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Effects of gate electrode work function on electrical characteristics of pentacene-based field-effect devices

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This paper presents the effects of the work function of an indium tin oxide (ITO) gate electrode on the electrical characteristics of two pentacene-based field-effect devices—metal-insulator-semiconductor (MIS) capacitors and field-effect transistors (FETs). The ITO work function was varied by employing base and acid treatments. Flat-band voltage shifts of the MIS capacitors were found to result from the shift in the work function. The current onset and threshold voltage of the FETs were also found to be influenced by the work function. These results demonstrate the correlation of the flat-band conditions of pentacene-based field-effect devices with the gate electrode work function. © *2010 American Institute of Physics*. [doi:10.1063/1.3486180]

Field-effect devices based on organic semiconductors have high mechanical flexibility and can possibly facilitate large-area processes at low temperatures; therefore, they are extensively investigated as potential building blocks for flexible electronic devices. For example, organic capacitors with metal-insulator-semiconductor (MIS) structures are useful for data storage applications in electric memories.[1,2] Examples of more fascinating applications include the integration of organic field-effect transistors (FETs) into bendable displays[3] and disposable chips,[4] as switching and/or driving elements. Therefore, considerable research effort has been made for improving charge injection and transport characteristics of organic field-effect devices through material synthesis,[5] interface modification,[6] and process optimization.[7] However, the flat-band conditions of these devices, which can greatly affect device performance, are often neglected.

In silicon technology, interfacial states between insulator and semiconductor layers and the difference between work functions of the gate metal and semiconductor are known to have a strong influence on the flat-band conditions of field-effect devices. Similarly, it has been reported that the built-in electric field at the insulator/organic semiconductor interface can alter the flat-band conditions of organic field-effect devices.[8] However, the underlying effects of a gate electrode work function on the electrical characteristics of these devices have not yet been explained clearly. This fundamental investigation is expected to aid a physical understanding of the operation of organic field-effect devices.

In this study, the capacitance- and current-voltage characteristics of pentacene-based MIS capacitors and FETs, respectively, fabricated with indium tin oxide (ITO) gate electrodes, were investigated. On the basis of literature on organic light-emitting diodes, base and acid treatments were used to vary the ITO work function. The change in the ITO work function was confirmed by ultraviolet photoelectron spectroscopy (UPS).

Pentacene-based MIS capacitors and FETs were fabricated on glass substrates with prepatterned ITO gate electrodes (sheet resistance: ~20 Ω per square). The ITO-patterned substrates were dipped for 8 min into aqueous solutions of tetrabutylammonium hydroxide (Aldrich, 40 wt % in H_2O) or phosphoric acid (Aldrich, 1 wt % in water); next, they were dried in a vacuum oven at 175 °C.[9] Then, poly(4-vinylphenol) (PVP, Aldrich) with a cross-linking agent was spin-coated on the ITO gate-patterned substrates, which were subsequently baked at 185 °C for 1 h in a vacuum oven, yielding a 480-nm-thick cross-linked PVP (cPVP) film as a gate insulator. Next, a 60-nm-thick pentacene (Tokyo Chemical Co.) was thermally deposited through a shadow mask onto the insulator-coated substrates at a rate of 0.05 nm/s. Finally, Au was thermally evaporated on top of the pentacene layer using shadow masks to form top electrodes. The channel length and width of the FETs were 90 μm and 200 μm, respectively. The work function was measured by UPS using the He I line at 21.2 eV. Electrical characterizations of the MIS capacitors and FETs in air were carried out using an impedance analyzer (HP 4192A, Agilent Technologies) and a semiconductor analyzer (EL 421C, Elecs Co.)

A comparison of He I UPS spectra of three different ITO films is shown in Fig. 1; here, the Fermi level was calibrated using a cleaned pure Au metal surface. The base-treated and acid-treated ITO films showed considerable shifts (of +1.1 eV and −0.9 eV, respectively) from the cutoff energy of an untreated ITO film (16.4 eV). The work function of the untreated ITO film was calculated as 4.8 eV by subtracting the cutoff energy from the He excitation energy (21.2 eV); this work function is nearly consistent with previously reported values.[10] The work function values of the base-treated and acid-treated ITO films in this study were 3.7 and 5.7 eV. Nüesch *et al.* reported that changes of the order of 1 eV in the ITO work function can be induced by the adsorption of bases and acids on ITO due to the formation of ionic double layers, which in turn is initiated by the chemical adsorption of hydroxyl ions or protons, respectively.[9] Therefore, these results enable us to understand the influence of the work function of the ITO gate electrode on the electrical characteristics of pentacene-based field-effect devices.

