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Insights into charge balance and its limitations in simplified phosphorescent organic light-emitting devices

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ABSTRACT

Simplified phosphorescent organic light-emitting device (PHOLED), which utilizes only two organic layers, showed record-high efficiency when first introduced. It is quite surprising that this device can have such high efficiency without the use of complex carrier and exciton confinement layers that are common in the state-of-the-art PHOLEDs nowadays. Therefore, it is important to understand how good charge balance is in simplified PHOLED and why. In this work, we study the effects of altering charge balance in simplified PHOLED through means of changing layer thickness in the hole transport layer (HTL) and electron transport layer (ETL) as well as intentionally doping hole and electron traps in the HTL and ETL, respectively, on device efficiency. The results show that when using high carrier mobility charge transport materials, changing layer thickness does not impact charge balance appreciably. On the other hand, introducing charge traps in a thin layer within the HTL or ETL can, in comparison, influence charge balance more significantly, and proves to be a more effective approach for studying the factors limiting charge balance in these devices. The results reveal that simplified PHOLEDs are generally hole-rich, and that the leakage of electrons to the counter electrode is also a major mechanism behind the poor charge balance and efficiency loss in these devices. In order to optimize charge balance in simplified PHOLED, it is important to reduce hole transport in the device so that e-h ratio can be brought closer to unity, as well as eliminate electron leakage. Finally, we show that by simply using an electron blocking HTL, the efficiency of the device can be enhanced by as much as 25%, representing the highest reported for simplified PHOLEDs.

1. Introduction

Efficiency is one of the most important performance parameters in organic light-emitting devices (OLEDs). The internal quantum efficiency (IQE) of an OLED generally depends on three factors: (i) the fraction of excitons that can decay radiatively, (ii) the quantum yield of the light emitting material, and (iii) the charge balance factor. Adachi et al. showed that the incorporation of a heavy metal atom in the emitter can enhance spin–orbit coupling, thereby allows nearly all excitons to undergo radiative decay [1]. It has also been shown that these emitters have nearly unity quantum yield [2]. This leaves the last factor, charge balance, to require optimization in order to maximize IQE. In general, the charge balance term can be optimized by changing the thickness of the hole and electron transport layers [3], using p-doped and n-doped transport layers or, more commonly, by introducing charge and exciton blocking layers [4]. Most state-of-the-art devices therefore comprise 5 or more organic layers. Inevitably, this prolongs and complicates OLED fabrication. In 2011, however, Helander et al. introduced a simplified phosphorescent OLED (PHOLED) structure that exhibits record-high efficiency yet only employs two organic layers thus significantly simplifies fabrication [5]. Without the additional charge carrier blocking layers, it is quite surprising for these simplified devices to have this high efficiency. Therefore, it is important to investigate the extent of charge balance and the factors that influence it in these devices.

Towards that end, this work addresses the charge balance question in simplified PHOLEDs. More particularly, we study the effects of altering charge balance in these devices by means of changing the thickness of the charge transport layers or introducing charge traps in the transport layers. The results show that when using high carrier mobility charge transport materials,

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changing layer thickness does not impact charge balance appreciably. Therefore, unlike in conventional devices, this approach cannot be used for optimizing charge balance. Introducing charge traps in a thin layer within the hole transport layer (HTL) or electron transport layer (ETL) can, in comparison, influence charge balance more significantly, and proves to be a more effective approach for studying the factors limiting charge balance in these devices. The results reveal that simplified PHOLEDs are generally hole-rich, and that the leakage of electrons to the counter electrode is also a major mechanism behind the poor charge balance and efficiency loss in these devices. Finally, we show that by simply using an electron blocking HTL, the efficiency of the device can be enhanced by as much as 25%.

2. Experimental

In this work, simplified PHOLEDs consisting 4,4′-bis(carbazol-9-yl)biphenyl (CBP) or 4,4′,4″-Tris(carbazol-9-yl)triphenylamine (TCTA) as the hole transport layer and host, 2,2′,2″-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) or 1,3-Bis [3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPhB) as the ETL, and $Ir(ppy)_3$ or bis(2-phenylpyridine)(acetylacetonate)iridium(III) ($Ir(ppy)_2(acac)$) as the emitters are fabricated and tested. The optimized simplified PHOLED has the following structure: MoO_3 (5 nm)/CBP (30 nm)/CBP:$Ir(ppy)_3$ (5%) (15 nm)/TPBi (45 nm)/LiF (1 nm)/Al (80 nm). The organic materials CBP, TCTA, TPBi and BmPyPhB are obtained from Shanghai Han Feng Chemical Co. The iridium-based complexes are obtained from Luminescence Technology Corp. All materials are used as received without further sublimation. Prior to device fabrication, the ITO coated glass substrates are sonicated in acetone and isopropanol for 5 min each, in respective order. Devices are then fabricated in an Angstrom Engineering EvoVac system. All materials are thermally evaporated at a rate of 0.1–2 Å/s at a base pressure of 5×10^{-6} torr. The devices are kept in a N_2 environment during all measurements.

