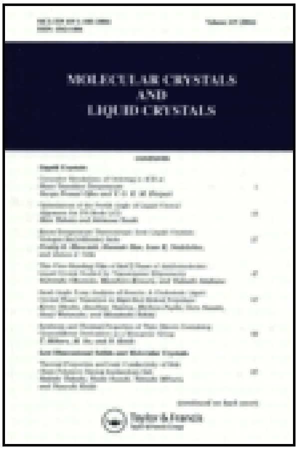


Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

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Version of record first published: 31 Jan 2007

To cite this article: Hyug Jong Kim, Min Wan Kim, Hyung su Kim, Hyu Suk Kim, Suk Hwan Kim, Sang Woo Lee, Byung Ho Choi, Bong Kyo Jeong & Hyuoung ho Lee (2006): Atomic Layer Deposition of Aluminum Oxide Thin Film on $BaMgAl_{10}O_{17}:Eu^{2+}$ Phosphor, Molecular Crystals and Liquid Crystals, 459:1, 239/[519]-245/[525]

To link to this article: http://dx.doi.org/10.1080/15421400600930375

*Mol. Cryst. Liq. Cryst.*, Vol. 459, pp. 239/[519]–245/[525], 2006

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400600930375



# Atomic Layer Deposition of Aluminum Oxide Thin Film on $BaMgAl_{10}O_{17}:Eu^{2+}$ Phosphor

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*An investigation is reported by the coating of $BaMgAl_{10}O_{17}:Eu^{2+}$ phosphor by aluminum oxide using atomic layer deposition. Aluminum oxide films were prepared the chamber temperature of 350°C using $Al(CH_3)_3$ and $H_2O$ as precursors and reactant gas, respectively. EDX and FTIR analysis showed the surface composition of coated phosphor was aluminum oxide. The photoluminescence intensity of coated phosphors showed 10.3 ~ 36% higher than that of uncoated. This means that the reactive surface is uniformly coated with stable aluminum oxide to reduce the dead surface layer without change of bulk properties.*

**Keywords:** aluminum oxide; atomic layer deposition; BAM phosphor; oxide coating; photoluminescence

## INTRODUCTION

$BaMgAl_{10}O_{17}:Eu^{2+}$ (BAM) has been widely used as a blue-emitting phosphor in backlight in liquid crystal display(LCD) and plasma

The study has been supported partially by Kumoh National Institute of Technology.
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display panel(PDP) because its high efficiency and chromaticity. However, the luminance decrease due to oxidation [1,2]. The degradation process is caused by several paths such as irradiation by ultraviolet photons, ion sputtering and baking process during PDP manufacture, etc. The problem of luminance decrease due to oxidation needs to solve. A few works were reported to overcome this problem [3–5].

In this study, ultrathin film $Al_2O_3$ on BAM was coated by atomic layer deposition using $Al(CH_3)_3$ and $H_2O$ as precursors and reactant gas, respectively. The effect of inorganic oxide passivation on the structural and optical properties of BAM phosphor was investigated as a function of film thickness.

## EXPERIMENTAL

The coating process was carried out in a vertical flow-type ALD reactor. Figure 1 shows a schematic diagram of ALD experimental system. $Al(CH_3)_3$ (trimethylaluminum; TMA) was evaporated from a boat at 25°C and was transpired with Ar carrier gas. The heating line was maintained at 100°C to prevent recondensation of $Al(CH_3)_3$. The chamber temperature was 350°C, controlling with a thermocouple. The working pressure in the reactor was about 0.5 torr. $H_2O$ was used as

