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Interface dipole: Effects on threshold voltage and mobility for both amorphous and poly-crystalline organic field effect transistors

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ABSTRACT

We report a detailed comparison on the role of a self-assembled monolayer (SAM) of dipolar molecules on the threshold voltage and charge carrier mobility of organic field-effect transistor (OFET) made of both amorphous and polycrystalline organic semiconductors. We show that the same relationship between the threshold voltage and the dipole-induced charges in the SAM holds when both types of devices are fabricated on strictly identical base substrates. Charge carrier mobilities, almost constant for amorphous OFET, are not affected by the dipole in the SAMs, while for polycrystalline OFET (pentacene) the large variation of charge carrier mobilities is related to change in the organic film structure (mostly grain size).

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1. Introduction

The control of the threshold voltage (V_T) of organic field-effect transistors (OFET) is still a key problem. Many groups have reported that the intercalation of a selfassembled monolayer (SAM) of molecules bearing a dipole between the gate dielectric and the organic semiconducting (OSC) film is an efficient way to modulate V_T over a large range of values (more than few tens of Volt) [1–11]. However, other OFET parameters (charge carrier mobility, on/off current ratio) can also be impacted by the SAMs due to their effects on (i) changes in the molecular orientation in the OSC, (ii) neutralization of surface defects, (iii) modification of surface roughness, interface dipole and surface energy. In most cases, a combination of these effects is likely, some of them being certainly dependent

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on both the dielectrics and the OSCs. As a consequence, several transistors parameters are modified simultaneously, and this feature prevents a clear understanding of the effect of the SAMs. We have previously demonstrated [9] that, in the case of an amorphous OSC, we can tune the threshold voltage alone, while keeping nearly unchanged the other electrical properties (hole carrier mobility, on/off ratio, subthreshold swing). Moreover, it is also difficult to precisely and quantitatively compare the results reported by various groups since, gate dielectric materials, OFET geometries, source and drain technologies are not systematically similar. For example, while a significant effect of SAMs on V_T has been reported in the literature, experimental results on the effect of the SAMs on the charge carrier mobility are sparse or not discussed with respect to the dipole moment of molecules [6–9].

Here, we report on a detailed comparison on the effects of dipolar SAMs on V_T and charge carrier mobility for both amorphous (polytriarylamine, PTAA) and polycrystalline (pentacene, P5) OFET made on the same SAMs and the same transistor base substrates (same gate dielectric and





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source–drain electrode technology and geometry). We show that the same V_T shifts are induced by the SAMs (10 different molecules with dipole moments ranging from –2D to 7D) for both PTAA and P5 OFETs. In particular, we observe that a linear correlation is obtained between V_T and the dipole-induced charge in the SAM (Q_{SAM}) instead of the net dipole moment of the molecule. We also observe that the dipole moments of the molecules used in the SAMs have no significant effect on the charge carrier mobility.

2. Materials and methods

2.1. Synthesis, monolayers, device fabrication

We used 10 molecules for SAM fabrication on the SiO_2 gate dielectric (Fig. 1). These molecules have dipole moments (along their long axis) ranging between ca. -2D and 7D (see Table 1, and calculation details below).

2.1.1. Molecules

PFTS ($C_6F_5-(CH_2)_3-SiCl_3$; 3-pentafluorophenyl-propyltrichloroSilane), MPTS (HS-(CH_2)_3-Si(OMe)_3; 3-mercaptopropyl-trimethoxysilane), APTS (H₂N-(CH₂)_3-Si(OEt)_3; 3-aminopropyl-triethoxysilane) and PhC₄ ($C_6H_5-(CH_2)_4-$ SiCl₃; 4-phenyl-butyl-trichlorosilane) were supplied by ABCR and used as received. TAATS (4-(Ph₂N)-C₆H₄-NHC(O)NH-(CH₂)_3-Si(OEt)_3; (1-(4-(diphenylamino)-phenyl)-3-(2-(triethoxysilyl)-propyl)-urea). This triarylamine derivative was synthesized in three steps according to a previously reported procedure [9]. PhC₁₀ and Py were obtained through multistep synthesis directly on the surface according to a published protocol [12]. tBuPh (1-(4-tert-butylbenzyl)-3-(3-(triethoxysilyl)propyl)urea), tBuPhNO₂ (1-(4-tert-butyl-2,6-dinitrobenzyl)-3-(3-(triethoxysilyl)propyl)urea) and tBuPhNH₂ (1-(2,6-diamino-4-tert-butylbenzyl)-3-(3-(triethoxysilyl)propyl)urea were obtained by multistep synthesis as described in the supporting information.

2.1.2. Self-assembled monolayers

Synthesis and characterization of the SAMs prepared from PFTS, MPTS, APTS, TAATS, Py and PhC₁₀ were described in our previous publications [9,12]. In brief, n⁺doped silicon wafers were freshly cleaned and oxidized to provide a dense array of silanol groups (=Si-OH), which are the anchoring sites for the organosilane molecules. Substrates were first cleaned by sonication in acetone. isopropanol then dichloromethane for 5 min. Wafers were dried under nitrogen flow then they were dipped into a freshly prepared piranha solution $(H_2SO_4-H_2O_2 2:1 v/v)$ at 100 °C for 30 min, or submitted to an oxygen plasma treatment (20 mTorr, 10 sccm O₂, 10 W, 300 s). They were rinsed thoroughly with deionized water then were dried under nitrogen stream. Caution: the piranha solution reacts violently with organic chemicals. Consequently, it should be handled with extreme care. The silanization reactions were carried out at room temperature in a nitrogen glovebox (<1 ppm H₂O and O₂). For tBuPh, tBuPhNH₂, and tBuPhNO₂ SAMs, the freshly cleaned oxidized silicon substrates were immersed in a 10^{-3} M solution of the corresponding organosilane in anhydrous toluene then samples were kept in the dark for 4 days. Concerning PhC₄ SAM, the freshly



Fig. 1. Chemical structures of dipolar molecules used in SAMs.

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