Highly efficient organic electroluminescent diodes realized by efficient charge balance with optimized Electron and Hole transport layers

M. A. Khan[a,*], Wei Xu[a], Fuxiang Wei[a], Yu Bai[a], X. Y. Jiang[a], Z. L. Zhang[a,b] W. Q. Zhu[a,b]
[a] Department of Materials Science, Shanghai University, Jiading 201800, P. R. China
[b] Key Laboratory of Advanced Display and System Applications, Ministry of Education, Shanghai University, Shanghai 200072, P. R. China

Abstract

Highly efficient organic electroluminescent devices (OLEDs) based on 4,7- diphenyl-1, 10-phenanthroline (BPhen) as the electron transport layer (ETL), tris (8-hydroxyquinoline) aluminum (Alq_3) as the emission layer (EML) and N,Ń-bis-[1-naphthy(-N,Ńdiphenyl-1,1´-biphenyl-4,4´-diamine)] (NPB) as the hole transport layer (HTL) were developed. The typical device structure was glass substrate/ ITO/ NPB/ Alq_3/ BPhen/ LiF/ Al. Since BPhen possesses a considerable high electron mobility of 5×10^{-4} cm^2 V^{-1} s^{-1}, devices with BPhen as ETL can realize an extremely high luminous efficiency. By optimizing the thickness of both HTL and ETL, we obtained a highly efficient OLED with a current efficiency of 6.80 cd/A and luminance of 1361 cd/m^2 at a current density of 20 mA/cm^2. This dramatic improvement in the current efficiency has been explained on the principle of charge balance.

Keywords: 4,7- diphenyl- 1, 10- phenanthroline (BPhen); Charge balance; Optimized thickness; Luminescence.

1 Introduction

Organic light-emitting diodes (OLEDs) have received much attention in recent years because of their potential application in full color flat-panel displays [1-5]. Since Tang and Van Slyke [6] reported vacuum-deposited bilayer OLEDs, consisting of a hole transport layer (HTL) and an emissive electron transport layer (ETL), great progress has been made in improving the performance. It is one of the key factors to enhance the efficiency and stability of the OLEDs for commercial display applications [7]. The operating mechanisms involve injection of electrons and holes to the organic emitter layers from the electrodes. On recombination, electrons and holes generate molecular excitons [8], which result in the emission of light from the emitter layer. It is, therefore, important to balance the number of holes and electrons in EL devices. In most OLEDs, the barrier height for holes is relatively lower than that for electrons, and the mobility of holes in an organic layer is larger by orders of magnitude than that of electrons. Thus, the injection and transportation of holes are easier than that of electrons [7]. To achieve a balanced injection of carriers, it is important to use the materials having high electron mobility and excellent transporting capabilities.

The most common material used for ETL is Alq_3. It possesses superior film stability and thermal endurance. However, due to its low electron mobility of 10^{-6} cm^2 V^{-1} s^{-1} and intrinsic degradation [9, 10], Alq_3 is not suitable for achieving efficient carrier balance. Experimental evidences strongly suggested that the excess amount of holes generate non-emissive cationic species and fluorescence quenchers in the Alq_3 [11, 12].

Hence, enhancement of electron conduction in the ETL of OLEDs is critical approach to both achieving the efficient carriers balance and improving the luminous efficiency. Although some electron transport materials, such as phenanthroline and oxadiazole compounds can be used as ETL due to their improved drift electron mobility [13-15], the device stability and thermal endurance have not fulfilled the requirement of commercial display applications [15, 16]. However, since 4,7- diphenyl- 1, 10-phenanthroline (BPhen) possesses a considerable high electron mobility (5×10^{-4} cm^2 V^{-1} s^{-1}) [15], devices with BPhen as ETL can realize an extremely high luminous efficiency. Recently, H. H. Fong et al. [17] reported that the efficiency of 4.4 cd/A was achieved at 20 mA/cm^2 with BPhen as ETL. This is almost 50% greater than that of conventional device (~ 3 cd/A) with Alq_3 as ETL.

In this paper, detailed study has been performed on OLEDs based on BPhen as ETL.

We achieved an extremely high luminous efficiency of 6.80 cd/A at 20 mA/cm^2 by optimizing the thickness of ETL and HTL. To the best of our knowledge, this is the best result ever reported with out doping of EML or ETL.

