Investigation into the potential of GAGG:Ce as a neutron detector

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Abstract

In this work we investigate the potential use as a thermal neutron detector of cerium-doped gadolinium aluminium gallium garnet (GAGG:Ce) coupled to a silicon photomultiplier (SiPM). The response to thermal neutrons has been measured, with two strong low energy neutron-indicative peaks clearly identifiable below 100 keV and additional γ peaks at higher energies. The neutron-related peaks are produced by a combination of contributions from excited states of the two isotopes ^{156}Gd and ^{158}Gd which can be clearly resolved in our GAGG scintillation detector. In particular, two peaks due to neutron-induced γ -ray emission are observed at approximately 82 keV and 260 keV, with best achieved energy resolutions of $24.1 \pm 0.2\%$ and $22.7 \pm 0.7\%$ respectively. Three different scintillator volumes (0.1 cm^3 , 0.4 cm^3 , and 1 cm^3) were investigated and the respective results for each configuration will be presented.

Our findings show that a GAGG-SiPM based detector can be used as a compact, efficient thermal neutron detector in a low γ -ray contamination environment.

Keywords: Neutron detection, GAGG:Ce, silicon photomultiplier.

1. Introduction

Cerium-doped gadolinium aluminium gallium garnet (GAGG:Ce - $\text{Gd}_3\text{Al}_2\text{Ga}_3\text{O}_{12}$) is a relatively new scintillator showing good promise as the potential detector

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Table 1: Typical values of GAGG:Ce characteristics compared to common alternate PET scintillators[1, 5].

	GAGG:Ce	BGO	LYSO:Ce
Light Yield (photons/MeV)	40-55000	8-10000	~32000
λ_{peak} (nm)	520	480	420
Density (g/cm ³)	6.63	7.13	7.1
Z_{eff}	54	74	66
Energy Resolution (%)	~5.2	~9-15	~7.9

of choice for PET[1, 2] and SPECT[3, 4] applications. GAGG has many characteristics making it highly suitable for use as a γ -ray detector, particularly for
5 a PET system. It is one of the brightest scintillators available today with a light yield of 44,000 photons/MeV which is slightly greater than that of NaI(Tl) (38,000 photons/MeV). It has a density approaching the levels of BGO and LYSO (6.63 g/cm³), and a gadolinium mass fraction of ~50.8% resulting in an
10 effective atomic number of 54.

Furthermore, with an energy resolution of ~5% at 662 keV, equivalent to that of NaI(Tl)[6], γ -ray spectroscopy applications become more feasible, especially when considering that the time constant of the scintillation light is fast enough (68-92 ns[7]) that higher rates can be accommodated before pulse pile up
15 becomes an issue. It has also been reported that GAGG:Ce exhibits some degree of pulse shape discrimination (PSD) [7, 8], making identification of the type of incident radiation possible. However the peak emission wavelength, as shown in Table 1, is higher than most scintillators, so care must be taken when coupling GAGG scintillator to a SiPM to ensure a well-matched optical efficiency.

20 The focus of this paper is to exploit the high neutron-capture cross section of the gadolinium isotopes present in GAGG, combined with the high attenuation coefficient for efficient detection of neutron-induced γ rays. These combined properties mean that GAGG has the potential to offer a very clean signature for

