COMPARATIVE STUDY OF LUMINESCENCE PROPERTIES OF Lu_2SiO_5:Ce AND $YAlO_3$:Ce SINGLE CRYSTAL SCINTILLATORS FOR USE IN MEDICAL IMAGING

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Abstract
The luminescence properties of Lu_2SiO_5: Ce (LSO: Ce) and $YAlO_3$: Ce (YAP: Ce) crystals were studied for use in medical X-ray imaging. LSO: Ce and YAP: Ce are non-hydroscopic with 7.4 g/cm^3 and 5.5 g/cm^3 density, and short decay time (40 ns and 30 ns respectively) scintillators. Evaluation was performed by determining the absolute luminescence efficiency (emitted light flux over incident x-ray exposure) in energies employed in general X-ray imaging (40-140 kV) and in mammographic X-ray imaging (22-49 kV). Additionally the light emission spectrum at various X-ray energies (22-140 kV), and the spectral compatibility to optical photon detectors incorporated in medical imaging systems, were determined. Furthermore, light transmission measurements were carried out in order to estimate the light transmission efficiency and to calculate their intrinsic conversion efficiency. The light emission performance of the two scintillation materials studied was found adequately high for x-ray imaging.

1. Introduction

Cerium (Ce^{3+}) doped scintillators are of particular interest for medical imaging, because of their very fast response. The latter is due to an electric dipole transition in Ce ion energy states [1,2]. Yttrium Aluminum Perovskite, $YAlO_3$:Ce (YAP:Ce) is a non hygroscopic scintillator emitting blue light (300-450nm) with a very fast decay time of 30ns [3, 4]. Lutetium oxyorthosilicate Lu_2SiO_5:Ce(LSO:Ce) has been recognized as one of the best scintillating materials [5, 6], due to its high density of 7.4 g/cm^3, fast decay time of 40 ns, suitable emission wavelength peaking at 420 nm, non hygroscopicity and excellent chemical stability compared to other scintillators.

These properties are very attractive for x-ray imaging since: (i) their light spectral distribution shows high compatibility to several existing optical sensors (i.e. charge coupled devices, photocathodes, radiographic films), (ii) fast decay time is a prerequisite for dynamic real-time imaging. LSO:Ce and YAP:Ce scintillator based image detectors have already been evaluated and employed in Positron Emmission Imagers, in synchrotron and $\gamma-$ray imaging [4-9]. A common denominator in testing the performance of scintillator materials is their luminescence efficiency or light yield (photons/MeV). In our study, the luminescence efficiency of LSO:Ce and YAP:Ce single crystal scintillators was examined under exposure conditions employed in medical diagnostic radiology.

2. Materials and Methods

2.1. Theory

The absolute luminescence efficiency (η_A) of a scintillation crystal in x-ray imaging can be given by the ratio [10-13]

$$\eta_A = \dot{\Psi}_\lambda / \dot{X} \tag{1}$$

where $\dot{\Psi}_\lambda$ is the light energy flux, emitted by a an excited scintillator and $\dot{X}$ is the exposure rate incident on the phosphor, emitted by an x-ray tube. Absolute efficiency is expressed in units of [$\mu W \cdot m^{-2} / mR \cdot s^{-1}$ or Efficiency Units (E.U.)].

The fraction of the absorbed x-ray energy converted into light energy within the scintillation material is often called the x-ray to light conversion efficiency (η_C) given as follows [14]:

$$\eta_C = \frac{\overline{E}_\lambda}{E_g} \cdot \left(\frac{S \cdot Q}{\beta}\right) \tag{2}$$

where $\overline{E}_\lambda$ is the mean energy of the emitted light photons, E_g is the forbidden energy gap between the valence and the conduction energy bands, S is the transfer efficiency of the electron-hole pair expressing the fraction of electron-hole energy transferred to the site of the activator (Ce^{3+}), Q is the absorption efficiency of the activator, expressing the fraction of transferred electron-hole pair energy absorbed at the activator site and β is a parameter characterizing the excess energy, above E_g, required to be absorbed so as to allow for an electron-hole pair generation. The mean energy of light photons $\overline{E}_{\lambda}$ may be determined from the spectrum of light $S_P(\lambda)$ emitted by the scintillator crystal.

