Supplementary Information

Unlocking High-Performance Zinc Batteries via Haloacetamide-Regulated Nucleation and Interface Chemistry

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[1]Author contributions
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Detailed calculation

1. The calculation formulas of Zn-ion transference number

The polarization was measured with a polarization potential (ΔV) of 10 mV in a Zn||Zn symmetric cells and Bruce–Vincent–Evans equation (Equation (1)) was used to calculate the Zn-ion transference number ($t_{Zn}{}^{2+}$).

$$t_{Zn^+} = \frac{I_s(\Delta V - I_0 R_0)}{I_0(\Delta V - I_s R_s)} \quad (1)$$

where I_0 and I_s are the initial current and steady-state current of the polarization currents, R_0 and R_s are the initial and steady-state interfacial resistances of Zn/electrolyte.

2. COMSOL Multiphysics simulation

COMSOL Multiphysics 6.2 was used to perform the Finite Element Analysis (FEA) simulations, wherein the Secondary Current Distribution module was paired with the Transport of Diluted Species. To observe the behavior of zinc ions in the presence of an electric field, the electrolyte potential (ϕ) was first computed with respect to the Butler-Volmer kinetics expression where α is the transfer coefficient for the anode and cathode, i_o is the exchange current density, σ is the conductivity, F is Faraday's constant, and η is the overpotential.

$$-\sigma\nabla\phi = i_o(\exp\left(\frac{\alpha_a F\eta}{RT}\right) - exp\left(\frac{-\alpha_c F\eta}{RT}\right))$$

The obtained electrolyte potential was then coupled with the Nernst-Einstein relation to observe the migration of zinc ions in an electric field. Initial concentration values were first set to 2 M, and the study was set to run for 100 seconds. It should be noted however that the simulations represent an ideal environment in thermal equilibrium. Nevertheless, the obtained results successfully showed the "tip effect" from the uncoated, bumpy system.

Table S1. Parameter values for the COMSOL simulation

Variable name	Value
Electrolyte conductivity	5.7 S m^{-1}
Exchange current density	10 A m^{-2}
Anodic & cathodic transfer coefficients	0.5
Diffusion coefficient	3.3E^{-10} m^2 s^{-1}
Initial bulk concentration	2 M

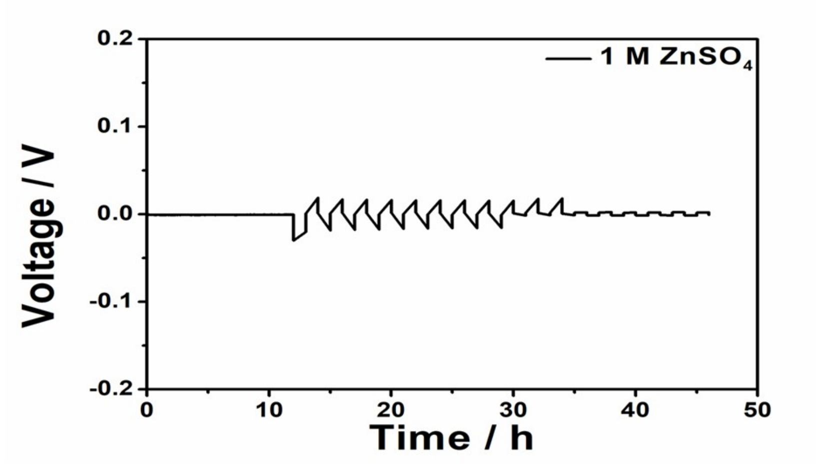

Figure S1. Chronopotentiometry profiles using 1 M $ZnSO_4$ electrolyte.

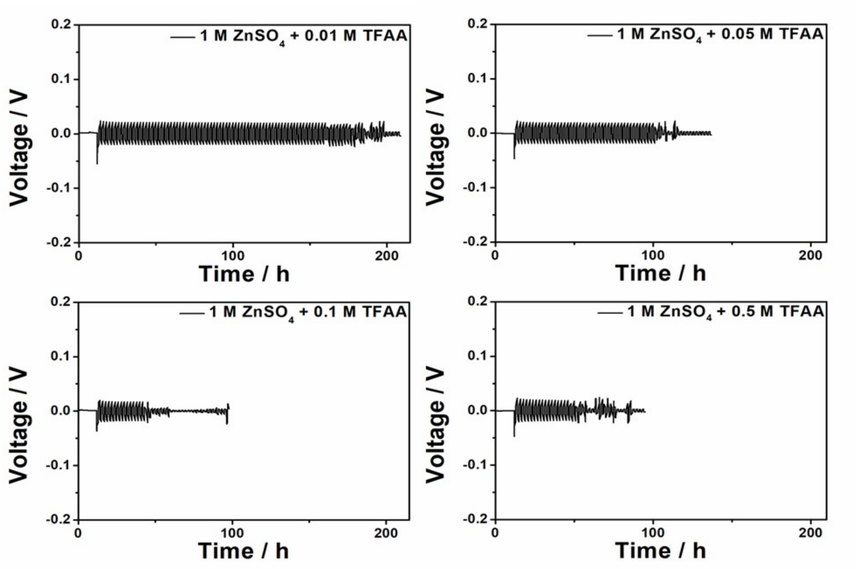

Figure S2. Chronopotentiometry profiles in 1 M $ZnSO_4$ electrolytes containing various concentrations of TFAA.

