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The Influence of Gate Insulator Dipoles on Charge Transport in Solution-Processed Top-Gate Organic Field-Effect Transistors with High Mobility and Operational Stability

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1. Introduction

Organic field-effect transistors (OFETs) employing solution-processable organic semiconductors have recently attracted much interest because they enable the large-area fabrication of flexible electronic circuits using low-cost solution processes such as spin coating, ink-jet printing, microcontact printing, screen printing and so on. The electrical characteristics of solution-processed OFETs depend largely on the fabrication processes and the charge mobility of bottom-gate OFETs is known to be strongly affected by the substrate treatment prior to the deposition of organic semiconductors. It has been reported that the field-effect mobilities of bottom-gate OFETs based on a typical polymer semiconductor of regioregular poly(3-hexylthiophene) (P3HT) are significantly increased by the hydrohobic treatment of substrates with self-assembled monolayers (SAMs) [1-3]. This is attributed to the improvement of the orientation of crystallinity of P3HT molecules through the formation of π-stacked lamellar structures.

In previous study, we have fabricated P3HT FETs with bottom-gate, top-gate, and double-gate configurations, and found that top-gate P3HT FETs show high mobilities without SAM treatment processes [4]. In this study, we show top-gate P3HT FETs with different gate insulators exhibit high mobilities and operational stability. We also discuss the influence of dipoles within gate insulators on charge transport in solution-processed top-gate OFETs.

2. Experiments

The schematic diagram of fabricated P3HT FETs with top-gate configurations is shown in Fig. 1. The fabrication processes are as follows. Source and drain electrodes (Cr/Au) were defined on glass substrates by photolithography. After the substrates were ultrasonically cleaned in acetone and isopropanol bath, the surface of the substrates were treated with UV/O_3. Anhydrous chlorobenzene solution of regioregular P3HT was spin-coated onto the substrates and fabricated P3HT thin films were annealed in vacuum at 100 °C for 1 hour. Then, CYTOP^{TM} (Asahi Glass), poly(4-chlorostyrene) (PCS), or poly(methyl methacrylate) (PMMA) was spin-coated on the P3HT layers as a gate insulator, whose relatve dielectric constants k are 2.1, 3.2, and 3.9, respectively. Finally, Al gate electrodes were deposited on gate insulators.

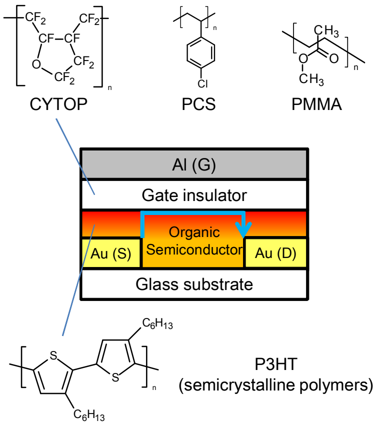

Fig. 1 A device structure of top-gate P3HT FETs and molecular structure of P3HT and polymer gate insulators.

The electrical characteristics of these fabricated FETs were measured in N_2 atmosphere using Keithley 6430 and 2400 source meters.

3. Results and discussion

Figure 2 shows the typical transfer characteristics of top-gate P3HT FETs with CYTOP, PCS, and PMMA gate insulators. Average field-effect mobilities of the devices

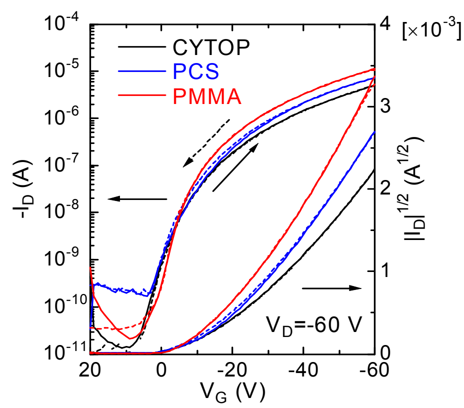

Fig. 2 Typical transfer characteristics of top-gate P3HT FETs with CYTOP, PCS, and PMMA insulators.

with CYTOP, PCS, and PMMA insulators are 2.6 ± 0.1 × 10^{-2}, 1.8 ± 0.4 × 10^{-2}, and 2.6 ± 0.3 × 10^{-2} cm^2/Vs, respectively. The obtained mobilities are much larger than those of bottom-contact P3HT FETs and are almost independent of the relative dielectric constant of gate insulators. We also investigate the operational stability of the top-gate P3HT FETs against gate bias stress. Figure 3(a) shows the variation in the transfer characteristics of the top-gate P3HT FET with the CYTOP insulator before and after applying V_G=–60 V for 10^4 s. The shifts of threshold voltage ΔV_{th} of P3HT FETs with CYTOP, PCS, and PMMA insulators are shown in Fig. 3(b). Although the operational stability of these devices depends slightly on the type of gate insulators, the obtained ΔV_{th} values are much smaller than those reported in the conventional bottom-gate P3HT FETs with SiO_2 gate insulators [3]. These observed high stability of top-gate P3HT FETs is attributable to the highly ordered microstructures formed at P3HT film surfaces [5], resulting in extremely low densities of carrier traps at the P3HT/insulator interfaces.

