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Photoluminescence Characteristics of $Sr_2SiO_4{:}Pb^{2+}$ Nano Phosphor by a Combustion Method

JONG-MIN KIM,[1] SANG-JOON PARK,[2] HYON-HEE YOON,[2] KYUNG-HWAN KIM,[1] AND HYUNG-WOOK CHOI[1]

[1]Department of Electrical Engineering, Kyungwon University, Sujeong-gu, Seongnam, Gyeonggi-do, Korea
[2]Department of Chemical Engineering, Kyungwon University, Sujeong-gu, Seongnam, Gyeonggi-do, Korea

In this study, lead doped strontium silicate ($Sr_2SiO_4{:}Pb^{2+}$) phosphor was synthesized by a combustion method and the luminescent characteristics were investigated. This was done using X-ray diffraction (XRD), a Scanning Electron Microscope (SEM), and by measuring the photoluminescence (PL). At different temperatures, the XRD patterns showed the pure Sr_2SiO_4 phase. The intensity of the XRD peak showed its highest value at 1000°C. The PL intensity was observed to improve with an increasing sintering temperature (up to 1000°C), but a quenching effect appeared at 1200°C. The emission spectrum for a 291 nm excitation showed a single band, which peaked at 425 nm. The SEM images showed that with an increased sintering temperature the phosphor particles became spherical.

Keywords Combustion method; light-emitting diode; phosphor; Sr_2SiO_4

Introduction

Liquid crystal display (LCD) technology has recently attracted an increasing interest in a variety of applications such as personal computer monitors, portable notebooks, mobile phones, car navigation systems, and in viewfinders for digital cameras and video recorders, etc. In particular, large-scale flat panel displays need a high-performance full color LCD that exhibits a high resolution, a uniform surface illuminance and a long life. The backlight module (BLU) is one of the critical components that determine the quality of the LCD, in which the stability of the fluorescence phosphors plays an important role [1]. There has been an increased interest, recently, in the development of white light-emitting diodes (white LEDs) because of their wide application in consumer electronics and solid state lighting [2,3].

Address correspondence to Prof. Hyung-Wook Choi, Department of Electrical Engineering, Kyungwon University, San 65 Bokjeong-dong, Sujeong-gu, SeongNam, Gyeonggi-do, 461-701, Korea (ROK). Tel.: (+82)31-750-5562; Fax: (+82)31-750-5491; E-mail: chw@kyungwon.ac.kr

White LEDs are known to have potential as a solid-state lighting source and in LCD backlighting applications because of their low power consumption and long lifetime. Recently, various different structures of white LEDs were studied in order to determine the optimal white light suitable for the human eye. At first the combination of the three fundamental colors, red, green and blue LEDs, which act as a multiple-wavelength LED, was investigated with the aim of producing white LEDs. However, this method required a complicated electrical design to control the light intensity and uniformity. This combination also had different light output degradation rates and a low emission efficiency in regards to the red-emitting InGaN compounds [4].

In recent years, LEDs utilyzing phosphor to generate white light became the present focus of research in the lighting industry. White LEDs could therefore be applied to an LCD BLU. Generally, blue phosphors are very important in the white LED strategy, via the combination of an ultraviolet light-emitting diode (UV-LED) with blue, green and red phosphors.

In this work before synthesis the phosphors, we take the silicate for the materials. Silicate materials can be applied to various technique such as display and so on. Silicate-base phosphor is one of the important thing among the rest. It has stability under high irradiation power and high temperature. Besides, it can be produced at relatively low cost with the other materials. The aim of this work was to investigate the effect of activator on the structural and luminescent properties of blue-emitting Strontoum-Silicate (Sr_2Sio_4) phosphor with Lead doped. And for a small spherical particles with smooth and round surfaces paticales, sr2sio4 phosphors were synthesized by combustion method.

The sol-gel combustion process is one of non-alkoxide sol-gel method, which is an efficient method for the preparation of nanocrystalline materials. But its reaction is too violent to control, and the powders are difficult to collect. Therefore, our synthesis has the advantages of short process control and low reaction temperature as compared with the former combustion method, and has the ability to synthesize materials with high purity better homogeneity and high surface area in one single step [5].

In this study, Sr_2SiO_4:Pb^{2+} phosphor precursors were synthesized using the combustion method.