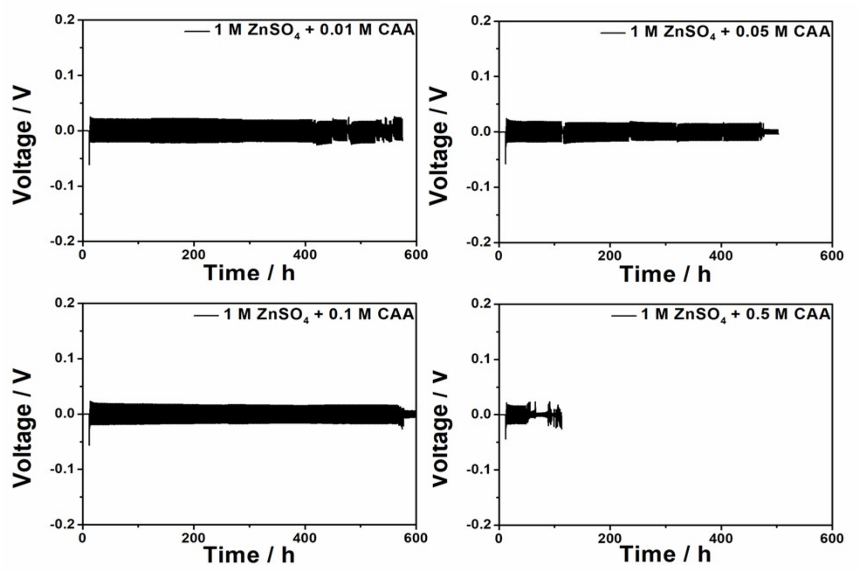

Figure S3. Chronopotentiometry profiles in 1 M $ZnSO_4$ electrolytes containing various concentrations of CAA.

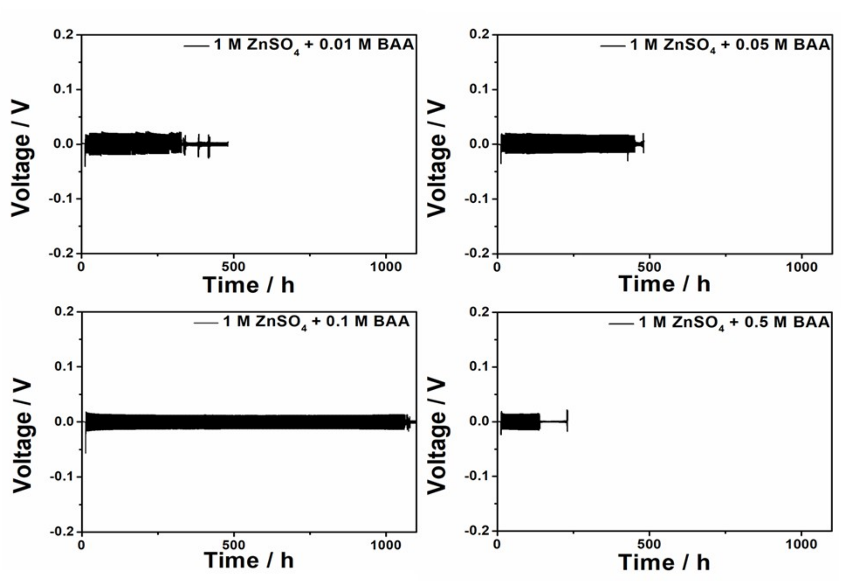

Figure S4. Chronopotentiometry profiles in 1 M $ZnSO_4$ electrolytes containing various concentrations of BAA.

