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Organic field effect transistors (OFETs) in environmental sensing and health monitoring: A review

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ABSTRACT

Organic field effect transistors (OFETs) have been the focus of sensing application research during the last two decades. In comparison to their inorganic counterparts, OFETs have multiple advantages such as low-cost manufacturing, large area coverage, flexibility, and readily tunable electronic material properties. To date, various organic semiconductors (OSCs), both polymers and small molecules, have been extensively researched for developing active channel layers in OFETs, thus enhancing their sensitivity and selectivity. However, OFET devices still need to be optimized to demonstrate reliable performance at the device level and in sensing applications. This review begins with an introduction of the OFETs with an emphasis on their geometry, materials (OSCs), fabrication process, and data analysis. After this, multiple applications are discussed, and the progress regarding sensing elements and precisions is highlighted. Finally, the challenges and possible future directions of OFET arrays in embedded sensing platforms are presented.

1. Introduction

Because of the ever-growing threat to human health and safety, a large number of researchers have focused on the topic of environmental sensors. The development of a common and cost-effective platform for sensing different environmental parameters is indeed the need of the hour. For the past four decades, researchers have been trying to address the same with an "electronic nose" (e-nose) platform [1]. Such a device requires mimicking an organic, biological nose to build an artificial detection platform. Like the olfactory receptor neurons (ORNs) that take care of sensing in the nose, a chemical block taking care of sensing analytes is used in the artificial structure [2]. The current e-nose platform is a multisensor package with provisions to detect minute quantities of chemicals such as a CNG leak, mining gases, and chemical warfare agents. Despite extensive research dating back decades and by the use of the same principle of combinatorial selectivity, e-nose still cannot match the performance standard of a biological nose [3]. The advantage of an e-nose is that it can be designed to perform desired tasks in limited applications, for example, for the breath analysis of patients with tuberculosis (TB) [4].

Environmental sensing and health monitoring can be classified as the sensing of chemical analytes, ionizing radiation, pressure, humidity, and temperature, which is the focus of this review. For a sensor to be efficient, it should satisfy the following criteria:

(a) Maximum surface-to-volume ratio of the sensing element [5,6]
(b) Quick response and recovery times
(c) High selectivity [7]
(d) Stability [8,9]
(e) Reusability [10], and
(f) High sensitivity [11]

Initially, when the microelectromechanical systems (MEMS) [12], fabricated using microfabrication technology, are used for

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numerous sensing applications, the high fabrication cost and the level of complexity involved prohibited this technology to be implemented on a broad scale. For example, micro accelerometers used in car airbags, microphones in cellular communication, and cantilevers in atomic force microscope (AFM) [12] are all based on microfabrication technology. In recent years, biological micro-electromechanical systems (bio-MEMS) have also shown promising results in both research and industry [13]. Second, field-effect transistors (FETs), although patented in 1926 by Julius Edgar, were first realized as practical devices by physicists only in 1947 and were purely based on an inorganic semiconductor. The major disadvantage of the inorganic FET-based sensors is the cost involved in the manufacturing process and the cost of semiconductor materials (Si, Ge, GaN, and GaAs). Despite faster switching speeds and reduced operating voltages, sensors made up of these inorganic FETs can address only certain applications because of a lack of sensitivity and/or selectivity for various environmental conditions. Spectroscopy is the third commonly used technology to detect analytes, different physiological parameters, and other physical properties. Morpho, an ion mass spectroscopy-based handheld device, is successful and used by the US army in the event of explosion to analyze the explosives used. All of the above techniques need complex units to be built at high manufacturing costs. Moreover, the data produced from different sensors are in different forms, thus making it a challenge to build e-nose platforms consisting of various pattern recognition approaches for extracting any meaningful information out of these sensing platforms. Orthogonality in sensing, although discussed widely as a way to address the selectivity issues, remains a challenge for experimentalists.

Organic materials have been studied for more than five decades, and there are numerous applications for these materials in the field of microelectronics. In the microfabrication industry alone, these polymers are used as gate insulators, photoresists, sacrificial layers, and encapsulation. However, currently, they are also being used as an active organic semiconducting thin film for OFETs. In addition to their application as an electronic device, OFETs are also being used for multiple sensing applications such as chemical sensing, biological sensing, humidity sensing, and ionizing radiation sensing. There has been a tremendous push to improve the performance of the OFET sensors to attain higher sensitivity and selectivity by using state-of-the-art techniques. However, organic electronic devices still need to be optimized to demonstrate reliable performance. Such a research on optimization could contribute to the evolution of low-cost and highly reliable sensors to assess the environment and human health, thus leading to a plethora of applications.

This review article discusses the feasibility and different aspects of the OFET sensors for environmental sensing and biosensing applications. However, there are review articles available on OFET-based sensing because of the ever-increasing output of scientific publications [14–17], a specific topic of multiparametric sensing, and the probability of integrating OFET-based sensors into a system has been chosen. This article also provides an overview of the advantages associated with the OFET sensors with the hope of pushing the boundaries of the research.

2. Sensing mechanisms for sensors

Sensing mechanisms for different gases, chemicals, pressure, and irradiations are complex and diverse because of various physical and chemical effects. In some cases, the sensing materials may change physical properties such as stress, temperature, and doping. Additionally, the limit of detection (LOD) of various analytes varies from one sensor to another, and the necessity to adhere to LOD, in turn, depends on analyte concentrations. In view of this, a few important (and widely used) mechanisms pertaining to sensors are discussed in this section. Fig. 1 shows two schemes, where we propose the mechanisms for multiparameter sensing in (a) and we show the sensing capability of OFET for similar environmental/biological parameters in (b).