Experiment

Synthesis

Lead doped strontium silicate (Sr_2SiO_4:Pb^{2+}) phosphor was synthesized by a combustion method and its luminescent characteristics were investigated. A detailed description of the method can be found in the original works of Pail *et al.* [6,7]. $Sr(NO_3)_2$ (99.995%, Aldrich), SiO_2 (99.9%, Aldrich), $Pb(NO_3)_2$ (99.999%, Aldrich) were used as starting materials. The $Sr(NO_3)_2$ and SiO_2 were mixed together at a 2:1 mole ratio then lead nitrate and distilled water were added. Urea was used as the fuel and ammonium nitrate as an oxidizer. The parameters were measured and are shown in Table 1.

A flowchart for the preparation of the phosphor powders is described in Figure 1. The urea and ammonium nitrate solution was heated to 80°C and continuously stirred using a magnetic bar stirrer. Next, this metal solution was dropped into the

Table 1. The mole ratio of the $Sr_2SiO_4:Pb^{2+}$ used by the combustion method at various temperatures

Materials	Mole ratio					Temp.
$Sr_{2-x}SiO_4:Pb_x$	Concentration of Pb^{2+}					600°C
	x = 0.01	x = 0.02	x = 0.05	x = 0.07	x = 0.1	800°C
Urea	20					1000°C
Ammonium	20					1200°C

fuel, after which it was heated for 30 minutes at 80°C. The solution was then transferred to a pre-heated furnace set at 500°C. Different samples of the mixture were then sintered in the furnace for 3 hours at temperatures between 600 and 1200°C.

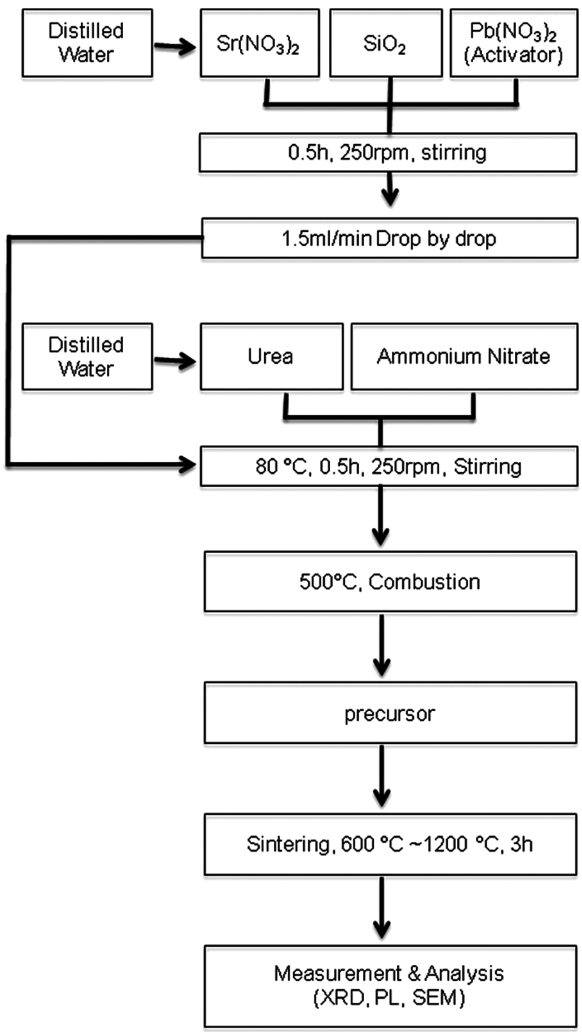

Figure 1. The flowchart for the preparation of the phosphor powders.

Measurements

The crystalline development of the resulting samples was checked by X-ray diffraction (XRD, model D/MAX-2200) using CuKα-radiation in the range of $2\theta = 20 \sim 80°$. The measurement of the photoluminescence (PL) spectra was carried out with a 150 W Xe lamp (spectrofluorometer, FP-6200, JASCO). The morphology and the size of the prepared particles were investigated with a field-emission scanning electron microscope (FE-SEM, model S-4700, HITACHI).

