

Analysis of Influencing Factors on Air-Stable Organic Field-Effect Transistors (OFETs)

Yong PAN¹, Aifeng LV¹, Lifeng CHI^{2*}

¹Physical Institute and Center for Nanotechnology (CeNTech), University Münster, Wilhelm-Klemm-Str. 10, Münster 48149, Germany

²Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Functional Nano and Soft Materials Laboratory (FUNSOM), Soochol University, Renai Rd. 199, Suzhou 215123, China

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The primary emphasis in this paper is on the major developments in the field of air-stable organic field-effect transistors (OFETs) over the past 20 years. The studies about the factors influencing the stability of OFETs, including air, humidity, oxygen and temperature, are reviewed and analyzed. The possible mechanisms that result in the degradation of OFETs, such as the penetrating of H₂O molecules, the doping effect of oxygen or the crystalline structure difference caused by temperature, are summarized. At same time, the reason why the field-effect mobility and the on/off current ratio of the transistor might change greatly with different ambient is concluded. The overall lives of OFET-based sensor in the detection of hazardous gases including nitrogen dioxide and ammonia are discussed, several breakthrough findings and technologies about how to solve the problem of instability of OFETs are also presented.

Keywords: OFETs, stability, sensor, air, humidity, oxygen, temperature.

1. INTRODUCTION

During the past many years, the performance of organic field-effect transistors (OFETs) has been continually improved and become more and more possible in industrial applications [1–3]. As a new developing sensing platform, OFET-based sensors, due to their high sensitivity, low production costs, room temperature detection, have also attracted intriguing attention owing to their advantages of plenty organic material resources, mechanical flexibility, and microarray compatibility, but the problem of instability of OFETs is still the focus of researchers, especially the interface property of dielectrics. The influencing of oxygen, air, moisture, is the most important factors to influence the OFETs. Although these mechanisms are not confirmed, some people have involved in developing a high performance OFETs for many years and have made great efforts to explain the mechanisms [4–6]. Today, the OFET devices are currently being driven by two methods, the optimization of device structures and the development of new organic semiconductors, while the major challenge in OFETs research is the development of practical OFET materials that ensure not only high performance but also high stability in the operational environment, so the mechanism study influencing the stability of OFETs is surely urgent.

2. AIR-STABLE ORGANIC FIELD-EFFECT TRANSISTORS

The environmental instability of OFETs has been attributed to chemical impurities, molecular structure, ionization potential, broken conjugation, water, oxygen,

and light exposure. With the development of conductive polymers whose highly conductive characteristic can be used as electrodes in OFETs, a lot of all-polymers OFETs have been studied. More than ten years ago, Henning et al. had reported on the fabrication of all-polymer OFET comprising conductive poly(ethylenedioxythiophene) poly(styrene sulfonic acid) systems (PEDOT-PSS), as electrode material, poly(3-hexylthiophene)I(P3HT) as semiconductor, as well as polymer substrates and dielectrics. The OFET was measured after 30 days storage under ambient oxygen and humidity conditions, it could be found that both the maximum on-current and the off-current rose greatly, but the on/off ratio thereby decreasing from 97 to 81, they thought the good performance of devices was probably related with the top-gate setup and the purity of the materials chosen [7]. On the other hand, as most of organic semiconductors exhibited only single carrier (electron or hole) operation mode due to intrinsic impurities level, so the ambipolar characteristic had shown its potential as alternatives for the research of OFETs which could be operated either p- or n-type in a single device, but if the measurement processes were carried out in air ambient, the deterioration of electrical characteristics might result in [8].

The stability of organic semiconducting polymers toward oxidative doping is related to their ionization potentials, i.e., the highest occupied molecular orbital (HOMO) levels. Amanda et al. explored a method for increasing the stability of polythiophenes toward oxidative doping by lowering the HOMO energy level through the addition of electronwithdrawing alkyl carboxylate substituents [9], the regioregular ester-functionalized polythiophenes were found to have charge mobilities to 0.07cm²V⁻¹s⁻¹. Later, Tomo Sakanoue investigated the electrical characteristics of single-component ambipolar

* Corresponding author. Tel.: +49-251-8333651; fax: +49-251-8333602. E-mail address: chilf@suda.edu.cn (L. F. Chi)

OFETs by controlling the device structure and the measurement conditions, he found the hole mobility was barely affected by the exposure while the electron mobility was significantly affected [10].