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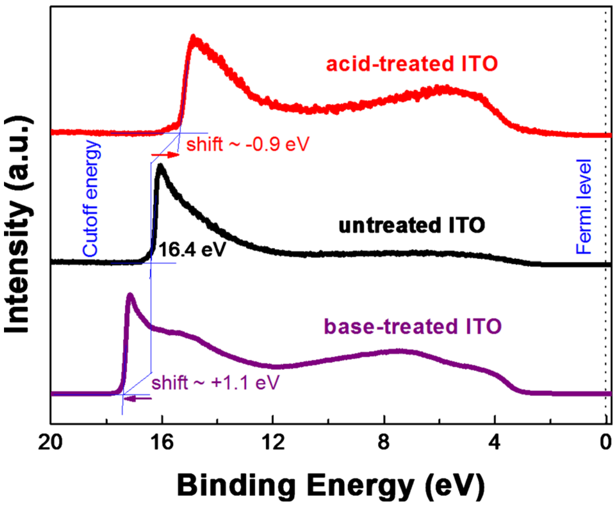

FIG. 1. (Color online) UPS spectra (He I, 21.2 eV) of ITO films without and with acidic and basic aqueous treatments.

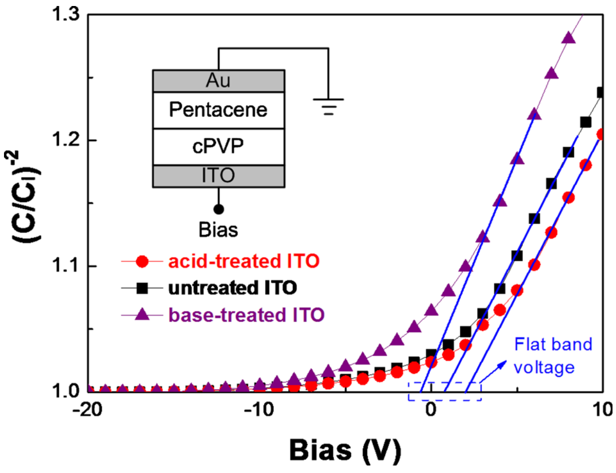

FIG. 2. (Color online) Plots of $(C/C_i)^{-2}$ vs V for MIS capacitors with different ITO electrodes. The inset shows the schematic illustration of the fabricated MIS capacitor.

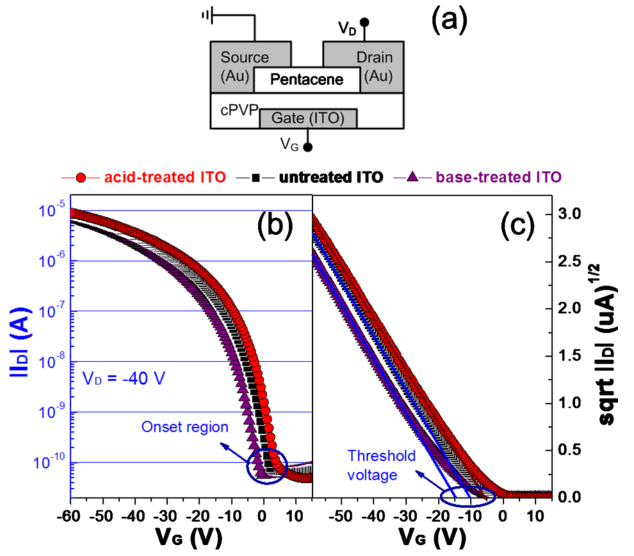

FIG. 3. (Color online) (a) Schematic of pentacene-based FET. (b) $\log_{10}|I_D|$ vs V_G and (b) sqrt$|I_D|$ vs V_G characteristics of pentacene-based FETs with different ITO gate electrodes.

