

# Photoluminescence property of magnetoplumbite-type $\text{LaAl}_{12}\text{O}_{18}\text{N}:\text{Eu}^{2+}$ phosphor and their XANES spectra

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## 1 Introduction

White light emitting diodes (LEDs) have received much attention as high efficiency, safe and environmental friendly devices. In white LED applications, blue LED light is combined with yellow light emitted from a phosphor to obtain white light. To improve the color rendering properties of white LEDs, both green and red light emitting phosphors have been used with yellow phosphor. Such phosphorous materials have been developed among silicon oxynitride and alumino-silicon oxynitrides. However, study on aluminum (oxy)nitrides is limited.

Three emission peaks for  $\text{Eu}^{2+}$  doped AlON with an impurity phase of magnetoplumbite(MP)-type  $\text{EuAl}_{12}\text{O}_{19}$  has been reported by Kikkawa et al. [1]. They attributed the multiple emissions to the presence of the different coordination environments around  $\text{Eu}^{2+}$  with different O/N ratios in the MP structure. The MP-type oxynitride has been reported only in lanthanum aluminum oxynitride,  $\text{LaAl}_{12}\text{O}_{18}\text{N}$ . However, the crystal structure and photoluminescence properties of  $\text{Eu}^{2+}$  doped MP-type lanthanum aluminum oxynitride have not yet been reported.

In this study,  $\text{Eu}^{2+}$  doped MP-type  $\text{LaAl}_{12}\text{O}_{18}\text{N}$  was prepared using aluminum glycine gel precursors followed by post-annealing of the calcined precursor. The luminescence property was discussed in relation to the crystal structure and Eu valency.

## 2 Experiment

The calcined precursors were prepared through ammonolysis of oxide powders obtained from aluminium glycine gel as described in Ref [1]. The calcined precursors were post-annealed at 1700 °C for 3 h under a nitrogen gas pressure of 0.2 MPa in a gas pressure furnace.

Crystalline phases were characterized using powder x-ray diffraction (XRD). Luminescence property was measured by a spectrofluorometer. X-ray absorption of Eu L<sub>III</sub>-edge was measured in transmission mode at the beam line 9C in Photon Factory. Neutron diffraction measurements were performed using HERMES installed at JRR-3 in JAEA. The Rietveld program RIETAN-2000 [2] was used for structural refinement.

## 3 Results and Discussion

After post annealing at 1700 °C for 3 h under a nitrogen gas pressure of 0.2 MPa, pure MP-type  $\text{LaAl}_{12}\text{O}_{18}\text{N}$  doped with Eu were obtained from 0% to 30% of Eu doping. Eu doped products exhibited broad blue emission at 440 nm with a shoulder peak at 464 nm under excitation at 254 nm. No characteristic sharp peaks due to  $\text{Eu}^{3+}$  ion were observed, which confirms the presence of only  $\text{Eu}^{2+}$  ions in the oxynitride phosphor. The  $\text{Eu}^{2+}$  was supported by a single absorption peak at 6972 eV in the XANES spectra for the Eu L<sub>III</sub>-edge as shown in Fig. 1. Eu doped  $\text{CaAl}_{12}\text{O}_{19}$  had a single emission at 415 nm.

Structural refinement using neutron diffraction showed the lanthanum site occupied by  $\text{Eu}^{2+}$  is split into *2d* and *6h* sites at the intermediate layer in the MP-type aluminium oxynitride. The shoulder emission peak in the emission spectrum of the oxynitride is related to the  $\text{Eu}^{2+}$  site splitting induced by the two kinds of anions.

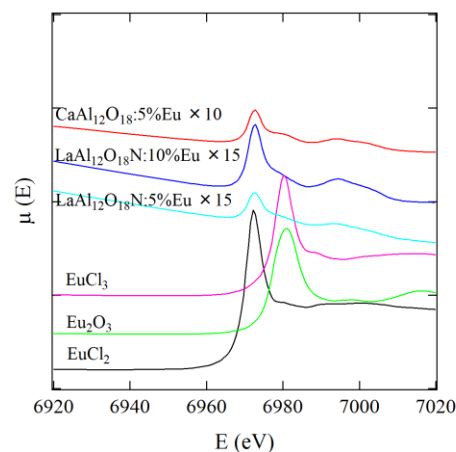


Fig. 1 Eu L<sub>III</sub>-edge XANES spectra of Eu doped MP-type  $\text{LaAl}_{12}\text{O}_{18}\text{N}$  and  $\text{CaAl}_{12}\text{O}_{19}$ .

## References

- [1] S. Kikkawa, et al., *J. Am. Ceram. Soc.*, **91** (2008) 924.
- [2] F. Izumi, et al., *Mater. Sci. Forum*, **321-324** (2000) 198.

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