

An aerial photograph of a university campus. In the upper left, there is a large, circular, tiered seating area, possibly a stadium or amphitheater. The campus is filled with various academic and administrative buildings, some with flat roofs and others with more complex structures. There are several large parking lots with many cars. The campus is surrounded by dense green trees and grassy areas. In the lower right, there are some industrial-looking structures, possibly water towers or storage tanks. The overall scene is a mix of urban development and natural landscape.

# Battery Research Overview

Jeff Chamberlain

March 20, 2013

# The national need for energy storage is driven by:

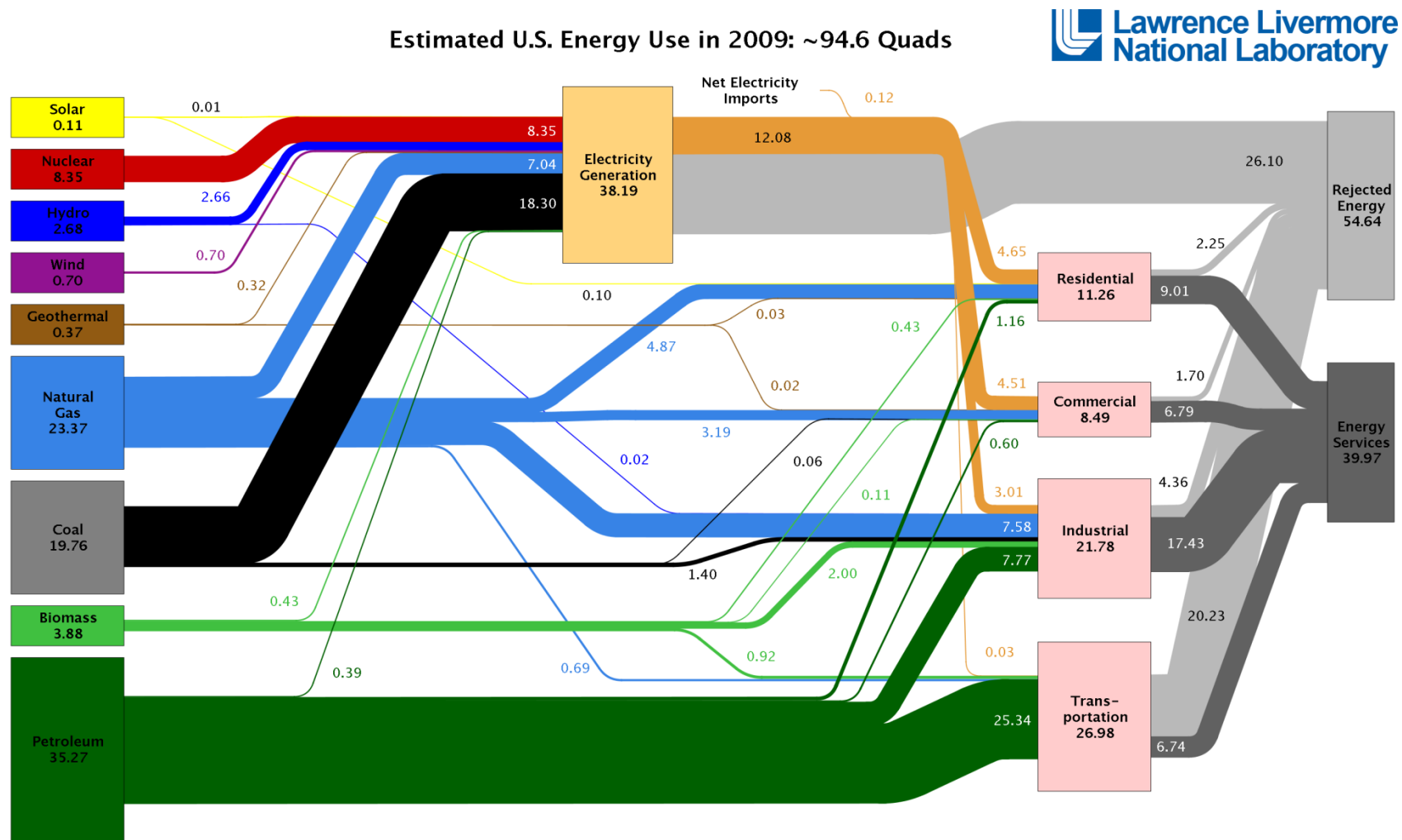
- Security
- Economy
- Environment

The projected doubling of world energy consumption in 50 years.

A growing demand for low- or zero-emission energy sources.

Part of the solution entails the transformation of our transportation and stationary storage technologies...

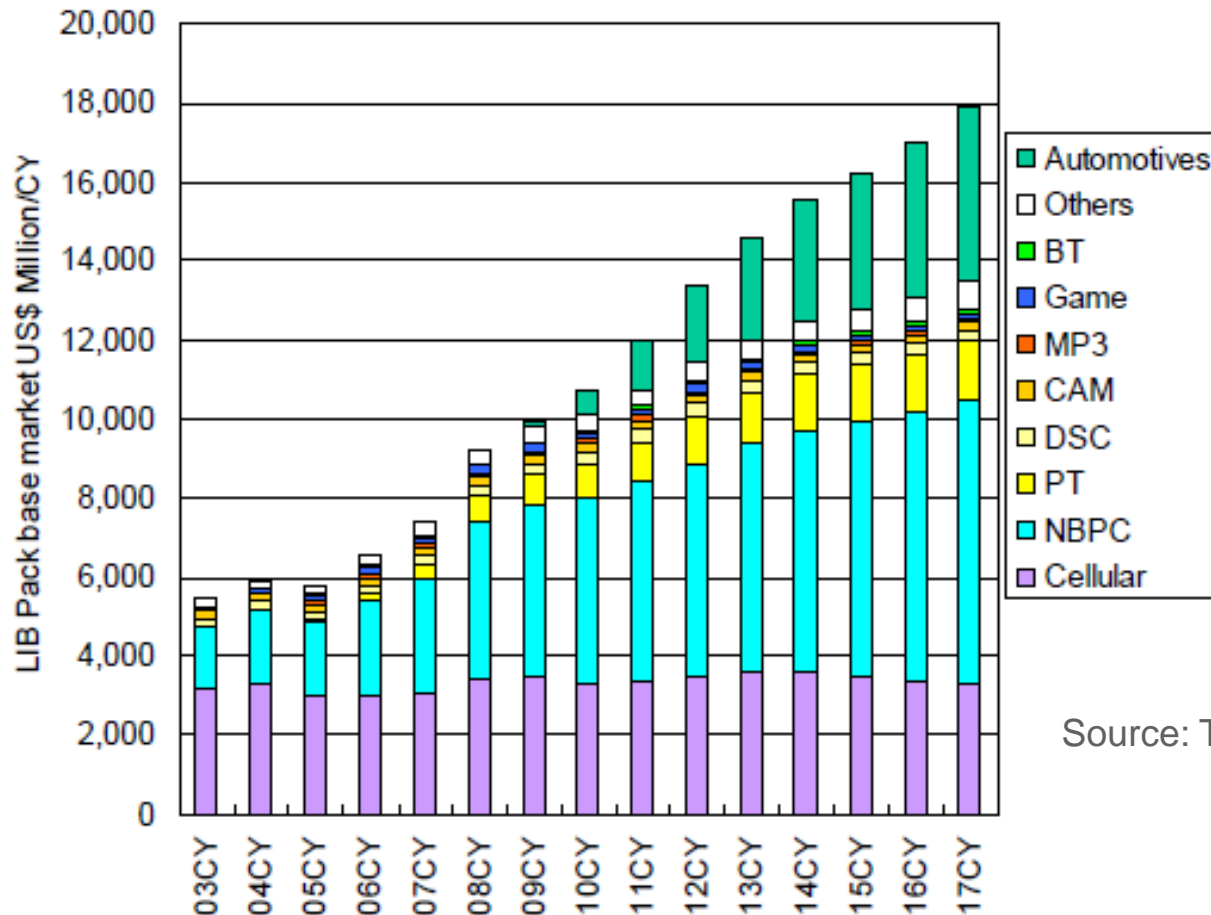
# Energy flow chart shows relative size of primary energy resources and end uses in U.S.



