Electronic Supplementary Information for

Deposition of ZnO on Bismuth Species Towards Rechargeable Zn-Based Aqueous Battery

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1. Supplementary methods

Composite electrode mass ratio: For the Zn electrode without additives, Zn (96.4 wt%) and polyvinylidene fluoride (PVDF) (3.6 wt%) was utilized. For the Zn electrode with Super-P (SP) additive, Zn, (86.0 wt%) SP, (10.4 wt%) and polyvinylidene fluoride (PVDF) (3.6 wt%) was utilized. For the Zn electrode with ZnO additive, Zn, (86.0 wt%) ZnO, (10.4 wt%) and polyvinylidene fluoride (PVDF) (3.6 wt%) was utilized. For the Zn electrode with Bi_2O_3 additive, Zn, (86.0 wt%) Bi_2O_3, (6.2 wt%) and polyvinylidene fluoride (PVDF) (3.6 wt%) was utilized. For the Zn electrode with all the additives, Zn, (69.4 wt%) SP, (10.4 wt%) ZnO, (10.4 wt%) Bi_2O_3, (6.2 wt%) and polyvinylidene fluoride (PVDF) (3.6 wt%) was utilized. For the Ag electrode, Ag_2O (Alfa Aesar) (80.0 wt%), SP (12.0 wt%), and polyvinylidene fluoride (PVDF) (8.0 wt%) was utilized.

2. Supplementary Figures

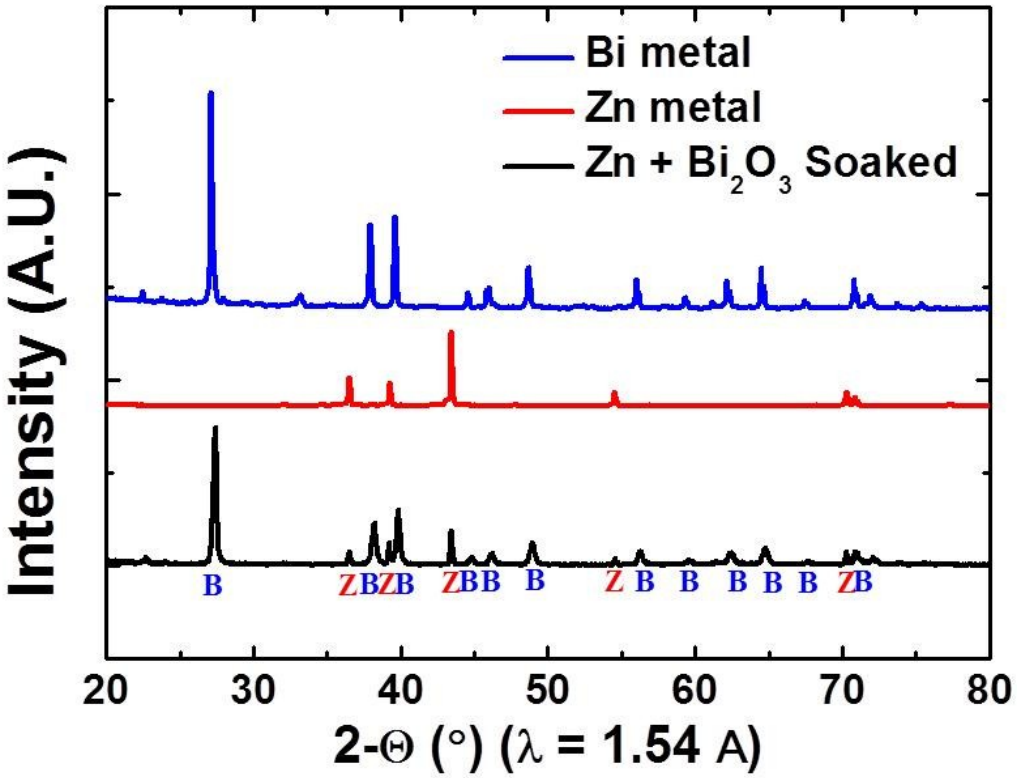

Figure S1: Same mass of Zn and Bi_2O_3 particles are mixed and soaked in the electrolyte. PXRD is obtained after soaking, the Bi_2O_3 reflections are completely vanished and only Zn and Bi metal reflections are present.