To get the high μ_{FET} of thin-film-based OFETs, which is the most important parameter in using, Kazuo et al. had ever synthesized DPh-BTBT, 2,7-diphenyl[1]benzothieno[3,2-b][1]enzothiophene [11], this high-quality homogeneous thin films were easily obtained by vacuum deposition on the bare Si/SiO₂ substrates of top-contact-type OFET. Treated with octyltrichlorosilane (OTS) and fabricated at 100 °C, the devices with $\mu_{\text{FET}} = 2.0 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and $I_{\text{on}}/I_{\text{off}} \sim 10^8$ were reproducibly obtained, and the mobility showed a slight decrease after 250 days storage. Considering from the chemical structure, it was obvious that when the number of benzene rings increased, the HOMO level became higher and the HOMO–LUMO (Lowest Unoccupied Molecular Orbital) gap became narrower. Such high-lying HOMO level made the molecule susceptible to air-oxidation in ambient conditions, and narrow HOMO–LUMO gap would cause readily photo-induced excitation that might lead subsequent chemical reactions [12, 13]. Yutaka et al. also designed and synthesized carbonyl-bridged bithiazole as a new electronegative unit for air-stable n-type OFET, which contributed both to lowering the lowest unoccupied molecular orbital energy level and to stabilizing the anionic species, the electron mobilities and the on/off ratios were $0.06 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and 10^6 respectively [14]. Electrochemical measurements and X-ray analysis of C-BTz revealed that the combination of carbonyl-bridging and electron-accepting thiazole rings contributes to a low LUMO energy level for charge-carrier transport. After 1 year storage under ambient condition, the results indicated that physically adsorbed oxygen and/or water did degrade the device performance, while the C-BTz molecule did have an intrinsic air stability when it was used as an active material for n-channel OFETs.

P3HT has been widely studied for its wide commercial availability and high carrier mobility, but it is not easy to measure P3HT-based OFET in ambient conditions because of its rapid undesirable interactions with atmospheric oxygen and moisture. If the P3HT-based OFET was

prepared with chloroform and hexamethyldisilazane (HMDS), it took only about 10 min in air for the off current to increase by one order of magnitude. While if it was prepared by 1,2,4-trichlorobenzene (TCB) and OTS, only a slight degradation was caused after 120 min exposure to air [15]. In all the factors concerning air stability of P3HT, crystallization was very crucial because a tight and ordered packing of P3HT molecules would provide effective resistance to atmospheric oxygen and water molecules penetrating the film and doping the P3HT at the semiconductor-insulator interface. In addition, many solvents may lead to different molecular packing and interface dipoles, which might also affect the HOMO and influence the stability of OFETs [16]. In 2010, Yanmin Sun et al. studied four different interfaces [17]: bare SiO₂, SiO₂ modified with OTS (SiO₂-OTS), SiO₂ coated a thin PMMA layer (SiO₂-PMMA), and SiO₂ substrate coated with PMMA and subsequently treated with OTS (SiO₂-PMMA-OTS), very different air stability was observed by modification of the interface, and it was believed that the improved stability was a consequence of H-bonding interactions the acidic OH groups on the silica surface and the basic carbonyl groups on PMMA.

As most of n-channel OFETs could not be operated well in air, but for application in the future, n-type organic semiconductors are highly desired. Yoshinobu et al. fabricated a n-channel OFET with a film of N,N'-bis(4-trifluoromethylbenzyl)perylene-3,4,9,10-tetracarboxylic diimide (PTCDI-TFB) which had two electron-withdrawing trifluoromethylbenzyl groups and was expected to have a high electron affinity [18]. By calculating, the LUMO level was to be 4.8eV and the electron affinity of PTCDI-TFB was also estimated to be 4.8eV. Another approach to improve the stability of n-channel OFETs was the use of organic compounds possessing strong electron-withdrawing units as the semiconducting materials, such as Fang-Chung Chen et al. improved the air stability of n-channel OTFTs based on N,N'-dioctyl-3,4,9,10-perylene tetracarboxylic diimide (PTCDI-C8) by modifying the device dielectric surface using hydroxyl-free polymer insulators [19], Fig. 1.

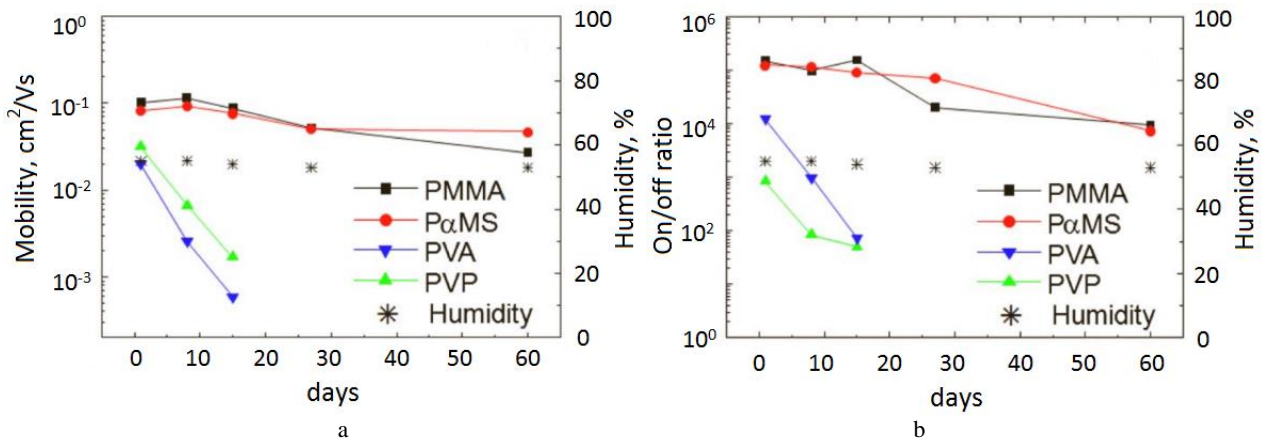


Fig. 1. Air-stability measurements of PTCDI-C8 OTFTs: a—electron mobilities and relative humidity as a function of time; b—on/off ratios plotted as a function of time. Between each measurement, the devices were stored in a cabinet, in which the relative humidity was also controlled at 50 %–55 %

