



# High mobility organic field-effect transistors based on defect-free regioregular poly(3-hexylthiophene-2,5-diyl)

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## ABSTRACT

We report on organic field-effect transistors (OFETs) prepared using defect free (100% regioregular) poly(3-hexylthiophene-2,5-diyl) (DF-P3HT) as semiconductor and cross-linked poly(vinyl alcohol) (cr-PVA) as gate insulator. High field-effect mobility ( $\mu_{\text{FET}}$ ) of  $1.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  is obtained and attributed to the absence of regioregularity defects. These transistors have transconductance of  $0.35 \mu\text{S}$  and the DF-P3HT film shows larger crystallites ( $\sim 80 \text{ \AA}$ ) than a highly regioregular ( $>98\%$ ) material ( $\sim 32 \text{ \AA}$ ). Devices with increased  $\mu_{\text{FET}}$  ( $2.8 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) could be obtained at the expense of the On-Off current ratio, which was reduced by one order of magnitude, when poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) treatment was applied to the dielectric surface. Our results suggest that the interaction of charged sites at the dielectric surface with regioregularity defects of the P3HT is an important factor degrading  $\mu_{\text{FET}}$  even at very low concentration of regioregularity defects.

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## 1. Introduction

In spite of some recently reported high field-effect mobilities  $\mu_{\text{FET}}$  (larger than  $20 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) [1] in macroscopically aligned conjugated polymers, researchers still find the mobility as one of the limiting factors for commercial application of organic field-effect transistors (OFETs) [2,3]. Without any treatment or extra fabrication step, polymer OFETs usually still demonstrate  $\mu_{\text{FET}}$  of only  $0.01\text{--}0.15 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  e.g. using poly(9,9'-dioctylfluorene-co-bithiophene) (F8T2) [1–6], poly[5,5'-bis(3-dodecyl-2-thienyl)-2,2'-bithiophene] (PQT) [7–10], or regioregular poly(3-hexylthiophene-2,5-diyl) (rr-P3HT) [11–13]. In previous studies,  $\mu_{\text{FET}}$  of P3HT could be found to be related to the casting solvent [11], carrier concentration [14], number average molecular weight [15,16], and regioregularity [17]. Strategies based on extra steps during film-formation method to increase  $\mu_{\text{FET}}$  and correspondingly to improve OFET performance have recently been developed and proved to be effective [18–20].

P3HT is one of the most experimentally investigated material regarding OFET [12–20] and organic solar cells [21] applications, owing to its simple chemical structure, high chemical stability, good semiconducting properties, spectral absorption matching solar spectrum, controlled synthetic method and wide commercial availability. The development of synthesis methods has now enabled production of highly regioregular P3HT with good control of molecular weight and polydispersity [22–24].

Effects of regioregularity and molecular weight ( $M_n$ ) on the morphology and crystalline structure of P3HT have been studied in OFET and solar cell applications context [16,25,26]. It is well established that the degree of regioregularity of rr-P3HT strongly affects its ability to form highly ordered domains [27]. Regiorandom P3HT is well known to suffer from torsional disorder and packing irregularities, showing inter-chain spacing of around  $4\text{--}4.5 \text{ \AA}$ , as determined through X-ray diffraction (XRD) patterns at room temperature [28]. OFETs prepared with regiorandom P3HT have been reported to have mobilities in the range of  $10^{-5}\text{--}10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  [29], while in the case of 91% rr-P3HT, relatively higher mobilities in the range of  $0.05\text{--}0.10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  were observed [17,30]. Even small variations of regioregularity between 90 and

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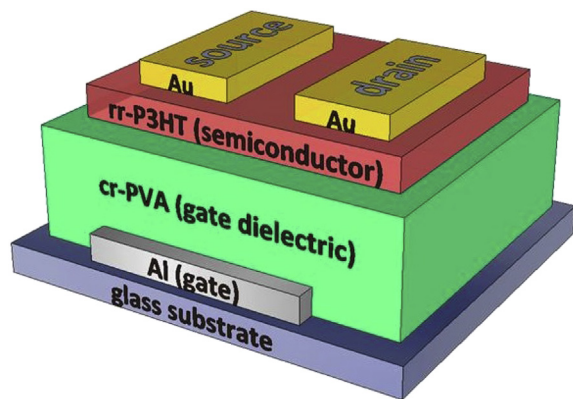


Fig. 1. Schematic structure of the OFETs used in this work.

96% significantly affect device performance [17,31]. Higher regioregularity results in an increased number of head to tail linkages of polymer units, in principle allowing considerable overlapping of the delocalized molecular orbitals of neighboring molecules, hence, facilitating charge transport [32].

In this study we investigate Al-bottom gate Au-top-source and –drain OFETs (see Fig. 1) prepared with cross-linked poly(vinyl alcohol) (cr-PVA) as gate dielectric and DF-P3HT (defect-free 100% regioregular P3HT) as semiconductor (see Fig. 2). We maintain the film processing conditions as used in earlier studies of some of us [18–20]. As a consequence, regioregularity is expected to be the major primary factor that leads to a different result of  $\mu_{FET}$  and OFET performance. An expression derived from drift-diffusion model is used to estimate the effective channel thickness from source to drain near the insulator/semiconductor interface, which corresponds to the charge transport bottleneck region along the channel.  $\mu_{FET}$  was hence evaluated as a function of gate-voltage ( $V_{GS}$ ) and distance from the insulator/semiconductor interface in the bottleneck.

