

Supporting Information

Synergistic Effect of TMSPi and FEC in Regulating the Electrode/Electrolyte Interfaces in Nickel-Rich Lithium Metal Batteries

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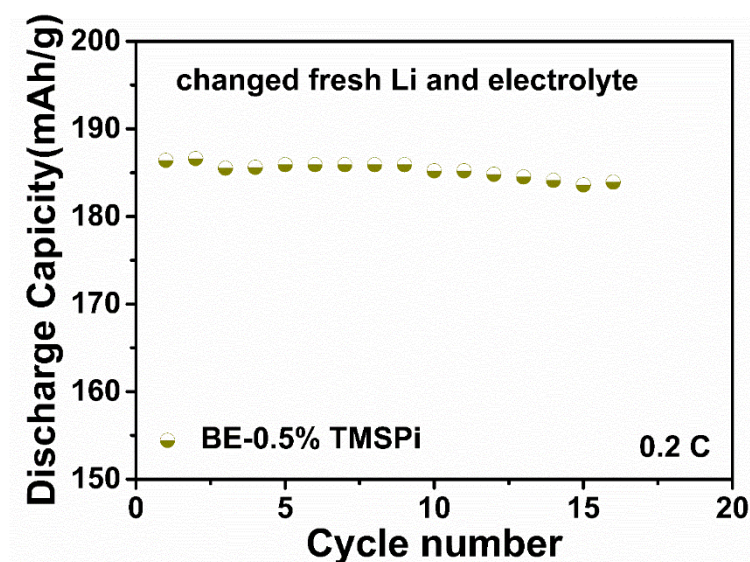


Figure S1. Charge/discharge profile of the reassembled NCM811||Li with BE-0.5% TMSPi.

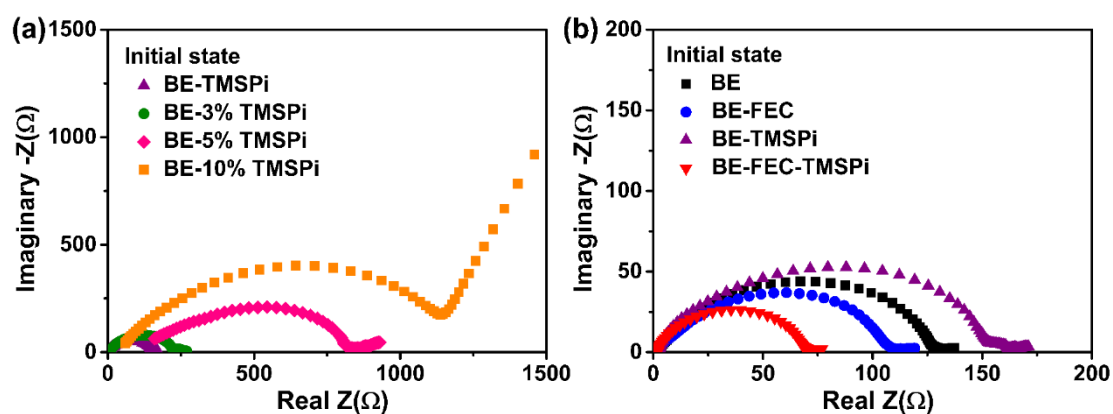


Figure S2. Nyquist plots of Li||Li symmetric cells after standing for 24 h (a) Electrolytes with different content of TMSPi. (b) Electrolytes in different additives.

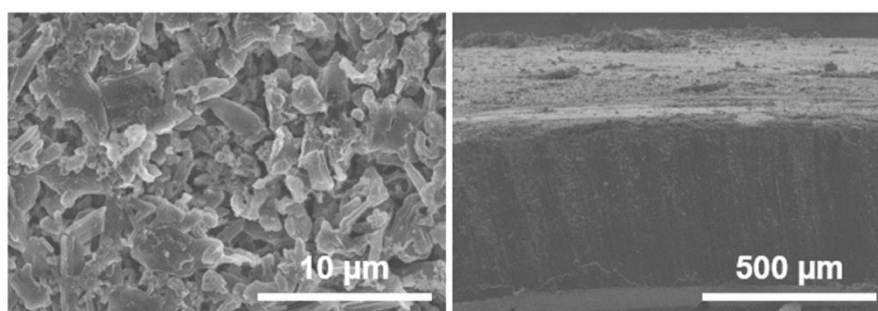


Figure S3. SEM pictures of lithium metal cycled in BE-TMSPi.

