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Evaluation of Eu:LiCAF for neutron detection utilizing SiPMs and portable electronics

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ABSTRACT

With the increasing cost and decreasing availability of ^{3}He, there have been many efforts to find alternative neutron detection materials. Lithium calcium aluminum fluoride (LiCAF) enriched to 95% ^{6}Li doped with europium was evaluated here as a replacement material for ^{3}He. Wafers 0.5 cm thick, consisting of LiCAF crystals in a rubberized matrix, were embedded with wavelength shifting fibers (WSF) and mated to silicon photo-multipliers (SiPMs) to measure the photon response in a flux of neutrons from a DD neutron generator. Excellent discrimination was realized between neutrons and gammas, and both pulse-height discrimination and pulse-shape analysis were explored. A Figure of Merit (FoM) of 1.03 was achieved. By applying pulse-shape analysis, a simple neutron count output was generated by utilizing a low-pass filter to suppress fast pulses from the SiPM output and subsequently applying a threshold to the remaining signal. Custom electronics were built to bias the SiPMs, then amplify, filter, discriminate, and digitize the LiCAF/WSF scintillation photons, resulting in a digital pulse that can easily be counted with any microcontroller or field programmable gate array. A significant advantage of LiCAF is that it can be fabricated into any shape/size (when embedded in a rubberized matrix), and the light output and transparency is sufficient to allow for thicker scintillators which enable detection of both thermal and epithermal neutrons. This work demonstrated that Eu:LiCAF is capable of discriminating gammas from neutrons and is a potential replacement material for ^{3}He, especially for nuclear security applications and neutron spectroscopy.

1. Introduction

Neutron detection has been investigated for decades, and it has been an enduring goal of the nuclear community to develop accurate and inexpensive neutron detection and spectroscopy techniques. The goal of this work was to focus on a detection medium that can effectively replace ^{3}He based neutron detectors, while also possessing properties that allow it to perform well as a neutron counter in a layered neutron spectrometer setup. ^{3}He has long been the material of choice for detecting neutrons, but high cost and limited supplies have created the impetus to find a replacement material. LiCAF (lithium calcium aluminum fluoride) is the material of choice for this work because of its desirable properties: non-hygroscopic, low γ sensitivity, available in larger sizes, transparent and high light yield [1,2]. ZnS: ^{6}LiF and ZnS: $^{10}B_2O_3$ are other alternatives that have been recently studied, and while ZnS has many desirable properties as a scintillator, there are also significant disadvantages including a long afterglow time (upwards of 100 μs) and opacity to its own light [3]. ^{10}B based neutron detectors have the advantage of a larger neutron interaction cross section, however, the reaction products are lower energy than ^{6}Li and include gammas [4].

LiCAF has been previously evaluated by Viererbl et al. and separately by the Pacific Northwest National Laboratory (PNNL) [1,2]. Viererbl et al. focused on the ability to discriminate signals from neutron and gamma radiation and PNNL evaluated LiCAF for application in a portal monitoring system. Using only pulse-height analysis, Viererbl et al. found that gamma radiation with energies above 1400 keV started to interfere with the neutron peak from a 0.5 cm thick wafer of Eu:LiCAF [2]. The discrimination capability, however, is highly dependent on the size and density of small grains (scintillator crystals) in the rubber, and also the geometry of the detector. PNNL found that the LiCAF neutron detector's sensitivity for a bare and moderated ^{252}Cf source is 1.01 ± 0.09 and 1.54 ± 0.23 cps/ng respectively with large rubberized Eu:LiCAF detectors measuring 100 cm long, 26 cm wide and 3 cm thick [1]. This is approximately 40%–60% of the value suggested as a requirement for portal monitors [5].

