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Lithium Metal Protection for Li-S Batteries

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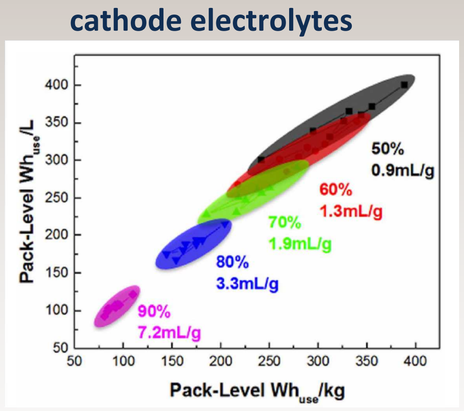
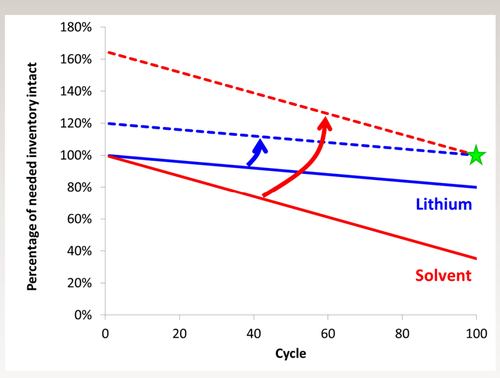
Motivation

- Electrolyte quantity in the Li-S battery drives energy density, cycle life, and cost
- Li deposition / dissolution disrupts the traditional SEI, resulting in continuous consumption of electrolyte salt and solvent
- Decouple the electrolyte requirements of the Li anode from the S-cathode: enable a Li anode compatible with range of S-cathode electrolytes



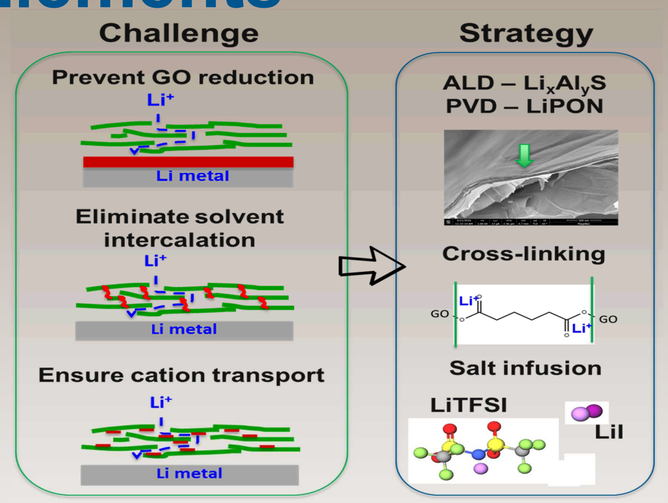
Li-S battery energy density as function of initial electrolyte reserves[1]



Estimate of excess inventory required to sustain 100 cycles at 99.8% CE

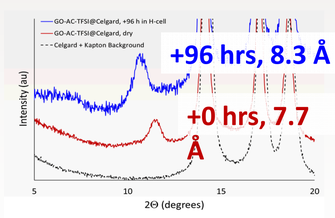
Integrated Design Elements

- Materials toolkit
  - ALD thin films for electrical insulation of GO from Li surface and/or internal fast-ion conduction
  - Cross-linking monomers functionalized on GO sheets for sealing membrane and fixing intersheet GO spacing
  - Lithium salt infusion forming high dielectric strength cation conducting phase in GO membrane

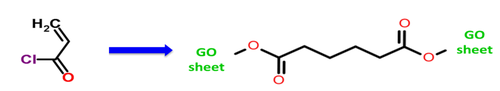


Design Driver: Solvent Rejection

- GO membranes swell in solvent
  - XRD indicates degree of solvent uptake by change in GO d-spacing
  - H-cell experiment: tracking solvent permeation by *ex-situ* GCMS and salt permeation by *in-situ* AC impedance spectroscopy

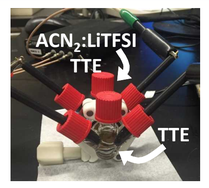


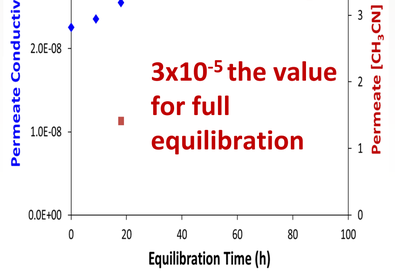
XRD of GO membrane after soaking in electrolyte



Acryloyl chloride functionalized GO sheets enables cross-linking of membrane by UV

H-cell set-up

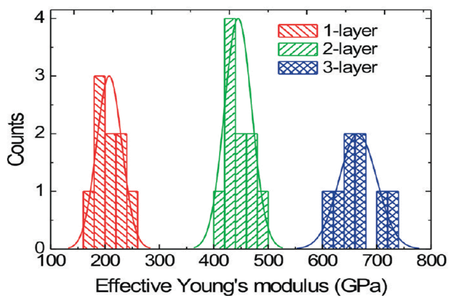




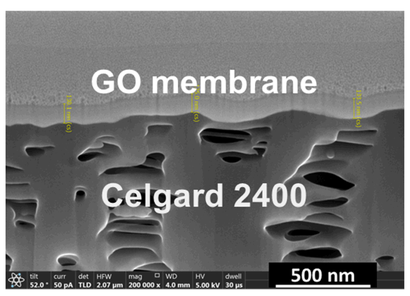
H-cell: tracking solvent and salt permeation through GO membrane

