P-71: Deposition and Luminescent Characterization of Y₃Al₅O₁₂:Ce Thin Film Phosphor

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Abstract

Trivalent cerium (Ce^{3}) activated yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) phosphor thin films were deposited on quartz glass substrates by rf magnetron sputtering. The effects of sputtering parameters and annealing condition on the luminescent properties were investigated. The sputtering parameters were O_2/Ar gas ratio, rf power, and deposition time. The films were annealed at 1200 °C for 5 hours in N_2 +vacuum atmosphere. Polycrystalline YAG:Ce thin film phosphor could be obtained with a gas ratio of $O_2/(Ar+O_2)=0.5$ after post-annealing. PL spectra excited at 450 nm showed a yellow single band at 550 nm.

1. Introduction

LEDs have been extensively used in markets such as traffic lights, automobile brake light, backlights of liquid crystal displays (LCD), and mobile telephone. LEDs have merits of high brightness, high power efficiency, low applied voltage and long lasting life. White LEDs are spotlighted for next generally illumination. Recently, white LED organizing indium galliun nitride (InGaN) of blue light and YAG yellow phosphor at epoxy is commercialized. [1],[2]

YAG:Ce phosphor are cubic garnet structure, and widely used in yellow phosphor. Usually, the Ce³⁺ emission is in the ultraviolet (UV) or blue spectra. However, trivalent Ce³⁺ activated YAG:Ce phosphor showed yellow band at 550 nm.^{[3]-[5]} The yellow emission is intense enough to compete with the residual blue light that escapes through the phosphor and then to produced white light.^[6] Recently, trivalent Ce³⁺ activated YAG:Ce phosphor of white LED manufacturing techniques have been commercialized.

At present, commercial white LEDs are fabricated with LED and YAG:Ce powder. However, thin film YAG:Ce was rarely reported.

Thin films YAG:Ce has not only advantages of easy fabrication process, but also high efficiency. The blue light intensity of LED can be reduced during passing through the epoxy, so YAG:Ce powders in the epoxy are exposed to weakened blue light.

While on the other, thin films YAG:Ce phosphor is just adhered to the surface of blue LED, so it can directly gain LED's light energy.

In this work, we synthesized the sputtering target with trivalent cerium activated YAG powder. And YAG phosphor thin films were deposited by rf magnetron sputtering. The effects of sputtering parameters and annealing condition on the luminescent properties were investigated.

2. Experiment

We prepared a sputtering target with commercialized YAG:Ce phosphor powder by conventional ceramic process. YAG:Ce thin film phosphor were deposited on quartz glasses by rf magnetron sputtering. The effects of sputtering parameters and annealing condition on the luminescent properties observed. The sputtering parameters were O₂/(Ar+O₂), rf power, and deposition time. Details are shown in Table. 1. The films were annealed at 1200 °C for 5 hours in vacuum atmosphere, purging N₂ gas.

The crystalline of thin films were analyzed by XRD (X-ray diffractometer, SIEMENS D5005), and luminescence was measured by PL (Photoluminescence, PSI Darsa-5000) system. SEM (Scanning electron microscope, JEOL JSM-6500F) was used to observe the structure and thickness of thin films. All samples had the same thickness by controlling the deposition time.

Table 1. Sputtering parameter for YAG:Ce films

Parameter	Conditions	
Target size	Ф 2"	
rf Power	150W	
Substrate temperature	550℃	
$O_2/(Ar+O_2)$	20~50%	
Working pressure	5mTorr	
Target-Substrate distance	5cm	

3. Results and Discussions

XRD patterns of YAG:Ce phosphor thin films as a function of thickness are shown in Fig. 1. As-deposited films were almost amorphous, so XRD spectra could not be detected. However, after annealing, they were transformer to the polycrystalline phases. Atoms in as-deposited films were rearranged to organize the crystalline phase due to the thermal energy during the annealing process. XRD peaks are in exact accordance with those of JCPDS card. With increasing the film thickness, peak became more apparent, and peak intensities increased.

