ความไม่เป็นสัดส่วนของยีลด์แสงและการแยกชัดพลังงานสำหรับผลึก $LaCl_3$(10%Ce) และ CsI(Na)

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บทคัดย่อ

งานวิจัยนี้ได้ทำการศึกษาการตอบสนองด้านซินทิลเลชันของผลึก $LaCl_3$(10%Ce) ขนาด ∅13 มม. ×13 มม. และ CsI(Na) ขนาด ∅10 มม. ×10 มม. สำหรับรังสีแกมมาในช่วงพลังงาน 22.1 keV ถึง 1,274.5 keV จากการศึกษาความไม่เป็นสัดส่วนของยีลด์แสง (non-proportionality of light yield) และการแยกชัดพลังงาน (energy resolution) ของผลึกทั้งสองชนิด ด้วยหลอดทวีคูณแสง Photonis XP 5200 พบว่าการแยกชัดพลังงานรังสีแกมมา 661.6 keV จาก ^{137}Cs มีค่า 4.6 ± 0.2% และ 6.5 ± 0.3% สำหรับ $LaCl_3$(10%Ce) และ CsI(Na) ตามลำดับ นอกจากนี้ยังพบว่าการแยกชัดพลังงานของ $LaCl_3$(10%Ce) มีความสัมพันธ์เชิงเส้นโดยประมาณกับค่าผกผันของรากที่สองของพลังงาน ในขณะที่การแยกชัดพลังงานของ CsI(Na) มีลักษณะกึ่งราบ (semi-plateau) ในช่วงพลังงาน 100 keV ถึง 300 keV $LaCl_3$(10%Ce) แสดงความไม่เป็นสัดส่วนของยีลด์แสงประมาณร้อยละ 4 เมื่อลดพลังงานจาก 1,274.5 keV ถึง 22.1 keV ซึ่งดีกว่า CsI(Na) ที่แสดงความไม่เป็นสัดส่วนประมาณร้อย 17 บทความนี้ได้อภิปรายถึงการแยกชัดพลังงานรวมและการแยกชัดในตัวของผลึกที่ศึกษาในแต่ละช่วงพลังงาน

คำสำคัญ : ซินทิลเลเตอร์ / $LaCl_3$(10%Ce) / CsI(Na) / ความไม่เป็นสัดส่วนของยีลด์แสง / การแยกชัดพลังงาน / การแยกชัดในตัว

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Light Yield Non-proportionality and Energy Resolution of $LaCl_3$(10%Ce) and CsI(Na) Crystals

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Abstract

The scintillation response of ∅13 mm × 13 mm $LaCl_3$(10%Ce) and ∅10 mm × 10 mm CsI(Na) crystals were studied for γ-ray energies ranging from 22.1 keV to 1,274.5 keV. The light yield non-proportionality and energy resolution were measured with a Photonis XP5200 PMT. The energy resolution, obtained in this work, for 661.6 keV peak from ^{137}Cs, are 4.6 ± 0.2 % and 6.5 ± 0.3 %, respectively, for $LaCl_3$(10%Ce) and CsI(Na). $LaCl_3$(10%Ce) showed approximately a linear relationship between energy resolution and the inverse square root of the energy, while the step-like curve with a semi - plateau in the energy range between 100 keV and 300 keV was observed for CsI(Na). The $LaCl_3$(10%Ce) showed a light yield non – proportionality of about 4% upon lowering energy from 1,274.5 keV to 22.1 keV, which is better than that of about 17% obtained for CsI(Na). The total and intrinsic energy resolutions are discussed.

Keywords : Scintillator / $LaCl_3$(10%Ce) / CsI(Na) / Non-proportionality of Light Yield / Energy Resolution / Intrinsic Resolution

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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 yield, fast response time, high stopping power, good energy resolution, good proportionality of light yield, minimal afterglow and low production costs. Good reviews on development of inorganic-scintillators and development of scintillation detectors for γ-ray spectrometry have been published by van Eijk [1], Moszynski [2], and recently by Lecoq et al. [3].

