Supporting Information

Non-volatile Organic Transistor Memory Devices Using the Poly(4-vinylpyridine)-based Supramolecular Electrets

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Experimental

Materials

Poly(4-vinylpyridine) (P4VP) (Mw~60000), Phenol, 2-Naphthol, and 2-Hydroxyanthracene were purchase from Aldich.

Sample preparation

The complexes of P4VP(Phenol)$_x$ (P4VP(2-Naphthol)$_x$ and P4VP(2-Hydroxyanthracene)$_x$) with different molar ratio x of Phenol (2-Naphthol and 2-Hydroxyanthracene) to the repeating unit of P4VP were prepared from chloroform solution. In each case, Phenol (2-Naphthol and 2-Hydroxyanthracene) and chloroform were first mixed together until a clear solution was obtained. P4VP was subsequently added, followed by mechanical stirring for about 12 h at room temperature.

Fabrication of the Pentacene Memory Devices.

The transistor-type memory devices based on a pentacene thin film were fabricated on a wafer with a thermally grown 300-nm thick SiO_2 dielectric on highly doped *n*-type Si as a gate electrode. The solution of P4VP(Phenol)$_x$, P4VP(2-Naphthol)$_x$, or P4VP(2-Hydroxyanthracene)$_x$ in chloroform was spin-coated at 1000 rpm for 60 s on a wafer. Thereafter, the polymer thin films were dried under vacuum (10^{-6} torr) at 100 ^oC for 1 h to remove residue solvents. The thickness of the prepared thin film was estimated to be 65~70 nm. The thin film of pentacene was prepared by thermally deposition with a deposition rate of 0.3~0.4 nm s^{-1} at 90 ^oC under vacuum (10^{-7} torr) to form a 50-nm-thick film. The top-contact source and drain electrodes were defined by 80 nm-thick gold through a regular shadow mask, and the channel length (L) and width (W) were 50 and 1000 μm, respectively. The current-voltage (*I–V*) characteristics of the devices were measured by using a Keithley 4200-SCS semiconductor parameter analyzer in a N_2-filled glove box.

Characterization

Atomic force microscopy (AFM) measurements were obtained with a NanoScope IIIa AFM at room temperature. Commercial silicon cantilevers with typical spring constants of 21-78 Nm^{-1} was used to operate the AFM in tapping mode.

The morphology of the thin film was determined using a field emission scanning electron microscope (SEM, JEOL JSM-6330F). The FE-SEM images were taken using a microscope operated at an accelerating voltage of 10 kV. Before imaging, the samples were sputtered with Pt.Infrared spectra were obtained using a Bruker VECTOR22 FTIR spectrometer. Samples were prepared by casting one drop of the chloroform solution (~10 mg/ml) directly on potassium bromide crystals. The FTIR measurements were taken at room temperature. The electrical characterization of the memory device was performed by a Keithley 4200-SCS semiconductor parameter analyzer in a glove box. For the capacitance measurement, metal-insulator-semiconductor (MIS) structure was fabricated by depositing gold electrodes on the polymer-coated n-type Si(300) wafers. The capacitance of the bilayer dielectrics was measured on the MIS structure using Keithley 4200-SCS equipped with a digital capacitance meter (model 4210-CVU).

