

Interface Engineering: An Effective Approach toward High-Performance Organic Field-Effect Transistors

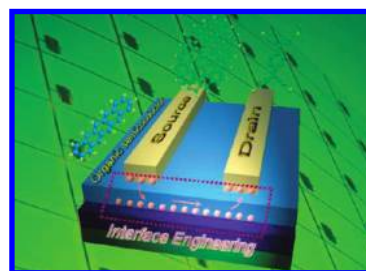
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CON SPECTUS

By virtue of their excellent solution processibility and flexibility, organic field-effect transistors (OFETs) are considered outstanding candidates for application in low-cost, flexible electronics. Not only does the performance of OFETs depend on the molecular properties of the organic semiconductors involved, but it is also dramatically affected by the nature of the interfaces present. Therefore, interface engineering, a novel approach towards high-performance OFETs, has attracted considerable attention. In this Account, we focus on recent advances in the study of OFET interfaces—including electrode/organic layer interfaces, dielectric/organic layer interfaces, and organic/organic layer interfaces—that have resulted in improved device performance, enhanced stability, and the realization of organic light-emitting transistors.



The electrode/organic layer interface, one of the most important interfaces in OFETs, usually determines the carrier injection characteristics. Focusing on OFETs with copper and silver electrodes, we describe effective modification approaches of the electrode/organic layer interfaces. Furthermore, the influence of electrode morphology on device performance is demonstrated. These results provide novel approaches towards high-performance, low-cost OFETs.

The dielectric/organic layer interface is a vital interface that dominates carrier transport; modification of this interface therefore offers a general way to improve carrier transport accordingly. The dielectric layer also affects the device stability of OFETs. For example, high-performance pentacene OFETs with excellent stability are obtained by the selection of a dielectric layer with an appropriate surface energy.

The organic/organic layer interface is a newly investigated topic in OFETs. Introduction of organic/organic layer interfaces, such as heterojunctions, can improve device performance and afford ambipolar OFETs. By designing laterally arranged heterojunctions made of organic field-effect materials and light-emitting materials, we realized both light emission and field effects simultaneously in a single OFET.

The preceding decade has seen great progress in OFETs. Interface engineering provides a simple but effective approach toward creating high-performance OFETs and will continue to make essential contributions in the development of useful OFET-based devices. The exploration of novel interface engineering applications also merits further attention; the design of gas sensors through a more complete understanding of interface phenomena serves as just one example.

Introduction

Organic field-effect transistors (OFETs) have received considerable attention in the past decade due to their potential applications in organic integrated circuits such as radio frequency identification (RFID) tags, smart cards, and organic active matrix displays.^{1,2} The field-effect mobility of

OFETs is now comparable to that of devices based on amorphous silicon. However, it is still far from satisfactory for real applications. The focus of recent attention has been devoted to improving device performance and stability, to reducing the power consumption and fabrication cost, to exploring new applications, and to developing

simple fabrication techniques. Overcoming these challenges relies on the development of novel organic semiconductors and optimization of devices. From the point of a device, the device performance is dominated by the properties of device components as well as the properties of their corresponding interfaces. In fact, it is well-accepted that interface modification is an excellent way to achieve high-performance OFETs, since it is an effective approach to improving mobility, to enhancing the device stability, to reducing the operating voltage, etc. Thus, interface engineering has become a general way to fabricate OFETs with excellent device characteristics.

A typical OFET is composed of a gate electrode, a gate dielectric layer, an organic semiconductor layer and source–drain electrodes. The device is operated as follows. When a gate voltage is pulsed, the carriers accumulate near the dielectric layer/organic layer interface and form a conductive channel. Carriers then inject from the source electrode into the organic layer and transport through the channel to the drain electrode. The two major processes, carrier injection and carrier transport, occur at the electrode/organic layer interface and the dielectric/organic layer interface, respectively. Therefore, the properties of these interfaces influence the device characteristics dramatically. Modification of the electrode/organic or dielectric/organic interfaces remains the most widely investigated approach to improving device performance. Other interfaces have been investigated more recently. It has been frequently reported that introducing an organic layer with an organic/organic layer interface leads to improved carrier transport and allows realization of ambipolar OFETs and light-emitting OFETs. Consequently, the organic/organic layer interface has become an important one in interface engineering. The organic layer/atmosphere interface also has an influence on device characteristics such as device stability or sensitivity to certain chemicals (Figure 1). However, we will not deal with such an interface in detail because it is very different from the other three interfaces mentioned above. In fact, the organic layer/atmosphere interface is a “surface” while the other three interfaces are “buried” ones. Despite great progress in the investigation of interfaces, a deeper understanding of interface phenomena and exploration of novel interface modification techniques are still required in order to improve the overall performance of OFETs. In this Account, we detail effective interface techniques (electrode/organic layer interfaces, dielectric/organic layer interfaces, and organic/organic layer interfaces) that lead to enhanced device performance and improved stability and also afford light-emitting OFETs.

