SAND2016-4938PE

An incomplete Overview of Integrated Computational Materials Engineering at Sandia National Laboratories

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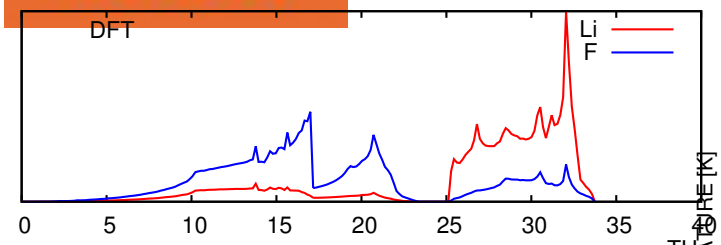

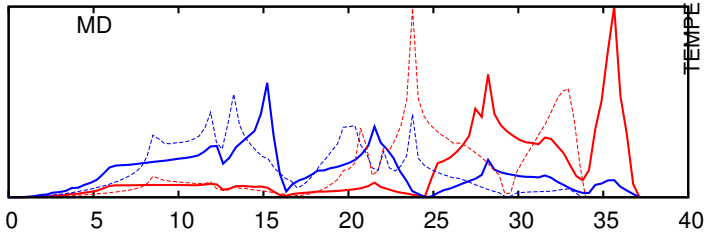

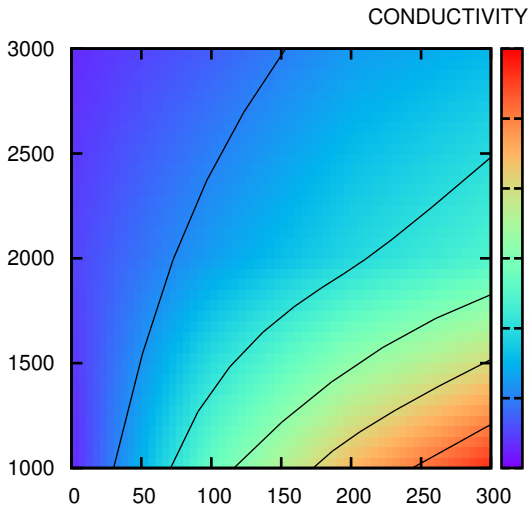

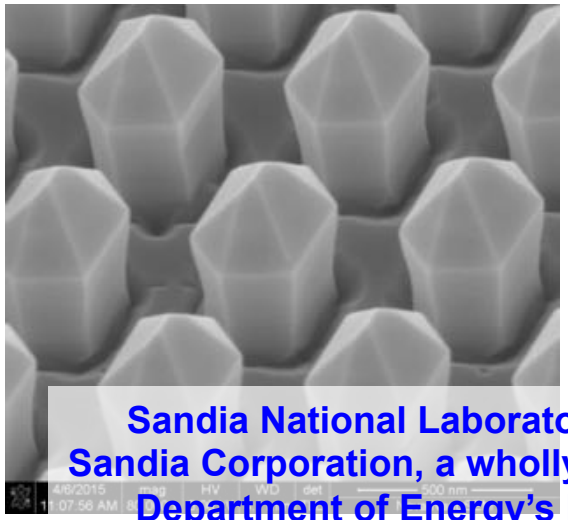

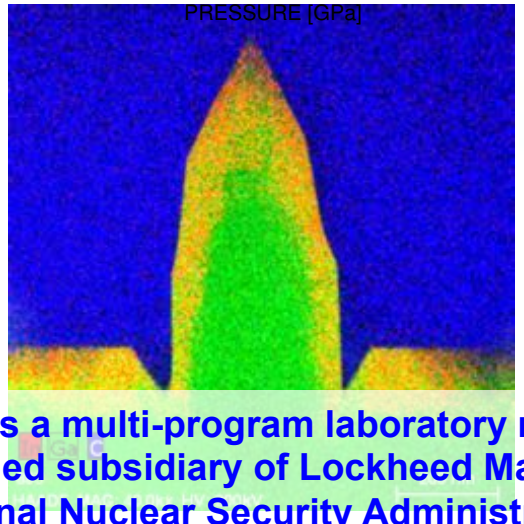

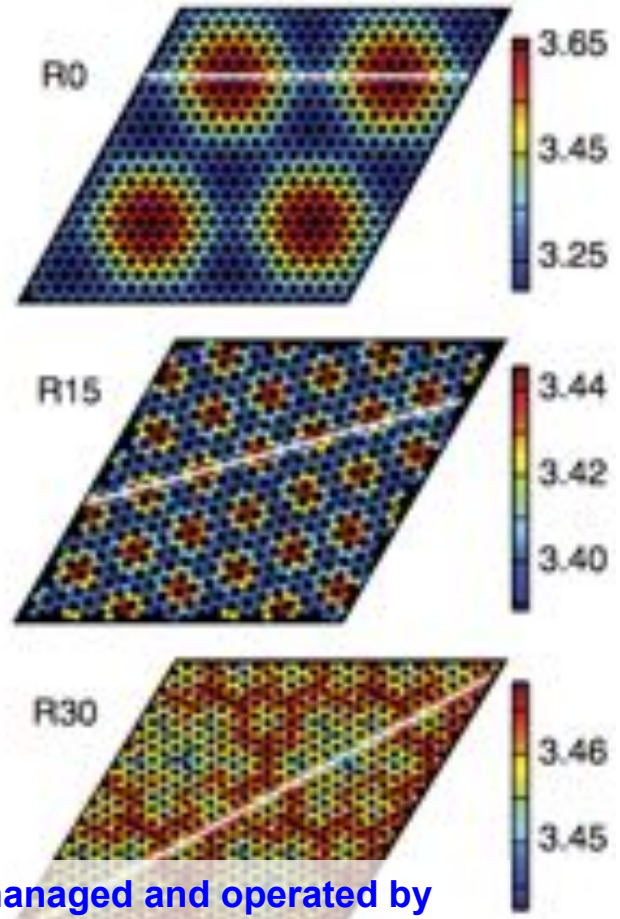

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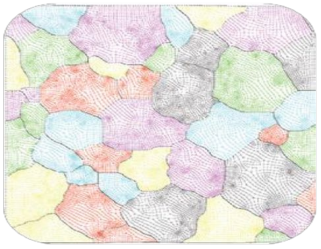
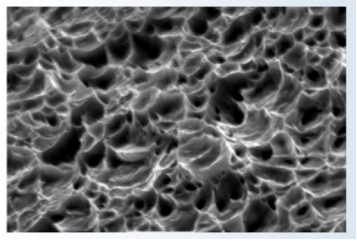
Laundry list

