UVU characteristics of YBO_3:Tb green phosphor prepared by spray pyrolysis

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Abstract

We applied the spray pyrolysis technique to prepare fine YBO_3:Tb particles with high photoluminescence, which could be used in the plasma display device as a green phosphor. Several preparation conditions were investigated in order to tail the vacuum ultraviolet characteristics of YBO_3:Tb particles when they were prepared by the spray pyrolysis. As a result, the optimized YBO_3:Tb particles showed the high photoluminescence intensity as well as fine size in comparison with the commercial one.

1. Introduction

Recently, flat penal displays (FPDs) such as plasma display, field emission display, and liquid crystal display have much attention because they can be made in light weight, thin thickness, and large screen size. Especially, plasma display panels (PDPs) are considered as the most potential display applicable to high-definition TV attachable on the wall as a home theater. The phosphor materials are of important parts in the PDPs because they directly influence the brightness and the lifetime [1]. So, much attention is given to design and prepare high efficient phosphor materials.

The phosphor materials for the PDPs should efficiently absorb vacuum ultraviolet (VUV) light emitting from the plasma discharge and transfer the absorbed light energy to the luminescent centers. Rare earth doped borates, silicates, and aluminates are well known as the most effective phosphor matrices. A represent-tative green phosphor using for the plasma display panels (PDPs) is Zn_2SiO_4:Mn which is known to have a little long decay time and high discharging voltage [2]. So, other phosphors such as YBO_3:Tb and $BaAl_{12}O_{19}$:Mn are additionally necessary to optimize the luminance property of PDP [3].

The luminance property of PDP phosphor materials is strongly affected by the preparation method. Most of commercialized phosphor materials being used in the PDPs are made by the solid-state reaction technique. In order to step with the fast growing of PDP technologies, the phosphor materials as a key component determining the quality should be retailed in terms of particle size, size distribution, and morphology. That is, smaller particle size, spherical shape, and higher brightness are progressively required for the fabrication of high quality PDPs. At these points, a new preparation process is needed because the conventional solid-state reaction technique hardly produces the phosphor particles satisfying the above described properties.

Spray pyrolysis has been considered as a promising technique that can produce spherical shaped and fine-size phosphor particles [4-5]. When the spray pyrolysis is applied to make

