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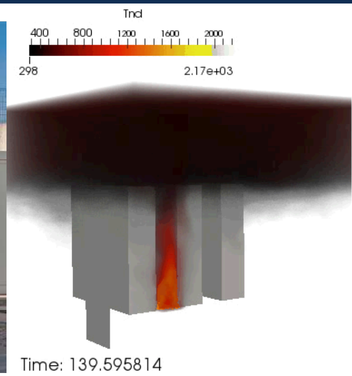

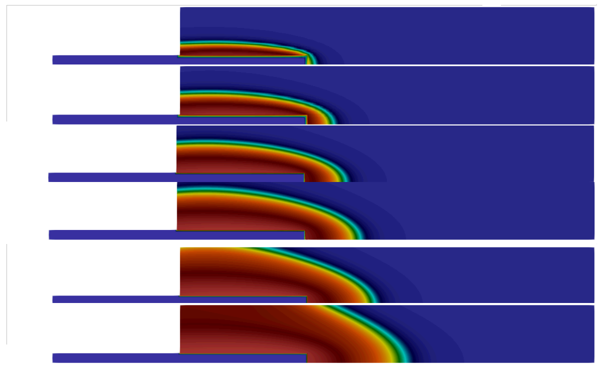

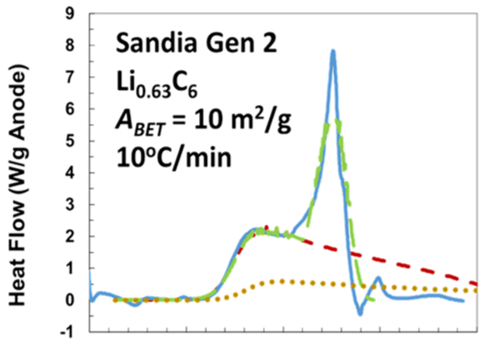

Modeling the limits of thermal runaway in Li-ion packs and designing tests to measure those limits

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Sandia National Laboratories

JANNAF meeting

December 4, 2017

SAND2017-XXXXC

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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,

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+

- 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?

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

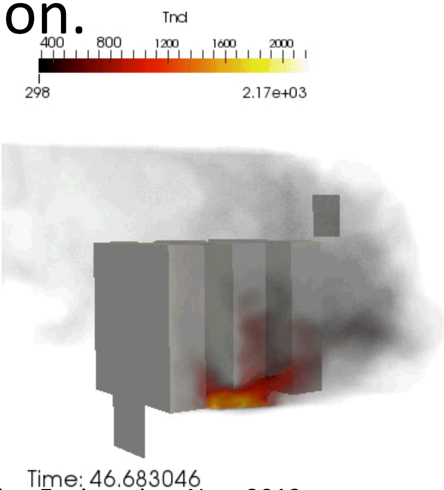
Approaches to designing in safety

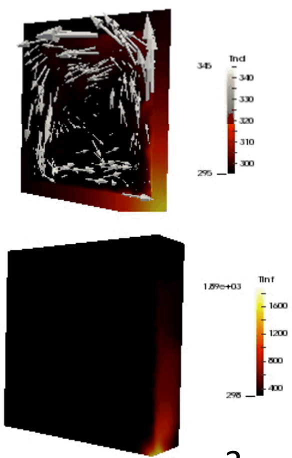
The current approach is to test our way into safety[1]

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

Consider supplementing testing with predictions of challenging scenarios and optimization of mitigation.

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

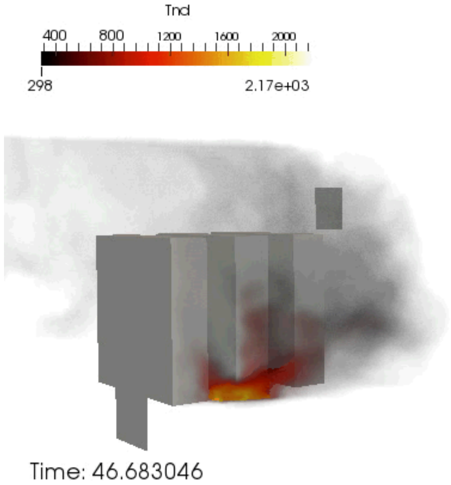


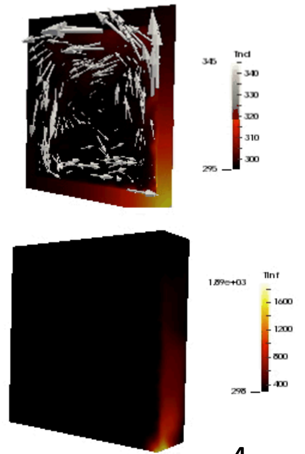


[1] 'Power Grid Energy Storage Testing Part 1.' Blume, P.; Lindenmuth, K.; Murray, J. EE – Evaluation Engineering. Nov. 2012.

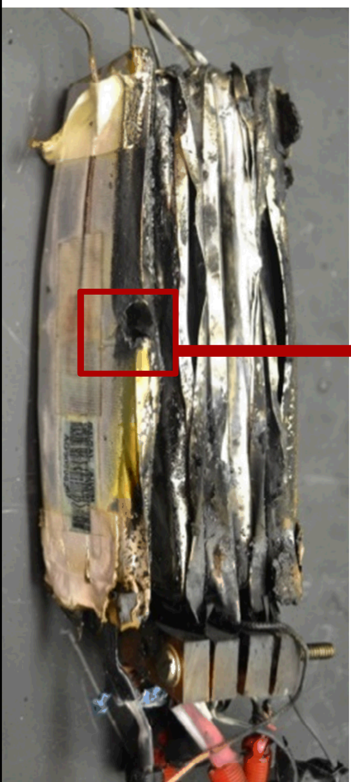
A range of risks to energy storage systems

- Internal short circuit
  - Soft short—gradual discharge.
  - Hard short—sudden discharge.
- Mechanical damage,
- External heating or internal overheating.
- Overcharge.



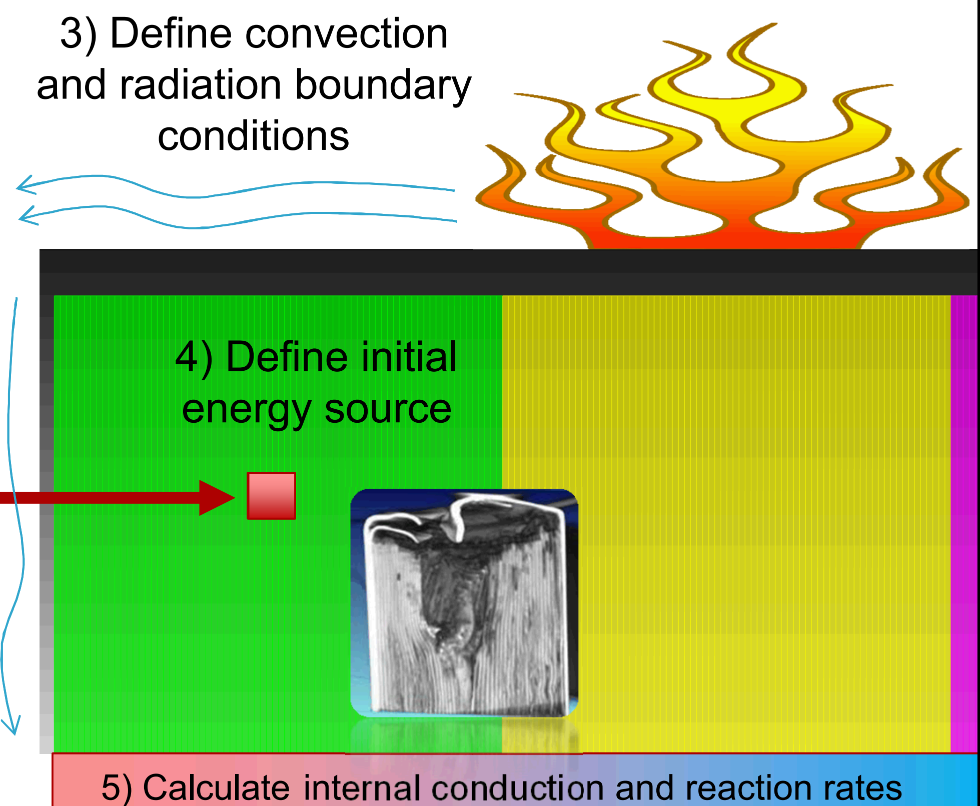


Models for thermal runaway processes



1) Simplify & Discretize Geometry

2) Define initial composition, thermal properties, reactions (species & energy source terms)



Models Need Parameters

- Preliminary chemistry model from literature
  - Based on Dahn group from 2000, 2001
  - Derived from calorimetry data (ARC and DSC)
  - Needs to be recalibrated—not expressed in fundamental cell characteristics.
- Empirical chemical reactions
  - SEI decomposition $2\ ROCO_2Li \rightarrow Li_2CO_3 + prod$
  - Cathode-electrolyte $CoO_2 + C_3H_4O_3 \rightarrow \frac{1}{3}Co_3O_4 + prod$
  - Electrolyte-salt $C_3H_4O_3 + LiPF_6 \rightarrow prod$
  - Anode-electrolyte $C_6Li + C_3H_4O_3 \rightarrow Li_2CO_3 + prod$
- This model form has been utilized repeatedly for oven-test runaway prediction.

