

การศึกษาเปรียบเทียบซินทิลเลเตอร์ $\text{Lu}_{1.8}\text{Y}_{0.2}\text{SiO}_5:\text{Ce}$ และ $\text{LaCl}_3:\text{Ce}$ สำหรับวัดสเปกตรัมรังสีแกมมา

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บทคัดย่อ

งานวิจัยนี้ได้ศึกษาการตอบสนองด้านซินทิลเลชันของผลึก $\text{Lu}_{1.8}\text{Y}_{0.2}\text{SiO}_5:\text{Ce}$ (LYSO:Ce) และ $\text{LaCl}_3:\text{Ce}$ โดยการกระตุ้นด้วยรังสีแกมมา และใช้หลอดทวิคูณแสงเป็นอุปกรณ์วัดแสง จากการวัดการแยกชัดพลังงาน (energy resolution) ของผลึกทั้งสองชนิด ด้วยหลอดทวิคูณแสงเบอร์ XP 5200B พบว่าการแยกชัดพลังงานรังสีแกมมา 662 keV จาก ^{137}Cs มีค่าร้อยละ 4.5 ± 0.2 และร้อยละ 8.2 ± 0.4 สำหรับ $\text{LaCl}_3:\text{Ce}$ และ LYSO:Ce ตามลำดับ โฟโตแฟรกชัน (photofraction) ที่พลังงาน 662 keV สำหรับ LYSO:Ce มีค่าร้อยละ 26.1 ซึ่งสูงกว่าค่าร้อยละ 15.7 สำหรับ $\text{LaCl}_3:\text{Ce}$ นอกจากนี้ ยังได้ทำการวัดความไม่เป็นสัดส่วนของยิลด์แสง (non-proportionality of the light yield) และการแยกชัดพลังงานที่แปรตามพลังงานรังสีแกมมา และนำไปสู่การคำนวณค่าการแยกชัดในตัวของผลึก บทความนี้เน้นถึงความเกี่ยวพันระหว่างการแยกชัดในตัวกับความไม่เป็นสัดส่วนของยิลด์แสงสำหรับซินทิลเลเตอร์

คำสำคัญ : การแยกชัดพลังงาน / การแยกชัดในตัว / $\text{LaCl}_3:\text{Ce}$ / LYSO:Ce / ความไม่เป็นสัดส่วนของยิลด์แสง / โฟโตแฟรกชัน / ซินทิลเลเตอร์

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Comparison of $\text{Lu}_{1.8}\text{Y}_{0.2}\text{SiO}_5:\text{Ce}$ and $\text{LaCl}_3:\text{Ce}$ Scintillators in γ -Ray Spectrometry

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Abstract

The scintillation response of $\text{Lu}_{1.8}\text{Y}_{0.2}\text{SiO}_5:\text{Ce}$ (LYSO:Ce) and $\text{LaCl}_3:\text{Ce}$ scintillators were compared under γ -ray excitation using photomultiplier tube (PMT) readout. For 662 keV γ -rays (^{137}Cs source), energy resolution of $4.5\pm 0.2\%$ obtained for $\text{LaCl}_3:\text{Ce}$ coupled to XP5200B PMT is much better than that of $8.2\pm 0.4\%$ for LYSO:Ce. The estimated photofraction of 26.1% at 662 keV for LYSO:Ce is higher than that of 15.7% for $\text{LaCl}_3:\text{Ce}$. The non-proportionality of the light yield and energy resolution versus γ -ray energy were measured and the intrinsic resolution of the crystals was calculated. Special attention was devoted to the correlation between intrinsic resolution and non-proportional response of scintillators.

Keywords : Energy Resolution / Intrinsic Resolution / $\text{LaCl}_3:\text{Ce}$, LYSO:Ce / Non-proportionality of the Light Yield / Photofraction / Scintillator

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1. Introduction

Inorganic scintillators play an important role in detection and spectroscopy of energetic photons and nuclear particles. Important requirements for the scintillation crystals used in these applications include high light output, fast response time, high stopping power and good energy resolution. During the last two decades, new types of scintillators, in particular, Ce-doped inorganic scintillators were intensively studied and some of them were successfully industrialized, for recent reviews see [1-4].

