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Scintillation Properties of $Lu_3Al_5O_{12}$, Lu_2SiO_5 and $LaBr_3$ Crystals Activated with Cerium

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Abstract

The performances of Ce-activated $Lu_3Al_5O_{12}$ (LuAG:Ce), Lu_2SiO_5 (LSO:Ce) and $LaBr_3$ ($LaBr_3$:Ce) scintillator crystals were investigated for γ-ray detection. The light yield and energy resolution were measured using photomultiplier tube (XP5500B PMT) readout. For 662 keV (^{137}Cs source), an energy resolution of 3.5% obtained for $LaBr_3$:Ce is much better than that of 6.7% and 8.3%, respectively, for LuAG:Ce and LSO:Ce. The light yield non-proportionality and energy resolution versus energy of γ-rays were measured and the intrinsic resolution of the crystals was determined. The $LaBr_3$:Ce exhibits a good proportionality within 7% deviation from unity at 16.6 keV, which is much better than that of 22% and 45%, respectively, for LuAG:Ce and LSO:Ce. The photofraction was determined at 320, 662 and 835 keV for studied crystals and compared with the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCom program.

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1. Introduction

Research and development of new scintillator materials is mainly triggered by the rapidly growing needs of medical imaging and high energy physics. During the last two decades, new types of scintillators, in particular, Ce-doped inorganic scintillators were intensively studied and some of them were successfully developed for commercial production, for recent reviews see [1- 4].

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$Y_3Al_5O_{12}$:Ce (YAG:Ce) single crystal was reported in the literature as a fast oxide scintillator [5,6]. Isostructural $Lu_3Al_5O_{12}$:Ce (LuAG:Ce) has a higher density (6.67 g/cm^3) than YAG:Ce (4.56 g/cm^3), which is advantageous in the case of high energy gamma-ray detection [7,8]. Its emission spectrum at room temperature (RT) is peaked around 525 nm. The scintillation light yield within 1 μs time gate is about 12,500 ph/MeV and 22,000 ph/MeV, respectively, for LuAG:Ce and YAG:Ce crystals [9].

Lu_2SiO_5:Ce (LSO:Ce) [10] and $(Lu,Y)_2SiO_5$:Ce (LYSO:Ce) [11,12] have been developed as promising scintillators for positron emission tomography (PET) due to their desirable properties such as high density, fast decay time and high light output. LSO:Ce has a density of 7.4 g/cm^3 and an emission spectrum at RT is peaked around 410 nm. LSO:Ce exhibits a high light yield up to about 30,000 ph/MeV [13,14].

New Ce-doped $LaCl_3$ [15] and $LaBr_3$ [16] scintillators appeared with attractive properties due to high light output and very good energy resolution. $LaBr_3$:Ce has a density of 5.29 g/cm^3 and an emission spectrum at RT is peaked around 370 nm. $LaBr_3$:Ce exhibits a very high light yield above 60,000 ph/MeV and an excellent energy resolution of about 3% for 662 keV γ-rays. The excellent energy resolution of $LaBr_3$:Ce crystal is confirmed by its good proportional scintillation response and corresponding excellent intrinsic resolution [17].

In this paper, we report on the detection properties of γ-rays for LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals covering energies from 16.6 to 1274.5 keV. The light yield non-proportionality and energy resolution versus energy of γ- rays were measured and the intrinsic resolution of the crystals was determined after correcting the measured energy resolution for PMT statistics. The estimated photofraction in the pulse height spectra of 320, 662 and 835 keV γ-rays was determined for studied crystals and compared with the ratio of the cross-sections for the photoelectric effect to the total one calculated using WinXCOM program.

2. Experimental procedures

The uAG:Ce and LSO:Ce crystals with size of 10×10×5 mm^3 were supplied by Crytur Ltd(Czech Republic) and CTI (USA), respectively. The $LaBr_3$:Ce crystal encapsulated in an aluminum can with size of ∅13×13 mm^2 was supplied by Saint-Gobain (France).

Photoelectron yield and energy resolution were measured by coupling the crystals to a Photonis XP5500B PMT using silicone grease. In order to maximize light collection, the crystals were covered with several layers of white Teflon tape in a configuration of a reflective umbrella. The signal from the PMT anode was passed to an ORTEC 113 preamplifier and then to a Tennelec TC244 spectroscopy amplifier. The measurements were carried out with 3 μs shaping time constant in the amplifier. The PC-based multichannel analyzer (MCA), Tukan 8k [18] was used to record energy spectra.

