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RESEARCH ARTICLE

Improved stability of blue TADF organic electroluminescent diodes via OXD-7 based mixed host

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Abstract Thermally activated delayed fluorescence (TADF) organic light-emitting diodes (OLEDs) have been demonstrated in applications such as displays and solid-state lightings. However, weak stability and inefficient emission of blue TADF OLEDs are two key bottlenecks limiting the development of solution processable displays and white light sources. This work presents a solution-processed OLED using a blue-emitting TADF small molecule bis[4-(9,9-dimethyl-9,10-dihydroacridine) phenyl]sulfone (DMAC-DPS) as an emitter. We comparatively investigated the effects of single host poly(N-vinylcarbazole) (PVK) and a co-host of 60% PVK and 30% 2,2′-(1,3-phenylene)-bis[5-(4-tert-butylphenyl)-1,3,4-oxadiazole] (OXD-7) on the device performance (the last 10% is emitter DMAC-DPS). The co-host device shows lower turn-on voltage, similar maximum luminance, and much slower external quantum efficiency (EQE) roll-off. In other words, device stability improved by doping OXD-7 into PVK, and the device impedance simultaneously and significantly reduced from 8.6×10^3 to 4.2×10^3 Ω at 1000 Hz. Finally, the electroluminescent stability of the co-host device was significantly enhanced by adjusting the annealing temperature.

Keywords blue thermally activated delayed fluorescence organic light-emitting diode (TADF OLED), 2,2′-(1,3-phenylene)-bis[5-(4-tert-butylphenyl)-1,3,4-oxadiazole] (OXD-7), bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone (DMAC-DPS), stability

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1 Introduction

Since the first organic light-emitting diode (OLED) was reported by Tang and Vanslyke in 1987 [1], OLEDs have attracted much attention due to their high brightness, flexibility, relative ease of large-scale manufacturing, and potentially higher efficiency and lower cost. Thermally activated delayed fluorescence (TADF) materials have attracted more attention due to the nearly 100% internal quantum efficiency and reduced material price/toxicity. These types of materials are usually based upon donor-acceptor units either in a single molecule or in two different molecules forming an exciplex. However, weak stability and inefficient emission of blue TADF OLEDs are two of the key bottlenecks limiting the development of solution processable displays and white light sources.

In recent years, many new blue-emitting TADF materials with relatively complex molecular structures, including tert-butyl substituted hetero-donors, boron-based emitters, and acridan analogs with heavy group 14 elements, etc. have been reported [2–4]. However, the efficiency, brightness, and stability of blue TADF OLEDs, especially deep blue, remain challenging goals because it is difficult to find suitable hosts with enough high triplet state energy higher than that of the emitter. A co-host system with bipolar or exciplex features is an effective strategy to resolve this issue. New co-hosts, such as incorporating 3,5-di(N-carbazolyl)tetraphenylsilane (SimCP) as the hole transporter and 2,4,6-tris(m-(2-trifluoromethyl)phenyl)-1,3,5-triazine (oCF3-T2T) as the electron transporter, or exciplex forming co-hosts composed of (tris(4-carbazoyl-9-ylphenyl)amine (TCTA):((30-(4,6-diphenyl1,3,5-triazin-2-yl)-(1,10-biphenyl)-3-yl)-9-carbazole) (CzTrz) or 1,3-bis (N-carbazolyl)benzene (mCP):(3,5-di-3-pyridylphenyl)-2-methylpyrimidine (B3PYMPM), have been applied [5,6].

