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#### Introduction

Organic light-emitting diodes (OLEDs) have great application potential in the fields of high-resolution displays and solidstate lighting.<sup>1,2</sup> The emission color and electroluminescence efficiency of OLEDs greatly depend on the properties of organic emitters.<sup>3,4</sup> Due to the efficient singlet–triplet intersystem crossing (ISC) promoted by heavy metal atoms, phosphorescent Pt( $\pi$ ) complexes can theoretically realize 100% internal quantum efficiency in OLEDs by utilizing all electrically generated singlet and triplet excitons.<sup>5,6</sup> Therefore, the development of Pt( $\pi$ ) complexes is highly attractive. To date, the reported Pt( $\pi$ ) complexes are usually composed of one metal atom, and thereby the emission properties of these mononuclear Pt( $\pi$ ) complexes are determined by organic ligands. Occasionally, to further adjust the properties, an extra Pt( $\pi$ ) center is incorporated to

# Efficient dinuclear Pt(II) complexes based on the triphenylphosphine oxide scaffold for high performance solution-processed OLEDs<sup>+</sup>

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Dinuclear Pt(n) complexes have the potential to achieve high electroluminescence (EL) performance because of the enhanced phosphorescence emission induced by the extra metal center. However, to date, organic light-emitting devices (OLEDs) utilizing dinuclear Pt(n) complexes as emitters usually show low EL performance with external quantum efficiencies (EQE) less than 10%. In this work, with the triphenylphosphine oxide group as the scaffold core and different N-heterocycles (pyridine, thiazole, and quinoline) as the end-groups, dinuclear Pt(n) complexes PyPODPt, ThPODPt, and QuPODPt are synthesized to show bright emissions peaking at 500, 543, and 586 nm in solutions. The photoluminescence quantum yields are measured to be up to 0.96. More importantly, the solutionprocessed orange-red device based on QuPODPt exhibits outstanding EL performance with the EQE reaching 11.2%, which is among the highest EQEs reported for OLEDs employing dinuclear Pt(n)complexes. The superior device performance demonstrates the promising potential of triphenylphosphine oxide-based dinuclear Pt(n) complexes for OLED applications.

> form the corresponding dinuclear Pt(II) complex.<sup>7–9</sup> Because of the enhanced ISC induced by the second Pt(II) center, dinuclear Pt(II) complexes could show higher photoluminescence yields (PLQYs) than their corresponding mononuclear Pt(II) complexes, leading to significantly improved electroluminescence efficiencies.<sup>10–12</sup> However, compared with OLEDs based on other dinuclear organometallic complexes,<sup>13–15</sup> devices using dinuclear Pt(II) complexes as emitters usually exhibit relatively inferior performance with external quantum efficiencies typically below 10%.<sup>7,16–18</sup>

> In this contribution, we report the synthesis of three dinuclear  $Pt(\pi)$  complexes based on the triphenylphosphine oxide scaffold. The triphenylphosphine oxide group was adopted because its branched chemical structure is suitable to coordinate with more metal centers. Furthermore, the strong electron-withdrawing property of the triphenylphosphine oxide group can benefit the electron-injection/transport process in OLEDs.<sup>19,20</sup> The emission colors of the resultant dinuclear Pt(II) complexes could be tuned from green to orange-red by simply using different N-heterocycles. Furthermore, these dinuclear Pt(II) complexes showed impressively high PLQYs up to 0.96. Most importantly, the solution-processed orange-red OLED based on QuPODPt achieved the maximum EQE of 11.2%, which is among the best reported for OLEDs based on dinuclear Pt(II) complexes. This work sheds light on the potential of developing efficient triphenylphosphine oxide-based dinuclear Pt(II) complexes for high performance OLEDs.