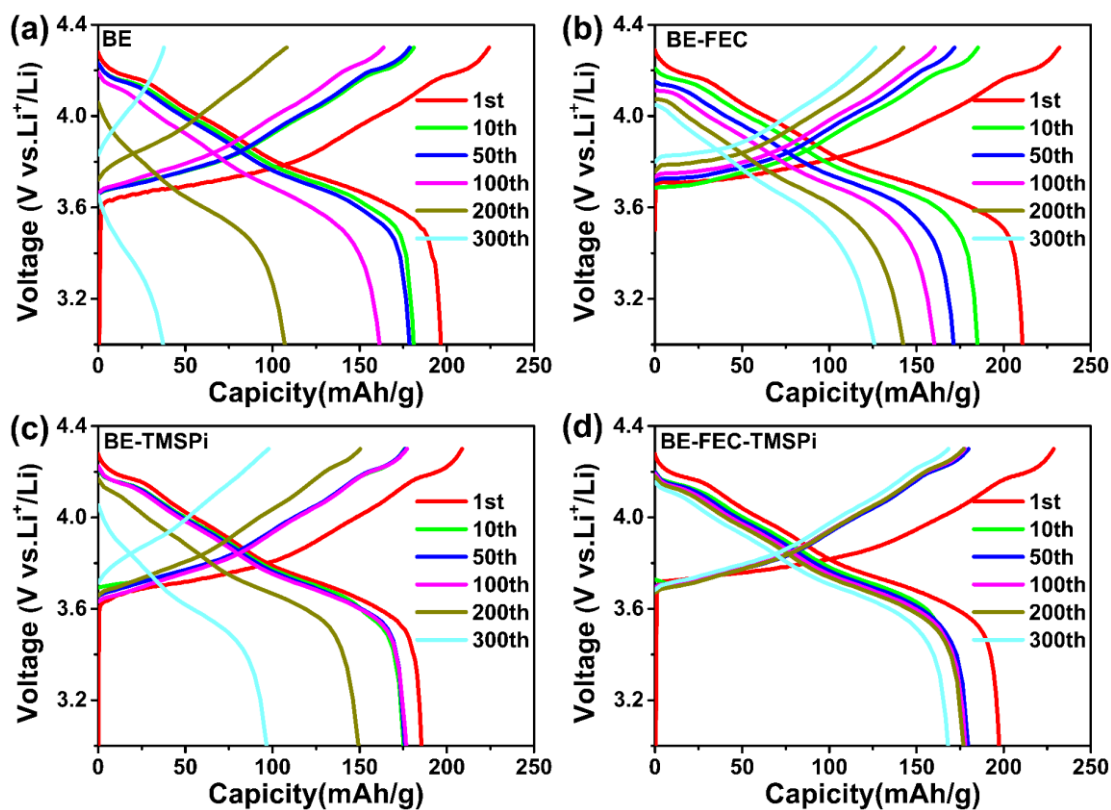


Figure S4. Voltage profiles of selected cycles of cells with different electrolytes: BE (a), BE-FEC (b), BE-TMSPi (c) and BE-FEC-TMSPi (d).

