Chapter 1

Polymer Dielectric in Organic Field-Effect Transistor

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Additional information is available at the end of the chapter

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Abstract

In this chapter, we aim to present an overview of the polymer dielectrics in organic field-effect transistors and their applications. In the first section, we give a short introduction of polymer dielectrics in organic field-effect transistors. We illustrate multilayer, hybrid, and cross-linked polymer dielectrics adopted in organic field-effect transistors. Then we introduce the available biomaterials engaged as polymer dielectrics in organic field-effect transistors. We mainly focus on the utilization of silk fibroin, DNA, and DNA base pair dielectrics. We end the chapter by presenting the applications of polymer dielectrics. We elaborate that the polymer dielectrics can function as the electrode buffer layer, as well as the organic field-effect transistor-based gas sensor, inverter, and memory.

Keywords: polymer dielectric, organic field-effect transistor, biomaterial, gas sensor

1. Introduction

Organic field-effect transistor (OFET) is an indispensable component in the field of organic electronics, which has been developed to realize low-cost, flexible large-area products, and biodegradable electronics [1–3]. Compared with the conventional silicon dioxide–based device, OFETs with polymer dielectrics are ideally compatible with flexible substrates and solution process [4]. Apparently, the solution processable polymer dielectric is very attractive, for it is compatible with spin-coating, casting, and printing at room temperature and under ambient conditions. Meanwhile, this capability has practical advantages when coupled with large-scale production using the patterning technique [5]. Moreover, as the function dependent on the structure of polymer dielectric is readily available, to design and synthesize a structure with certain function becomes feasible [6], which results in complementary kinds of polymer dielectrics. Recently, with more and more polymer biomaterials engaged in the OFETs to serve

as the dielectric, the resource of polymer dielectric has become environmentally friendly and very broad. Due to such kinds of polymer dielectrics, OFETs are available in versatile applications including sensor, inverter, and memory [7].

2. Introduction of polymer dielectric in organic field-effect transistor

2.1. Polymer dielectric in organic field-effect transistor

In a common configuration, the OFET consists of the source and drain electrodes, semiconductor, dielectric, and gate. The functional part of an OFET device is the current channel, which exists in the first few monolayers of the semiconductor upon the dielectric. Therefore, the interface between the dielectric and the semiconductor plays a crucial role in the device performance, which makes the requirements for polymer dielectric material for OFET rather stringent [8, 9]. The crucial parameter of a dielectric material is the maximum possible electric displacement (D_{max}) the dielectric can sustain:

$$D_{\max} = \varepsilon_0 k E_{\mathrm{B}} \tag{1}$$

where ε_0 is the vacuum permittivity, k is the dielectric constant, E_B is the dielectric breakdown field, and the capacitance per area C_i, which is defined as:

$$C_{\mathrm{i}} = \varepsilon_0 \frac{k}{d} \tag{2}$$

where d is the thickness of the dielectric. It is obvious that the capacitance magnitude is not only governed by the k value but also by the thickness of the dielectric.

The first detailed study of different polymer dielectrics in OFET was reported by Peng et al. in 1990 [10]. The OFET was fabricated on glass using five kinds of polymer dielectrics. They found that there was a strong correlation between the insulator's k value and the field-effect mobility. Then, in 1997, the first high performance plastic transistor was realized by Bao et al. [11]. They employed polyimide as the dielectric and all the essential components were printed directly on the plastic. The OFET had a field-effect mobility of 0.01–0.03 cm^2/Vs. In 2002, Klauk et al. made a step further achievement as the "all-polymer" circuit which integrated a 250 nm thick melamine cross-linked poly(vinyl pyrrolidone) (PVP) [12]. The cross-linked PVP with the capacitance of ~11–12 nF/cm^2 made the OFET yield a high carrier mobility of 3 cm^2/Vs. In the next year, Veres et al. reported that the interaction between the dielectric and the semiconductor plays a crucial role in the charge carrier transport. They found out that for a larger permittivity of the dielectric, the more charge carrier was localized at the surface of the dielectric [13]. Thus, there is a contradictory selection between the high dielectric material and the low permittivity material. In order to obtain such a balance, in 2004, Park et al. introduced double polymer layers as the dielectric in OFET [14]. This structure consisted of a thin PVP layer in contact with

the semiconductor, which could induce good charge transport properties and a thick poly(vinyl acetate) (PVAc) layer as the bottom layer to realize good dielectric properties.

In recent years, the tendency of the research on polymer dielectrics has been toward printable, flexible, and biocompatible materials. In 2015, Huang et al. developed a versatile self-healing polymer blend dielectric without salts and integrated it into the OFET [15]. This high capacitance of polymer blend dielectric could even induce the healing of the functional layer coated above it. In 2016, Jung et al. introduced a kind of cross-linked poly(methyl methacrylate) (PMMA) in an n-type OFET, which examined the application of the OFET for flexible circuit [16]. Schmidt et al. fabricated a low-voltage fully printed flexible OFET using three layers of dielectric of CYTOP (low-k), PVA (intermediate), and P(VDF-TrFE-CTFE) (high-k) [17]. Moreover, more and more biomaterials, such as glucose, indigo, and nucleic acid–based materials, are used in the OFETs [18]. Among them, DNA and silk fibroin (SF) are two widely researched dielectric biomaterial in OFET. For example, Liang et al. had adopted DNA-hexadecyltrimethyl ammonium chloride (CTMA) as the dielectric layer in OFET to successfully realize a nonvolatile memory [19]. Wang et al. employed the SF as the gate dielectric in OFET and obtained a high mobility of 23.2 cm^2/Vs and a low operating voltage of -3 V [20].

