

Bio-organic field effect transistors based on crosslinked deoxyribonucleic acid (DNA) gate dielectric

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Using DNA-based biopolymers purified from salmon waste, as an insulating layer, bio-organic field effect transistor (BioFET) devices were fabricated. Such devices exhibit current-voltage characteristics with low operational voltages as compared with using other organic dielectrics. The observed hysteresis in transfer characteristics of such BioFETs can be reduced using a crosslinking process. Such crosslinked DNA complex is used as a gate dielectric in n-type C₆₀ as well as p-type α -sexithiophene (T6) based BioFETs. © 2009 American Institute of Physics.

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Biopolymer-based materials possess unusual properties that are not easily replicated in conventional organic or inorganic materials. Additionally, biopolymers derived from DNA are a renewable resource and inherently biodegradable. Among natural biodegradable materials, the use of DNA-based biopolymers in organic electronic and photonic devices is rapidly becoming an area of interest in the photonics community.^{1–5} Organic field effect transistors, organic light emitting diodes, and nonlinear optical electro-optic modulators have been fabricated from DNA-based biopolymers. Such devices have demonstrated high performance that exceeds that of state-of-the-art devices made with currently available organic based materials.^{4–11}

However, the observed large hysteresis in BioFETs using DNA-hexadecyltrimethylammonium chloride (CTMA) complexes have been discussed as a performance limiting issue.⁵ Therefore, we analyzed the origin of this hysteresis and an ionic charge migration has been one of the main responsible factors for this hysteresis.¹² In this report, we present the idea of crosslinking the whole composite polymer for inhibiting the ion migration to improve the BioFET transfer characteristics.

The DNA used for this research was purified from waste materials of the salmon fishing industry through an enzyme isolation process by the Chitose Institute for Science and Technology.^{13,14} The molecular weight of the purified DNA measured $M_w=8000\ 000$ Daltons (Da), using gel phase electrophoresis. The purity measured assay=96% and the protein content measured is 2%.

However, the DNA is soluble only in aqueous solutions, the resulting films are too water sensitive and have insufficient mechanical strength, so it is not compatible with typical fabrication processes used for polymer-based optoelectronic devices. Also, many particulates are observed in the DNA films. Therefore, additional processing steps are performed

to render DNA films suitable for optoelectronic device fabrication with high film quality. This processing is accomplished by precipitating the purified DNA in water with a cationic surfactant complex CTMA, by an ion exchange reaction.^{13–16}

The resulting DNA-lipid complex became water insoluble and more soluble in organic solvents because of the alkyl chains of the CTMA. Adding the CTMA complex, DNA-CTMA compound was soluble in many of the alcohols including methanol, isopropanol, butanol, and alcohol/chloroform blend. When dissolved in the organic solvent, the DNA-CTMA was passed through a 0.2 μm filter to remove any large particulates.¹⁵ To spin cast a thin film for photonics applications, butanol is the solvent of choice because of its slow evaporation due to a high boiling point (116–118 °C), which ensures a smooth uniform film during the spin-coating process. Various weight ratios of DNA-CTMA in butanol were prepared to produce various film thicknesses. Thin films of 0.5 μm were obtained from 5% solution, using a spin speed rate of 1500 rpm, without ramping.

The resulting DNA-CTMA films, while of high optical quality, are soft and scratch easily. This makes them incom-

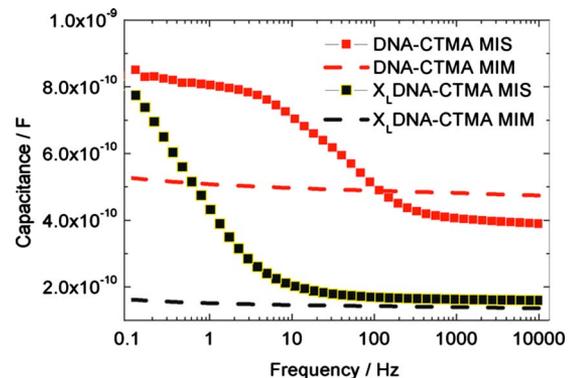


FIG. 1. (Color online) Room temperature capacitance-frequency characteristics of the MIM and MIS devices.