2. Experimental

Glass coated with indium-tin oxide (ITO) was used as starting substrate. After routine chemical cleaning, ITO was further treated by UV ozone. The devices were prepared by vapor deposition onto an indium tin oxide coated glass substrate with a sheet resistance of 20 Ω/square. The organic layers and the cathode layer were deposited by vacuum vapor deposition at 1.0×10^{-5} torr. Three series of devices, with different thickness of NPB and BPhen with a fixed thickness of Alq (30 nm) were fabricated. Each series consisted of fixed thickness of NPB with varying thicknesses of BPhen while Alq_3 (30 nm) thickness was kept constant for all devices.

The active area of the devices was 5×5 mm^2. The thickness of the organic layers was monitored by using quartz-crystal monitor. The current–voltage (I-V) and luminance characteristics were measured by Keithley 2400 Source Meter and Minolta LS-110 luminance meter.

3. Results and Discussion

Three series of devices with the following structures were fabricated:

Series A Devices: ITO/ NPB (20 nm)/ Alq_3 (30 nm)/ BPhen (x nm)/LiF (1 nm)/ Al (150 nm), where x = 30 nm, 50 nm, 60 nm.

Series B Devices: ITO/ NPB (40 nm)/ Alq_3 (30 nm)/ BPhen (x nm)/LiF (1 nm)/ Al (150 nm), where x = 30 nm, 50 nm, 60 nm.

Series C devices: ITO/ NPB (50 nm)/ Alq_3 (30 nm)/ BPhen (x nm)/LiF (1 nm)/ Al (150 nm), where x = 30 nm, 50 nm, 60 nm.

Current efficiency versus current density characteristics of all three series of devices are shown in figure 1 and Table 1 summarizes the performances of these devices. It can be seen from figure 1 and table 1 that in every series of devices there is a highly efficient device for a particular thickness combination of HTL and ETL. Table 1 shows that in Series A devices, Device A1 has a maximum current efficiency of 5.28 cd/A at a current density of 20 mA/cm^2 with 20 nm thick NPB and 30 nm thick BPhen.

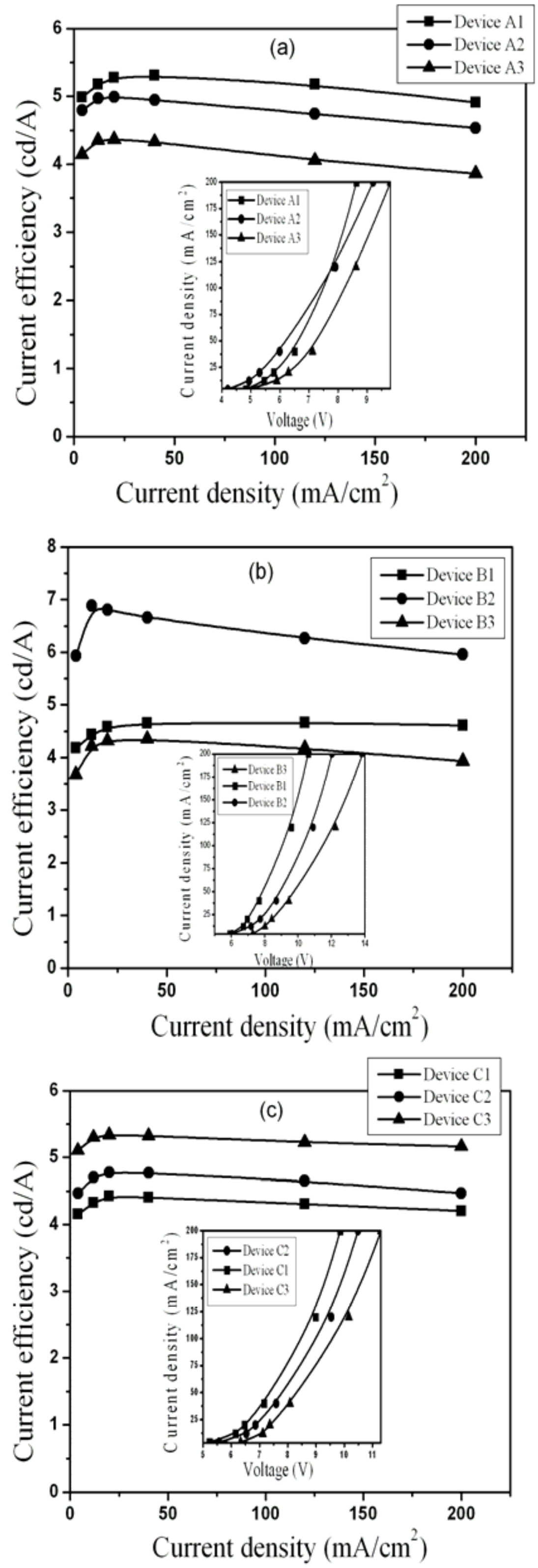


Figure 1. Current efficiency versus current density characteristics of three series of devices (a) Device A1, Device A2, Device A3 (b) Device B1, Device B 2,