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#### Results and discussion

The synthetic routes of the target dinuclear Pt(n) complexes are illustrated in Scheme 1. The synthesis details are provided in the ESI.<sup>†</sup> The key procedure was the synthesis of triphenylphosphine oxide-based ligands. The dibromo-substituted triphenylphosphine oxide moiety was firstly prepared and then reacted with different N-heterocycles through Suzuki–Miyaura coupling to form tetradentate ligands which could coordinate with two metal centres. The structures of the target dinuclear Pt(n) complexes were thoroughly characterized by <sup>1</sup>H, <sup>13</sup>C, and <sup>31</sup>P NMR and high resolution mass spectrometry analyses (see Fig. S1 and S2 in the ESI<sup>†</sup>).

The thermal stability of these dinuclear Pt(n) complexes was investigated by thermogravimetric analysis (TGA) under a  $N_2$ atmosphere. As revealed by the TGA curves (Fig. S3, ESI†), the decomposition temperatures ( $T_d$ ) of **PyPODPt**, **ThPODPt**, and **QuPODPt** were *ca.* 361, 341, and 256 °C (Table 1), respectively, demonstrating the high thermal stability of these dinuclear Pt(n) complexes. The high thermal stability would avoid the detrimental degradation during device fabrication and operation processes.

In CH<sub>2</sub>Cl<sub>2</sub> solution  $(3 \times 10^{-5} \text{ mol L}^{-1})$  at room temperature, these dinuclear Pt(II) complexes showed strong absorption bands in the region of 250–350 nm (Fig. 1b and Table 1), which were attributed to the spin-allowed singlet  $\pi$ – $\pi$ \* transitions of the metal perturbed ligands. In addition to the strong high energy absorptions, weak absorptions were observed in the range from 370 to 470 nm, which could be assigned to the charge transfer processes, including the intraligand charge transfer (ILCT), ligand-to-ligand charge transfer (LLCT), and metal-to-ligand charge transfer (MLCT) transitions.<sup>21</sup> Theoretical calculations revealed that the distances between the two Pt centers in **PyPODPt**, **ThPODPt**, and **QuPODPt** were 10.668, 10.686, and 10.686 Å, respectively, which were too far to form substantial Pt–Pt interactions. Therefore, the absorptions have

no contribution from metal-metal-to-ligand charge-transfer. The molar extinction coefficients  $(\varepsilon)$  for the low-energy absorptions of these dinuclear Pt(II) complexes were obviously larger than those of conversational mononuclear Pt(II) complexes.<sup>22,23</sup> For example, the  $\log \varepsilon$  of low-energy absorption for **PyPODPt** was 4.05, while that for the corresponding mononuclear Pt(n)complex decreased to 3.55,23 which was in good accordance with the conclusion that the incorporation of an extra metal centre could enhance the low-energy absorption.<sup>24</sup> The larger  $\varepsilon$ might suggest an enhanced MLCT process, which could increase the phosphorescence emission ability.<sup>25</sup> In addition, compared to conventional mononuclear Pt(II) complexes,<sup>22,23</sup> these dinuclear Pt(II) complexes showed shorter lifetimes (0.52-0.71  $\mu$ ). The shorter lifetime reflected the faster ISC process, which would facilitate the triplet radiation. Therefore, PyPODPt, ThPODPt, and QuPODPt displayed impressively high photoluminescence quantum yields (PLQYs) of up to 0.96 (Table 1), which was among the highest PLQYs reported for dinuclear Pt(II) complexes.<sup>7,10,11,16</sup> By applying different N-heterocycles, these dinuclear Pt(II) complexes exhibited lowenergy absorption peaks in the order of **PyPODPt** (378 nm) < ThPODPt (384 nm) < QuPODPt (420 nm), suggesting the gradually narrowed optical energy bandgaps ( $E_{gap}$ ). Consequently, the emission peaks measured in CH2Cl2 at room temperature were significantly red-shifted from 500 nm (PyPODPt) to 543 nm (ThPODPt) and 586 nm (QuPODPt), demonstrating the simplicity of largely adjusting the emission colors of dinuclear Pt(II) complexes based on the triphenylphosphine oxide scaffold. In 1 wt% doped 4,4'-bis(carbazol-9-yl)biphenyl (CBP) films, PyPODPt and ThPODPt showed slightly red-shifted emissions, while QuPODPt displayed the blue-shifted emission (Fig. 1c), indicating the relatively severe molecular aggregation behaviours of PyPODPt and ThPODPt than QuPODPt even in such a low doping level.<sup>26</sup> The theoretical calculations revealed remarkably large dipole moments  $(\mu)$  of these dinuclear Pt(II) complexes (Table 1). With a larger  $\mu$ , an emitter tends to aggregate, which



Scheme 1 Synthetic routes of the dinuclear Pt(II) complexes.

