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The Influence of Energy Transfer on the Color Temperature Change in Color-Tunable Organic Light Emitting Diodes with Interface Exciplex

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Abstract

The color-tunable white organic light-emitting diode (CT-WOLED) with wide correlation color temperature (CCT) has many advantages in optimizing the artificial light source to adapt to the human physiological cycle. The research on the change trend of CCT and the law of extending the change range of CCT will help to further improve the performance of this kind of device. The present work fabricated a series of CT-WOLED devices with a simple structure, which are all composed of two ultra-thin phosphor layers (PO-01 and Flrpic) and a spacer interlayer. The yellow interface exciplex (TCTA/PO-T2T) formed between the spacer layer (PO-T2T) and transmission material (TCTA) in EML will decrease the CCT value at low voltage. The relationship between the energy transfer in EML and CCT change trend is investigated by adjusting the interface exciplexes and the thickness of the interlayer or the phosphor layer in devices A, B and C, respectively. The results demonstrate that a simple OLED device with an interlayer inserted between two ultra-thin phosphor layers can achieve a wider CCT span from 3359 K to 6451 K at voltage increases from 2.75 V to 8.25 V.

Keywords

Interface Exciplex, Energy Transfer, Color-Tunable, WOLED

1. Introduction

In recent years, the organic light-emitting diode (OLED) has attracted many attentions of researchers and made significant progress due to its excellent performance, such as high luminance, light weight, bright colors [1] [2] [3]. On the one hand, OLED with stable spectrum can prevent the color distortion of the display device and greatly enhance the user's visual experience in the field of flat panel display [4] [5] [6] [7] [8]. On the other hand, the color-tunable OLED (CT-OLED) with unstable spectrum has also important application in the field of lighting industry. The color-tunable CT-OLED with a wide correlation color temperature (CCT) can simulate the change of sunlight, which is consistent with the human physiological cycle. In some specific occasions, such as military and medical fields, the demand for the high-performance CT-OLED is very necessary and urgent [9]-[15].

The CCT and the CCT span are very important parameters to CT-OLED. In order to explore the change rule of CCT value and further expand the change range of CCT, researchers have conducted extensive research. Zhao *et al.* investigate the change rule of spectrum and color coordinates in tandem OLED (TOLED) with different color vertical stacking structures by controlling the relative position of the light-emitting units. The maximum current efficiency of 20.4 cd/A is achieved, and the CIE coordinate changes from (0.63, 0.31) to (0.34, 0.27) when the voltage increases from 10 V to 22 V [16]. Zhang *et al.* develop a full-color tunable TOLED with an external quantum efficiency of up to 26.02% by using indium zinc oxide intermediate connection electrode to stack yellow quantum-dot LED with blue organic materials [17]. Although this kind of method is effective for the change of CCT value, TOLED involves a relatively complex structure and requires a higher driving voltage [16] [17] [18]. It is also a method to realize CCT adjustment by controlling the change of recombination zone (RZ) with voltage [18] [19] [20] [21]. Ying *et al.* demonstrate high efficiency CT-OLED by combining non-doped ultra-thin phosphorescent emitters with a blue phosphorescence-doped exciplex system. The resultant devices deliver a sunlight-style emission with a wide CCT span from 2143 to 7563 K and achieve the maximum current efficiency of 34.4 cd/A [19]. However, the role of blue exciplex in the device has not been discussed in detail by researchers. For CT-OLED devices with simple structure, the mechanism of how to further expand the range of CCT remains to be further clarified.

In this paper, we have investigated the changes of CCT in a simple OLED device with an interlayer inserted between two ultra-thin phosphor layers at low voltage and high voltage, respectively. At the low voltage, we plan to reduce CCT value by changing the material of interlayer (TPBi, PO-T2T or TPBi: PO-T2T) to form different interface exciplexes (TCTA/TPBi or TCTA/PO-T2T) in the light emission layer (EML), thus affecting the energy distribution in EML. At high voltage, we plan to increase the CCT value by increasing the thickness of the interlayer or by reducing the thickness of the yellow phosphor layer, because both methods can significantly reduce the yellow phosphor spectral intensity. Due to the use of interface exciplex, the designed CT-OLED achieves a wide CCT range only with a thin EML layer thickness. The optimized CT-OLED device achieves a maximum CCT change range (ΔCCT) of 2894 K. At the same time, the influence of interface exciplexes on the performance of CT-OLED is also discussed in detail.

