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# Numerical and experimental studies of mixed-host organic light emitting diodes

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ABSTRACT

Electrical characteristic and luminance of three mixed-host organic light emitting diodes (OLEDs): namely the uniformly mixed, step-wise graded and mixed, and continuously graded and mixed, were compared with the conventional hetero-junction OLED in both numerical and experimental studies. These mixed-host OLEDs were fabricated by a mixed-source thermal evaporation process, and half-cell devices were also fabricated to provide some input parameters for OLED simulations. The current efficiencies were largely influenced by their device structures and strongly agreed with the computed current balance factors. The improved mixed-host OLED performances can be discussed with aid from simulations, which include spatial distributions of electron and hole, carrier mobility, electric field profiles, the total recombination rates in the light emitting layer.



## 1. Introduction

In recent years, in contrast to the bi-layer heterojunction (HJ) organic light emitting diode (OLED), uniformly mixed (UM), step-graded mixed (SGM), and the continuously graded mixed (CGM) OLEDs were intensively investigated [1–5]. The concentration profiles of the electron transport and hole transport materials in the light emitting layers (EML) are shown in Fig. 1. The mixed-host OLEDs, in particular CGM-OLED was shown to significantly improve the device stability or operating lifetime by up to six times [1]. The improvement was attributed to the broadening of recombination zone in EML and the removal of bi-layer interfacial barrier which reduced the charge accumulation and heating effect. An experimental comparison of these four types of OLED in terms of luminance, current and power efficiencies was recently reported by us [6], which was largely attributed to the broadening of recombination zone in the light-emitting layer (EML).

Among the mixed-host OLEDs, only UM and SGM have been investigated by numerical simulations for explanations on improved performance. For examples, the electrical behavior and the spatial dependence of recombination rate in the SGM-OLED were numerically simulated, which generally agreed with the experiment [7]. An electrical model was also used to compute the recombination zone and carrier mobility in UM-OLED [8], which was based on the drift-diffusion, thermionic emission and recombination equations. In attempt to explain the enhanced device stability and the effect of electric field on recombination efficiency, a one-dimensional (1D) model [9] was used to simulate the electrical behaviors of UM- and SGM-OLEDs. It is noted that, to the best of our literature search, a more comprehensive comparison between the simulation and experiment for HJ-, UM-, SGM- and CGM-OLEDs for better understanding of the spatial profiles of charge-carriers, electric field, and recombination has probably not been reported.

In this work, half cells or electron-only and hole-only devices are fabricated for the purpose of obtaining the carrier mobility, highest occupied molecular orbit (HOMO) and lowest occupied molecular orbit (LUMO) from curve-fitting of current–voltage characteristics. All four types of OLED are fabricated for comparison with numerical simulations, namely the electrical and luminance characteristic and hence the current efficiency. Improved performance of mixed-host OLEDs, in particular the CGM-OLED, is discussed based on the simulated results for the spatial profiles of electron and hole, electric field and recombination zones and rates, which are correlated to the computed current balance factors.

## 2. Experimental

The fabrication method for HJ-, UM-, SGM- and CGM-OLEDs was previously reported [9]. In this work, the EML thickness was always

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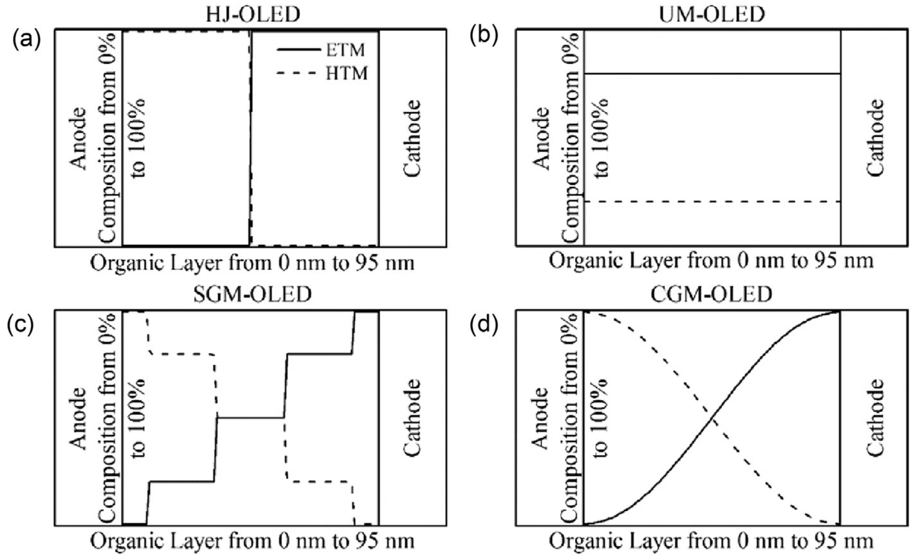


