LiF/ZnS Neutron Multiplicity Counter

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Abstract:

The availability of ^3He in recent years is becoming restricted with an order of magnitude price increase for this material. Alternatives to the use of ^3He for the detection of thermal neutrons are under investigation. One of the most challenging applications for ^3He alternatives is in neutron multiplicity counters that provide rapid assay of samples which contain an unknown amount of plutonium in a potentially unknown configuration. With appropriate detector design that has minimal gamma-ray sensitivity and a high detection efficiency even for triple coincidence events, the neutron single, double, and triple coincidence events can be used to extract information of three unknown parameters such as the ^{240}Pu -effective mass, the sample self-multiplication, and the (α, n) rate. This project is aimed at determining if commercially available ^3He alternatives can satisfy this challenging application. Using MCNP modeling the best alternative is based upon LiF/ZnS neutron-scintillator sheets and wavelength shifting plastic for light pipes. A four-panel demonstrator module has been constructed, tested, and compared with detailed modeling results. However, to attain that high-level of performance two primary design challenges must be address. They include building a fast electronics system and robust neutron/gamma-ray discrimination based on pulse shape analysis at high rates. A review of the current effort and the most recent findings will be presented.

Keywords: LiF/ZnS; neutron multiplicity counter; safeguards; He-3 alternatives

1 Introduction

The availability of ^3He in recent years is becoming restricted with an order of magnitude price increase for this material. This project grows from an earlier investigation that showed that for several applications there were viable alternatives to ^3He proportional counters. As a result, there is interest in investigating alternatives to the use of ^3He for the detection of thermal neutrons [1, 2]. One of the most challenging applications for ^3He alternatives is in neutron multiplicity counters (NMC). Neutron multiplicity counters are used to provide rapid, non-destructive assay of samples which contain a potentially unknown amount of plutonium in an unknown configuration. With appropriate detector design, the neutron single, double, and triple coincidence events can be used to extract information of up to three sample parameters. Neutron multiplicity counter designs have been refined over decades to provide the most amount of information in the least amount of time. However, most of that refinement work has relied upon ^3He tubes since the system must be insensitive to gamma-rays.

Since the late 1970's, ^3He -filled proportional tubes have been the mainstay technology upon which the capabilities of neutron coincidence and multiplicity counters have evolved. The epitome of that evolution is the ^3He -based epithermal neutron multiplicity counter (ENMC) which consists of 121 10-atm ^3He tubes embedded in high density polyethylene (HDPE); additional details of the ENMC design can be found in reference [3]. It has been extensively characterized and is capable of performing mass assays with 1% uncertainties in 15 to 60 minutes depending on size/activity of the sample. In addition, and

typical of ^3He proportional tubes, the ENMC is extremely stable over time and has a minimal sensitivity to gamma rays.

This paper describes the development of a neutron multiplicity counter design which uses an alternative to ^3He -filled tubes for neutron detection. Several alternatives were examined (BF_3 , boron-lined tubes) in a previous study [4] and the best performing alternative for neutron multiplicity applications identified by the study is lithium fluoride/zinc sulfide (LiF/ZnS) scintillation sheets. As a part of the effort to identify a ^3He alternative multiplicity counter, a four-panel demonstrator module was constructed, tested, and compared with modeling results. The findings indicate it may be possible to construct a full-scale neutron multiplicity counter with the same overall size as the ^3He -based ENMC and with improved performance. The remaining challenges are to demonstrate conclusively scaling up of the demonstration module would indeed approach the performance of the ENMC, determine the appropriate high speed electronics, and perform robust neutron/gamma-ray discrimination based on pulse shape analysis at high rates.

