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Scintillation Response of LSO:Ce and NaI:Tl Single Crystal Scintillators

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ABSTRACT

Scintillation response of two inorganic scintillators (LSO:Ce and NaI:Tl) were investigated. The light yield and energy resolution were measured using photomultiplier tube (PMT) readout. The light yield non-proportionality of both crystals was studied for γ-ray energies ranging from 31 to 1274.5 keV. The intrinsic resolution of both crystals versus energy of γ-rays has been determined after correcting the measured energy resolution for photomultiplier tube statistics. The photo fraction was determined for both crystals and compared with the cross sections ratio calculated using WinXCOM program. For 662 keV gamma rays (137 Cs source), the study showed that the light yield of LSO:Ce is superior to NaI:Tl, and its photo fraction is also higher than that of NaI:Tl.

Key words: energy resolution, light yield non-proportionality, LSO:Ce, NaI:Tl

INTRODUCTION

Inorganic scintillators play a major role in many fields of radiation detection, including medical imaging, astrophysics, high energy physics and exploring resources like oil. The last decade has seen the introduction of several new high luminosity scintillators, in particular Ce-doped complex oxide crystals that are promising candidates for these applications [1-4].

Lu₂SiO₅:Ce (LSO:Ce) [5] has been developed as promising scintillators for positron emission tomography (PET) due to its desirable properties such as high density, fast decay time and high light output. Crystal has the emission spectra peaking at 420 nm and exhibit the highest light yield up to about 30,000 ph/MeV [6].

In this paper, we present the scintillation response of LSO:Ce crystal and compare to these of the classical one (NaI:Tl). The light yield, energy resolution of both crystals was calculated. The estimated photofraction for both crystals at 662 keV γ-peak will also be discussed.

Materials And Methods

LSO:Ce crystal with the size of 10x10x2 mm³ was supplied by Saint-Gobain (France). Photoelectron yield and energy resolution were measured by coupling the samples to Photonis XP5200B PMT using silicone grease. In order to maximize light collection, the samples were optically coupled by silicone grease to the PMT and covered with several layers of white Teflon (PTFE) tape in a configuration of a reflective umbrella. The signal from the PMT anode was passed to a Canberra 2005 preamplifier and then to a Tennelec TC243 spectroscopy amplifier. The measurements were carried out with 4 µs shaping time constant in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8k7[7], was used to record pulse height spectra. Gaussian functions were fitted to the full energy peak, using procedures in the MCA, to determine their position and energy resolution.

The photoelectron yield, expressed as a number of photoelectrons released from the PMT photocathode per MeV (phe/MeV) of γ-energy deposited in the crystal, was determined by means of a single photoelectron method [8,9]. In this method the number of photoelectrons is measured by comparing the position of a full energy peak of γ-rays detected in the crystals with that of the single photoelectron peak from the PMT photocathode. The measurements of light yield non-proportionality and energy resolution were carried out for a series of X/γ-rays emitted by different radioactive sources in the energy range from 31 to 1274.5 keV. All

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measurements were carried out at room temperature (RT).

Results And Discussion

Photoelectron Yield and Energy Resolution:

Fig. 1 presents the pulse height spectra of 662 keV γ-rays from a $^{137}$Cs source measured with LSO:Ce and NaI:Tl detectors. The energy resolution of 10.3% obtained with LSO:Ce is superior compared to the value of 11.5% obtained with NaI:Tl. This is due to much higher photoelectron yield for LSO:Ce, see below. Note a higher photofraction in the spectrum obtained with LSO:Ce, as would be expected due to higher effective atomic number and density of the LSO:Ce material.

![Pulse Height Spectra](image)

**Fig. 1:** Pulse Height Spectra of γ-Rays from $^{137}$Cs(662 keV) Source as Measured with LSO:Ce and NaI:Tl Crystals.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Photoelectron Yield [phe/MeV]</th>
<th>Light Yield [ph/MeV]</th>
<th>Energy Resolution [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSO:Ce</td>
<td>8,830 ± 90</td>
<td>3,270 ± 3,300</td>
<td>10.3 ± 0.5</td>
</tr>
<tr>
<td>NaI:Tl</td>
<td>4,540 ± 90</td>
<td>1,520 ± 1,500</td>
<td>11.5 ± 0.6</td>
</tr>
</tbody>
</table>

