

Electrical Properties of Triisopropylsilyl Pentacene Organic Thin-Film Transistors by Ink-Jet Method

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Abstract: We fabricated triisopropylsilyl (TIPS) pentacene organic thin-film transistors (OTFTs) by ink-jet printing method and investigated their electrical properties. The film morphology of TIPS pentacene layer and the field-effect mobility of OTFTs were greatly affected by choice of the solvent which TIPS pentacene was dissolved in. Especially the boiling point of a solvent was a critical factor that determines the electrical properties of TIPS pentacene OTFTs. The field-effect mobility of an OTFT fabricated from anisole solution (boiling points of 155°C) was $0.04 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ while that of OTFT from chlorobenzene solution (boiling points of 132°C) was $0.01 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, suggesting that a solvent with a higher boiling point is preferable in the fabrication of ink-jet printed OTFTs.

Keywords: organic thin-film transistor; ink jet printing; chlorobenzene; anisole; field-effect mobility.

Introduction

Solution processed organic thin-film transistors (OTFTs) have attracted a considerable attention due to their potential advantages in organic electronics such as electronic paper and radio-frequency identification tags (RF-IDs) [1-3]. The solution-based process enables a simple and non-vacuum fabrication of organic devices such that extremely low-cost electronics can be achieved. Recently ink-jet printing method is widely adopted in organic electronics fabrication including organic light-emitting diode displays and organic transistors [4, 5]. The ink-jet printing method offers a fast and accurate patterning of organic layers without any costly photolithography process. The loss of rather expensive organic solutions can be significantly reduced by employing the ink-jet printing method.

There were several attempts to improve the electrical properties of ink-jet printed OTFTs, such as controlling the ink-jet process parameters [6] and surface treatment of gate insulator by employing a self-assembled monolayer (SAM). It has been reported that the organic solvent used in the semiconductor coating process had a significant influence on the film morphology, crystal structure of organic semiconductors and also their electrical properties [7]. Especially, the boiling point of a solvent had the most prominent effect on the electronic properties of OTFTs.

The purpose of the paper is to investigate the dependence of electrical properties of ink-jet printed triisopropylsilyl (TIPS) pentacene OTFTs on the boiling point of a solvent used in the ink-jet printing. For variation of boiling points, two types of organic solvents were employed in the device fabrication. They are chlorobenzene and anisole which have boiling points of 132°C and 155°C, respectively. We have also applied chloroform in the ink-jet process, however due to its extremely low boiling point (61°C), nozzle clogging was frequently occurred and could not apply in the ink-jet process. For this reason, OTFTs were also fabricated by using spin coating process and compared the results with the ink-jet printed devices.

The OTFT fabricated from chloroform solution showed field-effect mobility of $10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, whereas from chlorobenzene and anisole solutions the mobility was increased to $0.01 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $0.04 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively.

Experimental

OTFTs were fabricated with the widely used bottom-gate and bottom-contact geometry. 100-nm-thick silicon oxide barrier layer was deposited on a glass substrate by e-beam evaporation. Then, 100-nm-thick Al-Si gate electrode was deposited by dc sputtering and patterned. On the top of the gate electrode, an organic gate insulator, poly-4-vinylphenol (PVP) was spin-coated and thermally cured ($T_{\text{cure}} = 175 \sim 200^\circ\text{C}$). The PVP solution was mixed with poly (melamine-co-formaldehyde) methylated in propylene glycol monomethyl ether acetate. For source/drain electrodes, e-beam-deposited Cr (5 nm-thick) with a thermally evaporated Au (50 nm-thick) layer was used. Before ink-jetting TIPS pentacene on OTFT device, the PVP gate insulator surface was treated with hexamethyldisilazane (HMDS) by spin coating HMDS solution for 2 minutes with a rate of 3000 rpm/min. Finally, TIPS pentacene was ink-jetted using an ink-jet printing machine with 30 μm piezo-type nozzles. The average ink drop diameter was 31 μm , similar to the diameter of ink-jet nozzle. The average volume of a drop was 16.9 pl and the average drop velocity was 3.0 m/s. The sizes of the ink-jet printed drops were different with the solution used. The drop size from chlorobenzene solution was approximately 10 percent larger than that from anisole solution. Also, the drop size on a HMDS treated PVP surface was smaller than that on a bare PVP

surface, due to the difference in the surface tension of the PVP gate dielectric surface.

Figure 1(a) shows the schematic cross-section view of the fabricated OTFT device. The thickness of printed TIPS pentacene film was 20 ~ 50 nm depending on the concentration of solution (chlorobenzene and anisole) used for ink-jetting. The current-voltage characteristics of the transistors were measured in dark and air-ambient environment. Also, the morphologies of TIPS pentacene films were studied by atomic force microscopy (AFM).

