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Performance optimization of green tandem OLEDs with double emitting layers

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ABSTRACT

The utilization of a double emitting layer structure with well-matched energy levels has been shown to improve carrier balance and decrease operating voltages in organic light-emitting diodes (OLED). In this study, tandem OLEDs with double emitting layers (DEMLs) are successfully fabricated and optimized. Through adjusting the emitting layer thickness ratio and cavity length, the optimized green tandem OLEDs show driving voltages of 4.8 and 5.4 V, with current efficiency (CE)/power efficiency (PE) of 242.4 cd/A/158.6 lm W^{-1} and 237.1 cd/A/137.9 lm W^{-1} at 100 and 1000 cd m^{-2}, respectively. The ultra-high CE of 242.4 cd/A is the state of the art among all reported two-units green tandem OLEDs. Meanwhile, the device at the initial luminance of 10000 cd cm^{-2} also displays an extremely long LT50 of 734.8 h. In addition, the tandem OLED can emit pure green light with color coordinates of (0.27, 0.71), and has the ability to adjust the color coordinates by varying the cavity length.

1. Introduction

Displays and lighting sources with OLEDs (Organic Light Emitting Diodes) are currently in a rapid development stage due to their high brightness, low power consumption, extended lifespan, compact size, lightweight design, and flexibility [1,2]. An effective approach to ensure both low power consumption and high brightness is to construct tandem OLEDs by vertically stacking two or more electroluminescent (EL) units interconnected with a charge generation layer (CGL) [3,4]. This tandem structure not only improves current and power efficiency, but also reduces power consumption and extends operation duration [5,6]

In recent years, stacked structures have been utilized to enhance the optoelectronic performance of green-light OLEDs. Two key aspects for green-light OLEDs have been noted: 1) The development and application of different CGLs, including inorganic/inorganic, organic/inorganic, and organic/organic [7–11]. 2) The exploration of new structures, such as ultrathin structures, quantum dots, organic hybrid tandem structures, and double emitting layers structures [12–14]. Liao et al. [12] successfully obtained doping-free green phosphorescent tandem OLEDs with a current efficiency (CE) of 135.74 cd A^{-1} (EQE = 36.85 %) by utilizing Ir $(ppy)_2$(acac) as an ultrathin emissive layer. Chen et al. [14] designed a hybrid tandem structure by integrating QLED and OLED into tandem device, which achieved pure green emission with a full width at the half maximum (FWHM) of 44 nm, as well as a CE of 96.28 cd A^{-1} and EQE of 25.9 %. Ma et al. [10], Lee et al. [11], and Chang et al. [13] also reported efficient stacked green OLEDs with various types of CGLs. It is worth noting that Ma et al. [10] and Lee et al. [11] reported green tandem OLEDs with double emitting layers (DEMLs). Both achieved high current efficiencies of 205.9 cd A^{-1} and 200.9 cd A^{-1}, respectively. The design of the DEMLs with balanced electron and hole affinity enhances the injection and transport of charge carriers. Additionally, the well-matched energy levels in DEMLs devices promote better carrier balance and reduce non-radiative recombination. This highlights the advantages of tandem OLEDs with DEMLs. Thus, it is necessary to further study and optimize the various conditions of green tandem OLEDs with double-emitting layers.

In this work, green tandem light-emitting diodes with double-emitting layers were successfully fabricated by optimizing the thickness ratio of the DEML and the cavity length. The results show that the device achieves superior efficiency, long operating lifetime and low

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Table 1
The EL performances of the green single and tandem OLEDs.

Device		Single unit OLED	Tandem OLED
Voltage (V)	a	2.7	4.8
	b	3.3	5.4
	c	4.5	7.3
current efficiency (cd A^{-1})	a	109.8	242.4
	b	109.7	237.1
	c	101.7	221.3
power efficiency (lm W^{-1})	a	85.9	158.6
	b	69.3	137.9
	c	46.8	95.2
CIE1931 coordinates	c	0.26,0.70	0.27,0.71
max. luminance (cd m^{-2}) [V]		42533 [6V]	96488 [12V]
lifetime T50 (hour)	d	437.7	734.8

[a] Measured at 100 cd m^{-2}.
[b] Measured at 1000 cd m^{-2}.
[c] Measured at 10000 cd m^{-2}.
[d] Measured from L0 of 10000 cd m^{-2}.

operating voltage. Additionally, the optimal device can emit pure green light with color coordinates of (0.27, 0.71), and it has the ability to adjust the color coordinates by varying the cavity length.

