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Lower activation energy in organic field effect transistors with carbon nanotube contacts



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1. Introduction

Improving the charge transport in organic semiconductors (OSC) is of great interest because of their potential applications in low cost, large area flexible electronic and optoelectronic devices such as display, smart card, radio frequency identification tags and solar cells [1,2]. The OSCs are inherently disordered due to the presence of many grain boundaries and charge traps which both limit the intrinsic charge transport [3,4]. The low temperature charge transport mechanism in the OSCs is measured in field effect transistor (FET) geometry and is generally described by thermally activated hopping behavior [3-8]. The activation energy calculated from the hopping transport is considered to be a direct measure of disorder [2,4]. However, it has been pointed out that the measured activation energy of metal contacted organic FETs (OFETs) may not describe the intrinsic charge transport properties of the OSCs. This is because the charge transport in the OFETs is limited by the metal/OSC interface, and the measured activation energy is not only a result of disorder in the channel material but also the inter-

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ABSTRACT

We study temperature dependent charge transport properties of pentacene field effect transistors (FET) with carbon nanotube (CNT) electrodes. The field effect mobilities at different temperatures and gate voltages follow activated hopping behavior from which we calculate an activation energy of 48 meV for the CNT contacted pentacene FET. This value is lower than that of our control Pd contacted pentacene FET device (83 meV). The lower activation energy of the CNT contacted devices is attributed to improved CNT/pentacene interface.

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facial barrier [2,4,9,10]. When the metal electrodes are contacted with the OSCs, a barrier is formed at the metal/OSC interface due to presence of charge traps, Schottky barrier, dipole barrier and morphological discontinuity giving rise to a large contact resistance which limits the device performance [1–4]. Separating the role of the interfacial barriers and understanding the intrinsic transport properties of the OSCs turned out to be a significant challenge. Recently, it has been shown that compared to metal electrodes, carbon nanotube (CNTs) electrodes have a smaller contact resistance and form a significantly lower interfacial barrier with the OSCs [11,12]. This is attributed to the π - π interaction between the CNT and OSC [13,14]. Therefore, measurement of activation energy of the OFETs with CNT electrodes and comparing them with the OFETs fabricated with metal electrode offers an opportunity to understand the role of electrode/OSC interface and decipher the intrinsic transport properties of the OSCs.

In this paper, we present the temperature dependent electronic transport investigation of pentacene field effect transistors (FETs) with aligned array CNT electrodes. The devices were fabricated by thermally depositing pentacene thin film on prefabricated CNT source and drain electrodes, and the electronic transport properties of the devices were measured in the temperature range of 100–300 K. Our data follows thermally activated hoping behavior, from which we calculate a lowest activation energy for the CNT







contacted pentacene FET to be 48 meV at $V_g = -80$ V. This activation energy is lower than the calculated activation energy of 83 meV for our Pd contacted control device at same gate voltage, as well as other reported activation energies of the pentacene with metal electrodes. We attribute this smaller activation energy of the CNT contacted devices to improved CNT/pentacene interface compared to that of Pd/pentacene interface.

2. Material and methods

The CNT contacted pentacene devices were fabricated on a highly doped Si substrate covered with a 250 nm SiO₂ layer. The details of the CNT assembly and CNT electrode fabrication can be found in our recent publications [15,16]. In brief, single walled CNTs were aligned in a dense array between a Pd pattern of $5\,\mu m \times 25\,\mu m$ from an aqueous CNT solution (obtained from Brewer Science) via AC dielectrophoresis (DEP) (Fig. S1a) [17]. The solution contains mostly individual nanotubes with an average diameter of 1.7 nm [15]. The linear density of the CNTs in the array is $\sim 30/\mu m$, determined from the scanning electron microscopy (SEM) image. We used dense CNT arrays (~30 CNT/µm) for fabricating the electrodes because it has been found that higher density array electrodes show better FET performance than the lower density array electrode devices [18]. The typical resistance of a dense CNT array is \sim 400 Ω (Fig. S1b) corresponding to a sheet resistance of $\sim 2 \text{ k}\Omega/\Box$ [17]. In addition, the CNT arrays do not show any gatevoltage dependence, indicating the metallic behavior of the arrays [16]. Both the low sheet resistance and metallic behavior of the CNT arrays make them ideal for electrode fabrication.

