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<th>Tunable Threshold Voltage of Organic CMOS Inverter Circuits by Electron Trapping in Bilayer Gate Dielectrics</th>
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Thus, it is necessary to place the switching voltage of the leads to the inverter is very susceptible to electrical noise. This results in a small noise margin, which in turn the transistor may be close to either the ground voltage or the power supply. This demonstrates the importance of controlling the switching voltage of the transistor elements in order to achieve the maximum noise immunity of the CMOS inverter [5], [6].

Several approaches have been recently introduced to tune the $V_{th}$ in organic transistor circuit. The $V_{th}$ tuning can be obtained by varying a doping concentration of Au nanoparticles (NPs) [7] or using polar self-assembled monolayers (SAMs) [8], [9] in a gate dielectric. Although the $V_{th}$ is well controlled with these techniques, the $V_{th}$ is only defined at manufacturing step [7]–[9]. To dynamically control the $V_{th}$ after fabrication, a number of methods have been developed. The tunability of the $V_{th}$ results from using space charge polarization [10] or charge-trapping polymer [11] or ferroelectric gate dielectrics [12], or Al floating-gate [13], [14]. However, the limitations of such methods are required long switching time and large switching voltage [10], [11] due to gradual formation of space charge polarization [10] or poor device performance resulting from the rough surface of ferroelectric materials [13] or instability of the shifted $V_{th}$ because the trapped charges easily leak through a SAM blocking-layer [13].

In our recent works [15], we have demonstrated highly stable transistor memories based on a double gate dielectric structure of poly(perfluoralkenyl vinyl ether) (CYTOP) and SiO$_2$. The $V_{th}$ can be modulated thanks to electron trapping at the interface between CYTOP and SiO$_2$. Taking the advantage of that, we have focused on the memory functionality by using the different levels of the $V_{th}$ to present the binary values. The resulting memory transistors exhibit high performance with a large on/off ratio and excellent retention stability of data storage [15], [16].

In this work, we extend the findings to construct electrically tunable CMOS inverter circuits with a bilayer-dielectric structure of CYTOP and SiO$_2$. We firstly, in both $n$-channel fullerene and $p$-channel pentacene OFET elements, examined the tunability of $V_{th}$ and its stability after tuning through a current measurement. We found that a change in the drain current of the OFETs followed a stretched-exponential distribution with a long time constant of 10$^5$ s. In the latter part of this report, we describe additional experiments on control of the CMOS inverter circuit. The voltage transfer characteristic of the inverter was positively shifted after applying the program gate voltages for both transistor types. At a program gate voltage of 60 V for $p$-channel OFET, the inverting voltage of 4.8 V was realized, which approximates the half supply voltage. This brings about the maximum immunity of the circuits against...
Fig. 1 Illustration of OFET and CMOS inverter circuit layout and chemical structures of materials utilized in our experiments together with equivalent circuit of inverter.

the effect of electrical noise.

2. Experimental Procedure

Figure 1 presents the schematic illustration of the inverter and the chemical structures of the used materials together with the equivalent circuit of the CMOS inverter. The transistor circuit was fabricated on a heavily doped Si (n+Si, resistivity: 1–100 Ω cm) gate electrode coated with a SiO₂ (50 nm) dielectric layer. The silicon wafer was cleaned by ultrasonication (in acetone for 5 min, in detergent for 10 min, twice in pure water for 5 min, and in IPA for 10 min) and subjected to UV-O₃ treatment. A CYTOP electron-trapping layer (CTL-809 M, Asahi Glass) was spin-coated onto the gate dielectric at 2000 rpm for 60 s using a 0.5 wt% CYTOP solution (CT-Solv. 180) and dried at 100°C for 2 h. The CYTOP thickness of 10 nm was the optimized value as presented in our earlier work [15]. Fullerene (C₆₀, Nanom purple SUH, purity: 99.9%) and pentacene (Sigma-Aldrich Company) were thermally deposited through shadow masks onto the CYTOP layer to form the 40-nm-thick semiconductor layers for the n-channel and p-channel OFETs, respectively. To produce transistor configuration, 70-nm-thick Al and 50-nm-thick Au source-drain electrodes were formed on C₆₀ and pentacene layers by thermal evaporation through shadow masks. The L and W of the OFETs were set at 75 and 5000 μm, respectively. Finally, to complete the inverter circuit, a 100-nm-thick Ag interconnection line was vacuum-deposited through a shadow mask. The base pressure during the vacuum deposition was of 2×10⁻⁶ torr. Electrical characteristics of the OFETs and CMOS inverter were measured with a Keithley 4200 semiconductor parameter analyzer in a dry nitrogen atmosphere at room temperature in a dark probe station.

