GAMBE: Thermal Neutron Detection System Based on a Sandwich Configuration of Silicon Semiconductor Detector Coupled with Neutron Reactive Material

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Abstract

Silicon semiconductor detectors are used efficiently for neutron detection when coated with a suitable material. They detect secondary reaction products resulting from the interaction of thermal neutrons with a neutron sensitive material such as ^{6}LiF. In the present work, the efficiency of the thermal neutron detector system, GAMBE, is discussed. This detector system based on two silicon sensors of 1 cm^2 active area and a layer of ^{6}LiF (1.5 $\pm$ 0.6) mg/cm^2 thick in a sandwich configuration. This arrangement achieves total and coincidence detection efficiency of (4.1 $\pm$ 0.5)% and (0.9 $\pm$ 0.3)% respectively. The coincidence method defines a true neutron hit by the simultaneous signal recorded by the two sensors facing the conversion film. This coincidence methodology is applied to enhance the rejection factor of fake hits due to high gamma background conditions up to 10^8 as discussed in previous work. Geant simulation indicates that total and coincidence detection efficiency up to 55% and 18% are possible using an advanced design of stacked detectors.

Keywords: neutron detector, semiconductor detector, coated semiconductor detector, neutron detection, neutron conversion.

1. Introduction

Detection of thermal neutrons is a fundamental topic in many areas of nuclear science. It is used for varying applications such as reactor instrumentation, special nuclear material detection (SNM) [1], particle physics, material science [2] and radiation safety [3]. However, neutrons are neutral particles which cannot interact with matter by means of Coulomb force, which forms an energy loss mechanism for charged particles and electrons. Also, neutrons can travel through many centimeters of matter without any interaction and thus, can be totally unseen by a conventional detector [4]. Therefore, there are different types of materials which are used as a converter for thermal neutrons, which usually are in thermodynamic equilibrium with the surrounding medium and their most probable energy at a room temperature of 290 K is 0.025 eV. These neutrons interact with the nucleus of these conversion materials and as a result, the neutron may produce secondary radiation, or the energy and direction of this neutron change significantly. Secondary radiation arising from these neutrons interactions are mainly heavy charged particles. These particles can be produced by neutron-induced nuclear reactions, or they may result from the nuclei of the absorbing material itself, which gain enough energy from an eventual collision with a neutron. Consequently, the detection of thermal neutrons depends on this secondary radiation, and most neutron detectors utilise some materials with a large absorption cross section for a high detection efficiency. Moreover, the reaction products must have the capability to leave the material with positive detection energies [5, 6]. The three most often studied neutron reactive materials for such a detector are ^{6}Li, ^{10}B, or natural ^{157}Gd [7].

^{6}Li has a thermal neutron absorption cross section (σ) of 900 b, which decreases by increasing neutron energy [5]. The primary reaction of neutron interactions with ^{6}Li is ^{6}Li(n,α)^{3}H; this reaction produces an alpha particle (at 2.05 MeV) and a triton (at 2.73 MeV) in opposite directions with total Q-value of 4.78 MeV. Although ^{6}Li has a smaller thermal neutron absorption cross section than ^{10}B and ^{157}Gd, the higher energy reaction products make it attractive for thermal neutron detection. Furthermore, the low atomic density and the low mass density of ^{6}Li result in a large reaction product range exceeding the ranges of the reaction products for ^{10}B film, with a sufficient range for triton L_H = 126.77 μm and an efficient range for alpha L_α = 19.05 μm. ^{6}Li could be used in the pure form as a neutron reactive material although it demands cumbersome handling procedures because of its corrosiveness and chemical reactivity. Due to this chemical reactivity of pure ^{6}Li, it could alternatively be used in the form of ^{6}LiF, which is more stable, however, the range of reaction products will be affected with L_H = 29.25 μm and L_α = 4.64 μm [8].

