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REVIEW ARTICLE

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Progresses in organic field-effect transistors and molecular electronics

Abstract In the past years, organic semiconductors have been extensively investigated as electronic materials for organic field-effect transistors (OFETs). In this review, we briefly summarize the current status of organic field-effect transistors including materials design, device physics, molecular electronics and the applications of carbon nanotubes in molecular electronics. Future prospects and investigations required to improve the OFET performance are also involved.

Keywords organic semiconductors, organic field-effect transistors (OFETs), molecular electronics

1 Introduction

Organic field-effect transistor (OFET) is a three-terminal device whose characteristics can be modulated by the electrical field. It is composed of organic conjugated molecules as active channels, inorganic or polymer insulators as dielectric layers and metals or conductive polymers as electrodes. Since the first report in 1986 [1], OFETs have drawn more and more attention because of their low cost, flexibility as well as the capability for large area preparation. Bells Lab, IBM cooperation and many research institutes have ventured into this field [2, 3]. In the past decade, many novel organic semiconductors were synthesized and device performances were significantly improved. OFETs are becoming a significant and much discussed topic in organic electronics. Here, we present a few important progresses in organic field-effect materials, device physics and molecular electronics mainly based on our own results.

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2 Device physics of OFETs

2.1 Basic principles of OFETs

An OFET is a kind of device that controls the on/off states by applying a voltage on the gate electrode to form charge accumulation at the organic-dielectric interface. When the source-drain voltage V_{ds} equals zero, no current forms in the channel as the charges along the channel are uniformly distributed. Once $V_{ds}<V_{gs}-V_{th}$ (here V_{gs} is the gate voltage and V_{th} is defined as the threshold voltage), charges get to be a gradient distribution along the channel so that the current can be controlled by the applied source-drain electric field. Here, the device is operated in the linear mode, I_{ds} can be given by Ref. [4],

$$I_{ds}= \mu [(W/L)C_i (V_{gs}-V_{th}) V_{ds}-1/2V_{ds}^2]$$

If $V_{ds} >V_{gs}-V_{th}$, the device is operated under saturation region, I_{ds} is then given by,

$$I_{ds}= \mu (W/2L)C_i (V_{gs}-V_{th})^2$$

Here, μ is the field-effect mobility, W is the channel width, L is the channel length, C_i is the capacitance of the insulator per unit area. The structure and output characteristics of OFET resemble that of inorganic MOS transistors; one difference lies in the mechanism of the charge transportations. For example, for p type inorganic transistors, the minority carriers accumulate at the interface of the channel and the insulator when inversion occurs; however, organic field-effect devices have no inversion.

2.2 Parameters of OFETs

The most important parameters of OFETs are the mobility, on/off ratio, threshold voltage and sub-threshold slope. The mobility determines the possible bit speed of the organic integrated circuits, mobility in the magnitude of 1 cm^2/V s is essential for high-speed commercial applications. For other possible applications, mobility of at least 0.01 cm^2/V s and on/off ratio larger than 10^3 are required [5]. The field mobility is related to the structures of the molecules, especially

the conjugated degree and the aggregate status of the organic active materials [6]. The purity of the materials, the quality of source and drain contacts as well as the temperature have significant influences on the device performance [7]. Up to now, pentacene has been the most ideal material for OFETs; the highest performance of pentacene-based OFET was reported by 3M cooperation with mobility up to 3.3 cm^2/V s and on/off ratio exceeded 10^6 by depositing pentacene films on the alumina substrate with phosphonohexadecane SAM layers [8]. Further investigations for the radio frequency identification (RFID) tags were also taken based on this material [9].

On/off ratio is also critical for the applications in organic digital circuits, electronic papers and OFET-driven OLEDs [10] (e.g., for OLED driving, the mobility should be larger than 1 cm^2/V s and $I_{on}/I_{off} > 10^8$, V_{th} be closed to 0 V). Material purification, device optimization and the post-processing such as annealing are the most common ways to improve the I_{on}/I_{off} [11]. Sirringhaus et al. [12] and Lin et al. [13] reported OFETs based on bis(dithienothiophene) and pentacene with I_{on}/I_{off} as high as 10^8. The threshold voltage V_{th} is determined by trap density near the organic-insulator interface and the quality of source drain contacts. Recently, it has been found that the dipole layer and the channel modification can also change the threshold voltage [14].

