

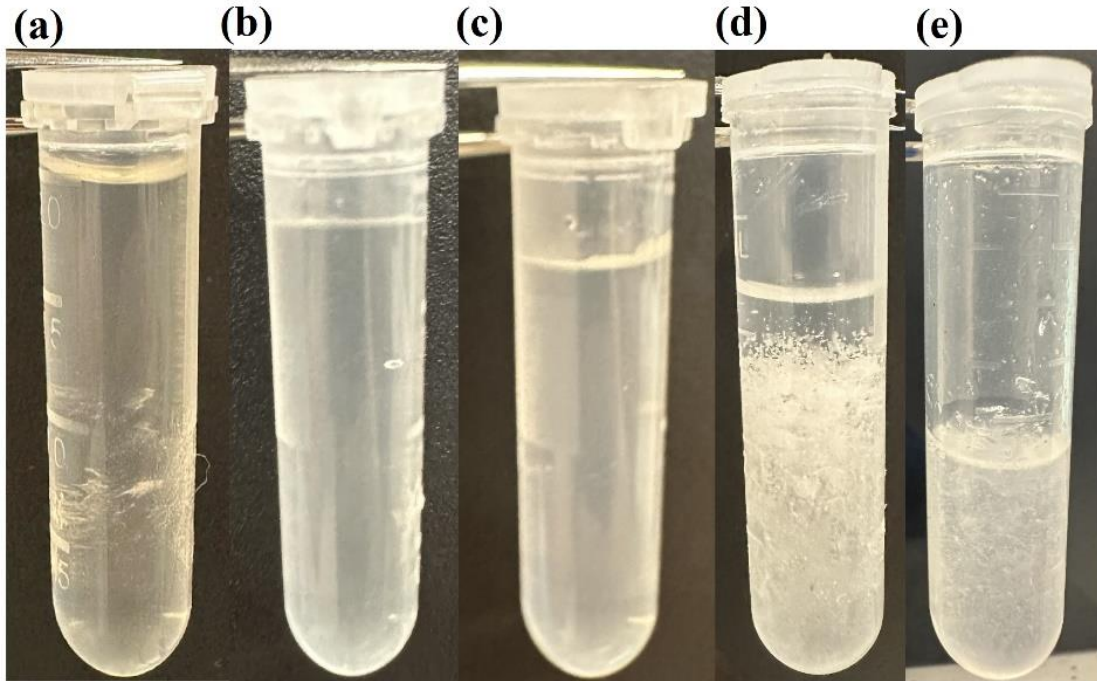
Supplementary Materials

Regulating the solvation environment of hybrid electrolytes towards high-temperature zinc-ion storage

