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ABSTRACT

The field of organic electronics is advancing quickly towards ultra low-cost, low-end applications and is expected to provide the necessary technology required for flexible/printed electronics. Here we address the need for solution processed low-voltage complementary logic in order to reduce power consumption of organic circuits and hence enable their use in portable, i.e. battery-powered applications. We demonstrate both p- and n-channel solution processed high performance organic field-effect transistors that operate at voltages below |1.5| V. The reduction in operating voltage is achieved by implementing ultra-thin gate dielectrics based on solution processed self-assembled monolayers. This work demonstrates the feasibility of fabricating low-voltage complementary organic circuits by means of solution processing.

1. Introduction

Organic materials are expected to play a major role in low-cost, low-end electronics in the very near future. The vast majority of state-of-the-art organic devices are based on vacuum deposited organic semiconductors. Vacuum deposition allows easy fabrication of unipolar circuits employing a single material that conducts either holes (p-channel) or electrons (n-channel). Despite the simplicity in fabricating such unipolar circuits there are major drawbacks associated with this technology. In particular, high power consumption, low gain and narrow noise margins hinder performance of unipolar logic. The alternative is complementary logic (employing both p- and n-channel devices), known from conventional inorganic microelectronics to provide significantly better performance and lower power consumption [1]. However, patterning of p- and n-channel devices on the same substrate by vacuum deposition is complicated and hence, potentially, more expensive. Processing organic semiconductors from solution (i.e. by spin-coating, inkjet printing, gravure printing, etc.) could provide a cheap method for large area/volume production.

Another major drawback associated with most organic devices and circuits reported to date is the high operating voltage, i.e. >20 V [1,2]. In order to make the technology compatible with portable and battery powered applications the devices have to operate at only a few volts. In organic field-effect transistors (OFETs) the geometric capacitance (C_i) of the gate dielectric, and therefore the density of mobile charge induced by the gate voltage (V_G) in the transistor channel, is inversely proportional to the dielectric thickness. Reducing its thickness from a few hundred nanometres to a few nanometres enables transistor operation at significantly smaller gate voltages [3–5]. Previously Klauk et al. have achieved this with solution processed organic ultra-thin gate dielectrics based on a self-assembled monolayer (SAM) of octadecylphosphonic acid (ODPA) functionalised on partially oxidised aluminium gate electrodes [6,7].

Here we employ a similar approach to fabricate low-voltage OFETs employing a side-chain fluorinated fulleropyrrolidine and poly(3-hexylthiophene) (P3HT) as solution processed organic semiconductors for the n- and p-channel, respectively (Fig. 1a). Fullerenes are known from the literature as excellent n-channel organic semiconductors [8,9]. We previously demonstrated high

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performance, low-voltage devices based on soluble fullerene derivatives [10]. P3HT was previously demonstrated by Park et al. [11] to be a suitable p-channel material for the fabrication of solution-processed low-voltage OFETs employing a methyl terminated trichlorosilane based SAM as the gate dielectric. By combining these n- and p-channel OFETs we demonstrate the feasibility of solution processed low-voltage organic complementary circuits.

2. Experimental

OFETs were fabricated with ultra-thin gate dielectrics based on a monolayer of ODPA. Al gate electrodes were patterned on cleaned glass substrates by thermal evaporation in vacuum through shadow masks. The Al surface was briefly exposed to oxygen plasma before immersion in a 5 mM solution of ODPA molecules in isopropanol (IPA). After the treatment the samples were dried in a furnace at 140 °C under nitrogen for several hours. Then residual molecules were removed by rinsing with neat IPA [10].

The organic semiconductors were spin coated in a nitrogen atmosphere from chlorobenzene solutions followed by thermal evaporation of metal contact electrodes. The P3HT film was annealed under a nitrogen atmosphere at 90 °C for 1 h before testing. The semiconductor film thickness was typically in the range of 50–200 nm.

The electrical characterization was performed under high vacuum (10^{-5} mbar) employing an Agilent Semiconductor Parameter Analyzer. Samples were briefly exposed to air during transport from the nitrogen atmosphere to the vacuum chamber.