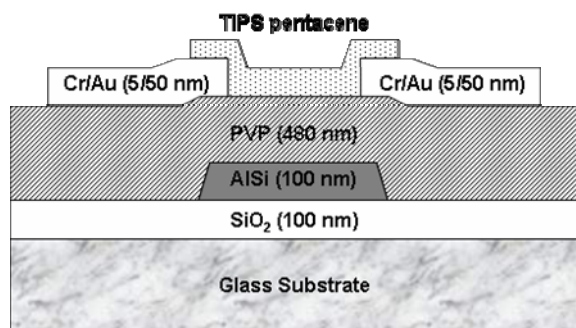


Figure 1. A schematic cross-section view of the fabricated OTFT device.

Results and Discussion

The $I_{DS}-V_{GS}$ and $\sqrt{I_{DS}-V_{GS}}$ curves of an OTFT with ink-jet printed TIPS pentacene are shown in Fig. 2(a). As described above, HMDS surface treatment was carried out before ink-jetting TIPS pentacene solution on the OTFT device. The field-effect mobility of the ink-jet printed OTFT was $0.015 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and the on/off ratio was $10^5 \sim 10^6$. Also, the electrical characteristics of a spin coated device are shown in Fig. 2(b). The spin coated device exhibited a field-effect mobility of $0.021 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and the on/off ratio was 10^4 . The rather low current on/off ratio is attributed to the fact that the active layer pattern is not defined in spin coated device and leakage current flows through the active layer. On the other hand, in ink-jet printed device the channel pattern is defined resulting in lower leakage current.

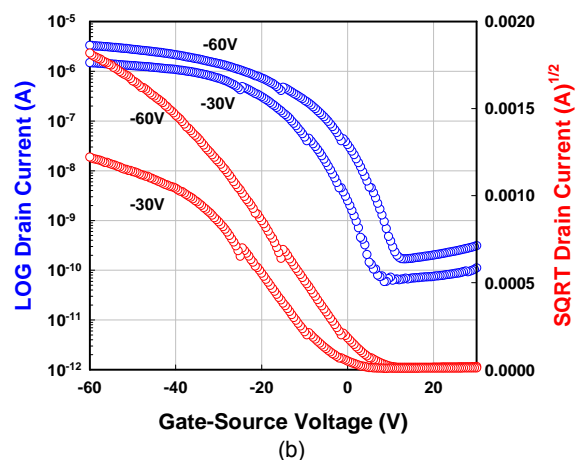
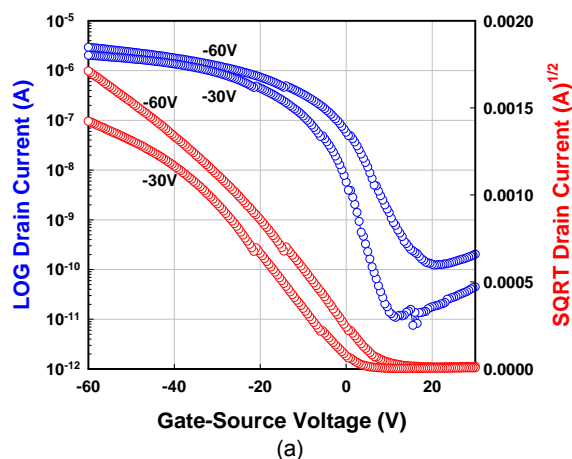


Figure 2. $I_{DS}-V_{GS}$ and $\sqrt{I_{DS}-V_{GS}}$ characteristics of (a) ink-jet printed OTFT and (b) spin coated OTFT. In this case, chlorobenzene was used as a solvent.

In Fig. 3, the AFM images of spin coated and ink-jet printed TIPS pentacene films are shown. The surface morphology and crystal structure of TIPS pentacene film significantly depend on the solvent used for coating process and also the coating method. Particularly, the boiling point of the solvent has a great influence on the film morphology. Figure 4 shows the optical microscope images which were obtained from spin coated TIPS pentacene films. From a solution with a higher boiling point (chlorobenzene), TIPS pentacene film had a dendrite-like morphology (Fig. 4(a)), whereas with a lower boiling point solution (chloroform), amorphous structure was observed. It was also confirmed from X-ray diffraction (XRD) results that TIPS pentacene films spin coated from chlorobenzene solution exhibited a preferential orientation of (001)-axis normal to the surface, while the film from chloroform had amorphous phase.

Figure 3(c) and 3(d) show the surface images of TIPS pentacene drops which were ink-jet printed on PVP gate insulator. The size of printed ink drop was varied from $60 \sim 80 \mu\text{m}$, which depends on the surface treatment of PVP layer. The AFM images were acquired from the center region of the drop with a size of $15 \mu\text{m} \times 15 \mu\text{m}$. The surface morphology of the ink-jet printed TIPS pentacene drops was quite apart from the spin coated films and in both ink-jet printed samples, no distinguishable structure was observed, such as grains or dendrite structure. Especially, TIPS pentacene film ink-jet printed from chlorobenzene had relatively low surface roughness (root-mean-square roughness of 0.97 nm) compared to other films.

