Investigation of the Performance of Donor–Acceptor Conjugated Polymers in Electrolyte-Gated Organic Field-Effect Transistors

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Electrolyte-gated organic field-effect transistors (EGOFETs) are gaining interest for application in bioelectronic devices. However, robust performance in terms of charge-carrier mobility, on-to-off drain current ratio \( I_{\text{on}}/I_{\text{off}} \), and turn-on speed are required for real application. Here, donor-acceptor (D-A) conjugated polymers, namely poly[2,5-(2-octyldodecyl)-3,6-diketopyrrolopyrrole-alt-5,5-(2,5-di(thien-2-yl)thieno[3,2-b]thiophene)] (PDPPDTT) and indacenodithiophene-co-benzothiadiazole (PIDTBT), are evaluated in EGOFETs. The operational performance of these materials is compared to that of the well-established conjugated polymer, poly[2,5-bis(3-hexadecylthiophen-2-yl)thieno[3,2-b]thiophene] (PBTTT).

1. Introduction

Electrolyte-gated organic field-effect transistors (EGOFETs) are low voltage electronic devices that utilize an electrolyte as the gate insulator.[1] The electrical double layers (EDLs) established at the interface of the electrolyte and the organic semiconductor (OSC)/gate terminal lead to a high capacitance (~\(4 \mu \text{F cm}^{-2}\)) that enables low voltage operation (i.e., \(\leq 1 \text{ V}\)) of these devices.[3] The transduction of voltage to current efficacy, in EGOFET, partly depends on the capacitive coupling of the electrolyte with the OSC and the gate electrode. The coupling must be electrostatic to guarantee charge transport under dominant field-effect mode. This differentiates the EGOFETs from the organic electrochemical transistor (OECT).[3] as the OSC must be impermeable to ions, and chemically as well as structurally stable when in direct contact with the electrolyte. However, the design guidelines for the active layer of EGOFET are still unclear although it appears that a high degree of crystallinity and interdigitated side chains are important to impede the uptake of ions to deliver improved device performance.[3] Small molecules and liquid crystalline polymeric semiconductors have been used as the active layer of EGOFETs for a variety of bioelectronic applications including sensing, triggering and mimicking biological or chemical phenomena.[6–8]

Semiconducting polymers with donor–acceptor moieties in the backbone (D–A polymers) are attracting great interest as they display high field-effect mobility in OFETs (>1 cm² V⁻¹ s⁻¹) with ideal operating characteristics.[9–15] These materials, while having less crystalline microstructures, outperform semicrystalline polymers (such as regioregular poly(3-hexylthiophene) (P3HT) or poly[2,5-bis(3-hexadecylthiophen-2-yl)thieno[3,2-b]thiophene] (PBTTT)). The rapid charge transport is ascribed to the enhanced polymer backbone planarity, the decoupling of energetic and side-chain disorder, and shorter intermolecular π–π stacking distances.[10,14] In addition, the operational stability of the materials is also improved and OFETs derived from thiophene–thiazole D–A polymers show comparable bias stress stability (excluding off-bias driven instability) to that of amorphous silicon thin-film transistors.[9,16–18] The deep highest
occupied molecular orbital (HOMO) energy levels (≤−5.1 eV) of D–A polymers reduce the likelihood of oxidation of the intrinsic polymers in use or on extended storage.[10,14,18] These characteristic performances may be advantageous in EGOFETs.

Here, the performance of EGOFETs based on two D–A polymers with a planar conjugated backbone (poly[2,5-(2-octyldodecyl)-3,6-diketopyrrolopyrrole-alt-5,5-(2,5-di(thien-2-yl)thieno [3,2-b]thiophene)] (PDPPDTT) and indaceno-dithiophene-co-benzothiadiazole (PIDTBT) were evaluated. Normalised transconductance (μ × C), on-to-off drain current ratio \( I_{on}/I_{off} \), subthreshold swing (SS), and switch-on time of these materials were determined and compared against that of well-established PBTTT. Both materials performed well with state-of-the-art electrical figures-of-merit. The mobility extracted for PDPPDTT and PIDTBT devices was almost double that of the PBTTT based device and outclassed most of what had been reported to date in EGOFETs. The \( I_{on}/I_{off} \) was one (PDPPDTT) and two (PIDTBT) orders of magnitude higher than that of PBTTT and compared favorably to the highest performing EGOFETs presented in the literature. Furthermore, the EGOFETs based on the D–A polymers turn on at two to ten times faster than the PBTTT device with improved subthreshold swings.

