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Recent Technological Advances in Fabrication and Application of Organic Electrochemical Transistors

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Abstract

Organic electrochemical transistors (OECTs), where conjugated polymers undergo doping/ de-doping from an electrolyte, have been widely studied for applications ranging from switching elements, artificial synapses to transducers for biological sensing. The concurrent transport of electronic and ionic charges within the channel provide OECTs with excellent amplification capability and efficient ion-to-electron transduction at low operating voltages ($<1V$). Herein, the latest research advances in the fabrication techniques and applications of OECTs are summarized. Particular focus is given on the emerging applications including sensors, neuromorphic devices and electrophysiological monitoring. Finally, the future challenges and opportunities of conventional OECTs and novel all-solid-state OECTs are discussed.

Keywords: Organic electrochemical transistors, conjugated polymers, doping-dedoping process, fabrication technologies, biological sensors

1. Introduction

In recent years, organic electrochemical transistors (OECTs) have been intensively studied based on the advancement of various conjugated polymers with high conductivity, inherent flexibility, and facile processability.^[1-3] The architecture of OECTs is similar to that of organic field-effect transistors (OFETs), except that an ion-conducting electrolyte is used between the conjugated polymer channel and gate electrode, which is one of the defining characteristics of OECTs and contributes to

their unique electronic properties.^[4-6] In an OECT, ions from an electrolyte penetrate the channel and modulate the bulk conductivity through electrochemical doping. Moreover, the color and volume of a conjugated polymer can also be typically controlled upon changing the polymeric redox state. These characteristics indicate the basic mechanism of operation in OECTs.

Transducing between electronic and ionic currents is extremely complex due to the intrinsic physics of different charge carriers. The prevalent use of organic semiconductors, rather than their inorganic counterparts, for transduction purposes is due to its “soft” character,^[7, 8] which can facilitate the uptake of ions throughout the bulk during the electrochemical doping and dedoping process,^[9, 10] resulting in an efficient ion-to-electron signal conversion.^[11-14] The classical organic semi-conducting material used for OECTs is an ionomer mixture called PEDOT:PSS (poly(3,4-ethylenedioxythiophene) polystyrene sulfonate), comprising of a conjugated PEDOT phase and a deprotonated PSS phase. Holes in the PEDOT phase is compensated by the negatively charged sulfonate ions in the PSS phase, making the PEDOT phase conducting. In the absence of a gate bias ($V_{GS}=0$ V), the flow of mobile holes within the polymer maintains a hole current when a drain voltage (V_{DS}) is applied (ON state). By applying a positive gate voltage ($V_{GS}>0$ V), cations from the electrolyte are injected into the channel and PSS anions are charge compensated, initiating the de-doping of PEDOT, reducing the hole density and thus lowering the drain current (OFF state). Such mode of operation is known as depletion mode; the OECT is ON in the absence of a gate bias. It is also possible to operate the OECT in accumulation mode if the conjugated polymer does not consist of any native dopants. Accumulated-mode OECT is typically in the OFF state and application of a negative gate voltage leads to injection of anions into the semiconducting polymer, initiating an electrochemically driven hole accumulation in the channel, leading to the ON state. Recently, naphthalene diimides (NDIs)-based polymers^[15], ladder-type polymers such as poly(benzimidazobenzophenanthro-line) (BBL)^[16], fullerene derivatives with glycolated side chains^[17] and random copolymer with glycol side chain such as P-90^[9, 18] were reported to be promising n-type materials to develop accumulation mode

OECTs with stable operation in aqueous environments. Another class of promising p-type materials for accumulation mode OECTs is semiconducting polymers based on benzodithiophene or bithiophene core with glycol side chains^[19, 20], glycolated thiophene (g2T-T) polymers^[21], poly(3-hexylthiophene) (P3HT)^[22, 23], conjugated polyelectrolytes based on poly(6-(thiophene-3-yl)hexane-1-sulfonate) tetrabutylammonium (PTHS)^[24], polythiophene derivative with ethylene glycol-based side chains (P3MEEMT)^[12] and poly(6-(thiophen-3-yl)hexane-1-sulfonate) tetrabutylammonium (PTHS:TEA)^[25]. Generally, the material must have simultaneously an aggregated, interconnected structure that supports efficient hole transport, and an amorphous phase to hydrate, and thus enable the transport of ions. The in-depth introduction on active materials for OECTs can be found in several review papers^[26-30].

The key feature of OECT is that the transition of doping/dedoping states occurs over the entire channel volume, as opposed to a thin interfacial region like in OFETs, and thus the total number of charge carriers is a function of the volume of the semiconductor channel. Therefore, large modulations in the drain current (I_{DS}) of the polymer channel between the source and drain contacts can be achieved for low-gate voltages (<1 V).^[31, 32] The corresponding transconductance ($g_m = \partial I_{DS} / \partial V_{GS}$) is typically large, on the order of millisiemens (~ 2.7 mS – 52.74 mS) for micrometer-scale devices^[3, 14, 31, 33, 34], making OECTs ideal ion-to-electron transducers. On the basis of this principle, the OECT characteristics are normally sensitive to ion concentration in the electrolyte, which is suitable for ion sensors^[35] and pH sensors^[36]. Moreover, by selective surface modification of the gate electrode with specific nanomaterials or enzymes, change in effective gate voltage can be monitored as a function of the concentration of chemical and biological analytes, which has attracted much attention for the development of chemical sensors^[36] and biological sensors^[37]. Furthermore, the soft, biocompatible organic materials as OECT channel enable an amiable interface with cells and tissues in aqueous environments (water-based electrolytes), thus extremely suitable for incorporating with bioelectronics including the monitoring of cell coverage, barrier tissue arrangement

and brain activity.^[38, 39] More advanced form of OECT arrays have been demonstrated as transparent active microelectrode arrays^[40], sensors for mapping activation potential in cardiac tissues^[41], and high temporal resolution electrophysiology arrays^[42]. More recently, OECTs are also developed for neuromorphic computing and memory matrices, where the transition between semiconducting and conducting states in these redox polymers offers a bistability in the current-voltage behaviors.^[43, 44]

In the aspect of fabrication technology, the simple device structure of OECT allows facile solution and printing processes such as inkjet printing^[45] and screen-printing^[46] and thus present unique advantages for low-cost and large area electronics. Another distinct advantage of OECT is that the electrical performance tends to be less affected by the surface roughness of the substrates and therefore suitable for flexible and wearable applications which require a variety of non-planar substrates. There have been numerous reports on flexible and stretchable OECT devices^[47-51], conformal OECTs integrated with natural and synthetic fibers^{[52],[53]} for sweat, neurotransmitters, and glucose sensors, as well as circuits built on paper substrates^[54, 55], and green OECT-based electronic circuits manufactured in living plants^[56], which demonstrate the versatility of OECT device design and indicate the rapid development of OECT technology and application. A summary of the various fabrication techniques and applications for OECT is presented in **Figure 1**.

In this review, we summarize the recent state of the art in OECTs, mainly focusing on the various fabrication processes and emerging applications of OECTs including sensors, neuromorphic devices, electrophysiological monitoring and the integrated electronic devices. Finally, the future challenges and opportunities of conventional OECTs and novel all-solid-state OECTs are discussed.

Figure 1. Schematic illustration of organic electrochemical transistors (OECT) in development today across facile fabrication techniques and broad range of applications. Printing process: screen printing, adapted with permission,^[57] Copyright 2019, Wiley-VCH; spray printing, adapted with permission,^[58] Copyright 2020, American Chemical Society. Laser-based patterning: laser ablation patterning, adapted with permission,^[59] Copyright 2012, Wiley-VCH; laser patterned OECTs, adapted with permission,^[60] Copyright 2020, IOP Publishing Ltd. Nanoimprint lithography: S/D nanoimprint, adapted with permission,^[61] Copyright 2016, Wiley-VCH; active layer nanoimprint, adapted with permission,^[62] Copyright 2013 SID. Fiber-based patterning: nanofiber-based, adapted with permission,^[63] Copyright 2019, American Chemical Society; woven-based, adapted with permission,^[53] Copyright 2011, Wiley-VCH. Biological sensor: glucose sensor, adapted with permission,^[64] Copyright 2019, Springer Nature; ion sensor, adapted with permission,^[65] Copyright 2018, Wiley-VCH. Logic circuit: Inverter and NAND, adapted with permission,^[46] Copyright 2019, Springer Nature; Wheatstone bridge, adapted with permission,^[66] Copyright 2017, Wiley-VCH. Neuromorphic device: nanowire artificial synapse, adapted with permission,^[67]

Copyright 2016, American Association for the Advancement of Science; short-term synapse, adapted with permission,^[68] Copyright 2019, Wiley-VCH. Electrophysiological monitoring: EEG signal, adapted with permission,^[69] Copyright 2019, American Association for the Advancement of Science; EMG signal, adapted with permission,^[42] Copyright 2016, Wiley-VCH.

