Eu^{2+} Activated Green Phosphor $Ba_2CaMgSi_2O_8$:Eu^{2+}

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Abstract

In this paper, we report Eu^{2+} activated green phosphor $Ba_2CaMgSi_2O_8$:Eu^{2+}. This phosphor absorbs ultroviolet radiation and emits a green visible light. The phosphors were synthesized by conventional solid state reaction method. Reagent grade $BaCO_3$, $CaCO_3$, MgO, SiO_2, Eu_2O_3 were used as raw materials. The raw materials were mixed thoroughly with an appropriate amount of ethanol in an agate mortar and then dried at 90 ℃ for 2 hours. The mixture was sintered at 900 ℃ for 2 hours and reheated at the mild reducing atmosphere 5% H_2 gas mixed with 95% N_2 gas at about 900 ℃ to 1250 ℃ for 2 hours. The photoluminescence spectra of the phosphor powders were measured by a fluorescent spectrophotometer. The crystal structure of phosphor powders were investigated by X-ray diffractometer .

1. Introduction

In recent years, high efficiency luminescent materials under electron, blue visible light, and UV radiation have been drawn interest. The stability of oxide phosphors in high vacuum and the absence of corrosive gas emission from the oxide phosphors under electron bombardment offer advantages over commonly used sulfide phosphors. So various types of multicomponent oxide phosphors have been widely studied for using in flat panel display technologies, especially in FED and LED. We have studied Eu^{2+} activated green phosphor $Ba_2CaMgSi_2O_8$: $Eu^{2+}$for the application to the UV LED of long wavelength range (380~400nm). The luminescence intensity as well as the excitation wavelength for UV-LED (380~400nm) applications could be improved by modification of composition and heattreatment compared to the previously reported oxide green phosphors, $Ba_2MgSi_2O_7$:Eu^{2+} and $Ba_2Si_1O_4$:Eu^{2+}.

2. Experimental

The phosphors were synthesized by conventional solid state reaction method. The high purity $BaCO_3$, $CaCO_3$, MgO, SiO_2, Eu_2O_3 were used as raw materials. According to the nominal composition $Ba_{3-x-y}A_yMgSi_2O_8$:Eu_x (A = Ca and Sr, $0 < x < 1$, , $0 < y \leq 1$) the raw materials were weighed and mixed thoroughly with an appropriate amount of ethanol in an agate mortar and then dried at 90℃ for 2 hours. The mixture was sintered at 900℃ for 2 hours and reheated at the mild reducing atmosphere 5% H_2 gas mixed with 95% N_2 gas at 1200℃ for 2 hours. The photoluminescence spectra of the phosphor powders were measured by a fluorescent spectrophotometer. The crystal phases of the oxide phosphor samples were characterized using X'Pert X-ray diffractometer (Phillips Co.).

3. Results and discussion

The excitation and emission spectra with the variation of Eu contents are shown in Fig.1 and Fig.2