In 2014, a high-efficiency blue-emitting TADF small molecule, bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl] sulfone (DMAC-DPS), was first reported by Zhang et al. [7]. However, the reported DMAC-DPS based blue OLED device structure was complicated and difficult to fabricate. Zhang et al. reduced the device complexity and fabricated non-doped DMAC-DPS TADF OLEDs with external quantum efficiency (EQE) value up to 19.5% [8]. Although the non-doped device demonstrated high efficiency, the turn-on voltage was still high and the device stability was worse. Recently, Duan et al. investigated the influence of interlayer compatibility on the lighting properties and stability of DMAC-DPS-based blue TADF devices. It was concluded that intralayer compatibility has an important impact on the device's EQE, while interlayer compatibility influenced on device stability [9]. By adjusting the intra- and intermolecular interplays of some novel hosts, such as phosphanthrene oxide host 5,10-diphenyl-phosphanthrene 5,10-dioxide (DPDPO2A), the Dexter energy transfer to dopant DMAC-DPS was facilitated [10–12]. Due to its large band gap, high triplet energy, and close polarity (dipole moment of 7.98) with emitters, an evaporation-based processed bis[2-(diphenylphosphino)phenyl]ether oxide (DPEPO) has also been widely chosen as host material for DMAC-DPS emitters [13,14]. Wu et al. proposed a universal design strategy for white TADF OLEDs by doping orange TADF emitters into blue TADF hosts to develop an effective exciton-confined emitting layer (EML) structure, where DMAC-DPS was used as the host. This design was compared with single-layer and double-layer emissive devices, and the EQE and device operational stability were both significantly improved [15]. To fabricate hybrid white OLEDs, Zhao et al. employed a non-doped DMAC-DPS-based blue EML and an ultrathin iridium complex, a bis[2-(4-tertbutylphenyl)benzo thiazolato-N,C-2′]iridium(acetylacetonate)[(tbt)(2)Ir(acac)] based yellow EML, by optimizing the DMAC-DPS thickness and achieving a warm white OLED with maximum current efficiency, power efficiency, and EQE values of 34.9 cd/A, 29.2 lm/W, and 11.4%, respectively [16]. Yang et al. employed synergetic solvents or mixed hole injection layers to improve the film morphology or hole injection of a solution-processed DMAC-DPS-based TADF OLED [17,18]. Particularly, as an excellent blue-emitting material, DMAC-DPS has demonstrated extensive applications in vacuum-evaporation and solution-processed OLEDs as doped and non-doped TADF emitters or hosts.

Generally, organic compounds and polymers involving the 1,3,4-oxadiazole unit exhibit good stability to heat and oxidation, and the conductivity is higher than carbon-containing aromatic systems [1], such as the W-shaped bipolar derivatives containing accepting oxadiazole and donating carbazole moieties designed by Hladka et al. [20]. Particularly, 2,2′-(1,3-Phenylene)-bis[5-(4-tert-butylphenyl)-1,3,4-oxadiazole] (OXD-7) is popular owing to its photoluminescence (PL) and electroluminescence (EL) bands in the blue-light region together with high luminescent brightness, good air stability, and film formation. Also, OXD-7 can easily capture and transport electrons. Usually OXD-7 is individually applied as a host for evaporation-processed deep blue TADF OLEDs [21,22], or the formation of an exciplex state used for Ir-complex-based phosphorescent OLEDs (PHOLEDs) to obtain longer lifetimes and better lighting performances [23]. OXD-7 could form a bipolar co-host in solution-processed OLEDs with many hole-transport hosts, such as TCTA, and poly(N-vinylcarbazole) (PVK) [24]. In addition, apart from functioning as a host, OXD-7 can also be used as an emitter. Zhang et al. applied OXD-7 as an emitting material and achieved an efficient UV OLED, which showed a high EQE of 1.43% at a peak of 388 nm [25].

However, there has been limited reports on OXD-7-based mixed co-hosts for improved performance and stability of solution-processed DMAC-DPS-based OLED devices. Herein, we combine the advantages of DMAC-DPS and OXD-7, and comparatively investigate the effects of single host PVK and a co-host of 60% PVK and 30% OXD-7 on device performance (the last 10% is emitter DMAC-DPS). The co-host device shows lower turn-on voltage, similar maximum luminance, and much slower EQE roll-off. By adjusting the annealing temperature, the device stability of the co-host device is significantly improved.

2 Experimental

All materials, including poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), PVK, DPEPO, DMAC-DPS, OXD-7, 4,7-diphenyl-1,10-Phenanthroline (Bphen), lithium fluoride (LiF), and aluminum (Al) were purchased commercially and directly used without further purification. The molecular structures of these materials are displayed in Fig. 1(a). Indium tin oxide (ITO, 15 Ω/sq, 150 nm) on glass substrates were consecutively cleaned via ultrasonic baths in detergent, deionized water, acetone, and isopropanol, each for about 15 min, and then finally treated in an ultraviolet-ozone chamber for 30 min before use. Hole injection layer (HIL) PEDOT:PSS was fabricated by spin-coating onto the clean ITO substrates at 3000 r/min for 60 s and annealed at 130°C for 15 min.