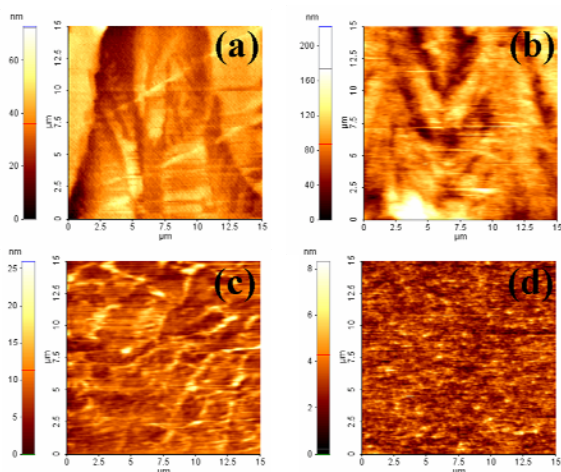


Figure 3. AFM images of TIPS pentacene films; (a) spin coated from anisole solution, (b) spin coated from chlorobenzene solution, (c) ink-jet printed from anisole solution, and (d) ink-jet printed from chlorobenzene solution. The AFM images were obtained from contact mode measurement with a size of $15\ \mu\text{m} \times 15\ \mu\text{m}$.

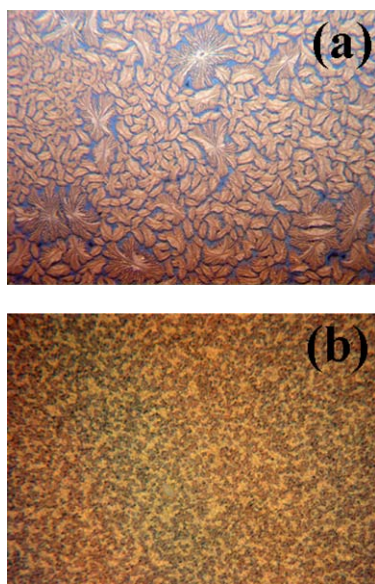


Figure 4. Optical microscope images (500X) of TIPS pentacene films; (a) spin coated from chlorobenzene solution, and (b) spin coated from chloroform solution.

It has been reported that an organic solvent with a higher boiling point is preferable as a solvent for making organic semiconductor solutions because of its slow evaporation rate [7]. With a slow evaporation rate, the solution has a longer time for drying and therefore the crystallinity of the organic semiconductor layer may be enhanced [7]. In the case of the ink-jet printing method, a solvent with a higher boiling point is preferable because the nozzle clogging occurs when a low boiling point

solvent such as chloroform (boiling points of 61°C) is employed.

In order to find out the effect of boiling point on the electrical properties of ink-jet printed OTFTs, OTFTs were fabricated with solutions having different boiling points. We used chlorobenzene and anisole (boiling points of 132°C and 155°C , respectively) as solvents ink-jetted on a same sample. In Fig. 5, the transfer curves of ink-jet printed OTFTs are displayed. The field-effect mobility of OTFT from anisole solution was $0.04\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ while that of OTFT from chlorobenzene solution was $0.01\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$. This effect of solvent on the field-effect mobility of OTFTs is clearer in spin coated devices. In spin coated devices, the field-effect mobility enhances by $10 \sim 20$ folds just by changing the solvent from chlorobenzene to anisole.

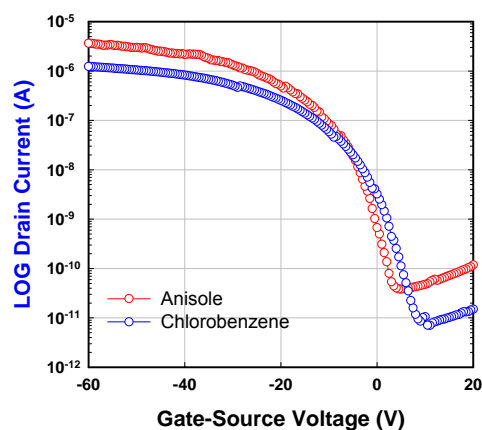


Figure 5. Comparison of transfer curves of ink-jet printed TIPS pentacene OTFTs with different solvents.

Conclusions

We fabricated TIPS pentacene OTFTs by using ink-jet printing method and investigated their electrical properties by varying solvents. The OTFT fabricated from anisole solution (which has the highest boiling point) showed field-effect mobility up to $0.04\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ while that from chlorobenzene solution showed mobility of $0.01\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$. There was a close relationship between the boiling point of a solvent and film morphology of TIPS pentacene layer. The TIPS pentacene obtained from higher boiling solvent had a dendrite-like structure, whereas when coated by a solvent with a lower boiling point, an amorphous structure was achieved. It can be concluded that in solution processed OTFTs, a solvent with higher boiling point is preferable in achieving highest electrical properties.

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