# Influence of the thickness of $N,N'$-Bis(naphthalene-1-yl)-$N,N'$-bis(phenyl) benzidine layer on the performance of organic light-emitting diodes

**Bo Jiao · Zhaoxin Wu · Xingwei Yan · Xun Hou**

**Abstract** We investigated the influence of the thickness of hole-transport layer, $N,N'$-biphenyl-$N,N'$-bis(1-naphthyl)-$(1,1'$-biphenyl)-4,4'-diamine (NPB), on the performance of the typical bi-layer organic light-emitting diodes (OLEDs). It was found that both the current efficiency and the power efficiency of bi-layer OLEDs were improved when the thickness of the hole-transport layer varied from 30 to 120 nm. By investigating the hole-injection efficiency of ITO/NPB contact with various thicknesses of NPB film, we found that the hole-injection efficiency was reduced with the thickness of NPB layer increasing from 60 to 180 nm, which improved the injected carriers balance in devices and increased the efficiency of the bi-layer OLEDs.

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## 1 Introduction

Since the organic light-emitting diodes (OLEDs) were reported by Tang et al. [1], much attention has been attracted due to their promising application in planar lighting and flat panel displays [2, 3]. In Tang's work, the OLEDs have the bi-layer structure, in which the $N,N'$-Bis(3-methylphenyl)-$N,N'$-bis(phenyl)benzidine (TPD)-like diamine serves as a hole-transport layer, and tris-(8-hydroxyquinoline) aluminum ($Alq_3$) is the emission and electron-transport layer. Based on this typical bi-layer structure, the more complicated structure of OLEDs with hole-injection layer, electron-injection layer, hole-block layer, and multiple-layer structures were developed for high efficiency and stability [4–7].

As for the bi-layer OLEDs, in order to obtain the high performance, the thickness of electron-transport and emission layer ($Alq_3$) has been optimized by optical interference effect [8, 9]. So et al. showed the maximum of efficiency of device with an approximately 60-nm thickness of $Alq_3$ layer [8]. Wu et al. also studied the optimal thickness of $Alq_3$ with different metallic cathode such as Ca, Mg, Ag, and Al [9]. For the hole-transport layer (HTL), it is found that the optimal-thickness value of HTL strongly depends on the device configuration [10–13]. For OLEDs with buffer layer of anode, such as copper phthalocyanine, Qiu et al. reported that the optimal thickness of HTL (TPD) is around 40 nm [10]. For doped OLEDs, however, Zhou et al. reported that the optimal thickness of HTL (NPB) is around 12 nm [11]. When it comes to the typical bi-layer OLEDs, Zhang et al. have investigated the dependence of the performance of the typical bi-layer OLEDs on the HTL (NPB) thickness (from 0 to 60 nm) in detail and reported that the optimal thickness of the NPB layer is no less than 20 nm [12]. However, the exact value of the optimal thickness of HTL cannot be obtained from their report. Based on the above results, the 20–60-nm thickness was always assumed to be the optimal HTL thickness for the typical bi-layer OLEDs.

In this paper, we investigated the dependence of the performance of OLEDs with typical bi-layer structure,

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ITO/$N,N'$-biphenyl-$N,N'$-bis(1-naphthyl)-(1,1'-biphenyl)-4,4'-diamine (NPB)/tris-(8-hydroxyquinoline) aluminum (Alq$_3$) (60 nm)/LiF (0.5 nm)/Al (100 nm), on the thickness of NPB film (from 30 to 180 nm) and found that both the current efficiency and the power efficiency of devices can be improved with thickness of NPB film from 30 to 120 nm, and optimal thickness of NPB film for bi-layer OLEDs is about 120 nm instead of 20–60 nm. The dependence of the hole-injection efficiency of ITO/NPB contact on the thickness of NPB films was also experimentally studied. We found that the increase of thickness of NPB film will lead to decrease of hole-injection efficiency, which will be the main reason for the increase of performance of devices by improvement of the balance between the injected holes and electrons in devices.