2. Materials and Methods

All devices are fabricated on glass substrates with square surface resistance (15 Ω/sq). Indium tin oxide (ITO) pre-evaporated on the glass surface is used as anode. And the limited area of luminescence is 3 × 3 mm^2. Before the experiment, the glass substrates are ultrasonically cleaned by detergent, ethanol, deionized water and isopropanol successively. After cleaning, the glass substrates are dried and cooled for 1 h. The deposition rate of all organic materials during the evaporation process is 0.05 Å/s - 2 Å/s, and the evaporation rate of the cathode material aluminum is 3 Å/s. Electro-optical data and spectrum are measured and recorded by a PR655 spectrometer and a computer-controlled Keithley 2400 digital power.

In our experiment, 1,1'-bis[4-(di-p-tolylamino)phenyl]cyclohexane (TAPC) and 1,3,5-tri[(3-pyridyl)-phen-3-yl) benzene(TmPyPB) act as the hole transport layer (HTL) and the electron transport layer (ETL), respectively[15]. Iridium(III)bis(4-phenylthieno[3,2-c]pyridinato-N,C2')acetylacetonate(PO-01) and iridium(III)bis[((4,6-difluorophenyl)-pyridinato-N,C2')] picolinate (FIrpic) are adopted as yellow and blue ultra-thin phosphor layers, respectively [6]. 4,4',4''-tri(N-carbazolyl) triphenylamine (TCTA), 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi) and (1,3,5-Triazine-2,4,6-triyl) tris (benzene-3,1-diyl)tris(diphenyl-phosphine oxide) (PO-T2T) are both the transport materials and the host materials in EML [2] [21] [22]. The device structure diagram of the emission layer material is shown in Figure 1.

3. Result and Discussion

Firstly, we investigate the effect of the interlayer material in EML on performances of CT-OLED. As shown in Figure 1, TPBi, PO-T2T and TPBi: PO-T2T are selected as interlayer materials in device A1, A2 and A3, respectively. There

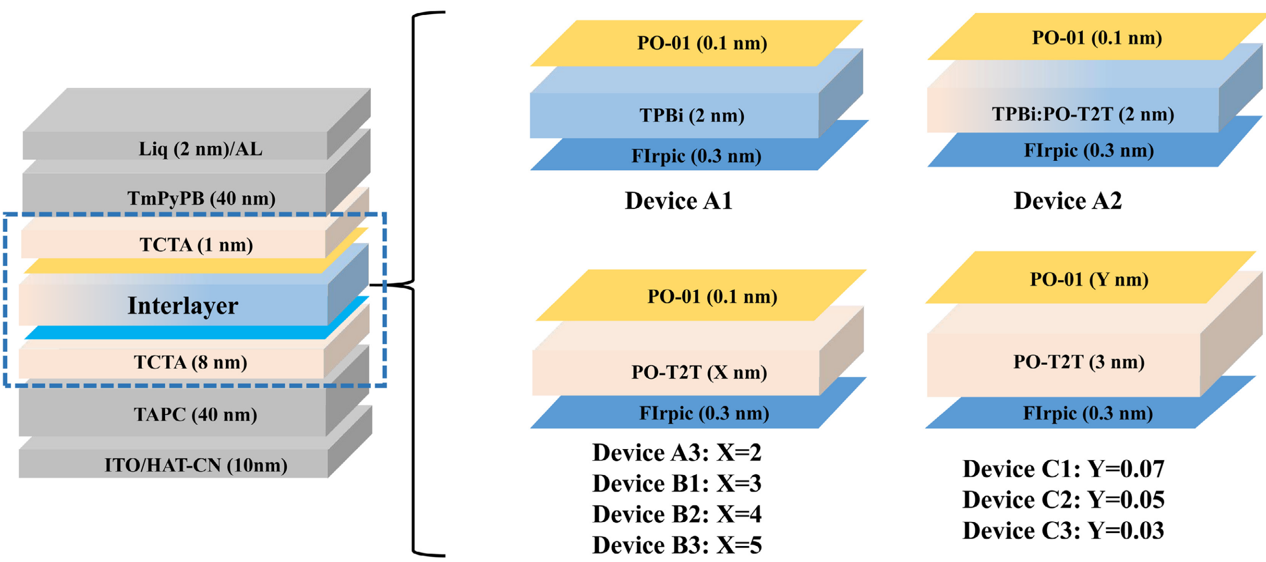


Figure 1. The schematic structural diagrams of all CT-WOLEDs used in this work.