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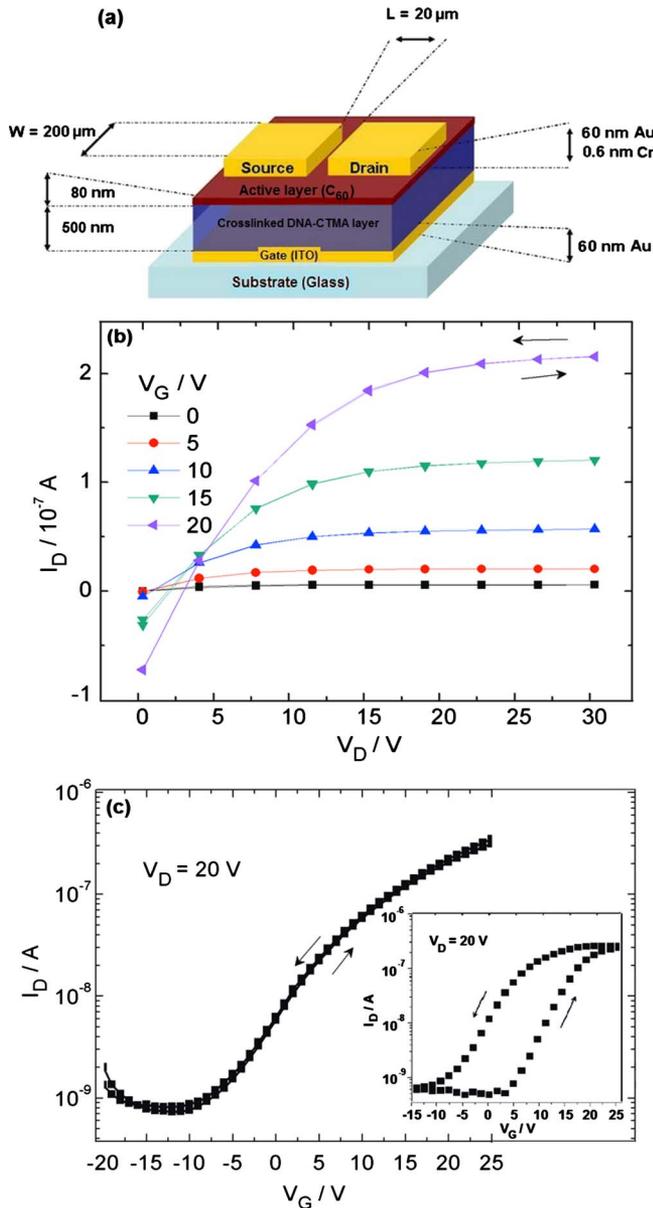


FIG. 2. (Color online) (a) Diagram of the n-channel BiOFET device geometry. (b) Output characteristics of BiOFET with C_{60} as a semiconductor with crosslinked DNA-CTMA film as a dielectric at different gate voltages. (c) Transfer characteristics at $V_{DS} = 20 \text{ V}$. Inset: Transfer characteristics of BiOFET with C_{60} as a semiconductor with non-crosslinked DNA-CTMA film as a dielectric.

patible with more aggressive processing techniques, which are often required to fabricate a photonic device. One solution to this problem is to crosslink the DNA-CTMA films. A crosslinking agent, poly(phenylisocyanate)-co-formaldehyde (PPIF), which has one crosslinking site located at every phenylene unit, is used for improving the uniformity of the crosslinking reaction between the DNA biopolymer chains and cationic surfactant complex CTMA. The resulting crosslinked DNA-CTMA films are significantly harder than the noncrosslinked films and are also resistant to the solvents used to process the films.

To prepare a crosslinked DNA-CTMA film, the DNA-CTMA is dissolved in butanol at a concentration of 0.51 M. An amount of 81.2 mol % PPIF, with respect to DNA-CTMA, is dissolved separately in butanol at a concentration of 0.51 M. Both the DNA-CTMA in butanol and PPIF in

