

Concentrated Electrolyte in Zinc Rechargeable Aqueous Batteries Studied by *in-operando* X-ray Diffraction

Bizualem Wakuma Olbasa¹, Chen-Jui Huang(黃貞睿)¹, Fikadu Wubatu Fenta¹, Shuo-Feng Chiu(邱碩峯)¹, Meng-Che Tsai(蔡孟哲)¹, Bikila Alemu Jote¹, Tamene Tadesse Beyene¹, Wei-Nien Su(蘇威年)^{*2}, Hongjie Dai(戴宏杰)^{*3} and Bing-Joe Hwang(黃炳照)^{*1,4,5}

1 Nano-Electrochemistry Laboratory, Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 106, Taiwan,

2 Nano-Electrochemistry Laboratory, Graduate Institute of Applied Science and Technology,

3Department of Chemistry, Stanford University, Stanford, California 94305, United States,

4National Synchrotron Radiation Research Center, Hsin-Chu 30076, Taiwan,

5Applied Research Center for Thin-Film Metallic Glass, National Taiwan University of Science and Technology, Taipei 106, Taiwan

Abstract

Recently, zinc (Zn) has become a promising ideal anode material for rechargeable aqueous batteries. However, vigorous growth of zinc dendrite during plating/stripping, low CE of the cell, and short cycle life hinder its practical applications in rechargeable batteries. Here in, we report a cost-effective concentrated electrolyte (4.2 M ZnSO₄ + 0.1 M MnSO₄) in which improves cycling stability of zinc with an average coulombic efficiency (ACE) of ~99.21% cycled for more than 1000 hours at a current density of 0.2 mA/cm² within Zn||Cu cells. However, frequently used diluted electrolyte (2 M ZnSO₄ + 0.1 M MnSO₄) only produces ACE of ~97.54% and relatively short cell cycle life. We further used *in-operando* XRD to study the effect of concentrated electrolyte on the intercalation/deintercalation of Zn²⁺ in MnO₂ cathode material. The developed concentrated electrolyte shows the synergetic effects of enhanced solvation/desolvation process, electrostatic shielding and Le Chateier's principle. Consequently, significant suppression of Zn dendrite formation and dissolution of MnO₂ from cathode electrode are obtained. A highly stable and reversible Zn||MnO₂ cell retaining about 88.4% of capacity retention is achieved after more than 1200 cycles under 3 C-rate and 83.7% of capacity retention after cycled for more than 600 cycles under 1 C-rate.

Keywords- Zinc; MnO₂; *In-operando* XRD; ZIB