Supporting Information

Data-driven Electrolyte Design for Lithium Metal Anodes

Sang Cheol Kim[1†], Solomon T. Oyakhire[2†], Constantine Athanitis[1], Jingyang Wang[1], Zewen Zhang[1], Wenbo Zhang[1], David T. Boyle[3], Mun Sek Kim[2], Zhiao Yu[3], Xin Gao[1], Tomi Sogade[1], Esther Wu[1], Jian Qin[2], Zhenan Bao[2], Stacey F. Bent[2,4*], Yi Cui[1,4,5*]

[1]Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA.

[2]Department of Chemical Engineering, Stanford University, Stanford, California 94305, USA.

[3]Department of Chemistry, Stanford University, Stanford, California 94305, USA.

[4]Department of Energy Science and Engineering, Stanford University, Stanford, California 94305, USA.

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

[†]These authors contributed equally to this work

[*]Corresponding authors. Y.C., yicui@stanford.edu; S.F.B., sbent@stanford.edu

Supporting Note

Feature Data Extraction

The process of feature data extraction is detailed in the Methods section. As an example to illustrate the process, the solvent carbon ratio of 1 M LiTFSI DOL-DME (1:1 vol) 3 wt% $LiNO_3$ is calculated in this Note. First, the molecular weight (MW) and density of each of the solvents is acquired. DME has a MW of 90.12 g/mol and a density of 0.868 g/mL, and DOL has a MW of 74.08 g/mol and a density of 1.06 g/mL. The volumetric ratio of the solvents is 1:1, therefore we can calculate the elemental makeup of the solvent containing 0.5 L of DOL and 0.5 L of DME. The elemental makeup of 1 M LiTFSI can be easily calculated by taking the elemental makeup of 1 mole of salt. It is assumed that the salt content does not significantly alter the total volume. Then the lithium nitrate additive is also included as salt. The 3% of the total weight of the solvents is taken as the mass of $LiNO_3$, which is converted into moles. When all the components are summed up, 1 liter of electrolyte has 42.73 moles of carbon, 91.09 moles of hydrogen, 29.34 moles of oxygen, 6 moles of fluorine, 1.47 moles of nitrogen, 2 moles of sulfur, and 1 mole of lithium, adding up to 174.09 moles of atoms. The number of carbon atoms in the solvent is 40.73 moles. Taking the ratio of 40.73 and 174.09 results in a solvent carbon ratio of 0.234.

Supporting Figures

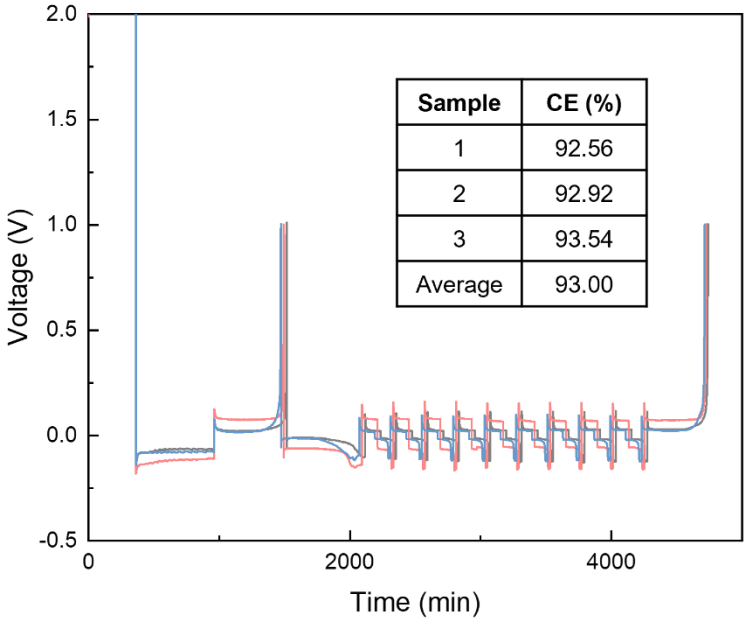


Fig. S1. Coulombic efficiency measurement of 1 M LiFSI EC-DEC.

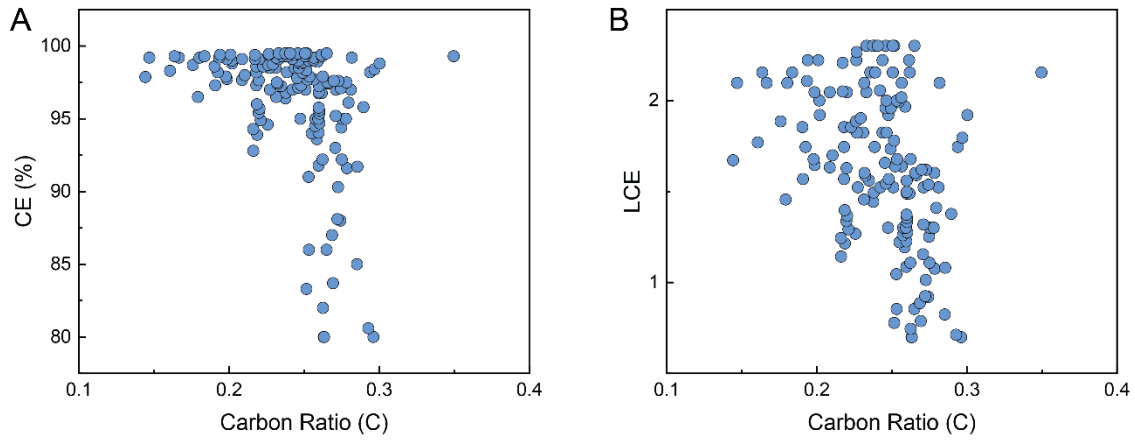


Fig. S2. Scatterplot of the data set showing carbon ratio versus a) CE and b) LCE.

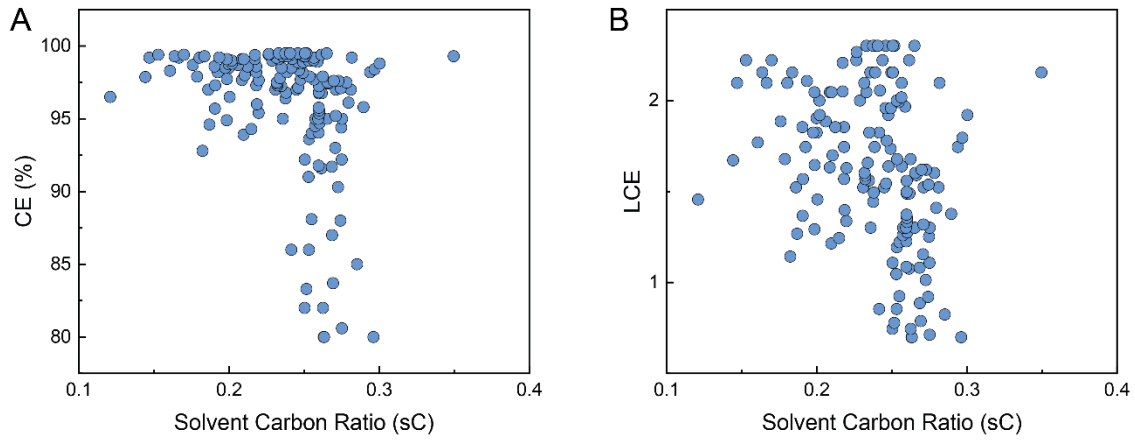


Fig. S3. Scatterplot of the data set showing solvent carbon ratio versus a) CE and b) LCE.

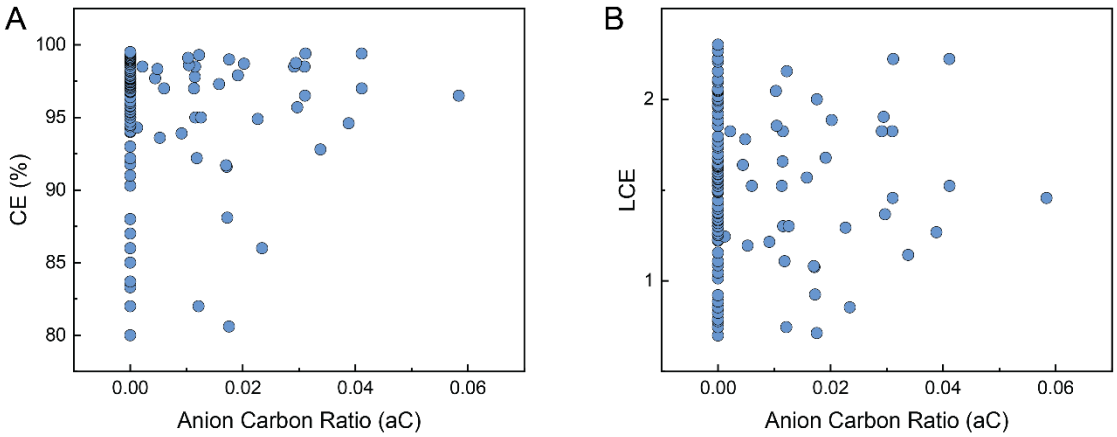


Fig. S4. Scatterplot of the data set showing anion carbon ratio versus a) CE and b) LCE.