2. Results and Discussion

An array of sixteen bottom contact EGOFETs were fabricated on polyethylene naphthalate (PEN) substrates (Figure 1a) following procedures detailed in the Experimental Section. Figure 1b shows a schematic of an individual device consisting of an interdigitated gold source/drain electrode patterned on the PEN and spin-coated with the OSC, which was either PBTTT, PDPPDTT, or PIDTBT (Figure 1c,d). Circular wells for the aqueous electrolyte were formed by affixing a laser-patterned double-sided tape (DST) to this substrate and the device completed by filling the cell with de-ionised water (Millipore Milli-Q system, 18.2 MΩ cm, 6 μL) and inserting a gold wire (diameter of 1 mm) to act as the gate electrode (Figure 1c). The DST was used to confine the electrolyte over the channel area and define the relative position of the gate electrode to conducting channel. This is important for accurate comparison of performance between devices as the position of the gate electrode relative to the OSC affected the electrical performance of the EGOFET (Figure S1, Supporting information).

2.1. Surface Morphology of PBTTT, PDPPDTT, and PIDTBT by AFM

The morphology of the thin films of PBTTT, PDPPDTT, and PIDTBT deposited on the contact electrodes was assessed by atomic force microscopy (AFM) in tapping mode. AFM height micrographs of these thin films are presented in Figure 2. The surface morphologies of the three polymers films were different. PIDTBT film showed gross surface features leading to a root-mean-square (RMS) of 8.8 nm. In contrast, PBTTT and PDPPDTT showed a uniform surface topography with a network of structured aggregates that were homogeneously distributed over 10 μm² and an RMS roughness of 3 and 2.6 nm, respectively. Differences in the size and shape of the aggregates in PBTTT and PDPPDTT film were observed. In PDPDDTT, the aggregates appear as nanofibers. Similar morphology was recently observed by Y. Xi and co-workers for this polymer.[19] However, the authors deposited PDPPDTT film from a mixture of chloroform with methanol at different methanol loading (10%, 15%, and 20%). They suggested that the mixture of polar poor solvent (methanol) with chloroform (good solvent for dissolving PDPPDTT) was the reason for PDPPDTT self-organisation into nanofibers.[19] Here, the nanofiber formation was probably due to the high concentration of PDPPDTT (7 mg mL⁻¹) in a high boiling point solvent (1,2-dichlorobenzene (DCB)). The slow evaporation of the solvent allows the self-organized and homogeneously distributed nanofiber morphology seen in Figure 2b to develop. For PBTTT, the interconnected aggregates resemble the fibrillary crystalline structure observed by Paterno et al. in slowly evaporating (12 h) drop casted PBTTT film deposited from hot orthodichlorobenzene solution.[20] The average peak to valleys volume of the fibrillar aggregates in PBTTT is two times larger than that of the nanofibers seen in the PDPPDTT film. Previous reports have shown that PBTTT films have more and intense grazing incidence X-ray diffraction peaks and a large proportion of the volume of crystallite than diketopyrrolopyrrole and indaceno-dithiophene based D–A conjugated polymers.[21]
2.2. EGOFETs Characteristics of D–A Conjugated Polymers and PBTTT

The transfer and output characteristics of EGOFETs based on PBTTT, PDPPDTT, and PIDTBT semiconductors are presented in Figure 3. The electrolyte used was de-ionised water (Millipore Milli-Q system, resistivity of 18.2 MΩ cm). The three devices display p-channel response with identifiable linear, pinch-off, and saturation regions (Figure 3b). No deviation from linearity at low drain voltage was observed in the output curves for all three materials. Hence, no evidence of charge injection problems was noticeable in these devices. The transfer curves of all the fabricated devices show small drain current hysteresis (Figure 3a; Figure S2, Supporting Information). This indicates, that the coupling of the gate electrode with the conducting channel was essentially electrostatic. Besides the small drain hysteresis, it can be perceived that the trends of the gate current ($I_{GS}$), which is a combination of ionic (dominant) and electronic current, differ from that of the transfer characteristics (Figure S2, Supporting Information). Also, $I_{GS}$ was ≈ two orders of magnitude lower than the on-state drain current ($I_{DS}$) of the corresponding device at maximum gate amplitude. Hence, the drain current modulations of water-gated devices based on the three polymers were not influenced by the gate current. Yet, despite the channel and gate terminals superposition and that the geometry for all the fabricated devices and the electrolyte employed were identical, the magnitudes of $I_{GS}$ recorded at −0.5 V gate bias differ. $I_{GS}$ recorded with the PBTTT based device was three times higher than those of PDPPDTT and PIDTBT, which were in a similar range (<3.5 nA). The small drain hysteresis and $I_{GS}$ observed in these devices indicates that almost no electrochemical reaction occurred, so they operated in a dominant field–effect charge transport mode.

