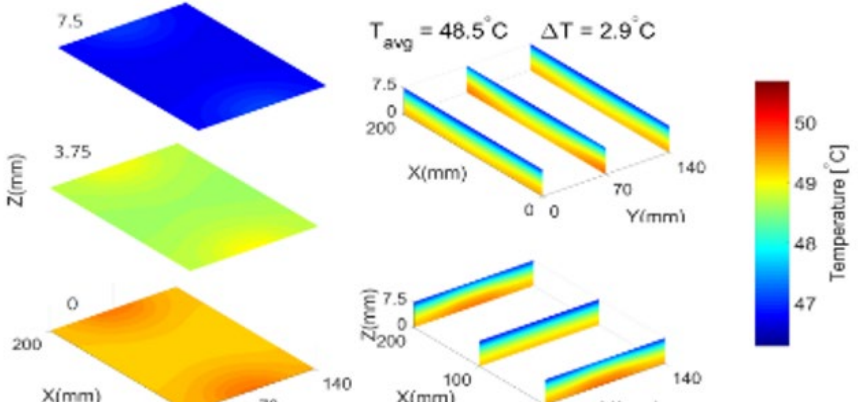
BAT459
U.S. DEPARTMENT OF ENERGY
VEHICLE TECHNOLOGIES OFFICE ANNUAL MERIT REVIEW

XCEL
eXtreme Fast Charge Cell Evaluation of Lithium-ion Batteries

HEAT GENERATION CONCERNS ASSOCIATED WITH EXTREME FAST CHARGING



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June 2021
Web Conference

OVERVIEW

Timeline

- Start: October 1, 2017
- End: September 30, 2021
- Percent Complete: 94%

Barriers

- Cell degradation during fast charge
- Low energy density and high cost of fast charge cells

Budget

- Funding for FY20 – $5.6M

Partners

- Argonne National Laboratory (ANL)
- Idaho National Laboratory (INL)
- Lawrence Berkeley National Laboratory (LBNL)
- National Renewable Energy Laboratory (NREL)
- SLAC National Accelerator Laboratory
- Oak Ridge National Laboratory (ORNL)

RELEVANCE — BATTERY THERMAL IMPLICATIONS

- Life, cost, performance, and safety of energy storage systems are strongly impacted by temperature.

Objectives of Heat Generation Thrust:

- Provide **feedback/solutions** to **industry** on the **battery thermal challenges** associated with extreme fast charging (XFC)
- Understand **temperature nonuniformity** within cell(s) during XFC
- Develop techniques for *in-operando* **interior temperature measurements**
- Identify limitations of using high specific energy density cells
- Identify **thermal areas of concern** with existing battery systems
- Identify how changes to the battery chemistry and cell design affect cells **efficiency** and **performance**
- Identify state-of-the-art thermal management strategies and how these can be applied to future battery electric vehicles

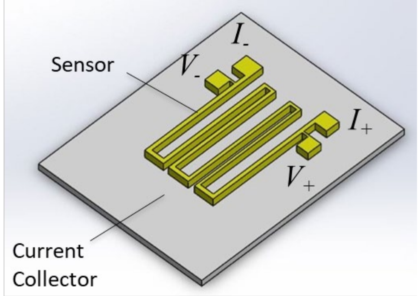
FY2020 MILESTONES

Milestone	Due Date	Status
Develop a model that provides thermal boundaries during charge and discharge for two scaled thermal management systems.	12/31/20	**Completed**
Model tradeoffs between elevated temperature fast charge benefit versus detriment to calendar/cycle lifetime. Define the critical parameters that affect heat generation within a cell.	12/31/20	**Completed**
Set up ANL facilities/fixtures to perform fast charging experiments at elevated temperatures and generate initial test results.	3/31/21	**Completed**
Incorporate electrochemistry into 3D cell model and elucidate differences between small test cells and large cells used in vehicles. Update 3D cell and module design strategies to suppress lithium (Li) plating, temperature, and aging heterogeneity.	9/30/21	On-track