Oxidation and reduction mechanisms have been in use for a long period to exploit the goal of sensing. Recently, Tian et al. used ZnO nanorods and Pt nanoparticles for the detection of ethanol and NO_2 gas [18]. The detection process is explained with the help of a change in the resistance of the pristine Pt/ZnO/nanosheet stack using reduction (Ethanol) and oxidation (for NO_2) mechanisms. Immediately upon exposure to air, O^- species accumulate around the Pt nanoparticles. These species, when they interact with ethanol, increase the accumulation of electrons on the surface, thereby decreasing the resistance of the pristine Pt/ZnO/nanosheet. In the case of NO_2 sensing, a reverse phenomenon increases the resistance while forming a depletion layer of O^{2-} ions on the ZnO surface. Thus, a chemiresistive sensor was demonstrated successfully with help of a stack of materials (Pt/ZnO/nanosheet) that have excellent charge carrier transport properties. Zhou et al. reported the intersheet sensing mechanism of reduced graphene oxide (RGO) while exposed to CO_2, which is neither a reducing nor an oxidizing gas [19]. The results obtained by the group were explained by the change in resistance due to the structural deformation rather than the charge exchange behavior of RGO. The flowrate of the carrier gas plays a key role in the electrical characteristics of the device. A higher flow rate creates compressive stress on the RGO film, thereby reducing the intersheet distance, which in turn reduces the electrical resistance. On the other hand, a low flow rate creates tensile stress, thereby increasing the electrical resistance. Finally, an optimal flow rate of 300 ml/min has been chosen for the CO_2 sensing.

Quartz crystal microbalance (QCM), a mass-sensitive device, has been known to be a reliable sensor because of its high sensitivity and measurement accuracy. Sauerbrey's equation is a well-known mass-sensitive principle and is used in the QCM

$$\Delta f = -2.26*10^{-6} f_0^{-2} \frac{\Delta m}{A} \quad [1]$$

where Δf is the shift in the frequency, Δm is the change in the mass of the electrode after the analyte interaction, f_0 is the resonant frequency of the bare QCM, and A is the quartz crystal active area. Sensitivity to analyte is calculated as $\Delta f/\Delta C$, where C is the minimum concentration of the analyte for which the sensor responds. Zhang et al. has recently reported on sensing humidity with the polyaniline (PANI)/GO composite. The highest sensitivity of 20 Hz/%RH was reported for 3 layers of the PANI/GO composite as compared to the higher number of layers [20]. Photoluminescence-based sensing applications span across a broad area ranging from explosives to biological analytes. The process involved in optical sensing makes noninvasive and hazardous condition measurements easy. Here, a single excitation light source is used to probe the device under test, and the emission peaks are monitored. The emitted peak carries the information related to the target analyte or the effect due to the physical parameter under measurement. Different applications such as metal ion sensing, gas sensing [21], and glucose monitoring were successfully demonstrated with this mechanism. Similarly, as discussed in the introduction part, FET devices have a different mechanism of sensing. The active channel layer current is modulated when an interaction occurs between the semiconductor and the interacting parameter under consideration. The following equation provides more details on the current modulation with regard to device parameters

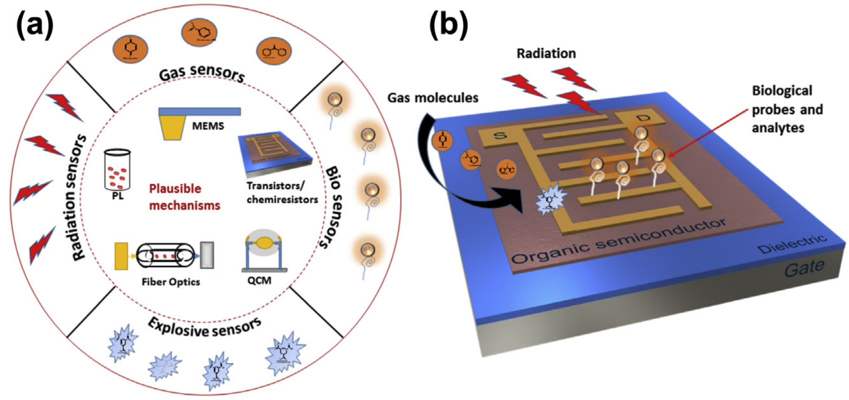

Fig. 1. (a) Scheme of applications and mechanisms and (b) proposed schematic for an OFET-based multienvironmental parameter sensor.

$$I_D(lin) = \frac{Cox.\mu}{2}\frac{W}{L}\left(2(V_{GS} - V_T)V_{DS} - {V_{DS}}^2\right) \quad [2]$$

$$I_D(sat) = \frac{Cox.\mu}{2}\frac{W}{L}(V_{GS} - V_T)^2 \quad [3]$$

where I_D is the drain current, C_{ox} is the capacitance of the dielectric layer, μ is the effective mobility, W and L are the width and length of device, V_{GS} is the gate bias, V_{DS} is the drain bias, and V_T is the threshold voltage.

3. Overview of OFETs

Organic electronic devices (OEDs) have greatly advanced and are presently making their foray into the commercial world. Organic thin film-based displays already exist in the market, such as mobile phones and PC monitors. The basic building blocks of these displays, the OSCs, have been synthesized and studied for more than five decades. Their main advantages are their roll-to-roll processing, mechanical flexibility, low/room temperature processing, and potentially low-cost manufacturing. Organic channel materials (OSCs) have also attracted much research, where the charge carrier transportation is taken care of by the π-conjugated system of OSC. Much like conventional transistors, OFETs allow a controlled current to flow between the source and drain electrodes through the modulation of gate voltage. The generic term used for most types of organic transistors is OTFT/OFET; however, there exist several categories of organic transistors depending on the active channel current modulation mechanism, which are out of the scope of this review. On the basis of their device structure distinguished by the order in which the OSC, dielectric, and metals are deposited, OFETs are primarily categorized as the following.

a. Bottom gate/bottom contact (BGBC)
b. Bottom gate/top contact (BGTC)
c. Top gate/bottom contact (TGBC)
d. Top gate/top contact (TGTC)

