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Advanced Materials Research Vols. 199-200 (2011) pp 1789-1795
Online available since 2011/Feb/21 at www.scientific.net
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doi:10.4028/www.scientific.net/AMR.199-200.1789

Luminescence and Scintillation Properties of Ce-Doped YAP and LuYAP Crystals

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Keywords: Energy Resolution, Luminescene, Luyap:Ce, Scintillation, YAP:Ce

Abstract. The luminescence and gamma-ray detection properties of cerium-doped scintillators, namely, $Lu_{0.3}Y_{0.7}AlO_3$:Ce ($Lu_{0.3}Y_{0.7}$AP:Ce), $Lu_{0.7}Y_{0.3}AlO_3$:Ce ($Lu_{0.7}Y_{0.3}$AP:Ce), and $YAlO_3$:Ce (YAP:Ce) were investigated. UV excitation and emission spectra of studied crystals were compared. The light yield and energy resolution were measured using photomultiplier tube (PMT) readout. The light yield non-proportionality and energy resolution versus gamma-ray energy were measured and the intrinsic resolution of the crystals was determined after correcting the measured energy resolution for PMT statistics. For 662 keV gamma rays (^{137}Cs source), YAP: Ce showed the highest light yield of 28,500 ph/MeV and the best energy resolution of 4.4 % , while its photofraction is worst.

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, good energy resolution, good proportionality of light yield, minimal afterglow and low production costs. Good reviews on development of inorganic scintillation detectors/systems have been published by Moszynski [1], van Eijk [2], and recently by Lecoq et al.[3].

The last decade has been the introduction of several new scintillators for medical imaging, in particular cerium-doped crystals as YAP:Ce or LSO:Ce exhibit better luminescence and scintillation properties compared to the classical ones {BGO, NaI(Tl), CsI(Tl)}. During last years many efforts were devoted to the development of heavy scintillators based on cerium-doped crystals, especially, crystals of perovskite type (Lu_xY_{1-x}AP:Ce) were developed for x = 0-1.

In this paper, we present the luminescence and gamma-ray detection properties of $Lu_{0.3}Y_{0.7}$AP:Ce, $Lu_{0.7}Y_{0.3}$AP:Ce and YAP:Ce. The photoelectron yield and energy resolution at 662 keV were measured. The light yield non-proportionality and energy resolution as a function of gamma-ray energy were measured, and the intrinsic resolution was determined. The estimated photofraction for crystals at 662 keV gamma rays will also be discussed.

Methodology

The $Lu_{0.7}Y_{0.3}$AP:Ce crystal with size of 10x10x5 mm^3 was supplied by Opto Materials S.r.l. (Italy). The $Lu_{0.3}Y_{0.7}$AP:Ce crystal with size of 5x5x9 mm^3 and the YAP:Ce crystal with size of 10x10x5 mm^3 were supplied by Crytur Ltd.(Czech republic).

UV excitation and emission spectra were obtained using a Hitachi F2500 fluorescence spectrophotometer. The measurements of the emission spectra were made using excitation wavelengths corresponding to the excitation bands found for the studied crystals. All measurements were performed at room temperature.

Each crystal was optically coupled to a Photonis XP5200B PMT using silicone grease. 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 TC244 spectroscopy amplifier. A shaping time constant of 4 μs was used with studied crystals. The energy spectra were recorded using a Tukan PC-based multichannel analyzer (MCA) [4].

The photoelectron yield, expressed as a number of photoelectrons per MeV (phe/MeV) for each gamma-peak, was measured by Bertolaccini method [5,6]. 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.

The measurements of photoelectron yield and energy resolution were carried out for a series of gamma rays emitted by different radioactive sources 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.

Results and Discussion

UV Excitation and Emission Spectra. YAP:Ce and $Lu_{0.7}Y_{0.3}AP$:Ce luminescence spectra are shown in Figs.1 (a) and (b), respectively. The spectra are normalized to the maximum peak for a better comparison. Emission and excitation spectra of both crystals were measured in near-UV and visible range at room temperature. Excitation spectra consist of broad absorption bands between 220 and 350 nm attributed to the optical transitions from Ce^{3+} 4f-ground state to the crystal field split 5d-excited levels. Broad band emission spectra of both crystals are roughly the same within 320 – 440 nm range. It can be seen that the overlap of excitation and emission bands is slightly larger for $Lu_{0.7}Y_{0.3}AP$:Ce than YAP:Ce. This should affect the absorption of emitted light, especially for thick sample.