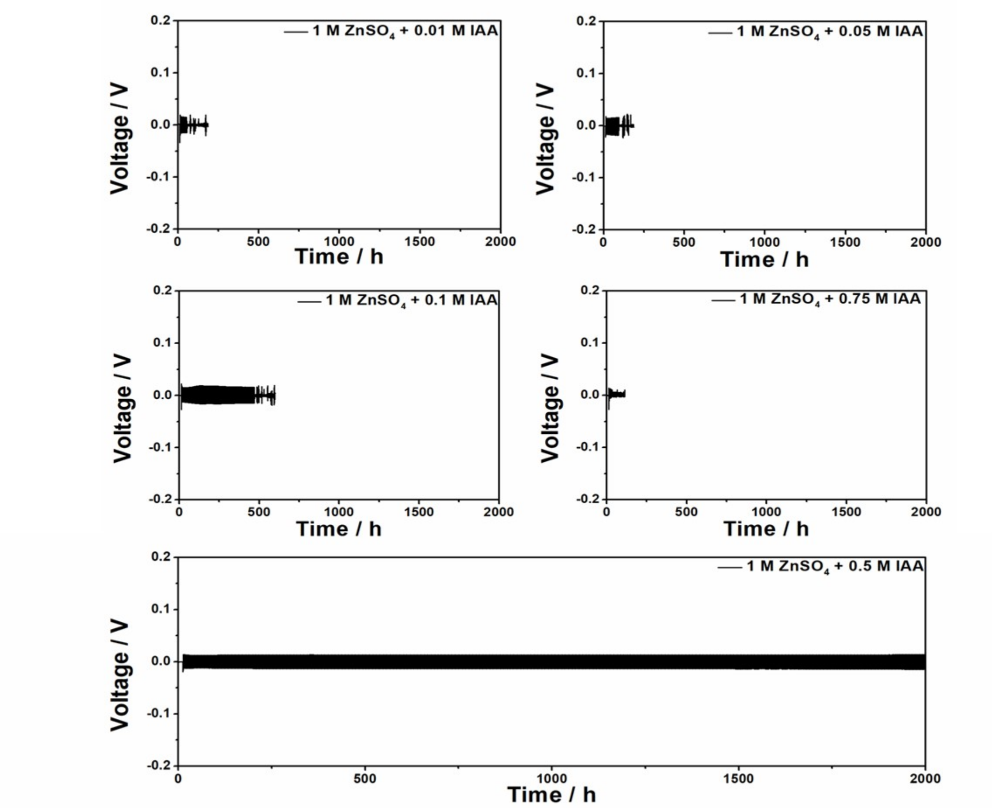

Figure S5. Chronopotentiometry profiles in 1 M $ZnSO_4$ electrolytes containing various concentrations of IAA.

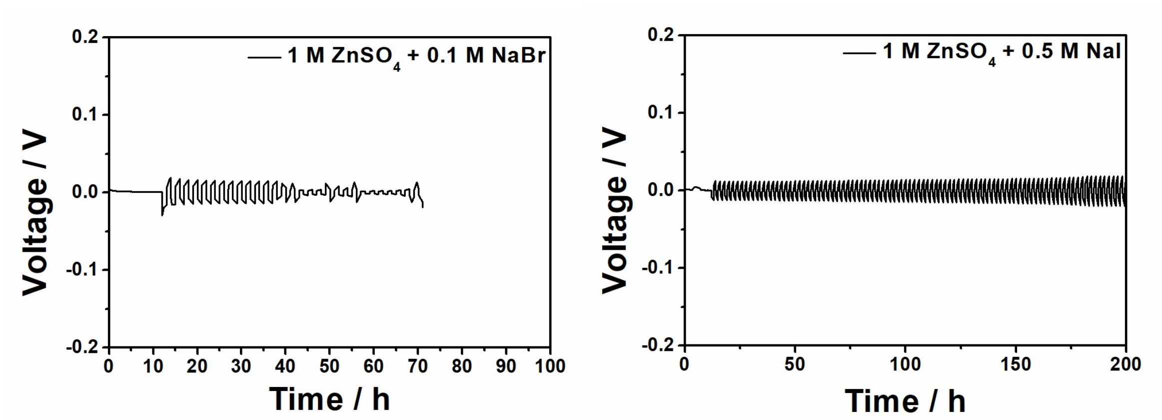

Figure S6. Chronopotentiometry profiles in 1 M $ZnSO_4$ electrolytes with the addition of 0.1 M

NaBr or 0.5 M NaI.

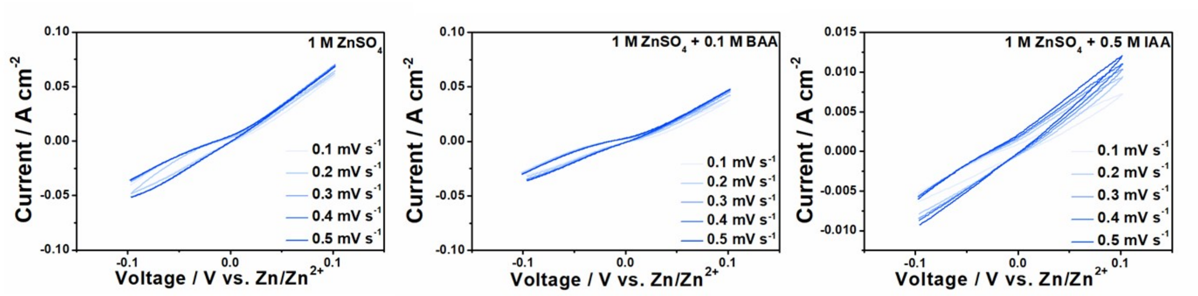

Figure S7. Cyclic voltammetry curves of Zn||Zn symmetric cells in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

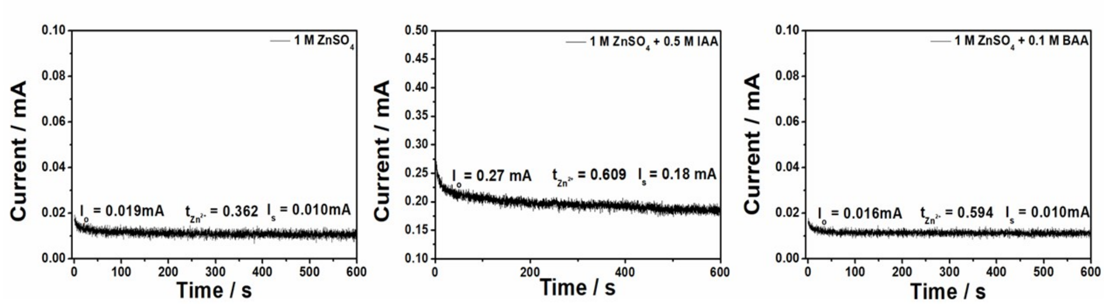

Figure S8. Chronopotentiometry results of Zn||Zn symmetric cells in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

