



A new approach to efficiency enhancement of polymer light-emitting diodes by deposition of anode buffer layers in the presence of additives

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ABSTRACT

Here we report a new approach for the preparation of anode buffer layer for efficient polymer light-emitting devices (PLEDs) by using glycerol to modify relative low conductivity poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS). This new type of anode buffer layer allows for a 50–90% increase in device performance for green emitting phosphorescent PLEDs in terms of luminous efficiency, and external quantum efficiency, while 90–150% in power efficiency, as compared to devices fabricated using commercially available PEDOT:PSS. The green emitting phosphorescent PLEDs with this modified anode buffer exhibit very high efficiencies, representing a significant step forward to matching and exceeding the efficiencies reported to date with vacuum-deposited small molecular devices. We anticipate that these findings can provide a simple experimental procedure for improvement of PLEDs.

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1. Introduction

Polymer light-emitting diodes (PLEDs) are devices that convert electricity to light as a result of radiative decay in semiconducting organic layers [1]. These devices are of

intense scientific and technological interest due to their potential applications in flat-panel displays and solid-state lighting. However, the low efficiency of PLEDs, compared to vacuum-deposited small molecule based organic light-emitting devices (OLEDs) [2–5], limits their feasibility for commercialization. Balanced charge carriers injection and transport is needed to achieve efficient devices [6]. PLEDs have therefore evolved from simple sandwich prototypes towards multilayer structures, wherein each layer plays a specific function, such as charge carrier injection, transport, unwanted carrier blocking and electron–hole pair recombination zone control [7]. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) is a com-

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mercial product that has been widely used as a hole injecting and transporting material [8–10] due to its electronic, optical and film forming properties [11–13]. Significantly, optical quality PEDOT:PSS thin films can be readily deposited on various substrates by solution processing techniques, such as spin-coating, inkjet printing, and roll-to-roll printing. [8–16].

Substantial efforts have been dedicated to develop PEDOT:PSS films with high electrical conductivity. One of the primary motivations has been that low conductivity leads to lower PLEDs performance since the average electric field across the device is decreased and Joule heating can lead to a decrease in operational lifetime [15,16]. However, high-conductivity is necessary when PEDOT:PSS films are used as the anode itself [15,17,18]. Atop ITO, PEDOT:PSS functions as a buffer layer to remedy electrical shorts and stabilize the work function. Under these conditions it is the out-of-plane resistance that contributes more significantly to the device resistance.

In this communication, we demonstrate that low conductivity PEDOT:PSS in combination with a novel high-conductivity, glycerol-modified PEDOT:PSS layer can be used to fabricate phosphorescent PLEDs with high luminous efficiency (LE), power efficiency (PE) and external quantum efficiency (EQE). This manuscript is organized as follows. The difference in PLEDs performance as function of different commercially available PEDOT:PSS formulations are provided first as a baseline measure. Subsequently, we show that the addition of glycerol as an additive during the spin-coating step leads to large increases in PEDOT:PSS conductivity. Finally, we integrate two different PEDOT:PSS layers with different electrical properties to fabricate PLEDs that displays remarkably improved LE, PE and EQE. These discoveries are anticipated to enable further improvement of the present efficiency of PLEDs and may minimize the efficiency gap relative to vacuum-deposited small molecular devices.

2. Experimental details

A host–guest system, consisting of poly(vinylcarbazole) (PVK) blended with 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) and green emitting iridium complex, Iridium tris(2-(4-tolyl)pyridinato-*N,C*^{2'}) (Ir(mppy)₃), is used as emitting layer. PVK and PBD were purchased from Aldrich and used as received. Ir(mppy)₃ was purchased from America Dyes Sources. The three types of PEDOT:PSS was purchased from H.C. Starck, Inc.