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each γ-peak, was measured by Bertolaccini method [19,20]. In this method the number of photoelectrons is measured by comparing the position of a full energy peak of γ-rays detected in the crystals with that of the single photoelectron peak from the PMT photocathode. The measurements of light yield non-proportionality and energy resolution were carried out for a series of X/γ-rays emitted by different radioactive sources in the energy range from 16.6 to 1274.5 keV.

3. Results and discussion

3.1 Light yield and energy resolution

Fig. 1 presents the pulse height spectra of 662 keV γ-rays from a ^{137}Cs source as measured with LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals at RT. The energy resolution of 3.5% obtained with $LaBr_3$:Ce is superior compared to the value of 6.7% and 8.3% , respectively, obtained with LuAG:Ce and LSO:Ce. This is due to much higher photoelectron yield and very good proportionality of light yield for $LaBr_3$:Ce, see below. Note a higher photofraction in the spectrum obtained with LSO:Ce, as would be expected due to higher effective atomic number and density of the LSO:Ce material.

Photoelectron yield was determined using 662 keV γ-rays from a ^{137}Cs source. $LaBr_3$:Ce exhibits the photoelectron yield of 12,320 phe/MeV, which is much larger than that of 9,990 ph/MeV and 3,730 phe/MeV, respectively, for LSO:Ce and LuAG:Ce. The number of photoelectrons measured for studied crystals was recalculated to the number of photons assuming the quantum efficiency of 29%, 33%, and 18%, respectively, for the XP5500B PMT at the peak emission of $LaBr_3$:Ce (370 nm), LSO:Ce (410 nm) and LuAG:Ce (525 nm). The light yield of about 42,500 ph/MeV, 30,300 ph/MeV and 20,700 ph/MeV was obtained, respectively, for $LaBr_3$:Ce , LSO:Ce and LuAG:Ce. The results summarizing the photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals are presented in Table 1. Despite a much higher photoelectron yield, LSO:Ce shows much worse energy resolution with respect to LuAG:Ce. The reason is a much higher contribution of intrinsic resolution for LSO:Ce, see below.

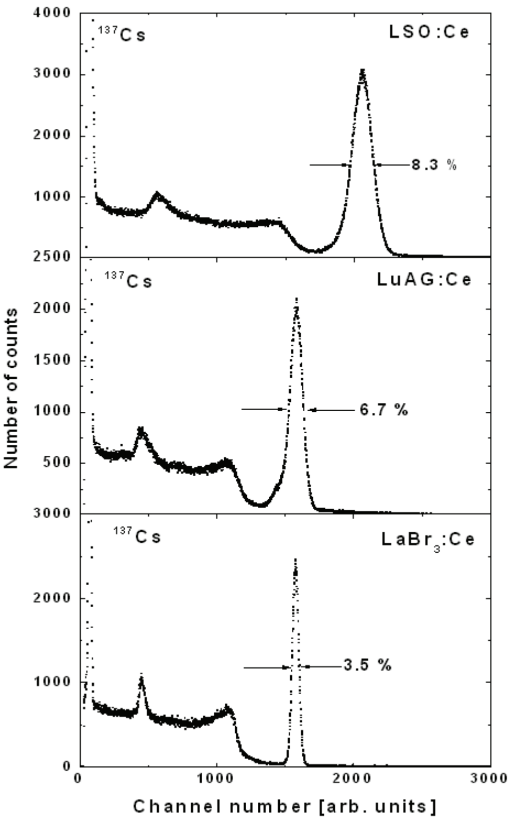

Fig. 1. Pulse height spectra of 662 keV γ - rays from a ^{137}Cs source as measured with LSO:Ce, LuAG:Ce and $LaBr_3$:Ce crystals.

Table 1. Photoelectron yield, light yield and energy resolution at 662 keV γ-rays for the studied crystals as measured with the XP5500B PMT.

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	Energy resolution [%]
LuAG:Ce	3,730 ± 200	20,700 ± 2000	6.7 ± 0.3
LSO:Ce	9,990 ± 500	30,300 ± 3000	8.3 ± 0.3
$LaBr_3$:Ce	12,320 ± 600	42,500 ± 4000	3.5 ± 0.1

The energy resolution (ΔE/E) of a full energy peak measured with a scintillator coupled to a photomultiplier can be written as [21]

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

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 energy 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 XP5500B 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 [21].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [21,22] 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 presents the overall energy resolution (ΔE/E) as a function of γ-ray energy, measured for the studied crystals. Over the energy range from 16.6 to 1274.5 keV, the overall energy resolution of $LaBr_3$:Ce is much better than that of LuAG:Ce and LSO:Ce. Fig.3 presents a direct comparison of the intrinsic resolution for the studied crystals.

