Simulation and design of a neutron detector based on Boron-Loaded linear alkyl benzene (LAB) liquid scintillator

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<u>Abstract</u>

A Boron-Loaded linear alkyl benzene (LAB) liquid scintillator (LS) neutron detector has been designed to detect neutrons in high gamma field environment. The detector is made robust by piping the light from a remotely located LS module by an optical fibre. Here we describe a GEANT4 based model to optimize the design of the LS detector. This model includes the physics of neutron interaction with Boron-10, light scintillation by the LAB and light transport in the optical fiber. All the detector components including the scintillator, light guides and an approximation of the photomultiplier tube response, are simulated. The results show that for unidirectional beam of thermal neutrons, a small detector with 70 % neutron detection efficiency can be achieved by loading the LAB with 4.5% Boron-10 and by using a 2 meter optical fibre. The simulated output results are compared to actual measurement.

1. Introduction

Neutron detection has broad applications in many fields. Measuring neutron flux is critical for a nuclear reactor to operate properly; for non-proliferation systems they can be used to identify nuclear materials; security systems can also rely on neutron detectors to trace illicit nuclear material; in radiological protection as a personal dosimeter; finally they can also be useful for waste management organisations to detect contamination [1, 2].

There is a need in the nuclear industry for small, robust, field deployable, thermal neutron detectors that can measure small neutron fluxes in an environment with a relatively large background of gamma rays. Helium-3 filled ionization chambers tubes have been extensively used for this purpose because of their good neutron-to-gamma discrimination capability. However, the industry is currently experiencing a global shortage of Helium-3. As a result, it is necessary to design neutron detectors relying on mechanisms such as boron-loaded liquid scintillators that are different, but just as effective as He-3 detectors.

The mechanisms for detecting neutrons are always based on indirect methods. In fact, two basic types of neutron interactions with matter are generally considered. First, the neutron can be scattered by a nucleus, transferring some of its kinetic energy to the nucleus. If enough energy is transferred, the recoil nucleus ionizes the surrounding material, thereby producing electron-ion pairs that can be detected. Second, the neutron can interact with the nucleus via a nuclear reaction. The products from these reactions are

often ionizing particles, such as protons, alpha, gamma rays, and fission fragments that can also initiate the detection process [3].

Detectors based on nuclear reactions can use solid, liquid, or gas-filled detection media. Although the choice of reactions is limited, the detecting media can be quite varied leading to many options. A Boron-Loaded liquid scintillator (LS) has been suggested as an efficient neutron detector [4]. One of the most important scintillators is linear alkyl benzene (LAB) doped with wavelength-shifters PPO (2.5 diphenyl-oxazonale) and bisMSB (1.4 bis-(o-methyl-styryl)-benzene) [5]. It is produced easily, has a high light output as well as a high flash point (130 °C).

In order to optimize the detector, it is important to simulate its geometry in a radiation transport code. The role of each component of the detector has to be examined. Here, we decided to use GEANT4 to simulate a LAB based neutron detector because it can simulate the necessary radiation/interaction and does the optical photon transport.

GEANT4 stands for GEometry ANd Tracking [6]. It is a Monte Carlo based particle simulation package. It was developed by the European Organization for Nuclear Research (CERN) and can be used to model and simulate the interaction of many different particles with matter. GEANT4 is fully capable of simulating the transport of most particles through any user-defined geometrical arrangement of materials. With the cooperation of scientists throughout the world, many different models have been implemented for use in GEANT4.

In this paper we present a simulation of a neutron detector based on Boron-Loaded linear alkyl benzene (LAB) liquid scintillator using the GEANT4 Monte Carlo code. The impact on the neutron detection efficiency of the geometrical, chemical, and optical parameters will be detailed. In addition a comparison between experiments and simulated results will be discussed.

