Luminescence properties of $LuYSiO_5$: Ce, Gd_2SiO_5: Ce, and CsI: Tl single crystal scintillators under x-ray excitation, for use in medical imaging systems.

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***Abstract*—The luminescence of $LuYSiO_5$: Ce (LYSO: Ce) and Gd_2SiO_5: Ce (GSO: Ce) crystals was studied for use in tomographic medical x-ray imaging. Both crystals were compared to CsI: Tl scintillator, used in x-ray and gamma-ray imaging. LYSO: Ce and GSO: Ce are high density (7.1 g/cm^3 and 6.71 g/cm^3 respectively), high atomic number (71 for Lu and 64 for Gd), non-hydroscopic, and short decay time (40 ns and 60 ns respectively) scintillators. CsI: Tl is a slightly hygroscopic, high light yield (≥10^4 photons/MeV) scintillator but of relatively slow decay time (>200 ns). Evaluation was performed by determining: 1) the absolute luminescence efficiency (emitted light flux over incident x-ray exposure) in x-ray energies employed in general x-ray imaging (40-140 kV) and in mammographic x-ray imaging (22-49 kV), 2) the light emission spectrum, determined at various x-ray energies (22-140 kV), and 3) the spectral compatibility to optical photon detectors incorporated in medical imaging systems. The light emission performance of the three scintillation materials studied, were found adequately high for x-ray imaging. LYSO: Ce and GSO: Ce were found most efficient (between 8 and 12 $\mu W \times m^{-2} / mR \times s^{-1}$) in the range from 40 to 60 kV. LYSO: Ce and GSO: Ce were found adequately most compatible with the S-20 photocathode (0.902 and 0.903 respectively against 0.763 of CsI: Tl) and adequately compatible to a-Si photodiode (0.705 and 0.737 respectively against 0.851 of CsI: Tl).**

Index Terms—**Inorganic scintillators, LYSO, GSO, Luminescence Efficiency, Matching Factor.**

I. Introduction

Scintillators coupled to optical photon detectors (photocathodes, photodiodes etc.) are used in most of the current medical diagnostic imaging modalities, using x-rays or gamma rays, inorganic scintillators are employed [1].

Scintillator detectors using Cesium Iodide crystal doped with Thallium, (CsI: Tl), show high gamma ray detection efficiency. The most important features of CsI(Tl) are its high light yield (≥10^4 photons/MeV) and its emission spectrum centered at about 550 nm [2]. This allows the use of photodiodes as optical detectors to detect the emitted light spectrum [3].

Due to the above characteristics, CsI (Tl) has been widely used in many medical imaging applications (e.g. gamma-ray cameras, x-ray computed tomography, digital radiography and mammography, dentistry (intra-oral radiography), fluoroscopy, etc) [1]. One undesirable feature of CsI(Tl) scintillator is its relatively high decay time (>200ns) which limits its use in fast medical imaging applications, i.e. Spiral Computed Tomography and Positron Emission Tomography (PET). As a result there is much interest in introducing new scintillator materials (Table I).

Cerium-based (Ce^{3+}) scintillators have a much faster response. A good compromise between fast response (40 ns) and relatively high light yield (26000 ph/MeV) seems to be Cerium (Ce^{3+}) doped Lutetium oxyorthosilicate, (Lu_2SiO_5:Ce or LSO:Ce) scintillator. However it appeared to be very difficult to grow large, stress free crystals of which small entities can be cut efficiently. These large crystals show inhomogeneities in light production and decay time and their energy resolution is poorer than the expected when using theoretical models [4].

Gadolinium oxyorthosilicate (Gd_2SiO_5 or GSO), doped with cerium (Ce^{3+}) ion activator is a high density ($\rho = 6.71\ g/cm^3$), high effective atomic number ($Z(Gd) = 59$) and high radiation detection index ($\rho Z_{eff}^4 = 84 \times 10^6$) crystal and due to the presence of Ce^{3+} ion activator, it exhibits a fast response with a decay time of 60*ns*. In addition, its low cost, as compared to LSO, led GSO: Ce to be used in commercial positron emission tomography detectors [5]. However due to the relatively low light yield (≥8000 ph/MeV), GSO: Ce remains not the scintillator of choice.