2. Experimental section

A single-unit OLED with the following structure was fabricated: ITO (120 nm)/HAT-CN (20 nm)/TAPC (25 nm)/BCzPh:Ir(ppy)$_2$acac (1:7 %; 10 nm)/CBP:Ir(ppy)$_2$acac (1:7 %; 20 nm)/TPBi (45 nm)/LiF (1 nm)/Mg:Ag (1:10 %; 13 nm). The architecture of the tandem OLED was as follows: ITO (120 nm)/HAT-CN (20 nm)/TAPC (25 nm)/BCzPh:Ir(ppy)$_2$acac (1:7 %; 10 nm)/CBP:Ir(ppy)$_2$acac (1:7 %; 20 nm)/TPBi (45 nm)/Alq3:Yb (1:5 %; 30 nm)/HAT-CN (20 nm)/TAPC (30 nm)/BCzPh:Ir(ppy)$_2$acac (1:7 %; 10 nm)/CBP: Ir(ppy)$_2$acac (1:7 %; 20 nm)/TPBi (43 nm)/LiF (1 nm)/Mg:Ag (1:10 %; 13 nm).

The organic materials include HAT-CH (1,4,5,8,9,11-hexaazatriphenylene -hexacarbonitrile), TAPC (1,1-Bis[(di-4-tolylamino) phenyl]cyclohexane), BCzPh (9,9′-diphenyl-9H,9′H-3,3′-bicarbazole), CBP (4,4′- bis(9-carbazolyl)biphenyl), Ir(ppy)$_2$acac, Alq3, and TPBi (1,3,5-tris(N -phenylbenzimiazole-2-yl), while the metal materials consist of Mg, Ag, and Yb. The device structures of both a conventional single-unit OLED and a tandem OLED with two EL units are illustrated in Fig. 1. The energy levels and molecular structures of the materials used in the designed green tandem OLEDs are presented in Fig. 2. To fabricate the OLEDs, a 150 nm thick layer of patterned ITO with a sheet resistance of 15Ω/square was sputtered onto glass substrates by using a shadow mask. The ITO surface was cleaned with ultrasound and dried at 120 °C, followed by treatment in a UV-ozone instrument for 10 min. Subsequently, the substrates were transferred to a vacuum chamber for OLED production. All layers were grown sequentially by thermal evaporation under vacuum conditions (7.5×10^{-7} Torr). The deposition rates of organic and metal films were set to 0.5 Å S^{-1} and 1 Å S^{-1}, respectively. The active emissive area of the OLEDs measured 2mm × 2 mm. The current density-voltage-brightness characteristics of the devices were evaluated using a Keithley 2400 sourcemeter equipped with a calibrated silicon photodiode. EL spectra were captured using a Photo Research SpectraScan PR655. The lifetimes of OLED devices were obtained by using an OLED lifetime testing system (RTS101) under constant current mode.

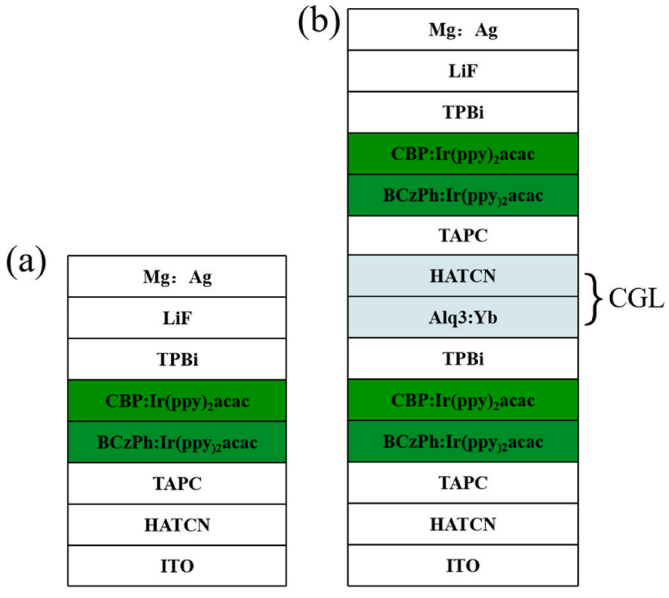

Fig. 1. Structure schematic diagrams of (a) conventional single-unit OLEDs and (b) tandem OLEDs including two EL units and charge generation layer using Alq3:Yb/HAT-CN.