Coated semiconductor diodes such as silicon with a thin film of neutron reactive material have been discussed for decades as neutron detectors [9]. Neutrons interacting in the reactive layer cause the spontaneous ejection of the secondary reaction products entering the adjacent semiconductor detector. This secondary radiation creates numerous electron-hole pairs whose charge can be measured through a shaped voltage pulse. These sensors offer valuable features, such as low weight, bias voltage, battery consumption and a high count rate capability [10]. The high density of the semiconductor allows an optimum de-

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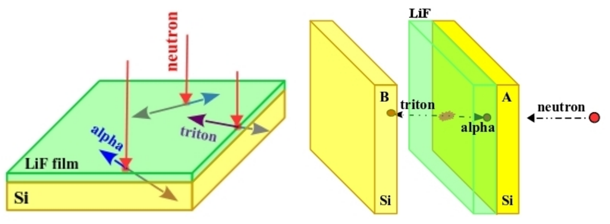
tector compactness, because of the short ranges of the reaction products. That suggests silicon as a best choice for thermal neutron detector also because of the relatively low atomic number of silicon (Z = 14), which decreases gamma-ray interaction probability [11]. In fact, the sensitivity to gamma radiation is expected to be low in Si wafer for a thickness range of 30−300 μm. For instance, Si sensors of 300 μm thick has gamma-ray detection efficiency up to 100% for gamma photons of 10 keV, falling approximately to 1% for 150 keV [12].

In this work, the feasibility of using enriched ^{6}Li with silicon sensors in a sandwich configuration as a thermal neutron counter, called GAMBE, has been investigated. Tests have been performed using ^{6}Li in the form of ^{6}LiF coating film which was applied to a silicon semiconductor radiation sensor where the reaction products will be measured. The basic design consideration was studied using Geant4 simulations to identify the optimal thickness of ^{6}LiF film, which was capable of achieving the highest total and coincidence detection efficiency. In addition, experimental measurements were carried out using an ^{241}Am-^{9}Be neutron source with the detector placed in a particular position 75 cm away from the neutron source. Thus, the thermal neutron flux and the GAMBE detection efficiency were determined at this position as presented. Finally, it has been suggested that using an advanced configuration of a stacked detector will improve the whole GAMBE performance.

2. Geant4 modelling approach

Geant4 is a toolkit for simulating the passage of particles through matter. It includes a complete range of functionality such as tracking, geometry, physics models, and hits. It has been created exploiting software engineering, object-oriented technology and implemented in the C++ programming language. It has been used for a variety of applications in particle physics, nuclear physics, accelerator design, space engineering, and medical physics [13].



(a) Planar structure (2π geometry). (b) Sandwich configuration (4π geometry).

Figure 1: Principle of neutron detection using a) planar Si semiconductor coated with ^{6}LiF film b) GAMBE, thermal neutron detector.

Planar design is the most straight forward adaptation of semiconductor detector for neutron detection. However, it has its limitation. Firstly, the neutron capture probability in the converter is increasing with increasing the converter layer thickness in one hand, but on the other hand, the chance that the neutron capture reaction products will reach the detector sensitive part may be severely reduced with the growing of this converter layer. Therefore, an optimal converter thickness of ^{6}LiF material has to be found. Secondly, only those charged particles which are ejected in the direction of diode interface would be detected. This is known as 2π geometry as presented in fig. 1(a) allowing only up to half of the primary reaction products to generate e−h pairs inside the depletion region of semiconductor material. Hence, sandwich stacking as shown in fig. 1(b) will lead to 4π collection of primary reaction products; as the detection of either alpha or triton becomes possible and this increases the detection efficiency.

Simulations have been performed using Geant4 to identify the optimal ^{6}LiF film thickness where neutron detection efficiency is the highest using a sandwich configuration of two silicon detectors. The sandwich detector geometry consists of two 12.5×12.5 mm^2 silicon diodes which are assigned as A and B with a thickness of 300 μm. The sensitive ^{6}LiF film with an adjustable thickness is located between the two Si sensors where it adheres to sensor A and the two sensors are separated by a 300 μm gap (see fig. 1(b)). There is also a 100 nm Al contact covering the active region of both silicon sensors.