Devices fabricated using PDPPDTT displayed the best transduction of voltage to current within the chosen characterisation window (0 V ≥ $V_{SD}$ ≥ −0.5 V; 0.35 ≥ $V_{GS}$ ≥ −0.5 V). The output characteristics of the PDPPDTT device, in contrast to those of the two other polymers, show full saturation at all gate biases applied (Figure 3b). Equally, the square root of the drain current as a function of gate bias is consistent with an idealised field–effect transistor characteristic (quadratic current dependence on gate bias) in comparison to that of PBTTT and PIDTBT (Figure 3a).

To quantitively analyze the electrical characteristics of the developed devices, commonly adopted fitting and extraction methods in the saturation regime were used. Among the polymers investigated, PBTTT displays the highest $I_{DS}$ (~7 µA), which was almost twice that of PDPPDTT based devices (~3.6 µA) and more than an order of magnitude superior to the drain current of PIDTBT (~0.21 µA) at a gate bias of −0.5 V (Table 1). The low $I_{DS}$ observed in PIDTBT EGOFET can be attributed to the fact that the voltage necessary to turn the device on is higher than that of the two other polymers. The high threshold voltage ($V_{th}$) for PIDTBT should originate from the energetic difference between the surface potential of PIDTBT and that of the gate terminal ($E_F$–$E_{HOMO}$). As shown in Figure 3c, the HOMO energy level of PBTTT (~5.1 eV) and PDPPDTT (~5.2 eV) are closer to the work function of the gold gate terminal (~−4.9 to −5.1 eV) in comparison to that of PIDTBT (~−5.3 eV). As PIDTBT has the deepest HOMO level the devices fabricated using this polymer should have the highest $V_{th}$ and this is observed. However, the $V_{th}$ of devices fabricated using polymers PBTTT to PDPPDTT and PIDTBT does not scale linearly with the energetic difference, $E_F$–$E_{HOMO}$, as expected.

While EGOFETs based on PBTTT display high $I_{on}$, they have been reported to exhibit a large off-state current ($I_{off}$) of about a few tens of nano-amperes when an electrolyte is used as a gate insulator. Here, it was found to be ~50 nA for PBTTT, which is up to three orders of magnitude higher than that recorded for PDPPDTT (~1 nA), and PIDTBT (~12 pA) devices, respectively (Table 1). The high $I_{off}$ degrades the $I_{on}/I_{off}$ of PBTTT based EGOFET, which as presented here (2.1 ± 0.7 × 10$^2$) as in most
of the reported literature is in the magnitude of $10^2$. This ratio is much lower than that displayed by EGOFETs of PDP-PDTT ($3 \pm 1 \times 10^3$) and PIDTBT ($2.3 \pm 0.1 \times 10^4$) (Table 1). Furthermore, the SS of the D–A polymers PDPPDTT and PIDTBT were 125 mV dec$^{-1}$ and 62 mV dec$^{-1}$, respectively. These values were significantly lower than that of PBTTT (456 mV dec$^{-1}$) reported in this work and other literature (Table 1). However, the product of mobility and capacitance ($\mu \times C$) of PBTTT was double that of PDPPDTT and PIDTBT (Table 1). These values are in the same range as the majority of $\mu \times C$ figures (≈10$^{-2}$ μF V$^{-1}$ s$^{-1}$) found in the literature when water is the electrolyte (Figure 4a; Table S1, Supporting Information). However, the range is large as the highest and lowest $\mu \times C$ figures reported to date are 0.6 μF V$^{-1}$ s$^{-1}$ and 0.0003 μF V$^{-1}$ s$^{-1}$, respectively (Figure 4a; Table S1, Supporting Information).

