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Light Yield Non-proportionality and Energy Resolution of BGO Scintillation Crystals

[1]Akapong Phunpueok, [1]Voranuch Thongpool, [2]Sikarin Yoo-Kong & [2]Weerapong Chewpraditkul

[1]Rajamangala University of Technology Thanyaburi, Faculty of Science and Technology,Division of Physics, Pathumthani, Thailand
[2]King Mongkut's University of Technology Thonburi, Faculty of Science, Department of Physics, Bangkok, Thailand

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ABSTRACT

Nowadays inorganic scintillators play an important role in detection and spectroscopy of energetic photons and nuclear particles, in particular medical imaging. 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. The main advantages of $Bi_4Ge_3O_{12}$(BGO) are high stopping power and non-hygroscopic which make them very promising scintillator for medical imaging. The aims of this work are to perform the further study of light yield non-proportionality and energy resolution of different sizes of BGO crystals covering energies from 22.1 to 1,274.5 keV using photomultiplier tube (PMT) readout. The intrinsic resolution of all crystals has been determined after correcting the measured energy resolution for PMT statistics. The result demonstrates that the contribution from the non-proportional response of the scintillator is strong correlated with the intrinsic resolution of the scintillators and photofraction at 662 keV gamma rays (^{137}Cs source) depending on sizes of crystals.

Key words: energy resolution, intrinsic resolution, light yield non-proportionality, BGO

INTRODUCTION

At the present time inorganic scintillators play an important role in radiation detection in many sectors of basic and applied research, in particular medical imaging. 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 inorganic scintillation detectors have been published by Moszynski [1], van Eijk [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 [4-5] and oxide based scintillators[6-7]. 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, which levels at higher energies. Bismuth germinate ($Bi_4Ge_3O_{12}$, BGO) crystals advantages are such as large absorption coefficient, non-hygroscopicity and high density and effective atomic number. Due to these factors, BGO has been widely used as detector of X-/γ-rays in nuclear medicine and gamma ray spectroscopy. However, one of drawbacks of BGO crystals is poor energy resolution. The aims of this work are to perform a further study of light yield non-proportionality and energy resolution of three different sizes of BGO crystals covering energies from 22.1 to 1,274.5 keV. From the obtained data on photoelectron yield versus the energy of gamma rays and corresponding energy resolution, the light yield non-proportionality and the intrinsic energy resolution of tested crystals were calculated. The estimated photofraction for all tested crystals at 662 keV gamma peak will also be discussed. The mass attenuation coefficient of all crystals for 662 keV gamma rays was also measured by transmission method and compared with the theoretical values calculated by WinXCom program.

Materials And Methods

Three different size of BGO crystals with the dimensions of 15x15x10 and 15x15x5 mm^3 grown by the Czochralski method at the Shanghai Institute of Ceramics (P.R.China) and 7x7x1 mm^3 grown by the Bridgman method at the Shonan Institute of Technology (Fujisawa, Japan). The crystals were optically coupled to a Photonis XP5200B photomultiplier tube using silicone grease. All measurements were made using standard NIM level

Corresponding Author: Akapong Phunpueok, Rajamangala University of Technology Thanyaburi, Faculty of Science and Technology, Division of Physics, Pathumthani, Thailand
E-mail: aumaum18@hotmail.com Tel. +662-549-4193

electronics. The sources were positioned along the cylindrical axis of the scintillator and the PMT. The signal from the PMT anode was passed to a CANBERRA2005 preamplifier and was sent to a Tennelec TC243 spectroscopy amplifier. A shaping time constant of 4 µs was used with crystals. The energy spectra were recorded using a Tukan 8k [8] PC-based multichannel analyzer (MCA)[9]. The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each gamma peak, was measured by Bertolaccini method [10]. In this method the numbers of photoelectrons are measured by comparing the position of a full energy peak of gamma 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 gamma rays emitted by different radioactive sources (^{109}Cd, ^{241}Am, ^{133}Ba, ^{51}Cr, ^{137}Cs, ^{58}Co, and ^{22}Na) in the energy range between 22.1 and 1,274.5 keV. For each gamma peak, the full width at half maximum (FWHM) and centroid of the full energy peak were obtained from Gaussian fitting software of Tukan MCA. The total mass attenuation coefficients at 662 keV γ-rays for all crystals were determined using the good geometry arrangement of source (^{137}Cs), absorber (crystal) and detector (plastic scintillator). A narrow beam of γ-rays is defined by circular apertures (∅ 2 mm) in the Pb-collimators of source and detector, placed at a distance of 60 cm.

