



Solvent-dependent electrical properties improvement of organic field-effect transistor based on disordered conjugated polymer/insulator blends

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ABSTRACT

Poly(3-hexylthiophene) (P3HT):polystyrene (PS) blends prepared from different solvents were used as the semiconducting layers in organic field-effect transistors (OFETs). The relationship between the molecular arrangement, aggregation and charge transport in P3HT:PS blends with the boil-points and solubility of different solvents were systematically analyzed. Topographic investigation by atomic force microscopy carried out on blends with various solvents revealed a lateral phase separation of the two components, which was strongly influenced by the choice of solvent. Although the blend film was highly disordered, the OFETs performed as well as that with the pristine P3HT films. Moreover, 1,2-dichlorobenzene with the high boiling temperature was found to be more desirable for achieving distinct lateral aggregation of P3HT in the blend film, which led to the superior performance of the OFET. X-ray diffraction analyses and optical absorption measurements revealed that PS matrix made the arrangement of P3HT molecules more disordered, but introduced a more efficient intermolecular coupling of P3HT molecules. This could be ascribed to that, the PS matrix made the aggregating rates of the P3HT molecules slower, inducing a uniform distribution of P3HT molecules in the blend film. This was beneficial for the efficient charge transport in the blend film, which provided a pathway between the aggregations and acted as a tie-molecule in the blend film. Consequently, the field-effect mobility of the optimized OFET (1.6 wt% P3HT) increased three times and the current on/off ratio increased two times compared to that of the pure P3HT (8 wt%). This work will fill the gap of current research about semiconductor/insulator blend transistor.

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1. Introduction

Organic field-effect transistors (OFETs) have been extensively studied over the last several years owing to their potential impact in a wide range of electronic applications, such as drivers for flat-panel displays [1–3], complementary circuits [4,5], and various kinds of sensors [6–8]. Moreover, OFETs based on solution-processed semiconducting conjugated polymers have achieved interesting technological attributes, such as compatibility with simple direct-write printing techniques, general low-cost manufacturing approaches, and flexible plastic substrates [9]. However, owing to the presence of

amorphous regions in polymer films, thus, polymer-based OFETs usually exhibit lower charge carrier mobility comparing to devices based on small organic molecules [10].

To date, many polymer semiconductors with novel building blocks were synthesized to improve charge carrier mobility [11]. The charge carrier mobilities of polymeric materials have exceeded $1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, such as poly(bis-alkylthiophenyl thienothiophene) (PBTTT) [12], poly(cyclopentadithiophene-benzothiadiazole) [13,14], isoindigo-based polymers and diketopyrrolopyrrole (DPP)-based polymers [15–17]. Ong and Liu reported a result of breakthrough, which was an achievement of the charge carrier mobility as high as $10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ from poly(dithienylthieno[3,2-b]thiophene-DPP) [18]. Nevertheless, for a widespread application of these materials, easy processing, air stability and material costs are

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important issues that need to be improved. Nowadays, only a few of these polymer semiconductors can meet the requirement of high mobility and operational stability for logic circuits application in active matrix displays or low-cost intelligent labels [19].

Another efficient way to obtain the required properties for OFETs is to blend two or more materials with the aim of combining the advantageous properties of each, thus exceeding the performance of the individual components [20–24]. Among these blend film system, blending of semiconducting and insulating polymers have shown greater charge carrier mobilities and on/off ratios than the corresponding pure semiconducting polymers. Wang et al. reported that OFETs based on a poly(3-hexylthiophene) (P3HT)/poly(methyl methacrylate) (PMMA) (1/39 w/w) film prepared from a 4 wt% blend solution, showed a field-effect mobility enhancement from 0.001 to 0.022 cm² V⁻¹s⁻¹ [25]. Lu et al. demonstrated a polymer blend system with only 5 wt% of P3HT and 95% polystyrene, affording impressive charge mobilities as high as 0.5 cm² V⁻¹s⁻¹ [26]. Note, comparing to OFETs with neat organic semiconducting layer, these high performance blend OFETs can reduce cost significantly, since they need a much lower concentration of the polymer active semiconductor. Moreover, by changing the blend ratios, these films allow a fine tuning of the active layer's rheological and optical properties without compromising charge transport.

