Electronic Supplementary Information

Stabilizing High-Voltage LiCoO2 Cathode in Aqueous Electrolyte with Interphase-forming Additive

Fei Wang,[a] Yuxiao Lin,[b] Liumin Suo,[c] Xiulin Fan, [a] Tao Gao, [a] Chongyin Yang, [a] Fudong Han, [a] Yue Qi, [b] Kang Xu [d] and Chunsheng Wang[a]*

a. Department of Chemical and Biomolecular Engineering, University of Maryland, College Park , MD 20742 , USA
b. Department of Chemical Engineering and Materials Science, Michigan State University, East Lansing, MI 48824, USA
c. Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 02138, USA
d. Electrochemistry Branch, Sensor and Electron Devices Directorate, Power and Energy Division, U.S. Army Research Laboratory, Adelphi, MD 20783, USA.

E-mail: cswang@umd.edu

Keywords: aqueous lithium-ion battery, $LiCoO_2$, high voltage, high energy density, water-in-salt electrolyte

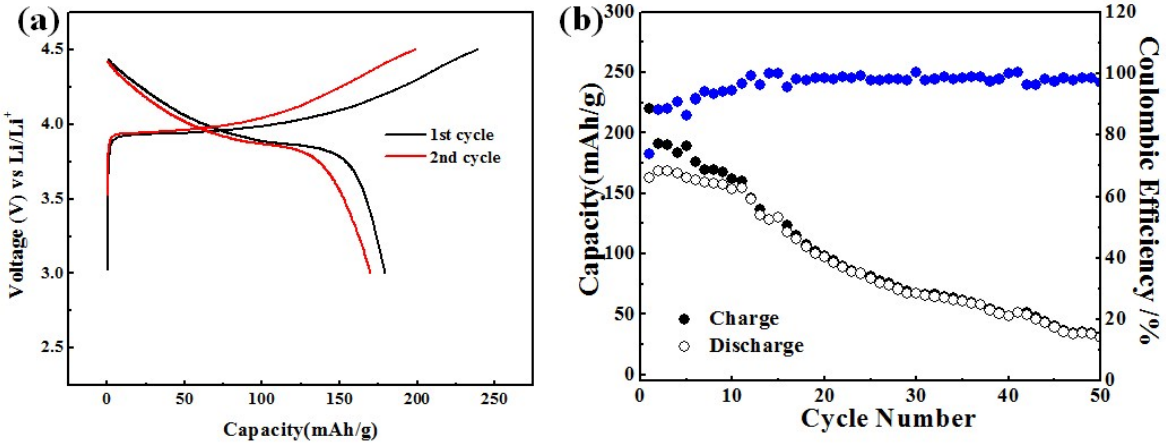

Fig. S1 (a) Typical voltage profile during first two charge and discharge cycles and (b) the cycling stability of $LiCoO_2$ in organic electrolyte (1M $LiPF_6$ in EC: DMC, 1:1 by volume) between 3.0 V and 4.5 V (vs Li^+/Li) at the constant current of 0.2 C with lithium as counter electrode and reference electrode.

