



Highly efficient green phosphorescent organic electroluminescent devices with a terbium complex as the sensitizer



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ABSTRACT

An effective device design strategy was reported to realize high performance green organic light-emitting diodes based on iridium complex by employing a trivalent terbium complex as the sensitizer. The low-lying energy levels of utilized terbium complex are beneficial in broadening recombination zone and facilitating carriers' balance on emitter molecules. No terbium ion characteristic emission was observed in co-doped devices ascribed to high triplet energy of the ligand of terbium complex. Consequently, the 0.4 wt% co-doped single light-emitting layer device obtained the maximum external quantum efficiency, current efficiency and power efficiency up to 27.6%, 114.23 cd/A and 101.92 lm/W, respectively. At 3.8 V, this device realized the practical brightness of 1000 cd/m² with inspiring external quantum efficiency and current efficiency as high as 27.5% and 113.71 cd/A, respectively. While the 0.2 wt% co-doped double light-emitting layers device displayed relatively lower efficiency, but higher brightness up to 205,629 cd/m².

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1. Introduction

Organic light-emitting diodes (OLEDs) have aroused extensive research interest due to their promising features, including flexibility, color tunability and low power consumption [1–3]. To achieve high efficiency, which is essential for practical display and lighting applications, iridium(III) complexes with short excitation lifetime are widely utilized in OLEDs owing to their advantage of achieving high efficiency [4–6]. Over the past decades, multifarious device design strategies, such as low interfacial resistance p-i-n device structure, low carriers injection barrier, stepwise multiple light-emitting layers (EML), carriers/exciton confinement layer, and host/guest system with effective energy transfer, have been reported to enhance the electroluminescent (EL) efficiency [7–10]. Although impressive EL efficiency has been reported constantly, EL

efficiency of these phosphorescent OLEDs drops rapidly with increasing current density, well known as notorious efficiency roll-off [11–15]. Therefore, it is crucial and urgent to develop highly efficient OLEDs with slow efficiency roll-off especially at high brightness.

According to previous investigations, the origin of efficiency roll-off can be ascribed to unbalanced carriers' distribution, triplet-triplet annihilation (TTA), exciton-polaron interaction and field-induced quenching [16,17]. Although several material design strategies such as decreasing exciton lifetime, reducing molecular aggregation and Förster radius have been previously demonstrated to be effective in suppressing the efficiency roll-off in some degree [18–23], EL performances of most obtained devices at relatively high brightness were still not adequate for commercialization. Recently, some groups have devoted to decrease the undesired TTA by optimizing the design of device structure [24–26]. For example, our group have demonstrated the feasibility of co-doping iridium(III) (Ir(III)) complex with low-lying highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels into electron dominant EML as sensitizer to achieve high performance OLEDs based on red-emitting platinum(II) (Pt(II))

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complex. Research results implied sensitizer molecules function as electron trappers and energy transfer ladders, which help to broaden recombination zone, balance carriers' distribution and facilitate the transfer of energy from host to emitter molecules, thus delaying EL efficiency roll-off. However, weak blue emission originating from the co-doped sensitizer was also observed, which unavoidably deteriorated the color purity of red OLED.

On the way of trying efforts to enhance EL efficiency and delay efficiency roll-off of OLEDs based on red-emitting Ir(III) complex iridium(III) bis(2-phenylquinoly- N,C^2')dipivaloylmethane ((PQ)₂Ir(dpm)) without sacrificing color purity, the widely used red-emitting trivalent europium complex Eu(TTA)₃phen (TTA = thenoyltrifluoroacetone, phen = 1,10-phenanthroline) was selected as sensitizer. Research results implied that emitters are considered to be electron trappers and energy transfer ladders due to its deep energy levels and well matched triplet energy (E_T). Furthermore, the relatively long excitation lifetime of Eu(III) ion is helpful in transmitting exciton energy from ligand TTA to not Eu(III) ion but emitter molecules. Although very weak Eu(III) ion emission can still be observed at relatively low current density, the pure red emission characteristic (peaked at 612 nm) of Eu(III) ion helps to ensure the color purity of co-doped red OLEDs. Finally, bright red EL device with maximum current efficiency (CE), power efficiency (PE) and brightness up to 58.98 cd/A (external quantum efficiency (EQE) of 21.0%), 61.73 lm/W and 100,870 cd/m², respectively, was achieved by optimizing the concentrations of Eu(TTA)₃phen and PQ₂Ir(dpm) in sequence. In contrast with red emitters, green and blue emitters are always accompanied with relatively wider energy gaps and higher E_T , which make it more difficult to select appropriated sensitizers in realizing pure green and blue PHOLEDs with high EL performances.

