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By introducing the naphthyl group into the 2,6-positions of anthracene, two different anthracene derivatives, 2,6-di(2naphthyl)anthracene (2,6-di(2-Na)Ant) and 2,6-di(1naphthyl)anthracene (2,6-di(1-Na)Ant) were synthesized. The tiny change of the substitution site of the naphthyl group leads to significant difference of the molecular packing and further exerts great impact on their optoelectronic properties. Thin film fieldeffect transistors for 2,6-di(2-Na)Ant show charge transport mobility up to 2.1 cm²·V⁻¹·s⁻¹, while 2,6-di(1-Na)Ant demonstrates no mobility. Moreover, thin film phototransistors of 2,6-di(2-Na)Ant show a photoresponsivity of 6.9×10³ A·W⁻¹, a high photosensitivity of 2.6×10⁶ and an excellent detectivity of 3.4×10¹⁶ Jones, which are one of the highest performances of thin film organic phototransistors reported till now.

Functional organic semiconductors with specific optoelectronic properties have been intensively investigated in recent years because of easy and various chemical modifications for organic molecules.¹⁻⁶ Among the diverse material systems reported so far, anthracene and its derivatives featuring excellent charge transport behaviours and high emission efficiency,⁷⁻⁹ have been attracting interest as a promising building block for optoelectronic devices, including organic field-effect transistors (OFETs), 10-16 organic light-emitting diodes (OLEDs)¹⁷⁻²⁰ and organic light-emitting transistors (OLETs).²¹⁻²³ To date, a series of anthracene derivatives have been synthesized and their optoelectronic property were investigated. For instance, Kaname Ito et al. first reported the field-

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Substitution site effect of naphthyl substituted anthracene

effect mobility of a series of anthracene dimers and trimers linked at the 2 and/or the 6 position of anthracene.¹⁰ Later, Meng et al. significantly improved the charge transport mobility of anthracene derivatives by incorporating the phenyl vinyl group into the 2,6positions of anthracene.^{12, 13, 24} Recently, by aromatic extension at the 2,6-positions of anthracene with carbon-carbon single bond, Hu et al. developed a series of high mobility ($\mu > 1 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$) emissive organic semiconductors, especially the star molecule 2,6diphenylanthracene (2,6-DPA), which displays charge transport mobility up to 34 cm²·V⁻¹·s⁻¹ and strong solid state emission efficiency of 41.2% and finds its applications in self-driven active organic light-emitting diodes.^{25, 26} 9,10 substituted anthracene derivatives were also widely studied by researchers.²⁷⁻³⁴ However, due to the large steric hindrance or rough thin film morphology, most of the 9,10 substituted anthracene derivatives exhibit rather poor charge transport property in organic thin film transistors.^{28, 30,} 33, 34 Most of the studies about anthracene derivatives focus on functionalization at the end sites (2,3,6,7) or the peri positions (1,4,5,9,10,11,12) of anthracene with functional substituents by tedious chemical reactions.9 Till now, very few studies focused on the modulation of the molecular packing structure and further tuning the optoelectronic property by changing the linking site of the substituents. Therefore, it is interesting to investigate the substitution effect of anthracene derivatives by fine tuning linking site of the substituents in an attempt to deeply understand the

performance optoelectronic materials.

In view of the optical and electrical properties of anthracene, by simply introducing the naphthyl group into the 2,6-positions of anthracene, two anthracene derivatives, 2,6di(2-Na)Ant and 2,6-di(1-Na)Ant were synthesized. The tiny difference of the substitution site of the naphthyl group leads to significant changes of the molecular packing and further exerts remarkable impact on their optoelectronic properties. 2,6-di(2-Na)Ant molecules exhibit good planarity and compact herringbone packing and plentiful intermolecular interactions can be observed while the large torsion angle between the naphthyl group and the anthracene core leads to much looser packing for 2,6-di(1-Na)Ant molecules in aggregation state.

aggregation structure-property relationship and to seek for high

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⁺Electronic supplementary information (ESI) available: Experiments, synthetic details, crystallographic information, calculation details. For ESI and crystallographic data in CIF or other electronic formats see DOI: 10.1039/c000000x

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Thin film field-effect transistors for 2,6-di(2-Na)Ant show charge transport mobility up to 2.1 cm²·V⁻¹·S⁻¹, while 2,6-di(1-Na)Ant demonstrates almost no mobility. Besides, phototransistors of 2,6-di(2-Na)Ant thin film show a highest photoresponsivity of 6.9×10^3 A·W⁻¹, a high photosensitivity of 2.6×10⁶ and a detectivity of 3.4×10^{16} Jones while the on/off ratio for diodes based on 2,6-di(1-Na)Ant thin film is 32.