To understand the high transconductance displayed by the PBTTT as compared to the D–A polymers, the capacitance and

Table 1. Summary of electrical figures of merit obtained from EGOFET.

|                | PBTTT       | PDPPDTT     | PIDTBT      |
|----------------|-------------|-------------|-------------|
| $\mu \times C$ [μF (Vs)$^{-1}$] | 0.05 ± 0.02 | 0.02 ± 0.01 | 0.013 ± 0.003 |
| $\mu$ [cm$^2$ (Vs)$^{-1}$] | 0.10        | 0.18        | 0.16        |
| $C$ [μF cm$^{-2}$] | 0.49 ± 0.06 | 0.11 ± 0.01 | 0.08 ± 0.02 |
| $I_{on}/I_{off} \times 10^3$ | 0.21 ± 0.07 | 3 ± 1       | 23 ± 1      |
| $I_{on}$ [μA] | 6.9 ± 4.0   | 3.6 ± 1.7   | 0.21 ± 0.03 |
| $I_{off}$ [nA] | 48 ± 1      | 1.1 ± 0.2   | 0.012 ± 0.006 |
| SS [mV dec$^{-1}$] | 456         | 125         | 62          |
| $V_{ds}$ [V]     | 0.02 ± 0.03 | −0.05 ± 0.03 | −0.36 ± 0.01 |
| $\tau_{on}$ [s] | 2.8 ± 0.3   | 1.2 ± 0.6   | 0.18 ± 0.02 |

Figure 3. Typical a) transfer and b) output characteristic of electrolyte gated organic field-effect transistor based on PBTTT, PDPPDTT, and PIDTBT. The electrolyte employed was de-ionised water purified with Milli-Q system (resistivity of 18 MΩ cm). c) Schematic representation showing how the Fermi level of the Au gate electrode ($E_F$ (Au) = −4.9 eV) compares to the frontier orbitals (HOMO and LUMO) of PBTTT, PDPPDTT, and PIDTBT.
mobility were decoupled. Usually, this is carried out by determining the capacitance of the EDL via techniques such as cyclic voltammetry and electron impedance spectroscopy measurement.\[27,28\] Here, an alternative method is used. The capacitance of the EDL \((C)\) at the interface between an electrolyte and an electrode is voltage-dependent.\[29\] It quantifies the change in surface charge \((\sigma)\) near the electrodes with the change in electric potential \((\phi_e)\). And it can be written as a differential capacitance:

\[ C = \frac{d\sigma}{d\phi_e} \]  

(1)

by the introduction of a derivative of time, Equation (1) becomes:

\[ C = \frac{d\sigma}{dt} \times \frac{dt}{d\phi_e} \]  

(2)

Considering that a variation of charge with time is a current \((I)\), Equation (2) can be rewritten as:

\[ I = C \times \frac{d\phi_e}{dt} \]  

(3)

