

## Cobalt free lithium batteries

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Lithium-ion batteries (LIBs) play an essential role in enabling the transition to a sustainable society with reduced carbon emissions by supporting clean energy generation, green transportation and more efficient energy use. It is widely expected that achieving a lower carbon and greener future will rely on the development of LIBs with high energy density (high capacity and high voltage), high environmental friendliness and low cost (Earth abundance).

a, Representative solution structures of 1.0 M LiFSI/EMC, 1.0 M LiFSI/FEMC and 3.4 M LiFSI/FEMC, as calculated via MD simulations. b, Raman spectra of the prepared electrolytes. The peak between 700 and 780  $\text{cm}^{-1}$  corresponds to the coordination environment of the FSI<sup>-</sup> anion.

Consequently, the considerably upshifted redox potential of  $\text{Li/Li}^+$  (and thus, simultaneously upshifted reaction potential of  $\text{SiO}_x$ ) in 3.4 M LiFSI/FEMC unburdens the kinetic support of the SEI, contributing to the decrease in the reductive decomposition of the electrolyte at the  $\text{SiO}_x$  surface.

a,b, Li 1s, F 1s, S 2p (a) and Si 2p (b) XPS spectra of the  $\text{SiO}_x$  electrodes cycled in 1.0 M LiFSI/EMC and 3.4 M LiFSI/FEMC. c, Schematic diagram of the SEI stability on cycling in electrolytes with different compositions.

Overall, stable cycling of the  $\text{SiO}_x$  anode is thermodynamically (upshifted electrode potential) and kinetically (anion-derived SEI) realized using 3.4 M LiFSI/FEMC, which is due to its distinct solution structure.

a, Charge and discharge curves of the  $\text{Li}|\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$  half-cell with 3.4 M LiFSI/FEMC. The inset shows the Coulombic efficiencies of the cells with the prepared electrolytes as a function of the cycle number. The first charge and discharge curves are shown in Supplementary Fig. 9. b,c, Oxidative stabilities of the electrolytes on Pt (b) and corrosion stabilities of the Al current collector (c) in the three electrolytes. The origin of low initial Coulombic efficiency can be a passivation film (SEI and CEI) formation (Fig. 4 and Supplementary Fig. 10).

In conclusion, 3.4 M LiFSI/FEMC provides unusual stabilities at the  $\text{SiO}_x$  anode and  $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$  cathode, which are incomparable to those of other electrolytes proposed so far (Fig. 6b).

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