SAND2013-6798C

Joint Center for Energy Storage Research Activities at Sandia National Laboratories

Kevin R. Zavadil, Sandia National laboratories

Challenges in Energy Storage Symposium, Northeastern University
(August 13-14/2013)

Messages

The Opportunity: Transportation and the Electricity Grid

The Outcomes: 5-5-5 and Three Legacies

Beyond Lithium Ion

JCESR Distinguishing Features

Embracing DOE's Discovery to Deployment Spectrum

Highlights

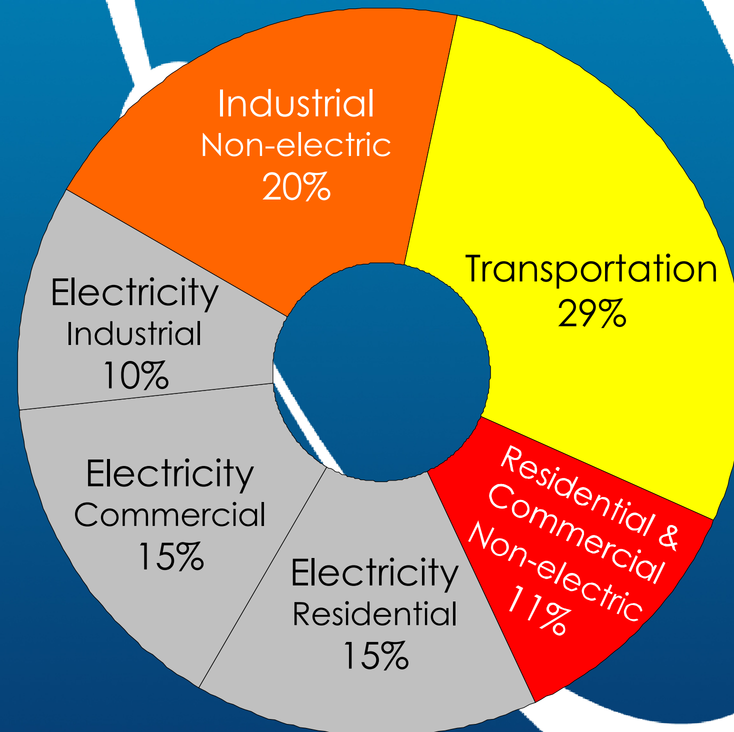
The Battery Development Ch llenge

Two biggest energy uses poised for transformational change

Transportation 29%
- Oil → Electricity
- Reduce foreign oil dependence
- Reduce carbon emissions

E tricity 40%
- oal → Gas → Wind and Solar
- luce carbon emissions
- S inable energy supply

Energy Demand



EIA Annual Energy Review 2009

The bottleneck for both transitions is
expensive, high density electrical energy storage

JCESR

JCESR Targeted Outcomes

ACHIEVING GOALS FOR LASTING LEGACIES

TRANSPORTATION

$100/kWh

400 Wh/kg **400** Wh/L

800 W/kg **800** W/L

1000 cycles

80% DoD C/5

15 yr calendar life

EUCAR

GRID

$100/kWh

95% round-trip efficiency at C/5 rate

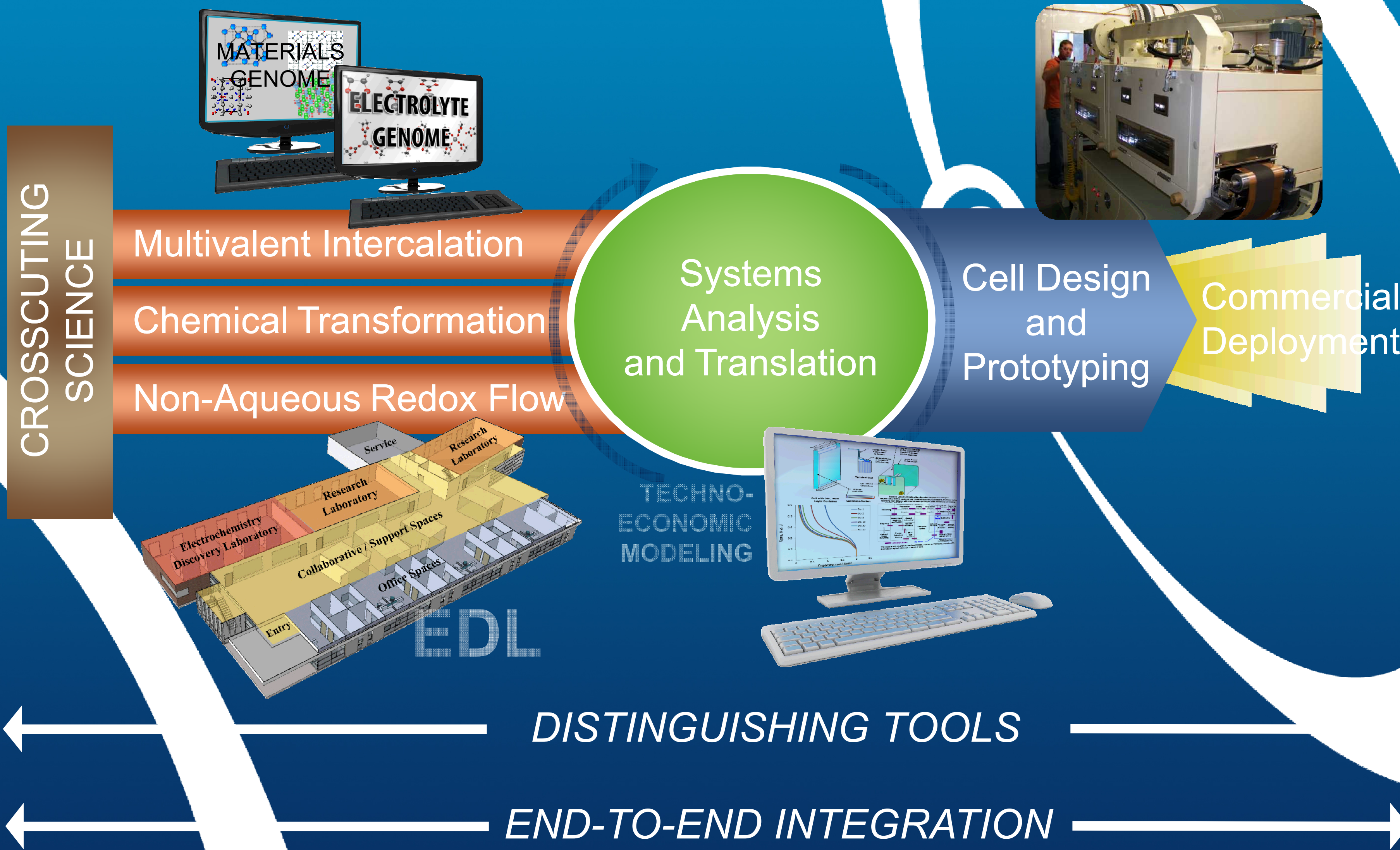
7000 cycles C/5

20 yr calendar life

Safety equivalent to a natural gas turbine

- Transformational goals: 5-5-5
  - 5 times greater energy density
  - 1/5 cost
  - within 5 years
- Legacies
  - Library of fundamental knowledge
    - *Atomic and molecular understanding of battery phenomena*
  - Pre-commercial prototypes for grid and transportation
  - New paradigm of battery development
    - *Build the battery from the bottom up*
    - *Systems-centric*
    - *End-to-end integration*

The JCESR Paradigm: Beyond Lithium Ion



The JCESR Team

5
National Laboratories

Argonne
Lawrence Berkeley
Sandia
Pacific Northwest
SLAC

5
Jniversities

U... sity of Illinois at ...hicago
Unive... of Chicago
Northwe... University
Universit... linois at Urbana-... paign
University ... higan

[illegible]
Private S... r Partner...

