สมบัติด้านซินทิลเลชันของผลึก $CsI(CO_3)$ และ CsI(Na) สำหรับการตรวจวัดรังสีแกมมา

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บทคัดย่อ

งานวิจัยนี้ได้ทำการศึกษาการตอบสนองด้านซินทิลเลชันของผลึก $CsI(CO_3)$ และ CsI(Na) ที่มีขนาด ∅10 มม. x 10 มม. สำหรับรังสีแกมมาในช่วงพลังงาน 16.6 keV ถึง 1274.5 keV จากการศึกษาความไม่เป็นสัดส่วนของแสงซินทิลเลชัน (non-proportionality of light yield) และการแยกชัดพลังงาน (energy resolution) ของผลึกทั้งสองชนิดด้วยหลอดทวีคูณแสงเบอร์ XP5200 พบว่า ผลึกทั้งสองชนิดแสดงความไม่เป็นสัดส่วนของแสงซินทิลเลชันมีค่าร่วมกัน โดยให้การแยกชัดพลังงานรังสีแกมมา 662 keV มีค่า 9.4 ± 0.5% และ 9.9 ± 0.5% สำหรับผลึก $CsI(CO_3)$ และ CsI(Na) ตามลำดับ นอกจากนี้ยังพบว่าการแยกชัดพลังงานของผลึกทั้งสองชนิดมีลักษณะกึ่งราบ (semi-plateau) ในช่วงพลังงาน 100 keV ถึง 300 keV การปรับแก้การแยกชัดพลังงานรวมที่วัดได้ด้วยส่วนที่มาจากความไม่แน่นอนทางสถิติของหลอดทวีคูณแสง สามารถนำไปสู่การแยกชัดในตัว (intrinsic resolution) ของผลึกแต่ละชนิดในแต่ละช่วงพลังงาน บทความนี้ได้อภิปรายถึงการแยกชัดพลังงานรวมและการแยกชัดในตัวของผลึก

คำสำคัญ : ซินทิลเลเตอร์ / $CsI(CO_3)$ / CsI(Na) / ความไม่เป็นสัดส่วนของแสงซินทิลเลชัน / การแยกชัดพลังงาน / การแยกชัดในตัว

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Scintillation Properties of CsI(CO_3) and CsI(Na) Crystals for Gamma Ray Detection

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Abstract

The scintillation response of CsI(CO_3) and CsI(Na) crystals with the same size of ∅10 mm x 10 mm were studied for γ-ray energies ranging from 16.6 keV to 1274.5 keV. The light yield non-proportionality and the energy resolution were measured with an XP5200 PMT. Both crystals showed a common light yield non-proportionality with yielding energy resolution of 9.4 ± 0.5% and 9.9 ± 0.5% at 662 keV γ-rays respectively, for CsI(CO_3) and CsI(Na). The step-like curves of energy resolution with a semi-plateau in the energy range between 100 and 300 keV have been observed for both crystals. The intrinsic resolution of the crystals versus energy of gamma rays has been determined after correcting the measured resolution for photomultiplier tube statistics. The overall and intrinsic energy resolutions are discussed.

Keywords : Scintillators / CsI(CO_3) / 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 X/γ-rays as well as neutrons and other energetic 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 [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 alkali halide scintillators, especially NaI(Tl), CsI(Tl) and CsI(Na) [4-9], and oxide based scintillators [10-14]. The scintillation response of alkali halides decreases as the photon energy increases, whereas oxide based scintillators in general show an increasing scintillation response with increasing photon energy at higher energy levels.

Recently, Moszynski et al. [15-17] studied the scintillation properties of the pure NaI and CsI at LN_2 temperatures with large area avalanche photodiodes (LAAPD) readout. High light output above 100,000 photons/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-proportional 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 output proportionality of doped halide CsI(Na) and $CsI(CO_3)$ crystals and to investigate the influence of activators. Obtained data on photoelectron yield and corresponding energy resolution lead to the contribution of the intrinsic energy resolution for tested crystals.

2. Experimental procedures

Two doped halide crystals, CsI(Na) and $CsI(CO_3)$, with the same dimensions of ∅10 mm x 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.

The crystals were optically coupled to ∅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) crystal and 12 μs was used with $CsI(CO_3)$ crystal. The energy spectra were recorded using a Tukan PC-based multichannel analyzer [18].

