

All-solution-process Red Phosphorescent and Blue Fluorescent Organic Light-emitting Diodes by blade coating

Yu-Fan Chang¹, Hsin-Fei Meng², Hsiao-Wen Zan¹, Hao-Wu Lin³, Ken-Tsung Wong⁴, Heh-Lung Huang⁵, Sheng-Fu Horng⁶

¹ Department of Photonics, Institute of Electro-Optical Engineering, National Chiao Tung University, Hsinchu, Taiwan
Phone: +886-9-39959317 E-mail: klorb@gmail.com

² Institute of Physics, National Chiao Tung University, Hsinchu, Taiwan
Phone: +886-3-5731955 E-mail: meng@mail.nctu.edu.tw

³ Department of Materials Science and Engineering, National Tsing Hua University, Hsinchu, Taiwan

⁴ Department of Chemistry, National Taiwan University, Taipei, Taiwan

⁵ e-Ray Optoelectronics Technology Co., Ltd.

⁶ Department of Electrical Engineering, National Tsing Hua University, Hsinchu, Taiwan

Abstract

Organic small-molecule materials that were designed for thermal evaporation were used to fabricate all-solution-processed organic light-emitting diodes (OLEDs). We developed a processing method to use these organic materials at a solubility of merely 0.1wt%, as in common organic solvents. In this study, we fabricated high efficiency all-solution-process red phosphorescent and blue fluorescent OLEDs. The red Phosphorescent OLEDs exhibited maximal efficiencies with 16.73 cd/A, 16.13 lm/W and 4.89%. In addition, Alq₃ was practically applied in blue fluorescent emitting devices and the efficiencies of the devices are 4.74 cd/A, 1.74 lm/W and 2.01%.

1. Introduction

In the last 20 years organic light-emitting diodes (OLEDs) based on small molecules evaporating in a vacuum have exhibited continuously improving efficiency and are now a promising technology for solid-state lighting and displays. The performance of vacuum processed OLEDs with multi-layer structures for charge balance and low driving voltage surpasses that of solution-processed OLEDs that usually involve conjugated polymers. In addition to featuring an optimized multilayer structure, small molecules have the advantages of higher purity, more flexibility in molecular design, and the absence of variation caused by molecular weight distribution compared with polymers. To reduce the high cost of large-area vacuum deposition, there has been a growing effort to fabricate small molecule OLEDs using a solution process. To manufacture OLEDs, the industry presently adopts a vacuum evaporation process, which requires expensive vacuum chambers and has low material use. Although the lifetime of the devices made using a vacuum process is acceptable, the cost of this process is considerably high. The processing cost can be considerably reduced through adopting solution coating; however, the lifetime of OLEDs manufactured through solution coating is low and few related studies have been published. In the application of products, sufficient operational lifetime of devices made using a solution coating process is critical.

Almost all small molecules can be blade-only coated if the solubility is a mere 0.1 wt%, as in certain general organic solvents. Therefore, blade coating enables realizing an exact optimized vacuum-deposited OLEDs structure through a solution coating process without any chemical modification. Redesigning materials and device structures is unnecessary. Such a fabrication platform solves the dilemma of balancing performance and cost in the production of OLEDs.

2. Experiment

Thin film and device fabrication

Figure 1 shows the procedure of the blade-only method. All the organic layers of the OLED devices in this study were fabricated using blade-only coating. We used a pipette to deliver the solution to the gap between the blade and the substrate. The organic solution was spread in the gap through capillary action. Subsequently, the blade was driven and the wet film completed the formation. Finally, we used hot wind and a hotplate to dry the wet film. The wet film rapidly formed a dry film. The processing details regarding the blade-only method have been previously reported. [1] The red emitter PER54 of the phosphorescent device was provided by e-Ray Optoelectronics Technology Co., Ltd. The devices exhibited co-dopants BUBD-1 and p-bis(p-N,N-diphenyl-aminostyryl)benzene (DSA-Ph) in a stable high-band gap host 2-Methyl-9,10-bis(naphthalen-2-yl)anthracene (MADN). [2, 3] The LiF (0.8 nm) / Al (100 nm) cathode was formed through thermal evaporation in a vacuum (3×10^{-6} Torr).

The device structures were as follows:

Device A: ITO (270 nm)/PEDOT:PSS(40 nm)/NPB(30 nm)/NPB:45%26DCzPPy:10%PER54(30 nm)/TPBI(35 nm)/LiF(0.8 nm)/Al(100 nm).

Device B: ITO(270 nm)/PEDOT:PSS(40 nm)/VB-FNPD(30 nm)/MADN:3%BUBD-1:0.1%DSA-Ph(50 nm)/TPBI(30 nm)/LiF(0.8 nm)/Al(100 nm).

Device C: ITO(270 nm)/PEDOT:PSS(40 nm)/VB-FNPD(30 nm)/MADN:3%BUBD-1:0.1%DSA-Ph(50 nm)/Alq₃(10 nm)/LiF(0.8 nm)/Al(100 nm).

The electroluminescence characteristics of the OLEDs were measured using Keithley 2400 and PR655 SpectroScan spectrometers.