An example was that the poly(methyl methacrylate)(PMMA) and poly- α -methylstyrene (P α MS) can improve the air stability of n-channel OTFTs because the complete absence of hydroxyl groups (electron traps) on the P α MS and PMMA surfaces led to enhanced n-channel conduction. Under ambient conditions, the electron mobility of the device incorporating PMMA as the modifying layer remained almost unchanged for the first 15 days and dropped to $0.03 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ after 60 days, he thought this method was general for n-channel OTFTs and could be used for other n-channel semiconductors.

To further illustrate the air stability of n-Channel OFET, more mechanisms were proposed and discussed. In general, thermodynamic stability of the charge carrier against ambient oxidants (e.g., O_2 and H_2O) and the kinetic barrier provided by densely packed fluoroalkyl side chains ($-\text{R}^{\text{F}}$) to keep O_2 and moisture out of the active conducting layer are two much discussed rationales to account for the air stability of n-channel OFET, while a thermodynamically stable charge carrier (radical anion) is especially important for n-channel OFET. Experimentally, the first reduction potential (E_{R1}) of an n-channel organic semiconductor (OSC) molecule in solution was used to estimate its reduction propensity and to correlate with the ambient thermodynamic stability of a charge carrier (which actually exists in the solid state). Several threshold values of E_{R1} and E_{LUMO} deduced from electrochemical measurements for air stability have been reported [20–22].

The charge carrier should be stable enough against ambient oxidants such as O_2 and H_2O , and it has been suggested that OSCs with large enough electron affinity (E_{A}) will possess air-stable charge carriers. Yu-Chang Chang et al. studied 47 existing n-channel OSCs with different molecular core structures and device configurations [23]. By analyzing datum, a correlation between calculated adiabatic E_{A} and air stability was established at the level of theory, the minimum value of E_{A} to achieve air stability against ambient oxidants is about 2.8eV. For example, the E_{A} of perylenetetracarboxylic diimides (PDI) was found to be close to the threshold value, therefore, it was not surprising that many PDI derivatives were air-stable. E_{A} of the eight compounds demonstrating long-term stability were in the range of 2.8–3.7eV at the above theory level. Although the electron mobility might decrease to about 60 % of its original value, some n-type OFET which could be easily solution-processed are fabricated with conventional techniques, and the on/off ratios remained almost unchanged after 30 days storage [24, 25]. Heterotriangulene polymer (PTA) had ever been selected as an alternative organic semiconductor for air stable OFET, as the crucial air stability of PTA was provided by the low lying HOMO level and wide bandgap, which exhibiting an enhanced hole transporting ability in organic light emitting diodes (OLEDs) together with an improved thermal and morphological stability [26, 27]. Kerstin Schmoltner et al. reported an air-stable p-type PTA for large-area OFET applications, the synthesized amorphous organic semiconductor was characterized concerning morphological, optical, electrical and interface related properties and revealed a saturation mobility of about $4.2 \times 10^{-3} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and an on/off current ratio of

$\sim 10^5$ in bottom-gate/bottom-contact (BG/BC) OFETs. After storing over 3 months, PTA OFETs did not show any significant changes in device performance, and the excellent stability was proved [28].

High HOMO energy level always leads to poor air stability, therefore, development of OFET materials with a low HOMO level and highly crystalline structure are need to overcome problem. In comparison with linear oligothiophenes, dendritic and hyperbranched oligothiophenes exhibited the relatively low-lying HOMO energy levels and could be used to improve air stability [29]. The best mobility of branched oligothiophenes was $0.012 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, it might provide extended delocalized π -electrons for enhancing charge transport [30]. Considering about this, Hung-Chin Wu et al. synthesized air stable branched octithiophene oligomer(8T) for OFET application [31], the 8T based OFETs showed the field effect mobility up to $2.12 \times 10^{-2} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and a high on/off ratio of 5.1×10^6 , the relatively low-lying HOMO energy level (-5.53eV) of 8T led to the outstanding air stability of the OFET device, it was attributed to the fact that the octadecyltrichlorosilane (ODTS)-treated surface possessed a hydrophobic long alkyl chain, preventing the influence of oxygen and moisture [32].

Rizwan Ahmed et al. once studied the photoinduced charge transfer between C60 semiconductor layer and parylene dielectric [33]. He used the bottom-gate and top-gate based C60 OFETs to compare the effects induced by light, it indicated clearly that the nature of the interface between the active layer and the dielectric played a major role in the light-induced threshold voltage shift, and it also proved that the light-induced threshold voltage shift strongly depended on the wavelength and intensity of the incident light as well as on the transverse electric field at the interface.

