Particle Size Effect on the Packaging Performance of YAG:Ce Phosphors in White LEDs

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The synthesis of cerium-doped yttrium aluminum garnet (YAG:Ce) phosphor of different sizes with uniform size distribution was carried out using solid-state reaction followed by grinding and sieving method. The effect of particle size distribution of YAG:Ce phosphors on the photoluminescence (PL) properties was investigated. The results demonstrate that the uniform size distribution and particle size affects the packaging performance in white light emitting diode (LED) applications. The YAG:Ce phosphors with different particle sizes were packaged in white LEDs using different amounts of each phosphors in order to get similar efficiency as that of commercially available YAG:Ce phosphors. It was observed that minimum amount of phosphor material is required for smaller particle size for getting the similar efficiency as that exhibited by commercially available YAG:Ce phosphors. The results are particularly interesting in view of reducing the cost of current LEDs by lowering the amount of phosphors without compromising the efficiencies of final LED package. A systematic study of YAG:Ce phosphors on the packing performance in white LEDs is reported.

Introduction

White light emitting devices (LEDs) have been considered as a replacement for conventional lighting systems because of their excellent properties such as high energy efficiency, long lifetime, low-power consumption

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and environmental protection.^{1–3} Generally, the white light was produced by a combination of blue LED with yellow luminescence from $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ yttrium aluminum garnet (YAG:Ce) phosphor materials.⁴ The yellow emission is sufficient enough to complement the residual blue light that escapes through the phosphor to produce white light. Thus, YAG:Ce has been the most suitable phosphor that can be utilized in the white LED market. The performance of phosphors in lighting applications is usually related to emission efficiency, quenching of luminescent centers and internal scattering. There have been numerous studies to improve YAG/Ce efficiency with focus on improving the YAG host quality, reducing defect quenching centers and reducing concentration quenching.^{5–7} The size reduction of phosphors was found to be the most effective way to improve the LED efficiency. The smaller particles possess a higher surface to volume ratio, which enhances the efficiency of absorption and emission.

It is desirable to have a fine particle size for high-resolution, optimum chromaticity, and brightness in white LEDs. The effect of particle size of YAG:Ce phosphors on the luminescent properties has been extensively studied.^{8,9} However, the effect of particle size of phosphors on the packaging performance of white LEDs and the cost reduction have not been studied in the literature. The cost reduction by improvement in the manufacturing process as well as in the luminescent properties is extremely valuable for applications in white LEDs. In this paper, we have synthesized four different sizes of YAG:Ce phosphors with narrow size distribution and studied the effect of particle size on the final packaging performance in white LEDs for the first time. One of the synthesized YAG:Ce samples (PS-1) resembles the particle size of the commercially available YAG:Ce phosphors. The characterization of YAG:Ce phosphors were carried out by X-ray diffraction, scanning electron microscope (SEM), and PL measurements. A systematic study of YAG phosphors on the packaging performance in white LEDs is reported.

Experimental Procedures

Synthesis of YAG:Ce phosphors were carried out by conventional solid-state reaction in which the reactants such as Y_2O_3 , Al_2O_3 , and CeO_2 were mixed in a stoichiometric amounts in order to get nominal composition of $(\text{Y}_{2.95}\text{Ce}_{0.05})\text{Al}_5\text{O}_{12}$ at around 1500°C for 2 h in

reducing atmosphere of 5% H_2 in 95% N_2 . The thoroughly grinded samples were then passed through different sieves in order to separate different particle sizes of YAG:Ce phosphors and obtain uniform size distribution. In this way, four different samples were collected with different particle sizes by repetitive sieving. The four YAG:Ce samples with decreasing particle sizes were designated as PS-1, PS-2, PS-3, and PS-4, respectively. The phase purity was analyzed by X-ray powder diffraction (XRD) measurements using an X'Pert PRO advanced automatic diffractometer (X'Pert PRO, Almelo, The Netherlands) with $\text{CuK}\alpha$ radiation operating at 45 kV and 40 mA. The SEM measurements were carried out by using Hitachi S2400 electron microscope (Hitachi, Tokyo, Japan). The particle size distribution was analyzed using Fritsch particle sizer “analysette 22” (Fritsch, Idar-Oberstein, Germany). The UV photoluminescence (PL) and photoluminescent excitation (PLE) spectra were collected at room temperature (RT) using a FluoroMax-3 and FluoroMax-P (Jobin Yvon, Edison, NJ) in the range of 300–500 and 470–650 nm, respectively.

Results and Discussions

The powder XRD measurements were carried out on all the YAG samples and as a representative, the XRD pattern of PS-1 along with the standard pattern of YAG is shown in Fig. 1. The observed peaks are in well agreement with the standard pattern with a cubic unit cell (space group: $\text{Ia}\bar{3}\text{d}$) from ICSD file 882048 and lattice parameters of $\sim 12 \text{ \AA}$, which indicate the formation of single-phase compositions. SEM measurements

Fig. 1. X-ray diffraction patterns of PS-1 along with the standard pattern of yttrium aluminum garnet phosphor.