3. Results and Discussion

The red emitter PER54 was designed for vacuum deposition, and the performance was high. We used this emitter to fabricate the all-solution-process OLEDs and achieved satisfactory efficiencies with 16.73 cd/A, 16.13 lm/W, and 4.89%. Alq₃ is not easily dissolved in general organic solvents. In this study, we use methanol as a solvent for Alq₃. Because the concentration was 0.1wt%, Alq₃ was not completely dissolved, and material deposited on the bottom of the solution remained. Therefore, we used a Teflon filter to filter out the dissolved material and obtain a clear solution. Subsequently, we used the clear solution to complete the thin film through blade coating. Then, we used blade coating and spin coating to complete the film coating of Alq₃. Simultaneously, we used Atomic force microscopy to observe various methods for forming the film surface. Obviously we blade coating film surface coating of its Rms = 1.18 nm, on the contrary, by the spin coating a thin film coating, the surface membrane of a clear projection and Rms = 34.9 nm. Subsequently, we applied this Alq₃ in blue fluorescent OLEDs, and achieved favorable characteristics. 2,2',2''-(1,3,5-Benzotriazolyl)-tris(1-phenyl-1-H-benzimidazol e) (TPBI) is commonly used in OLEDs to achieve high efficiency. However, previous studies have shown that TPBI exhibits problems related to stability.[4] Alq₃ is relatively stable but difficult to deposit using a solution process because of its extremely low solubility in common organic solvents resulting from the symmetrical molecular structure. In this study, Alq

was successfully deposited through blade coating at a considerably low concentration and was applied in blue fluorescent OLEDs. The manufactured device exhibited a peak current efficiency of 4.74 cd/A and a power efficacy of 1.74 lm/W. The results demonstrated an effective method to replace instable ETLs by more stable ETLs that exhibit low solubility. The large area device of the blue fluorescent OLEDs is shown in Figure 4. The illuminated area was 3 × 4 cm.

3. Conclusions

In summary, we developed a simple technique for fabricating all-solution-processed red phosphorescent OLEDs and blue fluorescent OLEDs. Only low solubility is required for the deposition, and the method was successful for most common unmodified small molecules. We fabricated all-solution-processed red phosphorescent OLEDs with high current efficiency (16.73 cd/A) and blue fluorescent OLEDs with favorable characteristics.

Figures and Tables

Table I EL characteristics of all-solution-processed OLEDs

Device	Current efficiency (cd/A)	Power efficiency (lm/W)
A	16.73	16.13
B	5.75	2.9
C	4.74	1.74

Maximal efficiency

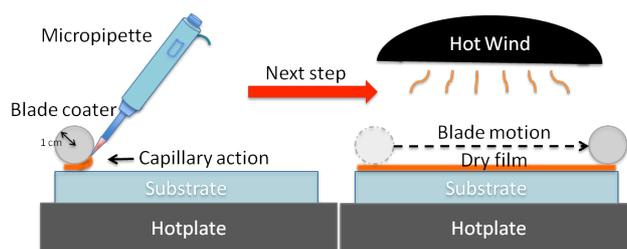


Fig. 1 The procedure of the blade-only method.

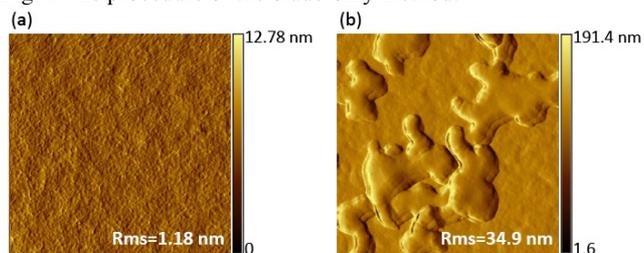


Fig. 2 AFM image of the Alq₃ film by (a) blade coating (Rms = 1.18 nm) and (b) spin coating (Rms=34.9).

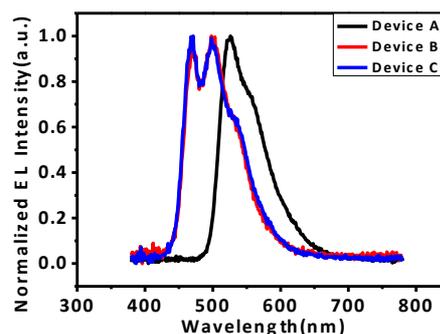


Fig. 3 The normalized EL spectrum of the all-solution-processed OLEDs.



Fig. 4 The picture of operated blue fluorescent device.

Acknowledgements

This work is supported by National Science Council of Taiwan under Contract NSC 101-2112-M-009-006-MY3.

References

- [1] C. Y. Chen, H. W. Chang, Y. F. Chang, B. J. Chang, Y. S. Lin et al., *J. Appl. Phys.* 2011, 110, 094501.
- [2] M. T. Lee, H.H. Chen, C. H. Liao, C. H. Tsai and C, H, Chen, *Appl. Phys. Lett.* **2004**, 85, 3301.
- [3] M. F. Lin, L. Wang, W.H. Wong, K. W. Cheah, H. L. Tam et al., *Appl. Phys. Lett.* **2006**, 89, 121913.
- [4] R. C. Kwong, M. R. Nugent, L. Michalski, T. Ngo, K. Rajan et al., *Appl. Phys. Lett.* **2002**, 81,162.