Luminescence efficiency of (Gd_2SiO_5:Ce) scintillator under x-ray excitation

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***Abstract*–Gd_2SiO_5:Ce (GSO:Ce) is a high-Z, non-hygroscopic fast emitting scintillator used in detectors of some positron tomography systems. The purpose of this study was to evaluate the luminescence of GSO:Ce scintillator at lower photon energies employed in other fields of medical imaging (e.g. x-ray computed tomography, general x-ray imaging, low energy gamma ray imaging). To this aim the absolute luminescence efficiency (emitted light flux over x-ray exposure), the matching factor (spectral compatibility to optical detectors) and the effective efficiency (combination of absolute efficiency with spectral compatibility) were experimentally determined. Various x-ray tube voltages ranging from 20 to 140 kV (employed in x-ray mammography, general radiography, and computed tomography) were used. In addition the behaviour of GSO: Ce under low energy gamma irradiation could be estimated from these data. Measurements were performed using an experimental set-up based on a photomultiplier coupled to an integration sphere. The GSO:Ce optical emission spectrum was measured under x-ray excitation using an optical grating monochromator. The efficiency of GSO:Ce was found to increase with increasing x-ray tube voltage, while the GSO:Ce spectrum, peaking at 440 and 490 nm, was found compatible with most optical detectors (photodiodes, photocathodes, charge coupled devices). However the spectral compatibility (0.77) and the effective efficiency were found highest for GSO coupled to photodiode.**

I. INTRODUCTION

Scintillators are materials, which emit light when exposed to ionizing radiation (X-rays, γ-rays etc). They are frequently used as radiation detectors in various medical imaging systems, used in diagnostic radiology and nuclear medicine (such as x-ray radiography, x-ray mammography computed tomography, gamma camera etc.). They are employed in powder, ceramic or single-crystal form, often coupled to optical photon detectors such as (photocathodes and photodiodes).

Out of the various scintillators employed in medical imaging modalities, particular interest is being paid on gadolinium oxyorthosilicate (Gd_2SiO_5 or GSO) often doped with cerium (Ce^{3+}) ion activator. In single crystal form, GSO:Ce was firstly produced in 1983 by Takagi and Fukazawa [1]. This crystal is of high density ($\rho = 6.71\ g/cm^3$), high effective atomic number ($Z(Gd) = 59$) and hence, high radiation detection index ($\rho Z_{eff}^4 = 84 \times 10^6$). In addition, due to the presence of Ce^{3+} ion activator, GSO:Ce exhibits a very fast response with a decay time of 60ns.

GSO:Ce has been previously studied under excitation with electrons, γ-rays [2], hard energy x-rays (above 500 keV) [3], charged particles [4], alpha and beta particles [5] as well as under UV excitation [6]. GSO:Ce has already been used in some positron emission imaging detectors.

The purpose of the present study was to examine the variation of luminescence emission efficiency of Gd_2SiO_5:Ce scintillator at various photon energies, lower than those corresponding to positron-electron annihilation. This was achieved in order to investigate the possibility of using GSO:Ce in imaging modalities other than positron emission (x-ray computed tomography, general x-ray imaging, nuclear imaging with low energy emitters). In addition the behaviour of this crystal under low energy gamma irradiation could be estimated from these data.

II. MATERIALS AND METHODS

A Gadolinium oxyorthosilicate crystal of dimensions $10mm \times 10mm \times 10mm$ doped with cerium 0,5% mol was supplied by Hitachi Chemical Co. Ltd [7]. This crystal was evaluated by experimentally determining the following luminescence parameters: (i) The absolute luminescence efficiency, (ii) the spectral matching factor and (iii) the effective efficiency.

Scintillators are often characterized by their absolute luminescence efficiency (AE) [8], which may be defined [9]-[11] as the ratio of the light energy flux (Ψ) emitted by an excited scintillation crystal over the incident X-ray or γ-ray exposure rate ($\dot{X}$) according to the equation:

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

where η_A represents the AE in units of [μW·s/mR·m^2]. AE is related to the radiation detection sensitivity of a scintillator. Of interest is to use high AE scintillator crystals, which may significantly reduce patient radiation dose burden required for various medical imaging examinations.

