

Title	An Organic Non-volatile Memory Using Photo-conductive Gate Dielectrics
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Citation	
Issue Date	2011-09
Type	Thesis or Dissertation
Text version	none
URL	http://hdl.handle.net/10119/9909
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An organic non-volatile memory using photo-conductive gate dielectrics

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Introduction: Recently, there have been many researches on organic non-volatile transistor memories. However, reducing programming/erasing (P/E) voltages is still the challenge to be overcome. To reduce the P/E voltages, the light is introduced. This dissertation focuses on the organic non-volatile memories using photo-conductive dielectric, poly vinylcarbazole (PVK), with the expect to reduce the P/E voltages by using light. As studied before, PVK is a photo-conductive material. It can generate charges under the light. By using PVK in the transistor as a charge-generator layer under the light, together with a charge trapping layer, CYTOP, as a floating gate, memory effect can occur in the transistor with the assistant of light in P/E processes.

Experiments. This research studied on three structures of memory using PVK. They are shown in Fig. 1. Here, PVK, PVA, PVK:PCBM, and CYTOP layers were fabricated by using spin-coating method. Pentacene and Au layers were fabricated by vacuum evaporation deposition. Channel length is 50 μm , and channel width is 2 mm.

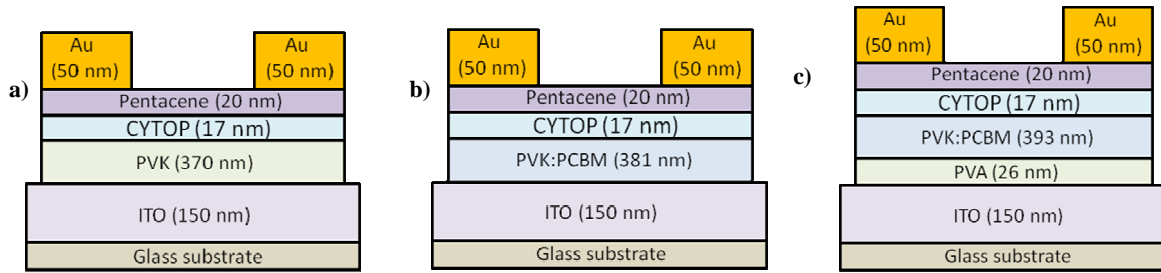


Fig. 1. Structure of devices in this research. a) ITO/PVK/CYTOP/Pentacene/Au, b) ITO/PVK:PCBM/CYTOP/Pentacene/Au, and c) ITO/PVA/PVK:PCBM/CYTOP/Pentacene/Au.

Results and discussions. The device of ITO/PVK/CYTOP/Pentacene/Au has typical output and transfer characteristics of an organic field effect transistor (OFET) (Fig. 2). In this case, PVK layer plays double roles, one is the charge generator and one is the gate insulator. Output characteristics reveal good contact between Au electrodes and pentacene. The linear and saturated regions are quite clear. It has the threshold voltage (V_{Th}) of -11.49 V, the subthreshold swing of 2.04 V/decade. The saturated hole field-effect mobility calculated based on equation $\mu = \frac{2LI_{Dsat}}{WC_i(V_G - V_{Th})^2}$ is 1.7×10^{-2} $\text{cm}^2/\text{V}\cdot\text{s}$ where PVK's capacitance is $5.65 \text{ nF}/\text{cm}^2$. In this equation, L is the channel length, W is the channel width, I_{Dsat} is the saturated drain current, V_G is the gate voltage, V_{Th} is the threshold voltage.

The shift between transfer curves in programed and erased states reveals the memory effect in the device (Fig.3). The device has the memory effect with a high on/off ratio of 8×10^5 by applying the P/E voltages of $\pm 10 \text{ V}$ for 1 s, under a $62.1 \text{ mW}/\text{cm}^2$ light intensity of 365 nm wavelength. The

current is measured under a drain voltage (V_D) of -5 V. The memory has long retention time and good endurance characteristics under dark (Fig.4). This device reveals the non-volatile memory behavior with low P/E voltage.

