

# Aerosol synthesis of phosphor particles based on $\text{Eu}^{3+}$ activated gadolinium oxide matrices



Metadata, citation and similar papers at core.ac.uk

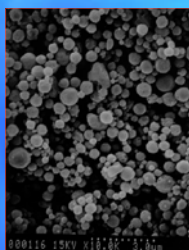
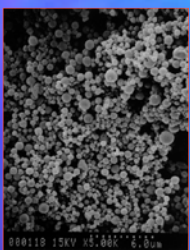
Provided by Serbian Academy of Science and Arts Digital Archive (DAIS)

Institute of Technical Sciences of  
Serbian Academy of Sciences and Arts  
11 000 Belgrade, K.Mihailova 35/IV  
Yugoslavia

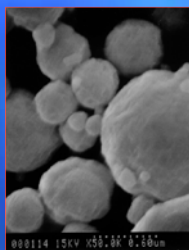
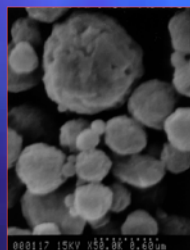
Japan Fine Ceramic Center  
2-4-1 Mutsuno, Atsuta-ku  
Nagoya, 456  
Japan

## ABSTRACT

The designs of advanced phosphor materials effort emphasize the ultrafine powder synthesis control ability. Particularly, when spherical, aggregate-free, uniformly sized particles are considered, luminance and resolution give an improved behavior on their application abilities compared with conventional coarse grained phosphors. Aerosol synthesis as a dispersion phase powder processing method was applied to synthesize the rare earth system  $\text{Eu}^{3+}$  activated gadolinium oxide matrices, commonly used as red phosphors in cathodoluminescence. The process involves aerosol formation ultrasonically (aerosol generator operating at 1.7 MHz) from the precursor salt solutions and control over the aerosol decomposition in a high-temperature tubular flow reactor at the temperatures up to 1173K. Consequently, spherical, solid, agglomerate-free, submicronic particles with the mean particle size below 900nm are obtained. The particle morphology, phase and chemical structure are revealed in accordance to various analysis methods (XRD, DTA, SEM-EMAX) and discussed in terms of precursor chemistry, process parameters and luminescent properties.

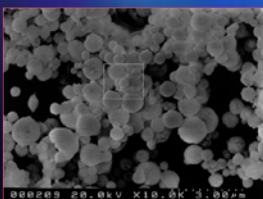


$\text{Gd}_2\text{O}_3$ :Eu as-prepared particles  
gas flow rate: 120 l/h  
residence time: 25.5 s

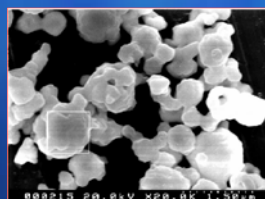


XRD of the as-synthesized powders: cubic  $\text{Gd}_2\text{O}_3$  phase (space group  $\text{Ia}_3$ , file card 43-1014) and a phase that is structurally similar with cubic  $\text{Gd}_2\text{TeO}_7$  (space group  $\text{Fm}\bar{3}\text{m}$ , file card 37-1400).

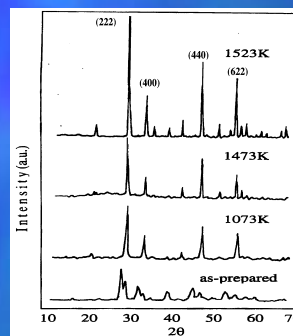
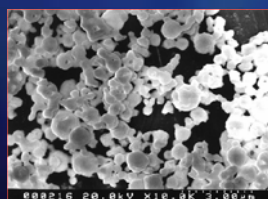
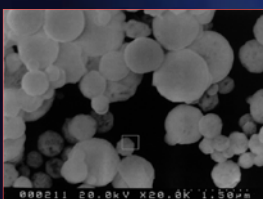
Red emission for the as-prepared powders, two dominant peaks at 615 and 624nm. The sharp emission band at 612nm is evident in all investigated samples annealed in the range from 1073-1473K.



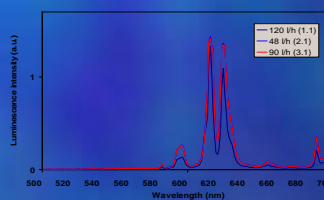
$\text{Gd}_2\text{O}_3$ :Eu particles  
gas flow rate: 120 l/h  
annealed at 1073K/10h



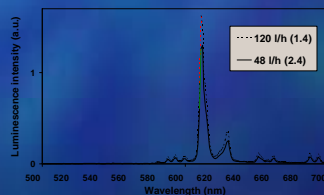
$\text{Gd}_2\text{O}_3$ :Eu particles  
gas flow rate: 90 l/h  
annealed at 1473K/8h



