A Three-Dimensional Thermal-Electrochemical Coupled Model for Spirally Wound Large-Format Lithium-Ion Batteries

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Kyu-Jin Lee*, Kandler Smith, Gi-Heon Kim
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Objectives

- Develop thermal and electrochemical models resolving 3-dimensional spirally wound structures of cylindrical cells

- Understand the mechanisms and interactions between local electrochemical reactions and macroscopic heat and electron transfer

- Develop a tool and methodology to investigate macroscopic designs of cylindrical Li-ion battery cells
Multi-Scale Physics in Li-Ion Battery Systems

Physics of Li-Ion Battery Systems in Different 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

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

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

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Thermodynamic properties
Lattice stability
Material-level kinetic barrier
Transport properties

10^{-10} 10^{-8} 10^{-6} 10^{-4} 10^{-2} 10^{0} [m]
Porous Electrode Model of Li-ion Battery

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

- Difficult to resolve heat and electron current transport in large cell systems
Computational Cost of Modeling Large Li-ion Cell

- Characteristic length of electrodes: $L_{elec}$
- Grids for the porous electrode model: $N_{elec}$

Number of grids for a full 3-D electrode porous model:

$$N_{cell} \sim (L_{cell} / L_{elec} \times N_{elec})^3$$

Number of grids in a model resolving mesoscale geometry: $\sim 10^{2-3}$

A full 3-D mesoscale cell model is extremely expensive.
### Multi-Scale Multi-Dimensional (MSMD) Model

**Description**

- Introduces separate computational domains for corresponding length scale physics
- Decouples geometry between the domains
- Has independent coordinate systems for each domain
- Uses two-way coupling of solution variables using multi-scale model schemes

**Advantage**

- Selectively resolves higher spatial resolution for smaller characteristic length scale physics
- Achieves high computational efficiency
- Provides flexible & expandable modularized framework
Large Cell Design Differences

**Prismatic cells**
- Stacking / folding / semi-winding
- Complex and slow production processes
- Better packing efficiency for modules
- Better heat transfer

**Cylindrical cells**
- Winding
- Simple and fast production processes
- Low manufacturing cost

Photo Credit: NREL-Dirk Long

Large Cell Design can Lead to Large Temperature Difference

- Anisotropic thermal conductivity of electrodes coated on current collectors

  \[ K_{\text{in-plane}} = 10-100 \text{W/mK} \]

  \[ K_{\text{through-plane}} \approx 1 \text{W/mK} \]

- Stacked electrodes
- Thin and wide shape helps thermal uniformity

- Wound electrodes
- Center region of cell heats up easily due to the poor radial thermal conductivity
Large Cell Design can lead to Large Electric Potential Difference

**Prismatic cell**
- Large number of small metal current collectors
- Electric current flows through small distance

**Cylindrical cell**
- A pair of long continuous metal current collectors
- Electric current flows through long distance.
- Tab design can critically impact on cell performance

*Example:*
- Cell volume: 0.21 mL
- Prismatic cell: 200 mm x 150 mm x 7 mm
- Cylindrical cell: radius: 25.85 mm  height: 100 mm
- Thickness of an electrode pair: 300 µm  Length of current collectors: ~ 7 m
### 2-D Cylindrical Cell Model

#### Sub-model choice for 2-D cylindrical cell model

<table>
<thead>
<tr>
<th>Particle domain sub-model</th>
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<th>Cell domain sub-model</th>
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<tr>
<td>1-D spherical particle representation model</td>
<td>1-D porous electrode model</td>
<td>2-D axisymmetric cell model</td>
</tr>
</tbody>
</table>

#### Applicable to continuous tab design

- Cylindrical cell
- Unwinding jellyrolls
- Continuous tab design
- Extended foil
- Axisymmetric assumption
Continuous Tab Cell Design Evaluation

Effects of “Aspect Ratio” of a Cylindrical Cell

10s pulse power capability comparison

- Large H design has almost 10% less power capability.

Large H  D[mm]: 14 H[mm]: 350

Nominal  D[mm]: 50  H[mm]: 107

9 min 5C discharge

Large D  D[mm]: 115 H[mm]: 20

Nominal  D[mm]: 50  H[mm]: 107

• Large H design has almost 10% less power capability.
Continuous Tab Cell Design Evaluation

Effects of “Aspect Ratio” of a Cylindrical Cell

- Previous study

10s pulse power capability comparison

HPPC, BSF = 78

- Large H design has almost 10% less power capability.

Large H  D[mm]: 14 H[mm]: 350

Large D  D[mm]: 115 H[mm]: 20

Nominal  D[mm]: 50 H[mm]: 107

9 min 5C discharge
Present Study: **Electrical Design Issue-Tab Configuration**

Current flows along the winding direction

- 2-D axisymmetric model is not applicable to a wound cell.
- Geometries and materials of electric current paths in spirally wound layer structure must be properly resolved.