2.2. Multilayer polymer dielectric

In OFETs, the bulk and interface properties of gate dielectric are both crucial in determining device performance. Therefore, two prerequirements have to be met for the dielectric material: (1) a high gate capacitance, which reduces the required gate voltage for sufficient charge accumulation in the channel, can be obtained by decreasing the film thickness or increasing the dielectric constant; (2) a trap-free interface between semiconductor and dielectric, which enhances the carrier mobility [17, 21]. A high *k* material has been considered as a good choice for the dielectric in OFET due to the excellent bulk insulating properties. However, the surface polarization of the high *k* material is much more higher than that of the low *k* material, which can significantly deteriorate the device performance. Therefore, to achieve high performance device, there should be a meticulous consideration between the high *k* and the low *k* materials. In this case, the utilization of the multilayer dielectric, combining a very thin low *k* layer at the interface to the semiconductor with a thick high *k* material for sufficient gate insulation is a reliable approach.

Yu et al. made an effective approach on multilayer dielectrics of PMMA and PVA in OFET [22]. Through analyzing the electrical characteristics of OFETs with various PVA/PMMA arrangements as shown in **Figure 1(a)**, it was found that one of the origins of the hysteresis was the trap in PVA bulk as well as at the interface of pentacene/PVA. Meanwhile, the results showed that the memory window was proportional to the amount of traps in PVA and the charge density at the interfaces of gate/PVA or PVA/pentacene. Then, the pentacene OFETs based on bilayer dielectrics of PMMA/SF was developed, as shown in **Figure 1(b)** [23]. The PMMA/SF bilayer dielectric exhibited a high field-effect mobility of 0.21 cm^2/Vs and a high current on/off ratio of 1.5×10^4. The performance enhancement was mainly attributed to the crystallization improvement of the pentacene and the smaller interface trap density at the SF/pentacene

interface. Meanwhile, a low contact resistance also indicated that a good contact of electrode/organic was formed.

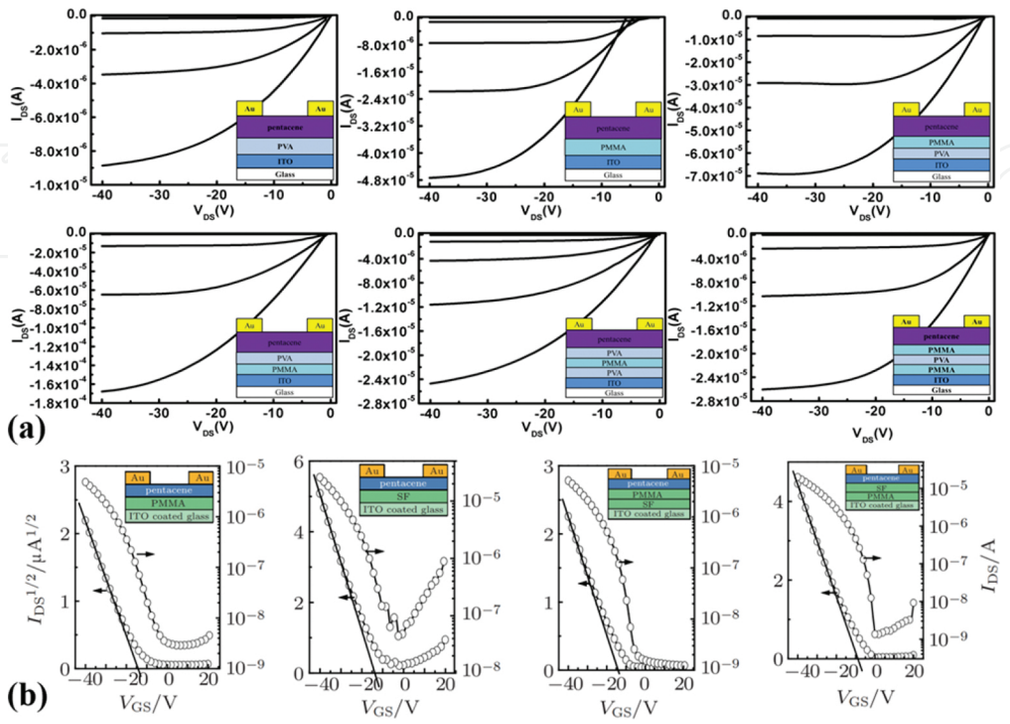

Figure 1. Configuration and performance of OFETs with (a) PVA and PMMA multi-layer dielectrics; (b) PMMA/SF bi-layer dielectrics [22, 23]. Copyright 2013, American Institute of Physics; Copyright 2014, IOP Publishing.

2.3. Hybrid polymer dielectric

An easily processable polymer typically has a low dielectric constant and good mechanical properties but requires large gate dielectric thicknesses due to high leakage current. One of possible solutions is to combine inorganic-organic or organic-organic materials as hybrid gate dielectrics. These complementary constituents ideally combine high permittivity of the inorganic inclusions with the high breakdown strength, mechanical flexibility, and easy processability of the organic counterparts [24]. Sun et al. explored a blending polymer of dielectric of polyphenyleneoxide (PPO) and polystyrene (PS) to enhance the performance of pentacene OFET [25], which could control both the inter-grain-enhancing process and the nucleation-controlling process. The optimized morphology of pentacene thus led to the enhancement of mobility to 3.6 cm^2/Vs. Yu and coworkers also employed the PMMA/zinc oxide (ZnO) hybrid as the dielectric for OFET [26]. The resulted morphology of the pentacene grown on the hybrid dielectric was responsible for the enhanced sensing performance to ammonia (NH_3) of the OFET.

