# Improving efficiency of organic light-emitting devices by optimizing the LiF interlayer in the hole transport layer*

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The efficiency of organic light-emitting devices (OLEDs) based on N,N'-bis(1-naphthyl)-N,N'-diphenyl-N,1'-biphenyl-4,4'-diamine (NPB) (the hole transport layer) and tris(8-hydroxyquinoline) aluminum ($Alq_3$) (both emission and electron transport layers) is improved remarkably by inserting a LiF interlayer into the hole transport layer. This thin LiF interlayer can effectively influence electrical performance and significantly improve the current efficiency of the device. A device with an optimum LiF layer thickness at the optimum position in NPB exhibits a maximum current efficiency of 5.96 cd/A at 215.79 mA/cm$^2$, which is about 86% higher than that of an ordinary device (without a LiF interlayer, 3.2 cd/A). An explanation can be put forward that LiF in the NPB layer can block holes and balance the recombination of holes and electrons. The results may provide some valuable references for improving OLED current efficiency.

**Keywords:** organic light-emitting devices, LiF, interlayer, efficiency

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## 1. Introduction

Organic light-emitting devices (OLEDs) have developed intensively over the past 24 years, owing to their great potential applications in solid-state lighting and large flat panel displays. Ever since an OLED with a multilayer structure was reported by C W Tang and S A VanSlyke, tremendous efforts have been made to improve device performance.[1−5] The optimization of charge injection and transport is very important for bright and efficient OLEDs. Lots of work has been done to improve the balance of holes and electrons injected into the emitting layer (EML)[6−11] and the insertion of an interlayer into the hole transport layer (HTL) has been found to be one of the most efficient approaches. LiF is an inorganic material that has widely been used as an electron buffer layer since it was first reported by Hung L S *et al.* in 1997.[12] It has been reported that inserting an ultrathin LiF layer between the ITO anode and organic layers (or in the middle of the HTL) can reduce the hole-mobility effectively, which can improve the efficiency of an OLED.[13,14] However, study of the effect of the thickness of the LiF interlayer, which acts as a hole blocking layer at the optimum location in NPB, has not been reported until now.

In this paper, an ultrathin LiF layer is inserted into an HTL to investigate the characteristics of these diodes and analyse the mechanism for the improvement of the efficiency of the device by varying the location and the thickness of LiF in NPB. Furthermore, this method had been applied to a blue emitting device in order to prove its universal applicability of current efficiency enhancement in different OLEDs.

## 2. Experiment

We conducted an experiment to clarify the role of LiF inside an OLED. The control device (CD) had the configuration of ITO/NPB (40 nm)/$Alq_3$ (50 nm)/LiF (0.8 nm)/Al (120 nm), which did not have a LiF interlayer in the HTL (NPB) on purpose. Then a LiF interlayer was inserted into the NPB of the CD with con-

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figurations of ITO/NPB ($x$ nm)/LiF (0.8 nm)/NPB (40-$x$ nm)/$Alq_3$ (50 nm)/LiF (0.8 nm)/Al (120 nm), ($x$ = 10 nm, 20 nm, 25 nm, 30 nm), of which the thickness of LiF was chosen to be 0.8 nm first. After the LiF interlayer reached its optimum position, we varied the thickness of LiF from 0.3 nm to 1 nm, in order to obtain its best thickness at the optimum location. All the organic layers as well as LiF and Al were deposited on a pre-cleaned ITO glass substrate with a sheet resistance of 20 Ω/sq, by using a thermal evaporation coating system at a pressure of about $2\times10^{-4}$ Pa. The deposition rates and thicknesses of the various layers were monitored using a quartz crystal oscillator. The evaporation rates were 0.1 nm/s–0.3 nm/s, 0.05 nm/s, and 1 nm/s for the organic layer, the LiF layer and the Al cathode, respectively. The current density ($J$)–luminance ($L$)–voltage ($V$) and electroluminescence (EL) characteristics of the devices were measured by a Keithley model 2400 programmable voltage–current source and a PR 650 spectra scan spectrometer. All measurements were carried out at room temperature.

