X-ray imaging resolution of phosphor screens prepared with different grains size and shape of granular Lu$_2$O$_3$:Eu

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Abstract. The influence of the grain shape and size on spatial resolution (ranging from nano to micro scale) of various Lu$_2$O$_3$:Eu phosphor screens was investigated. All screens were prepared using the sedimentation method. Three screens were prepared with spherical grains and sizes 50 nm, 200 nm and 5 $\mu$m, whilst two screens with rod-like shape grains and sizes 500 nm and 1-8 $\mu$m. All screens were coupled to a high resolution CMOS digital imaging sensor (Remote RadEye HR) consisting of 1200 x 1600 pixels with 22.5 $\mu$m pixel pitch. Experiments were performed under radiographic conditions, using 70 kVp tube voltage and 63 mAs tube load. Spatial resolution was assessed utilizing the Modulation Transfer Function (MTF). It was found that the influence of the grains shape on imaging performance was more crucial than the grain size. The rod-like grains showed very poor spatial resolution. The influence of grains size between 50 nm 200 nm and 5 $\mu$m was negligible on MTF values.

Keywords: Lu$_2$O$_3$:Eu; Nanophosphors; CMOS; X-ray imaging

1. Introduction

Indirect digital X-ray detectors, such as Complementary Metal Oxide Semiconductors (CMOS) and Charged Coupled Devices (CCDs), coupled with scintillating screens are used for many applications in medical imaging, airport security, industrial quality control and high energy physics [1]. The scintillating screens needs to fulfill characteristics, such as high density and high atomic number, to ensure efficient stopping power of ionizing radiation, but also high luminescence efficiency and low afterglow to ensure good energy resolution. The most traditionally used commercial scintillating materials are thallium doped cesium iodide (CsI:Tl), due to its good spatial resolution properties arising from columnar structure and terbium-doped gadolinium oxysulphide (Gd$_2$O$_2$S:Tb) due to its high X-ray absorption and light conversion efficiency [1-4]. Furthermore many scintillators have been investigated for X-ray imaging applications, such as Gd$_2$O$_2$S:Eu, Gd$_2$O$_2$S:Pr,Ce,F, Lu$_2$O$_3$:Eu, Gd$_2$O$_2$S:Pr employed in granular phosphors screens, and LuPO$_4$:Eu semitransparent thin films [5-9]. Apart from the intrinsic properties of the scintillating materials, the structural properties such as grain size and shape seem to influence image quality in order of light diffusion [10]. Nanomaterials have gain ground as radiation detectors with potential to reduce receiving dose by the operators or patients (in case of medical imaging) due to the increased luminescence efficiency and confinement [11,12]. On the other hand, the diffusion and propagation properties of light passing through nanoparticulated phosphors layers are still under investigation with ongoing theoretical and
experimental research, devoted to analysis of the physical and structural properties of those materials [13-18]. The aim of the current work is to provide a comparative investigation of the imaging performance of Lu₂O₃:Eu phosphors screens, prepared with different grain shape and size, and manufactured by the sedimentation method. More specific, three screens were prepared with spherical grains, and sizes 50 nm, 200 nm and 5 μm. Furthermore, two screens, were also prepared, with rod-like shape and sizes 500 nm and 1-8 μm. The screens were coupled to a CMOS optical sensor RadEye HR and the imaging performance was investigated through the Modulation Transfer Function (MTF). The experimental method was based on the guidelines published by the International Electrotechnical Commission (IEC) 62220-1-2:2005 [19]. Although this protocol has been replaced from IEC 62220-1-1:2015 [20,21] the first one was preferred for comparison purposes with previous results.

2. Materials and Methods

2.1. Nanophosphor screens preparation

Lu₂O₃:Eu Powders were synthesized by the following methods: a) the rod-like grains powders were synthesized by the hydrothermal method using ammonia for PH stabilization, b) the 200 nm grains by co-precipitation method using urea, c) the 5 μm grains powder was prepared by means of a Li₂SO₄ + SiO₂ flux method using a mixture of commercially offered Lu₂O₃ and Eu₂O₃, and d) the combustion method was used for the 50 nm grains powder, using glycine as the organic fuel. Afterwards, a suspension of the powder phosphor is prepared and allowed to sediment onto a substrate. For the preparation of the suspension, deionized water was used in which the above phosphor is practically insoluble. Sodium silicate water solution (Na₂SiO₃) or so called “waterglass” was used as binding material, which creates conditions of satisfactory adhesion of phosphorus layer. In addition the Na₂SiO₃ fill the voids between the grains. Subsequently, due to higher refractive index, compared with it of air, leads the emitting photons to refract with lower angle when they go through from one material to another (refractive index of Lu₂O₃=1.935, refractive index of Na₂SiO₃ = 1.53) [22]. The sedimentation was achieved by using a mixture consisting of 1000 ml of de-ionized water, 20 ml of Na₂SiO₃, and the appropriate amount of phosphor powder in a glass tube of 110 cm height. The substrate was placed at the bottom of the tube [23,24] and secured using a silicone optical compound (Visilox V-788) as an adhesive media between substrate and holder. This compound can be easily removed afterwards by using an alcohol based solution without leaving any light absorbing stains. The sedimentation is followed by the removal of fluid, slowly drying the screen without any movement for 24 hours, at room temperature. Afterwards was placed in an oven for 2 hours at a temperature of 150 °C, and then was slowly cooled at environment temperature. Five thin screens were prepared with thicknesses (coating density) 33.1 mg/cm² (200 nm spherical grains), 33.3 mg/cm² (50 nm spherical grains), 35.2 mg/cm² (5 μm spherical grains), 31.7 mg/cm² (500 nm rod-like grains) and 33.2 mg/cm² (1-8μm rod-like grains) on Borosilicate glass substrate 22x22 mm (Waldemar Knittel-GmbH) or round quartz substrate with diameter 30 mm.

