Investigation of the radiation absorption and light emission properties of a 25 mg/cm^2 Lu_2SiO_5:Ce (LSO) scintillating screen for use in x-ray digital mammography detectors

C. Michail[1,2], S. David[1,2], I.Valais[1,2], D. Nikolopoulos[1], I. Sianoudis[3], C. Nomicos[4], N. Dimitropoulos[5], G. Panayiotakis[2], D. Cavouras[1] and I. Kandarakis[1*]

[1] *Department of Medical Instruments Technology, Technological Educational Institution of Athens Ag. Spyridonos, Aigaleo, 122 10 Athens, Greece*

[2] *Department of Medical Physics, Medical School, University of Patras, 265 00 Patras, Greece*

[3] *Department of Physics, Chemistry and Materials Technology, Technological Educational Institution of Athens Ag. Spyridonos, Aigaleo, 122 10 Athens, Greece*

[4] *Department of Electronics, Technological Educational Institution of Athens, Ag. Spyridonos, Aigaleo, 122 10 Athens, Greece*

[5] *Department of Medical Imaging, "Euromedica" Medical center, Mesogeion 2, Athens, Greece*

[*] Corresponding author: kandarakis@teiath.gr

Abstract. *The aim of the present study was to examine the radiation absorption efficiency and the light emission efficiency of Lu_2SiO_5:Ce (LSO) powder scintillator under x-ray mammographic imaging conditions. LSO is an efficient and extremely fast scintillator, employed in positron emission tomography, which however has never been used in X-ray imaging. For the purposes of the present study a 25 mg/cm^2 thick scintillating screen was prepared in our laboratory, by sedimentation of Lu_2SiO_5:Ce powder. Absolute luminescence efficiency measurements were performed within the range of X-ray tube voltages (22-40 kVp) used in mammographic applications. Parameters related to X-ray detection, i.e. the energy absorption efficiency and the quantum detective efficiency were calculated. A theoretical model, describing radiation and light transfer, was employed to fit experimental data and to estimate values of the intrinsic conversion efficiency and the light attenuation coefficients of the screen. The Lu_2SiO_5:Ce powder scintillator material showed to exhibit high radiation absorption properties in comparison with Gd_2O_2S:Tb and CsI:Tl and considering its very short decay time, it may successfully be employed in digital mammography.*

Keywords: Inorganic Scintillators; Radiation detectors; Luminescence efficiency; Mammography.

1 INTRODUCTION

Inorganic scintillators (or phosphors) coupled to optical sensors (photodiodes, photocathodes, CCD arrays, films etc) are employed in a variety of radiation detectors used in medical imaging systems [1]. Cerium (Ce^{3+}) doped scintillators are of particular interest for medical imaging, because of their very fast response. The latter is dominated by an efficient $5d \rightarrow 4f$ electronic transition in the Ce^{3+} ion [2-3]. Lutetium oxyorthosilicate Lu_2SiO_5:Ce (LSO), discovered by Melcher and Schweitzer in 1992, has attracted a great attention due to its high effective atomic number, high density (7.4 g/cm^3), fast decay time (40 ns) and high light yield (26000 light photons/MeV)[4,5], suitable emission wavelength (420 nm) and excellent chemical stability compared to other scintillators. LSO:Ce, in single-crystal form, has been already used in detectors of Positron Emission Tomography (PET) scanners, in nuclear and high energy physics experiments and in environmental monitoring [5,6]. In the present study LSO:Ce scintillator, in powder form (phosphor), was examined under exposure conditions employed in mammography. Measurements were performed on a granular phosphor screen, prepared in our laboratory, using an X-ray mammography unit emitting molybdenum spectrum X-rays. To our knowledge, LSO:Ce has never been previously studied under mammographic exposure conditions. Absolute luminescence efficiency measurements were performed for various X-ray tube voltages (22-49 kVp) used in mammographic applications. Parameters related to X-ray detection such as the energy absorption efficiency (EAE) and the quantum detection efficiency (QDE) were calculated. A theoretical model, describing radiation and light transfer through scattering media was used to fit experimental data[8,9,18,19]. Intrinsic conversion efficiency and light attenuation coefficients of the screen were derived through this fitting. Emitted spectrum and spectral compatibility to optical sensors was determined by performing light emission spectrum measurements and by taking into account the spectral sensitivity of various optical sensors.

