

Microstructure transformations induced by modified-layers on pentacene polymorphic films and their effect on performance of organic thin-film transistor

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ABSTRACT

Phenyltrimethoxysilane was used to modify SiO₂ insulator and significantly enhanced the pentacene based organic thin-film transistors (OTFTs). The crystal structure, surface morphology, molecular structure and microstructure of pentacene polymorphic films with and without the modifications were investigated using X-ray diffraction (XRD), atomic force microscopy (AFM) and contact angle meter. XRD studies reveal a decreased tilt angle (θ_T) of pentacene molecules from *c*-axis toward *a*-axis, indicating that polymorphs transformation from the “triclinic bulk” phase to the “thin film” phase and orthorhombic phase occurs. AFM images show that the surface roughness of gate insulators has no influence on performance of the pentacene based OTFT. These results provide strong evidence that the performance improvement of OTFT after PhTMS modification of SiO₂ insulator surface is related to the microstructure transformation of the semiconductor. It suggests that the modified-layer may alter the molecular geometry and further induce structural phase transitions in the pentacene films for the performance improvement.

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1. Introduction

In recent years organic semiconductors have been extensively studied for various applications in ubiquitous

low cost electronics such as solar cells, sensors [1–3], light-emitting diodes, and organic thin-film transistors (OTFTs) [4,5]. The great application potentials have been recently boosted up as the OTFT performance becomes comparable to the amorphous silicon-based transistors [6,7]. It is known that a polycrystalline organic semiconductor film possesses an inherent order relative to an amorphous film for facilitating more efficient charge transport, and thus is the most critical component in an OTFT. Generally, thin film forms more preferable than a single crystal due to its well-defined supramolecular organization, of which the interface microstructure on the insulator layer is crucial for efficient charge transport in the OTFTs [8,9]. The microstructure of an organic semiconductor/gate

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insulator interface can be altered by its surface modification for performance improvement of OTFTs [10–12].

Pentacene is a promising organic semiconducting material for OTFTs due to its high mobility and good semiconducting properties. A thermal evaporation-deposited pentacene film on SiO₂ surface consists of a substrate-induced “thin film” phase and a “triclinic bulk” phase that can be detected only when its thickness is large enough. The triclinic structure of the pentacene single crystal composes two molecules in a unit cell with lattice parameters of $a = 0.628$ nm, $b = 0.771$ nm, $c = 1.444$ nm, $\alpha = 76.75^\circ$, $\beta = 88.01^\circ$, and $\gamma = 84.52^\circ$ [13], and the pentacene molecules are packed into a layered structure to form a herringbone pattern. Thickness-driven phase transformation from orthorhombic to “triclinic bulk” phase is believed to take place in the thin films [14,15]. However, the modified-layer-driven microstructure transformation of a pentacene polymorphic film at a fixed thickness has not yet been reported. Further, pentacene films on various modified gate insulator surfaces could reveal their polycrystalline structure and morphologies as well. Since the structural change of an organic semiconductor layer in the channel is critical to the OTFTs performance, there is a great need to investigate the growth dynamics and microstructure transformation of a pentacene film caused by modified-layers on the dielectric surface for fundamental insights, leading to further improvement of OTFT performance.

In this work, we fabricated a series of pentacene OTFTs with various interface modified-layers on thermally grown SiO₂ gate insulator by using phenyltrimethoxysilane C₆H₅Si(OCH₃)₃ (PhTMS) for performance improvement, while investigating the structural phase transformations induced by various modified-layer such as octadecyltrichlorosilane (OTS) and PhTMS on SiO₂ gate insulator. For the first time, we experimentally studied effects of the modified-layer on the molecular vibrational modes and structure change of the organic semiconductor. These microstructure transformations might imply the carrier transport improvement in pentacene OTFT devices.

2. Experimental

Top-contact (TC) pentacene OTFTs were fabricated with heavily doped silicon substrates by thermally growing 310 nm SiO₂ (SiO₂, dielectric constant $k = 3.35$) dielectric layer as the gate electrodes (Fig. 1a). The SiO₂/Si substrates were diced followed sequentially by cleaning in sulfuric acid/hydrogen peroxide (7:5 v/v) at room temperature for 45 min, rinsing thoroughly with de-ionized (DI) water, ultra-sonicating for 30 min in acetone and DI water, rinsing and drying under nitrogen. The cleaned substrates were immediately coated with PhTMS or OTS modified-layer via spin coating at a rate of 3000 rps for 30 s. The coating solutions containing 2.3% PhTMS and 2.3% OTS in toluene (v/v) were used, respectively for spin-coated modifications [16]. The molecular structures of PhTMS and OTS and chemical process between modified-layer and surface of SiO₂/Si substrate are shown in Fig. 1b–d, respectively. A 60 nm-pentacene thin film was subsequently deposited under a vacuum of $\sim 10^{-6}$ Pa at a deposition rate of 0.03 nm/s and room temperature for the substrate. Finally, Au source and drain electrodes (60 nm thickness) were deposited by thermal evaporation through a shadow mask. All devices were fabricated to have an identical channel width and length ratio of $W/L = 570 \mu\text{m}/57 \mu\text{m}$. All electrical characterizations were carried out under ambient conditions using Agilent Semiconductor Analyzer (E5270B) + Cascade probe station.

3. Results and discussions

The output characteristics of the fabricated devices in Fig. 2 show saturation behaviors at high V_{DS} for all devices. Interestingly, the saturation currents are increased remarkably with modified-layers, especially with PhTMS. As shown in Fig. 2c, the output characteristics of the PhTMS-treated OTFT device t display a pronounced curvature at low V_{DS} , indicating the existence of contact resistance (R_C), possibly due to the energy barrier for

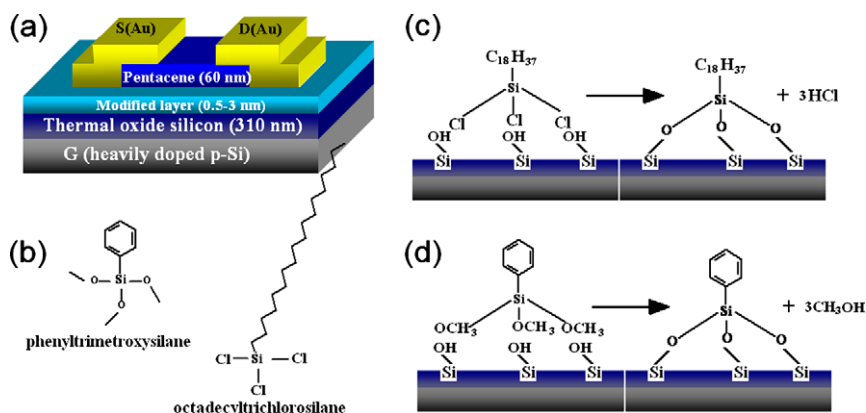


Fig. 1. (a) Schematic view of top-contact OTFT using different silane modified-layer/SiO₂ gate insulators; (b) structure of phenyltrimethoxysilane and OTS; (c) chemical process between OTS and surface of SiO₂/Si substrate; and (d) chemical process between phenyltrimethoxysilane and surface of SiO₂/Si substrate.

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