Performance of Novel Oxide Scintillator Ce:((La,Gd)$_2$Si$_2$O$_7$ with a High Energy Resolution

Akira Suzuki$^1$, Shunsuke Kurosawa$^{1,2}$, Toetsu Shishido$^1$, Jan Pejchal$^{1,3}$, Yuui Yokota$^1$, Yoshisuke Futami$^1$, and Akira Yoshikawa$^{1,2}$

$^1$ Institute for Materials Research, Tohoku University
$^2$ New Industry Creation Hatchery Center, Tohoku University
$^3$ Institute of Physics, AS CR

Contents:
1. Introduction
2. Evaluation of La-GPS with PMT
3. Evaluation of La-GPS with MPPC
4. Summary

Vienna Conference on Instrumentation 2013
Introduction: Food Monitoring in Fukushima

Fukushima Problem

All foods in Fukushima or surrounding area are checked
[ Foods < 100 Bq/kg, water < 10 Bq/kg ]

Application Requirements
- Energy resolution [ΔE/E < 5-6%]
- low intrinsic background
- high detection efficiency
Introduction: Compton Camera

\[ \Delta \phi \text{ Angular resolution (ARM)} \]

\[ \cos \phi = 1 - \frac{m_e c^2 K}{(E+K) E} \]

\( \Delta E/E \): energy resolution of absorber (scintillator) @ 662 keV, FWHM

Application Requirements
- Energy resolution \([\Delta E/E < 5 \%]\)
- low intrinsic background
- high detection efficiency

Kurosawa et al. (2008)

<table>
<thead>
<tr>
<th>( \Delta E/E )</th>
<th>Com. Angular Res. @662keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>10.5 %</td>
<td>4.2°</td>
</tr>
<tr>
<td>7%</td>
<td>3.1°</td>
</tr>
<tr>
<td>5%</td>
<td>2.3°</td>
</tr>
<tr>
<td>3%</td>
<td>2.1°</td>
</tr>
</tbody>
</table>

Doppler broadening (Ar) Zoglauer et al. (2003)
## Introduction: Various Scintillators

<table>
<thead>
<tr>
<th></th>
<th>Density (g/cm³)</th>
<th>attenuation length [mm] @511keV</th>
<th>Decay time constant (ns)</th>
<th>Light output (Relative)</th>
<th>Hydroscopic nature</th>
<th>Radiation Hardness</th>
<th>energy res. @662 keV [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tl:NaI</td>
<td>3.67</td>
<td>2.6</td>
<td>230</td>
<td>1</td>
<td>Strong</td>
<td>very weak</td>
<td>7</td>
</tr>
<tr>
<td>Tl:CsI</td>
<td>4.53</td>
<td>18.6</td>
<td>1050</td>
<td>0.85</td>
<td>Weak</td>
<td>very weak</td>
<td>7</td>
</tr>
<tr>
<td>BGO</td>
<td>7.13</td>
<td>large 1.1</td>
<td>300</td>
<td>0.07-0.12</td>
<td>No</td>
<td>weak</td>
<td>7</td>
</tr>
<tr>
<td>Ce:LSO</td>
<td>7.4</td>
<td>large 0.87</td>
<td>fast 40</td>
<td>0.4</td>
<td>No</td>
<td>strong</td>
<td>8</td>
</tr>
<tr>
<td>Ce:GSO</td>
<td>6.71</td>
<td>1.3</td>
<td>30-60</td>
<td>0.18</td>
<td>No</td>
<td>very strong</td>
<td>8</td>
</tr>
<tr>
<td>Ce:LaBr₃</td>
<td>5.3</td>
<td>2.2</td>
<td>20</td>
<td>1.6</td>
<td>Very strong</td>
<td>strong</td>
<td>3</td>
</tr>
</tbody>
</table>

Although halide scintillators have good properties, they are strongly hydroscopic.
Introduction: Gd$_2$Si$_2$O$_7$ (GPS) Scintillator

Gadolinium Pyro-Silicate Ce:Gd$_2$Si$_2$O$_7$ (Ce:GPS)

10 mol% Ce-doped GPS crystals grown by Floating Zone method [1].

Reference:
Introduction: Quenched Luminescence in Ce:GPS

Problem: Concentration Quenching

Much Higher Performance

Case: Ce:GSO [2]

We challenged to develop low Ce-doped GPS.