The device configuration is ITO/PEDOT:PSS (30 nm)/EML (*X* nm)/DPEPO (10 nm)/Bphen (30 nm)/LiF (0.8 nm)/Al (80 nm), and the EML incorporates a single host system of PVK:DMAC-DPS (9:1, wt%) and co-host system of PVK:OXD-7:DMAC-DPS (6:3:1, wt%). The

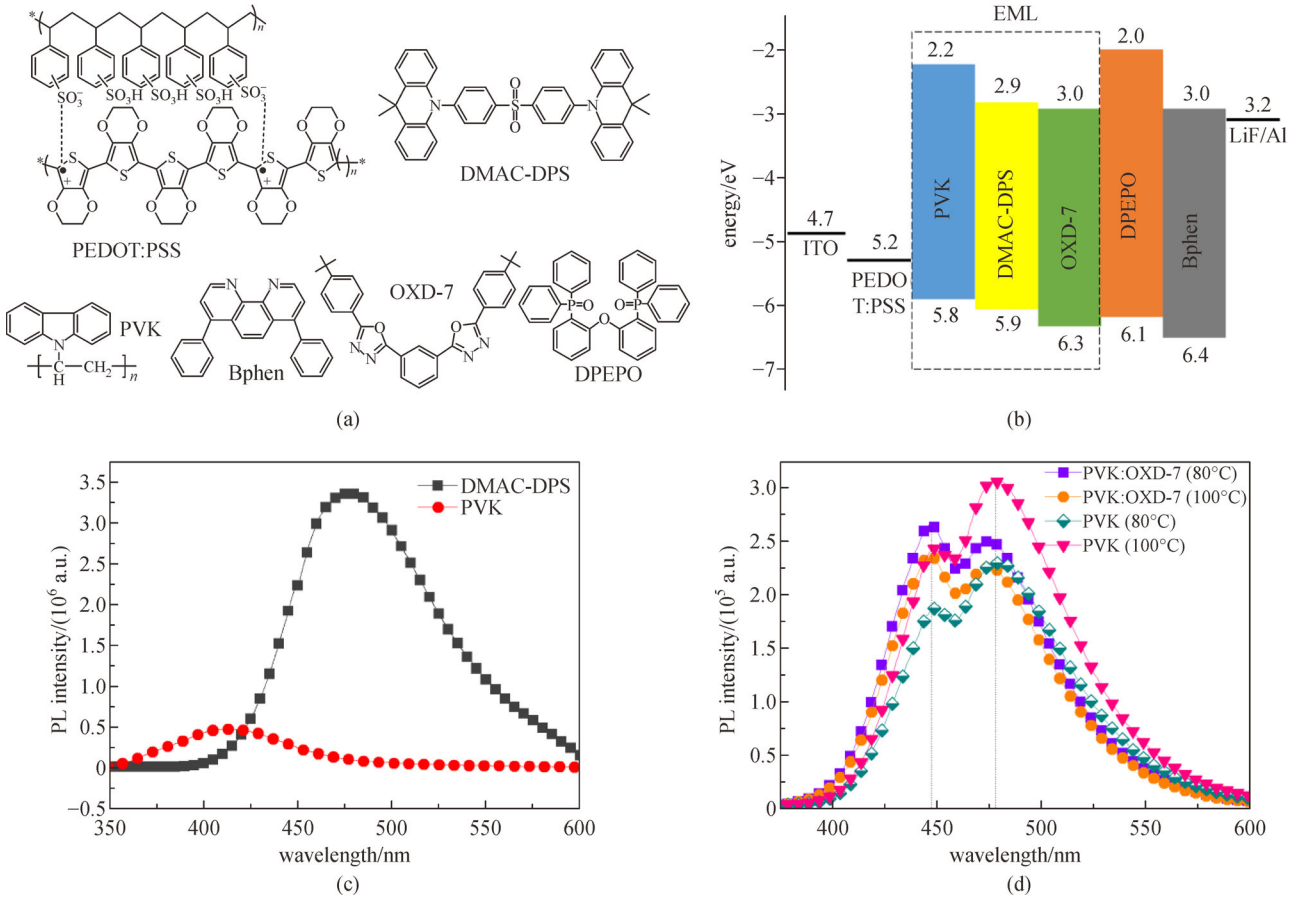

Fig. 1 (a) Molecular structures of PEDOT:PSS, DMAC-DPS, PVK, Bphen, OXD-7, and DPEPO. (b) Energy-level diagram of the co-host device. PL spectra of the pristine 40 nm DMAC-DPS film (c) and EML blend films (40 nm) with single host and co-host via annealing at 80°C and 100°C, respectively (d) on quartz substrates

