

Comparison of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ Scintillators in Gamma Ray Spectrometry

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Abstract. The scintillation response of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ scintillation crystals have been compared using photomultiplier tube readout for photon energies ranging from 22.1 to 1274.5 keV. $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ showed a light yield non-proportionality of about 20% upon lowering energy from 1,274.5 to 22.1 keV, which is better than that of about 39% obtained for $\text{Bi}_4\text{Ge}_3\text{O}_{12}$. $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ showed the light yield of 13,400 ph/MeV and energy resolution of about 8 % for 662 keV gamma rays from a ^{137}Cs source. The photofraction of $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ is better than that of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$. The intrinsic resolution of the crystals versus energy of gamma rays has been determined after correcting the measured energy resolution for photomultiplier tube statistics.

Introduction

Inorganic scintillators play an important role in detection and spectroscopy of energetic photons and nuclear particles. Important requirements for the scintillators used in these applications include high light yield, fast response time, high stopping power and good energy resolution. 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 [4-6] and oxide based scintillators [7-9]. 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.

The aims of this work are to perform a further study of energy resolution and light output of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ ($\text{Lu}_{0.7}\text{Y}_{0.3}\text{AlO}_3\text{:Ce}$) and BGO ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$) 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 tested crystals at 662 keV gamma peak will also be discussed.

Methodology

$\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ ($10 \times 10 \times 5 \text{ mm}^3$) and BGO ($15 \times 15 \times 4 \text{ mm}^3$) crystals, supplied by Proteus Inc.(USA) and Shanghai Institute of Ceramics (P.R.China), respectively, were studied.

Each crystal was optically coupled to a Photonis XP5200B photomultiplier tube (PMT) using 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 anode was passed to a CANBERRA2005 preamplifier and was sent to a Tennelec TC243 spectroscopy amplifier. A shaping time constant of $4 \mu\text{s}$ was used in all measurements. The energy spectra were recorded using a Tukan PC-based multichannel analyzer (MCA) [10].

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each gamma peak, was measured by Bertolaccini method [11,12]. In this method the number of photoelectrons is 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.

Results and Discussion

Energy Spectra and Photoelectron Yield. Fig. 1 presents a comparison of the energy spectra for 662 keV gamma rays from a ^{137}Cs source measured with $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals. The energy resolution of 7.9% obtain for $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is better than that of 9.1% for BGO. The energy resolution for the tested BGO crystal in this study is better than the value of 10.0 % observed by Moszynski et al.[8] for the BGO crystal ($\varnothing 9 \times 4 \text{ mm}^3$) supplied by Saint-Gobain. Note a higher photofraction in the spectrum measured with BGO, as would be expected due to a higher effective atomic number of the BGO crystal.

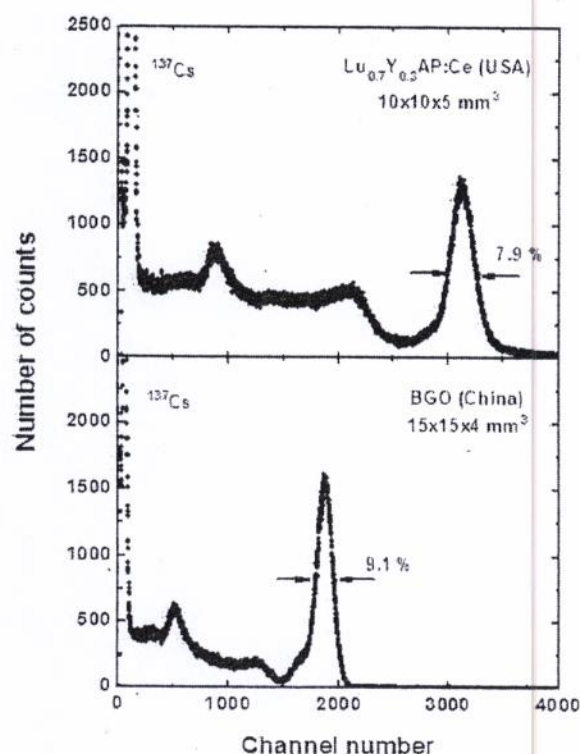


Fig. 1 Energy spectra of 662 keV gamma rays from a ^{137}Cs source measured with $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals.

