Light yield non-proportionality and intrinsic energy resolution of doped CsI scintillators

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Abstract. The light yield non-proportionality and energy resolution of doped CsI scintillators have been studied for γ-ray energies ranging from 16.6–1274.5 keV. The light yield non-proportionality of about 20% and the energy resolution of 7.6 ± 0.4% for 662 keV γ-rays have been achieved for ∅ 25 mm × 25 mm CsI(Na) crystal, coupled to an XP 5200 photomultiplier tube. The intrinsic resolution of the crystals vs. energy of γ-rays has been determined after correcting the measured resolution for photomultiplier tube statistics. The different shapes of intrinsic resolution curves observed with various samples of doped CsI are discussed.

Key words: scintillators • doped CsI • non-proportionality of light yield • energy resolution • intrinsic resolution

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Introduction

Inorganic scintillators play an important role in detection and spectroscopy of X/γ-rays as well as neutrons and charged particles. Important requirements for the scintillation crystals used in these applications include high light yield, fast response time, high stopping power, good proportionality of light yield, good energy resolution, minimal afterglow and low production costs. Good reviews on development of inorganic-scintillators and inorganic scintillation detectors/systems have been published by van Eijk [22], Moszynski [10], and recently by Lecoq *et al.* [8].

The phenomenon of non-proportionality response and its relation with energy resolution have been studied for many alkali halide scintillators, especially NaI(Tl), CsI(Tl) and CsI(Na) [1, 7, 9, 16, 17, 21], and oxide-based scintillators [2, 5, 11, 19, 20]. The light yield of alkali halides, expressed in photons per MeV of absorbed γ energy, decreases as the energy of γ-rays increases. On the contrary, oxide-based scintillators, in general, show an increasing light yield with increasing energy of γ-rays, which levels at higher energies.

Recently, Moszynski *et al.* [12–14] studied the scintillation properties of the pure NaI and CsI at LN$_2$ temperatures with large area avalanche photodiodes (LAAPD) readout. High light yield above 100,000 ph/MeV for CsI and the energy resolution of 3.8% for the 662 keV photopeak with NaI have been recorded for the best crystals. The studies showed that the intrinsic resolution of scintillators is strongly correlated with the non-proportionality response. However, both the non-proportionality and intrinsic energy resolution of pure NaI and CsI were affected by the impurities in the crystals studied.

The aims of this work are to perform a further study of energy resolution and light yield non-proportionality of doped halide CsI(Na) and $CsI(CO_3)$ crystals. From the obtained data on photoelectron yield vs. the energy of γ-rays and corresponding energy resolution, the intrinsic energy resolution of doped CsI crystals are determined.

Experimental procedures

Two doped halide crystals, CsI(Na) and $CsI(CO_3)$, with the same dimensions of ∅ 10 mm × 10 mm, supplied by CRYOS. Beta, Ukraine were studied. Both crystals were assembled by the manufacturer in the aluminum cases with a front glass window.

A CsI(Na) crystal of ∅ 25 mm × 25 mm supplied by Rexon, USA was used for comparison. The crystal was encapsulated by the manufacturer in an aluminum case with a front glass window.

Each crystal was optically coupled to a ∅ 52 mm Photonis XP 5200 photomultiplier tube using DC200 silicone grease. All measurements were made 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 an ORTEC 113 scintillation preamplifier and then to a Tennelec TC245 spectroscopy amplifier. A shaping time constant of 3 μs was used with CsI(Na) crystals. The primary decay time of $CsI(CO_3)$ crystal is 2 μs which is longer than that of 0.63 μs for CsI(Na) crystal. A shaping time constant of 12 μs was used with $CsI(CO_3)$ in order to sufficiently collect the scintillation light. The energy spectra were recorded using a Tukan PC-based multichannel analyzer [6].

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 16.6 keV and 1274.5 keV, as listed in Table 1. For isolated peaks, the energy resolution and centroid of the full energy peak were obtained by fitting a single Gaussian function. The background was fitted with a low order polynomial and subtracted from the spectrum. In order to separate the peak of interest from other partly overlapping peaks (e.g. Compton back-

