High-Efficiency and Long-Lifetime Fluorescent Blue Organic-Emitting Device

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ABSTRACT

In this paper, We have demonstrated a blue fluorescent organic light-emitting device (OLED) with a current efficiency of 19.2 cd/A at 100 cd/m², an estimated half-lifetime of 15611 hours at an initial luminance of 1000 cd/m², and a voltage of 4.9 V at 20 mA/cm² with a high electron mobility electron transport layer (ETL) material and high efficiency dopant material. The external quantum efficiency (EQE) in this optimized OLED is 8.32%, which is very close to the theoretical limit. Carrier balance condition is achieved due to the incorporation of the high mobility ETL, bis(10-hydroxybenzo[h]quinolino)beryllium (Bebq2), which can not only effectively increase the current efficiency and elongate the operation lifetime, but also reduce the driving voltage and increase the power efficiency. The EML consisted of 4,4'-bis[2-(4-(N,N-diphenylamino)phenyl)vinyl]biphenyl (DPAVBi) as the blue dopant and 9,10-bis(2'-naphthyl) anthracene (ADN) as the matrix. We found that the dopant concentration of DPAVBi did not affect the mobility value of the EML which is consistent with the J-V characteristics. Besides, although it is believed the bulk ADN is a kind of HTL materials, we found the electron mobility of ADN is one order of magnitude higher than its hole mobility in our blue OLEDs.

Keywords: OLED, lifetime, blue

1. INTRODUCTION

Organic light-emitting devices (OLEDs) have attracted much attention due to the advantages of light-weight, self-emitting, wide view-angle and the potential of low cost [1], [2]. Among the three primary colors, blue OLED is a technical bottleneck since it typically exhibits a lower current efficiency and shorter lifetime than the red and green devices [3], [4]. Typically, there are two major guest/host systems reported to exhibit good device performances. One is the combination of distyrylarylene (DSA) derivative as the emitting host material and DSA amine styrylamine 4,4'-bis(2-(9-ethyl-9H-carbazol-3-yl)vinyl)biphenyl (BCzVBi) as the emitting dopant, which is first presented by Hosokawa, et al. in 1995 [5]. The other is demonstrated by Shi and his colleague at Kodak in 2002 [6], who doped 2,5,8,11-tetra(t-butyl)-perylene (TBP) into the diphenylanthracene derivatives 9,10-di(2-naphthyl)anthracene (ADN). Recently, with the blue dopant p-bis(p-N,N-diphenyl-aminostyryl)benzene (DSA-Ph) in the 2-methyl-9,10-di(2-naphthyl)anthracene (MADN), a blue OLED achieved a high EL efficiency of 9.7 cd/A at 20 mA/cm² and 5.7 V with CIE_{x,y} (0.16, 0.32) and a half-decay lifetime of 46000 h with an initial brightness of 100 cd/m² [7].

In our study, we use the 9,10-bis(2'-naphthyl) anthracene (ADN) as the host material and 4,4'-bis[2-(4-(N,N-diphenylamino)phenyl)vinyl]biphenyl (DPAVBi) as the dopant material in the emitting layer (EML) to achieve a high current efficiency. Besides, we used the bis(10-hydroxybenzo[h]quinolino)beryllium (Bebq2) as the electron transport layer (ETL) material that exhibits the electron mobility of about one orders of magnitude higher than that of the typically used ETL material, tris-(8-hydroxyquinoline) aluminum (Alq3) [8]. Hence, the carrier balance condition is achieved which effectively reduces the driving voltage and increases the power efficiency. In this paper, the experiment details are given in section 2. Experimental results and the related discussions are presented in section 3. Finally, a summary is drawn in section 4.

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2. EXPERIMENTS

The structure of the devices were ITO/ hole-transport layer (HTL) (40nm)/ EML (45nm)/ETL (15nm)/ LiF(1.2nm)/Al (100nm) that was optimized with combinational experiments and optical simulation. The organic materials of HTL is N,N'-diphenyl-N,N'-bis(1-naphthyl)-1,1'-biphenyl-4,4'-diamine (NPB). The EML consists of DPAVBi doped in the ADN matrix with concentrations from 0, 1, 2, 3, 4, 8, and 10%, which are labeled as device 1, 2, to device 7, respectively. OLEDs in this study are fabricated in multi-chamber cluster evaporator under the pressure about 1×10^{-7} torr. The substrate is indium-tin-oxide (ITO) glass with sheet resistance 10 Ohm/sqr. O₂ plasma treatment is used to clean the ITO substrate surface and to improve the ITO work function before the deposition. The deposition rates of organic materials are typically 0.1 nm/sec. After evaporative deposition in the vacuum chamber, the devices are transported to the glove box with O₂ and H₂O concentration below 1 ppm. Encapsulation is completed in the glove box. Desiccant is used to absorb moisture that leaks into the device.