## 2 Experimental details

The bi-layer OLEDs was built on the glass substrates precoated with indium tin oxide (ITO) film and had a configuration of ITO/NPB /Alq$_3$ (60 nm)/LiF (0.5 nm)/Al (100 nm). The molecular structures of materials used and the configuration of the bi-layer OLEDs are shown in Fig. 1. The pressure during thermal evaporation for deposition
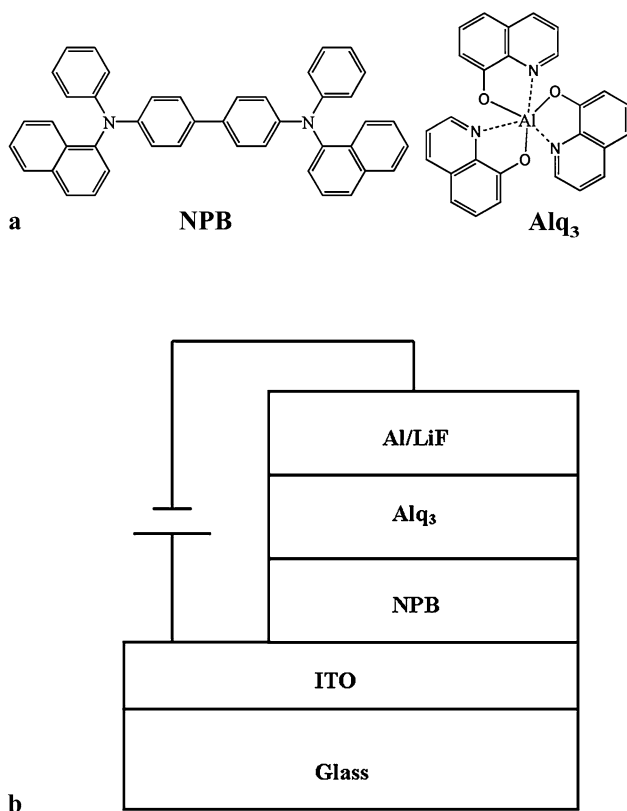




**Fig. 1** (**a**) Molecular structures of materials used and (**b**) configuration of devices

was $1 \times 10^{-3}$ Pa. NPB and Alq$_3$ were subsequently deposited at a rate of 0.3 nm/s. After the deposition of the organic layers, the LiF and Al were deposited at a rate of 0.05 and 0.5 nm/s without a vacuum break, respectively. The thickness of the films was determined in situ by a quartz-crystal sensor and ex situ by a profilometer. The emission area of the device was 12 mm$^2$.

The luminance–current–voltage (L–I–V) characteristics of the devices were measured using a computer-controlled sourcemeter (Keithley 2602) and a calibrated silicon photodiode. All the measurements were carried out at room temperature under ambient conditions.

## 3 Experimental results and discussion

Current density–voltage (J–V) and luminance–voltage (L–V) characteristics of bi-layer OLEDs with different NPB thicknesses are shown in Figs. 2(a) and 2(b), respectively. It was observed that increase of the NPB layer thickness from 30 to 180 nm shifts the J–V and L–V curves to the higher voltage, just as shown in Figs. 2(a) and 2(b). Current efficiency and power efficiency of these devices as functions of NPB thickness are shown in Fig. 2(c). As shown in Fig. 2(c), both the current efficiency and the power efficiency of bi-layer OLEDs were remarkably affected by the thickness of NPB. The current efficiency increases dramatically, close to 50%, with the thickness of NPB varying from 30 to 150 nm. The power efficiency is also improved, about 5%, with the NPB thickness varying from 30 to 120 nm. As we know, the power efficiency can be deduced from the current efficiency and corresponding operational voltage, just as shown in the equation [14]

$$\eta_P = \frac{\pi \eta_C}{V} \tag{1}$$

where $\eta_P$ and $\eta_C$ are the power efficiency and the current efficiency of the bi-layer OLEDs, respectively. $V$ is the corresponding applied voltage of the OLEDs. When the NPB thickness is varied from 30 to 120 nm, both the current efficiency and the operational voltage dramatically increase. Therefore, the increased percentage of the power efficiency is not as large as that of current efficiency, just as shown in Fig. 2(c). Taking the improvement of the current efficiency and the power efficiency into account, the optimal thickness of NPB film of bi-layer OLEDs, therefore, is about 120 nm instead of 20–60 nm.

