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Investigation of the imaging characteristics of the Gd_2O_3:Eu nanophosphor for high-resolution digital X-ray imaging system

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Abstract

For possible applications in high-resolution medical image systems, we manufactured a Eu^{3+}-doped Gd_2O_3 nanophosphor using the low-temperature solution combustion method, and evaluated its performance as an image sensor. The structural and optical characteristics of the fabricated nanophosphor were investigated using field-emission scanning electron microscopy (FE-SEM), X-ray diffraction (XRD), photoluminescence spectrum (PL), and luminescence decay time measurements. From the FE-SEM and XRD results, we established that the fabricated nanophosphor was formed of spherical particles of about 30–40 nm, and confirmed that the particles agglomerated as the sintering temperature increases. From PL spectra, a strong peak appeared near 611 nm, and the luminescent intensity was seen to be affected by the doping concentration of Eu. Gd_2O_3:Eu nanophosphor particles have shown the highest luminescent efficiency at a Eu concentration of 5 wt%. In the luminescent decay time measurements, the mean decay time was about 2.3–2.6 ms, about two times longer than that of the general bulk phosphor, and affected by the of Eu-doping concentration. For the investigation of the imaging characteristics of the fabricated nanophosphor, we connected the Gd_2O_3:Eu nanophosphor film to a commercial CMOS image sensor, obtained the X-ray images and evaluated the modulation transfer function (MTF) as a function of the Eu concentration. We were able to obtain high-resolution X-ray images and found that the Eu concentration also influenced the imaging characteristics.

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Keywords: Gd2o3:Eu nanophosphor; Digital X-ray imaging system; High resolution; CMOS image sensor

1. Introduction

The increased demand for high resolution in the field of digital X-ray imaging has stimulated research on X-ray conversion materials with high efficiency and high intrinsic resolution. These include phosphors that convert X-ray into light and visualize the internal structure of the human body, and the phosphor particle size becomes an important factor in determining the image resolution [1].

Recent research on nanophosphors using europium (Eu) activator have studied its application in display fields using fluorescent lamps and plasma display panels (PDP), etc. [2]. In some of this research, such nano sized phosphor particles are reported to be somewhat different in their electrical, optical, and structural characteristics. It is reported that these differences in electrical and optical characteristics of very small particles are caused by quantum effects due to their high surface to volume ratio, which increases the band gap by reduction of the number of allowable quantum states in the small particles, and improves surface and interfacial effects [3,4]. A phosphor with such small size particles can obtain high-resolution

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images by reducing the scattering of photons and at the same time by improving the luminescent efficiency.

In addition, the Gd_2O_3:Eu phosphor is reported to have high efficiency for radiation and to be excellent with respect to optical matching with image sensors (CMOS, CCD, etc.) [5]. Therefore, in this study, we evaluated its imaging performance by applying a Gd_2O_3:Eu nanophosphor film to a CMOS image sensor, and obtaining the X-ray images. Consequently, we propose the applicability of Gd_2O_3:Eu nanophosphors to digital X-ray imaging systems.

2. Materials and methods

2.1. Sample preparation

Gd_2O_3:Eu nanophosphor particles were synthesized using the low-temperature solution combustion method, using gadolinium acetate hydrate [$(CH_3CO_2)_3Gd$, 99.9%, Aldrich] and europium acetate hydrate [$(CH_3CO_2)_3Eu$, 99.999%, Aldrich] as starting materials. The synthesized Gd–Eu compounds are powders doped with 3, 5, 10, and 15 wt% of Eu, respectively, through sintering we manufactured nano crystals by maintaining the powder for 1 h at 500, 700, and 900 °C under oxygen. Using screen printing method, 5×5 cm sized, 200 °C thick Gd_2O_3:Eu nanophosphor films were manufactured for application in an imaging system.

