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Suppression of efficiency roll-off in highly efficient blue phosphorescent organic light-emitting devices using novel iridium phosphors with good electron mobility



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1. Introduction

In the past few years, white organic light-emitting diodes (WOLEDs) have drawn increasing attention because of their potential applications in full colour flat-panel displays and solid-state lighting [1-4]. Generally, there are two main methods to achieve white light emission including the mixture of red, green and blue three prime colors or blue and orange-yellow complementary colors in a device's configuration [5-9]. Obviously, the blue emitter is indispensable and its device performance is considerably significant to white OLEDs. Recently, fluorescent blue emitters combining with phosphorescent red and green emitters have been developed for white OLEDs [10-17]. However, due to the fact that only the singlet exciton of fluorescent blue emitter can contribute to the light emission, devices comprising fluorescent blue emitter may not have sufficient efficiency. For this reason, adopting blue

ABSTRACT

High-efficiency blue organic light-emitting diodes were reported by adopting two novel iridium phosphors. Due to phosphoryl moiety in ancillary ligands, both complexes $(dfppy)_2Ir(ppp)$ and $(dfppy)_2Ir(dpp)$ (dyppy = 2-(2,4-difluorophenyl)pyridine, ppp = phenyl(pyridin-2-yl)phosphinate, dpp = dipyridinylphosphinate) own high electron mobility which can balance the injection and transport of carriers. Furthermore, the double light-emitting layers with TcTa <math>(4,4',4''-tris(carbazol-9-yl)triphe-nylamine) and 26DCzPPy (2,6-bis(3-(carbazol-9-yl)phenyl)pyridine) hosts broaden the exciton formation zone and suppress efficiency roll-off. The optimized double light-emitting layers devices exhibited decent performances with peak current efficiency near 50 cd/A and external quantum efficiency above 20% as well as negligible efficiency roll-off.

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phosphorescent emitter becomes essential to achieve highly efficient white OLEDs because it can both utilize the singlet and triplet excitons for generating light emission. Though researchers have paid great attentions for developing stable and highly efficient blue iridium phosphors and designing appropriate device structures, blue phosphorescent OLEDs are still confronted with some problems such as low efficiency and rapid efficiency roll-off compared with other colors-emitting.

Furthermore, it is well known that charge balance is a very important factor in the operation of OLEDs, which has a significant impact on peak efficiency as well as efficiency roll-off. And in most OLEDs, because the hole mobility of the hole transport layer is higher than the electron mobility of the electron transport layer [18,19], the performances of OLEDs rely on the capability of electron transport. Therefore, the phosphorescent emitters owning good electron mobility would facilitate the injection and transport of electrons, which will balance the distribution of hole – electron, broaden the recombination zone and confine excitons well, leading to the suppressed excitons annihilation, as well as high device efficiency and low efficiency roll-off. Many groups have tried to introduce electron transport moieties (nitrogen-containing heterocycles, phosphoryl P=O group, etc) into Ir(III) complexes to



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improve their electron mobility. In our previous work, we also used tpip (tetraphenylimido-diphosphinate) derivatives with two P=O moieties as ancillary ligand for efficient iridium complexes and devices [20–22].

In this context, we proposed two novel blue phosphorescent iridium complexes (dfppy)₂Ir(ppp) and (dfppy)₂Ir(dpp) (dyppy = 2-(2,4-difluorophenyl)pyridine, ppp = phenyl(pyridin-2-yl)phosphinate, dpp = dipyridinylphosphinate) as dopants for blue phosphorescent OLEDs (Electronic Supporting Information). The phosphoryl P=O group in the ancillary ligand was used to improve the electron mobility of the complexes which will benefit the electroluminescence (EL) performances.