3. THE FACTORS INFLUENCING OFETs

3.1. Influence of humidity on OFET stability

In fact, most of sensors are affected easily by humidity and some can even be used as humidity sensor for their sensitivity to moisture [34, 35]. Humidity can affect the charge transport properties of OFET in many different ways. Such as that H_2O molecules can easily diffuse into these crevices and interact with the trapped carriers by altering the electric field at the grain boundaries, or the H_2O molecules might be incorporated into the films and change their morphology, all these factors will affect the hole mobility and translate into changes in the saturation current, then devices show decreased current output and mobility as the relative humidity (RH) is increased.

Degradation of organic thin-film field-effect transistors (OTFTs) with pentacene as the active material was studied by Yong Qiu et al., it was found that the field-effect mobility of the device decreased by 30% and the on/off current ratio decreased to one fifth after the OTFTs had been stored in atmosphere for 500 h. AFM and IR spectrum analysis revealed that the degradation was caused by H_2O molecules adsorbed on the pentacene film [36]. Atmospheric water might have a greater effect on the susceptibility of the device operating in air than O_2 . To

prove this, OFET based on P3HT was prepared by Satoshi et al. to determine the influence of moisture on device characteristics [37]. The device was operated and measured under different conditions, by comparing the experiment results, an increase in the off-state conduction and a deterioration in the saturation behavior of the output I_d could be observed. It might be the adsorption of water molecules on the surface possibly increased the charge carrier density because of their relatively large dipole momentum.

In 2004, Bäcclund et al. investigated the hygroscopic insulator field-effect transistor (HIFET) fabricated from solution processable polymers. In his device, the current modulation was improved in the presence of moisture, the static performance of the device characterized by current level and modulation at low voltages (less than 1V) was greatly enhanced by moisture [38]. Later, they further clarified the influence of moisture and the effect of different solvents on the performance of the device by using a PEDOT:PSS(poly(3,4-ethylenedioxythiophene) doped with poly(styrene sulfonic acid)) gate electrode in different RH atmospheres [39]. The I-V characteristics were quite similar in RH around 40 %–60 %, but changed significantly in lower humidity levels (RH 20 %), it might be the reason that the amount of the charge moving in the device increased with increasing humidity and the ions moved faster at higher humidity levels.

The device geometry (bottom or top contact device) and channel length are the other factors to influence the OTFT sensitivity by moisture. Dawen Li et al. selected a broader class of p-channel organic semiconductor materials including oligofluorene derivatives (C12FTTF and C6TTF) as well as pentacene as active layers for OTFTs, and two types of structures, i.e., top and bottom contact, with relatively large (200 microns) or small (25 microns) gaps between the source and drain electrodes were studied [40]. They found that OTFTs with small channel lengths and bottom contact structure had the highest sensitivity to RH by investigating the current response of an OTFT to humidity with different p-channel organic semiconductors and various device structures. They thought the reason was that the degradation of OTFT saturation current in a humid environment was expected to

be strongly dependent on the density of grain boundaries between source and drain electrodes [41].

Organic semiconductors that can be fabricated by simple processing techniques and possess excellent electrical performance are very important in the preparing of OFET. The stability of the electrical performance of OFETs based on thieno[3,2-b]-thiophene polymer with an ionization potential of 5.10eV was discussed by McCulloch et al. [42]. After 5 days storage in ambient air with humidity (~ 50 %), the field-effect mobility decreased by a factor of two, and the on/off ratio was reduced by two orders of magnitude. In comparison, the electrical performance of the device upon exposure to air with low humidity (~ 4 %) showed the stability for more than 20 days, Fig. 2.

Fewer n-type materials used in OFET had been reported, moreover most of the reported n-type organic semiconducting materials showed instability and sensitivity to air exposure. Junhyuk Jang et al. selected CYTOPTM fluoropolymer as a dielectric insulator to reduce the effect of interface trap sites and fabricate ambient air-stable C60 OFETs [43]. His research indicated that the interface trap density for electrons for n-type organic materials was significantly reduced by using the perfluorinated polymer as the gate dielectric layer even in ambient air. To clarify the role of the water in combination with the polymeric semiconductor, electrical and optical techniques were used carefully by Nikolai Kaihovirta et al. to monitor the electrolyte/semiconductor interface in an ion conducting membrane based OFET. With the using of a hydrous ion conducting membrane (PTFE:PFSA), they found that the careful balancing of the hydration-level in the membrane was necessary in order to prevent excess gate leakage currents and a deterioration of the transistor characteristics, that meant a dry membrane would not induce charges to the active channel [44]. The effects of moisture on the electrical characteristics of pentacene field-effect transistors (FETs) with the polyvinylpyrrolidone (PVPy) gate insulator was also reported by Jaehoon et al., it was found that the device performance rapidly degraded following exposure to ambient moisture [45].