Table 1 Photophysical, thermal, and energy level data for PyPODPt, ThPODPt, and QuPODPt

	$\lambda_{abs}{}^a$ (nm)	$\lambda_{\rm em}{}^a$ (nm)	$\lambda_{\rm em}^{\ \ b} ({\rm nm})$	PLQY <sup>c</sup>	$T_{\rm d}$ (°C)	HOMO/LUMO <sup><math>d</math></sup> (eV)	$\mu^{e}$ (D)
PyPODPt	258 (4.80), 282 (4.74), 378 (4.05)	500 (0.52 μs)	502 (1.44 µs)	0.96/0.36	361	-5.56/-2.70	6.4
ThPODPt	266 (4.47), 312 (4.39), 384 (3.95)	543 (0.62 µs)	545 (1.22 µs)	0.49/0.17	341	-5.67/-2.88	5.7
QuPODPt	260 (4.51), 296 (4.51), 356 (4.13), 420 (3.82)	586 (0.71 µs)	580 (2.37 µs)	0.79/0.62	256	-5.59/-2.95	5.7

<sup>*a*</sup> Measured in CH<sub>2</sub>Cl<sub>2</sub> (3 × 10<sup>-5</sup> mol L<sup>-1</sup>), log  $\varepsilon$  values and the corresponding lifetimes are shown in parentheses. <sup>*b*</sup> Measured in doped film. <sup>*c*</sup> Measured in CH<sub>2</sub>Cl<sub>2</sub>/1 wt% doped CBP film. <sup>*d*</sup> Calculated according to the equations:  $E_{HOMO} = -(E_{ox} + 4.8)$  eV and  $E_{LUMO} = E_{HOMO} + E_{gap}$  eV. <sup>*e*</sup> Theoretically calculated dipole moment at the ground state.



usually leads to concentration quenching.<sup>27</sup> Therefore, these dinuclear Pt(II) complexes showed reduced PLQYs in doped films. However, due to the larger molecular structure which could suppress the aggregation to some extent, the **QuPODPt** doped film still showed an impressively high PLQY of 0.62, demonstrating its great potential in fabricating efficient solution-processed OLEDs.

The electrochemical properties were investigated in CH<sub>2</sub>Cl<sub>2</sub> solution using the cyclic voltammetry (CV) method. According to CV measurements, PyPODPt, ThPODPt, and QuPODPt showed the oxidation potential  $(E_{\rm ox})$  values of 0.76, 0.87, and 0.79 V, respectively. Thus, the highest occupied molecular orbital (HOMO) levels  $(E_{HOMO})$  of PyPODPt, ThPODPt, and QuPODPt were calculated to be -5.56, -5.67, and -5.59 eV, respectively, according to the equation:  $E_{HOMO} = -(E_{ox} + 4.8) \text{ eV}.$ During the cathodic scanning, no obvious reduction waves were recorded. Therefore, the lowest unoccupied molecular orbital (LUMO) levels ( $E_{LUMO}$ ) were determined from  $E_{HOMO}$  and optical band gaps ( $E_{gap}$ ) according to the equation:  $E_{LUMO}$  =  $E_{\rm HOMO} + E_{\rm gap}$  eV.<sup>28</sup> The calculated  $E_{\rm LUMO}$  values for PyPODPt, ThPODPt, and QuPODPt were -2.70, -2.88, and -2.95 eV, respectively. Compared with the LUMO levels of other dinuclear Pt(II) complexes showing similar emission colors,<sup>10,12,24,29</sup> the deeper LUMO levels of PvPODPt, ThPODPt, and OuPODPt would promote the electron injection process to benefit the OLED performance.

