Journal of Luminescence 142 (2013) 86–91

Contents lists available at SciVerse ScienceDirect

Journal of Luminescence

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

Color-tunable and stable-efficiency white organic light-emitting diode fabricated with fluorescent-phosphorescent emission layers

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ARTICLE INFO

Article history:
Received 7 December 2012
Received in revised form
20 March 2013
Accepted 29 March 2013
Available online 10 April 2013

Keywords:
Organic light emitting diode
Fluorescence
Phosphorescence
Tunability

ABSTRACT

White organic light emitting diodes (OLEDs) were fabricated for color-tunable lighting applications. Fluorescent and phosphorescent hybrid emission layers (EMLs) were used to enhance the luminance and stability of the devices, which have blue-EML/CBP interlayer/green-EML/phosphorescent-sensitized-EML/red-EML structures. The influence of the composition and structure of the EMLs on the electroluminescence properties of the devices were investigated from the viewpoint of their emission spectra. The possible exciton harvesting, diffusion, transport, and annihilation processes occurring in the EMLs were also evaluated. A maximum luminance intensity of 7400 cd/m^2 and a highly stable current efficiency of 3.2 cd/A were obtained. Good color tunability was achieved for the white OLEDs; the chromatic coordinates linearly shifted from pure white (0.300, 0.398) to cold white (0.261, 0.367) when the applied voltage was varied from 10 to 14 V.

1. Introduction

As photoelectric technologies have been developed, organic light emitting diodes (OLEDs) have made a crucial technology revolution possible for flat-panel displays and lighting sources [1–4]. OLEDs are distinguished by their extremely low thicknesses, high flexibility, low weights, fast responses, and low power consumption. Nowadays, OLEDs are widely used in commercial applications, most commonly in mobile phones, portable media players, and tablet computers. OLEDs have been used extensively not only in displays but also for lighting applications. In general, the power efficiency is about 10–20 lm/W for electric bulbs and 60–90 lm/W for fluorescent lamps. Although the efficiency and lifetime of OLED lamps are comparable with those of fluorescent lamps, OLED lamps are more flexible and environmentally friendly.

For OLED indoor lighting, lamps should be bright sometimes for clear visibility, whereas at other times be of a relaxing, muted color. Therefore, color-tunable white OLEDs are desirable so that the emission color can be changed to provide either sunlight-like illumination or a more muted color depending on the specific requirements [5,6]. On the other hand, a color-tunable white OLED is also required for OLED backlighting applications in order to produce balanced lighting performance. For color-tunable white OLEDs, the emission color must be tunable from typical warm white, to bright sunlight, to cold white. These colors correspond to correlated color temperatures (CCTs) of 2700, 6500, and 7000 K, respectively. Cold white OLEDs are used for the screens of liquid crystal displays.

Jou has demonstrated OLEDs with wide CCT span from 2300 to 8200 K as well as sunlight-style CCT-tunable lamps [7,8]. The Commission Internationale del'Eclairage (CIE) chromaticity of this device can be varied from (0.48, 0.42) to (0.28, 0.29) by changing the voltage from 3 to 9 V. This chromaticity change occurs because of shifts in the carrier recombination zone of the device. Lee used an OLED structure of fluorescent blue/phosphorescent green/phosphorescent red emission layers (EMLs) to produce RGB white emission with a high brightness of 24,000 cd/m^2 and a CIE chromaticity ranging from nearly pure (0.30, 0.35) to warm white (0.35, 0.40) [9]. Choy reported that if a carrier barrier layer was introduced between the EMLs to shift the region in which the color could be tuned, the emission color could be adjusted by changing the voltage [10]. The color tunability of OLEDs was also discussed by Kim and Li [11,12]. Table 1 lists the structures of white OLEDs with fluorescent-phosphorescent (FP) hybrid EMLs; all the devices had unique features and showed good performance [8,9,13–17].