2. GEANT4 Simulation:

The principle of our neutron detector is based on the (n,α) reaction in boron-10 as follows:

$$B - 10 + n \rightarrow \begin{cases} 6\%: & Li - 7 (1.015 \, MeV) + \alpha (1.777 \, MeV) \\ \\ 94\%: & Li - 7 (0.840 \, MeV) + \alpha (1.470 \, MeV) + \gamma (0.478 \, MeV) \end{cases}$$

The numbers between parentheses are the kinetics energy that the particles take after the reaction.

An incident neutron with energy E_n may be captured by B-10. For thermal neutron the capture cross-section is about 3838 barns. The Q-value of this reaction is 2.792 MeV, with 2.31 MeV going to the charged particles and a 478 KeV to the gamma radiation (94% of the time). The choice of this mechanism is motivated by the high cross section of B-10 and more by the alpha particle that results. Alpha has a short range and therefore will stay in the detector, leading to an emission of a considerable number of photons. Thus, this reaction is monitored to determine the efficiency of the detector, which is defined by the capture rate of the B-10. The energy of the charged particles is deposited in the surrounding medium and converted to scintillation light. This physical process depends strongly on the nature of the particle, on its energy and on the scintillating molecule and solute [4].

Thus a photon emitting scintillator is loaded with Boron-10 to operate as the sensitive medium for neutron detection. The detector's geometry consists of a hollow cylinder filled with the LS Boron-loaded LAB. The hollow aluminum cylinder has 500 μ m wall thicknesses. The design of the detector head is based on three parameters: Boron-10 concentration, the height, and the diameter of the hollow cylinder. All the interior surfaces of the cylinder are reflective except the one on the top, which is totally transparent. At this surface, an optical fiber is placed to collect and transport the light produced by the scintillator to a light sensor such as Photomultiplier Tube (PMT) that transforms photons to an electrical pulse.

In nature, boron has two stable isotopes, B-10 (19.9 %) and B-11(80.1%) with the capture cross-section of B-11 being small compared to B-10. Furthermore the LAB can be loaded by boron to a maximum of 5% by weight. For these reasons, in the simulation, we have kept the mass of boron in the mixture constant to 5% and we have changed the enrichment of boron from 19.9% to up 90%.

GEANT 4 was used to simulate and optimize the above mentioned neutron detector. Although in the present paper the results of a parallel beam of neutrons incident on the detector are presented, different neutron beams with different geometry and energy distributions have been simulated. Figure 1 shows a typical view of the neutron capture simulation. Here, the neutrons are coming from the left in the x direction. All the incident neutrons are simulated within the section of the cylinder in Y-Z plane.





Figure 1: (a) View of the neutron capture simulation. The green lines on the left of the dots are for neutrons trajectories and the yellow ones are for gamma (that results from the decay of excited Lithium 7). (b) Shows a zoom around the point where the capture interaction takes place. The blue line shows the α trajectory and the white one is for Lithium 7.

The process of interaction of neutrons with Boron-10 can be easily set up using the QGSP_BERT_HP physics list provided by GEANT4 [6]. The physics behind this model represents more accurately the different neutron processes. In addition it uses high precision data (cross sections) for neutron transport below 20 MeV down to thermal energies and yields a good agreement with the experimental data.

The neutron capture efficiency is defined for a parallel beam as:

$$\eta = \frac{\text{number of neutrons captured in the cylinder}}{\text{number of neutrons entering in the cylinder}} \pm \Delta \eta$$
(1)

Where $\Delta \eta$ is the statistical standard deviation and is defined by Poisson Statistics:

$$\Delta \eta = \sqrt{\frac{\eta}{N}} \tag{2}$$

N is the numbers of events (or trials).

Forecasting the performance of the detector relies on three parameters that were examined separately: Boron-10 enrichment, radius of aluminum cylinder and neutron energy. In Figures 2 to 4, one can find the results we obtained respectively for the cases where variations in the radius of the detector, B10 enrichment, and the neutron energy spectrum were considered. Each simulation was run for 1 million neutrons. The statistical error for these simulations was calculated according to equation 2 and it is less than 0.1%.



Figure 2: Neutron capture efficiency versus the detector radius. The mass fraction of B-10 and the neutron energy are taken to be 4.5% and 0.025 eV respectively.