Density functional theory (DFT) and time-dependent DFT (TD-DFT) calculations were carried out to gain insights into the aforementioned properties. The electron density distributions of key molecular orbitals are depicted in Fig. 2. The calculated energies and the corresponding main assignments of each transition are summarized in Table 2. The calculated  $E_{\rm HOMO}$ 

and  $E_{\text{LUMO}}$  as well as the excitation energies were in good agreement with the experimental results. The theoretical results exposed that  $S_0 \rightarrow S_1$  and  $S_0 \rightarrow T_1$  excitations were mainly contributed by the HOMO-1  $\rightarrow$  LUMO, HOMO  $\rightarrow$ LUMO, and HOMO  $\rightarrow$  LUMO+1 transitions (Table 2). The calculated UV-vis absorption spectra of these dinuclear Pt(II) complexes showed good agreement with the experimental results as well (Fig. S4, ESI†), which indicated the mixed  $\pi$ - $\pi$ \* (*ca.* 35–58%), ILCT (*ca.* 10–14%), LLCT (*ca.* 24–26%), and MLCT (*ca.* 24–25%) features of the low-energy absorptions. For example, the major assignment for the  $S_0 \rightarrow S_1$  excitation was HOMO  $\rightarrow$ LUMO+1 for **PyPODPt**. As shown in Fig. 2, the HOMO of **PyPODPt** was mainly localized at the left arm (LA) with significant



Fig. 2 Key molecular orbital distributions of **PyPODPt**, **ThPODPt**, and **QuPODPt**.

Table 2 TD-DFT results for PyPODPt, ThPODPt, and QuPODPt based on optimized S<sub>0</sub> geometries

	MOs	Contributi	on from Pt and lig	gands to MOs (%)	Energy level (eV)	Main assignment for $S_0 \rightarrow S_1$ excitation/ $E_{cal}^a/f^a$	Main assignment for $S_0 \rightarrow T_2$ excitation/ $E_{cal}^a$
PyPODPt		Pt	L-PyPO	acac			
•	L+1	6.06	92.64	1.30	-1.70	$H \rightarrow L+1$ (49.3%),	$H-1 \rightarrow L (38.6\%),$
	$L^{a}$	5.34	93.46	1.20	-1.80	$H \rightarrow L (28.0\%),$	$H-3 \rightarrow L(13.0\%),$
	$H^{a}$	32.30	40.66	27.04	-5.45	$H-2 \rightarrow L+1 (10.5\%)$	$H \rightarrow L (11.0\%),$
	H-1	32.66	45.56	21.78	-5.54	/3.05 eV	$H-1 \rightarrow L+1 (9.3\%)$
	H-2	36.68	27.45	35.87	-5.77	/0.0380	/2.58 eV
	H-3	35.92	25.22	38.86	-5.88		
ThPODPt		Pt	L-ThPO	acac			
	L+1	5.41	93.67	0.92	-1.87	$H \rightarrow L$ (36.0%),	$H-1 \rightarrow L(27.4\%),$
	L	4.74	94.39	0.87	-1.99	$H \rightarrow L+1 (30.8\%),$	$H-3 \rightarrow L(17.4\%),$
	Н	31.46	38.92	29.62	-5.57	$H-2 \rightarrow L+1 (12.3\%),$	$H-3 \rightarrow L+1 (10.1\%),$
	H-1	31.97	54.35	22.38	-5.65	$H-2 \rightarrow L (8.0\%)$	$H \rightarrow L (9.6\%)$
	H-2	35.61	32.92	31.47	-5.77	/2.99 eV	/2.39 eV
	H-3	34.58	29.45	35.97	-5.87	/0.0669	
QuPODPt		Pt	L-QuPO	acac			
-	L+1	4.43	94.72	0.85	-2.06	$H \rightarrow L+1 (50.8\%),$	$H-2 \rightarrow L+1 (29.6\%),$
	L	4.12	95.04	0.84	-2.14	$H \rightarrow L(23.7\%),$	$H-2 \rightarrow L(11.4\%),$
	Н	31.74	39.49	28.77	-5.49	$H-2 \rightarrow L+1 (12.5\%)$	$H-3 \rightarrow L(9.8\%),$
	H-1	32.11	46.02	21.87	-5.58	/2.79 eV	$H-1 \rightarrow L(9.5\%)$
	H-2	36.86	35.33	27.81	-5.72	/0.0647	/2.33 eV
	H-3	36.14	33.16	30.70	-5.83		