2 MATERIALS AND METHODS

2.1. Theory

The light energy flux Ψ_λ emitted by a phosphor screen irradiated by an X-ray energy flux Ψ_X , may be given as follows:

$$\overline{\Psi}_\lambda = \int_0^{E_0} \overline{\Psi}_X(E)\overline{\eta}_\varepsilon(E)\eta_c \int_0^{W_0} \overline{\psi}_Q(E,w)\overline{g}_\lambda(\sigma,w)dwdE \tag{1}$$

where E_0 is the maximum energy of the X-ray spectrum, W_0 is the coating thickness of the phosphor, E is the X-ray photon energy, η_ε is the energy absorption efficiency (EAE), η_c is the intrinsic X-ray to light conversion efficiency (ICE), expressing the fraction of absorbed X-rays converted into light within the phosphor material. Ψ_Q is a function giving the probability of an interacting X-ray photon of energy E to be absorbed at a depth $W<W_0$. g_λ is the fraction of light photons, created at depth W, that escape the phosphor and σ is an optical parameter accounting for the attenuation of light within the phosphor, which is called the reciprocal of the light diffusion length [7, 8, 9]. The second integral in (1) expresses the fraction of light photons, created at various depths within the phosphor, that escape the phosphor layer. This fraction is defined as the light transmission efficiency (LTE) of the phosphor [8, 10-12]. The first integral denotes integration over the energies of the X-ray spectrum (molybdenum target / molybdenum or aluminum filter). Mean values in (1) express averaging over the area of the detector. The absolute luminescence efficiency (η_A) of a phosphor is given by the relation [13]:

$$\eta_A = \dot{\Psi}_\lambda / \dot{X} \tag{2}$$

where $\dot{X}$ is the exposure rate incident on the phosphor.

QDE is defined as the fraction of incident photons interacting with the scintillator. For polyenergetic X-rays the QDE of a scintillator layer of coating thickness W is calculated as follows:

$$QDE(E) = \frac{\int_0^{E_0} \Phi_0(E)(1 - e^{-(\mu_{tot,t}(E)/\rho)W})dE}{\int_0^{E_0} \Phi_0(E)dE} \tag{4}$$

where Φ_0(E) is the X-ray photon fluence (photons per unit of area) incident on the scintillator. The spectrum of Φ_0(E) may be given in terms of the X-ray energy fluence spectrum. $\mu_{tot,t}$ is the X-ray total mass attenuation coefficient of the scintillator. EAE is the fraction of the locally absorbed energy, representing more accurately the efficiency of a detector to capture the useful X-ray imaging signal (i.e., the spatial distribution of primary X-ray absorption events). EAE may be calculated by the relation (5):

$$EAE(E) = \frac{\int_0^{E_0} \Phi_0(E)E\left(\frac{\mu_{tot,en}(E)}{\mu_{tot,t}(E)}\right)(1-e^{-(\mu_{tot,t}(E)/\rho)W})dE}{\int_0^{E_0} \Phi_0(E)EdE} \quad (5)$$

where $\mu_{tot,en}$ is the total mass energy absorption coefficient of the scintillator. This coefficient includes all mechanisms of energy deposition locally at the point of X-ray interaction within the scintillators mass. All secondary photons (e.g. K-characteristic fluorescence X-rays created just after the primary interaction effect, Compton scattering photons) are assumed to be lost.

