Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supporting Information

Calculation of Reynolds numbers:

$Re = \rho U D_h / \eta$

- Density (ρ) and viscosity (η) are obtained from the references [1-3].
- Average velocity (U) is calculated based on the volumetric flow rate (Q) and the channel dimensions (L and H).
- Hydraulic diameter (D_h) is calculated from channel dimensions.

Table S1. Re values for 1M sulfuric acid based on the density, viscosity, and average velocities.

L [mm]	H [mm]	D_h [m]	Q [ml/h]	ρ [kg/m^3]	η [Pa.s]	U [m/s]	Re
1	1	0.001	300	1060	0.001208	0.08	73
1	1	0.001	600	1060	0.001208	0.17	146
1	1	0.001	900	1060	0.001208	0.25	219
1	1	0.001	1200	1060	0.001208	0.33	292

Table S2. Re values for 0.5M sulfuric acid based on the density, viscosity, and average velocities.

L [mm]	H [mm]	D_h [m]	Q [ml/h]	ρ [kg/m^3]	η [Pa.s]	U [m/s]	Re
1	1	0.001	300	1030	0.001102	0.08	78
1	1	0.001	600	1030	0.001102	0.17	156
1	1	0.001	900	1030	0.001102	0.25	234
1	1	0.001	1200	1030	0.001102	0.33	312

Table S3. Re values for 20% NaCl solution based on the density, viscosity, and average velocities.

L [mm]	H [mm]	D_h [m]	Q [ml/h]	ρ [kg/m^3]	η [Pa.s]	U [m/s]	Re
1	1	0.001	300	1147.4	0.00142	0.08	67
1	1	0.001	600	1147.4	0.00142	0.17	135
1	1	0.001	900	1147.4	0.00142	0.25	202
1	1	0.001	1200	1147.4	0.00142	0.33	269

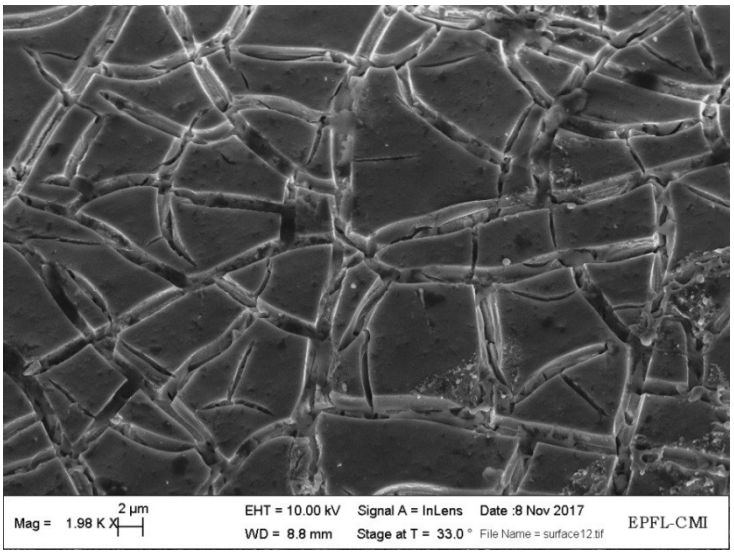

Figure S1. The SEM image of the electrodes surface: the cracks show the effect of annealing steps. Although these cracks increase the active area to some extent, this electrode is relatively flat as an electrocatalyst.

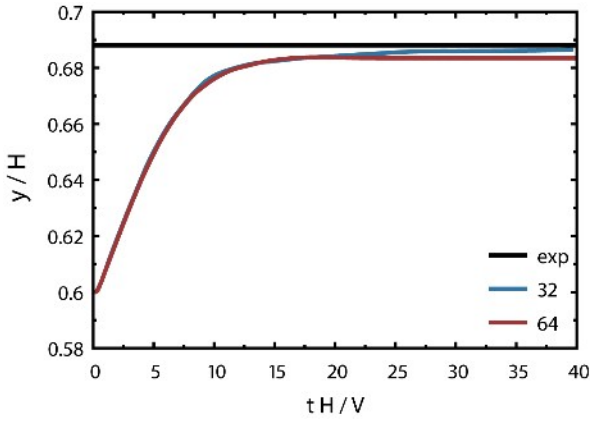

Figure S2. Lateral position of the bubble center: simulations at resolution of 32 and 64 cells per channel height in comparison to the experimental data [4] for the equilibrium position. Parameters of the flow are $Re = 7$, $Ca = 0.16$ and bubble radius $R/H = 0.24$ with channel height H and centerline velocity V.

