

Improved Photoluminescence of $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$ Under VUV Excitation

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Abstract

We applied the spray pyrolysis technique to prepare Mn-doped $\text{BaMgAl}_{10}\text{O}_{17}$ (BAM) particles with high photoluminescence, which could be used in the plasma display device as a green phosphor. Several preparation conditions were investigated in order to tailor the vacuum ultraviolet (VUV) characteristics. Some portions of barium were replaced with strontium to improve the luminescent intensity of BAM:Mn particles under VUV excitation. The content of Mn and Sr was optimized to obtain high luminescent efficiency under VUV excitation. Finally, the optimized BAM:Mn green particles showed higher photoluminescence intensity than that of commercial Zn_2SiO_4 and comparable with commercial barium-aluminate phosphor.

1. Introduction

An enhanced performance of displays and lamps requires an improved quality of the phosphors for high brightness and long-term stability. One way to meet this requirement is to discover new phosphors. The other way is to enhance performances of the existing phosphor materials in terms of shape, crystallinity, purity, mean size, size distribution, and dopant distribution. The latter way is frequently applied in the well-established displays such as cathode ray tubes (CRTs), plasma display panel (PDP), and fluorescent lamps (FLs).

Recently, plasma display panels (PDPs) are considered as the most potential display applicable to high-definition TV attachable on the wall as a home theater. The phosphor materials are of important parts in the PDPs because they directly influence the brightness and the lifetime. So, much attention is given to design and prepare high efficient phosphor materials [1, 2].

For the application to PDPs, the phosphor materials

should efficiently absorb the vacuum ultraviolet (VUV) light emitting from the plasma discharge and transfer the absorbed light energy to the luminescent centers. A representative green phosphor currently used in the plasma display panels (PDPs) is $\text{Zn}_2\text{SiO}_4:\text{Mn}$ which is known to have a little long decay time and high discharging voltage [3]. So, other phosphors such as $\text{YBO}_3:\text{Tb}$ and $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$ are additionally necessary to optimize the luminance property of PDP [4].

The characteristics of phosphor particles are strongly affected by synthetic process. The conventional solid-state reaction process has disadvantages in controlling the morphology and maintaining uniformity in composition of phosphor particles. Therefore, many new processes to overcome these disadvantages are under investigation. Spray pyrolysis is one of the promising processes for the preparation of multi-component powders like phosphor particles. Phosphor particles synthesized by spray pyrolysis have spherical shape, fine size, and relative uniformity in phase composition because of the micro-scale reaction within a droplet and the lack of milling process. However, despite these advantages, phosphor particles prepared by spray pyrolysis suffer from problems such as hollowness and high porosity, which cause the reduction in brightness and long-term stability. Recently, we have developed many new techniques to overcome the problems mentioned above [5].

In this work, spray pyrolysis was applied to prepare Mn-doped $\text{BaMgAl}_{10}\text{O}_{17}$ (BAM) as the green phosphor for PDP. In order to obtain spherical-shaped particles, the spray solution was controlled. It was also tried to improve the luminescence intensity of BAM:Mn particles under VUV excitation by replacing some portion of magnesium with strontium and optimize the luminescent intensity by varying the content of Mn and Sr.

2. Experimental

A pilot-scale ultrasonic spray pyrolysis process used in this work was schematically shown in Figure 1. The apparatus consists of an ultrasonic aerosol generator with six vibrators (1.7 MHz), a quartz tube (length - 1200 mm and inner diameter - 50 mm), and a particle collector. The flow rate of air used as a carrier gas was 40 L/min. The reactor temperature was fixed as 900 °C.

