

Exceptional long-life Performance of lithium-ion batteries using ionic liquid-based electrolytes

Giuseppe Antonio Elia^{1†}, Ulderico Ulissi^{2,3†}, Sangsik Jeong^{2,3}, Stefano Passerini^{2,3,*}
and Jusef Hassoun^{4*}

¹ *Technische Universität Berlin, Research Center of Microperipheric Technologies, Gustav-Meyer-Allee 25, 13355 Berlin, Germany*

² *Helmholtz Institute Ulm (HIU), Helmholtzstrasse 11, 89081 Ulm, Germany*

³ *Karlsruhe Institute of Technology (KIT), P.O. Box 3640, 76021 Karlsruhe, Germany*

⁴ *Department of Chemical and Pharmaceutical Sciences, University of Ferrara, Via Fossato di Mortara, 44121, Ferrara, Italy*

† *These authors equally contributed*

Corresponding Authors: jusef.hassoun@unife.it; Stefano.passerini@kit.edu

Supplementary Information

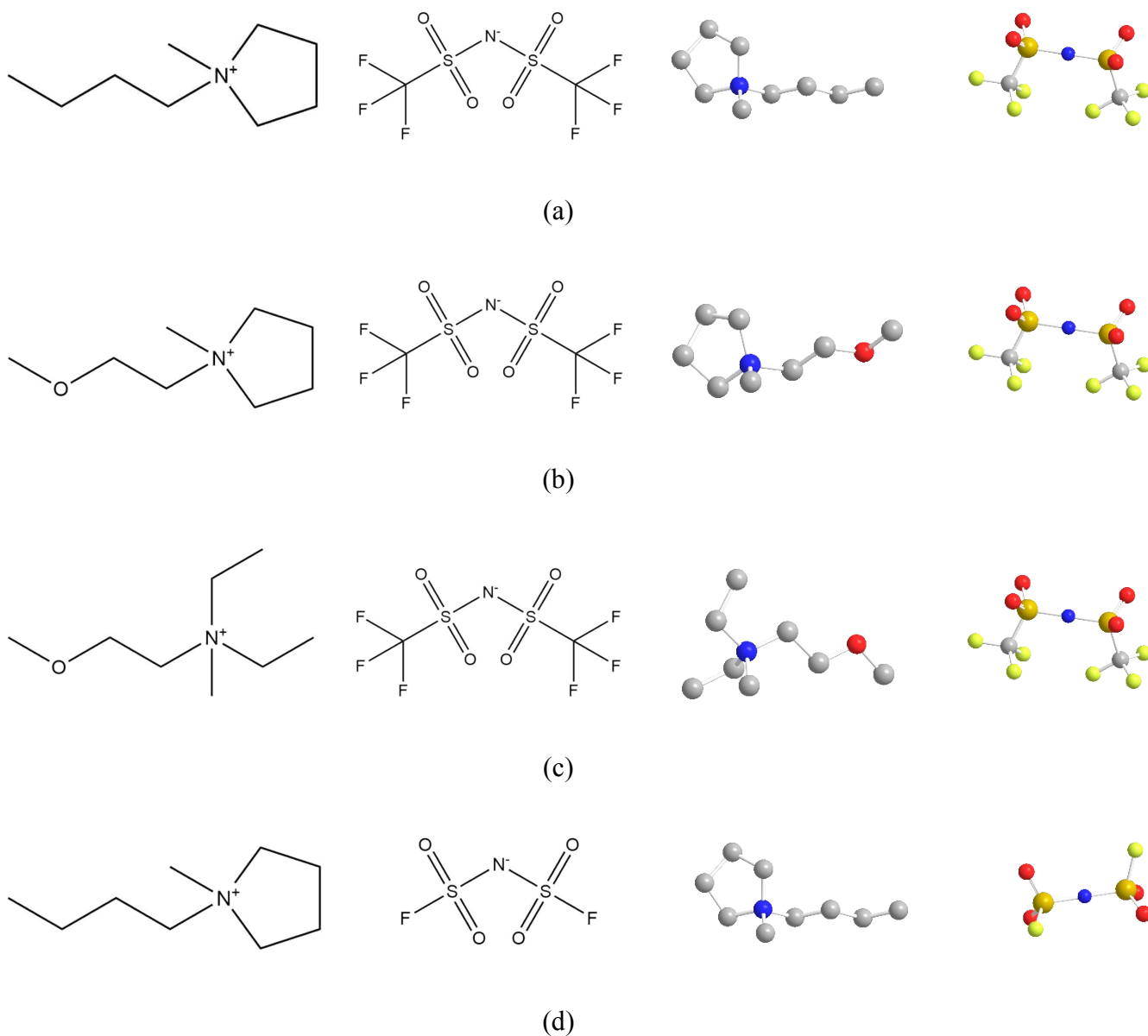
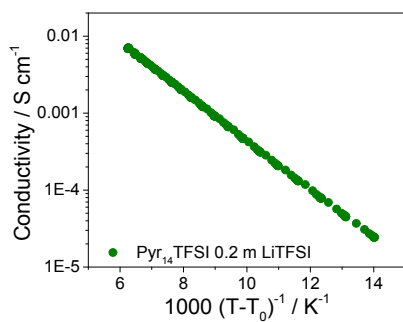
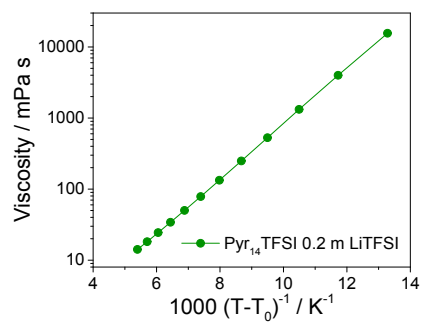


