Supplemental Information

Self-Flooding Behaviors on Fuel Cell Catalyst Surface: An In-Situ Mechanism Investigation

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|	Pt	Pt*	O_{OH^-}	H_{OH^-}	O_{H3O^+}	H_{H3O^+}	O_{H2O}	O_{H2O}
ε(kcal/mol)	15.718	4.000	0.158	0	0.156	0	0.155	0
σ(Å)	2.542	2.542	3.214	0	3.214	0	3.166	0
charge	0	0	-1.35	0.35	-0.4166	0.4722	-0.8476	0.4238

**Table. S1** Interaction parameters between atoms and the charge distribution.
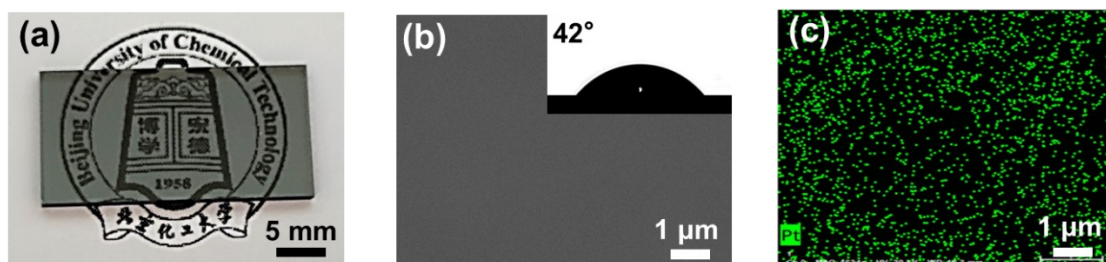
Fig. S1 (a) Digital image and (b) SEM image of the Pt-coated ITO electrode, inset: the water droplet contact angle. (c) Elementary mapping of Pt.
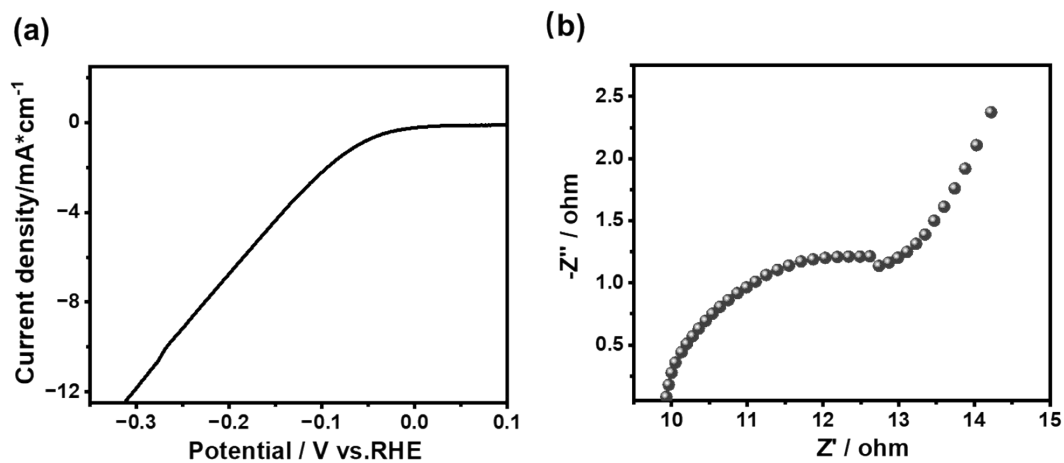

