Computer-Aided Engineering of Batteries for Designing Better Li-Ion Batteries

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Energy Storage R&D Program

- **DOE ES Charter:** Advance the development of batteries and electrochemical energy storage devices to enable a large market penetration of hybrid and electric vehicles.
- **DOE ES Focus:** Increase performance at lower cost while meeting weight, volume, and safety targets.

**NREL ES Objective:** Support DOE and industry to achieve energy storage targets through research and development, testing, analysis, design, and modeling.
Modeling and Design Tools

Most models are wrong, but some are useful.....

• Simulation and computer-aided engineering (CAE) tools are widely used to speed up the research and development cycle and reduce the number of build-and-break steps.

• Use of CAE tools has enabled the automakers to reduce product development cost and time while improving the safety, comfort, and durability of many components and vehicles.

• There is a need to have several user-friendly, 3D, fully-integrated CAE software tools available for the battery community across many scales.

[Diagram showing various levels and models related to battery design and simulation]
The Current State of Battery Modeling

- There are a number of battery models in academia, national labs, and industry, but they either
  - Include relevant physics details, but mostly neglect engineering complexities, or
  - Include relevant macroscopic geometry and system conditions in 3D, but use too many simplifications in fundamental physics.
- There are a number of custom battery codes available; however, they require expert users.
- The Battery Design Studio software suite has been in the forefront of battery simulations and now is being integrated into CD-adapco’s CAE environment.
- Validation of data is the key to building confidence.
The Current State of Battery Modeling

• With DOE funding, national laboratories, industry, and universities have developed many models for simulating lithium-ion battery (LIB)
  o cost,
  o life,
  o performance (electro-thermal, electrochemical), and
  o abuse reactions.

• So far, these models have not been fully integrated into 3D CAD for design of battery packs and linked with ease, especially for engineering purposes.

• Realizing the need, DOE has initiated the CAEBAT project to bring together these battery models to develop suites of battery CAE tools.
DOE’s CAEBAT Program

- **Objective:** Incorporate existing and new models into software tools for design of cells and packs.
- **Goal:** Shorten development and design cycles and optimize batteries for improved performance and safety, long life, and low cost.
NREL’s Role in the CAEBAT Project

1. As project coordinator, NREL supports DOE to achieve the CAEBAT objectives:
   - Provide input/support to DOE for the CAEBAT project plan
   - Coordinate activities among national laboratories
   - Support industry and universities through competitively-placed subcontracts
     - 50%-50% cost sharing with three teams
     - Work started in June 2011 to develop CAE tools

2. Enhance and further develop existing electrochemical, thermal, abuse reaction, and internal short circuit models for use by industry and CAEBAT participants.

<table>
<thead>
<tr>
<th>Model</th>
<th>Length Scale</th>
<th>Geometry</th>
<th>Physics/Application</th>
</tr>
</thead>
</table>
| Electro-thermal FEA & Fluid-dynamics (CFD) | 1D, 2D, & 3D | 1D, 2D, & 3D | • Electrical, thermal & fluid flow  
  • Performance, detailed cooling design  
  • Commercial software (restrictive assumptions) |
| Electrochemical-thermal (MSMD) | 1D, 2D & 3D | 1D, 2D & 3D | • Electrochemical, electrical & thermal  
  • Performance, design               |
| Electrochemical-thermal-degradation (MSMD-life) | 1D, 2D & 3D | 1D, 2D & 3D | • Electrochemical, electrical & thermal  
  • Cycling & thermal-induced degradation  
  • Performance, design, life prediction |
| Thermal abuse reaction kinetics | Thermal network, 2D & 3D | Thermal network, 2D & 3D | • Chemical & thermal  
  • Safety evaluation               |
| Internal short circuit | 3D           | 3D             | • Chemical, electrical, electrochem. & thermal  
  • Safety evaluation               |
Battery Performance, Durability and Safety

Multi-physics Interactions Across Varied Length Scales

Electrode Scale
- Charge balance and transport
- Electrical network in composite electrodes
- Li transport in electrolyte phase

Cell Scale
- Electronic potential & current distribution
- Heat generation and transfer
- Electrolyte wetting
- Pressure distribution

Particle Scale
- Li diffusion in solid phase
- Interface physics
- Particle deformation & fatigue
- Structural stability

Module Scale
- Thermal/electrical inter-cell configuration
- Thermal management
- Safety control

System Scale
- System operating conditions
- Environmental conditions
- Control strategy

Atomic Scale
- Thermodynamic properties
- Lattice stability
- Material-level kinetic barrier
- Transport properties

[Diagram showing scales from atomic to system level with corresponding physical phenomena and length scales (10^-10 to 10^0 meters).]
Porous Electrode Model – Commonly Used

**Charge Transfer Kinetics at Reaction Sites**

\[ j^{Li} = a_i i_e \left\{ \exp \left[ \frac{\alpha_F}{RT} \eta \right] - \exp \left[ - \frac{\alpha_c}{RT} \eta \right] \right\} \]

\[ i_0 = k(c_e)^{\alpha_a} (c_{s,max} - c_{s,e})^{\alpha_s} (c_{s,e})^{\alpha_c} \quad \eta = (\phi_s - \phi_e) - U \]