2.2. Experimental Setup

Experiments were performed using a Philips Optimus X-ray unit. For the image quality measurements, an optical readout device including a CMOS Remote RadEye HR photodiode pixel array was used. The CMOS consists of 1200 X 1600 pixels with 22.5 μm pixel pitch and a fill factor of 0.8. The screens were held using a thin polyurethane foam layer for compression between the screens and a 1 mm thick graphite cover. The screens were directly coupled to the photodiode array, while at the top side neither absorptive nor reflecting layer was used. The experiments were carried out at 70 kVp X-ray energy and 63 mAs tube current product. The source to detector distance (SDD) was 180 cm. The exposure rate at the entrance surface of the experimental devices was measured with a calibrated dosimeter (Piranha RTI Electronics), while the photon fluence was measured with a portable Amptek XR-100T spectrometer.
2.3. Image quality - Modulation transfer function (MTF)

The modulation transfer function (MTF) was used to characterize the resolution properties of an imaging system, describing the variation of contrast with spatial frequency [25-27]. The MTF was measured by means of a PTW Freiburg tungsten edge test device. Images of the edge placed at a slight angle, were obtained. The edge spread function (ESF) was calculated by the extraction of a 1 x 1 cm² ROI, which covers a large portion of the active area of the CMOS sensor (2.7 x 3.6 cm²), with the edge roughly at the center. The ESF was differentiated to obtain the line spread function (LSF). Finally, the normalized LSF was Fourier transformed to give the pre-sampling MTF.

3. Results and Discussion

Figure 1 shows the MTF curves of all screens under the same exposure conditions. The higher MTF values are attributed to spherical grain screens. Comparing the MTF values of 50 nm and 200 nm spherical grain screens, it is obvious that the MTF values are comparable. Only at high frequencies from 7 to 12 cycles/mm the MTF of the 50 nm spherical grains screen appeared slightly higher. The 5 μm spherical grains screen appeared with slightly lower MTF values in comparison with the 50 nm and 200 nm spherical grains screens. To explain those differences the size dependence optical properties should be taken into account. Monte Carlo simulations of Mie scattering theory have looked into the dependence of light absorption probability on grain size [16]. For nano-sized grains the absorption probability is considered to be higher since the number of interactions of light is higher due to higher number of grains. For this reason those screens give rise to higher scattering of the emitted radiation compared to the screen made up of 5 μm grains. Thus, the lateral emitted photons become attenuated by scattering before they reach the screen edge and finally the detector. This may provide an advantage of nanograins where light diffusion is limited to lower solid angles within this layer and implies a more sharp angular distribution of light spread toward layer output. The MTF values for the rods-like grains screens are very low compared with the spherical grains screens. As mentioned above the optical properties as well as the spatial arrangement of the emitted light photons may affect the imaging resolution inversely. Another important reason which is certainly influencing the MTF values for those screens are the clusters created due to the sedimentation procedure. The phosphor grains do not precipitate uniform on the substrate surface, creating structural abnormalities with consequence the degradation of image resolution due to high structural noise.

Figure 1. MTF of Lu₂O₃:Eu scintillating screens coupled to the CMOS RadEye HR optical sensor.

4. Conclusions

In this paper the imaging resolution of Lu₂O₃:Eu nanophosphors screens were investigated. Screens with grains of different size and shape were prepared by the method of sedimentation. It was found that the influence of the grains shape on image resolution is more crucial than the grain size. Specifically, the rod-like grains shows very poor imaging resolution influenced by the non-uniformities of the screen. The spherical grain screens appeared with very good resolution properties for all frequencies range. The influence of grains size between 50 nm and 200 nm is negligible on
the MTF values. The 5 μm spherical grains screen appears with slightly lower MTF values for all frequency range, compared with the nano-sized spherical grains.

5. References
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