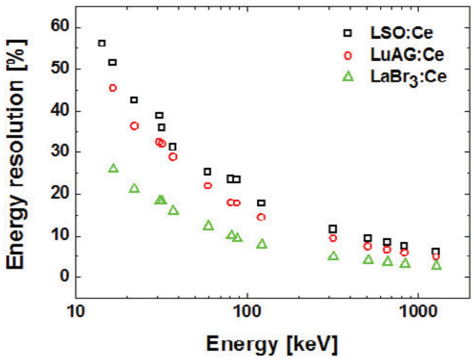

Fig. 2. Overall energy resolution of LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals.

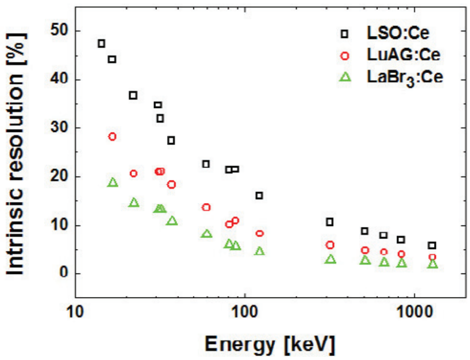

Fig. 3. Intrinsic resolution of LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals

To better understand the energy resolution of the studied crystals in γ-ray spectrometry, the contribution of various components to the overall energy resolution were 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 (2). From the values of ΔE/E and δ_{st} , the intrinsic resolution (δ_{sc}) was calculated using (3). Excellent energy resolution of $LaBr_3$:Ce is most likely associated with a lowest statistical error in the number of photoelectrons (δ_{st}) as well as a lowest contribution of intrinsic resolution (δ_{sc}). A poor energy resolution for LSO:Ce is mainly due to a very high contribution of δ_{sc} . This result suggested looking at the non-proportionality of light yield versus energy of γ-rays, as the non-proportionality of light yield is a fundamental limitation to δ_{sc} of the scintillators [21,22].

Table 2. The 662 keV energy resolution for the studied crystals coupled to the XP5500B PMT.

crystal	N [electrons]	ΔE/E [%]	δ_{st} [%]	δ_{sc} [%]
$LaBr_3$:Ce	8160	3.5	2.7	2.2
LuAG:Ce	2470	6.7	5.0	4.5
LSO:Ce	6610	8.3	3.0	7.7

3.2 Non-proportionality of the light yield

Non-proportionality of light yield is defined as the ratio of light yield measured at specific γ-ray energies relative to the light yield at the 662 keV γ-peak. Fig. 4 presents a comparison of the non-proportionality characteristics measured for all studied crystals. The most proportional scintillation response is obtained for $LaBr_3$:Ce with its non-proportionality only about 7% deviation from unity at 16.6 keV, which is much better than that of about 22% and 45%, respectively, for LuAG:Ce and LSO:Ce. The highest proportionality of the light yield for $LaBr_3$:Ce is related to a lowest contribution of δ_{sc} ,see Fig.3. This result confirms that the intrinsic resolution of a scintillator is mainly associated with the non-proportional scintillation response [21,22].

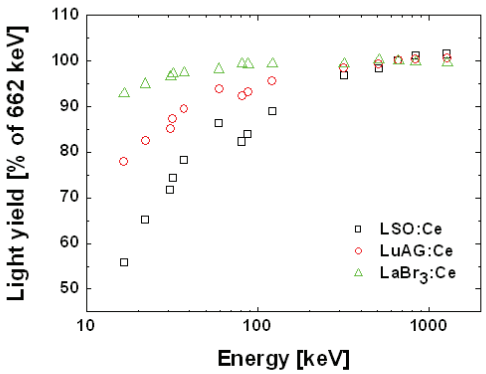

Fig. 4. Non-proportionality of light yield as a function of γ-ray energy for LuAG:Ce, LSO:Ce and $LaBr_3$:Ce crystals.