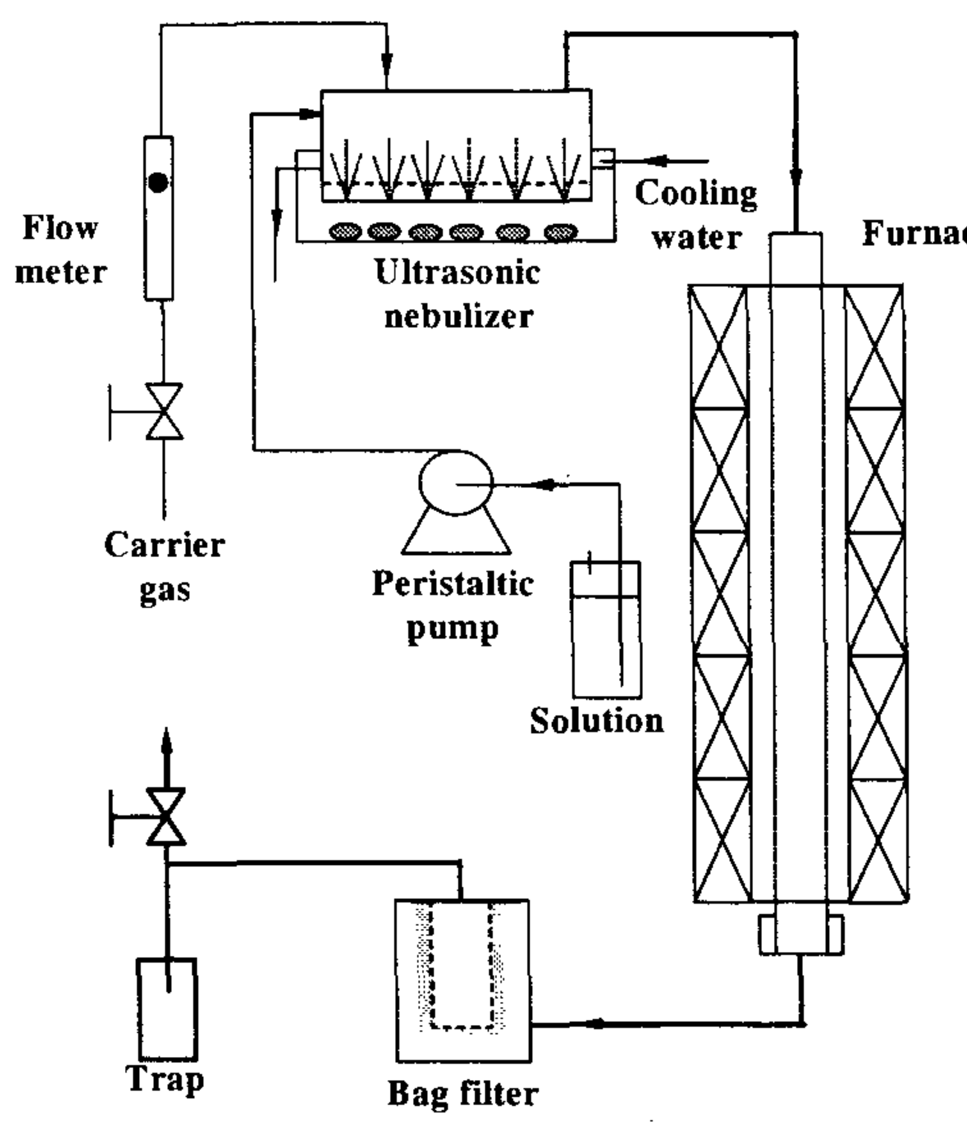

Figure 1. Schematic diagram of the used pilot-scale spray pyrolysis process

borate-type phosphor particles, however, the spherical shape is hardly obtainable due to the boron component of which the melting point is very low and easily evaporated during the post treatment at over 1000 °C. However, the process is much simpler and could easily control the phosphor property in terms of phase purity, particle size and morphology in comparison with the solid-state reaction. So, we applied the spray pyrolysis to make fine YBO_3:Tb particles and tried to improve the luminescent property under VUV excitation.

2. Experimental

Figure 1 shows the used pilot-scale spray pyrolysis process which consists of an ultrasonic aerosol generator with six vibrators (1.7 MHz), a quartz tube (length - 1200 mm and inner diameter - 50 mm), and a particle collector. The flow rate of air used as a carrier gas was 45 L/min. The reactor temperature was fixed at 800 °C, The residence time of droplets inside the reactor was about 0.6 sec.

A nitrate precursor solution to be sprayed was prepared by only dissolving yttrium nitrate, terbium nitrate, and boric acid into purified water. Also, the prepared nitrate solution was chemically modified by adding NH_4OH, so that the added boron turns into polycations. In order to optimize the morphological and luminous properties of $(Y_{1-x}, Tb_x)(BO_3)_{1+y}$ green phosphor particles, the content (x) of Tb activator and the excess quantity (y) of boron were changed.

The crystal phase and morphology of particles prepared were investigated with X-ray diffractometry (XRD) and scanning electron microscopy (SEM), respectively. The photoluminescence characteristics of the prepared particles were measured under vacuum ultraviolet(147nm) by Kr lamp.