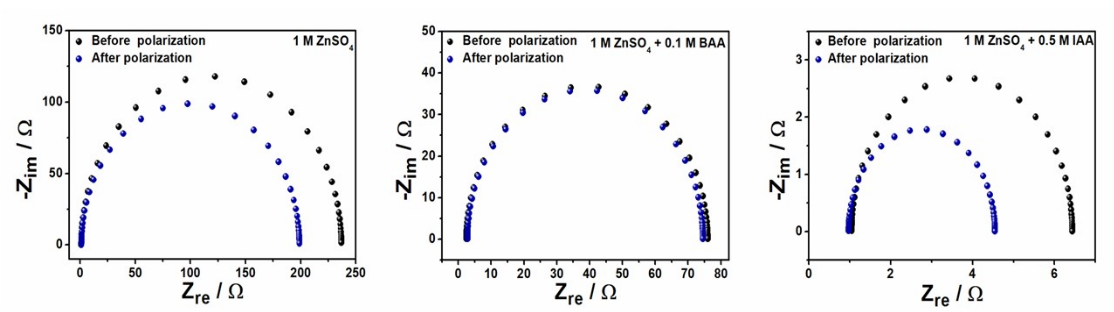

Figure S9. EIS spectra before and after polarization of Zn||Zn symmetric cells assembled with

1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

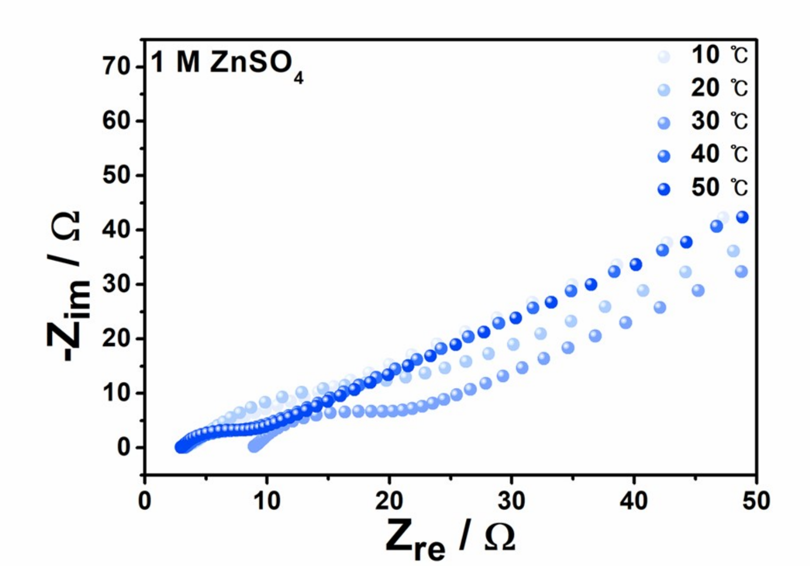

Figure S10. EIS spectra of Zn||Zn symmetric cells with 1 M $ZnSO_4$ electrolyte at different temperatures.

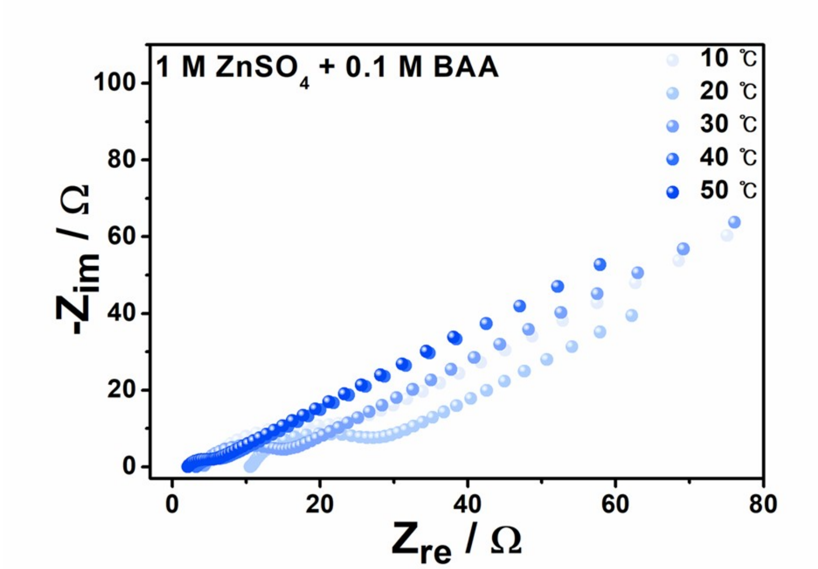

Figure S11. EIS spectra of Zn||Zn symmetric cells with 1 M $ZnSO_4$ electrolyte with bromoacetamide at different temperatures.

