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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 ($\mu W\ cm^{-2}$) 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.

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 $Ru(dtb\text{-}bpy)_3(PF_6)_2$ (where dtb-bpy is 4,4′-ditertbutyl-2,2′-bipyridine) [5]. $Ru(dtb\text{-}bpy)_3(PF_6)_2$ 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.

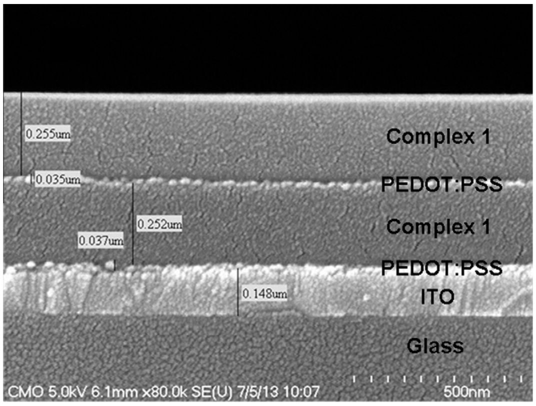

Fig. 1. SEM photograph of the cross section of device **T1**.

Table 1
Summary of EL characteristics of tandem LECs.

Device[a]	Bias (V)[b]	J_{max} (mA cm^{-2})[c]	t_{max} (min)[d]	L_{max} (μW cm^{-2})[e]	B_{max} (cd m^{-2})[f]	$\eta_{ext,\ max}$ (%)[g]	$\eta_{L,\ max}$ (cd A^{-1})[h]	$\eta_{p,\ max}$ (lm W^{-1})[i]	Lifetime (min)[j]
S1	2.73	7.05	51	119	30	2.46	2.40	2.69	396
S2	3.03	7.27	94	127	26	1.79	2.11	2.23	494
S3	4.00	45.90	4	673	298	2.69	2.27	1.78	7
T1	5.94	7.18	31	202	96	4.39	5.76	3.05	129
T2	5.96	7.09	32	218	89	5.52	5.62	2.97	129
T3	6.00	7.29	32	212	72	5.83	5.65	2.96	114
T4	7.28	26.69	8	655	300	5.41	4.78	3.75	10

[a] Devices **S1**, **S2** and **S3**: ITO (120 nm)/PEDOT:PSS (30 nm)/complex **1**/Ag (100 nm), where the thicknesses of complex **1** for devices **S1**, **S2** and **S3** are 250, 570 and 250 nm, respectively. Devices **T1, T2**, **T3** and **T4:** ITO (120 nm)/PEDOT:PSS (30 nm)/complex **1** (250 nm)/PEDOT:PSS/complex **1** (250 nm)/Ag (100 nm), where the thicknesses of the connecting PEDOT:PSS films for devices **T1, T2**, **T3** and **T4** are 31, 42, 55 and 55 nm, respectively.

[b] Bias voltage.

[c] Maximal current density.

[d] Time required to reach the maximal brightness.

[e] Maximal light output achieved at a constant bias voltage.

[f] Maximal brightness achieved at a constant bias voltage.

[g] Maximal external quantum efficiency achieved at a constant bias voltage.

[h] Maximal current efficiency achieved at a constant bias voltage.

[i] Maximal power efficiency achieved at a constant bias voltage.

[j] Time for the brightness of the device to decay from the maximum to half of the maximum under a constant bias voltage.

