Sustaining external quantum efficiency over wide range of current in a unipolar organic light emitting field effect transistor

Shih-Wei Chiu1, An Hsu1, Lei Ying2, Yong-Kang Liaw1, Kun-Ta Lin1, Jcng Ruan1, Ifor D. W. Samuel3 and Ben Bang-Yu Hsu1*

1 Department of Materials Science and Engineering, National Cheng Kung University, Taiwan.

2 Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou 510640, P. R. China.

3 Organic Semiconductor Centre, SUPA, School of Physics and Astronomy, University of St Andrews, U.K.

Keywords: Semiconducting polymers, light emitting transistors, conjugated polyelectrolytes, band bending

Organic light emitting field effect transistors (OLEFETs) with bilayer structures have been studied in order to obtain both current density and brightness by combining high-mobility materials and high emissive materials. These devices however are mainly hole-dominated, which cause the efficiency to degrade significantly at high brightness due to charge imbalance. To overcome this fundamental issue, we introduce a novel interfacial engineering ~2 nm into an organic/inorganic hybrid drain to significantly enhance charge capture. We show that the interfacially engineered device forms a bending electronic structure that further accumulate the injected electrons in the light emitting layer and thus effectively capture more holes. The numerical simulation of our capture model suggests that the Coulombic attractions of high density electrons will dominate when hole current is high. As a result, the effect of imbalanced charge transport is overcome, leading to a nearly constant external quantum efficiency (EQE) over a wide range of operating current, EQE~0.23 % over 3 orders of magnitude of current, 1.2 to 2700 mA/cm² corresponding to brightness 4 to 7700 cd/m². The same enhancement is retained after increasing EQE to ~0.51%. Having such tunable brightness with sustained emissive efficiency in an OLEFET will revolutionize the new applications in lighting and display technologies that desire high intensity.
Introduction

Organic light emitting field effect transistors (OLEFETs) have been an attractive emerging class of light emitting devices that combine the functions of electrical switching and optical emission (1-5). Their ability of being integrated with driving circuits into a single device makes OLEFETs convenient and versatile devices with the potential for low-cost display and lighting technologies. So far, OLEFETs can only be advanced on their emission intensity (6–11) or external quantum efficiency (EQE) (12–15), even though huge efforts have been put on. High EQEs in OLEFETs rely on the charged carrier transports to be balanced (16–19). Because electron and hole transports dominate in different voltage regimes, the overlapped operation window which shows the balanced charge transports is therefore small, leading to a severe limit of the highest available intensity. In contrast, high-mobility OLEFETs can reach high current and high intensity but increasing the current in hole-dominated OLEFETs also enhances the charge imbalance that degrades EQE (9-11). To gain both high intensity and high EQE together remains a big challenge in OLEFETs, meanwhile their EQEs will all drastically descend when the operating current is high.

Those bottlenecks above have restricted the development of display and lighting applications. Motivated by that, instead of pursuing the highest EQE, we seek to a new OLEFETs which not only can emit strong and controllable light intensity but sustain a EQE over a wide range of operating current, i.e light intensity proportionally responds to its driving current, for more practical use. Recently, high-mobility OLEFETs with polymeric heterostructures have been extensively investigated for applications that require high intensity such as electrically-pumped organic lasers (8–11). Based on this concept, we developed a bilayer OLEFET with a transparent organic/inorganic hybrid contact (drain) comprising of the specially designed interfacial electronic interactions between drain and p-type light emitting polymers. This interfacial engineering is delivered through a n-type conjugated polyelectrolyze (SPFN) particularly chosen for its higher electron conduction that can mediate the band alignment with p-type light emitting polymer. The electronic alignment will cause band bending to accumulate electrons, which cannot be achieved by the conventional neutral or ionic conjugated polyelectrolyzes (CPEs) (20–23).

