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Optical Properties of the (Y,Gd)BO₃:Eu³⁺ Phosphor Coated with Al₂O₃ for a Plasma Display Panel

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The surface of (Y,Gd)BO₃:Eu³⁺ phosphor, a red-emitting source in the plasma display panel (PDP), was coated with Al₂O₃ nano-particles. Colloidal alumina oxide was selected as a coating material for the surface modification of the (Y,Gd)BO₃:Eu³⁺ phosphor. A modified sol-gel method was used for coating the surface of the phosphor by Al₂O₃ nano-particles. Al₂O₃ nano-particles in the surface of the phosphor were observed by Scanning Electron Microscope. It was found that the luminance intensity was increased in the photo-luminescence (PL) by a suppression of the nonradiative recombination via surface defects. The experimental results suggest that the surface coating of phosphors with Al₂O₃ nano-particles leads to an increase in the luminance intensity of the PDP and the gas discharge lamps.

Keywords: Al₂O₃ nano-particles; (Y,Gd)BO₃:Eu³⁺ phosphor; red-emitting; sol-gel method; surface coating

INTRODUCTION

There have been various intensive efforts [1–5] to improve the poor characteristics of the luminance and the life span of the phosphors. The surface modification for improving the characteristics has been done for In₂O₃ [6], Y₂SiO₅ [7], and Y₂O₃ [8]. To avoid the phosphor degradation, coating the phosphor surface with MgO has been introduced in earlier works [2,11,12], indicating that a surface

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treatment is an effective way to improve the properties of phosphors. For the purpose of both increasing luminance and preventing the degradation of phosphors, several studies [1,5,9] regarding surface treatment have been done. The blue $\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$ coated with SiO_2 nano-particles is a typical example [1]. Thus, surface coating is known to be an important technique for improving phosphor characteristics.

The modified sol-gel method used for surface treatment in this work was a simple process, compared to other chemical methods [2,6–9]. In order to obtain the phosphors with better luminance performance, we investigated the optical properties of the $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphors coated with Al_2O_3 -nanoparticles.

EXPERIMENTAL

The commercial red phosphor $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ (LG CHEM, LDT) was coated with the Al_2O_3 by a modified sol-gel method where Al_2O_3 coating of the phosphor was performed by using colloidal Al_2O_3 (Al20; Nyacol Nano Technologies, Inc.) as the precursor material. To make the concentration of colloidal Al_2O_3 , fixed at a value, we diluted the Al_2O_3 with the de-ionized water. The $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphor was stirred in the diluted colloidal Al_2O_3 of about 0.04 wt% concentrations. Considering the zeta-potential of the solution, the pH of the solution was adjusted with NaOH at a constant pH 10. The solution was stirred with a magnetic bar for an hour. The phosphor suspensions were washed three times in ethanol. The Al_2O_3 -coated phosphors were obtained by filtering and drying at 80°C for 12 h.

In order to compare the electro-optical properties of the coated red phosphor with the non-coated one under the same driving condition, we fabricated a test panel which was shown in Figure 1. The coated and the non-coated red phosphor powders used in the test panel were mixed with a vehicle, composed of ethylcellulose (10 wt%), diethylene

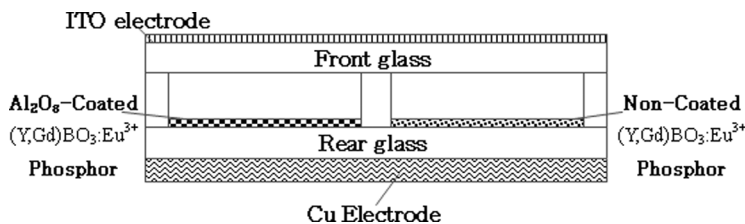


FIGURE 1 The cross sectional view of the test panel.

