

## Supporting Information

# A Nanofluidic Memristor Based on Ion Concentration Polarization

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## MATERIALS and METHODS

### Materials

Potassium chloride (KCl) was purchased from Thermo Fisher Scientific (Waltham, MA). Fluorescent polystyrene beads (green color; excitation: 488 nm) were purchased from Baseline Chromtech (Tianjin, China). Ultrapure water (18.2 M $\Omega$  cm) was obtained from a Millipore water purification system.

### Experiment

The device was bonded to a polydimethylsiloxane (PDMS) slab featuring inlet and outlet holes after an oxygen-plasma surface activation. KCl solution was injected shortly after the bonding to exploit the hydrophilic surface left by the plasma treatment, ensuring a bubble-free filling of the nanocapillaries. The measurements were performed with a source measure unit (SMU) Keithley 236 (Cleveland, OH) controlled using LabVIEW (National Instruments). Platinum electrodes (Leego Precision Alloy) were connected to the SMU and placed inside the reservoirs to apply the electric field across the nanochannels. The carboxyl-terminated 70-nm fluorescent polystyrene beads were diluted and dispersed in KCl solution to achieve a concentration of  $5.3 \times 10^9$  particles/ $\mu$ L (0.1% w/v) before being loaded into the device. Concurrently, the fluorescent time-series images were captured with an epifluorescence microscope (Eclipse, Nikon, Tokyo, Japan) equipped with an EMCCD camera (iXon3 897, Andor). All the images were processed using ImageJ (NIH, Bethesda, MD). The glass capillaries were characterized by using the scanning electron microscopy (SEM, JEOL JSM-6490, Japan).

### Microfabrication

A 100-mm silicon wafer was first thermally oxidized for 3  $\mu$ m thick oxide, and then transferred to a low-pressure chemical vapor deposition (CVD) furnace for another 3  $\mu$ m thick low temperature oxide (LTO). Trenches were structured 2  $\mu$ m wide and deep in LTO layer by UV lithography and advanced oxide etch (AOE). A 100 nm thick silicon nitride film was deposited as a diffusion-stop layer. A 6.5  $\mu$ m thick layer of phosphosilicate glass (PSG) was deposited through CVD and the resultant non-conformal step coverage profile transformed the trenches into self-enclosed triangular conduits. The phosphorus content of PSG was set at a target concentration of 8 mol % to facilitate the glass thermal reflow.<sup>1</sup> The subsequent thermal annealing at 1000 °C for 60 min was sufficient to turn the triangular conduits into cylindrical capillaries with a nominal diameter of 200 nm. To further shrink the capillary diameter below 100 nm, rapid thermal annealing (RTA) was performed at 1000 °C for several cycles with a duration 2 min/cycle. The capillary diameter was confirmed through SEM. Nanocapillary formation process is schematically described in Fig. S1.<sup>2</sup>

