Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Li^+ Transference Number and Dynamic Ion Correlations in Glyme-Li Salt Solvate Ionic Liquids Diluted with Molecular Solvents

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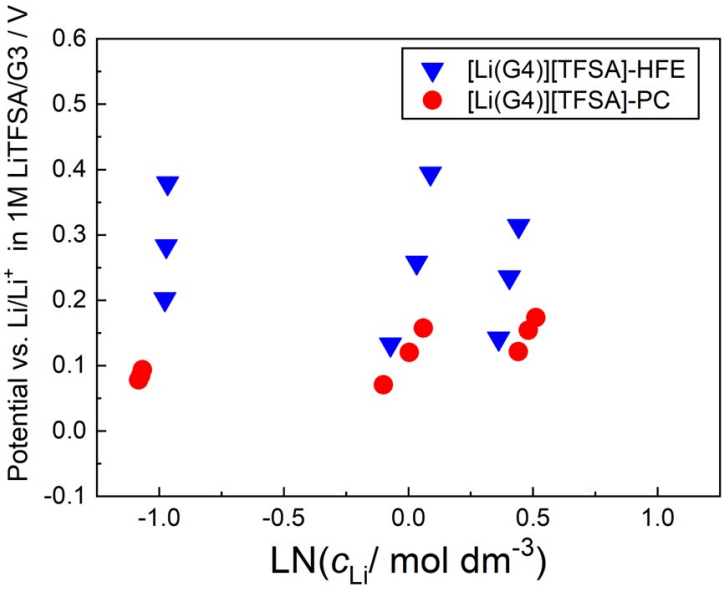

Figure S1. Plots of the Li/Li^+ electrode potential against the natural logarithm of the Li salt concentration in the [Li(G4)][TFSA] mixture with diluents (HFE or PC) at 30 °C. The reference electrode was Li/Li^+ in 1 mol dm^{-3} LiTFSA/G3.

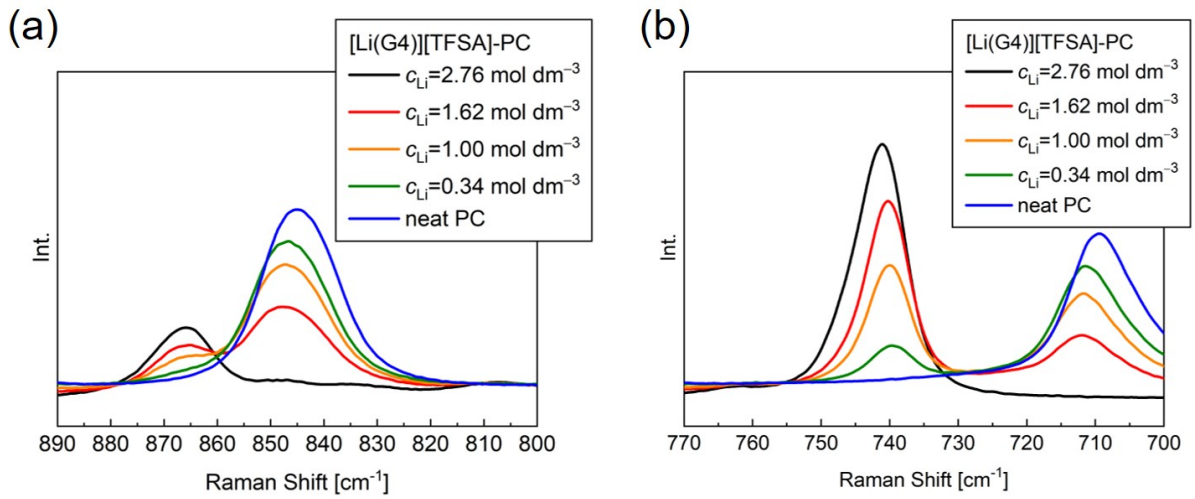

Figure S2. Raman spectra in the range of (a) 800-890 cm^{-1} and (b) 700-770 cm^{-1} for the [Li(G4)][TFSA]-PC.