Table 1. The radioactive sources and γ-ray energies

Source	Energy of γ-rays (keV)
[93]Mo	16.6 (K X-rays)
[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
[109]Cd	88
[57]Co	122
[51]Cr	320.1
[22]Na	511
[207]Bi	568
[137]Cs	661.6
[54]Mn	834.9
[207]Bi	1054
[22]Na	1274.5
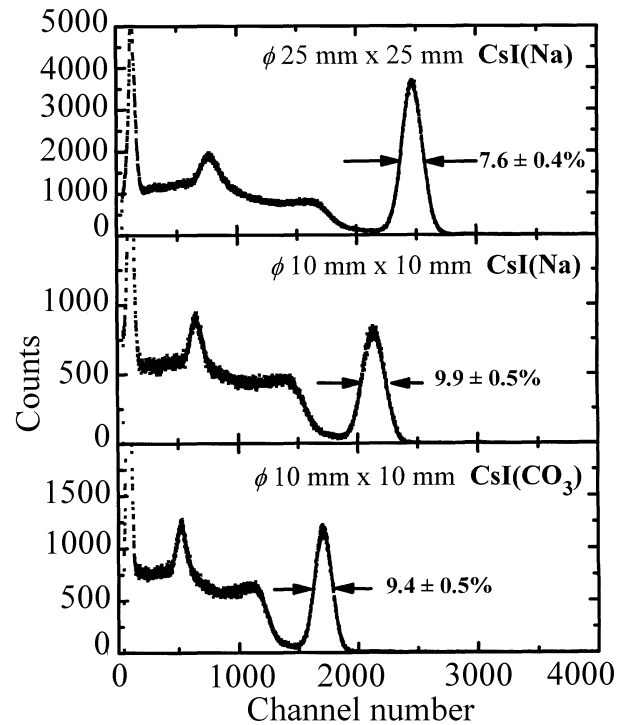
Fig. 1. Energy spectra of 661.6 keV γ-rays from a ^{137}Cs source, as measured with CsI(Na) and $CsI(CO_3)$ detectors.

scatter and K X-rays escape peaks), some peaks were fitted using two Gaussian functions with background subtraction, if necessary.

Results and discussion

Photoelectron yield and energy resolution

Figure 1 presents the energy spectra of 661.6 keV γ-rays from a ^{137}Cs source measured with $CsI(CO_3)$ and CsI(Na) crystals, coupled to XP5200 PMT. Note the better energy resolution of 7.6% obtained with large CsI(Na) in comparison to the value of 9.9% obtained with small CsI(Na) in spite of the fact that the volume ratio of the crystals is about 16 and thus a significant contribution from multiple Compton scattering in the large crystal.

Table 2 summarizes studies of tested CsI(Na) and $CsI(CO_3)$ samples. The photoelectron yield (N_{phe}) and energy resolution (ΔE/E), measured with XP5200 PMT at the optimal shaping time constant, are collected. The photoelectron yield, expressed in number of photoelectrons per MeV (phe/MeV) for each γ-peak, was measured by the Bertolaccini et al. method [3] used further in Ref. [15]. In this method the number of photoelectrons is measured directly by comparing

Table 2. Photoelectron yields and energy resolutions of CsI(Na) and $CsI(CO_3)$ crystals, coupled to XP5200 PMT, measured at 661.6 keV gamma rays

Crystal	Size (mm)	N_{phe} (phe/MeV)	ΔE/E (%)
CsI(Na)	∅ 25 × 25	3800 ± 140	7.6 ± 0.4
CsI(Na)	∅ 10 × 10	6700 ± 240	9.9 ± 0.5
$CsI(CO_3)$	∅ 10 × 10	4400 ± 150	9.4 ± 0.5

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. The single photoelectron spectrum was measured without the crystals, before and after measurements of the energy spectra with tested crystals. Note the better energy resolution of the large CsI(Na) for detection of high energy γ-rays in spite of the smaller photoelectron yield which is about two times less than the photoelectron yield for small CsI(Na). The 40% drop in the photoelectron yield between two CsI(Na) crystals can be associated with a different amount of doping or with additional codoping used by the crystal suppliers in the manufacturing processes.

It should be noted that the energy resolution at the 661.6 keV full energy peak for both the small samples is comparable in spite of the smaller photoelectron yield for CsI(CO$_3$) which is about 30% less than the photoelectron yield for CsI(Na).

Different energy resolutions and photoelectron yields observed with the tested samples suggested to compare the non-proportionality of the light yield and a contribution of intrinsic resolution to measured energy resolution [12].

Non-proportionality of light yield

Light yield non-proportionality as a function of energy can be one of the most important reasons for degradation in energy resolution of scintillators [5]. 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 662 keV γ-peak.

Figure 2 presents the non-proportionality characteristics of CsI(Na) and CsI(CO$_3$) crystals. Interestingly, both the small crystals exhibit a common curve within the experimental errors. Over the energy range from 17 keV to 1 MeV, the non-proportionality is about 30% for both the small CsI(Na) and CsI(CO$_3$) which is worse than that of about 20% for the large CsI(Na).