Source: LLNL 2010. Data is based on DOE/EIA-0384(2009), August 2010. If this information or a reproduction of it is used, credit must be given to the Lawrence Livermore National Laboratory and the Department of Energy, under whose auspices the work was performed. Distributed electricity represents only retail electricity sales and does not include self-generation. EIA reports flows for non-thermal resources (i.e., hydro, wind and solar) in BTU-equivalent values by assuming a typical fossil fuel plant "heat rate." The efficiency of electricity production is calculated as the total retail electricity delivered divided by the primary energy input into electricity generation. End use efficiency is estimated as 80% for the residential, commercial and industrial sectors, and as 25% for the transportation sector. Totals may not equal sum of components due to independent rounding. LLNL-MI-410527



# Economic Drivers are Enormous: transportation



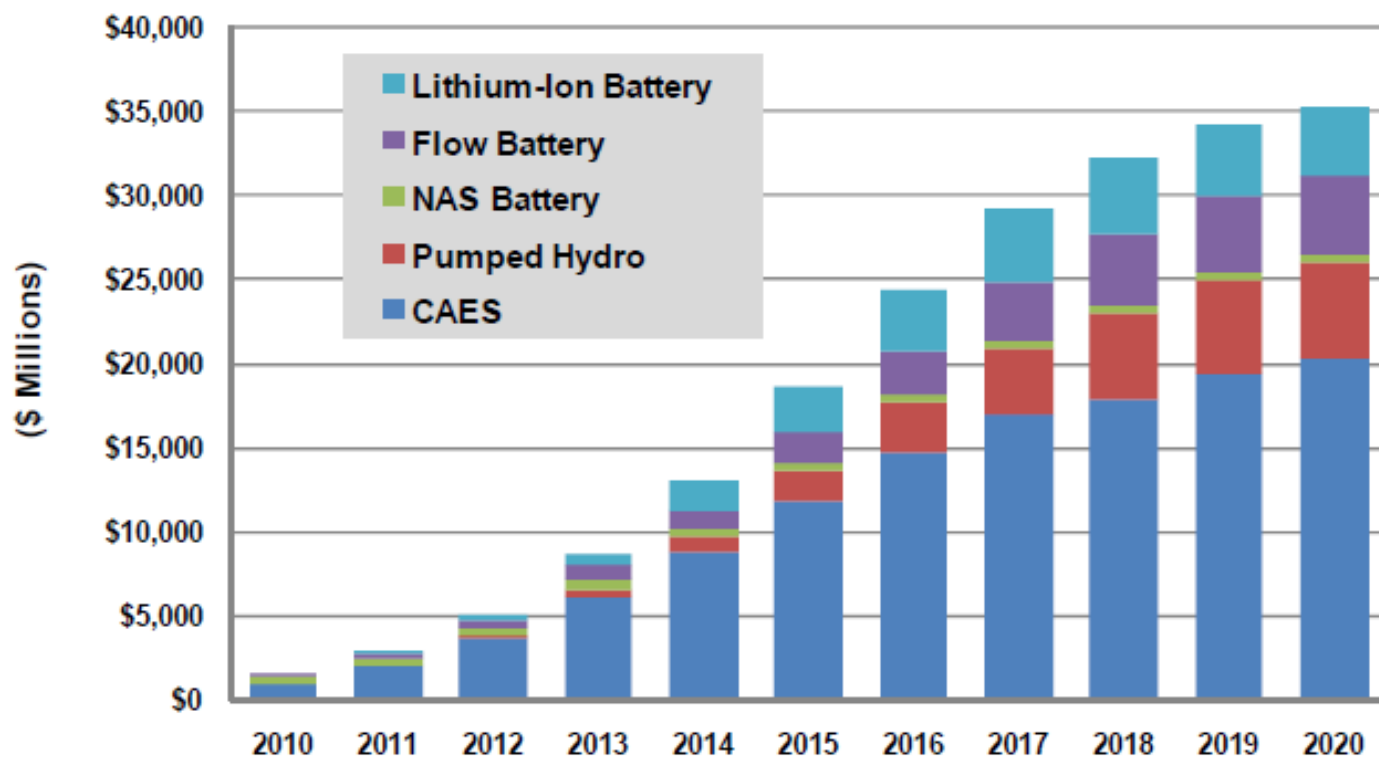
Source: Takeshita Report, 2008

- 5% penetration of PHEVs = \$18B in annual revenue, for battery packs alone (assuming current estimates of \$7500/pack)



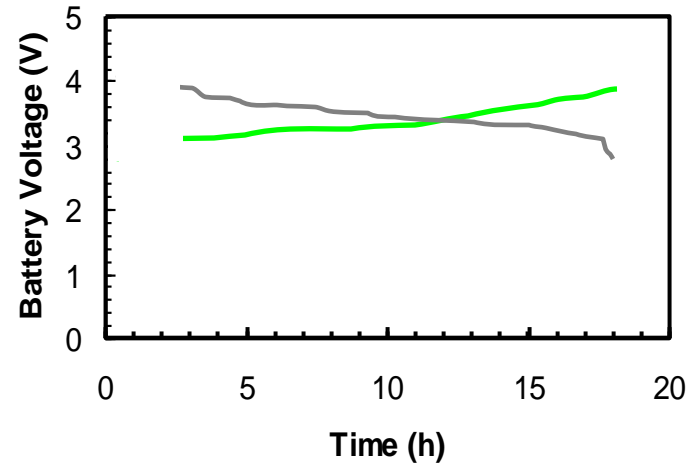
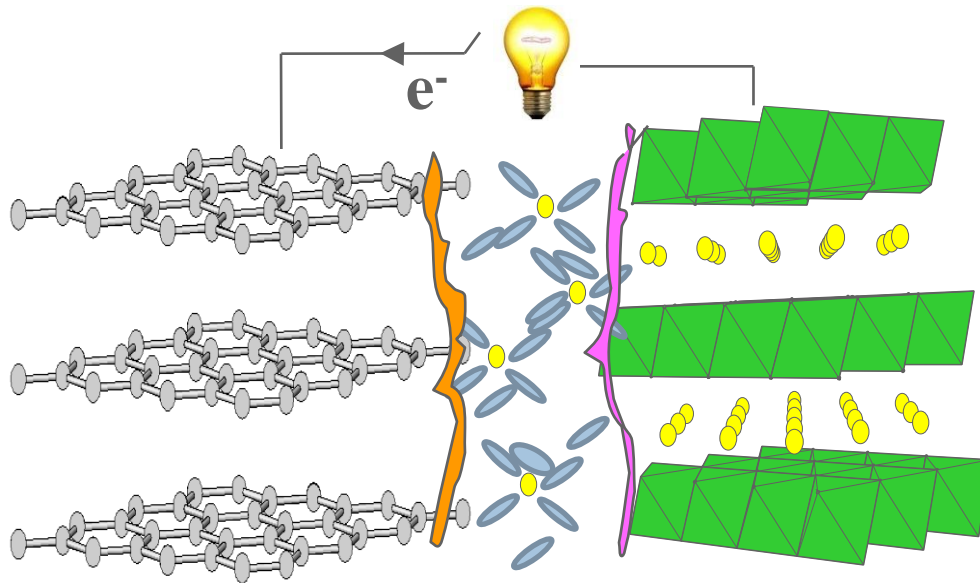
# Economic Drivers are Enormous: grid

**Chart 1.1** Installed Revenue Opportunity by ESG Technology, World Markets: 2010-2020



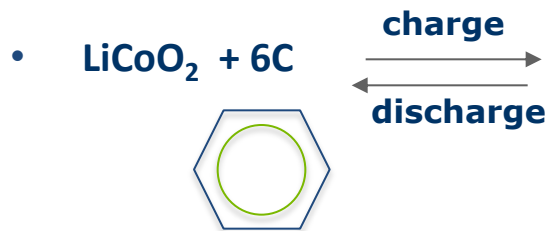
(Source: Pike Research)

# Schematic of a $\text{Li}_x\text{C}_6/\text{Li}_{1-x}\text{CoO}_2$ Li-Ion Cell Commercialized by Sony in 1991

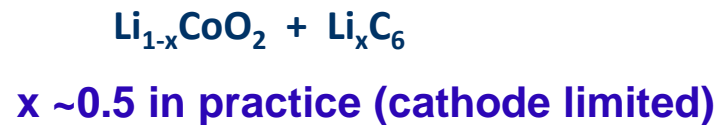


$\text{Li}_x\text{C}_6$  (Anode)

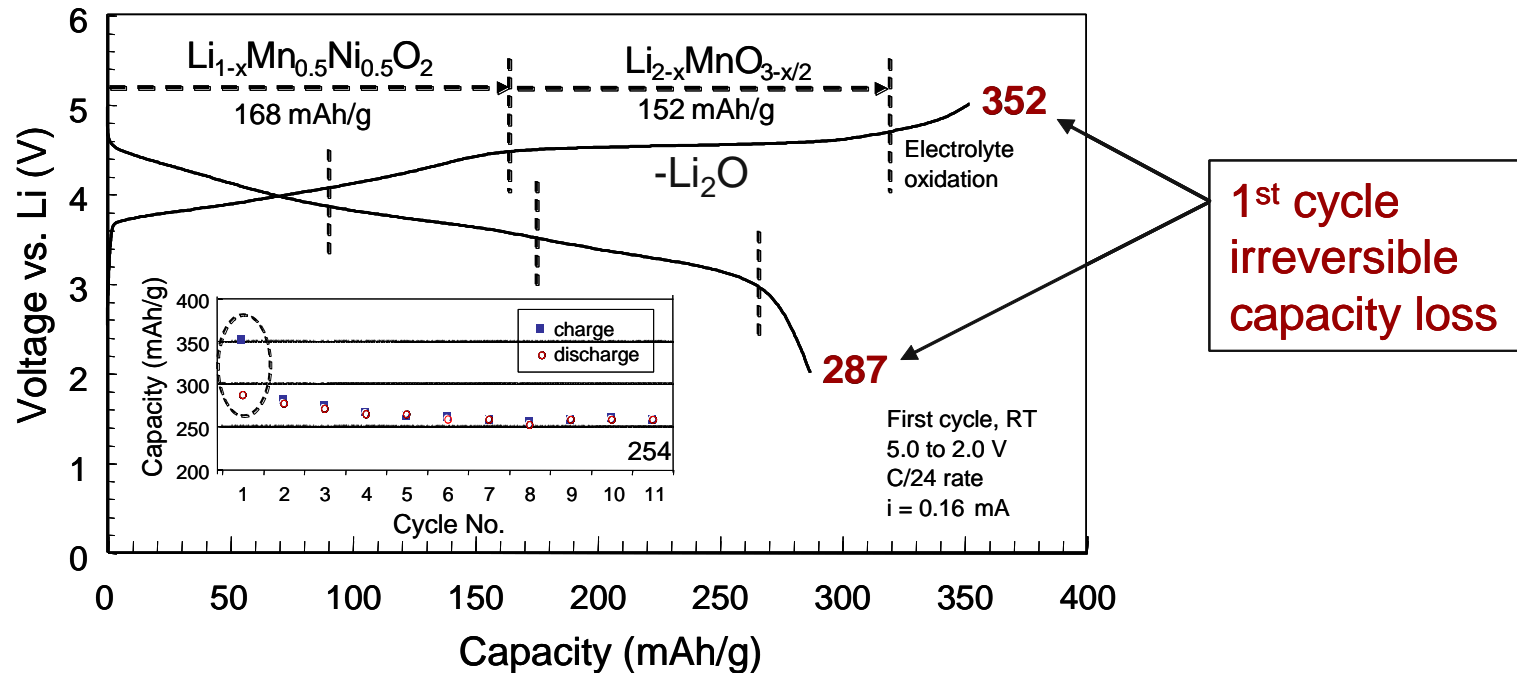
$\text{LiCoO}_2$  (Cathode)