Many reports cite the role of oxygen present in the atmosphere as an impediment to stability for the OFETs [22,23]. In this context, TGBC devices have been reported to be more stable than BG because of their OSCs being protected by dielectric and gate layers. However, for sensing devices, the stability remains a challenge due to interference from the environment. Further, Zang et al. reviewed device engineering aspects of OFET to improve sensor performance [24]. Interestingly, they were able to identify three key parameters (active layer thickness, receptor implantation (bilayer), and optimization of geometry) to tune the transduction signal and amplify it. In general, the active channel layer of an OFET is approximately several tens of nanometers and acts as an impediment for incoming analyte molecules. Hence, reduction in the channel thickness will always provide scope for improvement in sensitivity and response time [25]. Another important aspect in OFET-based sensing is specificity, and one needs to add multiple functional groups to the pristine OSC to achieve the same. This leads to the specific interaction of the desired functional groups with the corresponding analyte molecules, but this kind of approach is still under research, as it is prone to adverse effects like field-effect modulation deterioration. Eventually, a bilayer approach has been adopted for the device geometry, where another layer of receptor molecule binds covalently or noncovalently to the pristine organic semiconducting layer [26].

Pressure sensing applications were also demonstrated with the help of OFETs, where the geometry has been engineered by introducing an additional layer in the dielectric, as per the sensing needs. Yin and co-workers [27] exploited such solution-processed bilayer dielectrics to fabricate pressure sensors, where poly(-methyl methacrylate) (PMMA) and polyelectrolyte of polyacrylic acid (PAA) are combined together. PMMA/PAA exhibited improved electrical characteristics, as the new combination contributed to high capacitance and reduced leakage currents. A TGBC geometry OFET has been fabricated with a conjugated polymer (PIDT-BT) as an OSC and PMMA/PAA duo as a dielectric layer on top of a flexible substrate (PET sheet). Furthermore, it is shown that a reduced thickness in the PMMA layer down to 30 nm improves both capacitance and mobility. Additionally, a record sensitivity of 56.15 KPa^{-1} has been achieved in 19.2 ms at an operating voltage of −5 V.

4. Sensing using OFETs

Organic semiconducting materials have also been used as the receptor layer for devices in sensor applications. The advantage of using polymers is that the selectivity can be varied with the substitution of various functional groups on the main polymer. Subsequently, these organic materials with unique electronic and mechanical properties have been found suitable for the development of low-cost electronic devices. Depending on the chemical structures in these molecules, their stability varies and makes them susceptible to changes in the environment. For example, the oxygen content in air and humidity alters the transport properties of the OSCs. To address these problems, researchers use glove boxes with inert environments, where the oxygen and humidity levels are controlled. Subsequently, the devices are encapsulated inside the box before they are taken out. Table 1 presents significant information about OFETs used for sensing multiple analytes (gases, explosives, and VOCs), and they do not have any encapsulation.

4.1. OFET sensors for the detection of poisonous gases and explosives

The detection of poisonous gases has been given overwhelming importance in recent years. Like volatile organic compounds, ammonia (NH_3) is yet another dangerous analyte that is abundant in the atmosphere. Any decaying organic matter can produce NH_3, and under normal environmental conditions, it is a colorless, caustic (corrosive), and pungent-smelling gas. Jeong et al. used a DPP-based conjugated polymer to realize a highly sensitive flexible NH_3 gas sensing OFET [29]. Here, it is reported that the thinner devices (~10 nm DPP) annealed at 150 °C show a higher sensitivity of >90% upon exposure to 1000 ppm NH_3 gas (as shown in Fig. 2a) than thicker devices (~25 nm as shown in Fig. 2b). Further, the devices showed a negative shift in the transfer curves for prolonged exposure to NH_3 due to the formation of trap states in the tail state by chemical interactions. Interestingly, when tuned properly, dielectric layers also play a crucial role in the detection of analytes in OFETs. Recently [30], ZnO nanoparticles with polymethyl methacrylate (PMMA) mixed in a 1:1 (v/v) ratio was used for the deposition of a hybrid dielectric layer by spin coating. The authors [30] chose pentacene as the channel layer and ITO-coated glass as the substrate for two devices: (A) ZnO/PMMA hybrid dielectric and (B) only PMMA. The corresponding transient response is shown in Fig. 2d for both the devices with the same geometry as shown in Fig. 2c.

Device A is highly sensitive compared to device B because of the small grain size and adsorption of oxygen on the surface of the ZnO nanostructure. In the first case, the small grain boundaries indicate more NH_3-adsorbing sites, and in the second case, the oxygen adsorbed on the ZnO reacts with the NH_3 and releases electrons, thereby reducing the barrier height and leading to a decrease in ZnO resistance [30]. Similarly, the detection of trace chemical explosives using OFETs has been extensively investigated by the Katz Group at John Hopkins University, who are developing multiple platforms and processes to detect explosives that are both airborne and waterborne. Later, the same group demonstrated a dilute TNT analyte solution detector with the PQT12:TPT-TTF-blend OFETs [31]. Katz et al. chose BGBC geometry for the sensors, while both pristine PQT12 and its blends were used as a channel-cum-receptor layer. Four different blends made up of PQT 12 and TPT-TTF (1, 5, 10, and 20 wt%) were tested with different concentrations of TNT in IPA. A maximum response in terms of change in current $\Delta I/I$ (%) was obtained with PQT12:10% TPT-TTF blend when exposed to 10^{-4} mg TNT/ml IPA, which can be seen in Fig. 2e,f. The control studies conducted with pristine PQT12 did not show any response with either a varying concentration of TNT or with other analytes such as 2,4-DNT; 2,6-DNT; and amino-2,6-DNT, whereas the PQT12:10% TPT-TTF blend was highly selective to TNT among all these analytes and also showed varying sensitivity toward various concentrations of TNT in IPA. The sensing mechanism was ascribed to the TNT molecules that quench the functional groups of TTF, thereby allowing the de-trapping of holes and causing the current to increase in the channel.

