Bio-organic-semiconductor-field-effect-transistor based on deoxyribonucleic acid gate dielectric

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Organic-based field-effect transistors (OFETs) utilize organic semiconductor materials with low electron mobilities and organic gate oxide materials with low dielectric constants. These have rendered devices with slow operating speeds and high operating voltages, compared with their inorganic silicon-based counter parts. Using a deoxyribonucleic acid (DNA)-based biopolymer, derived from salmon milt and roe sac waste by-products, for the gate dielectric region, we have fabricated an OFET device that exhibits very promising current-voltage characteristics compared with using other organic-based dielectrics. With minimal optimization, using a thin film of DNA-based biopolymer as the gate insulator and pentacene as the semiconductor, we have demonstrated a bio-organic-FET, or BiOFET, in which the current was modulated over three orders of magnitude using gate voltages less than 10 V. © *2006 American Institute of Physics.*
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I. INTRODUCTION

Organic-based electronics has evolved into a separate research field producing thin-film devices such as displays,[1–3] sensors,[4] electronic labels,[5,6] ring oscillators,[7,8] and complementary integrated circuits. To fabricate a complementary integrated circuit, *p*-type materials and *n*-type materials that possess acceptable mobilities are required.[9,10] We have recently demonstrated an *n*-channel organic-based field-effect transistor, or OFET, using hot wall epitaxially grown, C_{60} thin films with electron mobilities as high as 1 cm^2/V s,[11] and these mobilities continue to increase upon optimization of the dielectric gate region.[12]

For the present, where might organic-based FETs, with low mobilites compared to silicon (Si), find a niche? It turns out that the operational voltage of the device can also be influenced by the dielectric gate region.[13] Polymer-based insulators have increasingly been investigated for potential use as a gate dielectric.[11–15] These polymeric materials are solution processable, are thermally stable with negligible thermal expansion, and render uniform films on both transparent glass and flexible plastic substrates. Due to their high optical transparency, they are suitable for optoelectronic devices, such as photoresponsive OFETs, and they posses relatively high dielectric constants. The interface between the organic semiconductor and the organic dielectric is critical for the charge carrier transport, as well as the hysteresis.[16] This hysteresis can be observed in the transfer characteristics (source-drain current I_{ds} as a function of the gate voltage V_{gs}) for most polymeric insulators used as a gate dielectric. It is presumed to be due to charge trapping and detrapping at the organic insulator/organic semiconductor interface and reduces at low temperatures.[17]

For metal insulator semiconductor devices the threshold or turn-on voltage is given by[18]

$$V_t = - (qn_0 d_s/C_i) + V_{fb},$$

where V_{fb} is the flat band potential (which accounts for the work function difference between the semiconductor and the gate electrode without considering fixed charges at the dielectric and at the interface), d_s is the thickness of the semiconductor, q is the elementary charge, and C_i is the capacitance of the gate insulator. The capacitance of the gate insulator, C_i is a ratio of the permittivity of the insulator material ε_i and the thickness of the gate region d_g. Increasing ε_i or decreasing d_g will reduce V_t.

Recently a pentacene-based OFET with a self-assembled polymer monolayer ultrathin gate insulator has been demonstrated.[19] There are also approaches such as the use of solid electrolytes as gate dielectrics with large capacitive coupling.[20]

We present here a deoxyribonucleic acid (DNA)-based biopolymer material which shows promise as an excellent gate insulator for OFETs.

II. EXPERIMENT

Spin cast films of DNA-based biopolymer, complexed with a cationic surfactant from butanol solution, forms a self-assembled structure that is optically transparent across a broad range of wavelengths and is easily processable.[21–24]

The DNA used for this research was purified DNA provided by the Chitose Institute of Science and Technology (CIST).[21,22] It was marine based, that was first isolated from

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FIG. 1. Freeze dried, purified salmon DNA.

frozen salmon milt and roe sacs through a homogenization process. It then went through an enzymatic treatment to degrade the proteins by protease. Proteins were then removed by controlling the pH level to 7.5. The DNA underwent a carbon treatment for decolorization, was filtered, and precipitated by adding acetone. The purified DNA was finally filtered from the acetone and freeze dried (see Fig. 1). The molecular weight of the purified DNA measured M_W =8 000 000 Daltons (Da), using gel phase electrophoresis. The purity measured assay=96% and the protein content measured is 2%.