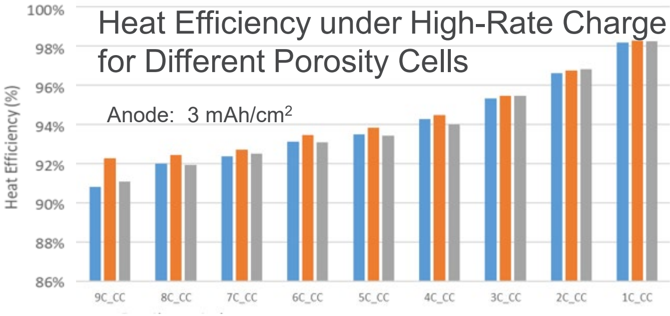
APPROACH — MEASURING HEAT GENERATION AND THERMAL TRANSPORT PROPERTIES FOR MODEL DEVELOPMENT

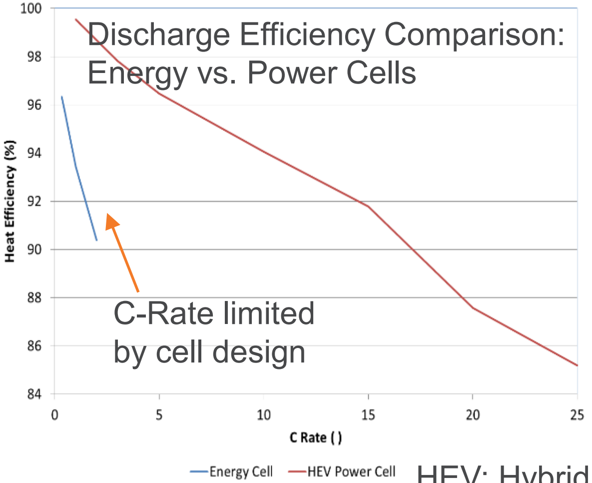
Identify critical parameters that affect heat generation in an electric vehicle (EV) cell.

