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Influence of substrate temperature and overlap condition on the evaporation behavior of inkjet-printed semiconductor layers in organic thin film transistors

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ABSTRACT

In this study, we investigate the evaporation behavior of inkjet-printed semiconductor layers of 6,13-bis(triisopropylsilylethynyl) pentacene (TIPS pentacene) to obtain well-oriented crystalline structures for the fabrication of high-performance organic thin film transistors (OTFTs). Variations in overlap and substrate temperature are considered to control the evaporation behavior and the resulting crystalline structures of the layers. The internal flow and corresponding contact line movement can be controlled via three representative inkjet-printing regimes. For inkjet-printing regimes that induce outward convective flow (the coffee stain effect), randomly-oriented TIPS pentacene crystalline structures are produced by irregular contact line receding. With an optimized inkjet-printing regime, uniform bottom contact line movement is generated by the unidirectional internal flow, resulting in well-oriented crystalline structures. All-inkjet-printed OTFTs with semiconductor layers inkjet-printed using the optimized regime exhibit a high field effect mobility of ~0.13 cm^2/Vs, due to the one-dimensionally-oriented TIPS pentacene crystal arrays.

1. Introduction

Brittle and rigid silicon-based inorganic semiconductors are increasingly being replaced with flexible polymer and molecule-based organic semiconductors in organic thin film transistor (OTFT) [1–3]. These OTFTs are then implemented in innovative flexible electronics [4–6]. Moreover, soluble organic semiconductor materials have gained considerable interest in the field of flexible electronics, because OTFT can be fabricated via simple and inexpensive solution processes [7–12].

The crystalline structure of the semiconductor layers is one of the important factors that determine the electrical performance of OTFT. High carrier mobility can be obtained from a well-oriented crystalline structure. In contrast, randomly-oriented crystalline structure in semiconductor layers results in significant degradation in the OTFT performance. In addition, the crystalline structure and orientation of solution-processed semiconductor layers are closely related to the evaporation behavior resulting from their drying processes [13–16]. It is therefore necessary to control evaporation behavior in solution processing to produce well-oriented crystalline structures for the fabrication of high-performance OTFT.

Various strategies have been developed to print semiconductor layers with well-oriented crystalline structures. External forces were intentionally applied to the drop-casted semiconductor solution on substrates in order to control evaporation behavior using gas injection [17,18], centrifugal force [19], and the movement of a meniscus solution [20,21]. Although crystalline structure in the semiconductor layers can be well-oriented, larger material consumption, sophisticated procedures, and difficulty in precise positioning of the semiconductor layer on the channel region are inevitable.

Inkjet printing is an attractive solution process for printed electronics and has potential to overcome these problems because of many advantages such as efficient material usage by drop on demand, simple and precise patterning without a mask, and eco-friendliness [4,22–27]. Even though there have been several studies on controlling the evaporation behavior of inkjet-printed semiconductor layers, the previous works have dealt with only a tiny, single, inkjet-printed semiconductor droplet when controlling the drying process and the resulting crystalline structure [28–30], leading to practical limitations in OTFT fabrication. Single semiconductor droplets would not be appropriate for conventional parallel-type source-drain electrodes. These single semiconductor droplets can only be used for specific OTFTs such as the ring-shaped OTFT; therefore, the strategy for inkjet-printing large semiconductor layers with well-oriented crystalline structures is required for the implementation of high-performance inkjet-printed OTFTs. However, few systematic studies have been conducted on the evaporation behavior of large-area semiconductor layers inkjet-printed using multi-droplets. It should also be noted that the semiconductor layers made of multi-droplets definitely represent a different drying behavior compared to that of a single semiconductor droplet on a substrate.

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In this study, overlap condition and substrate temperature were considered critical factors to control the evaporation behavior of inkjet-printed semiconductor layers using multi-droplets. Internal flow and the corresponding contact line movement of drying semiconductor layers can be controlled via these two printing parameters. For the inkjet-printing regimes which induce outward convective flow, randomly-oriented 6,13-bis(triisopropylsilylethynyl) pentacene (TIPS pentacene) crystalline structures are produced in the inkjet-printed semiconductor layers by irregular receding of the contact line. With optimized overlap conditions and substrate temperatures, uniform bottom contact line movement is generated due to the unidirectional internal flow, resulting in well-oriented crystalline structures. Finally, all-inkjet-printed OTFTs with well-oriented crystalline structures of semiconductor layers are fabricated, and their electrical performances are assessed.

2. Experimental details

2.1. Ink preparation

In order to synthesize a semiconductor solution, TIPS pentacene (Tokyo Chemical Industry) was dissolved in anisole solvent (Sigma-Aldrich) with a concentration of 15.3 mg/ml. For inkjet-printing dielectric layers, a cross-linked poly(4-vinylphenol) (PVP) solution was synthesized via magnetic-stirring of 0.6 g PVP (Sigma-Aldrich) and 0.2 ml poly(melamine-co-formaldehyde) (Sigma-Aldrich) with a mixture solvent of 3.4 ml ethanol (Daejung Chemical & Metals) and 1.6 ml 1-hexanol (Sigma-Aldrich) for 60 min. A commercially-available silver nanoparticle ink containing 52.1 wt.% silver nanoparticles (NPS-JL, Harima Chemical Co.) was used to print gates and source/drain electrodes in this study. A hydrophobic fluorocarbon (FC) solution (DS-1120, Harves) was purchased and inkjet-printed as received to define channel regions in the semiconductor layers.

