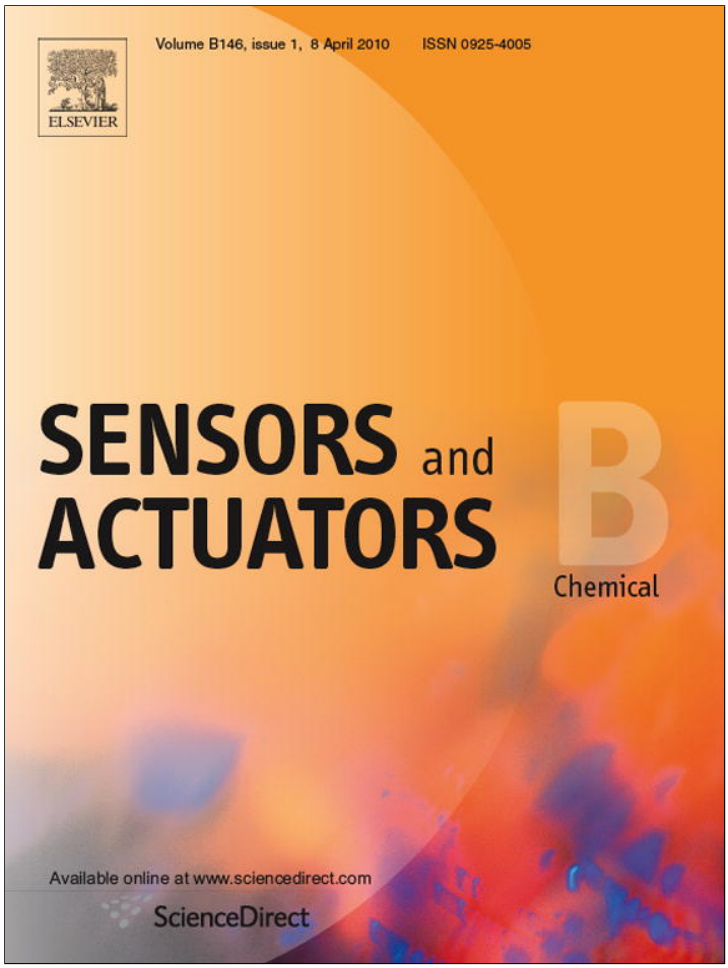

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The response characteristics of a gas sensor based on poly-3-hexylithiophene thin-film transistors

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ABSTRACT

This paper studies the response characteristics of a gas sensor for organic thin-film transistors (OTFTs), made from spin-coated poly-3-hexylthiophene (P3HT) on a thermally grown SiO_2/Si wafer. The gas response characteristics of OTFT sensors are observed from the change in the drain–source current, as a function of time, when the P3HT-based OTFT sensors of different channel widths are exposed to cycles of exposure to and the evacuation of NH_3 gas, with concentration ranging from 10 to 100 ppm at room temperature in normal atmosphere. The measured drain–source current decreases rapidly with time after exposure to NH_3 gas and the response characteristics of the drain–source current are seen to be higher for larger values of NH_3 gas concentration. Also, the response characteristics of the OTFT sensor show that there is a shift in the threshold-voltage as well as a change in mobility after exposure to NH_3 gas.

