Electronic Supplementary Information for

Adjustment of charge trap number and depth in molecular backbone

to achieve tunable multilevel data storage performance

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Figure S1. The cross-section profile of TPAAC 1-based sandwich-structure memory device.



Figure S2. Current-voltage (*I-V*) curves of the three molecules-based memory devices under multiple voltage loops.



Figure S3. Stability test of the memory device in all states under constant voltage stress or stimulus by read pulses at -1 V. The insets show the pulse shape.



Figure S4. X-ray diffraction (XRD) patterns of the thin films deposited on ITO substrate for the three molecules. It is shown that the three molecules all revealed high-intensity diffraction peaks, indicating that close packing between the neighboring molecules and highly ordered crystalline films were formed due to the strong intermolecular π - π stacking.



Figure S5. Tapping-mode AFM height images $(5 \times 5 \mu m^2)$ of the thin films vacuum-deposited on ITO substrate and corresponding cross-section profiles for TPAAC **1** (a, d), TPAVC **2** (b, d), and TPAAH **3** (c, f), respectively.



Figure S6. (a) UV-vis absorption spectra in THF solution and thin films deposited on quartz substrate of TPAVH **4**. (b) Cyclic voltammogram in 0.1 M TBAP/CH₂Cl₂ solution of TPAVH **4**. The ferrocene/ferrocenium redox couple is used as a standard (-4.8 eV, the onset potential is 0.57 eV). A scan rate of 100 mV s⁻¹ was used.



Figure S7. HOMO and LUMO energy levels for the two functional fragments (benzyl cyanide moiety and azobenzene chromophore).