2. OECT Fabrication Methods

In OECTs, the distinct volumetric redox processes determine the degree of electrochemical doping and therefore the channel geometry plays a critical role in determining the electrical performance. According to the OECT operation model developed by Bernards and Malliaras^[70], the transconductance (g_m) in the saturation regime is given by:

$$g_m = \frac{\partial I_{DS}}{\partial V_{GS}} = (W d/L) \mu C^* (V_{TH} - V_{GS})$$

which shows a linear function of g_m with channel geometry (Wd/L), where C^* is the volumetric capacitance of the channel, μ is the carrier mobility, V_{TH} is the threshold voltage, W , d , L are the channel width, thickness, and length respectively. On the other hand, the channel capacitance (C) is highly dependent upon the channel volume ($C \propto WL/d$) and the transient response time (τ_i) increases monotonically with channel geometry $\tau_i \propto d\sqrt{WL}$.^[4, 71] Therefore, one of the key challenges to fabricate OECTs lies in precise control of the channel area. The state-of-the-art fabrication processes for OECTs are dry lift-off and spin-coating processes that involve multiple photolithography steps to achieve the sub-micro scale patterned channel. Briefly, it comprises of photoresist-assisted patterning of source/drain pattern; followed by physical vapor deposition of insulating parylene layer to protect the electrode and a photoresist layer to protect the parylene during dry etching, the opening for the channel and contact pad, the deposition of channel layer and lift-off process.^[72, 73] The parylene layer prevents leakage gate current between gate electrolyte and source electrode and the lift-off process removes the excess semiconductor materials outside the channel area, thus reducing the parasitic effects. Although nanoscale OECTs ($L = 50$ nm) have been fabricated^[22], the conventional microfabrication photolithography technique poses certain limitations (i.e. high-cost, complicated process) that curb the commercialization of OECTs and may invalidate the large-area and scalable manufacturing. In addition, it also requires the assistance of various organic solvents such as acetone or photoresist developers which may delaminate or damage the

polymeric channel. In this section, we focus on other low-cost, facile techniques that have been developed to fabricate well-patterned and/or large-area OEETs.

2.1 Printing Processes

Conducting polymers, especially PEDOT:PSS, possess soft nature, solution processability, high conductivity and biocompatibility, which are suitable for the deposition of both active channel and electrode in a printing method^[74]. Various printing technologies have been developed to fabricate OEETs-based logic circuits or biosensors including screen-printing, spray-printing, aerosol-jet printing and inkjet printing.

Figure 2. (a) Illustration of the screen printing technology.^[57] Copyright 2019, Wiley-VCH. (b) The device configuration of an all-printed OEET with (c) the associated electrical performance. (d) Photographs of the OEETs based on a BCD decoder and (e) the corresponding logic voltage output sequence of segment of [1,0,1,1,0,1,0,1,1,1] with a input signal of [0,1,2,3,4,5,6,7,8,9].^[46] Copyright 2019, Springer Nature. (f) Illustration of a spray printing process using a commercial air-brush. Inset is the optical image of the spray coated PEDOT:PSS channel on various substrates. (g) Transient response of the spray-coated channels as a function of $d(WL)^{1/2}$.^[58] Copyright 2020, American Chemical Society. (h) Schematic illustration of the inkjet printing process and photographs of stretchable OEETs with the associated transfer characteristics under different strains prepared by inkjet printing technique.^[50] Copyright 2019, Wiley-VCH. (i) Photograph of glucose sensitive OEETs prepared by the inkjet printing technique.^[75] Copyright 2018, Springer Nature. (j) An alcohol sensing OEET fabricated by the inkjet printing technique.^[76] Copyright 2016, Springer Nature.

To date, screen-printing is one of the most popular approach to large-area deposition and has been widely utilized to prepare organic active layers for various applications such as solar cells, organic light emitting diodes (OLED), OFETs, and OEETs.^[77-79] The printing mechanism is based on forcing the ink through a screen or mask by applying pressure with a squeegee and forming the patterned area (see **Figure 2a**).^[57] Generally, the thickness of the printed layer relies on the thickness of emulsion of the screen and the viscosity of ink, which can be controlled in the range from hundreds of nanometers to several micrometers. However, this technique is not appropriate for high-resolution printed electronics because of its limited spatial resolution (typically more than 50 μm) and strong influence on the quality of the printed materials deriving from the speed of squeegee and viscosity of solution. Moreover, it also needs a different frame for each pattern and high material wastage in the printing process. Even so, the screen-printing technique is considered a simple, efficient, and low-cost method for the fabrication of large area OEET-based integrated circuits.^[46, 80-85] As shown in **Figure 2b**, coplanar OEETs were screen-printed, where a printable UV-curable electrolyte was sandwiched between the bottom channel and top-gate electrode.^[46] In such lateral OEETs, the printed PEDOT:PSS channel exhibited an ON/OFF ratio of $\sim 10^5$ and switching times of ~ 20 ms (ON-to-OFF) and 30 ms (OFF-to-ON), with a miniaturization of the OEET footprint down to ~ 1 mm^2 (See **Figure 2c**). Further, large-area screen-printed logic circuits including seven-bit shift register (consisting of 114 OEETs) and binary-coded decimal decoder (consisting of 87 OEETs) have recently been demonstrated (see **Figure 2d and 2e**). In addition,

Gustafsson et al. reported screen-printed vertical OECTs with channel dimensions of $W \times L \times d = 200 \times 200 \times (0.3-0.4) \mu\text{m}^3$, in which the electrolyte vertically bridges the bottom channel and top gate electrode.^[79] The low-voltage operation (1-3 V) and high current throughput (~mA scale) of screen-printed OECTs is beneficial to control the light emission in LEDs, in which the actual LED addressing was accomplished by an OECT-based decoder circuit. Organic electrochromic displays (OECDs), in which the device architectures and electrochemical switching mechanism are similar to that of OECTs, have also been fabricated using screen printing in a concurrent manufacturing process.^[81, 86] The all-screen printed OECTs and OECDs were monolithically integrated and manufactured on flexible plastic substrates for distributed smart sensor labels and targeted Internet of Things (IoT) applications. Thus, screen-printing technique makes it possible to develop large-scale logic circuits with arrays of sensors for wearable devices.

An alternative deposition technique is spray-printing, in which polymer solutions can be sprayed onto unrestricted surfaces with any roughness, textures and form factor which is a challenge for other printing techniques (see **Figure 2f**). The formation of active layers is achieved by sprayed droplets deposition along with a high-speed carrier gas, allowing lower material losses during deposition and easy scalability for large-area electronics. Among the spray deposition techniques, air brush spray method is low-cost and allows high throughput for industrial production. However, the resolution is also limited to tens of micrometers due to the requisite hard mask, and the quality of the sprayed films is easily influenced by the compositions of precursor solution and drying temperature of substrate^[87]. Using this facile air brush spray deposition technique, OECTs with excellent long-term stability on biodegradable ethyl-cellulose, nonplanar flexible natural leaf, wearable commercial adhesive bandages and polystyrene-*block*-poly(ethylene-ran-butylene)-*block*-polystyrene (SEBS) elastomer-based substrates were reported by our group (see **Figure 2f**).^[58] The device performance and solution viscosity were tuned by the addition of ionic liquid in PEDOT:PSS.^[3] The channel ($W \times L \times d = 1000 \times 100 \times (0.1-0.4) \mu\text{m}^3$) was well-defined through the use of a stencil mask. A transconductance of 3.7-7.4 mS, on/off ratio of $\sim 5 \times 10^2$ and switching time of 179 μs were demonstrated for the OECTs on these substrates. An inverter consisting of PEDOT:PSS-based OECT (working in depletion-mode) and glycolated thiophene polymer (g2T-T)-based OECT (working in

accumulation-mode) with both channels prepared by spray deposition exhibited a high gain of ~ 18 . Thus, spray printing is a facile technique to fabricate basic elements of a circuit, and is easily scalable for large area electronics. An all air brush spray-printed OECT was also reported by Hsu et al. by printing carbon nanotube (CNT) films as electrodes and PEDOT:PSS film as active channel layer on top of CNT films.^[88] Apart from the p-type semiconductors, an n-type poly(benzimidazobenzophenanthroline) (BBL) based OECT with channel dimensions of $W \times L \times d = 39000 \times 20 \times 0.18 \text{ } \mu\text{m}^3$ was also successfully prepared using the air brush spray printing method with a peak g_m of 9.7 mS.^[16]

Another promising alternative is the aerosol-jet printing (AJP) technique, of which the operating mechanism is very similar to general spray-printing process. In contrast, AJP is a contactless direct write approach that enables printing of the active materials on both 2D and 3D substrates. One advantage is that the semiconductor, gel electrolyte and gate electrode are all printable with well-defined patterns by AJP technique. Nevertheless, it is also an open challenge to control the edge profile instabilities of printed lines, since AJP process is highly affected by several parameters including the overspray issue and ink/substrate compatibility, thus causing a reproducibility problem.^[89] This technique has been widely explored to prepare the active layers for electrolyte-gated transistors (EGTs) and OECTs.^[89-93] Attractive features of AJP-based transistors such as high output conductance, low operation voltage and fast switching times have been demonstrated, thus exhibiting promising application in printed circuits. More recently, Giuseppe et al. demonstrated a fully printable OECT with silver as the metal electrode and PEDOT:PSS as the active material using AJP technique.^[89] The fabricated AJP-based OECTs with channel dimensions of $W \times L \times d = 200 \times 200 \times 0.2 \text{ } \mu\text{m}^3$ exhibited a peak g_m of 0.5 mS and on/off ratio of 10. They also reported a systematic investigation of AJP technique applied to PEDOT:PSS based films on flexible substrate, and provide a guide recipe to obtain high quality printed PEDOT:PSS films.

