

Supplementary Materials for
**Designing interphases for practical aqueous zinc flow batteries with high
power density and high areal capacity**

Shuo Jin *et al.*

Corresponding author: Yong Lak Joo, ylj2@cornell.edu; Lynden A. Archer, laa25@cornell.edu

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Figs. S1 to S17

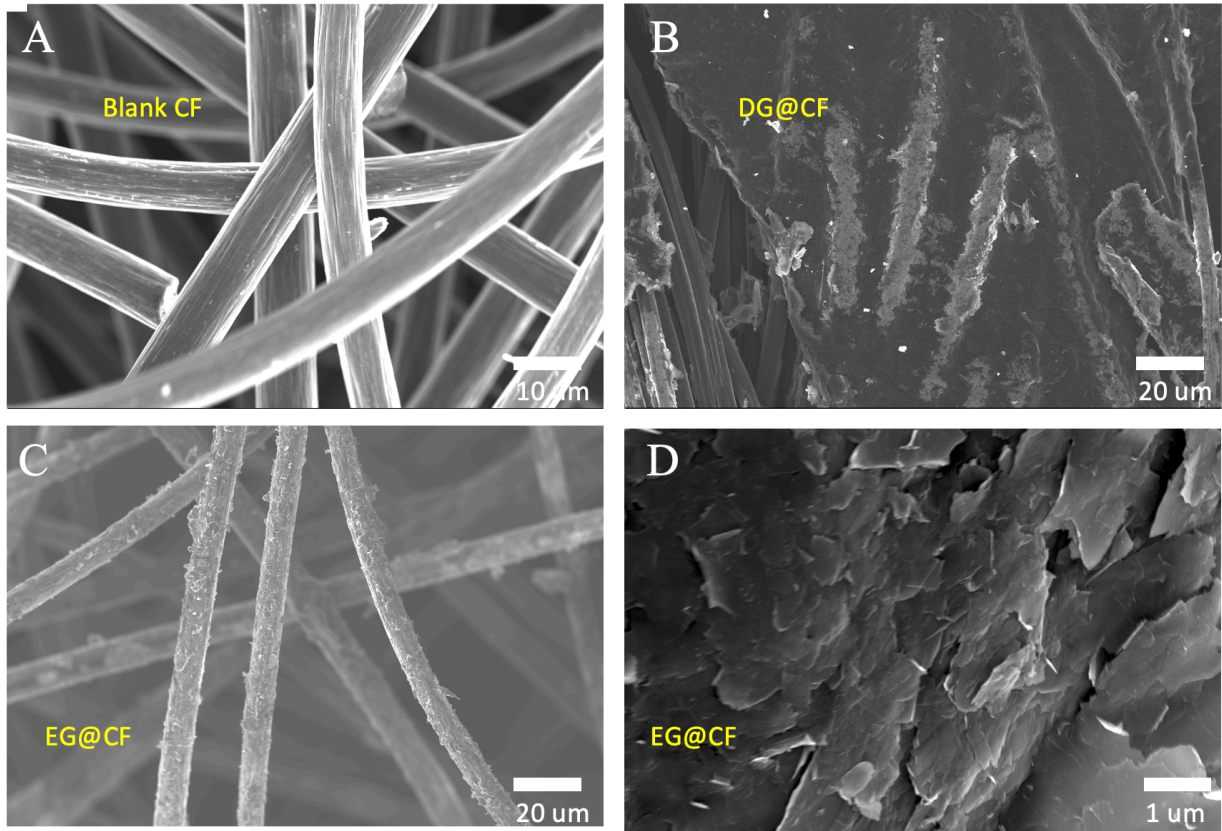


Figure S1. SEM images of graphene morphology. (A) blank carbon felt, (B) doctor-blade graphene at carbon felt, (C)(D) electrospray graphene at the carbon felt.

