Synthesis of Yellow-emitting NaAlSiO₄:Eu²⁺ Phosphors using SiO Powder as a Silica Source

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Phosphor-converted white light-emitting diodes (pc-white LEDs) have been extensively attracted as an alternative to conventional incandescent and fluorescent lamps. pc-white LEDs have superior features such as a high luminescent efficiency, low energy consumption, and a long lifetime.¹ In order to advance the present technologies on the pc-white LEDs for lamps, it is significant to develop novel phosphor materials of high luminescent efficiency. Recently, NaAlSiO₄:Eu²⁺ phosphor is receiving much attention for using pc-white LEDs because it has high thermal stability and shows strong broad emission band.²,³ However, it is necessary to improve the luminescent properties of NaAlSiO₄:Eu²⁺ phosphor for using pc-white LEDs. In this study, to improve the luminescence properties of NaAlSiO₄:Eu²⁺ phosphors, we used SiO powder as a silica source in the prepared process. We have previously determined that the luminescent properties of Eu²⁺-doped silicate phosphors were successfully improved by a small amount of SiO powder as a silica source due to high reactivity and reducing effect.⁴

The crystal structure of the NaAlSiO₄:10 mol% Eu²⁺ phosphors synthesized using x mol% SiO powder as a silica source were identified by powder X-ray diffraction (XRD). The samples with x ≤ 50 were obtained as a single phase of the hexagonal NaAlSiO₄ structure, while an unknown impurity phase was observed in the samples with 40 ≤ x.

The emission intensity was successfully enhanced by SiO powder using as a silica source in the prepared process. The maximum emission intensity was obtained for the sample with x = 10, where the relative emission intensity was 58% of that for the commercial Y₃Al₅O₁₂:Ce³⁺ (P-46) phosphor. The relative emission intensity is increased by 14% compared with that of the conventional NaAlSiO₄:10 mol% Eu²⁺ phosphor without SiO powder (44%) previously reported.⁵ The increase of the emission intensity can be attributed to the increase of the Eu²⁺ concentration in the phosphors. A small amount of SiO powder using in synthetic process as a silica source results in the increase of Eu²⁺ content in the phosphor because the SiO powder has high reactivity and reduction effects.

Fig. 1 shows the Eu-L₃ edges XANES spectra for the NaAlSiO₄:10 mol% Eu²⁺ with x = 0 and 20. The XANES spectra of the samples consisted of two absorption peaks at 6972 eV and 6980 eV, which correspond present of Eu²⁺ and Eu³⁺, respectively. This result indicates that the Eu²⁺ content in the phosphors was effectively increased by using SiO powder as a silica source during prepared processes.

The dependence of the emission intensities on the Eu²⁺ concentrations in the NaAlSiO₄:y mol% Eu²⁺ phosphors prepared in the presence of 10 mol% SiO powder as a silica source is shown in Figure 2. The emission intensity increases with the amount of the Eu²⁺ until it reaches a maximum at y = 7, and then decreases due to concentration quenching. The relative emission intensity of NaAlSiO₄:7 mol% Eu²⁺ phosphor was 68% of that of the commercial Y₃Al₅O₁₂:Ce³⁺ (P-46) phosphor. It is reported that the maximum emission intensity of the NaAlSiO₄:Eu²⁺ phosphors without SiO powder was observed at the composition of NaAlSiO₄:10 mol% Eu²⁺. However, in the present NaAlSiO₄:Eu²⁺ phosphor with SiO powder, maximum emission intensity was obtained for NaAlSiO₄:7 mol% Eu²⁺, which can be also explained by an increase in the Eu²⁺ content with SiO powder using in the prepared process. The increase of the Eu²⁺ content in the phosphors usually causes concentration quenching.

References