

Supporting Information

Enhancing Cycling Stability of Li-Rich Mn-Based Cathode Materials via Cyano Functional Additive

Dongwei Zhou^a, Zhanlin Yang^b, Shihao Wang^a, Guiyang Gao^a, Jie Zhu^a, Chengkun Zhang^a, Saichao Li^a, Baisheng Sa^b, Jie Lin^{*,a}, Dong-Liang Peng^{*,a}, Qingshui Xie^{*,a}

^a State Key Laboratory of Physical Chemistry of Solid Surface, Fujian Key Laboratory of Surface and Interface Engineering for High Performance Materials, College of Materials, Xiamen University, Xiamen 361005, China.

^b College of Materials Science and Engineering, Fuzhou University, Fuzhou 350108, China.

* Corresponding authors

E-mail addresses: linjie@xmu.edu.cn (J. Lin), dlpeng@xmu.edu.cn (D.-L. Peng), xieqsh@xmu.edu.cn (Q. Xie).

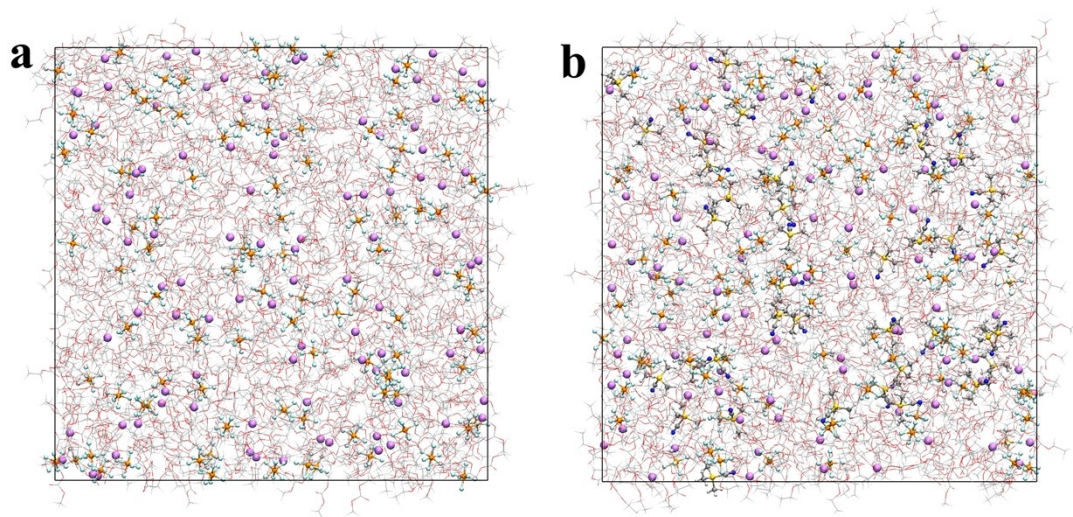


Fig. S1. MD simulation snapshots of the (a) baseline and (b) TMS-containing electrolytes.

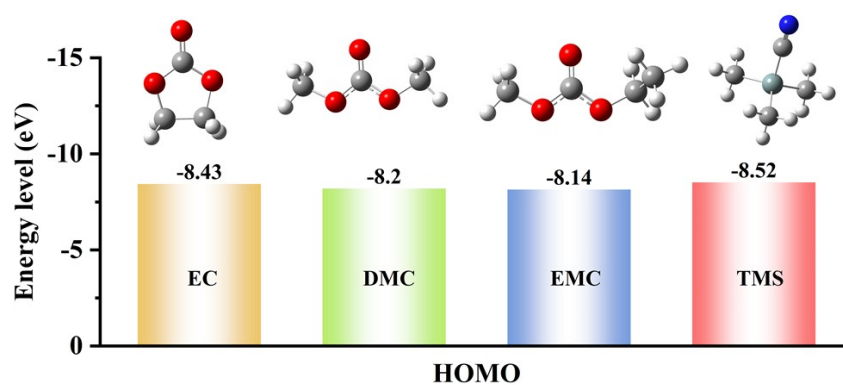


Fig. S2. Theoretical calculations of HOMO values for various solvents and additive.

