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Structural Characterization and Absolute Luminescence Efficiency Evaluation of Gd₂O₂S High Packing Density Ceramic Screens Doped with Tb³⁺ and Eu³⁺ for further **Applications in Radiology**

Anna Dezi^{1*}, Elenasophie Monachesi¹, Michela D'Ignazio¹, Lorenzo Scalise², Luigi Montalto², Nicola Paone², Daniele Rinaldi³, Paolo Mengucci³ George Loudos⁴, Athanasios Bakas⁵, Christos Michail⁶, Ioannis Valais⁶, Christine Fountzoula⁷, George Fountos⁶, Stratos David^{4, 6}

¹Faculty of Engineering, Università Politecnica delle Marche, Ancona, Italy

²Dipartimento di Ingegneria Industriale e Scienze Matematiche, Università Politecnica delle Marche, Ancona, Italy

³Dipartimento SIMAU, Università Politecnica delle Marche, Ancona, Italy

⁴Department of Biomedical Engineering, Technological Educational Institute of Athens, Athens, Greece

⁵Department of Radiology & Radiation Therapy, Technological Educational Institute of Athens, 12210 Athens, Greece

⁶Radiation Physics, Materials Technology and Biomedical Imaging Laboratory, Department of Biomedical Engineering, Technological Educational Institute of Athens, Egaleo, 12210 Athens, Greece

⁷Department of Medical Laboratories, Technological Educational Institute of Athens, 12210 Athens, Greece

Corresponding Author: David Stratos, sdavid@teiath.gr

Abstract. Rare earth activators are impurities added in the phosphor material to enhance probability of visible photon emission during the luminescence process. The main activators employed are rare earth trivalent ions such as Ce⁺³, Tb⁺³, Pr³⁺ and Eu⁺³. In this work, four terbium-activated Gd₂O₂S (GOS) powder screens with different thicknesses (1049 mg/cm², 425.41 mg/cm², 313 mg/cm² and 187.36 mg/cm²) and one europium-activated GOS powder screen (232.18 mg/cm²) were studied to investigate possible applications for general radiology detectors. Results presented relevant differences in crystallinity between the GOS:Tb doped screens and GOS:Eu screens in respect to the dopant agent present. The AE (Absolute efficiency) was found to rise (i) with the increase of the X-ray tube voltage with the highest peaking at 110kVp and (ii) with the decrease of the thickness among the four GOS:Tb. Comparing similar thickness values, the europium-activated powder screen showed lower AE than the corresponding terbium-activated.

Introduction

The investigation for luminescence materials, able to absorb X-rays efficiently and convert their energy into optical photon energy, starts as direct consequence of the Rontgen's discover in 1857 that introduces the X- rays in the field of Medical Imaging. During the years, the luminescence materials were utilized for α and γ radiation and become object of important applications. At present the radiation detectors are formed by two main parts: the scintillator (phosphor) that basically emit light when exposed to ionizing radiation (X-rays, γ -rays) [1] and the photo-detector that converts the emitted light into the useful diagnostic image. Among several efficient materials, the Gd₂O₂S (GOS) host showed the highest figure-of-merit in luminescence properties, due to its high light yield, elevated density and effective atomic number [2]. To enhance the probability of visible photon emission during the luminescence process, small amount of impurity, called activators, is commonly added to

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IOP Conf. Series: Journal of Physics: Conf. Series 931 (2017) 012029

inorganic scintillators. The main activators employed are rare earth (RE) trivalent ions as Tb^{+3} , Pr^{3+} , Ce^{+3} and Eu^{+3} [3].

The purpose of the present study, is to examine various features of five GOS scintillators with several thicknesses and two different dopant agents (Tb^{+3} and Eu^{+3}), formulate a structural characterization and evaluate the luminescence efficiency of them.

Materials and methods

The five scintillators investigated in this study are under the form of high packing density ceramic screens: four terbium-activated Gd_2O_2S (GOS:Tb) screens with different thicknesses (1049 mg/cm², 425.41 mg/cm², 313 mg/cm² and 187.36 mg/cm²) and one europium-activated Gd_2O_2S (GOS:Eu) screen (232.18 mg/cm²). Each screen has been prepared using phosphor powders mixed with Potassium Bromide (KBr) powder in various coating thicknesses using a hydraulic press under vacuum, following the methodology of disk preparation in Infrared Spectroscopy.