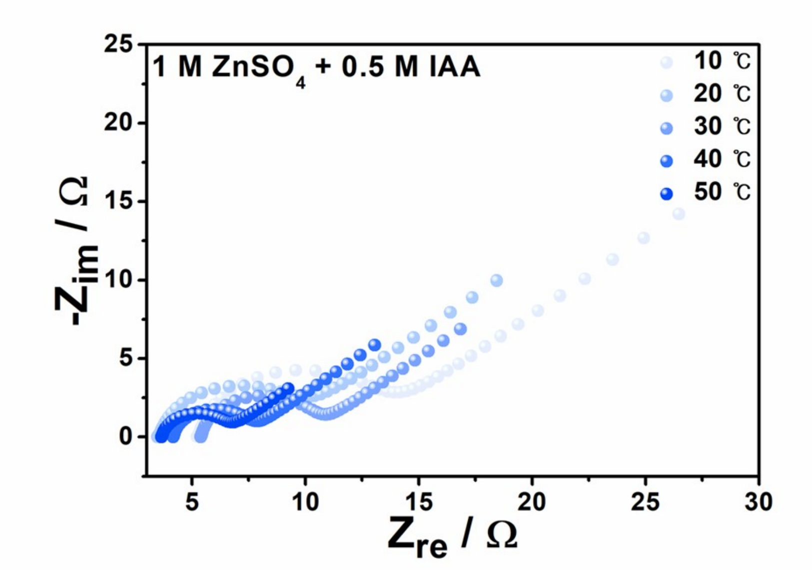

Figure S12. EIS spectra of Zn||Zn symmetric cells with 1 M $ZnSO_4$ electrolyte with iodoacetamide at different temperatures.

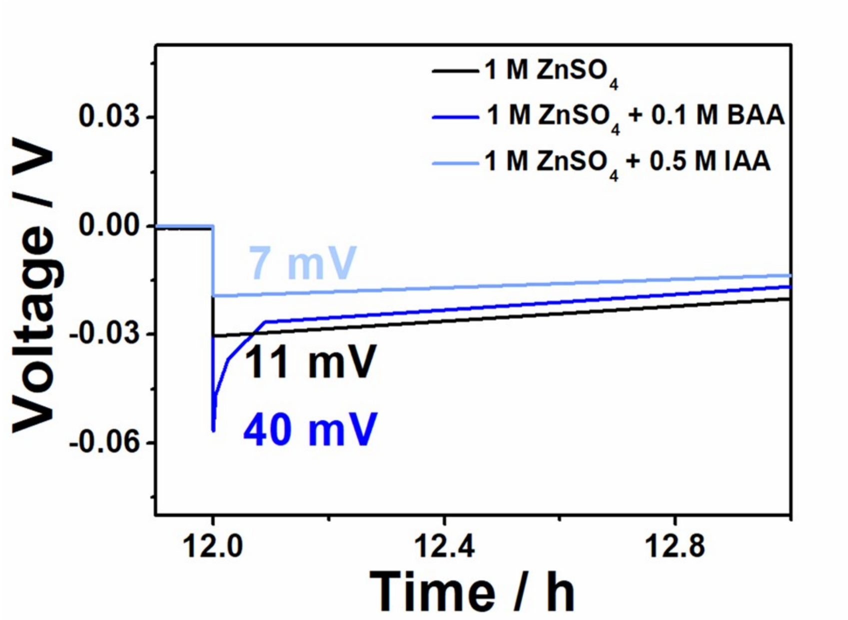

Figure S13. Overpotential comparison during Zn||Zn symmetric cell cycling in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

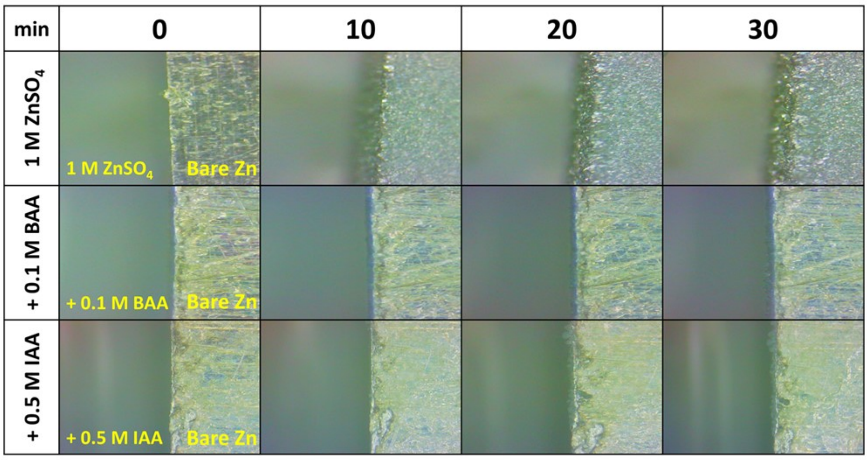

Figure S14. In situ optical microscopy images showing Zn-ion deposition on Zn metal electrodes in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

