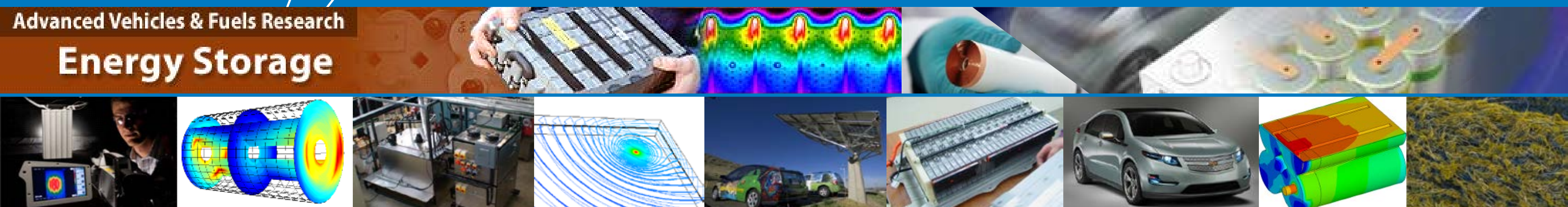
NREL
NATIONAL RENEWABLE ENERGY LABORATORY

Advanced Models and Controls for Prediction and Extension of Battery Lifetime



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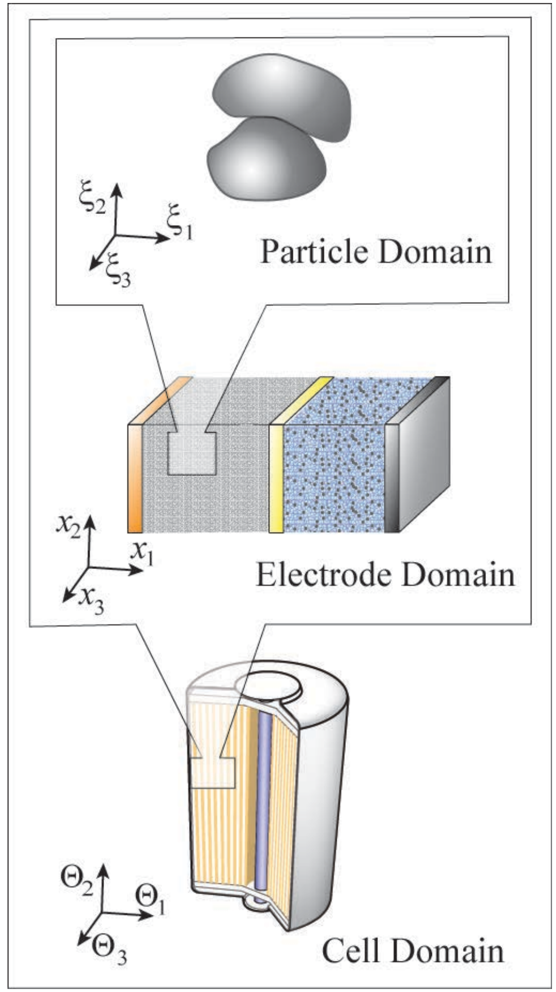
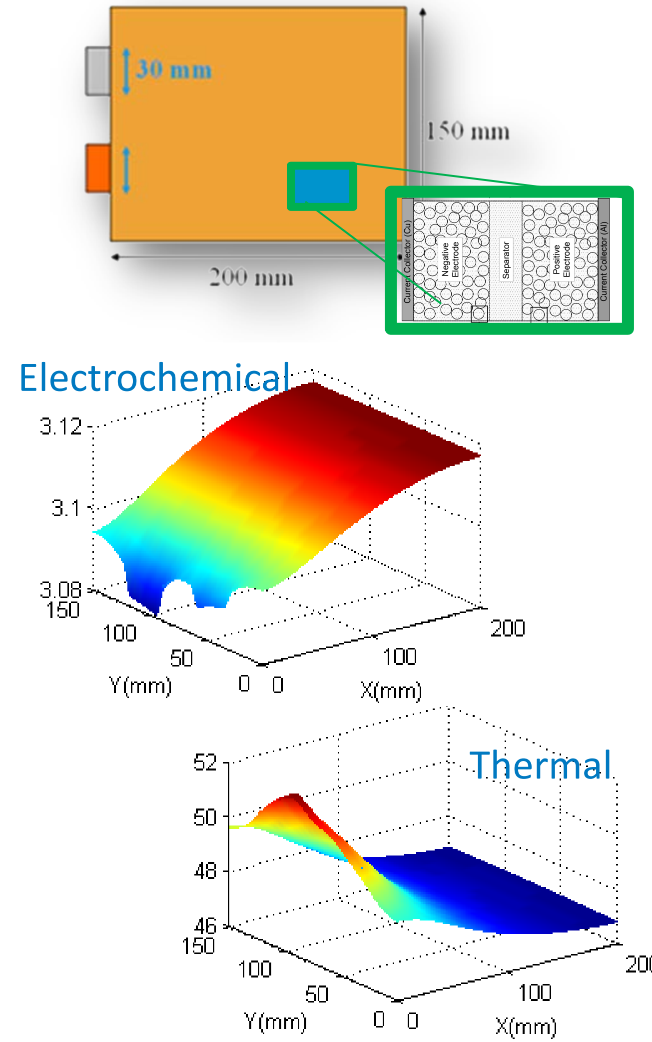
NREL Electrochemical/Thermal/Life Models

Multi-Scale Multi-Domain (MSMD) model

- Inter-domain coupling of field variables, source terms
- Efficient, flexible framework for physics expansion
- Leading approach for large-cell computer-aided engineering models





Kim et al. (2011) "Multi-Domain Modeling of Lithium-Ion Batteries Encompassing Multi-Physics in Varied Length Scales", *J. of Electrochemistry*, Vol. 158, No. 8, pp. A955–A969

NREL Electrochemical/Thermal/Life Models

Life-predictive model

- Physics-based surrogate models tuned to aging test data
- Implemented in system design studies & real-time control
- Regression to NCA, FeP, NMC chemistries

NCA = Nickel-Cobalt-Aluminum
FeP = Iron Phosphate
NMC = Nickel-Manganese-Cobalt

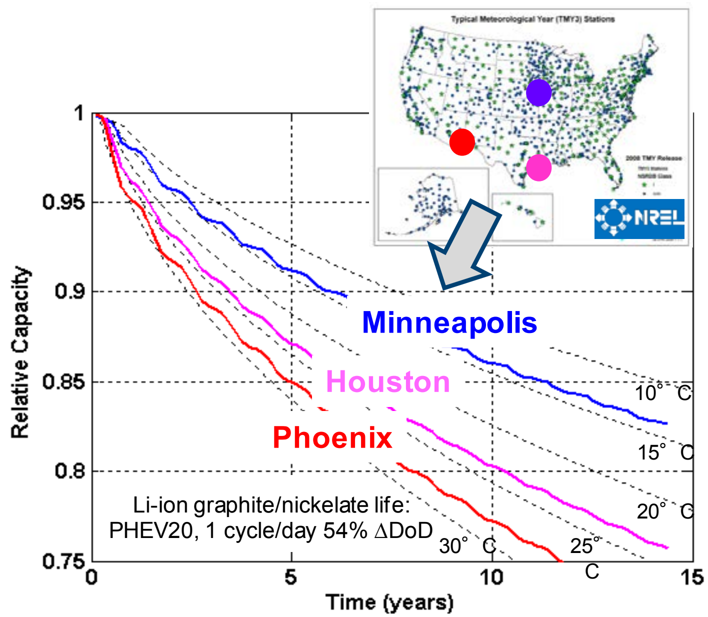
Calendar fade
- SEI growth
- Loss of cyclable lithium
- Coupled with cycling
- $a_1, b_1 = f(\Delta DOD, T, V_{oc}, \ldots)$

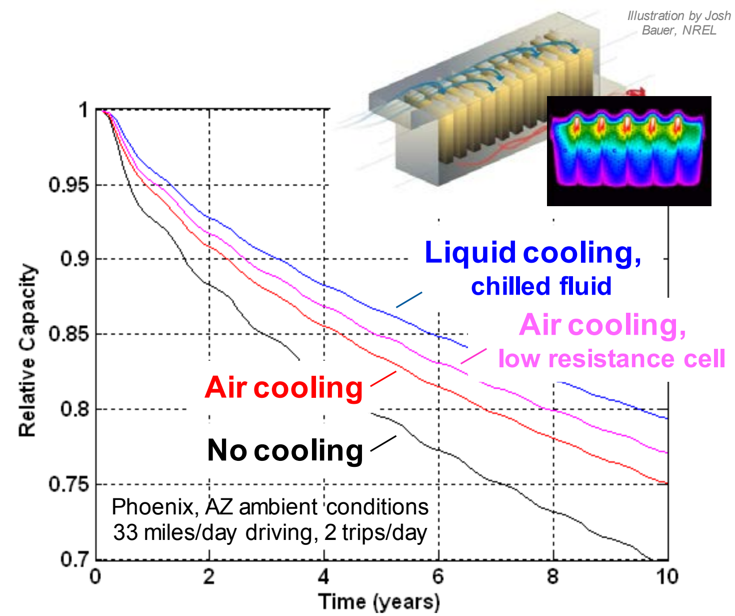
Cycling fade
- Active material structure degradation and mechanical fracture
- $a_2, c_2 = f(\Delta DOD, T, V_{oc}, \ldots)$

Relative Resistance $R = a_1 t^z + a_2 N$

Relative Capacity $Q = \min(Q_{Li}, Q_{sites})$

$Q_{Li} = b_0 + b_1 t^z + \ldots$ $Q_{sites} = c_0 + c_2 N + \ldots$





Challenges and Needs for Life/Degradation Models

Challenges:

- **No standard process for life certification**
- **Predictive models must consider some 5 to 10 coupled degradation mechanisms**
  - Electrochemical/thermal/mechanical mechanisms not yet fully understood or modeled
- **Lifetime uncertainty absorbed in excess design and warranty costs of xEV battery systems**