Fig. S2 (a) Polarization curve and (b) AC impedance curve of the Pt-coated ITO electrode.
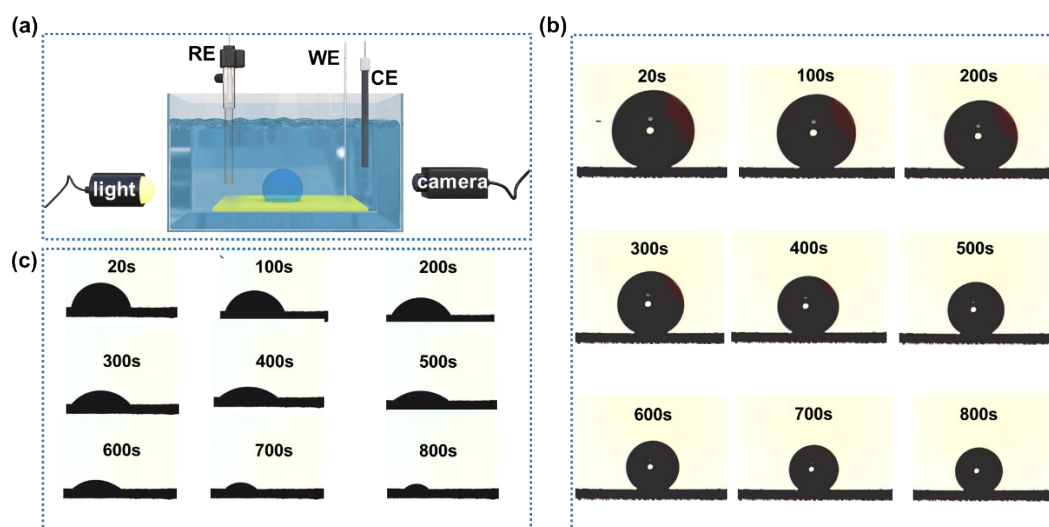
Fig. S3 (a) The configuration of in situ monitoring of bubble consumption process based on microscopy with side-view perspective. Snapshots of the processes bubble consumption on (b) aerophobic and (c) areophilic electrode.
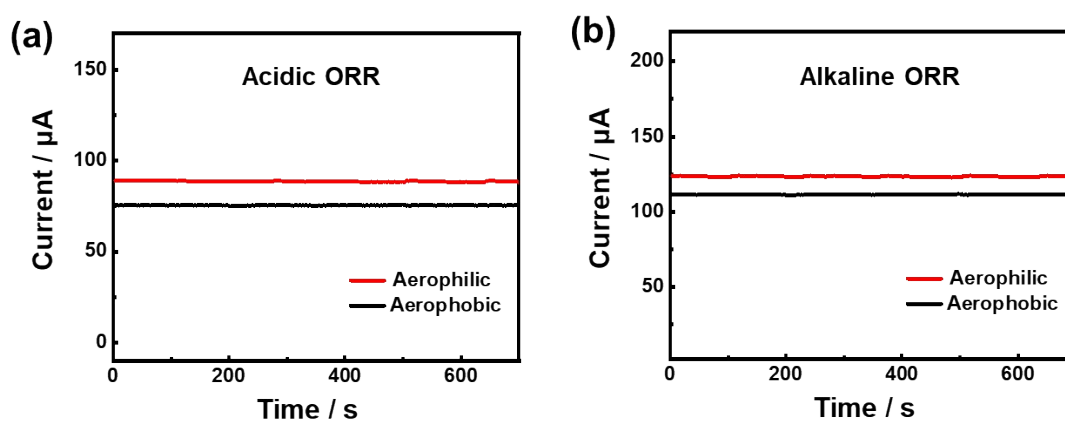