When a scintillation crystal is to be incorporated into a medical imaging detector a major consideration is the spectrum of the emitted light and its spectral compatibility with the spectral sensitivity of various optical photon detectors. Spectral compatibility can be estimated by the spectral matching factor (SMF), which is defined by the ratio [12], [13]:

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

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 light.

Effective efficiency (EE) is defined as the product of AE by the spectral matching factor corresponding to a specific scintillator-optical photon detector combination. That is:

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

where η_{Eff} is the effective efficiency (EE) of scintillator-optical detector block and α_S is the spectral matching factor of the optical detector [14]. Equation (3) provides information for the overall efficiency of the scintillator-optical detector system.

GSO:Ce was irradiated by X-rays in a Philips Optimus X-ray unit (tungsten target) and in a G.E. Senographe DMR Mammography unit (molybdenum target). Various X-ray tube voltages from 22 to 140kV were employed. The beam was filtered by 20mm of aluminium (general radiography) or by 30mm of Perspex (mammography), in order to simulate beam hardening by patient's body. The X-ray exposure rate was measured at the crystal's position using Radcal 2026C dosimeter. Light energy flux measurements were performed as follows. The light emitted by the irradiated crystal was measured by a calibrated photomultiplier (EMI 9798B) equipped with an extended sensitivity S-20 photocathode. The crystal was positioned on the integration sphere (Oriel 70451) input port whereas the calibrated photomultiplier was adapted on the integration sphere's output port (Fig. 1.). Integration sphere reduces experimental errors due to illumination non-uniformities. The photocathode of the photomultiplier was directly connected to a Cary 401 vibrating reed electrometer. This was performed in order to avoid electronic noise amplification due photomultiplier's dynode high voltage. The exact light flux of the crystal was determined by performing the following corrections on the experimental data:

(i) Correction by dividing with the integration sphere throughput (τ). The latter is defined as the ratio of the total light flux (Ψ_e) exiting sphere's output port to the total flux (Ψ_i) at the input port. τ depends on the sphere's reflectance properties, diameter, and number of ports.

$$\tau = \frac{\Psi_e}{\Psi_i} = \frac{\rho A_e / A_s}{(1 - \rho(1 - A_p / A_s))} \qquad (3)$$

where A_e is the area of the exit port, A_s is the surface area of sphere, A_p is the sum of all port areas and ρ is the sphere wall reflectance ($0 \leq \rho \leq 1$). The calibration of the integration sphere was achieved using prototype light sources, LEDs (Kingbright Company), from violet to red colour. All LEDs selected, had a viewing angle 2ϑ (where intensity drops to 50%) of 20 degrees in average. LEDs spectra were measured using an optical grating spectrometer (Ocean Optics Inc., S2000) and the total emitted light flux was measured using a calibrated photomultiplier (EMI 9798 B). Each LED was mounted on the one port of the integration sphere whereas on the other port was mounted the calibrated photomultiplier. The selected energy in μW per mA of current passing through the prototype LED was measured for each LED. The average of the above measurements found a sphere throughput of approximately 15.6 %, which is lower than the one given by the integration sphere manufacturer's datasheet (~23%) [15]. This was expected, since portion of energy is absorbed in the interfacing adapter from the sphere port to the photomultiplier.

ii) Measurements were multiplied by the ratio of the total light flux emitted by the crystal towards the side of the photomultiplier coupled to the sphere, to the light flux collected by the photocathode's sensitive area. This ratio was calculated by taking into account the crystal's emissive area, the distance between the crystal and the photomultiplier, the photocathode's sensitive area and the angular distribution of the emitted light; angular distribution was approximated by a Lambertian one i.e. following the $\cos\,\vartheta$ law were ϑ is the angle of light emission.

