



# Alternating current-driven, white field-induced polymer electroluminescent devices with high power efficiency

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## ABSTRACT

A solution-processed, all-phosphor, three-color (i.e., blue, green, and red), alternating current-driven white field-induced polymer electroluminescent device (WFIPEL), with low operational voltage, high luminance, high efficiency, high color-rendering index (CRI), and excellent color-stability, was demonstrated. The devices employed poly(vinylidene fluoride–trifluoroethylene–chlorofluoroethylene) [P(VDF–TrFE–CFE)] dielectric modified by single-walled carbon nanotubes (SWNTs) to further improve the dielectric characteristics, as the insulating layer. This significantly lowers the driving voltage of the device. Moreover, hole-generation layer and electron-transporting layer with high conductivity were used to more efficiently form and confine excitons in the emissive layer. The resulting WFIPEL devices show significant improvements in performance as compared to previous reports. Specifically, the devices exhibit a low turn-on voltage of 10 V, a maximum luminance of 7210 cd m<sup>-2</sup>, a maximum current efficiency and power efficiency of 33.8 cd A<sup>-1</sup> and 10.5 lm W<sup>-1</sup>, and a CRI of 82. The power efficiency is even 10 times higher than the highest previous report (1 lm W<sup>-1</sup>).

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## 1. Introduction

White organic thin-film electroluminescent (TFEL) devices have drawn increasing attention due to their application in displays and general solid-state lighting [1–13]. Up until now, white organic TFEL devices are typically operated with low-power direct current (DC), e.g., white organic light-emitting diodes (OLEDs) [1–3,5,7–9,11–13] and white light-emitting electrochemical cell (LEC) [4,6]. Conceptually, in such devices a threshold voltage (or higher voltages) is applied between a pair of electrodes, and electrons are injected from the cathode while holes are injected from the anode. The carriers meet in the

EML and recombine to give light. Thus, such devices typically use high current densities to achieve high luminance. This results in efficiency roll-off at high luminance caused by triplet-triplet or triplet-charge annihilation in typical phosphorescent devices [14–17]. Moreover, such DC-driven approaches require power converters which introduce power losses, making devices sensitive to dimensional variations that lead to run away currents at imperfections.

TFEL devices from field-activation of organic small molecules or conjugated polymers, driven by an alternating current (AC) mode, may present a potential alternative to DC technologies by providing low-energy consuming solutions [10,18–32]. AC-driven modes can avoid using expensive power converters when they are integrated into AC lighting panels, which avoid power losses and complicated back end electronics as well. This makes devices insensitive to dimensional variations that lead to run away

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currents at imperfections. The AC field-induced EL devices are essentially a light emitting capacitor with an active layer and dielectric layers inserted between a metal electrode and a transparent conducting electrode. There have been several proposed advantages of this approach to organic light generation, including using the dielectric layer to effectively prevent electro-chemical reactions between the organic layers and the electrodes, avoiding quenching charge carriers by electrodes and protecting from degradation due to external moisture and oxygen in the atmosphere. Moreover, due to frequent reversal of the applied voltage, AC-driven EL devices avoids charge accumulation in the device [23], which may reduce the triplet-triplet or triplet-charge annihilation in the devices' operation. Most recently, we found that the usual concentration dependence of emission from the dye is not observed in a hybrid system comprising of a blue fluorophore and a red phosphor dye, due to the unique carrier injection characteristics of the field-induced device. This further suggests that greater color tunability may be found in such devices as compared to typical OLED structures [27].