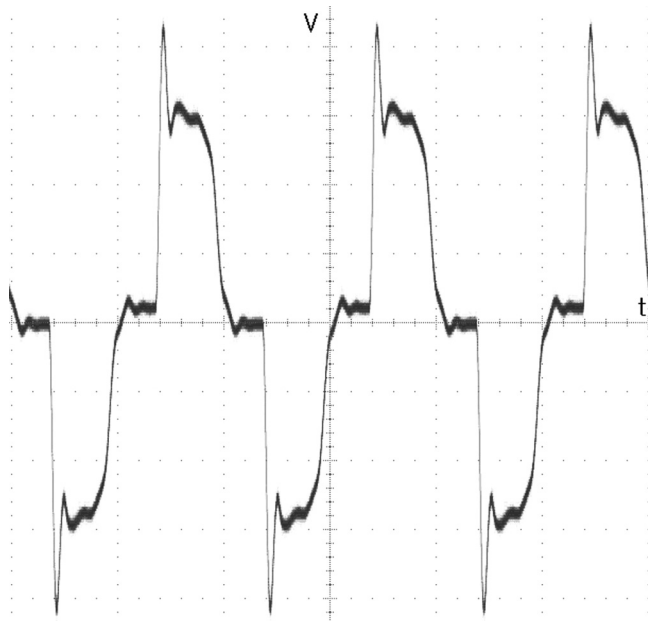


FIGURE 2 The waveform of the driving pulse (duty 20%, 20 kHz, 2.2 kV).

glycol monobutyl ether (10 wt%), and diethylene glycol monobutyl acetate (80 wt%). The dispensed thick films of phosphors were fired at 450°C for 30 min. to remove the organic components from the phosphors.

In the experiment of discharging the test panel, Xe gas pressure was fixed at 300 Torr and Ne gas pressure at 75 Torr. A driving pulse with a frequency of 20 kHz and a duty rate of 20% were applied to the test panel. The waveform of the driving pulse was plotted in Figure 2. All measurements were performed at room temperature. The comparison of the material properties between the Al_2O_3 -coated and the non-coated red phosphors was done by help of EDS (Energy Dispersive X-ray Spectrophotometer, HORIBA, Japan), XPS (X-ray Photoelectron Spectrometer, ESCALAB 250 XPS Spectrometer, VG Scientifics), SEM (Scanning Electron Microscope, HITACH S-4200), a spectrophotometer (SpectraPRO 2150i) and a luminance-meter (Minolta, CS-100).

RESULTS AND DISCUSSION

The SEM images of the non-coated and the Al_2O_3 -coated red phosphors are represented in Figure 3. The surface of the non-coated red

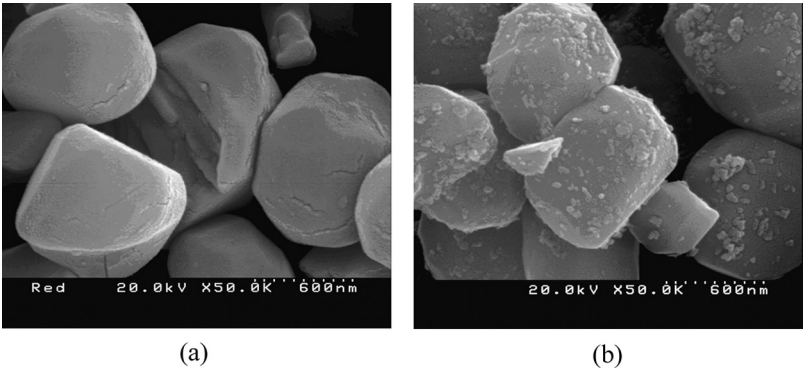


FIGURE 3 SEM Image of the phosphors (a) before and after (b) Al_2O_3 coating.