Needs:

- **Predict lifetime more accurately, with less test data**
  - Critical to capture accelerating fade effects nearing end-of-life
- **Provide engineering feedback for cell, pack and system control designs**

Outline

- **Models**
  - Surrogate life models (cell level)
  - Physics life models (cell level)
  - System life (pack level)

- **Lifetime extension**
  - Thermal control
  - Charge control
  - Cell electrochemical-based control
  - Active cell balancing
  - Prognostic-based supervisory control

Models

- **Surrogate life models (cell)**
  - Present state of art for lifetime prediction
  - Ranking of importance of mechanical-coupled degradation mechanisms on electrode site-loss
- **Physics life models (cell)**
  - Coupling of solid mechanics with electrochemical/ thermal physics
- **System life (pack)**
  - Vehicle & pack thermal models
  - Cell performance & aging process variation

NREL Life Predictive Model

Calendar fade

- SEI growth
- Loss of cyclable lithium
- Coupled with cycling
- a_1, b_1 = f(ΔDOD,T,V_{oc},…)

Cycling fade

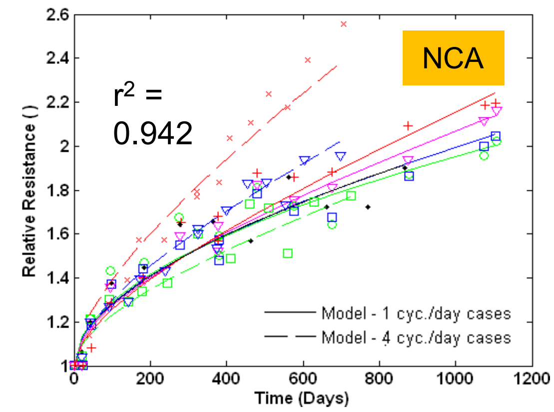
- Active material structure degradation and mechanical fracture
- a_2, c_2 = f(ΔDOD,T,V_{oc} ,…)

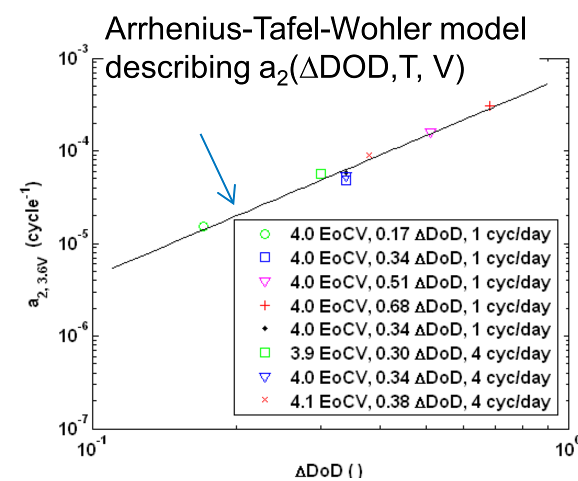
Relative Resistance $$R = a_1 t^z + a_2 N$$

Relative Capacity $$Q = \min(Q_{Li}, Q_{sites})$$

$$Q_{Li} = b_0 + b_1 t^z + \ldots$$

$$Q_{sites} = c_0 + c_2 N + \ldots$$





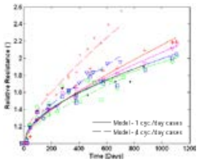
•Data: J.C. Hall, IECEC, 2006.

- **Statistical regression to experimental data**
- **Correct separation of calendar vs. cycling mechanisms**
- **In rate form, extensible to untested scenarios**

NREL Life Model Framework

Data

A. Resistance growth during storage
   Broussely (Saft), 2007:
   - T = 20°C, 40°C, 60°C
   - SOC = 50%, 100%



B. Resistance growth during cycling
   Hall (Boeing), 2005-2006:
   - DoD = 20%, 40%, 60%, 80%
   - End-of-charge voltage = 3.9, 4.0, 4.1 V
   - Cycles/day = 1, 4

C. Capacity fade during storage
   Smart (NASA-JPL), 2009
   - T = 0°C, 10°C, 23°C, 40°C, 55°C

   Broussely (Saft), 2001
   - V = 3.6V, 4.1V

D. Capacity fade during cycling
   Hall (Boeing), 2005-2006: (see above)

NCA

Regression of candidate models

1. Fit local model(s)
2. Visualize rate-dependence on operating condition
3. Hypothesize rate-law(s)

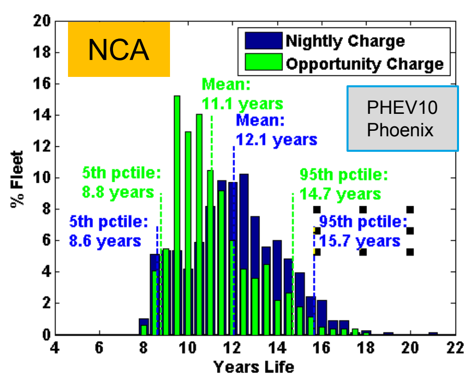
$$\theta_T = \exp\left[\frac{-E_a}{R}\left(\frac{1}{T(t)} - \frac{1}{T_{ref}}\right)\right] \qquad \theta_V = \exp\left[\frac{\alpha F}{R}\left(\frac{V_{oc}(t)}{T(t)} - \frac{V_{ref}}{T_{ref}}\right)\right] \qquad \theta_{\Delta DoD} = \left(\frac{\Delta DoD}{\Delta DoD_{ref}}\right)^{\beta}$$

4. Fit rate-laws(s)
5. Fit global model(s)

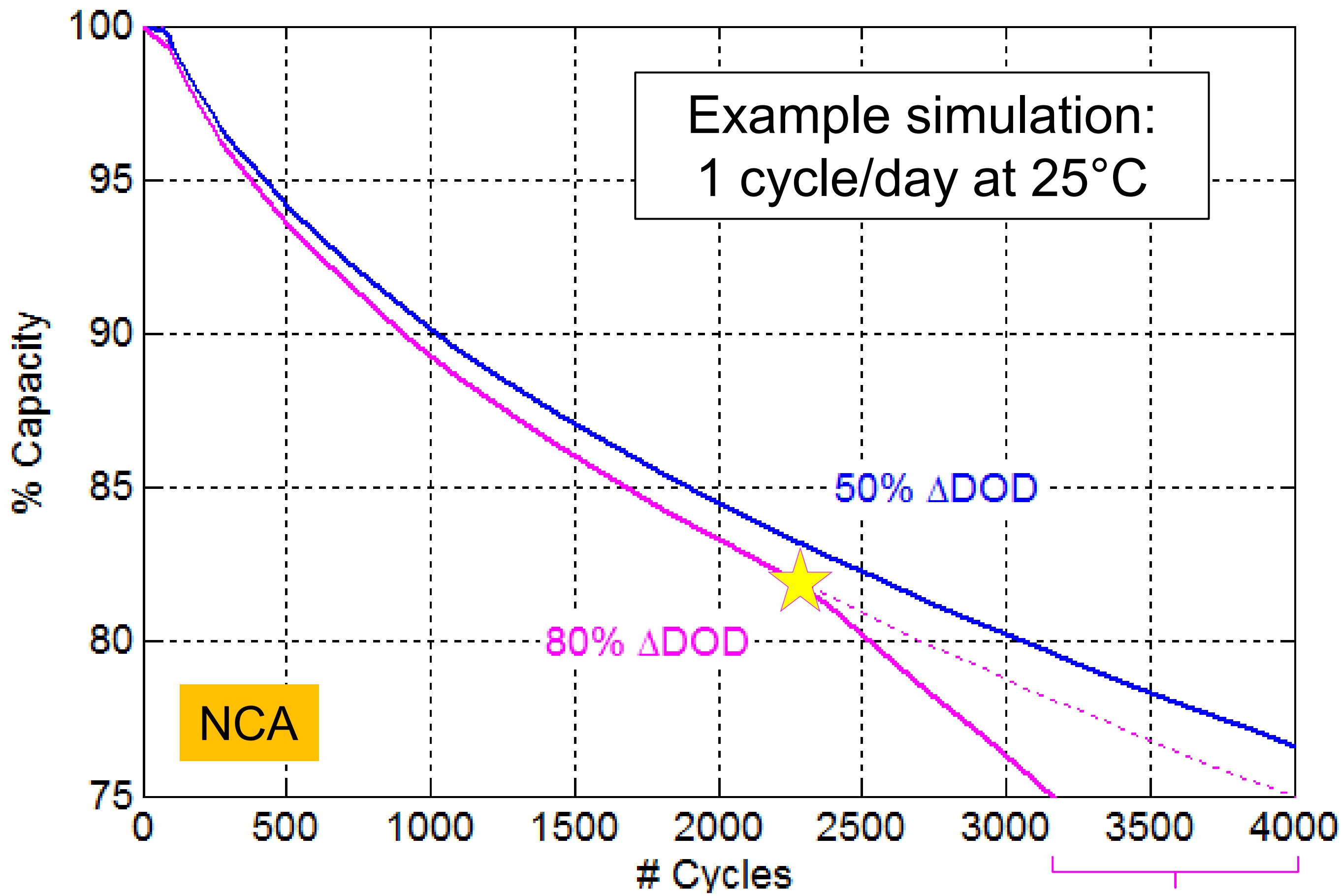
Model selection based on statistics

#	Storage Model	Parameters	RMSE (% capacity)	R2	Adjusted R2
7	q = 1 + b1*t^0.5	b1(T,Voc)	2.06	0.925	0.923
8	q = 1 + b1*t^z	b1(T,Voc), z	2.01	0.929	0.926
9	q = 1 + b1*t^z	b1(T,Voc), z(T)	2.03	0.929	0.925
10	q = 1 + b1*t.^0.5 + b2*t	b1(T,Voc), b2	1.99	0.930	0.927
11	q = 1 + b1*t.^0.5 + b2*t	b1(T,Voc), b2(T)	2.00	0.931	0.926
12	q = 1 + b1*t.^0.5 + b2*t	b1(T,Voc), b2(T,Voc)	1.87	0.941	0.936