Because of the unlimited potential of white OLEDs for solid-state lighting applications, color-tunable white OLEDs were investigated in this study. The EML was deposited with a structure of blue-EML/interlayer/green-EML/phosphorescent sensitized (PS)-EML/red-EML, in which RGB and FP hybrid EMLs were prepared for obtaining a broadband white emission. An interlayer for manipulating exciton diffusion was deposited, and a PS-EML was used to enhance the luminance and stability of the devices.

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http://dx.doi.org/10.1016/j.jlumin.2013.03.060

Table 1
Structures and features of white OLEDs with FP EMLs.

Device structure	Feature	Ref.
BCPO:FIrpic/TPBi/TCTA:Ir(ppy)$_3$/TPBi: Ir(ppy)$_3$:PO-01:Ir (piq)$_2$(acac)	Three EMLs with different host materials; using TPBi as CML and ETL; four blackbody radiation-complementary emitters	[8]
CBP:BCzVBi/CBP/CBP:Ir(ppy)$_3$/CBP:Ir(piq)$_3$	Stacked F-blue/P-green/P-red EMLs; undoped CBP as singlet-exciton separator	[9]
mCP/mCP:FCNIrpic/TPBiI:Ir(ppy)3:Ir(pq)$_2$(acac)/TPBi	Higher triplet energy of undoped mCP and TPBi leading to effective confinement of triplet excitons in EML	[13]
mCP:Ir(ppy)$_3$:DCJTB/ mCP:Ir(ppy)$_3$/mCP/ mCP:FIrpic	Using selectively codoped layer to manage energy transfer path and to enable efficient phosphorescent sensitization	[14]
NPB/NPB:DCJTB/CBP/CBP:Ir(piq)$_3$/TPBi	Solving efficiency roll-off issue with a FIP EML structure	[15]
mCP/mCP:FIrpic:DCJTB/TAZ:Ir(ppy)$_3$/TAZ	Reducing population of triplet excitons in the EML and using phosphor doped ETL to recycle excitons	[16]
CBP:BCzVBi/CBP/CPB:Ir(ppy)$_3$:DCJTB/CBP/CBP:BCzVBi	Employing a phosphor sensitizer combined with blue and red fluorescent dopants	[17]

CML, carrier modulating layer; ETL, electron-transporting layer; F, fluorescence; P, phosphorescence; FIP, fluorescence-interlayer-phosphorescence.

The device showed good efficiency stability and white color tunability. The electroluminescence (EL) properties were analyzed and investigated in detail.

2. Experimental process

For the preparation of white OLEDs, an indium tin oxide (ITO) glass with a resistivity of 15 Ω/sq was used as the substrate. In addition, N,N′-bis(naphthalen-1-yl)-N,N′-bis(phenyl)-benzidine (NPB), 4,7-diphenyl-1,10-phenanthroline (Bphen), 4,4′-bis(carbazol-9-yl)biphenyl (CBP), 4-(dicyanomethylene)-2-tert-butyl-6-(1,1,7,7-tetramethyljulolidin-4-yl-vinyl)-4H-pyran (DCJTB), 1,4-bis[2-(3-N-ethylcarbazoryl)vinyl]benzene (BCzVB), and tris-(phenylpyridine)-iridine [Ir(ppy)$_3$] were used as organic source materials. Herein, 30-μm-thick NPB and 50-nm-thick Bphen layers were used for the hole-transporting layer (HTL) and electron-transporting layer (ETL), respectively [18]. Bphen not only shows a high conductivity (5.2×10^{-4} cm^2/V s) for electrons but also can block holes in the emission layer [19]. Thus, it increases the recombination rate of electron-hole pairs. Furthermore, CBP was used as the host material in all EMLs [15]. BCzVB [20], Ir(ppy)$_3$ [17], and DCJTB were selected as the blue, green, and red dyes, respectively [21].