3. Results and discussion

The geometrical capacitance of the AlO_x–ODPA dielectric was measured to be approximately 600–800 nF/cm^2 in good agreement with previously reported values [6]. Upon functionalization with ODPA, the surface of the gate electrodes becomes extremely hydrophobic due to the low and dispersive surface energy of the monolayer. This is confirmed by a high water contact angle of >110° in agreement with prior reports [6,10]. As a result, the deposition of high mobility, small molecule organic semiconductor solutions into homogeneous films by spin coating on such surfaces is typically problematic. Previous surface energy analysis explains the wetting behaviour [10]. Smooth films and working FETs were obtained with a particular side-chain fluorinated fulleropyrrolidine (F17-DOPF) (Fig. 1) [12]. It was shown in the previous report that chlorobenzene solutions of this molecule exhibit suitable surface energy characteristics for deposition onto the AlO_x–ODPA dielectric [10]. Excellent n-channel transistor characteristics were obtained with Al source and drain top contacts at operating voltages below |1.5| V (Fig. 2). Electron injection into the lowest unoccupied molecular orbital (LUMO) of F17-DOPF (LUMO ≈ 3.5–3.8 eV) is achieved due to its good alignment with the metal workfunction of Al (≈4.1 eV) [12]. The same device architecture was also used to fabricate p-channel transistors employing P3HT as the organic semiconductor but using gold (Au, work function ≈5.1 eV) source/drain electrodes to facilitate hole injection into the highest occupied molecular orbital (HOMO) of P3HT (≈4.8–5.1 eV) [13,14]. P3HT formed acceptable films on the dielectric following spin coating from a chlorobenzene solution, presumably thanks to the high molecular weight of the polymer increasing the viscosity of the solution [15]. P3HT based p-channel devices exhibited lower saturation mobility, higher threshold voltages ($|V_T|$) and lower on/off current ratios than the n-channel OFETs, although they were comparable to previously reported low-voltage P3HT transistors [11]. The gate leakage currents were always at least one order of magnitude lower than the drain current (I_D). Representative values for the characteristic performance parameters of p- and n-channel transistors are given in Table 1. The field-effect mobility (μ) and V_T were calculated from a linear fit to a plot of $\sqrt{I_D}$ in saturation vs. V_G (Fig. 2) using Eq. (1), assuming the gradual-channel approximation is satisfied [16].

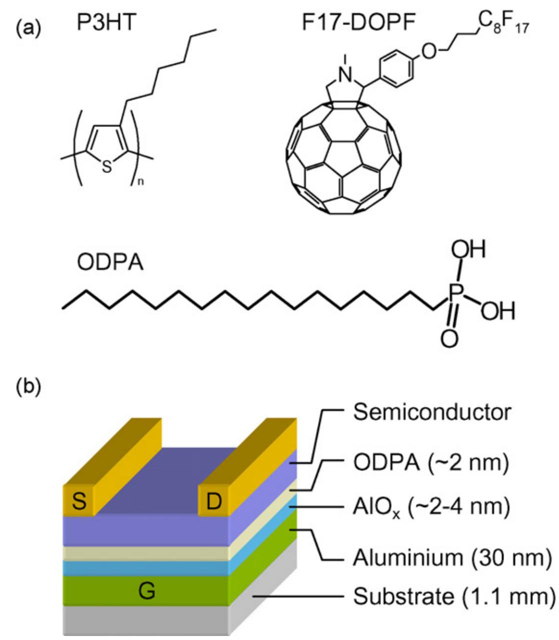

Fig. 1. (a) Molecular structures of the organic semiconductors and SAM insulator used for device fabrication. Poly(3-hexylthiophene) (P3HT), side-chain fluorinated fulleropyrrolidine (F17-DOPF) and octadecylphosphonic acid (ODPA). (b) Schematic structure of the bottom-gate, top-contact device architecture employed.

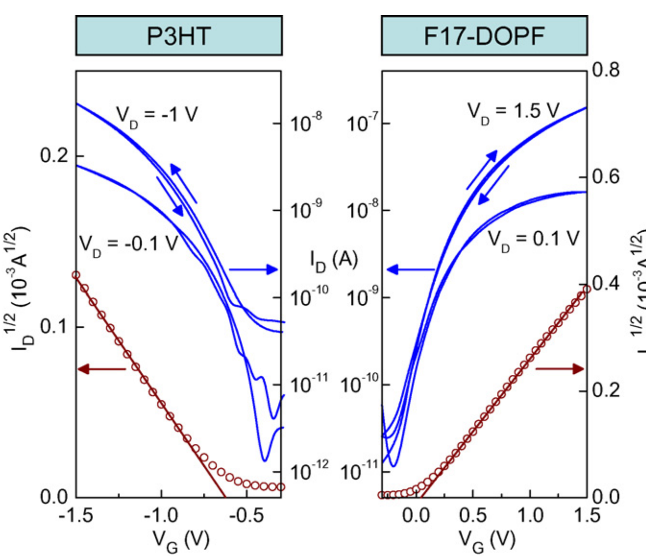

Fig. 2. Transfer characteristics of low-voltage OFETs employing solution processed P3HT (after annealing) and F17-DOPF as the organic semiconductors. The dimensions of the transistor channels were W = 1 mm and L = 60 μm for both devices.