Figure 1a shows our complete device structure. A pair of the n-type hybrid drain and its counter p-type gold electrode (source), which forms a hybrid contact-OLEFET (HC-OLEFET) is introduced for more balanced charge injections. The source is composed in order of PATBT (30 nm)/Au (90 nm) and the drain is composed of SPFN (2 nm)/Au:Ag (1:7 nm)/MoOx (30 nm), where PATBT is the p-type semiconducting polymer. SPFN and the bimetallic contact (Au and Ag) are the organic and inorganic parts of the hybrid drain. Super yellow (SY) is the p-type light emitting polymer. Their chemical structures are provided in Figure 1b. MoOx is added as a protection layer to prevent the hybrid drain from oxidation. The large energy mismatch between SY and Ag is reduced by SPFN with the polar amino side group and the lowest unoccupied molecular orbital (LUMO) in middle, therefore increasing electron injection. The right of Figure 1a is the energy diagram showing materials energy relation.
The top scheme of Figure 1c is the electronic structures of n-type SPFN and p-type SY before and after they contact. Once two materials contact, their Fermi levels $E_F$ (electron chemical potentials) will reach a new thermal equilibrium so aligning the SPFN $E_F$ with the SY $E_F$. This alignment energetically bends the SY LUMO downward and forms a potential well. This bending effect was shown by an emerging spectral feature around 141 nm in the SY/SPFN sample, which is absent in SY and SPFN spectra (the bottom of Figure 1c). After biasing the HC-OLEFETs, electrons and holes will start to transport and accumulate on the SY interface (see Figure 1d). Those accumulated electrons in SY have low mobility and will greatly enhance capture efficiency. As the result, the EQE degradation can be greatly reduced by factor of 2.5 to 4.0 compared to recent works\textsuperscript{9,11}. To study how the EQE can sustain, we also made a charge capture model. The simulated result suggests that Coulombic capture originated from electrons can facilitate efficient electron-hole recombination. At the extreme case of static electrons, EQE can reach a constant. Our detection of sustained EQE supports our model.

**Experimental Methods**

When fabricating bilayer OLEFETs, we first made the bottom transistors and used diamond lapping films to rub the substrate for grooves with 50~100 nm width and $\leq 10$ nm depth. The nano-grooved SiO$_2$ substrates were passivated by decyltrichlorosilane (DTS). After passivating DTS, we spin-coated poly(3,6-dialkylthieno[3,2-b]thiophene-co-bithiophene) (PATBT, purchased from Merck Chemicals Ltd) at 5000 rpm for 40 sec from hot chlorobenzene solution at 90 °C. Then, a 30 nm-thick PATBT was successfully coated onto the DTS-passivated nano-grooves. Groove density has been shown critical to tune charge carrier mobilities of organic transistors (8). Therefore, we measured the mobilities for different rubbing times on the substrates as demonstrated in Figure S1. Based on that, we chose 4 rubbing cycles and 6 rubbing cycles for two devices with high and low mobilities.

On the PATBT, 40 nm gold (Au) were thermally deposited through the silicon shadow mask for source electrodes (See Figure S2). Then we spin-coated the light emitting polymer, Super Yellow (SY) from Merck, at 2000 rpm 50 sec from a 0.6 wt% solution in toluene to produce a SY layer ~100 nm. After baking the PATBT/SY bilayer at 80 °C for 20 minutes, we spun the conjugated polyelectrolyte, poly[9,9-bis(6''-(N,N-dimethyl)-hexyl)-2,7-fluorene]-alt-[3,7-dibenzothiophene-S,S-dioxide] (SPFN) at 3000 rpm for 50 sec and baked at 70 °C for 10 minutes. The drain electrodes were deposited on SPFN in the sequence of Au (1 nm), Ag (7 nm), and molybdenum oxide (MoO$_x$, 30 nm) through the silicon mask for drain electrodes. Here Au played a role as a wetting layer to smoothen the following layers. We also prepared a hybrid drain without Au (SPFN/Ag/MoO$_x$) for comparison (refer the smooth/rough films to Figure S3ab). The 7 nm Ag layer was covered by MoO$_x$ layer to prevent the hybrid drain from oxidation. This pair of asymmetric electrodes, the hybrid drain (SPFN/Au:Ag/MoO$_x$) and the source (Au), formed a hybrid contact-OLEFET (HC-OLEFET) with a contact size $0.1 \times 0.0012$ cm$^2$. The transistor channel was fabricated in 0.1 cm width and 40 µm length. Commercial highly-doped n-type silicon wafers with dry thermal SiO$_2$ were
used as the gate electrodes. The thickness of SiO$_2$ is 300 nm and its capacitance is 10.7 nF/cm$^2$. Figure 1b shows the chemical structures of PATBT, SY, and SPFN.

**Figure 1.** (a) Actual image of a HC-OLEFET and its schematic asymmetric contacts with electrons (blue circles) and holes (orange circles) accumulated below hybrid drain shown in the electronic structures on the right. (b) Chemical structures of PATBT, Super Yellow (SY), and the n-type conjugated polyelectrolyte (SPFN). (c) The top scheme is the electronic structures of SY and SPFN before and after they contact. The UV-VIS absorption spectra of SY, SPFN, and SY/SPFN are below. A new feature on 545 nm appears in the SY/SPFN spectrum and corresponds to the bending effect in the top scheme. (d) The energy diagram when biasing the device. The blue and orange area are the electrons and holes accumulated due to the bending effect.

