NATIONAL RENEWABLE ENERGY LABORATORY

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

Space Power Workshop
Los Angeles, CA

April 18, 2011

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

NREL/PR-5400-51151

This research activity is funded by the US Department of Energy (Dave Howell)

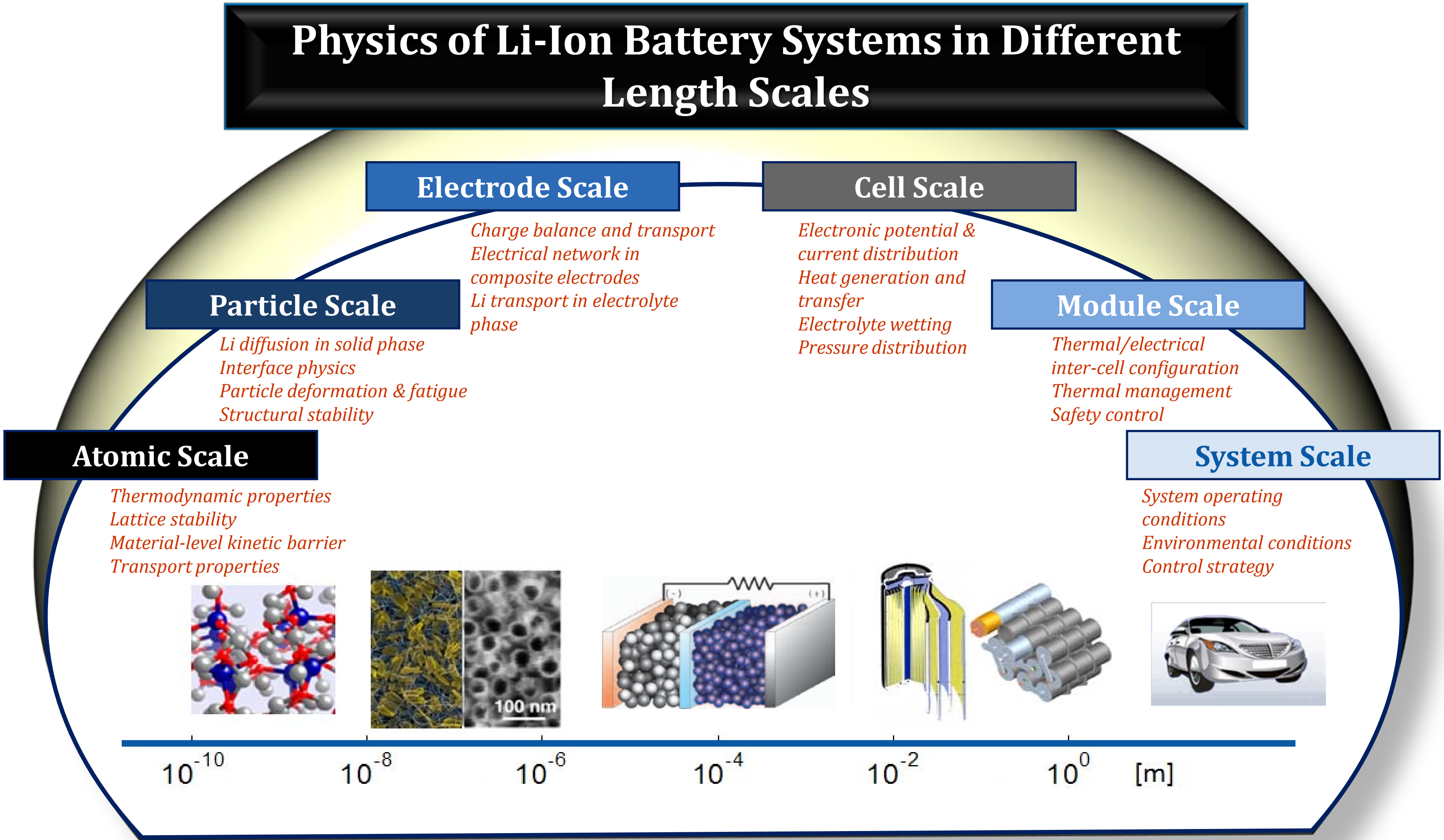
Objectives

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

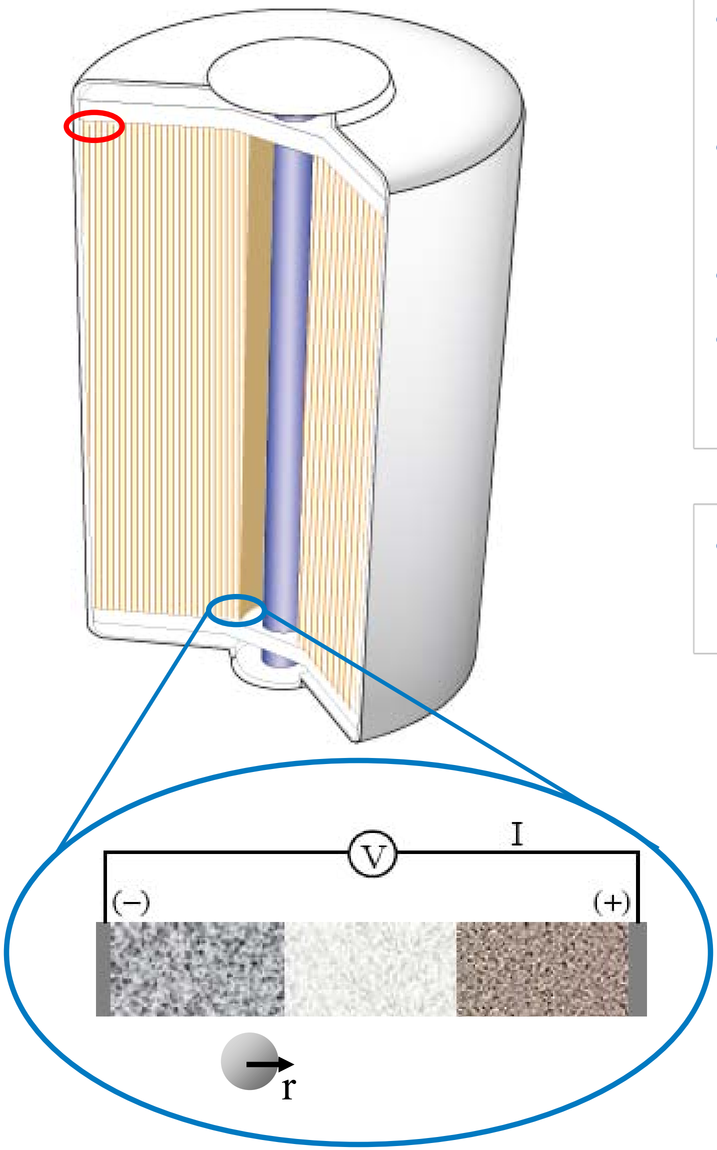
- **Understand the mechanisms and interactions between local electrochemical reactions and macroscopic heat and electron transfer**

- **Develop a tool and methodology to investigate macroscopic designs of cylindrical Li-ion battery cells**

Multi-Scale Physics in Li-Ion Battery Systems

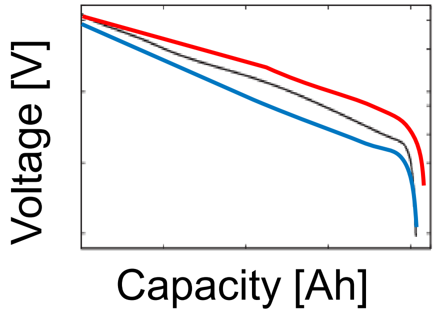


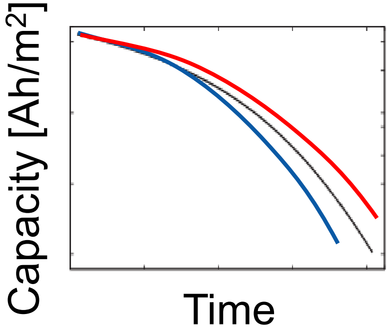
Porous Electrode Model of Li-ion Battery



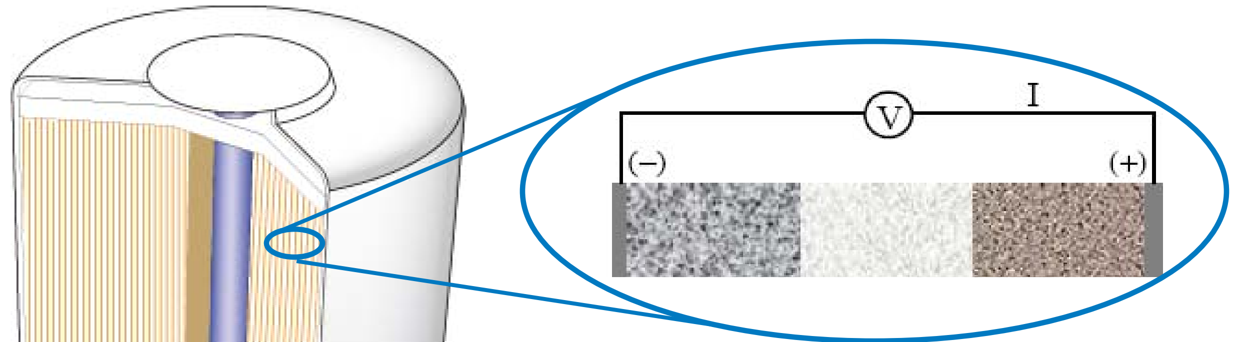
- Pioneered by Newman group (Doyle, Fuller, and Newman 1993)
- Captures lithium diffusion dynamics and charge transfer kinetics across electrodes
- Predicts current/voltage response of a battery
- Provides design guide for thermodynamics, kinetics, and transport across electrodes

- **Difficult to resolve heat and electron current transport in large cell systems**





Computational Cost of Modeling Large Li-ion Cell

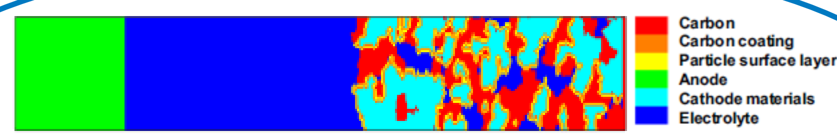


- Characteristic length of electrodes: L_{elec}
- Grids for the porous electrode model: N_{elec}

