



Study of shifting of recombination zone in multi-emissive layer organic light emitting devices and its effect on color stability

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ABSTRACT

Color stability in multi-emissive layer organic light emitting devices (Me-OLEDs) has been investigated. Me-OLEDs were based on multiple emitters with a common host CBP doped with three dopants, BCzVBi, Ir(ppy)₃ and Ir(btp)₂acac for blue, green and red emission. A major variation in CIE coordinates were found from (0.312, 0.294) to (0.236, 0.267) with increase in voltage from 6 to 9 V. This coordinate shift in Me-OLEDs has been attributed to the shifting of recombination zone with increase in voltage. In order to support our experimental observation, the EL spectrum of Me-OLEDs has been expressed as a linear combination of EL intensities of OLEDs with its individual constituting layers as emitters. In this way, the contribution of each layer in the EL spectrum of Me-OLEDs has been evaluated at each voltage. It has been observed from these calculations that the contribution of red emitter decreases from 47% to 33.33% and blue emitter increases from 38% to 51.67% with the increase in voltage from 6 to 9 V. This supports our hypothesis of shifting of recombination zone with the change in voltage. This shifting has been attributed to the field dependency of electron mobility in CBP. Me-OLED with CBP layers between the emitters was fabricated to improve the color stability. Significant improvement in color stability was achieved without changes in current efficiency in Me-OLED with interlayers.

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1. Introduction

Electroluminescence (EL) devices based on organic materials are widely used in display applications. The structure of these devices is simple having organic thin layers sandwiched between two electrodes. Charge carriers are injected from electrode into emissive layer. These injected charge carriers form a bound state known as excitons and their radiative relaxation leads to the generation of light [1]. Different device structures have been proposed to get the optimum exciton formation [2–9] and thereby producing high efficiency.

Multiple emissive layer structure is one of the prominent structures to get the high efficiencies in organic light emitting devices (OLEDs) [10]. High luminescence can be obtained at lower values of current in multi-emissive layer OLEDs (Me-OLEDs) as compared to single emissive layer OLEDs [11,12]. White light is usually generated by using multi-emissive layer architecture with red, green and blue emissive layers [10,13–15]. Generally, recombination in each layer is controlled by introducing charge blocking

layers in between the emissive layers [16], varying the layer thickness [17] or by adjusting dopant concentrations [18]. White OLEDs (WOLEDs) of desired color purity are obtained by balancing the emission from red, green and blue emissive layers. Such device structures were proposed by D'Andrade and Forrest [10] in which iridium(III) bis(4,6-di-fluorophenyl)-pyridinato-N,C^{2'} (Flrpic) was used for blue, bis(2-phenyl benzothiazolato-N,C^{2'}) iridium(III) (acetylacetonate) (Bt₂Ir(acac)) for yellow and iridium(III) bis[2-(2'-benzothienyl)-pyridinato-N, C^{3'}] (Ir(btp)₂acac) for red emission and 4,4'-bis(carbazol-9-yl)1,1'-biphenyl (CBP) as host material. They have reported a WOLED with high color rendering index (CRI) > 80, with the Commission Internationale de l'Eclairage (CIE) coordinates of (0.37,0.40) and power efficiency of 6.0 lm/W. However the operating voltages for these devices were quite high due to the high combined thickness of emissive layers. This method is widely used for vacuum evaporated small molecular devices since the thickness of layer can easily be controlled [10,13–15].

Although this method is quite efficient, it contains some serious issues such as high operating voltage and variation in EL spectrum with applied voltage. Problem of high operating voltage can be solved by employing p-i-n type structure using p-type doping by strong acceptor like 2,3,5,6-tetrafluoro-7,7',8,8'-tetra-cyanoquinodimethane (F₄-TCNQ) [19,20], molybdenum oxide (MoO₃) [21], and n-type doping by strong donor like lithium (Li) metal [22–25]. These doped layers are helpful in reduction of

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voltage drop across the conducting but non-emissive transport layers allowing total voltage drop to occur across the emissive layers. The other problem associated with the color stability is quite appreciable in Me-OLEDs [15,26,27]. In Lee et al. [15] work the CIE coordinates of device vary from (0.28,0.34) to (0.41,0.39) with the variation of current densities from 0.255 to 151 mA/cm². They used Flrpic as blue, Bt₂Ir(acac) as orange and bis(1-phenylisoquinoline) (acetylacetonate) iridium(III) (Ir(piq)₂(acac)) as red dopant with the common host 1,3-bis(9-carbazolyl) benzene (mcp). They have also studied the color stability of WOLEDs. Previously many authors have worked on the shifting of recombination zone and color stability in WOLEDs [15,26,27]. This variation in EL spectrum or CIE coordinates is due to the shifting of recombination zone with changing voltages. Since WOLEDs have a wide applicability in display devices, color stability becomes more important. Study of shifting of recombination zone in case of WOLEDs is a crucial issue. In present work, we have addressed the problem of color stability by studying the shifting of recombination zone in Me-OLEDs with voltage by using an analytical fitting of experimental results.

2. Experimental

Indium tin oxide (ITO) coated glass substrates having a sheet resistance of 20 Ω/□ were used for the device fabrication and were purchased from Vinkarola. Substrates were patterned and cleaned subsequently in deionized water, acetone, trichloroethylene and propanol for 20 min each using an ultrasonic bath. Cleaned substrates were dried in vacuum oven at 120 °C for 30 min and then loaded into vacuum chamber. Organic layers were deposited onto substrate under a pressure of 4 × 10⁻⁶ Torr at a rate of 0.4 Å/s. Metal electrode (aluminum) was deposited at a rate of 5 Å/s. For the deposition of doped layers the rate for the host and the guest were monitored in situ by using quartz crystal monitor and controlled by using a shutter. The size of each pixel was 5 mm × 5 mm. Thickness of films were measured by quartz crystal monitor and confirmed by using ellipsometer. EL spectrum was measured with a high resolution spectrometer (Ocean optics HR-200 CG UV-NIR). The current density–voltage–luminescence (J–V–L) characteristics were measured with a luminescence meter (LMT-1009) interfaced with a Keithley 2400 programmable current voltage digital source meter. All measurements were carried out at room temperature under ambient conditions.