$\text{Lu}_2\text{SiO}_5\text{:Ce}$ (LSO:Ce) [5] and $(\text{Lu,Y})_2\text{SiO}_5\text{:Ce}$ (LYSO:Ce) [6,7] have been developed as promising scintillators for positron emission tomography (PET) due to their desirable properties such as high density, short decay time and high light output. LYSO:Ce has a density of 7.11 g/cm^3 and an emission spectrum at room temperature (RT) is peaked around 420 nm. LYSO:Ce exhibits a high light yield up to about 34,000 ph/MeV with poor energy resolution around 7.5 - 9.5% for 662 keV γ -rays [8].

Recently, Ce-doped LaCl_3 [9] and LaBr_3 [10] scintillators have been discovered with attractive properties due to high light output and very good energy resolution. $\text{LaCl}_3\text{:Ce}$ has a density of 3.79 g/cm^3 and an emission spectrum at RT is peaked around 350 nm. $\text{LaCl}_3\text{:Ce}$ exhibits a high light yield above 49,000 ph/MeV and very good energy resolution of about 3.2% for 662 keV γ -rays.

In view of high light output and very good energy resolution of $\text{LaCl}_3\text{:Ce}$ for γ -ray spectroscopy, while the attractive properties of LYSO:Ce are high light output and detection efficiency for γ -rays ($\rho Z_{\text{eff}}^{3-4}$) due to high density and effective atomic number (Z_{eff}). It seems, therefore, worthwhile to investigate and explore their capabilities for γ -ray detection.

In this paper, we present the comparative study on scintillation response of LYSO:Ce and $\text{LaCl}_3\text{:Ce}$ covering energies from 22.1 keV to 1274.5 keV using photomultiplier (PMT) readout. From the obtained data on photoelectron yield versus the energy of γ -rays and corresponding energy resolution, the light yield non-proportionality and the intrinsic energy resolution of both crystals were calculated. Special attention was devoted to the correlation between intrinsic resolution and non-proportional response of scintillators. Some aspects of photofraction will also be discussed.

2. Experimental procedures

The $\text{Lu}_{1.8}\text{Y}_{0.2}\text{SiO}_5\text{:Ce}$ crystal with size of $10 \times 10 \times 5 \text{ mm}^3$ and $\text{LaCl}_3\text{:Ce}$ crystal encapsulated in an aluminum can with size of $\varnothing 13 \times 13 \text{ mm}^2$ were supplied by Saint-Gobain (France). According to the manufacturer, the nominal cerium doped level is 10% for $\text{LaCl}_3\text{:Ce}$ and less than 1% for LYSO:Ce.

Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonic XP5200B PMT using silicone grease. In order to maximize light collection, the crystals were wrapped in a reflective, white Teflon tape on all sides (except the one coupled to the PMT). The signal from the PMT anode was passed to a CANBERRA2005 preamplifier and was sent 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 8k [11] was used to record energy spectra. Gaussian functions were fitted to full energy peaks using procedures in the analyzer to determine their positions and FWHMs. It included also the analysis of complex double peaks, characteristic of KX-rays and those exhibiting an escape peak.

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each γ -peak, was measured by Bertolaccini method [12, 13]. 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 photocathode, which determines the gain of PMT. The measurements of photoelectron yield and energy resolution were carried out for a series of γ -rays emitted by different radioactive sources in the energy range between 22.1 keV and 1274.5 keV, as listed in Table 1.

Table 1 The radioactive sources and γ -ray energies

Source	Energy of γ -rays (keV)
^{109}Cd	22.1 (K X-rays)
^{133}Ba	30.9 (K X-rays)
^{137}Cs	32.1 (K X-rays)
^{241}Am	59.5
^{133}Ba	81
^{57}Co	122
^{51}Cr	320.1
^{22}Na	511
^{137}Cs	661.6
^{54}Mn	834.9
^{22}Na	1274.5