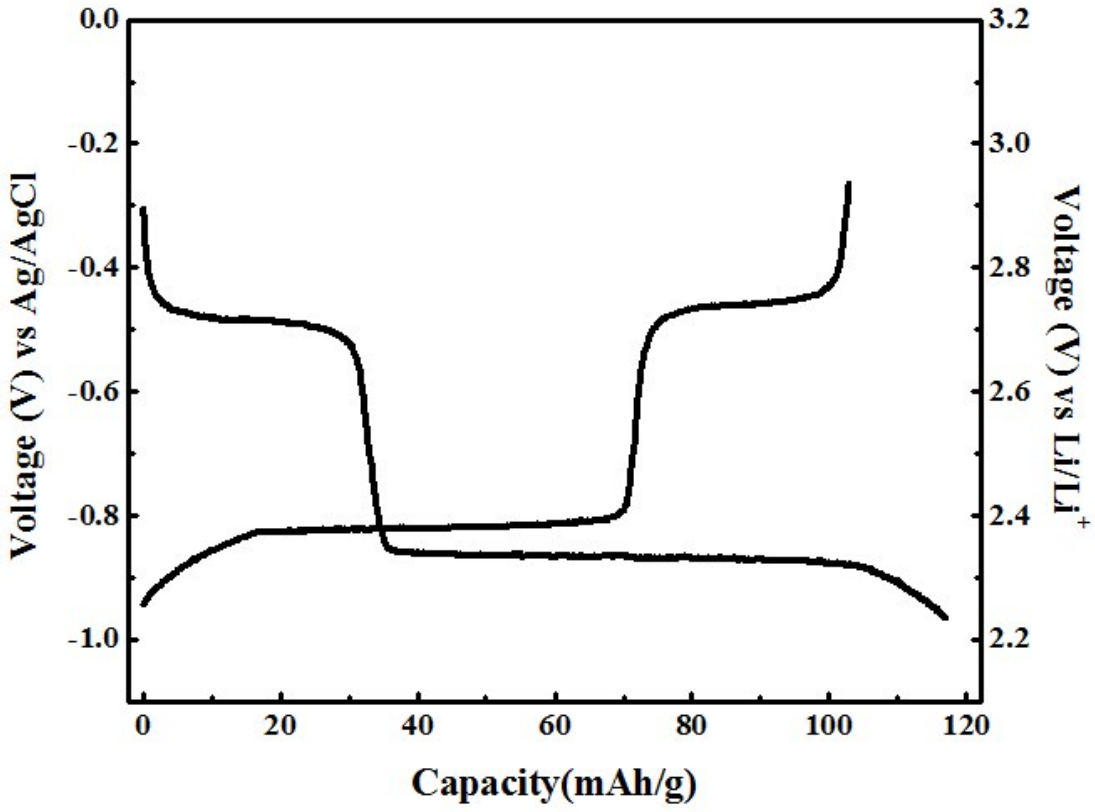

Fig. S2 The typical voltage profile of Mo_6S_8 in water-in-salt electrolyte at the constant current of 0.2 C with Pt as counter electrode and the Ag/AgCl as the reference electrode.

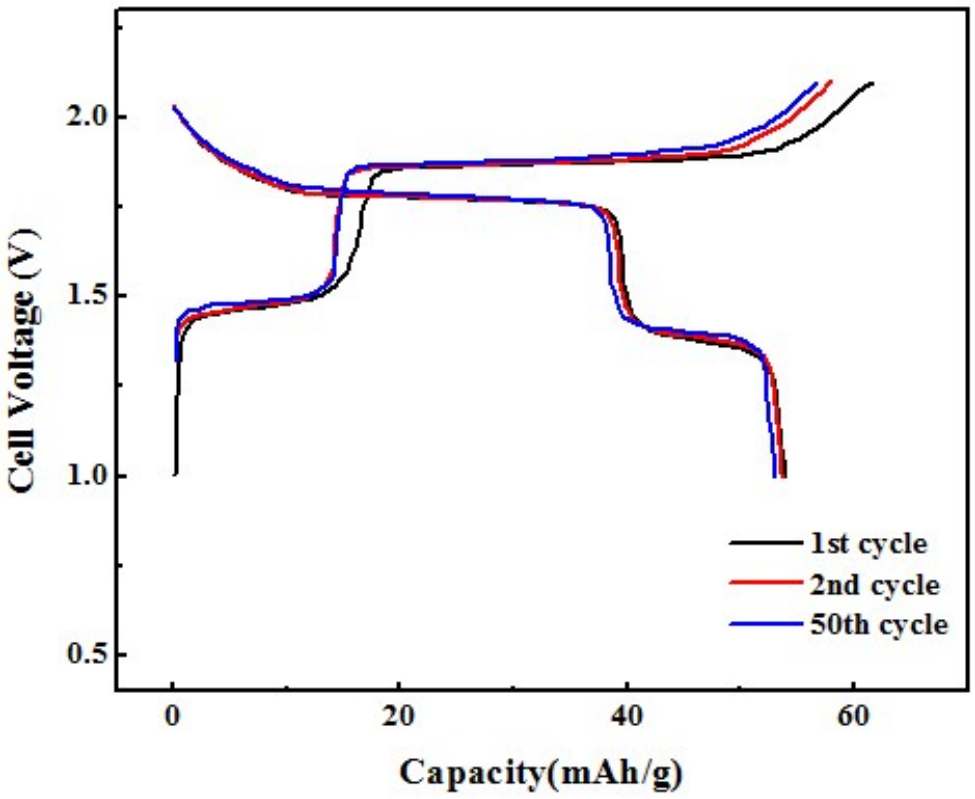

Fig. S3 The typical voltage profile of the full aqueous Li-ion cell employing Mo_6S_8 and LVLCO cycled between 1.0 V and 2.1 V in 21m LiTFSI electrolyte at constant current of 0.5 C.