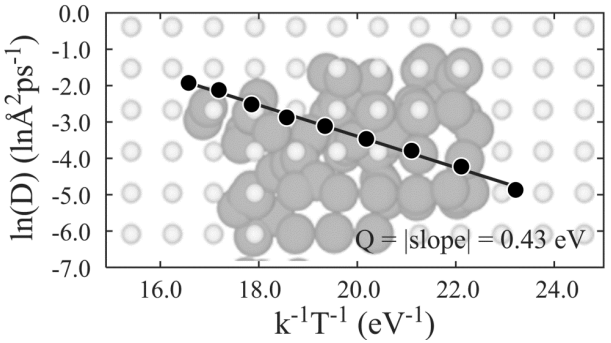




- **Predicting performance margins**
  - Goal is to provide a science-based, probabilistic underpinning for design and analysis capabilities that links microstructure differences to macro-scale property variability. **http://www.sandia.gov/PPM/**
- **Innovative Development, Selection and Testing to Reduce Cost and Weight of Materials for Components**
  - Identify alternatives to high-cost metals (annealed 316/316L stainless steel) for high-pressure components in hydrogen fuel cell systems
- **Organic Semiconducting Materials for Thin-Film Optoelectronic Devices**
  - Design/develop a organic polymer near-infrared photodetector
- **MOF@ALD: A New, Integrated Materials Platform to Improve Dye-Sensitized Solar Cells**
  - Develop a metal organic framework based dye-sensitized solar cell
- **Aging of TlBr**
  - TlBr has superior radiation properties but ages rapidly which implies vacancy concentration are be orders of magnitude higher than expected

Laundry list

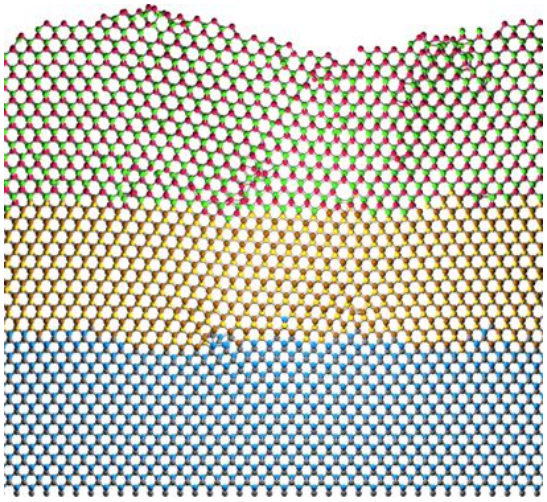
- **Compliant Nanoepitaxy: The Next Materials Revolution**
  - LED lighting is drastically reducing power requirements for lighting but more green is needed in the output. Indium is typically used to increase green output but causes excessive lattice mismatch and strain
- **DOE: HyMARC**
  - The goal is to understand hydrogen diffusion in hydrogen storage materials, which also impacts many Sandia problems especially on materials aging.



- **Soot inception modeling**
  - Basic question: what determines whether a hydrocarbon molecule can attach to a growing soot particle? The answer is crucial to predicting when soot forms (in combustion engines).
- **Transport properties at extreme conditions**
  - Sandia has an interest in material behavior at extreme conditions where experimental measurements are difficult. In some cases simulation can provide reliable estimates and physical insight

Laundry list

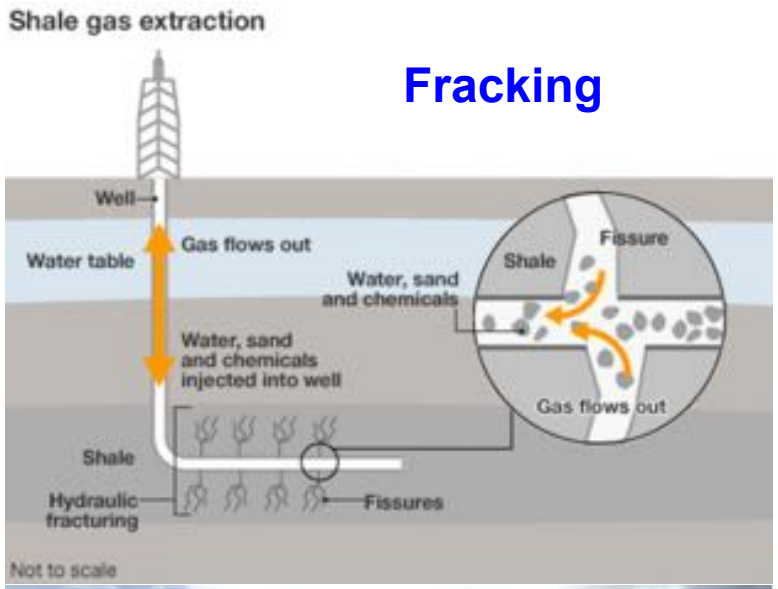
- **Fundamental models of ^{3}He bubble evolution**
  - Problem: ^{3}He bubbles nucleate and grow in aging metal tritides due to the radioactive decay of tritium. This bubble growth threatens the structural integrity of the tritide and it eventually leads to rapid release of He gas. Our goal is to develop and parameterize a model that predicts under what conditions this release occurs in a range of Pd alloys**.**
- **Sun-Shot: BRIDGE**
  - The goal is to use molecular simulations to understand defects in CdTe/ CdS solar cells.