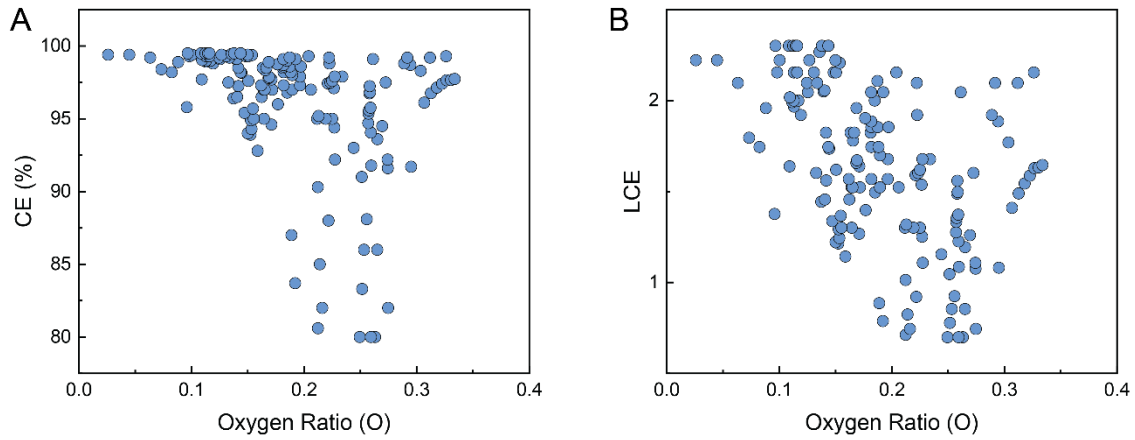


Fig. S5. Scatterplot of the data set showing oxygen ratio versus a) CE and b) LCE.

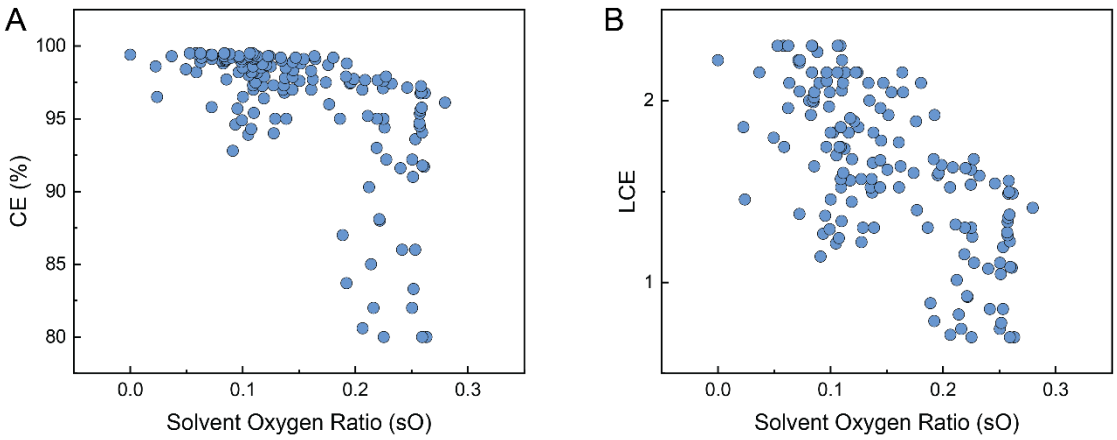
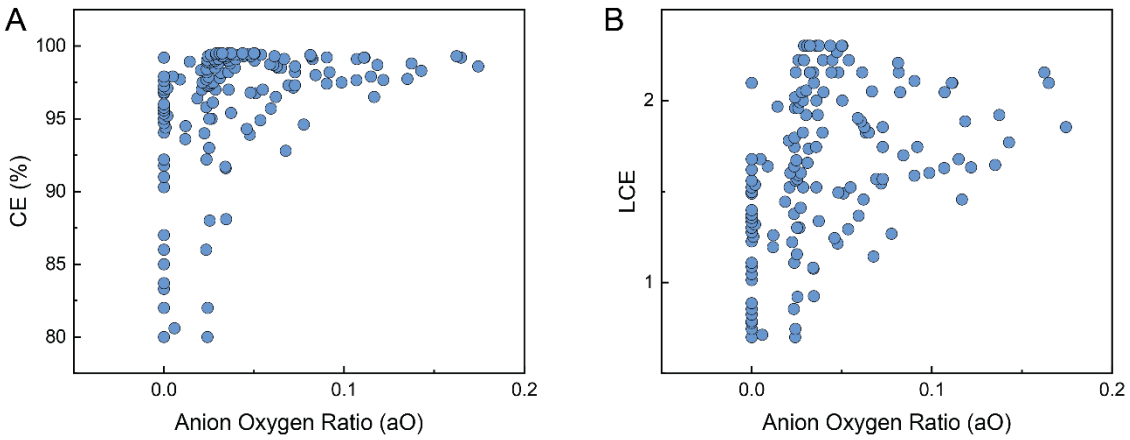


Fig. S6. Scatterplot of the data set showing solvent oxygen ratio versus a) CE and b) LCE.



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69 **Fig. S7.** Scatterplot of the data set showing anion oxygen ratio versus a) CE and b) LCE.

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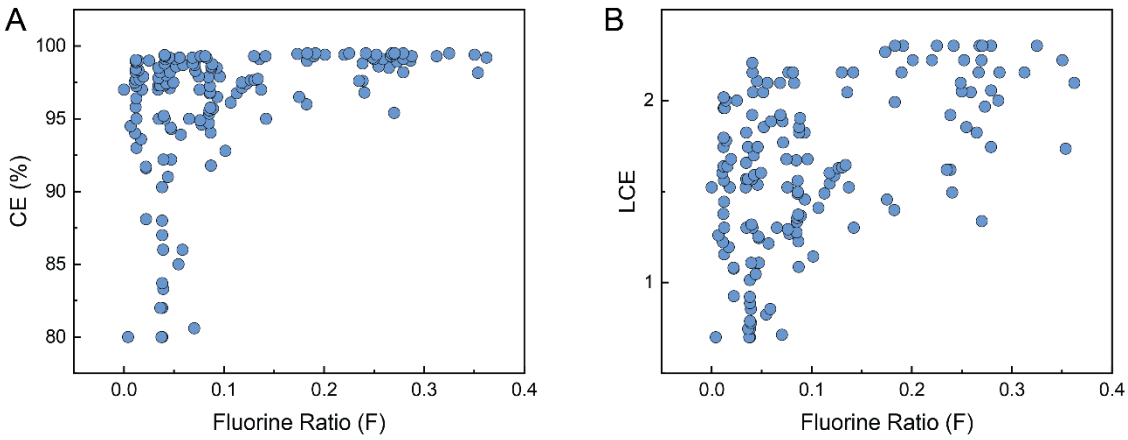

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73 **Fig. S8.** Scatterplot of the data set showing fluorine ratio versus a) CE and b) LCE.

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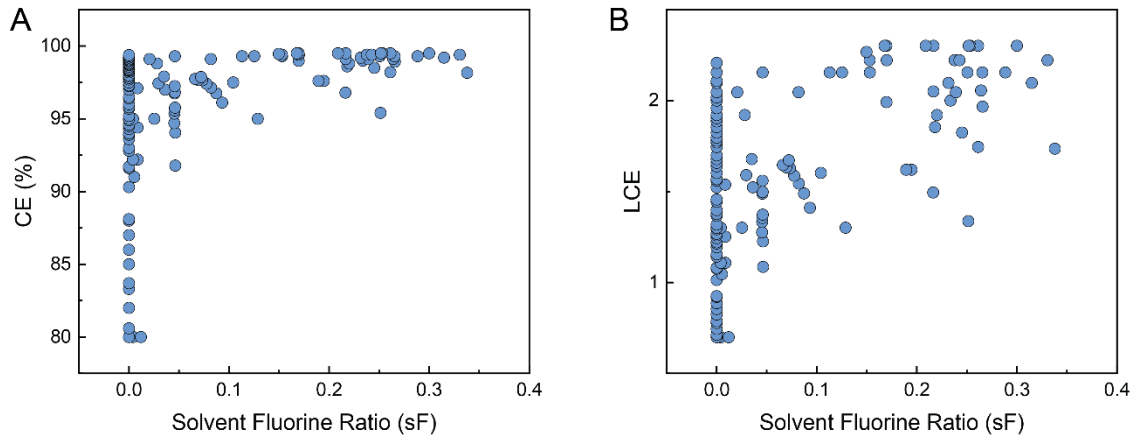

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76 **Fig. S9.** Scatterplot of the data set showing solvent fluorine ratio versus a) CE and b) LCE.

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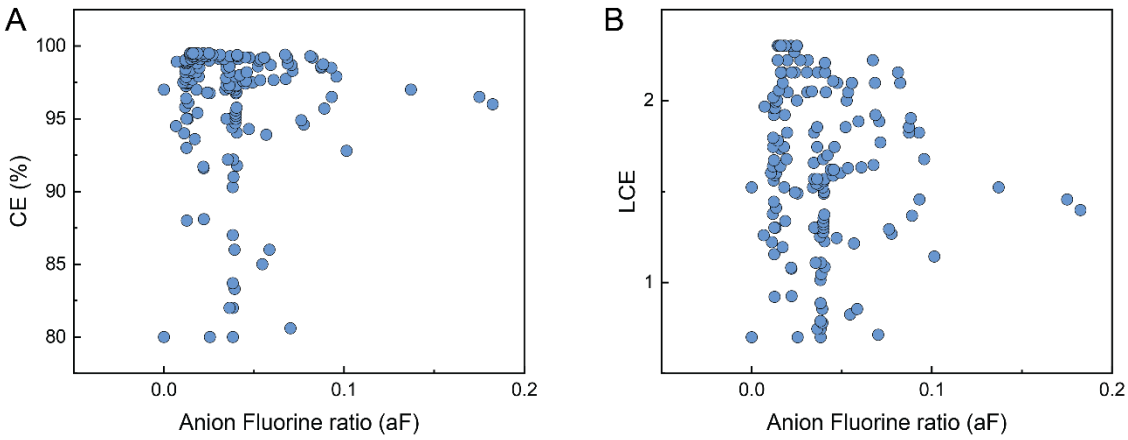

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79 **Fig. S10.** Scatterplot of the data set showing anion fluorine ratio versus a) CE and b) LCE.

