



High efficiency yellow organic light-emitting diodes with optimized barrier layers



Ye Mu^a, Shiming Zhang^{a,b,*}, Shouzhen Yue^a, Qingyang Wu^a, Yi Zhao^{a,*}

^a State Key Laboratory on Integrated Optoelectronics, College of Electronics Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, PR China

^b Département de Chemical Engineering, École Polytechnique de Montréal, Montréal, Québec H3C3J7, Canada

ARTICLE INFO

Article history:

Received 12 January 2015

Received in revised form 22 June 2015

Accepted 7 August 2015

Available online 25 August 2015

Keywords:

Yellow OLED

PO-01

M-EML

Interlayers

ABSTRACT

High efficiency Iridium (III) bis (4-phenylthieno [3,2-c] pyridinato-N,C^{2'}) acetylacetonate (PO-01) based yellow organic light-emitting devices are fabricated by employing multiple emission layers. The efficiency of the device using 4,4',4''-tris(N-carbazolyl) triphenylamine (TCTA) as potential barrier layer (PBL) outperforms those devices based on other PBLs and detailed analysis is carried out to reveal the mechanisms. A forward-viewing current efficiency (CE) of 65.21 cd/A, which corresponds to a maximum total CE of 110.85 cd/A is achieved at 335.8 cd/m² in the optimized device without any outcoupling enhancement structures.

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1. Introduction

Organic light-emitting diodes (OLEDs) have drawn particular attention due to their potential application in solid-state lighting and flat panel display [1]. Tremendous effort has been devoted to engineering device architectures to satisfy commercial requirements [2,3]. From the point of view of device architectures, multiple-emission layer (M-EML) structure is proved to be an efficient way to facilitate the confinement of electrons and holes within the emission layer (EML), thus increased carrier recombination efficiency will be realized [4,5]. Employing M-EML structure is also an efficient way to improve emission efficiency and adjust emission spectrum [6,7]. In this work, we realized high efficiency PO-01 based YOLED utilizing M-EML structures by engineering devices with different interface buffer materials. The different carrier mobilities, together with different energy-level alignments with the EMLs, allow us to gain insight into the selection rules of buffer layers and reveal the underlying mechanisms. Having taken advantage of the carrier confinement in the M-EML, along with selected potential barrier layer (PBL) and optimized EML numbers, YOLED with a forward-viewing current efficiency (CE) of 65.21 cd/A, i.e., maximum total CE of 110.85 cd/A, is obtained at 335.8 cd/m².

2. Experimental section

The fabrication and measurement procedures are described in detail elsewhere [7]. For the total (maximum) CE calculation, a factor of 1.7 is applied to the forward-viewing CE [9]. We used MoO_x, N,N'-Bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine (NPB) and 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TpPyPB) as hole-injection layer (HIL), hole-transporting layer (HTL) and electron-transporting layer (ETL), respectively. 4,4'-N,N'-dicarbazole-biphenyl (CBP): 6% PO-01 is used as the EML. Two typical hole-transporting materials: 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC), and 4,4',4''-tris(N-carbazolyl) triphenylamine (TCTA), and other two typical electron-transporting materials: tris-[3-(3-pyridyl)mesityl]borane (3TPYMB), and 1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene (TPBi) are used for PBLs.

3. Results and discussions

The first set of devices is demonstrated as follows and the corresponding configurations are shown in the inset of Fig. 1: ITO/MoO_x (10 nm)/NPB (40 nm)/PBL (5 nm)/EML (10 nm)/PBL (5 nm)/EML (10 nm)/TpPyPB (50 nm)/LiF (1 nm)/Al (100 nm). Here EML is CBP: PO-01 (6 wt.%), and PBL stands for TAPC (**device A**), TCTA (**device B**), 3TPYMB (**device C**) and TPBi (**device D**), respectively. Normalized EL spectra of devices, with the schematic energy-level diagrams of devices A, B, C, and D are shown in Fig. 1 [8–13], to study the carrier and exciton confinement as well as recombination profile in the devices. As depicted in Fig. 1, the EL

* Corresponding authors at: State Key Laboratory on Integrated Optoelectronics, College of Electronics Science and Engineering, Jilin University, 2699 Qianjin Street, Changchun 130012, PR China.

E-mail addresses: shiming.zhang.ca@ieee.org (S. Zhang), yizhao@jlu.edu.cn (Y. Zhao).