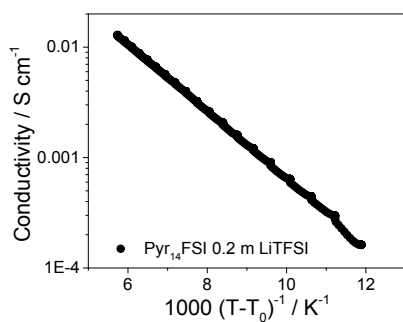
Figure S1 Structural details of the studied ionic liquids. a) *N*-butyl-*N*-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide (Pyr₁₄TFSI), b) *N*-methoxy-ethyl-*N*-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide (Pyr_{12O1}TFSI), c) *N*-*N*-diethyl-*N*-methyl-*N*-(2-methoxyethyl)ammonium bis(trifluoromethanesulfonyl)imide (DEMETFSI), d) *N*-butyl-*N*-methylpyrrolidinium bis(fluorosulfonyl)imide (Pyr₁₄FSI)



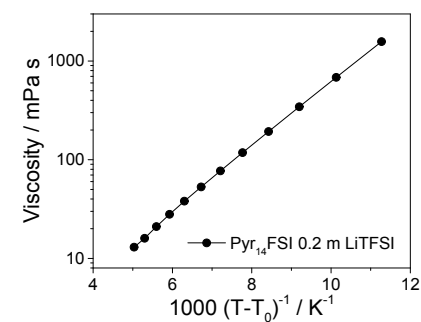
(a)



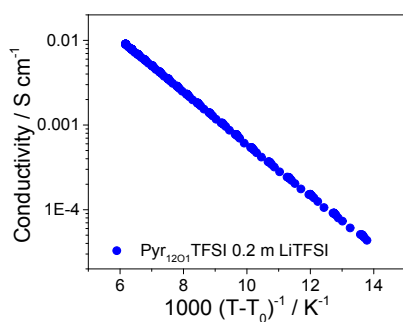
(b)



