# Fine-tuning the thicknesses of organic layers to realize high-efficiency and long-lifetime blue organic light-emitting diodes*

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By using *p-bis*($p-N$, *N*-diphenyl-aminostyryl)benzene doped 2-tert-butyl-9, 10-*bis*-$\beta$-naphthyl)-anthracene as an emitting layer, we fabricate a high-efficiency and long-lifetime blue organic light emitting diode with a maximum external quantum efficiency of 6.19% and a stable lifetime at a high initial current density of 0.0375 A/cm$^2$. We demonstrate that the change in the thicknesses of organic layers affects the operating voltage and luminous efficiency greater than the lifetime. The lifetime being independent of thickness is beneficial in achieving high-quality full-colour display devices and white lighting sources with multi-emitters.

**Keywords:** organic light-emitting diode, blue emission, lifetime, organic layers thickness

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## 1. Introduction

Organic light-emitting diodes (OLEDs) have attracted much attention, for they possess the advantages of easy fabrication, low driving voltage, and high luminance. The high-efficiency and long-lifetime OLEDs are expected to serve as the next generation flat panel display devices and light sources.[1−5] Development of primary red–green–blue emitters remains an important challenge to realizing full-colour display and lighting. In particular, it is difficult to generate high-performance pure blue emission because the intrinsically wide band-gap makes it hard to inject charges into emitting materials. As a result, the efficiency and lifetime of blue-light-emitting devices are usually not as good as those of their green or red counterparts.[6,7]

Of various blue emitting materials, distyrylarylene derivatives were regarded as one of the most promising materials, however, the maximum luminous power efficiency obtained has been only 1.5 lm/W due to a lack of suitable host materials.[8−10] Shi and Tang[11] developed another major blue emitter, utilizing the diphenylanthracene derivative 9, 10-*di*(2-naphthyl)anthracene (ADN) as host and 2, 5, 8, 11-tetra-(*t*-butyl)-perylene (TBP) as dopant to obtain a maximum current efficiency of 3.5 cd/A. However, the efficiencies of these OLEDs were not high enough for blue fluorescence emitting. In 2004, Lee *et al.*[12] reported on a new blue OLED in which a new material was utilized based on ADN, named 2-methyl-9, 10-*di*(2-napthyl)anthracene (MADN) as host, a maximum current efficiency of 9.7 cd/A was obtained by suitably choosing *p-bis*($p-N$, *N*-diphenyl-aminostyryl)benzene (DSA-ph) as dopant. Since that time, researchers have made a further study of ADN derivatives to improve the efficiency and thin-film morphological stability,[13] for example, 2-tert-butyl-9, 10-*bis*-($\beta$-naphthyl)-anthracene (TBADN) was possessed of a deeper blue emission of Commission Internationale d'Eclairage (CIE) (0.13, 0.19) and higher

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glass-transition temperature ($T_g$) of 126 °C.[14]

It has been reported that the change in the thickness of the organic functional layer in the device can affect the electroluminescence (EL) properties of the OLED.[15] However, there are few studies of the relationship between the operation lifetime and the thicknesses of different organic layers in the device. Therefore, in this work, we first are concerned with a blue OLED with an emitting layer (EML) of TBADN doped with DSA-ph. Then we particularly investigate the effect of thickness variations of the organic functional layers on the operational lifetime of the OLED, comparing with the same effect on EL properties.

## 2. Experiment

The device structure of the blue OLED is as follows. IZO (220 nm)/2-TNATA: F4-TCNQ 2 wt% ($X$ nm)/NPB(10 nm)/TBADN: DSA-ph 5 wt%($Y$ nm)/Alq(20 nm)/LiF(0.6 nm)/Al(100 nm), where indium zinc oxide (IZO) and aluminum (Al) are, respectively, the anode and cathode, the organic layer stack consists of 2 wt% of tetrafluorotetracyanoquinodimethane (F4-TCNQ) doped 4, 4, 4-{*N*-(2-naphthyl)-*N*-phenylamino}-triphenylamine (2T-NATA) as the hole injection layer (HIL), 1, 4-*bis*[*N*-(1-naphthyl)-$N'$-phenylamino]-4, 4′ diamine (NPB) as the hole transport layer (HTL), 5 wt% of DSA-ph doped TBADN as the emitting layer (EML), *tris*-(8-hydroxyquinoline)aluminum (Alq) as the electron transport layer (ETL), and lithium fluoride (LiF) as the electron injection layer (EIL). All organic layers and the Al cathode were deposited by high vacuum ($10^{-4}$ Pa) thermal evaporation onto a clean glass substrate precoated with a 220-nm thick IZO layer which had been UV-ozone treated before the evaporation process. The layer thickness was controlled *in situ* using a quartz crystal monitor. We varied the HIL and EML thickness simultaneously while keeping the total thicknesses of two layers constant. Accordingly, we name these five blue OLEDs Devices 1 through 5. After the organic layers and metal depositions, the devices were encapsulated in the glove box with $O_2$ and $H_2O$ concentrations both being below 1 ppm.

