

Effect of preparation temperature on the formation of Sr_2CeO_4 phosphor particles in the spray pyrolysis

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Abstract— Sr_2CeO_4 phosphor particles were prepared by spray pyrolysis at various preparation temperatures. The effect of preparation temperatures on the morphology, crystal structure and photoluminescence characteristics of the post-treated Sr_2CeO_4 phosphor particles was studied. Phase pure Sr_2CeO_4 phosphor particles were not produced by spray pyrolysis without post-treatment. The optimum post-treatment temperature to produce the Sr_2CeO_4 phosphor particles with high photoluminescence intensity was 1,000 °C in spray pyrolysis. The spherical morphology of the as-prepared particles obtained at high preparation temperatures above 1,400 °C had maintained after post-treatment at 1,000 °C. The relative photoluminescence intensities of the Sr_2CeO_4 phosphor particles varied with the preparation temperatures in the spray pyrolysis. The as-prepared particles obtained by spray pyrolysis at preparation temperatures below 1,400 °C converted into phase pure Sr_2CeO_4 phosphor particles after post-treatment at 1,000 °C. The optimum preparation temperature of the as-prepared particles was 1,400 °C to produce the Sr_2CeO_4 phosphor particles with spherical shape and high photoluminescence intensity in the spray pyrolysis.

Key words: Phosphor, Spray Pyrolysis, Ceramic

INTRODUCTION

Sr_2CeO_4 phosphor emitting blue-white light has efficient luminescence characteristics under electron beam and ultraviolet light [Danielson et al., 1998; Tang et al., 2002; Chen et al., 2004; Jiang et al., 1999; Yu et al., 2003; Masui et al., 2003]. Application of Sr_2CeO_4 particles as a phosphor in displays and lamps requires good morphology as well as high luminescence characteristics. The mean size and morphology of phosphor particles affects the characteristics of phosphor material in application to displays and lamps. Sr_2CeO_4 phosphor particles have been prepared by many phosphor preparation processes because the characteristics of phosphor particles are strongly affected by the synthetic process. Especially, liquid solution methods including citrate-gel, co-precipitation and hydrothermal methods were applied to the preparation of fine sized Sr_2CeO_4 phosphor particles [Jiang et al., 1999; Yu et al., 2003; Masui et al., 2003; Kholmam et al., 2004]. Spray pyrolysis is one of the promising processes for the preparation of improved phosphor particles with spherical shape, fine size, narrow size distribution, and non-aggregation characteristics [Kang et al., 2000, 2002, 2003; Roh et al., 2003; Shimomura et al., 2004; Kang and Choi, 2002; Jung et al., 2004]. Sr_2CeO_4 phosphor particles were also prepared by spray pyrolysis process, in which the prepared phosphor particles had spherical shape and fine size [Kang and Choi, 2002].

In this work, Sr_2CeO_4 phosphor particles were prepared by spray pyrolysis at various preparation temperatures. In spray pyrolysis, the effect of preparation temperatures on the morphology, crystal structure and photoluminescence characteristics of the post-treated Sr_2CeO_4 phosphor particles was studied.

EXPERIMENTAL

The solution of salts was atomized with six ultrasonic spray generators with 1.7 MHz resonator and introduced into a hot reaction column, where the droplets were dried, decomposed, and/or crystallized in dispersed phase. The length and inner diameter of the alumina reactor were 1,000 and 25 mm, respectively. The flow rate of carrier gas was 10 l/min. The spray solutions were obtained by adding strontium and cerium nitrate precursors into distilled water. The overall solution concentration was 0.5 M. The preparation temperature in the spray pyrolysis varied from 800 to 1,600 °C. The as-prepared particles were post-treated at temperatures between 700 and 1,200 °C for 3 h in muffle furnace.

The crystal structures of the particles were studied by X-ray diffraction (XRD, RIGAKU, D/MAX-RB) with $\text{Cu-K}\alpha$ radiation ($\lambda=1.5418$). The morphologies of particles were investigated by scanning electron microscopy (SEM). Photoluminescence measurement was performed with a spectrofluorophotometer (SHIMADZU, RF-5301PC) by using a Xe lamp excitation source.