Predictive model



Knee in Fade Critical for Predicting End of Life



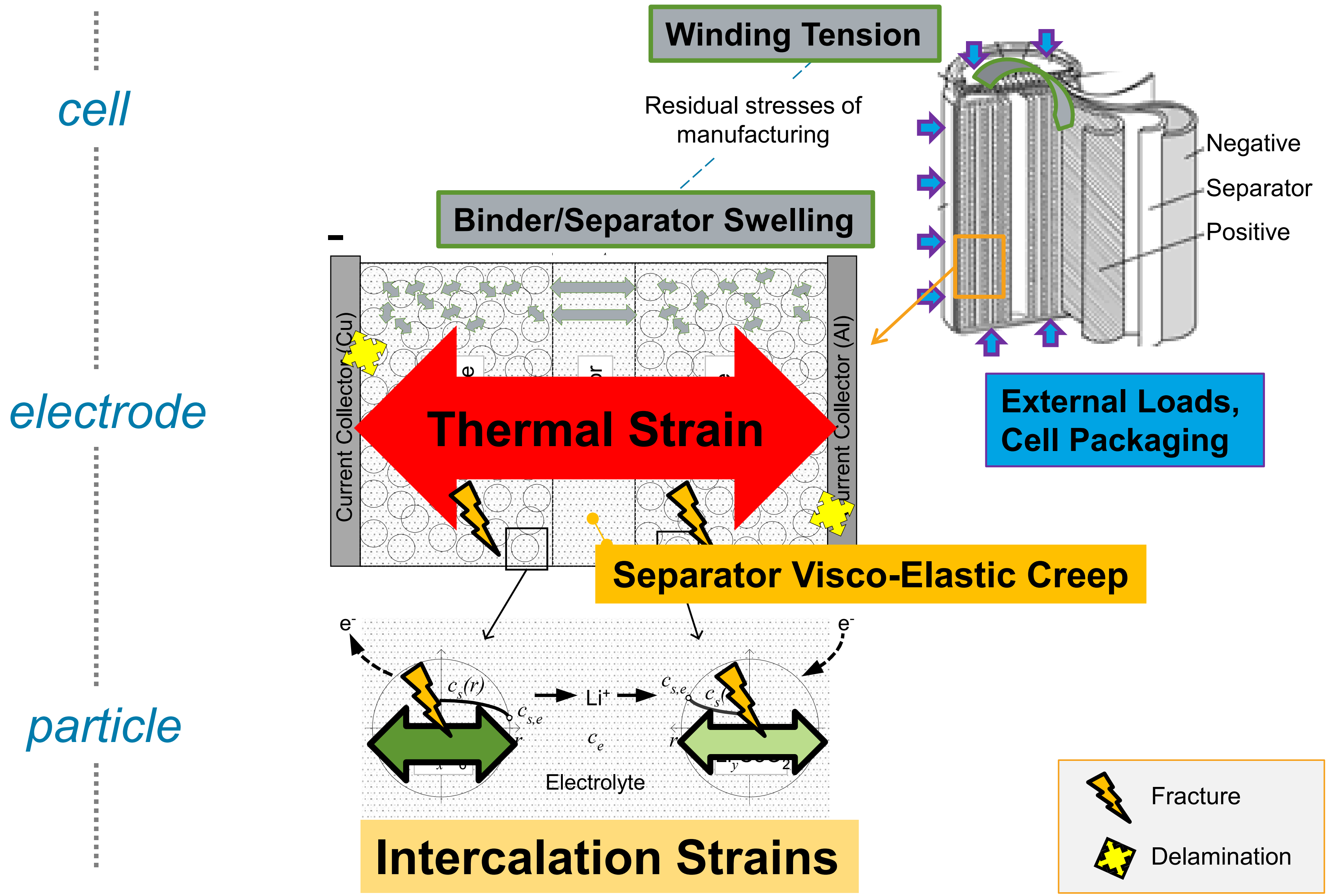
<u>50% DOD:</u>
Graceful fade (controlled by lithium loss)

<u>80% DOD:</u>
Graceful fade transitions to sudden fade ~2300 cycles

Life over-predicted by 25% without "knee"

Hypothesis based on analysis of aging data: Transition from Li loss (chemical w/ weak mech. coupling) to site loss (mechanical)

Sources of Mechanical Stress in Li-ion Cells



Open Areas in Mechanics Modeling

1) At which length-scale do stresses most impact battery life, e.g.:

- Particle-level fracture
- Active material bulk expansion/contraction
- Thermal expansion/contraction
- Polymer creep (binder, separator)

2) Optimal packaging of jellyroll (esp. pouch cells)

- Performance
- Lifetime

3) Multi-scale linkage to 3D automotive cell level

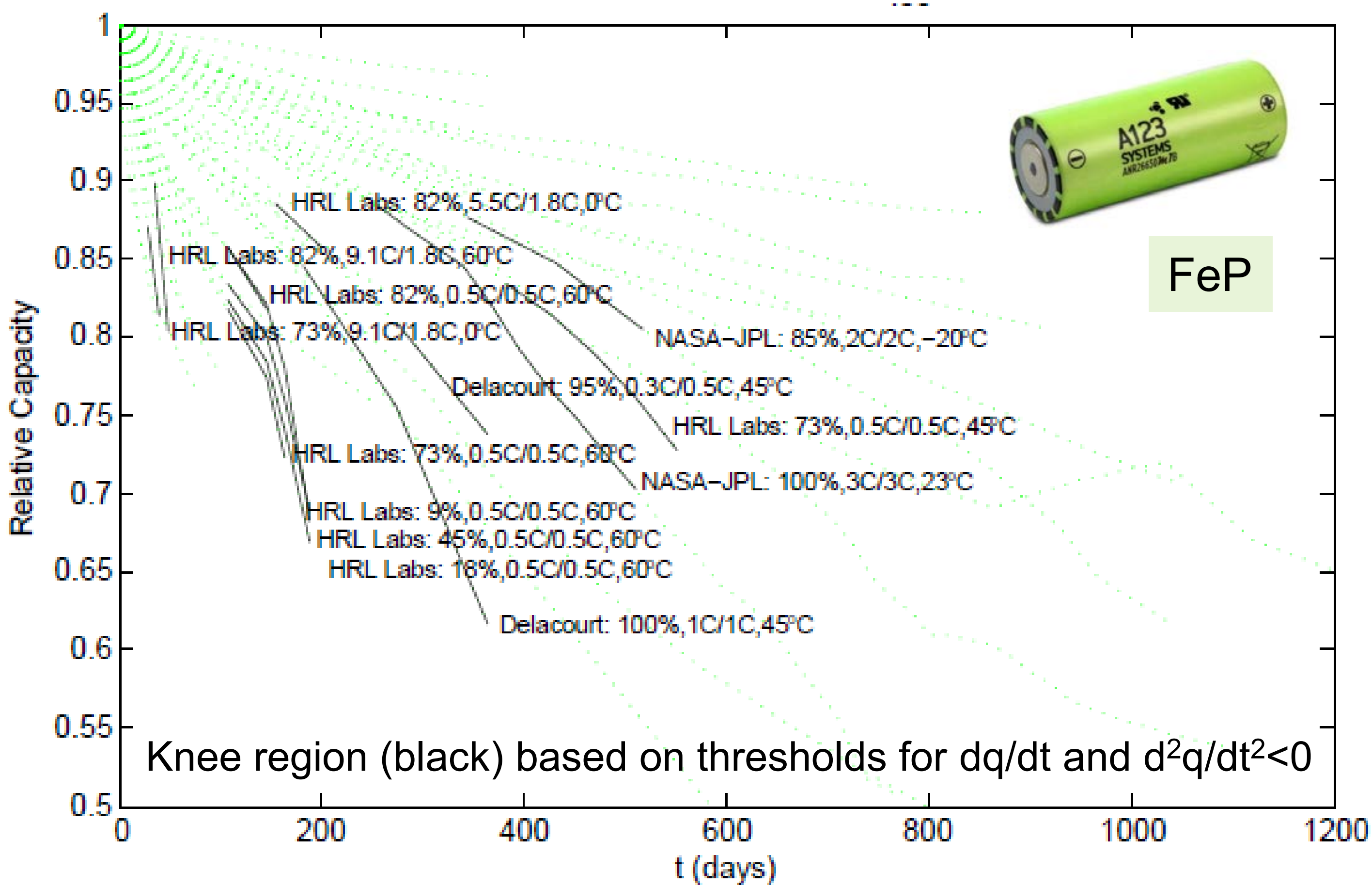
- Grains → particles → particles + PVDF + carbon black → composite electrode → neg./sep./pos. electrode sandwich → jellyroll → cell
- Accompanying property measurement

4) Linkage of mechanical stress with life (capacity, resistance)

Group	Reference	Length-scale	Material	Experiment	Model	Findings
GM	Qi, JES 2010	Atomic	Graphite		X	
Brown						
Harva						
Harva						
ASU						
GM,U						
GM,U						
UNO						
CU-Bo						
CU-Bo						
Umich						
Brown						
LBNL						
Aache						
LBNL						
SCaro						
MIT						
MSU						Multi-scale approach: Newman model + 2D mech model of particle/sandwich array
						Accurate separator stress prediction will require visco-elastic relaxation and thermal stress