The efficiency of OLEDs is determined by the balance between the hole and electron currents injected from the anode and cathode, respectively. In typical bi-layer OLEDs, holes are the majority carrier, and electrons are the minority carrier [15]. So, the performance of the electrode/organic
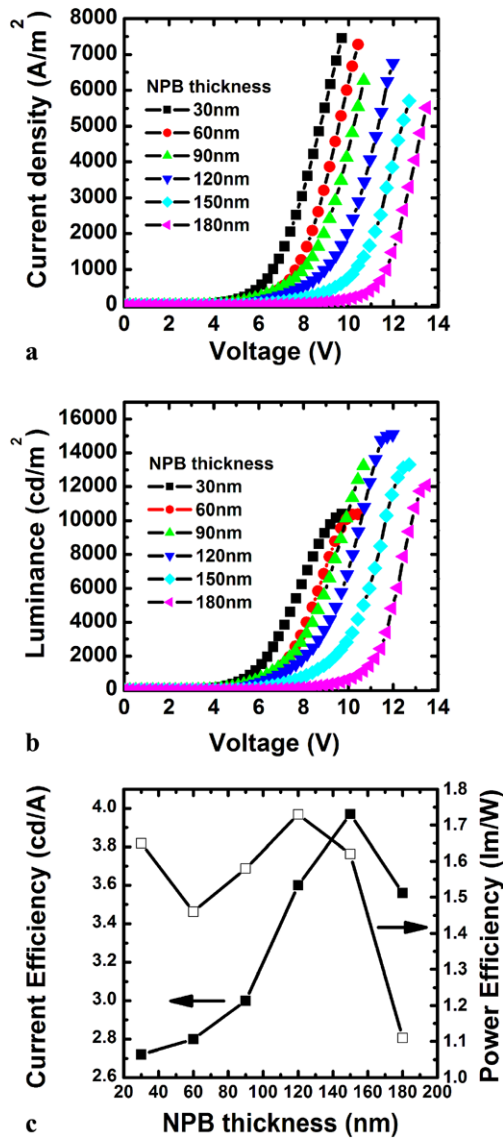
**Fig. 2** (**a**) J–V and (**b**) L–V characteristics of the devices with NPB thicknesses varying from 30 to 180 nm. (**c**) The current efficiency and power efficiency of these device as functions of NPB thickness at a current density of 445 A/m²

contacts to supply the organic bulk with charge plays an important role for OLEDs performance [16, 17]. A figure-of-merit for contact performance is the injection efficiency, defined by [17–19]

$$\eta = \frac{J_{INJ}}{J_{BULK}} \tag{2}$$

where $J_{INJ}$ is the current density supplied by the contact, and $J_{BULK}$ represents the maximum current density that organic layer can carry in the case of ideal ohmic contact, which is,

for trap-free materials, space-charge limited (SCL) current given by Mott–Gurney law as

$$J_{BULK} = J_{SCL} = \left(\frac{9}{8}\right)\varepsilon\varepsilon_0\mu\frac{E^2}{L} \tag{3}$$

where $\varepsilon\varepsilon_0$ is the dielectric constant, $E$ is the electric field, $L$ is the film thickness, and $\mu$ is the carrier mobility, which is dependent on the electric field and given by the Poole–Frenkel equation as

$$\mu = \mu_0 \exp(\beta\sqrt{E}) \tag{4}$$

where $\mu_0$ is the zero-field mobility, and $\beta$ is Poole–Frenkel factor. By combining (3) and (4), the SCL current density can be expressed by [20]

$$J_{SCL} = \left(\frac{9}{8}\right)\varepsilon\varepsilon_0\mu_0 \exp(\beta\sqrt{E})\frac{E^2}{L} \tag{5}$$

To understand our experimental results in Fig. 2(b), the hole-injection efficiency with various thicknesses of NPB films was investigated. The NPB bulk layer is established to be trap-free by analysis of transport and Xerographic measurements. It has been established that transport occurs by hopping in a Gaussian manifold with a nominal width of 0.1 eV [17, 21, 22]. So the $J_{BULK}$, is space-charge limited (SCL) current expressed by (5).

In order to calculate the $J_{BULK}$ of NPB film, two parameters, the zero-field mobility $\mu_0$ and the Poole–Frenkel factor $\beta$, should be estimated at first. For thin organic film with the thickness from 60 to 180 nm, two methods can be used to evaluate those two parameters. One method is the time-delay measurement of transient electroluminescence signal, which, however, is not suitable for our study because the transient time is determined by slower electron carrier in Alq₃ [23]. Another method is the space-charge-limited current measurement. Recently, Chu et al. presented the hole mobility of NPB at various thickness (50–1000 nm) by the space-charge-limited current measurement [24–26]. They found that, because of the effect of interfacial trap states, the hole mobility of NPB increases as the thickness increases [25, 26].[1] Based on the data ($\mu_0$ and $\beta$) obtained from their report, $J_{BULK}$ of NPB films with the thickness from 60 to 180 nm are calculated by (5), which are shown in Fig. 3. As showed in Fig. 3, the $J_{BULK}$ calculated from the SCLC model are much larger than the realistic current that the organic material carries due to the poor injection performance of the ITO/NPB contact.