The phenomenon of non-proportionality response and its relation with energy resolution have been studied for many classical scintillators, especially for NaI(Tl), CsI(Tl) and CsI(Na) [4-9]. The light yield, expressed in photons per MeV (ph/MeV) of absorbed γ energy, for NaI(Tl) and most other alkali halides decreases as the energy of γ-rays increases. Contrary to alkali halides, oxide based scintillators in general show an increasing light yield with increasing energy of γ-rays, which levels at higher energies [10-14]. NaI(Tl) and CsI(Tl) crystals provide high light yield with rather poor energy resolution of about 6% to 7% (FWHM) at 661.6 keV γ-rays. Non-proportionality in light yield can be one of the important reasons for degradation in energy resolution of these scintillators [13].

Recently, a cerium-doped lanthanum chloride–$\mathrm{LaCl_3}$(Ce) with attractive scintillation properties has been discovered [15]. $\mathrm{LaCl_3}$(Ce) has an emission peak at 350 nm, a density of 3.79 g/cm^3 and is hygroscopic. $\mathrm{LaCl_3}$(10%Ce) has high light yield output (49,000 ph/MeV), high energy resolution (3.1% for 662 keV),and fast principal decay time (25.5 ns) [16]. Therefore, these scintillation properties make $\mathrm{LaCl_3}$(Ce) a very promising and is considered to be a viable alternative to traditional halide scintillators for gamma ray spectroscopy and nuclear medical imaging applications.

In this paper, we present the comparative study on scintillation response of $\mathrm{LaCl_3}$(10%Ce) and CsI(Na) covering energies from 22.1 keV to 1,274.5 keV. 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 are determined.

2. Experimental procedures

A $\mathrm{LaCl_3}$(10%Ce) crystal used in this study was supplied by Saint Gobain, with the dimension of ∅13 mm x 13 mm. A CsI(Na) crystal from the same manufacturer with the dimension of ∅10 mm x 10 mm was used for comparison. Both crystals were assembled by the manufacturer in the aluminum cases with a front glass window. Each crystal was optically coupled to a ∅52 mm Photonis XP5200 photomultiplier tube (PMT) using silicone grease.

All measurements were carried out using standard NIM level electronics. The sources were positioned along the cylindrical axis of the scintillator and the PMT. The signal from the PMT was passed to a scintillation preamplifier and then to a spectroscopy amplifier. A shaping time constant of 3 μs was used with $\mathrm{LaCl_3}$(10%Ce) crystal and 4 ms was used with CsI(Na) crystal. The energy spectra were recorded using a Tukan PC-based multichannel analyzer.

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 1,274.5 keV. For isolated peaks, the full width at half maximum (FWHM) and centroid of the full energy peak were obtained by fitting with a single Gaussian function. Some peaks were fitted with two Gaussian functions in order to better separate the peak of interest from other partly overlapping peaks.

3. Results and discussion

3.1 Photoelectron yield and energy resolution

Fig.1 presents the energy spectra of 661.6 keV γ-rays from a ^{137}Cs source measured with LaCl$_3$(10%Ce) and CsI(Na) detectors. It is seen that LaCl$_3$(10%Ce) gives better energy resolution than CsI(Na). The energy resolution of 4.6 ± 0.2% obtained with LaCl$_3$(10%Ce) is much better than the value of 6.5 ±0.3% obtained with CsI(Na). The poorer energy resolution for LaCl$_3$(10%Ce) in this study than that reported in Refs. [15-17] could be associated with its lower light yield, see below. The obtained resolution for LaCl$_3$(10%Ce) is close to the value of 4.2% observed by van Loef et al. [18] and Balcerzyk et al. [19], respectively, for the ∅16 mm × 19 mm crystal and the ∅25 mm × 25 mm crystal, supplied by Saint Gobain. These results indicate that the energy resolution of LaCl$_3$(10%Ce) is not strongly influenced by the crystal size.

The energy resolution of 6.5% for the tested CsI(Na) crystal in this study is much better than the value of 9.9% observed by Chewpraditkul et al. [20] for an equal sized CsI(Na) crystal supplied by CRYOS.Beta, Ukraine. The superior energy resolution of the tested sample could be attributed to a much larger photoelectron yield together with its better proportionality of the light yield, see below.