Results and Discussion

Photoelectron Yield and Energy Resolution:

Fig 1. presents the energy spectra of 662 keV gamma rays from a ^{137}Cs source measured with three different size of BGO detectors. It is seen that small BGO (7x7x1mm^3) gives little better energy resolution than medium BGO (15x15x5 mm^3) and large BGO (15x15x10 mm^3). The energy resolution of 8.9% obtained with small BGO is little better than the value of 9.1 and 9.4% obtained with medium BGO and large BGO, respectively. The energy resolution of 8.9 % for the tested small BGO crystal in this study is better than that of 10.0% for sample observed by Moszynski et al.[11] for BGO crystal (∅ 9x4 mm^3) supplied by Bicron at room temperature. Note KX-rays escape peak in the spectrum measured with small BGO crystals is very clear than that of other BGO crystals.

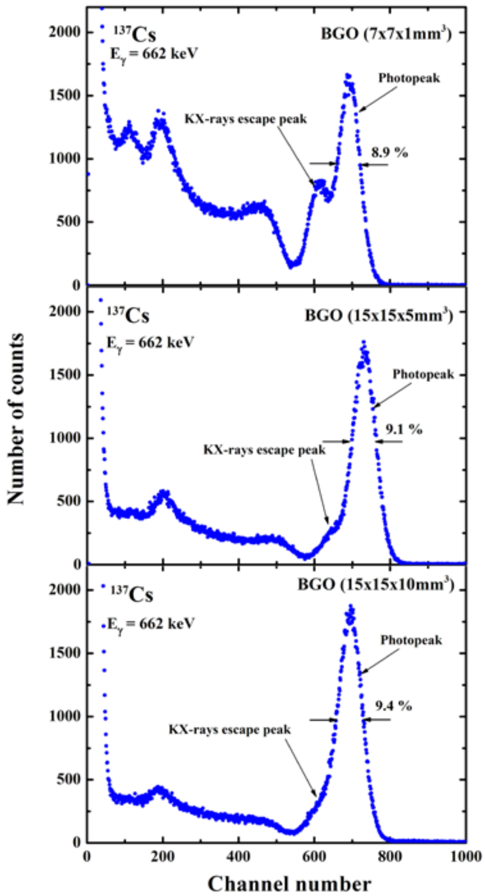

Fig. 1: Pulse Height Spectra of Gamma Rays from ^{137}Cs(662 keV) Source as Measured with three different sizes of BGO crystals.

Table 1: Photoelectron Yield, Light Yield and Energy Resolution at 662 keV Gamma Rays for Tested Crystals as Measured with the Photonis XP5200B PMT.

Crystal	Photoelectron Yield [phe/MeV]	Light Yield [ph/MeV]	Energy Resolution [%]
Small BGO	1840 ± 90	8900 ± 900	8.9 ± 0.5
Medium BGO	1780 ± 90	8600 ± 900	9.1 ± 0.5
Large BGO	1770 ± 90	8600 ± 900	9.4 ± 0.5

Table 1 summarizes comparative measurements of photoelectron yield, light yield and energy resolution at 662 keV gamma rays for the tested crystals coupled to the Photonis XP5200B PMT, as measured at 4 µs shaping time constant in the spectroscopy amplifier. The small BGO showed a photoelectron yield of 1,840 phe/MeV corresponding to about 8,900 Photon/MeV (ph/MeV), while medium BGO and large BGO showed about 1,770 phe/MeV corresponding to about 8,600 ph/MeV, at the PMT photocathode quantum efficiency (QE) of 20.7% for peak emission of 480 nm. Note a significantly higher light yield of 8,900 ph/MeV for the small BGO crystal, by about 130%, compared with a bigger sized sample in Ref. [11]. This result shows an improvement of light output for the tested crystal. The different thickness (5 vs 10 mm) of the crystals shall little affect the absorption of emitted light decreasing the light yield (1,780 vs 1,770 phe/MeV) of the BGO crystals. The energy resolution (ΔE/E) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [12]

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

where δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution and δ_{st} is the statistical contribution of the 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, and equals to 0.1 for an XP5200B PMT. The transfer component depends on the quality of optical coupling of the crystal and PMT, homogeneity of quantum efficiency of the photocathode 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 [12]. The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [12] and many effects such as inhomogeneities in the scintillator which can cause local variations 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 follows

