Performance comparison between ceramic Ce:GAGG and single crystal Ce:GAGG with digital-SiPM

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Performance comparison between ceramic Ce:GAGG and single crystal Ce:GAGG with digital-SiPM

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ABSTRACT: The Gd₃Al₂Ga₃O₁₂ (Ce:GAGG) is a new inorganic scintillator known for its attractive properties such as high light yield, stopping power and relatively fast decay time. In this study, we fabricated a ceramic Ce:GAGG scintillator as a cost-effective alternative to single crystal Ce:GAGG and, for the first time, investigated their performances when coupled to the digital silicon photomultiplier (dSiPM) — a new type of photosensor designed for applications in medical imaging, high energy and astrophysics. Compared to 3 × 3 × 2 mm³ sized single crystal Ce:GAGG, the translucent ceramic Ce:GAGG, which has a much lower transmittance than the single crystal, was determined to give an output signal amplitude that is approximately 61% of single crystal Ce:GAGG. The energy resolution of the 511 keV annihilation peak of a ²²Na source was measured to be 9.9 ± 0.2% and 13.0 ± 0.3% for the single and ceramic scintillators respectively. On the other hand, the coincidence resolving time (CRT) of ceramic Ce:GAGG was 307 ± 23 ps, better than the 465 ± 37 ps acquired with single crystals — probably attributed to its slightly faster decay time and higher proportion of the fast decay component. The ceramic Ce:GAGG may be a promising cost-effective candidate for applications that do not require thick scintillators such as x-ray detectors and charged particle detectors, and those that require time-of-flight capabilities.

KEYWORDS: Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators); Gamma detectors; Particle detectors; X-ray detectors

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1 Introduction

The development of new inorganic scintillation materials is crucial to improve a radiation detector’s performances for use in fields such as medical imaging, homeland security, and astrophysics. The recently developed cerium doped Gd$_3$Al$_2$Ga$_3$O$_{12}$ (Ce:GAGG) single crystal scintillator is an attractive alternative for room temperature radiation detection and related applications. The high atomic number and the high density of Ce:GAGG gives it a high stopping power. Additionally, it demonstrated high light output with relatively fast decay time and is not self-radiating or hygroscopic [1–3]. The potential of single crystal Gd-based scintillators grown with Czochralski method, such as Ce:GAGG or the more recent cerium doped gadolinium fine aluminium gallate (Ce:GFAG), have shown their potential as possible candidates for positron emission tomography (PET) or gamma spectroscopy when coupled to a silicon photomultiplier (SiPM) [4, 5]. However, while the single crystal Ce:GAGG is bright with light output of $\sim 56,000$ photons/MeV, has a high density of 6.63 g/cm$^3$ and decays with a decay time of 90 ns due to the 5d–4f transition in Ce$^{3+}$ ion [6, 7], the single crystal requires elaborated efforts to grow leading to relatively high production cost.

In comparison, transparent ceramics are known to have several advantages over single crystals, such as cost effectiveness (large-scale production, uniform scintillator and high process yield), feasibility of shape controlling, and better mechanical properties [8, 9]. A major challenge in fabricating ceramic scintillators is its transparency. The presence of defects within a scintillator affects the propagation path of the emitted light photons to the photodetector. In general, polycrystalline
crystals, such as ceramic scintillators, are more translucent than transparent due to residual pores, grain boundaries, secondary phase defect, double refraction, inclusions and surface roughness [10]. Recently, there have been several trials for mass production of Gd-based ceramic scintillators in a cost effective manner compared to the costly single crystal growing method presently used to fabricate such scintillators and results are promising [11, 12].

While similar Ce:GAGG ceramic scintillators have been developed and basic performances such as optical and scintillation properties have been reported [13, 14], few studies on ceramic scintillator readout with SiPM and timing performances have been carried out. For this paper, we fabricated ceramic Ce:GAGG scintillators and the performance (energy and time resolution) of these scintillators are compared with single crystal Ce:GAGG elements when coupled to a dSiPM. Basic properties such as the transparency and decay times are also reported.

2 Experimental method

2.1 Sample and material preparation

We prepared two types of Ce:GAGG scintillators: one is the single crystal (C&A, Japan) grown by Czochralski method and other one is a ceramic type (Chosun Refractories Co. Ltd., South Korea) manufactured by conventional ceramic processing method. To fabricate the ceramic scintillator, Gd$_2$O$_3$, Ga$_2$O$_3$, Al$_2$O$_3$ and Ce powder of 4N purity were mixed and grinded. Green sample compacts were formed by employing cold isostatic pressing (CIP) process and the powder mixture was subsequently sintered at 1700 °C. Each ceramic scintillator slab was about 30 × 50 × 3 mm$^3$ in size.