**Fig. 1.** Device structures of the (a) conventional HJ-OLED, and the mixed-host (b) UM-, (c) SGM- and (d) CGM-OLEDs, where solid and dotted lines represent TPD (hole transport) and $Alq_3$ (electron transport) concentrations, respectively.

maintained at 95 nm and cell size of $3 \times 3$ mm$^2$. The indium-doped tin oxide (ITO) coated glass with optical transmittance of 90% and sheet resistance of 20Ω/sq was used, which was patterned into two strips (3 mm width), cleansed in UV bath with acetone, IPA and deionized water sequentially for 15 min each. These were then dried by $N_2$ gas, followed by heating at 100 °C for 15 min.

For UM- and CGM-OLEDs, a mixed source of hole-transport, *N,N′*-Bis(3-methylphenyl)-*N,N′*-diphenylbenzidine (TPD) and the electron-transport, Tris(8-hydroxyquinolinato) aluminum ($Alq_3$) was first prepared at selected ratio by dissolving in chloroform and then dried in quartz crucible at 60 °C. The mixed source was thermally evaporated (Edward, E306) at base pressure of $4 \times 10^{-4}$ Pa. TPD and $Alq_3$ have a large difference in boiling points ($T_b$), 170 °C and 430 °C respectively. Therefore, we observed that initial deposition on ITO was always TPD, followed by mixed-deposition of TPD and $Alq_3$ when the heating temperature of mixed source was increased, then a pure $Alq_3$ layer after TPD was totally vaporized. The concentration profile of CGM-OLED was controlled by a slow heating rate (3.33 mA/s) of the mixed source, whereas that of UM-OELD required a fast heating rate (10 mA/s). For SGM-OLED, five thinner sub-layers of EML were separately deposited with different TPD:$Alq_3$ weight-ratios of (1:0), (3:1), (1:1), (1:3) and (0:1). To improve electron injection, an ultrathin layer of sodium citrate (NaCt) was deposited between the EML and aluminum (Al) cathode.

Half-cell devices of TPD and $Alq_3$, respectively, of two thicknesses (90 nm and 180 nm) were fabricated for the purpose of extracting the carrier mobility, HUMO and LUMO values as input parameters for subsequent OLED simulations. These required current–voltage characteristic measured from half cells to be curve-fitted accurately by the fitting module of the simulation software (SimOLED), as shown in Fig. 2.

## 3. Simulation

A commercially available software, SimOLED (Sim4tec GMbH) was used for half-cell and OLED simulations. The carrier mobilities for electron and hole in mixed-host layers were calculated using the Poole–Frenkel model [10], based on extracted information from half-cells. As the software was not designed for a continuously

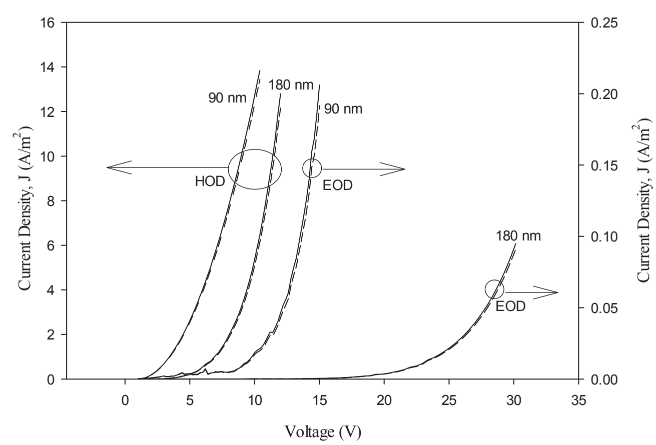


**Fig. 2.** Experimental (solid lines) and curve-fitting (dashed lines) for current–voltage (J–V) characteristics from single-layer, half-cell devices, namely hole-only (HOD) and electron-only (EOD) devices.

graded concentration profile in the mixed-host layer, the EML of CGM-OLED was divided into 40 sub-layers of different TPD:$Alq_3$ ratios yet uniformly mixed. Similarly, the SGM-OLED consisted of 5 sub-layers for its EML.

### *3.1. Simulation model*

The electrical model of 'SimOLED' [11] is based on the Poisson equation (1), Continuity equation (2), Drift–diffusion equation (3) and Exciton rate equation (4):

$$\frac{dE}{dx} = \frac{q}{\varepsilon\varepsilon_0}(p - n) \tag{1}$$

$$\frac{dn}{dt} = \frac{dJ_n}{qdx} - R - T \tag{2}$$

$$J_n = qp\mu_n E + qD_n\frac{dn}{dx} \tag{3}$$