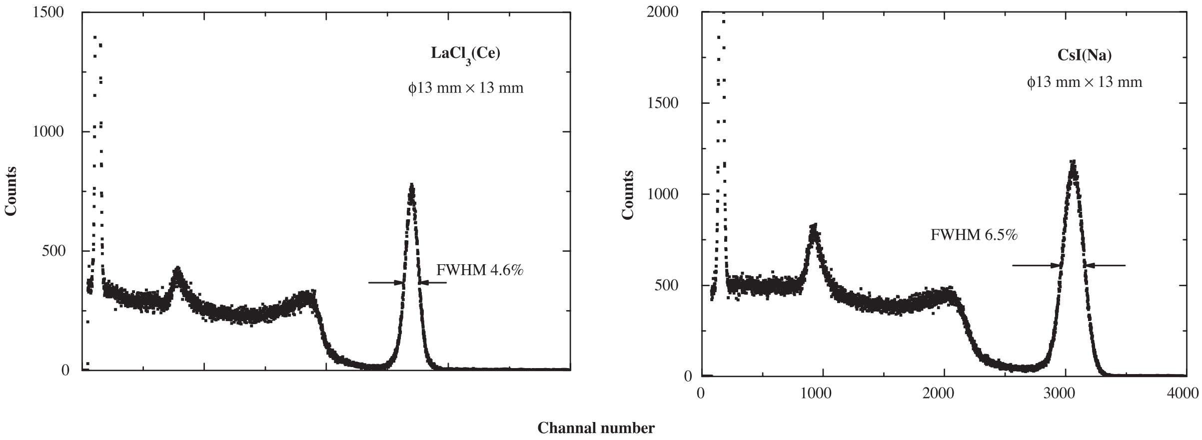

Fig. 1 Energy spectra of 661.6 keV γ-rays from a ^{137}Cs source, as measured with LaCl$_3$(10%Ce) and CsI(Na) detectors.

Fig. 2 presents a similar set of spectra measured with 59.5 keV γ-rays from an ^{241}Am source. The energy resolution is 14.6% for LaCl$_3$(10%Ce) and 12.6% for CsI(Na). Note the better energy resolution of CsI(Na) for detection of low energy γ-rays, whereas the energy resolution of LaCl$_3$(10%Ce) is much better than that of CsI(Na) for detection of high energy γ-rays, see below.

The photoelectron yield, expressed in number of photoelectrons per MeV (phe/MeV) for each γ-peak, was measured by the Bertolaccini et al. method [21-22]. In this method the number of photoelectrons is measured directly by comparing the position of the full energy peak of γ-rays detected in the crystal with that of the single photoelectron peak from the photocathode, which determines the gain of PMT. By removing the source and increasing the amplifier gain, the single photoelectron spectrum was recorded without the crystal.