## 3. Results and discussion

It is universally accepted that the mobility of a hole transport material is much greater than that of an electron transport material for general organic materials.[15] From the energy level diagram (Fig. 1), we can see that the total barrier (1.0 eV) for holes from an ITO anode to $Alq_3$ is lower than that (1.2 eV) for electrons from a cathode to $Alq_3$, which indicates that holes can enter into an emitting layer much more easily than electrons. This imbalance will reduce the recombination rate of electrons and holes in the EML and consequently lead to performance degradation of devices. In this report, we use LiF as the hole blocking material to prevent the holes from entering into the HTL, thereby improving the balance of carrier injection in view of its insulating property. The current density–luminance–voltage ($J$–$L$–$V$) and the current efficiency-current density characteristics (in which $x$ was varied from 10 nm to 30 nm) are presented in Fig. 2(a) and Fig. 2(b), respectively. Compared with the CD, the device with the LiF interlayer apparently exhibits a lower current density and lower luminance, but higher current efficiency. The maximum current efficiencies are 4.77 cd/A, 5.33 cd/A, 5.93 cd/A, 5.07 cd/A for the devices where the LiF interlayers are located in NPB 10 nm, 20 nm, 25 nm, and 30 nm away from anode, respectively, compared with 3.2 cd/A for the CD (Fig. 2(b)). When the LiF interlayer is located 10 nm away from the ITO anode, both the hole injection ability from ITO to NPB and the transport characteristics in NPB are reduced due to the accumulation of holes on the left-hand side of the NPB/LiF interface and the blocking effect of the LiF interlayer. Furthermore, the accumulation of holes might create an internal electric field, whose direction points from the left-hand side of the NPB/LiF interface to the ITO/NPB interface. Then the inverted internal electric field will cause a force which can prevent hole injection and transportation into the EML and lead to a weaker hole injection, a lower carrier recombination rate and an immediate lower luminance.[14] With the value of distance $x$ (shown in Fig. 1) increasing, hole injection and transport become relatively easier as the intensity of the internal electric field at the ITO/NPB interface becomes weaker. When the $x$ increases to 25 nm away from the anode, the efficiency of the device presents the highest value of 5.93 cd/A at 11 V due to the maximum balance between the holes and electrons in the EML. However, as the $x$ increases to 30 nm or more, the location of the LiF interlayer is very close to the recombination zone and then some electrons may pass through the interlayer and reach the anode. Consequently, the number of electrons would become less than that of holes in EML, which will break the balance of hole-electron pairs in this layer and again lead to the decrease of the current efficiency of device. However, as shown in Fig. 2(a), when the distance increases to more than 25 nm away from the ITO anode, the driving voltages become almost identical, which is due to a similar hole injection and transport process. That is to say, the hole injection and transport characteristics are insensitive to the accumulation of holes when the distance is more than 25 nm.