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 each γ peak, the full width at half maximum (FWHM) and centroid of the full energy peak were obtained from Gaussian fitting software of Tukan MCA.

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	1,054
^{22}Na	1,274.5

3. Results and Discussion

3.1 Light yield and energy resolution

Fig. 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. The energy resolution of 9.4 % obtained with CsI(CO_3) is comparable to the value of 9.9 % obtained with CsI(Na).

Fig. 2 presents a similar set of spectra measured with 122 keV γ-rays from a ^{57}Co source. The 122 keV photopeak is accompanied by satellite peak at lower energy, due to the escape of characteristic KX-rays of Cesium and Iodine. To determine the energy resolution, the photopeak was fitted with two Gaussian peaks. The resolution is 14.2% for CsI(CO_3) and 12.5% for CsI(Na). Note the better energy resolution of CsI(Na) for detection of low energy γ-rays.

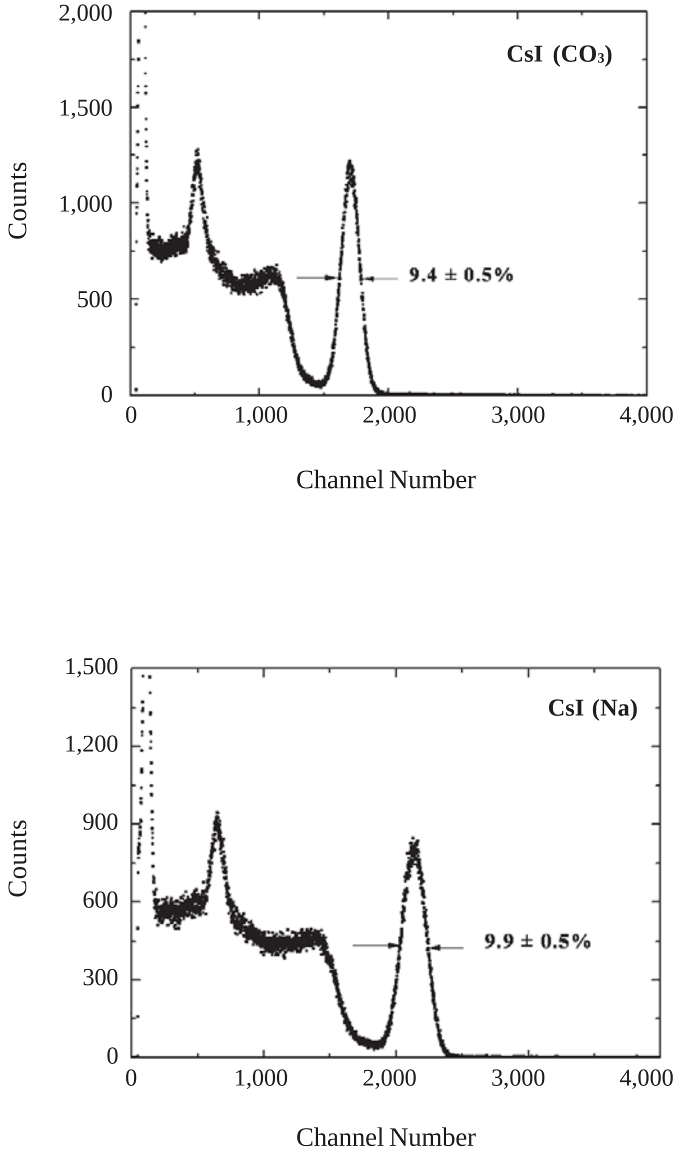

Fig. 1 Energy spectra of 661.6 keV γ-rays from a ^{137}Cs source, as measured with CsI(Na) and $CsI(CO_3)$ crystals.

