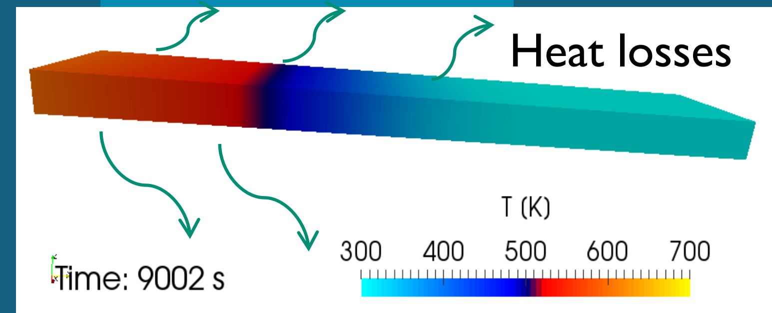
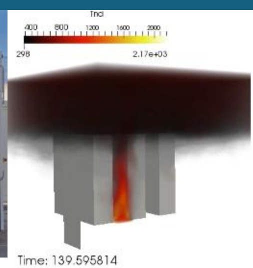
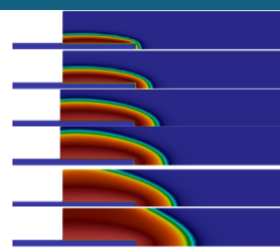
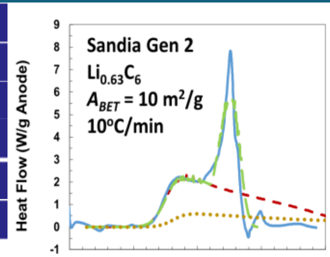
Predicting and mitigating cascading failure of thermal runaway in stacks of Li-ion pouch cells









Presented by

Andrew Kurzawski, Randy Shurtz, Loraine Torres-Castro, Joshua Lamb, John Hewson

Ninth Triennial International Aircraft Fire and Cabin Safety Research Conference October 28-31, 2019

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Thermal runaway and cascading failure

Validated reliability and safety is one of four critical challenges identified in 2013 Grid Energy Storage Strategic Plan

- Failure rates as low as 1 in several million
- Potentially many cells used in energy storage
- Moderate likelihood of ‘something’ going wrong

Increased energy densities and other material advances lead to more reactive systems

A single cell failure that propagates through the pack can have an impact even with low individual failure rates.

How do we decrease the risk?

Single Cell
~0.5-5 Ah

Strings and large
format cells
~10-200 Ah

EV Battery Pack 100s-
1000s cells
10-50 kWh

Stationary storage
system 1000s or more
individual cells
MWh+

www.nissan.com
www.internationalbattery.com
www.samsung.com
www.saft.com

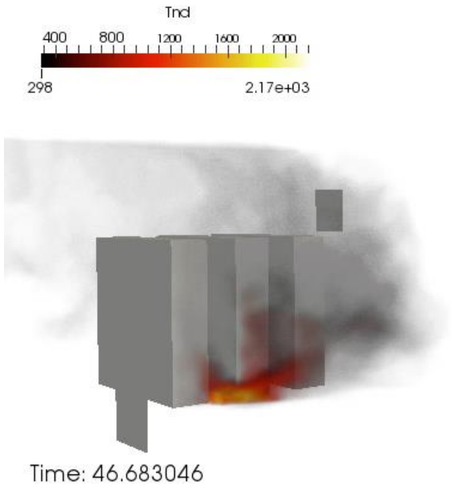
Approaches to designing in safety

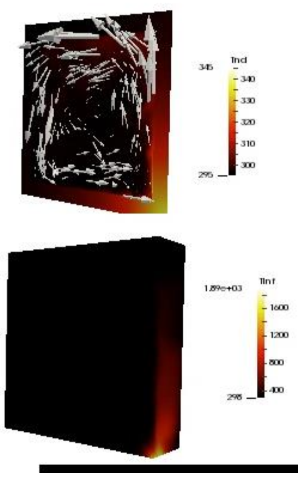
The current approach is to test our way into safety

- Large system (>1MWh) testing is difficult and costly.

Supplement testing with predictions of challenging scenarios and optimization of mitigation.

- Develop multi-physics models to predict failure mechanisms and identify mitigation strategies.
- Build capabilities with small/medium scale measurements.
- Still requires some testing and validation.





Cascading failure testing with passive mitigation

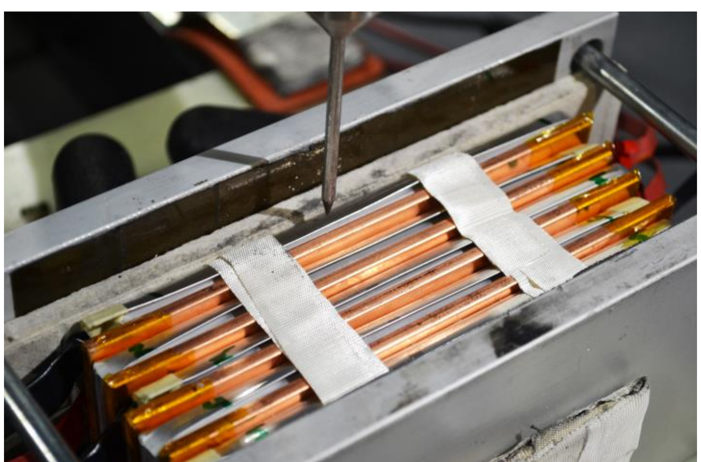
$LiCoO_2$ 3Ah pouch cells

5 closely packed cells with/without aluminum or copper spacer plates

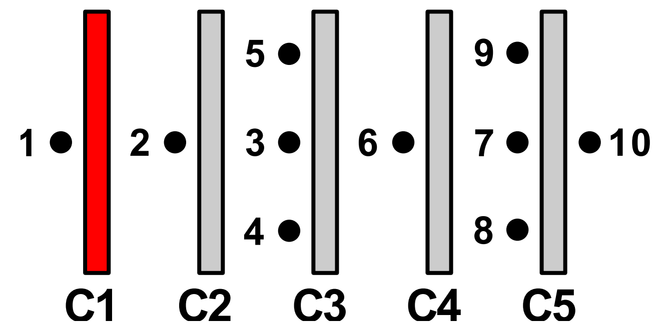
- Spacer thicknesses between 1/32" and 1/8"
- State of charge (SOC) between 50% and 100%

Failure initiated by a mechanical nail penetration in the outer cell (cell 1)

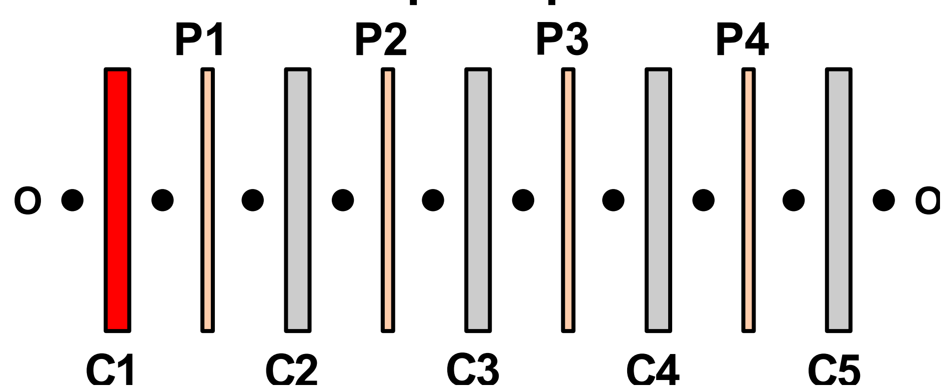
Thermocouples (TC) between cells and spacers (if present)



