Organic thin-film transistors and circuits manufactured by sub-femtoliter inkjets

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Abstract

We have successfully manufactured high-quality top-contact organic thin-film transistors using inkjet technologies with sub-femtoliter droplet volume. Silver fine lines were directly patterned by inkjet on pentacene channel layers. The minimum width of silver lines was 1µm with without the need for pre-patterning or surface pretreatments. The mobility was 0.3 cm^2/Vs.

1. Introduction

Organic thin-film transistors (TFTs) and their integrated circuits (ICs) are expected to play an important role in realizing flexible, large-area electronics including e-paper (1,2), flexible RFID tags (3,4) and large-area sensors (5-7). As cost-effective methods to manufacture organic TFTs, printing processes such as inkjet printing (7-9), micro-contact printing (10,11), and screen printing (7,12,13), have attracted much attention. In printed organic TFTs, source/drain electrodes are printed before the deposition of organic semiconductor layer to form TFTs with bottom contact geometry (7,14,15). When source/drain electrodes are deposited on semiconductors by vacuum processes to form TFTs in the top contact geometry, the device performances could be improved significantly. In order to manufacture top-contact organic transistors by printing, the damage induced to the organic semiconductors by the solvent in inks during the pattering of the over layers has to be suppressed.

In this work, we have fabricated high-performance organic TFTs with the top contact geometry by inkjet technologies with picoliter and sub-femtoliter droplet volumes. Silver fine lines were directly patterned by two inkjet systems on the pentacene channel layer without the need for any photolithographic pre patterning or any surface pretreatment. With controlling waveforms that are applied to piezoelectric actuators in the picoliter inkjet nozzles, the volume of ink droplets containing Ag nanoparticles ejected from the nozzles can be changed from 17 to 1.4 pl. TFT characteristics are improved significantly when the ink volume is reduced less than 3 pl. The devices manufactured using the droplets of 1.4 pl exhibit the mobility of 0.3 cm^2/Vs and the on-off current ratio exceeding 10^6, which are comparable characteristics to organic TFTs whose source/drain electrodes are formed by vacuum evaporation. Furthermore, sub-femtoliter inkjets were employed to further reduce the device dimensions; the minimum width of silver lines was 2µm with the sub-femtoliter inkjet system. We have manufactured p-type channel and n-type channel organic transistors with source/drain contacts prepared by the sub-femtoliter inkjet to fabricate complimentary inverter circuits.

2. Picoliter inkjet for organic TFTs

First, we investigate organic transistors that are manufactured by picoliter inkjet. We use a picoliter inkjet printing machine that equipped heads driven by piezoelectricity. As inks for electrodes, silver nanoparticles suspended by tetradecane are used (7,13,16). In order to reduce the volume of ink droplets ejected from the inkjet nozzle, the voltage applied to piezoelectric elements of the inkjet heads is systematically changed from 12 V to 25 V. In particular, two different waveforms of the voltage shown in Fig. 1(a) are applied to the heads. Dots of silver inks are printed on polyimide film substrate by inkjet and calcinated at 220 °C for 1 hour in air. The volume of ink droplet is shown in Fig. 1(b) as a function of the voltage.

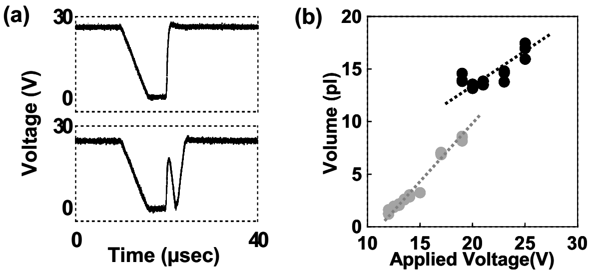

Fig. 1: (a) The two waveforms used in this experiment. (b)Volume of silver ink droplets as a function of the voltage applied to the piezoelectric actuators of the inkjet printer. The black and gray plots are the volumes obtained on using a single pulse waveform and two serial pulse waveforms, respectively.

The two-serial-pulse waveform can eject droplets whose volume is much smaller than the single-pulse waveform, as shown in Fig. 1 (a). The minimum volume reaches as small as 1.4 pl. When the voltage in the two-serial-pulse waveform is increased, two droplets of inks are ejected simultaneously. Thus, the single-pulse wave is applied when the larger volume exceeding 10 pl is needed.

