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A bio-inspired memory device based on interfacing *Physarum polycephalum* with an organic semiconductor

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The development of devices able to detect and record ion fluxes is a crucial point in order to understand the mechanisms that regulate communication and life of organisms. Here, we take advantage of the combined electronic and ionic conduction properties of a conducting polymer to develop a hybrid organic/living device with a three-terminal configuration, using the *Physarum polycephalum* Cell (PPC) slime mould as a living bio-electrolyte. An over-oxidation process induces a conductivity switch in the polymer, due to the ionic flux taking place at the PPC/polymer interface. This behaviour endows a current-dependent memory effect to the device. © 2014 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [<http://dx.doi.org/10.1063/1.4902817>]

In the landscape of organic bioelectronics^{1,2} and, in particular, electroactive materials,³ an important aspect is represented by the capability of recording signals useful to understand principles of living organisms. In particular, nervous systems of living beings use the same elements for recording and processing information. Thus, differently to what occurs in present computers, information plays an active role in these systems; it is not only memorized but also modifies the strength of the interconnections between elements of the nervous system, adapting it to better resolving similar problems in the future. This fundamental feature is responsible for the learning of living beings. Bio-inspired computers must be based on these principles. Thus, we need an element with properties similar to those of synapses; it must vary its conductivity state according to the history of the signal passing through it. Recently, a new type of electronic element, memristive device, has been widely considered as promising candidate for such kinds of circuits. In particular, organic memristive devices⁴ were successfully used as synapse analog,⁵ allowing the construction of circuits capable of Hebbian type of learning.⁶ In contrast with computers, such kind of bio-inspired memory does not imply complete erase of the information. The connection can be suppressed by additional learning, but, once obtained, the information is practically always accumulated in the system. The peculiar properties of organic semiconductors, such as the ability to conduct both *ions* and *electrons*,^{7,8} their synthetic tunability, and ease of processing, have been exploited in several classes of devices that range from Organic Light Emitting Diodes (OLEDs)⁹ to organic photovoltaics^{10,11} and Organic Thin Film Transistors (OTFTs).^{12,13} Low-cost, simple manufacture processing, and versatile geometry allow a straightforward integration of organic-based technology into lab-on-a-chip devices. In addition, the ability of organic semiconductors to operate in aqueous environment and their proved biocompatibility^{14–16} make them the ideal interface between electronics and biological systems.¹⁷ However, despite quick advancements of organic bioelectronics in recent years, only few examples have been described about interfacing conducting polymers with living systems. In most cases, they are related to studies focused on cell adhesion,^{18–20} measuring neuronal activity,²¹ or developing sensors for toxicology.^{22–26}

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A challenging and fascinating frontier of bioelectronics is the integration of living systems with organic-based devices, towards the development of hybrid living/organic tools, where the living system plays a key role in the device operation. To this regard, living systems characterized by intrinsic smart behaviours result particularly interesting, due to their ability to actively interact with the environment. Among these systems, the *Physarum polycephalum* Cell (*PPC*) slime mould is well suited for the implementation of the functional properties of smart living systems into electronic devices. *PPC* belongs to the family of myxomycetes, taxonomically classified as Protozoas or simply slime mould.^{27,28} *PPC* is a multinuclear mass of protoplasm, a single cell with a myriad of nuclei, able to regenerate autonomously, even if a part is cut away. Its foraging behaviour consists of optimized networks of protoplasmic tubes branching towards nutrients.²⁹ During its life cycle, *PPC* produces a certain amount of slime, a sort of “external brain” that allows the *PPC* remembering already trodden paths, in order to not retrace them when seeking for food or attractants.³⁰ Because of these unique features, *PPC* has been widely studied, since many years, in different research fields like unconventional computing,³¹ networks modelling and development,^{32,33} biorobotics,³⁴ and biochemistry.^{35,36}

As far as the interfacing of organic semiconductors with living cells is concerned, in a recent work,³⁷ we have interfaced the *PPC* slime mould with poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), one of the most ubiquitously used conducting polymers.³⁸ The choice of *PPC* was made because it represents a macroscopic living cellular model, well-fitting with the purpose of interfacing a living organism with an organic device. In our study, we demonstrated the possibility of using *PPC* for the implementation of a hybrid living-organic electronic device, namely an Organic Electrochemical Transistor (OECT), showing both sensing and memristive properties when used in a three or two-terminal configuration, respectively. In the light of this memristive behaviour, we believe that it is of great interest to investigate in a more detailed way the role of the interface between *PPC* and PEDOT:PSS and their effective contribution to the properties showed by our *PPC*-based OECT/memristor. It is worth to mention that the memristive properties of our device are a unique prerogative resulting from the coupling of Ag/*PPC*/PEDOT:PSS. In fact, by using saline solutions commonly used in OECT applications, e.g., PBS (Phosphate Buffered Saline), the device does not show neither hysteresis in the negative voltage branch nor the pinch-off at 0 V, which is another fingerprint of memristive devices.