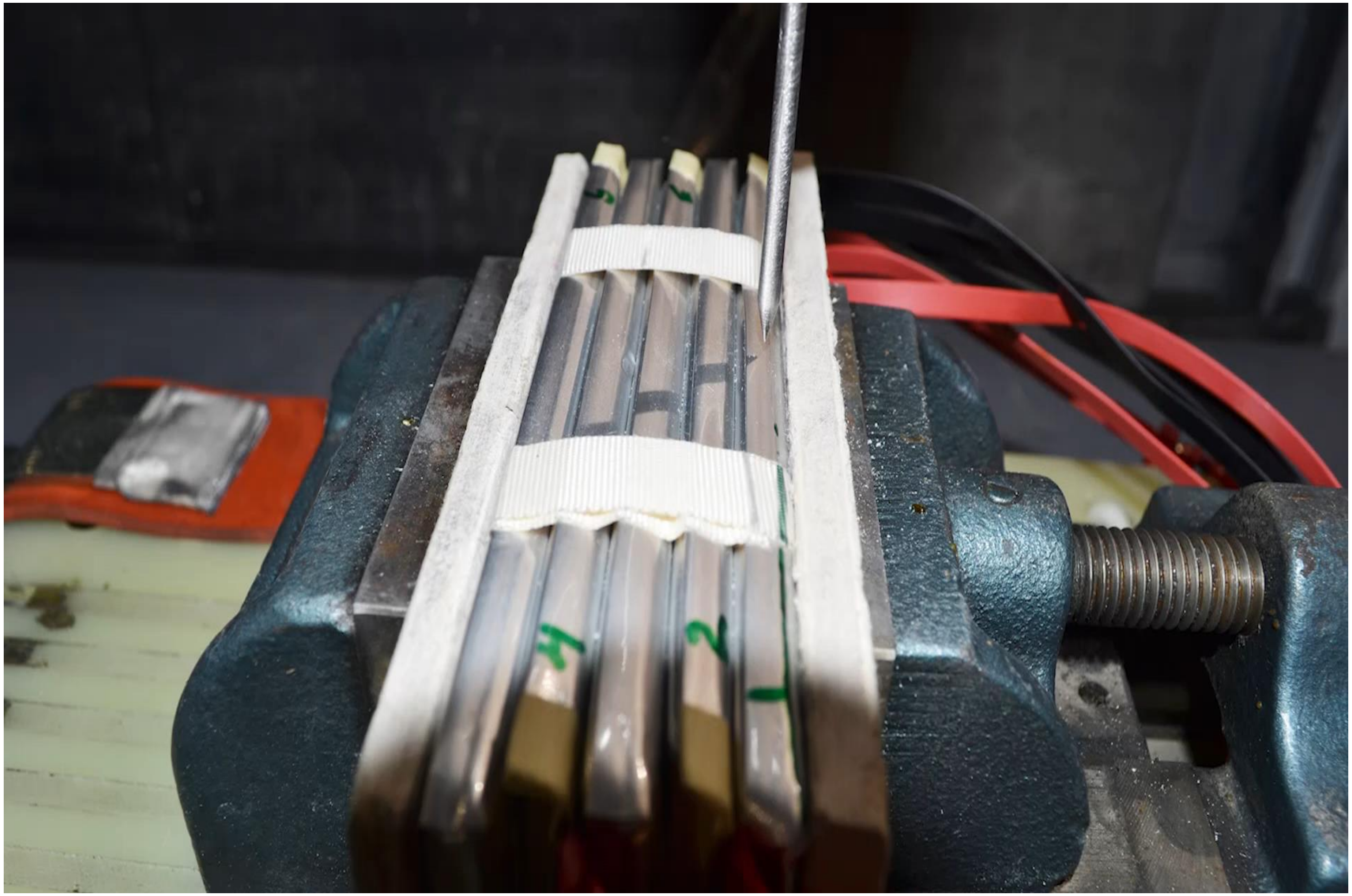
Thermocouple Locations



Thermocouple Locations with spacer plates



Cascading failure testing



Finite element model for full cells in thermal runaway

Discretization in one direction (***x***)

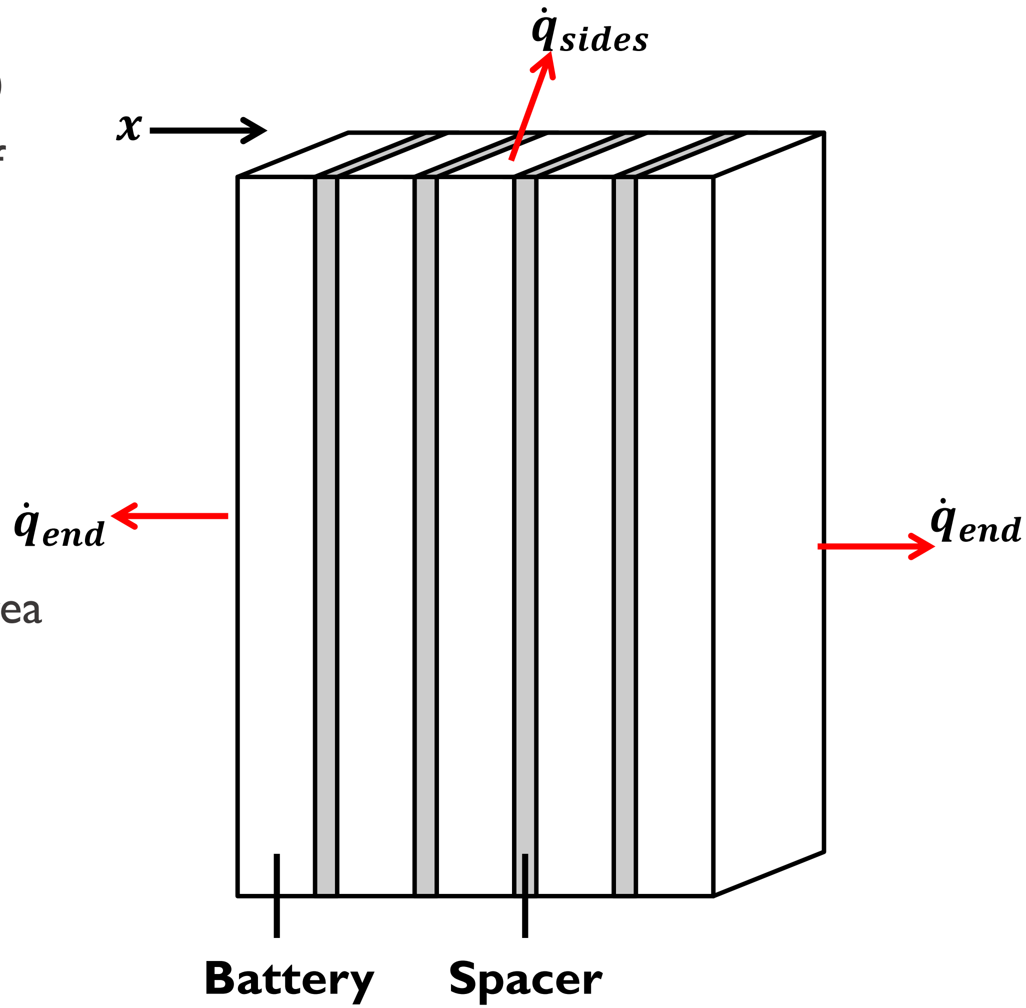
Modeled as a quasi 1-D domain of thin hexahedron elements

Multi-layered system

- Lumped battery material
- Spacers
- End block insulators

Convective heat transfer to surroundings (scaled by surface area to volume ratio for thin domain)

Heat conduction with chemical sources inside battery material



Finite element model equations

Energy conservation:

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

Mass conservation for species i with N_r reactions:

$$\frac{\partial \rho_i}{\partial t} = \sum_{j=1}^{N_r} (\nu''_{ij} - \nu'_{ij}) r_j$$

Energy source:

$$\dot{q}''' = -\sum_{j=1}^{N_r} \Delta H_j r_j$$

Chemical source terms for thermal runaway

Preliminary chemistry model from literature

- Based on Dahn group (1999-2001)
- Derived from calorimetry
- Good onset predictions
- Under-predicts peak temperature

