

## Continuous Nanocomposite Coatings on a Phosphor for the Enhancement of the Long-term Stability

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### Introduction

$Y_2O_3:S:Eu$  is a phosphor material emitting red light due to the presence of  $Eu^{3+}$  activator. This phosphor, with its sharp emission line for good calorimetric definition and high luminescence efficiency, is extensively used in the phosphor screen of display devices and X-ray detectors of digital mammography. The physical mechanism of phosphor excitation in the field emission display (FED) and conventional cathode ray tube (CRT) displays is the same. However, FEDs operate at much lower voltages, which makes it necessary to use higher current densities in order to maintain the same output luminance. Also, at low acceleration voltages, the electron penetration depth is small compared to the phosphor particle size, and therefore, phosphors exhibit low efficiency due to the loss processes associated with the surface. To suppress this surface recombination and give more long-term service, the surface coating of  $Y_2O_3:S:Eu$  is an essential technique. The surface coatings can also reduce phosphor surface degradation, such as oxidation, associated with display fabrication processes. Silica is a good coating material because it consists of light elements and has low density, which exhibits minimal losses in electron penetration.

In this study,  $Y_2O_3:S:Eu$ , a red phosphor, coated with silica nanoparticles or nanocomposites composed of silica nanoparticles and polymeric materials such as PMMA and PVP was prepared via sol-gel process. The PL intensity and long-term stability of phosphors coated with silica nanoparticles or nanocomposites were investigated.

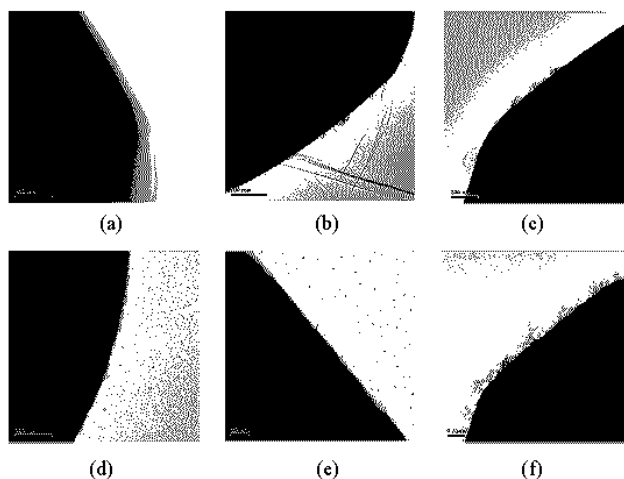
### Materials and Experimental

Tetraethylorthosilicate (TEOS) was used as a precursor of silica. Absolute methanol and reagent grade ammonium hydroxide were used as a solvent and a base catalyst, respectively. 1-vinyl-2-pyrrolinone (VP) and methyl methacrylate (MMA) were used as monomers for the synthesis of PVP and PMMA, respectively. [2,2'-Azobis(2-methylpropionitrile)] (AIBN) was used as an initiator.  $Y_2O_3:S:Eu$  red phosphor supplied by Samsung SDI, Korea was used for this study.

