Organic Electronics 16 (2015) 164–170

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Electrical transport mechanism of single monolayer pentacene film employing field-effect characterization

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ARTICLE INFO

Article history:
Received 3 September 2014
Received in revised form 8 October 2014
Accepted 14 October 2014
Available online 15 November 2014

Keywords:
Single molecular layer
Field effect mobility
Hopping transport
Non-Poole–Frenkel effect

ABSTRACT

We employed a novel electrode-contact architecture to enable operation of single monolayer pentacene-based field-effect transistor with a high electrical performance, whose mobility reaches as high as 0.31 $cm^2 V^{-1} s^{-1}$, the highest among any pentacene-based monolayer devices ever reported. A temperature dependent charge transport was systematically carried out to elucidate the carrier transport mechanisms within even single layer of molecules. The carrier mobilities are found to exhibit a pure Arrhenius type at the high temperature regime (above 170 K), by contrast, a pronounced turning point has been observed when the measurement temperature is below 170 K, possible mechanisms were ascribed to the distribution attribute of trapped states among the grain boundary. Furthermore, an electric field dependent mobility characterization shows a unique non-Poole–Frenkel type behavior at room temperature, which shows much different from the case occurred in multiple-layer devices where the quantity of permitted percolation routes among different monolayers guarantees an mobility enhancement as an electric field increasing. But for the case of single monolayer, the electric field-induced potential reduction effect is competing with a drop of percolation path arising from the directional movement of carrier under a strong electric field. The depth understanding of carrier transport within one monolayer may be helpful for optimizing the design of OFETs for better device applications.

1. Introduction

Organic field effect transistor (OFET), one of the most fundamental devices of organic semiconductor, is a significant component in application of organic logic circuit, OLED driving unit and various organic sensors, etc. It is very meaningful to deeply understand the charge transport mechanisms in OFET for the aim of improvement of devices' design. Therefore, a considerable amount of works have been and are being done on the description of the charge transport in OFET [1–3]. On one hand, several phenomenological models have been employed to describe the charge transport mechanisms in organic semiconductors. Traditionally, multiple trapping and release (MTR) [4] and multiple hopping model [5] were applied to characterize the transport occurred in organic polycrystalline films. On the other hand, it is widely accepted that the quality of the molecular stacking and the organic-dielectric interface properties play a fatal role in OFET. One unique feature in OFET is, charge transport, under the gate electric field, mainly occurs within a few nanometers with respect to the interface between dielectric layer and active layer [6,7]. Therefore, a considerable amount of work has been done trying to describe the charge transport within the interface of OFET.

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http://dx.doi.org/10.1016/j.orgel.2014.10.051
1566-1199/

One effective way to study the charge transport within the interface is to fabricate transistors with an ultrathin organic active layer, which helps us realize much more intrinsic electrical characterization of the interface by avoiding the impact of charge carriers distributed far from the interface. Several research groups have made efforts on studying transport properties of ultrathin OFET, however encountering challenges. Jung and his coworkers [8], fabricated single pentacene molecule layer transistor with traditional top source-drain contacts, first got the electrical characteristics of pentacene single monolayer film, of which the mobilities are only around 0.01 $cm^2\,V^{-1}\,s^{-1}$; Asadi et al. renewed the record to 0.05 $cm^2\,V^{-1}\,s^{-1}$ by employing SAM modified bottom contact electrodes to improve the carrier injection efficiency [9]. Even though, the mobilities are much lower than those of devices with thicker pentacene film. The major cause of poor performance may come from the inferior metal–organic contacts, since charge injection at the metal–organic interface is rather important for devices' performance. For an ultrathin film or even the single molecule layer film, conventional methods of electrode fabrication are damageable to the molecules' arrangement in the fragile organic layer. Consequently, few systematic electrical characterization work has been done on the ultrathin OFET because both the contact resistance can pre-dominate over the channel resistance and the poor organic molecular arrangement near the contact may deteriorate the electric properties of the devices.