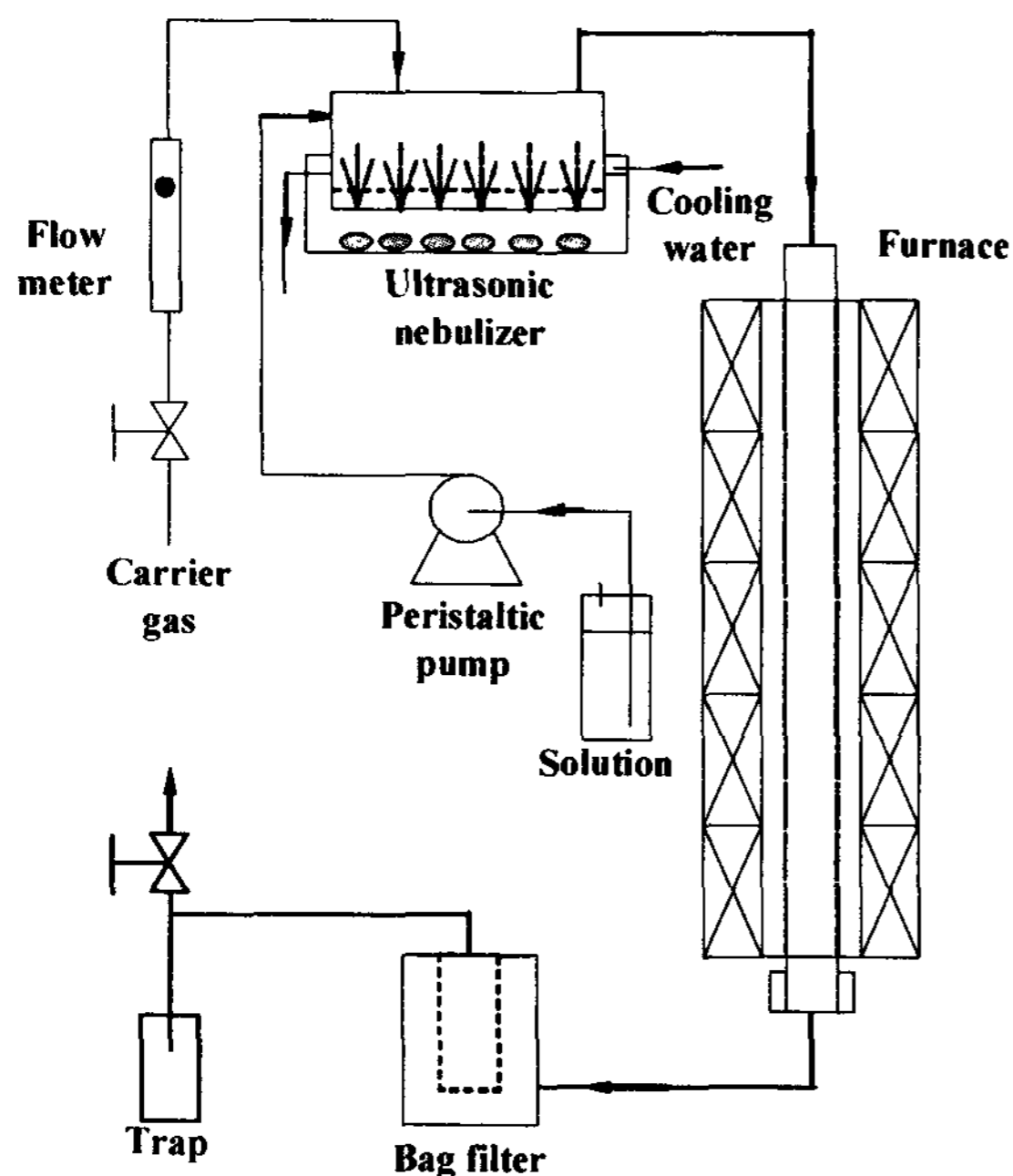


Fig 1. Schematic diagram of the used pilot-scale spray pyrolysis process.

The $\text{BaM}:\text{Mn}^{2+}$ phosphor particles were prepared by the spray pyrolysis from aluminum polycation precursor solutions. More detailed procedure for the preparation of aluminum polycation was elsewhere in the reference [6]. Barium, magnesium, manganese salts were used as the precursors and dissolved in the prepared aluminum polycation solution. The total concentration of the precursor solution was kept as 0.8 M. The content of Mn activator was varied from 0.01 to 0.4 in a molar ratio with respect to Mg. Also, the influence of Sr substitution for the Ba sites was monitored.

All as-prepared particles were posttreated at 1400 °C for 3 h under the reduction atmosphere. The morphology of prepared phosphor particles were identified by scanning electron microscopy (SEM).

The photoluminescence characteristics of prepared particles were measured under vacuum ultraviolet (147 nm) generated from a Kr_2 excimer lamp.

