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Photoluminescence Characteristics of YAG:Ce Phosphor by Sol-Gel Method

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1. Introduction

The white LEDs(Light Emitting Diodes) have been extensively used in lighting devices such as room illuminations, car headlights and display devices such as LCD(Liquid Crystal Display) TVs, monitors, cellular phones, digital cameras, camcorders. This is due to various advantages such as low applied voltage, power efficiency, high brightness and long lifetime. Therefore, study for white LED is coming along very actively. Recently, a number of LEDs that emits blue light (at 440~480nm) efficiently has been developed. It is possible to express white light luminescence using the blue LED.[1]

The cerium(Ce) activated yttrium aluminum garnet(YAG) is well known as phosphor of the blue excitation and yellow emission for white LED. YAG phosphor is prepared conventionally by solid state reactions. This method exist stable intermediate phases such as YAP(Yttrium Aluminum Perovskite, $YAlO_3$) and YAM(Yttrium Aluminum Monoclinic, $Y_4Al_2O_9$) due to high sintering temperature(1600~1800℃), milling process for a long time and low reactivity of raw materials.[2] Whereas, sol-gel method is possible to fabricate nano particle into low temperature processing and uniform mixing.

In this study, the Ce doped YAG phosphor precursors were synthesized by a sol-gel method. The crystalline phase compositions, particle size, morphologies and optical properties of the synthesized powders were evaluated using the XRD, SEM and PL.

2. Experimental

The starting materials for YAG:Ce phosphors were $Y(NO_3)_3$•$6H_2O$(99.9%, Aldrich), $Al(NO_3)_3$•$9H_2O$(99.997%, Aldrich), $Ce(NO_3)_3$•$6H_2O$(99.999%, Aldrich). Citric acid and ethylene glycol were used as reagent and solvent. The solution was prepared by dissolving proper amounts of yttrium nitrate, aluminum nitrate, and cerium nitrate into distilled water which gives Y:Al:Ce mole ratio of 2.88:5:0.12. The solution dissolved using hot plate and magnetic bar at room temperature for 0.5h. Then citric acid and ethylene glycol were added to the solution. Citric acid and ethylene glycol in a 1:1.5 ratio were employed as gelation reagents, and the molar ratio of all metal ions to citric acid was maintained at 1:3. Then the solution was heated to 130℃ and stirred until the mixture became transparent. After mixing and thoroughly stirring, the solution was evaporated at 300℃ to initiate the gelation reaction. In length of time, the color of the solution changed from pale yellow to bright yellow, and a large number of brownish gases emitted. When the excess solvent was removed, the solution became more viscous without any turbidity or precipitation. At last, a dark brown glassy resin-like substance was observed. After further firing at 300℃, a dry dark solid mass was formed. The precursor powders, obtained by grinding the dry gels, were sintered in a loosely sealed alumina crucible in air. In order to analyze the intermediate products at various temperatures in the sintering process, the precursor powders were sintered in air at 800, 1000℃ for 2h using alumina crucible on the box furnace.

3. Results and Discussion

The crystal chemical purity of the materials was checked by XRD. The XRD patterns of the YAG:Ce phosphor sintered at various temperatures and precursor are shown in Fig.1. We can't see any diffraction peak in the precursor, indicating that the no heating powder remains amorphous. From sintering temperature 800℃, the peaks of YAG cubic garnet phase appeared. It is noted that the sol-gel derived precursor was transformed into the crystalline YAG phase during sintering. YAG phase was crystallized noticeably at sintering temperature 1000℃ for 2h, which was in a good agreement with JCPDS diffraction file 33-0040. It is indicated that sol-gel method is possible to synthesize lower sintering temperature than conventional solid state reaction method.

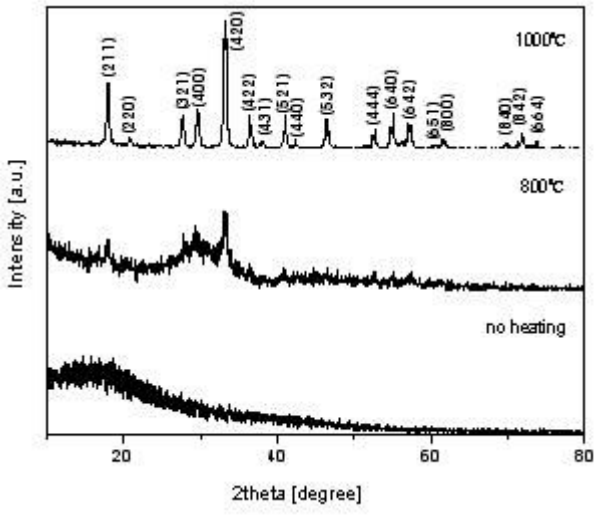

Fig. 1 XRD patterns of the YAG:Ce phosphor

Fig. 2 shows the SEM images of prepared particles at different sintering temperatures of precursor. The surface morphologies of the precursor have rough shape. With increased sintering temperature, phosphor particles have spherical shape and fine size. Uniform and spherical nano particles of YAG:Ce with homogeneous structure were obtained by sol-gel method at 1000℃ for 2h. The mean size of particles measured from SEM image was less than the 100nm.