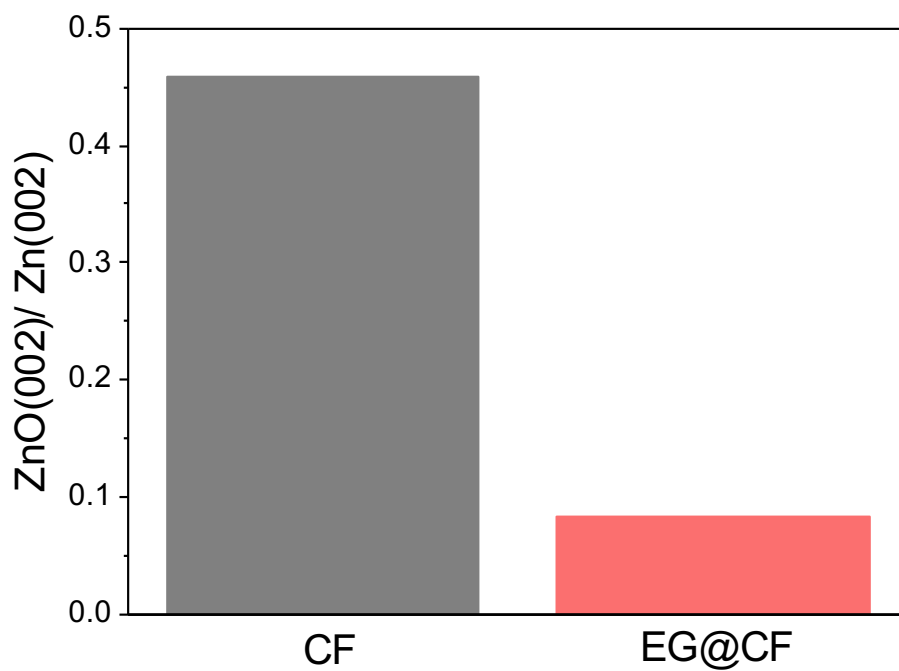


Figure S2. Ratio of ZnO(002)/Zn(002) after cycling at different electrodes. The ratio of ZnO(002)/Zn (002) at the CF surface and EG@CF surface extracted from the Fig. 3E.

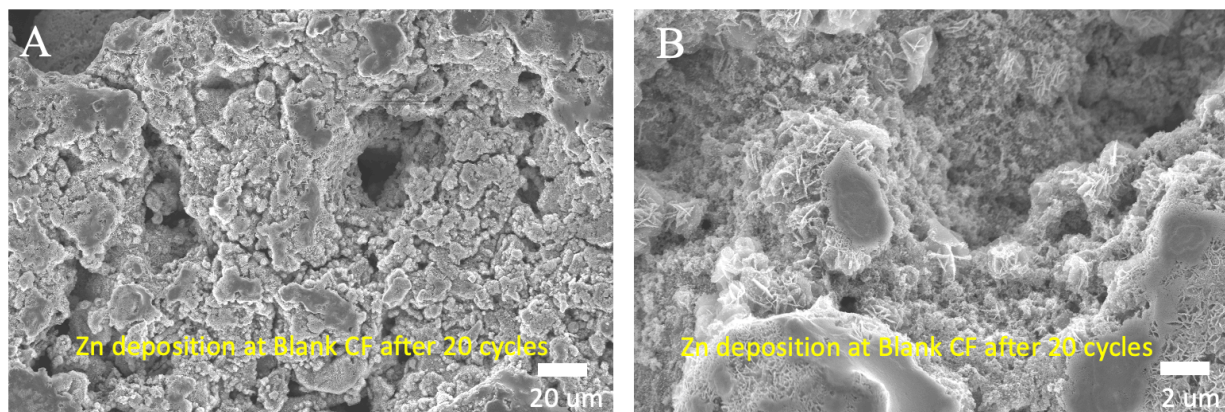


Figure S3. SEM images of Zn electrodeposition in the blank CF. SEM of the Zn electrodeposition at the blank carbon felt after 20 cycles. 40 mA/cm², 10mAh/cm².

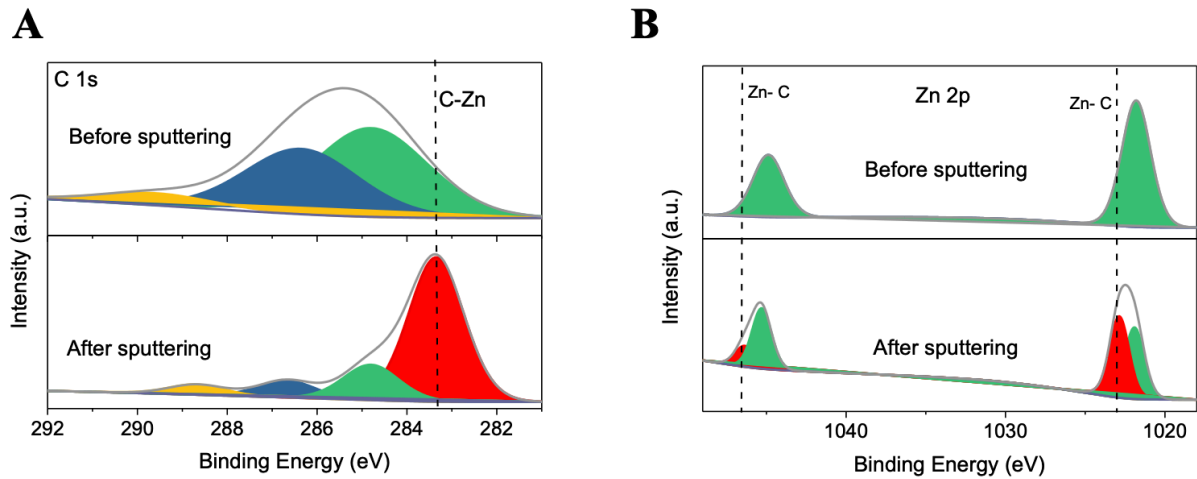


Figure S4. XPS of Zn deposits (1 mAh/cm^2) in the EG@CF. (A) C 1s before sputtering (top) and after sputtering (bottom). (B) Zn 2p before sputtering (top) and after sputtering (bottom). The C-Zn bond appeared after sputtering the Zn deposits at the top layer.