After OLED Fabrication, we use Keithley 2400 source meter for obtain I-V characteristics. Minolta CS-1000 photometer is used to measure the luminance, the spectrum, and the CIE coordinate of the device. The active pixel size of a device is 0.5 mm x 0.5 mm. In our operation lifetime measurement, we drive the devices under DC constant current of initial brightness of 12500, 10000, 7500, 5000 cd/m² and recorded every 5 minutes. All these measuring instruments are connected to the personal computer and controlled by Labview programs.

In order to obtain the carrier transport characteristics, a transient electroluminescence (TREL) is used to measure the carrier mobility [9]. Our measurement system is shown in Figure 1. In the setup, we use a HP 8114A pulse generator to supply rectangular voltage pulses to our devices, with a repetition rate of 20 to 2000 kHz and a duty cycle of 0.5 %. Driving voltage of an OLED is varied from 6 to 14V. The series resistance is 2 k Ω . A silicon photodiode, connected to the oscilloscope, TDS210, was fixed on top of the emitting area to record the transient EL signals. We can see the photo-signal and electrical-signal shown on the oscilloscope display. Those data was recorded by a computer for evaluating the carrier mobility.

Fig. 1 Schematic diagram of the TREL measurement setup

3. RESULTS AND DISCUSSIONS

The resulting J-V curves are shown in Fig. 2. Turn-on voltages of the devices are around 2.5 V. When the current density is 100 mA/cm², the driving voltage is only about 6.1 to 6.7 V. We can note that dopant concentration did not affect the J-V characteristics very much. Typically, if a dopant organic material acts as a trap in a matrix, the driving voltage will increase with increasing dopant concentration. However, in our DPAVBi/ADN devices, the driving voltage is nearly the same. And no obvious trend of voltage-shift is observed with varying dopant concentration. Hence, we

attribute the voltage difference comes from the inevitably experimental errors. Compared with other OLEDs, the driving voltage of these devices are quite low. Such a low driving voltage comes from the use of the high electron mobility transport layer material, Bebq2.

Fig. 2 J-V curves of the different devices.

Figure 3 shows the current efficiency versus the current density. At 100 cd/m^2 , the current efficiencies with different dopant concentration were 8.5, 9.3, 17.6, 19.2, 18.3, 8.0, 9.3 cd/A, respectively. CIE coordinate of devices were (0.178, 0.213), (0.175, 0.220), (0.159, 0.238), (0.154, 0.238), (0.154, 0.242), (0.160, 0.270) and (0.159, 0.275), respectively. We can see the current efficiency increases then decreases with increasing dopant concentration. That is a typical concentration quenching effect. In our devices, the optimized dopant concentration is between 2 to 4 %. Within this region, current efficiency can be as high as 13 cd/A under the current density of 0.1 A/cm^2 . Such value is the highest value in a fluorescent blue OLED ever reported which comes from better charge-balance in our blue OLEDs. With increasing the dopant concentration, the x-value of CIE coordinates decreased then increased and the y-value increased monotonically. The initial blue shift results from the more efficient energy transfer from the host to the dopant with increasing dopant concentration. However, such a dopant material, DPAVB_i, was reported to have a hole-transport characteristic [5]. When the dopant concentration exceeds 6%, the holes inject into the EML and recombination zone shifts towards the cathode. Such an optical interference induced the anomalous x-value increase that was also confirmed by the optical simulation model.

Fig. 3 Current efficiency versus luminance of the eight devices.

Results of the accelerated operation lifetime of device 4 with different initial luminance values are shown in Fig. 4. With increasing the initial luminance value, the brightness decrease is faster. The half lifetime is 5.58, 16.56, 27, and 109.8hrs with initial luminescence 12500, 10000, 7500, and 5000 cd/m^2 respectively. We use the well-known equation $L \cdot t_{1/2}^n = C$ to obtain the half lifetime under different initial luminances [10]. Where n is the acceleration coefficient, $t_{1/2}$ is half lifetime. The best fitting acceleration coefficient value is 3.088. Hence, we can deduce that the estimated half lifetime 15611 hours is achieved at an initial luminance of 1000 cd/m^2 .

