Microelectronics Reliability xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Microelectronics Reliability

journal homepage: www.elsevier.com/locate/microrel

Simple-structured efficient white organic light emitting diode via solution process

Deepak Kumar Dubey, Meenu Singh, Snehasis Sahoo, Jwo-Huei Jou *

Department of Materials Science and Engineering, National Tsing Hua University, Hsinchu 30013, Taiwan

ARTICLE INFO

Article history:
Received 14 January 2017
Received in revised form 6 July 2017
Accepted 18 July 2017
Available online xxxx

Keywords:
OLEDs
Single emissive layer
Wet process
Chromaticity stable

ABSTRACT

High-efficiency white emission is crucial to the design of energy-saving display and lighting panels, whereas solution-process feasibility is highly desirable for large area-size and cost-effective roll-to-roll manufacturing. In this study, we demonstrate highly-efficient, bright and chromaticity stable white organic light emitting diodes (OLEDs) with solution-processed single emissive layer. The resultant best white OLED shows excellent electroluminescence performance with forward-viewing external quantum efficiency, current efficiency and power efficiency of 22.7%, 48.8 cd A^{-1} and 27.8 lm W^{-1} at 100 cd m^{-2}, respectively, with a maximum luminance of 19,590 cd m^{-2}. Furthermore, we also observed an increment of 112% in the power efficiency, 86.9% in the current efficiency and a decrement of 39.2% in the external quantum efficiency at 100 cd m^{-2} as the doping concentration of blue dye was increased from 10 wt% to 25 wt% in the devices. The better efficiency performance may be attributed to the effective exciton-confining device architecture and low-energy barrier for electrons to inject from the hole-blocking electron-transport layer to the host layer.

1. Introduction

After the demonstration of first succinct organic light-emitting diode (OLED) with two-layer structure in 1987 by C.W. Tang [1], flat-panel displays and lighting applications based on OLED technology have grown dramatically because of their attractive features such as simple fabrication process, ultra-thin structure, wide viewing angles, impressive color rendering, light-weight, and high compatibility with flexible substrates [2,3]. In particular, white light-emitting OLEDs are known to be an ideal light source, which not only acts as a backlight for OLED display [4] but also can be utilized as an area light source for decorative and general lightings, or lighting in galleries, hospitals, and museums, because OLEDs do not emit ultraviolet radiation [5].

Typically, OLEDs are fabricated using either vacuum evaporation or solution process. Current white OLEDs consist of two or more multiple layers of different materials in precise optoelectrical design [6]. Such multilayer structures allow the separation of the charge-injecting, charge transporting and light-emitting functions to the different layers, thus leading to a marked increase in efficiency and lifetime [7], which can be easily achieved stepwise in vacuum evaporation process. However, the vacuum deposition technique is faced with many shortcomings such as inefficient utilization of materials and high energy consumption, which remains a great challenge for the mass-production of cost-effective white OLEDs [8,9]. However, solution-processed is deemed more superior in enabling flexible, large area size roll-to-roll production, and consequently more cost-effective [10,11]. In the solution based fabrication process, OLED devices generally exhibit efficiency much lower than their dry-processed counterparts, especially at high luminance. Markedly improving the efficiency of solution-processed OLEDs is hence crucial.

To achieve high device efficiency, three major approaches can be applied, namely, (i) design and synthesis of electroluminescent (EL) active materials, (ii) design and employment of efficiency-effective device architectures, and (iii) incorporation of internal and external light out-coupling technologies. From a device structure perspective, it includes the employment of low interfacial resistance P — I — N structures [12], low carrier-injection barriers [13], balanced carrier injection [14], carrier and exciton confinement [15], stepwise emissive layers [16], carrier modulation layers [17], structures enabling exciton to generate on host or on both host and guest [18], structures facilitating host-to-guest energy transfer [19], and co-host structures [20]. Mostly all are applicable only for vacuum deposition technique. Only a few approaches can be utilized in the solution process unless any certain special techniques are adopted.