Empirical chemical reactions:

- SEI decomposition (Richard 1999)

$$(CH_2OCO_2Li)_2 \rightarrow Li_2CO_3 + gas$$

- Anode-electrolyte (Shurtz 2018)

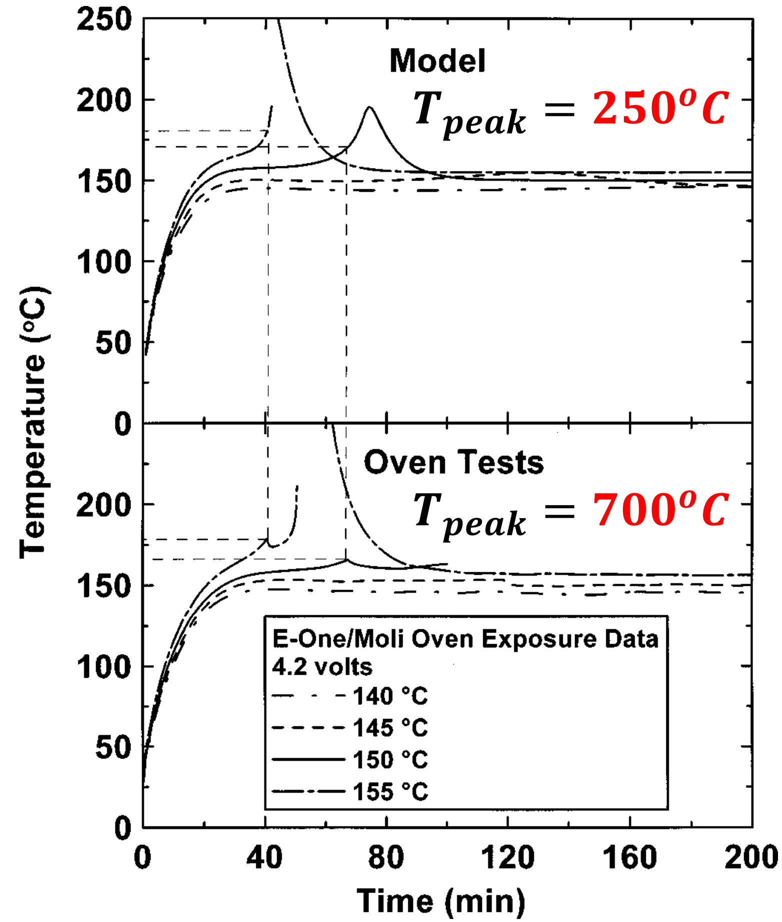
$$2C_6Li + C_3H_4O_3 \rightarrow 2C_6 + Li_2CO_3 + gas$$

- Cathode-electrolyte (Hatchard 2001)

$$CoO_2 + \frac{2}{15}C_3H_4O_3 \rightarrow \frac{1}{3}Co_3O_4 + gas$$

- Short-circuit

$$C_6Li + CoO_2 \rightarrow C_6 + LiCoO_2$$



Hatchard, T. D., D. D. MacNeil, A. Basu and J. R. Dahn (2001). Journal of the Electrochemical Society **148**(7): A755-A761.

Anode-electrolyte calorimetry and modeling

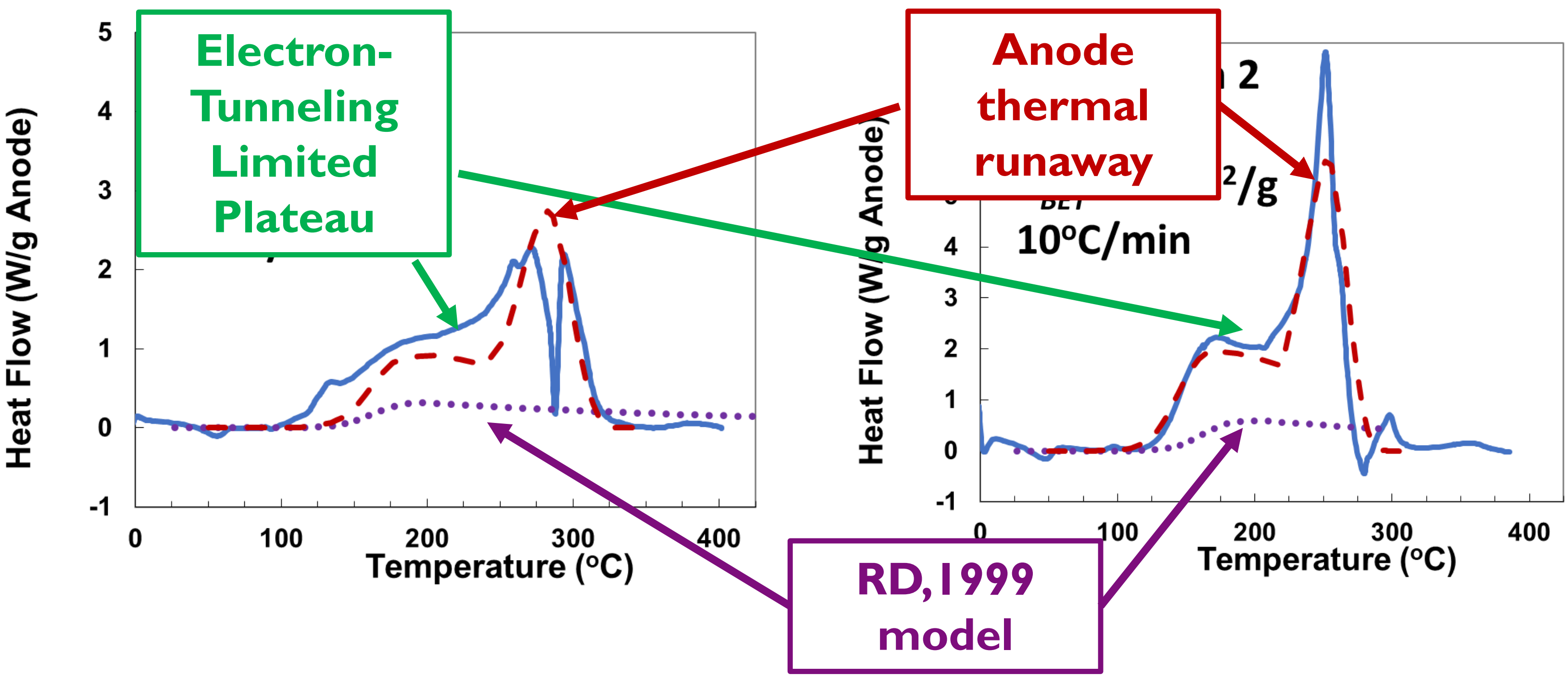
Anode-electrolyte calorimetry suggest several regimes during thermal runaway

