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White Organic Light-Emitting Diodes with Thermally Activated Delayed Fluorescence Emitters

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Abstract

Recently, thermally activated delayed fluorescence (TADF) organic light-emitting diodes (OLEDs) have attracted both academic and industrial interest due to their extraordinary characteristics, such as high efficiency, low driving voltage, bright luminance, lower power consumption, and potentially long lifetime. In this chapter, various approaches to realize white OLEDs (WOLEDs) with TADF emitters have been introduced. The recent development of WOLEDs based on all TADF emitters, WOLEDs based on TADF and conventional fluorescence emitters, and WOLEDs based on TADF and phosphorescence emitters is highlighted. Particularly, the device structures, design strategies, working mechanisms, and electroluminescent processes of the representative high-performance WOLEDs with TADF emitters are reviewed. Moreover, challenges and opportunities for further enhancement of the performance of WOLEDs with TADF emitters are presented.

Keywords: white, organic light-emitting diodes, thermally activated delayed fluorescence, lighting, display

1. Introduction

In 1987, Tang and his coworker reported the first organic light-emitting diodes (OLED) [1]. Since then, OLEDs have been the object of intense research because of the superior properties, including bright luminance, excellent efficiency, fast response, good stability as well as the flexible characteristic [2–5]. Currently, there are a large number of available commercial applications in our life, such as televisions, lamps as well as cell phones. In addition, after the invention of OLED, other kinds of LEDs (e.g., polymer LED, quantum-dot LED, nanoplatelet

LED, and perovskite LED) were also reported [6–10]. By dint of the strategies used in OLEDs, the performance of these LEDs can be greatly enhanced. Besides, with the increasing understanding of the insight of OLEDs, the concepts utilized in OLEDs can also be applied to other optoelectrical devices, which is beneficial to the development of related fields [11–13].

To satisfy the requirements of energy-saving lighting and high-quality displays, white OLED (WOLED) has been considered to be one of the most promising candidates. Since the pioneer WOLEDs made by Kido and his coworkers, the WOLED technology is greatly improved [14, 15]. During the last 24 years, the power efficiency (PE) was enhanced from less than 1 lm/W to more than 100 lm/W [16–18], indicating that WOLEDs are promising for the lighting and displays filed. In terms of lighting application, WOLEDs require a standard fluorescent tube efficiency (40–70 lm/W) and $\geq 10,000$ h of lifetime at the luminance of ≥ 1000 cd/m² [19–21]. Besides, the color rendering index (CRI) above 80 is required for the indoor lighting, and the Commission International de L'Eclairage (CIE) chromaticity coordinates of WOLEDs should be located near white light equal-energy point (0.33, 0.33) [22–25]. Moreover, for the high-quality lighting, other characterization parameters (e.g., correlated color temperature (CCT), color stability, and driving voltage) of WOLEDs are also required to be taken into account [26–30].

In 2012, Adachi et al. made a breakthrough on the thermally activated delayed fluorescence (TADF) material, which has been considered as the third-generation OLED emitter [31–33]. Different from the conventional fluorescent emitters in which only the singlet excitons (25%) can emit light since the radiative decay of triplet excitons (75%) is spin forbidden, TADF emitters could harness both singlet and triplet excitons since triplet excitons can be harvested as delayed fluorescence through their upconversion from a lowest triplet state to a lowest singlet state by inducing an efficient reverse intersystem crossing (RISC) [34–38]. Therefore, similar to phosphorescence emitters, a maximum internal quantum efficiency (IQE) of 100% can be realized [39–42]. Due to the outstanding properties (e.g., free noble metal, high efficiency, low driving voltage, bright luminance, lower power consumption, and potentially long lifetime), TADF emitters have been actively investigated to develop WOLEDs [43–45]. Although efficient TADF emitters were only demonstrated 6 years ago and WOLEDs with TADF emitters were just reported 4 years ago, the performance of WOLEDs with TADF emitters has been improved step by step [46]. For example, WOLEDs with TADF emitters can exhibit nearly 20% external quantum efficiency (EQE) [47], which is comparable to state-of-the-art phosphorescence WOLEDs and fluorescence/phosphorescence hybrid WOLEDs [29, 30, 48–54]. Thus, WOLEDs with all TADF emitters have great potential to the lighting and display field.

