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Semiconductor:polymer blend ratio dependent performance and stability in low voltage flexible organic field-effect transistors



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ABSTRACT

Keywords: Organic field-effect transistors (OFETs) Flexible TIPS-pentacene TIPS-pentacene:polymer blend ratio Bending The critical role of the mixing ratio of the semiconductor and polymer solutions on the performance and electromechanical stability of the flexible blend OFETs is investigated. An improvement in the OFET performance was observed with increasing fraction of polymer in the solution. The maximum field-effect mobility improved from 0.08 for neat TIPS-pentacene OFETs to 0.24, 0.25 and 0.57 cm² V⁻¹ s⁻¹ for semiconductor:polymer blend OFETs with mixing ratio of 3:1, 1:1 and 1:3 respectively. Under stress conditions of $V_{\rm DS} = V_{\rm GS} = -5$ V for 1 h, the 1:3 blend devices outperformed other categories of devices with very small drain current decay of ~2% and 11% and small threshold voltage shift of 0.05 V and 0.27 V before and after 1.27% (R_{bend} = 5 mm) tensile strain application respectively. These devices also exhibited superior reliability with least performance spread over 100 continuous transfer characteristics measurement cycles before and after application of strain.

1. Introduction

Aftermath of extensive research interest and accompanying advancement in the device performance have made the solution processed organic field effect transistors (OFETs) comparable to high mobility organic single crystal field effect transistors [1-8]. Along with achieving superior performance in devices, however, performance reliability is also a major concern in several standard applications of flexible electronics where a high degree of performance invariability is expected from devices under strained situations [9-12]. When the high performance and high degree of electro-mechanical stability are the prime objectives for a solution processed flexible OFETs, blending organic semiconductor with an insulating polymer binder is certainly one of the best possible and conventional choice to achieve both aforementioned features together [13-17]. Our research group has recently reported the improved electrical performance and stability of TIPSpentacene:PS blend OFETs over neat TIPS-pentacene OFETs on both rigid and flexible platforms [18-20]. Semiconductor:polymer blend OFETs derive their remarkable electrical performance due to occurrence of vertical phase separation between semiconductor and polymer films [19-22]. Numerous factors influence the electrical performance of blend OFETs which include deposition strategy, material properties of the polymer binder (molecular weight, crystallinity etc.), solvent used, and mixing proportion of the semiconductor and the polymer solutions [14,16,23-29]. The mixing ratio of semiconductor and polymer solutions is one of the crucial parameters, which has been given its due

consideration while exploring its effects on the electrical performance of blend OFETs. Semiconductor:polymer mixing ratio, which has the potential to alter the device performance by several orders of magnitude by affecting overall film formation, also impacts the operational stability of devices. However, the relation between the mixing ratio and the electrical stability of semiconductor:polymer blend OFETs is scarcely explored [28,30].

In this report, the dependency between the mixing ratio of the semiconductor and polymer solutions and the electro-mechanical stability of the blend OFETs is explored. Rigorously explored semiconductor material TIPS-pentacene has been used with insulating polymer polystyrene as the semiconductor-polymer pair. Polystyrene which is one among the few insulating polymeric binders reported, capable of forming phase-separated structures with small molecule organic semiconductors, was chosen to form blend with semiconductor. As per the literature available, polystyrene does not interrupt the π - π stacking of the TIPS-pentacene molecules and improves the uniformity in the molecular morphology and the active layer coverage within the device [13,30,31]. Polystyrene also has good degree of solubility in common organic solvents. The electrical performance of OFETs was observed to improve with increasing polymer content in the solution. The maximum field-effect mobility improved from 0.08 for neat TIPSpentacene OFETs to 0.24, 0.25 and $0.57\,\text{cm}^2\,\text{V}^{-1}\,\text{s}^{-1}$ for semiconductor:polymer blend with mixing ratio of 3:1, 1:1 and 1:3 respectively. The 1:3 semiconductor:polymer blend OFETs exhibited much lesser drain current decay ($\sim 2\%$) under the gate bias stress in