will be three different energy transmission modes because that TPBi and PO-T2T can form blue and yellow interface exciplexes with TCTA on both sides of EML, respectively. Since the overall thickness of EML is only about 11 nm, the movement of RZ at the interface between the interlayer and TCTA with the change of voltage is little. The change of energy transmission path has a major impact on device performance.

Table 1 shows the electro-optical data and spectral data of all devices. Due to more balanced carrier distribution in EML, the maximum power efficiency and luminance of device A1 reach to 62.8 lm/W and 19350 cd/m^2, which are the highest among all devices, respectively. The relatively stable spectrum with voltage change also shows that the device A1 has the most balanced carrier distribution and energy transmission, as shown in Figure 2(b) and Figure 3(a). In order to realize the change of spectrum and CCT with voltage, we modify the energy transmission path in EML by replacing interlayer material with TPBi: PO-T2T in device A2 and PO-T2T in device A3. Because the yellow interface exciplex (PO-T2T/TCTA) has lower energy levels (T_1 = 2.35 eV) than the blue interface exciplex (TPBi/TCTA, T_1 = 2.80 eV), the introduction of PO-T2T will increase the Förster energy transfer from RZ to the yellow phosphor layer (PO-01) as shown in Figure 3(b) and Figure 3(c) [23] [24]. At low voltage, the yellow phosphor layer in device A3 obtains more energy by Förster energy transfer than the bule phosphor layer (FIrpic), so the blue spectral intensity in device A3 is suppressed, and the CCT of this device is also reduced. The CCT value of device A3 (2693 K) is the lowest when the voltage is 2.75 V. With the increase of voltage, the blue phosphor layer obtains more energy by directly capturing excitons, which leads to the enhancement of the blue spectral intensity and CCT value. Although the spectrum of device A3 becomes more unstable, it also extends the range of CCT from 123 K in device A1 to 707 K in device A3.

Table 1. EL performance of WOLEDs A1 - A3, B1 - B3 and C1 - C3.

Device	CD_{max} (mA/cm^2)	L_{max} (cd/m^2)	CE_{max} (cd/A)	PE_{max} (lm/W)	CCT (K)		ΔCCT (K)	$CIE_{x,y}$ (1931)	
					2.75 V	8.25 V		2.75 V	8.25 V
A1	343.08	19350	55.9	62.8	3295	3418	123	(0.454, 0.485)	(0.448, 0.490)
A2	380.00	14390	38.1	39.9	3172	3656	484	(0.468, 0.497)	(0.434, 0.496)
A3	379.66	15510	30.1	31.2	2693	3400	707	(0.503, 0.486)	(0.448, 0.488)
B1	339.59	8021	15.4	14.2	3120	5606	2486	(0.473, 0.498)	(0.329, 0.485)
B2	325.35	7166	13.5	12.1	3384	6297	2913	(0.458, 0.509)	(0.296, 0.514)
B3	317.61	6205	12.5	10.9	3359	6451	3092	(0.462, 0.514)	(0.289, 0.516)
C1	323.51	7311	14.5	13.1	3209	5728	2519	(0.467, 0.501)	(0.323, 0.482)
C2	316.52	7247	14.2	12.7	3254	5923	2669	(0.463, 0.499)	(0.315, 0.485)
C3	333.38	7041	13.4	11.9	3378	6272	2894	(0.454, 0.500)	(0.300, 0.484)

CD_{max}, L_{max}, PE_{max} and CE_{max} are the maximum current density, luminance, power efficiency and current efficiency.