The fabrication of PLEDs followed well-established processes and can be referenced elsewhere [16]. For the preparation of glycerol-modified PEDOT:PSS layer, solution blending consisting of glycerol (1.0 wt%) dispersed in the commercially obtained PEDOT₈₀₀₀ solution was dropped cast at a speed of 2000 rpm on the top of the prepared PEDOT₈₀₀₀. All dispersion were filtered with 0.45 μm PVDF filter before spin-coating. It is important to disperse glycerol by using a high frequency mixer. The obtained PEDOT films were then dried on a hotplate at 160 °C for 10 min. The current density–luminance–voltage (*J*–*L*–*V*) characteristic was measured using a Keithley 236 source-measurement

unit and a calibrated silicon photodiode. The luminance was calibrated by a spectrophotometer (Photo Research, Model: SpectraScan PR-705). The external quantum efficiency of device was collected by measuring the total light output in all directions in an integrating sphere (IS-080, Labsphere).

The conductivity of PEDOT:PSS film at room temperature was measured by the two-point probe or four-point probe method or *I*–*V* curve sweeps in dry glove box filled with N₂ gas using a Keithley 2410 source meter. AFM images were obtained with a Nanoscope IIIa instrument (Digital Instruments), in the tapping mode. A crystal silicon tip, with a resonant frequency of 300–350 kHz and a spring constant of 20–100 N m⁻¹ was used. The scanner was a 10 μm piezo scanner and the scan rate was 1–2 Hz. Off-line image processing and analysis (grain size, roughness) were performed using DI off-line software (Veeco Corp).

3. Results and discussion

Three different commercial PEDOT:PSS versions were used to fabricate phosphorescent PLEDs. The three PEDOT:PSS versions are (together with their abbreviations in this manuscript): BAYTRON P (PEDOT_p), BAYTRON P Al 4083 (PEDOT₄₀₈₃), and BAYTRON P CH 8000 (PEDOT₈₀₀₀); their typical electrical conductivity are ~1 S cm⁻¹, ~10⁻³ S cm⁻¹ and ~10⁻⁵ S cm⁻¹, respectively [19]. The emitting layer (EML) used throughout the studies contains 70 wt% of poly(vinylcarbazole) (PVK), 29 wt% of 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) and 1 wt% of Iridium tris(2-(4-tolyl)pyridinato-*N,C*^{2'}) (Ir(mppy)₃). Fig. 1a presents the resulting current density–luminance–bias (*J*–*L*–*V*) characteristics. One observes that *J* is dependent on the conductivity of the PEDOT:PSS layer. The current density of the PEDOT₈₀₀₀ device is controlled by the high resistance of the buffer layer, as a result the peak luminance of 14369 cd m⁻² is obtained at a current density of 52 mA cm⁻² (see Table 1 for a summary of relevant device characterization), much lower than that of the PEDOT_p or PEDOT₄₀₈₃ device. However, we note that this device exhibits better control over the leakage current in the small voltage region (0–3 V). For example, as shown in Fig. 1a, the leakage current before turn-on is ~10⁻⁵ mA cm⁻², at least 2–3 orders of magnitude lower than those of PEDOT_p and PEDOT₄₀₈₃ devices.

The luminous efficiency–current density (LE–PE–*J*) characteristics of the PLEDs described above are shown in Fig. 1b. Foremost, one can observe a peak LE of 86 cd A⁻¹ for the PEDOT₈₀₀₀ device at 0.15 mA cm⁻² (6.0 V), which corresponds to an EQE of 26%. The peak PE (52 lm W⁻¹) was obtained at 0.030 mA cm⁻² (5.1 V). At a high forward view luminance of 1000 cd m⁻², the efficiency slightly decreased to 77 cd A⁻¹ (corresponding to an EQE of 23%) and PE still retained as high as 30 lm W⁻¹. These performance parameters are superior relative to those observed with PEDOT_p or PEDOT₄₀₈₃ (see Table 1) and are consistent with the fact that the peak LE and EQE occur in the small current density region [20]. We also note that the efficiencies are competitive with, and even exceed, those of vacuum-deposited OLEDs reported to date [2–5]. In contrast, a peak

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