Effectively enhancing ion diffusion in superconcentrated ionic liquid electrolytes by co-solvent additives

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SUPPLEMENTARY INFORMATION

Supplementary Information

Table S- 1: Errors ($E = \frac{abs(Experiment - Model)}{Experiment} 100\%$) for predicted and experimental diffusivities (%) calculated at 80 °C.

	Li	FSI	N1113	Co-solvent
100IL	75	36	62	
20EC	42	9	14	8
20DME	36	15	42	23

Table S- 2: Donnor number and dielectric constant of EC and DME.

	Donnor number	Dielectric constant
EC	16.3[1]	89.8[2]
DME	20.0[1]	7.2[2]

Table S- 3: Number of ions/molecules used in the simulations.

System	Li	FSI	N1113	EC	DME
100IL	320	640	320		
20EC	320	603	283	227	
20DME	320	603	283		222

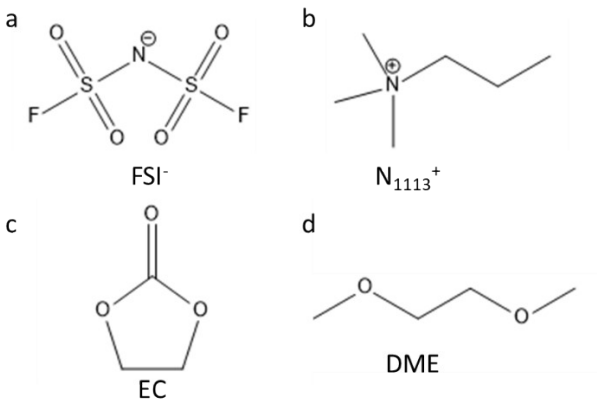

Figure S- 1: (a to d) Chemical structures of FSI, N1113, DME and EC.

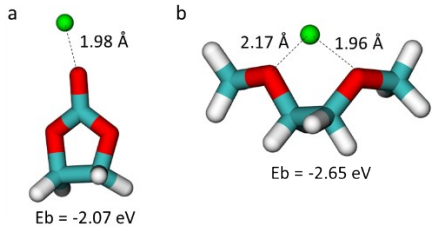

Figure S- 2: (a) Li-EC and (b) Li-DME binding energies calculated using density functional theory at a B3LYP/6-31G(d,p) level of theory with Gaussian 09 software based on the equation Eb= E(Li-solvent)-E(Li)-E(solvent). Literature reports Eb's of -2.24 eV and -2.84 eV for Li-EC and Li-DME, respectively[3].

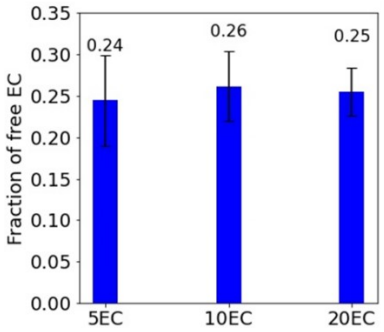

Figure S- 3: (a) Fraction of EC that is not coordinated with Li at 5, 10 and 20 wt% EC concentrations.

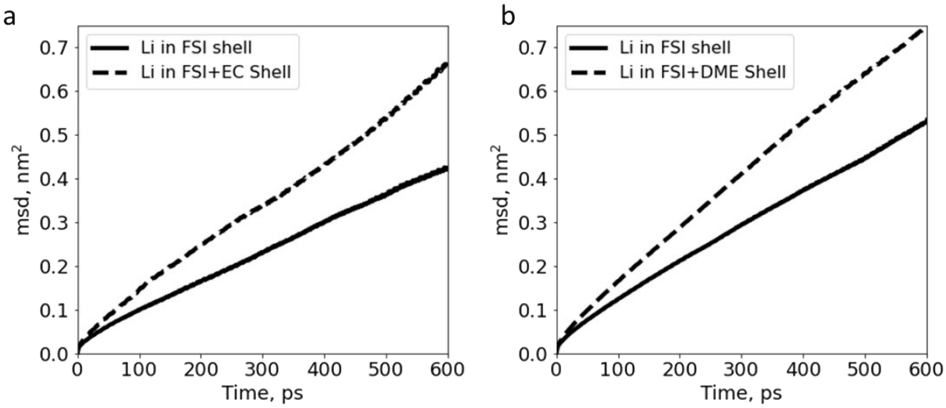

Figure S- 4: (a and b) MSD functions for Li in full FSI shell (solid lines) and hybrid shells (dashed lines). The left and right plots correspond to the 20 wt% EC and the 20 wt% DME systems, respectively.