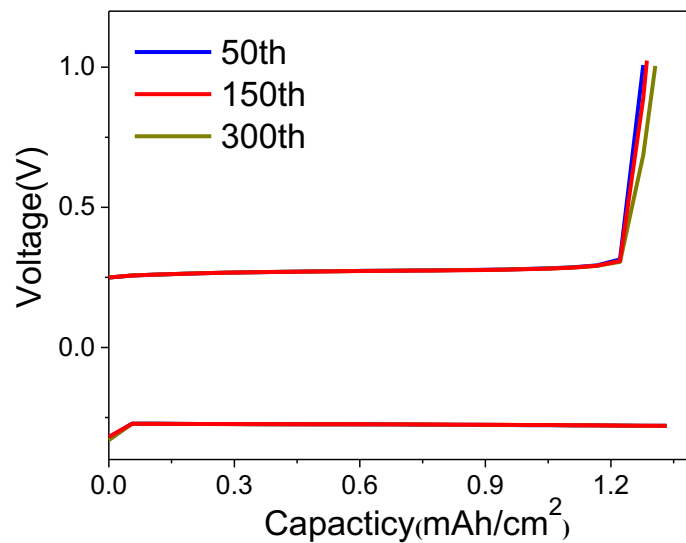


Figure S5. Voltage profile of Zn-EG@CF half cells. Voltage-Capacity curve at different cycles in Fig.2F for the EG@CF. 40 mA/cm², 1.33 mAh/cm².

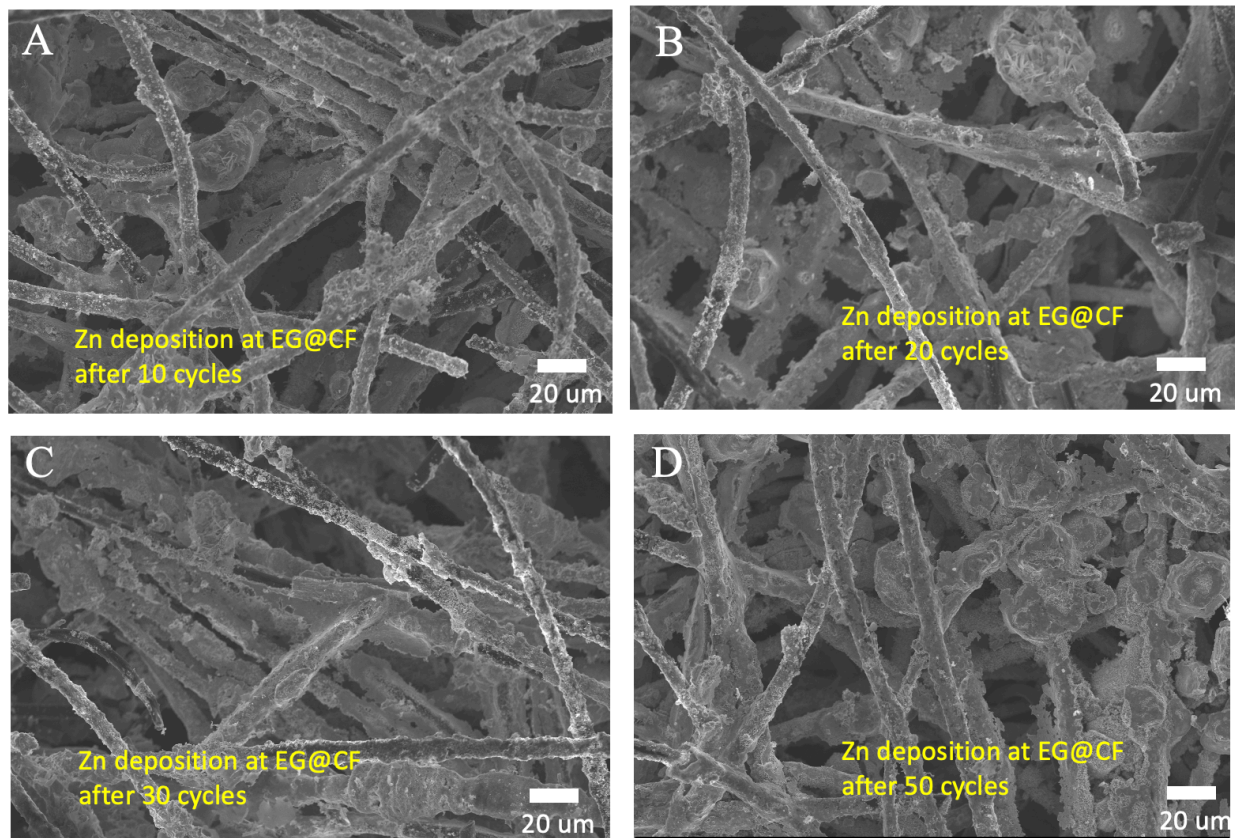


Figure S6. SEM images of Zn electrodeposition in the EG@CF. SEM of the zinc electrodeposition at different cycles for the EG@CF. 40 mA/cm², 10mAh/cm².