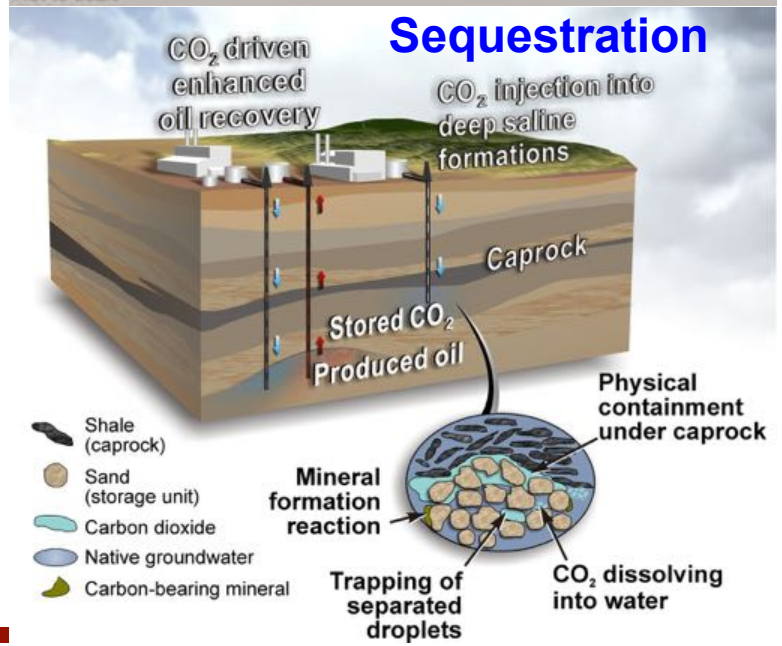


- **Predicting fracture toughness in geomaterials**
- **Synthetic screening of battery electrolytes**

Fracture of geomaterials

- **Goals:**
  - Develop fundamental understanding of the chemical-mechanical mechanisms that control subcritical cracks in low-permeability geomaterials
  - Link atomic-scale insight to macroscale observables and directly address how chemical environment affects mechanical behavior.
- **Why Atomistic Simulation?**
  - Cracks start at the atomistic scale – a crack is initiated by the breaking of bonds (e.g., Si-O) at the rock-fluid interface.
  - Crack tip formation and crack propagation is impacted by interfacial fluid and surface chemistry (e.g., development of surface charge, impact of adsorbed species along fracture surface).





Chemical Effects on Subcritical Rock Fracture

Stress-Corrosion Cracking

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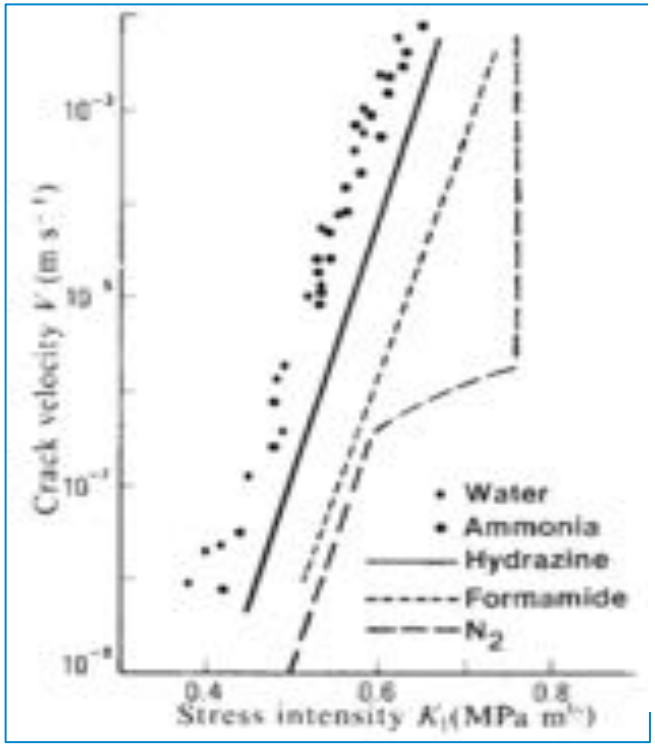
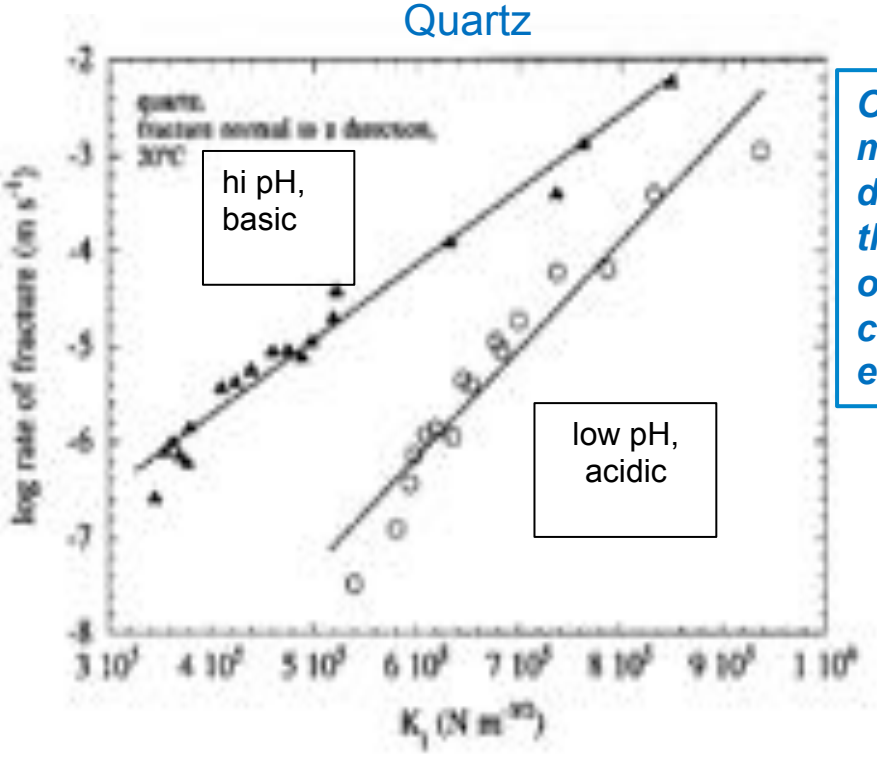
Silica Glass