energy levels of all the layers in the device are depicted in Fig. 1(b). The EML solution was prepared by mixing PVK and DMAC-DPS or PVK, OXD-7, and DMAC-DPS to obtain a 10 mg/mL chloroform solution. After stirring overnight, the mixed solution was filtered with a 0.45 μm filter before use. The solution was spin-coated onto HIL at 3000 r/min for 30 to 60 s and annealed at 80°C to 100°C for 30 min. Other organic materials were consecutively thermally evaporated onto the EML to obtain a complete OLED device. All organic materials and metals were deposited at an evaporation rate of approximately 0.6 and 5.0 Å/s at a pressure of 1.5×10^{-4} Pa, respectively. The active area (2 μm × 2 μm) was decided by the overlapping of anode and metal. The molecular structures of the organic materials and energy-level diagrams of the devices are shown in Fig. 1.

All device measurements were completed immediately after manufacturing without packaging at room temperature. The current density-voltage-luminance (*J-V-L*) characteristics and EL spectra were measured by a testing kit system consisting of a Keithley 2400 source-meter and Photo Research PR-650 spectrophotometer. The images of films were measured by atomic force microscopy (AFM) (Nanonavi SPA-400SPM, Japan). All PL spectra were measured by a spectrofluorometer (FLSP920, Edinburgh Instruments Corp., UK). The impedance spectroscopy was measured with a computer controlled programmable Agilent 4294A and Wayne Kerr 6500B precision impedance analyzer at a constant ac bias of 50 mV.

3 Results and discussion

To understand emission color stability, we measured the PL spectra of pristine DMAC-DPS and PVK films and blend films (40 nm) of PVK:DMAC-DPS as well as PVK:OXD-7:DMAC-DPS prepared at annealing temperatures of 80°C and 100°C, as shown in Figs. 1(c) and 1(d), respectively. The pristine DMAC-DPS and PVK films were prepared via the same process with the EML demonstrating a structureless peak emission at 478 and 425 nm, respectively (Fig. 1(c)). When DMAC-DPS is mixed with PVK or PVK:OXD-7 via solution-processing, the PL emission presents a main peak at 478 nm together

with an acromion near to 449 nm which is likely the emission from PVK when comparing with Fig. 1(c). The identical spectrum shape for the PVK and PVK:OXD-7-based blend films suggests that no exciplex is formed between PVK and OXD-7 with the complete energy transfer from host to DMAC-DPS.

Figure 2 demonstrates the AFM images of the EML films with single host or co-host prepared at various annealing temperatures on quartz substrates. It is seen that for both single host and co-host EML films, 100°C annealing treatment leads to smoother films. The root mean square (RMS) roughness decreases from 0.95 to 0.90 nm for single host EML, and from 0.68 to 0.49 nm for co-host EML, respectively. Compared with single host EML, the RMS roughness of the co-host EML decreases, implying that the incorporation of OXD-7 into PVK helps to improve the flatness of the EML film.

To gain insights into the co-host effect on OLED characteristics, we fabricated devices with hosts of both PVK:OXD-7 and PVK. The EL characteristics are shown in Fig. 3, and the device structure, as well as the corresponding performance parameters, are summarized in Table 1.

