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Dependence of the performance of the organic electroluminescent devices upon the deposition rate of organic thin films

Y. Qiu *, D.Q. Zhang, J. Qiao, Y. Shao

Department of Chemistry, Tsinghua University, Beijing 100084, People's Republic of China

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Abstract

Typical organic electroluminescent (EL) devices with triphenyldiamine derivative (TPD) and tris-(8-hydroxyquinoline) aluminum (Alq_3) as the hole transport layer and electron transport layer, respectively, have been fabricated. The EL properties as a function of the deposition rate of the organic materials were investigated. It was found that a TPD deposition rate of around 0.2–0.3 nm/s is the optimum for the maximum luminous efficiency. The optimum deposition rate of Alq_3 is 0.3–0.4 nm/s. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: Electroluminescence; Luminous efficiency; Thin film; Deposition rate

1. Introduction

Organic electroluminescent (EL) devices have been extensively studied since Tang and Vanslyke [1] developed a double-layered organic thin-film EL device in 1987. Recently, research of the EL devices has been carried out with the emphasis on the modification of multilayer structure [2–4], realization of multicolor display [5,6] and design of new organic materials [7,8]. To our knowledge, the dependence of EL properties on the film forming conditions has not been sufficiently investigated although the maximum luminance as a function of TPD deposition rate was reported [9]. However, it is not easy to obtain the exact value of the maximum luminance, since the luminance increases with voltage, such an effort always results in the device breakdown.

In this paper, the typical organic EL devices with the structure ITO/TPD/Alq_3/Mg:Ag were fabricated at different deposition rates of TPD and Alq_3, respectively. And the EL properties as a function of the deposition rate were measured. It was found that the film deposition rate is not only related to the luminance at a certain voltage but also to the maximum luminous efficiency. Our results will help to get high luminous efficiency by controlling the film forming conditions during the fabrication process.

2. Experimental

ITO-coated glass was used as the substrate for organic EL device, and the sheet resistance is about 60 Ω/square. The routine cleaning procedure included rubbing in a detergent, rinsing in deionized water, sonication in the mixture of acetone and alcohol and finally drying in a cleaning air flux. All the devices have the same double-layer structure of ITO/TPD/Alq_3/Mg:Ag, where TPD is the hole-transport layer, and Alq_3 is the emissive as well as electron transport layer. All organic layers were prepared by conventional vapor deposition. The thicknesses of TPD and Alq_3 were about 40 and 60 nm, respectively. The vacuum pressure during the deposition was about 2×10^{-3} Pa. In the experiment, devices were fabricated with the deposition rate of the TPD film changed from 0.09 to 0.67 nm/s by changing the source temperature, while the deposition rate of Alq_3 film was kept at 0.1 nm/s.

Similarly, devices were fabricated with the deposition rate of the Alq_3 film changed from 0.03 to 0.7 nm/s while the deposition rate of TPD film was kept at 0.2 nm/s. All the devices were fabricated continuously without breaking the vacuum.

* Corresponding author. Fax: +86-10-662771964.
E-mail address: qiuy@mail.tsinghua.edu.cn (Y. Qiu).

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3. Results and discussion

Current–voltage characteristics of the EL device, with different TPD deposition rates, are presented in Fig. 1. The J–V curve does not shift significantly with increasing the TPD deposition rate. The similarity of the J–V characteristics for several conditions can be understood from the FN tunneling theory [10]. Tunneling current from the electrode is expressed as a function of electric field E and potential barrier height Φ as follows:

$$J \propto E^2 \exp\left[-8\pi(2m^*)^{1/2}\Phi^{3/2}(3\mathrm{qh}E)^{-1}\right]$$

Here, m^* is the effective mass of an electron or hole, Φ is the potential barrier height, and E is the electric field ($E = V/d$, in which V is the applied voltage and d is the thickness of the film). This equation is valid in the case with a triangular barrier shape. Since the thickness of the active organic layers are the same, then J is only a function of V, the voltage applied to the device. Therefore, the J–V characteristics under different conditions were similar.

Although the J–V characteristics are almost similar, the corresponding brightness is quite different in each case (Figs. 2 and 3). Fig. 2 shows the dependence of B–V characteristics on the deposition rate of TPD.

