Research on the Performance of Blue-Green Tetra-Tandem Organic Light-Emitting Diodes

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Abstract

In this work, we present a blue-green tandem organic light emitting diodes (BG-OLEDs) with triplex blue emitting units and single green emitting unit. The analysis results of optical performances such as front efficiency, spectral variation, and viewing angle in different BG-OLED devices were explained. The blue-green tandem OLEDs introduced in this work provides a cost-effective way to extend the applications of QDs for full-color displays.

Author Keywords

Blue-green tandem OLED device; quantum dots; color conversion layer; full-color displays

1. Introduction

Quantum Dot-Organic Light Emitting Diodes (QD-OLEDs) use a QD film as a color conversion layer (CCL). The blue light of the QD-OLEDs comes from the electroluminescence (EL) of the blue OLED, and the green and red light come from the photoluminescence (PL) of the QD color conversion layer, which converts the blue light from the OLED to the green and red light of the QD [1].Among RGB OLEDs, the blue OLED has a short lifetime and low efficiency and deteriorates rapidly causing a burn-in issue. As representative technologies, the structure of tandem OLED was explored[2]. As the number of EL units of tandem OLED increases, the efficiency and lifetime of device improve since the high luminance can be achieved even at the low current. As a result, the tandem OLED structure shows the higher efficiency and lifetime compared with the single OLED [3].

In QD-OLED display, the conversion efficiency of green quantum dots by blue backlight is unsatisfactory. To enable efficient blue-to-green or blue-to-red photo-conversion, we adding a green emitting unit in the blue backlight, using blue and green backlight to conversion red and green quantum dots, to improve the brightness of QD-OLED display. In this work, we present a tandem OLED structure that combines triplex blue emitting units and single green emitting unit device to enhance the backlight brightness and lifetime.

2. Experiment

To fabricate the tandem OLED, the ITO/Ag/ITO patterns were engraved at the beginning, and the effective emitting area was about 3 mm × 3 mm. To ensure the substrate was thoroughly cleaned, the following procedures were followed: the substrate was soaked in acetone, isopropanol, and deionized water sequentially then cleaned using an ultrasonic vibration cleaner for 30 min. After that, the substrate was blown dry with 99% pure nitrogen (N2) and then put into an oven for 20 min to bake at a temperature of 150 ℃. Hence, moisture residue on the substrate could be avoided. In the next step, the organic and metallic layers were deposited using a thermal vacuum deposition system (at $4 \times 10{-}7$torr).

The organic material was deposited on the substrate at a rate of 0.5–1.5 Å/s in the order of hole injection layer/hole transport layer/ electron barrier layer/emitting layer/ hole barrier layer /electron transport layer (HIL/HTL/EBL/EML/HBL/ETL). Charge generate layer (CGL) was deposited on the substrate at a rate of 1 Å/s. The metal materials Yb/Mg/Ag cathode were deposited on the substrate at a rate of 0.1 Å/s and 3 Å/s, respectively.

3. Result and Discussion

3.1 Micro-cavity Theory

A planar micro-cavity structure consists of a reflective anode, a semi-transparent cathode, and organic layers sandwiched in between. The anode and cathode are parallel mirrors and they form a Fabry-Pérot resonator, which satisfies the following equation (1).

$$\frac{2\pi}{\lambda}\sum_m 2n_m\, d_m \cos\theta_0 - \phi_1(\lambda) - \phi_2(\lambda) = 2k\pi \quad (1)$$

where λ is the resonant wavelength or the peak wavelength; nm and dm are the refractive index and the thickness of the organic layer inside the two electrodes (the absorption of organic materials is neglected); θ_0 is the internal observation angle from the surface normal of the micro-cavity; $\Phi_1(\lambda)$ and $\Phi_2(\lambda)$ are the phase changes upon reflection corresponding to the effective reflectivity of R_1 and R_2 at the interfaces of the opaque anode/organic material and the organic material/ semi-transparent cathode, respectively; and k is the mode number.

When designing the tandem OLED device, the optical cavity conditions should be simultaneously satisfied in all units depending on the position of all emitting layers[4]. Indeed, it is difficult to simultaneously control the optical cavity conditions and match the superb charge balance property since the output spectrum intensity varies significantly with the device thickness variation [5]. According to the theoretical calculation and simulation, four structure OLEDs can be derived according to the position of the green light-emitting unit: GBBB, BGBB, BBGB, BBBG.