We found, however, that the purified DNA was soluble only in water, so it is not compatible with typical fabrication processes used for polymer-based devices. We also observed many particulates in the DNA films. Therefore, we performed additional processing to render DNA more suitable for device fabrication with better film quality. This processing was accomplished by precipitating the purified DNA in

water with a cationic surfactant complex, hexadecyltrimethy-lammmonium chloride (CTMA), by an ion exchange reaction.[21–24]

The resulting DNA-lipid complex was water insoluble and more mechanically stable due to the long alkyl chain of the CTMA. Adding the CTMA complex, DNA-CTMA could now be dissolved using solvents more compatible with device fabrication, such as ethanol, methanol, butanol, or a chloroform/alcohol blend. When dissolved in the organic solvent, the DNA-CTMA was passed through a 0.2 μm pore size syringe filter to remove the large particulates.[23,24]

Preliminary investigations have suggested that DNA-CTMA is a promising optical waveguide quality material with tunable resistivities several orders of magnitude lower than other polymers with comparable optical loss.[23–29] In addition, it is derived from waste and is, hence, abundant and affordable and is also a green material. Being derived from biowaste, it is a non-fossil-fuel-based polymer.

The solvent of choice for this research was butanol. Its high boiling point produced very uniform spin deposited thin films. Various weight ratios of DNA-CTMA in butanol were prepared to produce various film thicknesses. Thin films of 2.7 and 0.2 μm were obtained from 10% and 2% solutions, respectively, using a spin speed rate of 1500 rpm, without ramping.

First we fabricated a metal-insulator-metal (MIM) device structure for the characterization of the dielectric properties of DNA-based biopolymer film. For this measurement, the DNA-based biopolymer was sandwiched between a chromium/gold (Cr/Au) bottom electrode, which was evaporated on top of a quartz substrate and a Au top electrode.

We then fabricated a bio-organic field effect transistor (BiOFET) device structure using a top contact geometry, evaporating a pentacene organic semiconductor film on top of a layer of DNA-CTMA, serving as the gate dielectric,
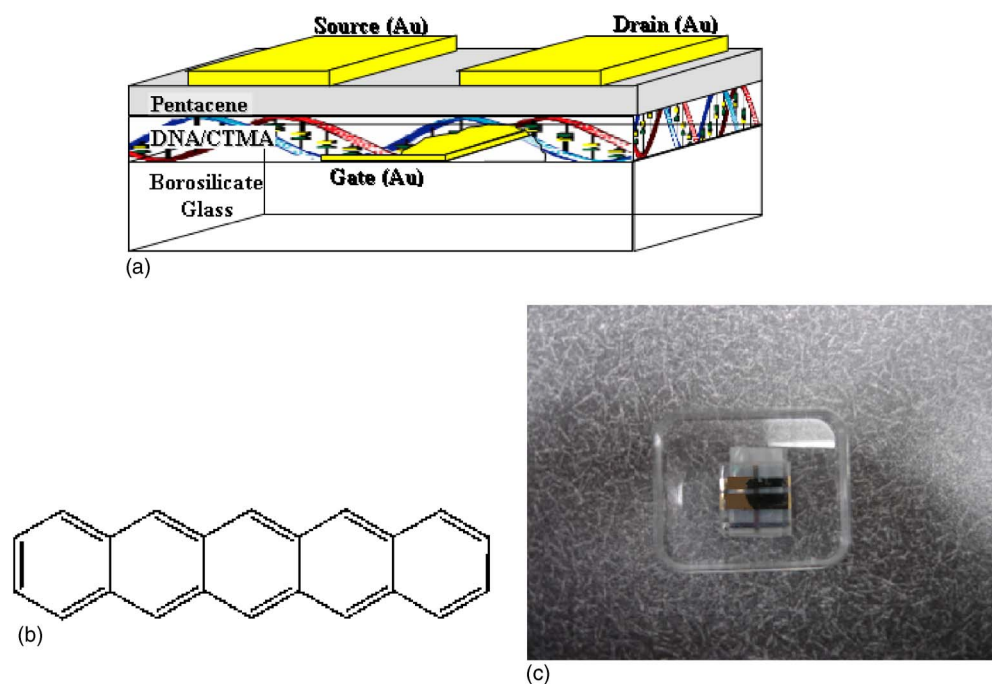

