

Mechanisms of Magnesium Ion Transport in Pyrrolidinium Bis(trifluoromethanesulfonyl)imide- Based Ionic Liquid Electrolytes

Sebastian Jeremias,[†] Guinevere A. Giffin,^{‡,§} Arianna Moretti,^{†,‡,§} Sangsik Jeong,^{†,‡,§} Stefano*

*Passerini**^{†,‡,§}

[†] Institute of Physical Chemistry and MEET Battery Research Center, University of Muenster,
Corrensstrasse 28, 48149 Muenster, Germany.

[‡] Helmholtz Institute Ulm (HIU) Electrochemical Energy Storage, Helmholtzstrasse 11, 89081
Ulm, Germany.

[§] Karlsruhe Institute of Technology (KIT), P.O. Box 3640, 76021 Karlsruhe, Germany.

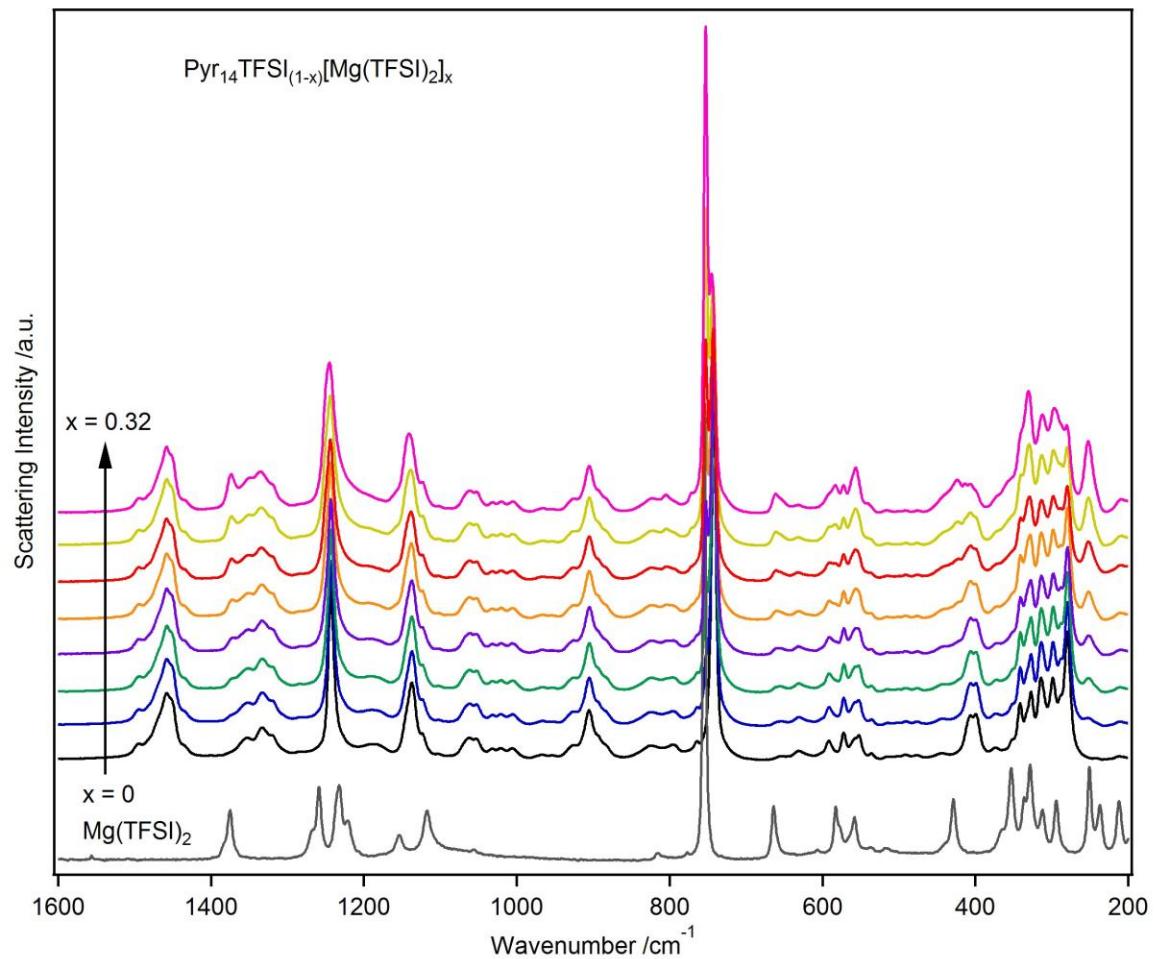


Figure S1. Raman spectra of the Mg²⁺-Pyr₁₄TFSI electrolytes.