contribution from the Pt centre (ca. 32.30%), the phenyl ring (ca. 33.94%) chelated to the Pt centre, and the acetylacetone (acac) ligand (ca. 27.04%), while the LUMO+1 was mainly contributed from the L-PyPO ligand with ca. 71.71% electron density located on the left arm and ca. 18.93% electron density located on the right arm (RA). Therefore, the HOMO  $\rightarrow$ LUMO+1 excitation consisted of  $\pi$ - $\pi$ \* ( $\pi$  orbital of the phenyl ring chelated to the Pt centre  $\rightarrow \pi^*$  orbital of the same phenyl ring), ILCT (the phenyl ring chelated to the Pt centre  $\rightarrow$  pyridyl rings), LLCT (acac  $\rightarrow$  L-PyPO), and MLCT (the Pt centre  $\rightarrow$ **L-PyPO**) transitions. Clearly, the  $S_0 \rightarrow S_1$  and  $S_0 \rightarrow T_1$  excitations of these dinuclear Pt(II) complexes all possessed similar properties. Different from the previous situations in which acac usually contributed very little or negligibly to the transitions of Pt(n)complexes,<sup>10,18,23,30</sup> it is worth mentioning that acac in these dinuclear Pt(II) complexes obviously participated in the transitions, indicating that the properties of these dinuclear Pt(II) complexes could be further adjusted by substituting or even changing the acac ligands.

The phosphorescence emission properties were investigated by analyzing the natural transition orbitals (NTO) based on the optimized  $T_1$  geometries. As shown in Fig. 3, although **ThPODPt** possessed two pairs of degenerate hole  $\rightarrow$  particle  $(H \rightarrow P)$  transitions, the T<sub>1</sub> states of all three complexes were substantially (around 99%) contributed by the H  $\rightarrow$  P transitions. The hole orbitals were mainly concentrated on the Pt center (9-22%) and the segment (70-80%) chelated with this Pt center, while the particle orbitals were predominantly (92-94%) contributed by the segment chelated with this Pt center. Because the organic ligands made a high contribution to both hole and particle orbitals (Fig. 3 and Table S1, ESI<sup>+</sup>), the efficient phosphorescence emissions mainly originated from the triplet  $\pi$ - $\pi$ \* transitions mixed with notable MLCT characters. Therefore, PyPODPt, ThPODPt, and QuPODPt displayed broadened and structured emission spectra (Fig. 1).<sup>22</sup>

The EL properties were investigated by fabricating solutionprocessed OLEDs with the simple configuration of ITO/ PEDOT:PSS (30 nm)/PVK (25 nm)/EML (20 nm)/TmPyPB



Fig. 3 NTO investigation of PyPODPt, ThPODPt, and QuPODPt.

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Fig. 4 (a) Device structure and energy level alignment. (b) EL spectra. (c) Curves of EQE vs current density. (d) Curves of CE and PE vs current density.

