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Supporting Information

A multifunctional phosphite-containing electrolyte for 5 V-class $LiNi_{0.5}Mn_{1.5}O_4$ cathodes with superior electrochemical performance

*Young-Min Song, Jung-Gu Han, Soojin Park, Kyu Tae Lee and Nam-Soon Choi**

School of Green Energy, Ulsan National Institute of Science and Technology (UNIST), Ulsan 689-798, Republic of Korea
E-mail: nschoi@unist.ac.kr

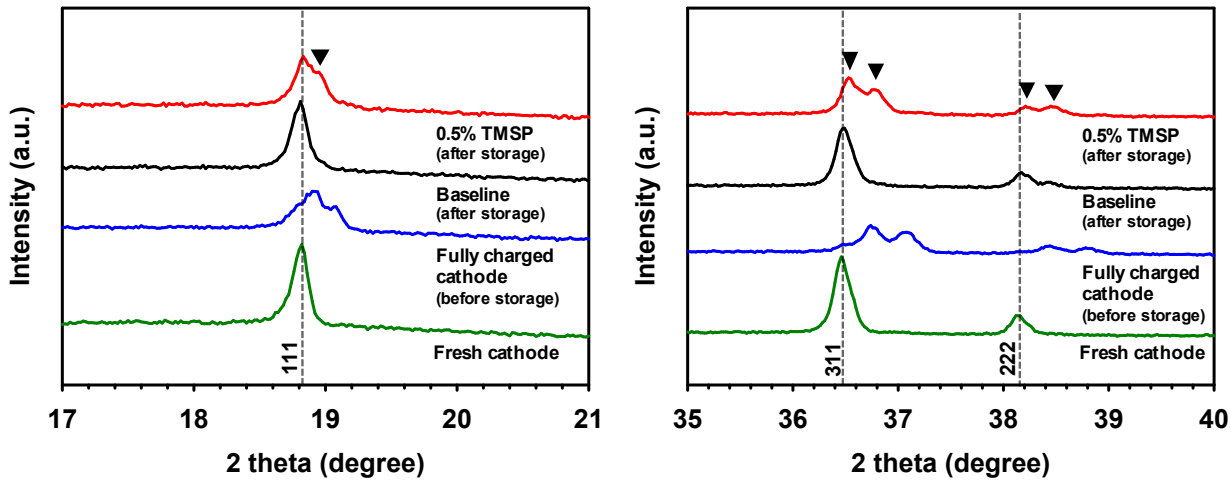

Fig. S1. XRD patterns of fully charged $Li_xNi_{0.5}Mn_{1.5}O_4$ cathode before and after stored at 60°C for 1day. The XRD pattern of fresh $LiNi_{0.5}Mn_{1.5}O_4$ cathode is displayed for comparison. The XRD data reveals that a fully charged cathode stored in the TMSP-added electrolyte at 60°C experiences less re-lithiation (self-discharge).

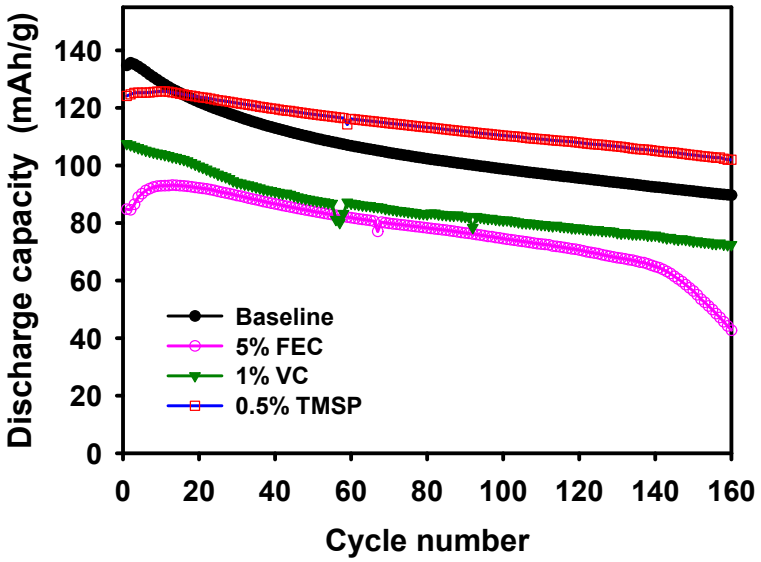

Fig. S2. Comparison of cycling stability of $LiNi_{0.5}Mn_{1.5}O_4$ cathodes with various electrolytes at 60°C at a current density of 60 mA/g.

