Electronic Supplementary Information

Tunable Electrical Memory Characteristics of Brush Copolymers Bearing Electron Donor and Acceptor Moieties

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Fig. S1. Synchrotron 2D GISAXS patterns of 40 nm thick polymer films measured with $\alpha_i = 0.160^\circ$ at 25 °C: (a) PVPK₅BEOXD₅; (b) PVPK₂BEOXD₈.

Fig. S2. Synchrotron 2D GIWAXS patterns of 40 nm thick polymer films measured with $\alpha_i = 0.183^\circ$ at 25 °C: (a) PVPK₅BEOXD₅; (b) PVPK₂BEOXD₈.



Fig. S3. Synchrotron XR profles of polymer films (ca. 40 nm thick) in contact with silicon substrate, Al bottom electrode, and Al top electrode (ca. 10 nm thick): (a) PVPK₅BEOXD₅; (b) PVPK₂BEOXD₈. The symbols are the measured data and the solid line represents the fit curve assuming a homogeneous electron density distribution within the film. The inset shows a magnification of the region around the two critical angles: $\alpha_{c,f}$ and $\alpha_{c,s}$ are the critical angles of the film and the substrate (silicon substrate or Al electrode), respectively.



Fig. S4. Synchrotron XR analysis of PVPK₈BEOXD₂ films (ca. 40 nm thick) in contact with silicon substrate, Al bottom electrode, and Al top electrode (ca. 10 nm thick): (a) electron density profile of the polymer film coated on the Al bottom electrode supported with a silicon (Si) substrate, which was obtained by analysis of the data in Fig. 4a; (b) electron density profile of the polymer film in the sample specimen of Si/Polymer/Al structure, which was obtained by analysis of the data in Fig. 4a.



Fig. S5. Synchrotron XR analysis of PVPK₅BEOXD₅ films (ca. 40 nm thick) in contact with silicon substrate, Al bottom electrode, and Al top electrode (ca. 10 nm thick): (a) electron density profile of the polymer film coated on a silicon (Si) substrate; (b) electron density profile of the polymer film coated on the Al bottom electrode supported with a Si substrate; (c) electron density profile of the polymer/Al structure; (d) electron density profile of the polymer/Al structure made by the combination of the electron density profiles in (b) and (c). All the results were obtained by analysis of the XR data in Fig. S3a.



Fig. S6. Synchrotron XR analysis of PVPK₂BEOXD₈ films (ca. 40 nm thick) in contact with silicon substrate, Al bottom electrode, and Al top electrode (ca. 10 nm thick): (a) electron density profile of the polymer film coated on a silicon (Si) substrate; (b) electron density profile of the polymer film coated on the Al bottom electrode supported with a Si substrate; (c) electron density profile of the polymer/Al structure; (d) electron density profile of the polymer/Al structure made by the combination of the electron density profiles in (b) and (c). All the results were obtained by analysis of the XR data in Fig. S3b.



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Fig. S7. Typical *I–V* curves of the Al/Polymer(40 nm thick)/Al devices, which were measured by negative voltage sweeps with a compliance current set of 0.01A: (a) $PVPK_8BEOXD_2$; (b) $PVPK_5BEOXD_5$; (c) $PVPK_2BEOXD_8$. The electrode contact area was $0.5 \times 0.5 \text{ mm}^2$. The switching-OFF run in (c) was carried out with a compliance current set of 0.1 A.



Fig. S8. Retention times of the ON- and OFF-states of the Al/Polymer(40 nm thick)/Al device: (a) PVPK₈BEOXD₂; (b) PVPK₅BEOXD₅; (d) PVPK₂BEOXD₈. All devices were probed with a reading voltage of +0.5 V, except for the ON-state of PVPK₂BEOXD₈ (+3.5 V).



Fig. S9. Voltage-current plots for the Al/Polymer(40 nm thick)/Al devices in the OFF-state: (a) PVPK₈BEOXD₂; (b) PVPK₅BEOXD₅; (c) PVPK₂BEOXD₈. The symbols are the measured data. In (a) PVPK₈BEOXD₂ and (c) PVPK₂BEOXD₈, the solid lines are the fit obtained with the ohmic current model for low-voltage regime and the trap-limited SCLC conduction model for high-voltage regime, respectively. In (b) PVPK₅BEOXD₅, the solid line is the fit obtained with the trap-limited SCLC conduction model.

