



# Extracting evolution of recombination zone position in sandwiched solid-state light-emitting electrochemical cells by employing microcavity effect



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## ABSTRACT

Techniques of probing for time-dependent evolution of recombination zone position in sandwiched light-emitting electrochemical cells (LECs) would be highly desired since they can provide direct experimental evidence to confirm altered carrier balance when device parameters are adjusted. However, direct imaging of recombination zones in thin emissive layers of sandwiched LECs could not be obtained easily. In this work, we propose an alternative way to extract evolution of recombination zone position in sandwiched LECs by utilizing microcavity effect. Recombination zone positions can be estimated by fitting the measured electroluminescence spectra to simulated output spectra based on microcavity effect and properly adjusted emissive zone positions. With this tool, effects of modified carrier transport and carrier injection on performance of LECs are studied and significantly altered carrier balance can be measured, revealing that microcavity effect is useful in tracing evolution of recombination zone position in sandwiched LECs.

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## 1. Introduction

Recently, organic light-emitting diodes (OLEDs) have attracted intense attention due to their potential applications in flat-panel displays and solid-state lighting [1,2]. Compared with conventional OLEDs, solid-state light-emitting electrochemical cells (LECs) [3,4] possess several promising advantages. LECs possess electrochemically doped regions, i.e. p-type doping near the anode and n-type doping near the cathode, which are induced by spatially separated ions under a bias. Such doped regions significantly reduce carrier injection barriers at electrodes, giving balanced carrier injection, low operating voltages, and consequently high power efficiencies. As a result, LECs generally require only a single emissive layer, which can be easily deposited by solution processes and can conveniently utilize air-stable electrodes. The emissive materials

of LECs can be roughly divided into two categories: fluorescent polymers [3] and phosphorescent cationic transition metal complexes (CTMCs) [5]. Compared with polymer LECs, LECs based on CTMCs show several further advantages and have drawn much research interests in recent years [5–34]. In such devices, no ion-conducting material is needed since these CTMCs are intrinsically ionic. Furthermore, higher electroluminescent (EL) efficiencies are expected due to the phosphorescent nature of CTMCs. Green [18] and white LECs [24,34] based on Ir (III) complexes have been shown to exhibit high device efficiencies up to 40 and 15 lm/W, respectively.

To realize highly efficient LECs, spatial control of the recombination zone for reducing exciton quenching in close proximity to electrodes [35,36] would be an important issue. However, it is difficult to directly observe the recombination zone in sandwiched LECs due to their thin emissive layers (<1 μm). A feasible alternative way is direct optical probing of the recombination zone in planar LECs with much larger interelectrode gaps (up to mm) [37–43]. With this approach, several important issues affecting

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device performance of LECs such as cationic effects of salts [39], work functions of electrodes [41], doping processes [38,40] and recombination zone positions [42,43] were studied with clear experimental evidence. Nevertheless, these experiments were performed on planar devices using interdigitated electrodes with spacings much larger than the interelectrode spacings of sandwiched devices and the electric fields would be significantly different in magnitudes for planar and sandwiched LECs. Since ion mobility, carrier injection efficiency and carrier mobility, which affect the recombination zone position, would be field dependent, measured EL characteristics of planar LECs would not necessarily match those of sandwiched LECs. To study device physics and to further improve device performance of sandwiched LECs based on CTMCs, for which promising device efficiencies have been achieved [11,13,15,18,20,24,26,34], feasible techniques to acquire evolution of recombination zone position of sandwiched LECs under driving are highly desired. In this work, we propose a novel technique to dynamically probe recombination zone position of sandwiched LECs by utilizing microcavity effect. Microcavity structures of sandwiched LEC devices modify wavelength dependent optical outcoupling efficiencies and thus lead to tailored EL spectra when the recombination zone is moving. Hence, the recombination zone positions of sandwiched LECs can be estimated by fitting measured EL spectra to simulated EL spectra with proper emitting zone positions. With this technique, effects of carrier trapping [26,29] and carrier injection efficiency [33] on carrier balance of sandwiched LECs can be studied with clear experimental evidence. It would be useful in optimizing device efficiencies of sandwiched LECs.