To realize highly efficient AC EL devices, two major factors have to be taken into account. First, the dielectric constant ( $k$ ) of insulating layers must be high to lower the driven voltage and improve the breakdown voltage [24]. Second, excitons must be efficiently created to increase the light intensity [23]. Recent conceptual advancements have led to many exciting approaches to fabricate AC EL devices. Tsutsui et al. utilized nanoparticles in an attempt to increase charge generation under high electric fields and a bright surface blue fluorescent emission of  $81 \text{ cd m}^{-2}$  at 250 V and 300 kHz was observed [18]. The emission is due to the recombination of holes and electrons that are generated from the nanoparticles embedded in organic layers. Park et al. also developed a high-performance AC EL device with management of charge carriers injection through highly dispersed SWNTs in the fluorescent polymer matrix [22], which emitted green light with a brightness of approximately  $350 \text{ cd m}^{-2}$ . Using inorganic dielectrics with molecular doping charge generation layers in a small molecule emitter, as reported by Leo et al. [23–25], the TFEL approach exhibited a red phosphorescent emission with a luminance up to  $1500 \text{ cd m}^{-2}$  (and  $5000 \text{ cd m}^{-2}$  with optically optimized output coupling). We have also reported a solution-processed device using multi-walled carbon nanotubes (MWNTs) as a charge injection channel and generation source in the polymer matrix. In this device a green phosphorescent emission of  $100 \text{ cd m}^{-2}$  was achieved [26]. Nevertheless, considering future applications, further challenges are inevitably posed, especially in getting white emission. Although a hybrid AC EL device, which featured an emissive layer (EML) composed of a blue polymer and inorganic colloidal quantum dots, exhibited effective color mixing, giving rise to white EL [10], the luminance of this device is just  $620 \text{ cd m}^{-2}$  and the color does not cover the whole visible spectrum. Very recently, an AC-driven white EL device using high frequency color-mixing has been reported with warm-white color coordinates (0.43, 0.44), a color-rendering index (CRI) of 76.5, a maximum

power efficiency of  $1 \text{ lm W}^{-1}$ , and a luminance value of  $1000 \text{ cd m}^{-2}$  [29]. The requirements of low voltage, high luminance, high efficiency, desired CRI, and excellent color-stability in one device must be satisfied simultaneously, but have not yet to be achieved.

In this work, we demonstrate a solution-processed, all-phosphor, three-color (i.e., blue, green, and red), AC-driven white field-induced polymer EL (WFIPEL) device, with low operational voltage, high luminance, high efficiency, high CRI, and excellent color-stability. The device we described is to use a high  $k$  relaxor ferroelectric polymer dielectric, poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) [P(VDF-TrFE-CFE)], blended with SWNTs to further modify the  $k$ , as the insulating layer. This lowers the driving voltage of the device (keeping the same field in the emitter). Moreover, we employ a high conductivity hole generation layer (HGL): poly(3-hexylthiophene) (P3HT) doped tetrafluoro-tetracyano-quinodimethane (F4TCNQ) and a high conductivity electron transport layer (ETL): 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene (TmPyPB) doped lithium carbonate ( $\text{Li}_2\text{CO}_3$ ) to more efficiently form and confine excitons in the EML. The excitons are formed by the recombination of the generated holes from HGL and the injected electrons from ETL. The resulting WFIPEL devices show significant improvements in performance as compared to previous reports. Specifically, the devices show a low turn-on voltage of 10 V, a maximum luminance of  $7210 \text{ cd m}^{-2}$ , a maximum current efficiency and power efficiency of  $33.8 \text{ cd A}^{-1}$  and  $10.5 \text{ lm W}^{-1}$ , and a CRI of 82. To the best of our knowledge, this is one of the highest values in the reported literatures. The power efficiency is even 10 times higher than the highest previous report [29].

## 2. Experimental

### 2.1. Materials

The high- $k$  relaxor ferroelectric terpolymer dielectric, P(VDF-TrFE-CFE) 62.6/29.4/8 mol%, poly( $N,N'$ -bis(4-butylphenyl)- $N,N'$ -bis(phenyl)benzidine) (P-TPD), P3HT and poly ( $N$ -vinylcarbazole) (PVK) were purchased from PiezoTech, American Dye Source, and Sigma-Aldrich, respectively. F4TCNQ, 1,3-bis(2-(4-*tert*-butylphenyl)-1,3,4-oxadiazole-5-yl)benzene (OXD-7), bis(3,5-difluoro-2-(2-pyridyl) phenyl-(2-carboxypyridyl)iridium (Flrpic), fac-tris(2-phenylpyridine)iridium(III) [Ir(ppy)<sub>3</sub>], Bis(2-methyl-dibenzo[*f,h*]quinoxaline) (acetylacetonate) iridium(III) [Ir(MDQ)<sub>2</sub>(acac)], TmPyPB were purchased from Luminescence Technology Corp., Taiwan. All materials were used as received without further purification.

### 2.2. Device fabrication

All WFIPEL devices were fabricated on a glass substrate with a 100-nm-thick pre-coated ITO film, with a sheet resistance of approximately  $10 \Omega/\square$ . First, the substrates were cleaned in an ultrasonic bath with acetone followed by methanol and isopropanol for 30 min each. The ITO substrates were subsequently dried in a vacuum oven for 2 h and treated with UV-ozone for 20 min. The dielectric layers

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