$$I_{D,sat} = \frac{WC_i}{2L} \cdot \mu \cdot (V_G - V_T)^2 \quad (1)$$

Table 1
Summary of low-voltage OFET characteristics.

	μ_{sat} (cm^2/V s)	V_T (V)	$|S|$ (mV/dec)	I_{on}/I_{off}
P3HT	0.006	−0.6	250	>10^2
F17-DOPF	0.02	0.1	180	>10^3

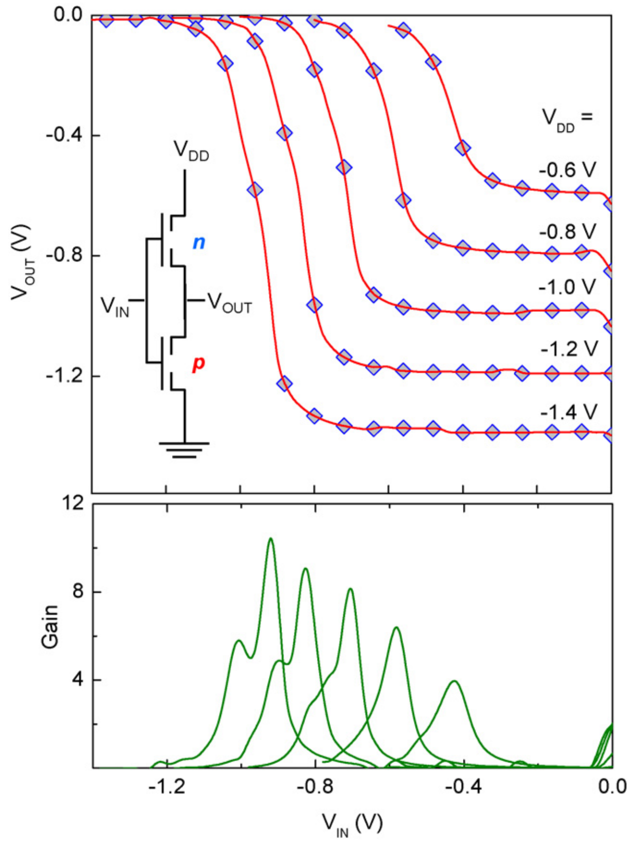

Fig. 3. Output characteristics and the corresponding gains of a complementary inverter fabricated using p- and n-channel low-voltage OFETs. *Inset*: Circuitry of the complementary inverter.

Here, L and W are the length and width of the transistor channel respectively. The subthreshold slope S was calculated using Eq. (2).

$$S = \frac{\partial V_G}{\partial(\log_{10} I_D)} \quad (2)$$

The solution processed p- and n-channel devices were then combined to produce complementary voltage inverters employing the circuitry shown in the inset of Fig. 3. The operating characteristics of the inverter (Fig. 3) show high gain (defined as $\partial V_{OUT}/\partial V_{IN}$) of the order of >10. The noise margin was about 60% of the maximum theoretical value, i.e. $V_{DD}/2$. Moreover, the hysteresis between forward and backward scans was negligible. As expected from complementary logic, the circuit current during quasistatic operation of the inverter was low (~1 nA) resulting in static power dissipation below 5 nW per logic gate. This power consumption is slightly higher than the value reported by Klauk et al. (<1 nW per logic gate) [6]. Improvement of the power consumption characteristics is expected upon patterning of the organic semiconductors. Finally, we note that the overall inverter performance is limited by the operating characteristics of the p-channel device and could be improved by implementing superior hole transporting materials. This work is currently under way and will be reported in the future.

4. Conclusion

We have demonstrated solution-processed low-voltage hole and electron-transporting OFETs enabled by the use of an ultra-thin SAM gate dielectric. By combining the two types of transistors we have also demonstrated the feasibility of fabricating low-voltage complementary logic circuits employing solution processing techniques. This work is a significant step towards the realisation of low-voltage, low-power organic electronics employing low-cost fabrication methods.

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