Model-Based Design and Integration of Large Li-ion Battery Systems

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Outline

• DOE’s CAEBAT program

• Battery physics
  o Performance
  o Degradation
  o (Safety – omitted here. See Dr. Gi-Heon Kim’s separate presentation at Battery Safety this week)

• Selected NREL modeling research

• Gaps and next efforts in model development
DOE’s CAEBAT Program

- Shorten time and cost for design of electric drive battery systems
- Integrate accomplishments of disparate battery modeling activities. Make them accessible as design tools for industry
- Led by Vehicle Technologies Office with support of US Army TARDEC
  - CAEBAT-1 (2010): Electrochemical-thermal (ECT)
  - CAEBAT-2 (2013): + computational efficiency, mechanical crush
  - CAEBAT-3 (2015): + microstructure
- Teams combining industry, national labs, universities
- Three commercially available toolsets with >60 licenses to date
Performance of Lithium-Ion Batteries Occurs Across Varied Length Scales

Practical computer-aided engineering (CAE) tools require fast, efficient frameworks and sub-models including reduced order models.
Degradation Similarly Occurs Across Various Length Scales

Chemistry
- SEI growth
- Li plating
- Electrolyte decomposition
- Gas generation

Particle
- Surface fracture, active area growth
- Bulk fracture, damage of transport paths
- Phase evolution, voltage droop

Electrode
- Particle displacement, electrode creep, delamination, isolation
- Separator pore closure
- Salt precipitation
- Pore clogging

Cell
- 3D electrical, thermal, mechanical non-uniformity
- Tab effects
- Stack/wind

System
- Thermal & mechanical non-uniformity & boundary conditions
- Electrical duty-cycle

Not all degradation modes are fully understood. Life can be predicted, but only with sufficient cell aging test data.
NREL Modeling & Research

• Fast electrochemical simulation
• Framework for efficient extension of electrochemistry to 3D cell & pack domains
• Chemical reaction modeling: SEI growth & Li plating
• Mixed material electrodes
• Mechanics:
  • Particle & electrode diffusion-induced damage
  • Cell-scale pressure management
Fast Electrochemical Simulation

Frequency domain technique used to four PDEs governing electrochemical dynamics to a set of ~13 ODEs

- **Previous**: Model reduction took 1-2 hours, only represented one battery design
- **Accomplishment**: Single pre-calculated reduced model valid for all battery designs

100x faster than typical finite-volume models. Similar speed as circuit models, but also predicts electrochemical potentials & concentrations based on design parameters

\[ \dot{x} = f(x, u) \]
\[ y = h(x, u) \]

Extending Electrochemistry to Cell and Pack

NREL Multi-Scale Multi-Dimensional (MSMD) Model
Modular architecture, linking interdisciplinary battery physics

MSMD Realizations in Various Geometries

Stack Pouch

Wound Prismatic

Wound Cylindrical

Elementary Chemical Reactions w/ CSM


Validates square-root-of-time SEI growth models
Elementary Chem. Reactions on Arbitrary Geometry

- Overcome limiting assumption of homogenization in most battery models (e.g. Li plating)

Steps encapsulated in python script `jpg2dxf.py`

- `crop`
- `threshold`
- `smooth` ("morphological opening")
- `vectorize`

Steps to convert an SEM image to a computational mesh

Extremely Fine                       Moderate         Extremely Coarse

Study of surface-effects by varying geometry threshold value

Electrolyte Distribution within the anode during charge
Modeling of Mixed Material Electrodes

- Multiple chemistries, particle sizes, morphologies often blended for optimal power/energy/life characteristics
- MSMD Discrete-Diffusion Particle Models
  - Sphere
  - Rod
  - Flake
  - Arbitrary 3-D

http://www.caer.uky.edu/elecrochemical/research/research.shtml
Particle-to-Electrode ECM Models w/ TAMU

- Concentration gradient drives particle fracture
- Inhibits diffusivity and performance

- Order-reduced and integrated in electrode-scale models

<table>
<thead>
<tr>
<th>Lithiation → 4C</th>
<th>Delithiation → 4C</th>
</tr>
</thead>
<tbody>
<tr>
<td>mol/m³</td>
<td>mol/m³</td>
</tr>
<tr>
<td>Y direction (x10^-5) [m]</td>
<td>Y direction (x10^-5) [m]</td>
</tr>
<tr>
<td>X direction (x10^-5) [m]</td>
<td>X direction (x10^-5) [m]</td>
</tr>
</tbody>
</table>

- Raw data
- Non-dimensional ROM

- Order-reduced and integrated in electrode-scale models

Table 2. Scaling Factor and Fitting Parameter in Eq. (12)