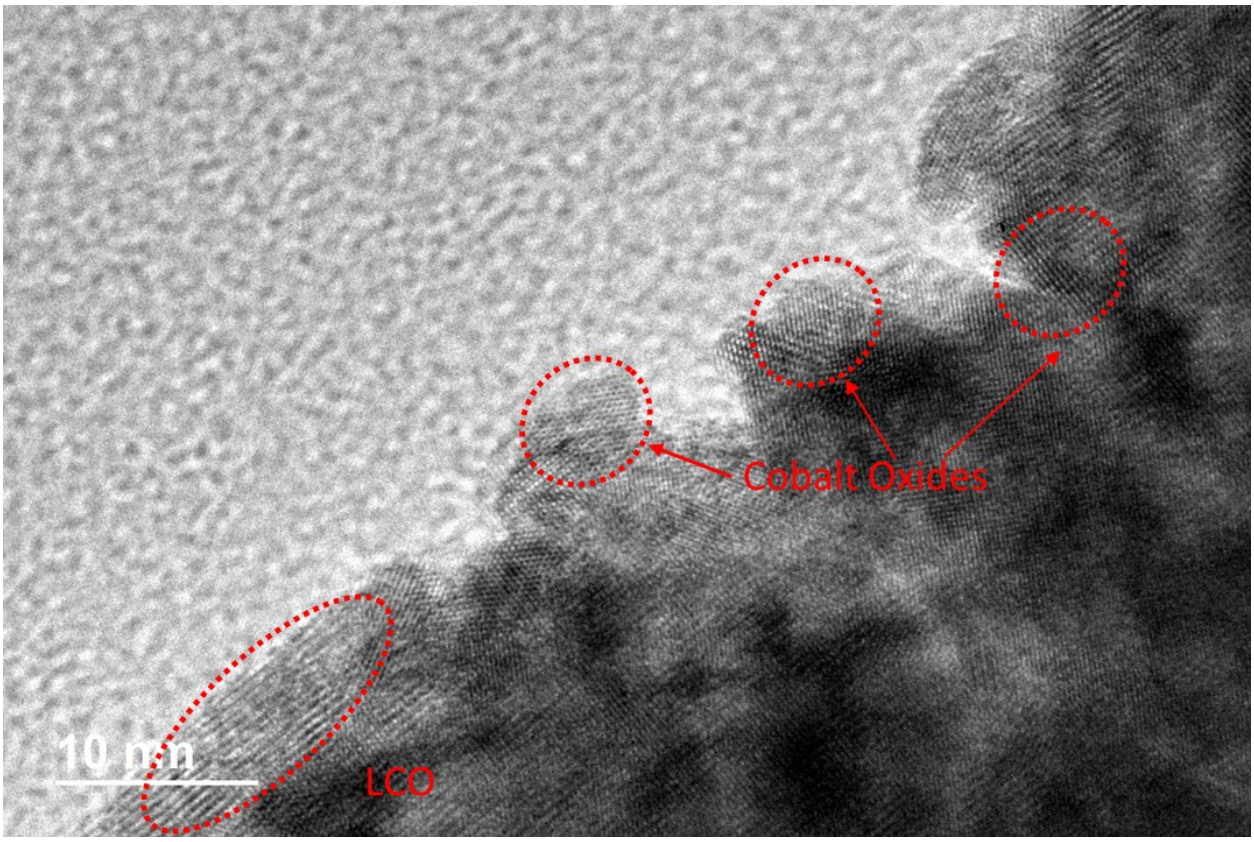

Fig. S4 The TEM image for the HVLCO electrodes after cycling in the 21m LiTFSI electrolyte.

