



Stabilization of space charge polarization in ion-dispersed gate dielectric layer of organic transistors by ultraviolet illumination for write-once read-many memory



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ABSTRACT

In this work, a new type organic field effect transistor (OFET) based write-once read-many memory (WORM) device was developed. The device uses an ultraviolet (UV) cross-linkable matrix polymer mixed with ionic compounds to form an ion-dispersed gate dielectric layer. Under an applied gate voltage bias, migration of cations and anions in opposite directions forms space charge polarization in the gate dielectric layer, resulting in change of the electrical characteristics. It is shown that, with UV illumination to cross-link the matrix polymer, the formed space charge polarization can be stabilized. Therefore, the OFET can be operated as a WORM with the applied voltage bias to define the polarization and in turn the stored data, and the UV illumination to stabilize the stored data.

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1. Introduction

Organic non-volatile memories (NVMs) have attracted tremendous research interest for their potential of realizing data storage at very low cost on arbitrary substrates such as plastic and paper [1]. In the past, electrically reprogrammable organic NVMs have been of the research focus [2]. Another important class of NVMs is the write-once read-many memory (WORM), which can be programmed only once and then be accessed for many times during the whole product life [3]. The organic WORM would be desired for a wide range of disruptive electronic applications in item identification, anti-counterfeiting, and brand protection, which are cost-sensitive and require the stored data unable to be modified any more once it is programmed. WORMs are normally implemented by fuses or anti-fuses with conductive line or metal/insulator/metal structures [4–6], which, however, require large current for the programming and are also difficult for realizing high density level memories, since for two terminal WORMs array, extra attention needs to be paid for avoiding cross-talk by adding a rectifying diode to each cell [7]. Transistor based memories are appealing since they allow direct integration of the memory element with the transistor circuits, and can potentially provide higher density

level. However, despite of plenty of studies on organic field effect transistor (OFET) based reprogrammable NVMs with various methods such as floating gate [8], polymer electret [9] or ferroelectric dielectric [10], there is very few work on OFET based WORMs.

Our previous work shows that, for OFETs with an ion-dispersed polymer gate dielectric layer, the bulk polarization of the gate dielectric can be modulated by the applied gate voltage bias to adjust the electrical characteristics of the OFETs [11,12]. However, when the external voltage bias is removed, the formed polarization will relax gradually, and the electrical characteristics tend to recover to its original condition. Based on the same device architecture, this work uses an ultraviolet (UV) cross-linkable matrix polymer poly (vinyl cinnamate) (PVC) to replace the previous poly-methylmethacrylate (PMMA) for forming the ion-dispersed gate dielectric layer. It is found that, by UV cross-linking of the matrix polymer, the formed space charge polarization at an applied voltage bias can be stabilized. As a result, the OFET device can be operated as a WORM, being programmed by applying a voltage bias to define the polarization and in turn the stored data, followed by UV light illumination to stabilize the stored data.

2. Experimental

The fabricated bottom-gate top-contact device structure, and the chemical structures of the matrix polymer PVC and the

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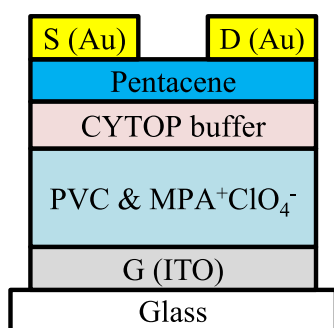
ionic compound 10-methyl-9-phenylacridinium perchlorate ($\text{MPA}^+\text{ClO}_4^-$) used for the ion-dispersed gate dielectric layer are shown in Fig. 1. Patterned indium tin oxide (ITO) on glass substrate was used as the gate electrode. PVC was dissolved in N, N-Dimethylformamide (DMF) at a concentration of 100 mg/mL, and then blended with $\text{MPA}^+\text{ClO}_4^-$ at a molar ratio of 50:1. The mixed solution was spin-coated onto the sample at 3000 rpm for 1 minute, followed by heating at 100 °C for 1 h, forming a 395 nm thick film on the ITO gate electrode. Then a 79 nm thick dielectric CYTOP film was deposited as the buffer layer by spin-coating to prevent possible chemical doping of $\text{MPA}^+\text{ClO}_4^-$ into the semiconductor layer. The 50 nm thick pentacene semiconductor layer was thermally evaporated at a deposition rate of 0.02 nm/s. Then the 60 nm gold (Au) source and drain electrodes were deposited by thermal evaporation through a shadow mask at the deposition rate of 0.03 nm/s to define the channel length (L) and width (W) of 50 μm and 2000 μm , respectively. Finally, the sample was encapsulated with a glass lid in nitrogen environment for the following tests.