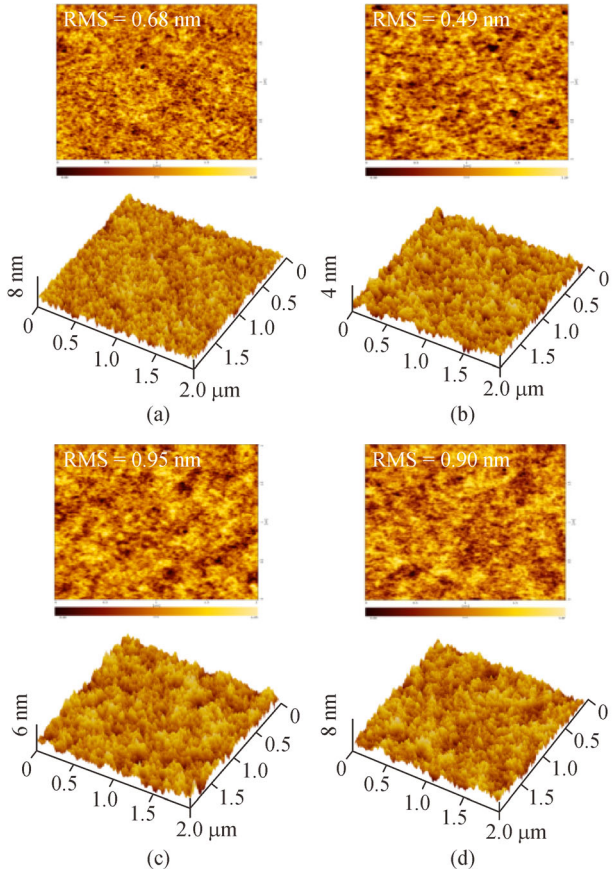

Fig. 2 AFM surface topographic and 3D images for different EML blend films on ITO substrates. (a) PVK:OXD-7:DMAC-DPS EML annealed at 80°C for 30 min. (b) PVK:OXD-7:DMAC-DPS EML annealed at 100°C. (c) PVK:DMAC-DPS EML annealed at 80°C. (d) PVK:DMAC-DPS EML annealed at 100°C

In combining the lighting characteristics shown in Fig. 4 and the EL parameters demonstrated in Table 2, it can be concluded that the EML spin-coated for 30 s achieves better EL performance than that spin-coated for 60 s. This is likely ascribed to the decreased electrical properties of the thinner EML spin-coated at 60 s. Although single host devices show higher CE, PE, and EQE, the co-host devices demonstrate lower turn-on voltages at about 3.2 V. The corresponding EL spectra are inserted in Fig. 3(c), which are shown to be structureless and similar to the PL spectrum of the pristine DMAC-DPS film, but different from the PL spectra of the EML shown in Fig. 1(d). It implies that in the EL process all of the host energy is transferred to DMAC-DPS. The EL intensities of PVK:OXD-7 devices are stronger than that of PVK devices because at the same bias voltage, the luminance values of PVK:OXD-7 devices are larger than that of PVK devices before reaching a maximum value.

The normalized EQE spectra are inserted in Fig. 3(d), and at low current density, the PVK devices show relatively high EQE, but faster efficiency roll-off, while PVK:OXD-7 devices depict much higher EQE at relatively high current density and slower efficiency roll-off. Therefore, the incorporation of OXD-7 significantly improves the device stability.

The AFM results suggest that the annealing temperature for preparing EMLs has a significant effect on the film morphology and device performance. To further improve the EL performance of the co-host device, we fabricated the EML of PVK:OXD-7:DMAC-DPS (6:3:1, wt%)-based OLED devices via spin-coating at 2000 r/min for 30 s and annealed at 80°C and 100°C for 30 min, respectively. The EML is thicker (20 nm) than that (15 nm) of the device shown in Fig. 3. The EL characteristics are shown in Fig. 4, and the corresponding performance parameters are summarized in Table 2. The thicker EML contributes to larger turn-on voltage but higher CE, PE, and EQE for the same device annealed at 80°C and spin-coated for 30 s. In addition, it is obvious that annealing temperature for the EML significantly influences EL properties. The luminance at 100°C is much higher than that at 80°C. The EL peak emissions are located at 500 and 480 nm for 80°C and 100°C, respectively. The device prepared at 80°C indicates better EL (CE, PE, and EQE) characteristics at relatively low luminance, while the device prepared at 100°C demonstrates good EL characteristics at relatively high luminance (as shown in Figs. 4(b) and 4(c)). It is seen that the device annealed at 100°C denotes slower efficiency roll-off at relatively high brightness, indicating higher device stability.

The roll-offs of CE and PE of the device annealed at

Table 1 EL characteristics of ITO/PEDOT:PSS (30 nm)/host:DMAC-DPS (15 nm)/DPEPO (10 nm)/Bphen (30 nm)/LiF (0.8 nm)/Al (80 nm) with various EMLs by spin-coating at 3000 r/min for 30 s or 60 s via annealing at 80°C for 30 min

host	time/s	L_{max}/(cd·m^{-2})	CE_{max}/(cd·cm^{-2})	PE_{max}/(lm·W^{-1})	EQE_{max}/%
PVK	30	228	2.08	1.18	1.37
PVK	60	217	1.63	0.85	1.12
PVK:OXD-7 (6:3, wt%)	30	207	0.59	0.41	0.51
PVK:OXD-7 (6:3, wt%)	60	238	0.46	0.29	0.40

Notes: L_{max} is the maximum luminance; CE_{max} is the maximum current efficiency; PE_{max} is the maximum power efficiency; EQE_{max} is the maximum external quantum efficiency.

