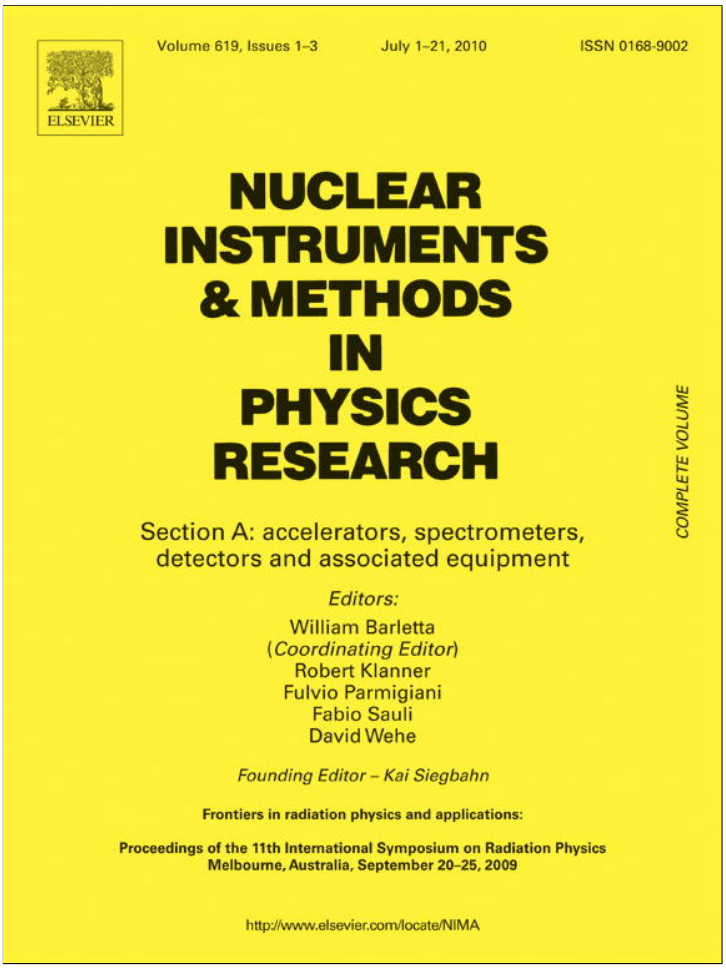

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Synthesis and scintillation properties of nano Gd_2O_3(Eu) scintillator for high resolution X-ray imaging applications

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ABSTRACT

Gd_2O_3:Eu scintillators with nano-crystalline structures were successfully synthesized through a precipitation method and subsequent calcination treatment as a converter for X-ray imaging detectors. In this work, a simple precipitation process was carried out using diethanolamine (DEA) as a precipitant to prepare nano-crystalline Eu-doped Gd_2O_3 powders. Scintillation properties such as luminescent spectra, light intensity and decay time were measured by varying the 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 emitted a strong red light at near 611 nm under photo- and X-ray luminescence for its potential X-ray imaging detector applications.

1. Introduction

In the last decade, indirect detection methods using charge-coupled device (CCD), complementary metal oxide semiconductors (CMOS) or amorphous silicon flat panel with scintillators has been widely used in digital X-ray imaging for medical and industrial applications. The conventional scintillators such as thallium-doped cesium iodide (CsI:Tl), terbium-doped gadolinium oxysulphide (Gd_2O_2S:Tb) and europium-doped gadolinium oxide (Gd_2O_3:Eu) were used for conversion of X-ray into visible light which in turn generates electrical charges in the above electronic imaging devices. However, scintillators with higher light output and better spatial resolution are still under development for application to X-ray microscopy or nano-imaging technology. It has been reported that the scintillator with nano-particle sizes unlike conventional screen with micro-crystalline phosphor shows somewhat different optical, structural properties in luminescence and image resolution, and so on. In this work, Gd_2O_3:Eu scintillators with nano-particles were synthesized by using simple precipitation process, and their scintillation properties were investigated as a function of various calcination temperature of the synthesized powder in heat-treatment [1–3].