To guarantee electronic activity and optical transparency, the hybrid drain must be
thin (<< optical wavelength) and smooth to maximize the electronic performance and optical output. Otherwise, an injection barrier will be introduced due to a thick and rough SPFN. Optical scattering on the rough surface (Figure S3) will decrease the transmission and turn the hybrid drain bluish and opaque as shown in the inset of Figure S4a. To test the transparency of the hybrid drains, we put two SPFN-coated glass substrates with and without 1 nm-Au beside the devices to share the same deposition procedure. The transmission spectra from the multilayer drains on glass substrates were collected using a reflective spectrometer (Shimadzu UV3600). All the transmission spectra were normalized to the spectrum of a clear glass substrate (see Figure S4a).

The electroluminescence spectrum of an operating HC-OLEFET at \( V_{DS} = V_{GS} = -100 \) V was collected by a mini-spectrometer (Ocean Optics USB2000+) through a 10× objective, shown in Figure S4b. To simultaneously measure the electrical and optical performance of HC-OLEFETs, we utilized two Keithley source measurement units and a calibrated photomultiplier tube. All HC-OLEFETs were tested in a \( \text{N}_2 \) glovebox with \( \text{O}_2 < 10 \) ppm. The topographic images of the hybrid drains were obtained by the Bruker Innova.

**Results and discussions**

For optimizing both optical emission and electrical conduction, we require a thin hybrid drain and to have its surface as smooth as possible. Figure 2a shows the topographic images of Ag without (left) and with (right) Au by atomic force microscope. In the case without Au, Ag forms larger particles that scatter more light, so the hybrid drain looks opaque. In contrast, with Au acting as a wetting layer, the hybrid drain looks transparent (See the inset of Figure S4a). Using software (ImageJ and NanoScope) to analyze Figure 2a, we got averaged particle sizes 10.0 nm and 4.0 nm with their surface roughness \( R_q = 1.3 \) nm and \( R_q = 0.7 \) nm for the hybrid drain without and with Au layer (Figure S3). We measured the transmittance of Au-added hybrid drain and obtained \~80\% at wavelength range 550 to 630 nm which covers the maximum electroluminescence of SY (refer to Figure S4ab).

The energy mismatch for electrons between Ag and SY was reduced by SPFN. To quantify the extent of mismatch reduction, we utilized Kelvin probe force microscope (KPFM) to measure the surface potential of SY and SY/SPFN. Ahead of the measurement, the SPFN was smoothened by 3-time methanol flushing to avoid possible jittering potentials caused by KPFM tip hitting aggregates (see Figure S5). Figure 2b shows the results of potential mapping of SY and SY/SPN with ITO as a reference. The potentials are uniform and are reliably read as \( \Phi_{SY} = -296 \) and \( \Phi_{SY/SPFN} = -85 \) mV (set \( \Phi_{ITO} \) as the reference). Accordingly, the mismatch is reduced by 211 mV so the electron conduction shall be higher. With sequentially methanol-flushed SPFN sample scanned by conductive AFM (cAFM), we found that the currents from ITO, SY, SPFN to the Au-coated cAFM tip increase with flushing times and exhibit inverse dependence of topographic heights (as shown in the red circles in Figure 2c). The I-V curves obtained by cAFM do show a 10-fold increase of electron conduction from 10 to 101 pA (see Figure S6 for details). Because the deeper highest occupied molecular orbitals (HOMO) of SPFN compared to that of SY are particularly designed to block the hole current in SY, the improved conduction is mainly attributed to the electrons.
and is expected to enhance recombination current for light emission.

Figure 2. AFM images of the hybrid drains. (a) The topographies of the hybrid drains without 1 nm Au, the averaged particle size $r=10$ nm and surface roughness $R_q=1.3$ nm (left) and with 1 nm Au, $r=4$ nm and $R_q=0.7$ nm (right) (b) The topographies and their surface potentials mapping for ITO/SY (left) and ITO/SY/SPFN (right). The insets show the surface potentials: $\phi_{SY}=-296$ mV and $\phi_{SY/SPFN}=-85$ mV (Reference $\phi_{ITO}=0$ mV). (c) The topographic images (left) and their current mapping (right) of SY/SPFN washed for (i) 1, (ii) 2, and (iii) 3 times. The red circles demonstrate the opposite tendency between height and current.