3. Results and discussion

3.1 Energy spectra and light yield

Fig. 1 presents a comparison of the energy spectra of 662 keV γ -rays from a ^{137}Cs source measured with LYSO:Ce and $\text{LaCl}_3\text{:Ce}$ crystals under the same conditions. The energy resolution of 4.5% obtained with $\text{LaCl}_3\text{:Ce}$ is superior compared to the value of 8.2% obtained with LYSO:Ce. The obtained resolution for $\text{LaCl}_3\text{:Ce}$ is close to the value of 4.2% observed by van Loef et al. [14] and Balcerzyk et al. [15], respectively, for the $\varnothing 16\text{ mm} \times 19\text{ mm}$ crystal and the $\varnothing 25\text{ mm} \times 25\text{ mm}$ crystal, delivered by Saint Gobain. However,

all these results are poor compared to the value of about 3.2% reported for $\text{LaCl}_3\text{:Ce}$ with small size of $\varnothing 8\text{ mm} \times 5\text{ mm}$ [9] and $\sim 1\text{ cm}^3$ samples [16]. It could be associated with the lower quality and lower light output of the studied sample, see below. Note a higher photofraction in the spectrum measured with LYSO:Ce, as would be expected due to a higher effective atomic number and density of the LYSO:Ce crystal.

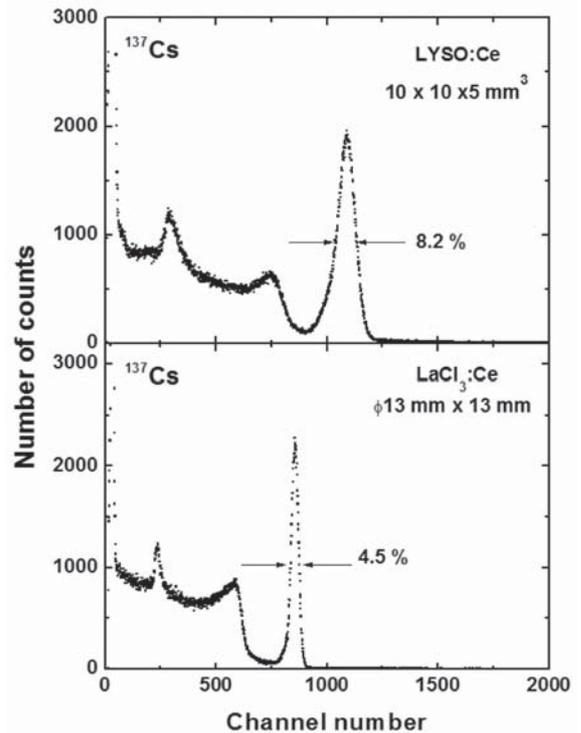


Fig. 1 Energy spectra of 662 keV γ -rays from a ^{137}Cs source measured with LYSO:Ce and $\text{LaCl}_3\text{:Ce}$ crystals under the same conditions.

Fig. 2 presents the energy spectra of γ -rays from a ^{22}Na source measured with LYSO:Ce and $\text{LaCl}_3\text{:Ce}$ crystals under the same conditions. Note a high energy resolution of 3.4% for the 1.274 MeV peak measured with $\text{LaCl}_3\text{:Ce}$. For LYSO:Ce, the obtained energy resolution is 6.1%.

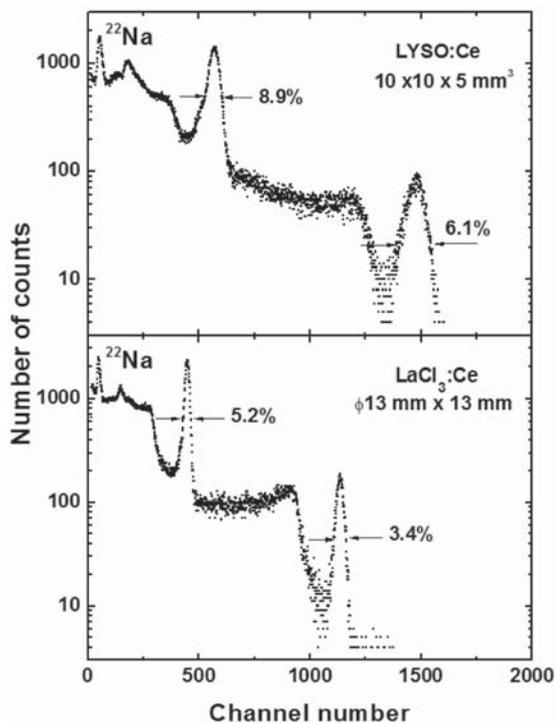


Fig. 2 Energy spectra of γ -rays from a ^{22}Na source measured with LYSO:Ce and $\text{LaCl}_3\text{:Ce}$ crystals under the same conditions.

