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Synthesis and Luminescent Properties of Sr_2SiO_4 Phosphors

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Abstract. $Sr_2SiO_4:Eu^{2+}$ phosphors and $Sr_2SiO_4:Eu^{2+}$, Dy^{3+} persistent phosphors were synthesized by solid-state reaction method at 1300℃ using $SrCO_3$, SiO_2(silica: 3 μ m and fumed silica: 7nm) , Eu_2O_3(0.01 to 0.06 mol% Eu) and Dy_2O_3(0.005 to 0.02 mol% Dy) powders. The amount of the stable β- Sr_2SiO_4 phase had decreased and the amount of the α'-Sr_2SiO_4 increased with the increase of the Eu content. The solid solution of Eu^{2+} ion stabilized α'-Sr_2SiO_4 at room temperature. The emission color of the $Sr_2SiO_4:Eu^{2+}$ products changed from the turquoise blue to yellow with the increase of the Eu content. The maximum emission peak position changed to the higher wavelength with the increase of the Eu content. The emission peak at 490nm(green color) was from β- Sr_2SiO_4 phase and that at 560nm(yellow color) was from α'-Sr_2SiO_4 phase. The change of the phase content in the products affects the color and the emission peak. The emission intensity of the products from fumed silica is stronger than the products from silica. $Sr_{1.98-x}SiO_4:Eu_{0.02}$, Dy_x persistent phosphors products showed the persistent emission for a few minutes with the naked eyes. The behavior was observed from all products. The product from fumed silica at x = 0.01 showed the strong emission for tens of seconds.

1. Introduction

In recent years, phosphors and long persistent phosphors were actively studied. Phosphors can convert light energy into a visible light, and long persistent phosphors store light energy and emit another color-light in dark.

Phosphors were used for Fluorescent Lamp, Plasma Display Panel(PDP), etc. Long persistent phosphors were used for Emergency Plate, Dial Plate, etc. Also, since phosphors showed the variety luminescent color, phosphors were used for white light emitting diodes (LEDs) which were widely studied. White LEDs composed of blue LEDs and yellow emitting phosphors have been developed and are widely applied to lighting systems in these days. As yellow phosphors, $Y_3Al_5O_{12}:Ce^{3+}$($YAG:Ce^{3+}$) and strontium silicate materials are mainly used for white LEDs[1-3].
Since $Sr_2SiO_4:Eu^{2+}$ phosphors have the merits of the stability under high irradiation powers and the durability in the packaging resin, they are also commercially used in white LEDs instead of $YAG:Ce^{3+}$[2,4]. $Sr_2SiO_4:Eu^{2+}$ phosphor has two phases of α'-type(monoclinic) and β-type(orthorhombic). β-Sr_2SiO_4 is stable at room temperature and transforms to α'-Sr_2SiO_4 at 85℃

Recently, $Sr_2SiO_4:Eu^{2+}$, Dy^{3+} have attracted attention since they show high water-resistant and luminescence characteristics[5].

In the present work, the effects of the particle size of raw silica, and the Eu content on the crystal structure and the luminescence properties of Sr_2SiO_4: Eu^{2+} powders were examined. Moreover, the effect of the Dy addition on the crystal structure and the luminescence properties of Sr_2SiO_4: Eu^{2+}, Dy^{3+} persistent phosphors were also examined.

2. Experimental

The Sr_2SiO_4 phosphors and persistent phosphors were synthesized by the general solid-state reaction. $SrCO_3$(Aldrich, 99.9%), Eu_2O_3(Aldrich, 99.5%), Dy_2O_3(Aldrich, 99.9%) and SiO_2(two kinds of silica particles : silica: 3 μm and fumed silica: 7nm) were used as raw materials. These were mixed with small amount of ethanol as the dispersing agent for 1h. The resulting slurry was dried at room temperature. The dried mixtures were heated at 1300℃ in the atmosphere (10%H_2 + 90%N_2) for 3 h.

All products were examined for the phase formation using the X-ray diffractometer (Shimadzu, XRD-6300) with CuKα radiation. The photoluminescence characteristics of the products were evaluated using the USB 4000-UV-Vis fiber optic spectrometer (Ocean optics), using the black-light (peak wave length: 356nm) as an excitation source. After irradiating the black-light for 5 min, the afterglow time was observed by the naked eyes until the products becomes dark. All measurement were carried out at room temperature.

3. Results and discussions

3.1. Characteristics of $Sr_{2-x}SiO_4$:Eu_x phosphors

3.1.1. XRD analysis of the products

XRD patterns of the products with various amounts of Eu, $Sr_{2-x}SiO_4$:Eu_x prepared from silica and fumed silica, are shown in Figures 1 and 2. Both products at x = 0.01 has α'-Sr_2SiO_4 and β-Sr_2SiO_4 phases. However, the products at x = 0.06 has only α'-Sr_2SiO_4 phase. Therefore, the solid solution of Eu^{2+} ion stabilized α'-Sr_2SiO_4 at room temperature.

