

Optical Properties of the $\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$ Phosphor Coated with Al_2O_3 by Using a Modified Sol-Gel Method

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The surface of $\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$ (BAM) phosphor, blue-emitting source in the plasma display panel (PDP), was coated with Al_2O_3 nano-particles. The surface modification of the phosphor was performed by a modified sol-gel method using the colloidal alumina as surface coating material. The surface morphologies of non-coated and Al_2O_3 coated BAM phosphors were compared by SEM (Scanning Electron Microscope). We observed the oxide nano-particles on the surface of Al_2O_3 coated phosphors. We also observed that the luminance intensity in the photo-luminescence (PL) was increased. This change in the PL intensity can be explained by a suppression of the nonradiative recombination via surface defects. The experimental results suggest that the surface modification of phosphors with nano-particles of oxide leads to an increase in the luminance intensity of red phosphor in the PDP and the gas discharge lamps.

Keywords BAM phosphor; blue-emitting; nano-particles; sol-gel method; surface coating; Al_2O_3

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 In_2O_3 [6], Y_2SiO_5 [7], and Y_2O_3 [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 treatment is an effective way for improving 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, and this study also aimed at the same purpose.

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

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Al_2O_3 coated phosphors with better luminance performance, we investigated the optical properties of the BAM phosphors coated with Al_2O_3 -nanoparticles.

Experimental

The commercial blue phosphor BAM (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 colloidal Al_2O_3 with the de-ionized water. The BAM 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 two times in ethanol. The Al_2O_3 -coated phosphors were obtained by filtering and drying at 80°C for 12 hrs.

In order to compare the electro-optical properties of the coated BAM phosphors with the non-coated one under the same driving condition, we fabricated a test panel which as seen in Figure 1. The Al_2O_3 -coated and the non-coated BAM phosphor

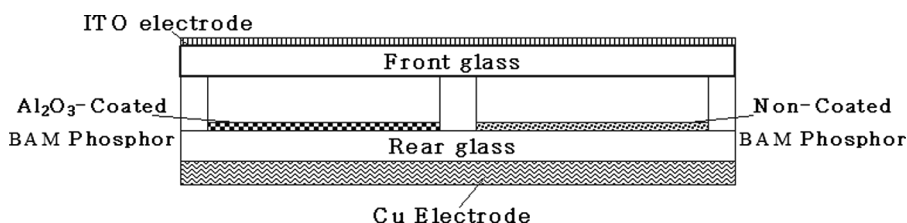


Figure 1. The cross sectional view of the test panel.

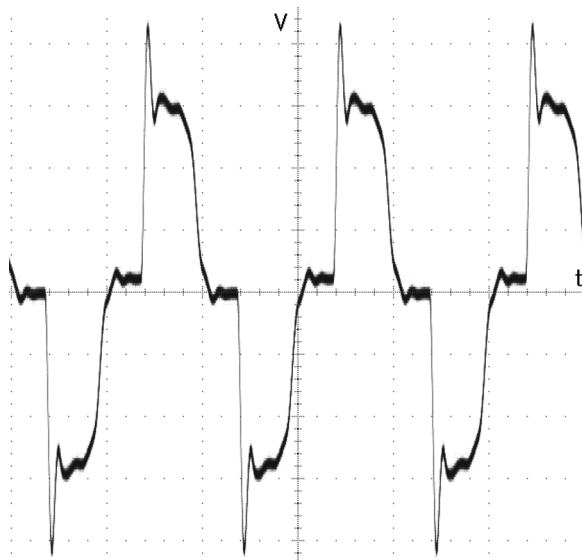


Figure 2. The waveform of the driving pulse. (duty 20%, 20 kHz, 2.2 kV).

powders dispensed in the test panel were mixed with a vehicle, composed of ethyl-cellulose (10 wt%), diethylene glycol monobutyl ether (10 wt%), and diethylene glycol monobutyl acetate (80 wt%). The vehicle was stirred at 80°C for 24 hours. The dispensed thick phosphor films 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 200 Torr and Ne gas pressure at 100 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 the 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 SEM (Scanning Electron Microscope, EDS (Energy Dispersive X-ray Spectrophotometer, HORIBA, Japan), XPS (X-ray Photoelectron Spectrometer, ESCALAB 250 XPS Spectrometer, VG Scientifics), 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 Al_2O_3 -coated red phosphors are represented in Figure 3(a). The surface of the non-coated BAM phosphor being 1~2 μm in size can be clearly seen. Nano-sized particles coated on the surface of Al_2O_3 coated phosphors are also seen in Figure 3(b).

