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Temperature-dependent ambipolar electrical characteristics of pentacene-based thin-film transistors: The impact of opposite-sign charge carriers

Tsung-Jun Ho [a], Horng-Long Cheng [a,*], Liang-Yun Chiu [a], Wei-Yang Chou [a], Fu-Ching Tang [b]

[a] Department of Photonics, Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan
[b] Department of Physics, National Cheng Kung University, Tainan 701, Taiwan

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ABSTRACT

The temperature-dependent electrical and charge transport characteristics of pentacene-based ambipolar thin-film transistors (TFTs) were investigated at temperatures ranging from 77 K to 300 K. At room temperature (RT), the pentacene-based TFTs exhibit balanced and high charge mobility with electron (μ_e) and hole (μ_h) mobilities, both at about 1.6 cm^2/V s. However, at lower temperatures, higher switch-on voltage of *n*-channel operations, almost absent *n*-channel characteristics, and strong temperature dependence of μ_e indicated that electrons were more difficult to release from opposite-signed carriers than that of holes. We observed that μ_e and μ_h both followed an Arrhenius-type temperature dependence and exhibited two regimes with a transition temperature at approximately 210–230 K. At high temperatures, data were explained by a model in which charge transport was limited by a dual-carrier release and recombination process, which is an electric field-assisted thermal-activated procedure. At $T < 210$ K, the observed activation energy is in agreement with unipolar pentacene-based TFTs, suggesting a common multiple trapping and release process-dominated mechanism. Different temperature-induced characteristics between *n*- and *p*-channel operations are outlined, thereby providing important insights into the complexity of observing efficient electron transport in comparison with the hole of ambipolar TFTs.

1. Introduction

Organic semiconductors (OSCs) have been extensively studied for application in various electronics and photonics. The semiconductor and charge transport (CT) properties of OSCs are relative to their molecular properties (i.e., delocalized π-molecular orbitals). Electron and hole carriers transfer rely on the CT between the highest occupied highest occupied molecular orbital and the lowest unoccupied molecular orbital of OSCs, respectively; thus, these OSCs are intrinsically capable of conducting both electrons and holes, that is, ambipolar carrier transportation [1]. Ambipolar thin-film transistors (TFTs) can exhibit both *n*- and *p*-channel electrical characteristics according to the given gate bias (V_G, positive for *n*-channel and negative V_G for *p*-channel), thus enabling more diversified applications in a simple way such as complementary-like inverters, light-emitting TFTs, memory, sensor, photodetectors, and so on [1,2]. Moreover, the behaviors of the hole and electron carriers could be investigated together with the ambipolar TFTs. The relative transport capability of electrons versus holes in OSC-based devices has attracted a great deal of interest in fundamental research, as well as in various devices [1–4]. To date, in contrast to unipolar CT properties, little information is available on the ambipolar CT properties of organic TFTs (OTFTs) because of the difficulty in realizing efficient and balanced hole and electron mobilities [2,4–6]. Nevertheless, understanding the fundamental CT phenomena of OSCs in devices is important to extend the fundamental knowledge and develop novel applications of various devices, as well as further optimize device performance.

Temperature (*T*)-dependent measurements are widely used for examining the CT behavior and mechanism of semiconductors. The *T*-dependent CT behavior of unipolar OTFTs have been extensively studied [4,7–10], particularly for *p*-channel devices. For OSCs with polycrystalline structure, the thermally activated behavior of field-effect mobility (μ) is generally observed and usually interpreted by multiple trapping and release (MTR) mechanism [11]. The MTR model assumes that CT occurs through delocalized states and impeded by impurities, defects, grain boundaries, and so on. These trapped carriers can be thermally released to reach the conduction band, where they can be trapped again.

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* Corresponding author.
E-mail address: shlcheng@mail.ncku.edu.tw (H.-L. Cheng).

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Alternatively, the hopping transport (HT) model also provides a possible scenario for Arrhenius-like μ-T dependence [12]. In this model, the states for charge carriers are considered localized because of the weak intermolecular coupling between adjacent organic molecules. At elevated T values, the thermal energy is sufficient to overcome small energy barriers between the localized states. In contrast to unipolar CT, very few studies on the effects of T on ambipolar OTFTs have been conducted [3,4,13]. For ambipolar OTFTs, the presence of the opposite-sign carriers has large effects in electrical characteristics [2]. Recently, we showed that the recombination and dissociation of electron–hole pairs, that is, the dual-carrier recombination and release (DCRR) process, can occur in the active channel, thereby significantly affecting electrical and mobility behaviors [5]. To date, our understanding of the effects of T on the ambipolar CT behaviors of OTFTs remains limited. To the best of our knowledge, no studies have examined whether the DCRR process have large impact on μ-T behaviors.

In this study, we investigated the effects of T on the ambipolar CT properties of pentacene-based OTFTs. Pentacene-based ambipolar OTFTs have been realized by several previous undertakings [2,14,15]. More recently, we showed that pentacene has great potential as an efficient and balanced ambipolar transistor semiconductor [16]. Hole mobility (μ_h) and electron mobility (μ_e) values above 1.0 cm^2/Vs were obtained by fine-tuning the interfacial properties between semiconductor and gate dielectric. Excellent samples were provided to further investigate the μ-T relationships of both electron and hole carriers in the same active channel under similar microstructural characteristics. Furthermore, we discussed the role of DCRR processes in the μ-T relationships, highlighting CT-T behaviors of ambipolar OTFTs completely different from that of unipolar OTFTs.

