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Photoluminescence property of 15R-sialon:Eu^{2+} phosphors and their XANES spectra

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Introduction

Sialon ceramics are the most widely studied and used high temperature structural materials over the past many years. Recently, both α- and β-sialons have been used as host materials for RE cations to give luminescence, e.g., Ca-α-sialon:Eu^{2+} [1] and β-sialon:Eu^{2+} (green) [2]. The luminescence allows sialon materials to find new functional applications in displays and white light-emitting diodes (LEDs), due to superior thermal and chemical stabilities.

In addition to the α- and β-sialons, there are other types of sialon phases. It is well known that in the phase diagram of Si_3N_4, SiO_2, Al_2O_3, and AlN, at the compositions between β-sialon and AlN, there are several AlN-polytypoid phases, e.g., 8H, 15R, 12H, 21R, 27R and 2H. In our preliminary study, Eu^{2+} doped 15R-sialon powders prepared by gel nitridation and post-annealing were found to exhibit a blue light emission under 254nm excitation [3]. However, the phase pure Eu^{2+} doped 15R sialon has not been obtained, and effect of Eu concentration on the luminescence was also not studied.

In this study, Eu^{2+} doped 15R-sialon ($SiAl_4O_2N_4$) was synthesized from the powder mixture of Si_3N_4, SiO_2, Al_2O_3, AlN, and Eu_2O_3 by gas-pressure sintering, and photoluminescence property was discussed with Eu valency.

Experimental

The raw powders were homogeneously mixed with ethanol, and the mixtures were fired in a gas pressure sintering furnace at 1800°C for 3h under a nitrogen pressure of 0.5MPa. The Eu concentration was varied from 0 to 1.5mol% with respect to 15R-sialon.

Phase identification was conducted by powder X-ray diffraction (XRD). Photoluminescence property was measured by a spectrofluorometer. X-ray absorption of Eu L_{III}-edge was measured in transmission mode at the beam line 9C in Photon Factory.

Results and discussion

The formation of 15R-sialon was observed after gas pressure sintering at 1800°C. At 0 and 0.05 mol% Eu, in addition to the major 15R phase, two kinds of impurities β-sialon and AlN were observed. At 0.1 and 0.5 mol% Eu, the impurity was only the residual AlN. When the Eu concentration increased to 1 mol% and above, single phase of 15R-sialon was formed without these impurities. These Eu doped 15R phases exhibit broad blue emission spectra with maximum at 445 nm and a shoulder at about 465nm under UV light excitation.

Figure 1 shows Eu L_{III}-edge XANES spectra of the synthesize 15R phase with various Eu contents. All products contained both Eu^{2+} and Eu^{3+}. The ratio of Eu^{2+}/Eu^{3+} increases with increasing Eu contents. However, no Eu^{3+} emission (line spectra around 620nm) was observed in PL measurement. These results indicate that a part of Eu^{2+} enter into the 15R-sialon phase resulting in the broad blue emission. Eu^{3+} might form a liquid phase during the sintering, and remain as an amorphous phase. The two emission peaks suggest that there are two kinds of Eu^{2+} site having difference coordination environment.

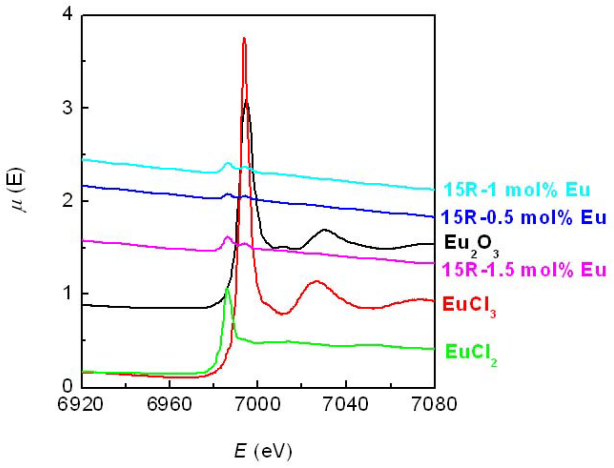

Fig. 1 Eu LIII-edge XANES spectra of Eu doped 15R-sialon products and references.

References

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