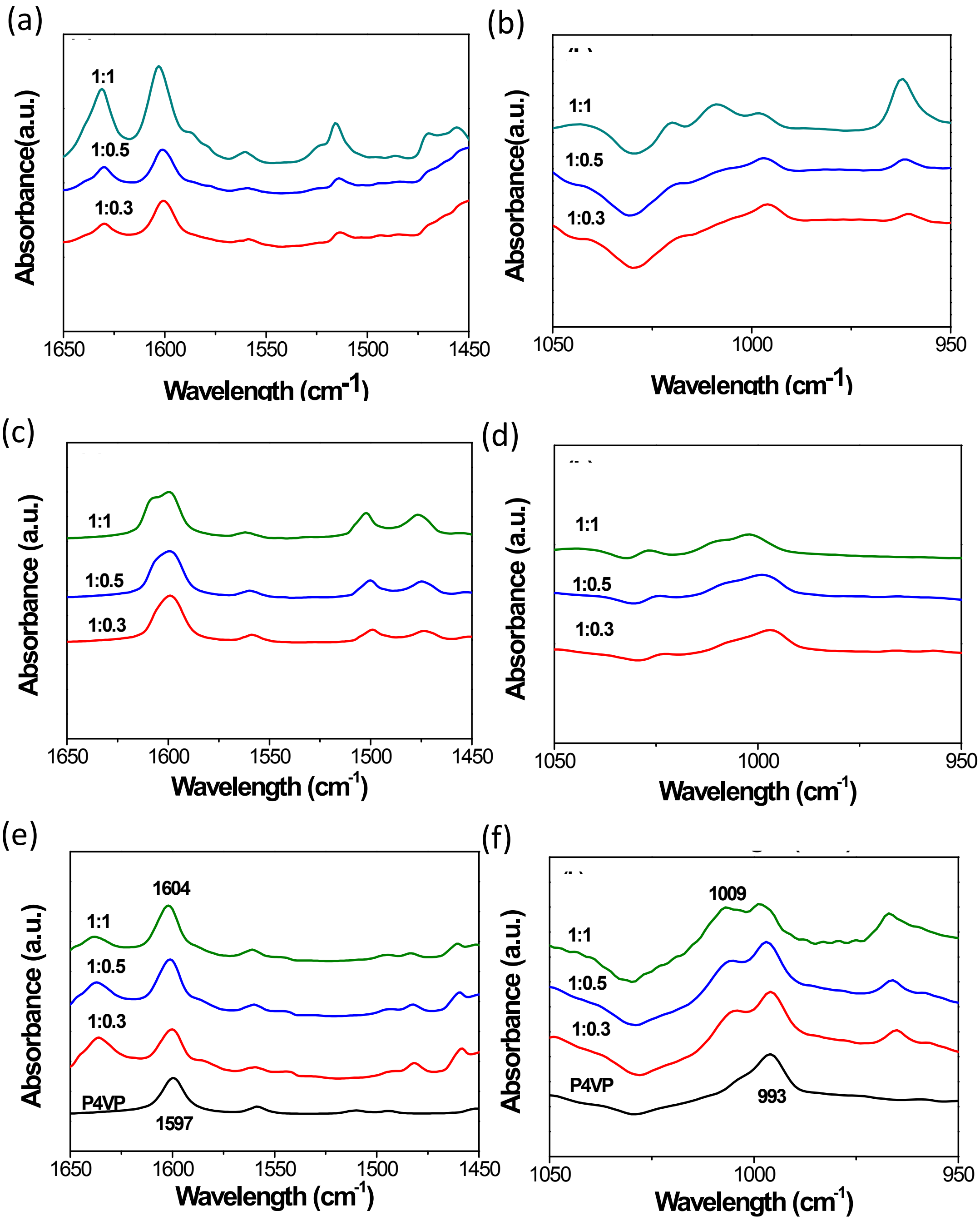

Figure S1. FTIR spectra of (a, b) P4VP(Phenol)$_x$, (c,d) P4VP(2-Naphthol)$_x$ and (e,f) P4VP(2-Hydroxyanthracene)$_X$: (a, c, e) 1450-1650cm^{-1} region; (b, d, f) 950-1050 cm^{-1} region, respectively.

