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Thermal neutron detection using alkali halide scintillators with ^{6}Li and pulse shape discrimination

Erik Brubaker, Dean Dibble, and Pin Yang

***Abstract*—An ideal ^{3}He detector replacement for the near- to medium-term future will use materials that are easy to produce and well understood, while maintaining thermal neutron detection efficiency and gamma rejection close to the ^{3}He standard. Toward this end, we are investigating the use of standard alkali halide scintillators interfaced with ^{6}Li and read out with photomultiplier tubes (PMTs). Thermal neutrons are captured on ^{6}Li with high efficiency, emitting high-energy α and triton (^{3}H) reaction products. These particles deposit energy in the scintillator, providing a thermal neutron signal; discrimination against gamma interactions is possible via pulse shape discrimination (PSD), since heavy particles produce faster pulses in inorganic scintillating crystals. We constructed and tested two classes of detectors based on this concept. In one case ^{6}Li is used as a dopant in polycrystalline NaI; in the other case a thin Li foil is used as a conversion layer. We present results from these investigations, including measurements of the neutron efficiency and gamma rejection for the two detector types.**

I. Introduction

THERE are many ideas for thermal neutron detectors to replace ^{3}He, as necessitated by the global shortage of ^{3}He. Most if not all consist of one of the handful of other nuclei with high thermal neutron capture cross-sections, along with a medium sensitive to the products of that capture reaction. In the case that the capture agent is not the same as or part of the sensitive medium, the interface between them must be considered, and finally, for many applications a low sensitivity to gamma radiation must be maintained. For e.g. cargo screening and other homeland security applications, a level of 10^{-6}–10^{-7} is desired to successfully identify a neutron signal in the presence of the relatively higher expected gamma background flux.

Our proposed detectors use ^{6}Li as the capture agent, alkali halide crystalline scintillators as the sensitive medium, and PSD for gamma rejection; two ways of interfacing ^{6}Li with NaI or CsI are investigated. PSD in alkali halides is not commonly used in radiation detection, but has been studied [1], [2], [3].

II. Lithium as a dopant

First, we have considered ^{6}Li as a dopant in NaI. Since lithium is also an alkali metal it is possible that Li could replace Na in the crystalline structure. We have successfully formed a solid solution with $\approx 5\%$ molar concentration of lithium, and produced polycrystalline NaI(Li,Tl) samples via hot pressing. The samples in their airtight housing are shown in Fig. 1. There is much room for refinement in the process, specifically to improve the transparency of the samples.

Fig. 1. Polycrystalline NaI(Li,Tl) samples tested for thermal neutron sensitivity. Diverse color and transparency can be seen in these samples, indicating the sensitivity of the material to variations in the production process and dopant concentration.

Of course, LiI is itself a commonly used scintillator with thermal neutron sensitivity [4]. Its primary disadvantages are low light yield, long scintillation decay time, and the absence of PSD to improve gamma rejection. The goal of this work is to create a material with primarily NaI-like scintillation properties, but containing some Li for thermal neutron sensitivity.

III. Lithium as a conversion layer

Second, we have interfaced a thin layer ($\approx 50\ \mu$m) of ^{6}Li foil with NaI and CsI crystals, as seen in Fig. 2. Initially, a single scintillating crystal was used, such that only one of the back-to-back reaction products can escape the foil into the crystal. A sandwich detector is also envisioned, where a coincident high-dE/dx event in two crystals would provide a highly specific thermal neutron signal. Most results shown here are from an assembly with a CsI(Tl) crystal, which was chosen partly for the simpler handling (lower hygroscopicity than NaI) and partly for its excellent PSD properties. Due to the reactivity of lithium, these detectors must be assembled in a controlled environment such as a dry box, and must be well sealed against air and moisture. Once a suitable container was developed, the assemblies were observed to be stable over months, at least by visual inspection.

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Brubaker and Dibble are with Sandia National Laboratories, Livermore, CA.

Yang is with Sandia National Laboratories, Albuquerque, NM.

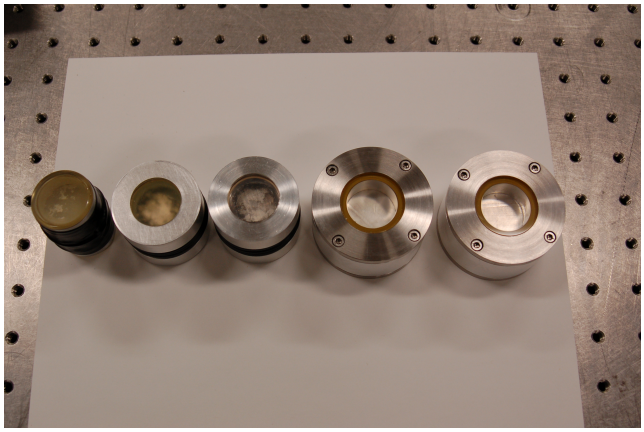

Fig. 2. Five tested detectors using Li foil interfaced with an alkali halide scintillator. Good hermetic sealing was needed to achieve stable detectors with no degradation of the Li foil.