Fig. S4 Comparison of background currents of ORR on aerophobic and aerophilic electrode surfaces in (a) acidic and (b) alkaline medium.
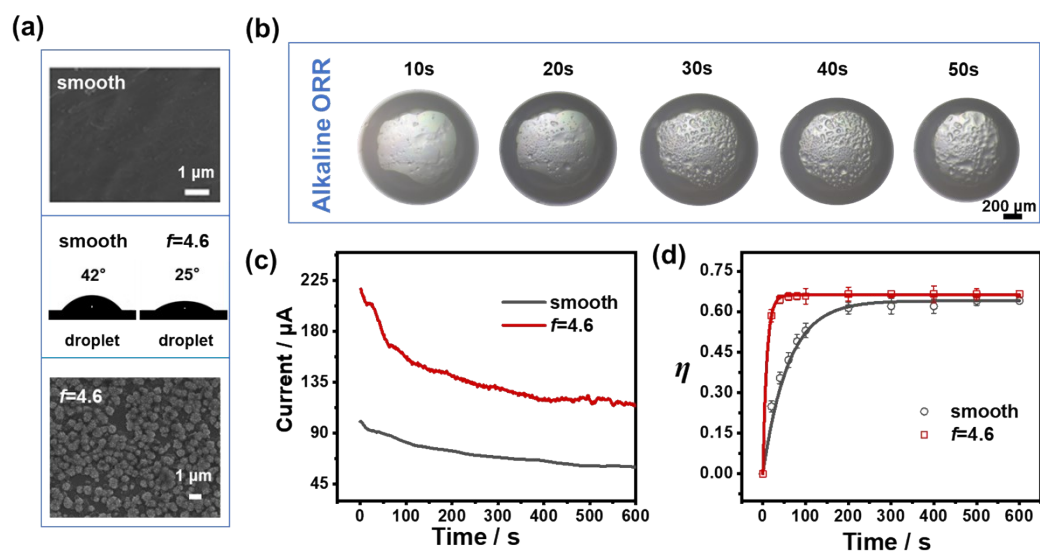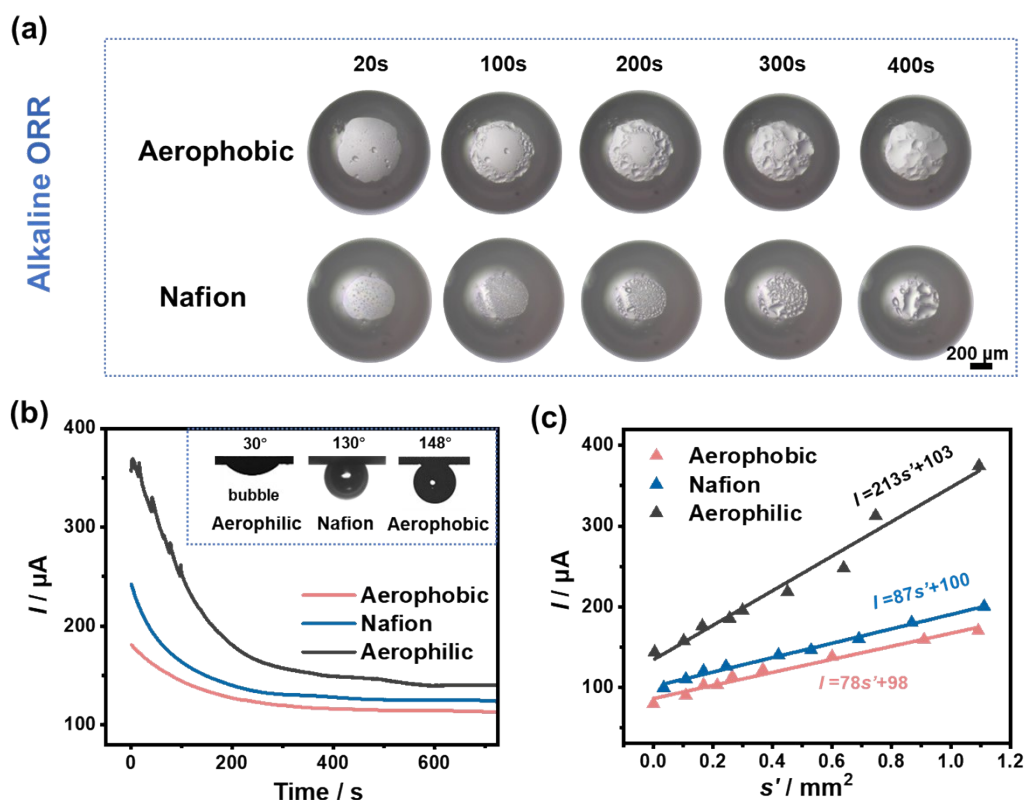
Fig. S5 (a) SEM images and water contact angles of smooth (up) and