Aitken *et al.* measured the non-proportional response for the thin CsI(Na) crystal (∅ 10 mm × 0.8 mm) and observed the non-proportionality of about 28% in the energy range from 22 keV to 1 MeV [1]. Mengesha *et al.* also measured the non-proportional response for
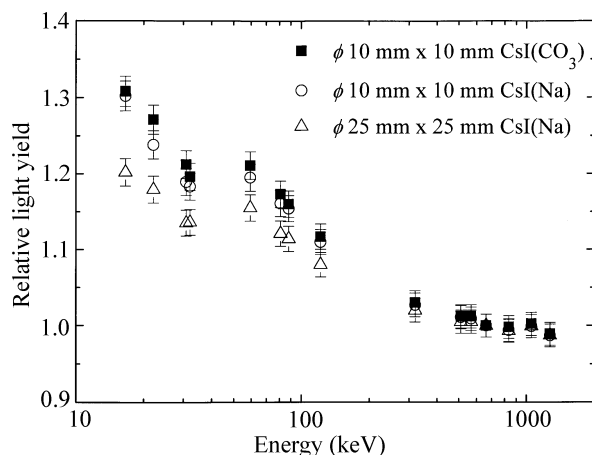
an equal sized (∅ 10 mm × 20 mm) CsI(Na) and CsI(Tl) crystals [9] in this energy range. They reported the non-proportionality of about 19% and 14% for CsI(Na) and CsI(Tl), respectively. Over the same energy range, the non-proportionality of about 17% was observed for a ∅ 9 mm × 9 mm CsI(Tl) crystal [18]. These results indicate that crystal thickness may be attributed to possible self-absorption of light in the scintillators, especially for low-energy photons, detected mainly at the top layer of the crystal. Different non-proportionality curves of tested samples should be reflected in their intrinsic resolutions.

Energy resolution

The energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled to a photomultiplier tube (PMT) can be written as [16]:

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

where δ_{sc} is the intrinsic resolution of the crystal; δ_p is the transfer resolution, and δ_{st} is the PMT contribution from the photoelectron statistics.

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

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

where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for the XP 5200 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 [16].

The intrinsic resolution of a crystal is connected with the δ-rays energy fluctuations, the non-proportional response of the scintillator [5, 7, 16] and many effects such as non-homogeneity 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:

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

Figures 3 and 4 present the measured energy resolution vs. energy of γ-rays for small CsI(Na) and CsI(CO$_3$) crystals. Both tested crystals exhibit step-like curves with a semi-plateau in the energy range between 100 and 300 keV. For lower and higher energies, the resolution decreases linearly with energy. Similar patterns of the step-like curves with a semi-plateau in the same energy
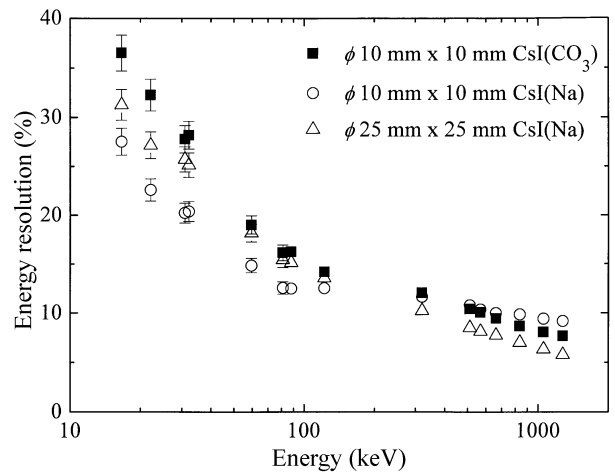