Graphite building block



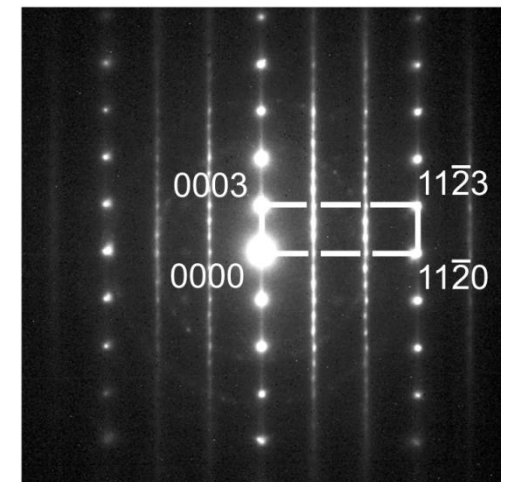
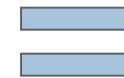
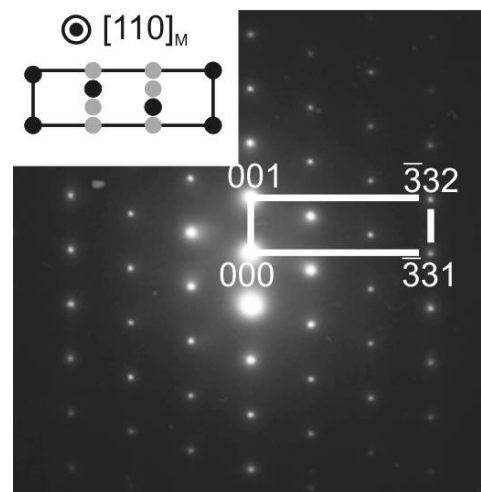
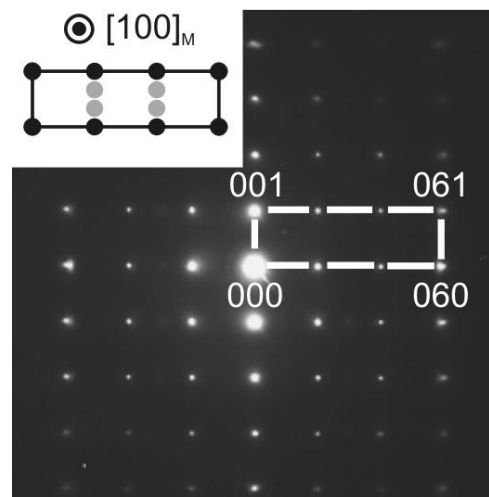
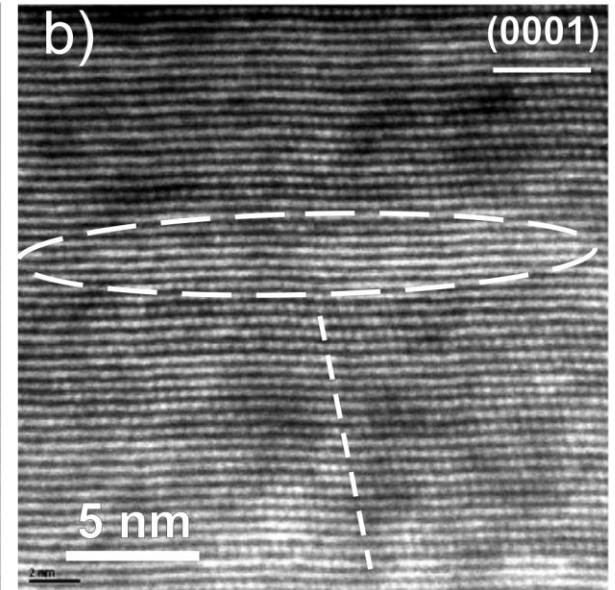
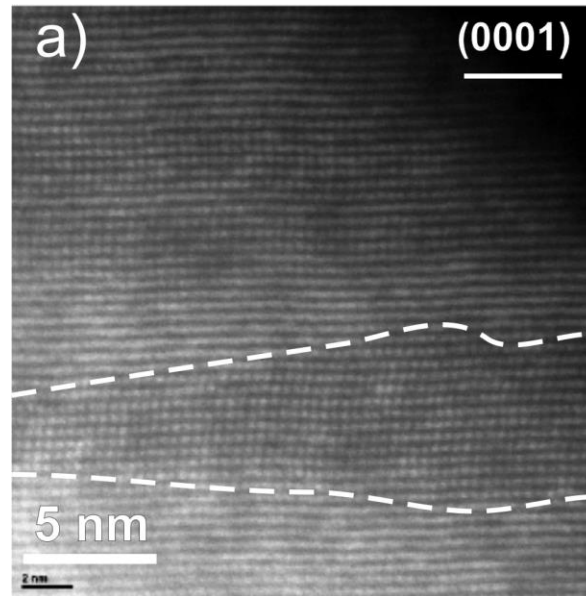
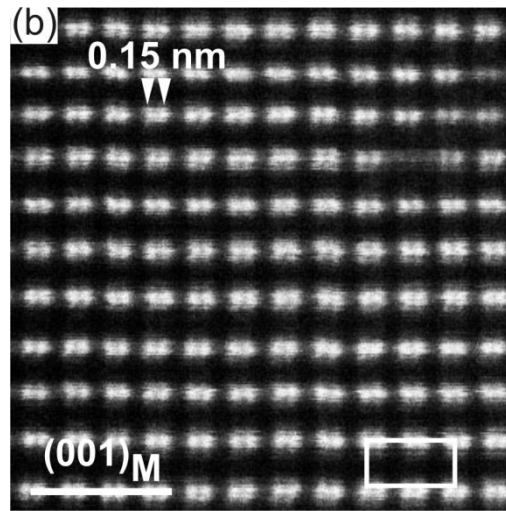
# Electrochemistry of a $\text{Li}/0.3\text{Li}_2\text{MnO}_3 \cdot 0.7\text{LiMn}_{0.5}\text{Ni}_{0.5}\text{O}_2$ Cell



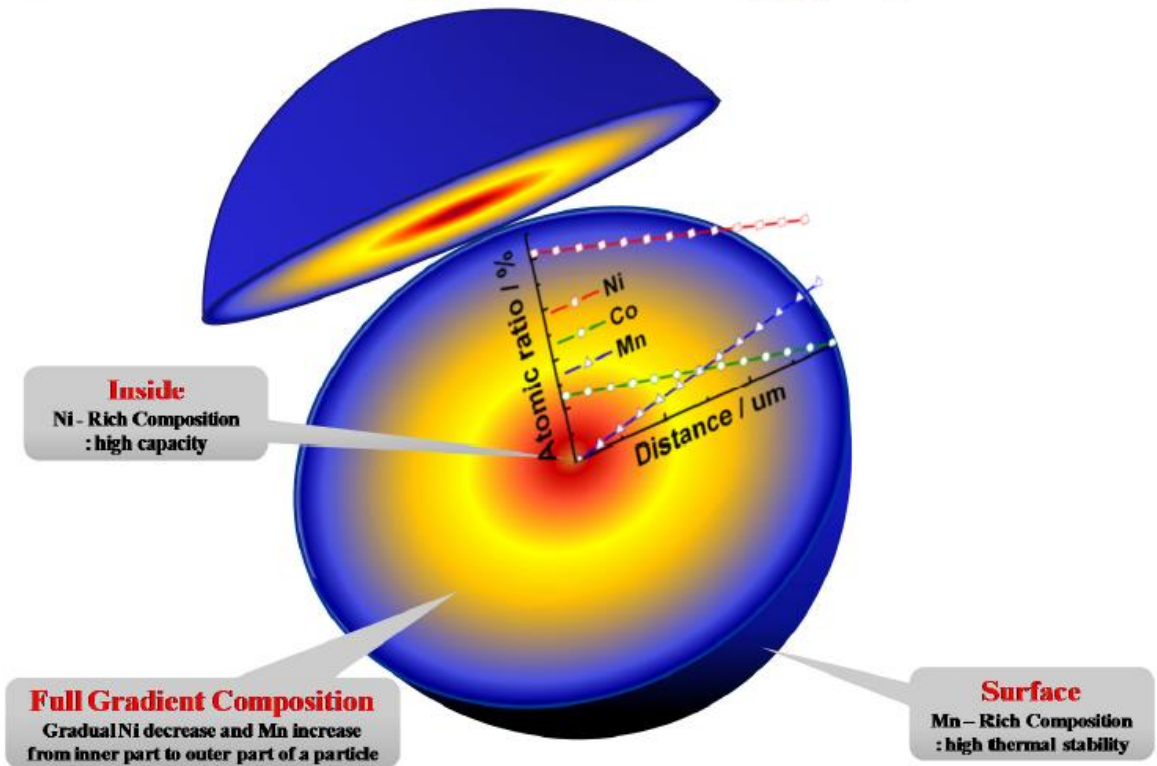
- Theoretical capacity of  $\text{LiMn}_{0.5}\text{Ni}_{0.5}\text{O}_2$  Component: 184 mAh/g
- Theoretical capacity of  $\text{Li}_2\text{MnO}_3$  Component: 158 mAh/g
- Theoretical charge capacity (total): 342 mAh/g
- Coulombic efficiency: 82% (1<sup>st</sup> cycle); >99% (10<sup>th</sup> cycle)
- Capacity (10<sup>th</sup> cycle): 254 mAh/g