3. Results and Discussion

In the first, the luminescent intensity of YBO_3:Tb under VUV (147 nm) excitation was monitored by changing the content of Tb activator in order to find an optimal concentration giving the highest photoluminescence intensity. Figure 2 shows the emission spectra of $(Y_{1-x}, Tb_x)BO_3$ particles prepared by the spray pyrolysis using a nitrate solution, wherein yttrium, terbium nitrate, and boric acid are dissolved with no chemical additive. All samples showed four peaks at around 480, 547, 580, and 620 nm. These peaks are due to the transition of excited electrons from $^5D_4$ to ^7F_j [6]. The highest emission intensity was obtained at 10 mol. % of Tb content (x=0.1).

The as-prepared YBO_3:Tb particles in the spray pyrolysis process have amorphous phase due to the short residence time. So, the post heat treatment is essentially needed to obtain high crystalline. The crystallinity and phase purity of phosphor are known to be crucial factors affecting the photoluminescence intensity. The phase purity of prepared YBO_3:Tb was analyzed by XRD diffraction patterns. Figure 3 shows XRD patterns of commercial and prepared YBO_3:Tb particles which were thermally treated at 1050 °C. It was confirmed that high crystalline was prepared and no impurity was formed. In general, the post heat treatment at higher temperature could produce higher crystallinity, but there is an optimal temperature at which the highest luminescent intensity will be obtained because too high temperature could induces the destruction of crystal phase as well as high agglomeration between individual particles. At this point of view, the emission