The current–voltage–luminescence characteristics were measured by a Keithley 2400 source meter and a PR-650 luminance colour meter. The luminance and spectra of each device were measured toward a vertical orientation for the substrate. The lifetime of the device was tested by the lifetime test meter developed in our department. The details of the test procedure were as follows. We drove all of the OLEDs at a constant current density of 0.0375 A/cm$^2$, and at a room temperature of 25.0 °C. Furthermore, the electroluminescent properties of our OLEDs (e.g., voltage, current, and luminous power efficiency) were tested at different current densities of 1.0 A/cm$^2$, 0.1 A/cm$^2$, and 0.01 A/cm$^2$, respectively, in regular time intervals, and the effective emitting area was 2 mm ×2 mm. The fundamental characteristics of each device are summarized in Table 1.

**Table 1.** Layer setup, EL performance, and chromaticity for each investigated sample.

| Device | Thickness of HIL/nm | Thickness of EML/nm | Voltage/V @ 1000 cd/m$^2$ | Max EQE/% | CIE @ 1 A/cm$^2$ | |
|---|---|---|---|---|---|---|
| | | | | | $x$ | $y$ |
| 1 | 95 | 15 | 4.1 | 5.26 | 0.175 | 0.319 |
| 2 | 85 | 25 | 4.7 | 5.88 | 0.179 | 0.336 |
| 3 | 75 | 35 | 5.4 | 6.19 | 0.185 | 0.348 |
| 4 | 60 | 50 | 6.3 | 5.71 | 0.193 | 0.369 |
| 5 | 40 | 70 | 8.0 | 4.21 | 0.218 | 0.429 |

## 3. Results and discussion

### 3.1. Effect of thickness variations of organic layers on electroluminescence characteristics

Figure 1 shows the current density–voltage ($J$–$V$) and luminance–voltage ($L$–$V$) characteristics of the blue OLEDs. With HIL thickness decreasing or EML thickness increasing, the operating voltage increases clearly. The operating voltages are 4.1 V, 4.7 V, 5.4 V, 6.3 V, and 8.0 V at a luminance of 1000 cd/m$^2$ for Devices 1–5, respectively (see Table 1). There are two factors contributing to this change of operating voltage with the variations of the thicknesses of the organic layers. The p-type F4-TCNQ doped 2T-NATA layer as a HIL has a conductivity of approximately $10^{-5}$ S/cm and forms a quasiohmic con-

tact with IZO.[16] Numerical simulations reveal that lower barrier height between the IZO and the HIL and higher mobility of the HIL are beneficial to a lower turn-on voltage when a thicker HIL than HTL is used.[17] On the other hand, the increase of the thickness of the EML gives rise to an increase in the electric field applied to the EML, resulting in a higher operating voltage.[18]

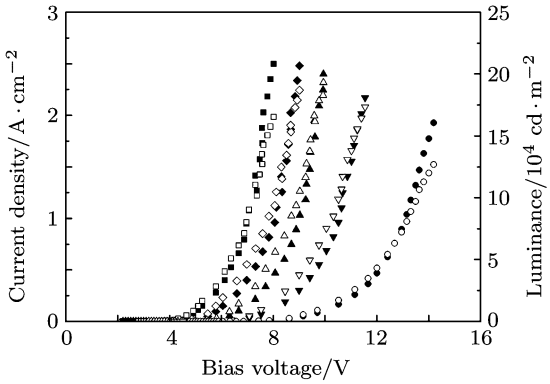



**Fig. 1.** The $J$–$V$ curves (solid symbols) and $L$–$V$ curves (open symbols). Square, rhombus, triangle up, triangle down, and circle symbols represent Devices 1–5 respectively.