We additionally test the application of PEDOT:PSS(poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)) and anionic and cationic surfactant treatment to the OFET aiming at obtaining enhancement in charge transport and  $\mu_{FET}$ . Recent reports have indicated that the improvement of the gate insulator/channel semiconductor interface properties using chemical treatments [18–20,33] to passivate charge traps can increase the output current at low voltages.

## 2. Experimental

DF-P3HT was synthesized via Nickel Zero Catalyzed Kumada Catalyst-Transfer Polymerization applying continuous flow synthesis (see Supplementary Material for details). The number

average molecular weight ( $M_n$ ) of 41 kDa was confirmed by Size Exclusion Chromatography (SEC) against tetrahydrofuran (THF) as mobile phase and polystyrene as standard. Regioregularity of the sample used in transistor preparation was confirmed using  $^1\text{H}$  NMR characterization at 303 K of a 10 mg/mL P3HT solution in  $\text{CDCl}_3$  on a Bruker AVANCE III 600 NMR spectrometer operating at 14.1 T, through the absence of the peak at 7.00 ppm [34]. Electronic grade regioregular poly(3-hexylthiophene-2,5-diyl), EG-rr-P3HT, was purchased from Sigma-Aldrich (CAS #156074-98-5,  $M_n = 54,000\text{--}75,000$  Da, >98% head-to-tail regioregularity [35]) and was used for comparison in some devices.

Bottom-gate top-contact OFETs preparation was carried on soda lime glass substrates that were sequentially cleaned using acetone, ultra-pure deionized water (resistivity >18.2  $\text{M}\Omega\text{ cm}$ ) and isopropanol, for 20 min each, in ultrasonic bath. A 100 nm thick Al layer, patterned using a shadow mask, was evaporated on top of the glass substrates at  $4 \times 10^{-6}$  Torr to form the gate electrode. Poly(vinyl alcohol) (PVA) (Sigma-Aldrich,  $M_w = 130$  kDa) dissolved in water at a concentration of 60 mg/mL was stirred for 1 h at 60 °C after which ammonium dichromate (AD), used as a cross-linking agent, was added to the PVA solution at AD:PVA w/w ratio of 0.25. The deposition of the gate insulator layer was carried out by spin-coating PVA:AD over the Al layer at a rotation speed of 4000 rpm for 60 s. The Al/PVA films were then exposed to ultraviolet (wavelength of 365 nm, power of 8 W) for 10 min and annealed in vacuum at 100 °C for 1 h, resulting in crosslinked PVA (cr-PVA). The thickness of cr-PVA layer was measured as 300 nm, using a Bruker DektakXT surface profiler.

When explicitly indicated in the text, samples included a PEDOT:PSS treatment made with an aged solution of PEDOT:PSS (Baytron P, 11 years old, conductivity of  $\sim 0.07\text{ Scm}^{-1}$ ) [18,36]. The PEDOT:PSS was spin-coated on top of cr-PVA layer at a speed of 4000 rpm for 60 s and sequentially annealed in vacuum for 10 min at 100 °C, resulting a 20–30 nm thick layer, as estimated using profilometry and optical absorbance.

For the formation of semiconducting layer, 7 mg/mL P3HT (DF-P3HT or EG-rr-P3HT) solution in toluene was magnetically stirred at 60 °C for 1 h and spin coated onto the cr-PVA at 1500 rpm for 60 s to form a 40 nm thick layer. The P3HT films were then annealed at 100 °C for 30 min in vacuum. Top electrode formation was carried out by evaporating a 100 nm thick gold (Au) layer, patterned using shadow mask, on top of the P3HT film at  $4 \times 10^{-6}$  Torr. Channel length and width of OFETs were 100  $\mu\text{m}$  and 2 mm respectively.

An Agilent 4284-A LCR meter operating at 1 kHz was used to measure capacitance in Al/cr-PVA/Au sandwich structure. OFET characterization was done in dark ambient environment using a Keithley 2602 dual source meter.

X-ray diffraction (XRD) measurements were performed in an XRD 7000 Shimadzu X-Ray Diffractometer, using  $\text{Cu K}\alpha$  X-ray radiation with an incident angle of  $0.2^\circ$ . The diffraction patterns were calibrated relative to the Si(111) diffraction peak of the powder

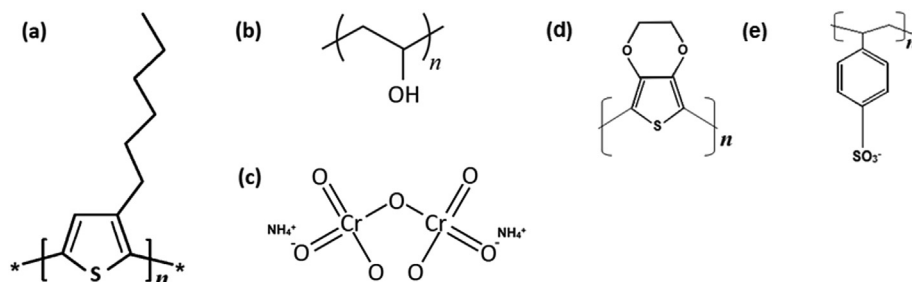


Fig. 2. Structures of the chemical compounds used in this work: (a) P3HT; (b) PVA; (c) ammonium dichromate (AD); (d) PEDOT and; (e) PSS.

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