Apart from the screen printing and spray printing techniques, the inkjet printing method has attracted substantial attention to fabricate high performance OECTs since it offers a contactless, stencil-free, and material-efficient approach to pattern the active layer.^[94-96] Inkjet printing offers the digital fabrication of customizable designs at atmospheric conditions, that enable to achieve a variety of patterns as required. The

channel thickness ranging from 100 nm to 2300 nm and minimal line resolution as low as 10 μm of the printed PEDOT:PSS film can be easily controlled on substrates with very small to large size areas, therefore this technique is ideal for small and medium production volumes.^[75, 76, 97, 98] For instance, a peak g_m of 10 mS and on/off ratio of 10^3 were achieved for the inkjet-printed OECTs on plastic polyethylene foils with channel dimensions of $W \times L \times d = 1000 \times 500 \times 2.095 \text{ } \mu\text{m}^3$.^[97] Furthermore, it provides possibilities to print active layers on top of any 2D or 3D substrate. For instance, as presented in **Figure 2h**, advancements in stretchable OECTs with well-patterned channels have been made on biomimicking PDMS substrate by the development of inkjet printing techniques.^[50] The stretchable OECTs could be attached on different regions of human hands and exhibited stable performance even under 30% strain state. Further, the inkjet printing technique enables fabrication of OECTs on eco-friendly paper substrates, which offers an inexpensive technique to construct disposable biosensors (see **Figure 2i-j**).^[75, 76, 98] However, the low-viscosity inks tend to be adsorbed by the rough and porous paper substrates, which results in spreading forms with wavy boundaries and an inadequate electrical property.^[94] Some strategies have been developed including: (1) modifying paper substrate with a hydrophobic layer^[99] or precoating a smoothing underlayer^[100]; (2) employing photopaper^[101] with a hydrophobic surface or nanocellulose paper^[102] with lower surface roughness and dense nanofibrous network structure to restrain ink permeation and overspreading. Another challenge for inkjet printing lies in the complex drying behavior of the printed materials, called the coffee-ring effect^[103]- a non-uniform ring-like profile is formed during the transport of solute from the center to the edge upon the drying of the printed ink on the substrate surface, thus negatively affecting the morphology and performance of patterned devices. To obtain desired morphology and homogeneity of active layer, several approaches have been proposed to minimize the coffee ring effect by investigating and optimizing the drying process in inkjet printing.^[74, 104] One way is to maintain a temperature gradient between the ink and the substrate surface. By increasing the substrate temperature (25-60 °C higher than the ink), the solvent evaporation can be stimulated, which reduces the in-flight droplet

diameter and suppress the hydrodynamical spreading to form the coffee ring.^[105, 106] Another way is the addition of high-boiling solvent, which can induce the surface tension gradient of the ink and retard the solvent evaporation at the contact edges.^[103] Furthermore, optimizing the ink concentration and incorporation of additive agents (e.g., nonionic small molecule surfactant, Triton X-100) can regulate the rheological and wettability properties of the ink formulations.^[107, 108]

Nanoimprint lithography (NIL) is a molding process that uses a stamp to replicate features in nanoscale over large areas with long-range order.^[109, 110] NIL offers an effective and low-cost technique to replicate nanoscale features as it does not require expensive optics and multiple photolithographic steps. It can replicate nanoscale features below 10 nm on organic polymers and is therefore a potential approach to overcome the practical limits of photolithography.^[111] As an emerging high-resolution patterning method, it basically requires a stamp, an imprint fluid, and a solid substrate. When a stamp containing surface relief patterns is pressed into the imprint fluid, a replicated patterning with a 1:1 ratio between features on stamp and substrate can be achieved due to the direct mechanical contact. After removing the stamp, an imprinted polymer pattern is left on the substrate. There are two common types including thermal and UV methods for the curing or hardening process. In thermal NIL, it makes use of thermoplastic polymer resist which needs to be heated above its glass transition temperature (T_g) to soften the polymer during the mold filling process and cooled below its T_g before demolding process. The application of thermal nanoimprint lithography is therefore limited on flexible substrates due to the high T_g (> 100 °C). On the other hand, UV nanoimprint lithography uses a UV source to solidify the resist and is performed at room temperature and low pressure.

Figure 3. Nanoimprint lithography of OECTs. (a) Schematic of the fabrication steps for P3HT-based side gate transistor using UV NIL process. (b) Optical microscopy and SEM images of the imprinted patterns of electrodes, channel and semiconductor (SC) areas for transistors.^[61] Copyright 2016, Wiley-VCH. (c) Schematics of the NIL patterning of PEDOT:PSS. Right image is the OECT geometry structured by NIL, the width W of the source-drain line and the gap G between the gate and source-drain line.^[62] Copyright 2013 SID.

To the best of our knowledge, OECTs based on NIL have only been reported by UV photocuring process. **Figure 3a** shows the fabrication flow of a typical UV NIL process and also the side-gate structure of P3HT-based transistor.^[61] A PDMS stamp was used to define the capillary channels and electrodes area, in which a short gate distance ($\sim 4.6 \mu\text{m}$), channel length ($\sim 1.5 \mu\text{m}$) and width ($\sim 30 \mu\text{m}$) were achieved (see **Figure 3b**). This technique for printed transistors enables a self-aligning and high-resolution patterning of active organic layers. PEDOT:PSS-based OECT was also fabricated by UV NIL technique using a silicon (Si) stamp as follows: i) press the Si stamp into the resist and remove the stamp after UV-curing, ii) leaving the passivated mask for etching process, iii) oxygen plasma etching and remove the resist, iv) inkjet printing of electrolyte on the sample (see **Figure 3c**).^[62, 112] A gap of $\sim 2 \mu\text{m}$ between side-gate and channel and channel width of $13 \mu\text{m}$ were obtained, and the reduced device geometry was demonstrated to improve the on/off ratio of $\sim 10^4$ and

switching speed of 30 ms for NIL-based OECTs, thus suitable for applications in logic circuits. These reports indicate that NIL plays a complementary role with photolithography for the fabrication of OECTs with high-resolution patterning in roll-to-roll manufacturing process. Much of this potential remains underexplored due to the following challenges for employing NIL in one's research: 1) the patterns are prone to defects and fragile which may induce the collapse of the replica, especially for high-aspect-ratio features, 2) the issue of polymer shrinkage during curing which induce pattern placement errors and leads to low reliability, and 3) the high fabrication cost and careful maintenance of nanoscale stamps.

2.2 Laser-based patterning technique

Figure 4. Laser-based patterning of OECT structures. (a) Schematic of HAZ generation following the laser-based patterning.^[113] Copyright 2015, Wiley-VCH. (b) The OECT array through laser-based patterning i) Computer-aided design image of the OECT array, ii) The channel area after laser ablation, iii) Printing PEDOT:PSS on the channel area, iv) After

baking of the printed PEDOT:PSS.^[59] Copyright 2012, Wiley-VCH. (c) Schematic of the fabrication process. 1. Stacking structure. 2. Laser ablation to open the channel with minimal damage to substrate. 3. Dispersion of PEDOT:PSS. 4. Baking to fabricate the transistor. 5. Operating the OECT. Image on the right side shows selection of laser power for optimal and complete cutting through the electrode without damage to the substrate.^[60] Copyright 2020, IOP Publishing Ltd.

Laser-based patterning technique is based on the ablation of polymer surface caused by the exposure to laser pulse energy. In general, infrared lasers are commonly used for polymer ablation by breaking the chemical bonds and removing the surface molecules by the interaction of photothermal effect and thermal conductivity of polymer, thus patterning the channel or electrodes area for OECTs. In this technique, the laser processing parameters, consisting of pulse width, wavelength, and laser power, are highly relevant to laser-based patterning performance.^[114] Particularly, laser pulse width has to be tuned critically due to the generation of heat affected zone (HAZ). As shown in **Figure 4a**, although HAZ is not exposed directly to the laser beam, the influence by the absorbed heat from the laser etching process leads to undesirable change in the interface of polymer-substrate.^[113] The thermal diffusivity of both polymer and substrate material has a great impact on the HAZ size, which basically determines the resolution of laser-based patterns. Thus, it is desirable to select suitable laser parameters to minimize the HAZ, according to the thermal diffusivity of polymer and substrate of interest.

Laser-based patterning of conjugated polymers for OECTs is a relatively difficult process due to the morphological changes brought about by the HAZ in conjugated polymers. Unoptimized laser patterning of conjugated polymers can result in distortion of the polymer-substrate interfaces and creation of pores in unexpected areas. Although the developed design of energy optimization can be used to minimize distortion of conjugated polymers^[114, 115], direct laser exposure on conjugated polymer films is not suggested as a good approach for the fabrication of high quality patterns, as it adversely affects the interface between electrode and conjugated polymers. Some efforts into laser-based patterning of OECTs have been made to ablate insulating polymer and open the channel area on flexible substrates. A combination of

screen-printing of conducting electrodes and dielectric layers, inkjet-printing of conjugated polymers, and laser-ablation patterning of channels was reported to fabricate fully isolated OECTs (see **Figure 4b**).^[59] The hybrid manufacturing approach allowed for the architecture of vertical OECT devices with on/off ratio of ~600 and switching times of ~100 ms. An OECTs array with 17×50 pixels was fabricated on A4 size (297 mm × 210 mm) flexible sheet substrates, which demonstrated a high potential in industrial application. Moreover, a multi-stacked structure was ablated with minimal damage to the hydrophobic insulating substrate by optimizing the laser power from 0.1 W to 5.0 W, as shown in **Figure 4c**.^[60] The laser patterning method provides self-alignment of source/drain contacts, channel and insulation, and allows for conjugated polymers to be physically dragged across surface and wick into laser cut OECT channels. Using the laser system, a channel length of 25 μm (smallest cut width), channel width of 700 μm and thickness of 115.5 nm were defined for OECTs, which eliminated the overlap between the channel and S/D contacts. A g_m of 2.82 mS and good reproducibility were achieved for laser-patterned OECTs. This approach enables the fabrication of free-standing and through-hole OECT channel, thus presents unique strategy for patterning channels that may be limited with traditional patterning methods.

