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

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

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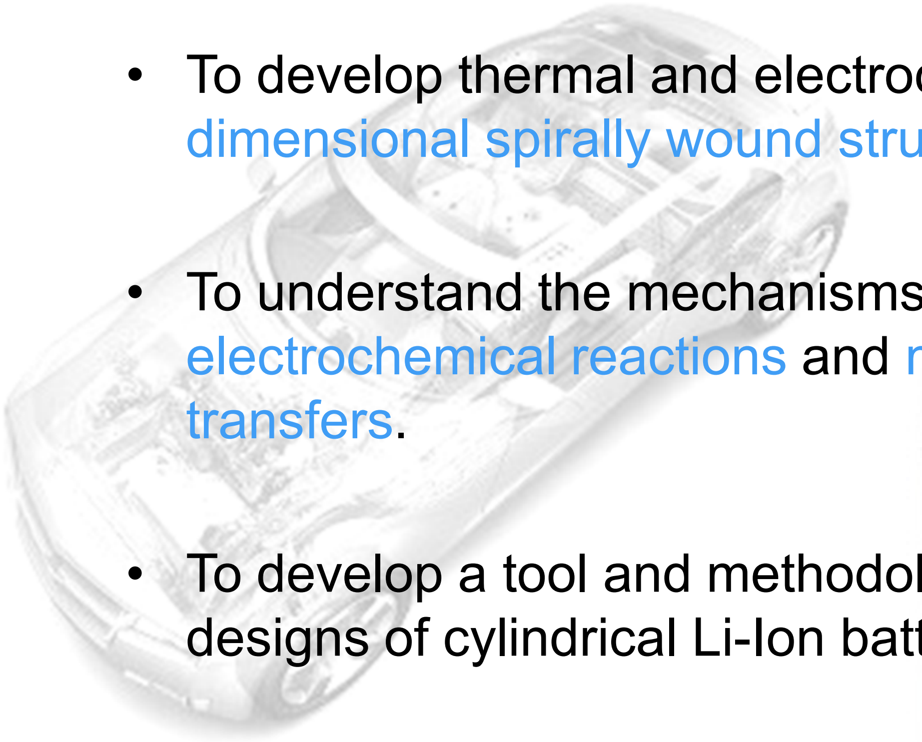
Objectives of this Study

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



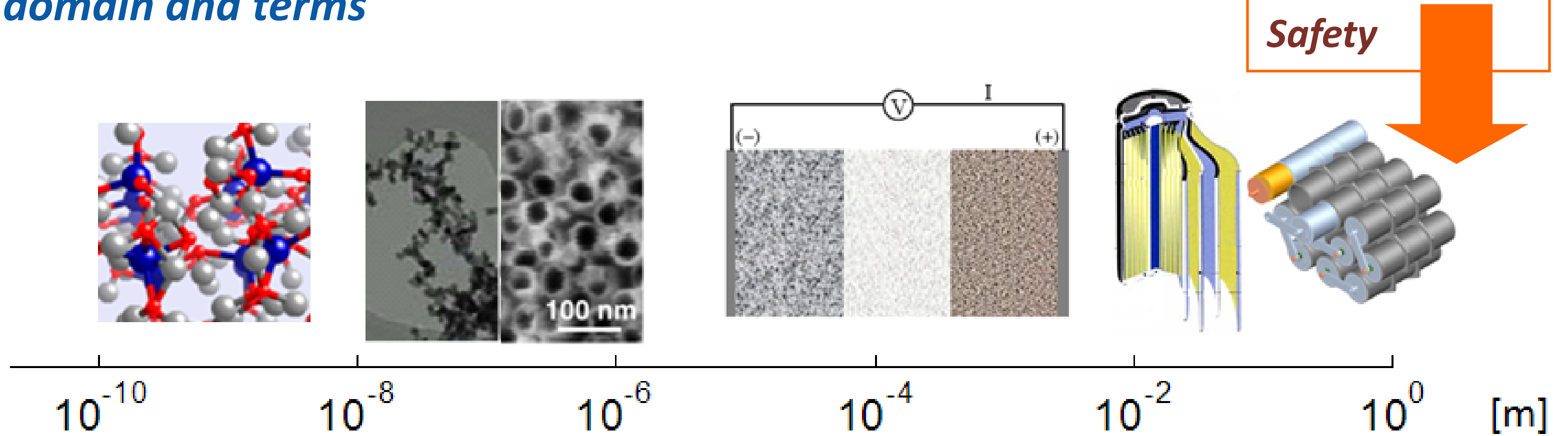
- To develop thermal and electrochemical models resolving 3 dimensional spirally wound structures of cylindrical cells.
- To understand the mechanisms and interactions between local electrochemical reactions and macroscopic heat and electron transfers.
- To develop a tool and methodology to support macroscopic 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

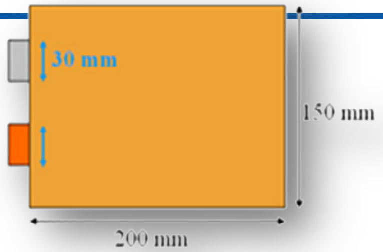
Performance
Life
Cost
Safety



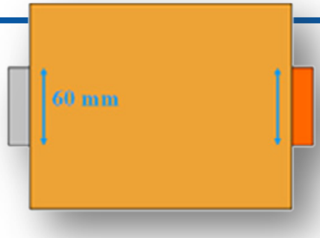
Design of Materials	Design of Electrode Architecture	Design of Transport at Electrode/Electrolyte	Design of Electron & Heat Transport

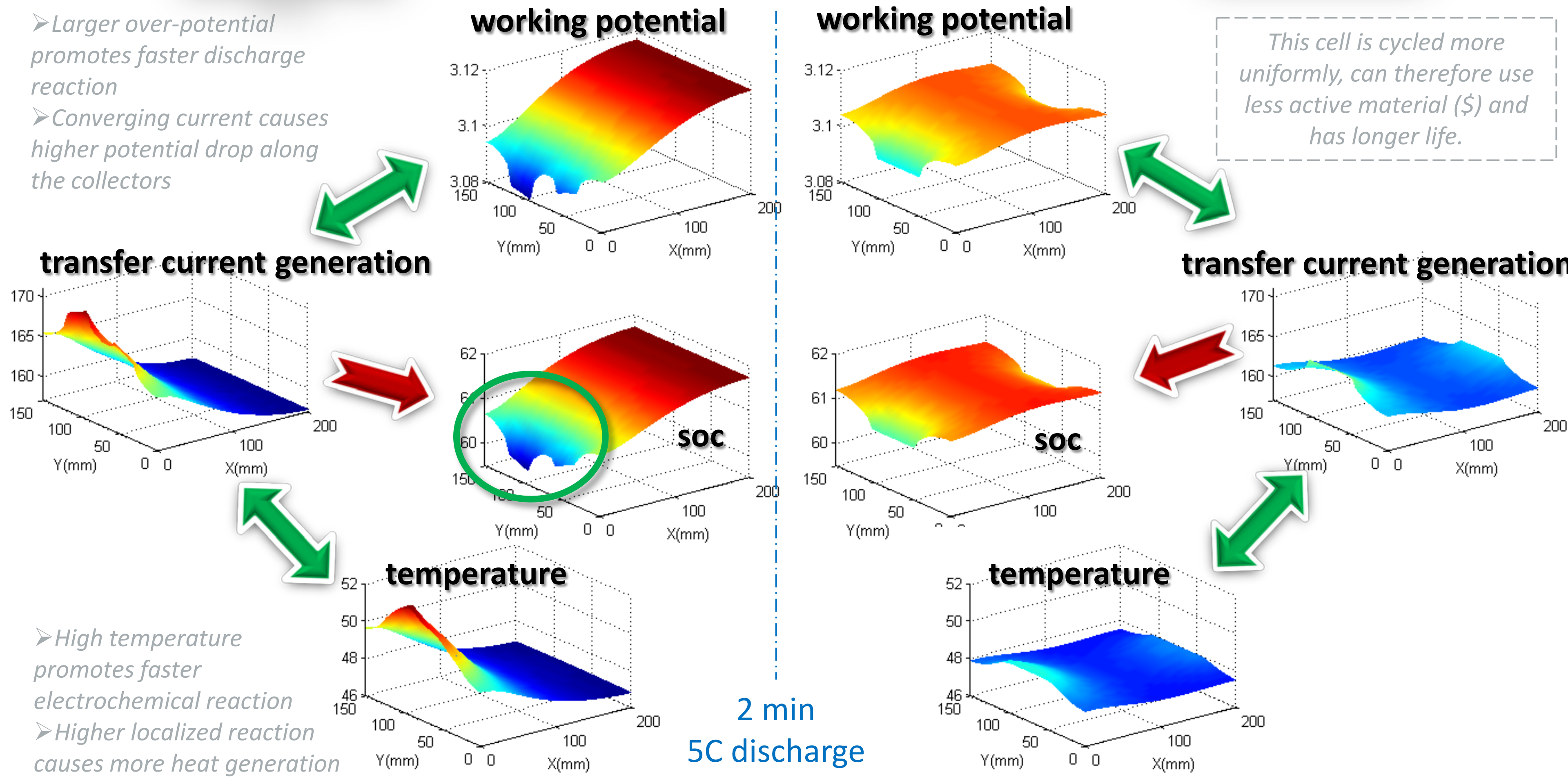
- Wide range of length and time scale physics
- Design improvements required at different scales
- Need for better understanding of interaction among varied scale physics