We performed transfer and output I-V characteristics on two different HC-OLEFETs to get more insights into the optical response of increased electrons. The two devices have mobilities $\mu=0.23$ cm$^2$/Vs and $\mu=0.52$ cm$^2$/Vs corresponding to their
rubbing times (6 and 4 respectively). Their electrical and optical performance were presented in Figure 3 and listed in Table 1. Then the mobilities can be extracted from the transconductance equation $I_{DS} = \frac{WC}{2L} \mu (V_{GS} - V_{th})^2$, where W and L are the channel width and length, C is capacitance and $I_{DS}$, $V_{GS}$, $V_{th}$ are source-drain current, gate-to-source voltage and threshold voltage respectively. The transfer I-V in Figure 3a are operated at $V_{DS} = -100$ V. The results show that both devices have a tendency for brightness to increase with $I_{DS}$ and the device with higher mobility ($\mu = 0.52 \text{ cm}^2/\text{Vs}$) reach the highest brightness. However, when investigating the output I-V by scanning the source-drain voltage ($V_{DS}$) through 0 to -100 V with varied $V_{GS}$, as shown in Figure 3b, the brightness tended to roll-off at large current for 0.52 cm$^2$/Vs mobility device while approached saturation (no roll-off) for 0.23 cm$^2$/Vs one. We conclude that the lower mobility device gets higher efficiency. In Figure 3c, we filmed the electroluminescence together with the output measurements to ensure their performance reliability. The luminescent area (pixel) at large current ($V_{GS} < -20$ V) was bright and uniform, indicating no voltage loss found along the electrodes. In OLEFETs often only part of pixel emits light, but here we see the entire length of the pixel emitting light. Furthermore, the ultrathin hybrid drains ($< 12$ nm) remarkably remain intact after operating at high voltages.

We further looked into the roll-off at the high current density regime in 0.52 cm$^2$/Vs mobility device, which implied a substantial efficiency degradation there. Considering the possible interactions involved in external quantum efficiencies (EQE) that relate to carrier densities, photoluminescence quantum yield and capture efficiency can be significantly decreased by singlet quenching and imbalanced charge capture at high current density. Previous studies reveal that a current density of at least 6 A/cm$^2$ is needed to induce substantial triplet-singlet and polaron-singlet quenching (24–26). Since our highest current density is only around 2.7 A/cm$^2$, the main mechanism for the efficiency roll-off at high current regimes is due to imbalanced charge capture. Normally, capture efficiency decreases when one charge carriers (typically holes) move faster. Conventional solutions for that are either slowing down hole transport or accelerating electrons. Here we present an alternative way that enables us to maintain the efficiency even at high current.

According to the transconductance equation, it is reasonable that we revisit Figure 3a by taking the square root of $I_{DS}$ and brightness, denoted SRI$_{DS}$ and SRB, to clarify the correlation between hole-only transport and its capture by electrons. In Figure 4ab, the SRI$_{DS}$ and SRB versus $V_{GS}$ curves for both devices are almost identical, implying that the optical output also follows the transistor-based equation. The brightness-to-$I_{DS}$ curves in Figure 4c indeed approximate a linear relationship and show a sustained efficiency over 3 orders of magnitude of $I_{DS}$ (see Figure 4d). So that such devices can fully respond to a wide range of operating current while retain a nearly constant EQE (~0.51 % for 0.23 cm$^2$/Vs and ~0.23% for 0.52 cm$^2$/Vs drop 14~20 % over 4 order of magnitude of current). It is generally known that higher current density shall amplify charge imbalance, and EQE is expected to drop significantly at high hole density. A recent study (9) showed an EQE of 0.8% at a brightness ~10,000 cd/m$^2$, but fell to 0.4% at the highest brightness 29,000 cd/m$^2$. Another work (11) demonstrated 0.1% EQE at a brightness ~10,000 cd/m$^2$ but dropped to ~0.02% at the highest brightness ~20,000 cd/m$^2$. To account for the sustained EQE, we made a capture dynamic model based on Langevin recombination. We will demonstrate that the effect of charge imbalance can be reduced by the accumulation of electrons on the SY/SPFN interface. And we’ll show our model is supported by our experimental results.
Figure 3. Electrical and optical measurements for the devices with $\mu = 0.23 \text{ cm}^2/\text{Vs}$ and $\mu = 0.12 \text{ cm}^2/\text{Vs}$. (a) Transfer I-V characteristics at $V_{DS} = -100 \text{ V}$ (b) Output I-V characteristics. (c) Video images of electroluminescence from the output measurements at $V_{DS} = -100 \text{ V}$ and $V_{GS} = 0$ to $-100 \text{ V}$ in a $-20 \text{ V}$ interval.

