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Synthesis, crystal structures, optoelectronic properties and resistive memory application of π -conjugated heteroaromatic molecules

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ABSTRACT

Three π -conjugated small molecules **DPBT**, **DEPBT** and **DEPBPy**, based on bithiazole or bipyridine were designed and synthesized. In **DEPBT** and **DEPBPy**, the central core and terminal phenyl groups are connected by C=C double bonds, whereas in **DPBT**, they are combined through C-C single bonds. The effects of the molecular structural changes on their crystal structures, optoelectronic properties and thin film morphologies were comparatively studied. In addition, their resistive memory performance was primarily investigated for the first time. Both **DEPBT**- and **DEPBPy**-based devices exhibit typical binary memory behavior, while **DPBT**-based device presents no memory property. The experimental results confirm that the introduction of C=C bridged bonds into molecules could enhance the molecular interaction, which plays an important role in the molecular arrangement on the thin film, and has a significant contribution to charge transport in memory devices. This work demonstrates that aryl vinyl terminated bithiazoles and bipyridines both are good promising candidates for organic memory storage.

1. Introduction

Organic memory devices (OMDs) have attracted particular attention due to their potential advantages of flexibility, low fabrication cost and ultrahigh-density capacity [1]. Among the potential materials for OMDs, organic small molecules with good memory property have earned significant progress in the past few decades [2]. Organic small molecules with well-defined structures are coalesced via intermolecular force, such as van der Waals force, π - π stacking or hydrogen bonding interactions, instead of strong covalent bonds in inorganic semiconductors [3]. As we all known that enhancing intermolecular interaction could increase the ordering on a molecular scale and lead to distinct solid-state organization, which often results in excellent charge-transport property [3c,4]. Thus, massive efforts have been paid to enhance intermolecular interaction of organic small molecules, such as introducing heteroatoms (N, Si, O or S, *etc.*), C=C double bonds or C=C triple bonds into the conjugated backbone, and great success has been achieved in the promotion of OMDs performance [5].

Bithiazole (BT) and bipyridine (BPy) both are important aromatic heterocycles, which have many advantages including excellent chemical stability, energy level tunability and easily-modified structure [6]. Organic semiconductor materials based on bithiazole or bipyridine moieties have been widely applied in organic fieldeffect transistors (OFETs) [7], organic solar cells (OSCs) [8] and organic light-emitting diodes (OLEDs) [9]. Nevertheless, the use of them in OMDs has still been very limited. To the best of our knowledge, only a few ruthenium complexes with bipyridine as ligands have been used in OMDs [10]. In addition, the resistive memory application of bithiazole-based small molecules has not been reported. Therefore, it is a significant work to develop diversified bithiazole- or bipyridine-based organic memory materials and study the relationship between molecular structure and device performance.

Herein, we synthesized two phenyl vinyl-bridged 4,4'-bithiazole- or bipyridine-based π -conjugated small molecules, namely,



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2,2'-di((*E*)-styryl)-4,4'-bithiazole (**DEPBT**) and 4,4'-di((*E*)-styryl)-2,2'-bipyridine (**DEPBPy**). In contrast, the C–C single bond compound 2,2'-diphenyl-4,4'-bithiazole (**DPBT**) was also prepared, as shown in Scheme 1. The effects of the molecular structural changes on their crystal structures, optoelectronic properties and filmforming abilities were comparatively studied. In addition, their resistive memory performance was primarily investigated. It is proved that the C=C bridged bond extends molecular conjugated backbone and enhances intermolecular interaction, which is beneficial to improve the film quality as well as the electrical memory performance. Both **DEPBT**- and **DEPBPy**-based devices exhibit typical binary memory behavior, while **DPBT**-based devices presents no memory property. This work demonstrates that aryl vinyl terminated bithiazoles and bipyridines both are good promising candidates for organic memory storage.

2. Results and discussion

2.1. Synthetic procedures

The phenyl terminated compound **DPBT** and the intermediate **3** were prepared through the Hantzsch reaction according to the literatures [11]. The styrene-terminated target molecules **DEPBT** and **DEPBPy** were prepared through the aldol-type condensation reaction according to the published procedure [12]. The detailed synthesis routes are depicted in Scheme 1. The molecular structures of **DEPBT** (CCDC 1507339) and **DEPBPy** (CCDC 1580398) have been successfully verified by X-ray single crystal diffraction.