3 Organic semiconductor materials

Organic transistors can be fabricated by thermal vacuum deposition, spin coating, screen printing, pattern stamping using different organic semiconductors, insulators and electrodes [15]. For years, chemists have been working toward the designing and synthesis of novel organic semiconducting materials with high mobility and superior stability. Early studies on OFETs focused on oligothiophenes and polythiophenes, the earliest OFET was just fabricated by electrochemical polymerization of thiophenes [1]. Other conjugated small molecules, oligomers and their derivatives based on thiophenes, pyrroles, carbazoles, benans, fluorines and condensed-nuclei aromatics showed organic field-effect characteristics.

Organic small molecules are required to be conjugated and have coplanar conformation atoms to get better stacking for high transistors performance. The most ideal material so far is pentacene with high mobility above 1 cm^2/V s. In recent years, more and more materials with high mobility have been synthesized. Li et al. [16] reported OFETs based on indolo[3, 2-b]carbazoles with mobility of 0.2 cm^2/V s, Merlo et al. [17] found the mobility of hybrid acene-thiophene molecules was as high as 0.5 cm^2/V s, Naraso et al. [18] fabricated OFETs based on tetrathiafulvalene (TTF) derivatives with mobility of 0.42 cm^2/V s. As for n type materials, Haddon et al. [19] fabricated OFETs based on C_{60} with electron mobility of 0.08 cm^2/V s in early years. In 2001, Patrick et al. [20] reported perylene derivatives with mobility of 0.6 cm^2/V s. In 2005, Ando et al. [21] and Letizia et al. [22] reported fluorine-substituted n type organic semiconductors with electron mobility up to 0.3 cm^2/V s and 0.51 cm^2/V s, respectively. Not long ago, Gundlach et al. [23] synthesized an n type material with mobility as high as 0.6 cm^2/V s and applied it in logic gates. Chesterfield et al. [24] elevated the mobility of perylene derivatives to 1.7 cm^2/V s, Ando et al. [25] also reported another thiazole oligomers based n type material with high mobility up to 1.83 cm^2/V s. Besides, some organic polymers are also semiconductors, which are more easily synthesized than small molecules. Polymer semiconductor devices can be prepared by spin coating, inject printing and screen printing. Generally, conjugated polymers form highly disordered polycrystalline films, the mobility is relative low (around 10^{-7} to 10^{-2} cm^2/V s) as hopping between polymer chains determines the charge carrier transportations [26, 27]. Recently, Roy et al. [28] reported polymer-based OFETs with mobility as high as 10^{-1} cm^2/V s, but it was still one magnitude lower than small molecules.

We have synthesized tens of organic materials and their derivatives, such as asymmetrically substituted phthalocyanine, ladder tetraazapentacenes [29], cyclo pyrroles [30, 31], condensed pentathienoacene [32, 33]. We also fabricated OFETs [34] and nano electronic devices [35] based on these materials.

3.1 Pentacene-like semiconducting compounds

So far, pentacene is thought to be one of the most ideal organic semiconductors; chemists and physicists have investigated it from molecule structure to the device physics in order to improve its mobility. It is widely accepted that good π-π conjugated and stronger molecule-molecule interactions are essential for higher-mobility materials. Thus, substitutional pentacene-like molecules are expected to have superior field-effect performances.