XRD spectra of YAG:Ce films deposited with various gas ratio of O₂/(Ar+O₂) are shown in Fig. 2. At low oxygen pressure, YAG:Ce films were not synthesized due to the deficient oxygen source. With increasing oxygen partial pressure, peak intensities

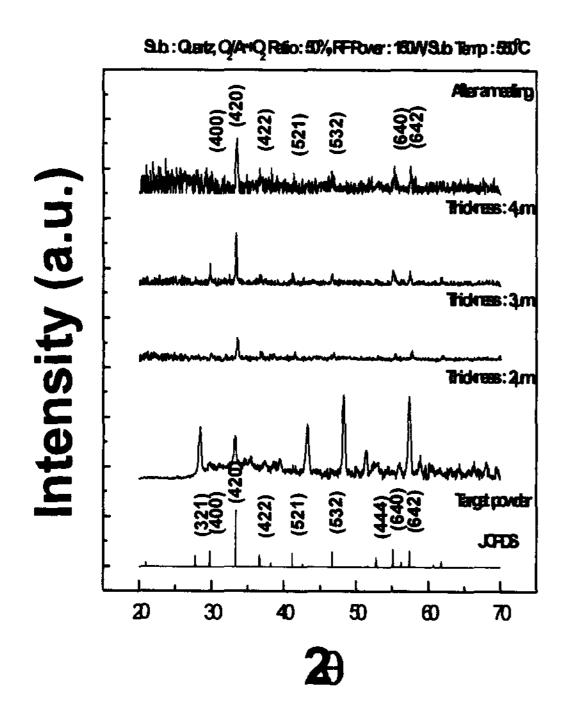


Fig. 1 XRD patterns of YAG:Ce films with variation of thickness.

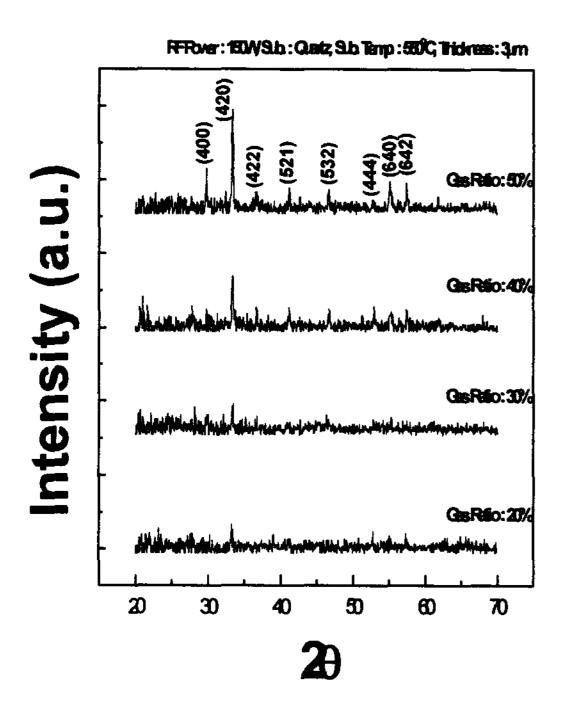


Fig. 2 XRD patterns of YAG:Ce films deposited with various gas ratio of O₂/(Ar+O₂).