4.2. OFET sensors for volatile organic compounds (VOCs)

Organic compounds are chemicals that contain carbon, and VOCs are compounds that easily become vapors and gases. In OFETs, source-drain currents flow in the first few monolayers (in the nm range) at the interface of OSC and dielectric layers. To sense analytes efficiently using OFETs, the channel current in these monolayers needs to be altered by the diffused analyte molecules. Thus far, with the help of porous structures and by employing ultrathin films, the conducting channel currents have been altered, thereby leading to a gas sensing response of OFET. Khim et al. used the wire-wound bar coating method to attain highly crystalline and ultrathin films (<2 nm) that can be used for chemical sensing, as shown in Fig. 3a [37]. Further, a thickness-dependent sensitivity investigation of the active channel layer (of two polymers) in OFET was carried out while proving that different morphologies were obtained for different thicknesses. This kind of coating method can be used generically for various platforms (substrates), both flexible and nonflexible. Further, there is no discussion on data obtained and its significance in developing an e-nose system. Addressing this issue, Alona et al. [35] used polycyclic aromatic hydrocarbons (PAH), a p-type semiconductor for the detection of various VOCs, and a classification of these analytes was carried out with multivariate analysis, as depicted in Fig. 3b. The sensitivity of each PAH variant during exposure to different analytes (VOCs) was depicted with the help of a normalized drain current ($\Delta I/I_0$), hole mobility

Table 1
Summarizing OFET sensors for various analytes.

OFET Configuration	Organic Semiconductor	Analytes	Detection Limit	Selectivity study	Reference
BGTC	P3HT/ZnO/GO	NO_2	1 ppm	No	[28]
BCTG/BGTC	DPP (LGC-D148)	NH_3	1000 ppm	No	[29]
BGTC	Pentacene (ZnO + PMMA dielectric)	NH_3	10 ppm	No	[30]
BGBC	PQT12 + TPT-TTF	TNT	1 ppt	Yes	[31]
BGTC	DNTT	NH_3	10 ppb	Yes	[32]
BGTC	Spirobifluorene-based polymer	H_2S	1 ppb	Yes	[33]
BGTC	PDPPHD-T3	xylenes	40 ppm	Yes	[34]
BGTC	Polycyclic aromatic hydrocarbon	VOCs	–	Yes	[35]
BCTG	Tips-Pentacene + PTAA	Cysteine	0.01 mM	No	[36]
		PEIE	0.04 wt%	No	
BGTC	DPP-TT	NH_3	10 ppm	No	[37]
		Ethanol	1000 ppm	No	
		Ethylene	1000 ppm	No	
BGTC	P3HT-azide/caliz[8]arene	Methanol in toluene	1 vol%		[38]
		Ethanol in toluene	1 vol%		
		n-Hexane in Toluene	1 vol%		
		Toluene in Methanol	2 vol%		
BGTC	DPP2T-TT	NH_3	<1 ppb	Yes	[39]
	C8-BTBT/PEI	CH_2O	<1 ppb	No	
BGTC/BCTG/PG	pBTTT-C14	BuOH	<100 ppm	No	[40]

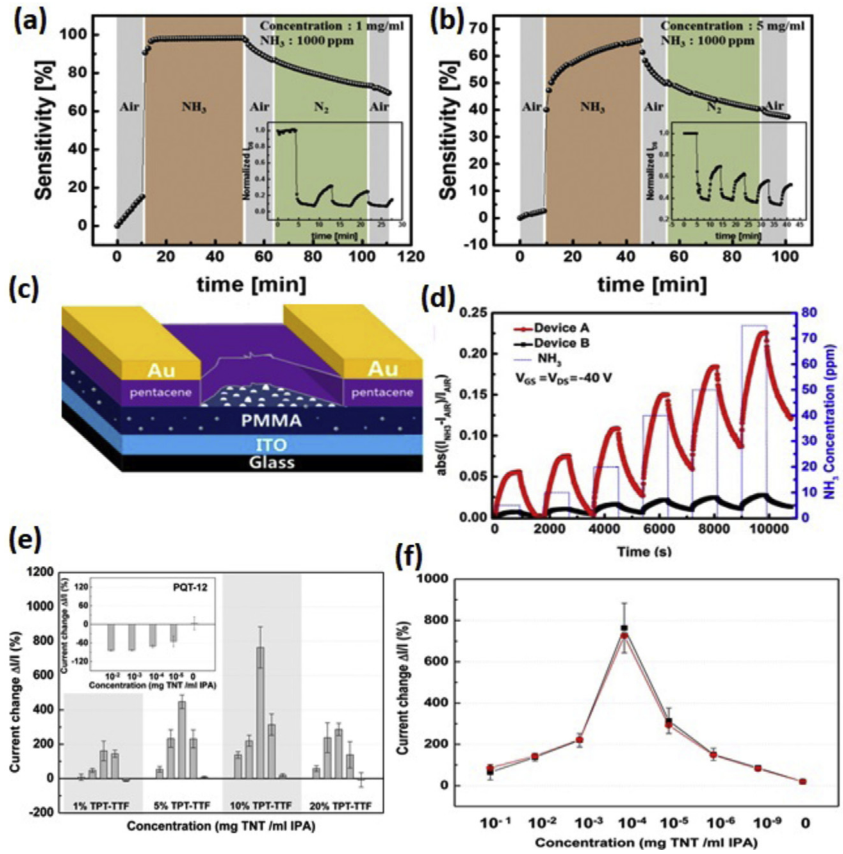

Fig. 2. (a) and (b) Sensitivity of 10 nm and 25 nm NH_3 gas sensors, adapted from Jeong et al. [29]; (c) and (d) OFET geometry and transient analysis of the same with a ZnO/PMMA composite-based dielectric exposed to NH_3 gas with various times; adapted from Han et al. [30] (e) Bar graph of TNT-exposed devices to multiple concentrations of TPT-TTF; and (f) Current change ΔI/I (%) at VD = VG = −40 V of blended PQT12:10% TPT-TTF with a maximum at ~10^{-4} (mg TNT/ml IPA); adapted from Katz et al. [31].