Active Site Loss Visible in Cell Aging Data

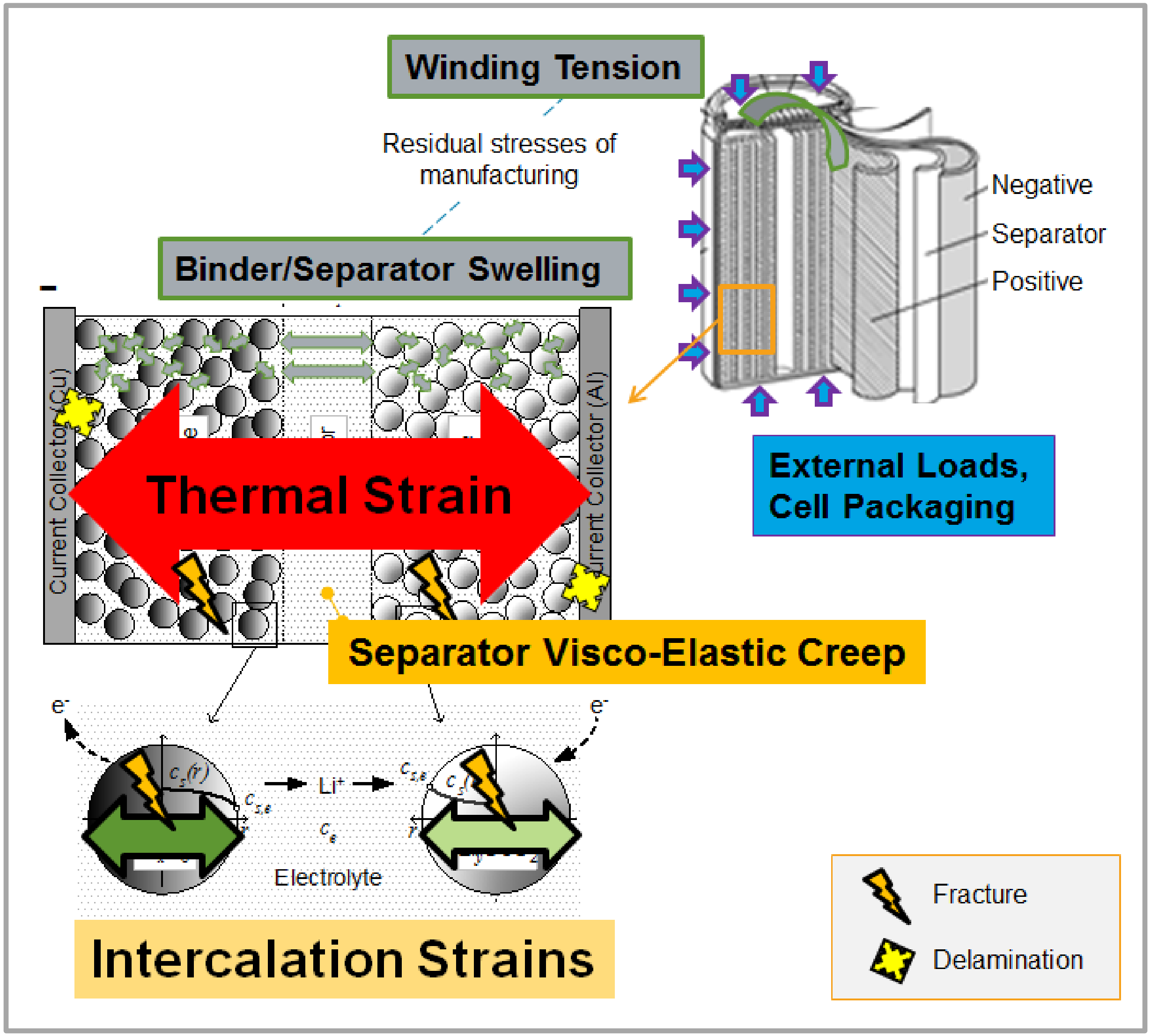
- Iron-phosphate meta-dataset combines tests from multiple labs – 50+ tests
- In "knee-region" of capacity fade data below, graphite site loss (mechanical process) has exceeded Li loss (predominantly chemical process)



Mechanical Stress Effects Contributing to Active Site Loss

Dependence on operating parameters:

- **C-rate** (intercalation gradient strains)
- **DOD** (bulk intercalation strains)
- **Low T** (exacerbates Li intercalation-gradients)
- **High T** (exacerbates polymer creep of binder, separator)
- **ΔT** (thermal strains)

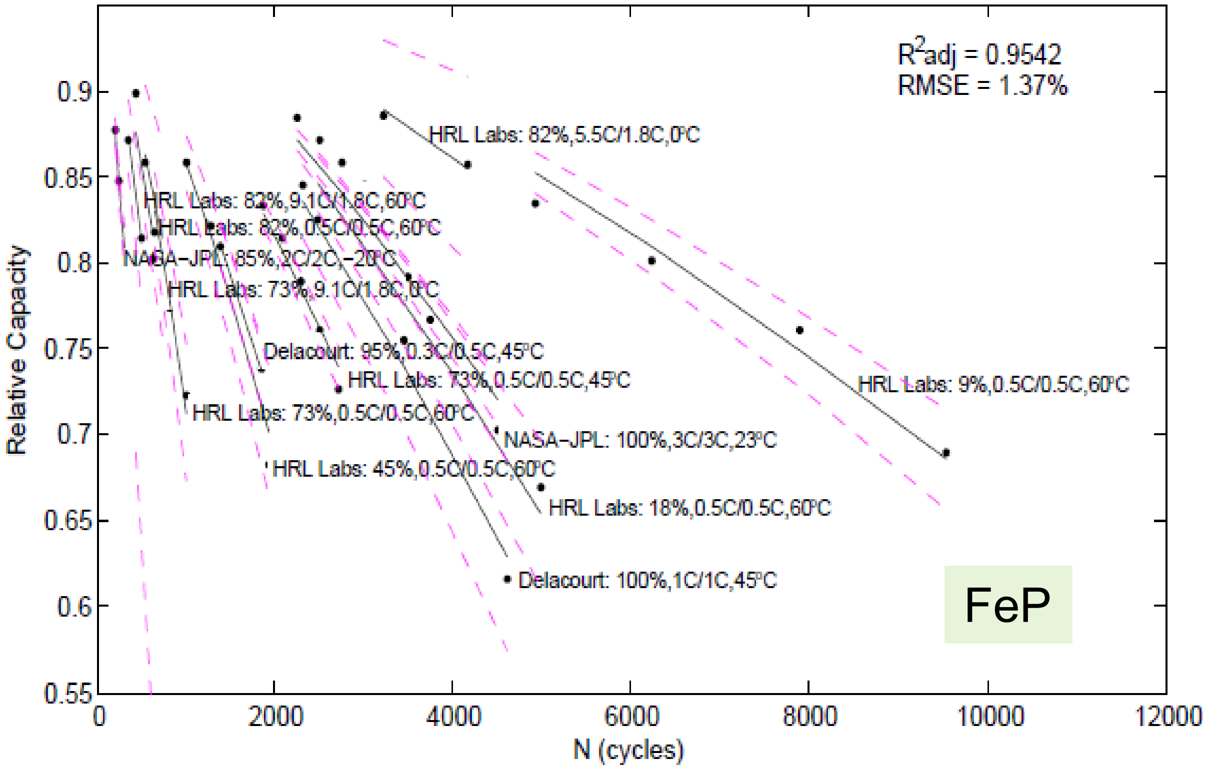


Hypothesized Active Site Loss Model

$$q = \min(q_{Li}, q_{sites}).$$

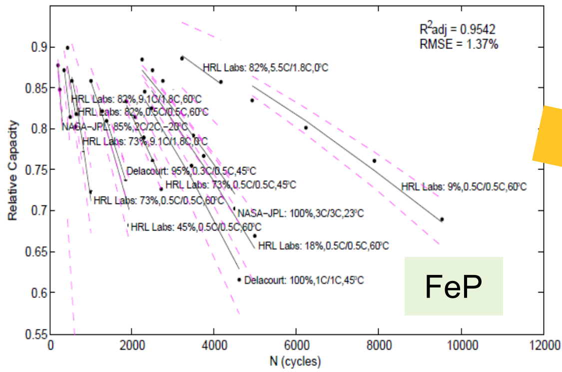
$$\boxed{q_{Li} = b_0 + b_1 t^z + b_2 N} \qquad \boxed{q_{sites} = c_0 + c_2 N}$$

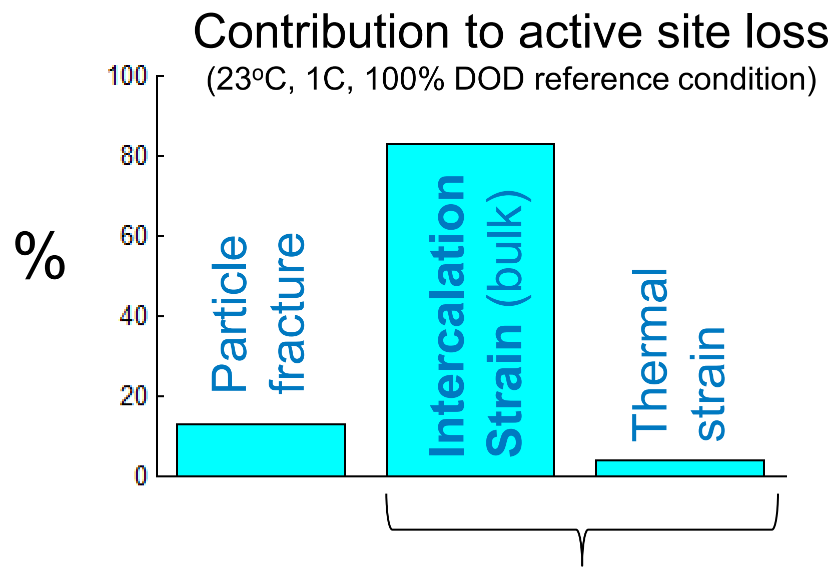
$$c_2 = c_{2,ref}\left\{ \underbrace{\exp\left(\frac{-E_a^{\text{binder}}}{R}\left(\frac{1}{T}-\frac{1}{T_{ref}}\right)\right)}_{\text{accelerated polymer failure at high T}}\left[\underbrace{m_1\, DOD}_{\text{bulk intercalation strain}} + \underbrace{m_2\,\Delta T}_{\text{bulk thermal strain}}\right] + \underbrace{m_3 \exp\left(\frac{-E_a^{\text{intercal.}}}{\mathrm{R}}\left(\frac{1}{T}-\frac{1}{T_{ref}}\right)\right)\left(\frac{C_{rate}}{C_{rate,ref}}\right)\left(\sqrt{\frac{t_{pulse}}{t_{pulse,ref}}}\right)}_{\text{intercalation gradient strain, accelerated by low temperature}} \right\}.$$