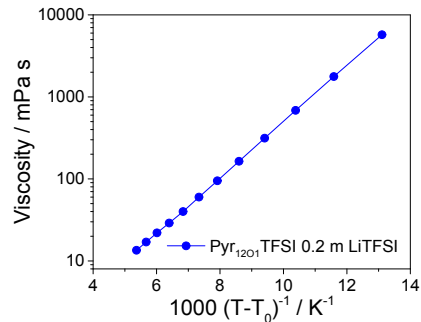
(c)



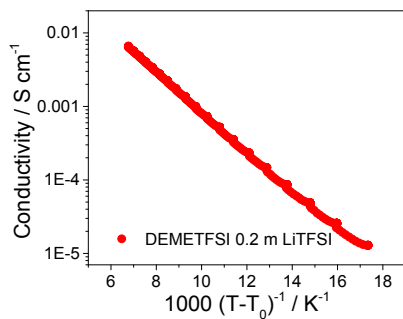
(d)



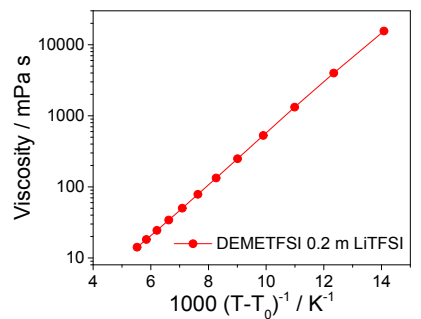
(e)



(f)

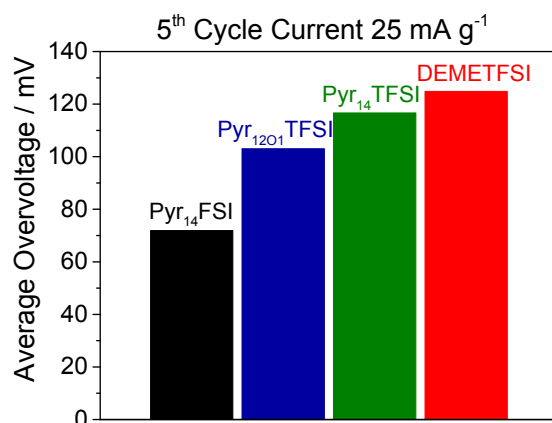


(g)

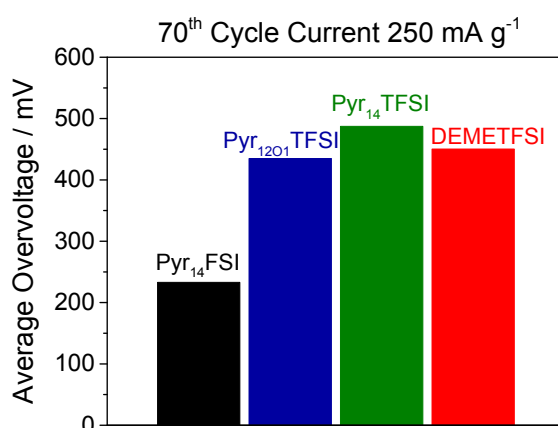


(h)

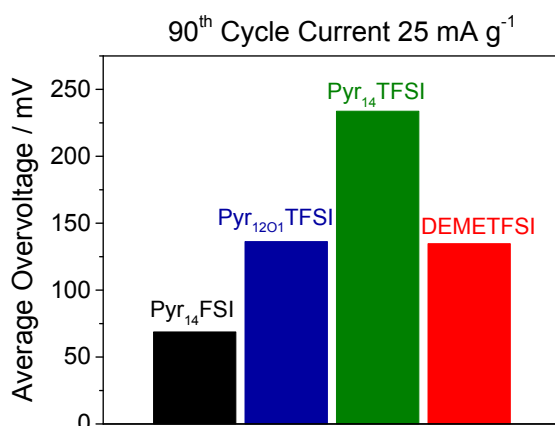
Figure S2 Conductivity and viscosity Vogel-Tammann-Fulcher (VTF) plots of Pyr₁₄TFSI-LiTFSI(a,b), Pyr₁₄FSI-LiTFSI (c,d), Pyr₁₂₀₁TFSI-LiTFSI (e,f) and DEMETFSI-LiTFSI (g,h).