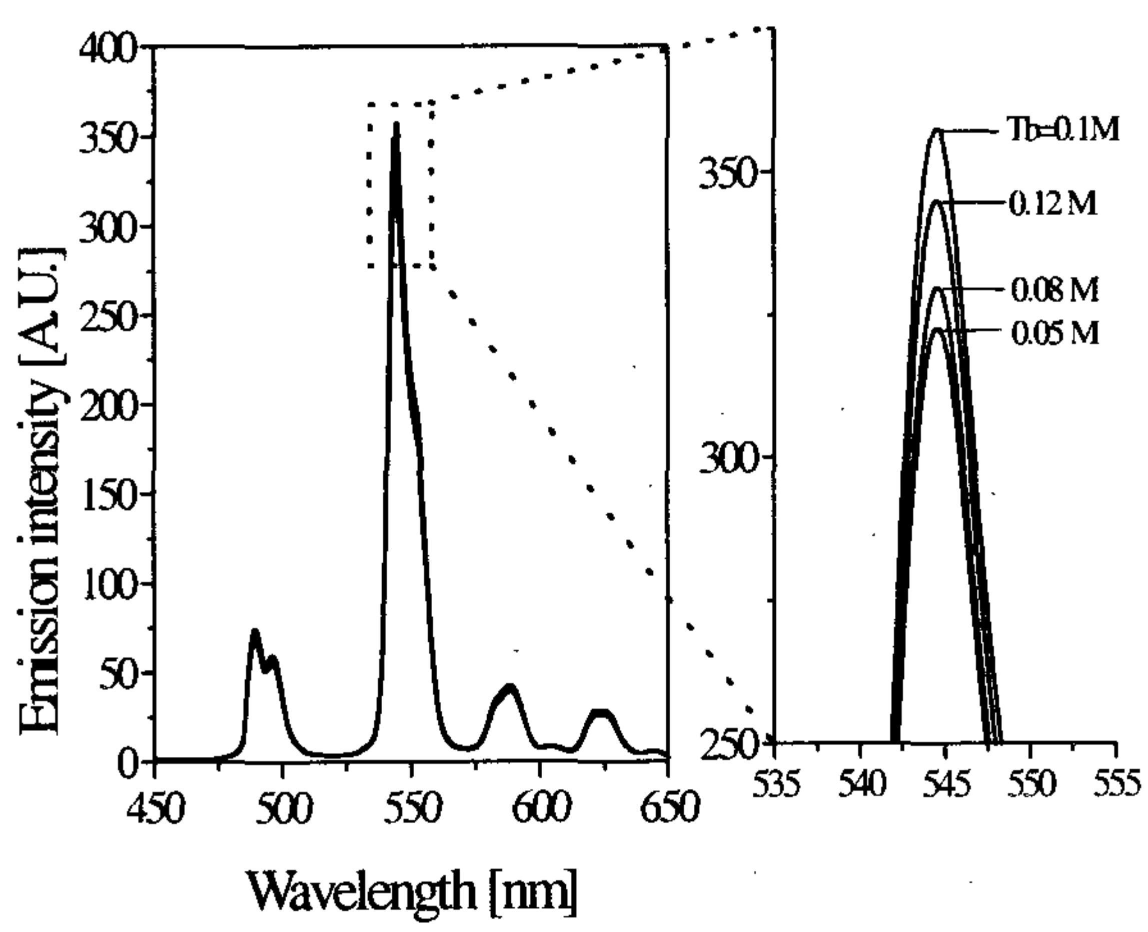

Figure 2. Effect of Tb content on the emission intensity of $(Y_{1-x}, Tb_x)BO_3$ green phosphor particles prepared by spray pyrolysis and post heat treated at 1050 °C.