rough (bottom) electrodes. (b) Typical snapshots of bubble/electrode interface during ORR in alkaline electrolytes at different time points with the roughness factor of 4.6. Time-dependent variation of (c) current and (d) droplet coverage η ($\eta=(s-s')/s$) with smooth and rough electrodes.
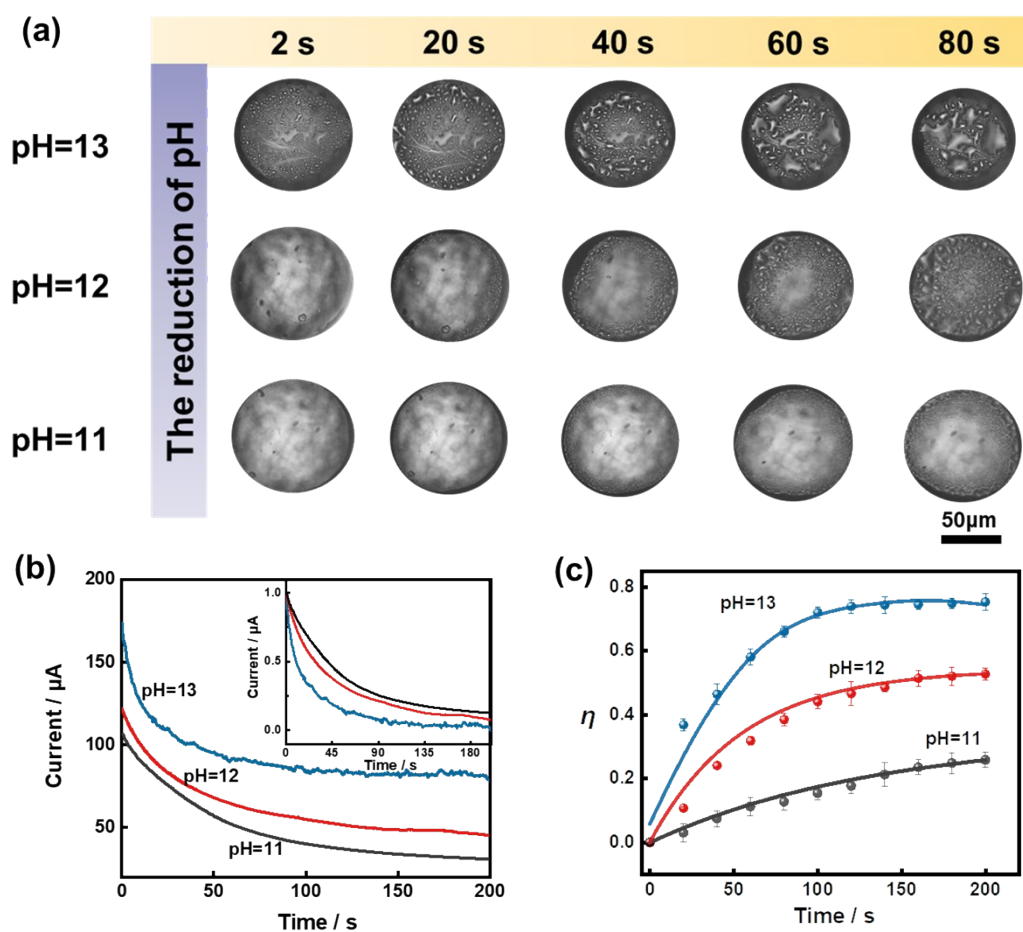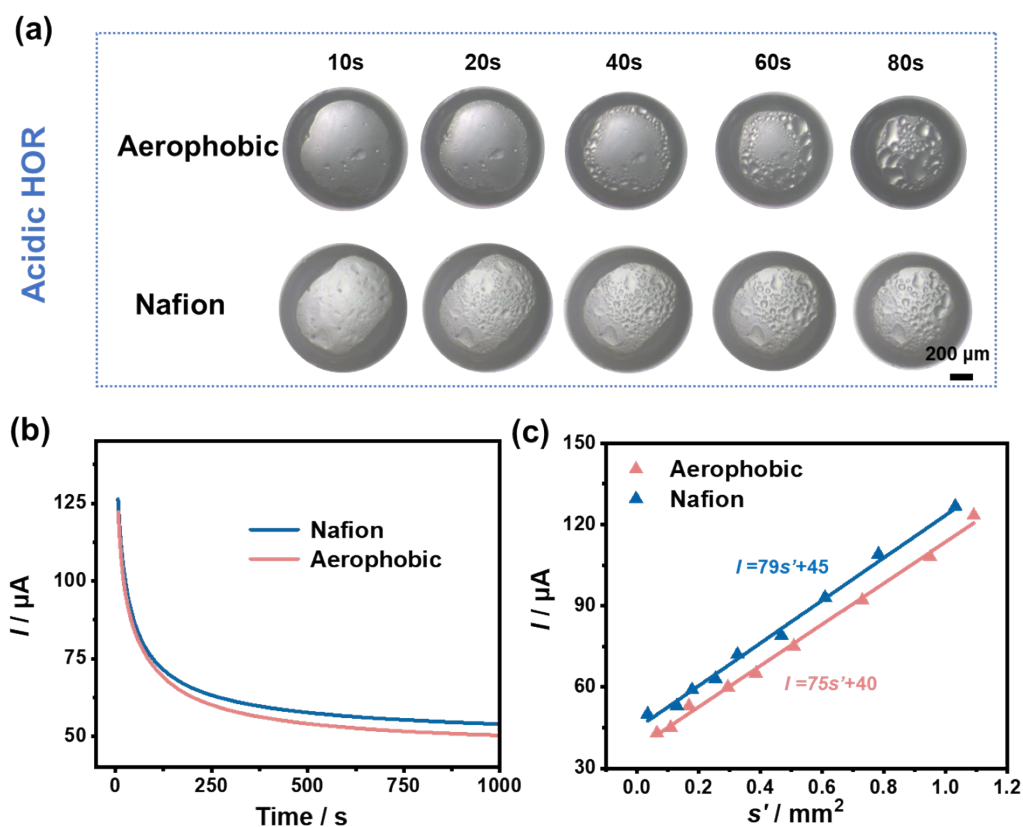
Fig. S6 (a) Variations of bubble/electrode interfaces at different pH value from 13 to 11. Time-dependent variations of (b) current and (c) droplet coverage η.
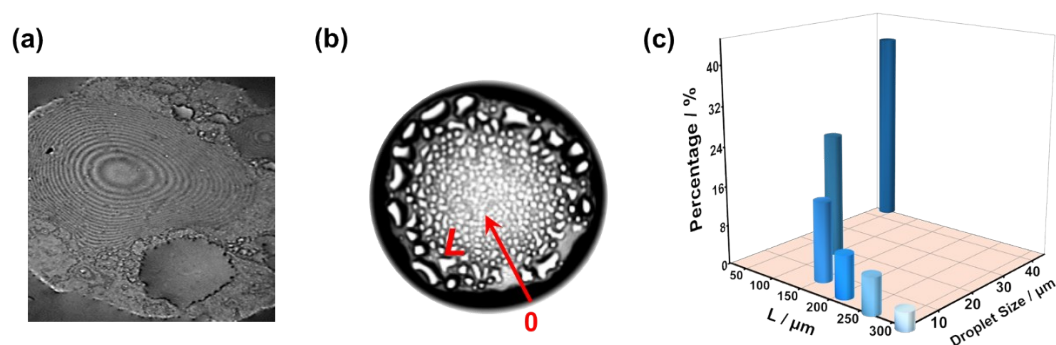

