



# Enhancing device efficiencies of solid-state near-infrared light-emitting electrochemical cells by employing a tandem device structure



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## ABSTRACT

Compared to near-infrared (NIR) organic light-emitting devices, solid-state NIR light-emitting electrochemical cells (LECs) could possess several superior advantages such as simple device structure, low operating voltages and balanced carrier injection. However, intrinsically lower luminescent efficiencies of NIR dyes and self-quenching of excitons in neat-film emissive layers limit device efficiencies of NIR LECs. In this work, we demonstrate a tandem device structure to enhance device efficiencies of phosphorescent sensitized fluorescent NIR LECs. The emissive layers, which are composed of a phosphorescent host and a fluorescent guest to harvest both singlet and triplet excitons of host, are connected vertically via a thin transporting layer, rendering multiplied light outputs. Output electroluminescence (EL) spectra of the tandem NIR LECs are shown to change as the thickness of emissive layer varies due to altered microcavity effect. By fitting the output EL spectra to the simulated model concerning microcavity effect, the stabilized recombination zones of the thicker tandem devices are estimated to be located away from the doped layers. Therefore, exciton quenching near doped layers mitigates and longer device lifetimes can be achieved in the thicker tandem devices. The peak external quantum efficiencies obtained in these tandem NIR LECs were up to 2.75%, which is over tripled enhancement as compare to previously reported NIR LECs based on the same NIR dye. These efficiencies are among the highest reported for NIR LECs and confirm that phosphorescent sensitized fluoresce combined with a tandem device structure would be useful for realizing highly efficient NIR LECs.

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## 1. Introduction

Near-infrared (NIR) organic light-emitting devices (OLEDs) would be promising NIR light sources offering advantages of light weight, low power consumption and compatibility with large area and flexible substrates. Therefore, they have received much attention due to their potential applications in telecommunications, displays and

bio-imaging [1]. However, sophisticated multilayer structures and low-work-function cathodes are generally required for NIR OLEDs to optimize device efficiencies, influencing their competitiveness with other solid-state NIR emitting technologies. In contrast with conventional NIR OLEDs, solid-state NIR light-emitting electrochemical cells (LECs) could possess several superior advantages. Generally, LECs require only a single emissive layer, which can be easily processed from solutions, and can conveniently use air-stable electrodes. The emissive layer of LECs contains mobile ions, which can drift toward electrodes under an applied bias. These spatially separated ions

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consequently induce doping (oxidation and reduction) of the emissive materials near the electrodes, i.e. p-type doping near the anode and n-type doping near the cathode [2,3]. The doped regions induce ohmic contacts with the electrodes and facilitate carrier injection, giving low operation voltages and high power efficiencies.

A few literatures about solid-state NIR LECs based on small-molecule cationic transition metal complexes (CTMCs) [4–7] and polymers containing pendant CTMC groups [8] have been reported. Nevertheless, compared with visible light-emitting CTMCs [9–24], deteriorated photoluminescence quantum yields (PLQYs) of NIR light-emitting CTMCs [4,5,7] are generally measured due to the energy gap law, which states that the nonradiative decay rates of CTMCs increase as the energy gaps decrease [11]. Furthermore, self-quenching of excitons in neat films also limits the device efficiency of NIR LECs based on neat films of CTMCs. Therefore, neat-film CTMC-based NIR LECs typically exhibited external quantum efficiencies (EQEs) lower than 0.1% photon/electron [4–8]. To improve device efficiencies of NIR LECs, NIR LECs based on phosphorescent sensitized fluorescence have been reported [25]. In phosphorescent sensitized fluorescence [25–28], both singlet and triplet excitons in the host could be harvested due to effective Förster energy transfer [29] from triplet excitons of the phosphorescent host to singlet excitons of the fluorescent guest. With well developed CTMC hosts possessing good carrier balance, commercially available efficient fluorescent ionic NIR laser dyes [30] can be conveniently utilized as guests to achieve efficient NIR electroluminescence (EL). In addition, self-quenching of excitons could be mitigated in host-guest emissive layers. As a result, the reported phosphorescent sensitized fluorescent NIR LECs exhibited EQEs of ca. 1% [25]. However, these efficiencies are still not enough for practical applications and further improving device efficiencies of NIR LECs would be required.

