



Forming semiconductor/dielectric double layers by one-step spin-coating for enhancing the performance of organic field-effect transistors

Chuan Liu^a, Yun Li^a, Takeo Minari^{a,b}, Kazuo Takimiya^c, Kazuhito Tsukagoshi^{a,d,*}

^a International Center for Materials Nanoarchitectonics (WPI-MANA), National Institute for Materials Science (NIMS), Tsukuba, Ibaraki 305-0044, Japan

^b RIKEN, Wako, Saitama 351-0198, Japan

^c Department of Applied Chemistry, Graduate School of Engineering, Hiroshima University, Higashi-Hiroshima, Japan

^d Core Research for Evolutional Science and Technology (CREST), Japan Science and Technology Agency (JST), Kawaguchi, Saitama 332-0012, Japan

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ABSTRACT

We report one-step formation of the gate dielectric and conduction channel for enhancing the performance of organic field effect transistors (OFETs). The resulting OFET with the semiconductor/dielectric bi-layers spun in ambient conditions exhibits μ_{FET} up to $1.6 \text{ cm}^2/\text{Vs}$ and on-off ratio higher than 10^6 , no additional treatment needed. Contact angle measurements and absorption spectra reveals that a well-defined semiconductor-top and dielectric-bottom film form after spin-coating the mixture of the two components, which is due to the surface induced self-organized phase separation. Compared to the single layer semiconductor film, the staggered film exhibits over 5 times higher mobility and nearly 90% reduced hysteresis in OFET. The higher performance is attributed to the simultaneous optimization in the dielectric interface and semiconductor crystallization. The approach is significant for the fabrication of low cost, easy processed and high performance OFETs.

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1. Introduction

Organic field effect transistors (OFETs) based on semiconducting small molecules have been intensively investigated [1]. Developing simple, reliable solution process to extract the intrinsic property of organic semiconducting molecules is one of the main goals. Generally, the semiconductor/dielectric interface are formed via successive deposition of films, e.g. by spin-coating, in solution processed OFETs [2]. This procedure yet may increase the risk of inducing impurities to the interface during the deposition of the second layer, especially when fabricated in air [3]. Hence forming the semiconductor/dielectric interface free of exposure to atmosphere would be beneficial for

realizing reliable, high performance and simplifying the fabrication process.

At the meantime, the OFET fabrication also needs optimization in both the interfacial conditions and the semiconductor crystallinity, because few interfacial traps and crystal boundaries are considered to be crucial for high mobility charge transport [1]. The former is mostly achieved by growing self-assembly monolayers (SAMs) on the dielectric interface which block interfacial traps and lower polarity disorder [4], while the latter is usually controlled by post thermal or solvent annealing [5–7]. However, the use of SAMs may cause de-wetting in spin-coating [8], and shows limited impact on modifying the morphology of the spin-coated semiconducting layers [9]. Therefore simultaneous optimization of the dielectric interface and semiconductor crystallinity remains a challenge for solution processed OFETs.

Here we report the formation of the semiconductor and dielectric layers in one step spin-coating, with the dielectric interfacial conditions and semiconductor

* Corresponding author at: International Center for Materials Nanoarchitectonics (WPI-MANA), National Institute for Materials Science (NIMS), Tsukuba, Ibaraki 305-0044, Japan.

E-mail address: TSUKAGOSHI.kazuhito@nims.go.jp (K. Tsukagoshi).

crystallinity simultaneously optimized. The semiconducting 2,7-didodecyl[1]benzothieno[3,2-b][1]benzothiophene (C12-BTBT) [10] and polymer dielectric poly(methyl methacrylate) (PMMA) self-organize into a staggering bi-layer structure after the spin-coating [11]. Compared to the as-spun C12-BTBT film on SiO₂, the OFETs with staggered film exhibit mobility more than 5 times higher and hysteresis nearly 90% reduced. With the film spun in ambient conditions, the FETs exhibit mobility up to 1.6 cm²/V s and on-off ratio higher than 10⁶.

2. Experimental

2.1. Film fabrication and characterization

The semiconductor C12-BTBT was synthesized as described in [12]. The substrate Si/SiO₂ wafers were cleaned in an ultrasonic bath in a succession of acetone and isopropanol for 10 min each. For the single component film, the semiconductor C12-BTBT was dissolved in chloroform solution (0.4 wt.%) and spin-coated on Si/SiO₂ substrates (500 rpm for 5 s, 2000 rpm for 40 s). For the mixture film, C12-BTBT and PMMA (Fluka, Mw = 100 k) were mixed in the mass ratio of 1:1 and dissolved in chlorobenzene solvent (1.2 wt.% in total). Then the solution was spin-coated onto the substrates (500 rpm for 5 s, 2000 rpm for 40 s). The above procedures were all processed in ambient conditions.