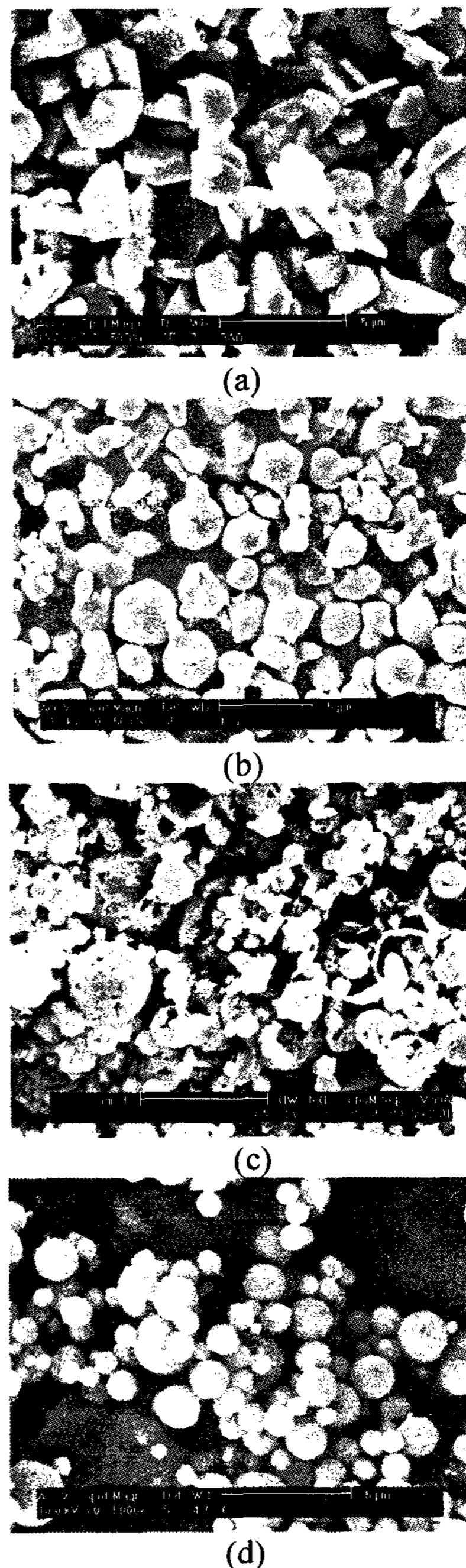


Fig 2. SEM photos;(a) commercial $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$, (b) commercial $\text{Zn}_2\text{SiO}_4:\text{Mn}$, (c) hollow BAM particle prepared from the nitrate solution, and (d) Dense BAM:Mn prepared from the aluminum polycation.

3. Results and Discussion

When the phosphor particles are prepared by the spray pyrolysis, the solution condition greatly influences the morphology of finally obtained powder as a consequence of the different particle formation mechanism. Figure 2 show SEM photos of BAM:Mn phosphor particles prepared and commercial available $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$ (called 'BAO' in this work) and $\text{Zn}_2\text{SiO}_4:\text{Mn}$ particles. All lab-made samples in Figure 2 were posttreated at 1400 °C. The commercial BAO particles which are synthesized by the solid-state reaction route have a plate shape due to the crystallographic characteristic of barium hexaaluminate. Also, the commercial $\text{Zn}_2\text{SiO}_4:\text{Mn}$ phosphor has irregular shape and the size of about 2~3 μm . As shown in Figure 2 (c), when using the nitrate solution wherein only nitrate precursors are dissolved in water, the prepared Mn-doped $\text{BaMgAl}_{10}\text{O}_{17}$ particles showed hollow and porous structure. On the other hand, the BAM:Mn particles prepared by using the aluminum polycation had spherical shape with filled morphology.

