

Bio-molecules and Organic Dielectrics for Organic Field-Effect Transistors

Th. Birendra Singh and Niyazi Serdar Sariciftci

Linz Institute of Organic Solar Cells (LIOS), Physical Chemistry,
Johannes Kepler University Linz, Altenbergerstrasse, A-4040, Austria

Abstract

Organic field-effect transistors (OFETs) based on solution-processible polymeric as well as small molecular semiconductors have been improved in their performance during recent years. This article presents novel organic field-effect transistors developed by using various organic dielectrics and biomolecules such as deoxyribonucleic acid (DNA). Ambipolar charge transport in organic field-effect transistors using polyvinyl alcohol (PVA) have also been discussed. C₆₀ based high mobility OFETs and its applications to integrated circuits is also reviewed. The interface of the organic semiconductor and organic dielectric insulator influences the device performance and stability.

1. Introduction

Organic thin-film electronics has developed to a promising technology in the last decade with prototypes of organic integrated circuits for radio frequency identification tags (RFID-tags) [1, 2] and thin-film transistor (TFT) arrays for active matrix displays [3]. Organic field-effect transistors (OFETs) have also been fabricated in arrays to drive electrophoretic display pixels [4]. To date Bell Labs succeeded making organic integrated circuits with as many as 1888 transistors using vacuum evaporation techniques [2]. Using the same techniques, Infineon Technology has similar results on integrated circuits on special papers [5]. Polymer Vision, Philips Research Laboratory came up with flexible 4.7-inch QVGA active matrix display containing 76,800 organic transistors [6]. As the number of transistors per circuit increases there is an increasing need for circuits characterized by low power dissipation, high noise margin, and greater operation stability. The performance of the individual transistor limits the switching speed in an integrated circuit, which can be roughly estimated by the ratio of mobility and channel length of the transistor [3]. To obtain higher switching speed, the search for higher mobility materials is therefore important along with the effort to downscale the transistor geometry. Current benchmark for high mobility materials among various organic semiconductors are pentacene [7] and fullerenes (for both $\mu \sim 6$ cm²/Vs) [8, 9] for p-type and n-type, respectively.

Organic field-effect transistors have been fabricated with various device geometries as depicted in **Figure 1a-d**.

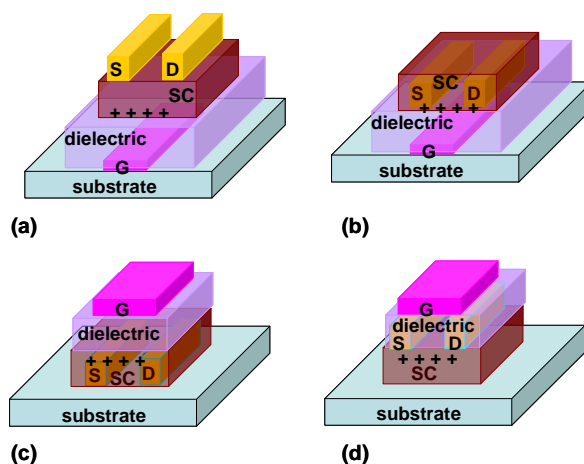


Figure 1. Schematic of the bottom-gate organic field-effect transistors (OFETs) with (a) top contact (b) bottom contact structures. Schematic diagram of a (c) top-gate/bottom contact OFETs using a standard TFT device structures and (d) top-gate /top contact is also shown.

The most commonly used device geometry is bottom gate with top contact partly because of borrowing the concept of thin-film silicon transistor (TFT) using thermally grown Si/SiO₂ oxide as gate dielectric. Due to the advantage of having commercially available high quality Si/SiO₂ substrate, it has dominated the whole community. Recently it has been shown that organic dielectrics are also promising for high performance OFETs [10-19]. Organic dielectrics (i) can be solution-processed, (ii) provide smooth films on transparent glass and plastic substrates, (iii) are suitable for opto-electronics like photo-responsive OFETs due to their high optical transparency, (iv) can be thermally stable up to 200 °C with a relatively small thermal expansion coefficient, and (v) can possess a rather high dielectric constant up to 18. The physics of organic dielectrics is a well developed branch of science and technology and will not be further discussed here. We simply display the chemical structures of commonly used organic dielectrics (**Figure 2**)

We present here an overview of OFETs using various organic dielectrics and bio-molecules such as deoxyribonucleic acid (DNA). High performance basic circuit element using high mobility C₆₀ OFETs have also discussed.