We synthesized ladder tetraazapentacenes semiconductors (Fig.1(**1**)) with mobility up to 0.02 cm^2/V s [29]. Recently, another pentacene-like semiconducting compound pentathienoacene was synthesized as shown in Fig.1(**2**) by collaborating with Prof. Jingui Qin's group at Wuhan University. The energy gap E_g of pentathienoacene is 3.29 eV

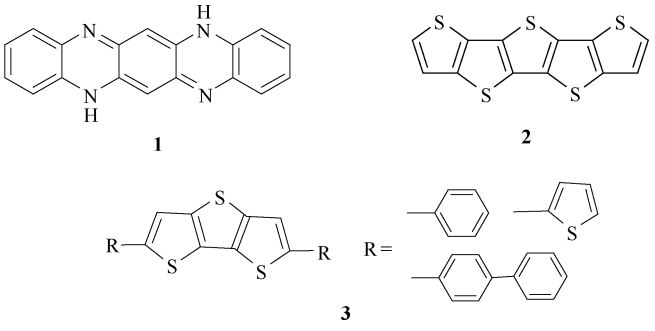


Fig. 1 Pentacene-like semiconducting molecules
(**1**) ladder tetraazapentacenes [29], (**2**) pentathienoacene [33] and (**3**) dithieno [3,2-*b*:2′,3′-*d*]thiophene [36]

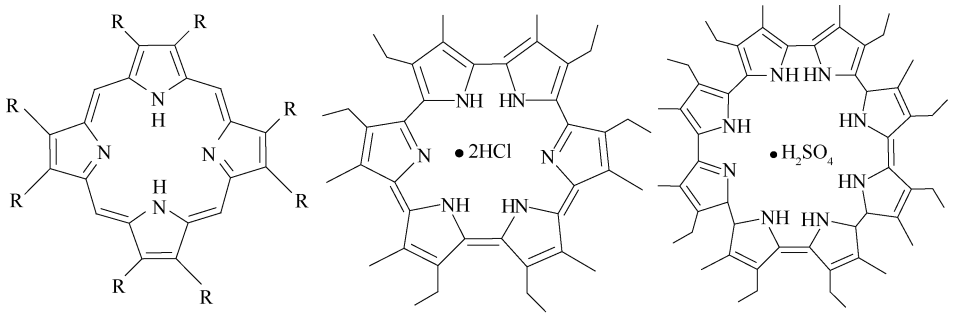
(for pentacene it is only 1.85 eV), the decomposition temperature is 272ºC and the OFET mobility is 0.045 cm^2/V s, indicating that pentathienoacene is a very promising material for high-performance transistors with excellent stability [33]. We further fabricated transistors based on derivatives of dithieno[3,2-*b*:2′,3′-*d*]thiophene (Fig.1(**3**)). These fused thiophene derivatives exhibit excellent field-effect performances, with mobility as high as 0.42 cm^2/V s, indicating fused thiophenes are a class of ideal building blocks for high organic-field materials [36].

3.2 Ring- and star-shaped compounds

Ring-shaped π conjugated molecules are a special class of organic semiconductors that provide the possibility of conjugation dimensions extensions. Ring-shaped molecules are important as they can easily be assembled to fabricate optical and electrical devices. We designed a series of cyclopyrroles (Fig.2) and using these we prepared Langmuir-Blodgett (LB) film based OFETs. The mobility of cyclo[4]pyrrole (Fig. 2(**4**)) and cyclo[8]pyrrole (Fig. 2(**6**)) were as high as 0.014 cm^2/V s and 0.68 cm^2/V s, respectively [30, 31].

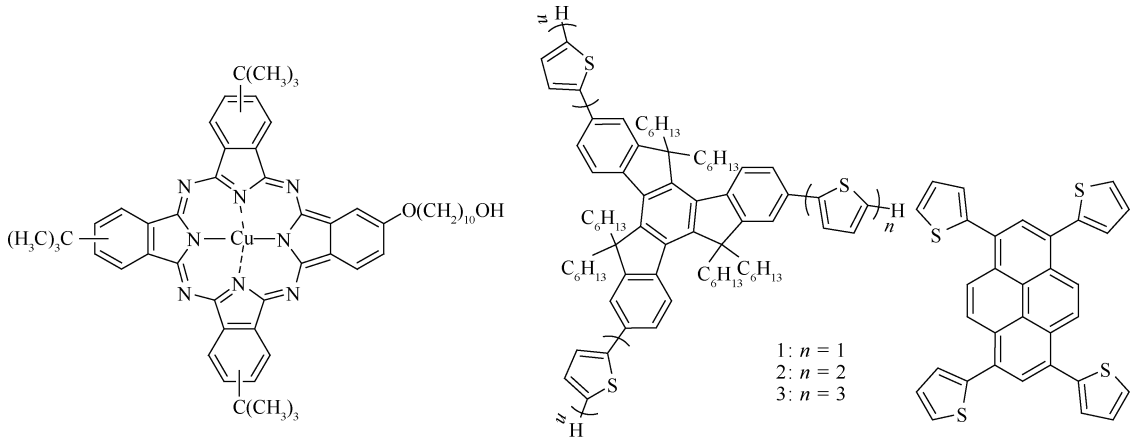
Organic complex compounds are drawing more and more attention in the case of optical and electrical devices. Phthalocyanine is a typical dye and optical electrical material that has been widely used in optical electronic devices. Its unique thermal stability makes it an ideal field-effect material that is suitable for potential commercial applications. We fabricated film [37, 38] and single crystal [39] phthalocyanine-based OFETs and sensors. The solubility of phthalocyanine is very limited, so we synthesized asymmetrically substituted phthalocyanines (Fig.3(**7**)) and further fabricated LB film based diodes [40], transistors [41] and sensors [42].

