Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Suppressing Water Clusters by "Hydrotropic" Ionic Liquid for Highly Stable Aqueous Lithium-ion Batteries

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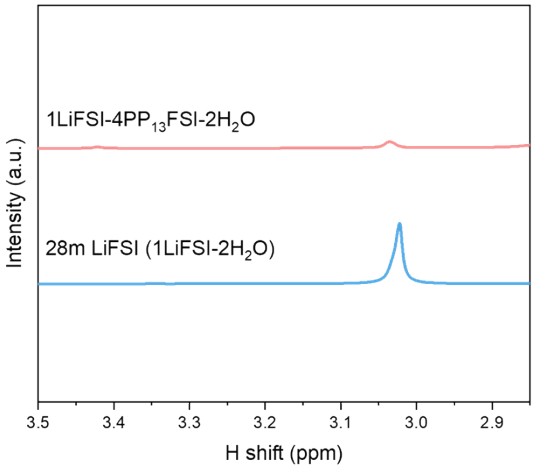

Figure S1. ^{1}H measurement for different aqueous electrolytes with and without $PP_{13}FSI$.

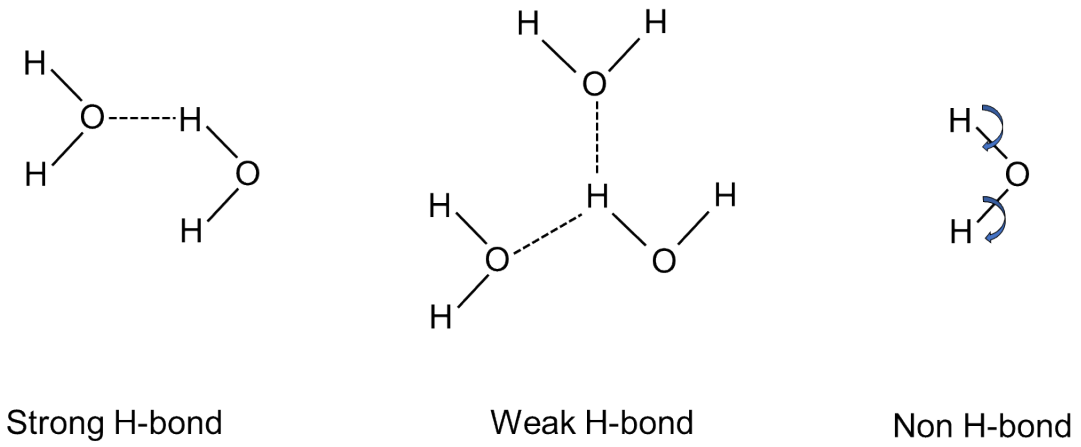

Figure S2. Corresponding vibrational structure of three types of O-H bonds.

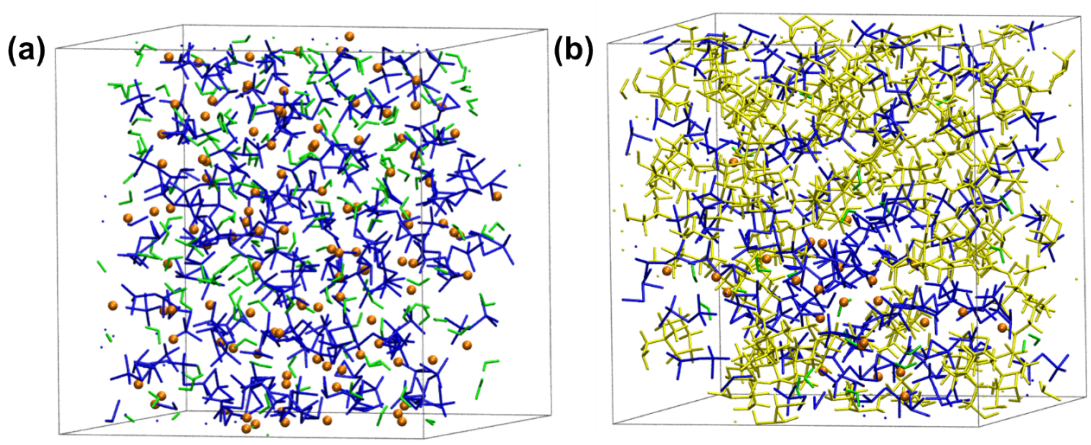

Figure S3. MD simulations results for **a)** 35 m LiFSI and **b)** 1LiFSI-4PP_{13}FSI-1H_2O.

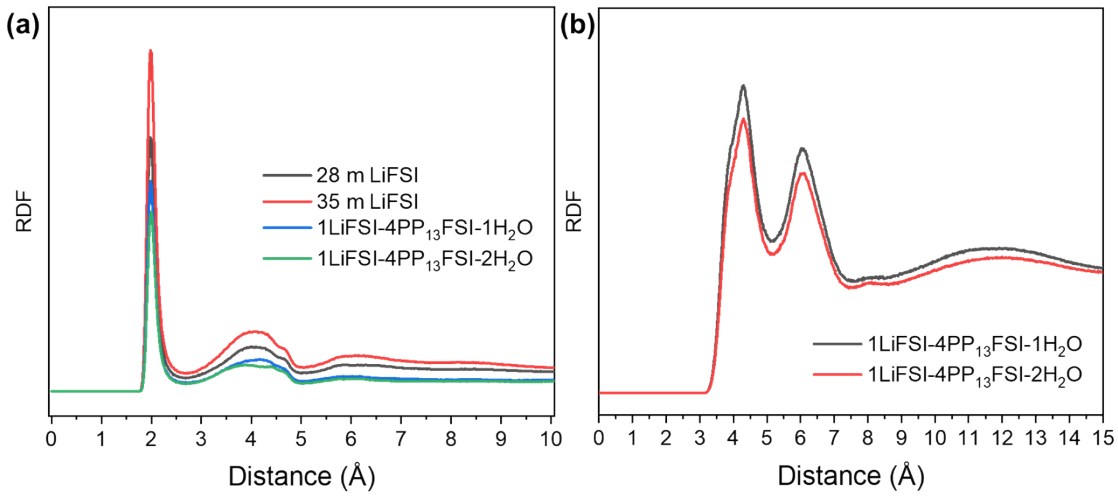

Figure S4. RDF plots for **a)** Li-O (FSI^-) and **b)** Li-N (PP_{13}^+) in different electrolytes.