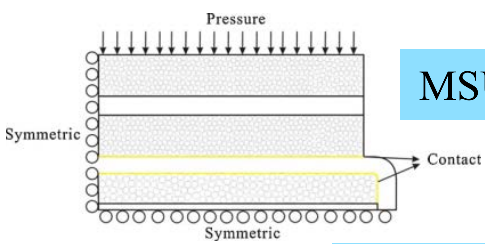


Model successfully describes 13 aging conditions from 0°C to 60°C

Bulk Strains Show Strongest Correlation with Capacity Fade in Knee Region







MSU/Xiao (2011)

Princeton/Peabody (2014)

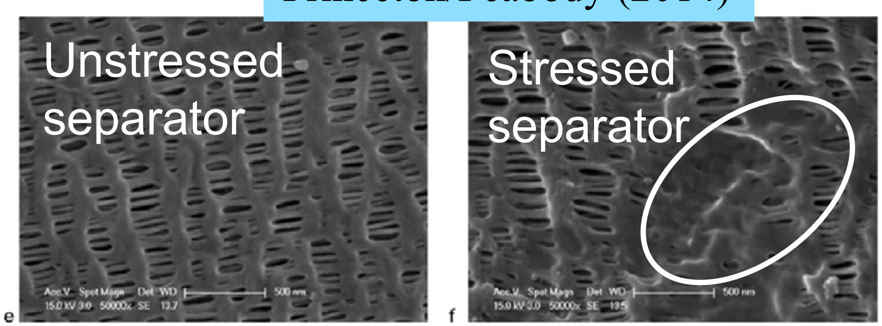
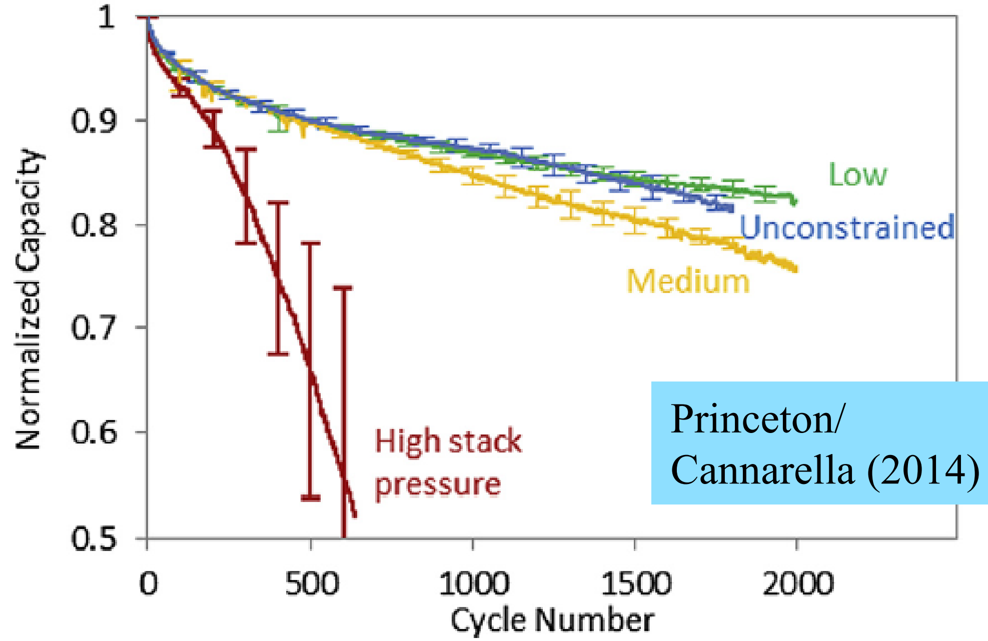


Fig. 3. Images of viscoelastic creep induced pore closure. SEM characterization of separator membranes after compressive stress testing. (a) Unstressed, (b) 5 MPa, (c) 10 MPa, (d) 30 MPa from separator only tests. Increasing total strain associated with higher stress decreases the pore areal density. (e) Unstressed separator from 90 mAh battery, (f) separator from a 90 mAh battery stressed at 30 MPa. The presence of electrolyte does not prevent stress-induced pore closure. The scale bar represents 500 nm for all of the images.

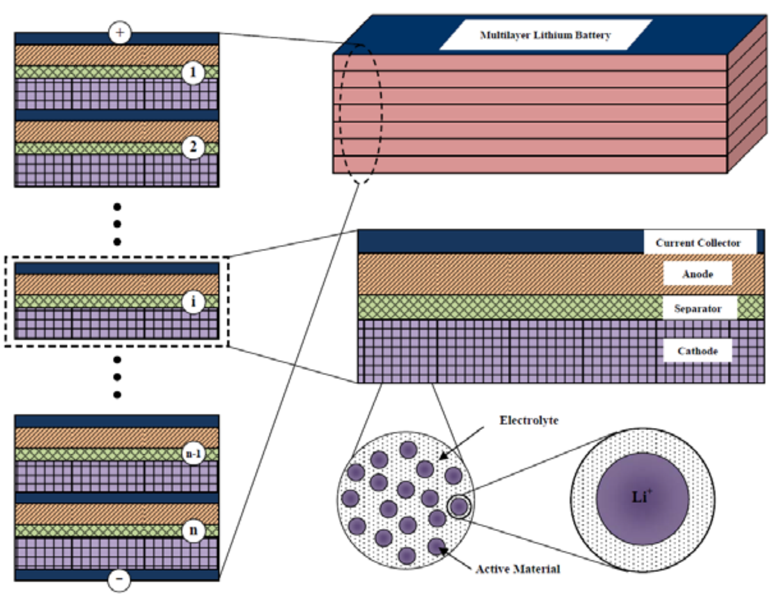
Important factors to capture in 3D CAE models to optimize cell lifetime, mechanical constraint

3D Electrochemical/Thermal/Mechanical Model

- **Objective:** Create cell design tool to predict lifetime, optimize mechanical packaging of large format cells



Multi-layer Li-ion battery discretized with solid shell elements



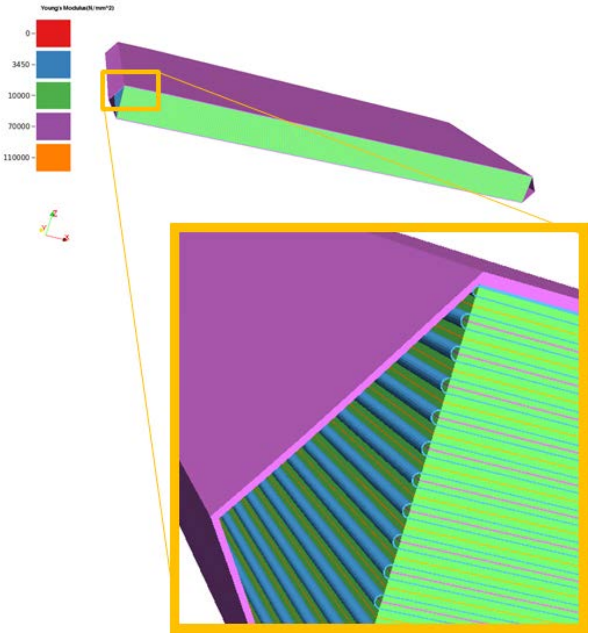
- **Approach:**

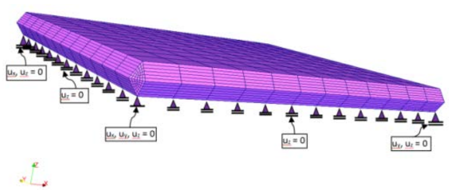
NREL MSMD Electrochemical/Thermal

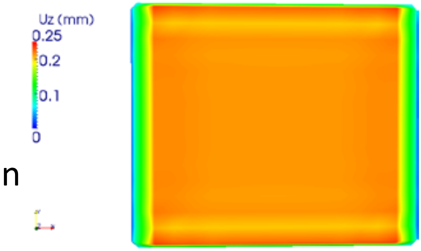
CU–Boulder Solid Mechanics

Echem/Thermal/Mechanical Model – Pouch Cells

- **ΔSOC & ΔT dependent stress (cell level) more significant contributor to capacity loss than diffusion-induced gradients (particle level)**
- **CU-Boulder and NREL developed multi-physics 3D model of commercial pouch cell**
- **Calibrated model versus electrical/thermal performance and measured changes in cell thickness with SOC, T, age**
- **Largest stresses at edge of electrode stack with separator wind**
- **Non-negligible in-plane displacements**
- **More significant stress/strain effects induced by electrochemical/thermal bulk changes rather than 3D gradients across the cell**
- **Temperature rise from electrochemical/thermal model important for capturing magnitude of mechanical strain**