In an EGOFET, the electrolyte separates the conducting channel and the gate terminal. Hence, the current in Equation (3) is the \(I_{GS}\) of EGOFET. Because of the low hysteresis seen in the transfer curves and the small recorded \(I_{GS}\), the gate leakage can be neglected, and therefore “1” is the displacement current. Hence, the effective capacitance of the EDL is simply the slope of the displacement current as a function of gate sweep rate. Initially, the \(I_{CS}\) versus \(V_{CS}\) curves were recorded at different gate bias sweep rates (See Figure S3a, Supporting Information). Then, the peak currents \((I_{CS})\) were extracted and plotted against the gate sweep rate (See Figure S3b, Supporting Information) to calculate the capacitance. The peak gate current, for example, \(I_{CS}\) at a gate bias of \(-0.5\, \text{V}\), was used as it corresponds to the current at which the capacitance is close to the steady-state value. The capacitance of the EDL was extracted by linear fitting of data points. Dividing this value by the source/drain electrode area gives the capacitance per unit area of the EDL in the fabricated EGOFETs. The effective mobility and capacitance of all the materials obtained by this method are summarised in Table 1. The calculated capacitances range from \(0.49 \pm 0.06\, \mu\text{F cm}^{-2}\) for PBTTT, \(0.11 \pm 0.01\, \mu\text{F cm}^{-2}\) for PDPPDTT, and \(0.08 \pm 0.02\, \mu\text{F cm}^{-2}\) for PIDTBT. The difference in these values demonstrates that the capacitance of an EGOFET device depends on the OSC employed as the conducting channel. This is consistent with previous studies, as the reported effective capacitances in EGOFET, when water is the electrolyte, range from \(0.08\, \mu\text{F cm}^{-2}\) for PNDIT2 to \(5.3\, \mu\text{F cm}^{-2}\) for diF-TES-ADT (Table S1, Supporting Information). The dispersion may originate from the difference in the OSC structure, and it seems that the values are lower for less crystalline OSC films (Table S1, Supporting Information). Given the calculated capacitances, it appears that the high \(\mu \times C\) of PBTTT originated from the higher capacitance rather than a high mobility of the semiconductor as the extracted mobility of PBTTT was similar to that reported in the literature \((0.10\, \text{cm}^2\, \text{V}^{-1}\, \text{s}^{-1})\). This value is almost two times lower than that of PDPPDTT and PIDTBT, which were \(0.18\) and \(0.16\, \text{cm}^2\, \text{V}^{-1}\, \text{s}^{-1}\), respectively.

The D–A polymer-based devices presented in this work are amongst the highest performing water-gated EGOFET reported to date.\[3,6,27,30–35\] Figure 4b shows a comparison of the mobility and \(I_{on}/I_{off}\) ratios reported for EGOFETs based on pristine OSC and blends of materials against the values obtained in this work. All devices in Figure 4b were fabricated in a bottom-contact top-gate architecture and the electrolyte employed was water. However, the channel geometry, gate electrode, and the bias applied to the terminals may be different. In terms of mobility and \(I_{on}/I_{off}\) (Figure 4b), it is clear that the D–A polymers devices of this work outperform EGOFETs in which the active layer is a pristine molecular or polymeric semiconductor.
and PDPPDTT, decreased after saturation. The increase of $I_{DS}$ reach a maximum value in less than 10 ms (Figure 5a,b,c). They display a sharp increase of $I_{DS}$ followed by a slow increase toward saturation. $I_{DS}$ of PIDTBT based device, in contrast to that of PBTTT and PDPPDTT, decreased after saturation. The increase of $I_{DS}$ can be correlated with the change in the $I_{GS}$. Upon switching the gate bias from 0 V to −0.5 V, the $I_{GS}$ increases sharply to reach a maximum value in less than 10 ms (Figure 5a,b,c). This corresponds to an ionic displacement under the effect of the electric field established between the gate and source-channel-drain. This is followed by an exponential decrease to a steady-state value. The variation in the rate of increase of $I_{DS}$ describes the state of coupling between the majority charge carriers in the semiconductor and ions present in the EDL. One explanation for this behavior is that the EDL capacitance increases with a rate that dictates the response time of the device. Here this time ($\tau_{on}$) depends on the conducting channel employed in the device (Table 1). The times required to reach 90% of the maximum drain current were 2.8, 1.2, and 0.18 s for PBTTT, PDPPDTT, and PIDTBT, respectively.

Due to the fast response to gate bias change, high $I_{on}/I_{off}$ and small SS ones may expect the D–A polymers PDPPDTT and PIDTBT, to be more sensitive to change in gate surface potential when compared to the PBTTT based EGOFET devices.

3. Conclusion

In this work, the electrical performances of three polymeric semiconductors were investigated in the electrolyte-gated transistor configuration. The results show that the planar D–A polymers, namely PDPPDTT and PIDTBT, can be water-gated to produce devices that operate in pure or at least dominant field-effect charge transport mode. The figures of merit of devices based on the two materials were systematically compared against that of an established polymer in the field, PBTTT. Except for the capacitance, the electrical characteristics of the devices, including field-effect mobility, SS, $I_{on}/I_{off}$ and switch-on time for devices based on D–A polymers were superior to those fabricated with PBTTT. Also, in terms of mobility and $I_{on}/I_{off}$, the D–A polymers-based devices reported here outclass most of the highest performing EGOFETs reported to date. These results show that D–A polymers with a planar conjugated backbone enable the development of robust EGOFETs that are well appropriate for applications in bioelectronic and environmental science.

