INITIAL RESULTS FOR THE DEVELOPMENT OF A SMALL FIELD OF VIEW GAMMA-RAY IMAGER USING A LUAG:Pr SCINTILLATOR

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Abstract. In this study we present the initial results for the construction and the evaluation of a small field of view gamma ray detector based on a R8900U-00-C12 position sensitive photomultiplier tube (PSPMT) coupled to a 2x2 LuAG:Pr scintillator array with 5x5x5mm³ crystal size elements. We have used a resistor network readout circuit, reducing the 6X and 6Y photomultiplier anode signals to 4 position signals (Xa, Xb, Yc and Yd). A Field Programmable Gate Array (FPGA) Spartan 6 LX16 was used for triggering and signal processing of the signal pulses acquired using a free running sampling technique. We have obtained raw images and energy histograms, under various gamma ray energies irradiation. Energy resolution and linearity of the detector response under various gamma ray energies ranging from 81 to 1332.5 keV were measured. The system presented an excellent performance in terms of energy resolution, measured 9.3%@511KeV and 8.5%@662keV.

1 INTRODUCTION

Single crystal scintillators coupled to photodetectors have been used in many fields such as high energy physics, astro physics, medical imaging, and sources detection ^[1]. Depending on the type of application different requirements in the components are set. For example, medical imaging requires mainly scintillators with high stopping power, high light yield, and fast decay ^[2]. Cerium doped inorganic single crystal scintillators such as LSO:Ce, LYSO:Ce, LuAP:Ce, LuYAP:Ce, GSO:Ce, YAP:Ce, LaBr₃;Ce and LaCl₃:Ce have already been used or suggested for use in clinical Positron Emission Tomography (PET), animal PET and other nuclear medicine or combined modalities ^[3-5]. Up to now, Ce³⁺ doped materials have been well studied to obtain a very fast and efficient emission center. Ce³⁺ emission centers in these complex oxide hosts are characterized by (i) broad $5d_1 \rightarrow 4f$ emission peaking mainly in the near UV-visible spectral range and (ii) fast decays in the range of tens of nanoseconds. Following few previous studies ^[6-7] the interest in Pr³⁺ doped hosts was renewed due to "faster-than-cerium" decay characteristics based on the same $5d_1 \rightarrow 4f$ emission transitions in UV spectral region ^[8-11].

The production of Pr^{3+} doped aluminum garnets, $Y_3Al_5O_{12}$ (YAG) and $Lu_3Al_5O_{12}$ (LuAG), prepared by the Czochralski method in bulk single crystal form have been reported ^[12]. The latter single crystal (LuAG:Pr) is characterized by a very fast decay time (20 ns) and good light yield (53% of NaI:Tl). LuAG:Pr has a shorter emission wavelength at 310 nm than Ce³⁺ doped scintillators, which typically emit at wavelengths above 400nm. The density of LuAG is 6.67 g/cm³ and its effective atomic number is 59 ^[13]. Table 1 shows a comparison between LuAG:Pr and some popular cerium-activated scintillators such as LSO:Ce, LYSO:Ce, LuYAP:Ce, GSO:Ce, YAP:Ce, YAG:Ce, LaBr₃;Ce and LaCl₃:Ce. LuAG:Pr was discovered in 2005 when its photo- and radio-luminescence properties were studied ^[14]. This specific material in scintillator array form coupled with APD and SiPM arrays has been used in Positron Emission Mammography and combined MRI-PET/ToF-PET detectors ^[15-16].

Scintillator crystal	Density	ρZ^4_{eff}	Hygro-	Light yield	Decay	Emission	Refra
	(g.cm ⁻³)	(10 ⁶)	scopicity	(ph/MeV)	time	peak	ctive
					(ns)	(nm)	Index
Lu ₂ SiO ₅ :Ce (LSO)	7.4	143	No	30 000	40	420	1.82
(LuY)2SiO5:Ce (LYSO)	7.1	63	No	30 000	45	420	1.82
(LuY)AlO3:Ce (LuYaP:Ce)	6.1	-	No	10 000	20	380	2.1
Gd ₂ SiO ₅ :Ce (GSO)	6.71	84	No	9 000	60	440	1.85
YAIO ₃ :Ce (YAP)	5.5	7	No	21 000	30	350	1.94
Y ₃ AI ₅ O ₁₂ :Ce (YAG)	4.6	39	No	16 700	80	530	1.82
LaBr ₃ ;Ce	5.2	-	Yes	63 000	17	380	2.05
LaCl ₃ :Ce	3.8	-	Yes	49 000	18	350	1.90
Lu ₃ Al ₅ O ₁₂ (LuAG:Pr)	6.73	62.9	No	20 000	20	310	1.84

Table 1. Properties of the LuAG:Pr and other most common cerium-activated scintillation materials.