These devices possess similar structures except for thicknesses of connecting PEDOT:PSS layers of 31, 42 and 55 nm for devices **T1**, **T2** and **T3**, respectively. The operating voltages are selected to reach similar current densities in tandem and single-layered devices (ca. 7 mA cm^{-2}). Higher voltages are required for devices with thicker connecting PEDOT:PSS layers to achieve similar current densities. Fair comparison of device efficiencies should be performed under similar current densities since phosphorescent device efficiencies are sensitive to current densities due to triplet–triplet annihilation [33]. Evolution of EL spectra with time has also been observed in relatively thicker LECs (ca. 450 nm) based on the same complex [25]. It may be attributed to moving of recombination zone, which alters the output EL spectra from a microcavity structure [25,34]. It may not result from degradation of the emissive material since the EL spectra remained almost unchanged during 10 h operation for thinner (250 nm) single-layered LEC devices (**S1**). Constructive interference of the emitted light in a microcavity structure leads to enhanced output EL intensity at ca. 625 nm for tandem LECs (Fig. 2(a–c)). Moving of recombination zones in tandem LECs alters the wavelength at which constructive interference occurs and modifies the EL spectra with time. However, for thinner devices with approximately half thicknesses (**S1**), wavelength spacing between neighboring constructive interference would be much larger and thus enhanced EL intensity would more likely take place at spectral regions outside the emission spectrum of complex **1**, leading to relatively stable EL spectra even when the recombination zone is moving.

When the electrochemically doped layers are well established, p-doped/intrinsic (undoped)/n-doped (p–i–n) structure was getting stable and recombination zone is approaching a fixed position, rendering a stabilized EL spectrum. The stabilized EL spectra of tandem devices **T1** (5.94 V), **T2** (5.96 V), **T3** (6.00 V) and single-layered device **S1** (2.73 V) are compared in Fig. 3. Commission Internationale de l'Eclairage (CIE) coordinates of the EL spectra of devices **S1**, **T1**, **T2** and **T3** are (0.672, 0.328), (0.646, 0.354), (0.650, 0.350) and (0.648, 0.351), respectively. Owing to different microcavity structures, tandem devices (**T1**, **T2** and **T3**) showed EL spectra with more concentrated red emission while single-layered device (**S1**) exhibited enhanced emission at the near infrared region. Since the spectral overlap between the EL spectrum and the luminosity function [35] is larger for tandem devices (Fig. 3), it would be beneficial for tandem devices to obtain higher brightness as compared to single-layered devices with similar quantum efficiencies. Discrepancies in EL spectra of these three tandem devices (**T1**, **T2** and **T3**) can be observed (Fig. 3). Hence, the recombination zone positions, which affect the output EL spectra, would be different in these devices. It reveals that carrier balance would be altered when the thickness of connecting PEDOT:PSS layer in tandem LECs is adjusted.

The main advantages of tandem LECs are enhanced light outputs and quantum (or current) efficiencies under similar current densities. Time-dependent light output, brightness and EQE of tandem devices (**T1**, **T2** and **T3**) and single-layered device (**S1**) are compared in Fig. 4(a–c), respectively. The peak light outputs (μW cm^{-2}) of tandem devices **T1**, **T2** and **T3** were enhanced by factors of 1.70, 1.83 and 1.78, respectively, when compared to that of single-layered device **S1** under similar current densities (Table 1). These results confirm that both upper and lower cells connected in series (Fig. 1) contribute to output EL emission. It is noted that enhancement ratios in peak brightness of tandem devices **T1**, **T2** and **T3** as compare to that of single-layered device **S1** are as high as 3.20, 2.97 and 2.40, respectively, which are significantly higher than enhancements in light outputs (cf. Fig. 4a and b). Additional enhancements in brightness of tandem devices result from better matching of the luminosity function and their EL spectra (Fig. 3). Hence, additionally increased lumen or candela values can be obtained for tandem