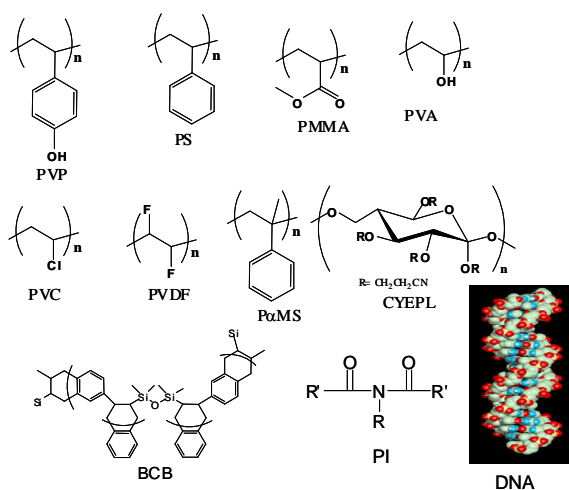


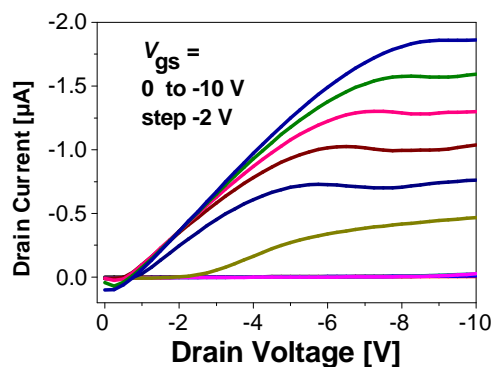
Figure 2: Chemical structure of some commonly used organic dielectric. PVP: Poly(4-vinyl phenol); PS: Polystyrene; PMMA: polymethyl-methacrylate; PVA: polyvinyl alcohol; PVC: polyvinylchloride; PVDF: polyvinylidene fluoride; P α MS: poly[α -methylstyrene], CYEPL:cyano-ethylpullulan and BCB: divinyltetramethyldisiloxanebis(benzocyclobutene),PI(po ly-imid),DNA (Deoxyribonucleic acid).

2. Results and Discussion:

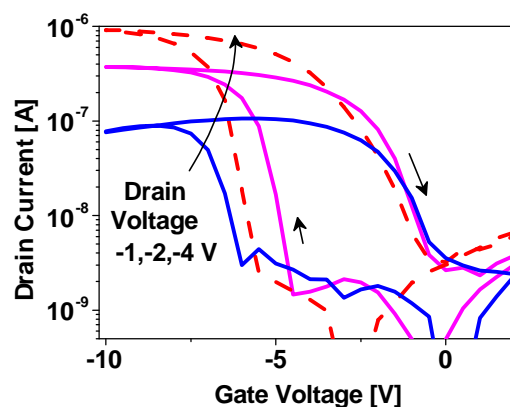
2.1. (a) Bio Organic Field-Effect Transistors (BIOFETs)

The DNA used for this research was purified DNA provided by the Chitose Institute of Science and Technology (CIST) [20-21]. It was marine-based, first isolated from frozen salmon milt and roe sacs through a homogenization process. It then went through an enzymatic treatment to degrade the proteins by protease. We found, however, that the purified DNA was soluble only in water, so not compatible with typically 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, hexadecyltrimethylammonium chloride (CTMA), by an ion exchange reaction [22-24].

Typical output characteristics of the BiOFET device we fabricated are plotted in Fig. 3(a). As can be seen, the BiOFET, with a 200 nm thick film of DNA-CTMA and a pentacene semiconductor, was able to modulate the drain current over three orders of magnitude using gate voltages of less than 10 V. Fig. 3(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. 3(b). The channel length ($L = \sim 20 \mu\text{m}$) and width ($W = \sim 1.5 \text{ mm}$), were used, along with the capacitance ($C_i = 1.15 \text{ nF/cm}^2$), to extract a saturated regime mobility of $0.05 \text{ cm}^2/\text{Vs}$ in our device, using the standard transistor equation,



(a)



(b)

Figure: 3 (a) BiOFET output characteristics; drain current, I_{ds} vs. versus 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 . [25]

in which there is no correction for contact resistances. We determined the linear mobility to be $10^{-2} \text{ cm}^2/\text{Vs}$. 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.