(a)

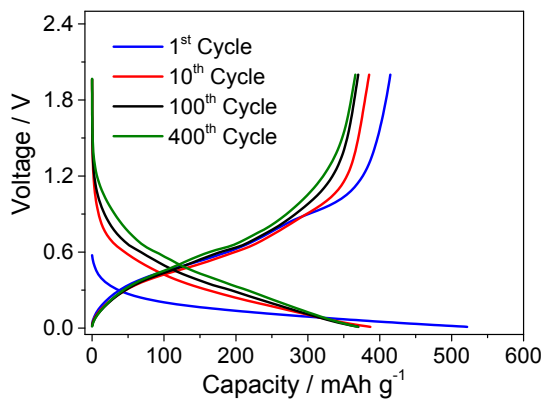


(b)

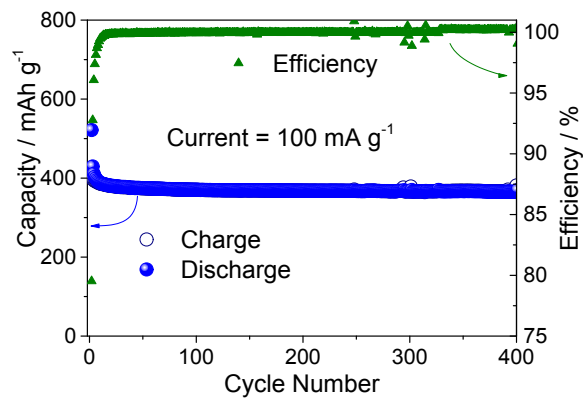


(c)

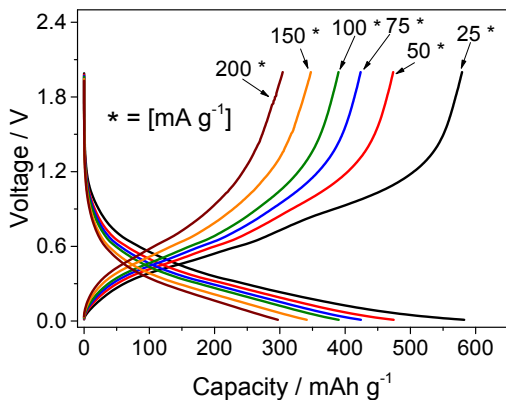
Figure S3 Average polarization during (a) the 5th cycle (measured at 25 mA g⁻¹; 0.12 mA cm⁻²), (b) 70th cycle (measured at 250 mA g⁻¹; 1.2 mA cm⁻²) and (c) 90th cycle (measured at 25 mA g⁻¹; 0.12 mA cm⁻²) of the Li/IL/LFP cells employing the Pyr₁₄FSI-LiTFSI (black), the Pyr₁₂₀₁TFSI-LiTFSI (blue), the Pyr₁₄TFSI-LiTFSI (green) and the DEMETFSI-LiTFSI (red) electrolytes. Temperature 40 °C temperature. Voltage cut off 2.2-4 V.



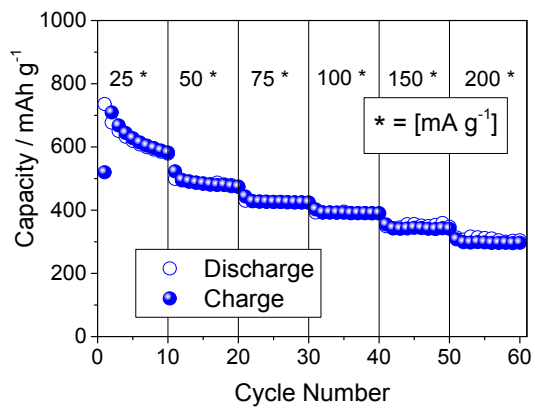
(a)



(b)



(c)



(d)