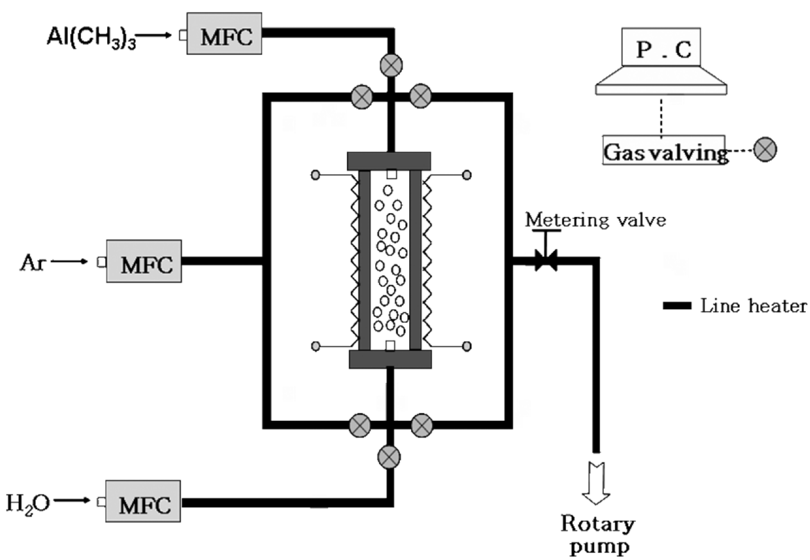


**FIGURE 1** A schematic diagram of ALD experimental system.

a reactant gas and Ar as carrier and purge gas. The opening and closing sequences of the air valves were controlled by using a personal computer. The exposure times were 1 s for the source material, then 4 s for the purging and a reactant gas was transpired for 1 s followed by the purging for 4 s. The average particle size was about 5 μm.

The composition of the films coated on phosphor powders was examined by Energy Dispersive X-ray Spectrometer(EDX, Inca Energy Analysis, Oxford). The interface structure was also measured by Fourier Transform Infrared Spectroscopy(FTIR, 460 Plus, JASCO). Field Emission Scanning Electron Microscopy(FE-SEM, JSM-6500F, Jeol) was used to investigate the surface morphology. The photoluminescence and lifetime of coated phosphors were measured using Spectroradiometer(PL, CS1000, Minolta).

## RESULTS AND DISCUSSION

ALD films are deposited by a respective process of single layer(or less than a layer) deposition sequences. Each sequence consists of several gas-surface interaction that are all self-limiting. However, in sol-gel process, there are many process variables, such as concentration of precursors, pH and temperature of solutions, which can affect the surface morphology [6–8]. However, in ALD process, Figure 2 showed that rough surface of uncoated phosphor became smoother and clearer as the number of ALD cycle increased from 300 to 700 cycles. On the contrary, it was reported that in sol-gel process, the surface of coated phosphor became rougher than uncoated [6].

Aluminum element could be detected by EDX analysis shown in Table 1. Al element peak increased, however, the other elements (Ba, Mg and Eu) consisting of phosphor decreased as the number of ALD cycle increased. Figure 3 shows the FTIR spectra of uncoated and coated phosphors. As can be seen, peaks for aluminum oxide are seen at the frequency of $\sim 1{,}000\,cm^{-1}$. As ALD cycle increased, the height of aluminum oxide peaks increased, due to thicker aluminum oxide films on the surface of phosphors. As a result, the surface of phosphor could be confirmed to be coated by aluminum oxide.

Figure 4 showed PL spectra of $BaMgAl_{10}O_{17}:Eu^{2+}$ phosphors with various ALD cycles. Up to 500 ALD cycles, PL intensity increased, but decreased above 600 cycles. This means that the reactive surface is coated with the aluminum-oxygen precursor and is stabilized by aluminum oxide coating and also its absorption is almost negligible due to ultrathin films up to 500 ALD cycles. Surface has high free energy due to the abrupt discontinuation of the bulk. The excess free energy reduces by rearrangement of aluminum oxide. This