In this study we present the evaluation of a small field of view gamma ray detector based on a R8900U-00-C12 position sensitive photomultiplier tube (PSPMT) coupled to a 2x2 LuAG:Pr scintillator array with 5x5x5mm³ crystal size elements. We have obtained raw images and energy spectra, under various gamma ray energies irradiation. Energy resolution and proportionality of the detector response were measured using controlled gamma sources with energies varying from 81 to 1332.5 keV. For comparison purposes results from a BGO discrete scintillator array with the same thickness are also presented.

2 MATERIALS AND METHODS

The small field of view gamma ray detector is based on a Hamamatsu R8900U-00-C12 position sensitive photomultiplier tube (PSPMT). The PSPMT (Figure 1) has a square field of view (FOV) with external size $30 \times 30 \times 30$ mm³. The minimum effective area of the photocathode is 23.5×23.5 mm² and it is made of bialkali material. The absorption wavelength ranges between 300 and 650nm which matches (25% quantum efficiency at 310nm) the wavelength of the LuAG:Pr scintillator. Charge collection is carried out through an 11 metal channel dynode stage and a 6X+6Y cross plate anode. Very recently this optical detector was evaluated as a high performance gamma imaging probe for axillary sentinel lymph node mapping and more details for its readout and performance can be found in this work ^[17].

The 6X+6Y anode signals were reduced to 2X+2Y using a standard resistive chain (Figure 2a) and then these four signals were fed to a custom pre-amplification circuit (Figure 2b)^[18].



Figure 1. Hamamatsu R8900U-00-C12



Figure 2. a) Resistive chain network, b) pre-amplification circuit of the four position signals

The 4 position signals (Xa, Xb, Yc and Yd) were digitized using a free running sampling technique ^[19]. An FPGA (Spartan 6 LX16) was used for triggering and signal processing of the pulses ^[20]. For each scintillation event these four signals are summed in order to obtain the entire energy information, while the position calculation is carried out by using the Anger logic:

$$X = \frac{Xa}{Xa + Xb} \quad \& \quad Y = \frac{Ya}{Ya + Yb}$$

We acquired raw images and energy histograms of a 2x2 LuAG:Pr scintillator array under various gamma ray energies ranging from 81 to 1332.5keV. A BGO pixilated scintillator was used as a reference crystal at 511keV. The BGO array consists of 6x6 scintillator elements with 2x2x5 mm³ pixel size and 2.3 mm pitch. Figure 3, shows the images of those scintillators used in this study. LuAG:Pr was manufactured by Furukama and BGO by Hilger Crystals. Both scintillators were coupled with Visilox V-788 optical grease to the PSPMT entrance window. We used standard 1µCi radioactivity gamma sources (Ba¹³³, Co⁵⁷, Na²², Cs¹³⁷ and Co⁶⁰) for gamma ray excitation.



Figure 3. Images of the LuAG:Pr (left) and BGO (right) scintillator arrays

3 RESULTS AND DISCUSSION

Figure 4 shows the acquired raw images of a 2x2 LuAG:Pr scintillator array under Na²² excitation. On the left, the scintillator array was coupled directly to the PSPMTs entrance window, while on the right the array coupled with Visilox V-788 optical grease. In both cases, counts produced from the natural radioactivity of Lu¹⁷⁶ were subtracted.





Figure 4. Flood maps of 2x2 array of 5x5x5mm³ LuAG:Pr scintillator excited with Na²² coupled without optical grease (left) and with grease (right). The natural radioactivity of Lu¹⁷⁶ was subtracted.

For comparison purposes, a raw image of a 6x6 array of 2x2x5mm3 BGO scintillator excited with Na22 (left), the peak to valley plot of an horizontal profile (middle) and the energy spectrum of one crystal element are presented in Figure 5.



Figure 5. Raw image of 6x6 array of 2x2x5mm³ BGO scintillator excited with Na²² (left). Peak to valley plot of one horizontal profile (middle).Energy spectrum obtained from crystal element of raw image (right).