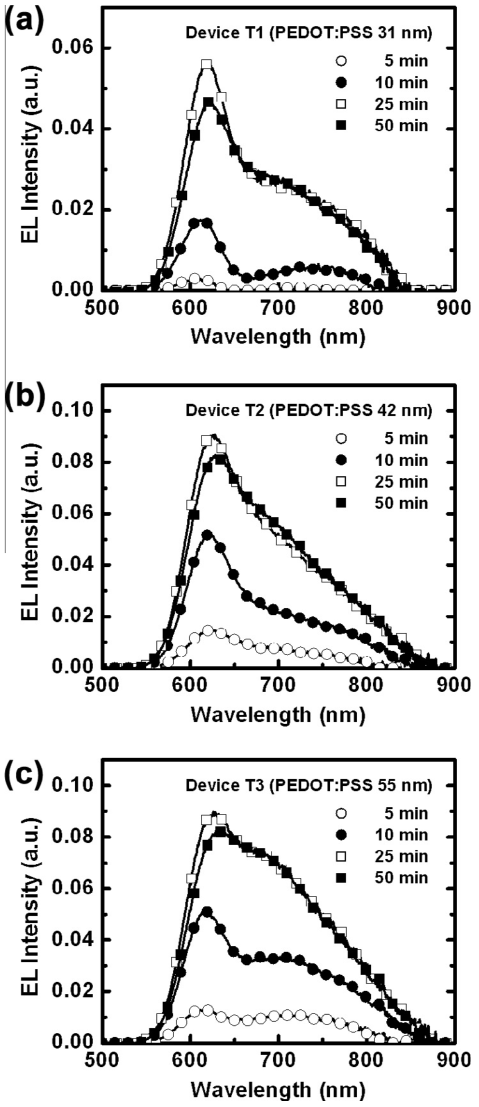

Fig. 2. Time-dependent EL spectra of devices (a) **T1** (5.94 V), (b) **T2** (5.96 V) and (c) **T3** (6.00 V).

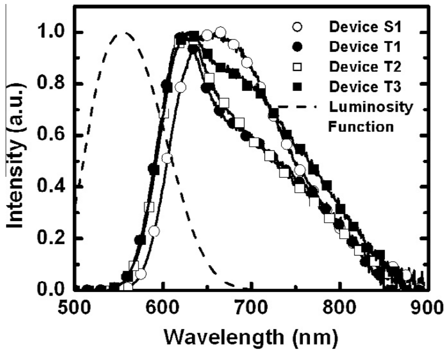

Fig. 3. Stabilized EL spectra of tandem devices **T1** (5.94 V), **T2** (5.96 V), **T3** (6.00 V) and single-layered device **S1** (2.73 V). The luminosity function is also shown for comparison of spectral overlap between the luminosity function and these EL spectra.

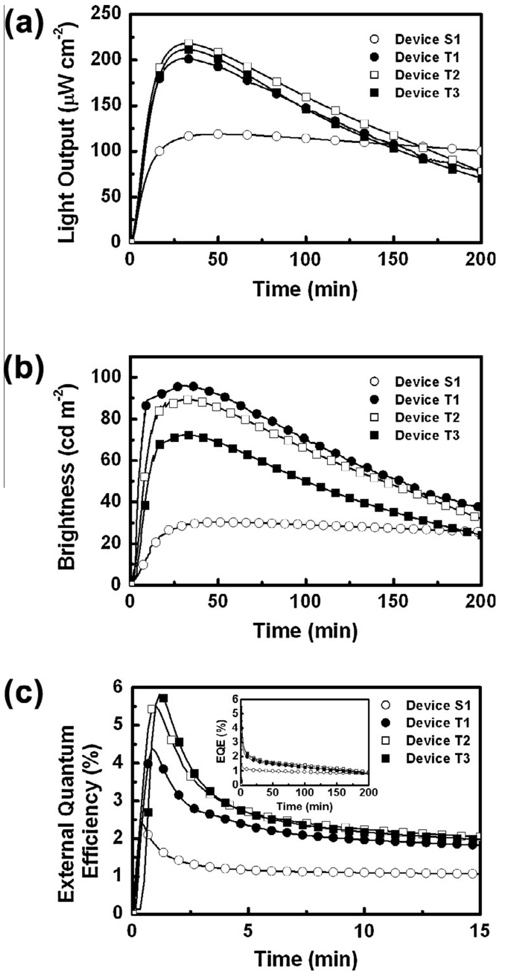

Fig. 4. (a) Light output, (b) brightness and (c) external quantum efficiency as a function of time for tandem devices **T1** (5.94 V), **T2** (5.96 V), **T3** (6.00 V) and single-layered device **S1** (2.73 V). Inset of figure (c): time-dependent external quantum efficiency during 0–200 min.