Microcalorimeter: Heat Generation



Sensor for Spatially Resolved Heat Transport Properties

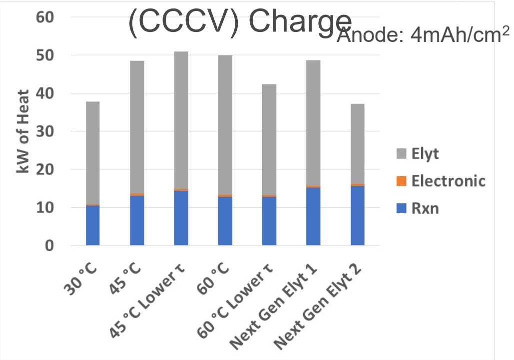


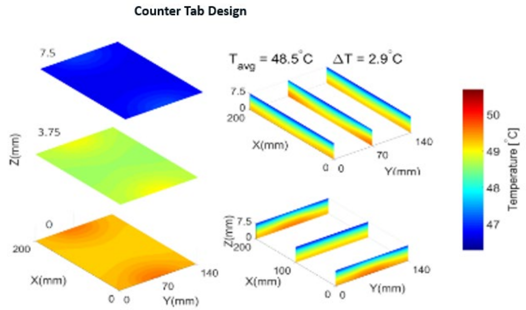


HEV: Hybrid Electric Vehicle

1D/3D Model Development

100 Kilowatt-Hour (kWh) Pack Under 6C Constant Current Constant Voltage (CCCV) Charge





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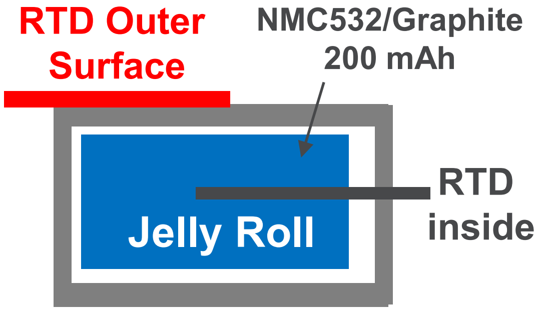
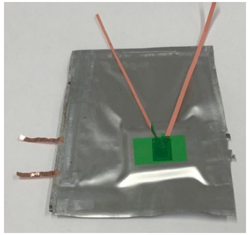
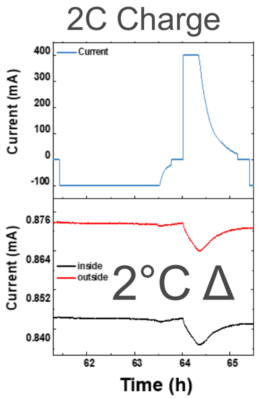
APPROACH: MEASURE AND UNDERSTAND TEMPERATURE VARIATION WITHIN EV CELLS

Benefit: Temperature inhomogeneity is often hypothesized to be a culprit in observed inhomogeneous degradation such as local Li plating, local state of charge (SOC) variation, and local solid electrolyte interphase (SEI) thickness variation. Measuring internal temperature will allow for correlation between hot spots and evidence of degradation.

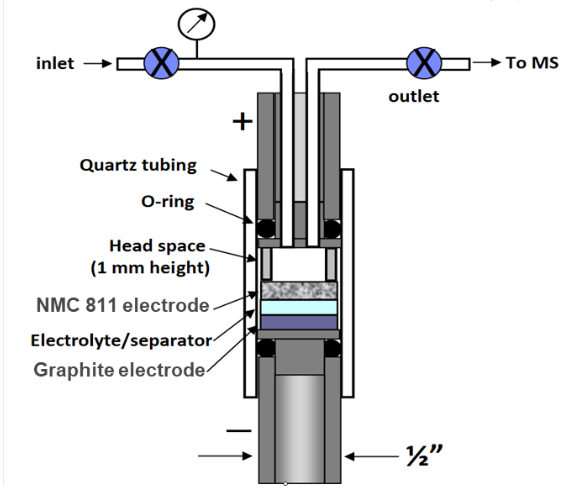
Resistance temperature detector (RTD) **stable** in electrolyte and under cycling



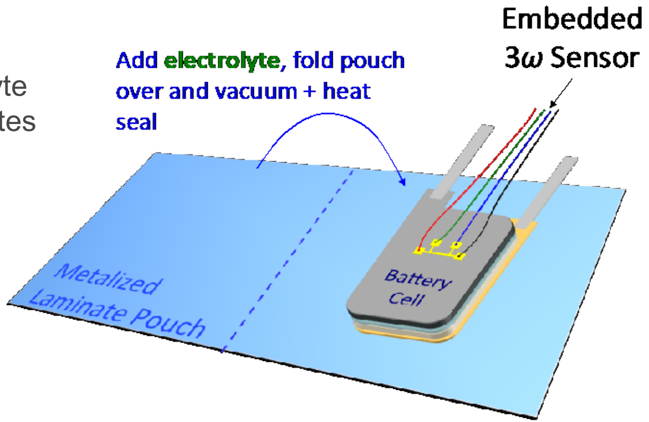




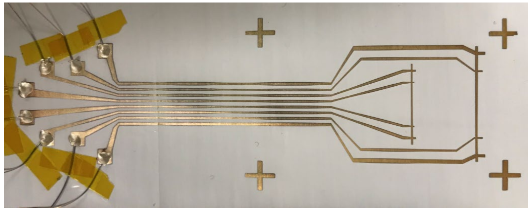
Determine the effect of temperature on electrolyte gassing behavior. Inform the design of electrolytes tailored to elevated temperature operation.



Above: Differential electrochemical mass spectrometry (DEMS) unit.