- Initiation – Plateau -Runaway

Anode-electrolyte reactions generate heat

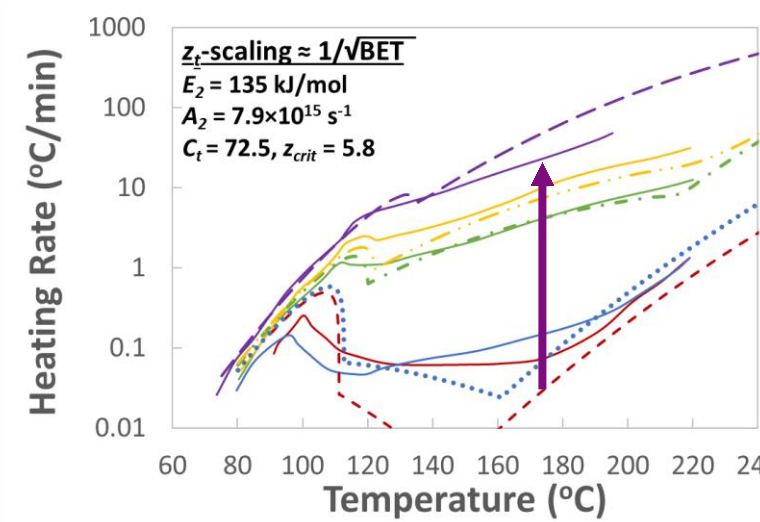
- Could raise cell temperatures $\sim 650^{o}C$
- Nominal reaction:

$$2LiC_6 + C_3H_4O_3 \rightarrow 2C_6 + Li_2CO_3 + C_2H_4$$

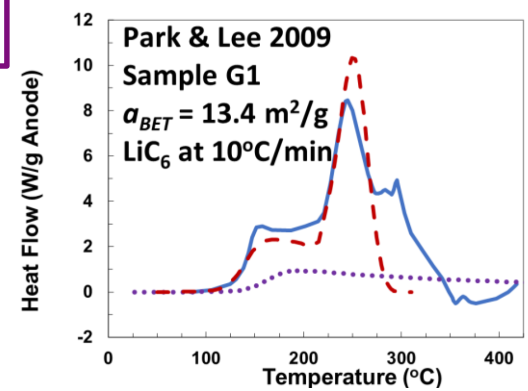


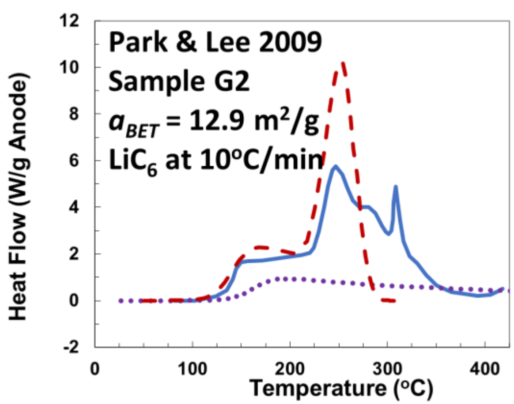
More predictions with the comprehensive model

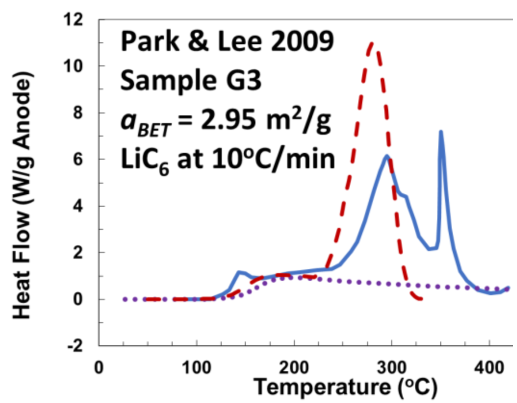
Predicting the full range of behavior over a range of particle sizes

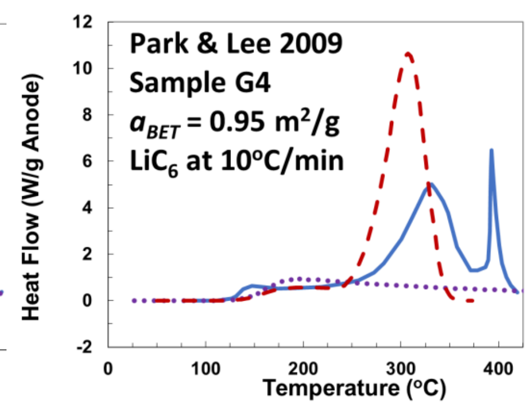


Increasing specific area





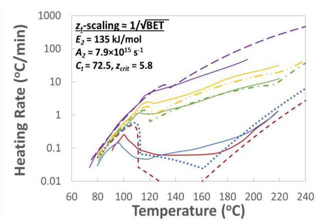


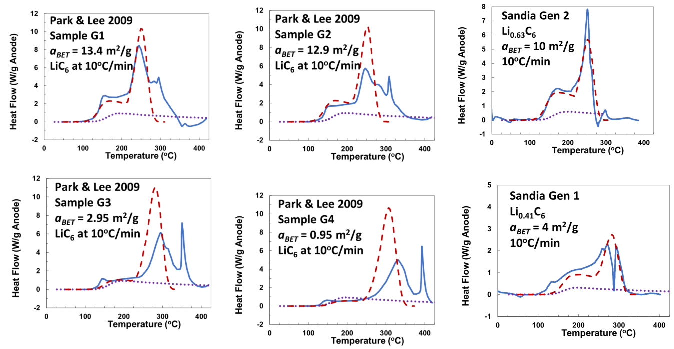


Increasing specific area

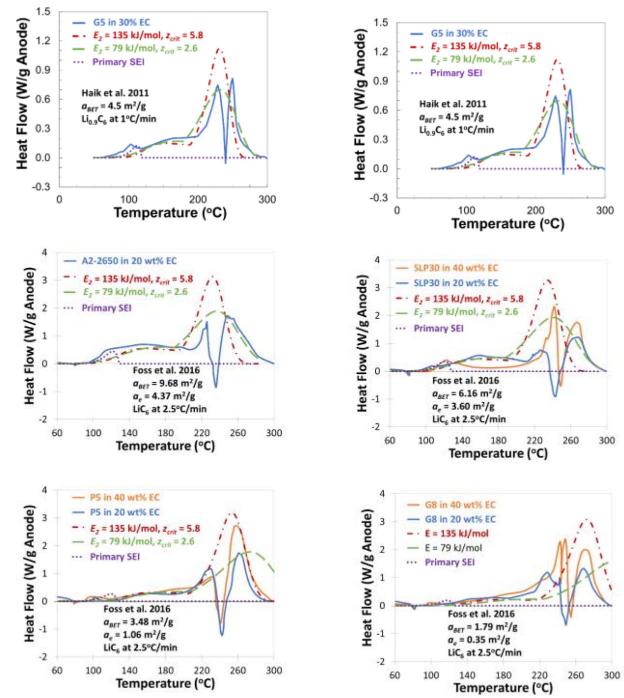
Many predictions with the comprehensive model 24 x DSC, 5 x ARC

Shown earlier

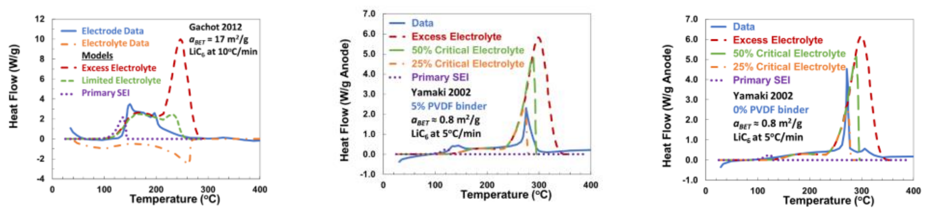




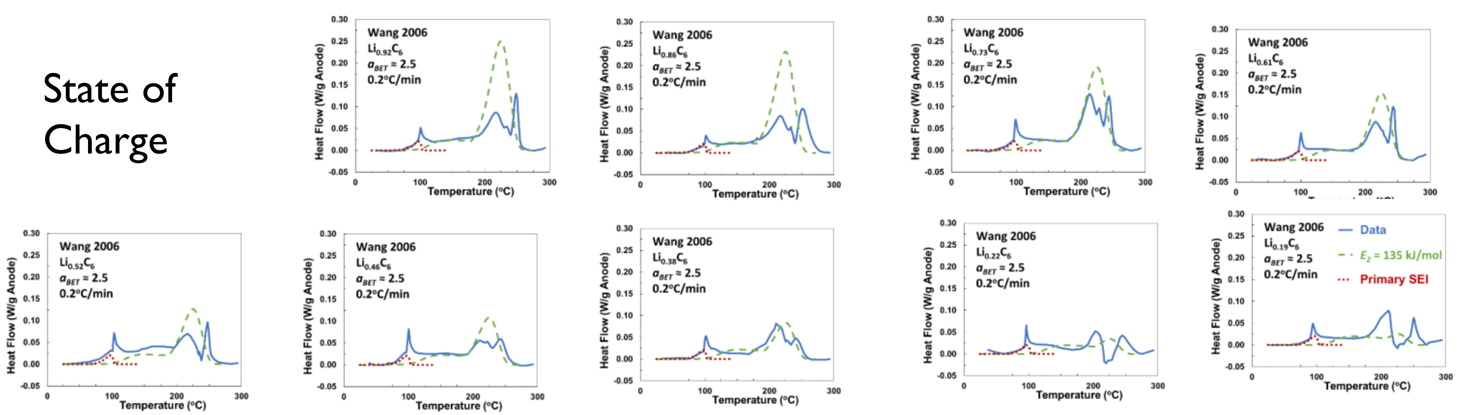
Detailed area measurements



Limiting Electrolyte



State of Charge



A comprehensive model for anode-electrolyte runaway

Edge Area Effect

Limiting Electrolyte

Electron-Tunneling Limiter

$$-\frac{dx_i}{dt} = x_i \frac{a_e}{a_0} \frac{m_E}{(m_{50} + m_E)} A_2 \exp\left(-\frac{E_2}{RT}\right) \exp(-z_t)$$