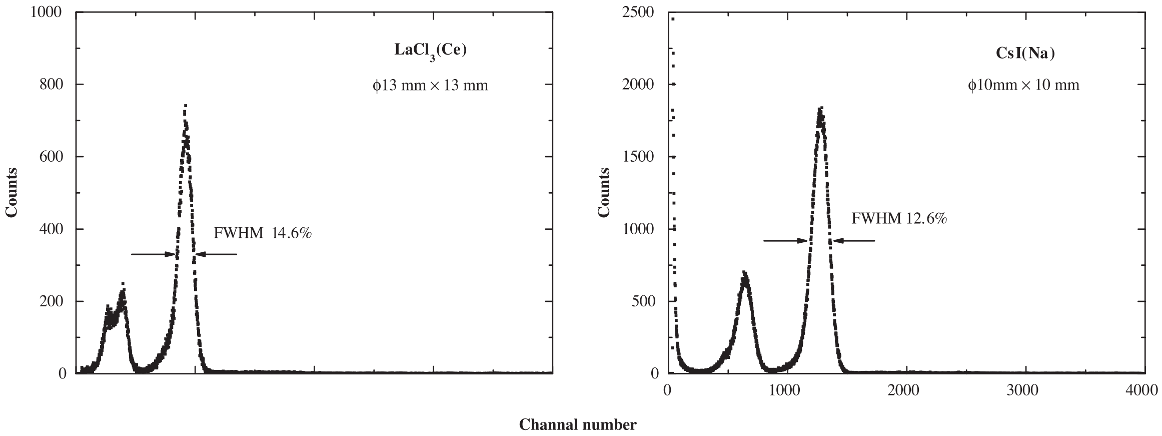

Fig. 2 Energy spectra of 59.5 keV γ-rays from a ^{241}Am source, as measured with $LaCl_3$(10%Ce) and CsI(Na) detectors.

In the measurements with the Photonis XP5200 PMT, the $LaCl_3$(10%Ce) showed a photoelectron yield of 8,300 ± 300 phe/MeV at 661.6 keV and 3 ms shaping time constant. This value corresponds to about 32,000±2,200 ph/MeV at the PMT photocathode quantum efficiency (QE) of 26% for peak emission of 350 nm. Note a significantly lower light yield of the tested $LaCl_3$(10%Ce) crystal, by about 35%, compared to those quoted for small samples in Refs.[15-17], and by about 11% compared with a ∅ 25 mm x 25 mm sample in Ref [19]. The tested CsI(Na) showed a photoelectron yield of 10800 ±400 phe/MeV at 4 μs shaping time constant. This value corresponds to about 41,500 ±2,900 ph/MeV at a QE of 26% for peak emission of 420 nm. This value is close to the advertised value of 41,000 ph/MeV. Note a significantly higher photoelectron yield of the tested CsI(Na) crystal, by about 40%, compared with an equal sized sample in Ref [20].

3.2 Non-proportionality of 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 [13]. The non-proportionality is defined here as the ratio of photoelectron yield measured for photopeaks at specific γ-ray energy relative to the yield at 661.6 keV γ-peak.

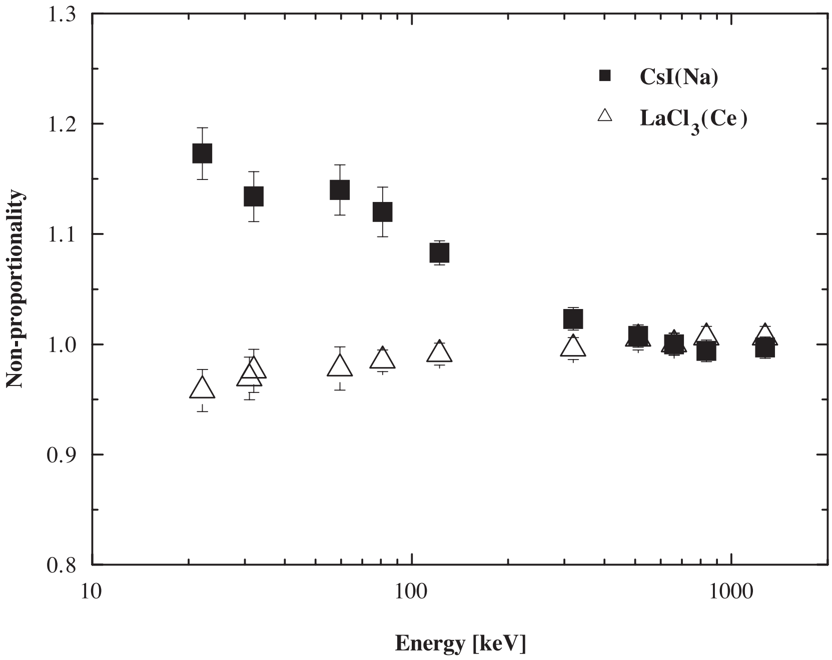

Fig. 3 Non-proportionality in the light yield of $LaCl_3$(10%Ce) and CsI(Na) crystals.