Moreover, the polymer dielectric can not only serve as the dielectric layer in OFET but can also play a role in the semiconducting layer [27]. Yu et al. made it a step further to adopt the poly(3-hexylthiophene) (P3HT)/PS hybrid as the semiconductor [28]. The relationship between the

molecular arrangement, aggregation, and charge transport in P3HT:PS blends with the boiling points and solubility in different solvents like chloroform (CF), o-xylene (XY), chlorobenzene (CB), and 1,2-dichlorobenzene (DCB) was systematically analyzed, as shown in **Table 1**. The result showed that DCB with the highest boiling temperature was beneficial to achieve distinct lateral aggregation of P3HT in the blend film. An optimized composition of 1.6 wt% P3HT in the PS matrix with three times increase of mobility and two times increase of current on/off ratio were obtained, compared to that of the pure P3HT (8 wt%). Li et al. adopted a blend of 6, 13-bis(triisopropylsilylethynyl)pentacene (TIPS-pentacene) and PS as the semiconducting layer [29]. They synthesized the PMMA and PS functionalized with both propargyl and azido groups by free radical copolymerization to serve as the dielectric to obtain a high performance OFET with a field-effect mobility of 0.59 cm^2/Vs, and an on/off current ratio of 10^5. Feng et al. fabricated a solution processed bottom-gate bottom-contact OFET, which could be able to sustain hybrid low-/high-voltage operation [30, 31]. In their devices, a channel engineering approach was used to obtain low-voltage operation, by inducing phase separation with a blend of TIPS-pentacene and PS to form an ultrathin high crystalline channel. Since the approach did not rely on enlarging the gate dielectric capacitance, the low-voltage OFET with a relatively thick dielectric layer was shown to be able to sustain high-voltage operation.

		μ (cm^2/V s)	V_T (V)	*SS* (V/dec)	On/off ratio (10^2)
DCB	P3HT	0.010 ± 0.002	-1.5 ± 0.2	10.0 ± 0.2	2.7 ± 0.1
	P3HT:PS	0.030 ± 0.002	0.2 ± 0.05	6.0 ± 0.2	6.2 ± 0.1
CB	P3HT	0.004 ± 0.0005	3.0 ± 0.2	9.5 ± 0.2	1.4 ± 0.1
	P3HT:PS	0.010 ± 0.002	4.0 ± 0.2	6.0 ± 0.2	2.9 ± 0.1
XY	P3HT	0.010 ± 0.002	0.5 ± 0.05	9.5 ± 0.2	2.1 ± 0.1
	P3HT:PS	0.005 ± 0.0005	7.5 ± 0.2	11.0 ± 0.2	2.7 ± 0.1
CF	P3HT	0.002±0.0005	-5 ± 0.2	15 ± 0.2	1 ± 0.1
	P3HT:PS	–	–	–	–

Table 1. Characteristics of pure P3HT and P3HT:PS blend OFETs based on different solvents [27]. Copyright 2015, Elsevier.

2.4. Cross-linked polymer dielectric

Commonly, there are two problems that exist in polymer gate dielectrics. One is the stability and the other one is the electrical robustness [29]. Meanwhile, when multilayer polymer dielectrics are applied, the elimination of the dissolution or swelling problem of the upper layer toward the under layer is desired. Cross-linking method can not only enhance solvent resistance and thermal stability, but also improve the electrical robustness of dielectric materials [32]. Moreover, the interference of each dielectric in the multilayer structure can be solved through the cross-link process. Commonly, the cross-linking in the dielectric layer can be achieved by photo or thermal reactions [33–35].

Li et al. synthesized the PMMA and PS functionalized with both propargyl and azido groups by free radical copolymerization, which could be effectively cross-linked by thermal azide-alkyne cycloaddition reaction at a relatively low temperature of 100°C as shown in **Figure 2** [29, 36]. This bifunctional approach significantly improved the efficiency of cross-linking reactions in the solid state and substantially enhanced the solvent resistance of the cross-linked dielectric layers. The OFET exhibited a high device performance with a field-effect mobility of 0.59 cm^2/Vs, and an on/off current ratio of 10^5 as mentioned above in the "hybrid polymer dielectric" part. At the same time, they synthesized two azide functionalized polymers by free radical copolymerization [37, 38]. Each new polymer was effectively cross-linked with a small molecule cross-linker by a thermally activated reaction at 100°C. This cross-linking method is compatible with plastic substrates for flexible electronic applications.

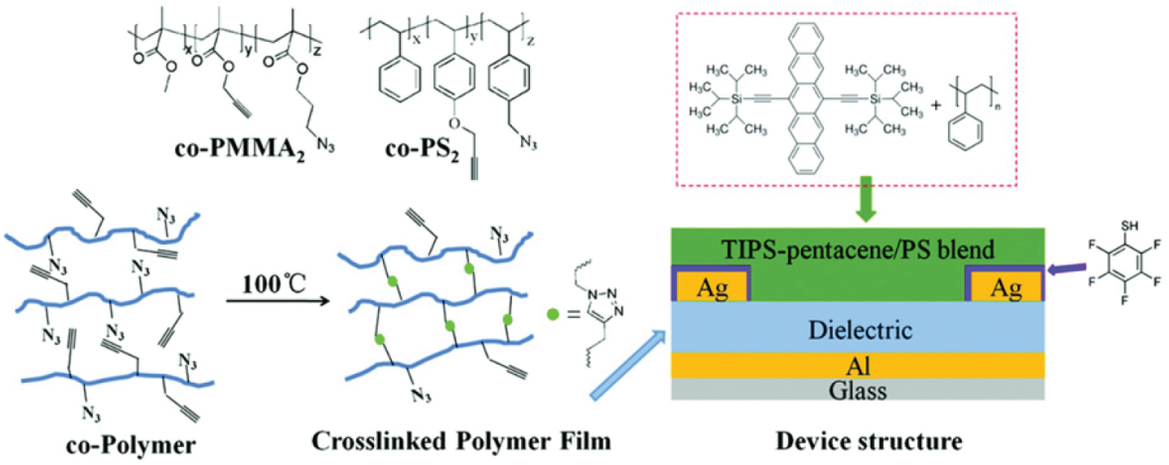

Figure 2. Schematic of the OFET and the chemical structures of the relevant materials [29, 36]. Copyright 2015, RSC Publishing; Copyright 2014, RSC Publishing.