It is well known that polymeric semiconductors often exist with disorder feature but suffer from low charge mobilities. Based on the observation that inorganic polycrystalline semiconductors exhibit better charge transport than their amorphous analogues, a lot of effort in synthesizing of semiconducting polymers and developing of deposition techniques was made aiming at enhancing crystallization. However, several reported polymer semiconductors seem to refute old paradigms exploited to attain high mobility [27–29]: low crystallinity or even amorphous polymers now perform as well as semi-crystalline materials [18,30]. Similar phenomenon was found in conjugated polymers/insulating polymers blend system [26]. The microstructure and growth mechanism of small molecule/polymer blends have been widely analyzed [31]. However, the interplay between electrical properties and many factors, such as the rate of solvent evaporation, the rate of crystallization of individual component in conjugated polymers/insulating polymers, is far from being clearly articulated, especially for those blend polymers with disordered structure but exhibit commensurate performance as highly ordered pure conjugated polymers.

In this paper, P3HT with self-organizing properties was selected as a semiconducting material in blend film. Polystyrene acted as the insulating polymer, and was blended with P3HT. Several solvents (chloroform, o-xylene, chlorobenzene and 1,2-dichlorobenzene) with different properties, such as boiling point, evaporation rates and solubility to PS and P3HT were investigated. Then, atomic force microscopy (AFM), X-ray diffraction (XRD), and UV–Visible absorption spectra were used to characterize the film morphology, crystallinity, and intermolecular connection, respectively. Finally, the improved performance OFETs consisted of these polymer blend films with less P3HT active material were realized on glass substrates.

2. Experimental section

2.1. P3HT/PS blend preparation

P3HT (MW = 45,000 g mol⁻¹, 93% regioregular) was purchased from Luminescence Technology Corp., PS (MW = 280,000 g mol⁻¹) and the solvents were purchased from Sigma–Aldrich. All the materials were used as-received without further purification. P3HT and PS were dissolved in chloroform (CF), o-xylene (XY),

chlorobenzene (CB) and 1,2-dichlorobenzene (DCB) (The summary of properties of aforementioned organic solvents was shown in support information as Table S1) with a concentration of 8 wt% and then mixed to form the P3HT/PS blend solution (v/v = 1:4).

2.2. Device preparation

Fig. 1 shows the molecular structures of P3HT active material and PS insulating material, and the schematic of the bottom-gate top-contacted OFET with P3HT/PS blend as organic active layer. The OFETs were processed according to the following procedure. Indium tin oxide (ITO) coated glass was used as substrate and gate electrodes. Prior to deposition of the dielectric layers, the substrates were successively ultrasonic cleaned in acetone, deionized water and isopropyl alcohol. PMMA, functioned as the gate dielectric, was deposited on ITO substrate via spin coating at room temperature (25 °C) and baked in an oven at 90 °C for 2 h.

Subsequently, P3HT:PS blend solutions were deposited onto dielectric layer using an on-the-fly-dispensing spin-coating approach (2000 rpm) [32], in which the solution was dispensed when the spin-coater motor was already operating at fixed rotation speed, to realize the uniform blend films. The samples were heated for an additional 20 min at 120 °C in a vacuum oven to remove the residual solvent and allow thermal annealing for P3HT:PS film. It was widely reported that additional annealing for P3HT film could facilitate the carrier transport [33,34]. Finally, the source and drain electrodes of 50 nm gold (Au) were thermally deposited on the blend film and patterned with a shadow mask. The length and width of the channel were 100 μm and 1 cm, respectively. The electrical characteristics of all the devices were measured with a Keithley 4200-SCS Source Measure Unit under ambient conditions. The field-effect mobility of devices was extracted in the saturation regime from the highest slope of $|I_{DS}|^{1/2}$ vs. V_G plots by using Eq. (1):

$$I_{DS} = \left(\frac{WC_i}{2L}\right)\mu(V_G - V_T)^2 \quad (1)$$

where L and W are the channel length and width, respectively. C_i is the capacitance (per unit area) of the dielectric, V_G is the gate voltage, and I_{DS} is the drain-source current.

2.3. Film characterization

The morphologies of P3HT/PS blend films were analyzed by atomic force microscopy (AFM) (Agilent, AFM 5500) images. The

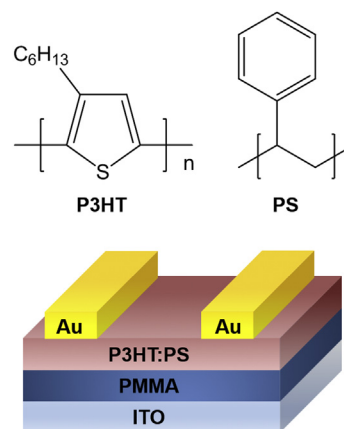


Fig. 1. Molecular structures of P3HT and PS, along with a schematic structure of OFET based on P3HT/PS blend in this study.

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