T.A. Michalske & S.W. Freiman, Nature (1982)

Quartz



P. M. Dove, J. Geophys. Res. (1995)

Orders of magnitude difference in the rate of fracture with chemical environment

r_{SiO} =A x exp(-H+b K/KbT)

rate of bond breaking — chemical+mechanical

Mechanisms for stress-corrosion:
diffusion, ion exchange, dissolution, microplasticity, pressure solution,…

Connection to Aging

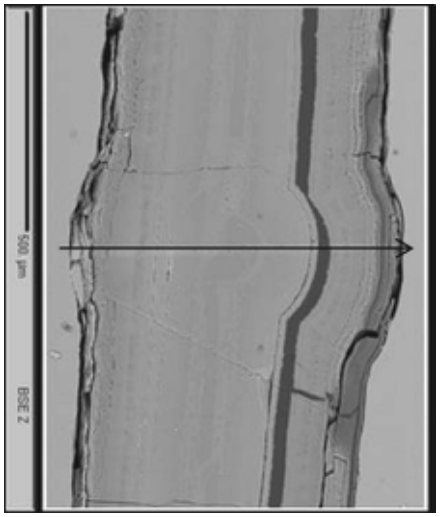
Serendipity experiments show that over the long run chemical changes dominate

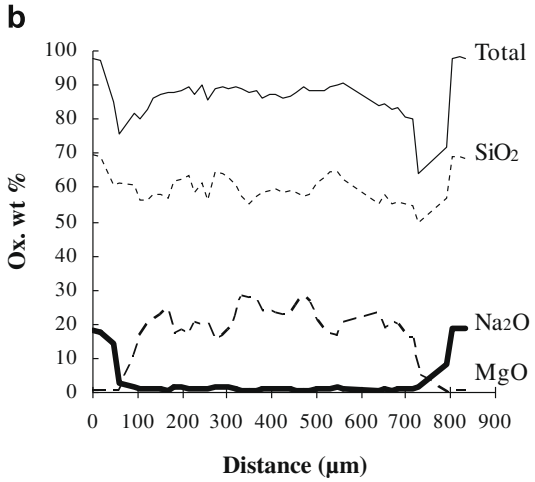
A fractured roman glass block altered for 1800 years in seawater: Analogy with nuclear waste glass in a deep geological repository

Aurelie Verney-Carron [a,*], Stephane Gin [a], Guy Libourel [b]

Using stained glass windows to understand the durability of toxic waste matrices

Jérôme Sterpenich [a,b], Guy Libourel [a,c,*]