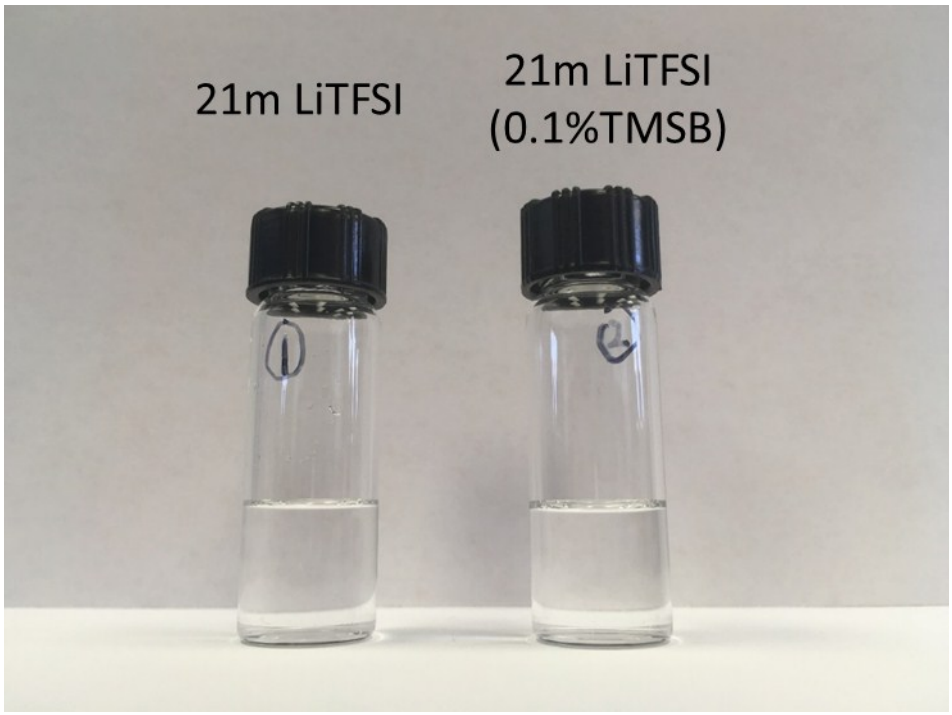

Fig. S5 The photograph of the 21m LiTFSI electrolyte and 21m LiTFSI-0.1wt% TMSB electrolyte.