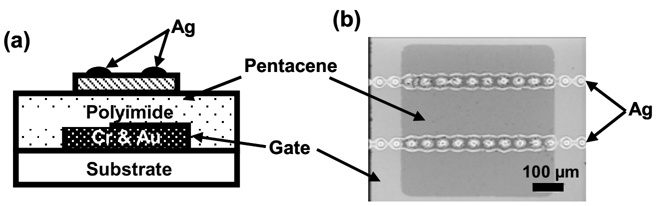

Fig. 2: (a) Cross-sectional view of the manufactured organic transistor with top contact geometry. The gate and organic semiconductor layers are patterned by vacuum evaporation, and the polyimide dielectric layer is spin-coated. Silver nanoparticles are printed on the pentacene layer by inkjet printing. (b) Micrograph of the manufactured device.

Organic TFTs are manufactured using printed source/drain electrodes with varying the volume of silver ink from 1.4 to 17 pl. Figure 2 (a) shows the cross-sectional view of the organic TFT with the top contact geometry. A gate electrode comprising 5 nm thick Cr and 50 nm thick Au layers are evaporated through a shadow mask on a 75-μm thick polyimide film substrate. A polyimide precursor is spin-coated and cured at 180 °C for 1 h in nitrogen to form a 450 nm thick gate dielectric layer. On the dielectrics, a 50 nm thick pentacene channel layer is deposited by thermal evaporation with using a shadow mask. Finally, silver nanoparticles are printed by inkjet at the substrate temperature of 90 °C to form source/drain electrodes. In this writing, the voltage applied on the piezoelectric elements is changed from 12 V to 25 V. The silver ink droplets are deposited with 50 μm spacing. The patterned silver lines are calcinated at 130 °C for 3 hrs in nitrogen.

The micrograph of the fabricated device is shown in Fig. 2 (b). The silver electrodes become wider on pentacene film than that on the polyimide dielectrics, because the surface energy on pentacene film is higher. The TFTs using 1.4 pl and 17 pl ink droplets, which are simply referred 1.4 pl TFT and 17 pl TFT, respectively, in this paper, exhibit 60 μm and 130 μm in the line width of the electrodes, respectively.

Figures 3 (a) and (b) show the source-drain current (I_{DS}) of the organic TFTs as a function of the source-drain voltage (V_{DS}) for the 1.4 pl and 17 pl TFTs, respectively. The gate voltage (V_{GS}) is changed from 20 V to –40 V in steps of 10 V. The 1.4 pl TFT shows excellent characteristics in the linear regime. This result clearly indicates that the contact resistance can be suppressed by reducing the volume of the ink droplet. The mobility of the 1.4 pl and 17 pl TFTs are 0.3 and 0.09 cm^2/Vs, respectively, in the saturation regime. In contrast, on-off ration does not depend on the volume and both exhibit the ratio of 10^6. These characteristics are comparable to that of organic TFTs using Au source/drain manufactured by thermal evaporation.

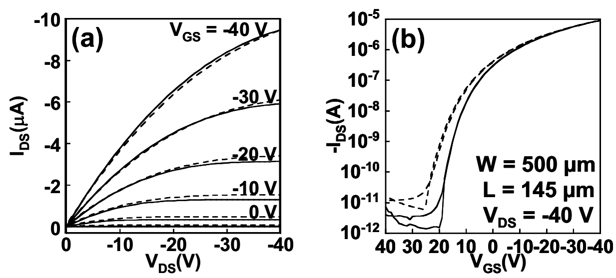

Fig. 3: Transistor characteristics of the 1.4 pl TFT. (a) Source-drain current (IDS) is show as a function of the source-drain voltage (VDS). (b) The transfer curves of the devices are shown (VDS = –40 V). The dashed lines in each graph represent the transistor characteristics after 3 weeks.

Figure 4 shows mobility as a function of the volume of silver ink droplets used for manufacturing these devices. The mobility increases significantly when the volume decreases less than 3 pl and reaches to 0.3 cm^2/Vs at 1.4 pl. This result shows that the ink volume significantly affects on the transistor characteristics.

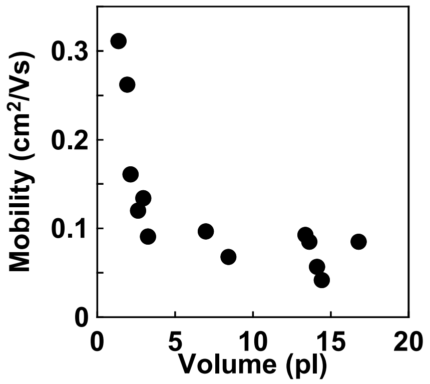

Fig. 4: Mobility in the saturation regime as a function of the volume of the silver ink droplets used for manufacturing these devices.

The TFTs are kept in nitrogen for 3 weeks. The device characteristics are shown as dashed line in Fig. 3. Although the 17 pl TFT exhibits the decrease in on current and the increase in off current, while the 1.4 pl TFT does not exhibit significant changes. The current decreases significant when the volume decreases. These results clearly demonstrate that reduction in the ink volume in 1 pl regime is crucial to obtain high mobility of 0.3 cm^2/Vs as well as good stability over many weeks on printed organic TFTs.