2.2. X-ray crystallogtaphy

To study the molecular structure-packing mode correlations, the single crystals of **DEPBT** and **DEPBPy** were successfully obtained in the process of vacuum sublimation, which could be compared to that of known compound **DPBT** [13]. The single crystal structures of **DEPBT** and **DEPBPy** are shown in Fig. 1a–c. Similar with **DPBT**, **DEPBT** crystallizes in the monoclinic space group P_{21}/c , with unit cell parameters a = 15.3811(6) Å, b = 5.38288(18) Å, c = 11.1802(5) Å, $\alpha = 90^{\circ}$, $\beta = 100.416(4)^{\circ}$, $\gamma = 90^{\circ}$ (Fig. S9 and Table S1 in Supporting Information). As displayed in Fig. 1a, the bithiazole core is in a *trans* coplanar configuration, and the lateral phenyl rings twist from the bithiazole plane in the same direction with small dihedral angle of ~16.22°. In the crystal packing (Fig. 1b), **DPBT** and **DEPBT** molecules both adopt a stacking mode of *J*-type aggregation along *b* axis through intermolecular π - π interactions. For **DEPBT**, the introduction of **C**=**C** double bonds extends the conjugated



Scheme 1. Synthesis routes and molecular structures. Reagents and conditions: (i) Br₂, CHCl₃, reflux, 3 h; (ii) MeOH, reflux, 3 h; (iii) Acetic anhydride, reflux, Ar, 24 h.



Fig. 1. Single crystal structures: (a) The front and side view of **DEPBT**; (b) Intermolecular interaction of **DEPBT**; (c) The front and side view of **DEPBPy**; (d) Intermolecular interaction of **DEPBPy**.

backbone, thereby extending the π -conjugated area and enhancing close packing between the adjacent parallel molecules [4a,4c]. Meanwhile, the neighboring molecular columns of **DEPBT** are linked by multiple C–H···N and C–H···S hydrogen bonds.

As depicted in Fig. 1c, the difference between the molecular structures of **DEPBT** and **DEPBPy** is the central core. **DEPBPy** crystallizes in the triclinic space group P-1, with unit cell parameters a = 9.4384(8) Å, b = 9.4791(7) Å, c = 12.8095(10) Å, $\alpha = 105.343(7)^{\circ}$, $\beta = 95.778(7)^{\circ}$, $\gamma = 117.504(8)^{\circ}$ (Fig. S10 and Table S2 in SI). Surprisingly, there are two different twisted conformations in crystal structures of **DEPBPv**. For both conformational molecules, the bipyridine cores are also in a *trans* coplanar configuration, and the dihedral angles between the bipvridine plane and phenyl rings increase to 25.13° and 24.73°, respectively. In the crystal packing (Fig. 1d), there are also multiple $C-H\cdots\pi$ and C-H…N interactions that exist in neighboring molecules. Different with **DEPBT**, the adjacent parallel molecules of **DEPBPy** only slightly overlaps, resulting in a weak π - π interaction. Different packing modes of the three compounds may lead to the differences in their film-forming and intermolecular charge transfer abilities [4a,5c,14].

2.3. Thermal property

The thermal properties of the three synthesized compounds were evaluated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) measurements, as shown in



Fig. 2. (a) TGA graphs of DPBT, DEPBT and DEPBPy; (b) Normalized UV–Vis absorption spectra of DPBT, DEPBT and DEPBPy in DMF.

(a)

Fig. 2a and Fig. S11. For **DPBT**, **DEPBT** and **DEPBPy**, the decomposition temperatures (T_d , 5% weight loss temperatures) are 276.9, 328.6 and 343.4 °C, the melting temperatures (T_m) are observed at 189.8, 242.8 and 265.0 °C, respectively (Table 1). It shows that all the three compounds exhibit good thermal stability for device applications. T_d and T_m of **DEPBT** and **DEPBPy** are obvious higher than that of **DPBT**, which is probably attributed to the stability of C=C double bonds [4c,15].