Table 1. Electrical and optical performance values extracted from the transfer I-V curves of the devices with hole mobility 0.23 and 0.52 cm$^2$/Vs.

| Hole mobility (cm$^2$/Vs) | On/off ratio | Current density (mA/cm$^2$) | Brightness (cd/m$^2$) | EQE (%) |
|--------------------------|--------------|----------------------------|-----------------------|---------|
| 0.23                     | $3 \times 10^4$ | $0.9 - 750$                | $7 - 4800$            | $\sim 0.51$ |
| 0.52                     | $1 \times 10^5$ | $1.2 - 2700$               | $4 - 7700$            | $\sim 0.23$ |

Considering that charge recombination is the consequence when the transporting electrons and holes meet in SY, we start with the capture efficiency $\eta_c$

$$\eta_c = \frac{J_c}{J_p + J_n - J_c}$$  \hspace{1cm} (Eqn.1)

, where subscripts n and p denote electron and hole, $J_c$ is the capture current
density, and \( J_n = ne\mu_n E \) and \( J_p = pe\mu_p E \) are the electron and hole current densities respectively. We adopted the Langevin recombination model in our model since it is commonly used in disordered organic semiconductors like SY. The Langevin model is only valid when charge mean free path \( \lambda \) is much smaller than thermal capture radius \( r_c \) of Coulomb potential. For SY, \( \lambda \) is typically in the scale of hopping distance \( \sim 1 \) nm and \( r_c = e^2/(4\pi\varepsilon_{SY} k_BT) \sim 18 \) nm is estimated at room temperature \( (25^\circ C) \) with dielectric constant of SY \( \varepsilon_{SY} \sim 3.2\varepsilon_0 \). Therefore, we reasonably model \( J_c \) as the form of \( J_c = eRl \) with the Langevin recombination rate \( R = \gamma np = e(\mu_p + \mu_n)np/\varepsilon_{SY} \) and the mean distance of electron-hole capture \( l \).

Charge carrier transport in a poor conductor like SY will result in charge displacement and restoration, called dielectric relaxation. We treat the transit lengths of electron \( l_n \) and hole \( l_p \), carrier drift velocities times dielectric relaxation times \( (\tau = \varepsilon_{SY}/ne\mu) \), as their maximum dielectric response lengths. So the maximum interaction distance between electrons and holes is defined as the geometric mean of \( l_n \) and \( l_p \), that is \( l \sim l_n l_p/(l_n + l_p) \). With SY thickness 100 nm and operating voltages \( V_{DS}, V_{GS} = -100 \) V, we can calculate \( p \sim 1.1 \times 10^{16} \) 1/cm\(^3\) from \( I_{DS} \) and obtain \( l_p = 18 \) \( \mu \)m. In the extreme condition of full recombination \( (\mu_n/\mu_p = 0) \), see the supporting information S6–S7 for the detailed calculation), \( l_n \) is bounded by 0.18 \( \mu \)m. Both transit lengths are much larger than Coulombic capture radius \( r_c \), which means multiple capture events can happen within \( l \). However, because transporting electrons may escape from being captured, we assume the successful recombination proportion \( \beta l/l_n \) with \( \beta \) as the recombination probability. And the Eqn. 1 can be written as

\[
\eta_c = \frac{J_c/J_p}{1 + J_n/J_p - J_c/J_p} \sim \frac{(1 + \frac{\mu_n}{\mu_p}) \cdot \frac{\beta l}{l_n}}{1 + \frac{\mu_n}{\mu_p} - (1 + \frac{\mu_n}{\mu_p}) \cdot \frac{\beta l}{l_n}} \quad (\text{Eqn. 2})
\]

Since \( l_p \) is much larger than SY thickness \( (100 \) nm), holes can be considered homogeneously distributed in SY. On the other hand, electron transport in SY is expected to be slowed down by the mismatch energy between SY and Ag. The smaller \( l_n \) then dominate the mean interaction distance \( l \) \( (l \sim l_n \text{ when } l_p \gg l_n) \) as we expected in hole-dominated environment. Subsequently, we replace \( l/l_n \) with \( l_p/(l_n + l_p) \) in Eqn. 2 and assume the equation also valid in hopping scale for the later simulation study. In the environment full of holes, electrons only diffuse a short distance around the SY/SPFN interface and quickly recombine with holes. As a result, the capture behavior is diffusion-limited, which satisfies the Langevin model.
Figure 4. (a)(b) Plots of the square roots of $I_{DS}$ (SRI$_{DS}$, black square) and brightness (SRB, red circle) vs. $V_{GS}$ for the devices with $\mu = 0.23$ cm$^2$/Vs and 0.52 cm$^2$/Vs. (c) Brightness vs. $I_{DS}$ curves for the two devices. (d) EQE vs. $I_{DS}$ curves for both devices at $V_{DS} = -100$ V. (e)(f) Simulated capture efficiency $\eta_c$ at different $\mu_n/\mu_p$ (solid lines) for the two devices. Solid squares are experimental data. The resulting diffusion length $\lambda_n = 18$ nm labeled in red are correlated to the Coulombic capture length at 21 °C.