Figure 3 shows the influence of the activator concentration on the emission intensity of BAM:Mn phosphor particles when they are excited by VUV (147 nm). The concentration quenching was observed when the content of Mn was over 10 at. % with respect to Mg element. So, we fixed the Mn concentration at 10 at. %. Figure 4 shows the photoluminescence spectra obtained under VUV (147

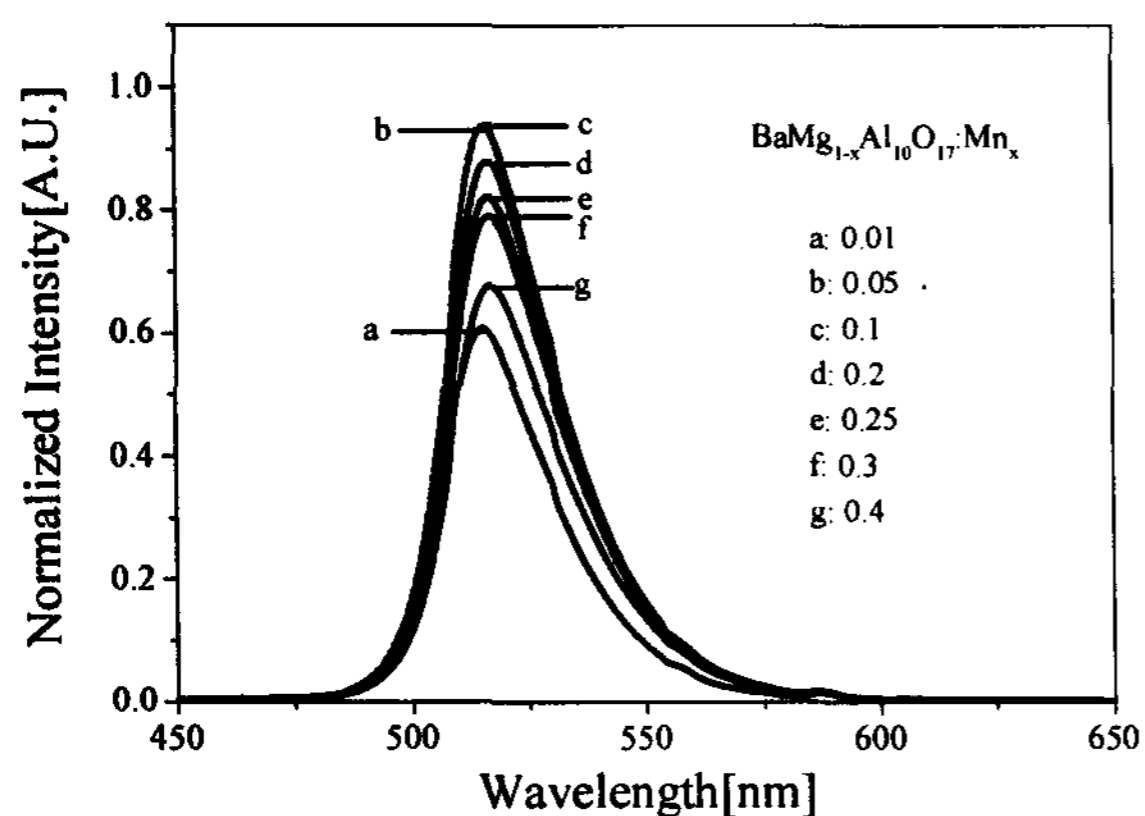


Figure 3 Effect of Mn concentration on the photoluminescence intensity of BAM:Mn phosphor particles prepared by the spray pyrolysis using the aluminum-polycation solution