Fig. 2. Non-proportionality curves of studied CsI(Na) and CsI(CO$_3$) crystals.
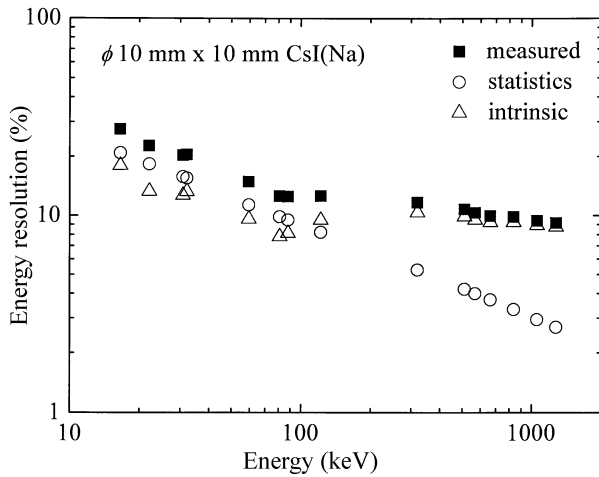
Fig. 3. Energy resolution and contributed factors vs. energy of $\varnothing$ 10 mm × 10 mm CsI(Na) crystal. The errors are of the same magnitude as the symbol size.
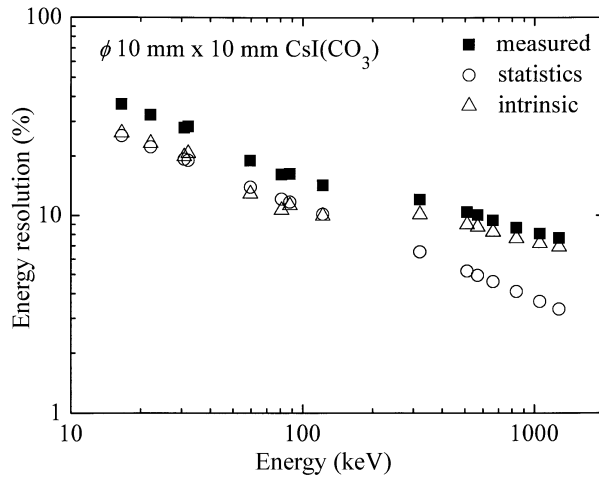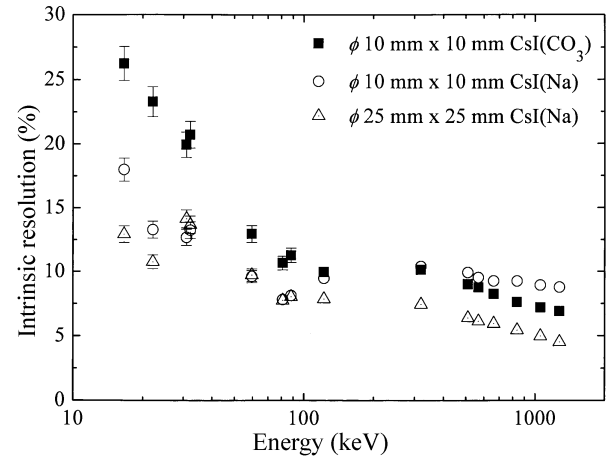

Fig. 4. Energy resolution and contributed factors vs. energy of $\varnothing$ 10 mm × 10 mm CsI(CO$_3$) crystal. The errors are of the same magnitude as the symbol size.

Fig. 5. Overall energy resolution of studied CsI(Na) and CsI(CO$_3$) detectors.

Fig. 6. Intrinsic resolution of studied CsI(Na) and CsI(CO$_3$) crystals.

range was also observed in Ref. [12] for the pure CsI crystals. 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. These results indicate that at energies above 300 keV the energy resolution is mainly due to the intrinsic resolution. In the low energy region the contribution of PMT resolution is of importance.

The measured overall energy resolution and the intrinsic resolution for the studied crystals are presented in Figs. 5 and 6, respectively. At energies below 100 keV the contribution of the intrinsic resolution for CsI(CO$_3$) is much higher than that for small CsI(Na). The photoelectron yield of small CsI(Na) around 60 keV is 8000 phe/MeV which is significantly larger than the yield of 5300 phe/MeV for CsI(CO$_3$). These are the main reasons for the better energy resolution of small CsI(Na) detector below 100 keV. However, despite the larger photoelectron yield, the energy resolution of small CsI(Na) detector significantly degrades as compared with the CsI(CO$_3$) detector at energies above 800 keV. The reason is its higher contribution from the intrinsic resolution.

For energies above 100 keV, the plots show a much lower contribution of the intrinsic resolution for the large CsI(Na) crystal, see Fig. 6. This together with its better proportionality of the light yield (see Fig. 2) are the main reasons for the superior overall energy resolution at energies above 300 keV (see Fig. 5), since the PMT contribution to the overall energy resolution at energies above 300 keV is small, especially for small crystal, with relatively high light yield. Consequently, the observed energy resolution is mainly due to the intrinsic resolution.

Table 3 summarizes the data relevant to the energy resolution of the 661.6 keV photopeak, performed for CsI(Na) and CsI(CO$_3$) scintillators in this study. 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 Eq. (2). From the values of $\Delta E/E$ and δ_{st}, the intrinsic resolution δ_{sc} is calculated using Eq. (3).

