Supporting Information

In-situ Probing of Mass Exchange at the Solid Electrolyte Interphase in Aqueous and Non-aqueous Zn Electrolytes with EQCM-D

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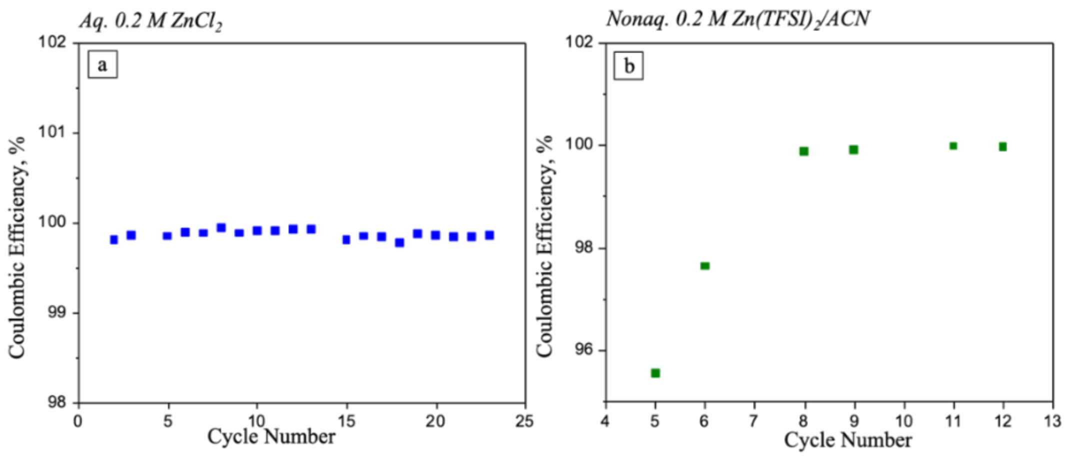

Figure S1. Coulombic efficiency for (a) aqueous 0.2 M $ZnCl_2$ electrolyte cycled between -0.5 V and 0.8 V at 0.05 V/s (cycles 1, 4 and 14 are not shown); (b) nonaqueous 0.2 M $Zn(TFSI)_2$ in ACN electrolyte cycled between 3.2 V to -0.5 V for cycles 4-6, 3.5 V to -0.7 V for cycles 7-9, 3.6 V to -1.0 V for cycles 10-12 at 0.05 V/s (cycles 1-3 and 4, 7, 10 are not shown).

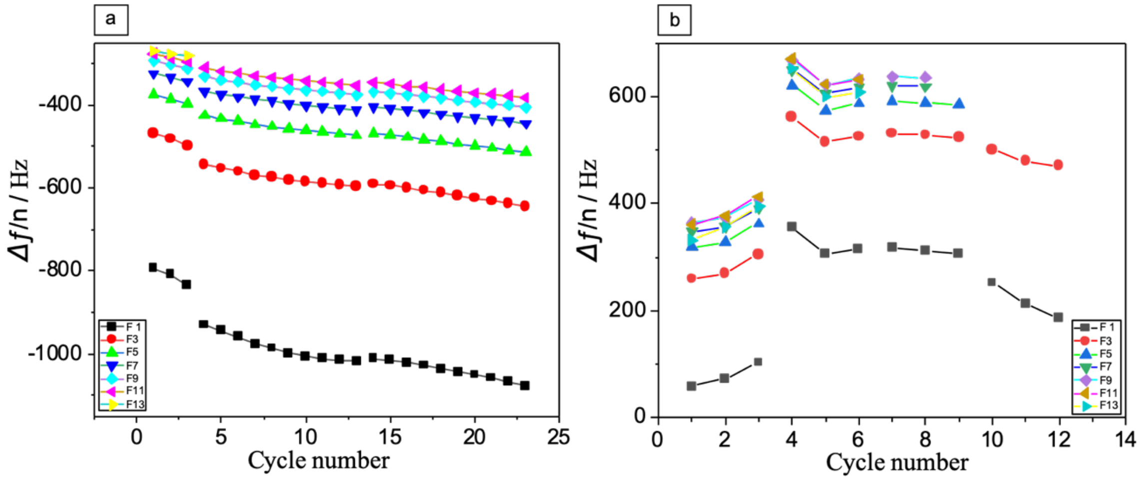

Figure S2. The baseline frequency shift at each CV cycle for aqueous Zn electrolyte (panel a) and non-aqueous Zn electrolyte (panel b) obtained directly from EQCM-D data.