Herein, we first introduced the basic concepts of TADF emitters, which are beneficial to comprehend WOLEDs with TADF emitters. Then, we summarize the main approaches to realize WOLEDs with TADF emitters in recent years. More specifically, we highlight the recent development of WOLEDs based on all TADF emitters, WOLEDs based on TADF and conventional fluorescence emitters, and WOLEDs based on TADF and phosphorescence emitters. Particularly, the device structures, design strategies, working mechanisms, and electroluminescent processes of the representative high-performance WOLEDs with TADF emitters are

reviewed. Finally, challenges and opportunities for further enhancement of the performance of WOLEDs with TADF emitters are presented.

2. Fundamental concepts of TADF emitters

Due to the effect of spin statistics, when holes injected from the anode meet electrons injected from the cathode, singlet and triplet excitons will be formed with a ratio of 1:3 [55]. In the case of the first-generation OLED emitters (i.e., conventional fluorescence emitters), only the singlet excitons (25%) can emit light since the radiative decay of triplet excitons (75%) is spin forbidden, as shown in **Figure 1**. Therefore, the EQE of fluorescence WOLEDs is usually below 5%, considering that the outcoupling factor is ~20%. For the second-generation OLED emitters (i.e., phosphorescence emitters), they can not only harvest triplet excitons via the triplet-triplet energy transfer but also harvest singlet excitons via the singlet-triplet intersystem crossing process (ISC) due to the heavy-atom effect [56]. Thus, the EQE of phosphorescence WOLEDs can be as high as 20%.

In terms of the third-generation OLED emitters (i.e., TADF emitters), a small energy gap (ΔE_{ST}) between singlet (S_1) and triplet (T_1) excited states is required and can be attained by carefully designing organic molecules [31]. Generally, the S_1 level was considerably higher in energy than the T_1 level by 0.5–1.0 eV, because of the electron exchange energy between these

Figure 1. An energy diagram of a conventional organic molecule. H is hole and e is electron. Reproduced from Ref. [31].

levels. However, to enhance thermal upconversion (i.e., $T_1 \rightarrow S_1$ RISC), the molecular design of TADF materials requires small ΔE_{ST} , typically less than 0.2 eV, to overcome competitive non-radiative decay pathways, leading to highly luminescent TADF materials [57]. In addition, to enhance the photoluminescence efficiency of TADF materials, the geometrical change in molecular conformation between its ground state (S_0) and S_1 states should be restrained to suppress non-radiative decay. As a result, the maximum theoretical IQE of TADF emitters can be 100%.

3. Approaches to realize WOLEDs with TADF emitters

3.1. Basic aspects of WOLEDs with TADF emitters

Similar to phosphorescence emitters, the use of TADF emitters is very promising to achieve WOLEDs. This is because TADF emitters can (1) harness triplet excitons, (2) exhibit excellent efficiency, and (3) show usually broad emission spectra with rather large full width at half maximum of about 100 nm, which is wider than that of conventional fluorescent materials because of their charge-transfer nature [31–47].

To attain the high performance, the device structures, design strategies, working mechanisms, and electroluminescent processes of the WOLEDs with TADF emitters should be well manipulated. For example, unlike conventional fluorescence emitters, the T_1 of TADF emitters is necessary to be considered when designing a WOLED architecture, since hosts or nearby layers with low T_1 would quench the triplet excitons, which leads to the low efficiency. Besides, the location of TADF emitters is needed to be investigated, since the energy transfer would occur between the contacted different emitters (e.g., energy can transfer from high-energy TADF emitters to low-energy emitters). To date, various approaches have been reported to develop WOLEDs with TADF emitters, such as the exploitation of all TADF emitters, the combination of TADF and conventional fluorescence emitters, and the mixture of TADF and phosphorescence emitters.

3.2. WOLEDs with all TADF emitters

The most directed approach to develop WOLEDs with TADF emitters is the exploitation of all TADF emitters. That is to say, all blue, green, and red emitters are TADF materials. By selecting high- T_1 hosts and charge transport materials, high-performance WOLEDs can be attained. More specifically, the conventional fluorescent host should possess high T_1 , which should be particularly higher than the blue TADF materials. Otherwise, the triplet excitons of emitters would be quenched by the host, resulting in low performance. Moreover, high- T_1 charge transport materials should be selected, which is used to confine the triplet excitons in the EML, leading to the excitons being well consumed.