Multi-Physics Interaction - Previous study



Comparison of two 40 Ah flat cell designs





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

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

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

2 min
5C discharge

Porous Electrode Model

Charge Transfer Kinetics at Reaction Sites

$$j^{Li} = a_s i_o \left\{ \exp\left[\frac{\alpha_a F}{RT} \eta \right] - \exp\left[-\frac{\alpha_c F}{RT} \eta \right] \right\}$$

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

Species Conservation

$$\frac{\partial c_s}{\partial t} = \frac{D_s}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_s}{\partial r} \right)$$

$$\frac{\partial (\varepsilon_e c_e)}{\partial t} = \nabla \cdot \left(D_e^{eff} \nabla c_e \right) + \frac{1 - t_+^o}{F} j^{\mathrm{Li}} - \frac{\mathbf{i}_e \cdot \nabla t_+^o}{F}$$

Charge Conservation

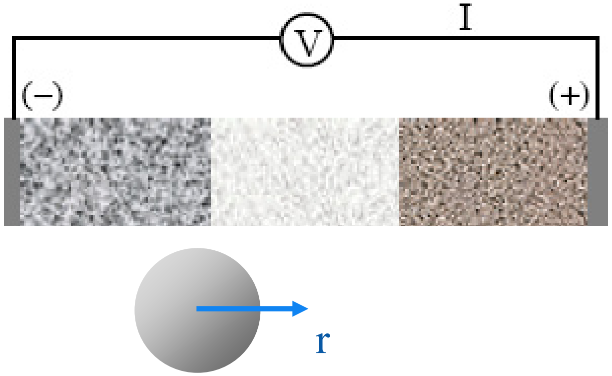
$$\nabla \cdot \left(\sigma^{eff} \nabla \phi_s \right) - j^{\mathrm{Li}} = 0$$

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

Energy Conservation

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

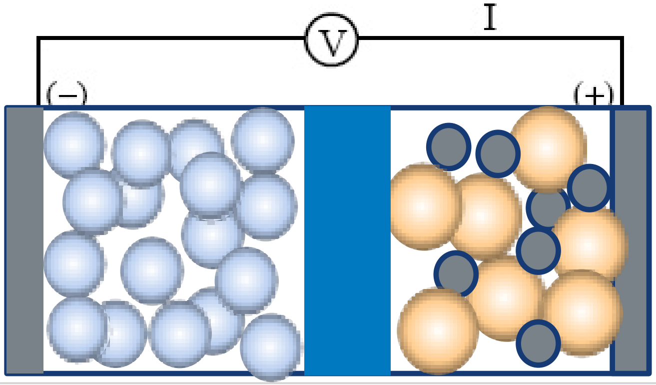
$$q''' = j^{Li} \left(\phi_s - \phi_e - U + T \frac{\partial U}{\partial T} \right) + \sigma^{eff} \nabla \phi_s \cdot \nabla \phi_s + \kappa^{eff} \nabla \phi_e \cdot \nabla \phi_e + \kappa_D^{eff} \nabla \ln c_e \cdot \nabla \phi_e$$



- 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



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- Captures ***lithium diffusion dynamics*** and ***charge transfer kinetics***
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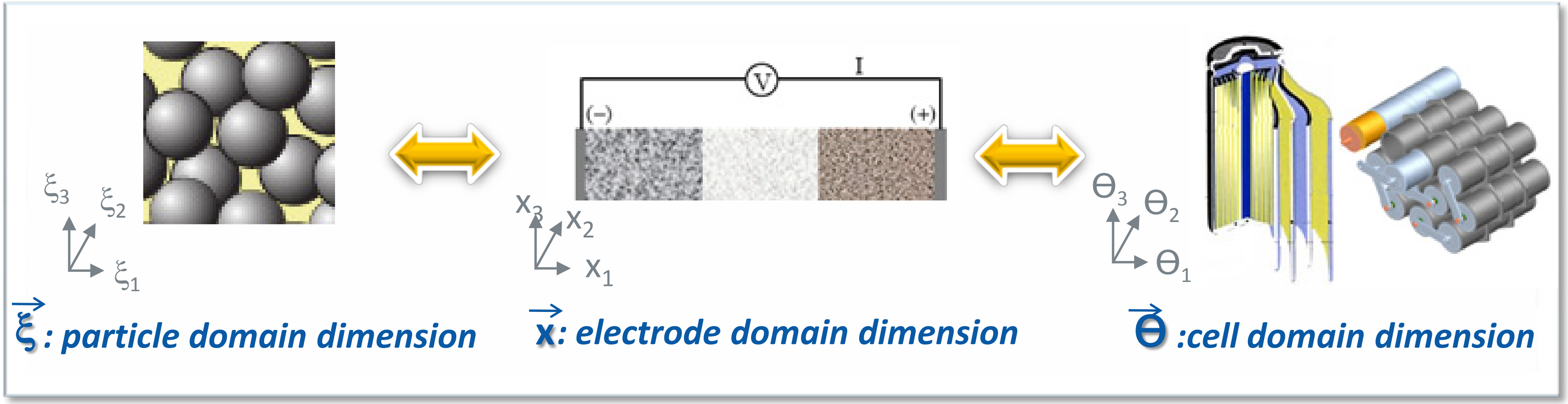
Multi-Scale Multi-Dimension (MSMD) Model



- 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)*

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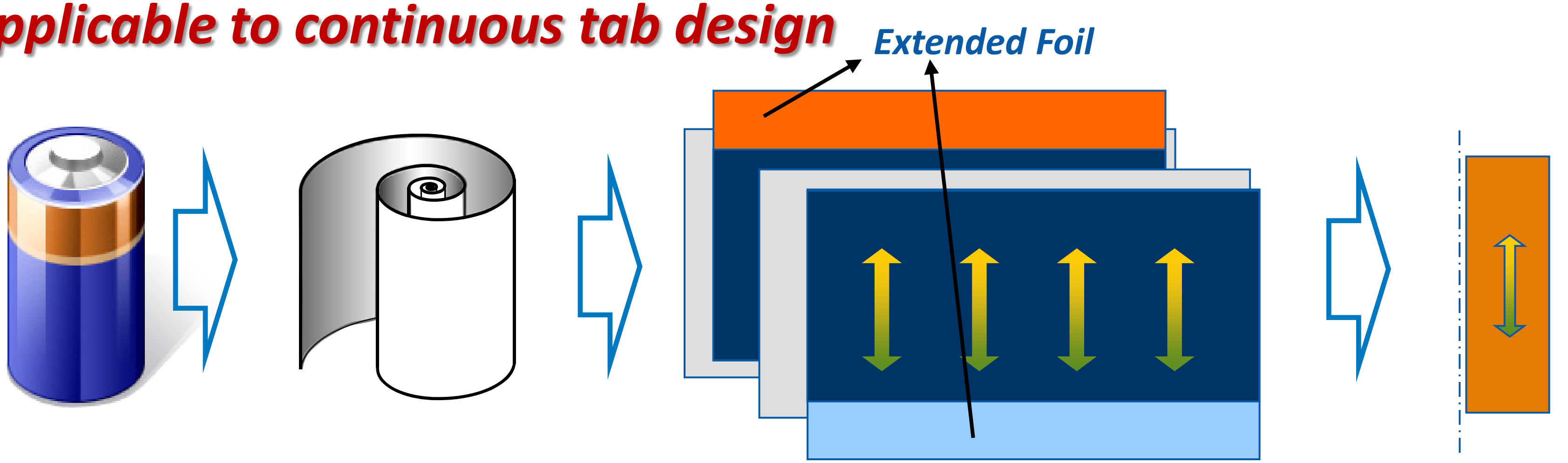
Previous Development of Wound Cell Model

