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Solution-Processed Large-Area Discrete Organic Thin-Film Transistors

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Abstract

In order to fully print high-performance organic thin-film transistors (OTFTs), a homogeneous dewetting process on large-area microdroplet arrays was developed to deposit discrete organic semiconducting thin films and the interface-modified layers of transition-metal oxides (including MoO₃, V₂O₅ and WO₃), resulting in the high field-effect mobility of 13.1 cm² V⁻¹ s⁻¹.

1. Introduction

Dewetting is considered as an attractive method to prepare thin films.[1] A homogeneous dewetting on large-area MDAs was well developed, which was caused by gravity-assisted deformation of droplets on a tilted substrate. It was found that increasing tilted angle enabled the deformation of droplets, thus leading to the homogeneous receding of upper contact lines from top to bottom on the MDAs. Moreover, this method allowed the deposition of discrete organic semiconducting thin films for fully-printed organic thin-film transistors (OTFTs). In particular, when applied a tilted angle of 90 degree, the obtained films exhibited the optimal surface morphology, which was also understood through a theoretical simulation. Furthermore, the dewetting behavior of water enabled the selective deposition of a transition-metal oxide (MoO₃) for modifying semiconductor/electrode interfaces to lower contact resistance. We found that the contact resistance significantly decreased from 14.9 kΩ cm (undoped) to 3.8 kΩ cm (MoO₃-doped) in the fully-printed OTFTs, which is consistent with the result of the field-effect mobility (μ_{FET}) increase from 9.2 cm² V⁻¹ s⁻¹ (before treatment) to 13.1 cm² V⁻¹ s⁻¹ (after treatment).

2. General Instructions

As the substrate was horizontally placed (the titled angle, $\alpha = 0^\circ$), the droplets of anisole had an axisymmetric shape and started dewetting toward their geometric centers, thus yielding radially grown crystalline films. And then, the substrate was inclined with $\alpha = 45^\circ$, wherein the upper contact line of a microdroplet was observed to recede along the oblique plane, while the bottom contact line was still pinned due to the nonaxisymmetric droplets induced by gravity. The shape deformation in droplets could render upper regions thinner but bottom regions thicker. Subsequently, well-aligned thin films grew following this unidirectional

dewetting. Furthermore, the receding motion of contact lines was speeded with the larger α of 90°, which induced much more uniform thin films to grow. It is also observed that the dewetting could be controlled as the increase of tilted angles on a large-scale MDA.

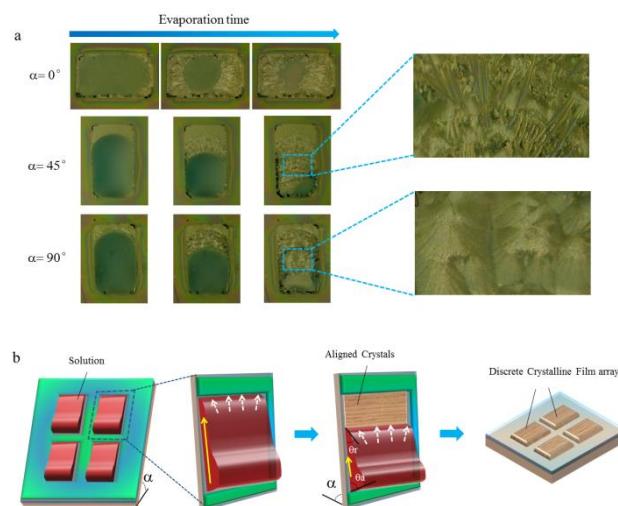


Fig. 1 A homogeneous dewetting process on large-area MDAs

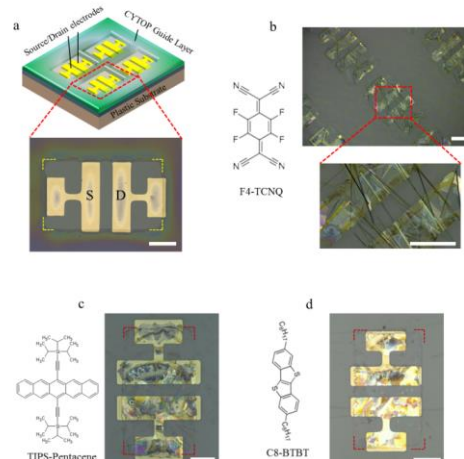


Fig. 2 Deposited thin films of organic semiconductors

The homogeneously unidirectional dewetting can be realized even in the fabrication of thin active layers for solution-processed OTFT devices. The evaluation of deposited

