Synthesis and scintillation characterization of nanocrystalline Lu_2O_3(Eu) powder for high-resolution X-ray imaging detectors

2012 JINST 7 C03048

(http://iopscience.iop.org/1748-0221/7/03/C03048)

PUBLISHED BY IOP PUBLISHING FOR SISSA MEDIALAB

RECEIVED: *November 7, 2011*
ACCEPTED: *February 27, 2012*
PUBLISHED: *March 29, 2012*

THE 9th INTERNATIONAL CONFERENCE ON POSITION SENSITIVE DETECTORS,
12–16 SEPTEMBER 2011,
ABERYSTWYTH, U.K.

Synthesis and scintillation characterization of nanocrystalline Lu_2O_3(Eu) powder for high-resolution X-ray imaging detectors

B.K. Cha,[a,1] **S.-M. Yong,**[b] **S.J. Lee,**[b] **D.K. Kim,**[b] **J.H. Bae,**[b] **G. Cho,**[b] **C.-W. Seo,**[a] **S. Jeon**[a] **and Y. Huh**[a]

[a] *Korea Electrotechnology Research Institute, Ansan 426-170, Republic of Korea*
[b] *Korea Advanced Institute of Science and Technology, Daejeon 305-701, Republic of Korea*

E-mail: Goldrain99@gmail.com

ABSTRACT: Lu_2O_3:Eu(C_{Eu}:5mol%) powder scintillators with nanocrystalline structures were successfully synthesized via a precipitation method and subsequent calcination treatment as a conversion material for X-ray imaging detectors. In this work, a homogeneous precipitation process was carried out using DEA(diethanolamine) as a precipitant to prepare nanocrystalline Eu-doped Gd_2O_3 powders. The microstructures, crystal structure and scintillation properties such as luminescent spectra, decay time and light intensity were measured as a function of calcination temperature in heat-treatment of the synthesized powder. The sample prepared at 1200 °C calcination temperature showed the highest light intensity. And the scintillator showed a strong red emission light at near 611nm under photo- and X-ray luminescence for its potential X-ray imaging detector applications.

KEYWORDS: X-ray detectors; Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators); Scintillators and scintillating fibres and light guides; X-ray radiography and digital radiography (DR)

[1] Corresponding author.

doi:10.1088/1748-0221/7/03/C03048

Contents

2012 JINST 7 C03048

1 Introduction

In the last decade digital X-ray imaging detectors using CCD arrays, CMOS and amorphous silicon flat panel (a-Si:H) in combination with various scintillation screens have been widely used for medical and industrial applications. Traditionally, the promising scintillation materials such as thallium-doped cesium iodide(CsI:Tl) and terbium-doped gadolinium oxysulphide(Gd_2O_2S:Tb) and europium-doped gadolinium oxide(Gd_2O_3:Eu) were used to convert X-rays into visible light. However, new scintillators with high X-ray stopping power, excellent conversion efficiency and better spatial resolution are still researched and developed for low radiation dose and high spatial resolution applications. Among various scintillation materials, europium-doped lutetium oxide (Lu_2O_3:Eu) has high density (ρ=9.44g/cm^3) and high X-ray to light conversion efficiency as well as high effective atomic number [1, 2]. It has been reported that the scintillator with nanocrystalline and submicron-sized powder unlike conventional screen with micro-crystalline phosphor shows somewhat different optical, structural properties in luminescence and image resolution, and so on. In this work, Lu_2O_3:Eu scintillators with nano particles were synthesized by using simple precipitation process, and their scintillation characteristics were investigated as a function of various calcination temperature of the synthesized powder in heat-treatment [3, 4].