From the above discussion, we can see that polymer dielectric is widely researched in OFET due to its inherent advantages of solution processability and flexibility. With the abundant resources, we can choose polymer dielectric with certain properties to serve different functions in hybrid and multilayer structures. Moreover, through cross-linking method, the atmosphere stability of polymer dielectric can be significantly enhanced, which brings a wider utilization for OFET and its electronic device.

3. Biomaterial-engaged polymer dielectric

3.1. Biomaterial in organic field-effect transistor

Nowadays, the research field of organic electronics is becoming more and more interdisciplinary, since biomaterial possesses superior attributes of chemical abundance, biodegradability, and low cost. OFETs are frequently used in the detection of biomaterials, whereas biomaterials are engaged in OFETs to serve as one of the functional layers as shown in **Figure 3** [18, 39–41].

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Substrates made of caramelized glucose, edible hard gelatin, and commercially available plastics based on potato and corn starch provide examples for metabolizable or biodegradable substrates [18]. DNA and DNA base pairs have been used as gate dielectric in OFETs and memory elements [42–44]. Beta-carotene and indigo have been employed as the p-type and n-type materials in OFETs, exhibiting the charge mobility of ~10^{-4} cm^2/Vs [45, 46].

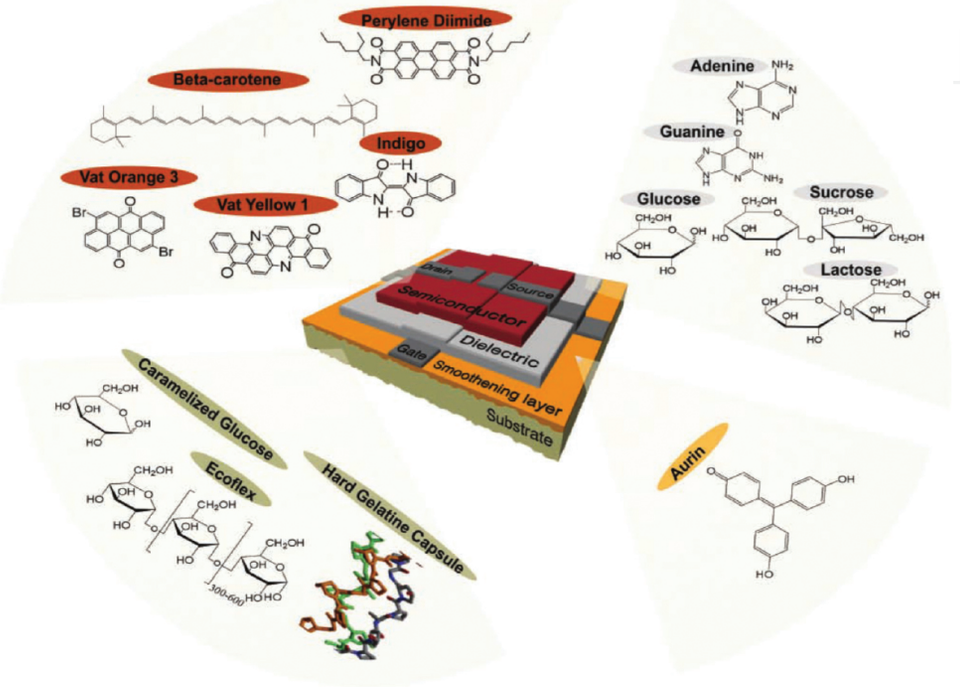

Figure 3. Utilization of natural materials or materials inspired by nature in OFETs [18]. Copyright 2010, Wiley.

3.2. Silk fibroin dielectric

Silk fibroin is one of the silk proteins emitted by the silkworm, which forms the structural center of silk with sericin around it. It is a natural biopolymer consisting of the repeated amino acid sequence of alternating glycine (gly) and alanine (ala) as shown in **Figure 4(a)** [47]. Until now, silk fibroin has been employed in various electronic fields, including the contact lenses [48], the platform for transistors [49], and photonic devices [50]. These applications are benefited from its unique characteristics of optical transparency, electrical insulation, and flexibility.

Yu et al. reported an enhanced performance pentacene OFETs consisting of PMMA/SF bilayer dielectric in 2014 [23]. The SF had good dielectric properties as shown in **Figure 4(b)**. The surface morphology of SF is very uniform with the root-mean-square surface roughness value of 1.3 nm as shown in **Figure 4(d)** (we can compare it with the smooth surface of PMMA shown in **Figure 4(c)**). The OFETs had a relatively high mobility of 0.21 cm^2/Vs with an enhanced on/off ratio of 1.5 × 10^4. This was mainly attributed to the crystallization improvement of the

pentacene grown on SF (as shown in **Figure 4(e)** and **(f)**) and the smaller interface trap density at the SF/pentacene interface. Then, the utilization of SF-engaged OFET to function as the nitrogen dioxide (NO_2) sensor was further studied [51]. In this research, SF was deposited on the top of PMMA to act as the dielectric layer, resulting in an increase of NO_2 sensing performance.