To compare the performances between ceramic Ce:GAGG and single crystal Ce:GAGG, longer polished samples of 3 × 3 mm$^2$ in cross-sectional area were acquired and cut into shorter (thinner) elements. Two 3 × 3 × 2 mm$^3$ and 3 × 3 × 5 mm$^3$ elements of each scintillator type as shown in figure 2 were assessed for their performances. As a result of the processing procedure, all sides
Figure 2. Photograph of the various Ce:GAGG samples used in this study. (a) Ceramic Ce:GAGG and (b) single crystal Ce:GAGG. The smaller elements are $3 \times 3 \times 2 \text{ mm}^3$ and the larger pieces are $3 \times 3 \times 5 \text{ mm}^3$ in size.

of the $3 \times 3 \times 5 \text{ mm}^3$ scintillators were polished, but only the 4 lateral sides were polished for the shorter $3 \times 3 \times 2 \text{ mm}^3$ scintillators; i.e. the other two sides (top and bottom) were as-cut (rough).

2.2 Digital silicon photomultiplier (dSiPM)

The photosensor used in this study was the digital silicon photomultiplier (dSiPM) from Philips. The dSiPM is basically a conventional analog-SiPM (array of micro-sized Geiger-mode avalanche photodiodes call microcells) integrated with on-die electronics. Each microcell in the dSiPM has its own 1-bit on-die analog-to-digital converter in the form of a CMOS inverter. Therefore, any incoming light photon that triggers a microcell produces a digital pulse; the final dSiPM output of a radiation event is thus the sum of all of digital pulses triggered by the incoming photons (energy information), as well as its time stamp (time information). While an analog SiPM requires dedicated readout circuitries which add electrical noise to the signal with each subsequent stage down the readout chain, electrical noise is minimized as all signals are digitized at the microcell level within the dSiPM [15, 16]. In addition, as each microcell has its own control circuit, microcells with high dark-count rates can be disabled. These properties facilitate detector prototyping and excellent performances have been reported when coupled with a scintillator [17].

The dSiPM package comprises of two dSiPM modules (DPC3200-22-44 sensor arrays) that are connected to a data acquisition module (DAQ base unit) through dedicated cables. During acquisition, data (such as time stamps and number of triggered microcells) are transmitted to a PC from the base unit via a USB cable and stored as a file to be processed. All control and configuration of the dSiPM are also performed at the PC.

Each dSiPM module consists of 4 tiles. Each tile consists of $4 \times 4$ dies and each die is composed of four ($2 \times 2$ array) dSiPM pixels. There is a time-to-digital converter (TDC) at each die for timestamp generation as explained above. The TDC can be configured to give the time stamp of the first arrival photon (trigger level “scheme 1”), $\sim 2.3^{\text{th}}$ photons (trigger level “scheme 2”) and so on. After triggering, the signal is checked against a preset “validation threshold” level. Any event that do not cross the validation threshold within a certain time (called “validation interval”),
is discarded. Events that cross the “validation threshold” within the “validation interval” period are counted for a short time period (“integration interval”) and stored in the PC.

2.3 Experimental setup

Each scintillator described in section 2.1 was wrapped with several layers of Teflon tape on five sides and coupled to a pixel of a digital silicon photomultiplier (dSiPM) photosensor with the silicone optical grease (Saint-Gobain Crystals, BC-630). A dSiPM pixel was $3.2 \times 3.875 \text{ mm}^2$ in active area (slightly bigger than the scintillators) allowing some margin for alignment errors.

Each dSiPM module was attached to a PC cooling fan via thermal conductive compound and was placed in a temperature-controlled cooling chamber. This can keep the modules at a constant temperature as low as $5 \degree C$ depending on the chamber temperature setting.

We used a $^{22}\text{Na}$ source to acquire both the energy and time resolutions and all measurements were conducted in a cooling chamber, maintaining the temperature of the dSiPM at a stable temperature of about $10 \pm 1 \degree C$ as shown in figure 3. This small temperature fluctuation is expected not to have appreciable effect on the results as dSiPM digital signal (the number of microcells fired) and Ce:GAGG light output are known to have weak dependence on the temperature [18]. To suppress dark count noise events, 20% of the noisiest microcells were turned off. The dSiPMs were kept in complete darkness and set as follow: trigger level “scheme 1”, validation threshold “scheme 16”, validation length “40 ns” and integration interval “165 ns”.