2.2. (b) Ambipolar Organic Field Effect Transistors (AOFETs)

Surface energy of a chosen dielectric plays an important role in the orientation and packing of the organic semiconductors on top of it. Figure 4 shows that organic semiconductor pentacene, can be ambipolar on polyvinylalcohol as dielectrics [26]. If the standard Si/SiO₂ dielectric gate transistor geometry is used, normally pentacene will only give p-type OFET operation. This demonstrates that using organic dielectrics like PVA can have fundamental influence on the character of the OFETs.

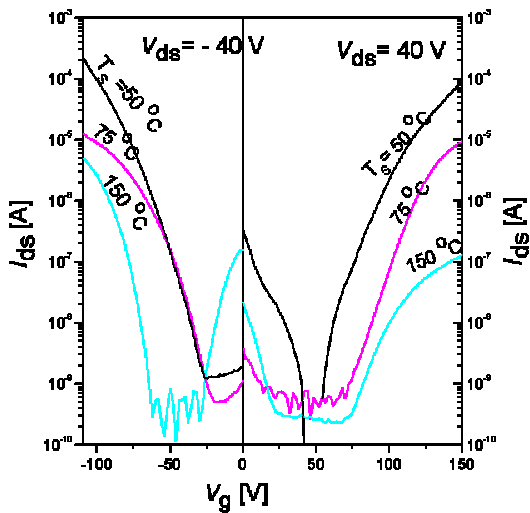


Figure 4: Transistor transfer characteristics of ambipolar pentacene OFETs using polyvinyl alcohol gate dielectric for which pentacene was deposited at different elevated temperatures showing both electron-enhanced mode as well as hole-enhanced mode.

2.3. (c) Ring Oscillators using C₆₀ OFETs

Depending on the elevated substrate temperature C₆₀ films grown by hot wall epitaxy can have striking film morphology as shown in Figure 5 can give charge carrier mobility ranging from 0.6 to 6 cm²/Vs [8,9]. Since C₆₀ exhibit high electron mobility, various types of C₆₀ based unipolar inverters have been fabricated in author’s laboratory. It also provides the possibility of integrating a number of such inverters for the construction of integrated complementary-like ring oscillators (see Figure 6). This was achieved by connecting an odd number of inverters in series and providing a feedback from the output of the last inverter to the input of the first, by borrowing the concept from literature [27]. A schematic representation of the circuit is shown Figure 6 a-b. The output characteristic of the oscillator circuit is shown in Figure 6c. Oscillating frequency as high as 30 KHz can be obtained from these ring oscillators.

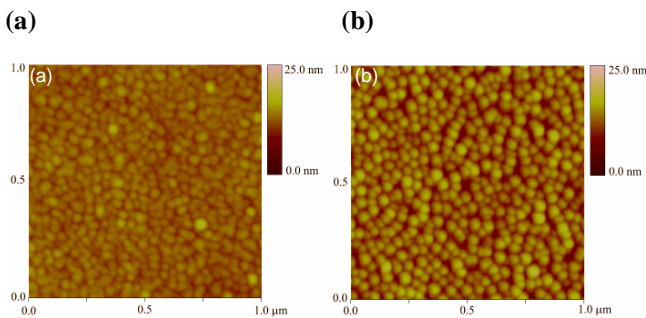


Figure 5: AFM topography images of (a) 5 nm C₆₀ film grown on BCB substrates grown at 25 °C (b) 5 nm C₆₀ film grown on BCB substrates with elevated substrate at 250 °C.