2.3 Fiber-based OECT

Fiber-based structures are highly desirable for wearable electronics that are expected to be light-weight, space-efficient, soft, flexible, and conformable.^[116] The fiber structure can be integrated into stretchable, bendable or twistable electronic devices with various functionalities and configured into threadlike fiber devices that can be easily woven together into textiles. For wearable fabric devices, the porous network geometry helps to keep human skin air-permeable and comfortable, whereas conventional planar wearable devices prevent sweat evaporation from the surface of the human skin.^[117] Recently, various fiber structures have been manufactured by advanced nanotechnologies, making it achievable to build electronic devices directly on the surface or inside of single fiber with a typical micron-sized thickness.^[118-121]

Fibers or textiles are also attractive carriers for OECTs, with great promising applications in wearable electronics such as non-invasive and continuous monitoring of physiological parameters. Unlike FETs that require well defined insulator layer and small feature sizes^[122, 123], OECTs present relative ease of manufacture without any narrow and critical dimensions for low voltage operation. As shown in **Figure 5a**, non-planar cross-junction configuration is an attractive design for OECTs on fibers, where two fibers act as substrates for channel and gate films, with an electrolyte bridging the junction between the fibers.^[53, 124-126] The active part of the channel area comprises the entire cylindrical film that is in contact with the electrolyte, which presents similar switching characteristics to planar OECTs. The mechanical toughness of conjugated polymers inherently ensures strong adhesion along the fibers, and enable them to withstand mechanical stretch and scratch during weaving and knitting process, thus allowing for the manufacture of flexible OECT fabrics and textiles (see **Figure 5b**).^[124]

Figure 5. (a) Operation mechanism of fiber-based OECTs.^[127] Copyright 2018, Wiley-VCH. (b) The image and transfer characteristics of fiber-based OECTs constructed at the junction of

two PEDOT:PSS-coated polyamide monofilaments with 100 μm diameter.^[124] Copyright 2007, Springer Nature. (c) The design of a core-shell conductive nylon fiber coating with PEDOT:PSS. The OECT device structure constructed by fiber-based channel and gate. Flexible fiber and textile devices after embedding OECTs by a conventional weaving machine showed a stable performance at different bending states.^[128] Copyright 2018, Wiley-VCH. (d) The mechanism for PEDOT:PSS microfiber formation in an acidic coagulation bath, where the PEDOT:PSS microfiber is coagulated, whereas excess PSS chains are dissolved by the sulfuric acid solution. The single-strand channel dimension-independent PEDOT:PSS microfiber OECT can be used as sweat sensor.^[129] Copyright 2018, Springer Nature.

Several approaches have been explored to fabricate OECTs in fiber or textile architectures using individual or multiple fibers. At first, Kim et al. developed a simple wet-spinning process to fabricate single-strand PEDOT:PSS conducting microfiber and assembled substrate-free microfiber-based OECT devices for wearable sweat sensors.^[129] Another PEDOT:PSS hydrogel fiber with the diameter of 400 μm was fabricated by facile syringe-injection into a cavity and the hydrogel fiber-based OECT with channel length of 10 μm presented a maximum g_m of ~ 1.4 mS at $V_{GS} = -0.2$ V.^[130] For the coating of conducting polymers on fibers substrate, in-situ polymerization of organic conducting polymers on fibers is a facile approach by the consecutive growth of monomers of conducting polymers in an ice bath. For example, a polypyrrole (PPy) layer was successfully polymerized on polyamide 6 (PA6) fibers and the treatment of poly(vinyl alcohol-co-ethylene) (PVA-co-PE) or reduced graphene oxide (rGO) on the surface of PA6 fiber helped to form a large-area of intertwined PPy nanowires network, which could improve the ion adsorption, charge transfer, and carrier transport.^[63, 131, 132] The assembled PPy-based OECT showed a typical on/off ratio of 10^2 , switching speed of 0.34 s and excellent bending stability within 500 cycles.

Another approach is vapor-deposited conjugated polymer film on monofilament fibers. Reactive vapor deposition (RVD) was used to conformally coat nylon fibers with a stably doped conducting polymer film (PEDOT-Cl). A fixed-channel-length, parallel OECT was then embroidered by straight-stitching the conducting fiber (channel) and a stainless steel thread (gate) onto a hydrophobic silk backing fabric.^[127] Using this simple embroidery approach, large-area OECT arrays were rapidly created

onto a 2×2 inch silk fabric, and the output current for each individual device remained quite stable while the entire front face of the fabric devices was covered in a liquid electrolyte solution. Another PEDOT-based OECT on microporous expanded polytetrafluoroethylene (ePTFE) membrane has also been reported by vapor-deposition, which can act as “breathable” gas sensors.^[113]

In addition, dip-coating method can be used to fabricate conducting fibers by simply immersing fibers into the conducting polymer solution and pulling it out at a uniform speed. For instance, natural cotton fibers soaked into PEDOT:PSS solution can be used as a channel of OECT for liquid electrolyte saline sensing.^[52] To improve the bending stability of fibrous OECTs, a multi-layer structure on Nylon fiber was designed, that was first coated with metal electrode layer, followed by a highly conductive PEDOT:PSS channel layer and parylene passivated layer as shown in **Figure 5c**.^[128] The conducting fiber with a Ti/Pt/Nylon fiber as gate was used to assemble a fiber-based OECT, which can be woven as both warp and weft with cotton yarns by using a conventional weaving machine, resulting in flexible and stretchable fabric devices for glucose sensing with much more stable signals than planar devices owing to the capillary effect in fabrics. Other than the requirement of supporting fibers, PEDOT:PSS microfibers with controllable diameter (10-100 μm) and conductivity (300-1000 S/cm) can also be prepared by a simple wet-spinning process using a sulfuric acid-based coagulation medium (see **Figure 5d**).^[129] The microfiber-based substrate-free OECTs exhibited an on/off ratio of 10^2 for gates voltages from -0.4 V to 0.4 V. A novel two-terminal single-strand OECT-based sweat sensor was fabricated and mounted on human skin for wearable healthcare monitoring applications. Thus, fabrication of OECTs in fiber form can serve as a good approach to realise noninvasive wearable sensors and present unique advantages in the analysis of external biological fluids (e.g. sweat, saliva and urine).^[83]

3. Applications of OECTs

3.1 Biological sensors

Figure 6. (a) A flexible integrated system comprising miniaturized ion-selective OECTs with TFTs. The KCl concentration was outputted as a drain current output for a K^+ selective OECT.^[65] Copyright 2018, Wiley-VCH. (b) Schematic diagram of label-free DNA sensor based on OECTs, which was integrated with a flexible microfluidic system. The surface of gate electrode was modified and immobilized with single-stranded DNA probes.^[133] Copyright 2011, Wiley-VCH. (c) Wearable OECT-based noninvasive sweat cortisol sensor. The device was functionalized with molecularly selective nanoporous membrane biorecognition based on molecularly imprinted polymers.^[134] Copyright 2018, American Association for the Advancement of Science. (d) Schematic diagram of highly selective enzyme biosensors based on OECTs. Potential drops between the gate and channel of the OECT in the presence of uric acid in electrolyte.^[135] Copyright 2014, Wiley-VCH.

The rise of bioelectronics, which combines the areas of electronics and biology, inspires enormous attention to develop novel biomedical devices for disease diagnosis and therapy. Conjugated polymers can offer an ideal bio-interface between the sensor element and the biological environment due to their intrinsic biocompatibility, and mechanical matching. In particular, OECTs hold a key feature for application in

bioelectronics, which can provide local amplification and thus as a transducer of biological events with high fidelity. Gate modification is an excellent technique for transducing various biological signals by controlling the electrochemical potential at the gate electrode.^[136, 137] Furthermore, OECTs offer a convenient approach to amplify fast-scan cyclic voltammetry, allowing for rapid and low-cost instrumentation compared to commercially available tools. Several advantages such as high selectivity, high sensitivity and low limit of detection can be achieved to detect targeted biomarker in the presence of interfering elements using OECTs, which is comparable or better than those acquired using sophisticated electrochemistry techniques such as differential pulse voltammetry.^[138] For now, preliminary studies have revealed the huge potential of OECTs for the sensing of various ions, biomolecules (e.g. enzymatic sensing, cortisol sensing, nucleotide sensing, immune sensing, glucose sensing, metabolite detection) and integrality of cell or barrier tissue (**Table 1**).

Generally, the ion concentration in electrolyte has an influence on the OECT characteristics.^[11, 97] The change of ion concentration in the electrolyte can be delineated by the variation of channel current of an OECT. Thus, on the basis of this principle, there is a unique advantage for OECTs to sense ions in the electrolyte, which is of great importance in healthcare diagnostics. For now, various ion sensors based on OECTs have been successfully developed.^[5, 139, 140] Chan et al. systematically investigated the ion-sensitive properties of an PEDOT:PSS-based OECT in aqueous solution with different cations, including H^+ , K^+ , Na^+ , Ca^{2+} , Al^{3+} .^[141] The results demonstrated that the transfer behaviors of OECTs shift to lower gate voltage horizontally while increasing the cation concentration in electrolyte. However, certain factors such as the choice of organic semiconductor materials, the design of device structure, and the use of appropriate gate electrode are critical for ionic sensitivity. Besides, another important challenge for use of OECTs in practical applications such as environmental monitoring, water quality detection and biomedical research is the ionic selectivity of OECTs. Ion-selective electrodes have been used to implement selective ion concentration measurements, where the potential drops across an ion-selective membrane (ISM) located on either side of ISM

could differentiate different ion sources in electrolyte.^[35, 65] For example, Sessolo et al. integrated a K⁺-selective membrane with a PEDOT:PSS-based OECT to demonstrate the ability of monitoring K⁺ with a low limit of detection (1.5×10^{-5} M) and in a reversible manner.^[35] The sensitivity in the 10^{-4} - 10^{-1} M K⁺ concentration range approached 50 $\mu\text{A dec}^{-1}$, which is 10 times higher than that of Na⁺ ions, thus proving the ion selectivity of ISM-based OECT. In a similar configuration as shown in **Figure 6a**, Street and co-workers reported a flexible miniaturized ion-selective OECT sensors array for multianalyte detection or spatial mapping of analytes.^[35, 65] The reported ISM consisted of an ionic salt of interest and an ionophore dissolved in a hydrophobic polymer membrane, which was deposited on top of the channel. When the ISM was exposed to a specific analyte, some of the cations diffused into the analyte and the hydrophobic anions were trapped in the ISM, generating a phase-boundary potential between the ISM and electrolyte, thus inducing the membrane potential's ionic concentration dependence of 61 mV dec^{-1} . Moreover, since the ionophore selectively solvated the ions on demand, it can eliminate the effect of other ion sources, making the membrane potential predominantly a function of the selected ionic concentration in the analyte.