In this paper, we fabricated pentacene-based organic single monolayer transistor with a novel method by imprinting an elastic electrode directly onto the monolayer film, and deliberate controlling for the interface and growth parameters has been carried out, so as to reach a field effect mobility as high as 0.31 $cm^2\,V^{-1}\,s^{-1}$, a value that is much higher than those of any previous work. Using devices of such high quality, we systematically performed characterization of 2D electrical properties for single monolayer pentacene OFETs. A large measurement temperature range spanning from 300 K to 130 K was covered to exam the temperature dependent mobility. At the mean time, a unique non-Poole Frenkel relationship between mobility and lateral electric field was also found. Possible mechanisms are fully discussed at a frame of carrier hopping between the trapped states among the grain boundaries.

2. Experimental

In the fabrication of the devices, heavily n-doped silicon wafers with thermally grown oxidized layer of 300 nm were employed as substrates/gate electrodes, toluene solution of polystyrene (PS) was spin-coated onto the substrate as a modified layer. After which, one monolayer (a nominal thickness of 1.5 nm) pentacene (Aldrich Co.) film was accurately deposited by physical thermal deposition (Auto-306, Edwards Co.) under pressure of 1.2×10^{-7} mbar with a deposition rate of 0.025 nm/s which was calibrated and monitored by a quartz oscillator.

Conventional thermal deposition of gold was not suitable for fabrication of drain and source electrodes on single monolayer film any longer, since the gold beam emitting with high kinetic energy is destructive to the fragile layer. Instead we developed a novel method to form the ohmic drain and source electrodes utilizing a soft imprinting with an elastic mechanical contact to ensure a device operating with a channel length of 100 μm and width of 2000 μm, the detailed fabrication process can be found in our previous work [10].

The morphology of pentacene single monolayer films were characterized using a Dimension 3100 (Bruker Co.) atomic force microscope (AFM) at tapping mode. The electrical characteristics of the devices were measured using a Keithley 4200 semiconductor analyzer and a home-made ultrahigh vacuum four-probe system under pressure of 1×10^{-8} mbar, at temperature ranging from 130 K to 300 K, drain–source voltage (V_{ds}) fixed at −3 V, −6 V, −9 V, −12 V, −15 V, −60 V, respectively. When the measurement temperature was lower than 130 K, the OFET signal dropped significantly, probably due to the frozen of carriers into the deep trap states [11]. Another possible explanation may come from the drain-source contact at which temperature the channel was probably broken arising from the strain under low temperature. The electrical signal can be recovered when the temperature rises again. Temperature window from 130 K to 300 K is somehow enough to character the unique transport properties for a 2D pentacene OFET. Mobility of the devices at saturate region and linear region were respectively obtained using the following two expressions

$$I_{sat} = \frac{\mu W C_{OX}}{2L}(V_G - V_T)^2 \quad (1)$$

$$I_{lin} = \frac{W}{L} C_{OX}\mu\left[(V_G - V_T)V_{ds} - \frac{V_{ds}^2}{2}\right] \quad (2)$$

where V_G is the gate voltage, V_T the threshold voltage, and C_{ox} the capacitance of the dielectric layer per unit area.

3. Results and discussion

3.1. Fundamental characteristics

Fig. 1a shows the schematic cross-section of a device structure used in our research. Polydimethylsiloxane (PDMS) covered with 40 nm thick Au film, forming a soft template, ensures the ohmic contact between gold electrodes and pentacene single monolayer film. By this means, we raised the recorded highest mobility [9] from 0.05 $cm^2\,V^{-1}\,s^{-1}$ to 0.31 $cm^2\,V^{-1}\,s^{-1}$, on/off ratio 10^5 as shown in Fig. 2b. The output curves are demonstrated in Fig. 2a. The perfect curves at different gate voltages focused at zero point and the relative quick-start drain current at low drain voltage suggested the injection barrier for carrier can be neglected small. Considering the carrier injection length for single monolayer device can be neglected [12], we could get intrinsic electrical characteristic of the channel. This guaranteed a systematical study about the charge transport mechanism in the interface of OFET.