Dow
JCI
Applied Materials
Clean Energy Trust

JCESR Affiliates

CENTRALIZING FORCE FOR BATTERY FIELD

~ 40 Organizations

Communicating

and

Collaborating

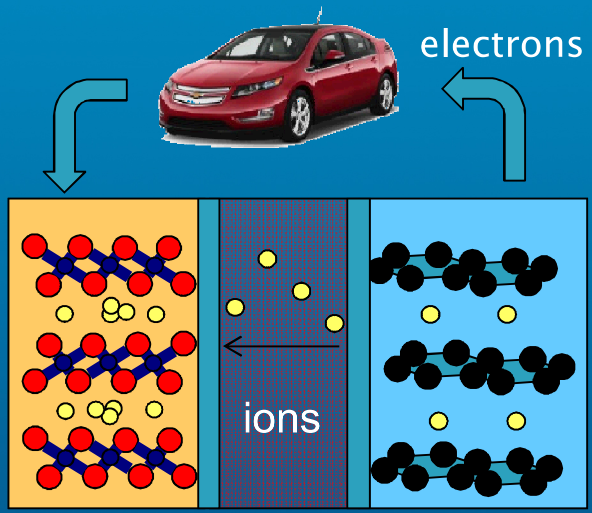
with the

Energy Storage

Community

Beyond Li-ion Storage Concepts

CROSSCUTTING SCIENCE
- Multivalent Intercalation
- Chemical Transformation
- Non-Aqueous Redox Flow



Multivalent Intercalation

- Li^{+} → Mg^{++}, Y^{+++}, . . .

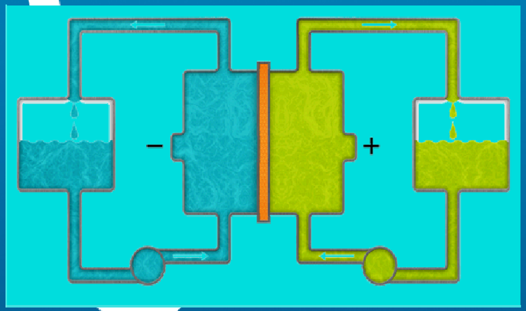
le or triple energy stored and released

Chemical Transformation

Inte ation → chemical reaction

i-O_2, Li-S, Na-S, . . .

All d store and release energy



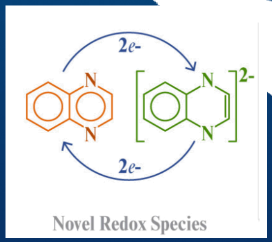
Non-aqueous Redox Flow

Flowable electrodes

- solutions or suspensions
- no structural constraints
- rich horizon of unexplored redox couples

Low cost / high capacity

organic materials

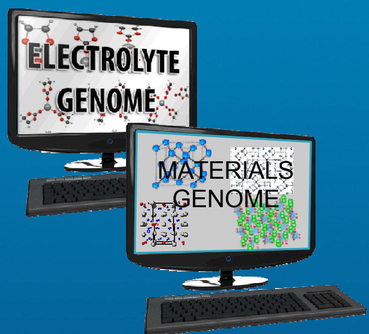


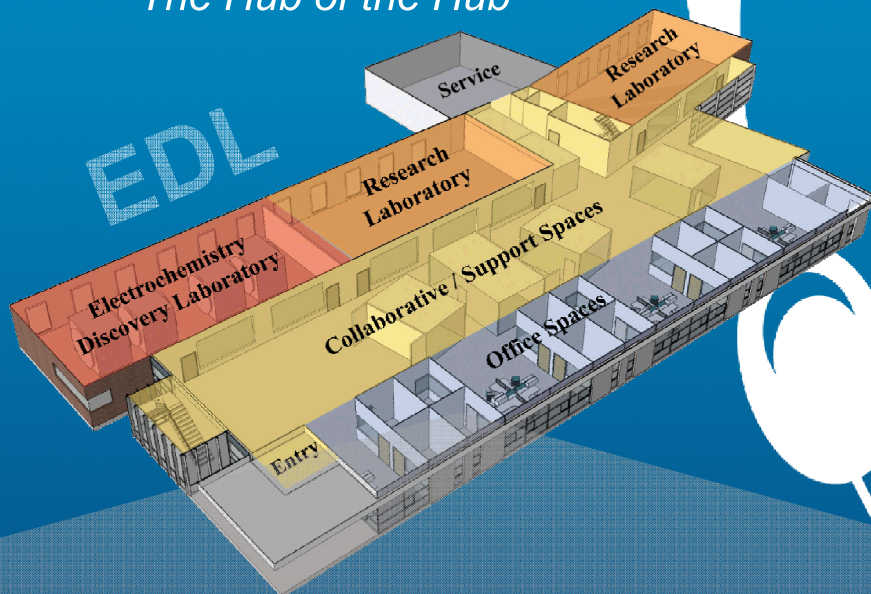
Highest potential, least understood opportunities

The Electrochemical Discovery Laboratory

The Hub of the Hub

CROSSCUTTING SCIENCE
- Multivalent Intercalation
- Chemical Transformation
- Non-Aqueous Redox Flow