RESULTS AND DISCUSSION

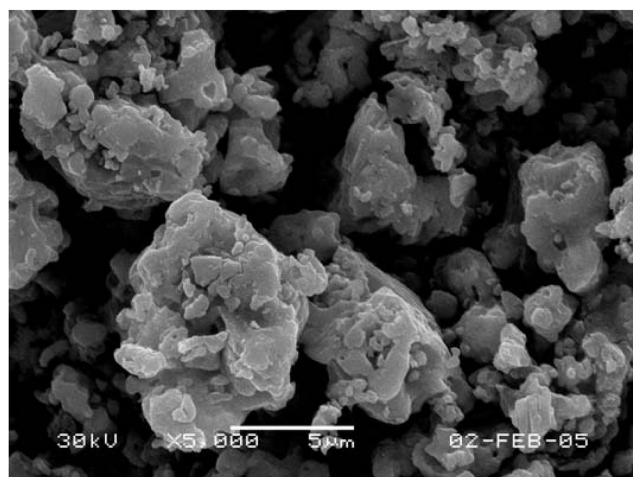
The Sr_2CeO_4 phosphor particles prepared by spray pyrolysis had poor photoluminescence intensities even at high preparation temperature of 1,600 °C. In the aqueous solution methods, phase pure Sr_2CeO_4 phosphor particles with high photoluminescence intensities were obtained at post-treatment temperatures below 1,000 °C [Jiang et al., 1999; Yu et al., 2003; Masui et al., 2003; Kholmam et al., 2004]. In spray pyrolysis, the crystal structure of Sr_2CeO_4 phosphor appeared at a preparation temperature of 1,400 °C. However, phase pure Sr_2CeO_4 phosphor particles were not produced by spray pyrolysis without post-treatment. The short residence time of parti-

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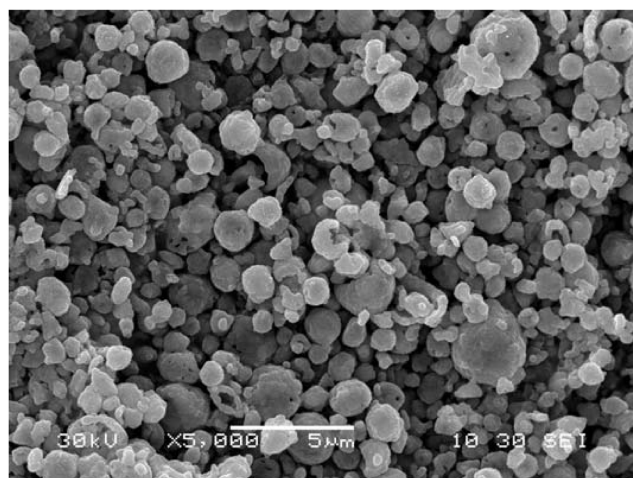
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cles inside the hot wall reactor did not offer enough time for the crystallization of Sr_2CeO_4 phosphor particles in spray pyrolysis. The residence time of particles inside the hot wall reactor was 0.3 sec at a preparation temperature of 1,600 °C. The Sr_2CeO_4 phosphor particles prepared by spray pyrolysis had pure crystal structure and high photoluminescence intensities after post-treatment. The optimum post-treatment temperature to produce the Sr_2CeO_4 phosphor particles with high photoluminescence intensity was 1,000 °C in spray pyrolysis. Phase pure Sr_2CeO_4 phosphor particles were obtained at low post-treatment temperature of 800 °C because of high mixing degree of each component comprising the as-prepared particles. The photoluminescence intensity of the Sr_2CeO_4 phosphor particles post-treated at 800 °C was 80% of that of the phosphor particles post-treated at 1,000 °C.

The as-prepared particles obtained by spray pyrolysis had spherical shape regardless of the preparation temperatures. On the other hand, the morphology of the post-treated Sr_2CeO_4 phosphor particles was affected by the preparation temperatures in the spray pyrolysis. The spherical shape of the as-prepared particles obtained at



(a) 800 °C



(b) 1400 °C

Fig. 1. SEM photographs of post-treated Sr_2CeO_4 phosphor particles.

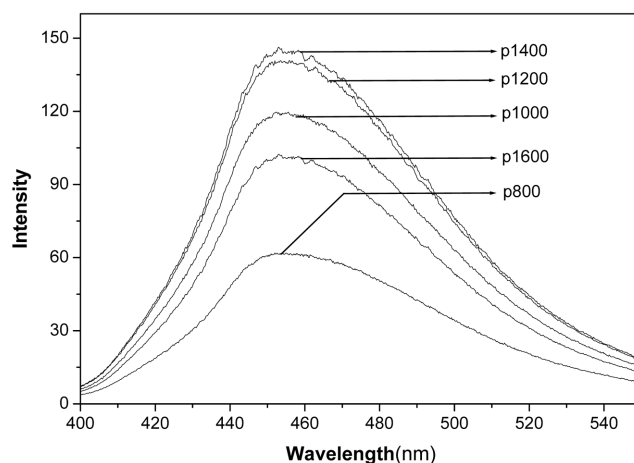


Fig. 2. Emission spectra of Sr_2CeO_4 phosphor particles prepared at different preparation temperatures.

800 °C disappeared after post-treatment at 1,000 °C. Aggregated and non-spherical shape Sr_2CeO_4 phosphor particles were obtained as shown in Fig. 1(a). However, the morphology of the as-prepared particles obtained at high preparation temperatures above 1,400 °C had maintained after post-treatment at 1,000 °C. Fig. 1(b) shows the SEM photograph of the Sr_2CeO_4 phosphor particles, in which the as-prepared particles obtained at 1,400 °C were post-treated at 1,000 °C. The post-treated Sr_2CeO_4 phosphor particles (Fig. 1b) had spherical shape and non-aggregation characteristics.