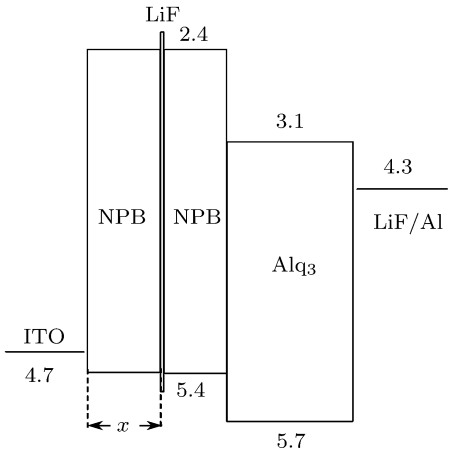



**Fig. 1.** Energy level diagram of the devices.

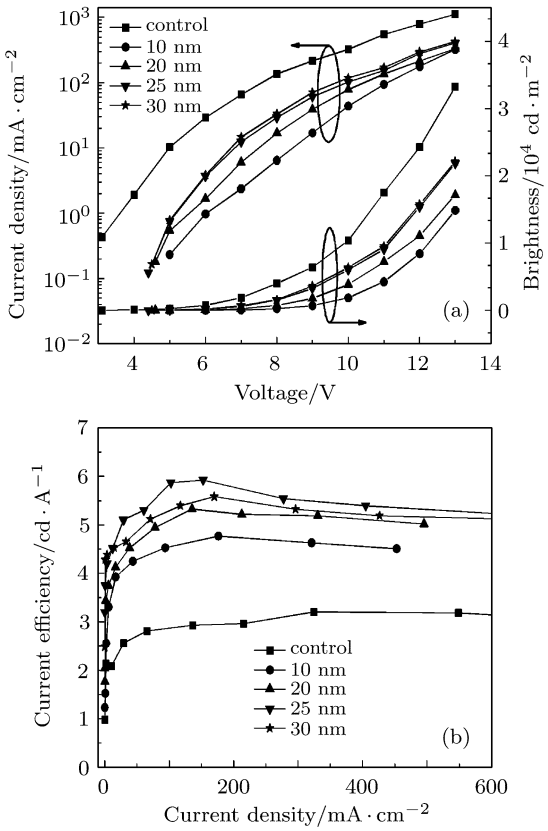



**Fig. 2.** Results for the CD and devices with different locations of LiF ($x$ = 10 nm, 20 nm, 25 nm, 30 nm). Panel (a) shows $J$–$L$–$V$ characteristics and panel (b) indicates current efficiency–current density characteristics.

The thickness of LiF interlayer is then optimized at the optimum location. Four types of devices were fabricated with the following structures: ITO/NPB (25 nm)/LiF ($y$ nm)/NPB (15 nm)/$Alq_3$ (50 nm)/LiF (0.8 nm)/Al (120 nm), ($y$ = 0.3 nm, 0.5 nm, 0.8 nm, 1.0 nm, respectively). It is clearly shown in Fig. 3(a) that with the thickness of the LiF interlayer increasing, both the current density and the corresponding luminance decrease at the same voltage, which should be induced by the hole blocking effect of the LiF interlayer and the internal electric field mentioned above. So the number of holes be reduced, thereby resulting in the balance of carriers, which means that the efficiency can be improved. When the thickness of the LiF interlayer reaches 0.8 nm, the current efficiency of the device exhibits the highest value of 5.96 cd/A at 215.79 mA/cm$^2$, which indicates that 0.8 nm of the LiF interlayer is the optimum thickness for balancing holes and electrons in the EML. However, if the LiF layer in NPB becomes thicker (or thinner), there will be less (or more) holes than electrons reaching the recombination zone, so a new imbalance will be created and the efficiency will be degraded accordingly (as shown in Fig. 3(b)).

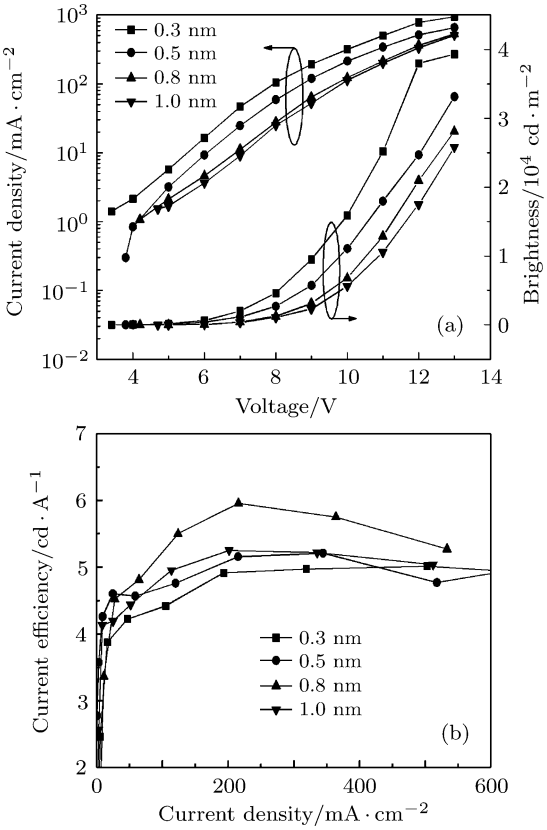



**Fig. 3.** Results for the device with different thicknesses ($y$ = 0.3 nm, 0.5 nm, 0.8 nm, 1.0 nm) of LiF interlayer at the optimum location. Panel (a) shows $J$–$L$–$V$ characteristics, and panel (b) indicates current efficiency–current density characteristics.

