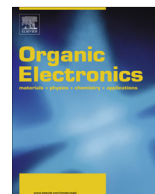




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Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Liquid electrolyte positioning along the device channel influences the operation of Organic Electro-Chemical Transistors

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ARTICLE INFO

Article history:

Received 11 March 2014

Received in revised form 12 August 2014

Accepted 18 August 2014

Available online xxx

Keywords:

Organic Electro-Chemical Transistor

Electrochemical sensors

Polymers hydration

Moving-front technique

Super-hydrophobic surface

ABSTRACT

In this work, we show the influence of the liquid electrolyte adsorption by porous films made of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate), PEDOT:PSS, on the operation of an Organic Electro-Chemical Transistor with an active channel based on these polymeric films. In particular, the effect of film hydration on device performance is evaluated by studying its electrical response as a function of the spatial position between the electrolyte and the channel electrodes. This is done by depositing a PEDOT:PSS film on a super-hydrophobic surface aimed at controlling the electrolyte confinement next to the electrodes. The device response shows that the confinement of ionic liquids near to the drain electrode results in a worsening of the current modulation. This result has been interpreted in the light of studies dealing with the transport of ions in semiconducting polymers, indicating that the electrolyte adsorption by the polymeric film implies the formation of liquid pathways inside its bulk. These pathways, in particular, affect the device response because they are able to assist the drift of ionic species in the electrolyte towards the drain electrode. The effect of electrolyte adsorption on the device operation is confirmed by means of moving-front measurements, and is related to the reproducibility of the device operation curves by measuring repeatedly its electrical response.

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1. Introduction

Organic Electro-Chemical Transistors (OECTs) are nowadays playing a crucial role as joining link between the largely investigated organic electronics and the newest frontier of research addressed to bio-inspired applications. This dual purpose is made possible by the fact that OECTs can operate as electronic switches, at very low voltages

(<1 V), by performing an ion-to-electron transduction. Hence, OECTs can be used as electronic switches or components of logic gates [1], but also in sensoristics or as a tool for the monitoring of biological-driven phenomena, such as enzymatic interactions [2] and processes in presence of cells [3] and for the detection of biomolecules, such as specific antigens [4] or DNA [5].

The OECT channel is made of an organic semiconducting material, acting as the transducer element, defined by two metallic electrodes. The device channel is separated from the gate electrode by means of a gate-electrolyte.

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Importantly, the gate-electrolyte exploits ionic liquids or electrochemical species in solution, making the OECT a versatile tool suitable for the operation in liquid phase. The ion-to-electron transduction is based on the injection of ionic species from the electrolyte towards the underneath polymeric film upon the application of a gate bias. The typical OECTs exploit channels made of a highly doped polymer, the p-type oxidized poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) and, specifically, their operation result in a channel current lowering due to the interaction between PEDOT:PSS and cations. Therefore, the injection of positively charged species into the polymer represents the key issue for the comprehension of OECTs working features.

Despite the wide literature production showing the potentialities of OECTs, relatively few works have been directly focused on the comprehension of their working features [6–9] and optimal operating conditions [10–13], unlike it happens in the case of the widely studied Organic Field Effect Transistors (OFETs) [14].

Among the studies presented so far, our attention has been attracted by two factors that can influence the OECT response. First, in real OECTs a nonlinear de-doping is expected to take place along the device channel as a consequence of the applied channel bias voltage [15]. This nonlinear de-doping profile is confirmed by the gradient in the electrochromic effect, i.e. a polymer color change due to doping and/or de-doping processes taking place upon the application of a suitable bias between the electrode immersed in the electrolyte and the polymeric channel. In the case of PEDOT:PSS, a bright blue color in correspondence of stronger de-doped portions of the channel has been observed [15]. A second aspect that one should take into account in the context of the OECT operation regards the marked tendency to adsorb water showed by PEDOT:PSS, due to the hydrophilic character of the PSS fraction. A significant film swelling upon hydration has been shown to induce a strong change in conductivity [16,17]. In addition, the formation of some liquid pathways, connected to the hydration of PEDOT:PSS and occupying most of the polymer bulk, has been demonstrated to assist the transport of ions injected within the film [18]. In particular, the hydration of the polymer has been demonstrated to take place on a timescale ranging from tens to hundred seconds, leading to the formation of the liquid pathways assisting the drift of metallic cations in the film. Cations are also expected to drift in regions of the polymer where, due to the hydration timescale and the device geometry, no penetration of liquid has taken place yet. In this case, cationic drift is related to the formation of the so-called doping front [19,20], that corresponds to the insulator/conductor interface propagating in the polymer under the effect of a bias voltage.

In the light of the above considerations, we expect that the features of the de-doping process can be connected to the electrolyte positioning within the OECT active channel. In this respect, it is worthwhile wondering how the film formation of liquid pathways eventually influences the efficiency of the de-doping processes. Therefore, in order to shed light on the role played by film hydration in the context of the device operation, in this work we study

the influence of the electrolyte positioning with respect to the device electrodes on its response. To this end, we measure the typical OECT kinetic curves as a function of the electrolyte positioning within the region comprised between the active channel electrodes. Specifically, these curves are used for evaluating the OECT sensing capability and consist in the acquisition of the channel current as a function of time, recorded by fixing the channel voltage and by varying in a step-like mode the voltage between the gate electrode and the grounded channel electrode. The active channel in this work is deposited on a superhydrophobic substrate aimed at controlling the confinement of some electrolyte microvolumetric drops next to the source and drain electrodes. In this way, the impact of the combined effect of the electrolyte positioning and adsorption can be efficiently studied.

2. OECT: device operation and materials

In this section, we provide a more detailed overview of both the transport properties of the p-type PEDOT:PSS and the OECT working mechanisms.

The prototypal conducting polymer used in OECTs is the oxidized PEDOT:PSS. PEDOT PSS is a highly porous material showing a morphology made of two separated phases consisting of PEDOT-rich conducting clusters surrounded by PSS⁻ insulating shells [21]. When used in the fabrication of OECTs, PEDOT:PSS is treated by 'secondary doping' methods through a treatment with inert organic solvents, such as ethylene glycol (EG), in order to significantly enhance its intrinsic conductivity [22]. The enhancement of its holes charge transport, strongly desired in OECT applications, is related to a rearrangement of the hydrophobic PEDOT-clusters and hydrophilic PSS shells. In particular, the addition of EG causes a conformational change of the PEDOT chains, from coiled to linear along the in-plane direction [23], resulting in a lengthening of PEDOT-rich clusters and a constriction of PSS insulating barriers [24], favoring thus the inter-cluster charge carrier hopping. Even if the charge transport in the polymer is enhanced, allowing the operation of the OECT at very low voltages, the hydrophilic character of PSS-based shells induces a strong effect in terms of the overall hydration film properties. In this respect, it has been widely reported that the polymer hydration plays an important role in terms of PEDOT:PSS hole transport. In fact, a strong conductivity change, depending on the materials processing conditions and on the percentage content of the relative humidity in air, occurs [16,25,26].

The general scheme of an OECT consists of a conducting polymeric channel equipped with a couple of electrodes, named source (grounded) and drain, in contact with an electrolyte having an electrode, the gate, immersed in it. The device can be seen as the combination of an ionic circuit, specifically an electrochemical cell where the conducting polymer acts as the working electrode and the gate is the reference electrode, and an electronic one, the polymer, made of a series of infinitesimal resistors between the source and drain electrodes. The interaction between circuits is promoted at the interface between the polymer and the electrolyte [27].

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