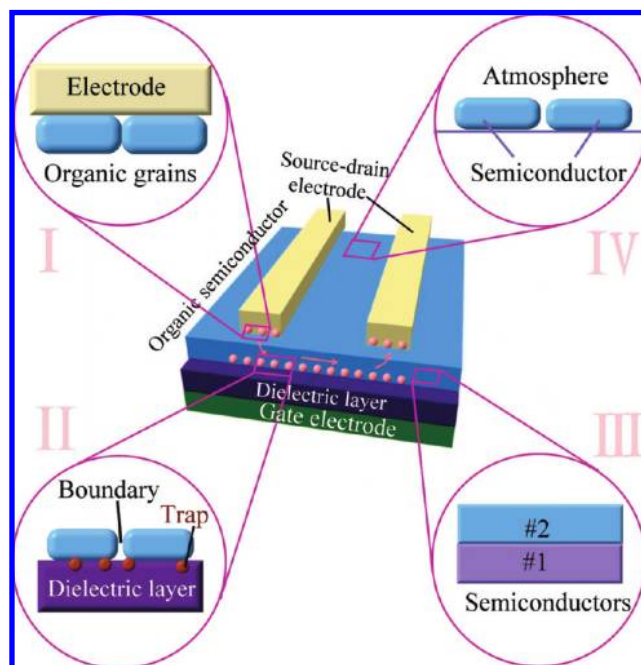


FIGURE 1. Interfaces in top-contact OFETs: (I) electrode/organic layer interface; (II) dielectric/organic layer interface; (III) organic/organic layer interface; (IV) organic layer/atmosphere interface.

Important Interfaces in OFETs

The electrode/organic layer interface has a key influence on carrier injection. In an ideal OFET, ohmic contact is assumed, such that the contact resistance is much lower than the channel resistance. However, Schottky contact is usually present in real OFETs and leads to large contact resistance. Therefore, the carrier injection barrier, which is determined by the work function of the electrodes and the energy level of the organic semiconductors, is an important parameter by which to evaluate the electrode/organic layer interface. For many OFETs, especially the n-type ones, reduction of carrier injection barrier is one of the most efficient ways to improve electrode/organic layer interface and device performance. Besides the energy barrier, the device structure also influences the contact resistance. The typical OFET device structure can be divided into top-gate bottom-contact (TGBC), top-gate top-contact (TGTC), bottom-gate bottom-contact (BGBC), and bottom-gate top-contact configurations (BGTC) (Figure 2). It has been well-accepted that BGTC and TGBC OFETs usually exhibit lower contact resistance than BGBC and TGTC ones. This is mainly due to the different carrier injection paths and injection areas. BGTC and TGBC devices possess large injection areas and favored injection paths compared with BGBC and TGTC devices (Figure 2), which leads to lower contact resistance. The contact condition of the electrode/organic interface is also dependent on the device geometry. For example, the source–drain electrodes are deposited before the depo-

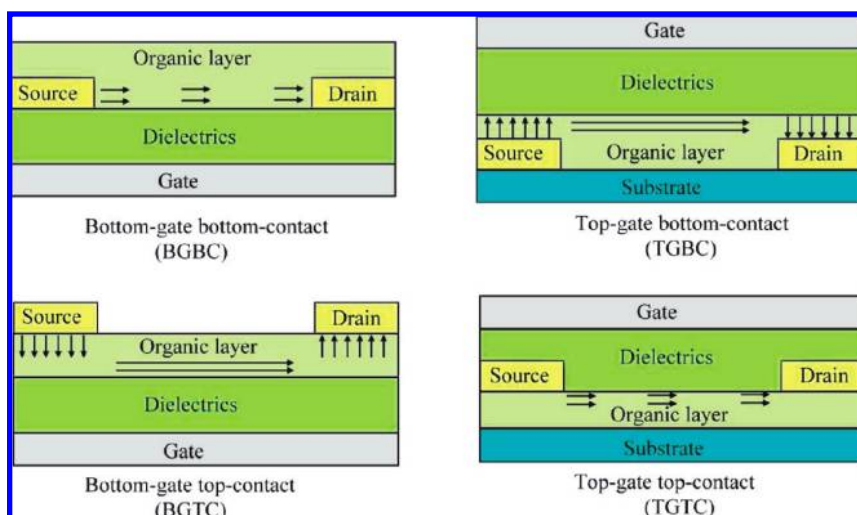


FIGURE 2. Carrier injection and transport scheme of four typical OFET geometries.

sition of the organic layers for BGBC OFETs. Unfortunately, the metal electrodes usually have a negative influence on the semiconductor deposition and bring on large boundary density at the electrodes/organic layer interface. As a result, the poor contact condition contributes in large part to the contact resistance. In fact, improvement of BGBC OFET contact conditions is an important subject since BGBC geometry is a feasible one for industrial applications. In a word, the energy barrier, carrier injection area, and contact condition are three major factors that determine the electrode/organic layer interface.

As mentioned above, carrier transport in OFETs occurs in the conductive channel, which is located on the few molecular layers near the dielectric layer.³ The dielectric/organic layer interface is thus the most important interface in OFETs. For OFETs with a given organic semiconductor, the dielectric/organic interface is affected by the surface energy, roughness, and trap density of the dielectric layer. The surface energy mainly influences the morphology of deposited films. If the dielectric layer possesses similar surface energy to that of the organic semiconductor, the films usually exhibit large grains and low boundary density, which favor effective carrier transport.⁴ The dielectric layer roughness also influences the morphology of the organic layer. High dielectric layer roughness usually results in small grain size and high boundary density.⁵ The trap density at the dielectric/organic interface is another influence on carrier transport. It is well-known that high trap density results in fewer mobile carriers and low device performance. Note that the electrons are more easily trapped than the holes. Therefore, controlling trap density is essential in the fabrication of high-performance OFETs especially for n-type and ambipolar devices. It has been reported that many devices based on p-type organic semiconductors exhibit n-type or ambi-

polar operation after modification of dielectric layers or selecting dielectric layers with low trap density.^{6,7}

Multilayer structures are frequently employed to obtain high performance in organic light-emitting diodes and organic solar cells.^{8,9} In traditional OFETs, the organic layer usually consists of a single semiconductor. Recently, OFETs with an organic layer made of two different semiconductors have been reported.^{10,11} Organic layers composed of a blended system (with two organic semiconductors blended into a single layer) and double layer structures are the two most common configurations. In both cases, an organic/organic layer interface is introduced. The interface is designed to improve the device performance or realize two types of functionality simultaneously. The double layer structure possesses a clear organic/organic layer interface. Since two different organic semiconductors are evaporated separately, the morphology of the second layer can be influenced by the initially evaporated layer, which can serve as a buffer layer. The double layer structure also plays an important role in attaining ambipolar operation and organic light-emitting transistors. It is mainly because the organic/organic layer interface favors the formation of two conductive channels in the OFETs and ensures the transport of two different carriers. Consequently, efficient transport of both holes and electrons can be obtained simultaneously in the conductive channel.