Substitutions have great influence on the optical and electrical properties of organic semiconducting molecules. We collaborated with Prof. Jian Pei at Peking University and synthesized star-shaped thiophene derivatives (Fig.3(**8**)) [32]; the mobility of the device was 1.03×10^{-3} cm^2/V s. Recently, we further synthesized pyrene-based butterfly-shaped molecules and used them as organic-field active layers (Fig. 3(**9**)); the mobility of the device was 3.7×10^{-3} cm^2/V s [43]. These molecule design concepts establish the foundation for the further designing of two- and three-dimensional conjugated molecules.



4 **5** **6**

Fig. 2 Cyclopyrroles [30, 31]
(**4**) cyclo[4]pyrrole, (**5**) cyclo[6]pyrrole and (**6**) cyclo[8]pyrrole



7 **8** **9**

Fig. 3 Modified organic field-effect materials
(**7**) asymmetrically substituted phthalocyanine [38], (**8**) Star-shaped [32] and (**9**) butterfly-shaped [43] organic semiconducting compounds

4 Novel field-effect transistors

4.1 Field-effect transistors based on single molecules

It is an important field that utilizes single molecules to fabricate devices with determined properties; computing with molecules has been a dream of the chemist and the physical scientist for years [44]. Studying the charge transfer inner as well as inter the molecules to find out the interactions between atoms and electrons, is very fundamental and important for fabricating single molecules devices (e.g., molecule rectifiers, molecule wires and molecule memories) and developing molecules electronics. We have also done some basic work in this field; recently we self-assembled thioacetyl-end-functionalized poly(para-phenylene ethynylene) between gold nano-electrodes, and observed the light-switching and transistor characteristics [35].

4.2 Organic field-effect transistors based on LB films

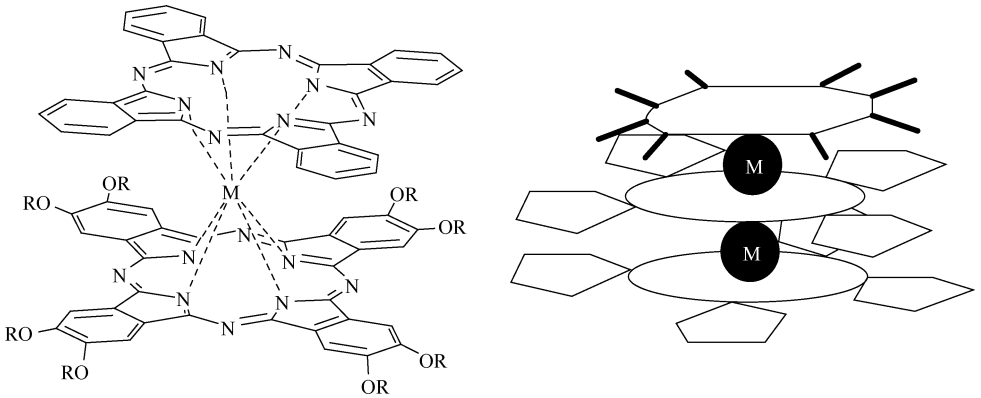
Langmuir-Blodgett (LB) technology is one of the most promising techniques for preparing thin films with the thickness of a few nanometers (a monolayer) on a solid substrate. It enables precise control of the monolayer thickness, homogeneous deposition of highly organized monolayer over large areas and allows making multilayer structures with varying layer composition. It has been widely used in the production of electrically, optically and biologically active components on a nanometer scale. The mobility of early OFETs based on LB films was very low, around 10^{-7}～10^{-3} $cm^2/V\,s$ only [45, 46]. We worked together with Prof. Jianzhuang Jiang's group at Shandong University and synthesized heteroleptic bis (phthalocyaninato) rare-earth complexes (Fig. 4(**10**), (**11**)); the mobility of OFETs based on the LB films using the two compounds was 6.4×10^{-4} $cm^2/V\,s$ and 1.7×10^{-3} $cm^2/V\,s$ [47]. Recently, we designed amphiphilic tris(phthalocyaninato) rare-earth triple-decker complexes and their field mobility reached 0.24～0.60 $cm^2/V\,s$. This experiment enhanced the performance of LB film based OFETs to a new level [48].