Fig. 3. A cave was constructed of HDPE and paraffin to moderate the fission spectrum neutrons emitted from ^{252}Cf and increase the flux of thermal neutrons through the detector.

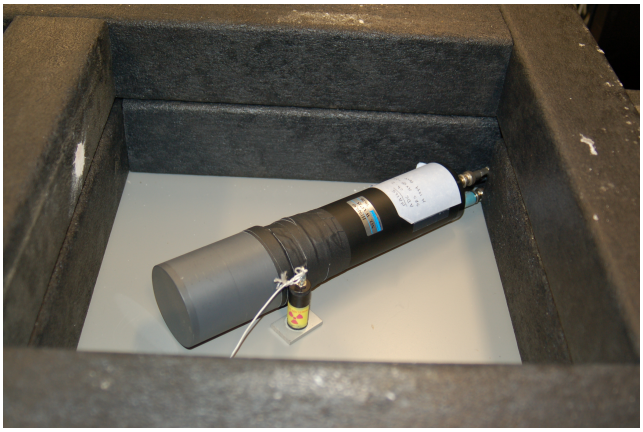

Fig. 4. The ^{252}Cf source (cylinder with string attached) and the detector under test were placed in the center of the cave on an aluminum plate.

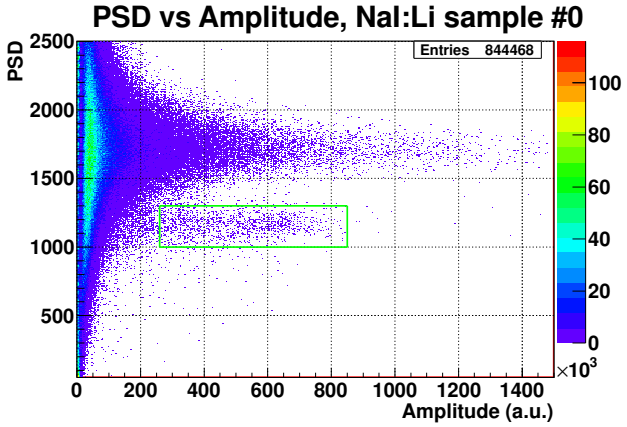

Fig. 5. Pulse shape parameter vs pulse amplitude for polycrystalline Li-doped NaI in the presence of thermal neutrons. The band at 1700 in the pulse shape parameter corresponds to gamma events, while the band at 1200 corresponds to the triton and alpha recoils from thermal neutron capture on ^{6}Li embedded in the material.

IV. Measurements

For these feasibility studies we used natural lithium salts and foil, with about 7% ^{6}Li abundance. Each sample or detector assembly was wrapped in a diffuse reflector and hermetically sealed in a housing with plastic window. The unit to be tested was coupled with optical grease to a PMT. Pulse shapes were acquired with a 500 MS/s digitizer and written to disk for offline analysis.

Our neutron source was a sealed ^{252}Cf source. In order to increase the flux of thermal neutrons, we placed both source and detector inside a cave constructed of HDPE and paraffin to provide moderation of the neutrons (Fig. 3 and Fig. 4). Tests with a ^{3}He detector of known sensitivity indicated a thermal neutron flux of 12.2 ± 1.2 (syst.) $\mathrm{n/cm^2/s}$, and this number was used for efficiency measurements below. The systematic uncertainty is estimated by considering variations in the measured flux with time and position of the source and detector within the cave.

Offline analysis of the digitized pulses proceeded as follows. First a baseline value was determined for each pulse using about 200 pre-trigger samples. If the RMS of the pre-trigger sample values was large, the baseline was considered unreliable and the pulse rejected. This step significantly improved the number of gamma pulses misreconstructed in the signal region of low PSD values. Pulse amplitude was calculated as the integral of the acquired pulse over baseline, while the pulse shape parameter corresponds to the number of samples (2 ns per sample) required to integrate from 10% to 80% of the area under the pulse. (Or 50% to 90% in the case of the NaI(Li,Tl) doped sample, which was optimized independently.)

V. Results

Results from a polycrystalline Li-doped NaI sample are shown in Fig. 5, and for Li foil adjoined to CsI(Tl) in Fig. 6. The scatter plots show a pulse shape parameter vs. the pulse amplitude. For each detector, we show results from a run with a ^{252}Cf fission-spectrum neutron source in the moderating HDPE/paraffin cave.