Cerium doped Lutetium Yttrium Oxyorthosilicate, $(Lu,Y)_2SiO_5$:Ce (LYSO: Ce), is a mixed LSO/YSO non-hygroscopic crystal that offers high density (7.1 g/cm^3, 5-10%Y), high light output (≥30000 ph/MeV), good energy

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resolution (~10%) and short decay time (40ns) that makes it ideal in a wide range of γ-ray detection applications [6].

In the present study we comparatively investigate the light emission characteristics of LYSO: Ce, GSO: Ce and CsI: Tl single crystal scintillators under x-ray medical imaging conditions. To this aim four parameters related to the luminescence emission spectrum and emission efficiency were studied using experimental methods. A similar approach, with LYSO, LSO and BGO crystal scintillators has been reported by Pepin *et al*, 2004, for phoswich PET detectors [7].

II. MATERIALS AND METHODS

A. Definitions

Scintillators emission efficiency (light yield) may be evaluated by measuring the absolute luminescence efficiency (AE) [8, 9], which has been defined [9-11] as the ratio of the light energy flux ($\dot{\Psi}_\lambda$), emitted by an excited scintillator, over the incident exposure rate ($\dot{X}$):

$$AE = \eta_A = \frac{\dot{\Psi}_\lambda}{\dot{X}} \qquad (1)$$

where η_A is the absolute efficiency expressed in AE-units [$\mu W \cdot m^{-2}/mR \cdot s^{-1}$]. AE may be used to describe the radiation detection sensitivity of energy integrating detectors, i.e. detectors producing a signal directly related to the total energy absorbed within the scintillator's mass.

Spectral compatibility may be estimated by the spectral matching factor (SMF), which has been defined by the ratio [12]:

$$SMF = \alpha_S = \frac{\int S_P(\lambda) S_D(\lambda) d\lambda}{\int S_P(\lambda) d\lambda} \qquad (2)$$

where S_P is the spectrum of the light emitted by the scintillator, S_D is the spectral sensitivity of the optical photon detector and λ denotes the wavelength of the emitted light.

The effective luminescence efficiency (EE) expresses the detection efficiency corresponding to a specific scintillator optical photon detector combination [14]. EE can be calculated by multiplying the absolute luminescence efficiency by the corresponding spectral matching factor [10]:

$$EE = \eta_{Eff} = \eta_A \cdot \alpha_S \qquad (3)$$

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

EE may be used to express the overall efficiency of a scintillator-optical detector combination.

The fraction of the absorbed x-ray energy converted into light energy within the scintillation material is often called x-ray to light conversion efficiency (η_C) given as follows[15]:

$$\eta_C = \overline{E}_\lambda \cdot \frac{1}{E_g} \cdot \left(\frac{S \cdot Q}{\beta}\right) \qquad (4)$$

where E_g is the forbidden energy band gap, S is the electron-hole pair energy transfer efficiency expressing the fraction of electron-hole energy transferred to the activator site, Q the absorption efficiency of the activator site, expressing the fraction of transferred electron-hole pair energy absorbed at the activator site and β is a constant which characterizes the excess energy above E_g required to be absorbed so as to allow for an electron-hole pair to be generated.