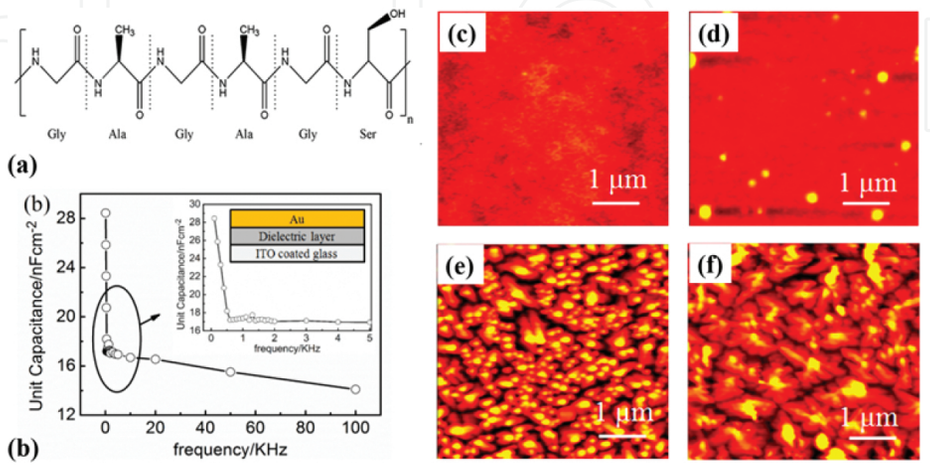

Figure 4. (a) Molecular structure of SF; (b) capacitance versus frequency (C-F) property of SF, the inset shows the C-F image in a frequency range of 0–5 KHz and structure of the device; AFM images of (c) PMMA; (d) SF; pentacene grown on (e) PMMA and (f) SF [23]. Copyright 2014, IOP Publishing.

3.3. DNA and DNA base pair dielectric

DNA is a complicated macromolecule consisting of four base pairs of guanine, adenine, thymine, and cytosine. Singh et al. first reported the utilization of DNA in OFET as a dielectric layer in 2006 [52]. They used a DNA-based biopolymer, derived from salmon milt and roe sac waste by-products for the gate dielectric, in which the current was modulated over three orders of magnitude using gate voltages less than -10 V. In most studies, since the purified DNA dissolves only in water, it should be modified through a cationic surfactant (hexadecyltrimethyl ammonium chloride, CTMA) cation exchange reaction to enhance solubility, processing, and stability.

Stadler et al. reported the utilization of DNA-CTMA as the gate dielectric in n-type methanofullerene as well as p-type pentacene-based OFETs working at low-voltage levels and low gate leakage currents [43]. They further realized a nonvolatile memory element based on the large hysteresis in the transfer characteristics of these DNA-based OFET. As DNA is soluble only in water, which is not compatible with most organic solvents, Yu et al. first introduced the spray coating method to fabricate DNA film. In 2016, spray-coated DNA on top of the PMMA dielectric was used to fabricate a DNA-functioned NO_2 sensor based on OFET as shown in **Figure 5(a)** and **(b)** [53]. The high-sensing performance is ascribed to the negatively-charged phosphate groups in DNA molecules, which can interact with NO_2 analytes as shown in **Figure 5(c)**.

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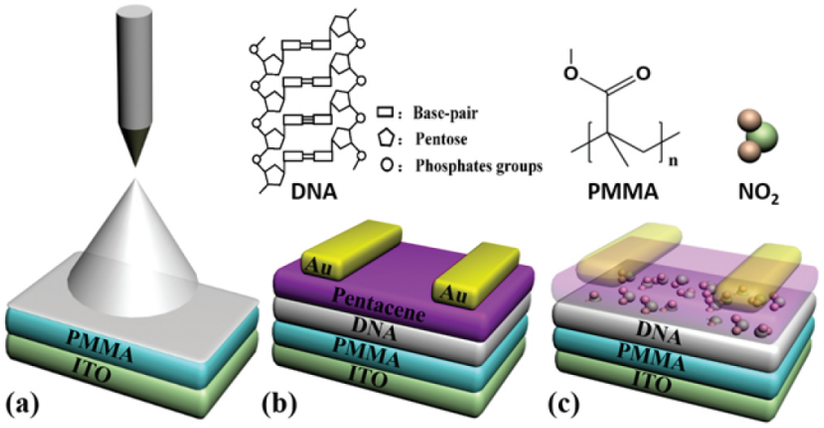

Figure 5. (a) Schematic representation of spray-coating technique; (b) device architecture of OFET sensor and the molecular structures of DNA and PMMA; (c) representation of the sensing mechanism of OFET-based NO_2 chemical sensor [53]. Copyright 2016, Elsevier.

Nature provides an overwhelming diversity of materials for human being. Hence, looking for natural or nature-inspired materials appears to be a promising route for the fabrication of fully biodegradable and biocompatible organic electronics. In the future work, to make the all biomaterial- based device, which is compatible with human body, is an attractive area of the biomaterial-engaged polymer dielectrics.

4. Application-related polymer dielectric

4.1. Electrode buffer layer in organic field-effect transistor

Dielectric polymers can not only serve as the dielectric layer in the OFETs, but can also be adopted as the modification layer at the interface between the semiconductor and the electrodes, due to their smooth and hydrophobic surface properties. Especially, the high performance bottom-contact OFET is required for its potential application in the large-scale industrialized production due to the remarkable advantage in the size-controlled fine lithography processing [54]. One of the major limits in fabricating high performance OFET in bottom-contact configuration is the large contact resistance at organic/electrode interface, which results in a pronounced current loss. This makes the employment of the electrode buffer layer necessary.