1. Introduction

Organic thin-film transistor (OTFT) devices have been widely investigated for use in such applications as flexible displays, smart cards, memory devices, and sensors [1–6] because of their desirable properties. OTFTs make it possible to create portable, light-weight devices that are thinner than those made with traditional transistors, they also have added advantages of consuming lower power yet they are able to achieve higher performances. Furthermore, devices based on organic semiconductors can be cheaply made because they can be fabricated using low-temperature processing methods and, potentially, with roll-to-roll manufacturing using solution processes [6,7]. The device performance of OTFTs based on such organic semiconductors as P3HT is comparable to that of hydrogenated amorphous silicon thin-film transistors (a-Si:H TFTs) [8,9]. P3HT used as conductive polymers is of particular interest due to their high electrical conductivities, and their large number of possible chemical variants. Also, P3HT-based OTFTs are of particular interest due to their self-organizing properties in forming microcrystalline structures, very stable p-type polymer semiconductors and having the highest field-effect mobilities among the soluble organic semiconductors. Many research groups have reported the high field-effect mobilities of 0.1 $cm^2\,V^{-1}\,s^{-1}$ using P3HT through the optimization of device fabricating conditions, such as choosing appropriate solvents, dielectric treatments, and deposition methods [10]. It has been reported in particular that the organic solvent used in the semiconductor coating process has a significant influence on the film morphology, the crystal structure of the organic semiconductors, and also on their resulting electrical properties [11]. Previous research has reported the effect of ammonia gas on the conductivity and field-effect mobility of the P3HT-based OTFT as a gas sensor [12]. The P3HT-based OTFT sensors have been shown to provide good sensing responses to a range of gases, with sensitivities in the low-ppm levels, good repeatabilities, and stable operation in an air environment [13]. In view of these advantages, gas sensors based on OTFTs are currently a topic of major interest among researchers involved in developing gas sensors for various applications; these include areas where the detection of impurities in gases used for semiconductor manufacturing as well as medical and food science applications are required. Several properties of organic semiconductors, such as their capacitances and resistivities, change in response to the presence of gases and volatile organic compounds [14] and so a number of OTFT parameters, such as the saturation-current (I_{sc}), the off-current (I_{off}), the threshold-voltage (V_{th}), and the subthreshold-slope (SS), could serve as the basis for realizing multi-parametric gas sensors [15–23].

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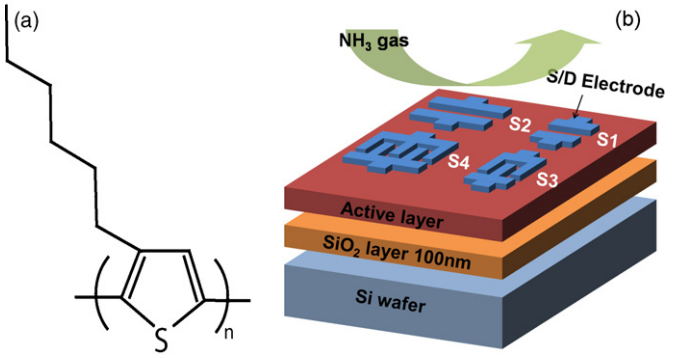

Fig. 1. (a) The molecular structure of P3HT and (b) schematic illustration for the fabrication of bottom-gated top-contacting OTFTs.

This paper describes the fabrication of a gas sensor based on OTFTs obtained from spin-coated P3HT on a thermally grown SiO_2/Si wafer. Gas sensors based on OTFTs respond to a direct interaction between the impinging gas molecules and the organic semiconductor layer, without the need of any additional sensing element. It is shown that gas sensors based on P3HT OTFTs can effectively monitor for NH_3 gas in the atmosphere.

2. Experiment

Fig. 1 shows (a) the molecular structure of P3HT and (b) the schematic illustration of bottom-gated, top-contacting P3HT OTFT sensors for four-type electrodes, such as channel widths of 500 μm (S1), 2 mm (S2), 7.8 mm (S3) and 13 mm (S4). The dielectric layer of the OTFT device has been fabricated by thermally growing 100 nm of SiO_2 on heavily p-doped silicon wafers. The P3HT active layer is spin-coated on a thermally grown SiO_2/Si wafer at a spin rate of 4000 rpm/min for 30 s. Here, the polymer binder is used, from a solution of P3HT (Sigma–Aldrich Co.) in a toluene solvent (1%, w/w). The spin-coated P3HT active layer is then annealed, using a hotplate at 110 °C, for 4 min. Finally, 150-nm-thick gold electrodes, used as the source and the drain, are deposited through a shadow mask by thermal evaporation (DOV Co., Ltd.) under a vacuum of 2×10^{-6} Torr at room temperature. The OTFT sensors are fabricated with varying channel widths from 500 μm to 13 mm and the morphology of the spin-coated P3HT OTFT sensors have been analyzed using scanning electron microscopy (SEM, Hitachi S-4300) at 15 kV and with an atomic force microscope (AFM, XE-100 system). Once the devices have been fabricated, in order to evaluate the electrical performances of the transistors, all the current–voltage (I–V) characteristics of the OTFT devices have been measured using the semiconductor characterization system (Keithley SCS-4200) in a dark box at room temperature. To evaluate the response characteristics to NH_3 gas, the OTFT devices have been placed into a sealed chamber without vacuum pumping in an air environment and NH_3 gas is introduced into a sealed chamber through a mass flow controller (MFC). The gas response characteristics of the OTFT sensors are measured via the changes in the drain–source current, as a function of time, for the OTFT sensors with different channel widths as they are exposed to the NH_3 gas at room temperature.