Figure 3: Neutron capture efficiency versus the B-10 concentration. The cylinder radius and the neutron energy are taken to be 2.5 mm and 0.025 eV respectively.



Figure 4: Semi-Log plot of detection efficiency of neutron versus neutron energy. The B-10 mass fraction and the detector radius are taken to be 90% and 2.5 mm respectively.

The capture efficiency increases with the radius and B-10 enrichment until it reaches a plateau. This is because the neutrons must travel several mean free paths before they are all absorbed in the low density LAB (0.86 g cm⁻³). This is ensured either by increasing the size of the detector or decreasing the mean free path of the neutron by increasing its absorption cross section (higher B-10 enrichment). On the other hand the capture efficiency decreases with the neutron energy and is practically zero above 100 keV. This behaviour is explained by the decrease of the absorption cross section of B-10 which has a $1/\sqrt{E_n}$ dependence [3]. The slowing-down process for neutrons is not taken into account in this simulation. Accordingly, a higher B-10 concentration and a larger detector radius yield a better capture efficiency. This conclusion remains valid for other kinds of neutron sources such as a point, volumetric or planar diffuse sources. Due optical fibre availability and the planned application, to reduce the gamma sensitivity, we have fixed the cylinder radius to 1 mm and the boron-10 concentration to 4.5%. This ensures a capture efficiency of 70%.

3. Light propagation:

Following the neutron interaction with the B-10, the energy deposited by the resultant charged particles (α and Li-7) is converted to scintillation light. The alpha particle will transfer all its energy within a range of micrometers to LAB molecules. The LAB molecules then transfer the energy to the fluor (PPO) and wavelength shifter (Bis-MSB). The amount of scintillation is well described by the Birks' semi empirical law [4, 7]:

Luminescence 🌣 Energy dissipated in scintillator

L = S.E Or, in differential form $\frac{dL}{dr} = s \frac{dE}{dr}$ (3)

Where S is the scintillator efficiency, E the energy dissipated in the scintillator and L the number of photons emitted.

In LS LAB, we estimated that an alpha particle produces nearly 750 photons per MeV [8, 9]. The heavier particles don't produce light even if they have high kinetic energy [4], so we can neglect the light output from the lithium-7 produced in the neutron interaction with B-10.

The key ingredient in a successful detector is the number of photons that reach the PMT. A certain fraction of them will interact with the photocathode material, producing a certain average number of photoelectron. The probability that a measurable pulse at the anode will be generated increases with the number of photoelectrons [4]. Consequently, we gain significantly by optimizing the photon collection system.

For this simulation, we consider an optical fiber with a refractive index of 1.60 for the core and 1.49 for the cladding. The core has a radius of 1mm and the cladding is 0.1 mm thick. The length of the fiber is fixed to 2 m. One end of the fibre is coupled to the top of the LAB filled aluminum cylinder and the other end is coupled to the PMT.

In GEANT4, the scintillation process is well supported by the G4Scintilation module based on the theory of light emission. Light transport is based on Maxwell and Fresnel equations. In order to increase the number of photons collected in the front of the PMT, many detector configuration were analyzed taking into consideration the diameter of the LAB cylinder compared to that of optical fiber, the technique used to couple the fiber to the LAB cylinder, the properties of the inside surface of the cylinder (reflectivity, diffuse and/or specular reflection) and the height of the LAB cylinder.

As shown in Table 1, photons collection efficiency is very sensitive to the interior surface total reflectivity (R_{tot}) (sum of diffuse (R_d) and specular (R_s) reflectivity). It increases by a factor of 7 by varying the total reflectivity from 50% to 98%. It also increases in average by a factor of 30 % when passing from pure to 70% specular reflection. Theoretically we can achieve a photon collection efficiency of 9.45 % by considering a cylinder having an internal surface that has a total reflectivity of 98 %, 70% of it being specular.

