

Ultra-high mobility in defect-free poly(3-hexylthiophene-2,5-diyl) field-effect transistors through supra-molecular alignment

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ABSTRACT

Performance enhancement of organic field-effect transistors (OFETs) based on solution-processable conjugated polymers (CPs) holds critical significance for the realization of cost-effective commercial applications such as organic light-emitting diode displays. One of the most critical performance parameters is the charge-carrier field-effect mobility (μ_{FET}) that is significantly influenced by the molecular arrangement in a CP. In this article, floating film transfer method (FTM) is utilized for the deposition of a CP—defect-free poly(3-hexylthiophene-2,5-diyl) (DF-P3HT)—which results in the formation of aligned supra-molecular assemblies. When applied as the active layer in OFET devices, μ_{FET} reaching as high as 8.0 cm²/V.s (6.3 cm²/V.s on average) is obtained. The value of μ_{FET} observed in the current study is the highest value reported so far for P3HT based OFETs (~5 times higher as compared to when DF-P3HT is deposited using spin coating).

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1. Introduction

Solution-processable polymers offer a viable alternative to be used as semiconductors in field-effect transistor (FET) devices because of cost-effectiveness and easy fabrication techniques. Polymeric semiconductor based FETs have now surpassed μ_{FET} values previously demonstrated using amorphous silicon (a-Si) thin-film transistors (TFTs) [1–5]. This broadens the possibilities of the integration of polymeric semiconductor based OFETs in electronic applications such as radio frequency identification tags (RFID) and organic light-emitting diode (OLED) displays [6]. For instance in the case of OLED displays, to operate the backplane circuitry at a sufficient speed, a minimum transistor mobility requirement of >1.5 cm²/V.s and a more stringent mobility requirement of 5–10 cm²/V.s has been estimated [7,8]. Most of the currently manufactured glass-based OLED displays consist of polycrystalline Si TFTs that can achieve μ_{FET} values > 20–50 cm²/V.s [7]. However, in order to switch to flexible displays, the mechanical properties of polymeric semiconductor based OFETs make them a better suited candidate to be integrated in the backplane of OLED displays [9–12]. Regioregular poly(3-hexylthiophene-2,5-diyl) (rr-

P3HT) is one of the most experimented conjugated polymers (CPs) for FET devices (chemical structure shown in Fig. 1) [13,14]. Because of the ability of rr-P3HT molecules to self-assemble in both solution and solid-state, three dimensional ordering of polymer chains can be obtained which results in high mobility of charge-carriers [15]. Recently, μ_{FET} of 1.2 cm²/V.s was demonstrated in defect-free P3HT (DF-P3HT) based OFETs [16]. Although this value is close to the minimum mobility requirement to operate the OLED backplane circuitry, it does not achieve the more stringent mobility requirement of 5–10 cm²/V.s [7,8]. In this article we show that this landmark can be achieved by optimizing the deposition method of DF-P3HT. We also include an additional insulator surface treatment step (with vitamin C) in pursuit of obtaining passivation of charge traps at the channel interface for further improvement of μ_{FET} .

Previously it has been shown that μ_{FET} of P3HT based FETs can be considerably improved by choosing better deposition techniques that favor the molecular order and/or molecular alignment [17–25]. One of these techniques is the floating film transfer method (FTM). Some reports on the deposition of P3HT using FTM can be found in the literature [17,26–30]. Different from spin coating deposition, FTM results in the formation of a thin floating film of P3HT on top of a viscous liquid substrate. Control over a few important parameters yields a uniform floating film consisting of stretched and aligned macromolecules. In principle, polymer solution is dropped on top of a viscous ethylene glycol (Eg) and

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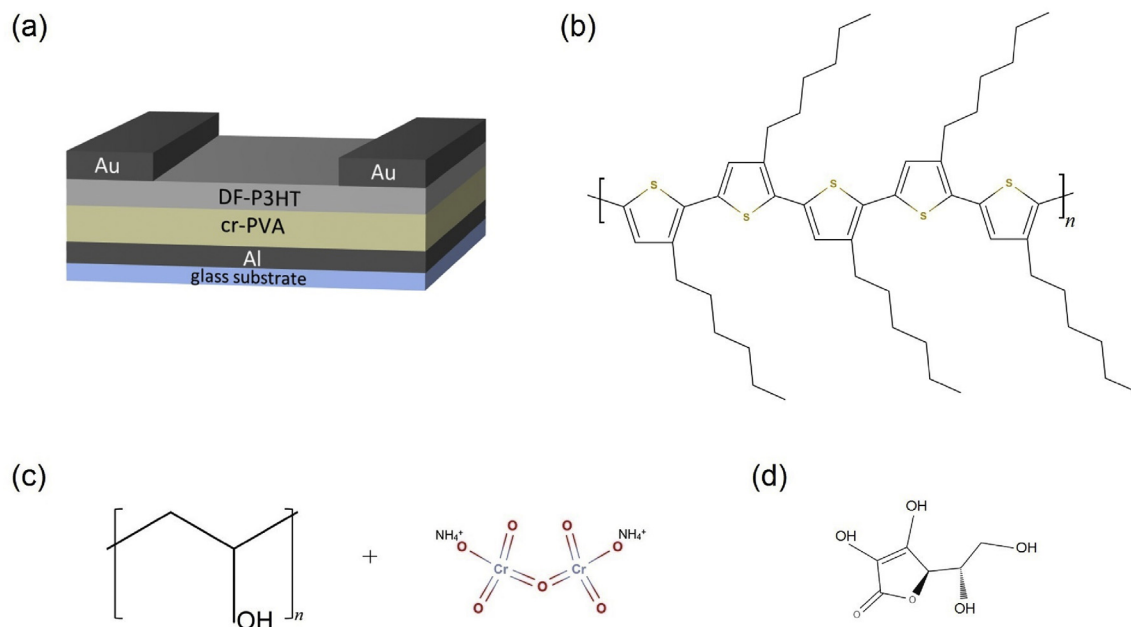