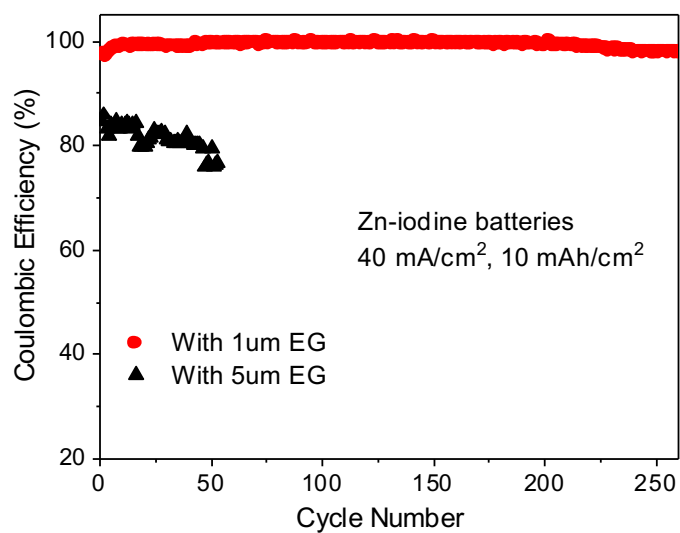


Figure S7. Thickness regulation of EG layers for Zn-Iodine flow batteries. The thickness optimization of EG layers for full Zn-Iodine batteries. Increasing the coating thickness of EG layers leads to the decrease of CE.

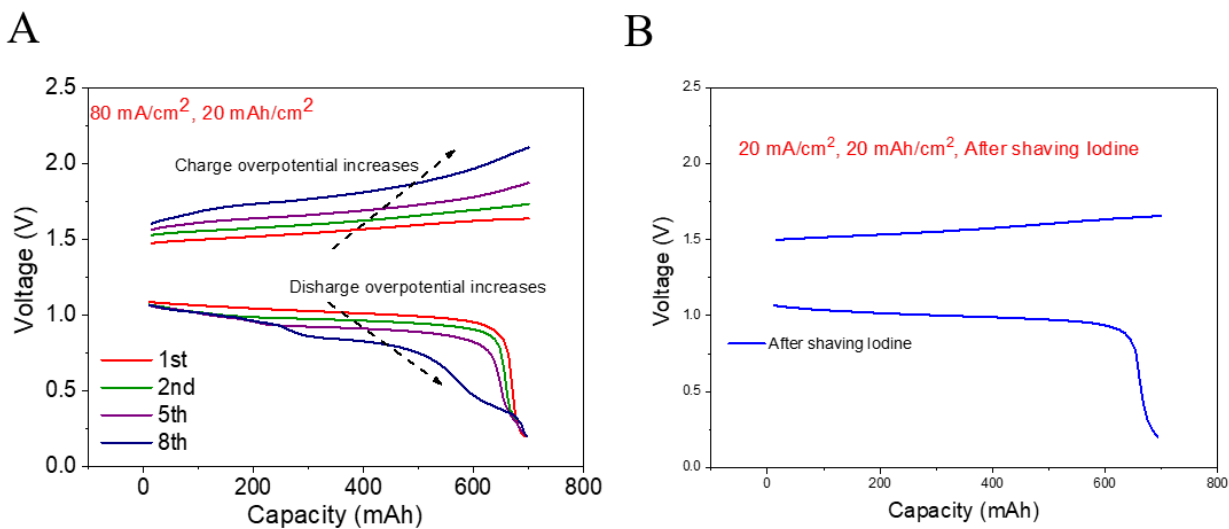


Figure S8. Voltage profile of Zn-Iodine flow batteries using EG@CF as anode and blank CF as cathode. (A) the voltage-capacity curve for the first few cycles. (B) After removing a slice of solid iodine, cycling the same battery again.