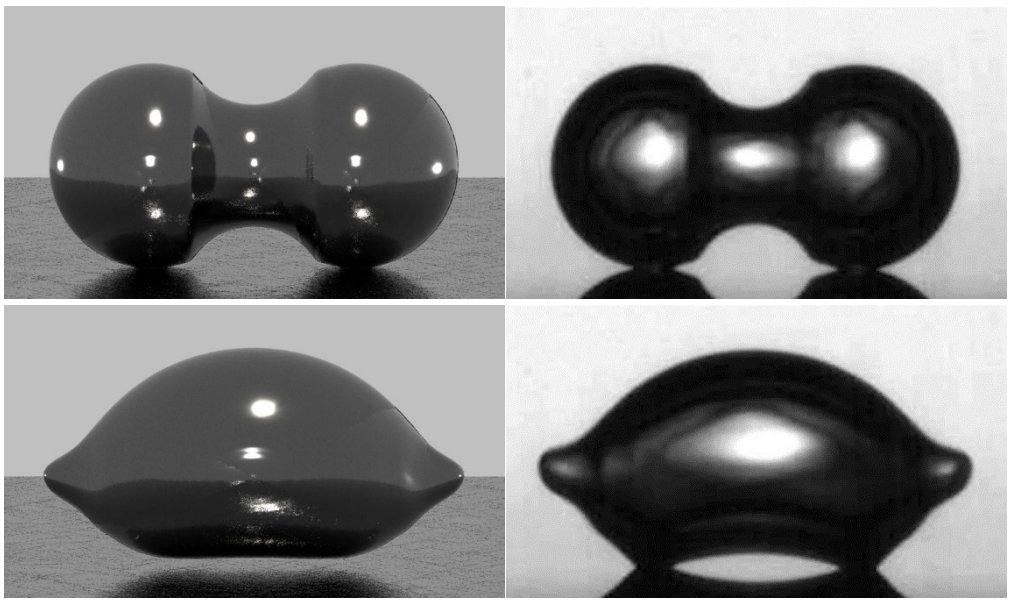

Figure S3. Snapshots of bubbles after coalescence from the simulation (left) in comparison to the experiment (right) [5].

