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journal homepage: www.elsevier.com/locate/optlastec

Full length article

Remarkable improvement in electroluminescence benefited from appropriate electron injection and transporting in ultraviolet organic light-emitting diode

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ARTICLE INFO

Article history:

Received 23 October 2015

Received in revised form

20 February 2016

Accepted 10 March 2016

Available online 17 March 2016

Keywords:

OLED

Ultraviolet emission

Electron injection and transport

Carrier balance

Impedance spectroscopy

ABSTRACT

Suitable thickness of LiF and 4,7-diphenyl-1, 10-phenanthroline with slightly weakened electron injection and transporting is proposed to match the intractable hole injection capacity in ultraviolet organic light-emitting diode (UV OLED). By using this strategy, the device performance is remarkably improved. With 4,4'-bis(carbazol-9-yl)biphenyl (CBP) and 3-(4-biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ) as emitters, the UV OLED shows maximum radiance of 5.8 mW/cm² and external quantum efficiency of 2.1% with emission peak of ~380 nm predominantly from TAZ and noticeable shoulder emission of ~410 nm from CBP. The retarded electron injection and transporting contribute to optimizing hole–electron recombination zone and balance within the emitting layers, which accounts for the improved electroluminescent intensity. The detailed mechanism is further clarified with impedance spectroscopy.

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1. Introduction

Ultraviolet organic light-emitting diode (UV OLED) combining flexibility and low-cost manufacturing makes it promising application to high density information storage and exciting light source. In documented works, UV OLED is hugely inferior to the conventional visible counterpart in terms of radiance (luminance) and efficiency [1–6]. One of major problems of counteracting performance improvement is the mismatch of energy level between the work function of indium-tin-oxide (ITO) anode and the highest occupied molecular orbital (HOMO) level of organic emitting layer, which results in rather high hole-injection barrier and serious hole–electron imbalance [6–8]. Fortunately, interfacial engineering paves the way for optimizing carrier balance and thus enhancing electroluminescent (EL) intensity of UV emitters. Wide energy-gap organics of 4,4'-bis(carbazol-9-yl)biphenyl (CBP) and 3-(4-biphenyl)-4-phenyl-5-tert-butylphenyl-1,2,4-triazole (TAZ), behaving high mobility and bipolar transporting, are considered as

the ideal candidates for UV emitters and/or host materials in OLEDs [5,9]. Yu et al. constructed UV OLED with radiance of 0.013 mW/cm² by using Ce-doped CBP as emitting layer [10]. Enhanced radiance of 1.2 mW/cm² and external quantum efficiency (EQE) of 1.25% is demonstrated in CBP-based UV OLED [8]. In our early works, the radiance of CBP-based UV OLED is enhanced to 2.2 mW/cm² with EQE of 0.72% by using graded hole-injection and -transport structure [3]. Shinar et al. demonstrated CBP-based UV OLED with emission peak of 385 nm by using microcavity structure [11]. They also demonstrated adjustable UV/blue emission of 373–469 nm from PVK-doped CBP [12]. TAZ is recently demonstrated to be an efficient UV emitter with emission peak of ~380 nm [5,13], which attracts much attention in contrast to the CBP emission of ~410 nm. However, TAZ features rather deep HOMO level of 6.6 eV, which extremely challenges hole injection capacity. We keep in mind that carrier balance plays a key role in governing device performance. In this study, in contrary to the conventional idea of enhancing hole injection capacity, we circumvent the problem of carrier balance by engineering electron injection and transporting characteristics. Remarkably improved device performance is readily realized by adjusting the thickness

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of electron injection layer LiF and electron transport layer 4,7-diphenyl-1,10-phenanthroline (BPhen). Using both CBP and TAZ as emitting layers, enhanced radiance of 5.8 mW/cm^2 and EQE of 2.1% have been demonstrated with appropriate thickness of LiF and BPhen. The detailed mechanism is further elucidated with impedance spectroscopy [14,15].

2. Experiments

The UV OLEDs with structure of ITO/MoO₃ (3 nm)/CBP (20 nm)/TAZ (15 nm)/BPhen (y nm)/LiF (x nm)/Al (200 nm) were fabricated in a multi-source deposition chamber under a vacuum of 4×10^{-4} Pa. MoO₃ is served for promoting hole injection capacity from ITO anode to adjacent organic layer. CBP is served as hole transport and emitting layer. TAZ is served as electron transport and emitting layer. BPhen ($y=20, 80$ nm) is served as electron transport layer as well as compartment between the emitting layer TAZ and cathode Al with a purpose of adjusting electron transporting and reducing non-radiative recombination [16]. LiF ($x=1, 2.5, 4$ nm) is selected as electron injection layer for engineering electron injection capacity from Al cathode to adjacent BPhen layer. Fig. 1 shows the schematic structure of studied UV OLEDs and the molecular structures of organic materials used. The layer thickness and deposition rate were monitored and controlled in situ with an oscillating quartz thickness monitor.

The photoluminescence (PL) spectra were measured with a Hitachi F4500 fluorescence spectrophotometer. The EL and current density versus voltage (J - V) characteristics were measured with a 7IGF10 Spectra Scan and Keithley 2400 Source Meter. The impedance spectroscopy measurements were carried out with amplitude of ac signals of 50 mV by using a computer controlled programmable Agilent 4294A Precision Impedance Analyzer.