154Gd 21.5 H ε: 100.00%	155Gd 5.32 D ε: 100.00% β: < 0.10%	156Gd 5.35 D ε: 100.00%	157Gd 71 Y ε: 100.00%	158Gd 180 Y ε: 83.40% β: 16.60%	159Gd STABLE 100% β: 100.00%	160Gd 72.3 D β: 100.00%	σ(n,γ) (barns) Unknown
154Gd 240.4 D ε: 100.00%	154Gd STABLE 2.18%	154Gd STABLE 14.80%	156Gd STABLE 20.67%	157Gd STABLE 15.65%	158Gd STABLE 24.84%	159Gd 18.479 H β: 100.00%	154G 155G 156G 157G 158G 159G 160G
2.21E+4	8.59E+1	8.27E+4	1.76E	2.53E+5	2.39E+2		
153Gd 15.317 Y ε: 72.13% β: 27.86%	154Gd STABLE 52.19%	156Gd 8.601 Y β: 99.98% ε: 0.02%	157Gd 4.753 Y β: 100.00%	158Gd 15.19 D β: 100.00%	159Gd 35.18 H β: 100.00%	159Gd 45.9 M β: 100.00%	
1.37E+4	3.12E+2	1.35E+3	3.76E+3	1.00E+2	1.10E+2		
151Gd 90 Y β: 100.00%	152Gd STABLE 26.75%	153Gd 46.284 H β: 100.00%	154Gd STABLE 25.75%	155Gd 22.3 M β: 100.00%	156Gd 9.4 H β: 100.00%	157Gd 8.03 M β: 100.00%	
1.51E+4	2.06E+2	4.20E+2	8.37E				

Figure 1: Natural abundances and thermal neutron capture cross sections for the region around gadolinium (amended from NuDat 2.7[12]).

thermal neutron detection. Previous authors have reported the neutron detection performance of earlier gadolinium-containing scintillators, namely cerium-doped gadolinium orthosilicate (GSO - $\text{Gd}_2\text{SiO}_5:\text{Ce}$)[9] and cerium-doped gadolinium iodide ($\text{GdI}_3:\text{Ce}$)[10]. In particular, the performance of GSO as a thermal neutron detector was measured followed by a report on a prototype thin-GSO neutron detector[11].

The gadolinium isotopes from mass 154 to 158 are stable, although only two isotopes, ^{155}Gd and ^{157}Gd , have significant thermal neutron cross sections. The relevant section of the chart of the nuclides[12] is shown in Figure 1 with the respective abundances and (n,γ) cross section displayed for each isotope. Both these gadolinium isotopes have extremely large thermal neutron capture cross sections and reasonably high natural abundances. ^{155}Gd has a natural abundance of 14.8% and a cross section of 60,740 b and makes a substantial contribution to the total neutron capture of gadolinium. However the majority of the response is from ^{157}Gd which has a natural abundance of 15.7% and a cross section of 253,700 b. Overall, the effective $\sigma(n,\gamma)$ for natural gadolinium is 49,000 b.

The implication of the gadolinium thermal neutron capture cross section becomes apparent when the current status of thermal neutron detection technology is considered. The availability (and cost) of ^3He has been widely reported[13–15]. Alternatively, ^6Li and ^{10}B [16, 17] are both commonly used in detection

Figure 2: The low-lying level structure for the rotational bands of ^{156}Gd and ^{158}Gd , adapted from [9, 22–24]. The γ ray transition energies are given alongside the competing internal conversion electron energy (in brackets).

materials. Gaseous detectors using ^{10}B are restricted to a relatively low pressure, and therefore low efficiency, but require a large detector volume[18, 19]. If ^6Li and ^{10}B are loaded into scintillators the achievable loading fraction is limited by degradation of the scintillation characteristics[20, 21]. In comparison, the thermal neutron capture cross section of gadolinium coupled with its mass fraction in GAGG means that relatively small crystals offer almost 100% thermal neutron capture efficiency.

The reaction products of (n, γ) reactions on ^{155}Gd and ^{157}Gd are ^{156}Gd and ^{158}Gd . The reaction Q-value energies are 8.536(2) and 7.937(2) MeV respectively. (n, γ) reactions produce a large number of high energy γ decays, typically above 1 MeV, which are not of primary interest in this work. The resulting de-excitations also produce transitions through numerous low-lying states, which are much more likely to be stopped within a relatively small detector crystal. The level scheme for the first three excited states in each of the nuclides of interest is given in Figure 2. The first excited states in ^{156}Gd and ^{158}Gd are at 88.97

Figure 3: The 1 cm cube from Epic Crystal Co. and its mounting on the SensL J-series SiPM.