Characteristic length of a cell: L_{cell}

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

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

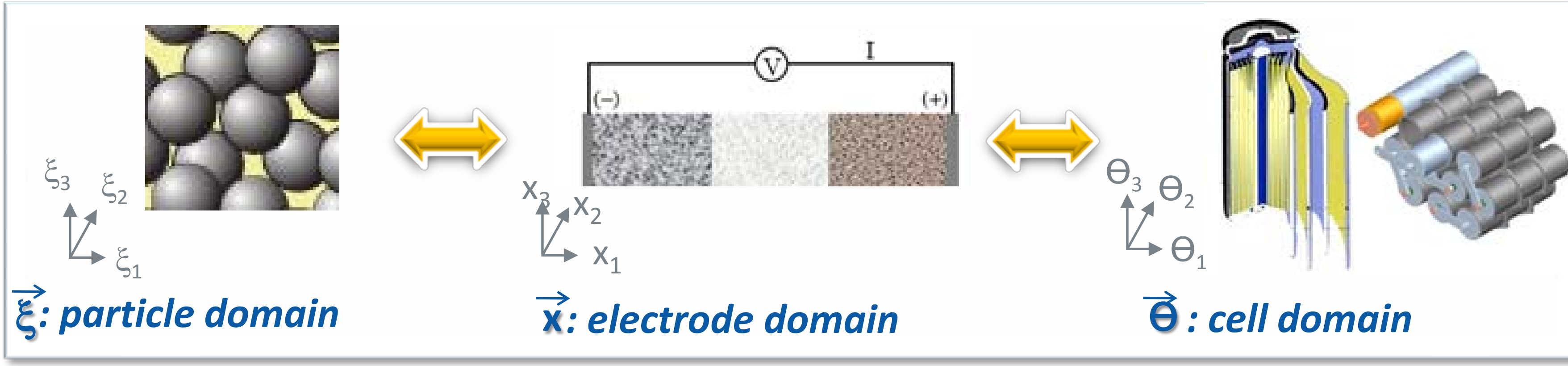


Liu and Siddique, 218th ECS , 2010 *Micro-structure Reconstruction*

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

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

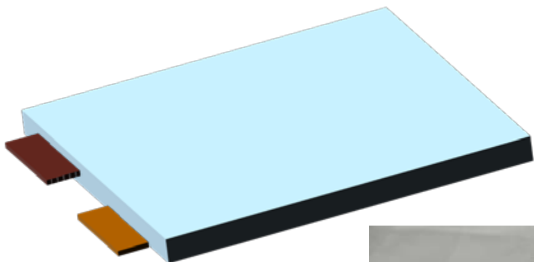


Photo Credit: NREL-Dirk Long

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

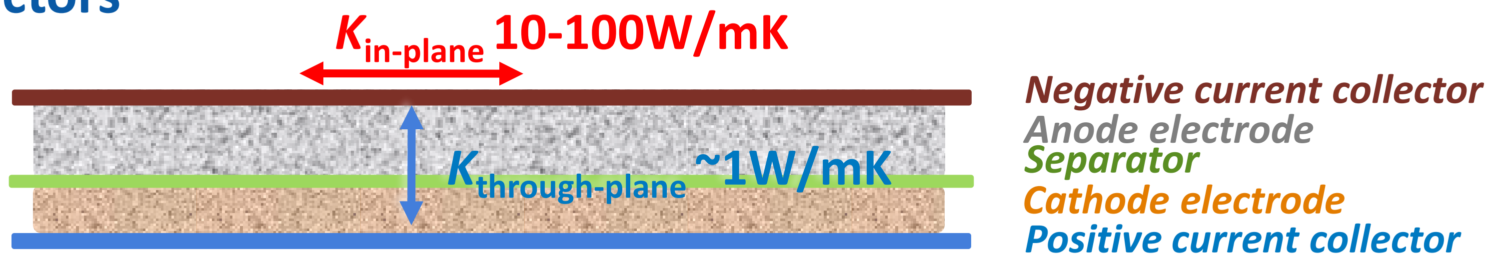
Cylindrical cells

Photo Credit: http://en.wikipedia.org/wiki/List_of_battery_sizes

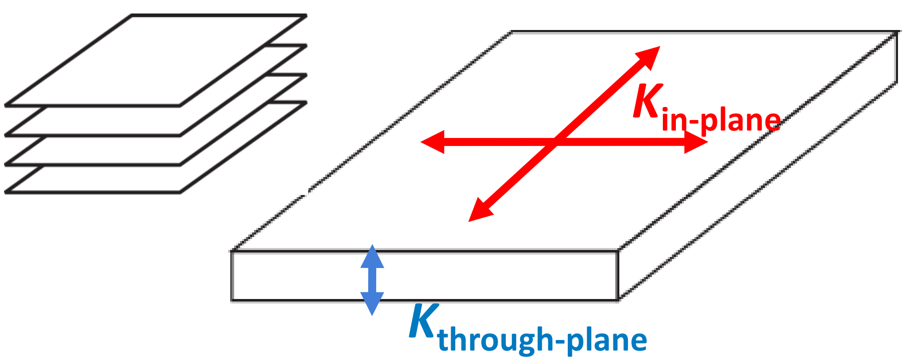
- *Winding*
- *Simple and fast production processes*
- *Low manufacturing cost*

Large Cell Design *can Lead to* Large Temperature Difference

- **Anisotropic thermal conductivity of electrodes coated on current collectors**

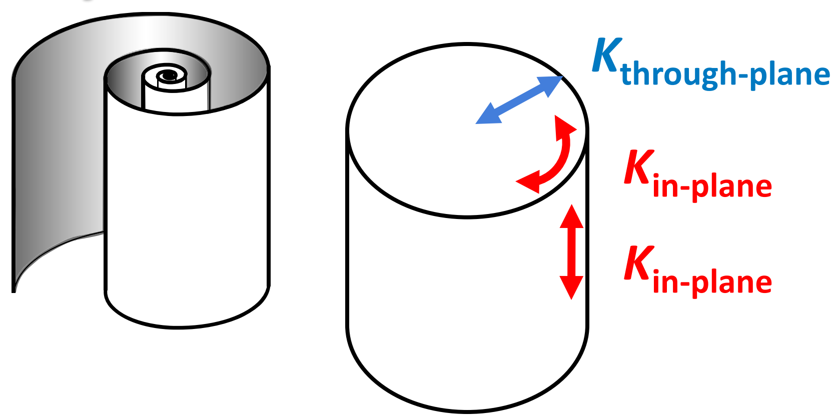


Prismatic cell



- Stacked electrodes
- Thin and wide shape helps thermal uniformity

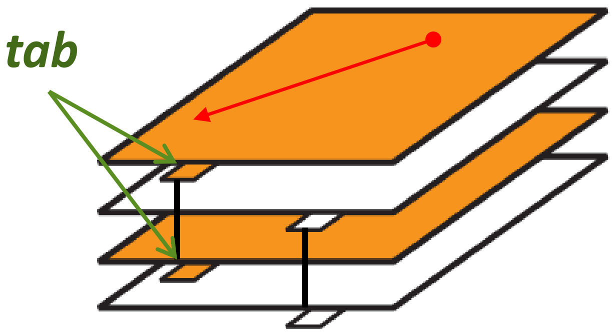
Cylindrical cell



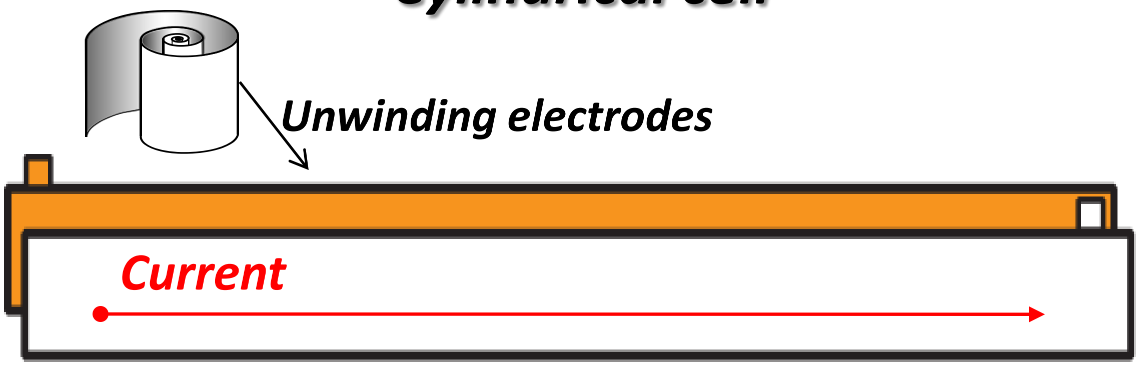
- Wound electrodes
- Center region of cell heats up easily due to the poor radial thermal conductivity

Large Cell Design *can Lead to* Large Electric Potential Difference

Prismatic cell



Cylindrical cell



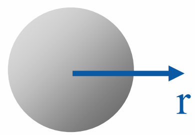
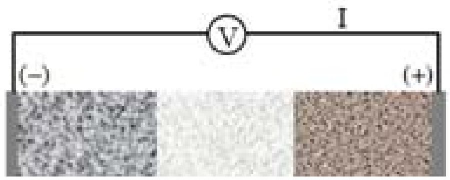
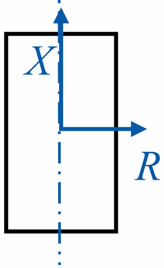
- Large number of small metal current collectors
- Electric current flows through small distance