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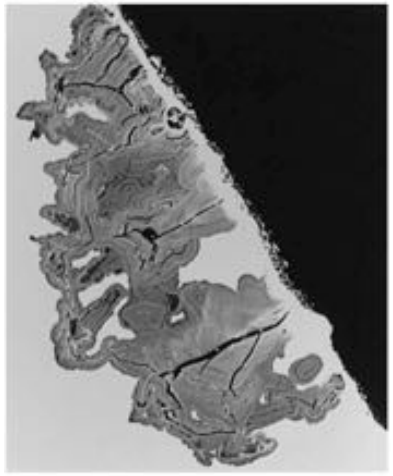
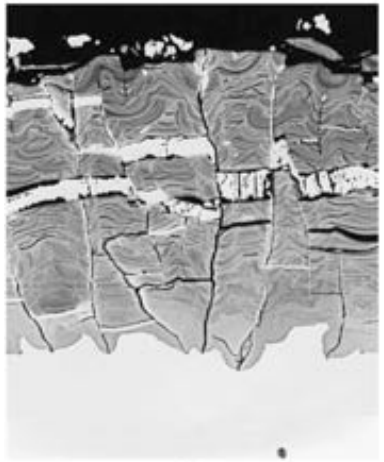
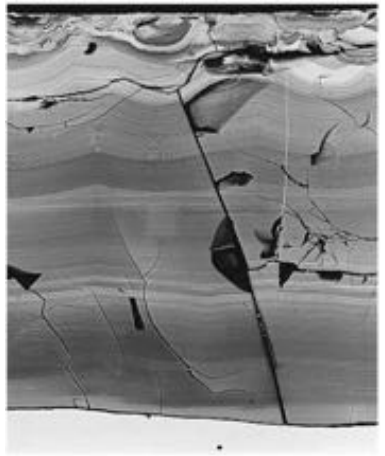
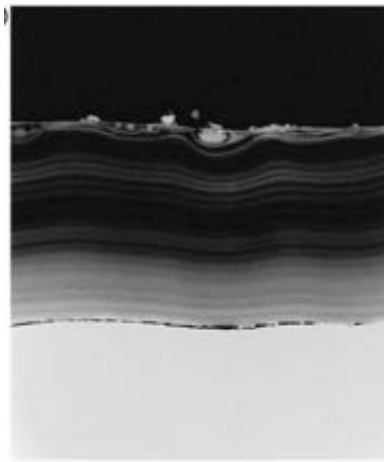
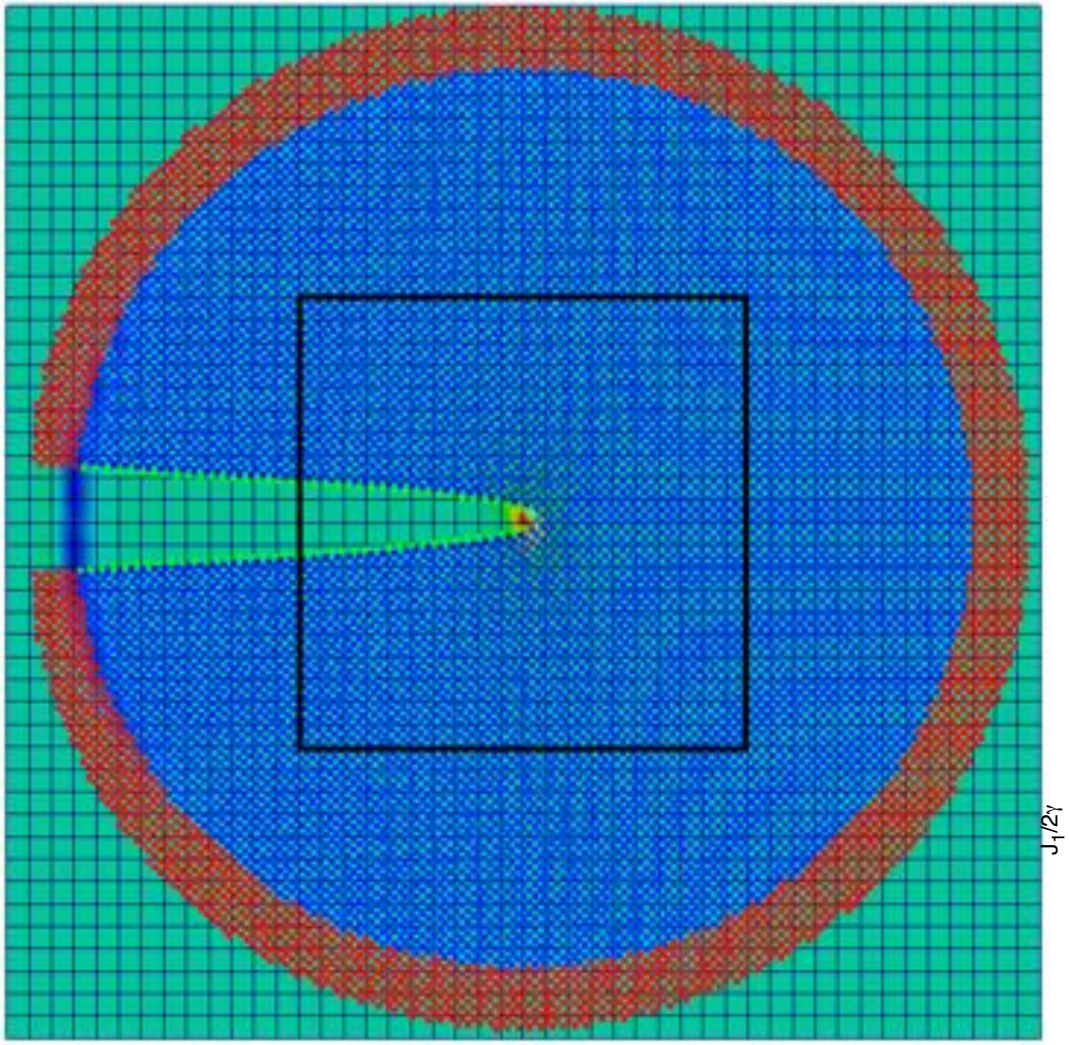




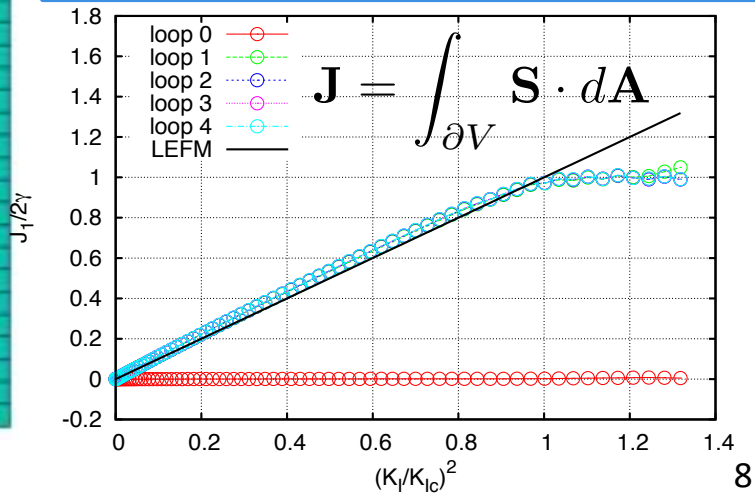




Method to upscale atomistic simulation to continuum fracture toughness



- A **slit** is inserted through bonds crossing x1 < 0, x2 =0
- A **far-field displacement** from an elastic solution is applied to the red annulus of atoms
- **Atomic data is upscaled** to continuum stress and displacement fields
- **J is evaluated** on the black contour



Developing practical Li-air batteries

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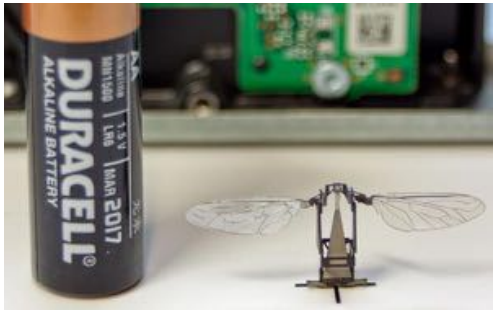
Lighter, Smaller