It can be seen that the B–V curves are significantly different due to the different deposition rate of TPD film. The luminance of the devices driven at a certain voltage strongly depends on the deposition rate of TPD film. This result is similar with that reported by Mandai et al. [9]. Fig. 3 shows the relationship between the brightness and current density at different TPD deposition rates. For a given current density, the luminance of device B and C are considerably higher than that of the device A, D and E, showing that they have larger external quantum efficiencies. For example, at a current density of 10 mA/cm^2, the luminance values (expressed in cd/m^2) of the devices are 270 (A), 390 (B), 325 (C), 224 (D) and 185 (E), respectively, indicating different efficiency of hole–electron recombination. The values, reported by Mandai et al. [9], shows that the surface morphology of TPD films is changed considerably by deposition rate, and strongly influences the maximum luminance of the EL device. In our experiment, however, the atomic force microscopy (AFM) observation revealed that the change in the surface morphology of TPD and Alq_3 thin films at different deposition rate is negligible. We hold that the surface morphology change is not responsible for, or at least not the main cause to, the effect of the deposition rate upon the device performance. It is possible that some other properties of the thin films, such as the charge carrier mobility, have been changed due to the change of the condensed state, which is caused by the different deposition rate. Such a result is in agreement with the most recently reported work by Aziz et al. [11] that device aging is not caused by morphological changes in the hole transport layer. The further research on this issue will be published elsewhere.

In our work, it was demonstrated that there also exists a strong dependence of the maximum luminous efficiency

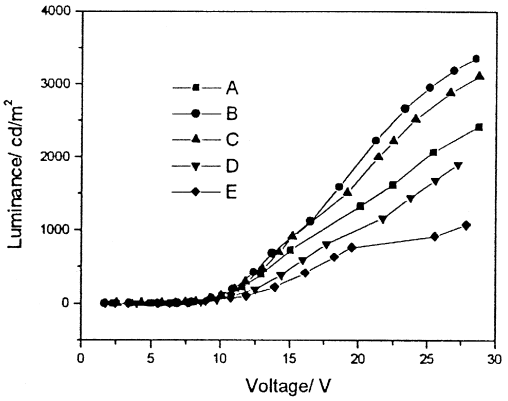

Fig. 2. B–V characteristics of the devices with different TPD deposition rates: (A) 0.07 nm/s, (B) 0.2 nm/s, (C) 0.3 nm/s, (D) 0.45 nm/s, (E) 0.67 nm/s.

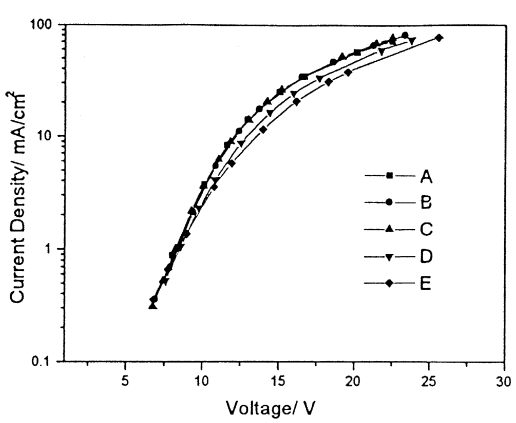

Fig. 1. J–V characteristics of the devices with different TPD deposition rates: (A) 0.07 nm/s, (B) 0.2 nm/s, (C) 0.3 nm/s, (D) 0.45 nm/s, (E) 0.67 nm/s.

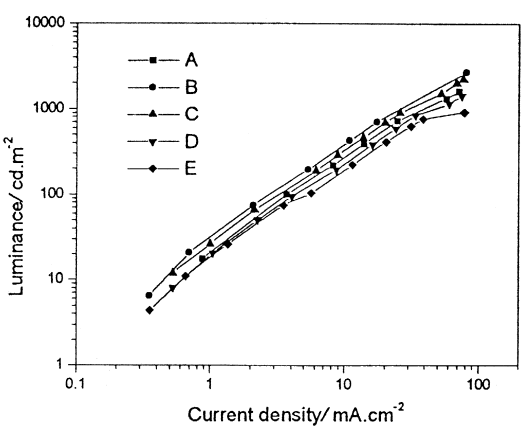

Fig. 3. B–J characteristics of the devices with different TPD deposition rates: (A) 0.07 nm/s, (B) 0.2 nm/s, (C) 0.3 nm/s, (D) 0.45 nm/s, (E) 0.67 nm/s.