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

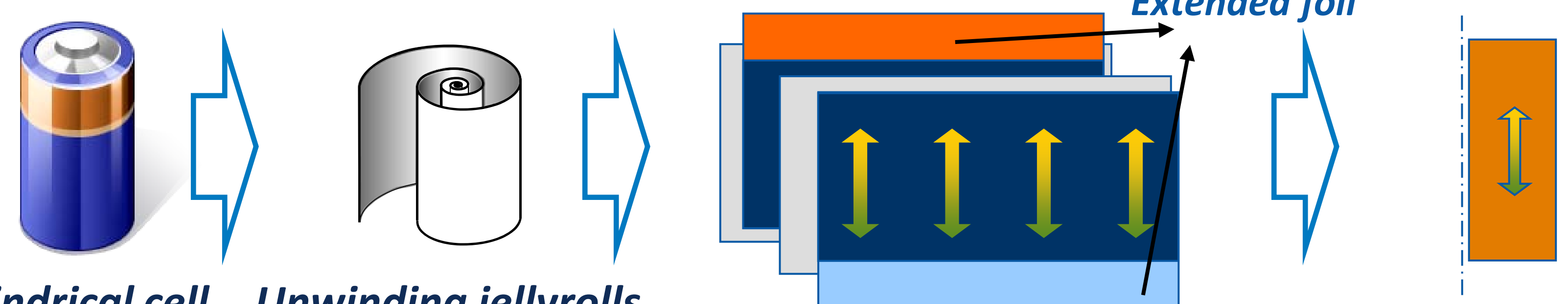
Example: *Cell volume: 0.21 mL*
Prismatic cell: 200 mm x 150 mm x 7 mm
Cylindrical cell: radius: 25.85 mm height: 100 mm
Thickness of an electrode pair: 300 µm ➡ *Length of current collectors: ~ 7 m*

2-D Cylindrical Cell Model – Previous study

<u>*Sub-model choice for 2-D cylindrical cell model*</u>

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

Applicable to continuous tab design

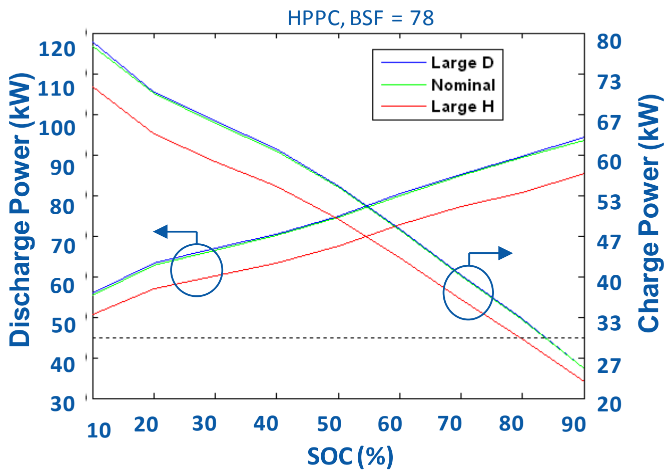


Cylindrical cell *Unwinding jellyrolls* *Continuous tab design* *Axisymmetric assumption*

Continuous Tab Cell Design Evaluation

Effects of "Aspect Ratio" of a Cylindrical Cell – Previous study

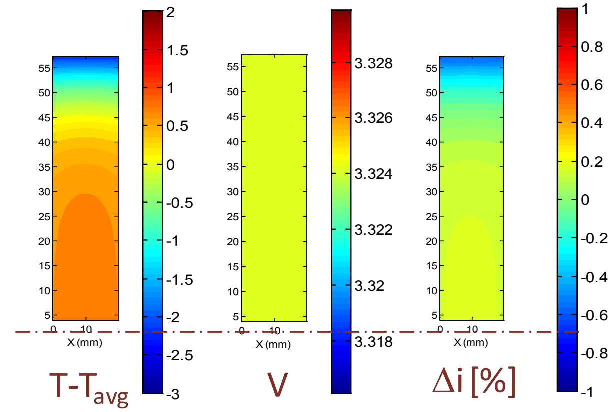
10s pulse power capability comparison



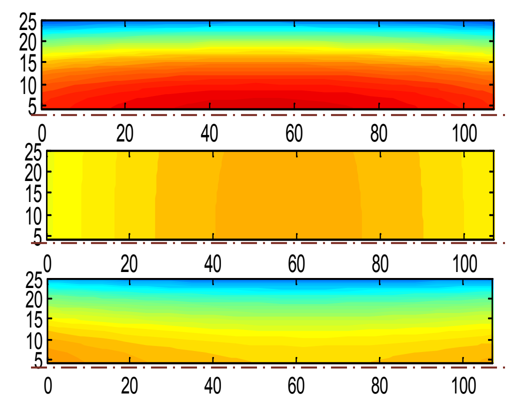
- Large H design has almost 10% less power capability.

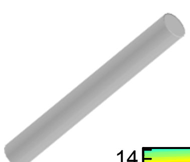
9 min 5C discharge

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

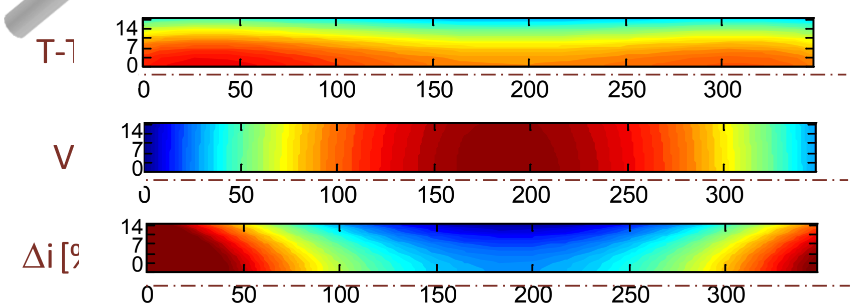


Nominal D[mm]: 50 H[mm]: 107





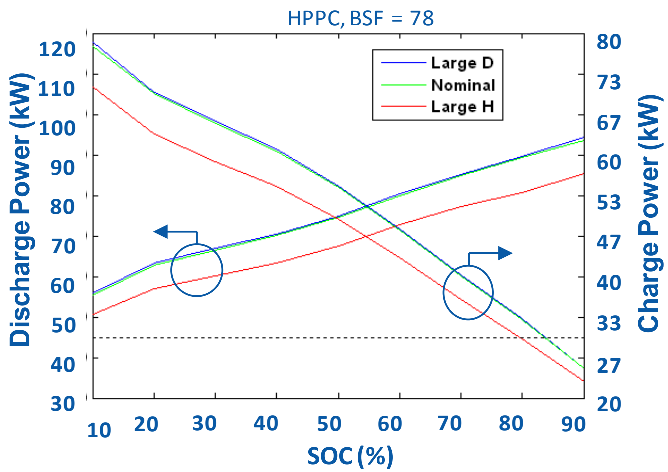
Large H D[mm]: 14 H[mm]: 350



Continuous Tab Cell Design Evaluation

Effects of "Aspect Ratio" of a Cylindrical Cell – *Previous study*

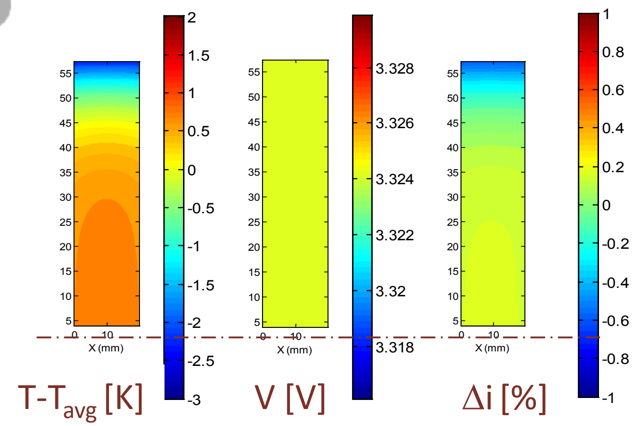
10s pulse power capability comparison



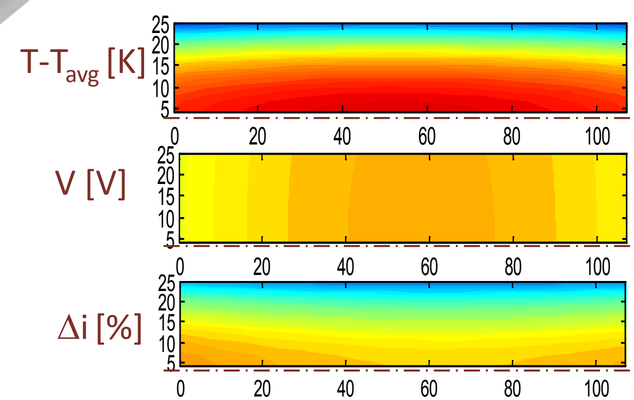
- Large H design has almost 10% less power capability.

9 min 5C discharge

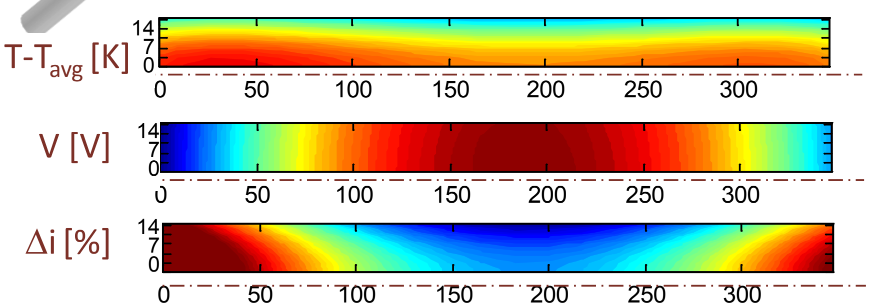
Large D D[mm]: 115 H[mm]: 20



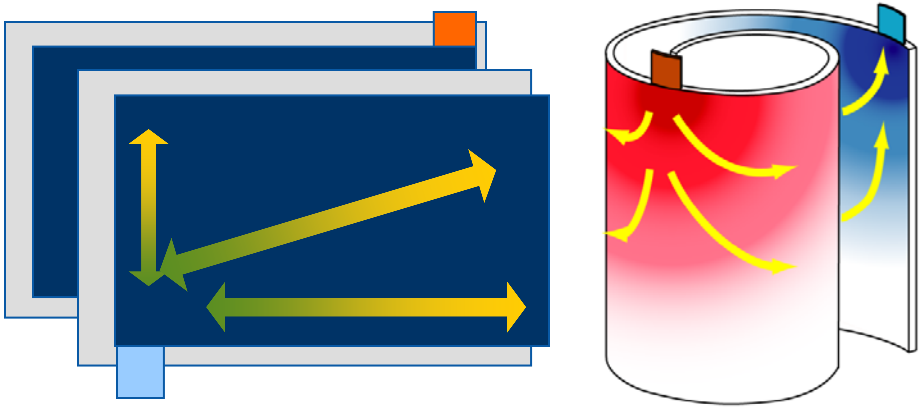
Nominal D[mm]: 50 H[mm]: 107



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



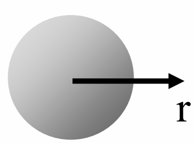
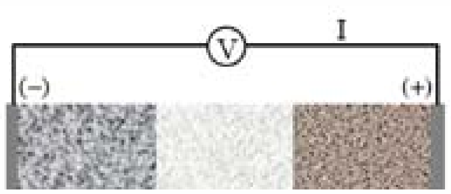
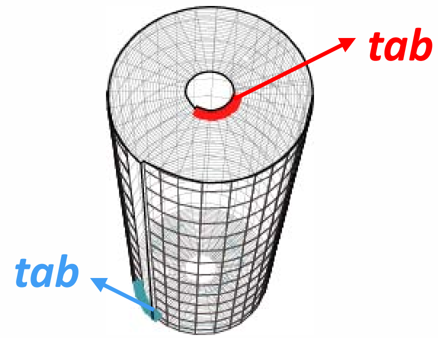
Present Study: *Electrical Design Issue-Tab Configuration*