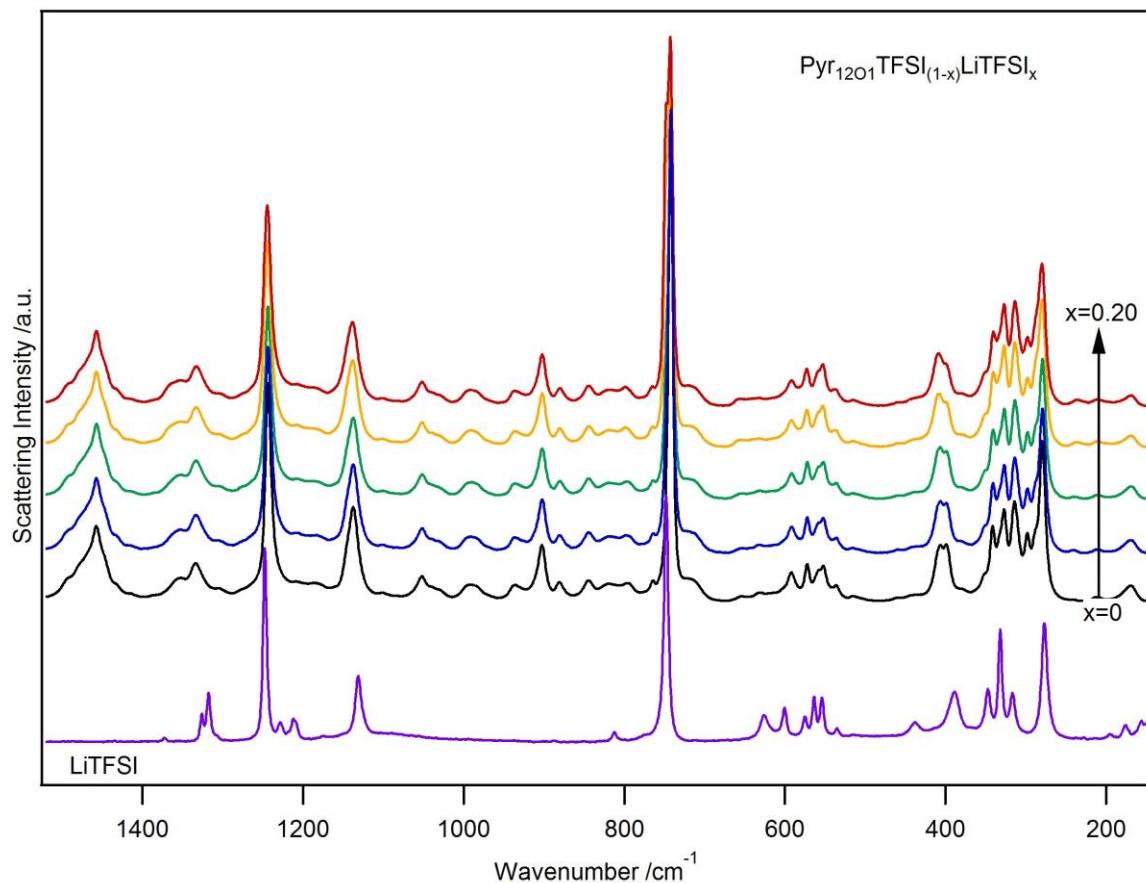


Figure S2. Raman spectra of the Li^+ - $\text{Pyr}_{12\text{O}_1}\text{TFSI}$ electrolytes.

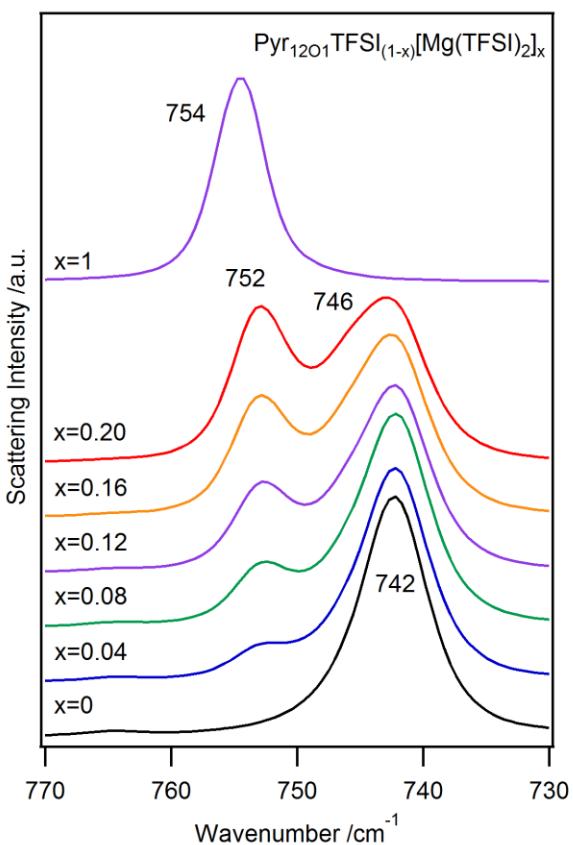


Figure S3. Raman spectra of the Mg^{2+} - $\text{Pyr}_{12}\text{O}_1\text{TFSI}$ electrolytes in the $730\text{-}770\text{ cm}^{-1}$ region (TFSI⁻ expansion-contraction mode).