2.2. Inkjet printing of semiconductor layers

In this study, all functional inks were ejected through single piezoelectric printheads (MicroFab Co.) with different nozzle diameters. A printhead with a nozzle diameter of 30 μm was utilized to print electrodes and semiconductor layers. Inkjet-printed dielectric layers were produced using a nozzle diameter of 100 μm. All inkjet-printing processes were performed under ambient conditions.

In order to investigate the evaporation behavior of inkjet-printed semiconductor layers, inkjet-printed PVP layers were initially fabricated on a glass microscope slide and then semiconductor layers (1.8 mm × 1.8 mm) were inkjet-printed on the PVP layers using various overlap conditions ranging from 40 to 70%. The overlap condition used in this study is described in Fig. 1. The distance between individual inkjet-printed droplets on the substrate becomes closer in the inkjet-printing process for higher overlap conditions. A raster scan method was utilized to fabricate the semiconductor layers. The same overlap condition was applied to the inkjet-printed lines in both x and y-directions of semiconductor layers for the raster scan printing process. Prior to inkjet-printing semiconductor layers, sequential C_4F_8 and O_2 plasma surface treatments of the PVP layers were carried out to avoid dewetting of the inkjet-printed TIPS pentacene solution on the PVP layers. A capacitively coupled parallel plate plasma treatment system with a radio frequency (RF) power supply of 13.56 MHz was used for plasma surface treatment. C_4F_8 gas was inserted into the plasma chamber at a flow rate of 40 sccm. After the C_4F_8 gas pressure of 380 mTorr was stabilized in the chamber, RF power of 100 W was supplied to generate C_4F_8 plasma discharge for 3 min and hence to produce a uniform FC film on the PVP layers, Then, the C_4F_8 plasma-treated PVP layers were exposed to O_2 plasma for 30 s using 30 sccm flow rate and 150 W RF power, making the PVP layers hydrophilic because the pre-deposited hydrophobic FC film on the PVP layers is etched away by this subsequent O_2 plasma treatment. In addition, the substrate temperature was varied from 30 to 40 °C during inkjet-printing of the semiconductor layers.

2.3. Fabrication of all-inkjet-printed OTFT

An all-inkjet-printed OTFT with a bottom-gate and top-contact configuration was fabricated following the procedure shown in Fig. 1. A glass microscope slide (Corning 2948, 75 mm × 25 mm) with 1 mm thickness was used as a substrate. The microscope slide was made from soda-lime glass. Before the inkjet-printing process was performed, the substrate was cleaned via ultrasonification with deionized water for 5 min, followed by a UV/O_3 treatment for 8 min. A bottom-gate electrode was produced by inkjet-printing silver ink onto the substrate. After sintering the gate electrode at 150 °C for 60 min, the PVP solution was inkjet-printed onto the bottom-gate electrode to form a dielectric layer (4 mm × 4 mm). The inkjet-printed dielectric layer was then cross-linked at 150 °C for 60 min to prevent subsequent deformation in the sintering process for source and drain electrodes. The thickness and specific capacitance of the cross-linked dielectric layers were 1.6 μm and 2.7 nF/cm^2, respectively. Plasma surface treatments with C_4F_8 and O_2 were performed to increase the surface wettability of the dielectric layer before printing semiconductor layers. A semiconductor layer was inkjet-printed on the plasma-treated dielectric layer using TIPS pentacene solution. Before inkjet-printing source and drain electrodes, a hydrophobic FC layer was inkjet-printed on the semiconductor layer to define the channel region so that inkjet-printed silver ink could be self-aligned, leading to well-defined source and drain electrodes with a channel length of 150 μm and a width of 800 μm. Finally, the inkjet-printed source and drain electrodes were thermally sintered at 120 °C for 60 min.

2.4. Characterization

We recorded the evaporation behavior of the drying semiconductor layers using a CCD camera equipped in the printing system after inkjet-

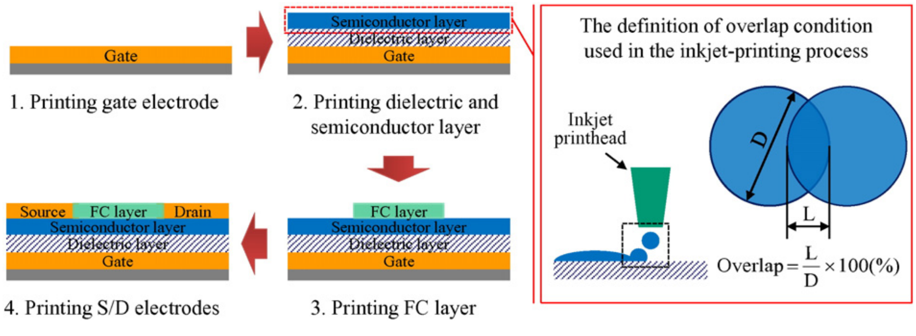

Fig. 1. Fabrication process of an all-inkjet-printed OTFT with a bottom gate and top contact configuration.