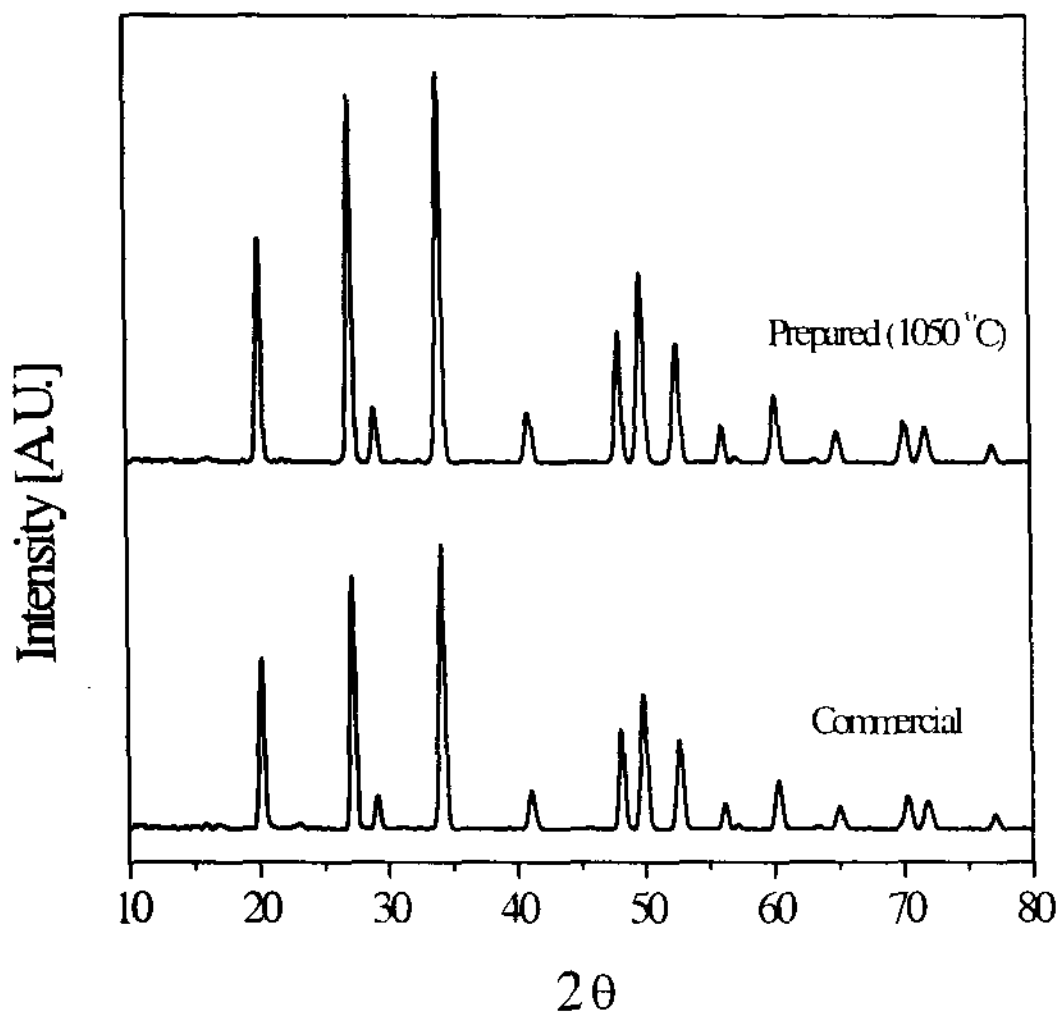

Figure 3. XRD patterns of commercial and prepared YBO_3:Tb particles

intensity of YBO_3:Tb particles prepared by the spray pyrolysis was measured with changing the heat-treatment temperature to find the optimal temperature in terms of VUV characteristics and morphological properties. As a result, it was found that YBO_3:Tb phosphor particles have highest photoluminescence intensity when they were treated at 1150 °C.

Particles prepared by spray pyrolysis generally have a spherical shape, but hollow structure which results in the deformation to a irregular shape and agglomeration during the high temperature post-treatment process. In addition, it is very difficult for borate-type phosphor to maintain complete spherical shape even after the high temperature treatment due to the ductility of boron component. When YBO_3:Tb phosphor particles were prepared by the spray pyrolysis using the nitrate solution, the final obtained particles after the post treatment had an irregular shape like a rod type. So, in this work, the spray solution was modified by NH_4OH in order to control the particle morphology as well as the VUV characteristics. As a result, YBO_3:Tb particles with a spherical-like morphology was obtained by the spray