2. Experimental procedure

A simple precipitation process was carried out using DEA as a precipitant to prepare the nano-crystalline 5 mol% Eu-doped Gd_2O_3 powders. In this procedure, $Eu(NO_3)_3 \cdot 6H_2O$ and $Gd(NO_3)_3 \cdot 6H_2O$ were dissolved in absolute ethanol with continuous stirring to form a clear homogeneous solution. To the above solution, 0.25 mol DEA was added dropwise with continuous stirring. The solution immediately turns into white thickened slurry, which was stirred vigorously and precipitated at room temperature (RT). A small amount of DI water was added to the precipitate and was allowed to stand for a few hours to ensure complete precipitation and then washed systematically with DI water and ethanol by centrifugation. The bright white powder obtained was dried at 60 °C for 12 h. And subsequent heat treatment was conducted from 600 to 1400 °C for 3 h in the electric furnace [4].

The microstructure, morphology and the existing phase of the synthesized Gd_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 20–90°. Photoluminescence (PL) and decay time were measured by means of a Czerny–Turner spectrometer and intensified charge coupled device camera using a Nd:YAG laser with 266 nm excitation source. A relative light output of samples was measured using a lens-coupled CCD camera (Andor DV-434) and X-ray source (LISTEM, BRS-2) with 4.3 mm spot size and inherent 0.8 mm Al filter [5].

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3. Results and discussion

The XRD pattern results of the powders prepared from precipitation method according to different calcination temperature are shown in Fig. 1. X-ray peaks at the powder of Gd_2O_3:Eu calcinated at 700–1200 °C temperature has (211), (222), (400), (440) and (622) and are consistent with cubic crystal structures of Gd_2O_3 (JCPDF cards 00-012-0797). The phase transformation from the cubic to monoclinic structure was induced at 1300 and 1400 °C temperature and the peaks correspond to crystallographic planes of the monoclinic phase (JCPDF cards 00-043-1015). Also, as the calcination temperature increases, the diffraction peak width was reduced and showed more sharp XRD pattern peaks.

In order to investigate the morphology and particle size of the synthesized powders, the SEM images of the Gd_2O_3:Eu powders as different calcination temperature ranging from 700 to 1400 °C are presented in Fig. 2. As the calcination temperature increase, the average diameters of the spherical particles were significantly increased from nano- to micrometer size. The Gd_2O_3:Eu powders with 700–1100 °C calcination temperature showed the particle sizes with average 20–500 nm particle sizes in agreement with the Debye–Scherrer equation using the diffraction peak full-width at half-maximum (FWHM). The powders synthesized at over 1200 °C temperature showed the particle shapes with average 5 μm sizes due to grain growth of Gd_2O_3:Eu precursor during heat-treatment.

The effect of calcination temperature on the photoluminescence (PL) intensities and emission spectra is displayed in Fig. 3. The PL intensities of samples increased rapidly as the temperature increased until 1200 °C. However, the intensities were decreased above 1300 °C temperature. Moreover, the main emission peak of the Gd_2O_3:Eu scintillator with cubic structure was observed at