3. Sub-femtoliter inkjet for organic TFTs

Then, the sub-femtoliter inkjet system was used (17). This sytem can prepare top contact transistors with a channel length of 1 μm and the width of the silver lines of 2 μm. The amount of organic solvent dispensed during sub-femtoliter inkjet printing is extremely small (typically ~0.7 fl). The silver nanoparticle calcination temperature after sub-femtoliter inkjet printing was 130 °C. We have found that the morphology of the organic semiconductors was not disturbed. The transistors which combine short channel length with small contact resistance and small parasitic capacitance are demonstrated.

The transistors employ vacuum evaporated aluminum gate electrodes patterned by shadow masking and a gate dielectric based on a combination of a 3.6-nm-thick layer of aluminum oxide and a 2.1-nm-thick molecular self assembled monolayer (SAM) of n-octadecylphosphonic acid (18). The aluminum oxide film results from a brief oxygen plasma treatment required to form a sufficient density of hydroxyl groups for molecular adsorption, and the SAM is prepared from a 2 propanol solution at room temperature. The gate dielectric capacitance is 0.7 $\mu F/cm^2$, so the transistors operate with voltages between 2 and 3 V. 30 nm thick films of pentacene and hexadecafluorocopperphthalocyanine ($F_{16}CuPc$) are deposited in vacuum and patterned by shadow masking to provide the semiconductor films for the p-channel and n-channel transistors, respectively.

The field-effect mobility of the transistors is dependent on the deposition conditions. Pentacene transistors exhibit the mobility of between 0.03 cm^2/Vs (substrates not heated during pentacene deposition) and 0.3 cm^2/Vs (pentacene deposited onto substrates held at a temperature of 60 °C during the vacuum deposition). The mobility of 0.3 cm^2/Vs is by far the highest mobility reported for organic transistors with patterned gates and a channel length below 2 μm. A mobility of n-type channel $F_{16}CuPc$ transistors is 0.02 cm^2/Vs and an on/off current ratio of greater than 10^4 when substrate temperature is 90 °C. These parameters are essentially identical to those of organic transistors with evaporated metal contacts, indicating the excellent performance of the printed contacts.

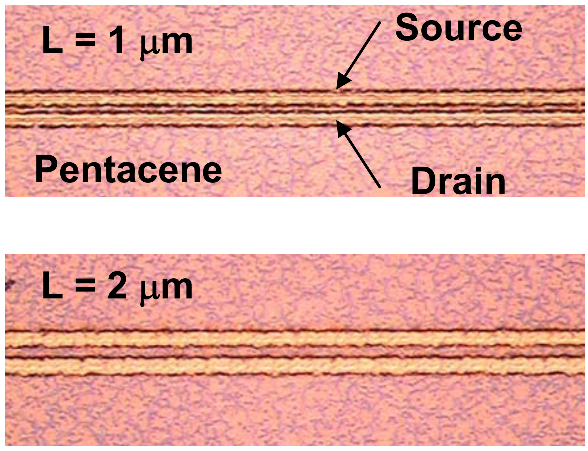

Fig. 5: Optical microscope images of pentacene TFTs with channel length of 1 μm and 2 μm after calcination.

Taking full advantage of p-type pentacene and n-type $F_{16}CuPc$ transistors, SAM based gate dielectric, and inkjet printed source/drain contacts, we have prepared organic complementary inverters. The pentacene transistor has a channel length of 50 μm, the $F_{16}CuPc$ transistor has a channel length of 5 μm, and both devices have a channel width of 60 μm. The inverter operates with supply voltages between 1.5 and 3 V and with a small signal gain greater than 10. From a circuit design perspective, complementary circuits have several advantages over circuits based on a single carrier type, including greater noise margin, lower power consumption, and faster switching speed.

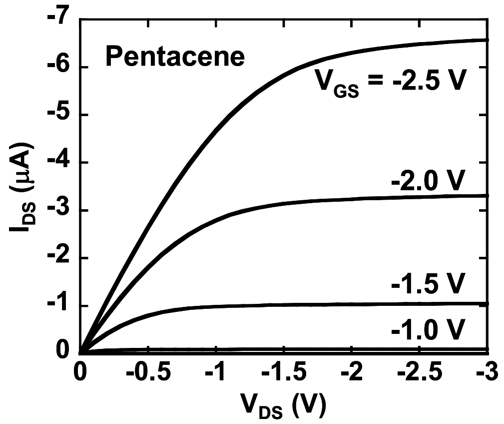

Fig. 6: Output characteristics of a p channel pentacene TFT with a channel length of 1 μm and a channel width of 300 μm. The measurements were carried out in air.

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