2 Study of ^3He alternatives

A study was performed previously to investigate the feasibility of various ^3He alternatives for use in neutron multiplicity counters [4]. Three thermal neutron detection alternatives were investigated: boron-trifluoride (BF_3) gas-filled tubes, boron-lined tubes, and layers of lithium fluoride/zinc sulfide scintillation sheets. A common figure of merit (FOM) was chosen to allow a comparison of the performance of different systems based on the three alternatives. This FOM is evaluated from the neutron detection efficiency, ϵ , and the neutron die-away time (the characteristic time a neutron is in the system prior to capture), τ as: $FOM = \frac{\epsilon^2}{\tau}$. In units of percentage squared per microsecond, the ENMC value of ~ 190 was the target FOM against which the three alternative configurations were measured. Given the definition of this FOM, it is clear that for alternative systems to succeed, configurations had to be designed that maximized the counting efficiency while minimizing the die-away time. In addition to attaining the FOM target, a viable replacement of the ENMC should also have a footprint similar to the ENMC's outside dimensions, which is approximately $65 \times 65 \text{ cm} \times 100 \text{ cm}$ and has an active region that is 71-cm tall.

The results for the highest performing notional designs identified by this study for each of the alternatives are presented in . The study with BF_3 examined several configurations and concluded with an optimized design consisting of a collection of 155, 5.08-cm diameter, 1 atmosphere tubes with a footprint about 50% wider than the existing ENMC. While the efficiency is similar to the ENMC, the larger amount of moderator increased the die-away time and lowered the overall FOM. For the boron-lined tubes, an optimization study was performed to determine the appropriate number of tubes along with the thickness of the boron coating on the tube walls. The optimal thickness was 1 micron and 4725 tubes each of 0.4 cm diameter were used. The detector footprint was approximately 13% wider than the ENMC. However, as shown in Table 1, the best boron-lined design had a lower efficiency and higher die-away time than the ENMC leading to a considerably lower FOM. The LiF/ZnS design used LiF/ZnS neutron-scintillator sheets and wavelength shifting plastic (WLSP) for light pipes and had the same width as the ENMC. Unlike the others two systems, the predicted neutron detection efficiency is similar to the

ENMC but with a reduced die-away time. The FOM is approximately double that of the ENMC. Given the results of the study, the decision was made to pursue the LiF/ZnS design.

It should be noted that the total capture efficiency in the LiF/ZnS sheets were tallied in the MCNP simulation and then reduced by a phenomenological Validation Correction Factor (VCF) obtained by comparing the predicted capture efficiency to measured laboratory results. Before construction and testing of the small-scale demonstration system described below, a few prototype LiF/ZnS based portal monitor systems were modeled and gave VCF values of ~ 0.5 - 0.6 . Comparison of more precise modeling of the demonstrator module discussed here gives a VCF of ~ 0.84 , and that is the value used to convert simulated total-capture efficiency for the LiF/ZnS studies into expected total count efficiencies and their FOM's.

Table 1. Summary of performance of notional designs using ^3He alternatives.

System	ENMC	BF ₃	Boron-lined	LiF/ZnS
ϵ (%)	66	57	39	63
τ (μs)	23	44	37	10
FOM	189	74	41	391

3 Demonstrator System

The results of the modeling study indicated that LiF/ZnS had the highest performance among the investigated alternatives, thus a small-scale demonstrator system was constructed. Detector materials in addition to the LiF/ZnS were also tested to ensure that they would perform as needed.

Experiments were performed with panels constructed of LiF/ZnS sheets layered with either non-scintillating polymethyl methacrylate (PMMA) plastic or wavelength shifting plastic light pipes. Photomultiplier tubes (PMTs) manufactured by Hamamatsu (R7724B) were attached to both ends of the panels. Superior light collection was obtained using the WLSP but with the cost of increased gamma-ray sensitivity. It is supposed that light propagation is more advantageous in the WLSP due to the isotropic re-emission of the ZnS scintillation light. The light entering the PMMA is not at optimal angles for propagation down the plastic light pipe leading to greater loss of light. Modeling is underway to confirm this hypothesis but the superior performance of WLSP was observed in laboratory measurements.