Fig. Operation stability with the dopant DPAVBi concentration at 3%

When the following two hypotheses are satisfied, the electron mobility of the EML with different dopant concentration can be obtained by using these devices utilizing TREL method. First, the hole mobility of NPB and the electron mobility of Beq2 is much higher than the electron mobility of EML, i.e., ADN with different dopant concentration of DPAVBi. Hence, the voltage drop is mainly across the EML layer. Second, the electron mobility is much larger than its hole mobility in EML and the recombination zone is at the NPB/EML interface. That means, when a voltage pulse is applied, the holes and electrons are injected from NPB and Beq2 rapidly and electrons transport through the EML and recombine the holes near the HTL/EML interface. In order to proof our assumptions, two further experiments are designed to obtain the hole mobility value of ADN those are: ITO/ NPB (10nm)/ DPAVBi:ADN (50 and 100 nm)/ NPB (10nm)/ Alq3 (15nm)/ LiF(1.2nm)/Al (100nm), those are labeled as device 8 and 9, respectively. In such a device, ADN is sandwiched by two NPB to ensure that there are only holes transporting through the ADN. In such a structure, the holes transport through the NPB, ADN and second layer NPB to recombine with electrons in Alq3.

Fig. shows the J-V curves of devices 8 and 9. Under the same current density, the driving voltage of device 9 is nearly double as compared to the device 8, which means that the applied voltage mainly drops on this layer. From this result we can estimate that hole mobility of NPB and electron mobility of Alq3 is much large than the hole mobility of ADN. That is to say, the time needed for a hole carrier injected from anode to travel through these organic layer and finally reach the NPB/Alq3 interface is primarily spent on ADN region. Therefore, we can determine the hole mobility value of ADN via the measurement of delay time, which is the duration that holes transport through the ADN. A green emission shown in Fig. 6 means the recombination zone was indeed at the NPB/Alq3 interface. The peak is very close to 517 nm, which is the emission peak of Alq3. That means the recombination zone is really at the NPB/Alq3 interface and delay time we measured in TREL system is does the duration that holes transport through the ADN.

Fig. 5 Current density versus voltage plot of devices 8 and 9.

Fig. 6 Spectra of devices 8 and 9.

Hence, the mobility of the EML can be obtained and can be fitted Poole-Frenkel model:

$$\mu = \mu_0 e^{\left(\frac{\beta\sqrt{E}}{kT}\right)}$$

Where μ_0 is the zero field mobility, β is Frenkel constant.

In room temperature(300k), the values of the μ_0 and β of the EML are:

$$(a) \mu_{e,EML} : \quad \mu_0 = 1.3757 \times 10^{-7} \text{ (cm}^2/\text{V*s)}, \quad \beta = 1.57 \times 10^{-23} \text{ (} J\sqrt{m/V} \text{)}$$

$$(b) \mu_{h,EML} : \quad \mu_0 = 4.3446 \times 10^{-11} \text{ (cm}^2/\text{V*s)}, \quad \beta = 2.50 \times 10^{-23} \text{ (} J\sqrt{m/V} \text{)}$$

Besides, we also measure the hole mobility of NPB and the electron mobility of Beq2 through the time-of-flight measurement [11] and obtain:

$$(c) \mu_{h,NPB} : \quad \mu_0 = 1.8 \times 10^{-4} \text{ (cm}^2/\text{V*s)}, \quad \beta = 1.53 \times 10^{-24} \text{ (} J\sqrt{m/V} \text{)}$$

$$(d) \mu_{e,Beq2} : \quad \mu_0 = 4 \times 10^{-5} \text{ (cm}^2/\text{V*s)}, \quad \beta = 6.3342 \times 10^{-24} \text{ (} J\sqrt{m/V} \text{)}$$

We can clearly see that the electron mobility value of the EML is one to two orders of magnitude higher than hole mobility regardless of the dopant concentrations. That means the ADN is kind of ETL material in our blue OLED. Besides, the DPAVB is not a trap inside the ADN host which is consistent with the J-V measurement since the driving voltage has no obvious trend between voltage-shift and dopant concentration. Also, the hole mobility of NPB and the

electron mobility of Beq₂ is much higher than that of electron mobility of ADN. That means the voltage drop is mainly on the EML during device operation and it explains the low driving voltage of our blue OLED.

4. CONCLUSIONS

In summary, we demonstrate a fluorescence blue OLED with high efficiency, low driving voltage, and good reliability. When the dopant concentration is 3%, there is a high current efficiency of 19.2 cd/A at 100 cd/m². The lifetime of device 4 (3% DPAVBi) is estimated about 15611 hours with initial luminance 1000 cd/m².

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