3. Results and discussion

In order to test the extent of charge balance in simplified PHOLED and its dependence on the charge transport layer thickness, we first study the effects of increasing the thickness of the HTL (CBP) in simplified PHOLED on device efficiency. The device structure used in this study is MoO_3 (5 nm)/CBP (x nm)/CBP:$Ir(ppy)_3$ (5%) (15 nm)/TPBi (45 nm)/LiF (1 nm)/Al (80 nm), where x = 30, 60, 90, 120, 150, 180 or 210. Fig. 1(a) shows the current density vs. voltage (J–V) characteristics of these devices. As expected, increasing the CBP thickness results in a shift in the characteristics to higher voltages. Despite the shift, the turn-on voltage is essentially the same for all devices, indicating charge injection has not been altered. Fig. 1(b) shows the current efficiency of the devices at 100 cd/m^2 versus the CBP thickness. As can be seen, the efficiency trend exhibits an oscillating pattern. This curve is not different from the commonly observed microcavity effect trend that occurs when the ETL thickness is varied [6,7], although with a much smaller oscillation amplitude (note the x axis does not cross y axis at 0 cd/A). This “weaker” microcavity can be attributed to the fact that light reflection at the ITO/organic interface is less than that at the organic/metal interface. This trend is however different from what is commonly observed in conventional fluorescent OLEDs (for example, one utilizing NPB and Alq_3 as HTL and ETL, respectively). In those devices increasing the HTL thickness generally leads to a monotonic (rather than an oscillating) change in efficiency as a result of the decrease in hole transport and the consequent increase in the electron–hole (e–h) ratio [3]. In contrast, in our experiment, despite the fact that driving voltage increases with the HTL thickness, indicating the charge balance also changes, such monotonic shift in current efficiency is not seen, and the trend is dominated by microcavity effects. For example, when the devices with 40 nm and 180 nm CBP are compared, with both representing nearly maximum constructive interference conditions, the current efficiency is almost the same. It is therefore reasonable to assume that charge balance does not change significantly even when the HTL thickness is increased by more than 4 times. Although the stark difference between the trend observed here versus that observed in conventional OLEDs [3] can at first glance appear surprising, when one considers that the hole mobility in our HTL, CBP (2×10^{-3} cm^2 V^{-1} s^{-1} at the applied field of 0.5 MV/cm), is one order of magnitude higher than that in NPB (1×10^{-4} cm^2 V^{-1}s^{-1} at the applied field of 0.5 MV/cm) [8,9], the much weaker effect of CBP thickness on charge balance becomes understandable.

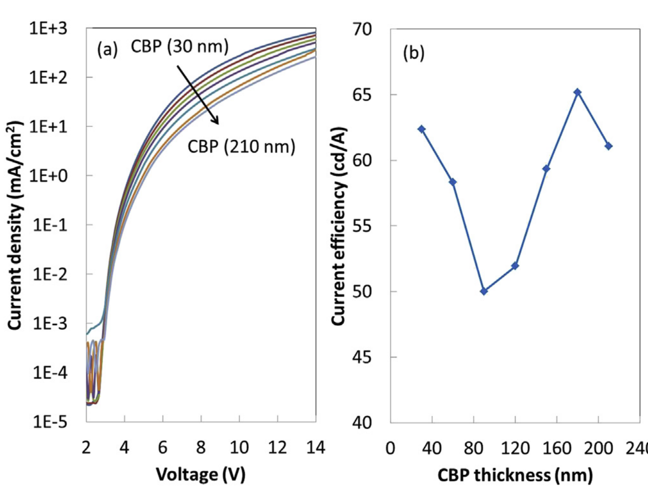

Fig. 1. (a) Current density–voltage characteristics of devices with various CBP thickness. (b) Current efficiency vs. CBP thickness of these devices at 100 cd/m^2.

Given the strong dependence of device current efficiency on microcavity effects, the traditional method of probing charge balance via monitoring device efficiency while varying layer thickness is clearly ineffective in devices utilizing high carrier-mobility materials. Therefore, for examining the factors influencing charge balance, we resort to a different technique: measuring device delayed EL. Delayed EL is the persistent EL that is emitted from a device after the end of the forward bias. In this technique, a device is driven using a square pulse with a pulse width of 0.5 ms (the pulse is sufficiently long enough for prompt EL to reach its steady-state intensity). An optical shutter opens to collect delayed EL 0.3 ms after the end of the forward bias pulse, which is significantly longer than the lifetime of $Ir(ppy)_3$ triplet state lifetime (<1 μs) and thus ensures the absence of any contributions from prompt EL in the collected signal. As such, any collected signal will arise from the radiative decay of excitons that are formed after the end of the forward bias pulse. A detailed description of the delayed EL measurement setup and signal detection protocol is reported elsewhere [10]. One common source of delayed EL is the recombination of charges that were initially trapped but get released after the end of the forward bias pulse. In order to identify contributions by this mechanism to the observed delayed EL, a 0.5 ms reverse bias pulse (which produces a field of 0.74 MV/cm) is applied on the device during the delayed EL signal collection, and subsequent changes in delayed EL characteristics are monitored. Therefore, by comparing the delayed EL signal with and without the reverse bias for a given HTL thickness, the residual charge concentration, and thus the