2. Experimental

This study employed the inverted staggered TFT configuration with a heavily doped silicon wafer as the gate electrode, poly (methyl methacrylate) (PMMA)-modified silicon dioxide (SiO_2) as the gate dielectric, and silver (Ag) source and drain electrodes as described elsewhere [5,16]. To reduce the electron traps, a PMMA buffer layer, which was prepared from 1.0 wt% p-xylene solution, with minimized coverage of the polar ester methyl groups, which act as electron traps, were prepared by spin-coating directly onto the SiO_2 layer [16]. Then, a ca. 60 nm pentacene active layer was thermally sublimed at a rate of 0.3 Å/s upon the gate insulator surface at a pressure of 6×10^{-6} torr. Then, the source and drain electrodes were deposited through a shadow mask on the pentacene surface. The channel width (W) and channel length (L) of the TFTs are 2000 μm and 100 μm, respectively. All electric measurements were performed in a dark vacuum chamber with a liquid nitrogen cooling system and incorporated with Lakeshore 332 to control the T value from room T (RT) down to 77 K. The TFTs were characterized using a Keithley 4200-SCS semiconductor parameter analyzer. The capacitance (C_i) value of the metal/gate dielectric/metal diodes was evaluated using an Agilent E4980 LCR meter.

3. Results and discussion

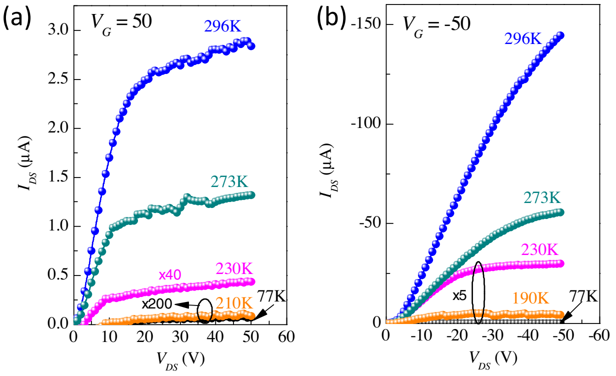

Fig. 1. Output characteristics of pentacene-based ambipolar OTFTs measured at different temperatures: (a) n-channel operations and (b) p-channel operations. Gate voltage (V_G) is also indicated.

Fig. 1 shows the comparison of the typical output current–voltage (I–V) characteristics of the pentacene-based OTFTs operating at a gate voltage (V_G) of |±50| V and measured at different selected T values. For both n- and p-channel operations, the output drain current (I_{DS}) significantly decreased with decreasing T. At T < 210 K, for n-channel operations, only a very small I_{DS} was detected, and its level was very close to the gate current (I_G). For p-channel operations, however, field-effect characteristics were still detected when T decreased to 77 K (the saturated I_{DS} of ca. 0.01 μA at $V_G = -50$ V). The observed significantly large I_{DS} of p-channel operations than that of n-channel operations is a result of large switch on (V_{on}) for n-channel operations [5]. Obviously, the n-channel operation, i.e., electron transport, of pentacene-based OTFTs is more sensitive to environmental T compared with that of p-channel operation, i.e., hole transport.

Fig. 2 shows the comparison of the typical transfer I–V characteristics, in which V_G was swept from -20 V (+20 V) to +100 V (-50 V) and then back to -20 V (+20 V) for n-channel (p-channel)

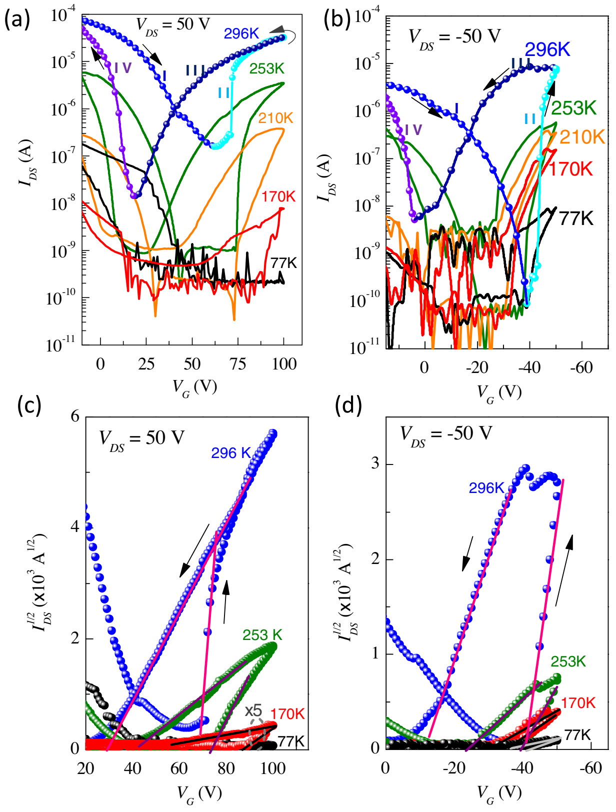

Fig. 2. Comparison of transfer characteristics, swept in both directions, of pentacene-based ambipolar OTFTs measured at different temperatures: (a) n-channel operations and (b) p-channel operations. Plot of $I_{DS}^{1/2}$ vs. V_G to calculate charge mobility: (c) n-channel operations and (d) p-channel operations. Solid lines fit the data using the standard MOSFET square-law drain current equation. Arrows indicate the sweep direction. The drain-to-source voltage (V_{DS}) is also indicated.