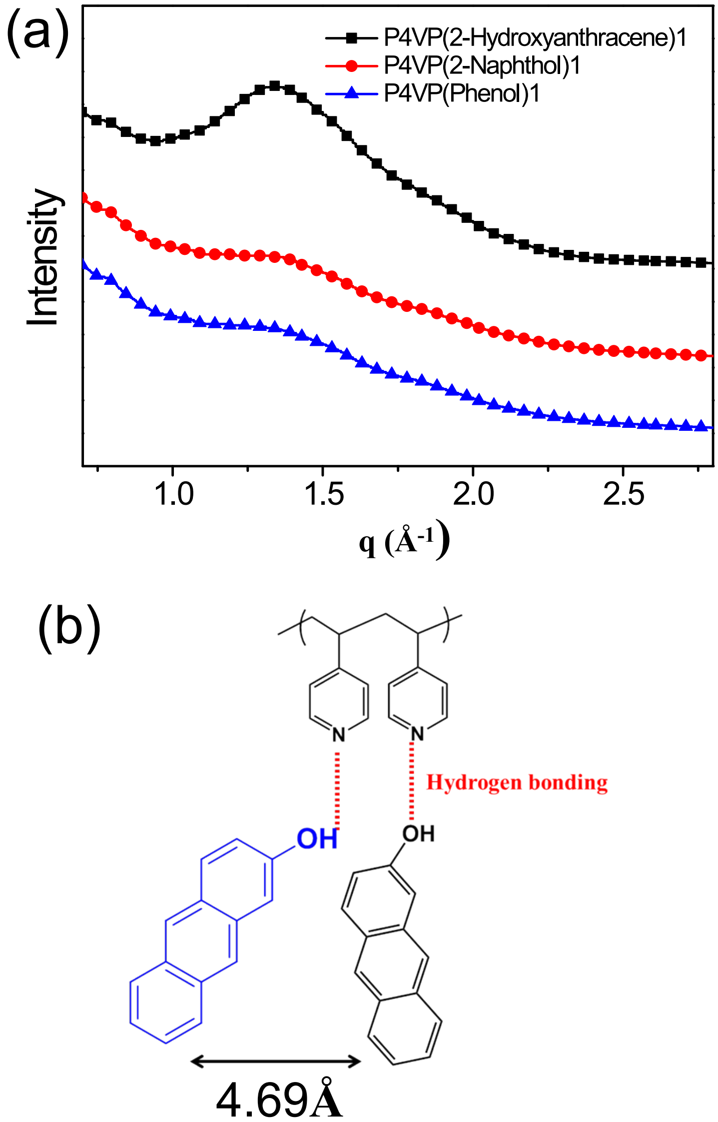

Figure S2. X-ray diffraction diagrams of the pentacene thin film with supramolecular polymer as electrets

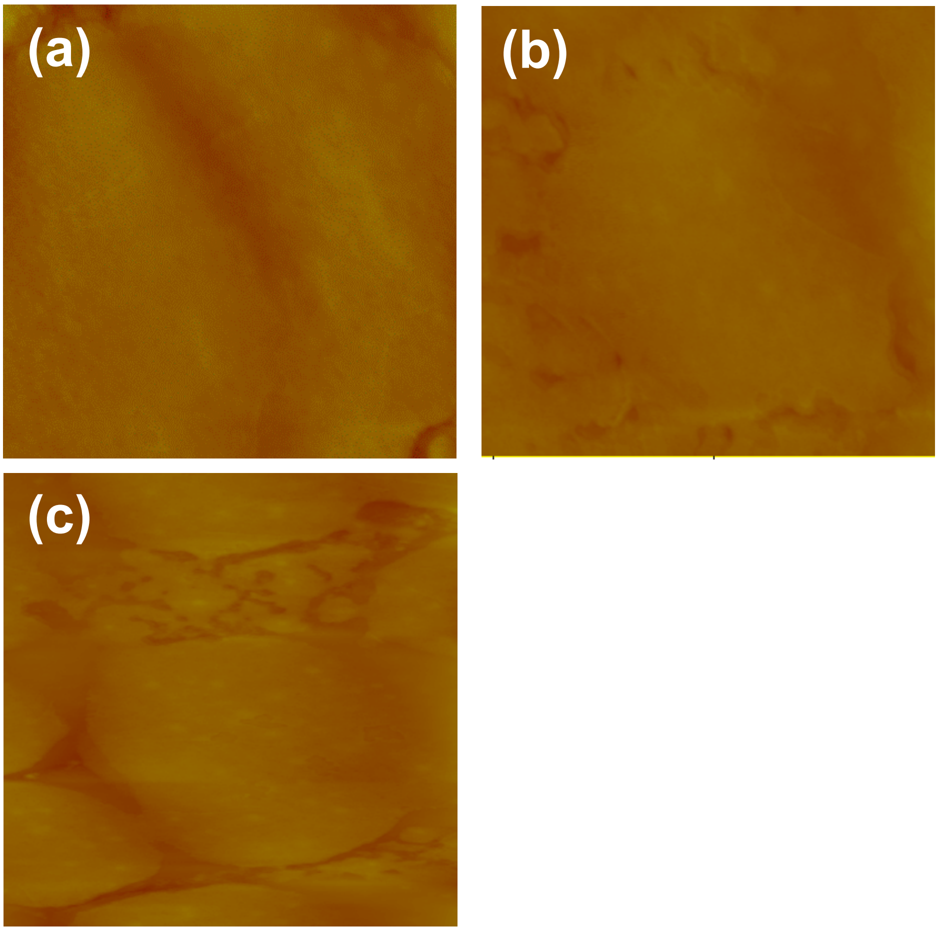

Figure S3. AFM topographic images of (a) P4VP(2-Hydroxyanthracene)$_{0.3}$, (b) P4VP(2-Hydroxyanthracene)$_{0.5}$, and (c) P4VP(2-Hydroxyanthracene)$_{1}$ on 3 μm x 3 μm area.