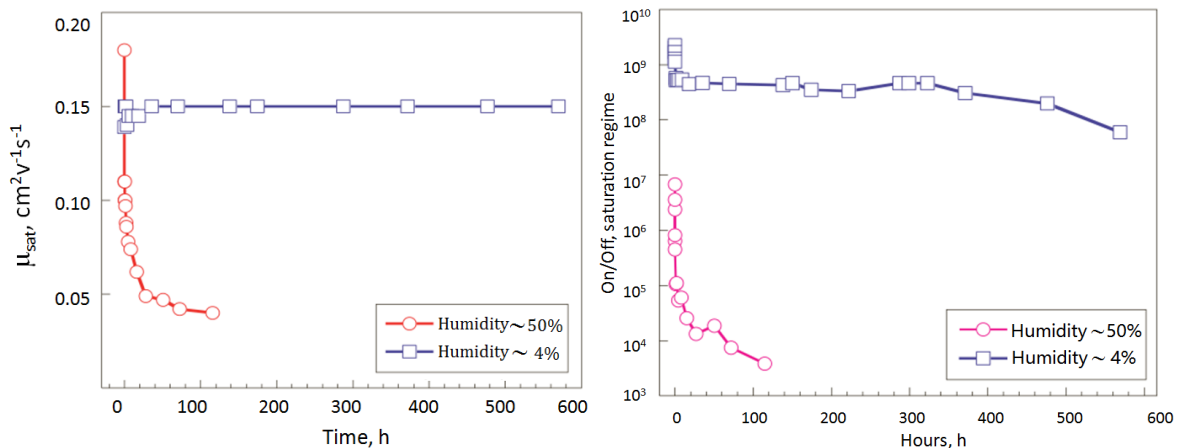


Fig. 2. Stability of FET devices. Transfer characteristics for polymer thieno[3,2-b]thiophene shown on prolonged exposure to change in mobility and ON/OFF ratio with time in both ambient and low-humidity air

Se Hyun Kim et al. employed a 150nm SiO_x film as an encapsulation layer to protect pentacene OFETs from H₂O, chemical and thermal damage [46]. It could be found that the exposure of the SiO_x-capped device to ambient air could cause the decreases in μ_{FET} , the on-current and positive shift of the turn-on voltage, it implied that H₂O molecules penetrating through the SiO_x layer into the pentacene film and/or the pentacene/dielectric interface resulted in the degradation of device performances. Bovine serum albumin (BSA) was ever chosen as the gate dielectric material to fabricate OFETs and to illustrate its ability to increase the drain current at low voltage by Chun-Yi Lee et al. [47]. In a relative humidity of 47 %, the $\mu_{\text{FET,sat}}$ value increases from 0.3 cm² V⁻¹s⁻¹ to 4.7 cm² V⁻¹s⁻¹ and the V_{TH} reduces from -16 V to -0.7 V. The susceptibility to degradation in both ambient and aqueous environments could also prevented organic electronics from sensor using. To overcome this barrier, solution-processed OFETs which were based on a hygroscopic biomaterial with albumin as the gate dielectric and poly(3-hexylthiophene) as the semiconductor were fabricated by Mingchao Ma et al. [48]. It was proved that the device performance could be improved significantly with the increasing of RH, and the results suggest moisture were potentially important in applications of biosensors.

3.2. Influence of oxygen on OFET stability

Molecular oxygen is a kind of strong electron acceptor leading to p-type conductivity of most organic semiconductors, so the charged p-type organic semiconductors are easily attacked in a reaction which can reduce water. Such as P3HT, a reversible decrease in the on/off-ratio of OFETs might be produced because of the enhanced bulk conductivity in the active P3HT layers [49]. In previous studies, the reversible and irreversible oxygen effects on polythiophenes (PT) was explained by the reversible formation of a charge-transfer (CT)-complex between molecular oxygen and the PT molecules [50], the existence of this metastable complex had been shown by its UV/Vis absorption band at the low energy edge of the P3HT absorption, but the information was not enough to completely understand the oxygen-induced effects in PT.

The methods to solve the problem of air stability of OFET were concluded as follow. First, kinetic barriers might limit the diffusion of air contamination to the channel region [51]. Second, thermodynamic control of the solubility of oxygen in the thin film was possible to tune the redox potentials of the organic semiconductors itself [52]. The third method was the device encapsulation by dielectric layers, which could also prevent the diffusion of oxygen and water vapors to the channel region of OFETs [53, 54]. In 2004, L. LÜer et al. studied the oxygen-induced fluorescence quenching(FQ) of excited states of regioregular P3HT in thin films to get some information about the mechanisms [55]. He thought the kinetics of FQ consisted of a fast component which was fully reversible, and a slow component which was partially irreversible, it was ascribed to collisional quenching of excited singlet states after oxygen diffusion into the bulk of the film and form reversibly charged states within 1 ps.

Toshio Nishi et al. investigated the interfacial

electronic structure for titanyl phthalocyanine (TiOPc) prepared under ultrahigh vacuum (UHV) and O₂ atmosphere by ultraviolet photoelectron spectroscopy (UPS) [56], the results indicated that the p-type semiconducting behavior of the TiOPc film deposited in O₂, was consistent with the supposed p-type doping by O₂ working as an electron acceptor. It was the first direct evidence of the doping effect by O₂ based on the interfacial electronic structure with quantitative information about the relative position of Fermi level (E_{F}) and the HOMO level, a clear conversion from n-type to p-type behavior was observed when the film was exposed to oxygen [57], X-ray photoelectron spectroscopy (XPS) of TiOPc film showed clearly the presence of extra oxygen forms for films deposited in moderate (10-6Torr) vacuums [58]. Two years later, the same working group of Toshio Nishi et al. studied the oxygen effect on the electronic structure of the interface between C₆₀ and highly-oriented pyrolytic graphite by UPS, they noticed the removal of band bending near the interface, this change of the electronic structure and the deduced possible origins were consistent with the drastic decrease of the electrical conductivity of the C₆₀ film fabricated in UHV at the exposure to O₂, so the influence of the atmosphere on the charge transport and the device characteristic of C₆₀ film was fatal [59].