2. Experimental

All the organic materials used in this study were obtained commercially except for (dfppy)₂Ir(ppp) and (dfppy)₂Ir(dpp) which were synthesized and purified in our laboratory. Indium-tin-oxide (ITO) coated glass with a sheet resistance of 10 Ω /sq was used as the anode substrate. All the organic layers were deposited with the rate of 0.1 nm/s under high vacuum ($\leq 2.0 \times 10^{-5}$ Pa). The EMLs were prepared by co-evaporating dopant(s) and host material from two individual sources, and the doping concentrations were modulated by controlling the evaporation rates of dopant(s). MoO₃, LiF and Al were deposited in another vacuum chamber (${\leq}8.0\times10^{-5}\,\text{Pa})$ with the rate of 0.01, 0.01 and 1 nm/s, respectively. The EL characteristics were measured by using a programmable Keithley source measurement unit with a silicon photodiode. The EL spectra were measured with a calibrated Hitachi F-7000 fluorescence spectrophotometer. The EQE was calculated based on the photo energy measured by the photodiode, the EL spectrum, and the current pass through the device.

3. Results and discussion

To measure the electron mobility of the complexes, transient electroluminescence (TEL) experiments based on the devices of ITO/TAPC (di-[4-(*N*,*N*-ditolyl-amino)-phenyl]cyclohexane, 50 nm)/ (dfppy)₂Ir(ppp) (Device A), (dfppy)₂Ir(dpp) (Device B), FIrpic (Device C) (60 nm)/LiF (1 nm)/Al (100 nm) were conducted. Because FIrpic (Ir(III)bis(4,6-(difluorophenyl)pyridinato-*N*,*C*')picolinate) is the most widely used blue emitter due to its good device performances and has similar molecular structure with (dfppy)₂Ir(ppp)

and (dfppy)₂Ir(dpp) [23,24], it was chosen as the reference in this study. Fig. 1 shows the energy level diagram of HOMO and LUMO levels (relative to vacuum level) for materials investigated in this work and their molecular structures. Here, TAPC functions as hole-transport layer, while Ir(III) compounds function as both emissive and electron-transport layer.

In transient EL measurements the time-dependent EL response is monitored upon exciting the OLEDs with a rectangular voltage pulse. Fig. 2 lists the transient EL signals for Device A/B/C and electric field dependence of charge electron mobility of complexes in the thin films. A finite delay time (t_d) , which is between the application of a voltage pulse and the onset of EL signal, is determined by the arrival of the slower charge carrier of the injected carriers at the emission zone [25-28]. Therefore, the slower charge carrier mobility is directly correlated with delay time. The most important advantage of the transient EL method is that the short pulse voltage is hard to degrade the performance of OLEDs, which is considerably important for the reproducibility of the experiments. Because the hole mobility of TAPC (~ $1.0 \times 10^{-2} \text{ cm}^2/\text{V} \cdot \text{s})$ [29–31] is much higher than the electron mobility of the iridium complexes, the voltage drop across the TAPC layer and the built-in electric field between organic/organic interface is negligible [32]. Bearing this in mind, the electron mobility can be roughly calculated with the equation of $\mu_e = d^2/(t_d \cdot V)$, where *d* is the thickness of the emitting layer, V is the driving voltage.

In the figure the t_d is the time from zero to the onset of EL signal and it also can be observed that these values via the driving voltages increase greatly. The calculated electron mobility data of (dfppy)₂Ir(ppp), (dfppy)₂Ir(dpp) and FIrpic are between $6.77-6.92 \times 10^{-6}$ (t_d : 0.52-0.66 µs), 7.50-7.84 × 10^{-6} (t_d : 0.48–0.58 µs) and 4.60–5.14 \times 10⁻⁶ (t_d : 0.70–0.98 µs) cm²/V·s, respectively, under the driving voltage from 8 to 10 V and an electric field range from 1150 to 1300 $(V/cm)^{1/2}$. Both (dfppy)₂Ir(dpp) and (dfppy)₂Ir(ppp) possess the higher electron mobility values than that of widely used electron transport (aluminum(III)tris(8-hydroxyguinolinato, material Alq_3 $4.74-4.86 \times 10^{-6} \text{ cm}^2/\text{V} \cdot \text{s}$) and Firpic. Furthermore, the data of (dfppy)₂Ir(dpp) are a little better than that of (dfppy)₂Ir(ppp). The good electron transport ability of (dfppy)₂Ir(ppp) and (dfppy)₂Ir(dpp) would facilitate the injection and transport of electrons, which will broaden the recombination zone, balance the distribution of hole-electron and reduce leakage current. Consequently, the results would lead to the suppressed triplet-triplet



Fig. 1. The energy level diagrams of the devices and molecular structures of materials investigated in this work.

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