



High-efficiency hybrid organic–inorganic light-emitting devices



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ABSTRACT

We report efficient red, orange, green and blue organic–inorganic light emitting devices using light emitting polymers and polyethylenimine ethoxylated (PEIE) interlayer with the respective luminance efficiency of 1.3, 2.7, 10 and 4.1 cd A⁻¹, which is comparable to that of the analogous conventional devices using a low work-function metal cathode. This is enabled by the enhanced electron injection due to the effective reduction of the ZnO work-function by PEIE, as well as hole/exciton-blocking function of PEIE layer. Due to the benign compatibility between PEIE and the neighboring organic layer, the novel phosphorescent organic–inorganic devices using solution-processed small molecule emissive layer show the maximum luminance efficiency of 87.6 cd A⁻¹ and external quantum efficiency of 20.9% at 1000 cd m⁻².

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1. Introduction

Organic light emitting diodes (OLEDs) have attracted broad attention from academia and industry due to their potential applications in flat-panel displays, solid-state lighting and laser diodes [1,2]. The basic device structure of OLEDs consists of organic functional layers sandwiched between two electrodes, which have relatively high and low work-function (WF) to inject holes and electrons into the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of organic materials. After two decades of development, they are now the most advanced technology in the field of organic electronics. However, the storage and operating stability of devices warrant further improvement. In particular, low WF metals such as calcium and barium are commonly used to facilitate electron injection, which are very reactive and sensitive towards ambient oxygen and moisture and therefore need to be rigorously encapsulated [3]. In order

to address this problem, air-stable metal oxides such as TiO₂ [4], ZnO [5–7], ZrO₂ [8], MgO and HfO₂ [9] have been investigated as electron-injection layers, leading to the development of hybrid organic–inorganic light emitting devices. For some devices, metal oxides modified indium–tin–oxide (ITO) and high WF metals are employed as the electron injection and hole injection contacts. However, electron injection in such devices is often limited because of the large offset between the conduction band minimum of ZnO at ca. –3.6 eV [10] and the LUMO level of organic semiconductors typically ranging from –2 to –3 eV. The ZrO₂ electron-injection layer with the conduction band minimum of ca. –3.0 eV was used to enhance electron injection into red, green and blue light emitting polymers (LEPs), enabling the generation of electroluminescence (EL) spanning the visible light region [8]. Nevertheless, the use of wide band-gap metal oxides such as ZrO₂ significantly increases the drive voltage [11] and the luminance efficiencies of red, green and blue devices need to be improved. Various electron injection materials including self-assembled dipolar materials [12,13], poly-electrolytes [14,15], alkali metal compounds such as cesium carbonate (Cs₂CO₃) [5,16], ion liquid [17] and carbon nanotube [18] have been utilized to improve electron

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injection. Among above mentioned electron injection materials, alkali metal compounds are commonly adopted. Indeed, incorporation of a Cs_2CO_3 interlayer has been shown to enhance the current of electron-only devices by ca. one order of magnitude, leading to high-efficiency organic–inorganic light emitting devices with the maximum external quantum efficiency (EQE) of 7.3% [16]. However, there is a large difference between the surface energy of crystalline and highly polar Cs_2CO_3 and organic materials [19], resulting in poor wetting of the overlying organic layer and non-conformal interface, which are prone to induce leakage current and may adversely affect the luminance efficiency and stability of light emitting devices. Crystallization of phosphorescent composite emissive layer (EML) when deposited on top of Cs_2CO_3 layer has been reported by Bolink et al. [20]. In addition, Cs_2CO_3 is highly hygroscopic and difficult to handle under common laboratory conditions. Furthermore, different treatments of Cs_2CO_3 lead to variations in device properties and interpretation of device physics [11]. Polyelectrolyte materials have been used as interfacial modifiers to improve the contact between metal oxides and organic layers, but the reported polyelectrolyte electron injection materials for hybrid organic–inorganic light emitting devices [14,15], which have a conjugated main chain and therefore possess relatively low triplet energy, may relax the confinement of triplet excited states formed inside the emission zone [11]. Phosphorescent organic–inorganic devices using a Cs_2CO_3 doped ZnO electron injection layer for suppression of EML crystallization and a tris(2-phenylpyridine)iridium ($(\text{Ir}(\text{ppy})_3)$) derivative as phosphorescent emitter show the maximum luminance efficiency of 15 cd A^{-1} [20], which is inspiring as the use of phosphorescent emitters would significantly enhance the luminance efficiency due to the effective harvest of triplet excited states for light emission. To improve the efficiency of phosphorescent organic–inorganic light emitting devices, electron injection materials which possess high electron injection and hole/exciton blocking capability and benign compatibility with phosphorescent emitter-containing EML are required. We note that insulating polymers such as poly(ethylene oxide) (PEO) [21,22], poly(ethylene glycol) (PEG) [23] and polyethylenimine ethoxylated (PEIE) [24,25] can improve electron injection/extraction in various organic electronic devices. These polymers possess the following advantages: (1) they are air-stable and can be processed from aqueous or alcohol solution, forming homogeneous thin films; (2) they work as the compatibilizer for metal oxide layers to keep the morphology integrity of the adjoining EML; (3) they possess large excited state energy to confine excitons inside the EML; (4) decreased chemical doping of the neighboring organic layer can be beneficial for device stability, which make them suitable as electron-injection and hole/exciton-blocking layers in combination with metal oxide layers. We report inverted light emitting devices using PEIE modified ITO as electron injection contact in the previous work [26]. Herein, we show that combination of PEIE and ZnO electron injection layer leads to red, green and blue organic–inorganic light emitting devices with improved luminance efficiencies compared to the devices using PEIE electron injection layer.