# HAADF-STEM



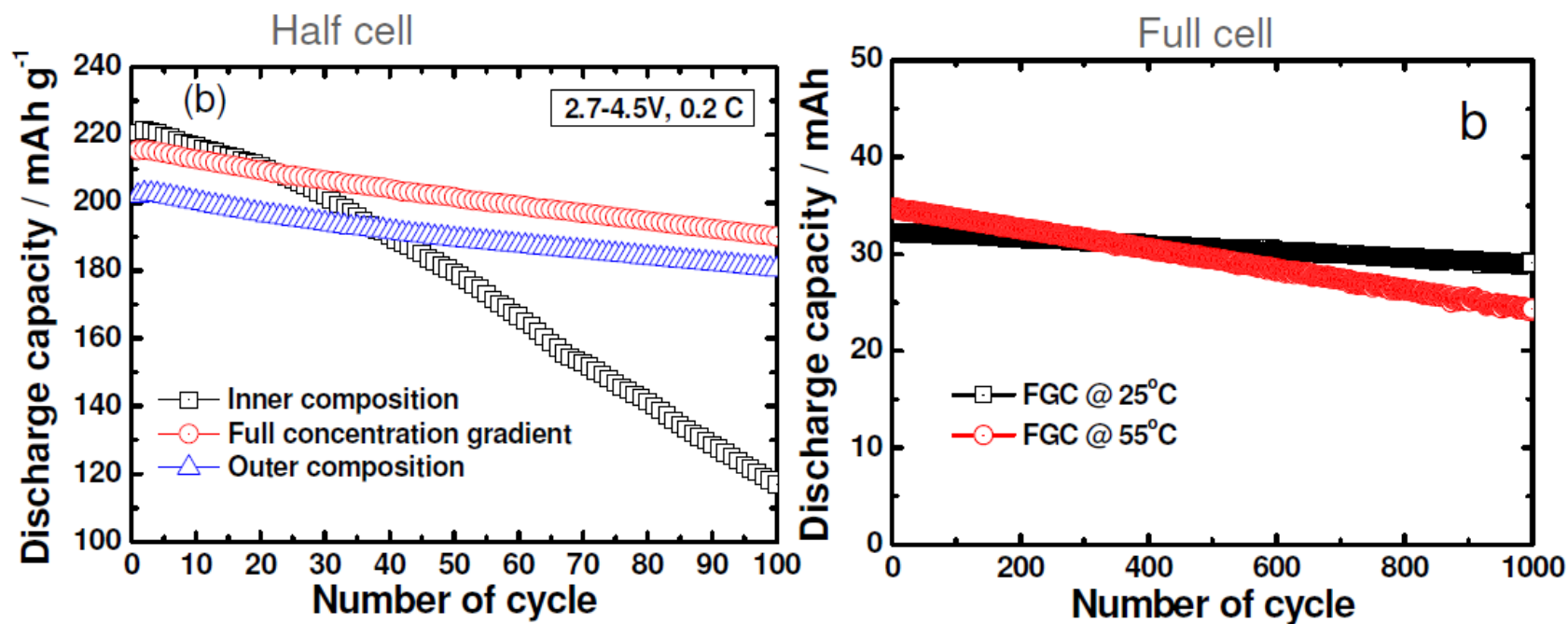
# High Energy Continuous Gradient materials (FGM) with average composition $\text{LiNi}_{0.6}\text{Co}_{0.10}\text{Mn}_{0.30}\text{O}_2$



Schematic diagram of the full concentration gradient lithium transition metal oxide particle with the nickel concentration decreasing from the center toward outer layer and the concentration of manganese increasing accordingly.



# Half and Full Cell Cycling performance of High Energy Full Gradient materials (FGM) at 25 and 55°C



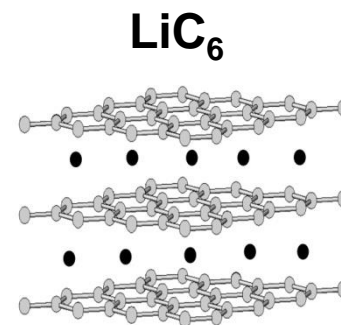
Full concentration gradient shows high capacity and very limited capacity fade after 1000 cycles at 55°C in a full cell,  
Electrolyte used is LiPF<sub>6</sub>/EC:EMC with 1%VC



# Li-Ion Batteries: Anode Materials

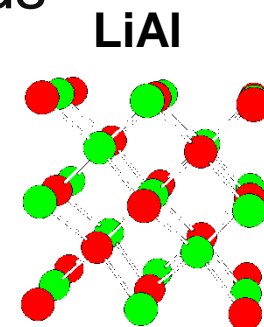
## ■ Carbon

- **Graphite:**  $<100$  mV vs.  $\text{Li}^0$
- Moderate capacity (372 mAh/g)
- Highly reactive, surface protection necessary



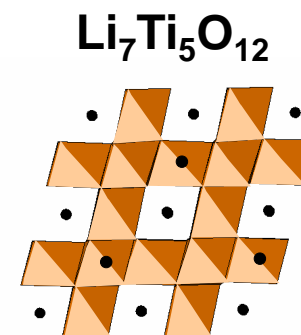
## ■ Metals, Semi-metals and Intermetallic Compounds

- Al, **Si**, CoSn,  $\text{Cu}_6\text{Sn}_5$ :  $<0.5$  V vs.  $\text{Li}^0$
- High gravimetric/volumetric capacities (1000-4000 mAh/g)
- Large volume expansion on reaction with lithium
- Reactive, surface protection required
- ***Greatest opportunity and challenge***

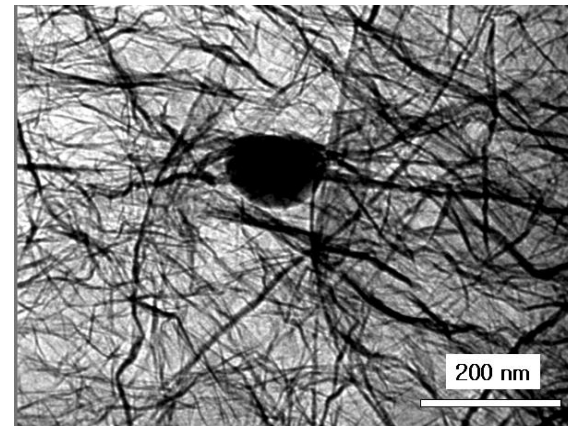
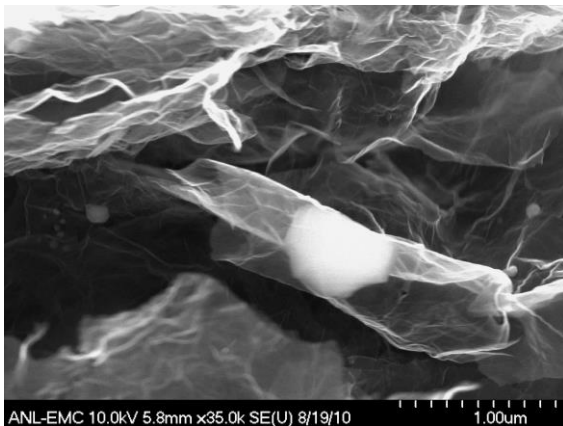
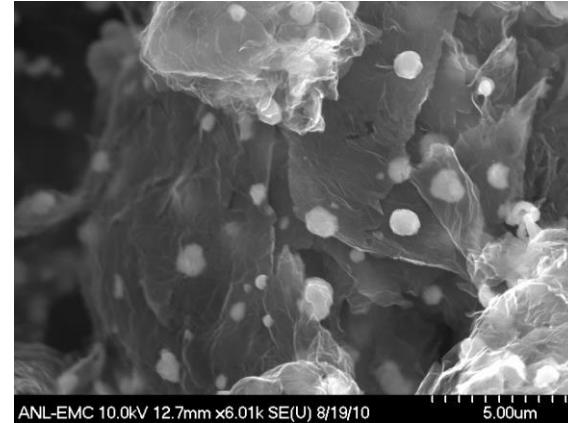
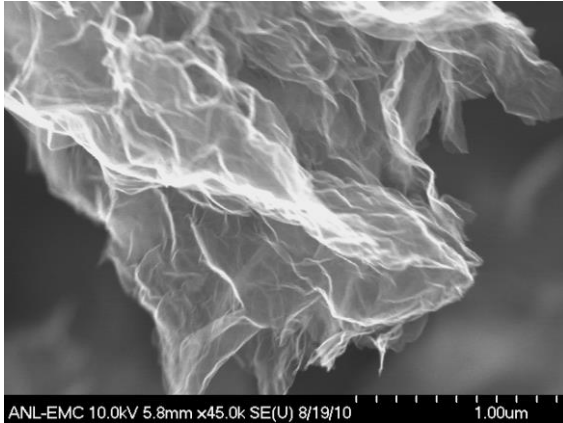