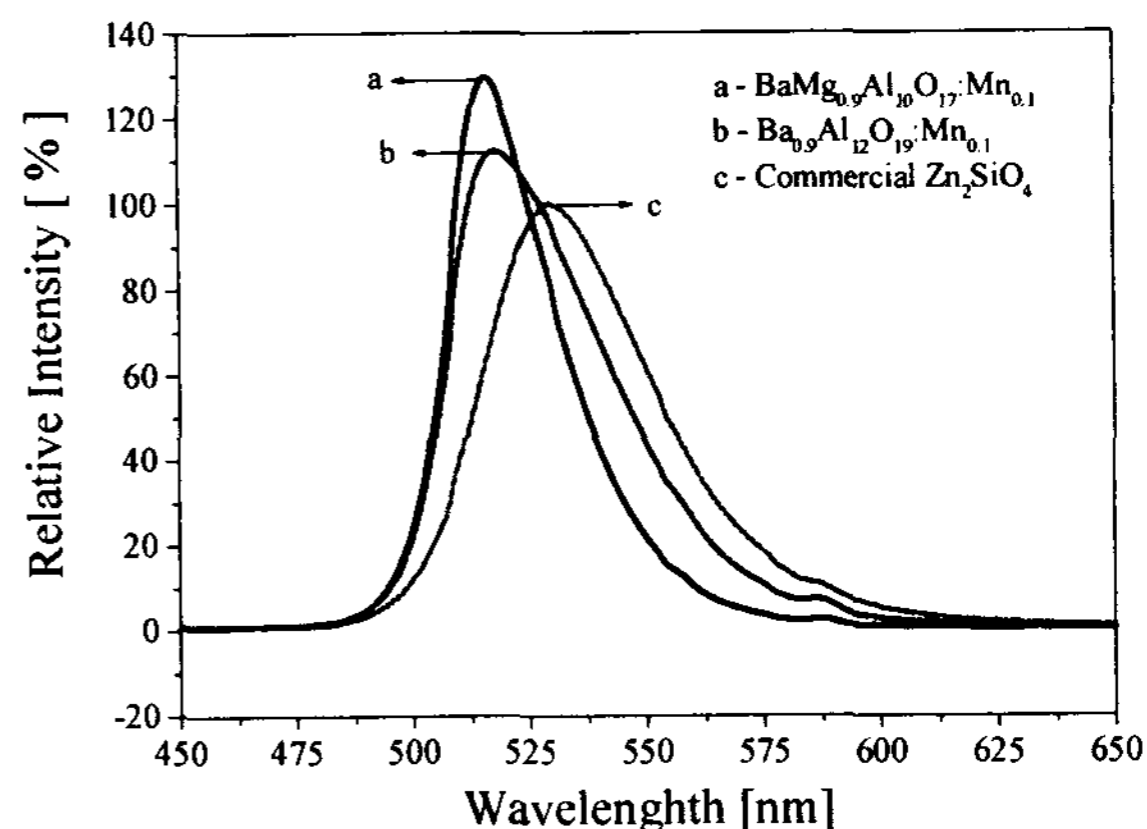


Figure 4 Emission spectra of lab-made $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$, $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$, and commercial $\text{Zn}_2\text{SiO}_4:\text{Mn}$ phosphor ($\lambda_{\text{ext}} = 147 \text{ nm}$).

nm) for $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$ (lab-made), $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$ (lab-made), and commercial $\text{Zn}_2\text{SiO}_4:\text{Mn}$ particles. Both BAO:Mn and BAM:Mn showed higher luminescent intensity compared with the commercial $\text{Zn}_2\text{SiO}_4:\text{Mn}$ particles. $\text{BaMgAl}_{10}:\text{Mn}$ phosphor also had higher luminescent intensity than that of $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$ phosphor. However, the band width of $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$ particles was greater than that of the $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$ particles, which means that the BAM:Mn phosphor has lower brightness.

To improve the brightness of BAM:Mn phosphor, in this work, strontium was substituted into the Ba sites at a fixed Mn concentration (10 at. %). As a result, the improvement in the photoluminescence intensity under VUV excitation was achieved as shown in Figure 5. It was found that the photoluminescence intensity of BAM:Mn phosphor was influenced by the Sr substitution. The highest luminescent intensity of $(\text{Ba}_{1-x}, \text{Sr}_x)\text{MgAl}_{10}\text{O}_{17}:\text{Mn}$ phosphor was achieved at $x = 0.4$. When 100 % of Ba was replaced with Sr, the $\text{SrMgAl}_{10}\text{O}_{17}:\text{Mn}$ still have good photoluminescence intensity comparable with the $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$ particles as shown in Figure 5. The luminescent intensity of $(\text{Ba}, \text{Sr})\text{MgAl}_{10}\text{O}_{17}:\text{Mn}$ phosphor was about 15 % improved in comparison with the BAM:Mn phosphor and 98 % of the commercial barium aluminate. The wavelength at the peak in the photoluminescence spectra, the relative emission intensity and the brightness for commercial

zinc silicate and barium aluminates were summarized in Table 1.