## FIGURES

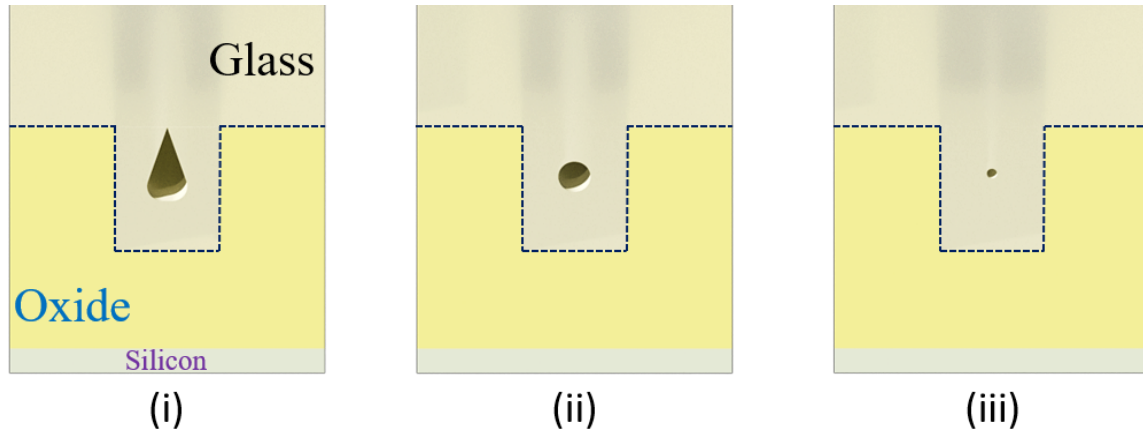


Figure S1. Nanocapillary formation process illustrated by cross-sectional diagrams. (i) Glass deposition and the formation of a self-enclosed triangular void inside a trench. (ii,iii) Glass reflow through rapid thermal annealing: (ii) void shape transformation and (iii) void diameter shrinking.

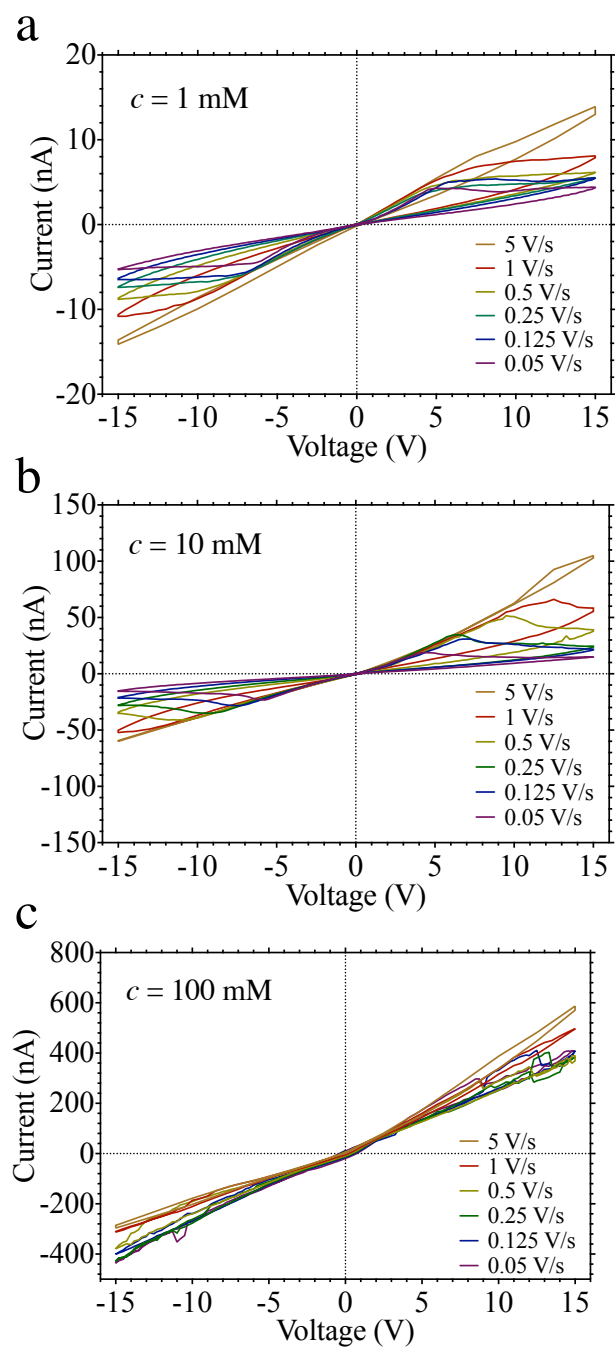


Figure S2. Hysteric  $I - V$  curves measured at various sweep rates from 70-nm nanocapillary device using (a) 1 mM, (b) 10 mM, and (c) 100 mM KCl as the background electrolyte.

