



# Electrochemical doping for lowering contact barriers in organic field effect transistors

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## ABSTRACT

By electrochemically p-doping pentacene in the vicinity of the source-drain electrodes in organic field effect transistors the injection barrier for holes is decreased. The focus of this work is put on the influence of the p-doping process on the transistor performance. Cyclic voltammetry performed on a pentacene based transistor exhibits a reversible p-doping response. This doped state is evoked at the transistor injection electrodes. An improvement is observed when comparing transistor characteristics before and after the doping process apparent by an improved transistor on-current. This effect is reflected in the analysis of the contact resistances of the devices.

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## 1. Introduction

Organic fused ring molecules such as pentacene excel in terms of their interesting electronic properties as thin film semiconductors with charge carrier mobilities in the range of up to  $23 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  [1–3]. The implementation of organic small molecules as active material to organic field effect transistors (OFETs) and furthermore to complex and flexible integrated circuits (ICs) has shown great progress over the last decade [4–7]. However, fast, organic device circuits operating in MHz regime may still not be feasible with state-of-the-art organic transistors. Two major limitations are revealed addressing the comparatively low mobilities of organic disordered thin films on the one hand and contact barriers on the other hand. While optimization of deposition techniques has yielded highly oriented, crystalline organic films with carrier mobilities beyond

$10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  [8] contact issues turn out to be more complex. In this work we concern the improvement of metal-organic junctions in OFETs. Implemented as active layers organic  $\pi$ -conjugated molecules are intrinsic semiconductors. Injection properties are crucial, as OFETs switch between the accumulation and the depletion of injected charge carriers. At the metal – intrinsic organic semiconductor junctions neither ohmic contacts have been observed, nor clear indications for a Mott-Schottky limit have been shown. There is agreement that the charge injection is limited by significant contact barriers. Implemented in a device structure gold-pentacene junctions for example exhibit contact resistances in the range of  $\text{M}\Omega$  [9,10]. An integration of OFETs to complex circuit arrays will cumulate the contact barrier effects, leading to high driving voltages, high threshold voltages and parasitic capacitances. Thus state-of-the-art organic electronic circuits are limited to operation in the kHz regime [4].

The work presented here targets the problem of barriers at metal-organic junctions. As discussed the energy level alignment at organic-metal interfaces is unfavorable for

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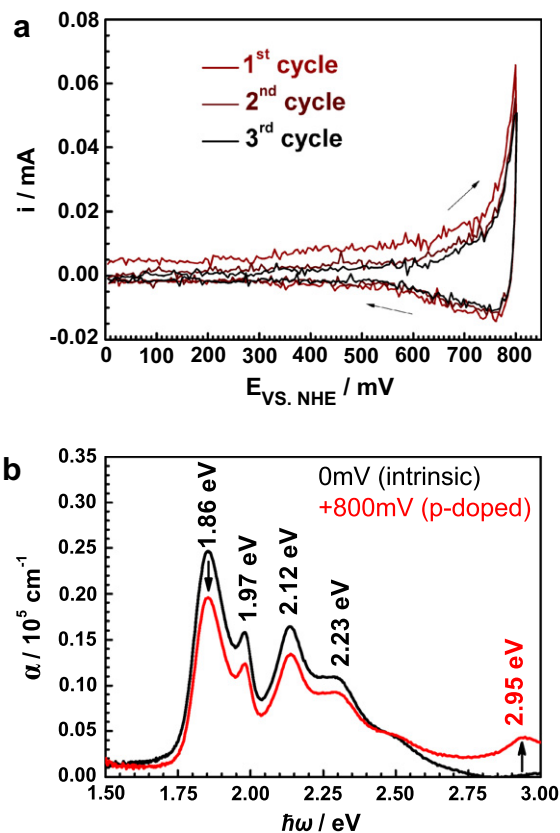
injection. In particular the frequently studied pentacene-gold interface exhibits a significant barrier for holes [11,12]. The energy offset varies between 0.5 and 0.9 eV [13]. In order to reduce them the energy alignment at contacts has to be optimized. One strategy is to apply thin organic interlayers, for instance self assembled molecules (SAMs), which selectively attach to the contact metal. It has been demonstrated that SAM treatment enhances the transistor currents [8,14] and that the energy levels can be tuned towards a more favorable situation for injection [15].