For the simulation, each event begins with the generation of a random position within the sensitive film volume where a thermal neutron will be captured. From this point, one alpha is assigned an arbitrary direction with an energy of 2.05 MeV and a triton is assigned the opposite direction with an energy of 2.73 MeV. This model assumes that neutron capture is distributed uniformly within the converter film. This is a good approximation for the thickness range under investigation because the probability of a thermal neutron absorption would be constant over the entire volume of the converter film. For each event (alpha or triton), the energy deposited in each of the two silicon sensors is computed.

In this simulation the thermal neutron detection efficiency, ε, is calculated by [6]

$$\varepsilon = \frac{n}{N} \times P(x) = \frac{n}{N} \times \{1 - exp(-\frac{N_A}{w_A} \times \rho \times \sigma \times x)\} \qquad (1)$$

where $\frac{n}{N}$ is the ratio of the detected charged particles (n) by Si detector to the number of generated neutrons (N) within the film. $P(x)$ is the probability of an incident thermal neutron being captured as a function of the reactive film thickness, x. N_A is Avogadro's number, w_A the atomic or molecular weight of the reactive film, ρ the density of the reactive film and σ the thermal-neutron cross-section, for ^{6}Li 940 b. A neutron is counted by detecting either alpha or triton as a single or/and a coincident event, this is defined as total detection efficiency of the detector (ε_t). Detecting neutron capture products in coincidence is a method based on detection both of reaction products (alpha and triton) by two Si sensors at the same time and, thus, the coincidence detection efficiency (ε_c) of the detector is defined.

The simulated thermal neutron detection efficiency for a range of ^{6}LiF film thicknesses is displayed in fig. 2 which shows the effect of ^{6}LiF film thickness increment on both total and coincidence detection efficiencies. Since, the detection efficiency depends on both the probability that neutron to be captured and the chance that secondary particles born in the ^{6}LiF film will be capable of reaching the sensitive detector volume. The total and coincidence detection efficiencies increase up to a certain value of a ^{6}LiF film thickness then they will decrease. Results indicate that the optimal film thicknesses for the highest total and coincidence detection efficiencies of 7.5 and 1.1% are 8.14 and 1.16 mg/cm^2 respectively.

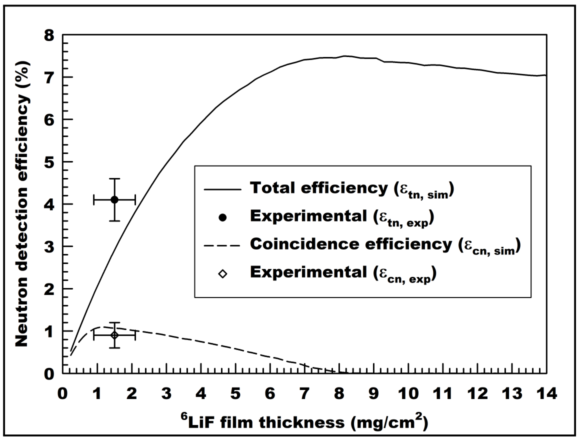


Figure 2: Variation of thermal neutron detection efficiency as a function of ^{6}LiF film thickness.

