# SSDM2018 Organic Electronics Fabricated by Room-Temperature Printing Techniques

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#### Abstract

In our current researches, we focus on the development of room-temperature printing techniques for position/direction/morphology-controlled depositing of organic and inorganic materials for electronic applications. By the utilization of this general strategy, various materials including metal nanoparticles, organic semiconductors, inorganic dopants, bio-macromolecules and even nanowires have already been employed for fabricating thin films and electronic devices as well. And also, this process can be implemented at ambient atmosphere so that a wide variety of substrates, like polymer, paper and biomaterial can be applied.

#### 1. Introduction

Internet of Things (IoT) device was defined for the network to connect every single object, including displays, memory devices, RFIDs and power sources. Basically, realization of IoT relays on manufacture and integration techniques for large-area flexible electronic devices. However, regular electronic devices are generally fabricated using photo- or electron-beam lithography for patterning, which are indeed highly developed methods, but have poor efficiency, high cost and difficulty for fabricating flexible devices. Therefore, to exploit an alternative method instead of lithography has been highly concerned.

In this sense, we have developed room-temperature printing methods for large-scale manufacturing flexible electronics. [1-5] We achieved a room-temperature printing of OTFT devices and 1- $\mu$ m-resolution printing technique, fabricated high-performance OTFT devices on flexible substrates with mobility exceeding 10 cm<sup>2</sup> V<sup>-1</sup>s<sup>-1</sup>. [2] In addition, OTFTbased non-volatile memory with DNA as the dielectric was also printed using our technique. [4] Now, we are moving our goal toward other devices including sensors and actuators. The main advantage of our solution-based process shows the ability to fabricate high-resolution electronic circuits on a large-scale flexible substrate. Thus, we believe our process is promising to realize the large-scale fabrication of IoT devices. In this proposal, we are going to apply these fabrication methods to practical manufacturing of large-scale IoT devices.

#### 2. General Instructions

2.1 Homogeneous dewetting on microdroplet arrays for largearea fabricating OTFT devices

Unidirectional dewetting enables the production of large-area thin films in low cost and high efficiency. Herein

we report the homogeneously unidirectional dewetting on large-area microdroplet arrays, which was induced via the gravity-induced deformation in droplets combined with alternating lyophilic/lyophobic patterns. This process allowed the scaling-up deposition of thin films including organic semiconductors and transition metal oxides as the autogenous shrinkage of droplets, which further enabled the fabrication of large-area organic thin-film transistor (OTFT) arrays. Results indicated that the field-effect mobility and on/off ratio of fully-printed OTFTs exceeded 13 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> and 10<sup>8</sup>, respectively. Therefore, this dewetting method will be promising to realize the roll-to-roll manufacture of large-area flexible electronics.

Growing Crystals



Fig. 1. Fully-printed OTFT arrays with aligned crystalline thin films showing high mobility.

2.2 Patterning large-area crystalline thin film arrays via a centrifugal-force-driving dewetting

Controlling the crystal growth direction enables highperformance and high-uniformity organic devices. In this work, a centrifugal-force-driving dewetting on the rotating plate was developed for controlling the molecular orientation and growth direction as well. It was found that the crystal growth direction can be tuned by both adjusting tilt angle and rotating speed. When the rotating plate had a tilt angle of 60 degree, the device exhibited the highest mobility up to 8 cm<sup>2</sup>  $V^{-1}s^{-1}$ , while the lowest of 4 cm<sup>2</sup>  $V^{-1}s^{-1}$  was determined in the devices fabricated on the horizonal placed plate. In addition, the device performance was found to be rather uniform comparing with those by other methods, like shearing and dipcasting.



Fig. 2. Centrifugal-force-drived dewetting on the rotating plate developed for controlling the molecular orientation and growth direction and the resultant device performance

2.3 Layer-by-Layer printing of non-volatile OTFT memory

Manufacturing non-volatile memory transistor device arrays using a printing technique is highly desired in the rollto-roll production of integrated circuits. Here, we demonstrate that the biomacromolecule of DNA can be homogeneously oriented via a solution process in butanol, which thus can be employed as the dielectric with a densely packed structure and a good insulating property. This allows to fabricate integrated organic thin-film transistor (OTFTs) memories on a large-area flexible substrate at ambient atmosphere. Combining with the result of the low frequency dependence of capacitance and the long-time retention characteristic of more than 100 seconds, this solution-processed DNA-complex was revealed to be a ferroelectric-like dielectric. Besides, the printed memories exhibit hole mobility as high as 0.65 cm<sup>2</sup>V<sup>-</sup> <sup>1</sup>s<sup>-1</sup> and a large memory window up to 13 V. Therefore, this approach is promising for printing large-scale flexible OTFT memories and thus realizing highly integrated electronics.



Fig. 3. Fully-printed OTFT memory with dielectric DNA 2.4 Vapor-Driven Spreading kinetics for Fabricating One-Dimensional Nanostructures with In-Plane Alignment

Spreading of the droplet into a thin liquid film on a certain solid surface followed by solvent drying is the crucial process for the nanostructure formation. However, such a thin liquid film was commonly observed to rupture due to the instability on a given surface. Here, we develop a technique to control the dynamical wetting of a solution droplet by the cosolvent vapor, which yielded a reversible spreading/dewetting process between the spherical droplet and the stable ultra-thin liquid layer on a hydrophobic surface. Our theoretical model indicates that this process was governed by the sorption of co-solvent vapor within the droplet which affects the surface free energy and thus lowers the contact angle (and film thickness which relates to disjoining pressure of the solid/liquid interface). Furthermore, the obtained ultra-thin liquid films allowed the in-plane alignment in one-dimensional nanostructures. In particular, in-plane aligned organic single crystals unveiled a high field-effect mobility achieved up to 9.1 cm<sup>2</sup>/V s.



Fig. 4. OTFT devices with In-plane crystalline films and device performance

### 3. Conclusions

This method will be promising for low-cost, high-efficiency manufacturing of high-performance flexible electronics toward large-area IoT devices.

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