The number of photoelectrons produced by the studied crystals in the XP5200B PMT was determined by relating the position of the full energy peak of 662 keV γ -rays to the position of the single photoelectron peak. Table 2 summarizes comparative measurements of photoelectron yield, light yield and energy resolution at 662 keV γ -rays for the studied crystals, as measured at 4 μs shaping time constant in the amplifier. The number of photoelectrons measured for both crystals was recalculated to the number of photons assuming the quantum efficiency of 26% and 27% for the XP5200B PMT, respectively at the peak emission of $\text{LaCl}_3\text{:Ce}$ (350 nm) and LYSO:Ce (420 nm).

Note a significantly lower light yield of 35,500 ph/MeV from the studied $\text{LaCl}_3\text{:Ce}$ crystal, by about 30% compared to that of 49,000 - 50,000 ph/MeV reported for small samples [9], [16]. The

low light output of the studied $\text{LaCl}_3\text{:Ce}$ is one of the important reasons for degradation in its energy resolution. The studied LYSO:Ce showed the light yield of 36,600 ph/MeV. This value is comparable to the value of 34,100 ph/MeV measured with 1 cm^3 sample [8].

Interestingly, despite a slightly higher photoelectron yield, LYSO:Ce showed much worse energy resolution compared with $\text{LaCl}_3\text{:Ce}$. It suggested looking at the non-proportionality of the light yield versus γ -ray energy.

Table 2 Photoelectron yield, light yield and energy resolution at 662 keV γ -rays for the studied crystals as measured with the XP5200B PMT

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	Energy resolution [%]
LYSO:Ce	$9,890 \pm 500$	$36,600 \pm 3000$	8.2 ± 0.4
$\text{LaCl}_3\text{:Ce}$	$9,230 \pm 400$	$35,500 \pm 3000$	4.5 ± 0.2

3.2 Non-proportionality of light yield

Light yield non-proportionality as a function of energy is one of the most important reasons for degradation in energy resolution of established scintillators [17]. The non-proportionality is defined here as the ratio of photoelectron yield measured at specific γ -ray energies relative to the photoelectron yield at the 662 keV γ -peak.

Fig.3 presents the non-proportionality characteristics of $\text{LaCl}_3\text{:Ce}$ and LYSO:Ce crystals. $\text{LaCl}_3\text{:Ce}$ is clearly superior to LYSO:Ce in terms of light yield proportionality. Over the energy range from 22.1 keV to 1274.5 keV, the non-proportionality is about 4% for $\text{LaCl}_3\text{:Ce}$, which is much better than that of about 35% for LYSO:Ce. The high proportionality of $\text{LaCl}_3\text{:Ce}$ is one of the important reasons behind its high-energy resolution. This result indicates that the non-proportionality is influenced by the host crystal properties, as the

doping agent is the same in both studied crystals. The different non-proportionality characteristics of the studied crystals should be reflected in their intrinsic resolutions, as it is known that the non-proportionality in the light yield is a fundamental limitation to the intrinsic energy resolution [17].

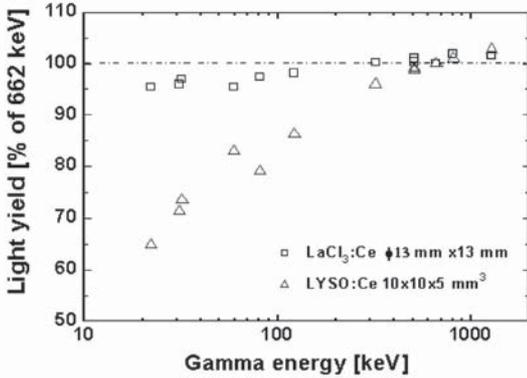


Fig. 3 Non-proportionality of the light yield as a function of γ -ray energy, measured with LYSO:Ce and LaCl₃:Ce crystals. Error bars are within the size of the points.

3.3 Energy resolution

The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a PMT can be written as [18]

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2, \quad (1)$$

where δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution and δ_{st} is the statistical contribution of PMT to the resolution.

The statistical uncertainty of the signal from the PMT can be described as

$$\delta_{st} = 2.355 \times 1/N^{1/2} \times (1 + \epsilon)^{1/2}, \quad (2)$$

where N is the number of the photoelectrons and ϵ is the variance of the electron multiplier gain, equal to 0.1 for an XP5500B PMT.

