

Low-Voltage, High-Gain, and High-Mobility Organic Complementary Inverters Based on N,N'-Ditridecyl-3,4,9,10-Perylenetetracarboxylic Diimide and Pentacene

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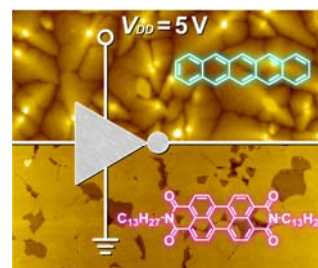
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The authors describe an organic complementary inverter with N,N'-ditridecyl-3,4,9,10-perylenetetracarboxylic diimide as an n-type semiconductor and pentacene as a p-type semiconductor. Each transistor of the inverter exhibited high carrier mobility: 1.62 cm²/Vs for an n-type drive transistor and 0.57 cm²/Vs for a p-type switch transistor. The gain of the inverter reached 125. Another inverter using Ta₂O₅ as a high κ gate dielectric performed well with a gain of 500 and an operation voltage of only 5 V.



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Organic thin-film transistors (OTFTs) have attracted considerable interest in recent years because of their many potential applications, such as flexible, large-area, and low-cost electronic devices [1,2]. The mobility of OTFTs has increased significantly, and the mobility of discrete OTFT devices has already overcome that of hydrogenated amorphous silicon TFTs [3,4]. In this context, organic transistors should be applied to monolithic circuits with several OTFTs. Complementary metal oxide semiconductor (CMOS)-like circuits have easier circuit design and lower power consumption essentially. Recently, CMOS-like organic circuits have been reported by several groups [5,6]. For example, Klauk *et al.* demonstrated organic CMOS-like logics: NOT and NAND gates and a 5-stage ring oscillator using an organic self-assembly monolayer as a gate dielectric. The logics worked below 3 V and consumed very low power: only about 1 nW per gate [7]. Consequently, organic CMOS-like circuits should be hopefully

applied to portable devices that will be driven by a non-contact power supply or by small batteries in the future.

An inverter is a basic building block for complex integrated circuits. Organic inverters require high gain, low operation voltage, absence of hysteresis, and high switching speed to be effectively used in integrated circuits. The gain of the inverter is expressed by the 1st-order derivative of an output voltage as a function of the input voltage curve: $-dV_{out}/dV_{in}$. Hence, a higher gain induces a higher noise margin. The switching speed of the inverters is ideally limited by the channel transit time of charge carriers and the charging and discharging times of the gate capacitance. Hence, being of high mobility and miniaturizing circuit design is important. Thus, fabricating organic CMOS inverters of high mobility OTFTs is greatly advantageous. Although the high-gain and low-voltage organic CMOS inverters were previously reported, there is no report about organic CMOS inverters composed both of high mobility n- and p-type transistors to the best of our knowledge.

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Pentacene is a well-known p-type material with high mobility in a thin-film state. High hole mobility of pentacene thin-films has exceeded that of hydrogenated amorphous silicon, reaching $5.5 \text{ cm}^2/\text{Vs}$ [8]. However, comparable n-type OTFT materials with mobility are scarce. High performance n-type OTFTs [9-11] have been reported by some researchers, and we have also demonstrated a high mobility n-type OTFT with N,N'-ditridecyl-3,4,9,10-perylenetetracarboxylic diimide (PTCDI-C13, see Figure 2(a) to determine its structure) [12]. PTCDI is well known as an n-type OTFT material [13], and alkyl-substituted PTCDI (PTCDI-Cn) has better n-type transistor operation [11,13,14]. We have demonstrated that an n-type OTFT using PTCDI-C13 with a longer alkyl chain exhibits a high field effect electron mobility of $2.1 \text{ cm}^2/\text{Vs}$ by just annealing it after device fabrication [12]. The reason is that the annealing improves the grain morphology and crystallinity of PTCDI-C13 thin films due to the high motion freedom of the long alkyl chain. In this letter, we report an organic CMOS inverter based on high mobility PTCDI-C13 and pentacene as n- and p-type OTFTs, respectively.

The inverters were fabricated in a top-contact configuration, shown in Fig. 1. The device was prepared on a heavily doped p-type silicon wafer with a 200-nm-thick thermally grown SiO_2 layer, which was utilized as a gate electrode and a gate dielectric (specific capacitance of $15 \text{ nF}/\text{cm}^2$). A PTCDI-C13 (Aldrich) layer was thermally deposited at $0.3 \text{ \AA}/\text{s}$ onto a prepared substrate at room temperature. During the deposition, the PTCDI-C13 layer was patterned with a shadow mask to create a definite separation between n- and p-type semiconductors. The film was thermally treated at 140°C in a vacuum for 1 hour and then slowly cooled to room temperature while maintaining the vacuum. A pentacene (Tokyo Chemical Industry) layer was also prepared by thermal evaporation at a deposition rate of $0.3 \text{ \AA}/\text{s}$ next to the PTCDI-C13 layer. The pentacene layer was also patterned with another shadow mask. Finally, Au was deposited onto the semiconductor films through another shadow mask to form contact electrodes. The channel width and channel length of each transistor were $2000 \mu\text{m}$ and $100 \mu\text{m}$. The inverter characteristics were measured in a vacuum chamber having four individual probe terminals at room temperature using two Keithley 2410 source meters.