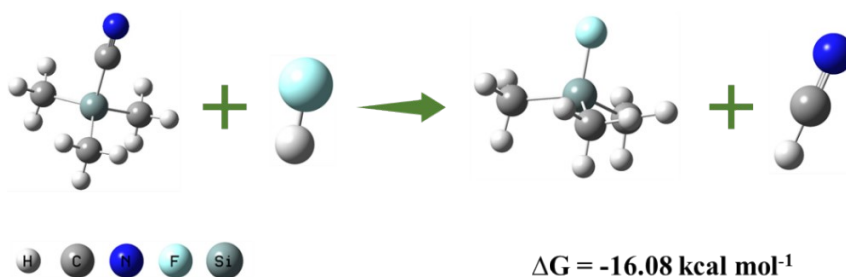


Fig. S3. The Gibbs free energy (ΔG) of TMS reacting with HF.

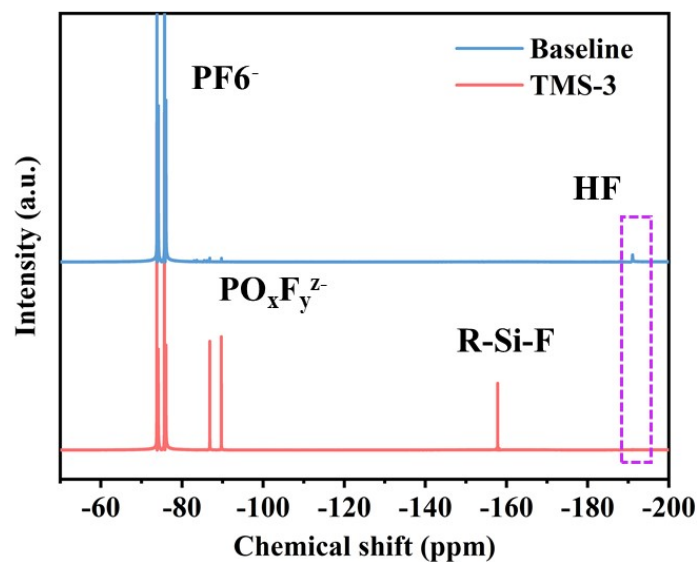


Fig. S4. ^{19}F NMR spectra of the baseline and TMS-containing electrolytes.

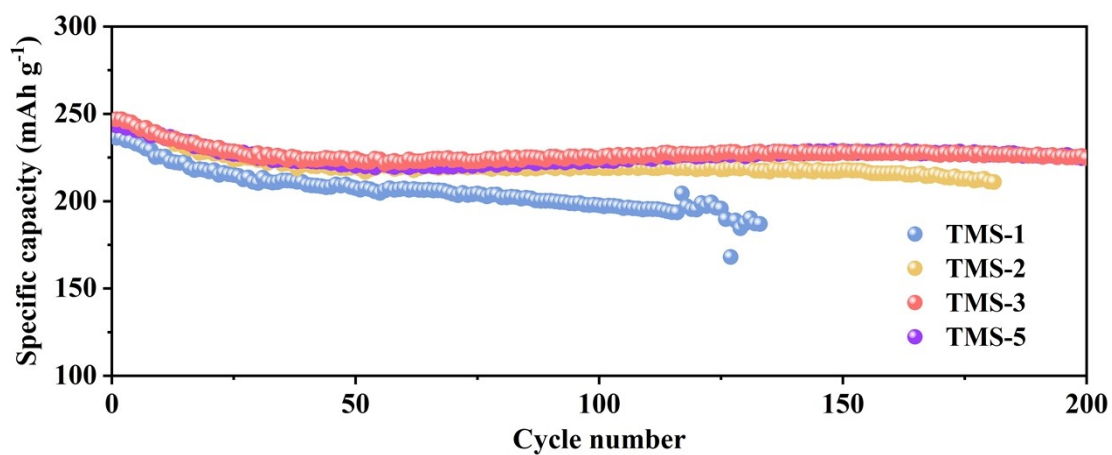


Fig. S5. Cycling performance of Li||LRM batteries in the electrolytes with different contents of TMS at 1 C.