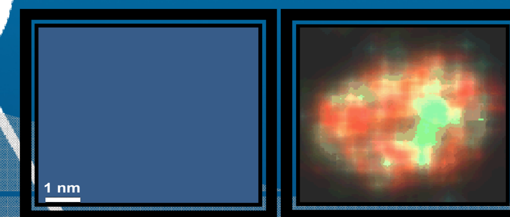




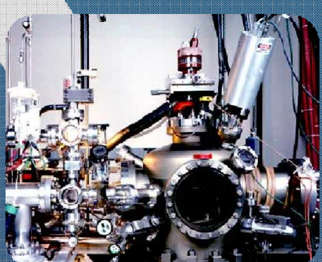
Cell Design and Proto

Techno-economic Modeling

COMPOSITE SYSTEMS

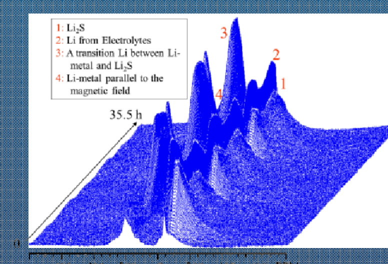


SYNTHESIS



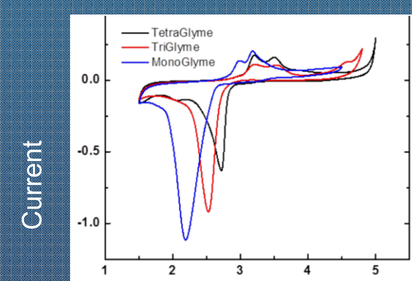
THIN FILM
SINGLE CRYSTAL
MBE•CVD•PVD

SPECTROSCOPY



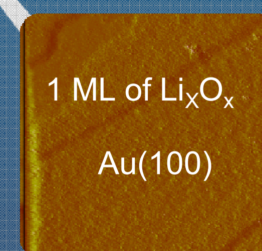
XPS•LEED•FTIR•UPS•AES
•RAMAN•SFG•NMR

ELECTROCHEMISTRY



RDE•RRDE

SURFACES•INTERFACES

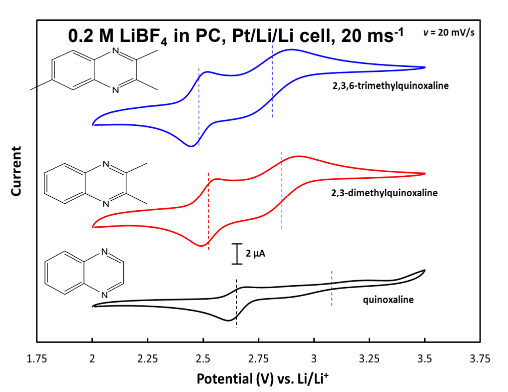


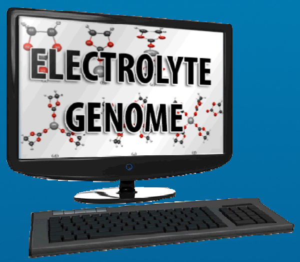
STM•AFM

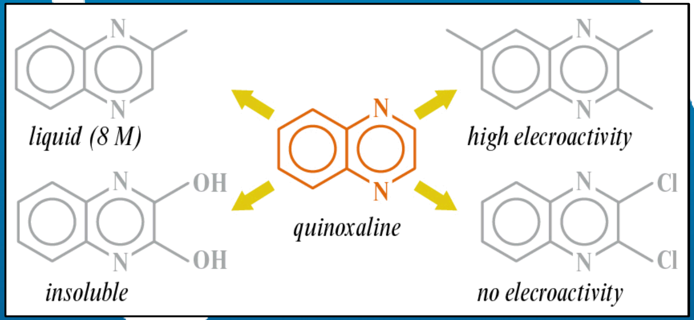
Building an Electrolyte Genom

A new horizon for designing novel electrolytes and redox-active olecules

CROSSCUTTING SCIENCE
- Multivalent Intercalation
- Chemical Transformation
- Non-Aqueous Redox Flow

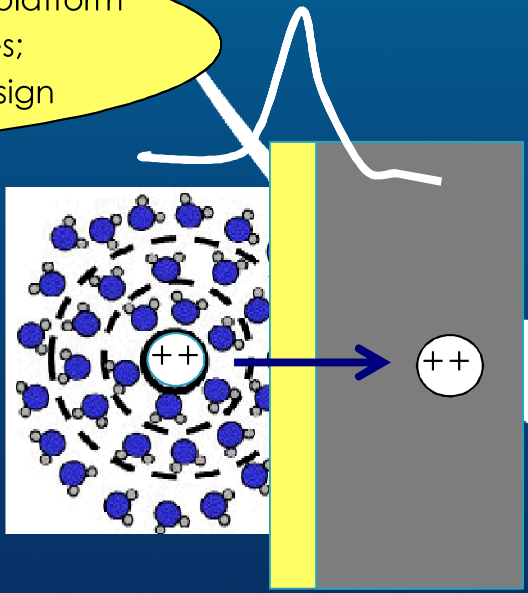




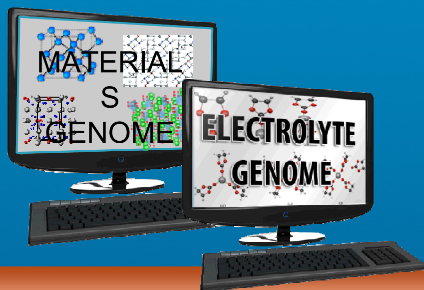


Computational structure/composition/property platform
10^4-10^5 solvents, salts, and redox molecules;
organized for interactive searching and design

- edox activity
- bility against cathode / anode
- ation structure and mobility
- So ion / desolvation dynamics
- Sol y
- Ene torage capacity