2.2. Experiments

LSO:Ce was purchased in powder form (Phosphor Technology Ltd, code: ZBK58/N-S1) with mean grain size of approximately 8 μm. The size of the grain is very crucial for both phosphor efficiency [14] and image resolution. The latter is degraded with increasing phosphor grain while efficiency increases at the same time. However, it is generally accepted that sizes in the range from 5 to 10 μm give a satisfactory compromise between resolution and efficiency [15,16]. The phosphor was used in the form of thin layer (test screen) to simulate the intensifying screens employed in X-ray mammography. For the purposes of the present study, a 25 mg/cm^2 thick scintillating screen was prepared by sedimentation of Lu_2SiO_5:Ce powder on a fused silica substrate (spectrosil B). During the sedimentation process, sodium orthosilicate (Na_2SiO_3) was used as binding material between the powder grains [17]. The phosphor screen was exposed to X-rays on a General Electric Senographe DMR Plus mammographic unit, employing X-ray tube voltages ranging from 22 to 49 kVp with molybdenum anode target and molybdenum filter. The filter changed automatically from molybdenum to rhodium (Rh) and aluminum (Al) as tube voltage increased. The X-ray beam was filtered by a 30 mm thick block of Perspex to simulate beam hardening by human breast [18]. Tube voltage was checked using an RMI model 240 multifunction meter. Incident exposure rate measurements were performed using a Radcal 2026C ionization chamber dosimeter (Radcal Corp. USA) at the powder phosphor's position. The absolute luminescence efficiency was determined, according to (2), by performing X-ray exposure and light flux measurements. Absolute luminescence efficiency expressed in units $\mu W \times m^{-2}/(mR \times s^{-1})$ or Efficiency units (E.U.). Light flux measurements were performed in transmission mode, where the non-irradiated screen side's light was measured. Transmission mode simulates X-ray-imaging modalities such as, digital detectors, image intensifiers, front screens of radiographic cassettes and computed tomography detectors [19]. Light flux, $\dot{\Psi}_\lambda$, measurements were performed using an experimental setup comprising a light integration sphere (Oriel 70451) coupled to a photomultiplier (EMI 9798B) connected to a Cary 401 vibrating reed electrometer. Light flux measurements were also corrected for the spectral mismatches between the emitted light and the spectral sensitivity of the photocathode (S20) of the photomultiplier. To determine both the mean light photon energy $\overline{E}_\lambda$ and the spectral matching factor α_S, the emitted light of the LSO:Ce powder phosphor was measured by an Ocean Optics HR 2000 optical spectrometer (Ocean Optics Inc.), while the spectral sensitivities of the optical detectors were obtained from manufacturers data. Spectrometer light measurements were performed under X-ray excitation. The light emitted by the irradiated powder LSO:Ce phosphor was transferred to the spectrometer through a 2.0 m long, 400 μm fiber optic, (Avantes Inc. FCB-UV400-2, Colorado, USA). Corrections for light signal degradation due to fiber optic light losses were taken into account. To estimate the compatibility of emitted light with spectral sensitivity of the photodetector, the spectral matching factor α_S was calculated by the relation (3):

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

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

2.3. Theoretical calculations

Using relations (1) and (2) absolute luminescence efficiency may be calculated as a function of the intrinsic physical properties of the phosphor material. The physical quantities employed in relation (1) were expressed as follows: (i) Incident X-ray energy flux Ψ_X was expressed and calculated using a theoretical model [20] that describes the energy spectral distribution of the X-rays produced by a molybdenum target X-ray tube. (ii) The quantum detection efficiency (QDE), η_Q, as well as the energy absorption efficiency (EAE), η_ε, were calculated by considering exponential X-ray absorption within the phosphor material, determined by the X-ray absorption and attenuation coefficients and the thickness of the phosphor layer. These coefficients of the LSO:Ce powder phosphor were calculated using the corresponding coefficients of Lu, Si, O, obtained from data as tabulated by NISTR [21]. (iii) The absorption probability function Ψ_Q, was calculated by the formula (6):

$$\overline{\psi}_Q(E, w) = \frac{\overline{\Psi}_X(\mathrm{E})\mu(\mathrm{E})\exp[-\mu(\mathrm{E})w]dw}{\int_0^{w_0} \overline{\Psi}_X(\mathrm{E})\mu(\mathrm{E})\exp[-\mu(\mathrm{E})w]dw} \tag{6}$$

where the numerator gives the probability for an X-ray photon to be absorbed at depth w within the phosphor and the denominator gives the total probability of X-ray absorption within the whole phosphor layer. $\mu_{(E)}$ is the X-ray absorption coefficient of the phosphor material. (iv) g_λ, giving the fraction of light escaping to the output per X-ray absorbed, was expressed by the formula (7)[8, 9]

$$\overline{g}_\lambda(\sigma, w) = \frac{\rho_1[(\beta+\rho_0)e^{\sigma w} + (\beta-\rho_0)e^{-\sigma w}]}{(\beta+\rho_0)(\beta+\rho_1)e^{\sigma w_0} - (\beta-\rho_0)(\beta-\rho_1)e^{-\sigma w_0}} \tag{7}$$