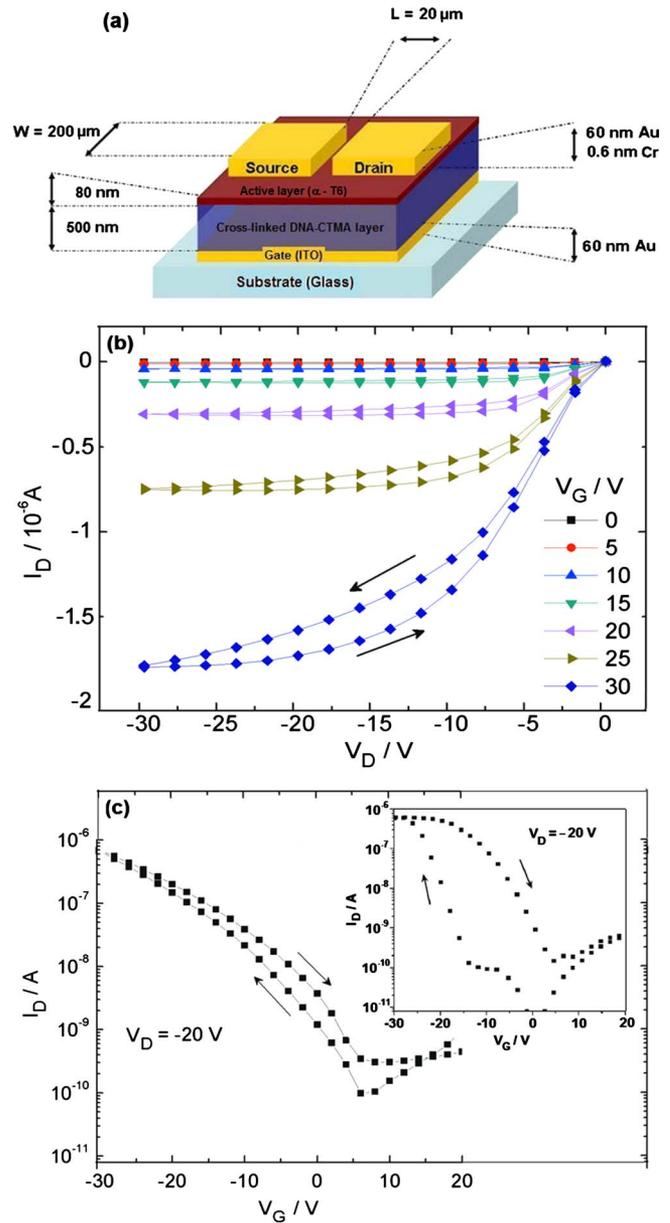


FIG. 3. (Color online) (a) Diagram of the p-channel BiOFET device geometry. (b) Output characteristics of BiOFET with $\alpha\text{-T6}$ as a semiconductor with crosslinked DNA-CTMA film as a dielectric at different gate voltages. (c) Transfer characteristics at $V_{DS} = -20 \text{ V}$. Inset: Transfer characteristics of BiOFET with $\alpha\text{-T6}$ as a semiconductor with non-crosslinked DNA-CTMA film as a dielectric.

butanol solutions are mixed in a 60°C oven for 1 to 2 h. Once fully dissolved, the DNA-CTMA in butanol solution is added to the PPIF in butanol solution and the resulting solution is mixed for an additional 2 h in the 60°C oven. The DNA-CTMA-PPIF solution is then filtered through a $0.2 \mu\text{m}$ pore size syringe filter and left to sit overnight in the 60°C oven. It is spin cast onto a substrate using the same spin parameters as the non-crosslinked DNA-CTMA films. The substrates are baked in an 80°C oven for 5 min and then cured in a vacuum oven at 175°C for 15 min.

First we fabricated metal-insulator-metal (MIM) and metal-insulator-semiconductor (MIS) devices for the characterization of the dielectric properties of DNA-CTMA and DNA-CTMA-PPIF films. Such films were sandwiched between a 0.6 nm/60 nm chromium/gold (Cr/Au) bottom elec-

trode, which was evaporated on top of a quartz substrate and a 60 nm Au top electrode. An HP model 4248A instrument was used for the capacitance versus frequency measurements. The plots of the dielectric response of the DNA-CTMA and DNA-CTMA-PPIF films (Fig. 1) show that dc capacitance of DNA-CTMA-PPIF is ~ 0.8 nF/cm². From the measurement of capacitance per unit area, we determined a dielectric constant of $\epsilon_{\text{DNA/CTMA/PPIF}}=5.4$.

We then fabricated an n-channel BiOFET, evaporating 80 nm C₆₀ [Fig. 2(a)] and p-channel BiOFET evaporating 80 nm α -T6 [Fig. 3(a)] as organic semiconductors. We fabricated device structures using top contact geometry on top of a layer of 0.5 μ m DNA-CTMA-PPIF, serving as the gate dielectric. For two types of BiOFETs, 60 nm Au electrode was employed as the bottom contact and Cr/Au (0.6 nm/60 nm) electrodes were used as the source-drain contacts; the channel length L of the device was 20 μ m and the channel width was $W=200$ μ m, giving a W/L ratio of 10. All the device electrical characterization was carried out under an argon environment inside the glove box. An Agilent model E5273A with a two source-measurement unit instrument was employed for the steady state current-voltage (I - V) measurements. All measurements were performed at a scan rate of 0.2 V/s.

Figure 2(b) shows the output characteristics of n-channel BiOFET device. For an applied positive drain-source voltage V_{ds} , above a positive gate voltage, V_{gs} , a saturated drain current, I_{ds} , is observed in the range of 2×10^{-7} A for applied V_{gs} of 20 V. Transfer characteristics with applied large V_{ds} of 20 V are shown in Fig. 2(c). In case of using crosslinked DNA-CTMA film as a gate dielectric, hysteresis is vanished. We also determined the mobilities from linear regime using the standard transistor equation¹⁷ to be 9.25×10^{-3} and 9.34×10^{-3} cm²/V s for n-channel BiOFET devices using crosslinked DNA-CTMA and non-crosslinked DNA-CTMA films as a gate dielectric, respectively.

Similarly, the output characteristics and transfer curves of the p-channel BiOFET are shown in Figs. 3(b) and 3(c). The hysteresis in the transfer characteristics can also be reduced by using crosslinked DNA-CTMA dielectric. The mobilities from linear regime are 0.42 and 0.09 cm²/V s for p-channel BiOFET devices using crosslinked DNA-CTMA

and non-crosslinked DNA-CTMA films as a gate dielectric, respectively.

In summary, we have fabricated BiOFETs using a crosslinked DNA-based biopolymer for the gate dielectric. Our results show that it is possible to reduce the hysteresis as shown in previous works^{5,6} by using crosslinked DNA-CTMA film heavily improving the transistor function. Cigdem Yumusak gratefully acknowledges the financial support of European Science Foundation (ESF Organizational Project) for her scientific visit to Linz Institute for Organic Solar Cells (LIOS) to perform these experimental studies.

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