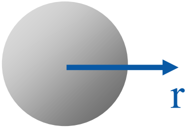
Applicable to continuous tab design



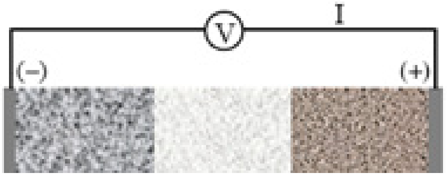
Cylindrical cell **Unwinding jellyrolls** **continuous tab design** **Axisymmetric assumption**

Sub-model choice for spirally wound continuous tab cells *- Previous study*

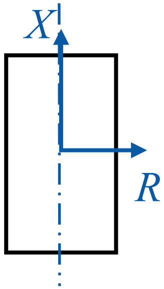
: particle domain sub-model *: electrode domain sub-model* *:cell domain sub-model*



1D spherical particle representation model



1D porous electrode model

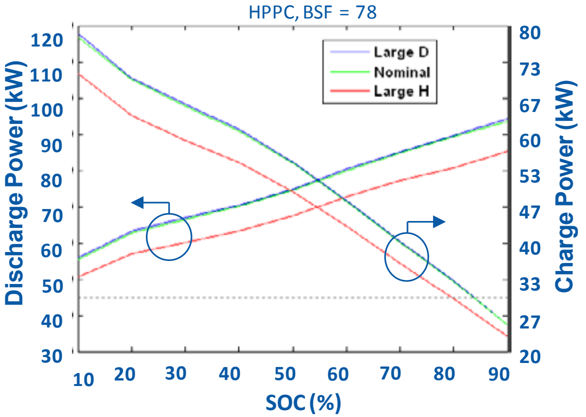


2D axisymmetric cell model

Tab-less Wound Cell Design Evaluation

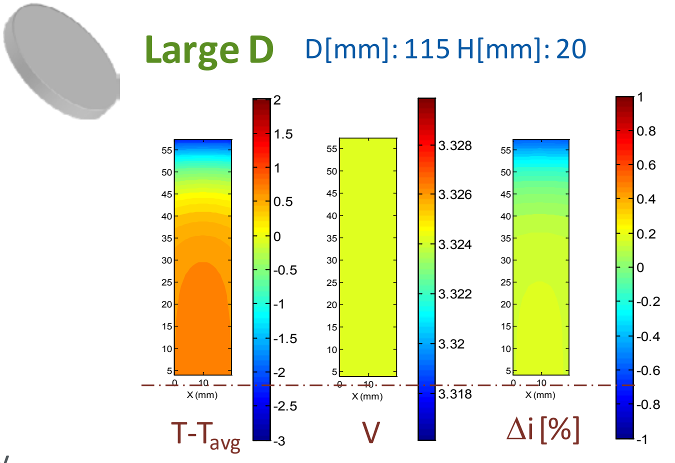
Effects of “Aspect Ratio” of a Cylindrical Cell *- Previous study*

10s pulse power capability comparison

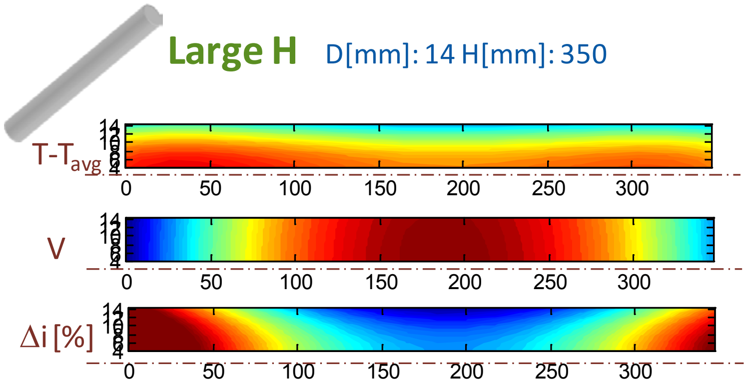


- Large H design has almost 10% less power capability.

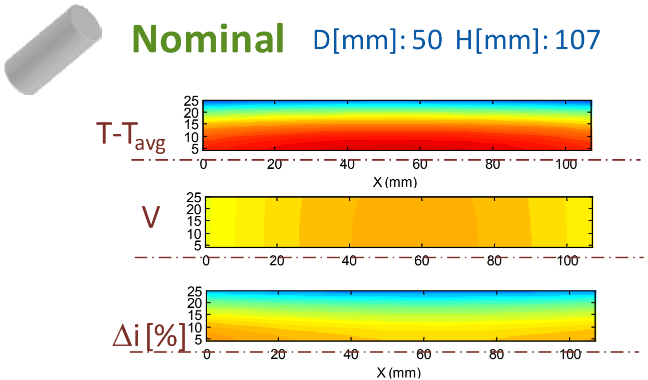
9 min 5C discharge



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



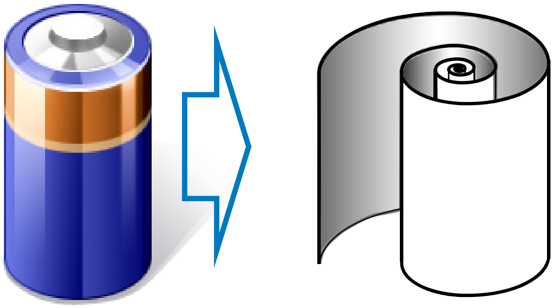
Large H D[mm]: 14 H[mm]: 350



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

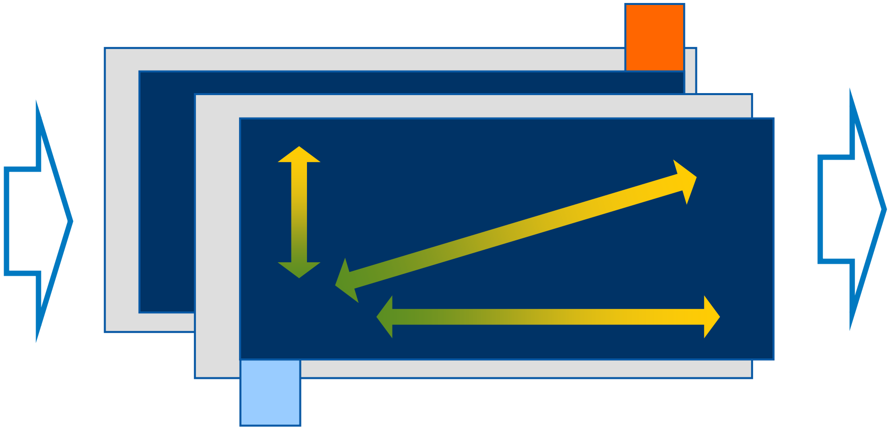
Present Study

Tab configuration: number & size

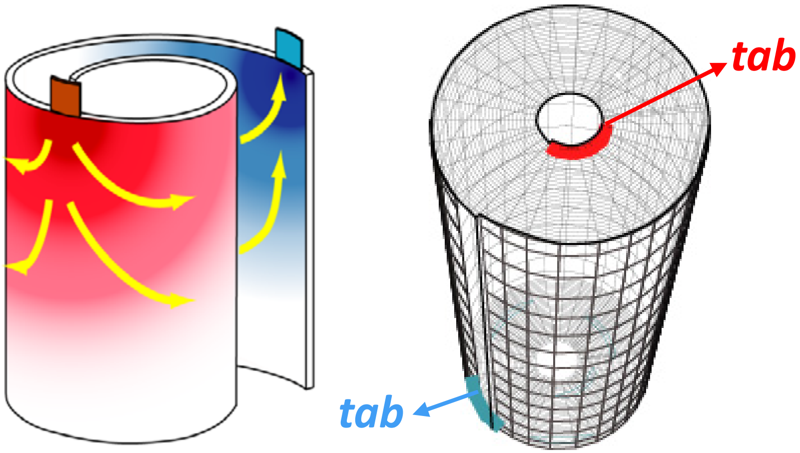


Cylindrical cell **Unwinding jellyrolls**

- 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.