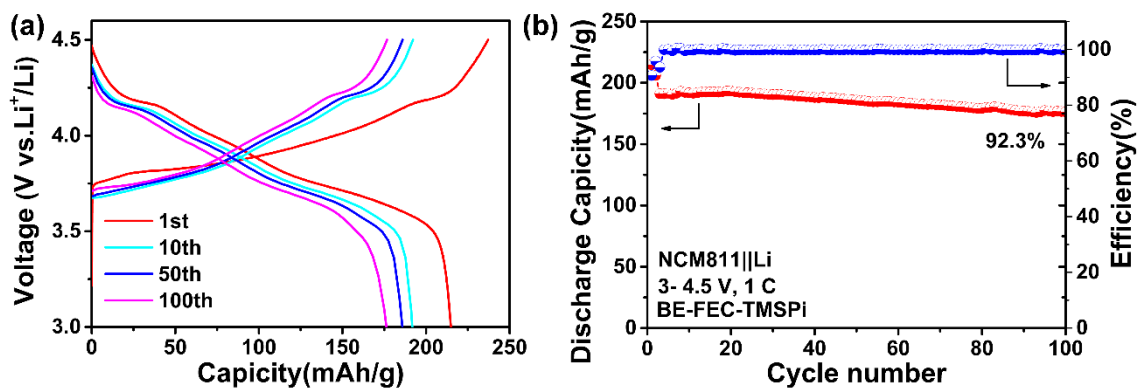


Figure S5. (a) Voltage profiles of selected cycles for the cell with BE-FEC-TMSPi; (b) Cycle performance of NCM811||Li cell in BE-FEC-TMSPi between 3 ~ 4.5 V at 1 C rate.

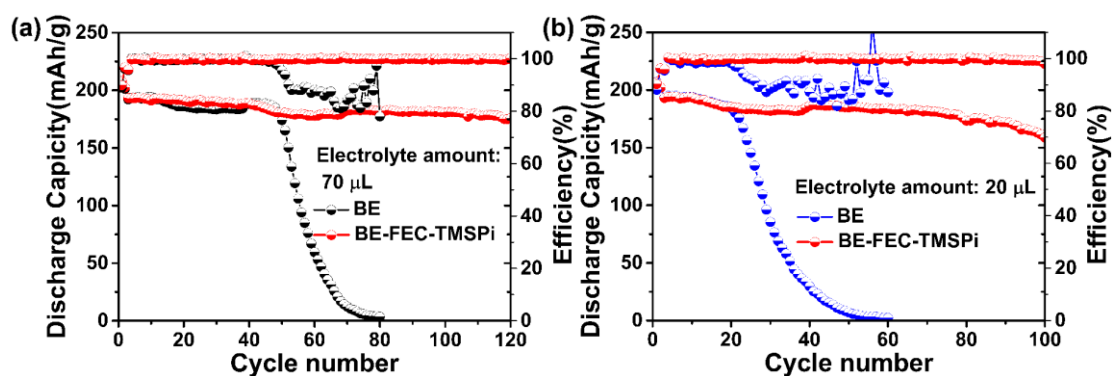


Figure S6. Cycle life test of LMBs full cells with the N/P of 7.1 (cathode: 7.3 mg cm^{-2} , anode: $50 \text{ } \mu\text{m}$ Li foil) at 0.5 C . (a) with $70 \text{ } \mu\text{L}$ electrolyte. (b) with $20 \text{ } \mu\text{L}$ electrolyte.

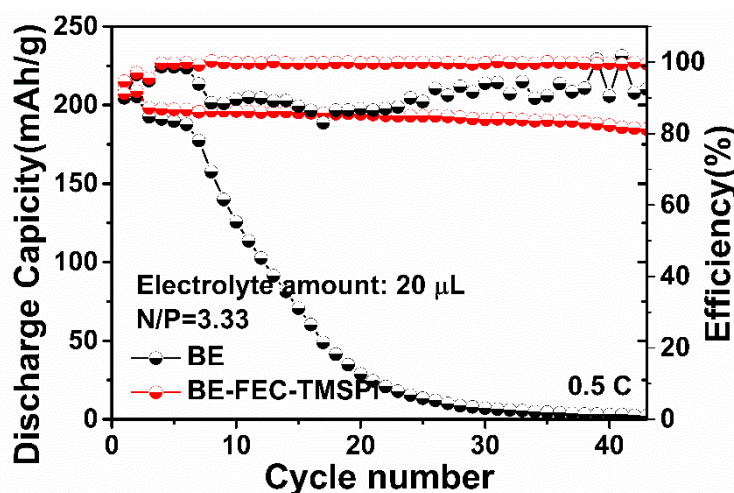


Figure S7. Cycle life test of LMBs full cells with the N/P of 3.3 (cathode: 16.5 mg cm^{-2} , anode: $50 \text{ } \mu\text{m}$ Li foil) with $20 \text{ } \mu\text{L}$ electrolyte at 0.5 C rate.