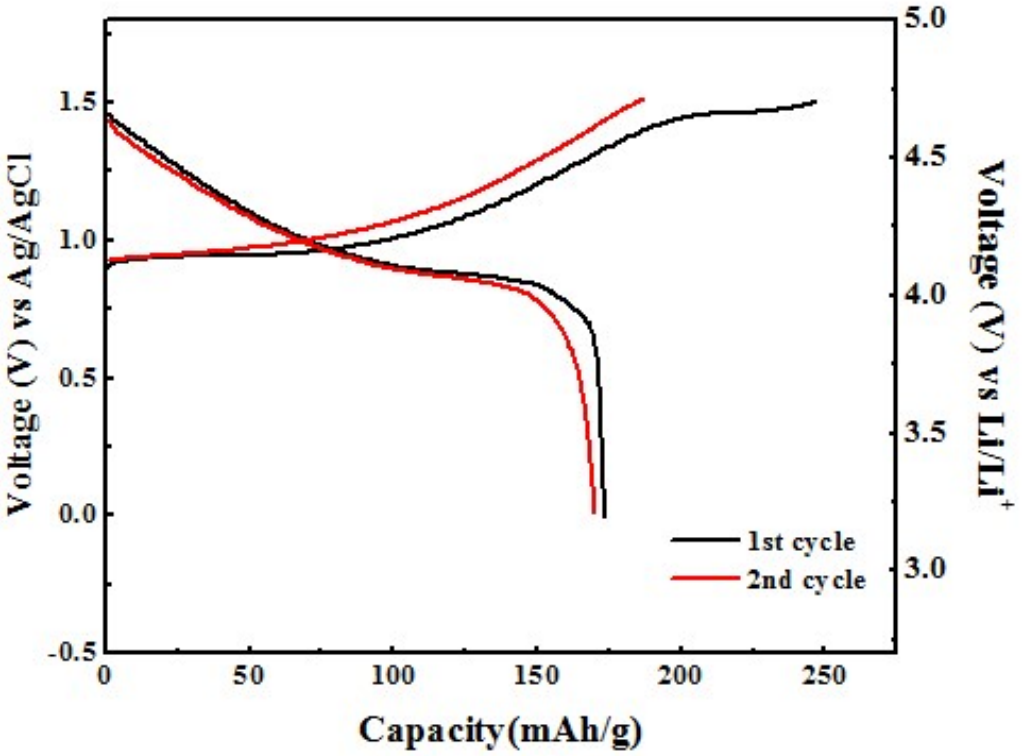

Fig. S6 The typical potential profile of $LiCoO_2$ between 1.5 V to 0.0 V (vs Ag/AgCl) in 21m LiTFSI electrolyte-0.1wt % TMSB at 0.2 C measured in a three-electrode cell using active carbon as a counter and Ag/AgCl as a reference electrode.

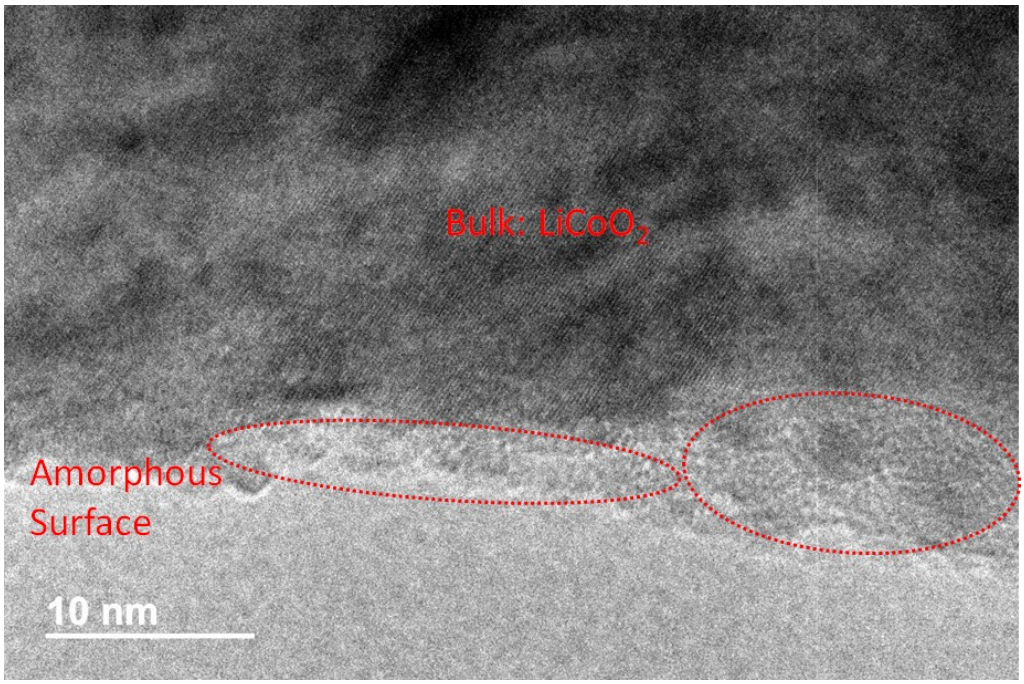

Fig. S7 TEM image for the HVLCO electrodes after the first cycle in 21m LiTFSI-0.1wt% TMSB electrolytes.