The effect of the emmitted light spectrum on the overall detector (scintillator-optical detector) performance may be estimated by determining the spectral compatibility of the scintillator's emitted light to the spectral sensitivity of the optical photon detector. Spectral compatibility may be estimated by the spectral matching factor (SMF), which has been defined by the ratio [15]

$$\alpha_S = \frac{\int S_P(\lambda) S_D(\lambda) d\lambda}{\int S_P(\lambda) d\lambda} \quad (3)$$

where $S_P(\lambda)$ is the spectrum of the light emitted by the scintillator and $S_D(\lambda)$ is the spectral sensitivity of the optical detector coupled to the scintillator.

The overall efficiency corresponding to a specific scintillator-optical photon detector combination has been expressed by the effective efficiency (EE) [16]. EE expresses the fraction of emitted light energy flux that may be captured by the optical photon detector per unit of incident radiation exposure rate. EE is given as the absolute luminescence efficiency multiplied by the corresponding spectral matching factor [12]:

$$EE = \eta_{Eff} = \eta_A \cdot \alpha_S \quad (4)$$

where η_{Eff} denotes the effective efficiency (EE) [16].

2.2 Experiments

The LSO:Ce and YAP:Ce crystals examined in this study had dimensions of 10mm×10mm×20mm, with concentration of 0,1% Ce. Both crystals had all their surfaces polished. The crystals were irradiated by x-rays using the following imaging units: (i) A General Electric Senographe DMR x-ray mammography unit equipped with a molybdenum (Mo) anode target and molybdenum filter and (ii) A Philips Optimus x-ray unit with a tungsten (W) anode target and 2mm Al filter. In the x-ray mammography unit the filter changed automatically from molybdenum to rhodium (Rh) and aluminum (Al) filters as tube voltage increased from 22 to 49 kVp. In general x-ray unit the tube voltage varied from 40 to 140 kVp. The samples were not exposed to strong UV light, including sunlight, at any time during the course of the irradiation measurements. For measurements performed under x-ray mammographic conditions, a 30 mm thick block of Perspex was used to simulate beam hardening by human breast [17]. Similarly a 20 mm thick Al block was used to simulate beam hardening by the human body in general x-ray imaging conditions [18].

The absolute luminescence efficiency was determined, according to (1), by performing x-ray exposure and light flux measurements. The exposure rate, $\dot{X}$ in (1), was measured at the crystal's position using a Radcal 2026C dosimeter (Radcal Corp., USA). Light energy flux, $\dot{\Psi}_{\lambda}$, measurements were performed using an experimental setup comprising a light integration sphere (Oriel 70451) coupled to a photomultiplier (EMI 9798B), previously described by Valais et al [19]. The samples were wraped with several layers of Teflon tape with only one end face left to couple to the window of the integration sphere. The intrinsic energy conversion efficiency was determined according to relation (2). To determine the value of η_C, parameters Q and S were assumed to be equal to 1. Values for the parameter β, for inorganic scintillators, range between 2 and 3 [2], but in some cases of crystalline scintillators can exceed the value of 5 [1, 2]. The energy gap, E_g, for each material was obtained from the literature [20, 21]. The light transmission efficiency, $\eta_{\lambda}(\lambda)$, was determined from light transmission measurements obtained by a Perkin-Elmer UV/Visible lamda 15 Spectrophotometer (Perkin-Elmer Life And Analytical Sciences, Inc., USA).

To determine both the mean light photon energy $\overline{E}_{\lambda}$ and the spectral matching factor α_S, the emitted light spectrum $S_P(\lambda)$ of the LSO:Ce and YAP:Ce crystals were measured using a wavelength range from 200 to 900 nm, using a grating optical spectrometer (Ocean Optics Inc., HR2000). Spectrometer light measurements were performed under x-ray excitation. X-ray measurements were performed at various x-ray tube voltages up to 140 kVp. Spectral sensitivity ($S_D(\lambda)$) data were obtained from corresponding manufacturer's (Hamamatsu, EMI, etc.) datasheets. Six optical photon detectors currently used in a large variety of imaging detectors (digital and conventional radiography, fluoroscopy, computed tomography, nuclear medicine, etc.) and their spectral matching factor with LSO:Ce and YAP:Ce spectra were examined (Table II). These optical detectors were the following: (i) GaAs photocathode, (ii) extended S20 EMI photocathode with quartz window, (iii) GaAsP Hamamatsu photocathode, iv) a-Si:H/108H amorphous silicon photodiode corresponding to intrinsic layer thickness of 800nm (108H), (v) Si/S1133 Hamamatsu crystalline silicon photodiode and (vi) CCD S100AB SITe®.

