

Fabrication of Organic Field Effect Transistors Based on Printing Techniques and the Improvement of FET Properties by the Insertion of Solution Processable Buffer layers

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Abstract

In this study, we have fabricated the bottom contact (BC) type all-printed organic field effect transistors with multi-walled carbon nanotubes (MWCNTs) or Ag nano-particle (Ag NP) electrodes and their multi-layers (or composite) as printed source-drain (S-D) electrodes. The OFETs with MWCNT S-D electrodes exhibited higher hole mobility and on/off ratios than the devices with Ag NP S-D electrodes owing to lower contact resistance at the MWCNT/semiconductor interface. The high sheet resistance of MWCNT electrodes was considerably reduced by mixing with Ag NPs or multilayer deposition of Ag and MWCNTs and MWCNTs acts as a solution processable buffer layer. On the other hand, a thin film of molybdenum oxide (MoO₃) considerably enhanced hole injection in the Ag/MoO₃ S-D electrodes at the p-type OFETs, but also reduced the ON/OFF ratio resulting to the normally on operation.

1. Introduction

Organic field-effect transistors (OFETs) based on solution-processed conjugated small molecules and polymers have attracted increased interest as key components of printed and flexible electronics because of their suitability for the fabrication of economically friendly, flexible electronic circuits over large areas at low temperatures. The patterning and multilayer deposition techniques of component materials such as electrodes, dielectric layer, and organic semiconductors are becoming more important for practical applications [1-2]. The softlithography techniques are additive-type of high-resolution printing techniques, and it is useful for the fabrication of multilayers of soluble materials [3-7]. Especially, nano-transfer printing (nTP) involves the pattern transfer of a dried film from a silicone elastomer such as polydimethylsiloxane (PDMS) at low pressure and temperature. Therefore, the nTP techniques seems to be a strong candidate for fabricating the multilayers of patterned soluble materials which are easily damaged by organic solvents and traditional photolithography techniques [4-7]. Moreover, the adhesion strength between PDMS and

pi-conjugated materials, such as organic semiconductors and carbon nanotubes, is relatively lower than that in the metal-PDMS interface. Therefore, we considered it is advantageous to fabricate the OFETs with solution processed electrodes with CNTs and soluble polymer semiconductors using the nTP techniques, without using a troublesome damaging processes.

In this study, we have fabricated the bottom gate-bottom type polymer based FETs based on the softlithography including the patterning of source-drain (S-D) electrode materials and polymer semiconductor layers. We have deposited regioregular poly(3-hexylthiophene) (P3HT) as the polymer semiconductor, and poly(methyl methacrylate) (PMMA) or cross-linked poly(vinylphenol) (CLPVP) as the polymer gate insulators. We have compared the FET properties of OFETs with several types of S-D electrodes with the silver (both evaporated and solution processed Ag), multiwalled carbon nanotubes (MWCNTs), composite of Ag/MWCNTs and Ag/MWCNTs multilayers.

2. Experimental

Firstly, Indium-tin-oxide (ITO)-coated glass substrates were prepatterned by conventional photolithography and acid treatment, followed by the removal of the resist film. Secondly, polymer gate insulators (PMMA or CLPVP) were formed by spin-coating and a heat-treatment in a nitrogen atmosphere. The S-D electrodes were deposited onto a patterned PDMS (Shin-Etsu Chemical SIM-240 or 260) stamp with a typical depth in the range of 20-30 μm. Here, PMMA thin films were inserted between PDMS and Ag (or Au) electrodes to reduce the adhesion between PDMS and metal electrode, whereas MWCNTs (and CuPc/Au stack) were directly deposited on PDMS. Here, CuPc was act as a hole injection buffer layer and the release layer of the transfer printing of Au electrodes. P3HT (or PCBM as a N-type layer) was spincoated on the other flat PDMS from chloroform (or dichlorobenzene) solution. The semiconductor layers were patterned by selective lift-off process and then transferred to

