Metal-organic pentacene derivative with well ordered morphology for the application of low voltage organic thin film transistors

V. A. L. Roy, Zong-Xiang Xu,Yong-Gang Zhi, Sze-Chit Yu, and Chi-Ming Che*

Department of Chemistry and the HKU-CAS Joint Laboratory on New Materials, The University of Hong Kong, Pokfulam Road, Hong Kong SAR, China

Abstract

Metal organic pentacene based low voltage organic thin film transistors with field effect mobility as large as 0.8 $cm^2/V^{-1}.s^{-1}$ and on/off current ratio larger than 10^6 have been fabricated. Thin films deposited by evaporation at different deposition rate has different morphology which leads to a difference in transistor characteristics. The films with a deposition rate of 2A/sec has better morphology and also the transistor behavior. AFM (atomic force microscope) and STM (scanning tunneling microscope) were used to understand the morphology and ordering of the molecules on the silicon surface which helps the transistor to operate at low voltages. The results presented here show a strong correlation between molecular ordering and the need of well-ordered films for the performance of organic thin film transistors (OTFT's).

Introduction

Organic materials have been drawn intensive attention as active layers in diverse electronic applications, especially for organic thin film transistors (OTFTs) [1-6], organic light-emitting diodes (OLEDs) [7-9] and organic solar cells [10-12]. Pentacene is one of the most well characterized organic semiconductor which demonstrates the remarkable field-effect mobilities in OTFTs (0.3-0.7 $cm^2V^{-1}s^{-1}$on SiO_2/Si [13], 1.5 $cm^2V^{-1}s^{-1}$on modified SiO_2/Si [14], and 3 $cm^2V^{-1}s^{-1}$on polymer gate dielectric [15]). Despite these advances, the practical applicability of pentacene has been rendered by the poor solution processability in organic solvents and poor stability in ambient environment [16]. The progresses in modifying the pentacene structure are recently highlighted [17-20]. A recent notably example is the modification of pentacene to 6,13-bis(triisopropylsilylethynyl)pentacene (**1**) by Anthony and co-workers for OTFT applications [21-23]. The field-effect mobilities of **1** have been reported as 0.05 $cm^2V^{-1}s^{-1}$ on octadecyltrichlorosilane (OTS) gate dielectric treatment and 0.4 $cm^2V^{-1}s^{-1}$with self-assembly layer configuration, respectively.

Herein, we introduce an optimized organic field effect transistors based on 6,13-bis(triisopropylsilylethynyl) pentacene **1** as active material. The compound was readily prepared with product yields up to 91% using a synthetic strategy based on a simple iterative and convergent Pd/Cu-coupling and desilylation protocol. Later, the material was purified by train sublimation process. We are interested in this molecule because of its molecular ordering with a smooth morphology. For device fabrication, thin films were deposited by thermal evaporation under high vacuum at room temperature on patterned silicon dioxide substrates. The charge carrier mobility at room temperature extracted from FET measurements

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is up to 0.8 $cm^2/V^{-1}s^{-1}$. It is explained that better morphology of the films and molecular ordering is important for the charge transport. Thin films were deposited with a deposition rate of 0.1, 2, & 5 Å/s on top of patterned substrates under high vacuum conditions at room temperature. It is explained that the difference in the deposition rate affects strongly the mobility and it is seen in the morphology of the films. One mono-layer (ML) of the films at the optimal deposition rate studied by STM, reveals a clear molecular ordering which could be a possible reason for the low voltage operation & high mobility of the transistor.

The OFET is based on the common substrate-gate structure. Gate oxide SiO_2 layer (100 nm, permittivity = 3.9) was thermally grown on n-type Si substrates (the gate electrode). Image reversal photolithography technique was used to form opening on the photoresist layer for the source and drain patterns on the gate oxide. The substrate was then loaded onto a thermal evaporation system. Source and drain metal layer, consisting of the lower Ti adhesion film (10 nm) and the upper Au conductive film (50 n.m) was deposited by thermal evaporation. After metal film deposition, standard lift-off process in acetone were used to remove excess metal films on top of the photoresist pattern, leaving behind the Ti/Au source/drain contact pattern.

The source/drain metal pattern on gate oxide substrate was thoroughly rinsed and cleaned in IPA (isopropyl a1cohol) and deionized water respectively, prior to drying under nitrogen atmosphere. The profile of the Au electrode was characterized with AFM. The profile was observed to have a smooth slope and regular pattern along the entire channel width.