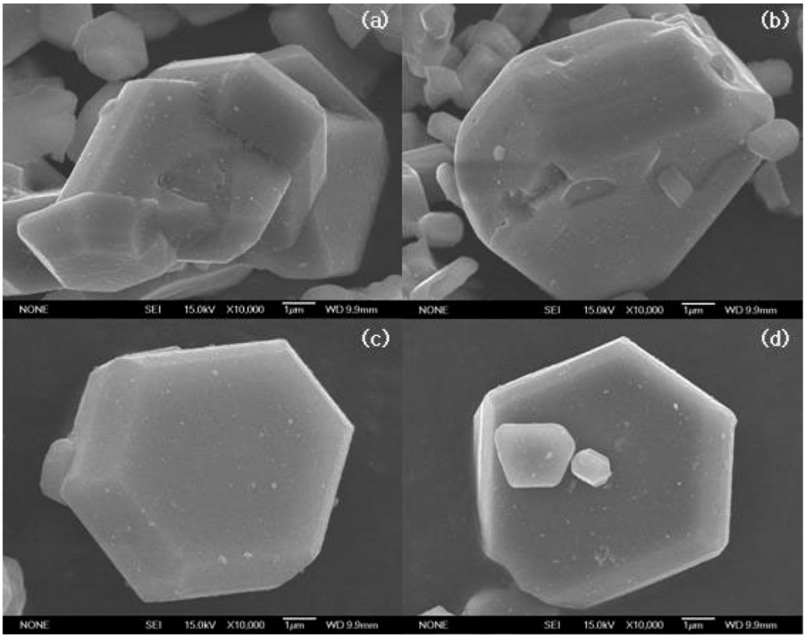


**FIGURE 2** FE-SEM photograph of $BaMgAl_{10}O_{17}:Eu^{2+}$ phosphors (a) uncoated, aluminium oxide coated by ALD (b) 300, (c) 500 and (d) 700 cycles.

phenomenon may also attribute to higher PL intensity [9]. Above 600 cycles, however, PL intensity decreased due to the absorption of thicker films.

In order to verify the resistance of the coated phosphor to moist air, PL intensity of phosphor was measured for 2,000 h in air as shown in

**TABLE 1** EDX Analysis of $BaMgAl_{10}O_{17}:Eu^{2+}$ Phosphors

| | EDX analysis (arb.%) | | | | |
|---|---|---|---|---|---|
| | O | Al | Ba | Mg | Eu |
| uncoated | 60.59 | 32.10 | 3.38 | 3.41 | 0.52 |
| coated by | | | | | |
| 300 cycles | 59.32 | 33.51 | 3.31 | 3.39 | 0.47 |
| 400 cycles | 58.74 | 34.25 | 3.29 | 3.28 | 0.44 |
| 500 cycles | 58.49 | 34.78 | 3.11 | 3.24 | 0.38 |
| 600 cycles | 57.78 | 35.86 | 2.99 | 3.08 | 0.29 |
| 700 cycles | 58.05 | 36.06 | 2.75 | 2.87 | 0.27 |

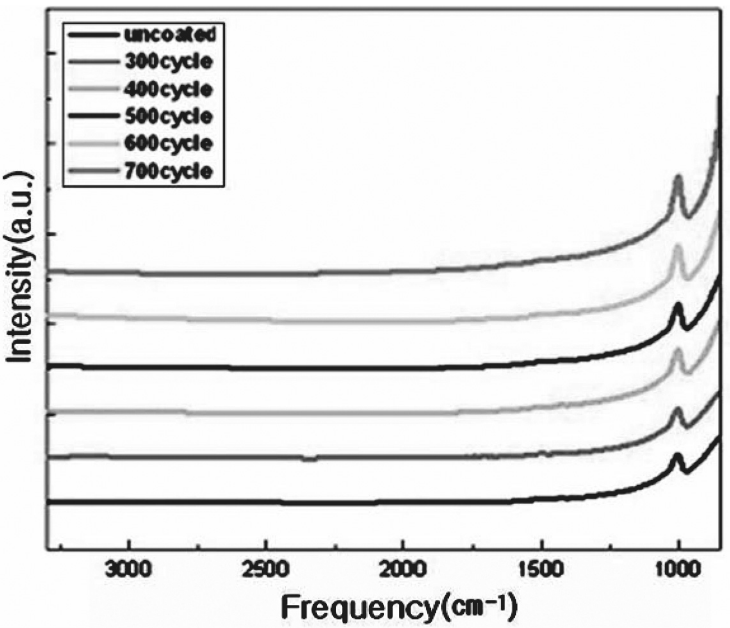


**FIGURE 3** FTIR spectra of $BaMgAl_{10}O_{17}$:$Eu^{2+}$ phosphors with various ALD cycles.



Figure 5. After 2,000 h, PL intensities of uncoated and coated phosphor(500 cycles) were 6.2 cd/m$^2$ and 10.74 cd/m$^2$, respectively. This value showed about 73.2% increase. Until now, it was generally reported that initial intensity of uncoated phosphor was higher than that of coated phosphor [6–8]. The inverse effect is probably due to ALD growth mechanism. It was also convinced that the films coated by ALD were more uniform, continuous and free of surface defects than that by sol-gel or CVD.