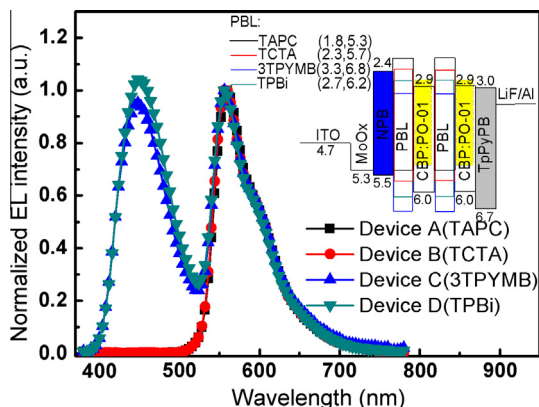


Fig. 1. The normalized EL spectrum of devices A–D at a current density of 5 mA/cm². The inset shows the schematic energy-level diagrams of devices A–D.

spectra of devices A and B show standard PO-01 emission with a peak emission at 556 nm. While for devices C and D, in addition to the PO-01 emission, a strong emission-peak centering at 448 nm, which is assigned to NPB emission [14], is observed, implying considerable amount of holes are blocked at the NPB/PBL interface due to the electron preferable transport nature of 3TPYMB and TPBi [15,16]. In this case, most holes will be used for inefficient fluorescent NPB emission, and few holes can reach the EML to contribute for efficient PO-01 phosphorescent emission, which subsequently results in an extreme low CE of devices C and D compared to that of device A and B (see Fig. 2). The above discussions reveal that, to obtain high CE, it is required to promote hole transport to the subsequent EMLs to suppress the NPB emission, i.e., a hole-transport dominated material is more preferable for the PBL.

TAPC and TCTA, which are served as PBLs for devices A and B, are two typical materials with high hole mobilities. However, it is observed in Fig. 2 that, although the hole mobility of TAPC is nearly 70 times higher than that of TCTA [17], surprisingly, the peak CE of device B is, on the contrary, 1.5 times higher than device A. Aiming to investigate the hidden reasons for the above phenomena, another set of examination devices (T-series) were demonstrated. Fig. 3 shows the J–CE curves of the T-series devices and inset illustrates device configurations of T₁, T₂, T₃ and T₄. It is observed that high CE are obtained in the devices (T₁ and T₃) where the EML is close to TpPyPB ETL regardless the selected PBL. This result suggests that electrons dominate the CE because EMLs in

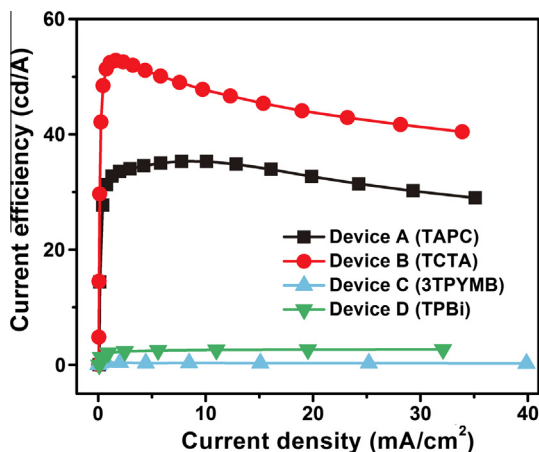


Fig. 2. CE versus current density characteristics of devices A–D.

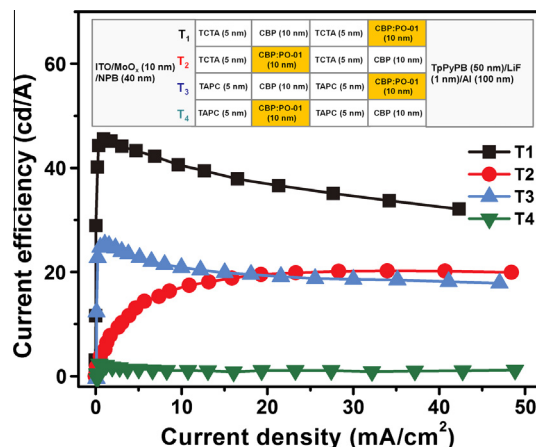


Fig. 3. CE versus current density characteristics of devices T₁–T₄. Inset shows the device structures of T₁–T₄.

devices T₁ and T₃ have higher electron densities with respect to that of devices T₂ and T₄, despite the fact that the latter ones have higher hole densities. In addition to the above facts, a more significant output from Fig. 3 is that, among the high CE devices, the peak CE of device T₁ (TCTA PBL) is approximately 1.8 times higher than that of device T₃ (TAPC PBL). According to this hint, the reason that CE of device B outperforms device A is able to be clarified. As shown in the inset of Fig. 1, the lowest unoccupied molecular orbital (LUMO) energy level of TAPC is 1.8 eV (0.5 eV higher than that of TCTA), and electron mobility of TAPC is also supposed to be lower than TCTA [18,19]. Thus, as compared to device T₁, more electrons injected from the cathode, in device T₃, will be blocked