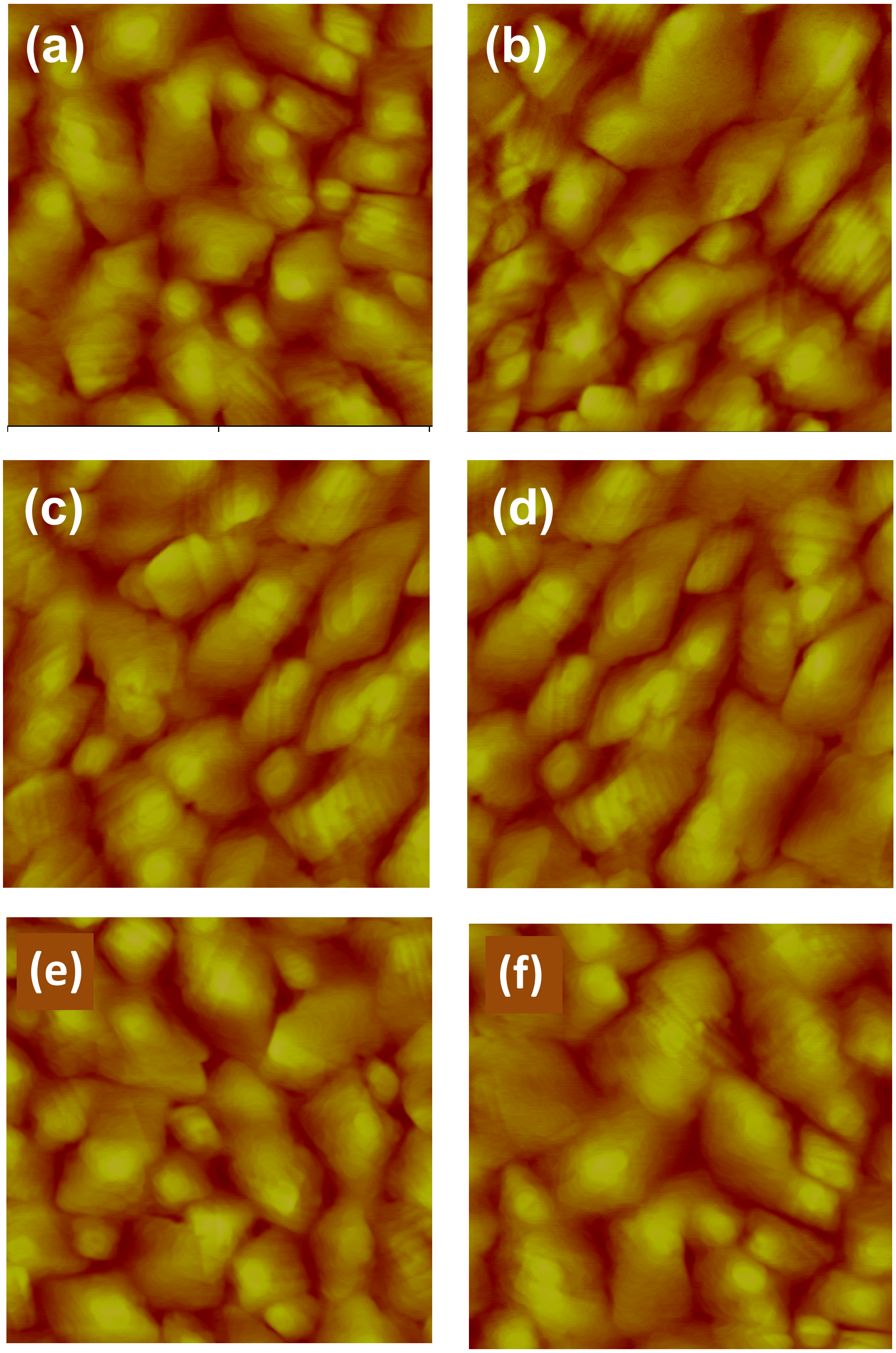

Figure. S4. AFM topographic images of Pentacene on P4VP surface with different supramolecular electrets: (a) P4VP, (b) P4VP(Phenol)$_{1}$, (c) P4VP(2-Naphthol)$_{1}$, (d) P4VP(2-Hydroxyanthracene)$_{1}$, (e) P4VP(2-Hydroxyanthracene)$_{0.3}$ and (f) P4VP(2-Hydroxyanthracene)$_{0.5}$, on 3 μm x 3 μm area, respectively.