3. Experimental work

A 6 mol/l ^{6}LiF solution was prepared by dissolving 3 g of ball milled ^{6}LiF powder (Sigma-Aldrich 95% enriched ^{6}Li) in 20 cc ethanol. The ^{6}LiF solution was mixed with a ratio of 1:1 with a solution of 1 w/v% polyvinylpyrrolidone (Sigma-Aldrich PVP, MW 700000) in ethanol with a molar concentration of 1.43×10^{-4} mol/l, which is used as an adhesive material. This mixture of ^{6}LiF/PVP was precipitated on the surface of the Si sensor (total area of 1.25×1.25 cm^2). The mass of ^{6}LiF/PVP solution to be poured on Si sensor substrate is estimated as a function of the required ^{6}LiF film thickness. The mass of the poured solution was measured with a scale, where (2.4 ± 0.1) mg of the mixture was applied to the Si sensor to cover the whole surface area of 1.25×1.25 cm^2. This precipitated mixture was dried at room temperature to avoid cracks and to form a uniform film over the area of Si substrate. In order to characterise the surface roughness of the deposited film, an Atomic Force Microscope (AFM) and a scanning Keyence VHX5000 Digital Microscope (KVDM) were used. It was found that the precipitated ^{6}LiF film on the silicon sensor is not uniformly distributed and has a surface roughness of 2.5 μm. This results in an error in the mass distribution of ± 0.6 mg/cm^2 over the whole area of the formed ^{6}LiF film, which can be considered as a reason for the variation of the detection efficiency. Consequently, the determined thickness of the precipitated ^{6}LiF film is (1.5 ± 0.6) mg/cm^2.

Experimental measurements have been carried out using sandwich detector configuration with and without neutron converter material, in order to differentiate between neutron and gamma-ray events. ^{6}LiF film (1.5 ± 0.6) mg/cm^2 thick has been used as a neutron converter in this sandwich configuration (see fig. 1(b)). In each measurement, the whole GAMBE assembly was oriented in such a manner that coated Si diode sensor A was back irradiated by thermal neutrons whereas sensor B was facing them. The entire sensor-converter system was mounted in an aluminium box designed to eliminate photoelectric noise as well as to decrease the effect of gamma-ray background. The detector has been placed in a particular position in front of a 1 Ci ^{241}Am-^{9}Be neutron source, where neutrons are emitted as part of the reaction Be(α,n)C*.

Thermal neutron flux has been measured using the ^{3}He detector tube as can be seen in fig. 3. The tube is 50 cm away from the end of the neutron tank, and the 1 Ci ^{241}Am-^{9}Be neutron source is 25 cm inside the water tank. This position is referred as the calibration position. The 3He detector tube is an industry standard 2 in. (5 cm) diameter, 36 in. (90 cm), active length, filled to a pressure of 2 atm, and operating at a high voltage of 1100 V. Its typical thermal neutron detection efficiency is > 60%. Furthermore, the neutron sensitivity of these detectors is 236 cps/nv (nv is thermal neutron flux, neutrons/cm^2/s). Approximately 3 cps/nv per cm active tube length assuming no degradation of performance over the lifetime of the detector.

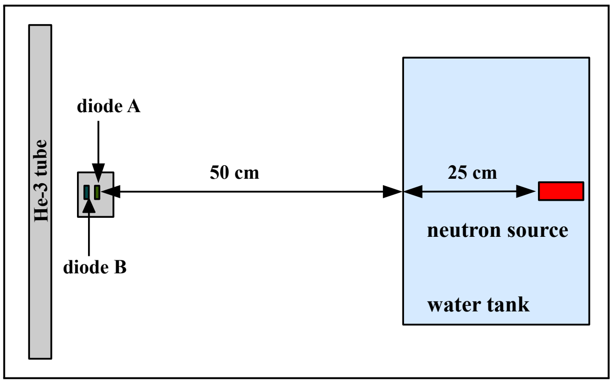


Figure 3: Experimental layout where the detector is at the calibration position.

Another detector such as the NMS017NG3 neutron survey monitor has been used to characterise the variation in the neutron flux along the length of the ^{3}He detector tube. This detector has demonstrated that the neutron flux at the center of the ^{3}He detector tube is 1.40 times greater than over the entire length of the ^{3}He detector tube.