($\Delta\mu_h/\mu_{h0}$), and threshold voltage ($\Delta V_t/V_{t0}$) values. In the case of discontinuous PAH (PAH-2, PAH-3, and PAH-7), transistors exhibited positive and negative V_t, for with HTS and without HTS surfaces, whereas in the case of PAH-4, the opposite effect was observed.

Apart from that, a discriminant factor analysis (DFA) was implemented using software (SAS JMP) to distinguish between various VOCs. Formaldehyde (HCHO) is one such VOC that gets off-gassed from furniture and poses a significant danger to human health. Zhang et al. [39] showed that introducing nanopores to an OFET sensor can improve its sensitivity to low concentrations of HCHO while reducing the response time and retaining the recovery characteristic to confounding factors (e.g., temperature and moisture). Although they did not explain the improvement in molecular ordering because of the introduction of nanopores, high charge carrier mobilities were obtained with the use of a new receptor layer, polyethyleneimine (PEI). The presence of a PEI layer on top of C_8-BTBT caused electron doping of the latter and decreased the current. Further, when carbonyl groups of HCHO interact with amine groups of PEI, the C_8-BTBT layer is de-doped and currents are regained. Thus, both factors (nanopores and PEI layer) improved the sensitivity and response time of the OFET sensor.

4.3. OFET sensor for ionizing radiation dosimetry

Owing to their sensitivity toward various environmental conditions, gases, and chemicals, OFETs are gaining acceptance for various sensing platforms and security applications. However, the unstable nature of the organic semiconducting materials is also well reported [41,42]. Within the emerging field of OFET sensors, researchers have reported the use of OFETs as sensors/detectors for ionizing radiation using organic semiconducting materials such as P3HT, pentacene, ZnPc, and CuPc [43,44]. It has been demonstrated that the sensing is due to the impact of high-energy ionizing radiation on organic semiconducting materials, which can easily penetrate through the thin passivation layer. Therefore, passivated OFETs can also act as sensors for ionizing radiation while ignoring the environmental instability of organic semiconducting materials [43]. For different types of irradiation, Table 2 summarizes various OSCs used as an active semiconducting layer for an OFET.

Upon exposure to γ-rays, an increase in the p-type doping and energy stage generation in the band gap of pentacene has been studied, and the pentacene OFETs were proposed as dosimeters for ionizing radiation of γ-rays by Shilpa et al. [43]. Similarly, the thin films of CuPc and ZnPc were reported to have linear dependence of absorption in B- and Q-bands upon exposure to γ-irradiation by Divya et al. Therefore, OFETs with CuPc or ZnPc were proposed to be used as real-time dosimeters for γ-rays [44].

Basirico et al. [45] investigated X-ray-induced photocurrents in TIPS-pentacene thin films, while Lai et al. [47] employed changes in the electrical characteristics of TIPS-pentacene and proposed the OFETs as a detector with changes in the electrical characteristics upon exposure to X-rays. Fig. 4a shows the experimental setup, and Fig. 4b,c shows the change in I_{DS} for the OFET upon X-ray exposure. Along similar lines, Paternò et al. [49] exposed a P3HT-based OFET to neutron radiation and demonstrated changes in the electrical properties. While Basiricò et al. [50] studied the degradation of the mobility of TIPS-pentacene-based OFETs upon exposure to proton irradiation, a study was conducted by Kim et al.