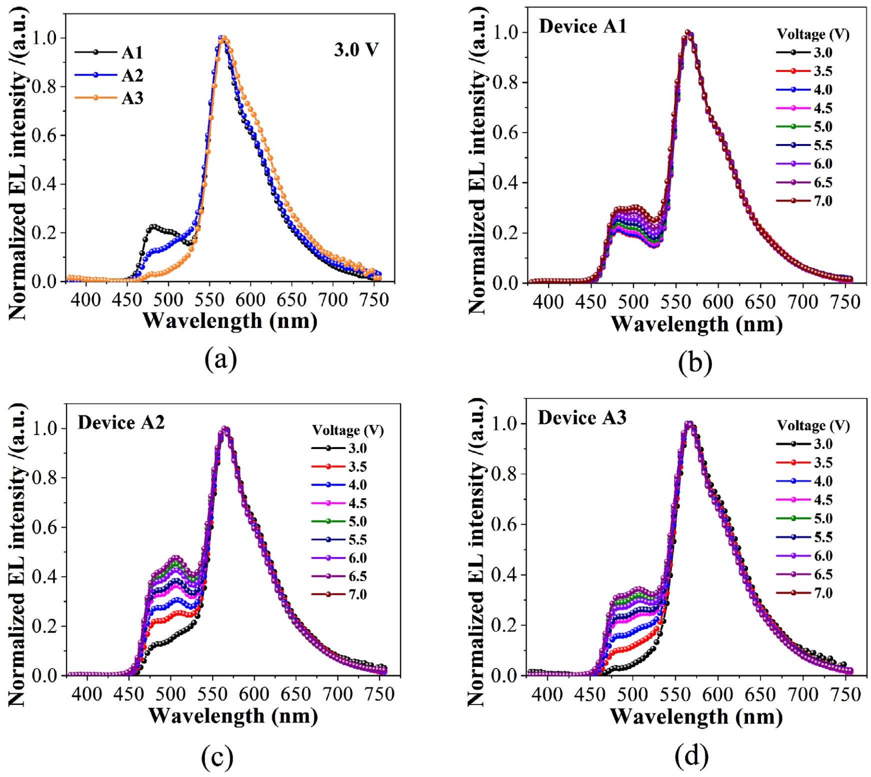


Figure 2. (a) The normalized EL spectra of devices A1 - A3 at 2.75 V; (b)-(d) The normalized EL spectra of devices A1 - A3 at different driving voltages.

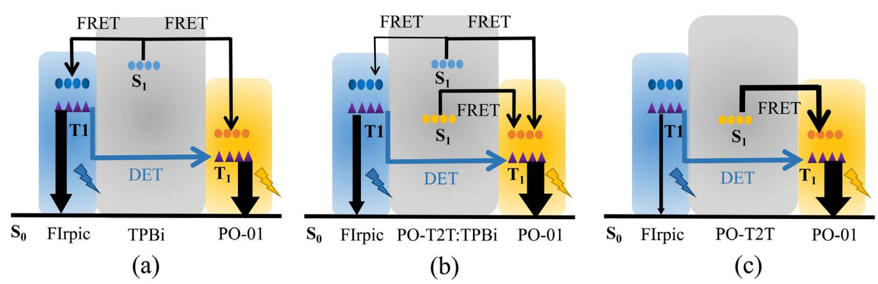


Figure 3. (a)-(c) The Energy transfer path diagram of device A1, A2 and A3 under low voltage in this study. S1, T1, S0, represent the singlet state level, triplet state level, ground state level, respectively. FRET and DET are Förster energy and Dexter energy transfer process.

Secondly, in order to improve the blue spectral intensity at high voltage, we also prepare a series of devices B by increasing the thickness of the interlayer (PO-T2T) in EML on the basis of device A3, which is conducive to suppressing the Dexter energy transfer from the blue phosphor layer to the yellow phosphor layer. The thicknesses of the interlayer (X) are 2 nm, 3 nm, 4 nm and 5 nm in devices A3, B1, B2 and B3, respectively. The increase in the thickness of the interlayer results in a further significant decrease in the maximum current density, efficiency and luminance because two ultra-thin phosphor layers cannot work at the same time. Although the value of CCT also increases compared with that of device A3 at low voltage, the enhancement of blue light spectrum at high voltage leads to more increase of CCT, which also makes the adjustable range of CCT larger. The CCT span of device B3 is 3092 K from 2.75 V to 8.25 V as shown Table 1, Figure 4(a) and Figure 5(b), which is the largest of all devices. Figures 4(a)-(c) indicate the normalized EL spectra of devices B1 - B3 at different driving voltage. At low voltage, the yellow phosphor spectrum of device B is significantly higher than that of blue phosphor. With the increase of voltage, the intensity of blue phosphorescence spectrum increases rapidly due to the inhibition of Dexter energy transmission by the interlayer as shown in Figure 5(a). Moreover, the yellow phosphor spectrum gradually decreases and disappears with the increase of voltage, which is different from the fact that the yellow phosphor spectrum of device A3 has been in a saturated state. At high voltage, the weakening