where σ is the light attenuation coefficient of the phosphor, which is equal to the reciprocal of the light photon diffusion length [8-10] and it is given as a function of the optical scattering coefficient *(s)* and the optical absorption coefficient *(α)*:

$$\sigma = [\alpha(\alpha + 2s)]^{1/2} \tag{8}$$

ρ_0, ρ_1 are optical parameters expressing the reflection of light at the front and back phosphor surfaces defined as:

$$\rho_n = (1 - r_n)/(1 + r_n), \quad n = 0,1, \tag{9}$$

where ρ_n denotes the optical reflection coefficients at the front (0) and back (1) screen surfaces. β is an optical parameter which is equal to ρ, corresponding to the case of a very thick phosphor with no light transmission through it. β has been also expressed as a function of α and s [8]:

$$\beta = [\alpha/(\alpha + 2s)]^{1/2} \tag{10}$$

β and ρ_n were determined as described previously [8,12-13]. These data were used in order to fit relation (1) to the experimental absolute luminescence efficiency measurements. Using the Levenberg-Marquardt technique [22], best fit was obtained, for specific values of the parameters η_c and σ, in relations (1) and (2). These values, together with β and ρ_n, were then adopted as the intrinsic optical properties of the phosphor. All secondary photons, e.g., K-characteristic fluorescence X-rays were not taking into consideration because the highest atomic number (Z) element, Lutetium, in the LSO:Ce scintillator has the K-absorption energy at 67keV ,which exceeds the examined energy region.

3 RESULTS AND DISCUSSION

Figure 1 shows the variation of absolute luminescence efficiency of the LSO:Ce screen with X-ray tube voltage. Points represent experimental data. A better fit to the data was obtained for η_c values varying from η_c=0.09 to 0.11 and σ varying from σ=71 to 75cm^{2}g^{-1}. The value of the intrinsic conversion efficiency (η_c), as estimated by the fitting, is lower than the corresponding value of Gd_2O_2S:Tb (η_c=0.2) phosphor [23, 24], which is used in conventional and digital radiographic detectors. It is approximately equal to the η_c values of CsI:Na, CsI:Tl and NaI:Tl phosphors (η_c=0.10), used in a large variety of radiation detectors [25]. The optical attenuation coefficient σ was found higher than that of Gd_2O_2S: Tb phosphors (σ =30cm^{2}g^{-1}). This may be explained by considering that the lower mean wavelength light of LSO:Ce (420 nm) exhibits higher attenuation than the light emitted by Gd_2O_2S:Tb (545 nm). An important observation from Figure 1 is that absolute luminescence efficiency maintains high values within a range of X-ray tube voltages from 25 to 36 kVp. This property is of interest for mammographic imaging.

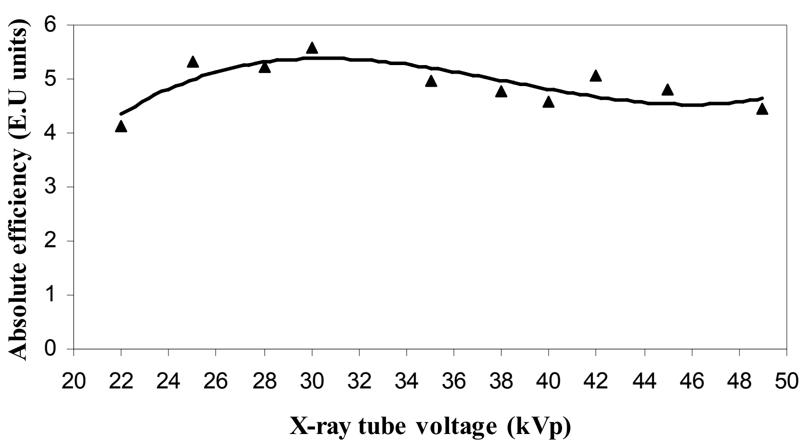

Figure 1 Variation of the absolute luminescence efficiency of the LSO:Ce powder phosphor with X-ray tube voltage. Points correspond to experimental values. Efficiency units: $\mu W \times m^{-2} / (mR \times s^{-1})$