Fig. 1 UV-vis absorption spectra and calculated frontier molecular orbitals for 2,6-di(2-Na)Ant (a, c) and 2,6-di(1-Na)Ant (b, d).

According to the reported literature, starting from 2,6dihydroxyanthracene-9,10-dione, the two compounds were successfully synthesized after reduction reaction, esterification reaction and Suzuki coupling, successively.^{21, 25} The synthetic details were shown in **Scheme 1**. It should be noted that 2,6di(1-Na)Ant demonstrates much better solubility than 2,6di(2-Na)Ant, indicating its potential application in solution processed organic optoelectronics. The UV-Vis absorption spectra were shown in **Fig. 1**. The absorption of 2,6-di(2-Na)Ant thin film show obvious bathochromic shift (33 nm) compared with solution, which is much larger than that of 2,6-di(1-Na)Ant (13 nm), indicating stronger intermolecular interactions of 2,6-di(2-Na)Ant than 2,6-di(1-Na)Ant. From the UV-Vis absorption for thin film, the λ_{noset} of thin film was 454 nm and 436 nm respectively for 2,6-di(2Na)Ant and 2,6-di(1-Na)Ant. By using the empirical formula $E_g = 1240/\lambda_{onset}$, the energy gap for 2,6-di(2-Na)Ant and 2,6-di(1-Na)Ant was estimated to be 2.73 eV and 2.84 eV. The fluorescence properties were also studied for comparison (**Fig. S1**). The photoluminescence quantum yield (PLQY) of 2,6-di(2-Na)Ant demonstrated to be 61.2% in solution and 29.2% for single crystals, while the PLQY value measured for 2,6-di(1-Na)Ant was 30.8% and 17.5%, respectively for dilute solution and single crystals.

Ultraviolet photoelectron spectroscopy (UPS) was used to measure the energy levels of the two compounds (as thin film state). The HOMO level estimated from UPS was -5.33 eV and -5.61 eV for 2,6-di(2-Na)Ant and 2,6-di(1-Na)Ant, respectively. The UPS energy distribution curve is presented in Fig. S2. The HOMO energy level (2,6-di(2-Na)Ant: -5.25 eV; 2,6-di(1-Na)Ant: -5.66 eV) estimated based on the onset potential and the empirical formula E_{HOMO} =-(E_{ox}^{onset} +4.8- $E_{Fc/Fc+}^{onset}$) eV corresponds well with the UPS measurements. The LUMO energy level (2,6-di(2-Na)Ant: -2.60 eV; 2,6-di(1-Na)Ant: -2.77 eV) was calculated from the bandgap and the HOMO energy level. DFT calculations were also carried out to gain insights into the frontier molecular orbitals of the two compounds, as shown in Fig. 1c and Fig. 1d. The calculation results suggest no significant difference for the frontier molecular orbitals for the two compounds. Thermal gravimetric analysis (TGA) were conducted to test the thermal stability. Due to the same molecular weight, the thermal stability of 2,6-di(2-Na)Ant and 2,6-di(1-Na)Ant show ignorable difference, sublimating at 368 °C and 370 °C respectively, as shown in Fig. S3. The good thermal and chemical stability might lay the foundation of the two compounds for future application in severe environment.