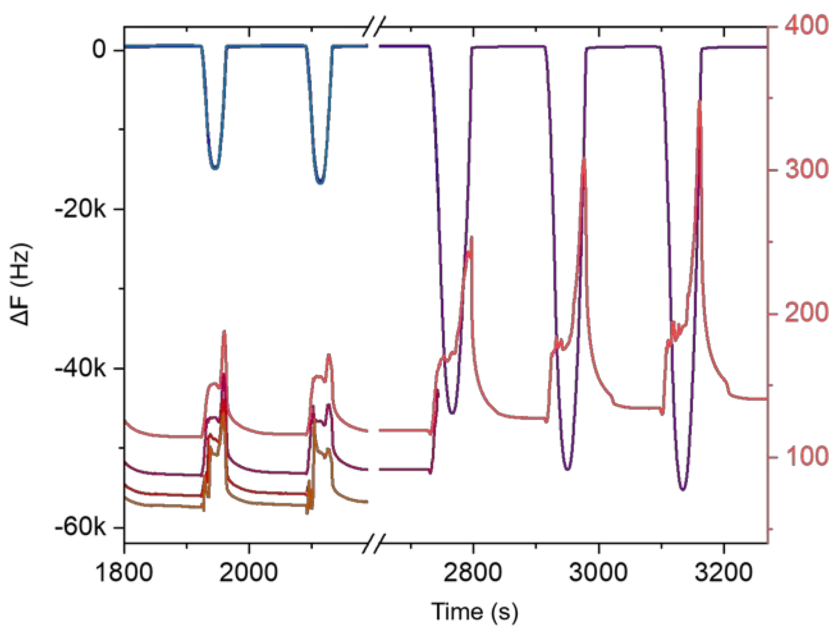

Figure S3. Frequency and dissipation shifts (n=3, 5, 7, 9) for non-aqueous $Zn(TFSI)_2$ electrolyte for CV cycles 8-12.

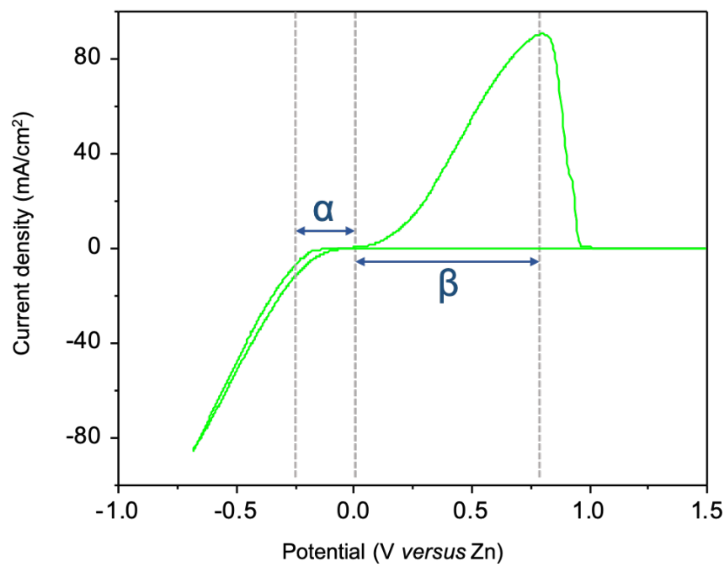

Figure S4. 0.2 M $Zn(TFSI)_2$ in ACN 9th CV cycle α and β voltage parameters used to calculate the overpotential. Plating overpotential (η_α) is defined as the difference between 0 V and voltage point after the plating is initiated while the stripping overpotential (η_β) denotes the difference between 0 V and the highest voltage point during stripping.