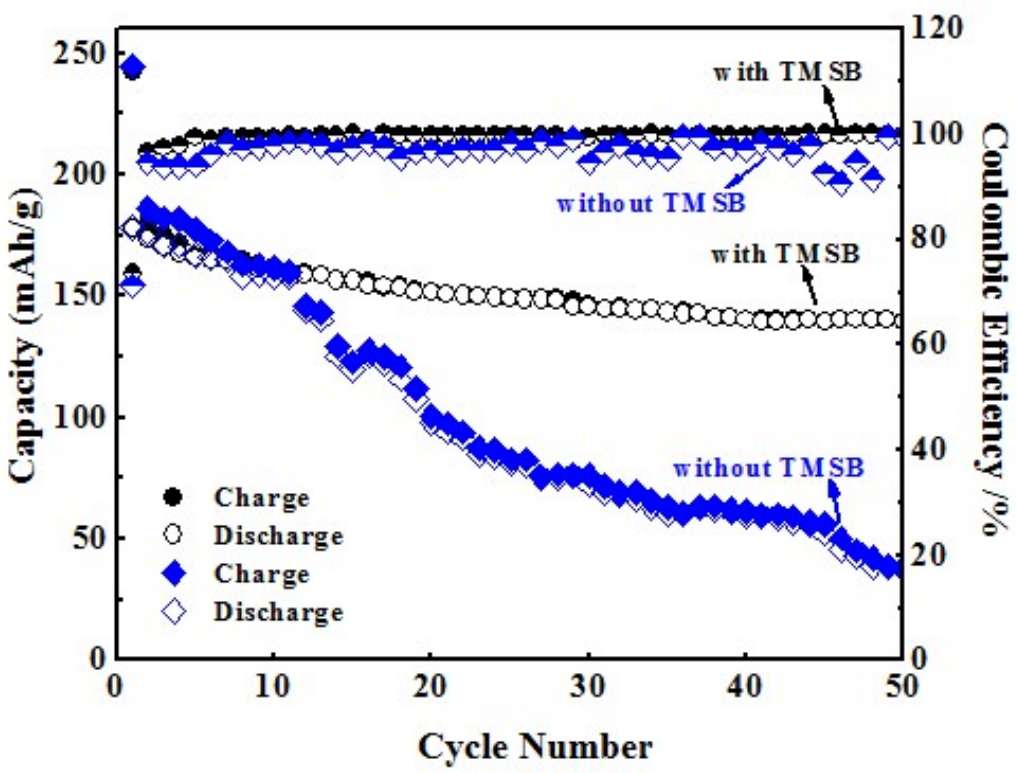

Fig. S8 The comparison of the cycling stability for the $LiCoO_2$ in the 0.1wt % TMSB added electrolyte and the TMSB-free electrolyte at 0.5 C rate measured in the three-electrode cells using active carbon as counter and Ag/AgCl as reference electrodes.