To probe the molecular packing of the two isomers, high quality crystals suitable for x-ray diffraction analysis were grown successfully. Due to the poor solubility in organic solvent, high quality crystals of 2,6-di(2-Na)Ant for x-ray diffraction analysis were grown by Bridgeman method (CCDC: 1901585). Single crystal x-ray diffraction results show that 2,6-di(2-Na)Ant belongs to monoclinic $P2_1/n$ space group with lattice constant a = 5.9799(3) Å, b = 7.5334(4) Å, c = 46.696(3) Å, $\alpha =$ 90°, β = 99.26(3)°, γ = 90°. As shown in **Fig. 2a**, the molecule is as long as 22.356 Å, with a torsion angle of 1.49° between the naphthyl group and the anthracene core, showing very good planarity. The torsion angle of 2,6-di(2-Na)Ant is much smaller than that of 2,6-DPA, indicating much larger conjugation length of 2,6-di(2-Na)Ant. 2,6-di(2-Na)Ant molecules adopt classical herringbone packing mode with a herringbone angle of 49.5°, much larger than 2,6-DPA (Fig. 2b). Every molecule interacts with four neighbour molecules in the herringbone plane with distances between 2.8 Å~2.9 Å (Fig. 2c). The comparable intermolecular interactions and conjugation length indicates the good charge transport property of 2,6-di(2-Na)Ant. Notably, there is not only strong C-H- π interactions in the herringbone layer, but also comparable interactions between adjacent herringbone layers, indicating the better charge transport property in the perpendicular direction than 2,6-DPA and DPVAnt (Fig. S4).^{13, 25} While for 2,6-di(1-Na)Ant, high quality crystals suitable for x-ray diffraction analysis can be obtained from both solvent evaporation method and the physical vapour transport method (CCDC: 2017537). X-ray Published on 02 September 2020. Downloaded on 9/3/2020 7:20:56 AM.

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diffraction results reveal that 2,6-di(1-Na)Ant belongs to monoclinic $P2_t/n$ space group with lattice constant a = 10.5482(2) Å, b = 14.0880(2) Å, c = 7.72920(10) Å, $\alpha = 90^\circ$, $\beta = 99.621(2)^\circ$, $\gamma = 90^\circ$. As shown in **Fig. 2d**, due to the large steric hindrance, the 2,6-di(1-Na)Ant molecule demonstrates a large torsion angle (54.27°) between the naphthyl group and the anthracene core. Though the molecules adopt the traditional herringbone packing mode (**Fig. 2e**) and every molecule interact with four neighbour molecules (**Fig. 2f**), the molecular packing appears much looser than that of 2,6-di(1-Na)Ant (0.682) than that of 2,6-di(2-Na)Ant (0.736).³⁵ Based on the crystal structures, theoretical calculations were also carried out to deeply understand the intrinsic charge transport

difference of the two compounds (**Fig. S5** and **Table. S1**). Both molecules have strong multi C-H- π interactions in the herringbone plane, and 2,6-di(2-Na)Ant molecules demonstrates six charge transfer pathways with much higher transfer integrals (46.98 meV, 32.05 meV, 24.53 meV) than that of 2,6-di(1-Na)Ant (12.69 meV), while the reorganization energy for 2,6-di(2-Na)Ant (145 meV) is smaller in comparison with that of 2,6-di(1-Na)Ant (166 meV). These results suggest better charge transport property of 2,6-di(2-Na)Ant than 2,6-di(1-Na)Ant.

Micro morphology is substantial for the electrical property of the thin film device.³⁶ Self-assembly monolayers and substrate temperature (Ts) can affect the



Fig. 3 Morphology and X-ray diffraction of the thermal deposited thin film of 2,6-di(2-Na)Ant (a-d) and 2,6-di(1-Na)Ant (e-h); atomic force microscopy of 2,6-di(2-Na)Ant thin film deposited at Ts = 80°C (i) and the corresponding step height (j).

nucleation mechanism and further influence the morphology. Here, thin film of 2,6-di(1-Na)Ant and 2,6-di(2-Na)Ant deposited on bare SiO₂/Si, octadecyltrichlorosilane modified SiO₂/Si (OTS/SiO₂/Si) substrate and poly(amic acid) substrate (PAA/ITO) with Ts = r.t. were systematically investigated. As the atomic force microscopy results shown in Fig. 3a-3c, for 2,6-di(2-Na)Ant, the film consistency is well on bare SiO₂/Si, OTS/SiO₂/Si substrate or PAA/ITO substrate, and large