The transfer component depends on the quality of optical coupling of the crystal and PMT, homogeneity of quantum efficiency of the photo-

cathode 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 [18].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [17,18] and many effects such as inhomogeneities 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 δ_p is negligible, intrinsic resolution δ_{sc} of a crystal can be written as

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

Figs. 4 and 5 present the measured energy resolution versus energy of γ -rays for LaCl₃:Ce and LYSO:Ce crystals, respectively. Other curves shown in Figs.4 and 5 represent the PMT resolution calculated from the number of photoelectrons and the intrinsic resolution of the crystals calculated from Eq. (3). For LaCl₃:Ce crystal, the statistical contribution is slightly higher than the intrinsic resolution over the energy range from 50 keV to about 400 keV. At energies from 500 keV to 1274.5 keV, the statistical contribution and the intrinsic resolution are comparable. In contrast, the energy resolution for the LYSO:Ce crystal is mainly contributed by the intrinsic resolution over the whole range of energies.

Fig. 6 presents a direct comparison of the intrinsic resolution for the studied crystals. The intrinsic resolution at high energies is almost a factor of two better for LaCl₃:Ce, which reflects to a better proportionality of the light yield, see Fig. 3.

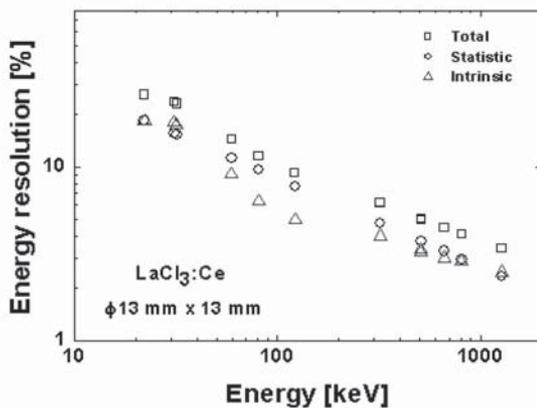


Fig. 4 Energy resolution and contributed factors versus energy of $\text{LaCl}_3:\text{Ce}$ crystal. Error bars are within the size of the points.

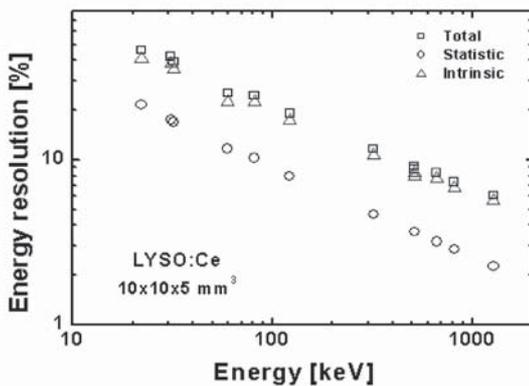


Fig. 5 Energy resolution and contributed factors versus energy of $\text{LYSO}:\text{Ce}$ crystal. Error bars are within the size of the points.

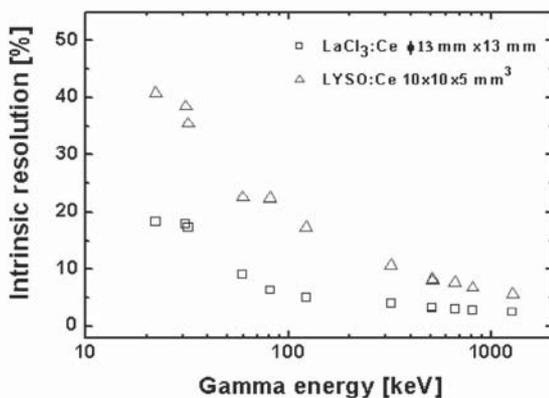


Fig. 6 Intrinsic resolution of $\text{LaCl}_3:\text{Ce}$ and $\text{LYSO}:\text{Ce}$ crystals versus energy of γ -rays. Error bars are within the size of the points.