To simulate our capture dynamics model, we fit Equ.2 with electron density $n$, and the recombination probability $\beta$ as the fitting parameters. The actual data $\eta_c$ versus $I_{DS}$ is extracted from EQE in Figure 4d (see the relation between $\eta_c$ and EQE in section S6). Figure 4ef are the simulation results for the two devices with different mobilities ratios $\mu_n/\mu_p$. $\beta$ is around the same order of magnitude $10^{-5}$ for all $\mu_n/\mu_p$. The corresponding diffusion lengths $\lambda_n$ can be obtained from $n$ and $\beta$, in which $\beta = \Delta n/n \sim \lambda^2/\lambda_n^2$ and $\lambda_n = \sqrt{D_n \tau_n} = \sqrt{eKT/\hbar^2}$. The black solid squares represent the experimental data. We can see the curves fit the experimental data better with higher $\mu_n/\mu_p$. For the 0.23 cm$^2$/Vs device, when $\mu_n/\mu_p > 1.4 \times 10^{-3}$ where $\lambda_n = 18$ nm, all the results nearly follow the same curves (the same behavior retains for the 0.52 cm$^2$/Vs
device with $\mu_n/\mu_p > 4.4 \times 10^{-3}$). We notice that 18 nm is the Coulombic capture radius in our cases; the electron density $n < p$ ($n > p$) $\sim 1.1 \times 10^{16}$ $1/cm^3$ when $\lambda_n > 18$ ($\lambda_n < 18$) nm. It indicates a dynamic transition around $\lambda_n = 18$ nm. We therefore suggest that when electrons have higher mobilities and longer diffusion lengths ($\lambda_n > 18$ nm), their capture behaviors are limited by the Coulombic capture (for detailed data, see Table S1). As a result, the effect of Coulombic capture due to accumulated electrons will overcome that of amplified transport imbalance, leading to a sustained efficiency. On the other hand, when electron mobility is low, those slow-moving electrons cannot go far but accumulate within the bending energy well on the interface of SY/SPFN. Consequently, the band bending-induced electron accumulation produces higher electron density than holes. Thereby the efficiency is higher and retaining. In Fig 4ef, $\eta_c$ shows a sustained magnitude over 3 orders of $I_{DS}$ for $\mu_n/\mu_p = 10^{-4}$ and $\lambda_n$ around the hopping scale. In the most extreme condition where $\mu_n/\mu_p \rightarrow 0$ and $n \rightarrow \infty$, $\eta_c$ ultimately reaches a constant (see Figure S7).

Conclusions

In summary, we have developed a HC-OLEFET with transparent organic/inorganic hybrid contact on top of the emissive SY. The hybrid drain which is composed of SPFN and bimetallic contact can induce band bending on the interface of SY/SPFN. So electrons can further accumulate along with electron injection enhancement to compensate charge imbalance. We have shown a sustained high EQE held by two different HC-OLEFETs over 3 orders of magnitude of current. And the lower mobility device has higher efficiency. Through modelling the capture dynamics, we found that the capture efficiency is dominated by Coulombic capture when electron mobility is high and by the band bending effect when electron mobility is low. As a consequence, a stable EQE over a wide range of brightness is achievable by carefully engineering interfacial electronic structures and hence overcome the long-standing fundamental challenge in OLEFETs. This method can also be applied to most p-type organic semiconductors whose charge transports are imbalanced. We show that such practically viable light emitting devices can provide a platform toward high-efficiency display and lighting technologies.

Acknowledgment

The authors are grateful to the financial support of the Ministry of Science and Technology, Taiwan (MOST 110-2112-M-006-025-MY2). We appreciate Dr. Byoung Hoon Lee’s help on the transmission spectrum of the hybrid drain in the supporting information. Dr. Bernard Haochih Liu provided the Bruker Dimension Icon to take a part of cAFM data. We thank to Dr. Feng-Yin Chang’s valuable comments.
References

(1) Muccini, M.; Nazionale, C. PROGRESS ARTICLE A Bright Future for Organic Field-Effect Transistors. *Nat Mater* **2006**, *6*, 605–613.