**Species Conservation**

\[ \frac{\partial c_z}{\partial t} = \frac{D_z}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial c_z}{\partial r} \right) \]

\[ \frac{\partial (\varepsilon_e c_e)}{\partial t} = \nabla \cdot (D_T^{\text{eff}} \nabla c_e) + \frac{1 - i_0^2}{F} j^{Li} - \frac{i_e}{F} \nabla i_0 \]

**Charge Conservation**

\[ \nabla \cdot (\sigma^{\text{eff}} \nabla \phi_s) - j^{Li} = 0 \]

\[ \nabla \cdot (\kappa^{\text{eff}} \nabla \phi_s) + \nabla \cdot (\kappa^{\text{eff}}_D \nabla \ln c_e) + j^{Li} = 0 \]

**Energy Conservation**

\[ \rho c_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + q''' \]

\[ q''' = j^{Li} \left( \phi_s - \phi_e - U + T \frac{\partial U}{\partial T} \right) + \sigma^{\text{eff}} \nabla \phi_s \cdot \nabla \phi_s + \kappa^{\text{eff}} \nabla \phi_e \cdot \nabla \phi_e + \kappa^{\text{eff}}_D \nabla \ln c_e \cdot \nabla \phi_e \]

- Pioneered by John Newman’s group at the University of Berkeley (Doyle, Fuller, and Newman 1993)
- Captures lithium diffusion dynamics and charge transfer kinetics
- Predicts current/voltage response of a battery
- Provides design guide for thermodynamics, kinetics, and transport across electrodes

- Difficult to apply in large-format batteries where heat and electron current transport critically affect the battery responses
NREL’s MSMD Model Framework

Through the multi-year effort supported by DOE, NREL has developed a modeling framework for predictive computer simulation of LIBs known as the **Multi-Scale Multi-Dimensional (MSMD)** model that addresses the interplay among the physics in varied scales.

- Introduce *multiple computational domains* for corresponding length scale physics
- *Decouple LIB geometries* into separate computation domains
- *Couple physics* using the predefined inter-domain information exchange
- Selectively resolve higher spatial resolution for smaller characteristic length scale physics
- Achieve high computational efficiency
- Provide flexible & expandable modularized framework

Segregation of Time and Length Scales

Self-Balancing Nature

→ Continuum approach with thermodynamic representation for sub-domain system

Kinetic/dynamic representation

Electronic conductivity is much higher in metal current collectors than in composite electrode matrix

\( \sigma_{ce} \ll \sigma_{cc} \)

Lithium transport is much faster in liquid electrolyte than in solid particles

\( e.g., \ D_s \ll D_e \)

Geometry Decoupling

Domain Invariant

Domain Average
MSMD Protocol for Transferring Information

\[ \phi_s, \phi_e \]

\[ \phi_+ \]
\[ \phi_- \]

\[ \eta \]

\[ i''_\xi \]

\[ \bar{i}''_x, \bar{i}''_x(\Theta) = \frac{\int_{A_x} i''_x(\xi, x, \Theta) dA_x}{A_x} \]
MSMD Protocol for Transferring Information

\[ i''(x, \Theta) = \frac{\int_{A_x} i''(\xi, x, \Theta) dA_x}{A_x} \]
MSMD Protocol for Transferring Information

\[ T \]

\[ \Phi_+ \quad \Phi_- \]

\[ \phi_s \quad \phi_e \]

\[ c_e \]

\[ q_x'' \quad q_x''' \quad \bar{q}_x'' \quad \bar{q}_x''' \]

\[ \eta \quad c_s \]

\[ \bar{i}_x'' \]

\[ i''_{\xi} \]

\[ q''_{\xi} \quad q'''_{\xi} \]

\[ j''_{\xi} \]

\[ \Theta \]

\[ j''(\Theta) = \frac{\int_{A_x} i''(x, \Theta) dA_x}{A_x} \]

\[ q''(\Theta) = \frac{\int_{A_x} q''(x, \Theta) dA_x}{A_x} \]

\[ \bar{q}_x''(\Theta) = \frac{\int_{V_x} q''(x, \Theta) dV_x}{V_x} \]

\[ i''_{\xi}(x, \Theta) = \frac{\int_{A_{\xi}} i''_{\xi}(\xi, x, \Theta) dA_{\xi}}{A_{\xi}} \]

\[ q''_{\xi}(x, \Theta) = \frac{\int_{A_{\xi}} q''_{\xi}(\xi, x, \Theta) dA_{\xi}}{A_{\xi}} \]

\[ q'''_{\xi}(x, \Theta) = \frac{\int_{V_{\xi}} q'''_{\xi}(\xi, x, \Theta) dV_{\xi}}{V_{\xi}} \]
Hierarchical Architecture of MSMD

- **Modularized flexible framework** for multi-scale multi-physics battery modeling
- **Expandable development platform** providing “pre-defined but expandable communication protocol”

**Particle Domain**
- Charge transfer kinetics
- Li transport in active particles
- ...

**Electrode Domain**
- Charge balance in solid composite electrode matrix
- Charge balance in liquid pore channels
- Li transport in electrolyte
- ...