Figure 2 and 3 illustrate the variation of calculated QDE and EAE with X-ray tube voltage for the 25 mg/cm^2 LSO:Ce screen. For comparison purposes similar calculations were performed for Gd_2O_2S:Tb and CsI:Tl screens. The first point to note is that EAE differs significantly from QDE. The differences between EAE and QDE show that only a fraction of the total amount of radiation energy detected is locally imparted (EAE). Hence, only this fraction contributes to accurate spatial registration of photons and accurate image formation. As it may be seen, at 28 kVp, EAE (0.62) is approximately 15% lower than QDE (0.73). Calculations also showed that the LSO:Ce powder scintillator has approximately 10% higher values of

QDE and 4.5% higher values of EAE than Gd_2O_2S: Tb. CsI:Tl scintillator has the lowest X-ray detection in this region of energy.

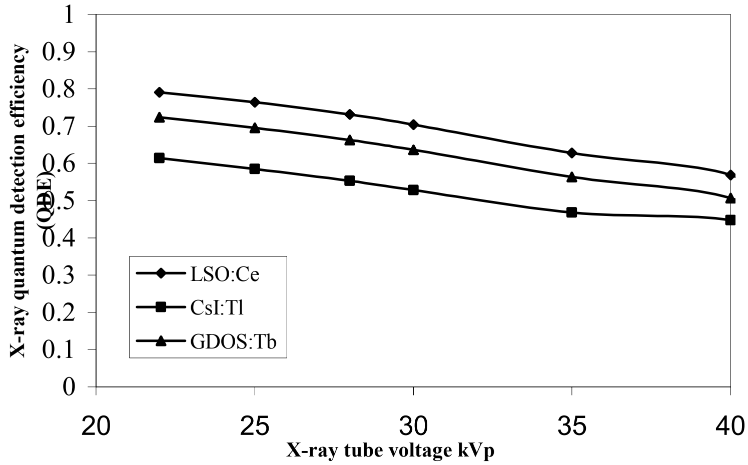

Figure 2 Variation of calculated QDE of LSO:Ce, CsI:Tl and Gd_2O_2S:Tb with X-ray tube voltage for 25 mg/cm^2 powder screens.

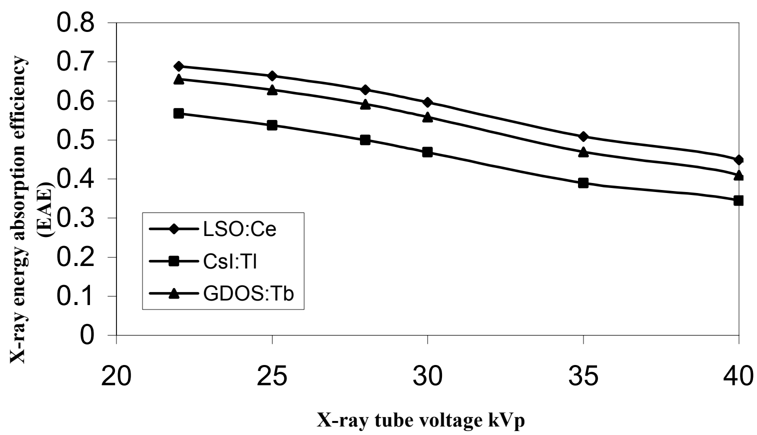

Figure 3 Variation of theoretical QDE of 25 mg/cm^2 LSO:Ce, CsI:Tl and Gd_2O_2S:Tb phosphor screens with X-ray tube voltage.

Figure 4 shows the measured light emission spectrum of the LSO:Ce phosphor under excitation by X-rays of 30 kVp tube voltage. The peak value of the light spectrum was found at 420 nm. The long tail on the right part of the spectrum should be ascribed to the $5d \rightarrow 4f$ electronic transitions of the Ce^{3+} ion, located at the Ce2 center. The Ce2 emission, much weaker and not well resolved (as compared to the Ce1 emission at 393-423 nm), occurs at 500 nm[5] . It is of significance to note these measurements were conducted at room temperature.