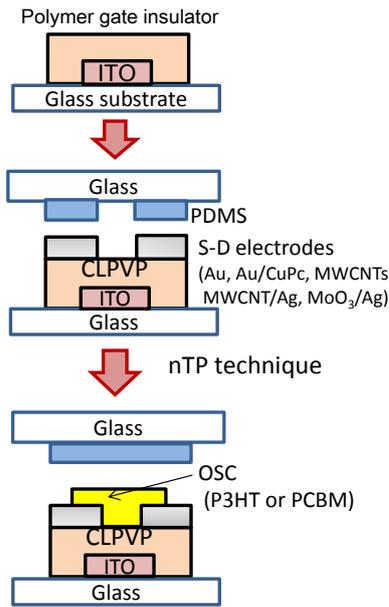


Fig. 1 Illustration of fabrication process of all-printed OFETs by nTP techniques.

the substrates in nitrogen atmosphere. The maximum process temperature for the pattern transfer was 100 °C and the typical pressure was in the range of 1-2 MPa.

FET properties were measured in a vacuum chamber after the heat-treatment in order to remove the adsorbed water and oxygen molecules during the fabrication in an atmosphere (air).

3. Results and Discussion

Figure 2 and Table I compares the FET properties of bottom contact type P3HT FETs with various S-D electrodes on CL-PVP gate insulators at $V_G = -40V$. It is clearly obtained that the P3HT FET with MWCNT buffer layer exhibit highest drain current owing to the low contact resistance at electrode/P3HT interface, whereas the FET with Ag NPs printed S-D electrode shows low drain current. The work function of the printed Ag NPs electrodes was estimated as 4.5 eV, whereas that of MWCNT was nearly 4.9-5.0 eV. The work functions of Au, and Au/CuPc were larger than Ag but slightly smaller than MWCNTs. It is therefore, considered that the drain current in Fig. 2 corre-

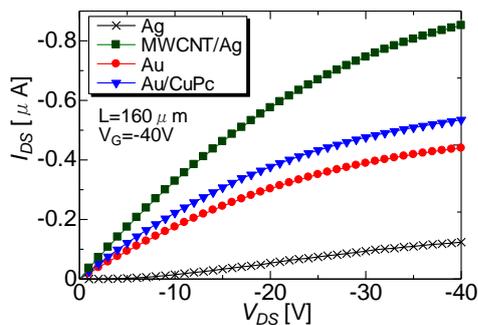


Fig. 2 The output properties of the bottom contact P3HT FETs on CL-PVP gate insulators with various S-D electrodes.

Table I FET parameters of P3HT FETs obtained from Fig. 2

(L=80μm)	V_{th} [V]	μ_h [$cm^2/(V\cdot s)$]	On/Off ratio	S parameters [V/dec]	Contact resistance] $R_c \cdot W$ [$M\Omega cm$] (@ $V_{GS} = -40V$)
Au	8	1.3×10^{-2}	1.1×10^5	6	4.96
Au/CuPc	12	1.5×10^{-2}	3.0×10^5	5	2.41
Ag	13	8.2×10^{-4}	8.3×10^4	7	281
Ag/MWCNT	13	1.9×10^{-2}	2.4×10^5	5	0.64

sponds to the order of the work function of S-D electrodes. Similar tendency was again obtained for the devices in the typical transfer line method (not shown here). We therefore considered that the MWCNTs not only act as S-D electrodes but also act as a hole injection buffer layers. It is interestingly known that the P3HT FETs with Ag/MoO₃ S-D electrodes exhibited highest hole mobility but the it was totally difficult to obtained the OFF states due to their very strong electron accepting properties. The detail will be discussed in the conference.

4. Conclusions

We have fabricated the all-printed organic field effect transistors with MWCNTs or Ag nano-particle (Ag NP) electrodes and their multi-layers (or composite) as printed source-drain (S-D) electrodes. The OFETs with MWCNT S-D electrodes exhibited higher hole mobility and on/off ratios than the devices with Ag NP S-D electrodes (even higher than the device with Au and Au/CuPc S-D electrodes) owing to the lower contact resistance at the MWCNT/semiconductor interface. The high sheet resistance of MWCNT electrodes was considerably reduced by mixing with Ag NPs or multilayer deposition of Ag and MWCNTs and MWCNTs acts as a solution processable buffer layer. On the other hand, although the MoO₃ layers enhances the hole mobility and reduces the contact resistance, the strong electron accepting properties of MoO₃ was resulting to the difficulty in the Off states. We therefore concluded that the MWCNT/Ag stack or composites seems to be a promising candidate of the printed electrodes of organic electronic devices.

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