All the BGO scintillator elements are clearly identified. The energy resolution (Rs) of BGO under 511 keV excitation was measured equal to 22% (Figure 5 right). Figure 6, shows the energy spectra of LuAG:Pr under Ba¹³³, Co⁵⁷, Na²², Cs¹³⁷ and Co⁶⁰ excitation.



Figure 6. Energy spectra of LuAG:Pr scintillator obtained from crystal elements of raw images. The radioactive sources used for excitation are Ba¹³³, Co⁵⁷, Na²², Cs¹³⁷ and Co⁶⁰ covering an energy area from 81 to 1332.5 keV.

Table 2 shows the energy resolution values (FWHM) of the LuAG:Pr scintillator achieved under various gamma ray excitation gamma sources and 3 different high voltages applied on the PSPMT. In each case high voltage was selected, in order to optimize the systems performance.

Scintillator Material	Energy Resolution @ Gamma energy								
LuAG:Pr	Ba¹³³	Co⁵⁷	Na²²	Cs¹³⁷	Co⁶⁰				
	81keV	122 & 136.5keV	511 & 1274.5keV	662keV	1173.2 & 1332.5keV				
5x5x5mm ³	31%	23%	11.3%	9.7%	7.2% & 4.2%				
without grease	(-887V)	(-887V)	(-783V)	(-783V)	(-783V)				
5x5x5mm ³	25%	18.6%	9.4% & 4.3%	8.5%	5.3% & 4%				
with grease	(-887V)	(-887V)	(-750V)	(-750V)	(-750V)				

Table 2. Energy resolution of the LuAG:Pr scintillator under various gamma ray irradiations and different (optimal) high voltages applied on the PSPMT.

The energy resolution of LuAG:Pr values measured in this study was slightly higher than those reported previously. The energy resolution of LuAG:Pr at 662 keV was equal to 8.5% while in the study of T. Yanagida et al (2010) [15] the energy resolution achieved was 4.6%. The main reason for this is mainly due to the reflector material that we used. Teflon tape used in this study (see Figure 3) absorbs UV photons, thus the PSPMT did not collect a large portion of the laterally scintillation photons produced by LuAG:Pr. BaSO reflector material performs more efficiently and it will be used in future studies. A second reason for this may be due to the fact that in the PEM detector of T. Yanagida *et al* larger LuAG:Pr crystals were used (8 mm than 5mm thick) compared to our setup.

Figure 7, shows the relationship between the ADC centroid channel of the photo peak location and the gamma energy used. The detector presents excellent proportionality in the energy range from 511keV to 1332.5keV. Similar results were obtained at lower gamma energies from 81 to 511keV with the need of slightly higher high voltage in PSPMT (noted in table 1).



Figure 7. Plot that shows the proportionality of LuAG:Pr scintillator. The vertical axis shows the ADC centroid channel of the photopeak location and the horizontal the energy of the gamma ray in keV.

4. CONCLUSIONS

The small field of view gamma detector based on LuAG:Pr scintillator and the R8900U-00-C12 PSPMT has an excellent performance in terms of energy resolution 9.3% @ 511KeV and 8.5% @ 662keV compared with 22% @511keV of BGO. Taking into account its excellent proportionality, it's very fast response (20 ns) and its good light yield (53% of NaI:Tl), LuAg:Pr could be the scintillator of choice for future PET or SPECT detector modules.

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REFERENCES

- [1] G. Blasse, B.C. Grabmaier, Luminescent Materials. Springer-Verlag Berlin, Heidelberg, 1994
- [2] M. Nikl, Scintillation detectors for x-rays, Meas. Sci. Technol. 17, 37-54, 2006
- [3] C.W.E. Van Eijk, 'Inorganic scintillators in medical imaging' Phys. Med. Biol. 47, 85-106, 2002
- [4] A. Lempicki, J. Globo, "Ce-doped scintillators LSO and LuAP," Nucl. Instrum. Methods Phys. Res. A., 414, 333-344, 1998
- [5] Byoungjik K., Wonho L., David K. W. "Comparative Measurements on LaBr3(Ce) and LaCl3(Ce) Scintillators Coupled to PSPMT" *IEEE Nuclear Science Symposium Conference Record*, 2, pp. 861-864, 2004
- [6] Van Eijk C.W. E., Dorenbos P., and Visser R., "Nd³⁺ and Pr³⁺ doped inorganic scintillators," *IEEE Trans. Nucl. Sci.*, 41,738–741, 1994
- [7] Mares J. A., Nikl M., Beitlerova A., Horodysky P., Blazek K., Bartos K., and D'Ambrosio C. Scintillation Properties of Ce³⁺ - and Pr³⁺- Doped LuAG, YAG and Mixed Lu_xY_{1-x}AG Garnet Crystals