## ■ Metal Oxides

- **$\text{Li}_4\text{Ti}_5\text{O}_{12}$**  ( $\text{Li}[\text{Li}_{1/3}\text{Ti}_{5/3}]\text{O}_4$ ) Spinel: 1.5 V vs.  $\text{Li}^0$
- Low capacity (175 mAh/g)
- **Very high rate capability**
- Stable in nanoparticulate form

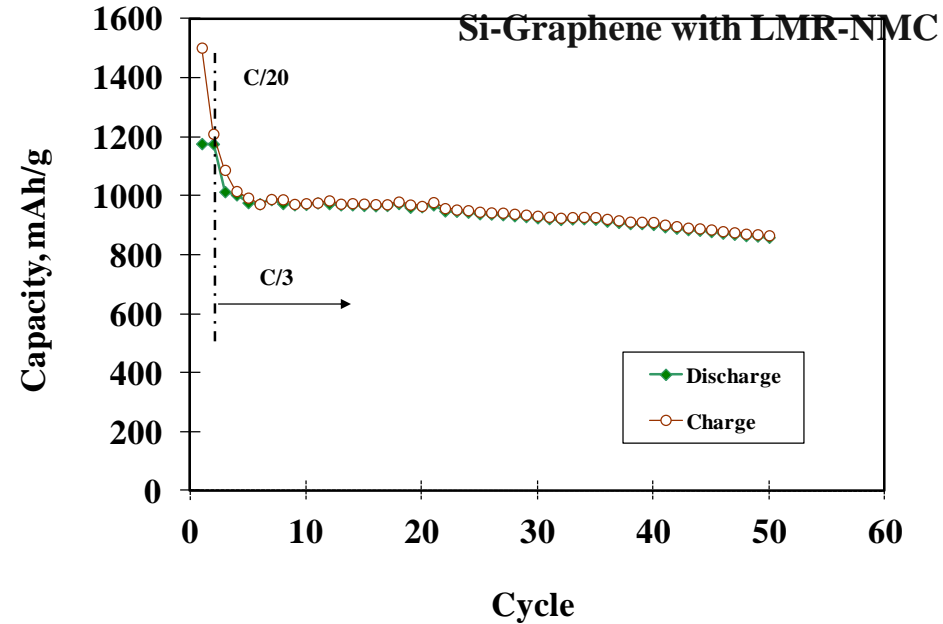
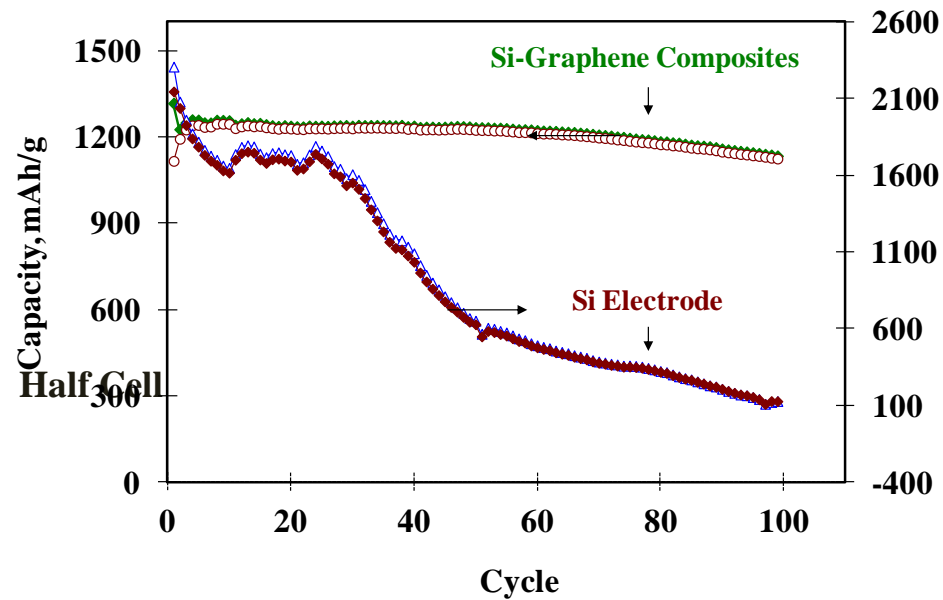


# Si-Graphene composites prepared through Gas Phase Deposition



*Silicon particles uniformly embedded inside graphene layers*

# Cell Data



*Si-Graphene possesses reversible capacity of 1100 mAh/g in 100 cycles*

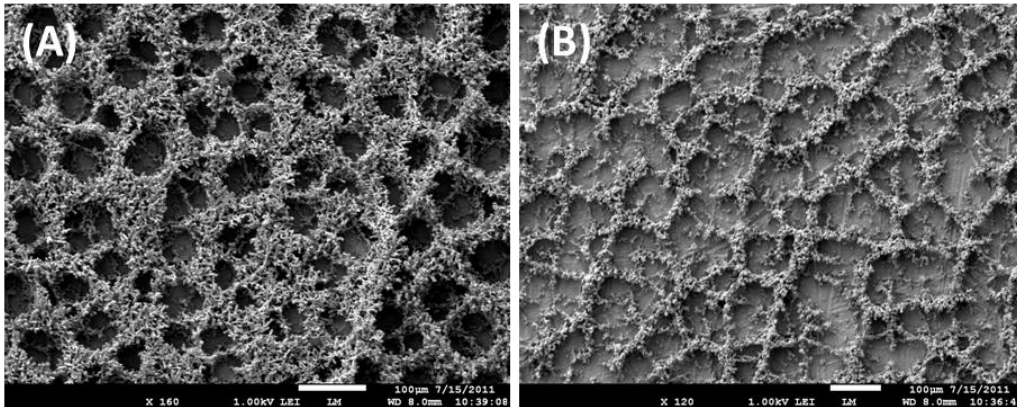
*340 Wh/Kg energy density could be achieved by this chemistry\**

\*Based on Argonne's Battery Design Model



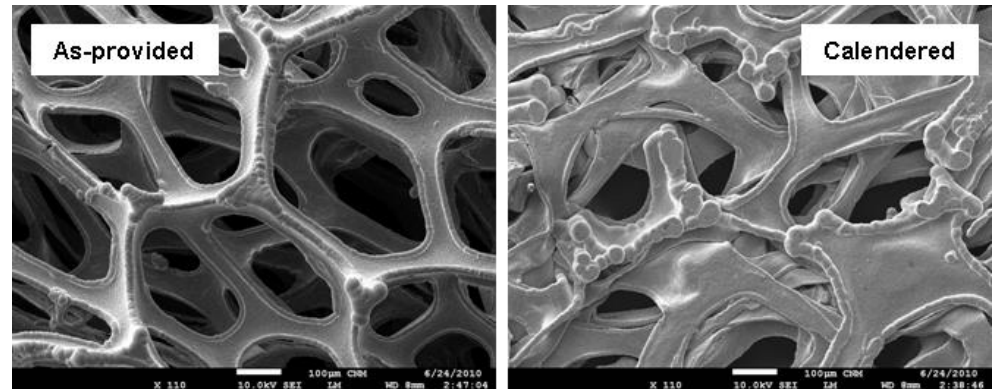
# Silicon on 3-D Architectures:

## Substrates: Copper foam synthesis



(left) Electrodeposited Cu foams with same Sn deposition performed on each. (A) 1 mM chloride concentration in Cu bath, (B) 4 mM chloride concentration in Cu bath

(right) Calendered commercial foams (CircuitFoil) before and after calendaring to 100 μm.

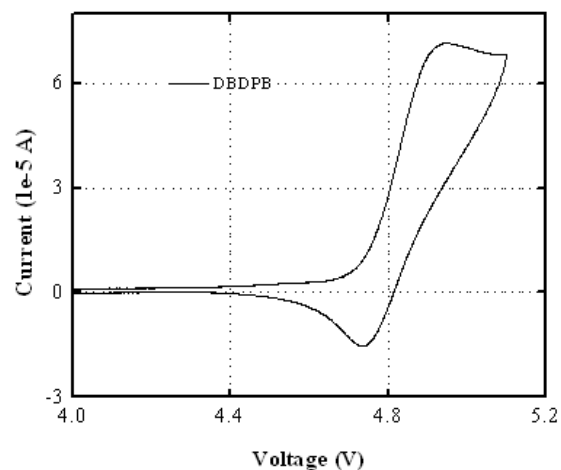
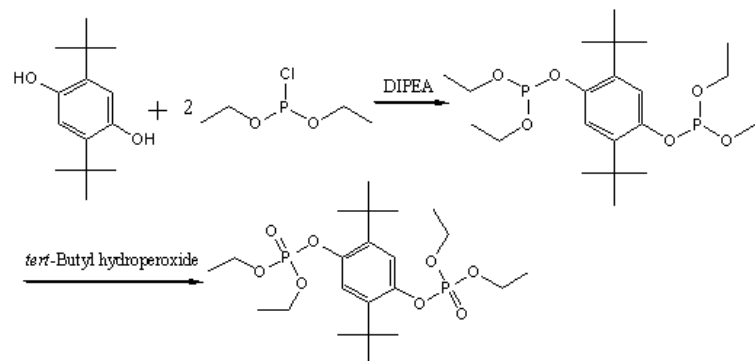


The porosity, thickness, and surface roughness of homemade foams is highly tunable. Commercial foams, however, offer the ease of reproducibility. More commercial vendors will be sought in order to have varying porosities.