2.4 Energy and time resolution

The energy resolution was computed from the 511 keV annihilation peak of the $^{22}\text{Na}$ energy spectrum. However, the finite number of microcells comprising a SiPM (3200 for a dSiPM pixel) is not enough to produce a linear output with varying radiation energy (number of incident light photons). That is, the probability of interaction of two or more photons on the same microcell increases as the number of incident photons increases, leading to saturation (non-linearity) effects [19]. Correction of this effect is crucial for unbiased measurement of the energy resolution for SiPM based detector, otherwise the energy resolution may be distorted and produce unrealistically good energy resolution. To correct for our data, the equation below was applied [20, 21].

$$p = -N \cdot \ln(1 - k/N)$$  \hspace{1cm} (2.1)

where, $N$ is the number of counts of available active cells (total number of microcells that are turned on), $k$ is the number of counts of triggered cells, and $p$ is the number of counts of corrected incident photons.
However, as light photons tend to spill over to neighboring pixels (there is a 200 μm thick protective glass over the sensor), the value $N$ is not a constant. Using $N=3200$ (number of microcells in a dSiPM pixel) would therefore not be accurate. To assess the true energy resolution, we applied the method described in [5]. Rather than simply using a constant value for $N$, we determined $N$ by iterative calculation using the 511 keV and 1.274 MeV photopeaks of $^{22}$Na as references. The final corrected spectrum was then fitted using Gaussian function and a bi-exponential curve. The biexponential fitting was performed on the background around the 511 keV annihilation peak (mostly Compton scatters of the 511 keV and 1.274 Mev gamma radiation) and the energy resolution was acquired from the Gaussian function (see figure 5).

The schematic diagram for the measurement of the coincidence resolving time (CRT) for the pair of scintillator samples is shown in figure 4. The two scintillators under test were setup to face each other with a separation of 5 cm and irradiated with $^{22}$Na source placed between two dSiPMs in the cooling chamber. Data for two dSiPMs in coincidence were using the same dSiPM setting as described earlier and with an additional timing window setting of 20 ns. The time stamps from the two modules (if they fall within the coincidence window) are subtracted, and the histogram of the time differences are plotted to acquired timing spectrum. Only events that fall within ±30% of the 511 keV full energy peak were used to plot the timing spectrum on which a Gaussian curve was fitted to acquire the FWHM CRT.

2.5 Optical properties: decay time and transmittance

In order to elucidate the reasons behind the resulting energy and timing performances of the single crystal and ceramic scintillators, we measured their respective decay time and optical transmittance as follows. The dSiPM is unable to output complete signal waveforms while analog SiPMs are known to filter signals due to their large capacitances. Therefore, a $3 \times 3 \times 2$ mm$^3$ Ce:GAGG
scintillator, wrapped with Teflon reflector, was coupled to a PMT (Hamamatsu, H11934-100) with optical grease and were excited by a $^{137}$Cs source. The PMT was placed in a dark box and the output was connected directly to a fast oscilloscope. A large number of full-energy waveforms were acquired using a digital oscilloscope (Lecroy, WaveRunner 604Zi). The waveforms were aligned at the peak position, superimposed, and the average of data at each time point was calculated. A bi-exponential curve (equation (2.2)) was then fitted on the averaged data.

$$y = A_1 e^{-x/\tau_1} + A_2 e^{-x/\tau_2} \quad (2.2)$$

where $A_1$ and $A_2$ are the amplitudes of the fitted curves, and $\tau_1$ and $\tau_2$ are the slow and fast components of the decay, respectively. The intensity ($I_1$ and $I_2$) of the curves are calculated with the equation [22] below:

$$I_1 = \frac{A_1 \tau_1}{A_1 \tau_1 + A_2 \tau_2} \quad \text{and} \quad I_2 = \frac{A_2 \tau_2}{A_1 \tau_1 + A_2 \tau_2} \quad (2.3)$$

Finally, the optical transparency of both types of scintillators was measured with a UV-VIS-NIR spectrophotometer (Agilent Technologies, Cary 5000). The $3 \times 3 \times 5 \text{mm}^3$ ceramic Ce:GAGG scintillator was wrapped around four sides ($3 \times 2 \text{mm}^2$ surfaces) using Teflon tape and its transmittance was measured. It was placed on the holding plate of the spectrometer and a narrow beam of light in the range of 200–800 nm was allowed to penetrate the scintillator via a small slit. The output signal obtained for the single crystal scintillator was used as the reference to calculate the relative transmittance of the ceramic scintillator.