(40 nm)/LiF (1 nm)/Al (100 nm) (Fig. 4a). Polymers PEDOT:PSS [poly(ethylenedioxythiophene):poly(styrenesulfonate)] and PVK [poly(9-vinylcarbazole)] were used as hole-injection and transport materials, respectively. The emissive layer (EML) was prepared by doping the dinuclear Pt(II) complex into CBP at a very low concentration of 1 wt%. Due to its relatively high electron mobility and significantly deep HOMO level, TmPyPB [1,3,5tri(*m*-pyridin-3-ylphenyl)benzene] was chosen to act as both electron transport and hole blocking layers at the same time. The device performance is presented in Fig. 4, and the key EL data are summarized in Table 3. These devices showed typical turn-on voltages in the range of 5.0-5.4 V for solution-processed OLEDs.<sup>31–33</sup> As shown in Fig. 4b, the EL spectra of these devices were similar to the PL spectra of doped films, except that weak emissions around 400 nm from the CBP host were observed.<sup>34</sup> Device A based on PyPODPt showed the peak EQE, current efficiency (CE), and power efficiency (PE) of 6.85%, 21.3 cd  $A^{-1}$ , and 11.7 lm  $W^{-1}$ , respectively. At a luminance of 100 cd  $m^{-1}$ this device still gave decent efficiencies of 6.82%, 21.1 cd  $A^{-1}$ , and 11.0 lm W<sup>-1</sup>, indicating a low efficiency roll-off. Based on

Table 3 Key EL performance of OLEDs based on PyPODPt, ThPODPt, and QuPODPt

Devices	$\lambda_{\rm EL} \left( nm \right)$	$V_{\text{turn-on}}^{a}$ (V)	$EQE^{b}$ (%)	$\operatorname{CE}^{b}(\operatorname{cd}\operatorname{A}^{-1})$	$\mathrm{PE}^{b} (\mathrm{Im} \mathrm{W}^{-1})$
A	502, 538	5.1	6.85/6.82	21.3/21.1	11.7/11.0
В	546, 590	5.0	4.52/3.19	10.3/6.85	6.20/3.26
С	582, 608	5.4	11.2/8.80	21.7/16.8	11.8/8.28

 $^a$  Driving voltage at *ca.* 1.0 cd m<sup>-2</sup>.  $^b$  Efficiencies in the order of the peak value/at a luminance of 100 cd m<sup>-2</sup>.

ThPODPt, device B displayed inferior performance with EQE, CE, and PE of 4.52%, 10.3 cd  $A^{-1}$ , and 6.20 lm  $W^{-1}$ , respectively. Their relatively low efficiencies were related to the low PLQY of the ThPODPt doped film. Furthermore, the inefficient energy transfer from the host to **ThPODPt**, supported by the relatively strong emission from CBP in device B (Fig. 4b), was also responsible for the unsatisfactory performance of device B. Fortunately, device C achieved the highest EQE, CE, and PE of up to 11.2%, 21.3 cd  $A^{-1}$ , and 11.7 lm  $W^{-1}$ , respectively (Fig. 4c and d). To the best of our knowledge, these are among the highest efficiencies reported for OLEDs based on dinuclear Pt(II) complexes (Table S2, ESI<sup>+</sup>).<sup>7,10-12,16,18,35</sup> In addition to the high PLQY of the QuPODPt doped film, the excellent EL performance of device C was also inseparable from the relatively more efficient energy transfer, as supported by the weakest emission from the CBP host among these devices. It is also worth mentioning that device C showed orange-red emission with a remarkably large full width at half maximum over 105 nm, which indicated the great potential of QuPODPt in fabricating OLEDs that can emit high-quality white light.

#### Conclusion

To conclude, three efficient dinuclear Pt(n) complexes were developed based on the triphenylphosphine oxide group with different *N*-heterocycles. These thermally stable complexes showed green to orange-red emissions with impressively high PLQYs among all the reported dinuclear Pt(n) complexes to date. Accordingly, even at a low doping level of 1.0 wt%, solution-processed OLEDs based on these complexes could show high EL efficiencies. Particularly, the orange-red emitting device based on **QuPODPt** displayed the highest EQE, CE and PE of 11.2%, 21.3 cd  $A^{-1}$ , and 11.7 lm  $W^{-1}$ , respectively, which is among the most efficient OLEDs based on dinuclear Pt( $\pi$ ) complexes. This work demonstrates that triphenylphosphine oxide is a promising scaffold for developing highly efficient dinuclear Pt( $\pi$ ) complexes.

#### Conflicts of interest

There are no conflicts to declare.

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