NREL National Renewable Energy Laboratory
Innovation for Our Energy Future

Multi-Scale Multi-Dimensional Model
for Better Cell Design and Management

1st International Conference on Advanced Lithium Batteries for Automotive Applications
Argonne, September 15–17, 2008

Gi-Heon Kim
Kandler Smith

National Renewable Energy Laboratory

NREL/PR-540-44245

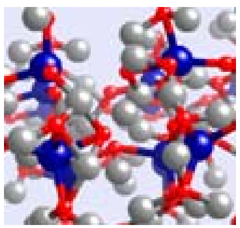
Presented at the 1st International Conference on Advanced Lithium Batteries for Automotive Applications held September 15-17, 2008 in Argonne, Illinois

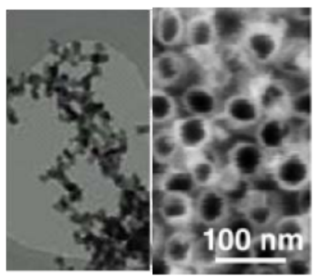
Multi-Scale Physics in Li-Ion Battery

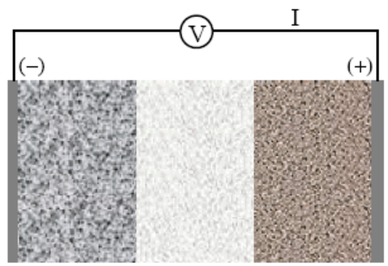
Requirements & Resolutions

"Requirements" are usually defined in a macroscale domain and terms.

Performance
Life
Cost
Safety







10^{-10} 10^{-8} 10^{-6} 10^{-4} 10^{-2} 10^{0} [m]

Design of Materials

Voltage
Capacity
Lattice stability
Kinetic barrier
Transport property

Design of Electrode Architecture

Li transport path (local)
Electrode surface area
Deformation & fatigue
Structural stability
Surface physics

Design of Electrodes Pairing and Lithium Transport

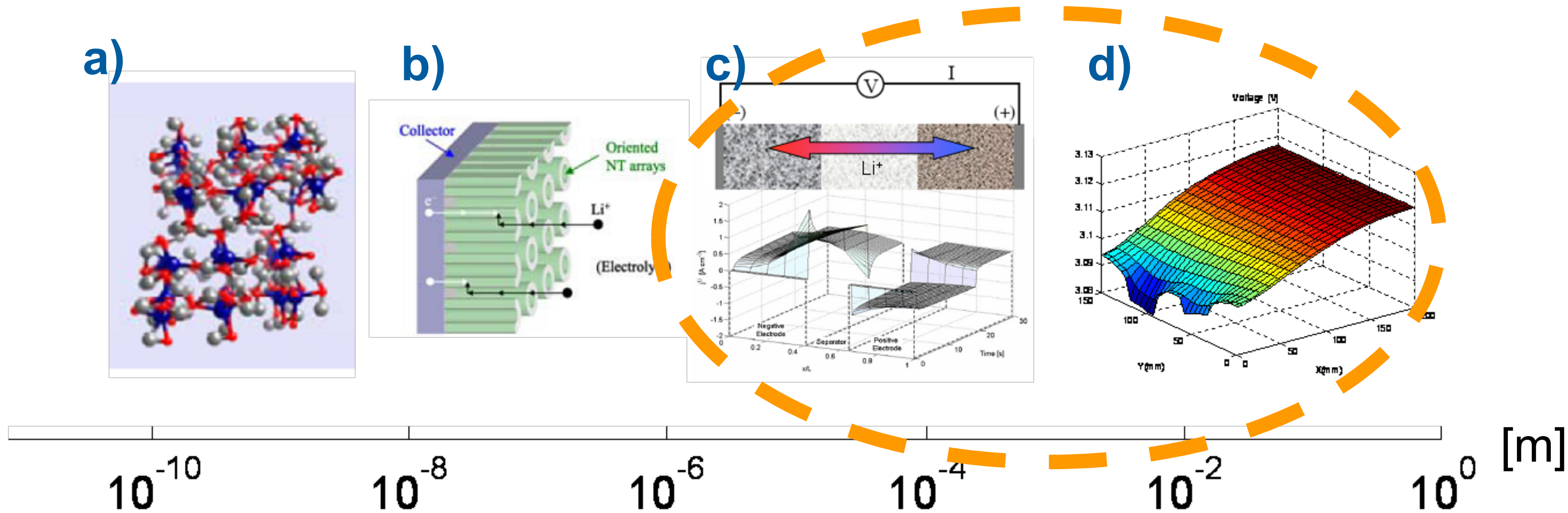
Electrodes selection
Li transport
Porosity, tortuosity
Layer thicknesses
Load conditions

Design of Electron Current & Heat Transport

Electric & thermal connections
Dimensions, form factor
Component shapes

Need a Multi-Scale Model?

Numerical approaches focusing on different length scale physics



a) Quantum mechanical and molecular dynamic modeling
b) Numerical modeling for addressing the impacts of architecture of electrode materials
c) 1D performance model capturing solid-state and electrolyte diffusion dynamics
d) Cell-dimension 3D model for evaluating macroscopic design factors

Why macro-scale transport becomes critical?

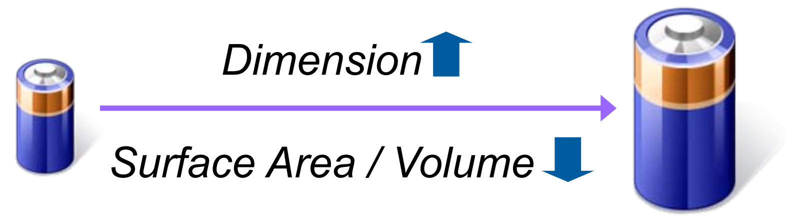
Sub-electrode scale physics
Kinetics
Li diffusion
Ion transport
Heat dissipation

Design of current and heat flow paths

Spatial variation of …

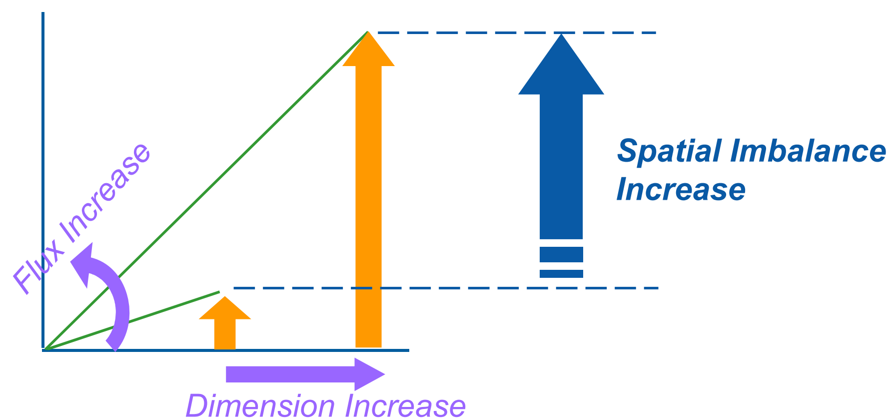
- Electric potentials
- Temperatures

Size Effect



Flux (gradient) ↑
Barrier (distance) ↑

Spatial Difference (gradient x distance) ↑↑



Approach in the Present Study

Multi-Scale Multi-Dimensional (MSMD) Modeling

To address ...

- Multi-scale physics from sub-micro-scale to battery-dimension-scales
- Difficulties in resolving microlayer structures in a computational grid

Simulation Domain **=** **Macro Grid** **+** **Micro Grid**
(Grid for Subgrid Model)

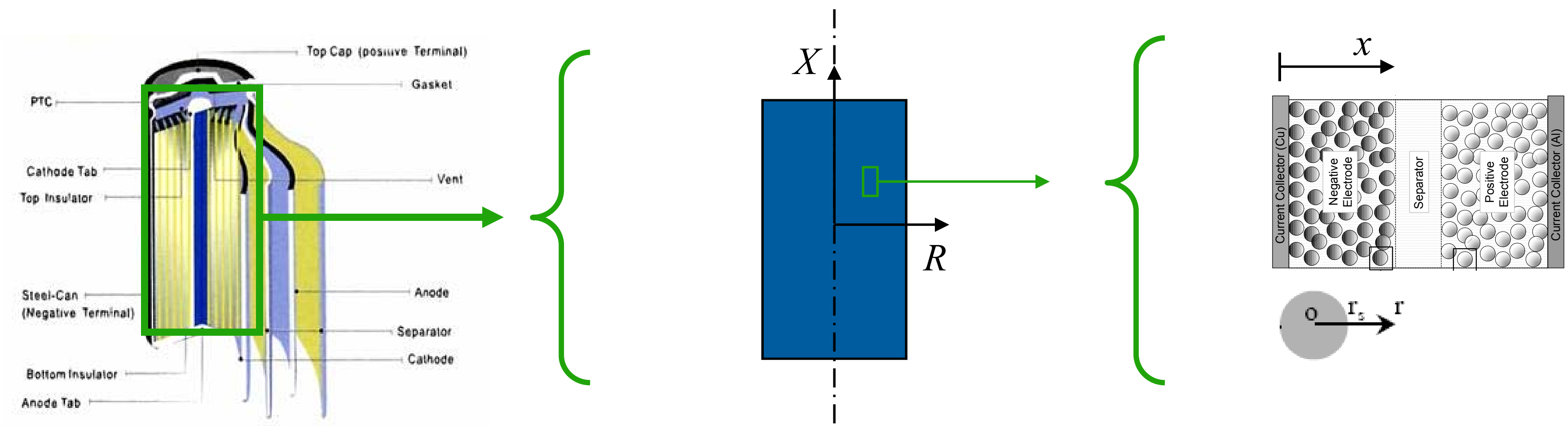


Image source: www.dimec.unisa.it

Solution Variables

Detailed Structure ≈ **Cell Dimension Transport Model** + **Electrode Scale Submodel (1D)**

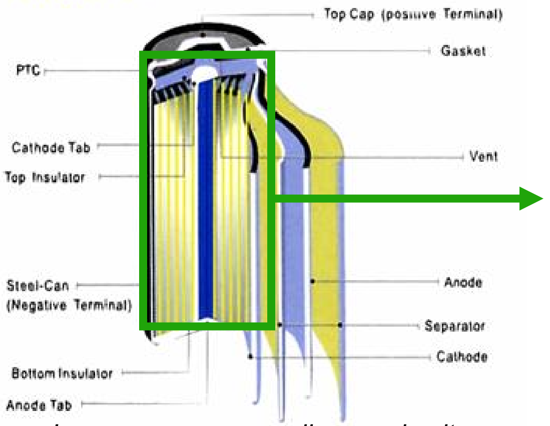
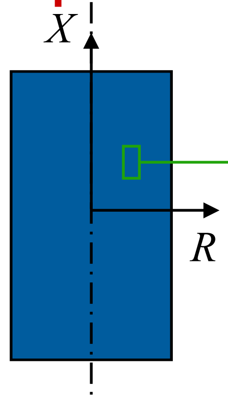
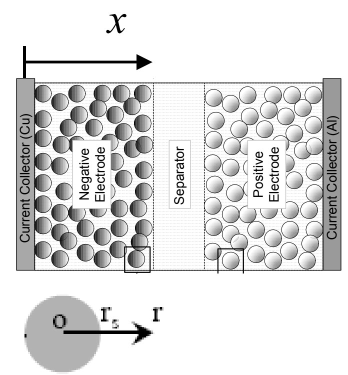


Image source: www.dimec.unisa.it





$$T(X,R,t)$$
$$V(X,R,t)$$
$$i(X,R,t)$$
$$SOC(X,R,t)$$
$$Q(X,R,t)=\int_x Q_i \frac{Adx}{V}$$

