



Effect of electron injection and transport materials on efficiency of deep-blue phosphorescent organic light-emitting devices

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ABSTRACT

We investigate the performance of Flr6-based deep-blue phosphorescent organic light-emitting devices (PHOLEDs) with three different electron transport materials, bathocuproine (BCP), 4,7-diphenyl-1,10-phenanthroline (BPhen), and tris[3-(3-pyridyl)mesityl]borane (3TPYMB), and study the effect of doping alkaline metals (Li and Cs) into these charge transport materials. External quantum efficiency (η_{EQE}) of $(20 \pm 1)\%$ and peak power efficiency (η_p) of (36 ± 2) lm/W were achieved maintaining Commission Internationale de L'Eclairage (CIE) coordinates of $(x = 0.16, y = 0.28)$ in p-i-n dual-emissive-layer (D-EML) deep-blue PHOLEDs with 3TPYMB as the electron transport material and 3TPYMB:Cs as the electron injection layer. The high efficiencies are attributed to the high triplet energy of 3TPYMB as well as the increased conductivity of 3TPYMB:Cs.

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1. Introduction

Highly efficient phosphorescent organic light-emitting devices (PHOLEDs) [1–3] have attracted great interests from both academia and industries due to their advantages for solid-state lighting [4] and full-color display [5] applications. Blue light emission is a necessary component for such applications; however, compared to their longer-wavelength counterparts (such as red and green), the performance of blue PHOLEDs has remained a challenge due to the higher exciton energies involved in these devices. A widely studied blue electrophosphorescent dye, irid-

ium(III) bis[(4,6-difluorophenyl)-pyridinato-*N,C*^{2'}]picolinate or Flrpic [6], typically yields greenish-blue emissions with the major vibronic peak at a wavelength of $\lambda = 475$ nm and Commission Internationale de L'Eclairage (CIE) coordinates of approximately (0.17, 0.34) [7]. It has been shown recently that the efficiency of Flrpic-based blue PHOLEDs can be significantly improved by using host and charge transport materials with high triplet energies [8–10], and maximum luminous efficiencies approaching 50 cd/A and power efficiencies exceeding 30 lm/W have been reported [8,10].

Our work has been focused on a deep-blue phosphor, iridium(III) bis(4',6'-difluorophenylpyridinato)tetrakis(1-pyrazolyl)borate or Flr6 [11]. With the lowest vibronic peak at $\lambda = 458$ nm and CIE coordinates of (0.16, 0.26)

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[12,13], FIr6-based PHOLEDs show deeper blue emissions than FIrpic-based devices, and therefore are more advantageous for lighting and display applications. However, the higher triplet energy (T_1) of FIr6 than that of FIrpic (2.72 eV vs. 2.62 eV) [11] makes it even more challenging to efficiently generate and confine excitons on the phosphor molecules. Also note that the deeper blue emission of FIr6-based devices leads to an approximately 20% lower photopic sensitivity by the human eyes compared to that of FIrpic devices.

Recently we have shown that external quantum efficiencies (η_{EQE}) up to 18% can be achieved in FIr6-based PHOLEDs by using the hole transport material of 1,1-bis-(di-4-tolylaminophenyl)cyclohexane (TAPC, $T_1 = 2.87$ eV) [14]. Furthermore, we employed a dual-emissive-layer (D-EML) structure using *p*-bis(triphenylsilyl)benzene (UGH2) and *N,N'*-dicarbazolyl-3,5-benzene (mCP) as the host materials for FIr6 in the two adjacent emissive layers to maximize the exciton formation on FIr6 molecules, and applied the *p-i-n* device structure with highly conductive charge injection/transport layers to lower the device operating voltage. As a result, a high power efficiency of $\eta_p = 25$ lm/W at a luminance of $L = 100$ cd/m² was demonstrated, which was only slightly reduced to $\eta_p = 20$ lm/W at $L = 1000$ cd/m² [15].

Here, we compare performance of FIr6-based D-EML PHOLEDs with three different electron transport materials, bathocuproine (BCP), 4,7-diphenyl-1,10-phenanthroline (BPhen), and tris[3-(3-pyridyl)mesityl]borane (3TPYMB) [16–19], and study the effect of doping alkaline metals (Li and Cs) [20] into these charge transport materials. We show that $\eta_{EQE} = 20\%$ and maximum $\eta_p = 36$ lm/W can be achieved in devices with 3TPYMB as the electron transport material and 3TPYMB:Cs as the electron injection layer. The high efficiencies are attributed to the high triplet energy of 3TPYMB, $T_1 = 2.95$ eV [19], compared to 2.5 eV for BCP and BPhen [16,17], as well as the increased conductivities due to alkaline doping.