Fig. 1. a) Schematic structure of the OFET devices used in this work. Chemical structures of the used materials: b) P3HT; c) PVA + AD and; d) ascorbic acid (vitamin C).

glycerol (G1) mixture (3:1), which expands outwards, followed by rapid solidification and formation of a floating polymer film (Fig. 2a). The propagation of polymer solution is controlled by the viscosity of Eg:G1 hydrophilic liquid substrate and the molecular interaction between the top layers of hydrophilic liquid substrate and the hydrophobic polymer molecules [27,30]. The viscosity of Eg:G1 hydrophilic liquid substrate acts as a compressive external force and hinders the solution expansion [27,30]. The resultant force due to simultaneous compression/expansion allows the alignment of polymer molecules perpendicular to the spreading direction of polymer solution [27,30]. The floating film is then transferred onto the desired substrate by stamping, which permits the choice of aligning polymer molecules either parallel or perpendicular to the stamping direction. FTM has also been experimented successfully on a few other polymeric materials including poly(3,3'-didodecyl-quaterthiophene) (PQT-C12), poly[2,5-bis(3-tetradecylthiophen-2-yl)thieno [3,2-b]thiophene] (PBTTC-C14) and poly(9,9'-dioctyl-fluorene-alt-bithiophene) copolymer (F8T2) [27,31,32]. In a recent work, Pandey et al. demonstrated that μ_{FET} of non-regioregular P3HT (NR-P3HT) based OFETs can be improved by two orders of magnitude, to

$3.4 \times 10^{-3} \text{ cm}^2/\text{V.s}$, when NR-P3HT is deposited by FTM as compared to when deposited by spin coating [17]. Motivated by that report, we applied FTM to deposit DF-P3HT for OFET fabrication. Correspondingly, we have been able to achieve μ_{FET} value up to $8 \text{ cm}^2/\text{V.s}$, which to our knowledge is the highest ever reported for P3HT based FETs.

2. Experimental section

OFETs were prepared in bottom-gate top-contact geometry on top of glass substrates—schematic structure is shown in Fig. 1a. The glass substrates were first cleaned sequentially with acetone, ultra-pure deionized water and isopropanol (20 min each), followed by UV/Ozone treatment for 30 min. Gate electrode was formed by thermal evaporation of $\sim 50 \text{ nm}$ aluminum (Al) layer at a base pressure of $\sim 10^{-6}$ Torr. Poly(vinyl alcohol) (PVA) (chemical structure shown in Fig. 1) obtained from Sigma-Aldrich (M_w : 130 kDa) was dissolved in ultra-pure deionized water at a concentration of 60 mg/mL followed by magnetic stirring for 120 min at 60°C . Sequentially, ammonium dichromate (AD) (chemical structure shown in Fig. 1) was added to the PVA solution

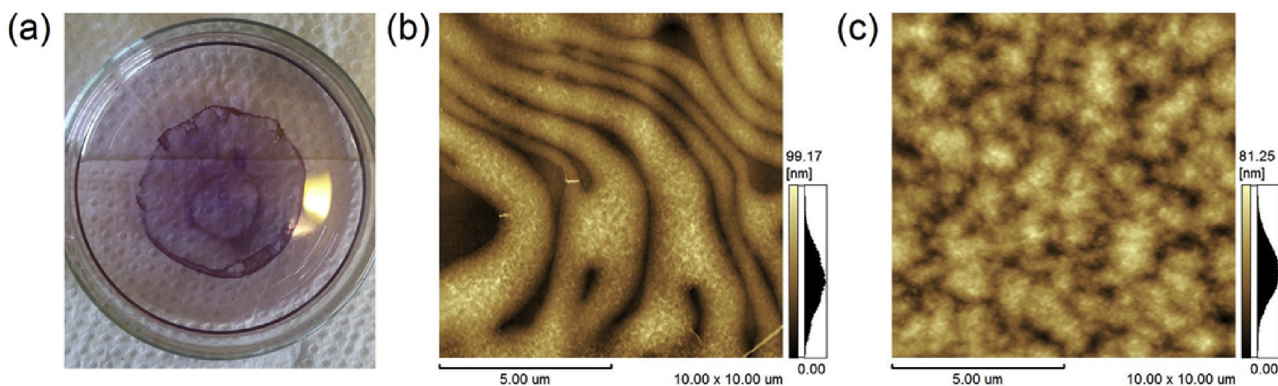


Fig. 2. a) Image of solidified DF-P3HT film floating over Eg:G1 (3:1). Tapping mode AFM images of DF-P3HT thin films deposited over cr-PVA substrate; b) by FTM deposition (R_{rms} : 15 nm), and; c) by spin coating (R_{rms} : 12 nm).

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