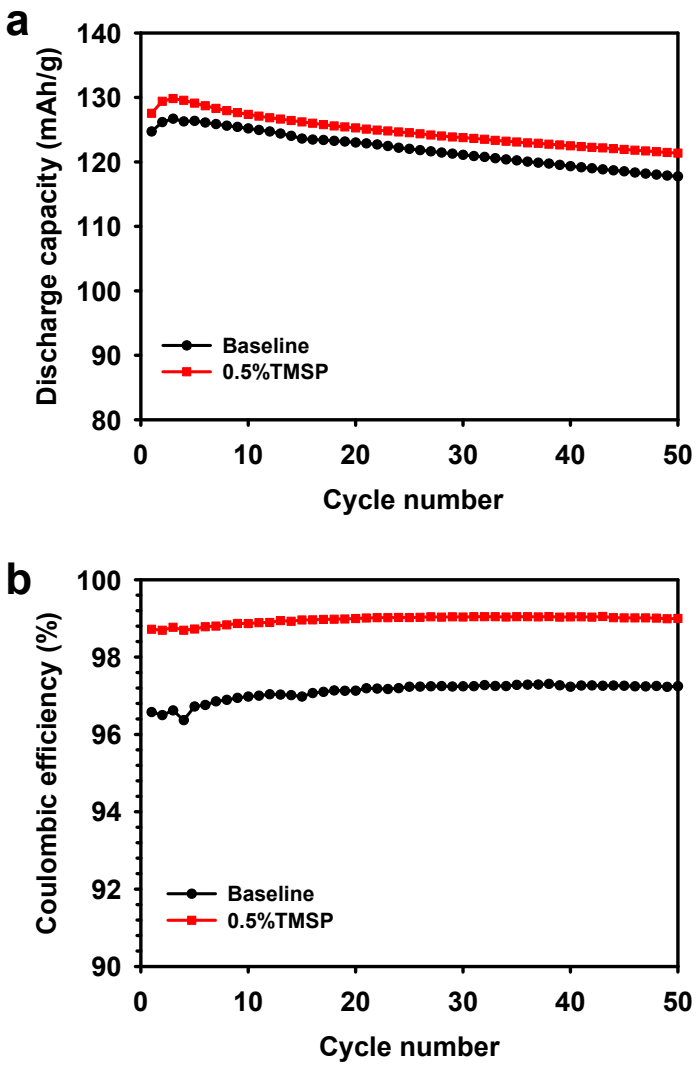

Fig. S3. (a) Discharge capacity, (b) Coulombic efficiency of the $LiNi_{0.5}Mn_{1.5}O_4$ cathodes in baseline and TMSP-added electrolytes at 30°C at a current density of 60 mA/g.

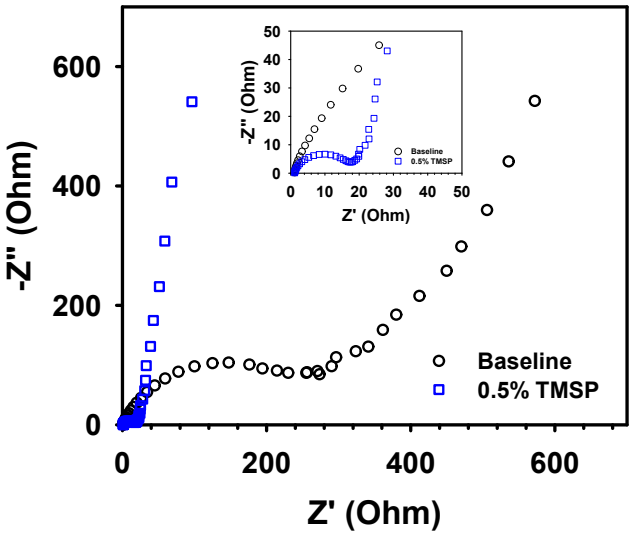

Fig. S4. EIS spectra for $LiNi_{0.5}Mn_{1.5}O_4$ cathodes precycled in baseline or 0.5% TMSP-added electrolyte.