<table>
<thead>
<tr>
<th>Relation</th>
<th>a</th>
<th>b</th>
<th>M</th>
</tr>
</thead>
<tbody>
<tr>
<td>CSE and C (T &gt; 0°C)</td>
<td>0.01942</td>
<td>0.35</td>
<td>M₁ = \left( \frac{C \cdot \text{Rate}}{T} \right)^{0.34}</td>
</tr>
<tr>
<td>CSE and C (T &lt; 0°C)</td>
<td>0.01942</td>
<td>0.35</td>
<td>M₂ = \left( \frac{C \cdot \text{Rate}}{T} \right)^{0.34}</td>
</tr>
<tr>
<td>CSE and Microcrack Density (T &gt; 0°C)</td>
<td>0.0015</td>
<td>0.657</td>
<td>M₃ = \left( \frac{C \cdot \text{Rate}}{T} \right)^{0.26}</td>
</tr>
<tr>
<td>CSE and Microcrack Density (T &lt; 0°C)</td>
<td>0.0016</td>
<td>0.8443</td>
<td>M₄ = \left( \frac{C \cdot \text{Rate}}{T} \right)^{0.23}</td>
</tr>
</tbody>
</table>

Cell Electrochemo-Mechanical Model w/ CU-B


Impact of severe pressure on separator

Strain at end of full charge

Through-plane

Vertical direction
Cell Resistance/Capacity Life Model

- Surrogate models for physical mechanisms regressed to aging test data
- Integrated in control algorithms and BLAST systems analysis model

- SEI growth & damage
- Particle fracture
- Electrode isolation
- Electrolyte decomposition
- Gas generation & delamination
- Li plating

<table>
<thead>
<tr>
<th>Mechanism</th>
<th>Trajectory equation</th>
<th>State equation</th>
<th>Fitted parameter</th>
<th>Physics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffusion-controlled reaction</td>
<td>( x(t) = k \cdot t^{1/2} )</td>
<td>( \dot{x}(t) = \frac{k}{2} \cdot x(t) )</td>
<td>( k ) - rate ((p=1/2))</td>
<td>(E)(C)(T)(M)</td>
</tr>
<tr>
<td>Kinetic-controlled reaction</td>
<td>( x(t) = k \cdot t )</td>
<td>( \dot{x}(t) = k )</td>
<td>( k ) - rate ((p=1))</td>
<td>(E)(C)(T)</td>
</tr>
<tr>
<td>Mixed diffusion/kinetic</td>
<td>( x(t) = k \cdot t^{p} )</td>
<td>( \dot{x}(t) = k \cdot \left( \frac{k}{x(t)} \right)^{p} )</td>
<td>( k ) - rate ((p=0)), (0.4 &lt; c &lt; 1)</td>
<td>(E)(C)(T)(M)</td>
</tr>
</tbody>
</table>

Illustration by Josh Bauer, NREL

Liquid cooling, chilled fluid
Air cooling, low resistance cell
No cooling

Relative Capacity

Phoenix, AZ ambient conditions
33 miles/day driving, 2 trips/day

Calendar fade
- SEI growth (partially suppressed by cycling)
- Loss of cyclable lithium
- \( a_0(\Delta DOD, T,V) \)

Cycling fade
- Active material structure degradation and mechanical fracture
- \( a_2(\Delta DOD, T,V) \)

Relative Capacity

\( R = a_1 \cdot t^{1/2} + a_2 \cdot N \)

Relative Resistance

\( Q = \min (Q_{Li}, Q_{active}) \)

Gr/NCA

Gr/FeP

\( R^2_{adj} = 0.5611 \)
RMSE = 0.041
Life Model Validation at Pack Level

ARPA-E AMPED project led by Eaton Corporation (PI Dr. Chinmaya Patil)
- Demonstrating 30% smaller Eaton HEV battery with prognostic-based control
- Model accuracy maintained from cell-to-pack level (2-3% capacity, 7% resistance)

Cell Model Identification
- 25 cells, 6 months
- Constant temperature & cycling

Pack Model Validation
- Cell model + temperature distribution
- 4-season temperature & variable cycling

- Resistance Model Error (%)
  - Time (months)
  - Capacity Model Error (%)
  - Time (months)
Filling the Gaps

• Electrode microstructure simulation
• ECT parameter identification
3D Microstructure Model: Overcoming Limitations of Today’s 1D Porous Electrode Models

- Enable virtual design of battery electrodes to shorten design cycle
- Create platform to explore new physics and geometries

Stochastic reconstruction & meso-scale physics

Electrode Design Inputs

Microstructure Model
- Geometry
- Physics
- High Performance Computing

Effective properties for upscaling

Electrode fabrication, Tomography, electrochemical testing

Validated electrochemical performance
Electrochemical/thermal parameter identification is an intrinsically under-determined problem. NREL is developing sequential approach starting from smallest length scale with appropriate model at each length scale regressed to data.
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