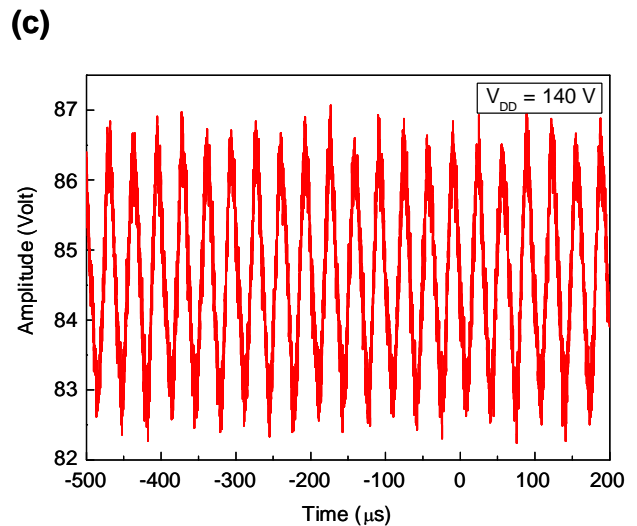
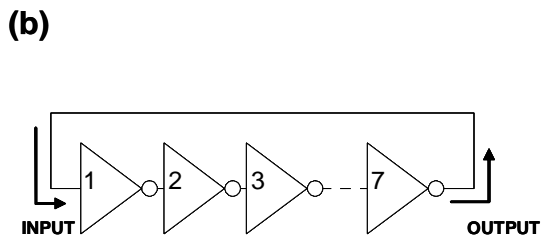
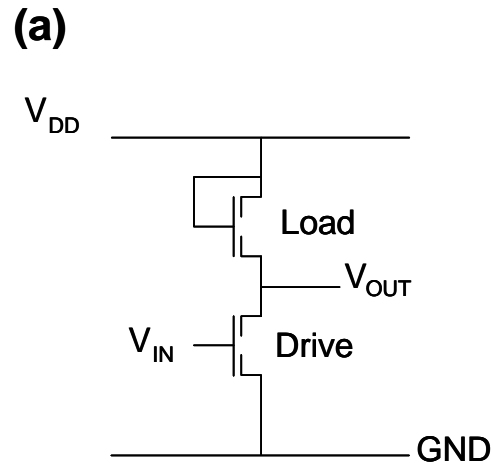


Figure 6 (a) shows the scheme of the unipolar inverter circuit using C₆₀ OFETs (b) Shows the schematic circuit of a ring-oscillator consisted of seven inverters. (c) Oscillating signal measured at the output of the ring oscillator.

3. Acknowledgments

We acknowledge Dr. Thomas D. Anthopoulos and Dr. Dago M. de Leeuw of Philips Research Laboratories, Eindhoven and Prof. Dr. Siegfried Bauer and Dr. Reinhard Shrówdiauer of Soft Matter Physics, JKU Linz, Dr. James G. Grote, Air Force Research Laboratory, Wright-Patterson Air Force Base, Ohio, USA, Prof. Dr. Gilles Horowitz of ITODYS, CNRS-UMR 7086, University Denis-Diderot, Paris and colleagues; Dr. Helmut Sitter of Institute of semiconductor physics, JKU for the collaborations. We also acknowledge financial support from Austrian Science Foundation, FWF project No. P16891-N08 and N00103000.