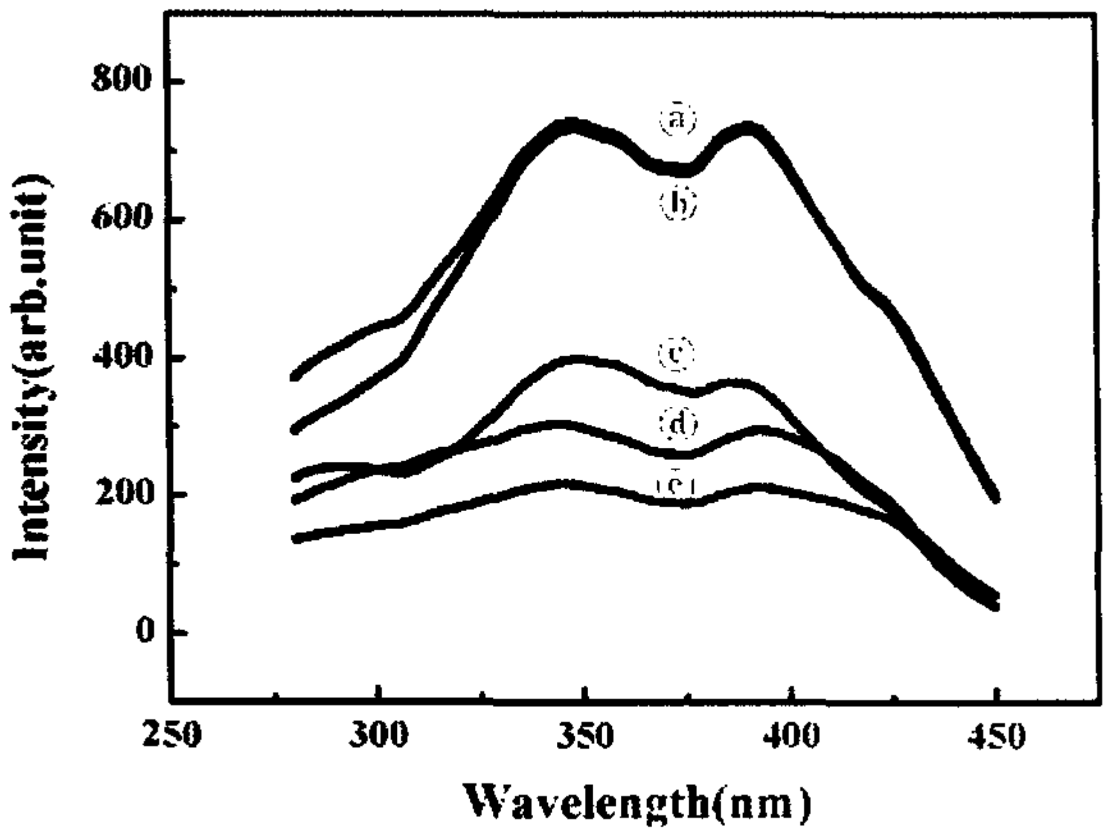

Fig. 1. PL excitation spectra of $Ba_{3-x-y}A_yMgSi_2O_8$:Eu_x (a: x=0.05, b: x=0.1, c: x=0.005, d: x=0.2, e: x=0.4 , y=1)

