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## Journal of Luminescence

journal homepage: [www.elsevier.com/locate/jlumin](http://www.elsevier.com/locate/jlumin)On the response of GdAlO<sub>3</sub>:Ce powder scintillatorsC. Michail<sup>a,\*</sup>, N. Kalyvas<sup>a</sup>, I. Valais<sup>a</sup>, S. David<sup>a</sup>, I. Seferis<sup>b</sup>, A. Toutountzis<sup>a</sup>, A. Karabotsos<sup>c</sup>, P. Liaparinos<sup>a</sup>, G. Fountos<sup>a</sup>, I. Kandarakis<sup>a</sup><sup>a</sup> Department of Medical Instruments Technology, Technological Educational Institute of Athens, 122 10 Athens, Greece<sup>b</sup> Department of Medical Physics, Medical School, University of Patras, 265 00 Patras, Greece<sup>c</sup> Department of Conservation of Antiquities and Works of Art, Technological Educational Institute of Athens, 122 10 Athens, Greece

## ARTICLE INFO

## Article history:

Received 7 February 2013

Received in revised form

18 June 2013

Accepted 27 June 2013

Available online 4 July 2013

## Keywords:

Inorganic scintillators

Radiation detectors

GdAlO<sub>3</sub>:Ce

## ABSTRACT

The aim of the present study was to investigate the luminescence efficiency (XLE) of gadolinium aluminum perovskite (GdAlO<sub>3</sub>:Ce) powder scintillator. This powder phosphor, also known as GAP:Ce scintillator, is a non-hygroscopic material, emitting blue light with short decay time. For the purposes of this study, five scintillating screens with coating thicknesses, 14.7, 31.0, 53.7, 67.2 and 121.1 mg/cm<sup>2</sup>, were prepared in our laboratory from GdAlO<sub>3</sub>:Ce powder (Phosphor Technology, Ltd) by sedimentation on silica substrates. The light emitted by the phosphors under investigation was evaluated by performing measurements of the absolute luminescence efficiency (AE), X-ray luminescence efficiency and detector quantum gain (DQG) under X-ray exposure conditions with tube voltages ranging from 50 to 140 kV. The quantum detection efficiency (QDE) and energy absorption efficiency (EAE) were also evaluated. The spectral compatibility of GdAlO<sub>3</sub>:Ce, with various existing optical detectors, was investigated after emission spectra measurements. A theoretical model, describing radiation and light transfer, was used to fit experimental AE data. This has allowed the estimation of optical attenuation coefficients of the scintillator. GdAlO<sub>3</sub>:Ce exhibited higher QDE and EAE values, compared to aluminium perovskite (YAlO<sub>3</sub>:Ce) but lower absolute efficiency values. Absolute efficiency was found to increase with increasing X-ray tube voltage, although for values higher than 120 kVp a decrease was observed.

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## 1. Introduction

Most medical imaging detectors are based on scintillator–optical detector (photodiodes, photocathodes, films etc.) combinations. Cerium (Ce<sup>3+</sup>) doped scintillators or phosphors exhibit the property of very fast response. The latter is dominated by the very efficient 5d → 4f electronic transitions in the Ce<sup>3+</sup> ion [1–4]. Previous studies have shown that yttrium aluminium perovskite (YAlO<sub>3</sub>:Ce) also known as YAP:Ce has attractive properties [5,6]. On the other hand gadolinium based scintillators (e.g. Gd<sub>2</sub>O<sub>2</sub>S:Tb) are widely used in X-ray imaging applications. Using gadolinium (Gd) which is heavier than yttrium (Y), higher absorption efficiency is expected. In cerium doped gadolinium aluminium perovskite (GdAlO<sub>3</sub>:Ce also known as GAP) powder scintillator, yttrium has been replaced by gadolinium [7–14]. GAP:Ce has been studied thoroughly in the past [7–18], however, it has never been tested under X-ray radiography conditions. It has been used in electronics as a dielectric layer for flash memory devices [19], as a light converting material substrates for use in light emitting diode (LED) substrates [20] and optical ceramic materials [21,22]. Initial