For device fabrication, the ITO surface was cleaned with an organic solvent, dried with high-purity nitrogen gas, and then subjected to oxygen plasma treatment. All the organic materials were evaporated on the ITO glass layer by layer at a deposition rate of 0.5–0.8 Å/s at 5×10^{-6} Torr. The deposition rate of the materials can be controlled by adjusting the heating current provided to the crucible. The thicknesses of the films were monitored in situ using a 5 MHz quartz crystal thickness monitor. Finally, a 1-nm-thick LiF electron injection layer and 200-nm-thick Al cathode were deposited on top of the organic film at a deposition rate of 2–3 Å/s. The emission area of the devices was 0.24 cm^2. To measure the EL properties of the devices, a Keithley 2410 programmable voltage–current source was used. The luminance and CIE coordinates were measured using a Minolta Chroma Meter model CS-100A, and a Newport OSM-400 spectrophotometer was used to measure the EL spectra at the maximum luminance intensity of the devices. All the measurements were carried out in air at room temperature, and the devices were not encapsulated.

3. Results and discussion

In this study, a fluorescent-phosphorescent hybrid white OLED with multiple EMLs was fabricated and its characteristics were evaluated. The device structure and energy level diagram are shown in Fig. 1(a). The emission and absorption spectra of the CBP, BCzVB, Ir(ppy)$_3$, and DCJTB are shown in Fig. 1(b). Good

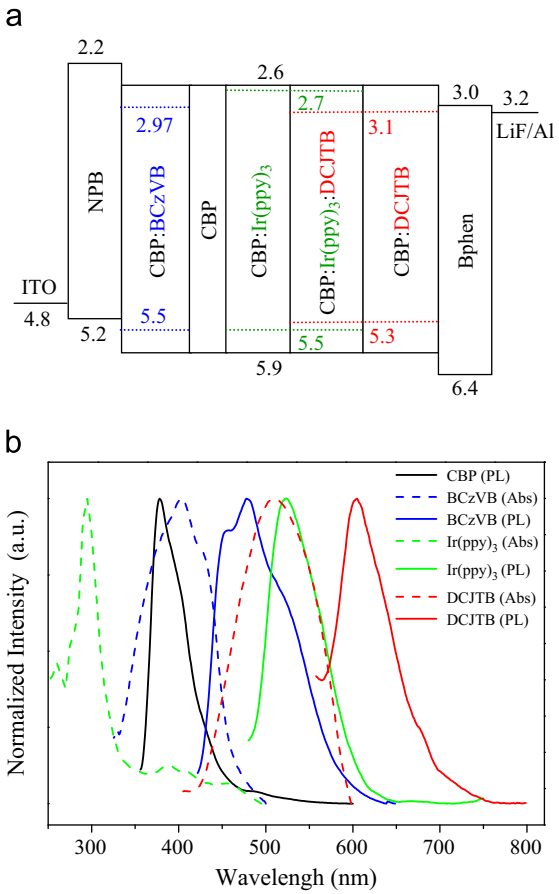

Fig. 1. (a) Energy level diagram of the device, and (b) emission and absorption spectra of CBP, BCzVB, Ir(ppy)$_3$, and DCJTB.

spectral overlaps were observed between the CBP emission and BCzVB absorption and between the Ir(ppy)$_3$ emission and DCJTB absorption. Therefore, effective Förster energy transfer is possible between these molecules [21]. On the other hand, the Förster energy transfer is ineffective between CBP and DCJTB as well as between the CBP and Ir(ppy)$_3$ molecules. However, Dexter energy transfer is possible if molecules are close enough to each other [22]. Our experimental results showed that the highest brightness of a blue OLED was obtained when a 20 nm-thick blue EML (B-EML) prepared from CBP:7 wt% BCzVB was deposited. Adjacent to the B-EML was a green CBP:Ir(ppy)$_3$ EML (G-EML), that can efficiently utilize singlet and triplet excitons to achieve near 100% quantum efficiency [23]. Fig. 2 shows the current density and luminance intensity as a function of the applied voltage for the G-EML doped