High-performance alternating current field-induced chromatic-stable white polymer electroluminescent devices employing a down-conversion layer

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ABSTRACT

In this work, a high-performance alternating current (AC) filed-induced chromatic-stable white polymer electroluminescence (WFIPEL) device was fabricated by combining a fluorophor Poly(9,9-dioctylfluorene) (PFO)-based blue device with a yellow down-conversion layer (YAG:Ce). A maximum luminance of this down-conversion FIPEL device achieves 3230 cd m^{-2}, which is 1.41 times higher than the device without the down-conversion layer. A maximum current efficiency and power efficiency of the down-conversion WFIPEL device reach 19.7 cd A^{-1} at 3050 cd m^{-2} and 5.37 lm W^{-1} at 2310 cd m^{-2} respectively. To the best of our knowledge, the power efficiency is one of the highest reports for the WFIPEL up to now. Moreover, Commison Internationale de L'Eclairage (CIE) coordinates of (0.28, 0.30) is obtained by adjusting the thickness of the down-conversion layer to 30 μm and it is kept stable over the entire AC-driven voltage range. We believe that this AC-driven, down-conversion, WFIPEL device may offer an easy way towards future flat and flexible lighting sources.

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1. Introduction

Alternating current (AC) field-induced electroluminescent (EL) devices have recently gained considerable attention as a potential alternative to standard organic light-emitting diodes (OLEDs) [1–16]. These devices are essentially a light-emitting capacitor with an active layer and dielectric layers inserted between a metal electrode and a transparent conducting electrode. This structure gives a noticeable advantage that the dielectric layer can effectively prevent electro-chemical reactions between the organic layer and the electrodes, protecting the device from degradation caused by external moisture and oxygen in air [5]. Besides, due to frequent reversal of AC operation, charge accumulation in devices can be effectively avoided [7]. More importantly, AC EL devices can be easily designed and operated for display and solid-state lighting directly from 110/220 V and 50/60 Hz AC power lines on the wall by impedance matching of the device to the driving source [8,16].

For solid-state lighting sources and full-color display, white light technologies are a major focus among organic-based lighting devices because of its merits of high resolution and possible flexibility for large-scale production. By definition, white emission can be generated by mixing three primary colors (red, green and blue) or complementary colors (blue and yellow or orange) in the emitting layer. In both cases, color response inevitably varies with drive current, operating temperature, and operating time [17]. Down-conversion structures provide a potential alternative which uses a blue emitter incorporating a yellow or orange phosphor layer. From a materials point of view, high-efficiency blue emitters are crucial for device performance. Blue fluorophore Poly(9,9-dioctylfluorene) (PFO) is a commonly used blue emitter [13]. More recently, some new heterocyclic blue or blue-green fluorophores such as pyrazoloquinolines azulenes [18] and azafluoranthenes [19,20] have been reported. The down-conversion device architecture was first introduced to white OLEDs by Duggal et al. [21] By using a blue polymer OLED with inorganic phosphors and organic dyes, they achieved white emission by carefully varying the thickness and the concentration of the organic phosphors. In this method, some photons originating from the blue emitter are absorbed by the phosphors and re-emitted at longer wavelengths to generate white light, and one can avoid issues such as color shift that results from the complexity of having different colors mixed within a single emissive layer.

In this work, we fabricated an AC-driven, down-conversion, chromatic-stable white field-induced polymer electroluminescent

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(WFIPEL) device. The down-conversion WFIPEL device exhibits a maximum luminance of 3230 cd m^{-2}, which is 1.41 times higher than the device without this down-conversion layer. The maximum current efficiency and power efficiency of the down-conversion WFIPEL device reach 19.7 cd A^{-1} at 3050 cd m^{-2} and 5.37 lm W^{-1} at 2310 cd m^{-2} respectively. The power efficiency is one of the highest reports for the WFIPEL up to now [6,13,22]. Furthermore, stable EL spectra with Commison Internationale de L'Eclairage (CIE) coordinates of (0.28, 0.30) were also achieved.

2. Experimental

Fig. 1a shows the schematic WFIPEL device structure with (Device A) and without (Device B) a down-conversion phosphor, and the chemical structures of the molecules are presented in Fig. 1b. The down-conversion material is a yellow emitting phosphor YAG:Ce purchased from GTP. The dielectric layer is comprised of a ferroelectric polymer poly(vinylidenefluoride-co-trifluoroethylene) [P(VDF-TrFE)] with a high dielectric constant [23]. A solution processable thermally polymerizable poly(N,N'-bis(4-butylphenyl)-N,N'-bis(phenyl)benzi-dine) (Poly-TPD) [24] doped tetrafluoro-tetracyano-quinodimethane (F4TCNQ) [25] is used as the hole generation layer (HGL). On top of the HGL is a blue fluorophor PFO [13] as the emitting layer (EML). A high-electron-mobility (1.0×10^{-3} cm^2 V^{-1} s^{-1}) material 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) is used as the electron trans-porting layer (ETL) [26]. Two devices were fabricated on ITO substrates with a sheet resistance of 10 Ω/□. Cleaned ITO was first treated by UV-ozone for 20 min. The dielectric layer was spin-coated using a 15% P(VDF-TrFE) solution in dimethylformamide at 2000 rpm, followed by thermal annealing at 135 °C for 2 h in a nitrogen-filled glove box. For Device A, the down-conversion phosphor YAG:Ce was first mixed with a phosphor vehicle (purchased from DuPont) at 50:50 (w:w) and then spin-coated on the reverse side of the ITO at 2000 rpm and thermally annealed at 120 °C for 2 h. The HGL of Poly-TPD:F4TCNQ in chlorobenzene (12 mg mL^{-1}) was spin-coated at 3000 rpm and then dried at 120 °C for 30 min in nitrogen-filled glove box for both devices. Then the EML layer (PFO in chlorobenzene at 10 mg mL^{-1}) was spin-coated at 2000 rpm. Following the spin-coating step, both devices were transferred into a thermal evaporator in a nitrogen atmosphere. The device fabrications were then completed by thermal evaporation of TmPyPB (0.3 nm/s, 40 nm), LiF (0.05 nm/s, 1 nm) and aluminum (1 nm/s, 100 nm) in succession without breaking vacuum ($\sim 5 \times 10^{-4}$ Pa). Therefore, structures of the devices are phosphor (x μm)/glass/ITO/P(VDF-TrFE)(2.4 μm)/Poly-TPD:F4TCNQ(120 nm)/PFO (150 nm)/TmPyPB(40 nm)/LiF(1 nm)/Al (100 nm) (x=0, 17, 30, 45, 62). The overlap between the ITO and Al electrodes was 3 mm × 3 mm as the active emissive area of the devices.