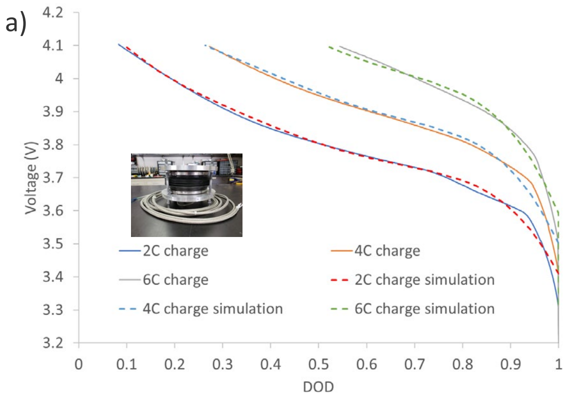


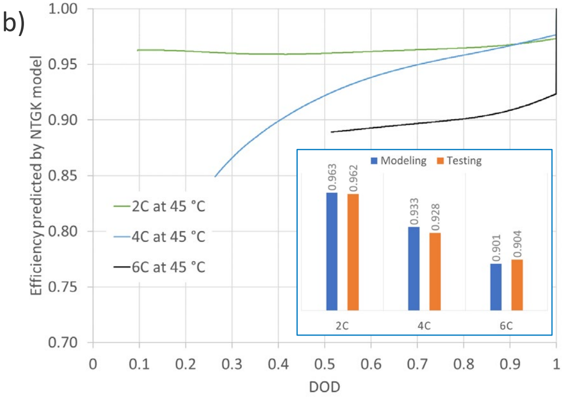
Sweep heating frequency to measure thermal transport properties at different distances from sensor. Used to assess internal temperatures.

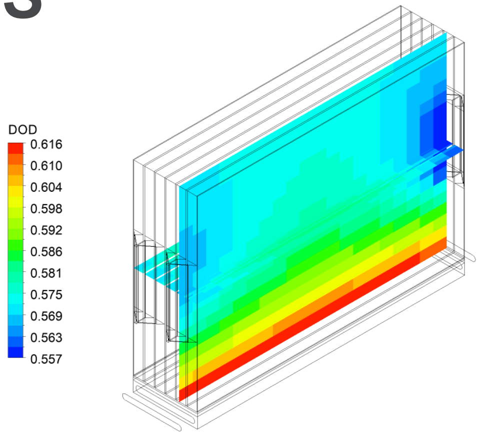


Prototype of exterior 3ω sensor.