The EDS of the non-coated and the multi-coated BAM phosphors are shown in Figure 4. As shown in the Figure 4(b), the Al related peak was observed, and thence we can see that the nano-particles as seen in Figure 3(b) are related to the Al peak in the Figure 4(b). In the Table 1, wt% and atomic% of components calculated from analysis of the EDS in Figure 4 were shown. The value related Al was increased from 30.68 atomic% to 31.89 atomic%, and thence we can confirm that the nano-particles as seen in Figure 3(c) are composed of SiO_2 and Al_2O_3 .

In order to reconfirm Al_2O_3 nano-particles on the surface of the coated phosphors, we performed the XPS experiments. Figure 5 represents XPS results of the non-coated and the Al_2O_3 -coated BAM phosphors. The binding energy peak at 76.27 eV, related to an Al element was detected from the XPS results of the

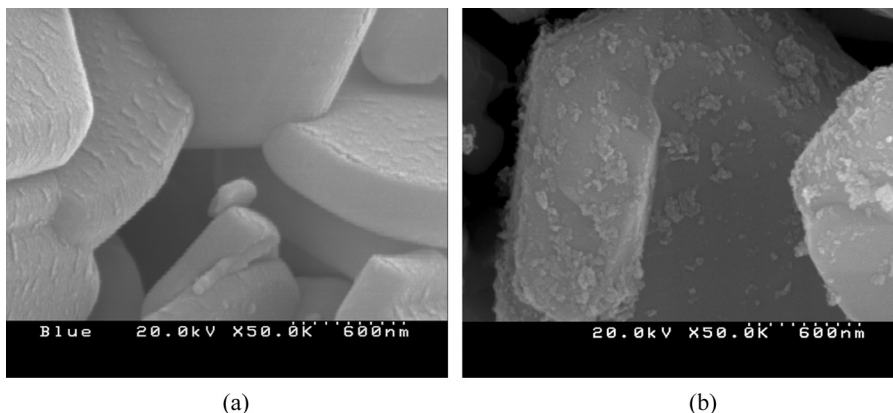
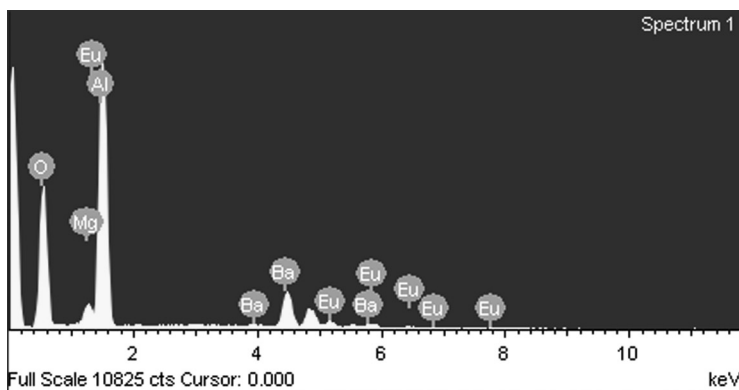
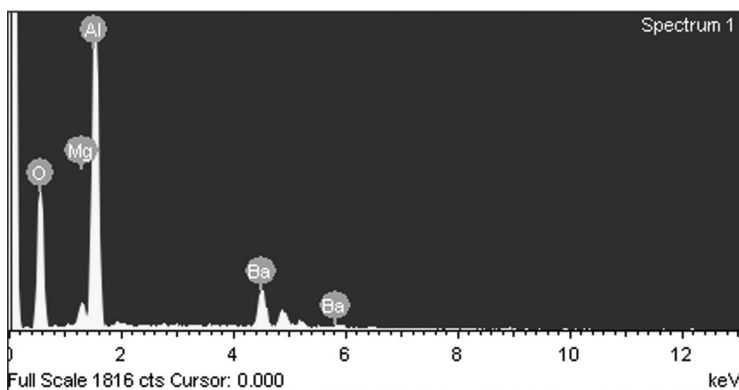


Figure 3. SEM Image of the BAM phosphors (a) before and after Al_2O_3 -coating (b).