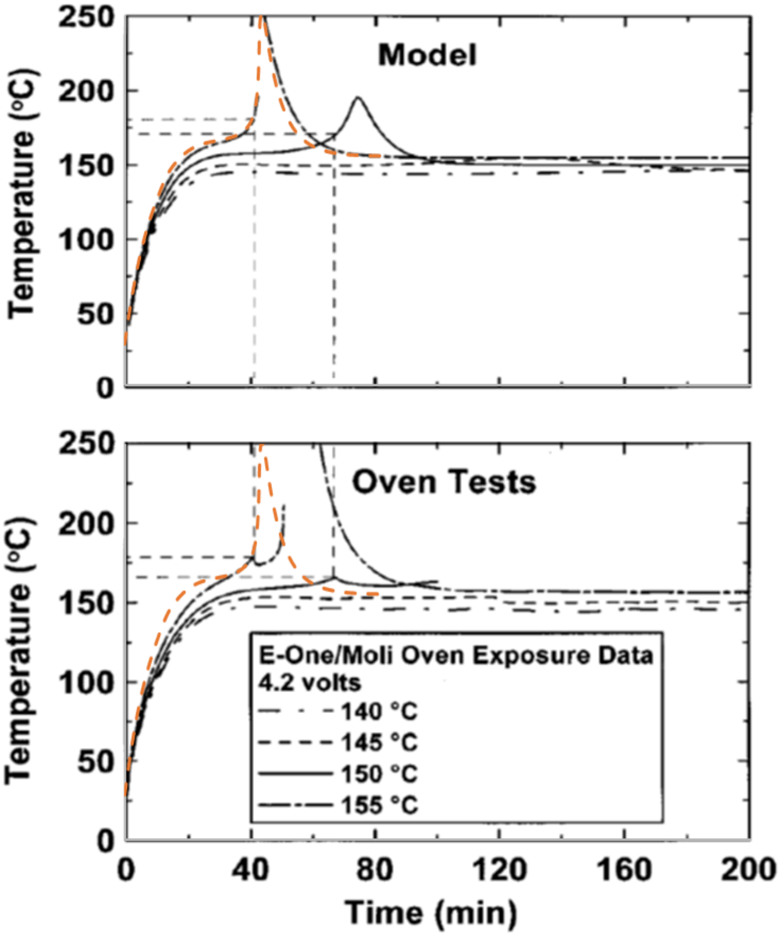


Figure 2. A comparison of oven exposure test results to model predictions: (top) model predictions and (bottom) oven test results for 18650 E-One/Moli Energy cells charged to 4.2 V.

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

Thermal and electrochemical reactants are same

- Discharge reaction: $C_6Li + CoO_2 \rightarrow C_6 + LiCoO_2$
- Charged materials are reactive.

- SEI decomposition $2\,ROCO_2Li \rightarrow Li_2CO_3 + prod$
- Cathode-electrolyte $CoO_2 + C_3H_4O_3 \rightarrow \frac{1}{3}Co_3O_4 + prod$
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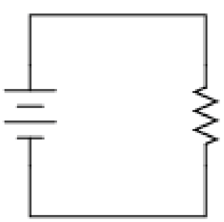
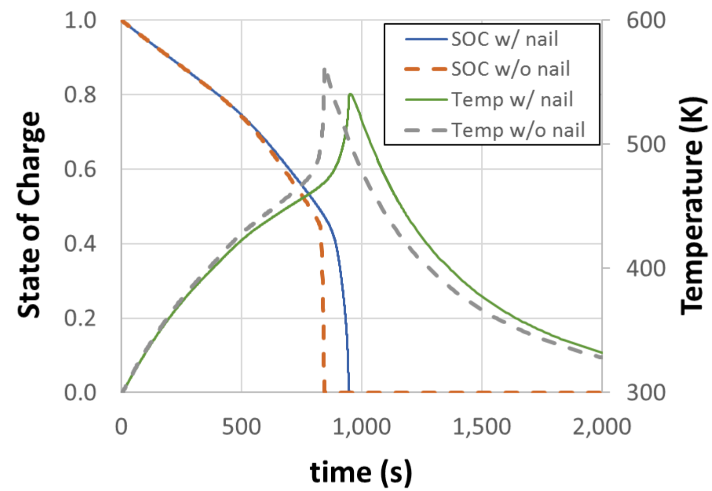
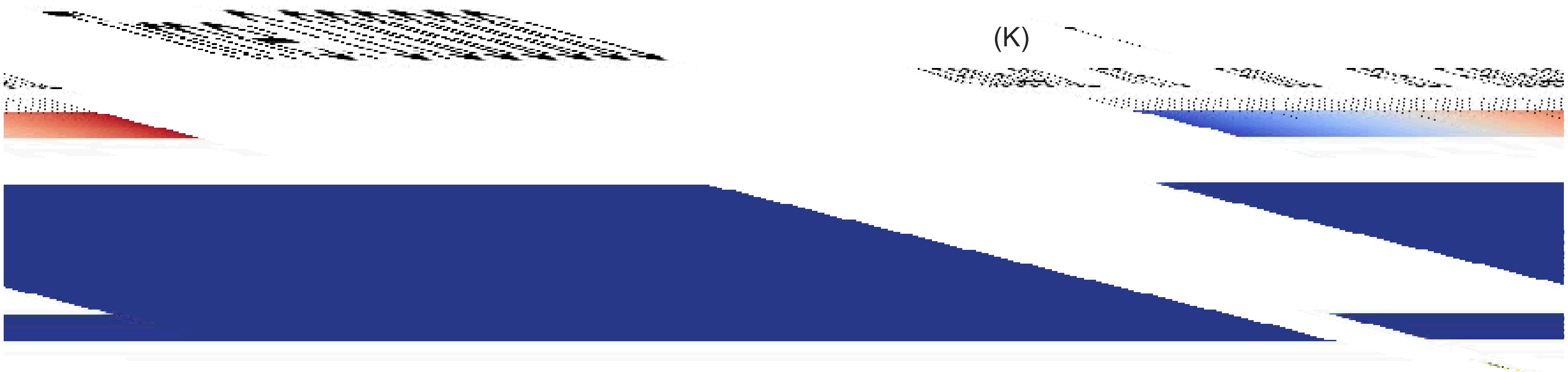


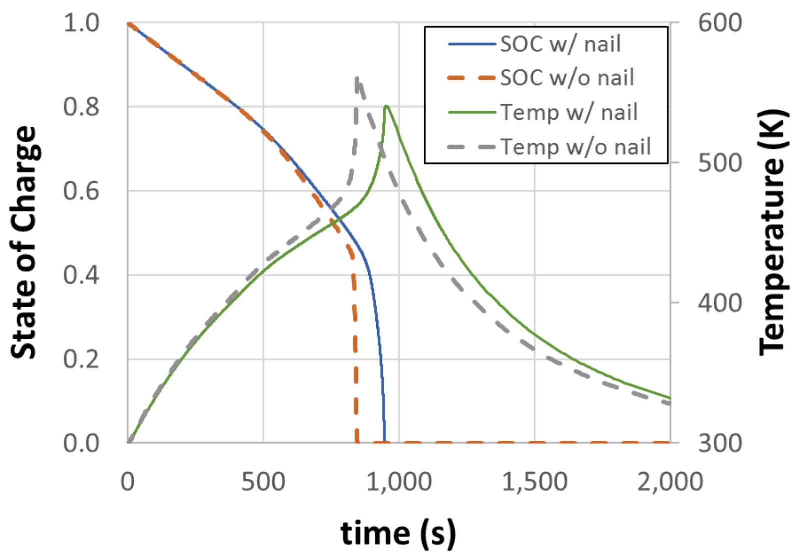
Simulated short circuit plus thermal runaway

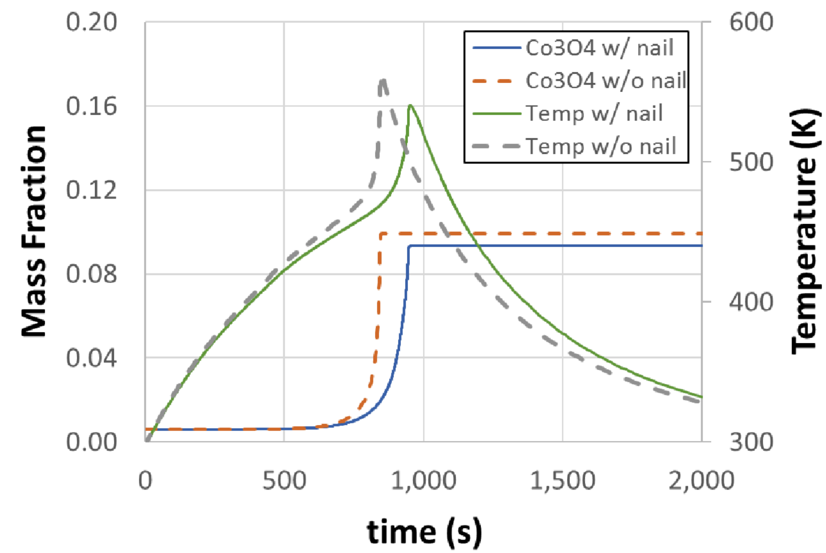


Short-circuit induced runaway in meshed 18650 with nail



R = 1.4 ohm, h = 7 W/m^2/K, Meshed 18650 with 50% heat release in nail



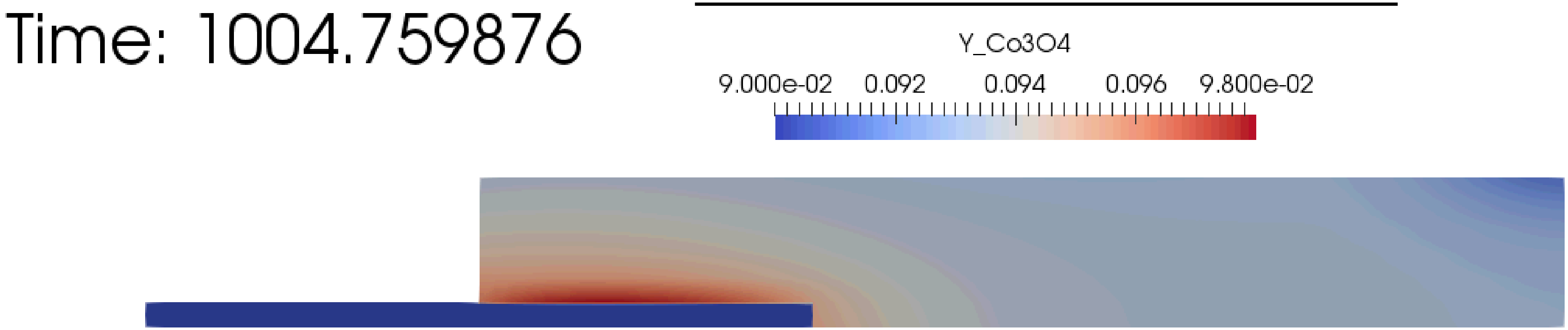


- Effects of inhomogeneity increase as scale increases beyond the lumped-capacitance regime.