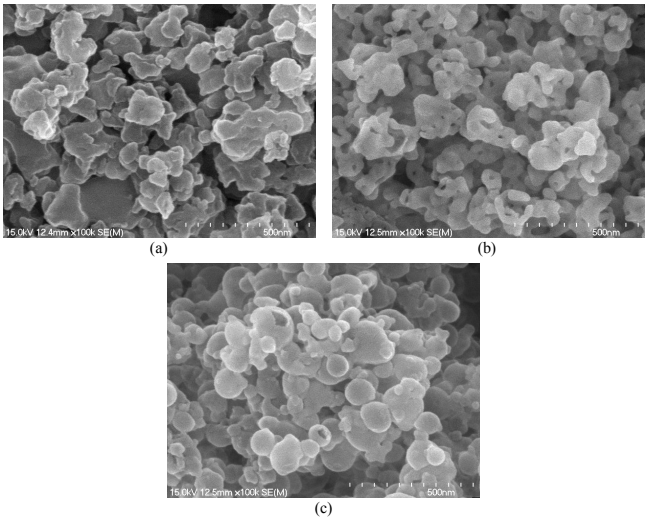

Fig. 2 SEM images of the YAG:Ce phosphor (a) no heating, (b) 800℃, (c) 1000℃

Fig. 3 presents the excitation spectra of the YAG:Ce phosphor sintered at different temperatures. When sintered at 800℃, hardly any excitation band appeared. However, when sintering temperature reaches 1000℃, the excitation band is located at 466nm, respectively. This excitation band is due to the electron transition from the grand state of Ce^{3+} to the different crystal field splitting component of excited 5d state of Ce^{3+}. Obviously, the broad band from 410 to 510nm is most intense, which makes it possible for the phosphors to apply with blue LED.

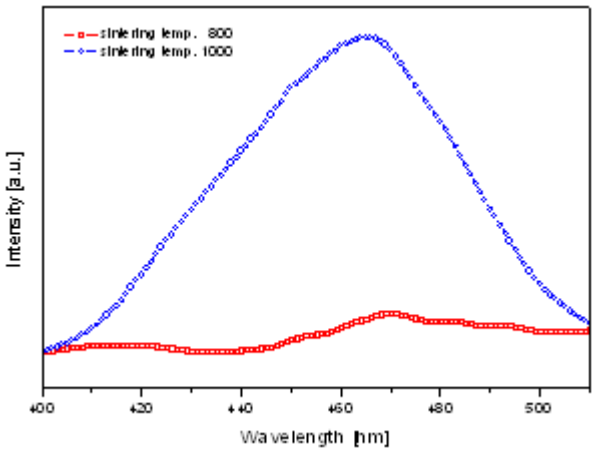

Fig. 3 Excitation spectra of the YAG:Ce phosphor

The emission spectra of the YAG:Ce phosphor sintered at different temperatures are shown in Fig. 4. With increased sintering temperature, emission band showed dramatic increase. There is broad emission band located from 480 to 600nm, which is an ideal yellow secondary light that components the blue light emitted by blue emission to yield white light.

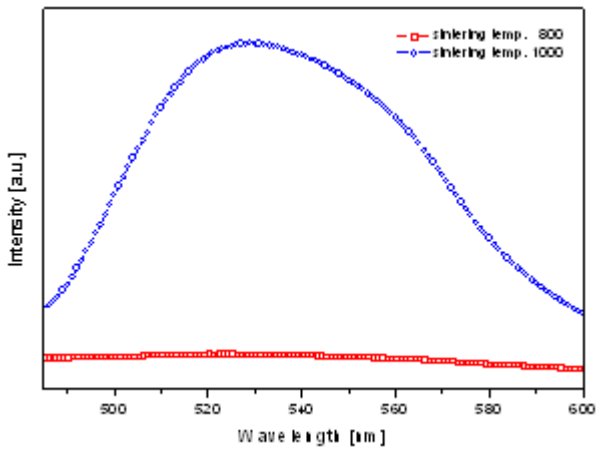

Fig. 4 Emission Spectra of the YAG:Ce phosphor

4. Conclusions

The YAG:Ce phosphors have been synthesized by a sol-gel method and the precursors are sintered using the box furnace. Following are the summary and conclusions of this study:

(1) The sol-gel method was possible to synthesize the lower temperature than the conventional solid state reaction method.

(2) The (420) main peak of YAG:Ce phosphor is increased under the effect of sintering temperature. The crystal property was found to be at its best at sintering temperature 1000℃ and the PL property depends on the crystal property.

(3) The amorphous precursor transformed directly pure phase YAG without intermediate phases such as YAP and YAM. SEM images shown that nano sized particles were of spherical shape.

(4) The best PL properties have been observed at sintering temperature 1000℃ respectively. In the YAG:Ce phosphor, the excitation spectrum located from 410 to 510nm and the emission spectrum located from 480 to 600nm.

These results indicate that YAG:Ce phosphors have wide application as phosphor of the blue excitation and yellow emission for white LED.

References

[1] Y. Pan, M. Wu, and Q. Su, Journal of Physics and Chemistry of Solids, 65 (2004) 845.

[2] X. Li, H. Liu, J. Wang, H. Cui, and F. Han, Materials Research Bulletin, 39 (2004) 1923.