The detection of nucleic acid has aroused great interest because of its extensive applications in gene expression monitoring and viral and bacterial identification. Some reported DNA sensors based on organic thin film transistors (OTFTs) showed a relatively low sensitivity to DNA, being unable to efficiently detect the presence of DNA in picomolar (pM) or even femtomolar (fM) range.^[142, 143] As shown in **Figure 6b**, an OECT-based DNA sensor was developed by Yan's group, has extended the detection limit to 10 pM.^[133] In this work, OECTs with PEDOT:PSS as the channel polymer were fabricated in a flexible microfluidic system. Single stranded (ss) DNA probes were modified on the surface of gate electrode. The transfer characterizations showed obvious gate voltage shifts after the hybridization of ssDNA with different concentrations of complementary DNA targets and no appreciable shift was recorded using non-complementary DNA species. The mechanism to sense DNA owes to the modulation of the effective gate surface potential induced by the immobilization and

hybridization of DNA molecules on gate surface and consequently, the shift in potential modulates the source-drain current.

Immunosensors are a class of biological sensors to recognize the interaction of antibodies with specific antigens. Generally, either antibodies or antigens can be the immobilized biorecognition molecule on the transducer, which placed in contact with the analyte solution containing specific antibody/antigen marker to detect the specific antibody-antigen interaction. There are two methods to functionalize OECTs as immunosensors, including immobilizing antibody or antigen molecules on surface of conducting polymer^[144-146] or gate electrode^[147-149]. As reported, OECT-based immunosensor was firstly developed in 2002, by immobilizing goat antirabbit anti-immunoglobulin G (anti-IgG) in PEDOT polymer matrix during polymerization.^[145] Antibody-antigen binding resulted in the backbone conformation and film conductivity change in PEDOT polymer, and a concentration of antigen rabbit IgG as low as 1×10^{-10} g/ml can be detected with a response time of 3 minutes. To improve the sensitivity of OECT-based immunosensor, Kim et al. designed a PEDOT:PSS OECT for the detection of prostate specific antigen/ α 1-antichymotrypsin (PSA-ACT).^[146] By immobilizing PSA polyclonal antibodies (PSA pAb) conjugating with gold nanoparticles (Au-NPs) on PEDOT:PSS surface, the limit of detection of PSA-ACT can be improved to 1 pg/ml from 4 ng/ml for OECT without Au-NPs. The improved sensitivity was attributed to the interaction of AuNPs-PSA pAb with PSA-ACT complex leading to a more negative charge on the surface and thus induced higher doping effect in PEDOT:PSS. For gate-modified OECT immunosensors, an additional potential can be generated during antibody-antigen binding on gate electrode to shift the effective gate voltage which will affect the dedoping of PEDOT:PSS. Recently, PEDOT:PSS-based OECTs with gate electrodes modified with antibodies (e.g., poly (3-aminophenylboronic acid) (PABA), human epidermal growth factor receptor 2 (HER2) and anti-human IgG), have been successfully used to identify cancer or normal cells, HER2 proteins and human IgG with ultra-low limit of detection (down to 10^{-14} g/ml), which is several orders of magnitude lower than that of reported electrochemical methods, owing to the inherent amplification function of

OECT.^[147-150] The progress of OECT-based immunosensors open a way for developing highly sensitive and low-cost biosensors for the detection of various antibodies or antigens in immunoassay technology.

Even though most OECT-based biosensors focused on the analyte detection in saline solutions, it is also possible for OECT-based biosensor to operate under other environmental conditions such as sensing (i) gas molecules in breath^[76], (ii) metabolites in body fluids including sweat^[134], saliva^[135], and (iii) integrity of barrier tissue in cell culture media^[151]. With the increasing requirements for mantic clinical diagnosis and personalized health care, an invasive manner in commercially available technologies to monitor physiological level mostly requires drawing blood from patients, which introduces pain, inconvenience, and possible microbial infection.^[152] In this regard, noninvasive biosensors offer a painless strategy for continuous and long-term monitoring for patients who need to be monitored routinely. Body fluids, such as sweat, saliva, and tears, include abundant biomarkers, which can provide distinct capability while extracted and analyzed in a noninvasive method, thus offer alternatives to blood to monitor physiological levels.^[153-155] However, due to the rich mixtures in the body fluids, achieving a reliable sensing with excellent sensitivity and selectivity for noninvasive biosensors still requires further exploration.

Cortisol, a steroid hormone, is a biomarker for physiological stress detection which presents an optimum level from 0.02 to 0.5 μM in sweat. The effect on stress plays a major part in causing or triggering major cardiovascular disease and stroke. Most reported stress sensors are based on physical sensing such as heart rate and temperature. However, the influence of surrounding conditions that are non-stress related can contribute to false positives in these sensors. Self-monitoring of cortisol in a dynamic and non-invasive manner, alternative to blood sample, such as in sweat is therefore in high demand. Yet, there is no reliable method of objectively and continuously measuring the "stress hormone" cortisol for point-of-care applications. A recently reported strategy for cortisol sensing introduced a molecularly imprinted polymer (MIP)-based selective nanoporous membrane into a PEDOT:PSS OECT.^[134] The planar Ag/AgCl gate electrode was functionalized with the acrylate-based

molecularly selective membrane as an electrochemical transducing layer for cortisol recognition as shown in **Figure 6c**. The SEBS elastomer substrate provided excellent flexibility and stretchability of the integrated wearable device. This wearable device offered many superiorities such as noninvasiveness, ease of accessibility and user comfort as compared to other traditional blood analysis for cortisol sensing, and has achieved a sensitive and selective detection for cortisol in the level of 10 nM-10.0 μ M. The sensing mechanism relies on the fact that the molecular membrane can be tuned to be ion-permeable or ion-impermeable in the absence and presence of the analyte respectively, due to the sealed and blocked membrane pores, which strongly modulates the conductivity of the OECT channel, thereby giving rise to a sensing event. A similar biosensor based on OECTs with gate electrodes modified with MIP film were developed and used for the chiral recognition of amino acid enantiomers (D/L-tryptophan and D/L-tyrosine) with a detection limit of 2 nM.^[156]

OECTs can also act as metabolite transducers for the detection of various enzymes, such as glucose, uric acid (UA), dopamine (DA), cholesterol, and lactic acid, which are vital for clinical diagnostics, health monitoring, food safety, environmental monitoring, etc.^[157-161] For the detection of metabolites, the sensing mechanism owes to the selective interaction of a redox enzyme with the metabolite molecules, which leads to the transfer of an electron to the gate of OECT. The channel current change depends on the concentration of the metabolite in electrolyte. As shown in **Figure 6d**, a flexible OECT-based enzymatic sensor was developed by modifying the gate electrode with suitable enzymes (e.g. PANI/Nafion-graphene/Pt) and graphene oxide (GO), which exhibited sensitive and selective response to UA, cholesterol, and glucose with low detection limits ($\sim$ 10 nM, $\sim$ 100 nM and $\sim$ 30 nM, respectively).^[135] The levels of biomarkers in saliva were also detected successfully as a non-invasive biosensor. In addition, the combination of OECTs with a fully filament-integrated fabric paves the way for novel wearable non-invasive biosensing in sweat or urine, whereas combination with textiles allows for the design of multi-analyte sensing platforms and remarkable mechanical compatibility with human body. The fibrous device structure improved the specific surface area and the conductivity of the active

filament. Recently, OECT-based biosensors with functionalized gates and weaved onto the nylon fibers or cotton yarns, were designed for the detection of DA, UA and glucose, which showed low detection limit (down to ~ 1 nM) and outstanding repeatability compared to some planar OECT biosensors.^[63, 128] Interestingly, an all-polymer biofuel cell was demonstrated to generate enough power to operate a P-90-based OECT for glucose detection from bodily fluids, by converting the chemical energy of glucose and oxygen into electrical power.^[64]

Figure 7. (a-b) Schematic representation of monitoring of cell layer coverage using OECTs. With increased barrier functionality, ionic transport into the film is restricted, whereas “leaky” cells allow certain degree of ionic transport. (c) The comparison of normalized g_m of different cells monitored using OECT biosensor.^[162] Copyright 2015, Royal Society of Chemistry. MDCK-I is short for Madin Darby Canine kidney epithelial cells with “tight” barrier tissue cells; Caco-2 is a human colon cancer derived cell; Hela cell is human cervical cancer derived cell; Hek cell is human embryonic kidney cell. (d) Several challenges for future biological applications including suitable biocompatible material choice for electrodes and surface biofunctionalization to further improve the selectivity.