luminescence results were published by our group in the past [7]. In the present study, a systematic investigation of the GdAlO<sub>3</sub>:Ce was performed. Absolute luminescence efficiency measurements were performed for various X-ray tube voltages (50–140 kVp). Parameters related to X-ray detection such as the energy absorption efficiency (EAE) and the quantum detective efficiency (QDE) were calculated. Emitted spectrum and spectral compatibility to optical sensors were determined by performing light emission spectra measurements and by taking into account the spectral sensitivity of the optical detectors. Quality metrics such as quantum gain (DQG) was estimated. An analytical model was used to predict optical properties of the GdAlO<sub>3</sub>:Ce scintillator [5,23].

## 2. Materials and methods

## 2.1. Calculations

## 2.1.1. Attenuation coefficients for compounds

Attenuation coefficients for compounds (materials comprised of ≥ 2 elements) can be determined as the weighted average (by mass) of the individual mass attenuation coefficients of the

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compound's constituent elements, as

$$\left(\frac{\mu}{\rho}\right)_{\text{compound}} = \sum_{i=1}^N m_i \left(\frac{\mu}{\rho}\right)_i \quad (2.1)$$

where  $m_i$  is the mass fraction (fraction of the element's mass contribution to the total mass) and  $(\mu/\rho)_i$  is the mass attenuation coefficient of element  $i$  in the compound. This is important for estimating attenuation probabilities of compounds and materials that cannot be easily measured and particularly for computer simulations [24,25].

### 2.1.2. Quantum detection and energy absorption efficiency (QDE & EAE)

The efficiency of a scintillator to detect photons is conventionally described by the quantum detection efficiency (QDE), which is defined as the fraction of incident photons interacting with the scintillator mass [1]. However accurate X-ray detection may be determined by considering only those X-ray photons that deposit an amount of energy in the phosphor mass. This is because only these X-rays can generate light signals (scintillations) which can be detected by the optical sensor and contribute to image formation. The fraction of energy depositing photons is expressed through the energy absorption efficiency (EAE). QDE as well as EAE were evaluated analytically [1] as described in previous studies [26]. The required values for the total attenuation and the total energy absorption coefficients of GdAlO<sub>3</sub>:Ce scintillator were calculated from tabulated data of energy absorption and attenuation coefficients of gadolinium, aluminum and oxygen [27,28].

## 2.2. Experiments

GdAlO<sub>3</sub>:Ce was purchased in powder form (Phosphor Technology Ltd, England, code: UM58#9438) with a mean grain size (estimated by ultrasonic dispersion with a coulter counter having 100 μm aperture) of approximately 8.9 μm at the 95% of the volume and quartile deviation of 0.32 (Phosphor Technology Ltd., datasheet). GdAlO<sub>3</sub>:Ce has  $Z_{\text{eff}} = 56.2$ , refractive index = 2.02 [21] and a very fast decay time of the order of a few ns [29]. The forbidden energy band-gap between the valence and the conduction energy bands of the GdAlO<sub>3</sub>:Ce scintillator material is  $E_g = 5$  eV [2,3,30,31].

Particle size and morphology parameters of the GdAlO<sub>3</sub>:Ce powder phosphor were verified via scanning electron microscope (SEM) micrographs using the Jeol JSM 5310 scanning electron

microscope (SEM) collaborating with the INCA software. Gold was used to obtain a figure from a site of interest of the GdAlO<sub>3</sub>:Ce specimen. For the elementary particle analysis carbon thread evaporation process was used. Carbon was flash evaporated under vacuum conditions to produce a film suited for the GdAlO<sub>3</sub>:Ce SEM specimen in a BAL-TEC CED 030 carbon evaporator ( $\sim 10^{-2}$  mbar). The phosphor was used in the form of thin layers to simulate the intensifying screens employed in X-ray imaging. Five screens from 14.7 to 121.1 mg/cm<sup>2</sup> thick were prepared by sedimentation of GdAlO<sub>3</sub>:Ce powder on fused silica substrates (spectrosil B). The screen coating thicknesses correspond to thicknesses calculated thicknesses of 39.2, 82.67, 143.2, 179.2 and 322.9 μm assuming a density of 7.5 g/cm<sup>3</sup> and a packing density of 50% [5,17]. Sodium orthosilicate (Na<sub>2</sub>SiO<sub>3</sub>) was used as binding material between the powder grains [26].