3. Result and discussion

We have fabricated Me-OLEDs with varying concentration of Ir(btp)₂acac to study the shifting of recombination zone. The device configurations were:

Device 1: ITO (120 nm)/ 0.4 wt% F₄-TCNQ doped α-NPD (35 nm)/5 wt% BCzVBi doped CBP (20 nm)/5 wt% Ir(ppy)₃ doped CBP (4 nm)/0.5 wt% Ir(btp)₂acac doped CBP (12.5 nm)/BALq (30 nm)/LiF (1 nm)/Al (150 nm).

Device 2: ITO (120 nm)/0.4 wt% F₄-TCNQ doped α-NPD (35 nm)/5 wt% BCzVBi doped CBP (20 nm)/5 wt% Ir(ppy)₃ doped CBP (4 nm)/0.65 wt% Ir(btp)₂acac doped CBP (12.5 nm)/BALq (30 nm)/LiF (1 nm)/Al (150 nm).

Device 3: ITO (120 nm)/0.4 wt% F₄-TCNQ doped α-NPD (35 nm)/5 wt% BCzVBi doped CBP (20 nm)/5 wt% Ir(ppy)₃ doped CBP (4 nm)/0.75 wt% Ir(btp)₂acac doped CBP (12.5 nm)/BALq (30 nm)/LiF (1 nm)/Al (150 nm).

Where 4,4'-bis(9-ethyl-3-carbazovinylen)-1,1'-biphenyl (BCzVBi), iridium(III) tris(2-phenyl-pyridinato-N,C^{2'}) (Ir(ppy)₃) and Ir(btp)₂acac were used as blue, green and red dopants, respectively with

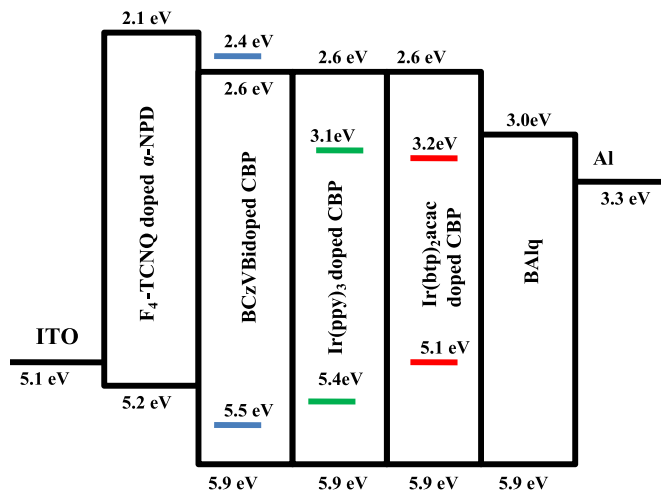


Fig. 1. Schematic energy level diagram for Me-OLED.

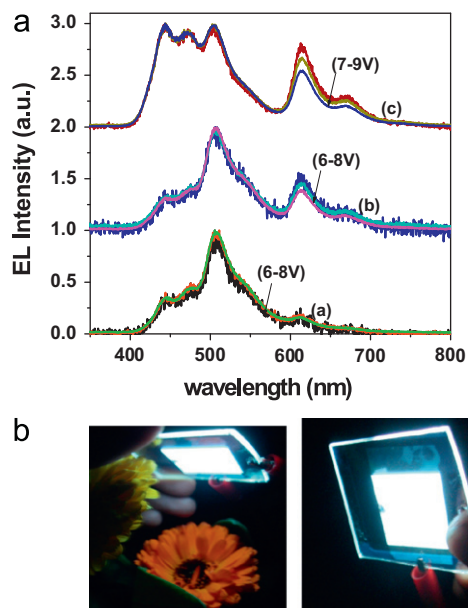


Fig. 2. (a) Normalized EL spectrum for Me-OLEDs with 0.5 wt% (curve a), 0.65 wt% (curve b) and 0.75 wt% (curve c) Ir(btp)₂acac doped CBP. (b) Photographs of device 3.

the common host CBP. Fig. 1 shows the schematic energy level diagram for Me-OLED.

Fig. 2(a) shows the EL spectrum for devices 1, 2 and 3 (curves a, b and c respectively). EL spectrum for device 1 (curve a) has four peaks centered at wavelengths 445 nm, 475 nm, 510 nm and 615 nm. Peaks at wavelengths 445 nm and 475 nm are due to BCzVBi [28]. Peak at wavelength 510 nm is due to Ir(ppy)₃ [28] and peak at wavelength 615 nm is completely attributed to Ir(btp)₂acac [28]. However, shoulder peak of Ir(btp)₂acac was not observed due to small contribution of red emission by Ir(btp)₂acac [28]. No wavelength shift in peak position was observed with the variation of voltage from 6 to 8 V. As the doping concentration of Ir(btp)₂acac was increased to 0.65 wt% in device 2 (curve b), intensity of peaks due to Ir(btp)₂acac increases and the shoulder peak at 670 nm aroused due the triplet emission from Ir(btp)₂acac [28]. When Ir(btp)₂acac doping concentration was further increased to 0.75 wt% in device 3, intensity of peaks due to Ir(btp)₂acac increases. The device has CIE

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