Optical Characteristics of Spherical Phosphors for Low-Voltage FED Operation

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Abstract

The aerosol pyrolysis technique was introduced to prepare the spherical phosphors for enhancing low-voltage phosphor efficiency. The density, the shape, and the crystallinity of phosphors were controlled by thermal decomposition temperature and phosphor annealing condition. The particle size was adjusted by the precursor concentration and reactor pressure. It was fond that the efficiency of phosphors synthesized in this work was superior to the commercial products at the low-voltage excitation by 1.5 times and the screen efficiency was also higher than that of any value reported in literature.

Introduction

Field emission displays(FED) are an newer technology that does not have as yet the benefit of any high volume manufacturing experience. Hence there are likely to be critical needs for cost effective manufacturing. General areas of anticipated technology needs could be identified but specific details such as phosphor materials must await the experience of initial trials. The anticipated needs and current challenges for phosphor materials are unique to FED. The phosphor should be efficient enough to guarantee the required luminance at the driving condition and they must be stable. In particular, both high and low voltage FED designs may suffer from outgassing of materials in the vacuum area over time. Phosphors can be a source of outgassing, which is detrimental to the electron emission from microtip. However, the most critical issue in FED manufacturing is the efficiency of phosphors[1].

The luminous efficiency of color phosphor is dependent upon the excitation condition. Generally, the high luminous efficiency was obtained by reducing the current density. Recent activities of FED suppliers give us a hint on what operation may be more realistic. All the suppliers, including Candescent in USA, took high-voltage option. As of our knowledge, only Futaba Co., in Japan has insisted the low-voltage option. In SID'99, Futaba Co., demonstrated a nice panel that operated near 700 anode voltage. We believe that it is obviously related to the manufacturing cost.

Experimental

The aerosol pyrolysis method was used to prepare the spherical phosphors.[2] In particular case of Y_2O_3:Eu, the precursor solution was prepared by dissolving $Y(NO_3)_3 6H_2O$(99.99%) and $Eu(NO_3)_3 6H_2O$(99.99%) into deionized water. The content of europium was adjusted for doping the host lattice and getting the maximal cothodoluminescent intensity. The concentrations of precursor solutions were varied from 0.1 M to 1.8 M. It could control the particle size to be obtained. The precursor solutions were stirred for 3 hours and then made to the liquid drop of aerosols by ultrasonic aerosol generator. The aerosols were thermally decomposed to the spherical yttrium oxide particles in a tubular furnace of above 600°C temperature as shown in Fig. 1. N_2(99.9%) with the velocity of 1 liter/min was used as carrier gas. The reactor temperatures were varied from 600°C to 1000°C. The as-collected yttrium oxide particles were then heat-treated for crystallization and activation at around 1300°C for a few hours. For maximizing the optical intensities, several heating cycles were tried. The particle shapes and crystalline were examined by SEM(Secondary Electron Microscope) and XRD(X-Ray Diffractometer). The CL brightness and color coordinates of Y_2O_3:Eu powder and screen were measured by chromameter(Minolta CS-100) in the vacuum chamber(10^{-6}torr) at 400V, 50μA/cm^2 and DC mode excitation condition.

The synthesized phosphor powders were screened onto ITO glass by electrophoretic deposition method. The film thickness was also optimized for the maximal CL intensity. The screen efficiency was investigated.

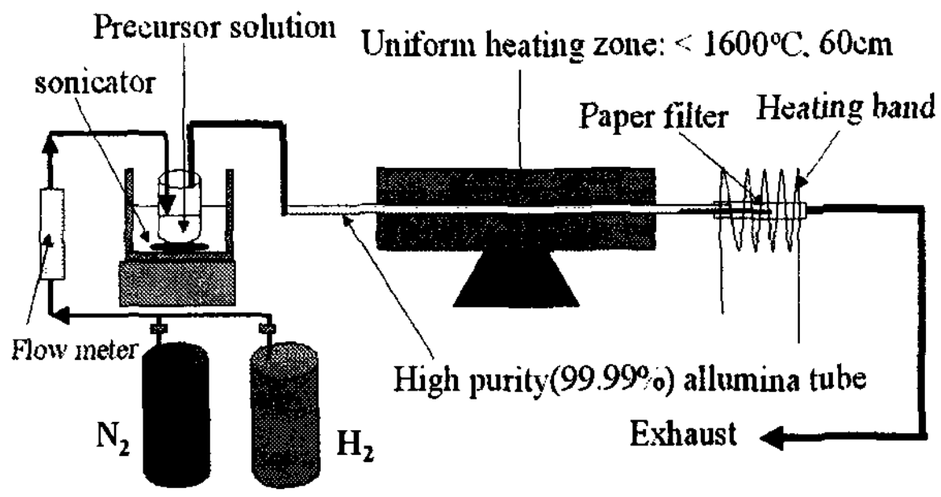

Fig. 1. Schematic diagram of aerosol pyrolysis equipment

Result and Discussion

Our group has developed the synthetic process to prepare spherical phosphors, which named as aerosol pyrolysis method.[5] It is not a conventional diffusion process, but a dynamic process that the phosphors are formed in the nanosecond period from a liquid aerosol. Therefore, the shape and crystal structure of phosphors might be unstable while it has the intrinsic advantages over conventional diffusion process such as easy control of particle size, uniform distribution of activators in host lattice, and automatic formation of spherical shape. In this process, the preparation of precursors, reactor temperature for thermal decomposition, residence time, and annealing conditions are typical process parameters for high quality of spherical phosphors.

