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# A Thermally Stable Anthracene Derivative for Application in Organic Thin Film Transistors

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#### ABSTRACT

A novel multifunctional anthracene derivative 2,6-bis(dibenzo[*b*,*d*]furan-3-yl)anthracene) (BDBFAnt), by coupling anthracene moiety with dibenzo[*b*,*d*]furan units attached to its peripheral position, has been designed and synthesized for application in OTFT devices. Introduction of dibenzo[*b*,*d*]furan units in OTFTs is the first example and is highly innovative due to their suitable carrier transport ability, excellent thermal stability, fine film-forming properties, rigid planarity and outstanding fluorescence. The fabricated OTFTs demonstrate a decent hole mobility (3.0 cm<sup>2</sup>/V·s) and favorable green solid fluorescence with excellent thermal stability (up to 220 °C), which expound on the great potential of our material in organic electronics.

KEYWORDS: BDBFAnt, Dibenzo[b,d]furan, Thermal stability, Strong solid fluorescence

## 1. INTRUCTION

During the last couple of decades, organic semiconductors (OSCs), as the active layer in organic thin film transistors (OTFTs), have attracted much attention due to their promising applications in large area and mechanically flexible electronics such as electronic textiles, radio frequency identification tags and transparent electronics.[1-3] Indeed, a remarkable enhancement has been achieved in the performance, especially p-type OSCs,[4] because extensive efforts have been devoted to the improvement of OTFTs for both academic and industrial applications.[5, 6] However, practical application of OTFTs such as integration into consumable electronic equipment requiring exposure to temperatures above 100 °C is still challenging because of their

low mobility and air and thermal instability.[7] To our delight, many groups have been focusing developing the thermally stable OSCs such as polythiophene derivatives,[8] on benzothieno[3,2,b]benzothiophene (BTBT) derivatives,[9-12] linear acenes[13] and so on. Takao [9, 12, 13] et al has made massive research on developing the thermally stable OSCs based on BTBT derivatives. They developed a novel semiconductor dinaphtho[2,3-b:2',3'-f] thieno [3,2,b]thiophene (DNTT)[9] and provided evidence of thermal stability by demonstrating a steady mobility of 1.2 cm<sup>2</sup>/V•s in their TFTs after annealing up to 100 °C. In the same year, they also reported TFTs based on DNTT derivatives (DPh-DNTT)[12] exhibiting a mobility of 1.6  $cm^2/V$ •s with slight decrease in the mobility upon heating the TFTs up to a temperature of 250 °C. In addition, Tian et al[10] developed a BTBT derivative (2,2'-bi[1]benzo-thieno[3,2b][1]benzothiophene) which presented mobility up to 2.12  $\text{cm}^2/\text{V}$ -s and demonstrated high thermal stability as evident from the slight degradation after annealing at temperature up to 250 °C. Impressively, thermally stable OSCs have achieved a great breakthrough and OTFTs based on DPh-BBTNDT [11] displayed high mobility (up to 7 cm<sup>2</sup>/V•s) and excellent thermal stability (up to 300 °C). However, these OSCs have the disadvantages of complex synthesis procedure of the sophisticated reactants, which limits their application in devices.

According to an extensive research to date, anthracene derivatives are regarded as one of the most potential candidates for OTFTs and OLEDs due to their inherent advantages such as ease in chemical modification, excellent air stability, high mobility and high fluorescence.[6, 14-19] In addition, it is known that an effective method to obtain OSCs with lager  $\pi$ -conjugation and excellent thermal stability is oligomerization of small acenes.[10] Taking this into consideration, we intend to introduce thermally stable fused aromatic rings such as dibenzofuran (DBF), dibenzothiophene (DBT) [20, 21] into the anthracene core. Thereto, DBF moiety is widely

