Supplementary Materials for

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**Supplementary Text**

**Supplementary Note 1: Carrier accumulation in device C1.**
The TmPyPB/LiF/Al cathode is efficient for electron injection and has only a low threshold voltage of ~0.4 V (42). Figure S5b shows that holes start to inject at 2.8 V and emission is observed at 3.1 V (Figure S1b). It indicates that both holes and electrons injection are efficient for device C1. Besides, the TAPC/TCTA junction has been demonstrated to not behave like an injection barrier when the bias voltage is over 2.8 V (41). Based on their HOMO and LUMO energy levels, electron injection barrier from TmPyPB and hole injection barrier from TCTA into CBP are both less than 0.2 eV. On the other hand, TCTA and TmPyPB are respectively efficient electron and hole blockers. Thus, the capacitance increases in devices C1 (Figure S5b) is mostly attributed to carrier accumulation in the emitting layer (41, 43-45).

**Supplementary Note 2: “Marburg model” of the conventional devices.**
According to “Marburg model” (Figure S6a), the current of an operating OLED can be expressed as (13, 37, 46-47):

\[
I_h = I_R + I_h' + \frac{dQ_h}{dt} \quad (S1);
\]

\[
I_e = I_R + I_e' + \frac{dQ_e}{dt} \quad (S2);
\]

\[
I_{ext} = I_h + \frac{dQ_A}{dt} + I_e' = I_e + \frac{dQ_c}{dt} + I_h' \quad (S3);
\]

where, \(I_{ext}, I_h\) and \(I_e\) are respectively the external current, injected hole and electron currents at the electrode/organic interfaces. \(I_R, I_h'\) and \(I_e'\) are respectively the electron-hole recombination current and the hole and electron leakage currents beyond the recombination zone. \(Q_A\) and \(Q_h\) are respectively hole charges at the surface of anode and in the device interior. \(Q_c\) and \(Q_e\) are corresponding electron charges.

The “Marburg model” can be used to model carrier accumulation in device interior. Using equations (S1, S2), we can obtain an equation of recombination current \(I_R\):

\[
I_R = I_h - I_h' - \frac{dQ_h}{dt} = I_e - I_e' - \frac{dQ_e}{dt} \quad (S4).
\]

In hole-dominated OLEDs (Figure S6b), the terms of \(I_e'\) and \(dQ_e/dt\) are negligible, and equation (S4) of \(I_R\) can thus be simplified as follow:

\[
I_R = I_h - I_h' - \frac{dQ_h}{dt} = I_e \quad (S5).
\]

**Supplementary Note 3: “Marburg model” of the devices with LHDL.**
Figure S6c shows the proposed working mechanism of an operating hole-dominant OLED with a LHTL for current balance. For the time being, it is assumed that lateral current \(I_L\) is not directly injected from the ITO anode. That is all anode current \((I_h)\) is first injected vertically into the device via the ITO anode. After this, part of the injected hole would spread laterally outside the anode-cathode overlapping region leading to lateral current \(I_h\). After reaching a steady state, current balance should be maintained for the both the regions inside and outside the anode-cathode overlapping region. For the total current (sum of current inside and outside of the electrode overlapping region), we can write:

\[
I_h + I_{e'} = I_e + I_{eL} + I_h' \quad (S6);
\]

where, \(I_e, I_{eL}\) are respectively the cathode injection currents inside and outside the electrode overlapping region, and \(I_h\) is the anode injection current. \(I_e'\) and \(I_h'\) are respectively the electron...
and hole leakage current flow through the whole device. Considering that present device C1 and most other high efficiency devices with suitable hole and electron blocking layers, the leakage currents are negligible and thus equation (S6) becomes,

\[ I_h = I_e + I_{eL} \quad (S7). \]

Considering the current balance outside the electrode overlapping region, we have,

\[ I_{eL} = I_{hL} = I_L \quad (S8). \]

Equation S7 becomes: \( I_h = I_e + I_L \quad (S9). \)

This implies that with a non-zero \( I_L \), \( I_h \) and \( I_e \) can be different suggesting the hole-electron current balance can be achieved via \( I_L \). This gives rise to the third scenario not described by Fave et al (13, 37). However, it should be note that this is obtained with the assumption that \( I_L \) is not due to direct lateral injection from ITO anode. If this assumption is not valid, equation S9 will become: \( I_h + I_L = I_e + I_L \). This means that \( I_h \) has to be equal to \( I_e \). This gets back to the second scenario described by Fave et al (13, 37), implying that accumulation of major carriers would be needed to achieve the current balance. The above analysis suggests that whether we can achieve current balance without involving (or with much reduced) carrier accumulation, depend on whether the lateral current \( I_L \) is a direct injection from anode or not.

