



Solution-processable tandem solid-state light-emitting electrochemical cells



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ABSTRACT

Compared to organic light-emitting diodes (OLEDs), solid-state light-emitting electrochemical cells (LECs) exhibit simple single-layered structure and low operating voltages due to in situ electrochemical doped layers. However, device efficiencies of LECs are usually lower than those of sophisticatedly designed OLEDs. Furthermore, device efficiencies and lifetimes of LECs degrade significantly as brightness increases. In this work, we demonstrate tandem LECs to obtain nearly doubled light outputs ($\mu\text{W cm}^{-2}$) in comparison with single-layered LECs under similar current densities. Since the output EL emission is modified by microcavity effect of the device structure, the EL spectra of tandem LECs exhibit EL emission peak at ca. 625 nm while the EL spectra of single-layered LECs center at ca. 660 nm. Better spectral overlap between the EL spectrum of tandem LECs and the luminosity function results in further enhanced candela values, rendering a tripled brightness (cd m^{-2}). The device efficiencies can be optimized by adjusting the thickness of the connecting layer between the two emitting units of the tandem devices. The peak external quantum efficiency achieved in tandem LECs is up to 5.83%, which is higher than twice of that obtained in single-layered LECs due to improved carrier balance. When single-layered and tandem LECs are biased under higher voltages to reach similarly higher brightness, tandem LECs show higher device efficiencies and longer lifetimes simultaneously. These results indicate that device efficiencies and lifetimes of LECs can be improved by employing a tandem device structure.

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1. Introduction

Recently, organic light-emitting diodes (OLEDs) have received intense attention due to their potential applications in displays and solid-state lighting [1]. Compared with conventional OLEDs, solid-state light-emitting electrochemical cells (LECs) [2,3] have several superior advantages. In LECs, electrochemically doped regions, i.e. p-type doping near the anode and n-type doping near the cathode, are formed by spatially separated ions under a bias. Electrochemically doped regions significantly reduce carrier injection barriers at electrodes, giving balanced carrier

injection, low operating voltages, and consequently high power efficiencies. Thus, LECs are generally composed of only a single emissive layer, which can be easily deposited by solution processes and can conveniently utilize air-stable electrodes. Emissive materials of LECs can be classified into two categories: fluorescent polymers [2] and phosphorescent cationic transition metal complexes (CTMCs) [4]. As compared to polymer LECs, LECs based on CTMCs exhibit several advantages and have attracted much research interests in recent years [4–26]. In these 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.

Several highly efficient single-layered LECs exhibiting external quantum efficiencies (EQEs) over 10% have been

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reported [9,11,14,19,21,26]. However, device efficiencies achieved in these LECs are still lower than those obtained in sophisticatedly designed multilayered OLEDs [27]. To improve device efficiencies of single-layered devices, tandem device structures would be feasible to obtain multiplied EQEs and current efficiencies (cd A^{-1}) under similar current densities. Tandem OLEDs have been shown to exhibit rather high current efficiencies [28,29]. Tandem OLEDs are generally composed of two or more emissive units connected in series via thin connecting layers. When a bias is applied, electrons and holes are generated within the connecting layer and then are injected into adjoining emissive units. Thus, EL efficiencies of tandem OLEDs can increase linearly with the number of emissive units. In spite of successful realizing enhanced device efficiencies in tandem OLEDs, there has been no report up to now on tandem LECs, which would additionally exhibit advantages such as simpler device structures and lower operating voltages. Instead, lateral cascaded structures were reported for LECs to reach multiplied radiant fluxes and EQEs [30,31]. The main difficulty of preparing multilayered devices by solution processes is that deposition of the new layer often dissolves the previous one. Proper choosing of solvents used in spin coating of each layer to avoid dissolving the previous layer would be critical in fabricating tandem LECs. In addition, efficiencies of LECs are especially sensitive to the current density [32]. To achieve high efficiencies, which are typically measured at low current densities, it is not easy to obtain high luminance values in single-layered cells. High luminance at low current densities can be realized in a tandem structure. In this work, we show the demonstration of tandem LECs by connecting two single-layered LECs vertically. Under similar current densities, doubled light outputs and device efficiencies can be obtained in tandem LECs as compared to single-layered LECs. When biased to achieve relatively higher brightness, both device efficiencies and lifetimes can be improved in tandem LECs. These results confirm that tandem structures are useful in enhancing device efficiencies of LECs and in improving lifetimes of LECs at relatively higher brightness.