(2) Zhang, C.; Chen, P.; Hu, W. Organic Light-Emitting Transistors: Materials, Device Configurations, and Operations. *Small* **2016**, *12* (10), 1252–1294. https://doi.org/10.1002/smll.201502546.

(3) Liu, C. F.; Liu, X.; Lai, W. Y.; Huang, W. Organic Light-Emitting Field-Effect Transistors: Device Geometries and Fabrication Techniques. *Advanced Materials* **2018**, *30* (52), 1–34. https://doi.org/10.1002/adma.201802466.

(4) Chaudhry, M. U.; Muhieddine, K.; Wawrzinek, R.; Sobus, J.; Tandy, K.; Lo, S. C.; Namdas, E. B. Organic Light-Emitting Transistors: Advances and Perspectives. *Adv Funct Mater* **2020**, *30* (20), 1–11. https://doi.org/10.1002/adfm.201901282.

(5) Zaumseil, J.; Sirringhaus, H. Electron and Ambipolar Transport in Organic Field-Effect Transistors. *Chem Rev* **2007**, *107* (4), 1296–1323. https://doi.org/10.1021/cr0101143.

(6) Namdas, E. B.; Hsu, B. B. Y.; Liu, Z.; Lo, S.-C.; Burn, P. L.; Samuel, I. D. W. Phosphorescent Light-Emitting Transistors: Harvesting Triplet Excitons. *Advanced Materials* **2009**, *21* (48), 4917–4961. https://doi.org/10.1002/adma.200900919.

(7) Seo, J. H.; Namdas, E. B.; Gutacker, A.; Heeger, A. J.; Bazan, G. C. Solution-Processed Organic Light-Emitting Transistors Incorporating Conjugated Polyelectrolytes. *Adv Funct Mater* **2011**, *21* (19), 3667–3672. https://doi.org/10.1002/adfm.201100682.

(8) Hsu, B. B. Y.; Seifter, J.; Takacs, C. J.; Zhong, C.; Tseng, H. R.; Samuel, I. D. W.; Namdas, E. B.; Bazan, G. C.; Huang, F.; Cao, Y.; Heeger, A. J. Ordered Polymer Nanofibers Enhance Output Brightness in Bilayer Light-Emitting Field-Effect Transistors. *ACS Nano* **2013**, *7* (3), 2344–2351. https://doi.org/10.1021/nn305566u.

(9) Chaudhry, M. U.; Muhieddine, K.; Wawrzinek, R.; Li, J.; Lo, S. C.; Namdas, E. B. Nano-Alignment in Semiconducting Polymer Films: A Path to Achieve High Current Density and Brightness in Organic Light Emitting Transistors. *ACS Photonics* **2018**, *5* (6), 2137–2144. https://doi.org/10.1021/acsphotonics.8b00011.

(10) Nam, S.; Chaudhry, M. U.; Tetzner, K.; Pearson, C.; Groves, C.; Petty, M. C.; Anthopoulos, T. D.; Bradley, D. D. C. Efficient and Stable Solution-Processed Organic Light-Emitting Transistors Using a High- k Dielectric. *ACS Photonics* **2019**, *6* (12), 3159–3165. https://doi.org/10.1021/acsphtotonics.9b01265.

(11) Park, Y. J.; Song, A. R.; Walker, B.; Seo, J. H.; Chung, K. B. Hybrid ZnON–Organic Light Emitting Transistors with Low Threshold Voltage <5 V. *Adv Opt Mater* **2019**, *7* (7), 1–6. https://doi.org/10.1002/adom.201801290.

(12) Swensen, J. S.; Soci, C.; Heeger, A. J. Light Emission from an Ambipolar Semiconducting Polymer Field-Effect Transistor. *Appl Phys Lett* **2005**, *87* (25), 253511. https://doi.org/10.1063/1.2149986.

(13) Zaumseil, J.; Donley, C. L.; Kim, J.-S.; Friend, R. H.; Sirringhaus, H. Efficient Top-Gate, Ambipolar, Light-Emitting Field-Effect Transistors Based on a Green-Light-Emitting Polyfluorene. *Advanced Materials* **2006**, *18* (20), 2708–2712. https://doi.org/10.1002/adma.200601080.

(14) Schidleja, M.; Melzer, C.; von Seggern, H. Investigation of Charge-Carrier Injection in
Ambipolar Organic Light-Emitting Field-Effect Transistors. *Advanced Materials* 2009, 21 (10–11), 1172–1176. https://doi.org/10.1002/adma.200801695.