Electrode/Organic Layer Interface Engineering: An Excellent Way To Improve Carrier Injection and a Novel Way To Improve Carrier Transport

Improvement of Carrier Injection. To ensure efficient carrier injection, a suitable source–drain electrode has to be energetically compatible with the organic semiconductor. High

work function electrodes are thus required for most p-type OFETs. Gold has been the most widely applied source–drain electrode for OFETs to date. However, the high cost of gold is an adverse factor in practical applications. On the other hand, low-cost electrodes, such as Al, Cu, and Ag, are unsuitable for most p-type OFETs due to their relatively low work function. Therefore, electrode/organic layer interface engineering is necessary to ensure effective carrier injection and reduce device fabrication costs.

Introduction of a buffer layer between the source–drain electrode and organic layer is a common approach for modification of BGTC OFETs. The technique is frequently applied with the aim of both reducing the energy barrier and preventing metal atom penetration into organic layers. Taking pentacene OFETs as an example, an energy barrier of about 1 eV is present when Al serves as the source–drain electrodes. This large barrier results in low device performance of $2.8 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. When a very thin layer of MoO_3 is inserted as a buffer layer between Al and pentacene, the injection barrier is reduced dramatically,¹² resulting in an increased extracted mobility of $0.4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Metal atom penetration, a well-known phenomenon during electrode deposition for top-contact OFETs, can also be avoided by introducing a buffer layer. In pentacene top-contact devices, the gold atoms usually penetrate into a few pentacene layers and form clusters^{13,14} which have an adverse effect on hole injection. If a copper phthalocyanine (CuPc) layer (1–4 nm in thickness) is deposited before evaporation of the Au electrode, it can inhibit such penetration and result in improved device performance.¹⁵ In the above-mentioned buffer layer introduction processes, additional compound deposition is required. In fact, a buffer layer can also be formed during the electrode evaporation. We recently discovered that copper is an excellent source–drain electrode for many p-type OFETs.¹⁶ The device performances of many top-contact OFETs with copper electrodes are similar to those of Au-based devices, although this seems unreasonable from the point of energy barrier. Taking pentacene-based OFETs as an example, the hole injection barrier is about 0.5 eV when Cu is used as the electrode, while the value is 0.1 eV for the Au-based device. By using X-ray photoelectron spectroscopy, we examined pentacene films and observed an obvious O 1s peak under high vacuum conditions. This indicates that oxygen is present in the pentacene films even under high vacuum and suggests the formation of Cu_xO at the copper/pentacene interface during the copper deposition process. Since Cu_xO and pentacene possess matched energy levels, formation of the former during the deposition process gives a buffer layer that reduces the carrier injection

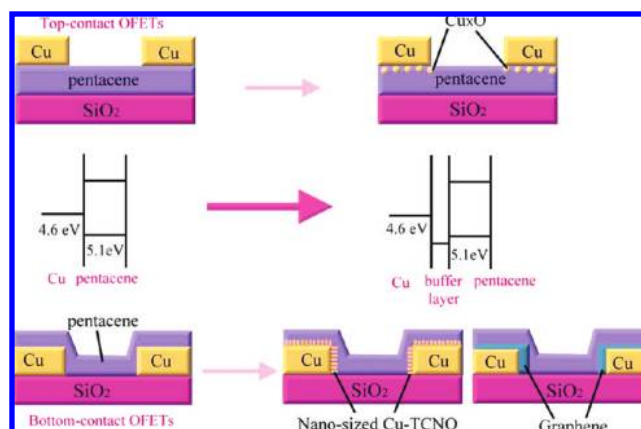


FIGURE 3. Illustration of Cu source–drain electrode modification techniques for both BGTC and BGBC pentacene OFETs.

barrier (Figure 3). More interestingly, the Cu_xO also can be formed during the storage of devices under ambient atmosphere. For example, the performance of CuPc and vanadyl phthalocyanine (VOPc) based devices with Cu top-contact electrodes increased dramatically once the devices were stored in air.

For BGBC OFETs, the grains around the source–drain electrodes are usually smaller than the ones in the channel because of the poor compatibility between metals and organic semiconductors. Therefore, enlarging the grain size near the electrode is an important goal for the engineering modifications of bottom electrodes. Self-assembled monolayer (SAM) modification is the most effective way to solve this problem.^{17,18} With modification of a gold electrode by thiol-based SAMs, the pentacene grains around the Au electrodes become much larger, which results in dramatically enhanced mobility in both linear and saturated regions.^{17,18} Modification of copper or silver electrodes is much more challenging because both enhancement of work function and improvement of compatibility with the organic semiconductor are required. We recently demonstrated an effective interface modification approach for Cu and Ag electrodes (Figure 4a,b).¹⁹ As expected, the output curves exhibit the typical S-shape when bare copper and silver work as the source–drain electrodes (Figure 4c), due to the contact resistance caused by large injection barrier and poor contact. Once the Cu and Ag source–drain electrodes are chemically modified with 7,7,8,8-tetracyanoquinodimethane (TCNQ), the formation of Cu–TCNQ and Ag–TCNQ enhances the work function of the electrodes to about 5.6 eV. The hole injection barrier is thus reduced dramatically (Figure 3). Furthermore, the gains on the modified electrodes are similar to those in the conductive channel due to the good compatibility between the modified electrodes and pentacene. Both reduced hole injection