The instability (oxidation sensitivity) of the compounds often lies in the low ionization potential, that is, a high-lying HOMO energy level. Within a particular class of compounds, an increased effective conjugation length leads to energy-rich HOMO levels and thus to an increased susceptibility towards oxidation [60]. Knipp et al. reported that the mobility of pentacene-based OFETs was not affected by exposure to dry O₂ and decreased upon exposure to air with moisture. Yan et al. had also shown that some components of air including N₂/O₂ and N₂/H₂O had no effect on the electric characteristics of OFETs based on copper phthalocyanine (CuPc) with a HOMO energy level of -5.1eV, these results indicated that wet oxygen was one of the factors causing the degradation of the performance of OFETs based on p-type organic semiconductors with high HOMO energy levels [61, 62].

The stability of OFETs based on two oligo-p-phenylenevinylenes with different HOMO energy levels was studied by Tomoyuki. By experiments, the OFET with a HOMO energy level of -5.55eV showed long-term air stability for a period exceeding one year, while the field-effect mobilities of organic semiconductors with a HOMO energy level ranging from -4.94 to -5.20eV decreased with time, it meant that the organic semiconductors with a relatively low HOMO energy level had good chemical stability [63]. Daisuke Kumaki et al. paid attention on the influence of O₂ and H₂O on the transistor characteristics in the p- and n-type OTFTs fabricated on the SiO₂ gate insulator without surface treatment of silane-coupling agent [64]. In both p- and n-type OTFTs, The V_{th} shifts to the positive direction indicated that negative charges were generated on the SiO₂ gate-insulator surface by exposure to ambient air and dry air, the possibility that the electrochemical reaction at the interface was related to the deprotonation of SiOH with O₂.

Bo-Chieh Huang et al. developed a simple technique

(oxygen plasma treatment) to find the charge trapping measurement for OTFTs. They prepared the pentacene-based OTFTs with and without oxygen plasma treatment in order to display the relationship between charge trapping and transfer characteristics of OTFTs [65]. In their works, the μ of OTFTs with oxygen plasma treatment was much higher than that of OTFTs without oxygen plasma treatment, and positive V_{th} shift was observed. In n-type OFETs, anions could form and had high reduction powers, so it could reduce oxygen molecules diffused into the layer of organic molecules. [66].

3.3. Influence of temperature on OFET stability

The influence of temperature is another important factor to the stability of OFETs. In 2000, Gilles Horowitz et al. performed current–voltage measurement on polycrystalline sexithiophene (6T) thin film transistors at temperatures ranging from 10 to 300 K [67]. They found that carrier mobility would increase quasilinearly with gate voltage at room temperature. Different substrate temperatures during the deposition process could influence the ordering and orientation of the molecules. While K.Xiao et al. observed high order and strong orientation of CuPc molecules on Si/SiO₂ by using XRD and TEM analysis at different substrate temperatures for a gate voltage $V_G = -50$ V during the deposition process, which affected directly the morphology of thin films [68]. The channel conductivity was clearly thermally activated with a thermal activation energy, it was because of the defects, disorder and domain boundaries in the polycrystalline film. When the substrate temperature for deposition of CuPc reached to 120 °C, a mobility of $3.75 \times 10^{-3} \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ could be gotten.

It has been recognized that ordering of the organic film structure is very important for achieving high field-effect mobilities. The dependence $\mu(T)$ in pentacene thin film OFETs varies from thermally activated to temperature independent with $\mu(300\text{K})$ increasing from $0.3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ to $1.2 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ [69]. Podzorov et al. used parylene as a material as the gate insulator and fabricated the FETs at the surface of single crystals of rubrene. It demonstrated the hole-type conductivity with the field-effect mobility up to $1.0 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and the on/off ratio up to 10^4 at room

temperature. The temperature dependence of the mobility depended strongly on the value of $\mu(300\text{K})$ [70]. In 2004, Hamadani et al. reported his measurements of the parasitic contact resistance and the true channel resistance in bottom contact P3TH FET from room temperature down to 77 K [71], once parasitic contact resistances were taken into account, the mobility of solution-cast P3HT could approach $1.0 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ at room temperature, these results indicated that performance of P3HT-based OFETs could be limited more by contact physics than by the intrinsic transport physics in the polymer itself. In 2004, Jianfeng Yuan et al. investigated the effects of OFET which used F₁₆CuPc as active layer and tantalum pentoxide (Ta₂O₅) as insulator[72], he demonstrated that the threshold voltage shift (ΔV_T) was temperature dependent with activation energy of 0.51eV, and the activation energy of ΔV_T might come from temperature dependent of hopping conduction in the insulator, Fig. 3.