Table 1 summarizes comparative measurements of photoelectron yield and energy resolution at 662 keV gamma rays for the tested crystals. The $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ showed a photoelectron yield of 3,950 phe/MeV. This value corresponds to about 13,400 photons/MeV (ph/MeV) at the PMT photocathode quantum efficiency (QE) of 29.4% for peak emission at 375 nm. The tested BGO showed a photoelectron yield of 2,390 phe/MeV. This value corresponds to about 11,600 ph/MeV at a QE of 20.7% for peak emission at 480 nm. A high light output of tested BGO seems to be not correct, compared with the announced value between 8,000 - 9,000 ph/MeV. In the light of the study of modern PMTs, the unexpected excess of photoelectrons can be measured with XP5200 PMT, reflected later on by the excess of photons, reported recently in Ref.[13]. Note the light yield of 13,400 ph/MeV for $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is higher than the value of 8,530 ph/MeV measured with small sample ($2 \times 2 \times 10 \text{ mm}^3$) in Ref.[14].

Table 1 Light yield and energy resolution at 662 keV gamma rays for tested crystals

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	$\Delta E/E$ [%]
$\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$	$3,950 \pm 400$	$13,400 \pm 1,300$	7.9 ± 0.4
BGO	$2,390 \pm 200$	$11,600 \pm 1,200$	9.1 ± 0.5

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 [4]. 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. 2 presents the non-proportionality characteristics of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals. $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is clearly superior to BGO in terms of light yield proportionality. Over the energy range from 22.1 to 1,274.5 keV, the non-proportionality is about 20 % for $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$, which is better than that of about 39 % for BGO. The higher proportionality of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ should be reflected in its better intrinsic resolution, see below.

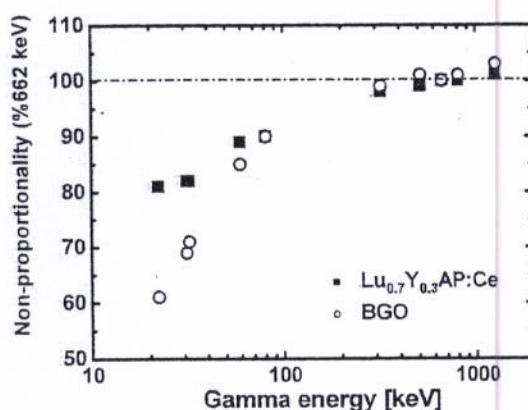


Fig. 2 Non-proportionality in the light yield of BGO and $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ crystals. Error bars are within the size of the points.

Energy Resolution. The energy resolution ($\Delta E/E$) of a full energy peak measured with a scintillator coupled to a PMT can be written as [5]

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

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.

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 [5].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [5] 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

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

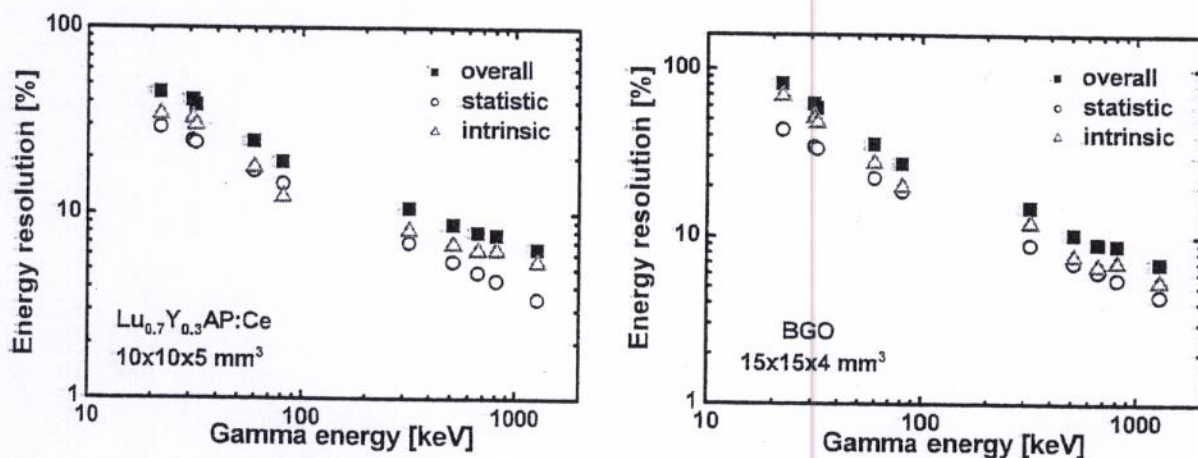


Fig. 3 Energy resolution and contributed factors versus energy of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals. Error bars are within the size of the points.