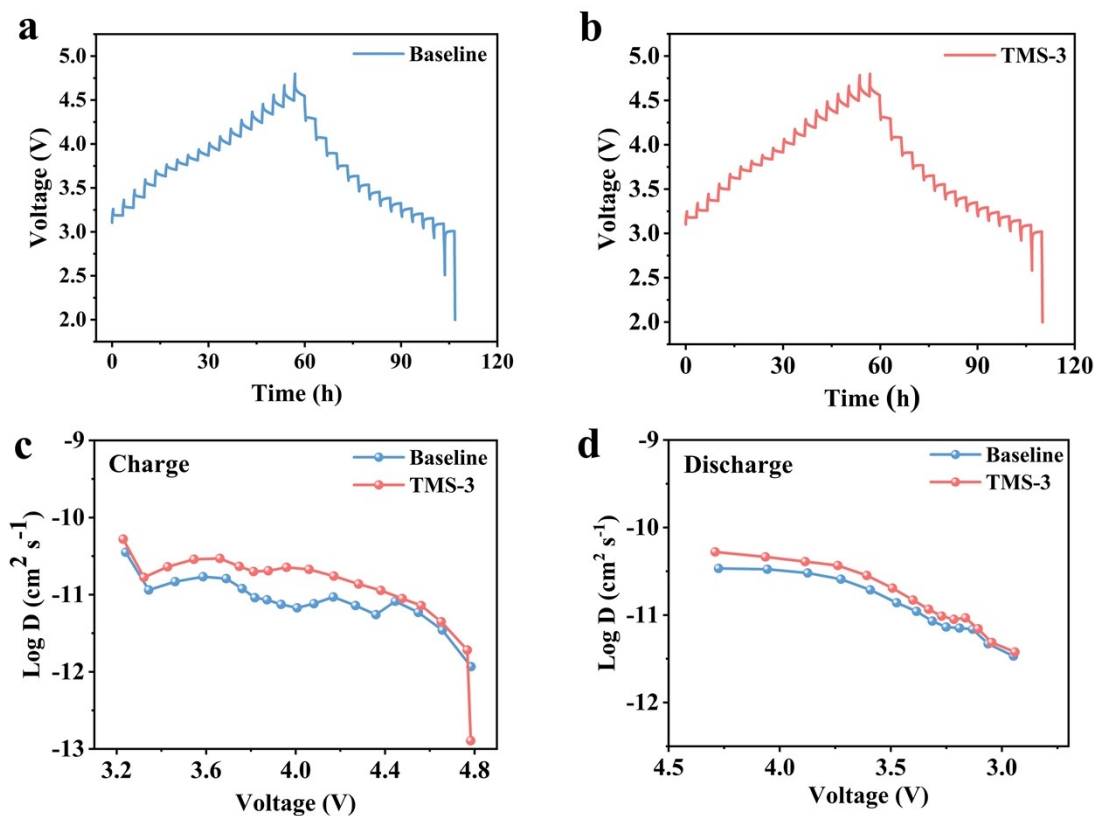


Fig. S6. (a, b) GITT curves of Li||LRM batteries (a) without and (b) with TMS additive. (c, d) The calculated Li^+ diffusion coefficient during (c) charge and (d) discharge process.

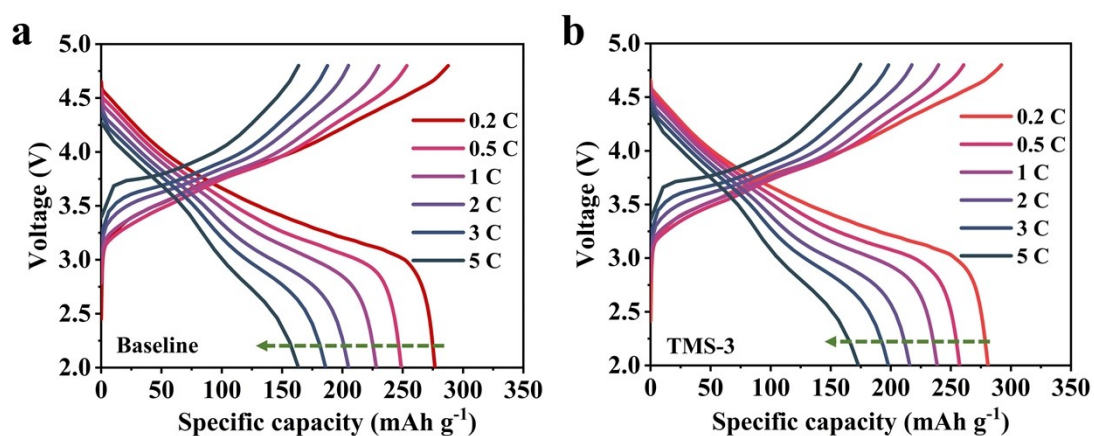


Fig. S7. The charge and discharge curves of Li||LRM batteries (a) without and (b) with TMS additive in the rate test.