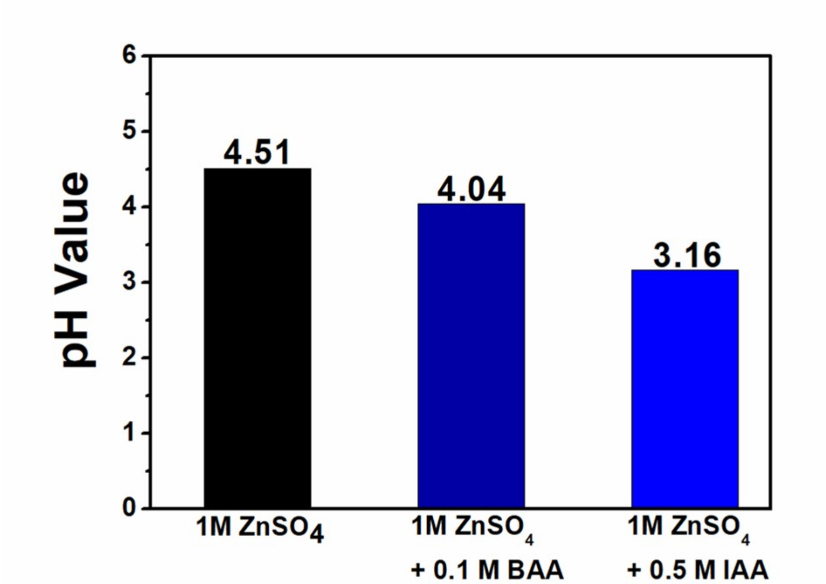

Figure S15. pH values of 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

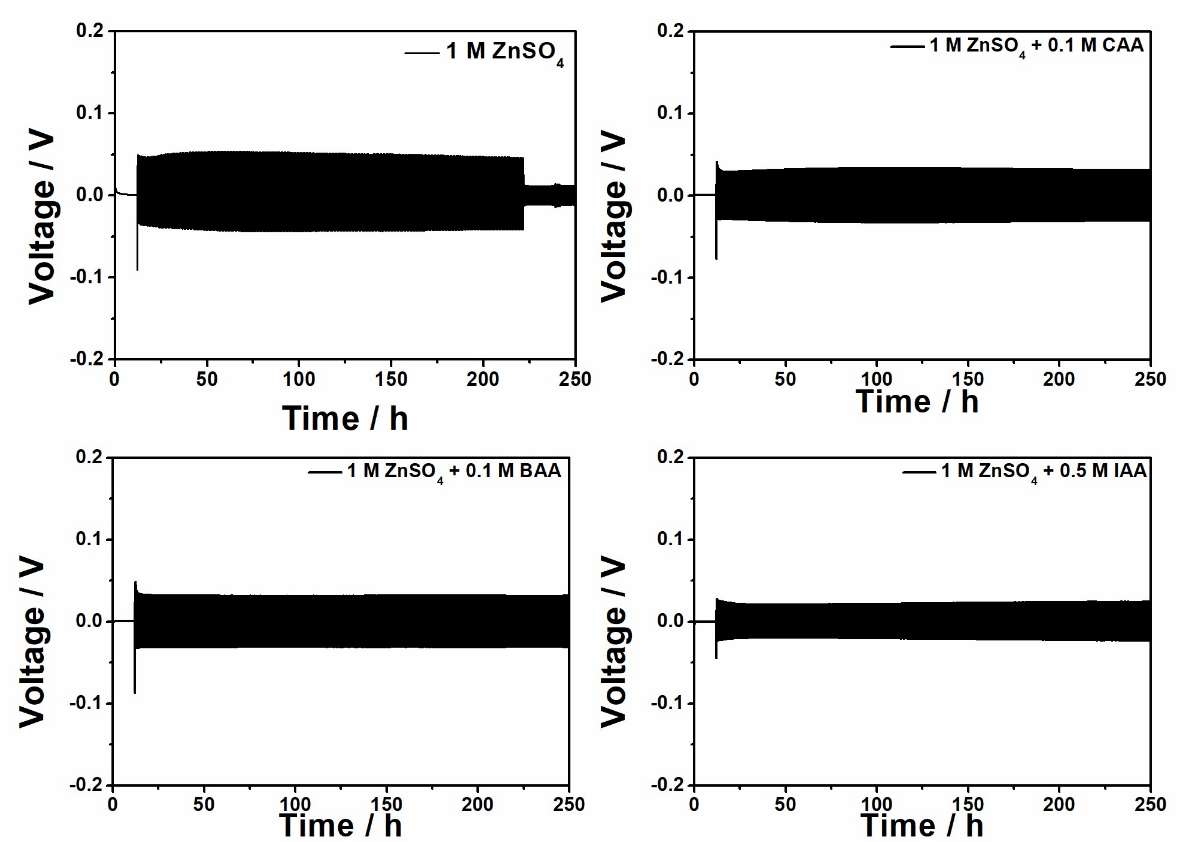

Figure S16. Chronopotentiometry profiles of Zn||Zn symmetric cells in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide at 5 mA cm^{-2} and a fixed capacity of 5 mAh cm^{-2}.

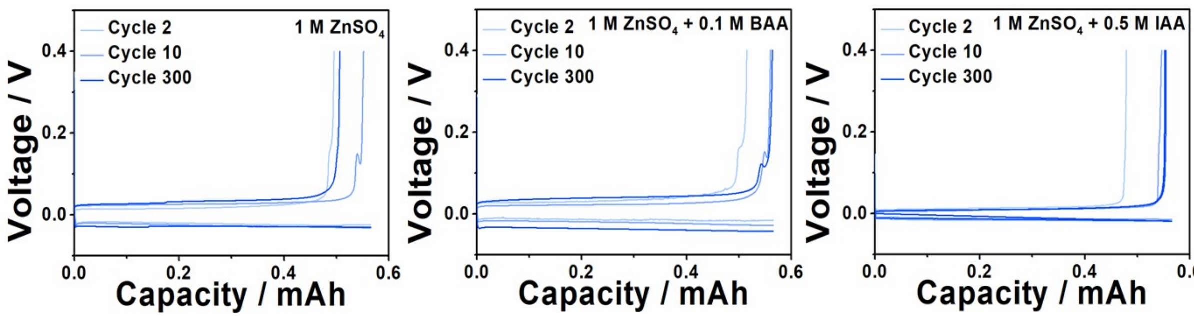

Figure S17. Voltage profiles of Zn||Cu half-cell in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