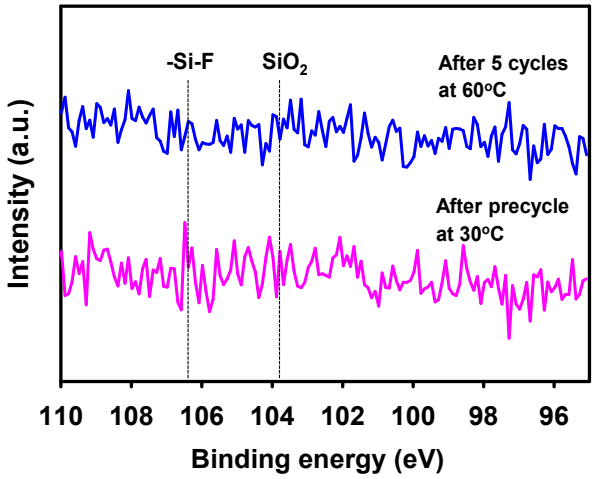

Fig. S5. Si 2p XPS spectra of the $LiNi_{0.5}Mn_{1.5}O_4$ cathodes cycled in TMSP-added electrolyte.

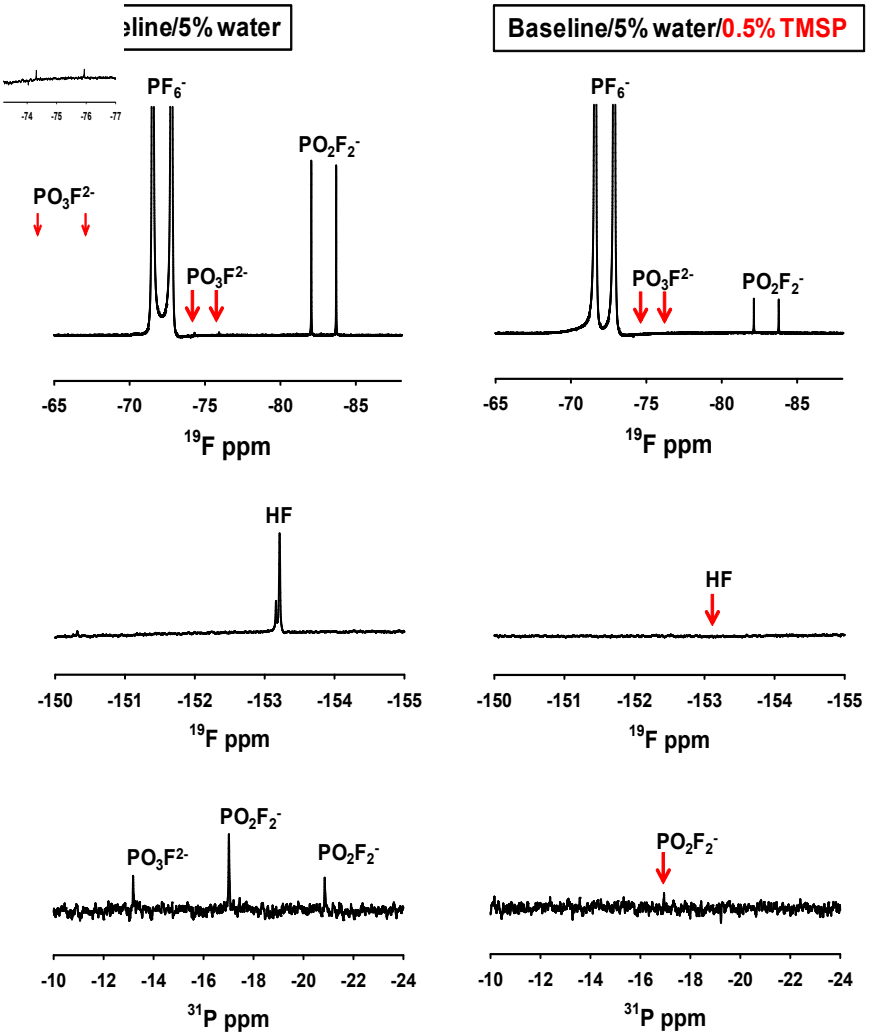

Fig. S6. ^{19}F and ^{31}P NMR spectra of the electrolyte with and without 0.5wt% TMSP after hydrolysis tests at room temperature. HF was completely removed from the electrolyte in presence of TMSP.