4. Reference:

- [1a] Clemens W, Fix W, Ficker J, Knobloch A. 2004. Ullmann A. From polymer transistors toward printed electronics. *J. Mat. Res.* 19:1963
- [1b] Horowitz G. 2004. Organic thin film transistors: From theory to real device. *J. Mat. Res.* 19:1946
- [2] Crone B, Dodabalapur A, Lin Y-Y, Filas RW, Bao Z, et al. 2000. Large-scale complementary integrated circuits based on organic transistors. Li, *Nature* 403:521
- [3] Huitema HEA, Gelinck GH, van der Putten JBPH, Kuijk KE, Hart CM, et al. 2001. Plastic transistors in active-matrix displays. *Nature* 414:599
- [4a] Rogers JA, Bao Z, Baldwin K, Dodabalapur A, Crone B, et al. 2001. From the Cover: Paper-like electronic displays: Large-area rubber-stamped plastic sheets of electronics and microencapsulated electrophoretic inks. *Proc. Natl. Acad. Sci.* 98:4835
- [4b] Sundar VC, Zaumseil J, Podzorov V, Menard E, Willett RL, et al. 2004. Elastomeric Transistor Stamps: Reversible Probing of Charge Transport in Organic Crystals. *Science* 303:1644
- [5] Eder F, Klauk H, Halik M, Zschieschag U, Schmid G, Dehm C. 2004. Organic electronics on paper. *Appl. Phys. Lett.* 84:2673
- [6] Huitema HE, Gelinck GH, Van Veenendaal E, Cantatore E, Touwslager FJ, et al. 2003. A flexible QVGA Display with Organic Transistors. IDW 1663
- [7] Kelley TW, Muyres DV, Baude PF, Smith TP, Jones TD. 2003. High performance organic thin film transistors, in *Organic and Polymeric Materials and Devices*, edited by P.W.M. Blom, N.C. Greenham, C.D. Dimitrakopoulos, and C.D. Frisbie. (*Mater. Res. Soc. Symp. Proc.* 771, Warrendale, PA), L6.5.1
- [8] Singh ThB, Marjanović N, Matt GJ, Günes S, Sariciftci NS, et al. 2005. *Organic Thin-Film Electronics*, edited by A.C. Arias, N. Tessler, L. Burgi, and J.A. Emerson (*Mater. Res. Soc. Symp. Proc.* 871E, Warrendale, PA), I 4.9.1.
- [9] Montaigne Ramil A, Singh ThB, Haber NT, Marjanović N, Günes S, et al. 2006 Influence of Film Growth Conditions on Carrier Mobility of Hot Wall Epitaxially Grown Fullerene based Transistors. *J. Cryst Growth*, 288: 123
- [10] X. Peng, G. Horowitz, D. Fichou, and F. Garnier, *Appl. Phys. Lett.* 57, 2013 (1990)
- [11] H. Klauk, M. Halik, U. Zschieschang, G. Schmid, W. Radlik, *J. App. Phys.* 92, 5259 (2002)
- [12] R. Parashkov, E. Becker, G. Ginev, T. Riedl, H. H. Johannes, and W. Kowalsky, *J. Appl. Phys.* 95, 1594 (2006).
- [13] J. Park, S. Y. Park, S. Shim, H. Kang and H. H. Lee, *Appl. Phys. Lett.* 85, 3283 (2004).
- [14] R. Schroeder, L. A. Majewski and M. Grell, *Appl. Phys. Lett.* 83, 3201 (2003)
- [15] D. Knipp, R. A. Street, B. Krusor, J. Ho, R. B. Apte, *Mat. Res. Soc. Symp. Proc.* 708 ,BB.10 (2002).
- [16] R. Schroeder, L. A. Majewski and M. Grell, *Adv. Mater.* 16, 633 (2004)
- [17] Th. B. Singh, N. Marjanović, G. J. Matt, N. S. Sariciftci, R. Schwödiauer and S. Bauer., *Appl. Phys. Lett.* 85, 5409 (2004).
- [18] R. C. G. Naber, C. Tanase, P. W. M. Blom, G. H. Gelinck, A. W. Marsman, F. J. Touwslager, S. Setayesh and D. M. De Leeuw *Nat. Mat.* 4, 243 (2005).
- [19] Th. B. Singh, N. Marjanović, P. Stadler, M. Auinger, G. J. Matt, S. Günes, N. S. Sariciftci, R. Schwödiauer and S. Bauer, *J. Appl. Phys.* 97, 083714 (2005).
- [20] L. Wang, J. Yoshida, N. Ogata, S. Sasaki, and T. Kajiyama, *Chemistry of Materials*, 13(4), 1273 (2001).
- [21] G. Zhang, L. Wang, J. Yoshida and N. Ogata, *SPIE Proc. – Optoelectronic, Materials and Devices for Communications*, eds., Q. Wang and T. Lee, 4580, 337 (2001).
- [22] K. Tanaka, Y. Okahata, *J. Am. Chem. Soc.*, 1996, 118, 10679
- [23] H. Kimura, S. Machida, K. Horie, Y. Okahata, *Polymer Journal*, 1998, 30, 708
- [24] J. Grote, N. Ogata, J. Hagen, E. Heckman, M Curley, P. Yaney, M. Stone, D. Diggs, R. Nelson, J. Zetts, F. Hopkins and L. Dalton, *SPIE Proc. – Nonlinear Optical Transmission and Multiphoton Processes in Organics*, eds., A. Yates, K. Belfield, F. Kajzar and C. Lawson, 5221, 53, (2003).
- [25] Th. B. Singh, N.S. Sariciftci, J. Grote, F. Hopkins, *Journal of Applied Physics* 100 (2006), 024514.
- [26] Singh ThB, Meghdadi F, Günes S, Marjanović N, Horowitz G, et al. 2005. High-Performance ambipolar pentacene organic field-effect transistors on poly(vinylalcohol) organic gate dielectric. *Adv. Mater.* 17:2315
- [27] T. D. Anthopoulos, D. M. de Leuw, E. Cantatore, P. van 't Hof, J. Alma and J. C. Hummelen, *J. Appl. Phys.* 98, 054503 (2005).