B. Experiments

The LYSO: Ce and GSO: Ce crystals used in this study were supplied by Photonic Materials Ltd., Scotland, U.K., and by Hitachi Chemicals Co., Ltd, Japan, respectively with dimensions of 10mm×10mm×10mm, and doped with 0.5% mol of cerium (Ce^{+3}). The CsI: Tl crystal used in this study was supplied by CRYOS-Beta Ltd., Ukraine with dimensions of 50mm in diameter and 5mm in thickness. This 5mm crystal was chosen among a set of five CsI: Tl crystals with different thicknesses (between 1 to 7mm) due to its higher luminescence characteristics, using the same x-ray exposure parameters. The crystals were irradiated by X-rays using: (i) A Philips Optimus x-ray unit with a tungsten anode target and 2mm Al filter (to simulate conditions of general purpose computed tomography) and (ii) A General Electric Senographe DMR x-ray mammography unit equipped with a molybdenum anode target and molybdenum filter (to simulate conditions of tomographic breast imaging). The filter changed automatically to rhodium (Rh) and aluminum (Al) filters as we increased from medium to higher mammographic voltages. The whole range of available x-ray tube voltages varied from 22 to 49 kV in the mammography unit and from 40 to 140kV in the general radiography unit.

The absolute luminescence efficiency was determined according to equation (1), by performing x-ray exposure and light flux measurements. The exposure rate was measured at the crystal's position using a Radcal 2026C dosimeter (Radcal Corp., USA). Light energy flux measurements were performed using the experimental setup described by Valais *et al* , 2005 [16].

The spectral matching factor was determined using equation (2). The emitted light spectrum $S_P(\lambda)$ of LYSO: Ce, GSO: Ce and CsI: Tl crystals was measured using the Ocean Optics optical grating spectrometer. Measurements were performed using x-ray excitation. Spectral sensitivity ($S_D(\lambda)$) data were obtained from corresponding manufacturer's (Hamamatsu, EMI, etc.) datasheets. Six optical photon detectors and their spectral matching factor with LYSO: Ce, GSO: Ce and CsI: Tl crystal spectra were examined (Table II).

III. RESULTS AND DISCUSSION

Fig. 1 shows the variation of the absolute luminescence efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals with x-ray tube voltage. Measurements were obtained using the general radiography unit and employing the full range of available tube voltage values from 40 to 140 kV. The fitted curve shown in the figure was obtained by a logarithmic fitting. A similar non-linear response has been also documented for NaI:Tl by Valentine *et al*, 1998 [18].

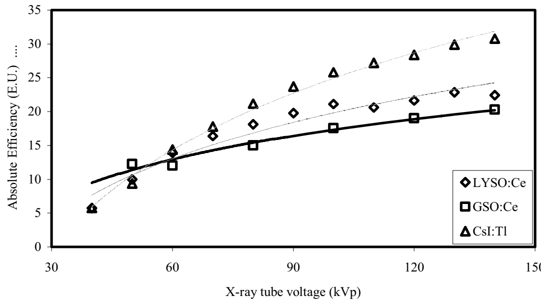

Figure 1 Variation of absolute luminescence efficiency (AE) of LYSO: Ce, GSO: Ce and CsI: Tl crystals for Radiographic X-ray tube voltages, between 40 and 140 kV. AE units: μW· s/ mR· m^2. Points: measured data, line: fitted curve.

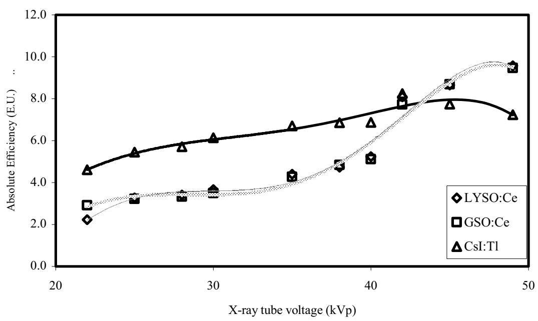

Figure 2. Variation of absolute luminescence efficiency (AE) of LYSO: Ce, GSO: Ce and CsI: Tl crystals for Mammography x-ray tube voltages, between 22 and 49 kV. AE units: μW· s /mR · m^2. Points: measured data, line: fitted curve.