## 3 Results

### 3.1 Energy resolution and light yield

Figure 5 shows the pulse height spectra acquired with the single crystal Ce:GAGG and the ceramic Ce:GAGG under the excitation from a $^{22}$Na source. Both saturated and corrected spectra are given. After employing saturation correction (see section 2.4), the energy resolution for each scintillator was obtained from the Gaussian fitted curve. However, we did not apply saturation correction on the $3 \times 3 \times 5 \text{mm}^3$ ceramic Ce:GAGG due to its distorted energy peak and absence of a distinct 1.274 MeV peak. Comparing $3 \times 3 \times 2 \text{mm}^3$ Ce:GAGG scintillators, the single crystal yielded better energy resolution ($9.9 \pm 0.2\%$) than that of ceramic scintillator Ce:GAGG ($13.0 \pm 0.3\%$). The $3 \times 3 \times 5 \text{mm}^3$ single crystal Ce:GAGG yielded an energy resolution of $8.8 \pm 0.2\%$ — an unexpected value considering that shorter crystals should have better light collection.

Comparing the position of saturation corrected annihilation photon peak of the two mm thick ceramic and single crystal scintillators, the dSiPM signal amplitude (corrected number of fired microcells before converting into keV, not shown in figure) of the ceramic scintillator was 39% lower than that of the single crystal. Between single crystal elements, the output of the five mm thick scintillator was 15% lower than its two mm thick counterpart due to internal reflection losses in the longer crystal or sample variation.

### 3.2 Transparency and decay time

The transmittance of the single crystal Ce:GAGG was measured to be much higher than that of ceramic Ce:GAGG at room temperature in air. For both single crystal and ceramic Ce:GAGG,
Figure 5. Saturated energy (left column) and corrected energy spectra (right column) of the tested Ce:GAGG with a $^{22}$Na source. (a) Single crystal saturated spectrum ($3 \times 3 \times 2\,\text{mm}^3$) and (b) its corrected spectrum; (c) single crystal saturated spectrum ($3 \times 3 \times 5\,\text{mm}^3$) and (d) its corrected spectrum; (e) ceramic scintillator saturated spectrum ($3 \times 3 \times 2\,\text{mm}^3$), and (f) its corrected energy spectrum; (g) ceramic scintillator saturated spectrum ($3 \times 3 \times 5\,\text{mm}^3$). Saturated correction was not performed on the last data.
Figure 6. Relative transmittance spectra of two mm thick ceramic Ce:GAGG (red) and single crystal Ce:GAGG (blue) obtained from UV-VIS-NIR spectrometer (Agilent Technologies, Cary 5000). The transmittance of the translucent ceramic Ce:GAGG is nearly two orders of magnitude lower than the single crystal.

![Relative transmittance spectra of two mm thick ceramic Ce:GAGG (red) and single crystal Ce:GAGG (blue).](image)

Figure 7. The decay curve, after superimposing and averaging multiple waveforms (blue), of single crystal Ce:GAGG (left) and ceramic Ce:GAGG (right) excited by $^{137}$Cs at room temperature. The fitted curve is plotted in red.

![Decay curve of single crystal Ce:GAGG (blue) and ceramic Ce:GAGG (right).](image)

we could observe the absorption band of $4f \rightarrow 5d$ transition of Ce$^{3+}$ around 400 nm and the main transmittance was over 550 nm.

The averaged waveforms used to measure decay time of the single crystal and ceramic Ce:GAGG are shown in figure 7. From the results of the fitting, the fast and slow decay components of the single crystal Ce:GAGG were 92.5 ns ($I_1$ of equation (2.3) = 56.9%) and slow 220 ns ($I_2$ = 43.1%) respectively, while that of the ceramic scintillator were 88.5 ns ($I_1$ = 72.6%) and 313 ns ($I_2$ = 27.4%).