applied in organic semiconductors particularly phosphorescent organic light-emitting diodes (PhOLEDs)[22] due to its suitable carrier transport ability, excellent thermal stability, fine filmforming properties, rigid planarity and outstanding fluorescence.[23-26] However, to our knowledge analogous research on the OTFT devices is not yet reported, hence we take the lead to combine anthracene moiety with DBF units to design the OSC for OTFT devices and aim to increase the thermal stability and fluorescence. In this context, BDBFAnt, a new kind of anthracene derivative, has been designed and synthesized via simple and efficient Suzuki reaction. BDBFAnt exhibits high thermal stability, outstanding fluorescence and fine mobility. From this point of view, BDBFAnt is regarded as a potential candidate for multifunctional electronic devices requiring high mobility and outstanding fluorescence, such as organic light emitting transistors which are multifunctional devices combining the current switching amplification of OFET and the light emission functionality of OLED [27, 28] and electrically pumped organic lasers [29-31] that require organic semiconductors with high fluorescence to generate optical excitation and high carrier mobilities to ensure fast transport of the injected charges to the recombination zone promoting further charge injection. The fabricated devices are still far away from practical application due to shortage of organic semiconductors with both high mobility and strong fluorescent emission. It remains a challenge to obtain OSCs with both high mobility and good fluorescent emission because of the contradictory structural requirements. Achieving high mobility always demands strong  $\pi$ -interactions and planar structure, while obtaining high emission typically requires suppressing  $\pi$ -interactions to avoid exciton quenching.[32]

Here, we envisioned a planar molecular structure by specifically designing the dibenzofuran anthracene compound with dibenzo[b,d] furan moiety artificially substituted at 3 position. The

rational motives to design the molecules are: (i) to use the DBF units at 2-, 6- position of anthracene core in order to extend the molecular  $\pi$  conjugation and tune the molecular packing, (ii) to increase the fluorescence and the thermal stability of anthracene core.

Scheme 1 illustrates the synthetic route to our new organic semiconductor material. The final yellow colored product was synthesized via Suzuki coupling and was purified three times by gradient sublimation. The compound was characterized single crystal X-Ray analysis, elemental analysis and high-resolution mass spectrometry (see more details in ESI)



Scheme 1 synthetic route to compound BDBFAnt

# 2. RESTULT AND DISCUSSION

The detailed synthesis routes, characterization of BDBFAnt and the OTFT device fabrication steps are described in the supporting information.

The thermal properties of BDBFAnt were investigated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). As shown in Fig. S1 (A), the TGA curves illustrate the

decomposition temperature (5% weight loss) of ~ 460 °C under both nitrogen and atmosphere environment, which is an evidence of good thermal stability.[12] Fig. S1 (B) shows the DSC of BDBFAnt, there is no peak in the DSC scan from 25 to 360 °C, whereas, sharp melting peaks at ~ 390 °C can be seen which correspond to the phase transition (Fig. S1 (C)). These results exhibit excellent thermal stability of our material.



**Fig. 1** (A) UV-Vis (red) and PL spectra (blue) of BDBFAnt in thin films (B) Cyclic Voltammetry of BDBFAnt in thin films.

The optical properties of the thermally evaporated thin films of BDBFAnt were studied using UV-Visible absorption and photoluminescence (PL) spectra. As depicted in Fig. 1(A), the energy band gap of 2.61 eV is evaluated from the absorption onset of 475 nm. The peak positions of UV absorption from 400 to 500 nm are similar to anthracene. Whereas, the new peaks in the wave length range of 300 to 380 nm is ascribed to the absorption of end-capping dibenzofuran units. In PL spectra (Fig. 1 (A)), BDBFAnt thin films exhibit strong greenish fluorescence and the florescent quantum efficiency (QE) of about 49 % (Fig. S2). As shown in Fig S3 the PL spectra of BDBFAnt in the thin film shows red-shifted emission as compared to that of anthracene. It is known that the anthracene exhibits blue fluorescence and the PLQY of anthracene in ethanol is