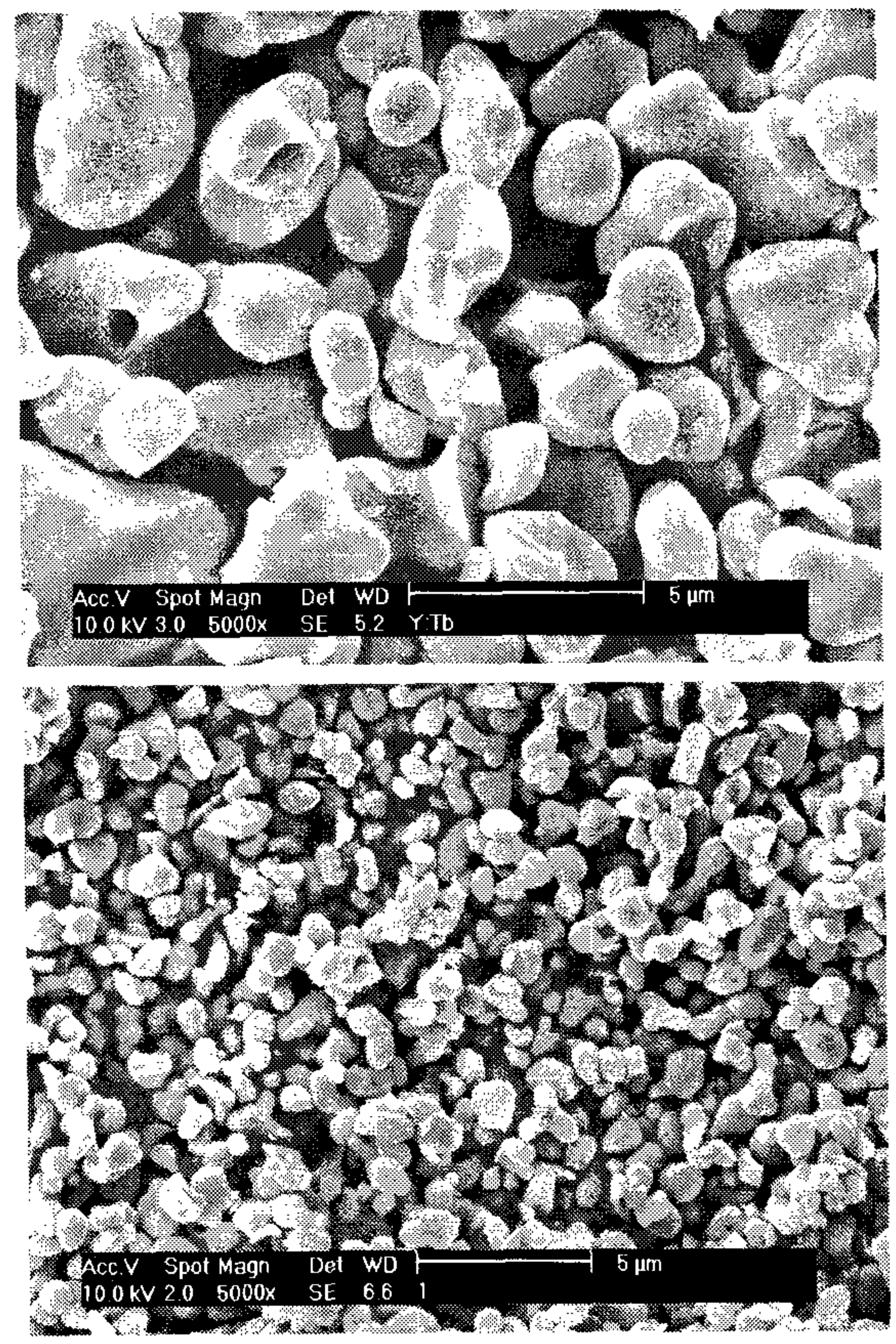

Figure 4. SEM photos of commercial (upper) and prepared (down) YBO_3:Tb

pyrolysis as shown in Figure 4. The optimized YBO_3:Tb particles have fine size and narrow size distribution compared with the commercial one.

Boron is very volatile and could be evaporated during the preparation or post heat-treatment process. Therefore, the excess boron is required to obtain stoichiomatically well established YBO_3:Tb with high luminescent intensity. According to the investigation on the effect of excess boron(y) on the luminous property of YBO_3:Tb phosphor, the highest luminescence intensity under VUV (147 nm) excitation was at about 25 % excess.

Figure 5 shows the photoluminescence spectra obtained by the excitation of vacuum ultraviolet (147 nm) light for the prepared YBO_3:Tb particles at optimal conditions found out. In this work, in order to optimize the PL intensity and the morphological property, the