Supplementary Note 4: Extraction of diffusion coefficient using Warburg short model.

As shown in Figure 2, the PE4083/PE8000 shows a typical semi-infinite linear diffusion, while the PE4083-MeOH/PE8000 shows a transmissive finite-length linear diffusion. To extract the diffusion coefficient \( D \), the low-frequency impedance spectra are fitted by using Warburg Short (Ws) model (transmissive boundary) with the following equation:

\[ Z_{WS} = \frac{R}{(j\omega)^{P}} \tanh[(j\omega)^{P}] \quad (S10), \]

Where, \( R \) is the diffusion resistance, \( \omega \) is the angular frequency, \( P \) is a constant between 0 and 1. The parameter \( T \) is related to the diffusion length \( L \) and the diffusion coefficient \( D \) \( (T = L^2/D) \). With the data \( L=0.5 \) mm and \( T=0.68 \) s, the diffusivity \( D \) of the PE4083-MeOH/PE8000 layer is \( \sim3\times10^{-3} \) cm\(^2\)/s. With \( L<0.5 \) mm and \( T=135.5 \) s, the diffusivity \( D \) of the PE4083/PE8000 layer is determined to be lower than \( 1.8\times10^{-5} \) cm\(^2\)/s.

Supplementary Note 5: Capacitance-frequency characteristics.

Depending on the DC bias, capacitance of OLEDs can be written as (48):

\[ C = \frac{dQ}{dv} = \frac{dQ_s + dQ_t - dQ_r}{dv_{ac}} \quad (S11). \]

Here, \( Q_s \) is the charge amount at the surface of electrodes, \( Q_t \) is the trapped charge in the device interior, and \( Q_r \) is the charge consumed by recombination process. When the frequency of alternating current (AC) small signal is low, fast recombination process can consume the slow carriers injected by small signal, easily resulting in a rapid decrease in capacitance and even negative capacitance (48).

When bias voltage is zero, the internal trapped charge \( Q_t \) and the recombination charge \( Q_r \) make no contribution to capacitance. The capacitances of devices C2 and D2 under zero bias are contributed by electrode surface charging process in response to AC small signal. Devices C2 and D2 exhibit similar capacitance at intermediate and high frequencies (Figure S15), which is determined by intrinsic dielectric properties of the parallel-plate capacitor system. In the low frequency region, the interfacial state density for charging dominates the capacitance (48), and its contribution will decrease with the frequency increases since it is not able to respond to the high-frequency AC signal. As shown in Figure S15, device D2 only has a much higher capacitance than
device C2 at low frequency. It indicates that higher geometric capacitance is caused by higher acceptor density of PEDOT:PSS films treated by MeOH.

**Supplementary Note 6: Lateral current driven by diffusion behavior.**

Inside the diffusion layer, holes will diffuse along the x-direction and injected into the HTL in the y-direction. We now consider an elementary volume of length $\delta x$ as shown in Figure S21. Under steady state condition, the net rate of hole diffusion into the elementary volume shall equal to the rate of hole injection into the HTL and described as,

$$D \frac{\partial^2 \rho}{\partial x^2} - k \rho = 0 \quad (S12);$$

where, $k$ is the hole injection rate and $\rho$ is the volume density of hole. Solving the equation gives:

$$\rho = \rho_0 \exp \left( - \frac{k}{D} x \right) \quad (S13);$$

where, $\rho_0$ is the hole density at the side edge of ITO where $x = 0$.

Additionally, diffusion flux $J$ can be defined by Fick’s first law:

$$J = -qD \frac{d\rho}{dx} = q \rho_0 \sqrt{Dk} \exp \left( - \frac{k}{D} x \right) \quad (S14).$$
Figure S1. Characteristics of devices C1, C2, D1 and D2.
(a) Maximum efficiencies, (b) current-voltage-total luminescence, (c) current efficiency-current and (d) power efficiency-current characteristics of devices C1, C2, D1 and D2.
Figure S2. Parameters and calculated results of the rate equation for device C1.
(a) Triplet lifetime of green phosphorescent material Ir(ppy)$_3$. (b) Calculated triplet ($n_T$) and polaron ($n_P$) densities in device C1.
Figure S3. Device performances of common green phosphorescent OLEDs without anode buffer layer or hole buffer interlayer.