3. Results and Discussion

Figures 1 and 2 shows the variation of the absolute luminescence efficiency of LSO:Ce and YAP:Ce crystals in the x-ray tube voltage from 22 kVp to 140 kVp, i.e. in the mammography region, 22-49 kVp (fig. 1), and in the general x-ray imaging region, 50-140 kVp (fig. 2). The fitted curve shown in the figure was obtained by polynomial fitting.

Under both mammographic and general x-ray imaging conditions, the absolute efficiency curves showed a nonlinear increase with increasing x-ray tube voltage. This behavior was similar to efficiency data previously obtained by our group on different scintillator materials [19].

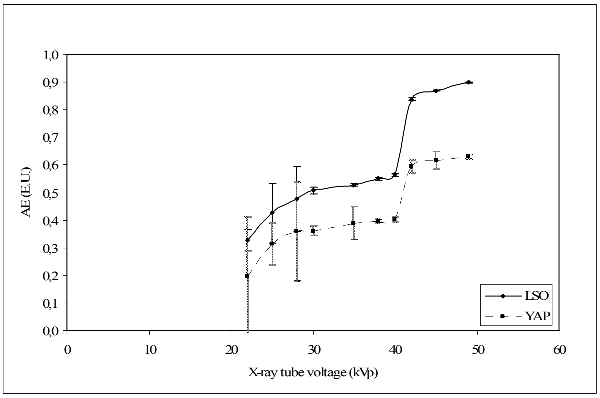

Figure 1. Variation of absolute luminescence efficiency (AE) of LSO:Ce and YAP:Ce crystals for mammographic x-ray tube voltages, between 22 and 49 kVp. AE units: $\mu W \cdot m^{-2} / mR \cdot s^{-1}$. Points: measured data, line: fitted curve.

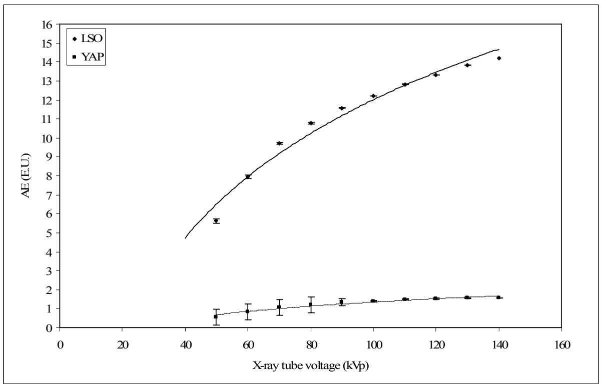

Figure 2. Variation of absolute luminescence efficiency (AE) of LSO:Ce and YAP:Ce crystals for radiographic x-ray tube voltages, between 50 and 140 kVp. AE units: $\mu W \cdot m^{-2} / mR \cdot s^{-1}$. Points: measured data, line: fitted curve.

The intrinsic conversion efficiency (η_C) was estimated to be significantly higher for LSO:Ce than for YAP:Ce. Corresponding results, shown in Table II, were calculated using relation (2) and published data [2, 20, 21].

Table 1: Theoretical maximum intrinsic conversion efficiency of LSO:Ce and YAP:Ce scintillators.

