



A physical-based equivalent circuit model for an organic/electrolyte interface



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ABSTRACT

The aim of this work is to develop an equivalent circuit model for the metal-organic semiconductor-electrolyte structures that are typically used as transducers in biosensor devices. In particular, a perylene derivative material is implemented in the active layer of a gold-semiconductor-electrolyte stack. Our approach is extending the standard range of the bias voltages applied for devices that operate in water. This particular characterisation protocol allows to distinguish and investigate the different mechanisms that occur at the different layers and interfaces: adsorption of ions in the semiconductor; accumulation and charge exchange of carriers at the semiconductor/electrolyte interface; percolation of the ionic species through the organic semiconductor; ion diffusion across the electrolyte; ion adsorption and charge exchange at the platinum interface. We highlight the presence of ion percolation through the organic semiconductor layer, which is described in the equivalent circuit model by means of a de Levie impedance. The presence of percolation has been demonstrated by environmental scanning electron microscopy and profilometry analysis. Although percolation is much more evident at high negative bias values, it is still present even at low bias conditions. The very good agreement between the model and the experimental data makes the model a valid tool for studying the transducing mechanisms between organic films and the physiological environment.

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1. Introduction

The flexibility and biocompatibility of many organic semiconductors are properties that easily prompted the use of these materials for their implementation in chemical and biological sensing [1]. Moreover, organic thin-film transistors (OTFTs) allow the integration of a sensor and an amplifier in a single device. In fact, it has been widely demonstrated the potentiality of these technologies in gaining better performances with respect to inorganic devices, such as silicon TFTs or micro-electrode arrays [2–5].

Among the different device structures, it was recently reported that the organic cell stimulating and sensing transistors (O-CSTs) are capable to interface with neuronal cells directly plated on top of the organic layer [6,7]. The mechanism of transduction of ionic/electronic currents at the interface between the organic layer and the biological environment has to be fully investigated in order to master and optimize the performances of the overall device sensor.

At this purpose, we simplified the architecture of the three-electrode device in order to throughout investigate the different physical phenomena occurring in the O-CST system. We employed the electrochemical impedance spectroscopy (EIS) technique as investigating probe to extrapolate a complete model of the device. In this scenario, our results may be easily implemented without loss of generality to a plethora of other devices at the interface with the physiological/electrochemical environment such as water-gated transistors [8–10], dye sensitized solar cells (DSCs) [11],

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super-capacitors [12,13], light-emitting electrochemical cells (LECs) [14,15], etc.

To the best of our knowledge, the different models that have been as far developed for similar devices [16–25] describe only in part the whole device physics. In particular, they omit the description of the diffusion layer in the electrical double layer (EDL) or they include mathematical artifices like constant phase elements (CPEs) to describe the endogenous effects of surface irregularity, roughness, porous interfaces, frequency dispersion in many processes, or other non-idealities.

Accordingly, the present work aims at developing an equivalent circuit model for the metal-organic semiconductor-electrolyte stack structure in which every circuit element is correlated to a particular physical phenomenon. We kept the model as simple as possible, avoiding the use of CPEs. In fact, the introduction of more sophisticated circuit elements in the model may result in loss of physical meaning due to the increase in the number of fitting parameters. The consequent pointless complication in the extrapolation of the parameters would make difficult the interpretation of the results, such as discerning among different phenomena.

2. Materials and methods

To get a clearer understanding of the working principles of the O-CST (depicted in Fig. 1), we use a simple 2-terminal device shown in Fig. 2. The 2-electrode device architecture is used in order to: i) specifically investigate the device physics at the bulk and the semiconductor electrolyte interface of the organic material ii) reduce parasitic couplings among the O-CST electrodes as much as possible; and iii) perform measurements on devices with a larger area (roughly 2 cm^2) for increasing the signal-to-noise ratio (SNR) and, consequently, the measurement accuracy.

In the following, we will refer to this structure as the stack device.