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- [8] Pejchal J., Nikl M., Mihokova E., Mares J. A., Yoshikawa A., Ogino H., Schillemat K. M., Krasnikov A., Vedda A., Nejezchleb K., and Mucka V., "Pr³⁺-doped complex oxide single crystal scintillators," *J. Phys. D: Appl. Phys.*, 42, 055117-1–055117-10, 2009
- [9] Yanagida T., Yoshikawa A., Kokota Y., Kamada K., Usuki Y., Yamamoto S., Miyake M., Baba M., Kumagai K., Sasaki K., Ito,N.Abe M, Fujimoto Y., Tanaka H., Fukabori A., dos Santos T. R., Takeda M., and Ohuchi N., "Development of Pr:LuAG scintillator array and assembly for positron emission mammography," *IEEE Trans. Nucl. Sci.*, 57, 1492–1495, 2010
- [10] Swiderski L., Moszynski M., Nassalski A., Synfeld-Kazuch A., Szczesniak T., Kamada K., Tsutsumi K., Usuki Y., Yanagida T., and Yoshikawa A., "Light yield non-proportionality and energy resolution of praseodymium doped LuAG scintillator," *IEEE Trans. Nucl. Sci.*, 56, 934–938, 2009
- [11] Ogino H., Yoshikawa A., Nikl M., Kamada K., and Fukuda T., "Scintillation characteristics of Prdoped Lu₃Al₅O₁₂ single crystals," J. Cryst. Growth, 292, 239-242, 2006
- [12] Drozdowski W., Lukasiewicz T., Wojtowicz A., Wisniewski D., Kisielewski J., "Thermoluminescence and scintillation of praseodymium-activated YAG and LuAG crystals," J. Cryst. Growth, 275, 709-714, 2005
- [13] Chewpraditkul W., Sreebunpeng K., Nikl M., Mares J. A., Nejezchleb K., Phunpueok A., Wanarak C., "Comparison of Lu₃Al₅O₁₂:Pr³⁺ and Bi₄Ge₃O₁₂ scintillators for gamma-ray detection," *Rad. Meas.*, 47, 1-5, 2012
- [14] Nikl M., Ogino H., Krasnikov A., Beitlerova A., Yoshikawa A., Fukuda T., "Photo- and radioluminescence of Pr-doped Lu₃Al₅O₁₂ single crystal," *Phys. Stat. Sol. A.*, 202, 4-6, 2005
- [15] T. Yanagida, A. Yoshikawa, Y. Yokota, K. Kamada et al., "Development of Pr:LuAG Scintillator Array and Assembly for Positron Emission Mammography," *IEEE Trans. Nucl. Sci.*, 57, 1492-1495, 2010
- [16] Nakamori T., Kato T., Kataoka J., Miura T., Matsuda H., Sato K., Ishikawa Y., Yamamura K., Kawabata N., Ikeda H., Sato G. and Kamada K. "Development of a gamma-ray imager using a large area monolithic 4 × 4 MPPC array for a future PET scanner," *JINST*, 7, 2012, DOI:10.1088/1748-0221/7/01/C01083
- [17] Georgiou M., Loudos G., David S., Papadimitroulas P., Liakou P., Georgoulias P., "Optimization of a gamma imaging probe for axillary sentinel lymph mapping," *JINST, in press 2012.*
- [18] Olcott P. D., Talcott J. A., Levin C. S., Habte F., Foudray A. M. K., "Compact Readout Electronics for Position Sensitive Photomultiplier Tubes," IEEE Trans. Nucl. Sci. 52, 21, 2005
- [19] M. Streun, G. Brandenburg, H. Larue, E. Zimmermann, K. Ziemons, H. Halling, "Pulse recording by a free running sampling," *IEEE Trans. Nucl. Sci.*, 48, 524-526, 2001
- [20] SP601 Hardware User Guide, Xilinx Inc., UG518 (v1.4), 2010 (datasheet available online at http://www.xilinx.com