Barrier Growth

Variable Area Effect

Critical Barrier

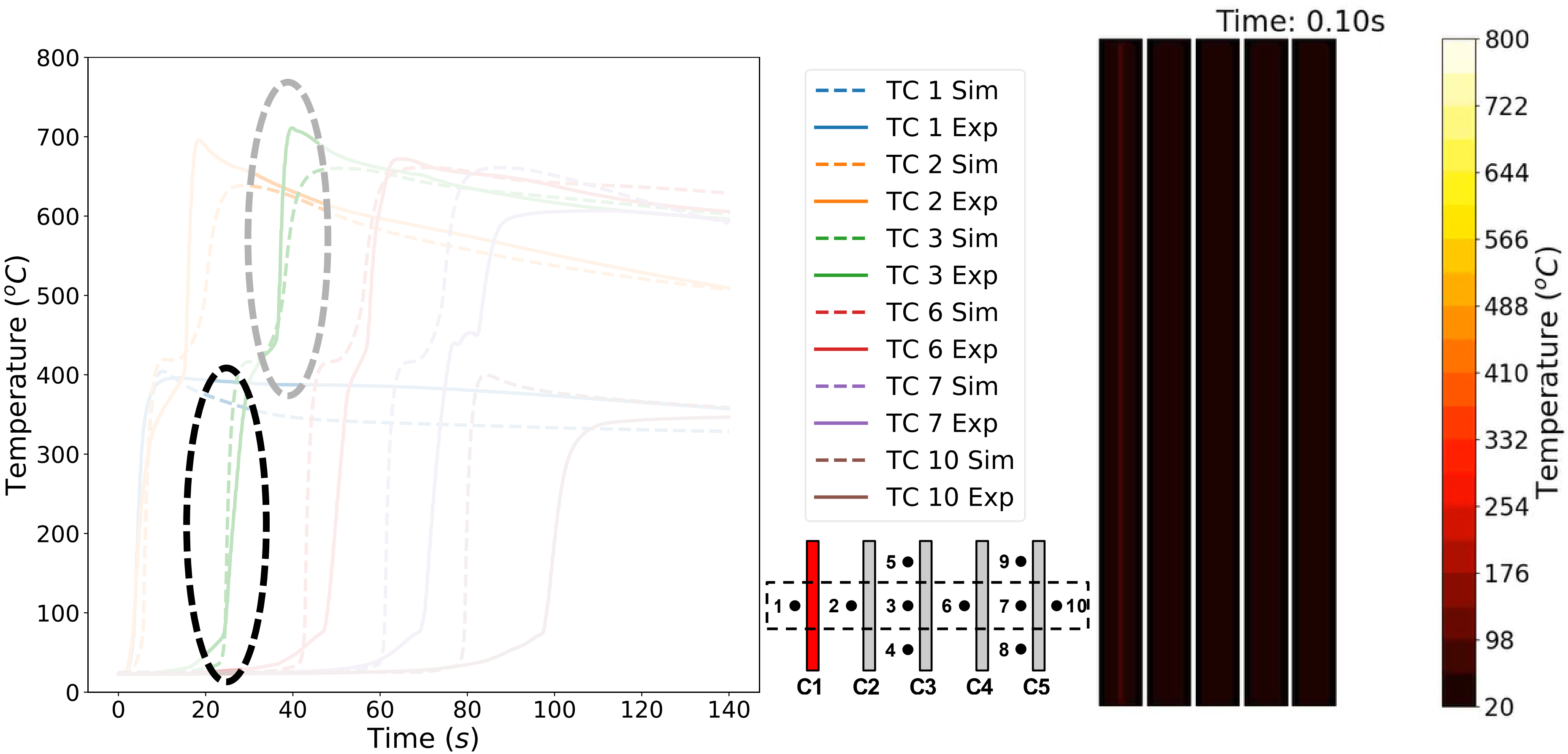
Allows Acceleration

$$\frac{dz_t}{dt} = -\frac{dx_i}{dt} \frac{C_t}{\left(\frac{a_{BET}}{a_0}\right)^{n_t}} \text{ for } z_t < z_{crit} \text{, and } \frac{dz_t}{dt} = 0 \text{ otherwise}$$