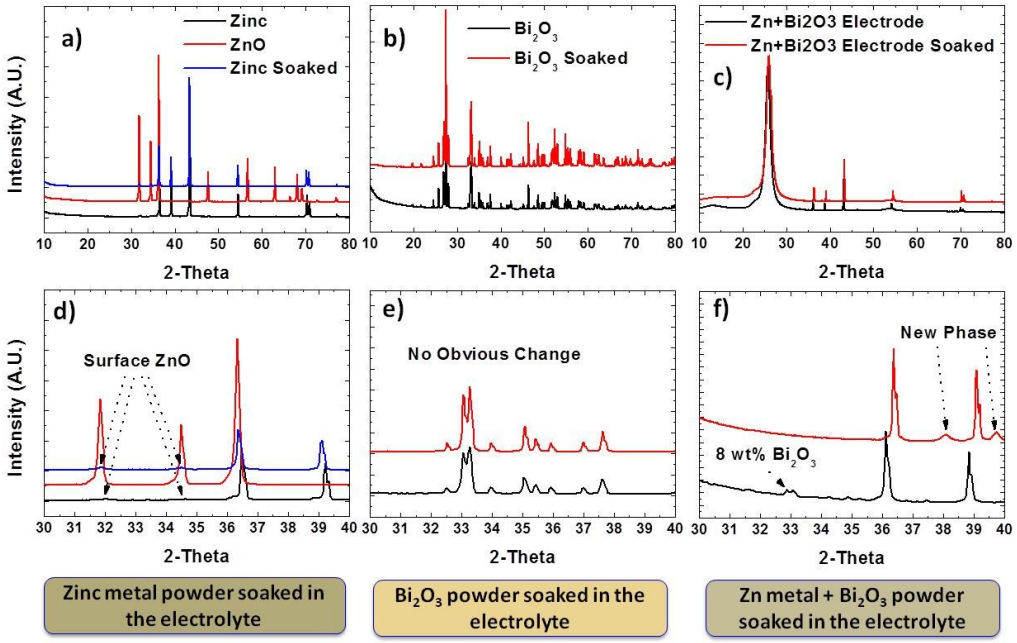

Figure S2: a) Wide range and d) narrow range of PXRD of Zn particle soaked in the electrolyte. b) Wide range and e) narrow range of PXRD of Bi_2O_3 particle soaked in the electrolyte. c) Wide range and f) narrow range of PXRD of Zn + Bi_2O_3 electrode soaked in the electrolyte. The electrode samples contain PVDF polymer binder.