OECTs have also been used as alternative technology to detect the presence of live cells and sense the integrity of barrier tissue. When a live cell appears around the channel of OECTs, its membrane is polarized, giving rise to an additional potential. The cell’s polarization generates a potential shift to reduce the effective gate voltage and affects the electrochemical doping/de-doping in channel. Barrier tissue, comprising of epithelial or endothelial cells, can modulate paracellular ion flux

between different bodily compartments.^[163] However, a non-integrity of barrier tissue such as those caused by toxic events mostly renders a dysregulation, resulting in disease or dysfunction. Unlike current optical techniques or electrical impedance measurement for the investigation of in vitro cell cultures, OECTs have been demonstrated to sense the presence of a cell monolayer directly grown on the active layer interface, where cell layer integrity and health directly correlates to the measured electrical signals.^[151] As shown in **Figure 7a-b**, the g_m of OECT is expected to fall with increasing cell coverage and increasing barrier functionality, which is especially useful in determining cell confluency. Other OECT devices were designed to monitor the cellular vitality of various cell lines in real-time, and showed differentiable response to leaky-barrier (NIH-3T3, mouse embryonic fibroblast) and strong-barrier (CaCo-2, human colorectal epithelial; MDCK-I, Madin Darby Canine kidney epithelial cells) cell lines growth and detachment by selecting suitable specific channel dimensions (see **Figure 7c**).^[162, 164] Others also used PEDOT:PSS-based OECT lactate sensors to monitor the basal metabolic activity and predict metastatic potential of tumor cells, and P3HT-based OECT bacterial sensor to detect the membrane disruption by possible antimicrobial compounds and assess the potency of such compounds.^[66, 165] Magnus et al. reported a hydrogel-like and long-range conducting PEDOT:S:H wire grew along the xylem channels of a stem of garden rose with surprisingly high conductivity (>0.1 S/cm).^[56] The developed in situ OECT devices utilizing surrounded cellular domains including confined electrolytic compartments, demonstrated the mechanism of ion exchange with the extracellular medium and charge compensation at the gate electrode, making it an OECT-based electronic plant for feedback-regulated control of plant physiology. However, some significant challenges for future biological applications also exist, such as the fabrication of high density transistors at biological scale that are biocompatible, and minimize scarring in in vivo applications^[166], and suitable biocompatible material choice for electrodes and surface biofunctionalization to further improve the sensitivity, selectivity and stability (**Figure 7d**).

Table 1. Summary of OECT-based biological sensors

Type	Analyte	Device structure	Active materials	Detection range	References
Ions	K ⁺	PVC-ISM	PEDOT:PSS	15 μ M – 100 mM	[35]
	K ⁺	ISM	PEDOT:PSS	1 μ M – 100 mM	[65]
	Na ⁺ , K ⁺	ISM-free	p(T15c5- <i>ran</i> -EDOT)	10 μ M – 1 M	[167]
	Cu ²⁺	Vertical	Polycarbazole	2.5 μ M – 100 μ M	[168]
	Ca ²⁺ , NH ₄ ⁺	ISM	PEDOT:PSS	10 μ M – 100 mM	[155]
	Ag ⁺ , Ca ²⁺	Vertical	PEDOT:PSS	100 μ M – 100 mM	[140]
Hormone	Dopamine	Weaving-fiber -based	PPy	1 nM – 1 μ M	[63]
	Adrenaline	Cotton-fiber-based	PEDOT:PSS	1 μ M – 1 mM	[169]
	Dopamine	Planar	PEDOT:PSS	1 μ M – 4 mM	[170]
	Cortisol	Wearable OECT patch	PEDOT:PSS	10 nM – 5 mM	[134]
Metabolite	Lactate	Planar	PEDOT:PSS	10 μ M – 300 μ M	[66]
	Lactate	Solid OECT	PEDOT:PSS	300 μ M – 100 mM	[171]
	Lactate	Planar	P-90	10 μ M – 10 mM	[157]
	Uric acid	Flexible Planar	PEDOT:PSS	300 nM – 500 μ M	[135]
	Amino acid	Vertical	PEDOT:PSS	2 nM – 10 μ M	[156]
Nucleotide	DNA	Vertical	PEDOT:PSS	0.1 nM – 12.5 nM	[172]
	DNA	Flexible planar	PEDOT:PSS	10 pM – 5 μ M	[133]
	RNA	Vertical	PEDOT:PSS	2 pM – 20 nM	[173]
Antibody- antigen	Antigen IgG	Unknown	PEDOT	10 ⁻¹⁰ –10 ⁻⁷ g/ml	[145]
	PSA-ACT	Planar	PEDOT:PSS	10 ⁻¹² –10 ⁻⁷ g/ml	[146]
	Human IgG	Planar	PEDOT:PSS	10 ⁻¹⁸ –10 ⁻¹¹ M	[147]
	HER2	Vertical	PEDOT:PSS	10 ⁻¹⁴ –10 ⁻⁷ g/ml	[148]
Enzyme	Glucose	Vertical	PEDOT:PSS	5 nM – 500 μ M	[174]
	Glucose	Weaving-fiber -based	PEDOT:PSS	30 nM – 3 mM	[128]
	Glucose	Planar fluidic system	PEDOT:PSS	10 mM	[175]
	Glucose	Planar	P-90	10 nM – 20 mM	[64]
	Glucose	Skin-attachable OECT	PEDOT:PSS	1 μ M – 100 mM	[176]
	Alkaline phosphatase	Vertical	PANI	1–4 nM	[177]
Cell	Bacterial membrane disruption	Planar	P3HT	1–50 μ M	[165]
	Cell growth	Planar	PEDOT:PSS	–	[151]
	Cell vitality	Vertical	PEDOT:PSS	–	[164]

Note: Vertical device structure means the OECT devices are placed in a beaker filled with electrolyte and individual gate electrode is immersed in the electrolyte.

3.2 Logic Circuits

Logic gates are the basic building blocks of any digital system, which includes AND, OR, XOR, NOT, NAND, NOR and XNOR gates. Every terminal of these gates is in one of the two binary conditions, false (0) and true (1), which are represented by different voltage levels. Logic circuits are able to carry out a series of logic actions such as controls for displays, computer operation, security systems, industrial control and internet-of-things. All logic actions, however complicated, can be analyzed into several simple actions, by connecting the inputs of these basic logic gates and outputting the arithmetic action. Digital circuits such as shift registers, encoders, decoders, multiplexers, and demultiplexers have been developed to transfer serial or binary-coded signals into exclusive output terminals, which change the carrying way of digital codes.^[178-180] OFETs have been explored in various integrated logic circuits at frequencies ranging from 10^3 to 10^6 Hz^[181, 182], allowing relatively swift signal processing, however the operating voltage used to drive the logic circuits are typically beyond 10 V.^[183-185] The high g_m , on/off ratio and low operating voltage have attracted great attentions for researchers to integrate OECTs in logic circuits. However, the primary drawback of OECTs is the relatively slow switching behavior, owing to the ionic diffusion effect in OECT configuration. Nonetheless, OECTs also offer many promising opportunities, for example, electrochromic displays due to the colour-switching phenomenon of conjugated polymers and security features on packages as the system-on-a-sheet label.^[186] Berggren's group has printed the PEDOT:PSS-based active-matrix electrochromic display on paper using a standard label printing press.^[55, 187] To realize OECT-based digital circuits, many attempts have been reported to apply OECTs in inverters, NAND, oscillators, signal amplifier and wheatstone bridge logic circuits, etc.^[82, 188, 189] Up to now, various technologies have been developed to manufacture different large-area and high-volume OECT-based logic circuits, including screen-printing^[46, 79, 81, 86], photolithographic patterning^[190], nanoimprinting^[61, 112], inkjet printing^[75, 95], aerosol-jet-printing^[89], cut-and-stick method^[191], etc.

Figure 8. Logic circuits based on OEETs. (a-b) Circuit diagrams and optical images of an inverter and a two-input NAND gate based on all-printed OEETs. The supply voltage V_{DD} and V_{SS} were +5 V and -5 V, respectively. (c) Monolithic integration of the decoder and electrochromic display. All-printed OEETs located between the decoder and display. The digit '4' is visible on the display after addressing the decoder with binary input sequence [0,1,0,0].^[46] Copyright 2019, Springer Nature. (d) Circuit diagram and optical image of OEET-based signal amplifier. Scale bar, 20 μm .^[69] Copyright 2019, American Association for the Advancement of Science. (e) Circuit diagram of the Wheatstone bridge sensor circuit. The load resistors are on the drain side of the OEETs. Right image is the chronopotentiometric recording upon pulsing the gate of each OEET for different values of the drain load resistor.^[66] Copyright 2017, Wiley-VCH.

Inverter and NAND gate are the most basic elements in a circuit and are commonly used for the construction of combinational and sequential logic circuits. Woven OEET inverters and multiplexer has also been developed on fibres in 2007, which open an avenue for large-area OEET-based micro-electronics circuits.^[124] Recently, ultrathin conformable OEET-based NAND and NOR digital logic gates were fabricated on 2- μm -thick parylene-C supports, that could accurately perform the corresponding arithmetic.^[69] Furthermore, Berggren's group reported OEET-based inverters and two-input NAND gates by screen printing technology as shown in

Figure 8a-b, in which the output voltage levels, upon switching, rely on voltage division of a resistor ladder (R_1 , R_2 , R_3).^[46] They also built a binary-coded decimal 4-to-7 decoders and 7-bit shift registers, including more than 100 all-printed OECTs, thus minimizing the number of terminals required to drive electrochromic displays (ECDs). The OECTs were located between the decoders and ECDs, and enabled to drive the display components upon receiving the correct input logic voltage levels from the decoder circuit (**Figure 8c**). The OECTs used as drivers for display was also demonstrated by controlling the light emission in traditional LEDs, in which actual LED addressing was implemented by an OECT decoder.^[81] The monolithic integration of OECT-based display driver circuits and displays presents a promising application in smart labels within packaging and Internet of Things (IoT).