## 2. Experiment

### 2.1. Materials

The emissive complex used in this study is Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub> (where dtb-bpy is 4,4'-ditertbutyl-2,2'-bipyridine) [6]. The hole transporting material *N,N'*-dicarbazolyl-3,5-benzene (mCP) with a high ionization potential was utilized to impede hole injection into Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub> emissive layer. Both Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub> and mCP were purchased from Luminescence Technology Corp. and were used as received. The low-gap cationic fluorescent near-infrared (NIR) laser dye 3,3'-diethylthiatricarbocyanine iodide (DTTCI) [25] was utilized as the carrier trapper in Ru(dtb-bpy)<sub>3</sub>(PF<sub>6</sub>)<sub>2</sub> emissive layer. DTTCI was purchased from Sigma–Aldrich Co. and was used as received.

### 2.2. LEC device fabrication and characterization

Indium tin oxide (ITO)-coated glass substrates were cleaned and treated with UV/ozone prior to use. A poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) layer was spin-coated at 4000 rpm onto the ITO substrate in air and baked at 150 °C for 30 min. For **Device III**, an mCP layer (~20 nm) was spin-coated at 5000 rpm from chlorobenzene solutions on the PEDOT:PSS

layer under ambient conditions and baked at 60 °C for 6 h in a nitrogen glove box (oxygen and moisture levels below 1 ppm) while this step was skipped for **Devices I and II**. The emissive layers (~450 nm, as measured by profilometry) were then spin-coated at 3000 rpm from the acetonitrile solutions of complex **1** (**Devices I and III**) and complex **1** containing 0.1 wt.% DTTCI (**Device II**) under ambient conditions. The concentration of the solutions used for spin coating of the emissive layers is 250 mg/mL. 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). The electrical and emission characteristics of LEC devices were measured using a source-measurement unit and a Si photodiode calibrated with the Photo Research PR-650 spectroradiometer. All device measurements were performed under a constant bias voltage (2.5 V) in a nitrogen glove box. The EL spectra were taken with a calibrated CCD spectrograph.

## 3. Results and discussions

Since the thickness of the organic layer of the LECs (450 nm) is comparable to the visible optical wavelength and a highly reflective metal (Ag) is used as the cathode, the emission properties of the emissive layer can be modified in such a microcavity structure, which alters the optical mode density within it and spectrally redistributes the EL spectrum. The output EL spectrum of a bottom emitting OLED device can be calculated approximately by using the following equation [44]:

$$|E_{ext}(\lambda)|^2 = \frac{T_2 \frac{1}{N} \sum_{i=1}^N \left[ 1 + R_1 + 2\sqrt{R_1} \cos\left(\frac{4\pi z_i}{\lambda} + \varphi_1\right) \right]}{1 + R_1 R_2 - 2\sqrt{R_1 R_2} \cos\left(\frac{4\pi L}{\lambda} + \varphi_1 + \varphi_2\right)} \times |E_{int}(\lambda)|^2$$

where  $R_1$  and  $R_2$  are the reflectance from the cathode and from the glass substrate, respectively,  $\varphi_1$  and  $\varphi_2$  are the phase changes on reflection from the cathode and from the glass substrate, respectively,  $T_2$  is the transmittance from the glass substrate,  $L$  is the total optical thickness of the cavity layers,  $|E_{int}(\lambda)|^2$  is the emission spectrum of the organic materials without alternation of the microcavity effect,  $|E_{ext}(\lambda)|^2$  is the output emission spectrum from the glass substrate,  $z_i$  is the optical distance between the emitting sublayer  $i$  and the cathode. The emitting layer is divided into  $N$  sublayers and their contributions are summed up. Since the width of p-n junction estimated by capacitance measurements when p- and n-type layers were fully established was shown to be ca. 10% of the thickness of the active layer of LECs [45], the emitting layer width should not broader than tenth of the active layer thickness and thus an emitting layer width of 45 nm and  $N = 45$  were estimated in optical simulation. The PL spectrum of a thin film (450 nm) of complex **1** coated on a quartz substrate was used as the emission spectrum without alternation of the microcavity effect since no highly reflective metal layer is present in this sample. Based on this simulation method, the time-dependent recombination zone location of LECs can be estimated by fitting

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