Table S1 I(003)/I(104) value of fresh and cycled cathodes

Sample	I(003)/I(104)
Fresh	1.71
BE	1.04
BE-FEC	1.43
BE-TMSPi	1.65
BE-FEC-TMSPi	1.45

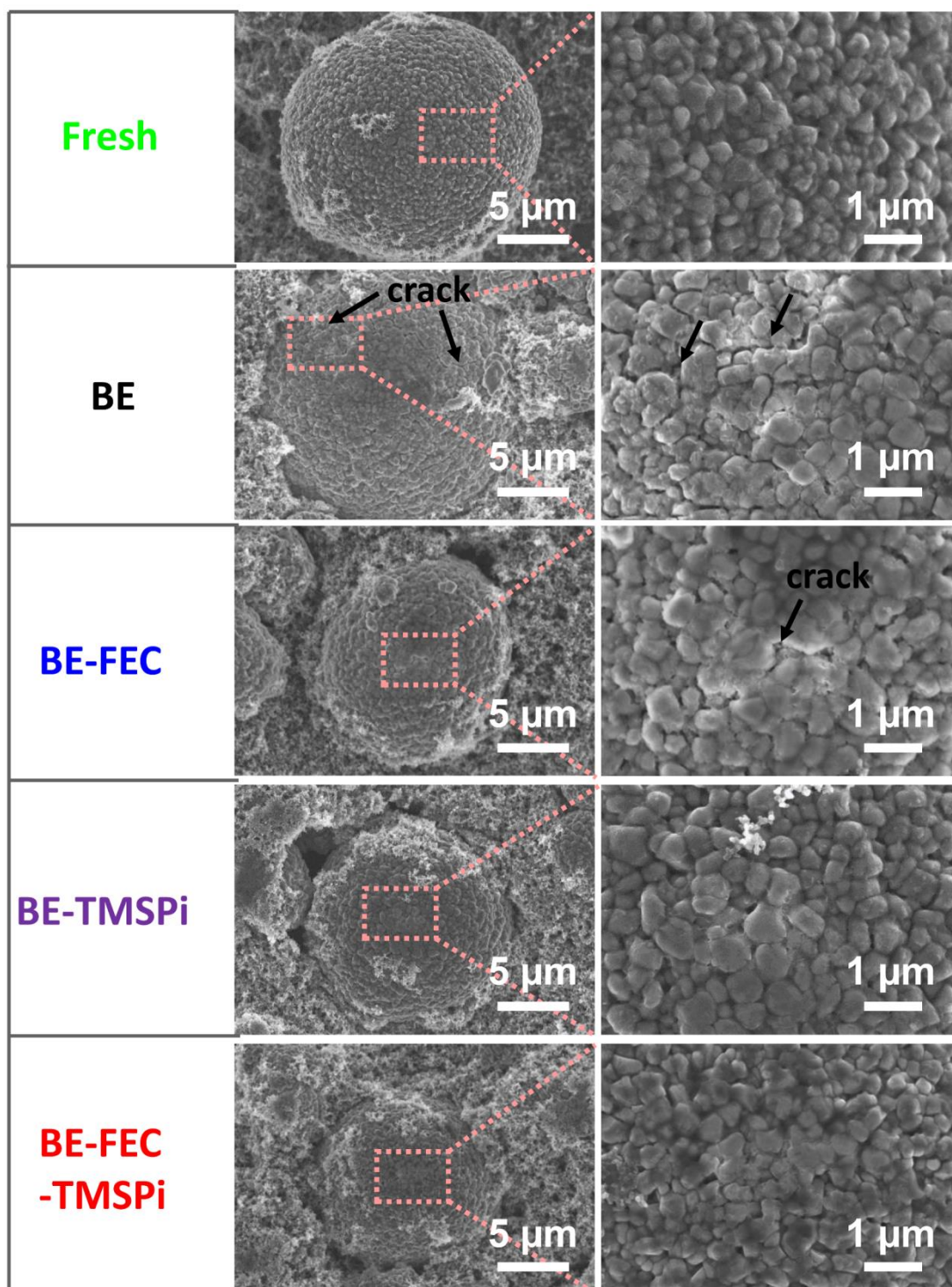


Figure S8. SEM images of Fresh NCM811 electrode and the cycled NCM811 electrode after 200 cycles at 1 C in various electrolytes.