Current flows along the winding direction

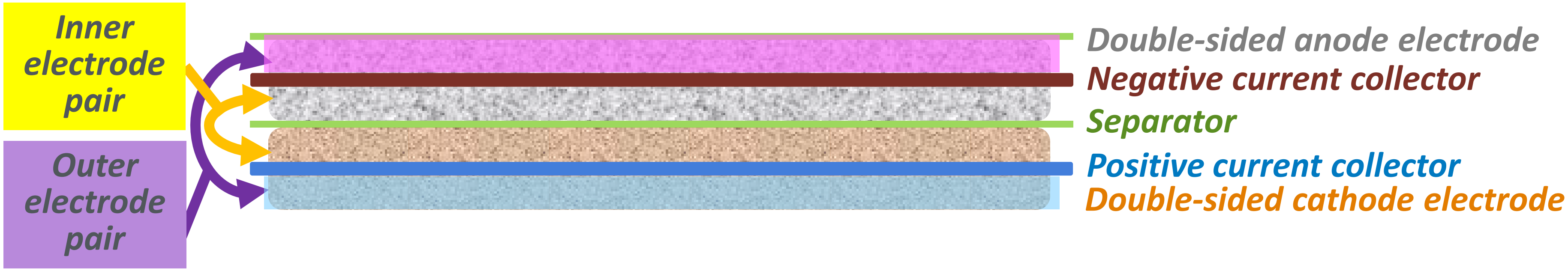
- 2-D axisymmetric model is not applicable to a wound cell .
- Geometries and materials of electric current paths in spirally wound layer structure must be properly resolved.

Sub-model choice for 3-D cylindrical cell model

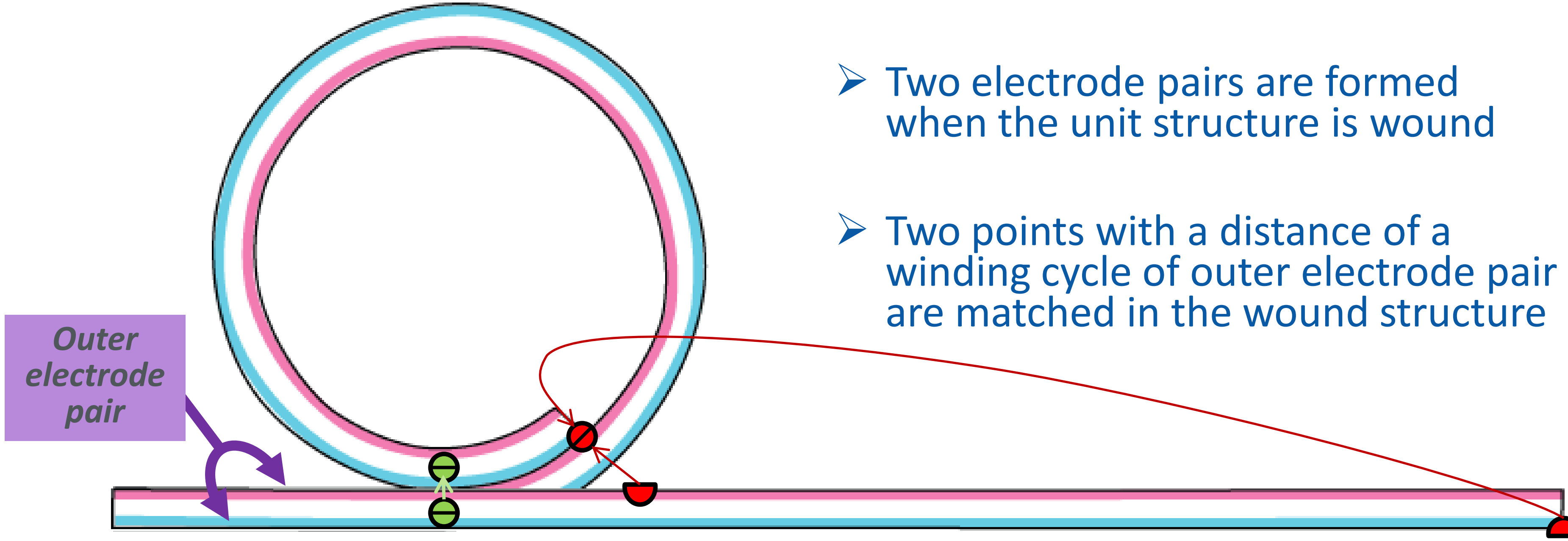
Particle domain sub-model	*Electrode domain sub-model*	*Cell domain sub-model*
		
1-D spherical particle representation model	**1-D porous electrode model**	**3-D spiral wound cell model**

