Title: 6Li Foil Thermal Neutron Detector

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ABSTRACT

In this paper we report on the design of a multilayer thermal neutron detector based on $^6$Li reactive foil and thin film plastic scintillators. The $^6$Li foils have about twice the intrinsic efficiency of $^{10}$B films and about four times higher light output due to a unique combination of high energy of reaction particles, low self absorption, and low ionization density of tritons. The design configuration provides for double sided readout of the lithium foil resulting in a doubling of the efficiency relative to a classical reactive film detector and generating a pulse height distribution with a valley between neutron and gamma signals similar to $^3$He tubes. The tens of microns thickness of plastic scintillator limits the energy deposited by gamma rays, which provides the necessary neutron/gamma discrimination. We used MCNPX to model a multilayer Li foil detector design and compared it with the standard HLNCC-II ($18^3$He tubes operated at 4 atm). The preliminary results of the $^6$Li configuration show higher efficiency and one third of the die-away time. These properties, combined with the very short dead time of the plastic scintillator, offer the potential of a very high performance detector.

INTRODUCTION.

Comparison of $^{10}$B and $^6$Li based neutron-capturing films: Neutron measurements are commonly used in Nuclear Safeguards and $^3$He detectors are the backbone technology of these measurements. The increasing needs for neutron detection and the current shortage of $^3$He gas have lead to rigorous R&D efforts on alternative neutron detection technology. The unique combination of $^3$He properties (near 100% efficiency for thermal neutrons, inherent insensitivity to gain and HV variations, and reasonable tolerance to gamma radiation) present a real challenge to candidate replacement technologies. This challenge is especially severe for neutron coincidence counting applications where high detection efficiency, low dead time, short die-away time and high stability are essential. Two most common thermal neutron capturing materials ($^{10}$B and $^6$Li) have become a major object of research for detector technology. Because of lithium metal’s chemical reactivity, the compound LiF is commonly used for detector development. The maximum calculated thermal neutron efficiency for single-ended readout is about 4% with $^{10}$B at ~10 μm film thickness and 4.3% with $^6$LiF films at ~25 μm [1]. For pure $^6$Li films, the maximum thermal neutron detection efficiency of 11.6% is obtained for film thickness of ~95 μm [1]. While the short range of $^{10}$B alpha particles makes this material advantageous for $^{10}$B-lined gas proportional counters, $^6$Li is preferred choice for scintillation detectors also for the better light yield due to the secondary particle produced.
The emerging off-the-shelf technology based on $^{10}\text{B}$-lined proportional counters [2,3] is obtaining the efficiency of $^3\text{He}$ detectors at the cost of generating overlapping neutron and gamma distributions as well as creating signal processing electronics challenges stemming from the lack of true plateau and the higher capacitance of multiwire anode configuration. For scintillation applications, the most common use has been neutron sensitive ZnS(Ag)$^{6}\text{LiF}$ scintillation sheets sandwiched with wavelength shifters [5]. Using ZnS(Ag)$^{6}\text{LiF}$ allows up to 9mg/cm² areal density of $^6\text{Li}$, but, because of severe charge absorption in LiF and light self-absorption in ZnS scintillator, only a small fraction of an incident particle’s energy is converted and transported to the PMTs as a light signal. The pulse height spectrum has a $1/E$ energy distribution, with most of the signal coming from noise and gammas [5] Gamma discrimination is achieved by excellent pulse-shape discrimination property of the ZnS(Ag)$^{6}\text{LiF}$ scintillator. The mismatch of refraction index between ZnS and nonscintillating converters and light guides additionally reduces the light transport along the lightguide. Recent status of this technology has been reported on 2010 SORMA conference [6,7]. A step forward in using ZnS/LiF is its sandwiching between light-guide strips instead of non-scintillating fibers [9]. This approach eliminates additional distortion of neutrons pulse height spectrum in wavelength shifting fibers.

In all of these technologies the intrinsic efficiency and quality of neutron and gamma pulse-height distribution are limited by the short range (due to self absorption) of charged particles in the neutron capturing film. Use of pure $^6\text{Li}$ foil as a neutron capturing film could substantially improve the pulse height distribution while maintaining reasonable efficiency, because of the few times longer range of charged particles in lithium metal. Calculations and experimental measurements show an intrinsic efficiency of up to 12% for single-sided readout and nearly twice that much for double-sided readout of charged particles [9]. An alternative technique was used in Ref. [10] with flat sheets of $^6\text{Li}$ metal and double-sided readout with multi-wire anode [11]. Because of microphonics and high anode capacitance, this system had to operate at very high multiplication gain that caused long-term instability due to outgasing from the supporting structure.