3. Results and discussion

As shown in Fig. 1(b), the simple bottom-gated, top-contacting OTFT devices are fabricated using P3HT thin films and this structure has generally been used for OTFT devices because it provides much better performance than the bottom-contacting structure [24,25]. However, it is difficult to perform the drain–source patterning for the bottom-gated, top-contacting structures by using photolithography because the formation by metal deposition through the shadow masks results in large size devices with channel lengths/widths of around a few hundred microns.

Fig. 2 shows (a) a typical two-dimensional SEM image and (b) an AFM image of the surface morphology of the P3HT film deposited on the p-type Si wafer with the spin-coating method. It can be seen from the figure that the surface morphology of the spin-coated thin film is dense and crack-free. The grains are round in shape and uniform in size, ranging from 1 to 2 μm, and the inset optical image shows the interdigitated drain–source electrode having channel length and width of 100 μm and 13 mm, respectively. As can be seen in Fig. 2(b), the AFM image demonstrates that the large crystals of the P3HT film, with a surface roughness of 150 nm, are grown directly on the SiO_2/Si wafer so that the surface of the spin-coated P3HT film is both homogeneous and smooth. Generally, the sensing performance of organic thin films is influenced by various factors, including the film composition, the organic semiconductor (OSC) molecular structures, the degree of crystallization, the surface morphology and the grain boundaries. Also, it is well known that the grain boundaries in particular play an important role in realizing effective tin oxide gas sensors, and it has been reported that the grain boundaries are also critical in organic thin films [26].

The OTFTs operate in the accumulation mode, since positive carriers are generated in the P3HT organic semiconductor layer when a negative gate voltage is applied. As shown in Fig. 3(b), the OTFT exhibits a clear p-channel transistor behavior. The drain current increases linearly with an increase in drain voltage in the low-drain-voltage regime, whereas it tends to saturate in the higher drain-voltage regime because of the pinch-off of the accumulation layer. Here, several parameters of the OTFTs, when exposed to NH_3 gas, can be greatly influenced by their interaction with the OTFTs active layer, and these can be seen as resulting in a change of the drain–source current, which, in the saturated regime, is given by plotting the $-\sqrt{I_{DS}}$ versus V_G, and fitting the data to the following

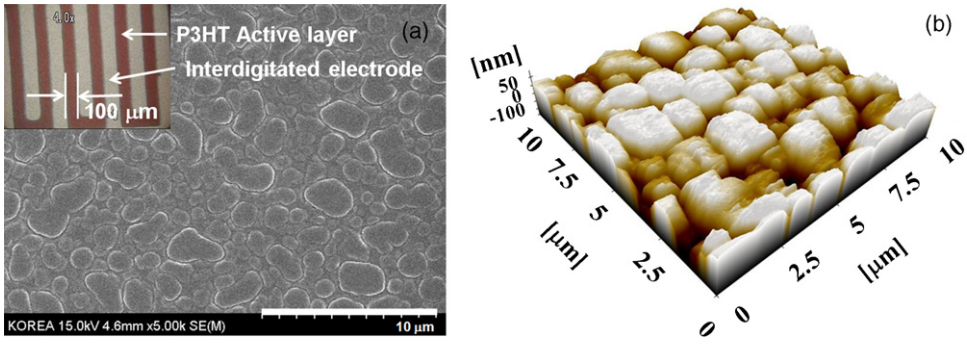

Fig. 2. (a) A typical two-dimensional SEM image and (b) AFM image for the surface morphology of the P3HT film deposited on the p-type Si wafer by the spin-coating method. The inset optical image shows the interdigitated drain–source electrode. The channel length and widths are 100 μm and 13 mm, respectively.