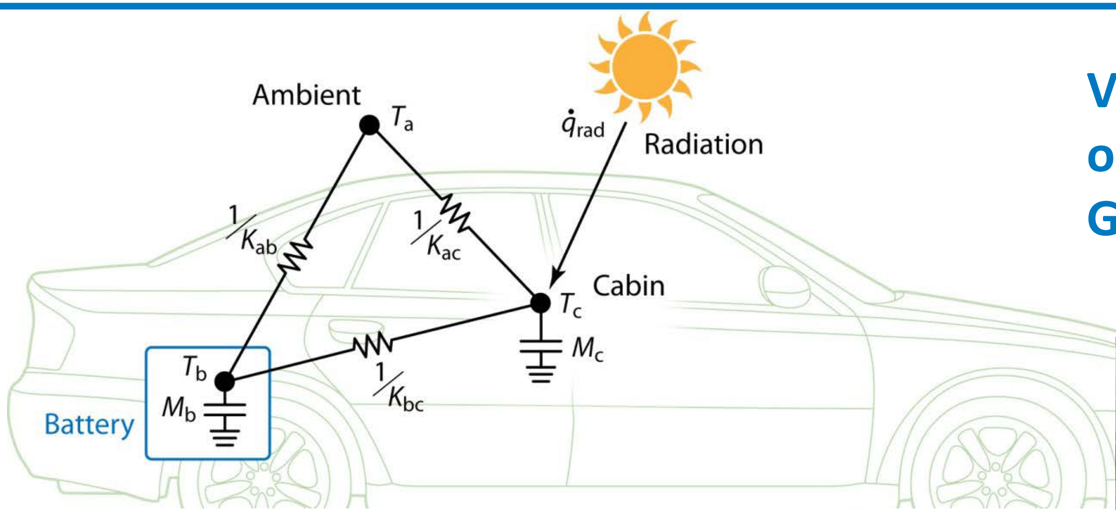






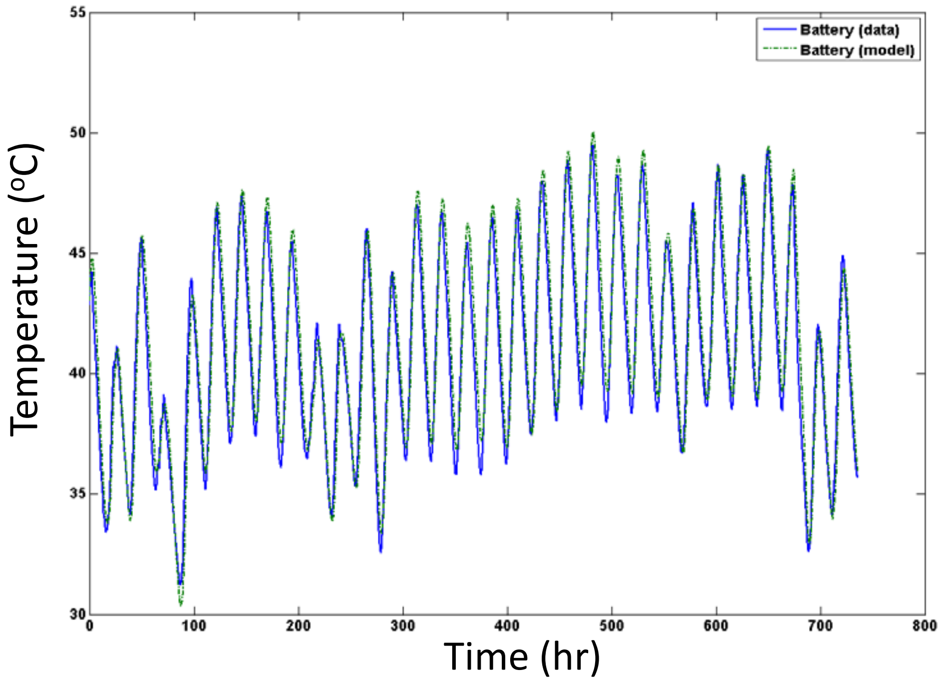
Displacement in thickness direction during charging

Capturing Vehicle & Ambient Impacts on Life



Vehicle thermal model fit to 3 days of Gen II Prius data recorded in Golden, CO in winter

Photo credits: Kandler Smith NREL

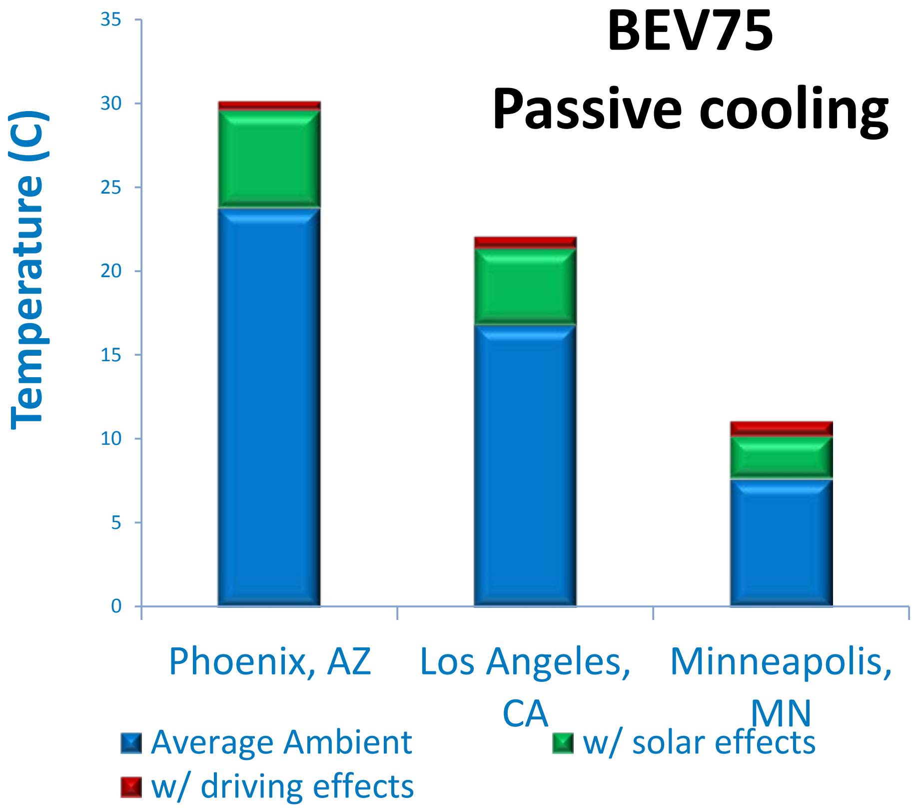


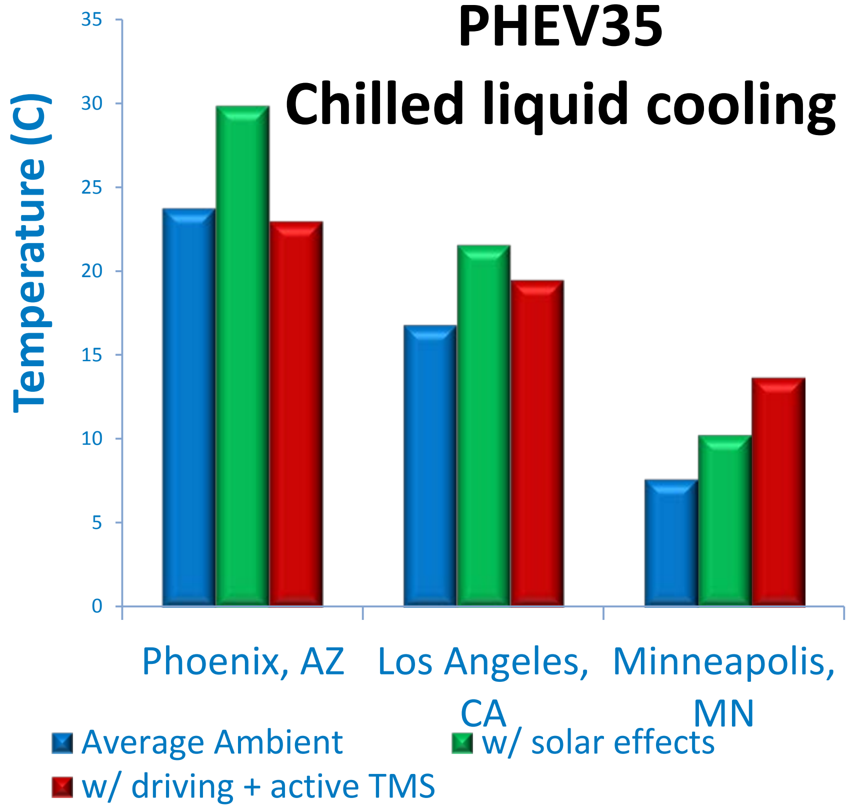
Same model predicts Prius battery temperature fluctuation in Phoenix, AZ

- **Winter: within ½ °C**
- **Summer: within 1°C**

(Passenger cabin fluctuations within 6°C)