The spectral matching factor was determined using (2). The scintillator's emitted light spectrum $S_P(\lambda)$, of GSO:Ce crystal was measured using an Ocean Optics (Ocean Optics Inc., S2000) optical grating monochromator. Measurements were performed using both X-ray and UV excitation (Fig. 4(a), 4(b)).

A fibre optic 1.0m long (Avantes Inc. FCB-UV400-2) was transferring the light from the scintillating crystal to the spectrometer. The UV source (D_2 UV lamp, Perkin-Elmer) was positioned 10cm away from the crystal with approximately 45^0 angle with the crystal's surface. The X-ray tube was positioned 3cm over the scintillation crystal. Tube voltage was set primarily to 100kV, 100mA and then to 140kV, 63mA with 1sec exposure time. Light signal degradation due to fibre optic was taken into account. $S_D(\lambda)$ (spectral sensitivity) data were obtained from corresponding manufacturer's (Hamamatsu, EMI, etc.) datasheet.

Four optical photon detectors currently used in a large variety of X-ray and digital imaging detectors (digital radiography, computed tomography, nuclear medicine) and their spectral matching factor with GSO (Table I), namely an Si/S1133 Hamamatsu crystalline silicone photodiode, two a-Si:H 104H/108H amorphous silicone photodiodes corresponding to intrinsic layer thickness of 400nm (104H) and 800nm (108H), an extended S20 EMI photocathode and a GaAsP Hamamatsu photodiode, were examined.

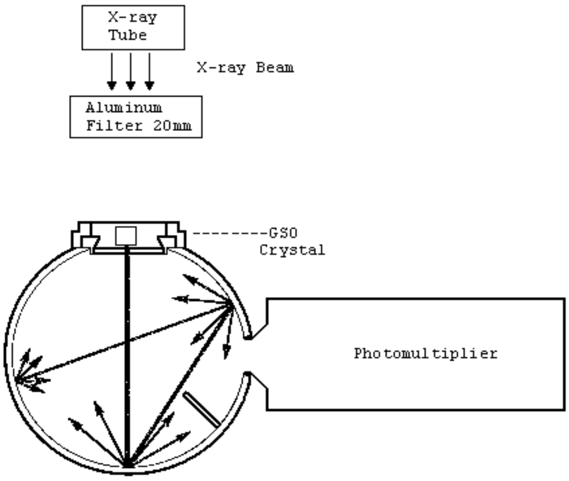

Fig. 1. Experimental set up for absolute efficiency measurement, using integration sphere.

III. RESULTS AND DISCUSSION

The variation of measured absolute efficiency of the GSO:Ce crystal with x-ray tube voltage using the general radiography unit is shown in Fig. 2.

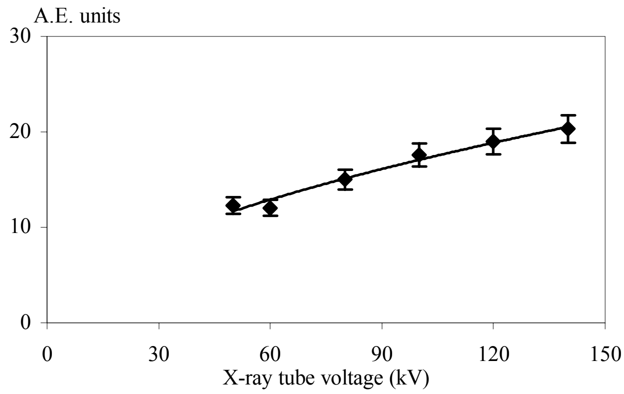

Fig. 2. Variation of absolute efficiency (AE) of GSO:Ce crystal for X-ray tube voltages between 40 kV and 140 kV. AE units: μW s/mR m^2.

Fig. 3 shows variation of absolute efficiency (AE) of the GSO:Ce crystal with x-ray tube voltages using the mammography unit.