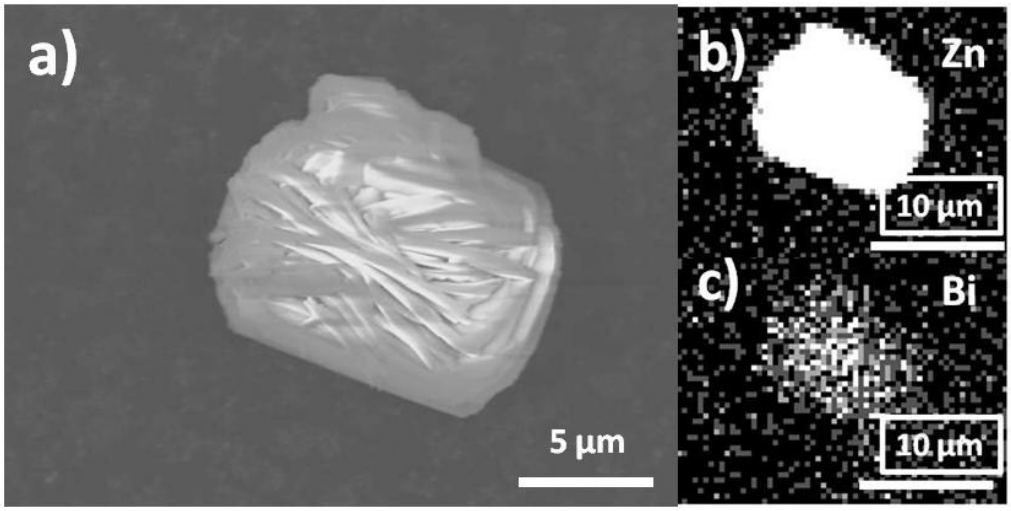

Figure S3: a) SEM image of the bismuth species after 50 cycles. b) EDS mapping of Zn $K\alpha_1$ and c) Bi $M\alpha_1$. The EDS mapping is performed on the same particle as the SEM image (a).