The demonstrator system is shown in Figure 1 and Figure 2. Five thin sheets of 0.5 mm thick LiF/ZnS sheets manufactured by Eljen (EJ-426HD2) were placed between six pieces of 7 mm thick WLSP also manufactured by Eljen (EJ-280). Tapered light guides were attached to each end and photomultiplier tubes were attached to the light guides. Each stack of LiF/ZnS and WLSP was then placed in a light tight aluminum outer housing. The four panels were arranged to form a box around a sample region and testing was performed.

The LiF/ZnS is used in large sheets to increase the surface area and neutron detection probability. One challenge in using LiF/ZnS as opposed to ^3He is gamma-ray sensitivity. ^3He tubes with the appropriate electronics threshold are extremely insensitive to gamma rays. However, with LiF/ZnS and WLSP, gamma rays can interact with either material and generate light which is then detected by the PMTs. Fortunately, there is a difference in the shape of the light pulse that is generated by gamma-ray induced events and neutron induced events. Neutron-induced events tend to have pulses with sharp rises but tails that can continue for several microseconds. Gamma-ray-induced events tend to be less bright and have pulses with sharp rises and short tails. For the testing that has been performed to-date, an analysis which integrates the charge in the tail and compares it to the total charge using the ratio of two time windows has been very successful at separating the neutron and gamma-ray events. A further reduction in gamma-ray sensitivity has been achieved by requiring coincidence between the two PMTs. More details on neutron and gamma-ray event selection will be presented in later sections.

4 Model Validation

The simulated efficiencies were determined with MCNP [5] and report the total neutron captures. The actual light transport was not performed in the MCNP model. In the LiF/ZnS system, energy or light losses could occur in the various detector materials which will reduce the efficiency. The experiment plan proceeded by comparing modeling results with laboratory measurements of the efficiency of the system as well as the die-away time. More details are provided in Ref. [6] but a short summary follows.

A bare ^{252}Cf source was used as the source for the die-away and efficiency measurements. The die-away measurement was performed using analog electronics which required a coincidence between the top and bottom PMTs in a slab. The analog signals from each PMT were amplified using an Ortec Fast Timing Filter Amplifier and then discriminated with an Ortec Constant Fraction Discriminator. The logic outputs from each pair of discriminators (corresponding to the two PMTs from a single slab) were sent to an Ortec Coincidence Module. The output of that module was then ORed together and sent to a Canberra JSR14 shift register to determine the die-away time via the gate-width variation technique. Gate widths were varied from 2 microseconds to 64 microseconds with a constant pre-delay of 2 microseconds and an accidental gate of 4.5 milliseconds. The doubles rates as a function of gate width were fit with an exponential function yielding a measured die-away time of 12.4 microseconds. This agrees well with the simulated die-away time of 11.8 microseconds. No neutron/gamma-ray discrimination was performed in this setup (other than the coincidence requirement which disfavors gamma-ray events) and so some gamma-ray events were included in the die-away time measurement. However, the size of the effect was investigated in an earlier set of measurements by surrounding the ^{252}Cf source with a layer of lead that reduced the gamma-ray flux by approximately 9 times. The die-away time decreased by approximately 7% indicating that the vast majority of events being analyzed by the JSR14 are due to neutrons. The die-away time will be measured again in the near future with a system capable of discriminating neutron events from gamma-ray events.

The neutron detection efficiency measurements were performed using Pixie-4 digitizers from XIA¹ and the same bare ²⁵²Cf source as was used for the die-away time measurement. In addition to reporting a pulse height and time for each event, several microseconds of waveform data were recorded for each event. This allowed a simple two window pulse shape discrimination to be applied which reduced sensitivity to gamma rays. The efficiency was calculated given the known source strength and then compared with the total number of detected neutron events. These results were found to be 84±13% of the simulated neutron efficiency thus giving a validation correction factor which can be used to scale other modeling results. The uncertainty is driven by the uncertainty in the activity of the Cf-252 source. Measurements with a source with a smaller uncertainty are planned for the near future. The difference between the measured and modeled results is hypothesized to be due to light losses since those effects were not modeled in the simulation. The measurement provides a scaling factor for the simulated results assuming the final geometry of each slab is not significantly different from the demonstrator slabs.