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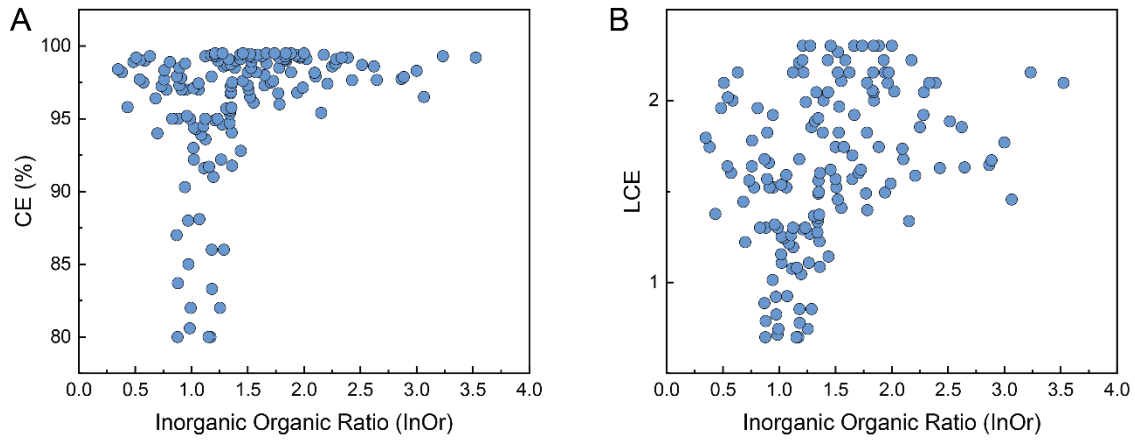

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83 **Fig. S11.** Scatterplot of the data set showing inorganic to organic ratio versus a) CE and b) LCE.

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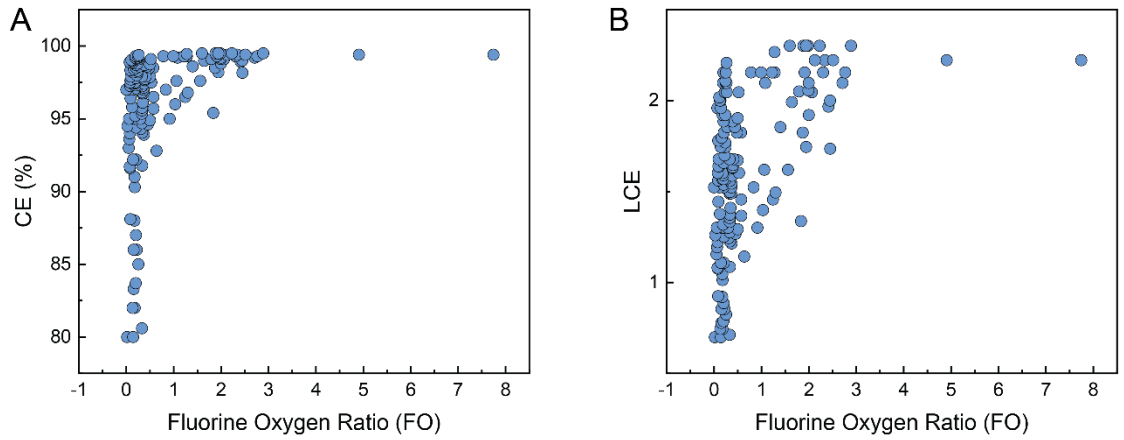

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87 **Fig. S12.** Scatterplot of the data set showing fluorine to oxygen ratio versus a) CE and b) LCE.

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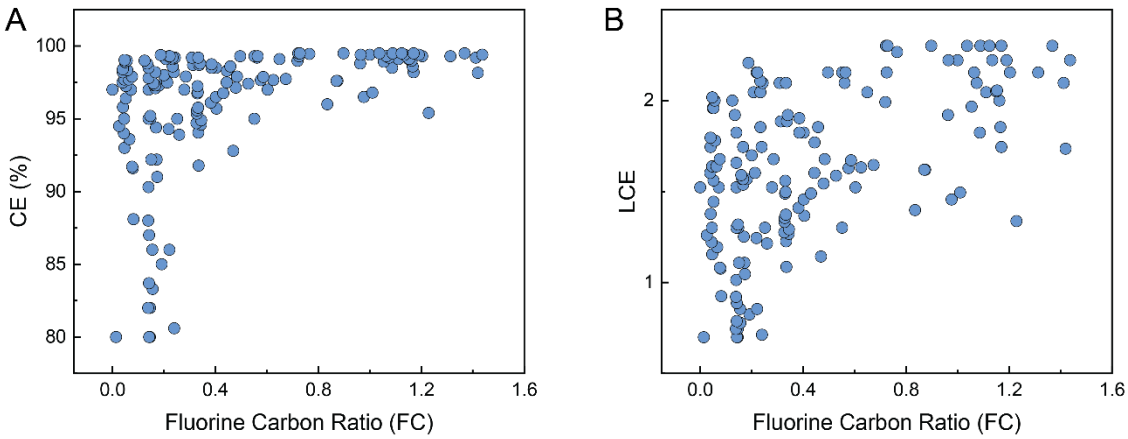

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90 **Fig. S13.** Scatterplot of the data set showing fluorine to carbon ratioe versus a) CE and b) LCE.

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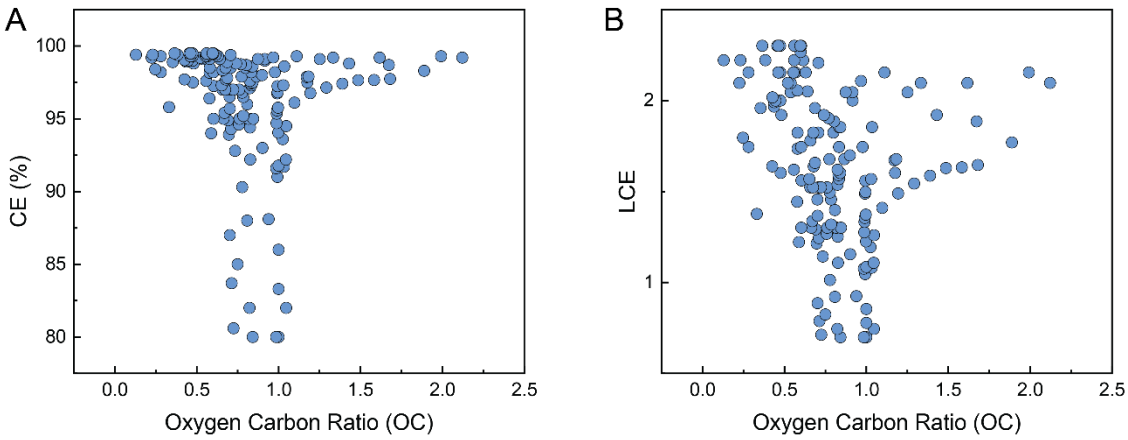

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94 **Fig. S14.** Scatterplot of the data set showing oxygen carbon ratio versus a) CE and b) LCE.