Ambient Effects on Battery Average Temperature



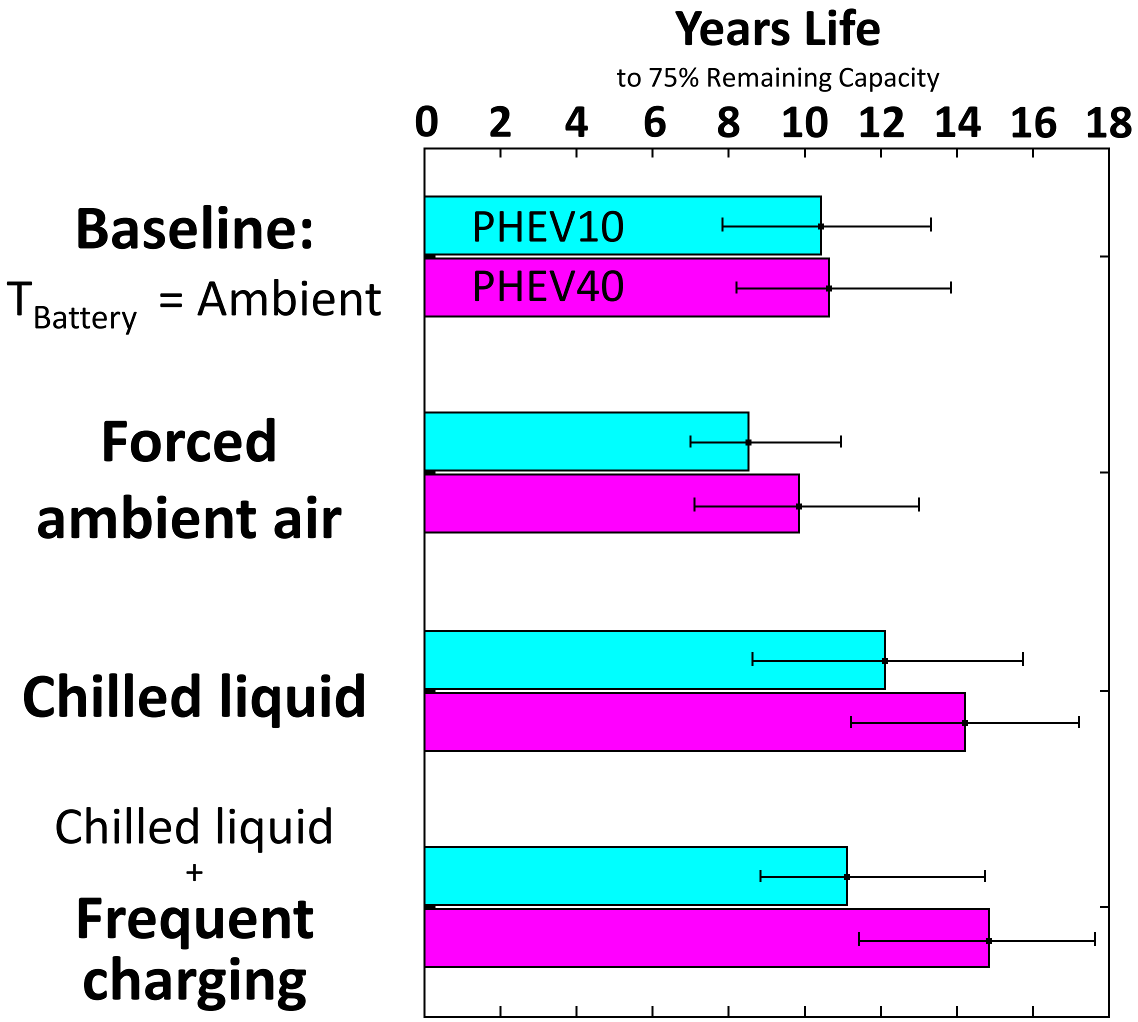


- **Ambient conditions dominate**
- **Thermal connection with passenger cabin, parking in shaded structures strongly influence battery life**

- **Battery temperature and lifetime weakly coupled to ambient conditions**

PHEV Life Variability – Phoenix, Arizona

Simulation of 782 drive cycles. Error bars show 5th to 95th percentile drive cycles



Lifetime Variation

Drive cycle/annual mileage: **±25%**

Value of chilled liquid vs. forced air battery thermal management:

- PHEV10: **+34%**
- PHEV40: **+42%**

(equates to \$500-\$600 savings in reduced pack total energy at \$300/kWh)

Frequent charging:

- PHEV10: **-8%** (more cycles)
- PHEV40: **+4%** (shallower cycles)

Sub-Ambient Standby Cooling Topologies

- Options: Chilled liquid, air, refrigerant evaporative plate or thermoelectrics (TE)
- Shown here: TE device on busbars

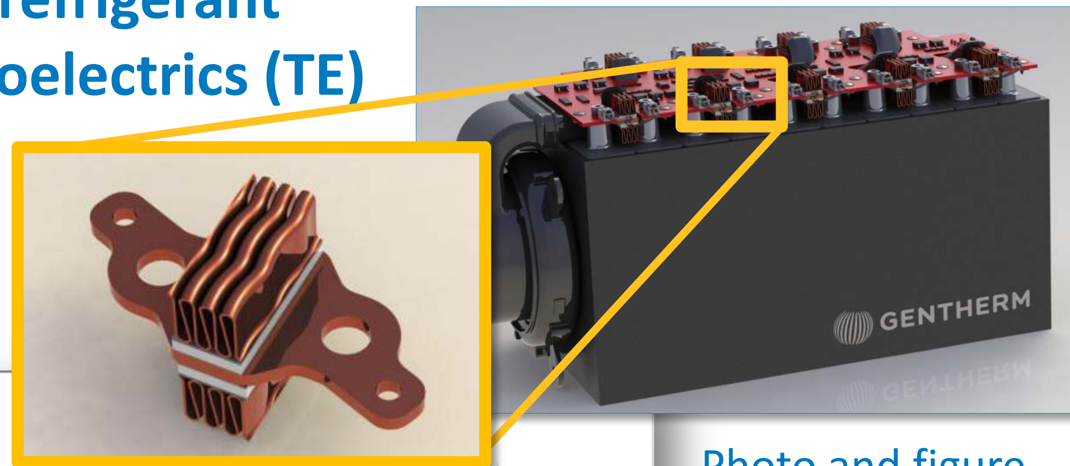
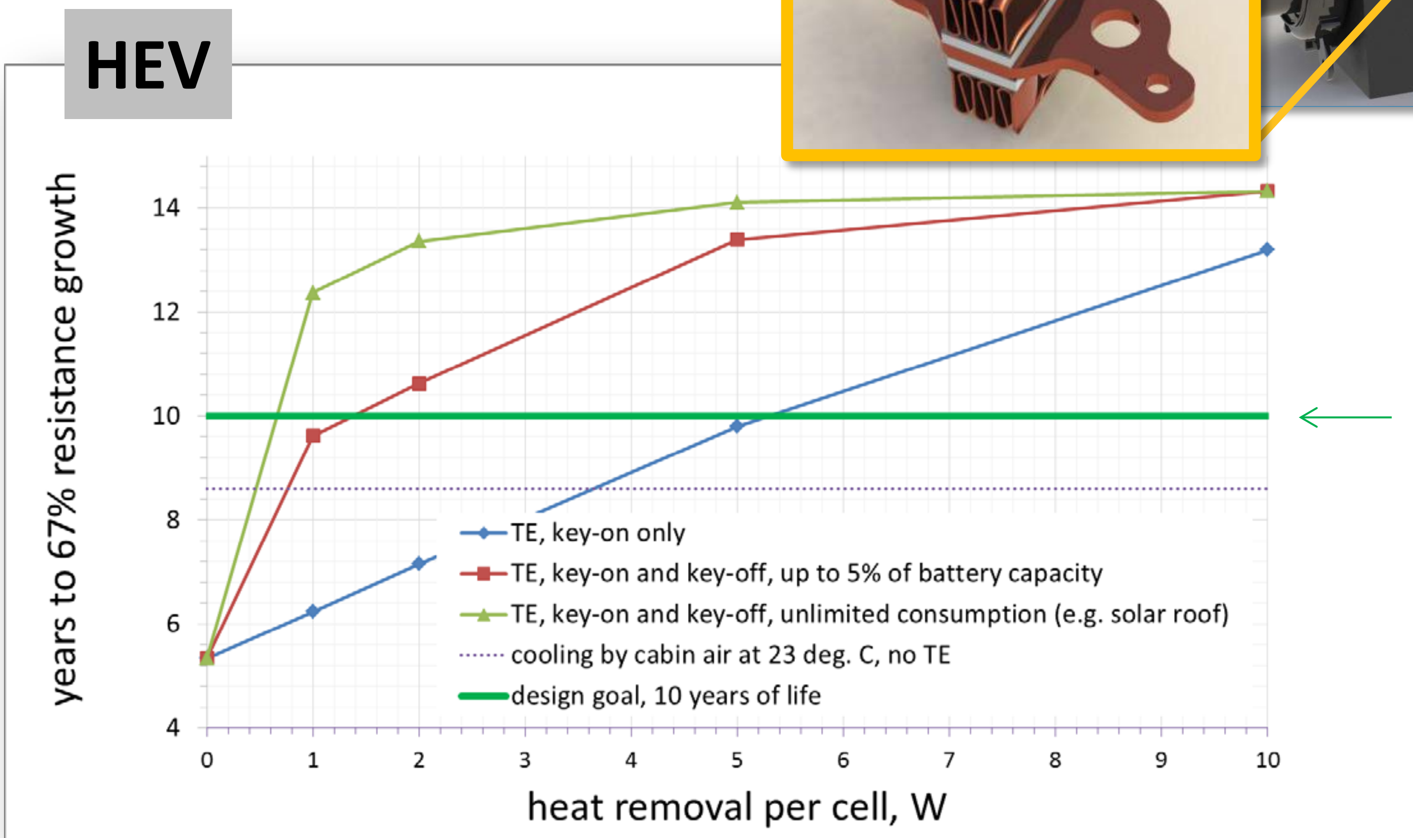
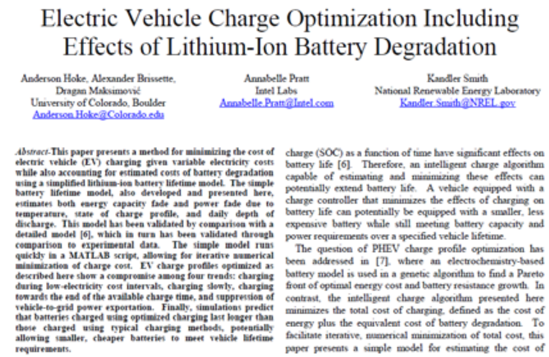


Photo and figure credit: Gentherm



10 yr target life

Optimized Charging Strategies



Electric Vehicle Charge Optimization Including Effects of Lithium-Ion Battery Degradation

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Abstract-This paper presents a method for minimizing the cost of electric vehicle (EV) charging given variable electricity costs while also accounting for estimated costs of battery degradation using a simplified lithium-ion battery lifetime model. The simple battery lifetime model, also developed and presented here, estimates both energy capacity fade and power fade due to temperature, state of charge profile, and daily depth of discharge. This model has been validated by comparison with a detailed model [6], which in turn has been validated through comparison to experimental data. The simple model runs quickly in a MATLAB script, allowing for iterative numerical minimization of charge cost. EV charge profiles optimized as described here show a compromise among four trends: charging during low-electricity cost intervals, charging slowly, charging towards the end of the available charge time, and suppression of vehicle-to-grid power exportation. Finally, simulations predict that batteries charged using optimized charging last longer than those charged using typical charging methods, potentially allowing smaller, cheaper batteries to meet vehicle lifetime requirements.

charge (SOC) as a function of time have significant effects on battery life [6]. Therefore, an intelligent charge algorithm capable of estimating and minimizing these effects can potentially extend battery life. A vehicle equipped with a charge controller that minimizes the effects of charging on battery life can potentially be equipped with a smaller, less expensive battery while still meeting battery capacity and power requirements over a specified vehicle lifetime.

