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Flexible pentacene thin film transistors as DNA hybridization sensor

Jung-Min Kim[1], Sandeep Kumar Jha[1], Rohit Chand[1], Dong-Hoon Lee[1], and Yong-Sang Kim[1,2]

[1]Department of Nano Science and Engineering, Myongji University, Gyeonggi 449-728, Korea
[2]Department of Electrical Engineering, Myongji University, Gyeonggi 449-728, Korea
E-mail: kys@mju.ac.kr

Abstract—**A DNA hybridization sensor using pentacene thin film transistors (TFTs) is an excellent candidate for disposable sensor applications due to their low-cost fabrication process and fast detection. We fabricated pentacene TFTs on flexible substrate for the sensing of DNA hybridization. The 100 mer ss-DNA (poly A/poly T) or 100 bp ds-DNA (poly A/poly T hybrid) are deposited from a solution on pentacene layer. The electrical characteristics of devices were studied as a function of DNA immobilization, single- and double-strand DNA and DNA concentrations. The DNA molecules were immobilized directly on the surface of the pentacene, thereby producing a dramatic change in the electrical properties of the devices. Based on these results, we propose that a "label-free" detection technique for DNA hybridization is possible through direct measurement of electrical properties by the immobilization of DNA on pentacene TFTs.**

Keywords-DNA hybridization sensor; pentacene thin film transistor ; disposable sensor; label-free

I. INTRODUCTION

Completion of Human genome project has necessitated rapid development in the field of nucleic acid diagnostics. The detection and quantification of DNA hybridization is also of great importance in many applications, such as medical diagnostics, forensic science, genotyping, and pathogen detection [1-5]. Traditional methods for detection of DNA mainly focus on radio labeled system or optical detection using fluorochrome tagged oligonucleotides. These detection techniques have limitations due to the complications in sample preparation as well as the necessary usage of complex and expensive optical systems, along with health risk. Compared with these techniques, label-free electronic methods promise to offer sensitivity, selectivity, and low cost for the detection of DNA hybridization. Recently, DNA hybridization sensors, using the "label-free" method, have been studied with much interest, including electrochemical detection [6], carbon nanotube network field-effect transistors [7], atomic force microscopy (AFM) [8], surface plasmon resonance (SPR) [9], genetic field effect transistor (FET) [10], and microcantilevers [11]. Among these methods, a DNA hybridization sensor using organic thin film transistors (OTFTs) is an excellent candidate for the application as disposable sensors, due to their potentially low-cost fabrication process and quicker response time [12]. Moreover, due to their biocompatibility and flexibility, an organic semiconductor material offers great opportunity for integration with biological systems.

For these reasons, in the present work, we fabricated organic TFTs using pentacene on flexible substrates as a biosensor for DNA hybridization. Pentacene was the choice for the organic semiconductor material due to its excellent electrical properties and ease in immobilization of DNA on it. The target DNA was immobilized on the pentacene surface through physical adsorption without requiring any binding agents thereby reducing the use of reagents and fabrication cost. The adsorbed DNA on the OTFT attracts holes from channel region, causing a change in the resultant channel current during analysis. The magnitude of this change was significantly different for single stranded DNA (ss-DNA) and double stranded DNA (ds-DNA), thereby allowing us to sense the DNA hybridization.

II. EXPERIMENTAL

A. *Facrication of pentacene thin film transistor*

The top-contact pentacene TFTs were fabricated on a PES substrate. Indium tin oxide (ITO) was used for devising gate electrode. The gate insulator composed of poly (4-vinylphenol) (PVP) was deposited over the ITO gate electrode to a thickness of 480 nm by spin coating and was baked at 200 ^{o}C for 1 hour. The PVP solution was prepared by dissolving PVP (10 wt% of solvent) and methylated poly (melamine-co-formaldehyde) (5 wt% of solvent) as a cross-linking agent in propylene glycol methyl ether acetate as solvent. The pentacene active layer was patterned through the shadow mask by thermal evaporation at a rate of 0.1 Å/s to a thickness of about 70 nm at a high vacuum ($< 5x10^{-6}$ torr). The source and drain electrodes were made up of Au layer of 100 nm thickness, which were deposited by thermal evaporation using a shadow mask. The pentacene TFTs obtained had a channel length (L) and width (W) of 100 and 1000 μm, respectively.