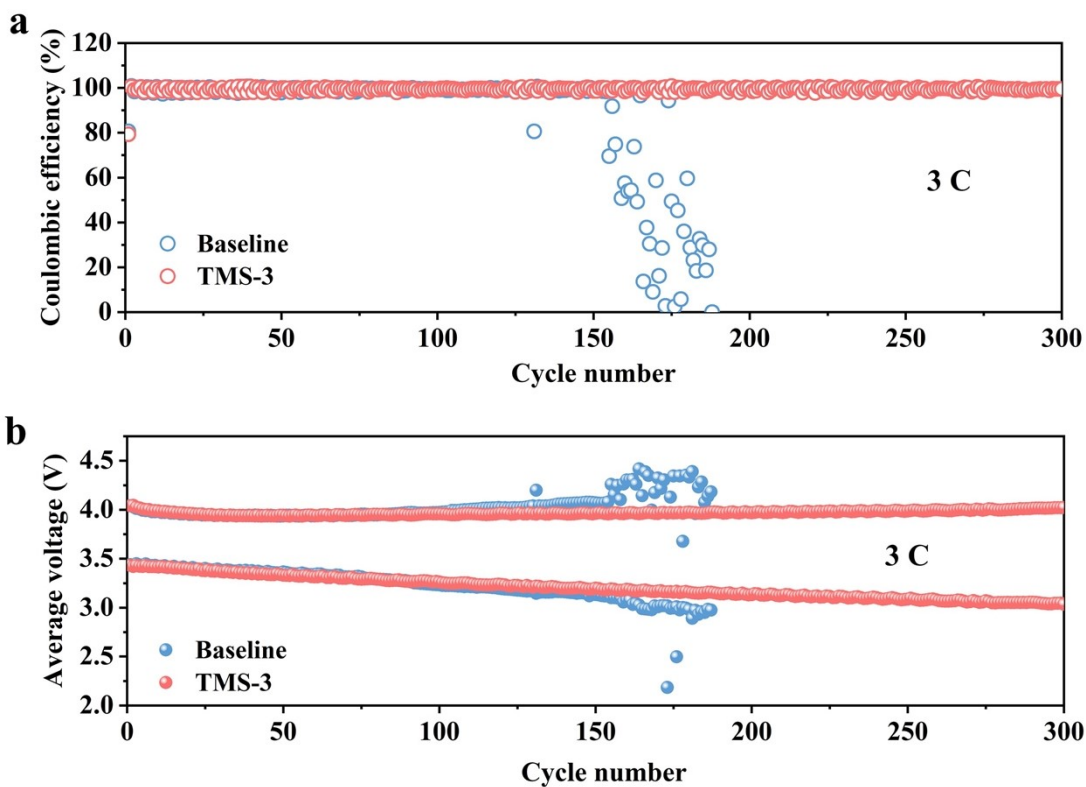


Fig. S8. (a) Coulombic efficiency and (b) the corresponding average voltage with different electrolytes at 3 C.