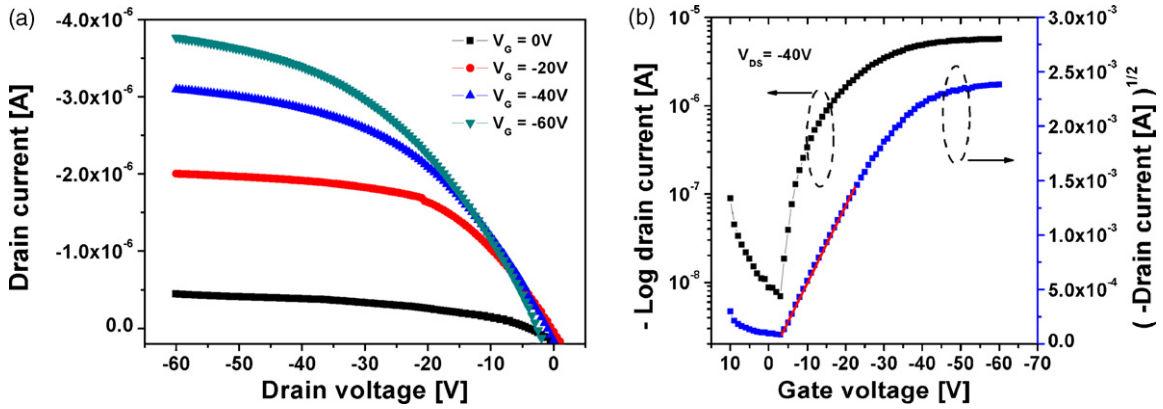

Fig. 3. The characteristics of (a) output curve (I_D–V_D) and (b) transfer curve ($\log_{10}(I_D)$–V_G and $-\sqrt{I_D}$–V_G) for an OTFT with a P3HT film deposited on a thermally grown SiO_2/Si wafer.

equation:

$$I_d = \frac{W\mu C_i}{2L}(V_g - V_{th})^2 \quad (1)$$

where C_i (3.45×10^{-8} F cm^{-2}) is the capacitance of the dielectric layer, W = 13 mm, L = 100 μm, and V_{th} is the threshold voltage. From the transfer characteristics curves of standard OTFT in Fig. 3(b) and using Eq. (1), the field-effect mobility μ has been calculated to be 5.5×10^{-3} cm^2 V^{-1} s^{-1}, when the threshold-voltage V_{th} is about −3 V. The on/off ratio and subthreshold-slope SS is above 0.82×10^3 and 9.1 V/dec when V_G is scanned from −60 to +10 V, respectively. Generally, high field-effect mobilities of above 0.1 cm^2 V^{-1} s^{-1} of P3HT-based OTFT have been demonstrated through the optimization of device preparation by choosing appropriate solvents, such as chloroform, thiophene, xylene, cyclohexylbenzene (CHB), 1,2,4-trichlorobenzene (TCB), toluene and chlorobenzene. As a result, the field-effect mobilities obtained here are lower than previously reported values [10,11] but it is expected that higher field-effect mobilities can be realized through the optimization of the device preparation by choosing appropriate solvents as previously mentioned. All the electrical measurements of the OTFT sensors have been performed at room temperature in an air environment.