Results and Discussion

Figure 2 shows the XRD patterns of the Sr_2SiO_4:Pb^{2+} sintered at different temperatures. At the different temperatures the XRD patterns show the pure Sr_2SiO_4 phase. In this structure, Sr^{2+} ions locate at two kinds of unequivalent lattice sites, and their coordination numbers are 9 and 10, respectively. O^{2+} ions locate at three kinds of unequivalent lattice sites, which space group is D_{2h}, and Si^{4+} ions locate at the center of the oxygen tetrahedron. It can be seen that the structure of Sr_2SiO_4 has the higher symmetry. When the Pb is introduced into the Sr_2SiO_4 structure, it takes the place of the Sr^{2+}. In case of precursor were observed not only the Sr_2SiO_4 phase, but also the impurities peaks due to the have not synthesized. When the temperature was at 1000°C, the diffraction peaks became sharper and stronger. The Sr_2SiO_4 phase crystallized with results that are in good agreement with the JCPDS diffraction file 38-0271. However at 1200°C the Sr_2SiO_4 peaks were markedly weakened.

The XRD patterns of the Sr_2SiO_4:Pb^{2+} powders at various concentrations of Pb^{2+} are shown in Figure 3. The added amount of Pb^{2+} ranged from 0.01 to 0.1 mole. The Sr_2SiO_4 peaks, with the (103) main peak, appeared at all concentrations of the Pb in the XRD patterns.

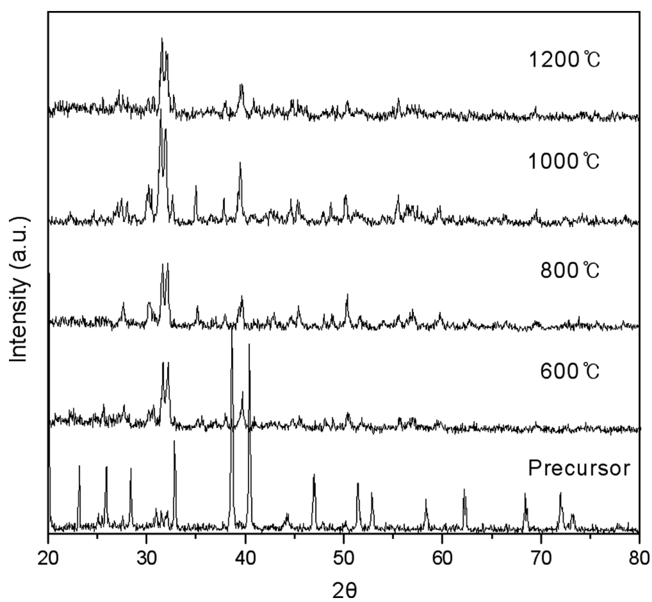

Figure 2. The XRD patterns of the Sr_2SiO_4:Pb^{2+} sintered at different temperatures.

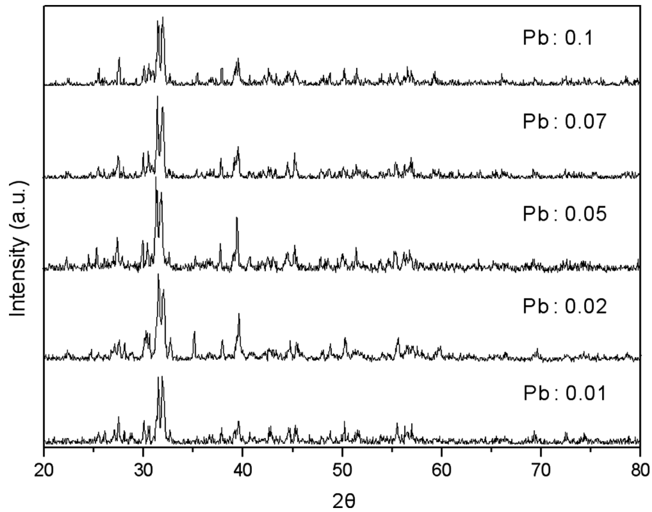

Figure 3. The XRD patterns of the $Sr_2SiO_4:Pb^{2+}$ powders at various concentrations of Pb^{2+}.