3. Results and discussion

The effect of LiF thickness on device performance is initially performed, and a series of devices of ITO/MoO₃ (3 nm)/CBP (20 nm)/TAZ (15 nm)/BPhen (20 nm)/LiF (x nm)/Al (200 nm) with $x=1$ (Device A), $x=2.5$ (Device B) and $x=4$ (Device C) are fabricated. Fig. 2 shows the radiance, EQE and J - V characteristics of device A–C. One sees that enhanced radiance is observed when the LiF interlayer reaches a suitable thickness of 2.5 nm (Device B). The maximum radiance of Device B reaches 2.8 mW/cm^2 (@ 325 mA/cm^2 and 8 V). In comparison, the corresponding Device A with conventional 1 nm LiF exhibits a maximum radiance of only 2.0 mW/cm^2 (@ 405 mA/cm^2 and 8 V). It is obvious that suitable-

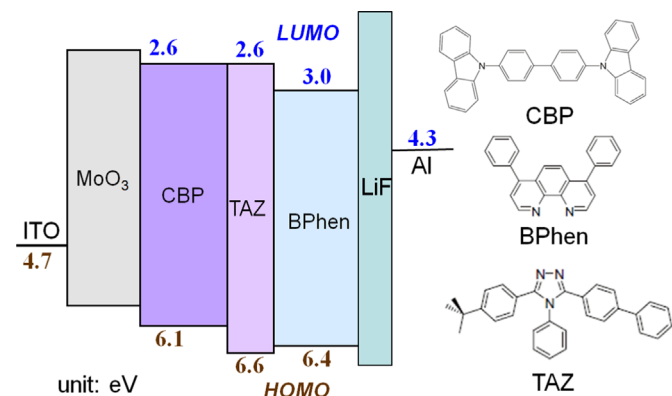


Fig. 1. Schematic energy level of device structure and the molecular structures of organic materials used.

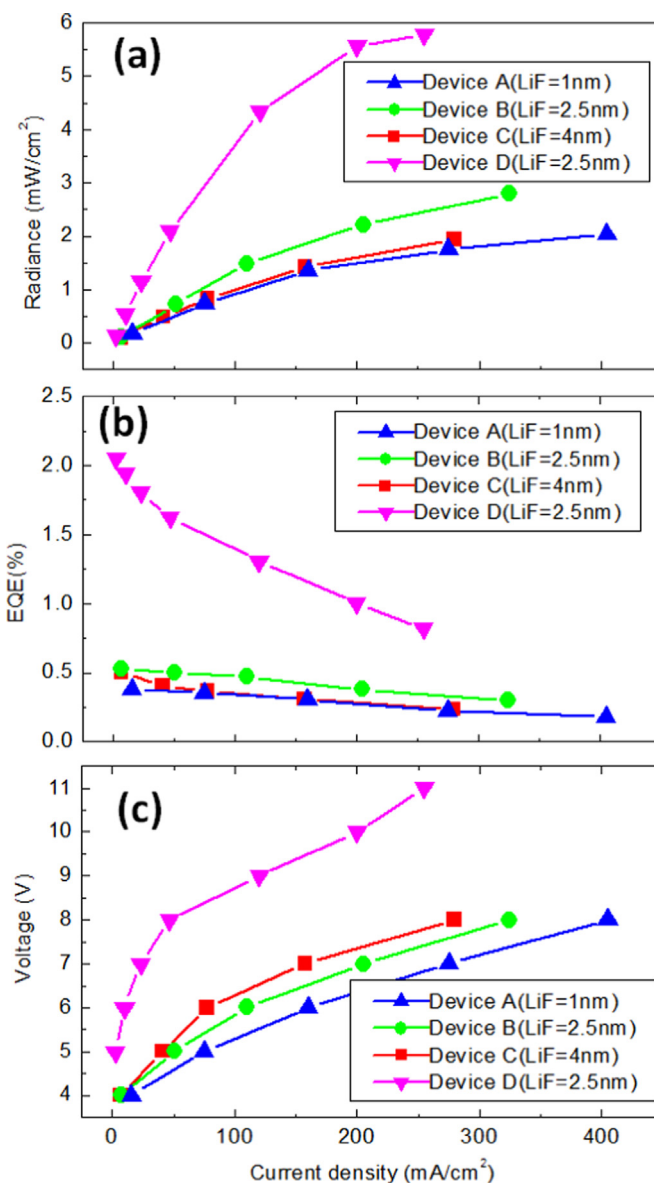


Fig. 2. The (a) radiance, (b) EQE and (c) J - V characteristics of Device A (BPhen/LiF=20/1 nm), Device B (BPhen/LiF=20/2.5 nm), Device C (BPhen/LiF=20/4 nm) and Device D (BPhen/LiF=80/2.5 nm).

thickness LiF improves carrier balance within the emitting layers of CBP and TAZ and thus promoting device performance. Device B also behaves enhanced EQE of 0.53% @ 7.2 mA/cm^2 , which has been enhanced by $\sim 40\%$ in comparison with Device A (0.38% @ 15.5 mA/cm^2).

The EL spectra of Device A–C and PL spectra of CBP and TAZ (thin films) are shown in Fig. 3. It can be seen that the EL spectra of these studied devices are composed of a peak emission of ~ 410 nm and a noticeable shoulder emission of ~ 380 nm. Both CBP and TAZ serve as UV emitters. The emission of ~ 410 nm is resulted from CBP while that of ~ 380 nm is predominantly emitted from TAZ [3,5]. This can also be verified by the PL characteristics of CBP and TAZ, as shown in Fig. 3(b). The difference in EL and PL emission peak may originate from crystallinity of CBP (or TAZ) thin film formed by thermal evaporation [13]. As shown in the J - V curves in Fig. 2(c), thick LiF interlayer requires high voltage. This accounts for the fact that thick LiF counteracts electron injection and benefits hole–electron recombination zone within TAZ layer, which strengthens the shoulder emission of ~ 380 nm. In

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