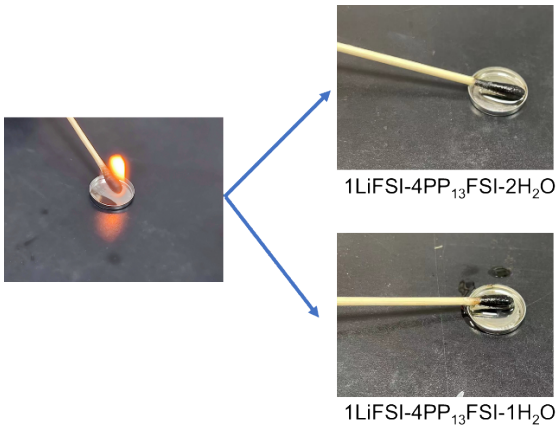

Figure S5. Flammability testing. An ignited cotton swab was immersed in the 1LiFSI-4PP_{13}FSI-2H_2O a) and 1LiFSI-4PP_{13}FSI-1H_2O b) electrolyte. The fire of cotton swab was extinguished in 1LiFSI-4PP_{13}FSI-xH_2O-based electrolytes.

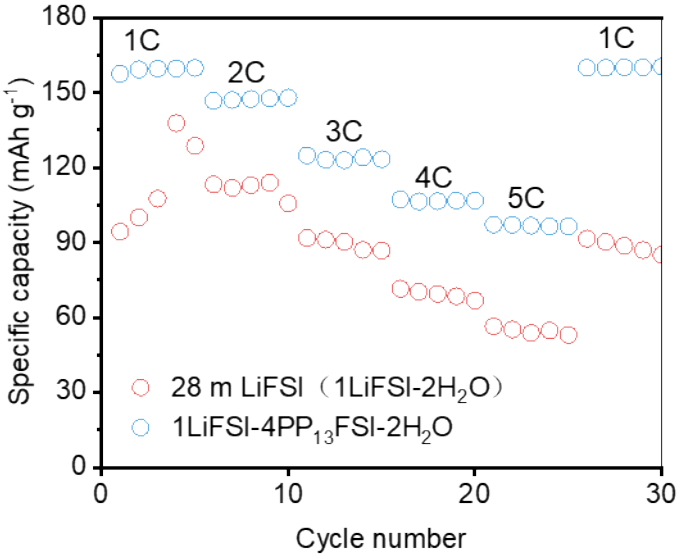

Figure S6. Rate performance of aqueous L-$Li_4Ti_5O_{12}$‖$LiMn_2O_4$ full cells with the 28 m LiFSI electrolyte (red circles) and the 1LiFSI-4PP_{13}FSI-2H_2O electrolyte (blue circles).

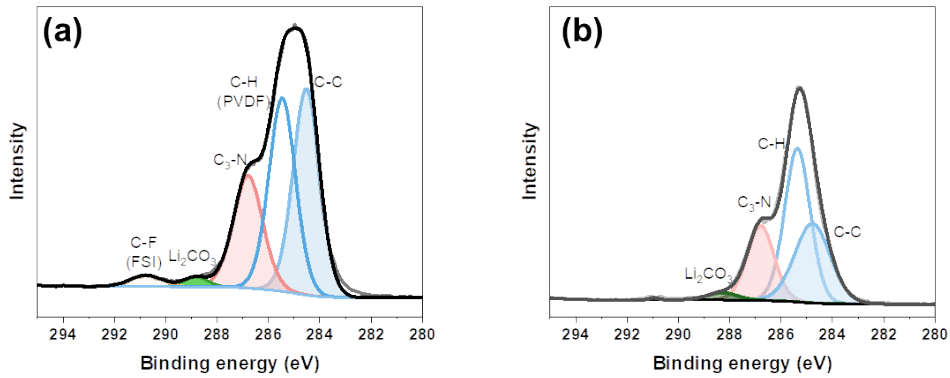

Figure S7. C 1s XPS spectra of cycled $Li_4Ti_5O_{12}$ electrodes in: **a)** 1LiFSI-4PP_{13}FSI-2H_2O electrolyte, **b)** 1LiFSI-4PP_{13}FSI-1H_2O electrolyte. (30^{th} cycle)

Table S1. Ratios of FSI^- anions coordinated with Li^+ in the first coordination shell based on MD simulations for different electrolytes.

Electrolytes	**Ratios of Li-FSI coordination in the first coordination shell of Li^+**
28 m LiFSI	80.11%
35 m LiFSI	82.38%
1LiFSI-4PP_{13}FSI-2H_2O	87.79%
1LiFSI-4PP_{13}FSI-1H_2O	95.99%

Table S2. The number of molecules employed for MD simulations of different aqueous electrolytes.

Electrolytes	Number of molecules		
	LiFSI	H_2O	$PP_{13}FSI$
28 m LiFSI	200	400	0
35 m LiFSI	200	320	0
1LiFSI-4PP_{13}FSI-2H_2O	100	200	400
1LiFSI-4PP_{13}FSI-1H_2O	100	100	400