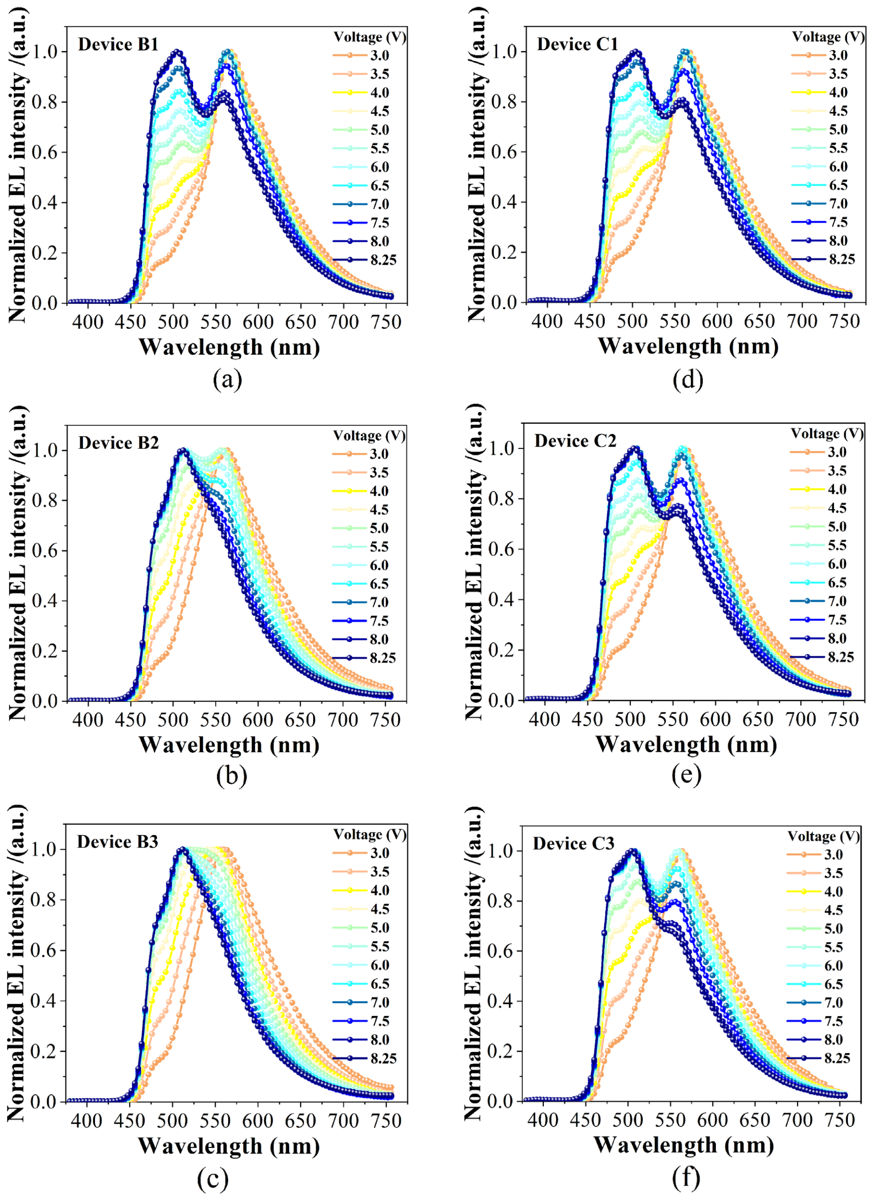
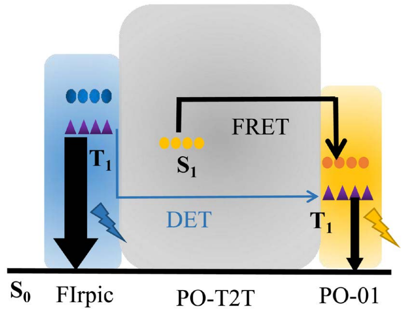
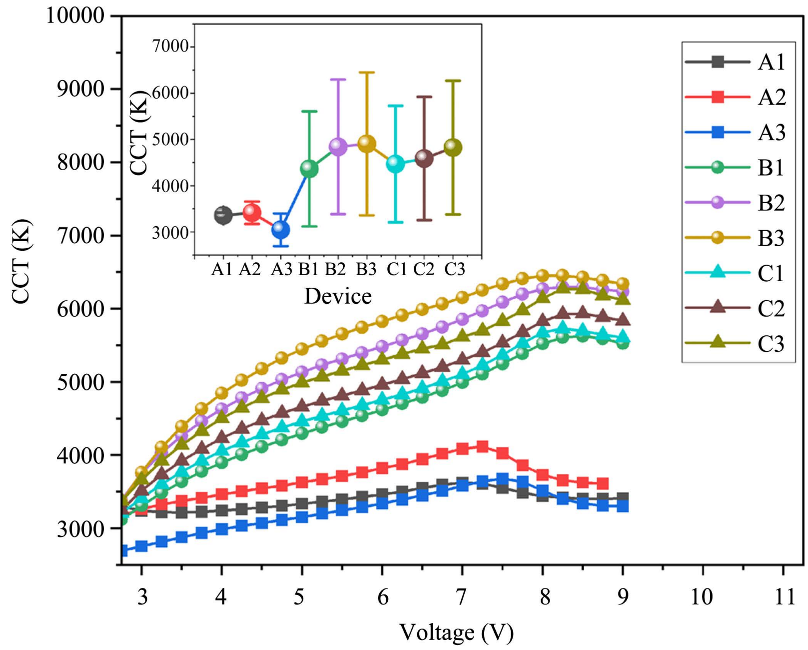