(a) Device structure of common green phosphorescent OLEDs without or without anode buffer layer and hole buffer interlayer. These devices are marked as device C(x, y). x is 0 or 1, for absence or presence of PE4083 hole buffer layer, and y is 0 or 1, for absence or presence of a TCTA hole buffer interlayer. (b) Current density-voltage-brightness and (c) EQE-current density characteristics of these green phosphorescent OLEDs.
Figure S4. Structure of hole-only (C1h) and electron-only (C1e) devices.
Figure S5. Characteristics of single-carrier devices for device C1. (a) Current density-voltage characteristics of devices C1h and C1e. (b) Capacitance-voltage characteristics of device C1, and the red spheres (at ~3.1V) show its EL onset (Figure S1b). The accumulated hole surface density can be estimated as: \[1.1 \text{nF} \times 4 \text{V} / (1.6 \times 10^{-19} \text{C} \times 3 \text{mm} \times 5 \text{mm}) = 1.8 \times 10^{11} \text{cm}^{-2}.\]
Figure S6. Marburg models for the conventional devices and the proposed devices with LHDL.

(a) Schematic view of an operating OLED according to the Marburg model. (b) Schematic view of an operating conventional hole-dominant OLED for current balance, (c) schematic view of an operating hole-dominant OLED with LHDL for current balance.
Figure S7. Chemical compositions of different types of PEDOT:PSS layers.
(a) S2p X-ray photoelectron spectroscopic spectra of different types of PEDOT:PSS layers. The peaks observed at the binding energy of 168 eV correspond to the sulfur signal from PSS chains, and the peaks around 164 eV correspond to the sulfur signal from PEDOT chains. (b) Schematic diagram of composition and configuration changes in different types of PEDOT:PSS layers (39).
Figure S8. Size parameters of PEDOT:PSS layers for resistance and capacitance measurements.

Size parameters of PEDOT:PSS layers used for resistance measurements (a=1 mm, b=10 mm, c= layer thickness) and capacitance/impedance measurements (a=0.5 mm, b=8 mm, c= layer thickness).
Figure S9. Device structures of devices C1, C2, D1 and D2.
Figure S10. Conductivity characteristics of PEDOT:PSS layers.
Figure S11. Lateral real versus imaginary impedance spectra of PEDOT:PSS layers. Lateral real versus imaginary impedance spectra of (a) PE4083 and (b) PE4083-MeOH layers. The insets show their equivalent circuits and fitting parameters, and all the fitting is conducted with the ZView software.
Figure S12. Transient EL response of * point of device D2 (labelled as D2*).

The voltage pulse width for D2* point was 480 µs, and all the voltage pulse heights correspond to 6 V. The initial part of the transient EL was magnified, as shown in the inset. The green vertical arrow in the inset shows the EL onset.
Figure S13. Transient EL response of device D2 measured by using the repeated pulse voltage.

(a) Pulse voltages with a fixed bias voltage of 6 V at the first half of the cycle (0~80 µs) and a reverse bias voltage ranging from 0 V to -6 V at the second half of the cycle (80~160 µs). (b) Transient EL characteristics of device D2 measured by using the repeated pulse voltage with a reverse bias voltage ranging from 0 V to -6 V. The reverse bias voltages can remove the residual charges in PEDOT:PSS such that the transient EL characteristics will not be influenced by the residual charges in the repeated cycles.
Figure S14. Structure of hole-only (C2h, D1h and D2h) and electron-only (C2e, D1e and D2e) devices.
These single carrier devices were prepared by modifying the structure of devices C2, D1 and D2.
Figure S15. Capacitance-frequency characteristics of devices C2 and D2 without direct current (DC) bias voltage.
Figure S16. Brightness-voltage characteristics of devices C1, C2 and the ITO-cathode overlapping area of devices D1 and D2.
Figure S17. Ultraviolet photoelectron spectra of PE4083, PE8000 and PE4083-MeOH films.
Figure S18. Current-voltage characteristics of single-carrier devices C1e, C1h, D1e and D1h (-e and -h represents electron-only and hole-only device, respectively).
Mott-Schottky analyses (49) on the single-carrier devices are performed by measuring the capacitance:

