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High efficiency phosphorescent organic light-emitting diode by incorporating an electron transport material into emitting layer

Fuh-Shyang Juang[1,*], Shun-Hsi Wang[1], Yu-Sheng Tsai[1], Bo-Syen Hsieh[1], Yun Chi[2], and Han-Ping, Shieh[3]

[1] Institute of Electro-Optical and Materials Science, National Formosa University, Yunlin, 632, Taiwan
Phone: +886-5-631-5029 E-mail: fsjuang@seed.net.tw
[2]Department of Chemistry, National Tsing Hua University, Hsinchu, 300, Taiwan
[3]Department of Photonics & Display Institute, National Chiao Tung University, Hsinchu, 300, Taiwan

1. Introduction

Organic light emitting diodes (OLEDs) have recently received much attention due to their potential applications in solid-state lighting [1-4] and flat-panel displays. The charge balance is an important factor in OLED performance. However, carrier transport in most OLED devices is highly imbalanced. In this study, the electron transport material was doped into the emitting layer (EML) as a mixed-host structure to balance the carriers and confine the recombination zone within EML. Furthermore, the high efficiency white PHOLED was produced by doping $Os(bpftz)_2(PPh_2Me)_2$ into buffer layer (TCTA). The effects of carrier transport layer thicknesses on the optoelectronic properties of PHOLEDs were also discussed.

2. Experimental

PHOLEDs were fabricated on pre-patterned ITO substrates with a sheet resistance of 13.59±5 Ω/square. The substrates were cleaned via acetone, isopropyl alcohol, DI water followed by O_2 plasma treatment. All organic and metal layers were vacuum deposited under 2×10^{-6} Torr. A 40 nm thick of EHI608 (produced by e-Ray EO Tech. Co., Ltd.) used as hole injection layer, 1,1-bis[(di-4-tolylamino) phenyl] cyclohexane (TAPC) used as hole transport layer (HTL), 4,4',4''-tris(N-carbazolyl) triphenylamine (TCTA) [5] doped with 10 wt% of ($Os(bpftz)_2(PPh_2Me)_2$) [6] used as red-EML, intrinsic TCTA used as the interlayer (IL), 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) [7] mixed into the TCTA (1:1) used as host of blue-emission, iridium-bis-(4,6- (difluorophenyl-pyridinato-N,C2) (FIrpic) used as blue dopant, and ris[3-(3-pyridyl)-mesityl]borane (3TPYMB) [8] used as the electron transport layer (ETL). The cathode consisted of 0.5 nm thick CsF followed by 200 nm thick Al. Table I shows the adjustment parameters for different PHOLED structures and Figure 1 shows the energy band structure of white device with the mixed-host structure (TmPyPB:TCTA). Spectra Scan PR650 and Keithley 2400 were employed to measure the current-voltage-luminance (I-V-L) characteristics.

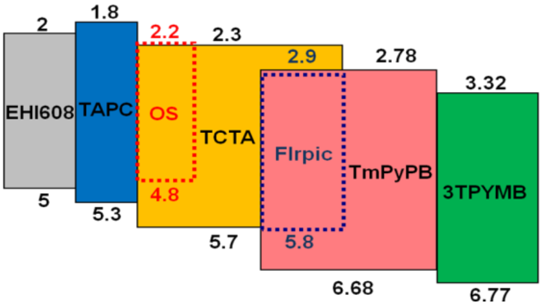

Fig. 1. Energy band structure of white PHOLED.

3. Results and discussion

Hole mobility of TAPC (~1.0×10^{-2} cm^2/Vs) is two orders of magnitude higher than the electron mobility of TmPyPB (~7.0×10^{-4} cm^2/Vs). Hence, it is expected that the OLED device is strongly hole dominant. To improve the charge balance, hole injection amount and barrier were adjusted by inserting a hole transport-type host (TCTA) as the buffer layer between HTL and EML. Moreover, the hole transport-type host (TCTA) was combined with TmPyPB as a mixed-host structure. The introduced mixed-host structure was effective to improve the carrier injection and balance due to the highly electron mobility of TmPyPB and carrier can directly injected from intrinsic interlayer (TCTA or TmPyPB) to EML. In addition, the high triplet energy (T_1=2.78) of TmPyPB led to triplet exciton was effectively confined within EML. The charge carrier confine ability is dominated to the energy barrier between HOMO and LUMO level of EML with nearby carrier transport layers. Hence, the used mixed-host structure exhibited a larger energy barrier, which resulted in the effectively confinement of charge carrier and recombination zone within ultra-thin EML (10 nm). Device A shows a power efficiency 12 lm/W and a driving voltage 4.8 V at a luminance 1000 cd/m^2 (as shown in Table II, Fig. 2 and 3).