After the DEP assembly, the CNT electrodes of 4 µm channel length and 25 µm channel width were fabricated through oxidative cutting of aligned CNT arrays by electron beam lithography (EBL) and precise oxygen plasma etching (Fig. S1c,d) [16,17]. For the control experiment, palladium (Pd) electrodes with geometry similar to that of the CNT electrodes were fabricated using the standard EBL process. Finally, pentacene (obtained from sigma Aldrich) was thermally deposited onto the CNT and Pd electrodes at a pressure of 2×10^{-6} mbar and a deposition rate of 0.2 Å/s. The thickness of the pentacene film is 30 nm. The atomic force microscopy (AFM) images of deposited pentacene films on both the CNT and Pd electrodes show nearly uniform morphology (Fig. S1e,f) [17]. The devices were then bonded and loaded into a cryostat. The electric transport measurements of the devices were performed using a HP 4145B semiconductor parametric analyzer.

3. Results and discussion

The current-voltage (I_d-V_d) characteristics at different gatesource voltages (V_g) at room temperature of typical CNT/pentacene and Pd/pentacene devices are shown in Fig. 1(a) and (c) respectively. The maximum output current at $V_d = -80$ V and $V_g = -80$ V of the CNT/pentacene device is \sim 90 μ A, which is significantly higher than that of the control Pd/pentacene device of 8.2 µA. Fig. 1(b) and (d) shows the transfer characteristics $(I_d - V_g)$ of the CNT/pentacene and Pd/pentacene device, respectively. From the slope of the transfer curve (dI_d/dV_g) , the linear field effect mobility of the devices at $V_d = -20$ V is calculated using standard formula: $\mu = (L/WC_iV)(dI/dV_g)$, where, C_i is the capacitance per unit area of the gate insulator (13.8 nF/cm²) [16]. The extracted current onoff ratio and mobility for the CNT/pentacene device are 1×10^5 and 0.48 cm²/Vs respectively; whereas these values are 2×10^4 and 0.04 cm²/Vs respectively for the Pd/pentacene device. The higher output current, mobility and current on-off ratio are typical for the CNT/pentacene device and is in agreement with previous reports [12,18–20]. The improved performance of the CNT con-



Fig. 1. Room temperature output characteristics (I_d – V_d) at V_g = 0 to -80 V with an interval of -10 V of (a) CNT/pentacene device and (c) Pd/pentacene device. Room temperature transfer characteristics (I_d – V_g) at V_d = -20 V for (b) CNT/pentacene and (d) Pd/pentacene devices. The channel length and width of both the CNT and Pd electrode are 4 µm and 25 µm respectively.

tacted pentacene device compared to the Pd contacted device has been attributed to the improved charge carrier injection from CNTs to pentacene due to a strong π - π bonding between the CNTs and pentacene as well as field emission characteristics of the CNTs owing to their unique one dimensional geometry [12,18,21]. Although, the work function of both the Pd and CNT are very close to highest occupied molecular orbital (HOMO) level of pentacene. when the Pd electrodes are contacted with organic semiconductors, the dipoles barriers are formed at the interface giving rise to a large effective Schottky barrier and charge carrier transport though the interface is limited by this barrier [12]. On the other hand, due to the strong π - π bonding between the CNT and organic materials, significant dipole barriers may not form between CNTs and pentacene giving rise to a better interfacial contact and lower barrier height. In addition, due to their unique one-dimensional structure, the CNTs have large electric field emission properties which can lead to a higher charge injection from CNT electrodes into the pentacene and enhance the performance of the CNT/ pentacene devices [21].

In order to better quantify the reason for improved performance of the device with CNT electrode, we performed temperature dependent electronic transport measurements of our devices. Fig. 2a and b shows the temperature dependent transfer characteristics in the temperature range of 300–100 K of the CNT/pentacene and Pd/pentacene devices, respectively. As we can see, the current of the CNT/pentacene device is much higher than that of the Pd/ pentacene device for all temperatures and the magnitude of current decreases with decreasing temperature for both devices. We calculated the mobility at different temperatures and gate voltages and plotted the mobility as a function of gate-voltage for CNT/ pentacene and Pd/pentacene devices, as shown in Fig. 2c and d respectively. These figures show that the motility is larger at higher gate-voltage, and mobility decreases with decreasing the magnitude of gate-voltage.

From Fig. 2(c) and (d), we extract mobility at different temperatures for a few fixed gate voltages. We found that our temperature dependent mobility data for both CNT contacted and Pd contacted devices can be fitted with activated hoping behavior:

$$\mu = \mu_0 \exp\left[\frac{-E_a}{k_b T}\right] \tag{1}$$

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