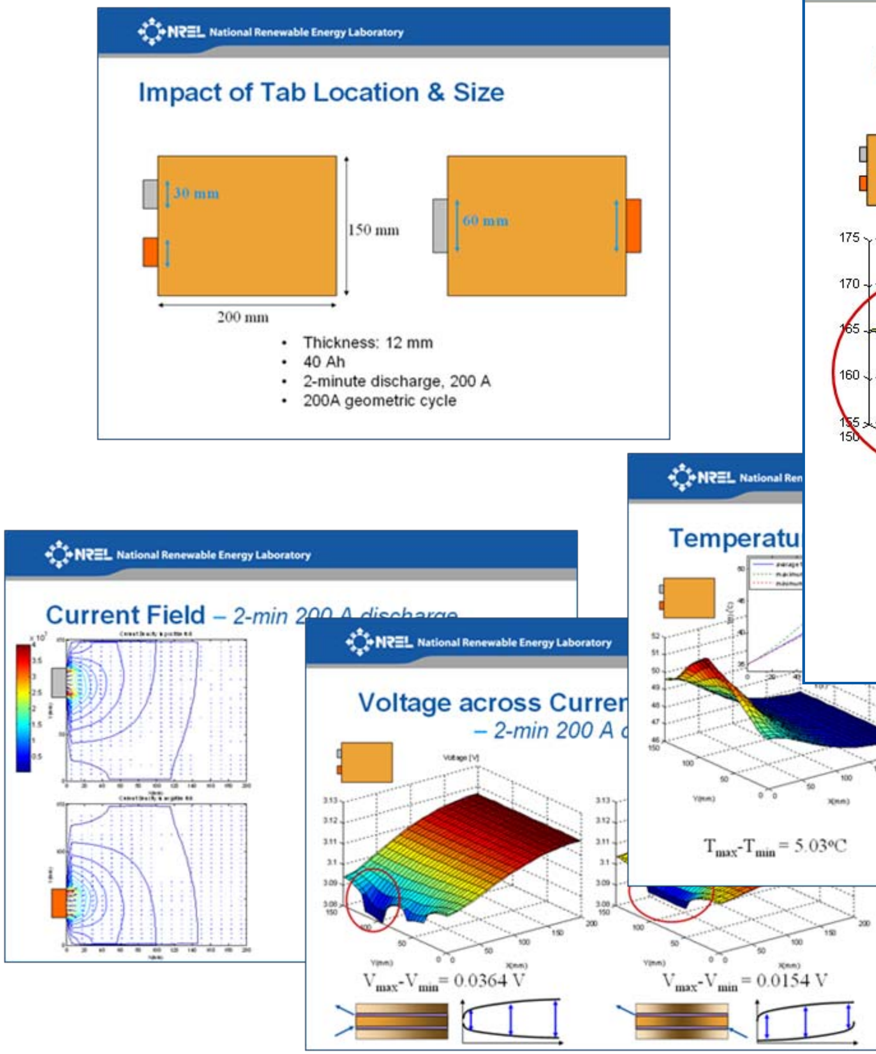
⟺

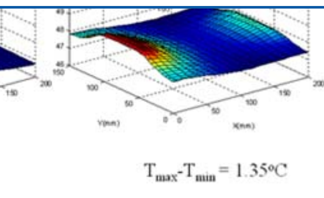
$$\phi_s(X,R,x,t)$$
$$\phi_e(X,R,x,t)$$
$$c_s(X,R,x,r,t)$$
$$c_e(X,R,x,t)$$
$$j_{Li}(X,R,x,t)$$
$$Q_i(X,R,x,t)$$

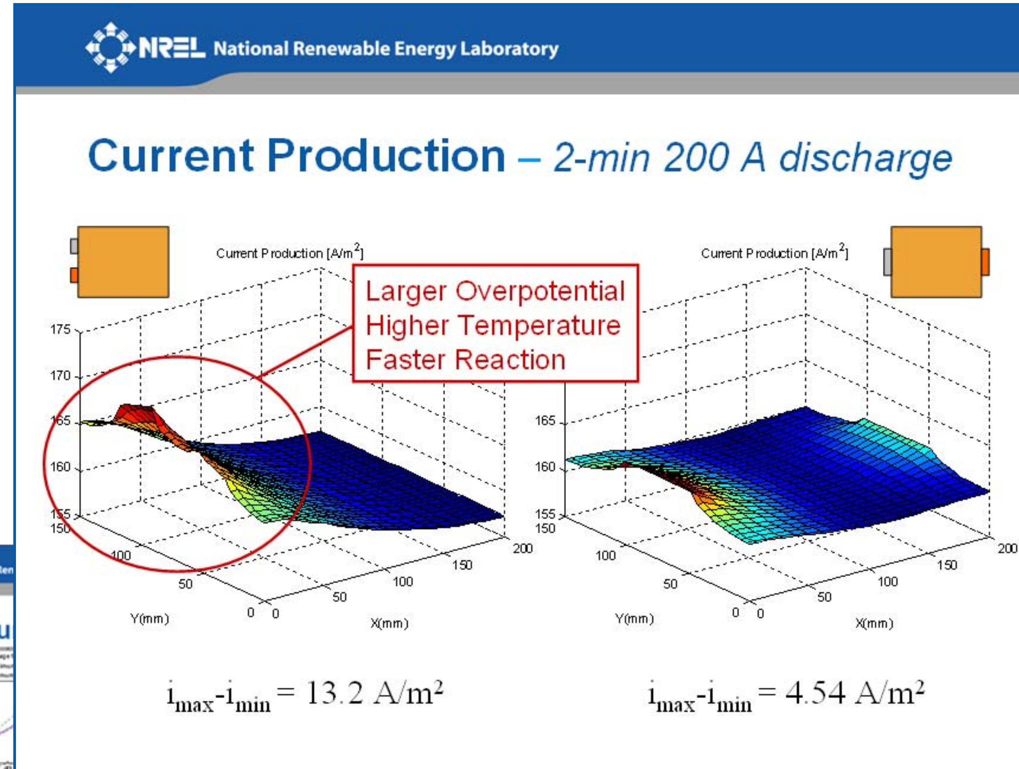
NOTE:
Selection of solution scheme for either grid system is independent of the other.

Previous Study

AABC 08, Tampa, May 2008







"Poorly designed electron and heat transport paths can cause excessive nonuniform use of materials and then deteriorate the performance and shorten the life of the battery."

Analysis

Comparison with Experimental Results

- *Model Validation against JCS VL41M Test Data*

Macro-Scale Design Evaluation Analysis

- *Impacts of Aspect Ratio of a Cylindrical Cell*

Comparison with Experimental Results

- *Model Validation against JCS VL41M Test Data*

The JCS VL41M cell was chosen as a candidate for several reasons:

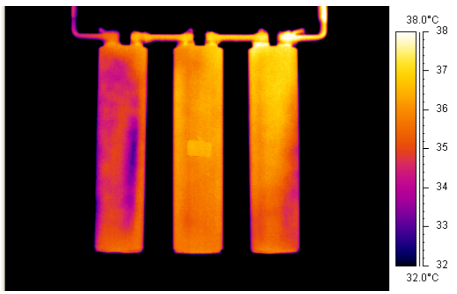
- *1-D electrochemical model was previously validated vs. VL41M current/voltage data.*
- *Thermal imaging experiments were recently run.*
- *Future calorimeter test data will allow for further refinement & validation of the model.*

Macro-Scale Design Evaluation Analysis

- *Impacts of Aspect Ratio of a Cylindrical Cell*

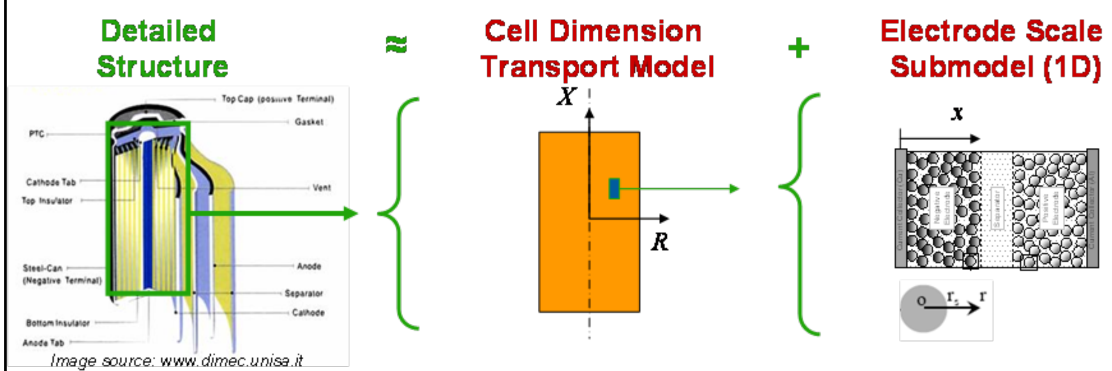
Approach

JCS VL41M Thermal Imaging Test



Data used for model validation

Multi-Scale Multi-Dimensional (MSMD) Model

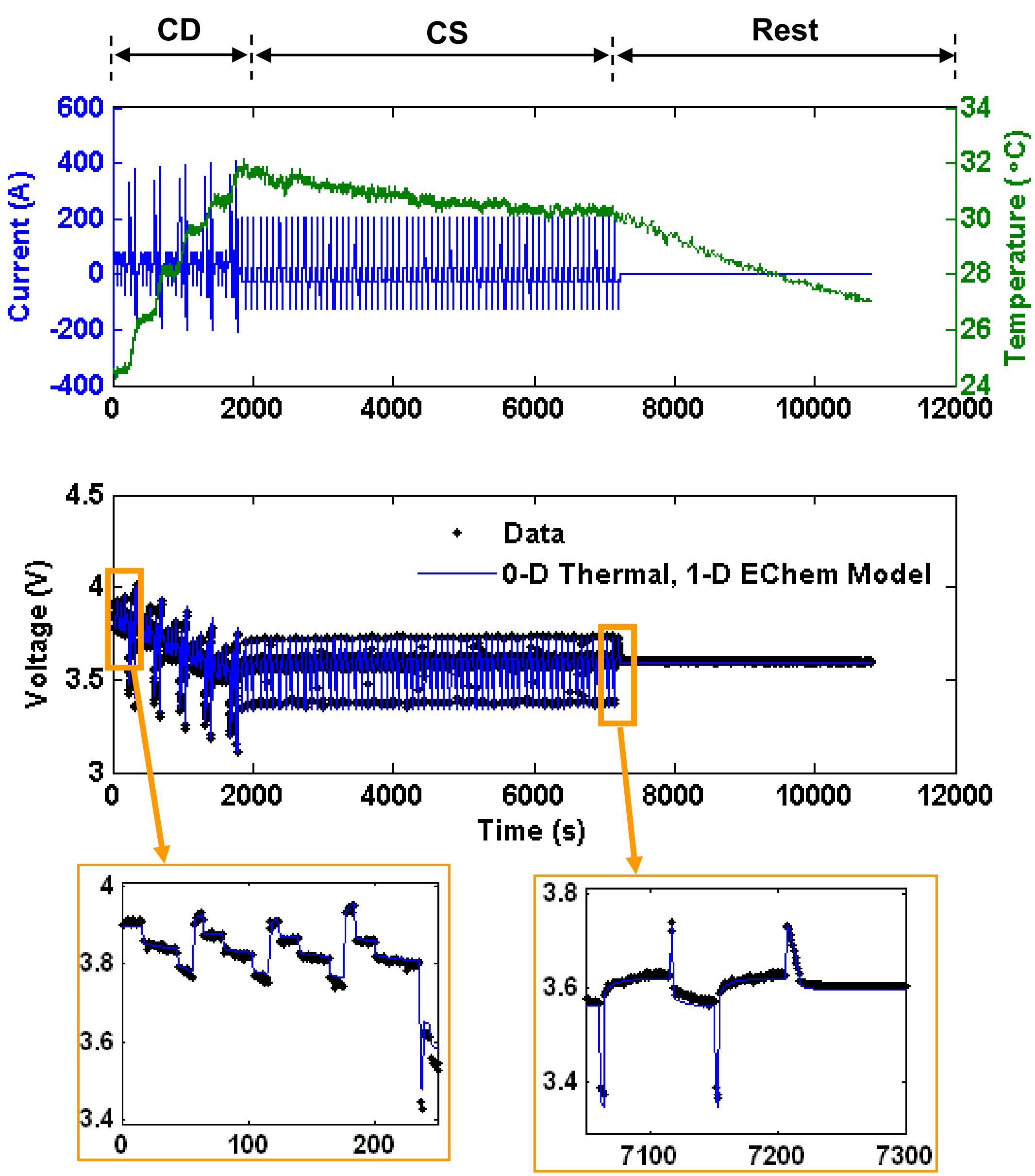


1) 1-D Electrochemical Model Validation
   - Measured current & temperature profiles used as inputs to model
   - Model predicts voltage & heat generation rate

2) Multi-Scale Multi-Dimensional ("MSMD") Model Validation
   - Utilized 3D thermal model results to extract thermal boundary conditions
   - Measured surface temperature compared to model prediction of jelly-roll surface temperature.

3) MSMD Model Predictions
   - Multidimensional features

1) 1D Electrochemical Model Validation



Measured current and skin temperature* profiles from thermal imaging test used as inputs to lumped thermal/1-D electrochemical model.

Model voltage prediction compares favorably with data.