In this work we introduce electrochemical contact doping as a novel strategy to improve metal-organic contacts. Here the hole injection is enhanced by electrochemically treating as-prepared OFETs. The approach tends to a p-doping of organic molecules adjacent to the injection electrodes. The goal is to create a *metal to doped to intrinsic* junction (metal-p-i junction). P-doping pins the Fermi level to the contact metal forming a narrow Schottky-junction and an improvement of hole injection [16].

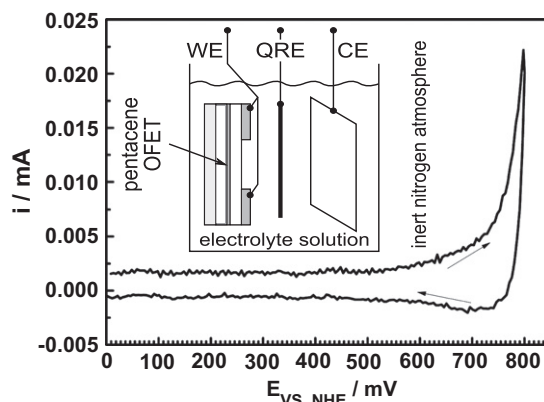
The crucial part is to selectively p-dope molecules just in the vicinity of the electrodes. Molecules in the channel must not be affected. Electrochemical doping particularly satisfies this requirement, as it impacts the proximate electrode surrounding solely. Thus contact-doped OFETs render a MOSFET-related structure, where the source-drain electrodes face a narrow heavily p-doped semiconductor layer. The OFET channel itself is unaffected by the treatment. The metal-p-i junction is evoked by applying electrochemical doping directly to an as-prepared pentacene FET. We control the p-doping concentration by a profound electrochemical study. All parameters for creating the metal-p-i junction are presented chronologically step-by-step starting from electrochemistry and the contact doping followed by the transistor characteristics and finally a careful analysis of the contact resistance.

## 2. Experimental

Bottom-gate/top-contact (staggered) OFETs were used for a complementary study. We chose materials with high performance and good resistibility against electrochemical treatment. Aluminium deposited by vacuum deposition on a glass substrate served as gate electrode. For the gate dielectric 30 nm alumina was grown by anodic oxidation. The oxide was covered with divinyltetramethyldisiloxane – bis(benzo-cyclobutene) (BCB, Dow Chemicals). The cross-linked resin is fabricated by spin coating from a 1 wt.% precursor solution in mesitylene at 1500 rpm followed by an annealing step at 180 °C. The resulting hybrid dielectric has a geometric capacitance of 55 nF cm<sup>-2</sup> [17]. Anodic alumina has excellent properties in terms of high dielectric breakdown fields [18–20]. BCB is applied for reducing shallow traps, for enhancing surface properties and for its thermal and chemical stability [21]. For the semiconductor 50 nm of pentacene (Aldrich) were applied by thermal evaporation at a constant substrate temperature of 50 °C during deposition and a rate of 0.4 Å s<sup>-1</sup>. Gold source/drain contacts of 80 nm thickness were evaporated using a shadow mask with a channel width of 1 mm and a channel length varying from 40–120 μm.



**Fig. 1.** (a) Cyclic voltammogram (CV) showing the reversible p-doping of pentacene thin film on ITO. (b) In-situ absorption recorded for intrinsic (black) and p-doped (red) pentacene. A new feature arises at 2.95 eV and the intensity of the four characteristic absorption features decrease. The relative drop in intensity is proportional to a p-doping concentration of approximately 1%. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** Applying electrochemical contact doping to an OFET: Cyclic voltammogram showing the p-doping of pentacene recorded on an as prepared transistor. The inset shows the schematic of the setup, where source/drain electrodes in the staggered OFET act as the working electrode.

The electrochemical treatment and the electrical characterization were performed under nitrogen atmosphere, for the electrolyte solution an aprotic system was used

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