**Sub-model choice for 3-D cylindrical cell model**

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<td>3-D spiral wound cell model</td>
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</table>
**Cell Domain Model:** Spirally Wound Cell Model

**Unit structure:** Double-paired electrodes on single-paired current collectors

- **Inner electrode pair**
- **Outer electrode pair**

- **Double-sided anode electrode**
- **Negative current collector**
- **Separator**
- **Positive current collector**
- **Double-sided cathode electrode**

**Winding:** Alternating radial placement of double-paired electrodes

- Two electrode pairs are formed when the unit structure is wound.
- Two points with a distance of a winding cycle of outer electrode pair are matched in the wound structure.
Spiral Cell Structures: *Alternatively* layered jelly roll

A current collector has two electrode pairs in both sides
Non-uniform electrical potential along current collectors
Non-uniform charge transfer reaction across electrodes

Non-uniform potential along the current collectors occurs from electric current in the winding direction
Modeling Case

- Diameter 40 mm, inner diameter 8 mm, height 100 mm form factor
- Positive tabs on the top side, negative tabs on the bottom side
- 10-Ah capacity

**Tab locations for 5-tab case**

**5C constant current discharge**

SOC\textsubscript{ini} = 90%

Natural convection:

\[ h_{\text{inf}} = 5 \text{ W/m}^2\text{K} \]

\[ T_{\text{amb}} = 25^\circ\text{C} \]

\[ T_{\text{ini}} = 25^\circ\text{C} \]

Nickel oxide-based cathode

Graphite-based anode
Modeling Results

- 5 tabs in each current collector
- 5C discharge for 5 min

**Electric potential**

- Positive current collector
- Negative current collector

**Electrochemical reaction rate**

- Inner electrode pair
- Outer electrode pair

Current mainly flows in the winding direction

High generation rate of transfer current near tabs
Modeling Results

State of Charge

- Inner electrode pair
- Outer electrode pair

Temperature

- Radial heat transfer from tabs
- Temperature difference is relatively small

More usage of electrode near tabs
Modeling Results: \textit{Parametric Study}

- Different tab numbers (2, 5, 10 and continuous tab) on cell performance
- 10-Ah capacity, 5C discharge

![Graph showing output voltage vs. SOC with different tab numbers](image)

$SOC_{ini} = 90\%$
Modeling Results: *Parametric Study*

**Temperature**

- Natural convection: $h_{inf} = 5 \text{ W/m}^2\text{K}$
- $T_{amb} = 25^\circ\text{C}$
- $T_{ini} = 25^\circ\text{C}$

- 2 tabs
- 5 tabs
- 10 tabs
- Continuous tab
- 2-D model
Modeling Results: Parametric Study

**Generated Heat**

- 2 tabs
- 5 tabs
- 10 tabs
- Continuous tab
- 2-D mode

**Natural convection:**
- $h_{inf} = 5 \text{ W/m}^2\text{K}$
- $T_{amb} = 25^{\circ}\text{C}$
- $T_{ini} = 25^{\circ}\text{C}$
Innovation for Our Energy Future

Modeling Results: *Parametric Study*

- High rate of discharge with a moderate heat transfer condition
- Heat generation dominates temperature distribution in the system
### Modeling Results: Parametric Study

**Electrochemical reaction rate comparison**

*in the inner electrode pair at 5 min*

<table>
<thead>
<tr>
<th>Tab Count</th>
<th>Δi''/i''_{avga}</th>
<th>Color Bar</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 tabs</td>
<td>32.2%</td>
<td>0.2%</td>
</tr>
<tr>
<td>5 tabs</td>
<td>6.6%</td>
<td>0.2%</td>
</tr>
<tr>
<td>10 tabs</td>
<td>2.2%</td>
<td>0.2%</td>
</tr>
<tr>
<td>Continuous tab</td>
<td>0.2%</td>
<td>0.2%</td>
</tr>
</tbody>
</table>

**Modeling Results:**

- **Δi''/i''_{avga}**
- **2 tabs**
- **5 tabs**
- **10 tabs**
- **Continuous tab**
Modeling Results: *Parametric Study*

**Temperature deviation comparison**

*at 5 min*

<table>
<thead>
<tr>
<th>Temperature</th>
<th>ΔT</th>
<th>ΔT at 5 min</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.25°C</td>
<td>0.19°C</td>
<td>0.37°C</td>
</tr>
<tr>
<td>0.78°C</td>
<td>0.78°C</td>
<td>3.25°C</td>
</tr>
<tr>
<td>0.37°C</td>
<td>0.37°C</td>
<td>0.78°C</td>
</tr>
<tr>
<td>0.19°C</td>
<td>0.19°C</td>
<td>0.37°C</td>
</tr>
</tbody>
</table>

**Continuous tab**
Conclusions

• Used Multi-Scale Multi-Dimensional model to evaluate large-format cell designs by integrating micro-scale electrochemical processes and macro-scale heat and electrical current transport.

• **Spatial non-uniformity** of battery physics, which becomes significant in large batteries, requires 3 dimensional model.

• **Developed macro-scale domain model** resolved **spirally wound structures** of lithium-ion batteries.

• **Modeled effects of tab configurations** and **the double-sided electrode structure**.

• Increasing the number of tabs in spiral-wound cells would be preferable to manage internal heat and electron current transport, and to achieve uniform electrochemical kinetics.

• The spiral-wound cell model provides **quantitative information** regarding optimization of cell design including tab location and number.
US. Department of Energy, Vehicle Technology Program
Dave Howell, Hybrid Electric Systems Team Leader
Brian Cunningham, CAEBAT Coordinator

National Renewable Energy Laboratory
Ahmad Pesaran, Energy Storage Team Leader