Fig. 4(a) displays the change in the drain–source current as a function of time when the P3HT-based OTFT sensors of different channel widths are exposed to various exposure/evacuation cycles of NH_3 in concentrations ranging from 10 to 100 ppm, performed at room temperature in the atmosphere. The measurements of the time dependence of the drain–source current in the OTFT sensors can be made at a gate voltage of 0 V and a drain–source voltage of −1 V. The channel lengths of the OTFT sensors are fixed at 100 μm, and the channel widths are varied from 500 μm to 13 mm. The measured drain–source current characteristics for different channel widths are seen to be attained more rapidly at higher NH_3 gas concentrations. Also, the drain–source currents are higher for larger values of NH_3 concentrations and large channel widths of the OTFTs. When 100 ppm NH_3 gas is introduced to the measurement chamber, the drain–source current of the OTFT sensor, with a channel width of 13 mm, is changed from −0.263 to −0.181 μA for 200 s. After that, recovery is obtained by degassing the chamber for 300 s in an air environment. The OTFT sensors demonstrated fast linear responses, and are able to recover completely to their original baseline when the OTFT sensors were removed from the NH_3 gas. It can be clearly observed that the drain–source currents decrease due to an increase in NH_3 concentration and the response of the OTFT sensors can be calculated from the graph curve of Fig. 4(a). The relative drain–source current change, $\Delta I_{DS} = I_{DS}/I_0$ is plotted against time for each concentration of the NH_3 gas. Here, I_{DS} is the drain–source current, I_0 is the baseline drain–source current before being exposed to the NH_3 gas. When 10, 25, 50, 75 and 100 ppm of NH_3 gas is injected in the OTFT sensors, for a channel width of 13 mm, it can be observed that the response of the sensor is changed by 0.14, 0.17, 0.22, 0.27, and 0.31, respectively. Fig. 4(b) shows the response characteristics for the P3HT-based OTFT sensors of different channel widths versus different concentration levels of NH_3 gas. As shown in Fig. 4(a), the variational rate of the drain–source current increases due to the increasing concentration of NH_3 gas and

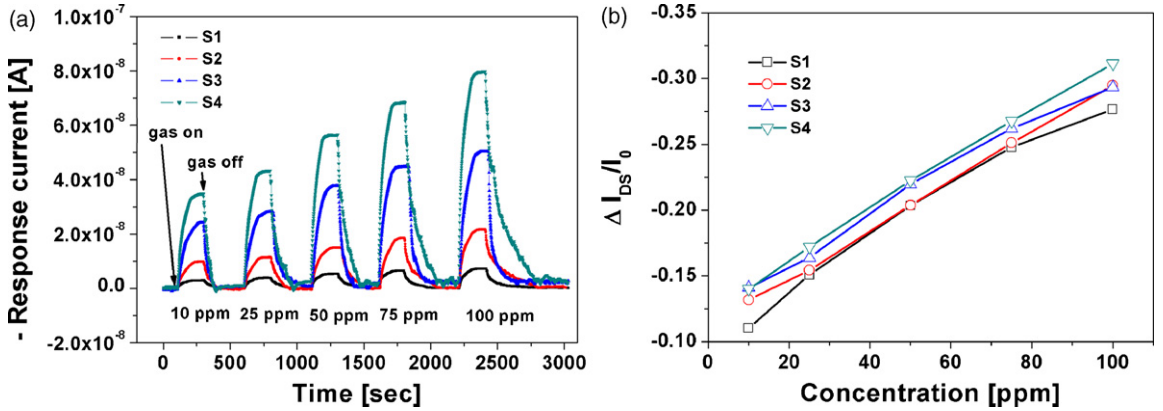

Fig. 4. (a) The change in drain–source current as a function of time when P3HT-based OTFTs sensors of different channel widths are exposed to cycles of exposure and evacuation to NH_3 concentrations ranging from 10 to 100 ppm at room temperature in normal atmosphere. OTFTs sensors are measured for their time dependence of the drain–source current at a gate voltage of 0 V and a drain–source voltage of −1 V. The channel length of the OTFTs devices is fixed at 100 μm and the channel widths are changed from 500 μm to 13 mm. (b) The response characteristics for the P3HT-based OTFTs of different channel widths versus different concentration levels of NH_3 gas.