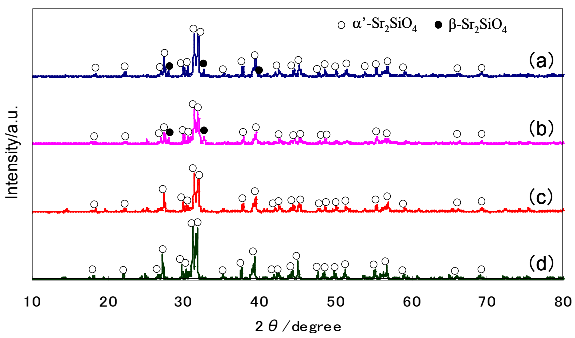

Figure 1. XRD pattern of $Sr_{2-x}SiO_4$:Eu_x using silica : (a) x =0.01, (b) x =0.02, (c) x =0.03, (d) x =0.06.

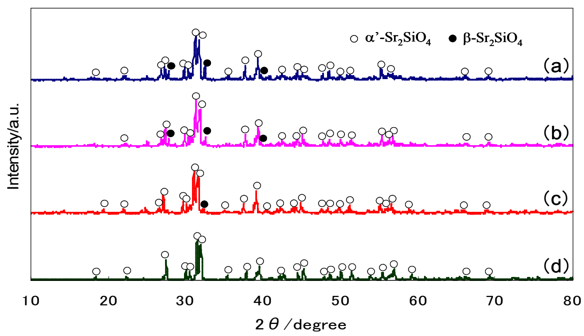

Figure2. XRD pattern of $Sr_{2-x}SiO_4$:Eu_x using fumed silica : (a) x =0.01, (b) x =0.02, (c) x =0.03, (d) x =0.06.

3.1.2. Luminescence characteristics

Emission spectrum of the products are depicted in Figures 3 and 4. The emission spectrum are broad, ranging from 400nm to 650nm.The emission color of the products changed from the turquoise blue to yellow with the increase of the Eu content. The maximum emission peak position changed to the higher wavelength with the increase of the Eu content. The emission peak at around 490nm(green color) was from β-Sr_2SiO_4 phase and that at around 560nm(yellow color) was from α'-Sr_2SiO_4 phase. The change of the phase content in the products affects the color and the emission peak. The emission intensity of the products from both silica at x = 0.01 becomes maximum value. The emission intensity of the products from fumed silica is stronger than the products from silica.

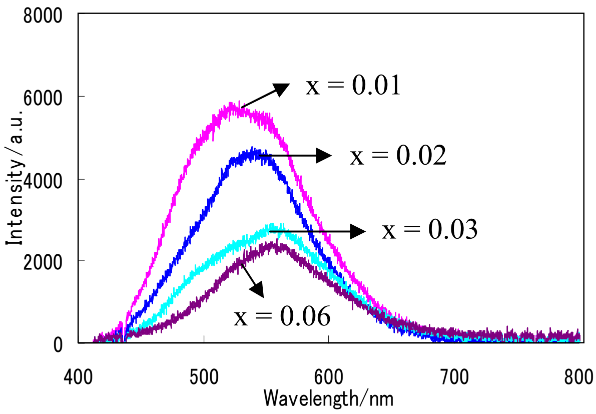

Figure 3. Emission spectrum of $Sr_{2-x}SiO_4$:Eu_x using silica : (A) x =0.01, (B) x =0.02, (C) x =0.03, (D) x =0.06.

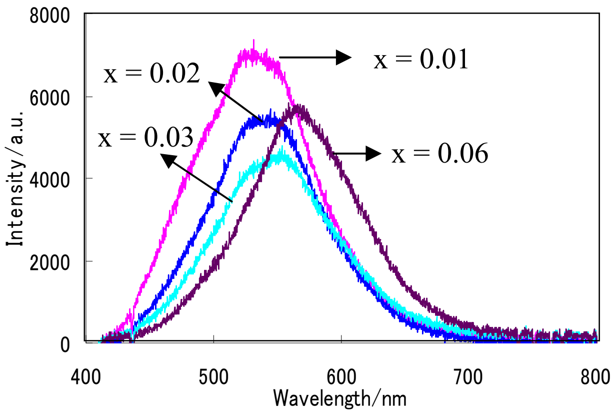

Figure 4. Emission spectrum of $Sr_{2-x}SiO_4$:Eu_x using fumed silica : (A) x =0.01, (B) x =0.02, (C) x =0.03, (D) x =0.06.

3.2. Characteristics of $Sr_{1.98-x}SiO_4$:$Eu_{0.02}$, Dy_x persistent phosphors

3.2.1. XRD analysis of the products

XRD patterns of the products with various amounts of Dy, $Sr_{1.98-x}SiO_4$:$Eu_{0.02}$, Dy_x prepared from silica and fumed silica, are shown in Figures 5 and 6. The products from fumed silica at x = 0.01 has only α'-Sr_2SiO_4 phase. On the other hand, the products from silica at x = 0.02 has only α'-Sr_2SiO_4 phase. Therefore, the solid solution of Dy^{3+} ion stabilized α'-Sr_2SiO_4 at room temperature. But, the optimum value of the Dy contents is different from silica particle size.