Table I Parameters for different PHOLEDs. (unit: nm)

No	HTL	R-EML	Buffer layer	B-EML	ETL-1	ETL-2
	TAPC	(concen.)	TCTA	(concen.)	Tm	3TP
A	20	-	7	FIr:TCTA:Tm (0.2: 0.4: 0.4) 10	5	20
B	25		5			
C						35
D (White)		Os: TCTA (0.1: 0.9) 3	intrinsic interlayer 2			45

As discussed above, carrier transport in most OLED device is highly imbalanced. In order to improve the charge balance in OLED, thicknesses of HTL (TAPC) and buffer layer (TCTA) was adjusted (Device A and B, Table I), respectively. Hole injection amount and transport distance from HTL to EML can be effectively controlled due to the different hole mobility of TAPC (~1.0×10^{-2} cm^2/Vs) and TCTA (1.0×10^{-4} cm^2/Vs). From the results, Device B exhibited a enhanced power efficiency of 14.3 lm/W (see Fig. 3) and the driving voltage decreased from 4.8 V to 4.3 V at a luminance of 1000 cd/m^2 (see Table II and Fig. 2).

Table II Optoelectronic properties of PHOLEDs at 1000cd/m^2

No	at 1000 cd/m^2		CIE at 3V	CIE at 8V
	V (volt)	J (mA/cm^2)		
A	4.8	5.45	(0.142, 0.271)	(0.142, 0.267)
B	4.3	5.1	(0.142, 0.276)	(0.141 0.273)
C	4.4	3.2	(0.143, 0.306)	(0.145, 0.308)
D	5	3.1	(0.279, 0.358)	(0.327, 0.314)

Next, the thickness of ETL (3TPYMB) was adjusted (Device C and B, Table I) to improve the electron current. From I-V curves of Fug. 2, it is observed that the increased ETL thickness leads to the current density of device significantly decreased. This is attributed to the total resistance of device increased. However, the hole mobility of TAPC is three orders of magnitude higher than the electron mobility of 3TPYMB (~1.0×10^{-5} cm^2/Vs). This is indicated that the resistance of TAPC is lower than that of 3TPYMB. Therefore, the mainly cross pressure of device was situated on 3TPYMB. Although the total current density of device decreased with increasing the ETL thickness, the cross pressure of ETL became large. However, the larger cross pressure led to the electron current increased. This indicated that the increased ETL thickness can improve the electron current, which results in the carrier balance and luminance efficiency enhancement. Figure 4 shows the normalized electro-luminescence (EL) spectra of blue PHOLEDs with different ETL thicknesses. As the hole injection reduced (Device B) and ETL thickness increased (Device C), the shoulder peak around 474 nm was increased. It is suggestion that the amount of electron was huger than the hole, which resulted in the optical effect through the recombination zone shift [9]. Device C shows a power efficiency 22.3 lm/W at a luminance of 1000 cd/m^2 (see Fig. 3).

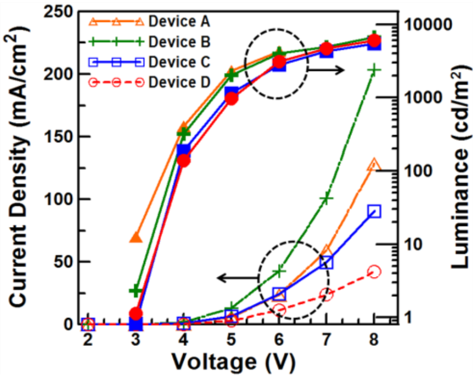

Fig. 2. I-V-L characteristics of different PHOLEDs.