4. Results and Discussion

4.1. Thermal neutron flux

Thermal neutron flux has been determined at the calibration position corresponding to a detection rate of (1013.01 ± 0.07) cps by ^{3}He thermal neutron detector. As a result, the determined neutron flux is 6 nv (n/s/cm^2). It is this figure that has been used to calculate the total and coincidence detection efficiency of the thermal neutron detector.

4.2. GAMBE detection efficiency

The detection efficiency has been determined by integrating the area under the curve for both total and coincidence events and then dividing the results by the neutron flux (n/cm^2/s) through the converter film. All events belonging to gamma-ray interaction with the sandwich detector of bare silicon sensors, either they are single or coincidence events have been subtracted from the obtained energy spectrum as presented in fig. 4.

This subtraction affects the count rate especially at lower energy range where there is a possibility of a combination between α-particle and γ-ray interaction with silicon detector. However, this step insures that the evaluated detection efficiency is an absolute thermal neutron detection efficiency.

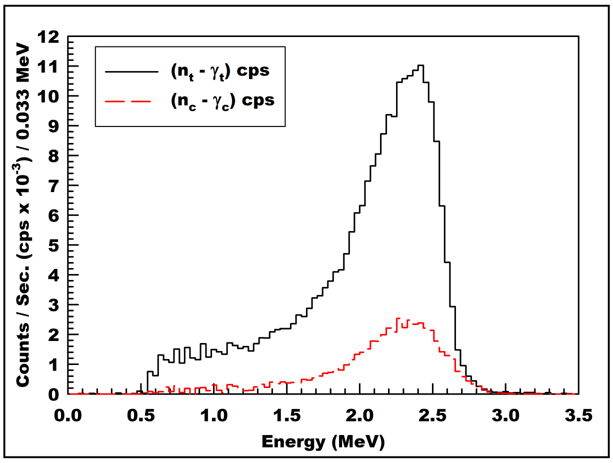


Figure 4: Spectra of energy deposited in both Si sensors of the detector as a function of a ^{6}LiF film (1.5 $\pm$ 0.6) mg/cm^2 thick.

Results show that ^{6}LiF film of (1.5 $\pm$ 0.6) mg/cm^2 thick in a sandwich detector configuration can achieve total and coincidence detection efficiency of (4.1 $\pm$ 0.5)% and (0.9 $\pm$ 0.3)% respectively. The results are compatible and in agreement with the theoretical investigation as displayed and compared in fig. 2, where the variation of the surface roughness of the converter film has been taken into consideration. Although, the coincidence detection methodology affects and decreases the detection efficiency of, GAMBE, thermal neutron detection system, It provides a very good method for rejecting the spurious hits coming from gamma photons, which are usually present in the neutron field under measurement.

5. Neutron detection efficiency enhancement

A method has been proposed to improve the thermal neutron detection efficiency by using a stacked detector configuration. The concept of this method was based on eq. (1) by introducing a new factor m in the probability of thermal neutron absorption $P(x)$. Where m represents the number of detectors of sandwich configuration with a converter material in the stack as depicted in eq. (2).

$$\varepsilon = \frac{n}{N} \times P(x) = \frac{n}{N} \times \{1 - exp(-\frac{N_A}{w_A} \times \rho \times \sigma \times x \times m)\} \quad (2)$$

Hence, the purpose of multi-layers approach is to increase the thermal neutron detection efficiency by increasing the active area where the neutron could be captured. However, The detection efficiency is not expected to scale linearly with increasing the number of stacked detectors. This is because the initial neutron flux will be attenuated by each of the neutron sensitive ^{6}LiF film and as a result, the neutron flux decreases for each subsequent detector. Moreover, in each reactive layer a proportion of incident neutrons are captured and not all result in a detected event. It should be pointed out that the rate of increment in the detection efficiency reduces, as the number of detectors in the stack increases.

