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# Highly efficient inverted top emitting organic light emitting diodes using a horizontally oriented green phosphorescent emitter



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## ARTICLE INFO

## Article history:

Received 5 May 2014

Received in revised form 24 July 2014

Accepted 2 August 2014

Available online 16 August 2014

## Keywords:

Top emission organic light emitting diodes

Transparent conducting oxide top electrode

Horizontal emitter dipoles

Exciplex forming co-host

Optical analysis

## ABSTRACT

We report a highly efficient phosphorescent green inverted top emitting organic light emitting diode doped with Ir(ppy)<sub>2</sub>tmd [bis(2-phenylpyridine)iridium(III)(2,2,6,6-tetramethylheptane-3,5-diketonate)] as the horizontally oriented emitter in an exciplex forming co-host system. The device showed a maximum current efficiency of 120.7 cd/A, a maximum external quantum efficiency (EQE) of 27.6% and the power efficiency of 85.9 lm/W at 1,000 cd/m<sup>2</sup>. Moreover the efficiency roll off was small long-lasting to 20,000 cd/m<sup>2</sup> with EQE's and current efficiencies of 26.0% and 113.7 cd/A at 10,000 cd/m<sup>2</sup> and 24.5% and 107.6 cd/A at 20,000 cd/m<sup>2</sup>, respectively. Optical analysis of the efficiencies and emission spectra of the device is also reported.

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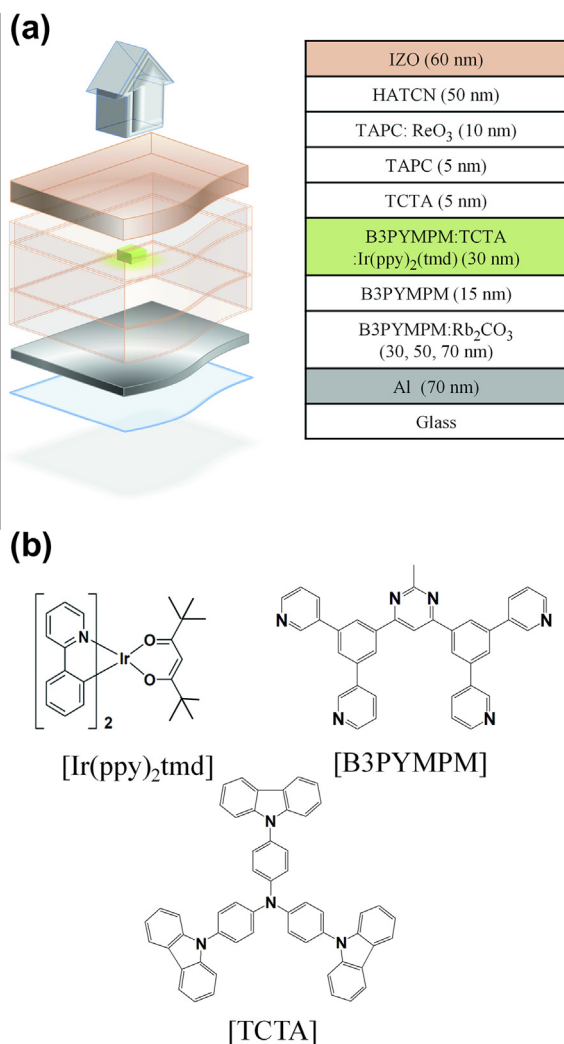
Top emitting organic light emitting diodes (TEOLEDs) have shown interests in solid state lighting and display. Especially, the research of inverted TEOLEDs with a bottom cathode is important for the future applications adopting metal foil substrate for roll to roll process or the display backplane using n-type oxide thin film transistors [1–5]. In recent years, orientation of the transition dipole moments of an emitter is considered as an important factor influencing the quantum efficiency and the out-coupling efficiency in OLEDs. Numerous papers have demonstrated that horizontally oriented emitting dipoles result in higher outcoupling efficiency than vertically oriented dipoles due to the change of the electromagnetic field distribution of the dipole oscillations [6–11]. Not only spin coated polymers but also vacuum evaporated fluorescent and phosphorescent emitters were reported to have preferred horizontal orientation, which enhances the out-

coupling efficiency of OLEDs [12–18]. Recently our group reported high efficiency bottom emission OLEDs with external quantum efficiencies (EQEs) over 30% by doping heteroleptic iridium complexes having high quantum yield and horizontal orientation of transition dipole moments in exciplex co-host systems [19–25].

In this letter, we report a highly efficient inverted TEOLED using an exciplex-forming co-host system doped with the horizontally oriented phosphorescent green dopant bis(2-phenylpyridine)iridium(III)(2,2,6,6-tetramethylheptane-3,5-diketonate) [Ir(ppy)<sub>2</sub>tmd]. The emitter has a preferred orientation of the transition dipole moment in the emitting layer (EML) with the horizontal to vertical transition dipole ratio of 0.78:0.22 and the photoluminescence quantum yield (PLQY) of 96% [20]. The device showed an external quantum efficiency (EQE) of 27.6% at 1,000 cd/m<sup>2</sup> and the current efficiency of 120.7 cd/A. Moreover, the device showed low efficiency roll-off with the EQE value of 24.5% and the luminous efficiency of 107.6 cd/A at 20,000 cd/m<sup>2</sup>.