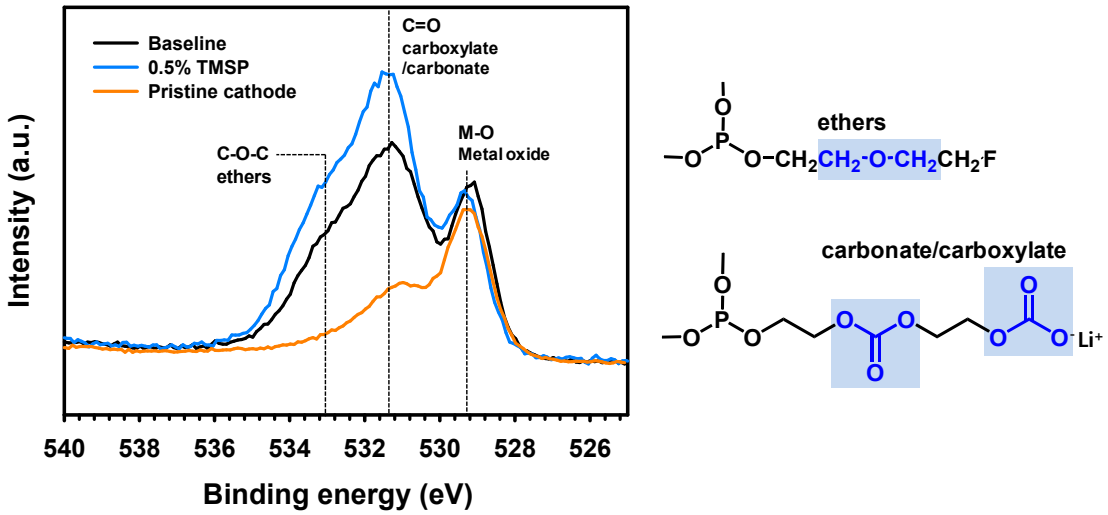

Fig. S7. O 1s XPS spectra of the $LiNi_{0.5}Mn_{1.5}O_4$ cathodes before and after 5 cycles in the baseline and TMSP-added electrolytes at 60°C.

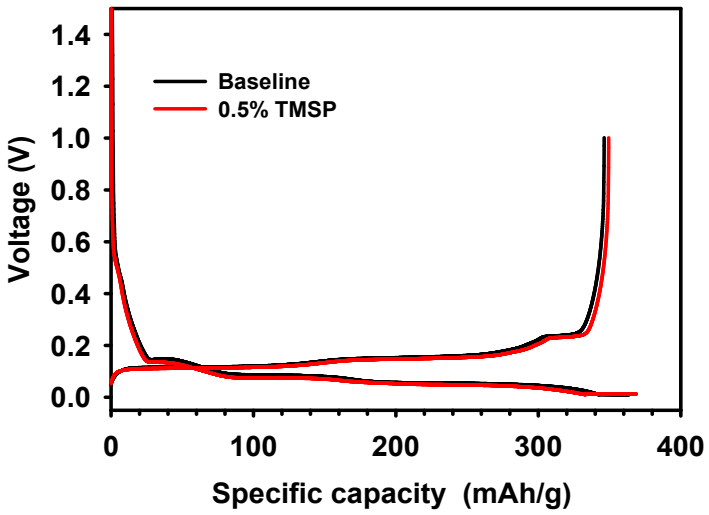

Fig. S8. Voltage profiles of the graphite anode with or without 0.5% TMSP for a current density of 35 mA/g (electrode capacity = 2.55 mAh/cm^2) during precycling.