60 and 79.51 keV respectively, which are populated by E2 transitions of 199.22 and 181.95 keV from the 4^+ 2nd excited states. In turn the 2nd excited states are themselves populated by E2 transitions of 296.4 and 277.6 keV from the 6^+ 3rd excited states. These γ transitions are the source of key neutron signatures for gadolinium-containing scintillators. However, it should be noted that the 2^+ 65 first excited states in both isotopes can also decay via internal conversion processes, and this mechanism produces an additional low energy neutron-related peak due to the escape of characteristic gadolinium K-shell X-rays.

2. Experimental Method

2.1. Scintillators

70 Three samples of GAGG:Ce were acquired for this work: a smaller cube measuring 7.6 mm was provided by Advatech U.K. Ltd.; a second slightly larger 1 cm cube, and a thin slice 10 mm x 10 mm x 1 mm were both provided by Epic Crystal Co. Ltd. Both cubes were wrapped in PTFE tape to improve light collection; the 7.6 mm cube was only polished on a single face, whereas 75 the 1 cm cube had all faces polished, whilst the slice was polished on the two larger surfaces.

2.2. SensL SiPM

The SiPM used in this work was a SensL J-series 2x2 array, model number ArrayJ-60035-4P-EVB. Each individual SiPM has an active area of 6.07 x 6.07 mm², which combined for a total package surface area of 155.25 mm², with a total of 22,292 microcells per SiPM. The SiPM was readout by a bespoke breakout board which combined the four individual SiPM outputs into a single signal; the board and its mounting can be seen in Figure 3. Each scintillator was optically coupled to the SiPM with Dow Corning high vacuum silicone grease to ensure maximum light transmission.

2.3. Data Acquisition and Pulse Height Analysis

All γ ray and neutron data were acquired with an Ortec EasyMCA and its accompanying Maestro software. The SiPM bias supply was set to 27.2 V, or an overvoltage of 2.5 V as had previously been established for these devices[25]. The SiPM output signal was fed into a Canberra 2005 preamplifier and then an Ortec 672 spectroscopy amplifier with a shaping time of 2 μ s and a gain of 10x.

The detectors were calibrated using the the primary photopeaks from the γ sources ²⁴¹Am, ⁵⁷Co, ²²Na, and ¹³⁷Cs. Additionally, to improve the energy calibration in the low energy region around the first excited states of ¹⁵⁶Gd and ¹⁵⁸Gd, the 32.3 keV barium X-ray peak from ¹³⁷Cs was also used in the calibration.

The thermal neutrons used in this work were produced from a 18.5 GBq AmBe source located within a moderating water tank measuring 80 cm x 76 cm x 94 cm. The role of the water tank is to provide shielding from, and moderation of, the fast neutrons emitted from the Be(α ,n) reaction. The detector sits on top of the water tank, offset approximately 50 cm from the source, shielded by 35 cm of water. The dose rate immediately outside the water tank was monitored with a Berthold LB6411 Neutron Probe, which recorded a rate of ~ 2 n μ Sv/hr throughout the experiment. In order to accurately determine the neutron sensitivity of our detector a complex shielding arrangement was necessary, shown in Figure 4. A dominant source of potential background is

Figure 4: Sketch of the experimental setup sitting on top of the neutron tank. The AmBe source is located off the bottom of the figure surrounded by a water moderator.

the $^1\text{H}(n,\gamma)^2\text{D}$ reaction[26] from the water which causes a significant γ flux. This is shielded from our detector by 5 cm of lead. In turn, the emission of characteristic lead X-rays from the shielding material must be removed using a
 110 ~ 1 mm layer of copper wrapped around the detector. To avoid scattered γ rays a further layer of lead (~ 3 mm) was placed above the detector and to maximise neutron intensity a polyethene reflector (2.5 cm) was built around the whole assembly. Finally, additional layers of cadmium (a 3 mm thick sheet and a further 1 mm thick scattering “hat”) could be put into place to remove thermal
 115 neutrons for background measurements.