The capacitance-voltage (C-V) characteristics of pentacene-based MIS capacitors were analyzed to examine the influence of the ITO work function on the flat-band voltage (V_{FB}) of these devices. Measurements were performed using a small ac signal with an amplitude of 10 mV at 100 kHz. It should be noted that the MIS capacitor is the simplest of all field-effect devices; it is a two-terminal device as shown in the inset of Fig. 2. From the plots of $(C/C_i)^{-2}$ versus V in Fig. 2, the flat-band voltage was extracted by extrapolating to the x-intercept at $(C/C_i)^{-2}=1$, where C_i is the capacitance of the cPVP layer. The obtained flat-band voltage for the MIS capacitor with the untreated ITO electrode was approximately 0.9 V, whereas those for the devices with the base-treated ITO and acid-treated ITO were -0.3 V and 1.9 V, respectively. The key features of these results are the shifts in the flat-band voltage of the MIS capacitors with the treated ITO electrodes with respect to that of the device with the untreated ITO electrode; these shifts are similar to the variations of the ITO work function values observed in Fig. 1. In principle, the flat-band voltage of an MIS capacitor is closely related to the difference in the work functions of the gate metal and semiconductor (Φ_{MS}) and to the charges (Q_{if}) at the insulator-semiconductor interface, as follows:[11]

$$V_{\mathrm{FB}} = \Phi_{\mathrm{MS}} - \frac{Q_{if}}{C_i}.$$

Since the interface charges between the cPVP layer and the pentacene layer of the fabricated MIS capacitors are similar, the flat-band voltage shifts in our results can be estimated from the difference in the measured ITO work function values by assuming a constant Fermi level of the pentacene layer. Indeed, the abovementioned flat-band voltage shifts of -1.2 and $+1.0$ V for the devices with the base-treated and acid-treated ITO electrodes correspond considerably to the differences of -1.1 and $+0.9$ V in the ITO work function values. Therefore, the flat-band voltage shift observed by us can be attributed to the change in the ITO work function, indicating the significant effect of the ITO work function on the flat-band conditions of a pentacene-based MIS capacitor.

As the MIS capacitor is a fundamental building block in complex MIS device structures, the results for MIS capacitors can be extended to characteristic investigations of FETs shown in Fig. 3(a). The transfer characteristics of the fabricated pentacene-based FETs were measured by applying a drain voltage (V_D) of -40 V, while the gate voltage (V_G) was varied from +15 to -60 V in sweep steps of -0.5 V. Figure 3(b) shows that the shifts in the onset characteristics of the FETs are reminiscent of shifts of the flat-band voltages of MIS capacitors. These different onset characteristics of the FETs are thus indicative of different flat-band conditions at different work functions of ITO gate electrodes, as described in the literature.[12] It was also observed that the early onset of the FET with the acid-treated ITO gate electrode led to an increase in the drain current (I_D), unlike the case of the device with the untreated ITO gate electrode. The lowest drain current was obtained by the latest onset of the FET with the base-treated ITO electrode. The threshold voltages of the fabricated FETs were extracted from Fig. 3(c), in which changes in the threshold voltage induced by treatments of the ITO gate electrode are quite apparent: the extracted threshold voltages for the acid-treated ITO, untreated ITO, and base-treated ITO FETs are -5.3 V, -10.6 V, and -14.6 V, respectively. It is noteworthy that the threshold voltage shifts in the FETs are more pronounced than the flat-band voltage shifts in the MIS capacitors. This is probably attributed to the fact that the threshold voltage is that gate bias beyond the flat band at which a channel charge sheet is induced; the threshold voltage is then given by the sum of voltages across the semiconductor and insulator layer. Further quantitative analyses are required to elucidate the relation between the threshold voltage of an organic FET and a gate work function. Meanwhile, the field-effect mobilities in the saturation region were almost identical ($\sim 0.32\ \mathrm{cm}^2/\mathrm{V\ s}$), irrespective of the flat-band conditions of the fabricated FETs. Accordingly, these results indicate that the modification of a gate work function mainly results in a shift in the onset voltage and the consequent shift in the threshold voltage in the pentacene-based FET.

In summary, we demonstrated the correlation between the flat-band conditions of pentacene-based field-effect devices and the ITO work function, whose value could be varied by subjecting the ITO gate electrodes to aqueous treatments using base and acid solutions. For MIS capacitors, the shift in the flat-band voltage agreed well with the difference in the ITO work function values, indicating that the shift was caused mainly by the change in the work function of the ITO electrode. The shift in the onset characteristics of the FETs due to the change in the ITO work function was found to cause the pronounced shift in the threshold voltage. We believe that the results of our study can be applied to the common practice of controlling the performance of organic field-effect devices.

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