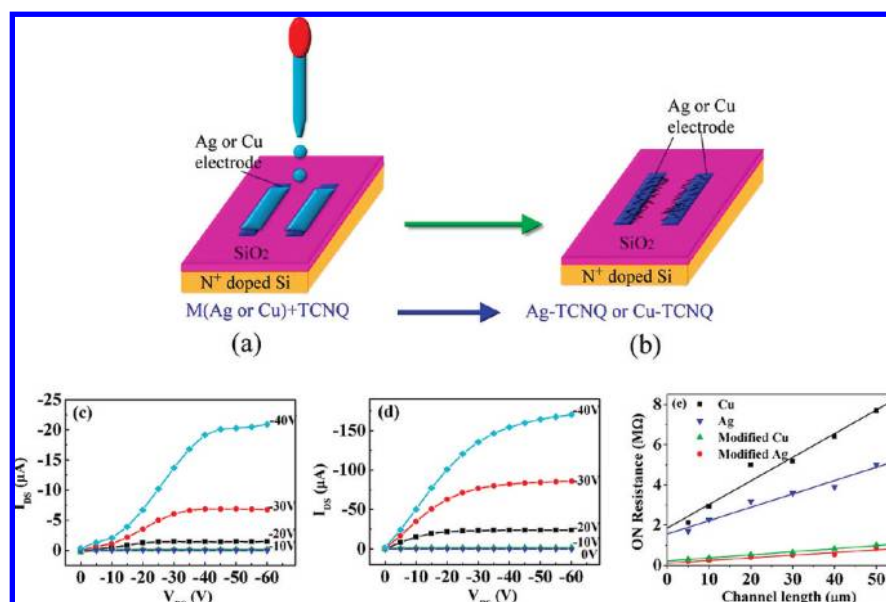


FIGURE 4. (a) Dropwise addition of TCNQ solution onto octadecyltrichlorosilane (OTS)-modified SiO₂ with Ag or Cu source–drain electrodes. (b) The modified source–drain electrode. Output characteristics of the devices with source–drain electrode of (c) Cu and (d) Cu–TCNQ modified Cu electrode. (e) Contact resistance of pentacene OFETs with different electrodes.¹⁹

barrier and improved electrodes/organic layer contact contribute to the reduction of contact resistances. The contact resistance of pentacene OFETs with Cu–TCNQ and Ag–TCNQ modified electrodes are 0.21 and 0.12 MΩ, respectively, which are about 1 order of magnitude lower than those of the devices with bare Cu and Ag electrodes (1.8 and 1.56 MΩ) (Figure 4e). Consequently, OFETs with modified electrodes exhibit excellent device characteristics (Figure 4d). We obtained high field-effect mobility for both p-type and n-type organic semiconductor based OFETs with modified electrodes. Taking pentacene OFETs with Ag–TCNQ modified electrodes as an example, the mobility increases from 0.02 to 0.18 cm² V⁻¹ s⁻¹. The value is comparable to the one for Au top-contact OFETs (0.15 cm² V⁻¹ s⁻¹).

Similar results were observed with graphene as the source–drain electrodes. Graphene, a 2-D graphite, possesses a similar structure to many organic semiconductors such as pentacene and tetracene (Figure 5). Therefore, strong π – π interactions between graphene and pentacene are expected. When the graphene serves as the bottom-contact electrode, the strong interaction between graphene and pentacene can result in the pentacene molecules lying flat on the electrode and forming a buffer surface for subsequent molecule packing (Figure 5).^{20,21} We observed that the sizes of pentacene grains on graphene are larger than those on SiO₂.²⁰ Even a single grain across the graphene/channel interface can be obtained. This leads to improved electrode/organic layer contact. Also, from the point of view of carrier injection barrier, the hole injection barrier reduced 0.3 and 0.7 eV after modifica-

tion of Cu and Ag electrodes, respectively, when it is assumed that graphene has similar work function of 4.8–4.9 eV (the work function of Cu and Ag are 4.6 and 4.2 eV). As a result, the improved contact and reduced carrier injection barrier result in low contact resistance. The pentacene OFETs with graphene-modified electrodes exhibit contact resistance of 0.16–0.18 MΩ, which is also much lower than the devices with bare Cu and Ag electrodes. The low contact resistance leads to dramatically improved mobility, higher than 0.5 cm² V⁻¹ s⁻¹. Surprisingly, we obtained almost identical saturated mobility for different devices with channel lengths range from 5 to 50 μm. The result further testified the small contact resistance since the mobility of the BGBC OFETs usually decreases with a reduction of channel length ($L < 50$ μm) due to the influence of contact resistance. Another interesting observation about graphene modification is the wide applicability for metal electrodes, such as Au, Ag, and Cu. More recently, we also fabricated pentacene OFETs with graphene-modified Au electrodes. By the control of modification conditions and pentacene deposition, field-effect mobility up to 1.2 cm² V⁻¹ s⁻¹ has been achieved even for devices with channel length of 5 μm (Figure 5d,e).²² The mobility is one of the best results for BGBC OFETs with narrow channel length.