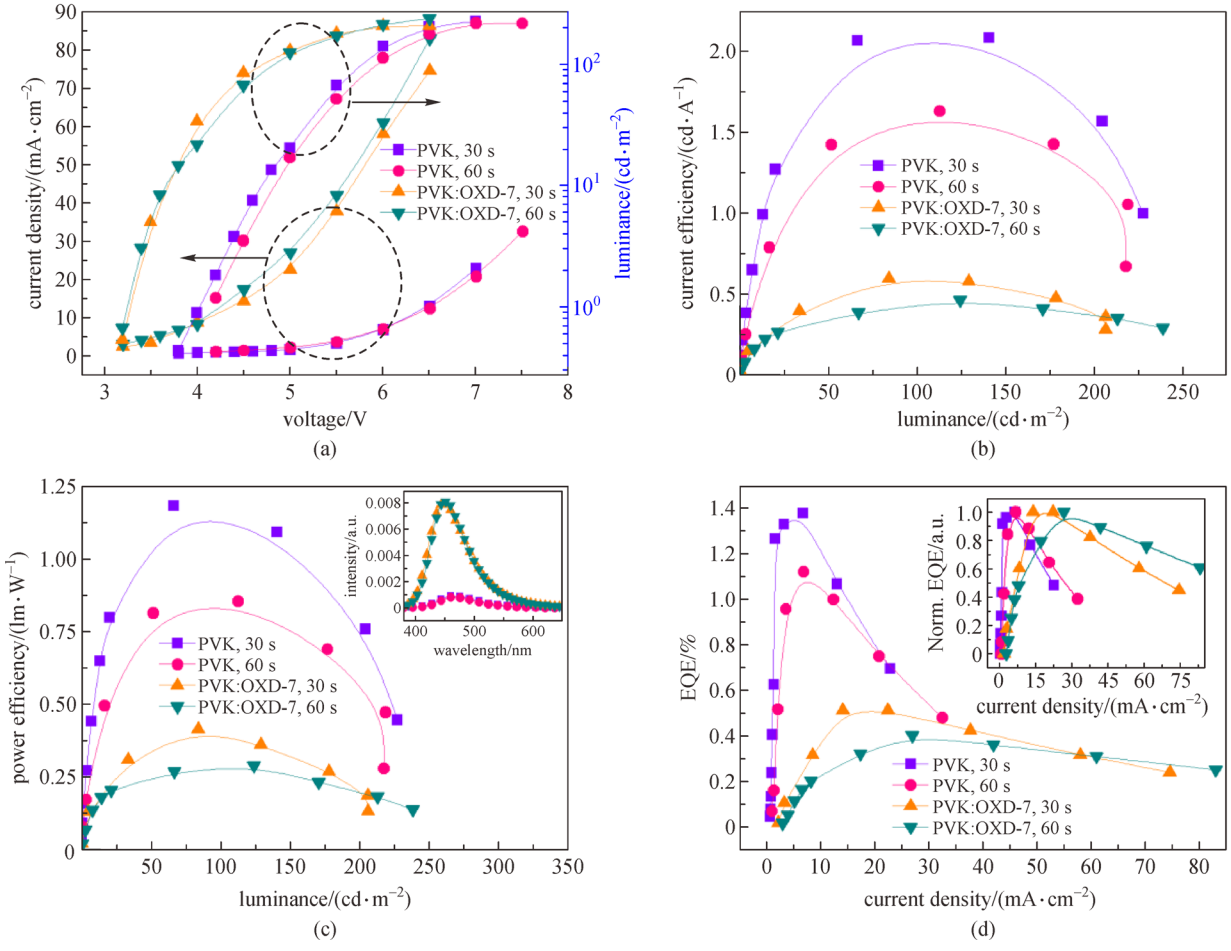

Fig. 3 EL characteristics of the TADF devices with PVK:DMAC-DPS or PVK:OXD-7:DMAC-DPS EML by spin-coating at 3000 r/min for 30 s or 60 s and annealing at 80°C for 30 min. (a) Current density-voltage-luminance (*J-V-L*). (b) Current efficiency (CE)-luminance. (c) Power efficiency (PE)-luminance, inset of (c) is the corresponding EL spectra. (d) EQE-current density, inset of (d) is the normalized EQE spectra