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**Fig. 1.** Schematic diagram of the inverted TEOLEDs (a) device structures of inverted TEOLEDs. (b) Molecular structures of organic materials in the emitting layer.

Fig. 1a and b shows the device architecture of the inverted TEOLEDs and the chemical structure of the materials used for the devices. A 70 nm thick Al metal cathode and organic layers were successively deposited at a base pressure of  $<5 \times 10^{-7}$  Torr by thermal evaporation without breaking the vacuum. 15 wt% rubidium carbonate ( $\text{Rb}_2\text{CO}_3$ ) doped bis-4,6-(3,5-di-3-pyridylphenyl)-2-methylpyrimidine (B3PYMPM) was used as a n-doped electron injection layer (EIL) and undoped B3PYMPM was used as an electron transport layer (ETL). Co-deposited B3PYMPM and 4,4',4''-tris(N-carbazolyl)-triphenylamine (TCTA) with the 1:1 ratio and doped with 8 wt%  $\text{Ir}(\text{ppy})_2\text{tmd}$  (0.46:0.46:0.08 M ratio) were used as an EML to take advantage of the exciplex forming character of the materials [15–21]. Undoped 1,1-bis-(4-bis(4-methyl-phenyl)-amino-phenyl)-cyclohexane (TAPC) and TAPC doped with 8 wt% rhenium oxide ( $\text{ReO}_3$ ) were used as a hole transport layer and a p-doped hole injection layer (HTL, HIL), respectively. A 50 nm thick HATCN layer was deposited to protect

the underlying organic layers from the plasma damage and have good hole-injection properties from the sputter deposited IZO top electrode [22–26]. The device structure was optimized prior to the fabrication of the inverted TEOLEDs using the classical dipole model [27,28]. The current density–voltage–luminance ( $J$ – $V$ – $L$ ) characteristics of the devices were measured using a Keithley 2400 semiconductor parameter analyzer and a Photo Research PR 650 spectro-photometer. Angular-dependent EL spectra were measured using an optical fiber and an S2000 miniature fiber optic spectrometer (Ocean Optics).

Fig. 2a exhibits a simulated contour plot of the relative maximum achievable luminous intensity in the normal direction as functions of the thickness of EIL and HIL. The simulation results indicate that the highest current efficiency can be obtained with the EIL and HIL thicknesses of 30 nm and 10 nm, respectively with the fixed thicknesses of B3PYMPM (15 nm), TCTA (5 nm), and TAPC (5 nm), respectively. Fig. 2b shows the current density–voltage–luminance ( $J$ – $V$ – $L$ ) characteristics of the inverted TEOLEDs with the 30 nm, 50 nm, and 70 nm thick EILs, respectively. The device with the 30 nm thick EIL showed the highest luminance, which is consistent with the simulation result. The turn-on voltage of the optimized device was 2.8 V at 22  $\text{cd}/\text{m}^2$  and the driving voltages were 4.0 V and 5.8 V at 1,000  $\text{cd}/\text{m}^2$  and 10,000  $\text{cd}/\text{m}^2$ , respectively. The electroluminescence spectra in the normal direction and angle dependent intensity distribution of the devices matched very well with the simulation results as shown in Fig. 2c and d, respectively. Some discrepancy between them might come from the shift of emission zone in the devices depending on the thickness of the ETL and/or optical birefringence of organic materials which has not been considered in our optical modeling.

Fig. 3a represents the current and power efficiency of the inverted TEOLEDs. The optimized device with 30 nm thick EIL exhibited the current and power efficiencies of 120.7  $\text{cd}/\text{A}$  and 85.9  $\text{lm}/\text{W}$  at 1,000  $\text{cd}/\text{m}^2$  and 113.7  $\text{cd}/\text{A}$ , 56  $\text{lm}/\text{W}$  at 10,000  $\text{cd}/\text{m}^2$ . The device also showed high EQEs of 27.6% at 1,000  $\text{cd}/\text{m}^2$  and 26.0% at 10,000  $\text{cd}/\text{m}^2$  (Fig. 3b). Moreover, the efficiency roll off is small with the current efficiency of 107.6  $\text{cd}/\text{A}$ , the power efficiency of 46.4  $\text{lm}/\text{W}$ , and the EQE of 24.6% at 20,000  $\text{cd}/\text{m}^2$ . The power efficiency and external quantum efficiency of the devices were calibrated using the viewing angle-dependent electroluminescent (EL) intensity and spectra. Log scale plots of the efficiencies can be found in Supplementary Information (Fig. S1) to get more information on the region of low luminance. Performances of the devices are summarized in Table 1.

Optical simulation of the EQE of the devices was performed using the experimentally obtained values of  $\text{PLQY} = 0.96$  and  $\theta = 0.78$ . The PL quantum yield was measured using an integrating sphere [29,30] and the sample consisted of a 50 nm thick emitting layer (TCTA: B3PYMPM:Ir(ppy)<sub>2</sub>(tmd), 1:1 M ratio and 8 wt%) on a quartz substrate. Fig. 4 represents the maximum achievable EQEs from the optically optimized device structure as a function of  $\text{PLQY}$  and  $\theta$  of the emitter under the assumption of negligible electrical loss ( $J$ ). The simulation predicted that EQE of 31.9% is achievable if there is no elec-

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