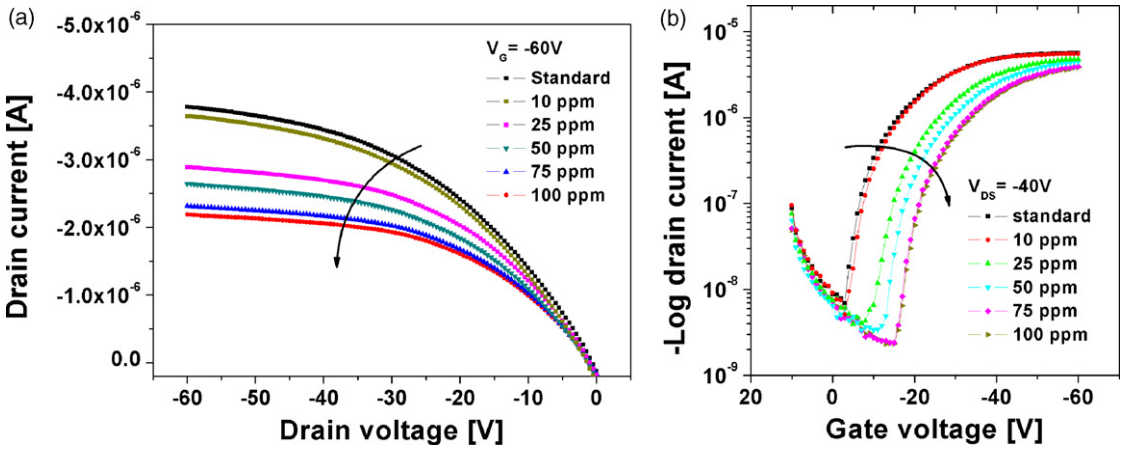

Fig. 5. (a) The output characteristics (I_D–V_D) and (b) the transfer characteristics ($\log_{10}(I_D)$–V_G) of the OTFT sensor exposed to NH_3 gas in concentrations ranging from 10 to 100 ppm, compared with those not exposed to the gas.

the channel widths of the OTFT sensors but the response characteristics also increase with the channel width of the OTFT sensors, as shown in Fig. 4(b). This induces the response characteristics due to the interdigitated electrode structure of the OTFT sensors. As shown in Fig. 4(b), among the OTFT sensors with different channel widths exposed to NH_3 gas of a fixed concentration, those with long channel widths show higher response characteristics than those with shorter channel widths.

Fig. 5 shows (a) the output characteristics (I_D–V_D) and (b) the transfer characteristics (I_D–V_G) of the OTFT sensor (S4) exposed to NH_3 gas in concentration ranging from 10 to 100 ppm, compared with those not exposed to the gas. From the plots shown in Fig. 5(a), it can be noted that the output drain–source current, I_{DS}, decreases in the presence of NH_3 when compared to the output current obtained without NH_3 gas. Here, the decrease in the drain–source current can be explained by a change in the carrier transfer, due to the adsorption of NH_3 molecules on the P3HT film. Molecules adsorbed on the active layer or sites in between gain boundaries of the P3HT thin film produce a larger carrier density leading to drain–source current decreases. The presence of polar molecules is known to change the rate of charge transportation in organic materials by increasing the amount of energetic disorder through charge–dipole interactions [22,23,26,27]. The V_{th} of the OTFT sensor is thus gradually shifted in the negative direction in comparison to the V_{th} of standard OTFT sensors without exposure to NH_3 gas, as shown in Fig. 5(b) (where the sensors are exposed to NH_3 gas in concentrations ranging from 10 to 100 ppm). The shift direction in V_{th} can be explained by the interactions between the dipolar molecules adsorbed on the active layer or sites in between gain boundaries of the P3HT thin. Generally, the negative shifts in V_{th} are induced due to dipolar molecules such as methanol, acetone and methylene chloride which interact with organic semiconductors through Van der Waals interactions. Here, polar molecules behave as acceptor-like deep trap states for the charge carriers moving at the interface between the organic semiconductor layer and the insulator [28,29]. Therefore, a shift in V_{th} is related to the

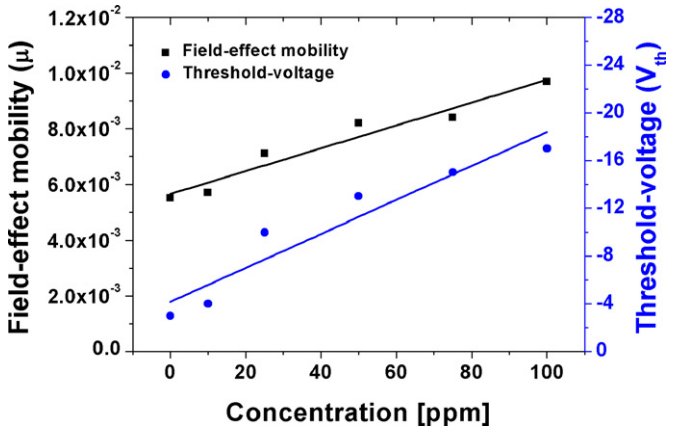

Fig. 6. The variations in the field-effect mobility and the threshold voltage of OTFT sensor during the NH_3 gas exposure in concentration ranging from 0 to 100 ppm.