2 Materials and methods

A simple precipitation process was carried out using diethylamine(DEA) as a precipitant to prepare the nanocrystalline 5mol% Eu-doped Lu_2O_3 powders. In this procedure, first, the $Lu(NO_3)_3{\cdot}xH_2O$ was dissolved in 50 ml of ethanol with continuous stirring to form a clear homogeneous solution. To the clear solution above, DEA was added drop-wise with vigorous continuous stirring. The solution immediately turned in to thick whitish slurry, which was further stirred in the same vigorous manner and precipitated. A small mound of DI water was added to the precipitate and allowed to stand for a few hours to ensure completed precipitation. After the reaction, the final products were systematically washed with DI water and ethanol by centrifugation. The obtained powder was subsequently dried at 60^{o}C for 12 h and calcined from 600 to 1200^{o}C with different temperatures and consistent 5h time in electrical furnace. And $Eu(NO_3)_3{\cdot}\ 6H_2O$ was dissolved in the

ethanol to form the homogeneous Eu-doped Lu_2O_3 powder. To the above solution, (100-x) mol% of calcined Lu_2O_3 powder was dispersed under sonication for 2 h. And then DEA was added to form a Eu-doped Lu_2O_3 in accordance with above procedures. By the reaction, round-shaped and homogenously dispersed Lu_2O_3(Eu) with several hundred nm particle size was obtained [5].

The microstructure, morphology and the crystal structures of the synthesized Lu_2O_3:Eu were performed by FE-SEM (JEM-2100F HR) and high resolution X-ray diffraction(Ultima IV, RIGAKU) with an analysis range 2θ of $20\sim70^\circ$. Furthermore, a Czerny-Turner spectrometer and intensified charge coupled device camera as well as Nd:YAG laser with 266nm excitation were used for photo-luminescence (PL) and decay time measurement. A relative light output of all fabricated samples was measured using a lens-coupled CCD camera (Andor DV-434) and X-ray source (LISTEM, BRS-2) with 4.3mm spot size and inherent 0.8mm Al filter [4].

3 Results and discussion

The crystalline structure of the fabricated scintillating powders was investigated by XRD analysis. The XRD pattern results of the powers obtained from precipitation method as a function of different calcination temperatures are shown in figure 1. X-ray peaks at the powder of Lu_2O_3:Eu calcinated at 600–1200°C temperature has (211), (222), (400), (440), and (622) and are well consistent with cubic crystal structures of Lu_2O_3(standard JCPDF cards 12-0728). Also, as the calcination temperature increases, the diffraction peak width was reduced and showed more sharp XRD pattern peaks [6]. SEM measurement of the Lu_2O_3:Eu powders as different calcination temperatures ranging from 600 to 1200°C was used to investigate the morphology and particle size of the synthesized powders. Figure 2 shows SEM images of nanocrystalline Lu_2O_3:Eu powders with 5mo% Eu concentration at different calcination temperatures of 600–1200°C and 5h. The morphology of fabricated Lu_2O_3:Eu powders was spherical shape and its particle size was dependent on the calcinations temperature. As the calcination temperature increases, the average diameters of the spherical particles were slightly increased. The Lu_2O_3:Eu powders with 600–1200°C calcination temperature showed the particle sizes with average 100–600nm particle sizes in agreement with the Debye-Scherrer equation using the diffraction peak full-width at half-maximum(FWHM). The Lu_2O_3:Eu powders calcined at high temperature showed the increased particle size due to grain growth of Lu_2O_3:Eu precursor during heat-treatment.

The effect of different calcination temperature on the PL (photo-luminescence) intensities and emission spectra is shown in figure 3. The PL measurements were done at room temperature. It seems that their emission spectra are all similar. However, The PL measurements show that the scintillator's emission intensities according to different calcination temperatures change significantly. The PL intensities of Lu_2O_3:Eu scintillators increased rapidly as the temperature increases until 1200°C. Moreover, the main emission peak of the Lu_2O_3:Eu scintillator with cubic structure was observed at 611nm($^5D_0 \rightarrow {}^7F_2$) wavelength, which correspond to a typical red emission transition of Eu^{3+}. The broadening of emission peaks with decreasing calcination temperature was also observed in the emission spectra. The photoluminescence peak of the nanocrystalline powders was very similar to the emission peak of the microcrystalline powder [7]. After excitation at 266nm, the decay curve of the emitted photons of the Lu_2O_3:Eu scintillator with different calcination temper-

