

# Gamma irradiation induced method in preparation of $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ phosphors: the effect of dose towards luminescent properties

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**Abstract.** A novel gamma irradiation induced synthesis method of  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors was investigated in the presence of cetyltrimethylammonium bromide (CTAB). The effect of irradiation doses (50-150kGy) on structural and morphology analysis as well as luminescence properties were characterized by X-ray diffraction (XRD), field emission scanning microscopy (FESEM) and photoluminescence spectrometer (PL). The results show that gamma radiation is potentially induced formation of  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors from radiation reduction and/or precipitation of insoluble compounds as the hexagonal phase structure was formed without any impurities as proven in XRD pattern. The morphologies were observed that the obtained  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors possess sphere structure with smooth surface at 100 kGy irradiated dose. PL spectroscopy reveals that the strongest red emission peaks is located at 626 nm under 325 nm light excitation, which corresponds to  $^5\text{D}_0 \rightarrow ^7\text{F}_2$  transition of  $\text{Eu}^{3+}$  ions. An optimized dose for excellent luminescent was observed at 100 kGy. The results suggested that the  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors may have a beneficial approach in field of imaging device or media.

## 1. Introduction

Trivalent europium activated gadolinium oxysulfide ( $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ ) phosphors is an important luminescent material especially for imaging display either in medical or industrial applications. The material featured high density and refractive index thus perform high light absorption and energy transfer efficiency with a broader excitation band and shows red emission. Radiation-induced processes have some advantages over conventional chemical methods:

- Temperature independent
- High purity of yield materials
- Narrow size distribution of particles
- Simple method

In this work, we have investigated the potential of gamma irradiation method to synthesize  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors as well as the effect on luminescent properties.

## 2. Experimental details

Gadolinium (III) nitrate hexahydrate ( $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ , 99.9%), europium (III) nitrate ( $\text{Eu}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ , 99.9%) and cetyltrimethylammonium bromide (CTAB,  $\geq 98\%$ ) were purchased from Sigma-Aldrich. Ammonium sulfate ( $(\text{NH}_4)_2\text{SO}_4$ ) was purchased from Merck. All reagents were



analytical grade and used without further purification. Deionized water (18 $\Omega$ ) was used as solvent throughout the experiment.

In this study, gadolinium nitrate,  $\text{Gd}(\text{NO}_3)_3$  0.5 M,  $\text{Eu}(\text{NO}_3)_3$  0.05 M and ammonium sulfate,  $(\text{NH}_4)_2\text{SO}_4$  were weighed in suitable stoichiometric ratio and dissolved in deionized water as described detail in our previous work [1]. 1% mol of CTAB was added into the solution and stirred with a magnetic stirrer continuously for 2 hours before being portioned into four samples and irradiated in  $^{60}\text{Co}$  gamma cell (1.45 kGy/h) with irradiation doses at 50kGy, 100kGy and 150kGy. The precipitate samples obtained were centrifuged and washed with ethanol and deionized water for several times. Afterwards, the solid precipitates were dried overnight in air at ambient temperature and hydrogenation treatment was carried out towards dried precipitates at 900°C for 2 hours.

### 2.1. Characterization

The gamma source of  $^{60}\text{Co}$  with a dose rate of 1.45 kGy/h model MDS NORDION / Gamma Cell 220 Excel was used under atmospheric pressure and room temperature. The X-ray diffraction (XRD) measurements of  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors samples were carried out in the reflection mode with a Bruker X-ray diffractometer that operated at 40 kV voltages and a current of 40 mA with  $\text{Cu K}\alpha$  radiation ( $\lambda=0.15406$  nm). The morphology and size of the samples were inspected by the field emission scanning electron microscopy (FESEM-Carl Zeiss, Supra 35VP) imaging. Meanwhile, the photoluminescence spectra of the samples were gathered over a range between 300 to 800 nm of wavelength via FLSP920 Edinburgh spectrometer.

## 3. Results and Discussion

The X-ray diffraction pattern of  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors at different irradiation doses are shown in FIGURE 1. All samples show same diffraction patterns including none-irradiated sample (control) and these well-defined diffraction peaks can be indexed to pure hexagonal  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phase according to JCPDS No: 00-026-1422 without any impurity peaks.