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

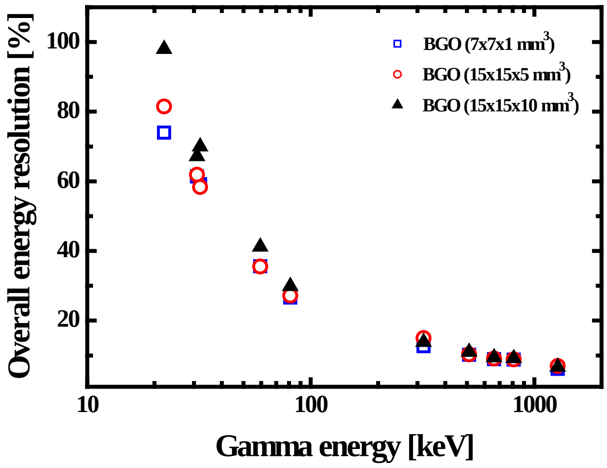

Fig. 2: Overall Energy Resolution of Three Different Sizes of BGO Crystals.

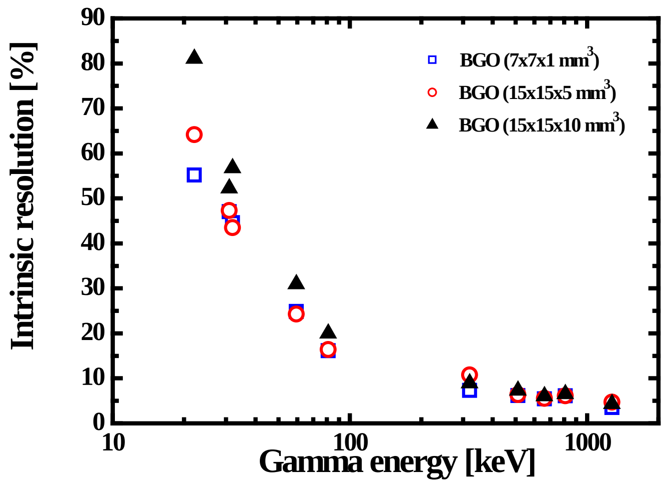

Fig. 3: Intrinsic Resolution of Three Different Sizes of BGO Crystals.

Fig. 2 presents a direct comparison of the overall energy resolution for all tested BGO crystals. The overall energy resolution of large BGO crystal is worse than that of small and medium BGO crystals in the energy range below 80 keV, which is, reflected by a worse intrinsic resolution (see Fig. 3).The intrinsic resolution of large BGO crystal is worse than that of other BGO crystals in the energy range below 80 keV, which is, reflected by a inferior proportionality of the light yield (see Fig. 4).

Table 2: Analysis of the 662 keV Energy Resolution for Three Different Sizes of BGO Crystals.

Crystal	N [electron]	ΔE/E [%]	δ_{st} [%]	δ_{sc} [%]
Small BGO	2150 ± 220	7.4 ±0.4	5.3 ± 0.3	5.2 ± 0.3
Medium BGO	1670 ± 170	9.0 ± 0.5	6.1 ± 0.3	6.7 ± 0.3
Large BGO	1220 ± 120	8.9 ± 0.4	7.1 ± 0.4	5.4 ± 0.3

To better understand the energy resolution of all tested crystals in gamma ray spectrometry, the contribution of various components to the overall energy resolution was analyzed for 662 keV photopeak, and the results are presented in Table 2. The second column gives N, the number of photoelectrons produced in the PMT. The third column gives ΔE/E, the overall energy resolution at 662 keV photopeak. The PMT contribution (δ_{st}) was calculated using Eq.(2). From the values of ΔE/E and δ_{st} , the intrinsic resolution (δ_{sc}) was calculated using Eq.(3).

The superior energy resolution of small BGO crystal as compared to other BGO crystals is mainly due to a small contribution of both δ_{st} and δ_{sc} , which seems to follow a high light output and good proportionality of the light yield, respectively, for small BGO crystal. For the superior energy resolution of small BGO crystal as compared to other BGO crystals is mainly due to a small contribution of both δ_{st} and δ_{sc} , which seems to follow a high light output and good proportionality of the light yield, respectively, for small BGO crystal.

Non-proportionality of the 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 gamma ray energy relative to the yield at 662 keV gamma peak.

Fig. 4 presents the light yield non-proportionality characteristics of three different sizes of BGO crystals in the energy range of 22.1 to 1,274.5 keV. Large BGO crystal is clearly poorer than other BGO crystals in terms of light yield proportionality. To better understand, the degree of non-proportionality (σ_{np}) proposed by Dorenbos [14] was calculated with this energy range, we obtain a value of 0.19 for small BGO, 0.18 for medium BGO and 0.24 for large BGO. The degree of non-proportionality of small and medium BGO crystals is lower than that of large BGO crystal, reflecting in lower intrinsic resolution for small and medium BGO crystals.