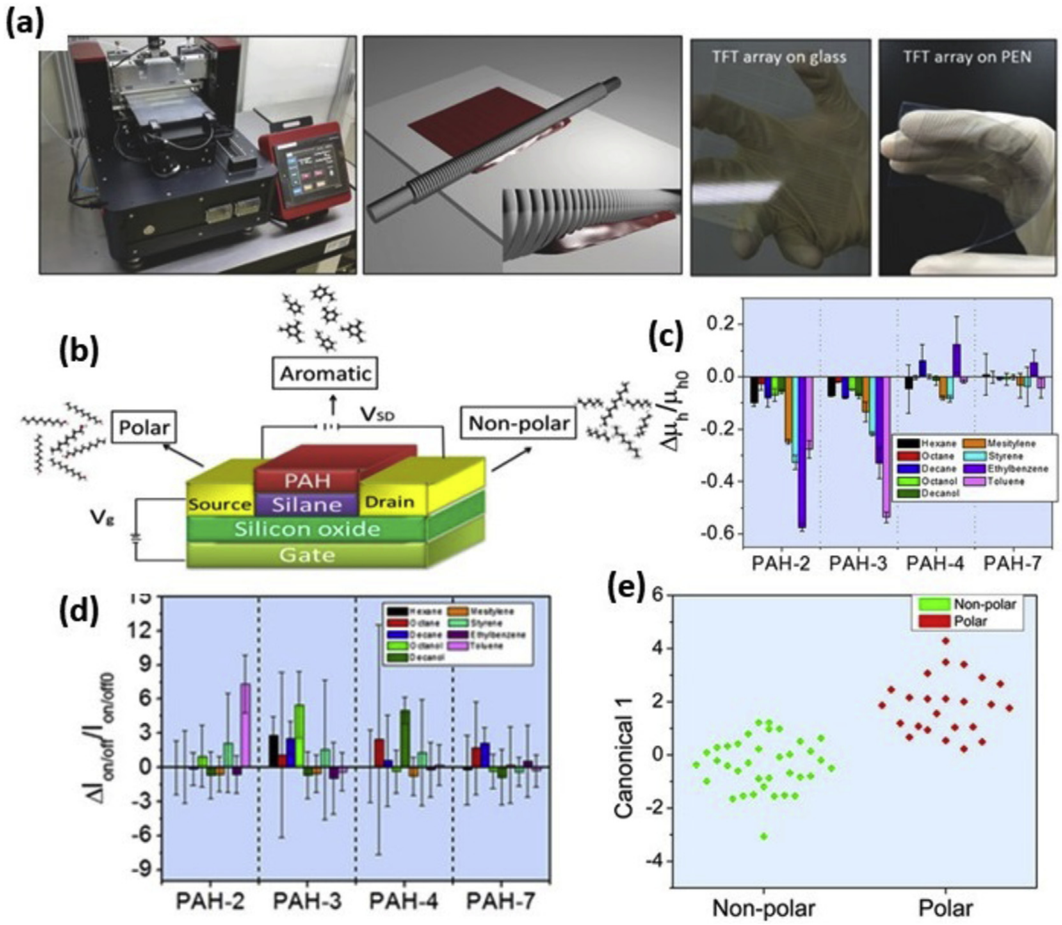

Fig. 3. (a) Bar-coating machine and the bar used for fabricating TFTs on glass and PEN sheet, adapted from Khim et al. [37]; (b) schematic of OFET exposed to different analytes; (c) normalized mobility (Δμh/μh0) values while exposing the devices studied to different analytes; (d) normalized average current ratio (Δ I_{on}/I_{off}) values while exposing the devices studied to different analytes; (e) discriminant factor analysis (DFA) of the polar materials and the nonpolar materials at different concentrations; adapted from Alona et al. [35].

[52] on rubrene-based BGTC OFET. They observed the change in the threshold voltage of an OFET with rubrene upon exposure to electron beam irradiation, which concluded that an n-type doping was induced upon electron beam irradiation to rubrene, as shown in Fig. 4d,e.

4.4. Organic field effect transistor-based biosensors

OFET-based biosensors show promise for detecting low concentrations of biomolecules and studying biochemical interactions [17,53]. A remarkably low concentration (10^{-16} M) of dopamine was detected using an electrolyte-gated OFET [54]. The biosensor used for detection was fabricated using a transducer material (Pt NPs decorated uniformly on the rGO surface) on a graphene substrate (Fig. 5a). The large-scale graphene worked as a highly conductive template for Pt NP decoration, which was the main catalyst for the oxidation of dopamine, and the sensitivity of the biosensor was dependent upon the amount of Pt decoration. In another electrolyte-gated OFET, during the detection of the inflammatory cytokine TNF-α, a minimum detection level of 10^{-10} M was achieved [55]. The OFET device contained a sensing element of an anti-TNF-α antibody-functionalized Au-gated electrode and a channel of a 15 nm thick pentacene film (Fig. 5b). This device was integrated with a microfluidic system for electrochemical characterization (Fig. 5c). The adsorption of TNF-α on the sensing element decreased the hole density, and consequently, the Fermi level of pentacene was increased.

The large electrical double-layer capacitance (*ca.* 10 mF cm^{-2}) in electrolyte-gated OFETs is formed at the interface between the OSC layer and electrolyte solution, which is useful for detecting the sub-femtomolar concentration of proteins [56]. With this idea, the functional bio-interlayer was introduced between the dielectric

Table 2
Summarizing the reported literature for ionizing radiation sensing and corresponding organic semiconducting materials.

Type of Ionizing Radiation	Energy of Irradiation	Organic Semiconducting Materials	OFET Configuration	Ref.
γ-rays (^{60}Co)	1.3 MeV	Pentacene, CuPc	BGBC, BGTC	[43,44]
X-rays	Several hundreds of keV	P3HT	BGBC	[45–48]
Neutron radiation	Energy in the beam of neutrons ranges from 0.025 eV to >10 MeV to >20 MeV	P3HT, PBTTT (thiophenes)	BGBC	[49]
Proton irradiation	3–10 MeV	TIPS-pentacene, pentacene	BGBC	[50,51]
Electron beam irradiation	Ranges from keV to MeV	Rubrene	BGTC	[52]

