Electronic Supplementary Information (ESI) for

Reliable Organic Memristors for Neuromorphic Computing by Predefining a Localized Ion-Migration Path in Crosslinkable Polymer

Hea-Lim Park^a, Min-Hwi Kim^b, Min-Hoi Kim^{c*}, and Sin-Hyung Lee^{d*}

^aDepartment of Materials Science and Engineering, Gwanak-ku, Seoul National University,

Seoul 151-600, Republic of Korea

^bSchool of Electrical and Computer Engineering, Gwanak-ku, Seoul National University, Seoul

151-600, Republic of Korea

^cDepartment of Creative Convergence Engineering, Hanbat National University, Yuseong-ku,

Daejeon 305-719, Republic of Korea

^dSchool of Electronics Engineering, and School of Electronic and Electrical Engineering,

Kyungpook National University, Bukgu, Daegu 702-701, Republic of Korea

Corresponding Author

*E-mail: <u>mhkim8@hanbat.ac.kr</u> (M.-H, Kim), <u>sinhlee@knu.ac.kr</u> (S.-H. Lee)



Fig. S1 UV-vis absorption spectra of PVCi films according to the UV treatment time.



Fig. S2 Conductive filament formation in Device 1 with various compliance currents of the writing process. (a) Three types of electroforming processes with different compliance current values of 6×10^{-7} A (the left panel), 10^{-6} A (the middle panel), and 4×10^{-6} A (the right panel). (b) Field emission scanning electron microscopy images of the devices after the forming processes.



Fig. S3 Conceptual schematic diagram depicting the steps for defining the local ion-migration path (LIP) in the proposed organic memristor.



Fig. S4 Current-voltage curves of the vertical-type device consisting of the crosslinked polymer medium (PVCi).



Fig. S5 Electroforming processes for triggering (a) r-Device and (b) m-Device. The forming voltages of Device 1 and Device 2 are about 5.5 V and 5.0 V, respectively.



Fig. S6 Current variations in (a) r-Device and (b) m-Device at $V_{read} = 0.2$ V after the repeated 50 cycles of the writing and erasing processes.



Fig. S7 (a) A schematic of the lateral-type organic memristor. (b) Fabricating steps for the lateral-type organic memristors (r-Device and m-Device). In the case of m-Device, the photo-crosslinking process was performed after the electroforming process to define the LIP.



Fig. S8 Transient responses of m-Device to the switching voltage pulses. (a) Voltage pulse with the amplitude of 3.6 V was utilized for writing and (b) the pulse with the amplitude -2.0 V was utilized for erasing. The switching times for writing and erasing were about 300 µs and 300 µs, respectively.



Fig. S9 Short-term plasticity in m-Device. (a) Under the voltage pulse with the amplitude of 2.0 V, the device showed the volatile-memory characteristics. (b) The relaxation of the device in current level. The experimental data is fitted by an exponential decay function.



Fig. S10 Multi-level memory characteristics of m-Device. The voltage-pulse amplitudes for potentiation and depression were 4.8 V and -2.7 V, respectively. The time width of each pulse was 100 μs long.



Fig. S11 The nonlinear transmission characteristics of m-Device. The conductance of the device was effectively modulated by the dual-voltage sweeps.



Fig. S12 Energy consumption of m-Device in each pulse for the potentiation and depression processes illustrated in Fig. 6d.