devices. It reveals that microcavity effect can be utilized to tailor EL spectra for further improving brightness of tandem LECs. The peak EQEs of tandem devices **T1**, **T2** and **T3** reach 4.39%, 5.52% and 5.83%, respectively. As compared to single-layered device **S1**, the peak EQEs are enhanced by ratios of 1.78, 2.24 and 2.37 for tandem devices **T1**, **T2** and **T3**, respectively. Thicker connecting PEDOT:PSS layers lead to higher EQEs. It would arise from modified carrier balance of the tandem devices when the thickness of the connecting PEDOT:PSS layer is adjusted. As shown in previously reported results of time-of-flight experiments, 2,2′-bipyridine based ionic ruthenium complexes exhibit higher electron mobilities than hole mobilities [36]. For the upper emitting unit of the tandem device or the single-layered device **S1**, i.e. PEDOT:PSS and Ag are used for anode and cathode, respectively, electron would be the excess carrier when electrochemical doped layers facilitate balanced carrier injection. A thicker connecting PEDOT:PSS layer in the tandem device would impede electron flow injected into the lower emitting unit, rendering improved carrier balance. The device efficiency of the upper emitting unit is similar to that of the single-layered device **S1** while the lower emitting unit is more efficient when the number of electrons is reduced. Therefore, overall device efficiency of the tandem device can be higher than twice of the corresponding single-layered device.

Although higher light outputs and device efficiencies can be obtained in tandem LECs, the device lifetimes of tandem LECs are lower than those of single-layered LECs (Fig. 4a and b). It may be related to exciton quenching near the doped regions while the recombination zone is moving toward electrodes [25]. Furthermore, residual water in the spin-coated PEDOT:PSS layer is difficult to completely remove with simple baking [37]. Degradation due to quenching species $[(bpy)_2(H_2O)RuORu(OH_2)(bpy)_2]^{4+}$ may also contribute to decreased device lifetimes [38]. Further studies will be required to clarify the dominant mechanism responsible for deteriorated device lifetimes of tandem LECs.

Enhancement in device efficiency can also be achieved by increasing the thickness of the emissive layer of LECs since the distance between the recombination zone and electrodes would be larger and exciton quenching near electrodes can be mitigated consequently [26]. To study the possibility of enhancement in device efficiency from significantly increased device thickness of the tandem device, a single-layered device (**S2**) with a thickness similar to that of a tandem device (570 nm) was fabricated and its EL characteristics are shown in Table 1 for comparison. Compared to thinner device **S1**, thicker device **S2** showed similar light output under a similar current density. However, device **S2** exhibited slightly lowered device efficiencies than device **S1** possibly due to altered carrier balance under a reduced electric field. These results confirm that only increased thickness of the emissive layer can not lead to doubled device efficiency of the tandem device. Both contributions of EL emission from the upper and the lower emitting units of the tandem device account for such efficiency enhancement.