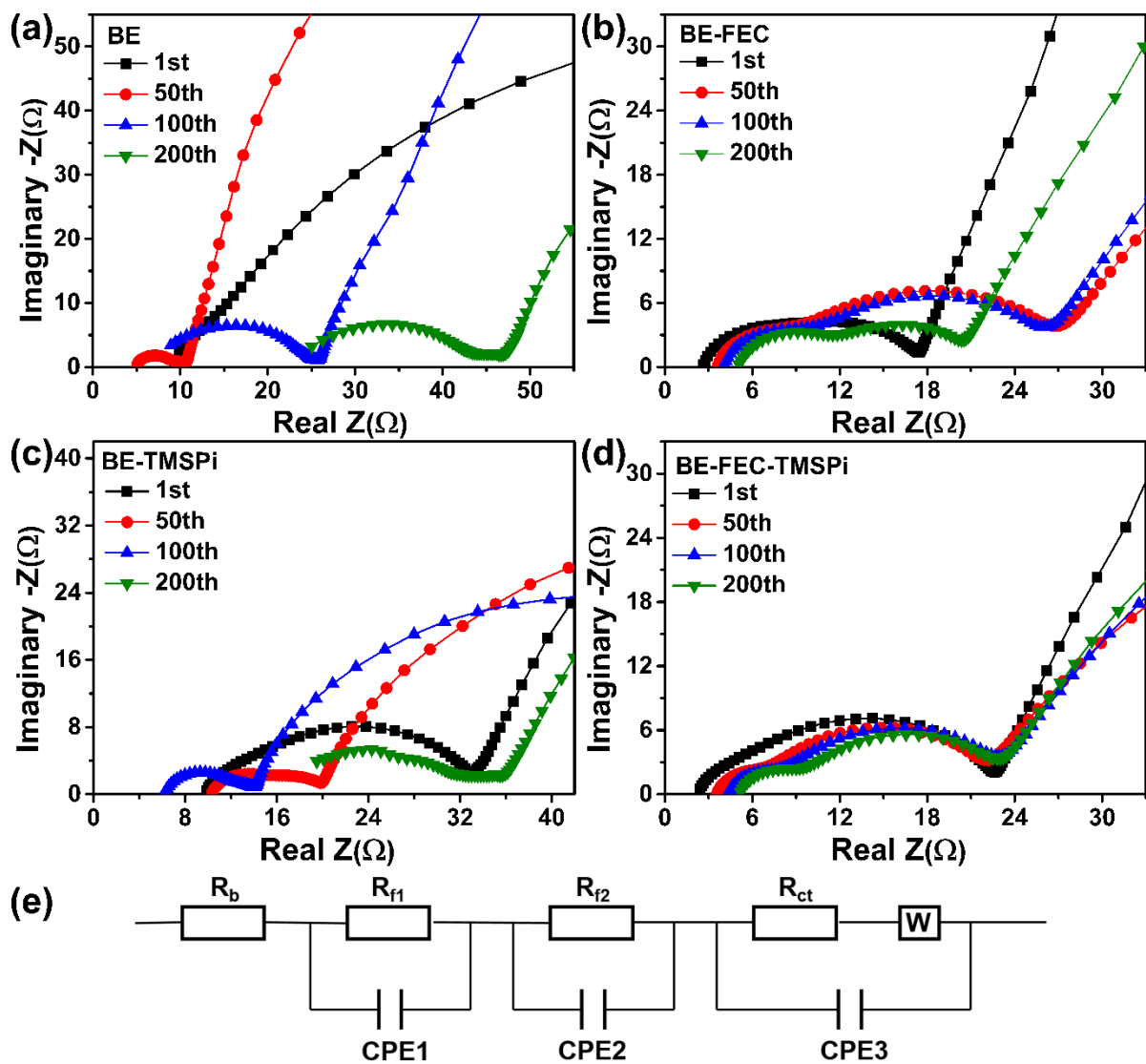


Figure S9. (a-d) An enlarged view of the high-frequency region and (e) the corresponding equivalent circuit.