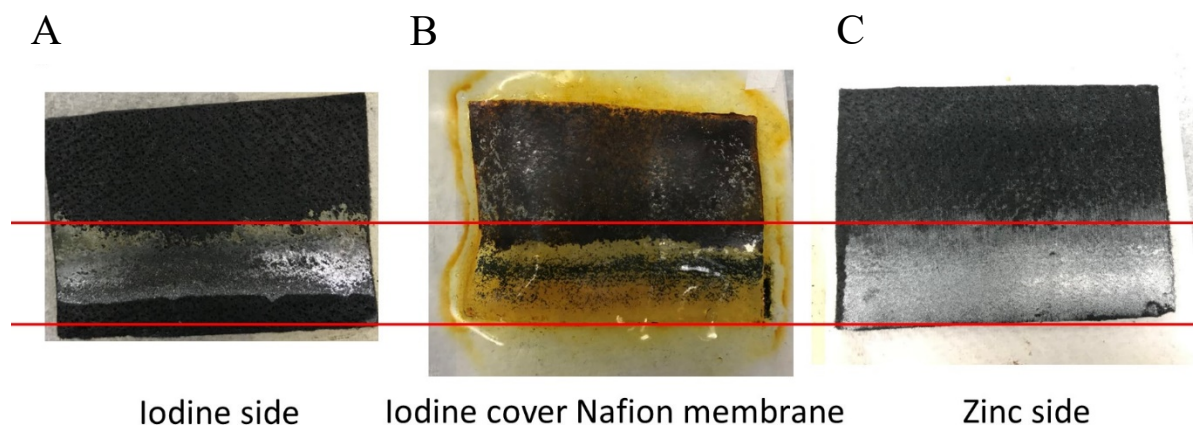


Figure S9. Nafion membrane with a slice of shaved solid iodine after cycling. (A) carbon felt at the iodine side; (B) Nafion membrane; (C) EG@CF at the zinc side. The morphology means the reaction only happens at the shaved place.

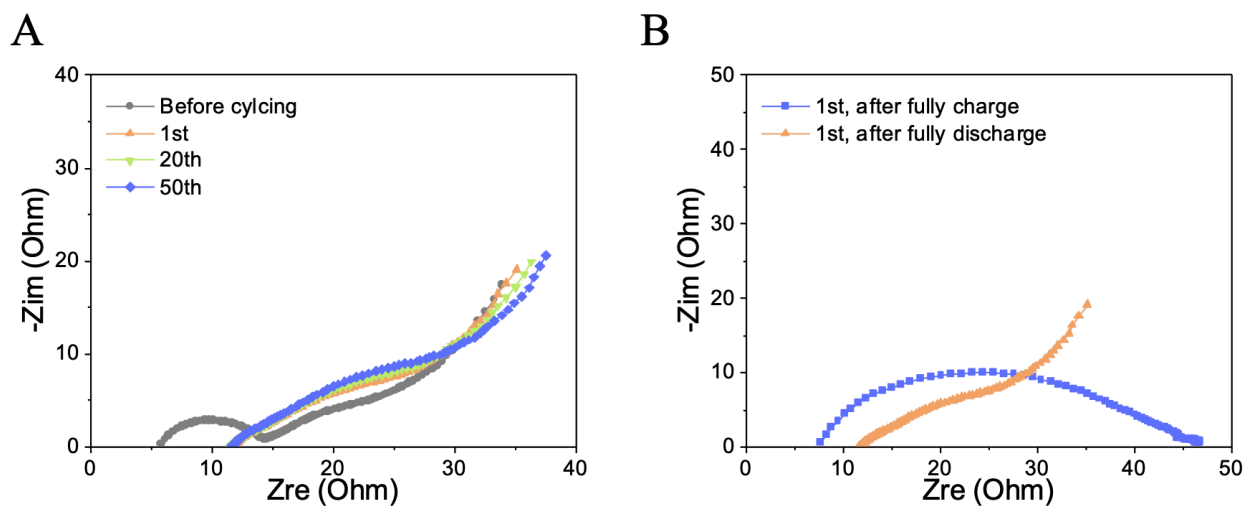


Figure S10. EIS of Zn-Iodine full batteries at different cycles. (A) The EIS spectrums were got after 1st, 20th, 50th fully cycles, which means the battery cell was at a fully discharge state. (B) The EIS was got after fully charged and discharged states at the 1st cycle, respectively. The battery cells were cycled at 80 mA/cm², and 20mAh/cm².