3. Results and discussion

To achieve high efficiency and long operational lifetime, it is necessary to optimize the structure of tandem OLEDs. A key aspect of this optimization strategy is to adjust the thickness ratio of the double-emissive layer (DEML) [15,16]. Specifically, the thickness ratio of the double emitters (BCzPh:Ir(ppy)$_2$acac and CBP:Ir(ppy)$_2$acac) in the two light-emitting units is set at 5 nm:25 nm, 10 nm:20 nm, and 15 nm:15 nm, separately. They are labeled as T1, T2, and T3, respectively. The electroluminescent (EL) characteristics of these DEML tandem OLEDs are illustrated in Fig. 3 and their performances are summarized in Table S1. These parameters included voltage (V), current efficiency (CE), power efficiency (PE), CIE1931 coordinates (x,y), maximum luminance (cd m^{-2}), and T50 lifetime (h). Notably, the J-V-L characteristics of all devices are almost identical (Fig. 3(a)), while the emission spectra and corresponding CIE coordinates exhibit slight variations. The emission peak range is from 520 to 530 nm (Fig. 3(b)). As shown in Fig. 3 (c) and (d), compared to T3 and T1, the device T2 at a luminance of 100 cd cm^{-2} has the highest current efficiency (CE) of 192.9 cd A^{-1} and power efficiency (PE) of 126.2 lm W^{-1}. Furthermore, the lifetimes T50 (time to decay to half the initial brightness, with an initial luminance of 10000 cd m^{-2}) for devices T1, T2, and T3 are 543.6, 722.1, and 614.5 h, respectively. The voltage versus time curves (Fig. 3(f)) reveal that the operating voltage of device T2 is lower in comparison to devices T1 and T3. As time goes on, the operating voltage of device T2 slowly increases. This is due to its more effective charge carrier recombination. During the lifespan testing, we monitored the temperature changes of the devices (Fig. S1). Interestingly, device T2 emitted a lower temperature compared to devices T1 and T3 when illuminated. This indicates that the carrier recombination center of device T2 is closer to the center of the emitting layer, leading to reduced non-radiative recombination. Three devices also exhibit high color stability according to brightness changes from 100 to 10000 cd m^{-2} (Figs. S2–S3).

Fig. 4 illustrates the operational mechanism of devices T1, T2, and T3. The holes with recombination centers in device T1 primarily accumulate at the HTL/EML1 interface due to the low hole mobility and thin EML1 thickness. As the thickness of EML1 increases in device T2, hole permeability also increases, causing recombination centers to shift towards the cathode. This promotes the radiative recombination of electrons and holes in the emissive layer. At this stage, hole mobility reaches its optimal level. The thicker EML1 in device T3 enhances hole transmission distance, resulting in minimal movement of recombination centers. Consequently, triplet excitons cannot efficiently transfer to EML2 [17,18], leading to reduced efficiency and shortened lifespan in device T3. To test the theory further, transient EL testing have been conducted (Fig. S4). The testing results indicate that Device T2 exhibited a slower EL decay rate compared to devices T1 and T3. This delayed

