



Tunable chromaticity stability in solution-processed organic light emitting devices

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ABSTRACT

Efficient solution-processed color-stable and color-tunable white organic light emitting diodes (OLEDs) have been realized by judicious selection of the host materials for the emission layers. The color-tunable OLED demonstrates the unique characteristic of modulating the electroluminescence by using the applied voltage of the device and displays color temperatures ranging from 1600 K to 4600 K around the daylight locus, with a peak external quantum efficiency of 13.6% and a peak current efficiency of 22.5 cd A⁻¹. On the other hand, the chromaticity-stable device shows a negligible color change, from 300 to 2000 nits. The manipulation of chromaticity is attributed to the energy transfer dynamics of the hosts and dopants under different electric fields.

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1. Introduction

The progression of artificial lighting began with incandescent bulbs and fluorescent tubes, followed by the development of the modern light emitting diode (LED) and the organic light emitting diode (OLED). Artificial lighting has evolved from point light sources (e.g., incandescent bulbs) to line sources (e.g., fluorescent tubes) and, ultimately, soft two-dimensional surface illumination (e.g., OLEDs). The aforementioned unique characteristics, together with the high color rendering index (CRI) and continuous tunability in intensity of OLED technology, result in its recognition as the most promising technique for next-generation solid-

state light, especially for indoor applications [1]. Normally, stable chromaticity is required for common lighting applications [2–4], yet in some special applications, an operating voltage tunable chromaticity is required. For example, a sunlight-simulated light source can be used for growing plants indoors. For daily lighting, people can freely choose the most appropriate color temperature while doing various activities [5–8]. The desired chromaticity function was previously achieved by complicated vacuum-deposited multilayer stacking. For instance, owing to the shifting of the recombination zone at various operation voltages, a blue/red/blue [5] or red/blue/red [6] sandwiched structure was utilized to achieve chromaticity-stable devices. In chromaticity-tunable devices, deliberate manipulation of the recombination zone in different emission layers under different applied voltages is usually required. These approaches have been proven to effectively give OLEDs their required functionality; however, because of the complex emission layer (EML) structures, increasing fabrication complexity and cost are inevitable. Compared to

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vacuum deposition OLEDs, solution-processed OLEDs possess several significant advantages, including high material utilization, easy fabrication of multi-dopants in a single emission layer, low fabrication cost, and adequate compatibility with the roll-to-roll processing technologies [9,10]. Therefore, ever-increasing research endeavors have been dedicated to developing solution-processed OLEDs over the past few years [11–18].

In this report, we demonstrate a simple but effective way to fabricate efficient solution-processed small-molecule phosphorescent white OLEDs with controllable chromaticity. A simple hole transporting layer (HTL)-single EML-electron transporting layer (ETL) architecture was applied in which the solution-processed EML consisted of blue emissive dopant iridium(III), bis(4',6'-difluorophenylpyridinato)picolinate (Flrpic) and red emissive dopant, Os(btfp)₂(pp2b), blended into the hosts. The required operation voltage dependent chromaticity characteristics (color-stable or color-tunable) were easily manipulated by judicious selection of host materials with different energy levels. The photophysical mechanism and excitonic recombination pathways were investigated using time-resolved electroluminescence (TR-EL). The color-stable white OLED with 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy) as the host, which has a lower-lying highest occupied molecular orbital (HOMO) and lower unoccupied molecular orbital (LUMO), exhibited a very stable EL chromaticity between 300 and 2000 nits. Nevertheless, the highly chromaticity-tunable white OLED (W-OLED) yielded EL with a correlated color temperature (CCT) capable of shifting from 1600 to 4600 K, as demonstrated by utilizing 4,4',4''-tris(N-carbazolyl)-triphenylamine (TCTA) with higher-lying HOMO and LUMO levels as the EML host. An additional yellow emissive dopant Os(bptz)₂(dppb) was applied to the chromaticity-tunable W-OLED to make the emission closer to the daylight locus across the entire tunable range. A promising external quantum efficiency (EQE) of up to 13.6% and power efficacy of 14.5 lm W⁻¹ were achieved. The simple approach reported here provides a guideline for fabrication of low-cost solution-processed W-OLEDs with the chromaticity characteristics desired in a variety of applications.