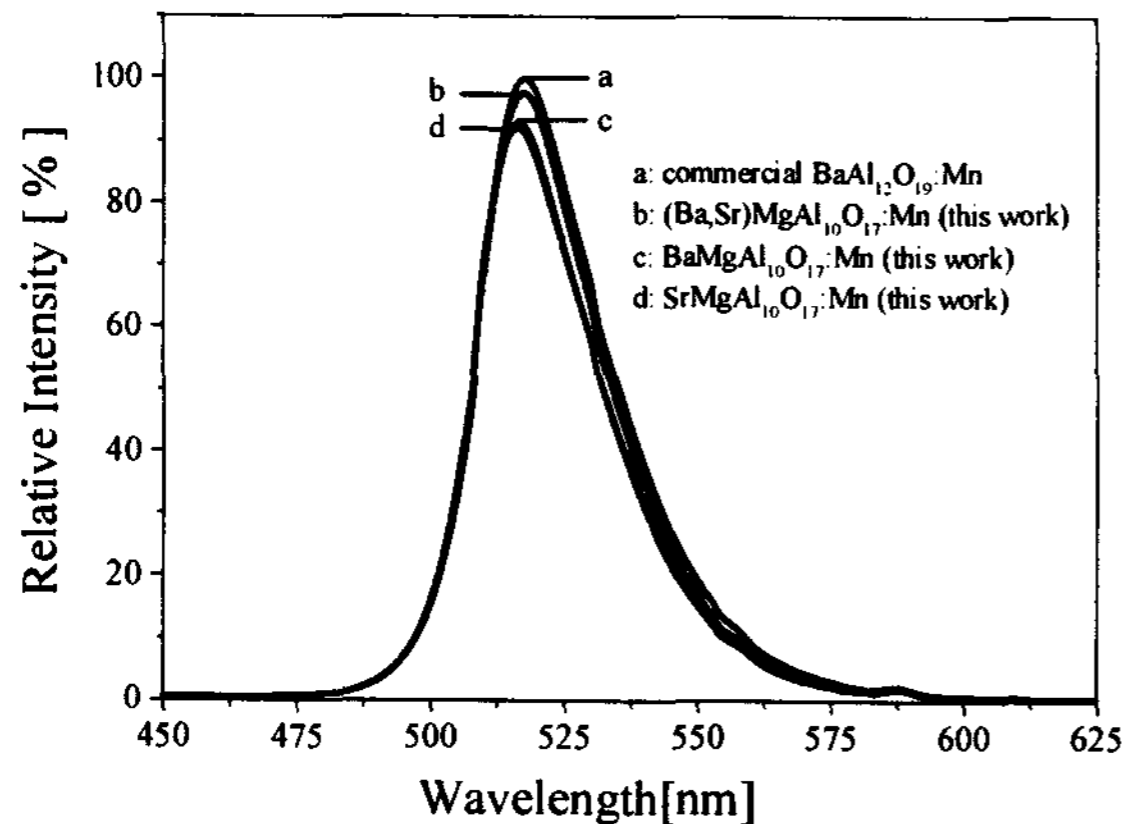


Figure 5 Emission spectra of commercial barium aluminate, $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$, $(\text{Ba,Sr})\text{MgAl}_{10}\text{O}_{17}:\text{Mn}$, and $\text{SrMgAl}_{10}\text{O}_{17}:\text{Mn}$ under VUV excitation (147 nm).

4. Conclusions

The spherical $\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$ green phosphor particles were prepared by spray pyrolysis. The optimal Mn concentration was 10 at. %. It was found that substituting Sr in the Ba sites enhances the luminescence intensity which was about 14 % improved in comparison with the BAM:Mn particles. The highest photoluminescence intensity was obtained when 40 at. % of Ba was replaced with Sr. The optimized $(\text{Ba,Sr})\text{MgAl}_{10}\text{O}_{17}:\text{Mn}$ green phosphor prepared by the spray pyrolysis is expected to be successfully used for the fabrication of high-resolution PDPs instead of the commercial one prepared by the solid-state reaction.

Table 1 Optical properties of green phos 1

	λ_{max} (nm)	Peak intensity (%)	Brightness (%)
$\text{Zn}_2\text{SiO}_4:\text{Mn}$ (commercial)	530	100	100
BAO (Commercial)	517	151	94
$\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$	518	112	102
$\text{BaMgAl}_{10}\text{O}_{17}:\text{Mn}$	517	130	87
$(\text{Ba/Sr})\text{MgAl}_{10}\text{O}_{17}:\text{Mn}$	517	148	90

5. Acknowledgments

This work was supported from Information Display R & D Center, one of the 21st Century frontier R & D Program funded by the Ministry of Science and Technology of Korea.

6. References

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