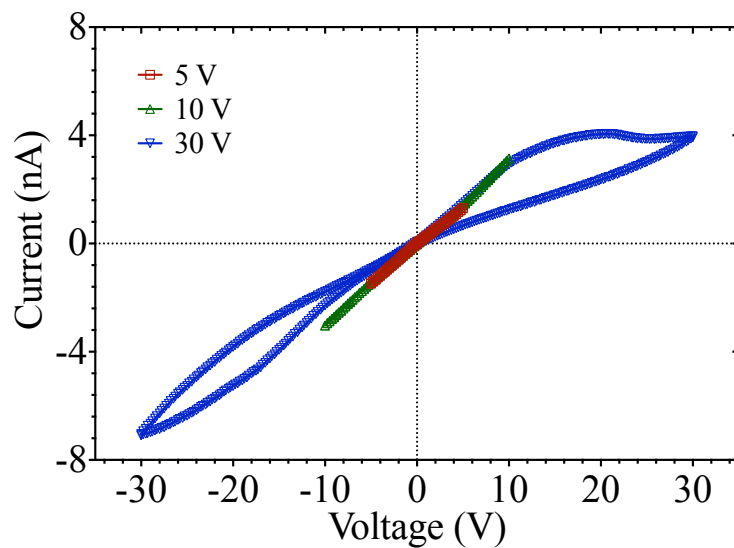


Figure S3. The sweep range dependence of  $I - V$  characteristics from 70-nm nanocapillary device (1 mM KCl).

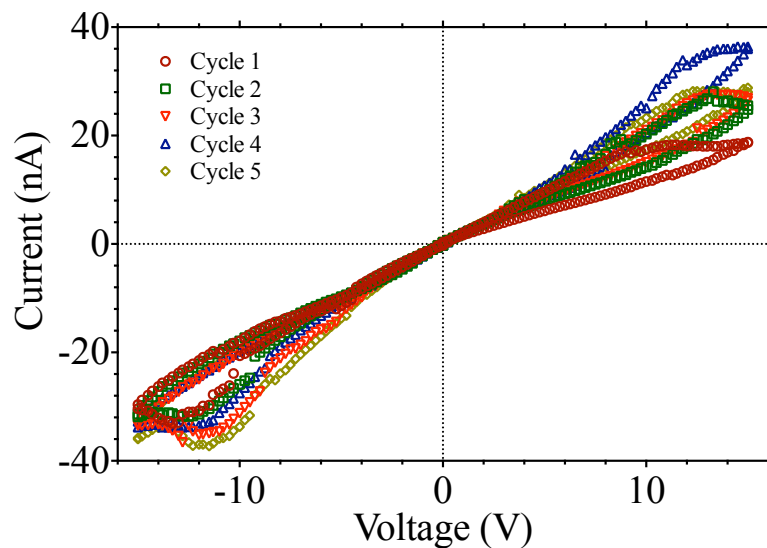


Figure S4. The  $I - V$  characteristics obtained in five consecutive cycles from 70-nm nanocapillary device using 10 mM KCl as the background electrolyte. The sweep rate: 0.5 V/s.