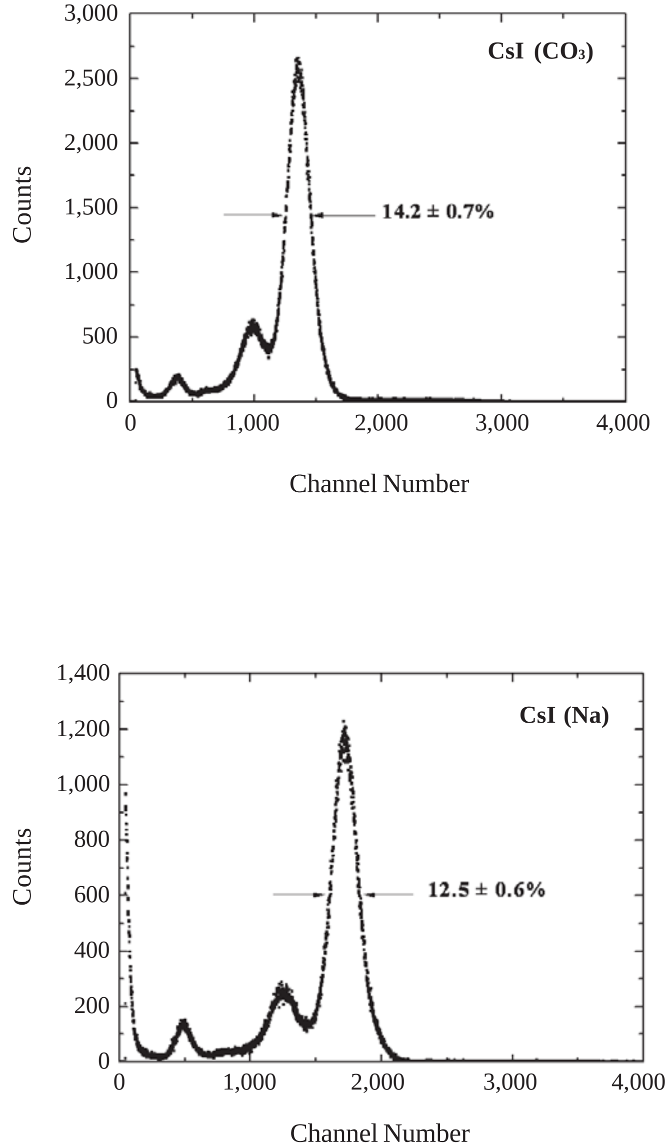

Fig. 2 Energy spectra of 122 keV γ-rays from a ^{57}Co source, as measured with CsI(Na) and CsI(CO_3) crystals.

Table 2 summarizes studies of tested CsI(Na) and CsI(CO_3) samples. The light yield and energy resolution, measured with XP5200 PMT at the optimal shaping time constant, are collected. The light yield, expressed in number of photoelectrons per MeV (phe/MeV) for each γ-peak, was measured by the Bertolaccini et al. method [19,20]. 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 measured without the crystals, and it was performed before and after measurements of the energy spectra with tested crystals. From these spectra with associated amplifier gains, the number of photoelectrons/MeV of full energy peaks at specific γ-ray energy were determined. It should be noted that the energy resolution at the 661.6 keV full energy peak for both 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 crystals suggested comparing the non-proportionality of the light yield and a contribution of intrinsic resolution to measured energy resolution [17].

3.2 Non-proportionality of light yield

Light yield non-proportionality as a function of energy is one of the most important reasons for degraded 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 662 keV γ-peak.

Table 2 Light yields and energy resolutions of CsI(Na) and CsI(CO_3) crystals coupled to XP5200 PMT, measured at 122 keV and 661.6 keV γ-rays.

Crystal	Light yield (phe/MeV)	$\Delta E/E$ (%)	Energy (keV)
CsI(Na)	7400 ± 270	12.5 ± 0.6	122
CsI(CO_3)	4900 ± 180	14.2 ± 0.7	
CsI(Na)	6700 ± 240	9.9 ± 0.5	661.6
CsI(CO_3)	4400 ± 160	9.4 ± 0.5	

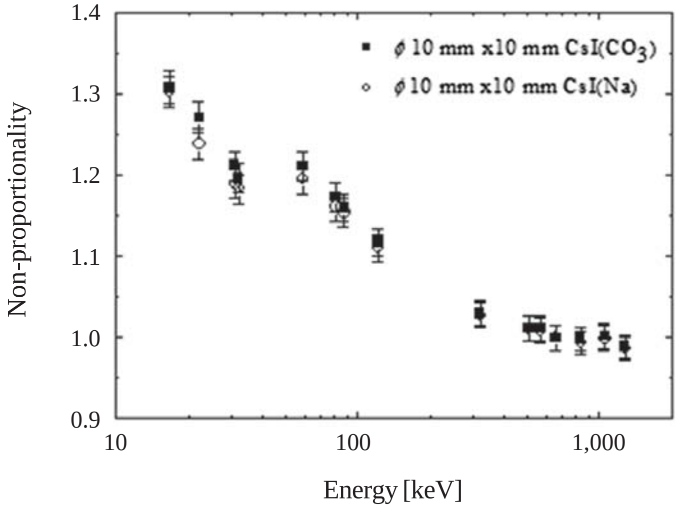

Fig. 3 Non-proportionality curves of studied CsI(Na) and CsI(CO_3) crystals.