The crystalline structure and the morphology of the organic semiconductor thin film can also be heavily influenced by the temperature (TD) at which the film is deposited. Th.B. Singh et al. investigated C₆₀-based n-channel OFET with mobility in the range of $0.4 - 1 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. C₆₀ OFETs exhibited a temperature dependent mobility with an Arrhenius behavior, and the devices characterised in inert atmosphere conditions showed high stability with an on/off ratio $> 10^4$ [73]. Two years later, they extended an extensive study of morphology and crystallinity of the fullerene films using atomic force microscopy and grazing-incidence x-ray diffraction, by experiments, they not only found the correlation of crystalline quality of the C₆₀ film and charge carrier mobility, but proved that a higher substrate temperature led to single crystal-like faceted fullerene crystals [74]. To investigate the intrinsic transport mechanism of OFETs, Takeo Minari et al. developed a method of fabricating single-grain OFETs with top-contact structure by using finely formed metal mask. The results indicated the saturation mobility of $1.11 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ at 300 K increased with a decrease in temperature, reaching $1.22 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ at 210 K [75].

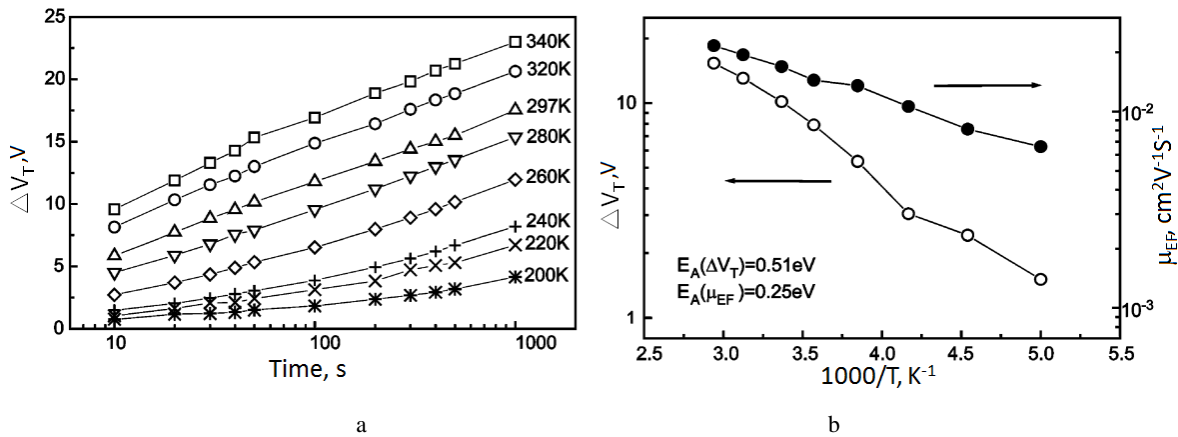


Fig. 3. a – time dependence of ΔV_T under various temperatures; b – temperature dependence of the threshold voltage shift ($V_{G,bias} = 50$ V, $t = 50$ s) and the field-effect mobility

In the aspect of mechanism, Lawrence Dunn et al. not only found that at higher temperatures $\mu_{\text{dynamic}} > \mu_{\text{FE}}$, at lower temperatures $\mu_{\text{dynamic}} < \mu_{\text{FE}}$, but also found $E_{a,\text{dynamic}}$ to be greater than $E_{a,\text{FE}}$ by approximately 7 meV at the same effective gate voltage [76].

It had been proven that the mobility of OFETs was determined by the layer-by-layer growth and the resultant crystallinity of the active layer, under a vacuum deposition condition, the structure of the organic semiconductor thin film was influenced by the growth conditions, e.g. substrate temperature, and so on. At a low temperature ($< 90^\circ\text{C}$), thin (10–20 nm) layers could easily be obtained by hot-wire chemical vapour deposited (HWCVD) or solution-processed technologies [77, 78]. By this technology, high-performance OFETs with high-k dielectric for low-voltage, high field-effect mobility (μ) and on/off ratio could be fabricated. During the procedure, the film of OFET deposited at different temperature could be analysed by ultraviolet-visible-near-infrared (UV-NIR) or X-ray diffraction patterns (XRD) [79]. In recently years, some people are focusing on biomaterial-based OFETs which can be used as temperature sensors because of the advantages including transparency, flexibility, thermal stability, high-temperature sensitivity and biocompatibility, and thus possess broad applicability for environmentally friendly electronics, implantable medical devices, etc [80].

4. STABILITY OF THE OFETs SENSORS

Many past and recent works successfully demonstrated the use of OFET in the field of gas sensing based on conjugated polymers, oligomers, or small molecules, such as the sensors used in humidity, ammonia, NO_x , formaldehyde, alcohol, explosive gas [81–84]. It is because, first, analyte molecules can diffuse through the semiconductor and approach the interface between the source and drain contacts, so the organic semiconductor can affect the carrier injection [85]. Secondly, the migration of analyte molecules into the organic semiconductor grain boundaries can deteriorate the carrier transport along the conduction channel of an OFET due to the introduction of localised charge traps [86], these interactions may have an impact on the conductance of the channel and the mobility of the carriers. Additionally, the

analyte molecules that diffuse deeper into the semiconductor film and migrate at the semiconductor-dielectric interface of an OFET can also affect the threshold voltage of the device, so the stability of OFET sensors is still a serious problem in using.