It is well known that the scintillation properties of Ce-doped rare-earth oxyorthosilicates are controlled by two distinct luminescence centers, known as Ce1 and Ce2 [23,24]. The presence of two competing scintillation processes is believed to be detrimental to the energy resolution of Ce-doped crystals [25]. In the recent study, the observed correlation of the intrinsic resolution of the LGSO:Ce crystals and the intensity of their afterglow suggested that the energy resolution of scintillation detectors may be affected also by a strong afterglow of the crystals [26]. It seems to suggest that the statistical spread of the population of the primary light and that of afterglow light [26] together with the existence of two competing scintillation processes in Ce-doped crystals [25] contribute to inhomogeneities of the scintillation efficiency across the crystal. It is well known that LSO:Ce exhibits a strong aftergrow [14]. Thus, the high δ_{sc} value for LSO:Ce, which is associated mainly with the non-proportionality in its light yield, can be affected also by inhomogeneities of the scintillation efficiency in this scintillator.

The main component of energy resolution for LuAG:Ce detector is δ_{st} , which is due to a rather low light output of this scintillator together with a low number of photoelectrons as measured with a blue-sensitive XP5500B PMT, which mismatch to the peak emission at 525 nm of LuAG:Ce. An improvement in the energy resolution for LuAG:Ce detector could be obtained by coupling to a higher green-sensitive photodetector such as Si-avalanche photodiode.

3.3 Photofraction

The photofraction is defined here as the ratio of counts under the photopeak to the total counts of the pulse height spectrum as measured at a specific γ-ray energy. The photofraction for LSO:Ce, LuAG:Ce and $LaBr_3$:Ce at 320, 662 and 835 keV γ-rays is collected in Table 3. For a comparison, the ratio of the cross-sections (σ-ratio) for the photoelectric effect to the total one calculated using the WinXCom program [27] is also given. The LSO:Ce exhibits higher photofraction than LuAG:Ce and $LaBr_3$:Ce in a similar trend as the σ-ratio obtained from the WinXCom program. The reason is due to higher effective atomic number (Z_{eff} = 66) and density (ρ = 7.4 g/cm^3) of the LSO:Ce with respect to those of LuAG:Ce (Z_{eff} = 58.9; ρ = 6.67 g/cm^3) and $LaBr_3$:Ce (Z_{eff} = 46.9; ρ = 5.29 g/cm^3). However, the measured photofractions for both LSO:Ce and LuAG:Ce crystals are closer to the σ-ratios than the values for $LaBr_3$:Ce. It may be due to a larger size (a factor of 2.5) of the studied $LaBr_3$:Ce sample.

Table 3. Photofractions for $LaBr_3$:Ce, LuAG:Ce and LSO:Ce crystals

γ energy (keV) Source	320 ^{51}Cr	662 ^{137}Cs	835 ^{54}Mn	
Photof. (%)	44.7	16.2	12.7	$LaBr_3$:Ce
σ- ratio (%)	31.3	8.7	5.9	
Photof. (%)	64.8	28.0	22.3	LuAG:Ce
σ- ratio (%)	52.8	19.9	14.2	
Photof. (%)	70.9	35.8	27.5	LSO:Ce
σ- ratio (%)	58.8	24.0	17.4	

4. Conclusions

The performances among Ce-activated $LaBr_3$, LuAG and LSO scintillators were investigated and compared in γ-ray spectrometry. The high energy resolution of 3.5% for 662 keV γ-rays obtained with $LaBr_3$:Ce is much better than the values of 6.7% and 8.3% obtained, respectively, for LuAG:Ce and LSO:Ce. The high light output and very good proportionality of $LaBr_3$:Ce are the important reasons behind its high energy resolution. It has a potential to replace NaI:Tl as the scintillator of choice for SPECT camera and γ-ray spectrometry. $LaBr_3$:Ce appears to be promising for PET, but a relatively low density and photofraction make it less attractive than LSO:Ce and LYSO:Ce.

The main advantages of LSO:Ce are high light yield and detection efficiency for γ-rays. This fact and together with the fast scintillation decay (~40 ns) make it an excellent scintillator for PET imaging.

An advantage of LuAG:Ce is its superior energy resolution with respect to LSO:Ce. A drawback of LuAG:Ce is its very intense slow component in the scintillation pulse [28,29], which is due to retrapping of charge carriers at shallow traps and appearance of the delayed radiative recombination at the Ce^{3+}-emission centers. It points to a chance to enhance its scintillation intensity of fast component determining both the energy and time resolutions, if related shallow traps could be suppressed. This fact together with the considerably fast scintillation decay (~60 ns) and moderate detection efficiency for γ-rays, would make LuAG:Ce the material of choice for γ-ray spectrometry and PET imaging.

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