2012 JINST 7 C03048

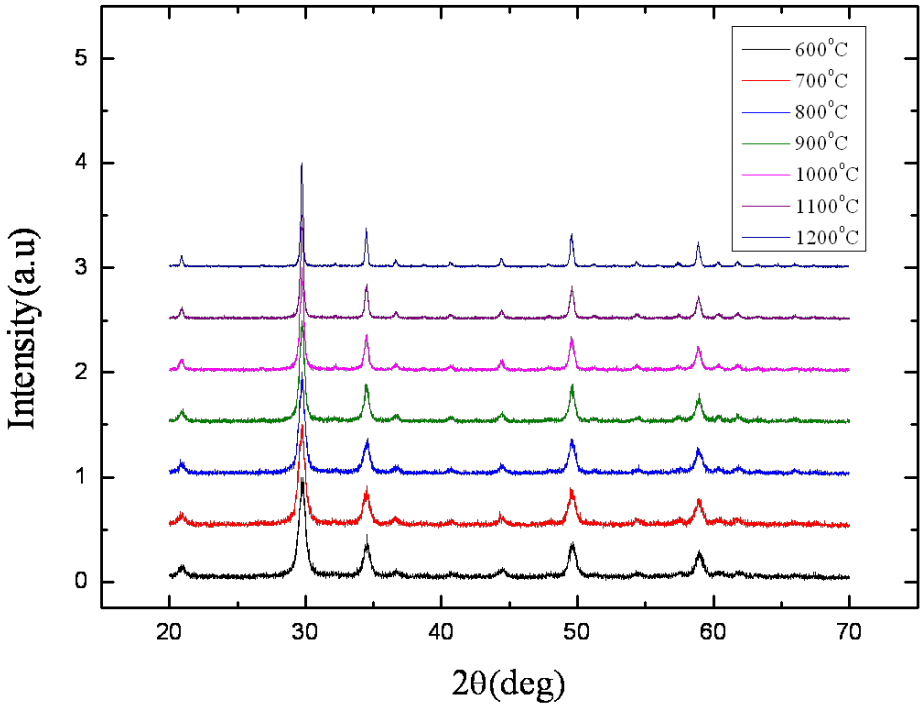

Figure 1. XRD of Lu_2O_3:Eu powders with different calcination temperature.