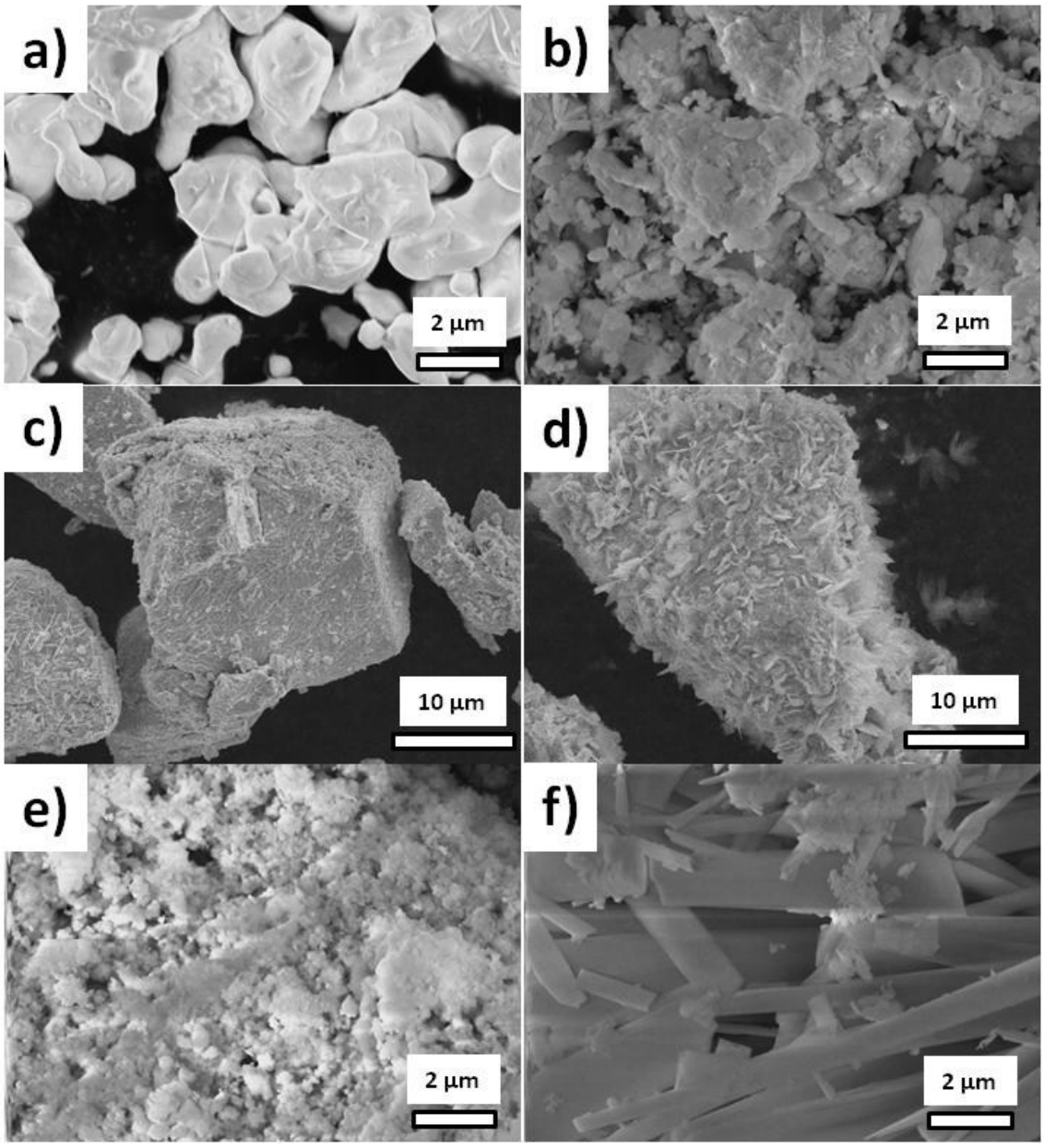

Figure S4: SEM images of the pristine a) Bi_2O_3, c) Bi, and e) NiO. SEM images of the b) Bi_2O_3 and d) Bi soaked in 6 M $Zn(NO_3)_2$ and f) NiO soaked in 0.5 M $Cu(SO_4)$. The respective particles in the respective solutions were soaked for overnight. The particles were collected by vacuum filtration and washed with copious amount of D.I. water. Then the samples were loaded onto the SEM holder using the carbon tape. The images were taken using 10 kV energy source.