As mentioned above, the injection area is also an important factor that influences contact resistance. Large contact area usually brings on low contact resistance. Consequently, introduction of source–drain electrodes with proper roughness is effective to reduce contact resistance for BGBC OFETs due to the following reasons. First, devices with a rough sur-

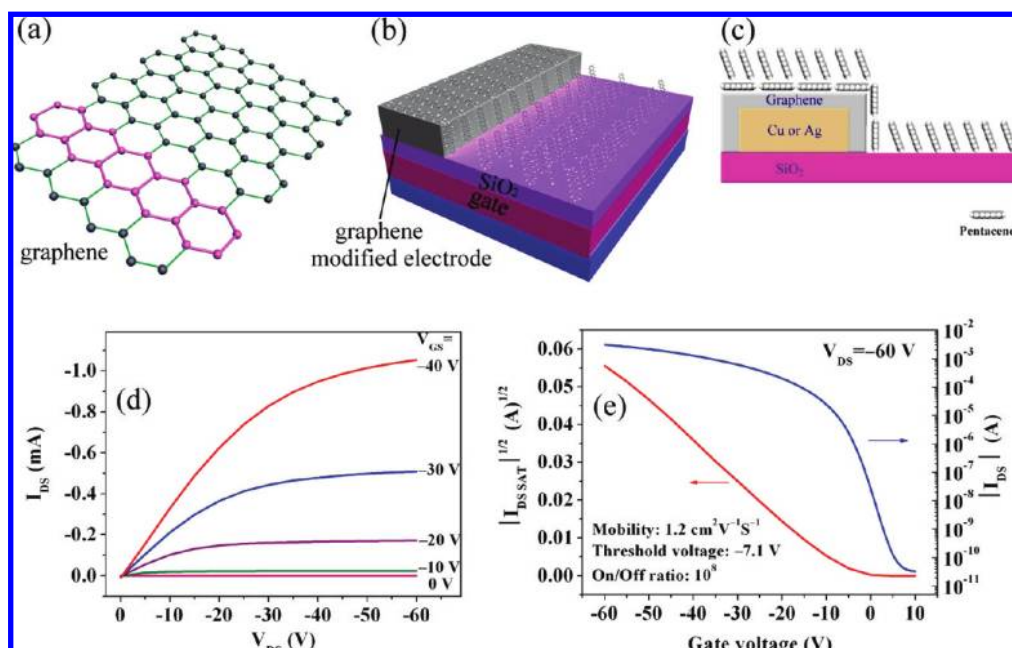


FIGURE 5. (a) Molecular structure of graphene, which possess a similar structure to pentacene. (b, c) Packing models of pentacene molecules on a graphene-modified electrode. (d) Output and (e) transfer characteristics of pentacene OFETs with graphene-modified electrodes.

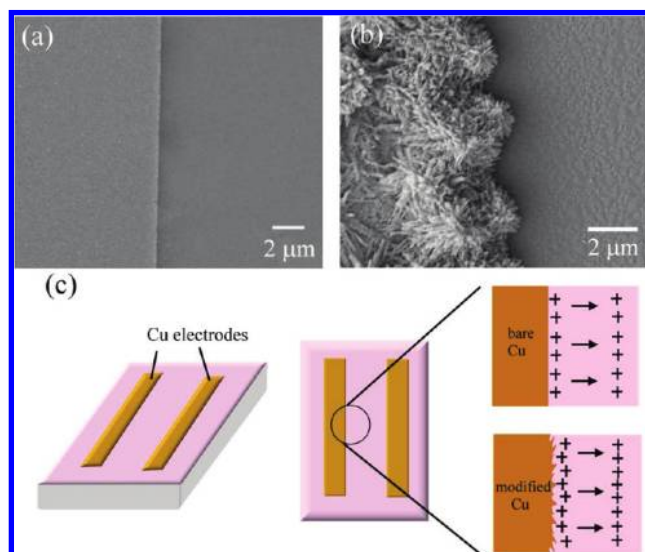


FIGURE 6. SEM image of OFETs with (a) bare Cu electrodes and (b) TCNQ-modified Cu electrodes. (c) Schematic illustration of improvement in the hole injection of the modified Cu electrode.²³

face possess larger electrode/organic contact areas than those with a flat electrode (Figure 6). Thus, many more carriers can inject into the channel under a certain voltage with rough source–drain electrodes, leading to reduced contact resistance. Furthermore, although the rough surface has a detrimental effect on the dielectric layer, source–drain electrodes with appropriate roughness do not lead to a significant adverse influence on the carrier injection.²³ We demonstrated that when a Cu electrode is modified by nanosized Cu–TCNQ with rough surface, it can serve as an excellent source–drain

electrode (Figure 6).²³ The modification roughness is an important factor that influences the contact area. For OFETs with Cu–TCNQ electrodes, modification of electrodes with roughness about 50–100 nm, which corresponds to about 2–5 times increase of surface area, is most suitable for the device fabrication. The large electrode/organic contact area ensures low contact resistance and contributes to the high extracted mobility. We achieved high mobility of $0.31 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ after nanosized Cu–TCNQ modification. This performance is about three times higher than a device without dense Cu–TCNQ nanostructure.^{21,24} This proved that controlling the electrode morphology is an effective technique to improve device performance.

Improvement of Carrier Transport. Source–drain electrodes are generally modified to optimize carrier injection. Little attention has been paid to improve carrier transport by modification of the electrode/organic layer interface because the interface is believed to have little influence on the conductive channel. In fact, the electrode can influence the growth of an organic layer near bottom source–drain electrodes. Gundlach et al. recently demonstrated induced crystallization of an organic layer by electrode interface modification.²⁴ By using pentafluorobenzene thiol (PFBT)-modified gold electrodes, 5,11-bis(triethylsilyl)ethynyl anthradithiophene is induced to grow with large grain domains near the source–drain electrodes, indicating that optimization of the source–drain electrodes is an alternative way to improve carrier transport. We recently achieved selectively polycrystal-