Current flows along the winding direction

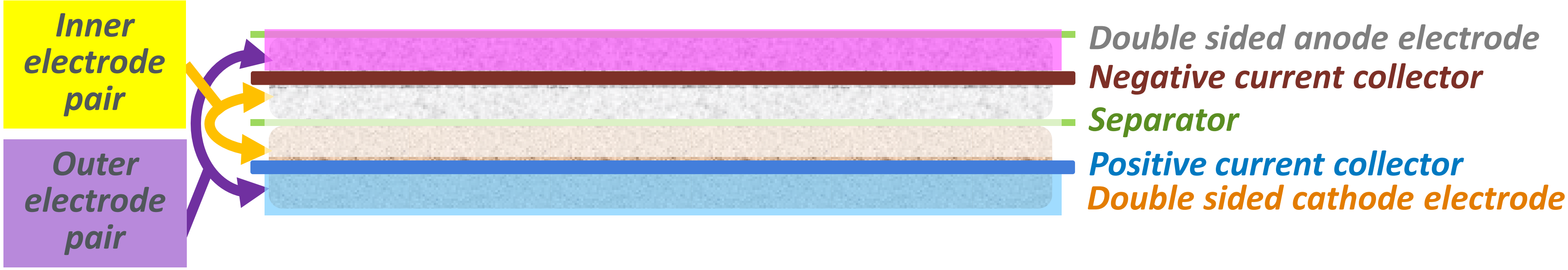


3 dimensional spiral wound geometry

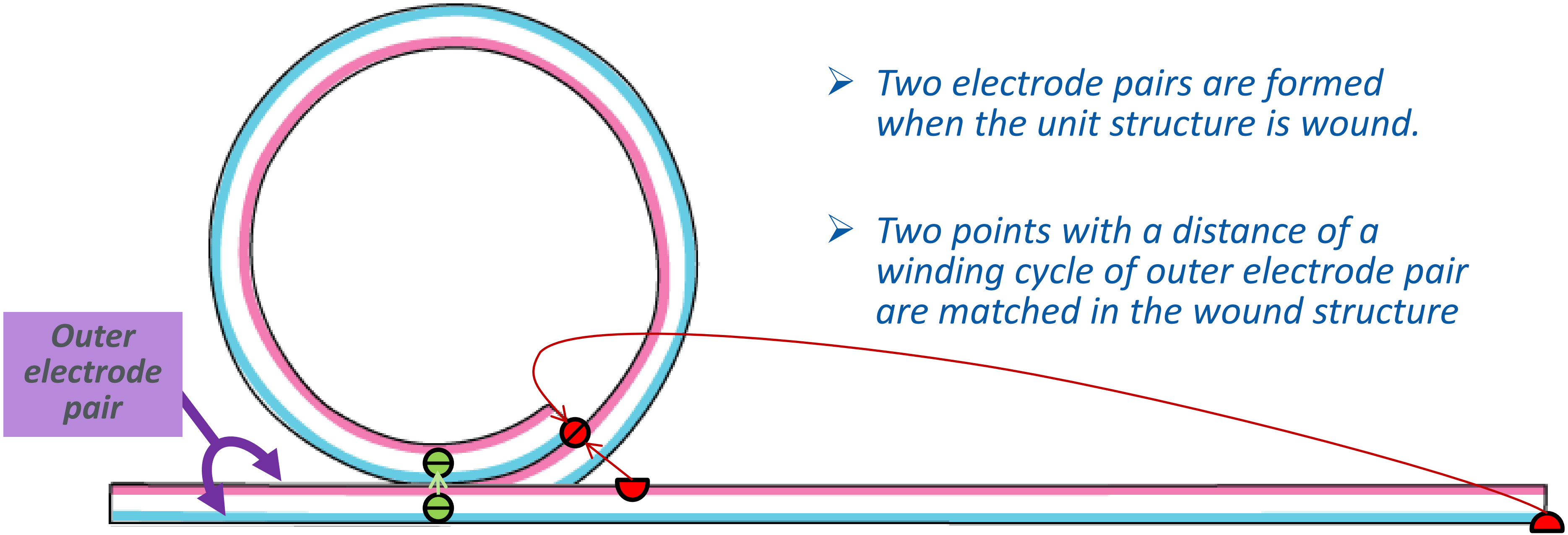
New Development of Cell Domain Model

Spirally Wound Cell Model:

Unit structure: *Double Paired Electrodes on Single-Paired Current Collectors*



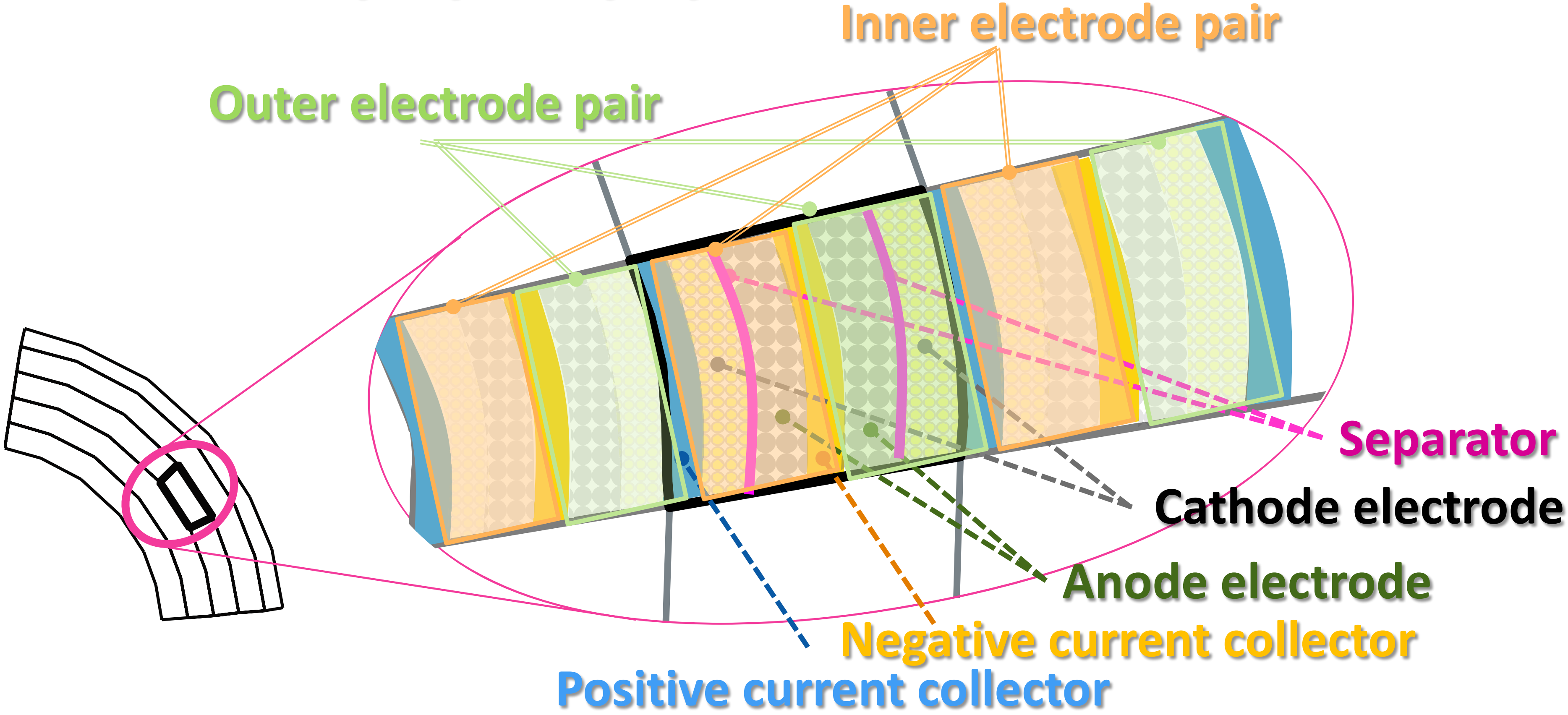
Winding: *Alternating Radial Placement of Double Paired-Electrodes*