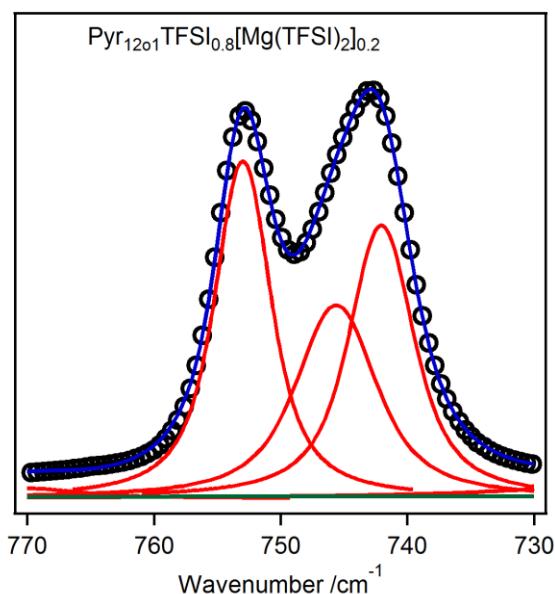


Figure S4. Example of the Voigt fit of the TFSI⁻ expansion-contraction mode.

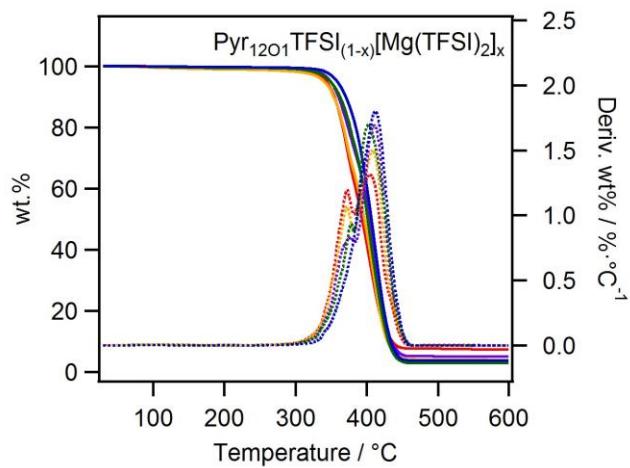


Figure S5. TGA profiles of Mg^{2+} -Pyr₁₂O₁TFSI electrolytes.

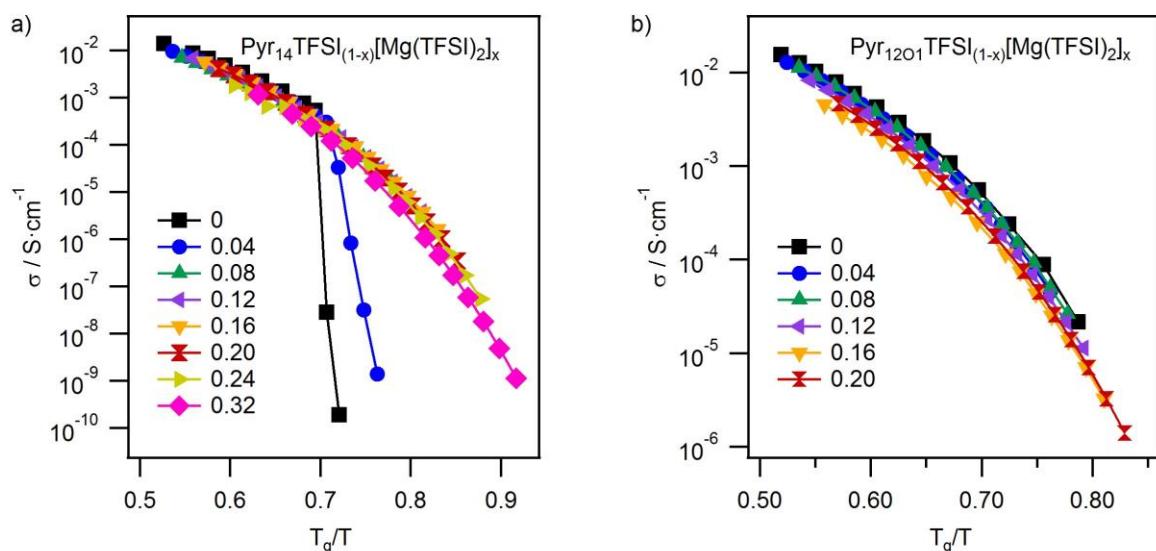


Figure S6. T_g -scaled Arrhenius plot of the conductivity of the Mg^{2+} -IL electrolytes.