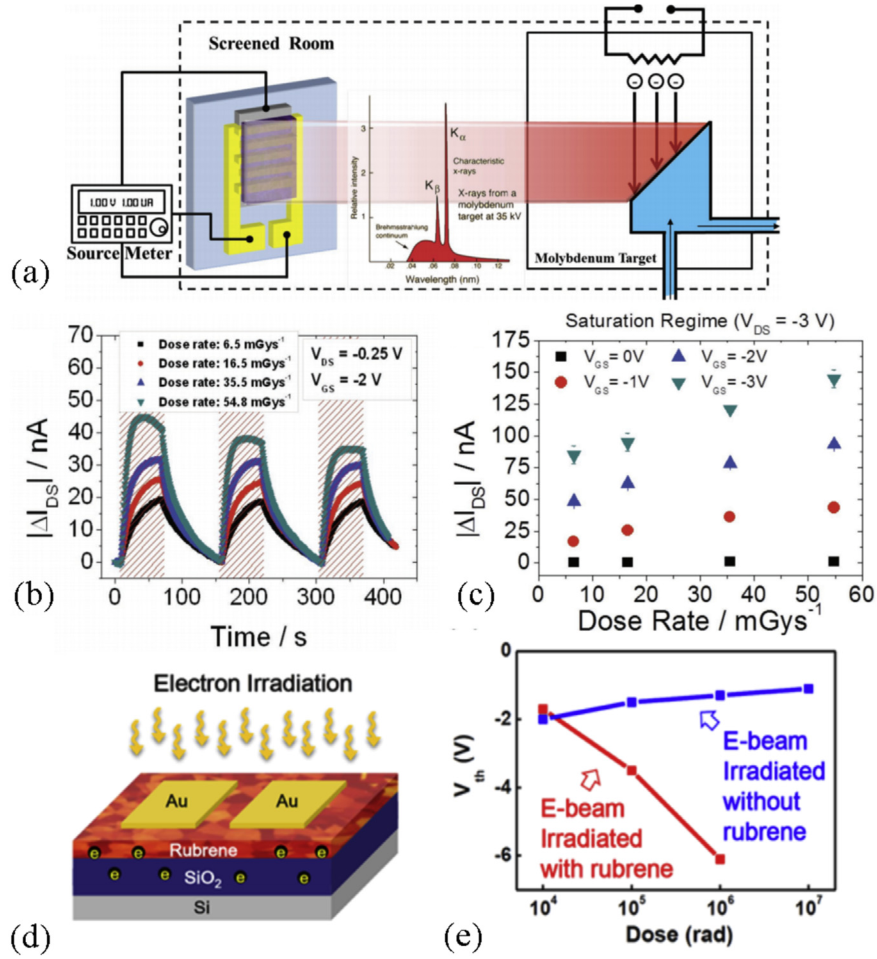

Fig. 4. (a) Experimental setup for TIPS-pentacene-based BGBC OFET exposed to X-rays; (b) Observed ΔI_{DS} when OFET exposed to X-ray irradiation for different doses; (c) Change in I_{DS} in the saturation regime with $V_{DS} = -3$ V demonstrating remarkably high sensitivity of approximately 1200 nC Gy^{-1}; adapted from Lai et al. [47]; (d) Schematic diagram of BGTC Rubrene OFET exposed to electron beam irradiation; and (e) Shift in the V_{TH} as observed upon electron beam exposure of rubrene OFET due to induced p-type doping adapted from Kim et al. [52].

and the OSC layers to fabricate highly efficient OFET-based biosensors. For the first time, this approach was followed by Magliulo et al. [57]. However, one major drawback of this device structure is a protein layer embedded under the OSC layer, which limits the availability of target molecules to reach the protein layer for detection.

Attempts have been made to tailor the sensing performance of the OFET-based biosensors to detect/catalyze the specific analytes [58,59]. However, it is important to anchor the bio-recognition elements/enzymes with the OSC layer for biosensing. To investigate the effect of protein detection parameters, Hammock et al. functionalized ultrathin PVP-HDA dielectric and DDFTTF-based OFETs using Au NP binding sites followed by a thrombin-specific aptamer attachment and a BSA treatment before thrombin exposure to the device, as seen in Fig. 5d [58]. A PDMS flow cell was designed to deliver thrombin directly on top of the OFET biosensor (Fig. 5e). During validation of thrombin sensitivity, the current response of the biosensor was decreased while increasing the thrombin concentration, and a nonlinear sensitivity was observed with a lower detection limit of 100 pM (Fig. 5f,g). Shen et al. used a plasma-assisted interfacial-grafting method to tailor the neighboring-conductive channel of OFET and also assembled an antenna onto the surface of OFET, which enables direct interaction of target analytes in the solution [59]. Because of direct and specific interaction, the sensor detected adenosine triphosphate (ATP) at a very low concentration of 0.1 nM. Further, reproducibility and selectivity were other enhanced properties.

Enzyme-based OFETs offer an advantage of excellent selectivity and sensitivity for specific analytes during electrochemical reactions [60,61]. Minami et al. used bipyridinium derivative as an electron-transfer mediator to functionalize the extended gate, which also supported the nitrate reductase enzyme [60]. Sodium dithionite in aqueous solutions served as an electron donor during nitrate reduction that was detected by changing the electrical characteristics of the OFET biosensor. The change in the channel conductance upon the addition of nitrate was noticed because of an enzymatic reaction. A low LOD (45 ppb) and high sensitivity were achieved. In another report, Minami et al. fabricated a novel flexible extended-gate OFET [61]. The surface was modified with lactate oxidase and horseradish peroxidase osmium-redox polymer to detect lactate levels selectively (Fig. 5h). The transfer characteristics of the fabricated OFET showed a negative shift of the transfer curve while increasing the lactate concentration (Fig. 5i). A linear relationship was seen in the low concentration range, and the LOD was 220 nM (Fig. 5j). Furthermore, the sensors were highly selective for lactate when measured in the presence of other biomolecules and ions (Fig. 5k). OFET-based biosensors thus opened up a novel opportunity for low-cost and flexible bioelectronic devices. There are, however, some technical challenges including the instability of the organic materials in aqueous media.