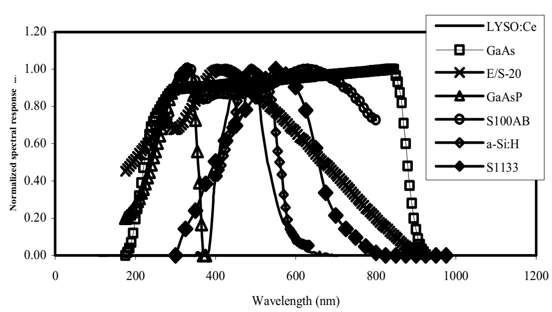

Figure 3. Normalized spectral response of LYSO: Ce compared to the spectral sensitivities of GaAs, ES-20, GaAsP,CCD S100AB, a-Si:H 108H and S1133 photodetectors.

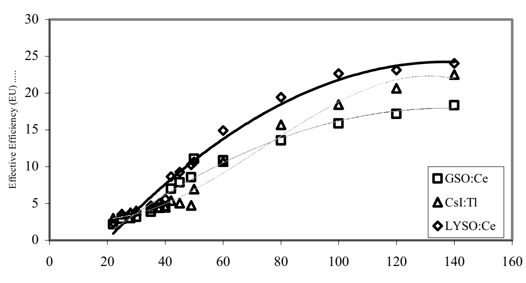

Figure 4. Effective efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals using S-20 EMI photocathode

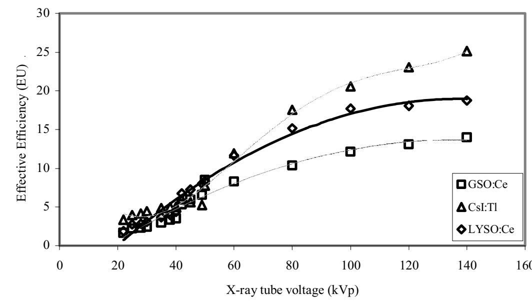

Figure 5. Effective efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals using aSi:H-108H amorphous silicone photodiode with intrinsic layer thickness 800nm

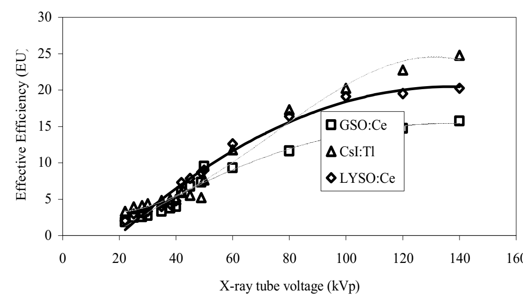

Figure 6. Effective efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals using Si/S1133 crystaline silicone Hamamatsu photodiode.

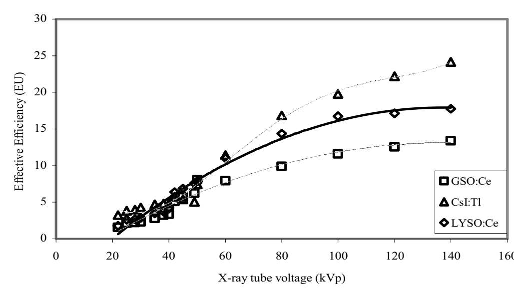

Figure 7. Effective efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals using GaAsP Hamamatsu photocathode

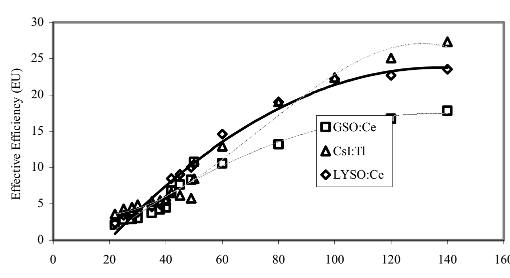

Figure 8. Effective efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals using CCD S100AB SITe photodetector