2.4. Optical and electrochemical properties

The UV-Vis absorption spectra of the three compounds in DMF solution (10^{-5} M) and thin film are exhibited in Fig. 2b and Fig. S12. In DMF solution, **DPBT** and **DEPBT** both present two absorption bands. The first strong absorption band appears at 270 nm for DPBT and 286 nm for **DEPBT**, owing to the existing phenyl substituents. The second absorption band at 295 nm for DPBT and 335 nm for **DEPBT** is corresponding to a π - π * transition within the conjugated backbone [7c,16]. DEPBPy has one characteristic absorption at 317 nm due to a π - π * transition, with a slight blue-shift compared with **DEPBT**. This result may be due to the different electron donoracceptor effect of the two compounds [10b]. Compared with **DPBT**, the absorption peaks of **DEPBT** and **DEPBPy** are significantly shift to lower energy band, which can be attributed to the extending conjugated backbone by C=C bridged bonds [4]. For all the three compounds (Fig. S12), the thin films show obvious broaden and red-shift peaks compared to that peaks in solution, indicating that a tight packing or molecular aggregation occurred in the solid state [17]. The absorption peaks (λ) and the absorption onsets (λ_{onset}) for estimating the optical band gaps (E_g^{opt}) are summarized in Table 1.

Due to bithiazole derivatives do not present good electrochemical activity in the cyclic voltammetry (CV) [16,17b], the different pulse voltammetry (DPV) was used to examine the electrochemical property for the synthesized compounds, as displayed in Fig. 3a–b. All potentials of the three compounds were calibrated with the ferrcocene/ferrocenium couple (Fc⁺/Fc) as internal standard in the DMF solution of 10^{-3} M with 0.1 M tetrabutylammonium hexafluorophosphate (Bu₄NPF₆) as a supporting electrolyte. In contrast to **DPBT** ($E_{ox} = 0.98$ eV), the other two compounds **DEPBT** ($E_{ox} = 1.30$ eV) and **DEPBPy** ($E_{ox} = 1.47$ eV) exhibit an obvious positive shift in the first oxidation potential. This phenomenon indicates that C=C double bonds increase the molecular electrochemical stability [4c,18]. Oxidation onset potentials (E_{ox}^{onset}), the highest occupied molecular orbitals (HOMOs) and the lowest unoccupied molecular orbitals (LUMOs) are summered in Table 1.

2.5. Morphology of the thin films

To investigate the surface morphology and film microstructure, atomic force microscopy (AFM), scanning electron microscope (SEM) and X-ray diffraction (XRD) measurements were performed on the films of **DPBT**, **DEPBT** and **DEPBPy**. As shown in Fig. 4a–d and Fig. S13, the AFM image of **DPBT** forms long strips and deposits loosely, leading to a non-continuous film, while the quality of

Table 1					
A summary of thermal.	optical and ele	ectrochemical of	data of DPBT ,	DEPBT and	DEPBPy

Compound	T _d (°C)	T _m (°C)	λ_{onset} (nm)	E_{g}^{opta} (eV)	$E_{\rm ox}^{\rm onset}$ (eV)	$E_{\rm HOMO}^{\rm b}$ (eV)	E_{LUMO}^{c} (eV)
DPBT	276.9	189.8	346	3.58	0.83	-5.29	-1.71
DEPBT	328.6	242.8	385	3.22	1.20	-5.64	-2.42
DEPBPy	343.4	265.0	346	3.58	1.39	-5.83	-2.25

^a E_g estimated from the onset absorption ($E_g = 1240/\lambda_{onset} \text{ eV}$).

^b $E_{\text{HOMO}} = -(E_{\text{ox}}^{\text{onset}} - E_{\text{Fc/Fc}}^{+} + 4.8) \text{ eV}.$

^c $E_{\text{LUMO}} = E_{\text{HOMO}} + E_{\text{g}}^{\text{opt}}$.

DPRT - DEPBT 0.**−** 0.2 0.4 0.6 0.8 1.0 1.2 1.4 0.1 0.2 0.5 0.6 0.7 1.6 0.4

(b)

Fig. 3. (a) DPV curves of **DPBT**, **DEPBT** and **DEPBPy** in DMF; (b) DPV curve of ferrocene in DMF.