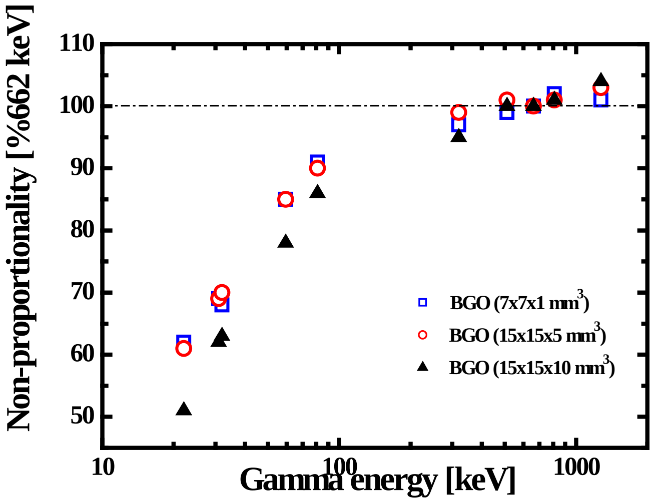

Fig. 4: Non-proportionality of the Light Yield of Three Different Sizes of BGO Crystals.

Photofraction:

Table 3: Photofraction at 662 keV Gamma Peak for all Tested BGO Crystals.

Crystal	Volume [cm^3]	Photofraction [%]	σ-ratio [%]
Small BGO	0.049	26.9 ± 2.7	20.5
Medium BGO	1.125	45.0 ± 4.5	20.5
Large BGO	2.250	53.3 ± 5.3	20.5

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 gamma ray energy. The photofraction for all tested BGO crystals at 662 keV gamma peak is collected in Table 3. For a comparison, the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCom program [15] are given too. The data indicate that large BGO crystal shows much higher photofraction than small and medium BGO crystals due to higher volume (2.250 vs 0.049 cm^3, 2.250 vs 1.125 cm^3) of the large BGO crystal. The bigger BGO crystal shows higher photofraction than smaller BGO crystal due to the thickness of the samples. It is the contribution of multiple Compton scattering to create a full energy peak. The small BGO crystal show the value of photofraction to be near to the value of the cross-section ratio (σ-ratio) obtained from WinXCom program.

A parallel beam of monoenergetic γ-rays is attenuated in an absorber according to the Lambert-Beer law:

$$I = I_o \exp(-\mu_m \rho t), \qquad (4)$$

where I_o and I are incident and transmitted intensities of gamma rays, respectively, μ_m is the mass attenuation coefficient, ρ is the density of the absorber, and t is the thickness of the absorber. The product $\mu_m\rho$ is called the linear attenuation coefficient. Theoretical values of the mass attenuation coefficients of mixture have been calculated by WinXCom program.

Table 5 shows the experimental $(\mu_m)_{ex}$ and theoretical $(\mu_m)_{th}$ values of the mass attenuation coefficients for all crystals at 662 keV gamma rays. The results are in good agreement within the experimental uncertainty.

Total Mass Attenuation Coefficient:

Table 4: Total mass attenuation coefficient at 662 keV gamma rays for all BGO crystals

Crystal	$(\mu_m)_{ex}$ [cm^2/g]	$(\mu_m)_{th}$ [cm^2/g]	RD* [%]
Small BGO	9.72 x 10^{-2}	9.95 x 10^{-2}	2.31
Medium BGO	9.69 x 10^{-2}	9.95 x 10^{-2}	2.61
Large BGO	9.61 x 10^{-2}	9.95 x 10^{-2}	3.42

* Relative difference between $(\mu_m)_{ex}$ and $(\mu_m)_{th}$

Conclusion:

In this work, the scintillation properties of three different sizes of BGO crystals were studied and compared in gamma ray spectrometry. The energy resolution of small BGO crystal is little superior than that of other BGO crystals due to a high light output and small contribution from its intrinsic resolution, reflecting a better proportionality of light yield between 22.1 and 1,274.5 keV. This study demonstrates that the contribution from the non-proportional response of the scintillator is strong correlated with the intrinsic resolution of the scintillators. The experimental results of total mass attenuation coefficients of all crystals are in good agreement with the theoretical values, calculated by WinXCom. In conclusion, the different thickness (1, 5, 10 mm) of the crystals shall affect the absorption of emitted light decreasing the light yield (1840, 1780, 1770 phe/MeV) of the crystals. The main advantages of BGO crystals are non-hygroscopicity and high photofraction due to its high effective atomic number and density which make them very promising scintillator for medical imaging.

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