B. *DNA immobilization and hybridization*

As shown in Fig. 1, ss-DNA was first immobilized by pipetting a 1-μℓ drop of deionized (D.I) water containing the DNA onto the pentacene TFTs channel and then air-drying for 60 min. Subsequently, 1 mℓ of D.I water was dropped slowly through slant onto the pentacene TFT channel for thorough washing of surface. The devices were then air-dried for 60 min before being characterized at room temperature in ambient air. Further, in order to validate DNA hybridization on same

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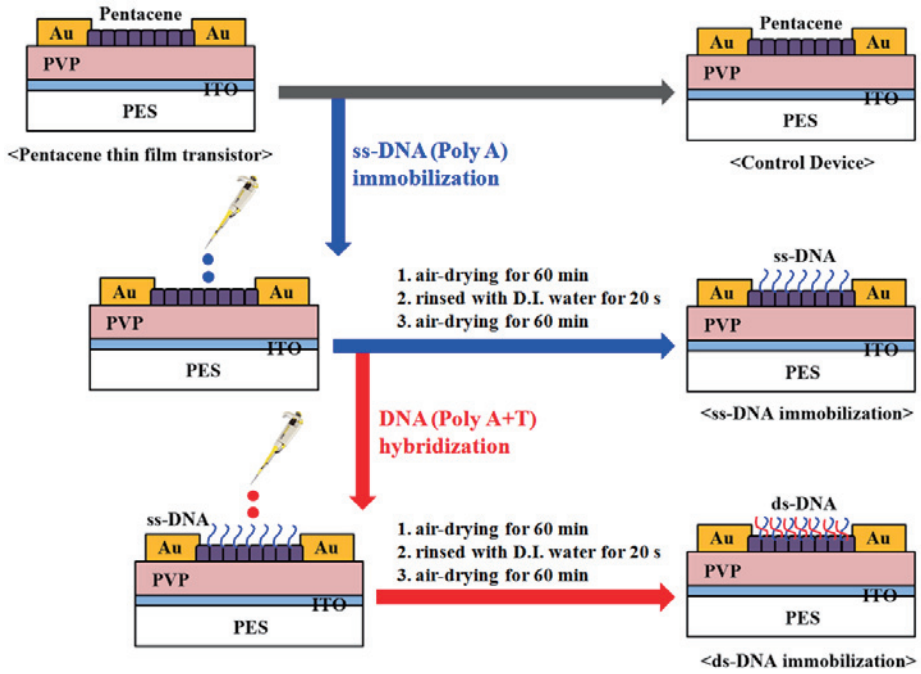

Figure 1. Scheme showing the DNA immobilization and hybridization in the OTFTs.

substrate, pentacene TFTs with immobilized ss-DNA (100 mer- Poly A) were used for immobilization of complementary ss-DNA (100 mer- Poly T). The devices were air-dried for 60 min, washed again with D.I water as before, and air-dried for 60 min before being characterized by Keithley 236 meter. Similarly, different length and concentrations of DNA oligos were used to determine their effect on device characteristics.

The immobilization of ss-DNA on the pentacene surface and subsequent hybridization of complementary strand was confirmed using fluorescent labeling of ds-DNA during hybridization process (Fig. 2). The fluorescent intercalator ethidium bromide (EtBr) was used for this purpose. The fluorescent images of labeled DNA or control devices were obtained using a fluorescence microscope (Olympus BX50, Japan).

C. Sensing mechanism

The devices were characterized at room temperature in ambient air using a Keithley 236 meter interfaced with a computer using a GPIB-software interface.

The DNA molecules were immobilized on the hydrophobic pentacene surface by physical adsorption through their hydrophobic interactions. These molecules also have negatively charged phosphate groups on their backbone, which profoundly affects the electrical performance of the pentacene TFTs. When the DNA molecules were immobilized on the pentacene surface, negative charge of DNA molecules attract holes from the channel region, thereby increasing the scattering of holes, while holes moves down from source to drain electrode. In this scenario, we expected the mean time between collisions or scattering (τ_{cp}) to decreases, thus decreasing field-effect mobility by equation

$$\mu_{FET} = \frac{v_{dp}}{E} = \frac{e\tau_{cp}}{m_p^*} \quad (1)$$

where, v_{dp} is the average drift velocity of the holes, E is electric field, e is the magnitude of the electronic charge and m^*_p is the effective mass of the hole.

The performance of the pentacene TFT devices was measured in terms of their output and transfer characteristics. In order to find the output characteristics of devices, the channel current (I_{DS}) was measured as a function of the drain-source voltage (V_{DS}) under a constant gate voltage (V_{GS}). Evaluation of transfer characteristics was carried by measuring the I_{DS} between the source and drain as a function of the V_{GS} under a constant V_{DS}. One of the important parameters of OTFT was the field-effect mobility of carriers in its channel region. The field-effect mobility of hole (μ_{FET}) was determined using the saturation drain current ($I_{DS,sat}$) which is given by

$$I_{DS,sat} = \frac{WC_{PVP}\mu_{FET}}{2L}(V_{GS} - V_{TH})^2 \quad (2)$$

where W is the width of the channel, L is the length of the channel, C_{PVP} is the capacitance per unit area of the PVP gate insulator, V_{GS} is the gate voltage and V_{TH} is threshold voltage.

