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

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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

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

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

System operating conditions
- Environmental conditions
- Control strategy

[Diagram showing scales and physics processes]
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

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**Diagram:**
- **Voltage [V]**
- **Capacity [Ah]**
- **Time**

**Table:**
- Capacity [Ah/m²]
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:

$$N_{cell} \sim \left( \frac{L_{cell}}{L_{elec} \times N_{elec}} \right)^3$$

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
  
  \[ K_{\text{in-plane}} \approx 10-100 \text{W/mK} \]
  
  \[ K_{\text{through-plane}} \approx 1 \text{W/mK} \]

  - Negative current collector
  - Anode electrode
  - Separator
  - Cathode electrode
  - Positive current collector

**Prismatic cell**

- Stacked electrodes
- Thin and wide shape helps thermal uniformity

**Cylindrical cell**

- 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

- Particle domain sub-model: 1-D spherical particle representation model
- Electrode domain sub-model: 1-D porous electrode model
- Cell domain sub-model: 2-D axisymmetric cell model

Applicable to continuous tab design

Cylindrical cell → Unwinding jellyrolls → Continuous tab design → Axisymmetric assumption

Extended foil
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

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

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

9 min 5C discharge
Continuous Tab Cell Design Evaluation

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**Present Study:** *Electrical Design Issue-Tab Configuration*

- 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**

- **Particle domain sub-model:** 1-D spherical particle representation model
- **Electrode domain sub-model:** 1-D porous electrode model
- **Cell domain sub-model:** 3-D spiral wound cell model
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*

**Negative current collector**
**Cathode electrode**
**Separator**
**Anode electrode**
**Positive current collector**

A current collector has two electrode pairs in both sides.
Spiral Cell Structures: \textit{Electrical potential fields and charge transfer reaction}

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**

- **Positive current collector**
- **Negative current collector**

**Tab configuration of each electrode pair**

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

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5C constant current discharge

SOC\(_{ini}\) = 90%

Natural convection:

- \(h_{inf} = 5 \text{ W/m}^2\text{K}\)
- \(T_{amb} = 25^\circ\text{C}\)
- \(T_{ini} = 25^\circ\text{C}\)

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

Top view

Bottom view

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

Innovation for Our Energy Future
Modeling Results: Parametric Study

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

![Output voltage graph with different tab numbers showing varying voltage levels.](image)

- $\text{SOC}_{\text{ini}} = 90\%$
Modeling Results: \textit{Parametric Study}

![Graph showing the relationship between discharge capacity and average temperature]

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

**Generated Heat**

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

**Natural convection:**

\[
h_{\text{inf}} = 5 \text{ W/m}^2\text{K} \\
T_{\text{amb}} = 25^\circ\text{C} \\
T_{\text{ini}} = 25^\circ\text{C}
\]
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

- 2 tabs: 32.2%
- 5 tabs: 6.6%
- 10 tabs: 2.2%
- Continuous tab: 0.2%

Δi''/i''avg ≈ i'' [A/m²]
Modeling Results: *Parametric Study*

**Temperature deviation comparison**

*at 5 min*

- **3.25°C**
- **0.78°C**
- **0.37°C**
- **0.19°C**

**ΔT**

- **2 tabs**
- **5 tabs**
- **10 tabs**
- **Continuous tab**

**T-T_{avg} [°C]**

- **0.19°C**
- **0.37°C**
- **0.78°C**
- **3.25°C**
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.
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