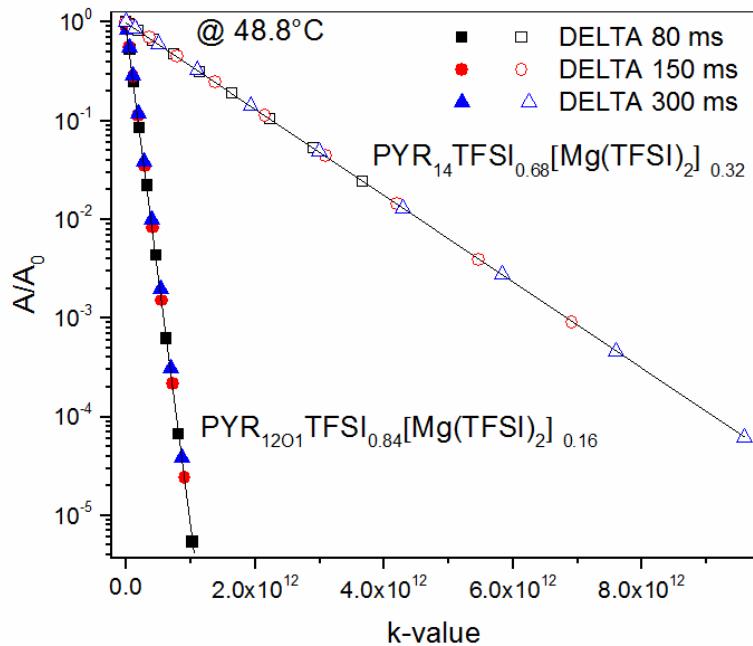


Figure S7. Relative 19F-signal area attenuation for $\text{Pyr}_{14}\text{TFSI}_{0.68}[\text{Mg}(\text{TFSI})_2]_{0.32}$ and $\text{Pyr}_{1201}\text{TFSI}_{0.84}[\text{Mg}(\text{TFSI})_2]_{0.16}$ electrolytes at specific observation times a) at 30°C b) at 48.8 °C. The same monoexponential behavior was seen in both the 19F and 1H decays for all compositions and all temperatures.

Table S1: VTF parameters PYR_{12O₁}TFSI and mixtures. The parameters were determined from

the data as plotted in figure 4 with the equation $\sigma = \sigma_\infty e^{\frac{-B}{T-T_0}}$

Mole fraction	A= $\log \sigma_\infty (\text{S}\cdot\text{cm}^{-1})$	B = $\frac{E_a}{k_B}$	T ₀ (K)
0	-0.31 ± 0.03	630 ± 20	170 ± 1
0.04	-0.35 ± 0.2	637 ± 9	173.9 ± 0.7
0.08	-0.28 ± 0.01	689 ± 6	173.7 ± 0.4
0.12	-0.15 ± 0.04	810 ± 20	170 ± 1
0.16	-0.31 ± 0.05	840 ± 20	173 ± 2
0.20	-0.07 ± 0.03	930 ± 20	173 ± 1

Table S2: Table S1: VTF parameters PYR_{12O₁}TFSI and mixtures. The parameters were

determined from the data as plotted in figure 4 with the equation $\sigma = \sigma_\infty e^{\frac{-B}{T-T_0}}$

Mole fraction	A= $\log \sigma_\infty (\text{S}\cdot\text{cm}^{-1})$	B = $\frac{E_a}{k_B}$	T ₀ (K)
0	-0.22 ± 0.01	692.45 ± 7	169.6 ± 0.6
0.04	-0.40 ± 0.01	661 ± 6	176.1 ± 0.5
0.08	-0.27 ± 0.04	790 ± 20	172 ± 1
0.12	0.02 ± 0.0203	930 ± 10	169.3 ± 0.6
0.16	0.03 ± 0.05	940 ± 30	174 ± 1
0.20	0.23 ± 0.04	1070 ± 20	173 ± 1
0.24	-0.4 ± 0.2	920 ± 70	186 ± 3
0.32	0.9 ± 0.1	1540 ± 70	176 ± 2