Electronic Supplementary Information (ESI)

Electrically Switched Asymmetric Interface for Liquid Manipulation

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Fig. S1 (a) Superhydrophobic substrate composed of PTFE on glass surface. (b) The contact angle of liquid droplet on the substrate surface is ~156° in oil, indicating the prepared PTFE/glass substrate is superhydrophobic.



Fig. S2 The contact angle of liquid droplet (2 μ L) on flat electrode surface in oil (a) is ~140° and in air (b) is ~74°.



Fig. S3 Electric field induced unidirectional liquid droplet motion on the wire electrode surface. Scale bars are 1 mm.



Fig. S4 Liquid droplet wetting and dewetting behavior on electrode surface by changing the electrode shape and direction. Scale bars are 3 mm.



Fig. S5 Schematic preparation of groove-microstructured Pt electrode by laser etching.



Fig. S6 Microscopic surface morphology of microstructured electrode surface with different groove width. (a) w=0 μ m, (b) w=50 μ m, (c) w=100 μ m, (d) w=200 μ m, (e) w=300 μ m, (f) w=400 μ m, (g) w=500 μ m and (h) w=600 μ m. Scale bars are 1 mm.



Fig. S7 Contact angle of groove-microstructured electrode surface in air along X and Y direction changes with different groove width.



Figure S8. Contact angle of liquid droplet on flat electrode surface changes with the applied voltage of -4~4 V.



Fig. S9 Comparison of cathode and anode gas generation rate on flat electrode surface using the drainage gas collection method.



Fig. S10 Electric field induced directional dichloromethane transport on electrode surface in water. Scale bars are 1 mm.



Fig. S11 Schematic liquid droplet wetting on the groove-microstructured electrode surface. For the groove-microstructured electrode surface with groove microstructure depth (*h*), intrinsic contact angle (ϑ_0), radius of liquid droplet (*R*) and the groove width (*w*), based on the relationship:

$$R = \frac{h}{1 - \sin(\pi - \theta 0)} = \frac{h}{1 - \sin\theta 0} \# (1)$$
$$\frac{w}{\frac{2}{R}} = \cos(\pi - \theta 0) = -\cos\theta_0 \# (2)$$

When the liquid droplet just contacts with bottom of the groove-microstructured electrode, the critical width of groove (w_c) can be calculated as:

$$w_c = \frac{2h\cos\theta0}{\sin\theta0 - 1} \#(3)$$

For the groove-microstructured electrode with intrinsic CA of ~140°, and h~80 µm, the critical width of groove (w_c) by calculating is 345 µm, *i.e.*, when w_c =345 µm, liquid droplet can contact with and just wet the bottom of groove-microstructured electrode.



Fig. S12 Liquid droplet wetting states on electrode surface with different groove width. (a) Liquid droplet wetting in the initial state on flat electrode surface. (b) Liquid droplet wetting in the Cassie state on groove-microstructured electrode surface with $w=0^{\sim}300 \ \mu\text{m}$. (c) Liquid droplet wetting in the Wenzel state on groove-microstructured electrode surface surface with $w>300 \ \mu\text{m}$. The contact area between liquid droplet and electrode surface can be described as follows: S_{III}>S_I>S_{II}.



Fig. S13 Cross-sectional SEM images of groove-microstructured electrode (w=400 $\mu\text{m}).$



Fig. S14 Liquid droplet contact angle on (a) flat and (b) groove-microstructured electrode surface during directional liquid droplet (10 μ L) motion at critical voltage (4 V) under the unbalanced force. $F_{E,c}$ activated on both ends of the liquid droplet, as calculated by equation (4) :

$$F_{E,c} = \int_{s}^{L} \gamma ow(\cos\theta + -\cos\theta -)dl\#(4)\#$$

where θ_{+} and θ_{-} are the CAs of liquid droplet under electric field on the anode and cathode surfaces, respectively. γ_{ow} is the interfacial tension between the oil and the water, and *dl* is the corresponding length of the liquid droplet on electrode surface. The calculated results indicate that groove microstructure facilitates liquid droplet motion.



Fig. S15 Adhesion force-distance charts of liquid droplet (2 μ L) on flat and groovemicrostructured electrode surface in oil, showing that groove-microstructure reduces the surface adhesion force.



Fig. S16 Liquid droplet wetting distance on combined electrode surface varies with groove width and volume in air. (a) w=0~600 μ m, (b) w=50~600 μ m, (c) w=100~600 μ m, (d) w=200~600 μ m, (e) w=300~600 μ m, (f) w=400~600 μ m, (g) w=500~600 μ m and (h) w=600 μ m.

Table S1 The effect of surface tension for liquid droplet with different compositions on themotion behavior.

Composition	Directional	Droplet splitting	Surface tension
	transport		(mN/m)
Deionized water	×	×	72.56
0.1 M PBS	V	×	68.21
0.1 M NaCl	V	×	67.95
0.1 M PBS+0.001 M DBAE	V	×	58.87
0.1 M NaCl+0.001 M DBAE	V	×	58.30
0.1 M PBS+0.005 M DBAE	V	×	54.44
0.1 M NaCl+0.005 M DBAE	V	×	54.28
0.1 M PBS+0.01 M DBAE	V	×	50.39
0.1 M NaCl+0.01 M DBAE	V	×	49.64
0.1 M PBS+0.05 M DBAE	V	V	42.44
0.1 M NaCl+0.05 M DBAE	V	V	42.20
0.1 M PBS+0.1 M DBAE	V	V	38.67
0.1 M NaCl+0.1 M DBAE	V	V	38.42
0.1 M PBS+0.2 M DBAE	V	V	34.77
0.1 M NaCl+0.2 M DBAE	V	V	34.12

Supplemental Movies

Movie S1 Droplet motion comparison on electrode surface at the voltage of 4 V and 10 V.

Movie S2 Droplet splitting comparison on flat and groove-microstructured electrode surface

at the voltage of 30 V and volume of 40 $\mu\text{L}.$