fig. 1, was used to record total charge produced in the LS by ^{10}B neutron capture. Gamma rays could also, by Compton effect, cause a scintillation process. For this

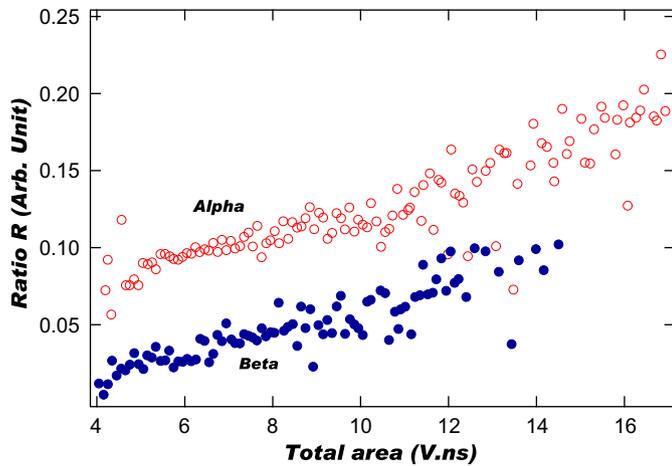


Fig. 5. Average of ratio R for alpha and beta pulses.

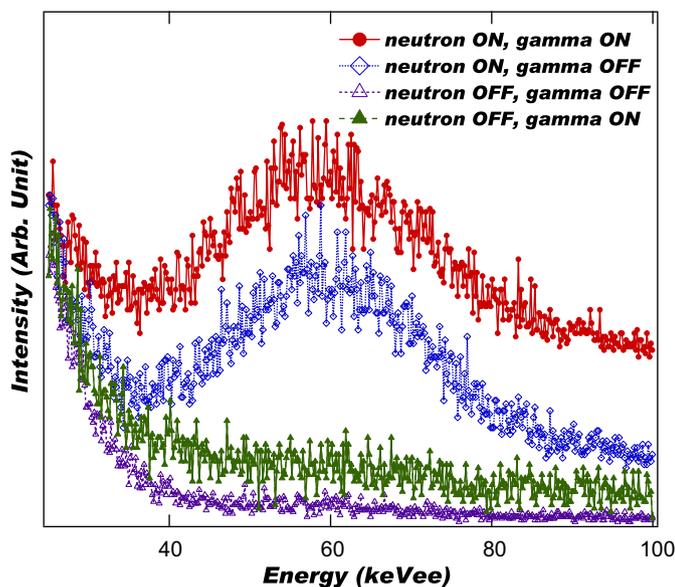


Fig. 6. Energy spectra recorded in four conditions by the boron loaded LAB based scintillator.

reason and in order to extract the neutron contribution in the detected signal, four types of measurements were performed using combinations of a cadmium sheet of 1 mm thickness (thermal neutron beam is attenuated by a factor of 5.10^6) and a lead brick of 50.8 mm thickness (gamma rays are attenuated by a factor of 60). For a good statistics, each spectrum was recorded for about 5 min. A ^{60}Co source was then used to calibrate each energy spectrum. As seen in Fig. 6, a broad peak (FWHM ~ 20 keV) is observed around 60 keVee (keV electron-equivalent) when the scintillator is excited by the neutron beam. This peak disappears completely when the neutron beam is turned off. As stated in Eq. 1, ^{10}B neutron capture leads to creation of an alpha particle with an average energy of 1.48 MeV. It was demonstrated early that in liquid scintillator, the quenching factor for 1–2 MeV alpha particles is in the range of 20–25 [8,9]. Hence the ^{10}B neutron

capture is expected to generate a peak in the range 60–75 keVee. Based on the correlation between the presence of neutrons and the expected equivalent energy for ^{10}B neutron capture [2], the peak in Fig. 6 can be definitely attributed to neutron capture by ^{10}B . The peak position corresponds to an alpha quenching factor of 25, which is in good agreement with published data for other organic scintillator [8,9]. To our knowledge, this is the first time such a measurement has been made with a Boron-loaded LAB-based scintillator. It is interesting to note that the position of the peak does not change with neutron energy. Also, as we can see in Fig. 6, gamma rays produce a very low background. This is very encouraging for the use of ^{10}B -loaded LAB-based scintillator for neutron detection in the presence of a high gamma ray background [10].

6. Conclusion

In this experimental work, we have characterized a liquid scintillator based on the LAB solvent by using internal and external sources. The results confirm the principle of using LAB-based liquid scintillator for neutron detection. Using the ^{212}Pb source to emulate the ^{10}B neutron capture, we have shown the capabilities of Pulse Shape Discrimination in the LAB-based scintillator. The results demonstrate that the optical emission due to an alpha particle has a longer time constant than that due to a beta particle. A figure of merit of 1.75 has been found for the scintillator. Measurements with a neutrons beam of energy equals to 14.56 meV have shown a new peak around 60 keVee due to the neutron capture by ^{10}B .

Acknowledgments

This work was supported in part by a grant from the Natural Science and Engineering Research Council (NSERC) of Canada. The research presented herein was made possible by a reflectometer jointly funded by Canada Foundation for Innovation (CFI), Ontario Innovation Trust (OIT), Ontario Research Fund (ORF), and the National Research Council Canada (NRC).

References

- [1] F. Glenn, Knoll, Radiation detection and measurement, 4th ed., John Wiley & Son LTD, USA, 2010.
- [2] T. Marrodán Undagoitia, F. von Feilitzsch, L. Oberauer, W. Potzel, A. Ulrich, Review of Scientific Instruments 80 (2009) 043301.
- [3] L.F. Miller, J. Preston, S. Pozzi, M. Flaska, J. Neal, Radiation Protection Dosimetry 126 (2007) 253.
- [4] M. Yeh, A. Garnov, R.L. Hahn, Nuclear Instruments and Methods in Physics Research A 578 (2007) 329.
- [5] A.G. Piepke, S.W. Moser, V.M. Novikov, Nuclear Instruments and Methods in Physics Research A 432 (1999) 392.
- [6] <<http://www.struck.de/>>.
- [7] L. Donald, Horrocks, The Review of Scientific Instruments 35 (1964) 334.
- [8] K.F. Flynn, L.E. Glendenin, E.P. Steinberg, P.M. Wright, Nuclear Instruments and Methods 27 (1964) 13.
- [9] H.M. O'Keefe, E. O'Sullivan, M.C. Chen, Nuclear Instruments and Methods in Physics Research A 640 (2011) 119.
- [10] Hyeon-Soo Wuon-Shik Kim, Ki-Hwan Kim, Yong-Uhn Kim, Kim, Ki-Hyon Kim, Journal of Radioanalytical and Nuclear Chemistry 215 (1997) 257.
- [11] CAEN, Digital Gamma Neutron Discrimination With Liquid Scintillators, CAEN, Viareggio, Application note AN2506, April 2011.