Figure 4. (a)-(f) The normalized EL spectra of devices B1 - B3 and devices C1 - C3 at different driving voltages.



(a)



(b)

Figure 5. (a) The Energy transfer path diagram of device B and C at high voltage; (b) voltage-correlation color temperature curves of all devices. The insert diagram shows the ranges of CCT span of all devices with increasing voltage from 2.75 V to 8.25 V.

of yellow phosphor layer further increases the spectral intensity of blue phosphor, thus further increasing the CCT value and expanding CCT span in device B3 with the thickest interlayer (5 nm).

Finally, we fabricate in order to improve the blue spectral intensity at high voltage, we also prepare a series of devices C by decreasing the thickness of the yellow phosphor layer (PO-01, Y) in EML on the basis of device B1. In devices B1, C1, C2 and C3, the thicknesses of yellow phosphor layer correspond to 0.1 nm, 0.07 nm, 0.05 nm and 0.03 nm, respectively. Reducing the thickness of the yellow phosphor layer increases the blue spectral intensity at high voltage and low voltage at the same time. Although the range of CCT is also expanding compared with device B1, the effect of yellow phosphor layer still exists at high voltage, so the effect of improving the CCT span is weaker than that of increasing the thickness of the interlayer.

4. Conclusion

In conclusion, we demonstrate that a simple OLED device with an interlayer inserted between two ultra-thin phosphor layers can achieve a wider CCT span from 3359 K to 6451 K at voltage increases from 2.75 V to 8.25 V. The relationship between the energy transfer in EML and CCT change trend is investigated by adjusting the interface exciplexes and the thickness of the interlayer or the phosphor layer in devices A, B and C, respectively. On the one hand, we find that the yellow interface exciplex is beneficial to reduce the CCT value at low voltage. On the other hand, increasing the thickness of the interlayer layer can not only improve the intensity of the blue spectrum at high voltage, but also reduce the role of yellow phosphor layer, thus increasing the CCT value at high voltage. Our results provide a promising method for preparing low-cost, simple CT-OLED device.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Ding, D.X., Wang, Z.C., Li, C.Y., *et al.* (2020) Highly Efficient and Color-Stable Thermally Activated Delayed Fluorescence White Light-Emitting Diodes Featured with Single-Doped Single Emissive Layers. *Advanced Materials*, **32**, Article ID: 1906950. https://doi.org/10.1002/adma.201906950