(15) Gwinner, M. C.; Kabra, D.; Roberts, M.; Brenner, T. J. K.; Wallikewitz, B. H.; McNeill, C. R.; Friend, R. H.; Sirringhaus, H. Highly Efficient Single-Layer Polymer Ambipolar Light-Emitting Field-Effect Transistors. *Adv Mater* 2012, 24 (20), 2728–2734. https://doi.org/10.1002/adma.201104602.

(16) Bürgi, L.; Turbiez, M.; Pfeiffer, R.; Bienewald, F.; Kirner, H.-J.; Winnewisser, C. High-Mobility Ambipolar Near-Infrared Light-Emitting Polymer Field-Effect Transistors. *Advanced Materials* 2008, 20 (11), 2217–2224. https://doi.org/10.1002/adma.200702775.

(17) Zaumseil, J.; McNeill, C. R.; Bird, M.; Smith, D. L.; Paul Ruden, P.; Roberts, M.; McKiernan, M. J.; Friend, R. H.; Sirringhaus, H. Quantum Efficiency of Ambipolar Light-Emitting Polymer Field-Effect Transistors. *J Appl Phys* 2008, 103 (6), 064117. https://doi.org/10.1063/1.2894723.

(18) Capelli, R.; Toffanin, S.; Generali, G.; Usta, H.; Facchetti, A.; Muccini, M. Organic Light-Emitting Transistors with an Efficiency That Outperforms the Equivalent Light-Emitting Diodes. *Nat Mater* 2010, 9 (6), 496–503. https://doi.org/10.1038/nmat2715.

(19) Nakano, H.; Saito, M.; Nakamura, H.; Adachi, C. Highly Balanced Ambipolar Mobilities with Intense Electroluminescence in Field-Effect Transistors Based on Organic Single Crystal Oligo(p-Phenylenevinylene) Derivatives. *Appl Phys Lett* 2009, 95 (3), 033308. https://doi.org/10.1063/1.3184588.

(20) Huang, F.; Hou, L.; Wu, H.; Wang, X.; Shen, H.; Cao, W.; Yang, W.; Cao, Y. High-Efficiency, Environment-Friendly Electroluminescent Polymers with Stable High Work Function Metal as a Cathode: Green- and Yellow-Emitting Conjugated Polyfluorene Polyelectrolytes and Their Neutral Precursors. *J Am Chem Soc* 2004, 126 (31), 9845–9853. https://doi.org/10.1021/ja0476761.

(21) Hoven, C. v.; Garcia, A.; Bazan, G. C.; Nguyen, T. Q. Recent Applications of Conjugated Polyelectrolytes in Optoelectronic Devices. *Advanced Materials* 2008, 20 (20), 3793–3810. https://doi.org/10.1002/adma.200800533.

(22) Huang, F.; Wu, H.; Cao, Y. Water/Alcohol Soluble Conjugated Polymers as Highly Efficient Electron Transporting/Injection Layer in Optoelectronic Devices. *Chem Soc Rev* 2010, 39 (7), 2500–2521. https://doi.org/10.1039/b907991m.

(23) Xiao, H.; Miao, J.; Cao, J.; Yang, W.; Wu, H.; Cao, Y. Alcohol-Soluble Polyfluorenes Containing Dibenzothiophene-S,S-Dioxide Segments for Cathode Interfacial Layer in PLEDs and PSCs. *Org Electron* 2014, 15 (3), 758–774. https://doi.org/10.1016/J.ORGEL.2014.01.006.

(24) Murawski, C.; Leo, K.; Gather, M. C. Efficiency Roll-off in Organic Light-Emitting Diodes. *Advanced Materials* 2013, 25 (47), 6801–6827. https://doi.org/10.1002/adma.201301603.

(25) Zhang, Y.; Forrest, S. R. Triplets Contribute to Both an Increase and Loss in Fluorescent Yield in Organic Light Emitting Diodes. *Phys Rev Lett* 2012, 108 (26), 1–5. https://doi.org/10.1103/PhysRevLett.108.267404.

(26) Staroske, W.; Pfeiffer, M.; Leo, K.; Hoffmann, M. Single-Step Triplet-Triplet Annihilation: An Intrinsic Limit for the High Brightness Efficiency of Phosphorescent Organic Light Emitting Diodes. *Phys Rev Lett* 2007, 98 (19), 8–11. https://doi.org/10.1103/PhysRevLett.98.197402.