Figure 4 shows SEM images of the $Sr_2SiO_4:Pb^{2+}$ sintered at different temperatures. The surface morphologies of the phosphor at 600°C had a rough shape. With an increased sintering temperature the phosphor particles became spherical, which occurs because the particles condense at high temperatures [7]. But at 1200°C, agglomerates formed particle was observed. Over 1200°C the particle became

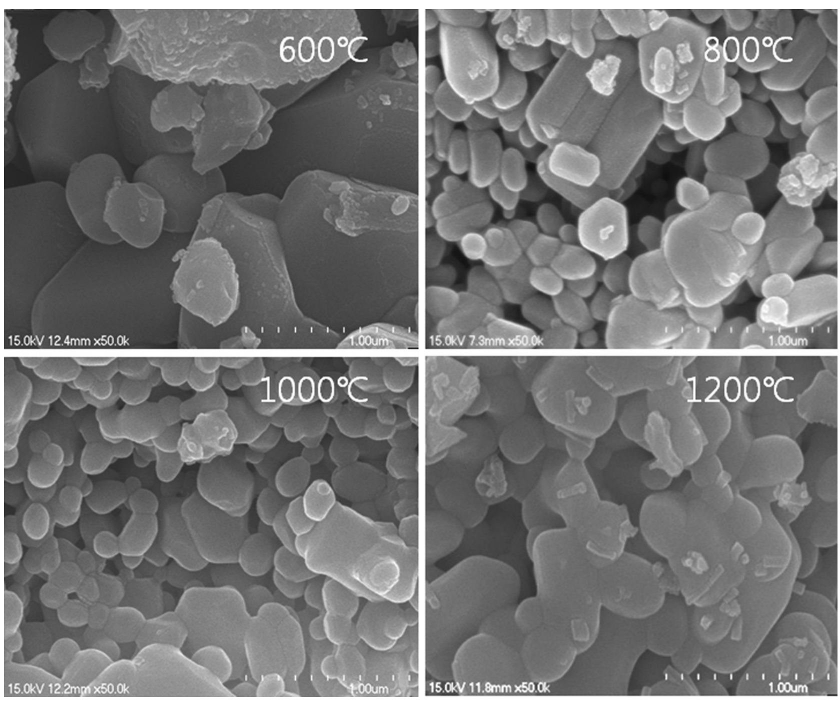

Figure 4. The SEM images of the $Sr_2SiO_4:Pb^{2+}$ sintered at different temperatures.

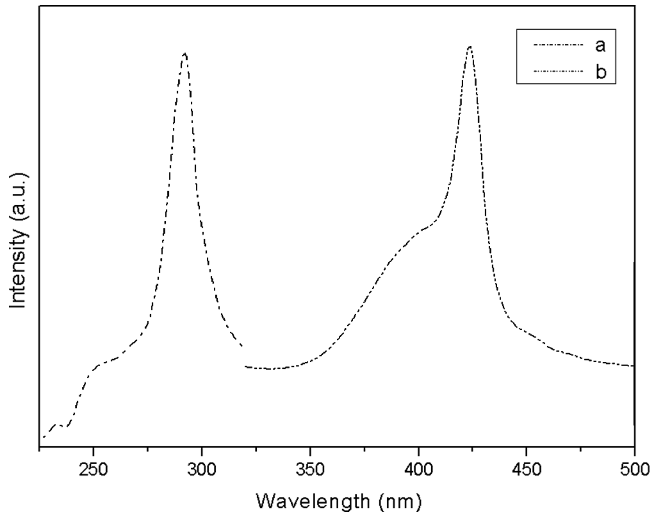

Figure 5. (a) The excitation and (b) emission spectra of the Sr_2SiO_4:Pb^{2+}.

cohesion and rough due to the high sintering temperature. At this point, we estimated that the characteristics of the phosphor powders had been improved by increasing the sintering temperature; however, defects form in the phosphor when it is heated beyond a critical temperature.

The excitation (Fig. 5a) and the emission (Fig. 5b) spectra of the precursor are shown in Figure 5. Figure 5a shows the excitation at a 291 nm emission and the emission for a 425 nm excitation is shown in Figure 5b.

Figures 6 and 7 show the PL emission spectra of the Sr_2SiO_4:Pb^{2+} heated at different temperatures and with various concentrations of Pb^{2+}. The luminescence of Pb^{2+} in host materials is diverse. It can be described by the $^1S_0 \rightarrow {}^3P_{0,1}$ transition