Additional Experimental Results:

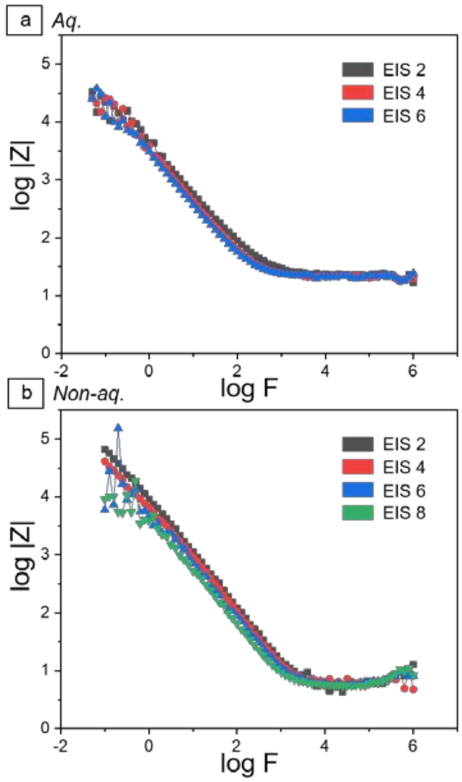

Figure S5. (a) Bode plot representing EIS measured at the 3rd, 13th and 23rd CV cycles for aqueous $Zn(Cl)_2$ electrolyte; (b) Bode plot for EIS measured at the 3rd, 6th, 9th and 12th CV cycles for non-aqueous $Zn(TFSI)_2$ electrolyte.

Table S1. Charge transfer resistance (R_{ct}) for aqueous and non-aqueous Zn electrolytes calculated from the Bode plot (Fig. S1).

	Aq $Zn(Cl)_2$	**Non-aq $Zn(TFSI)_2$**
	R_{ct} (Ohm)	R_{ct} (Ohm)
EIS 2	25339.90	65401.63
EIS 4	26507.73	41325.42
EIS 6	32031.53	16803.97
EIS 8	-	10216.49

Table S2. CE for 0.2 M aqueous $ZnCl_2$ electrolyte, cycles 1, 4, 14 are excluded

CV #	CE, %
2	99.80237
3	99.85029
5	99.84311
6	99.89153
7	99.87622
8	99.93057
9	99.87803
10	99.90485
11	99.89993
12	99.91833
13	99.91915
15	99.79642
16	99.84397
17	99.83901
18	99.76981
19	99.86632
20	99.85067
21	99.83504
22	99.83547
23	99.85688

Table S3. CE for 0.2 M non-aqueous $Zn(TFSI)_2$/ACN, cycles 1-3 and 4, 7, 10 are excluded

CV #	CE, %
5	95.55349
6	97.6379
8	99.8842
9	99.8995
11	99.97408
12	99.94918

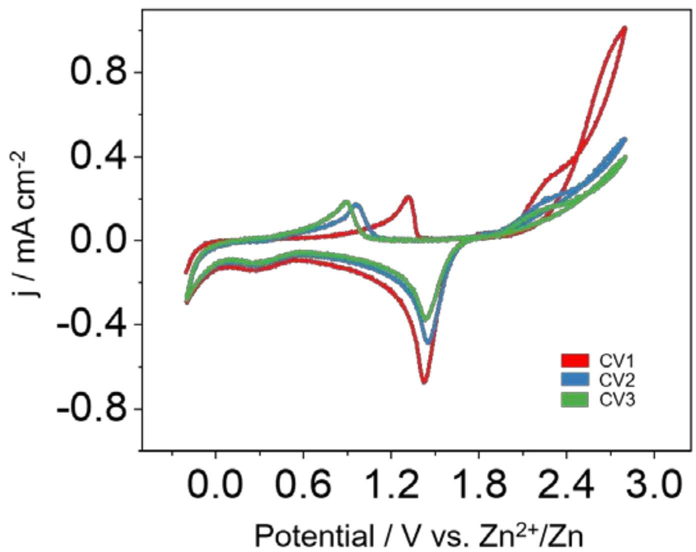

Figure S6. The first 3 CV cycles for Zn electrodeposition from non-aqueous $Zn(TFSI)_2$ electrolyte.