Table 1 summarizes comparative measurements of photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the tested crystals coupled to the Photonis XP5200B PMT, as measured at 4 μs shaping time constant in the spectroscopy amplifier. The LSO:Ce showed a photoelectron yield of 8,830 phe/MeV. This value corresponds to about 32,700 Photon/MeV (ph/MeV) at the PMT photocathode quantum efficiency (QE) of 27.0% for peak emission of 420 nm. The tested NaI:Tl showed a photoelectron yield of 4,540 phe/MeV. This value corresponds to about 15,200 ph/MeV at a QE of 29.9% for peak emission of 415 nm. Note the light yield of 32,700 ph/MeV for LSO:Ce is slightly higher than the value of 31,900 ph/MeV measured for big sample (6x6x10 mm³) in Ref. [6].

The energy resolution \((\Delta E/E)^2\) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [10]

\[
(\Delta E/E)^2 = (\delta_e)^2 + (\delta_p)^2 + (\delta_a)^2,
\]

where \(\delta_e\) is the intrinsic resolution of the crystal, \(\delta_p\) is the transfer resolution and \(\delta_a\) is the statistical contribution of PMT to the resolution. The statistical uncertainty of the signal from the PMT can be described as

\[
\delta_a = 2.355 \times \frac{1}{N^{1/2}} \times (1 + \varepsilon)^{1/2},
\]

where \(N\) is the number of the photoelectrons and \(\varepsilon\) is the variance of the electron multiplier gain, equal to 0.1 for an XP5200B PMT. The transfer component...
depends on the quality of optical coupling of the crystal and PMT, homogeneity of quantum efficiency of the photocathode and efficiency of photoelectron collection at the first dynode. The transfer component is negligible compared to the other components of the energy resolution, particularly in the dedicated experiments. The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [10, 11] and many effects such as inhomogeneities in the scintillator which can cause local variations in the scintillation light output and non-uniform reflectivity of the reflecting cover of the crystal. Overall energy resolution and PMT resolution can be determined experimentally. If $\delta_p$ is negligible, intrinsic resolution $\delta_{ic}$ of a crystal can be written as follows

$$ (\delta_{ic})^2 = (\Delta E / E)^2 - (\delta_{i})^2. \quad (3) $$

Figs.3 and 4 present the measured energy resolution versus energy of $\gamma$-rays for LSO:Ce and NaI:Tl crystals, respectively. Other curves shown in Figs.3 and 4 represent the PMT resolution calculated from the number of photoelectrons and the intrinsic resolution of the crystals calculated from Eq. (3). Apparently, the energy resolution for the both crystals is mainly contributed by the intrinsic resolution over the whole energy range from 31 to 1,274.5 keV.

![Graph 3: Overall Energy Resolution Contributed Factors versus Energy of LSO:Ce Crystal.](image_url)

![Graph 4: Overall Energy Resolution Contributed Factors versus Energy of NaI:Tl Crystal.](image_url)
Overall energy resolution of LSO:Ce and NaI:Tl detectors versus energy is shown in Fig. 5. The energy resolution of LSO:Ce is superior than that of NaI:Tl in the energy range from 31 to 320 keV.

Fig. 5: Overall Energy Resolution of LSO:Ce and NaI:Tl Crystals versus Energy of γ-rays.

Fig. 6: Intrinsic Resolution of LSO:Ce and NaI:Tl Crystals.

Fig. 6 presents a direct comparison of the intrinsic resolution for the tested crystals. The intrinsic resolution of LSO:Ce crystal is better than that of NaI:Tl crystal in the energy range from 31 to 320 keV.

Table 2: Analysis of the 662 keV Energy Resolution for LSO:Ce and NaI:Tl Detector.

<table>
<thead>
<tr>
<th>Detector</th>
<th>N [electron]</th>
<th>ΔE/E [%]</th>
<th>δε [%]</th>
<th>δε [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSO:Ce+XP5200B</td>
<td>5480 ± 550</td>
<td>10.3 ± 0.5</td>
<td>3.2 ± 0.2</td>
<td>9.8 ± 0.5</td>
</tr>
<tr>
<td>NaI:Tl+XP5200B</td>
<td>2840 ± 280</td>
<td>11.5 ± 0.6</td>
<td>4.6 ± 0.2</td>
<td>10.5 ± 0.5</td>
</tr>
</tbody>
</table>

To better understand the energy resolution of the tested crystals in γ-ray spectrometry, the contribution of various components to the overall energy resolution was analyzed for 662 keV photopeak, and the results are presented in Table 2. The second column gives N, the number of photoelectrons produced from the photocathode. The third column gives ΔE/E, the overall energy resolution at 662 keV.
photopeak. The PMT contribution (\( \delta_\text{PMT} \)) was calculated using (2). From the values of \( \Delta E/E \) and \( \delta_\text{e} \), the intrinsic resolution (\( \delta_{\text{int}} \)) was calculated using (3).