Protection Strategy *Materials Driven Approach*

- Foundation: graphene oxide membrane scaffold
  - Thin, strong, single ion (cation) conductor formed on existing separator material
  - High yield and shear strength for blunting Li dendrite formation
  - Solvent blocking, single ion (Li cation) conductor
  - GO is scaffold that supports diversity of functionalization schemes



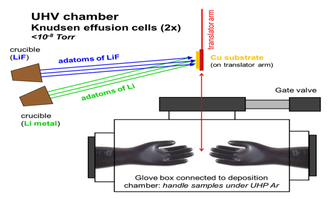
High Young's modulus for membranes of only two or three GO sheets – increase shear modulus by cross-linking[2]



Basic building block: laminar structure with continuous 2D tunable width slit for ion transport

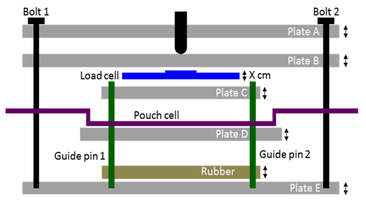
GO-Membrane / Li-Metal Interface

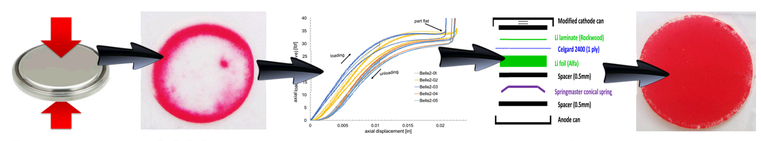
- Hypothesis: Li deposition / dissolution from Li metal anode possible only where surface contacts GO membrane
  - Practically achieved interface by means of compression of GO and Li at pressures at / beyond the Li yield strength
- Maintenance of formed GO-Li interface with restoring force (spring) in coin or pouch cell



Controlled study of interfacial effects conducted with Li evaporated directly upon GO

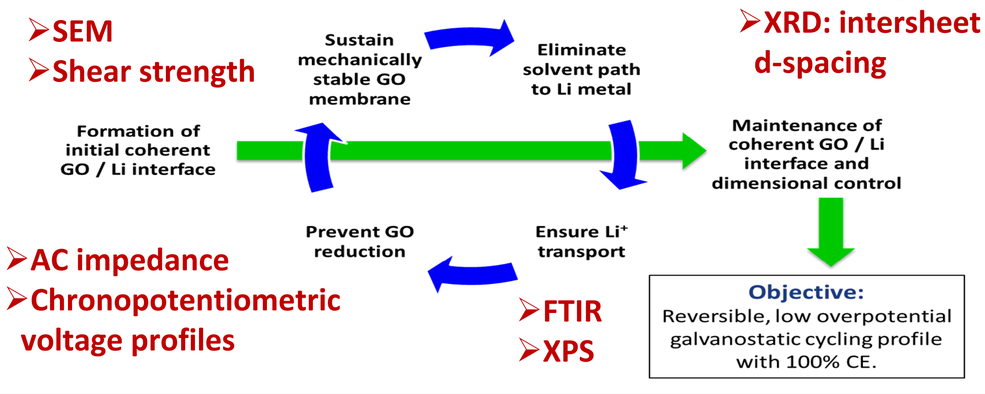
Cartoon of jig with in-line load cell for continuous monitoring of pouch cell compression





Redesign of 2032 for uniform compression at > Li yield strength

Critical Design Path

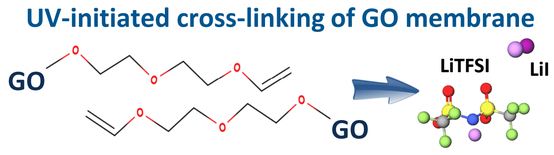


- SEM
- Shear strength
- Permeation monitoring
- XRD: intersheet d-spacing
- AC impedance
- Chronopotentiometric voltage profiles
- FTIR
- XPS

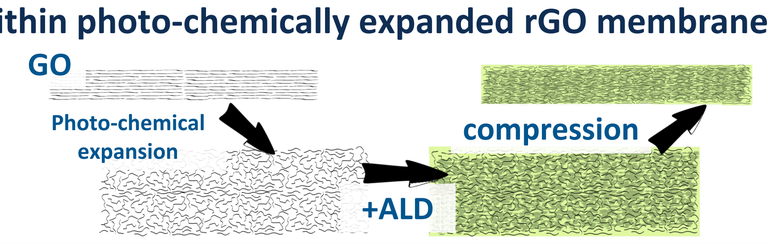
Future Work

PEGylation of GO membrane

Fast-ion conducting monomer designed for UV-initiated cross-linking of GO membrane



Fast-ion conducting, uniform coating ALD film within photo-chemically expanded rGO membrane



Acknowledgements

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References

U.S. DEPARTMENT OF ENERGY

National Nuclear Security Administration

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