around 27% [33], so the introduction of DBF groups significantly increase the intensity of fluorescence. But, the PLQY of BDBFAnt is lower than previously reported 9,10bis(dibenzo[b,d]furan-4-yl)anthracene [34], due to its planar structure and strong  $\pi$ -interactions which generate exciton quenching.[6, 35] The electrochemical properties of the thin films on GCE in CH<sub>2</sub>Cl<sub>2</sub>/TBAPF<sub>6</sub> solution were analyzed. As shown in Fig. 1 (B), the HOMO level is calculated to be -5.78 eV, about 0.7 eV more negative than pentacene (-5.1 eV), 0.4 eV more negative than bi-BTBT and at the same level as anthracene (~5.79 eV),[15] providing the evidence of higher oxidation stability. It has been proved that transistors based on materials with HOMO level range from -4.5 to -5.0 eV are unstable when exposed to ambient atmosphere because these materials are susceptible to oxidative doping.[36] The optical and electrochemical properties of BDBFAnt are summarized in Table S1.

With the intention to gain insight into geometry and stabilization effects of DBF moiety, we grew the single crystal via vapor deposition and, gratifyingly, the single crystal X-ray diffraction results are demonstrated in Fig. 2 (CCDC 1506173). The results reveal that the molecular packing of BDBFAnt is a typical herringbone structure, possessed by most of the high mobility organic semiconductors. From the X-ray Crystallographic data, the BDBFAnt crystal belongs to monoclinic I/ (2/c) space group with crystal parameters of a = 5.981 Å, b = 7.485 Å, c = 53.68 Å,  $\beta = 91.71^{\circ}$ . As shown in Fig. 2(D), C-H- $\pi$  interactions (distance: 2.88 Å) are observed between every single molecule and its adjacent molecules. However, the anthracene crystal belongs to monoclinic P/ (2<sub>1</sub>/n) space group with lattice constants a = 8.553 Å, b = 6.021 Å, c = 22.333 Å,  $\beta = 124.54^{\circ}$ . Moreover, multi C-H- $\pi$  interactions (distance: ~ 2.78 Å) are observed between every anthracene molecule and its nearest molecules.[37] The BDBFAnt molecule displays a nearly planar, symmetric molecular geometry, which leads to effective expansion of  $\pi$ -conjugation, also

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proved by an almost planar backbone structure of BDBFAnt with very low torsion angles of  $6.01^{\circ}$  and  $1.72^{\circ}$ . Additional  $\pi$ -conjugation, coplanarity and strong intermolecular interactions play the leading role in the carrier charge mobility of organic semiconductors.[38]



**Fig. 2** (A) Molecular length and two torsion angles (B) packing structure projected along a-axis (C) herringbone packing (D) short contacts in BDBFAnt

To study the charge transport properties, OTFT devices were fabricated with a top contact geometry. The thin films (40 nm) were deposited on Si/SiO<sub>2</sub> (200 nm) substrates modified by OTS at different substrate temperatures via vacuum deposition and then gold was evaporated as the source/drain electrodes. The performance of devices was measured by Angilent 1500B parameter analyzer under ambient conditions. Representative transfer and output curves at different substrate temperatures are demonstrated in Fig. 3 and the results are summarized in Table 1. From the results it is clear that the substrate temperature has a significant influence on the performance of BDBFAnt TFTs. The devices grown on OTS-treated substrate at 80 °C exhibit the best field effect mobility of  $3.0 \text{ cm}^2/\text{V}$  s, current on/off ratio ( $I_{on}/I_{off}$ ) of about  $8 \times 10^5$ , and the threshold voltage ( $V_T$ ) of nearly -37 V. We notice that the work function of gold was noticeably higher than the HOMO level, creating a noteworthy contact resistance between the semiconductor layer and the gold electrode, which restricts the device performance particularly the linear regime[10] and result in a relatively large threshold. With the increased substrate temperature, the performance of OTFTs enhances sharply under 80 °C. However, when the