- Error generally < 50 mV

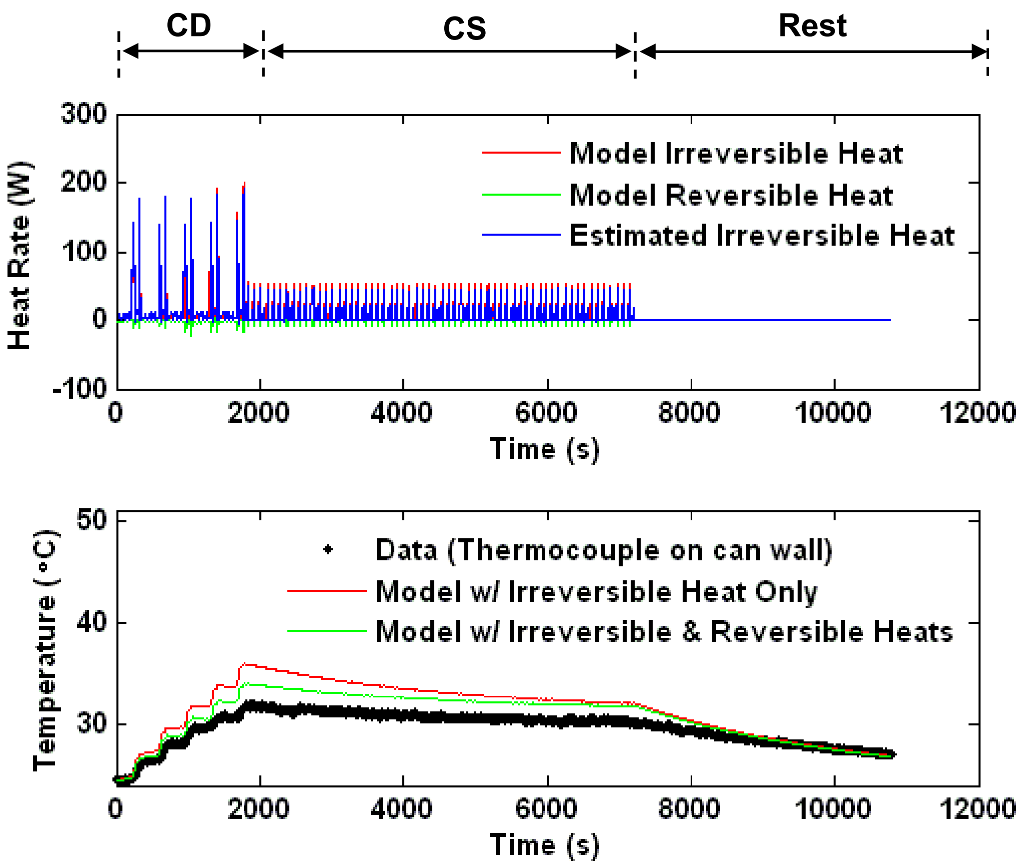
Test Profile:
5 charge-depletion cycles + 60 charge-sustaining cycles per USABC manual (BSF = 39)

* Skin temperature measured via thermocouple on can wall, 3" from bottom.

1) 1D Electrochemical Model Validation

Test Profile:

5 charge depletion cycles + 60 charge sustaining cycles per USABC manual (BSF = 39)



Irreversible heat generation rate predicted by 1-D electrochemical model compares well with calculated value using measured current and voltage and model open-circuit voltage.

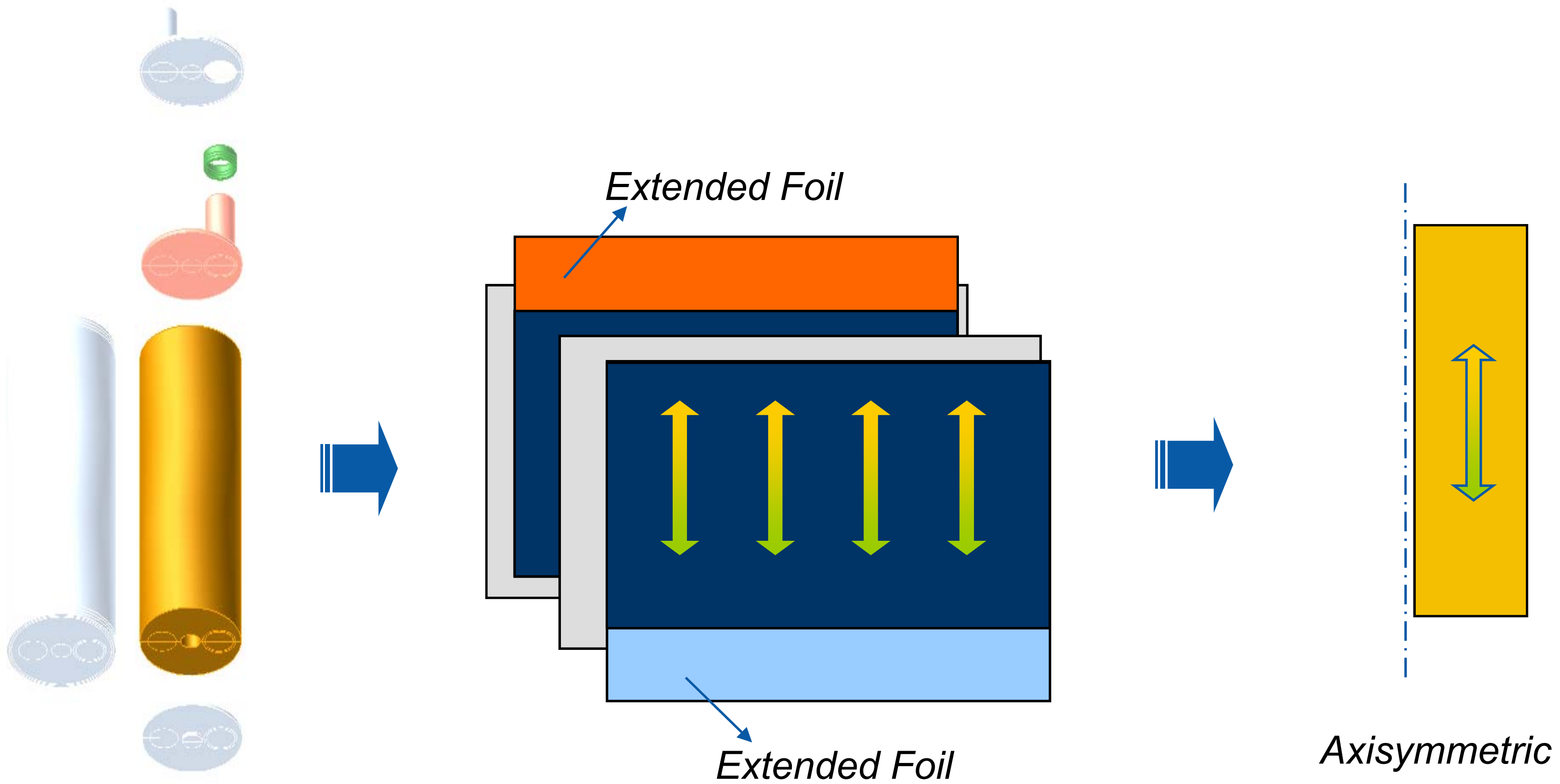
$$Q_{irr} = I_{meas}(OCP_{model} - V_{meas})$$

- Entropic heat effects seem to be non-negligible and may need to be included in the model.

* More rigorous heating rates and specific heat to be measured in upcoming calorimeter testing.

2) MSMD Model Validation

Assumption for Model Simplification

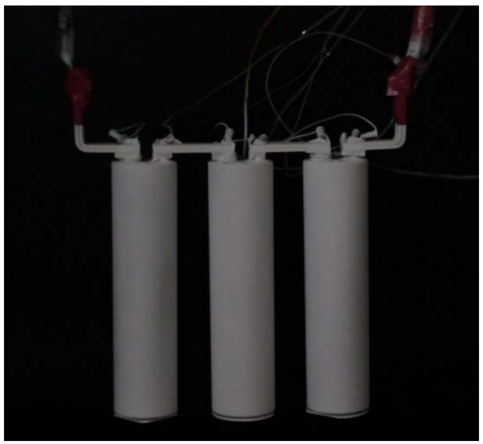


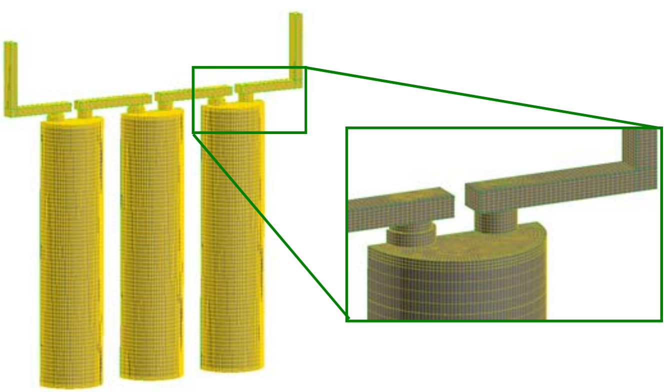
*Note: The schematics shown above do **not** represent actual* JCS VL41M.

2) MSDM Model Validation

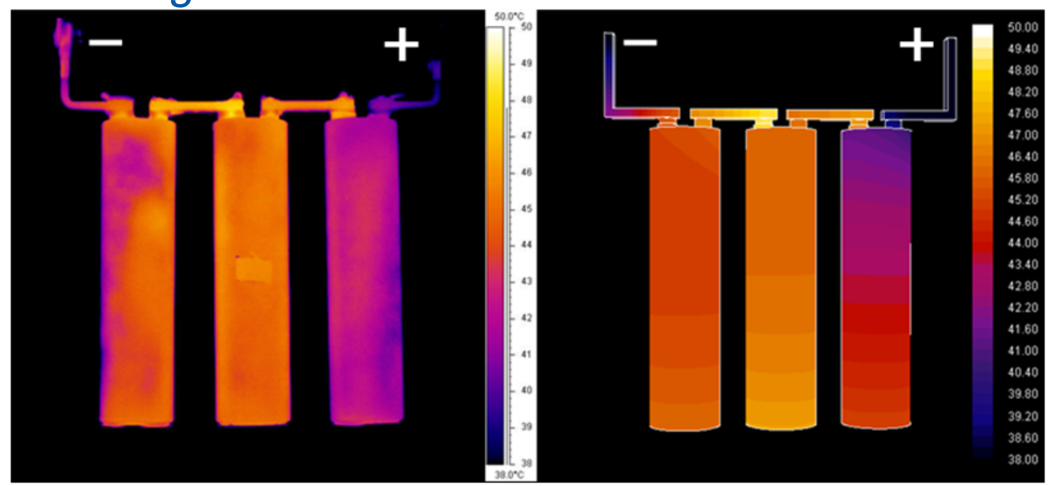
Retrieving information from 3D Thermal Model for MSMD model input

• Complex thermal pathway was captured in 3D thermal model, then appropriate thermal boundary condition was evaluated for MSMD model





IR Image *Model*

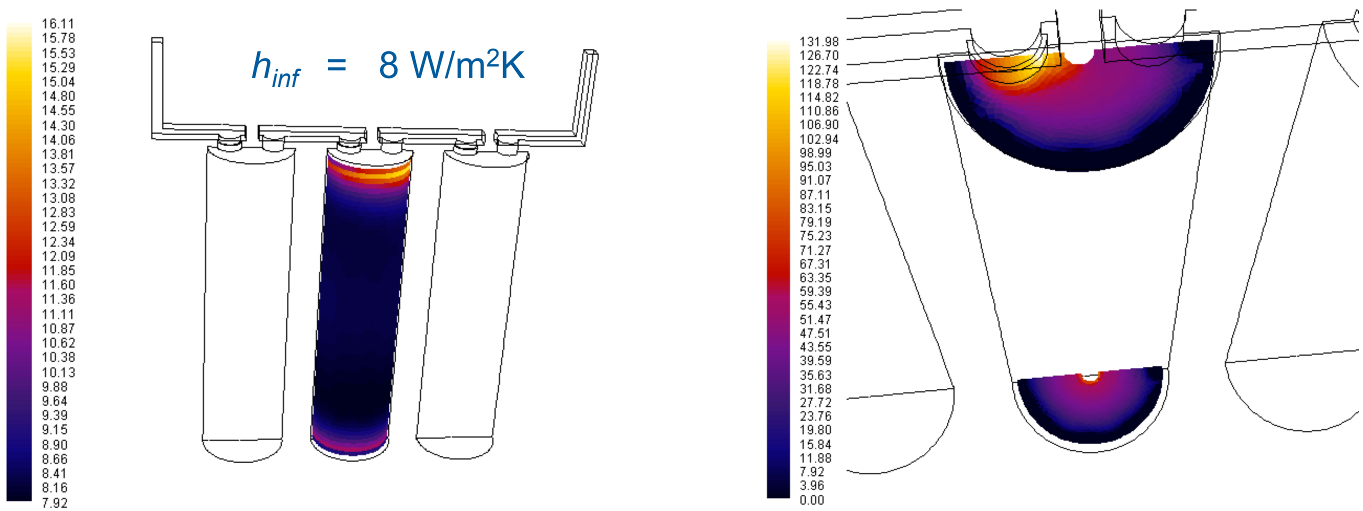


100 A Geometric Cycle - Steady

- General system response for temperature distributions at cell skins, terminals and bus bars is well predicted and reveals how heat is transferred through the 3 cell assembly.