Amplifying transducers have drawn great attention for clinical electrophysiology applications, in order to provide a first stage amplification of electrophysiological signals at the recording site, making the signal less sensitive to further noise collection. Based on this, OECT-based voltage amplifier combined with external medical electrodes was used to record electrocardiographic signals, and achieved much higher quality recordings upon operating the OECT in the saturation regime.^[190] In addition, a multistage amplification of physiological signals is required to ensure suitable gain and allow between-stage processing. As shown in **Figure 8d**, Khodagholy et al. designed cascaded OECTs to realize a multistage amplification and higher overall gain than a single OECT.^[69] A 25-mV-amplitude sine wave was input to the cascaded OECTs, which showed a four times increase in the amplification compared to a single OECT-based circuit. For biological sensing, a sensor circuit using reference-based conductance switching in OECT present sensitive and precise detection in highly interfering conditions, such as cell culture media. For example, a PEDOT:PSS-based signal-ON sensor circuit showed a 10-times increase in sensitivity to salt ions than that of single OECT sensor.^[192] Such sensor circuits connected two differently functionalized OECTs into a Wheatstone bridge layout, where one OECT acted as a reference and the other functionalized for lactate sensing (**Figure 8e**).^[66] The Wheatstone bridge sensor circuit enabled an intrinsic background subtraction, thus

allowing to eliminate any interference arising from other factors such as electro-oxidizable compounds and electrolyte evaporation.

3.3 Memory and Neuromorphic devices

Organic electronic materials exhibit attractive characteristics for neuromorphic computing, and largely motivate the fabrication of low-cost and biocompatible neuromorphic devices with low energy consumption and excellent tunability.^[193-198]

The ionic doping process in OECTs is analogous to large numbers of biological processes in human brain activity, including learning and memory behavior, information processing and signal transmission, which are modulated by ionic fluxes between neurons. In particular, electrochemical doping-based neuromorphic devices allow operation in a large conductivity range with more continuous states than organic memory devices. The electrochemical doping mechanism of OECTs also helps to enhance the coupling effect between ionic and electronic charge carriers, thus enabling efficient communication between electronics and biological systems.^[68, 199, 200]

Figure 9. Organic memory and neuromorphic devices based on OECTs. (a) Ionic doping process to modulate and retain the conductance states in OECTs for neuromorphic devices.^[196] Copyright 2018, Springer Nature. (b) Device structure and electronic states of a non-volatile organic neuromorphic device. Pre- and Postsynaptic layers are separated by an electrolyte layer transporting ions or protons.^[43] Copyright 2017, Springer Nature. (c) Schematic of biological neuronal network and an organic nanowire synaptic transistor that emulates the function of long-term synaptic plasticity of a biological synapse. The optical image is an array of 144 synaptic transistors fabricated on a 4-inch silicon wafer by the nanowire printing technique.^[67] Copyright 2016, American Association for the Advancement of Science. (d) Configuration of the 4x4 OECT array and principle of global input. The channels are in contact with a common electrolyte. In neural environment global parameters regulate the overall function of the brain.^[201] Copyright 2017, Springer Nature.

As shown in **Figure 9a**, for OECTs operating with depletion mode, the conducting channel polymer shows a high conducting state until ions from electrolyte are penetrated that induce the de-doping process of the conjugated polymer backbone.^[196] Those penetrative ions could return to the electrolyte while turning off

the gate voltage, causing the channel polymer to be reverted back to its conducting doped state. The reversibility of ionic movement between conducting polymer and electrolyte has been used to develop basic short-term (considered volatile) synaptic plasticity functions such as potentiation and depression effects in PEDOT:PSS-based OECTs. However, the self-discharge behavior of conducting polymer also leads to poor bistability, making fast operation and memory retention impossible. To attain long-term, non-volatile and bistable memory behavior of OECTs, Salleo's group modified the organic materials with polymeric rearrangement, PEDOT:PSS film partially reduced with poly(ethylenimine) (PEI), interfaced with a PEDOT:PSS presynaptic electrode via an electrolyte and postsynaptic electrode, as artificial synapse (**Figure 9b**).^[43] In their system, a positive presynaptic potential on gate can push cations to flow into the postsynaptic electrode through the electrolyte, thus decreasing the electronic conductivity of PEDOT:PSS/PEI. While enabling a continuous current by ion transport, the electrolyte also served as a barrier for electronic charge transport, retaining the conductance state after applying the presynaptic potential. The main virtue of this system is that the electrolyte barrier for state retention can be decoupled and the conductance state can be changed, allowing for ultralow switching voltage (~ 1 V) while maintaining a large number of non-volatile and reproducible states (>500). Another approach is modifying a ferroelectric polymer layer on the gate electrode to control the redox state of conducting polymer. As reported, the polarization switching behaviors of the ferroelectric polymer (PVDF-TrFE) can generate appropriate threshold voltage characteristics and bistability in electrochemical device that keep PEDOT in its highly conducting and fast operating regime.^[202, 203]

Despite the significant progress that has been made to emulate synaptic functions, current neuromorphic devices still consume orders of magnitude more energy than that of biological synapses (~ 10 fJ per synaptic event). To reduce the energy consumption of artificial synapses, it requires selection of suitable organic materials and design of devices such that the overall size of the organic devices is minimized. Tae-Woo Lee's group reported organic core-sheath-structured (PEO-P3HT) nanowire

artificial synapses with extremely low level of energy consumption (~ 1.23 fJ per synaptic event for individual nanowire device) (**Figure 9c**).^[67] In their system, the presynaptic spikes could provide an electrical field which helps ions from ionic gel to penetrate the PEO sheath or even the P3HT core and the ionic trapping/de-trapping processes are distinctly reversible. The artificial synapses facilitated the development of short-term and long-term plasticity, and could be constructed as highly integrated, large-scale (12×12 array) neuromorphic computing systems. The achievement opened new paths to integrating soft neuromorphic systems with nanometer feature size.

For OECTs, the common electrolyte can serve for single device or device array with large numbers of units, that allows us to design novel architectures for neuromorphic engineering. For example, the architectures comprise one channel and multiple gates or one gate and multiple channels.^[204] The state of the channel can be controlled by the applied signals to gate in a case of spatiotemporal coupling. George G. Malliaras and his co-workers demonstrated a behavior that is reminiscent of the global control of neurons in the brain using a 4×4 array of OECTs (**Figure 9d**).^[201] The common electrolyte established complex soft connections (without physical wiring) between individual OECT, and leveraged these connections to realize coincidence detection. This feature revealed that the electrolyte gating of OECT provides remarkable superiority to implement the networks of neuromorphic devices with higher complexity and soft connectivity.

Furthermore, neuro-inspired computational platforms made by OECTs interfaced with cellular systems^[205, 206], integrated ionic-based logic gates including inverters and NAND gates^[124, 188] and stretchable synaptic OECTs^[48] were also developed, which expand the functionality boundaries of the current neuromorphic devices. In general, the easily tailored features and continuously tunable conductance range reported in various organic materials and architectures for OECTs have demonstrated great perspective for neuromorphic computing applications.

3.4 Electrophysiological monitoring

Figure 10. (a) The signal amplitude associated with different electrophysiological measurements. (b) Recording of EOG signals by OECTs during eyeball movements in both horizontal and vertical directions.^[207] Copyright 2014, Wiley-VCH. (c) Recording of EEG signals by attaching OECTs on human scalp and the recorded time-frequency spectrogram during opening/closing eyes.^[69] Copyright 2019, American Association for the Advancement of Science. (d) Recording of ECG signals by attaching OECT array on the heart surface and the spatial distribution of the recorded ECG outputs.^[208, 209] Copyright 2018, American Association for the Advancement of Science; Copyright 2014, Wiley-VCH. (e) Recording of EMG signals by attaching OECT/OFET devices on a neuron concentrated area of muscle tissue of optogenetic mice with a blue laser continuous stimulation.^[42] Copyright 2016, Wiley-VCH.

Electrophysiological activity with high-quality signals of an organ or a tissue can provide valuable information about physiological and pathological functions of the brain and body for healthcare monitoring and better understanding of body functions/dysfunctions. A key element to record these signals is the electrodes that serve as ion to electron transducers and translate biological signal to electronic signals that the present technology can read. However, traditional rigid devices based on metal electrodes that are attached onto bodies to perform these recordings, suffer from an invasive problem and possible damage on organs and tissues.^[210] Soft electronic

devices with unique mechanical features not only can provide conformal contact with target surface of the body, but can also reduce the interfacial impedances and signal distortion caused by physical deformation of the device.^[211, 212] To improve the quality of electrophysiological signals, the integration of preamplifiers near the sensing electrodes is an effective means to realize the local signal amplification and minimize the influence of wiring noise.^[213, 214] Among the various active soft electronic devices, OECTs have vital advantages such as biocompatibility, high transconductance, low operating voltage, high signal-to-noise ratio and high temporal resolution, allowing them to be loaded on the target surface of organ or tissue directly and monitor electrophysiological signals even at a very low level of gate bias sensitively.

As reported, OECTs have shown tremendous application potential in measuring electrophysiological activities such as electrocardiography (ECG), electrooculography (EOG), electroencephalography (EEG) and electromyography (EMG). These potential amplitudes generated by body activities distribute in a range from tens of microvolts to tens of millivolts, which is the majority of the noninvasive signals in electrophysiology (**Figure 10a**). For example, EOG can be used to track eyeball movement for the analysis of drowsiness or sleepiness stages, and other applications such as visual system and cognitive linguistics.^[215-217] Malliaras's group used two OECTs with high transconductance (>1 mS at zero gate bias for OECT with channel dimensions of $W \times L \times d = 100 \times 100 \times 0.14 \text{ } \mu\text{m}^3$) to simultaneously monitor both horizontal and vertical eyeball movements as shown in **Figure 10b**.^[207] In general, the eye movement can generate a potential of $\sim 200 \text{ } \mu\text{V}$, which can serve as power supply (V_{GS}) to bias the gate. The recordings exhibited drain current changes in an equal or approximately equal amplitude while eyes moved left/right or up/down, proving the feasible application in EOG. They also used OECTs to record the amplitude, frequency and duration of spontaneous eye blinking, and demonstrated the prominent potential in monitoring drowsiness.