FIG. 2. (a) Schematic of the top contact BiOFET device. (b) Chemical structure of pentacene. (c) Photograph of the actual BiOFET device.
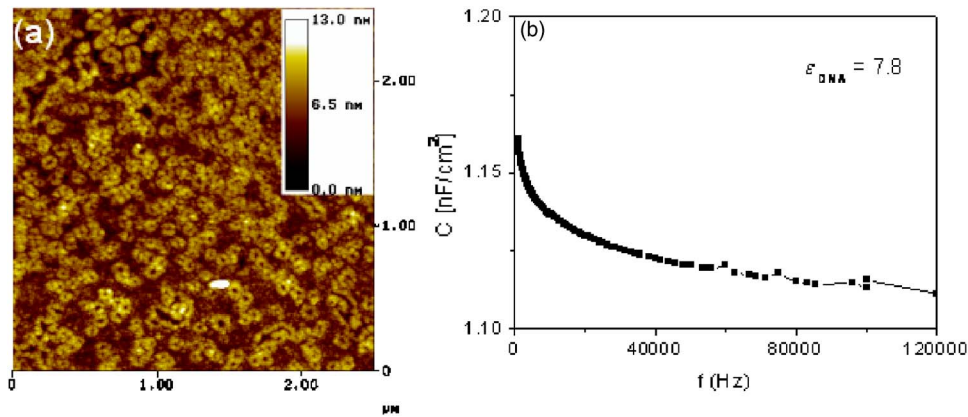
FIG. 3. (a) AFM tapping mode image of a DNA-CTMA thin film on quartz. (b) Capacitance-voltage (C-V) characteristics of the MIM device.

with the intention of reducing the operating gate voltage of the device. The BiOFET is schematically depicted in Fig. 2(a), and the chemical structure for pentacene is shown in Fig. 2(b). A photograph of the actual BiOFET device is shown in Fig. 2(c).

We started with a prepatterned Cr/Au bottom gate electrode, deposited on a glass/quartz plate. A 200 nm thick DNA-based biopolymer film was then spin deposited on top of the bottom gate electrode. This was followed by the evaporation of 50 nm thick layer of pentacene, at a rate of 0.2 Å/s at room temperature, at a base vacuum of 10^{-6} mbar. Finally Au top source and drain electrodes were deposited to complete the device.

An Agilent model E5273A, two source-measurement unit instrument, was employed for the steady state current-voltage (I–V) measurements. We then confirmed all electrical measurements using both a Keithley model 2400 and a Keithley model 236 source-measurement unit instrument. All measurements were performed at a scan rate of 0.2 V/s, unless otherwise stated. For the quasistatic capacitance-voltage (C-V) measurements, we used an HP model 4248A, precision *LCR* meter. The surface morphology and thickness of the dielectric and semiconductor thin films were measured in ambient condition with a Digital Instrument Dimension model 3100 atomic force microscope (AFM).

III. RESULTS

Our initial experiments dealt with the study of the thin film morphology of the DNA-based biopolymer. Figure 3(a) is an AFM image of the morphology of a 200 nm DNA-CTMA film, spin deposited on top of a quartz substrate. As can be seen, this produced a surface roughness of 10 nm. Features of self-organized structures can also be observed. Figure 3(b) is a plot of the dielectric response (C-V) of the DNA-CTMA film that was sandwiched between the two electrode (MIM) devices. Figure 3(b) indicates that DNA-CTMA has a stable capacitance for a wide range of frequencies with a dc capacitance of $\sim$1.15 nF/cm^2.