Recently, we have demonstrated tandem LECs to reach over doubled light outputs and device efficiencies by stacking two single-layered LECs via a thin connecting layer [31]. A similar approach employing a thin metal interlayer to produce tandem white light emitting devices was also reported recently [32]. It would be a simple way to enhance device efficiencies of phosphorescent sensitized fluorescent NIR LECs. In this work, we report tandem phosphorescent sensitized fluorescent NIR LECs based on emissive layers containing a phosphorescent CTMC host doped with a fluorescent ionic NIR dye. To clarify effects of emissive-layer thickness on device performance, tandem NIR LECs with various thicknesses are studied. Output EL spectra of tandem NIR LECs can be modified by adjusting the thicknesses of the emissive layers due to altered microcavity effect. Furthermore, improved device lifetimes are found in thicker devices due to reduced exciton quenching near electrodes. The peak EQE obtained in these tandem NIR LECs is up to 2.75%, which is over triple of that achieved in single-layered NIR LECs based on the same NIR dye [25]. The device efficiencies achieved are among the highest reported for NIR LECs and thus confirm that phosphorescent sensitized fluorescence combined with a tandem device structure would be useful for realizing highly efficient NIR LECs.

## 2. Experiment section

### 2.1. Materials

The host complex (**1**) used in the emissive layer of the NIR LECs was Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub> (where dtb-bpy is 4,4'-ditertbutyl-2,2'-bipyridine) [33]. The ionic NIR dye 3,3'-diethyl-2,2'-oxathiacarbocyanine iodide (DOTCI), which has been reported as an active material in efficient NIR dye lasers [30], was utilized as the guest material doped in the host. Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub> and DOTCI were purchased from Luminescence Technology Corp. and Sigma-Aldrich, respectively, and they were used as received.

### 2.2. Photoluminescent characterization

Photoluminescence (PL) characteristics of DOTCI in ethanol were recorded at room temperature using 10<sup>-5</sup> M solutions. The neat film of complex **1** for PL studies was spin-coated at 3000 rpm onto a quartz substrate using acetonitrile solution with a concentration of 80 mg mL<sup>-1</sup>. The thickness of the spin-coated neat host film was ca. 250 nm, as measured using profilometry. UV-Vis absorption and PL spectra were obtained with a Princeton Instruments Acton 2150 spectrophotometer. The exciting wavelengths of DOTCI in ethanol and neat film of complex **1** are 650 and 450 nm, respectively.

### 2.3. LEC device fabrication and characterization

Indium tin oxide (ITO)-coated glass substrates were cleaned and treated with UV/ozone prior to use. A thin poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) layer (30 nm) was spin-coated at 4000 rpm onto the ITO substrate in air and was then baked at 150 °C for 30 min. For the single-layered devices (**S1**, **S2** and **S3**), the emissive layers were then spin-coated at 3000 rpm from the mixed acetonitrile solutions of complex **1** and DOTCI (weight ratio of complex **1** and DOTCI = 99:1). The concentrations of the solutions used for spin coating of the emissive layers of devices **S1**, **S2** and **S3** are 80, 150 and 220 mg mL<sup>-1</sup>, respectively. The thicknesses of the emissive layers for devices **S1**, **S2** and **S3** are 250, 440 and 660 nm, respectively. For the tandem devices, the lower emissive layers were first spin-coated at 3000 rpm from the mixed acetonitrile solutions of complex **1** and DOTCI (weight ratio of complex **1** and DOTCI = 99:1) with concentrations of 80, 150 and 220 mg mL<sup>-1</sup> for devices **T1**, **T2** and **T3**, respectively. The thicknesses of the lower emissive layers for devices **T1**, **T2** and **T3** are 250, 440 and 660 nm, respectively. The connecting PEDOT:PSS layers were then spin-coated onto the lower emissive layers at 3500 rpm. The thickness of the connecting PEDOT:PSS layers is ca. 55 nm. Finally, the upper emissive layers were spin-coated by the same procedures used for spin-coating of the lower emissive layers. After spin coating, the samples were then baked at 70 °C for 10 h in a nitrogen glove box, followed by thermal evaporation of a 100-nm Ag top contact in a vacuum chamber (~10<sup>-6</sup> torr). Thicknesses of thin films were measured by ellipsometry. The electrical and emission

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