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Wafers of rubberized Eu:LiCAF were obtained from Tokuyama Corporation, Japan. First, the wafers were evaluated for their neutron response and the ability to discriminate neutrons from gammas. Wavelength shifting fibers (WSF) from Kuraray (B-3) were embedded in the wafers to convert the scintillation photons to a frequency that is optimized for SensL C-Series blue-sensitive silicon photo-multipliers (SiPMs). SiPMs have been used for many years in applications ranging from medical imaging, 3D ranging and sensing, biophotonics, high energy physics, and now for threat identification [6–9]. The scintillation light of particle detectors has historically been collected with traditional photo-multiplier tubes. However, recent advances have significantly reduced the dark noise of SiPMs. Coupling the low dark noise with the greater transparency and relatively high light output of LiCAF creates ideal conditions to introduce SiPMs to inelastic neutron scattering detectors [10].

Previous work with LiCAF indicates that it has the ability to perform well as a neutron detector. The scintillation properties of LiCAF make it desirable for use with silicon photo-multipliers because of its high light output, excellent transparency, and ability to discriminate neutrons from gammas. Custom electronics were developed as part of this work to readout, amplify, filter, and count the neutron pulses created in LiCAF by a neutron flux. The methodology and results are discussed herein.

2. LiCAF scintillator

Two compositions of LiCAF are available from the Tokuyama Corporation. It is available doped with europium or cerium, has an effective Z of 15 and density of 2.99 g/cm^3 [11]. There are a few primary differences between the dopants that led to Eu:LiCAF being chosen for this work. The light yield of Eu:LiCAF is approximately eight times that of Ce:LiCAF. The decay constant of Ce:LiCAF is 40 ns, while it is >1 μs for Eu:LiCAF. This is a disadvantage of Eu:LiCAF, as the shorter decay constant is desirable, however, since the material was tested in environments with a relatively low neutron flux, the longer decay constant was not a significant issue. The luminescent wavelength of the Eu:LiCAF is 360–390 nm as compared to the Ce:LiCAF at 280–320 nm. The optimal wavelength for the Sensl-C series silicon photomultiplier is the peak sensitivity region (approximately 425 nm) where the photon detection efficiency (PDE) is at 42% with an overvoltage of 5.0 V [12]. The photon detection efficiency is highly dependent on the overvoltage of the SiPMs. Finally, europium has a much larger neutron absorption cross section than cesium does, especially near the thermal energy region [13]. This is a disadvantage of the europium atoms, but it is not a serious issue because of the trace amounts of atoms that are present only in the Eu:LiCAF fibers embedded in the rubberized wafer; the primary interaction with the europium is (n, γ) and the gammas will not significantly interact with the low-Z material.

Eu:LiCAF/rubber (2×10^{21} ^{6}Li/cm^3) was used throughout this work. The neutron absorption percentage of the rubber-matrix LiCAF is approximately 5% higher than ^{3}He at 10 atm for 25 meV neutrons [11]. While the neutron absorption percentage is much larger for the pure LiCAF crystal, since it is largely dependent on the number of ^{6}Li atoms, the cost of the material is an order-of-magnitude higher. The neutron absorption percentage of pure cerium or europium doped LiCAF crystal is ~60% for a 1 mm thick sample (thermal neutrons), whereas it is only ~17% for the Eu:LiCAF/rubber used for this work. Thermal neutrons have a high cross section for absorption in ^{6}Li resulting in the following reaction:

$$ {}^{1}_{0}\mathrm{n} + {}^{6}_{3}\mathrm{Li} \longrightarrow {}^{3}_{1}\mathrm{H}\ (2.73\ \mathrm{MeV}) + {}^{4}_{2}\mathrm{He}\ (2.05\ \mathrm{MeV}). \quad (1) $$

Both the tritium and the alpha particles interact in the LiCAF crystal scintillator, emitting photons that are transported via the WSFs to the SiPMs, where a current is created. The current is then amplified and converted to a voltage signal, then filtered and converted to a digital signal using a comparator. The digital signals can then be counted/recorded using a field-programmable gate array (FPGA) or microcontroller.