Figure S4 Voltage signatures (a) and cycling behavior (b) of the Li/Pyr₁₄FSI-LiTFSI/Sn-C cell upon galvanostatic cycle tests at 100 mA g⁻¹ (0.25 mA cm⁻²). Voltage signatures (c) and cycling behavior (d), of the Li/Pyr₁₄FSI-LiTFSI/Sn-C cell upon cycle tests at increasing currents, i.e., 25, 50, 75, 100, 150 and 200 mA g⁻¹ (0.065, 0.1875, 0.25, 0.375, 0.5 mA cm⁻², respectively). Temperature 40°C. Cut off voltages 0.01-2 V.

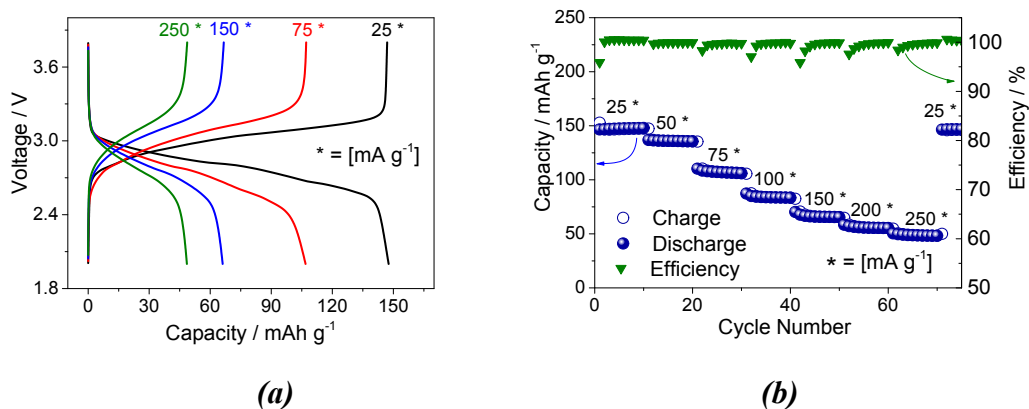


Figure S5 (a) Selected voltage profiles and **(b)** cycling behavior (including coulombic efficiency) of the Sn-C/Pyri₄FSI-LiTFSI/LFP full-cell galvanostatically cycled at increasing currents, i.e., 25, 50, 75, 100, 150, 200 and 250 mA g⁻¹ (0.12, 0.24, 0.36, 0.48, 0.72, 0.96, 1.2 mA cm⁻², respectively) at 20 °C. Voltage cut off 2-3.8 V. The specific capacity (mAh g⁻¹) and specific current (mA g⁻¹) values are given with respect to the active mass of the LFP cathode.