2. Experimental

Glass substrates precoated with an indium tin oxide (ITO) anode (sheet resistance $\sim 20 \Omega/\square$) were degreased in detergent and de-ionized water, and cleaned with ultrasonic baths of acetone and isopropanol consecutively for 15 min each. The substrates were then exposed to an ultraviolet-ozone ambient for 15 min immediately before being loaded into a high-vacuum thermal evaporation system (background pressure $\sim 3 \times 10^{-7}$ Torr), where all the organic and metal layers were deposited successively without breaking the vacuum.

The schematic energy level diagram of the D-EML *p-i-n* deep-blue PHOLEDs [15] and the molecular structures of FIr6 and the three electron transport materials are shown in Fig. 1 (energy levels are taken from the literature) [11,13,21,22]. The D-EML structure composed of 4 wt% FIr6 doped mCP (7 nm thick) and 25 wt% FIr6 doped UGH2 (15 nm thick) layers was used to maximize the exciton generation within the broadened charge recombination zone [15]. TAPC serves as the hole transporting/

electron blocking layer (HTL), whereas an undoped layer of BCP, BPhen, or 3TPYMB was used as the electron transporting/hole blocking layer (ETL). The thicknesses of both HTL and ETL were x nm with x varying from 5 to 20. A (40- x) nm thick *N,N'*-diphenyl-*N,N'*-bis(3-methylphenyl)-[1,1'-biphenyl]-4,4'-diamine (MeO-TPD) layer doped with 2 mol% tetrafluoro-tetracyanoquinodimethane (F₄-TCNQ) was used as the *p*-type hole injection layer (HIL) [21], whereas Li or Cs was doped into the ETL material to serve as the *n*-type electron injection layer (EIL), whose thickness was also (40- x) nm. As a comparison, “conventional”, i.e. non-*p-i-n* type, devices were also fabricated, which did not contain the *p*- and *n*-doped charge injection layers. The thicknesses of HTL and ETL in the conventional devices were both 40 nm.

For the alkaline doping of the electron transport materials, lithium metal was used as the Li source, whereas cesium carbonate (Cs₂CO₃), which decomposes during thermal evaporation to generate cesium atoms ($2\text{Cs}_2\text{CO}_3 \rightarrow 4\text{Cs} + \text{O}_2\uparrow + 2\text{CO}_2\uparrow$) [23], was used as the Cs source. The cathode consisting of a 0.5 nm thick layer of LiF followed by a 50 nm thick Al was deposited through an in situ shadow mask, forming active device area of 4 mm². 3TPYMB, mCP, UGH2, and FIr6 were purchased from Luminescence Technology Corp., and used as obtained without further purification.

Luminance (L)–current density (J)–voltage (V) measurements were carried out in ambient using an Agilent 4155C semiconductor parameter analyzer and a calibrated Newport silicon detector. The luminance was calibrated using a Konica Minolta LS-100 luminance meter assuming Lambertian emission pattern. Electroluminescence (EL) spectra were taken using an Ocean Optics HR4000 high-resolution spectrometer. The luminous, power, and external quantum efficiencies (η_L , η_p , and η_{EQE} , respectively) were derived based on the recommended methods [24]. The conductivities of nominally undoped or alkaline doped electron transport materials were obtained from the ohmic regions of the J - V characteristics of 100 nm thick films sandwiched between two Al electrodes.

3. Results and discussions

3.1. Conventional devices with different ETLs

Fig. 2 shows the comparison of η_{EQE} of three conventional devices with different ETLs. The layer thicknesses and ETL materials for these devices are summarized in Table 1. A maximum $\eta_{EQE} = 14\%$ is achieved at a current density of $J < 10^{-2}$ mA/cm² in device A with BCP as the ETL, compared to the maximum of $\eta_{EQE} = 16\%$ in device B with a BPhen ETL, which is obtained at $J \approx 0.1$ mA/cm². In device C which uses 3TPYMB as the ETL, η_{EQE} is substantially higher than in the other two devices, and reaches a maximum of 20%. This is the highest η_{EQE} ever reported for deep-blue PHOLEDs without any particular light extraction enhancement mechanism. As the light extraction efficiency in these planar-type OLEDs is generally believed to be 20% [2,25] (although there have been recent evidences that it could be slightly higher [10]), this suggests that the internal

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