3D Thermal and Electrochemical Model for Spirally Wound Large Format Lithium-ion Batteries

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Objectives of this Study

Behaviors of spirally wound large format Li-Ion batteries are affected by macroscopic designs of cells.

• To develop thermal and electrochemical models resolving 3 dimensional spirally wound structures of cylindrical cells.

• To understand the mechanisms and interactions between local electrochemical reactions and macroscopic heat and electron transfers.

• To develop a tool and methodology to support macroscopic designs of cylindrical Li-Ion battery cells.
Multi-Scale Physics in Li-Ion Battery

Requirements & Resolutions

“Requirements” are usually defined in a macro-scale domain and terms

- Performance
- Life
- Cost
- Safety

- Wide range of length and time scale physics
- Design improvements required at different scales
- Need for better understanding of interaction among varied scale physics
Multi-Physics Interaction

Comparison of two 40 Ah flat cell designs

- Previous study

Larger over-potential promotes faster discharge reaction
Converging current causes higher potential drop along the collectors

Transfer current generation

High temperature promotes faster electrochemical reaction
Higher localized reaction causes more heat generation

Working potential

This cell is cycled more uniformly, can therefore use less active material ($) and has longer life.

Temperature

2 min 5C discharge
Porous Electrode Model

Charge Transfer Kinetics at Reaction Sites

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

\[ i_0 = k (c_e)^{\alpha_a} (c_{s,max} - c_{s,e})^{\alpha_a} (c_{s,e})^{\alpha_c} \]

\[ \eta = (\phi_s - \phi_e) - U \]

Species Conservation

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

\[ \frac{\partial(e_e c_e)}{\partial t} = \nabla \cdot \left( D_{ee}^{\text{eff}} \nabla c_e \right) + \frac{1 - t^2_+}{F} j^{Li} - \frac{i_e}{F} r^t_+ \]

Charge Conservation

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

\[ \nabla \cdot \left( \kappa^{\text{eff}} \nabla \phi_e \right) + \nabla \cdot \left( \kappa_D^{\text{eff}} \nabla \ln c_e \right) + j^{Li} = 0 \]

Energy Conservation

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

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

- Pioneered by Newman group (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 resolve heat and electron current transport in large cell systems
Porous Electrode Model

Charge Transfer Kinetics at Reaction Sites

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

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

Species Conservation

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

\[
\frac{\partial \left( \varepsilon_e c_e \right)}{\partial t} = \nabla \cdot \left( \varepsilon_{\text{eff}} \nabla c_e \right) + \frac{1 - t_e^o}{F} j^{Li} - \frac{i_e}{F} \nabla t_e^o
\]

Charge Conservation

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

\[
\nabla \cdot \left( \kappa_{\text{eff}} \nabla \phi_e \right) + \nabla \cdot \left( \kappa_D \nabla \ln c_e \right) + j^{Li} = 0
\]

Energy Conservation

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

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

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Multi-Scale Multi-Dimension (MSMD) Model

- Introducing separate computational domains for corresponding length scale physics
- Geometry decoupling between the domains
- Using independent coordinate systems for each domain
- Two-way coupling of solution variables using multi-scale model schemes

- Selectively resolve higher spatial resolution for smaller characteristic length scale physics
- Achieve high computational efficiency
- Provide flexible & expandable modularized framework
Large Cell Design Issues

**Prismatic cell**

- Stacking electrodes coated on metal current collectors
- Less efficient manufacturing processes for mass production

**Cylindrical cell**

- Rolling electrodes coated on metal current collectors
- Well established manufacturing process for small batteries
- More difficulties for large cell design (cf. tap design, heat management, etc)
Large Cell Design Issues

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Previous Development of Wound Cell Model

Applicable to continuous tab design

Sub-model choice for spirally wound continuous tab cells

- Previous study

Cylindrical cell Unwinding jellyrolls continuous tab design Axisymmetric assumption

Extended Foil

: particle domain sub-model
: electrode domain sub-model
: cell domain sub-model

1D spherical particle representation model
1D porous electrode model
2D axisymmetric cell model
Tab-less Wound Cell Design Evaluation

Effects of “Aspect Ratio” of a Cylindrical Cell

- Previous study

10s pulse power capability comparison

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

9 min 5C discharge

- Large D: D[mm]: 115 H[mm]: 20
- Nominal: D[mm]: 50 H[mm]: 107
Present Study

**Tab configuration: number & size**

- 2D axisymmetric model is not applicable to a wound cell with a finite number of current tabs where lateral electric current is not negligible in current collector foils.
- Geometries and materials of electric current paths in spirally wound layer structure should be properly resolved.

Cylindrical cell Unwinding jellyrolls

Current flows along the winding direction 3 dimensional spiral wound geometry
New Development of 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.
We cannot expect uniform potential along the current collectors due to inevitable electric current in winding direction.
Modeling case

- Diameter 40mm, inner diameter 8mm, 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  
$\text{soc}_{ini} = 90\%$

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

**Tab configuration of each electrode pairs**

Inner electrode pair

Outer electrode pair
Modeling results

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

Electric potential

- Current mainly flows in the winding direction

Electrochemical reaction rate

- High generation rate of transfer current near tabs

- Current mainly flows in the winding direction

Top view

Bottom view
Modeling results

- **State of charge**
  - **Inner electrode pair**
  - **Outer electrode pair**
  - More usage of electrode near tabs

- **Temperature**
  - High rate discharge with a moderate heat transfer condition
  - Heat generation dominates temperature distribution in the system.
  - Temperature difference in the system is relatively small yet.
**Parametric study**

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

### Modeling results

**Cells with fewer tabs ...**
- lower output voltage
- higher average temperature

**soc\textsubscript{ini} = 90\%**
**Natural convection:**
- $h_{\text{inf}} = 5 \text{ W/m}^2\text{K}$
- $T_{\text{amb}} = 25^\circ\text{C}$
- $T_{\text{ini}} = 25^\circ\text{C}$

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**V\textsubscript{out} [V]**

**T\textsubscript{avg} [^\circ\text{C}]**

- 2D model
  - continuous tab
  - 10 tabs
  - 5 tabs
  - 2 tabs

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**SOC**

**Time [min]**

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- 2 tabs
- 5 tabs
- 10 tabs
- continuous tab
Electrochemical reaction rate comparison

in the inner electrode pair at 5 min

\[ i'' \text{ [A/m}^2\text{]} \]

\[ \Delta i'' / i''_{avg} \]

32.2% 
2 tabs

6.6% 
5 tabs

2.2% 
10 tabs

0.2% 
Continuous tab

\[ X \text{ [m]} \]

\[ Y \text{ [m]} \]
Modeling results

Temperature deviation comparison

$\Delta T$ at 5 min

$T - T_{avg}$ [°C]

- 2 tabs
- 5 tabs
- 10 tabs
- Continuous tab
Conclusion

• **A Multi-Scale Multi-Dimension model** was used for evaluating large format automotive cell designs by integrating micro-scale electrochemical process and macro-scale heat and electrical current transports.

• Spatial non-uniformity of battery physics, which becomes significant in large batteries, cause unexpected performance in spiral wound lithium-ion batteries.

• **A macro-scale domain model based on spirally wound structures** of lithium-ion batteries was developed to understand effects of tab configurations and the double sides electrodes structure.

• Spiral wound cells with more tabs would be preferable to manage cell internal heat and electron current transport, and consequently to achieve uniform electrochemical kinetics over a system.

• The spiral wound cell model can provide quantitative data in terms of finding the optimum number of tabs to battery manufacturers.
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