Device B 3 (c) Device C 1, Device C 2, Device C 3. The insets show the J-V characteristics.

In Series B devices, when the thickness of NPB is increased to 40 nm the most efficient device has shifted to 50 nm thick BPhen having a current efficiency of 6.80 cd/A at a current density of 20 mA/cm^2. Similarly, in Series C devices, the efficient device is again shifted to higher thickness of BPhen (60 nm). This phenomenon clearly indicates that there exists a direct relation between current efficiency and the thickness of both NPB and BPhen. That is, if the thickness of NPB is less, the amount of holes reaching the interface will be more and vice versa. Therefore, when the thickness of NPB is increased, the number of holes holes reaching the interface will obviously be decreased and in order to achieve a better balance of holes and

Table 1. Performances of all three series of devices at 20 mA/cm^2 with a fixed Alq_3 (30 nm) thickness.

Devices	NPB Thickness	Bphen Thickness	Voltage (V)	Luminance (cd/m^2)	Current Efficiency (cd/A)
Series A devices: ITO/NPB (20 nm)/Alq_3 (30 nm)/BPhen (x nm)/LiF (1 nm)/Al (150 nm)					
Device A1	20 nm	30 nm	5.77	1056	5.28
Device A2	20 nm	50 nm	5.20	999	4.99
Device A3	20 nm	60 nm	6.29	874	4.37
Series B devices: ITO/NPB (40 nm)/Alq_3 (30 nm)/BPhen (x nm)/LiF (1 nm)/Al (150 nm)					
Device B1	40 nm	30 nm	6.96	916.5	4.58
Device B2*	40 nm	50 nm	7.74	1361*	6.80*
Device B3	40 nm	60 nm	8.42	863.7	4.31
Series C devices: ITO/NPB (50 nm)/Alq_3 (30 nm)/BPhen (x nm)/LiF (1 nm)/Al (150 nm)					
Devices C1	50 nm	30 nm	6.47	884.9	4.42
Devices C2	50 nm	50 nm	6.85	955.5	4.77
Devices C3	50 nm	60 nm	7.36	1068	5.34

*Most efficient device of all

electrons, it is must to increase the thickness of BPhen. In this way, we can obtain a reasonable balance of carriers, giving rise to the enhancement of current efficiency. However, current efficiency attains a maximum value only for optimum thickness of both NPB and BPhen. In our case, the optimum thickness of NPB is 40 nm and that of BPhen is 50 nm.

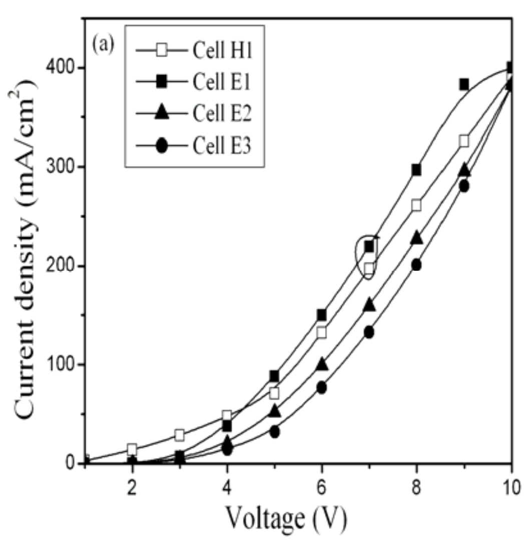
In order to understand the mechanism behind the dramatic increase in the current efficiency of OLEDs in our study, we demonstrated hole-only and electron-only devices. Hole-only (NPB based) devices have the following configurations:

Cell H1: ITO/ NPB (20 nm)/ Al (150 nm)
Cell H2: ITO/ NPB (40 nm)/ Al (150 nm)
Cell H3: ITO/ NPB (50 nm)/ Al (150 nm).

