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# Investigation of Solid-State Devices Using Deep-Ultraviolet Emitting $ZnAl_2O_4$ Thin Film

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## ABSTRACT

ZnO films were deposited on sapphire substrates by magnetron sputtering and annealed to produce zinc aluminate ($ZnAl_2O_4$) deep ultraviolet emitting phosphor thin films by mutual thermal diffusion between film and substrate. We explored the annealing temperature and time to find the optimum conditions in terms of luminescence intensity.

## 1 INTRODUCTION

UV light has high energy and is said to be appropriate for sterilization because it can decompose C=C bonds in cells. The currently used UV light sources, such as mercury lamps and UV LEDs, have problems in terms of cost and the risk of using mercury, and the need for new UV light sources is focused on high efficiency, low cost, and low environmental impact. In this study, we focus on $ZnAl_2O_4$, a deep UV emitting phosphor. [1]

Conventionally, powder samples have been prepared and evaluated [2], but it was difficult to analyze transmittance and refractive index due to the influences of surface scattering, so it would be possible to evaluate these basic physical properties by preparing thin film samples. In this research, ZnO films were deposited on sapphire substrates by sputtering and $ZnAl_2O_4$ thin films were prepared by thermal diffusion [3, 4]. These films were evaluated its optical properties for application to solid-state devices such as EL lamps.

## 2 EXPERIMENT

Fig.1 shows the experimental procedure and conditions for preparing the $ZnAl_2O_4$ thin film. ZnO layer of 300 nm and α-$Al_2O_3$ layer of 50 ~200 nm as a capping layer to prevent re-evaporation of ZnO film was deposited on a-plane sapphire substrates by RF magnetron sputtering. Thereafter, films were annealed at 980~ 1100 ºC for 40 to 75 hours in air. $ZnAl_2O_4$ films were formed by thermal diffusion of Zn and Al between deposited ZnO film and sapphire substrate. Crystal structure and element distributions of films were evaluated by X-ray diffraction (XRD) and field emission-scanning electron microscope analysis (FE-SEM), and luminescence property was analyzed by cathodoluminescence (CL).

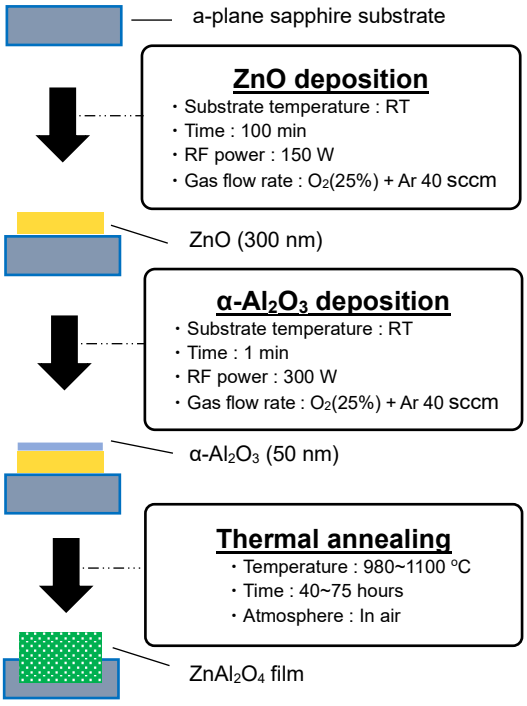


Fig.1 Experimental procedure of preparing $ZnAl_2O_4$ films.

## 3 RESULTS AND DISCUSSIONS

Figure 2 shows the CL spectra of $ZnAl_2O_4$ thin films prepared by annealing at 990°C to 1100°C for 75 hours. Ultraviolet emission from $ZnAl_2O_4$ was observed around 230 nm. Differences in the UV emission intensity were observed depending on the annealing temperature, and the sample prepared by annealing at 990°C showed the strongest UV emission intensity. The lower strength at higher temperatures is probably due to the reduction of oxygen defects and ZnO re-evaporation caused by high