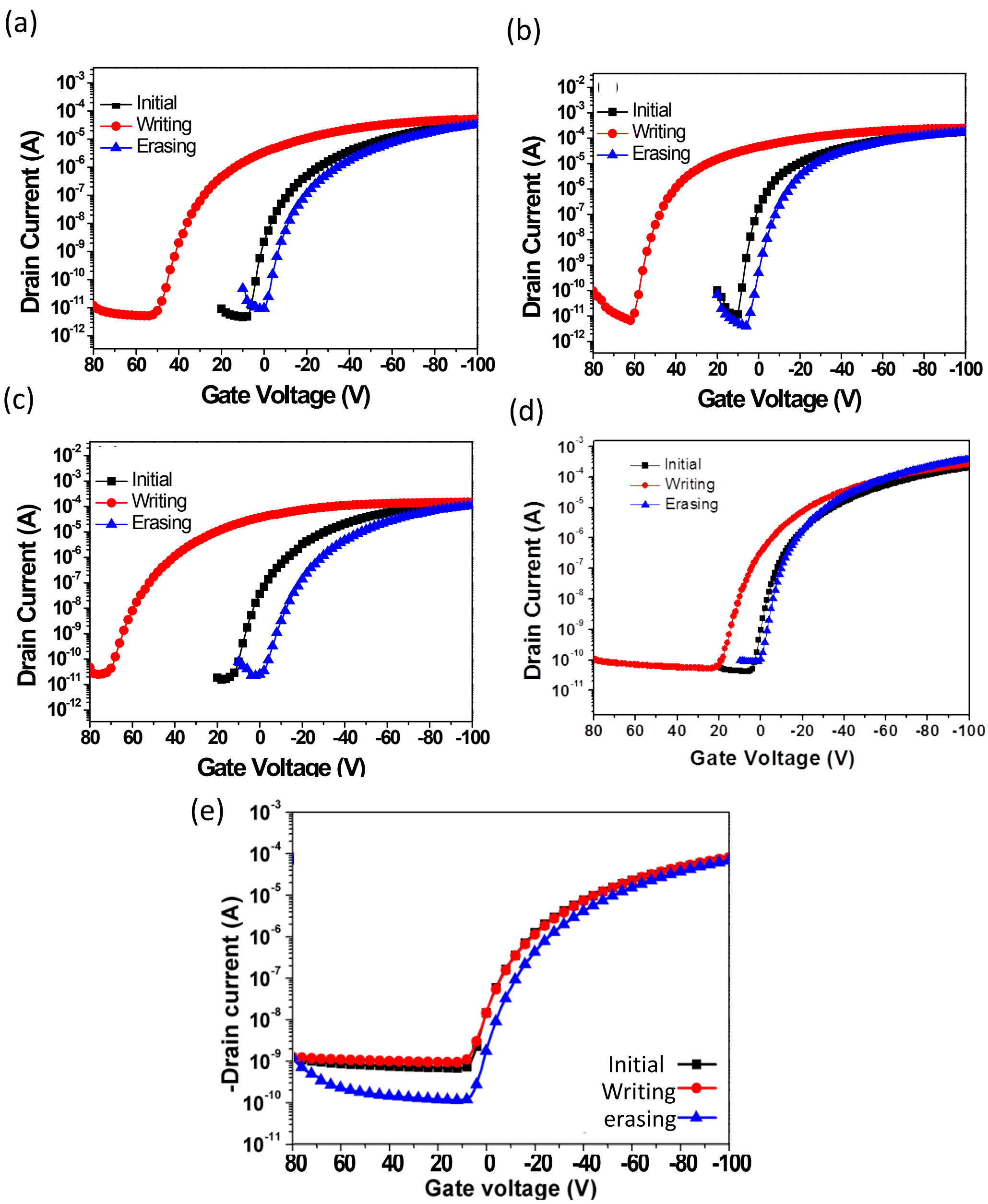

Figure S5. Shifts in transfer curves for Pentacene OFET memory device with (a) P4VP(2-hydroxyanthracene)$_{0.3}$, (b) P4VP(2-hydroxyanthracene)$_{0.5}$, (c) P4VP(2-hydroxyanthracene)$_{1}$, (d) pure 2-hydroxyanthracene, and (e) P4VP-only as polymer electrets, respectively.