The structural analysis was carried out through an X- ray powder diffractometer (Bruker D8 Advance), equipped with a copper anode tube (0.15418 nm) to analyze the variation of crystallinity of the powder screens and phosphor material in function of different amounts of chemical components. Scanning electron microscope (FESEM ZEISS SUPRA 40) was employed for distribution investigation of micro particle belonging to the phosphor material. This instrumentation allows a high-resolution observation of the three-dimensional topography of the sample with the possibility of obtaining also compositional information and images.

The fluorescence emitted light energy flux of the phosphor screens was measured 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. The excitation of the screens was conducted by the X-ray tube with rotating Tungsten anode (BMI General Medical Merate) and inherent filtration equivalent to 2 mm Al. This machine operates at various X-ray tube voltages, from 50 to 140 kVp, and it was used to simulate clinical irradiation conditions used in General Radiology. An alternative 20mm of Al filter was added to simulate beam hardening by human body [4].

2.1. Calculation of radiation detection parameters

The efficiency of a scintillator to detect photons is described by the quantum detection efficiency (QDE). QDE is the fraction of incident photons interacting with the scintillator mass described in [5]. For polyenergetic X-rays the QDE of a scintillator layer of coating thickness w is calculated according:

$$QDE(E) = \left[\int_0^{E_0} \Phi_0(E) \left(1 - e^{-\left(\frac{\mu_{tot,t}(E)}{\rho}\right) W} \right) dE \right] \left(\int_0^{E_0} \Phi_0(E) dE \right)^{-1}$$
(1)

where E_0 is the maximum energy of X-ray spectrum and $\mu_{tot,t}$ (E) / ρ is the X-ray total mass attenuation coefficient of the scintillator. $\Phi_0(E)$ is the X-ray photon fluence (photons per unit of area) incident on the scintillator [4].

X-ray imaging detectors are energy integrating systems, i.e., their output signal is proportional to the X-ray energy absorbed within the scintillator. Hence, when evaluating X-ray imaging systems, the calculation of the energy absorption efficiency (EAE) is also of importance. EAE may be calculated by the relation [4]:

$$EAE(E) = \left[\int_{0}^{E_{0}} \Phi_{0}(E)E(\frac{\mu_{tot,en}(E)}{\mu_{tot,t}(E)})\left(1 - e^{-\left(\frac{\mu_{tot,t}(E)}{\rho}\right)W}\right)dE\right]\left(\int_{0}^{E_{0}} \Phi_{0}(E)dE\right)^{-1}$$
(2)

Where $\Psi_0 = \Phi_0(E)E$ is the incident X-ray energy fluence and $\mu_{tot,en}$ is the total mass energy absorption coefficient of the scintillator. $\mu_{tot,en}$ includes all mechanisms of locally energy deposition at the first point of X-ray interaction within the scintillator's mass. All secondary photons, e.g., k-characteristic fluorescence X-rays, created just after the primary interaction effect, are assumed to be lost. Thus EAE, being a measure of the locally absorbed energy, represents more accurately the efficiency of a

IOP Conf. Series: Journal of Physics: Conf. Series 931 (2017) 012029

detector to capture the useful X-ray imaging signal (i.e., the spatial distribution of primary X-ray absorption events) [6].

The QDE and EAE can be increased by making the scintillator/phosphor screen thicker or by using materials which have higher values of atomic number and density. The radiation detection will in general be highest at low energies, and gradually decreasing with increasing energy following the exponential law [7]. If the material has an atomic absorption edge (i.e. k-absorption edge) in the energy region of interest, then the QDE increases dramatically above this energy, causing a local minimum in QDE for energies immediately after the absorption edge [8,9].