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comparison to 1:1, 3:1 blend and neat OFETs with drain current decay of 11.2%, 17.8%, and 41%, which resulted in threshold voltage shifts of 0.05 V, 0.3 V, 0.48 V and 1.1 V respectively. All types of the devices were recovered almost completely from the degrading effects of gate bias stress. OFETs with 1:3 semiconductor:polymer blend ratio showed an unchanged electrical behavior after undergoing 100 continuous characteristics measurement cycles with respect to all other types of OFETs, which demonstrated a large spread in their electrical characteristics.

2. Experiments

OFETs were fabricated in top-contact bottom-gate architecture on indium tin oxide (ITO) coated flexible polyethylene terephthalate (PET) substrates (thickness = $127 \,\mu$ m, surface resistivity $60 \,\Omega/sq$.). All the materials were purchased from Sigma-Aldrich and used directly without further purification. Substrates were cleaned using ultrasonic bath in methanol, de-ionized water and 2-propanol respectively, and then dried in a N₂ blow. A 40 nm thick dielectric layer of HfO₂ was deposited on these substrates by atomic layer deposition at 100 °C using tetrakis(dimethylamido)hafnium (TDMAH) and H₂O as precursors. 0.5 wt.% solution of TIPS-pentacene and poly(styrene) ($M_W \sim 280,000$) were prepared separately in toluene by stirring at 70 °C for 3 h. Various blend solutions were then prepared by mixing TIPS-pentacene and polymer stock solutions in 3:1, 1:1 and 1:3 ratio by volume, followed by stirring for 30 min. To make the active organic semiconductor layer, neat/blend solutions were dispensed on HfO2 deposited substrates. Just after drop cast, the substrates were covered with a glass petri dish to provide a solvent rich environment to the drying film. All solution preparations and sample processing steps were done in dark and ambient conditions. An Au layer (thickness of 200 nm) was thermally deposited through shadow masks under a high vacuum of 10⁻⁶ Torr to form Source-Drain contacts. Surface morphologies of crystals were captured by atomic force microscopy (AFM) using SPM XE-70 from Park Systems. X-Ray Diffraction (XRD) measurements were done using D8 Advanced system from Bruker. Electrical measurements were performed using Keithley 4200 SCS. All measurements were performed in ambient condition with the relative humidity \sim 20%. After characterization, the devices were stored in vacuum. Device performance is definite to vary with oxygen content of the air. Moreover, TIPS-pentacene is known to be highly air stable. However, relatively constant environmental conditions were maintained near measurement setup. μ_{sat} and V_{TH} were extracted from the highest slope of the linear fit of $|I_{DS}|^{1/2}$ vs. V_{GS} plots using the saturation region drain current equation for standard transistors,

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

where C_i is the capacitance density of the gate dielectric layer. Values of C_i for neat and blend devices have been measured from metal-insulatormetal (MIM) capacitors and metal-insulator-semiconductor (MIS) structure respectively and found to have values of 200, 31(\pm 5.9), 27.4(\pm 1.94), and 21.11(\pm 1.84) nF/cm² at 1 KHz for neat, 3:1, 1:1 and 1:3 blend films.

To explore the bending stability, devices were subjected to tensile strain along the channel direction by bending the substrate with radius R_{bend} of 5.0 mm. The strain on the device was calculated using the following equation:

$$\text{Strain}(\%) = \frac{\iota_{\text{sub}}}{2R_{\text{bend}}} \times 100 \tag{2}$$

where t_{sub} is the thickness of the substrate.