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crystalline grains can be obtained. The sharp x-ray diffraction peaks up to the sixth order indicate the strong crystallinity of the 2,6-di(2-Na)Ant on all substrates (Fig. 3d). The d-spacing (23.33Å) calculated from the first order diffraction (3.78°) is nearly the same as the molecular length (22.356Å) of 2,6-di(2-Na)Ant. Same as 2,6-DPA, the 2,6-di(2-Na)Ant molecules stand almost perpendicularly on the substrate, most favourable for charge transport.³⁷ In contrast, 2,6-di(1-Na)Ant molecules deposited on bare SiO₂/Si substrate (Fig. 3e) show nano/micro porous microstructure while the molecules on OTS/SiO₂/Si substrate (Fig. 3f) appeared to be oil-droplet-like, demonstrating rather poor film consistency and crystallinity. While for the 2,6-di(1-Na)Ant molecules deposited on PAA/ITO substrate, large crystalline dendrite can be obtained, as the optical microscopy shown in Fig. 3g. The XRD results (Fig. 3h) with no diffraction peak also suggest the poor crystallinity of the thin film on bare SiO₂/Si substrate or OTS/SiO₂/Si substrate. For 2,6-di(1-Na)Ant thin film on PAA/ITO substrate, the sharp peak of (2 2 0) suggests the enhanced crystallinity and oriented growth of 2,6-di(1-Na)Ant induced by PAA.38, 39 We choose OTS to further study the influence of substrate temperature on thin film morphology of 2,6-di(2-Na)Ant (Fig. S6). When deposited at 80°C, the crystal domain size reaches $\sim 2 \mu m$, much larger than at lower deposited temperature. Besides, the disordered rod-like crystals disappear as the substrate temperature increases and the crystalline domains exhibit obvious layer-by-layer growth mode, as shown in Fig. 3i. This can be further corroborated by correspondence of the step height (21.74 Å, Fig. 3j) with the molecular length (22.356 Å).





To investigate the electrical properties of the two compounds, thin film field-effect transistors (OTFTs) of the two compounds were fabricated with the vacuum deposited thin film mentioned above in bottom gate top contact geometry. For the compound of 2,6-di(2-Na)Ant, all the devices exhibit typical p type characteristics in ambient conditions (**Fig. S7**). **Table S2** summarized the performance of 2,6-di(2-Na)Ant-based OTFTs prepared at various conditions. The highest hole mobility 2.1 cm²·V⁻¹·s⁻¹ was achieved on OTS/SiO₂/Si substrate at T_{sub} = 80 °C with a threshold voltage of -13 V and an on/off ratio of 10^{7~}10⁸, calculated in the saturation region. The typical transfer and output electrical curve were displayed in **Fig. 4a** and **Fig. 4b**, respectively. The dependence of mobility

on gate voltage was displayed in Fig. S8. While for the compound of 2,6-di(1-Na)Ant, no field-effect mobility can be measured for the device built on SiO₂/Si, OTS/SiO₂/Si or PAA/ITO substrate, which is probably due to the twisted molecular structure and the as-resulted poor thin film continuity and crystallinity.



Fig. 5 (a) The photoresponsivity and photosensitivity of the phototransistors based on 2,6-di(2-Na)Ant thin film (OTS/SiO₂/Si); (b) the I-V curves of 2,6-di(2-Na)Ant thin film (OTS/SiO₂/Si) phototransistors with different light intensity; (c) the photo switch properties of 2,6-di(2-Na)Ant thin film (OTS/SiO₂/Si); (d) the photo switch properties of 2,6-di(1-Na)Ant thin film (PAA/ITO).