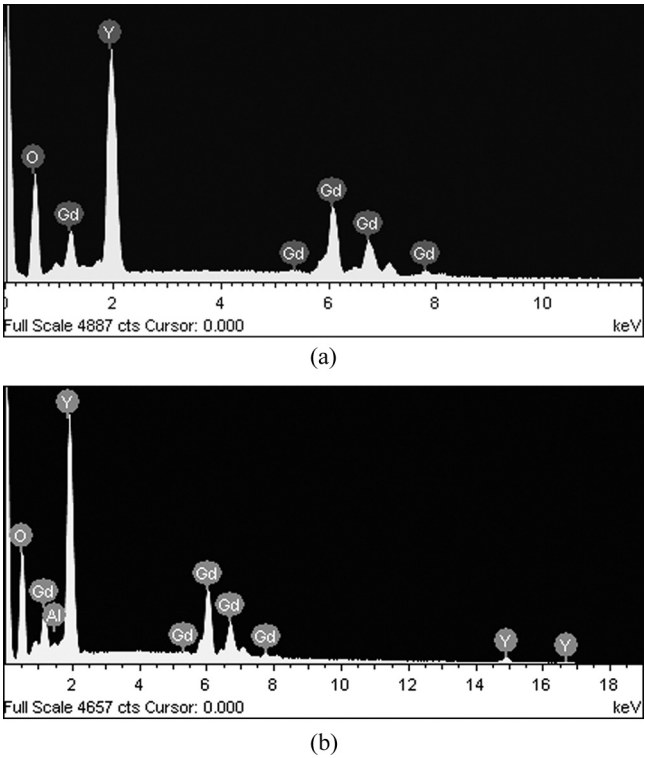


FIGURE 4 EDS of the phosphors (a) before and after (b) Al_2O_3 coating.

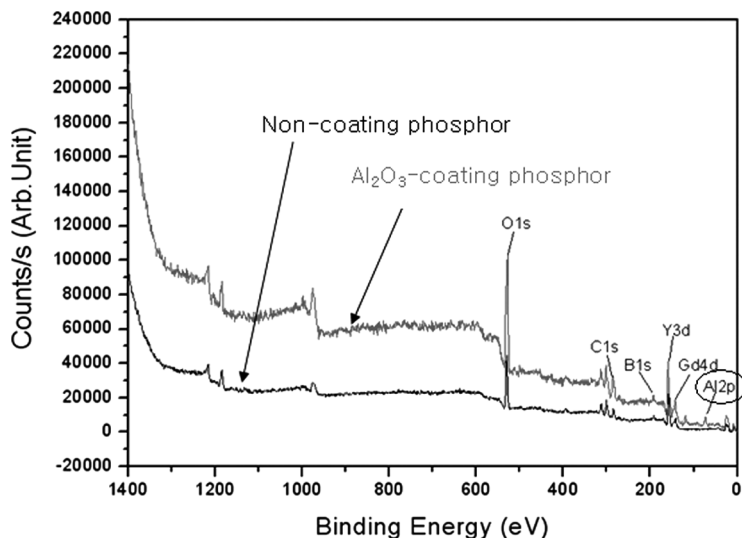


FIGURE 5 XPS of the phosphors before (down line) and after Al_2O_3 coating (upperline).

phosphor being $1\sim 2\mu\text{m}$ in size can be clearly seen. Nano-sized Al_2O_3 particles coated on the surface of phosphors are also seen.

The EDS of the non-coated and the Al_2O_3 -coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphors are shown in Figure 4. In the Figure 4(b), the Al related

TABLE 1 Atomic % of the Phosphors Before (a) and After Al_2O_3 Coating (b)

Name	At. (%)
(a) Non-coating Phosphor	
B1s	14.16
C1s	18.64
Eu3d5	0.13
Gd4d	2.75
O1s	50.46
Y3d	13.86
(b) Al_2O_3 -coating Phosphor	
Al2p	8.05
B1s	8.49
C1s	14.32
Gd4d	1.28
O1s	58.07
Y3d	9.79

peak was observed, and thence we can confirm that the particles as seen in Figure 3(b) are composed of Al_2O_3 .

