An Organic Electrochemical Transistor fabricated on Waxy-Sublimating Substrates

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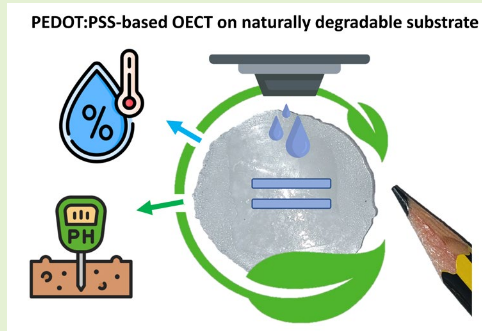
Abstract— **The global demand for zero-waste technologies requires the development of electronic devices with sustainable fabrication processes and (ideally) biodegradable materials and substrates. This article reports on the fabrication and electrical characterization of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS)-based organic electrochemical transistors (OECTs) on naturally degradable cyclododecane (CDD)-based substrates. The electrical performance of the devices is tested in a closed box flushed with nitrogen gas under ambient and different humidity conditions. The devices exhibit an I_{ON}/I_{OFF} of more than 10^3 and transconductance values up to 300 mS. As a proof-of-concept of the potential use of the PEDOT:PSS OECTs for biosensing and environmental monitoring, their application as pH sensors is presented. The sensitivity of 2.25 mS/pH and the signal amplification (up to 50 mS) obtained in this study offer a promising perspective that opens to the formulation of strategies for a more responsible approach to the production and recovery processes of electronic platforms, thus contributing to sustainable technological practices.**



***Index Terms*— transient electronics, organic electrochemical transistors, PEDOT:PSS**

I. Introduction

THE growing need for eco-friendly technologies and waste reduction has directed the interest of the materials science community towards the development of electronic platforms designed to degrade over time and ultimately vanish into the environment after stable operation. The ideal outcome is leaving no toxic residue and eliminating the necessity for recovery after use [1]. Thus, the rational choice of architectures, materials, and fabrication processes is the core of this emerging sustainable zero-waste electronics.

Organic electrochemical transistors (OECTs) are polymer-based devices in which the gate electrode modulates the bulk conductivity of an organic semiconductor through the injection of ions into the film from an electrolyte [2]. Thanks to the double ionic-electronic conducting nature of the materials employed as active layers — that are often also biocompatible — and the easy fabrication processes of the devices, OECTs have served as electronic transducers in a wide range of applications, including chemo-/bio- sensing [3], [4], bioelectronics [5], artificial intelligence [6] and energy harvesting [7]. Poly(3,4-ethylenedioxythiophene) doped with poly(styrene sulfonate) (PEDOT:PSS) is, by far, the most employed organic semiconductor for OECTs [8]. It is characterized by high current and transconductance (g_m) levels [9], easy functionalization, good chemical resistance, and environmental stability, enabling high sensitivity and good selectivity, thus making PEDOT:PSS based OECTs ideal devices to be employed in sensing applications [10].

Among the different sensing possibilities, pH evaluation is one of the most interesting because it is fundamental in several fields like environmental monitoring, medical diagnostics, and food safety [11]. PEDOT:PSS OECT-based pH sensor functionality is related to the variation of the ionic concentration in the organic semiconductor caused by the pH of the analyte [12]. When the hydrogen ion concentration in the PEDOT:PSS film increases, the concentration of carriers decreases and the microstructure of PEDOT:PSS membrane changes from continuous network to non-continuous clusters, resulting in a reduced conductivity of the film [13]. These sensors show benefits over traditional pH sensors, such as increased sensitivity and fast response times [14]. Moreover, being PEDOT:PSS solution-processable, it can be easily deposited using printing techniques [15] on various substrates, from glass and silicon wafers to plastic, paper and textiles to obtain cost-effective, portable and, eventually, disposable pH

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sensing devices [16].