3D MODELING OF LARGE-FORMAT CELLS





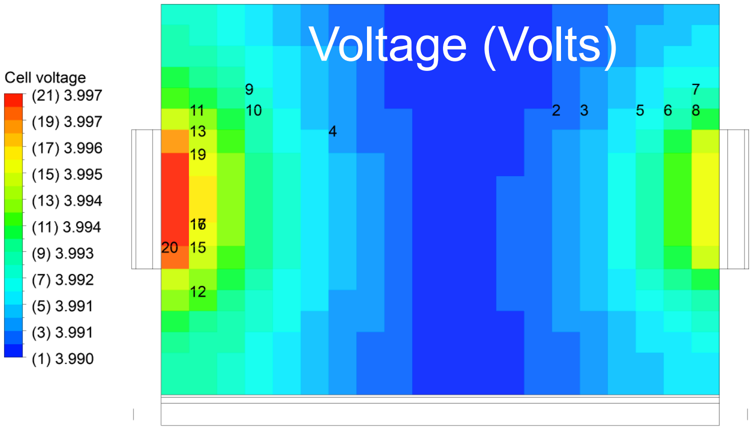


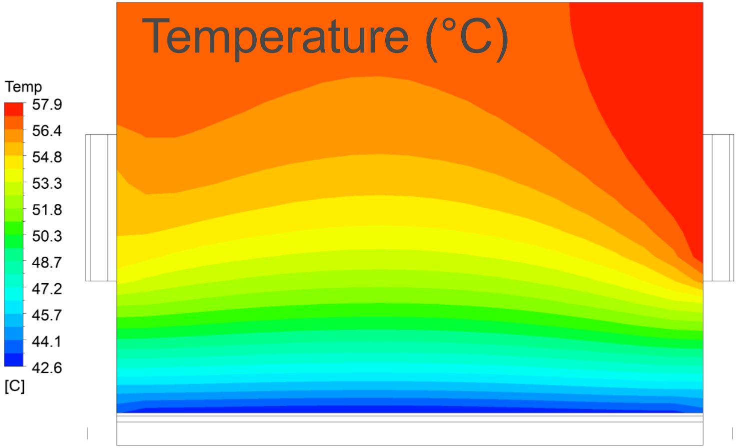
- An electrochemical-thermal model was created and validated with microcalorimeter testing of XCEL Round 2 32 milliamp hour (mAh) cells at 30°C and 45°C. Model charge capacity, voltage behavior, and charging energy efficiency match well with test results.

- 3D fast charging simulations utilizing CAEBAT MSMD software were performed for 25 Ah pouch cells scaled up from the Round 2 cell.

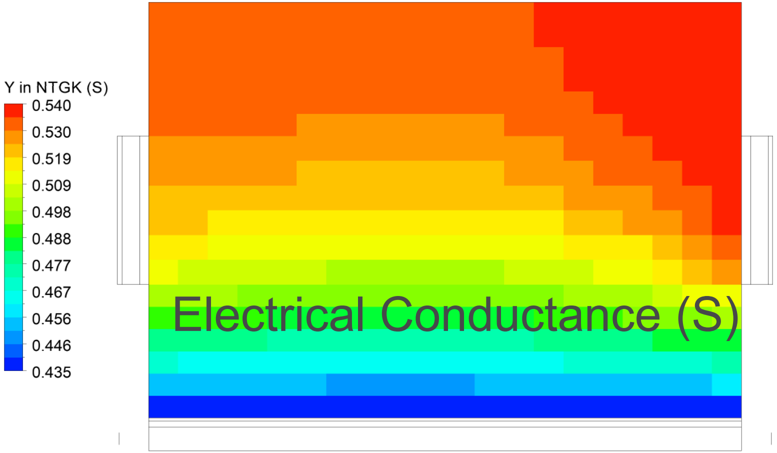
- The 3D model captures the heterogeneity caused by nonuniform electrical and thermal fields within a 3P2S module. The figure above shows the depth-of-discharge (DOD) heterogeneity associated with bottom cooling—as an example, the electrodes close to the tabs are being preferentially used/charged.

