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Effect of Alumina Nanocoating on Thermal Stability of $Y_3Al_5O_{12}$: Ce Phosphor

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Al_2O_3 film was successfully coated on the surface of YAG: Ce phosphor. The microscopic structure and thermal stability of Al_2O_3-coated phosphor were investigated. The results show that a smooth and uniform alumina nanocoating was formed on the surface of YAG: Ce phosphor particles, which revealed the formation of YAG: Ce-Al_2O_3 core-shell structure. The optical properties of Al_2O_3 coated phosphor have shown that the luminescent intensity of Al_2O_3-coated YAG: Ce phosphor decreases slightly. But the thermal stability became better and the resistant light decay has been improved *ca.* 20 %.

Key Words: YAG: Ce, Phosphor, Al_2O_3 coating, Nanocoating.

INTRODUCTION

It is worth mentioning that the full form white light emitting diodes (W-LEDs) have been widely considered as the 4th-generation light sources due to the advantages of small size, energy saving, environmental protection, fast response, long life, *etc*. Phosphorus play an important role in the realization process of white light emitting diodes[1,2]. The conventional $Y_3Al_5O_{12}$: Ce (YAG: Ce) phosphorus often has some deficiencies of agglomeration, poor thermal stability because of the internal factors. Many studies found that the formation of a coating on the surface of phosphorus with alumina, silica and other inorganic materials can segregate the phosphorus from the outside world, which can effectively reduce the external impact on the phosphorus[3,4]. So it can extend the life expectancy of the phosphorus. In addition, the coating process of phosphorus also effectively eliminates some surface defects of the phosphorus, such as metal-oxygen dangling bond.

Dry-process and wet-process have been reported for the coating on the surface of phosphorus. As a wet-process, the sol-gel method has been widely used in the preparation of thin film[5-8]. Sol-gel method is generally including two steps: the precursor of coating material is added to the solution of phosphorus, followed by adjusting the pH of the solution in order to coat the precipitated material on the surface of phosphorus[9]. Wang *et al.*[10], reported that the Al_2O_3-coated YAG: Ce phosphorus prepared by heating the suspension of aluminum isopropoxide mixed with YAG: Ce phosphorus, shows smaller light decay than uncoated, but the luminous intensity decreased obviously. Kajiyoshi *et al.*[11], obtained a smooth and uniform titania nanocoating on ZnS particles with the coating thickness in a range from 20-54 nm. Several coating materials such as Al_2O_3, SiO_2[12], $BaTiO_3$, In_2O_3, Y_2O_3[13] have been investigated, where alumina is a traditional coating material which is economically cheap and easy to obtain. In this paper, we presented a modified method for the preparation of Al_2O_3 nanocoating on the surface of YAG: Ce phosphorus in order to improve the thermal stability and the ability of resistant luminous decay.

EXPERIMENTAL

Sodium acetate (A.R.), aluminum nitrate nonahydrate ($Al(NO_3)_3\cdot 9H_2O$, A.R.), acetic acid (HAc, A.R.) and polysorbate are obtained from Chongqing Chuandong Chemical, Ltd., China. All these chemicals are used without further purification. YAG: Ce phosphor is commercial.

Coating methods: YAG: Ce phosphor was slowly added to aqueous solution of sodium hydroxide and then kept for 12 h. The pre-treated phosphorus was obtained by filtration, washing with water and drying.

The aqueous solution of aluminum nitrate (0.56 mol/L) and the suspension of YAG: Ce particles were prepared, respectively. Polysorbate (1 mL) and phosphorus (10 g) were added into 500 mL of NaOAc-HOAc solution (pH = 4.5-6.0), respectively, to form phosphorus suspension. The aluminum nitrate solution was slowly added to the phosphorus suspension at room temperature under vigorously stirring for 1 h. Then the pH of mixture was adjusted to 6.0-7.0 by addition of NaOH.

After filtering and drying, the resulting solid was heated at 300 ºC for 2 h, then obtaining Al_2O_3-coated YAG: Ce phosphor.

Characterization of coated phosphorus: The photoluminescence were measured by RF-5301 molecular fluorescence spectrometer equipped with xenon lamp as excitation source. The excitation and emission slit was 5 nm and the scan range was 300-800 nm. The relative brightness of phosphorus was investigated by JY-2003 phosphor relative brightness test meter with PE-5 phosphorus parameters measurement. The microscopic structure of phosphor was observed by TESCAN VEGAIILMU scanning electron microscope. The surface morphology of phosphorus particles were characterized by Tecnai G2F20 field emission transmission electron microscope. The composition of the samples was studied by INCA Energy 350 EDS method.

RESULTS AND DISCUSSION

Microstructural and compositional analysis: Fig. 1 shows the SEM images of (a) uncoated and (b) Al_2O_3-coated YAG: Ce phosphor particles. The particles without coating are well-distributed and spherical and their diameter is in range of 1-2 μm. As shown in Fig. 1(b), the diameter and shape of phosphor particles almost have no change compared with the uncoated YAG: Ce phosphor.