To gain insight into the influence of the dipoles of gate insulators on charge transport in solution-processed top-gate OFETs, we also fabricate top-gate OFETs using an amorphous organic semiconductor of poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA). The relationships between field-effect mobility and the relative dielectric constant of gate insulators in top-gate P3HT and PTAA FETs are shown in Fig. 4. The P3HT devices show high mobilities irrespective of gate insulating materials, whereas the mobilities of PTAA devices are significantly decreased when the dielectric constant *k* of the gate insulator becomes larger. Such decrease in mobility has been generally reported in OFETs and has been explained by the increase of energy width of the density of localized states due to dipole disorder [6]. The result in Fig. 4 strongly suggests that the influence of dipole disorder on field-effect mobility depends largely on the degree of crystallinity and packing of organic semiconducting molecules. The *k*-insensitive mobilities observed in top-gate P3HT FETs can be attributed to the presence of highly ordered edge-on orientations of P3HT molecules at the P3HT/insulator interface, which

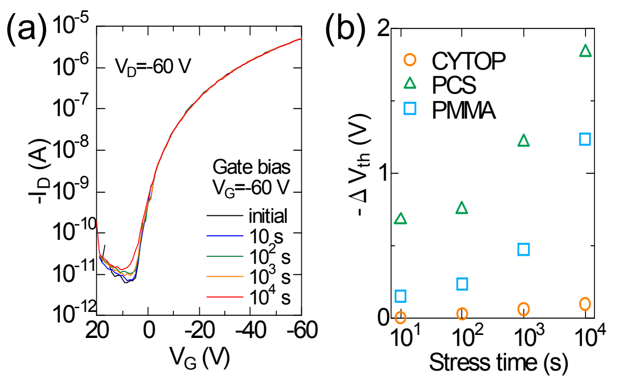

Fig. 3 (a) Transfer characteristics of a top-gate P3HT FET with a CYTOP insulator and (b) threshold voltage shifts ΔV_{th} of P3HT FETs with CYTOP, PCS, and PMMA insulators after gate bias stress of V_G=–60 V (V_D=0 V) for 10, 10^2, 10^3, 10^4 s

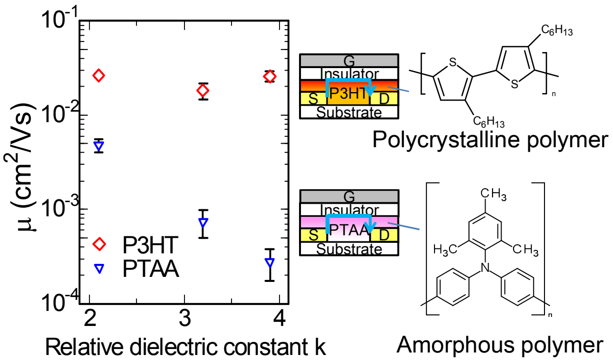

Fig. 4 Field-effect mobilities of top-gate P3HT and PTAA FETs with CYTOP, PCS, and PMMA gate insulators, whose relative dielectric constants *k* are 2.1, 3.2, and 3.9, respectively.

would suppress the electronic coupling between the semiconductor core and the dipoles of the gate insulators through the alkyl side chains of P3HT molecules. Our findings reinforce the superiority of a top-gate OFET with solution-processable polycrystalline organic semiconductors such as P3HT, and suggest an important strategy to fabricate OTFTs with low operational voltage in addition to high mobility and high operational stability by using high-*k* gate insulators and solution-processable organic semiconductors.

4. Conclusions

We have reported that top-gate P3HT FETs with polymer gate insulators show high field-effect mobilities and operational stabilities, which are less affected by the type of gate insulating materials. We have also investigated the influence of dielectric constant *k* of gate insulators on charge transport in solution-processed top-gate OFETs based amorphous PTAA. The obtained results indicate that the presence of highly ordered edge-on structures at the P3HT/insulator interface is responsible for the *k*-insensitive mobility in top-gate P3HT FETs. Our findings provide a possibility of the fabrication of high performance OFETs with low operational voltage using solution-processable organic semiconductors.

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