Figure 1. Demonstrator system of four paddles (three are visible) ready for insertion of the source for measurements.

¹ XIA LLC, Hayward CA

Figure 2. Left: Exposed WLSF with tapered light guide attached. Right: Fully assembled panel with a PMT on either end.

5 Full-scale designs

The 84% validation correction factor was used to scale the simulated efficiencies of the full-scale LiF/ZnS-based designs. Many designs were examined; the most promising designs are shown in Figure 3 and their properties are summarized in Table 2. The designs were constrained to have the same horizontal size as the existing ENMC system. The highest efficiency alternative design is the Lithium-based Neutron Multiplicity Counter 8 (LiNMC-8) so-called due to the eight panels surrounding the central sample chamber. Each panel consists of twenty layers of LiF/ZnS sheets between 21 layers of 7 mm thick WSP. For actual construction, it is assumed that each panel will be separated into 4 optically isolated parts with 5 layers of LiF/ZnS each. This will act to limit the overall event rate on any given channel. The corner pieces are smaller than the side pieces but increase the efficiency as demonstrated by contrasting the Table 2 results with the LiNMC-4. LiNMC-4 had the corner pieces removed thus reducing the efficiency but without significant effect on the die-away time. However, there are more criteria for selecting a final full-scale design than the FOM. In addition to a large FOM, the final design must be minimally sensitive to the location and configuration of the sample and to the neutron energy distribution emitted by the sample. Another consideration is the cost of instrumenting and reading output channels. The LiNMC-4 will have a lower cost than the LiNMC-8 but at the expense of sensitivity to the source placement due to the lack of axial symmetry. The LiNMC-4+ was then chosen as a compromise between the two systems. The addition of four wedge-shaped high density polyethylene blocks in the corners improves the efficiency with the cost of an increased die-away time. However, it improves the symmetry of the azimuthal detector response and maintains the smaller number of instrumented channels. As Table 2 shows, the LiNMC-4+ design is expected to perform similarly to the existing ENMC.

A constraint in developing the alternative multiplicity counter is the overall height of the system. Recall that the ^3He -based system that is serving as a model of performance is the ENMC and that system has an active region that is 71-cm tall with a total height of approximately 1-m. This compact size allows easy placement of sample containers into the counter. The demonstrator system has the same active height as the ENMC but the addition of the PMTs as opposed to the ^3He tube bases makes the current

system approximately 60-cm taller than the ENMC. The height of the active region of the ENMC was based on potential sample sizes and a desire to maintain a constant detection efficiency along the height of the sample volume. Therefore, the only place to reduce the height of the LiF/ZnS-based counter is to reduce the size of the PMTs. Silicon photomultipliers (SiPMs) may solve this problem by being compact and having compact readout electronics. However, SiPMs can have noise which may affect the low level threshold and the efficiency. The current plan is to continue to use PMTs but design the system with a side access port either through a sliding or swinging door mechanism.

Figure 3. The three most promising full-scale designs viewed from the top.

Table 2. Summary of modeled performance for promising full-scale designs.

System	ENMC	LiNMC-8	LiNMC-4	LiNMC-4+
ϵ (%)	66	63	44	55
τ (μ s)	23	10	10	15
FOM	189	391	185	203

6 High speed readout

The previous laboratory measurements with the small-scale demonstrator system used separate data acquisition systems for the efficiency and die-away time measurements. The final system will need to measure both quantities simultaneously while also retaining the ability to separate neutron events from gamma-ray events. One solution being investigated is the AlazarTech² ATS9626 2 channel, 16 bit, 250 MHz waveform digitizer with a field programmable gate array (FPGA) available for user applications. This digitizer has the ability to run in full waveform mode continuously with no dead time. This allows the collection of large amounts of data where no events are lost and that can be analyzed offline to

² Alazar Technologies Inc, Point-Claire, Quebec, Canada

refine various PSD algorithms. Similar high speed data processing is being developed for another project funded by the same US Department of Energy Office (see Acknowledgements) using the same digitizer hardware [7].