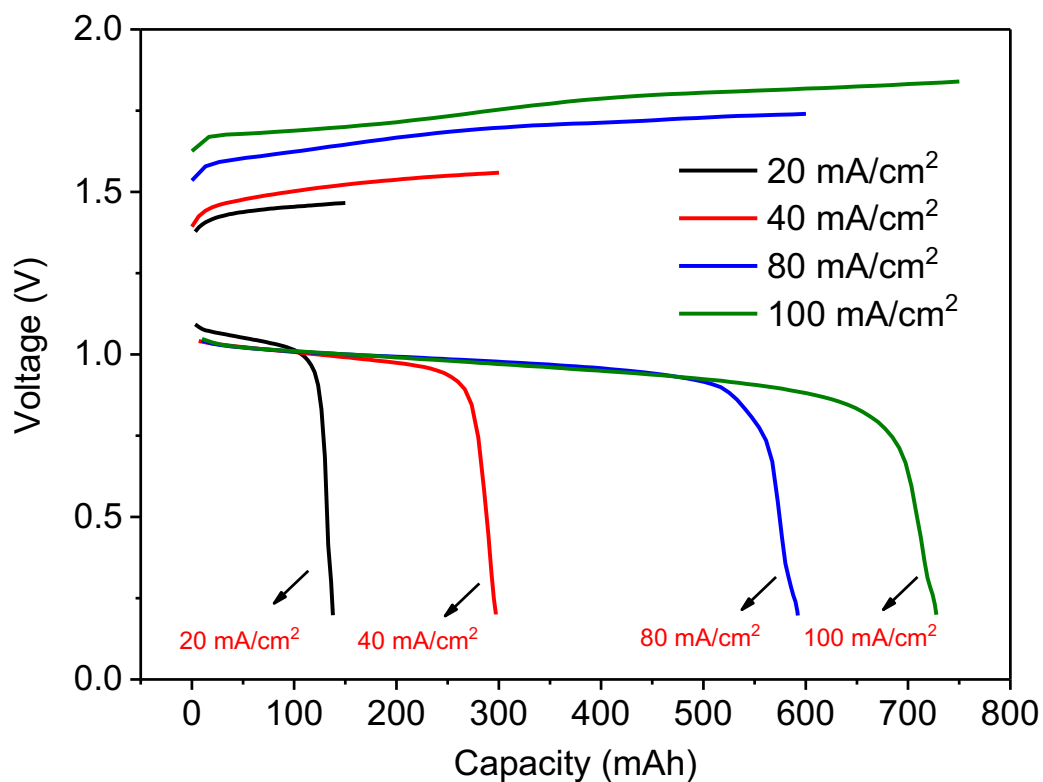


Figure S11. Capacity-voltage curve of Zn-Iodine flow battery at different current densities. The charge time is set as 15 mins. Here, we believe that the ohmic polarization at the charge process results from the high interfacial resistance during the charging process, which can be proved by the EIS at fig.S10B.

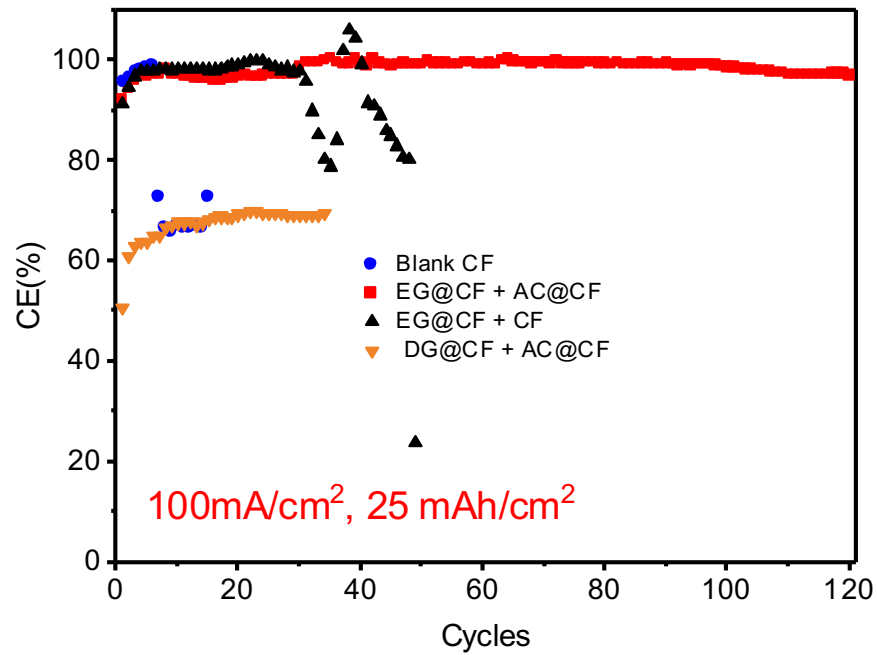


Figure S12. Performance of Zn-Iodine flow batteries using different electrodes. Comparison of cycling performance at different interfaces.