4.3 Carbon nanotube based transistors

Carbon nanotubes (CNTs) have enticed researchers since their discovery in 1991, offering an impressive combination of high strength and important electric properties. Strictly speaking, carbon nanotubes do not belong to the class of organic semiconductors, but they are often considered to be another aspect of organic electronics due to their significance in the field of micro- and nano-scale devices. Recently, CNTs have gained importance as they showed unique advantages and promising applications in flexible and organic composite transistors.

We fabricated the pillar-shaped and very interesting patterns of three-dimensionally aligned CNTs formed by pyrolysis of iron(II) phthalocyanine [49], and utilized them as the channel for transistors with mobility comparable to highly doped silicon as high as 61.6 $cm^2/V\,s$ [50] . We further doped CNTs with nitrogen [51] and fabricated field-effect transistors based on an individual CN_x/C nanotube by focus ion-beam technology, with electron mobility as high as 3.84×10^3 $cm^2/V\,s$ [52]. Temperature dependence of two-terminal transport experiments suggests that transportation was dominated by thermionic emission and tunneling through a 0.2 eV Schottky contact barrier [53].

We have prepared multi-walled carbon nanotubes (MWCNTs) coated with crystalline multi-walled rare-earth oxide [54], and zinc sulfide [55] achieving structures possibly have applications in catalyses, high density storage and functional devices. Recently, we successfully coated the MWCNTs by alumina as insulator for the CNT-based transistors [56]. We also functionalized single-walled CNTs with thiol groups to link them between predefined gold contact pads by a solution-based orientational self-assembly method [57].



M=Tb, Lu R=C_8H_{17} M=Eu^{3+}, Ho^{3+}, Lu^{3+}

10 **11**

Fig. 4 Layered rear-earth complex
(**10**) heteroleptic bis(phthalocyaninato) [47] and (**11**) amphiphilic tris(phthalocyaninato) [48]

5 Potentials and applications of OFETs

OFETs based on small molecules and polymer semiconductors will be extensively applied in electronic papers, RFID tags, OFET driving OLEDs [58]. In additional, investigations on trace matters and gas sensors using OFETs have been active for a long time since 1980s. Recently, OFETs were also used as mechanical sensors [59]; not long ago pentacene-based OFET arrays were modeled as human skins [60]. Other applications of OFETs such as the light [61] and photocurrent detectors [62] were realized. Prodigious progresses have been made in this field; however, several problems are still not solved. One is the development of OFETs with low operation voltage and high mobility, especially stable n type OFETs. Another challenge is the simplification of the device fabrication. There are also lots of unsolved theoretical issues on organic semiconductor charge carrier transportations. The model based on the inorganic MOS model is too tentative to explain OFET experimental results and should be further developed. Finally, single-molecule and single-electron organic devices should be further studied by intercrossing the organic material science and nano electronics. As investigations and technologies develop, these issues will be understood more clearly; thus OFETs will play significantly important roles in the new generation of devices.

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