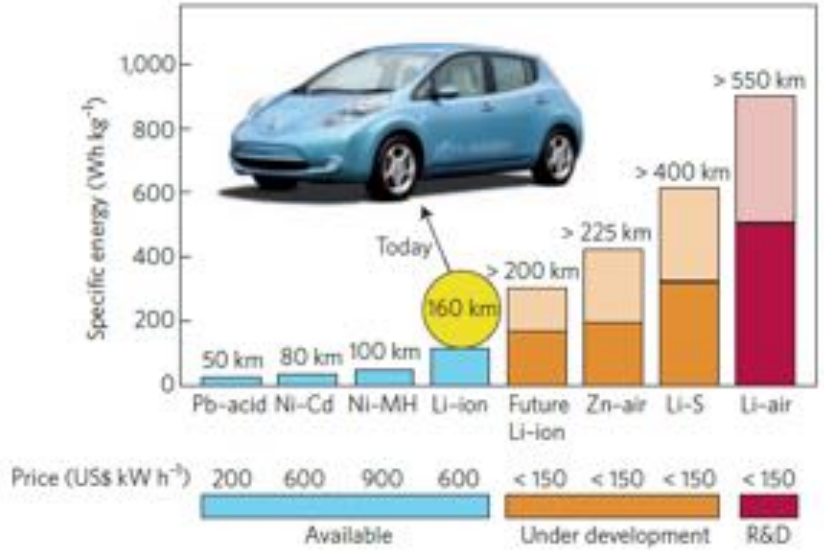
Grid Scale

Transportation

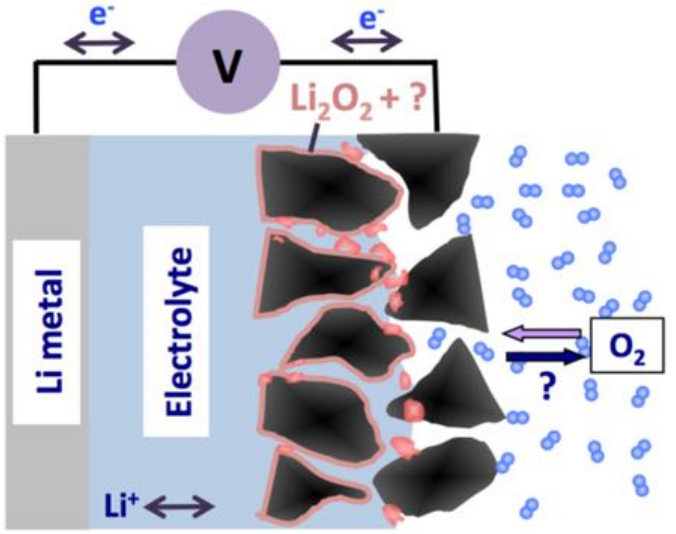
Portable Electronics



Microelectronics



Bruce et al. *Nat. Mater.*, 2011, **11**, 172–172.



Luntz and McCloskey, *Chem. Rev.*, 2014, **114**, 11721–11750.

Electrolyte downselect via simulation

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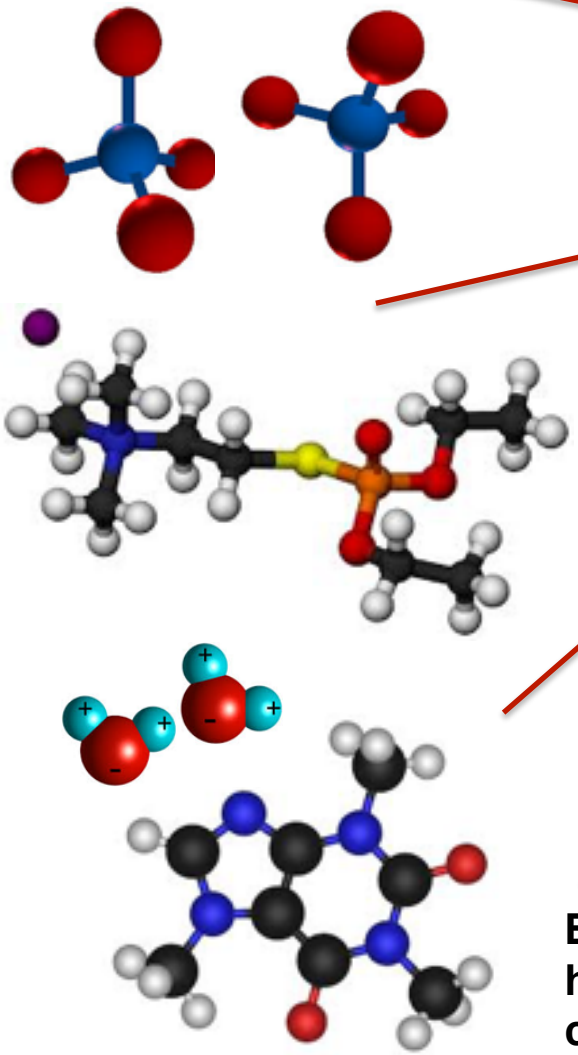
1000s of electrolyte candidates