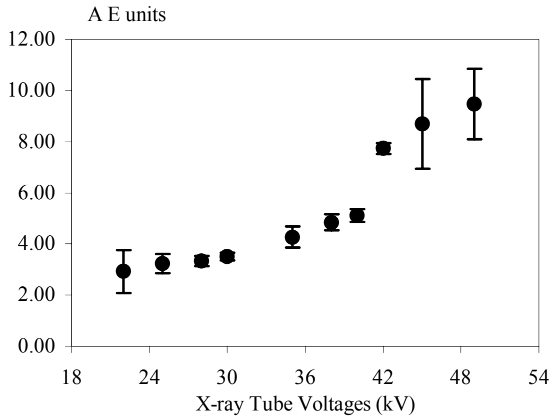

Fig. 3. Absolute efficiency of GSO:Ce crystal under Mammography exposure. AE units: μW s/mR m^2.

As it may be shown in Fig. 2, 3 absolute efficiency increases continuously with increasing x-ray tube voltage. The curve in Fig. 2 is of similar shape to data curves obtained by others using electron beam excitation and mono-energetic photon excitation [2], [4], [17]. This variation of absolute efficiency is explained by considering that, while x-ray tube voltage increases, the x-ray beam penetrates deeper within the scintillator block. Hence light is generated at points closer to the scintillator – optical detector interface. Thus light attenuation effects are of lower importance within scintillator's mass and the emitted light intensity is higher. Fig. 3 shows the variation of absolute efficiency in the mammographic energy range from 22 to 40 kV (molybdenum anode tube). 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. The absolute efficiency values obtained under both excitation conditions are comparable to corresponding values of the gadolinium oxysulfide (GOS) scintillator [8], [9] currently used in a large variety of x-ray imaging modalities

Fig. 4(a) and 4(b) show the optical emission spectrum of GSO: Ce crystal obtained under UV (a) and X-ray (b) excitation. As it may be observed the two spectra are similar. This was expected since the energy level scheme of the Ce^{+3} ion activator ($^2F_{5/2}$ and $^2F_{7/2}$ levels) remains the same in both types of excitation. The shape of the GSO: Ce emission spectrum plotted in Fig. 4 (a) and 4(b) matches the experimental results previously obtained by others [16].

Fig. 5 shows effective efficiency data corresponding to four optical detectors (Si and a-Si photodiodes, S20 and GaAsP photocathodes). As it may be observed highest effective efficiency was obtained for the a-Si:H photodiode.

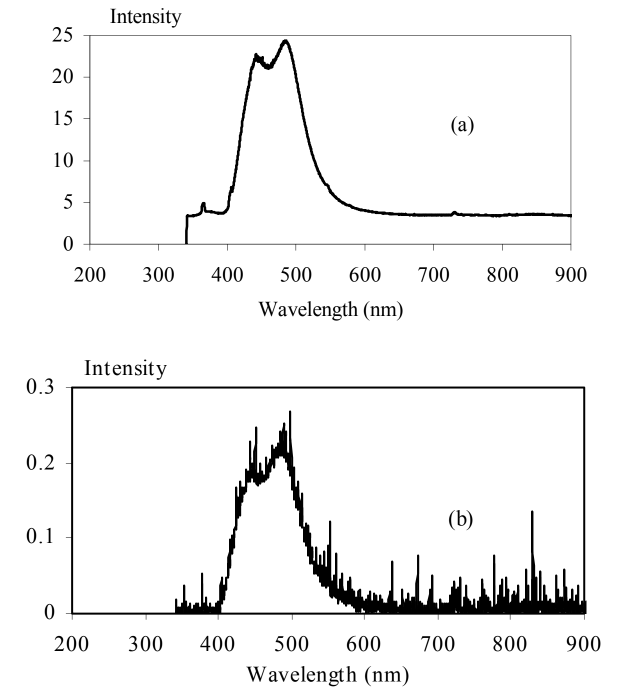

Fig. 4. (a) GSO:Ce optical spectrum at 20°C, under UV (D_2 lamp, Perkin-Elmer) excitation, (b) GSO:Ce optical spectrum at 20°C under X-ray (140kV, 63mA, 1sec, Philips Optimus) excitation.