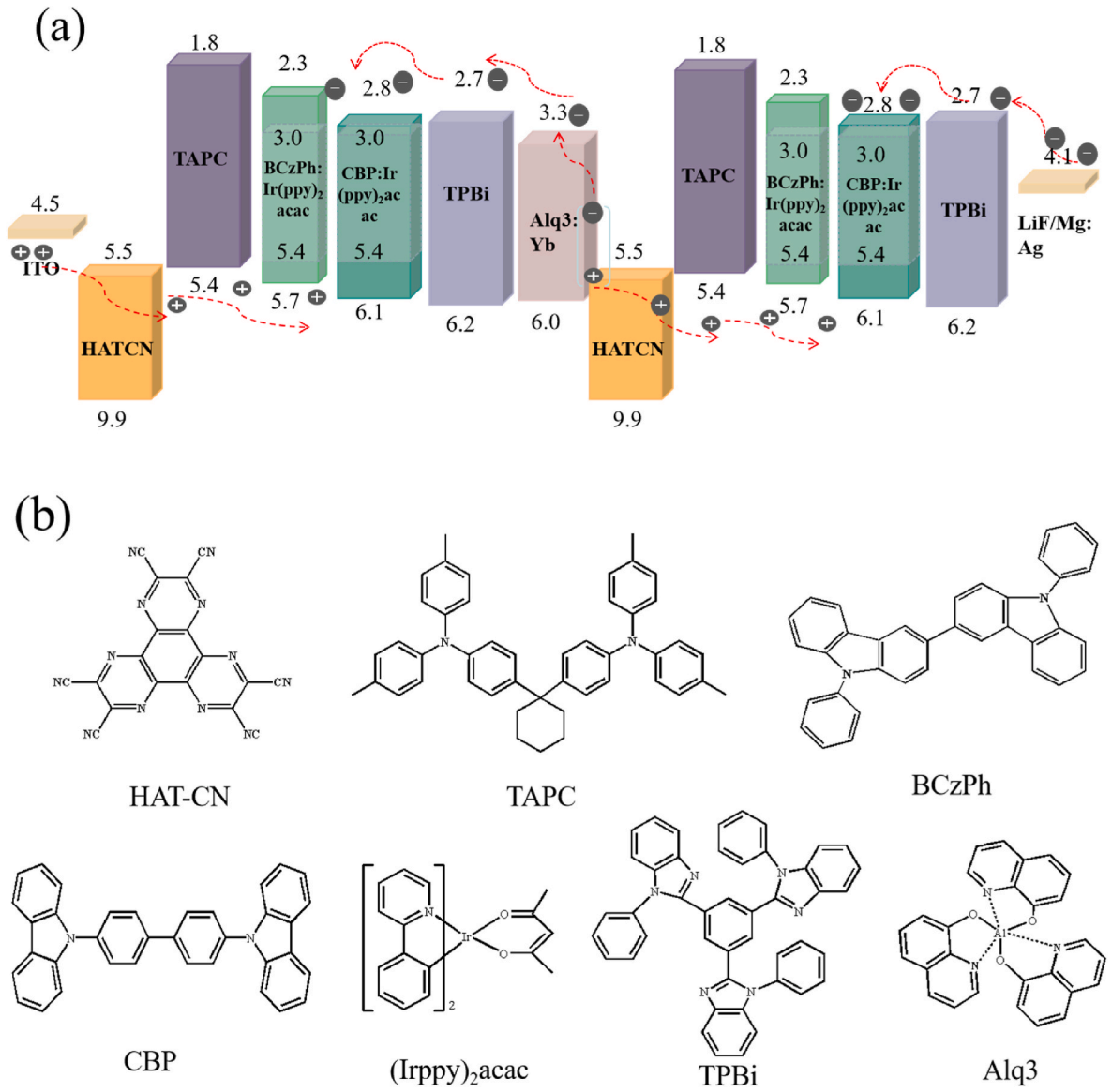

Fig. 2. (a) Energy level diagram and carrier transfer route of the designed tandem OLEDs (b) chemical structures of the materials employed in tandem OLEDs.

transient EL can be attributed to the recombination of trapped carriers, suggesting that the mixing ratio adjustment of the donor/acceptor blended host in the T2 device has achieved a relative balance of charge carriers.

The optimal resonant wavelength of the micro-cavity is influenced by the thickness of the functional layer. The thickness ratio of the DEML is fixed at 10 nm:20 nm for optimization, while the thickness of Electron Transport Layer 2 (ETL2) varies from 28 nm to 48 nm, denoted as devices A, B, C, D, and E. ETL2 is the electron transport layer TPBi located near the cathode (Fig. S5). As shown in Fig. 5(a) and (b) and Table S2, the brightness, CE and PE increase to their maximum values. However, there is a slight decrease when the thickness of ETL2 further increases. Notably, the device D with a 43 nm thickness of ETL2 achieves a brightness value of 96880 cd m^{-2}, a CE value of 242.4 cd A^{-1}, and a PE value of 158.6 lm W^{-1}. The comparative analysis in Table S3 presents the EL performance of our device with Alq3:Yb/HAT-CN CGL and other green tandem OLEDs with different CGL structures. The ultra-high CE of 242.4 cd A^{-1} is the state of the art among all reported two-units green tandem OLED. The emission spectrum of the devices shifts towards longer wavelengths and the full width at half maximum (FWHM) broadens with an increase in the thickness of ETL2. This suggests that the resonant wavelength is also influenced by the change in cavity length. (Fig. 5(c)). This indicates that the resonant wavelength also changes due to the cavity length change [19,20]. The CIE coordinates of the tandem OLED with a 28 nm-thick ETL2 (device A) are (0.22,0.72). With each 5 nm increase in ETL2 thickness, the CIE coordinates shift to (0.23,0.73), (0.25,0.72), (0.27,0.71), and (0.26,0.71) (Fig. 5(d)). Notably, the device A emits pure green light (0.22,0.72), which is closely consistent with the NTSC standard (0.21,0.71). In addition, adjusting the thickness of ETL2 allows for obtaining specific color coordinates. These devices also exhibit high color stability according to the brightness changes from 100 to 10000 cd m^{-2} (Figs. S6–S7).