The question of PHEV charge profile optimization has been addressed in [7], where an electrochemistry-based battery model is used in a genetic algorithm to find a Pareto front of optimal energy cost and battery resistance growth. In contrast, the intelligent charge algorithm presented here minimizes the total cost of charging, defined as the cost of energy plus the equivalent cost of battery degradation. To facilitate iterative, numerical minimization of total cost, this paper presents a simple model for estimating the cost of

CU-Boulder/Hoke (2014)

- **Reduce time spent at high SOC (delay charging)**
- **Avoid high C-rates to lower peak temperatures**

Cost function

$$c_{x,T} = c_{bat} \cdot \left(\underbrace{\int_{t_{ch}} \frac{1}{8760 \cdot L_x\left(T_{amb} + R_{th} \cdot |P(t)|\right)} dt}_{\Delta L/L \text{ due to charging}} + \underbrace{\frac{t_{max} - t_{ch}}{8760 \cdot L_x\left(T_{amb}\right)}}_{\Delta L/L \text{ while plugged in but not charging}} - \underbrace{\frac{t_{max}}{8760 \cdot L_x\left(P_{min} \cdot R_{th} + T_{amb}\right)}}_{\text{Baseline } \Delta L/L \text{ that would be expended by slow charging}} \right)$$

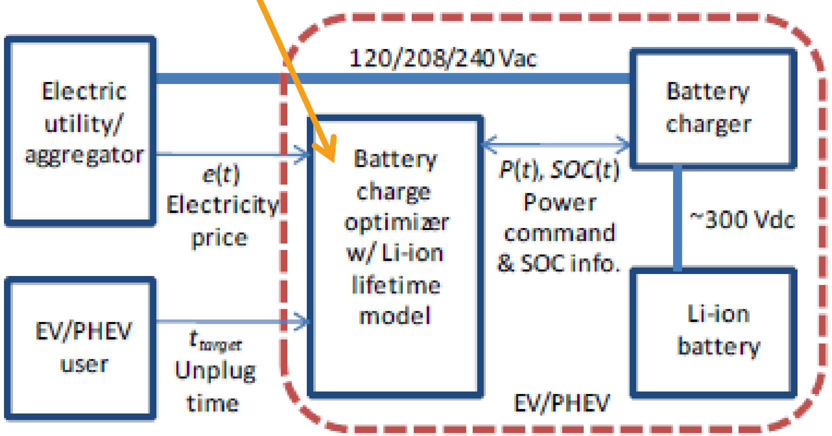
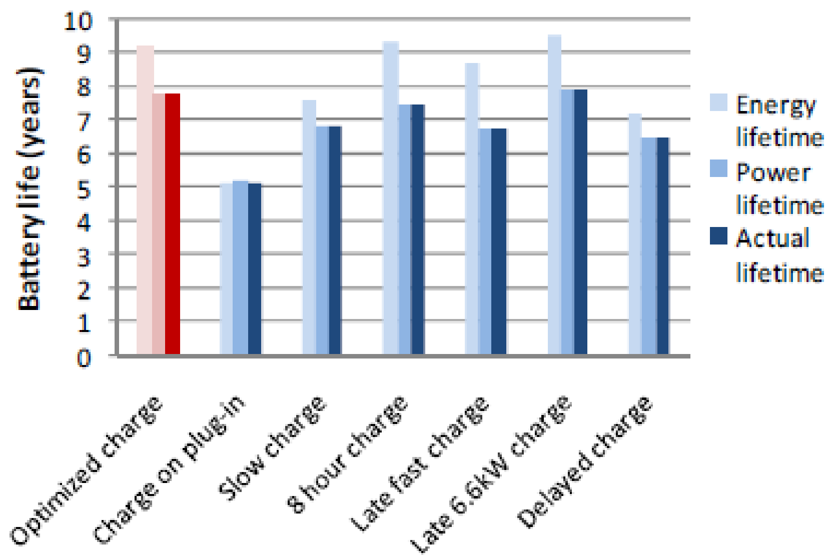
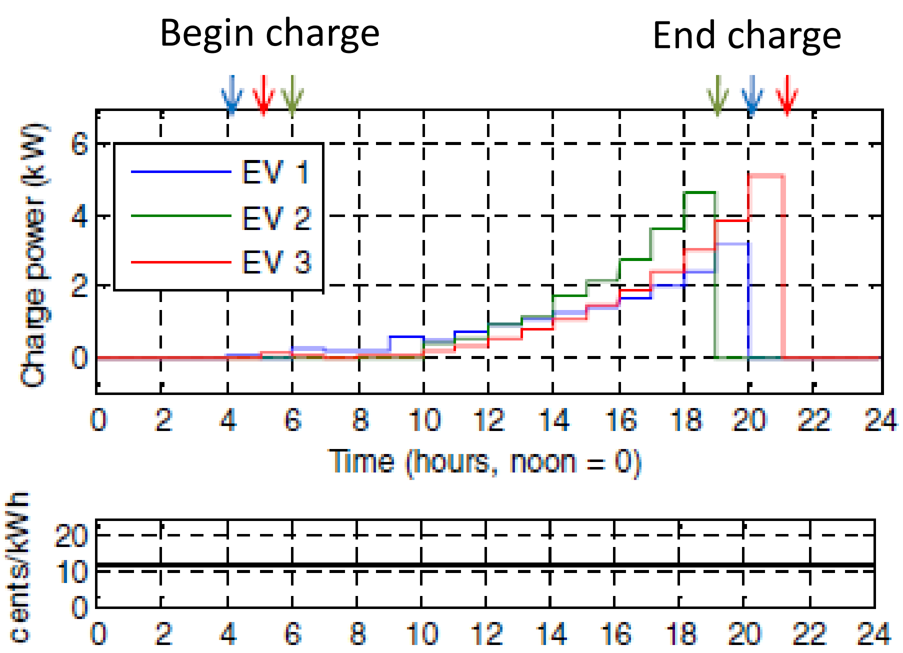


Fig. 1. EV/PHEV charge optimization schematic

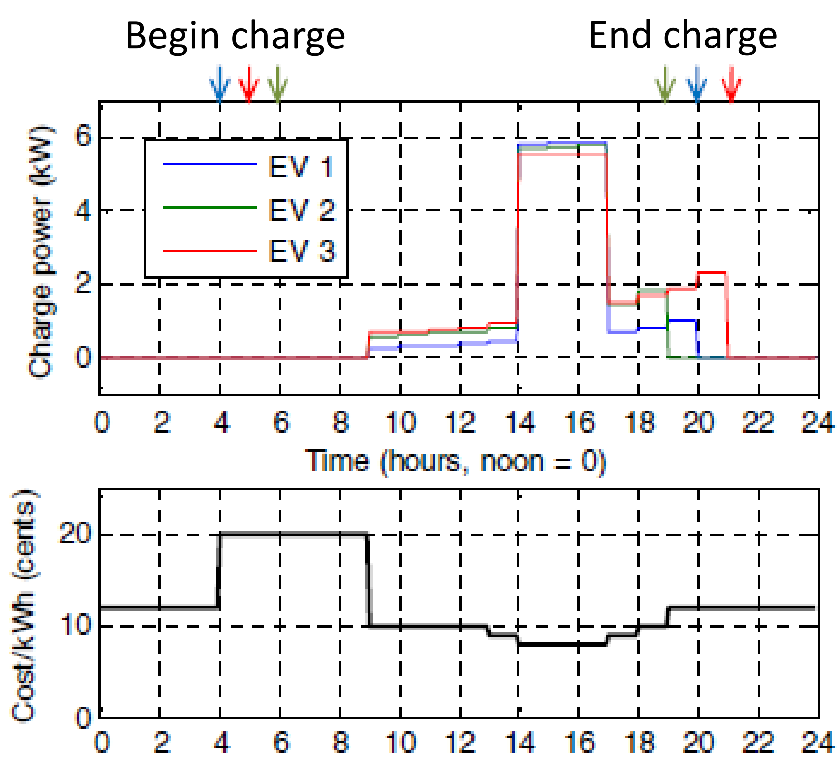


Optimized Charging Strategies

A) Constant energy cost



B) Variable energy cost



- Delayed charging best
- Response to price signals
- No V2G energy exported until electricity price $0.50/kWh

ARPA-E AMPED: Battery Management

Advanced Management and Protection of Energy Storage Devices

- Develop advanced sensing and control technologies to provide new innovations in safety, performance, and lifetime for grid-scale and vehicle batteries.

Eaton Corporation

Project: Downsized HEV pack by 50% through enabling battery prognostic & supervisory control while maintaining same HEV performance & life

NREL: Life testing/modeling of Eaton cells; controls validation on Eaton HEV packs

Utah State/Ford

Project: 20% reduction in PHEV pack energy content via power shuttling system and control of disparate cells to homogenous end-of-life

NREL: Requirements analysis; life model of Ford/Panasonic cell; controls validation of Ford PHEV packs

Washington Univ.

Project: Improve available energy at the cell level by 20% based on real-time predictive modeling & adaptive techniques

NREL: Physics-based cell-level models for MPC; implement WU reformulated models on BMS; validate at cell & module level

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