The JCESR Paradigm: Beyond Lithium Ion



CROSSCUTTING SCIENCE

Multivalent Intercalation

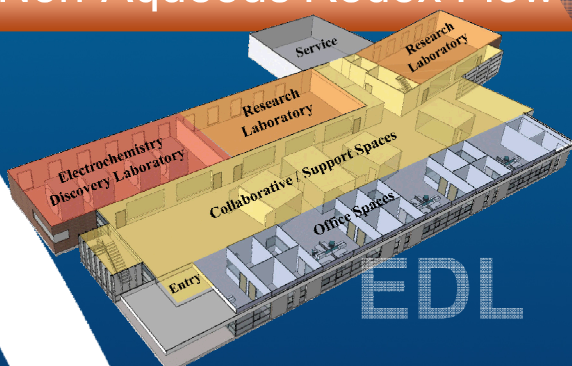
Chemical Transformation

Non-Aqueous Redox Flow

Systems Analysis and Translation

Cell Design and Prototyping

Commercial Deployment



EDL

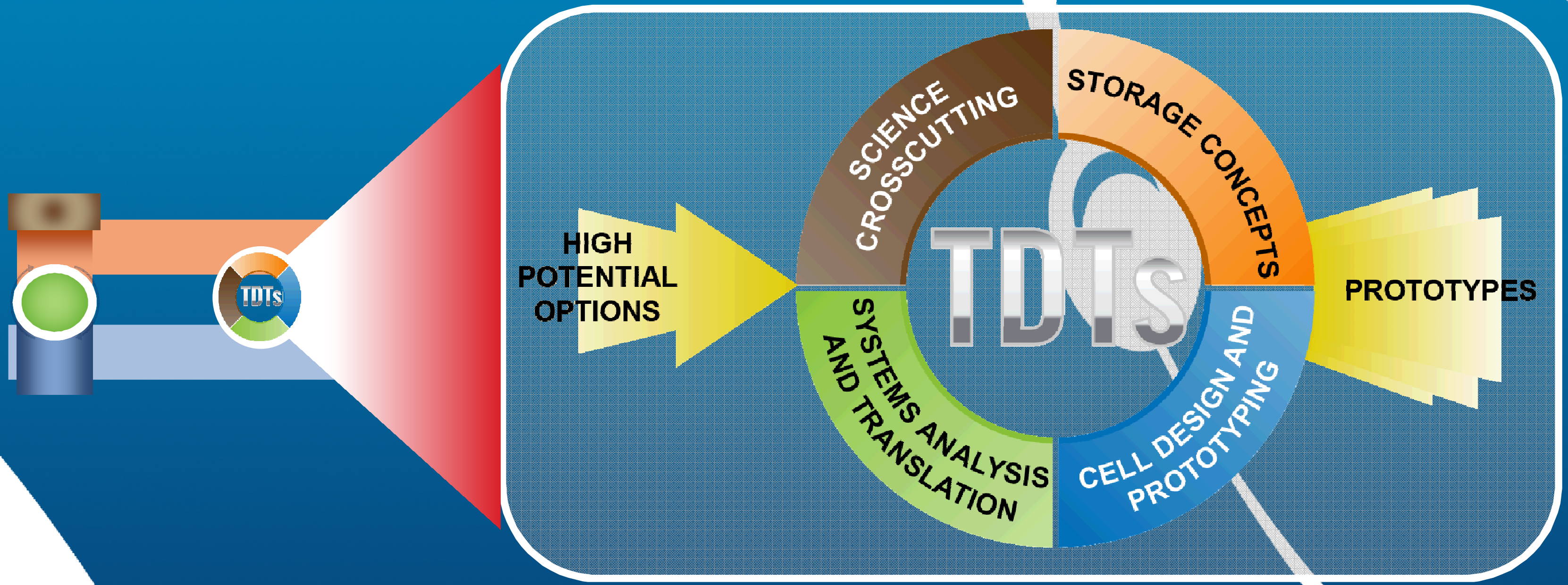
TECHNO-ECONOMIC MODELING

DISTINGUISHING TOOLS

END-TO-END INTEGRATION

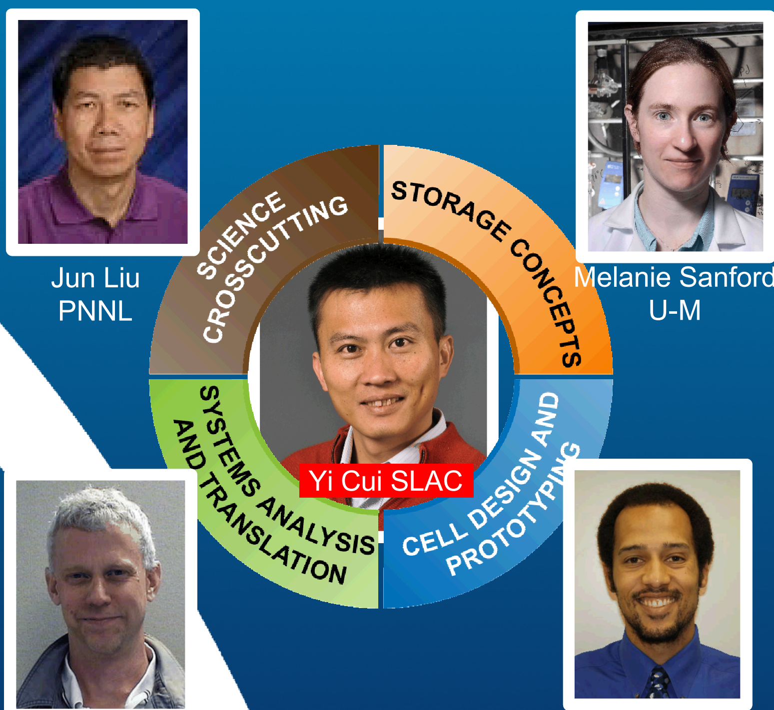
Translational Development Teams

INTEGRATING SCIENCE WITH ENGINEERING



- We kickoff two TDTs on Day 1 to design and prototype cells
  - Transportation – Mg Intercalation
  - Grid – Non-aqueous redox flow

TDT 1
Non-aqueous Redox Flow
$100/KWH, 95% EFFICIENCY AT C/5



TDT 2
Mg intercalation
$100/kWh, 400 Wh/kg and 400 Wh/L

Our Multi-Disciplinary Teams

JCESR: Reaching Across the Discovery-Desig Demonstration S trum

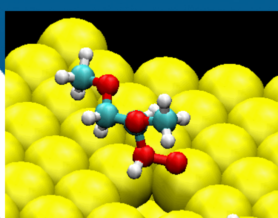
CROSSCUTTING SCIENCE

- Multivalent Intercalation
- Chemical Transformation
- Non-Aqueous Redox Flow

Systems Analysis and Translation

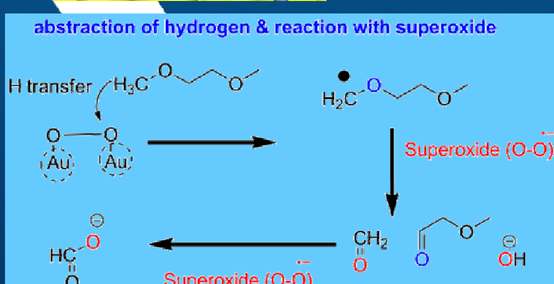
Cell Design and Prototyping

Commercia Deployment

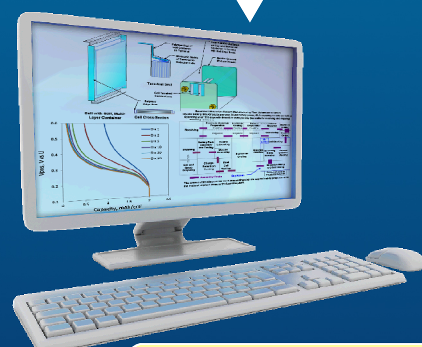


Electrochemical Discovery Lab

RDE, RRDE, FTIR, DFT



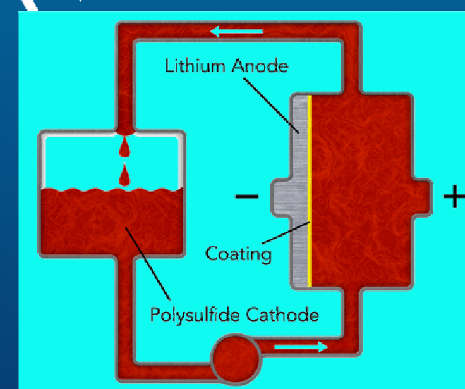
Reaction seq and active sites for electrolyt gen interaction



- Cell and system design
- Performance
- Cost

Building Li-air battery on the computer

(with GM)



Yang, Zheng, Cui DOI: 10

Non-aqueous Li-Polysul semi-flow battery

X10 increase in flow battery energy density

2000 cycles

Chemical Transformation

$S_2 + 4\ Li^+/e^- \leftrightarrow 2Li_2S$

600 Wh/kg (3X), ↑ achievable

Electrolyte Genome

$O_2 + 2\ Li^+/e^- \leftrightarrow Li_2O_2$

5X: quantify achievable limits

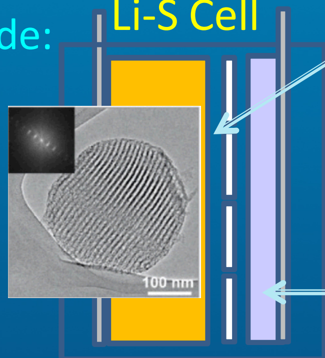
Positive Electrode:
New materials

- sulfur sequestration
- Li_2S cathodes

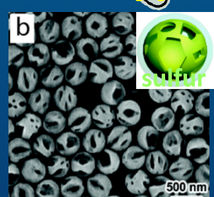
Mechanistic understanding
…mine -

- …eciation
- LPS… sport in nanos… red membra…

Li-S Cell



NRF Thrust Prototype

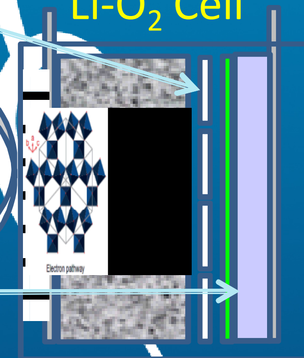


Electrolyte:
Control of dissolution-ppt

- solvent stability
- tuning solubility

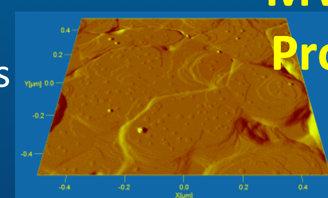
O_2

Li-O_2 Cell



Negative Electrode:
Manipulate electrodeposition

- additives for passivation
- interface studies
- Li informs Mg & MV's



MV Thrust Prototype

Positive Electrode:
New materials

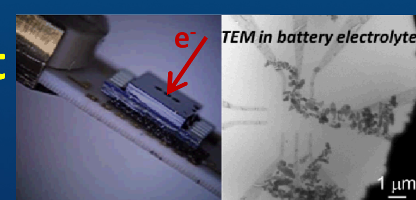
- catalysts for ORR
- porous materials
- new architectures

Mechanistic understanding:
Determine -

- limits of O_2 reversibility
- …eaction pathway an… lectrolyte role
- elect…

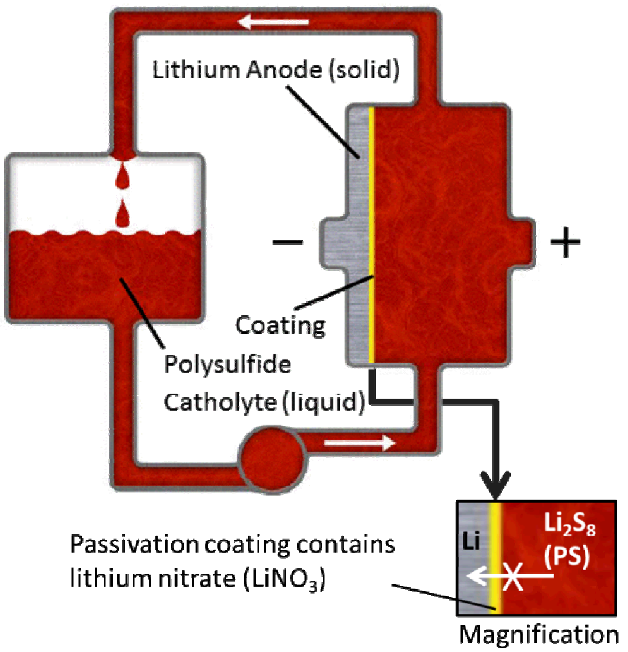
Cross Cutting Thrust

- computation
- *operando* probes (TEM, NMR, X-ray)



Create Library of Knowledge • Establish Rational Design • Enable Prototypes

Lithium/Polysulfide Battery for Large-scale Energy Storage



Design principle for Li/PS battery: Polysulfides (PS) are pumped through the system during operation and are stored in the tank in downtime. The passivation coating (magnification) is formed by exposing the Li to lithium nitrate and prevents the undesirable reaction between PS and the Li.