Adachi and his coworkers for the first time adopted such an approach to develop highly efficient WOLEDs with all TADF emitters [46]. **Figure 2** depicts the device structure: ITO/1,4,5,8,9,11-hexaazatriphenylene hexacarbonitrile (HAT-CN, 10 nm)/9,9',9''-triphenyl-9H,9'H,9''H-3,3':6',3''-tercarbazole (Tris-PCz, 35 nm)/10 wt% 1,2,3,4-tetrakis(carbazol-9-yl)-5,6-dicyanobenzene

Figure 2. The WOLED structures and energy level diagram. Reproduced from Ref. [46].

(4CzPN): 3,3-Di(9H-carbazol-9-yl)biphenyl (mCBP, green EML) (x nm)/6 wt% 4CzPN: 2 wt% 41,4-dicyano-2,3,5,6-tetrakis (3,6-diphenylcarbazol-9-yl)benzene (CzTPN-Ph): mCBP (red EML) (y nm)/10 wt% 9-(3-(9H-carbazol-9-yl)-9-(4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-9H-carbazol-6-yl)-9H-carbazole (3CzTRZ): 2,8-bis(diphenylphosphoryl) dibenzo-[b,d] thiophene (PPT, blue EML) (z nm)/PPT (50 nm)/LiF/Al. In this device, the T₁ level of mCBP host is 2.9 eV, which is much higher than that of blue, green, and red TADF emitters, ensuring the high efficiency. Besides, PPT has a high T₁ of 3.1 eV, suggesting a good confinement of the triplet excitons. By optimizing the charge generation zone via the adjustment of different EML thickness (the total thickness is set to be x + y + z = 15 nm), the WOLED achieved a maximum EQE of over 17%, a peak PE of 34.1 lm/W with CIE coordinates of (0.30, 0.38).

3.3. WOLEDs with TADF and conventional fluorescence emitters

Considering that triplets could be harnessed by TADF emitters, it is promising to realize the unity IQE by combining traditional fluorescent materials with TADF emitters [61]. For such an approach, the TADF molecules would act as the triplet harvester, which is used to harness the triplets for the conventional other-color fluorescence materials. As a result, highly efficient white light can be produced [42–44].

In 2016, Li et al. reported high-efficiency and high CRI WOLEDs with the chromaticity-adjustable yellow TADF emitter 2-(4-phenoxazinephenyl)thianthrene-9,9',10,10'-tetraoxide

(PXZDSO2) [47]. By combining the conventional deep-blue fluorescence emitter NI-1-PhTPA and PXZDSO2, the two-color WOLED showed a maximum EQE of 15.8% (device W3). Then, since the chromaticity of the EML containing PXZDSO2 could be tuned to yellowish green, they introduced a deep-red fluorescence emitter DBP (dibenzo[*f,f'*]-4,4',7,7'-tetraphenyl]diindeno[1,2,3-*cd*:1',2',3'-*lm*]perylene) subtly to fabricate three-color WOLED, achieving the most efficient ever EQE of 19.2% with a CRI of 68 (device W4) and the highest ever CRI of 95 with an EQE of 15.6% (device W6). The configurations are ITO/HATCN/TAPC/EMLs/TmPyPB/LiF/Al, in which device W3 has the EML of CBP: 8 wt% NI-1-PhTPA (10 nm)/CBP (3 nm)/CBP: 6 wt% PXZDSO2 (15 nm)/CBP (3 nm)/CBP: 8 wt% NI-1-PhTPA (10 nm), device W4 has the EML of CBP: 7 wt% NI-1-PhTPA (10 nm)/CBP (3 nm)/CBP: 3 wt% PXZDSO2 (5 nm)/CBP: 5 wt% PXZDSO2: 0.3 wt% DBP (5 nm)/CBP: 3 wt% PXZDSO2 (5 nm)/CBP (3 nm)/CBP: 7 wt% NI-1-PhTPA (10 nm), device W6 has the EML of CBP: 10 wt% NI-1-PhTPA (10 nm)/CBP (3 nm)/CBP: 5 wt% PXZDSO2: 0.35 wt% DBP (15 nm)/CBP (3 nm)/CBP: 10 wt% NI-1-PhTPA (10 nm). The device working mechanisms can be described as follows. For device W3, (1) since NI-1-PhTPA is a deep-blue fluorescence emitter and CBP: 6 wt% PXZDSO2 emits a yellow light with broad spectrum, high-performance two-color WOLEDs were realized; (2) given the almost equal T_1 level of NI-1-PhTPA and PXZDSO2, the efficiency roll-off occurs if they are directly in contact due to the triplet exciton quenching by NI-1-PhTPA; (3) the efficiency roll-off can be further induced as the formed triplet excitons of NI-1-PhTPA cannot be utilized by PXZDSO2; (4) to stabilize the recombination zone which occurs in whole EMLs since NI-1-PhTPA/CBP are bipolar and avoid triplet exciton quenching by NI-1-PhTPA, two 3-nm CBPs were inserted between the blue and yellow EMLs, restraining the inevitable Förster