Fig. 3 presents the non-proportionality characteristics of CsI(Na) and CsI(CO_3) crystals. Interestingly, both 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 crystals.

Aitken et al. measured the non-proportional response for the 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 the non-proportional response for an equal sized (∅10 mm x 20 mm) CsI(Na) and CsI(Tl) crystals [6] 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 22% was observed for a ∅10 mm x 10 mm CsI(Tl) crystal [17]. 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. As the results from this study, both an equal sized CsI(CO_3) and CsI(Na) crystals exhibit approximately a common non-proportionality curve, which is significantly worse than that of CsI(Tl) reported in the previous works. Further studies carried out with CsI(Tl), pure CsI at room and liquid nitrogen temperatures and pure NaI at room temperature seem to suggest that the observed effects of non-proportionality and the intrinsic resolution dependency on shaping time constant are characteristic of crystals with light pulses exhibiting two decay time components [2].

3.3 Energy resolution

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

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

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

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

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

where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.1 for the 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 [8].

The intrinsic resolution of a crystal is connected with the δ-rays energy fluctuations, the non-proportional response of the scintillator [4, 8, 13] and many effects such as inhomogeneities 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:

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

Figs. 4 and 5 present the measured energy resolution versus energy of γ-rays for 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 range was also observed in Ref. [17] 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. 6 and 7, respectively. At energies below 100 keV the contribution of the intrinsic resolution for CsI(CO_3) is much higher than that for CsI(Na). The light yield of small CsI(Na) around 60 keV is 8,000 phe/MeV which is significantly larger than the yield of 5,300 phe/MeV for CsI(CO_3). These are the main reasons for the better energy resolution of small CsI(Na) below 100 keV.

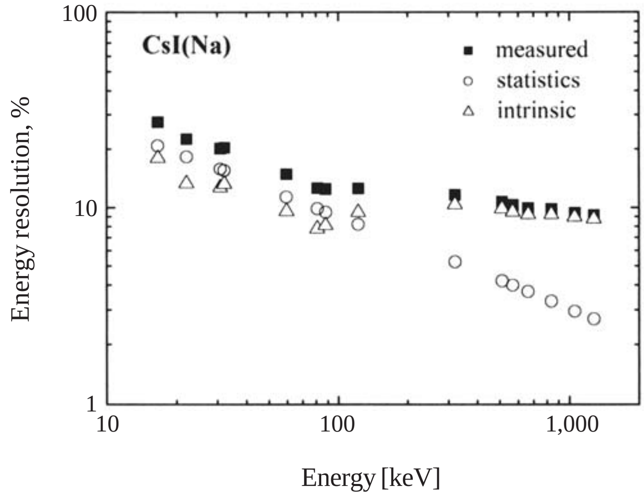

Fig. 4 Energy resolution and contributed factors versus energy of ∅10 mm x 10 mm CsI(Na) crystal. The errors are of the same magnitude as the symbol size.

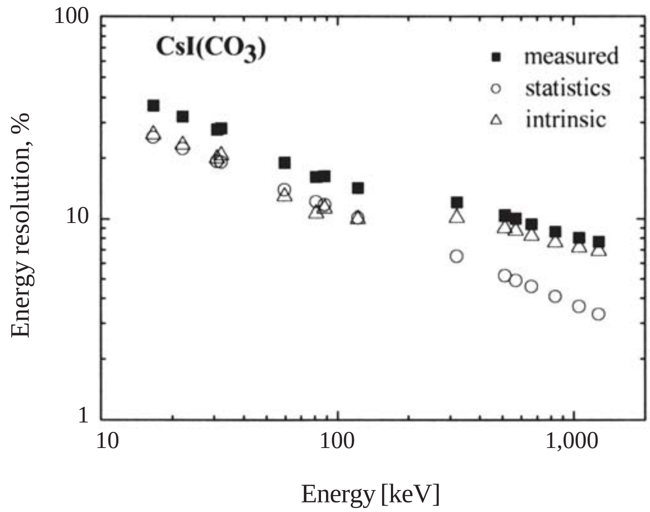

Fig. 5 Energy resolution and contributed factors versus energy of ∅10 mm x 10 mm CsI(CO_3) crystal. The errors are of the same magnitude as the symbol size.