3. Results and discussion

Fig. 1(a-d) shows the surface morphologies of the TIPS-pentacene

crystals obtained from four kinds of solutions. Surface morphologies for all four types of crystals were found to be identical, which suggest that terracing structure of the TIPS-pentacene crystal does not vary with the proportion of TIPS-pentacene in the solution. Fig. 1(e) shows the X-ray diffractograms for four types of TIPS-pentacene films. The degree of crystallinity of all the crystals resulting from the blend solutions was higher than that of the neat TIPS-pentacene. The full width at half of maximum (FWHM) was found to decrease with increasing polymer content; 0.18 for neat TIPS-pentacene films to 0.023 for 1:1 TIPS-pentacene:PS film, indicating an increase in the crystallinity. The crystallinity of TIPS-pentacene depends on its amount in the solution and the rate of solvent evaporation from the film. Though the semiconductor content decreases from neat TIPS-pentacene to 1:1 TIPS-pentacene:PS case, the crystalline order still improves, which can be explained with the kinetics of the film formation by solvent evaporation from the semiconductor:polymer blend solutions [32]. The process of solvent evaporation is quite complicated with interplay of various related factors like vapor pressure, temperature, surface area to volume ratio etc. [33,34]. In the initial stages, solvent evaporation is vapor pressure controlled and unaffected by the presence of the solute. However, as the solvent evaporates, solution turns more viscous and solvent evaporation is limited by the diffusion of the solvent molecules to the surface of the solution. Rate of solvent evaporation is relatively slower for the case of a blend solution than that for a neat TIPS-pentacene solution due to increased viscosity of the blend solutions resulting from the added polymeric insulator [35]. Rise in fraction of the polymer in the semiconductor:polymer blend solution leads to further increased viscosity. As the solvent evaporates from blend solution, the viscosity further increases and, free volume decreases. All these factors retard the rate of solvent evaporation. However, during the slow process of solvent evaporation, semiconductor molecules get the sufficient time to improve their arrangement and enhance crystalline order in the final film [36]. Thus, abundance of polymer in the solution results in to a slower rate of the solvent evaporation and eventually to a more crystalline final film. However for 1:3 TIPS-pentacene:PS film, the FWHM value increases slightly, which is associated more with the reduced semiconductor content rather than the reduced solvent evaporation rate dependent crystallinity.

Fig. 2 shows the electrical characteristics of OFETs corresponding to all the four cases. Table 1 summarizes various electrical parameters for all the four types of OFETs. 1:3 TIPS-pentacene:PS blend OFETs outperform other types of OFETs with an average and maximum field-effect mobility of 0.37(\pm 0.14) and 0.57 cm² V⁻¹ s⁻¹ while operating at a low voltage of -5 V. 1:1, 3:1 blend and neat OFETs show average field-effect mobility of 0.19(\pm 0.04), 0.18(\pm 0.04), and 0.05(\pm 0.01) with maximum of 0.25, 0.24 and $0.08 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ respectively. Higher performance of blend OFETs can be attributed to formation of a uniform dielectric-semiconductor interface through the process of vertical phase separation (resulting in TIPS-pentacene:PS:TIPS-pentacene tri layer structure [19]) and several associated factors [16,21,30]. Foremost among them can be the enhanced quality of the active semiconductor layer due to slowest solvent evaporation in polymer rich 1:3 blend films in comparison to all other cases as discussed before, which causes ameliorated charge transport in the corresponding films. Second reason can be the decreased dipolar disorder and carrier localization at the PS:TIPS-pentacene interface, which reduces broadening of density of states leading to a lower density of trap states than that in the neat device [37-40]. However, the reasons of increasing device performance with increasing polymer fraction are not yet completely understood. However, further deficiency of the semiconductor in the blend solution will obviously lead to performance roll-off, as also suggested by some previous reports [28,30,41]. This loss in the performance has been reasoned with the discontinuous active material film formation due to reduced semiconductor content.

Fig. 3(a) shows the decay in the normalized drain current at biasing conditions of $V_{\rm DS} = V_{\rm GS} = -5$ V for 1 h for all cases. Under constant

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