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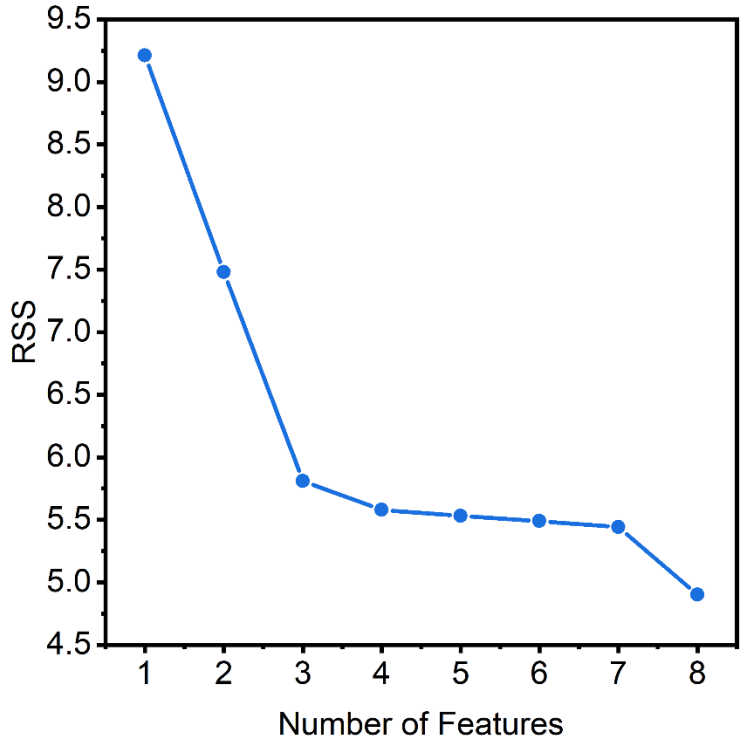
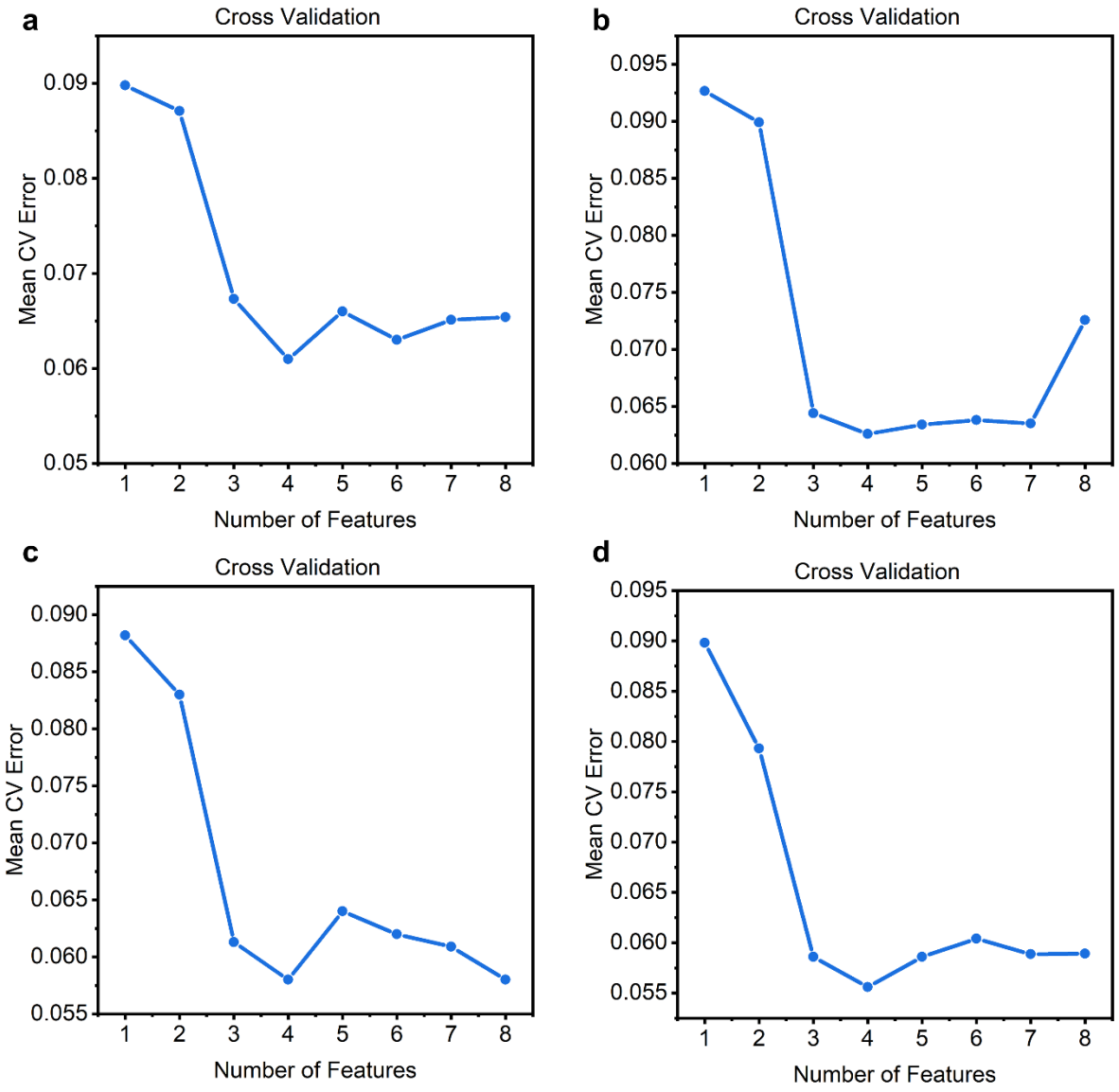


Fig. S15. Residual sum of squares (RSS) of subset selection models as a function of number of features.



114

115 **Fig. S16.** Cross-validation errors as a function of number of features for four random partitions of the
116 training data.

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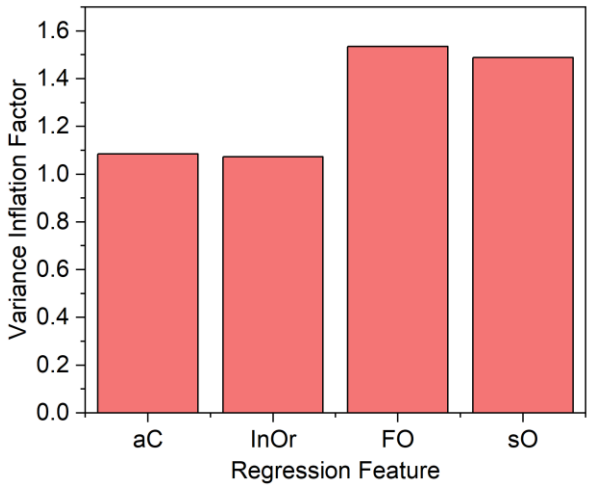

118

119 **Fig. S17.** Variation inflation factors for the four features in the final regression model.

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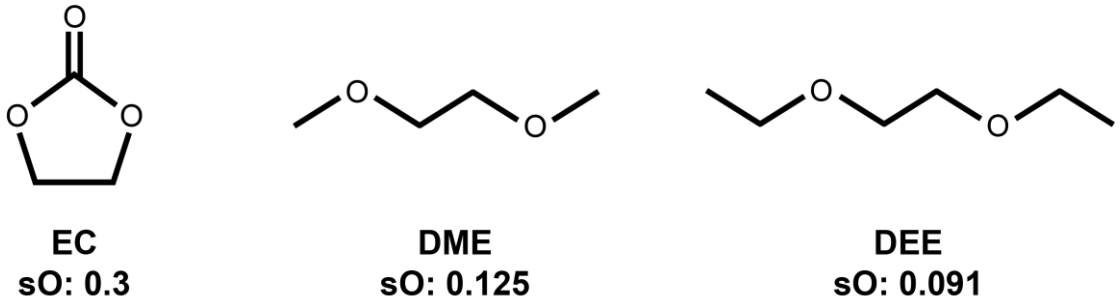


Fig. S18. Molecular structures of carbonates and ethers.

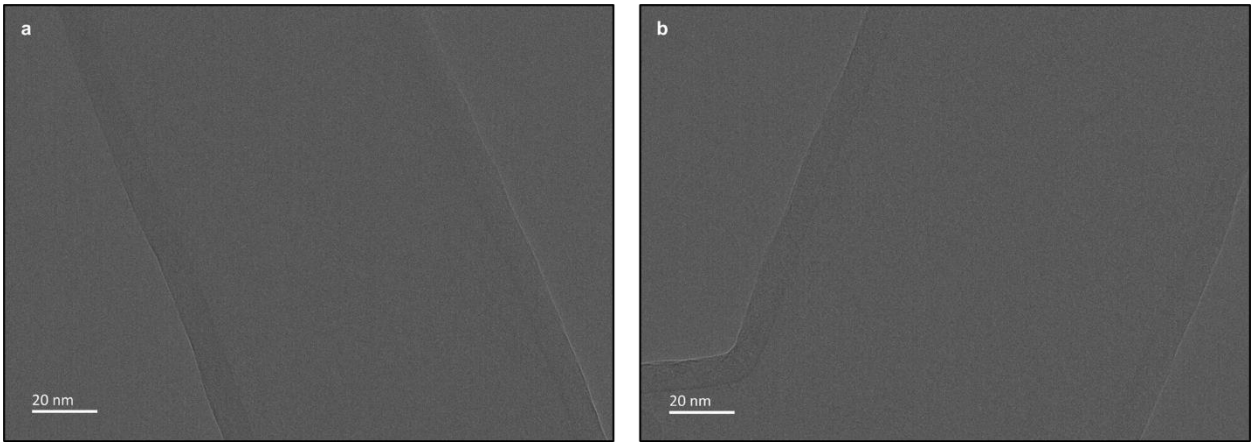


Fig. S19. Cryo-HR-TEM images of lithium deposits using 1 M LiFSI DME.

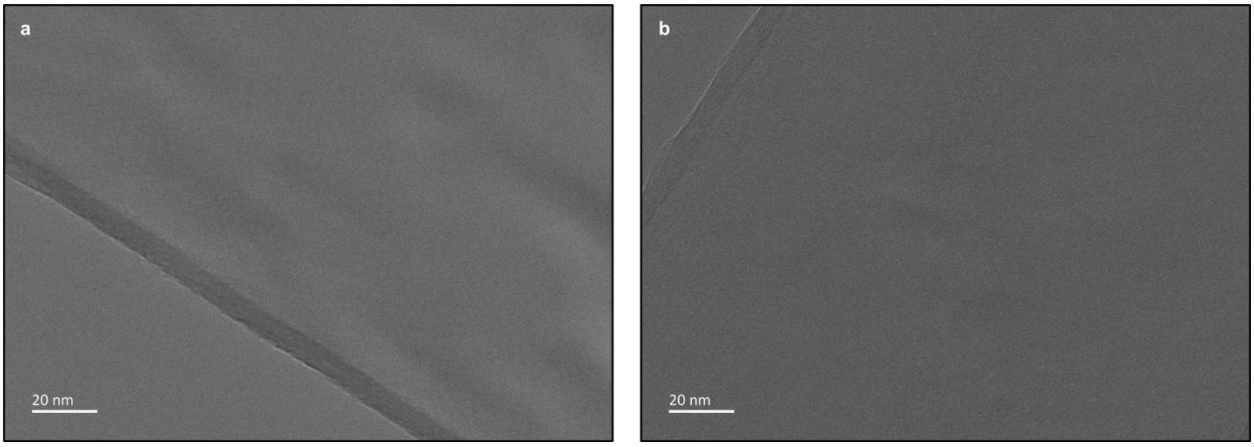


Fig. S20. Cryo-HR-TEM images of lithium deposits using 1 M LiFSI MTBE-toluene.