Therefore, it is highly desirable to develop a newly effective device that can render various channels to harvest all the excitons together with a reduced efficiency roll-off. In this work, we demonstrate the feasibility of fabricating a single emissive layer white OLED via solution process using two white light complementary color approach to avoid device complexity and to reduce cost. The resultant white OLED shows at 100 cd m^{-2} an external quantum efficiency of 22.7%, current efficiency 48.8 cd A^{-1} and power efficiency 27.8 lm W^{-1} with a maximum luminance of 19,590 cd m^{-2} without using any light out-coupling techniques.

* Corresponding author.
E-mail address: jjou@mx.nthu.edu.tw (J.-H. Jou).

http://dx.doi.org/10.1016/j.microrel.2017.07.076
0026-2714/

Table 1
Effect of the doping concentrations of blue dye, Flrpic, on the operation voltage (OV), power efficiency (PE), current efficiency (CE), external quantum efficiency (EQE), CIE coordinates, and maximum luminance of the white OLED devices studied with a wet processed, single emissive layer.

Device	Host	Guest		OV (V) PE (lm W^{-1}) CE (cd A^{-1}) EQE (%) CIE_{xy}[a]					Maximum Luminance (cd m^{-2})
		Blue dopant (wt%)	Yellow dopant (wt%)	@ 100/1000 cd m^{-2}					
W1	CBP	10	1	6.2/6.8	13.1/15.4	26.0/33.3	22.7/11.3	(0.44, 0.48)	19,590
W2	CBP	15	1	5.5/6.2	16.0/22.7	28.0/44.0	14.3/14.8	(0.44, 0.48)	19,240
W3	CBP	20	1	5.5/6.0	18.7/19.9	32.6/37.7	14.3/13.0	(0.44, 0.48)	18,420
W4	CBP	25	1	5.5/6.0	27.8/20.7	48.6/39.5	13.8/12.1	(0.44, 0.48)	18,610

[a] CIE_{xy} coordinates of studied WOLEDs at @100 (cd m^{-2}).

2. Experimental

2.1. Device fabrication

All the devices are composed of a 125 nm indium tin oxide (ITO) anode layer, a 35 nm poly(3,4-ethylene-dioxythiophene)-poly(styrenesulfonate) (PEDOT: PSS) hole injection layer (HIL), following a 30 nm single emissive layer (EML), a 32 nm 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi) electron transporting layer (ETL), a 0.8 nm lithium fluoride (LiF) electron injection layer (EIL), and a 150 nm aluminum (Al) cathode layer. Among, all the layers are deposited via spin-coating, while the ETL, EIL, and cathode are deposited via dry process.

The fabrication process included first spin-coating an aqueous solution of PEDOT: PSS at 4000 rpm for 20 s to form a HIL on a pre-cleaned ITO anode and then dried at 150 °C for 30 min to remove residual solvent. Before depositing the EML, the solution is prepared by dissolving the host and guest molecule in tetrahydrofuran at 50 °C for 40 min with stirring. The resulting solution is then spin-coated at 2500 rpm for 20 s under nitrogen and again the substrate is baked at 150 °C for 30 min to remove residual solvent. Followed by the TPBi, LiF, and Al, are deposited by thermal evaporation under high vacuum (5×10^{-4} Torr).

2.2. Device measurements

All devices were measured under atmospheric condition. The luminance, Commission International de l'Eclairge (CIE) chromaticity coordinates, and electroluminescence spectrum of the resultant devices were measured by using a Photo Research PR-655 spectrum. A Keithley 2400 electrometer, together with a Minolta CS-100 luminance meter, was used to measure the current–voltage (I–V) characteristics. The emission area of the devices was 2.5 mm^2, and only the luminance in the forward direction was measured.

3. Results and discussion

We fabricated solution-processed single emissive layer white OLEDs W1 to W4 by incorporating x wt% (x = 10, 15, 20, and 25) blue emitter, iridium (III) bis[(4,6-difluorophenyl)pyridinato-*N*,C2′]-picolinate (FIrpic) and a 1 wt% yellow emitter, iridium (III) bis(4-phenylthieno[3,2-*c*]pyridinato-*N*,C2′)acetyla-cetonate (PO-01) into 4, 4′-*N*, *N*′-dicarbazole-biphenyl (CBP) with the following device structure: ITO/PEDOT: PSS/1 wt% PO-01 and x wt% Flrpic doped in CBP/TPBi/LiF/Al.