aligned thin film arrays was performed by measuring charge carrier transport in bottom contact/top gate OTFTs (Figure 3a). The transfer and output characteristics of a selected transistor are exhibited in Fig. 3b and 3c. As calculated, the highest μ_{FET} in the saturated regime is $9.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, and the on/off ratio was exceptionally high up to 10^9 with a sharp threshold characteristic (Fig 3b), which is several times higher than those of other flexible OTFTs. In addition, the output curves in Fig 3c presented clear nonlinear increase at low drain voltage, suggesting the dominative non-ohmic high-resistance conduction at the semiconductor/electrode interfaces[2], which is normally observed when the energy gap between the semiconductor and metal electrodes was rather large, and acts as an energy barrier for charge injection.

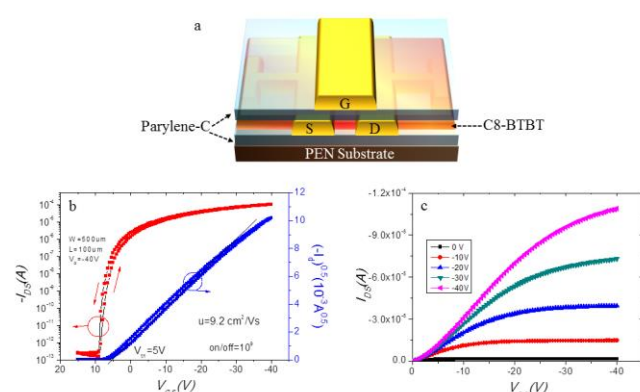


Fig. 3 Device structure and performance (undoped)

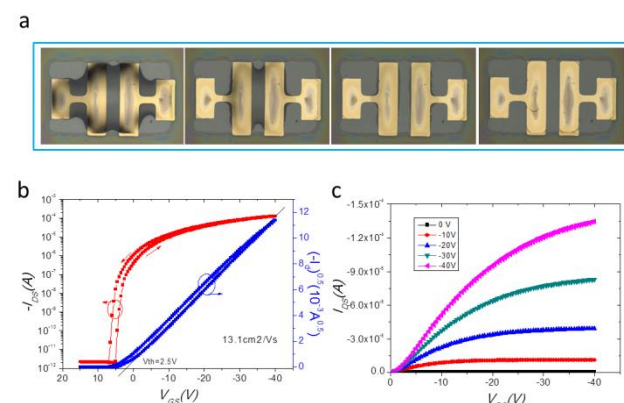


Fig. 4 Doping process and device performance (doped)

For further improving the devices' performance, we concentrated on the utilization of the induced homogenous dewetting to deposit doping layers at the interfaces of Au electrodes and organic semiconductors.[3] The hydrophilic source/drain electrodes on the hydrophobic substrate, combining with the Cytop guide layer, can naturally orient the location of aqueous dopant solution, consequently resulting in the precise deposition of dopants (Fig. 4a). Fig 4b and 4c shows the transfer and output characteristics of a transistor doped by MoO_3 . This transistor exhibited higher μ_{FET} of $13.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and smaller threshold voltage of 2.5V than

those of the undoped, which indicated that contact resistance had already decreased; this conclusion also can be verified by the following two proofs: on the one hand, the turn-on characteristics in Fig 4b became much sharper than that of the undoped and exhibited a quite small threshold slop of 0.3 V/decade; on the other hand, output curves tended to be more linear at low drain voltages in Fig 4c.

3. Conclusions

In summary, we induced a homogeneous dewetting on large-area MDAs by the gravity-assisted deformation in droplets on a tilted substrate. It was found that increasing tilted angle enabled the deformation of droplets, thus leading to the homogeneous receding of upper contact lines from top to bottom on the MDAs. Moreover, this method allowed the deposition of discrete organic semiconducting thin films for fully printed organic thin-film transistors. The obtained transistors exhibited high mobility and on/off ratio of $9.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and 10^9 , respectively. Furthermore, we developed a solution process for p-dopants that aqueous TMOs solutions can be precisely deposited onto the bottom electrodes to modify the interface state, which significantly enhanced the devices' characteristics, resulting in the contact resistance decreasing to $3.8 \text{ k}\Omega \text{ cm}$, much higher mobility and lower threshold voltage. The current procedure actually makes the fully solution-processed separate TFTs and doping of electrodes possible, which will be promising for high-resolution AM-LCDs.

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Appendix

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