In this paper, we present a study of the mechanisms arising from interfacing PEDOT:PSS with *PPC*. We recurred to a three-terminal device configuration, already used for organic memristive systems based on solid electrolytes and organic semiconductors,³⁹ where the *PPC* is used as *electrolyte* and the gate electrode, placed in it, is kept at zero-bias (silent gate) and used as a controlling electrode. In this configuration, we recorded the current flowing through the conducting polymer and monitored the gate current, the latter providing information on the contribution of the flux of ionic species taking place across the *PPC*/PEDOT:PSS interface. Interestingly, our device showed memory properties, associated to the formation of meta-stable redox states in the polymer, similar to those of write-once read-many memories (WORMs).⁴⁰ These results pave the way to the realization of innovative memory devices based on hybrid living/organic systems.

Physarum polycephalum has been cultivated in petri dishes with Agar 1.5% non-nutrient gel (Figure 1(a)), fed with oat flakes and kept in a dark and humidity controlled chamber. The colony was periodically replanted to new agar substrate.

The conducting channel, made of PEDOT:PSS, was patterned on square glass slides of 2×2 cm using standard methods,⁴¹ resulting in a 10×1 mm stripe (~ 100 nm thick). PEDOT:PSS was supplemented with di-ethylene glycole 20% and dodecylbenzenesulphonic acid (DBSA) 0.5%, in order to improve its conductivity and film formation properties, respectively.⁴² Source and drain electrodes were obtained by applying silver conductive paste at the extremities of the PEDOT:PSS channel. Fresh slime mould was placed directly onto the conducting channel, playing the role of the electrolyte. A silver wire, acting as the gate electrode, was immersed into the mould body and kept at zero-bias, acting as a reference silent gate electrode (Figure 1(b)).⁴

Electrical outputs were recorded using two Keithley 2400 SourceMeters, controlled by a home-made Matlab software. Measurements were carried out by recording the hysteresis loops

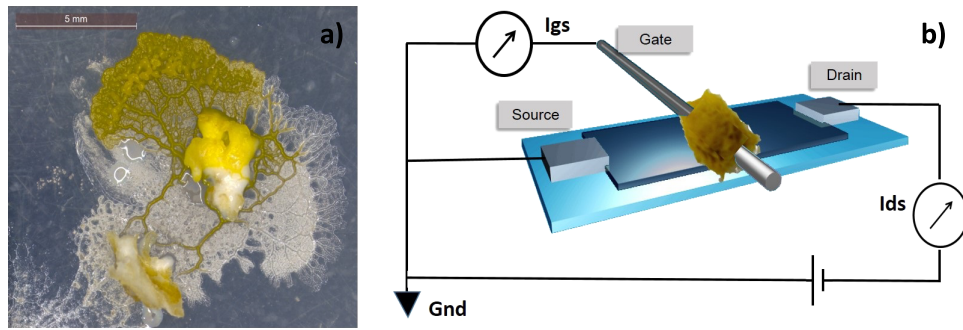


FIG. 1. (a) Optical micrograph of *Physarum Polycephalum* slime-mould, cultivated on agar non-nutrient gel. The yellow tree-like network is the mould, surrounded by a white-transparent area (the slime); two oats partially covered by *Physarum* can be also distinguished. (b) Schematic illustration of the OECT architecture. The yellow blob and the dark blue channel represent the PPC slime mould and the PEDOT:PSS thin film, respectively. A grounded Ag wire, placed directly in the PPC mould, acts as silent gate electrode, while source-drain potential is swept from positive to negative voltages.

showed by source-drain current I_{ds} . In detail, the source-drain voltage (V_{ds}) was swept in a symmetrical range ($-V_{ds}^*$, $+V_{ds}^*$), with 1 s long-lasting steps of 0.1 V, and by setting V_{ds}^* at 1, 3, 7.5, and 10 V. The hysteresis loops have been recorded by sweeping V_{ds} starting from 0 V to $-V_{ds}^*$, then from $-V_{ds}^*$ to $+V_{ds}^*$ and returning to 0 V. Simultaneously, the related gate-source current I_{gs} was also monitored. All measurements were performed on the same device, upon successive V_{ds} potential scans on progressively increasing voltage ranges.