Fig. 6 illustrates the electroluminescent (EL) characteristics of both the optimized tandem and single unit devices, the key properties are listed in Table 1. The J-V-L characteristics of single and tandem OLEDs are depicted in Fig. 6(a). The tandem OLEDs with an organic-metal junction charge generation layer (CGL) operate at 1000 cd m^{-2} at a voltage of 5.4V, which is less than twice the 3.3V operating voltage of a single OLED. This reduced operating voltage is attributed to the well-matched energy levels and triplet energies of the materials [21,22]. At a low current density of 4.5 mA cm^{-2}, the tandem OLED achieves a luminance of 10000 cd m^{-2}. However, ~~a~~ traditional single-unit OLED

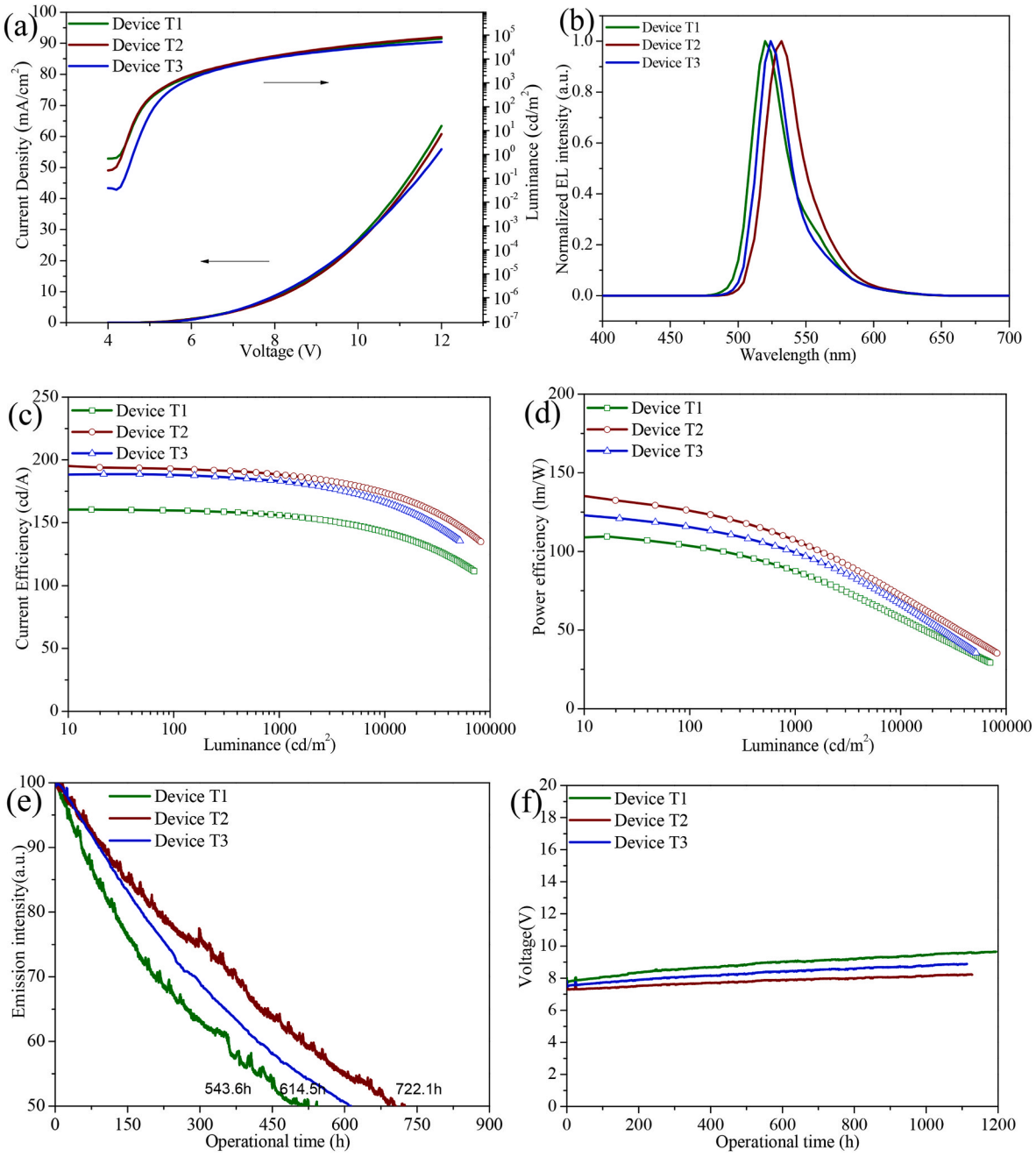

Fig. 3. (a) The current density−luminance−voltage characteristics; (b) the EL spectra; (c) brightness vs current efficiency; (d) brightness vs power efficiency; (e) Operation lifetime of devices at initial luminance of 10000 cd m^{-2}; (f) operating voltage vs time for tandem OLEDs T1, T2, and T3.