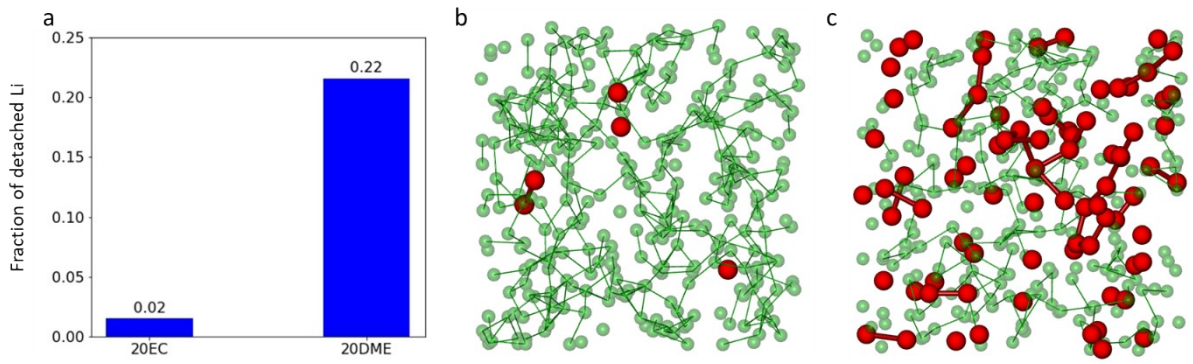

Figure S- 5: (a) Fraction of Li along with their solvation shell that are detached from the Li-FSI network for the 20 wt% EC and the 20 wt% DME systems. (b and c) Simulation snapshots that highlight Li that are detached from the Li-FSI network colored in red for 20EC and 20DME systems, respectively.

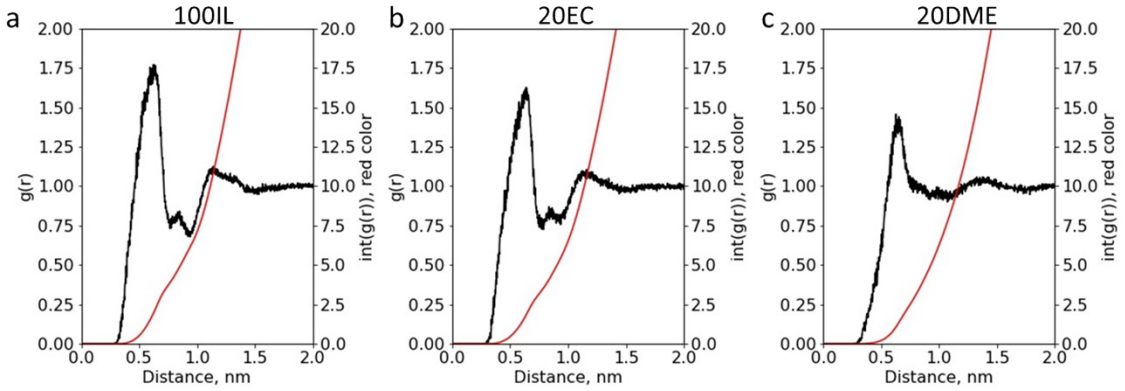

Figure S- 6: (a to c) Li-Li RDF's along with their integration curves, colored in red, for the 100IL, 20EC and 20DME systems, respectively. (d) Calculated average distance between Li and coordination number.

REFERENCES

1. J. Xu, J. Zhang, T. P. Pollard, Q. Li, S. Tan, S. Hou, H. Wan, F. Chen, H. He, E. Hu, K. Xu, X.-Q. Yang, O. Borodin and C. Wang, *Nature*, 2023, **614**, 694-700.
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3. X. Chen, X. Q. Zhang, H. R. Li and Q. Zhang, *Batteries & Supercaps*, 2019, **2**, 128-131.