## CONCLUSIONS

In sol-gel or CVD process, PL intensity of coated phosphor decreased normally than that of uncoated due to oxide absorption and the aggregate of phosphor powder was also a main barrier for application. In ALD system, however, an ultrathin and uniform film can be controlled without the aggregate of phosphor powder. Resistance of the phosphor to moisture has been enhanced by ALD coating the ultrathin layer of

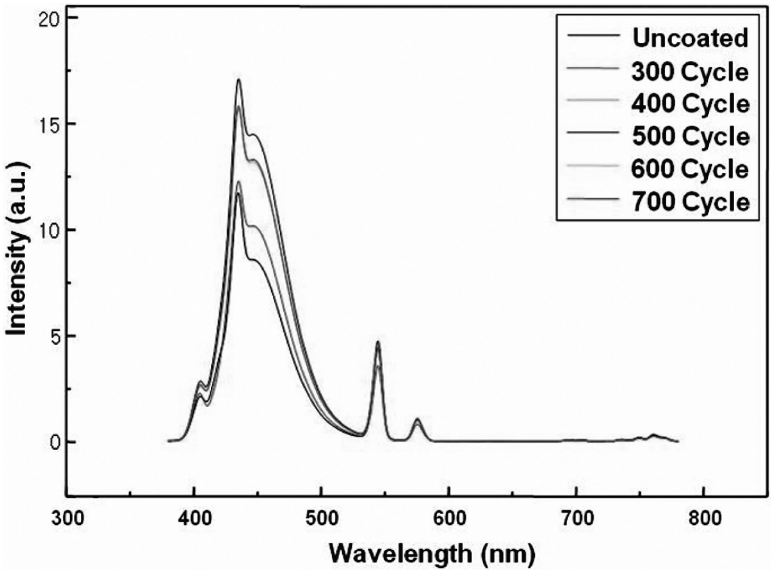


**FIGURE 4** PL spectra of $BaMgAl_{10}O_{17}:Eu^{2+}$ phosphors with various ALD cycles.

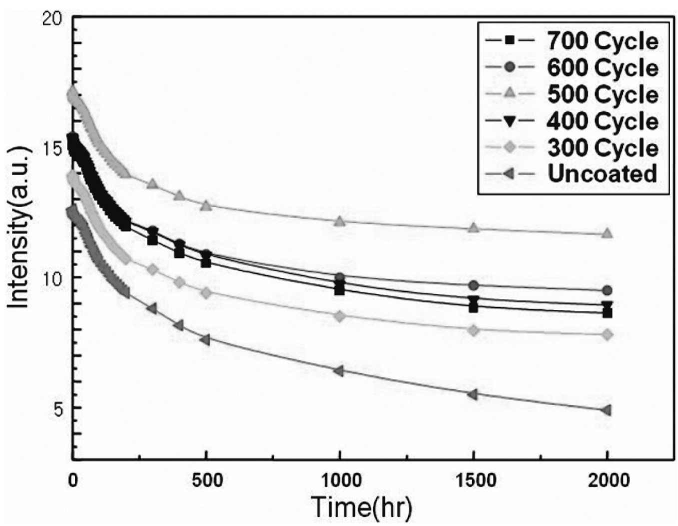


**FIGURE 5** PL lifetime spectra of uncoated and coated phosphors.

aluminum oxide on the surface of $BaMgAl_{10}O_{17}:Eu^{2+}$ phosphor. ALD process for oxide coating phosphors showed a remarkable improvement of PL intensity and PL lifetime.

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