From the measurement of capacitance per unit area, we obtained a dielectric constant of $\varepsilon_{DNA}=7.8$. This is in good agreement with earlier dielectric data taken for DNA-CTMA.[24]

Typical output characteristics of the BiOFET device we fabricated are ploted in Fig. 4(a). As can be seen, the

BiOFET, with a 200 nm thick film of DNA-CTMA and a 50 nm thick pentacene semiconductor layer, was able to modulate the drain current over three orders of magnitude using gate voltages of less than 10 V. Figure 4(b) is a plot of the transfer characteristics for the linear and saturated drain voltage. Pronounced hysteresis in the transfer characteristics can be observed in these devices, as indicated by the direction of arrows in Fig. 4(b).
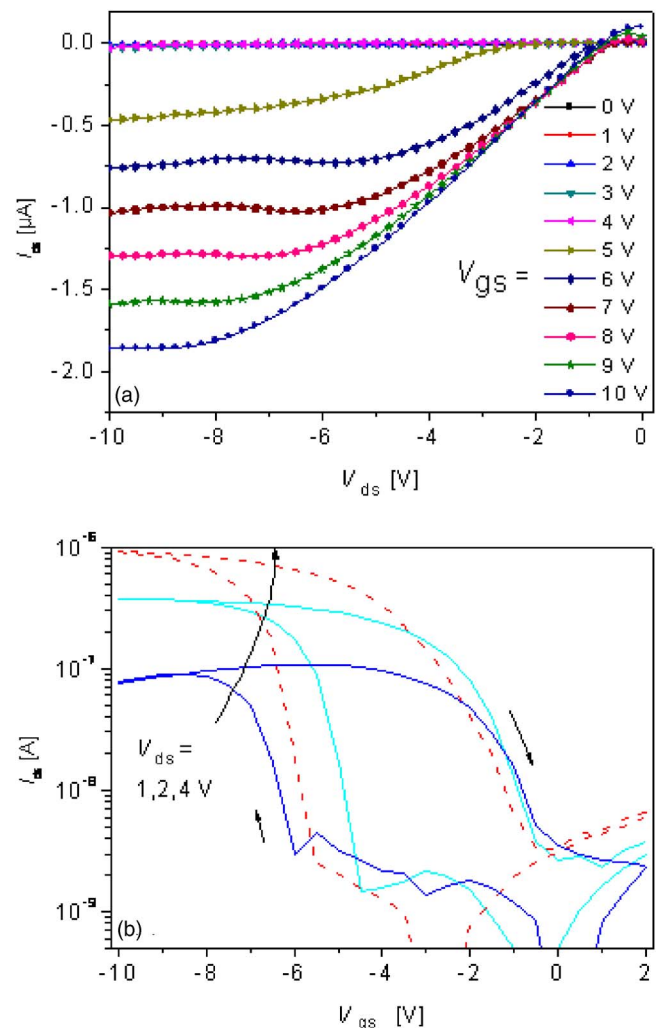

FIG. 4. (a) BiOFET output characteristics; drain current I_{ds} vs drain voltage V_{ds} for different gate voltages V_{gs}. (b) Transfer characteristics; I_{ds} vs V_{gs} for drain voltages, $V_{ds}=1$, 2, and 4 V.

The channel length ($L = \sim 20~\mu$m) and width ($W = \sim 1.5$ mm) were used, together with the capacitance ($C_i = 1.15$nF/cm^2), to extract a saturated regime mobility of 0.05 cm^2/V s in our device, using the standard transistor equation,[18] in which there is no correction for contact resistances.[30–32] We determined the linear mobility to be 10^{-2} cm^2/V s.

As shown in the transfer characteristics, a sizable hysteresis exists, presumably due to the motion of ionic charges present in the DNA-based biopolymer at the gate insulator/organic semiconductor interface.

IV. CONCLUSION

In this work we have demonstrated a promising, low gate voltage, BiOFET using a DNA-based biopolymer for the gate dielectric region. This suggests that the potential for rendering OFETS is more competitive for low frequency applications. This would prove advantageous from a low cost, high volume perspective, since this material is inexpensive and simply spin deposited without any special processing required.

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