Cell Domain Model: *Spirally Wound Cell Model*

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

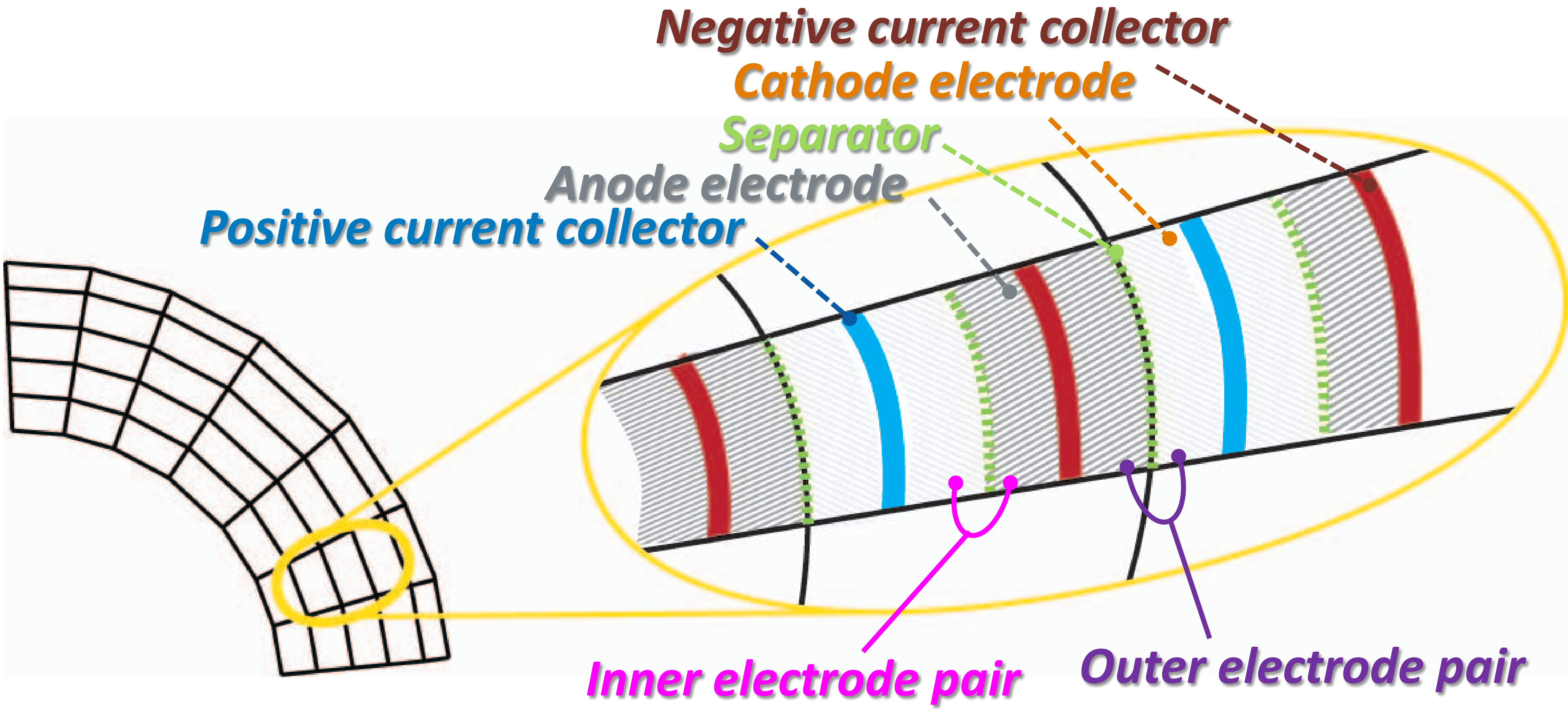


Winding: Alternating radial placement of double-paired electrodes



- 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

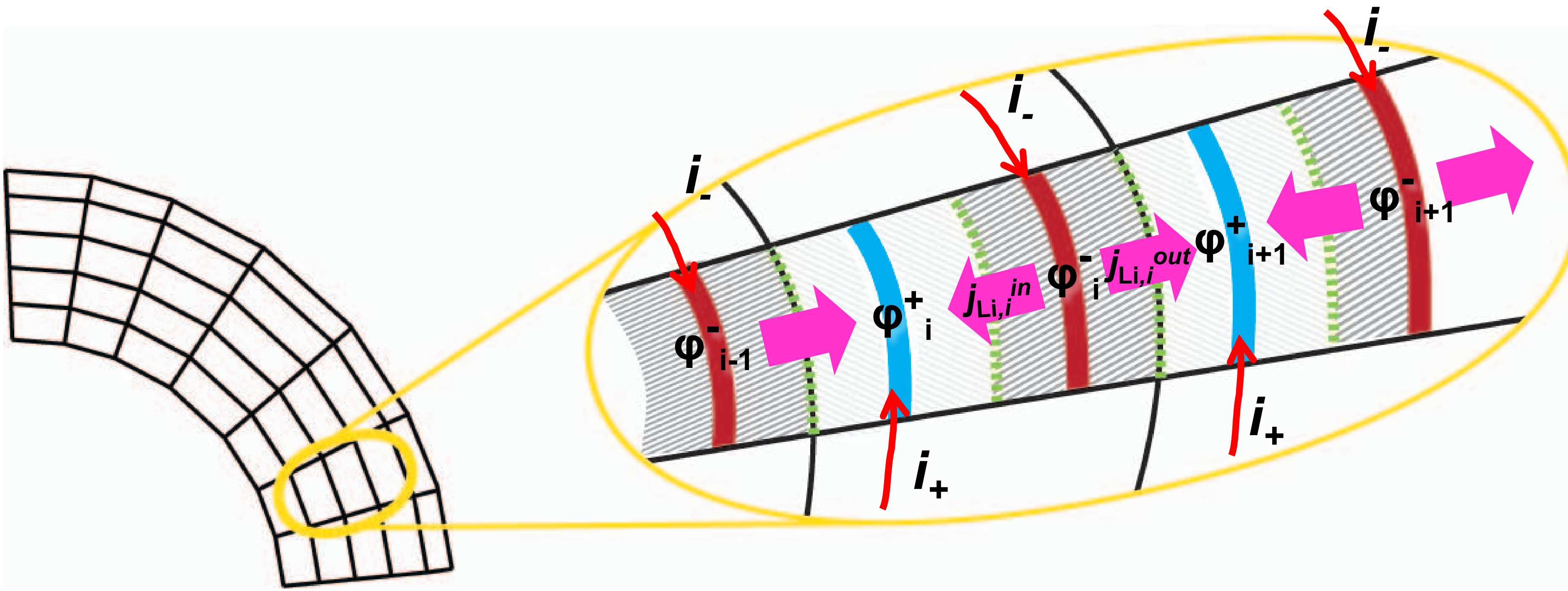
Spiral Cell Structures: *Alternatively layered jelly roll*



A current collector has two electrode pairs in both sides

Spiral Cell Structures: *Electrical potential fields and charge transfer reaction*

Non-uniform electrical potential along current collectors
Non-uniform charge transfer reaction across electrodes

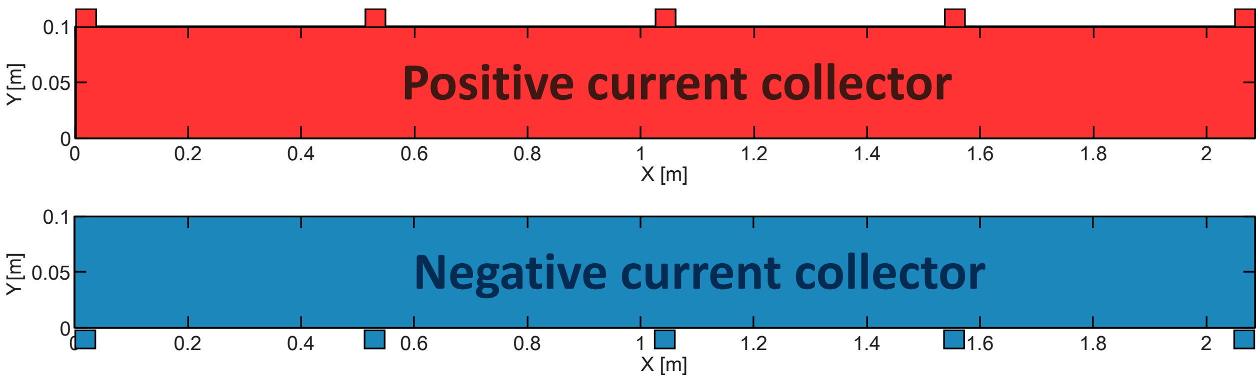


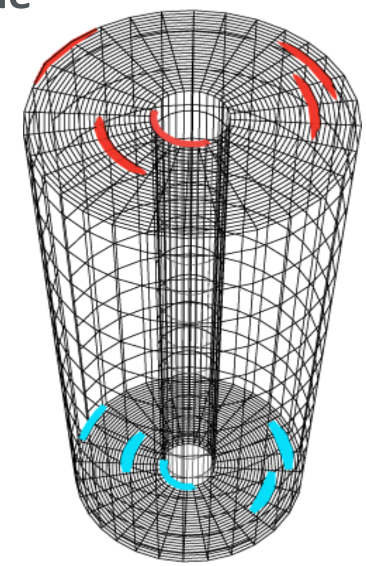
Non-uniform potential along the current collectors occurs from electric current in the winding direction

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

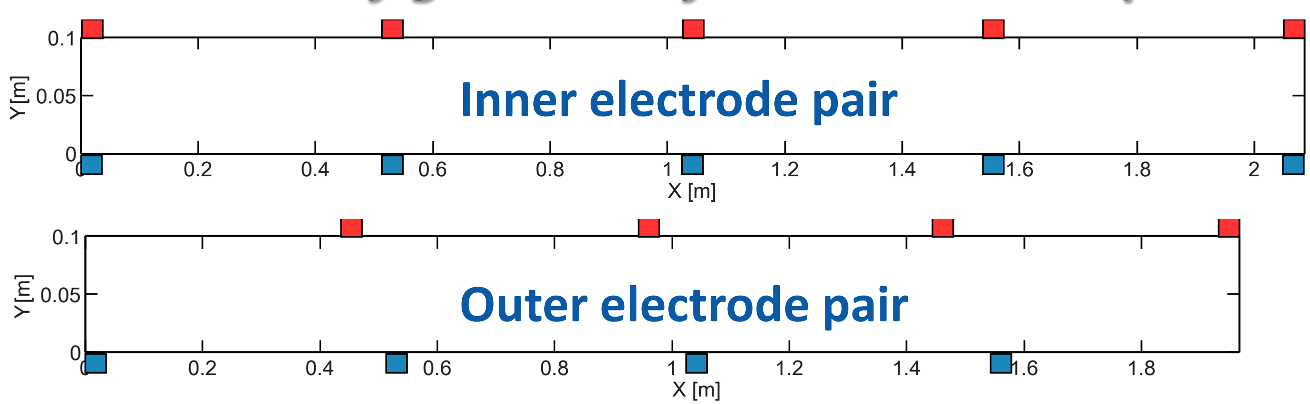




5C constant current discharge
SOC_{ini} = 90%
Natural convection:
h_{inf} = 5 W/m²K
T_{amb} = 25°C
T_{ini} = 25°C

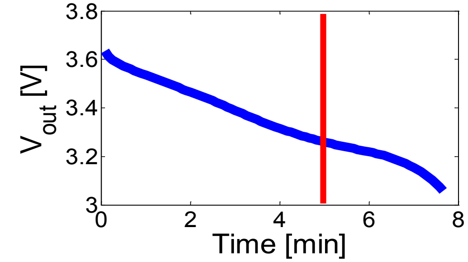
Nickel oxide-based cathode
Graphite-based anode