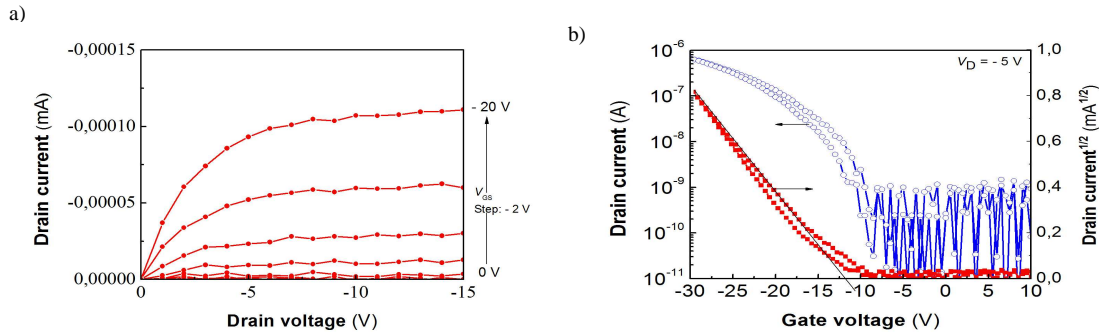


Fig. 2. a) Output and b) transfer characteristics of ITO/PVK/CYTOP/Pentacene/Au.

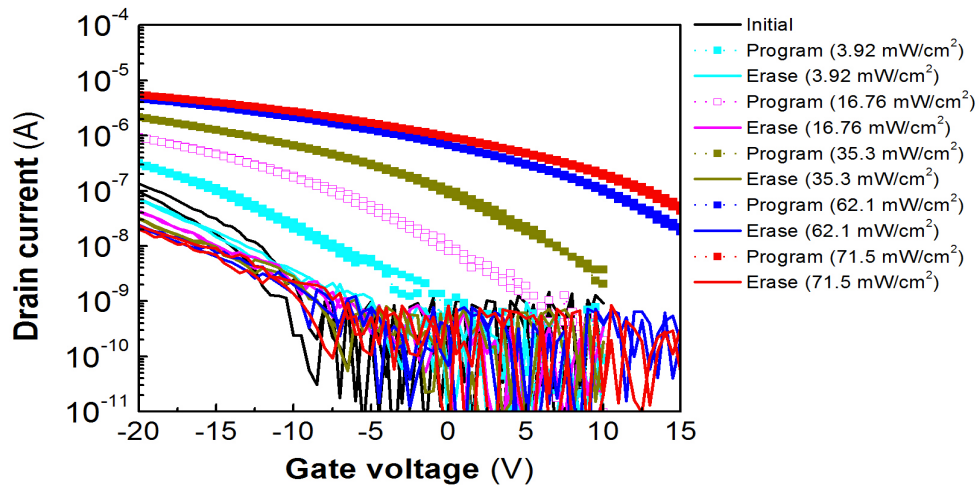


Fig. 3. Programmed and erased transfers. There is a shift between programmed state and erased state. Higher light intensity is applied, larger shift is obtained.

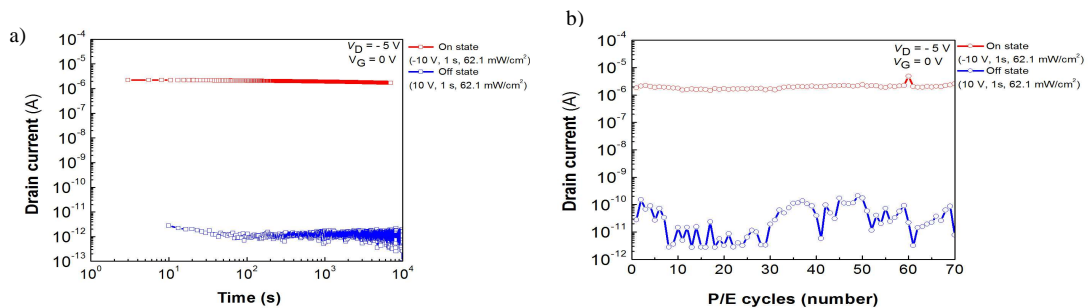


Fig. 4. a) Retention. b) Endurance characteristics of OFET based on ITO/PVK/CYTOP/Pentacene/Au

Although the device using PVK as an insulator layer has high on/off ratio, the required light intensity is high. This phenomenon may be due to the short time of charge-separation in PVK.

To reduce the required light intensity, fullerene derivative [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) was added into the PVK layer in the expect to create the charge separation, and hence, improve the conduction of charge-generator layer under the light. By adding PCBM to PVK with molar ratio of 1:10 between PVK and PCBM, the required light intensity is reduced, from 62.1 mW/cm² to 8.68 mW/cm². The shift between transfer curves in programmed and erased states are shown in Fig.5. To obtain the on/off ratio of 1×10⁶, the P/E voltages of ±10 V is applied for 1 s under 8.68 mW/cm² light intensity (Fig.6).