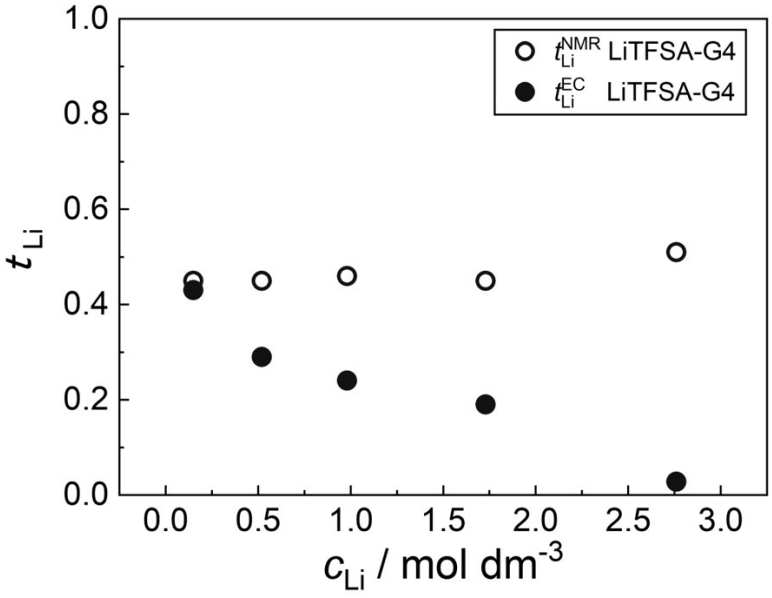

Figure S3. Concentration dependence of the Li transference numbers, t^{EC}_{Li} and t^{NMR}_{Li}, for the LiTFSA-G4 solutions.

Table S1. Salt concentrations and six experimentally obtained parameters for calculating the Onsager transport coefficients.

Sample	c_{Li} [mol dm^{-3}]	σ_{ion} [mS cm^{-1}]	t^{EC}_{Li}	D [10^{-7} cm^2 s^{-1}]			$\frac{d\varphi}{dln(c)}$
				D_{Li}	D_{anion}	D_{salt}	
[Li(G4)][TFSA]+2HFE	1.50	4.0	0.018	8.14	8.3	2.7	2.15
[Li(G4)][TFSA]+4HFE	1.04	5.2	0.018	15.6	16.3	3.1	1.83
[Li(G4)][TFSA]+15HFE	0.38	3.26	0.012	35.9	35.9	12.6	15.9
[Li(G4)][TFSA]+3PC	1.62	5.7	0.071	6.8	8.2	3.6	0.73
1M [Li(G4)][TFSA] / PC	1.00	7.3	0.12	13.0	17.1	9.0	0.57
[Li(G4)][TFSA]+30PC	0.34	4.8 [a]	0.16	21.3 [a]	30.3 [a]	9.2	1.05

[a]Ref 1.

Table S2. Five normalized transport coefficients of all the electrolytes at 30 °C.

Sample	$\sigma_{+}^{self}/\sigma_{ion}$	$\sigma_{-}^{self}/\sigma_{ion}$	$\sigma_{++}^{distinct}/\sigma_{ion}$	$\sigma_{--}^{distinct}/\sigma_{ion}$	σ_{+-}/σ_{ion}
[Li(G4)][TFSA]	0.80 [b]	0.77 [b]	−0.64 [b]	−0.45 [b]	−0.23 [b]
[Li(G4)][TFSA]+2HFE	1.13	1.15	−0.55	−1.09	−0.18
[Li(G4)][TFSA]+4HFE	1.15	1.20	−0.48	−1.17	−0.15
[Li(G4)][TFSA]+15HFE	1.55	1.54	−0.69	−1.54	−0.07
[Li(G4)][TFSA]+3PC	0.72	0.86	−0.042	−0.84	−0.14
1M [Li(G4)][TFSA] / PC	0.65	0.86	0.069	−0.83	−0.12
[Li(G4)][TFSA]+30PC	0.56	0.79	0.38	−0.79	−0.030

[b]Ref 2.

References:

1. K. Ueno, J. Murai, K. Ikeda, S. Tsuzuki, M. Tsuchiya, R. Tatara, T. Mandai, Y. Umebayashi, K. Dokko and M. Watanabe, *J. Phys. Chem. C*, 2016, **120**, 15792-15802.
2. K. Shigenobu , K. Dokko , M. Watanabe and K. Ueno , *Phys. Chem. Chem. Phys.*, 2020, **22** , 15214–15221.