A. Efficiency

A clear signal, identified as an additional band in the scatter plot at lower PSD values, is observed in each case in the presence of neutrons relative to the gamma-dominated background

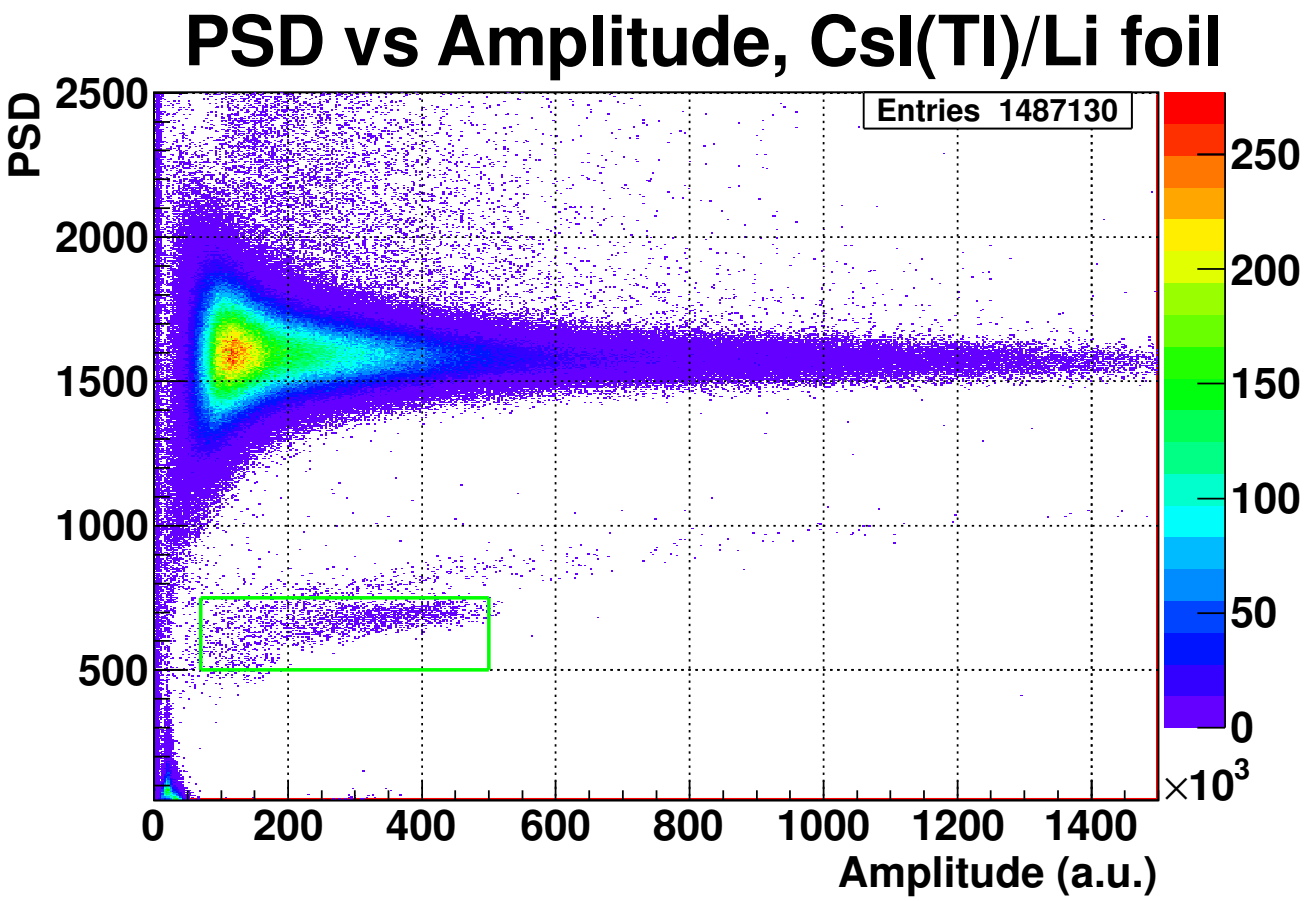

Fig. 6. Pulse shape parameter vs pulse amplitude for a CsI(Tl) crystal adjoined to a thin Li foil in the presence of thermal neutrons. The band at 1500 in the pulse shape parameter corresponds to gamma events, while the band at 500–650 corresponds to the triton or alpha recoils from thermal neutron capture on ^{6}Li in the foil.