The active organic layer with a thickness of 50 nm was deposited with a deposition rate of 0.1, 2, & 5 Å/s on top of the patterned substrates under high vacuum conditions at room temperature. The transistor output and transfer characteristics were measured with a probe station under nitrogen atmosphere inside a Mbraun MB20G using Keithley semiconductor parameter analyzer 4200 SCS.

Results and discussions

Figure 2 displays a tapping mode Atomic Force Microscope (AFM) images of a 10 nm film deposited at 0.1, 2, & 5 Å/sec respectively. Inspection of the image reveals the morphology of the film is smooth and roughness of the film is ca. 0.849 nm for films deposited at 0.1 and 0.810 for the films deposited at 2 Å/sec and 2.3 nm for the film deposited at 5 Å/sec. For the films deposited at 0.1 and 2 Å/sec have mobilities 0.64 and 0.8 respectively, while the films deposited at 5 Å/sec gave a mobility of 0.09 Å/sec. This implies morphology and the smoothness of the films is an important factor for OFET devices.

To confirm the molecular ordering, STM measurements were used to study one monolayer film on a heavily doped Silicon surface which was used for the transistor devices. Figure 3 shows 3-dimensional and flatten STM images with an

area of 100 nm^2 recorded at a bias voltage of 2 V. Here the molecules are observed to form periodic parallel rows. In our case, mainly this molecular ordering has an impact on the charge transport which helps the transistor to operate at low voltages with better mobility. The transistor has a low threshold voltage of 0.2 v and a negligible contact resistance explain the enhanced transport due to this molecular ordering.

Figure 4 (a) and (b) shows the I-V (Current-Voltage) and transfer characteristics of the device respectively, deposited at 2A/sec which has better morphology for the film. The transistor has a channel length and channel width of 20 and 300 μm respectively. The transfer characteristics were measured at drain voltage of -2 V. The field-effect mobility is 0.8 $cm^2V^{-1}s^{-1}$ measured at a gate-source voltage of -3 V. A threshold voltage of 0.2 V and on/off current ratio of 10^6 was obtained. This is one of the best mobilities and low threshold voltage reported for organic transistors without self assembly configuration. Figure 5 (a) shows the subthreshold region measured at drain-source voltage of -1 V and a subthreshold swing of 200 mV per decade was obtained. The current decay property and stability of the device under prolong bias was verified by transient measurements with a constant applied voltage to the gate and drain of -5 V. Figure 5 (b) depicits the characteristics of current decay as a function of time. Notably, the current was found to decay slowly in a non-single exponential decay manner. The device required several minutes of prolong bias for complete current decay down to the noise threshold, which shows the stability of the device.

In summary, we have demonstrated OTFTs based on a functionalized pentacene compound, 6,13-bis(triisopropylsilylethynyl)pentacene that exhibits pronounced improvement on field effect mobility up to 0.8 $cm^2V^{-1}s^{-1}$ with low threshold voltage and the results on AFM and STM explains the importance of film morphology and molecular ordering for planar charge transport in the performance of low voltage organic thin film transistors.

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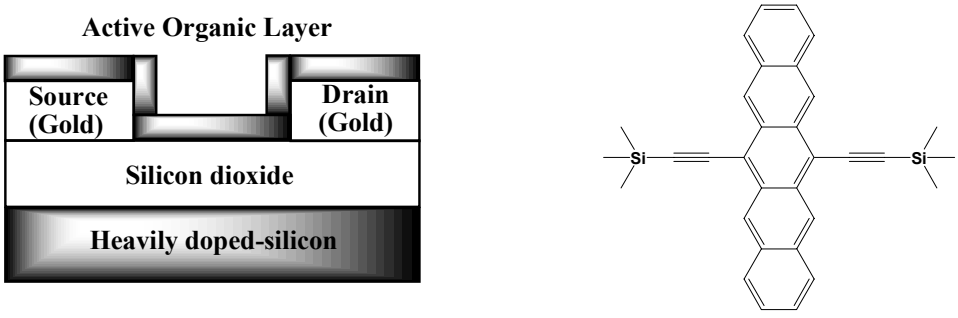

Fig. 1. Schematic structure of bottom contact OTFT and Chemical structure of 6,13-bis(triisopropylsilylethynyl) pentacene.