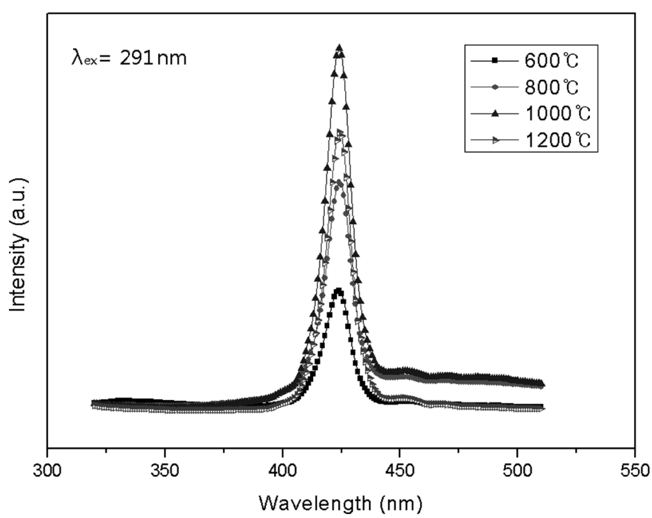

Figure 6. The PL emission spectra of the Sr_2SiO_4:Pb^{2+} heated at different temperatures.

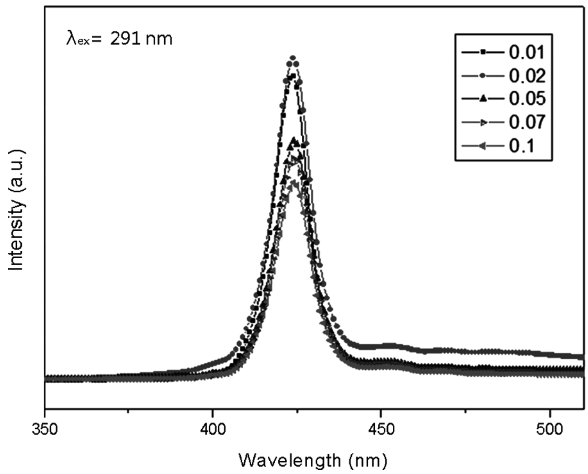

Figure 7. The PL emission spectra of the Sr_2SiO_4:Pb^{2+} powders at various concentrations of Pb^{2+}.

which originates from $(6s)^2$–$(6s)^1(6p)^2$ interconfigurational transition. Typically at room temperature, emission is observed from the $^3P_1$–$^1S_0$ transition [9], although at low temperatures the highly forbidden $^3P_0 \rightarrow {}^1S_0$ emission is also observed [10]. The maximum emission band of Sr_2SiO_4:Pb^{2+} was observed at 425 nm corresponding to the $^3P_1$ excited state level to the $^1S_0$ ground state transition upon excitation with 291 nm. The PL emission intensity of the phosphors with the starting composition of Sr_2SiO_4:Pb^{2+} increased with an increasing firing temperature up to 1000°C, due to an improved crystallinity as shown in Figure 6. However, at higher firing temperatures than 1000°C a quenching effect appeared. The intensity of the emission spectrum of the Sr_2SiO_4:Pb^{2+} phosphor was found to decrease when sintered at 1200°C. In Figure 7 the emission intensity increased with an increasing Pb concentration up to $x = 0.02$, and then decreased at $x = 0.05$ due to the concentration quenching. The best results for the Pb concentration (0.02 mol of Pb) and the annealing temperature (1000°C) are indicated in Figures 6 and 7.

Conclusions

In this study, Sr_2SiO_4:Pb^{2+} phosphors were synthesized via a combustion method. The effects of the sintering temperature and the Pb^{2+} concentration on the changes in structure and the luminescent properties were investigated. The PL intensity was observed to improve with an increasing sintering temperature (up to 1000°C), but a quenching effect appeared at 1200°C. The emission spectrum for the 291 nm excitation showed a single band which peaked at 425 nm, a blue emission. Doping with the Pb^{2+} ion resulted in a new UV-excited luminescent material with a single emission band, centered in the blue region with a wavelength of 425 nm. The emission peak intensities change depending on the concentration of Pb^{2+} in the host lattice and reached a maximum at 0.02 mol%. The Sr_2SiO_4 peaks, with the (103) main peak, appeared at all the sintering temperatures in the XRD patterns. The intensity of the XRD peak showed its highest value at 1000°C. Through SEM images it was shown that with an increasing sintering temperature phosphor particles became spherical at 1000°C. Nanosized powder of Sr_2SiO_4:Pb^{2+} with a spherical-like shape and smooth surfaces was successfully synthesized using combustion method. Due to its excellent color purity and emission intensity, this phosphor can be considered for a promising candidate for dispaly applications. These results indicate that Sr_2SiO_4:Pb^{2+} phosphors have a wide application as the blue emission phosphor for white LEDs and can be readily applied to LCD BLUs.

Acknowledgments

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