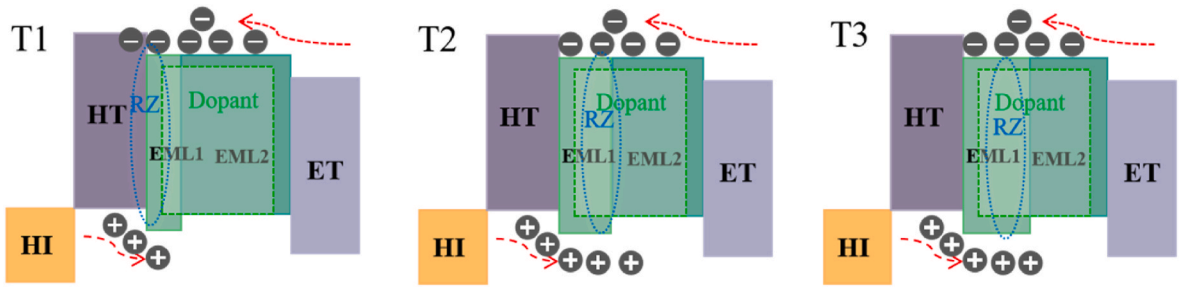

Fig. 4. Diagram of the working mechanism of the light-emitting unit in green tandem OLEDs with double emitting layer.

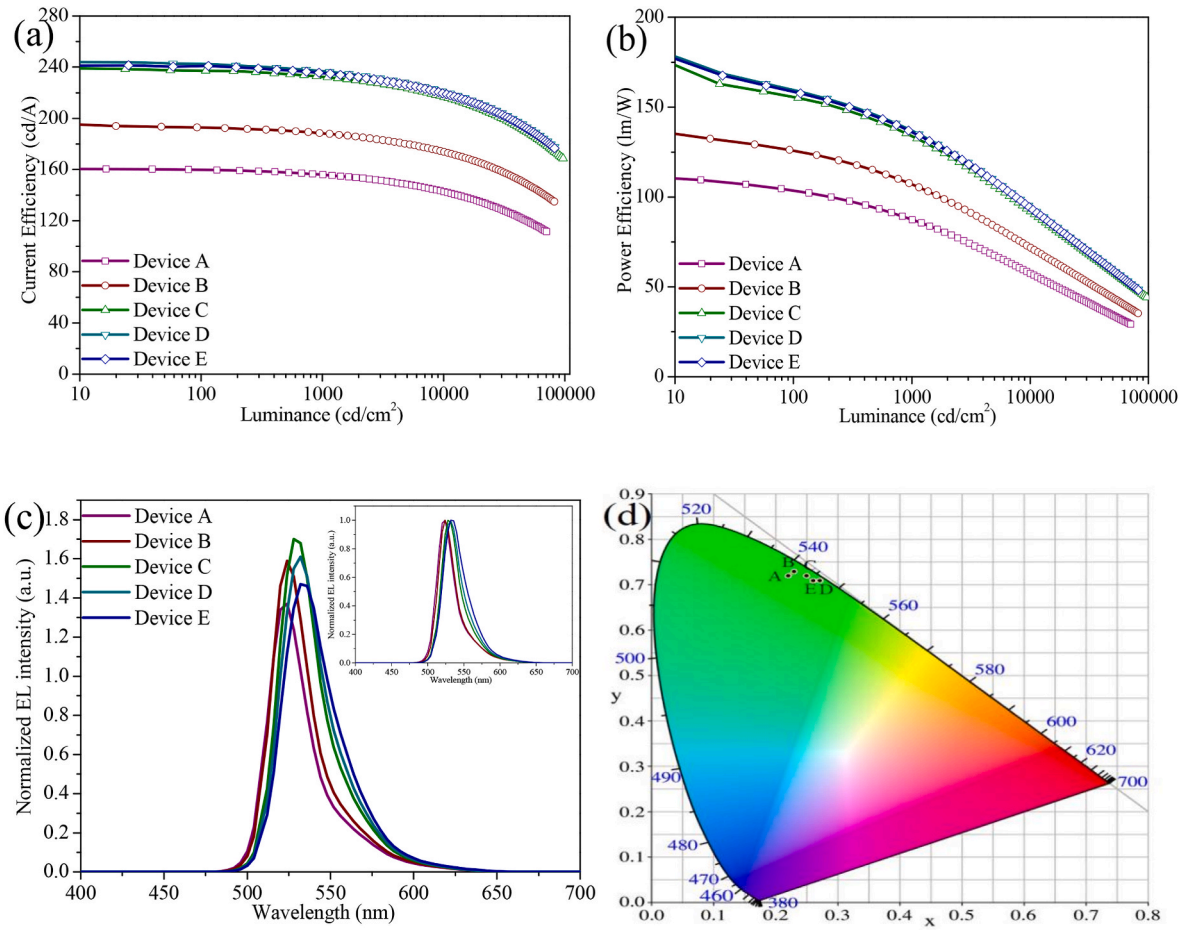

Fig. 5. (a) Brightness vs current efficiency; (b) brightness vs power efficiency; (c) the EL spectra; (d) the CIE chromaticity diagrams of tandem OLEDs.