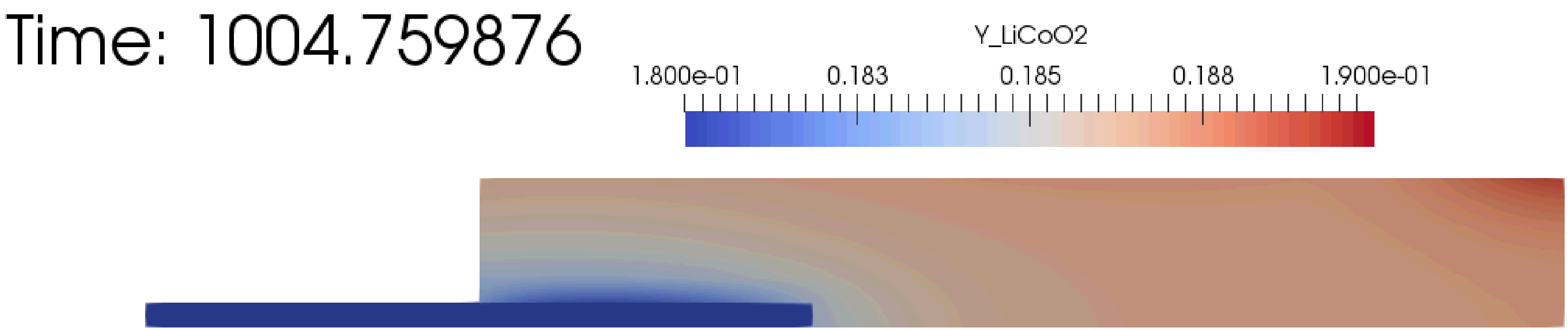
Relative importance of short-circuit versus thermal reactions

R = 1.4 ohm, h = 7 $W/m^2/K$, Meshed 18650 with 50% heat release in nail

Thermal Reaction Cathode Product



Short Circuit Cathode Product



Energetics of thermal runaway in high-temperature environments and under internal short-circuits

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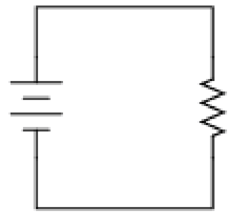
Simulated oven test

high
temp
environ

Simulated short circuit

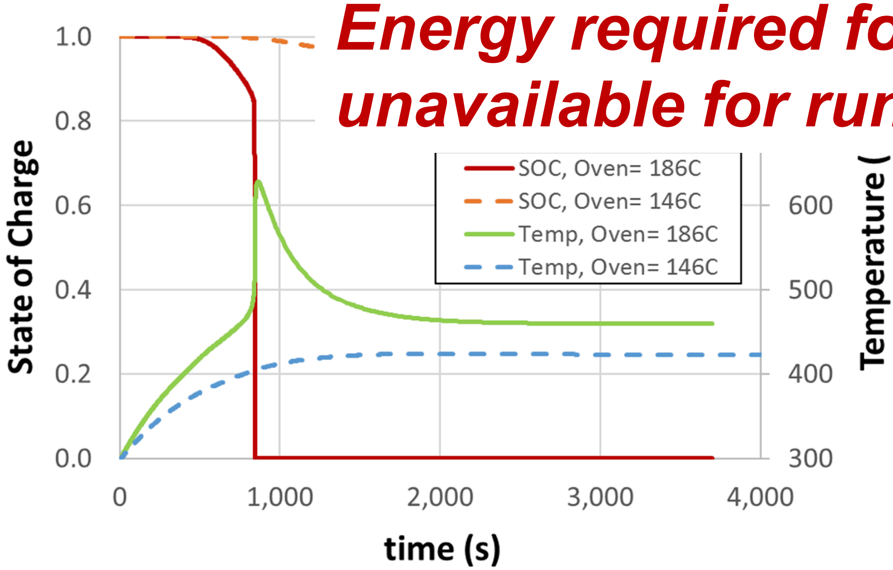


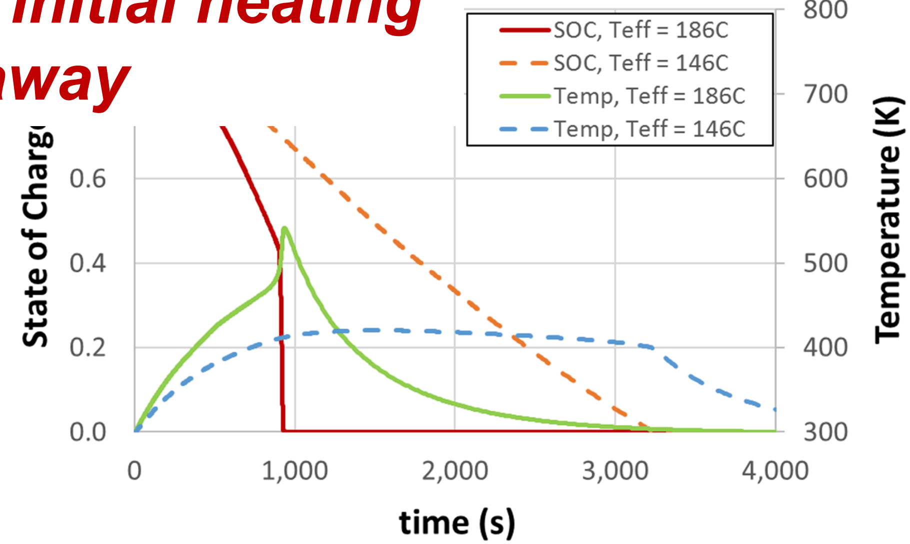
analogy with oven temperature

$$P = \frac{V^2}{R}$$

$$T_{eff} = T_{\infty} + P / h_{net} A$$

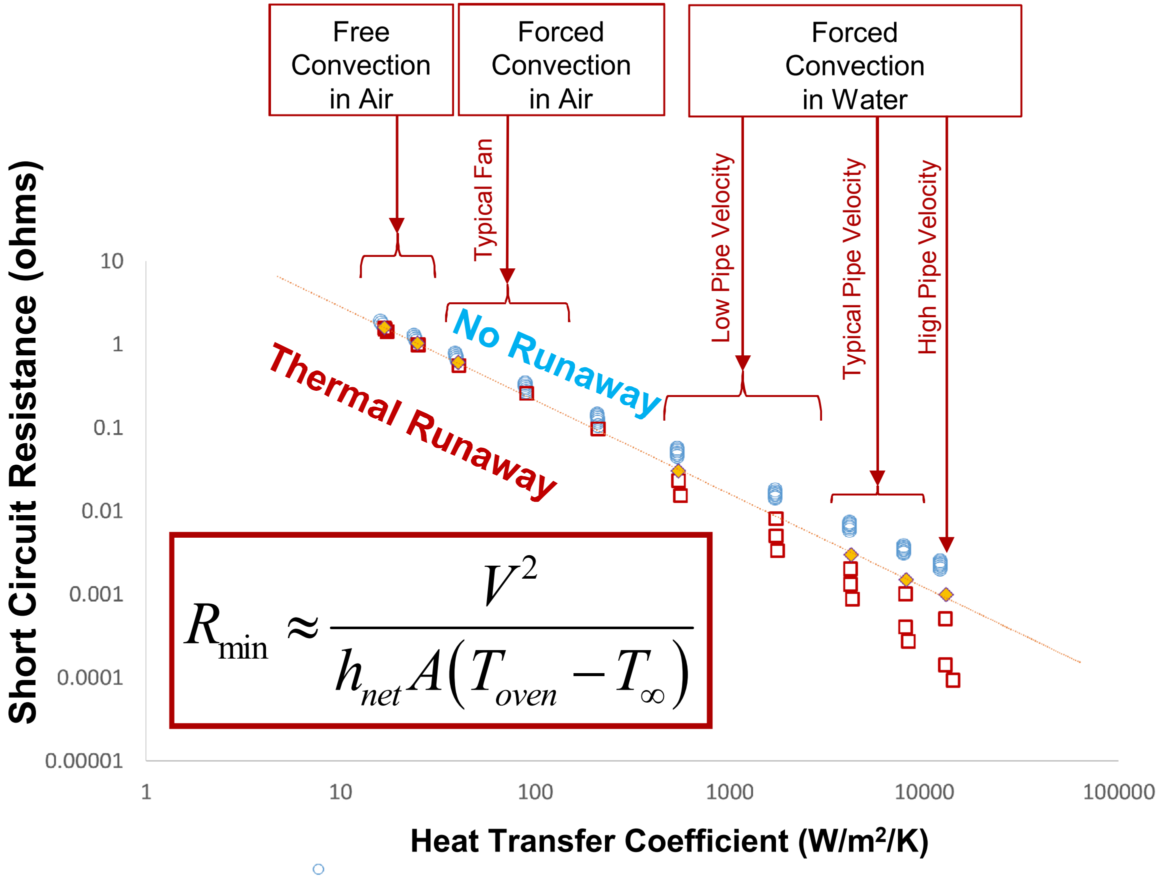
Energy required for initial heating unavailable for runaway