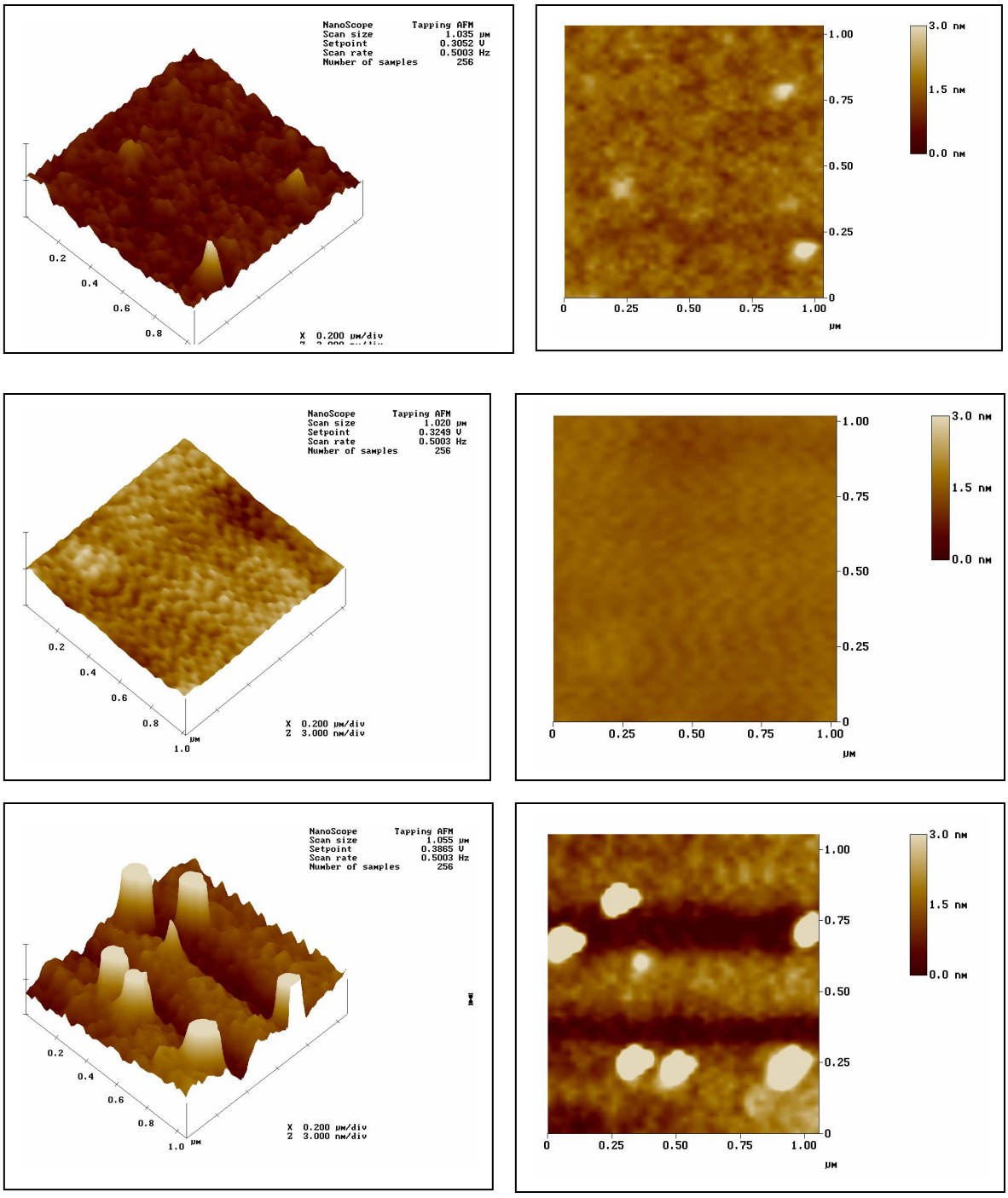

Figure 2: AFM tapping mode 3-dimensional and flatten images of 5 nm thick films deposited at 0.1, 2 and 5 A/sec respectively.