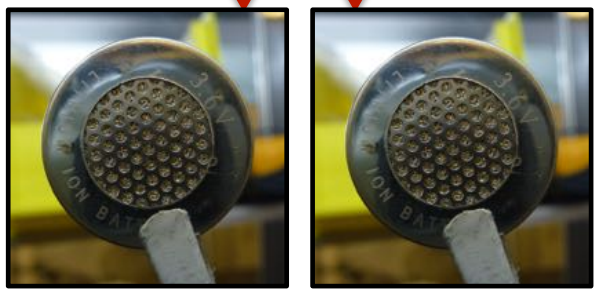


Prediction of transport properties via GK+MD

Diffusion & ionic conductivity

PDE battery model

power & capacity

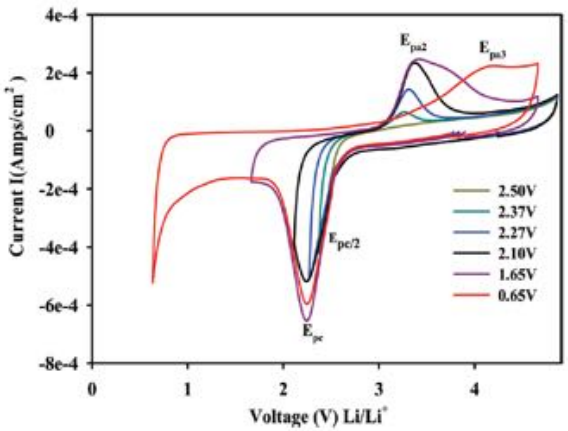


Electrochemical tests

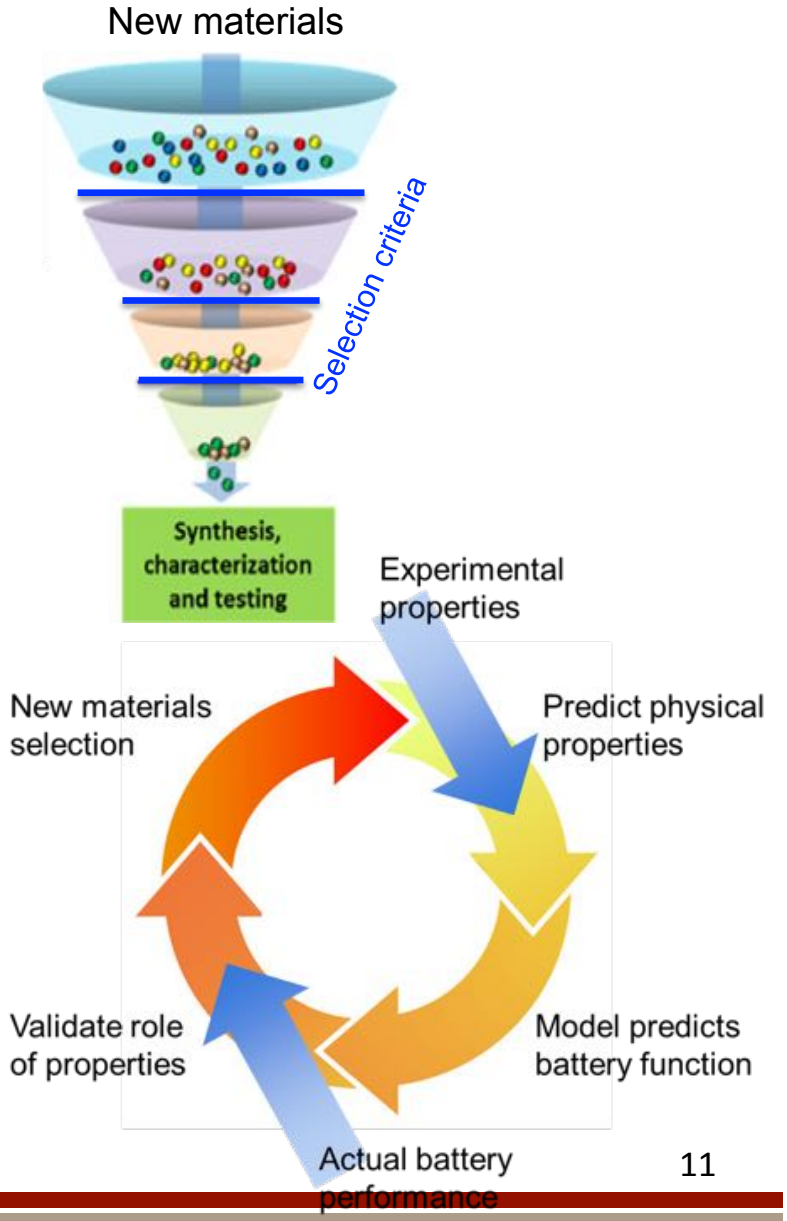
Electrolyte composition has greatest impact on cell function

Modeling and experimental feedback

- Predict actual battery function
  - Self-consistency between experimental inputs and validation
- Use voltammetry as a tool for validation

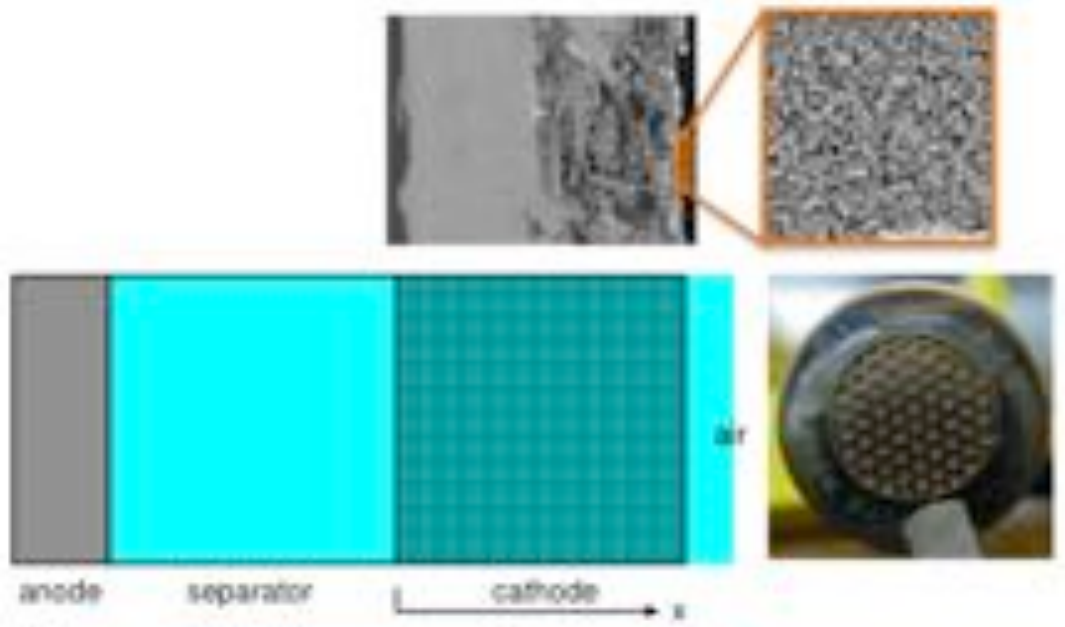


- Extract:
  - Diffusion coefficients of mobile species
  - Local O_2 concentration
  - Role of electrode geometry
  - Influence of electrolyte composition
    - Solvent, anion, salt concentration etc.