Table. 1 Tandem OLED device structure

Tandem OLED device structure			
GBBB	**BGBB**	**BBGB**	**BBBG**
Cathode	Cathode	Cathode	Cathode
ETL	ETL	ETL	ETL
EML4	EML4	EML4	EML4
HTL	HTL	HTL	HTL
CGL3	CGL3	CGL3	CGL3
ETL	ETL	ETL	ETL
EML3	EML3	EML3	EML3
HTL	HTL	HTL	HTL
CGL2	CGL2	CGL2	CGL2
ETL	ETL	ETL	ETL
EML2	EML2	EML2	EML2
HTL	HTL	HTL	HTL
CGL1	CGL1	CGL1	CGL1
ETL	ETL	ETL	ETL
EML1	EML1	EML1	EML1
HTL	HTL	HTL	HTL
Anode	Anode	Anode	Anode

3.2 Device Structure

The proportion of blue-green light emission intensity varies greatly with the position of the green light emitting unit in the tandem device. Equation (2) provides a qualitative understanding of the main effects produced by the micro-cavity. When the tetra-tandem device structure changes from GBBB to BBBG, the effective distance xi from the green emission dipole to the metal mirror increases, cos(4πx$_i$/λ) decreases, and the emission intensity of the green emission peak decreases.

$$|E_{cav}(\lambda)|^2 = \frac{\frac{(1-R_2)}{i}\sum_i\left[1+R_1+2(R_1)^{0.5}\cos(\frac{4\pi x_i}{\lambda})\right]}{1+R_1R_2-2(R_1R_2)^{0.5}\cos(\frac{4\pi L}{\lambda})}\times|E_{nc}(\lambda)|^2 \qquad (2)$$

Equation (2) provides a qualitative understanding of the main effects produced by the micro-cavity. where χ_i are the effective distances of the emitting dipoles from the metal mirror, R_1 and R_2 are the reflect distance of the metal and dielectric mirrors, respectively, L is the total optical thickness of the cavity, and $|E_{nc}(\lambda)|^2$ is the free space electroluminescence intensity at λ.

With the green emitting unit moves from semi-reflective cathode to full reflective anode, the green emission intensity is gradually increased, and the blue light emission intensity decreased. The main reason is that the longer the distance from the full reflective electrode is, the longer the transmission path of the light emitted by the light-emitting unit in the device is, and the OLED material has a certain absorption, so the light absorption ratio increasing and the light output ratio decreasing with the transmission path increasing.

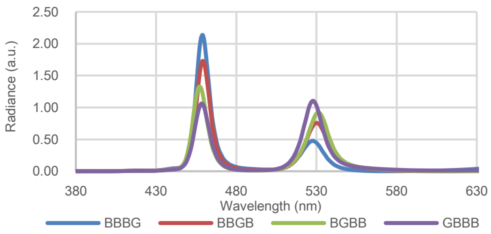

Figure. 1 Emission spectra of tandem OLEDs with different structures in current density 11mA/cm^2

Blue backlight can excite green and red light and emitting light from Blue pixels at the same time, green emitting unit can only excite red light and emitting light from G pixels, and can't emitting light from blue pixels, green light has at least one third loss, so priority to ensure the efficiency of blue light emitting unit is more beneficial to QD-OLED products. The structure of BBBG and BBGB devices is more suitable for the product requirements of QD-OLED.

Table. 2 IVL test data of tandem OLEDs with different structures

Structure	Voltage (V)	Luminance (cd/m^2)	Current Efficiency (cd/A)	EQE (%)	CIE_x	CIE_y	Main Peak (nm)	FWHM (nm)
BBBG	13.11	6683	60.15	41.89	0.159	0.158	459	10
BBGB	13.10	10350	93.15	43.35	0.161	0.253	459	10
BGBB	13.11	11640	104.76	38.04	0.168	0.349	459	11
GBBB	13.18	12790	115.11	44.18	0.169	0.330	457	11

3.3 View angle Dependence

View angle dependence is an important factors to evaluate OLED devices. Four structural devices were prepared respectively to evaluate the brightness attenuation under different viewing angles. It can be seen that the brightness of BBBG devices increases under large viewing angles. The CIE was remarkably shifted with the view angle increases, which leads to a large angle brightness increase.

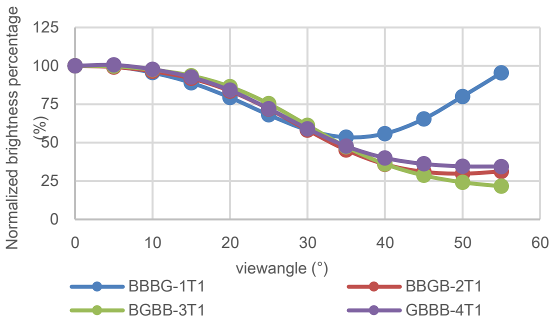

Figure. 2 The brightness of different tandem OLEDs under viewing angles ranging from 0° to 60°

The CIE shift of the other three devices was unobvious, and the brightness attenuation is the same as that of normal devices. Considering that QD-OLED display may have large viewing angles application scenarios, BBGB structure devices are more suitable for the requirements of QD-OLED display.