Yu et al. fabricated bottom-contacted OFET by using PMMA as an electrode buffer layer between P3HT layer and gold electrodes in OFETs and obtained a five-fold enhancement of whole mobility [55]. The relatively rough surface of gold (**Figure 6(a)**) is modified by PMMA (**Figure 6(b)**). The uniformity and hydrophobicity of PMMA surface were responsible for the remarkable reduction of contact resistance at P3HT/electrode interface while they enhanced the crystallinity of P3HT as shown in **Figure 6(c)**. Three polymer dielectrics of PMMA, PS, and polyvinylidene fluoride (PVDF) as the electrode buffer layers were further studied in top-contacted OFET to decrease the contact resistance (**Figure 6(d)**) [30]. All the OFETs incorporating with the buffer layers obtained a significant enhancement of the device performance,

whereas the device employing PMMA exhibited the highest charge mobility of 0.59 cm^2/Vs. This was due to the optimal surface energy and appropriate dielectric constant of PMMA, which are favorable for the growth of pentacene crystal.

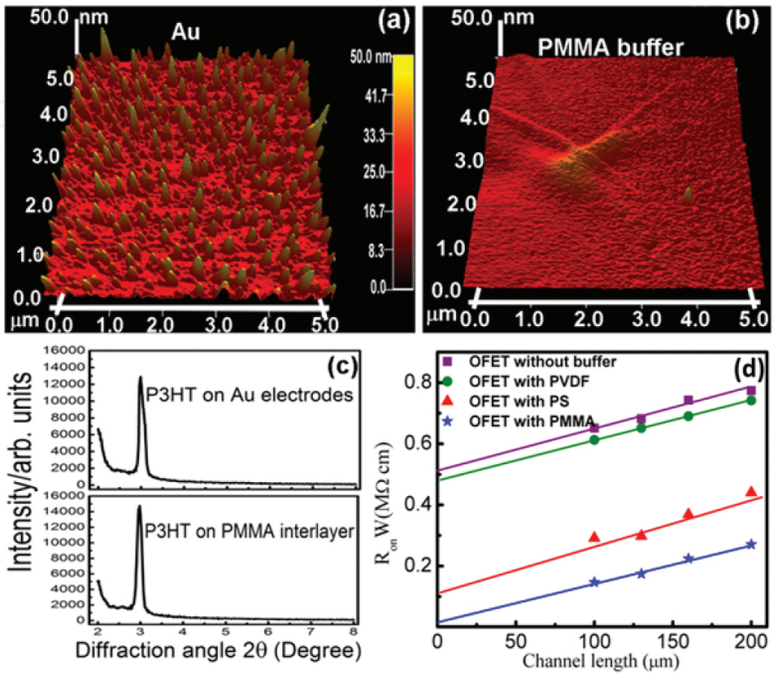

Figure 6. AFM images of (a) gold and (b) PMMA; (c) XRD analysis of P3HT; (d) contact resistance of OFETs [55]. Copyright 2013, American Institute of Physics.

4.2. Organic field-effect transistor-based gas sensor

OFET-based sensors have been intensively researched by virtue of their incomparable advantage of abundant material resource, physical flexibility, and elaborate array compatibility [56–59]. Moreover, OFET holds the unique capability of gate bias modulation, by which the signal can be easily amplified over orders of magnitude. This provides a promising future of OFETs for the ultra-low detection [60–64]. The controllable multiparameters of charge mobility, threshold voltage, and current on/off ratio make OFETs more suitable for selective detection [65]. Until now, OFET has been used to detect the solution, gas as well as the biomaterials [66].

Different kinds of polymer dielectrics perform differently in the OFET sensors. Yu et al. investigated the effect of various kinds of polymer dielectrics on the sensing performance of the OFET sensors. NH_3 gas sensors were fabricated based on pentacene OFETs using polymers including PVA, PVP, PMMA, or PS as the gate dielectric as shown in **Figure 7(a)** [67]. The OFETs with PS as the gate dielectric could achieve the detection limit as low as 1 ppm. Meanwhile, the recovery properties of OFETs with PS were also enhanced. The variation of the sensing performance of OFET sensors with different dielectrics was proved to be mainly induced by the different properties of dielectric/pentacene interfaces. Furthermore, the low-trap density of PS dielectric surface and the absence of polar groups in PS dielectric were responsible for

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the high performance of NH_3 sensors. Moreover, the OFET incorporating ZnO/PMMA hybrid dielectric was fabricated to detect NH_3 as shown in **Figure 7(b)** [26]. The OFETs exhibited a 23% current change under 75 ppm NH_3, as well as a remarkable shift of threshold voltage and field-effect mobility. The sensing mechanism was ascribed to the decreased grain size of pentacene formed on the ZnO/PMMA hybrid dielectric, facilitating NH_3 to diffuse into the conducting channel.