Device with PVK:PCBM as an gate insulator ($C_i = 4.7 \text{ nF/cm}^2$) also shows good OFET characteristics, with a saturated mobility of $6.2 \times 10^{-2} \text{ cm}^2/\text{V.s}$, a threshold voltage of -12.6 V, and a subthreshold swing of 1.86 V/decade.

Although the required light intensity is reduced, the retention time of memory is not good. The on current in this case decreases very quickly (after 10² s) due to discharge.

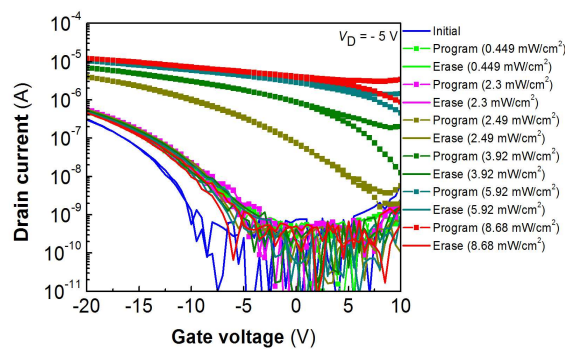


Fig. 5. Programmed and erased transfer characteristics of ITO/PVK:PCBM/CYTOP/pentacene/Au

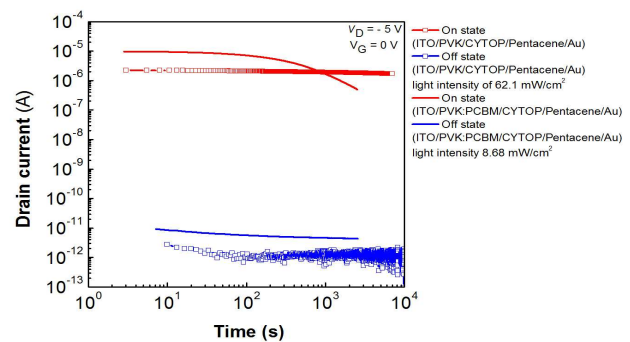


Fig. 6. Retention time of OFET based on ITO/PVK:PCBM/CYTOP/pentacene/Au compared to ITO/PVK/CYTOP/pentacene/Au

To prevent the discharge, a thin PVA layer was added into the transistor as a insulator layer. The device with PVA (26 nm), and molar ratio of 1:10 between PVK and PCBM, shows the good OFET characteristics with a threshold voltage of -9 V, a saturated mobility of $1.9 \times 10^{-2} \text{ cm}^2/\text{V.s}$, and a subthreshold swing of 3.79 V/decade where the insulator capacitance is 4.5 nF/cm^2 . There is large shift between programmed transfer curve and erased transfer curve (Fig.7). This device shows a high on/off ratio of 6.6×10^5 , with the P/E voltages of ±10 V applied for 1 s under a 8.68 mW/cm² light intensity. Long retention time and good endurance characteristics are demonstrated (Fig. 8). The dependence of on and off currents on the P/E times, on P/E voltages and on the light intensity were investigated (Fig.9). To obtained the on/off ratio of 10⁵, required P/E voltages are below ±10 V, P/E

times are below 1 s and light intensity is below 8.68mW/cm^2 . The on and off currents are measured at the drain voltage of -5 V and gate voltage of 0 V .

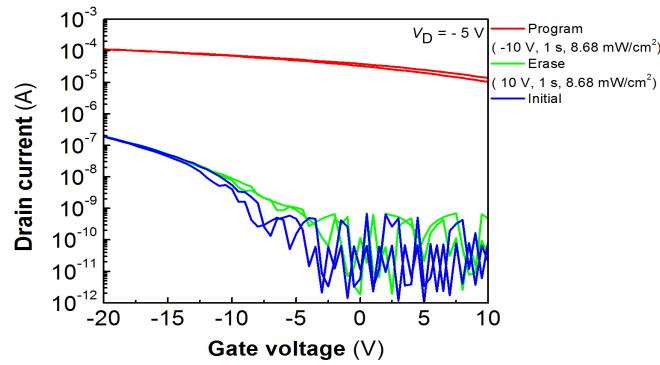


Fig. 7. Programmed and erased transfer characteristics.