Photons collection Efficiency (%)					
	$R_{s}=0$ %	$R_{s} = 30 \%$	$R_{s} = 50 \%$	$R_{s} = 70 \%$	$R_s = 100 \%$
	$R_d = 100 \%$	$R_d = 70 \%$	$R_d = 50 \%$	$R_d = 30 \%$	$R_{d}=0$ %
$R_{tot} = 0 \%$	0.55	0.55	0.55	0.55	0.55
$R_{tot} = 50 \%$	0.96	1.06	1.17	1.28	1.58
$R_{tot} = 85 \%$	2.07	2.5	3.19	3.20	4.11
$R_{tot} = 98 \%$	7.59	8.78	9.19	9.45	7.37

Table 1: Photon collection efficiency for different total reflectivity and specular/diffuse reflection. The cylinder is 1 mm in radius and 2 cm in height. The fiber has a 1mm radius and is 2 meter long. R_{tot} , R_s and R_d are total, specular and diffuse reflectivity respectively.

Another important result is the photons collection efficiency versus the cylinder height. As shown in Figure 5, even if the collection efficiency decreases with the cylinder height, the total number of photons collected increases until saturation with an increase in the cylinder height. This result can be understood by the fact that the probability for a photon to be absorbed by the detector wall increases with the distance between the location where it is born and the fibre. Above a certain distance, photons are totally absorbed by the cylinder's wall because they undergo a considerable number of reflections and do not reach the entrance to the optical fiber. As Figure 5 shows, 200 mm is the optimal length for the detector.



Figure 5: Variation of the photon collection efficiency and photons numbers on the PMT as function of the cylinder height containing the LAB. The results are for infinite size of a neutron beam (1 neutron/cm) and a cylinder with a total reflection of 98%, 70% of it being specular.

Figure 5 shows that in an environment with infinite size of a parallel neutron beam (size of the beam is bigger than the size of the detector), it is better to use a head detector with up to 200 mm height. On average 210 photons can reach the photocathode of the PMT. If the beam size is small compared to the detector, it is more functional if we guarantee a neutron interaction in the top of the detector near the optical fiber. On average 32 photons on PMT photocathode can be collected. In the two cases and with the moderns PMTs, a pulse can be generated leading to a true count. In practice, matching the height of the detector head to the beam size is an important factor to consider for successful detection.

4. Experiment:

To validate the GEANT4 calculation, an experiment setup for radiation detection (neutron and gamma) has been built. A sketch of it is shown in Figure 6.



Figure 6: Experimental setup for gamma/neutron detection.

A cell of LS LAB has been optically coupled to a PMT. The cell was exposed to a Cobalt-60 source that emits gamma rays at energy 1.173 and 1.333 MeV. A fraction of the light output is collected by the PMT (9266B electron tubes model). The LS LAB cylinder is 50.8 mm in diameter and has a height of 7.62 mm.

In parallel a GEANT4 simulation was conducted for this experiment. The same procedure was used as that described earlier, except there was no optical fiber and the cell was optically coupled to the PMT. Figure 7 shows a good agreement between the experiment and the simulated result. Neither spectra show the two photo-peaks of the cobalt-60. This is due to the low density of LAB combined to a small size of the cylinder. The gamma radiation does not deposit its entire energy in the LAB. Only the Compton edge can be seen. Furthermore the experimental spectrum agrees with the published data that used other type of material for gamma spectroscopy [3, 10]. This result confirms the principle of detection using LAB as a medium for scintillation.



Figure 7: Comparison between simulated (lozenge) and measured (square) spectra of LAB excited by Cobalt-60.

5. Conclusion

We have simulated a neutron detector based on liquid scintillator based on Boron-10 loaded linear Alkyl-Benzene. An optical fibre attached to a hollow liquid-filled cylinder was simulated. The field deployment is made robust by piping the light from the remotely located sensitive detector through an optical fibre. The simulation results show that it is possible to build a small neutron detector with efficiency up to 70 %. It was demonstrated that the inside surface reflectivity of the housing is a key element in the design of this detector. The initial results confirm the principle of using LAB as the solvent for a liquid scintillator and have shown good agreement between the GEANT4 calculations and the experimental results.

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