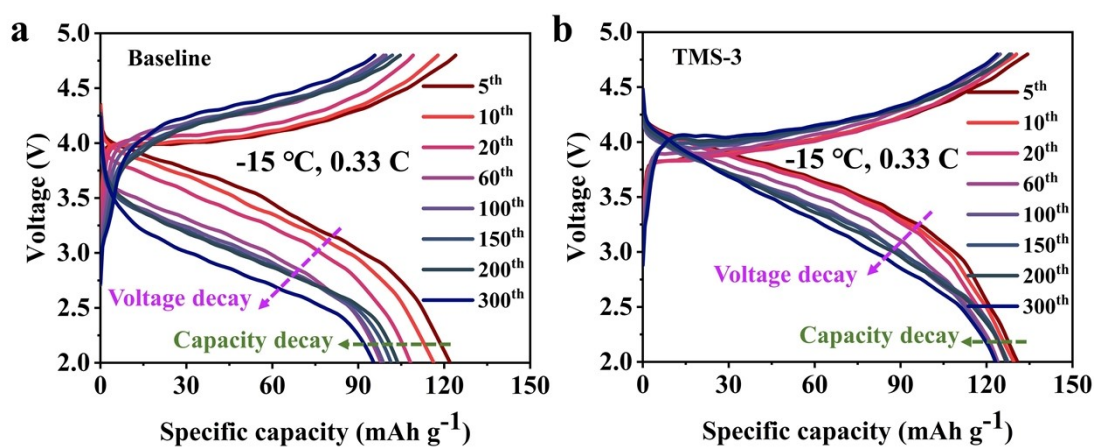


Fig. S9. The charge and discharge curves of Li||LRM batteries (a) without and (b) with TMS additive at 0.33 C and -15 °C.

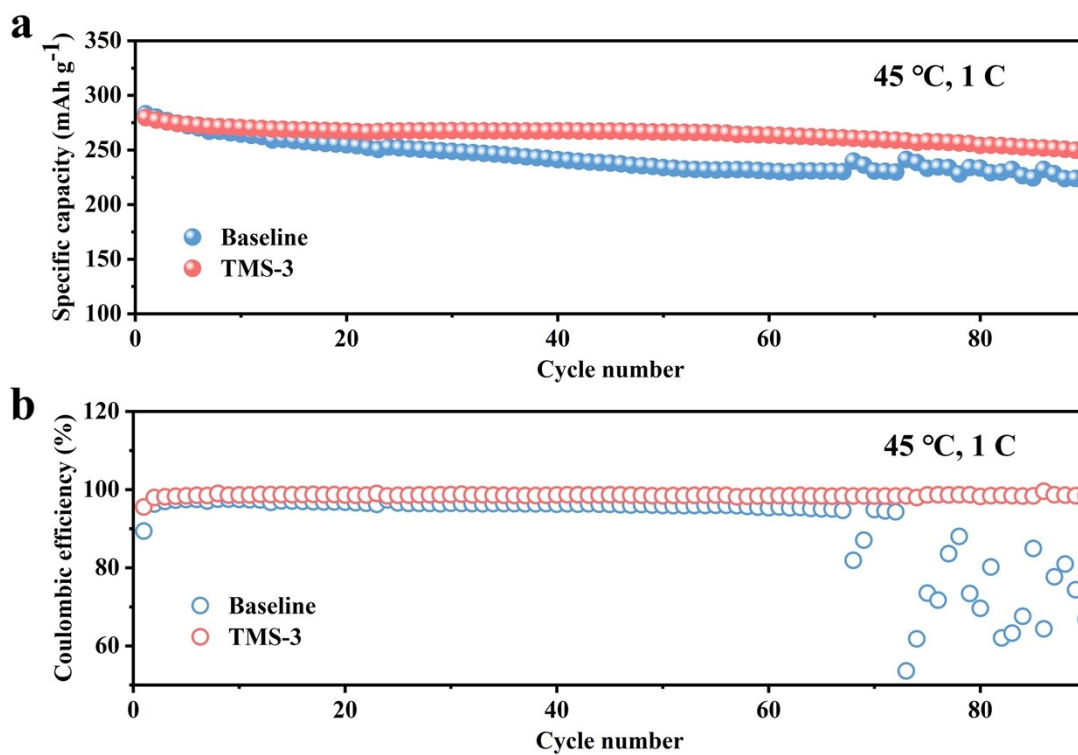


Fig. S10. (a) The Cycling performance with different electrolytes at 45 °C and 1 C and (b) the corresponding Coulombic efficiencies.