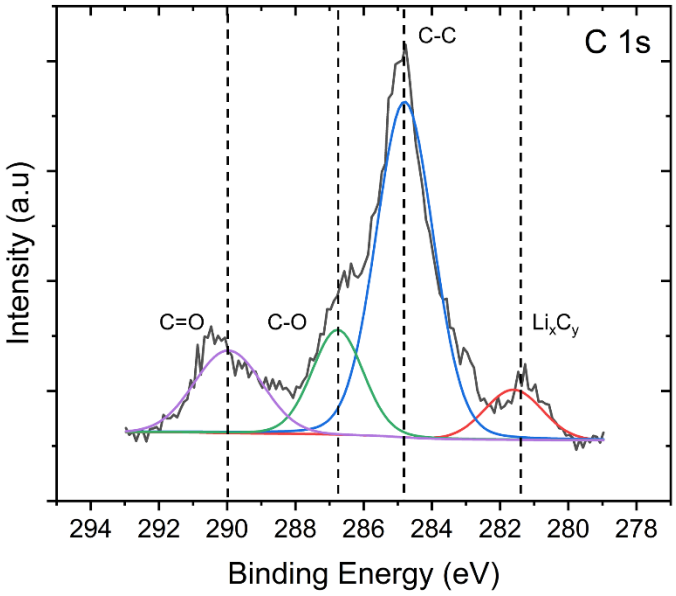


Fig. S21. XPS high resolution signals of C 1s species in the SEI formed using 1 M LiFSI MTBE-toluene electrolyte.

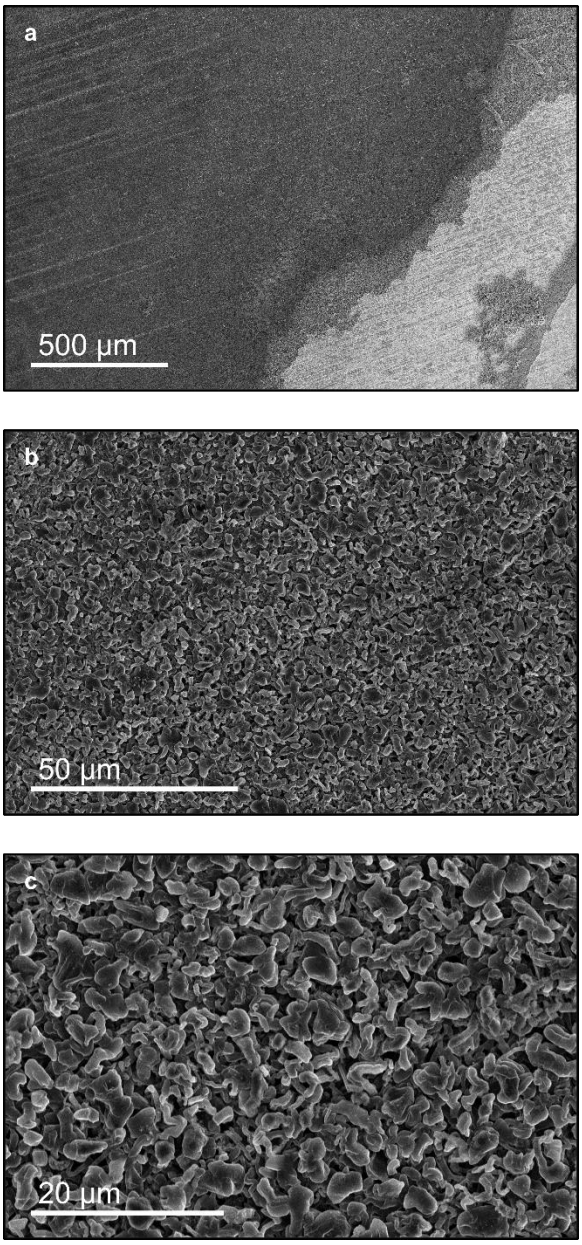


Fig. S22. Scanning electron microscopy (SEM) images of 1 M LiFSI DME.

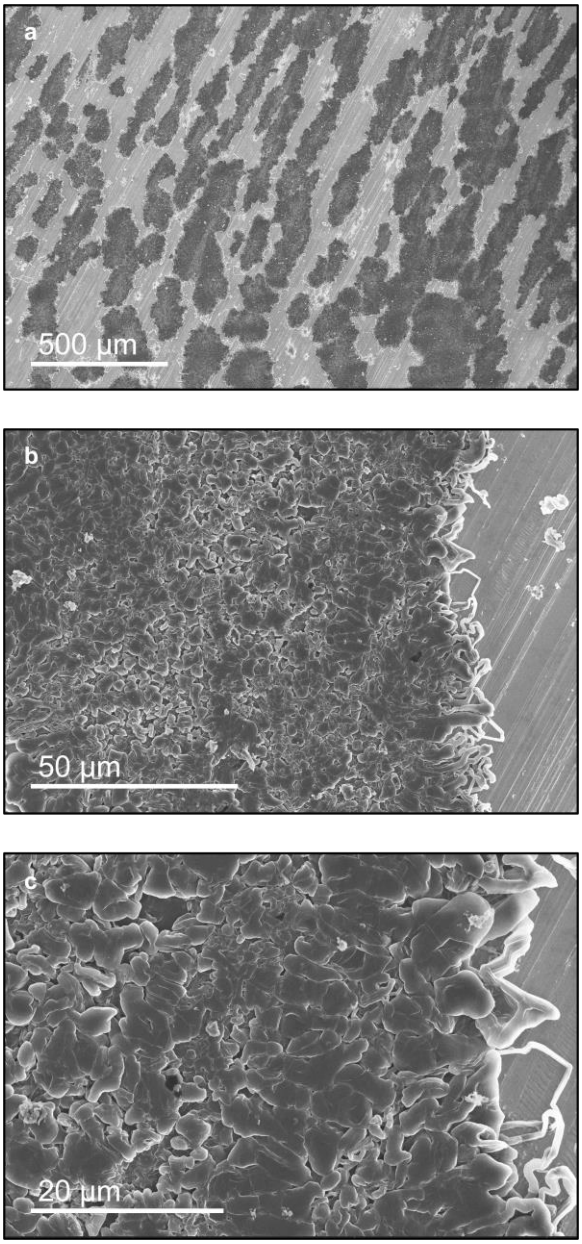
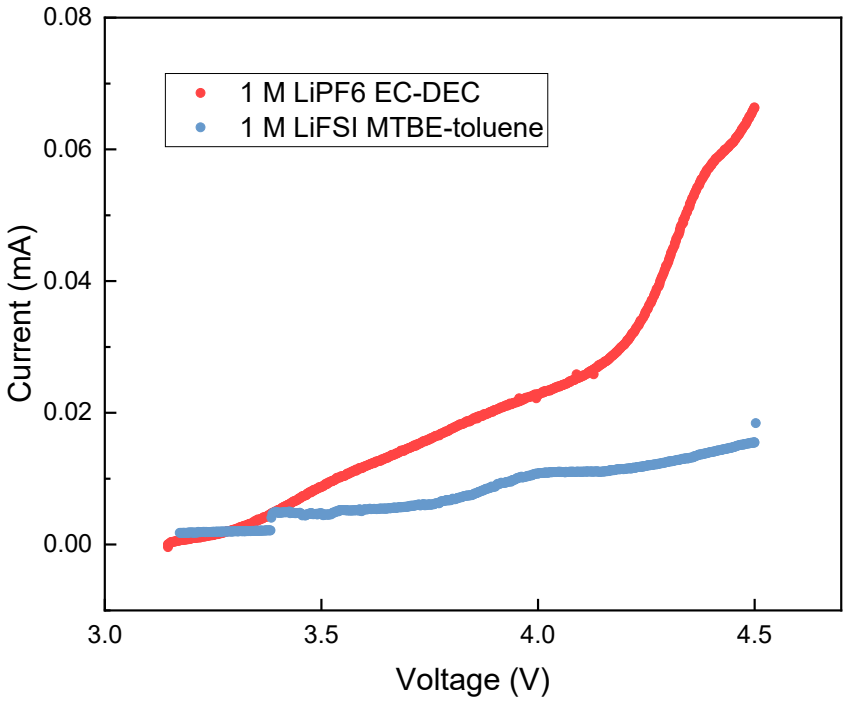


Fig. S23. Scanning electron microscopy (SEM) images of 1 M LiFSI MTBE-toluene.



152

153 **Fig. S24.** Li|Al oxidative stability of our fluorine-free electrolyte (1 M LiFSI MTBE-Toluene) compared
154 to a common carbonate electrolyte (1 M LiPF6 in 1:1 wt% of ethylene carbonate and diethyl carbonate)
155 using linear sweep voltammetry at a sweep rate of 10 mV s^{-1}.