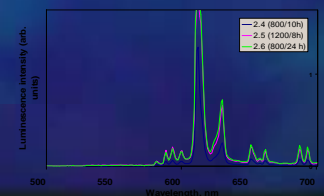
## Influence of the gas flow rate



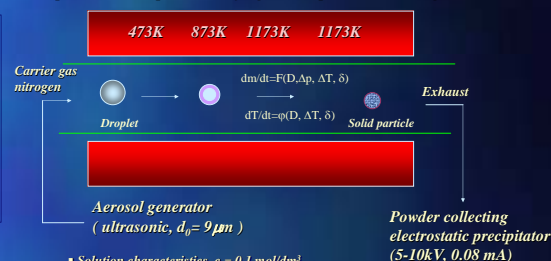
## Influence of the flow rate (annealing temperature 800C)



## Influence of the annealing temperature



## Evaporation- Precipitation- Drying- Decomposition-Sintering



• Solution characteristics,  $c = 0.1 \text{ mol/dm}^3$   
 $\text{Gd}(\text{NO}_3)_3 \cdot n\text{H}_2\text{O}$  ( $n=5.92$ ) and  
 $\text{Eu}(\text{NO}_3)_3 \cdot n\text{H}_2\text{O}$  ( $n=5.42$ )

Phosphor particles  
 $\text{Gd}_2\text{O}_3$ :Eu

Table I. Synthesis conditions and characteristics of  $\text{Gd}_2\text{O}_3$ :Eu phosphor particles

Sample	Flow rate (l/h)	Particle size (nm) / Specific surface area ( $10^4 \text{ m}^2/\text{m}^3$ )	Annealing temperature (K) / time (h)	Crystallite size (nm) / lattice microstrains (Scherrer eq <sup>12</sup> ; Voight function modeling <sup>13</sup> )
F1.1	48 / 63.1	770 / 8.74	As-prepared	17.10 / 102.6
F1.2			1073 / 10	25.66 / 45.57
F1.3			1073 / 24	68.46 / 6.4
F1.4			1473 / 8	136.85 / 1.6
F2.1	90 / 34.3	800 / 8.18	As-prepared	-
F2.2			1073 / 10	82.12 / 4.45
F2.3			1473 / 8	136.87 / 1.6
F3.1	120 / 25.5	890 / 7.5	As-prepared	14.66 / 139.6
F3.2			1073 / 10	-
F3.3			1073 / 24	17.18 / 102.5
F3.4			1473 / 8	136.81 / 1.60

## CONCLUSIONS

Aerosol synthesis was used for the preparation of red phosphor particles based on cubic  $\text{Gd}_2\text{O}_3$ :Eu. As-prepared particles are spherical, nonagglomerated, submicronic (<900 nm), comprised of nanocrystallites (~6nm). In order to control the particles crystal structure and to establish the conditions for stabilization of the low-temperature gadolinia cubic phase, the process parameters such as temperature distribution, gas flow rate and annealing temperatures were adopted. It was shown that the phase content influences luminescence causing the appearance of two dominant peaks at 615 and 624 nm for as-prepared particles when a two-phase particle structure is established. However, the sharp emission band at 612 nm is associated with single cubic gadolinium oxide. Particle morphology affects luminescence properties implying that particle aggregation and/or sintering caused the decrease of luminescence intensity.

## REFERENCES

- Phase diagrams. Materials Science and Technology, ed. A.M.Alper, vol. IV, Academic Press, New York, San Francisco, London (1976).
- P. Beck and P.H.Holloway. *Materials Science and Engineering*, R21[4] 171-219 (1989).
- H.Tsujiki, K.Soga, H.Inoue and A.Makishima. *Journal of the American Ceramic Society*, 81 [5] 1197-202 (1998).
- S.Ekambaram and K.C.Pati. *Journal of Alloys and Compounds*, 248 7-12 (1997).
- G.L. Messing, S.C.Zhang and G.V.Sayantha. *Journal of the American Ceramic Society*, 76 [11] 2707-805 (1993).
- A. Gero, I.Kokles, T.Pfingst and Y.Song. *Aerosol Science and Technology*, 19 411-52 (1993).
- O.Milosevic, M.Mirkovic and D.Uskokovic. *Journal of the American Ceramic Society Communications*, 76 [6] 720- (1996)
- O. Milosevic, pp. 103-111 in *Advanced Science and Technology of Sintering*, Edited by B. Stojanovic, M.V.Nikolic and V. Skovokhod, Kluwer Academic/Plenum Publishers, New York, 1999.
- Y.C.Kang, S.B.Park, I. W.Lenggoro and K.Okuyama. *Journal of Electrochemical Society*, 146 [7] 2744-747 (1999).
- Y.C.Kang, S.B.Park, I. W.Lenggoro and K.Okuyama. *Journal of Physics and Chemistry of Solids*, 60 379-384 (1999).
- R.Marek, T.Fukui, S.Ohara, H.Yoshida, M.Nishimura, T.Inagaki and K.Miura. *Journal of Materials Science*, 35 1-8 (2000)
- H. P.Klug and L.E.Alexander. John Wiley & Sons, New York, London, Sydney, Toronto, 1974
- D.Balzar and H.Lebetter. *Journal of Applied Crystallography*, 26, 97-103 (1993)