Phosphor coating with silica nanoparticles or nanocomposites was performed via sol-gel process. Various factors affected on particles size and morphology of nanocomposites in this process. Among them, the concentration of the precursor and pH of the solution were two main factors. Samples were prepared from four different methods coded P1, P2, P3, and P4. The details of each described below. P1: Silica nanoparticles were prepared from conventional sol-gel process and then surface coating of phosphors was performed by using a dip-coating method. P2: This method followed the same procedure as P1 except that nanocomposites containing silica nanoparticles and polymer prepared by sol-gel process were used as coating materials. P3: In this method, phosphors were dispersed in a solution containing silica precursor, i.e., TEOS and then polymerization was performed to coat onto the phosphors surface. P4: This method followed the same procedure as P3 except that a solution containing TEOS and organic monomer were used in preparing coating materials. For example, in preparing silica coated  $Y_2O_3:S:Eu$  by P3 method, a solution containing  $Y_2O_3:S:Eu$  5g, TEOS 1.165g,  $NH_4OH$  0.08 g,  $H_2O$  7.2g, and methanol 15.8g. were reacted at 65 °C for 1 hr. After reaction, this solution was filtered and dried in air at 80 °C for 3 hr. To prepare  $Y_2O_3:S:Eu$  coated with PVP/TEOS or PMMA/TEOS nanocomposite by method P4, VP 2.36g (or MMA 10.485g) and AIBN 0.0118g were added to the solution used in method P3. The surface morphologies of these coated phosphors were investigated with HR-TEM (model: JEM 3010, JEOL, Japan). To confirm improved long-term stability of the coated phosphors, aging of samples were performed and then the photoluminescence properties of the coated phosphors were compared with those of uncoated phosphors.

### Results and discussion

Silica coated  $Y_2O_3:S:Eu$  red phosphor was prepared from a sol-gel process. The average size of the prepared silica nanoparticles was smaller than 10 nm.



**Figure 1.** The surface morphology of  $Y_2O_3:S:Eu$ ; (a) pure  $Y_2O_3:S:Eu$ ; (b) silica coated  $Y_2O_3:S:Eu$  by method P1; (c)  $Y_2O_3:S:Eu$  coated with PVP/TEOS by method P4; (d)  $Y_2O_3:S:Eu$  coated with PMMA/TEOS by method P4; (e) sample (c) after firing at 500 °C; (f) sample (d) after firing at 500 °C.

Dip-coating methods, i.e., methods P1 and P2, were not effective on the phosphor coating. As shown in Figure 1- (b), phosphor surface was coated irregularly with small amount of silica nanoparticles. Since silica nanoparticles synthesized from TEOS contain hydroxyl groups on the surface, regular coating onto the hydrophobic phosphor surface could not be expected. Surface morphology of phosphor prepared by method P3 was similar with that prepared by method P1. Figures 1-(c) and 1-(d) exhibited surface morphology of phosphor coated by method P4. When PVP or PMMA used as a binder, large amount of silica existed on the surface of phosphor. When nanocomposite composed of PVP matrix and silica nanoparticles was used as coating materials, island-type aggregates were observed on the phosphor surface. The resulting morphology might stem from hydrophilicity of PVP. On the other hand, when hydrophobic PMMA used as a matrix for the embedding of silica nanoparticles, phosphor surface was coated uniformly. Figure 1-(e) and (f) showed surface morphology of phosphor after removing polymer with firing at 500 °C for one hour. Aggregates of silica nanoparticles still exist on the phosphor surface when nanocomposite containing PVP was used, while silica nanoparticles were evenly coated on the phosphor surface when nanocomposite containing PMMA was used. The PL intensities of the coated phosphors were slightly lower than that of the pure  $Y_2O_3:S:Eu$  except phosphor coated with nanocomposite containing PMMA.  $Y_2O_3:S:Eu$  phosphor coated with PMMA nanocomposite exhibited improved PL intensities in both before and after firing. Long-term stability of phosphors prepared here was compared with that of uncoated phosphor by conditioning at 85 °C and 85 % relative humidity for a week. Phosphors coated with silica nanoparticles or nanocomposites exhibited better long-term stability than uncoated pure phosphor.

### Conclusions

$Y_2O_3:S:Eu$  red phosphors coated with silica nanoparticles or nanocomposites composed of polymer matrix and silica nanoparticles were prepared via sol-gel process. Among various coating methods examined here, uniform coating of phosphor could be achieved by using method P4, i.e., phosphor surface coating in a solution containing hydrophobic monomer and TEOS. Furthermore,  $Y_2O_3:S:Eu$  red phosphor coated with nanocomposite composed of PMMA matrix and silica nanoparticles exhibited enhanced PL intensity and long-term stability.

### References

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