High Brightness and Long Lifetime OLED with Mixing Layer Technology

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ABSTRACT

In this paper, we demonstrate organic light emitting devices (OLED) that exhibit high brightness, low driving voltage and long lifetime. Devices with the brightness of 10,000 cd/m² can be achieved at 4 V by the use of the high mobility electron-transport layer (ETL) material, bis(10-hydroxybenzo[h]quinolino)beryllium (Bebq₂), and the mixing host (MH) technology. Electron mobility of Bebq₂ is two orders of magnitude higher than that of the typical ETL material, tris-(8-hydroxyquinoline) aluminum (Alq₃), from the time-of-flight (TOF) measurement and hence the driving voltage can be decreased. By co-evaporating the hole-transport layer (HTL) material and the ETL material as the host of the emitting layer, it reduces two volts in driving voltage because of its bipolar transport characteristics. MH technology can not only decrease the driving voltage, but also increase the device lifetime since it eliminates the sharp boundary of HTL/ETL interface and decreases the carriers piling up near this interface which causes the organic material degradation. Compared to the conventional heterojunction (HJ) OLED, operation lifetime of MH devices was enhanced by a factor of 4.

Keywords: OLED, mixing layer, lifetime.

1. INTRODUCTION

OLED is one of the most promising candidates for the next generation display technologies since it exhibits advantages such as self emissive, wide-view-angle, flexible-substrate-capability and potentially low cost [1], [2]. For recent years, OLED technologies also show a remarkable progress in material and device for improving the efficiency and the stability [3], [4].

Typically, an OLED consists of two-layer organic materials, a HTL and an ETL, with the thickness of several tens of nanometers, separately. High IQE material is usually doped inside the ETL and near the HTL/ETL interface to serve as emission centers and it is called emitting-layer (EML). With voltage applied to the device, the holes and the electrons are injected from the anode and the cathode, driven to near the HTL/ETL interface and blocked by the ETL material and the HTL material, respectively. It enhances the collision capture process and recombination happens. Then, it produces excitons and the energy will be transferred from EML host to the high IQE dopant materials to emit light. Typically, the size of a molecule is about several nanometers. Hence, such a structure in OLED is like a single HJ structure used in semiconductor light emitting diodes (LED) with some quantum dots inside the active layer. However, the driving voltage of an OLED is higher than that of a semiconductor LED and the operation lifetime is shorter.

Compared with semiconductor LED, we can see that there are two major differences. The first one comes from the difference in materials. Because an organic material usually has a wide band gap, its carrier concentration is quite low

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[5]. Crystallized organic solid may have the mobility compatible to amorphous silicon. However, in order to form a flat and uniform surface, amorphous organic structure for the OLED devices is necessary. It dramatically decreases the carrier mobility. Typically, carrier mobility of an organic material is six to eight orders of magnitude lower than that in semiconductor. As compared with typical semiconductor LED, the driving voltage of OLED device is three to four times larger although the semiconductor LED is much thicker than OLED device. Here, we use an ETL material, Bebq2, with electron mobility that is two orders of magnitude higher than the typical used ETL, Alq3. And it can effectively decrease the driving voltage.

The second difference between OLED and LED comes from the device structures. Since OLED device is typically a SH structure, many carriers accumulate near at the HJ interface. Such a strong built-in voltage and the space charges would deteriorate the organic materials. Besides, because the width of the emitting region is usually very narrow to several nanometers, the strong EM wave within this region might induce serious photochemical reaction and hence degrade the device performance [6]. It results in that the brightness of an OLED device decreases and the voltage increases at the same time with constant current driving. To overcome such a problem, MH structures are proposed [7], [8]. To mix the HTL organic and ETL organic material as the host of the EML creates a bipolar transport behavior. Without the sharp interface, the carrier piled-up effect can be much reduced and the lifetime would be extended. At the same time, the bipolar behavior of the mixed material also decreased the driving voltage and achieved high brightness.