With the goal of developing more sustainable electronics, one of the most interesting substrates are the naturally degradable ones, which decompose spontaneously over time [17]. For example, cyclododecane (CDD), a wax-like non-polar cyclic hydrocarbon solid, exhibits unique chemical, physical, and thermal properties that make it suitable for transient substrate applications [18]. In particular, CDD is chemically stable and inert under normal conditions, can be biodegraded over time, and maintains stability at temperatures below its boiling point (194°C). Here, exploiting the possibility of using the same material —PEDOT:PSS — for both the gate electrode and the active layer of the device, we focus on the fabrication of fully printed PEDOT:PSS-based OECTs on naturally degradable substrates. We explore the performance of these devices under various humidity conditions and as pH sensors in air. The fabricated devices show low-voltage operations, large signal amplification (up to 50 µS) and good sensitivity (2.25 µS/pH), which are the main benchmark parameters for this kind of applications.

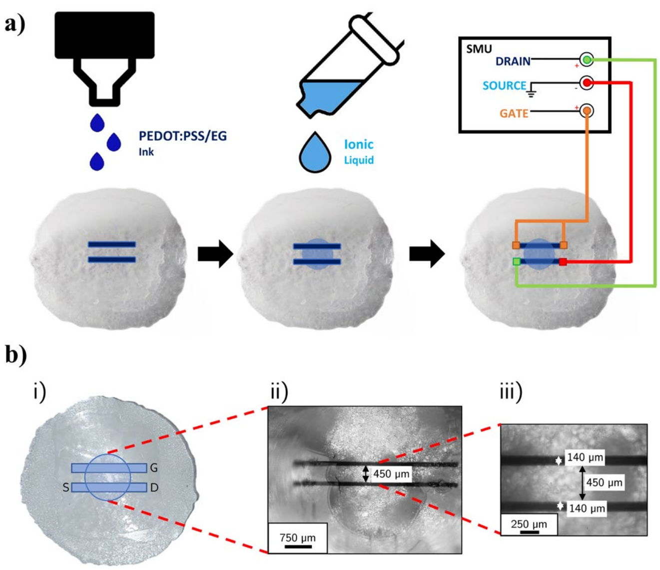

Fig. 1. All inkjet-printed PEDOT:PSS OECT on CCD/PVA/PVP transient substrates. a) Schematic of the printing fabrication steps and 3-electrode setup used to characterize the pH sensing transistor. b) Optical images of the inkjet-printed OECT on transient substrate.

II. Materials and Methods

A. Fabrication of the Substrates

0.3 g of grains of CDD were mechanically pressed for about 20 minutes with forces of 60 kN by using a two-column precision manual press at room temperature. The resulting CDD films have a diameter of 3–4 cm and are about 700 micrometers thick. More details on the fabrication procedure and the sublimation rate of these substrates are reported in [19]. A bilayer of polymer poly(vinyl alcohol) (PVA) and polyvinylpyrrolidone (PVP) was deposited by casting directly on top of the CDD substrate and defining an area of 2 × 2 cm^2. PVA was deposited from a water solution (8% by weight) and left drying overnight, while PVP was casted from a Chloroform solution (7 % by weight) and left to dry for a few minutes under a fume hood.

B. Fabrication of Organic Electrochemical Transistors

OECTs were inkjet-printed using a PEDOT:PSS ink modified with ethylene glycol (EG, 20%) with a drop-on-demand Dimatix Materials Printer 2800 (Fujifilm). As for the electrolyte, 1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIm TFSI) was deposited on top of the PEDOT:PSS line, working as the active channel (Fig. 1(a), more information about the electrical characterization can be found in the Methods section).

One of the two PEDOT:PSS line served as the gate electrode and the other as the channel layer of the OECT. The widths and lengths of the lines are equal to 3mm and 140µm, respectively, and the distance between the two lines is 450µm. The printing was carried out under ambient conditions, with a 16-nozzle cartridge (10 pL single-drop volume). Different overlapping layers of PEDOT:PSS were printed (5 or 10), with a 60 s pause between two consecutive printing steps to guarantee the deposition of a uniform film. OECTs with different channel widths (100µm, 200µm and 300µm) and different distance between the electrodes (90µm, 300µm, 450µm and 590µm) were printed both on transient substrates and glass slides for comparison.