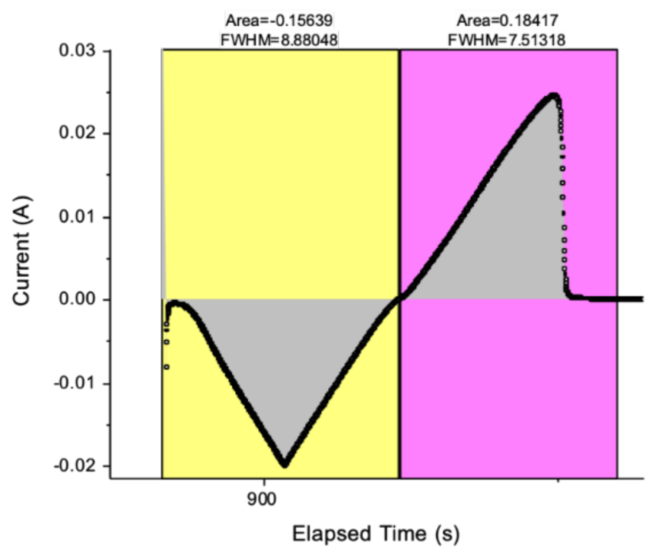

Figure S7. 4th CV for aqueous $ZnCl_2$ electrolyte showing integrated plating and stripping currents used for CE calculations.

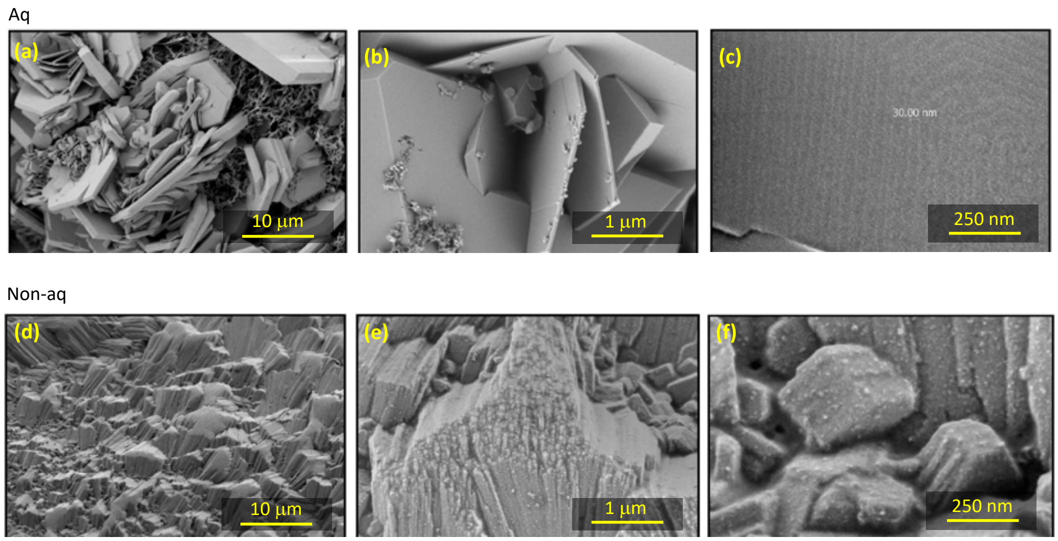

Figure S8. SEM images from the electrochemically deposited Zn of the 0.2 M $ZnCl_2$ in H_2O (panels a-c), and the non-aqueous 0.2 M $Zn(TFSI)_2$ in ACN (panels d-f).