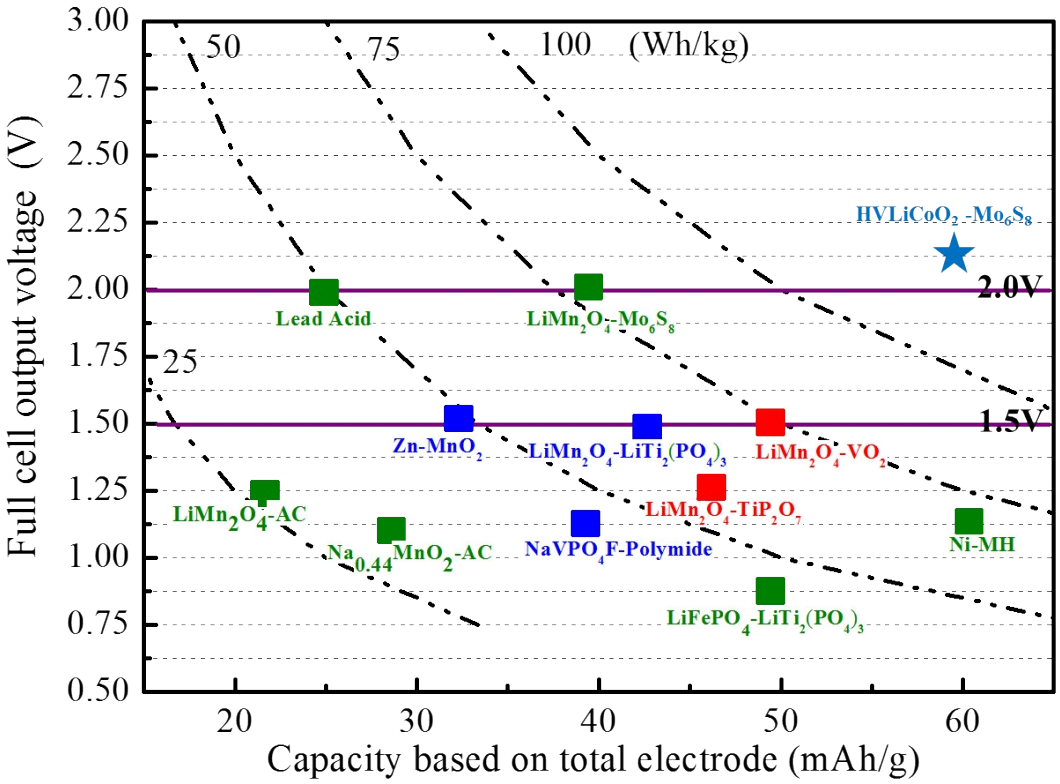

Fig. S9 Performances comparison for aqueous batteries based on various electrochemical couples.

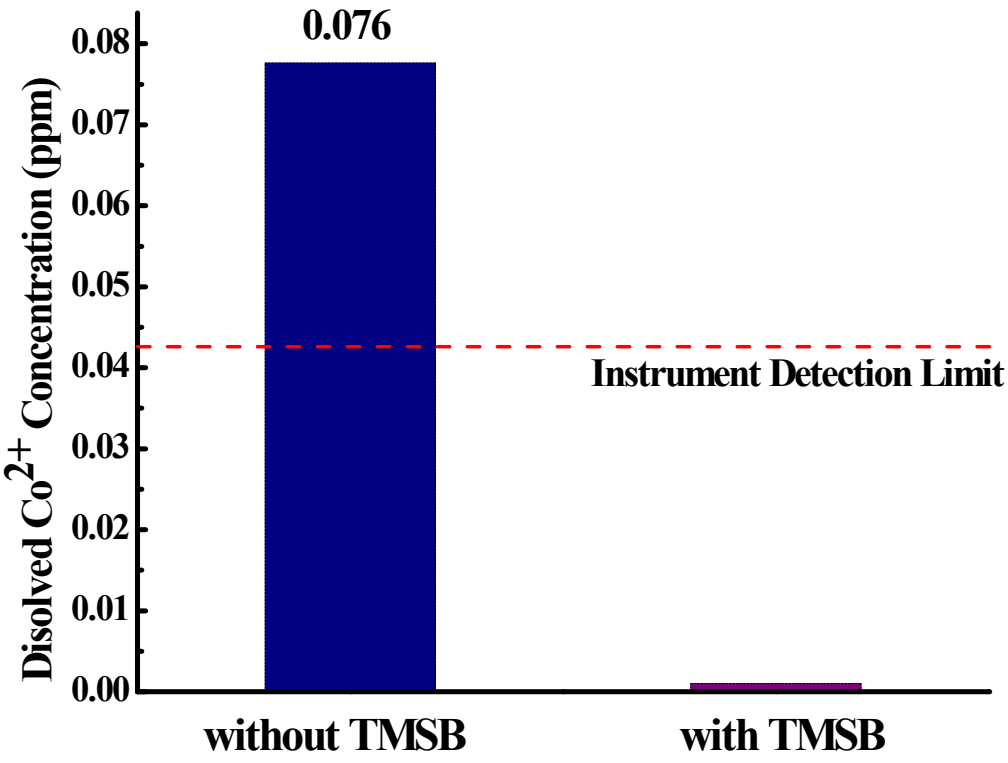

Fig. S10 The comparison for the dissolved Co concentrations in TMSB-added elecrolytes after 100 cycles and TMSB-free electrolyte after 50 cycles.