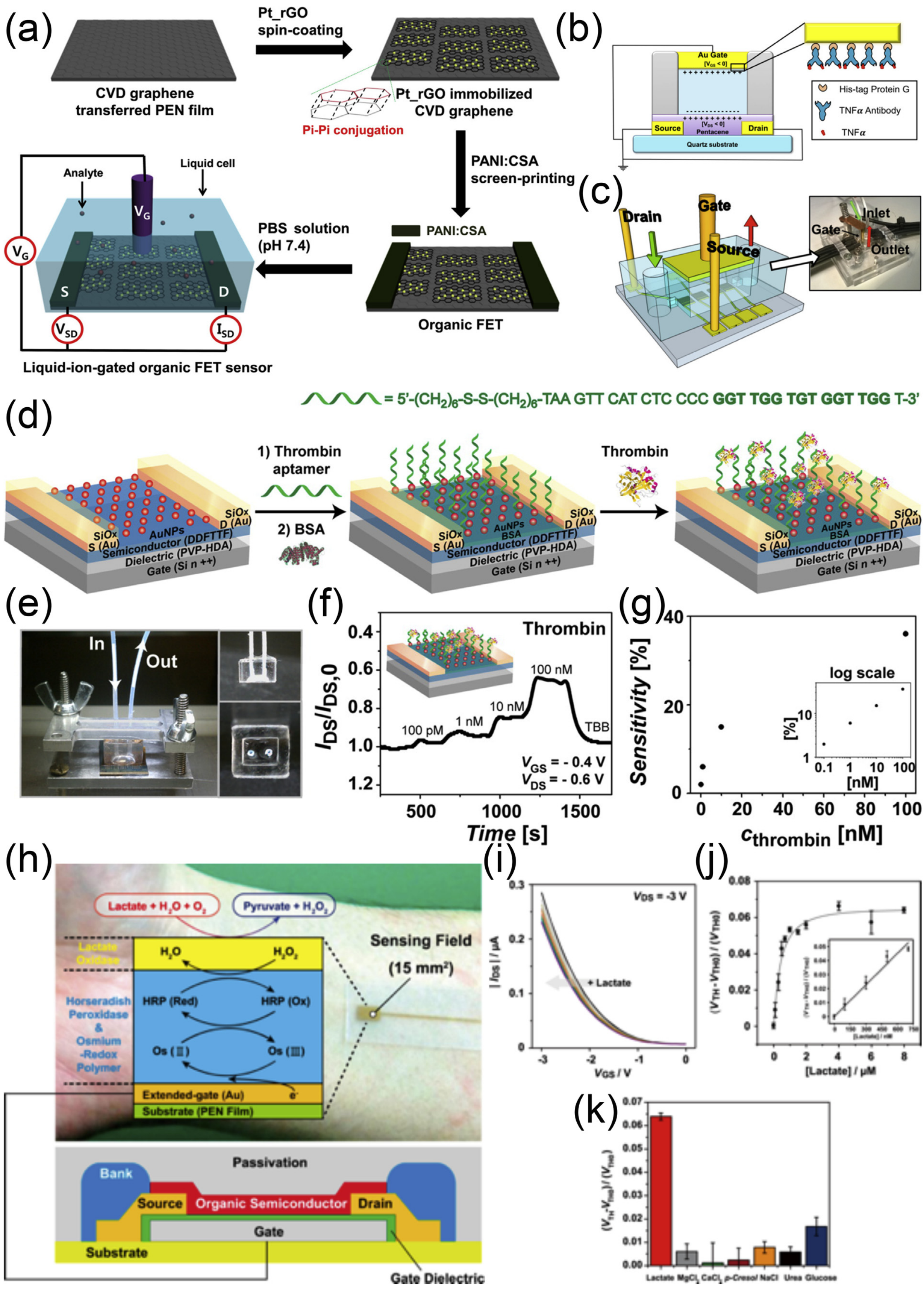

Fig. 5. (a) Schematic illustrating fabrication sequence of an OFET-based sensor, adapted from Jungkyun et al. [54]; (b) Schematics of the EGOFET device; (c) Drawing of the microfluidics with optical image showing experimental setup; adapted from Basiricò et al. [55]; (d) Fabrication of an OFET-based sensor for the detection of the target protein; (e) PDMS flow cell; (f) response to different concentrations of thrombin; (g) Calibration plot; adapted from Hammock et al. [58]; (h) Flexible OFET layout and optical image of OFET-based lactate biosensor attached on the wrist; (i) transfer characteristics; (j) calibration curve; and (k) selectivity test; adapted from Minami et al. [61].

5. Future directions of the research

An important aspect that requires considerable attention is the quantitative analysis of the threat that arises because of dangerous analytes such as dangerous gases, chemical vapors, explosives, VOCs, leaks of ionizing radiation, and/or various bio-chemicals. As discussed in this article, being sensitive to many of these analytes, OFET-based sensor platforms have to be “ghostbusters” with regard to even the minute concentration of these analytes. This means that either the active channel layer or the receptor layer in tandem with active channel should be spatially sensitive to the varying amounts of analytes. To achieve such a response to miniscule changes of analytes, one needs to have control over the composition of the OSC matrix, pore sizes in the material, or surface area of the nanoparticles in the receptor elements. Further, the thickness of the thin films under test (active channel-cum-receptor site) or the thinning of the receptor layer on top of the active layer of the OFET do not always improve the sensitivity [33]. The optimal thickness has to be figured out to achieve the highest sensitivity to the corresponding analyte, while being stable to other environmental factors. With its cost-effective, room temperature manufacturing process, flexibility, and versatility to be used for sensing applications, the OFET-based ionizing radiation sensing of various types needs further exploration for enhanced sensitivity and stability with different OSCs. With OFET being an electronic device, it is also possible to employ various circuit techniques to enhance sensitivity.

All the above problems need to be solved depending on the application and the necessity. It is also required to freeze all the fabrication process parameters, materials (their compositions), and characterization details discussed thus far, as it would help in generating data from identical devices. Additionally, relevant information about sensed analytes needs to be extracted carefully, and data analytics (DA) can be implemented to draw conclusions. Part of such DA is pattern recognition, which is the most important element in any sensing system with sensor arrays. It helps in identifying patterns of the data obtained from sensors and then classifying them in accordance with the algorithm. Data mining tools such as principal component analysis (PCA) are employed to reduce the higher dimensional data under test, and its output is used to build prediction models. Machine learning (ML) algorithms are used to learn from the data sets and build these prediction models. That is, we first train the algorithm with the training set and we then test the predicting accuracies with the test set of data. Subsequently, the test data can be used to assess the performance of a PR algorithm in accurately classifying sensor information. In this way, it helps in more accurately classifying or identifying the samples.

A singular platform that has the capability of embedding different OFET-based sensor arrays that can specifically detect and transduce multi-environmental parameters is a major challenge because of a wide band of variations and degradation of OSCs in the environment. For rapid commercialization, by reducing the complexity and cost, OFET sensors can be employed on flexible substrates with laser-scribed graphene (LSG) techniques [62]. The usage of LSG scribers on top of polymer substrates can be of help in realizing the source, drain, and gate contacts. Essentially, only a semiconductor-cum-sensor layer is deposited in such a case, and a device can be readily used as a sensor. Future research can focus on dealing with the long-term drifts associated with sensors, which cause problems in pattern recognition and lead to errors or false alarms. Individual OFET sensors need to be taken care of separately to deal with such issues. Finally, using an array of OFETs along with pattern recognition can lead to a very reliable e-nose platform.

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