Figure 2(a) shows the hysteresis loop of the drain current (I_{ds} vs. V_{ds}) obtained by sweeping the drain voltage between $V_{min} = -1$ to $V_{max} = +1$ V (black curve) and $V_{min} = -3$ to $V_{max} = +3$ V (red curve). In the first case, the device shows an ohmic behavior, while, in the second case, we observe a clear rectifying behaviour with a plateau in the negative branch and a slight hysteresis. Figure 2(b) represents the same measurement extended over a wider V_{ds} range (-7.5 , $+7.5$ V). Interestingly, when sufficiently high V_{ds} voltages (e.g., over a threshold between 6 and 10 V) are reached, the I_{ds} current drops abruptly by several orders of magnitude. This indicates a transition of PEDOT:PSS from its conductive to non-conductive state. We tentatively explain this phenomenon by a likely over-oxidation process, leading to a non-conducting and electrochemically inactive film. Over-oxidation of thiophene-based organic semiconductors was observed in thin films exposed to sufficiently high anodic potentials in a wet environment,^{43,44} that is similar to our case, where PEDOT:PSS is in contact with the slime mould. Irreversible conductivity switches due to over-oxidative mechanisms have been already described in literature for the specific case of PEDOT:PSS.^{45,46} Berggren's group has demonstrated that over-oxidation of PEDOT:PSS, upon exposure to large anodic potentials, can be exploited for subtractive patterning in the fabrication of ion pumps, where specific areas of the polymer can be made selectively non-conductive, in order to develop electrically insulating-ionically conducting

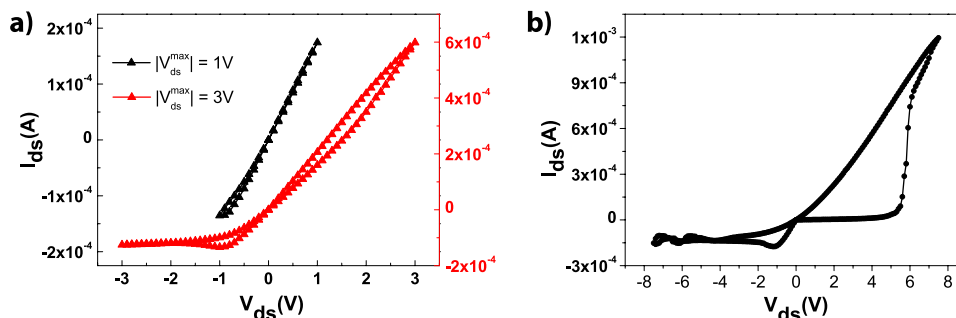


FIG. 2. (a) I_{ds} vs V_{ds} profiles measured for increasing voltage ranges ($(-1, +1)$ V, black curve; $(-3, +3)$ V, red curve). (b) Hysteresis loop measured in the V_{ds} range ($-7.5, +7.5$) V. Conductivity switch of PEDOT:PSS towards a less conductive form can be observed in the positive voltage branch (the switch can be related to an over-oxidation of the polymer).

channels. Such mechanism is related to cationic species, in particular divalent cations, present in the electrolyte, which greatly contribute to the abatement of the polymer conductivity. To this regard, in our specific case, it's important to recall that among other ionic species, the intracellular matrix of *PPC* is particularly rich of Ca^{++} .^{47,48} The mechanism causing the conductivity decrease was showed to be associated to the cleavage of the conjugation pathway, accompanied by the formation of sulfone, carbonyl, and carboxylic groups in some specific positions of the thiophene rings.⁴⁵ Even though high voltages are generally required, it was also demonstrated that the over-oxidation potential decreases at high pH. On the basis of all these results, we formulate our hypothesis of an over-oxidation process occurring when PEDOT:PSS is directly interfaced with *PPC* slime mould and appropriately biased.

In particular, the average threshold V_{ds} voltage necessary to induce the over-oxidation in PEDOT:PSS, when interfaced with *PPC*, results to be much lower than that used in ion pumps (+25 V). This could be due to the higher ionic content and the major complexity of the intracellular matrix of *PPC*, which could reasonably strongly interact with the conducting polymer.