3. Results and Discussion

Figure 2 shows the output characteristics of the OFETs. The drain current (I_D) was monitored while the drain voltage (V_D) was varied at the different gate voltage (V_G). In a low V_D region, a linear relationship between V_D and I_D was realized, suggesting efficient electron or hole injections at interfaces of C₆₀/Al or pentacene/Au. The I_D saturated at a high V_D because the conducting channels were pinched-off. These results indicate that the as-fabricated organic transistors exhibit standard n/p-channel field-effect operations. Table 1 presents the initial FET parameters in this study, where the field-effect mobility (μ) and the V_th were extracted by fitting the curves with the standard equation for I_D in the saturation regime. The estimated μ and other parameters obtained in our experiments are typical values in OFETs using C₆₀ or pentacene as the semiconducting layers [11], [15], [16].

Figure 3 shows the transfer characteristics of OFETs recorded after application of a positive program gate voltage. In all processes, the pulse width of the program voltage is 1 s and the source-drain electrodes were grounded [16]. As can be seen in Fig. 3, the transfer curves positively shifted in both types of OFETs. Indeed, upon application of a high positive program voltage to a gate electrode, electrons are injected from the source-drain electrodes through the semiconducting and CYTOP layers and subsequently trapped at the interface of CYTOP and SiO₂.
cial trapping can be understood due to the electrets property of CYTOP material [17] and has been discussed in detail in our previous works [15], [16]. The trapped electrons then deplete electrons in the n-channel C_{60} or induce additional holes in p-channel pentacene, leading to the \( V_{\text{th}} \) was shifted to more positive voltage. Note that, the trapped electrons at CYTOP/SiO\(_2\) interface remain there without the need for any applied voltage, resulting in a built-in electric field. During the measurements of transfer characteristics, for n-channel C\(_{60}\) OFET, when the applied electric field between the gate electrode and the semiconductor compensates the built-in electric field, the transistor operated similarly to the its initial state. Thus, the off-current of the device is almost unchanged (Fig. 3(a)). Meanwhile, in p-channel OFET, the built-in electric field leads to an increase in the number of holes accumulated in the pentacene channel that is the possible reason for an increase in the off-current (Fig. 3(b)).

We would like to confirm that the control of the \( V_{\text{th}} \) is a reversible phenomenon. After applying a negative erase gate voltage, the trapped electrons were removed from the interface, the \( V_{\text{th}} \) completely returns to the initial position. Figure 4 presents the characteristics of the reversible shifts in the \( V_{\text{th}} \). The change of the \( V_{\text{th}} \) increased as the gate voltage pulse was increasing. Under applications of program/erase gate voltage pulses between 0 and ±55 V (for the n-channel OFET device) and between 0 and ±45 V (for the p-channel OFET device), the \( V_{\text{th}} \) was reversely controlled, from +2.8 to +12.8 V for the n-channel OFET and from –4.4 to +4.6 V for the p-channel OFET. The required program voltage for p-channel device is larger than that for n-channel one (Fig. 4) because the facts that the larger injection barrier at the interfaces of Au/pentacene and electron transport through lower electron-mobility of pentacene [16].