Photo response characteristics of the two compounds were systematically studied. For 2.6-di(2-Na)Ant. three-terminal thin film organic phototransistors (OPTs) were prepared by vacuum deposition with bottom gate top contact architecture (Fig. 5a) using gold as the electrodes. The I-V curves of thin film (OTS/SiO₂/Si) phototransistors with different light intensity were displayed in Fig. 5b, demonstrating obvious photo response to white light. The transfer curves of the OPTs show obvious V_T shift as the incident optical power increases, which evidenced the dominant photovoltaic mode of the 2,6-di(2-Na)Ant-based thin film phototransistors.^{40, 41} The photoinduced excitons generate when photons are absorbed, the holes easily flow to the drain electrode whereas the electrons accumulate at the source electrode. The accumulated electrons at the source electrode help to lower the injection barrier and lead to the decrease of the contact resistance, and finally reflect on the shift of the threshold voltage and the increase of I_{DS} . Photosensitivity (P) and photoresponsivity (R) are the key parameters to evaluate the performance of phototransistor, given by the following equations: $P = I_{ph}/I_{dark} =$ $(I_{ill}-I_{dark})/I_{dark}$, $R = (I_{ill}-I_{dark})/(P_{ill}*S)$, where I_{ph} is the photogenerated current, P_{ill} is the incident illumination power on the channel of the device, I_{ijj} the drain current with illumination, I_{dark} the drain current in the dark state and S the area of the channel.42 The photoresponsivity and photosensitivity of the phototransistors are displayed in Fig. 5a. Devices show good response to white light with $R = 6.9 \times 10^3$

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A·W⁻¹, P = 2.6×10^6 and $I_{on}/I_{off} = 10^5$ at illumination intensity of 0.796 mW/cm². The P and R as a function of illumination intensity are shown in Fig. S9. The detectivity of the device is given by the equation $D^* = RS^{1/2}(2eI_{Dark})^{-1/2}$ and calculated to be 3.4×10¹⁶ Jones. As far as we know, the photosensitivity, photoresponsivity and the detectivity are one of the highest performances of thin film organic phototransistors reported till now (Table S3).41, 43, 44 The photo switch properties of the devices based on PAA/ITO and bare SiO₂/Si were also characterized for comparison (Fig. S10). For the thin film on PAA/ITO substrate, the devices show faster response to the light stimuli, indicating better interface contact. As for the compound of 2,6-di(1-Na)Ant, due to the poor gate modulation, two-terminal diodes were fabricated with the thin film prepared above. As shown in Fig. 5d, the devices with film on the PAA/ITO substrate demonstrate photo response with an on/off ratio of 32, while no photo response was observed for the thin film on bare SiO₂/Si or OTS/SiO₂/Si substrate which is probably because of the poor conductivity and the discontinuous morphology of the thin film.

Conclusions

In conclusion, by fine tuning the linking site of the naphthyl group, two different anthracene derivatives were synthesized and characterized. The small change of the chemical structure leads to significant impact on the packing structure, aggregation morphology and further influence the optoelectronic properties. Compared with the large torsion angle of the 2,6-di(1-Na)Ant molecules, 2,6-di(2-Na)Ant molecules demonstrate much better planarity and more compact classical herringbone stacking motif. Due to the better planarity and good compatibility with the substrate (bare SiO₂/Si, OTS/SiO₂/Si and PAA/ITO), 2,6-di(2-Na)Ant molecules demonstrate well film consistency and large crystalline domain. Thin film transistors based on 2,6-di(2-Na)Ant show a mobility up to 2.1 cm²·V⁻¹·s⁻¹ while 2,6-di(1-Na)Ant demonstrate no mobility. Phototransistors of 2,6-di(2-Na)Ant show good response to white light with R = 6.9×10^3 A·W⁻¹, P= 2.6×10^6 , D*= 3.4×10^{16} Jones, $I_{on}/I_{off} = 10^5$, which is among the highest performances reported for thin film organic phototransistors. While the two terminal diodes based on 2,6-di(1-Na)Ant thin film demonstrate weak photo response with I_{on}/I_{off} = 32. These results provide deep insights to understand the aggregation structure-property relationship and to seek for high performance optoelectronic materials.

Conflicts of interest

There are no conflicts to declare.

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Graphical and textual abstract for "Substitution site effect of naphthyl substituted anthracene derivatives and their applications in organic optoelectronics":



Two different naphthyl substituted anthracene derivatives were synthesized. The tiny change of the substitution site of the naphthyl group leads to significant difference of the molecular packing and further exerts great impact on their optoelectronic properties.