CAUSES OF HETEROGENEITY

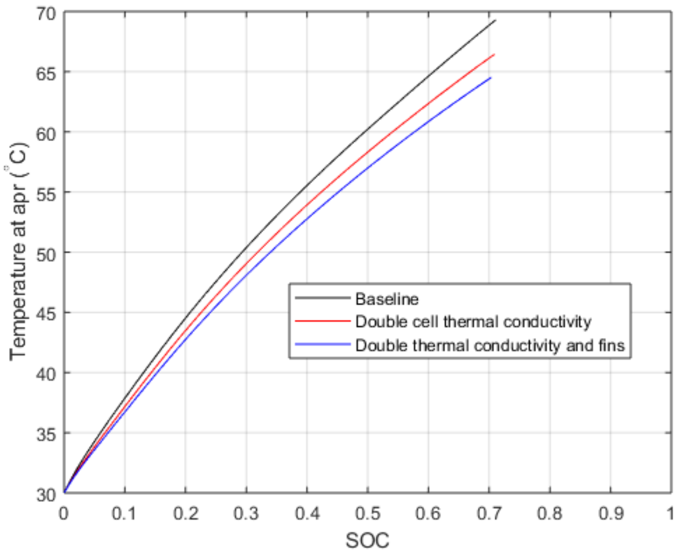


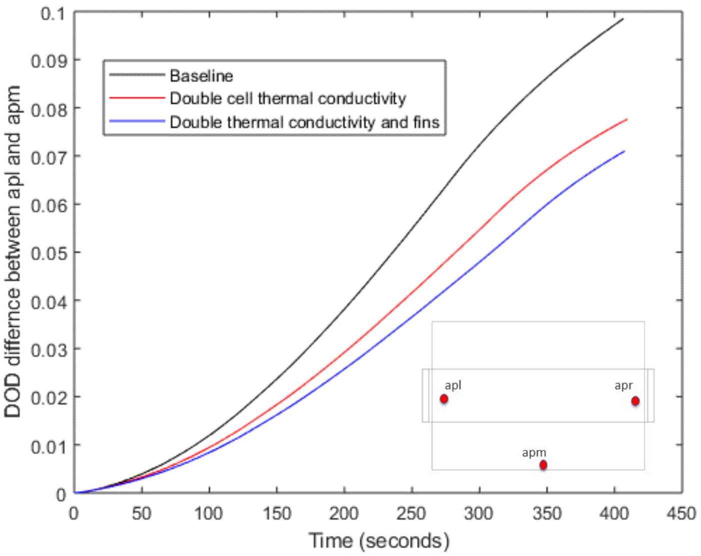


- Primary reasons for heterogeneity:
  - Higher current density near the tabs
  - Higher temperatures away from the cold plates leading to higher conductance of electrode.



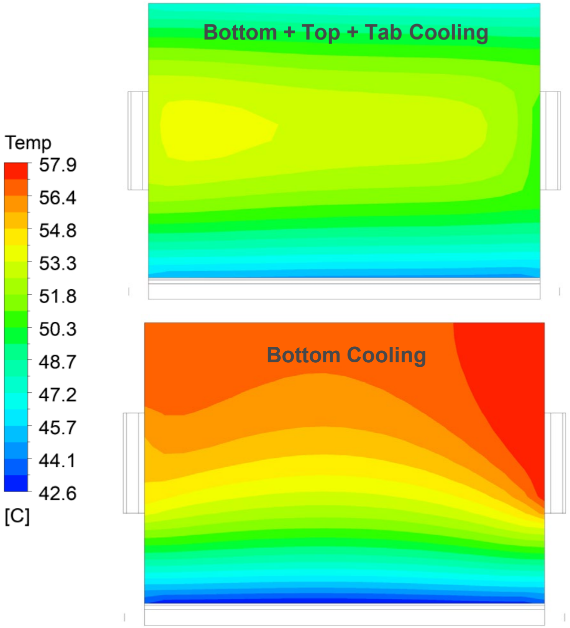
WHAT CAN BE DONE? ADJUSTING CELL THERMAL CONDUCTIVITY



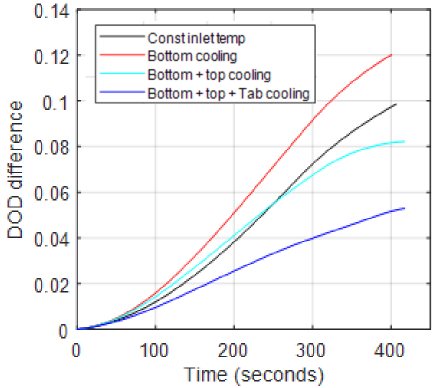


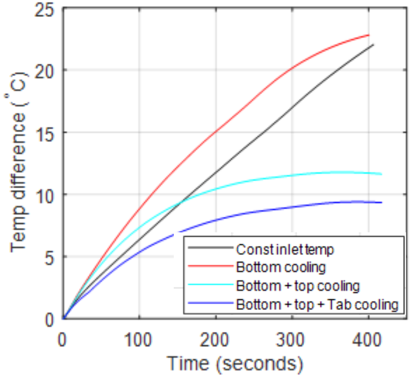
- Enhancing cell thermal conductivity and using thicker fins reduces module heterogeneity
- Heterogeneity is also affected by tab location, tab thickness, cell thickness and form factor, number of layers, and cooling options.

