## Supporting Information for

## Separator-Free and Concentrated LiNO<sub>3</sub> Electrolyte Cells Enable Uniform Lithium Electrodeposition

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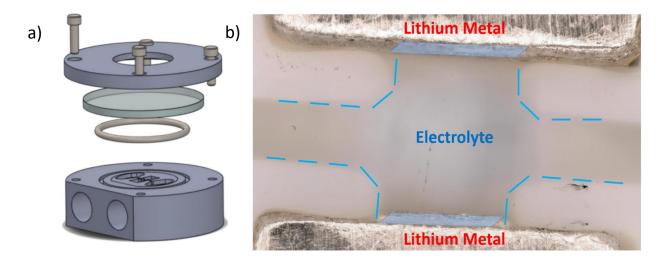
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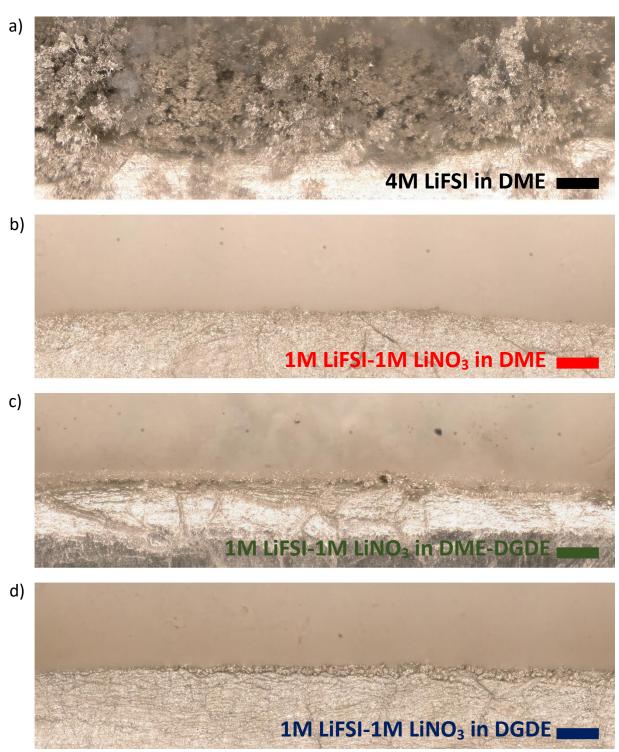
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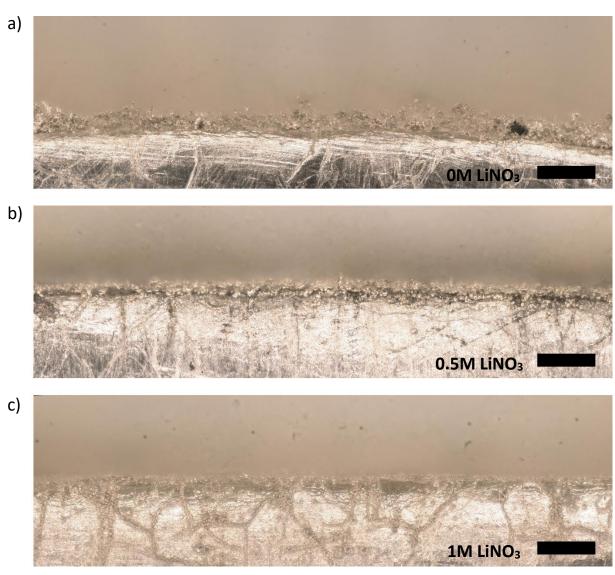
## SUPPORTING INFORMATION



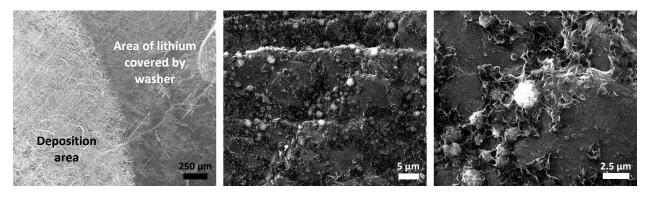
**Figure S1.** 3D CAD schematic of the optical cell utilized in this work (a). b) illustrates an assembled Li|Li visualization cell. The light blue outlines illustrate the walls of the optical cell that hold the lithium in place. The highlighted areas on the lithium electrodes are the deposition/dissolution sites. Only electrolyte, without a separator, spans the distance between the electrodes.