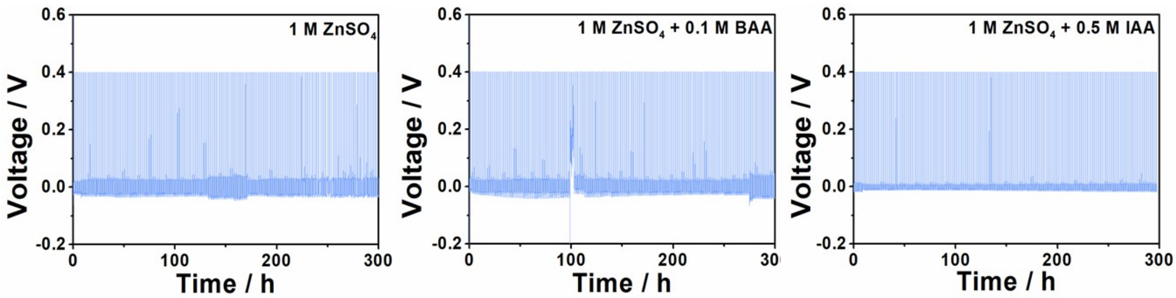

Figure S18. Voltage vs. time plot of Zn||Cu half-cells in 1 M $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

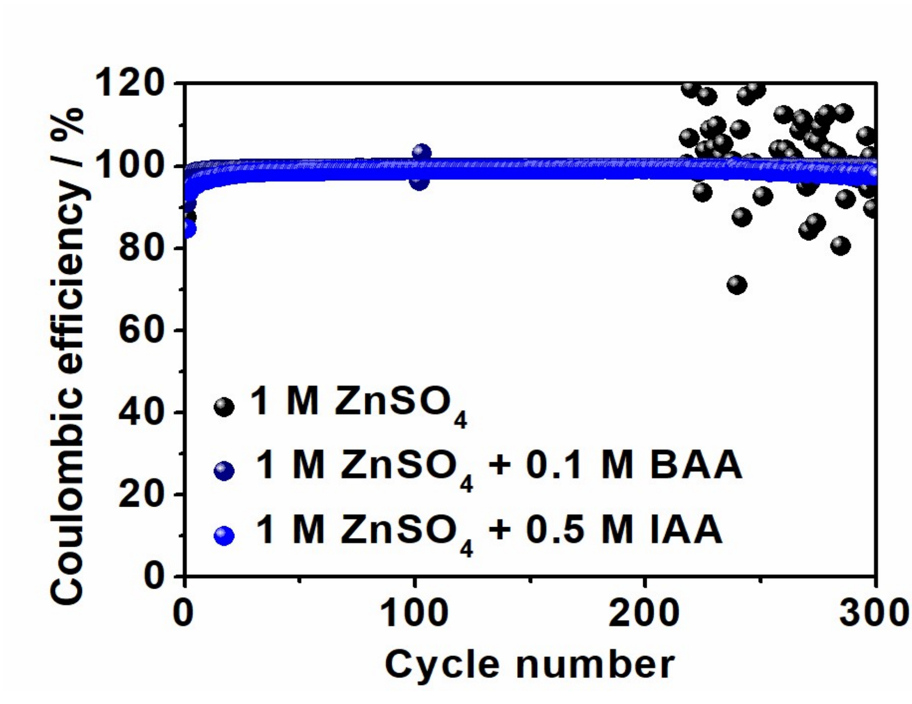

Figure S19. Coulombic efficiency of Zn||Cu half-cells in 1 M $ZnSO_4$ electrolytes with and without the addition of additives, bromoacetamide and iodoacetamide.

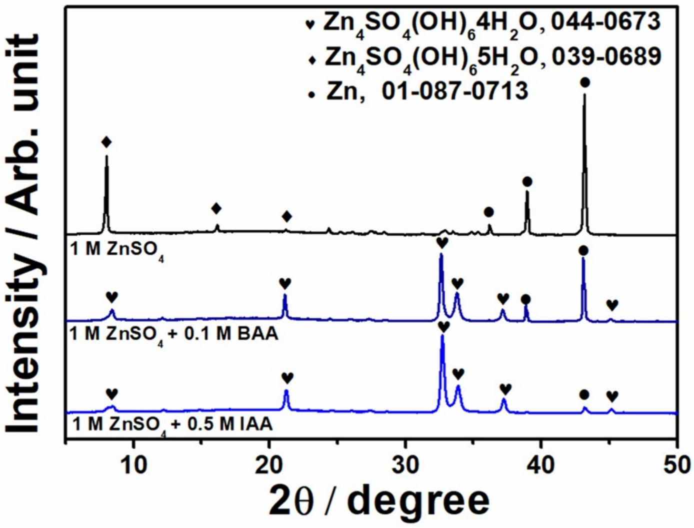

Figure S20. X-ray diffraction patterns of Zn metal electrode soaked for 5 days in 1 M $ZnSO_4$ electrolyte with and without bromoacetamide or iodoacetamide.