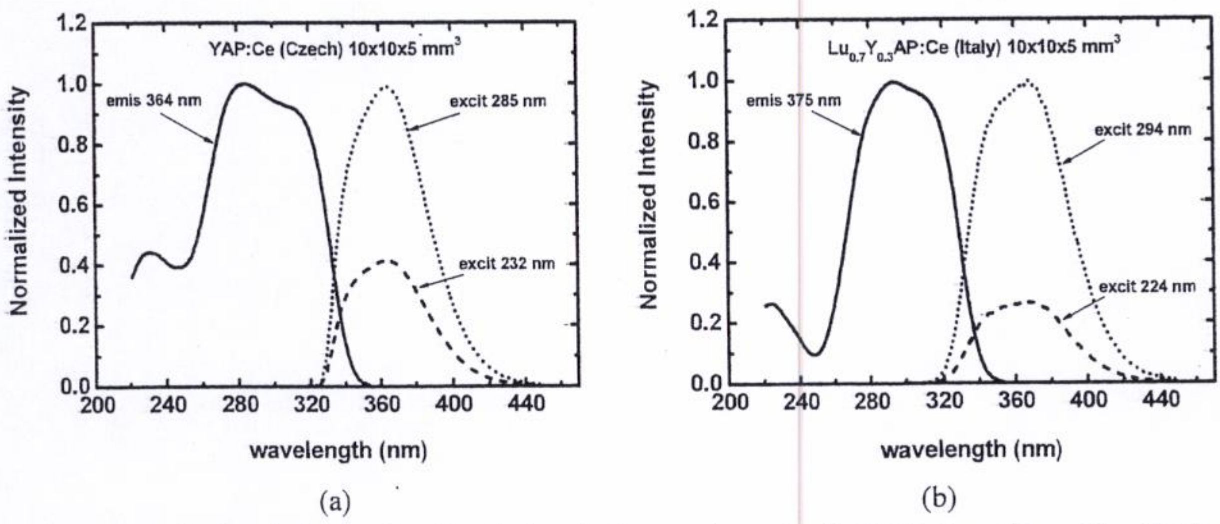

Fig. 1 UV excitation (left curve) and emission (right curve) spectra for YAP:Ce and $Lu_{0.7}Y_{0.3}AP$:Ce crystals.

Photoelectron Yield and Energy Resolution. Fig. 2 presents the energy spectra of 662 keV gamma rays from a ^{137}Cs source measured with each detector. It is seen that YAP:Ce gives better energy resolution than $Lu_{0.3}Y_{0.7}AP$:Ce and $Lu_{0.7}Y_{0.3}AP$:Ce. The energy resolution of 4.4% obtained with YAP:Ce is much better than the value of 9.2 and 9.0 % obtained with $Lu_{0.3}Y_{0.7}AP$:Ce and $Lu_{0.7}Y_{0.3}AP$:Ce, respectively. The energy resolution of 4.4% for the studied YAP:Ce crystal is much better than the value of 5.7% observed by Moszynski et al.[7] for an equal sized YAP:Ce crystal supplied by Preciosa Co.(Turnov, Czech Republic). Note a higher photofraction in the spectrum measured with LuYAP:Ce, as would be expected due to a higher effective atomic number and density of the LuYAP:Ce crystal.

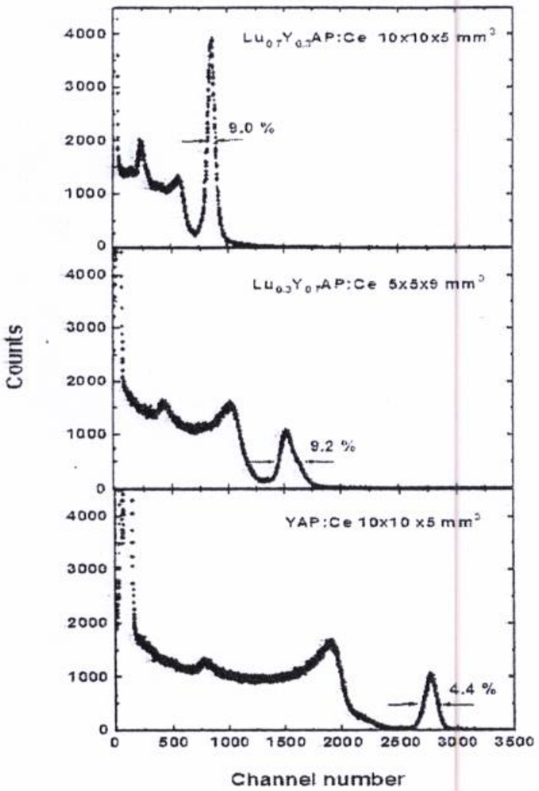

Fig. 2 Energy spectra of 662 keV gamma rays from a [137]Cs source measured withYAP:Ce, and LuYAP:Ce crystals.