- *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

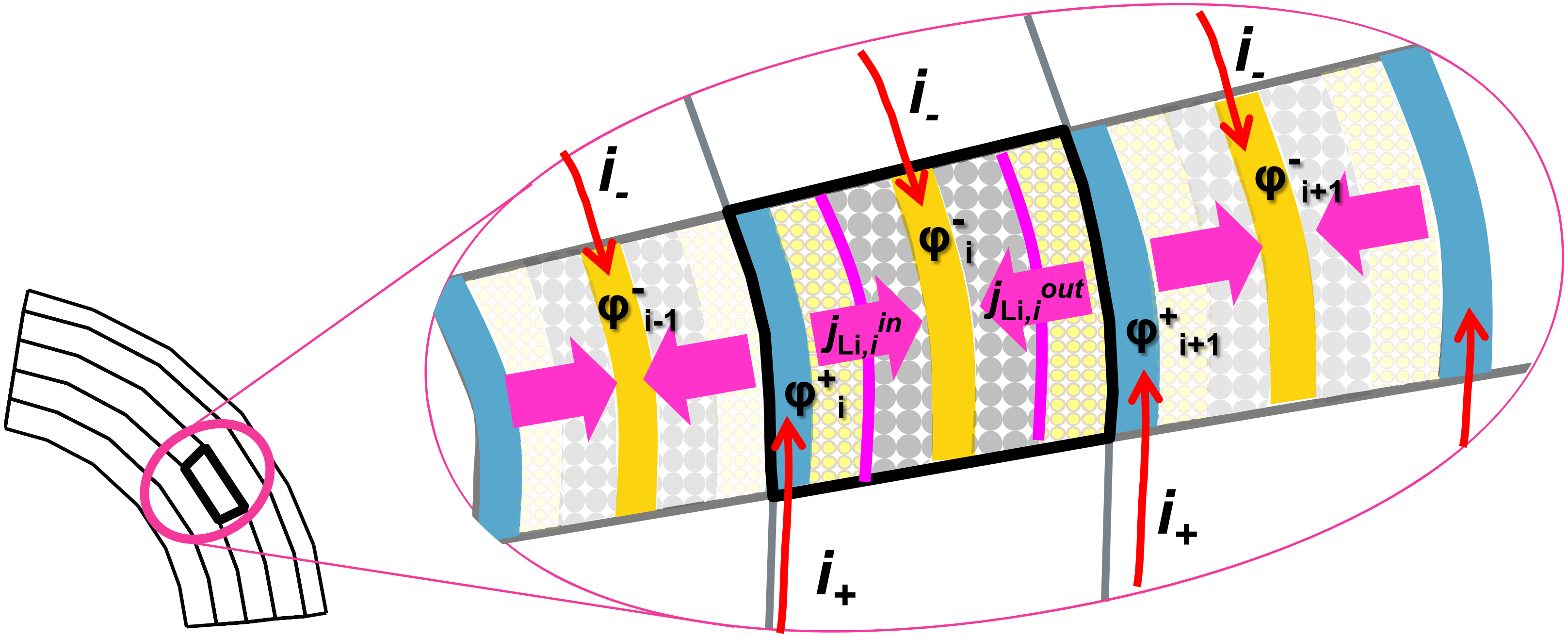


A current collector has two electrode pairs in both sides.

Spiral cell structures

Non-uniform electrical potential along current collectors

Non-uniform charge transfer reaction

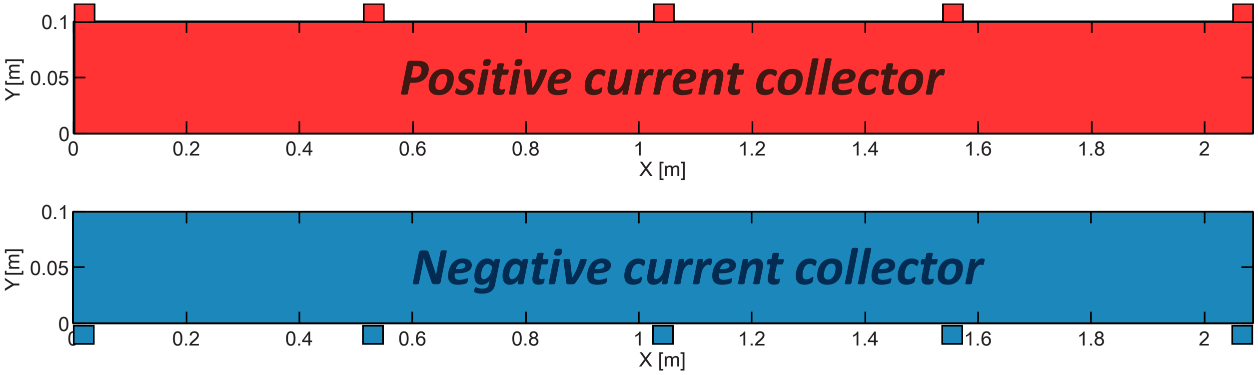


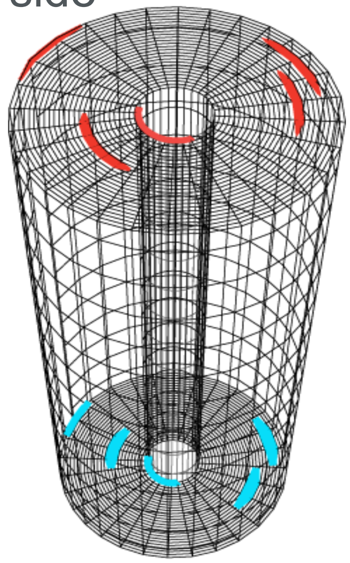
We cannot expect uniform potential along the current collectors due to inevitable electric current in winding direction.

Modeling case

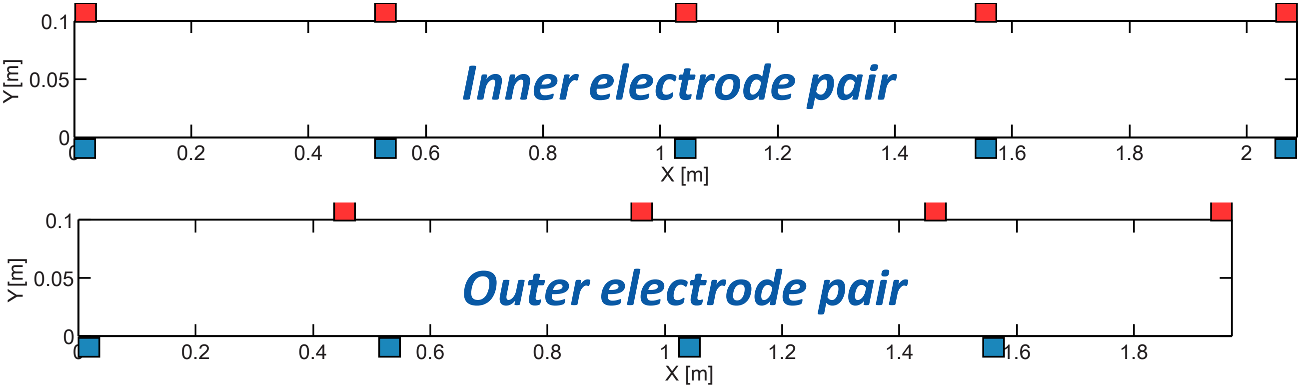
- ✓ Diameter 40mm, inner diameter 8mm, height 100 mm form factor
- ✓ Positive tabs on the top side, negative tabs on the bottom side
- ✓ 10 Ah capacity

Tab locations for 5 tab case





Tab configuration of each electrode pairs



5C constant current discharge

$soc_{ini} = 90\%$

Natural convection :