(a)



(b)

Figure 4. EDS of the BAM phosphors (a) before and after Al_2O_3 -coating (b).

non-coated and Al_2O_3 phosphors. Table 2(a) and (b) tell the relative atomic concentrations of the surfaces of the non-coated and the Al_2O_3 -coated BAM phosphors, obtained from the analysis of the XPS experiments in Figure 5. The relative concentrations of the alumina ($\text{Al}2p$) element corresponding to the $\text{Al}2p$ peaks at 176.27 eV

Table 1. Composition from EDS of the phosphors before (a) and after Al_2O_3 -coating (b)

(a)			(b)		
Element	Weight %	Atomic %	Element	Weight %	Atomic %
O K	40.13	61.18	O K	41.16	61.92
Mg K	4.33	4.34	Mg K	2.63	2.60
Al K	33.94	30.68	Al K	35.76	31.89
Ba L	19.30	3.43	Ba L	20.45	3.58
Eu L	2.30	0.37			
Totals	100.00		Totals	100,00	

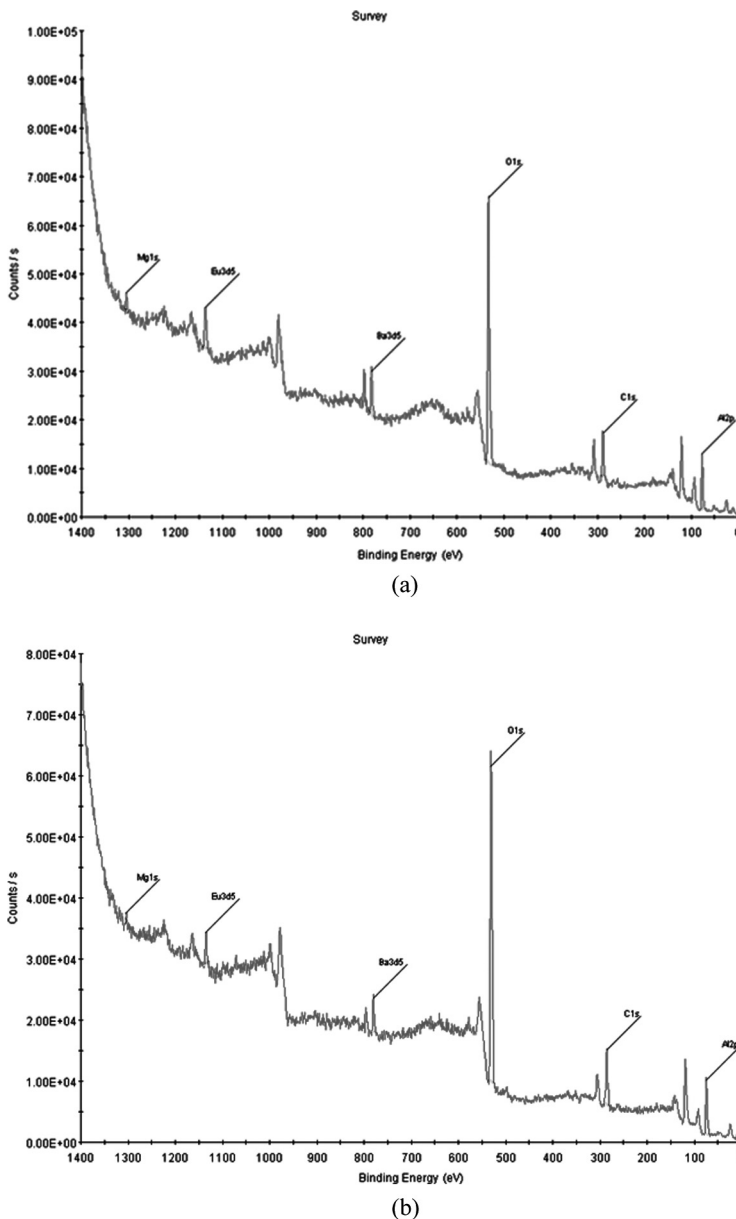


Figure 5. XPS of the phosphors before (a) and after single coating (b).

in Figure 5 was measured 31.54% in the surface of the non-coated BAM phosphors and 32.66% in that of the Al_2O_3 -coated one as listed in Table 2. The XPS results reveal strongly that the surfaces of Al_2O_3 -coated phosphors are coated with Al_2O_3 nano-particles.