DEPBT and DEPBPy films are better. This result is in accordance with the SEM measurements in Fig. S14. Subsequently, XRD experiments were carried out to measure the crystalline ordering within the films. According to the XRD patterns (Fig. 4d), only DEPBT film shows obvious diffraction peaks, no XRD peaks are observed from **DPBT** and **DEPBPy** films. These results suggest that **DEPBT** films form a more ordered crystalline arrangement [19]. There are diffraction peaks at $2\theta = 5.85^{\circ}$. 17.53° and 23.48° for **DEPBT**, corresponding to a d-spacing of 15.08 Å and its multiple diffractions (5.05 Å and 3.78 Å), respectively. The obtained dspacing of **DEPBT** is consistent with the long axis (a axis, ~15.38 Å) of unit cell obtained from single crystal X-ray diffraction, indicating **DEPBT** has a close π - π stacking in the condensed aggregate [20]. It is noted that the different morphology of the films could lead different charge transfer ability, which gives a different device performance [4,5b,19].

2.6. Resistive memory evaluation

The memory properties of the studied compounds were primarily determined by investigating the current-voltage (I-V)characteristics of "Al/Organic/ITO" sandwich devices, as shown in Fig. 5a–d and Fig. S15. Considering a thicker film can be avoided a metal-filament effect [21], all devices with an active thickness of about 120 nm was directly selected without further optimization (Fig. 5d). As shown in Fig. 5a–c, the I-V characteristics of "Al/ DEPBT/ITO" and "Al/DEPBPy/ITO" devices both exhibited flashtype binary memory behavior, while the **DPBT**-based device presents no memory characteristic. The resistive memory properties of the three compounds are in correspondence with their filmforming abilities, which shows that the introduction of C=C double bonds will affect the morphology of molecular films as well as the electrical memory performance. In addition, the reproducibility of the memory devices was examined on 50 independent cells of each compound. The proportions of **DEPBT-** and **DEPBPy-**based devices with bistable memory behavior are about 60% and 66%, respectively. The retention performance of "Al/Organic/ITO" devices were evaluated by a reading voltage of +0.1 V in the "ON" and "OFF" states under ambient conditions as depicted in Fig. S16. The current ON/OFF ratios (I_{ON}/I_{OFF}) are about 10^4 for both devices of

ferrocen



Fig. 4. (a) AFM image (20 μ m² × 20 μ m²) of **DPBT**-based film, (b) AFM image (4 μ m² × 4 μ m²) of **DEPBT**-based film, (c) AFM image (4 μ m² × 4 μ m²) of **DEPBPy**-based film, (d) XRD patterns for **DPBT**- and **DEPBPy**-based films on the ITO substrate.



Fig. 5. I–V characteristic of memory devices: (a) DPBT, (b) DEPBT and (c) DEPBPy. And (d) Cross-section SEM image of the device.

DEPBT and **DEPBPy**. The statistic distribution of V_{SET} were provided in Fig. S17. These results denote the reliable memory performance of the devices, which implies that the synthesized compounds could function as a potential storage material [2b,2c,19a]. Based on the experimental results and the literatures [15b,22], the electrical switching behaviors of the synthesized compounds may be illustrated by a possible "traps" mechanism. The detailed memory mechanism of the aryl vinyl terminated bithiazoles- or bipyridinesbased OMDs is under way in our laboratory.

3. Conclusions

Three bithiazole- or bipyridine-based π -conjugated small molecules, **DPBT**, **DEPBT** and **DEPBPy**, were synthesized and

characterized. Their single crystal structures, photophysical and electrochemical properties have been systematically investigated. In addition, their resistive memory characteristics were primarily studied for the first time. Both DEPBT- and DEPBPy-based devices exhibit typical binary memory behavior, while **DPBT**-based device presents no memory property. The experimental results confirm that the introduction of C=C bridged bonds into molecules could enhance the molecular interaction, which plays an important role in the molecular arrangement on the thin film, and has a significant contribution to charge transport in memory devices. This work demonstrates that aryl vinyl terminated bithiazoles and bipyridines both are good promising candidates for organic resistive storage, and could be considered as effective structural units for polymeric materials. The synthesis of a series of new aryl vinyl terminated bithiazoles and their application in organic resistive memory are under way in our laboratory.

4. Experimental section

4.1. Materials

Biacetyl (98%) was purchased from Alfa-Aesar (USA). Ethanethioamide (98%) and benzothioamide (98%) were purchased from Sinopharm Chemical Reagent Co., Ltd. (China). 4,4'-Dimethyl-2,2'bipyridine was purchased from J&K (China). Benzaldehyde (99%) were purchased from Beijing Innochem Science & Technology Co., Ltd. (China). All the solvents were purchased from Tianjinzhiyuan Chemical Reagent Co., Ltd. (China). All the reagents and solvents were of analytical grade without any purification.