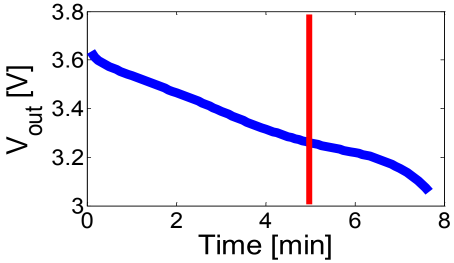
$h_{inf} = 5\ W/m^2K$

$T_{amb} = 25^oC$

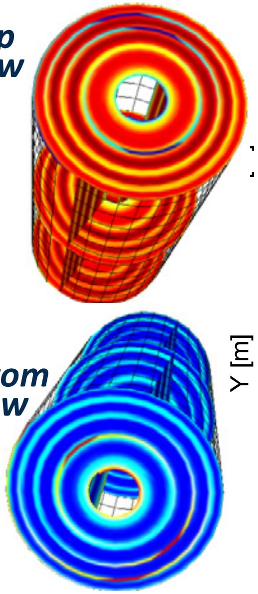
$T_{ini} = 25^oC$

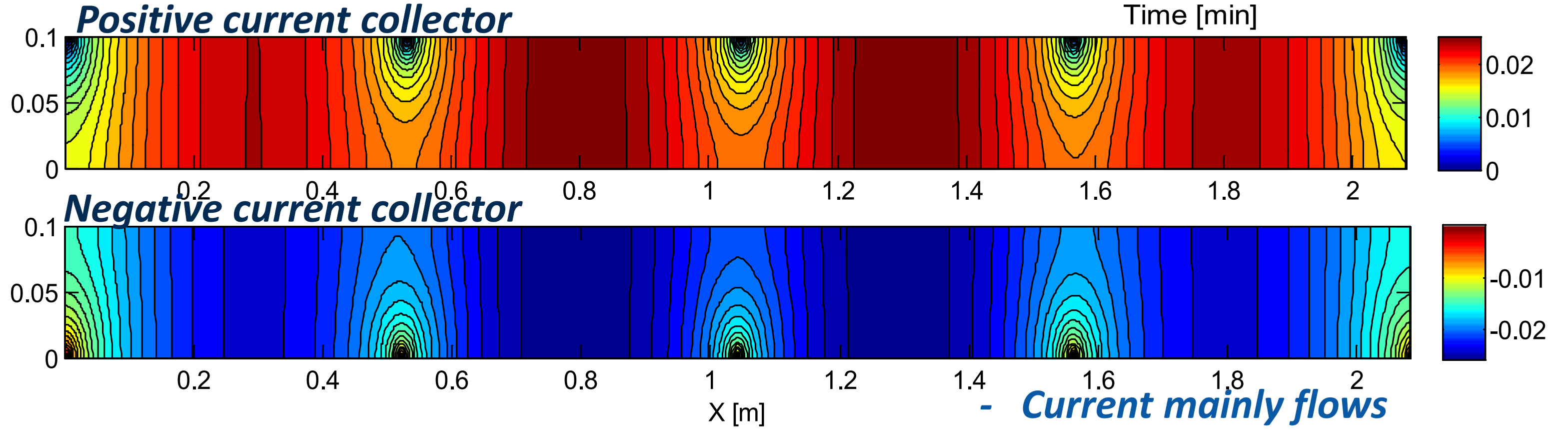
Modeling results

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



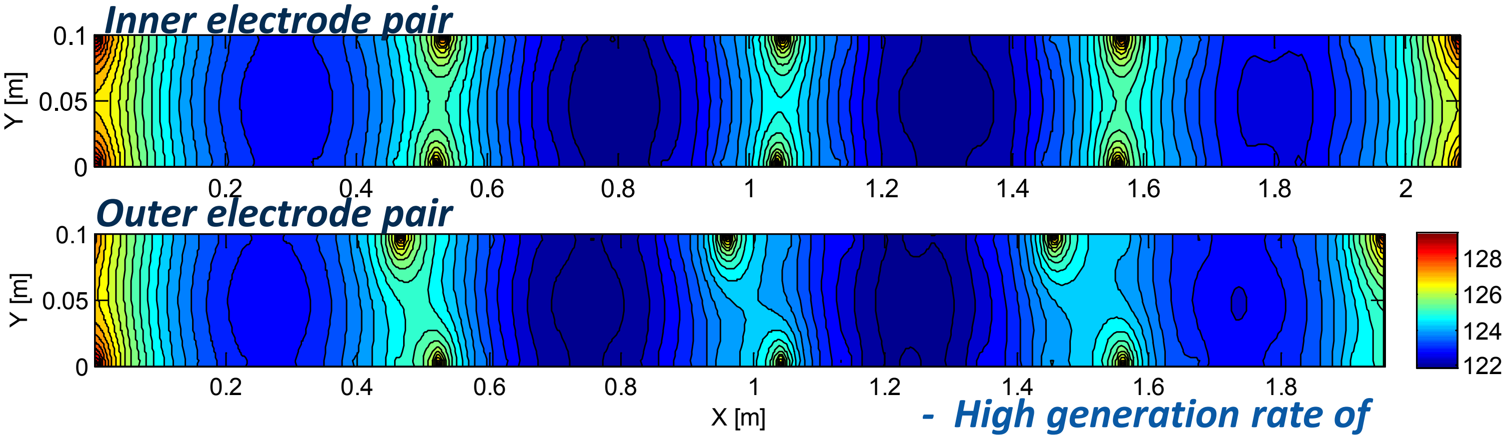
Electric potential

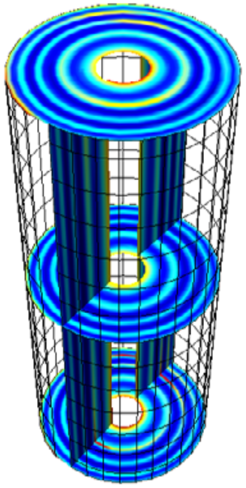




- Current mainly flows in the winding direction

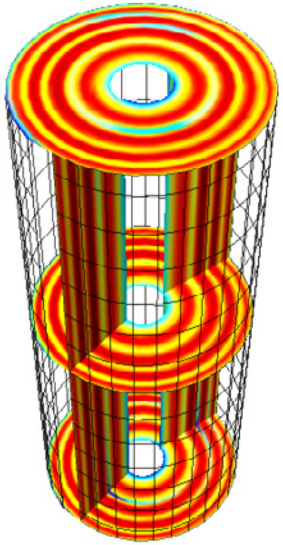
Electrochemical reaction rate



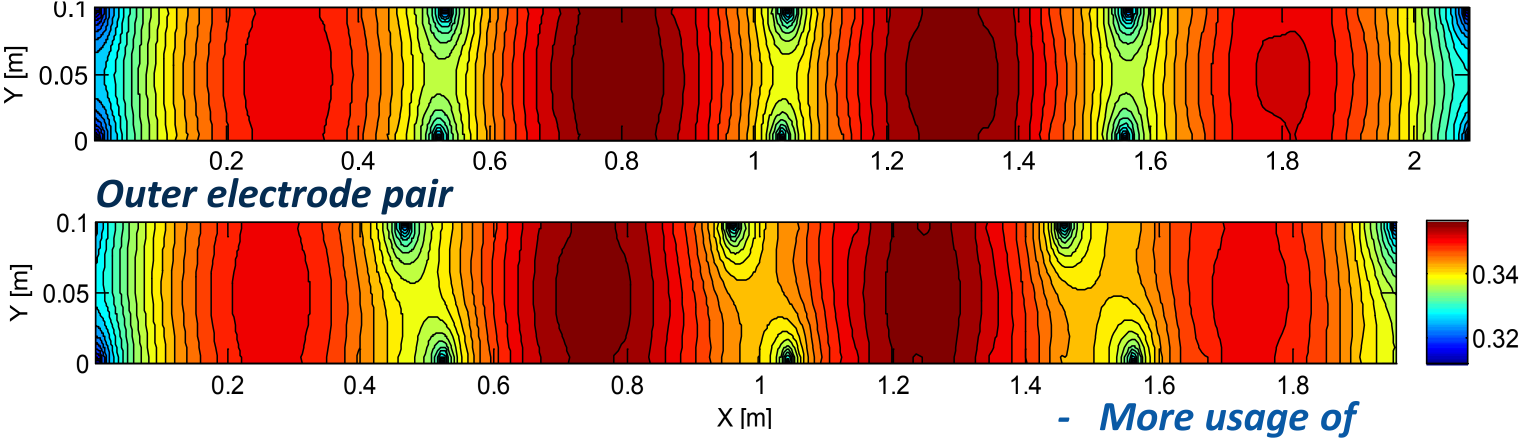


- High generation rate of transfer current near tabs

Modeling results



State of charge

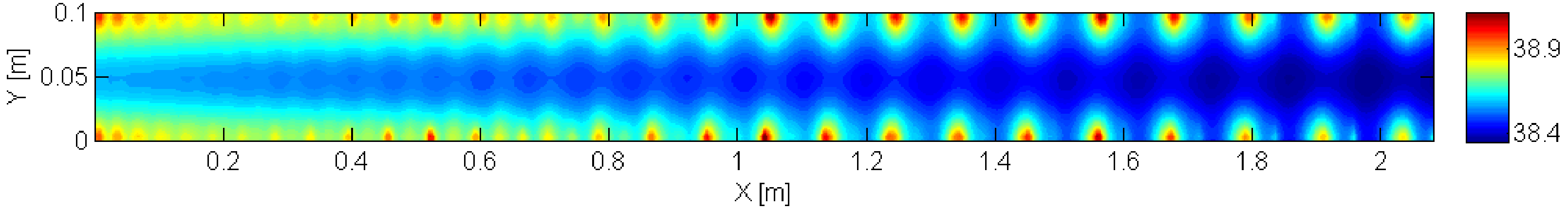


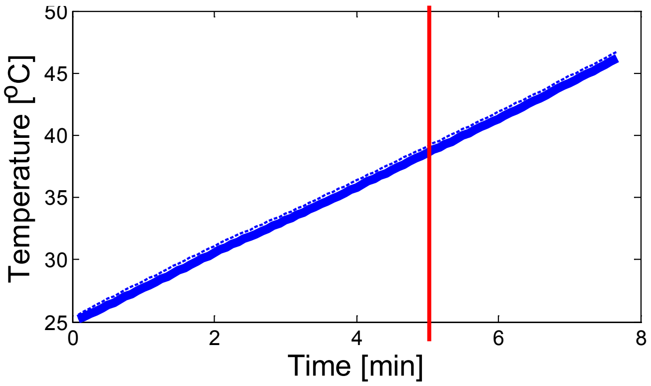