The superior energy resolution of LSO:Ce as compared to NaI:Tl is mainly due to a small contribution of both \( \delta_\text{e} \) and \( \delta_{\text{PMT}} \), which seems to follow a high light output (almost a factor of two) for LSO:Ce crystal.

**Non-proportionality of the Light Yield:**

Light yield non-proportionality as a function of energy can be one of the important reasons for degradation in energy resolution of scintillators\[11]\]. The non-proportionality is defined here as the ratio of photoelectron yield measured for photopeaks at specific \( \gamma \)-ray energy relative to the yield at 662 keV \( \gamma \)-peak.

Fig. 7 presents the light yield non-proportionality characteristics of LSO:Ce and NaI:Tl crystals in the energy range of 31 to 1,274.5 keV. LSO:Ce crystal is clearly poorer than NaI:Tl crystals in terms of light yield proportionality. To better understand, the degree of non-proportionality (\( \sigma_{\text{np}} \)) proposed by Dorenbos \[12]\] was calculated with this energy range, we obtain a value of 0.18 for LSO:Ce and 0.08 for NaI:Tl. The degree of non-proportionality of 0.18 for LSO:Ce and 0.08 for NaI:Tl is lower than that of about 0.22 for LSO:Ce and 0.12 for NaI:Tl in Ref.\[12]\], respectively. This result shows an improvement of \( \sigma_{\text{np}} \) for the tested crystals. Although LSO:Ce crystal is clearly poorer than NaI:Tl crystals in terms of light yield proportionality but its intrinsic resolution is slightly better. Other effects such as inhomogeneities in the NaI:Tl crystal and non-uniform reflectivity of the reflecting cover of the NaI:Tl crystal can be one of the important reasons for degradation in its intrinsic resolution.

![Graph showing non-proportionality of light yield](image)

**Fig. 7: Non-proportionality of the Light Yield of LSO:Ce and NaI:Tl Crystals.**

**Photofraction:**

<table>
<thead>
<tr>
<th>Crystal</th>
<th>( Z_{\text{eff}} )</th>
<th>Density [g/cm(^2)]</th>
<th>Volume [cm(^3)]</th>
<th>Photofraction [%]</th>
<th>( \sigma )-ratio [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSO:Ce</td>
<td>64.4</td>
<td>7.40</td>
<td>0.200</td>
<td>26.9 ± 2.7</td>
<td>24.0</td>
</tr>
<tr>
<td>NaI:Tl</td>
<td>50.8</td>
<td>3.67</td>
<td>0.785</td>
<td>18.4 ± 1.8</td>
<td>11.2</td>
</tr>
</tbody>
</table>

The photofraction is defined here as the ratio of counts under the photopeak (including the escape peak) to the total counts of the spectrum as measured at a specific \( \gamma \)-ray energy. The photofraction for tested LSO:Ce and NaI:Tl crystals at 662 keV \( \gamma \)-peak is collected in Table 3. For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCom program \[13]\] are given too. The data indicate that LSO:Ce crystal shows much higher photofraction than NaI:Tl crystal in a same trend with the cross-section ratio (\( \sigma \)-ratio) obtained from WinXCom program. The reason is due to much density and effective atomic number (\( Z_{\text{eff}} \)) of the LSO:Ce crystal although its volume is less.
Conclusion:

Scintillation response of LSO:Ce and NaI:Tl crystals were tested and compared in γ-ray spectrometry. The energy resolution of LSO:Ce is superior than that of NaI:Tl due to a high light output and small contribution from its intrinsic resolution. Moreover, inhomogeneities of dopant and some defects in the LSO:Ce and NaI:Tl crystals could affect the energy resolution, and the crystalline quality of this sample could be further improved. In conclusion, the main advantages of LSO:Ce are non-hygroscopicity, high density and photofraction which make it very promising scintillator for high energy γ-ray detection and medical imaging.

Acknowledgements

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References