3. Results

3.1. Energy Resolution and Light Yield

Pulse height spectra were collected using the calibration sources: ^{137}Cs , ^{22}Na , and ^{241}Am , for each of the scintillator samples. The standard comparison of the ^{137}Cs photopeak is displayed in Figure 5 for each of the scintillator
 120 samples. Additionally, further data is displayed in Table 2 for the other calibration points. The finish of the non-coupled surfaces was different for each crystal so direct comparisons should be made with care. However, all samples achieve sub-10% energy resolution at 661.7 keV, which is consistent with previously reported work on GAGG coupled to SiPM readout by Seitz et al[1].
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Figure 5: Uncalibrated ^{137}Cs spectra for each of the GAGG:Ce samples. Gaussian fits have been applied to the 661 keV peaks and the respective energy resolutions have been extracted.

Furthermore, the exact composition of commercially available GAGG:Ce varies between suppliers, with dopant levels, growth technique, and elemental ratios contributing to the variability of quoted light yields ranging from 22,000-56,000 pH/MeV[1, 7, 27–29].

Figure 5 demonstrates the variable light yield obtained from our samples. In particular, by comparing the two samples from the same manufacturer, the effect of internal attenuation within the scintillator is clearly observed. The photopeak from the 10 mm thick scintillator occurs at a significantly lower channel number than the 1 mm sample, due to the internal absorption of the scintillation light[1, 30–32].

3.2. AmBe Neutron Measurements

The calibrated AmBe spectra for the 10 mm thick scintillator is shown in Figure 6 with several of the key features marked. The combined 1st excited state to ground state transitions for ^{156}Gd and ^{158}Gd at ~ 82 keV is the most prominent feature and is well separated from a lower energy peak at ~ 43 keV. In addition two higher energy features are also observed at ~ 259 keV and ~ 517 keV.

Similar features were observed in Ref. [9] and an explanation of their origins was initially presented there. The 82 keV peak is the combination of the 88.9 and 79.5 keV γ rays from the first excited states of ^{156}Gd and ^{158}Gd , which

Table 2: γ ray photopeak energies for the calibration sources

Sample	E_{peak} (keV)	E_{res} (%)
1 mm	59.5	21.7 ± 0.2
	122	16.7 ± 0.6
	661	8.64 ± 0.06
7.6 mm	59.5	25.3 ± 0.07
	511	10.3 ± 0.1
	661	9.80 ± 0.05
10 mm	59.5	27.1 ± 0.2
	511	9.2 ± 0.1
	661	7.94 ± 0.06
	1274.5	5.6 ± 0.3

cannot be separately resolved. The lower energy peak at ~ 43 keV is caused by internal conversion decays occurring within the GAGG scintillator from the first excited states. The internal conversion coefficients are both greater than 1 (3.89 and 5.94 respectively[24]) indicating de-excitation is predominantly via this mechanism. The thermal neutron capture cross sections of ^{155}Gd and ^{157}Gd are sufficiently large that the thermal neutron reaction occurs within tens of micrometers of the surface of the detector. Hence there is a high likelihood of escape by the 43 keV gadolinium K-shell X-ray[33] for approximately half of the internal conversion events. These events produce the lower energy peak seen in Figure 6 due to the conversion electrons that only travel a few micrometres in the scintillator. Interestingly, the intensity of the lowest energy peak was far lower in [9] at $<50\%$ than that of the 1st excited state transition, compared to $\sim 90\%$ shown in this work. The gadolinium-containing scintillator discussed in [9], gadolinium orthosilicate (GSO), was measured to have an energy resolution of 9.6% at 661 keV for a 1 cm cube coupled to a PMT. Reports of the fast neutron response of GAGG by Korzhik et al also report the presence of a neutron-induced γ peak at ~ 90 keV[34].