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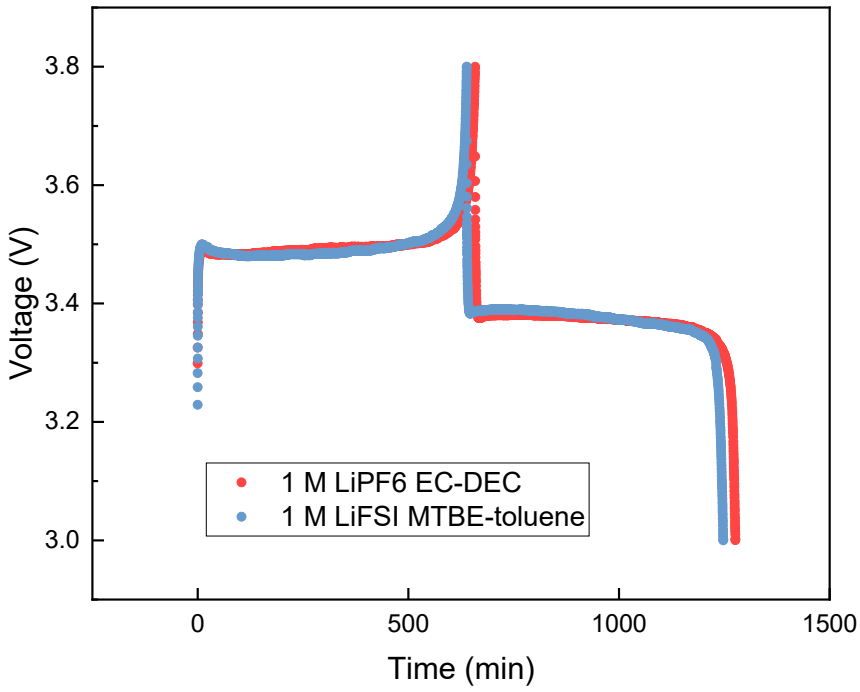

157

158 **Fig. S25.** First cycle of charge and discharge using LFP-Li cells show that our electrolyte is stable against
159 LFP cathodes.

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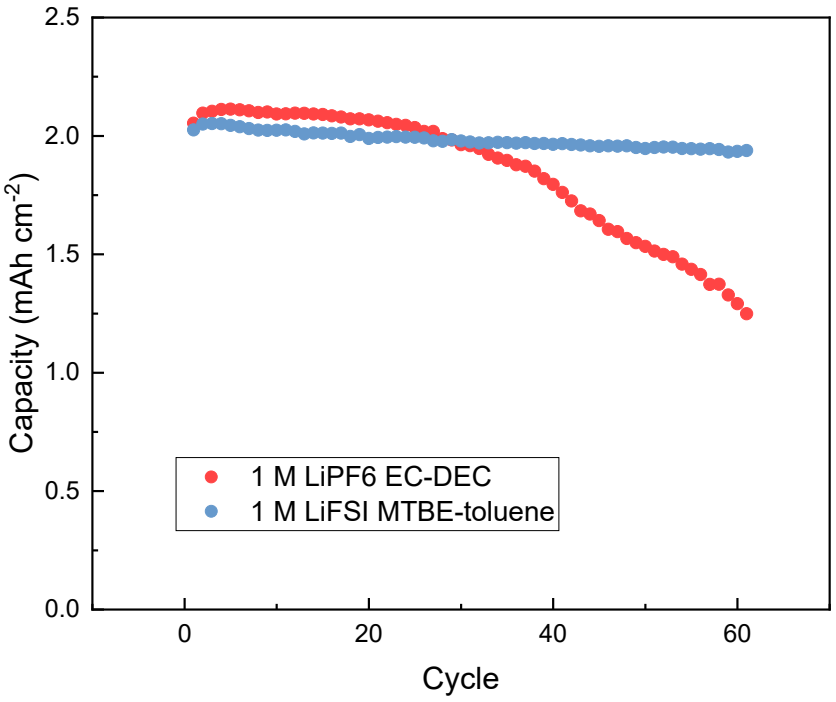

Fig. S26. LFP-Li coin cell cycling between 3.0 - 3.8 V using 0.2 mA cm^{-2}, showing superior cycling stability of the 1 M LiFSI MTBE-toluene electrolyte. 750 µm lithium electrodes were used in these experiments.

Supporting Tables

Table S1. Acquired training and test dataset for the machine learning model.

Electrolyte	CE (%)	LCE	Method	Current (mA/cm2)	Capacity (mAh/cm2)	Cycle
1 M LiClO4-PC + 5% FEC	80	0.699	Cycle	0.5	0.083	30
1 M LiPF6 EC-DMC (1:1 v) 2% VC	80	0.699	Cycle	0.6	0.44	50
1 M LiBF4 EC-DMC (1:1 v) 2% VC	80	0.699	Cycle	0.6	0.44	50
1M LiTFPFB PC	80.6	0.712	Cycle	0.5	1	50
1 M LiPF6 EC-EMC (3:7 wt) 2% wt VC	82	0.745	Aurbach	0.5	1	10
1 M LiTFSI EC-DMC (1:1 v) 2% VC	82	0.745	Cycle	0.6	0.44	50
1M LiPF6 EC-DMC (3:7 v/v)	83.3	0.777	Aurbach	0.4	0.8	10
1M LiPF6 EC-DEC (1:2 w/w)	83.7	0.788	Cycle	1	1	40
1.5 M LiAsF6-PC	85	0.824	Cycle	1	1	
1 M LiBETI EC-DMC (1:1 v) 2% VC	86	0.854	Cycle	0.6	0.44	50
1 M LiPF6 EC-DMC (1:2 vol)	86	0.854	Cycle	0.5	0.5	100
1 M LiPF6 EC-DEC (3:7 wt)	87	0.886	Cycle	0.1	0.1	160
1 M LiPF6-EC/DEC (1:1 vol.)	88	0.921	Cycle	0.5	0.5	100
0.6M LiTFSI 0.4M LiBOB EC-EMC (5:5 wt)	88.1	0.924	Aurbach	0.5	1	10
1M LiPF6 EC-DEC (1:1 w/w)	90.3	1.013	Cycle	1	1	40
1 M LiPF6 EC-DMC (1:2 vol) 2% TTFEB	91	1.046	Cycle	0.5	0.5	100
0.6M LiTFSI 0.4M LiBOB EC-EMC (7:3 wt)	91.6	1.076	Aurbach	0.5	1	10
0.6M LiTFSI 0.4M LiBOB EC-EMC (9:1 wt)	91.7	1.081	Aurbach	0.5	1	10
1 M LiPF6 FEC-DMC (1:1 v)	91.78	1.085	Cycle	1	1	50
1 M LiPF6 FEC-EC-DEC (2:9:9 vol)	92.2	1.108	Cycle	0.5	1	150
1 M LiTFSI EC-DMC (1:1 v) 5wt% FEC	92.2	1.108	Cycle	0.5	0.5	50
3.57 M LiTFSI DME	92.8	1.143	Aurbach	1	1	12
1M LiFSI EC/DEC (v 1:1)	93	1.155	Aurbach	0.5	1	10
0.1M LiDFP 0.4M LiTFSI EC-DMC (3:7 v/v)	93.6	1.194	Aurbach	0.4	0.8	10
0.84 M LiTFSI 2.7M LiDFP DME	93.9	1.215	Aurbach	1	1	12
1 M LiFSI in tetraglyme	94	1.222	Cycle	0.1	0.2	30
1 M LiPF6 FEC-DMC (1:1 v) 0.25wt% 12-Crown-4	94.05	1.225	Cycle	1	1	50
0.11 M LiTFSI 3.9 M LiDFP DME	94.3	1.244	Aurbach	1	1	12
1 M LiPF6 FEC-EC-DEC (2:9:9 vol) 0.05 M RbNO3 0.05M 18-crown-6	94.4	1.252	Cycle	0.5	1	150
0.1M LiDFP 0.4M LiFSI EC-DMC (3:7 v/v)	94.5	1.260	Aurbach	0.4	0.8	10
2M LiTFSI 2M LiDFOB DME	94.6	1.268	Aurbach	0.5	1	10
1 M LiPF6 FEC-DMC (1:1 v) 2wt% 12-Crown-4	94.7	1.276	Cycle	1	1	50