To clarify the effect of hole blocking induced by LiF interlayer, hole-only devices with the structures of ITO/NPB (25 nm)/LiF (0.8 nm)/NPB (15 nm)/Al (120 nm) and ITO/NPB (40 nm)/Al (120 nm) were designed and the dependencies of current density on voltage were measured and shown in Fig. 4. From the figure, it can be found that the current density of the device with the LiF interlayer is in fact lower than that of the device without this layer. Hence a lower current density should be induced by the insertion of a LiF interlayer and its hole blocking effect. Another two blue emitting devices with a kind of new blue emitting material (TC-1759) were also fabricated to demonstrate the universality of this approach. The structures were as follows: ITO/NPB (25 nm)/LiF (0.8 nm)/NPB (15 nm)/TC-1759 (50 nm)/Bphen (30 nm)/LiF (0.8 nm)/Al (120 nm) and ITO/NPB (40 nm)/TC-1759 (50 nm)/Bphen (30 nm)/LiF (0.8 nm)/Al (120 nm).

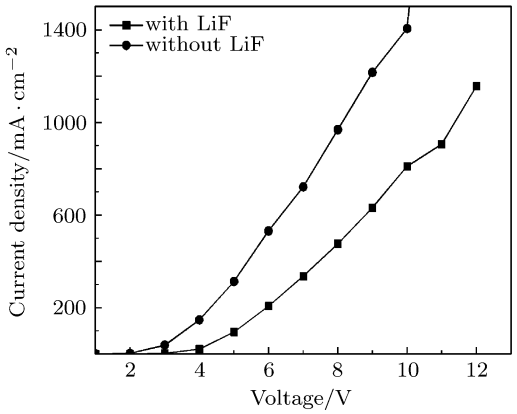



**Fig. 4.** Dependencies of current density on voltage for hole-only devices with and without a LiF interlayer.

It can be obviously seen in Fig. 5 that the blue emitting device with a LiF interlayer exhibits a low current density and high efficiency compared with those of the device without a LiF interlayer, which are similar to those of the device based on $Alq_3$ discussed above. The maximum current efficiency increases from 4.94 cd/A to 5.84 cd/A at low luminance ($\sim$130 cd/m$^2$). Thus the optimum LiF interlayer has some universal applicability of current efficiency enhancement in different OLEDs, owing to its hole blocking effect.

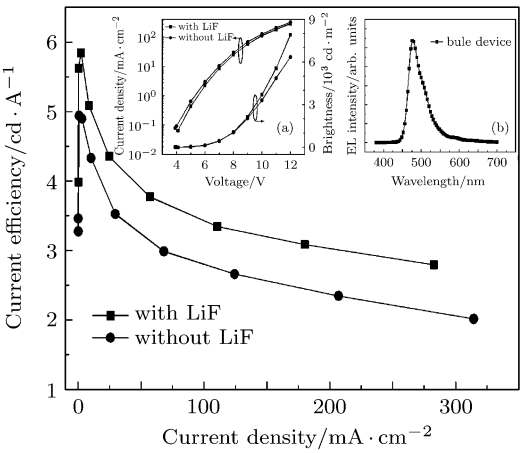



**Fig. 5.** Dependencies of current efficiency on current density, and dependencies of current density and brightness on voltage (inset (a)), EL intensity on wavelength (inset (b)).

## 4. Conclusion

We demonstrate an OLED efficiency improvement by optimizing the location and thickness of the LiF interlayer in the HTL to adjust the hole transport character and improve the balance between holes and electrons in the EML. When the LiF layer with 0.8-nm thickness is located in NPB, 25 nm away from ITO anode, the device shows the highest luminance of 28120 cd/m$^2$ at a fixed bias of 13 V and exhibits a maximum current efficiency of around 5.96 cd/A at 215.79 mA/cm$^2$, which is about 86% higher than that (3.2 cd/A) of the CD without a LiF interlayer. Since it serves as a hole blocking layer, a LiF interlayer inserted in the HTL reduces the number of holes from the anode. Consequently, the hole-electron balance is improved and the higher efficiency is achieved by their effective recombination. However, if the LiF layer is too thick, it can debase the brightness and current efficiency. Furthermore, a blue light-emitting device is successfully fabricated by inserting the optimum LiF interlayer into it and we have obtained an $Alq_3$-based device. This device also exhibits higher efficiency than the device without the interlayer, indicating that this experiment may provide some valuable references for improving OLED current efficiency.

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