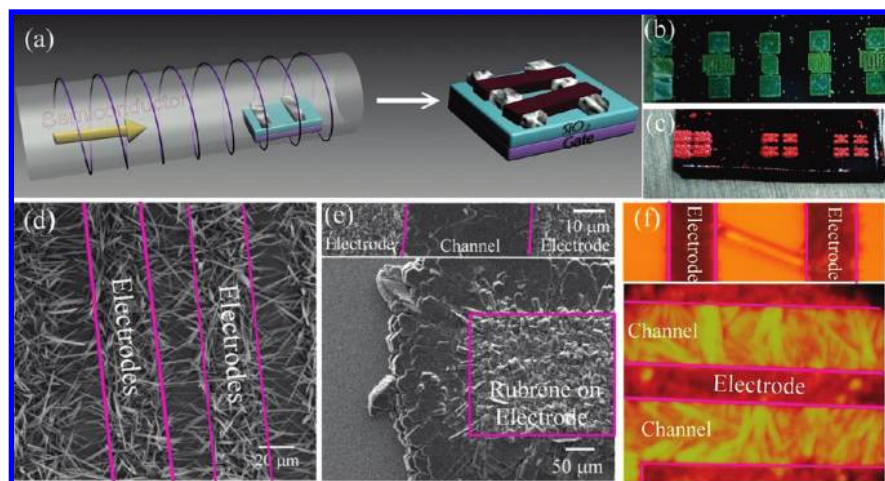


FIGURE 7. (a) Fabrication process of OFETs based on selective-patterned organic crystals. Optical micrograph of (b) TCNQ and (c) rubrene crystals on Cu–TCNQ modified electrodes. SEM images of (d) CuPc and (e) rubrene crystals near the Cu–TCNQ modified electrode and their corresponding devices. (f) Optical micrograph of PET crystals near the Cu–TCNQ modified electrode.

line growth of organic crystals by interface modification engineering. When the Cu source–drain electrodes were modified by nanosized Cu–TCNQ, two interesting phenomena were observed after vapor deposition process of organic crystals (Figure 7a). At first, preferential deposition of organic crystals near the electrodes was observed (Figure 7b).²⁵ Therefore, the crystal location can be controlled by the prepatterned electrodes. More interestingly, the crystalline growth direction can also be controlled by electrode modification. The π – π interaction direction is almost vertical to the electrode for many organic semiconductors like CuPc, rubrene, and perylo[1,12-*b,c,d*]thiophene (PET). This direction is the most favorable one for carrier transport. These two phenomena are related both to the morphology and to the molecular structure of the modified electrodes. According to classical nucleation theory, high surface energy lowers the barrier to heterogeneous nucleation.²⁶ Since the nanosized electrodes have rough surfaces, this leads to high surface energy and offers a superior surface for heterogeneous nucleation relative to a flat SiO₂ dielectric layer (Figure 8). The organic crystals are thus preferentially deposited near the modified electrodes. As for the vertical growth direction of these organic crystals, the interaction between electrodes and organic semiconductor and the strong intermolecular interactions both play a role. The interaction between electrodes and the organic molecules orients the first deposited molecule in a face-to-face packing with Cu–TCNQ. Subsequent deposited molecules form a well-ordered π – π stacking sequence with the first layer of deposited molecules. Therefore, the ordered crystal growth is transmitted to the channel by the electrode/organic semiconductor interactions and intermolecular π – π interactions. For many excellent organic semiconductors such as rubrene, strong π – π interac-

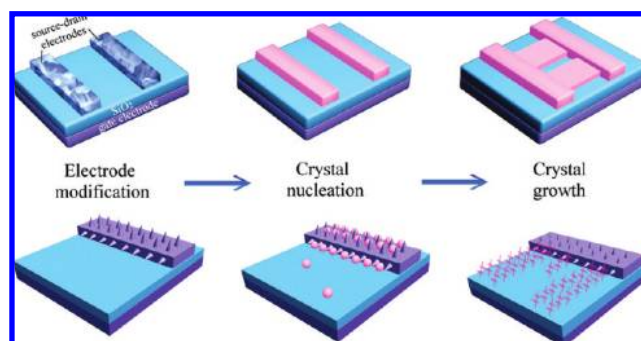


FIGURE 8. Illustration of the crystal growth process near the Cu–TCNQ modified electrodes.

tions dominate intermolecular interaction; their corresponding crystals thus can grow from the electrode with large size, which offers opportunity to fabricate polycrystalline or single crystal based OFETs. Since the packing direction is consistent with that of the source–drain field, it might bring on high device performance.

As mentioned above, both the location and crystal direction can be controlled. We thus can fabricate OFETs with selectively patterned crystals (Figure 7). The rubrene OFETs exhibit the highest mobilities, up to $4.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is comparable with rubrene single crystal devices with a SiO₂ dielectric layer.²⁷ The excellent device performance is ascribed to the following reasons. Although we fabricate the device with polycrystalline material, the crystal size is much larger than the grain in organic thin films. In fact, an OFET with rubrene crystals across the source–drain electrode, which is essentially a single-crystal device, can be easily obtained. The boundary density is thus minimized comparable to that of single crystal OFETs. On the other hand, the crystals grow from the electrodes and can ensure good electrode/crystal contact. These

two factors induce high device performances. Since the selective patterning technique of organic crystals is a simple while widely applicable one, it should be a promising approach for fabrication of high-performance OFETs.