2.2 M LiTFSI 0.8 M LiDFP DME	94.9	1.292	Aurbach	1	1	12
1 M LiPF6-EC/DEC (1:1 vol.) + 5 vol% FEC	95	1.301	Cycle	0.5	0.5	100
0.02M Li2S5 0.15M LiNO3 1M LiTFSI DOL-DME (1:1 vol)	95	1.301	Cycle	0.5	0.5	100
1 M LiFSI in DEG-FTriEG	95	1.301	Cycle	0.1	0.2	30
1 M LiDFOB EC-DEC (3:7 wt)	95	1.301	Cycle	0.1	0.1	500
1 M LiPF6 FEC-EMC-DMC (34.6:57.2:8.2 wt)	95	1.301	Cycle	1	1	100
1 M LiPF6 in EC/DEC + 0.15M LiDFP	95.2	1.319	Aurbach	0.5	1	10
1 M LiPF6 FEC-DMC (1:1 v) 1.75wt% 12-Crown-4	95.35	1.333	Cycle	1	1	50
LiFSI-1.4DME-3TFEB	95.4	1.337	Aurbach	0.5	1	10
1 M LiPF6 FEC-DMC (1:1 v) 1.5wt% 12-Crown-4	95.58	1.355	Cycle	1	1	50
3M LiTFSI DME	95.7	1.367	Aurbach	0.5	1	10
1 M LiPF6 FEC-DMC (1:1 v) 0.5wt% 12-Crown-4	95.76	1.373	Cycle	1	1	50
1 M LiFSI THF	95.8	1.377	Cycle	1	1	40
6 M LiPF6 EC-DEC (1:1 vol)	96	1.398	Cycle	0.5	1	90
1 M LiFSI FEC	96.11	1.410	Cycle	0.25	0.5	100
0.5 M LiFSI+0.5 M LiPO2F2/DME	96.4	1.444	Cycle	0.5	1	170
3.25M LiTFSI SL	96.5	1.456	Cycle	0.5	1	100
5M LiTFSI 1-4 (EA-DCM vol)	96.5	1.456	Cycle	0.2	0.5	50
1 M LiPF6 FEC-DMC (1:1 v) 1.25wt% 12-Crown-4	96.75	1.488	Cycle	1	1	50
2 M LiFSI FEC	96.76	1.489	Cycle	0.25	0.5	100
LiFSI-1.2DME-3BTFEC	96.8	1.495	Aurbach	0.5	1	10
1 M LiPF6 FEC-DMC (1:1 v) 0.75wt% 12-Crown-4	96.82	1.498	Cycle	1	1	50
1M LiPF6 FEC-DEC (1:1 w/w)	97	1.523	Cycle	1	1	40
0.1 M Li2S5 + 5% LiNO3 in 1 M LiTFSI in DOL/DME (1:1 vol)	97	1.523	Cycle	1	1	230
3 M LiFNSI in DOL/DME	97	1.523	Cycle	0.5	1.5	180
0.5 M LiTFSI 0.5 M LiNO3 DOL-DME (1:1 v)	97	1.523	Cycle	0.5	0.5	100
1 M LiClO4 EC-ES (1:1 vol)	97	1.523	Cycle	0.5	0.5	450
1 M LiPF6 FEC-EC-DEC (2:9:9 vol) 0.1 M RbNO3 0.1M 18-crown-6	97.1	1.538	Cycle	0.5	1	150
3 M LiFSI FEC	97.14	1.544	Cycle	0.25	0.5	100
1 M LiPF6 FEC-DMC (1:1 v) 0.1wt% 12-Crown-4	97.24	1.559	Cycle	1	1	50
1M LiFSI DME	97.25	1.561	Aurbach	0.5	1	10
3.2M LiFSI TEP	97.3	1.569	Aurbach	0.5	1	10
1 M LiTFSI in DOL/DME with 5wt% TMS-FNFSI	97.3	1.569	Cycle	0.5	1	110
LiFSI SL (1:3 molar)	97.3	1.569	Cycle	1	0.5	350
4 M LiFSI FEC	97.41	1.587	Cycle	0.25	0.5	100
1M LiFSI FEC-DEC (1:2 by volume)	97.43	1.590	Aurbach	0.5	1	10

1 M LiFSI/EC-ETFEC (3:7 vol)	97.5	1.602	Aurbach	0.25	0.5	10
5 M LiFSI PC	97.5	1.602	Cycle	0.5	1	100
1 M LiFSI 1,4-Dioxolane	97.5	1.602	Cycle	1	1	40
1M LiPF6 FEC-TFEC (1:1 w/w)	97.6	1.620	Cycle	1	1	40
1 M LiPF6 MTFP-FEC (9:1 vol)	97.6	1.620	Aurbach	0.5	1	10
5 M LiFSI FEC	97.65	1.629	Cycle	0.25	0.5	100
6 M LiFSI FEC	97.67	1.633	Cycle	0.25	0.5	100
1M LiFSI DMB	97.7	1.638	Aurbach	0.5	1	10
1 M LiPF6 EC-DMC (1:1 v), 0.5 M LiNO3 TEGDME, 70%	97.7	1.638	Cycle	0.5	1	80
7 M LiFSI FEC	97.74	1.646	Cycle	0.25	0.5	100
1M LiTFSI DME-DOL (1:1) 3% LiNO3	97.8	1.658	Cycle	0.5	1	350
1 M LiFSI FSA	97.87	1.672	Cycle	0.5	0.5	200
4.6 M LiFSI 2.3 M LiTFSI DME	97.9	1.678	Cycle	0.5	0.5	200
1 M LiPF6 EC-DMC (1:1 v), 0.5 M LiNO3 TEGDME, 50%	97.9	1.678	Cycle	0.5	1	200
1 M LiPF6 FEC-EMC-DMC (34.6:57.2:8.2 wt) 1wt% TMSB	97.9	1.678	Cycle	1	1	100
LiFSI SL (1:2.5 molar)	98	1.699	Cycle	1	0.5	450
1M LiFSI FEC-FEMC-D2 (234:280:793 by mol)	98.16	1.735	Aurbach	0.5	1	10
1 LiFSI - 3 TMS	98.2	1.745	Cycle	0.5	1	150
1.2M LiFSI DMC-BTFE (1:2 mol)	98.2	1.745	Cycle	0.5	1	60
4.6 M LiFSI DME	98.2	1.745	Cycle	0.5	0.5	200
1 M LiFSI MeTHF	98.2	1.745	Cycle	1	1	40
LiFSI DMC (1:1.5 mol)	98.3	1.770	Cycle	1	1	200
0.4M LiTFSI 0.6M LiNO3 DOL-DME (1:1 vol)	98.34	1.780	Cycle	0.5	1	250
1 M LiFSI DMeTHF	98.4	1.796	Cycle	1	1	40
1.2M LiFSI 0.15M LiDFOB EC-EMC-BTFE (0.175:0.825:2 mol)	98.5	1.824	Cycle	0.5	1	200
3.25M LiTFSI 0.1M LiNO3 SL	98.5	1.824	Cycle	0.5	1	100
3 M LiTFSI 3wt% LiNO3 DOL-DME (1:1 v)	98.5	1.824	Cycle	0.5	1	350
1 M LiTFSI DOL-DME (1:1 vol) 2wt% LiNO3	98.5	1.824	Cycle	0.5	1	150
LiFSI DMC-TTE (1:1.5:1.5 mol)	98.6	1.854	Aurbach	0.5	1	10
2M LiFSI 1M LiTFSI DOL-DME (1:1 vol)	98.6	1.854	Cycle	0.5	0.8	200
LiFSI-AN-VC (0.52:1:0.09 molar)	98.6	1.854	Aurbach	1	1	10
1M LiFSI 2M LiTFSI DOL-DME (1:1 vol)	98.7	1.886	Cycle	0.5	1	200
6 M LiFSI DMC	98.7	1.886	Cycle	0.5	1	200
3 M LiFSI DOL-DME (1:1 vol)	98.75	1.903	Cycle	1	1.62	100
1 LiFSI - 3 TMS- 3 TTE	98.8	1.921	Cycle	0.5	1	150
1M LiFSI+0.3 M LiNO3-FEC/GBL (1:2 v/v)	98.8	1.921	Cycle	0.5	1	200
8 M LiFSI PC	98.8	1.921	Cycle	0.5	1	200

1 M LiFSI DOL-DME (1:1 vol)	98.9	1.959	Aurbach	0.5	1	10
1 M LiFSI DEE	98.9	1.959	Aurbach	0.5	1	10
0.47M LiFSI DOL-BTFE (1:4 w/w)	98.92	1.967	Aurbach	0.5	1	10
1M LiFSI FDMB	98.98	1.991	Aurbach	0.5	1	10
2.2M LiFSI TEP (= 1:2 MR)	99	2.000	Cycle	0.2	1	140
1.3 M LiTFSI 0.04 M LiNO3 SL-D2 (1:2 wt)	99	2.000	Cycle	0.5	1	100
1.2 M LiFSI DEE	99	2.000	Aurbach	0.5	1	10
1 M LiFSI DEE	99.04	2.018	Aurbach	0.5	1	10
4M LiFSI DME	99.1	2.046	Cycle	0.2	1	>500
5 M LiFSI/EC-ETFEC (3:7 vol)	99.1	2.046	Aurbach	0.25	0.5	10
2 M LiFSI 1 M LiTFSI 3wt% LiNO3 DOL-DME (1:1 v)	99.1	2.046	Cycle	0.5	1	450
LiFSI-1.4TEP-3TTE	99.1	2.046	Aurbach	0.5	1	10
2.54M LiFSI DME-BTFE (1:4 w/w)	99.11	2.051	Aurbach	0.5	1	10
1M LiFSI DMC-BTFE (1:2 by mol)	99.12	2.056	Aurbach	0.5	1	10
1.2M LiFSI TEP-BTFE (1:3 mol)	99.2	2.097	Aurbach	0.5	1	10
5.5M LiFSI DMC	99.2	2.097	Aurbach	0.5	1	10
2M LiPF6 THF-MTHF (1:1 vol)	99.2	2.097	Cycle	0.5	1	100
1 M LiPF6 FEC-FEMC-D2 (2:6:2 wt)	99.2	2.097	Cycle	0.2	1	500
LiFSI-TEP 1:1 molar	99.2	2.097	Cycle	0.2	1	300
10 M LiFSI DMC	99.2	2.097	Cycle	0.5	1	200
4.5 M LiFSI/DME (1 : 1)	99.22	2.108	Cycle	0.5	1	500
LiFSI DME-TTE (1:1.2:3)	99.3	2.155	Aurbach	0.5	1	10
1M LiTFSI in FDMA/FEC (1:1 by vol)	99.3	2.155	Cycle	1	1	100
LiFSI-DME-FB (1:1.2:3 molar)	99.3	2.155	Cycle	1	1	500
LiFSI-TEP 1:1.5 molar	99.3	2.155	Cycle	0.2	1	300
10 M LiFSI EC-DMC	99.3	2.155	Cycle	0.5	1	240
1.33 M LiFSI DME-TTE (1:2 vol)	99.3	2.155	Cycle	0.2	0.5	100
LiFSI-2.2DMC-3TTE	99.3	2.155	Aurbach	0.5	1	10
1.2 M LiFSI F3DEE	99.3	2.155	Aurbach	0.5	1	10
1.2 M LiFSI F6DEE	99.3	2.155	Aurbach	0.5	1	10
4 M LiFSI DEE	99.38	2.208	Aurbach	0.5	1	10
1.28M LiFSI FEC-FEMC-D2	99.4	2.222	Cycle	N/A	2	100
1.2M LiTFSI 1M AN in FM	99.4	2.222	Cycle	0.5	1	200
0.5M LiTFSI 0.5M THF CH3F	99.4	2.222	Cycle	1	1	200
1 M LiFSI DME-FDMH (1:6 molar)	99.4	2.222	Aurbach	0.5	1	10
LiFSI-1.2DME-3BTFE	99.4	2.222	Aurbach	0.5	1	10
1.8 M LiFSI/DME (1 : 1) with HFME at 50% concentration	99.46	2.268	Cycle	0.5	1	500
1.8M LiFSI DEE-BTFE (1:4 w/w)	99.5	2.301	Aurbach	0.5	1	10
1.2M LiFSI DMC-BTFE (1:2 mol)	99.5	2.301	Aurbach	0.5	1	10
1M LiFSI DME-TFEO (1.2:3 mol)	99.5	2.301	Aurbach	0.5	1	10