Figure S7. (a) Magnified digital image of the as-formed droplets at the bubble/electrode interface. (b) A typical snapshot of bubble/electrode interface during the self-flooding process. (c) Statistical graph of the size and proportion of the droplets generated along with the distance from the edge of the bubble to the droplets.

Fig. S8 The flooding phenomena and influence on current in alkaline ORR with Nafion. (a) Typical snapshots of bubble/electrode interface during ORR in alkaline electrolytes at different time points with the Nafion and aerophobic surfaces. (b) Time-dependent current variations of the aerophobic, Nafion-modified and aerophilic surfaces. Inset, bubble contact angles of the three electrodes. (c) Fitted lines of net area (s') with current on the three electrodes.

Fig. S9 The flooding phenomena and influence on current in acidic HOR with Nafion. (a) Typical snapshots of bubble/electrode interface during HOR in acidic electrolytes at different time points with the Nafion and aerophobic surfaces. (b) Time-dependent current variations of the aerophobic and Nafion-modified surfaces. (c) Fitted lines of net area (s') with current on the two electrodes.
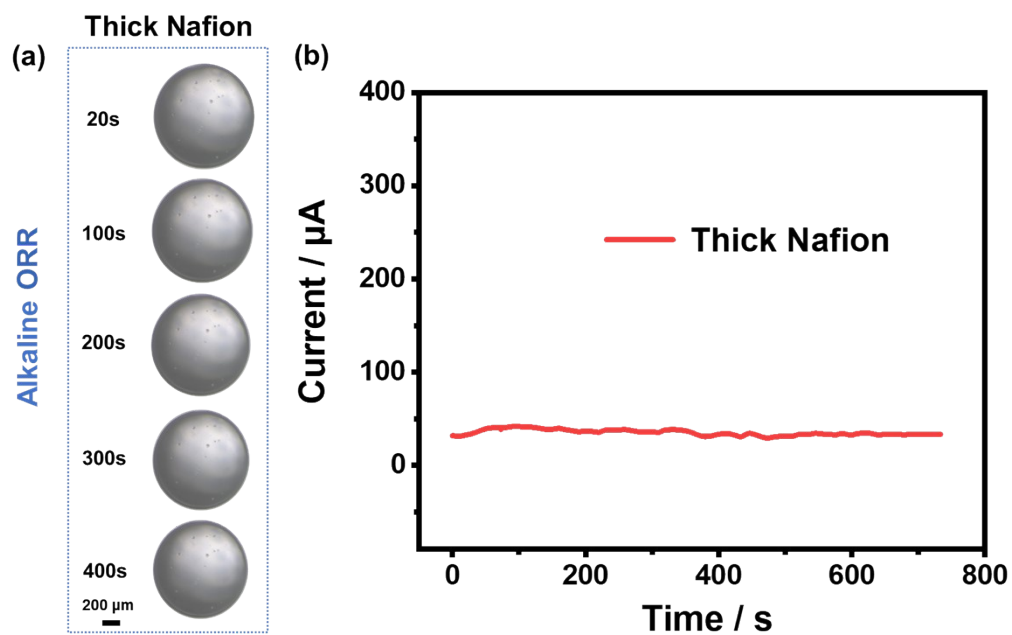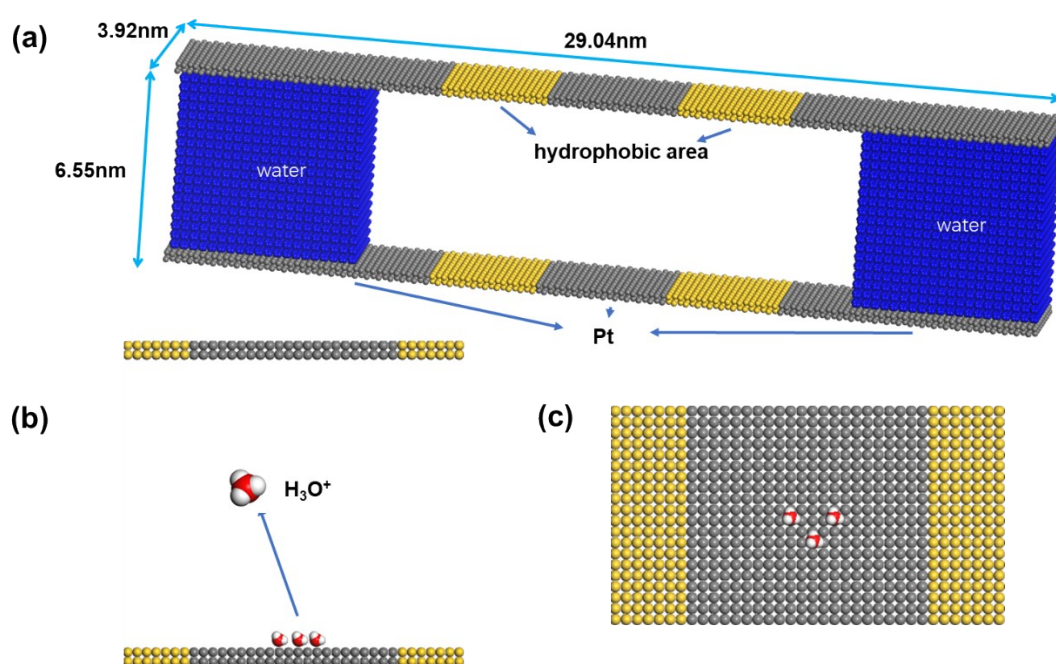

Fig. S10 (a) Typical snapshots of bubble/electrode interface during ORR in alkaline electrolyte at