Inner electrode pair

Outer electrode pair

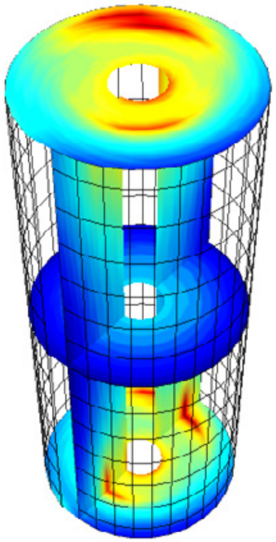
- ***More usage of electrode near tabs***

Temperature





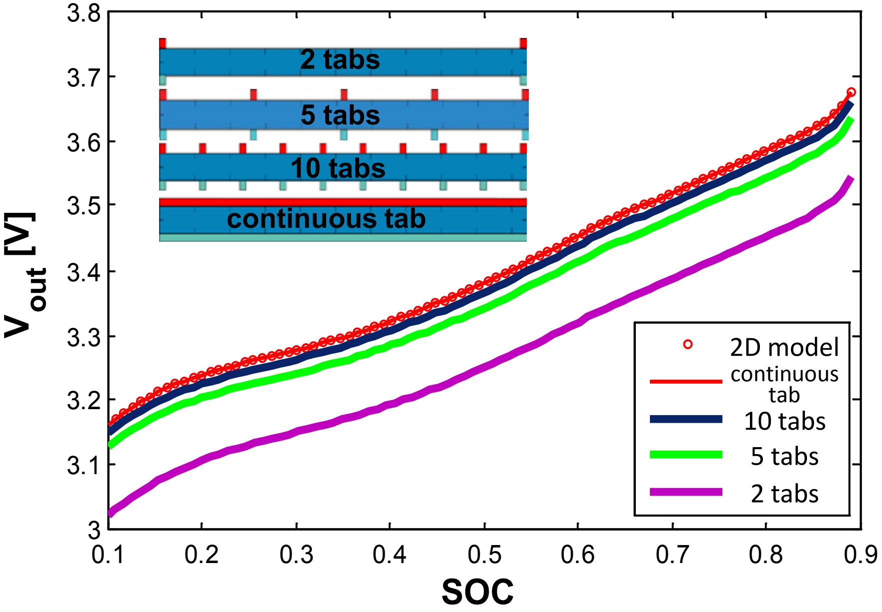
- ***High rate discharge with a moderate heat transfer condition***
- ***Heat generation dominates temperature distribution in the system.***
- ***Temperature difference in the system is relatively small yet.***

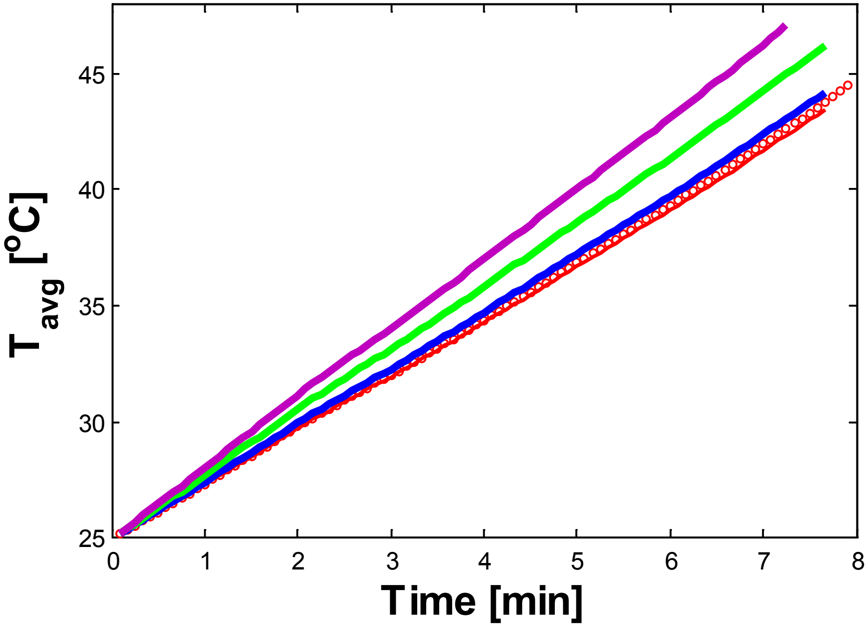


Modeling results

Parametric study

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





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Natural convection :
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T_{amb} = 25^{o}C
T_{ini} = 25^{o}C