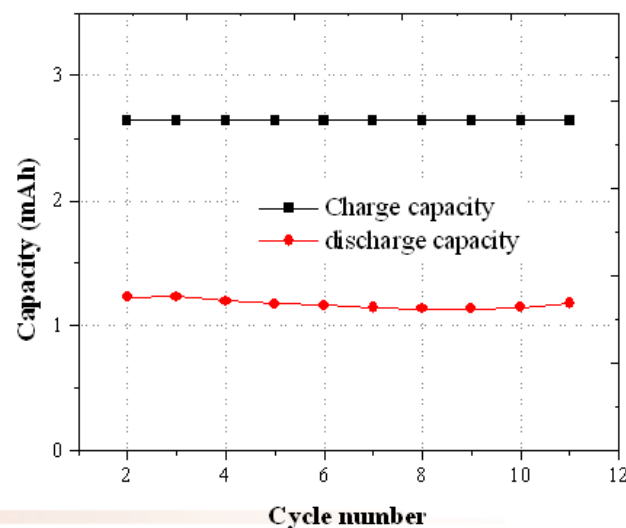
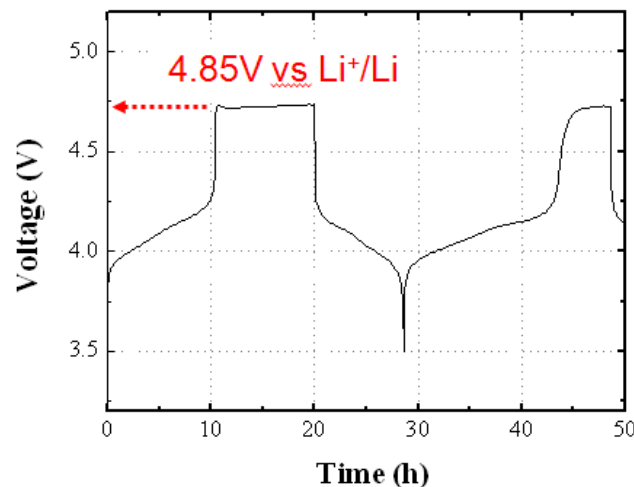


# High Voltage Redox Shuttle for Protection of 5V Cathode

To Further Increase the Redox Potential-Synthesis Tailored for High Voltage (4.8-5.0V) Cathodes



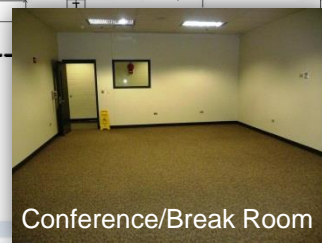
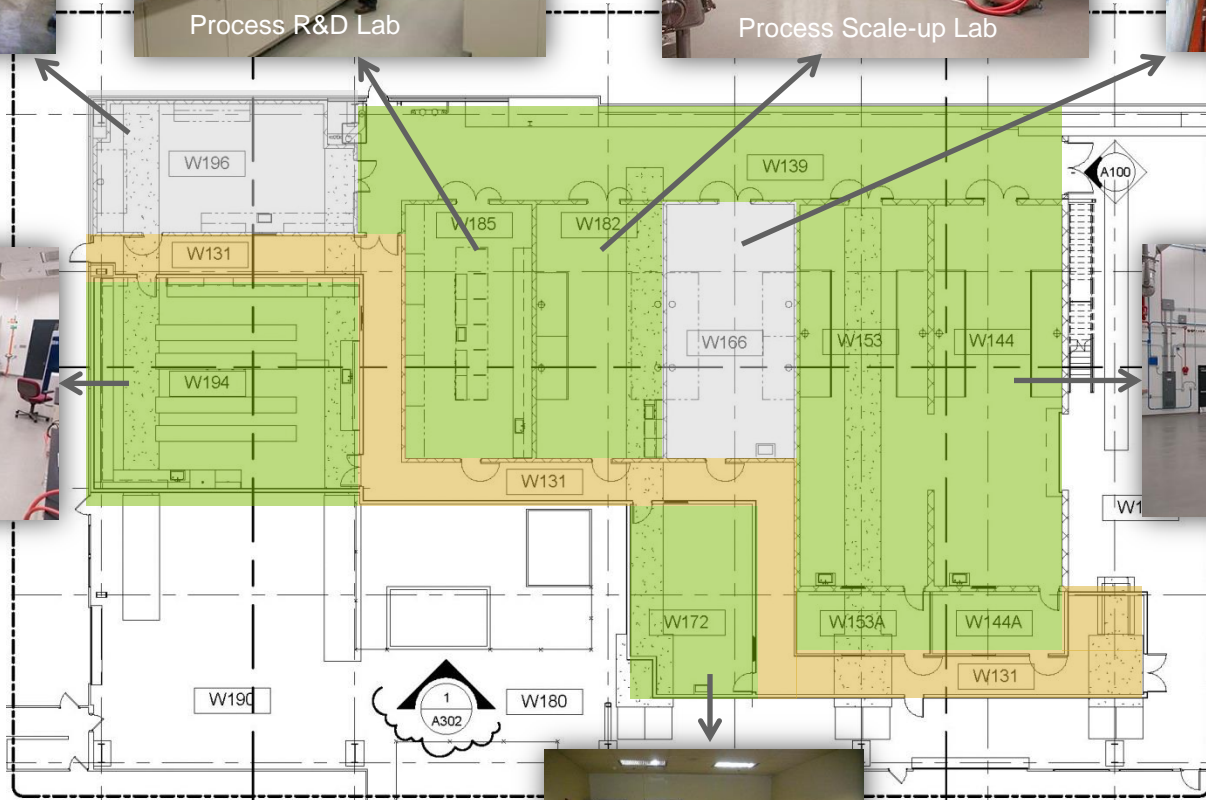
Cyclic voltammetry in 1.2 M LiPF<sub>6</sub> in EC/EMC (3:7 by weight); Scan rate is 100mV/s.



# Argonne's Materials Engineering Research Facility



# Argonne's Materials Engineering Research Facility



# Cell Fabrication Facility (CFF)



- The CFF was established by DOE-EERE, Vehicle Technologies Program, to fabricate commercial-grade sealed cells to facilitate the performance & life testing of promising advanced materials and cell chemistries developed on the ABR Program
  - Fabrication equipment housed in a new dry-room facility built for this purpose
  - Semi-automated equipment capable of coating, calendaring, & slitting electrodes; fabricating multi-electrode stacked pouch cells; & fabricating 18650 cylindrical cells
- ARRA funding used to procure cell formation, test, and characterization equipment

# Battery Test Laboratory

- New PC-based control & data acquisition system
- New software (PC compatible)
- New environmental chambers for testing cells and modules

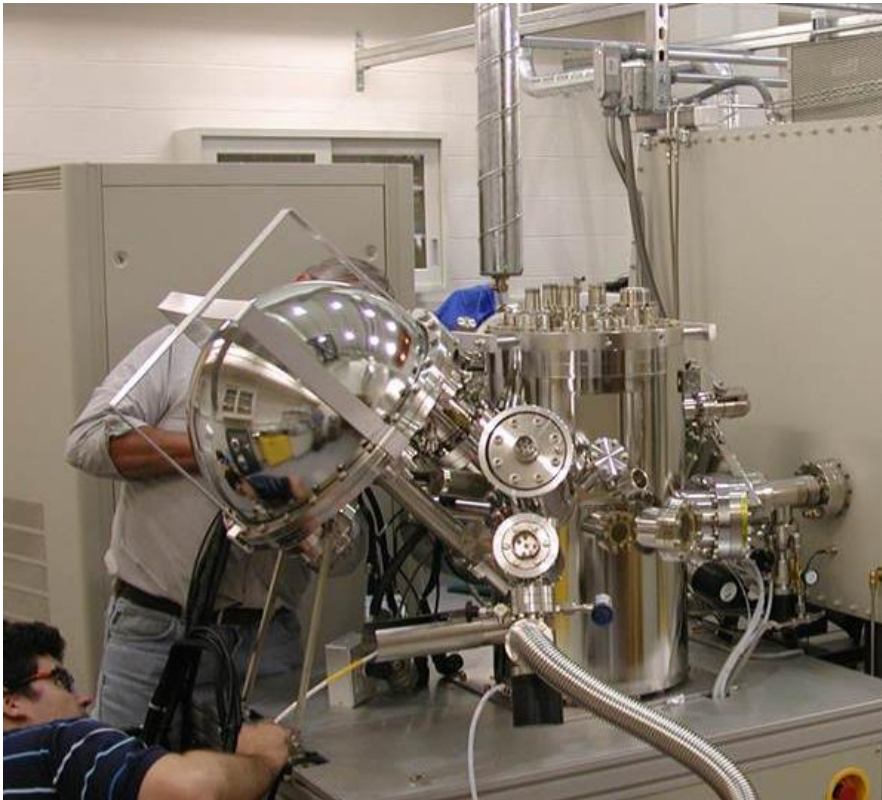
## ***Conduct independent performance & life tests:***

- DOE/USABC deliverables
- Non-DOE supported technologies
- ABR Program cells

## ***Utilize life test data to develop life prediction models for different technologies***



# New Post-Test Analysis Facility (PTF)



- Linked to & operated in a manner similar to the battery test facility
- ARRA funding was used to establish a new integrated post-test analysis facility incorporating a variety of teardown & diagnostic capabilities:



# JOINT CENTER FOR ENERGY STORAGE RESEARCH: AN OVERVIEW

Jeff Chamberlain | JCESR Deputy Director

# Start with the simple facts

- ▶ JCESR is a DOE Energy Innovation Hub, funded through the Basic Energy Sciences office of DOE, and led by Argonne National Laboratory
- ▶ Hubs are a bold initiative by DOE
  - Through science, focus on solving a single, societal problem
  - Integrated team effort
  - Rapid translation to societal impact