4.2. Characterization and instruments

¹H NMR and ¹³C NMR spectra were recorded on 400 and 100 MHz NMR instruments using CDCl₃ and DMSO-d₆ as the solvent and TMS as the internal standard. High resolution mass spectra (HRMS) was obtained on an Agilent LC-MSD-Trap-XCT spectrometer with micromass MS software using electrospray ionisation (ESI). Single crystal X-ray diffraction was conducted on a Bruker SMART CCD diffractor, using graphite monochromated Mo K radiation (λ) 0.071073 nm. Melting points were measured using a WC-1 microscopic apparatus and were uncorrected. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were taken on a STA449F5 analyzer at a heating rate of 10 $^{\circ}C \cdot min^{-}$ and under a N_2 flow rate of 60 mL min⁻¹. UV–Vis spectra was recorded in *N*,*N*-dimethylformamide solution of 10⁻⁵ M with Carry 5000 UV-Vis-NIR spectrometer. Different pulse voltammetry (DPV) was carried out by a CHI66E0 electrochemical analyzer with a three-electrode cell at a scan rate of 50 mV s⁻¹. A Pt wire was used as the counter electrode and an Ag/AgCl electrode was used as the reference electrode. Atomic force microscopy (AFM) images were recorded by a Bruker Multimode 8 microscope in tapping mode under atmospheric environment. Scanning electron microscope (SEM) images were performed on FEI Quanta 250 FEG scanning electron microscope. X-ray diffraction (XRD) patterns were measured on a Bruker D8-A X-ray diffractometer. The 2θ angle was scanned from 5° to 30°. All electrical measurements of the devices were characterized under ambient conditions with an Agilent B1500A semiconductor parametric analyzer.

4.3. Synthetic procedures

4.3.1. 1,4-dibromobutane-2,3-dione (2) [11a]

Biacetyl (5.0 g, 58.1 mmol) was dissolved in CHCl₃ (20 mL), Br₂ (6.0 mL, 116.2 mmol) was added dropwise while maintaining the reaction mixture was refluxed. The mixture was stirred for 3 h. The

solvent was removed under vacuum, and the residue was washed with an excess amount of CHCl₃. The crude product was further purified by crystallized from CHCl₃ and dried to offer **2**, as a light-yellow crystalline solid (7.90 g, 56%). M.p.: 117–119 °C. ¹H NMR (400 MHz, CDCl₃) δ 4.35 (s, 4H).

4.3.2. 2,2'-diphenyl-4,4'-bithiazole (DPBT) [23]

1,4-Dibromobutane-2,3-dione (**2**, 1.22 g, 5 mmol) was added in one portion to a solution of thiobenzamide (1.37 g, 10 mmol) in MeOH (30 mL), and the mixture was refluxed 3 h. The precipitate was filtered, washed with MeOH and dried to offer the product, as an off-white crystalline solid (1.31 g, 82%). The product was sufficiently pure to proceed to the next step. ¹H NMR (400 MHz, CDCl₃) δ 8.07–8.02 (m, 4H), 7.91 (s, 2H), 7.50–7.42 (m, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 168.3, 151.5, 133.5, 130.1, 128.9, 126.6, 115.3. HRMS (ESI): calcd. for C₁₈H₁₃N₂S₂ [M+H]⁺ 321.0515; found 321.0518.

4.3.3. 2,2'-dimethyl-4,4'-bithiazole (**3**) [24]

Compound **3** was obtained in a similar way to **DPBT**, as an offwhite solid in 67% yield and used without further purification. M.p.: 156–158 °C. ¹H NMR (400 MHz, CDCl₃) δ 7.56 (S, 2H), 2.73(S, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 166.2, 150.2, 114.3, 19.2.