2. Experimental

2.1. Materials

The yellow-emitting Os(II) complex Os(bptz)₂(dppb), showing an emission peak maximum at $\lambda = 604$ nm, phosphorescence lifetime $\tau_{\text{obs}} = 1.8$ μs and emission quantum yield $\Phi = 0.42$ in degassed CH₂Cl₂ solution, was synthesized from treatment of Os₃(CO)₁₂ with bptzH and dppb in sequence, using the methodology described in literature [19,20].

Selected spectral data of Os(bptz)₂(dppb): MS (FAB, ¹⁹²Os): m/z 1040 (M⁺). ¹H NMR (400 MHz, d₆-acetone, 298 K): δ 8.07–8.03 (m, 4H), 7.77–7.73 (m, 2H), 7.53–7.44 (m, 6H), 7.31 (t, $J_{\text{HH}} = 7.6$ Hz, 2H), 7.19 (t, $J_{\text{HH}} = 7.6$ Hz, 4H), 7.05 (t, $J_{\text{HH}} = 7.6$ Hz, 2H), 7.01 (d, $J_{\text{HH}} = 5.6$ Hz, 2H), 6.82 (d, $J_{\text{HH}} = 8.0$ Hz, 4H), 6.69–6.64 (m, 6H), 1.41 (s,

18H). ³¹P NMR (202 MHz, d₆-acetone, 298 K): δ 29.47 (s, 2P). Anal. Calcd. for C₅₂H₅₀N₈OsP₂: N, 10.78; C, 60.10; H, 4.85. Found: N, 10.69; C, 59.64; H, 5.35.

2.2. Device fabrication

The schematic diagram of the device structure and the chemical structures of the compounds used in this study are shown in Fig. 1. Indium tin oxide (ITO)-coated glass substrates were cleaned with detergent and bathed in the de-ionized water, acetone, and methanol for 15 min, each. The cleaned substrates were treated with UV-ozone for 30 min, and then poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) was spin-coated onto the substrates (~40 nm, measured by ellipsometer). The PEDOT:PSS layer was baked at a temperature of 135 °C on the hot plate for 30 min. A TCTA solution in chlorobenzene (10 mg/cc) was spin-coated onto the PEDOT:PSS layer and formed a 40-nm neat film [21]. The TCTA neat film was baked at 100 °C for 30 min to remove the residual solvent in the TCTA layer. The 20-nm-thick EML, which was applying TCTA or 26DCzPPy as a host and doped with phosphorescent dopants, was spin-coated (10 mg/cc in toluene) on the top of the TCTA layer. Blue emissive iridium(III) bis[(4,6-difluorophenyl)-pyridinato-N,C^{2'}]picolinate (Flrpic), yellow emissive Os(bptz)₂(dppb), and red emissive Os(btfp)₂(pp2b) dopants were utilized. The fabrication of spin-coated EML unavoidably eroded the TCTA layer, and the residual thickness of TCTA was found to be ~20 nm, as measured by ellipsometry. The samples were then baked on the hot plate at 80 °C for 30 min before being transferred from the glove box to the thermal deposition chamber. All the transfer processes were carried out in an inert atmosphere without exposure to air. The 2,2',2''-(1,3,5-phenylene)tris(1-phenyl-1H-benzimidazole) (TPBi)

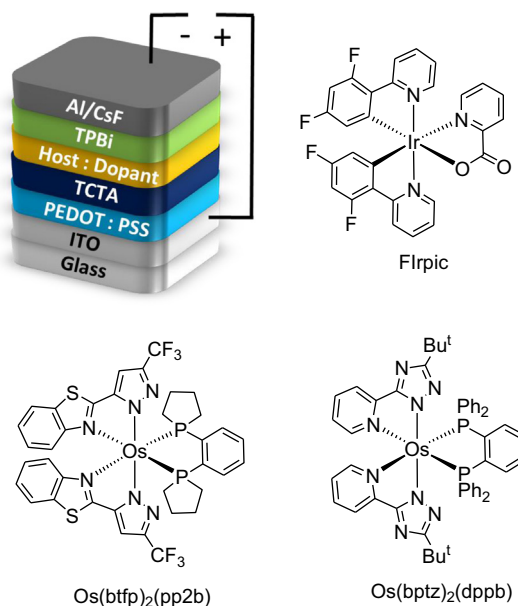


Fig. 1. Structural drawings of OLED devices and materials used in this work.

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