Work was performed at SLAC National Accelerator Laboratory (JCESR partner).

Y. Yang, G. Zheng and Y. Cui, *Energy Environ. Sci.*, **2013**, online edition, DOI: 10.1039/c3ee00072a

Scientific Achievement

Tailored the electrochemical properties of a novel lithium (Li)/polysulfide (PS) semi-liquid battery with high energy density and discovered a method to eliminate the need for a separator membrane.

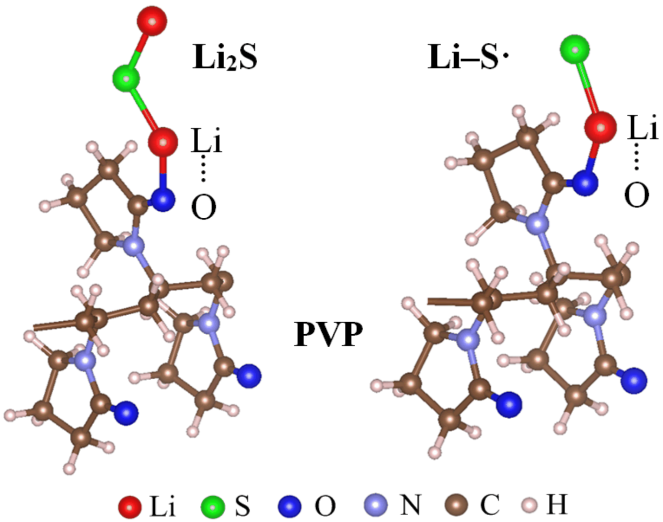
Significance and Impact

Research prototype design could lead to low-cost, long-life batteries that facilitate the large-scale use of solar and wind energy on the electrical grid.

Research Details

- Working research prototypes of the hybrid flow battery were made with the polysufide (Li_2S_8) in ether-based solvent as the liquid catholyte, and passivated metallic Li as the solid anode.
- The anode is treated with lithium nitrate to form a passivation layer that prevents direct chemical reaction between the Li anode and PS catholyte, eliminating the need for a membrane.
- Energy density of the system reaches 108 Wh/liter - three times that of the conventional vanadium flow battery - with a discharge voltage of 2.45 V.
- By keeping the charge/discharge voltage range narrow, all phases remain dissolved in the liquid, reducing degradation.

Rational Design of High-Performance Li_2S Cathodes



Using *ab initio* simulations, poly(vinylpyrrolidone) (PVP) binder was found to possess strong affinity for both Li_2S and lithium polysulfides. This bifunctional binder not only helps to form a uniform dispersion of active material and carbon in the electrode slurry, but also minimizes the loss of polysulfides into the electrolyte during cycling.

Work was performed at SLAC National Accelerator Laboratory (JCESR partner).

Z. W. Seh, Q. Zhang, W. Li, G. Zheng, H. Yao, Y. Cui, *Chem. Sci.* **2013**, DOI: 10.1039/c3sc51476e.

Scientific Achievement

Achieved record performance in Li_2S cathodes by using *ab initio* simulations to guide our rational selection of effective bifunctional binders.

Significance and Impact

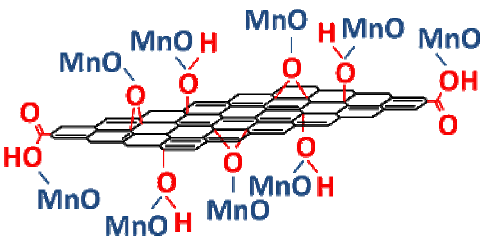
Rational materials design could lead to low-cost and high-performance Li_2S cathodes that can be paired with safer, lithium metal-free anodes for use in emerging applications such as vehicle electrification and grid-scale energy storage.

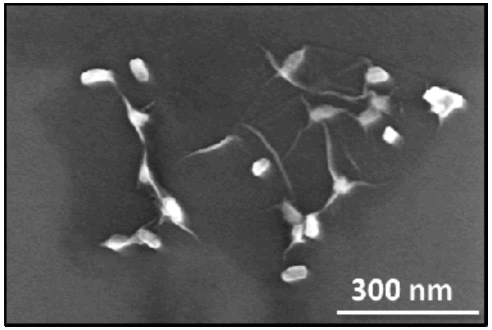
Research Details

- A rational approach was taken by first using *ab initio* simulations to elucidate the poorly-understood interaction between Li_2S and various functional groups found in macromolecular binders.
- Poly(vinylpyrrolidone) (PVP) was then selected as a promising bifunctional binder which exhibits strong affinity with both Li_2S and lithium polysulfides.
- Using PVP as a binder for Li_2S cathodes, we demonstrate unprecedented stable cycling performance over prolonged 500 charge/discharge cycles.

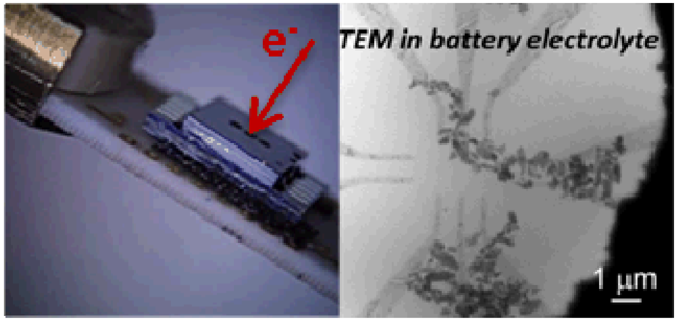
Research Activities at Sandia National Laboratories