Figure 2 exhibits the external quantum efficiencies (EQEs) each as a function of current for Devices 1–5. We find that EQEs increase before the current density reaches about 0.1 A/cm2 and then roll off. A maximum EQE of 6.19% at the high luminance of about 14000 cd/m$^2$ is achieved for Device 3. The higher efficiency in Device 3 than in all other devices is attributed mainly to the optimized hole–electron pair balance. In addition, the thicknesses of the HIL and EML in Device 3 cause an improved light out-coupling compared with in the other devices.

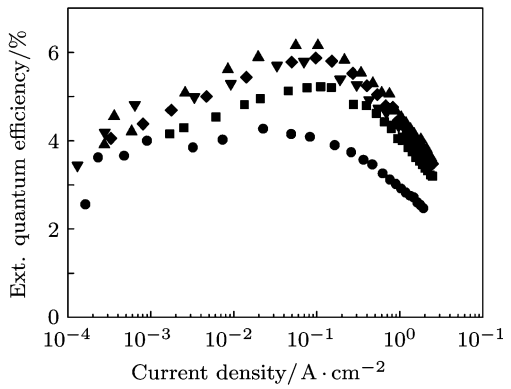



**Fig. 2.** EQEs measured for Devices 1–5 each as a function of current. Solid square, rhombus, triangle up, triangle down, and circle symbols represent Devices 1–5 respectively.

The EL emission spectra at a current density of 1.0 A/cm$^2$ of the blue OLEDs are shown in Fig. 3. The intense peak at 470 nm accompanied by vibronic sidebands at 504 nm and about 540 nm of EL emission spectra for Devices 1–5 are observed, while the intensities of two sidebands increase with the increase of the thickness of the EML, especially for Device 5. This phenomenon is attributed to the optical interference effect in the low-energy wavelength region as the thickness of the EML increases.[19] The maximum EQE and detailed CIE coordinates of each device with different thicknesses of HIL and EML are summarized in Table 1.

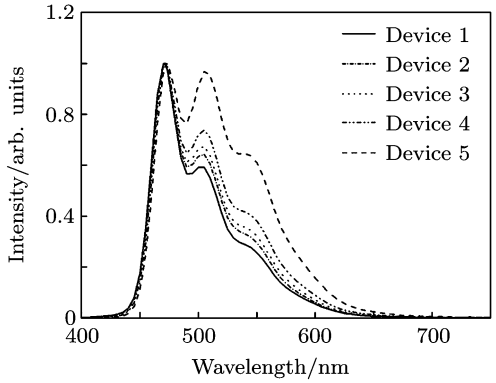



**Fig. 3.** Electroluminescence spectra of Devices 1–5 at a current density of 1 A/cm$^2$.

## 3.2. Effect of thickness variations of the organic layers on device lifetime

The lifetime of the OLED is affected by many factors except encapsulation and operating circumstance, such as ion migration, carrier accumulation at the interface, stability of material, etc. OLED lifetime could be increased by sublimation cleaning of all organic materials used[20,21] and using materials which have high glass-transition temperatures and are stable against charge carriers and excitons.[22] However, there are few reports on the role of the thicknesses of the organic functional layers.

Figure 4 shows that the relative luminance drops off and relative voltage rises with continued operation time. We can see that the trends of luminance drop and voltage rise at the same test current density of 1.0 A/cm$^2$ are almost the same, although the initial applied voltages are different, which are 7.0 V, 7.9 V, 8.9 V, 10.4 V, and 12.9 V for Devices 1–5, respectively.

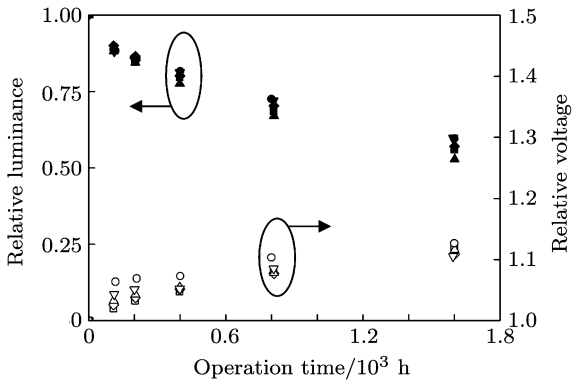



**Fig. 4.** Device operation stabilities for Devices 1–5 at a test current density of 1.0 A/cm$^2$. Solid symbols represent relative luminance (normalized) drop while open symbols denote relative voltage (normalized) rise. Square, rhombus, triangle up, triangle down, and circle symbols represent Devices 1–5 respectively.