In this investigation, to achieve excellent EL performances of OLEDs based on Ir(tfmppy)₂(tpip) (tfmppy = 4-trifluoromethylphenylpyridine, tpip = tetraphenylimido-diphosphate), a well-known green-emitting trivalent terbium complex Tb(acac)₃(phen-Cl) (acac = acetylacetonate, phen-Cl = 3-chlorophenanthroline) was selected as sensitizer and co-doping into electron dominant EML due to its low-lying energy levels and high E_T of ligand acac. Compared with reference device, Tb(acac)₃(phen-Cl) co-doped devices exhibited higher brightness, higher efficiencies, slower efficiency roll-off, and even higher color purity attributed to the improved carriers' balance and wider recombination zone. In addition, no Tb(III) ion emission was observed in all co-doped devices, which was attributed to the higher E_T of ligand acac and the lower HOMO level of Tb(acac)₃(phen-Cl). Finally, the optimized co-doped single-EML device obtained the maximum CE and PE up to 114.23 cd/A (EQE = 27.6%) and 101.92 lm/W, respectively, while the optimized co-doped double-EMLs device obtained the maximum brightness up to 205,629 cd/m². Even at certain brightness of 1000 cd/m² (3.8 V), inspiring EQE and CE as high as 27.5% and 113.71 cd/A, respectively, can still be retained by the co-doped single-EML device.

2. Experimental

All the organic materials used in this study except for Ir(tfmppy)₂(tpip), which was synthesized and purified in our laboratory, were obtained commercially and used as received without further purification. Indium-tin-oxide (ITO) coated glass with sheet resistance of 10 Ω/sq was used as the anode substrate. Prior to film deposition, patterned ITO substrates were cleaned with detergent, rinsed in de-ionized water and finally dried in an oven. All the organic layers were deposited with the rate of 0.1 nm/s under high vacuum ($\leq 3.0 \times 10^{-5}$ Pa). The doped and co-doped EMLs were prepared by co-evaporating dopant(s) and host material from two

or three individual sources, and the doping concentrations were modulated by controlling the evaporation rate of dopant(s). MoO₃, LiF and Al were deposited in another vacuum chamber ($\leq 8.0 \times 10^{-5}$ Pa) with the rates of 0.01, 0.01 and 1.0 nm/s, respectively, without being exposed to the atmosphere. The thicknesses of these deposited layers and the evaporation rate of individual materials were monitored in vacuum with quartz crystal monitors. A shadow mask was used to define the cathode and to make ten emitting dots with the active area of 9 mm² on each substrate. Current density-brightness-voltage (J - B - V) characteristics were measured by using a programmable Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a silicon photodiode. The EL spectra were measured with a calibrated Hitachi F-7000 fluorescence spectrophotometer. The EQE of EL device 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

Fig. 1 shows the device architecture used in this work and the energy levels of the used materials. In this device architecture, MoO₃ acts as hole injection layer, di-[4-(N,N -ditolyl-amino)-phenyl]cyclohexane (TAPC) and 1,3,5-tri(m -pyrid-3-yl-phenyl)benzene (TmPyPB) function as hole and electron transporting layers, respectively. Meanwhile, TAPC and TmPyPB serve also as electron and hole blocking layers, respectively, to confine charge carriers and excitons in the EML [27,28]. 4,4',4''-tri(9-carbazoyl)triphenylamine (TcTa) and 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy) serve as host materials, due to their matched E_T and energy gaps [29,30]. The highly efficient green-emitting Ir(III) complex Ir(tfmppy)₂(tpip) was selected as emitter, while the typical terbium complex Tb(acac)₃(phen-Cl) was co-doped into EML2 as sensitizer because of the high E_T of ligand acac and the low-lying energy levels [31,32]. The molecular structures of Ir(tfmppy)₂(tpip) and Tb(acac)₃(phen-Cl) were also depicted in Fig. 1. In this case, the HOMO and LUMO levels (−5.44 and −2.98 eV) of Ir(tfmppy)₂(tpip) are well matched with TcTa and 26DCzPPy [5,29,30], thus direct exciton formation on emitter molecules following carrier trapping is expected. Within EMLs, electrons would be trapped preferentially by Tb(acac)₃(phen-Cl) molecules due to its lower LUMO level of −3.23 eV. Considering the even lower HOMO level −6.31 eV of Tb(acac)₃(phen-Cl), transfer of holes onto Tb(acac)₃(phen-Cl) molecules would be negligible [5,30,32].

To determine the optimal doping concentration of Ir(tfmppy)₂(tpip), devices following the configuration of ITO/MoO₃ (3 nm)/TAPC (50 nm)/Ir(tfmppy)₂(tpip) (x wt%):26DCzPPy (10 nm)/TmPyPB (50 nm)/LiF (1 nm)/Al (100 nm) were firstly manufactured and examined. The details of the performance were summarized in Table 1, the 6 wt% doped device (defined as single-EML reference device (S-RD)) gave the maximum brightness, CE, PE and EQE up to 55,829 cd/m², 92.50 cd/A, 87.12 lm/W and 22.3%, respectively. At the operation voltage of 3.8 V, this device realized the certain brightness of 1000 cd/m² with CE of 89.57 cd/A (EQE = 21.6%). For comparison, double-EMLs devices were also manufactured and examined by inserting Ir(tfmppy)₂(tpip) (y wt%) doped TcTa film between HTL and bipolar EML as p-type EML. As depicted in Table 2, the maximum brightness, CE, PE and EQE up to 134,743 cd/m², 99.81 cd/A, 85.76 lm/W and 24.1%, respectively, were realized by the 6 wt% doped device (defined as double-EMLs reference device (D-RD)). At the operation voltage of 4.6 V, this device exhibited the practical brightness of 1000 cd/m² with CE of 97.91 cd/A (EQE = 23.7%).

To further enhance the EL performances of these devices, four other co-doped single-EML devices: ITO/MoO₃ (3 nm)/TAPC

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