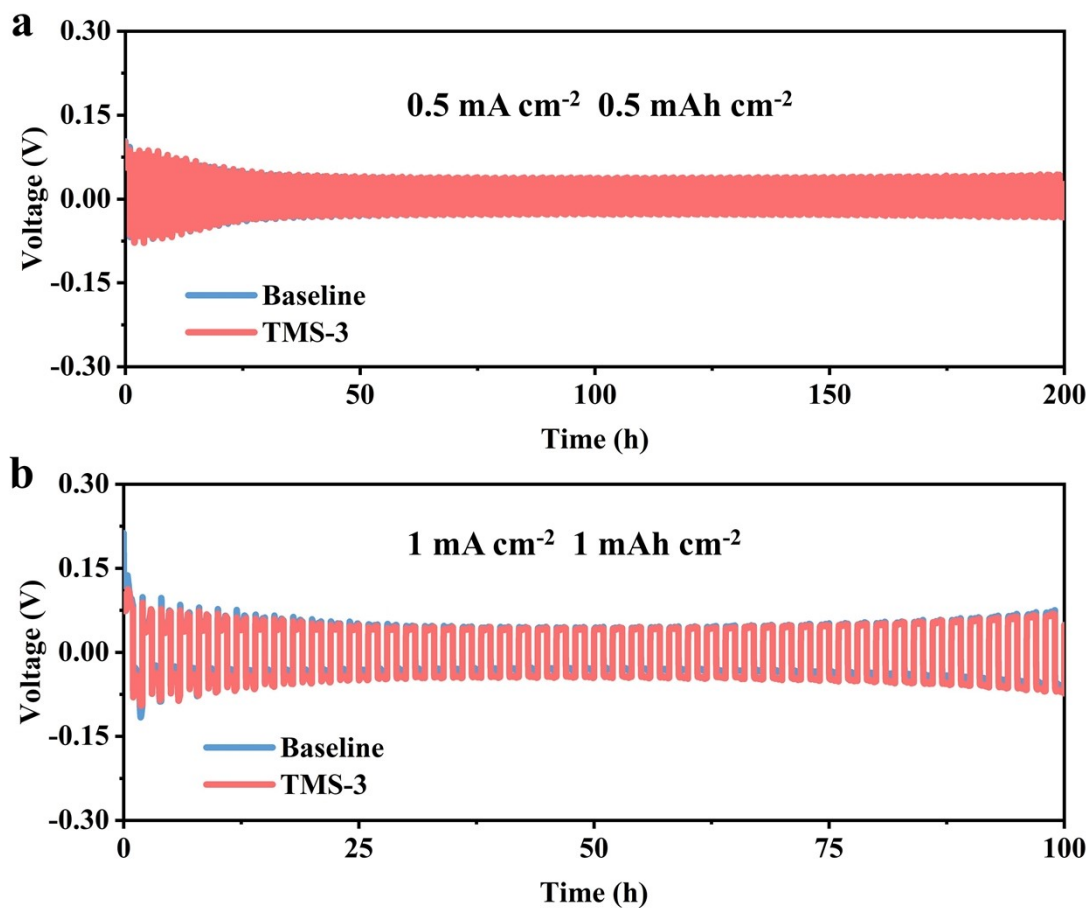


Fig. S11. Voltage curves of Li||Li symmetrical batteries with different electrolytes: (a) at 0.5 mA cm^{-2} with a capacity of 0.5 mAh cm^{-2} , (b) at 1 mA cm^{-2} with a capacity of 1 mAh cm^{-2} .

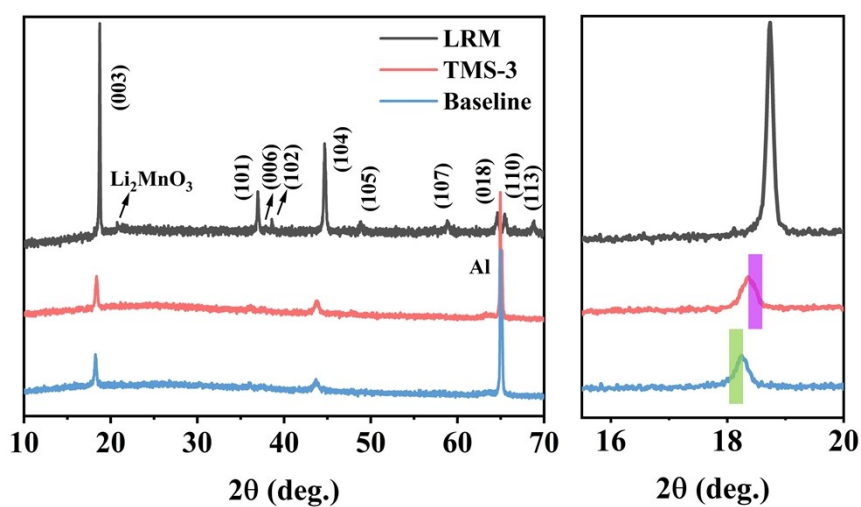


Fig. S12. XRD patterns of LRM cathodes without and with TMS additive after 100

cycles.