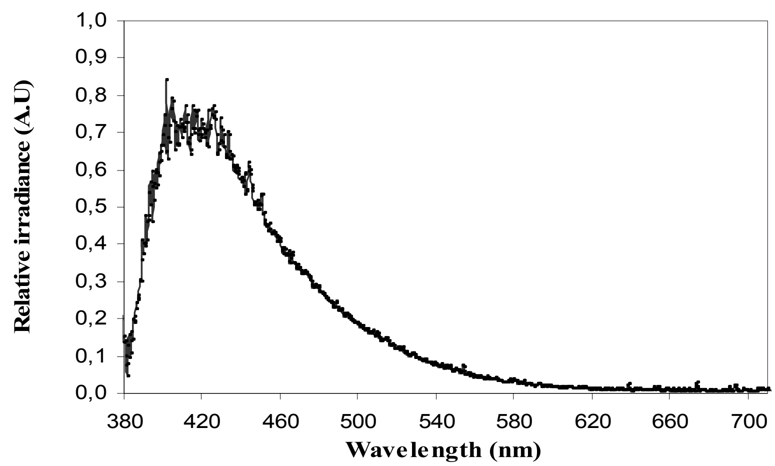

Figure. 4 Optical emission spectrum of LSO:Ce phosphor measured at 30 kVp for $25 mg/cm^2$ coating thickness.

Table 1 shows the values of the spectral matching factors of the LSO:Ce calculated according to relation (3). LSO:Ce exhibits excellent compatibility with the AgfaGS and KodakGR radiographic films. In addition it was found adequately compatible with the Amorphous Silicon (AmorSi) photodiode.

Optical Detectors	Lu_2SiO_5:Ce
GaAs	0.916
Si	0.320
AmorSi	0.580
MAMORAY	0.873
E/S 20	0.965
AgfaGS	0.960
KodakGR	0.965
FujiUM	0.896

Table 1 : Spectral matching factors of the LSO:Ce screen

4 CONCLUSIONS

In the present study, a LSO:Ce powder scintillator screen of 25 mg/cm^2 coating thickness was prepared and examined under X-ray mammographic conditions. The X-ray quantum detection efficiency and the X-ray energy absorption efficiency were found higher than currently employed materials (e.g. Gd_2O_2S:Tb and CsI:Tl) for detection of X-rays used in mammographic applications. The absolute luminescence efficiency maintains high values, within the mammographic energy range, while the intrinsic conversion efficiency was found close to that of CsI:Tl but lower than that of Gd_2O_2S:Tb. The emission spectrum of LSO:Ce screen showed excellent spectral compatibility with currently used detectors. Taking also into account its very fast response this phosphor could be considered for applications in X-ray mammographic imaging systems.

Acknowledgments

The project is co-funded by the European Social Fund & National Resources-EPEAEK II-ARXIMIDIS.

