

Supporting Information for: Complete Characterization of a Lithium Battery Electrolyte using a Combination of Electrophoretic NMR and Electrochemical Methods

Darby Hickson^{1,2}, David Halat^{1,2}, Alec Ho^{1,2}, Jeffrey Reimer^{1,2}, Nitash Balsara^{1,2}

¹*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States*

²*Department of Chemical and Biomolecular Engineering, University of California, Berkeley, Berkeley, California 94720, United States*

1. Salt Concentration Determination for LiTFSI/tetraglyme Electrolytes

Concentrations of the electrolytes used in this work are given in Table S1, showing the conversion between r , corresponding to the ratio of lithium ions to ether oxygens in tetraglyme ($r = ([\text{Li}^+]/[\text{O}])$) and molality, m , or moles of LiTFSI salt per kilogram of tetraglyme.

Table S1. Salt concentration equivalencies for LiTFSI/tetraglyme electrolytes

r ($[\text{Li}^+]/[\text{O}]$)	m (mol/kg)
0.008	0.18
0.016	0.36
0.032	0.72
0.048	1.08
0.064	1.45
0.08	1.79
0.096	2.16
0.112	2.52

2. Diffusion Coefficient Fitting

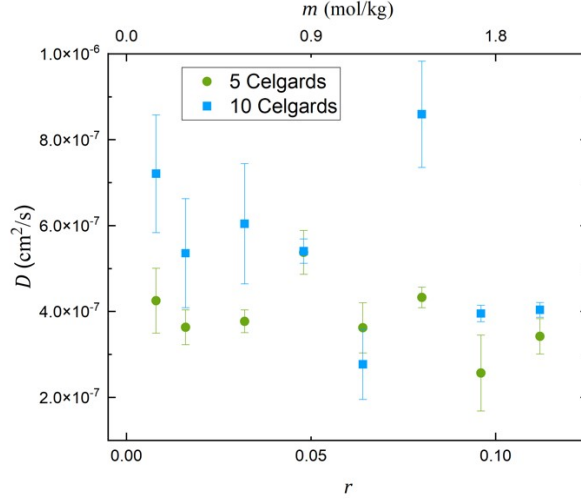


Figure S2.1. Comparison of averaged D from coin cells made with five and ten Celgards. For cells made with five Celgards, the first 15 minutes of relaxation were fit to determine D , while the first hour of relaxation was fit to determine D for the ten Celgard cells. Values are more-or-less consistent with each other.

3. Error determination

This section of the SI details equations used to determine error for all of the derived parameters in this work. Error formulas are based on standard error propagation. Error for measured parameters is based on standard deviation of multiple measurements.

Formula for determining error in $\frac{dU}{d\ln m}$ based on the finite difference method, where x_1 and y_1 refer to the $\ln(\text{molality})$ and OCV, respectively, for the first data point:

$$\delta\left(\frac{y_3 - y_1}{x_3 - x_1}\right) = \left|\frac{y_3 - y_1}{x_3 - x_1}\right| * \left|\frac{\sqrt{(\delta y_3)^2 + (\delta y_1)^2}}{y_3 - y_1}\right| \quad (\text{S3.1})$$

Error formula for thermodynamic factor based on electrochemical methods, $T_{f, \text{echem}}$:

$$\delta\left(1 + \frac{d\ln \gamma_{\pm}}{d\ln m}\right) = \left|1 + \frac{d\ln \gamma_{\pm}}{d\ln m}\right| \sqrt{\left(\frac{\delta \kappa}{\kappa}\right)^2 + \left(\frac{\delta D}{D}\right)^2 + \left(\frac{\delta \rho_{+}}{\rho_{+}}\right)^2 + 2 * \left(\frac{\delta\left(\frac{dU}{d\ln m}\right)}{\frac{dU}{d\ln m}}\right)^2} \quad (\text{S3.2})$$

Error formula for the transference number based on electrochemical methods, $t_{+, \text{echem}}^0$:

$$\delta t_{+}^0 = |t_{+}^0| \sqrt{\left(\frac{\delta \kappa}{\kappa}\right)^2 + \left(\frac{\delta D}{D}\right)^2 + \left(\frac{\delta \rho_{+}}{\rho_{+}}\right)^2 + \left(\frac{\delta\left(\frac{dU}{d\ln m}\right)}{\frac{dU}{d\ln m}}\right)^2} \quad (\text{S3.3})$$

Error formula for calculation of the thermodynamic factor using eNMR and concentration cells,
 $T_{f,eNMR}$:

$$\delta\left(1 + \frac{d\ln\gamma_{\pm}}{d\ln m}\right) = \left|1 + \frac{d\ln\gamma_{\pm}}{d\ln m}\right| \sqrt{\left(\frac{\delta t_{-}^0}{t_{-}^0}\right)^2 + \left(\frac{\delta\left(\frac{dU}{d\ln m}\right)}{\frac{dU}{d\ln m}}\right)^2} \quad (\text{S3.4})$$