We fabricated another organic complementary inverter that operates at a much lower voltage. Using high permittivity materials as gate insulators is very effective in decreasing operation voltages. We fabricated an organic complementary inverter using Ta_2O_5 ($\epsilon = 20\text{-}23$) as a high dielectric constant gate insulator, which was reported by Y. Iino *et al.* [15]. We formed 133-nm Ta_2O_5 using anodic oxidation of a Ta film deposited on a glass substrate in an aqueous solution of citric acid at an applied voltage of 70 V at room temperature. The specific capacitance of the Ta_2O_5 gate insulator was $140 \text{ nF}/\text{cm}^2$. A remaining tantalum layer was used as a gate electrode for an inverter. Annealing PTCDI-C13 at 200°C led to a smaller threshold voltage than at 140°C , but the mobility decreased. Consequently, we thermally treated PTCDI-C13 at 200°C . Other fabrications and measurements were the same as ones already mentioned, except for the channel length ($25 \mu\text{m}$).

Fig. 2 shows the drain current-drain voltage (I_D - V_D) plots of individual PTCDI-C13 and pentacene TFTs in the inverter fabricated on the SiO_2/Si substrate. Note that the characteristics were measured with three terminals: V_{in} as a gate, V_{out} as a source, and V_{DD} or V_{SS} as a drain electrode. The mobility, threshold voltage, on/off ratio, and sub-threshold swing for the PTCDI-C13 TFT were $1.62 \text{ cm}^2/\text{Vs}$, 38 V , 10^6 , and $2.57 \text{ V}/\text{decade}$, respectively, and these parameters for the pentacene TFT were $0.57 \text{ cm}^2/\text{Vs}$, -19 V , 10^5 , and $2.11 \text{ V}/\text{decade}$ for the pentacene TFT. Here, the mobilities were obtained from the saturation region. Each TFT showed high mobility, a high ON/OFF ratio, and a good subthreshold performance.

Fig. 3 shows the input-output (V_{in} - V_{out}) characteristics of the organic complementary inverter, where the V_{SS} terminal was shorted to the earth. As shown in the figure, this inverter exhibited good characteristics with a small hysteresis and a high gain of 125. The gain mainly depends on transconductances of n- and p-type TFTs at the midpoint voltage of the inverter. A TFT with high mobility exhibits a high transconductance. Because the PTCDI-C13 and pentacene TFTs showed high mobility, as shown in Fig. 2, the inverter had high gain. Midpoint voltages, V_M , for forward and backward bias sweeps were 41.7 V and 42.3 V from V_{in} - V_{out} characteristics. We also obtained V_M from the current gain factor (β) of each TFT using this equation [16]:

$$V_M = \frac{V_{DD} + V_{th}^p + V_{th}^n \sqrt{\beta_n/\beta_p}}{1 + \sqrt{\beta_n/\beta_p}} \quad (1)$$

where V_{th}^n and V_{th}^p are the threshold voltages for n- and p-type TFTs, and β_n and β_p represent the current gain factors of n- and p-type TFTs. The V_M for a forward bias sweep can be calculated using these parameters of the n-type TFT measured by the forward sweep and the parameters of the p-type TFT measured by the backward sweep. The measurement of the V_M for the backward bias sweep, on the other hand, should be made using these parameters of the n-type TFT measured by the backward sweep and the parameters of the p-type TFT measured by the forward sweep. The calculated V_M was 41.5 V for the forward sweep and 43.8 V for the backward sweep, corresponding to each measured V_M .

Finally, we describe a low-voltage-operational organic complementary inverter that uses Ta_2O_5 as a gate dielectric. Fig. 4 shows the I_D - V_D plots of individual PTCDI-C13 and pentacene TFTs. The mobility and threshold voltage for the PTCDI-C13 TFT were $0.21 \text{ cm}^2/\text{Vs}$ and 4.9 V , respectively, and these parameters for the pentacene TFT were $0.15 \text{ cm}^2/\text{Vs}$ and 2.1 V . The results of Fig. 5 show that the inverter works under an operation voltage of 5 V and exhibits good inverter characteristics with a gain of 150, an output voltage swing of 4.7 V , and a noise margin of 2.1 V . To obtain its intrinsic gain, we decreased the step voltage of V_{in} . The gain reached almost 500, as shown in the inset of the lower panel of Fig. 5. The mobilities were smaller than those of each TFT fabricated on SiO_2 . We believe that the surface condition of Ta_2O_5 might be different from that of SiO_2 . In addition, some hysteresis occurred. This hysteresis probably results from the hysteresis of both n- and

p-type transistors fabricated on Ta₂O₅ showing hysteresis due to charge trapping at the semiconductor/gate dielectric interface.