Dependency of light output on Ce concentration in Ce:GPS scintillator [1]

Reference
Introduction: (La, Gd)$_2$Si$_2$O$_7$ (La-GPS)

Ce:(La, Gd)$_2$Si$_2$O$_7$ (Ce:La-GPS)

**Conventional GPS**

| 10 mol% Ce |

**Novel La-GPS**

| 1 mol% Ce for emission center |
| 9 mol% La for stabilizing of crystal |

---

**Material Design**

Comparable Ionic Radius

$R_{Ce^{3+}} \sim R_{La^{3+}}$

---

Dieke diagram

Research Sample

Material Preparation

Starting Material:
$\text{Gd}_2\text{O}_3$, $\text{La}_2\text{O}_3$
$\text{SiO}_2$, $\text{Ce}_2\text{O}$
99.99% Purity Powder

Floating Zone (FZ) Method

As-grown Crystal

Under UV Lamp

Ce 1mol% doped La-GPS Crystal

5 mm
Transmittance

Ce$^{3+}$ 4f – 5d transitions

Measurement Instruments:
- Spectrometer: V-530 (JASCO)
- Excitation Source:
  - Deutrium lamp (200 – 360 nm)
  - Xe lamp (360 – 900 nm)
Photoluminescence

Measurement Instruments:
Spectrofluorometer: FLS920 (Edinburgh Instruments)
Excitation Source: Xe lamp Xe-900 (Edinburgh Instruments)
Broad scintillation luminescence was observed around 390 nm due to Ce\textsuperscript{3+} 5d–4f transitions under alpha-ray excitation. 

**Measurement Instruments:**

- Spectrofluorometer
- Excitation Source
- FLS920 (Edinburgh Instruments)
- \textsuperscript{241}Am Alpha ray Source
Photo-detector (I)

PMT

R7600U-200
Hamamatsu

18 mm

Ultra bi-alkali photocathode
Effective area: 18 x 18 mm

Quantum Efficiency (%)

La-GPS Emission Length

QE>40% @ 390 nm

Reference: R7600U Data Sheet (HPK)
Pulse Height Spectra with PMT

La-GPS $^{137}\text{Cs}$
- Escape peak (Gd Kα)
- 662 keV

La-GPS $^{133}\text{Ba}$
- 276 keV
- 303 keV
- 356 keV

La-GPS $^{22}\text{Na}$
- 511 keV

La-GPS $^{54}\text{Mn}$
- 835 keV

Temperature: Constantly 26.0 ± 0.1 °C in all the measurements.
Preamplifier: ORTEC MODEL 113
Shaper: ORTEC MODEL 572A, Shaping Time: 1 us
Light Output with PMT

Emission Wavelength:
- BGO: 480 nm
- La-GPS: 390 nm

When BGO ~ 8000 photon/MeV,
La-GPS ~ **39000** photon/MeV

\(^{137}\text{Cs} \ 662 \text{ keV} \)
Energy Resolution with PMT

La-GPS Energy Resolution ~ 4.7% FWHM at 662 keV

\[
\frac{\Delta E}{E} \% = (4.52) \cdot \left( \frac{\text{Energy (keV)}}{662 \text{ (keV)}} \right)^{-0.51}
\]
### Scintillation Decay

#### Measurement System

- **Crystal**
- **PMT**
  - Hamamatsu R7600U
- **Oscilloscope**
  - Tektronix TDS3052B

#### Exponential Fitting:

\[
I = I_0 + \sum_{n=1}^{2} A_n \exp \left( -\frac{t - t_0}{\tau_n} \right)
\]

- **I**: Intensity, **t**: Time, **\( \tau_n \)**: Decay Constant

#### Decay Time and Fitting Error

<table>
<thead>
<tr>
<th>Decay Time (ns)</th>
<th>( \tau_1 )</th>
<th>( \tau_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>45.97</td>
<td>345.6</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fitting Error (ns)</th>
<th>0.05</th>
<th>0.8</th>
</tr>
</thead>
</table>

| Ratio (%) | 79   | 21   |
Photo-detector (II)