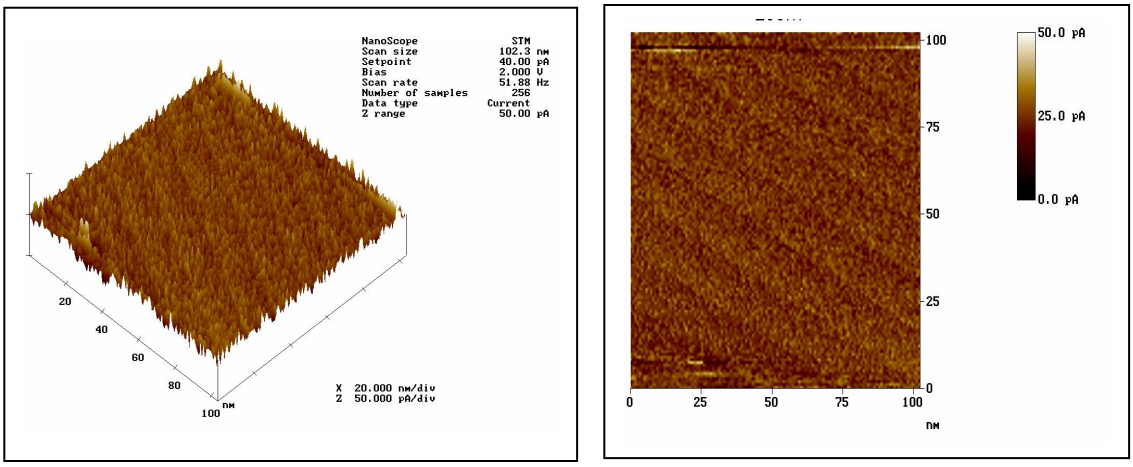

Figure 3: STM 3-dimensional and flatten images of one monolayer thin film deposited at 2 A/sec

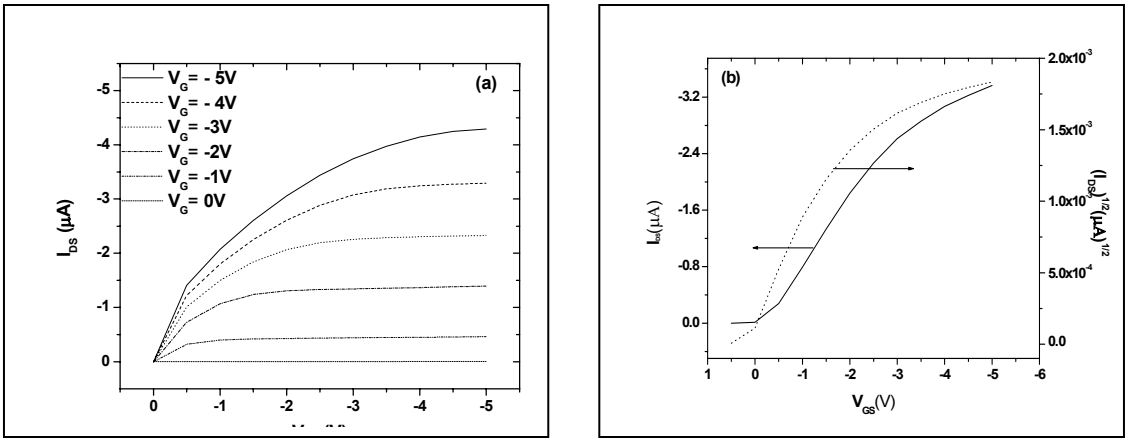

Figure 4: (a) Transistor output characteristics and (b) Transfer characteristics measured at drain-source voltage -2V.

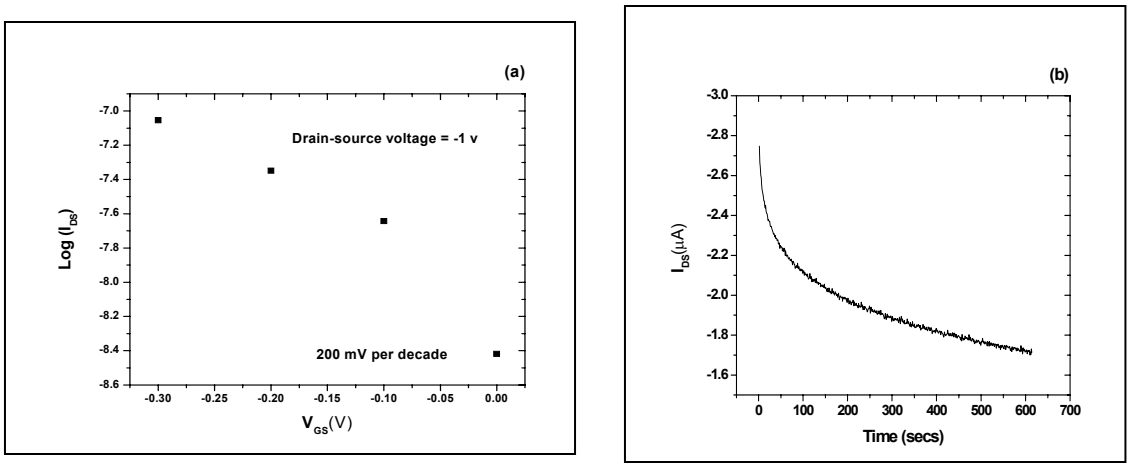

Figure 5: (a) Subthreshold region, showing a subthreshold swing of 200 mV per decade, (b) Transient characteristics, keeping drain-source and gate voltages at -5 V.