4. Experimental Section

Device Fabrication Procedure—Substrate Preparation: EGOFETs were fabricated in bottom contact top-gate architecture. A 125 µm PEN and a Platfix C325N4A4 adhesive sheet were cut into rectangular pieces of 20 mm × 30 mm with a CO2 laser cutter. Then, a silicon substrate was cut into rectangular pieces of 22 mm × 32 mm with a diamond tip. The silicon carriers were sonicated in acetone and isopropanol (IPA) for 5 min each, rinsed with IPA, and blown dry with a stream of nitrogen. PEN substrates were then laminated, using Bonsen Electronics Ltd. laminator Peach PL714, onto cleaned silicon carriers with the help of the Platfix adhesive.

Contact Electrode Patterning: The substrate fixed on silicon carrier (PEN/Si) was sonicated in IPA (5 min), rinsed with IPA, blown dry with a stream of nitrogen, and heated on a hotplate at 110 °C (5 min) to prepare for photoresist deposition. A solution of PGMI was spun onto
PEN/Si (1500 rpm, 1000 rpm s\(^{-1}\), 60 s), then the substrate was heated up to 150 °C for 5 min to fully dry the produced film and improve its adhesion to the PEN substrate. S1805 was then spun onto PGMI coated substrates (7000 rpm, 1000 rpm s\(^{-1}\), 60 s), followed by heating the substrate on a hotplate at 110 °C for 1 min to remove residual solvent. Contact electrodes of sixteen transistors (W/L: 11.6 mm/10 μm) designed with AutoCAD were patterned by a direct laser lithography machine (Microtech LW405) to expose some parts of the PEN/Si coated S1805/PGMI bilayer photoresist to a 405 nm laser beam. Then, the substrate was immersed in an MF-319 solution, and in water for 60 s each, rinsed with water, and blown dry with a stream of nitrogen to generate the same pattern as the design of the AutoCAD drawing exchange format file. This was followed by thermally evaporating 5 nm of chromium and 40 nm of gold with an Edward Auto 300 in high vacuum (10\(^{-7}\) mbar) at a rate of 0.1 nm s\(^{-1}\). The last step to obtain the electrode pattern onto the PEN/Si consisted of immersing the substrate in acetone for ~15 min of sonication to lift-off. The substrate was then peeled off the silicon carriers, sonicated in acetone and IPA for 15 min in each, rinsed with IPA, blown dry with a stream of nitrogen, and treated by UV/O\(_3\) for 15 min to remove organic contaminants.

**Organic Semiconductor Deposition**: PBTTT (Ossila Ltd. – \(M_w \approx 395\,000\) Da and \(M_n \approx 292\,200\) Da and \(M_w \approx 74\,900\) Da), and PIDTBT (\(M_w \approx 243\,000\) Da and \(M_n \approx 33\,000\) Da), were dissolved in 1,2-dichlorobenzene (heated at 80 °C for 1 h to produce 7 mg mL\(^{-1}\) of solutions of each. The three solutions were deposited on different substrates with patterned contact electrodes with the same spin-coating parameters (1500 rpm, 1000 rpm s\(^{-1}\), 60 s). All the substrates were heated at 140 °C for 1 h to fully dry the films. Double-sided tape (DSP) with circular holes was laser cut and fixed on the DSP. The hole was filled with 6 μL of de-ionised water (Millipore Milli-Q system, resistivity of 18.2 MΩ cm, 6 μL). A gold wire (1 mm width) cleaned with UV/O\(_3\) (15 min), isopropanol and water, serving as the gate electrode, was fixed on the DSP.

**Device Characterization Procedure—Surface Properties Metrology**: Atomic force microscope (Nasoscope V) was used to probe the organic semiconductor films surface morphology in tapping mode.

