***Supplementary Material***

**Eliminating Stubborn Insulated Deposition by Coordination Effect to Boost Zn Electrode Reversibility in Aqueous Electrolyte**

Yuzhuo Jianga, Xinyao Xiab, Siyi Qiana, Jing Zhangc, Pinxin Zhouc, Xuefang Gua,\*, Shu Tiana, Yijin Qiand,\*, Haoqing Jib, Jie Liua,\*, and Tao Qiana

a School of Chemistry and Chemical Engineering, Nantong University, Nantong 226019, China

b College of Energy, Key Laboratory of Advanced Carbon Materials and Wearable Energy Technologies of Jiangsu Province, Soochow University, Suzhou 215006, Jiangsu, China

c State Key Laboratory of Space Power-sources Technology, Shanghai Institute of Space Power-Sources, 2965 Dongchuan Road, Shanghai 200245, China.

d Deakin University, Institute for Frontier Materials, Waurn Ponds Campus, Locked Bag 20000, Geelong, Victoria 3220, Australia

E-mail: xuefang818@ntu.edu.cn, qianyij@deakin.edu.au and jliu93@ntu.edu.cn



**Figure S1.** Changes in UV-Vis of EDTA-2Na solution samples with and without Zn4(OH)6SO4·5H2O.



**Figure S2.** Scheme Caption CE of Zn||Cu cells with different mass ratio of EDTA-2Na in the control electrolyte cycled under 10 mA cm-2 and 5 mA h cm-2 conditions.

****

**Figure S3.** EIS plots of Zn||Cu cells with and without EDTA-2Na after various numbers of cycles.

****

**Figure S4.** Galvanostatic Zn plating/stripping in Zn||Zn symmetrical cells at 1 mA cm-2 and 1 mA h cm-2.

****

**Figure S5.** Galvanostatic Zn plating/stripping in Zn||Zn symmetrical cells at 2 mA cm-2 with different plating/stripping capacity.



**Figure S6.** CV curves of Zn||LiMnO4 battery in the control electrolyte at 0.5 mV s-1.



**Figure S7.** Typical charge/discharge curves for Zn||LiMnO4 cells in the control electrolyte with current density.

**Table S1.** The electrochemical performances of aqueous ZIBs using various additives.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Electrolyte | Cathode | Cathode cyclability | Current density | Refs |
| 1.26×10-8 M PbSO4 + 5% fumed silica + 1M Li2SO4 + 2M ZnSO4 | LiMn2O4 | 74.4% capacity retention after 300 cycles | 1 C | Mater. Today Energy 2017, 4, 34 |
| H3BO3 + 1M Li2SO4 + 1M ZnSO4 | LiMn2O4 | 78% capacity retention after 1000 cycles | 4 C | Chem. Eur. J. 2018, 24, 1667 |
| PAM/GO/EG | δ-MnO2 | 95.0% capacity retention after 100 cycles | 0.5 C | Front. Chem. 2020, 8, 603 |
| (NH4)6[Mo7O24]·4H2O + 1M ZnSO4 | NaV3O8·1.5H2O | 60% capacity retention after 500 cycles | 5 C | J. Mater. Chem. A 2021, 9, 7025 |
| Polyacrylamide + 0.1M MnSO4 | MnO2 | 87.2% capacity retention after 200 cycles | 0.8 C | Angew. Chem. 2019, 131, 15988-15994 |
| 5%EDTA-2Na + 1M ZnSO4 + 3M Li2SO4 | LiMn2O4 | 90.3% capacity retention after 150 cycles | 4 C | This work |