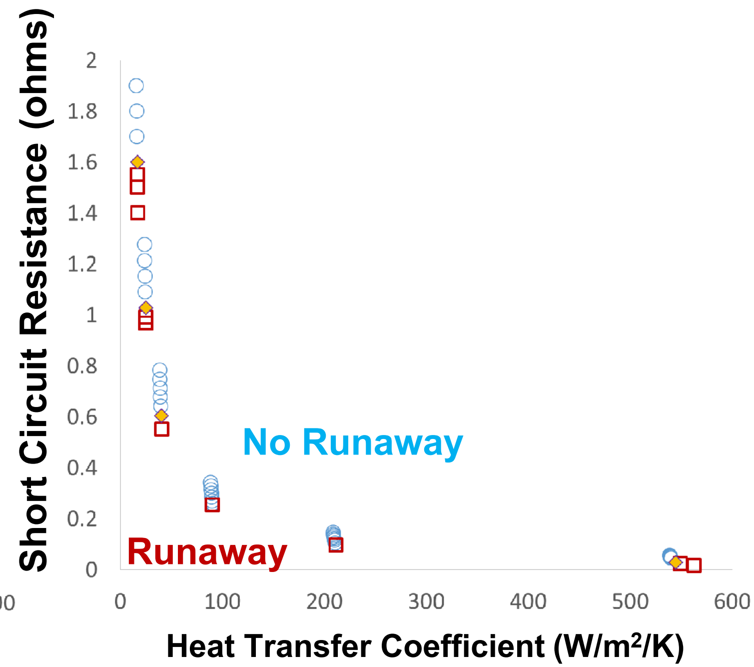


How Much Cooling to Suppress Runaway with Internal Short Circuit?

$$T_{eff} = T_\infty + P / h_{net} A \qquad P = \frac{V^2}{R}$$

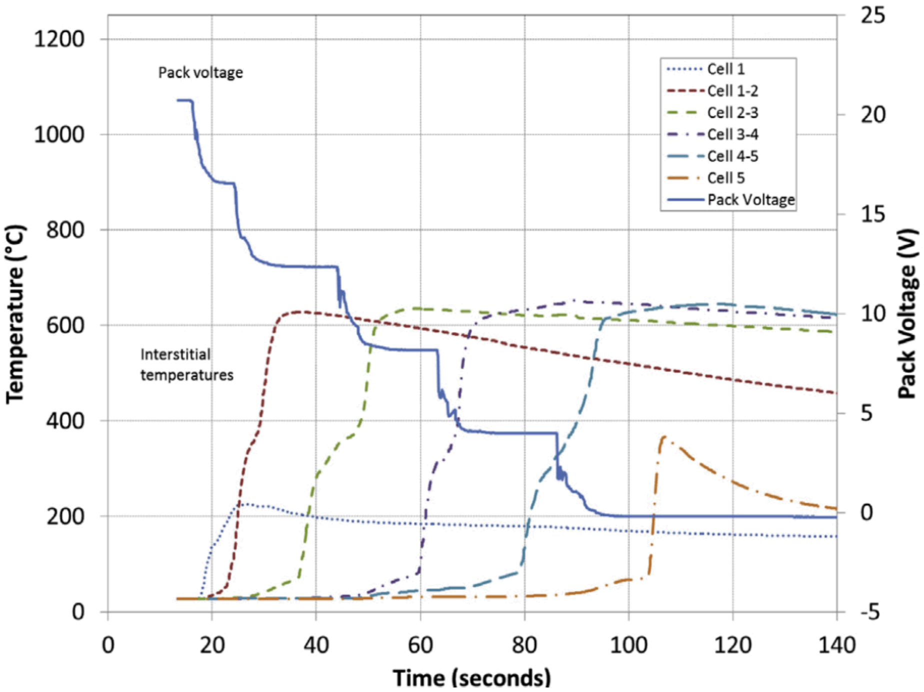


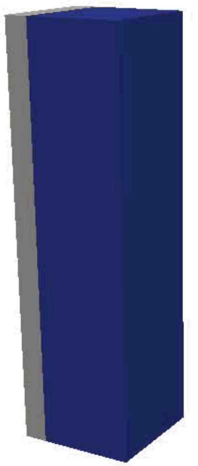
$$R_{min} \approx \frac{V^2}{h_{net} A \left(T_{oven} - T_\infty\right)}$$

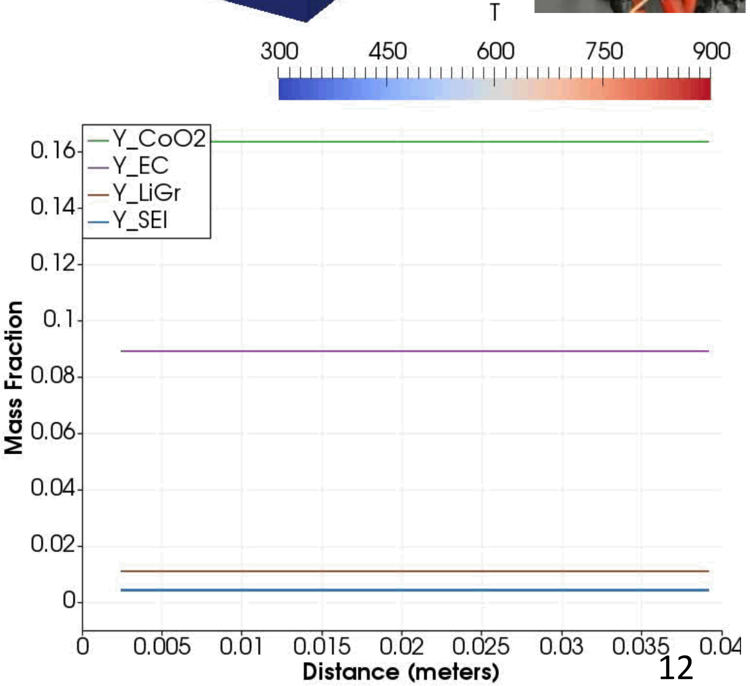


- Models can be used to estimate cooling requirements
  - Simulation shows homogeneous heating of 18650 cells (varying short resistance and cooling)
  - Internal temperature variation will be worse for large format systems and localized shorts

Cascading Propagation Observed in Li-Ion Packs



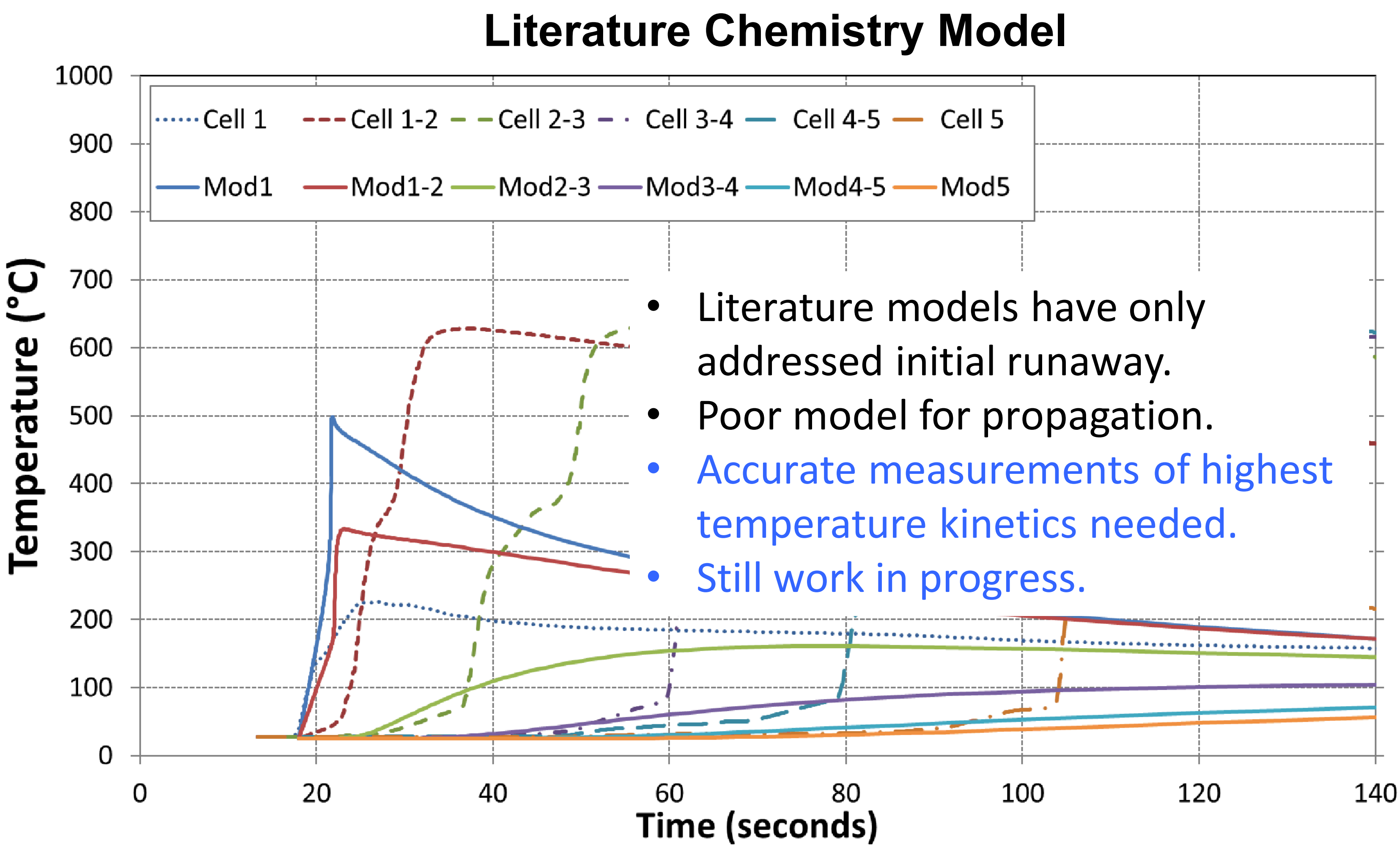




- Experimental propagation in 5 stacked pouch cells at Sandia
- Investigating effects of
  - State of charge
  - Intermediate layers
  - System geometry
- Good pack-scale model validation cases

Lamb, J., et al. (2015). Journal of Power Sources **283**: 517-523.