Optical images of an inkjet-printed OECT on a transient substrate are shown in Fig. 1(b), with the typical planar and symmetric configuration visible in the multiple enlargements, and in Supplementary Fig. 1(a), where the definition of the electrode, increasing the number of printed passes, is evaluated. The profiles of a 5- and 10-layer inkjet-printed electrodes on glass slide are shown in Supplementary Figure 1(b).

The OECTs were fabricated on the transient (CCD)/PVA/PVP substrates (see Fabrication of the substrates). The PVA layer was used to planarize the surface of the CDD layer and confer stability to the PVA–CDD interface. PVP was used to enable orthogonality in the multilayer coating (the PEDOT:PSS layer is printed from an aqueous dispersion which could dissolve the underlying PVA layer).

C. Electrical characterization

The electrical characterization of the transistors was performed under ambient conditions immediately after printing the devices using a Keithley SCS4200 parameter analyzer. The electrical measurements related to transfer characteristic were performed by sweeping the gate voltage V_{GS} from 2V to -0.5V and back with a step of 0.01V. The hold time was 60 seconds, and the sweep delay was 1 second. Indeed, the electrical measurements related to output characteristic were performed by sweeping the drain voltage V_{DS} from 0V to -0.5V and back with a step of 0.01V and the gate voltage V_{GS} from 2V to -0.5V with a step of 0.5V. The hold time was 60s, and the sweep delay was 0.5s The measurements under different humidity conditions were conducted inside a closed box where nitrogen gas was flushed in through an inlet valve. pH measurements were performed in ambient conditions using three buffer solutions with pH values of 4, 7, and 9. A full electrical characterization was also performed on PEDOT:PSS OECT inkjet-printed on glass slides following the same printing process (see Supplementary Information).

D. Materials

CDD was purchased from Kremer Pigmente GmbH & Co. KG (Aichstetten). PVA and PVP were purchased from Sigma Aldrich. PEDOT:PSS (CleviosTM PH 1000) was purchased from Heraeus.

III. Results and Discussion

A. PEDOT:PSS-based OECTs electrical characterization

The OECT principle of operation is schematically shown in Fig. 2(a). The channel current is generated by the flux of charge carriers (i.e., holes in PEDOT:PSS) determined by the V_{DS} voltage applied across the electrode acting as the channel. The gate voltage V_{GS} modulates the channel resistivity by the injection of cations from the electrolyte, whose effect is to un-dope the channel and reduce the electric conductivity until an OFF state is reached, as in a depletion mode field effect transistor.

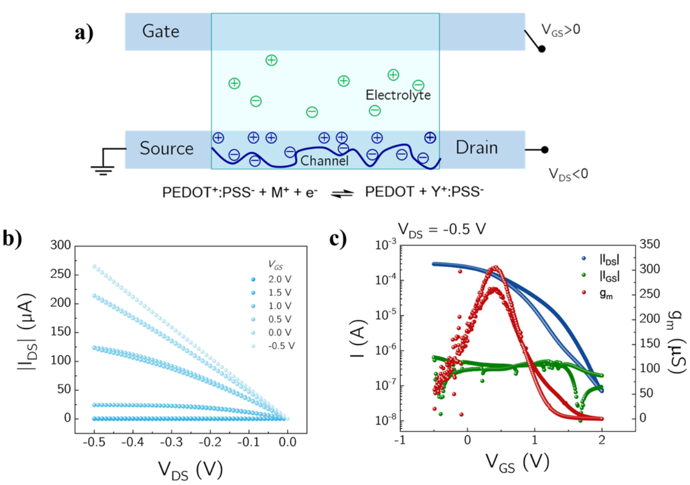

Fig. 2. Electrical characterization of OECTs with 10-layers inkjet-printed PEDOT:PSS on transient substrate. a) Schematic principle of operation of the OECT; b) Typical output characteristic measured at different gate voltages (from V_{GS} = 2.0 V to V_{GS} = -0.5 V, steps of 0.5 V); c) Typical transfer characteristic measured as a function of the gate voltage for a drain voltage of -0.5 V. Left axis, I_{DS} and I_{GS} in logarithmic scale, right axes: transconductance (g_m).