[2] Wang, B.Q., Kou, Z.Q., Tang, Y., *et al.* (2019) High CRI and Stable Spectra White Organic Light-Emitting Diodes with Double Doped Blue Emission Layers and Multiple Ultrathin Phosphorescent Emission Layers by Adjusting the Thickness of Spacer Layer. *Organic Electronics*, **70**, 149-154. https://doi.org/10.1016/j.orgel.2019.04.013

[3] Ying, S.A., Sun, Q., Dai, Y.F., *et al.* (2019) Precise Regulation of the Emissive Layer for Ultra-High Performance White Organic Light-Emitting Diodes in an Exciplex Forming Co-Host System Materials. *Chemistry Frontiers*, **3**, 640-649. https://doi.org/10.1039/C9QM00017H

[4] Wang, L.J., Kou, Z.Q., Wang, B.Q., *et al.* (2021) Realizing High Efficiency/CRI/Color Stability in the Hybrid White Organic Light Emitting Diode by Manipulating Exciton Energy Transfer. *Optical Materials*, **115**, Article ID: 111059. https://doi.org/10.1016/j.optmat.2021.111059

[5] Dai, X.D. and Cao, J. (2020) Study on Spectral Stability of White Organic Light-Emitting Diodes with Mixed Bipolar Spacer Based on Ultrathin Non-Doped Phosphorescent Emitting Layers. *Organic Electronics*, **78**, Article ID: 105563. https://doi.org/10.1016/j.orgel.2019.105563

[6] Li, A., Sun, M.Y., Yang, L.P., *et al.* (2021) Improved Efficiency, Stable Spectra and Low Efficiency Roll-off Achieved Simultaneously in White Phosphorescent Organic Light-Emitting Diodes by Strategic Exciton Management. *Organic Electronics*, **97**, Article ID: 106262. https://doi.org/10.1016/j.orgel.2021.106262

[7] Li, J., Chen, T.Q., Yang, J. and Cao, J. (2021) Realizing the Stable Spectrum in Four-Chromatic White Organic Light-Emitting Diodes by Controlling the Positions of Various Emitters in the Bipolar Interlayer. *Journal of Physics D: Applied Physics*,

54, Article ID: 165105. https://doi.org/10.1088/1361-6463/abdd67

[8] Chen, T.Q., Li, J., Cao, J. and Yang, J. (2021) Spectrum-Stable Tetra-Chromatic White Organic Light-Emitting Diodes with Red Emitter outside the Exciton Recombination Zone. *Optical Materials*, **117**, Article ID: 111150. https://doi.org/10.1016/j.optmat.2021.111150

[9] Jou, J.-H., Tai, T.-C., Liu, S.H., *et al.* (2019) Pseudo-Sunlight Organic Light-Emitting Diodes. *Optics & Laser Technology*, **112**, 494-499. https://doi.org/10.1016/j.optlastec.2018.11.046

[10] Imbrasas, P., Lenk, S. and Reineke, S. (2020) Organic Light-Emitting Diodes with Split Recombination Zones: A Concept for Versatile Color Tuning. *Organic Electronics*, **78**, Article ID: 105558. https://doi.org/10.1016/j.orgel.2019.105558

[11] Zhang, J.M., Wang, Y.H., Liu, S.H., *et al.* (2022) Color-Tunable Organic Light-Emitting Diodes with Ultrathin Thermal Activation Delayed Fluorescence Emitting Layer. *Applied Physics Letters*, **120**, Article ID: 171102. https://doi.org/10.1063/5.0084137

[12] Guo, F., Karl, A., Xue, Q.-F., *et al.* (2017) The Fabrication of Color-Tunable Organic Light-Emitting Diode Displays via Solution Processing. *Light, Science & Applications*, **6**, e17094. https://doi.org/10.1038/lsa.2017.94