# JCESR Targeted Outcomes

*ACHIEVING GOALS FOR LASTING LEGACIES*

- ▶ Transformational goals: 5-5-5
  - 5 times greater energy density → Beyond Li-ion
  - 1/5 cost
  - within 5 years
- ▶ Legacies
  - Pre-commercial prototypes for grid and transportation
  - Library of fundamental knowledge
    - Atomic and molecular understanding of battery phenomena
  - New paradigm of battery development
    - Science-based rational design
    - Systems-centric
    - End-to-end integration

# JCESR Team

5

## National Laboratories

ANL, LBNL,  
Sandia,  
PNNL, SLAC

5

## Universities

UIC, UC, NU,  
UIUC, UM

4

## Private-Sector Partners

Dow, JCI,  
AMAT,  
CET



# Today's Paradigm

Science Community

**ISOLATED  
ISOLATED**

Engineering Community

Science

Journal  
Articles/Patents

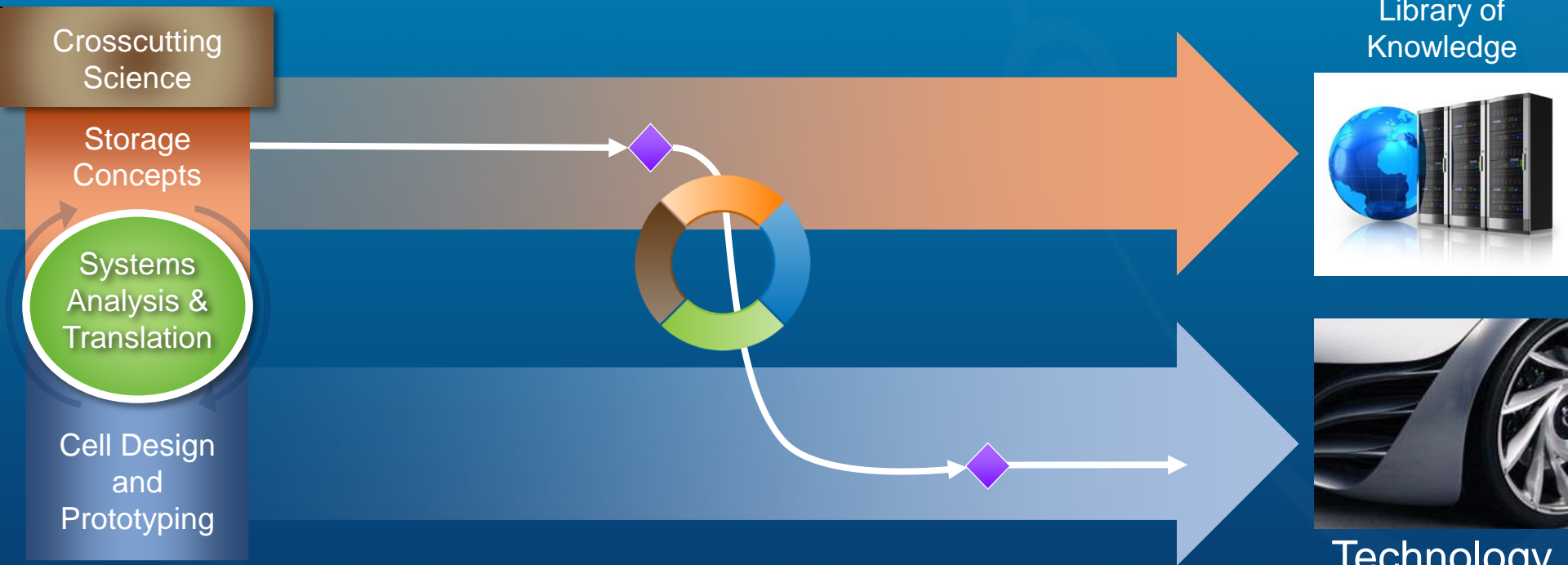


Technology

5% Improvement  
per year

- Component-centric • Sequentially organized •
- Incremental improvement •

# JCESR Paradigm



Science  
Library of  
Knowledge



Technology

5 Times Greater  
Energy Density

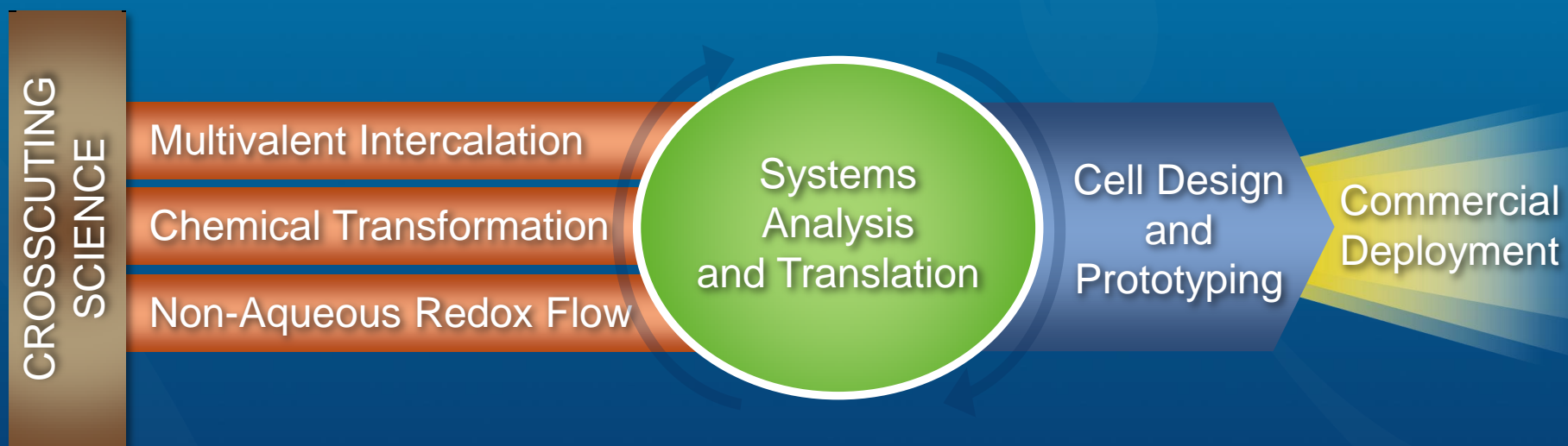
1/5 Cost

5 Years

- Science-based rational design • Parallel development •
- Systems-centric • End-to-end integration • Labs, Universities, Industry •

# Introduction

## *JCESR IN A SINGLE GRAPHIC*

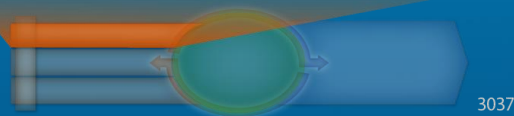


# Multivalent Intercalation

## POTENTIAL TO DOUBLE CAPACITY OF CATHODE

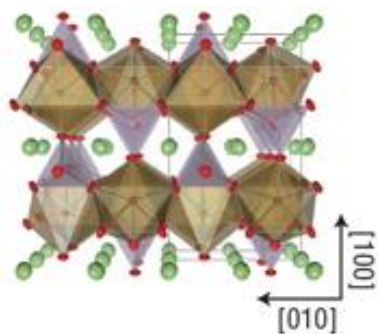
### Multivalent Intercalation

- Mobility in host structures
- Mobility across interfaces
- Stable and selective interfaces

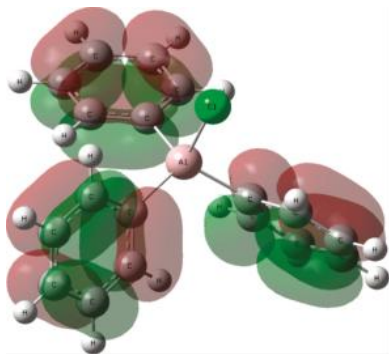


### Objectives

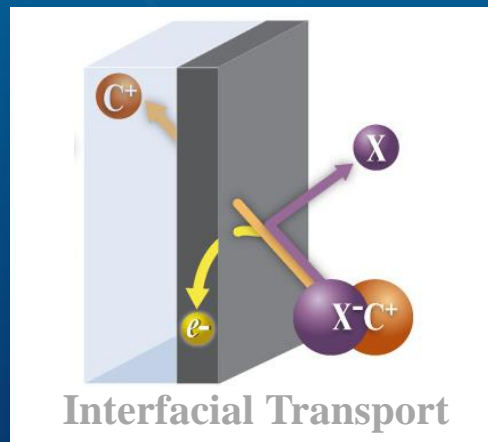
- Understand how highly charged cations move through solids, liquids and interfaces while under electrochemical control