The electrical properties of the devices were characterized with a Keithley 4200 Semiconductor Characterization System in a nitrogen-filled glove box at room temperature. The capacitance measurement of the dielectric was performed using an Agilent 4284A LCR meter. The infrared absorption spectra were recorded with a Thermo Scientific Nicolet 6700 Fourier transform infrared spectrometer. The UV-Vis absorption spectra were recorded with a JASCO V-570 spectrophotometer. The thicknesses of the films were measured with a Keyence VN-8000 atomic force microscope. The atomic force microscope (AFM) images of the surfaces of CYTOP and pentacene films were taken with a SII SPA-400 atomic force microscope. The ion-dispersed PVC film was illuminated through a commercial UV light source (Sen Lights Handy100) with the peak wavelength and intensity of 365 nm and 6.5 mW/cm², respectively.

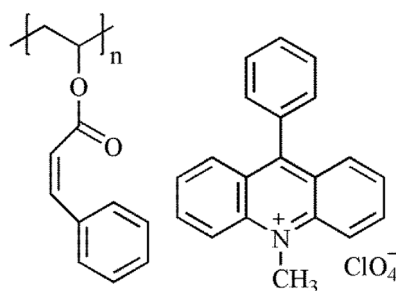
3. Results and discussion

Fig. 2 shows the measured transfer (I_D - V_{GS}) and output (I_D - V_{DS}) electrical characteristics of the as-fabricated devices, indicating typical field-effect transistor behaviors. The devices present a threshold voltage (V_{th}) of about -50 V, and a carrier mobility of around 0.12 cm²/(V s). The V_{th} value was estimated through a linear fit of the square root of I_D versus V_{GS} plot and its intercept at x -axis as illustrated in Fig. 2(a). The carrier mobility was calculated by fitting the plot of the square root of I_D versus V_{GS} with the following equation:

$$I_D = \frac{\mu C_G W}{2L} (V_{GS} - V_{th})^2 \quad (1)$$



(a)



(b)

where L is the channel length, W is the channel width and C_G is the gate dielectric capacitance per unit area. C_G was measured to be 4.5 nF/cm² at 20 Hz. The AFM image of the CYTOP film in Fig. 2(c) reveals a smooth surface with a root-mean-square roughness of about 0.5 nm. The pentacene film has relatively small grain sizes as shown in Fig. 2(d), and thus the devices present relatively low mobility. The crystalline quality of the pentacene film and in turn the device mobility can be improved by optimizing the deposition process. In this work, since the objective is to investigate the effects of UV crosslinking on stabilization of the space charge polarization for the OFET based WORMs, the mobility will not influence the conclusion to be made.

In previous work, it was found that, with an applied external electrical field, the migration of MPA^+ and ClO_4^- ions in the PMMA matrix polymer in opposite directions formed space charge polarization in the gate dielectric [11]. As a result, there was a shift of the electrical characteristics of the device with the direction and the magnitude depending on the polarity and strength of the applied electrical field. However, after removal of the external voltage bias, the ions tended to migrate back and the space charge polarization would relax gradually. Therefore, the electrical characteristics of the devices would recover to the original state.

In this work, with UV cross-linkable PVC as the matrix polymer dielectric to replace PMMA, the feasibility of stabilizing the formed space charge polarization by cross-linking the matrix polymer was investigated. The cross-linking process was conducted by UV illumination for 100 min through the back side of the sample, as shown in the inset of Fig. 3(a). From the measured Fourier transform infrared spectroscopy (FTIR) of PVC in Fig. 3(a), it can be observed that the absorption peak intensity around 1637 cm⁻¹, representing the amount of C=C bonding, decreases over the UV illumination time. The results confirm cross-linking of PVC film under UV illumination [13]. The observed shift of the C=O peak under UV illumination in Fig. 3(a) is attributed to the breaking of C=O bonds [13].

After UV illumination for 100 min, the weak absorption peak around 1637 cm⁻¹ still appears, indicating that the PVC film was not fully cross-linked with existing of small amount of C=C bonding. The main reason is that the ITO/glass substrate absorbs most of the UV light with wavelength less than 300 nm, while the required wavelength for cross-linking PVC is in the range of 250–300 nm [13], as shown in Fig. 3(b).

Before applying gate voltage bias to form space charge polarization, the influence of UV exposure on device performance was evaluated. The measured gate dielectric capacitance per unit area and the electrical characteristics of the device before and after UV exposure are nearly identical, as shown in Fig. 3(c) and (d). The results reveal that the UV illumination has negligible influence

Fig. 1. (a) The schematic diagram of the fabricated OFETs. (b) The chemical structures of the PVC matrix polymer (left) and the ionic compound $\text{MPA}^+\text{ClO}_4^-$ (right), which are used to form the ion-dispersed gate dielectric layer.

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