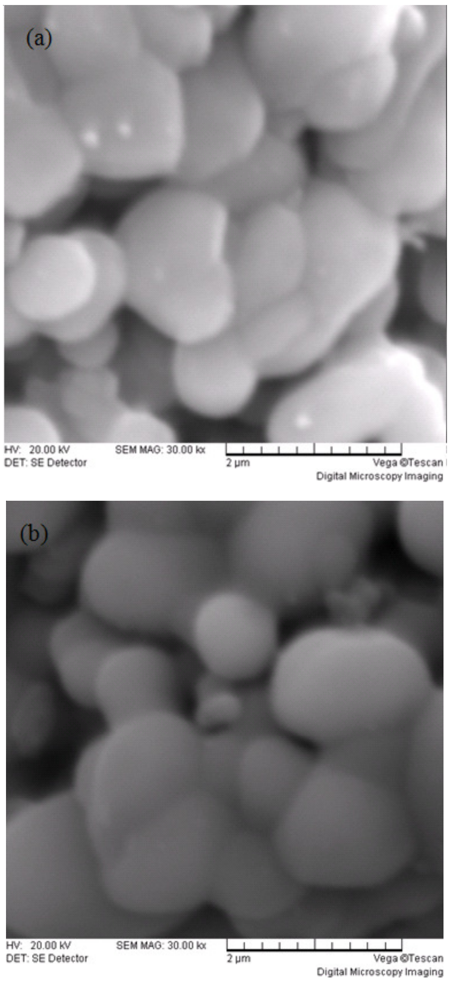

Fig. 1. SEM images of (a) uncoated and (b) Al_2O_3-coated YAG: Ce phosphor particles

Before coating, the raw YAG: Ce phosphor has a clean, smooth and even surface, as shown in Fig. 2(a) and no core-shell kind of structure was observed. TEM image of a typical alumina-coated YAG: Ce particle was displayed in Fig. 2(b), which reveals that the particle consists of a crystalline YAG: Ce core surrounded by a thin alumina layer. The YAG: Ce-Al_2O_3 interface is very sharp, showing little or no phase mixing. The typical surrounding oxide thicknesses are about 10-15 nm.

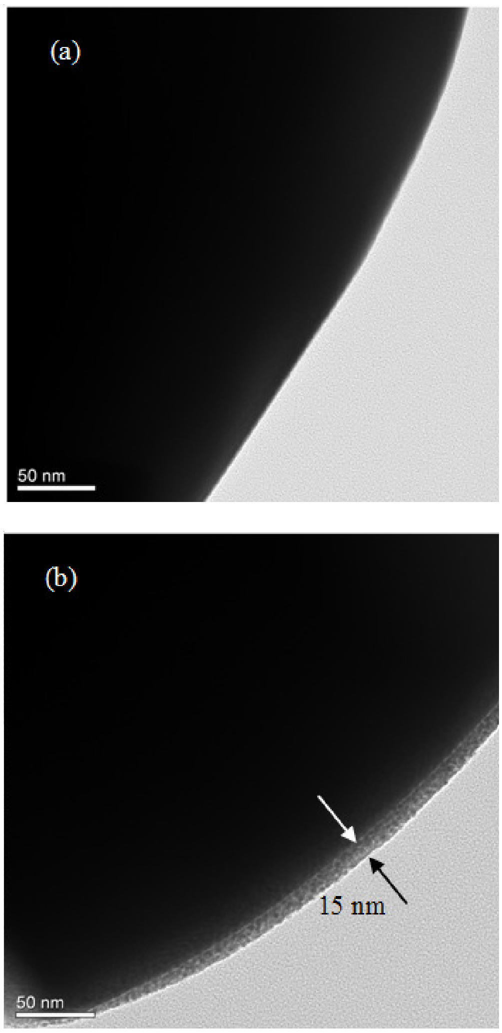

Fig. 2. TEM micrographs of (a) the original and (b) alumina-coated YAG: Ce particle

Fig. 3 displays the EDS spectrum of (a) uncoated and (b) Al_2O_3-coated YAG: Ce phosphor. It can be seem that the contents of Al obviously increase from 21.94-32.88 % compared with uncoated YAG: Ce phosphor. The atoms content of oxygen has little change. The peak of Ce is too weak due to low cerium concentration in YAG: Ce phosphor. Because of the coating of Al_2O_3 nano-layer, the atoms content of Y reduce significantly, from 44.38-34.24 %. The increase of aluminum and the reduction of yttrium suggest that Al_2O_3 nano-layer is successfully coated on the surface of YAG: Ce phosphor.