Both the gate electrode and the channel are printed using the same number of layers (10) and the same printing settings (as reported in Section B of Materials and Methods). Fig. 2(b) and 2(c) report the typical output characteristics, measured at different gate voltages (from V_{GS} = 2.0 V to V_{GS} = -0.5 V, steps of 0.5 V), and transfer characteristic, measured as a function of the gate voltage for a drain voltage of -0.5 V for 10-layer printed devices. As shown in Fig. 2(c), the maximum value of the transconductance g_m is 300 µS comparable with other devices fabricated on flexible substrates [20]. OECTs with large transconductance at low operating voltages (V_{GS} < 5V) are well-suited for high-performance biosensors, because transconductance is an important parameter in signal amplification and markedly influences the device sensing capabilities, including its detection limit.

OECTs with different architectures were fabricated and characterized changing the number of PEDOT:PSS layers, the channel width and the distance between the channel and the gate electrode. Details and characteristics of the different structures can be found in Supplementary Fig. 2. An analysis was performed on the peak value of the transconductance of 26 devices fabricated with the same printing settings but with different geometries (Supplementary Table 1 and Fig. 3). As demonstrated in [21], the OECT peak value of g_m scales with the *Wd/l* where W and l are the width and the length of the channel respectively, and d is the electrode thickness.

The thickness of the electrode is not directly proportional to the printing passes, because the first layers are partially absorbed by the substrate and the subsequent layers partly blend with each other [22]. As previously demonstrated [23], the operational range of all-PEDOT:PSS organic electrochemical transistors can be manipulated modifying the device geometry, as observed in structures where the gate serves as a polarizable electrode. To evaluate the influence of the thickness of the electrodes, which depends on the number of inkjet-printed layers, we compare OECTs with different layers. In Fig 3(a), the transfer characteristic of an OECT printed using the same number of layers for the gate and the channel electrodes is reported. To prove the symmetrical behavior of the device, the transfer characteristics are measured using alternatively the stripes either as the gate electrodes, or the channel, for a fixed drain voltage of -0.2V.

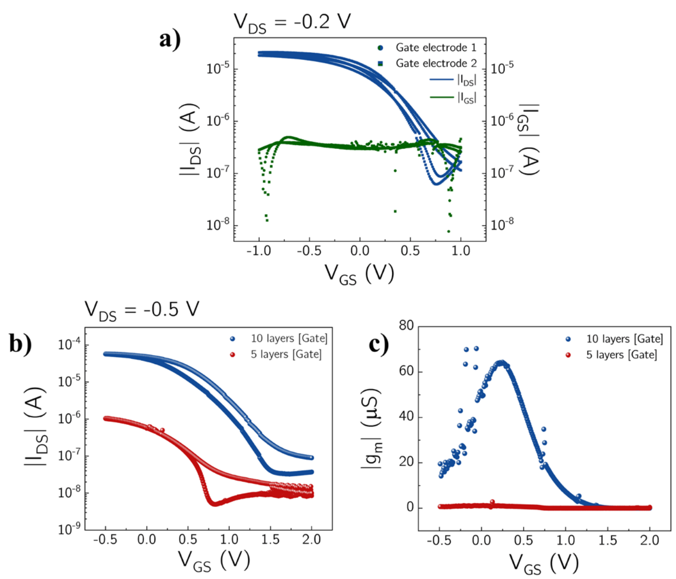

Fig. 3. Electrical characterization of the OECT on transient substrate. a) Electrical characterization of an OECT printed using the same number of layers for the gate electrode and the channel (for a drain voltage of -0.2V, logarithmic scale). b-c) Electrical characterization of an OECT printed using the different number of layers for the gate electrode and the channel; the transfer characteristics are measured using alternatively the stripes as the gate electrodes (for a drain voltage of -0.5V, logarithmic scale).

To evaluate the effect of gate electrode thickness on the electrical performance of the device, different architectures were tested.