Although many kinds of OFET sensors have been fabricated to detect different gases, the overall lifetime of the device for environmental stability is seldom reported. Junsheng Yu et al. created a kind of ammonia sensors based on bottom contact OFETs using pentacene as an active layer and polymermethylmethacrylate (PMMA) as an insulator. The variation of drain-source current was tested after 10 and 30 days storage in air when OFET sensor was exposed to different NH_3 concentrations [87], Fig. 4.

In 2013, Liqiang Li et al. fabricated another ammonia gas sensor with ultrathin micro stripes of dialkyl tetrathiapentacene (DTBDT-C6) [81]. After storage in air for 25–35 days, the sensors still worked well and the sensitivity was still in the range of initial detection. These series of stability tests indicated the good reversibility and stability under continuous operation and storage conditions, which were rarely met in the OSCs-based sensors reported so far. The degrading of OFET sensor also happened on the detecting of dimethyl methylphosphonate (DMMP), after three months storing, although the trends of detection results to DMMP were consistent with the fresh preparing OFET sensors, the mobility value and the sensitivity would drop, and the response and recovery were rather slow [88].

In spite of the problems mentioned above, a lot of efforts are being made to optimized these OFET sensors. Minseong Yun et al. reported on a new sensor based on top-gate OFETs wherein chemical sensing was driven by the diffusion of ionic species. They demonstrated that top-gate OFETs based on 6,13-bis(triisopropylsilyl)ethynyl pentacene (TIPS-pentacene) using a bilayer CYTOP/ Al_2O_3 dielectric could display excellent operational and environmental stability and were suitable for the implementation of reusable chemical/biological sensors because they primarily responded to charged species diluted in an aqueous media by rapidly shifting their threshold voltage [89].

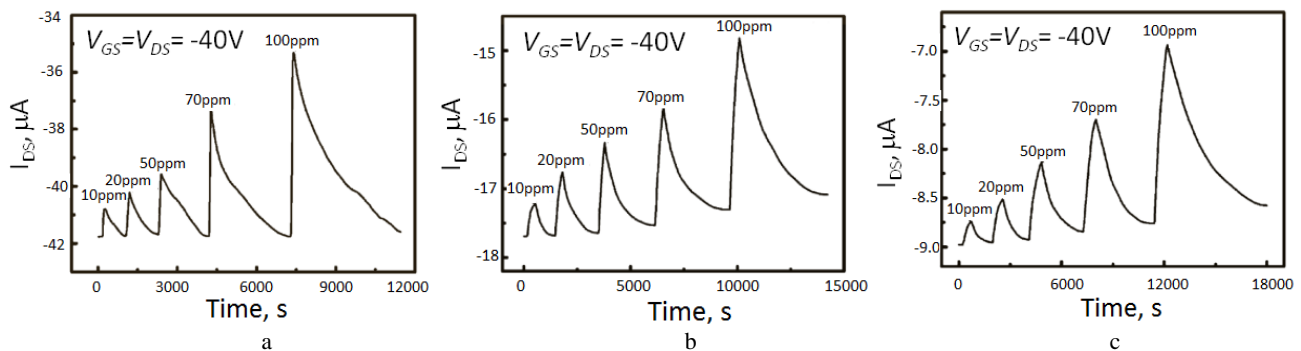


Fig. 4. Relationship between drain-source current and time when the OFET sensor exposed to various cycles of the exposure/evacuation different NH_3 concentrations ranging from 10 to 100 ppm, the OFET sensor: a–detected NH_3 after preparing; b–was stored in air for 10 days; c–was stored in air for 30 days

Shijiao Han et al. fabricated a kind of high-response OFET-based NO₂ sensor with the synergistic effect of zinc oxide/poly(methyl methacrylate) (ZnO/PMMA) hybrid dielectric and CuPc/Pentacene heterojunction, after being stored in atmosphere for 30 days, the variation of saturation current increased more than 10 times at 0.5 ppm NO₂, they thought the performance enhancement was due to the synergistic effect of the dielectric and organic semiconductor [90]. Although great achievement has been made, there are more and more experiments are needed to improve the stability of OFET sensors, the major findings of the studies in recent years have proved the fact that it is possible to solve the problems of stability of OFET sensor, of course, there is enormous research scope to extend the dimension of applicability in this field.

4. CONCLUSIONS

The present review attempts to give an overview of influencing factors of OFETs stored or operated in environment, and many research works related with stability of OFET are summarized. From the discussion above, we find that almost all of the OFETs or OFET-based sensors are easily degraded by moisture, oxygen, temperature or other factors existed in ambient, such as the penetrating of H₂O molecules, the doping effect of oxygen or the crystalline structure difference caused by temperature, field-effect mobility and the on/off current ratio of the transistor might decrease greatly because of the difference of ambient. Although there has been much development in designing and fabricating air-stable OFET to overcome this limitation during the past years, more studies are needed to understand device degradation mechanisms and improve the OFETs lifetimes. In conclusion, the stability of OFET is the most important in the application in the future, there is still enormous research scope to extend the dimension of applicability of OFETs.

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