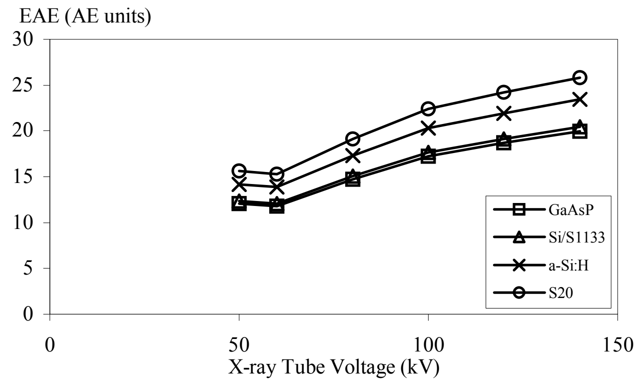

Fig. 5. Variation of effective efficiency of GSO:Ce with four optical detectors -○-S20, -x-a-Si:H, -Δ-Si/S1133 and -□-GaAsP.
AE units: μW s/mR m^2.

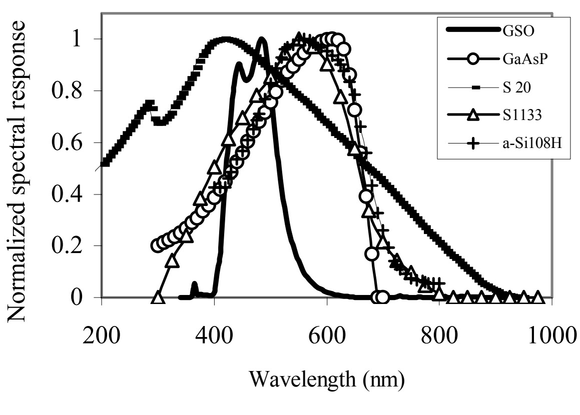

Fig. 6. Normalized light emission spectrum of GSO:Ce and spectral sensitivities of some optical photon detectors.

Fig. 6 is a plot of GSO:Ce emission spectrum measurements including three spectral sensitivity curves corresponding to photodetectors currently used in a large variety of conventional X-ray and digital imaging detectors (digital radiography, computed tomography, nuclear medicine). As it is observed in Fig. 6, GSO:Ce spectrum is lying well within the sensitivity limits of a-Si:H 104H/108H, Si/S1133, GaAsP photodiodes and S20 photocathode. This is also reflected from the results of the calculated SMFs in Table I, where all values are higher than 0.5.

TABLE I.
Spectral matching factor of GSO:Ce with optical detectors.

Optical Detectors	GSO:Ce
S-20 EMI	0.774
a-Si:H 104H/108H	0.756
Si/S1133 Hamamatsu	0.689
GaAsP Hamamatsu	0.659

IV. CONCLUSIONS

In conclusion, our measurements on the absolute efficiency of GSO: Ce shows that, under conditions employed in the present study, the light output of this scintillator increases with increasing x-ray photon energy. The values of absolute efficiency were found of the same order of magnitude to those of the currently employed GOS scintillator. In addition the emission spectrum, extending from 400 to over 550 nm and peaking at 440 and 490 nm, is well situated within the spectral sensitivities of all four optical detectors (photodiodes, photocathodes, charge coupled devices) considered in this study (Fig. 6). This is also reflected in the spectral matching factor values in Table I, which are all higher than 0.5. GSO:Ce was found with highest effective efficiency (Fig. 5) when combined to silicon a-Si:H photodiode. Taking into account the very short scintillation decay time of GSO:Ce, this material could be appropriate for imaging applications requiring relatively high x-ray tube voltages (e.g. computed tomography detectors, pixelated scintillation digital detectors, nuclear medicine imaging detectors etc.). GSO:Ce may also be considered for use in mammographic applications mainly because of the high absorption efficiency at low energies due to its high effective atomic number ($Z(Gd) = 59$) and its high density ($\rho = 6.71\ g/cm^3$).

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