A disadvantage of Eu:LiCAF is the relatively low α/β ratio. The difference between the ratio of absorbed energy and the light yield for the gamma radiation and heavy charged particles (HCP) is caused by the quenching dependence on the linear energy transfer (LET). The ratio for Eu:LiCAF is 0.2 [14]. This presents an issue for bulk Eu:LiCAF crystals, as the scintillation light from the high-energy HCPs is approximately equivalent to a 1 MeV gamma. The discrimination problem can be mitigated by controlling the geometry of the crystals since the range of the fast electrons induced by gamma rays is significantly longer than the HCP range. In the case of the rubberized LiCAF, controlling the size of the small LiCAF grains embedded in the rubber matrix is essential, and also the number and spacing of the small grains to optimize discrimination capability while not significantly sacrificing neutron detection efficiency. Using a smaller grain size of LiCAF in the rubber matrix is advantageous for discrimination purposes as it allows the fast electrons induced by gamma rays to easily escape the scintillator grain before depositing their full energy [15]. Another method of controlling the sensitivity to gammas is by reducing the overall LiCAF in the wafers (reducing the number of small grains). A drawback to the lower density of LiCAF is the reduced neutron detection efficiency.

Each wafer of rubberized Eu:LiCAF scintillator used throughout this work is 10 × 10 cm × 0.5 cm thick. There are also 30 WSF fibers embedded in both the X and Y axes through the wafer. The WSFs are desirable for signal readout because of the flexibility of the rubberized LiCAF and the custom geometries, which makes using traditional PMTs difficult to implement. However, the gammas and neutrons tend to interact with the WSFs (as they are very similar to plastic scintillation fibers) [15]. [15] found that the spectrum obtained from the WSFs without the Eu:LiCAF scintillator is almost the same as the one obtained with the Eu:LiCAF scintillator and the WSFs when using a ^{60}Co gamma source. The gamma/WSF signals are an undesirable side-effect of utilizing the wavelength-shifting fibers, however the scintillation pulses of the WSFs are on the order of nanoseconds, and an active low-pass filter will be utilized throughout this work to discriminate the gamma/WSF signals from the neutron pulses (~1 μs).

3. Electronics

Maintaining the portability of the detectors was a primary consideration in designing the electronics for the pulse counting and discrimination. Traditional PMTs were not used because of their size and power requirements; SiPMs offer similar specifications as PMTs without many of the disadvantages [6–9]. The advantage of using SiPMs is that they are extremely small, are insensitive to magnetic fields, operate ideally with relatively low voltage (30 V), and the output signal can be easily amplified and filtered with basic electronics. A disadvantage of SiPMs is that their detection efficiency and gain are highly dependent on temperature.

A schematic of the pulse counter circuit is shown in Fig. 1. The timing and gain of each component was carefully chosen to ensure that proper pulse-shape filtering and amplification can be achieved. The first essential component is the Sensl C-Series SiPM, which has a microcell size of 35 μm and a peak sensitivity of 425 nm. The stage 1 is a simple npn transistor used to buffer the current (unity gain) from the SiPMs and the stage 2 amplifier is an Analog Devices AD8007 (ultralow distortion high-speed amplifier, 650 MHz, 1000 V/μs slew rate) with a gain of +2. Initial testing was conducted with the SensL evaluation board (MicroFC-SMA-300xx-35u) to ensure that the light emitted from the LiCAF would result in a sufficiently high signal-to-noise ratio (SNR) after the inefficiencies from both the WSFs and the photon detection efficiency of the SiPMs (maximum of 42%). Fig. 2 shows the results from comparing the SensL evaluation board to the custom circuit using a BGO crystal and ^{68}Ge gamma source. A BGO scintillator was used for the early electronic testing because it has a well documented light output from the 511 keV annihilation gammas that could be used for comparison in simulations. The original signal from the evaluation board (green trace,