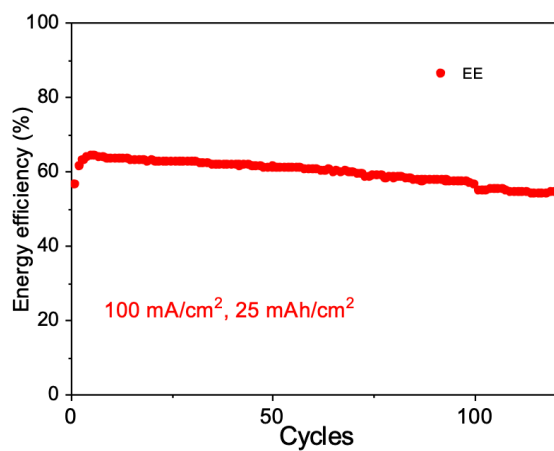
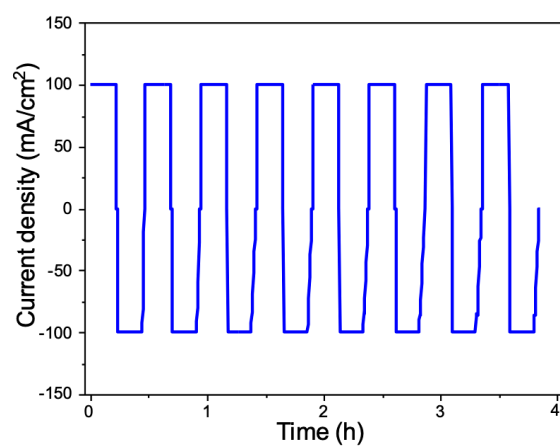
A**B**

Figure S13. Energy efficiency and current density for Zn-iodine flow batteries at high current density and areal capacity. (A) The energy efficiency for the Zn-Iodine flow battery at 100 mA/cm², 25 mAh/cm² for Fig.5D. (B) The current density-Time curve for Fig.5D.

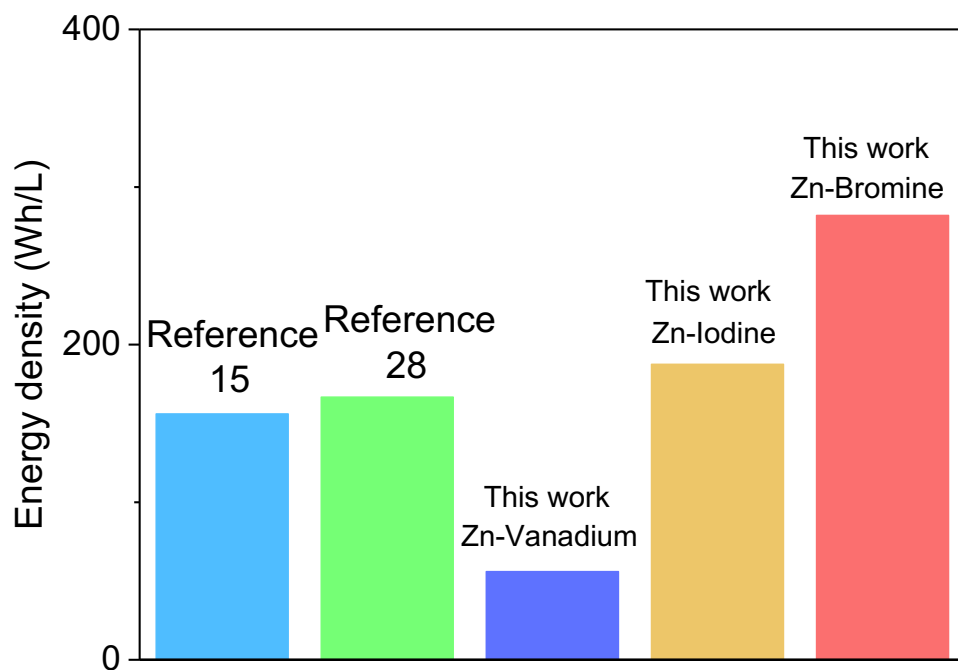


Figure S14. The energy density comparison of different flow battery systems. The references are the other state-of-the-art flow battery systems. Notice: all energy densities were calculated based on the volume of catholyte.

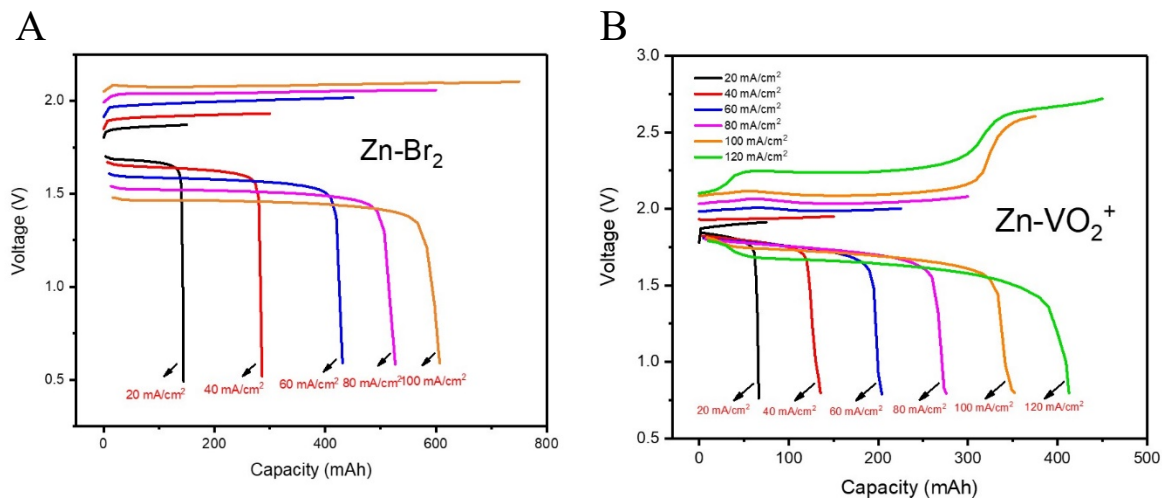
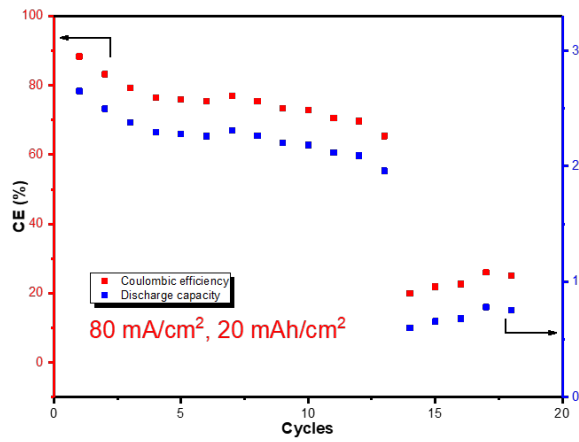