4.3.4. 2,2'-di((E)-styryl)-4,4'-bithiazole (**DEPBT**)

Compound **3** (98.1 g, 0.5 mmol) and benzaldehyde (127 µL, 1.25 mmol) were dissolved in 200 µL of acetic anhydride under argon atmosphere. The reaction mixture was heated to reflux for 24 h. After cooling to ambient temperature, the off-white product was isolated by centrifugation and washed with alcohol. The crude product was sublimated twice to give bright white crystal (119.2 mg, 64%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 7.99 (s, 2H), 7.75 (d, *J* = 7.2 Hz, 4H), 7.58 (s, 4H), 7.40 (dt, *J* = 24.9, 7.0 Hz, 6H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 166.7, 150.5, 135.4, 134.5, 129.1, 128.9, 127.4, 121.3, 116.0. HRMS (ESI): calcd. for C₂₂H₁₇N₂S₂ [M+H]⁺ 373.0828; found 373.0831.

4.3.5. 4,4'-di((E)-styryl)-2,2'-bipyridine (**DEPBPy**) [25]

Compound **4** (92.1 g, 0.5 mmol) and benzaldehyde (127 µL, 1.25 mmol) were dissolved in 200 µL of acetic anhydride under argon atmosphere. The reaction mixture was heated to reflux for 24 h. After cooling to ambient temperature, the off-white product was isolated by centrifugation and washed with alcohol. The crude product was sublimated twice to give bright white crystal (138.6 mg, 77%). ¹H NMR (400 MHz, CDCl₃): δ 8.68 (d, *J* = 5.1 Hz, 2H), 8.57–8.56 (m, 2H), 7.58 (d, *J* = 7.2 Hz, 4H), 7.47 (d, *J* = 16.4 Hz, 2H), 7.44–7.37 (m, 6H), 7.36–7.30 (m, 2H), 7.15 (d, *J* = 16.3 Hz, 2H). ¹³C NMR (100 MHz, CDCl₃): δ 156.6, 149.6, 145.8, 136.3, 133.4, 128.9, 128.7, 127.1, 126.2, 121.1, 118.3. HRMS (ESI): calcd. for C₂₆H₂₁N₂ [M+H]⁺ 361.1699; found 361.1705.

4.4. Memory device fabrication

Indium-tin-oxide (ITO) glass was pre-cleaned in turn with distilled water, acetone and alcohol, in an ultrasonic bath for 20 min. By physical vacuum vapor deposition method, the organic molecules that had been sublimated were heated in a quartz crucible, and the vacuum of the deposition system was about 7.0×10^{-4} Pa. The beginning deposition rate of **DPBT**, **DEPBT** and **DEPBPy** were about 0.3-0.7 Å·s⁻¹, and then the deposition rate were controlled by current properly with the film thickness increased. The deposited film thickness was thermally deposited onto organic surface at about 7.0×10^{-4} Pa through a shadow mask. The active device area of ~0.0314 mm² was obtained.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.tet.2020.131471.

References

- [1] (a) S. Gao, X. Yi, J. Shang, G. Liu, R.-W. Li, Chem. Soc. Rev. 48 (2019) 1531-1565;
 - (b) B. Hwang, J.-S. Lee, Adv. Electron. Mater. 5 (2019), 1800519;
 - (c) L. Zhou, J. Mao, Y. Ren, S.-T. Han, V.A.L. Roy, Y. Zhou, Small 14 (2018), 1703126
- (d) C. Wang, P. Gu, B. Hu, Q. Zhang, J. Mater. Chem. C 3 (2015) 10055-10065. [2] (a) Y. Li, C. Zhang, Z. Li, P. Gu, Z. Wang, H. Li, J. Lu, Q. Zhang, J. Mater. Chem. C 7
 - (2019) 3512-3521; (b) Y. Li, X. Zhu, Y. Li, M. Zhang, C. Ma, H. Li, J. Lu, Q. Zhang, ACS Appl. Mater.

Interfaces 11 (2019) 40332-40338; (c) Y. Li, Z. Wang, C. Zhang, P. Gu, W. Chen, H. Li, J. Lu, Q. Zhang, ACS Appl.

