Galvanic Corrosion Underlies Coulombic Efficiency Differences in High-Performing Lithium Metal Battery Electrolytes

Solomon T. Oyakhire[1,2‡], Sang Cheol Kim[3‡], Wenbo Zhang[3‡], Sanzeeda Baig Shuchi[1‡], Yi Cui[3,4,5*], Stacey F. Bent [1,4*]

1 Department of Chemical Engineering, Stanford University, Stanford, CA 94305, USA.

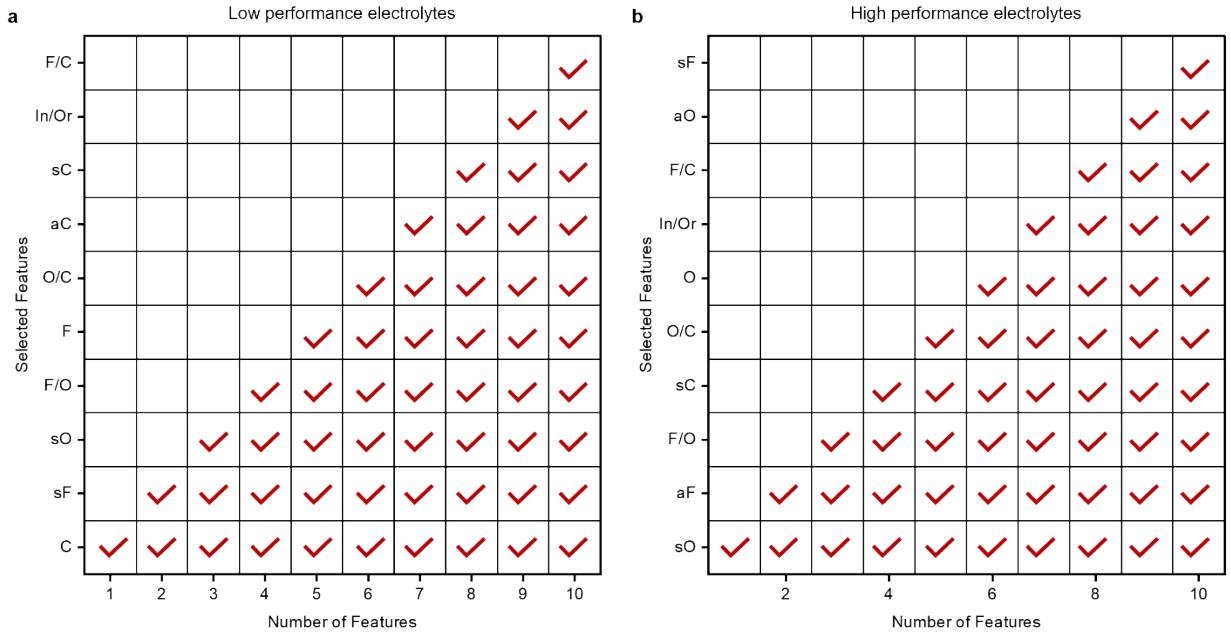
2 Department of Materials Science and Engineering, University of California, Berkeley, CA 94720, USA

3 Department of Materials Science and Engineering, Stanford University, Stanford, CA 94305, USA.

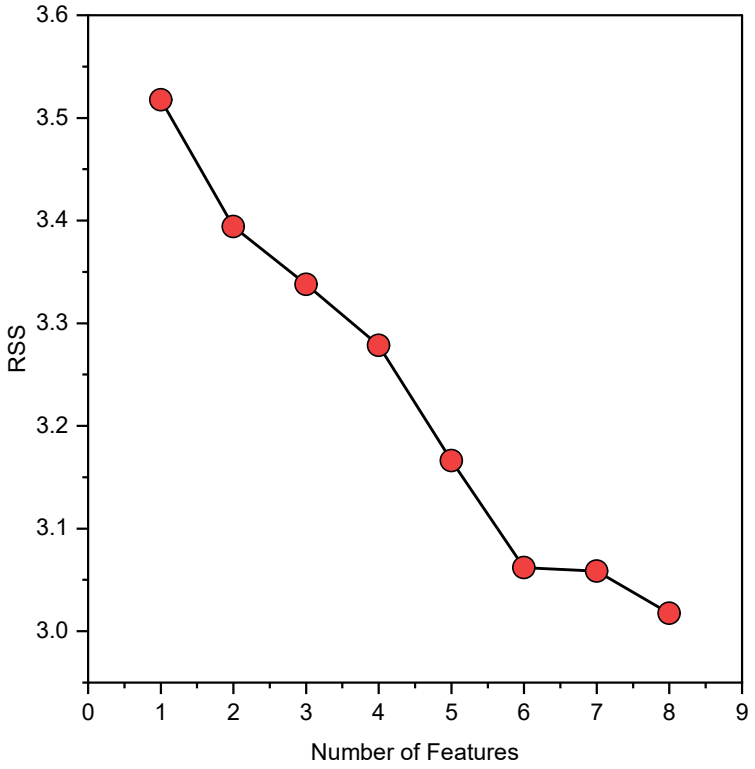
4 Department of Energy Science and Engineering, Stanford University, Stanford, CA 94305, USA.