Table 1 Photoelectron yield, light yield and energy resolution at 662 keV gamma rays for the studied crystals as measured with the Photonis XP5200B PMT

Crystal	Photoelectron yield [phe/MeV]	Light yield [ph/MeV]	Energy resolution [%]
YAP:Ce	7,560 ± 700	28,500 ± 2,900	4.4 ± 0.2
$Lu_{0.3}Y_{0.7}AP$:Ce	4,010 ± 400	15,400 ± 1,500	9.2 ± 0.4
$Lu_{0.7}Y_{0.3}AP$:Ce	2,250 ± 200	8,700 ± 900	9.0 ± 0.4

Table 1 summarizes comparative measurements of photoelectron yield, light yield and energy resolution at 662 keV gamma rays for the studied crystals coupled to the Photonis XP5200B PMT, as measured at 4 μs shaping time constant in the spectroscopy amplifier. The YAP: Ce showed a photoelectron yield of 7,560 phe/MeV. This value corresponds to about 28,500 Photon/MeV (ph/MeV) at the PMT photocathode quantum efficiency (QE) of 26.5% for peak emission of 360 nm. The studied $Lu_{0.7}Y_{0.3}AP$:Ce and $Lu_{0.3}Y_{0.7}AP$:Ce showed a photoelectron yield of 2,250 and 4,010 phe/MeV, respectively. These values correspond to 8,700 and 15,400 ph/MeV, respectively, for $Lu_{0.7}Y_{0.3}AP$:Ce and $Lu_{0.3}Y_{0.7}AP$:Ce, at a QE of 26% for peak emission of about 375 nm. Note a significantly higher light yield of 28,500 ph/MeV for the studied YAP:Ce crystal, by about 170 % compared with a same sized sample in Ref [7]. The studied $Lu_{0.7}Y_{0.3}AP$:Ce showed the light yield of 8,700 ph/MeV which is slightly higher than the value of 8,530 ph/MeV measured with small sample (2x2x10 mm^3) in Ref [8].

Non-proportionality of 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 [9]. 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.

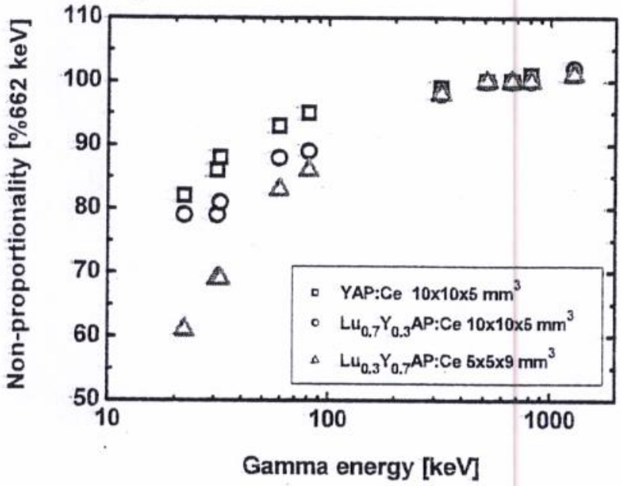

Fig. 3 Non-proportionality in the light yield of YAP:Ce and LuYAP:Ce crystals. Error bars are within the size of the points.

Fig. 3 presents the non-proportionality characteristics of YAP:Ce and LuYAP:Ce crystals. All crystals exhibit different non-proportionality curves. YAP:Ce is clearly superior to both LuYAP:Ce in terms of light yield proportionality. Over the energy range from 22.1 to 1,274.5 keV, the non-proportionality is about 18% for YAP:Ce, which is better than that of about 21 and 39 %, respectively, for $Lu_{0.7}Y_{0.3}AP$:Ce and $Lu_{0.3}Y_{0.7}AP$:Ce. The higher proportionality of YAP:Ce should be reflected in its better intrinsic resolution.

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 [1]

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

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 [1].

The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [1,9] 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

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

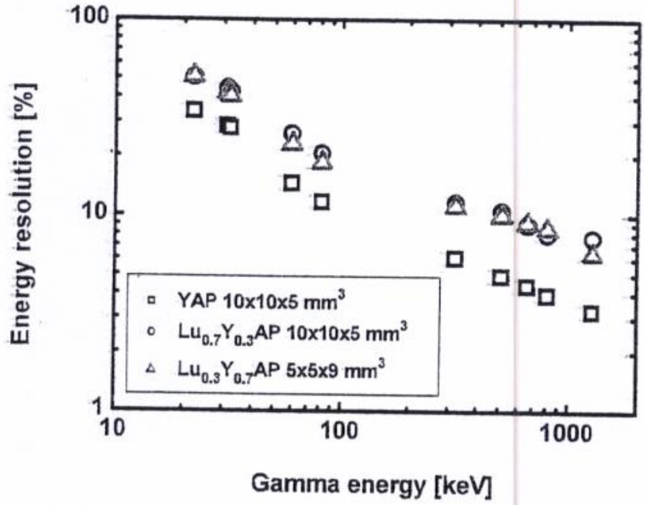

Fig. 4 Overall energy resolution of YAP:Ce and LuYAP:Ce crystals versus energy of gamma rays. Error bars are within the size of the points.