Fig. 3 presents the non-proportionality characteristics of $LaCl_3$(10%Ce) and CsI(Na) crystals. Both crystals exhibit different non-proportionality curves. $LaCl_3$(10%Ce) is clearly superior to CsI(Na) in terms of light yield proportionality. Over the energy range from 22.1 keV to 1,274.5 keV, the non-proportionality is about 4% for $LaCl_3$(10%Ce), which is much better than that of about 17% for CsI(Na). The higher proportionality of $LaCl_3$(10%Ce) is one of the important reasons behind its high-energy resolution.

Shah et al. [17] measured proportional response of $LaCl_3$(10%Ce) crystal (~1 cm^3 in size) under excitation with five γ-ray energies. They reported the non-proportionality of about 7% in the energy range from 60 keV to 1,275 keV. van Loef et al. [18] measured proportional response of an aluminum canned $LaCl_3$ (10%Ce) crystal (∅16 mm × 19 mm) and observed the flatness of its response within 5% in the energy range from 30 keV to 1,275 keV. Balcerzyk et al. [19] measured proportional response of an aluminum canned $LaCl_3$(10%Ce) with the size of ∅25 mm × 25 mm. They observed the non-proportionality of about 5% over the energy range from 17 to 1,275 keV. These results indicate that the crystal size does not have any influence on the light yield non-proportionality of $LaCl_3$(10%Ce) crystal.

The proportional response for tested CsI(Na) shows a different behavior from $LaCl_3$(10%Ce). Over the energy range from 22.1 keV to 1,274.5 keV, the non-proportionality in its light yield is about 17%. This value is much better than that of about 24%, over the same energy range, for an equal sized CsI(Na) crystal in our previous work [20]. This together with its much larger photoelectron yield are the main reasons for the superior energy resolution of the tesed CsI(Na) crystal supplied by Saint Gobain. Aitken et al. measured proportional response for a thin CsI(Na) crystal (∅ 10 mm x 0.8

mm) and observed the non-proportionality of about 28% in the energy range from 22 keV to 1 MeV [5]. Mengesha et al. also measured proportional response for a ∅ 10 mm x 20 mm CsI(Na) crystal [6]. They reported the non-proportionality of about 19% over the same energy range. These results indicate that crystal size does not seem to be relevant parameter in non-proportionality response of CsI(Na) scintillators. A better proportionality response of the tested CsI(Na) sample can be attributed to the better quality of the new CsI(Na) crystal used in this study.

3.3 Energy resolution

The energy resolution, ΔE/E, of the full-energy peak measured with a scintillator coupled to a photomultiplier tube can be written as [8]:

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

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

The statistical uncertainty of the signal from the PMT is described as:

$$\delta_{st} = 2.35 \text{ x } (1/N)^{1/2} \text{ x } (1+\varepsilon)^{1/2}, \qquad \textbf{(2)}$$

where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for the Photonis XP5200 PMT.

The transfer component is described by the variance associated with the probability that a photon from the scintillator results in the arrival of photoelectron at the first dynode. The transfer component depends on the quality of the optical coupling of the crystal and PMT, homogeneity of the quantum efficiency of the photocathode and efficiency of photoelectron collection at the first dynode. This component is negligible compared to the other components of the energy resolution in the modern scintillation detectors [8].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [4, 8, 13] and many effects such as inhomogeneity of the scintillator which can cause local variations in the scintillation light output and non-uniform reflectivity of the reflecting cover of the crystal.

The overall energy resolution and the PMT resolution can be determined experimentally. If δ_p is negligible, the intrinsic resolution of a crystal can be written as follows:

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

Figs. 4 and 5 present the measured energy resolution versus energy of γ-rays for CsI(Na) and $LaCl_3$(10%Ce) detectors. $LaCl_3$(10%Ce) exhibits approximately a linear relationship between resolution and the inverse square root of the energy as reported recently in Refs [18-19]. CsI(Na) exhibits step-like curve with a semi-plateau in the energy range between 100 keV and 300 keV. Similar pattern of the step-like curves was also observed in Ref. [23] for the pure CsI crystals. 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.