Figure 3. Function mechanisms of the use of singlets/triplets. (a) Device W3 and (b) devices W4 and W6. H, B, Y, and R represent CBP, NI-1-PhTPA, PXZDSO2, and DBP, respectively. Reproduced from Ref. [47].

energy transfer from NI-1-PhTPA to PXZDSO₂; (5) to reduce the triplet exciton energy loss via nonradiative transition process, blue-fluorescence emitter was dispersed in CBP for blue emission, leading to most excitons being generated at CBP; and (6) triplet energy transferred from CBP gives most of triplet excitons of PXZDSO₂ since triplet excitons typically have long diffusion lengths (≈ 100 nm), as shown in **Figure 3a**. Hence, an EQE of 15.8% was achieved for device W3. For device W4, (1) a deep-red fluorescence emitter DBP was conceived to be used; (2) PXZDSO₂ was an assistant host for DBP to realize a red-light emission due to an efficient energy transfer from the S1 of PXZDSO₂; (3) the doping concentration of PXZDSO₂ was decreased to reduce intermolecular aggregation and thus blue-shifted emission (20 nm), achieving green emission, complementary to emissions of NI-1-PhTPA and DBP; (4) a red EML of CBP: 5 wt% PXZDSO₂: 0.3 wt% DBP was inserted between two green EMLs of CBP: 3 wt% PXZDSO₂ to receive singlet exciton energy transferred from the PXZDSO₂ molecules in both sides to give both green and red emissions; (5) the two doped blue EMLs and CBP interlayers located at both sides of the green EMLs to give a blue emission and to confine the PXZDSO₂ triplet excitons, respectively, as shown in **Figure 3b**. Thus, an EQE of 19.2% was achieved for device W4. Furthermore, an EML consisting of improved DBP doping concentration was utilized instead of the green and red EMLs for candle-style warm WOLEDs (device W6), achieving a high CRI of 95.

3.4. WOLEDs with TADF and phosphorescence emitters

Since both TADF and phosphorescence emitters can harvest singlet and triplet excitons, the mixture of TADF and phosphorescence emitters is a significant approach to construct WOLEDs. By virtue of their respective advantages, high efficiency and long lifetime can be realized simultaneously [44, 58]. In particular, there is tremendous interest in mixing blue TADF emitters with green/red or complementary color phosphorescence emitters. This is because blue TADF materials (1) are naturally advantageous to achieve high triplet energies due to their reduced singlet-triplet splits, (2) can possess high efficiency, and (3) can harvest the triplets [59–61]. For this kind of WOLED (i.e., mixing blue TADF and other-color phosphorescence emitters), it is generally called hybrid WOLEDs [21].