2. EXPERIMENTS

In all of our experiments, we used indium-tin-oxide (ITO) glass substrate with low sheet resistivity (10 Ohm/sqr) and flat surface roughness ($R_a < 1$ nm). The size of the active region in our test pixel was 2 mm x 2 mm. Before organic deposition, O_2 plasma treatment was used to clean the ITO surface and increase the ITO work function. All deposition process was carried in a multi-chamber cluster evaporator. The base pressure was about 1×10^{-7} torr. Typically, deposition rate was controlled at 0.1 nm/sec. We used N,N'-diphenyl-N,N'-bis(1-naphthyl)-1,1'-biphenyl-4,4'-diamine (NPB) as the hole transporting material, 9-Benzothiazol-2-yl-1,1,6,6-tetramethyl-2,3,5,6-tetrahydro-1H,4H-11-oxa-3a-aza-benzoanthracene-10-one (C545T) as the green dopant material and Bebq2 as the electron transporting material. Table I illustrated the device structure used in the experiments. Bebq2 was also used as the host material of the emitting layer in our control device while NPB and Bebq2 were co-evaporated by the ratio of 1:1 in other experiments. The thickness of the total organic layer was kept at 100 nm and that of the EML is varied from 10 to 30 nm. The thickness of the HTL and the ETL were kept at the same value corresponding to different EML thickness. Dopant concentration of C545T in Bebq2 was 1.25% in mole ratio. LiF of 1.2 nm was used for better electron injection between the ETL and the aluminum cathode. The thickness of the aluminum cathode was 100 nm. After organic and metal deposition, devices were well encapsulated in the glove box with O_2 and H_2O concentrations below 1 ppm. Device performances were characterized by Keithley 2400 for current-voltage measurements and SR-2 photometers for the brightness measurements. Those instruments were connected to a personal computer and controlled by labview software. Lifetime measurements were conducted with DC constant current driving with the initial luminance set at 10,000 cd/m^2 .

Samples	HTL	EML			ETL
		HOST	HOST1+HOST2	Dopant	
	NPB	Bebq2	NPB+Bebq2	C545T	Bebq2
A	40	20		1.25%	40
B	45		10		45
C	40		20		40
D	35		30		35

Table 1 Experimental recipe of devices (Unit: nm)

3. RESULTS AND DISCUSSIONS

Figure 1 shows the charge carrier mobility Bebq2 as determined by time-of-flight (TOF) measurement. We can see that

the electron mobility of Beq2 is $6.96 \times 10^{-5} \text{ cm}^2/\text{V}\cdot\text{sec}$ at $2.5 \times 10^5 \text{ V/cm}$ which is two orders of magnitude larger than that of typical used ETL, Alq3 [9]. Typically, HTL materials exhibit higher mobility than ETL material over several orders of magnitude. That means, in OLED, the electric field strength on the ETL is much higher than that on HTL. Hence, with such a high mobility ETL material, device exhibits more balanced carrier transport and has a lower driving voltage.

Fig. 1 Typical log-log plots of time-of-flight transients for Beq2 at room temperature. The inset shows double linear plots of the time transient.

Fig2 shows the current-voltage characteristics of different samples. Sample A is our control device and exhibits lowest current density under the same driving voltage among all the samples. However, it still achieves a high current density of 1.09 A/cm^2 at only 7.5 V that contributes to the use of high electron mobility ETL. Sample B to sample D is our MH devices while the emitting layer thickness is varied from 10 to 30 nm . Compared the I-V characteristics of sample A and sample C, which have similar device structures except the host material of EML, we can see that the driving voltage has a 1.8 V reduction in MH case as shown at the inset of Fig 1.

From the I-V characteristics of sample B, C and D, we can observe that the current density is decreased with increasing emitting region. The emitting region was doped with C545T that acted as traps in the device and impeded the current transport in organic material. It would arise from the elimination of the sharp interface. Holes would have higher mobility in the emitting region and achieve balance conduction. Such a trap-charge-limited-current (TCLC) can be described approximately by the equation as follows [10]:

$$j_{TCLC} \propto \frac{V^{l+1}}{d^{2l+1}}$$

where j_{TCLC} is the current density, V is the voltage, d is the organic layer thickness and l is the parameter derived from the trap distribution. The value of l is larger with higher trap density. The lowest value of l is 1 . That corresponds to the trap-free space-charge-limited current and it is the maximum possible unipolar current that can sustain in organic materials. When the emitting layer increases from 10 , 20 to 30 nm , the l value corresponds to each device ranges from 4.80 , 5.38 to 5.91 . However, in traditional HJ case, l is 6.85 which is the highest value among all the experiments. Decrease of l value in MH devices results in the bipolar transport and decreases the probability of charge trapped by the C545T dopant. Hence, with such a MH technology, it decreases the l value and changes the carrier transport characteristics toward SCLC conduction.