2) MSMD Model Validation

Evaluating thermal boundary conditions at jelly-roll surfaces



Heat transfer coefficient at jelly-roll surfaces of the middle cell

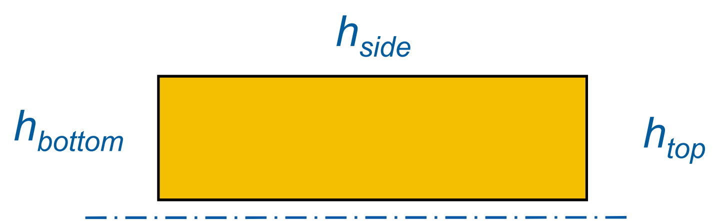
Area Weighted Averages

h_{top} = 22.6 W/m^{2}K

h_{side} = 8.7 W/m^{2}K

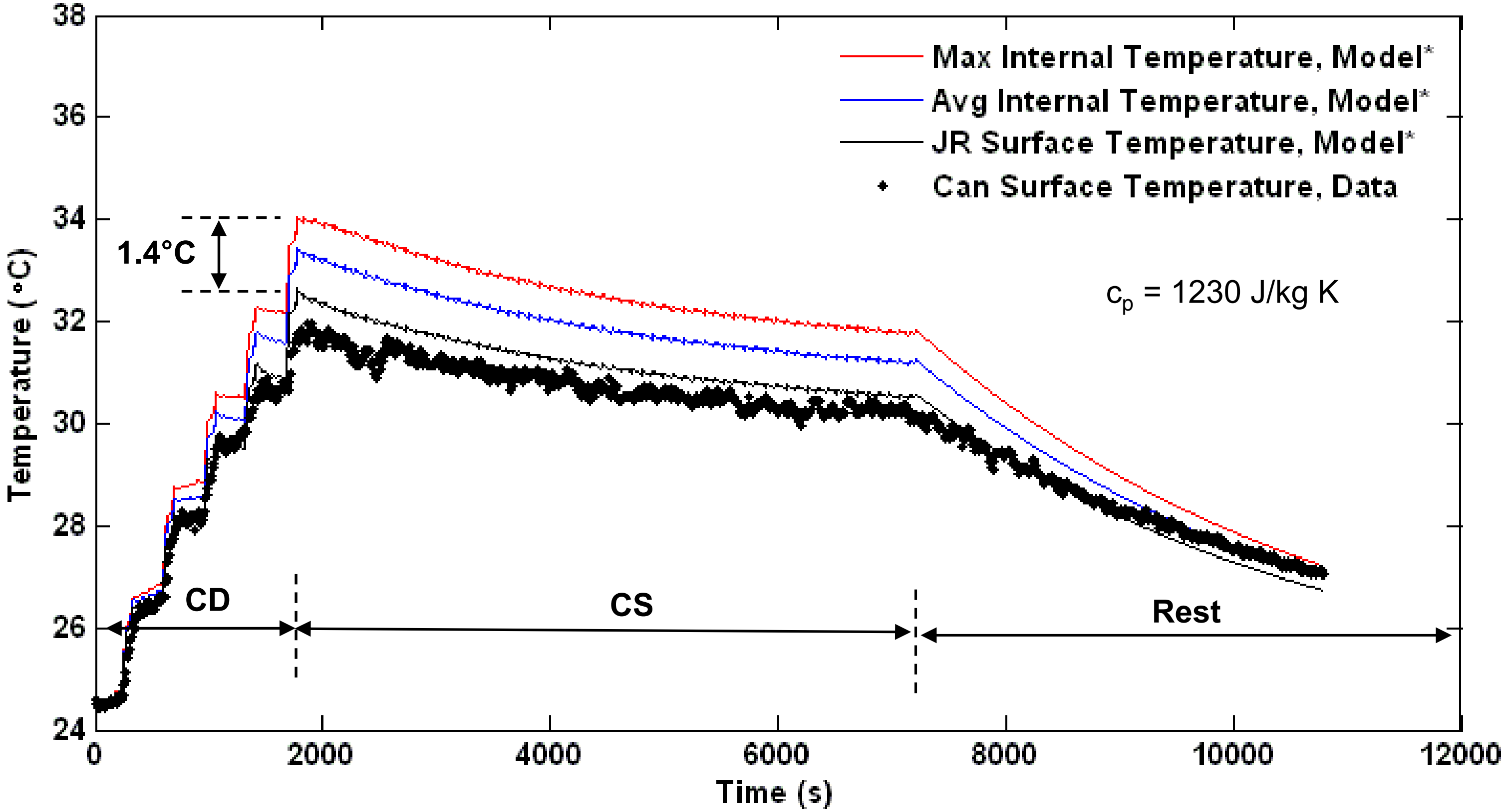
h_{bottom} = 12.4 W/m^{2}K

Axisymmetric MSMD Model



2) MSMD Model Validation

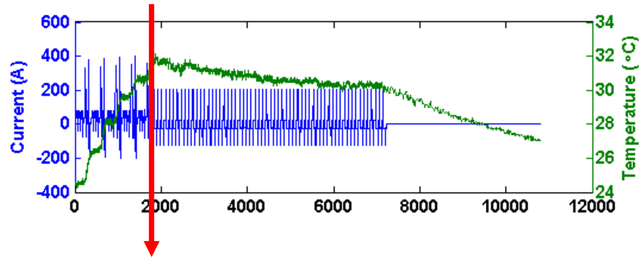
Comparison with Measured Temperature



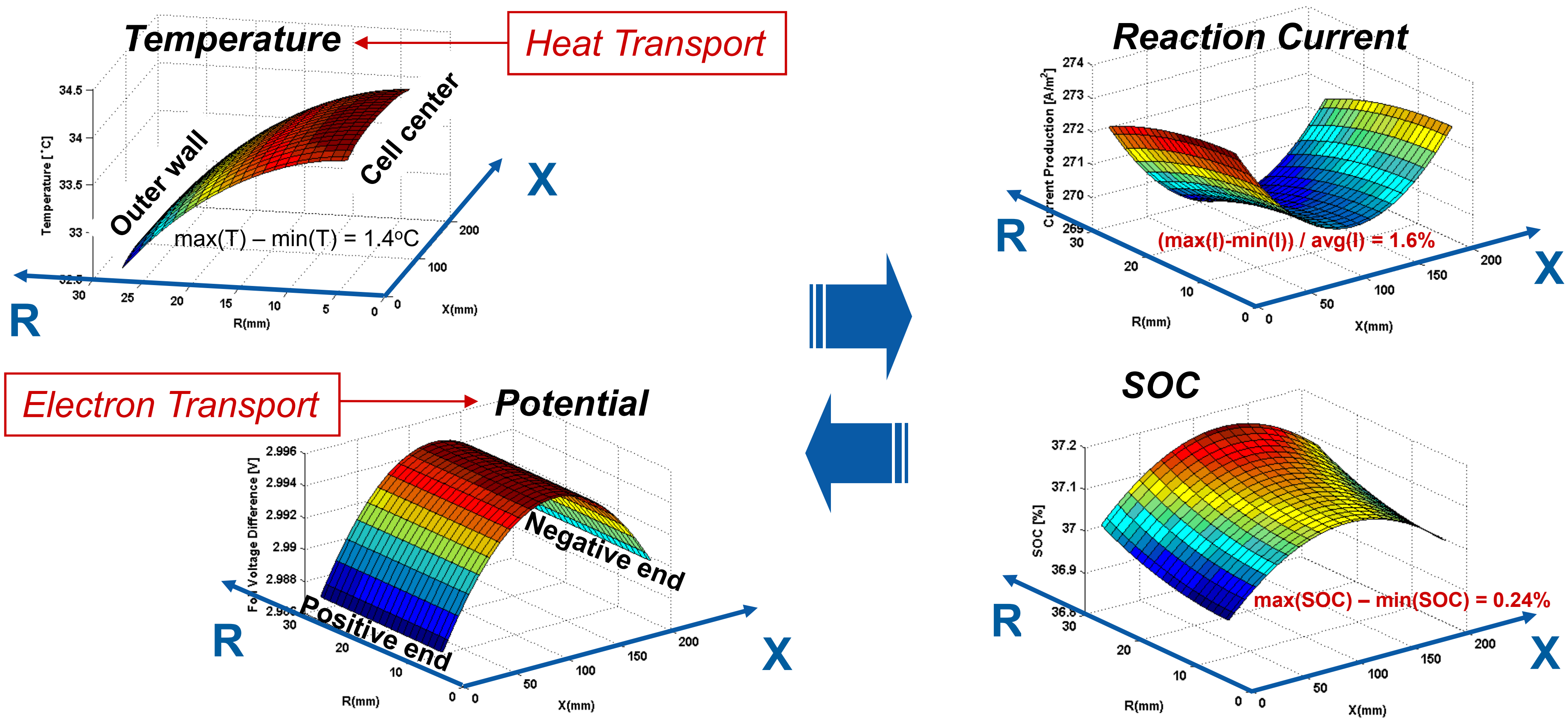
Measured can surface temperature and model-predicted jelly-roll temperature agree reasonably well. Without an internally-instrumented cell, it is not possible to directly validate the MSMD model's jelly-roll temperature predictions.