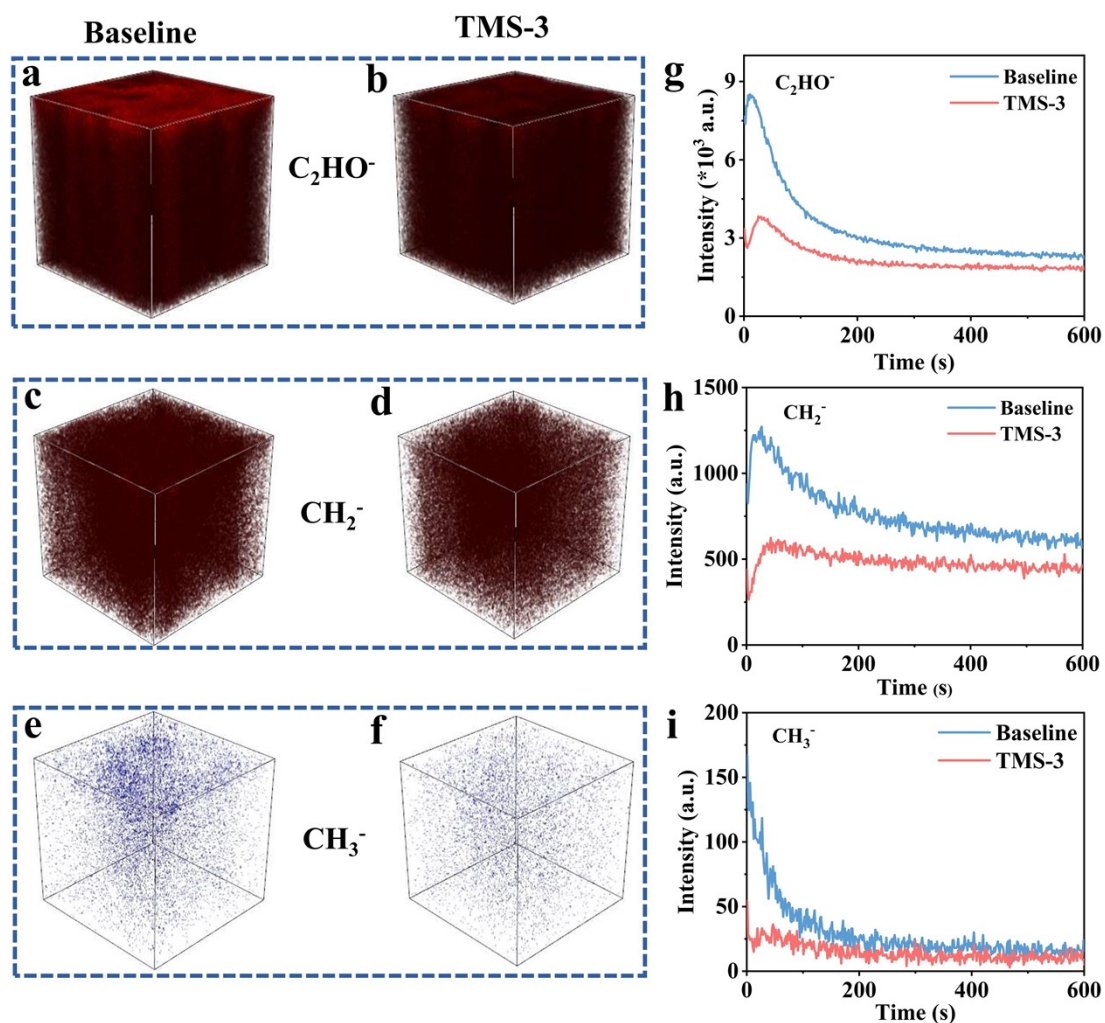


Fig. S13. TOF-SIMS 3D render images of fragments on the cycled cathodes with the (a, c, e) baseline and (b, d, f) TMS-3 electrolytes. The corresponding depth curves for (g) C_2HO^- , (h) CH_2^- , and (i) CH_3^- species.