materials

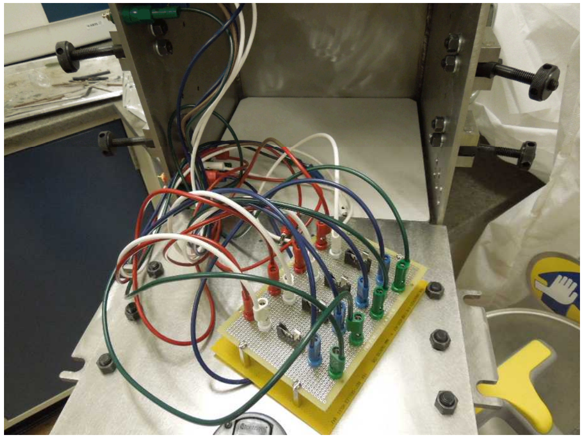




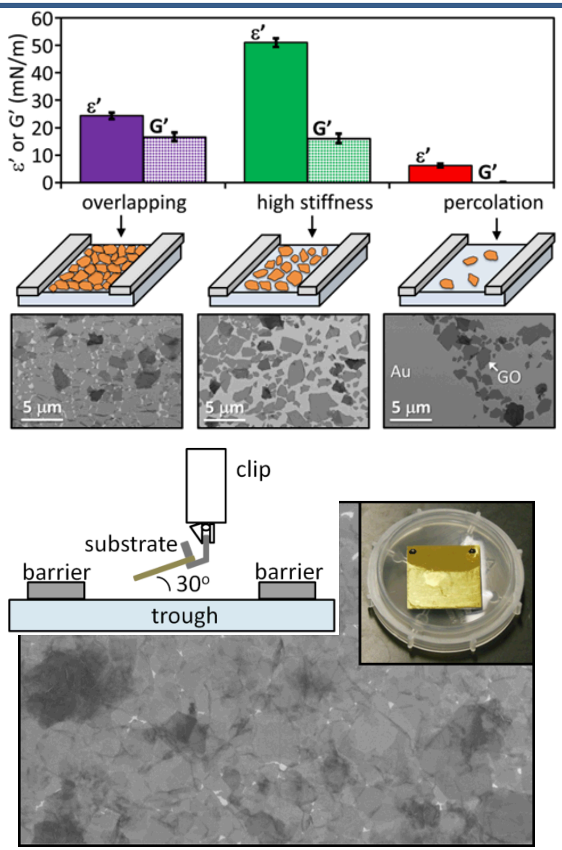
Fundamental studies & diagnostics



cell development & prototyping



Controlled Graphene-based Electrode Fabrication: Mechanical Properties of Water-Assembled Graphene Oxide Monolayers



GO monolayers at the water/air interface behave like a 2D solid. Acute angle dipping is used to preserve the 2D solid resulting in the transfer of a continuous GO film on *hydrophobic* Au.

K.L. Harrison, L.B . Biedermann, K.R. Zavadil, ACS Nano, submitted.

Scientific Achievement

Quantifying the mechanical properties of graphene oxide (GO) monolayers leads to a demonstrated ability to control its transfer to a wide variety of surfaces.

Significance and Impact

Graphene is an important building block material for electrochemical-based energy storage but its properties and therefore impact cannot be fully realized without simple strategies for its controlled assembly into coherent electrodes.

Research Details

– Liquid phase transfer using a Langmuir-Blodgett approach is pursued as a means of controlled self assembly.

– Oscillatory barrier measurements are used to probe the mechanical properties of the GO monolayer as a function of surface pressure (Π). A shear modulus indicates the GO forms a 2D solid and precautions must be taken to prevent solid rupture during transfer. Acute angle dips prevent rupture and allow transfer of continuous films independent of substrate.

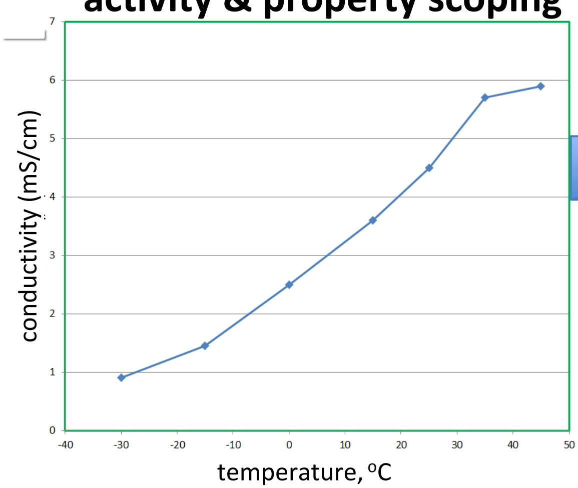
Work was performed at Sandia National Laboratories (JCESR partner)

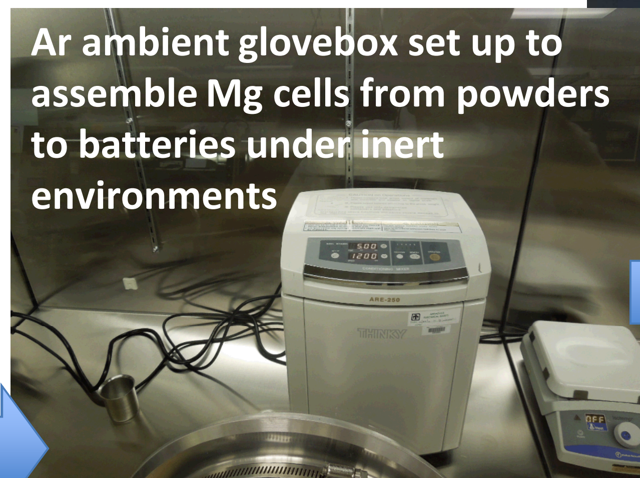
Cell Development and Prototyping (CDP) of Multivalent Ion Chemistries – C. Apblett

Early stage CDP activities serve several key functions - establish:

- **benchmark cell chemistry**
- **process flow**
- **synthesis – end use chain**
- **compatibility and stability**

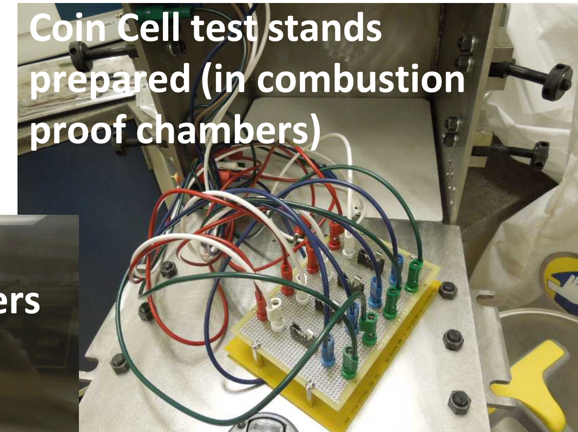
activity & property scoping





Ar ambient glovebox set up to assemble Mg cells from powders to batteries under inert environments

materials synthesis and assembly

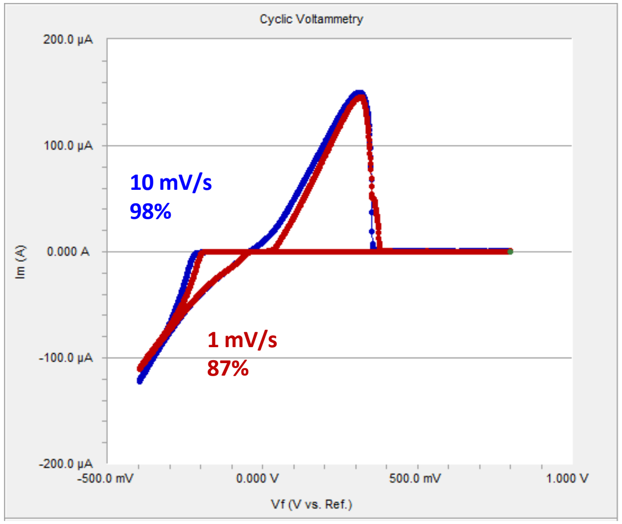


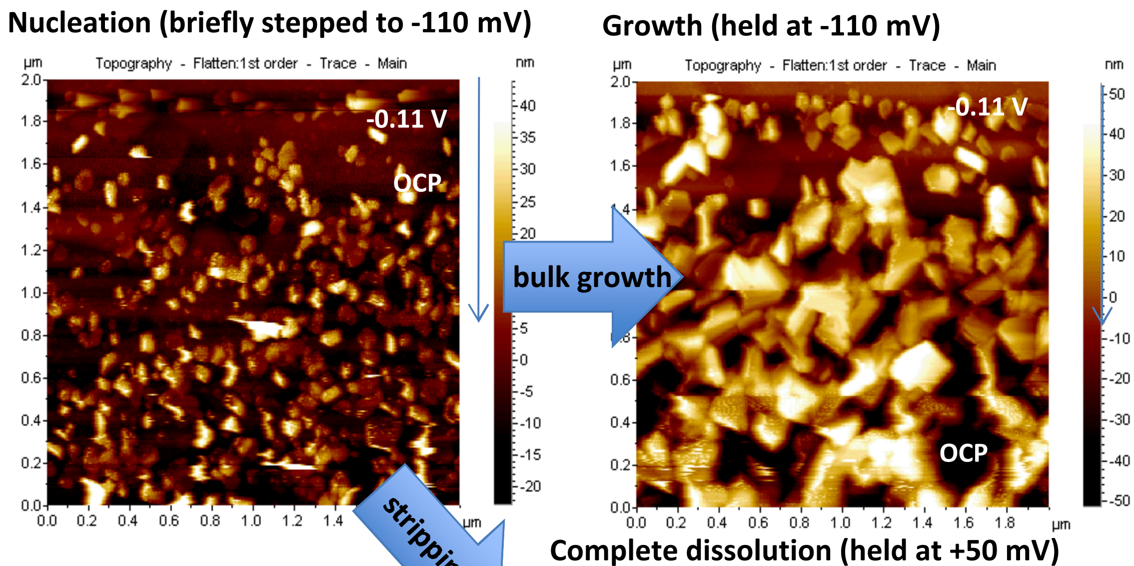
Coin Cell test stands prepared (in combustion proof chambers)