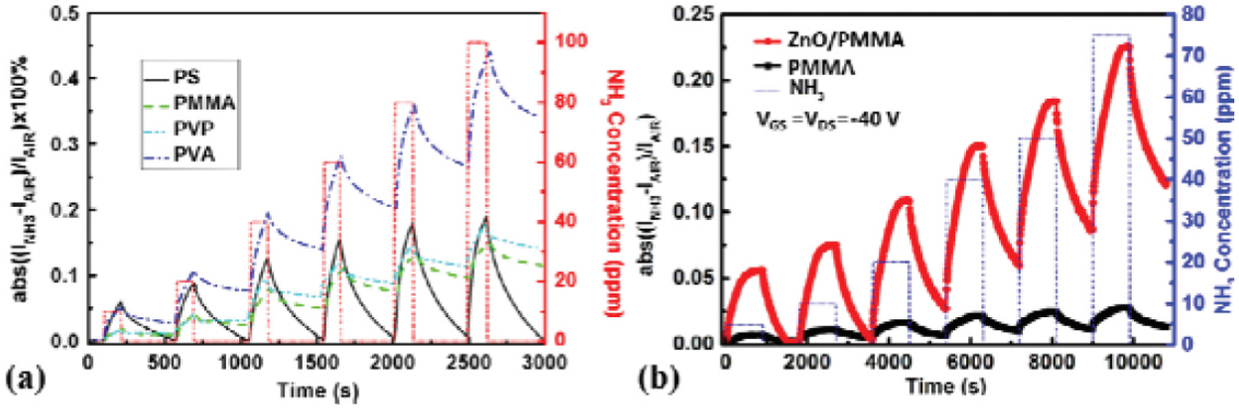

Figure 7. Real-time response curves of (a) OFET with PS, PMMA, PVP, and PVA as dielectric; (b) OFET with ZnO/PMMA and PMMA as dielectric exposed to NO_2 [26, 67]. Copyright 2014, Elsevier; Copyright 2013, Elsevier.

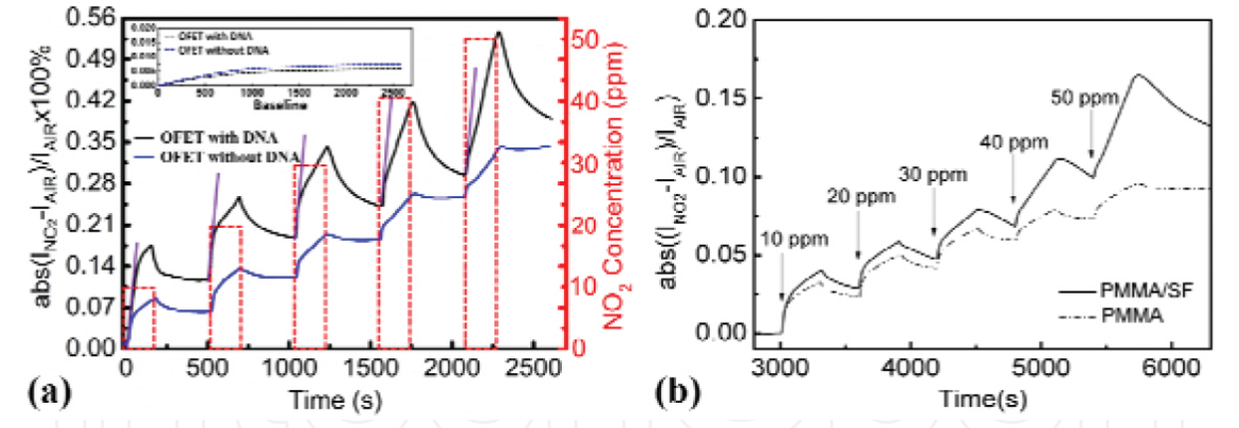

Figure 8. Real-time response curves of (a) OFET with and without DNA; (b) OFET with and without SF exposed to NO_2 gas in the concentrations ranging from 10 to 50 ppm [51, 53]. Copyright 2015, Springer; Copyright 2016, Elsevier.

In terms of the biomaterials discussed above, they also exhibit good sensing performance in OFETs. A DNA-based OFET sensor was realized for the detection of NO_2 as shown in **Figure 8(a)** [53]. In this sensor, DNA was introduced between the gate dielectric and the organic semiconductor via spray-coating to function as the detecting layer for NO_2 analyte. There were remarkable shifts of 14.7% in saturation current and 14.4% in charge carrier mobility after exposure to 10 ppm NO_2 analyte. With the concentration of NO_2 increased to 50 ppm, the shifts of 22.8% in saturation current and 16.6% in charge carrier mobility had been obtained. The

sensing performance was ascribed to the negatively-charged phosphate groups in DNA molecules, which could interact with NO_2 analytes and lead to a superior sensing performance of OFET incorporating with DNA (**Figure 5(c)**). Silk fibroin was also employed on top of PMMA dielectric in OFETs to detect NO_2 as shown in **Figure 8(b)** [51]. When exposed to a high NO_2 concentration of 50 ppm, the saturation current of the OFETs exhibited an increase of 16%. The superior sensing performance was due to the interaction between the hydroxyl and the amidogen of the SF biomaterial and NO_2 molecules at the interface of dielectric/organic layer.

4.3. Organic field-effect transistor-based inverter

Inverters are the basic and indispensable parts in the case of electronic circuits. Especially, complementary inverters, which usually consist of both p-type and n-type transistors, are widely applied in silicon-based circuits. Besides, due to the performance inconsistency of p-type and n-type organic semiconductors, unipolar inverter, which can be functionalized with only one type of transistors, is extensively studied in the field of organic electronics [68, 69]. The OFETs with a significant controllable threshold voltage (V_{th}) are indispensable to construct the high performance unipolar inverter. Ultraviolet (UV)/ozone (UVO) treatment can modify the physical and chemical characteristics of the polymer surface, leading to a shift of the V_{th} without changing the properties of the bulk material [70, 71].