Our experimental procedure is depicted in Fig. 3. The procedure

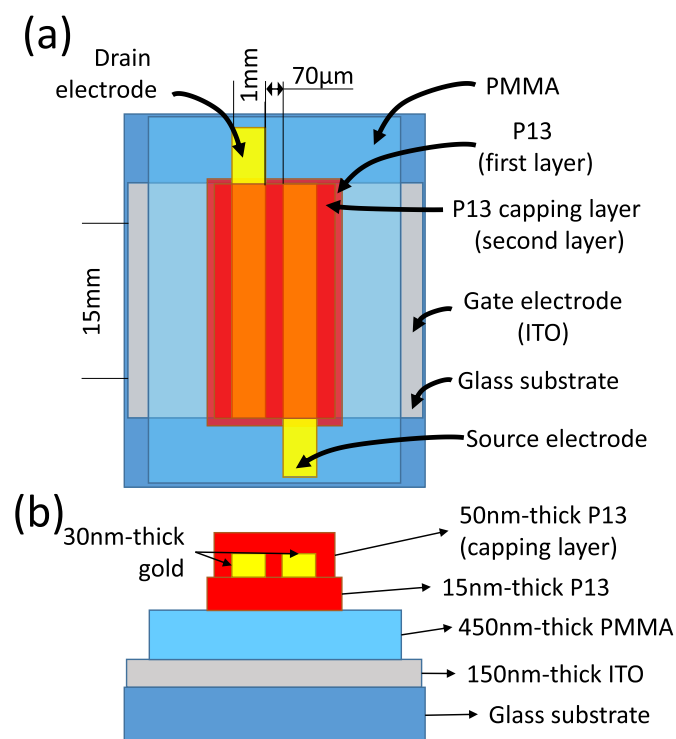


Fig. 1. (a) OCST top view. (b) OCST cross section.

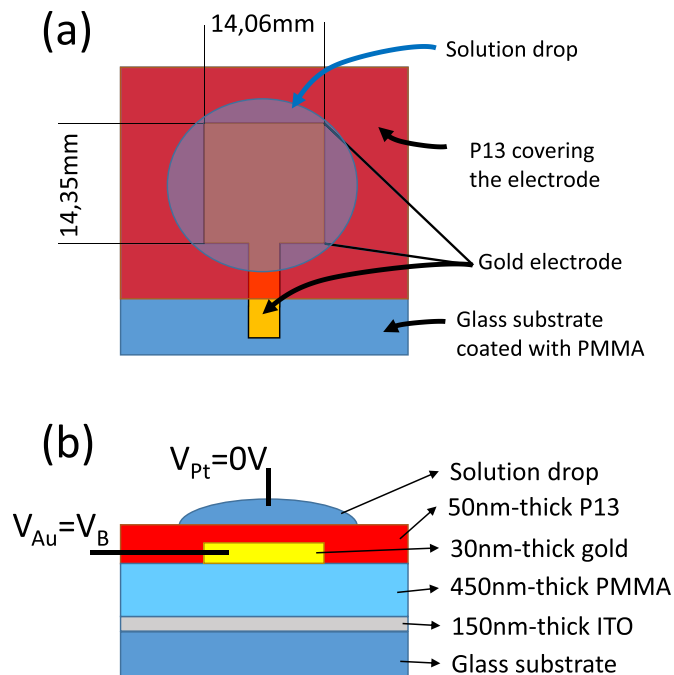


Fig. 2. (a) STACK top view. (b) STACK cross section showing the applied voltages at the platinum (V_{Pt}) and gold (V_{Au}) electrodes. (colors online).

can be divided into eight steps, which are listed here below and described in details in the following sub-Sections 2.1–2.5:

- (1) *Sample preparation*, described in Section 2.1.
- (2) *Surface analysis*. We used both environmental scanning electron microscopy (ESEM) and optical profilometry (see Section 2.2).
- (3) *PDL functionalization* (Section 2.3). Poly-D-lysine (PDL) is used to help cell attachment to the devices surface. We adopted the PDL treatment to keep as close as possible to the real operating condition.
- (4) *NaCl flush* (Section 2.3). Because we performed EIS measurements with a NaCl solution (concentration of 0.1 M), we used the same solution to clean the surface from any contaminant that might remain from the previous step.
- (5) *NaCl deposition* (Section 2.3). Approximately 0.1 ml of NaCl was put on top of the device for characterizations.
- (6) *EIS Measurements* at different bias conditions (Section 2.4).
- (7) *MilliQ wash* (Section 2.5). Immediately after the EIS measurements, devices were cleaned using an appropriate protocol using MilliQ water in order to remove NaCl residuals. After cleaning, devices were left to dry before the next step.
- (8) *Surface analysis*. Same as in point (2) for comparing the behaviour of the device before and after measurements.

2.1. Sample preparation

Stack devices (Fig. 2) were fabricated onto a soda-lime glass substrate pre coated with a 150 nm thick indium tin oxide (ITO) layer. The substrate was cleaned by means of multiple sonications in acetone and isopropanol solutions before a 600 nm-thick PMMA film was spin-coated on it. After an overnight annealing treatment in inert atmosphere the PMMA film thickness was reduced to about 450 nm. A 30 nm-thick gold was then evaporated onto the substrate defining the active area of the device as a

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