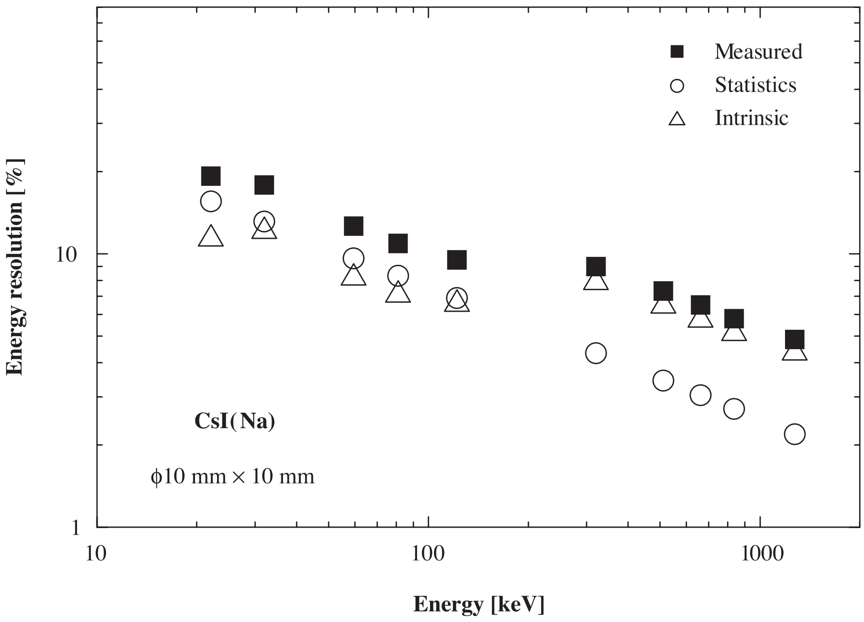

Fig. 4 Energy resolution and contributed factors versus energy of CsI(Na) crystal. Error bars are within the size of the points.

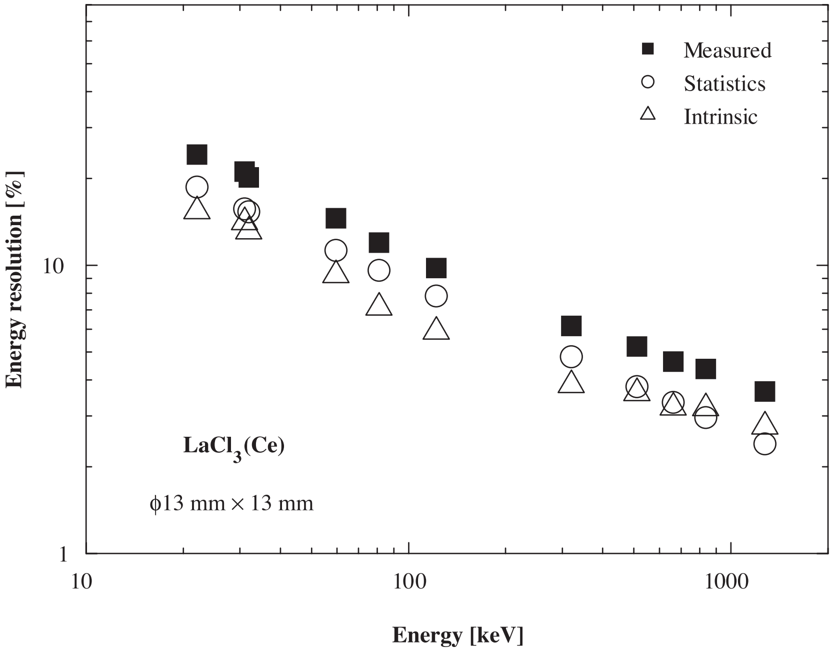

Fig. 5 Energy resolution and contributed factors versus energy of $LaCl_3$(10%Ce) crystal. Error bars are within the size of the points.

The overall energy resolution and the intrinsic resolution for the studied crystals are presented in Figs. 6 and 7, respectively. At energies below 100 keV, both crystals exhibit a comparable intrinsic resolution. The photoelectron yield of CsI(Na) around 60 keV is 12,000 phe/MeV which is significantly larger than the yield of 8,100 phe/MeV for $LaCl_3$(10%Ce). These are the main reasons for a slightly better energy resolution of CsI(Na) detector below 100 keV. However, despite a larger photoelectron yield, the energy resolution of CsI(Na) detector significantly degrades as compared with the $LaCl_3$(10%Ce) detector at energy above 200 keV. The reason is a much higher contribution from its intrinsic resolution.