Furthermore, the novel phosphorescent organic–inorganic light emitting devices using solution-processed small molecule EML exhibit the maximum EQE of ca. 22.6%.

2. Experimental

ITO substrates were cleaned sequentially using detergent solution, deionized water, acetone and ethanol in an ultrasonic bath, and were treated with UV–ozone for 10 min. ZnO layers were prepared by spin-coating 2-methoxyethanol solution of zinc acetate dihydrate onto ITO substrates and tempering the resultant films at 300°C for 1 h. Polyethylenimine 80% ethoxylated (PEIE) was applied on top of ZnO layer. Light emitting layers including poly[2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylene vinylene] (MEH-PPV), fluorene–amine copolymer (PFA), poly(phenylene vinylene) copolymer super yellow (SY), poly[9,9-dioctylfluorene-co-(bis-thienylene)benzothiadiazole] (PF-TBT) and 4,4',4''-tri(N-carbazolyl)triphenylamine (TCTA):1,3-bis[(4-tert-butylphenyl)-1,3,4-oxadiazolyl]phenylene (OXD-7)/a 4,4-bis-[2-(4,6-diphenyl-1,3,5-triazinyl)]-1,1-biphenyl derivative (DATE):fac-tris(2-3-(p-xylyl)phenyl)pyridine iridium (TEG) blend were deposited from the respective chlorobenzene solution under controlled nitrogen atmosphere. The thickness of MEH-PPV, PFA, SY and PF-TBT layer was 100 nm, while the thickness of TCTA:OXD-7/DATE:TEG layer was either 90 or 40 nm, as determined by a Dektak surface stylus profiler. Then the samples were transferred to a high-vacuum chamber integrated inside a glove-box without exposure to air. A 50 or 100 nm 1,1-bis((di-4-tolylamino)phenyl)cyclohexane (TAPC) layer was thermally evaporated for phosphorescent devices. Light emitting devices were completed by the deposition of the common top contact with the structure of MoO_3 (10 nm) and Al (150 nm). For electron-only devices, the CsF (1 nm)/Al (150 nm) top contact was thermally deposited onto a 300 nm MEH-PPV layer. For hole-only devices, a 50 nm poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) layer was applied on top of ITO substrate, which was tempered at 170°C for 10 min under ambient conditions. A 300 nm MEH-PPV layer was deposited on top of PEDOT:PSS layer, followed by thermal evaporation of the 10 nm MoO_3 and 150 nm Al. The thickness and deposition rate for evaporated layers were measured by a quartz crystal oscillator. And the deposition rate values for TAPC, MoO_3 , CsF and Al were 0.03, 0.02, 0.01 and 0.3 nm s^{-1} , respectively. The current density–luminance–voltage (J – L – V) characteristics of devices were measured by a programmed Keithley 2400 electrometer and a Konica-Minolta CS-100A chroma-meter. EL spectra of devices were recorded using an Ocean Optics USB4000-UV-VIS spectrometer. UPS and XPS measurements were performed with a Kratos Axis Ultra DLD spectrometer, where He (I) gas discharge lamp (21.2 eV) and Al $K\alpha$ line (1486.8 eV) were used as excitation sources for UPS and XPS.

3. Results and discussion

The schematic of device structure is shown in Fig. 1a. We choose MEH-PPV as the representative LEP to illustrate the function of PEIE layer in light emitting devices. As the

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