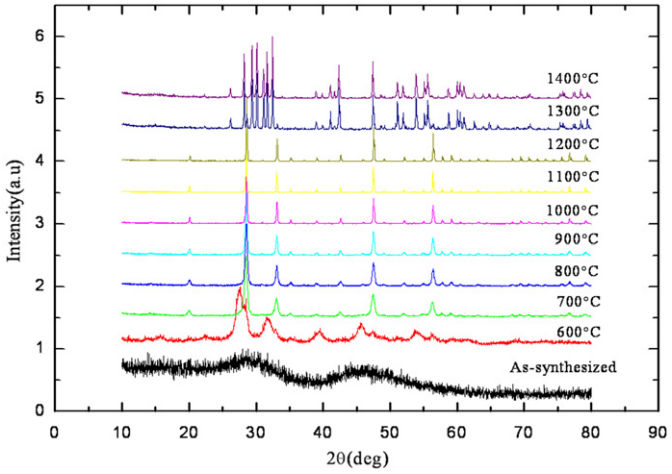

Fig. 1

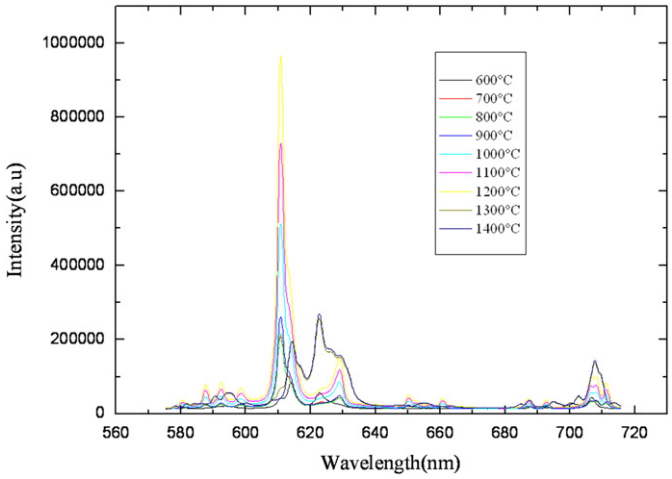

Fig. 3

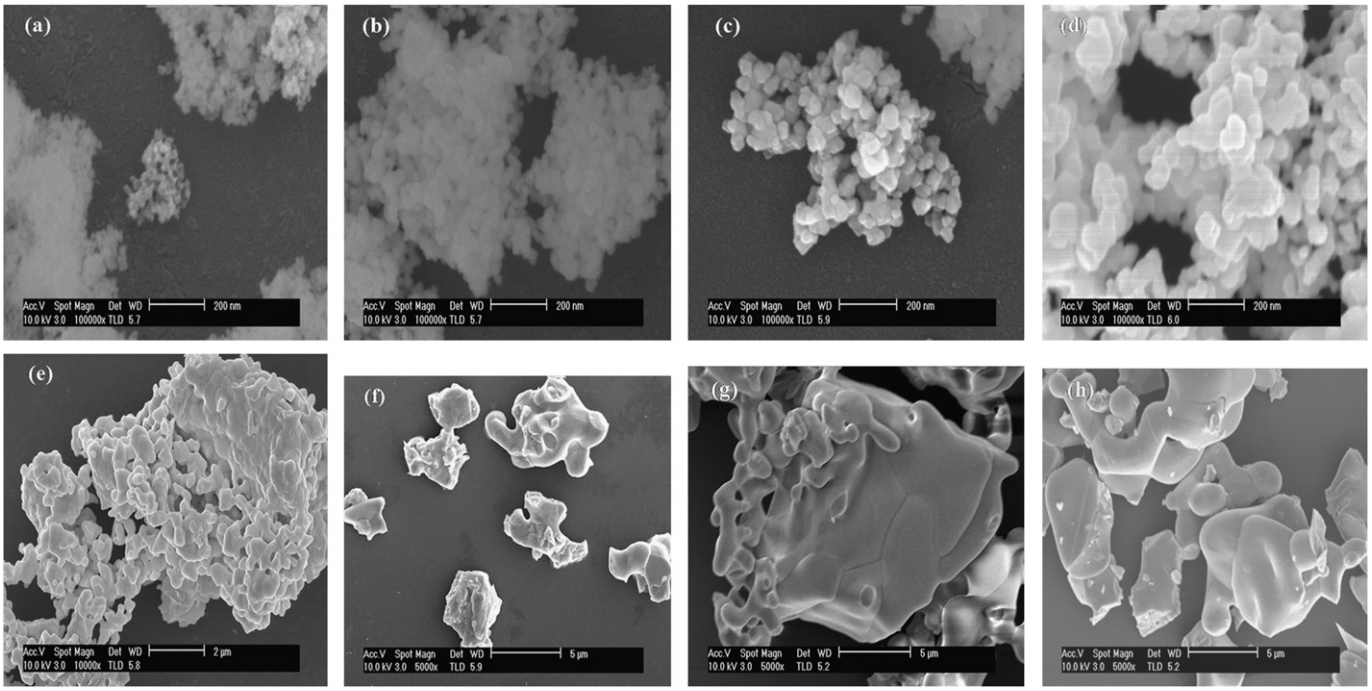

Fig. 2

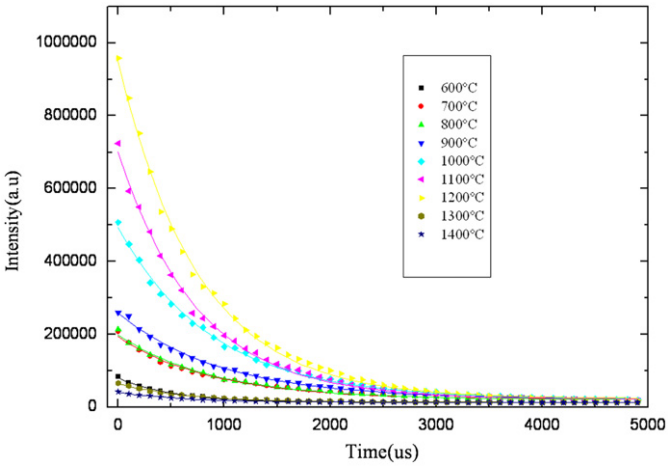

Fig. 4

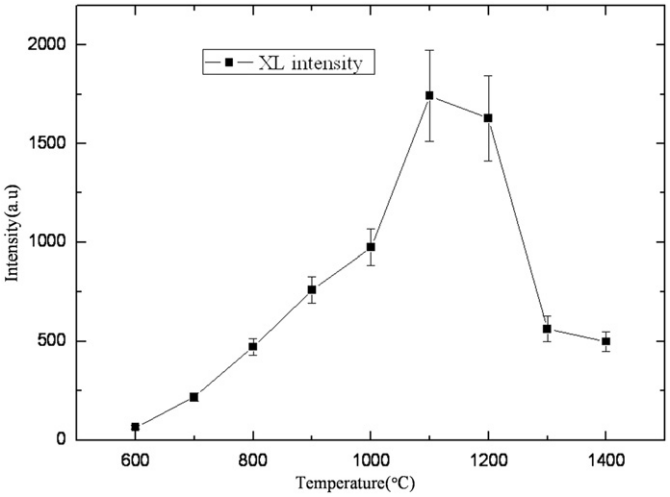

Fig. 5

611 nm ($^5D_0 \rightarrow {}^7F_2$) wavelength, which correspond to a typical red emission transition of Eu^{3+}. The photoluminescence of the powders with the monoclinic phase which are calcinated at 1300 and 1400 °C showed a strong emission peak at 623 nm wavelength.

After excitation at 266 nm, the decay curve of the emitted photons of the Gd_2O_3:Eu scintillator with different calcination temperature is presented as shown in Fig. 4. In our study, the decay times in the range 0.5–1.0 ms were measured with a little change as a function of calcination temperature. The light intensities of the Gd_2O_3:Eu scintillator with different calcination temperature were measured by X-ray excited luminescence under X-ray source exposure with 80 kVp and 30 mAs beam current. The highest light intensity was observed for the sample prepared at 1100 °C temperature as shown in Fig. 5.

4. Conclusion

Eu-doped Gd_2O_3(Gd_2O_3:Eu) scintillators with nano-particles were synthesized using a simple precipitation and subsequent calcination process for X-ray imaging detector applications. In this work, the scintillators with average 20 nm to 5 μm particle size were prepared according to different heat-treatment conditions ranging from 600 to 1400 °C calcination temperature. The morphology, particle sizes and crystal structure of the synthesized Gd_2O_3:Eu powders were observed by SEM and XRD analysis. The phase transformation from cubic to monoclinic structure was discovered at 1300 °C calcination temperature. When the Gd_2O_3:Eu scintillator were excited by UV light of 266 nm, dominant emission peak of samples with cubic structure was showed at 611 nm, on the other hands, the main peak of those with monoclinic structure was observed at 623 nm wavelength. The particle size and luminescence intensity of synthesized Gd_2O_3(Eu) powder were greatly influenced by calcination temperature. As calcination temperature increases, the particle size of Gd_2O_3(Eu) powder was considerably increased. As a result, the luminescent intensity was also increased. However, luminescence intensity of Gd_2O_3(Eu) powder with micronized particles calcinated at high temperature was no longer increased or decreased. The highest light intensity by photo- and X-ray luminescence was showed at 1200 and 1100 °C calcination temperature, respectively. And decay time of the Gd_2O_3:Eu samples showed the 0.5–1 ms ranges as a function of calcination temperature. In the near future, we will try to apply the nano-crystalline Gd_2O_3:Eu scintillator with high light intensity for high resolution X-ray imaging detectors.

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