Interestingly, after the conductivity switch, our *PPC*/PEDOT:PSS device shows an hysteretic behaviour in current-voltage characteristics ascribable to a memory-like effect, similar to that observed in other organic memory devices (OMD),⁴⁹ where typical features are hysteresis and bi-stability (meaning that the device shows two stable electrical states). In our case, we performed a cyclic current-voltage measurement, soon after the over-oxidation had taken place. A wider voltage range of the V_{ds} potential was investigated ($-10, +10$ V). As a result, we obtained an I_{ds} vs V_{ds} loop showing a large hysteresis area, with two well-defined lobes in each branch of voltages scanned (Figure 3(a)) and oxidation/reduction peaks (located around +4.5 V and -5 V, respectively). It has to be pointed out that the memory effect found in our device presented a non-volatile nature. In addition, this memory effect is similar to that showed by non-volatile WORMs,⁴⁰ where information is stored using the resistive switching above a certain voltage level and the actual state of the device is read at a lower voltage. WORM can be implemented by fuses (in which all lines are made conductive during production and selectively fused at the site of use) or anti-fuses (lines made conductive at the site of use). Noteworthy, fuse technology based on PEDOT:PSS has also been described.⁵⁰ Hence, our device could represent an innovative and biologically relevant memory device, based on the intrinsic integration of an organic semiconductor with a living system.

The ionic flux across the *PPC*/PEDOT:PSS interface would play a key role in both the rectifying behavior and conductivity switches observed in our hybrid system. As for other PEDOT:PSS-based electrochemical devices, the current flowing into the conducting channel can be modulated by reversible doping/de-doping upon injection of cations from an electrolyte set in contact with the polymer channel.⁵¹ The potential drop along the channel generates a current towards the drain electrode. This current includes two contributions: ionic species injected into the polymer and its intrinsic charge carriers. By measuring the current in the (silent) gate circuit (I_{gs} at $V_{gs} = 0$ V), we uncouple these two contributions and monitor the sole contribution of the ionic species entering PEDOT:PSS. In order to investigate the role of the ionic flux across the *PPC*/PEDOT:PSS interface, we monitored the I_{gs} current as a function of V_{ds} in presence of the memory effect (Figure 3(b)).

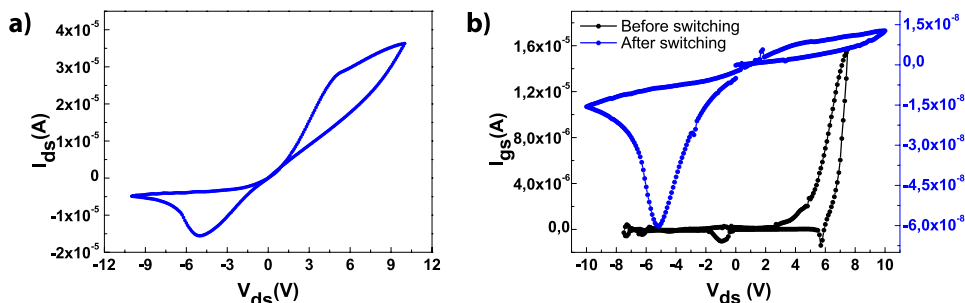


FIG. 3. (a) Hysteresis curve showing the typical behaviour of a memory device, measured on a *PPC*/PEDOT:PSS device soon after the conductivity switch has taken place. V_{ds} was swept in the ($-10, +10$) V range. (b) I_{gs} vs V_{ds} before (black curve) and after (blue curve) the occurrence of the conductivity switch.

During the voltage sweep, according to the sign of the potential acting on the conducting channel, charged species are expected to cross the *PPC*/PEDOT:PSS. In particular, for anodic potentials ($V_{ds} > 0$), anionic species are injected from the *PPC* bulk into the conducting polymer. The increase of I_{gs} as a function of positive potentials is directly related to the flux of cationic species towards the silent gate electrode placed in the *PPC* body. After the over-oxidation process, for $V_{ds} < 0$, the weight of a small peak in the I_{gs} current (placed at -1 V before the conductivity switch, Figure 3(b), black curve) strongly increases, along with a shift of its position towards higher negative potentials (~ -5 V). This peak is related to the hysteresis of I_{ds} , and, as a consequence, to the crossing of cationic species from *PPC* bulk to PEDOT:PSS. This passage is also responsible of the rectifying behavior associated with the de-doping of PEDOT:PSS film. The ionic drift recorded at the gate electrode demonstrates that the active role in the electrochemical changes involved in the memory effect observed is mostly played by the *PPC* and PEDOT:PSS bulks, rather than the *PPC*/PEDOT:PSS interface. Finally, we can argue that the observed memory effect is the result of the intimate coupling of PEDOT:PSS with a relatively complex living system such as *PPC*.

In conclusion, our study of a hybrid junction between a living system, the *Physarum polycephalum* Cell, and the conducting polymer PEDOT:PSS demonstrates an innovative bio-inspired device, showing memory effects induced by electrochemical changes occurring into the polymer upon application of high anodic potentials across the semiconducting PEDOT:PSS channel. We explain these effects in the light of an over-oxidation mechanism, triggered by the cationic flux across the *PPC*/PEDOT:PSS interface. The device can be accounted as an element with properties similar to those of synapses, useful to implement bio-inspired computation.

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