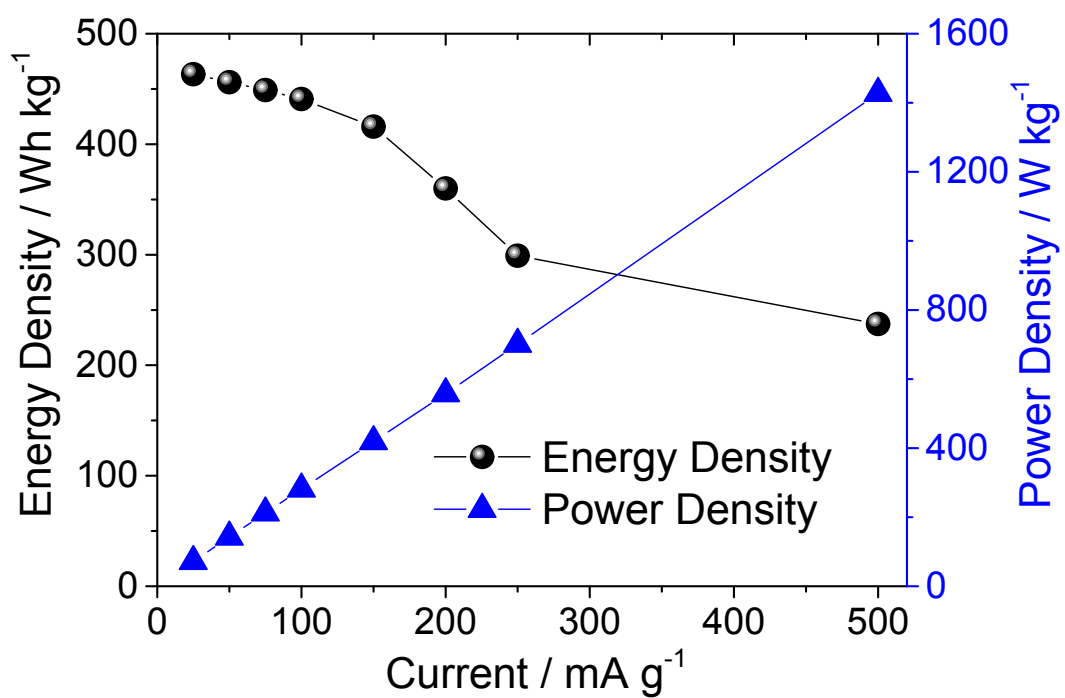


Figure S6 Energy and power densities of the Sn-C/Py₁₄FSI-LiTFSI/LFP lithium ion cell calculated basing on the data reported in Fig. 4 of the work. The values are given with respect to the active mass of the LFP cathode. Temperature 40 °C.