requires a higher current density of 9.8 mA cm^{-2} (Fig. 6(b)). This difference can be attributed to the superior charge generation and transfer properties of the CGL. When a forward bias is applied, the CGL generates opposite charges and injects them into the upper and lower EL units, causing both units of the tandem OLED to emit light simultaneously. Consequently, tandem OLEDs achieve the same brightness with lower current density compared with single OLEDs. Fig. 6(c)–(d) illustrates the efficiency curves of both the tandem and single unit devices. The current efficiency (CE) of the tandem OLED is approximately 2.2 times that of the single-unit OLED, achieving impressive values of 242.4 cd A^{-1}, 237.1 cd A^{-1}, and 221.3 cd A^{-1} at brightness levels of 100, 1000, and 10000 cd m^{-2}, respectively. Additionally, the power efficiency (PE) is also enhanced, reaching 158.6/137.9/95.2 (lm W^{-1}) at 100/1000/10000 cd m^{-2}. The superior efficiency of Ir(ppy)$_2$acac is attributed to its higher horizontal dipole moment and the efficient energy transfer from the exciplex host to the emitter [23,24]. Hole transport material (BCzPh) and electron transport material (CBP) were selected as electron donors and acceptors, respectively, to generate exciplex. Together with the green light emitter Ir(ppy)$_2$acac, an efficient EML host/guest system can be established. The Ir(ppy)$_2$acac of this EML structure has a high horizontal dipole ratio (about 80 %), which improves the external decoupling efficiency of the device and suppresses Coulomb scattering during energy transmission [13]. Enhancing the power efficiency of tandem OLEDs requires either an increase in the brightness or a reduction in the operational voltage at a certain current density. In our work, the voltage of the optimized tandem OLED is lower than twice that of the single-unit device. Additionally, at the same current density, the brightness of the tandem OLED is more than twice that of the single-unit OLED (as shown in Fig. 6(b)). Consequently, the power efficiency is also significantly enhanced compare to the single-unit device.

The electroluminescence (EL) spectra of the tandem and single unit devices are shown in Fig. 6(e), pure green emission was observed at 528nm/525 nm. The corresponding FWHM is 30nm/33 nm. This suggests that the exciton formation region is precisely located in the emission layer (EML) of each emission units (EUs). The spectral profiles reveal minor differences, as the tandem devices exhibit relatively narrow EL spectra without shoulder peaks. This discrepancy in the spectrum is believed to be due to the optical path difference caused by the complex architecture of tandem devices and related microcavities [25]. Fig. 6(f) presents the lifetime data of both devices. The estimated T50 (time taken for the luminance to drop to 50 % of the initial value of 10000 cd m^{-2}) for the single unit OLED is 437.7 h and it is 734.8 h for the tandem OLED. This difference can be attributed to two factors: 1) tandem devices require approximately half of the current density to achieve the same brightness. 2) tandem devices need the efficient charge generation layer (CGL) units and the effective forster resonance energy transfer (FRET) from host BCzPh/CBP to (Irppy)$_2$acac [26].

4. Conclusions

In this work, green tandem OLEDs with DEMLs were prepared successfully. the DEMLs thickness ratio and cavity length in tandem OLEDs were optimized. The optimized green tandem OLED achieves an ultra-high CE of 242.4 cd A^{-1} at the luminance of 100 cd cm^{-2}. This value is the state of the art among all reported two-units green tandem OLED. The lifetime T50 of device is 734.8 h at an initial luminance of 10000 cd cm^{-2}. Moreover, these devices demonstrate adjustable color purity by