Once the \( V_{\text{th}} \) can be controlled, its stability is the other important factor. In subsequent experiments, we studied the stability of the \( V_{\text{th}} \) after tuning to the new value. It was reported that an investigation of the change in the \( V_{\text{th}} \) of an OFET device can be converted to observation of the change in the \( I_{D} \), and the relationship between the drain currents at initial \( I_{D}(0) \) and at the waiting time \( t \) \( I_{D}(t) \) can be expressed by a stretched-exponential equation [18]:

\[
\frac{I_{D}(t)}{I_{D}(0)} = \exp \left[ -\left( \frac{t}{\tau} \right)^{\beta} \right],
\]

where \( \tau \) is the time constant and \( \beta \) is the dispersion parameter. To apply that knowledge for our devices, after programming, \( I_{D} \) was continuously measured over \( 10^{5} \) s at \( V_{G} = \pm 10 \) V and \( V_{D} = \pm 5 \) V for n-channel and p-channel OFETs (Inset of Fig. 5). The \( \mu \) was found to slightly decrease from 0.40 to 0.35 cm\(^2\) V\(^{-1}\) s\(^{-1}\) for the n-channel OFET and from 0.30 to 0.27 cm\(^2\) V\(^{-1}\) s\(^{-1}\) for the p-channel OFET, this is due to a decrease in the \( I_{D} \). Then, the \( I_{D}(t)/I_{D}(0) \) versus the waiting time were subsequently estimated. As can be seen in Fig. 5, the experimental data fit very well to the calculated results obtained with Eq. (1). The extracted \( \tau \) and \( \beta \) were shown in Table 2 where the \( \tau \) obtained from p-channel device is higher than that of n-channel OFET. This is probably due to, during bias process, the trapped electrons penetrate from the trap site through the pentacene layer slower than through the C\(_{60}\) layer because of lower electron-mobility of pentacene material. Significantly, the obtained \( \tau \) is almost one order of the magnitude larger than that in a normal OFET [18], suggesting a highly stable \( V_{\text{th}} \) obtained in our
For each curve, when an input voltage ($V_{\text{in}}$) was varied from 0 to supply voltage $V_{\text{DD}}$, the output voltage ($V_{\text{out}}$) swings from $V_{\text{DD}}$ to 0 V, indicating a typical inverting operation. The voltage gain, estimated by $-dV_{\text{out}}/dV_{\text{in}}$, increased with increasing the $V_{\text{DD}}$. This trend is similar to that in other circuit systems [14], [19]. The obtained gain can be comparable to that of previous organic inverters [7], [8].

It knows that the $V_{\text{ins}}$ occurs when both $n$-channel and $p$-channel devices are saturated and the $V_{\text{ins}}$ can approximately be given by the following equation [19]:

$$V_{\text{ins}} = \frac{V_{\text{DD}} - |V_{\text{th},n}| + V_{\text{th},p}}{1 + \frac{K_n}{K_p}},$$

where $V_{\text{th},n}$ and $V_{\text{th},p}$ are the threshold voltages for $n$-channel and $p$-channel OFETs, respectively; $K_n = (W/L)\mu_n C_i$; $K_p = (W/L)\mu_p C_i$, in which $C_i$ is the capacitance per unit area of the gate dielectric. Since the $V_{\text{in}}$ and the $\mu$ parameters of the $n$-channel device were different from those of the $p$-channel one as presented in Table 1, our initial inverter did not switch at the theoretical point of $1/2V_{\text{DD}}$ as evidently shown in Fig. 6. Thus, the voltage transfer characteristics should be relocated.

The controllable characteristics of the organic inverter circuit at $V_{\text{DD}}$ of 10 V are shown in Fig. 7, where each $V_{\text{out}}$-$V_{\text{in}}$ relationship was taken after applying a program gate voltage for the $n$-channel or $p$-channel OFETs. The positive shift of $V_{\text{th}}$ in both transistor types caused the switching voltage of inverter $V_{\text{ins}}$ tunes also to more positive voltage value. We would like to note that when applying a program voltage of 50 V for $n$-channel device (shifted $V_{\text{th}} = \sim 10$ V, Fig. 4), the $V_{\text{ins}}$ cannot be observed since it is actually larger than the $V_{\text{DD}}$. The changes in the $V_{\text{ins}}$ as a function of the gate voltage pulse at $V_{\text{DD}}=10$ V are shown in Fig. 8 (opened curves). The $V_{\text{ins}}$ was tuned over a wide range, from 3.3 to more than 10 V after programming the $n$-channel OFET (Fig. 8(a)) and from 3.3 to 6.6 V after programming the $p$-channel OFET (Fig. 8(b)). Interestingly, we found that, at the program voltage of 60 V, the $V_{\text{th}}$ of $p$-channel OFET was placed at $-2.9$ V (Fig. 4(b)), nearly equal to that of the initial $n$-channel device (2.8 V). This leads to the inverter was switched at a voltage of 4.8 V, which is close to the ideal value ($1/2V_{\text{DD}}$). On the other hand, to compare with the measured values, the $V_{\text{ins}}$ of the organic CMOS inverter was theoretically calculated using Eq. (2) and plotted as filled curves in Fig. 8. As can be seen in Fig. 8(a) and (b), the experimental data tend to follow the calculated results with a relatively small error.