At energies above 200 keV, the intrinsic resolution curve for CsI(Na) is characterized by a bell-shaped contour mainly due to the contribution from the non-proportionality of light yield to the full energy peak. For $LaCl_3$(10%Ce) we did not observe such a contribution, reflecting its very good proportional response between 22 keV and 1,274.5 keV.

Table 1 summarizes the data relevant to the energy resolution of the 661.6 keV photopeak, performed for CsI(Na) and $LaCl_3$(10%Ce) scintillators in this study. The second column gives N_{phe}, the photoelectron yield in phe/MeV produced in the PMT. The third column gives $\Delta E/E$, the overall energy resolution of the 661.6 keV photopeak. From the number of photoelectrons (N), the PMT contribution δ_{st} is calculated using (2). From the values of $\Delta E/E$ and δ_{st}, the intrinsic resolution δ_{sc} is calculated using (3).

Table 1 Energy resolution data at 661.6 keV γ-rays for CsI(Na) and $LaCl_3$(10%Ce) measured with the Photonis XP5200 PMT.

Crystal	N_{phe} (phe/MeV)	$\Delta E/E$ [%]	δ_{st} [%]	δ_{sc} [%]
CsI(Na)	10,800	6.5	3.0	5.7
$LaCl_3$(10%Ce)	8,300	4.6	3.3	3.2

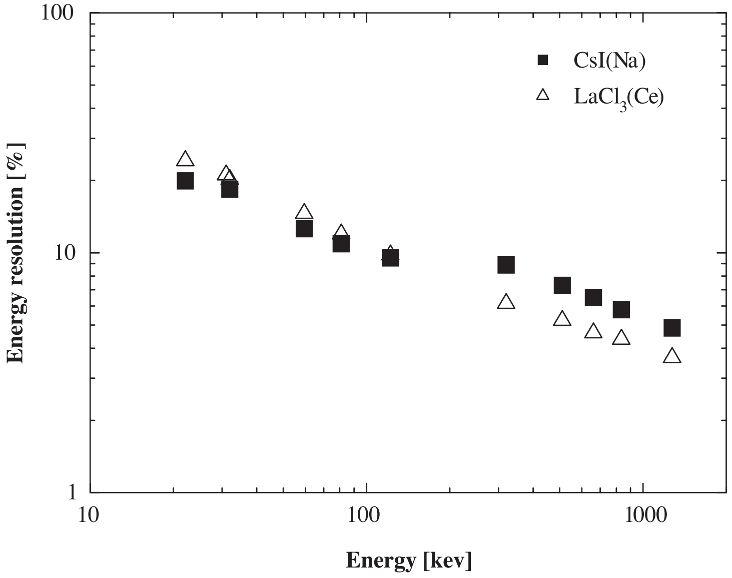

Fig. 6 Overall energy resolution of $LaCl_3$(10%Ce) and CsI(Na) detectors. Error bars are within the size of the points.