Mater. Interfaces 10 (2018) 15971-15979; (d) F. Zhou, B. Wu, H. Dong, Q. Xu, J. He, Y. Li, J. Jiang, J. Lu, Adv. Mater. 29

- 2017) 1604162. [3] (a) Y. Wang, L. Sun, C. Wang, F. Yang, X. Ren, X. Zhang, H. Dong, W. Hu, Chem.
- Soc. Rev. 48 (2019) 1492-1530;
- (b) C. Wang, H. Dong, L. Jiang, W. Hu, Chem. Soc. Rev. 47 (2018) 422-500; (c) W. Yi, S. Zhao, H. Sun, Y. Kan, J. Shi, S. Wan, C. Li, H. Wang, J. Mater. Chem. C 3 (2015) 10856–10861.
- [4] (a) Z. Shan, J. Shi, W. Xu, C. Li, H. Wang, Dyes Pigments 171 (2019), 107675; (b) T.T. Do, H.D. Pham, S. Manzhos, J.M. Bell, P. Sonar, ACS Appl. Mater. Interfaces 9 (2017) 16967-16976;
 - (c) J. Shi, Y. Li, M. Jia, L. Xu, H. Wang, J. Mater. Chem. 21 (2011) 17612-17614.
- [5] (a) K. Zhao, F. Yu, W. Liu, Y. Huang, A.A. Said, Y. Li, Q. Zhang, J. Org. Chem. 85 (2020) 101-107;
 - (b) H. Li, H. Li, Q. Zhang, J. Lu, Org. Electron. 73 (2019) 255-260;
 - (c) Y. Li, Z. Liu, H. Li, Q. Xu, J. He, J. Lu, ACS Appl. Mater. Interfaces 9 (2017) 9926-9934: (d) C. Wang, J. Wang, P.-Z. Li, J. Gao, S.Y. Tan, W.-W. Xiong, B. Hu, P.S. Lee,
 - Y. Zhao, Q. Zhang, Chem. Asian J. 9 (2014) 779-783;
 - (e) G. Li, K. Zheng, C. Wang, K.S. Leck, F. Hu, X.W. Sun, Q. Zhang, ACS Appl. Mater. Interfaces 5 (2013) 6458-6462.
- [6] (a) M. Bazargan, M. Mirzaei, A. Franconetti, A. Frontera, Dalton Trans. 48 (2019) 5476-5490;
- (b) Y. Lin, H. Fan, Y. Li, X. Zhan, Adv. Mater. 24 (2012) 3087-3106; (c) M. Hapke, L. Brandt, A. Lützen, Chem. Soc. Rev. 37 (2008) 2782-2797.
- [7] (a) Y.-L. Rao, A. Chortos, R. Pfattner, F. Lissel, Y.-C. Chiu, V. Feig, J. Xu, T. Kurosawa, X. Gu, C. Wang, M. He, J.W. Chung, Z. Bao, J. Am. Chem. Soc. 138 (2016) 6020-6027;

(b) J. Li, Y. Hu, C. Ge, H. Gong, X. Gao, Chin. Chem. Lett. 29 (2018) 423-428; (c) H. Usta, W.C. Sheets, M. Denti, G. Generali, R. Capelli, S. Lu, X. Yu, M. Muccini, A. Facchetti, Chem. Mater. 26 (2014) 6542-6556.

- [8] (a) M. Jeong, H.C. Jin, J.H. Lee, D.K. Moon, J.H. Kim, Dyes Pigments 173 (2020) 107927:
- (b) M. He, Z. Ji, Z. Huang, Y. Wu, J. Phys. Chem. C 118 (2014) 16518-16525; (c) P. Dutta, W. Yang, S.H. Eom, S.-H. Lee, Org. Electron. 13 (2012) 273–282.
- [9] (a) F. Wang, D. Liu, J. Li, M. Ma, Adv. Funct. Mater. 28 (2018), 1803193; (b) A. Faradhiyani, Q. Zhang, K. Maruyama, J. Kuwabara, T. Yasudab, T. Kanbara, Mater. Chem. Front. 2 (2018) 1306–1309.
- [10] (a) Y. He, Y. Huang, Y. Li, H. Li, Z. Chen, R. Jiang, Inorg. Chem. 58 (2019) 13862-13880; (b) M.-Y. Leung, S.Y.-L. Leung, D. Wu, T. Yu, V.W.-W. Yam, Chem. Eur J. 22

(2016) 14013–14021:

(c) B. Pradhan S. Das, Chem. Mater. 20 (2008) 1209–1211. [11] (a) G. Kurtay, M. Ak, M. Güllü, L. Toppare, M.S. Ak, Synthetic Met 194 (2014) 19-28

(b) G. Gavrel, P. Yu, A. Léaustic, R. Guillot, R. Métivier, K. Nakatani, Chem. Commun. 48 (2012) 10111–10113: (c) H.R. Khavasi, A. Ábedi, V. Amani, B. Notash, N. Safari, Polyhedron 27 (2008)