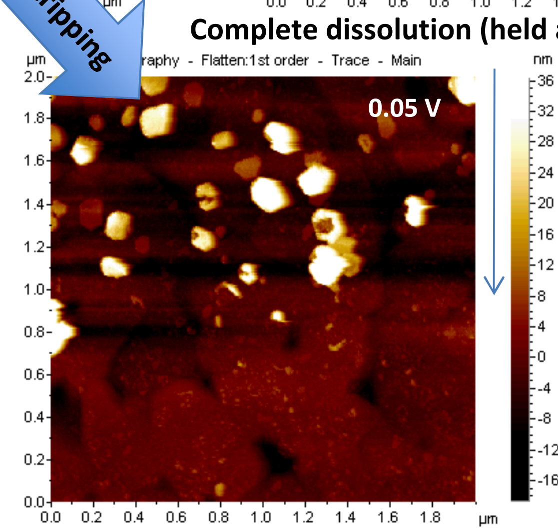
performance testing

Environmental chambers set up for temperature experiments

Multivalent Metal Cation Accommodation at the Anode: Origins of Efficiency and Cycleability – K. Zavadil







In situ metrology provides direct measurement of critical phenomenon:

- **morphology evolution and rate dependence**
- **balancing metastability and passivation**
- **solid electrolyte interphases**
- **partitioning at a host:electrolyte interface**

Dynamics of Li_2O_2 Formation & Spatial Distribution in the Li-O_2 System – K. Zavadil

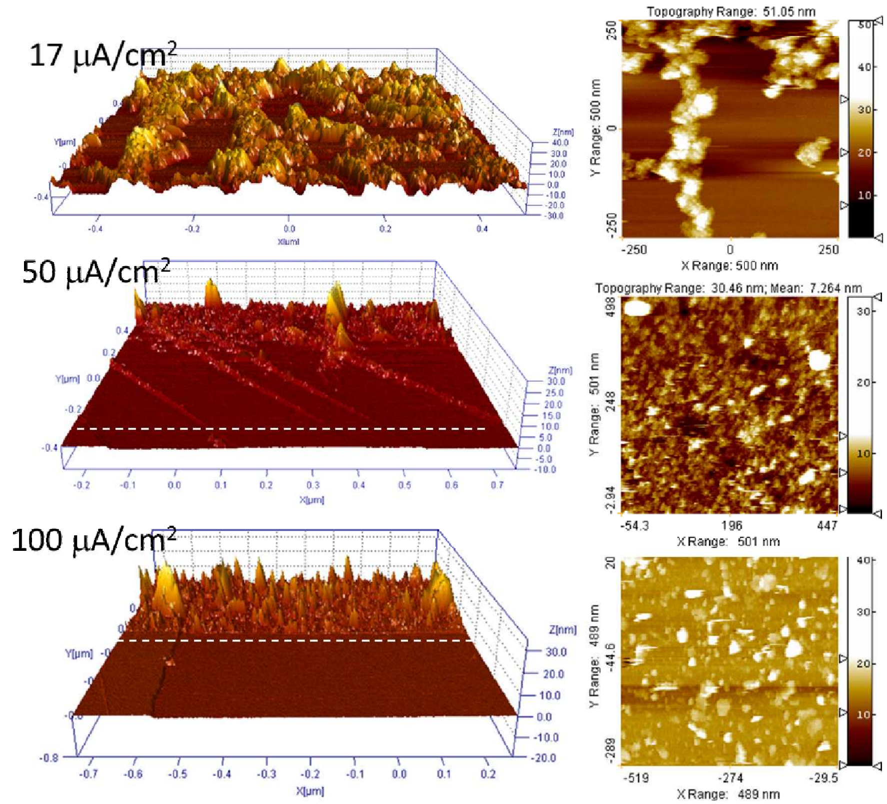
Alkali metal – Oxygen electrochemistry

- **promise of high theoretical energy density e.g. $2Li + O_2 \leftrightarrows Li_2O_2$**
- **system level energy density still shows promise**
- **O_2 is an attractive reporter molecule to gain new insight into energy storage redox phenomena**

Operando studies allow determination of critical processes:

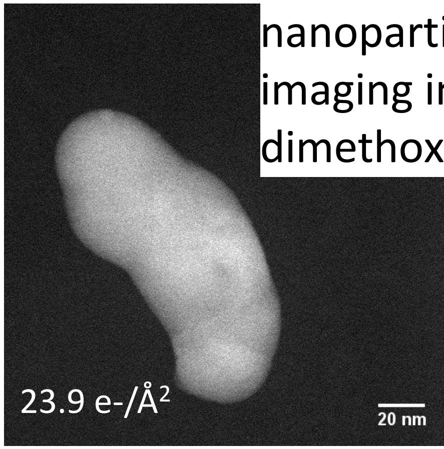
- **solvent mediation of nucleation and growth**
- **proximity relationships**
- **electrocatalyst role**

peroxide nucleation and growth on graphite

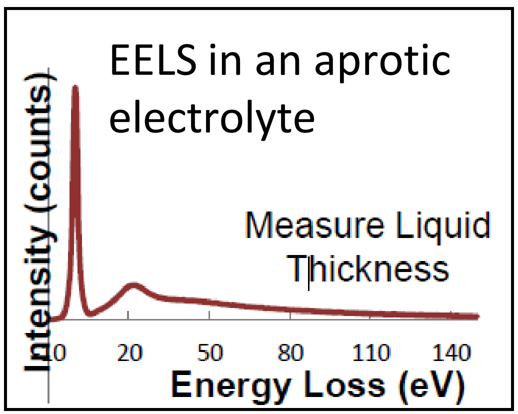


Visualization of Structural and Interfacial Changes with Energy Storage Processes – K. Jungjohann