To better understand the energy resolution of the studied crystals in γ -ray spectrometry, the contribution of various components to the overall energy resolution were analyzed for 662 keV photopeak, and the results are presented in Table 3. The second column gives N , the number of photoelectrons produced in the PMT. The third column gives $\Delta E/E$, the overall energy resolution at 662 keV photopeak. The PMT contribution (δ_{st}) was calculated using Eq.(2). From the values of $\Delta E/E$ and δ_{st} , the intrinsic resolution (δ_{sc}) was calculated using Eq.(3). The energy resolution of $\text{LYSO}:\text{Ce}$ is worse than that of $\text{LaCl}_3:\text{Ce}$ in spite of a comparable contribution of δ_{st} . The reason is a much higher contribution of δ_{sc} , related to its higher non-proportionality. This result confirms that the intrinsic resolution of a scintillator is mainly associated with the non-proportional light response [17,18].

Table 3 Analysis of the 662 keV energy resolution for $\text{LaCl}_3:\text{Ce}$ and $\text{LYSO}:\text{Ce}$ crystals

Detector	N [electrons]	$\Delta E/E$ [%]	δ_{st} [%]	δ_{sc} [%]
$\text{LaCl}_3:\text{Ce}$	6,110	4.5	3.3	3.0
$\text{LYSO}:\text{Ce}$	6,550	8.2	3.2	7.7

3.4 Photofraction

The photofraction is defined here as the ratio of counts under the photopeak to the total counts of the spectrum as measured at a specific γ -ray energy. The photofraction for $\text{LYSO}:\text{Ce}$ and $\text{LaCl}_3:\text{Ce}$ at 320 and 662 keV γ -peaks is collected in Table 4. For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one calculated using the WinXCom program [19] are given. The data indicate that $\text{LYSO}:\text{Ce}$ shows much higher photofraction than $\text{LaCl}_3:\text{Ce}$ for both γ -peaks in a same trend with the cross-section ratio (σ -ratio)

obtained from the WinXCom program. The reason is due to much higher effective atomic number and density of the LYSO:Ce crystal. However, the σ -ratio is closer to the measured photofraction for LYSO:Ce than for LaCl₃:Ce. It may be due to a smaller size (a factor of 2.5) of LYSO:Ce sample.

Table 4 Photofraction for LaCl₃:Ce and LYSO:Ce crystals

γ energy (keV) Source	320 ⁵¹ Cr	662 ¹³⁷ Cs	
Photof. (%)	38.8	15.7	LaCl ₃ :Ce
σ ratio (%)	33.8	9.8	
Photof. (%)	60.0	26.1	LYSO:Ce
σ ratio (%)	56.6	22.6	

4. Conclusions

The properties of LYSO:Ce and LaCl₃:Ce scintillators in γ -ray spectrometry are summarized in Table 5. The main advantage of LaCl₃:Ce is its superior energy resolution over the whole energy range from 22.1 keV to 1274.5 keV. The reason is very small contribution of the intrinsic resolution, reflected by its very good proportionality of the light yield. However, the energy resolution of 4.5% at 662 keV for this LaCl₃:Ce sample is worse than that of 3.2% for the small samples, due to much lower light yield by about 30% with respect to the small samples. A further improvement of crystal quality as well as a better encasement of crystal might improve the energy resolution and make it a good candidate to replace NaI:Tl ($\Delta E/E = 6.5\%$, $\rho = 3.67$ g/cm³, $Z_{\text{eff}} = 50$) as the scintillator of choice for SPECT camera and γ -ray spectroscopy.

Despite a slightly higher photoelectron yield, LYSO:Ce showed much worse energy resolution compared with LaCl₃:Ce. The reason is much higher contribution of intrinsic resolution for LYSO:Ce, reflected by a high non-proportionality of 35% at 22 keV for LYSO:Ce with respect to only 4% for

Table 5 Comparison of properties of LaCl₃:Ce and LYSO:Ce crystals

Property	LaCl ₃ :Ce	LYSO:Ce
Light yield	35,500	36,600
	ph/MeV	ph/MeV
Energy resolution at 662 keV	4.5 \pm 0.2%	8.2 \pm 0.4%
Proportionality	- 4% at 22 keV	- 35% at 22 keV
Density	3.79 g/cm ³	7.11 g/cm ³
Z_{eff}	49.5	65
Photofraction at 662 keV	15.7%	26.1%

LaCl₃:Ce. Our study confirms that the intrinsic resolution of the scintillator is strongly correlated with the non-proportionality in the scintillation response.

The main advantages of LYSO:Ce are high light yield and detection efficiency which make it very promising scintillator for PET medical imaging.

5. Acknowledgements

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