Basics of the model

- Assumptions
  - Irreversible electron transfer
    - Secondary reactions are fas
  - Flooded porous electrode
  - Metallic-like electrode conduction
  - No cross-species diffusion
  - Average concentration/ diffusion of O_2 is one dimensional
- Fundamental foundation
  - Butler-Volmer kinetics
  - Reaction-diffusion equation



Oxygen Reduction (Discharge)

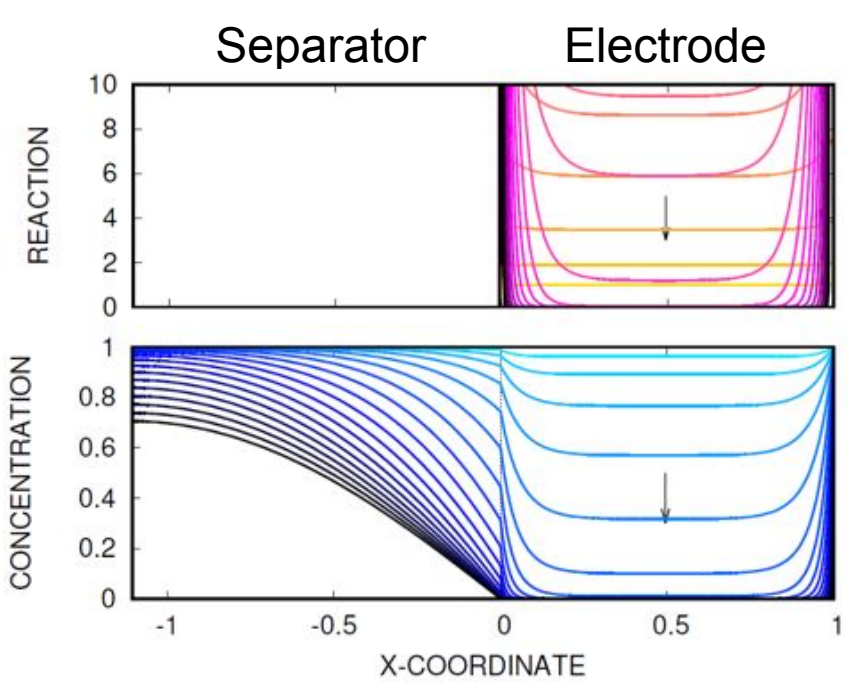
(1) $O_2 + e^- \rightarrow O_2^-$ (rate limiting)

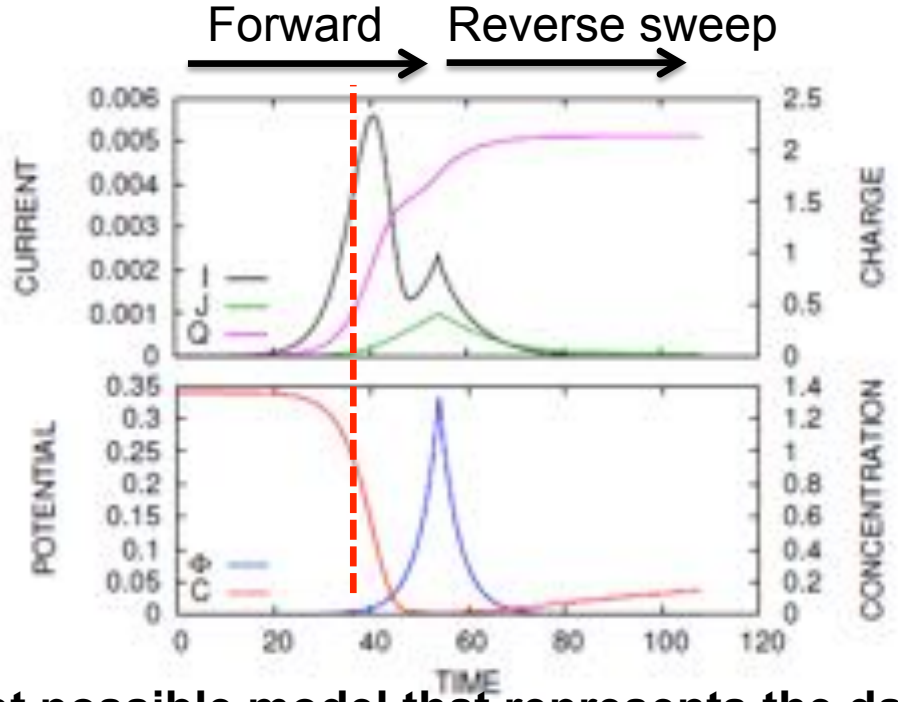
(2) $O_2^- + Li^+ \rightarrow LiO_2$

(3a) $2LiO_2 \rightarrow Li_2O_2 + O_2$

(3b) $LiO_2 + Li^+ + e^- \rightarrow Li_2O_2$

Diffusion and reaction dynamics





Simplest possible model that represents the data

- Oxygen is depleted, replaced by diffusion from separator and air interface
  - Reaction profile peaks at ends of electrode
- I varies with average of concentration across electrode
- Oxygen depletion → voltammetric peak current
- Bulk oxygen diffusion → secondary peak/tail
  - At long times, diffusion rate dominates