$$Q\,[W/g] = -\frac{dx_i}{dt} \frac{\Delta H_{rxn}}{W_a}$$

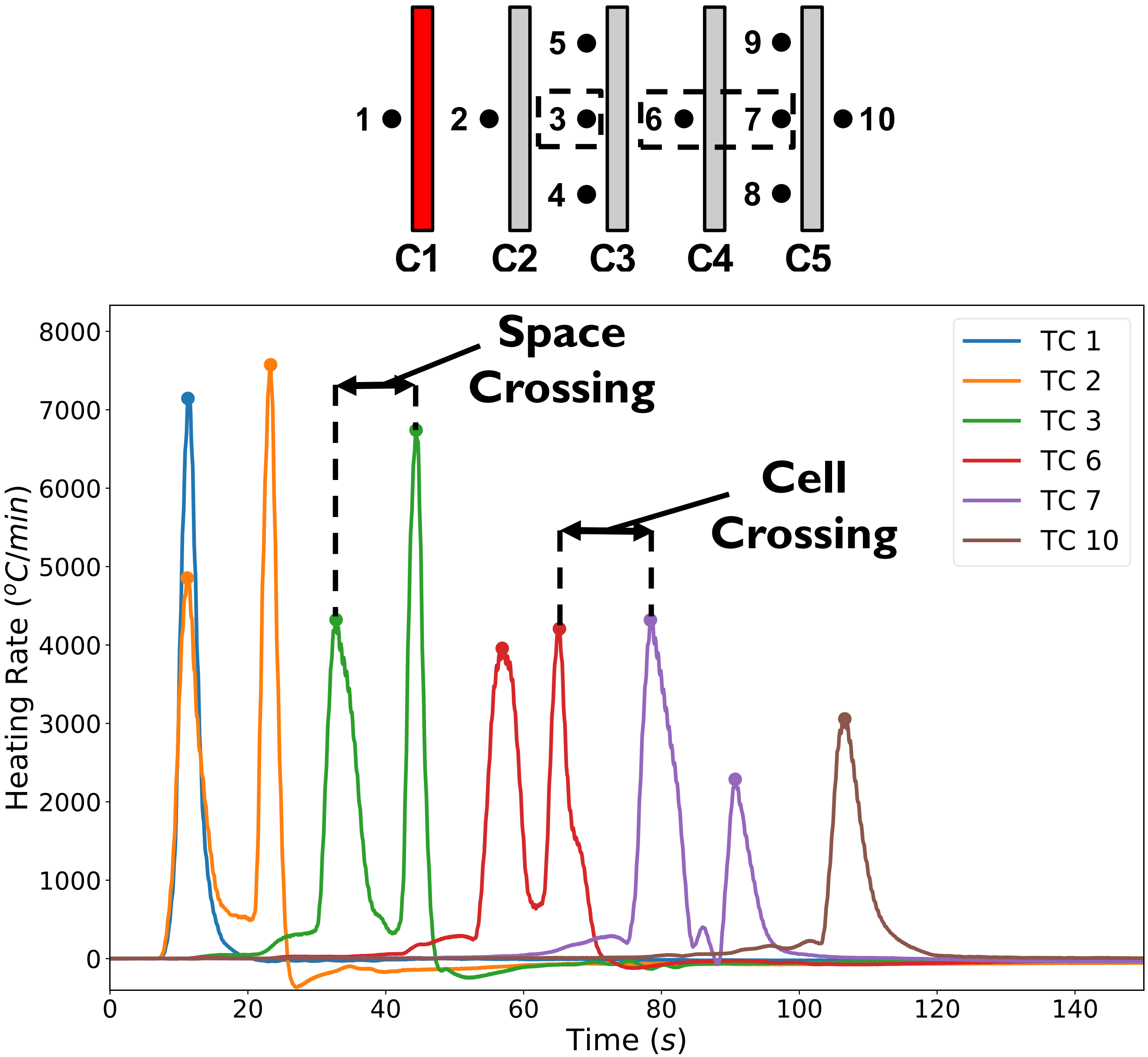
Heat Release with new ΔH_{rxn}

Simulation results: 100% SOC, no spacers



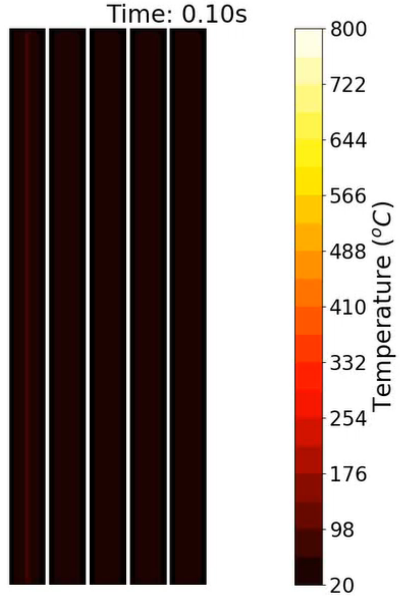
- Prediction of peak temperatures and cooling
- Cell crossing speed over-predicted

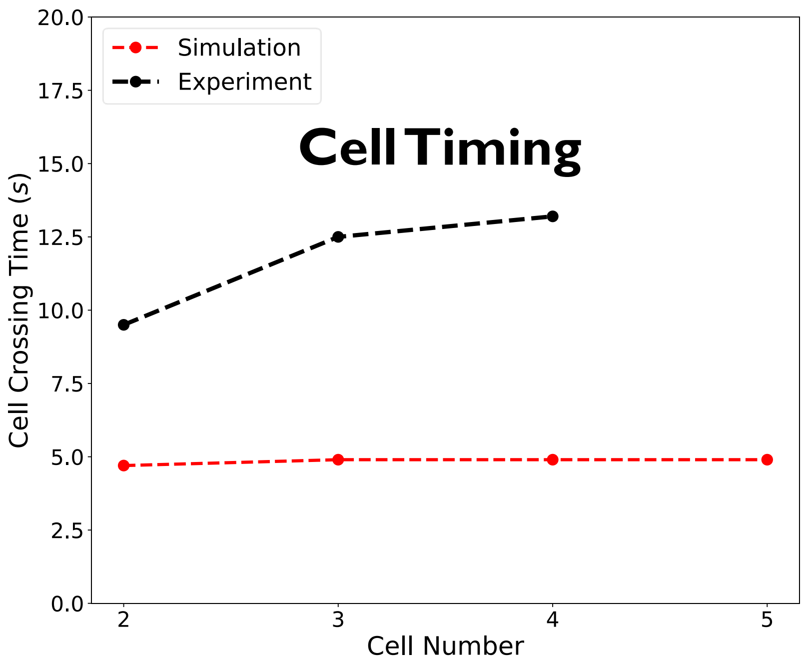
Deriving propagation times

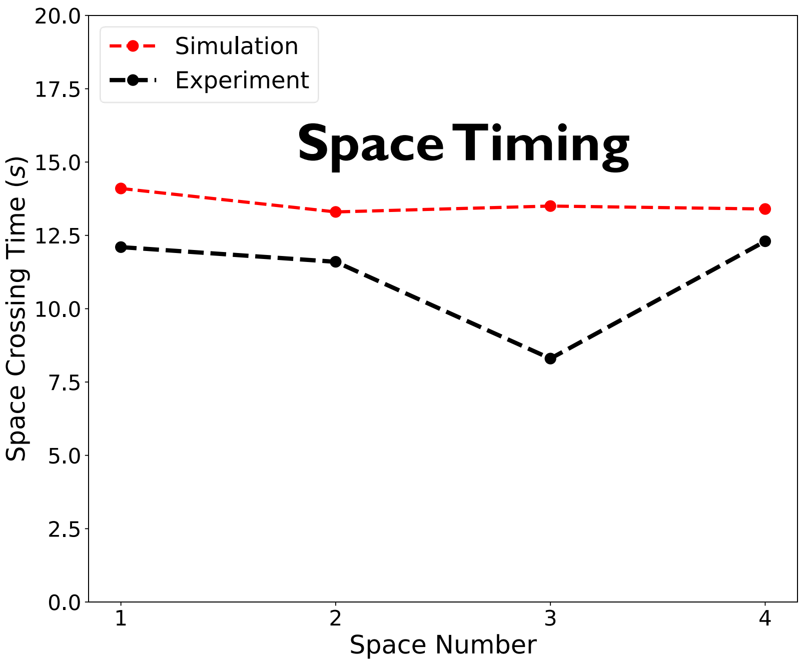


Predicted crossing times: 100% SOC, no spacers

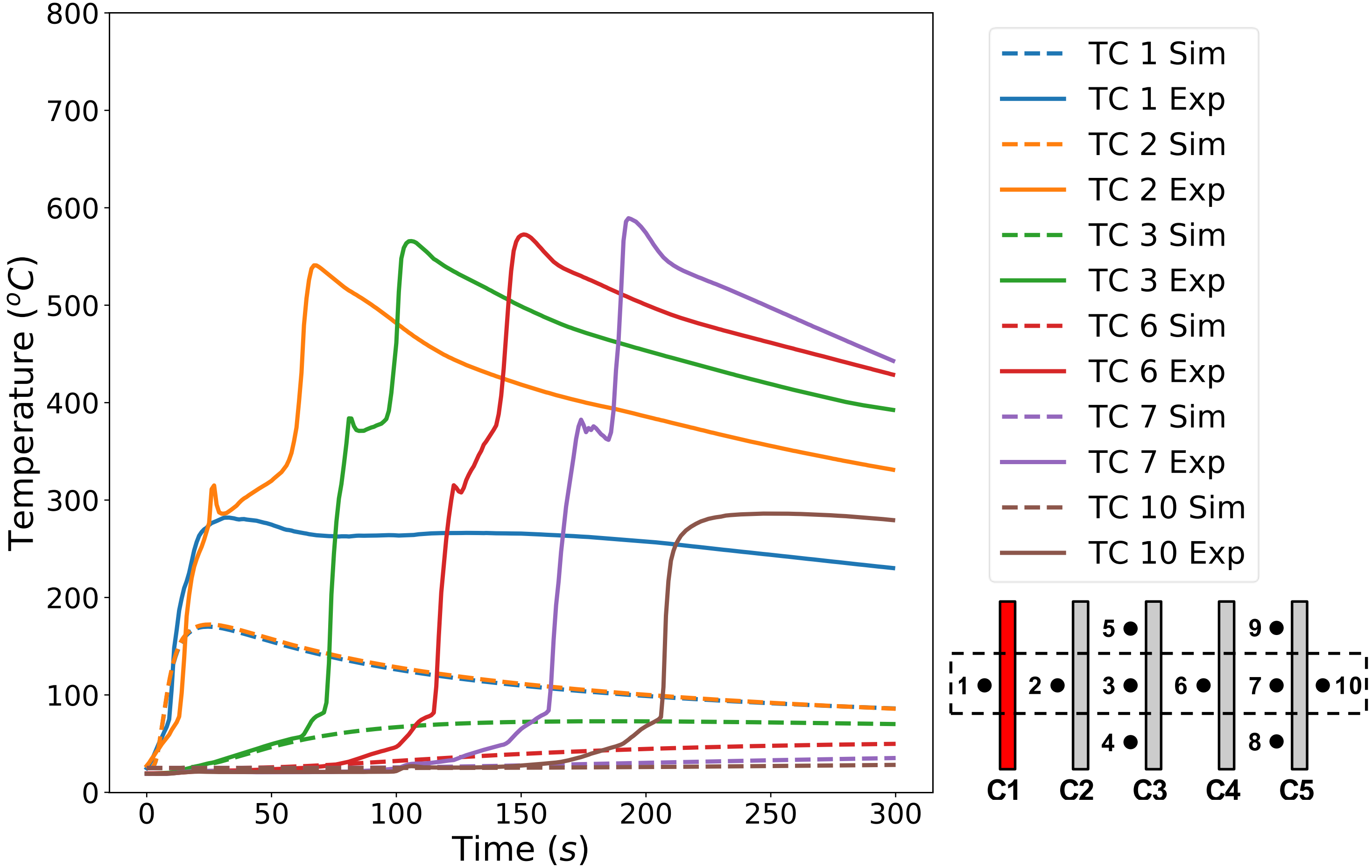
- Experimental cell and space crossing times are on the same order.
- Cell crossing times are under-predicted and space crossing times are over-predicted.