In 2014, Zhang and coworkers demonstrated hybrid WOLEDs via the use of blue TADF emitter, obtaining the peak efficiency as high as 47.6 lm/W [59]. The device structure is ITO/HATCN (5 nm)/NPB (40 nm)/TCTA (10 nm)/mCP: 4,5-bis(carbazol-9-yl)-1,2-dicyanobenzene (2CzPN, 11 nm, blue EML)/TAZ: 4 wt% (acetylacetonato)bis[2-(thieno[3,2-c]pyridin-4-yl)phenyl]iridium(III) (PO-01, 4 nm, orange EML)/TAZ (40 nm)/LiF (0.5 nm)/Al (150 nm), as shown in **Figure 4**. The factors for the high performance are as follows: (1) mCP is chosen to be the host for 2CzPN due to the wide energy gap and high T_1 of 3.0 eV, since the host of the TADF material plays an important role in determining the efficiency; (2) 2CzPN is placed nearest to the main recombination zone, ensuring that excitons can diffuse throughout the emissive region to produce a desired color-balanced output; (3) triplets formed on 2CzPN can be harvested by either energy transfer to the low-lying triplet states of the phosphor PO-01 (2.2 eV) or thermal upconversion to the emissive singlet states, eliminating the energy loss; and (4) the recombination zone is fixed as the voltage increases by 2CzPN due to its charge-trapping ability, achieving a stable white emission.

Figure 4 Schematic diagrams of the working mechanisms. The gray-filled rectangle represents the main exciton generation zone. PF is the prompt fluorescence while DF is the delayed fluorescence. RISC indicates the reverse ISC and ET denotes the energy transfer. Reproduced from Ref. [59] with permission from the Royal Society of Chemistry.

Although high-performance hybrid WOLEDs based on TADF materials have been demonstrated, there are still some problems, even for these state-of-the-art devices [59–61]. For example, (1) the driving voltages are somewhat high (e.g., 3.2 V at 1 cd m⁻² [61]); (2) the luminances are very low (e.g., only ~10,000 cd m⁻² [61]); (3) the efficiency at high luminance is not high (e.g., <6 lm W⁻¹ at 10,000 cd m⁻² [61]); (4) the CRI is not high enough; and (5) negligible attention has been paid to the lifetime of TADF-based hybrid WOLED.

To solve the issues, Luo et al. recently reported high-performance two-color and three-color hybrid WOLEDs [62]. The two-color WOLED exhibits (1) low voltage (i.e., 2.9 V at 1 cd m⁻²); (2) high luminance (103,756 cd m⁻²); (3) maximum EQE and PE of 23.5% and 70.92 lm W⁻¹, respectively; and (4) 21.59 lm W⁻¹ at 10,000 cd m⁻². The three-color WOLED exhibits (1) low voltage and high luminance (51,514 cd m⁻²); (2) superior CRI of 94; and (3) EQE and PE of 17.3% and 46.09 lm W⁻¹, respectively. The configuration of the two-color WOLEDs is ITO/HAT-CN (100 nm)/TAPC (20 nm)/mCP: 9,9',9'',9'''-((6-phenyl-1,3,5-triazine-2,4-diyl) bis(benzene-5,3,1-triyl))tetrakis(9*H*-carbazole) (DDCzTrz, 10 nm, 20%)/interlayers (3 nm)/bis[2-(2-hydroxyphenyl)-pyridine] beryllium (Bepp₂): bis(2-phenyl-4,5-dimethylpyridinato) [2-(biphenyl-3-yl)pyridinato] iridium(III) (Ir(dmp₂py)₂(dpp), 15 nm, 1:2%)/Bepp₂ (35 nm)/

LiF (1 nm)/Al (160 nm), where interlayers are none, mCP and 2,6-bis(3-(carbazol-9-yl)phenyl)pyridine (26DCzPPy) for devices W11, W12, and W13, respectively. The configuration of the two-color WOLED is ITO/HAT-CN (100 nm)/TAPC (20 nm)/mCP: DDCzTrz (10 nm, 20%)/26DCzPPy (interlayer, 3 nm)/Bepp₂: Ir(dmpy)₂(dpp): Ir(piq)₃ (15 nm, 1: 2%: 1.3%)/Bepp₂ (35 nm)/LiF (1 nm)/Al (160 nm). Unlike previous TADF-based hybrid WOLEDs, the bipolar interlayer is demonstrated to enhance the performance. Particularly, it is demonstrated that the use of interlayer can enhance the lifetime (2.3 times). The working mechanism of the two-color WOLED can be described as follows, which is beneficial to comprehend the reason why the bipolar interlayer can enhance the performance. For W11, since mCP and Bepp₂ are p-type and n-type materials, respectively, holes and electrons are easily accumulated at the mCP/Bepp₂ interface, forming singlet and triplet excitons, as shown in **Figure 5a**. The triplets on blue EML can (1) convert into singlets via the RISC procedure and then generate the blue emission, and (2) transfer to the low energy of yellow phosphor Ir(dmpy)₂(dpp) via the Dexter process and then generate part of yellow emission (the other part of yellow emission is originated from excitons on the yellow EML). However, the main exciton generation