We define a relative rise in voltage ($\Delta V$) with respect to the initial operating voltage ($V_0$) as RV and relative drop in luminance ($\Delta L$) with respect to the initial luminance ($L_0$) as relative luminance (RL), respectively, to investigate the temporal degradation process under a constant test current density of 1.0 A/cm$^2$. After operation for 1600 h, the values of $\Delta V$ are 0.819 V, 0.865 V, 1.032 V, 1.095 V, and 1.633 V, while the values of relative voltage (RV) are 11.70%, 10.87%, 11.60%, 10.51%, and 12.65% for Devices 1–5, respectively. With regard to the luminance change, the values of RL are obtained to be 44.10%, 42.97%, 47.22%, 40.26%, and 40.40%, exhibiting the same tendencies for all devices.

The degradation processes for Devices 1–5 at test current densities of 0.01 A/cm$^2$ and 0.1 A/cm$^2$ are illustrated in Fig. 5. The RV and RL exhibit almost the same values in Devices 1–5 although their EML thicknesses vary from 15 nm to 70 nm (or HIL thicknesses vary from 95 nm to 40 nm). This same trend for each device demonstrates that the lifetime of the OLED is hardly influenced by the thicknesses of organic layers.

Owing to almost the same degradation processes for Devices 1–5, we investigate Device 4 as an example for observing practical operation lifetime defined as the half-value time of the luminance for a constant current density. For Device 4, an initial luminance of 4500 cd/m$^2$ (Fig. 1) and a half-lifetime of 1600 h (Fig. 4) are obtained at a constant current density of 0.0375 A/cm$^2$. It is known that the relationship between lifetime and initial luminance is expressed by the equation: $t(L_0) = t(L_1) \times (L_1/L_0)^n$, in which $t(L_0)$ and $t(L_1)$ are lifetimes measured at initial luminance of $L_0$ and $L_1$ respectively, and $n$ is an acceleration coefficient whose value is generally between 1.5 and 2. Assuming that $n$ is 2, we obtain the lifetimes of 129600 h and 32400 h at the initial luminance of 500 cd/m$^2$ and 1000 cd/m$^2$, respectively.

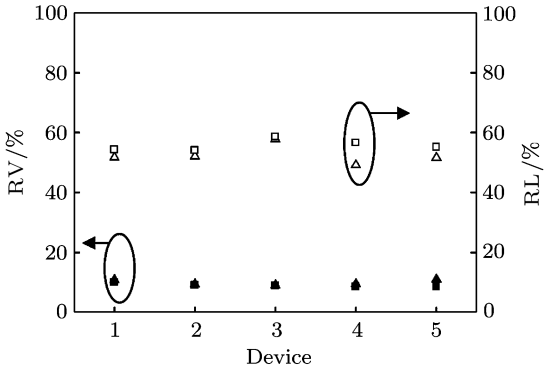



**Fig. 5.** Values of RV and RL of different devices and test current densities. Solid symbols correspond to RV while open symbols correspond to RL. Square and triangular symbols represent the cases of test current densities of 0.01 A/cm$^2$ and 0.1 A/cm$^2$, respectively.

In addition, we find that the difference of RV and RL under different test current densities of 0.01 A/cm$^2$ and 0.1 A/cm$^2$ is very slight. Taking Device 1 for example, we deduce that the values of RV are 10.0% and 10.8%, and the values of RL are 54.3% and 51.7% for test current densities of 0.01 A/cm$^2$ and 0.1 A/cm$^2$, respectively, after operation for 1600 h.

Therefore, we conclude that the operational lifetimes of our OLEDs are independent of the variations in both the organic layers' thickness and test current density. This implies that our device structures could be applied in a high-quality full-colour display and white lighting sources with multi-emitters. In order to achieve white emission, the most widely used method is to use three different colour-emitting (red, green, and blue) layers in one device unit.[23] Unfortunately, this kind of OLED usually suffers from non-uniformity degradation in each layer, limiting the device quality. Accordingly, our results reveal that the developed blue device structure in multi-emitting-layer OLEDs can overcome the drawback mentioned above to some extent and achieve high-stability OLEDs.

## 4. Conclusions

In this paper, we demonstrate that a maximum EQE of 6.19% and long-lifetime blue OLED can be realized by using DSA-ph doped TBADN as emitting layer. The electroluminescence properties of OLEDs

(e.g., operating voltage, EL spectra, and luminous efficiency) are found to be affected greatly by the changes in the thickness of the HIL and EML, while the operational lifetimes of devices are independent of the thicknesses of organic functional layers.

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