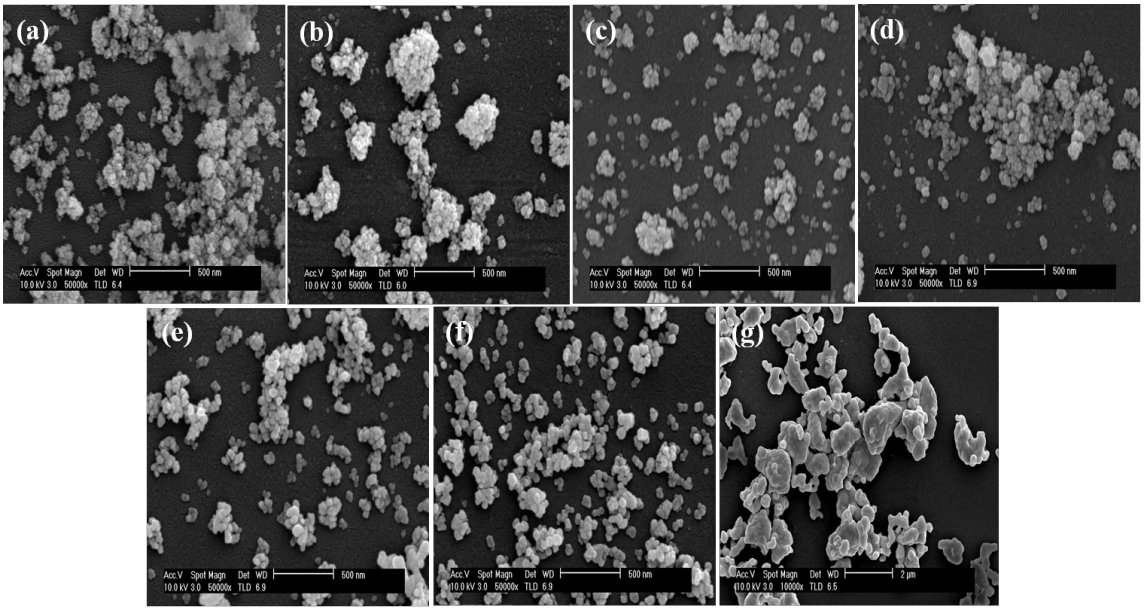

Figure 2. SEM images of Lu_2O_3:Eu powders with different calcination temperature; (a)600 ˚ C, (b)700 ˚ C, (c)800 ˚ C, (d)900 ˚ C, (e)1000 ˚ C, (f)1100 ˚ C, (g)12600 ˚ C.

ature is presented as shown in figure 4 and 5. In this work, the mean decay life times as a function of calcination temperature were observed in the range of 0.4–1.2ms.

The light intensities of the Lu_2O_3:Eu scintillator with different calcination temperature were

2012 JINST 7 C03048

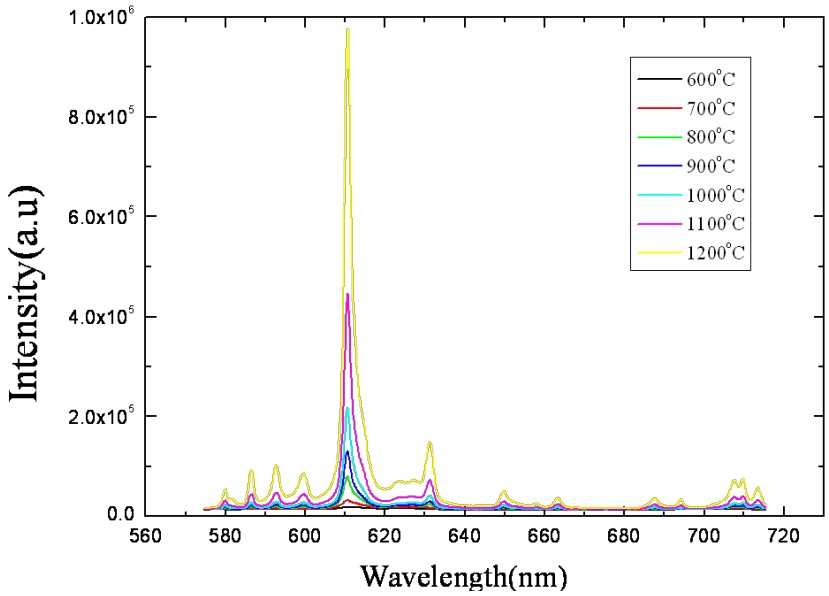

Figure 3. Emission spectra of Lu_2O_3:Eu powders with different calcination temperature.

measured by X-ray excited luminescence under X-ray source exposure with 80kVp and 30mAs beam current. The light output was obtained by measuring the average pixel value over region of interest (ROI) X-ray images. As the calcination temperature of Lu_2O_3:Eu scintillators increases until 1200°C, the light output increased significantly. The dramatic increase of light output between 800 and 1200°C temperature was observed from the results. The highest light intensity was observed for the sample prepared at 1200°C temperature as shown in figure 6.

4 Conclusions and future

Eu-doped Lu_2O_3(Lu_2O_3:Eu) scintillators with nanocrystalline structure were synthesized via a simple precipitation and subsequent calcination process for X-ray imaging detector applications. In this work, the scintillators with average 100 to 600nm particle size were prepared according to different heat-treatment conditions ranging from 600–1200°C calcination temperature. The spherical nanocrystalline particle and crystal structure with cubic phase of Lu_2O_3:Eu powders was discovered at 600–1200°C calcination temperature. When the Lu_2O_3:Eu scintillator were excited by UV light of 266nm, main emission peak of samples with cubic structure was showed at 611nm. The particle size and luminescent intensity of synthesized Lu_2O_3:Eu powder were significantly influenced by calcination temperature. There were no differences in emission peak of the luminescent spectra according to various calcination temperatures. As calcination temperature increases, higher crystalline size and higher scintillation intensity of Lu_2O_3:Eu powder were achieved. The highest light intensity by photo- and X-ray luminescence was showed at 1200°C calcination temperature. And decay life time of the Lu_2O_3:Eu samples showed the 0.4–1.2ms ranges as a function of calcination temperature. In the near future, we will try to fabricate and apply the nanocrystalline Lu_2O_3:Eu scintillation film with optimal synthesis condition for high-sensitive and resolution X-ray imaging detectors.