An electrical noise immunity of a digital CMOS circuit is quantified as the noise margin ($\text{NM}$) which is the maximum noise signal that can be superimposed on a digital signal without changing the function of the circuit [19]. In order to examine effect of the $V_{\text{th}}$ on the $\text{NM}$, in Fig. 9, we have estimated the $\text{NM}$ at the high ($\text{NM}_{\text{H}}$) and low ($\text{NM}_{\text{L}}$) devices even after tuning to the new value.

In organic CMOS technology, the changeable $V_{\text{th}}$ results in tunable switching voltage of inverter ($V_{\text{ins}}$), which in turn help to adjust the noise margin, avoiding the inadvertent switching of the circuit [5], [9]. To verify that hypothesis, we further studied on a CMOS inverter configuration constructed of $p$-channel pentacene and $n$-channel fullerene devices as discussed above. Figure 6 shows the voltage transfer characteristics and corresponding voltage gain of the organic CMOS inverter at various drain–drain voltages ($V_{\text{DD}}$).
logic levels from the selected voltage transfer characteristics at $V_{DD}=10$ V using the following equations [19]:

$$NM_H = V_{Out_H} - V_{In_H} \tag{3}$$

$$NM_L = V_{In_L} - V_{Out_L} \tag{4}$$

where $V_{In,L}$ and $V_{In,H}$ are the input low and high voltages, $V_{Out,H}$ and $V_{Out,L}$ are the output high and low voltages. The $V_{In,L}$ and $V_{In,H}$ are extracted at the points where the gain

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the program gate voltage of 60 V for p-channel device, the balance between the $\text{NM}_\text{L}$ (3.9 V) and the $\text{NM}_\text{H}$ (4.1 V) is significantly improved as shown in Fig. 9(c). This results in a greater noise immunity [5], [9], i.e., the circuit can operate more reliably.

We would like to stress here that, to balance between the $\text{NM}_\text{L}$ and the $\text{NM}_\text{H}$ at a certain $V_{\text{DP}}$, it is necessary to choose a suitable program gate voltage for $n$-channel or $p$-channel OFETs based on the tendency documented in Fig. 4 in order that results in a symmetrical $V_{\text{th}}$ for both transistor types.

4. Conclusion

In summary, we have demonstrated tunable threshold voltage OFETs and CMOS inverter circuit based on pentacene/C$_{60}$ and a bilayer-dielectric structure of CYTOP and SiO$_2$. The $V_{\text{th}}$ of OFETs can be electrically switched thanks to electron trapping at the interface of CYTOP and SiO$_2$ layers. The stability of the $V_{\text{th}}$ after programming was examined through an investigation of the change in transistor current, which fitted very well with a stretched-exponential equation with a time constant up to $10^6$ s. For organic CMOS inverter circuit, the voltage transfer characteristic was positively shifted after applying the program gate voltages for $n$-channel or $p$-channel elements. The noise margin $NM$ of the inverter was found to be strongly dependent on the $V_{\text{th}}$. Significantly, at a program gate voltage of 60 V, the $V_{\text{th}}$ of $p$-channel OFET was set to be nearly equal to that of the initial $n$-channel device, which provides an excellent balancing between the $\text{NM}_\text{L}$ and the $\text{NM}_\text{H}$ at supply voltage $V_{\text{DP}}$ of 10 V. This in turn results in the maximum electrical noise immunity of the circuits. The experimental data suggest that the organic transistor and inverter based on a bilayer-dielectric structure of CYTOP and SiO$_2$ can be used to fabricate reconfigurable complex organic CMOS integrated circuits.

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References

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