Effect of alumina nanocoating on the properties of YAG: Ce phosphor: Fig. 4 shows that the emission spectra of uncoated and Al_2O_3-coated YAG: Ce phosphor. The maximum emission wavelength did not change, which indicates that Al_2O_3

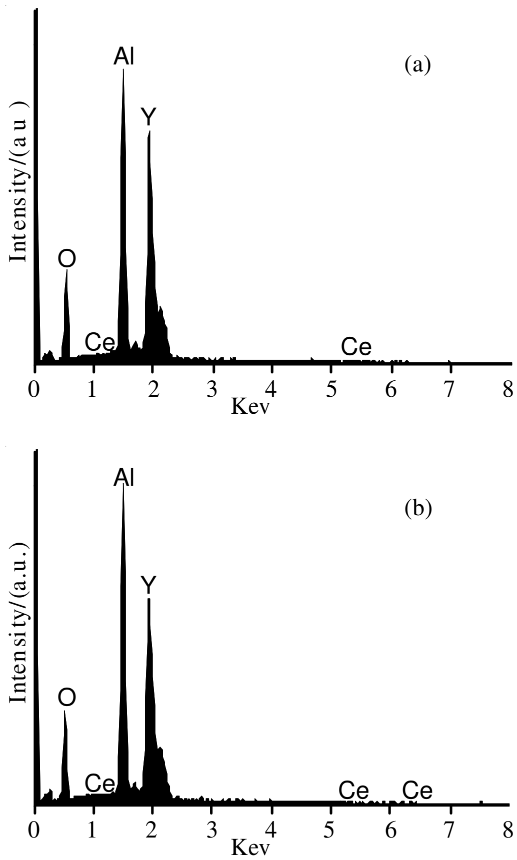

Fig. 3. EDS of Al_2O_3-coated YAG: Ce phosphor

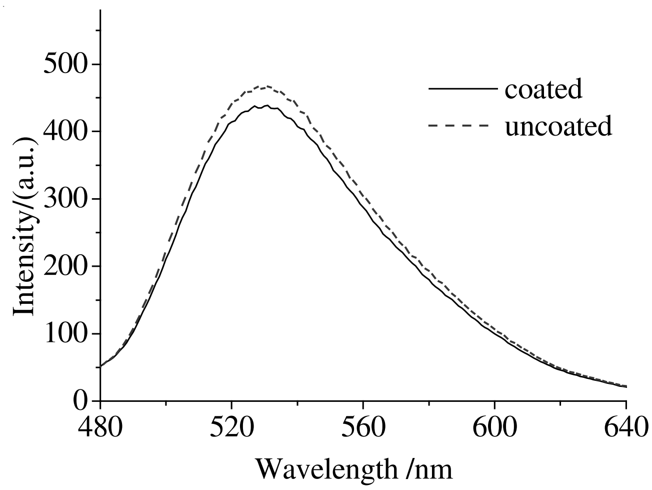

Fig. 4. Emission spectra of Al_2O_3-coated and uncoated YAG: Ce phosphor

nano-layer can not change the luminescent property of YAG: Ce phosphor. While the luminescent intensity of Al_2O_3-coated YAG: Ce phosphor decreases slightly after coating.

The relative brightness of YAG: Ce phosphor was detected by phosphor relative brightness test meter after heat-treatment for 0, 12, 24, 36, 48 and 60 h at the simulating operating temperature of 200 ºC, respectively. The brightness decay rates of uncoated YAG: Ce phosphor is 7, 13, 25, 31 and 50 %, respectively. While the relative brightness of the Al_2O_3-coated YAG: Ce phosphor is respectively 3, 9, 14, 25 and 34 %. Though the brightness of the coated YAG: Ce phosphor is slightly lower than that of uncoated phosphorus, the properties of thermal stability and resistant luminous decay can be improved obviously at 200 ºC.

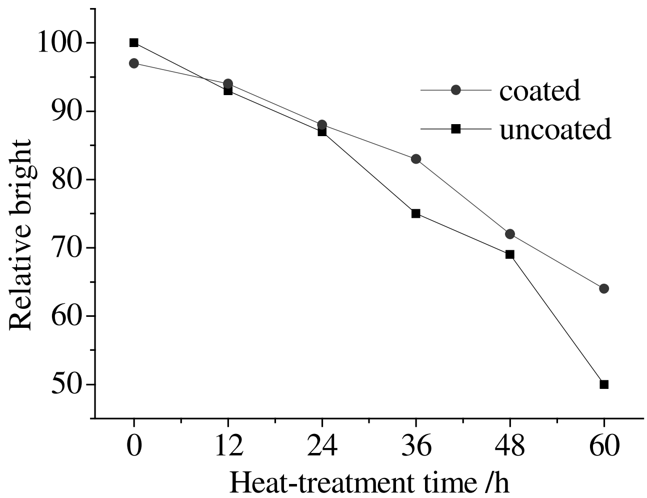

Fig. 5. Effect of heat treatment time on relative brightness of YAG: Ce phosphor

Conclusion

In this work, Al_2O_3 film was successfully coated on the surface of YAG: Ce phosphor. The optical properties of Al_2O_3 coated phosphor have shown that the luminescent intensity of Al_2O_3-coated YAG: Ce phosphor decreases slightly after coating. The thermal stability became better and the resistant light decay has improved about 20 %. It is valuable to extend the life of white light emitting diodes.

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