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Light Yield Non-Proportionality and Energy Resolution of $Lu_{1.8}Y_{0.2}SiO_5$:Ce and $LaCl_3$:Ce Scintillation Crystals

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Abstract. The scintillation response of $Lu_{1.8}Y_{0.2}SiO_5$:Ce (LYSO:Ce) and $LaCl_3$: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±0.2% obtained for $LaCl_3$:Ce coupled to XP5200B PMT is much better than that of 8.2±0.4% for LYSO: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.

Introduction

Inorganic scintillators play an important role in detection of energetic photons and nuclear particles. 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].

Lu_2SiO_5:Ce (LSO:Ce) [5] and $(Lu,Y)_2SiO_5$: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 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].

New 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. $LaCl_3$: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 this paper, we present the comparative study on scintillation response of LYSO:Ce and $LaCl_3$: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.

Methodology

The $Lu_{1.8}Y_{0.2}SiO_5$:Ce crystal with size of 10×10×5 mm^3 and $LaCl_3$:Ce crystal encapsulated in an aluminum can with size of ∅13×13 mm^2 were supplied by Saint-Gobain (France). According to the manufacturer, the nominal cerium doped level is 10% for $LaCl_3$:Ce and less than 1% for LYSO:Ce. Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonis XP5200B PMT using silicone grease. The signal from the PMT anode was passed to a CANBERRA 2005 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.

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.

Results and Discussion

Light Yield and Energy Resolution. Fig. 1(a) presents the energy spectra of 662 keV γ-rays from a ^{137}Cs source measured with LYSO:Ce and $LaCl_3$:Ce crystals under the same conditions. The energy resolution of 4.5% obtained with $LaCl_3$:Ce is superior compared to the value of 8.2% obtained with LYSO:Ce. The obtained resolution for $LaCl_3$:Ce is close to the value of 4.2% observed by van Loef et al. [14] and Balcerzyk et al. [15], respectively, for the ∅16 mm × 19 mm crystal and the ∅25 mm × 25 mm crystal, delivered by Saint Gobain. Note a higher photofraction in the spectrum measured with LYSO:Ce, as would be expected due to a higher effective atomic number (65 vs 49.8) and density (7.11 vs 3.79 g/cm^3) of the LYSO:Ce crystal.

Fig. 1(b) presents the energy spectra of γ-rays from a ^{22}Na source. Note a high energy resolution of 3.4% for the 1.274 MeV peak measured with $LaCl_3$:Ce. For LYSO:Ce, the obtained energy resolution is 6.1%.

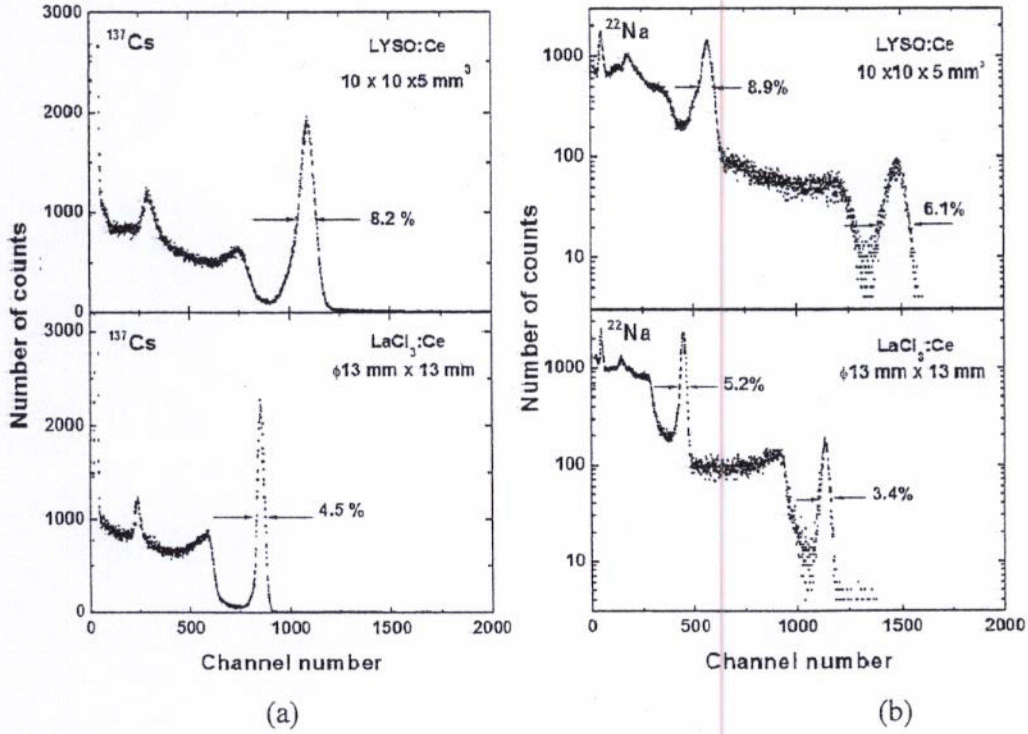

Fig. 1 Energy spectra of (a) 662 keV γ - rays from a ^{137}Cs source and (b) γ - rays from a ^{22}Na source measured with LYSO:Ce and $LaCl_3$:Ce crystals under the same conditions.

Table 1 summarizes comparative measurements of photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals. 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 $LaCl_3$:Ce (350 nm) and LYSO:Ce (420 nm).

Note a significantly lower light yield of 35,500 ph/MeV from the studied $LaCl_3$:Ce crystal, by about 30% compared to that of 49,000 – 50,000 ph/MeV reported for small samples [9,16]. 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 $LaCl_3$:Ce. It suggested looking at the non-proportionality of the light yield versus γ-ray energy.