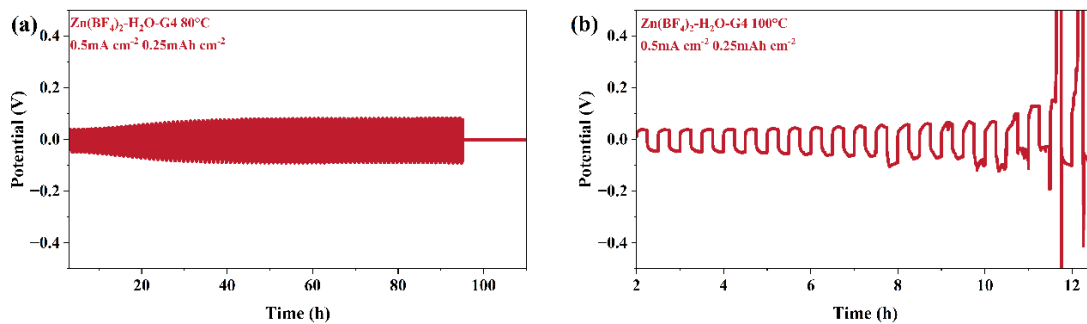
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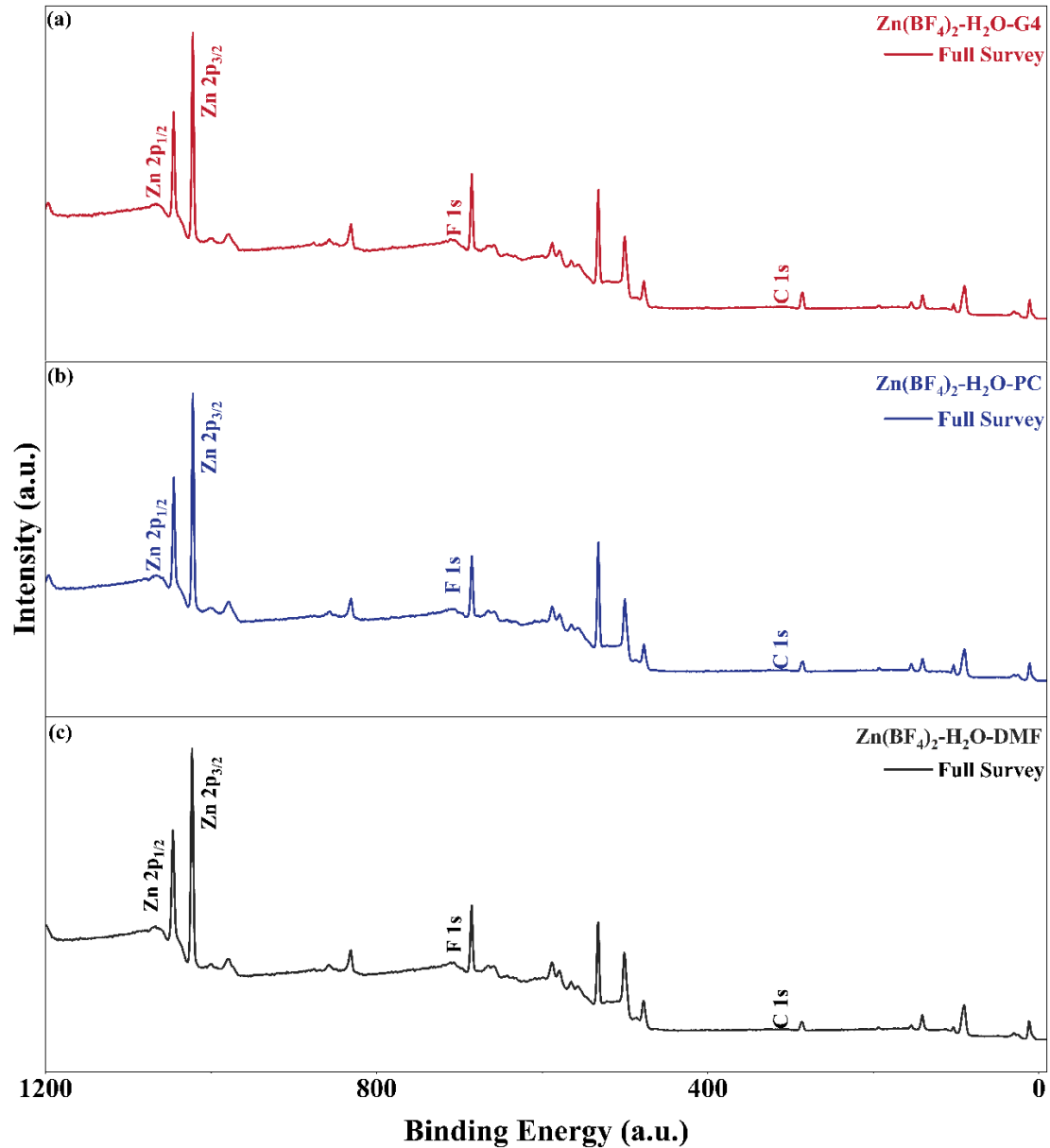
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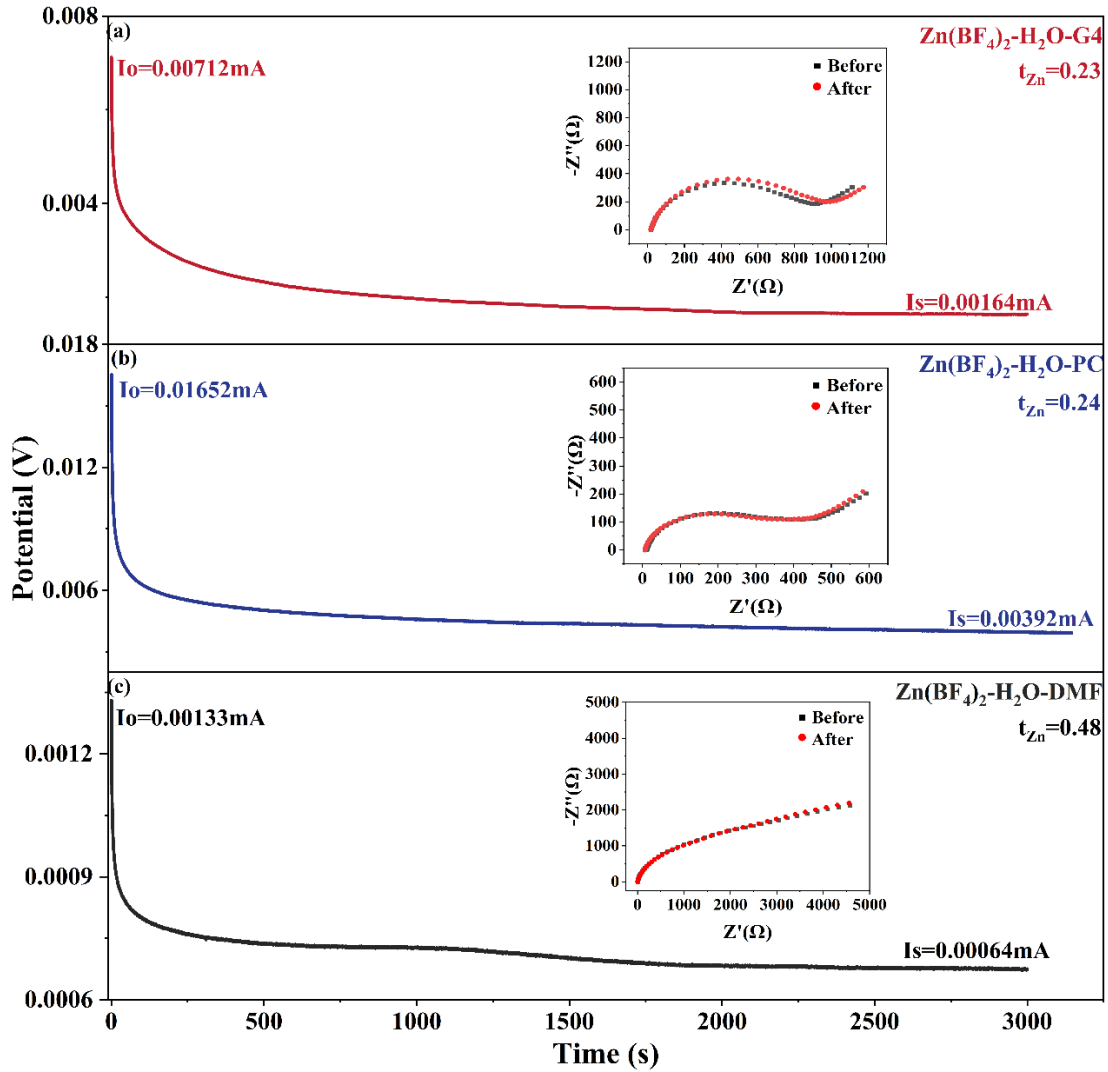
Supplementary Figure 1. Solubility of $\text{Zn}(\text{BF}_4)_2 \cdot x\text{H}_2\text{O}$ in five different organic solvents (a) G4, (b) PC, (c) DMF, (d) DEC, and (e) DMSO.