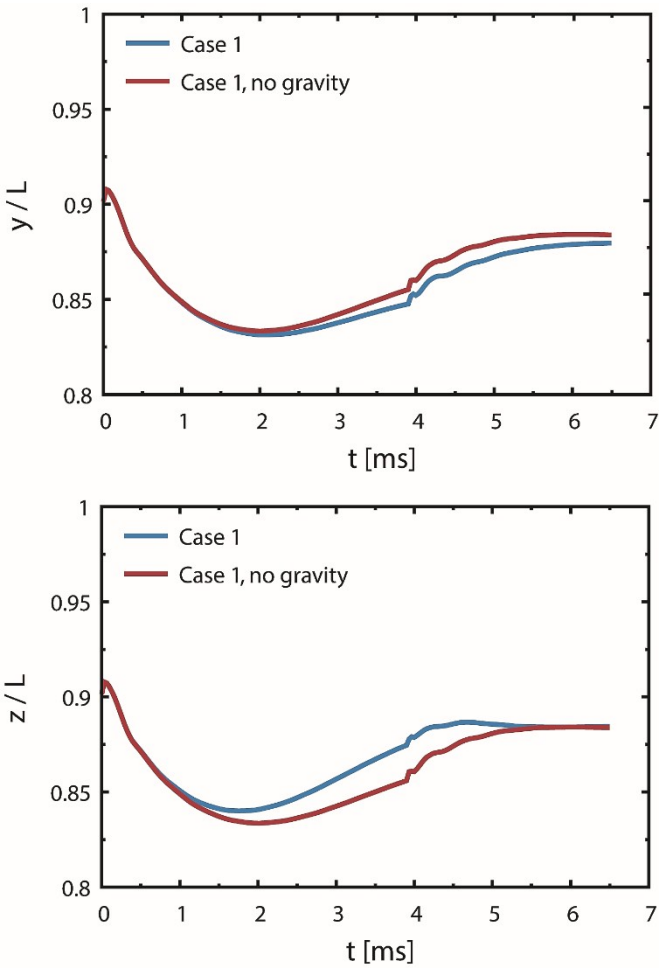

Figure S4. Comparison of the results for Case 1 in the main text with and without gravity for bubbles of 0.1 mm in radius. The gravity affects minimally the distance from the electrode (top) and from the lower wall (bottom).

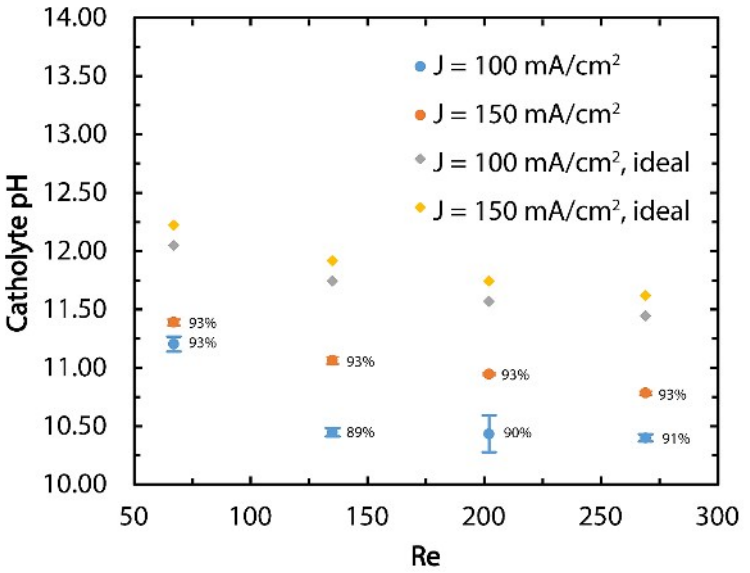

Figure S5. The measured and ideal catholyte pH values for a set of current densities and flow rates. The numbers next to each experimental data point shows the Faradaic efficiency. These values are in contrast to the Faradaic efficiencies from calorimetric measurements (Figure9) that increase with flow rate. This is likely due to the fact that in calorimetry measurements, some Cl_2 is lost in gaseous form, especially at low flow rates.

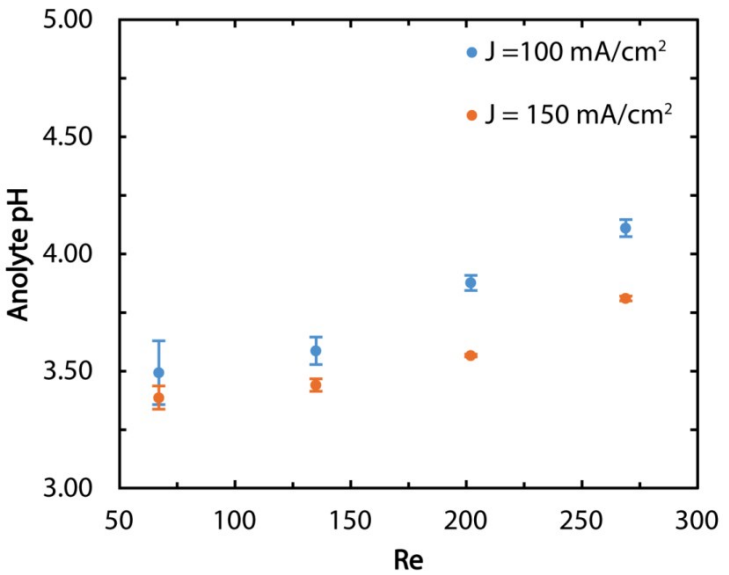

Figure S6. The measured anolyte pH values for a set of current densities and flow rates.