Figure 5 A schematic illustration of the working mechanism of (a) W11, (b) W12, and (c) W13. The gray-filled rectangles are the main exciton generation zones. The Dexter energy transfer can occur in W11, while it is prohibited in both W12 and W13. Reproduced from Ref. [62].

zone of W11 is narrow, unfavorable to the performance. Similarly, the main exciton generation zone of W12 is located at the mCP interlayer/Bepp₂ interface, as shown in **Figure 5b**. As a result, excitons are more easily harvested by Ir(dmppy)₂(dpp) instead of DDCzTrz since Ir(dmppy)₂(dpp) is close to the main exciton generation zone. However, a part of electrons can pass through the thin interlayer via the tunneling process and then meet holes, which can generate excitons to guarantee the blue emission. For W13, by way of the bipolar interlayer and the suitable energy levels of 26DCzPPy, both holes and electrons can be easily passed through 26DCzPPy, as shown in **Figure 5c**. As a result, excitons can be formed at both the mCP/26DCzPPy and 26DCzPPy/Bepp₂ interfaces, leading to a broad exciton generation zone, which ensure the high performance of W23. Besides, since the Dexter energy transfer from DDCzTrz to Ir(dmppy)₂(dpp) is also prevented due to the 3 nm 26DCzPPy, the yellow emission mainly results from excitons on the yellow EML.

Another effective approach to develop WOLEDs with TADF and phosphorescence emitters is the mixture of green TADF and other-color phosphorescence emitters. In this case, the TADF emitters are adopted as the emitters for WOLEDs because they may be compatible with phosphorescence emitters and not quench triplet excitons of the phosphorescence emitters, otherwise triplet excitons will be wasted.

Kim et al. reported this approach by combining a green TADF with red/blue phosphorescence materials to organize high-efficiency hybrid-type WOLEDs [45]. In their WOLED, energy transfer between a blue phosphorescent material and a green TADF emitter was efficient and could be managed by controlling the doping concentration of emitters. A maximum EQE of 20.2% was achieved by optimizing the device structure of the hybrid-type WOLEDs. The device structure is ITO (50 nm)/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, 60 nm)/TAPC (20 nm)/mCP (10 nm)/mCP: iridium(III) bis[(4,6-difluorophenyl)-pyridinato-N,C²]picolate (FIrpic): (4 s,6 s)-2,4,5,6-tetra(9H-carbazol-9-yl)isophthalonitrile (4CzIPN) (12.5 nm)/TPBI: Ir(pq)₂acac (12.5 nm, 3%)/diphenylphosphine oxide-4-(triphenylsilyl)phenyl (TSPO1, 35 nm)/LiF (1 nm)/Al (200 nm), where 4CzIPN is the green TADF emitter, and FIrpic and Ir(pq)₂acac are blue and red phosphorescence emitter, respectively, as shown in **Figure 6**. To explore the possibility of this type of WOLEDs, hybrid OLEDs with blue-emitting FIrpic and green-emitting 4CzIPN were first fabricated. By optimizing the concentration of FIrpic and 4CzIPN, a maximum EQE of the hybrid OLEDs was 19.2% at 5% FIrpic and 0.5% 4CzIPN. Given that the EQE of mCP: FIrpic OLED is <20%, such superior EQE of hybrid OLED suggests that 4CzIPN would not quench FIrpic triplet emission. In fact, T₁ of FIrpic can be transferred to 4CzIPN and then make a contribution to the 4CzIPN TADF emission. For this hybrid OLED, there are three main energy transfer processes, that is, energy transfer processes from mCP to FIrpic, mCP to 4CzIPN, and FIrpic to 4CzIPN dominate the blue and green emissions of the hybrid OLEDs. After the successful exploration of hybrid OLED, Kim et al. combined this system and red phosphorescence emitting layers, attaining high-efficiency WOLEDs. The factors for the high-performance of WOLEDs can be summarized as follows: (1) the hybrid OLEDs doped with FIrpic and 4CzIPN showed a high quantum efficiency, which ensure the high efficiency of blue-green-emitting layer. In this emitting layer, energy transfer from FIrpic to 4CzIPN is