Supplementary Figure 2. Cycling performance of the $\text{Zn}||\text{Zn}$ symmetric cells using G4-based hybrid electrolyte. Galvanostatic Zn plating/stripping under 0.5mA cm^{-2} and 0.25mAh cm^{-2} in temperatures of (a) 80°C and (b) 100°C .



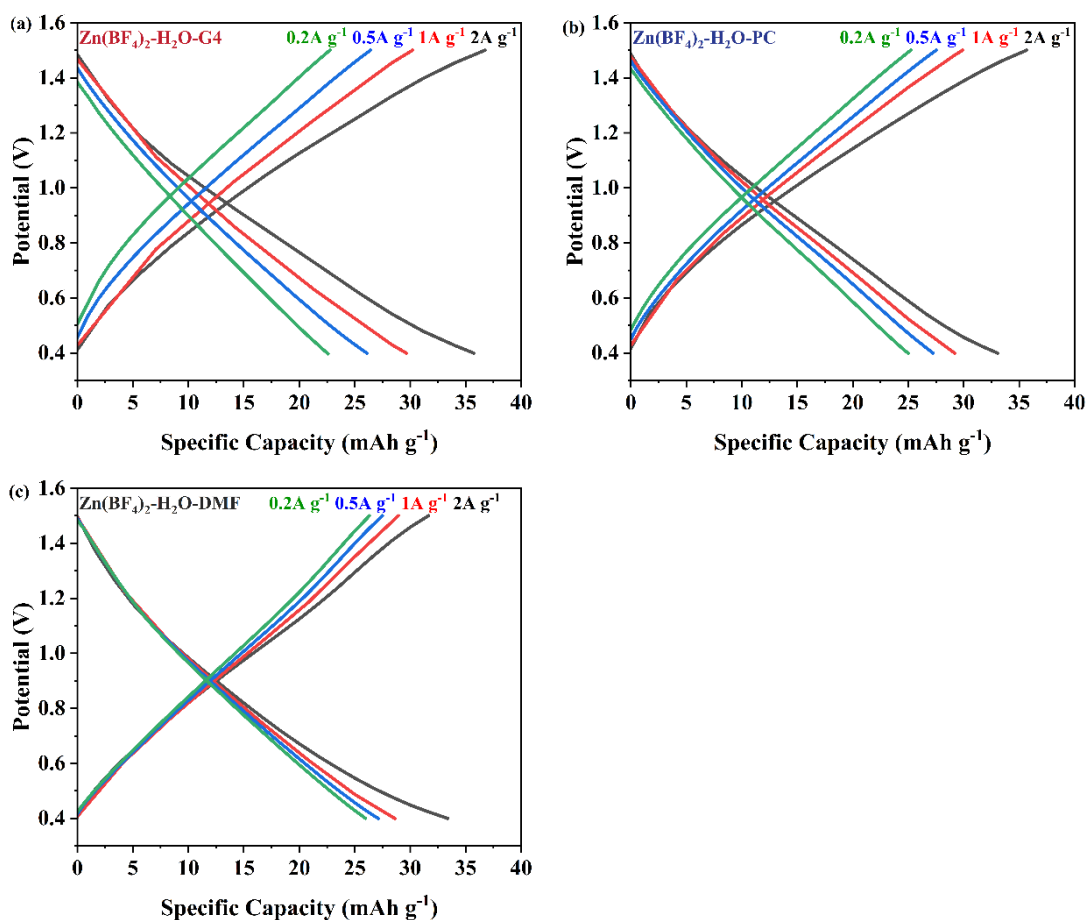
Supplementary Figure 3. Full survey XPS spectra of Zn anodes cycled in (a) G4-based, (b) PC-based, and (c) DMF-based hybrid electrolytes, after 20 cycles at 60 °C.



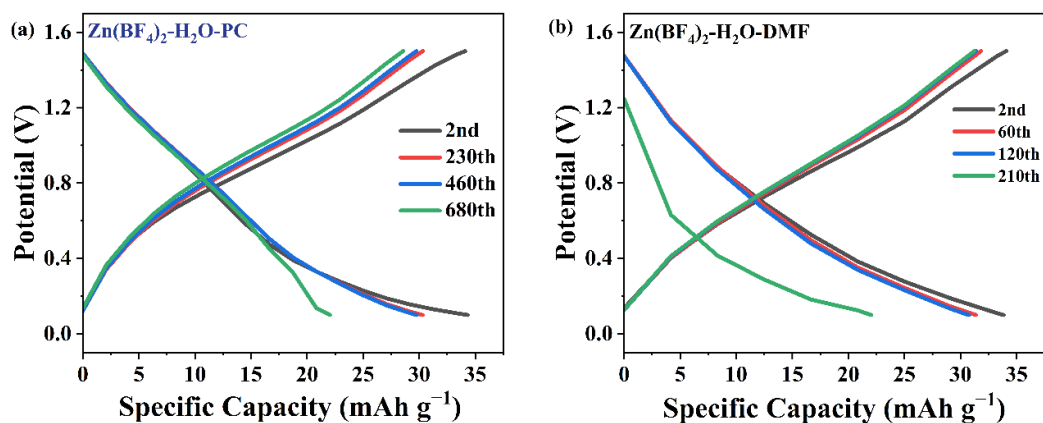
Supplementary Figure 4. Chronoamperometry (CA) polarization curves with an applied voltage of 10 mV for the symmetric cells consisting of Zn electrodes in (a) G4-based, (b) PC-based, and (c) DMF-based hybrid electrolytes. The transference number of Zn^{2+} (t_{Zn}) was calculated based on the formula:

$$t_{Zn} = \frac{I_s(\Delta V - I_o R_o)}{I_o(\Delta V - I_s R_s)}$$

where ΔV is the applied potential (10 mV), I_o and I_s are the initial and steady-state currents, and R_o and R_s are the initial and steady-state electrode resistances, respectively^[1].



Supplementary Figure 5. Rate performance of Zn||AC cells in (a) G4-based; (b) PC-based; (c) DMF-based electrolytes at 60°C.



Supplementary Figure 6. GCD curves of Zn||AC cells at 0.5A g^{-1} in (a) DMF-based electrolyte from the 2nd to 210th cycle, and (b) PC-based electrolyte from the 2nd to 680th cycle at 60 °C.

References

1. Kaminski GA, Friesner RA, Tirado-Rives J, Jorgensen WL. Evaluation and Reparametrization of the OPLS-AA Force Field for Proteins via Comparison with Accurate Quantum Chemical Calculations on Peptides. *J Phys Chem B* **2001**; 105: 6474-87. DOI: 10.1021/jp003919d