Tab configuration of each electrode pair



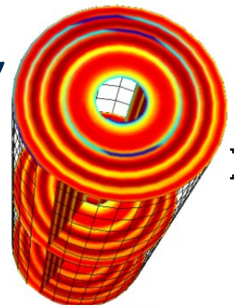
Modeling Results

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

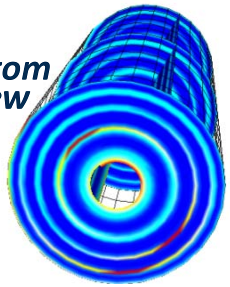


Electric potential

Top view

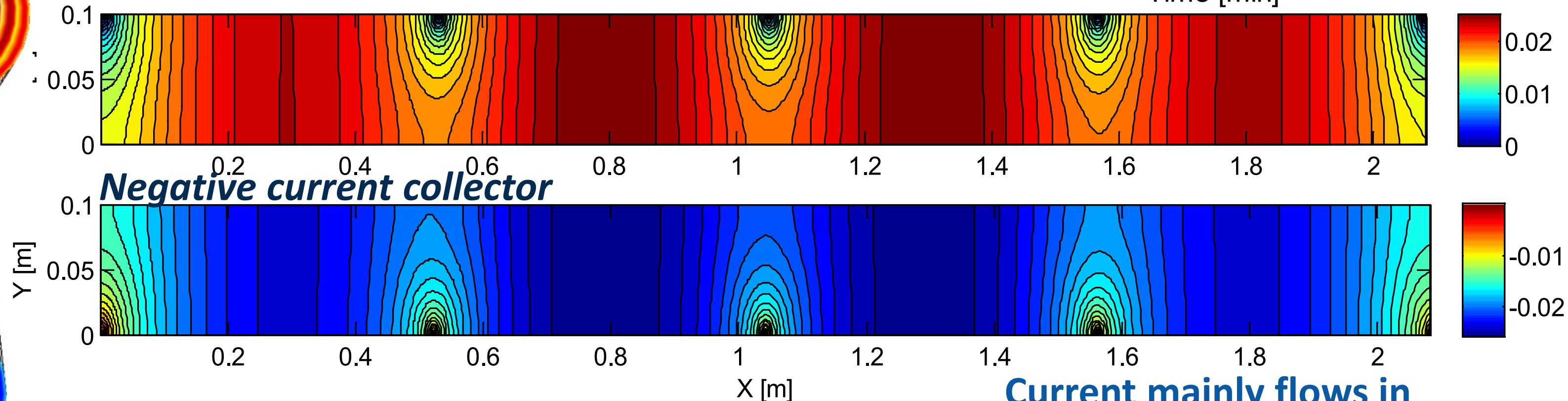


Bottom view



Positive current collector

Negative current collector

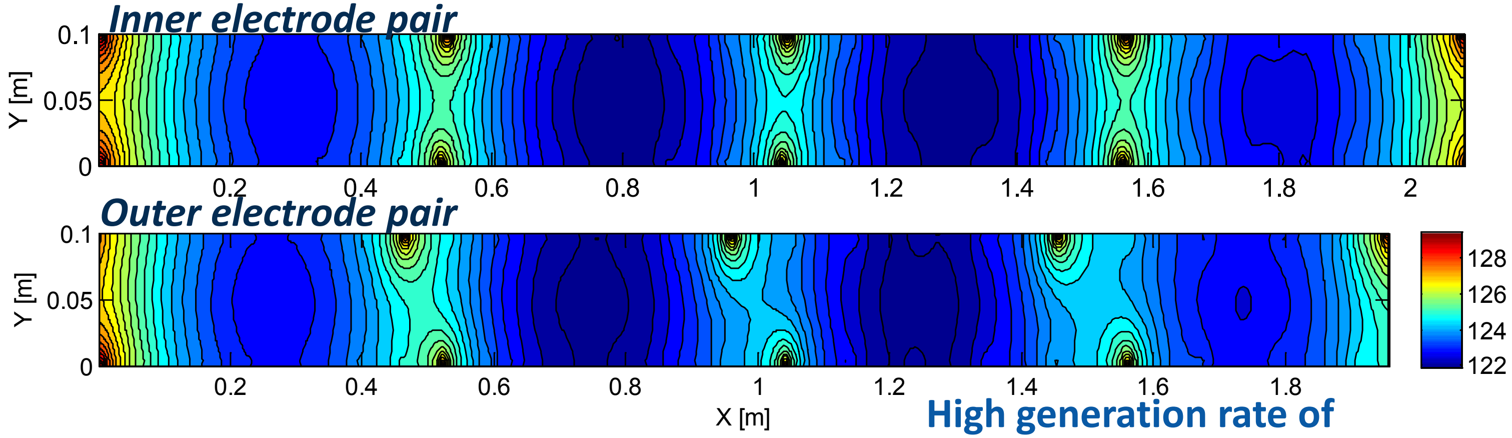


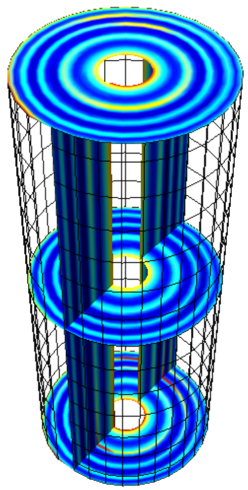
Current mainly flows in the winding direction

Electrochemical reaction rate

Inner electrode pair

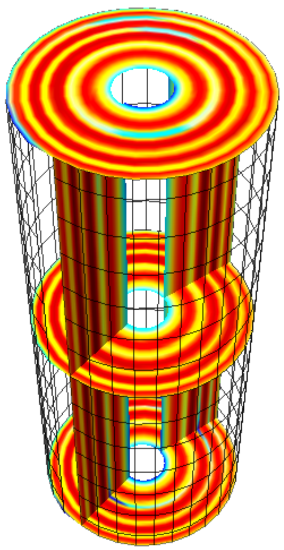
Outer electrode pair





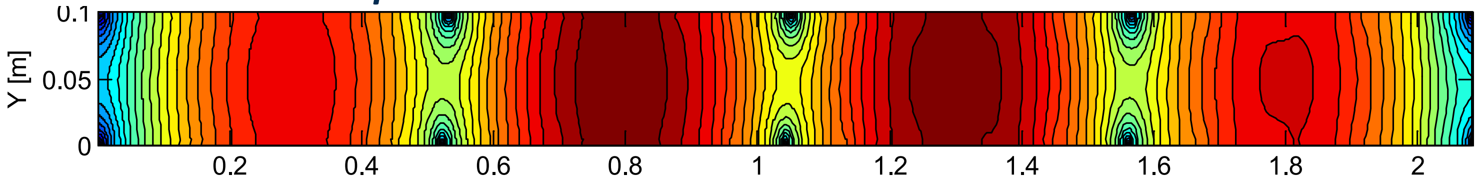
High generation rate of transfer current near tabs