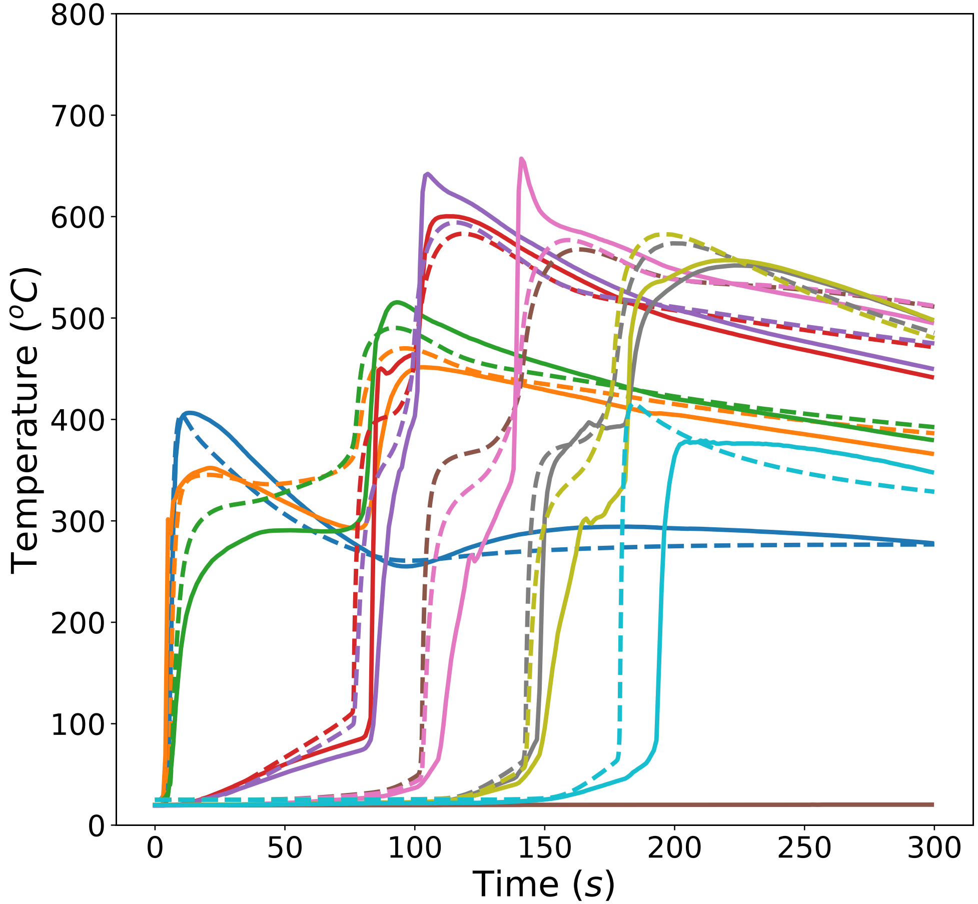


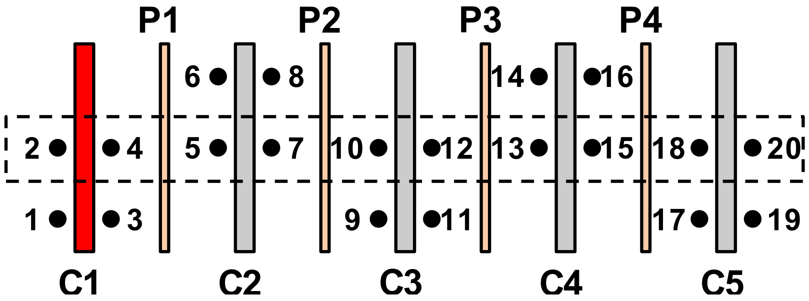
Simulation results: 80% SOC, no spacers



- Insufficient heat generation to initiate thermal runaway outside of the nail penetration region
- Experimental peak temperatures lower than 100% SOC

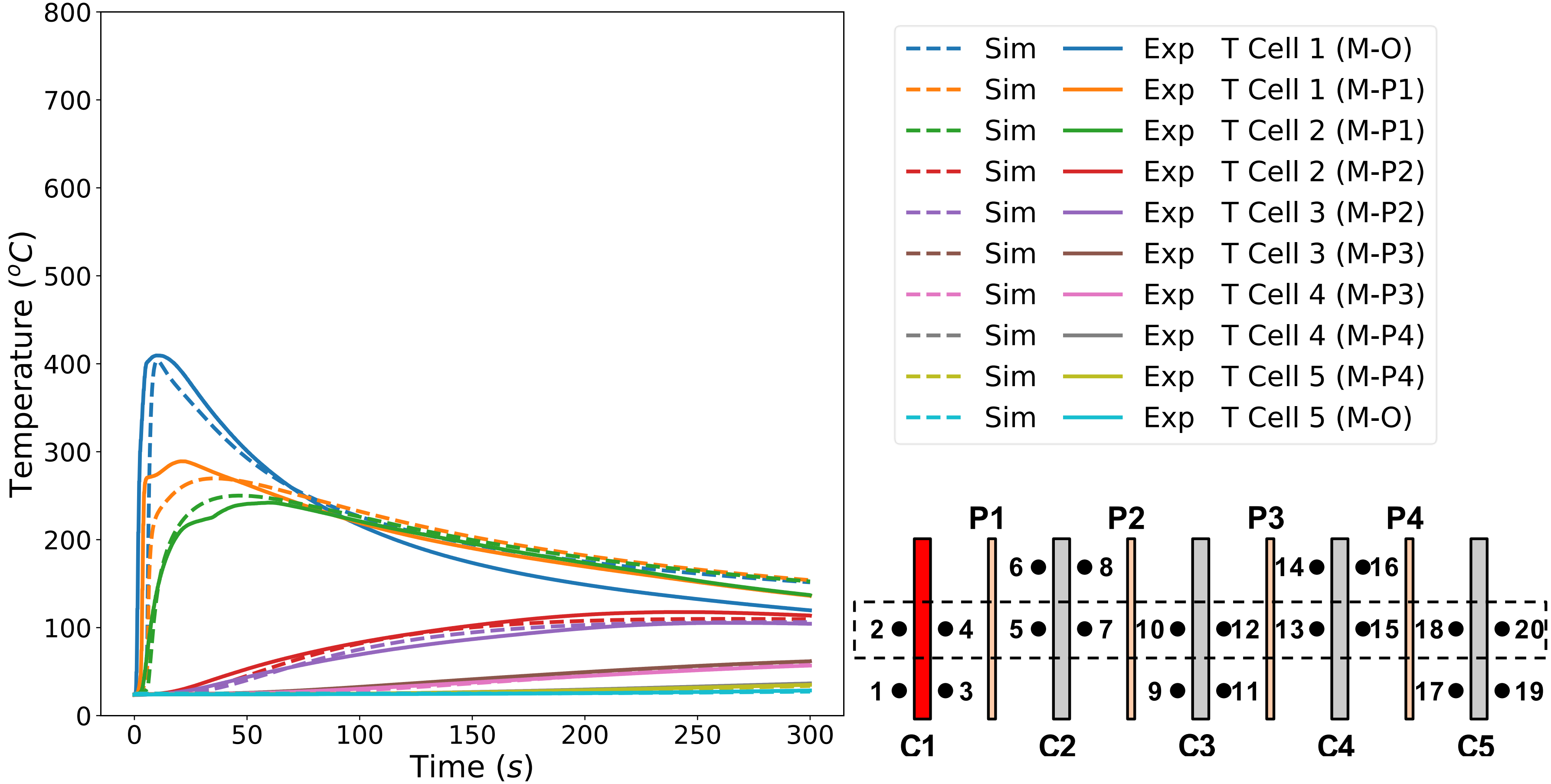
Simulation results: 100% SOC, 1/32” aluminum spacers