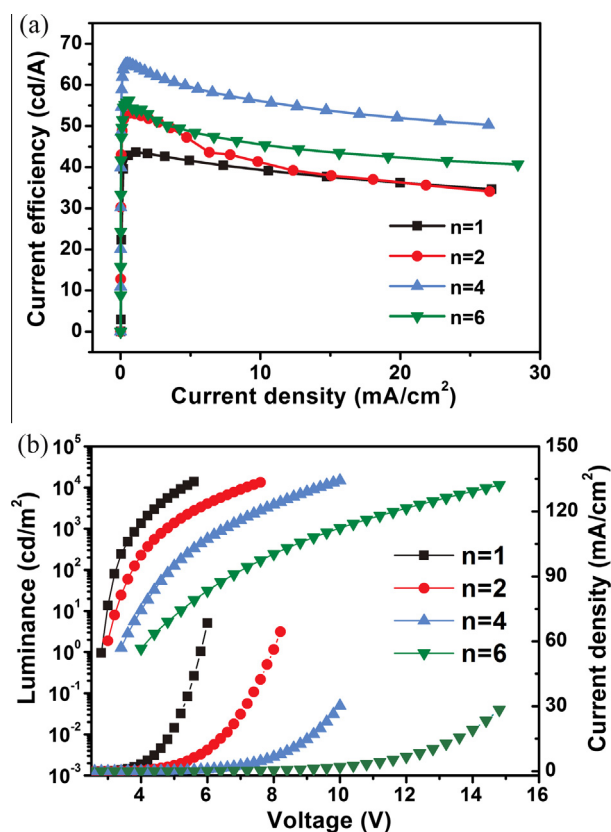


Fig. 4. (a) V–I–B curves of devices employing TCTA PBL with different EML numbers; (b) CE of the devices.

Table 1

Summary of the EL performances of YOLEDs with different EML numbers using TCTA as PBL. The total CE is obtained by applying a factor of 1.7 to the forward-viewing CE [9].

	$n = 1$	$n = 2$	$n = 4$	$n = 6$
Turn on voltage (at 1 cd/m ²)	2.8 V	3.0 V	3.4 V	3.8 V
Voltage at 1000 cd/m ²	3.8 V	4.8 V	6.6 V	9.8 V
Total CE	74.1 cd/A at 485.5 cd/m ²	85.0 cd/A at 369 cd/m ²	110.85 cd/A at 335.8 cd/m ²	95.54 cd/A at 339.6 cd/m ²

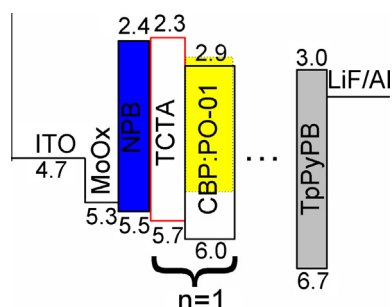


Fig. 5. The energy-level diagram of the devices with M-EMLs ($n = 1, 2, 4, 6$) structure.

at the TAPC/EML interface due to the inherent higher LUMO level and low electron mobility of TAPC. In this case, the superfluous electrons injected from the cathode in device T_3 are not able to be transported to the subsequent layers and have to be accumulated at the TAPC/EML interface which finally results in a severe triplet-polaron annihilation inevitably [20] and therefore a low CE. One can also note that device T_4 owns extremely low peak CE (4.0 cd/A at 0.5 mA/m²) as compared to device T_2 (see Fig. 3), suggesting negligible electron density in the EML, again a clear signifier of the severe electron accumulation in the TAPC/CBP interface in device T_3 , which is consistent with the above conclusion. Having taken into consideration of the distinguished CE of device T_1 , along with above analysis, we conclude herein the main contribution of this work, i.e., the selective rules for PBL: to harvest high CE, the PBL selected in this study not only needs to have good hole mobility (as aforementioned), but also should achieve a better energy level alignment with the EML and avoid a fatal low electron mobility, so that more balanced electron distribution can be expected in the EMLs.

The excellent performance of the device T_1 and T_2 inspired us to further boost the CE of devices utilizing TCTA as PBL. Fig. 4 (a) and (b) plots the V–I–B and CE curves of the devices with various EML numbers ($n = 1, 2, 4, 6$) using TCTA PBL, respectively. Table 1 summarizes the performances of devices and the schematic energy-level diagrams are shown in Fig. 5. A remarkable total CE of 110.85 cd/A (corresponding to a forward-viewing CE of 65.21 cd/A)

is finally achieved at 335.8 cd/m² in the device with 4 EMLs, which is the best values based on PO-01 emission reported in literatures so far [21,22]. The total CE can be maintained as high as 90 cd/A at 10,000 cd/m² (Fig. 4b), indicating balanced carrier distribution and effective exciton confinement are realized under different bias.

4. Conclusions

In summary, high CE is obtained in PO-01 based YOLED utilizing TCTA as PBL. The device can efficiently facilitate the carrier transport as well as regulate exciton distribution within the EML. The PBL in the device should (i) be a hole transport preferable material to suppress the emission of the HTL; and (ii) realize a better energy level alignment with the EML. It is anticipated this work should be a significant reference for further research toward high CE YOLEDs based on M-EMLs structures.

Acknowledgments

We acknowledge funding for this research from the National Key Basic Research and Development Program (973) of China under Grant No. 2010CB327701, and the National Natural Science Foundation of China (Grant No. 61275033).

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