Fig. 2, shows the variation of absolute luminescence efficiency of LYSO: Ce, GSO: Ce and CsI: Tl crystals in the mammographic x-ray tube voltage range (22-49 kV, molybdenum anode spectrum). As it may be seen, absolute luminescence efficiency increases, in a non proportional way, with increasing x-ray tube voltage. The fitted curve shown in the figure was obtained by a fifth order polynomial fitting. An interesting point to notice in Fig. 2 is that for LYSO: Ce at 42 kV a sharp increase was observed. This was attributed to the change of the mammography tube filter from Rh to Al. On the contrary, no significant variation was observed when the filter changed from Mo to Rh at lower energies (38 kV). The same behaviour was observed in a previous study of ours involving GSO: Ce crystal [17]. The above consideration did not apply to the CsI: Tl crystal, which shows no significant discontinuity in the whole energy range in this study.

Fig. 3, shows the normalized spectral response of LYSO: Ce crystal compared to the spectral sensitivities of the six afore mentioned optical photon detectors.

Figs 4 to 7 shows the luminescence effective efficiency of LYSO: Ce single crystal scintillator with some optical detectors. As one can observe luminescence effective efficiency curves are quite close although the optical detectors used in our graph were of different composition.

The best luminescence efficiency was found for GaAs photocathode (Matching Factor 0.92635.

The intrinsic conversion efficiency η_C for the scintillators used in this study was calculated using equation (4). For obtaining the maximum theoretical intrinsic conversion efficiency Q, and S were taken equal to 1. The constant β for inorganic scintillators is ranging between 2-3. For simplicity reasons its value were taken $\beta \approx 2.5$ [1, 19]. The energy gap E_g for each material were reported elsewhere [1, 20-21]. Our calculations are shown in Table III.

The AE values of LYSO: Ce (23E.U.) was found higher compared to that of GSO: Ce values (19E.U.) previously studied [18,22], which suggests that the former scintillator crystal could be useful in designing detectors for x-ray computed tomography as well as for the recently proposed breast computed tomography systems[23]. In addition, these data could be useful to obtain an estimation of the efficiency of the LYSO: Ce crystal under mono-energetic radiation used in ordinary nuclear medicine.

IV. Conclusions

In conclusion, our measurements showed that the absolute efficiency of LYSO: Ce scintillator crystal increased with x-ray tube voltage in the range of mammographic and general x-ray imaging energies.

The absolute efficiency of LYSO: Ce was found adequately high (22.5 $\mu W.m^{-2} / mR.s^{-1}$ at 140 kV). In addition, the LYSO: Ce's emission spectrum, extending from 390 to over 470 nm and peaking at about 430 nm, was found compatible (70%- 95%) to many currently employed optical photon detectors (amorphous and crystalline silicon photodiodes etc).

TABLE II.
Spectral Matching Factors of LYSO: Ce, GSO: Ce and CsI: Tl with some optical detectors

Optical Detectors	LYSO: Ce	GSO: Ce	CsI: Tl
GaAs Photocathode	0.92635	0.92707	0.94076
Extended S-20 Photocathode	0.90185	0.90294	0.76304
GaAsP Hamamatsu Photocathode	0.66715	0.65991	0.81042
a-Si:H 108H Photodiode	0.70491	0.68984	0.85120
Si/S1133 Hamamatsu Photodiode	0.76025	0.77425	0.83827
CCD S100AB SITe®	0.88449	0.87917	0.92654

TABLE III

Theoretical intrinsic conversion efficiency of LYSO: Ce, GSO: Ce and CsI: Tl scintillators

Parameter	LYSO: Ce	GSO: Ce	CsI: Tl
E_g	6.4	6.2	6.4
β	2.5	2.5	2.5
η_C	0.103	0.026	0. 13

V. Acknowledgments

-This work was financially supported by the EPEAEK program "Archimidis".

-The authors gratefully acknowledge Photonic Materials Ltd., Scotland, UK, for supplying the LYSO: Ce crystal, Hitachi Chemical Co. for supplying the GSO crystal and CRYOS Ltd., Ukraine, for supplying the CsI: Tl crystal.

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