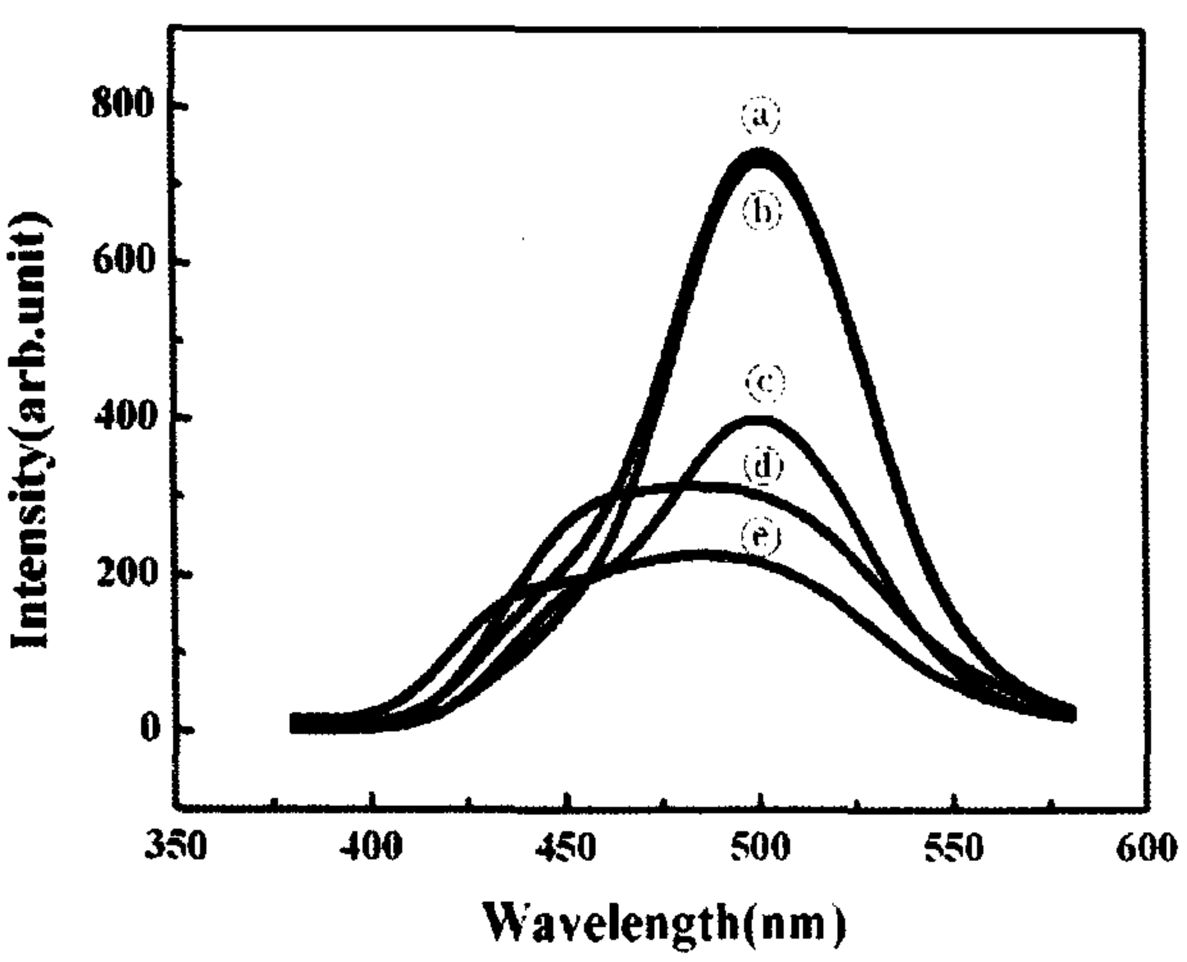

Fig. 2. PL emission spectra of $Ba_{3-x-y}A_yMgSi_2O_8$:Eu_x (a: x=0.05, b: x=0.1, c: x=0.005, d: x=0.2, e: x=0.4 , y=1)

As can be seen in Fig.2 the emission intensity of $Ba_{2-x}CaMgSi_2O_8$:Eu_x varied with the europium concentration. The main peak positions are in the range of 485nm to 505nm. The main emission peak widens from green (505nm) to blue-side. The relative intensity of the blue emission (about 450nm) increases with the increase of Eu contents. It reaches a maximum emission intensity at x=0.1 and then diminishes with increasing Eu content. The doping of Eu^{2+} in $Ba_2CaMgSi_2O_8$ host material results in excellent green emission. The PL spectra with the variation of annealing conditions about $Ba_{1.9}CaMgSi_2O_8$:$Eu_{0.1}$ are shown in Fig.3 and Fig.4.