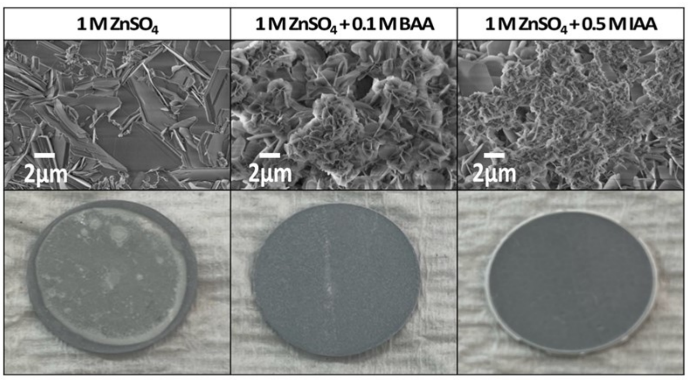

Figure S21. SEM images and digital photographs of Zn metal electrode soaked for 5 days in 1 M $ZnSO_4$ electrolyte with and without bromoacetamide or iodoacetamide.

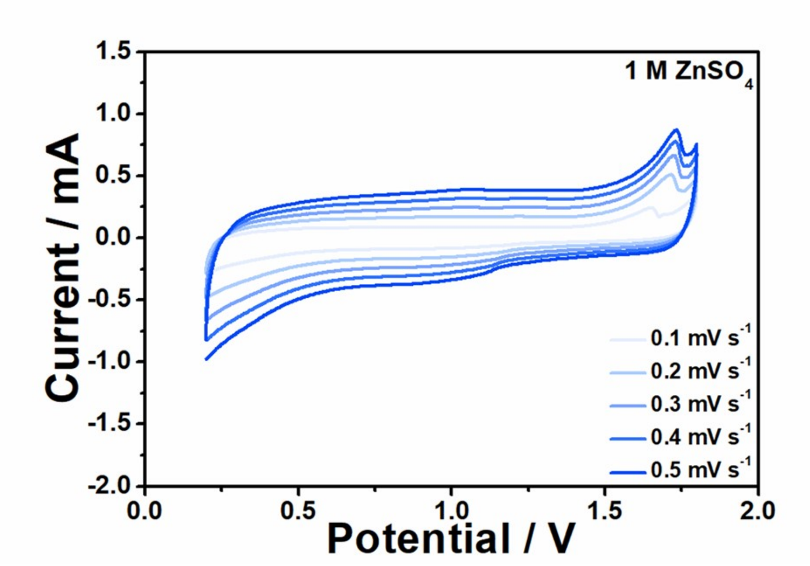

Figure S22. Cyclic voltammetry curves at various scan rates for $ZnSO_4$ electrolytes.

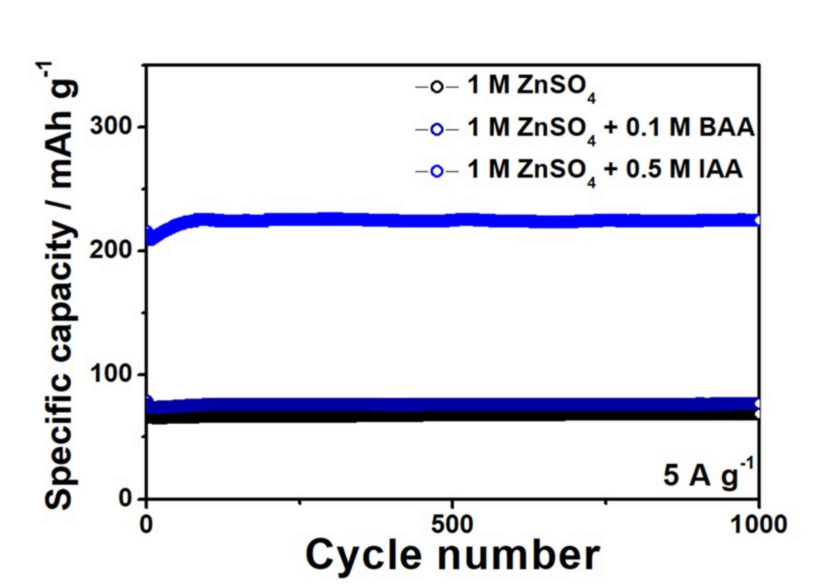

Figure S23. Galvanostatic charge–discharge curves at 5 A g^{-1} in $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.

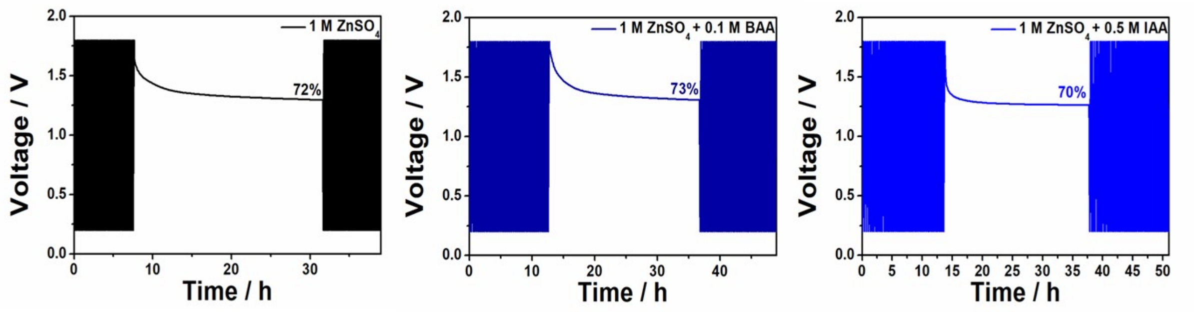

Figure S24. Self-discharge behavior of Zn||AC cells in $ZnSO_4$ electrolytes with and without bromoacetamide or iodoacetamide.