MITIGATION OF HETEROGENEITY BY MULTIPLE-SIDED COOLING



Temperature contours of a cell cross-section after 250 seconds under 6C charge





- Various cooling strategies analyzed for temperature heterogeneity in large EV cells:
  - Bottom cold plate + fin → 10% ΔSOC and 20°C ΔT (bad)
  - Bottom + top & tab cooling → 5% ΔSOC and 10°C ΔT (marginal)
  - Immersion + Bottom cooling → 5% ΔSOC and 5°C ΔT (ok)

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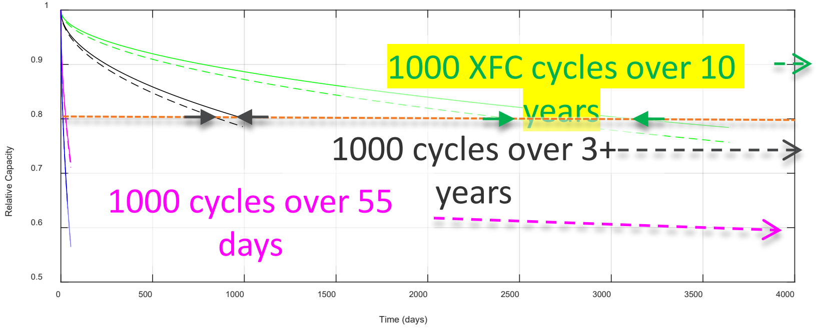
IMPACT OVER 10-YEAR LIFETIME SCENARIO—60°C CHARGING

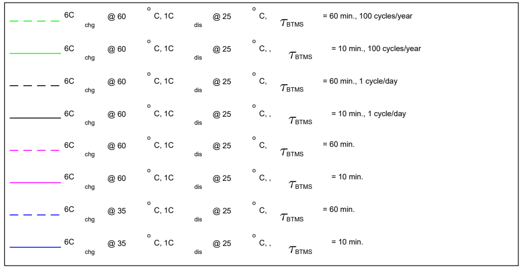
When 1,000 cycles are spread over 10 years, the benefit of rapid cooldown (extends calendar life) increases to 20% longer life