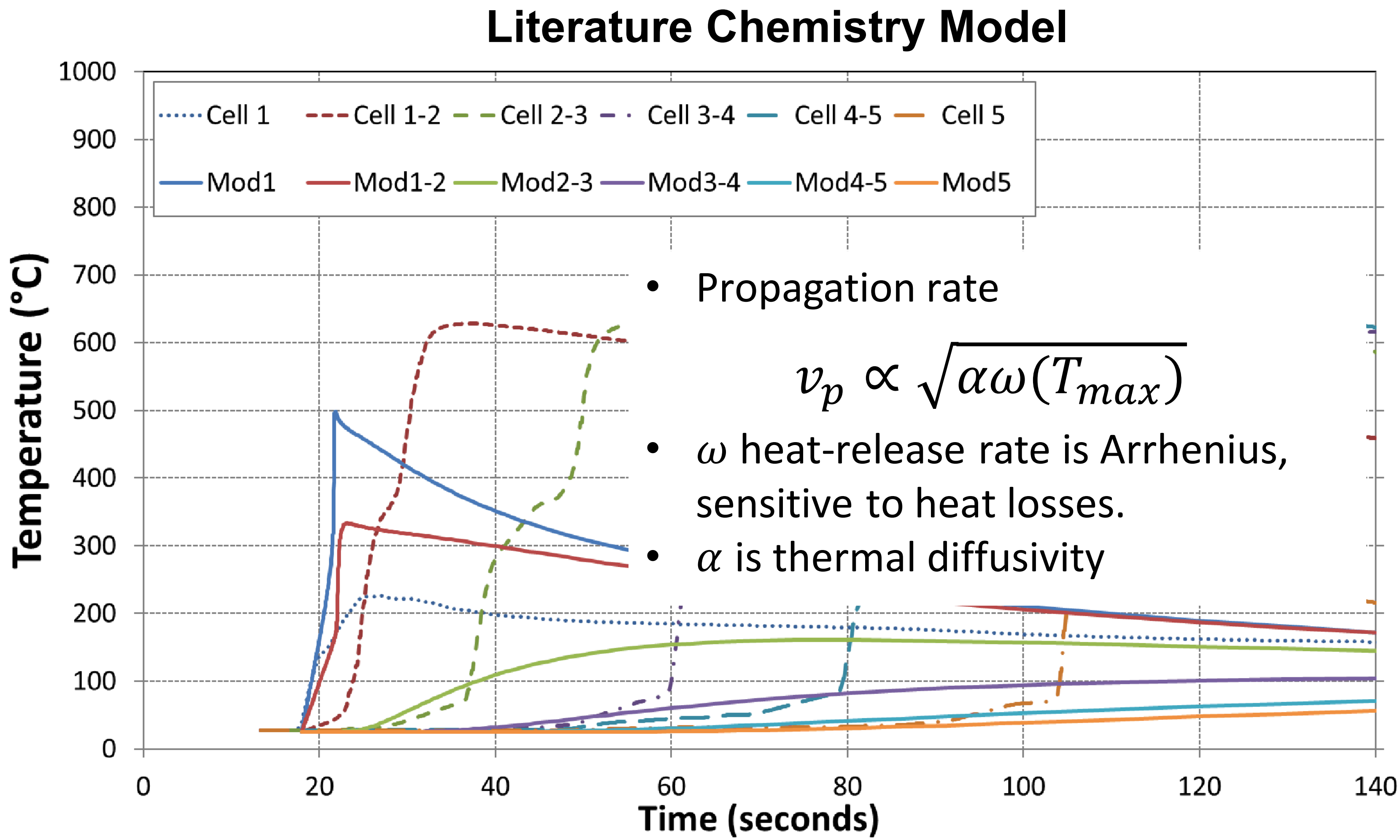
High-Fidelity Models Required for Cascading Failure



- Literature models have only addressed initial runaway.
- Poor model for propagation.
- Accurate measurements of highest temperature kinetics needed.
- Still work in progress.

Data from Lamb, J., et al. (2015). Journal of Power Sources **283**: 517-523.

High-Fidelity Models Required for Cascading Failure

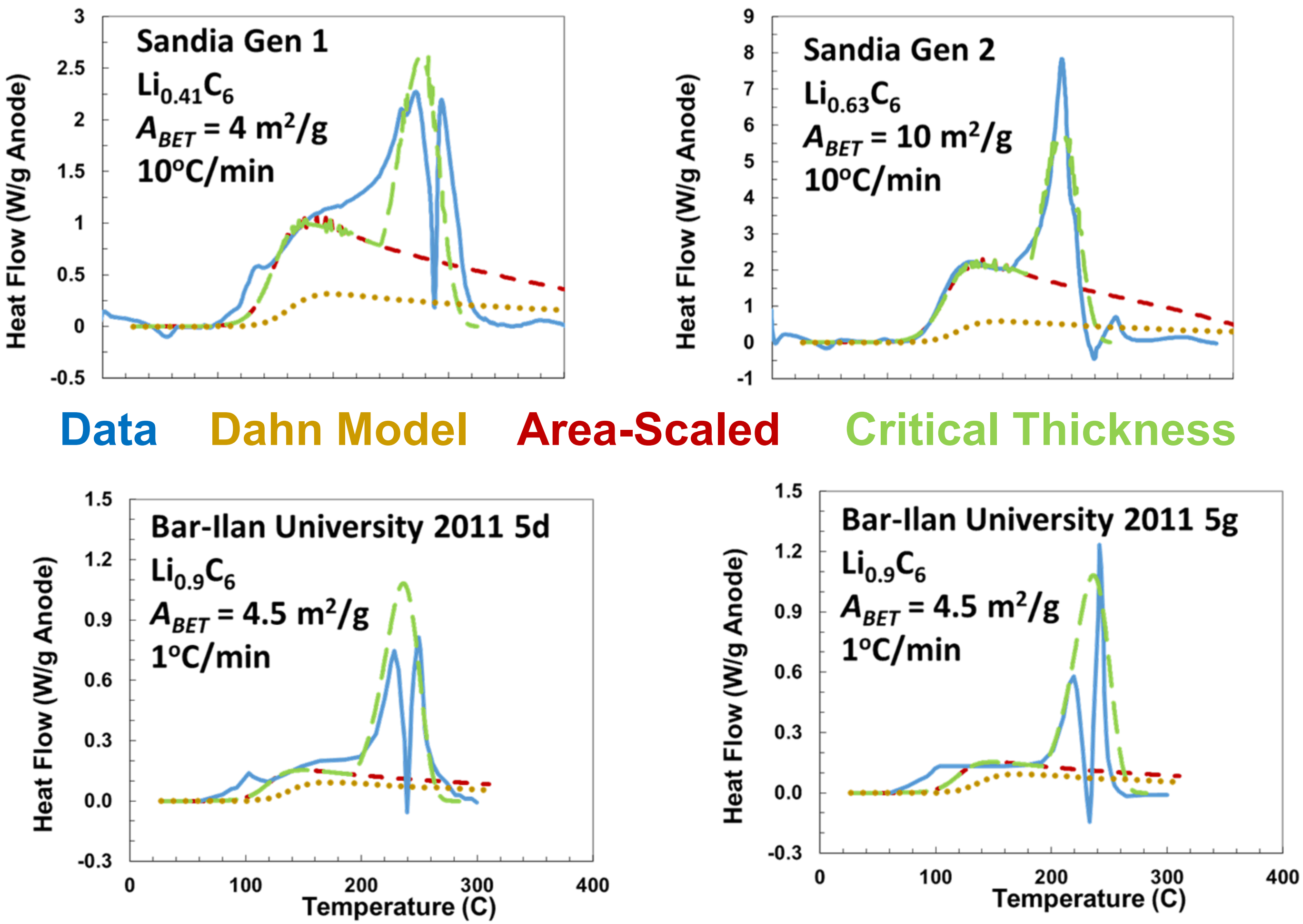


- Propagation rate

$$v_p \propto \sqrt{\alpha\omega(T_{max})}$$

- ω heat-release rate is Arrhenius, sensitive to heat losses.
- α is thermal diffusivity