**MPPC**

**S10362-33-050C**
Hamamatsu

Pixel Size: 50 x 50 μm
Number of Pixels: 3600
Effective area: 3 x 3 mm

**La-GPS Emission Length**

Reference: S10362-33-050C Data Sheet (HPK)
Pulse Height Spectra with MPPC

La-GPS ($^{137}$Cs)
- Escape peak (Gd K$\alpha$)
- 662 keV

La-GPS ($^{133}$Ba)
- 276 keV
- 303 keV
- 356 keV

La-GPS ($^{22}$Na)
- Escape peak (Gd K$\alpha$)
- 511 keV

La-GPS ($^{54}$Mn)
- 835 keV

Temperature: Constantly 26.0 ± 0.1 °C in all the measurements.
Preamplifier: Time Constant 5.4 us
Shaper: ORTEC MODEL 572A, Shaping Time: 1 us
Energy Resolution with MPPC

Fitting Function

\[ Ch = \beta \left( 1 - \exp \left( -\frac{N_{\text{photon}} \cdot E \cdot \text{PDE}}{N_{\text{Pixel}}} \right) \right) \]

- \( N_{\text{photon}} \): Number of photons which traveled into MPPC
- \( E \): Energy of gamma rays
- \( \text{PDE} \): Photon detection efficiency
- \( N_{\text{Pixel}} \): Number of pixels
- \( \beta \): Constant

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>Radiation Source</th>
<th>Energy Resolution (%, FWHM)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>MPPC</td>
</tr>
<tr>
<td>835</td>
<td>(^{54}\text{Mn})</td>
<td>8.5</td>
</tr>
<tr>
<td>662</td>
<td>(^{137}\text{Cs})</td>
<td>9.5</td>
</tr>
<tr>
<td>511</td>
<td>(^{22}\text{Na})</td>
<td>10.2</td>
</tr>
<tr>
<td>356</td>
<td>(^{133}\text{Ba})</td>
<td>10.7</td>
</tr>
</tbody>
</table>
## Comparison of La-GPS with Other Scintillators

<table>
<thead>
<tr>
<th></th>
<th>Density (g/cm³)</th>
<th>attenuation length[mm]@511keV</th>
<th>Decay time constant (ns)</th>
<th>Light output (Relative)</th>
<th>Hydroscopic nature</th>
<th>Radiation Hardness</th>
<th>energy res. @662 keV [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tl:NaI</td>
<td>3.67</td>
<td>2.6</td>
<td>230</td>
<td>1</td>
<td>Strong</td>
<td>very weak</td>
<td>7</td>
</tr>
<tr>
<td>Tl:CsI</td>
<td>4.53</td>
<td>18.6</td>
<td>1050</td>
<td>0.85</td>
<td>Weak</td>
<td>very weak</td>
<td>7</td>
</tr>
<tr>
<td>BGO</td>
<td>7.13</td>
<td>large 1.1</td>
<td>300</td>
<td>0.07-0.12</td>
<td>No</td>
<td>weak</td>
<td>7</td>
</tr>
<tr>
<td>Ce:LSO</td>
<td>7.4</td>
<td>large 0.87</td>
<td>fast 40</td>
<td>0.4</td>
<td>No</td>
<td>strong</td>
<td>8</td>
</tr>
<tr>
<td>Ce:GSO</td>
<td>6.71</td>
<td>1.3</td>
<td>30-60</td>
<td>0.18</td>
<td>No</td>
<td>very strong</td>
<td>8</td>
</tr>
<tr>
<td>Ce:LaBr₃</td>
<td>5.3</td>
<td>2.2</td>
<td>20</td>
<td>1.6</td>
<td>Strong</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Ce:La-GPS</td>
<td>---</td>
<td>---</td>
<td>46</td>
<td>0.6</td>
<td>No</td>
<td>---</td>
<td>4.7</td>
</tr>
</tbody>
</table>
Summary

- Novel Scintillator La-GPS was successfully developed.
- Although La-GPS is an oxide scintillator, it has a high light output and good energy resolution (FWHM) comparable with halide scintillators.

<table>
<thead>
<tr>
<th>Emission Wavelength (nm)</th>
<th>Light Output (photon/MeV)</th>
<th>Energy Resolution (%, FWHM at 662 keV)</th>
<th>Scintillation Decay Time (ns)</th>
<th>Hygroscopic Nature</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>PMT</td>
<td>MPPC</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.7</td>
<td>9.5</td>
<td>45.97 (79%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>345.6 (21%)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>None</td>
</tr>
</tbody>
</table>

Future Plan

- Measurement of time resolution for La-GPS scintillator with a MPPC