Contents lists available at ScienceDirect

Synthetic Metals

journal homepage: www.elsevier.com/locate/synmet

Crystallinity and performance improvement in solution processed organic field-effect transistors due to structural dissimilarity of the additive solvent

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ARTICLE INFO

Article history: Received 15 December 2015 Received in revised form 16 January 2016 Accepted 21 January 2016 Available online 15 February 2016

PACS: 85.30.Tv 72.80.Le

Keywords: Organic field-effect transistors Solution processing Non-solvents Crystallinity Intermolecular interactions

ABSTRACT

The effect of structural dissimilarity of the additive solvent from the main solvent on the properties of the solution and resulting crystals for solution processed organic field-effect transistors (OFETs) is comprehensively studied. Organic semiconductor TIPS-Pentacene was explored with toluene as the main solvent and benzene, cyclohexane, and hexane as additives for making semiconductor solutions. The solution formation with various solvents is explained by Hansen's solubility theory, according to which the binary solvent with higher structural dissimilarity between constituent solvents is a weaker solvent of the organic semiconductor; which eventually supports better molecular aggregation and crystallinity in the resulting crystallites. As investigated by XRD, the degree of crystalline order systematically increased with rising dissimilarity in component solvents; integrated intensity ratio rising from 0.52 for toluene to 0.54, 0.69 and 0.72 respectively for toluene/benzene, toluene/cyclohexane, and toluene/ hexane. The variation in degree of crystallinity is also reflected in electrical properties of OFETs, where the field-effect mobility improved from 0.05 cm²/(V.s) for toluene solvent to higher than 0.1 cm²/(V.s) for toluene/hexane.

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1. Introduction

The inevitably critical role of the organic semiconductors as the active material in the organic field-effect transistor (OFET) devices has caused the tremendously grown research interest in its material properties and deposition styles [1-3]. One such property is the crystalline nature of the deposited semiconductor, which is very crucial for an unhindered charge transport [4,5]. The charge transport occurs in a few monolayers of organic semiconductors close to semiconductor-dielectric interface and it is significantly influenced with the crystallinity of these monolayers [6,7]. These crystalline layers lead to superior characteristics because of the absence of grain boundaries in which charge carriers tend to be scattered or trapped during the transport process [1,5]. The overall crystallinity of the active layer depends on the procedure of depositing organic semiconductor. For vacuum grown semiconductors like Pentacene, methods to obtain crystalline films are well studied [8-10]. However, solution processing is preferred

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http://dx.doi.org/10.1016/j.synthmet.2016.01.013 0379-6779/© 2016 Elsevier B.V. All rights reserved.

for organic electronic device fabrication due to its advantage of low cost and simplicity. Spin coating, which is one of the most commonly used method for film formation [11,12], is kinetically unfavorable for growing highly crystalline films due to fast solvent evaporation [13], whereas techniques such as drop casting promote better molecular aggregation due to slow evaporation rate, and lead to large crystals with high degree of crystallinity [14]. To enhance the crystalline order in a solution deposited semiconductor, several schemes including solvent vapor annealing [15], evaporation controlled fluidic channel [16], solvent exchange [17,18], and dual solvent [19–23] have been applied. Dual solvent technique is one of the effective, simple, and very usual strategies to control the evaporation time of the solvent. Molecular structure of the additive has a profound effect on the overall evaporation rate of the solvent and the solvation of the organic semiconductor. These additives when added into the main solvent in a certain proportion have great potential to alter the intermolecular forces between semiconductor molecules, solvent molecules and semiconductor-solvent molecules by means of preferential solvation [24]. Many solution processed organic semiconductors show high degree of variation in their performances due to changes in





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solution formation and processing methods. 6,13-Bis(triisopropylsilylethynyl) pentacene (TIPS-Pentacene) is one such promising semiconductor which is reported with carrier mobility varying from 0.007 cm²/(V.s) to 2.7 cm²/(V.s) in recent years [25–27], is chosen for our study. There are many reports available in literature for TIPS-Pentacene devices fabricated with dual solvent scheme [19–21,23]; however, the critical role of the nonsolvent and its molecular structure in the solvation process of an organic semiconductor has not been addressed in detail and remain underexplored so far.