Parameter	LSO:Ce	YAP:Ce
E_g (eV)	6,40[b]	7,70[c]
β	5,60[d]	7,08[d]
Light Yield (photons/MeV)	27000[a]	21000[a]
η_C	0.081[d]	0.062[d]

[a] Data are from [1]
[b] Data are from [21], [c] Data are from [20]
[d] Calculated data

The intrinsic efficiency of LSO:Ce ($\eta_C = 0.099$) was estimated to be high enough compared to most of the currently used scintillators (CsI, BGO, Gd_2O_2S), in various medical imaging applications [1,2,9]

Figure 3 shows the light transmission measurements obtained using the Perkin-Elmer UV/Visible lamda 15 Spectrophotometer. In figure 3, transmittance expresses the fraction (%) of light passing through the crystals and reaching the spectrophotometer optical detector. Both crystals showed high transmittance (84.1% for LSO:Ce and 82.2% for YAP:Ce), in the visible region above 450 nm. These values seem to be higher than the corresponding light transmission values previously published for LSO:Ce [21]. In the region from 450 nm down to 410 nm, for LSO, and down to 360 nm, for YAP, transmittance showed a tendency to decrease very slightly. For lower wavelengths, transmittance was found to drop rapidly down to values close to zero. The absorption cut-off shifts were close to 390 nm for LSO and 330 nm for YAP mainly due to the cerium dopand [21].

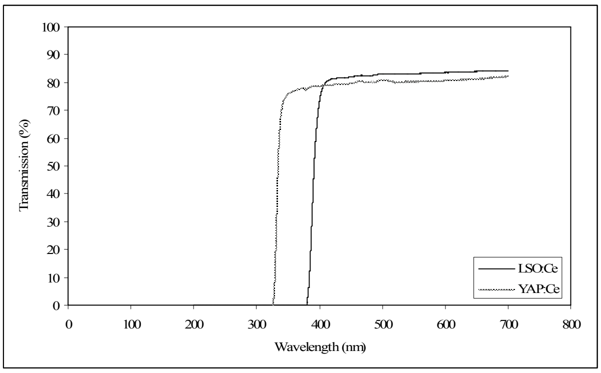

Figure 3. Light transmission measurements using a spectrophotometer Perkin Elmer Lamda 15. Above 600 nm or below 300 nm the transmittance of LSO:Ce and YAP:Ce was constant.

Figure 4 shows the spectral response of LSO:Ce and YAP:Ce crystals, respectively, under 140 kVp x-ray excitation. The measurements were performed at room temperature. LSO:Ce spectrum, extending from 390 to over 470 nm and peaking at about 420 nm was found very similar to the one reported by others [21]. The long tail on the right part of the spectrum should be ascribed to the *5d-4f* electronic transitions of the Ce^{3+} ion, which locates at Ce2 center. The Ce2 emmission, much weaker and not well resolved than Ce1 emmission at 393-423 nm, occurs at 500 nm. YAP:Ce spectrum, extending from 350 nm to over 410 nm and peaking at about 370 nm [19] was found to be well situated within the spectral sensitivity of most optical photon detectors examined. Since LSO:Ce x-ray excited spectrum is broader than YAP:Ce, the corresponding matching factor of LSO with the optical photon detectors it is expected to be greater than YAP:Ce (Table 2).

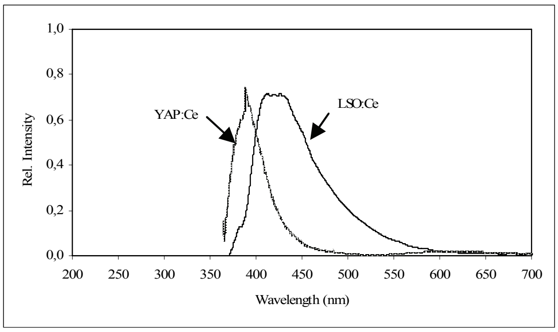

Figure 4. Spectral response of LSO:Ce and YAP:Ce under 140 kVp x-ray excitation. LSO:Ce spectrum is peaking at 420 nm and YAP:Ce spectrum peak is at 370 nm, respectively.

Table 2 summarizes the calculated spectral matching factors of LSO:Ce and YAP:Ce crystal scintillators with some optical photon detectors, using relation (5). The spectral sensitivity of the optical photon detectors (S_D), in Table 2, were taken from manufacturers' datasheets.

Table 2: Spectral Matching Factors of LSO:Ce and YAP:Ce with some optical detectors.