However, despite a larger light yield, the energy resolution of CsI(Na) significantly degrades as compared with the CsI(CO_3) at energies above 800 keV. The reason is its higher contribution from the intrinsic resolution.

What is the origin of this intrinsic resolution? In the present experiment, comparable intrinsic resolutions were determined for both crystals at energies between 100 and 700 keV. Note that the observed intrinsic resolution of the CsI(CO_3) is higher than that observed with the CsI(Na) at energies below 100 keV whereas the both crystals exhibit a common non-proportionality over the energy range from 17 keV to 1 MeV. It seems to suggest that the contribution of δ-rays energy fluctuations and the contribution of the non-proportional response of the scintillator are not the only dominant components for the halide crystals.

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 increases.

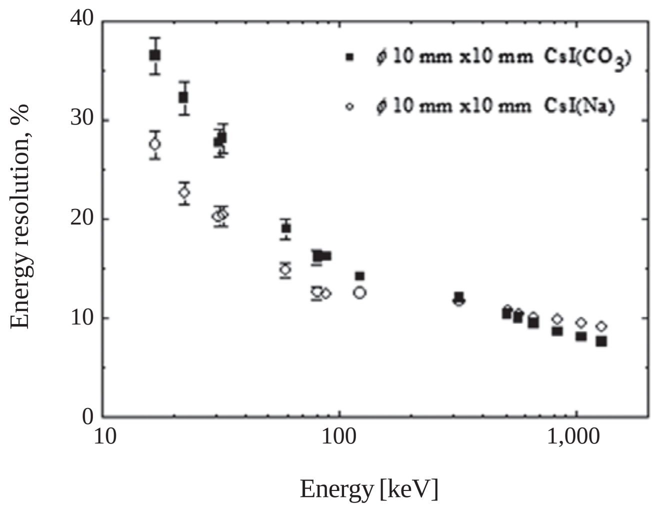

Fig. 6 Overall energy resolution of studied CsI(Na) and CsI(CO_3) crystals.

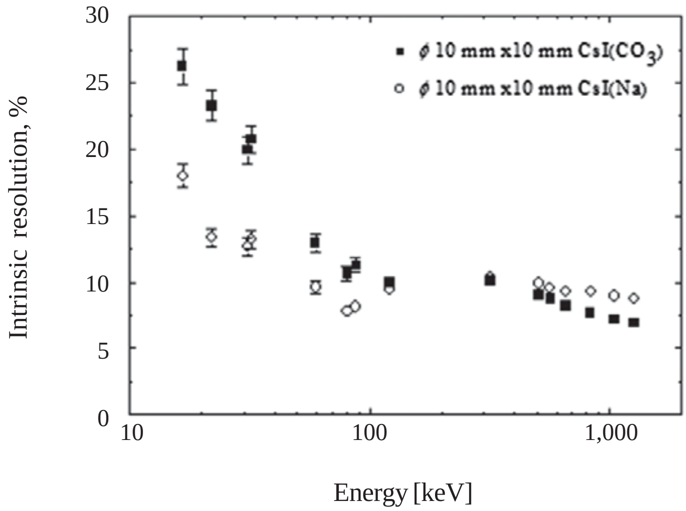

Fig. 7 Intrinsic resolution of studied CsI(Na) and CsI(CO_3) crystals.

4. 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 CsI(Na) crystal is better than that of an equal sized CsI(CO_3) crystal at energies below 100 keV due to a larger light yield (by almost about 50%) measured for CsI(Na) crystal as well as a much lower contribution of the intrinsic resolution for the CsI(Na) crystal at energies below 100 keV.

Between 16.6 keV and 1,274.5 keV the energy resolution for CsI(Na) and CsI(CO_3) shows the step-like curves with a semi-plateau in the energy range between 100 and 300 keV. Over this energy range, the non-proportionality in light yield is about 30% for both CsI(Na) and CsI(CO_3) crystals. Activators influence the non-proportionality response of alkali halide crystals, probably by changing scintillation decay time and emission spectrum, but they are not the only decisive factors. This study seems to confirm that the surface effects and δ-rays energy fluctuations are the main components in the intrinsic resolution for doped CsI scintillators, whereas the contribution from the non-proportional response of the scintillator is a fundamental limitation of obtainable energy resolution [8].

5. Acknowledgements

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