Figure 2. Florescence image of pentacene TFT after hybridization of DNA

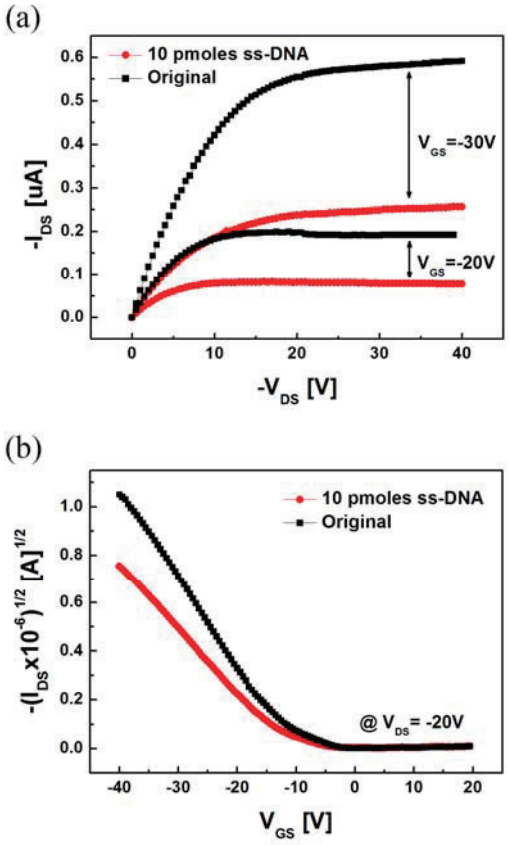

Figure 3. Performance of the pentacene TFTs with DNA immobilized on pentacene surface for 60 min: (a) output characteristics of two pentacene TFTs (original, ss-DNA), (b) transfer characteristics of two pentacene TFTs (original, ss-DNA).

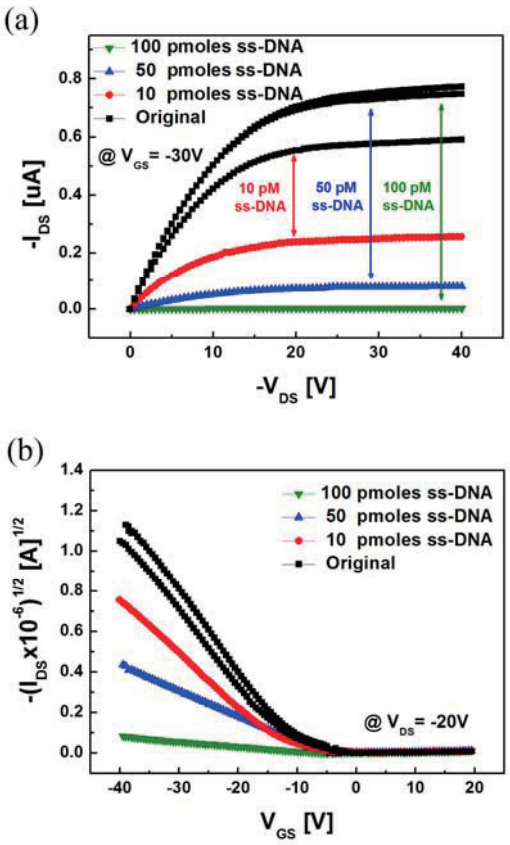

Figure 4. (a) Output characteristics and (b) transfer characteristics of a pentacene TFTs with 10, 50 and 100 pmoles DNA immobilized on pentacene surface.

III. Result and Discussion

The DNA molecules were immobilized on the hydrophobic pentacene surface by physical adsorption through their hydrophobic interactions. These molecules also have negatively charged phosphate groups on their backbone, which profoundly affects the electrical performance of the pentacene TFTs. This can be regarded as the sole reason for using the pentacene TFTs for fabrication of DNA hybridization sensors.