3) MSMD Model Prediction

Snapshots at the end of CHARGE DEPLETING cycles

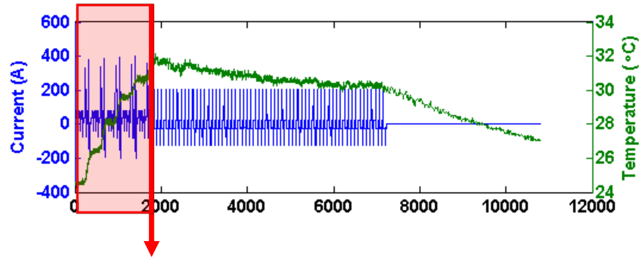


- t = 1770 s
- $T_{can\ wall}$ = 31.6°C
- Current = 409 A



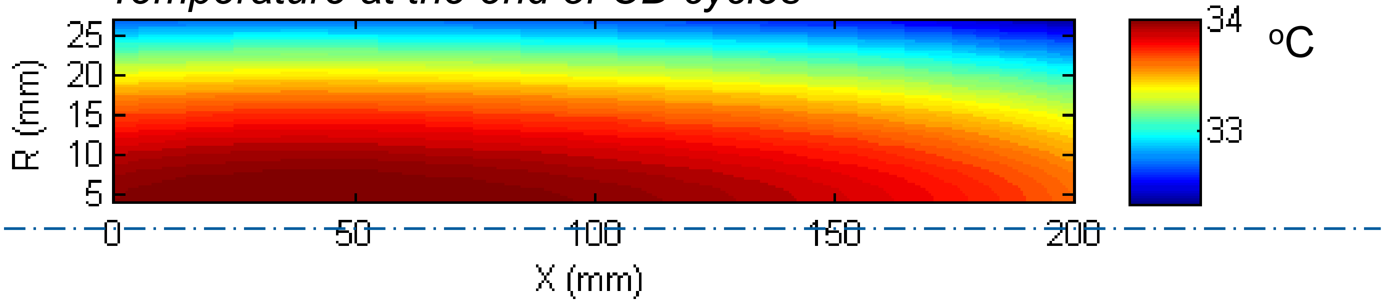
3) MSMD Model Prediction

Ah-throughput during CHARGE DEPLETING cycles

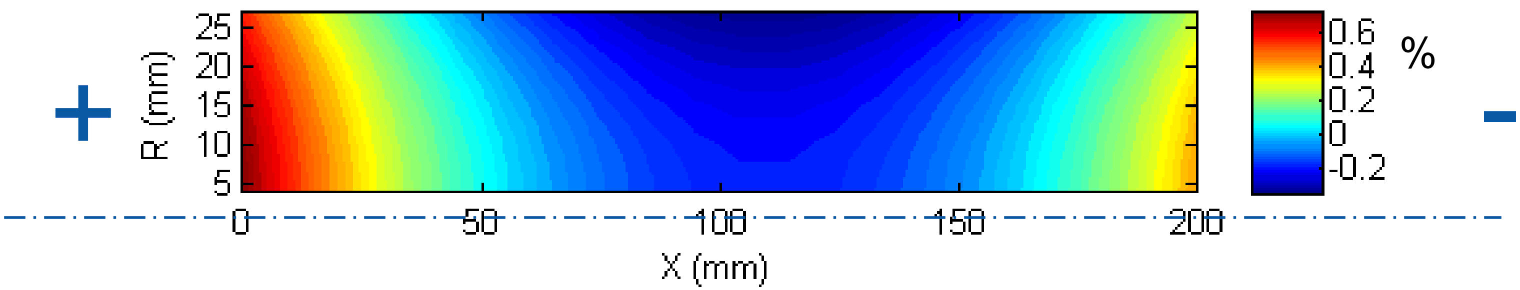


- t = 1770 s
- $T_{can\ wall}$ = 31.6°C
- Current = 409 A

Temperature at the end of CD cycles



Ah-throughput imbalance during CD cycling $(Ah/m^2 – Ah/m^2_{avg})/Ah/m^2_{avg}$



Analysis

Comparison with Experimental Results

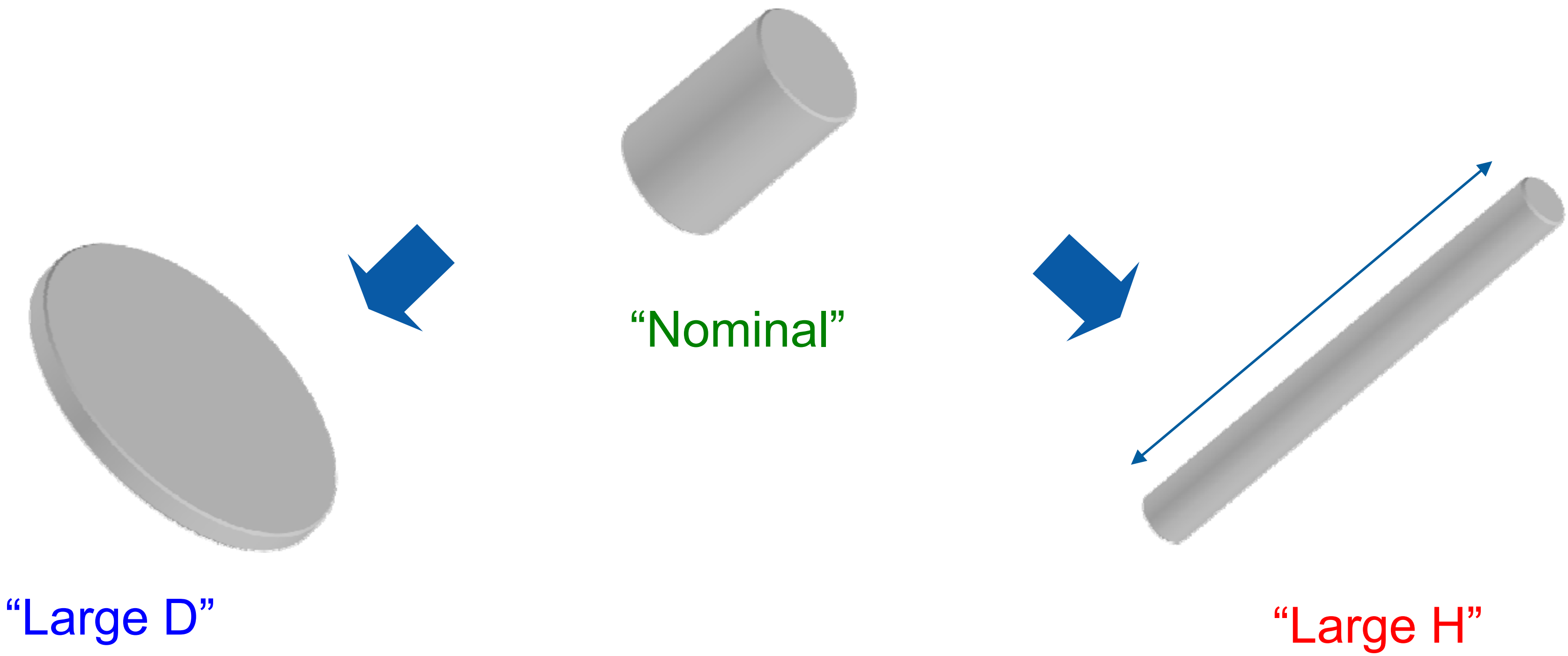
- *Model Validation against JCS VL41M Test Data*

The JCS VL41M cell was chosen as a candidate for several reasons:
- *1-D electrochemical model was previously validated vs. VL41M current/voltage data.*
- *Thermal imaging experiments were recently run.*
- *Future calorimeter test data will allow further refinement and validation of the model.*

Macro-Scale Design Evaluation Analysis

- *Impacts of Aspect Ratio of a Cylindrical Cell*

Aspect Ratio of Cylindrical Cells



PHEV10 application

- US06 cycle discharges 3.4 kWh in 12 minutes (~3C rate)

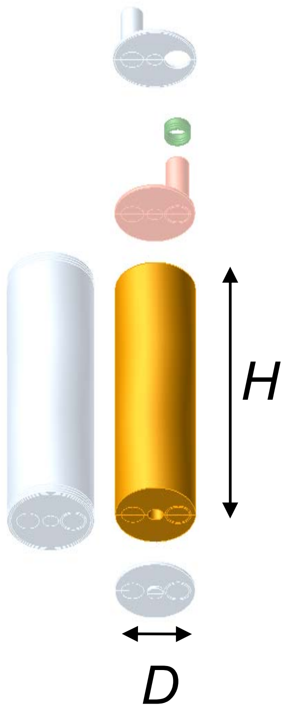
20 Ah cell

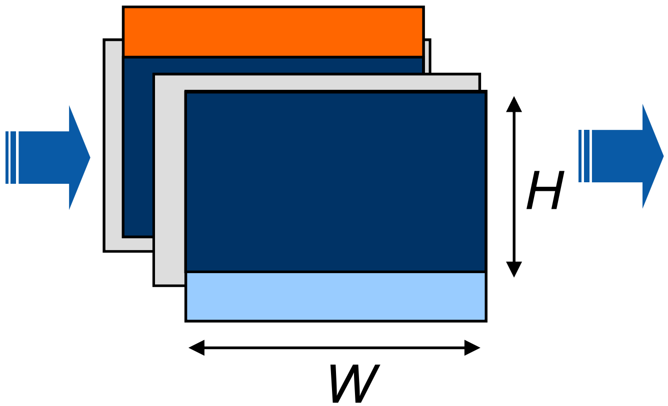
- Well suited for PHEV10
- BSF = 78 → V_{nom} ≈ 290V

US06 CD cycle

- P_{avg} = 14 kW, P_{RMS} = 32 kW

Brief Look at "What H/D Ratio Means"