EEG offers a valuable technique for the research and diagnosis of epilepsy, depth of anesthesia, coma, brain death and other focal brain disorders. On account of the

measurement necessity to get high quality EEG signals, strong chemical adhesives to bond electrodes with surface of brain scalp or invasive inside brain to ensure good physical contact are imperative, which may lead to tissue injury and reaction of the immune system to the electrode.^[71, 218] Flexible OEECTs have been demonstrated with a highly conformal contact with the brain surface and ultrahigh amplification function of the neuronal signals to detect the low magnitude EEG signals ($\sim 10\text{-}100\ \mu\text{V}$).^[21, 33, 39] Khodagholy and his co-workers designed a flexible and compatible internal ion-gated OEECT with high transconductance, fast response speed and excellent conformability, which was used for local amplification to record high-quality brain neurophysiological activity.^[69] An additional layer of ionic conducting gel between the interface of skin and electrodes was used to achieve low enough electrochemical impedance and minimize the skin redness or irritation as shown in **Figure 10c**. A clear oscillation of $\sim 8\text{-}12\ \text{Hz}$ (α) neurophysiological activity over the occipital region during wakefulness with eye closure was captured from the left occipital region, which is in keeping with a posterior dominant rhythm. In addition, higher frequencies ($13\text{-}25\ \text{Hz}$, β) ($30\text{-}50\ \text{Hz}$, γ) for the co-occurrence of brain oscillations were also implemented to highlight the ability of OEECTs for various neural computations and information transfer between cortical areas. Malliaras et al. also fabricated ultrathin ($\sim 4\ \mu\text{m}$) flexible probes with PEDOT:PSS-coated Au electrodes and OEECTs encapsulated in Parylene C to withstand mechanical deformation and reduce scarring on rat's skin.^[166] By translating the small bioelectric voltage inputs into large drain current changes, the PEDOT:PSS OEECT device showed a superior signal-to-noise ratio for the monitoring of low amplitude signals (44 dB), and showed great potential for the detection of the epileptiform spiking activity in humans undergoing brain surgery, as well as the distinction between awake and unconscious state after anesthesia.^[39, 219]

ECG as the main technique to record cardiac activity ($\sim 1\ \text{mV}$), can provide useful information about the normal function or abnormalities of the heart. Biscarini and his co-workers developed conformable and biocompatible OEECTs on a fully resorbable bioscaffold (poly(L-lactide-co-glycolide), PLGA) for the ECG

recording.^[209] As shown in **Figure 10d**, the gate electrode was placed on the skin close to the heart, while the channel was placed on the forearm with constant applied voltages ($V_{GS} = +0.5$ V and $V_{DS} = -0.3$ V) relative to the ground potential of the body. Each heartbeat cycle induces a macroscopic ionic current wave that spreads throughout the body and leads to a small additional potential on skin which can be collected by gate electrode and largely modulate the drain current by the signal amplification of OECTs. The bio-degradability and bio-absorbability of the bioscaffold made it possible for a controlled timescale and free removal of the implanted electronic devices from the regenerated tissue in future applications. However, accurate ECG recordings on cyclic contraction and expansion during normal beating of the heart require a high degree of device flexibility and stretchability, which can be realized by grid patterns on ultrathin substrate.^[220-222] Someya's group fabricated ultrathin (< 3 μm), stretchable and transparent active OECT arrays for mapping the spatial-temporal distribution of ECGs on heart surfaces of a rat with multiplexing.^[40, 208] The OECT array with antithrombotic ability enabled a stable measurement over long periods even in an environment where bleeding occurs due to its antithrombotic feature. However, the direct electrolyte-gated operation for commonly used OECTs limits their ability to measure biosignals only when electrolyte exists. Therefore, Someya's group developed an ultrathin gel electrolyte (~ 6 μm) by loading a nonvolatile ion-containing glycerol into the PVA and an acrylamide matrix.^[222] The as-fabricated solid OECT with channel dimensions of $W \times L = 50 \times 50$ μm^2 showed comparable g_m (~ 1.62 mS) and operating frequency (~ 100 Hz) to devices utilizing liquid electrolyte, which could operate steadily on dry biological surfaces for long-term ECG monitoring. To minimize the noise acquired from the external power sources and short interconnection wiring during signal acquisition, self-powered ultraflexible OECT devices by integrating with organic photovoltaics were developed to monitor the ECG signals with higher signal-to-noise ratios (~ 40.02 db) when applied to skin or other tissue, than devices biased by a commercial battery.^[223]

EMG recordings is useful for the analysis and evaluation of electrical activity

produced when muscle cells are electrically or neurologically activated, and have great prospect in clinical and biomedical applications such as identifying neuromuscular diseases, studying kinesiology and disorders of motor control, etc. Recently, an integrated OECT/OFET device with a total thickness of $\sim 2.0\ \mu\text{m}$ including the substrate and encapsulation layers was laminated on the surface of a rat's gracilis muscle for EMG monitoring (**Figure 10e**).^[42] The localized muscle movement was induced by blue light coupled optical stimulation, which would evoke myoelectric potential to modulate the drain current. Due to the excellent conformability and negligible mechanical interference during the dynamic muscles motion, the integrated device enabled recording of EMG signals with a higher time resolution ($<1\ \text{ms}$) and high stability which showed a uniform amplitude during repetitive stimulations at 2, 3, 4 Hz.

Furthermore, in order to maintain stable and long-term measurements, hydrogels or ionic liquids gels were used to establish a better contact of electronic device with the skin.^[224, 225] Overall, OECTs, as a new technique for recording high quality of electrophysiological activity, have attracted a great deal of attention in clinical medicine and health monitoring application, which broadens the application of OECTs. We believe OECTs can be explored to open new application areas in neuroprosthetics and relative artificial intelligence robots where high-quality recordings are required while driving artificial limbs and exoskeletons in the future.

4. Challenges and Perspectives

In this review, we systematically presented recent progress in OECTs including the fabrication techniques and potential applications. The massive research on the topic of OECTs demonstrates the insistent demands for a wide range of applications. As reviewed above, OECTs are desirable for multiple biological sensors, logic circuits, neuromorphic devices and electrophysiological monitoring devices. With regards to integration, self-powered OECTs enables accurate and continuous signal detection without the need for an external power supply or bulky connecting wires, revealing the great potential for next-generation autonomous electronics. Although extensive

progress in OECTs has been achieved, several challenges still exist for future practical applications.

The existing fabrication techniques for large scale OECTs such as printing and laser-pattern techniques still require either the use of a shadow mask, or precise alignment to pattern the channel area. The limited resolution of device geometry remains to be further reduced to nanoscale. Other efficient techniques such as gravure printing and reverse-offset printing are hoped to be applied for the fabrication of large-scale OECTs with high printing speeds and high-resolution patterning.

Thus far, much of the printing and application efforts have focused on PEDOT:PSS. However, these PEDOT:PSS films have limitations, where the acidity of PSS hinders the stability. In addition, the swelling behavior of PEDOT:PSS in an aqueous solution enables it to absorb water molecules and expand its volume, which is essential for efficient ion injection and transport. On the other hand, it also limits the electrochemical switching speed and affects the reversibility of the doping process. It's imperative that the ion pathways (amorphous regions) and hole/electron pathways (crystalline regions) should be balanced or inclined for the maximum OECT performance and desired applications. Many other conjugated polymers are typically hydrophobic, and thus impede ion penetration and slows down the transient response of the device. There is therefore a drive to look at the recent promising new OECT materials and explore them further in various printing techniques and evaluate their suitability in technological translations.

In OECT devices, the structure of the films and device performance are highly dependent on operating conditions such as electrolyte concentration, gate electrode capacitance, and the overall patterned device area. As reported, the aqueous electrolytes can ensure high operational stability, but the long-term device performance can change over time (months or even years) depending on storage or operating conditions (e.g., humidity, temperature, and presence of oxidants). On the other hand, the possible leakage of electrolyte is another issue for conventional OECTs in practical applications, thus needs a high-cost device encapsulation inevitably. Moreover, it is difficult to scale-down these devices for high density

applications, and flexible OECT devices due to the liquid electrolytes and packaging issues. Therefore, the development of a solid-state electrolyte can avoid the leakage problem and enable a long-term performance stability, which can extend the applications in wearable devices and electronic skins. The use of solid electrolyte technology also allows for the fine-tuning of ion-polymer interactions independent of the aqueous electrolyte and provides an encouraging way for OECT device optimization through interface engineering. However, since ion penetration is typically slower in a solid electrolyte as compared to a liquid media, the device switching response becomes slower. Approaches to develop active polymeric electrolyte materials for solid-state OECTs that would allow efficient ion uptake and transport are therefore highly sought after.

Wearable sensors based on large-scale OECTs array are expected to be developed for further smart diagnostic purposes, such as the monitoring of temperature and blood oxygen, or health status parameters extracted in sweat, etc. Flexible OECT devices should keep abreast with the development of large-scale integration and low power consumption to meet the demands of the application in neuromorphic computing, which requires combining and integrating millions of individual elements. Reducing the size of individual OECT device can achieve higher spatial resolution of OECTs array, allowing for high-level large-area integration. Vertical OECT is an efficient way to save device space, but with a complex fabrication process and high cost. The use of solid electrolyte is also likely compatible with space-saving capacity.

Fiber-based OECTs are highly desirable for wearable electronics that are expected to be flexible, light-weight, durable, and comfortable. Although some high-performance OECTs with fibrous or textile structures have been reported, the current state-of-arts of processing technologies are still expensive and complicated. In addition, for textile based OECTs, there is a need for durability, washability, abrasability during wear for practical implementation. Moreover, OECT devices aimed for biosensors and medical facilities should have satisfactory performance while being physically conformal, and entail the use of conformable,

degradable/resorbable materials that do not permanently disrupt the biological functions. The lack of excellent stretchability would obstruct their practical applications because of the restricted degree of freedom and insufficient mechanical robustness. At the current rate of progress and with the help of multidisciplinary research, we presume that most of these issues could be addressed soon and further broaden the application fields of OECTs.

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