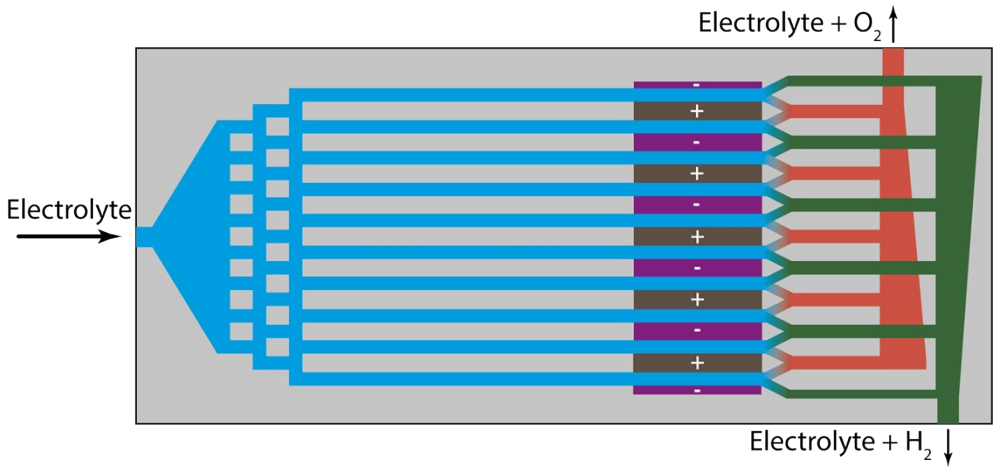

Figure S7. The schematic illustration of multiple cells in one row of a stack (top view): several of these rows will be stacked on top of each other in the final device and the electrodes will be inserted in form of metallic bars from the top layer all the way down towards the bottom layer. The electrolyte will be distributed between the cells through a network of pathways with equal hydraulic resistances. At the outlet, the mixture of electrolyte and H_2 is collected through dedicated interconnected channels (in green) and the same happens for the mixture of O_2 and electrolyte (in orange). Anodes are shown in brown color and plus signs and cathodes in violet and minus signs. For higher production rates, several of these stacks can be integrated into the electrolysis device.

Movies:

These videos are recorded at 4000 fps and play at 30 fps.

Movie S1: the upstream region of the flow based electrolysis cell operating at Re = 312 (1200 ml/h of 0.5 M H_2SO_4) and current density of 450 mA/cm^2.

Movies S2: the midstream region of the flow based electrolysis cell operating at Re = 312 (1200 ml/h of 0.5 M H_2SO_4) and current density of 450 mA/cm^2.

Movies S3: the downstream region of the flow based electrolysis cell operating at Re = 312 (1200 ml/h of 0.5 M H_2SO_4) and current density of 450 mA/cm^2.

Movie S4: the midstream region of the flow based electrolysis cell operating at Re = 234 (900 ml/h of 0.5 M H_2SO_4) and current density of 450 mA/cm^2, overlapped with results of simulation.

References:

1. http://www.labchem.com/tools/msds/msds/LC25770.pdf
2. http://www.labchem.com/tools/msds/msds/LC25790.pdf
3. Carvalho, G. R., et al. "Physicothermal properties of aqueous sodium chloride solutions." *Journal of Food Process Engineering* 38.3 (2015): 234-242.
4. P. Hadikhani, S. M. H. Hashemi, G. Balestra, L. L. Zhu, M. A. Modestino, F. Gallaire and D. Psaltis, *Lab Chip*, 2018, **18**, 1035-1046.
5. A. M. Soto, T. Maddalena, A. Raters, D. van der Meer and D. Lohse, J Fluid Mech, 2018, 846, 143-165.