The devices were driven by AC sinusoidal voltages, which were generated from a 200 MHz function/arbitrary waveform generator (Agilent 33220 A) connected to a Model PZD700A M/S amplifier

(Trek). The voltage and current were measured on an oscilloscope (Tektronix). The excitation spectra were measured using a LS50B luminescence spectrometer. The EL spectra were collected with an ILT950 spectroradiometer (International Light Technologies). The luminance was recorded with an ILT 1400-A photometer (Inter-national Light Technologies), either as a function of AC voltages applied to the devices (at a fixed frequency) or as a function of the frequency of applied AC voltages (keeping the applied AC bias constant). The phase characteristics are measured with a Keithley 595 quasistatic CV meter. Time-resolved AC electroluminescence response is detected using a fast photo diode. All measurements were carried out under ambient conditions at atmospheric pres-sure and room temperature.

3. Results and discussion

Fig. 2a shows the excitation spectra of the down-conversion phosphor and the normalized EL spectrum of the PFO-based blue FIPEL (Device B). The down-conversion phosphor was deposited on a quartz substrate with a thickness of 30 μm. Lights with wavelengths ranging from 400 nm to 500 nm are applied to excite the down-conversion phosphor, and the emission intensities are recorded at different wavelengths. It can be clearly seen that there is a peak appearing at around 460 nm in the excitation spectra of the down-conversion phosphor, which well matches the shoulder peak of the PFO EL spectrum. This can be understood that the down-conversion phosphor could absorb a fraction of photons emitted by the PFO-based blue FIPEL. Then the down-conversion phosphor reemits at a wavelength of 550 nm with strongest int-ensity, corresponding to the yellow range of the visible spectrum, which is just the complementary color of the blue. Therefore, white emission should be achieved by combining the blue emis-sion from PFO and the yellow emission from the down-conversion phosphor. To test this assumption, a device incorporating the PFO-based blue FIPEL with the down-conversion phosphor is fabricated (WFIPEL, Device A). The normalized EL spectra of the light gen-erated from Device A is shown in Fig. 2b. Compared to the EL spectra of Device B, there is a second maximum peak presented at 550 nm in Device A, due to which the white light is generated, consistent with the above-mentioned discussions. We can also see that the normalized EL spectrum of the WFIPEL is remarkably stable over the entire AC-driven voltage range. The CIE coordinates nearly keep the same from (0.28, 0.31) to (0.28, 0.30), the color rendering index (CRI) increases from 77.4 to 79.0, and the corr-elated color temperature (CCT) ranges from 8768 to 9743. Besides, the CIE coordinates of Device A can be adjusted by varying the thickness of the down-conversion phosphor layer for the same exited condition. Note that the chromaticity can be defined by only using x and y. As shown in Fig. 2c, decreasing the thickness of the down-conversion layer from 62 μm to 17 μm, leads to a decrease
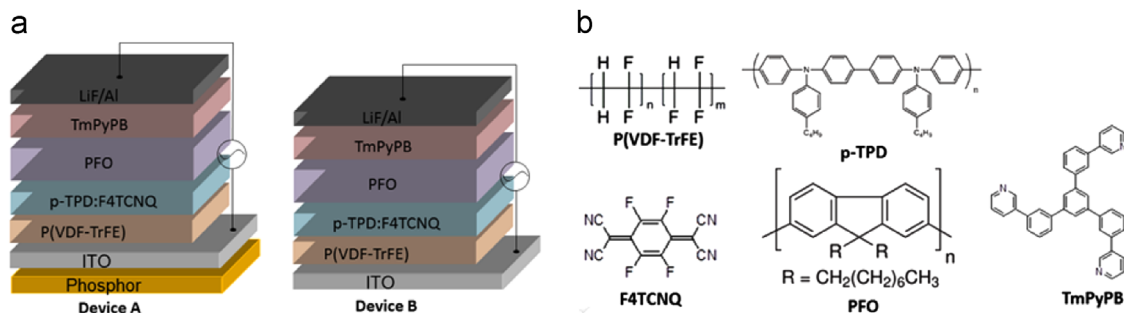

Fig. 1. (a) Schematic structures of the FIPEL devices. The device structures are phosphor (30 μm)/glass/ITO/P(VDF-TrFE) (2.4 μm)/PFO (150 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al (100 nm) for Device A and glass/ITO/P(VDF-TrFE) (2.4 μm)/PFO (150 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al (100 nm) for Device B. (b) The molecular structures used in this work.