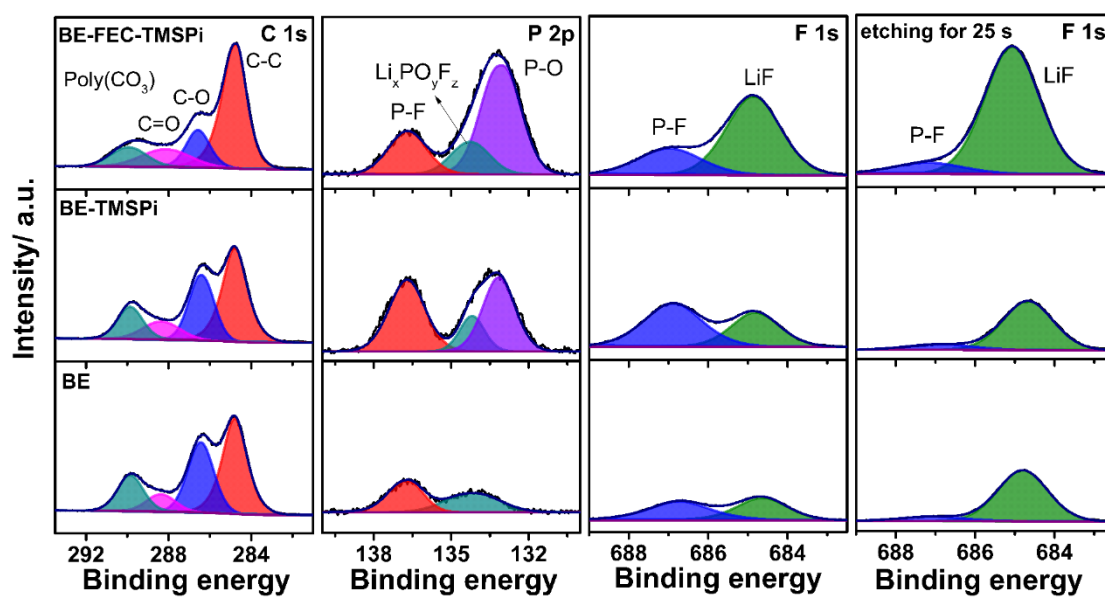


Figure S10. The surface and depth XPS spectra (C 1s, P 2p, F 1s) of Li metal anodes harvested from NCM811||Li cells after 50 cycles at 1 C in different electrolytes.

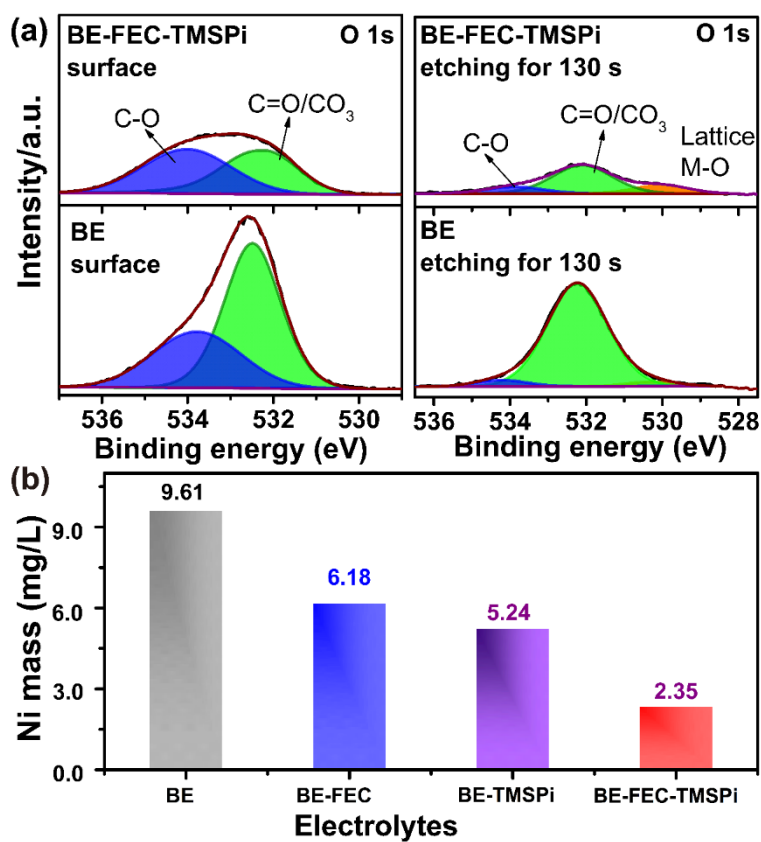


Figure S11. (a) O 1s in-depth XPS spectra s of NCM811 after 200 cycles in BE and BE-FEC-TMSPi. (b) Contents of Ni ions on lithium electrodes and separators from the NCM811||Li cells after 200 cycles in different electrolytes.