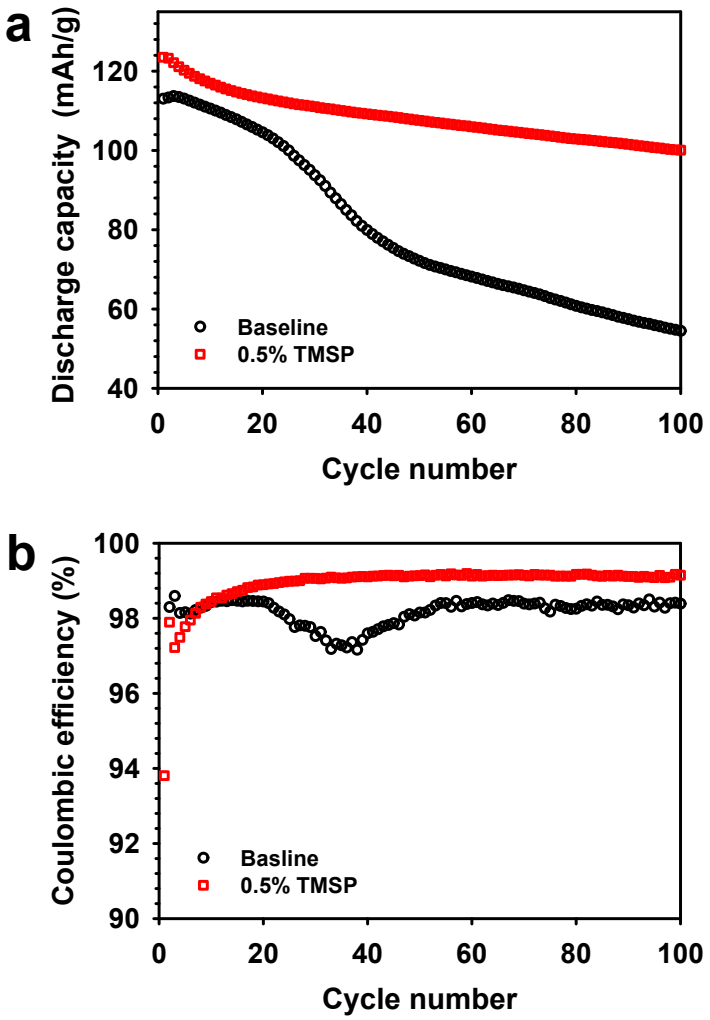

Fig. S9. (a) Discharge capacity, (b) Coulombic efficiency of graphite/$LiNi_{0.5}Mn_{1.5}O_4$ full cells in baseline and TMSP-added electrolytes at 45◦C at a rate of C/2.