[13] Jeon, Y.M., Noh, I., Seo, Y.C., *et al.* (2020) Parallel-Stacked Flexible Organic Light-Emitting Diodes for Wearable Photodynamic Therapeutics and Color-Tunable Optoelectronics. *ACS Nano*, **14**, 15688-15699. https://doi.org/10.1021/acsnano.0c06649

[14] Yang, M.L., Shikita, S., Min, H., *et al.* (2021) Wide-Range Color Tuning of Narrowband Emission in Multi-Resonance Organoboron Delayed Fluorescence Materials through Rational Imine/Amine Functionalization. *Angewandte Chemie International Edition*, **60**, 23142-23147. https://doi.org/10.1002/anie.202109335

[15] Zhang, J.M., Liu, S.H., Chen, Y.F., Zhang, L.T. and Xie, X.F. (2021) Simple-Structure Color-Tunable Fluorescent Organic Light-Emitting Devices with Chromaticity Difference beyond Five-Step McAdam Ellipses. *Journal of Physics D: Applied Physics*, **54**, Article ID: 505103. https://doi.org/10.1088/1361-6463/ac2642

[16] Zhao, D., Huang, W., Qin, Z., Wang, Z.J. and Yu, J.S. (2018) Emission Spectral Stability Modification of Tandem Organic Light-Emitting Diodes through Controlling Charge-Carrier Migration and Outcoupling Efficiency at Intermediate/Emitting Unit Interface. *ACS Omega*, **3**, 3348-3356. https://doi.org/10.1021/acsomega.8b00314

[17] Zhang, H., Su, Q. and Chen, S.M. (2020) Quantum-Dot and Organic Hybrid Tandem Light-Emitting Diodes with Multi-Functionality of Full-Color-Tunability and White-Light-Emission. *Nature Communications*, **11**, Article No. 2826. https://doi.org/10.1038/s41467-020-16659-x

[18] Lee, H., Cho, H., Byun, C.-W., *et al.* (2018) Color-Tunable Organic Light-Emitting Diodes with Vertically Stacked Blue, Green, and Red Colors for Lighting and Display Applications. *Optics Express*, **26**, 18351-18361. https://doi.org/10.1364/OE.26.018351

[19] Ying, S.A., Wu, Y.B., Sun, Q., *et al.* (2019) High Efficiency Color-Tunable Organic Light-Emitting Diodes with Ultra-Thin Emissive Layers in Blue Phosphor Doped Exciplex. *Applied Physics Letters*, **114**, Article ID: 033501. https://doi.org/10.1063/1.5082011

[20] Li, Y.L., Liu, N., Zhou, P.C., *et al.* (2021) Efficient and Color-Tunable Organic Light-Emitting Diodes for Rear Light Application on the Motor Vehicle. *Materials Science*, **27**, 264-268. https://doi.org/10.5755/j02.ms.24678

[21] Zhou, J., Kou, Z.Q., Wang, L.J., *et al.* (2021) Realizing High-Performance Color-Tunable WOLED by Adjusting the Recombination Zone and Energy Distribution in the Emitting Layer. *Journal of Physics D*: *Applied Physics*, **54**, Article ID: 265107. https://doi.org/10.1088/1361-6463/abf258

[22] Ying, S.A., Chen, Y.W., Yao, J.W., *et al.* (2020) High Efficiency Doping-Free Warm-White Organic Light-Emitting Diodes with Strategic-Tuning of Radiative Excitons by Combining Interfacial Exciplex with Multi-Ultrathin Emissive Layers. *Organic Electronics*, **85**, 105876. https://doi.org/10.1016/j.orgel.2020.105876

[23] Wang, X.Y., Zhang, Y.F., Yu, Z., *et al.* (2022) Overcoming Energy Loss of Thermally Activated Delayed Fluorescence Sensitized-OLEDs by Developing a Fluorescent Dopant with a Small Singlet-Triplet Energy Splitting. *Journal of Materials Chemistry C*, **10**, 1681-1689. https://doi.org/10.1039/D1TC05700F

[24] Jiang, W., Zhao, G.M., Chen, H.W. and Sun, Y.M. (2022) Novel Ternary Exciplex System Based on TCTA Dendrimer with a New Linking Type Amongst Various Functional Donors. *Journal of Materials Science*: *Materials in Electronics*, **33**, 11403-11413. https://doi.org/10.1007/s10854-022-08113-z