Fig. 3(b) shows the transfer characteristic curves (registered for a drain voltage of -0.5V) for a device with asymmetrical

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number of layers for the gate electrode and the channel. The higher the number of layers, the higher the conductivity of the electrode. Therefore, a thicker gate electrode (10 layers, blue curve) determines a higher modulation of the channel. Consequently, a quasi-flat transconductance was measured in the case of 5-layers gate electrode (Fig 3(c), red curve). As demonstrated in [24], larger thicknesses corresponded to higher gains for OECTs operating as switches in a transistor-like mode.

B. PEDOT:PSS-based OECTs in time, pressure and humidity conditions

We then evaluate the electrical performance of inkjet-printed OECTs on transient substrates over time and in controlled atmosphere. We considered two different devices: one single PEDOT:PSS 10-layer electrode (Fig. 4a-c) and a PEDOT:PSS OECT (Fig. 4b-d) with 10-layer electrodes and measured the current through the devices over the course of some days and in different humidity conditions.

From Fig. 4(a), it is clear that, already two days after the fabrication, the electrode becomes more resistive (i.e., from 25 kΩ to 165 kΩ) if stored in air at constant temperature (18°C), with the measurements made in air, at ambient conditions. Accordingly, the OECT also exhibits a lower drain current as the days pass (Fig. 4(b)).

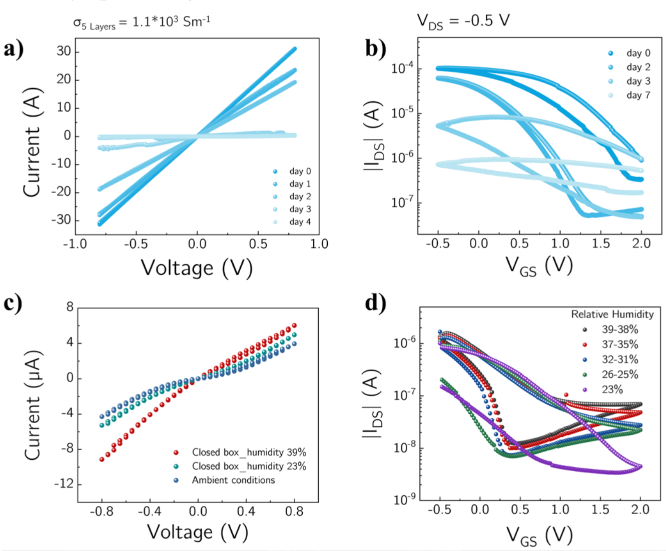

Fig. 4. Characterization of inkjet-printed electrodes and OECTs in time and in different ambient conditions. Conductivity of a 5-layer electrode (a) and transfer characteristic of an OECT (b) measured as a function of time. Conductivity of 10-layer electrode (c) and OECT transfer characteristic measured in a closed box with different humidity conditions (d).

Measurements have also been carried out in different humidity conditions in a closed box flushed with nitrogen gas and in ambient conditions (the box was open, and no gas was flushed in) (Fig. 4(c) and 4(d)). PEDOT:PSS conductivity increases with increasing humidity. However, after measuring the electrode conductivity with the box opened (i.e., initial humidity of 39% restored), the current does not return to its initial value (blue curve in Fig. 4(c)).

Humidity measurements have also been performed at constant temperature and pressure on the OECT. As in the previous case, as the humidity decreases from 39% to 23%, the device drain and gate current decreases, as shown in Fig. 4(d). Specifically, when the relative humidity (RH) drops below 25%, the performance evidently degrades for the same voltage range.

To verify that electrical performance varying the humidity is not affected by the transient substrate, the same device has been fabricated and characterized on a glass slide (see Supplementary Note 3). It can be seen that at a constant temperature and pressure, as the humidity in the closed box varies (from 29% to 23%), the current appears to decrease over time (see Supplementary Fig. 6), showing the same behavior as on the transient substrate.