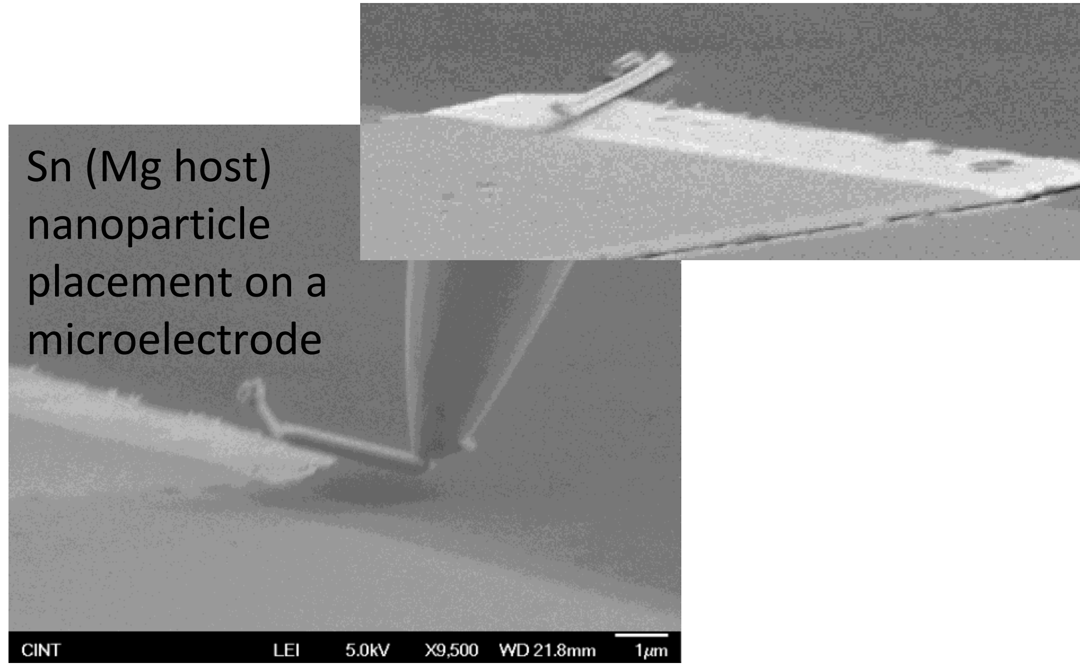
nanoparticle imaging in dimethoxyethane



EELS in an aprotic electrolyte



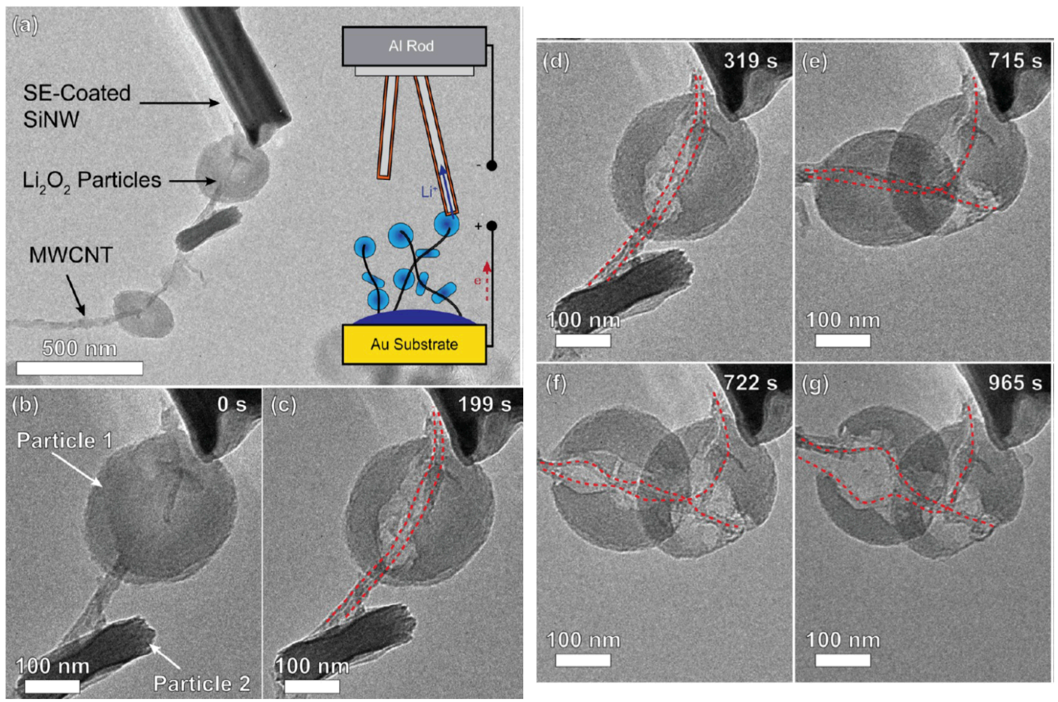
Sn (Mg host) nanoparticle placement on a microelectrode



Must understand and account for radiolysis damage during STEM imaging

Electrochemical TEM for Tracking Energy Storage Phenomena – a Sandia Area of Expertise

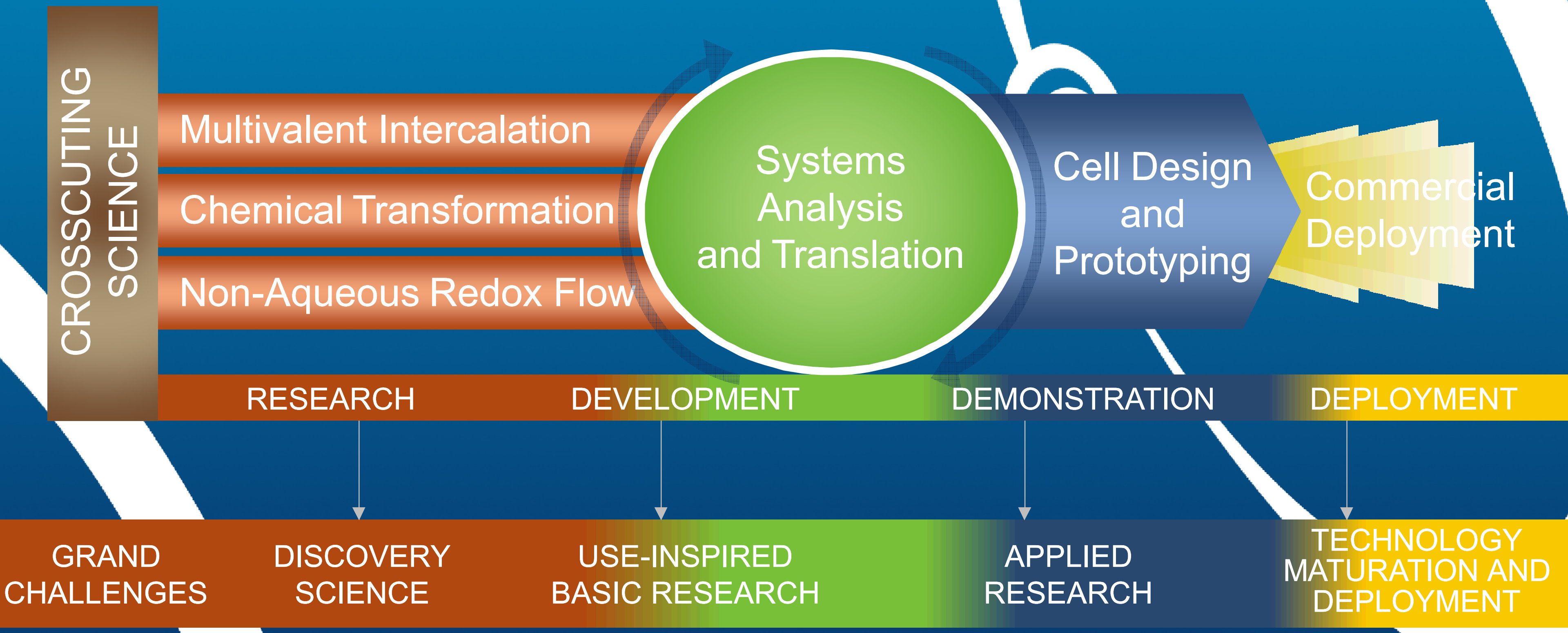
Solid state discharge studies as a precursor to operando measurement in an electrolyte



Shao Horn et al. Nano Lett 2013 (Sandia collaboration)

JCESR

JCESR Maps onto DOE's RDD&D Spectrum



Embracing DOE's Discovery to Deployment Spectrum

Ultimate Battery May Combine Storage Concepts

Multivalent metal anode

High mobility in stable electrolyte

Chemical reaction with flowable cathode