2.2. Experimental method

2.2.1. Sample evaluations

Field-emission scanning electron microscopy (FE-SEM) (using a JSM-6500F electron microscope) was used to examine the shape, size and distribution of the phosphor particles to examine the morphology and the size of the synthesized phosphor particles, we also used an X-ray diffractometer (Rigaku DMAX-IIIA), with an analysis range 2θ of 20–80°. The luminescence characteristics of the synthesized nanophosphor were examined by measuring the photoluminescence spectra and decay time dependence on Eu concentration and sintering conditions using a PL spectrometer (FS900CDT). For the measurement of PL spectra, we excited the phosphor with ultraviolet light of 254 nm, generated from a xenon lamp, and passed the light emitted from the excited phosphor through a monochromator to a photomultiplier tube (PMT).

2.2.2. Imaging evaluations

For the X-ray response and image performance evaluation, a commercial CMOS image sensor (Shad-o-Snap™ 1024, Rad-icon) was used. The detector part of the CMOS device consists of a 1024×1000 photodiode array with an active area of 4.8×4.93 cm and a pixel pitch of 48 μm. In obtaining the X-ray response signals and X-ray images, the settings of the X-ray generator (Shimadzu TR-500-125) were 74 kVp, 128 mAs with additional 21 mm Al filter, the RQA 5 radiation quality defined in IEC for measuring the modulation transfer function (MTF) [7]. The distance to the CMOS-based image sensor was fixed at 150 cm. We measured the pre-sampling MTF after obtaining images using a slit camera (Nuclear Associates, Model: 07-624-2222), and, in order to obtain the composite line spread function (LSF), the slit camera was slightly tilted (about 1.5–3°). The analysis was carried out using MATLAB 6.5.

3. Results and discussion

3.1. Gd_2O_3:Eu nanophosphor particles

For the analysis of the growth characteristics, the morphology, size and distribution of Gd_2O_3:Eu nanophosphor particles, we used FE-SEM and X-ray diffraction (XRD) after sintering the Gd–Eu compound, synthesized using the solution combustion method, at 500, 700, and 900 °C, respectively. Fig. 1(a) shows the dried Gd–Eu compound and (b) shows the shape and the size of the Gd_2O_3:Eu nanophosphor particles after sintering. The average particle size was about 30–40 nm, in agreement with the XRD results, and spherical particles are agglomerated. Spherical particles not only reduce light scattering compared to particles of irregular types, and increase film brightness by increased packing density, but also improve the resolution of the image [6].

Fig. 2 shows the XRD pattern dependence on the sintering temperature. As shown in the figure, the powder sintered for 1 h at 500 °C has X-ray peaks at (2 2 2), (4 0 0), (4 4 0), and (6 2 2), and consistent with the cubic crystal structure of Gd_2O_3. The cubic crystal structure of the Gd_2O_3:Eu phosphor shows its peak near the luminescence wavelength of 611 nm. On the other hand, the powders sintered for 1 h at 700 and 900 °C have an XRD pattern that has a monoclinic phase part at 630 nm luminescence wavelength together with the cubic phase. Also, as the sintering temperature increases, the diffraction width reduces, while the intensity of the diffraction in such crystal state increases. These facts show that sintering temperature contributes significantly to particle size and luminescence intensity. The particle size of the phosphor powder was inferred from the Scherrer equation using the diffraction peak full-width at half-maximum (FWHM) [8]. The results indicate that the phosphor powder has diameter

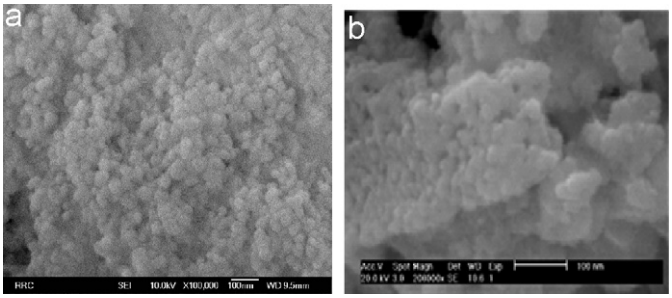

Fig. 1. FE-SEM images: (a) the dried Gd–Eu powder, (b) Gd_2O_3:Eu nanophosphor particle.