Electronic Supplementary Material (ESI) for Reaction Chemistry & Engineering.

Bayesian Optimization of Electrochemical Devices for Electrons-to-Molecules Conversions: The Case of Pulsed CO_2 Electroreduction

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Additional Figures

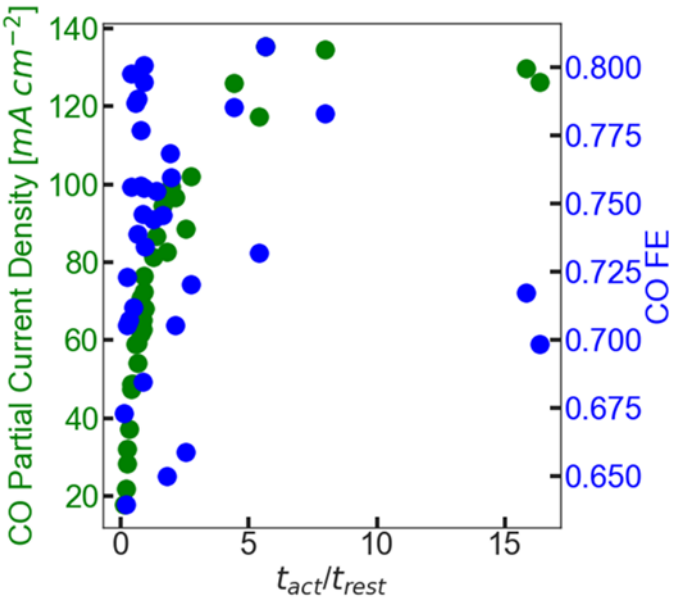

Figure S1. CO partial current density and CO FE as a function of the ratio of t_{act} to t_{rest}. Each experimental point represents one of the points in the 2D map experimentation campaign. The CO partial current density increases with larger ratios, until an upper limit is reached. The CO FE doesn't appear to be as dependent on the ratio.

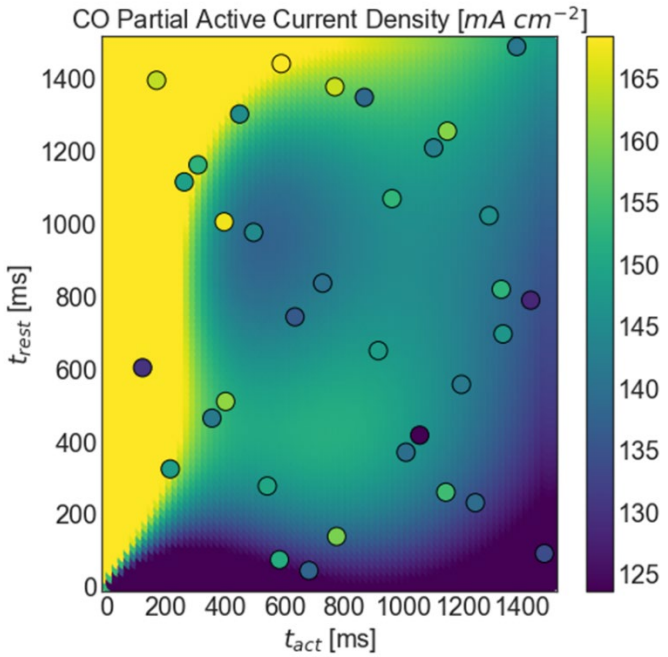

Figure S2. 2D map of CO partial current density during active time while varying t_{act} and t_{rest}. Experimental conditions are shown with a black outline. The background displays the GPR prediction based on the observed experimental values. Active current density was set to 200 mA cm^{-2} and resting current density was set to 0 mA cm^{-2}.

Accounting for device degradation

To account for the device performance degradation over the life of the membrane electrode assembly (MEA), a systematic process of baseline performance testing and normalization was performed.

1. Assemble MEA into cell.
2. Run and analyze an experiment using a constant current density of 200 mA/cm^2.
    a. Use the CO mol % value obtained from the microGC to normalize the subsequent pulse experiments in step 3
3. Following the constant current density experiment, perform 10 pulse experiments.
    a. Two of the 10 pulse experiments are repeated for each set of 10 experiments for the duration of the optimization campaign.
    b. The remaining 8 pulse experiments were chosen by the optimization campaign. **The normalized CO mol % values were used as the objective function values in the Bayesian optimization campaign.**
4. Once 10 pulse experiments were run, steps 2 and 3 were repeated.
5. After 2 trials of each pulse experiment, the MEA was changed for the next experiments in the BO campaign.

Table S1. Summary of the data for the two repeated pulse experiments (Step 3a) throughout the 3D optimization campaign.

Active pulse time [ms]	**Resting pulse time [ms]**	**Active current density [mA/cm^2]**	**Average normalized CO production (n=12)**	**Standard deviation of normalized CO production (n=12)**
982	965	240	0.61	0.09
756	159	286	1.08	0.33

Table S2. Summary of the data for the two repeated pulse experiments (Step 3a) throughout the 4D optimization campaign.

Active pulse time [ms]	**Resting pulse time [ms]**	**Active current density [mA/cm^2]**	**Resting current density [mA/cm^2]**	**Average normalized CO production (n=12)**	**Standard deviation of normalized CO production (n=12)**
182	587	163	57	0.47	0.08
1020	625	167	12	0.48	0.14