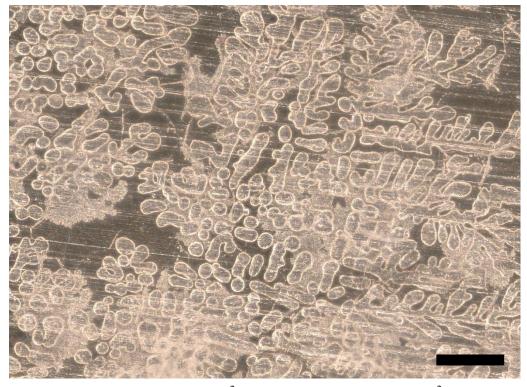
**Figure S2.** Lithium electrodes after passing 6 mAh cm<sup>-2</sup> of charge with a current density of 1 mA cm<sup>-2</sup>. Electrolytes utilized: (a) 4M LiFSI in DME, (b) 1M LiFSI with 1M LiNO<sub>3</sub> in DME, (c) 1M LiFSI with 1M LiNO<sub>3</sub> in DME and DGDE, and (d) 1M LiFSI with 1M LiNO<sub>3</sub> in DGDE. The scale bars are 200  $\mu$ m.



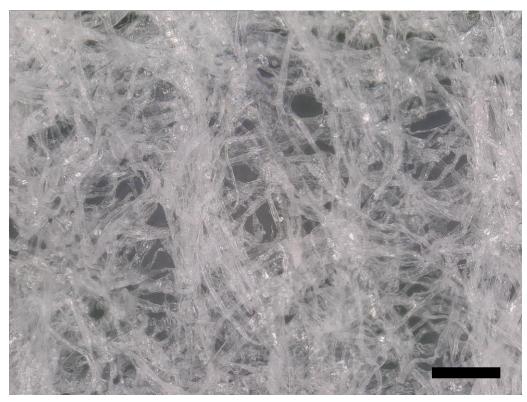
**Figure S3.** Lithium electrodes after passing 6 mAh cm<sup>-2</sup> of charge with a current density of 0.5 mA cm<sup>-2</sup> with 1M LiFSI co-salt and varying LiNO<sub>3</sub> concentrations: (a) no LiNO<sub>3</sub> salt, (b) 0.5M LiNO<sub>3</sub>, and (c) 1M LiNO<sub>3</sub>. Scale bars are 200  $\mu$ m.



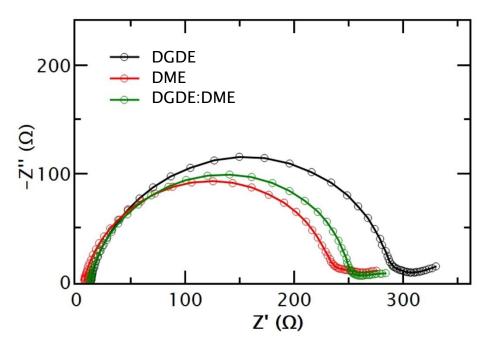
**Figure S4.** SEM images of a 1 mAh cm<sup>-2</sup> Li-deposit cycled at 1 mA cm<sup>-2</sup> using a 1M LiNO<sub>3</sub> with 1 M LiFSI in DGDE electrolyte. A washer was used in this electrodeposition.



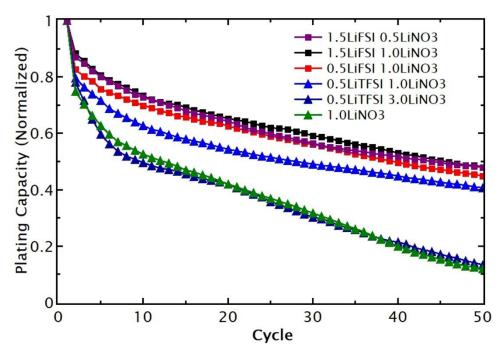
**Figure S5.** Optical image of a 1mAh cm<sup>-2</sup> Li-deposit cycled at 1 mA cm<sup>-2</sup> using a 1M LiNO<sub>3</sub> with 1 M LiFSI in DGDE electrolyte. A washer and polymer separator were used in this electrodeposition; the washer held the separator in place but did not press down on it where the deposition occurred. Scale bar is 100 μm.



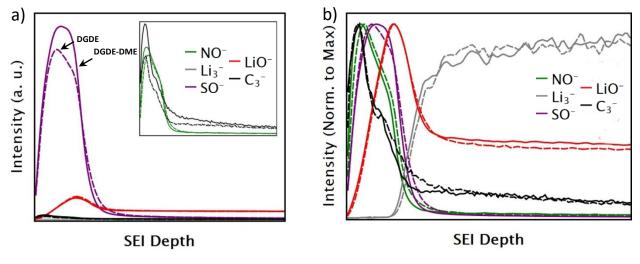
**Figure S6.** Optical image of Kimwipe. Scale bar is 200 μm.



**Figure S7.** EIS spectra of 1M LiNO<sub>3</sub> with 1M LiFSI electrolytes in DGDE, DME, and DGDE:DME (1:1 vol%) symmetrical Li|Li cells before cycling.



**Figure S8.** Retention of the coulombic capacity of LFP|Cu cells for different electrolytic solutions normalized to the first cycle. All electrolytes utilized the DGDE solvent.



**Figure S9.** Time-of-flight depth profiling of the DGDE:DME (solid) and DME (solid) based electrolytes. a) compares the SEI of lithium electrodes soaked in electrolyte for the DME and DME-DGDE electrolytes and b) shows the same SEI normalized to the maxima of each individual species.