band. The thermal neutron detection efficiency was measured by determining a signal region in the amplitude-PSD plot, and comparing the rate of events in that region to the total thermal neutron flux through the surface of the polycrystalline sample or foil, as determined by the independent measurement with a ^{3}He detector. The signal regions were determined by eye, and are represented by green boxes in Fig. 5 and Fig. 6)

Results are summarized in Table I. The measured efficiency corresponds to natural lithium foil or dopant, but using lithium 100% enriched in ^{6}Li would immediately increase the efficiency. An extrapolation to enriched materials is given in the table as well. Relative uncertainties of $\pm 20\%$ (measured) and $\pm 30\%$ (extrapolated) are dominated by systematic uncertainties in the thermal neutron flux (10%), a correction for the bulk crystal sensitivity to fast neutrons ($\approx 10\%$), and in the extrapolation itself ($\approx 15\%$).

B. Gamma rejection

For the Li-doped NaI, the observed pulse shape separation is not sufficient for good gamma-neutron discrimination, though improvements may be possible with changes in the material preparation. For Li foil coupled to CsI the PSD separation is excellent, so a quantitative estimate of the gamma rejection was made.

The CsI(Tl) crystal with Li foil conversion layer was tested in the presence of a strong ^{232}Th source to determine the gamma rejection capabilities. Figure 7 shows the PSD vs amplitude plot from that run. Using the ratio of the number of events falling in the thermal neutron signal region to the total number of events detected, we calculate an upper limit on the gamma sensitivity of this detector of 2×10^{-6}. Since there is a real thermal neutron background contributing to the detected signal events, and since not all gammas passing through the detector are registered and therefore included in the denominator, the real gamma sensitivity is likely close to 1×10^{-7} for the ^{232}Th spectrum.

VI. Conclusion

We have investigated two substantially different approaches to interfacing ^{6}Li with alkali halide scintillating crystals. In summary, alkali halide scintillators with ^{6}Li conversion layers show near-term promise as thermal neutron detectors for ^{3}He replacement. We demonstrated a thermal neutron detection efficiency of $\sim 0.8\%$ with natural Li foil and CsI(Tl), and extrapolate a single-foil efficiency of $\sim 10\%$, close to the theoretical maximum limited by the range of the reaction products in the Li foil. CsI(Tl) crystals also show good gamma rejection via pulse shape discrimination, on the order of 10^{-6}

TABLE I
SUMMARY OF THE MEASURED EFFICIENCY AND GAMMA REJECTION.

Sample	Active Area	Thermal n efficiency Measured (nat. Li)	Extrapolated (100% ^{6}Li)	γ rejection
NaI(Tl) + Li foil	5 cm^2	0.7%	8.4%	??
CsI(Tl) + Li foil	5 cm^2	0.8%	10%	$2 \times 10^{-6} - 1 \times 10^{-7}$
NaI(Tl):LiI(5%)	1.3 cm^2	3.4%	37%	Insufficient
Uncertainty		$\pm$20% (rel.)	$\pm$30% (rel.)	

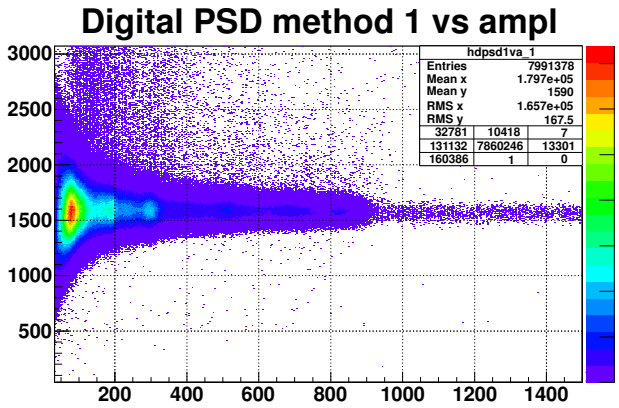

Fig. 7. Pulse shape parameter vs pulse amplitude for a CsI(Tl) crystal adjoined to a thin Li foil in the presence of a strong gamma flux from ^{232}Th. Events falling within the signal region outlined in Fig. 6 are used to measure an upper limit on the misidentification rate of gammas as thermal neutrons.

to 10^{-7}. NaI(Tl) crystals exhibit less powerful PSD, though improved gamma rejection could be achieved with thinner active crystal volumes. Alkali halide scintillators doped with ^{6}Li are an interesting system with potential, but need more investigation. These are new materials that require careful characterization, optimization of doping levels, and refinement of the production process. Both detector concepts have the potential for simultaneous gamma spectroscopy and thermal neutron detection with high efficiency, good gamma energy resolution, and gamma/neutron discrimination.

ACKNOWLEDGMENT

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