Another advantage of tandem devices is improved lifetimes when operated under higher current densities to achieve higher light outputs. To examine device lifetimes at higher brightness, single-layered (**S3**) and tandem (**T4**) devices were biased under higher voltages (4.00 and 7.28 V for device **S3** and **T4**, respectively) to achieve higher brightness (ca. 300 cd m^{-2}). The device structures of device **S3** and **T4** are the same as device **S1** and **T3**, respectively. As shown in Fig. 5a, the lifetime of the tandem device (**T4**) is ca. 1.5 times longer than that of the single-layered device (**S3**). Since two emitting units contribute to EL emission, a lower current density is required for a tandem device to reach similar brightness obtained from a single-layered device. To achieve a maximum brightness of 300 cd m^{-2}, a maximum current density of 45.9 mA cm^{-2} is necessary for the single-layered device (**S3**) while only 26.69 mA cm^{-2} in the tandem device (**T4**) can lead to a similar brightness. Thus, the tandem device, which is operated under a lower current density, suffers mitigated degradation and exhibits a longer lifetime in consequence. Irreversible multiple oxidation and subsequent decomposition under a high electric field were proposed as the mechanism for degradation of the emissive materials of LECs [39]. Even when operated under a higher current density, the tandem device (**T4**) still showed a doubled EQE (5.41%) as compare to the single-layered device (**S3**) (2.69%) (Fig. 5b). These results reveal that a tandem device structure can be utilized to simultaneously achieve higher device efficiency and longer device lifetime for LECs giving out relatively higher brightness. It would be a feasible way to improve deteriorated device efficiencies and lifetimes accompanied by increased brightness, which are commonly observed for solid-state LECs.

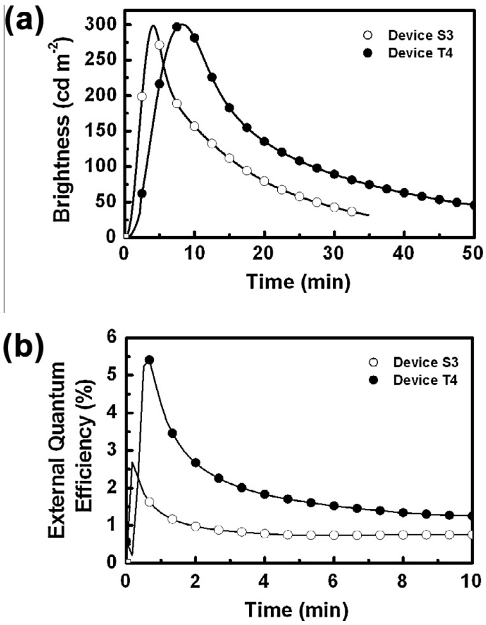

Fig. 5. (a) Brightness and (b) external quantum efficiency as a function of time for **T4** (7.28 V) and single-layered device **S3** (4.00 V).

The main focus of this report is to generally demonstrate tandem LECs for improving device efficiencies and lifetimes. Thus, these devices were fabricated by using commercially available compounds, which can be easily purchased by other research groups to repeat the same experiments. Based on similar tandem device structures, highly emissive iridium-based CTMCs [22,23,40] can be utilized to achieve even higher device efficiencies.

4. Conclusions

In summary, we have demonstrated tandem solid-state LECs by employing a thin PEDOT:PSS layer as a connecting layer between two emitting units. Light outputs (μW cm^{-2}) of the tandem devices were enhanced by ca. 1.8 times when compared to those obtained from the single-layered devices at similar current densities. Enhancements in brightness of the tandem devices can be further increased to ca. 3 times. Owing to differential microcavity effects in devices with different thicknesses, the single-layered devices showed EL emission centered at ca. 660 nm while enhanced EL emission occurred at ca. 625 nm was obtained for the tandem devices. Larger spectral overlap between the EL spectra of the tandem devices and the luminosity function results in higher lumen or candela values, rendering further enhanced brightness ($\mathrm{cd\,m^{-2}}$). Enhancement ratios of EQEs in the tandem devices can be optimized by adjusting the thickness of the connecting PEDOT:PSS layer to modify the carrier balance. The peak EQE obtained from the tandem LECs was up to 5.83%, which is 2.37 times higher than that of the single-layered devices. Furthermore, as compared to the single-layered device, the tandem device can also provide higher device efficiencies and longer lifetimes at a relatively higher brightness. Therefore, a tandem device structure would be an effective approach for solid-state LECs to improve device efficiencies and lifetimes, especially at a relatively higher brightness.

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