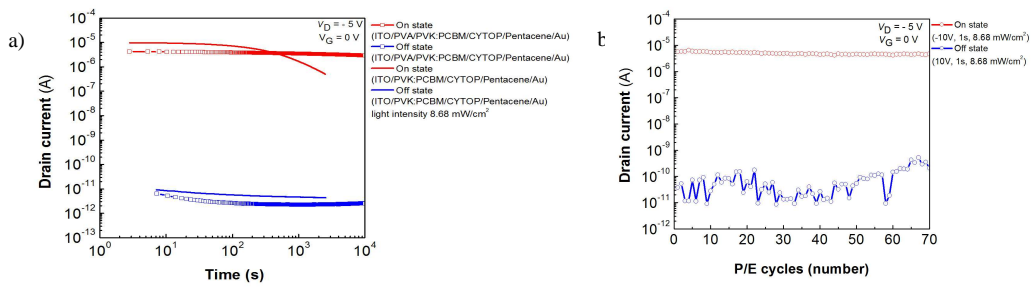


Fig. 8. a) Retention characteristic of OFET based on ITO/PVA/PVK:PCBM/CYTOP/Pentacene/Au, compared with the retention time of OFET based on ITO/PVK:PCBM/CYTOP/Pentacene/Au. b) Endurance

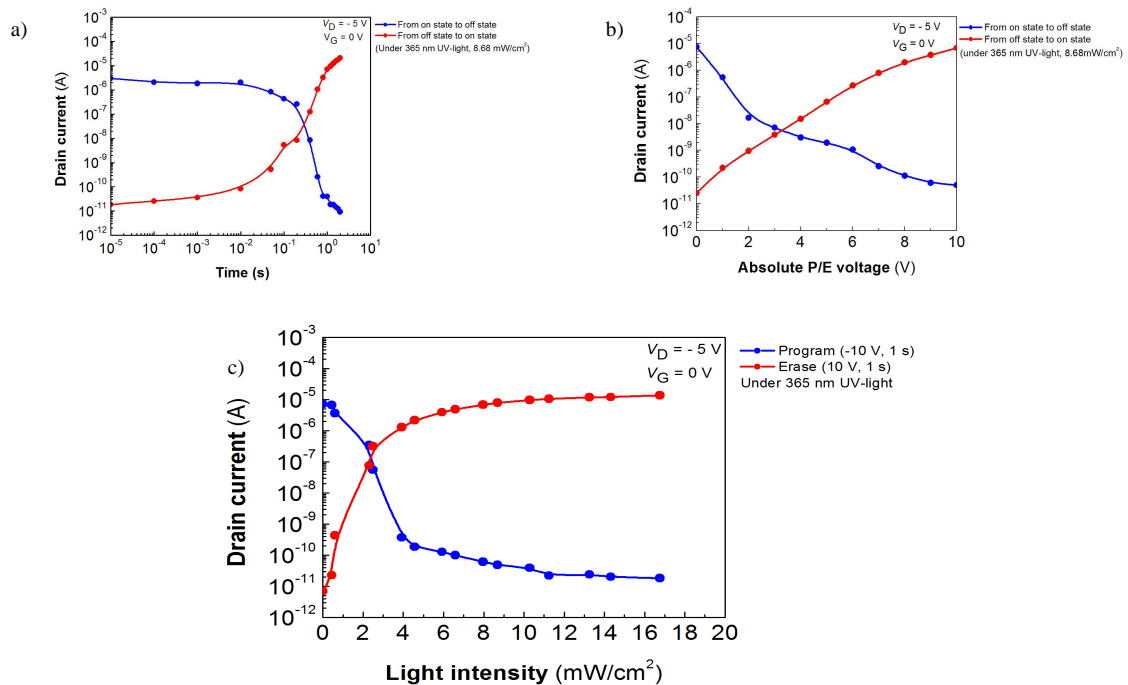


Fig. 9. a) On and off currents according to the P/E time. b) On and off current according to the P/E voltages. c) On and off current according to the light intensity.

To understand the memory effect in above devices, the sandwich devices of ITO/PVK/Au and ITO/PVK:PCBM/Au were fabricated. The current density-voltage (J - V) curves of these devices were investigated under the different wavelengths of light. The number of photo-electrons according to the wavelength of light was calculated from the J - V data (Fig.9 and Fig. 10). In both cases, the ratio between the number of photo-electrons and the number of coming photons shows the maximum at the wavelength of 340 nm, which is consistent with the maximum of PVK's absorbance. To confirm, the on/off ratio of memory based on ITO/PVA/PVK:PCBM/CYTOP/Pentacene/Au was measured under the different wavelengths. The dependence of on/off ratio on the wavelength of light also has the maximum at peak of the PVK absorbance (at 340 nm) (Fig.11). The results can indicate the roles of PVK and PCBM in these memories.