80°C are more drastic than those at 100°C, likely ascribed to the rougher film surface. As indicated in Fig. 2, for both single host and co-host EML films, the RMS roughness at 80°C is larger than that at 100°C. The rougher morphology causes partial short circuits or poor electrical contact in the thin OLED device, resulting in poorer performance and serious efficiency roll-off [26].

Besides, impedance spectroscopy, usually characterized by impedance versus frequency (*Z-f*), phase angle versus frequency (φ-*f*), and impedance versus bias voltage (*Z-V*), has gradually become an effective tool to investigate the electrical properties at the interface of organic layers for OLEDs [27,28]. We comparatively investigated the impedance spectroscopy for single host PVK, as well as OXD-7, and co-host PVK:OXD-7-based devices. Figure 5 illustrates the *Z-f* and φ-*f* characteristics for the three

Table 2 EL characteristics of ITO/PEDOT:PSS (30 nm)/PVK:OXD-7:DMAC-DPS (6:3:1, wt%, 20 nm)/DPEPO (10 nm)/Bphen (30 nm)/LiF (0.8 nm)/Al (80 nm) at various annealing temperatures (T) for the EML by spin-coating at 2000 r/min for 30 s

T/°C	L_{max}/(cd·m^{-2})	$V_{turn\text{-}on}$[a]/V	CE_{max}/(cd·cm^{-2})	PE_{max}/(lm·W^{-1})	EQE_{max}/%
80	36.5	7.5	3.15	1.17	1.41
100	243	5.8	2.32	0.82	1.31

Notes: a, measured at 1.0 cd/m^2; L_{max} is the maximum luminance; $V_{turn\text{-}on}$ is turn-on voltage; CE_{max} is the maximum current efficiency; PE_{max} is the maximum power efficiency; EQE_{max} is the maximum external quantum efficiency.