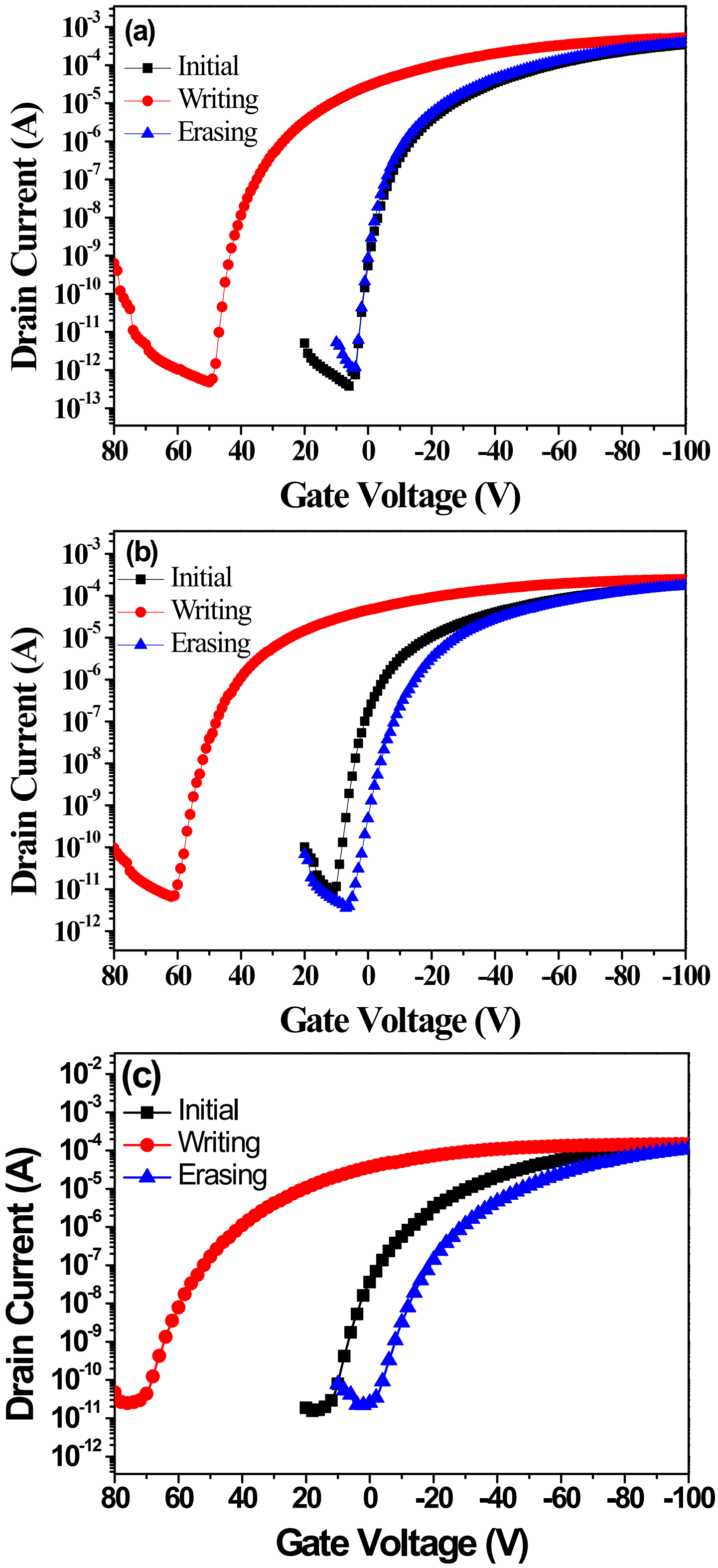

Figure S6. Shifts in transfer curves for Pentacene OFET memory device with (a) P4VP(Phenol)$_{1}$, (b) P4VP(2-Naphthol)$_{1}$ and (c) P4VP(2-Hydroxyanthracene)$_{1}$ as polymer electrets.