5 Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA.

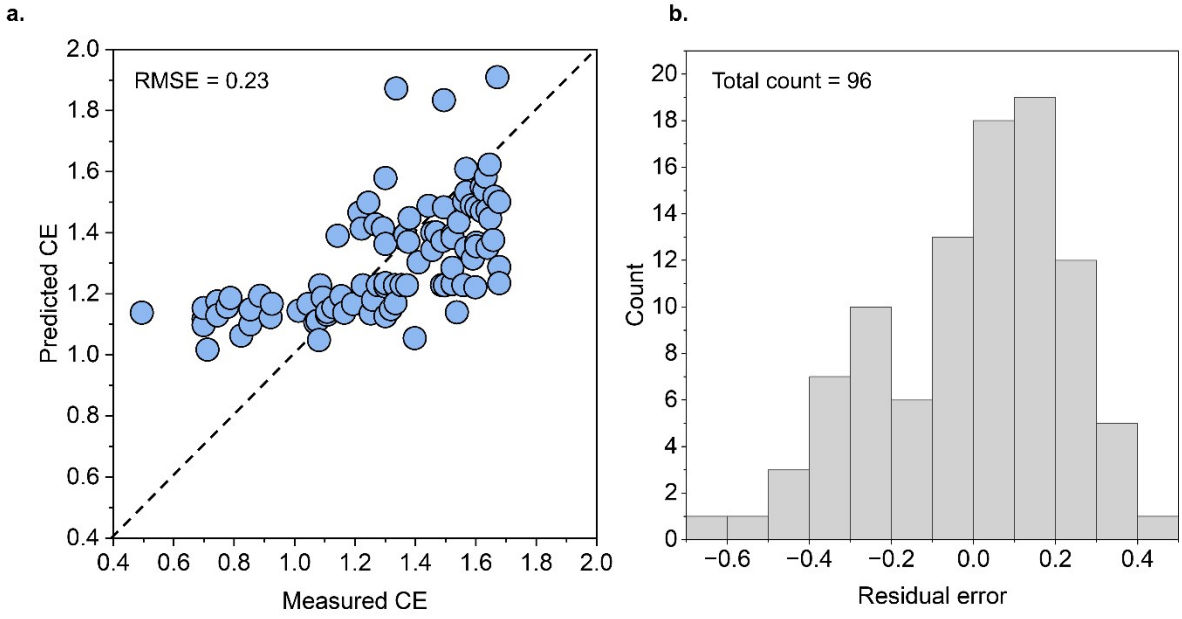
‡ equal contributions.



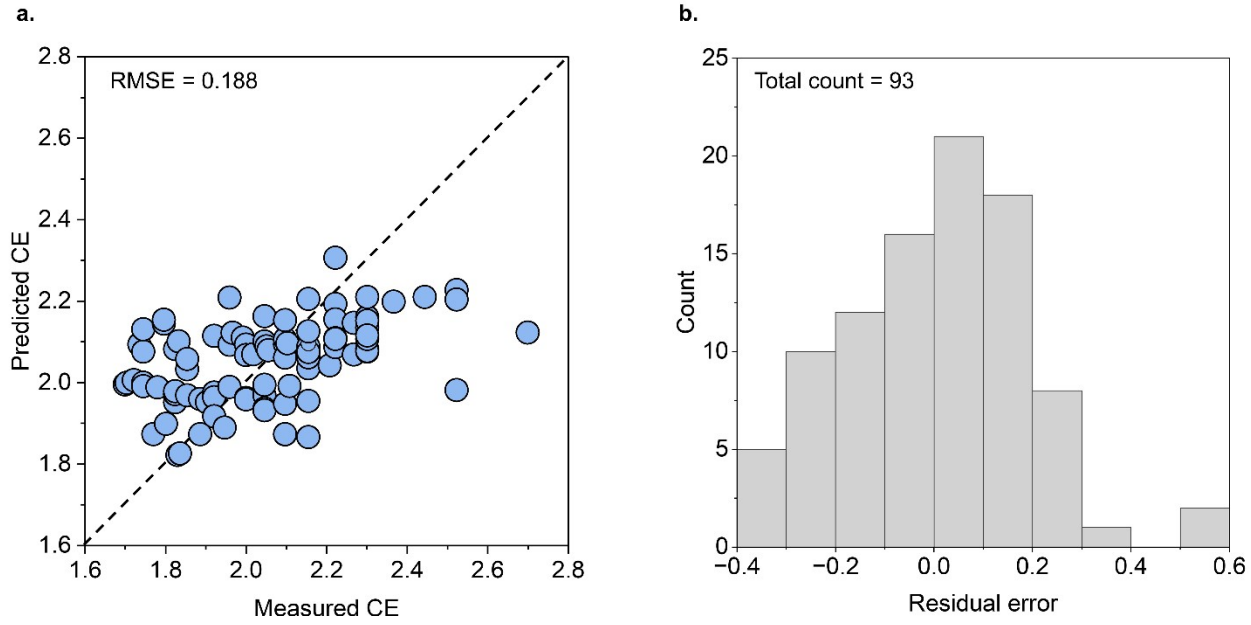
Supplementary Figure 1: **a, b** Forward stepwise selection applied to low and high-performance electrolytes, respectively.