**Cell Domain**
- Energy conservation
- Charge conservation in current collectors
- ...

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Modularized hierarchical architecture of the MSMD model allows *independent development of submodels* for physics captured in each domain.

The modularized framework facilitates collaboration with external expertise.
NREL’s Cell-Domain Models: Orthotropic Continuum Model

Cell Domain Models

- **SPPC (Single Potential-Pair Continuum) model:** applicable to stack prismatic cells, tab-less wound cylindrical/(prismatic) cells:

- **MPPC (Multi Potential-Pair Continuum) model:** applicable to alternating stacked prismatic cells:

- **WPPC (Wound Potential-Pair Continuum) model:** applicable to spirally wound cylindrical/(prismatic) cells:

- **Lumped model:** applicable to small cells

- Discussed in this presentation
SPPC: Single Potential-Pair Continuum

Cell Composite

*Arbitrary finite volume of cell composite*

Orthotropic Continuum Model

$\Phi_{i,j}$

Positive current collector

Negative current collector

Stacked cell

Wound cell with continuous tab

$k_{ij} = (k_t - k_p) e_i'^t e_j'^t + k_p \delta_{ij}$

$\sigma^\text{eff}_{ij} = (\delta_{ij} - e_i'^t e_j'^t) \epsilon^- \sigma^-$

$\sigma^+_{ij} = (\delta_{ij} - e_i'^t e_j'^t) \epsilon^+ \sigma^+$
### MSMD Application to Prediction of Large Stacked Prismatic Cell Behavior

#### Submodel Choice

<table>
<thead>
<tr>
<th>Submodel in the Particle Domain</th>
<th>Solution Method</th>
</tr>
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<tbody>
<tr>
<td>• 1D spherical particle model</td>
<td>• SVM (state variable method)</td>
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</tbody>
</table>

#### Submodel in the Electrode Domain

<table>
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<tr>
<th>Submodel in the Cell Domain</th>
</tr>
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<tbody>
<tr>
<td>• 3D Single Potential-Pair Continuum Model (<strong>SPPC</strong>)</td>
</tr>
</tbody>
</table>

- **SVM** (Support Vector Machine)
- **SPPC** (Single Potential-Pair Continuum Model)
- **SVM** (state variable method)
- **FV-LSM** (Finite Volume – Linear Superposition Methods)
Electric Current Transport

4C discharge / Single-side cooling
Electric Current Transport

4C discharge / Single-side cooling

cooled top

bottom
Non-Uniform Utilization

Wound Cells

- A pair of wide current collectors
- Two electrode pairs
- Cylindrical or prismatic cells

**Stacking :** Forming the first pair between inner electrodes

**Winding :** Forming the second pair between outer electrodes
WPPC (Wound Potential-Pair Continuum)

Concentric semi-circular winding

Anode electrode
Positive current collector
Separator
Cathode electrode
Unit stratum/finite cell volume

Applicable to flat wound prismatic cells

Cell with discrete tabs
# MSMD Application to Prediction of Wound Cylindrical Cell Behavior

<table>
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<tr>
<td><strong>Submodel in the Electrode Domain</strong></td>
<td></td>
</tr>
<tr>
<td>🔄 1D porous electrode model</td>
<td>• SVM</td>
</tr>
<tr>
<td><strong>Submodel in the Cell Domain</strong></td>
<td></td>
</tr>
<tr>
<td>🔄 3D Wound Potential-Pair Continuum Model (WPPC)</td>
<td>• FV-LSM finite volume – linear superposition methods</td>
</tr>
</tbody>
</table>

### Diagrams
- Particle Domain
- Electrode Domain
- Cell Domain

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Kinetics Response

Impact of electrical current transport design

Continuous-tabs (CT) cell

20-tabs (20T) cell

10-tabs (10T) cell

5-tabs (5T) cell

2-tabs (2T) cell

Kyu-Jin Lee, et al., April 2011
Non-uniform Kinetics during 4C Discharge

Electrode plate current density [A/m²] at inner-electrode pair

Time is 5 sec

2-tab 20Ah cell

10-tab 20Ah cell
Thermal Response

Impact of electrical current transport design

Continuous-tabs (CT) cell

20-tabs (20T) cell

10-tabs (10T) cell

5-tabs (5T) cell

2-tabs (2T) cell

Temperature imbalance at 4C discharge

Kyu-Jin Lee, et al., April 2011
Thermal Evolution during 4C Discharge

2-Tab Design

Time is 5sec

Y(m)
0.1
0.05
0

X(m)
0.5
1
1.5
2
2.5
3

50
40
30

core
surface

[Graph showing temperature distribution over time and space with a 2-tab design diagram]
The Road Ahead

Extending scales, higher fidelity, fully integrated system

First Principles

Porous Electrode Model

Meso-Scale Electrode Model

MSMD

CFD

Vehicle Simulation

10^{-10}  10^{-8}  10^{-6}  10^{-4}  10^{-2}  10^{0} [m]
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