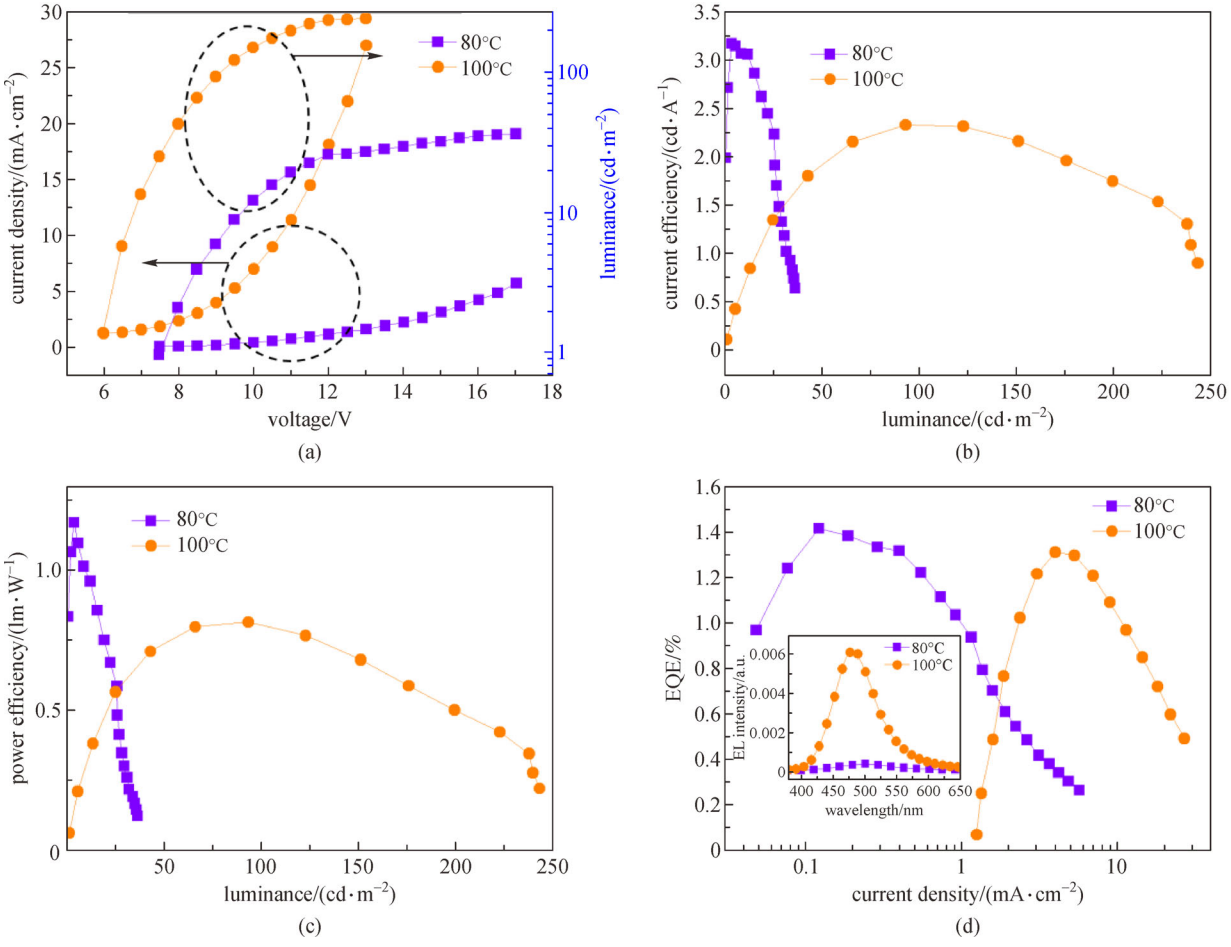

Fig. 4 EL characteristics of TADF devices with PVK:OXD-7:DMAC-DPS EML via spin-coating at 2000 r/min for 30 s. (a) Current density-voltage-luminance (*J-V-L*). (b) Current efficiency (CE)-luminance. (c) Power efficiency (PE)-luminance. (d) EQE-current density, inset of (d) is the corresponding EL spectra

devices without applying bias. It is obvious that the impedance gap among the three devices is large at frequencies below 40 kHz. The impedance values at 1000 Hz are 8.6×10^3, 4.2×10^3, and 2.7×10^3 Ω for PVK, PVK:OXD-7, and OXD-7 host-based devices, respectively, indicating that the doping of OXD-7 significantly reduces device resistance.

Host-guest interplay is crucial for achieving high efficiency in most OLED devices. It has been reported that the effects of guest interactions and guest ionization potential on charge transport, luminance, and carrier mobility are markedly different [29], where the single host PVK and bipolar co-host of PVK:OXD-7 significantly influence the guest interaction and molecule overlapping and thus, the device performance is different.

4 Conclusions

In conclusion, we have illustrated a solution-processed TADF OLED using blue-emitting small molecule DMAC-DPS as the emitter. By comparative investigations, the effects of PVK and a co-host of PVK:OXD-7 on the device performance determined that the co-host device showed

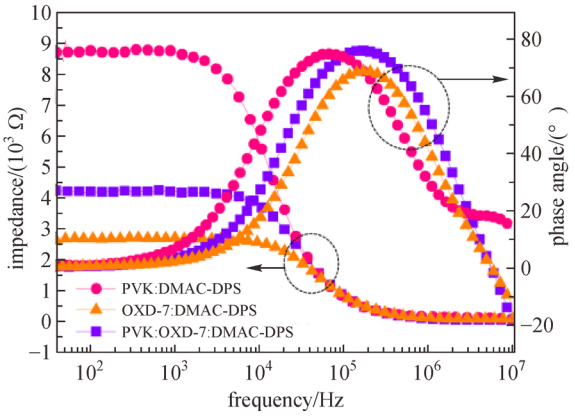

Fig. 5 Impedance spectroscopy characteristics (*Z-f* and *φ-f*) for the TADF OLEDs with hosts of PVK:OXD-7, PVK, and OXD-7, respectively

lower turn-on voltage, similar maximum luminance, and slower EQE roll-off, suggesting that doping OXD-7 into PVK improves device stability. Simultaneously, it significantly reduces device impedance from 8.6×10^3 to 4.2×10^3 Ω at 1000 Hz. By adjusting annealing temperature, the device stability of the co-host device was also significantly improved.

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