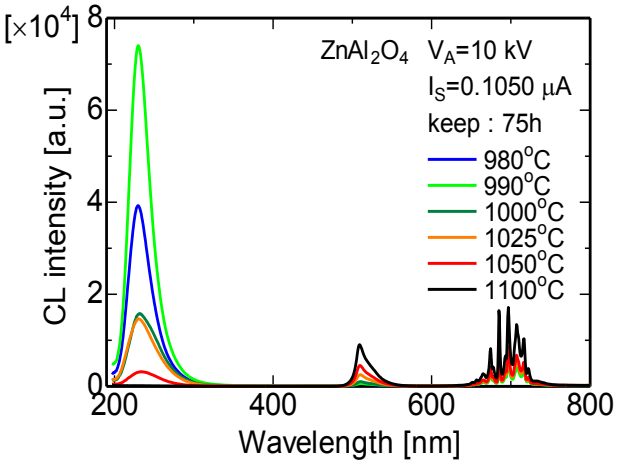


Fig.2 CL spectra of the samples annealed at several temperatures for 75 hours.

temperature annealing.

Figure 3 shows the relationship between the UV emission intensity of the sample annealed for 75 hours and calculated electron beam penetration depth. At each temperature, the increase in UV emission intensity with increasing excitation range was uneven. it indicates that quality of the $ZnAl_2O_4$ film was not uniform because of too high annealing temperature, that caused unbalance of diffusion speed and crystal formation.

Figure 4 shows the CL spectra of the sample annealed at 990°C. No significant change in the UV emission intensity with annealing time was observed, but the emission of the raw material, ZnO, was observed only after 40 hours of annealing. The required annealing time is likely to be 50 hours or more.

Figure 5 shows the relationship between the UV emission intensity and the calculated electron beam penetration depth. As the excitation range increased, the UV emission intensity increased proportionally. This result

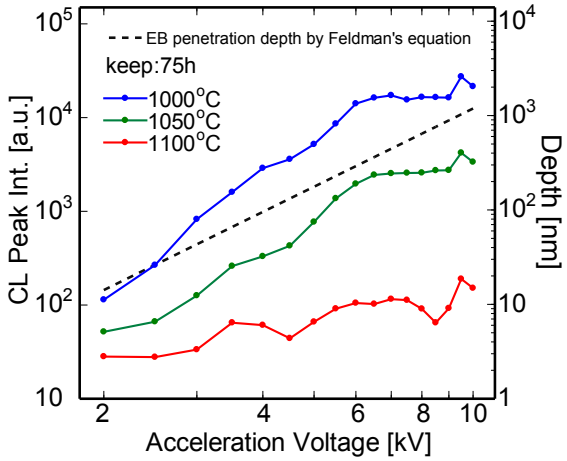


Fig.3 Relationship between ultraviolet intensity and calculated electron penetration depth (75 hours annealing).

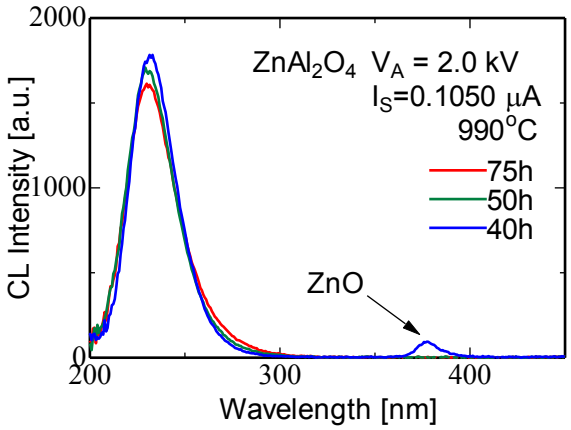


Fig.4 CL spectra of samples annealed at 990°C for several hours (excited surface region)

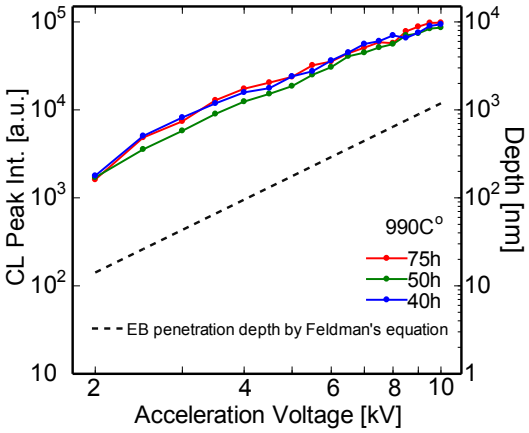


Fig.5 Relationship between ultraviolet emission intensity and calculated electron penetration depth.