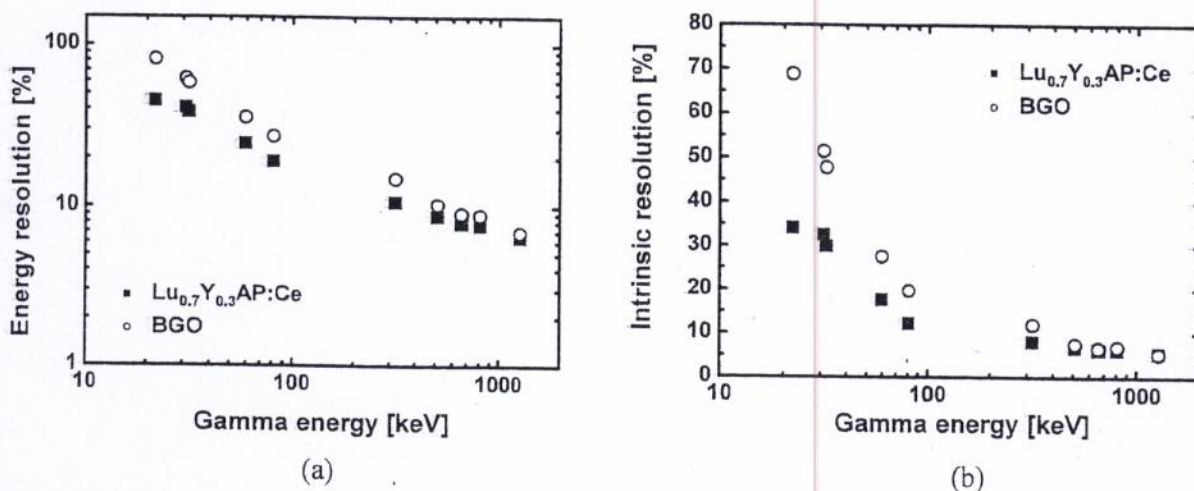


Fig. 4 Overall energy resolution and the intrinsic resolution of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals. Error bars are within the size of the points.

Fig.3 presents the measured energy resolution versus energy of gamma rays for $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals. 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).

Overall energy resolution and the intrinsic resolution of both crystals versus energy are shown in Fig.4 (a) and (b), respectively. The energy resolution for tested crystals is approximately inversely proportional to the square root of the energy. The energy resolution of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is better than that of BGO over the energy range from 22 to 810 keV. The intrinsic resolution of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is better than that of BGO in the energy range below 320 keV, which is, reflected by a better proportionality of its light yield (see Fig. 2).

To better understand the energy resolution of 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 $\Delta E/E$, the overall energy resolution at 662 keV photopeak. The PMT contribution (δ_{st}) was calculated using Eq.(2). From the values of $\Delta E/E$ and δ_{st} , the intrinsic resolution (δ_{sc}) was calculated using Eq.(3).

Table 2 Analysis of the 662 keV energy resolution for BGO and $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ detectors

Detector	N [electrons]	$\Delta E/E$ [%]	δ_{st} [%]	δ_{sc} [%]
$\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce} + \text{XP5200B}$	$2,620 \pm 260$	7.9 ± 0.4	4.8 ± 0.2	6.3 ± 0.3
BGO + XP5200B	$1,580 \pm 160$	9.1 ± 0.5	6.2 ± 0.3	6.6 ± 0.3

The photoelectron yield (and δ_{st}) of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is clearly superior to BGO. However, there is a little progress in energy resolution for $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ which due to a large contribution of its intrinsic resolution to the overall energy resolution.

Photofraction. 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 $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO 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 BGO shows much higher photofraction than $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ in a same trend with the cross-section ratio (σ -ratio) obtained from WinXCom program. The reason is due to much higher effective atomic number and volume of the BGO crystal.

Table 3 Photofraction at 662 keV gamma peak for BGO and $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ crystals

Crystal	Z_{eff}	Volume (cm^3)	Photofraction (%)	σ - ratio (%)
$\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$	60	0.5	23.6 ± 2.4	20.5
BGO	74	0.9	42.9 ± 4.3	30.8

Summary

In this work, the scintillation properties of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ and BGO crystals were studied and compared in gamma ray spectrometry. $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ showed the light yield of about 13,400 ph/MeV is better than that of about 11,600 ph/MeV measured for BGO. For 662 keV gamma rays, an energy resolution of 7.9 % obtained for $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is better than that of 9.1% obtained for BGO. The energy resolution of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ is better than that of BGO for energy lower than 320 keV due to a higher photoelectron yield and lower intrinsic resolution. The photofraction of BGO is superior to that of $\text{Lu}_{0.7}\text{Y}_{0.3}\text{AP:Ce}$ due to much higher effective atomic number and volume of the BGO crystal.

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