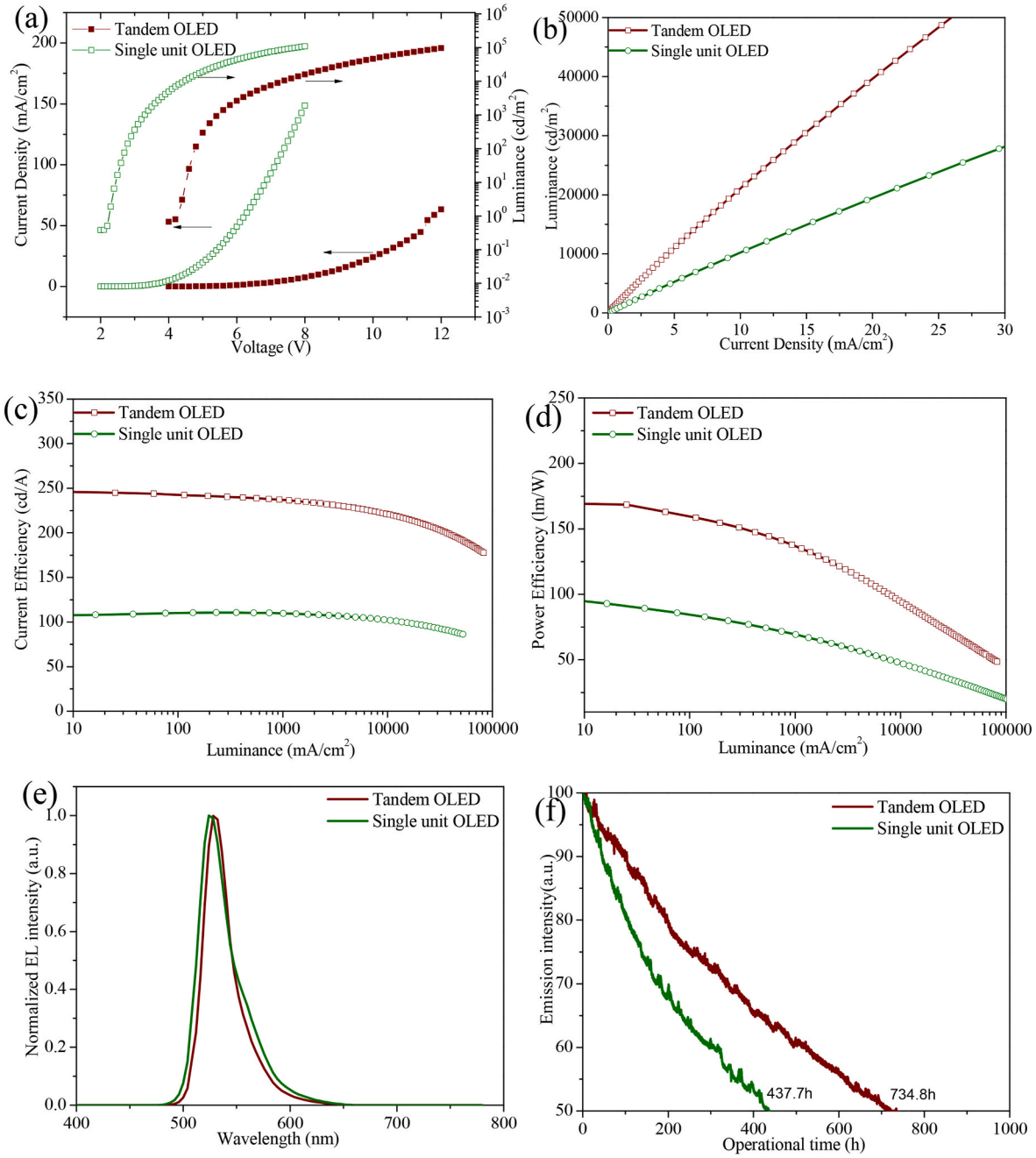

Fig. 6. (a) the current density–luminance–voltage characteristics; (b) the current density vs luminance characteristics; (c) brightness vs current efficiency; (d) brightness vs power efficiency; (e) normalized EL spectra at a luminance of 10000 cd m^{-2}; (f) lifetime characteristics at an initial luminance of 10000 cd cm^{-2} of optimized tandem OLED and single-unit OLED.

manipulating the cavity length. Our works revealed that the photoelectric performance and lifetime of the tandem green OLEDs can be significantly improved by introducing double emitting layers.

CRediT authorship contribution statement

Qiming Yang: Writing – original draft, Data curation, Conceptualization. **Fuli Qian:** Software, Resources, Investigation. **Guoru Gou:** Supervision, Resources, Conceptualization. **Tilu Wang:** Software, Investigation, Formal analysis. **Yu Duan:** Supervision, Resources, Conceptualization. **Chaoyu Lu:** Methodology, Investigation, Formal analysis. **Guanghua Wang:** Supervision, Formal analysis, Conceptualization. **Liangfei Duan:** Software, Project administration, Methodology. **Wen Yang:** Validation, Methodology, Investigation. **Yong Zhang:** Writing – review & editing. **Peizhi Yang:** Writing – review & editing, Validation, Supervision.

Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled.

Data availability

No data was used for the research described in the article.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jlumin.2024.120798.

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