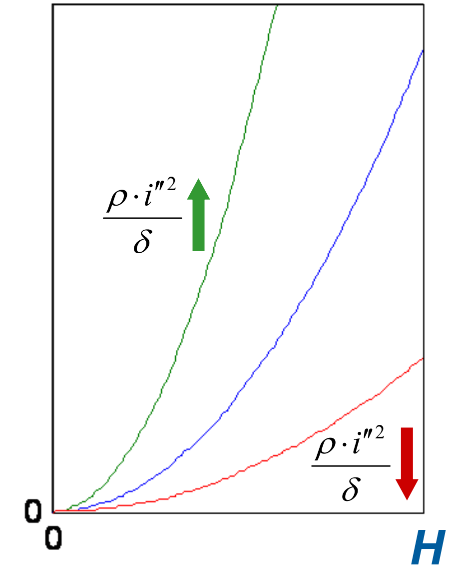




$$P_{loss,foil} \sim \frac{\rho \cdot i''^2}{\delta} H^2$$

$$\Delta V_{foil} \sim \frac{\rho \cdot i''}{\delta} H^2$$

i“: current [A/m²]
ρ: resistivity
δ: foil thickness

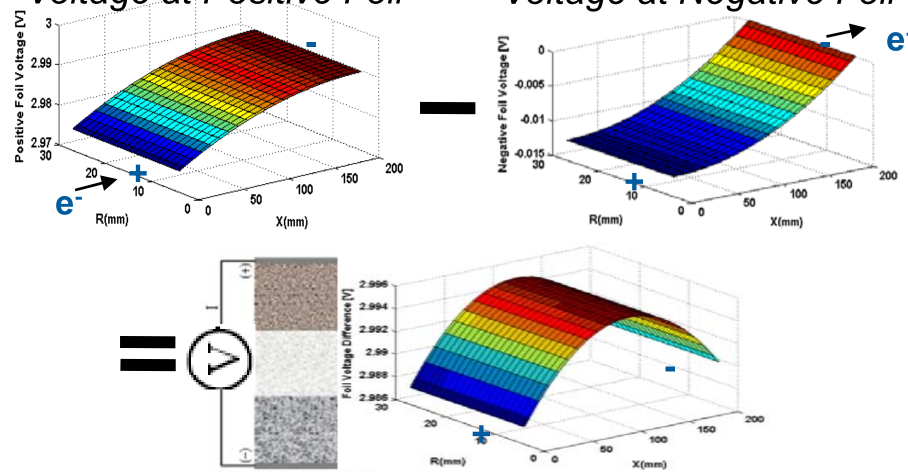


Volume = const

H x W = const

Foil thicknesses
Al: 20 µm
Cu: 15 µm

Voltage at Positive Foil — *Voltage at Negative Foil*

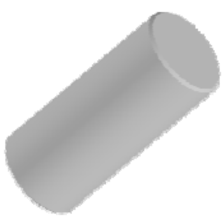


10s Power Capability Comparison

Large H

D[mm]: 28
H[mm]: 350

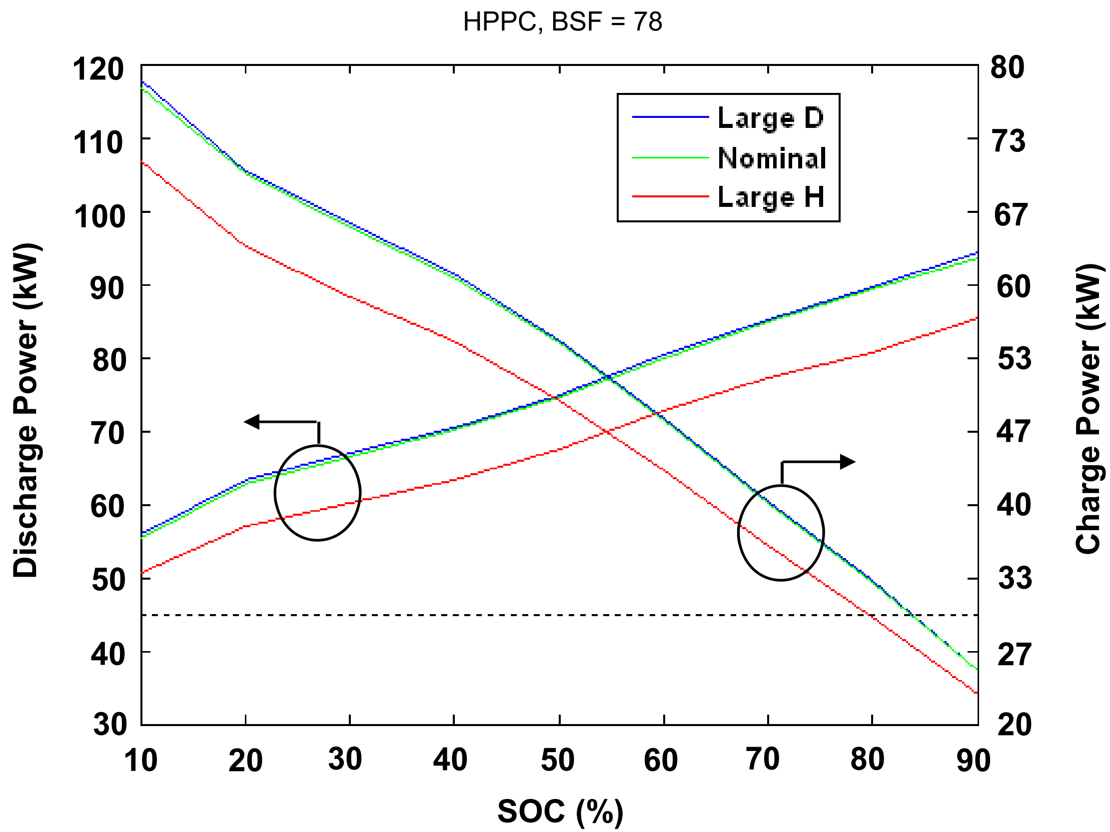
Nominal



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

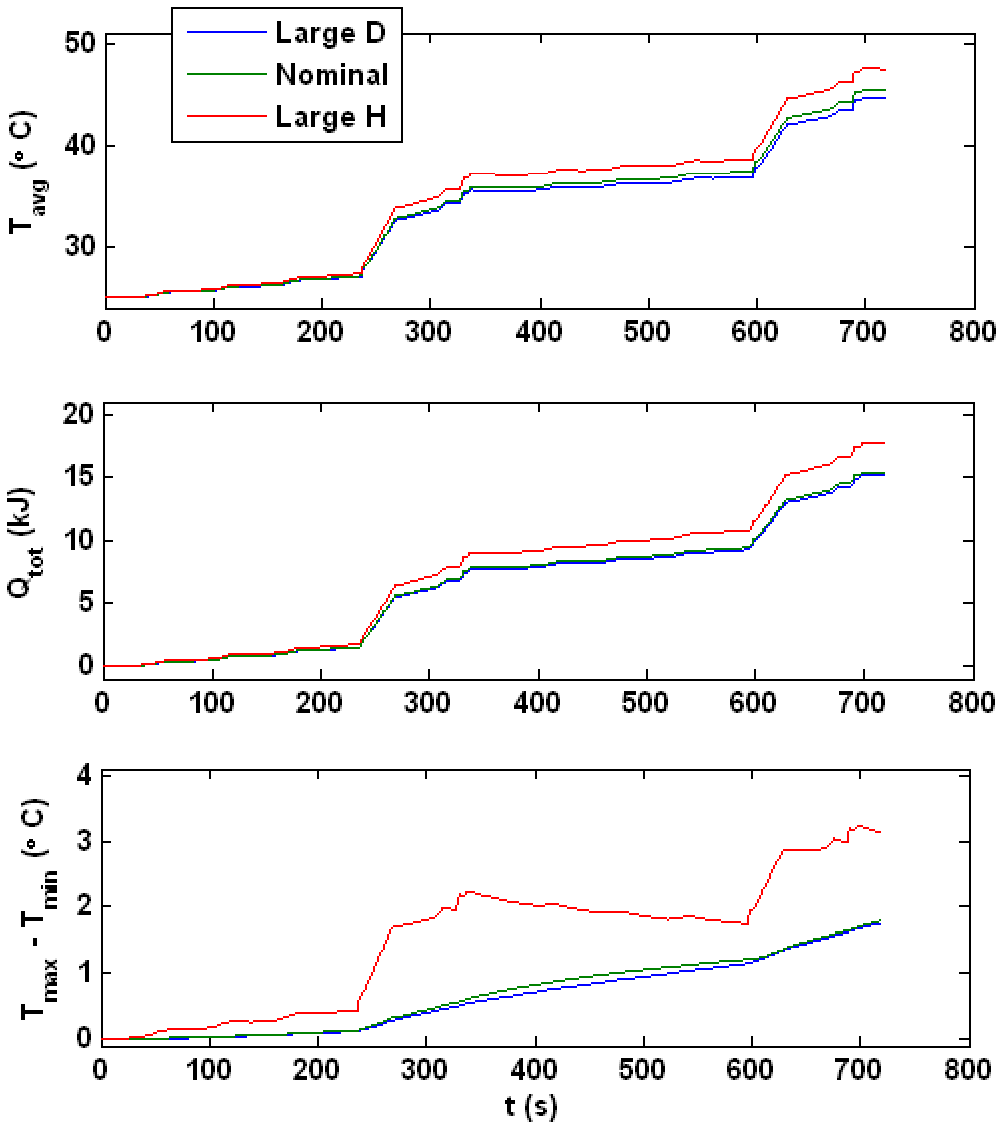
Large D

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



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

US06 CD Cycle x 2, Natural Convection



Large H cell has greatest temperature rise owing to long electronic current paths resulting in high foil heating.

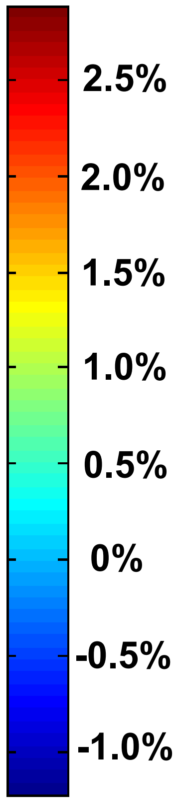
Foil heat contribution to total:

- 15% - Large H
- 1.7% - Nominal
- <0.1% - Large D

Large H cell has greatest internal temperature imbalance.

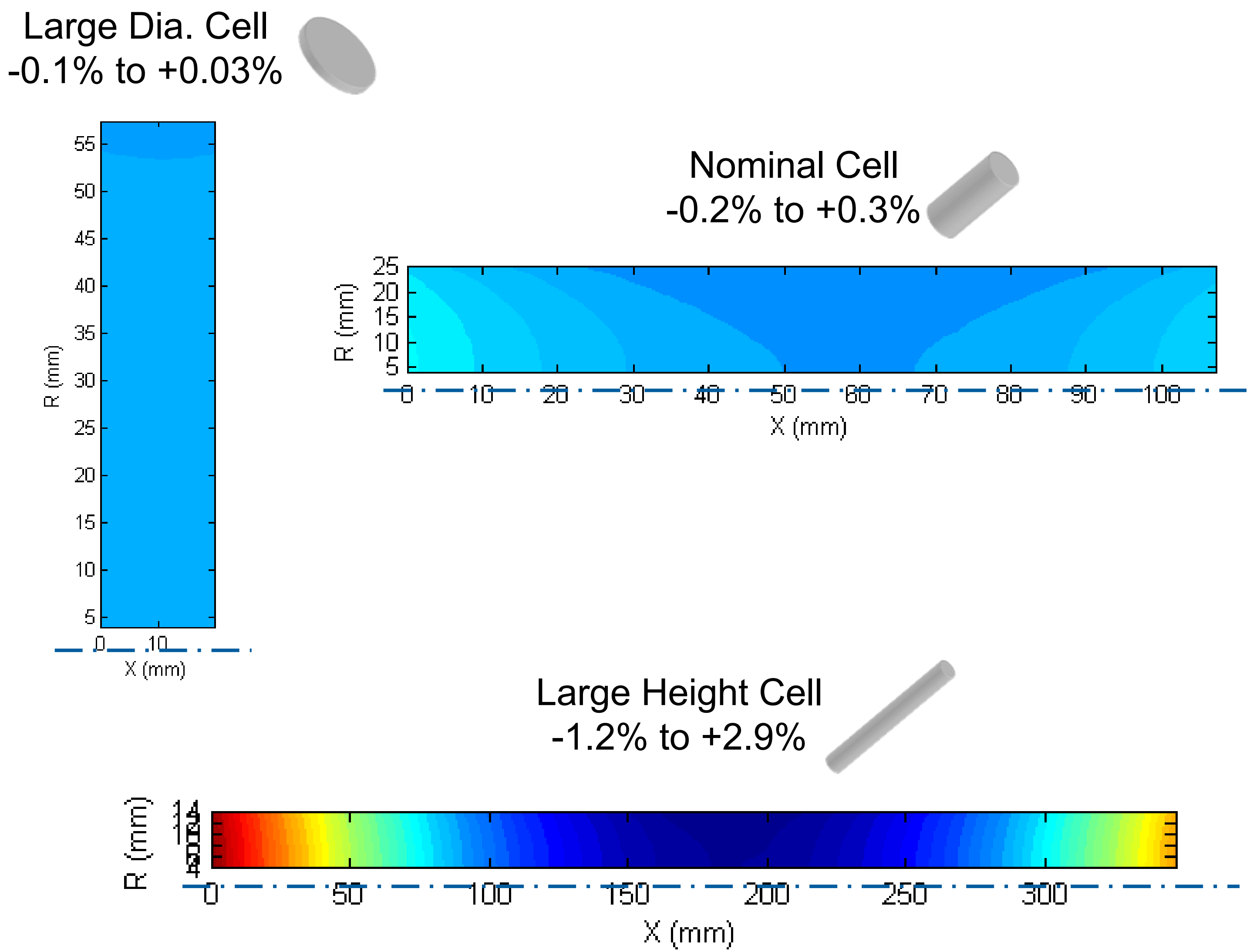
US06 CD Cycle x 2, Natural Convection

Amp-hour Throughput Imbalance



US06 CD cycle

$(Ah/m^2 - Ah/m^2_{avg})/Ah/m^2_{avg}$



Summary

- Nonuniform battery physics, which is more probable in large-format cells, can cause unexpected performance and life degradations in lithium-ion batteries.
- A Multi-Scale Multi-Dimensional model was developed as a tool for investigating interaction between micro-scale electrochemical process and macro-scale transports using a multi-scale modeling scheme.
- The developed model will be used to provide better understanding and help answer engineering questions about improving *cell design*, *cell operational strategy*, *cell management*, and *cell safety*.

- Engineering questions to be addressed in *future works* include …

What is the optimum form-factor and size of a cell?
Where are good locations for tabs or current collectors?
How different are measured parameters from their nonmeasurable internal values?
Where is the effective place for cooling? What should the heat-rejection rate be?
How does the design of thermal and electrical paths impact under current-related safety events, such as internal/external short and overcharge?

Acknowledgments

Vehicle Technologies Program at DOE

- Tien Duong
- Dave Howell

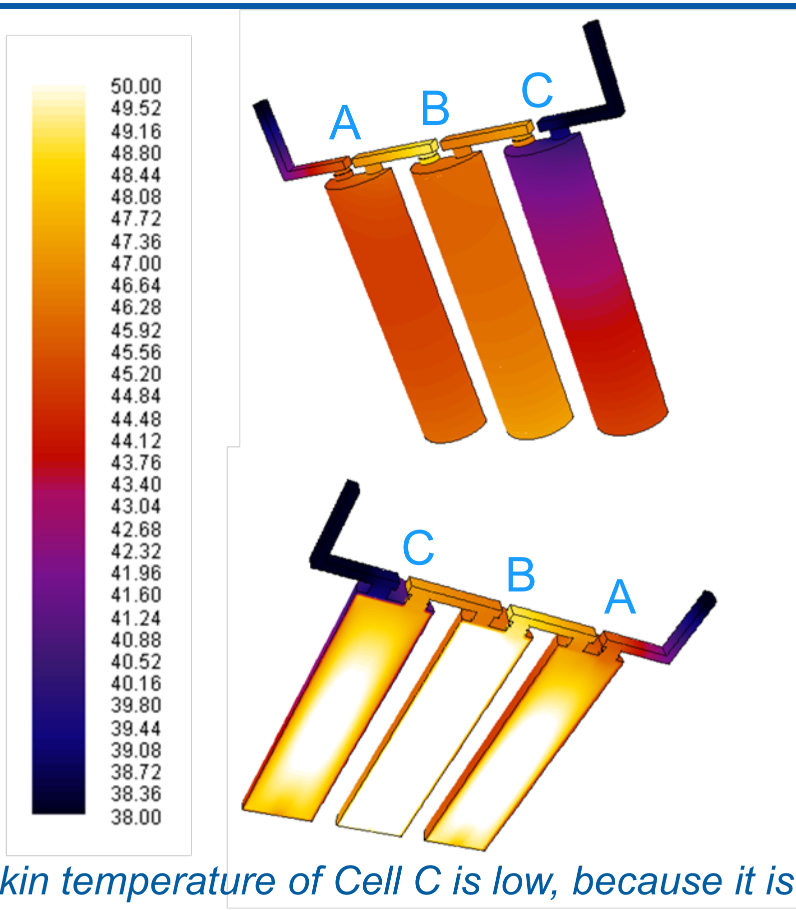
NREL Energy Storage Task

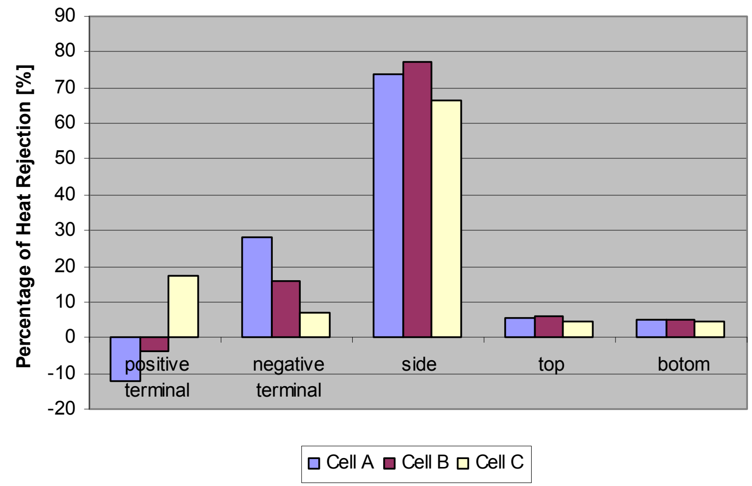
- Ahmad Pesaran

Thank you!