**LITHIUM (Li-6) GAS IONIZATION CHAMBER**

**Prior experience with Lithium foil detector:** Several years ago we have built a thermal neutron detector based on $^6\text{Li}$ lined ionization chamber with integrated body-moderator made from high-density polyethylene (HDPE) [12,13]. In this introductory section we describe the basic design and measurements of eight-cell detector array as a background work for our new detection concept. The exploded view of detector parts and final assembly are shown in Fig. 1. Two sets of four ionization chamber cavities are carved into three HDPE slabs. The solid anode plates are suspended by ceramic insulators. The side A and side B covers are sealed to the central slab using O-rings. All surfaces of HDPE except the internal peripheral surfaces are metalized. The internal surfaces (about 600 sq.cm each) of all cavities were laminated with 40 micron thick 95% enriched $^6\text{Li}$ foil. The detector assembly was filled with Ar +3% CH$_4$ gas mixture at one
atmosphere pressure. The field intensity was selected to be about 0.07 kV/cm/torr in order to tolerate increased concentration of hydrocarbons outgassing from the HDPE.

A reference charge with value of 25 fC was injected to each detection cell (in both the A and B sides of detection cells), and a $^{252}$Cf source was placed at the center of detector for 20 minutes and the combined pulse height spectra were recorded (see Fig. 2). The experimental results were compared with the modeled with MCNPX transport code [17]. The measurements show about 13% intrinsic efficiency and in good agreement (better than 10%) with modeling results: based on these results, we can be quite confident in using the Monte Carlo simulations in the design of Li-foil thermal neutron detector. The aging effects were estimated by comparison between pulse height spectra taken at the time of initial filling of detector and six months later. Based on prior experience, the most intensive out-gassing occurs during the first couple months after assembly of the detector. The pulse height spectra shown in Fig. 2 show a few percent gain degradation, but because most of the pulse height spectrum is above the event threshold this change does not affect the neutrons counting rate.
This work with the $^6$Li ionization chamber demonstrates the technical feasibility of using lithium foil for large area neutron detectors and the capability for handling the reactive lithium metal. Based on that experience we have developed a thermal neutron sandwich detector described in the next section.

**LITHIUM ($^6$Li) FOIL SANDWICH DETECTOR**

**Detection Concept:** This detection concept is similar to that described in Refs. [5,8] except the neutron sensitive ZnS/LiF scintillator is replaced with $^6$Li foil sandwiched between thin scintillating films laminated on long strips of light-guide material (Fig. 3). Multiple layers of reactive film and lightguide are stacked in a sandwich configuration for improved moderation and efficiency. The ends of the lightguide strips are interfaced to the PMTs using a fishtail lightguide. This detector concept relies on a good longitudinal light transport to the PMT's photocathodes. The double-sided readout of charged particles escaping the $^6$Li reactive layer doubles the intrinsic efficiency per layer while preserving a good pulse height distribution. Gamma discrimination is improved by the low energy deposition in the thin scintillator film. The very close refraction indexes (1.58 for PVD scintillator and 1.502 for PMMA lightguide) allow for low light losses during light transfer between the scintillator sheets and lightguide strip.

**Fig. 3** Lithium foil sandwich detector concept. The $^6$Li sheets are shown in red, the PVD scintillator film is shown in blue and PMMA lightguide strips and fishtail lightguide are show in yellow.
**Light transport measurements.** The light transport from the place of light generation in scintillation film to PMMA lightguide strips and the longitudinal light transport to the ends of lightguide strips and PMTs photocathode are critical elements of detector design. We constructed an experimental detector consisting of 20" long by 2" wide and 0.25" thick strips of PMMA lightguide laminated with 200-μm plastic scintillator film coupled to Hamamatsu PMTs (model R6232). This detector was tested with $^{239}$Pu alpha source (the alpha particles with 5.15-MeV energy have a similar light yield as 2.7 MeV tritons) for light transport properties. The accumulated spectrum taken different positions of the $^{239}$Pu source in presence of constant exposure to gamma radiation from $^{137}$Cs source is shown in Fig. 4.