From the modelling, it is expected that a multiple-layer proposal of 20 sandwich detectors can achieve total and coincidence detection efficiencies up to 55 and 18% respectively. The advanced stacked detectors design not only affects the detection efficiency but it also influences the optimum ^{6}LiF film thickness for a particular number of sandwich detectors in the stack. It is found that the optimum thickness for the individual ^{6}LiF neutron reactive films decreases as the number of detectors in the multiple layers configuration increases. The results of this study also indicate that for a specific number of sandwich detector there is an ideal thickness of ^{6}LiF film that maximises the total and coincidence detection efficiencies as displayed in figs. 5 and 6 respectively. In addition, figs. 5 and 6 illustrate that the range of ^{6}LiF thickness is dependent on which type of the detection efficiency will be used for neutron identification. Finally, the overall device design should incorporate a reasonable number of detectors compatible with the available power supply and output electronics.

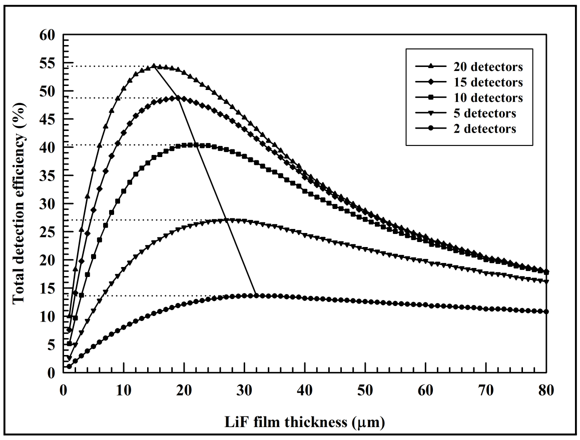


Figure 5: Total detection efficiency as a function of enriched ^{6}LiF film thickness and number of layers.

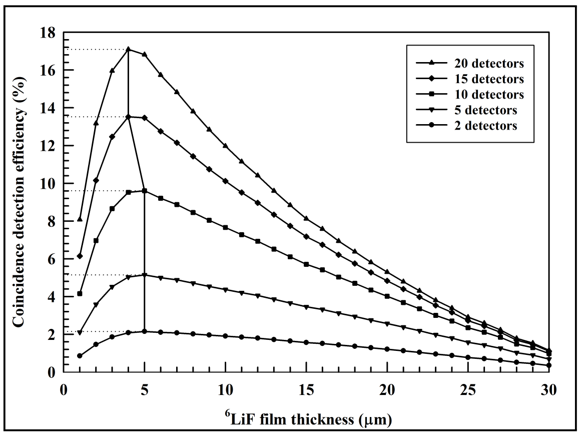


Figure 6: Coincidence detection efficiency as a function of enriched ^{6}LiF film thickness and number of layers.

6. Conclusion

The aim of the present research was to examine the practicality of using enriched ^{6}Li as a ^{6}LiF film with silicon sensors around 1 cm^2 active area in a sandwich configuration as a thermal neutron detector. The expected efficiency for this sandwich style has been described as a function of ^{6}LiF converter film thickness. For instance, the sandwich configuration allows for optimised total and coincidence detection efficiencies of 7.5 and 1.1% corresponding to ^{6}LiF film of 8.14 and 1.16 mg/cm^2 respectively. Tests have been performed using ^{6}LiF film of (1.5 ± 0.6) mg/cm^2 thick to examine the coincidence detection efficiency which will be capable of enhancing gamma-ray rejection factor. The detector was capable of achieving total and coincidence detection efficiencies of (4.1 ± 0.5)% and (0.9 ± 0.3)% respectively. In addition, if the detector stacking technique is used, dramatic increases in thermal neutron detection efficiency can occur. For instance, stacking 20 individual sandwich detectors can increase the total and coincidence detection efficiencies up to 55% and 18% which are comparable to the detection efficiency of ^{3}He detector tubes. Hence, the future work will focus on building the neutron detection system, GAMBE, which has a multilayer configuration in order to validate the theoretical study and to investigate the behaviour of the stacked detector with the background gamma radiation.

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