Figure 2 shows typical SEM photograph of as-collected Y_2O_3:Eu powder. As aforementioned, the particle size and shape could be controlled. In a reactor furnace, the liquid drops of aerosol are thermally decomposed and getting into spherical shapes of powders due to the surface tension. The reactor temperature should be higher than that of thermal decomposition for given precursors. Thermal decomposition temperature for the precursor of Y_2O_3:Eu phosphors is near 600°C in our case. However, the physical properties of as-collected sample are very sensitive to this temperature. The hollow type and broken shape of spherical shapes were often observed at relatively higher temperature. Also it has

influence on the optical properties as well.

The influence of particle size on the cathodoluminescence was investigated. In our synthetic method, particle size of phosphor was controlled by adjusting the mole concentration of solutes. In our operating condition($\approx$400V), CL intensity shows maximum near $\approx$ 1.3μm and slightly saturated with particle size. Now, CL intensities of our samples were compared with commercial ones. The measurements were done in phosphor technology center of excellence in Georgia Institute of Technology. Obviously, luminous efficiency of the spherical phosphors is found to be superior to the commercial products by 1.5 times, as shown in Fig. 3.

The luminous efficiency of a phosphor under electron beam excitation can be expressed in units of lumens per watt(lm/W), as following

$$\eta(lm/W) = \pi \frac{L(cd/m^2)}{Vi_a Du} \qquad (1)$$

where L, V, i_a, Du indicate the luminance in cd/m^2, anode voltage(V), anode current density(A/m^2), and duty cycle, respectively. We calculated the luminance efficiency of phosphor powder using this equation under above excitation conditions. There can be an inconsistency of data presentation due to the lack of an accepted characterization protocol and standard for display phosphor characterization. Lauren Shea summarized all these controversies well and provided low-voltage efficiency data for red, green, blue powders and screen.[3] As of our best knowledge, the most reliable data on the FED phosphors have been reported by Futaba group in SID annual meetings for last three years.[4]

Now, we prepared the phosphor screen by electrophoretic deposition method. Figure 4 shows the CL intensity from the phosphor screen. It corresponds to 2.0 lm/W. These data were from the vacuum chamber that the vacuum level could be kept as ~ 10^{-6} torr. This value is also higher than the reported value, even though the direct comparison with the data from Futaba is hard in light of excitation conditions. Recently, our group fabricated the test module and examined the optical properties of red phosphor screen. In our case of test module with red phosphor screen, luminous efficiency which we obtained was 1.24 lm/W, of which operating condition were of 400 V anode voltage and 60 V_{p-p} gate voltage inducing an average 30μA/cm^2 emission current density in 15 % pulse mode.[6] This value is also comparable to the best one from Futaba. However, we expect that this value can be increased by substantial amount when optimizing the preparation of phosphor screen as well as synthetic condition.

Summary

Based on the most reliable literature data so far, the optical properties of spherical phosphors were evaluated and their superiority at low-voltage excitation was demonstrated. This superiority may be ascribed to the low-scattering at the stack of spherical phosphors. Another possible factors may be from the unintentional oxygen vacancies and homogeneous activator distribution in host lattice, which are naturally favorable in aerosol pyrolysis method.

Acknowledgment

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References

[1] USDC Roadmap, NEMI, 1998.
[2] S. H. Cho, J. S. Yoo, and J. D. Lee, J. Electrochem. Soc., **145**, 3, 1998.
[3] Lauren E. Shea, The Electrochemical society Interface summer, pp24, 1998.
[4] See SID proceedings published in 1997, 1998, and 1999.
[5] US Patent No. 5,885,492
[6] S. H. Cho, S. H. Kwon, J. S. Yoo, C. W. Oh, J. D. Lee, K. J. Hong, and S. J. Kwon, J. Electrochem. Soc, revision for publication.