In order to reconfirm Al_2O_3 nano-particles on the surface of the coated phosphors, we performed the XPS experiments. The Figure 5

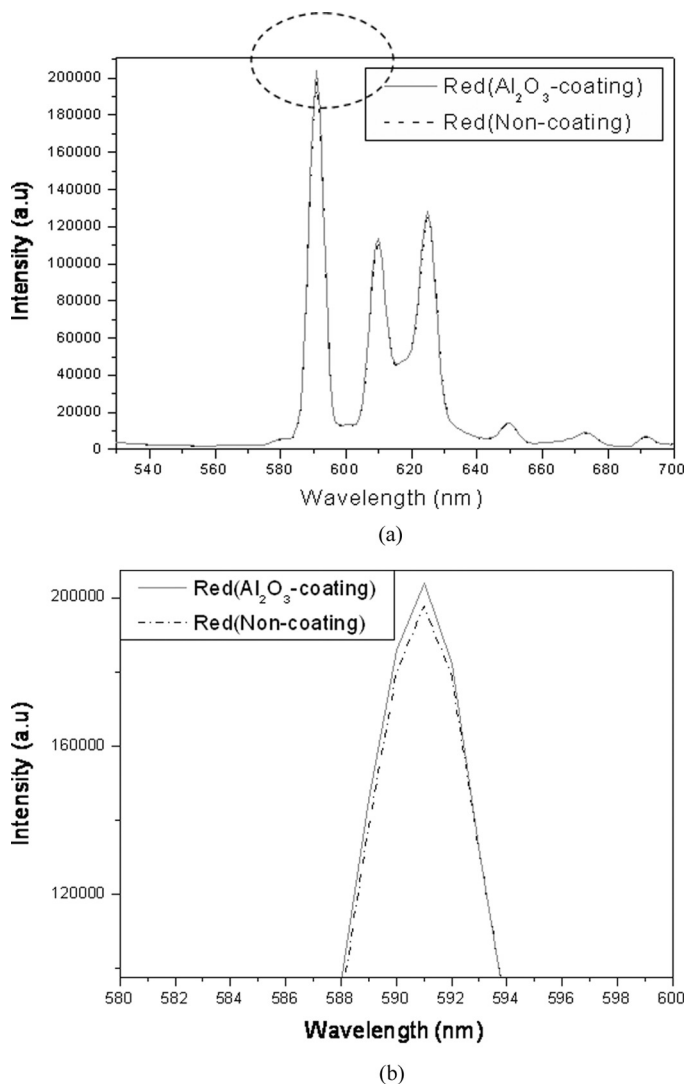


FIGURE 6 PL spectra of the phosphors (a) the full range PL spectrum and (b) the partial range PL spectrum dotted in the circle of (a).

show XPS results of the the non-coated and the Al_2O_3 -coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphors. Table 1(a) and (b) tell the relative atomic concentration of the surfaces of the non-coated and the Al_2O_3 -coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphors, obtained from the analysis of the XPS experiments in Figure 5. The binding energy peak at 74.1 eV, related to an Al element was detected from the XPS results. The relative concentrations of the alumina (Al_{2p}) element corresponding to the Al_{2p} peaks at 74.1 eV in Figure 5 was measured about 8% in the surface of the Al_2O_3 -coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphors as listed in Table 1 (b). The relative concentration of the oxygen (O_{1s}) element corresponding to the $\text{O}1s$ peaks at 530 eV in Figure 5 was increased form about 50% to 58% as listed in Table 1(a) and (b). The XPS results of Al_{2p} reveal strongly that Al_2O_3 nano-particles are coated on the surface of phosphors.

In Figure 6, the PL spectra of the non-coated and Al_2O_3 -coated red phosphors powders, excited by an ultraviolet (UV) with a wavelength of 254 nm, are shown. The PL spectrum shows that the intensity of Al_2O_3 -coated phosphor is higher than that of non-coated one without a wavelength shift in the core-level 4f–4f transition region, indicating that the presence of Al_2O_3 -particles on the surface effects on PL intensity. This can be explained, based on the suppression of the non-radiative recombination via surface defects [10].