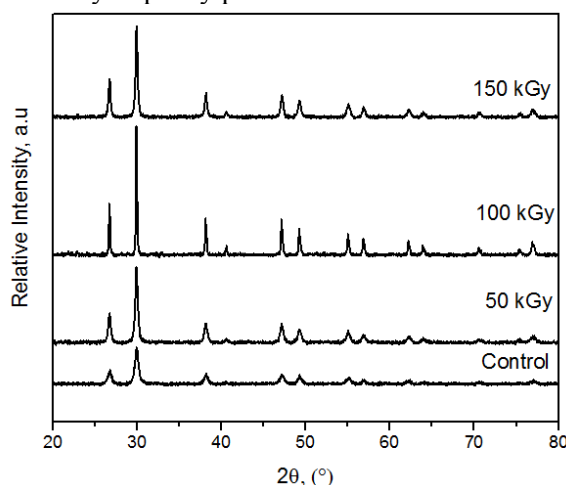


FIGURE. 1: XRD patterns of  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors of none-irradiated sample (control), 50 kGy, 100 kGy and 150 kGy irradiation dose.

The FESEM images of the produced  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors at different gamma irradiation doses were shown in FIGURE 2. In FIGURE 2(a), the none-irradiated samples exhibit agglomerated and rough surface of the particles. As the irradiation dose increases, the particle morphologies transformed into uniform spherical shape and the agglomeration also reduced. Therefore, the gamma irradiation could also produce defined structure as well as high homogeneity. The sample with 100 kGy

irradiation dose shows high homogeneity and smooth surface. After 150 kGy irradiation dose, the spherical shape becomes ruptured and slightly porous due to molecular damage of CTAB thus unable to protect the particle surface. In this case, CTAB played an important role in controlling the morphologies and sizes of any nanomaterials [2]. As expected, the low concentration of CTAB used in this work tends to formed sphere structures as the surfactant associates into micelles and controlled particles surface thus slow down the nucleation process which also avoided from the aggregation process.