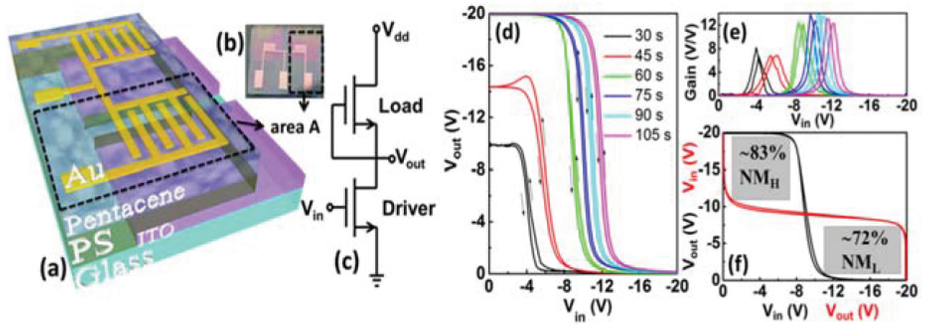

Figure 9. (a) Schematic configuration, (b) photograph, and (c) circuit diagram of unipolar inverter. (d) VTCs and (e) corresponding voltage gains of the inverters; (f) NMs of the inverter used OFET with 60 s UVO treated PS as load transistor [72]. Copyright 2014, American Institute of Physics.

PS was also adopted as the gate dielectric in OFET to fabricate a unipolar inverter as shown in **Figure 9(a)–(c)** [72]. This inverter was based on a significant variation of threshold voltage (V_{th}) of OFETs, which was realized by introducing UVO treatment to PS dielectric. A controllable V_{th} shift of more than 10 V was obtained in the OFETs by adjusting the treating time, and the unipolar inverters exhibited an inverting voltage near 1/2 driving voltage and a noise margin of more than 70% of ideal value as shown in **Figure 9(d)–(f)**. The dramatic controllable V_{th} of OFETs was attributed to the newly generated oxygen functional groups in the PS dielectric induced by UVO treatment. Guo et al. developed a novel organic/inorganic hybrid integration architecture to realize low-voltage complementary inverters with low temperature (not exceeding 150°C) solution-processed semiconductor and PVA dielectric layers [73]. The fabricated inverter had a voltage gain larger than 15 at an operation voltage of 3 V. The inverters

were further fabricated on a polyethylene naphthalate plastic substrate with the bottom-gate bottom-contact configuration [28]. In the devices, UV cross-linked PVA was used as the gate dielectric layer and the devices show a high DC voltage gain up to 67.3 at a supply voltage of 3 V. Also, the inverter with the low-operation voltage based on a channel engineering approach was fabricated [74]. The relatively thick (even with a 400-nm thick) and low dielectric constant polymer dielectric of PVA could be used in the inverter.

4.4. Organic field-effect transistor-based memory

OFET-based memory mostly relies on the hysteresis of the dielectric. As to a nonvolatile transistor, a voltage is added between the gate electrode and the semiconducting channel. Through the charge storage, the effective gate voltage within the device then differs from the applied voltage. The resulted polarization phenomenon creates an additional electronic state in the device. PVA is a commonly adopted polymer dielectric in OFET-based memory. The hysteresis mechanism of OFETs with PVA dielectric can be concluded in two aspects: the charge transport in PVA bulk and the charge trapping/detrapping process in PVA bulk and/or at the interface of organic semiconductor/PVA.

The research on multilayer dielectric of PMMA and PVA in OFET was also carried out [22]. Through analyzing the electrical characteristics of OFETs with various PVA/PMMA arrangements, it was found that one of the origins of the hysteresis was the trap in PVA bulk as well as at the interface of pentacene/PVA. Meanwhile, the results showed that the memory window was proportional to the amount of traps in PVA and the charge density at the interfaces of gate/PVA or PVA/pentacene. Therefore, the memory window could be controlled to around 0–10 V by tuning the thickness and combination of triple-layer polymer dielectrics strategy. Meanwhile, as the dielectric interfaces also greatly influenced the number of densities and the mobility of charge carrier in adjacent semiconductors, the surface issue should also be taken into consideration. Feng et al. fabricated a novel type of OFET-based write-once read-many memory (WORM) device [75]. The device used an ultraviolet cross-linkable matrix dielectric polymer of poly (vinyl cinnamate) (PVC) mixed with ionic compounds 10-methyl-9-phenyl-acridinium perchlorate ($MPA^+ClO_4^-$) to form an ion-dispersed gate dielectric layer. Under an applied gate voltage bias, the migration of cations and anions in opposite directions formed space charge polarization in the gate dielectric layer, resulting in a change of electrical characteristics. Through UV illumination method to cross-link the matrix polymer, the stability of the formed space charge polarization could be enhanced. Hence, the OFET could function as a WORM with the applied voltage bias to define the polarization, and at the same time the UV illumination could stabilize the stored data.

There is a wide utilization of OFETs functionalized by polymer dielectric. The most common adopted application is the OFET-based sensors. As the effective current channel lies at the interface between the semiconductor and the polymer dielectric, to modify the surface of the dielectric becomes an effective way to enhance the sensing performance. Moreover, polymer dielectric can also play a role in the electrode buffer layer as well as the inverter and memory, which makes the polymer dielectrics not simple function as the dielectric, but also as a versatile material.

5. Summary

In this section, we make a concise introduction of polymer dielectric in OFETs, including the multilayer, hybrid, and cross-linked polymer dielectric. In addition, we had a detailed overview of the available biomaterials of the silk fibroin and the DNA and DNA base pair as polymer dielectrics. As one of the most important layer in OFETs, polymer dielectric possesses a promising application as interface modification layer in OFETs, as well as the functional layer in OFET-based sensor, inverter, and memory. Hence, the polymer dielectric holds versatile inherent properties to be explored and is paced toward the future in the field of organic electronics.

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