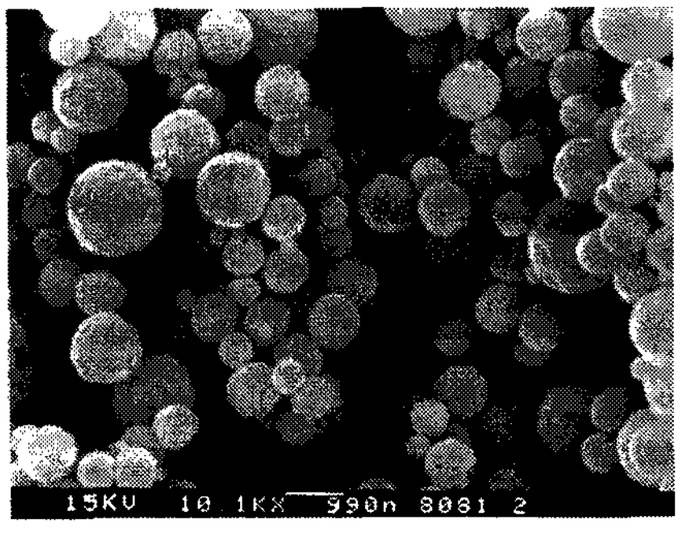

Fig. 2. SEM photograph of as collected Y_2O_3:Eu powder particles at 1100°C prepared by aerosol pyrolysis method.

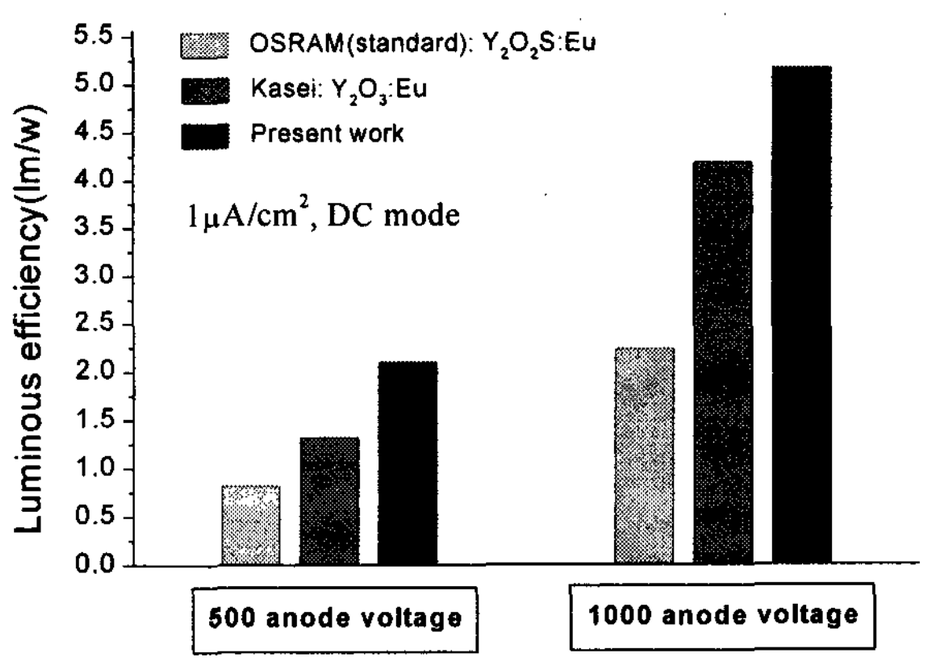

Fig. 3. Comparison of CL intensity of Y_2O_3:Eu powder phosphors.

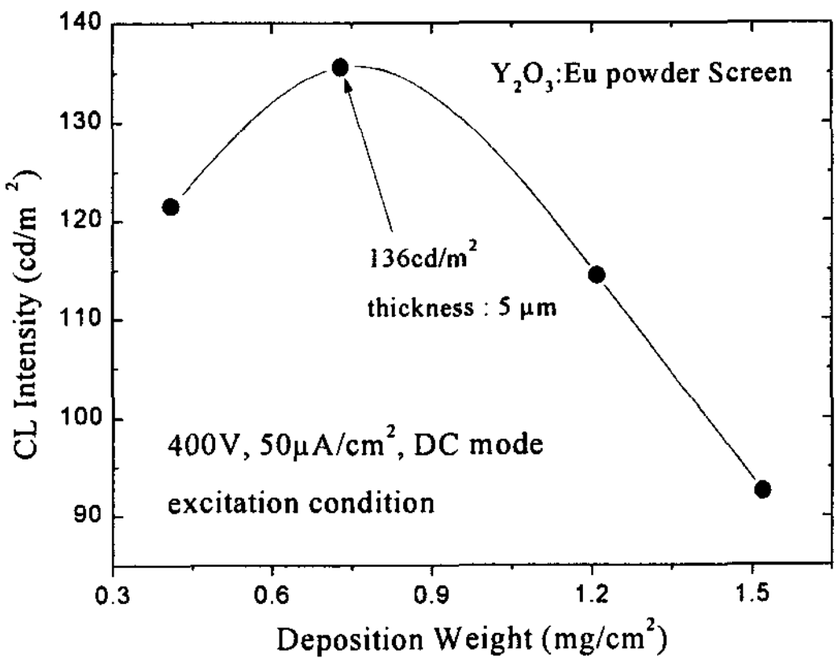

Fig. 4. CL intensity of Y_2O_3:Eu powder screen for the deposition weight.