In this paper, we demonstrate the effect of structural dissimilarity of various additives to the main solvent on the degree of crystallinity of the semiconductor film and performance of the solution processed TIPS-Pentacene OFETs. We followed a systematic approach to understand the role played by various active intermolecular forces in the solvation of a semiconductor which ultimately influences the crystalline order and the device performance. We found that increasing dissimilarity of the additive in the binary solvent mixture promotes better molecular aggregation in the semiconductor due to lesser ability to overcome the intermolecular forces between semiconductor molecules; resulting in a systematic increase in crystalline nature. Crystallites resulting from a binary mixture with high structural dissimilarity have shown better crystallinity (integrated intensity ratio rising from 0.52 to 0.72) and improved electrical characteristics (field effect mobility up to 0.15 cm²/(V.s)) as confirmed by experimental results.

2. Device fabrication and characterization

Highly doped *n*-type Si wafers with a 300 nm thick SiO₂ $(C_{ox} = 11 \text{ nF/cm}^2)$ layer were used as the substrates for making devices in bottom-gate top-contact configuration. TIPS-Pentacene was purchased from Sigma Aldrich and was used without further purification. Si/SiO₂ substrates were thoroughly cleaned in heated solvents of 2-Propanol, trichloroethylene, and methanol and copious amount of deionized water. A commonly used solvent and additive volume ratio of 80/20 was selected for the study [19,28]. 1.0 wt.% solutions of TIPS-Pentacene were prepared in toluene, toluene/benzene, toluene/cyclohexane, toluene/hexane solvents and stirred for 3 h at 70 °C. After dispensing the solution on substrates inclined at a small angle ($\sim 5^{\circ}$), substrates were covered with a Petri dish to provide a solvent rich environment for slow solvent evaporation. Subsequently, the substrates were heated at 80 °C to evaporate any residual solvent. A 200 nm thick Au layer was deposited under a high vacuum of 10⁻⁶ Torr to form Source-Drain contacts using shadow masks having separation of 150 µm to 250 µm to define channel length. The precise channel length and device width were also measured after complete device fabrication. The molecular structure of the various materials used and the device structure are given in Fig. 1. All the solution preparations and sample processing steps were done in dark and



Fig. 1. Chemical structures of TIPS-Pentacene (a), and various solvents; toluene (b), benzene (c), cyclohexane (d), and hexane (e); The drying behavior of a drop cast solution of TIPS-Pentacene (f), and Device structure of a top-contact TIPS-Pentacene OFET (g).

ambient conditions. To find the degree of crystallinity of various films, 1D Grazing Incidence X Ray Diffraction (GIXRD) measurements were performed using D8 Advanced system from Bruker by keeping the crystal alignment direction along the plane of incident X rays. 1D GIXD studies were preferred for higher accuracy and resolution which allowed precise measurement of diffraction peak width [29]. Surface morphology of crystals were captured using SPM XE-70 Atomic Force Microscope (AFM) from Park Systems. Electrical measurements were done using Keithley 4200 SCS. Field-effect mobility (μ) and threshold voltage ($V_{\rm TH}$) were extracted from the transfer characteristics in the saturation regime from the highest slope of $|I_{\rm DS}|^{1/2}$ vs. $V_{\rm GS}$ plots using the saturation regime current equation for standard transistors.

$$I_{\rm DS} = \frac{1}{2} \mu C_{\rm ox} \frac{W}{L} (V_{\rm GS} - V_{\rm TH})^2, \tag{1}$$

where C_{ox} is the capacitance density of the gate dielectric layer.

3. Solution dynamics and solubility analysis

In a solution, there are three kind of intermolecular interactions; solute-solute, solvent-solvent and solute-solvent. It is the affinity of the solvent for solute molecule, which regulates the level of all intermolecular forces in the solution and governs many important properties of the resulting solution and the film formed [30]. The affinity of a particular chemical for a material can be estimated using Hansen's solubility theory [31]. Solubility parameters for various solvents used in the study and TIPS-Pentacene are given in Table 1. ∂_d , ∂_p , and ∂_h are the three Hansen's

Table 1

Hansen's solubility parameters for various solvents and TIPS-Pentacene. Dual solvents have a volume ratio of 80:20.

Solvent/Material	Hansen's Solubility Parameters (MPa) ¹ / ₂			Mismatch from TIPS-Pentacene
	$\partial_{\mathbf{d}}$	$\partial_{\mathbf{p}}$	$\partial_{\mathbf{h}}$	(R)
Toluene	18.00	1.40	2.00	2.04
Benzene	18.40	0.00	2.00	1.69
Cyclohexane	16.80	0.00	0.20	3.96
Hexane	14.90	0.00	0.00	5.32
Toluene/Benzene	18.08	1.12	2.00	1.90
Toluene/Cyclohexane	17.76	1.12	1.64	2.33
Toluene/Hexane	17.00	1.12	1.60	2.78
TIPS-Pentacene	18.80	0.40	3.60	NA

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