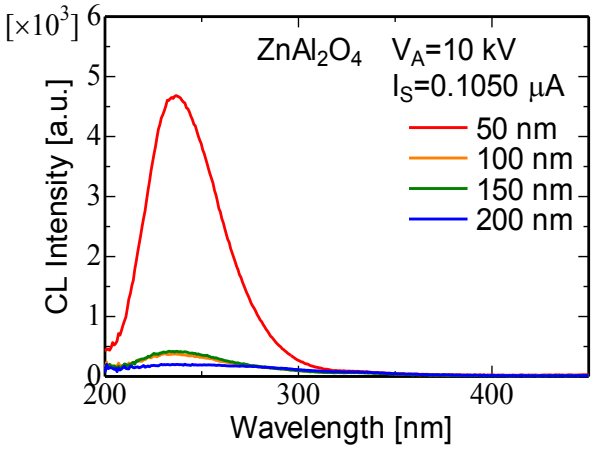


Fig.6 CL spectra of ultraviolet intensity of the samples with several thickness of α-$Al_2O_3$ cap layers.

suggests that a homogeneous luminescent layer is formed from the surface to the interior of the sample.

Figure 6 shows the UV emission intensity when the thickness of the alumina cap layer is varied. As the cap layer thickness was increased, the UV emission intensity decreased, and the UV emission intensity was highest at 50 nm deposition. It is possible that ZnO reacted with alumina in the cap layer, which is an amorphous phase, resulting in the formation of a poor luminescent layer. It indicates that good quality $ZnAl_2O_4$ phase was formed by the reaction of ZnO layer and sapphire substrate.

Figure 7 shows the SEM image and elemental mapping of the sample prepared by annealing at 990°C for 50 hours. For ZnO sputtering of about 300 nm, Zn was distributed in the thickness direction from the surface to about 750 nm.

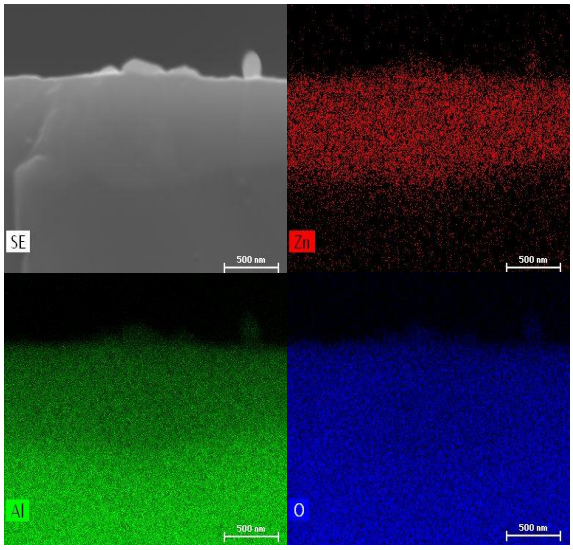


Fig.7 SEM images and elemental mapping of samples prepared by annealing at 990°C for 50 hours.

## 4 CONCLUSION

Deep UV emitting $ZnAl_2O_4$ thin films were prepared by thermal diffusion between ZnO film and sapphire substrate, and the thin film formation conditions were optimized in terms of UV emission intensity.

The best UV emission was obtained by annealing at 990°C, and the intensity of UV emission decreased with annealing at higher temperatures.

SEM and EDS measurements showed that Zn diffused to about 2.5 times the thickness of 300 nm ZnO sputter by annealing for 50 hours.

Annealing at 990°C for 50 hours is considered to be optimal.

## 5 REFERENCES

[1] Hiroko Kominami, Yoichiro Nakanishi, Proceedings of International Vacuum Nano-electronics Conference 2009, pp.67-68 (2009).

[2] Takeaki Ishinaga, Taku Iguchi, Hiroko Kominami, Kazuhiko Hara, Mamoru Kitaura, Akimasa Ohnishi, Physica Status Solidi C, 12/6, pp.797-800 (2015).

[3] Taro Ito, Hiroko Kominami, Yoichiro Nakanishi, Kazuhiko Hara, Proceedings of IDW ’15, PHp1-11L (2015).

[4] Kazuto.Kijima, Taro Ito, Hiroko Kominami, Yoichiro Nakanishi, Kazuhiko Hara, Proceedings of IDW ’17, PHp1-8L pp.659-660 (2017)