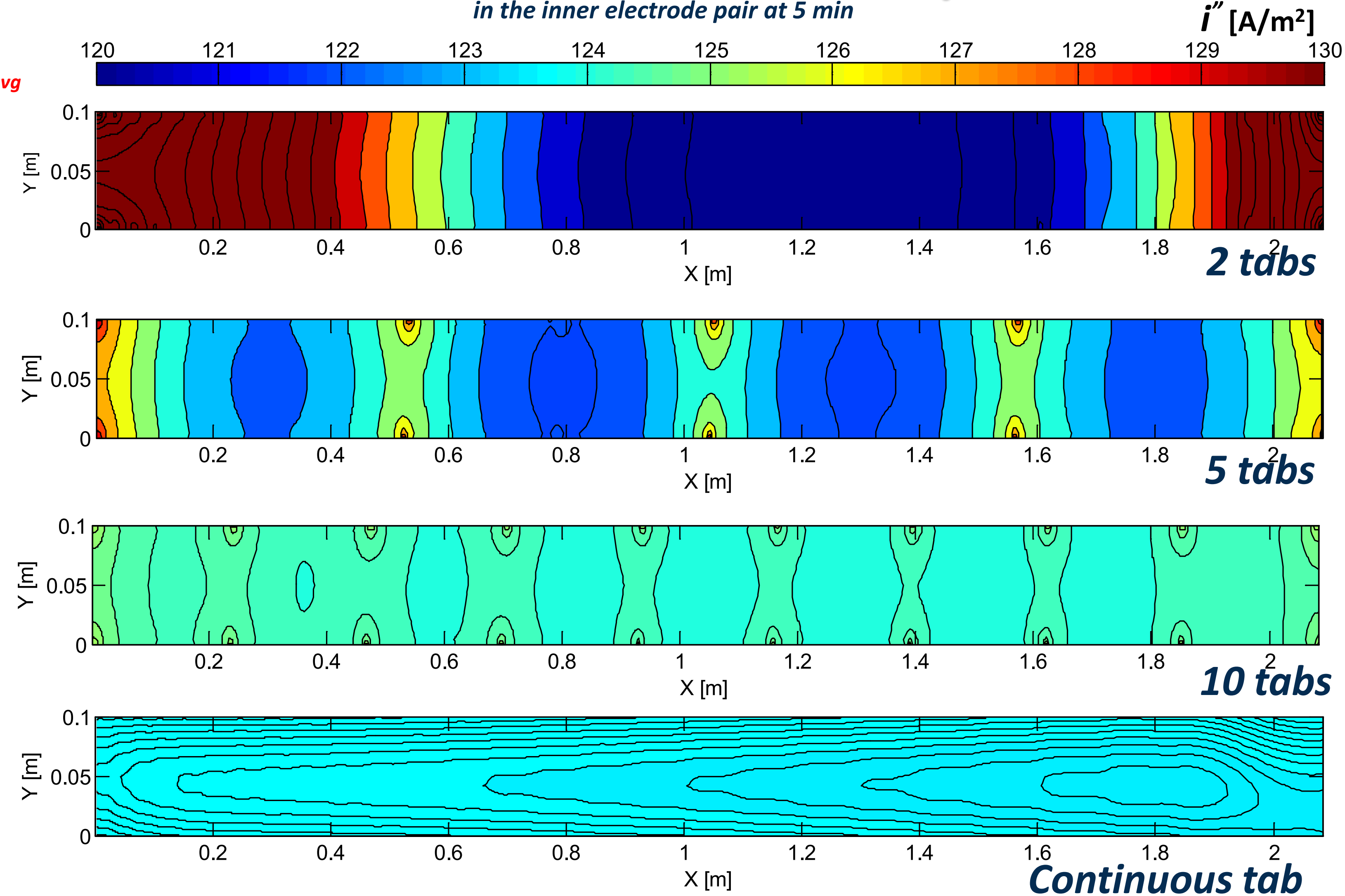
Cells with fewer tabs …

- ***lower output voltage***
- ***higher average temperature***

Modeling results - Parametric study

Electrochemical reaction rate comparison

in the inner electrode pair at 5 min



$\Delta i'' / i''_{avg}$

32.2% — **2 tabs**

6.6% — **5 tabs**

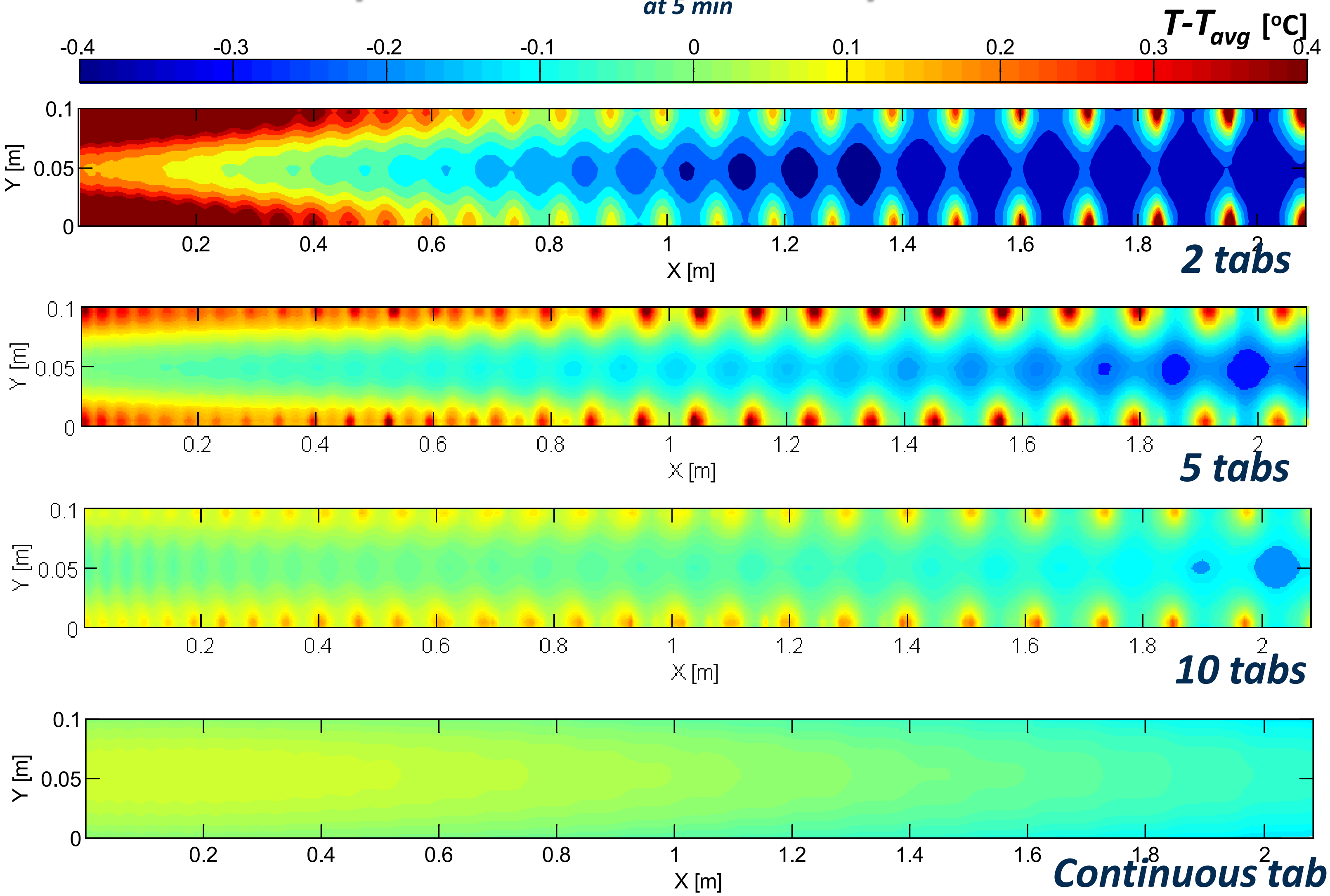
2.2% — **10 tabs**

0.2% — **Continuous tab**

Modeling results - Parametric study

Temperature deviation comparison

at 5 min



ΔT

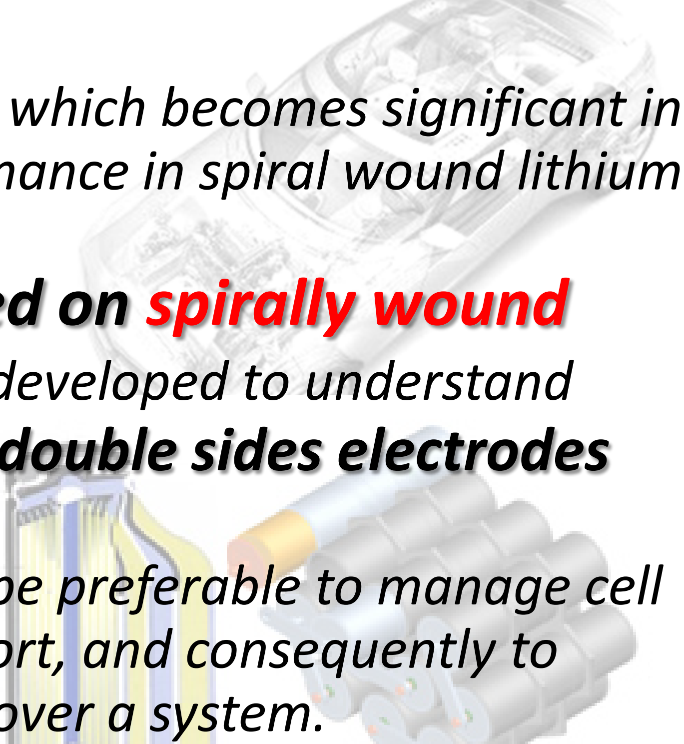
3.25°C — ***2 tabs***

0.78°C — ***5 tabs***

0.37°C — ***10 tabs***

0.19°C — ***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.*

Acknowledgments

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National Renewable Energy Laboratory

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