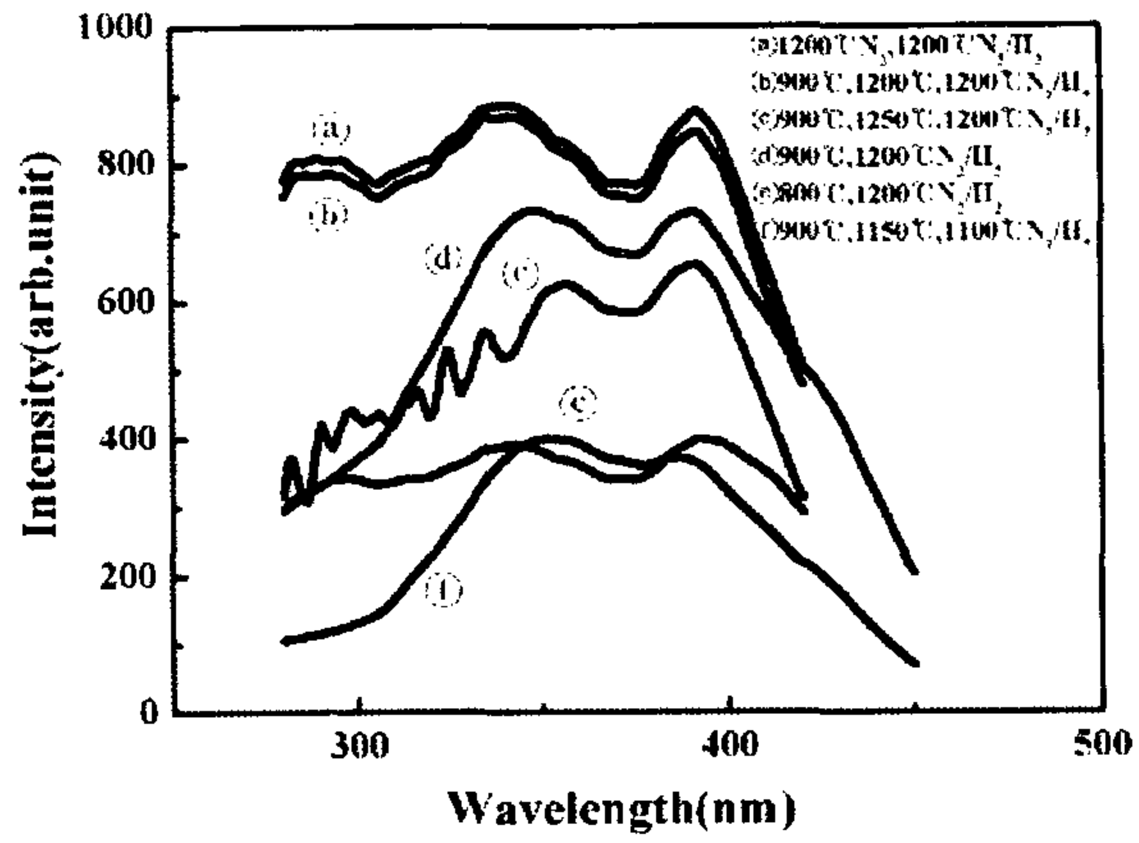

Fig. 3. PL excitation of $Ba_{1.9}CaMgSi_2O_8$:$Eu_{0.1}$

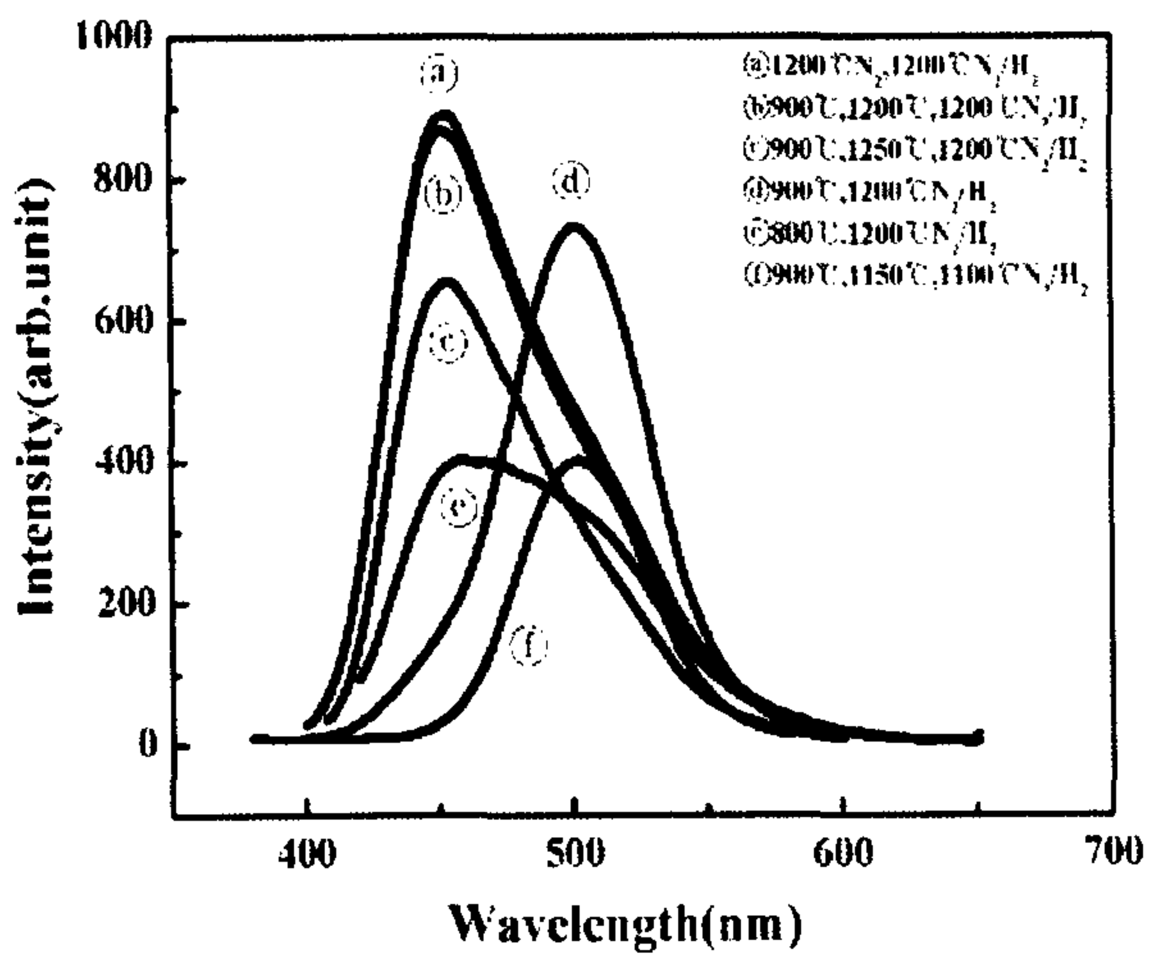

Fig. 4. PL emission spectra of $Ba_{1.9}CaMgSi_2O_8$:$Eu_{0.1}$

As can be seen in Fig. 3 the excitation spectra of the phosphors are similar in wavelength aspect but the emission main peak position and intensities vary greatly with heat-treatment conditions as in Fig. 4. The samples heat-treated in air at high temperature (1200℃, 1250℃) before the reducing treatment (N_2/H_2) at 1200℃ show blue-white emission. On the while the samples directly reduced under N_2/H_2 atmosphere after heat-treatment at 800~900℃ show green-white emission.

The X-ray diffraction patterns of $Ba_{1.95}CaMgSi_2O_8$:$Eu_{0.05}$,

$Ba_{1.9}CaMgSi_2O_8$:$Eu_{0.1}$, $Ba_{1.4}CaMgSi_2O_8$:$Eu_{0.6}$ samples fired at 900℃ with ambient air and reheated at 1200℃ with 5% H_2 gas mixture are shown in Fig.5. The XRD patterns of $Ba_3MgSi_2O_8$ (JCPDS # 10-74), $BaMg_2Si_2O_7$ (JCPDS # 10-44), $BaMgSiO_4$ (JCPDS # 16-573), $Ba_2MgSi_2O_7$ (JCPDS # 36-375), and $Ca_2BaMgSi_2O_8$ (JCPDS # 31-129) have been tested for comparison with the XRD patterns of Fig. 5. The some diffraction peaks of XRD patterns of our samples coincide from those of $Ca_2BaMgSi_2O_8$ (JCPDS # 31-129) type phase. But, the major phase of the highest intensity peak could not be identified.