Dielectric/Organic Layer Interface Engineering: An Effective Way To Improve Device Stability

Device stability is one of the biggest problems hindering commercial application of OFETs. It is generally accepted that the device stability is determined by organic semiconductors. In fact, the dielectric/organic interface also influences the stability in three different ways. First, the dielectric layer surface trap density affects the device performance dramatically, especially for n-type OFETs. Thus, optimization of a dielectric layer with low trap density can be used to improve stability. For example, *N,N*-dioctyl-3,4,9,10-perylene tetracarboxylic diimide (PTCDI-C8) is generally believed to be unstable in air. However, Chen et al. discovered that the stability of PTCDI-C8 based OFETs is related to the properties of the dielectric layer.²⁸ Modification of the SiO₂ surface with hydroxyl-free polymer insulators, such as poly(methyl methacrylate) (PMMA), afforded significant improvements in device stability. The hydrophobic nature of PMMA inhibits protonation of the siloxyl groups on the surface, leaving low electron trap density, and this is responsible for the enhanced stability.²⁸ Second, the dielectric layer can influence the morphology of the organic layers which in turn affect the device stability. In general, the organic layer boundary is more sensitive to air or water. Therefore, the boundary density and depth, which are important film parameters, can influence the device stability dramatically. We recently demonstrated that organic layers with large grain sizes, low boundary density, and low boundary depth have a beneficial effect on device stability.²⁹ In fact, optimization of the dielectric/organic interface is a general way to control the film morphology and thus should be a useful way to improve device stability. The third way that the dielectric/organic interface influences the stability is its influence on the aggregation of organic grains. For example, it has been observed that the stability of pentacene OFETs is also strongly related to the dielectric layer.^{30,31} Devices with OTS-modified dielectric layers exhibit much worse stability than those with bare SiO₂ or polystyrene (PS)-modified dielectric layers. We observed that the OTS-modified SiO₂ dielectric layer favors pentacene aggregation (Figure 9).³¹ No obvious aggregation was observed for devices with bare SiO₂ or PS-modified dielectric layers under the same conditions. Since these dielectric layers possess similar roughness, the dielectric layer surface

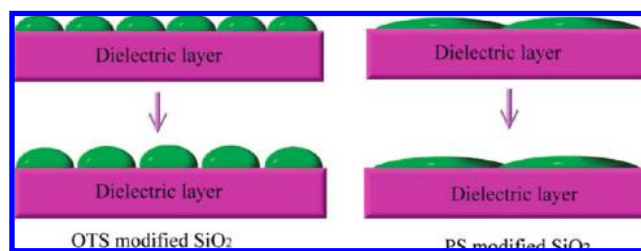


FIGURE 9. Illustration of pentacene grain aggregation on different dielectric layers.

energy should be responsible for aggregation of the pentacene film. When the surface energy of the dielectric layer is lower than that of the organic layer, the organic layer tends to aggregate in order to minimize the total surface energy, leading to instability of the film. For pentacene OFETs with OTS-modified dielectric layers, the surface energy of the modified surface is much lower than that of the pentacene layer. As a result, the pentacene layer readily aggregates, leading to rapid degradation of device performance. In contrast, the high surface energy of PS impedes pentacene aggregation and ensures excellent device stability for the devices with PS-modified dielectrics. Consequently, optimization of the dielectric/organic interface is an effective way to realize high-performance OFETs with excellent stability.

Organic/Organic Layer Interface Engineering: A Shortcut To Achieve Organic Light-Emitting Transistors

Organic light-emitting transistors (OLEFTs), a highly integrated organic optoelectronic device, achieve field effect and light emission in the same channel. Realization of two functions requires effective injection of both holes and electrons. Note that organic semiconductors for use in OLEFTs should possess both excellent field-effect properties and light-emitting properties. Unfortunately, most light-emitting materials exhibit poor field-effect mobility while excellent field-effect semiconductors usually possess unsatisfactory light-emitting properties. Introduction of two semiconductors into the channel is one way to resolve the problem. Therefore, organic layers having a blended composition or a double layer structure are usually employed in attempts to fabricate OLEFTs (Figure 10a,b).^{32,33} For the two kinds of devices, both p-type and n-type organic semiconductors are required to ensure injection and transport of holes and electrons. Although the devices obtained show good field-effect characteristics, the light-emitting properties are still not satisfactory. We have suggested a new device architecture, which we named laterally arranged heterojunction configuration.³⁴ As shown in Figure 10c, a heterojunction composed of organic field-effect materials and

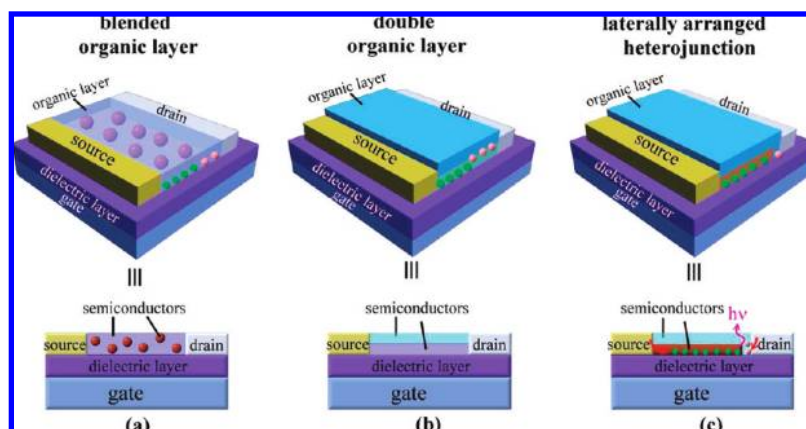


FIGURE 10. Structures of OFETs with organic layers made of two different semiconductors. The devices involve (a) a blended organic layer, (b) a double organic layer, and (c) a laterally arranged heterojunction.