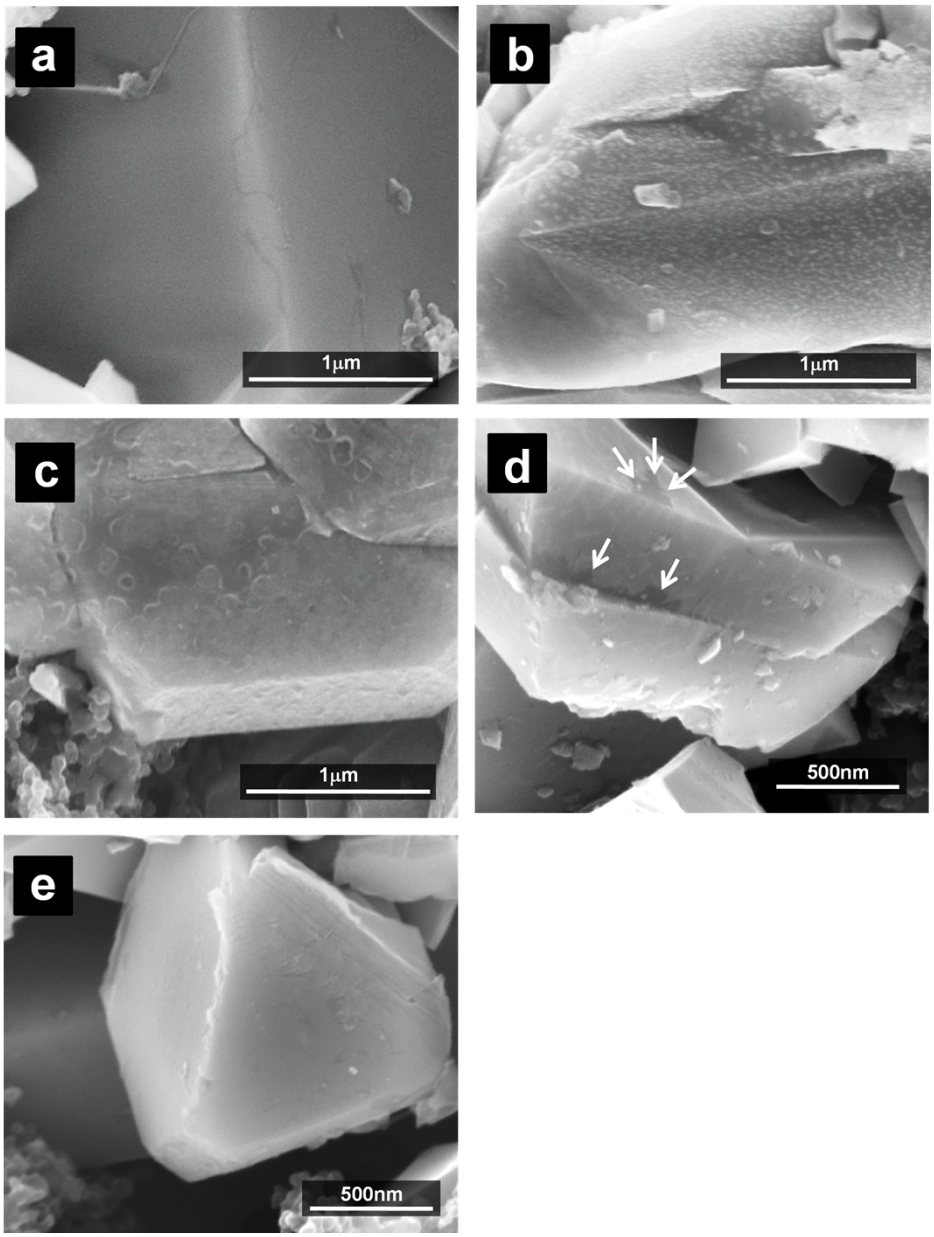

Fig. S10. FE-SEM images of the $LiNi_{0.5}Mn_{1.5}O_4$ cathodes after 5 cycles at 60°C: (a) pristine cathode, (b) baseline electrolyte, (c) 1% VC-added, (d) 5% FEC-added, and (e) 0.5% TMSP-added.