Modeling Results

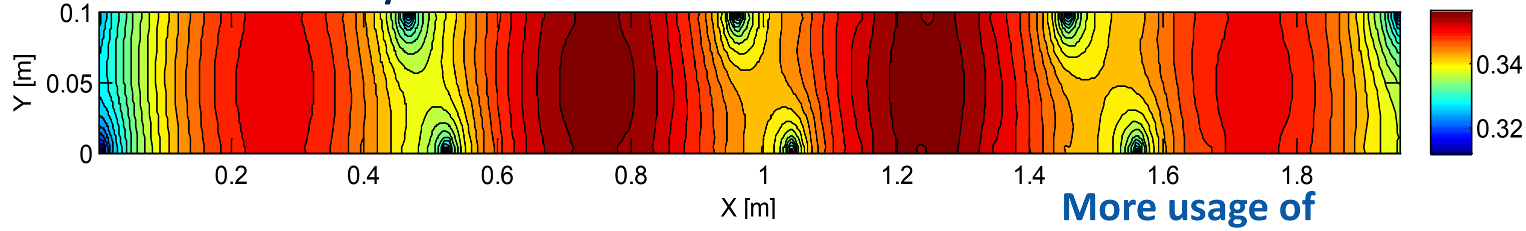


State of Charge

Inner electrode pair

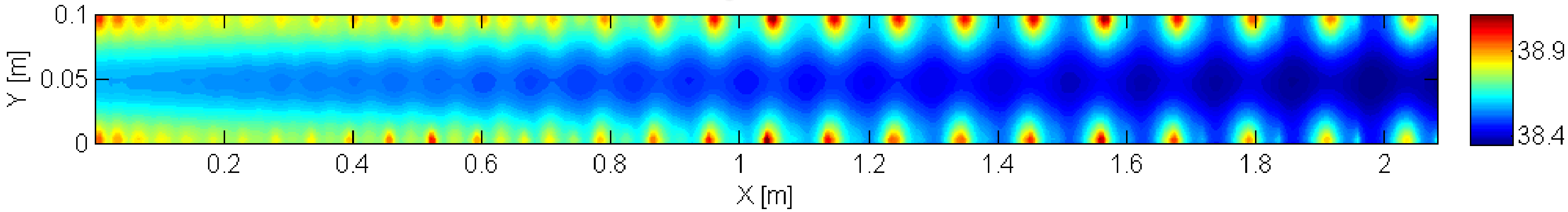


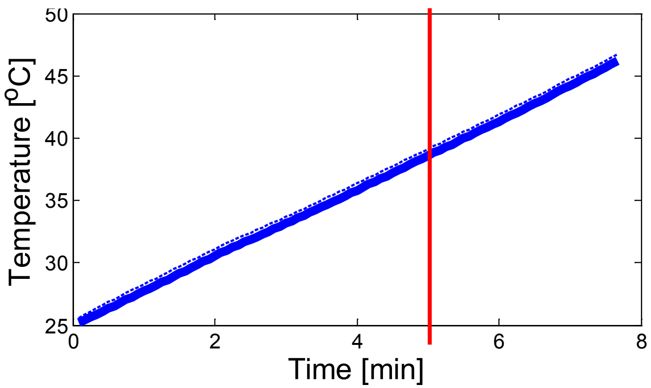
Outer electrode pair

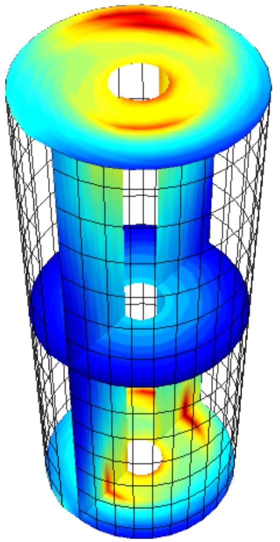


More usage of electrode near tabs

Temperature



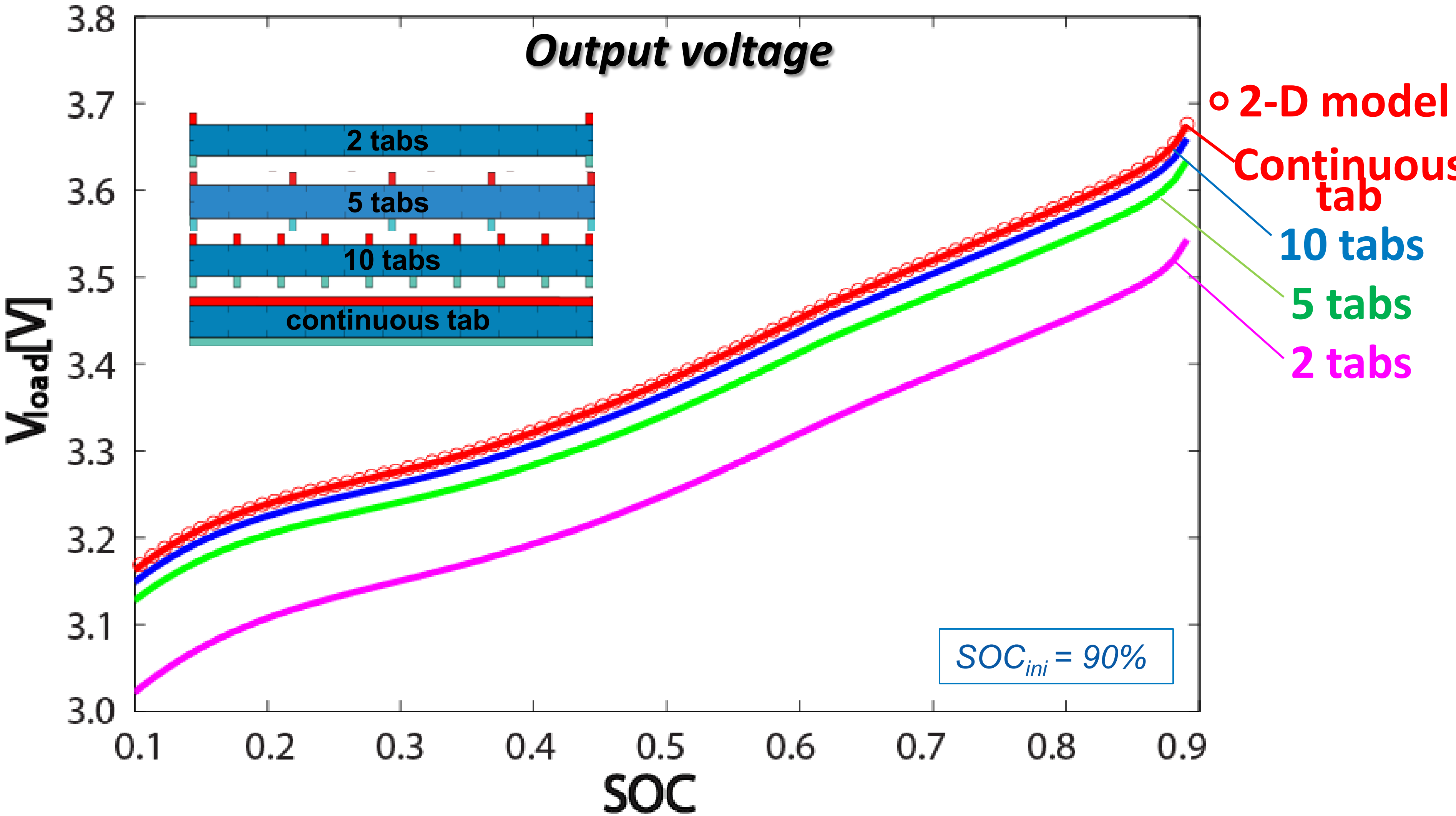




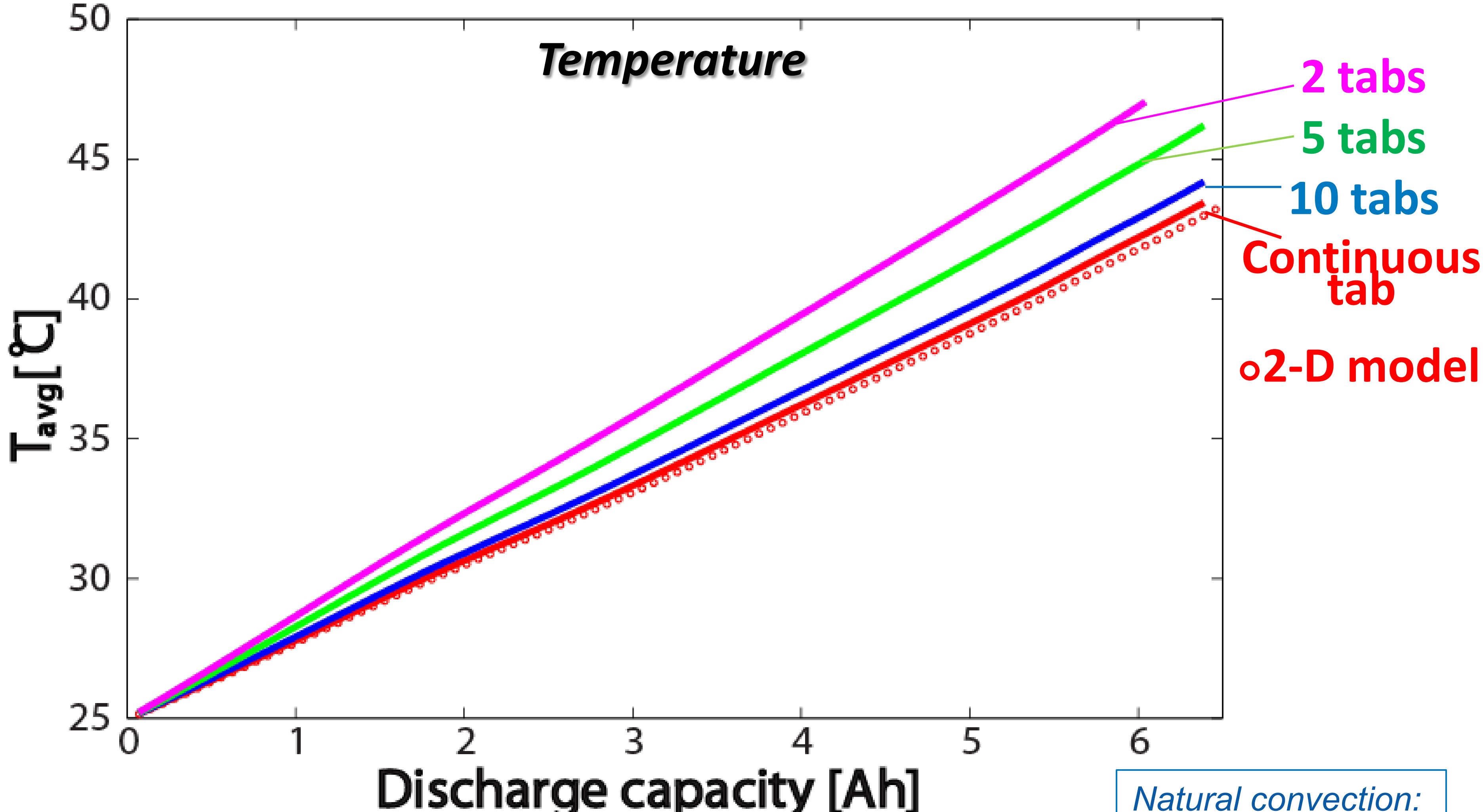
- **Radial heat transfer from tabs**
- **Temperature difference is relatively small**