Host Mobility



Ion/Solvent Interactions

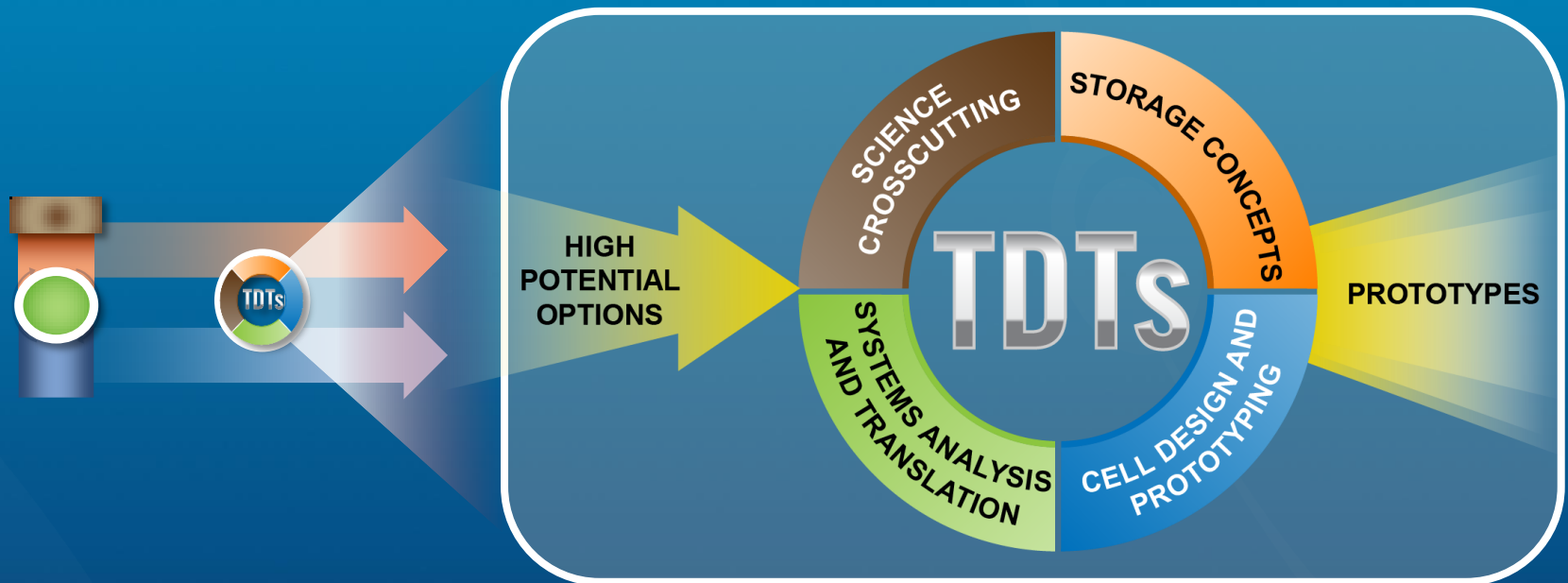


Interfacial Transport

Combining multivalent cathode and reversible metal anode has potential to achieve 5x Li-ion energy density

# Translational Development Teams

## *INTEGRATING SCIENCE WITH ENGINEERING*



- ▶ We kickoff two TDTs to design and prototype cells
  - Grid
  - Transportation

# JCESR Applies Several Mechanisms to Meet the 5-5-5

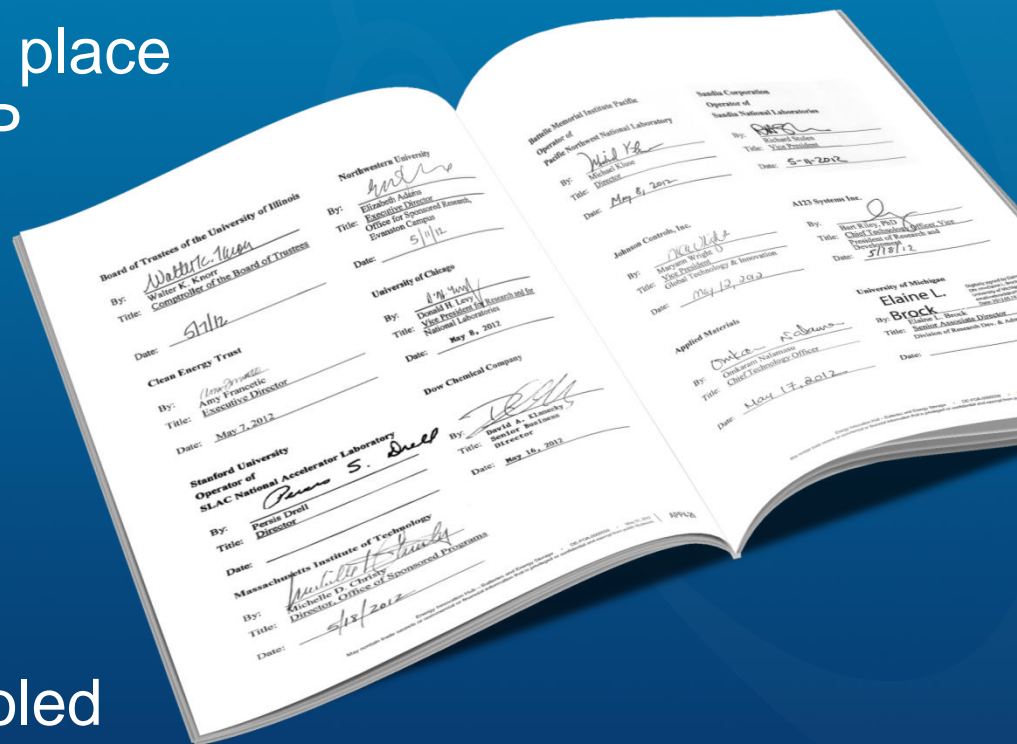
- ▶ RDD&D Spectrum
  - Systems Analysis and Translation
    - Techno-Economic Modeling
  - Translational Development Teams
  - Cell Design and Prototyping
- ▶ Intellectual Property Management
- ▶ Advisory groups
- ▶ Affiliates

RESULTS IN COMMON TEAM WORKING TO SHARED MISSION

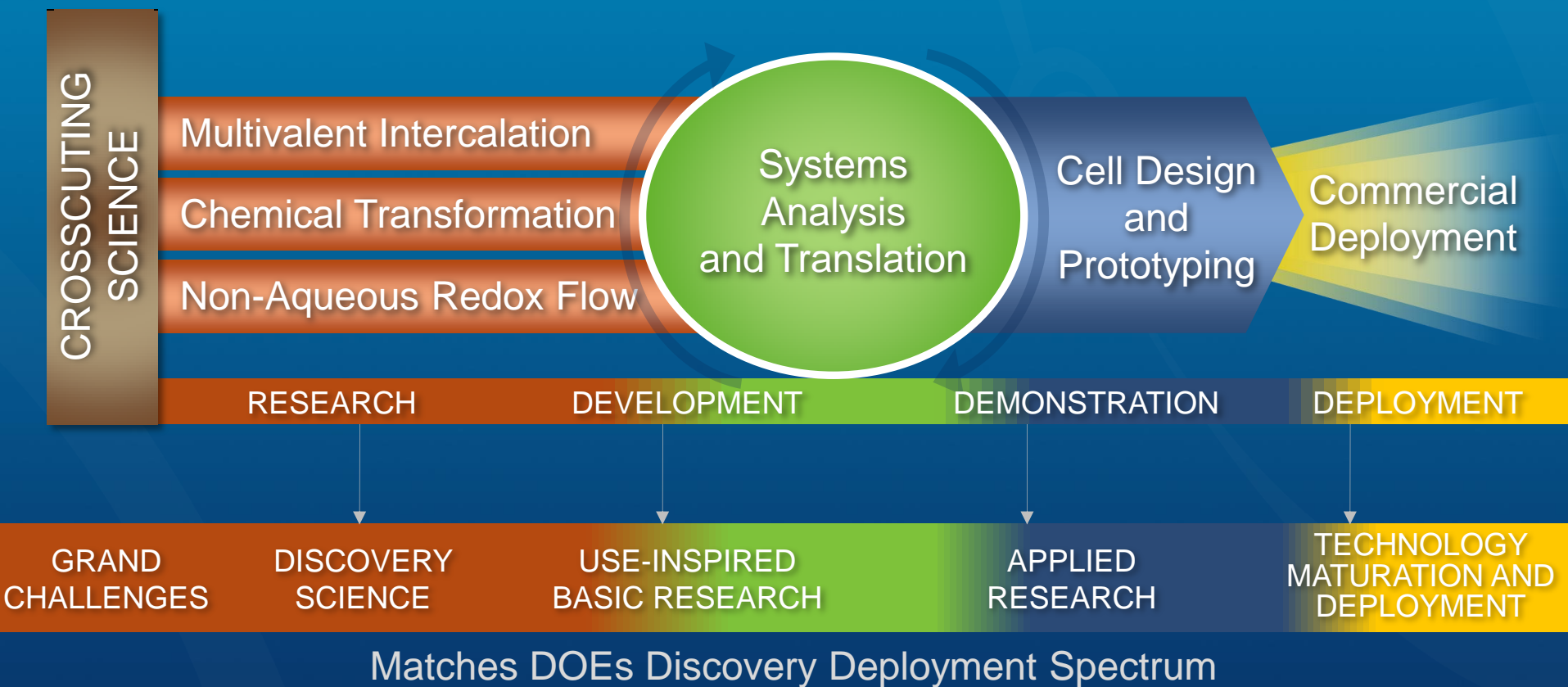
# IP Plan

## FACILITATES OPEN COLLABORATION AND TRANSLATION TO MARKET

- ▶ Agreement signed and in place to operate under single IP Management Plan
- ▶ All foreground IP pooled, with centralized licensing
  - Allows industrial concerns to see a clear path to commercialization
  - One stop shopping
- ▶ Some Background IP pooled



# JCESR Integrates All Aspects of RDD&D



# Working with DoD

- We have a high-powered energy-storage research engine sponsored primarily by DOE, both in Li-ion and with JCESR focusing on beyond Li-ion.
- We can deliver our innovations to DoD through DoD-approved suppliers.
- Branches of the military might serve in an advisory capacity to JCESR.



Questions?