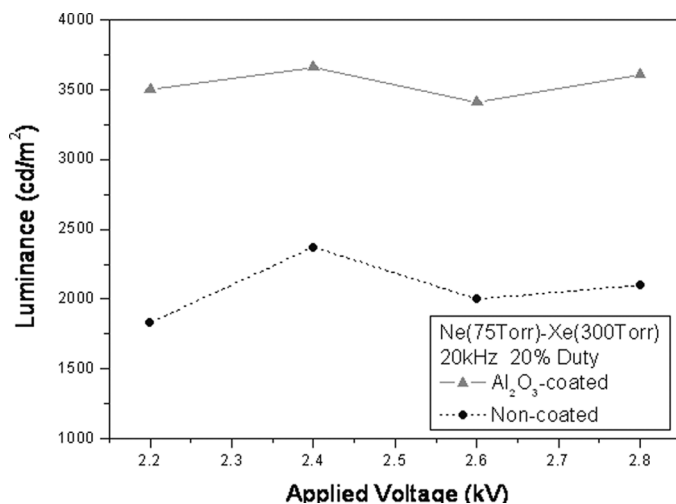


FIGURE 7 Luminance of the non-coated and Al_2O_3 -coated red phosphors in the test panel.

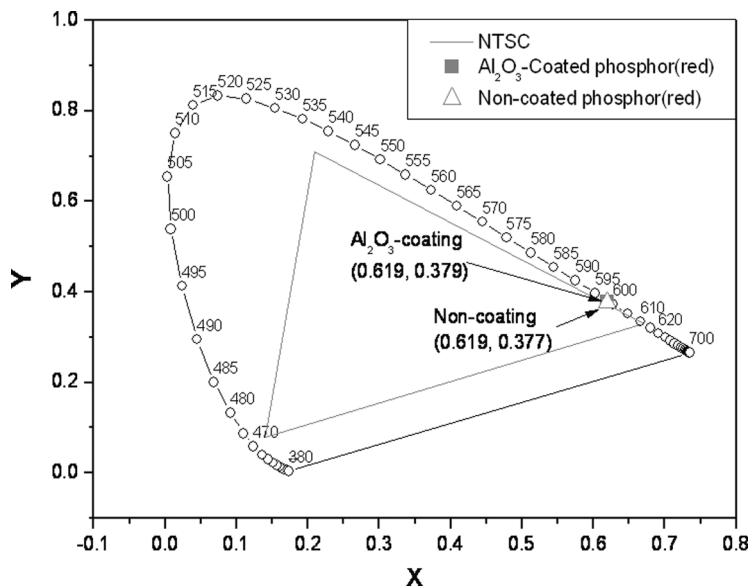


FIGURE 8 Color coordinates of the non-coated and Al_2O_3 -coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphors in the test panel. (duty 20%, 20 kHz, Xe 300 Torr and Ne 75 Torr).

In Figure 7, the luminance of the test panels is plotted as a function of the driving voltages. It is of interest that the test panel with the Al_2O_3 -coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphor shows much higher luminance than the panel with the non-coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphor. This fact implies that the surface coating of phosphors with a proper oxide is a way for improving the luminance of PDP devices or the gas discharge lamps, based on the same mechanism as explained in Figure 7.

Figure 8 shows that the color coordinates of the two phosphors, indicating that the surface coating gives rise to a negligible change in the color purity.

CONCLUSIONS

We prepared the $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphor coated with Al_2O_3 nano-particles by a modified sol-gel method and compared optical properties of the Al_2O_3 -coated phosphor with those of the non-coated $(\text{Y,Gd})\text{BO}_3:\text{Eu}^{3+}$ phosphor. The Al_2O_3 coating yields an increase in the both PL intensity of the phosphor powder and the luminance of

the test panel, due to the suppression of the nonradiative recombination via surface defects.

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