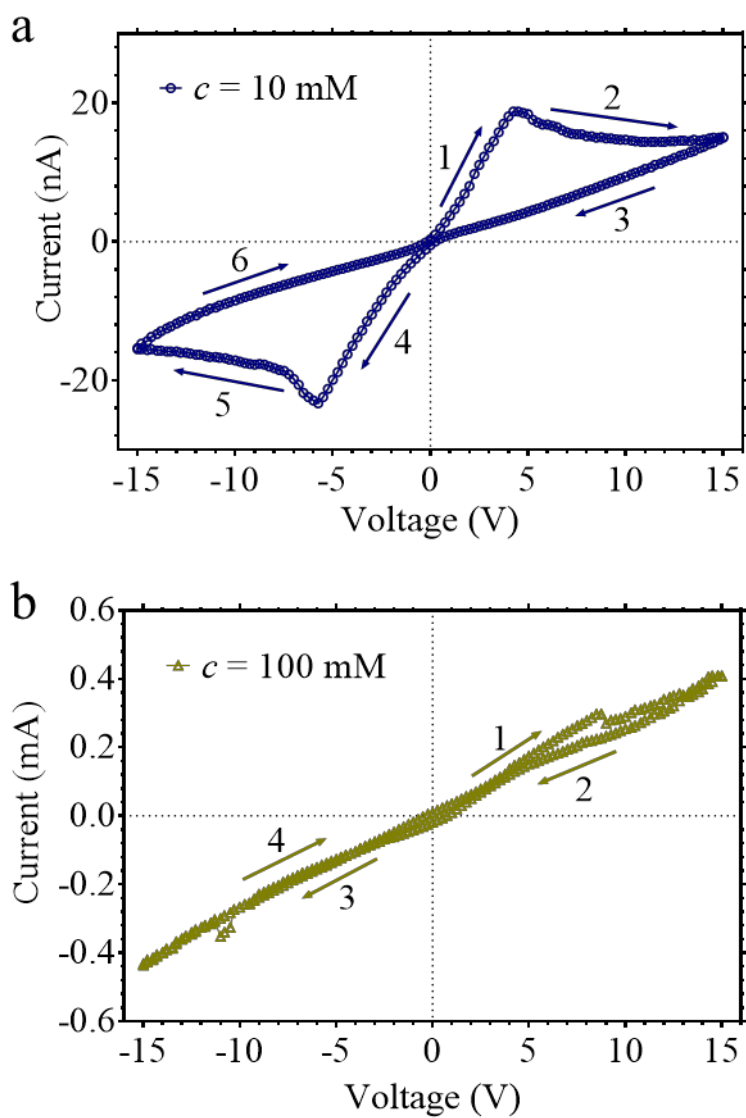


Figure S5. The hysteretic  $I - V$  curves obtained from 70-nm nanocapillary device using (a) 10 mM and (b) 100 mM KCl as the background electrolyte. The arrows and the numerals indicate the bias sweep direction and order. The sweep rate: 0.05 V/s.

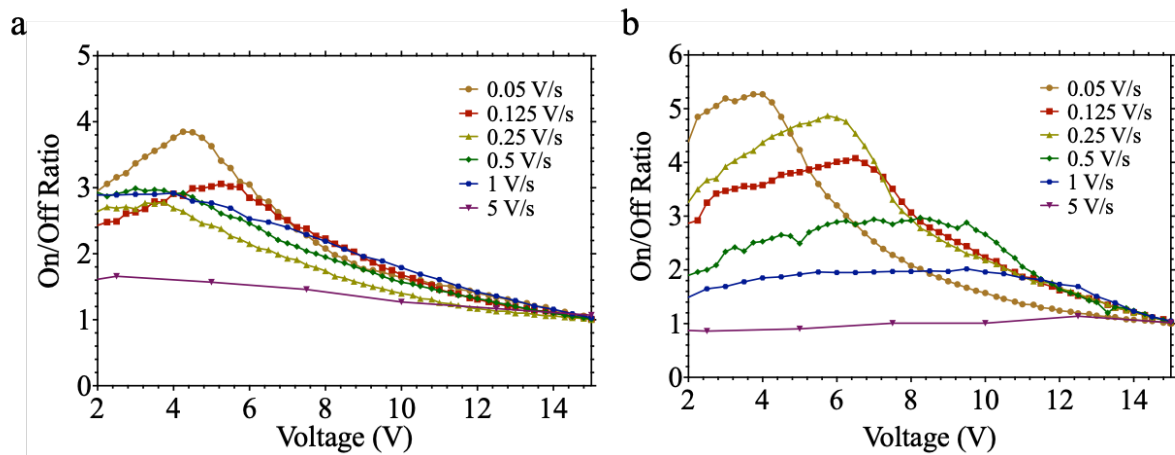


Figure S6. The ON/OFF ratio plots against the positive bias sweep for various sweep rates (legends) based on  $I - V$  curves obtained from 70-nm nanocapillary device using (a) 1 mM and (b) 10 mM KCl.



## REFERENCES

- (1) Y. Liu and L. Yobas, *Biomicrofluidics*, 2012, **6**, 046502.
- (2) N. S. Alvi and D. L. Kwong, *J. Electrochem. Soc.*, 1986, **133**, 2626–2631.