2012 JINST 7 C03048

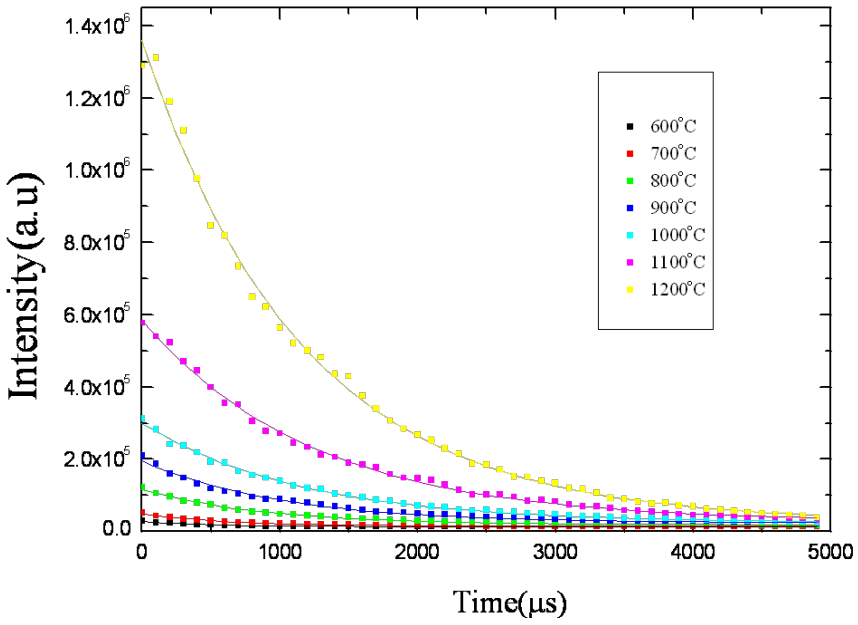

Figure 4. The decay time curves of Lu_2O_3:Eu powders with different calcination temperature at 611nm emission wavelength.

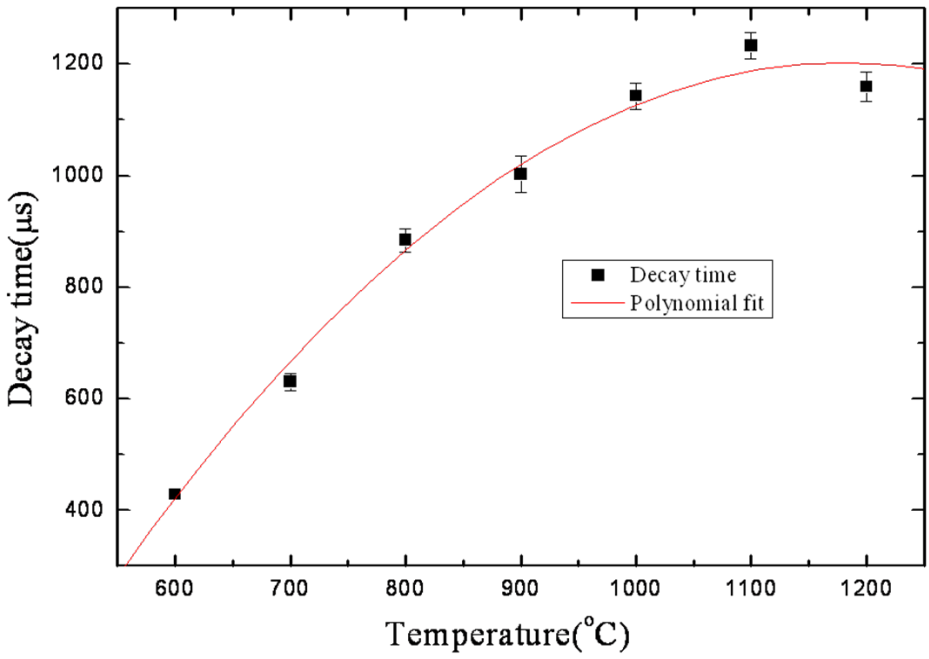

Figure 5. The mean decay time curves of Lu_2O_3:Eu powders as a function of calcination temperature.