1.7 M LiFSI MeTHF-TTE (1:1 vol)	99.5	2.301	Cycle	1	1	40
LiFSI-1DME-3TTE	99.5	2.301	Aurbach	0.5	1	10
LiFSI-1.2DME-3TFEO	99.5	2.301	Aurbach	0.5	1	10
1.2 M LiFSI F4DEE	99.5	2.301	Aurbach	0.5	1	10
1.2 M LiFSI F5DEE	99.5	2.301	Aurbach	0.5	1	10

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Table S2. Feature data and predicted efficiencies of candidate electrolytes for model verification.

No.	Electrolyte	FO	lnOr	sO	aC	Pred. LCE	Pred. CE
1	1 M LiFSI EC/DMC (1:1 v/v)	0.032	1.155	0.255	0.000	1.187	0.935
2	1 M LiFSI FEC/DMC (1:1 v/v)	0.209	1.413	0.256	0.000	1.241	0.943
3	1 M LiFSI EC/FEC/DMC (1:1:1 v/v/v)	0.147	1.335	0.265	0.000	1.184	0.935
4	1 M LiFSI DME/TEGDME(1:1 v/v)	0.073	0.692	0.124	0.000	1.691	0.980
5	1 M LiFSI TEGDME/DOL (1:1 v/v)	0.062	0.756	0.147	0.000	1.598	0.975
6	1 M LiFSI DME/TEGDME/DOL (1:1:1 v/v/v)	0.069	0.750	0.138	0.000	1.637	0.977
7	4 M LiFSI EC/DMC (1:1 v/v)	0.142	1.893	0.216	0.000	1.506	0.969
8	4 M LiFSI FEC/DMC (1:1 v/v)	0.273	2.117	0.213	0.000	1.569	0.973
9	4 M LiFSI EC/FEC/DMC (1:1:1 v/v/v)	0.222	2.001	0.221	0.000	1.508	0.969
10	4 M LiFSI DME/TEGDME (1:1 v/v)	0.203	1.267	0.107	0.000	1.882	0.987
11	4 M LiFSI TEGDME/DOL (1:1 v/v)	0.181	1.306	0.127	0.000	1.797	0.984
12	4 M LiFSI DME/TEGDME/DOL (1:1:1 v/v/v)	0.194	1.343	0.119	0.000	1.843	0.986
13	1 M LiFSI EC/DMC/TTE (1.25:1.25:7.5 v/v/v)	2.187	1.827	0.111	0.000	2.073	0.992
14	1 M LiFSI FEC/DMC/TTE (1.25:1.25:7.5 v/v/v)	2.336	1.888	0.108	0.000	2.103	0.992
15	1 M LiFSI EC/FEC/DMC/TTE (0.83:0.83:0.83:7.5 v/v/v/v)	2.239	1.858	0.112	0.000	2.077	0.992
16	1 M LiFSI DME/TEGDME/TTE (1.25:1.25:7.5 v/v/v)	2.808	1.629	0.075	0.000	2.228	0.994
17	1 M LiFSI TEGDME/DOL/TTE (1.25:1.25:7.5 v/v/v)	2.603	1.636	0.083	0.000	2.183	0.993
18	1 M LiFSI DME/TEGDME/DOL/TTE (0.83:0.83:0.83:7.5 v/v/v/v)	2.730	1.656	0.079	0.000	2.211	0.994
19	1 M LiPF6 EC/DMC (1:1 v/v)	0.149	1.174	0.261	0.000	1.170	0.932
20	1 M LiPF6 FEC/DMC (1:1 v/v)	0.335	1.361	0.260	0.000	1.222	0.940
21	1 M LiPF6 EC/FEC/DMC (1:1:1 v/v/v)	0.261	1.285	0.268	0.000	1.165	0.932
22	1 M LiPF6 DME/TEGDME (1:1 v/v)	0.256	0.649	0.125	0.000	1.687	0.979
23	1 M LiPF6 TEGDME/DOL (1:1 v/v)	0.213	0.715	0.149	0.000	1.591	0.974
24	1 M LiPF6 DME/TEGDME/DOL (1:1:1 v/v/v)	0.239	0.706	0.140	0.000	1.631	0.977
25	1 M LiTFSI EC/DMC (1:1 v/v)	0.135	1.260	0.248	0.012	1.077	0.916
26	1 M LiTFSI FEC/DMC (1:1 v/v)	0.303	1.442	0.246	0.013	1.123	0.925
27	1 M LiTFSI EC/FEC/DMC (1:1:1 v/v/v)	0.237	1.366	0.255	0.013	1.070	0.915
28	1 M LiTFSI DME/TEGDME (1:1 v/v)	0.376	1.197	0.187	0.012	1.353	0.956
29	1 M LiTFSI TEGDME/DOL (1:1 v/v)	0.187	0.805	0.143	0.010	1.497	0.968
30	1 M LiTFSI DME/TEGDME/DOL (1:1:1 v/v/v)	0.206	0.802	0.134	0.011	1.531	0.971
31	1 M LiFSI EC/DMC (1:1 v/v) 0.02M LiNO3	0.045	1.225	0.258	0.000	1.190	0.935
32	1 M LiFSI EC/DMC (1:1 v/v) 2% VC	0.185	1.360	0.282	0.000	1.114	0.923
33	2 M LiFSI DME/TEGDME (1:1 v/v)	0.127	0.884	0.118	0.000	1.758	0.983
34	3 M LiFSI DME/TEGDME (1:1 v/v)	0.169	1.075	0.112	0.000	1.821	0.985

Table S3. Coulombic efficiency measurement of 13 electrolytes for model verification.

Number	Electrolyte	CE	LCE	Predicted LCE
19	1 M LiPF6 EC-DMC	91.70	1.081	1.170
		92.45	1.122	
		90.22	1.009	
1	1 M LiFSI EC-DMC	91.33	1.062	1.187
		92.00	1.097	
		92.47	1.123	
9	4 M LiFSI EC-DMC-FEC	96.59	1.467	1.508
		96.56	1.464	
		96.64	1.474	
4	1 M LiFSI DME-TEGDME	95.33	1.331	1.691
		95.81	1.377	
		96.39	1.443	
33	2 M LiFSI DME-TEGDME	96.40	1.443	1.758
		96.52	1.458	
		97.50	1.602	
34	3 M LiFSI DME-TEGDME	97.68	1.634	1.821
		97.74	1.646	
		98.04	1.708	
14	1 M LiFSI FEC-DMC-TTE	98.64	1.865	2.103
		98.43	1.804	
		98.51	1.828	
18	1 M LiFSI DME-DOL-TEGDME-TTE	99.37	2.198	2.211
		99.49	2.294	
		99.53	2.330	
31	1 M LiFSI EC-DMC 0.02 M LiNO3	95.58	1.354	1.190
		95.63	1.359	
		94.90	1.292	
10	4 M LiFSI DME-TEGDME	98.39	1.793	1.882
		97.50	1.602	
		98.16	1.736	
25	1 M LiTFSI EC-DMC	93.99	1.221	1.077
		92.49	1.124	
		92.97	1.153	
15	1 M LiFSI EC-FEC-DMC-TTE	99.20	2.099	2.077
		99.17	2.081	
		99.26	2.133	
32	1 M LiFSI EC-DMC 2% VC	92.51	1.125	1.114
		93.31	1.174	
		91.85	1.089	

Table S4. Measured Coulombic efficiencies of the designed electrolytes.

Electrolyte	CE (%)	LCE	Average CE
1 M LiFSI MBE-toluene (1:1 vol)	99.58	2.375	99.51
	99.45	2.258	
	99.53	2.327	
	99.47	2.272	
1 M LiFSI MTBE-TTE (1:2 vol)	99.60	2.402	99.57
	99.47	2.277	
	99.56	2.360	
	99.64	2.448	
1 M LiFSI MTBE-toluene (1:1 vol)	99.73	2.567	99.64
	99.53	2.332	
	99.73	2.569	
	99.56	2.355	
1 M LiFSI DBE-toluene (1:1 vol)	99.68	2.498	99.70
	99.72	2.554	
	99.77	2.641	
	99.64	2.444	