Fig. 2 shows the emission spectra of the Sr_2CeO_4 phosphor particles, in which the as-prepared particles at different preparation temperatures were post-treated at 1,000 °C. The emission spectra of the Sr_2CeO_4 phosphor particles were measured under excitation wavelength of 285 nm showing the maximum peak intensity in the excitation spectra. The photoluminescence intensities of the Sr_2CeO_4 phosphor particles were affected by the preparation temperatures in the spray pyrolysis. All the phosphor particles had the same emission spectra having a maximum peak intensity at 455 nm. However, the relative photoluminescence intensities of the Sr_2CeO_4 phosphor particles varied with the preparation temperatures in the spray pyrolysis. The Sr_2CeO_4 phosphor particles obtained at the preparation temperature of 800 °C had low photoluminescence intensity after post-treatment at 1,000 °C. The maximum photoluminescence intensity of the post-treated Sr_2CeO_4 phosphor particles was obtained at a preparation temperature of 1,400 °C. The Sr_2CeO_4 phosphor particles obtained at a preparation temperature of 1,600 °C had lower photoluminescence intensity than that of the phosphor particles obtained at 1,400 °C after post-treatment.

XRD spectra of the Sr_2CeO_4 phosphor particles were investigated to explain the effect of the preparation temperature on the photoluminescence intensities of the Sr_2CeO_4 phosphor particles in spray pyrolysis. Fig. 3 shows the XRD spectra of the as-prepared and post-treated particles obtained by spray pyrolysis. The as-prepared particles obtained by spray pyrolysis at a preparation temperature of 1,000 °C had phase of CeO_2 . No peak lines corresponding to Sr_2CeO_4 phosphor particles were observed in the XRD spectrum of the as-prepared particles obtained at 1,000 °C. The peak lines of the Sr_2CeO_4 phosphor appeared at the preparation temperature of 1,200 °C, in

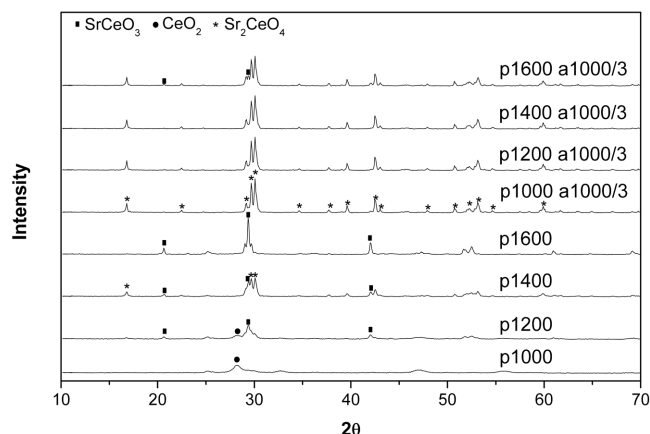


Fig. 3. XRD spectra of Sr_2CeO_4 phosphor particles.

which the particles had mixture peaks of SrCeO_3 , Sr_2CeO_4 and CeO_2 compounds. The as-prepared particles obtained at $1,400^\circ\text{C}$ had mixture peaks of Sr_2CeO_4 and SrCeO_3 compounds. SrCeO_3 was intermediate product in the preparation of Sr_2CeO_4 phosphor particles by spray pyrolysis. However, the as-prepared particles obtained at a preparation temperature of $1,600^\circ\text{C}$ had main peaks of SrCeO_3 compound. Sr_2CeO_4 decomposed into SrCeO_3 and SrO compounds at a preparation temperature of $1,600^\circ\text{C}$. The as-prepared particles obtained by spray pyrolysis at preparation temperatures below $1,400^\circ\text{C}$ converted into phase pure Sr_2CeO_4 phosphor particles after post-treatment at $1,000^\circ\text{C}$. However, the particles prepared by spray pyrolysis at a preparation temperature of $1,600^\circ\text{C}$ had impurity phase of SrCeO_3 after post-treatment at $1,000^\circ\text{C}$. In spray pyrolysis, some of strontium component formed by decomposition of Sr_2CeO_4 into SrCeO_3 and SrO evaporated at the preparation temperature of $1,600^\circ\text{C}$. Therefore, the phosphor particles prepared by spray pyrolysis at the preparation temperature of $1,600^\circ\text{C}$ had low photoluminescence intensity and impurity phase after post-treatment.

In spray pyrolysis, the preparation temperature of the as-prepared particles affected the morphology, crystal structure and photoluminescence characteristics of the Sr_2CeO_4 phosphor particles. The optimum preparation temperature of the as-prepared particles was $1,400^\circ\text{C}$ to produce the Sr_2CeO_4 phosphor particles with spherical shape and high photoluminescence intensity in the spray pyrolysis.

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