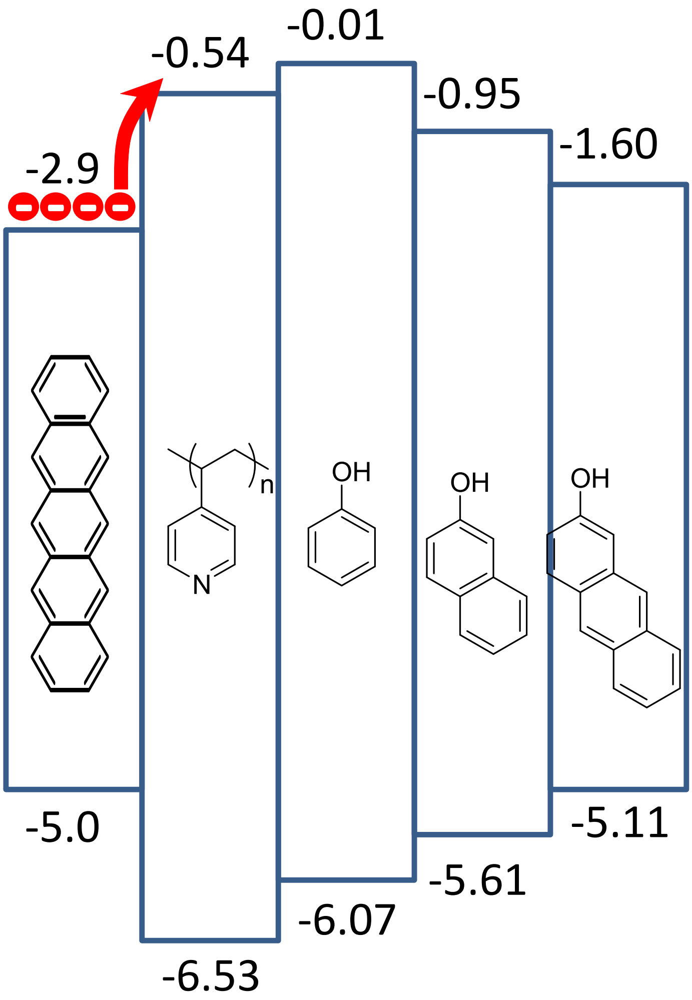

Figure S7. Energy diagram of pentacene/supramolecular electrets and the schematic charge trapping mechanism. Note that the energy levels of P4VP, phenol, 2-naphthol and 2-hydroxyantracene were calculated by Gaussian 03 using density functional theory (DFT) method and Becke's three-parameter functional with the Lee, Yang, and Parr correlation functional method (B3LYP) with 6-31G.