3.3 Coincidence resolving time (CRT)

The CRT spectra for the 4 pairs of crystals are shown in figure 8. We obtained a time resolution of 465 ± 37 ps ($3 \times 3 \times 2$ mm$^3$) and 492 ± 41 ps ($3 \times 3 \times 5$ mm$^3$) for the single crystal Ce:GAGG.
Figure 8. CRT spectra and Gaussian fitted curve obtained with a $^{22}$Na source using a pair of (a) single crystal Ce:GAGG $(3 \times 3 \times 2 \text{ mm}^3)$, (b) single crystal Ce:GAGG $(3 \times 3 \times 5 \text{ mm}^3)$, (c) ceramic Ce:GAGG $(3 \times 3 \times 2 \text{ mm}^3)$, and (d) ceramic Ce:GAGG $(3 \times 3 \times 5 \text{ mm}^3)$.

pair as shown in figure 8 (a) and (b), respectively. On the other hand, the time resolution of ceramic Ce:GAGG $(3 \times 3 \times 2 \text{ mm}^3)$ was found to be $307 \pm 23$ ps (figure 8(c)), better than that of single crystal scintillator. This degraded to $716 \pm 62$ ps (figure 8(d)) with thicker 5 mm ceramic scintillator.

4 Summary and discussion

The summary of our results is presented in table 1. In this study, we fabricated elements of ceramic Ce:GAGG scintillator as an alternative to its single crystal counterpart and evaluated their performance. The ceramic scintillator elements are opaque, which is typical for the polycrystalline ceramic materials. Upon measurement of its transmittance (two mm thick sample), the transmittance was about two orders of magnitude lower than for the single crystal Ce:GAGG of the same thickness. However, this does not reflect the true transmittance of the ceramic scintillator because most of the spectrophotometer’s light photons scatter as they penetrate the opaque ceramic scintillator and are not collected by photodetector of the spectrophotometer. A more elaborate setup would be required to effectively collect transmitted photons and assess its true transmittance.

The translucent nature of ceramic Ce:GAGG leads to incomplete light collection depending on the radiation interaction position (depth) within the scintillator. This phenomenon is manifested via the distorted 511 keV gamma peak (figure 5(g)) for the five mm thick ceramic crystal. With thinner two mm thick ceramic scintillator, a less distorted energy spectrum (511 keV gamma peak, see figure 5(f)) was acquired. The energy resolution, however, was about 30% worse than for the single crystal scintillators due to its translucent nature and lower light output as reflected in dSiPM signal amplitude (corrected number of microcells fired). Note that the dSiPM signal amplitude is
Table 1. Summary of key properties of the $3 \times 3 \times 2$ mm$^3$ single crystal and ceramic Ce:GAGG scintillators.

<table>
<thead>
<tr>
<th>parameters</th>
<th>Single crystal</th>
<th>Ceramic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Signal amplitude (AU)</td>
<td>1</td>
<td>0.61</td>
</tr>
<tr>
<td>Energy resolution (%)</td>
<td>9.9 ± 0.2 %</td>
<td>13.0 ± 0.3 %</td>
</tr>
<tr>
<td>Time resolution (ps)*</td>
<td>465 ± 37</td>
<td>307 ± 23</td>
</tr>
<tr>
<td>Decay time (ns)*</td>
<td>Fast (92.5, I = 56.9%)</td>
<td>Fast (88.5, I = 72.6%)</td>
</tr>
<tr>
<td></td>
<td>Slow (220, I = 43.1%)</td>
<td>Slow (313, I = 27.4%)</td>
</tr>
</tbody>
</table>

*: Error values denote the one sigma fitting errors.

not a true representation of the light yield due to scatters (and losses) within the ceramic scintillator. Therefore, both the output dSiPM signal and energy resolution may be improved for ceramic scintillator elements thinner than those used in this study.

The time resolution was better with a pair of two mm thick ceramic scintillator. Despite having a lower light yield, the time resolution was better than for the single crystal elements of the same size. This may be attributed to the higher proportion and slightly faster decay time of the fast decay component of ceramic scintillators [22].

5 Conclusion

A ceramic Ce:GAGG scintillator that can potentially be a cost-effective alternative to single crystal scintillator has been fabricated. Two mm thick ceramic scintillator was measured to show lower signal amplitude output and energy resolution, but better time resolution compared to the single crystal Ce:GAGG. In all, considering its translucent nature, ceramic Ce:GAGG can be a promising cost-effective candidate for applications that do not require thick scintillators such as x-ray detectors [23] and charged particle detectors [24, 25], those that require time-of-flight capabilities [26], and even PET detectors with non-conventional readout configurations [17, 27].

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References