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substrate temperature approaches 100 °C, the mobility of BDBDFAnt TFTs shows slight degradation. As shown in Fig 3(E)-(H), the output curves of the transistors exhibit decrease in current as the drain voltage, this behavior was ascribed to the existence of  $O_2$  and  $H_2O$  at the interface of semiconductor-dielectric.[39]



**Fig. 3** Typical transfer and output curves of OTFTs at (A) (E)  $T_{sub}=25\Box$ , (B) (F)  $T_{sub}=60\Box$ , (C) (G)  $T_{sub}=80\Box$ , (D) (H)  $T_{sub}=100\Box$ 

Table 1 Performance parameters of BDBFAnt OTFTs

T <sub>sub</sub>	$\mu_{max}$ (cm <sup>2</sup> /V·s)	$\mu$ (cm <sup>2</sup> /V·s)	V <sub>th</sub> (V)	$I_{on} / I_{off}$
25 °C(R.T.)	0.89	$0.59\pm0.22$	-35 to -45	$10^{5} - 10^{6}$
60 °C	1.63	$1.21\pm0.28$	-35 to -45	$10^{5}$ - $10^{6}$
80 °C	3.01	$2.53\pm0.36$	-35 to -45	$10^{5}$ - $10^{6}$
100 °C	2.04	$1.57\pm0.30$	-35 to -45	$10^{5}$ - $10^{6}$

Since morphology, crystallinity and preferred orientation are the crucial elements influencing the charge carrier transport in OTFTs, the thin films of BDBFAnt deposited on OTS-modified Si/SiO<sub>2</sub> were characterized by atomic force microscope (AFM) and X-Ray diffraction (XRD). As illustrated in Fig. 4 (A)-(D), the substrate temperatures have a significant effect on the surface morphology, which are supposed to affect the corresponding charge transport properties of the films. The step heights of the crystalline terraced structure in the AFM images can be clearly seen which is in good agreement with the extended molecular layers, indicating that the thin films of BDBFAnt are grown layer by layer. Likewise, the obtained d-space (2.63 nm) is nearly consistent with the length of the BDBFAnt molecule (2.58 nm), well in agreement with the results of AFM images. The results indicate that the molecules are almost perpendicular to the SiO<sub>2</sub> substrate and form a packed structure, which is actually a widespread characteristic of most of the high mobility organic semiconductors. We can see the dendrite growth in the AFM images, but with the increase in substrate temperature, the dendrites become less obvious and the films of BDBFAnt present large grain sizes with well-defined terraces with the increase in substrate temperature. However, when the  $T_{sub}=100$  °C, we notice that some subtle cracks (Fig. S4 white arrow) appear in the thin films, which may be the reason for the degradation of mobility. Additionally, the grain boundaries become larger and deeper, which could be unfavorable for charge transport.[40] As shown in Fig. 4 (F), majority of the main peaks in the thin films XRD patterns can be identified as (0 0 h) planes which are in agreement with the single crystal X-Ray diffraction. Similarly, as the substrate temperature increases, the peaks (004), (006) and (008) become much more intense than the peak (0 0 2), indicating that the orientation of the molecules is optimal, in favor of carrier transport. At the same time, we notice that the intensities of the peaks such as (1 1 2) and (1 1 4) increase when the substrate temperature is raised, illustrating

that there is another new orientation. Furthermore, it is easy to observe that the intensity of the peaks at  $T_{sub} = 80$  °C is the lowest among the peaks at other substrate temperatures. The best performance of the devices at  $T_{sub} = 80$  °C, indicates that the new orientation of the molecules is not suitable for carrier transport. These changes may be the reason for the charge mobility value exhibits the best at 80 °C and decreases when the substrate temperature goes either direction.[41]



**Fig. 4** AFM images of the semiconductor films deposited on OTS-treated wafer substrate at (A) R.T., (B) 60 °C, (C) 80 °C, (D) 100°C (E) step height of terrace at 80 °C (F) XRD of the thin films deposited at R. T., 60 °C, 80 °C, 100°C