Additional Slides

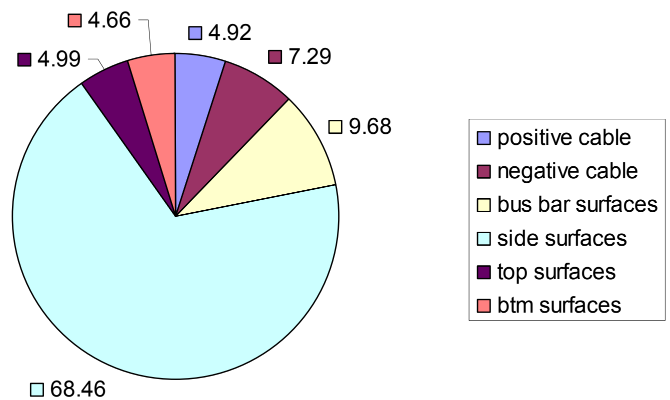
Heat Transfer – 100 A Geometric Cycle





Percentage of Heat Rejection from Each Cell

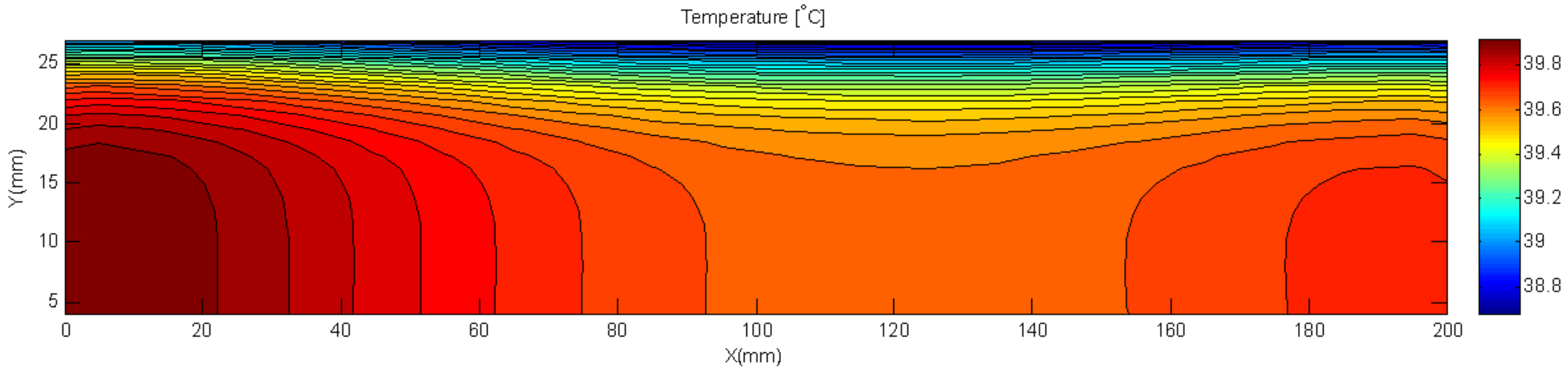
- *Skin temperature of Cell C is low, because it is directly connected to the cable through the positive terminal.*
- *There are inflows of heat through the positive thermals at Cell A and Cell B which are connected to the negative terminals of the neighbor cells.*
- *Most heat is rejected through cell side surfaces. About 10% of heat is dissipated at bus bar surfaces. 12% runs away through cables.*



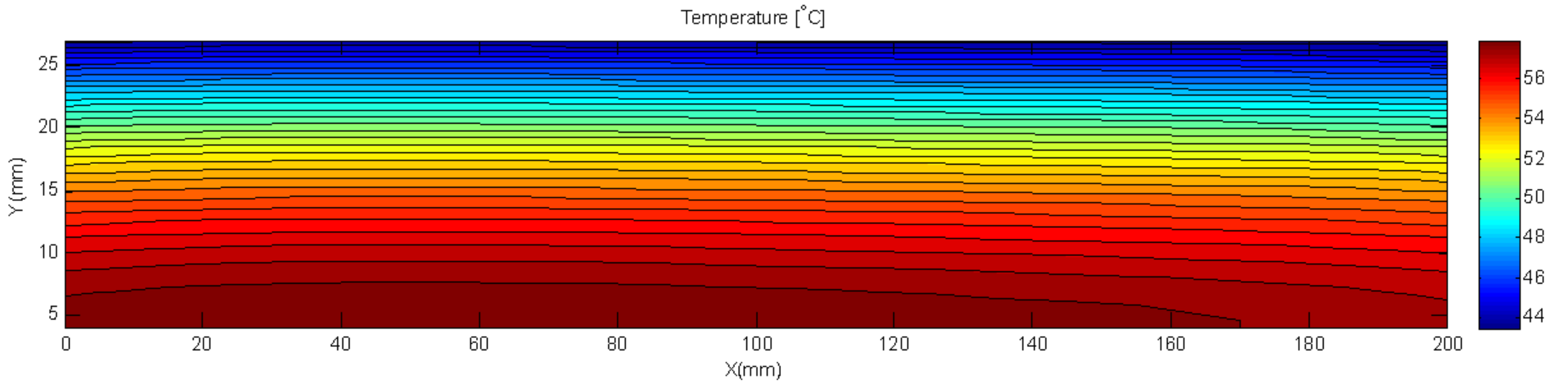
Percentage of Heat Rejection from Assembly

3) MSMD Model Prediction

Temperature Distribution after 30 sec 300 A discharge

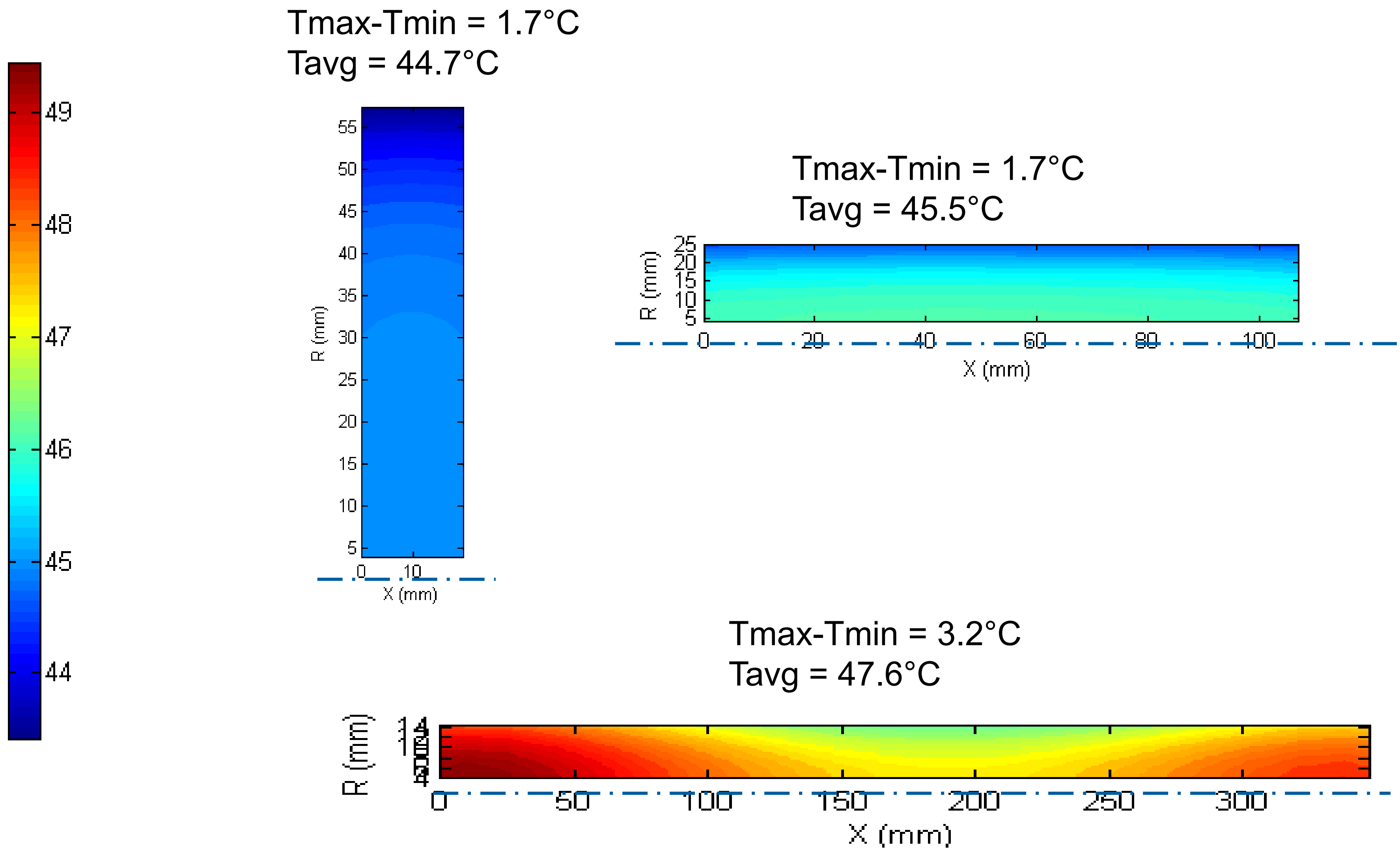


Temperature Distribution after 20 min 100 A geometric cycling

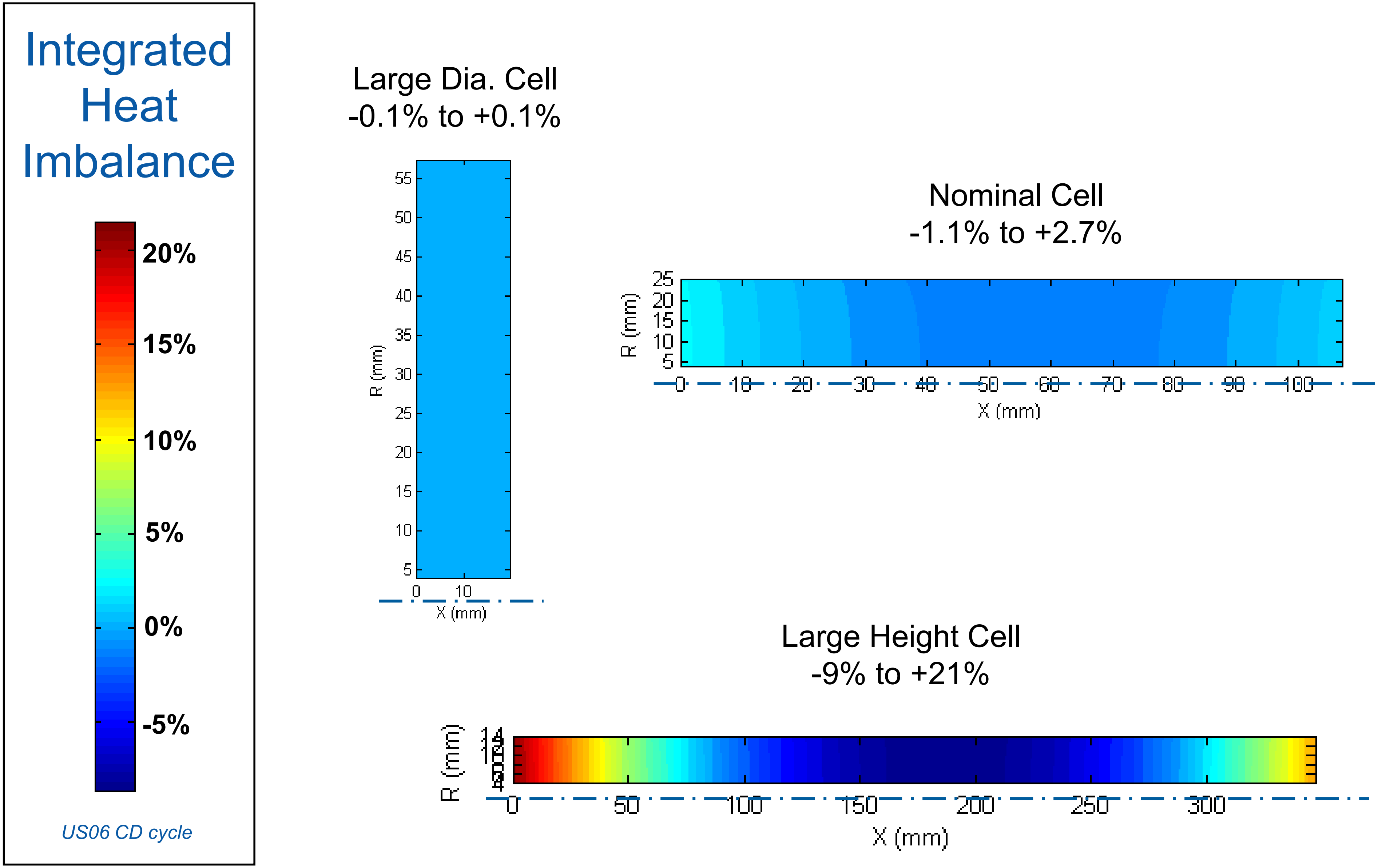


US06 CD Cycle x 2, Natural Convection

Temperature Distribution



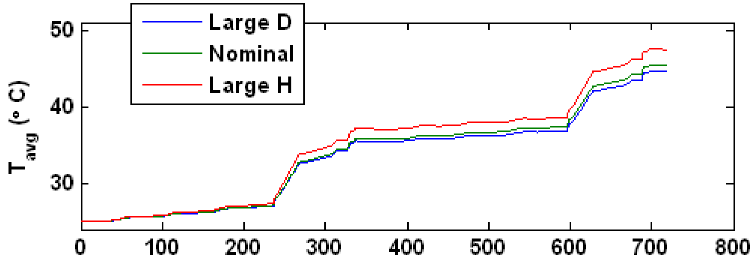
US06 CD Cycle, Natural Convection

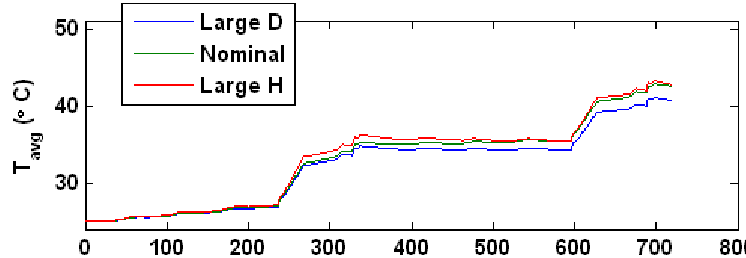


Forced Convection

Natural Convection
(h = 8 W/m² K)