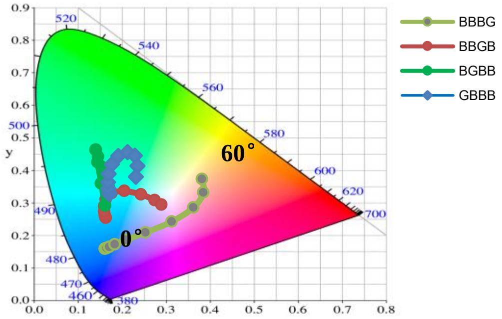

Figure. 3 The CIE color coordinates of different tandem OLEDs under viewing angles ranging from 0° to 60°

3.4 Lifetime Performance

The lifetime of the tetra-tandem device was tested under identical current (a) and blue brightness (b) conditions. The lifetime of BBBG and BBGB structure device is almost the same when test at the same current. When test at the same blue light brightness, due to higher blue light radiation intensity and lower current in BBBG structure device, the lifetime slightly surpasses that of BBGB device structures with a negligible difference.

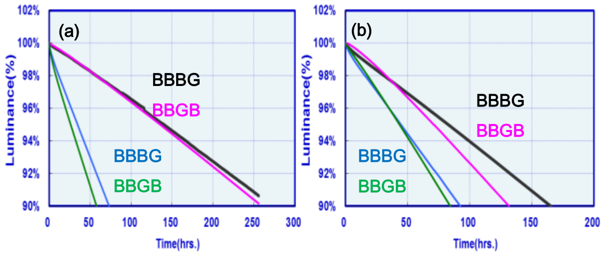

Figure. 4 Lifetime of tandem OLEDs with different structure

According to the requirement of QD-OLED display and the difference green-blue radiation intensity of tandem devices with different structures, view angle dependence characteristics, device lifetime and other aspects, the BBGB device is suitable for QD-

OLED display.

The OLED device structure optimization is mainly divided into optical and electrical performance, the optical performance mainly adjust the length of the device micro-cavity, which needs to satisfy simultaneously of the device micro-cavity length and the emission spectrum of the OLED device. The electrical properties mainly adjust the thickness of each film layer and doping ratio, optimize the excitions recombine center of the emitting layer. Since the optical and electrical properties of OLED devices affect each other, the electrical and optical properties of OLED devices are optimized respectively.

3.5 Electrical and Optical Performance

In order to avoid the optical performance influence of the device, bottom emitting device is used to optimize the electrical performance, and the micro-cavity of the bottom emitting device is weak, which can minimize the interference of the micro-cavity on the test results. To optimize the electrical performance of the device, the effect of the device is improved by changing the ETL thickness, ETL doping ratio, N-CGL and P-CGL doping ratio.

By optimizing the film layer which has great influence on the electrical performance of the device, the optimal device structure is ETL doping ratio 7:3, N-CGL doping ratio 9%, P-CGL doping ratio 9%.

Optical performance mainly optimizes the efficiency of the device by adjusting the length of the device cavity and the distance from the luminous center to the full reflect anode. Through the blue and green light spectra main peak position determines the cavity length of the tetra-tandem device, through the single-layer device to confirming the position of the first blue emitting unit, through the bi-tandem device to determining the position of the second blue light emitting unit, adjust the green emitting unit to the reflect anode position, optimizes the device structure with the highest efficiency to determine the green light emitting unit position, and finally determines the third blue light emitting unit position.

After optical performance optimization, the green light emission intensity is also unsatisfactory, which was inferred that the green emitting unit excitons recombine center is offset of the emitting layer. Therefore, we doping red dye in the green emitting layer to demonstrate the excitons recombination position of the green emitting unit.

It is found that the excitons recombine position of the green light-emitting unit is at the interface of the light-emitting layer and the electron barrier layer, indicating that the number of electrons in the green emitting unit is more than that of holes. The carrier balance of the emitting layer needs to be optimized by increasing the number of holes or decreasing the number of electrons.

The electrons and holes in the green light-emitting layer are mainly generated by N-CGL3 and P-CGL2 then transferred to the emitting layer through ETL and HTL, and HTL hole mobility is high, which has little influence on the number of holes in the emitting layer. Therefore, the effect of the device is observed mainly by reducing the NCGL doping ratio or increasing the ETL thickness to reduce the number of electrons, and increasing the PCGL doping ratio to increase the number of holes. By optimizing the carrier balance of the green light emitting unit, when the P-CGL doping ratio is 9%, the device voltage is lower, and the green radiation strength is obviously increased. This indicates that the excitons recombine center has moved to the center of the emitting layer.

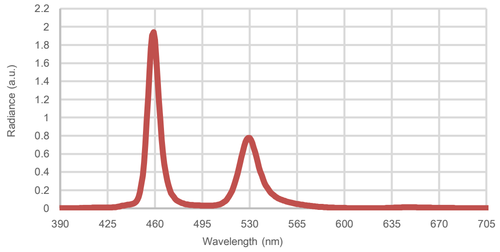

Figure. 5 Emission spectra of top emitting tetra-tandem OLEDs in current density 11mA/cm^2

4. Conclusions

In summary, we fabricated a blue-green tandem OLED devices and analyzed the optical performances such as EL spectra, front efficiency increase ratio, viewing angle and lifetime characteristics. The optical and electrical performance optimization of tetra-tandem device have been described in detail, and the optimal device structure suit for QD-OLED was achieved. We believe that this study provides an effective and convenient method to fabricate full-color QD-OLED displays for practical applications.

5. Reference

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