Optical Detectors	LSO:Ce	YAP:Ce
GaAs Photocathode	0,93689	0,88682
Extended S-20 Photocathode	0,98756	0,96518
GaAsP Hamamatsu Photocathode	0,66715	0,40124
a-Si:H 108H Photodiode	0,57946	0,49545
Si/S1133 Hamamatsu Photodiode	0,51123	0,32036
CCD S100AB SITe®	0,90449	0,82781

4. Conclusions

In conclusion, our measurements showed that the absolute efficiency of LSO:Ce (14,21 $\mu W \cdot m^{-2} / mR \cdot s^{-1}$ at 140kVp) scintillator crystal was found higher than the corresponding of YAP:Ce crystal (1,56 $\mu W \cdot m^{-2} / mR \cdot s^{-1}$ at 140 kVp) respectively. LSO:Ce luminescence efficiency was found higher than the corresponding of YAP:Ce crystal in the whole range of energies used in our study. In the mammographic energy range the difference between the AE values was smaller than the ones obtained in the radiographic energy region. This lead us to the assumption that both materials can be efficiently used for low x-ray energy imaging (below 50 kVp). The intrinsic conversion efficiency was estimated to be significantly higher for LSO:Ce. The light emission spectrum of the two scintillators, peaking at about 420 nm for LSO:Ce and at about 370 nm for YAP:Ce respectively, was found compatible to many currently employed optical photon detectors. Taking into account the very short scintillation decay time of both scintillators, they can potentialy be used in modern fast image producing x-ray computed tomography systems, especially those employed in combined PET/CT devices.

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References

[1] van Eijk C.W.E Phys. Med. Biol. 47, R85-R106, (2002)
[2] G. Blasse, B.C. Grabmaier. Luminescent Materials. (Springer-Verlag. Berlin, Heidelberg, 1994) p.85
[3] Gorin A., Kuroda K. et al. Nucl. Instr. and Meth A, Vol 510, 76-82, 2003.
[4] Hirota K., Toyokawa H. et al. Nucl. Instr. and Meth A, 510, 83-91, 2003.
[5] Melcher, C. L.; Schweitzer, J. S. Nucl. Instrum. Methods Phys. Res., A 314, 212. 1992
[6] Melcher, C. L.; Schweitzer, J. S., U.S. Patents 4,958,080; 5,025,151; 5,660,627
[7] C.L. Melcher, M. Schmand, M. Eriksson, et al., IEEE Trans. Nucl.Sci. 47 (2000) 965–968. 1992.
[8] M. Moszynski, M. Kapusta, D., et al. Nucl. Instr. and Meth A, 404, 157-165, 1998.
[9] M. Nikl, E. Mihokova, et al. Phys. Stat. Sol. 181, R10-R12, 2000.
[10] Ludwig, G.W., J. Electrochem. Soc. 118, 1152. 1971
[11] Kandarakis, I., Cavouras,. D., Nucl. Instr. Meth. Phys. Res. A. 417, 86. 1998
[12] I. Kandarakis, D. Cavouras, G. S. Panayiotakis, C. D. Nomicos, Phys. Med. Biol., vol. 42, pp. 1351-1373, 1997.
[13] J.M. Boone, SPIE Press, Bellingham, pp. 67-68, 2000.
[14] G. Blasse, J. Lumin, Vol. 68, pp. 225-240, 1994.
[15] I. Kandarakis, D. Cavouras, C.D. Nomicos, and G.S. Panayiotakis, Nucl. Instr. Meth. Phys. Res. B., Vol. 179, pp. 215-224, 2001.
[16] D. Cavouras, I. Kandarakis, G.S. Panayiotakis, A. Bakas, D. Triantis, and C.D. Nomicos, Br. J. Radiol., Vol. 71, pp. 766-772, 1998.
[17] J.M. Boone, Med. Phys., Vol. 29, pp. 869-875, 2002.
[18] T. Holm, R. Moseley, Radiology, pp. 898-903, 1964.
[19] I. Valais, I. Kandarakis, D. Nikolopoulos, I. Sianoudis, N. Dimitropoulos, D. Cavouras, C.D. Nomicos and G.S. Panayiotakis, IEEE Trans. Nucl. Sci. Vol. 52, pp. 1830-1835, 2005.
[20] R.H. Bartram, A. Lempicki, J. Lumin, Vol. 68, pp. 225-240, 1996.
[21] G. Ren, L. Qin, S. Lu, and H. Li, Nucl. Instr. Methods Phys. Res. A, Vol. 531, pp. 560-565, 2004.