

# Interface Engineering of Functional Material Thin Films for Electronic and Optoelectronic Applications



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By

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*Chapter 7*  
*Summary and Future outlook*

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In conclusion, we have systematically investigated the mechanism behind the molecular semicrystal growth while self-assembling over the air-liquid interface. It has been explained that polymer self-assembly is mainly governed by convective Marangoni flow, compressive viscous force, divergent evaporation flux, intermolecular  $\pi$ - $\pi$  interaction, and high conformational degrees of freedom of OSPs. With these complex flow patterns during spreading and solidification, many cumulative paracrystalline disorders manifest into the microcrystalline domains. In response to the compressive viscous flow and elasticity of the restricting air-solution interface, the planar PBTTT skeleton takes more edge-on conformation than allows lattice fluctuation due to backbone rigidity. At the same time, the most flexible P3HT (and PQT also) backbone balances some of the forces by adopting many microstructural defects.

The lower alkyl chain length in P3HT provides lesser resistance, leading to the highest out-of-plane carrier mobility in the case of edge-on stacking, whereas PBTTT shows maximum in-plane (both  $\parallel$  and  $\perp$ ) mobility. For PBTTT, its backbone's rotational invariance helps adopt a low-energy planar backbone conformation, and a tail-to-tail regiopositioning of 14C-alkyl chains on the bithiophene unit promotes the self-assembly process, minimizes steric hindrance from adjacent side-chains, enhances the crystalline order, and that facilitates the in-plane carrier transport. Nevertheless, all three OSPs processed through FTM show electrical anisotropy. Thus, high-quality active material layers can be fabricated through proper backbone engineering and manipulating the forces acting at the air-liquid interface during film formation. Given this, several interface engineering approaches have been adopted to tune the molecular stacking and orientation, and improve the film crystallinity, to enhance the in-plane and out-of-plane charge transport properties.

At first, a facile thin film processing method named SVA-FTM has been developed utilizing the key advantages of conventional FTM and solvent vapor saturated atmosphere that leads to almost five times greater mobility than the conventional FTM films in parallel OFETs.

Next, another novel film fabrication technique, SM-FTM, has been developed by improvising the conventional ribbon-FTM, where the macromolecular arrangement at the solution-liquid interface can be controlled by tuning the polymer-liquid substrate interaction. SM-FTM includes key advantages (similar to conventional ribbon FTM) like large-area and homogeneous film formation, layer-by-layer coating capability, compatibility with various substrates having smooth surface etc. along with control over molecular packing. The state-of-the-art rectification ratios of  $\sim 10^7$  in SBDs and almost four times increment in the effective out-of-plane mobility compared to the conventional FTM films have been achieved through SM-FTM. Further, the dynamics of polymer solution spreading over an air-liquid interface and the effect of DBSA-polymer interaction have been demonstrated through a simple model based on lubrication theory and thin-liquid film flow kinetics.

Finally, a band engineering approach has been explored, where a synergy has been found between PBTTT and an optimum percentage of NIR dye molecule, DPPCN, that facilitates enhanced average saturation mobility up to  $0.2 \text{ cm}^2/\text{Vs}$  (1.25 times increment). Moreover, the presence of a NIR-sensitive molecule in the system causes the inception of NIR sensitivity in the order of  $10^3$  and almost one order enhancement in photosensitivity for the red light,  $\sim 10^4$ .

Thus, exploring several fundamental properties, our study demonstrates a clear picture of structure-property correlation along with cheap and facile thin film fabrication methods

aiming towards solution-processed high-performing organic electronic and optoelectronic devices.

Next, we are planning to automate the FTM process along with a large area ( $> 100 \text{ cm}^2$ ) film fabrication suitable for industry-level applications. Moreover, we will use these techniques for the fabrication of the organic hole transport layer (HTL) and electron transport layer (ETL) in semitransparent perovskite solar cells (STPSCs).