Fig. 2 Comparison of current density versus voltage curve among the MH device (samples B, C and D) and the control device (sample A). The inset shows the comparison between sample A and sample C.

Figure 3 shows dependence between luminance and injection current density. Maximum brightness in sample A reaches 270,000 cd/m^2 while that in sample C and sample D can reach their maximum brightness at 284,000 cd/m^2 and 288,000 cd/m^2 , respectively. The slope of B-I curves corresponds to current efficiency (cd/A) and the inset shows the relationship between current efficiency and brightness among these samples. We can see that current efficiency of sample A is highest among all the samples while that of sample B and sample C are slightly lower. Sample D with thinnest EML has lowest current efficiency. That indicated that the absence of sharp HJ reduced the carrier confinement

Fig. 3 Comparison of brightness versus current curve among the MH device (samples B, C and D) and the control device (sample A). The inset shows the comparison of current efficiency and brightness.

effect and lower down the current efficiency. With increasing the thickness of MH may help to increase current efficiency. However, it is still somehow lower than that in the HJ case. It would be a trade-off between the voltage reduction and current efficiency.

Fig. 4 shows the spectrum of these four samples. It is a clear blue shift trend from HJ case to MH case. It may result from the optical interference effect or the different energy transfer path from the host to the dopant. To distinguish these two effects, we fabricated two more with typical HJ structures. The layer thickness is the same as sample B and sample D listed at Table 1 except the host material in the EML is Beq2 rather than MH. The spectrums of those three HJ devices are nearly the same as that of sample A with their peaks at 520 nm to 522 nm. However, in MH case, the spectrum peak shifts to 514 to 516 nm and it broadens at blue side. It may come from the energy transfer path from HTL material to C545T directly.

Fig. 4 Spectrum of different samples.

Fig. 5 Comparison of power efficiency versus brightness curve among the MH device (samples B, C and D) and the control device (sample A).

Power efficiency is one of the major indexes for OLED device performance evaluation. Although MH layer can effectively decrease the driving voltage as compared with typical HJ devices, the current efficiency is still slightly lower. Combined those two effect, we can see in Fig. 5, sample C and sample D of MH devices have highest power efficiency at the brightness less than and over 150,000 cd/m^2 , respectively. Although there exists a trade-off between driving voltage and current efficiency with MH technology, it is still beneficial in terms of power saving.

Since MH technology reduces the sharp heterojunction interface between HTL and EML and decreases the carriers piling-up near the interface, it can not only reduce the driving voltage but also increased the device lifetime as shown in Figure 6. Sample C exhibited the half-lifetime of 58.3 hours with the initial brightness at 10000 cd/m^2 that is nearly four times longer than that in the conventional device. Sample D has shorter lifetime that may come from the lower power efficiency at this brightness. However, it still has a lifetime of 49.4 hours that is much longer than that of HJ devices. In our experiments, thicker MH layer may not give to higher lifetime and better device performance that means device optimization is necessary. We can easily extract the operation lifetime would be near 12,000 hours with our MH devices under normal operation based on our statistical data that is sufficient long for some applications.

Fig. 6 Accelerated lifetime test of sample A, C and D.

4. SUMMARY

In summary, we have shown high brightness and long lifetime OLED devices by using high mobility ETL materials and MH technology. The high mobility ETL material, Beq2, exhibits the mobility over one order of magnitude than the traditional used ETL, Alq3. MH technology can effectively reduce the driving voltage because of its bipolar transport characteristics and long lifetime because of the HTL/EML interface blurring. Observation of blue shift in spectrum of MH devices shows different energy transfer paths in EML. Although MH would decrease current efficiency, the power efficiency still increases with proper device design. It can effectively decrease the power consumption. Finally, a four-time improvement in lifetime is shown in our optimized MH devices.

ACKNOWLEDGMENTS

This work was supported by the National Science Council, the Republic of China, under Grant No. NSC 92-2218-E-002-050.

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