\[ C^{-2} = \frac{2}{e\varepsilon A^2N} (V_{bi} - V), \]

where \( C \) is the measured capacitance, \( q \) is the elementary charge, \( \varepsilon \) is the permittivity, \( A \) is the active area, \( N \) is the doping density of the donor, and \( V_{bi} \) is the built-in potential.
Figure S20. Normalized brightness with different distance from the ITO-cathode overlapping region.
Figure S21. Carrier diffusion and injection behaviors in the lateral diffusion layer.
Figure S22. Sizes of ITO array electrodes and nominal light-emitting area $S^*$ for OLEDs with ITO array electrode and LHDL.
Figure S23. The real versus imaginary impedance spectrum of device D2AA’ with ITO grid array electrode under 8 V.
Figure S24. Infrared images of device D2AA’ after continuously operating.
(a) 0 min, (b) 5 min, (c) 10 min, (d) 30 min and (e) 60 min, and (f) its visible image after continuously operating for 60 min.
Figure S25. Resistance characteristics of different parts of device D2AA’.
Resistance characteristics of ITO anode ($R_{ITO1}$ and $R_{ITO2}$) and PEDOT:PSS film (Rx and Ry) of device D2AA’, and resistance characteristics of device D2LA (10 mm×12 mm) and its partial light-emitting area on a ITO finger (10 mm×50 μm).
Figure S26. Device performances of device PE-LA.
(a) Schematic diagram and image (operating at 4 V) of device PE-LA using only methanol-treated PEDOT:PSS as anode. (b) Current-voltage-luminous flux and (c) EQE-voltage characteristics of device PE-LA.
Figure S27. EQE-luminous flux characteristics of device C2LA, D2LA and D2AA'.
Figure S28. Normalized EL spectra of devices C2LA, D2LA and D2AA' at different voltages.
Figure S29. Variations of outcoupling efficiencies of dipole sources inside and outside the ITO-cathode overlapping area of device D2AA’.

(a) Power dissipation spectra (@520 nm) and (b) outcoupling efficiencies of dipole sources inside and outside the ITO-cathode overlapping area of device D2AA’. Using the classical theory of electromagnetism (50, 51), we have calculated the power dissipation spectra (Fig. S29a) of dipole sources inside and outside the ITO-cathode overlapping area (area with ITO and area without ITO). It is found that the absence of ITO in diffusion area would mainly change waveguided modes, and thus lead to variations of far-field mode and outcoupling efficiencies (Fig. S29b). Nevertheless, after convoluting the power dissipation spectra with the emission spectra, the overall outcoupling efficiencies of the two areas are similar as one is 20% and the other is 19%.
Figure S30. Transient PL characteristics (@520 nm) of devices corresponding to area with ITO and area without ITO of device D2AA’.

To investigate the influence of microcavity effect, we measured the transient PL characteristics (Figure S30) of devices corresponding to area with ITO and area without ITO of device D2AA’. It can be observed that Ir(ppy)_3 emitters of the two areas should have the same exciton lifetime. It indicates that the radiative recombination rates of the two areas are not changed by the Purcell effect, and thus have no contribution to the luminous flux and efficiency. It should be attributed to the absence of highly reflective materials at the anode side of both the two areas.
Figure S31. Generality of lateral hole diffusion on OLEDs with different emission materials. EQE-luminous flux characteristics of (a) blue phosphorescent device, (b) red phosphorescent device and (c) thermal active delay fluorescent (TADF) device, their emitting layer is TCTA:10wt% Firpic, CBP:10wt% Ir(MDQ)$_2$(acac) and CBP:10wt% CzDBA, respectively.
Table S1. Rate parameters determined by fitting the experimental EQE-j characteristic of devices C1.

| Device | $K_L$ (cm$^3$ s$^{-1}$) | $K_{TT}$ (cm$^3$ s$^{-1}$) | $K_{TP}$ (cm$^3$ s$^{-1}$) |
|--------|------------------------|---------------------------|---------------------------|
| C1     | 1.4×10$^{-11}$         | 7.6×10$^{-12}$            | 5.3×10$^{-12}$            |
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