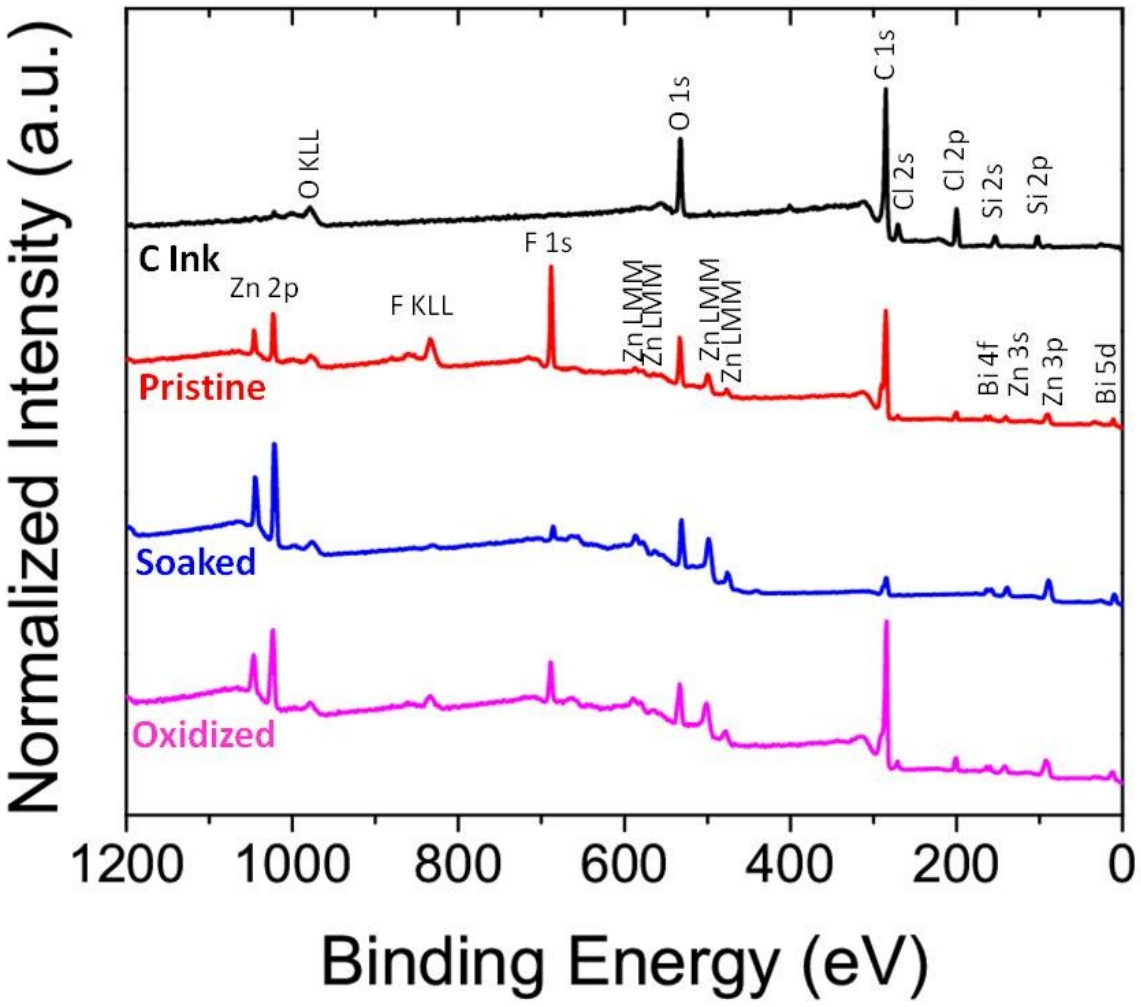

Figure S5: Survey scans of the Bi_2O_3 electrode. All the samples are in the form of electrodes printed on a PET substrate. The pristine sample is the Bi_2O_3 electrode after curing. The soaked sample is the Bi_2O_3 electrode after soaking in the electrolyte. The oxidized sample is the Bi_2O_3 electrode after first electrochemical oxidation or discharge.

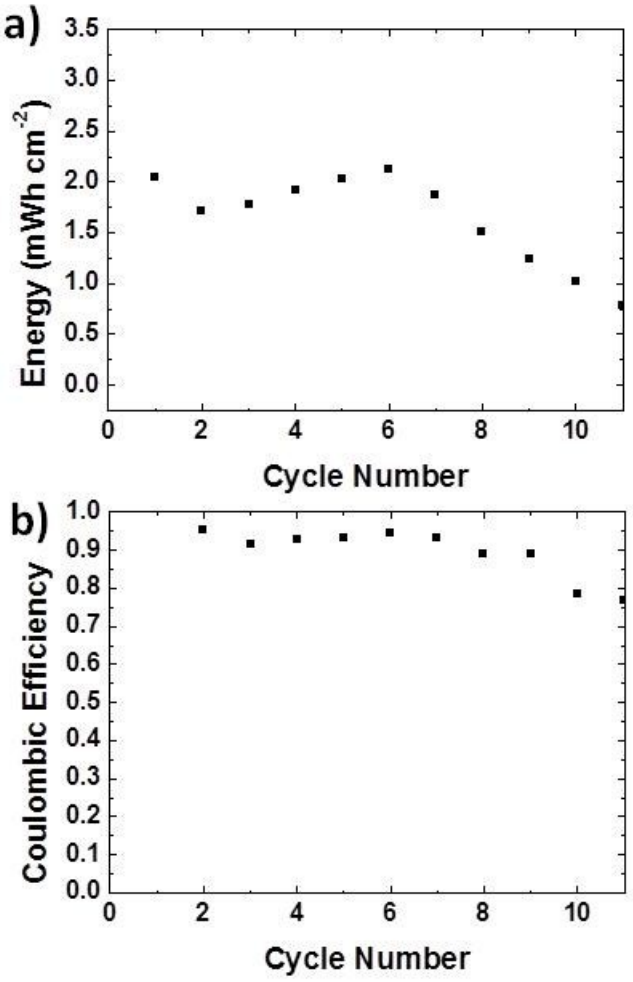

Figure S6: a) Energy density and b) coulombic efficiency of Zn-Ag full-cell battery performance with all the additives added in the Zn anode. The full-cell is cycled with current density at 4 mA cm^{-2} without a capacity limit.