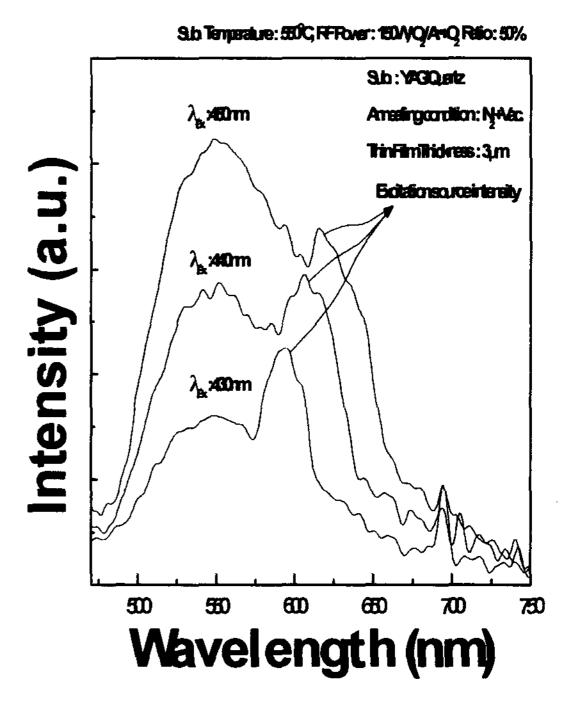


Fig. 3 PL spectra of YAG:Ce thin films excited at various wavelengths.

became higher. Finally, at 50%, well-developed polycrystalline YAG:Ce films could be obtained.

Fig. 3 shows PL spectra of YAG:Ce excited at various excitation wavelength. All peaks exhibited a yellow single band at 550 nm regardless of the excitation wavelength, but the intensities depended on it. In this experimental condition, at 450 nm that is the same with the emission wavelength of commercialized blue LED for white LEDs, highest intensity could be obtained. It was clear that emission peaks on the right side of main peaks at 550 nm were resulted from the light source during measuring, because they shifted to the long wavelength coinciding with increasing the excitation wavelength.

Fig. 4 shows PL spectra of YAG:Ce films deposited with various gas ratio. By adding oxygen in sputtering gas, emission intensity increased because of the better crystallinity. Prevariously described in Fig. 2, high quality YAG:Ce film could be obtained at the gas ratio of O₂/(Ar+O₂)=0.5, which exhibited high luminescent intensity. Annealing and the enhanced crystallinity contributed to the luminescent properties, because they reduced the defects such as point defect, strain, and quenching site, etc., which are related to nonradiative centers.

4. Conclusion

YAG:Ce thin film phosphor were deposited by rf magnetron sputtering method. As-deposited films were almost amorphous. But, the films were transformed to polycrystalline phase after post-annealing. Also, YAG:Ce thin film phosphor deposited at the

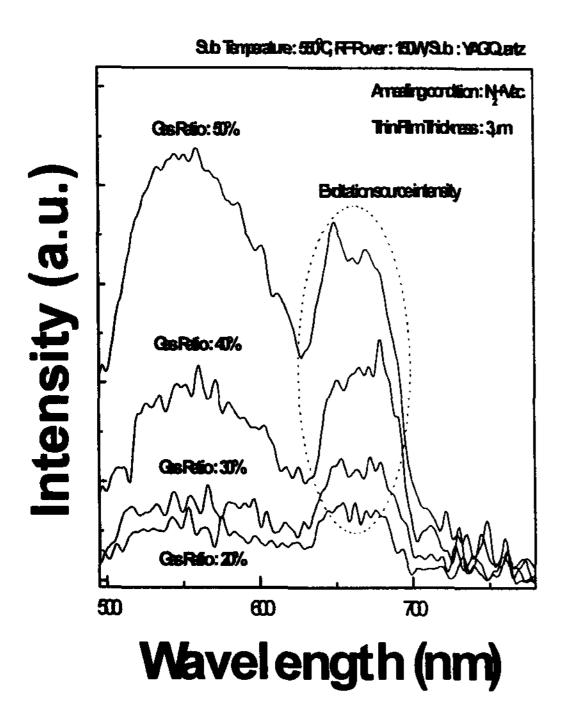


Fig. 4 PL spectra of YAG:Ce thin films deposited with various gas ratio of O₂/(Ar+O₂).

gas ratio of $O_2/(Ar+O_2)=0.5$ showed yellow band PL emission at 550 nm ($\lambda_{ex}=450$ nm)

5. Acknowledgements

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6. References

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