C. An application of PEDOT:PSS OECTs as pH sensor

We also explore the possibility to use the PEDOT:PSS OECTs as disposable sensors for real-time liquid pH detection, which would allow to measure pH levels in various aqueous environments, like biological fluids or soil. Theoretically, as the pH decreases, indicating an increased concentration of hydrogen ions, more ions can participate in modulating the on and off states of the OECT. This means that a greater number of hydrogen cations in the electrolyte can enter the PEDOT:PSS channel, leading to a rapid reduction in the drain current for a given gate voltage. In literature various articles have reported on the use of PEDOT:PSS-based OECTs for pH sensing applications on both rigid [25] and flexible [26] substrates.

At first, 10-layer PEDOT:PSS inkjet-printed single electrodes on transient substrate have been tested to study the change in conductivity of the material varying the pH of the analyte. In accordance to the literature [12], the conductivity of the film decreases increasing the pH of the solution (Supplementary Fig. 7). As shown in Supplementary Fig. 8, the same results have also been obtained on PEDOT:PSS electrodes printed on the glass slide.

Then, OECT-based pH sensors were fabricated printing 10-layer gate electrodes and 5-layer channels on the transient substrate. The inset in Fig. 5(a) shows the sketch of the measurement setup: during the pH sensing test, the ionic liquid is replaced by a drop of three different pH buffer solutions used as the electrolyte (red drop, pH 4; green drop, pH 7, blue drop, pH 9). Fig. 5(a) shows the transfer characteristics for three different pH concentrations at V_{DS} = -0.15V and Fig. 5(b) shows the transconductance for different gate voltages and different pH concentration, with values above 30 μS. An elevated transconductance value, above 10 μS (particularly in sensing applications based on OECT devices) enhances the signal-to-noise ratio and reduces the detection limit. Consequently, the sensor sensitivity is increased, as even a small input signal produces a substantial response in the output.

For comparison, Fig. 5(c) and 5(d) show the transfer characteristics and the transconductance obtained by printing the OECT on a glass slide using the same printing settings, geometrical parameters and pH concentrations as for the transient substrate case. Despite the different voltage range applied for the measurements, the transconductance as a

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function of the pH follows the same trend as in the transient substrate.

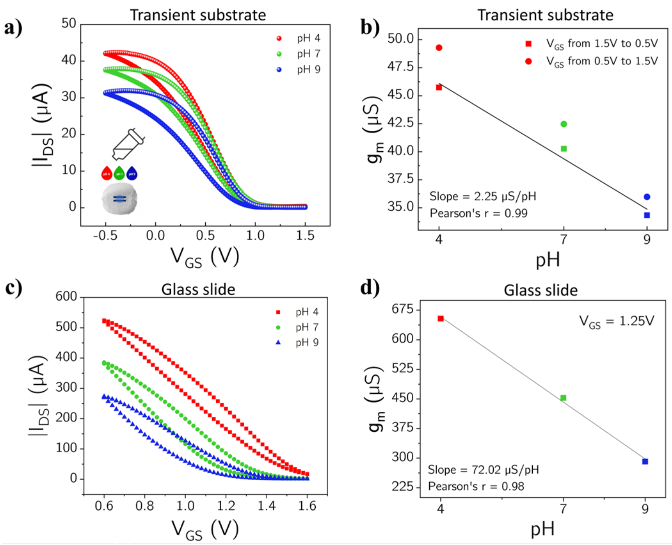

Fig 5. Response of OECTs to different pH values. a) Transfer curves at V_{DS} = - 0.15V and b) transconductance values at V_{GS} = 0.4V in different pH buffers on transient substrate. Inset: scheme of the measurement setup (potentiodynamic measurements). c) Transfer curves and d) transconductance curves in different pH buffers on glass slide.

IV. Conclusion

The fabrication of PEDOT:PSS-based OECTs on naturally degradable substrates represents a promising choice in the field of flexible and environmentally friendly electronics. We have studied the electrical performance of these devices in air and under various humidity conditions and explored the application as pH sensors, where we achieved very interesting transconductance value above 30 µS. The results highlight the feasibility of our process for the definition of OECTs on transient substrates, and also prove their potential as key players in the field of biosensing and environmental monitoring.

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