Acknowledgments

This research was supported by the cooperative R&D program (B551179-08-04-00) funded by the Korea Research Council for Industrial Science & Technology.

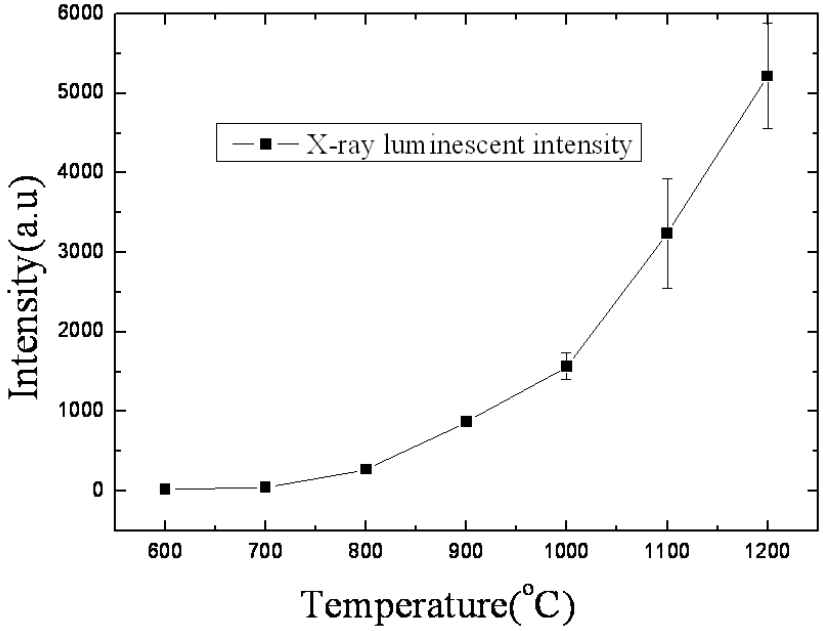

Figure 6. Light output of Lu_2O_3:Eu powders with different calcination temperature.

References

[1] A.R. Cowen, S.M. Kengyelics and A.G. Davies, *Solid-state, flat-panel, digital radiography detectors and their physical imaging characteristics*, *Clin. Radiol.* **63** (2008) 487.

[2] E. Zych, D. Hreniak and W. Strek, *Lu_2O_3:Eu, a new X-ray phosphor*, *Mater. Sci.* **20** (2002) 111.

[3] M. Mupparapu et al., *Development and application of a novel nanophosphor scintillator for a low-dose, high-resolution digital X-ray imaging system*, *Int. Cong. Ser.* **1281** (2005) 1256.

[4] B.K. Cha et al., *Synthesis and scintillation properties of nano Gd_2O_3(Eu) scintillator for high resolution X-ray imaging applications*, *Nucl. Instrum. Meth.* **A 619** (2010) 174.

[5] E. Zych et al., *Homogeneously precipitated Lu_2O_3:Eu nanocrystalline phosphor for X-ray detection*, *Sens. Actuat.* **B 109** (2005) 112.

[6] J. Sokolnicki, *Photoluminescence and structural characteristics of $Lu_2O_3:Eu^{3+}$ nanocrystallites in silica matrix*, *J. Solid State Chem.* **180** (2007) 2400.

[7] S. Cho et al., *Synthesis and characterization of Eu3+ doped Lu_2O_3 nanophosphor using a solution-combustion method*, *J. Sol-Gel. Sci. Technol.* **53** (2009) 171.

2012 JINST 7 C03048