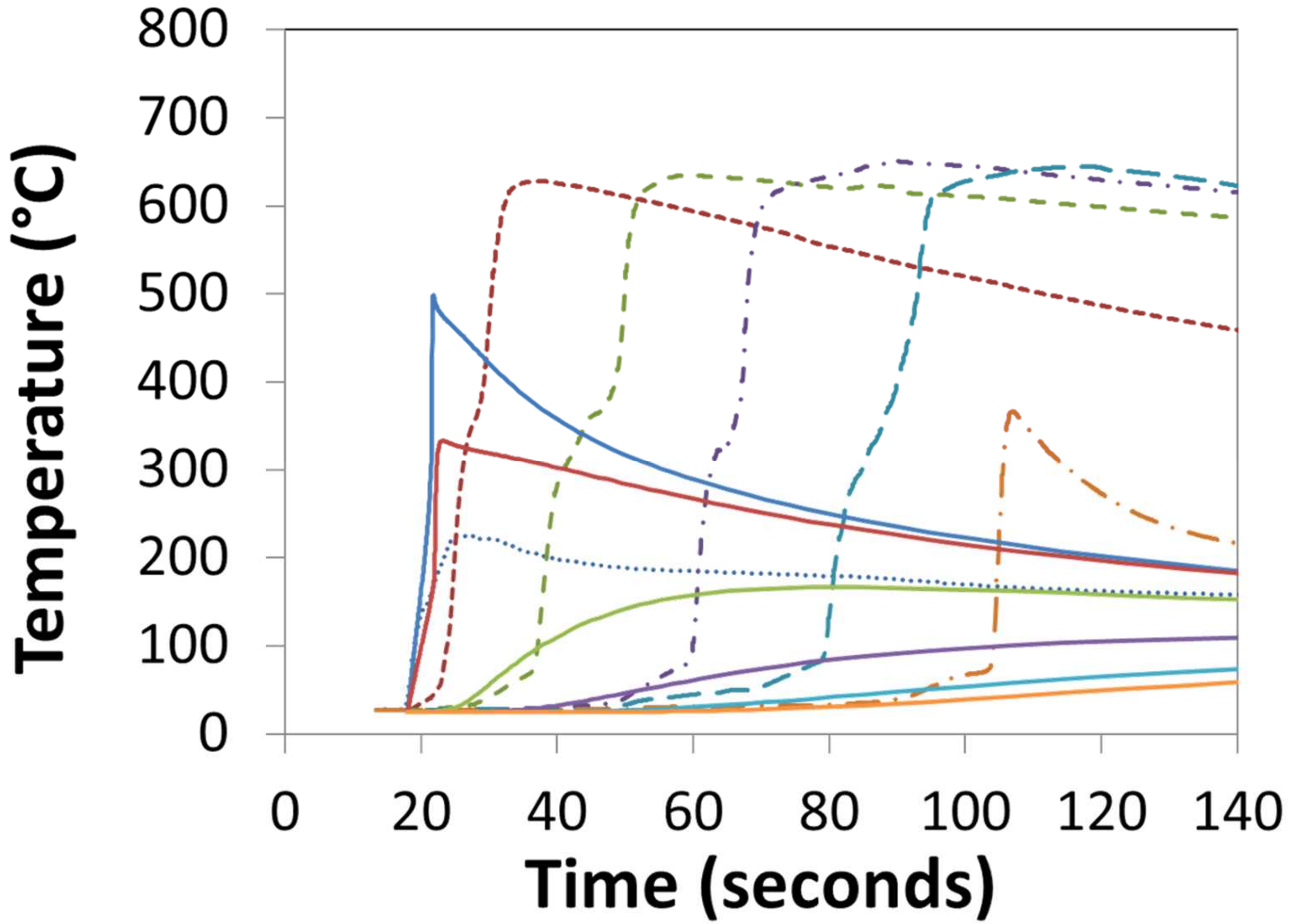
New model based on measureable quantities and thermodynamic material properties



Data from Lamb, J., et al. (2015). Journal of Power Sources **283**: 517-523.

Applying the new anode model to cascading failure

Dahn Model (Hatchard et al. 2001)



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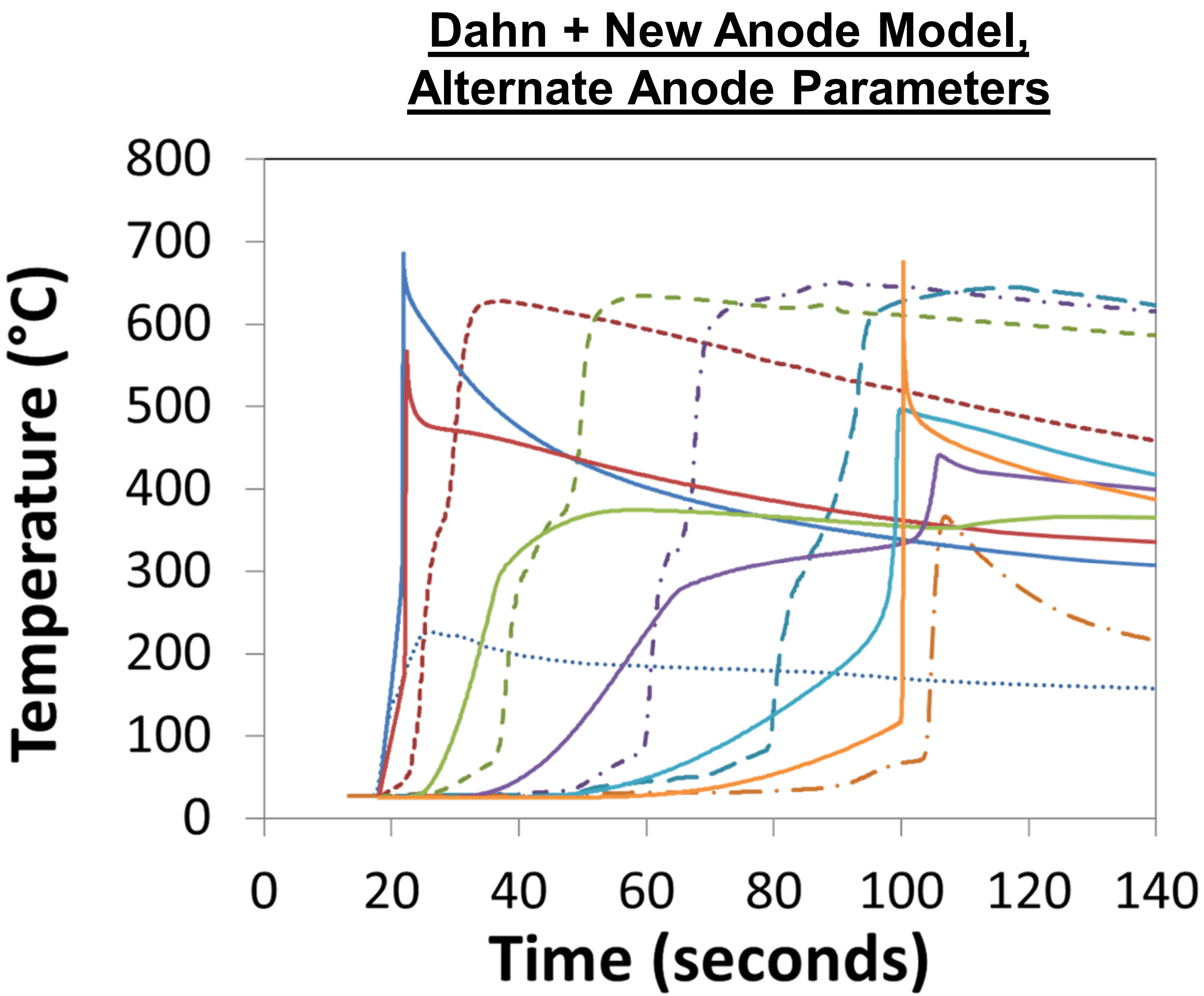
Applying the new anode model to cascading failure



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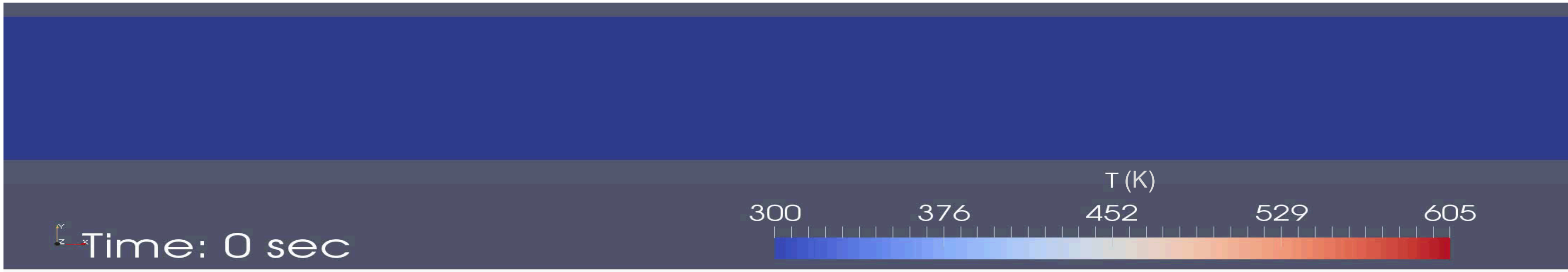
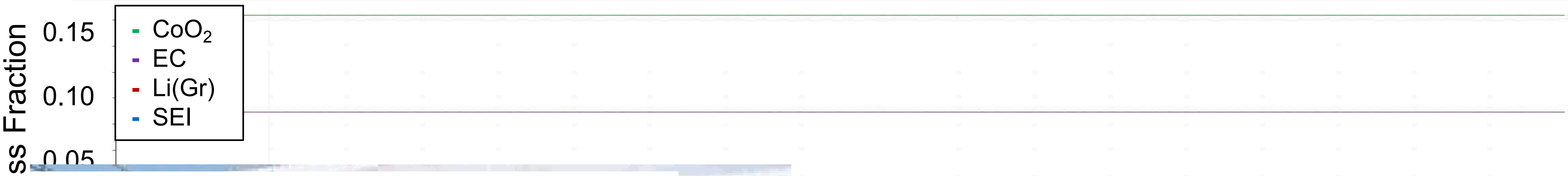
Extend modeling to Large Scales – Pulsating propagation