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[1]In Chu's work, the surface dipole was ascribed to the trap state. This surface dipole may be formed by C₆₀, which was used for a quasi-Ohmic contact between ITO and NPB, or may be formed by the direct interaction between ITO and NPB. Whilst the formation of surface dipole between ITO and NPB was also reported by He et al. [27]. So it is believed that the result of Chu's work can be used in the ITO/NPB contact.
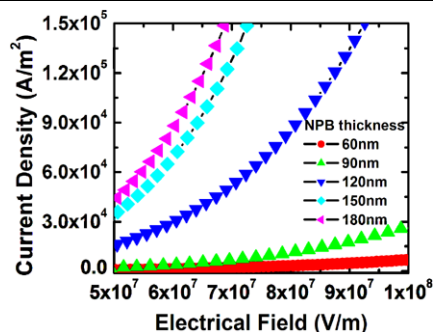
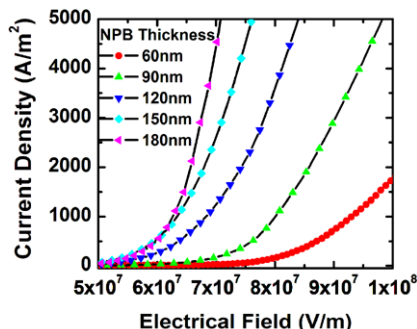**Fig. 3** The $J_{BULK}-E$ characteristics of the NPB layer with thicknesses varying from 60 to 180 nm



**Fig. 4** The $J_{INJ}-E$ characteristics of the hole-only devices ITO/NPB/Ag (100 nm) with NPB thicknesses varying from 60 to 180 nm

In order to get the $J_{INJ}$, the hole-only device, ITO/NPB ($x$ nm)/Ag (100 nm), where $x = 60$, 90, 120, 150, and 180 nm, are fabricated. Silver was chosen as the cathode with Fermi energy of 4.6 eV, and the lowest unoccupied molecular orbital (LUMO) level of NPB is 1.8 eV. The great offset between the Fermi energy of the cathode and the LUMO level of NPB served to reduce the efficiency of the electron injection and guarantee that the holes injected from the anode would dominate in the device.

The current density–electrical field ($J_{INJ}-E$) characteristics of hole-only devices are shown in Fig. 4. Because the hole mobility increases as the NPB layer thickness increases [25, 26], the current density $J_{INJ}$ increases with the increase of NPB layer thickness at constant electrical field.

Based on (2) and data in Figs. 3 and 4, we can calculate the injection efficiency of the ITO/NPB contact, which are shown in Fig. 5. Obviously, the injection efficiency of the contact decreases as the NPB layer thickness increases from 60 to 150 nm. The decrease of injection efficiency of the ITO/HTL contact is expected to improve the balance between hole and electron currents arriving at the recombination zone, which leads to the increase of current efficiency of devices. When the NPB thickness is larger than 150 nm, the current efficiency, however, decreases with the increase of the NPB layer thickness. This may be be-
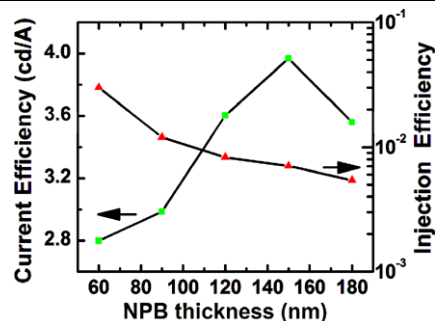


**Fig. 5** The injection efficiency of ITO/NPB contact (triangles) as a function of NPB thickness at an electric field of $6 \times 10^7$ V/m. The current efficiency of ITO/NPB/Alq$_3$/LiF/Al (squares) as a function of NPB thickness at a current density of 445 A/m$^2$

cause the balance between injected holes and electrons deteriorates when the hole injection becomes minor to the bulk organic emitting layer. In the previous research, in order to improve the balance of holes and electrons for the high performance of OLEDs, many methods were proposed to reduce the holes injection, for example, NPB layer is doped with rubrene [28]; the films of copper phthalocynanine (CuPc) [17], Alq$_3$ [29], and bathocuproine (BCP) are used as holes blocking layer in devices [30], etc. Comparing with these methods, however, our work provides a simpler and more practical way to improve the balance of hole and electron current by adjusting only the thickness of hole transport layer.

## 4 Conclusion

By comparing current and luminance characteristics of bilayer OLEDs with NPB layer thickness varying from 30 to 180 nm, we have found that both the current efficiency and the power efficiency of OLEDs with typical bi-layer structure increase with the increase of NPB layer thickness, and the optimal thickness is about 120 nm. The effects of hole-injection efficiency for various thicknesses of NPB were investigated. It is found that the hole-injection efficiency was reduced by increase of NPB film thickness, which was expected to improve the balance of carriers in devices and plays a more important role in the increase of the OLEDs performance. Our investigation also presented a simple way to improve the electron–hole balance in bi-layer OLEDs by adjustment of thickness of HTL for its high efficiency.

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