The influence of the immobilized DNA on pentacene surfaces was studied by fabricating the pentacene TFTs with 10 pmoles ss-DNA for 60 min (immobilization time). Fig. 3(a) shows the I_{DS} as a function of V_{DS} under different V_{GS} (the output characteristic), whereas, Fig. 3(b) shows I_{DS} as a function of the V_{GS} measured at a constant V_{DS} (the transfer characteristic). At the same applied V_{GS}, the original pentacene TFTs (without ss-DNA immobilization) showed a higher I_{DS} than the pentacene TFTs with the immobilized ss-DNA. The DNA molecules influence the field-effect mobility in the pentacene TFTs as given by equation (2). Original pentacene TFTs (without ss-DNA immobilization) have a field-effect mobility of μ_{FET}= 0.038 cm^2/Vs. Conversely, the pentacene TFTs with ss-DNA immobilization have a field-effect mobility of μ_{FET}= 0.019 cm^2/Vs. The electrical characteristic of the pentacene TFTs with the ss-DNA immobilization gives a lower I_{DS} and field-effect mobility due to the ss-DNA immobilization. Such dramatic changes of electrical properties were well expected, since the phosphate group on the DNA backbone imparts a net negative charge in the DNA molecules, which attracts holes from the channel region, thereby decreasing the I_{DS} and field-effect mobility.

In order to derive any correlation between the device response and the extent of the DNA concentration, the output and transfer characteristics of the devices were also measured by varying the concentration of the ss-DNA on the pentacene surface (Fig. 4). As DNA concentration was increased, the I_{DS} value decreased and so a decrease in the field-effect mobility from 0.038 cm^2/Vs to 0.019, 0.004 and 0.0001 cm^2/Vs for 10, 50 and 100 pmoles ss-DNA was observed respectively. Such a reduction in I_{DS} and field-effect mobility was due to the increase in the concentration of immobilized DNA on the pentacene's surface, which collectively attracts more holes from the channel region. This result indicates the possibility of dynamic response from devices having low concentration of DNA immobilized on pentacene.

Additionally, we also found a dramatic difference in the I_{DS} and pattern in field-effect mobility upon exposure to either 10 pmoles ss-DNA or ds-DNA. Fig. 5 shows the difference in the

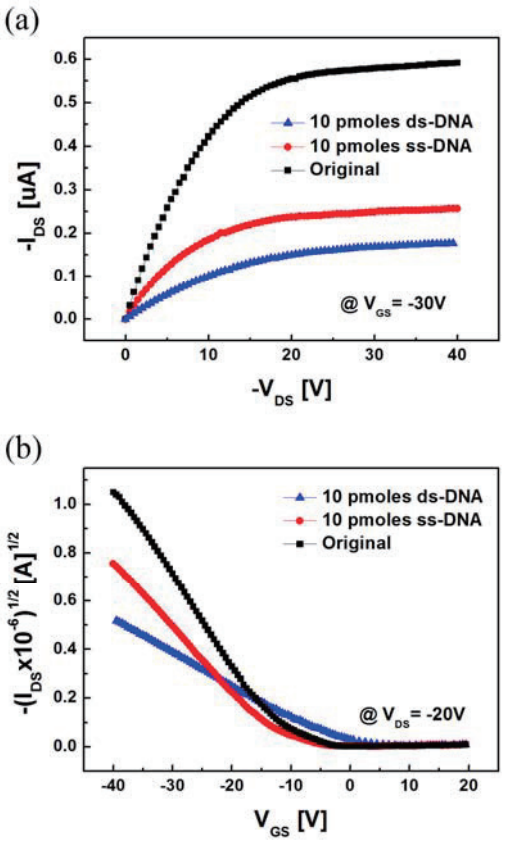

Figure 5. Performance of the pentacene TFTs with DNA immobilized on pentacene surface for 30 min. (a) output characteristics of three pentacene TFTs (original, ss-DNA, ds-DNA); (b) transfer characteristics of three pentacene TFTs (original, ss-DNA, ds-DNA).

sensor output and the transfer characteristics from the original pentacene TFTs (without ss-DNA) compared to the pentacene TFTs with immobilized ss-DNA or ds-DNA. The ds-DNA caused a higher ΔI_{DS} compared to that caused by the ss-DNA, since the ds-DNA carry more net negative charge. The field-effect mobility reduced to 0.019 and 0.005 cm^2/Vs for ss-DNA and ds-DNA, respectively. This enables the direct electrical detection of the DNA hybridization through the measurement of I_{DS} for the pentacene TFTs. The net difference in the I_{DS} (ΔI_{DS}) on the pentacene TFTs due to the single or double stranded DNA was the basis for the analysis of the DNA hybridization.

IV. Conclusion

The single and double stranded DNA was immobilized on the surface of the pentacene layer, producing a change in the performance of the pentacene TFTs. It is attributable to the negative charges on the DNA molecules having the ability to attract holes from the channel region. The electrical characteristic of the pentacene TFTs with the ds-DNA immobilization gives a lower current output and field-effect mobility since the ds-DNA carry more net negative charge. Therefore, we propose, in conclusion, that a "label-free" detection technique for DNA hybridization with high sensitivity and selectivity is possible to realize portable and disposable DNA sensor having applications in molecular biology laboratories, medical diagnostics, forensic investigations, genotyping, etc.

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