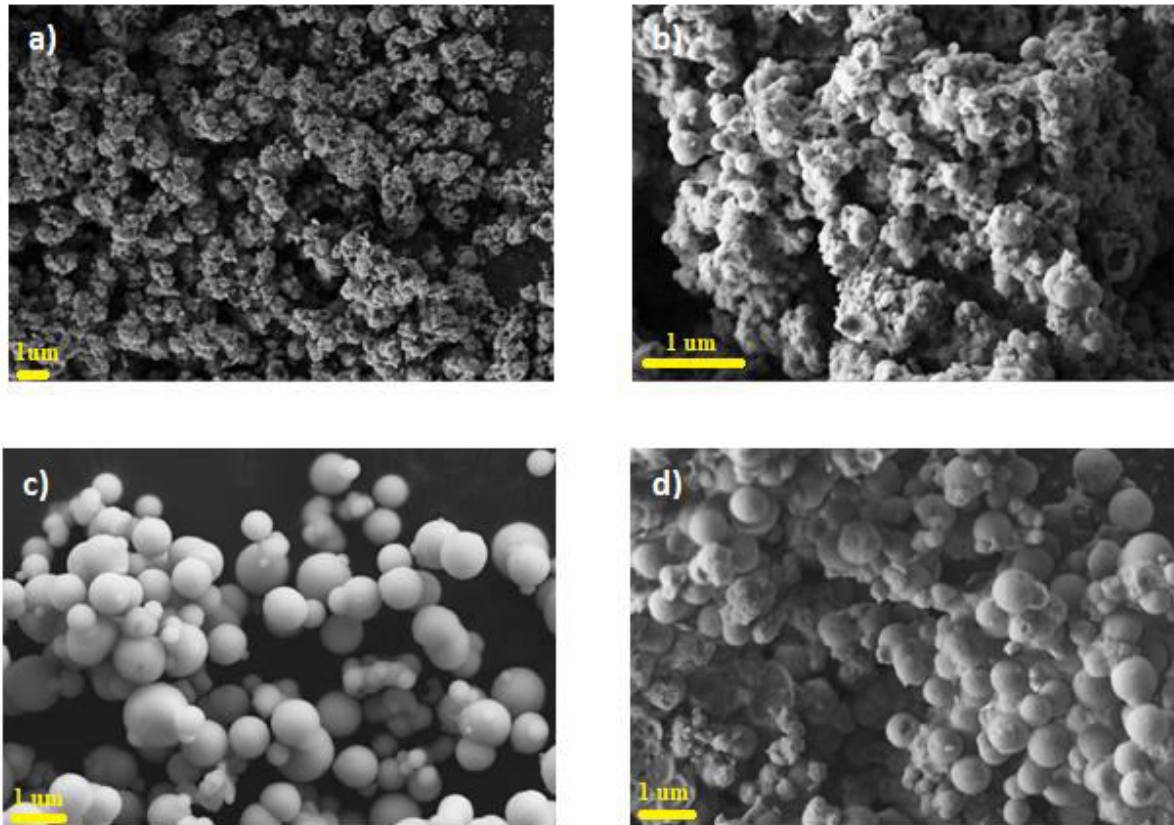


FIGURE 2.: FESEM images of  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors of non-irradiated sample/control (a) and by different irradiation doses b) 50 kGy, c) 100 kGy and d) 150 kGy.

FIGURE 3 shows the emission spectra of the  $\text{Gd}_2\text{O}_2\text{S}:\text{Eu}^{3+}$  phosphors at different gamma irradiation doses. Upon the excitation at 325 nm, the strongest red emission peaks which splits into two peaks at 616 nm and 626 nm arises from the forced electric-dipole  $^5\text{D}_0 \rightarrow ^7\text{F}_2$  transition of the  $\text{Eu}^{3+}$  ions. In this process, trivalent  $\text{Gd}^{3+}$  ions, as sensitizer, absorb ultraviolet excitation light and subsequently transfer energy to the neighboring  $\text{Eu}^{3+}$  ions act as activator, resulting in the overall red emission of  $\text{Eu}^{3+}$  ions [3]. An optimized dose that demonstrated excellent luminescent was observed at 100 kGy. It was found that the smooth surface of particles showed higher photoluminescence intensity [4]. The slight decrease in luminescence intensity after reaching a certain crystallite size was caused by the large size of the crystallite, which tends to scatter light in particle excitation, leading to a reduction in intensity in luminescence measurements (FIGURE 3(b) 7).

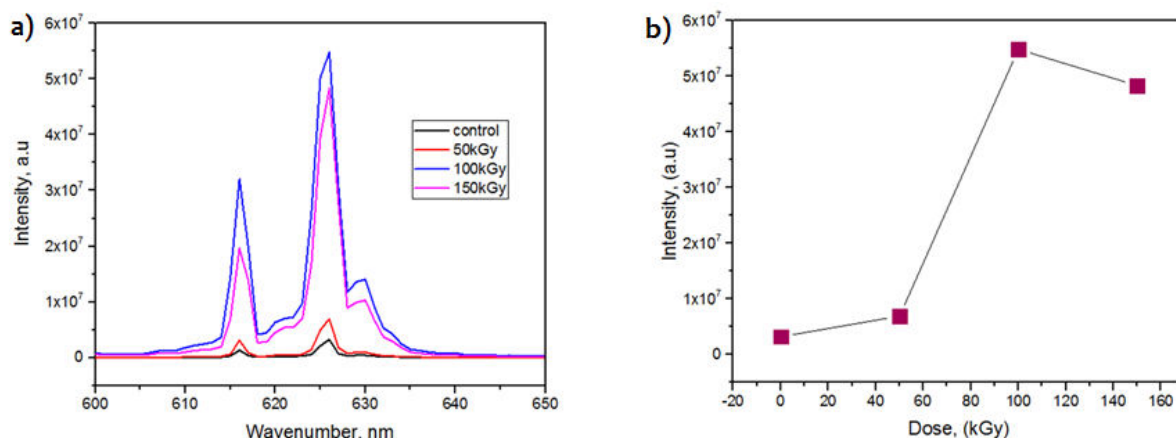


FIGURE. 3: (a) Photoluminescence emission spectra of Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphors at different radiation doses and control (none irradiated sample) and (b) relation between irradiation dose and luminescent intensity.

#### 4. Conclusion

In summary, Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphors have been successfully synthesized by gamma irradiation induced method combined with hydrogenation process. The CTAB played important roles in the formation of the spherical morphology as well as in producing smooth surface of particles. It was found that the optimized dose for excellent luminescent observed at 100 kGy and the strongest red emission peaks is located at 626 nm under 325nm light excitation, which corresponds to <sup>5</sup>D<sub>0</sub>→<sup>7</sup>F<sub>2</sub> transition of Eu<sup>3+</sup> ions. The results obtained suggest that the Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphors may have a beneficial approach in field of imaging device or media.

#### 5. Acknowledgment

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