A Carbonate-Free, Sulfone-Based Electrolyte for HV Lithium Batteries

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Harnessing the enhanced energy and power metrics offered by "5V-class" lithium-ion electrode chemistries relies on development of more robust electrolytes with expanded voltage and temperature stability windows. As a part of this effort, electrochemists have explored the promise of sulfones, highlighting their oxidative stability (generally >5.0V vs. Li/Li^+ compared to ~4.3V for conventional electrolytes), safety, and compatibility with high voltage systems.[1-3] Unfortunately, due to their high melting points, viscosity, and inability to independently form a stable solid electrolyte interphase (SEI), practical implementation of sulfones has generally been limited to use as minority co-solvents in more conventional electrolyte systems.

In this work we explore the effects of solvation, concentration, and salt chemistry on sulfone-based electrolyte performance. In graphite anode half-cells, we characterize the formation of a stable SEI which enables full utilization of the graphite electrode for the first time in an electrolyte with a sulfone as the primary solvent. In high voltage full cells, the oxidative stability of the system is confirmed by voltammetry and galvanostatic cycling measurements. Anode and cathode surface chemistry is explored extensively with XPS, and coupled with computational calculations, a mechanism for the formation and operation of both electrode/electrolyte interphases is proposed. These proposed mechanisms are further explored and verified by cryo-TEM and EELS of the cathode electrolyte interphase (CEI). These results suggest sulfones warrant continued consideration as a system for enabling safe, high performance, next generation Li-ion batteries.

References:

1. Xu, K.; Angell, C. A., Sulfone-Based Electrolytes for Lithium-Ion Batteries. *Journal of The Electrochemical Society* **2002,** *149* (7), A920-A926.
2. Xu, K., Nonaqueous liquid electrolytes for lithium-based rechargeable batteries. *Chem Rev* **2004,** *104* (10), 4303-417.
3. Xu, K., Electrolytes and Interphases in Li-Ion Batteries and Beyond. *Chemical Reviews* **2014,** *114* (23), 11503-11618.