2.2. Experimentally determined quantities

Scintillators or phosphors are often characterized by their absolute luminescence efficiency (ALE), which may be defined [9] as the ratio of the light energy flux ($\dot{\Psi}_{\lambda}$) emitted by an excited scintillator or a phosphor over the incident X-ray or γ radiation exposure rate \dot{X} according to the equation [9]:

$$ALE = \dot{\Psi}_{\lambda} \left(\dot{X} \right)^{-1} \tag{3}$$

The absolute luminescence efficiency is expressed in AE units $\left[\frac{\mu W \cdot m^{-2}}{mR \cdot s^{-1}}\right]$ [10]

When a phosphor scintillator 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= a_s), which is defined by the ratio [11]:

$$a_{s} = \left(\int S_{p}(\lambda)S_{D}(\lambda)d\lambda\right)\left(\int S_{p}(\lambda)d\lambda\right)^{-1}$$
(4)

where S_P is the light emitted spectrum by the scintillator, S_D is the spectral sensitivity of the optical photon detector and λ denotes the wavelength of the light [10].

Results and discussions

3.1. X-ray powder diffraction (XRD) analysis of the Gd₂O₂S screens

The microanalysis, done on the five powder screens, provides slightly different spectra. In figure 1 is illustrated two different XRD spectra regarding the Gd_2O_2S :Tb and Gd_2O_2S :Eu screens. The very narrow peaks for each corresponding chemical element provides that Gd_2O_2S material is chemically pure and very well crystallized. No dopant agent is found during the microanalysis mainly because the percentage of dopant inside the Gd_2O_2S phosphor material is too small (<1%). Even if the dopant is not visible, it modifies the crystallinity way of grow and consequently the position of the peaks of the Gd_2O_2S :Eu (figure 1b) compared to Gd_2O_2S :Tb.



Figure 1. a) Gd_2O_2S :Tb b) Gd_2O_2S :Eu. Structural analysis and study of the crystallinity. Two components are visible in the graph: in red the Gd_2O_2S and in blue the KBr. No presence of the dopant has been found. X axis is the 2-theta angle. Y axis is the normalized counts of the diffracted X-rays.

3.2. Homogeneity investigation of Gd_2O_2S powder screens

The mean areas and the standard deviations (SDs) coming from the analysis on the six SEM images, an example in figure 2, of the phosphor material, show an inhomogeneity in dimension of the particles due to the significant SDs, figure 3.



Figure 2. Characteristic SEM picture of Gd₂O₂S: Tb phosphor screen.



Figure 3. Mean area values with relative standard deviations

*3.3. X-ray detection efficiency in term of EAE and QDE in Gd*₂*O*₂*S powder screens*

Figure 4 illustrates the variation of calculated EAE and QDE with X-ray tube voltages (ranging from 40 to 140 kVp) for the five powder screens. Both EAE and QDE decreased with increasing energy and increased with increasing coating thickness. At low X-ray tube voltages, the thicker screen (1049 mg/cm²) absorbs large fractions of incident X-ray energy (e.g. 0.9 at 50 kVp). At higher voltages X-ray photons are more penetrating and X-ray energy absorption is lower (e.g. 0.55 at 90 kVp, 1049 mg/cm² screen).



Figure 4. a) Variation of calculated EAE of Gd_2O_2S screens with various coating thicknesses. **b)** Variation of calculated QDE of Gd_2O_2S screens with various coating thicknesses. The points represent the calculated values

The difference in magnitude between EAE and QDE shows 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.

3.4. ALE of Gd_2O_2S powder screens in the radiographic range (50-130 kVp) and spectral emission

In figure 5a the variation of absolute luminescence efficiency of the Gd_2O_2S powder phosphors with X-ray tube voltages used in radiographic applications, is presented. Points correspond to experimental values. The absolute luminescence efficiency is determined by the fluorescence emitted light energy flux measurements of each phosphor screen and incident exposure rate measurements.

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IOP Conf. Series: Journal of Physics: Conf. Series 931 (2017) 012029



Figure 5. a) Variation of absolute luminescence efficiency of the Gd_2O_2S screens with X-ray tube voltage. The vertical axis has E.U values (1 E.U equal to $\mu W \cdot m^{-2}/mR \cdot s$). b) Normalized spectral response of S-20 EMI photocathode together with normalized emission spectra of the Gd_2O_2S :Tb and Gd_2O_2S :Eu respectively in green and in red.