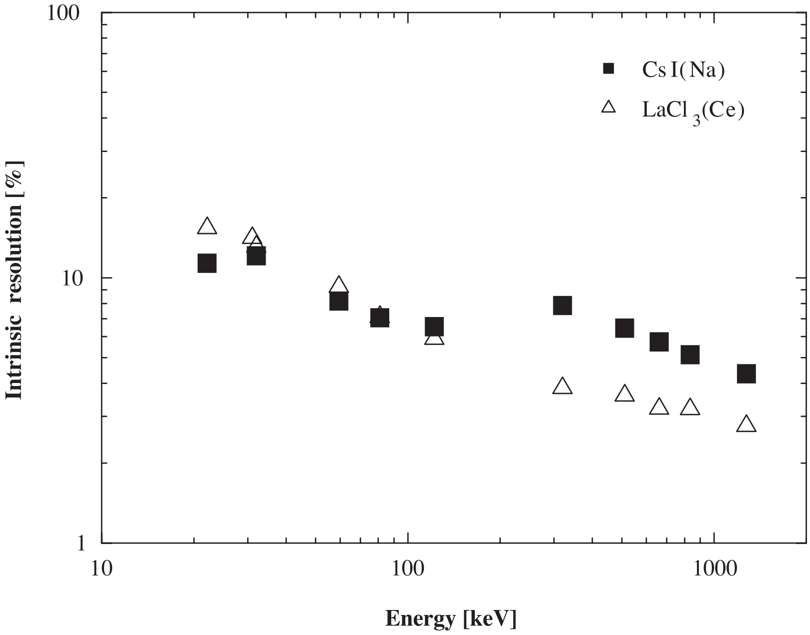

Fig. 7 Intrinsic resolution of $LaCl_3$(10%Ce) and CsI(Na) crystals. Error bars are within the size of the points.

Moszynski et al. [8] have performed a comparison of the intrinsic resolution with the non-proportionality component (δ_{np}) from primary electrons (photoelectrons, Compton electrons and Auger electrons) for a NaI(Tl) scintillator coupled to a PMT. The δ_{sc} at 661.6 keV energy was measured to be 5.8%, while δ_{np} was found to be about 2.6%. Consequently, the non–proportionality contribution from secondary electrons, namely δ-rays (δ_{δ}) was obtained to be about 5.2% from $[\delta^2_{sc} - \delta^2_{np}]^{1/2}$ by assuming that the δ_{sc} is weakly affected by the contribution of crystal inhomogeinity (δ_{inh}) and δ_p is negligible for PMT readout. The estimated value indicates that the contribution from δ-rays (δ_{δ}) is a major component in the NaI(Tl) intrinsic resolution. As shown in Table 1, intrinsic resolution δ_{sc} was measured to be 5.7% and 3.2%, respectively, for CsI(Na) and $LaCl_3$(10%Ce). The non-proportionality of light yield was measured to be 17% and 4%, respectively, for CsI(Na) and $LaCl_3$(10%Ce) while a typical value of about 15% was observed for NaI(Tl). It confirms further that the intrinsic resolution is strongly correlated with the light yield non–proportionality [8, 23]. For $LaCl_3$(10%Ce), since the non-proportionality of light yield is only about 4% in the energy range from 22 keV to 1,274.5 keV, the contribution of δ_{np} to the intrinsic resolution δ_{sc} is probably negligible. Consequently, we expect that for $LaCl_3$(10%Ce) , the δ_{sc} is mainly contributed by δ_{δ}. This study supports the conclusion of [8] that the scattering of secondary electrons in the crystal, namely δ-rays, mainly creates the intrinsic resolution.

4. Conclusions

In this work, the scintillation response of $LaCl_3$(10%Ce) and CsI(Na) crystals were studied and compared in γ-ray spectrometry. Between 22.1

keV and 1,274.5 keV the energy resolution for $LaCl_3$(10%Ce) decreases with the inverse square root of the gamma ray energy whereas for CsI(Na), the energy resolution shows the step-like curve with a semi-plateau in the energy range between 100 and 300 keV. $LaCl_3$(10%Ce) is a very proportional scintillator. Over this energy range the non-proportionality in its light yield is about 4% which is better than that of about 17% for CsI(Na) in this study.

At energies above 200 keV, the energy resolution of $LaCl_3$(10%Ce) is much better than that of CsI(Na) due to small contribution from its intrinsic resolution, reflecting a very good proportionality of light yield between 22.1 keV and 1,274.5 keV. However, below 100 keV the energy resolution of CsI(Na) is slightly better than that of $LaCl_3$(10%Ce) due to a larger photoelectron yield (by almost about 50%) measured for CsI(Na). This study demonstrates that the contribution from δ-rays is a major component of the intrinsic resolution.

In conclusions, the main advantages of $LaCl_3$(10%Ce) are good energy resolution at energies above 200 keV, good proportionality of light yield, and short decay time. These properties make $LaCl_3$(10%Ce) very promising to replace NaI(Tl) and CsI(Na) in γ-ray spectrometry and SPECT camera.

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