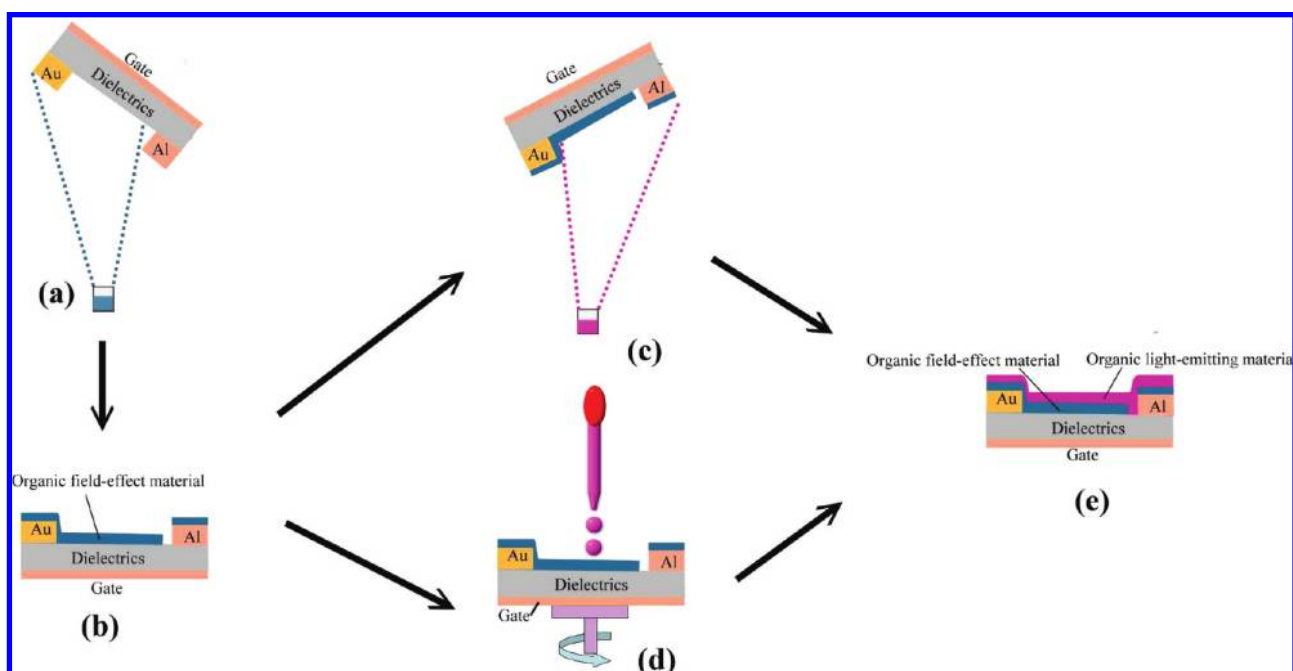


FIGURE 11. Schematic of the OLEFT fabrication process: (a) the substrates are mounted with an angle, and the organic field-effect semiconductors are evaporated; (b) the device structure after first deposition; (c) the substrates with oppositely inclined direction are mounted, and the organic light-emitting materials are deposited; (d) spin coating of polymer light-emitting material; (e) the laterally arranged heterojunction structures.

organic light-emitting materials is introduced vertical to the conductive channel. The unique structures are achieved by successively inclined deposition of the field-effect and light-emitting materials (Figure 11). After deposition of organic field-effect materials, a narrow gap of about 10–20 nm near the drain electrode was obtained, which can be used to fabricate a laterally arranged heterojunction. Interestingly, the device is also a unique structure for OLEDs where the anode (source) electrode, hole-transport material (field-effect material), light-emitting material, and cathode (drain) electrode are laterally arranged, thus offering a chance to control the electroluminescent intensity by changing the gate voltage.³⁴

Compared with OLEFTs based on single semiconductors, the new device structure possesses the following advantages. First, the energy levels can be optimized to ensure carrier injection of both holes and electrons since two different organic semiconductors are present in the channel. Then, one kind of carrier injects into the organic layer and transports through the channel to the heterojunction under the gate voltage, while the other kind of carrier transports through the short channel length to the heterojunction. Accumulation of high density holes and electrons occurs at the organic/organic layer interface due to the injection barrier at the heterojunction, which favors effi-

cient carrier combination and minimizes exciton quenching at the metal electrodes. Therefore, highly efficient light emission is obtained from the light-emitting materials and can be controlled by the gate voltage. More importantly, design of multifunctional semiconductors is not required since separate field-effect materials and light-emitting materials are introduced. We fabricated an OLEFT in such a device configuration with pentacene and tris(8-hydroxyquinolino)aluminum (Alq₃). The device exhibited good field-effect characteristics as well as satisfactory light-emitting properties even in ambient atmosphere. It is the first demonstration of air-stable OLEFT devices.

Conclusions

There has been great progress in OFETs in the past decade. Numerous novel organic semiconductors with outstanding properties have been designed, synthesized, and fabricated into OFETs. To realize actual applications, further efforts to improve overall device performance including high mobility, excellent stability, low cost, and low operating voltage are still required. Interface engineering offers novel ways to prepare high-performance OFETs. Herein, the interfaces of OFETs including the electrode/organic layer interface, dielectric/organic layer interface, and organic/organic layer interface are investigated to improve the device performance and stability, to reduce fabrication cost, and to realize organic light-emitting transistors. Despite these achievements, both opportunities and challenges for interface engineering still remain. To date, both the focus of attention and major achievements have been in the area of improving device performance by modification of the electrode/organic layer interface, and the dielectric/organic layer interface. Further investigation of the organic/organic layer interface and organic layer/atmosphere interface are still required. Exploration of novel applications of interface engineering also deserves further attention. As an example, interface engineering can play an important role in the design of gas sensors and multifunctional devices. In summary, with a deeper understanding of interface phenomena, better usage of interface properties, and exploration of novel interface modification approaches, interface engineering will dramatically boost further development of OFETs.

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FOOTNOTES

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