In Figure 6, the PL spectra of the non-coated and Al_2O_3 -coated BAM 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 any wavelength shifts as much as 5%,

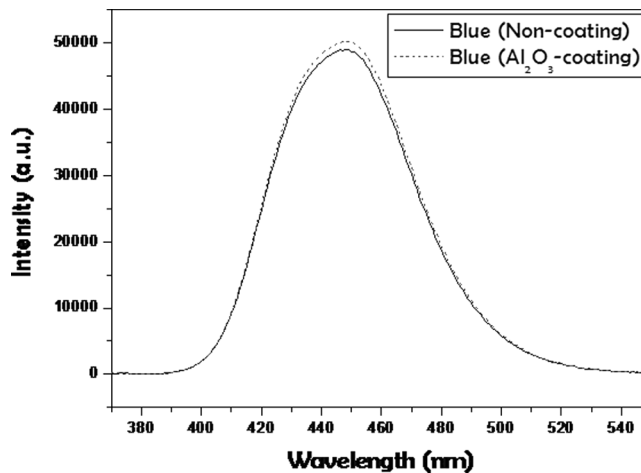
Table 2. Composition from XPS of the phosphors before (a) and after Al_2O_3 -coating (b)

(a)		(b)	
Name	At.%	Name	At.%
Al2p	31.54	Al2p	32.66
Ba3d5	0.84	Ba3d5	0.5
Cl s	16.24	Cl s	18.27
Eu3d	0.1	Eu3d5	0.15
Mg1s	1.05	M s1s	1.01
O1s	50.23	O1s	47.42

indicating that the presence of Al_2O_3 -particles on the surface of phosphors affects PL intensity. This can be explained, based on the suppression of the nonradiative recombination via surface defects [10] by a surface coating with oxides. Thus, the surface nano-coating can reduce the surface defects of phosphors, resulting in higher PL intensity.

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 BAM phosphor shows higher luminance than the panel with the non-coated BAM phosphor as much as about 15%. The luminance variation in Figure 6 differs from that seen in Figure 7 can be explained by the general feature that the luminance measured in the phosphor paste differs from that measured in the phosphor powder because of the dispersion properties involved. 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 6.

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

**Figure 6.** PL of the non-coated and the Al_2O_3 -coated BAM powder.

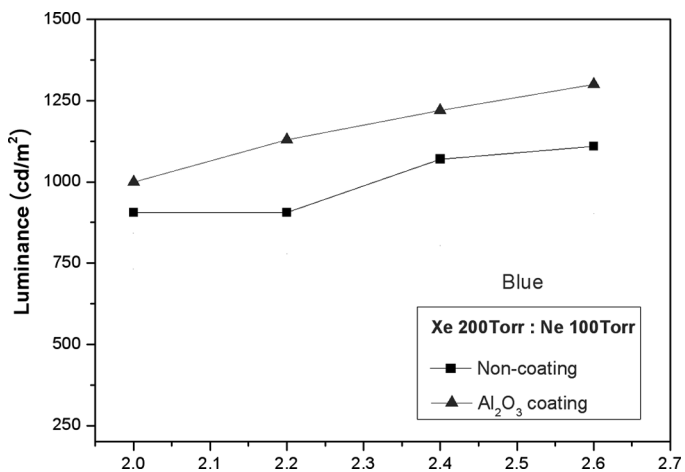


Figure 7. Luminance of the non-coated and Al₂O₃-coated BAM phosphors in the test panel.

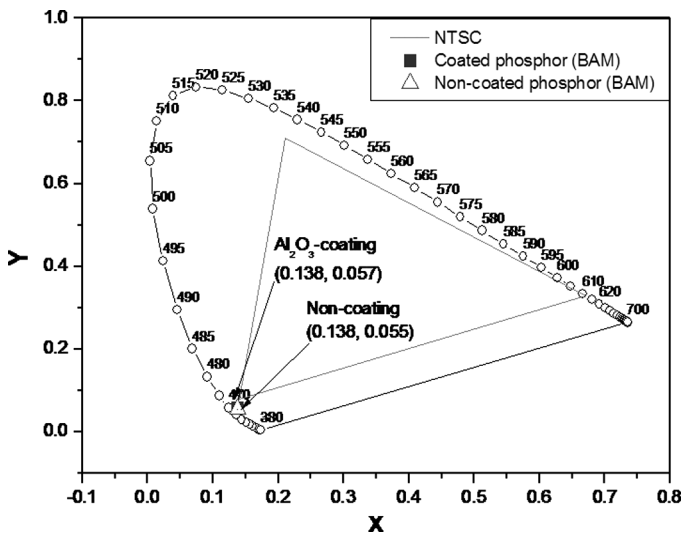


Figure 8. Color coordinates of the non-coated and Al₂O₃-coated BAM phosphors in the test panel. (duty 20%, 20 kHz., Xe 200 Torr and Ne 100 Torr).

Conclusions

We prepared the BAM phosphor coated with Al₂O₃ nano-particles by a modified sol-gel method and compared optical properties of the Al₂O₃-coated phosphor with those of the non-coated BAM phosphor. The Al₂O₃-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, suggesting an effective method to improve the phosphor performance of PDP.

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