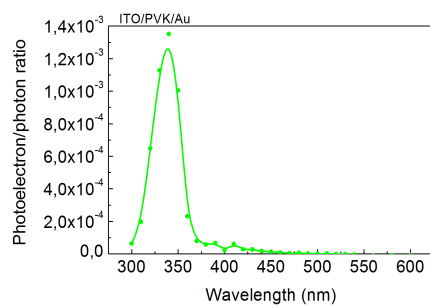


Fig. 9. Photoelectron/photon ratios in device ITO/PVK/Au depending on wavelengths of light.

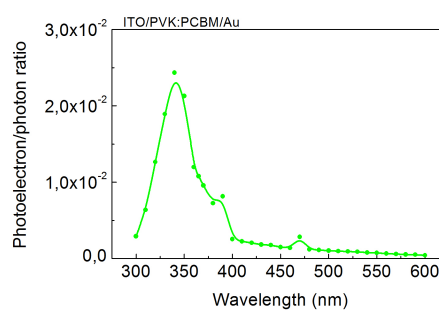


Fig. 10. Photoelectron/photon ratios in device ITO/PVK:PCBM/Au depending on wavelengths of light.

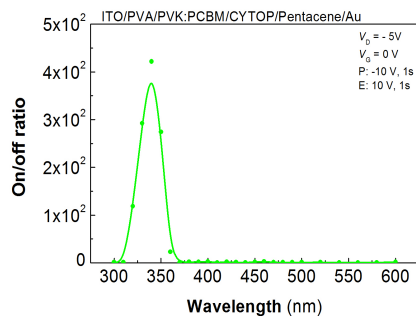


Fig. 11. On/off ratio of device ITO/PVA/PVK:PCBM/CYTOP/pentacene/Au depending on the wavelength of light.

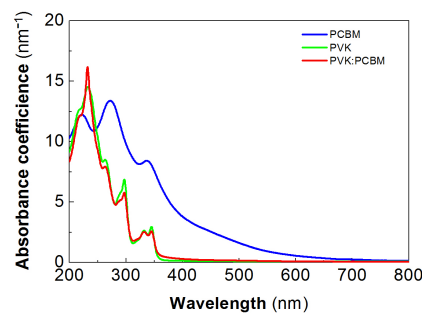


Fig. 12. Absorption spectra of PVK, PCBM, and PVK:PCBM blend film.

The memory effect in transistors using PVK is due to the charge generation of PVK under the light. Photo-electrons are generated by effect of light on PVK. Then, they are moved into the CYTOP layer by electric field. These electrons are trapped in the CYTOP layer, resulting in memory effect. However, the required light intensity is quite high in the case of OFET memory using only PVK. Adding PCBM into PVK layer can decrease the required light intensity. Because PCBM and PVK have different energy levels related to each other, therefore, charge can transfer between two material

molecules, making the charge separation easily. Hence, more photo-electrons can come to the CYTOP layer, increasing the on/off ratio with lower light intensity.

Figure 13 describes the charge transfer between PVK and PCBM when blend film is under the light. Electrons in the valence band of PVK molecules are excited to the conduction band of PVK molecules. These electrons relax to the conduction band of PCBM, making the long time of charge separation.

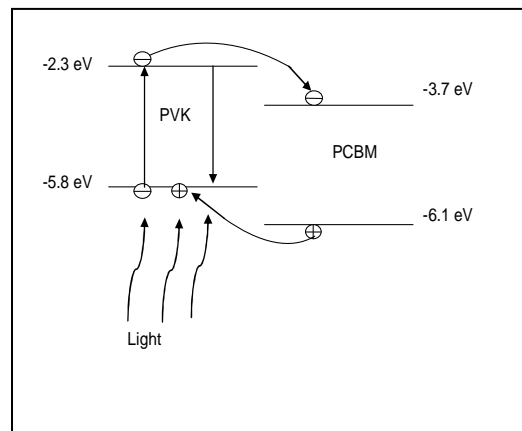


Fig. 13. Energy levels of PVK and PCBM

Conclusion. The memory based on PVK as a gate insulator shows the non-volatile behavior, but it needs high light intensity. The memory using PVK:PCBM as gate insulator layer has high on/off ratio with lower light intensity, but it has not long retention time. The memory using PVK:PCBM and using PVA insulator layer has high on/off ratio (6.6×10^5) with low P/E voltages (± 10 V) applied for 1 s under low light intensity (8.68 mW/cm^2) of 365 nm wavelength. It has good retention time and good endurance, can be demonstrated with the assistant of light. The memory also can be used in photo-applications and EPROM.