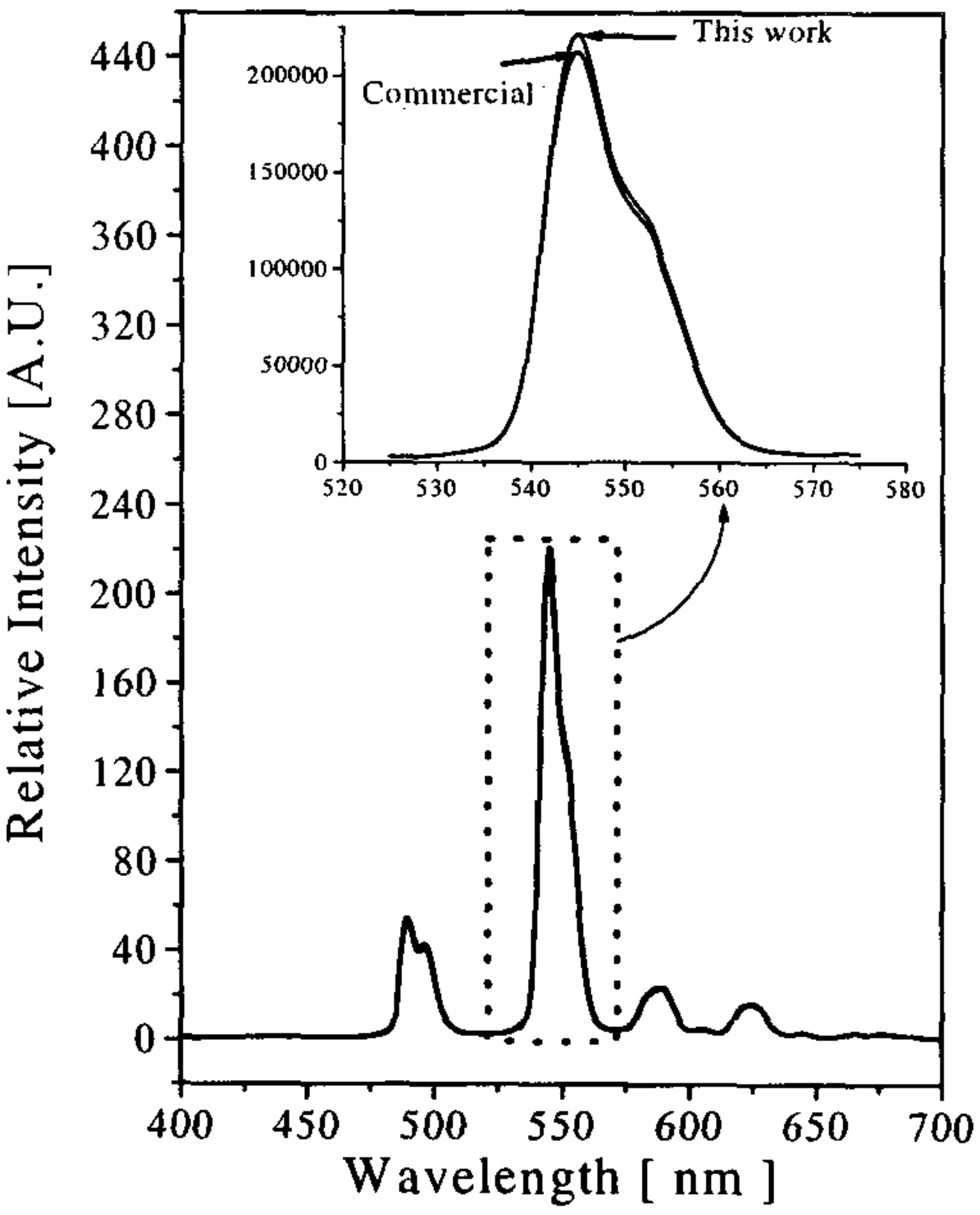

Figure 5. Emission spectra of the YBO_3:Tb particles prepared by spray pyrolysis and the commercial product under VUV excitation (147 nm).

spray solution was chemically modified by the NH_4OH solution. As a result, the PL intensity was about 14 % improved as well as the particle size was smaller than the commercial one. Finally, optimized YBO_3:Tb particles in spray pyrolysis had higher photoluminescence (PL) intensity than that of commercial one. Given that fine size or high luminescent intensity are required for the fabrication of PDPs of high resolution, the YBO_3:Tb particles prepared by the spray pyrolysis are expected to be successfully used as a green phosphor for next-generation PDPs.

4. Conclusions

High luminescence YBO_3 green phosphor with fine size was prepared by spray pyrolysis. In order to improve and optimize the VUV characteristics of the particles, the content of Tb or boron and the heat treatment temperature were optimized. The morphology and luminous property of YBO_3:Tb phosphor particles were successfully improved by modifying the spray solution with NH_4OH. The highest luminescent intensity of prepared $(Y_{1-x}, Tb_x)(BO_3)_{1+y}$ particles under VUV (147 nm) excitation was obtained when the content of Tb (x) and excess quantity of boron(y) were 0.1 and 0.25, respectively. Also, the optimal heat treatment temperature was 1150 °C.

5. Acknowledgements

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6. References

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