Figure 6: A typical γ -ray spectrum for thermal neutrons incident on a GAGG:Ce crystal (blue), and a background NaI:Tl spectrum (black).

In Figure 6 we also observe a γ peak at ~ 259 keV, this is a result of the rotational band structure of both ^{156}Gd and ^{158}Gd in which the 4^+ state de-excites in sum-coincidence through the 2^+ state to the ground state. The resulting γ peak is due to the combination of these transitions from both ^{156}Gd and ^{158}Gd isotopes, and was also observed in [9].

The background γ -ray spectrum of the neutron tank as acquired with a NaI(Tl) detector is also displayed in Figure 6 which shows a shallow, broad peak at ~ 560 keV as well as a shoulder around 260 keV. There has been limited data published on the associated γ -ray spectra of isotopic neutron sources but AmBe and PuBe (the source discussed in [9]) have shown peaks at 0.2 and 0.5 MeV[26]. Additionally, the thermal neutron capture cross section of NaI (^{23}Na : 0.43 b and ^{127}I : 6.2 b) results in a contribution from a 563 keV transition in ^{24}Na and numerous $100 < E_\gamma < 300$ keV decays from ^{128}I , which will contribute to the NaI spectrum in Figure 6.

Since the majority of interactions are likely to be close to the surface of the material additional scintillator volume will not improve the neutron detection capability, but rather will have the opposite effect by increasing the sensitivity to background (and internal) γ radiation. The effect of scintillator thickness is demonstrated in Figure 7 for the two pieces of scintillator from Epic Crystal Co. Ltd. The thinner scintillator no longer has the stopping power to fully

Figure 7: Neutron capture γ -ray spectra for GAGG:Ce scintillators of different thicknesses. The thinner scintillator has an improved peak-to-background ratio compared to the 10 mm thick sample.

Figure 8: The response of GAGG:Ce's neutron capture capability to successive layers of cadmium shielding.

resolve the ~ 517 keV peak but the other features remain. The ratio of the ~ 82 keV peak to an extrapolated background is approximately 3.44 ± 0.10 for the 10 mm scintillator compared to $\sim 7.75 \pm 0.46$ for the thinner slice. Furthermore, measurements with the 1 mm sample recorded the best energy resolutions for the neutron peaks at $24.1 \pm 0.2\%$ (~ 82 keV) and $22.7 \pm 0.7\%$ (~ 259 keV).

The impact of additional neutron shielding is displayed in Figure 8 by exploiting the large neutron resonances characteristic of cadmium. There is a

190 clear reduction in total integrated count with each extra layer of shielding. The
82 keV peak-to-background ratio similarly reduces as thermal neutrons are cap-
tured prior to the detector, from ~ 7.75 to 3.98 ± 0.29 for the ~ 3 mm cadmium
layer between the source and detector, and a further reduction to $\sim 1.44 \pm 0.14$
once scattered neutrons are removed as well with the addition of a cadmium
195 “hat”.

4. Conclusions

We have confirmed the sensitivity of GAGG:Ce to thermal neutrons, and
demonstrated the performance of a GAGG-based neutron detector using a
SiPM-coupled detector. In particular, the two low energy peaks observed at
200 43 keV and 82 keV provide a clean signature for thermal neutron detection in
GAGG, even in the presence of a strong γ field. However, the intrinsic proper-
ties that make GAGG a viable candidate for spectroscopic photon applications
such as PET, introduce potential limitations for exploitation as a neutron de-
tector. For an effective neutron detector, the strong detection efficiency for
205 γ rays should be minimised by reducing the scintillator volume. This can be
achieved without any significant reduction in neutron detection efficiency, and
tends to reduce the sensitivity to unwanted high energy γ rays and their as-
sociated Compton background. Smaller detector thicknesses also improve the
overall light collection by minimising internal attenuation. The optimal limit
210 for minimising the detector thickness is not yet known, but should be tested
in terms of scintillator fabrication, retention of neutron response, and γ -ray
insensitivity.

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