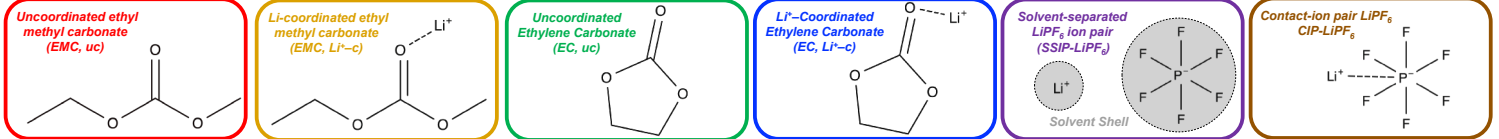


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Key Findings

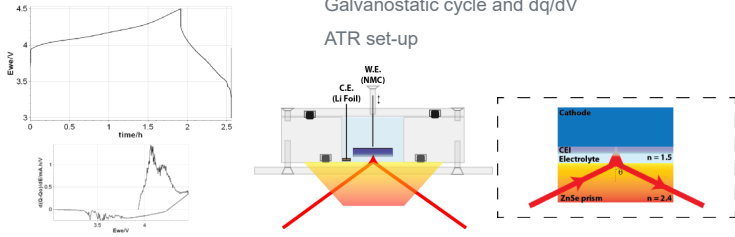
- EMC decomposes faster than EC, suggesting that EMC plays a greater role in the electrochemical reactions on the electrode surface, e.g., formation of a protective layer to prevent further electrolyte decomposition.
- We observe an increase [decrease] of the vibrational absorption peaks attributed to carbonates coordinated with Li^+ ions relative to the decrease [increase] of their uncoordinated counterparts, as expected when changing the local concentration of Li^+ ions near the electrode surface during deintercalation [intercalation] into the electrode.
- Features assigned to the Li^+ - PF_6^- contact ion pair increase in intensity relative to the uncoordinated PF_6^- anion when Li^+ concentration increases.

Molecular Species in the Gen2 Electrolyte:

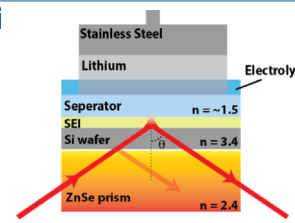


Model Cathode: NMC

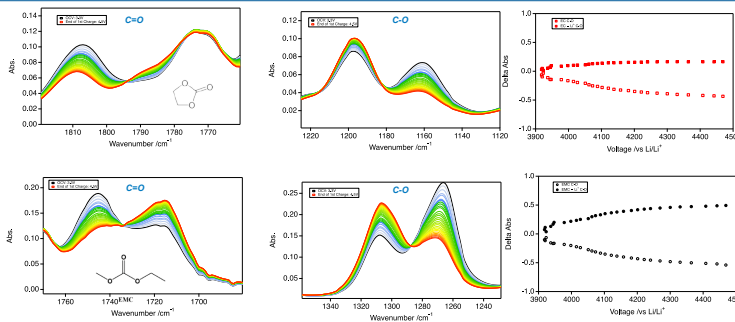
battery configuration, ATR-FTIR set up, Galvanostatic cycle and dq/dV



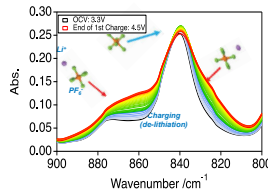
Model Anode: Si



- Cell configuration simulates coin cell environment
- Si wafer acts as both the anode and ATR crystal
- Evanescent wave probes ~80-800 nm (controlled by angle of incidence)

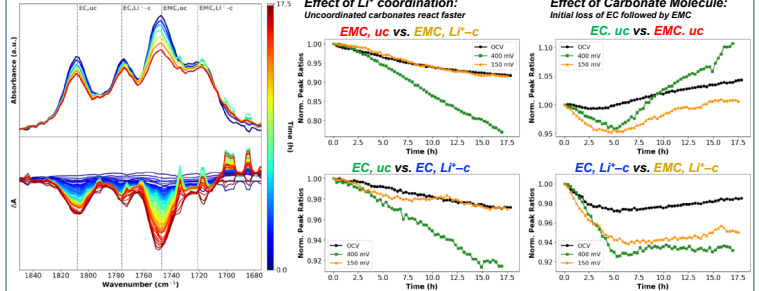


EC and EMC, C=O and C-O vib. abs., intensity change at EC vs EC - Li^+ and intensity change at EMC vs EMC - Li^+ , PF_6^- -- Li^+ (CIP) increases with charging b/c more and more Li^+ coming out of cathode.

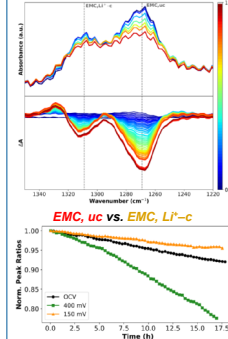


D. M. Seo et al., J. Phys. Chem. C 2015, 119, 14838-14866, Crease et al., Phys. Chem. Chem. Phys., 2017, 19, 374-386

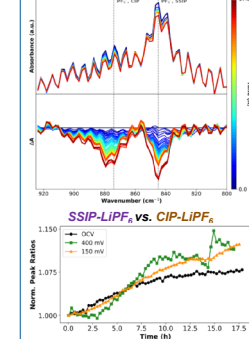
C=O Stretches in Gen2 Electrolyte



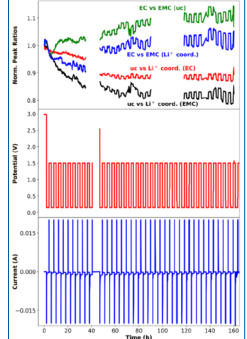
C(O)-O Carbonate Stretches



LiPF6 Stretches



Cycling to 150 mV vs Li



Conclusions

These observations give important information regarding the electrolyte solution structures and dynamics near the electrode surfaces. We look forward to applying these techniques to additional lithium-ion battery systems with different cathode or anode materials and **beyond lithium-ion systems** to characterize surface chemistry/evolution (on-going research) and solution structures/dynamics (in particular, (de)solvation behaviors) within the EEI, which can be a bottleneck to efficient ion transfer from the electrolyte to the electrode.