The thermal stability of BDBFAnt TFTs was investigated by annealing the devices ( $T_{sub} = 80 \text{ °C}$ ) in a glove box at selected temperatures for 30 minutes. Then the devices were allowed to cool to R.T. and measurements were performed. As shown in Fig. 5 (A), at annealing

temperatures below 180 °C, no obvious degradation in mobilities is observed. Whereas, at annealing temperature of 220 °C, the performance of OTFTs based on BDBFAnt is slightly decreased (from 3.0 to 1.8 cm<sup>2</sup>/V•s), which proves the excellent thermal stability of BDBFAnt-based devices. The performance of BDBFAnt TFTs show dramatic decrease at the annealing temperature up to 240 °C. Consequently, BDBFAnt is among one of the highest mobility materials with excellent thermal stability for OTFTs based on anthracene derivatives.

In order to investigate the effect of annealing temperature on the polycrystalline thin films of BDBFAnt, the thin films were studied using XRD, AFM, PL, UV-Vis respectively. As shown in Fig. 5 (B), when the annealing temperature is below 180 °C, it can be clearly seen that the XRD data exhibit almost the same peaks. Nevertheless, the intensity of peaks is drastically decreased after the annealing treatment up to 220 °C, suggesting that the crystalline quality was not good. The results reveal that the degradation of mobilities is attribute to poor ordering of the thin films. Interestingly, we found that annealing at 180 °C did not cause any spectral change in the PL spectra, however when the annealing temperature is increased to 220 °C, the intensity of ~610 nm is increased in Fig. 5 (C). Similarity the UV-Vis spectra (Fig. 5(D)) illustrate a corresponding change. The appearance of such an additional emission band upon heating has been attributed to spontaneous aggregation and excimer formation as the origin of low-energy emission [42-44], which impeded the mobility of charge carriers. Likewise, as shown in Fig. S5, the terraces of BDBFAnt films are almost the same at the annealing temperature below 220 °C. Whereas we notice that BDBFAnt molecules are gradually aggregated into thicker grains resulting a significant change in the morphology when the annealing treatment is above 220 °C. It is evidenced by analyzing the height profiles of grains in Fig. S6. With the increase of annealing temperature, all BDBFAnt molecules exhibit aggregation more quickly, leading to rather thicker

grains.[45] These phenomena of aggregation into thicker grains could result in the degradation in the performance of OFETs based on BDBFAnt.



**Fig.5** (A) The effect on mobility of different annealing temperature (B) The effect on XRD of different annealing temperature (C) The effect on PL of different annealing temperature (D) The effect on UV of different annealing temperature

#### **3. CONCLUSION**

In summary, by architecturally introducing the substituent at 3 position, we have designed and synthesized a novel semiconductor material based on anthracene derivative for application in OTFTs. To the best of our knowledge introduction of DBF moiety is the first ever example to exhibit excellent thermal stability not only as a compound itself but also as thin films used in OTFTs, which gives a particular research direction. Moreover, a decent hole mobility of up to 3.0 cm<sup>2</sup>/V•s is achieved for the thin film transistors with excellent thermal stability at temperature up to 220 °C. Furthermore, the BDBFAnt demonstrates a green fluorescence when exposed at the wavelength of 365 nm. Therefore, due to this property of high mobility and strong fluorescence along with excellent thermal stability, BDBFAnt may be employed in multifunctional device applications in future.

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found at in the online version.

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- A novel anthracene derivative BDBFAnt has been designed and synthesized for application in OTFT devices.
- BDBFAnt exhibits excellent thermal stability not only as a compound itself but also as thin films for OTFTs
- The BDBFAnt demonstrates a decent hole mobility with excellent thermal stability.
- The BDBFAnt demonstrates outstanding solid green fluorescence.