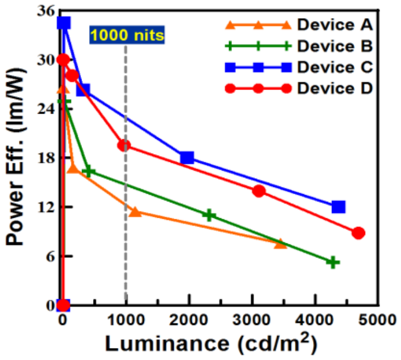

Fig. 3. Power efficiency-luminance curves of different PHOLEDs.

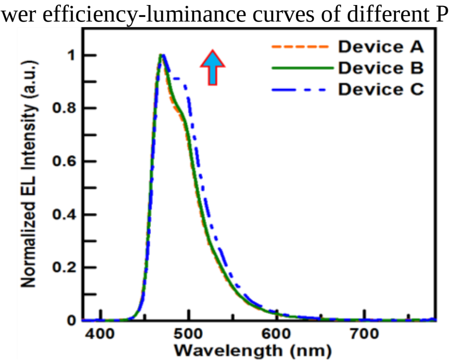

Fig. 4. EL spectra of different blue devices.

In addition, the osmium complex (Os(bpftz)$_2$(PPh$_2$Me)$_2$) was doped into a portion of buffer layer (TCTA) as red-emitter and combined with blue-emitter to product white PHOLED device (see Device D, Table I). The thin intrinsic interlayer (TCTA) can prove the diffusion of triple exciton from blue-EML to red-EML. Furthermore, by optimizing the device parameters, white PHOLED with a power efficiency 20 lm/W at the luminance of 1000 cd/m^2 can be achieved (see Fig. 3). From Table II and Fig. 5 the normalized EL spectra of Device D under different driving voltages, we could be observe the CIE coordinates red-shifted from (0.279, 0.358) to (0.327, 0.314). This is attributed to energy transferred to the lower-energy excited states of Os complexes at high-energy excitation densities. And the low-lying HOMO level of Os (4.8 eV) is substantially lower than those of TCTA, TmPyPB and FIrpic. It is well expected that the Os functioned as an effective hole traps in Device D since the red emission from Os was generally increased under higher voltage. In addition, the blue emission reduced slightly, due to the recombination zone moved to HTL/EML interface with bias increasing. At the practical brightness of 1000 cd/m^2 (driving voltage 4.5 V), Device D exhibits efficiencies of 32 cd/A and 20 lm/W with the CIE coordinate of (0.28, 0.35). And the maximum efficiencies of 36 cd/A and 30 lm/W could be achieved.

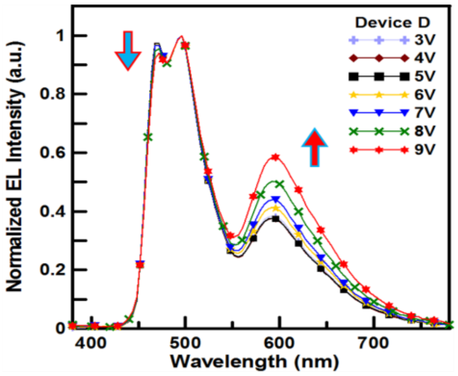

Fig. 5. EL spectra of white device under different biases.

3. Conclusions

The performances of PHOLEDs could be enhanced by doping ETL into EML as a mixed-host. Mixed-host structure device of TCTA:TmPyPB showed effective confined of recombination zone as well as balanced charge carrier injection, which resulted in highly improved efficiency of PHOLEDs. At a luminance of 1000 cd/m^2, blue PHOLED exhibited a better power efficiency 22.3 lm/W. Furthermore, a highly efficiency white PHOLED can be achieved by doping Os(bpftz)$_2$(PPh$_2$Me)$_2$ into buffer layer (TCTA) as red-emission and optimizing the thicknesses of buffer layer, HTL and ETL. From the experimental results, the optimization structure of white PHOLED shows a luminance efficiency 32 cd/A, power efficiency 20 lm/W and a low driving voltage of 4.5V at a luminance of 1000 cd/m^2 with the CIE coordinate of (0.28, 0.35). And the maximum luminance of 10200 cd/m^2, yield of 36 cd/A @ 4 V and power efficiency of 30 lm/W @ 3 V was obtained.

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