change of the work function at the interface between the organic semiconductor layer and the insulator, induced by the adsorption of the polar molecules. The shift in V_{th} can be estimated according to Poisson's equation, the result of which is given by [30]:

$$\Delta V_{th} = \frac{-N\mu_I}{\varepsilon} \quad (2)$$

where N is the density of the adsorption site, μ_I the number of dipole moments, and ε is the permittivity of the organic layer.

Fig. 6 shows the variations in the field-effect mobility and the threshold-voltage of OTFT sensors (S4) under NH_3 gas exposure in concentrations ranging from 0 to 100 ppm. As can be seen in Figs. 4 and 5, when the sensor is exposed to the gas, the drain–source current decreases rapidly, and when the gas supply is turned off the drain–source current increases sharply. The measured drain–source current changes more rapidly at a higher concentration of the NH_3 gas. Moreover, the V_{th} of the OTFT sensor

Table 1
Comparison of the electrical characteristics for the field-effect mobility, the threshold-voltage, the on/off current rations, and the sub-threshold swings, when the OTFT sensor is exposed to NH_3 gas in concentrations ranging from 0 to 100 ppm.

NH_3 concentration (ppm)	Field-effect mobility (μ)	Threshold-voltage (V_{th})	On/off ratio	Subthreshold-slope (SS)
Standard	5.5×10^{-3}	−3	0.82×10^{-3}	9.10
10	5.7×10^{-3}	−4	0.11×10^{-4}	8.70
25	7.1×10^{-3}	−10	0.26×10^{-4}	4.90
50	8.2×10^{-3}	−13	0.13×10^{-4}	3.02
75	8.4×10^{-3}	−15	0.16×10^{-4}	1.22
100	9.7×10^{-3}	−17	0.16×10^{-4}	1.2

shifts further toward the negative direction at higher concentrations of NH_3. The field-effect mobility increases gradually, when OTFT sensors are exposed to NH_3 compared with those not exposed to the gas. When the OTFTs are removed from the NH_3 gas, the drain–source current and field-effect mobilities return to their original values and the parameters for all the devices, after exposure to NH_3 gas, are summarized in Table 1.

Since it is easy to fabricate OTFT-based gas sensors on a variety of substrates using defined electrodes, arrays of multi-parametric gas sensors can be designed to detect various gases simultaneously. Moreover, the devices have the potential advantages of structural flexibility, low cost, and versatility in the sensor technologies used. Such sensors can also be considered for other applications, such as wearable electronics, e-textiles, and e-skins for robots.

4. Conclusions

Gas sensors have been fabricated based on OTFTs made from spin-coated P3HT on thermally grown SiO_2/Si wafers. These sensors provide a direct interaction between the impinging gas molecules and the organic semiconductor layer without the need for any additional sensing elements. In studying these devices, the time dependence of the drain–source current has been derived from the *I–V* curves of P3HT-based OTFT sensors of different channel widths exposed to NH_3 gas at concentration levels of up to 100 ppm. The drain–source current of the OTFT sensors has also been observed, as a function of time, when exposed to cycles of NH_3 gas sensing and recovery for different NH_3 concentrations ranging from 10 to 100 ppm and the drain–source currents are seen to change more rapidly at higher concentrations. Moreover, the response characteristics of the drain–source current are higher for larger values of NH_3 gas concentrations. Also, the response characteristics of the OTFT sensor, when exposed to NH_3 gas in concentrations ranging from 0 to 100 ppm, have shown results in the threshold-voltages being shifted and an increase in the field-effect mobilities. These results demonstrate that gas sensors based on P3HT OTFT sensors offer an effective way of monitoring NH_3 gas in the atmosphere.

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