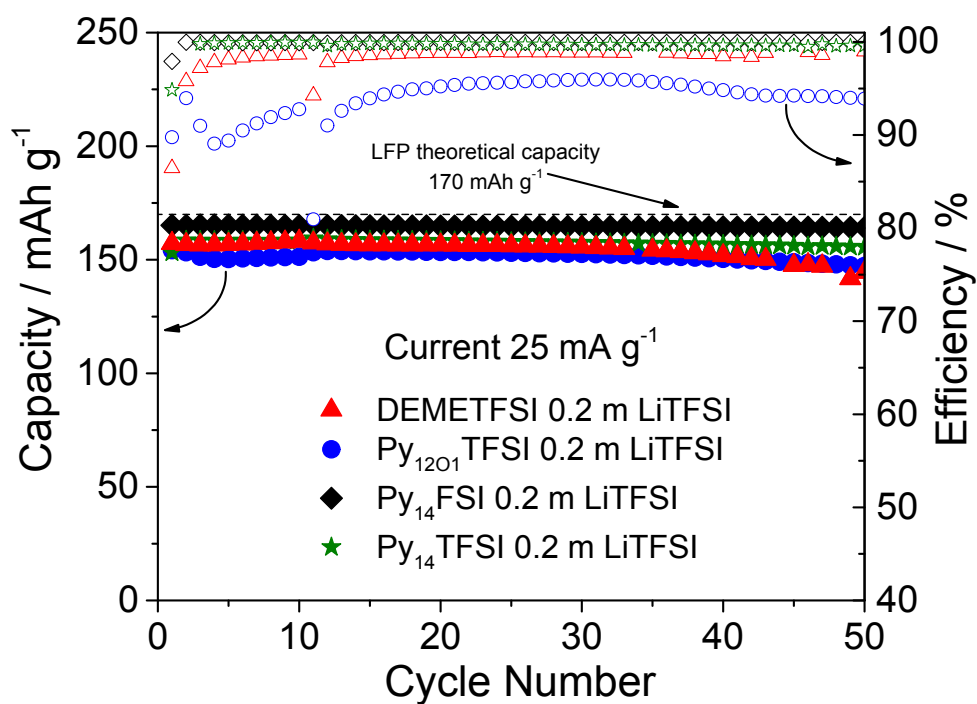
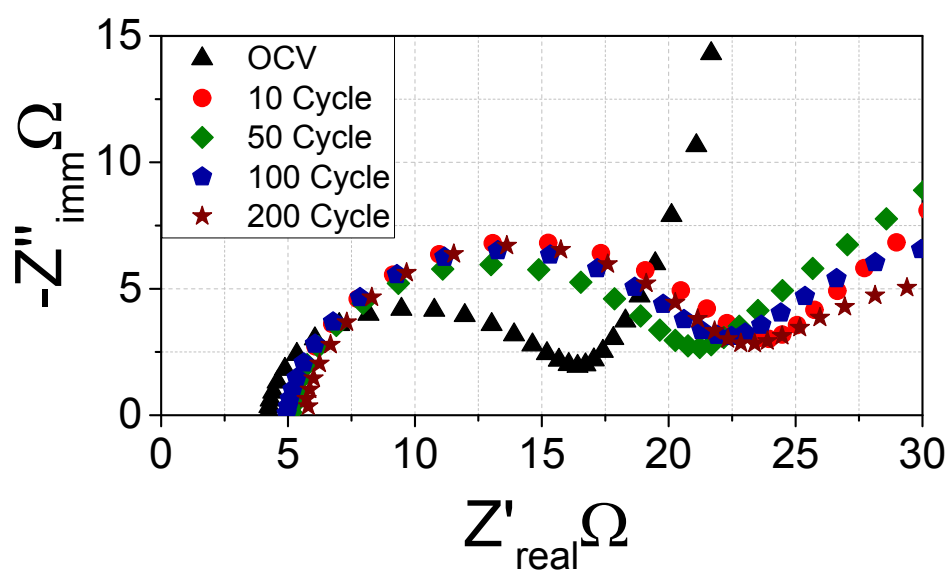
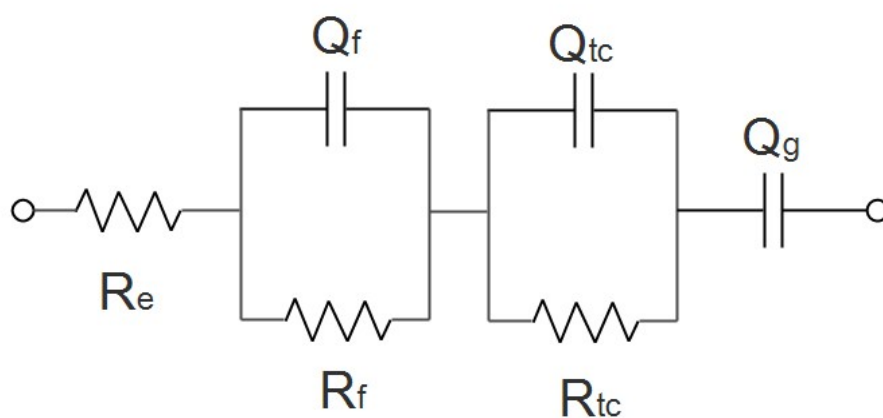


Figure S7 Cycling trend and columbic efficiency of the Sn-C/IL/LFP cells at 25 mA g⁻¹ current (0.12 mA cm⁻²). Voltage cut-off 2.2-4 V. Py₁₄TFSI-LiTFSI (green), Py₁₄FSEI-LiTFSI (black), Py₁₂₀₁TFSI-LiTFSI (blue), DEMETFSEI-LiTFSI (red). All measurements at 40°C. The capacity values are given with respect to the active mass of the LFP cathode.



(a)



(b)

Figure S8 a) Electrochemical impedance spectroscopy (EIS) Nyquist plots during cycling of the Sn-C/Py_{r14}FSI-LiTFSI/LFP cell. Frequency range 1 MHz-10 mHz (10 mV signal amplitude), cycling test at 100 mA g⁻¹ of current (referred to the LFP active mass). Voltage limits 2-3.8 V. Temperature 40 °C. b) R_e(R_fQ_f)(R_{tc}Q_{tc})Q_g equivalent circuit, where R_e represents the resistance of the electrolyte, R_f and Q_f the resistance and the capacitance of the SEI film, R_{tc} and Q_{tc} the resistance and capacitance of the charge transfer process and Q_g the cell capacitance, respectively.