In conclusion, we fabricated organic complementary inverters using PTCDI-C13 and pentacene which intrinsically have high carrier mobility. An inverter using SiO₂ as a gate dielectric performed well with a small hysteresis and a highest gain of 125. We also fabricated an inverter using Ta₂O₅ which has high permittivity materials as a gate dielectric, and the inverter functioned at only 5 V. The gain of this inverter was as high as 500, and the noise margin remained at 2.1 V.

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References

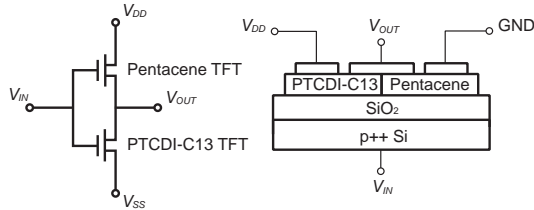


Figure 1 Circuit design and device structure of fabricated CMOS inverter.

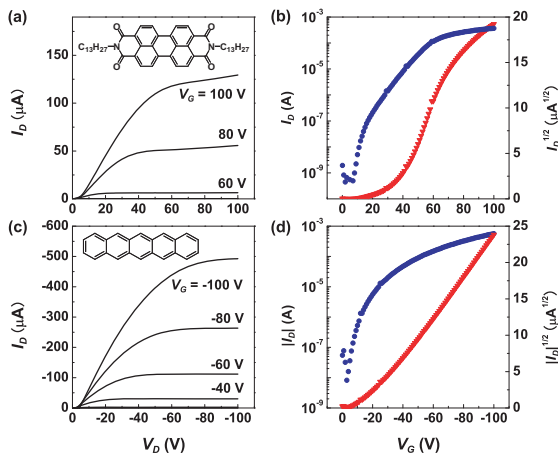


Figure 2 I_D - V_D and I_D - V_G characteristics of (a), (b) PTCDI-C13 and (c), (d) pentacene TFTs.

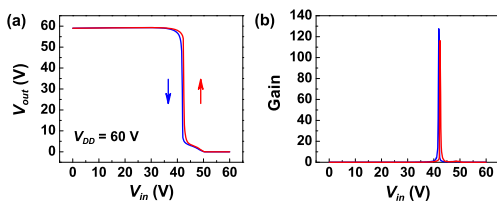


Figure 3 (a) V_{out} - V_{in} characteristics and (b) gain curves of CMOS

[1] C. D. Dimitrakopoulos and P. R. L. Malenfant, *Adv. Mater.* **14**, 99 (2002)
 [2] S. R. Forrest, *Nature* **428**, 911 (2004)
 [3] A. Facchetti, *Materials Today* **10**, 28 (2007)
 [4] C. R. Newman et al., *Chem. Mater.* **16**, 4436 (2004)
 [5] B. Crone et al., *Nature* **403**, 521 (2000)
 [6] H. Klauk et al., *IEEE Trans. Electron Devices* **52**, 618 (2005)
 [7] H. Klauk et al., *Nature* **445**, 745 (2007)
 [8] S. Lee et al., *Appl. Phys. Lett.* **88**, 162109 (2006)
 [9] S. Ando et al., *J. Am. Chem. Soc.* **127**, 14996 (2005)
 [10] T. D. Anthopoulos et al., *Appl. Phys. Lett.* **89**, 213504 (2006)
 [11] R. J. Chesterfield et al., *J. Phys. Chem. B* **108**, 19281 (2004)
 [12] S. Tatemichi et al., *Appl. Phys. Lett.* **89**, 112108 (2006)
 [13] G. Horowitz et al., *Adv. Mater.* **8**, 242 (1996)
 [14] P. R. L. Malenfant et al., *Appl. Phys. Lett.* **80**, 2517 (2002)
 [15] Y. Iino et al., *Jpn. J. Appl. Phys.* **42**, 299 (2003)
 [16] T. A. DeMassa and Z. Cicone, *Digital Integrated Circuits* (Wiley, New York, 1996), p.342

inverter using PTCDI-C13 and pentacene fabricated on SiO₂ gate insulator.

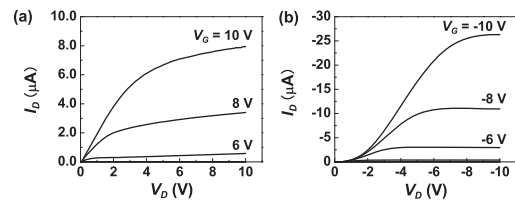


Figure 4 I_D - V_D characteristics of (a) PTCDI-C13 and (b) pentacene TFTs.

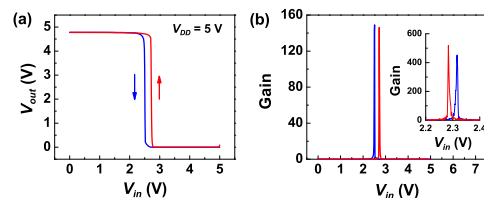


Figure 5 (a) V_{out} - V_{in} characteristics and (b) gain curves of CMOS inverter using PTCDI-C13 and pentacene fabricated on Ta₂O₅ gate insulator. The inset in the lower panel shows the precisely measured gain curve of the inverter.