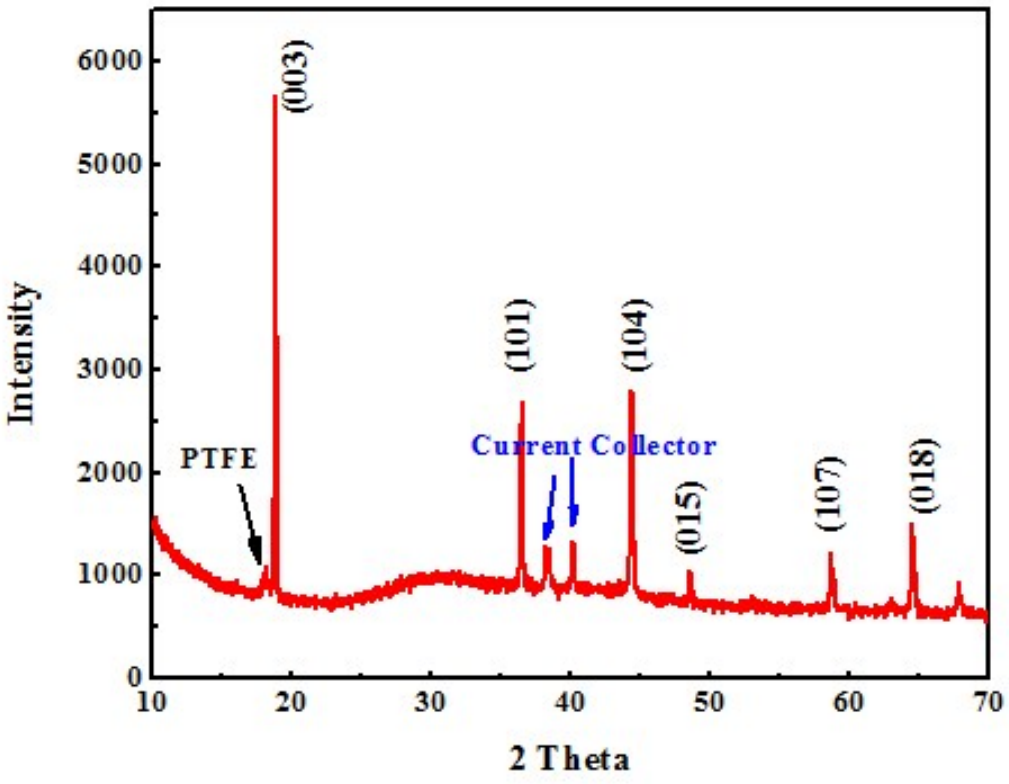

Fig. S11 The X-ray Diffraction (XRD) patterns for HVLCO electrodes after cycling in 21m LiTFSI-0.1%wt TMSB electrolytes for 100 cycles.

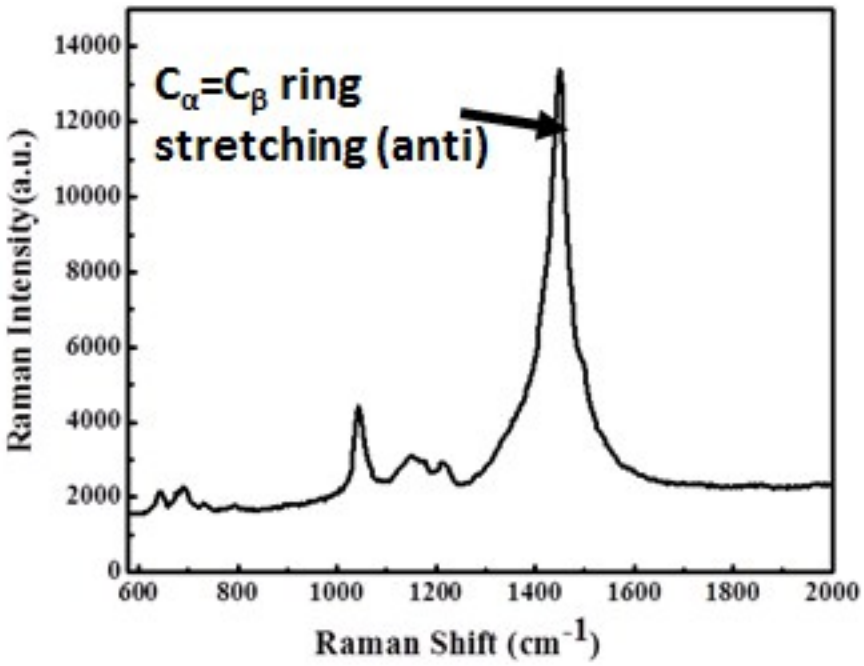

Fig. S12 Raman spectroscopy of HVLCO in 21m LiTFSI with thiophene additive. The electrochemical polymerization of thiophene was achieved during the charge process.