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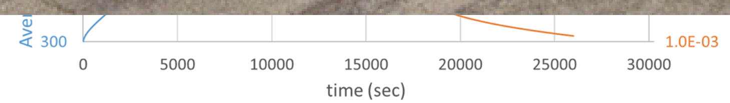
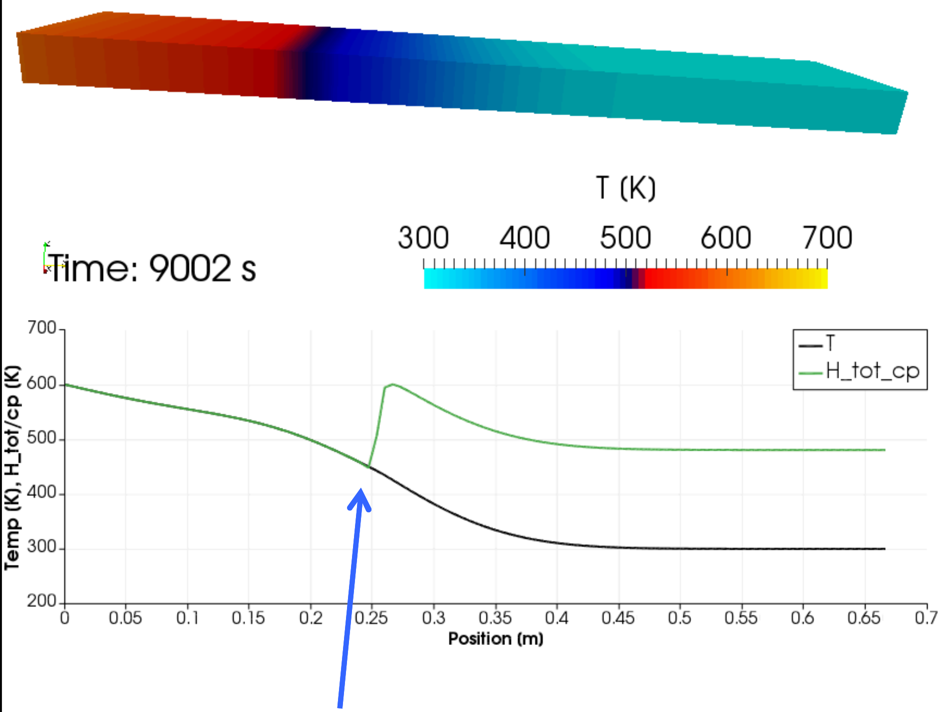
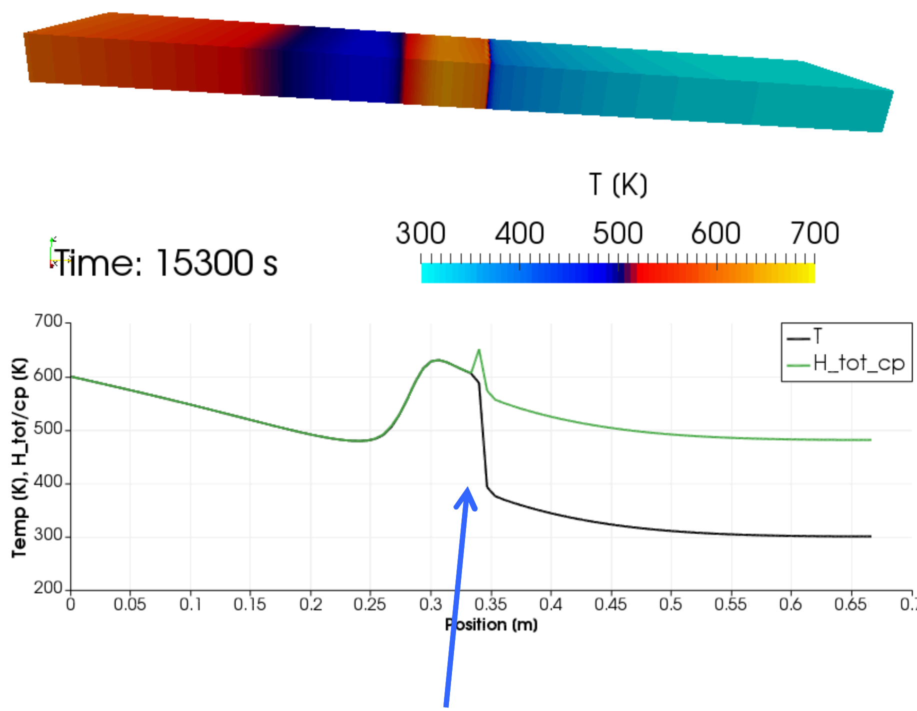


- Slow periods are best opportunity for cooling, but need to consider slow hidden reactions.

The mechanism of pulsating propagation

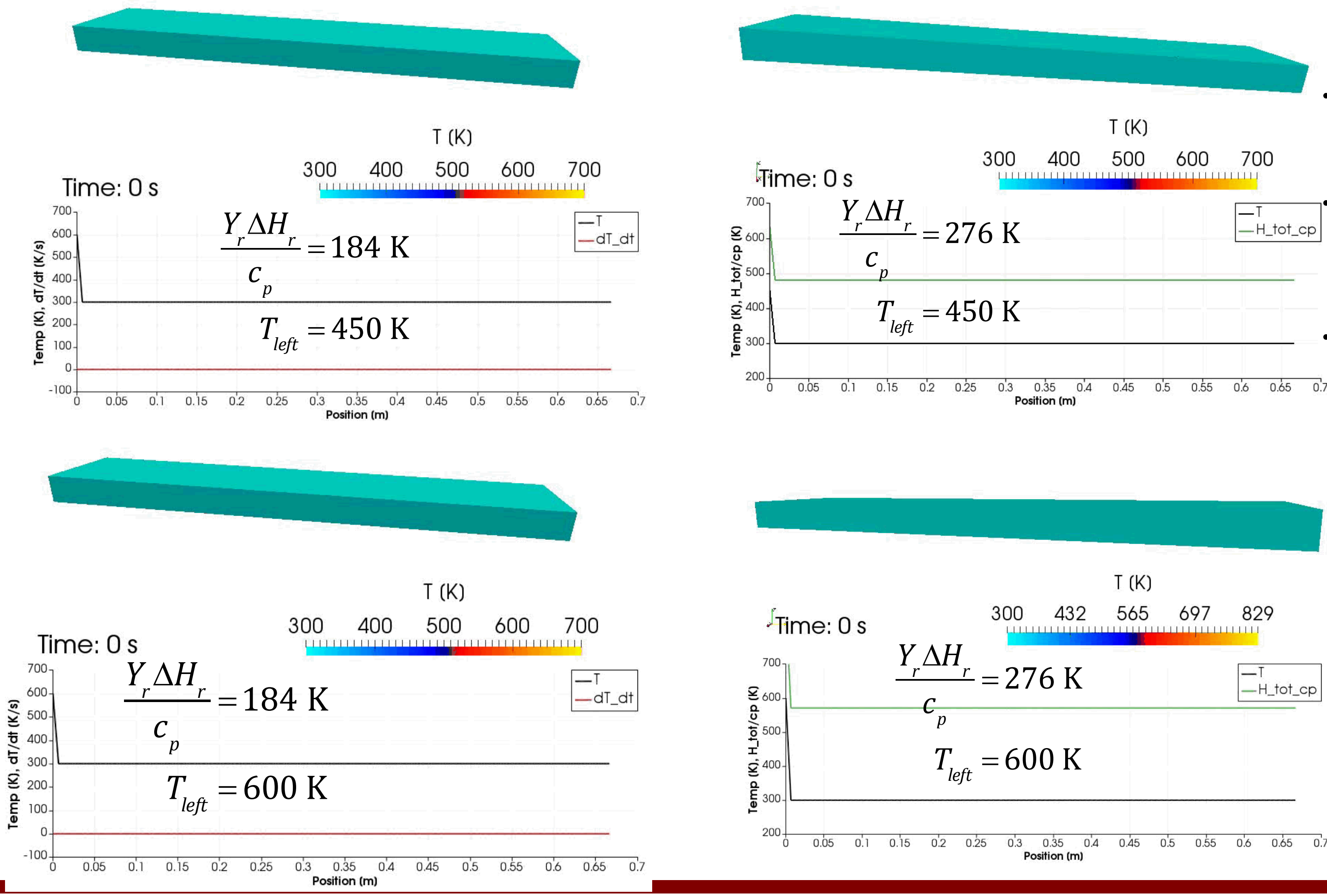
- Heat released is conducted upstream of reaction front, increasing the total enthalpy (sum of sensible and chemical enthalpy) $H_{TOT} = c_p T + Y_r \Delta H_r$
- Front propagates rapidly through preheated region with larger H_{TOT}.





- Slow propagation (low Temp), but preheating mixture ahead of reaction front.
- Rapid propagation (high Temp), into preheated mixture.