different time with thick-layer ionomer binding. (b) Plot of current (I) with time on the thick-layer ionomer binding surface.
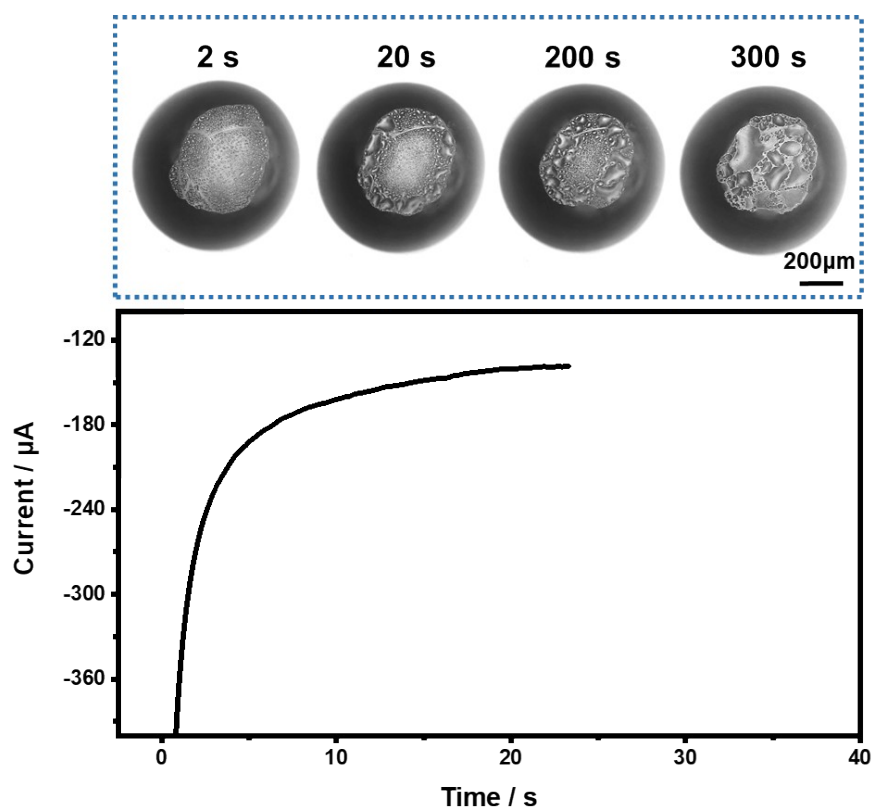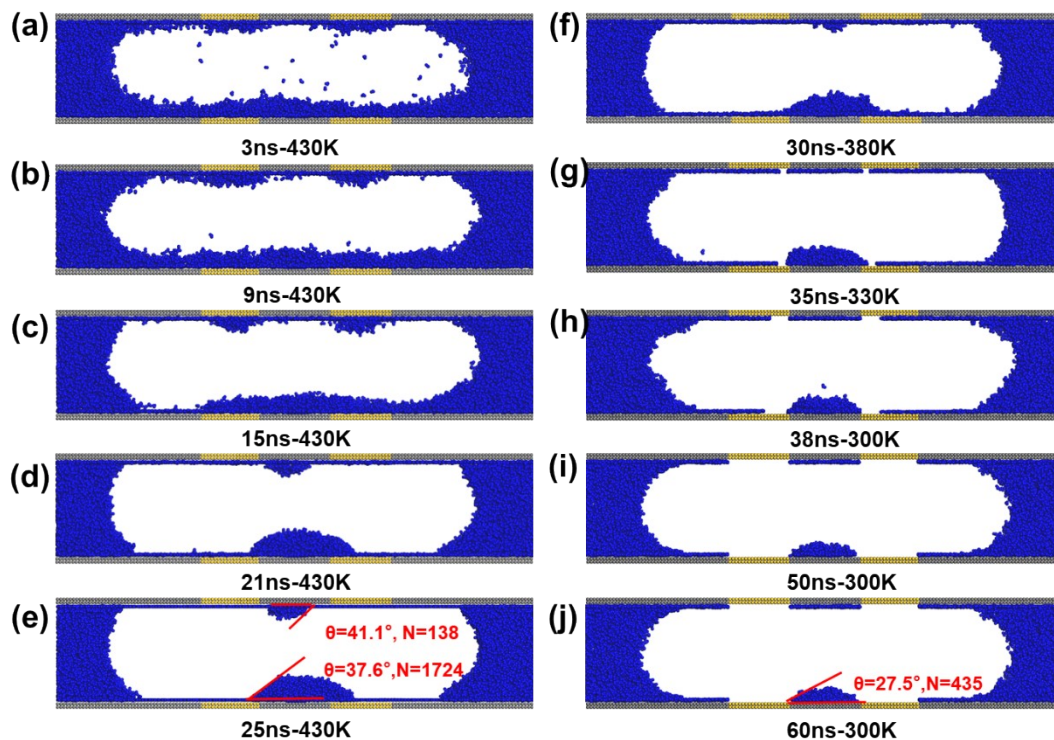