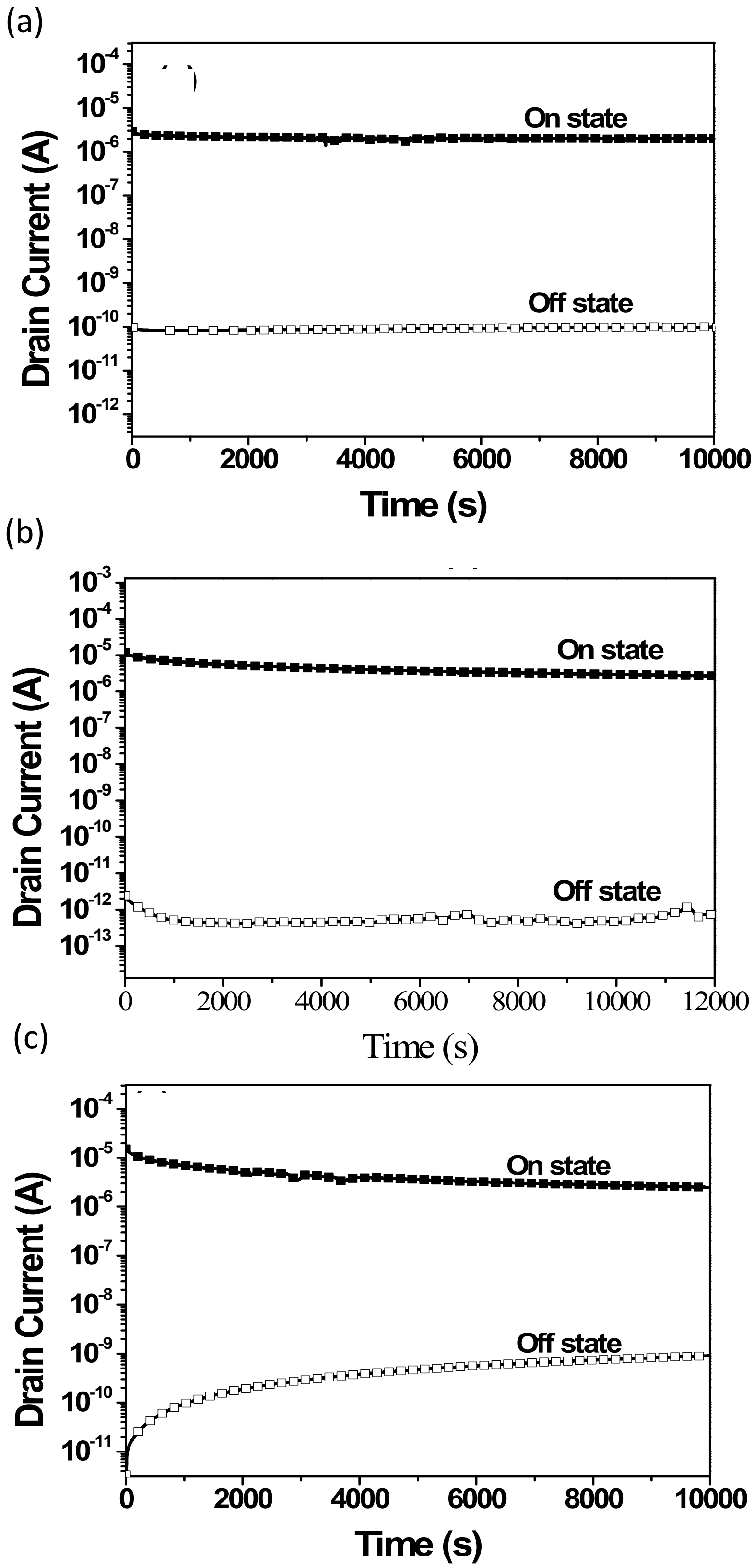

Figure S8. Retention time testing of the OFET memory devices based on Pentacene thin film with (a) P4VP(phenol)$_1$, (b) P4VP(naphthol)$_1$, and (c) P4VP(2-hydroxyanthracene)$_1$ as polymer electrets, respectively.

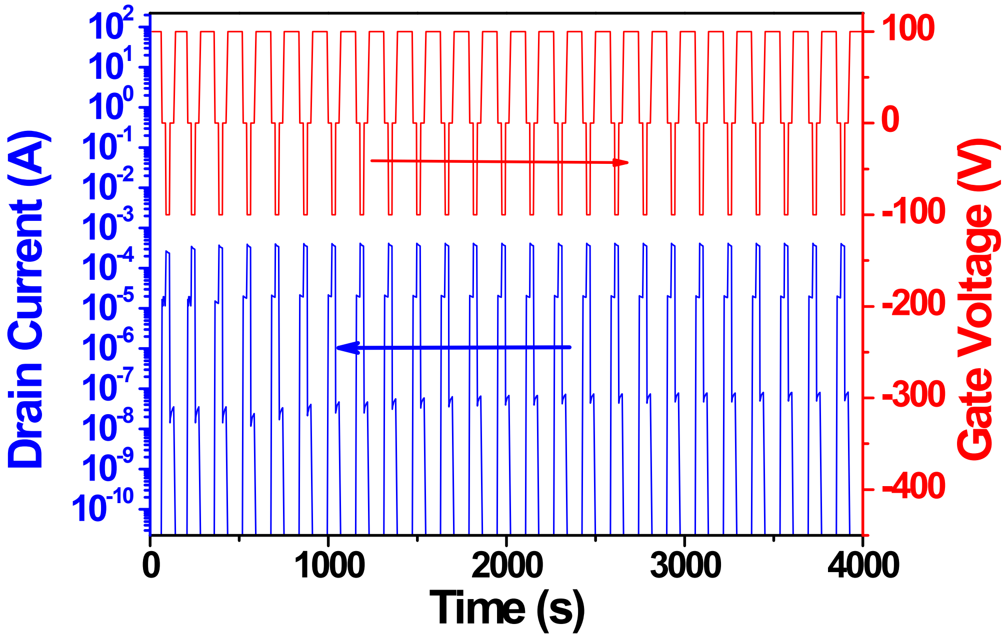

Figure S9. Reversible current response to the WRER cycles of Pentacene OFET memory device with P4VP(2-Naphthol)$_1$ as electret. The drain current was measured at V_d =100V. The writing, reading and erasing were at the gate voltages of -100, 0 and 100 V, respectively.