The absolute luminescence efficiency is found to increase for all screens continuously with increasing X-ray tube voltage up to 110 kVp. After the value of 90kVp, ALE values show a tendency to saturate especially in case of thinner screens. The thinner screen of 187.36 mg/cm² is found to exhibit highest ALE values at 110 kVp equal to 9 E.U. Absolute efficiency is found to decrease with increasing coating thickness of the Gd₂O₂S:Tb for the screens under investigation. The Gd₂O₂S:Eu screen compared with similar thickness values of Gd₂O₂S:Tb screen, shows lower ALE values.

Figure 5b shows the normalized emission spectra of the Gd_2O_2S powder phosphor screens doped with terbium (green dots) and with europium (red dots). Moreover, in the same graph the normalized spectral sensitivity of the extended S-20 EMI photocathode, utilized in this study for the emitted light energy flux measurements used in ALE calculation is illustrated. The spectral matching factor was calculated equal to 0.61 for Gd_2O_2S :Eu and 0.78 for Gd_2O_2S :Tb [12].

Conclusions

In conclusion, Gd_2O_2S :Tb and Gd_2O_2S :Eu powder screens developed by mixed with Potassium Bromide (KBr) using hydraulic press under vacuum, has perfect crystallinity with inhomogeneity's in the surface phosphor particle dispersion. The different dopant agents - even in a very small amount - it changes the crystallinity of the phosphor screens. The absolute luminescence efficiency of Gd_2O_2S :Tb and Gd_2O_2S :Eu screens under General Radiography conditions shows that increases with increasing X-ray photon energy but do not increase with the increasing of coating thickness, showing that thinner screens may exhibit higher ALE values. Finally, the Gd_2O_2S :Eu screen compared with similar thickness values of Gd_2O_2S :Tb screen, shows lower ALE values. In future works, we will try to construct thinner screens with the same technique solving the problem of its mechanical stability maybe by fused on a silica substrate for mechanical support. Possible applications of the analysed phosphor screens could be radiography applications to obese patient and industrial radiography in which hard X-rays are utilized and consequently thicker screens are needed to stop them.

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References

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- Valais I, Kandarakis I, Nikolopoulos D and Cavouras D 2004 IEEE Nucl. Sci. Symp. Conf. Rec. 5 2737.
- [2] Nikl M 2006 Meas. Sci. Technol. 17 37
- [3] Rodney P A 1997 Physical Processes in Inorganic Scintillators (CRC Press Boca Raton).
- [4] Michail C, David S, Bakas A, Kalyvas N, Fountos G, Kandarakis I and Valais I 2015 Radiat. Meas. 80 1.
- [5] Boone J M 2000 Handbook of Medical Imaging vol 12, ed Beutel J, Kundle H J and Van Metter RL (Bellingham: SPIE Press)
- [6] David S, Michail C, Seferis I, Valais I, Fountos G, Liaparinos P, Kandarakis I and Kalyvas N 2016 J. Lumin. 169 706.
- [7] David S. 2006 Master thesis (Patras).
- [8] Yaffe M J and Rowland J A 1997 Phys Med Biol. 41 39.
- [9] David S, Michail M, Valais I, Nikolopoulos D, Liaparinos P, Kalivas N, Kalatzis I, Efthimiou N, Toutountzis A, Loudos G, Sianoudis I, Cavouras D, Dimitropoulos N, Nomicos C, Kandarakis I and Panayiotakis G 2007 Nucl. Instrum. Meth. Phys. Res. A 571 346.
- [10] Michail C, Fountos G, Liaparinos P, Kalyvas N, Valais I, Kandarakis I and Panayiotakis G 2010 Med. Phys. 37(7) 3694.
- [11] Kandarakis I, Cavouras D, Nomicos C D and Panayiotakis G S 2001 Nucl. Instr. Meth. Phys. Res. B. 117 215.
- [12] Michail C, Valais I, Toutountzis A, Kalyvas N, Fountos G, David S, Kandarakis I and Panayiotakis G 2008 *IEEE Trans. Nucl. Sci.* **55(6)** 3703.