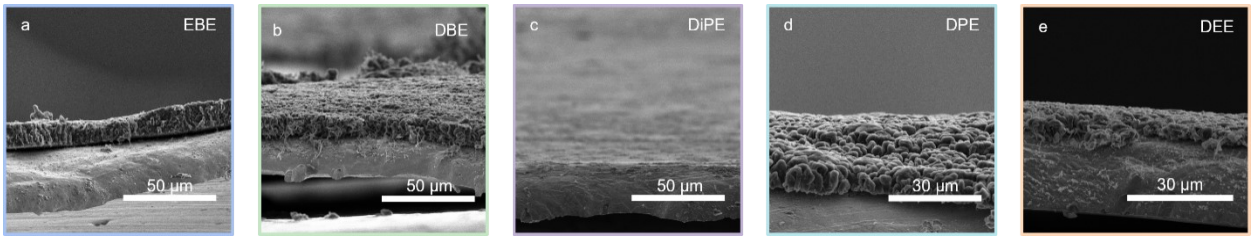
Supplementary Figure 2: Residual sum of squares as a function of number of features for the high-performance electrolytes.



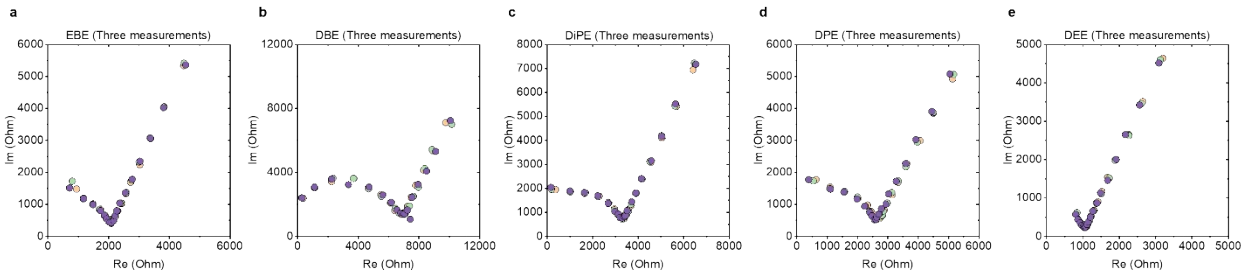
Supplementary Figure 3: Prediction performance of the four-feature model on low-performing electrolytes a. Comparison of predicted CE with measured CE. b. Residual error for all low-performing electrolytes, calculated as the difference between measured and predicted CE.



Supplementary Figure 4: Prediction performance of the four-feature model on high-performing electrolytes a. Comparison of predicted CE with measured CE. b. Residual error for all high-performing electrolytes, calculated as the difference between measured and predicted CE.



Supplementary Figure 5: Cross sectional view of lithium morphology formed in our high-performing electrolytes.



Supplementary Figure 6: Replicate Nyquist plots of lithium-ion conductivity measurements in symmetric stainless-steel Swagelok cells for our new electrolytes synthesized using 1M LiFSI in **a.** EBE, **b.** DBE, **c.** DiPE, **d.** DPE, **e.** DEE solvents, respectively.

Supplementary Table 1: Complete list of derivative features (products and ratios) used for correlation analysis.

Pearson Coefficient	Spearman Coefficient	Feature name	Feature formula
-0.99	-0.9	corrosion	$\int_0^{48} idt$
-0.35	-0.5	b	$\frac{Overpotential}{Impedance\ (t = 0h)}$
-0.18	-0.4	f	$\frac{Overpotential}{sO}$
0.42	0.8	o	$sO * F/O$
-0.14	-0.4	e	$\frac{Overpotential}{F/O}$
-0.25	-0.9	i	$Overpotential * Impedance\ (t = 0h)$
-0.52	-0.4	n	$\frac{F/O}{sO}$
0.52	0.8	q	$SEI\ anion\ content * \Delta R^{@}(24h)$
0.46	0.6	s	$\frac{SEI\ anion\ content}{F/O}$
-0.91	-0.7	v	$\frac{F/O}{\Delta R(24h)}$
0.91	0.9	x	$sO * \Delta R(24h)$
-0.81	-0.9	d	$\frac{Overpotential}{\Delta R(24h)}$

@ ΔR (t) represents SEI impedance measured at time t relative to SEI impedance measured at time t = 0h