The experiment shows good pulse height distribution from alpha particles with equivalent light yield as 2.7 MeV tritons. There is also a gap between the gamma source response and particles distribution, similar to that of $^3$He detectors. In final application, we will use about 30 μm thick
scintillation films (triton range in plastic scintillator is ~60-μm, calculated by SRIM code [17]).
These thin scintillator films will significantly reduce the sensitivity to gamma radiation.

**Pulse height spectrum and intrinsic efficiency.** In order to evaluate the performance of the sandwich detector, a set of Monte Carlo simulations have been performed using the MCNPX code; this code allows one to follow the alpha and triton particles subsequent to \(^{6}\text{Li}(n,\alpha)^{3}\text{H}\) nuclear reaction. For these simulations we have assumed lithium enriched at 95% in \(^{6}\text{Li}\) with density of 0.464 g/cm\(^3\). Figure 5 shows the results obtained from these calculations; the neutron reaction rate into the lithium film and the intrinsic efficiency for various event thresholds. It is worth mentioning the high tolerance of detection efficiency to variations of event threshold: for example at 40 μm thickness, increasing of event threshold from 1-MeV to 2-MeV leads to reduce of intrinsic efficiency from ~29% to ~23%. Beside the tolerance to instability of detector and electronics, this feature allows operation at very high gamma fields without significant loss of efficiency.

![Graph showing neutron reaction rate and intrinsic efficiency results](image)

**Fig.5 Neutron reaction rate and intrinsic efficiency results as obtained by MCNPX modeling.**

Even if both the alpha and triton particles deposit energy in the scintillator film, taking a conservative approach we ignore the alpha’s contribution to the light pulse; and use the triton’s energy deposition as an estimate for light pulse height distribution. Figure 6 shows the calculated pulse height spectrum for tritons. The gamma sensitivity is naturally limited by the 30 to 50 μm thickness of the plastic scintillator film.
Fig. 6 Triton pulse height spectrum as function of lithium foil scintillator film. The intrinsic
efficiency of the detector is also reported.

**Efficiency modeling of neutron detector assembly:** The $^6$Li foil sandwich detector features
described in the previous paragraphs (high efficiency and a well defined gap in the distribution
of gamma rays and charged particles) are promising in order to develop a new
coincidence/multiplicity detector for use in the field of the nuclear safeguards. To this aim, we
modeled an equivalent of the HLNCC-II [18], a typical neutron coincidence counter, by
substituting $^6$Li sandwich detectors for the $^3$He proportional counters. The detector system of the
HLNCC-II is composed of a cylindrical polyethylene body with 18 $^3$He proportional counters
(see Fig. 4 for a cross section view); a cadmium liner is present inside the sample cavity and
outside the coincidence detector. An equivalent detector was modeled in MCNPX as a polygon
of 10 sides with 12 layers $^6$Li (thickness 50 µm) and 13 light guides strips in each side. Again,
the cadmium liner was included in the simulations. The overall dimensions are the exactly same
of the HLNCC-II. The results obtained are reported in the Table 1. The efficiency calculated for
the HLNCC-II equivalent with $^6$Li is ~2 times the efficiency of the standard HLNCC-II, and die-
away time is ~1/3 of that seen with the standard HLNCC-II coincidence counter. Monte Carlo
simulations are in progress to optimize the arrangement of the HLCNN-II equivalent coincidence
counters.
CONCLUSIONS.

This paper presents the experimental and numerical results with $^6$Li foil neutron detector. In the first part of the paper we have described a $^6$Li lined ionization chamber previously built at Los Alamos National Laboratory. The results obtained show both very good neutron efficiency and demonstrated the feasibility of building a neutron detector based on $^6$Li foils. Because of the difficulties in using Physical Vapor Deposition (PVD) to deposit $^6$Li on the 3-dimensional surface of the detector and in order to design a more flexible neutron detector, we have developed a thermal neutron detector based on $^6$Li foil sandwiched between lightguide strips laminated with thin film of plastic scintillator. The double-sided readout of the lithium foil provides intrinsic efficiency per layer of $\sim 20-30\%$ depending of event threshold setting. The
pulse height spectrum measured with $^{239}$Pu and $^{137}$Cs sources shows good separation between gamma ray and charged particles responses and inherent insensitivity to instability of detector and changes in electronics gain. The MCNPX comparison model of the neutron coincidence counter based on $^6$Li shows higher efficiency than the standard HNLCC-II using $^3$He proportional counters, up to three times shorter die-away time and negligible dead time. The combination of these properties with the inherent insensitivity to detector and electronics instability makes this detector a good alternative of $^3$He technology.

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