Table 1 summarizes the electroluminance characteristics of the studied white OLEDs, including power efficiency, current efficiency, external quantum efficiency (EQE), operation voltage, CIE coordinates, and maximum luminance. Fig. 1(a) and (b) show the detailed device structure for all the studied OLEDs and their corresponding energy level diagram.

Fig. 2(a) shows the normalized electroluminescent spectra (EL spectra) of the studied white OLEDs at 1000 cd m^{-2}. The EL spectrum exhibits two primary emission peaks at 465 and 570 nm, which are attributed to the emissions of Flrpic and PO-01, respectively. All the fabricated device emitted white light having CIE coordinates of (0.44, 0.48) with broad spectra from 400 to 750 nm at 1000 cd m^{-2} luminance. Fig. 2(b) shows the EL spectra of device W4 at different operating voltages. The device W4 shows CIE coordinates of (0.448, 0.482) at 1000 cd m^{-2} and (0.442, 0.480) at 5000 cd m^{-2}, as shown in Fig. 4. The CIE coordinates only vary (0.002, 0.0005) per 1000 unit.

The reason why the spectra of white OLEDs are fairly stable may be attributed to three phenomena. Frist, due to the use of a single emissive layer and an effective carrier-confining architecture in the device, excitons can be effectively confined and recombine within the desired emissive layer [21]. Therefore, the recombination zone will barely shift as the applied voltage increases. Second, both guest molecules are uniformly dispersed throughout the entire thickness in the emissive layer and excitons can hence be generated uniformly. Third, all the resulted white OLEDs employed comparatively low doping concentrations so that less exciton quenching and hence less color variation will occur at high voltage [22].

At high voltage, we observed that CIE coordinate "x" changes more as comparing to CIE coordinate "y", therein leading to a stable emission. It can be attributed to the fact that at a higher voltage, the additional exciton energy could be more effectively transferred to the blue emitter

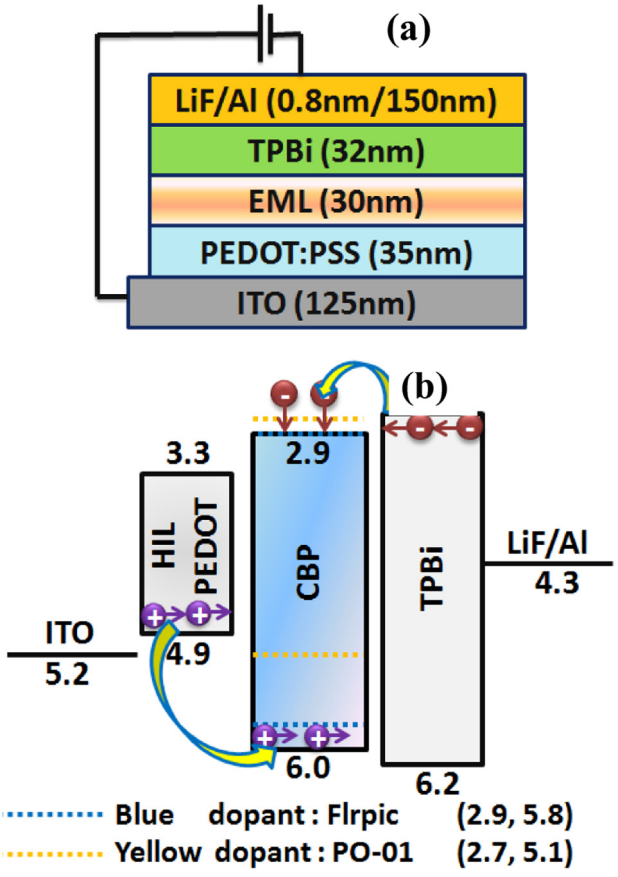

Fig. 1. (a) and (b) show the detailed device structure and schematic energy levels diagram of the studied white OLEDs, respectively.

Please cite this article as: D.K. Dubey, et al., Simple-structured efficient white organic light emitting diode via solution process, Microelectronics Reliability (2017), http://dx.doi.org/10.1016/j.microrel.2017.07.076