The effect of the fused silica substrates (spectrosil B) on the emitted light of the GdAlO<sub>3</sub>:Ce powder phosphors was also investigated by transmission and absorption measurements. The purpose of these measurements was to confirm that the emission wavelength of the phosphors does not influence drastically the absorption and scattering properties of the substrate. The transmission measurement was carried out with a Perkin-Elmer Lambda 15 UV/vis spectrophotometer.

The phosphor screens were exposed to X-rays on a Philips Optimus radiographic unit, with a dual-focus rotating tungsten anode, employing X-ray tube voltages ranging from 50 to 140 kVp. Tube filtration was 2.5 mm Al. An additional 20 mm filtration was introduced in the beam to simulate beam quality alternation by a human body [32].

### 2.2.1. Absolute efficiency (AE)

The light emission efficiency of a phosphor may be experimentally estimated under X-ray imaging conditions, by determining the absolute luminescence efficiency (AE) defined by Eq. (2.2):

$$\eta_A = \dot{\Psi}_\lambda / \dot{X} \quad (2.2)$$

where  $\dot{\Psi}_\lambda$  is the emitted light energy flux (energy of light per unit of area and time),  $\dot{X}$  is the incident exposure rate that excites the phosphor to luminescence. AE, is traditionally expressed, in units of  $\mu\text{W} \times \text{m}^{-2} / (\text{mR} \times \text{s}^{-1})$  thereafter referred to as efficiency units (E.U.). The S.I. equivalent of this unit is given in  $\mu\text{W} \times \text{m}^{-2} / (\text{mGy} \times \text{s}^{-1})$ , where mGy stands for the corresponding air Kerma. The light flux measurements were performed using an experimental set up comprising a light integration sphere (Oriell 70451) coupled to a photomultiplier (PMT) (EMI 9798B) which was

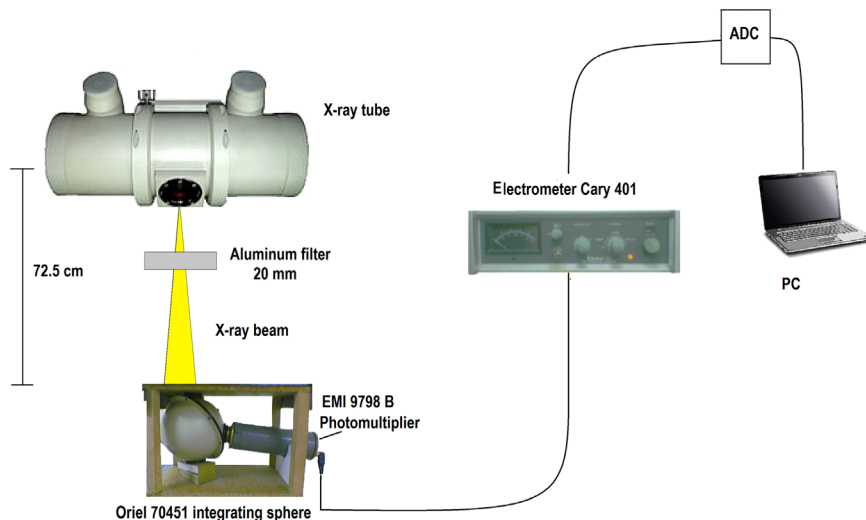


Fig. 1. Experimental set-up for the measurement of the emitted light energy flux comprising the integrating sphere, the PMT and the vibrating reed electrometer.

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