Figure 6. The energy level diagram and device architecture of the WOLED. Reproduced from Ref. [45].

efficient, leading to no non-radiative triplet exciton quenching of FIrpic by 4CzIPN. (2) The energy transfer from mCP to dopant materials in the FIrpic and 4CzIPN co-doped emitting layer is very efficient. (3) FIrpic activates the delayed emission of 4CzIPN through an efficient energy transfer, which resulted in the high quantum efficiency of the hybrid-emitting layer. (4) The balanced charge density in the emitting layer contributed to the high quantum efficiency. The TPBI: Ir(pq)₂acac-emitting layer efficiently injects electrons due to electron transport properties of TPBI, which improved charge balance in the emitting layer in combination with hole transport-type mCP: FIrpic: 4CzIPN-emitting layer.

4. Summary and outlook

As a novel kind of OLED emitter, TADF materials show many unique characteristics, which have been demonstrated to develop high-performance WOLEDs. Thanks to the hard endeavors of researchers, the performance of WOLEDs is now comparable to state-of-the-art phosphorescence WOLEDs and fluorescence/phosphorescence hybrid WOLEDs. In this chapter, the focus is the development of WOLEDs by manipulating TADF emitters. Specifically, we highlight the recent development of WOLEDs based on all TADF emitters, WOLEDs based on TADF and conventional fluorescence emitters, and WOLEDs based on TADF and phosphorescence emitters. Particularly, the device structures, design strategies, working mechanisms, and electroluminescent processes of the representative high-performance WOLEDs with TADF emitters are reviewed.

Although the performance of WOLEDs with TADF emitters has been enhanced over the past few years, there are still many challenges before they can be large-scale commercialized production, such as the efficiency, lifetime, and cost. However, it is deserved to point out that these issues are also hindrances for other kinds of WOLEDs. For example, there is still much room for the efficiency of WOLEDs to the theoretical limit of 248 lm/W (standard light source (D65) from 400 to 700 nm wavelength) [63]. Therefore, the photoluminescence quantum efficiency of the emissive materials (TADF or other-type emitters), the charge balance, and outcoupling efficiency of the devices should be further enhanced.

Besides, despite the efficiency of WOLEDs with TADF emitters that can be high enough, there are some other parameters that are needed to be enhanced. Particularly, the stability and efficiency roll-off of WOLEDs with TADF emitters still lags behind other kinds of WOLEDs. For example, fluorescence WOLEDs can show an extremely long lifetime of 150,000 h at an initial luminance of 1000 cd/m² [64], while hybrid WOLEDs based on conventional blue fluorescence emitters can possess a long lifetime of >30,000 h at 1000 cd/m² [65]. However, it is still difficult for WOLEDs with TADF emitters to achieve long lifetime, which may be attributed to the instability of TADF emitters. For example, Wang et al. recently reported the first WOLED with TADF emitters realizing long lifetime (2025 h at 1000 cd/m²) [66]. However, it is noted that the lifetime still cannot meet the requirement of large-scale commercialized productions. Hence, to solve this issue, stable TADF emitters are urgently explored [57]. In addition, fluorescence WOLEDs, hybrid WOLEDs based on conventional blue fluorescence emitters or even phosphorescence WOLEDs with extreme color stability ($\Delta\text{CIE} = (0.00, 0.00)$) in the whole luminance/driving voltage have been reported [28, 67–70]. However, the color stability of WOLEDs with TADF emitters is usually unstable, indicating that more efforts are required to manage this difficulty. Furthermore, the efficiency roll-off in WOLEDs with TADF emitters is not ideal, particularly for the PE roll-off [71]. As a consequence, only low efficiency can be attained at high luminances, which is not beneficial to the practical applications. To loosen this bottleneck, the charge balance, energy barriers between nearby layers, and materials selection should be well manipulated [72–76]. With the endeavor of academic and industrial researchers to enhance the materials design and device engineering, we believe that WOLEDs with TADF emitters can play a significant role in the marketplace in the near future, which is beneficial to our human society.

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Conflict of interest

The authors declare no competing financial interest.

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