Supporting Information

Polymer-Ceramic Composite Electrolytes for Lithium Batteries: A Comparison Between the Single-Ion-Conducting Polymer Matrix and Its Counterpart

Laura C. Merrill^a, Xi Chelsea Chen^{b,*}, Yiman Zhang^b, Hunter O. Ford^a, Kun Lou^c, Yubin Zhang^d,

Guang Yang^b, Yangyang Wang^e, Yan Wang^d, Jennifer L. Schaefer^a, and Nancy J. Dudney^b

^aDepartment of Chemical and Biomolecular Engineering, University of Notre Dame, Notre Dame, IN 46556, USA

^bChemical Sciences Division, ^eCenter for Nanophase Materials Sciences Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA

^cThe Bredesen Center for Interdisciplinary Research and Graduate Education, University of Tennessee, Knoxville, TN 37996, USA

^dDepartment of Mechanical Engineering, Worcester Polytechnic Institute, Worcester, MA 01609, USA

*Corresponding author email: <u>chenx@ornl.gov</u>



Figure S1: Conductivity comparison between LiTFSI containing films dry (solid), swelled in bulk TEGDME (half shaded circle), and swelled in 20 µL TEGDME.



Figure S2: (a) Storage modulus and (b) loss modulus versus temperature for the PEGDMA-co-STFSI polymer films with and without the ceramic particles The increase in storage modulus beginning around 70 °C is due to the loss of TEGDME, and the graph is cut off at 75 °C for this reason.



Figure S3: EDS mapping of SEM image at 5000x magnification from Figure 1e, all scale bars represent 5µm. (a) carbon, (b) oxygen, (c) sulfur, (d) silicon, (e) phosphorus, (f) titanium, (g) aluminum, and (h) is the SEM image.



Figure S4: Raw DSC data from the second temperature scan from -80 to 150 °C. Dashed lines represent the composite polymer electrolytes and the solid lines are the pristine polymer electrolytes. The peak in the LiTFSI sample is likely from unreacted monomer, as this sample was not washed with any solvent to prevent the loss of LiTFSI.



Figure S5: EIS throughout cycling of (a) PEGDMA-co-STFSI and (b) PEGDMA-co-STFSI with LICGCTM.



Figure S6: Transference number measurements for PEGDMA with LiTFSI (a) EIS measurements before and after polarization and (b) polarization for 10 hours at 10 mV. Transference number measurements for PEGDMA with LiTFSI and LICGCTM (c) EIS measurements before and after polarization and (d) polarization for 10 hours at 10 mV.



Figure S7: Transference number measurements for PEGDMA-co-STFSI (a) EIS measurements before and after polarization and (b) polarization for 10 hours at 10 mV. Transference number measurements for PEGDMA-co-STFSI with LICGCTM (c) EIS measurements before and after polarization and (d) polarization for 10 hours at 10 mV.



Figure S8: Lithium ion conductivity as a function of 1000/T. Lithium ion conductivity of the TEGDME swelled xPEGDMA with LiTFSI polymers, calculated as follows: $\sigma_{Li} = \sigma * t^+$.



Figure S9: Full Raman spectra from Figure 6.