Moszynski *et al*. [16] have performed a comparison of the intrinsic resolution with the non-proportionality contribution from primary electrons (Compton electrons, photoelectrons and Auger electrons) for NaI(Tl) scintillator coupled to a PMT. The δ_{sc} at 661.6 keV energy was measured to be 5.8%, while the non-proportionality

Table 3. Energy resolution data at 661.6 keV gamma rays for CsI(Na) and CsI(CO$_3$) measured with the XP 5200 PMT

Crystal	Size (mm)	$\Delta E/E$ (%)	δ_{st} (%)	δ_{sc} (%)
CsI(Na)	$\varnothing$ 25 $\times$ 25	7.6	4.9	5.8
CsI(Na)	$\varnothing$ 10 $\times$ 10	9.9	3.7	9.2
CsI(CO$_3$)	$\varnothing$ 10 $\times$ 10	9.4	4.6	8.2

component from primary electrons (δ_{np}) was found to be about 2.6%. Consequently, the non-proportionality contribution from the secondary electrons, namely δ-rays (δ_δ) was obtained to be about 5.2% from $[(\delta_{sc})^2 - (\delta_{np})^2]^{1/2}$ by assuming that the δ_{sc} is weakly affected by the contribution of crystal inhomogeneity (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 3, intrinsic resolution δ_{sc} of a large CsI(Na) was measured to be 5.8%. The measured non-proportionality of its light yield was about 18% in the energy range from 22 keV to 1274.5 keV which is slightly higher than the value of about 15% for NaI(Tl). This results indicate that the contribution of δ_δ is also a major component for the δ_{sc} in CsI(Na) by assuming that the intrinsic resolution is correlated with the non-proportional response and the non-proportionality component from primary electrons is also small for CsI(Na). The intrinsic resolution of small CsI(Na) was measured to be 9.2% which is much higher than the value of 5.8% for the large CsI(Na). By assuming that the contributions from δ_{inh} and light collection properties for a large CsI(Na) sample is negligible, an additional increase in the intrinsic resolution for small CsI(Na) sample is accounted mainly by the contribution from the effect of light collection and crystal inhomogeneity (δ_{inh}). Non-uniformity in the spatial distribution of the activator within the host CsI materials and non-uniform reflectivity of the reflecting capsule are expected to make a large contribution to the observed intrinsic resolution for both the small samples.

A lower intrinsic resolution for large CsI(Na) crystal at energies above 200 keV may also be due to some manufacturing details. It seems that the large CsI(Na) crystal has a different amount of doping or may be some additional codoping. Brecher *et al.* [4] have recently reported that adding Eu^{2+} ions to CsI(Tl) reduces both the afterglow and the light yield. An impact of codopant on energy resolution was also observed, although needs further exploiting.

Between 100 and 700 keV, both small crystals exhibit comparable intrinsic resolution, reflecting its approximately a common non-proportionality curve. A larger intrinsic resolution for CsI(CO$_3$) at energies below 100 keV may be due to an increases in surface effects, different in both crystals. These low energy photons, especially at energies below 40 keV, are mainly absorbed in the surface layer at the top of the crystal. If the scintillation or light collection efficiencies at the surface are somewhat different from the bulk, it will result in a larger spreading of the total light produced and intrinsic resolution increase. A better encapsulation of the crystal might improve the intrinsic resolution at energies below 100 keV, especially for the CsI(CO$_3$) crystal.

Conclusions

In this work, the scintillation properties of canned CsI(Na) and CsI(CO$_3$) crystals were studied. The study showed that the overall energy resolution of a small CsI(Na) detector is better than that of an equal sized CsI(CO$_3$) detector at energies below 100 keV due to a larger photoelectron yield (by almost about 50%) measured for a small CsI(Na) crystal as well as a much lower contribution of the intrinsic resolution for the CsI(Na) crystal at energies below 100 keV. At energies above 300 keV, despite the larger photoelectron yield, the energy resolution of both the small detectors significantly degrades as compared with those of the large CsI(Na) detector. The reason is much higher contribution of the intrinsic resolution for both small crystals, which may be explained by a large contribution from the effects of light yield non-proportionality, crystal inhomogeneity and non-unifrom reflectivity of the reflecting capsule. The large difference in the light yield and the intrinsic resolution between small and large CsI(Na) crystals indicates that some manufacturing details during the process of growing the ingots and quality of reflecting encapsulation of the crystals are also of importance.

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