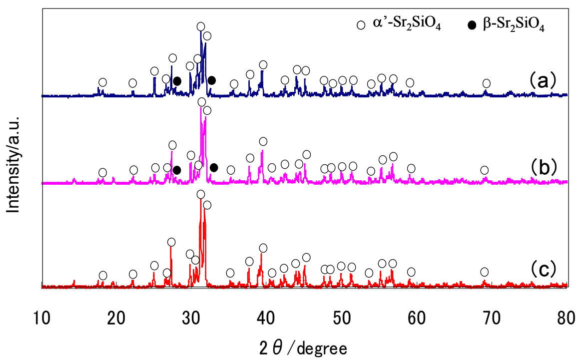

Figure 5. XRD pattern of $Sr_{1.98-x}SiO_4$:$Eu_{0.02}$, using silica : (a) x =0.005, (b) x =0.01, (c) x =0.02.

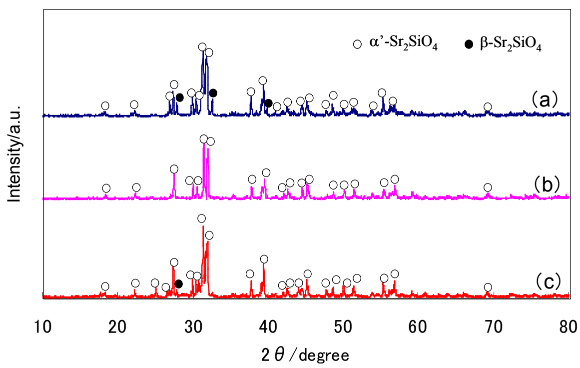

Figure 6. XRD pattern of $Sr_{1.98-x}SiO_4$:$Eu_{0.02}$, Dy_x using fumed silica : (a) x =0.005, (b) x =0.01, (c) x =0.02.

3.2.2. Luminescence characteristics

Emission spectrum of the products are depicted in Figures 7 and 8. The emission color of the products from fumed silica at x = 0.01 was yellow. However, the emission color of the other all products were lime green. The emission intensity of products from both silica at x = 0.01 becomes maximum value. The emission intensity of the products from fumed silica is stronger than the products from silica. The persistent emission was observed for a few minutes with the naked eyes. The behavior was observed from all products. The product from fumed silica at x = 0.01 showed the strong emission for tens of seconds.

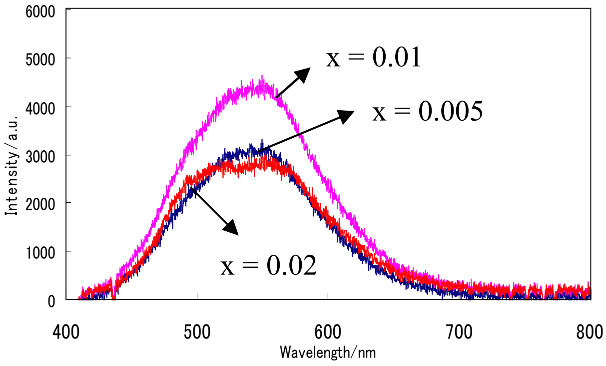

Figure 7. Emission spectrum of $Sr_{1.98-x}SiO_4$: $Eu_{0.02}$, Dy_x using silica : (A) x =0.005, (B) x =0.01, (C) x =0.02.

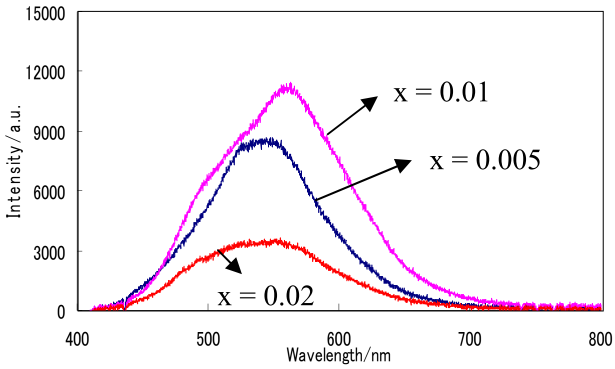

Figure 8. Emission spectrum of $Sr_{1.98-x}SiO_4$: $Eu_{0.02}$, Dy_x using fumed silica : (A) x =0.005, (B) x =0.01, (C) x =0.02.

4. Conclusions

The $Sr_{2-x}SiO_4$:Eu_x phosphors and $Sr_{1.98-x}SiO_4$:$Eu_{0.02}$, Dy_x persistent phosphors showed two phases, α'-type and β- type, and the solid solution of Eu^{2+} and Dy^{3+} ions stabilized α'-Sr_2SiO_4 at room temperature. Both products at x = 0.01 showed the strongest emission intensity. The emission intensity of the products from fumed silica is stronger than the products from silica. $Sr_{1.98-x}SiO_4$:$Eu_{0.02}$, Dy_x persistent phosphors showed the persistent emission for a few minutes with the naked eyes. The behavior was observed from all products. The product from fumed silica at x = 0.01 showed the strong emission for tens of seconds.

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