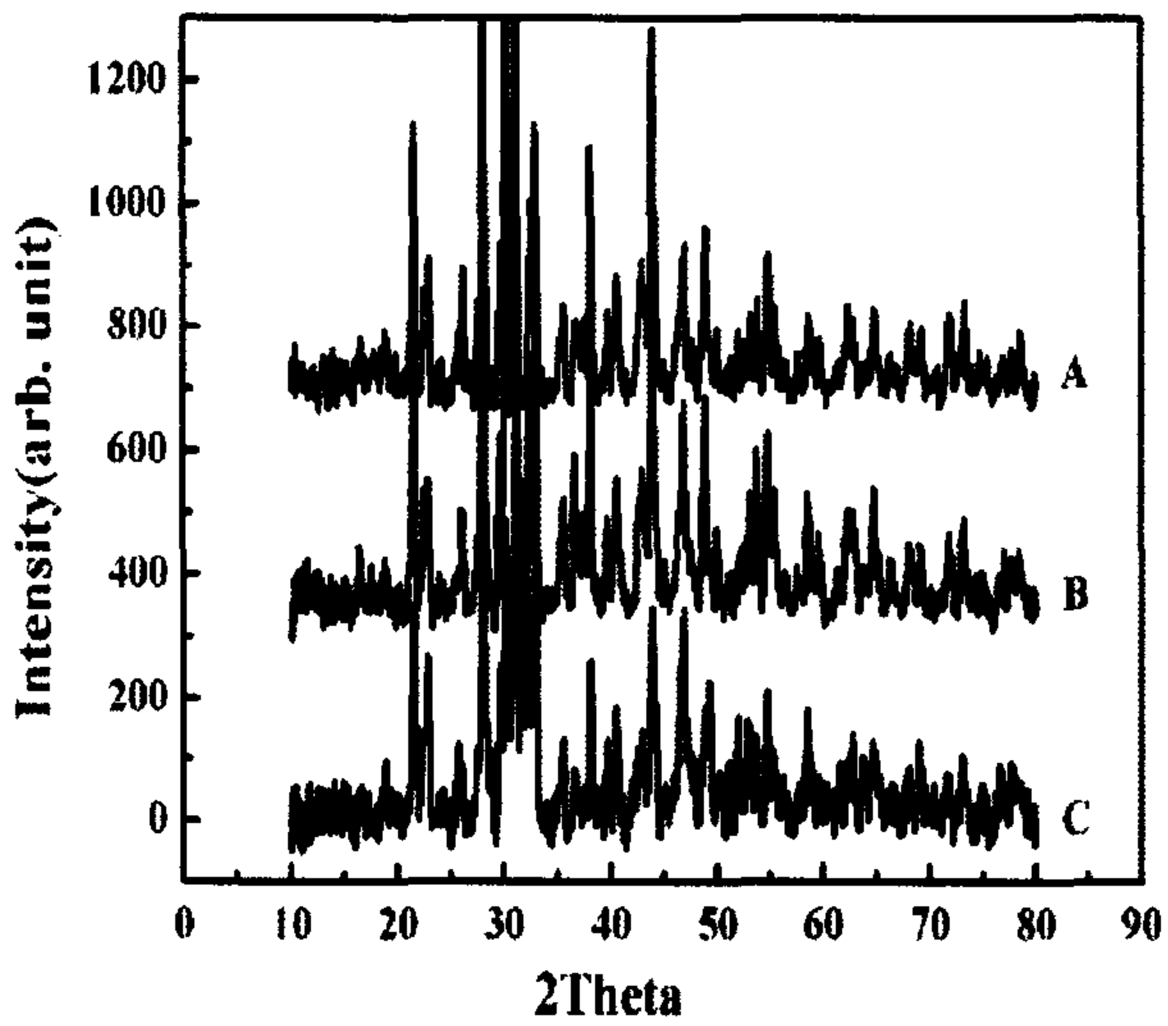

Fig. 5. The XRD patterns of $Ba_{3-x-y}A_yMgSi_2O_8$:Eu_x

(A: x=0.05, B: x=0.1, C: x=0.4 , y=1)

4. Conculsion

Green phosphors developed in this study can be applied in the light-emitting devices, especially UV-LED of 390-405nm range. The Eu^{2+} activated green phosphor of $Ba_2CaMgSi_2O_8$:Eu^{2+} have excellent photoluminescence properties. Especially, Eu-doped green phosphors are excited by 390nm ~ 405nm UV light and emit 500nm green light. The wavelength of the green light can be controlled by the amount of the Eu-doping and heat-treatment conditions. Compare to the previously reported oxide green phosphors, $Ba_2MgSi_2O_7$:Eu^{2+} and $Ba_2Si_1O_4$:Eu^{2+} the luminescence intensity of phosphors have been greatly improved. The present phosphors are composed of $Ca_2BaMgSi_2O_8$ (JCPDS # 31-129) type phase and other unidentified major phase.

5. Acknowledgements

This study was financially supported by Hoseo SERC(RRC supported by KOSEF).

6. References

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