1848-1854

- [12] E.N. Gulakova, A.G. Sitin, L.G. Kuz'mina, O.A. Fedorova, Russ. J. Org. Chem. 47 (2011) 245-252.
- [13] A.R. Mahjoub, A. Morsali, Z. Kristallogr, NCS 218 (2003) 121-122.
- [14] J. Zhang, J. Jin, H. Xu, Q. Zhang, W. Huang, J. Mater. Chem. C 6 (2018) 3485-3498
- [15] (a) J. Shi, L. Xu, Y. Li, M. Jia, Y. Kan, H. Wang, Org. Electron. 14 (2013) 934–941; (b) M. Jia, Y. Li, M. Huang, J.K. Kim, Y. Liu, S. Cao, Synthetic Met 266 (2020), 116431.
- [16] (a) T. Tao, Y. Peng, W. Huang, X. You, J. Org. Chem. 78 (2013) 2472-2481; (b) T. Tao, B. Ma, Y. Peng, X. Wang, W. Huang, X. You, J. Org. Chem. 78 (2013) 8669-8679 (c) T. Tao, H. Qian, K. Zhang, J. Geng, W. Huang, Tetrahedron 69 (2013)
- 7290-7299 [17] (a) J. Liu, Y. Sun, P. Moonsin, M. Kuik, C.M. Proctor, J. Lin, B.B. Hsu, V. Promarak, A.J. Heeger, T.-Q. Nguyen, Adv. Mater. 25 (2013) 5898-5903; (b) S. Ando, R. Murakami, J.-i. Nishida, H. Tada, Y. Inoue, S. Tokito, Y. 21 Yamashita, J. Am. Chem. Soc. 127 (2005) 14996-14997.
- [18] Y. Ie, M. Nitani, M. Karakawa, H. Tada, Y. Aso, Adv. Funct. Mater. 20 (2010) 907-913.
- [19] (a) X. Cheng, E. Shi, X. Hou, J. Shu, J. He, H. Li, Q. Xu, N. Li, D. Chen, J. Lu, Adv. Electron. Mater. 3 (2017) 1700107; (b) P. Gu, Y. Ma, J. He, G. Long, C. Wang, W. Chen, Y. Liu, Q. Xu, J. Lu, Q. Zhang, J. Mater. Chem. C 3 (2015) 3167-3172; (c) F. Zhou, J. He, Q. Liu, P. Gu, H. Li, G. Xu, Q. Xu, J. Lu, J. Mater. Chem. C 2 (2014) 7674-7680; (d) S. Miao, H. Li, Q. Xu, Y. Li, S. Ji, N. Li, L. Wang, J. Zheng, J. Lu, Adv. Mater. 24 (2012) 6210-6215.
- [20] Y. Li, H. Li, H. Chen, Y. Wan, N. Li, Q. Xu, J. He, D. Chen, L. Wang, J. Lu, Adv. Funct. Mater. 25 (2015) 4246-4254.
- [21] (a) Q. Liu, H. Dong, Y. Li, H. Li, D. Chen, L. Wang, Q. Xu, J. Lu, Chem. Asian J. 11 (2016) 512-519;

(b) Y.-C. Lai, T. Kurosawa, T. Higashihara, M. Ueda, W.-C. Chen, Chem. Asian J. 8 (2013) 1514-1522.

- [22] (a) W. Lin, S. Liu, T. Gong, Q. Zhao, W. Huang, Adv. Mater. 26 (2014) 570-606; (b) J. Shao, B. Cui, J. Tang, Y. Zhong, Coord. Chem. Rev. 393 (2019) 21-36; (c) B. Cui, Z. Mao, Y. Chen, Y. Zhong, G. Yu, C. Zhan, J. Yao, Chem. Sci. 6 (2015) 1308-1315.
- [23] Z. Li, Y. Wang, Y. Huang, C. Tang, J. Xu, X. Wu, H. Yao, Tetrahedron 67 (2011) 5550-5555.
- [24] B. Notash, N. Safari, H.R. Khavasi, V. Amani, A. Abedi, J. Organomet. Chem. 693 (2008) 3553-3557.
- [25] S. Salinas, M.A. Soto-Arriaza, B. Loeb, Polyhedron 30 (2011) 2863-2869.