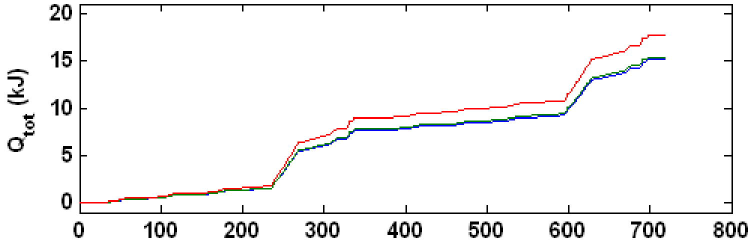
Forced Air Convection
(h = 30 W/m² K)

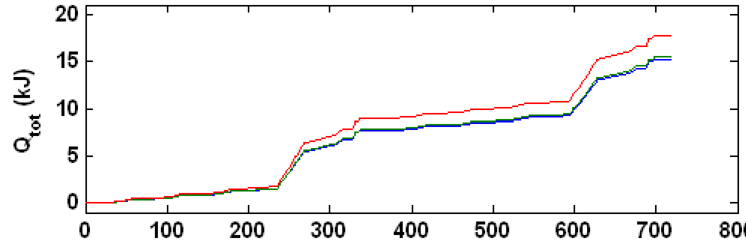




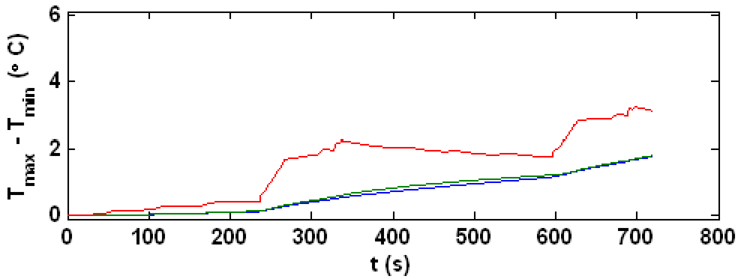
Average temperature lower

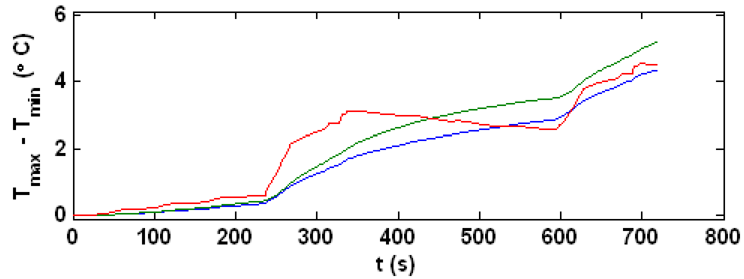
~ 5°C in Large H and Large D format, ~2°C in Nominal





Heat generation similar

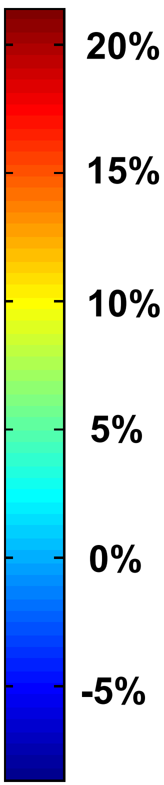




Temperature imbalance 1-3°C higher

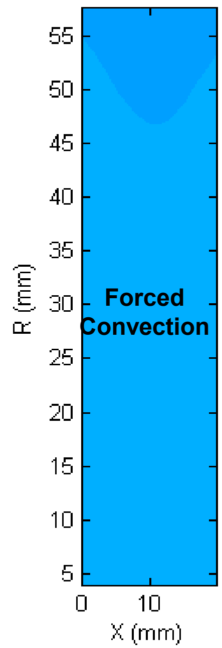
Forced convection – negligible impact on where heat is generated

Integrated Heat Imbalance

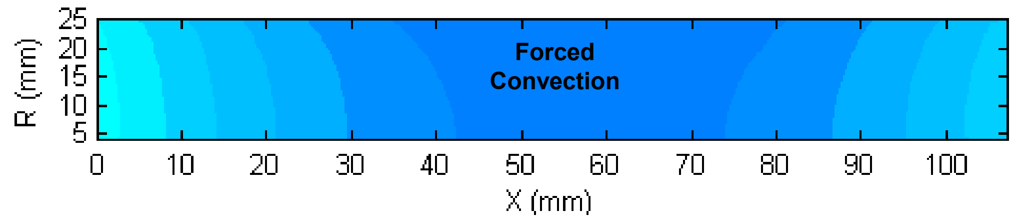


US06 CD cycle

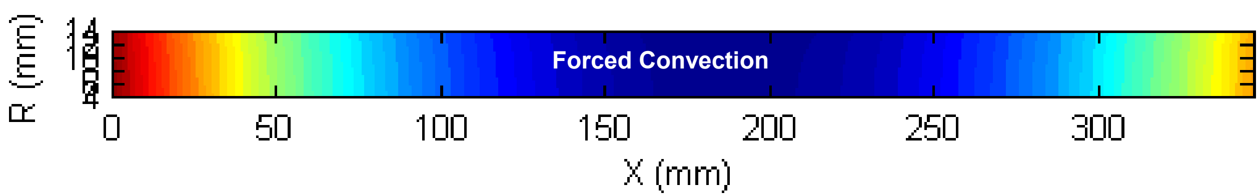
Large Dia. Cell
Natural: -0.1% to +0.1%
Forced: -0.3% to +0.2%



Nominal Cell
Natural: -1.1% to +2.7%
Forced: -1.2% to 2.8%

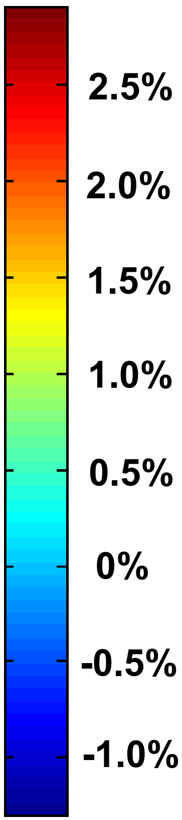


Large Height Cell
Natural: -9% to +21%
Forced: -9% to +21%



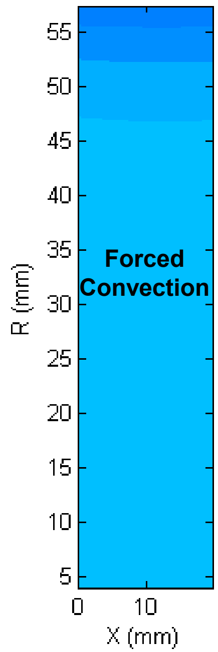
Despite additional thermal imbalance, forced convection does not drastically change localized material usage.

Amp-hour Throughput Imbalance

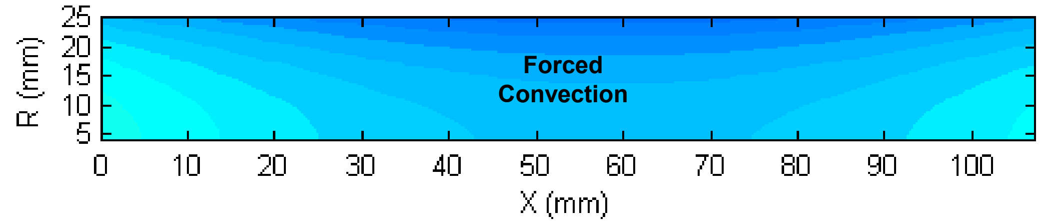


US06 CD cycle

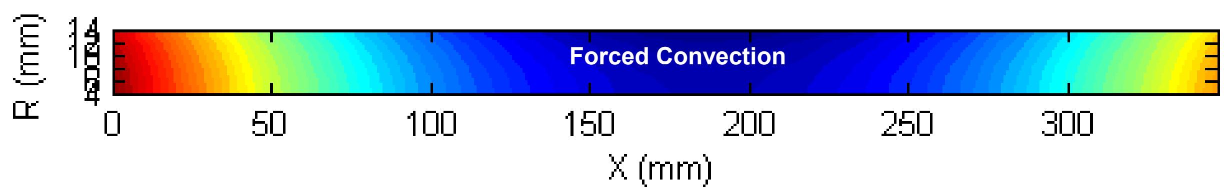
Large Dia. Cell
Natural: -0.1% to +0.03%
Forced: -0.3% to 0.07%



Nominal Cell
Natural: -0.2% to +0.3%
Forced: -0.3% to 0.4%



Large Height Cell
Natural: -1.2% to +2.9%
Forced: -1.3% to 2.9%

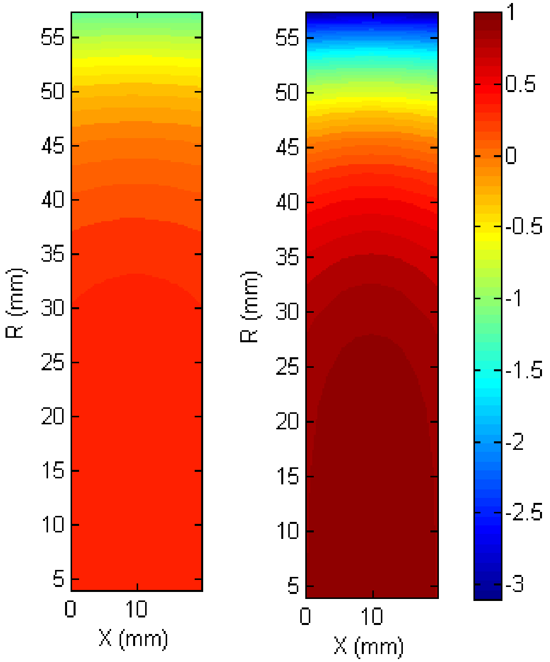


Comparison of natural and forced convection

at t = 690 s of US06 cycle

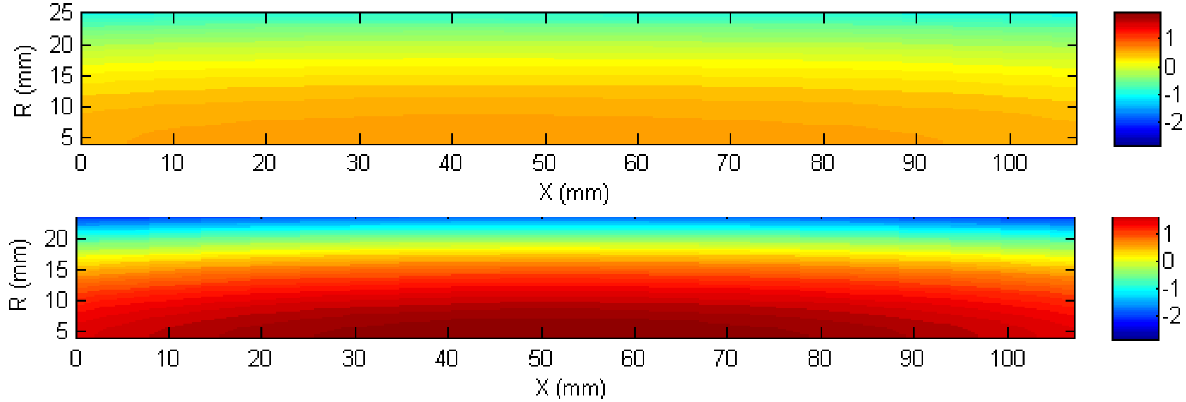
Large D

	T_{avg} (°C)	T_{max}-T_{min} (°C)
Natural	44.4	1.6
Forced	40.9	4.1



Nominal

	T_{avg} (°C)	T_{max}-T_{min} (°C)
Natural	45.2	1.7
Forced	42.6	4.8



Large H

	T_{avg} (°C)	T_{max}-T_{min} (°C)
Natural	47.2	3.2
Forced	43.0	4.4