**Electrical Characterization**: Transfer, output curves, gate current, and transient characteristics, were recorded in the air at ambient conditions for sixteen transistors (W/L: 11.6 mm/10 μm) designed with AutoCAD were patterned by a direct laser lithography machine (Microtech LW405) to expose some parts of the PEN/Si coated S1805/PGMI bilayer photoresist to a 405 nm laser beam. Then, the substrate was immersed in an MF-319 solution, and in water for 60 s each, rinsed with water, and blown dry with a stream of nitrogen to generate the same pattern as the design of the AutoCAD drawing exchange format file. This was followed by thermally evaporating 5 nm of chromium and 40 nm of gold with an Edward Auto 300 in high vacuum (10\(^{-7}\) mbar) at a rate of 0.1 nm s\(^{-1}\). The last step to obtain the electrode pattern onto the PEN/Si consisted of immersing the substrate in acetone for ~15 min of sonication to lift-off. The substrate was then peeled off the silicon carriers, sonicated in acetone and IPA for 15 min in each, rinsed with IPA, blown dry with a stream of nitrogen, and treated by UV/O\(_3\) for 15 min to remove organic contaminants.

**Conflict of interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Keywords**

donor–acceptor conjugated polymers, electrical double layer (EDL), electrolyte-gated organic field-effect transistor (EGOFET), PBTTT, PDPPDTT, PIDTBT

 Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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[1] L. Kergoat, B. Piro, M. Berggren, G. Horowitz, M. C. Pham, Anal. Bioanal. Chem. 2012, 402, 1813.
[2] J. Rivnay, S. Inal, A. Salleo, M. Berggren, G. G. Malliaras, Nat. Rev. Mater. 2018, 3, 17086.
[3] R. Porrazzo, S. Bellani, A. Luzio, E. Lanzarini, M. Caironi, M. R. Antognazza, Org. Electron. 2014, 15, 2126.
[4] A. Doumbia, M. Webb, M. L. Turner, J. Behrendt, R. J. Wilson, in Organic Sensors Bioelectronics X (Eds.: R. Shinar, I. Kymissis, L. Torsi), SPIE, San Diego, CA 2017, p. 21.
[5] J. Le Gall, F. Mouillard, T. N. Le, T. T. Vu, G. Mattana, R. Brayer, S. Zrig, V. Noël, B. Piro, Biosens. Biosens. Bioelectr. 2020, 157, 112166.
[6] C. Sun, Y. X. Wang, M. Sun, Y. Zou, C. Zhang, S. Cheng, W. Hu, Biosens. Bioelectr. 2020, 164, 112251.
[7] G. Forsch, F. Leonardi, A. Scala, F. Biscarini, A. Kovtun, A. Liscio, A. Mazzaglia, S. Casalini, Nanoscale 2015, 7, 20025.
[8] S. Desbie, M. di Lauro, S. Casalini, D. Guerin, S. Tortorella, M. Barbalinardo, A. Kyndiah, M. Murgia, T. Cramer, F. Biscarini, D. Vuillaume, Org. Electron. 2016, 38, 21.
[9] M. Kim, S. U. Ryu, S. A. Park, K. Choi, T. Kim, D. Chung, T. Park, Adv. Funct. Mater. 2020, 30, 1904545.
[10] C. B. Nielsen, M. Turbiez, I. McCulloch, Adv. Mater. 2013, 25, 1859.
[11] J. Tong, A. Doumbia, A. Alieva, M. L. Turner, C. Casagrande, ACS Omega 2019, 4, 11657.
[12] T. Marszalek, M. Li, W. Pisula, Chem. Commun. 2016, 52, 10938.
[13] Y. Yamashita, F. Hinkel, T. Marszalek, W. Zajczkowski, W. Pisula, M. Baumgarten, H. Matsui, K. Mülten, J. Takeya, Chem. Mater. 2016, 28, 420.
[14] X. Zhang, H. Bronstein, A. J. Kronemeijer, J. Smith, Y. Kim, R. J. Kline, L. J. Richter, T. D. Anthopoulos, H. Sirringhaus, K. Song, M. Heeney, W. Zhang, I. McCulloch, D. M. Delongchamp, Nat. Commun. 2013, 4, 2238.
[15] D. Venkateshvaran, M. Nikolka, A. Sadhanala, V. Lemaur, M. Zelazny, M. Kepa, M. Hurhangee, A. J. Kronemeijer, V. Pecunia, I. Nasrallah, I. Romanov, K. Broch, I. McCulloch, D. Emin, Y. Olivier, J. Cornil, D. Beljonne, H. Sirringhaus, Nature 2014, 515, 384.