- Temperature difference in TCs on either side of the plates under-predicted
- Cell crossing speed still over-predicted

Simulation results: 100% SOC, 1/16” copper spacers



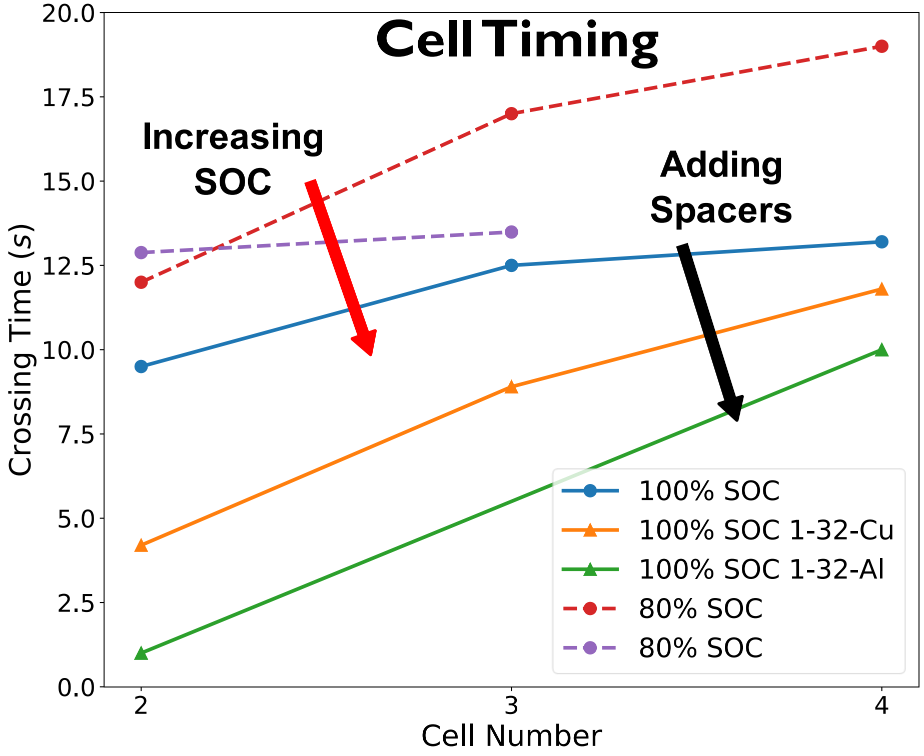
- No propagation in simulations and experiments

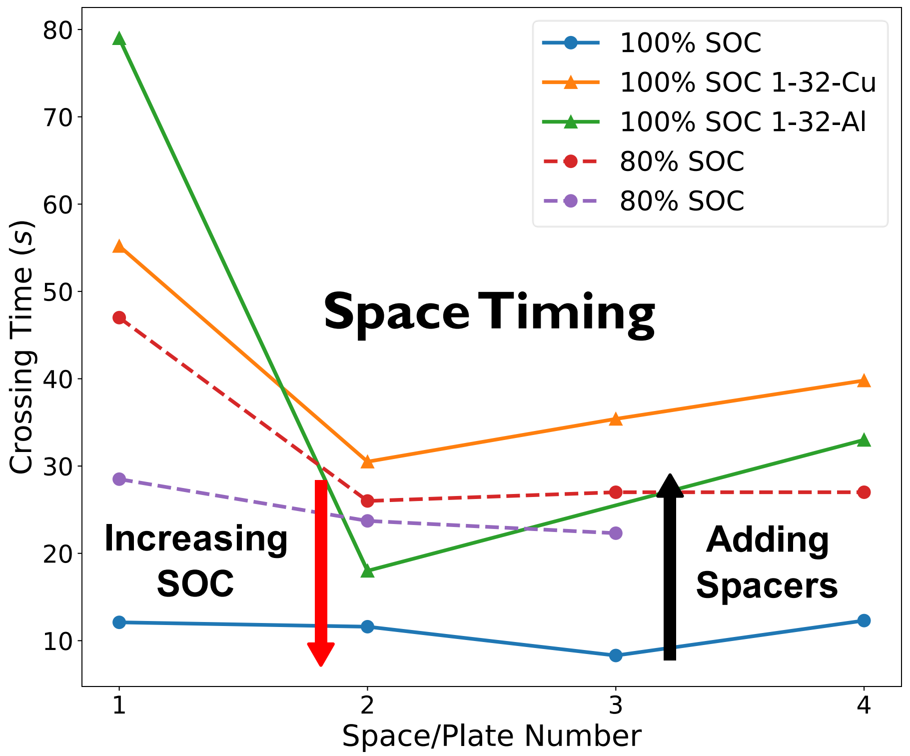
Cascading failure: propagation speeds

Adding spacers **increases** space crossing time, but **decreases** cell crossing time

Increasing state of charge (SOC) **decreases** both space and cell crossing time

Interplay between **heat capacity** of system and **energy release**





Heat capacity and SOC: limits of propagation

Interplay between **heat capacity** of system and **energy release:**

$$\text{Energy/Capacity} = Q_{cells}/(m_{cells}c_{p,cells} + m_{spacers}c_{p,spacers})$$

Case Description	Energy/Capacity (K)	Experiment	Simulation
100% SOC	940	Propagation	Propagation
1/32" Aluminum	819	Propagation	Propagation
1/32" Copper	778	Propagation	No Propagation
80% SOC	752	Propagation	No Propagation
1/16" Aluminum	725	Cell 2 Failure	No Propagation
75% SOC	705	Cell 2 Failure	No Propagation
1/16" Copper	663	Cell 2 Failure	No Propagation
1/8" Aluminum	590	No Propagation	No Propagation
1/8" Copper	512	No Propagation	No Propagation
50% SOC	470	No Propagation	No Propagation

Summary

Finite element model with chemical source terms was tested against experimental data.

- Captures trends at 100% SOC, over-predicts propagation velocity through cells.
- Model is under-conservative with predictions when heat capacity is increased and SOC is decreased.

There is a need for validated chemical source models tested at higher heat release rates.

Ongoing work to improve mechanistic understanding of thermal and chemical time scales.

- Comprehensive cathode models
- Transport limited reaction kinetics

Acknowledgements

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