Modeling Results: *Parametric Study*

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

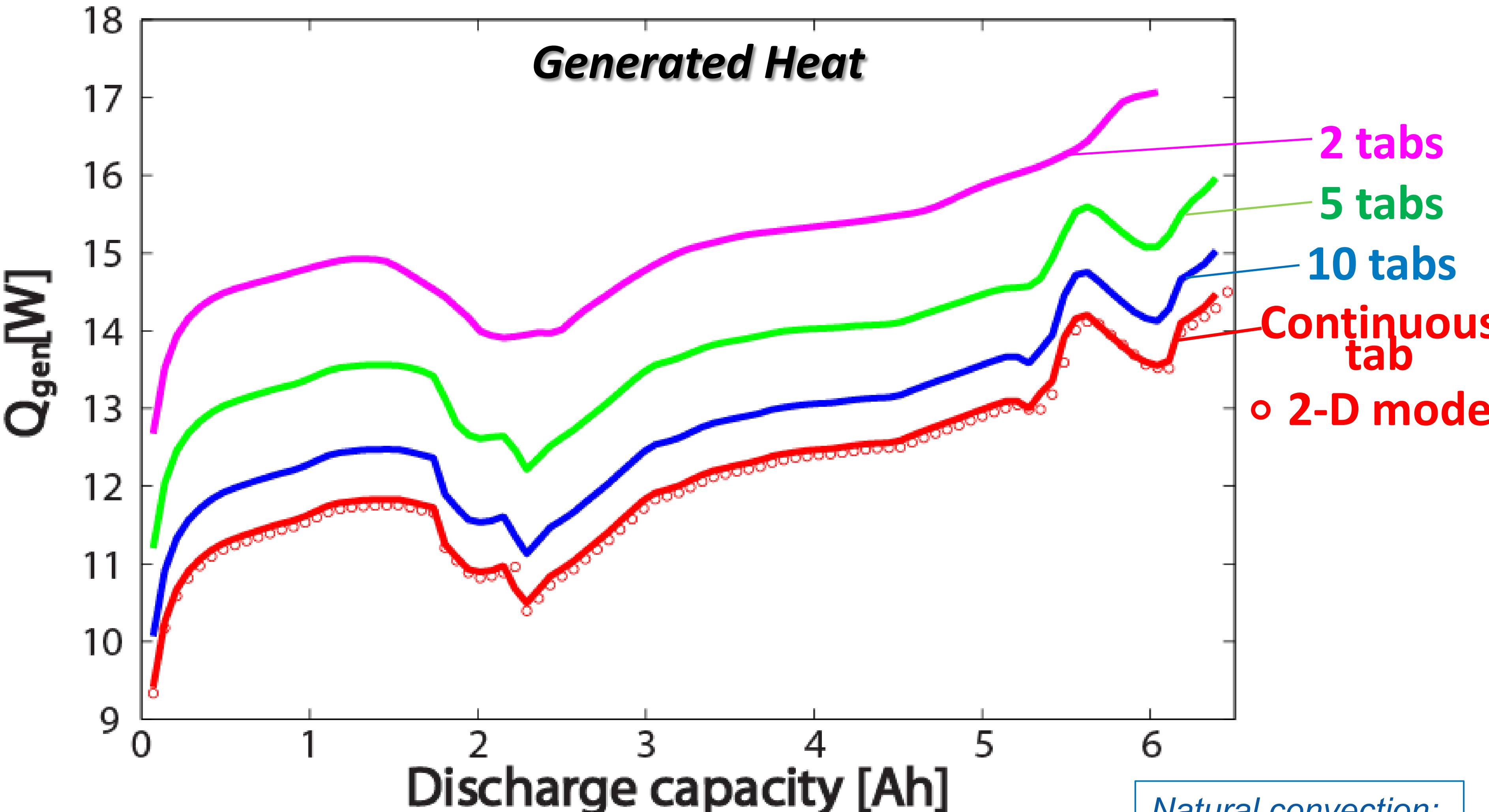


Modeling Results: *Parametric Study*



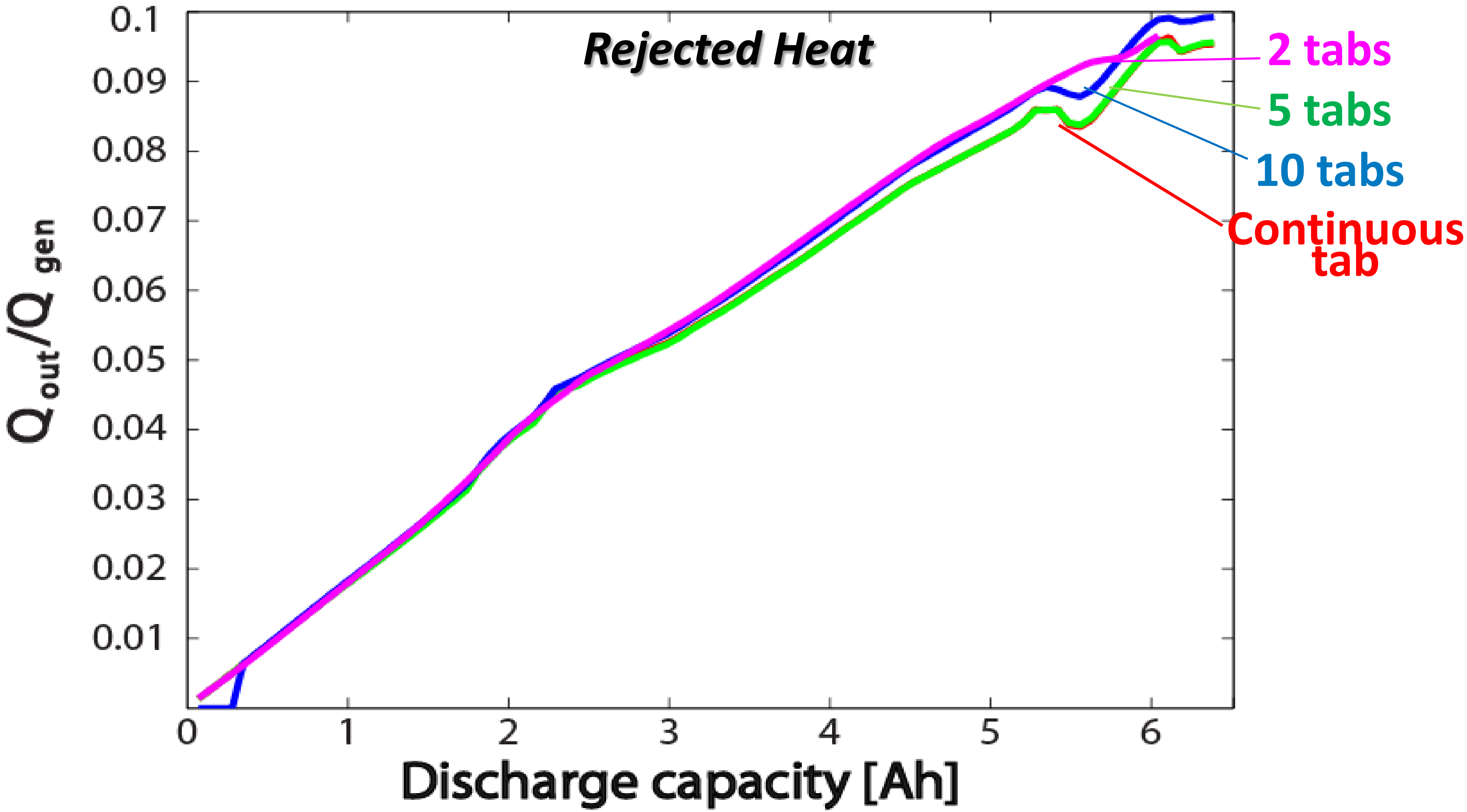
Natural convection:
h_{inf} = 5 W/m²K
T_{amb} = 25°C
T_{ini} = 25°C

Modeling Results: *Parametric Study*



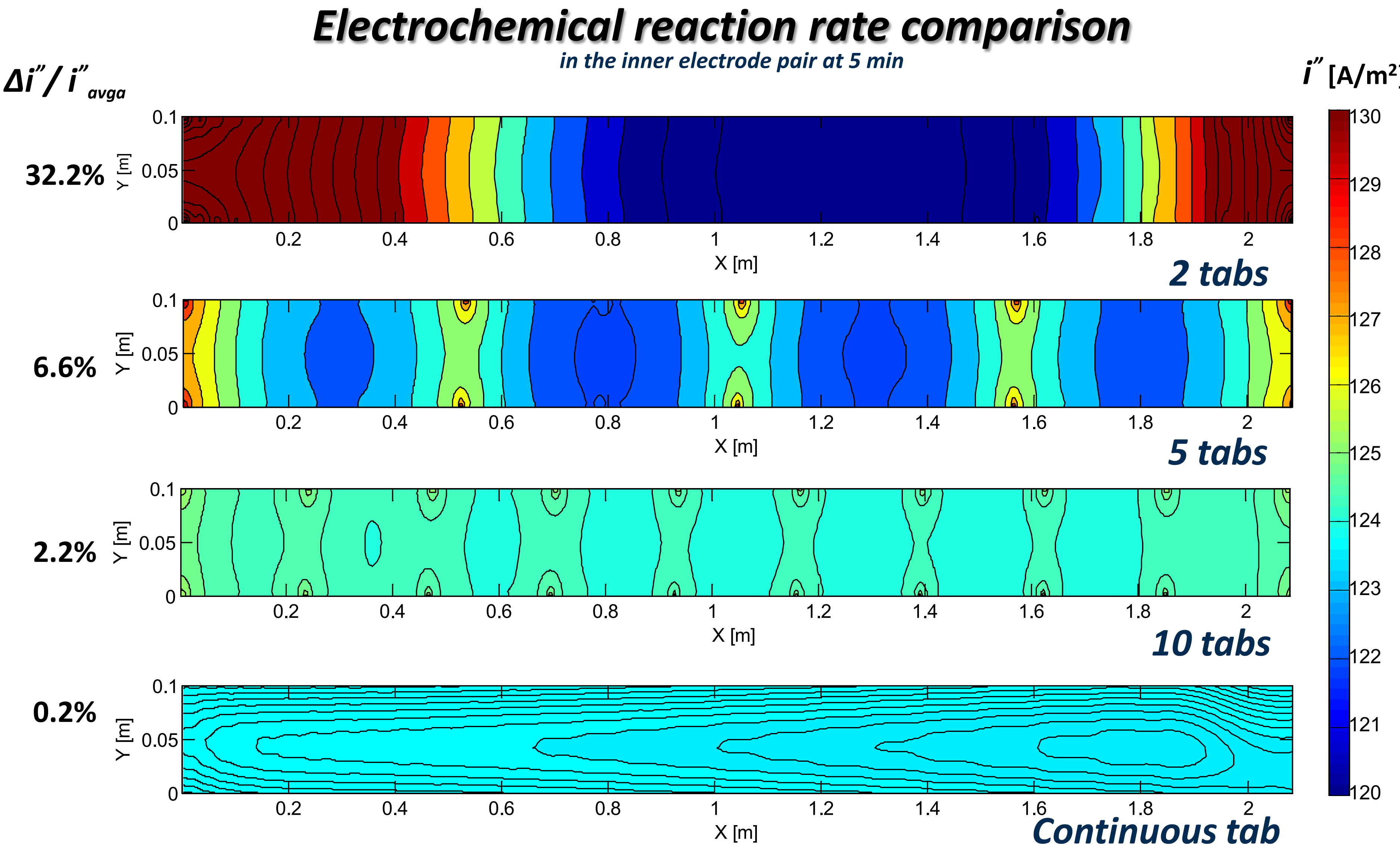
Natural convection:
h_{inf} = 5 W/m^{2}K
T_{amb} = 25°C
T_{ini} = 25°C

Modeling Results: *Parametric Study*



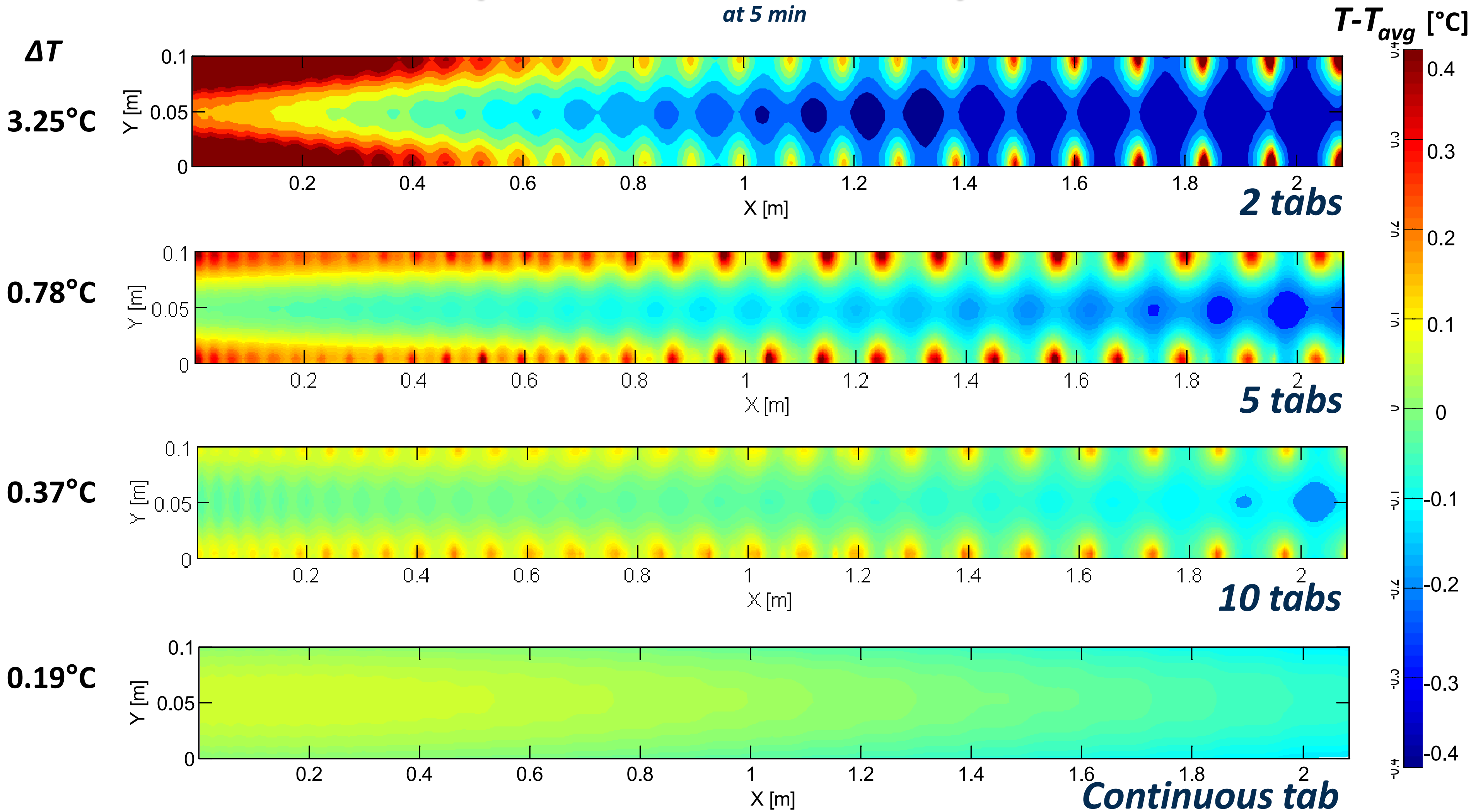
- High rate of discharge with a moderate heat transfer condition
- Heat generation dominates temperature distribution in the system

Modeling Results: *Parametric Study*



Modeling Results: *Parametric Study*

Temperature deviation comparison



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.

Acknowledgments

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*