Fig. 2. Particle size distribution of (a) PS-1, (b) PS-2, (c) PS-3, and (d) PS-4 yttrium aluminum garnet (YAG:Ce) phosphors. Inset shows scanning electron microscopic images of (a) PS-1 and (d) PS-4 YAG:Ce phosphors.

were carried out in order to observe the morphology of YAG samples. The particle size distribution of the YAG samples is shown in Fig. 2 and inset shows the SEM of PS1 and PS4 samples, which suggest different morphologies for YAG samples. The particle sizes with D10%, D50%, and D90% size distribution are shown in Table I. It was observed that the particle morphology is almost identical for all the samples with variation in particle sizes. The particle size of PS-1 was found to be 22.0 μm for D90% size distribution, which is similar to the commercially available YAG sample. The other samples show the particle sizes of 10.5, 9.3, and 4.4 μm for PS-2, PS-3, and PS-4, respectively.

Table I. The Particle Sizes Distribution of D10, D50, and D90%. Wt % of Yttrium Aluminum Garnet (YAG:Ce) Mean % of YAG Phosphor in Silicone

No.	Sample	P/N (D10%) (μm)	P/N (D50%) (μm)	P/N (D90%) (μm)	Wt % of YAG:Ce
1	PS-1	8.3	15.6	22.0	5.7
2	PS-2	—	7.2	10.5	4.5
3	PS-3	—	4.1	9.3	4.4
4	PS-4	—	2.2	4.4	3.7

Fig. 3. Photoluminescent excitation spectra of (a) PS-1, (b) PS-2, (c) PS-3, and (d) PS-4 yttrium aluminum garnet phosphors.

Figure 3 shows the photoluminescence excitation (PLE) of YAG:Ce phosphors with different particle sizes. The excitation spectra between 300 and 500 nm show two excitation peaks, which are similar to previous reports on YAG:Ce system. The excitation peaks were observed at 340 and 457 nm, which can be attributed to the Ce^{3+} transitions from $4f$ ground state to the $5d$ field splitting excited states. The broad excitation peaks suggest that YAG phosphors are similar with commercially available YAG phosphors for excitation efficiency and can be excited by a blue chip in the range of 450–470 nm. Figure 4 shows the emission spectra of YAG:Ce

Fig. 4. Photoluminescence spectra of (a) PS-1, (b) PS-2, (c) PS-3, and (d) PS-4 yttrium aluminum garnet phosphors at 460 nm excitation.

Fig. 5. I_v comparison (%) of white package LEDs of (a) PS-1, (b) PS-2, (c) PS-3, and (d) PS-4 yttrium aluminum garnet phosphors.

phosphors at 460 nm excitation. It was observed that the 532 nm emission peak was slightly blue-shifted as compared with bulk counterpart. This blue-shift was associated with the size of the YAG phosphors. For phosphors with smaller sizes, the lattice parameters were usually smaller than those of the bulk materials because of the huge surface stress. For a smaller lattice parameter consisting of a smaller atomic spacing, the ligands field was usually stronger and hence the blue shift was often observable for field-sensitive emission centers.

In order to understand the effect of particle sizes on the final packaging performance of YAG:Ce phosphors

Fig. 6. Plots of phosphor emission, excitation efficiency after packaging and amount of phosphors for packaging (wt %) against the particle sizes of (a) PS-1, (b) PS-2, (c) PS-3, and (d) PS-4 YAG:Ce phosphors.

in white LEDs, all the phosphor samples were mixed with silicone and uniformly applied over InGN chip. The different amounts of PS-1, PS-2, PS-3, and PS-4 were used in order to get the similar efficiency as that of commercially available YAG:Ce phosphor in white LEDs. The comparison of I_v (%) of white LEDs is shown in Fig. 5. It was observed that all the samples showed similar efficiencies in comparison with commercially available YAG:Ce phosphors. The different amounts of PS-1, PS-2, PS-3, and PS-4 phosphors are plotted against the particle sizes of the phosphors as shown in Fig. 6. Interestingly, the amount of phosphors required to get similar I_v efficiency depends on the particle sizes of the samples. The PS-4 sample required the lowest amount, which is 3.7 wt% as against 5.7 wt% for PS-1 sample. These results suggest that the reduction in particle size will require minimum amount of phosphor samples in comparison with bulk YAG:Ce phosphor without loss of efficiency of white LEDs. Although the emission efficiencies exhibited by smaller size particles are lower (as in Fig. 6), their amount required for packaging in white LEDs was found to be less. For white LED applications, it was observed that YAG:Ce phosphors with smaller particle size was better than fine-powder phosphors with a larger particle size because smaller particle size could reduce internal scattering when they were coated onto a bare LED surface. In brief, the studies on the effect of particle size on the packaging performance to investigate the amounts of phosphors required will be helpful in lowering the cost of white LEDs. The YAG:Ce phosphors with smaller particle size will be cost effective in terms of the reduction in the amount of phosphors utilized in white LEDs.

Conclusions

In summary, YAG:Ce phosphors exhibiting different sizes having uniform distribution were prepared by

solid-state reaction followed by grinding and sieving method. The effect of particle size distribution of YAG phosphors on the PL properties was investigated, which showed that the uniform size distribution and particle size affects the packaging performance in white LED applications. Interestingly, the amount of phosphors required to get similar I_v efficiency showed dependence on the particle sizes of the samples. The PS-4 sample having smaller particles required the lowest amount, which is 3.7 wt% without compromising the efficiency of white LEDs. The YAG:Ce phosphors with smaller particle size will be cost effective in terms of the reduction in the amount of phosphors utilized in white LEDs.

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