Figure S15. The capacity-voltage curve for extended flow battery systems. (A) Zn-bromine flow battery at different current densities; (B) Zn-Vanadium flow battery at different current densities. The charge time is set as 15 mins.

A



B

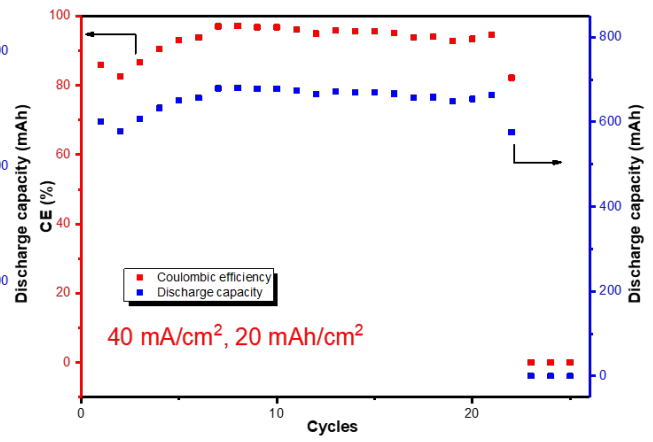


Figure S16. The control cases of cycling performance of the extended flow battery systems. (A) Zn-Vanadium flow battery at 80 mA/cm^2 , 20 mAh/cm^2 ; (B) Zn-Bromine flow battery at 40 mA/cm^2 , 20 mAh/cm^2 . The charge time is set as 15 mins.

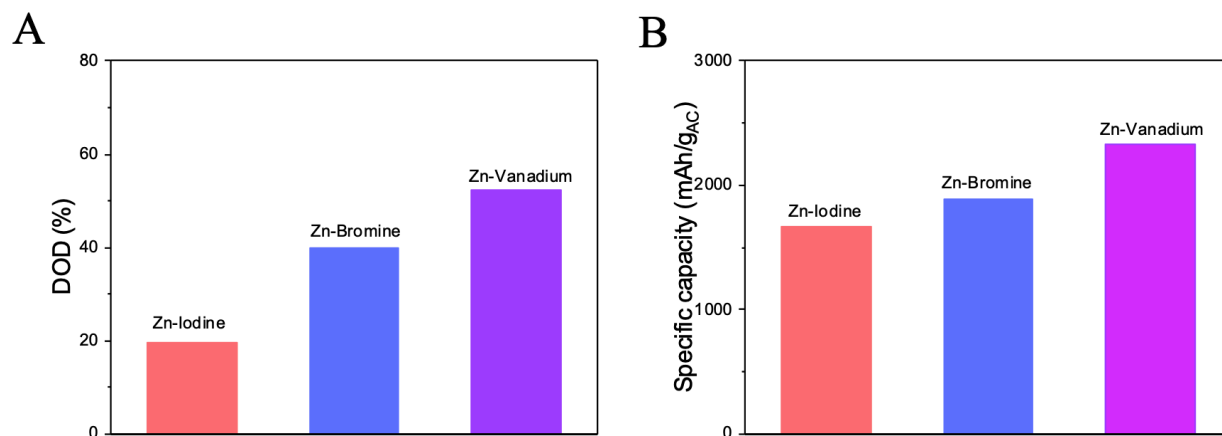


Figure S17. DOD and specific capacity of all flow battery systems in this work. (A) The discharge of depth (DOD) of the flow battery systems in this work. DOD was calculated based on the amount of catholyte. (B) Specific capacity of the flow battery systems in this work based on the active carbon (AC).