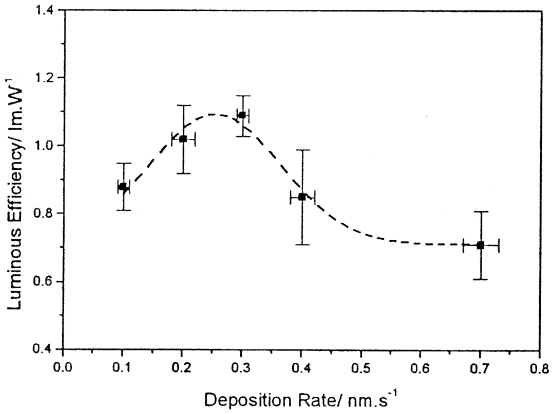

Fig. 4. Relationhip between maximum luminous efficiency and the deposition rate of TPD.

on the TPD deposition rate (Fig. 4). The luminous efficiency η (lm/W) is obtained according to the formula: η (lm/W) $= \pi B$ [cd/m^2]/VJ [W/m^2], where B is the brightness, V is the voltage and J is the current intensity. The η (lm/W) value changes with bias voltage and always reaches its maximum at a relative low voltage. Fig. 4 shows the relationship between maximum luminous efficiency and the deposition rate of TPD. Five devices were made at each point, and then the mean and the standard deviation were obtained which are shown in Fig. 4. The same result can be obtained from Figs. 3 and 4, i.e., the TPD deposition rate of ca. 0.2–0.3 nm/s is optimum for obtaining high performance devices.

In another experiment, when the deposition rate of the Alq_3 film was changed from 0.03 to 0.7 nm/s with the deposition rate of TPD film kept at 0.2 nm/s, the B–J curves of the devices (Fig. 5) and the relationship between maximum luminous efficiency and deposition rate (Fig. 6). At each point, five devices were made for the mean and the standard deviation indicate that the optimum deposition rate of Alq_3 film is 0.3–0.4 nm/s. It is interesting to note

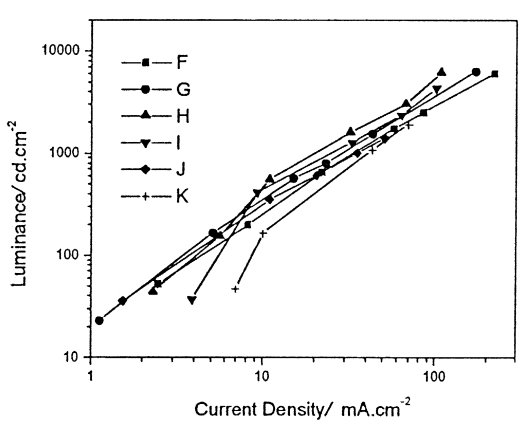

Fig. 5. B–J characteristics of the devices with different deposition rates of Alq_3: (F) 0.03 nm/s, (G) 0.09 nm/s, (H) 0.2 nm/s, (I) 0.27 nm/s, (J) 0.52 nm/s, (K) 0.68 nm/s.

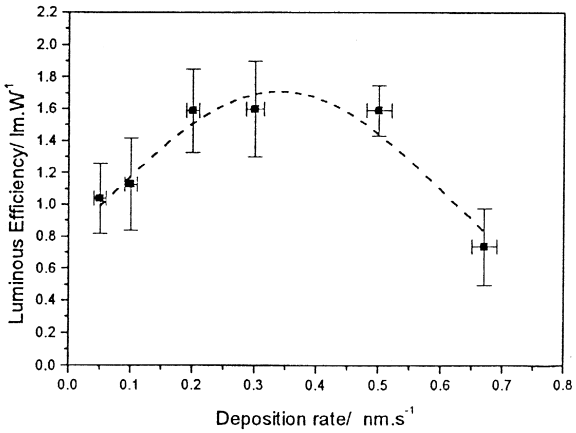

Fig. 6. Relationhip between maximum luminous efficiency and the deposition rate of Alq_3.

that the optimum deposition rate of Alq_3 is a bit larger than that of TPD.

4. Conclusion

It was revealed that the EL characteristics including the B–V curves, the B–J curves, and the maximum luminance efficiencies change with the deposition rate of TPD and Alq_3 films. It was found in our experiment that the optimum deposition rate of TPD film is around 0.2–0.3 nm/s and the optimum deposition rate of Alq_3 film is around 0.3–0.4 nm/s.

Acknowledgements

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