Table 1 Photoelectron yield, light yield and energy resolution for the studied crystals.

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	Energy resolution [%]
LYSO:Ce	9,890 ± 500	36,600 ± 3000	8.2 ± 0.4
$LaCl_3$:Ce	9,230 ± 400	35,500 ± 3000	4.5 ± 0.2

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.2 presents the non-proportionality characteristics of $LaCl_3$:Ce and LYSO:Ce crystals. $LaCl_3$: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 $LaCl_3$:Ce, which is much better than that of about 35% for LYSO:Ce. The high proportionality of $LaCl_3$:Ce is one of the important reasons behind its high-energy resolution. 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].

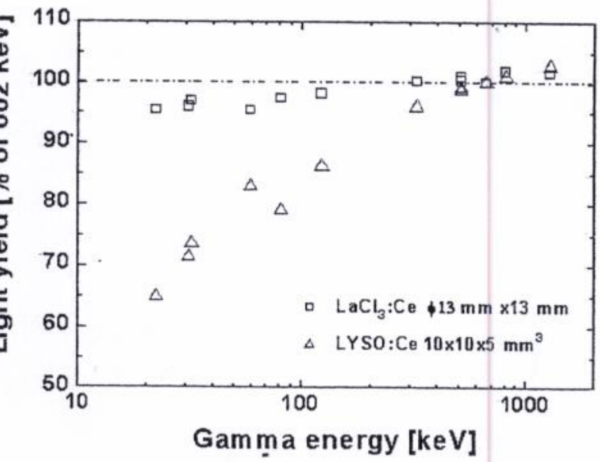

Fig. 2 Non-proportionality of the light yield as a function of γ-ray energy, measured with LYSO:Ce and $LaCl_3$:Ce crystals. Error bars are within the size of the points.

Intrinsic Resolution. The energy resolution (ΔE/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

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

where N is the number of the photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for an XP5200B PMT.

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 follows

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

Fig. 3 present the measured energy resolution versus energy of γ-rays for $LaCl_3$:Ce and LYSO:Ce crystals, respectively. Other curves shown in Fig. 3 represent the PMT resolution calculated from the number of photoelectrons and the intrinsic resolution of the crystals calculated from Eq. (3). For $LaCl_3$: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. 4 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 $LaCl_3$:Ce, which reflects to a better proportionality of the light yield, see Fig. 2.

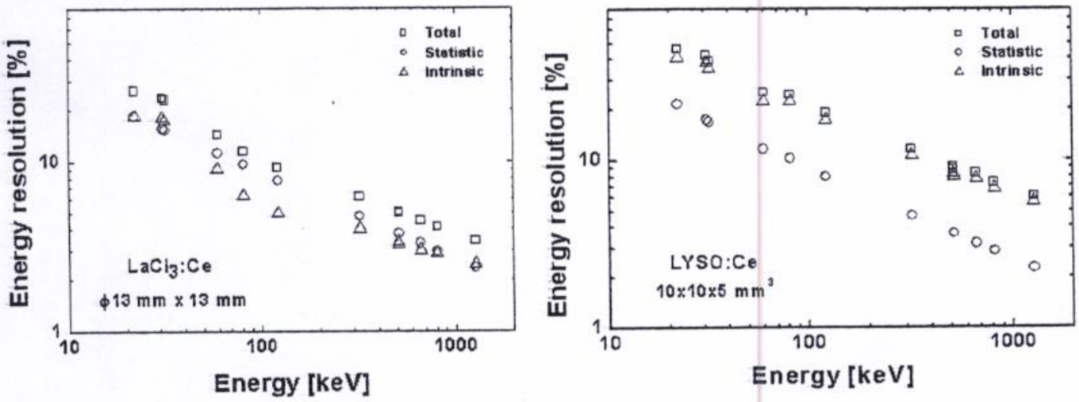

Fig.3 Energy resolution and contributed factors versus energy of $LaCl_3$:Ce and LYSO:Ce crystals. Error bars are within the size of the points.

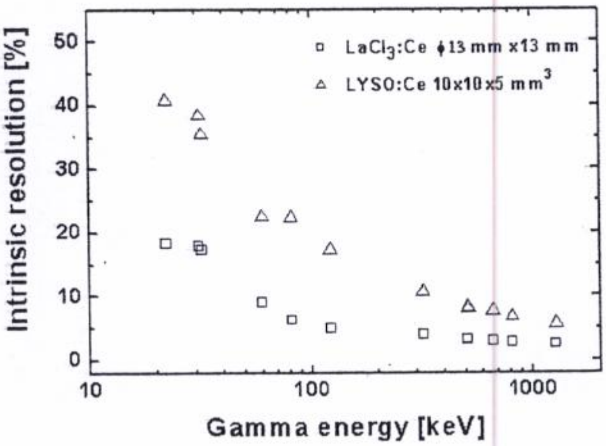

Fig. 4 Intrinsic resolution of $LaCl_3$:Ce and LYSO:Ce crystals versus energy of γ-rays. Error bars are within the size of the points.

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 2. The energy resolution of LYSO:Ce is worse than that of $LaCl_3$: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 of the light yield [17,18].

Table 2 Analysis of the 662 keV energy resolution for $LaCl_3$:Ce and LYSO:Ce crystals

Detector	N [electrons]	ΔE/E [%]	δ_{st} [%]	δ_{sc} [%]
$LaCl_3$:Ce	6110	4.5	3.3	3.0
LYSO:Ce	6550	8.2	3.2	7.7

Summary

The main advantage of $LaCl_3$: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 $LaCl_3$: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/cm^3, $Z_{eff} = 50$) 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 $LaCl_3$: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 $LaCl_3$: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.

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