An example of the digitized waveforms for two neutron events in the demonstrator module is shown in Figure 4. The waveforms are for the top PMTs and bottom PMTs and show how both PMTs observe the neutron induced reactions. The waveforms appear to consist of multiple short pulses which may require smoothing prior to more advanced analysis. The need for smoothing is currently under study.

Figure 4. Sample waveform data showing two neutron events separated in time by approximately 50 microseconds.

The full waveform mode is very useful for diagnostics and research and development tasks but requires large amounts of disk space for storage and significant computing resources to analyze the data after collection. However, for the final system the goal is to move as much of the analysis to the FPGA located on the ATS9626.

As a first test of the functionality of the FPGA, a set of multiple charge integration windows has been implemented. These windows are user configurable and integrate the waveform for certain preset amounts of time and then report the total charge observed in that window. This is a much smaller amount of information that must be recorded for each event but still allows a simple pulse shape discrimination (PSD) by comparing the charge collected in short gates and long gates. In this analysis, all the charge integration windows begin at the same time but extend for different amounts of time. The charge in a short gate is designated SG and the charge in a long gate is designated LG . Gamma ray events with their much shorter duration are expected to have similar amounts of charge in both the short and the long gate. However, neutron events will have more charge in the longer gates due to the longer tail. The pulse shape discrimination value is then typically determined using $(LG-SG)/LG$ which can also be thought of as the tail charge over the total charge. Gamma-ray-like events will cluster near 0 and neutron-like events will have larger values tending towards 1. The length of the charge integration

windows must be optimized for the best performance and studies are currently under way. However, initial results indicate that the system can perform the PSD while also preserving the timing information for each neutron event. The performance of the FPGA as a function of rate has not been tested yet.

7 Pulse shape discrimination

The simple charge window technique for suppressing the gamma-ray response described in the previous section is expected to fail at high rates especially if many gamma-rays are present in the sample. Multiple gamma-rays could arrive with the appropriate timing and mimic a neutron-induced event when simply recording the total charge in multiple gates. An effort is underway to perform a more sophisticated analysis of the pulse shapes to allow improved gamma-ray rejection at high rates.

One method under study is a template matching algorithm. Waveform data were collected for gamma-ray and neutron sources separately using the small-scale demonstrator system. Typical neutron and gamma-ray waveforms were then selected and summed over multiple events to generate templates with minimal statistical variation. These templates were then used in a minimization routine to determine the most likely combination of overlapping events that represent the data. More details about template matching minimization algorithm are available in Ref. [8].

Studies of the algorithm were performed with synthetic data created using the same neutron and gamma-ray event templates. The relative amounts and rates of neutron and gamma-ray events were varied and the algorithm was applied to all the different cases. Figure 5 summarizes the results of the pileup study over a rate range from 100 kHz to 1 MHz. It demonstrates that this algorithm can reliably separate neutrons from gamma rays in the 100 kHz range even when the gamma rays outnumber the neutrons 3 to 1. The current template matching algorithm is too complex for high-speed implementation on the FPGA on the ATS9626 but it may be able to be simplified while maintaining this level of performance. For example, a multiple charge window technique may be used to flag when piled up events occur and could allow for a deconvolution of the gamma-ray and neutron signals offline.

Figure 5. Performance summary for template matching algorithm using simulated neutron and gamma-ray data over a range of rates.

8 Conclusion

Based on previous and current studies, a promising new design for a ^3He -free neutron multiplicity counter has been identified. The design minimizes the number of instrumented channels lowering the cost while still achieving a performance similar to the existing ENMC. Validation of the models with a small-scale demonstrator has given confidence in the full-scale designs. High speed digitizer waveform data collected with the demonstrator system have yielded results similar to previous analog measurements. Real time PSD is being implemented and the current solution is expected to scale to the full-size system. The performance of the system at high rates of gamma-ray and neutron events will be tested in the near future but a potential solution has been identified.

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