Fig. S11 The variations of the bubble/electrode interface after stopping the reaction at 20 s, upper panel: typical snapshots.
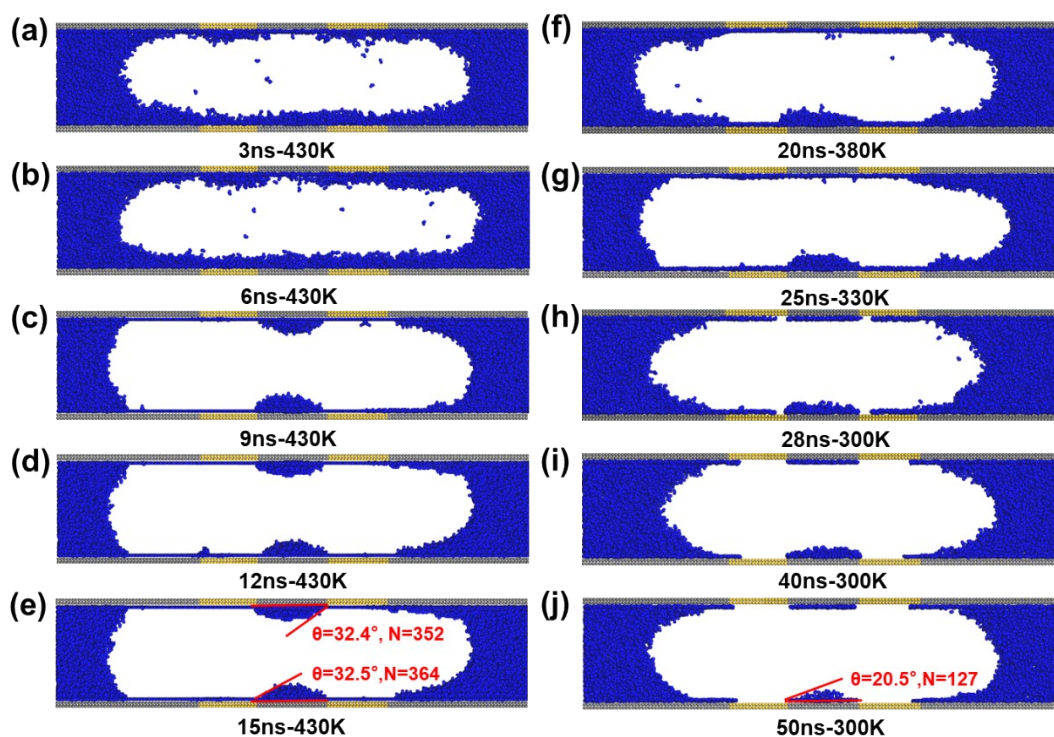
Fig. S12 (a) Initial structure of the simulation system. The size of the whole box is 29.04nm×3.92nm×10nm. The gray and yellow particle are all Pt atoms with different interaction parameters. There are 9120 H_2O molecules in the box. (b) and (c) The position of H_3O^+ be placed.

(a) 3ns-430K
(b) 9ns-430K
(c) 15ns-430K
(d) 21ns-430K
(e) 25ns-430K — θ=41.1°, N=138; θ=37.6°,N=1724
(f) 30ns-380K
(g) 35ns-330K
(h) 38ns-300K
(i) 50ns-300K
(j) 60ns-300K — θ=27.5°,N=435

Fig. S13 Snapshots of droplet nucleation on the electrode surface for alkaline ORR. The simulation temperature is set at 430K for first 25ns, and then adjusted to 300K at a 10K/ns speed.

(a) 3ns-430K
(b) 6ns-430K
(c) 9ns-430K
(d) 12ns-430K
(e) 15ns-430K — θ=32.4°, N=352; θ=32.5°,N=364
(f) 20ns-380K
(g) 25ns-330K
(h) 28ns-300K
(i) 40ns-300K
(j) 50ns-300K — θ=20.5°,N=127

Fig. S14 Snapshots of droplet nucleation on the electrode surface for acidic HOR. The simulation temperature is set at 430K for first 15ns, and then adjusted to 300K at a 10K/ns speed.