REFERENCES

[1] J. A. Rowlands, J. Yorkston. (2000) In *Handbook of Medical Imaging, Vol. 1, Physics and Psycophysics,* ed. by J.Beutel, H.L. Kundel, R.L. Van Metter, SPIE Press, Bellingham.
[2] Van Eijk, C. W. E. (2002), "Inorganic scintillators in medical imaging", Phys. Med. Biol., Vol. 47, pp. 85-106
[3] G. Blasse, B.C. Grabmaier. (1994) Luminescent Materials, Springer-Verlag. Berlin, Heidelberg.
[4] C. L. Melcher, J. S. Schweitzer (1992), Nucl. Instrum. Methods Phys. Res., A Vol.314, pp.212-213.
[5] C.L. Melcher, M. Schmand, M. Eriksson, et al., (2000) IEEE Trans. Nucl.Sci. Vol. 47 pp.965–968.
[6] E. Bescher, S.R. Robson . J.D. Mackenzie, B. Patt, J. Iwanczyk.E.J. Hoffman, (2000) "*New Lutetium Silicate Scintillators",* Journal of Sol-Gel Science and Technology Vol.19, pp.325–328,
[7] D. Engelkemeir (1956), "Nonlinear response of NaI(Tl) to photons".Rev. Sci. Instr. Vol. 27, pp.589-591.
[8] G.W.Ludwig (1971), "X-Ray Efficiency of Powder Phosphors", J. Electrochem. Soc., 118, pp. 1152-1159.
[9] Swank, R.K., (1973), "Calculation of modulation transfer functions of X-ray fluorescent screens". Appl. Opt. Vol.12, pp.1865-1866.
[10] H. Hamaker (1947), "Radiation and heat conduction in light scattering material", Philips. Res. Rep. Vol.2, pp.55-56.
[11] I. Kandarakis, D. Cavouras, E. Kanellopoulos, G. Panayiotakis, C.Nomicos (1998), "Experimental determination of detector gain, zero frequency detective quantum efficiency, and spectral compatibility of phosphor screens: comparison of CsI:Na and Gd2O2S:Tb for medical imaging applications", Nucl. Instr. Meth. Phys. Res. A. Vol. 417, 86.
[12] I. Kandarakis, D. Cavouras, P. Prassopoulos, E. Kanellopoulos, C.Nomicos, G. Panayiotakis (1999), "Evaluating Zn2SiO4: Mn phosphor for use in medical imaging radiation detectors". Appl. Phys. A. Vol.67, pp.521-525.
[13] D. Cavouras, I. Kandarakis, C. Nomicos, A. Bakas, G. Panayiotakis (2000), "Performance evaluation of $(Gd;La)_2O_2$:Tb phosphor for medical imaging applications under X ray excitation", Radiat. Meas. Vol.32, pp.5-7.
[14] J. Lindstrom, G.A. Carlsson, (1999), "A simple model for estimating the particle size dependence of absolute efficiency of fluorescent screens", Phys. Med. Biol. Vol.44, pp.1353-1354.
[15] B.A. Arnold (1979), "Physical characteristics of screen-film combinations. In: Haus, A.G. (Ed.), The Physics of Medical Imaging: Recording System, Measurements and Techniques. American Association of Physicists in Medicine, New York, pp. 30-33.
[16] A. M. Gurwich (1995), "Luminescent screens for mammography". Radiat. Meas. Vol.24, pp.325-328
[17] I. Kandarakis, D. Cavouras, G. Panayiotakis, C.Nomicos (1997), "Evaluating x-ray detectors for radiographic applications: A comparison of ZnSCdS:Ag with Gd_2O_2S:Tb and Y_2O_2S:Tb screens", Phys. Med. Biol., Vol. 42, pp. 1351-1373.

[18] I. Kandarakis, D. Cavouras, D. Nikolopoulos, A. Anastasiou, N. Dimitropoulos, N. Kalivas, E. Ventouras, I. Kalatzis, C. Nomicos, G. Panayiotakis (2005), “Evaluation of ZnS:Cu phosphor as X-ray to light converter under mammographic conditions” Rad. Meas. Vol. 39, pp.263-275
[19] I. Kandarakis, D. Cavouras, C. Nomicos, G. Panayiotakis (2001), “X-ray luminescence of ZnSCdS:Au, Cu phosphor using X-ray beams for medical applications”, Nucl. Instr. Meth. Phys. Res. B., Vol. 179, pp. 215-224.
[20] D.M. Tucker, G.T. Barnes, D.B. Chakraborty (1991), “Semi-empirical model for generating tungsten target X-ray spectra”. Med. Phys. Vol.18, pp. 211-215.
[21] E. Storm, H. Israel (1967). “Photon cross-sections from 0.001 to 100 MeV for elements 1 through 100”. Report LA-3753, Los Alamos Scientific Laboratory, University of California, CA.
[22] W. H. Press, B. P. Flannery, S. A. Teukolsky, W. T. Vetterling (1990), “Numerical Recipes in C: The Art of Scientific Computing”. Cambridge University Press, Cambridge.
[23] G E Giakoumakis, C D Nomicos, E N Yiakoumakis and E K Evangelou, (1990) “Absolute efficiency of rare earth oxysulphide screens in reflection mode observation”, Phys. Med. Biol., , Vol. 35, No 7, pp.1017-1023.
[24] G. E. Giakoumakis (1991), “Matching factors for various light-source-photodetector combinations”, Appl. Phys. A Vol.52, pp.7-9.
[25] G.E Giakoumakis and D M Miliotis, (1985), “Light angular distribution of fluorescent screens excited by x-rays”, Phys. Med. Biol., Vol. 30, No. I , pp.21-29.