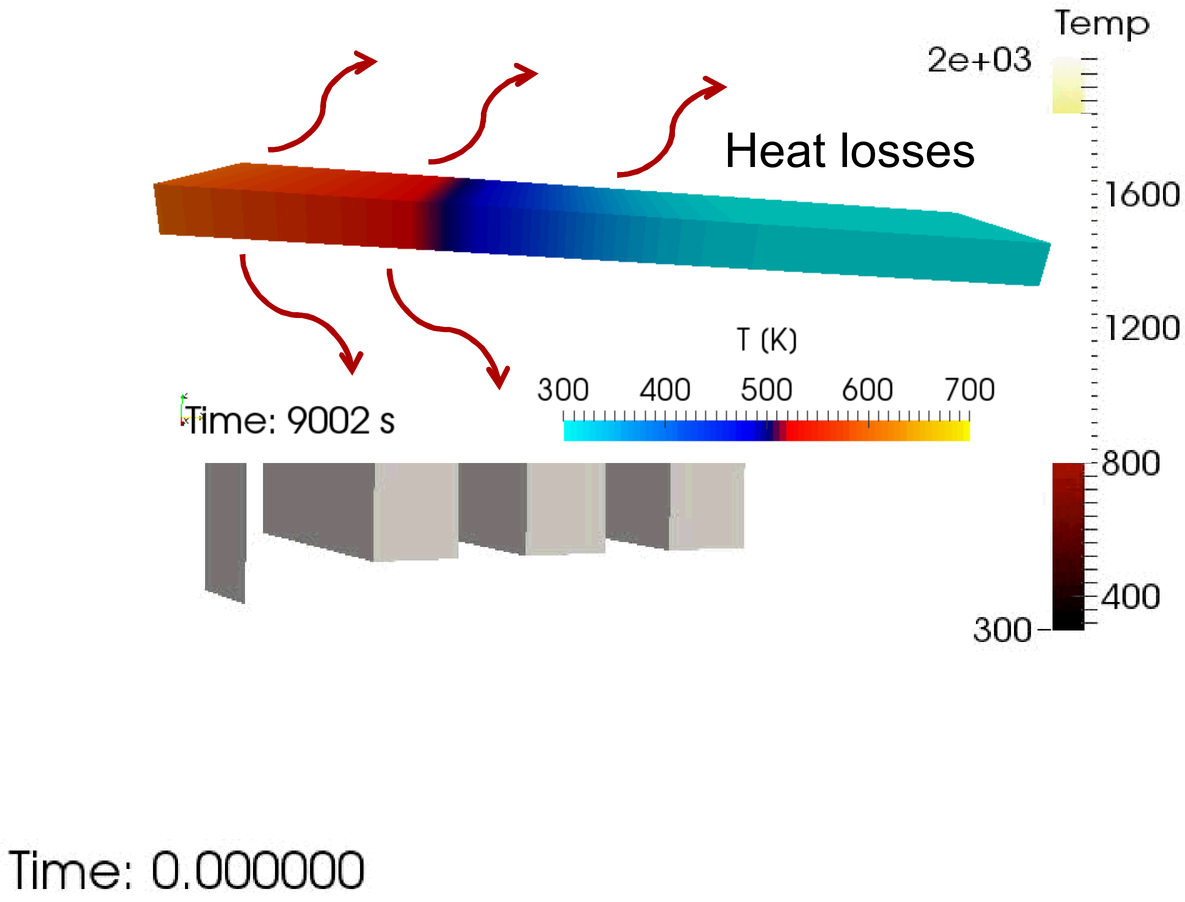
Parameter studies of propagation at large scales are possible with models



- Prediction and mitigation of cell-to-cell propagation is key to addressing risk.
- Single-step heat-release predictions with a range of heat release and boundary temps.
- Propagation across a large pack (80 cells here) exhibits pulsating instabilities.

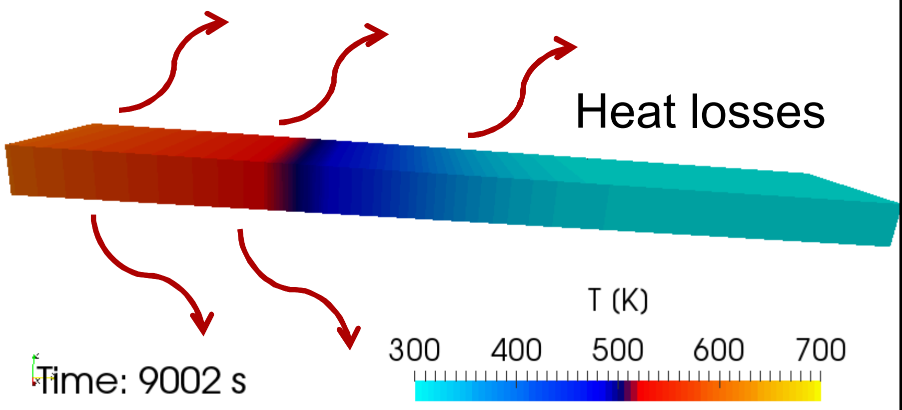
External risks to energy storage systems

- External fires:
  - Relatively low heat flux.
  - Installation integrated suppression.



Looking forward

- Thermal management can be an effective approach to managing thermal runaway risks.
- Heat capacity can inhibit ignition and propagation.
- Moderate cooling can quench propagation, and sometimes ignition.



- Simulations allow exploration of trade space if the physics are known.
  - But physics still must be observed and measured.
- → **Ultimate goal: *Predict criteria for cascading failure to act as a design tool in developing mitigation strategies.***

In closing

- Thermal runaway is a risk and potential barrier to development and acceptance.
- Heat release rates are moderate relative to potential dissipation.
- Multi-physics thermal models can potentially identify critical ignition and propagation trends.
- Quality measurements are key to parameter identification.
- Recent progress
  - Development of thermal source terms.
  - Identification of thermal ignition criterion.
  - Cell-to-cell propagation along homogenized pack structures.

Acknowledgements

- Supported by Imre Gyuk and the OE Electrical Energy Storage Program.

- Collaborative discussions with Heather Barkholtz, Lorraine Castro, Summer Ferreira, Josh Lamb, Chris Orendorff and Dave Ingersoll have been instrumental in understanding the range of possible topics that might be addressed.

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THANK YOU

QUESTIONS:

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High-Fidelity Models Required for Cascading Failure

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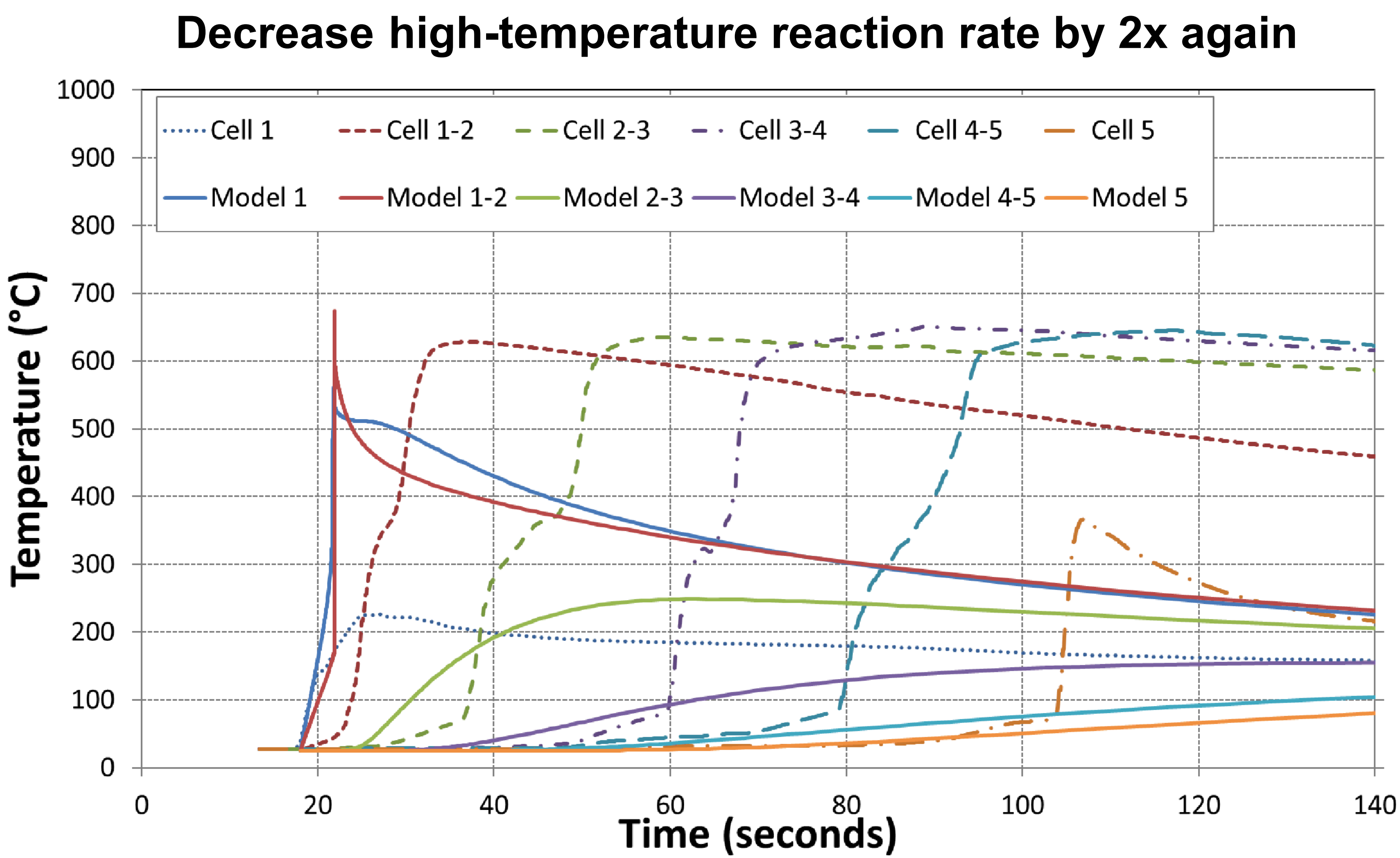

- Propagation predictions will improve with fidelity of high-temperature chemistry