The optical images of film were captured by using the microscope Nikon Eclipse LV100POL, with or without polarizer as described. The thicknesses of the films were measured by the surface profiler (Model P-16, KLA Tensor). The water contact angles of selected surfaces were measured by the contact angle goniometer (Imoto Corporation) with de-ionized water as the sessile drop. UV–Vis absorption spectra were measured from the film on glass substrates by the UV/Vis/NIR spectrophotometer JASCO V-570. The absorptions of glass substrate were deduced from the film signals.

2.2. Transistor fabrication and characterization

Highly doped n-type (100) silicon wafers with 50 nm SiO₂ layers were cleaned as described above. Films were deposited as stated above without any post annealing, followed by thermal evaporation of selected materials through shadow masks in vacuum ($<3 \times 10^{-4}$ Pa). For gold only contact, Au was evaporated in the rate of 0.1 Å/s and the final thickness is 40 nm. For contact doping, firstly FeCl₃ was evaporated in the rate of 0.1 Å/s for 1 nm thick film and secondly Au was evaporated as stated above. Channel width (W) and channel length (L) of the device are 1200 μm and 300 μm, respectively. Then the FETs were measured under vacuum ($<4 \times 10^{-3}$ Pa) using an Agilent 4156C semiconductor parameter analyzer. The saturation mobility of OFET, μ_{FET} , and threshold voltage, V_{th} , are extracted from saturation regime in the transfer characteristics by fitting $-I_{\text{d}} = [W/(2L) \times C_i \times \mu_{\text{FET}}] \times (V_{\text{g}} - V_{\text{th}})^2$,

where I_{d} is drain current, C_i is capacitance per unit area, V_{g} is gate voltage, and V_{th} is threshold voltage.

3. Results and discussions

3.1. Forming semiconductor and dielectric layers in one-step spin-coating

The C12-BTBT film spun from chloroform onto bare SiO₂ exhibit relatively low mobility in OFET with Au contact ($\mu_{\text{FET}} \sim 0.1$ cm²/V s, data shown in the next section). Yet the μ_{FET} of vacuum deposited C12-BTBT film reaches 3.9 cm²/V s on the octadecyltrichlorosilane (ODTS) SAM layer [10], and has a theoretical limit of 40~50 cm²/V s in the single crystalline state [13], implying possibility for further enhancement for the spin-coated film. However, spin-coating C12-BTBT onto the SiO₂ treated by hydrophobic hexamethyldisilazane (HMDS) causes severe de-wetting, not able to form a continuous film larger than 0.5 cm². Such de-wetting also happens when spin-coating onto bare SiO₂ in high boiling point solvent (e.g. chlorobenzene). Instead we adapt the polymer dielectric PMMA as the charge transport interface. To form the layers of C12-BTBT and PMMA in one step, we investigated the potential of self-organized phase separation method, i.e. the two solutes automatically separate and form bi-layer structure in spin-coating [11].

The mixture of C12-BTBT and PMMA in the chlorobenzene solvent was spin-coated onto the Si/SiO₂ substrates. The two components in the film are possibly evenly distributed in each other or phase separated, either laterally to form isolated domains or vertically to form staggered continuous layers [7]. We used orthogonal solvent to selectively dissolve the component and reveal the film structure. Firstly, the substrate was vertically held and partially immersed into acetone, which dissolve both C12-BTBT and PMMA, followed by drying with nitrogen air flow. This step exposed the SiO₂ surface (Fig. 1a, region I). Secondly, the substrate was partially immersed into cyclohexane, which dissolve C12-BTBT moderately (~ 2 mg/ml) but not PMMA, and dried. This step forms the region II in Fig. 1a, next to which is the surface of the pristine film (region III). Under the cross-polarized microscope, the region II is totally dark and thus amorphous, while the region III exhibits strong birefringence and thus is crystalline (Fig. 1a, with polarizer). Surface profile scanning across the regional edges clearly reveals two staggering layers (Fig. 1b), each about 20~40 nm thick. According to the solubility, the bottom layer is presumed to be PMMA and the top layer is presumed to be C12-BTBT. Therefore we propose a process of self-organized phase separation as illustrated in Fig. 1c.

We further provide evidences to confirm the film structure by measuring water contact angles and absorption spectra of each region. As shown in Fig. 2, the water contact angle of the bottom layer (region II) resembles that of the as-spun PMMA film ($\sim 68^\circ$), while the value measured on the top layer (region III) is almost the same with that on the as-spun C12-BTBT film ($\sim 100^\circ$). In UV–Vis absorption spectra, the as-spun film of the C12-BTBT/PMMA mixture exhibits a strong absorption peak at

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