2. Experiment section

The emissive complex (**1**) used in this study was $\text{Ru}(\text{dtb-bpy})_3(\text{PF}_6)_2$ (where dtb-bpy is 4,4'-ditertbutyl-2,2'-bipyridine) [5]. $\text{Ru}(\text{dtb-bpy})_3(\text{PF}_6)_2$ was purchased from Luminescence Technology Corp. and was used as received. 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) (Baytron P VP Al 4083) layer (30 nm) was spin-coated at 4000 rpm onto the ITO substrate in air and baked at 150 °C for 30 min. For single-layered devices (**S1**, **S2** and **S3**), the emissive layers were then spin-coated at 3000 rpm from the acetonitrile solutions of complex **1**. The concentrations of the solutions used for spin coating of the emissive layers of devices **S1**, **S2** and **S3** are 80, 190 and 80 mg mL^{-1} , respectively. The thicknesses of the emissive layers for devices **S1**, **S2** and **S3** are 250, 570 and 250 nm, respectively. For tandem devices (**T1**, **T2**, **T3**

and **T4**), the lower emissive layers (250 nm) were first spin-coated at 3000 rpm from the acetonitrile solutions of complex **1**. The connecting PEDOT:PSS layers were then spin-coated at 7500, 5500, 3500 and 3500 rpm for devices **T1**, **T2**, **T3** and **T4**, respectively. The thicknesses of the connecting PEDOT:PSS layers for devices **T1**, **T2**, **T3** and **T4** are 31, 42, 55 and 55 nm, respectively. Finally, the upper emissive layers (250 nm) were spin-coated at 3000 rpm from the acetonitrile solutions of complex **1**. 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 ($\sim 10^{-6}$ torr). Thicknesses of thin films were measured by ellipsometry. 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 constant bias voltages in a nitrogen glove box. The EL spectra were taken with a calibrated CCD spectrograph.

3. Results and discussions

Since all multilayered tandem LEC devices in this study were fabricated by solution processes, it is important to check the possibility of interlayer mixing during spin-coating, which would result in non-uniform layer thickness and thus poor reproducibility of device characteristics. Scanning electron microscope (SEM) photograph of the cross section of device **T1** is shown in Fig. 1. It is obvious that the interlayer boundaries are definite and a 2-unit tandem device structure is clearly present. Hence, both spin-coating of acetonitrile solution of complex **1** on a PEDOT:PSS layer and spin-coating of aqueous solution of PEDOT:PSS on a complex **1** layer would not induce significant interlayer mixing, ensuring successful construction of a tandem device structure.

To compare the device properties in different device structures, the EL characteristics of single-layered and tandem LECs were measured and are summarized in Table 1. All tandem LECs exhibited time-dependent EL spectra. Time-dependent EL spectra of devices **T1** (5.94 V), **T2** (5.96 V) and **T3** (6.00 V) are shown in Fig. 2(a–c), respectively.

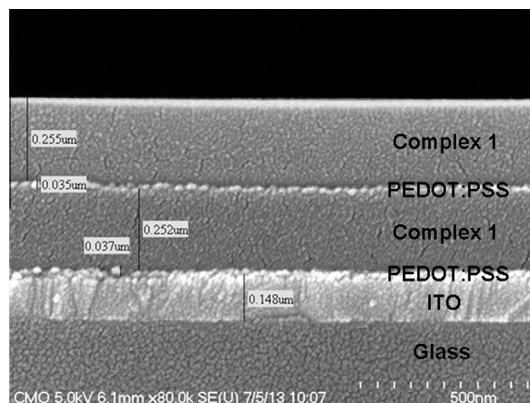


Fig. 1. SEM photograph of the cross section of device **T1**.

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