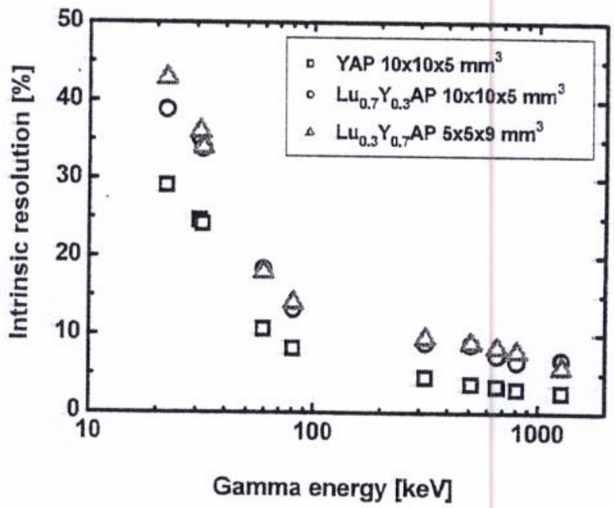

Fig. 5 Intrinsic resolution of YAP:Ce and LuYAP:Ce crystals versus energy of gamma rays. Error bars are within the size of the points.

Overall energy resolution of YAP:Ce and LuYAP:Ce detectors versus energy is shown in Fig.4. The energy resolution for all crystals is approximately inversely proportional to the square root of the energy. The energy resolution of YAP:Ce is superior than that of both LuYAP:Ce over the whole energy range from 22.1 to 1,274.5 keV.

Fig. 5 presents a direct comparison of the intrinsic resolution for the studied crystals. The intrinsic resolution of YAP:Ce crystal is better (almost a factor of two at high energies) than that of both LuYAP:Ce crystals, reflected by a better proportionality of the light yield (see Fig. 3).

To better understand the energy resolution of the studied 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 YAP:Ce and LuYAP:Ce crystals

Detector	N [electrons]	ΔE/E [%]	δ_{st} [%]	δ_{sc} [%]
YAP:Ce	5,000 ± 500	4.4 ± 0.2	2.9 ± 0.1	3.3 ± 0.1
$Lu_{0.3}Y_{0.7}AP$:Ce	2,650 ± 300	9.2 ± 0.4	3.9 ± 0.2	8.3 ± 0.3
$Lu_{0.7}Y_{0.3}AP$:Ce	1,490 ± 150	9.0 ± 0.4	5.2 ± 0.2	5.2 ± 0.2

The superior energy resolution of YAP:Ce as compared to both LuYAP:Ce 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 YAP:Ce crystal.

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 YAP:Ce and both LuYAP:Ce 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 [10] are given too. The data indicate that $Lu_{0.7}Y_{0.3}AP$:Ce shows much higher photofraction than $Lu_{0.3}Y_{0.7}AP$:Ce and YAP: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 density of the $Lu_{0.7}Y_{0.3}AP$:Ce crystal.

Table 3 Photofraction at 662 keV gamma peak for YAP:Ce and LuYAP:Ce crystals

Crystal	Z_{eff}	ρ (g/cm^3)	Photofraction (%)	σ- ratio (%)
YAP:Ce	34	5.4	5.3 ± 0.5	2.6
$Lu_{0.3}Y_{0.7}AP$:Ce	53	6.2	11.1 ± 1.1	10.9
$Lu_{0.7}Y_{0.3}AP$:Ce	60	7.1	27.5 ± 2.8	20.5

Summary

The luminescence and scintillation properties of YAP:Ce, $Lu_{0.7}Y_{0.3}AP$:Ce, and $Lu_{0.3}Y_{0.7}AP$:Ce crystals were investigated. Emission and excitation spectra of studied crystals are roughly the same with overlapping in the range 320-350 nm. This could affect their scintillation efficiency. The energy resolution of YAP:Ce is superior than that of both LuYAP:Ce 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 non-proportional response of the scintillator is correlated with the intrinsic resolution of the scintillators. Moreover, inhomogeneities of dopant and some defects in crystals could affect the energy resolution, and the crystalline quality of these samples could be further improved. $Lu_{0.7}Y_{0.3}AP$:Ce showed much higher photofraction than $Lu_{0.3}Y_{0.7}AP$:Ce and YAP:Ce due to its much higher effective atomic number and density.

Acknowledgement

This work was supported by King Mongkut's University of Technology Thonburi, Rajamangala University of Technology Thanyaburi, and the Office of the Higher Education Commission, Ministry of Education.

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