Electron only (Alq-BPhen based) devices have the following structures:

Cell E1: ITO/ Alq_3 (30 nm)/ BPhen (30 nm)/ Al (150 nm)
Cell E2: ITO/ Alq_3 (30 nm)/ BPhen (50 nm)/ Al (150 nm)
Cell E3: ITO/ Alq_3 (30 nm)/ BPhen (60 nm)/ Al (150 nm).

The motive behind using of both Alq_3 and BPhen in electron only devices is that, (i) they both are electron transport materials and (ii) to determine the efficient balance of charge carriers. Figure 2 shows the comparison of current density versus voltage characteristics of each hole-only device with all electron-only devices.



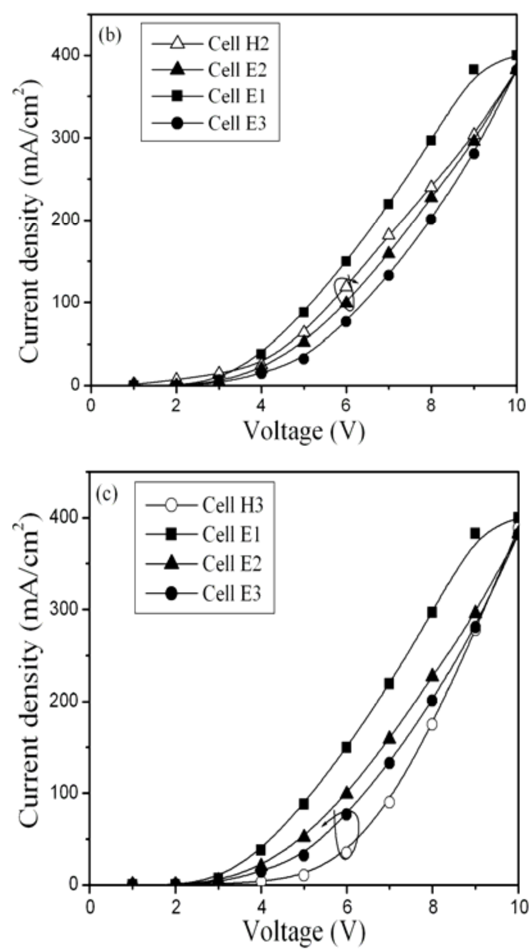


Figure 2. Comparison of current density – voltage characteristics of (a) Cell H1 with Cell E1, Cell E2 and Cell E3 (b) Cell H2 with Cell E1, Cell E2 and Cell E3, and (c) Cell H3 with Cell E1, Cell E2 and Cell E3.

The data indicate a reasonably good comparison of J-V characteristics of hole-only devices and electron-only devices. It is seen from figure 2 that there are a very close J–V curves only for those thicknesses of NPB and BPhen which were used for highly efficient devices, shown in figure 1. For example, Cell H1 is very close to Cell E1, Cell H2 is very close to Cell E2 and Cell H3 is very close to Cell E3. This comparison clearly indicates that there exists a perfect balance for both holes and electrons for the most efficient devices (Device A1, Device B2, and Device C3), shown in figure 1 and table 1.

Therefore, this study strongly suggests that the improvement in the device efficiency in our devices is due to the perfect balance of holes and electrons injected from opposite electrodes, resulting in an efficient radiative recombination in the emission zone. In addition, perhaps BPhen's ability of hole blocking can also be another reason for the improvement in the luminous gain. Hence, high radiative recombination is expected in Alq_3 emission layer. This highly efficient recombination results in high brightness and enhanced efficiency in our OLEDs.

4. Conclusion

In conclusion, we have demonstrated highly efficient organic light-emitting devices (OLEDs) by optimizing the thicknesses of both hole transport layer (HTL) and electron transport layer (ETL). Based on the principle of charge balance, we have presented hole-only (NPB based) and electron-only (Alq_3-BPhen) devices. By comparing the J-V characteristics of hole-only and electron-only devices, it has been found that there exits a highly efficient balance for both holes and electrons for particular thicknesses of NPB and BPhen in our devices. This efficient carrier balance leads to high luminous efficiency and high brightness. We have obtained a highly efficient device with a luminous efficiency of 6.80 cd/A and a luminance of 1361 cd/m^2 at a current density of only 20 mA/cm^2. This is the best result ever reported without doping of EML or ETL.

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