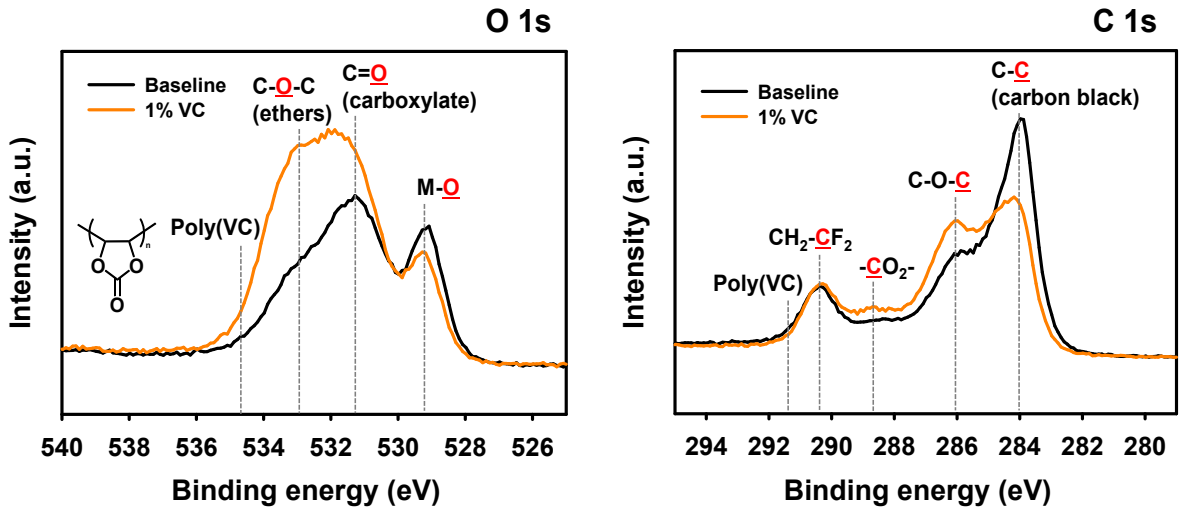

Fig. S11. O 1s and C 1s XPS spectra of the $LiNi_{0.5}Mn_{1.5}O_4$ cathodes cycled in baseline and VC-added electrolytes during 5 cycles at 60°C.

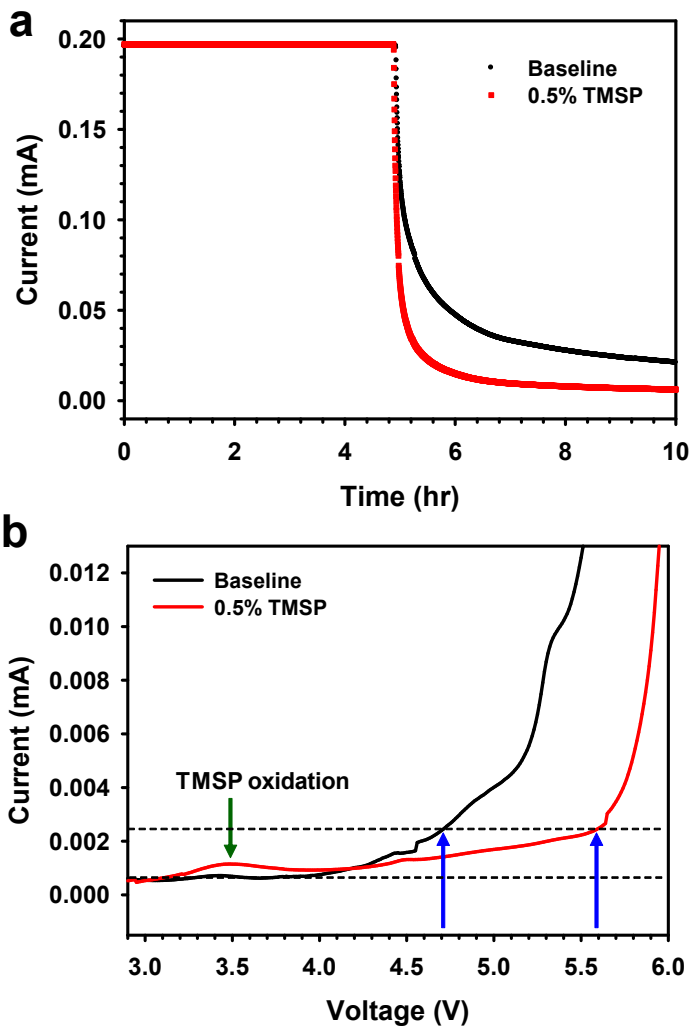

Fig. S12. (a) Potentiostatic profiles of Li/$LiNi_{0.5}Mn_{1.5}O_4$ half cells maintained at charging voltage of 5.0 V after charging up to 5.0 V at 30°C. (b) Linear sweep voltammetry of electrolytes with and without 0.5% TMSP additive. Stainless steel was used as a working electrode and the scan rate was 1 mV/s.