Synthesis of Yellow-emitting $NaAlSiO_4:Eu^{2+}$ 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, $NaAlSiO_4:Eu^{2+}$ 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 $NaAlSiO_4:Eu^{2+}$ phosphor for using pc-white LEDs. In this study, to improve the luminescence properties of $NaAlSiO_4:Eu^{2+}$ phosphors, we used SiO powder as a silica source in prepared process. We have previously determined that the luminescent properties of Eu^{2+}-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 $NaAlSiO_4$:10 mol% Eu^{2+} phosphors synthesized using x mol% SiO powder as a silica source were identified by powder X-ray diffraction (XRD). The samples with $x \leq 30$ were obtained as a single phase of the hexagonal $NaAlSiO_4$ structure, while an unknown impurity phase was observed in the samples with $40 \leq x$.

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 $Y_3Al_5O_{12}:Ce^{3+}$ (P-46) phosphor. The relative emission intensity is increased by 14% compared with that of the conventional $NaAlSiO_4$:10 mol% Eu^{2+} phosphor without SiO powder (44%) previously reported.[2)] The increase of the emission intensity can be attributed to the increase of the Eu^{2+} concentration in the phosphors. A small amount of SiO powder using in synthetic process as a silica source results in the increase of Eu^{2+} content in the phosphor because the SiO powder has high reactivity and reduction effects.

Figure 1 shows the Eu-L_3 edges XANES spectra for the $NaAlSiO_4$:10 mol% Eu^{2+} with x = 0 and 20. The XANES spectra of the samples consisted of two absorption peaks at 6972 eV and 6980 eV, which correspond presence of Eu^{2+} and Eu^{3+}, repectively. This result indicates that the Eu^{2+} 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 Eu^{2+} concentrations in the $NaAlSiO_4$:y mol% Eu^{2+} 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 Eu^{2+} until it reaches a maximum at y = 7, and then decreases due to concentration quenching. The relative emission intensity of $NaAlSiO_4$:7 mol% Eu^{2+} phosphor was 68% of that of the commercial $Y_3Al_5O_{12}:Ce^{3+}$ (P-46) phosphor. It is reported that the maximum emission intensity of the $NaAlSiO_4:Eu^{2+}$ phosphors without SiO powder was observed at the composition of $NaAlSiO_4$:10 mol% Eu^{2+}[4)]. However, in the present $NaAlSiO_4:Eu^{2+}$ phosphor with SiO powder, maximum emission intensity was obtained for $NaAlSiO_4$:7 mol% Eu^{2+}, which can be also explained by an increase in the Eu^{2+} content with SiO powder using in the prepared process. The increase of the Eu^{2+} content in the phosphors usually causes concentration quenching.

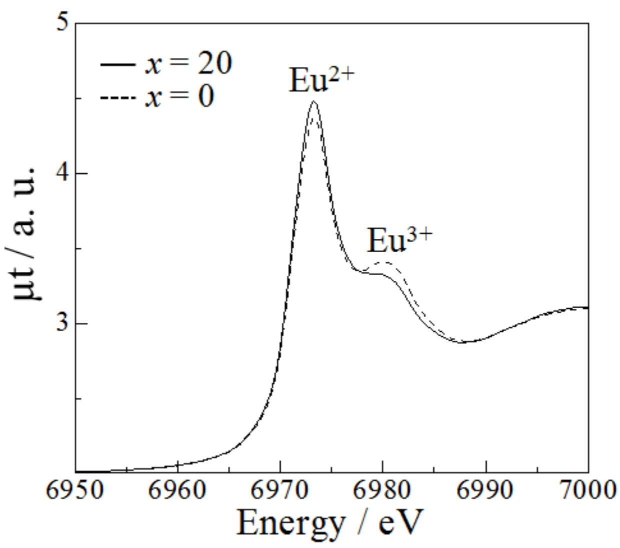

Fig. 1. Eu-L_3 edges XANES spectra for the $NaAlSiO_4$:10 mol% Eu^{2+} with x = 0 and 20.

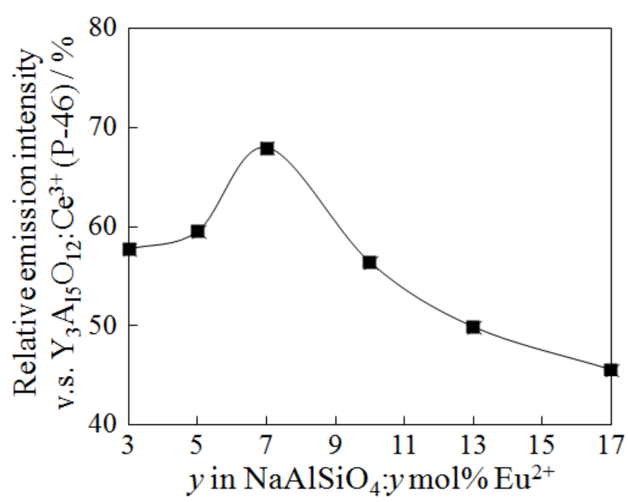

Fig. 2. Dependence of the emission peak intensity on the Eu^{2+} content in the $NaAlSiO_4$:y mol% Eu^{2+} ($3 \leq y \leq 17$) phosphors in the presence of 10 mol% SiO powder as a silica source. The excitation wavelength is 405 nm for $NaAlSiO_4$:y mol% Eu^{2+} and 460 nm of $Y_3Al_5O_{12}:Ce^{3+}$ (P-46).

References

1. E. F. Schubert et al., *Science*, **308**, 1274 (2005).
2. D. S. Jo et al., *J. Ceram. Process Res.*, **12**, 47 (2011).
3. D. S. Jo et al., *Opt. Mater.*, **34**, 696 (2012).
4. K. Toda, *J. Ceram. Process Res.*, **12**, 256 (2011).