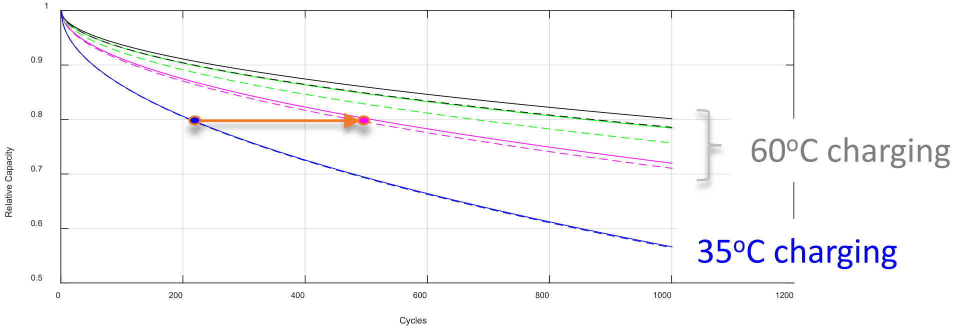


- 20% longer life
- 2.5% more capacity

- 17% longer life
- 1.5% more capacity

- 7% longer life
- 0.7% more capacity

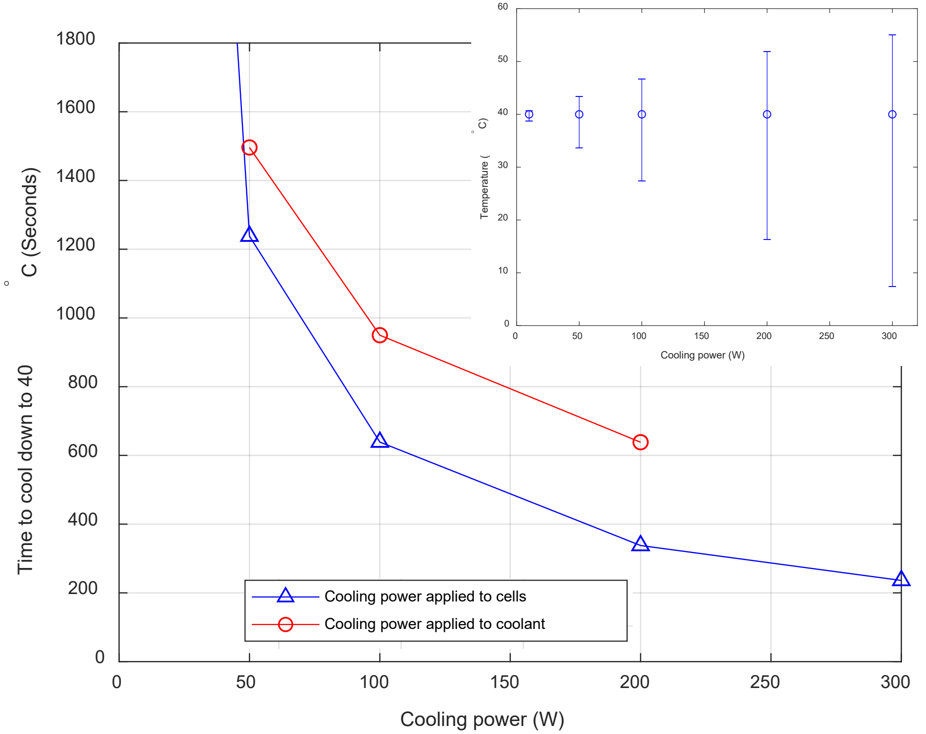




150% longer cycle life when XFC at 60°C vs. 35°C*

**Model accounts for cathode cracking and SEI/cathode electrolyte interface (CEI) growth, not Li plating. Li plating will also see benefits.*

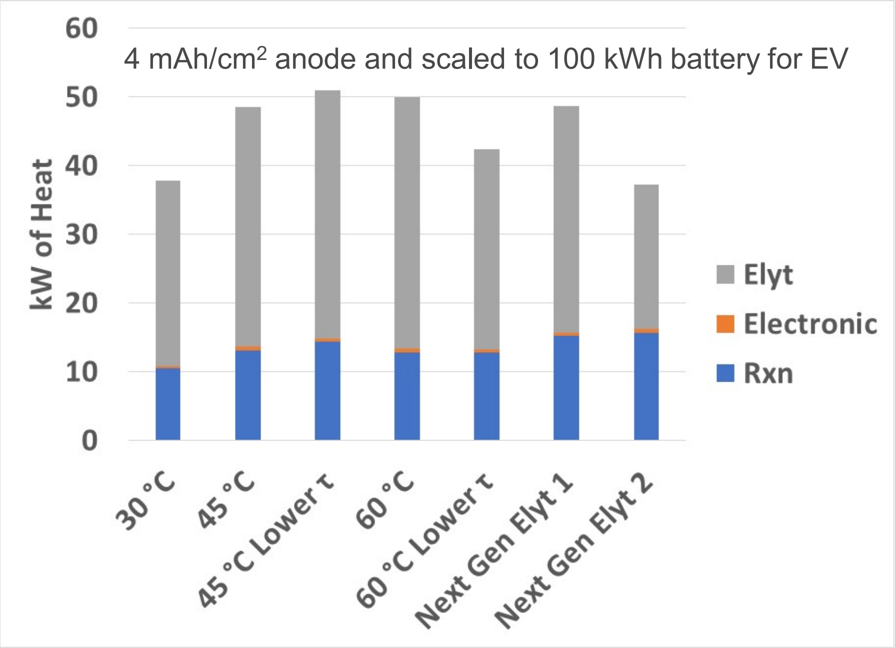
RAPID COOLING OF CELLS FROM 60°C TO 40°C

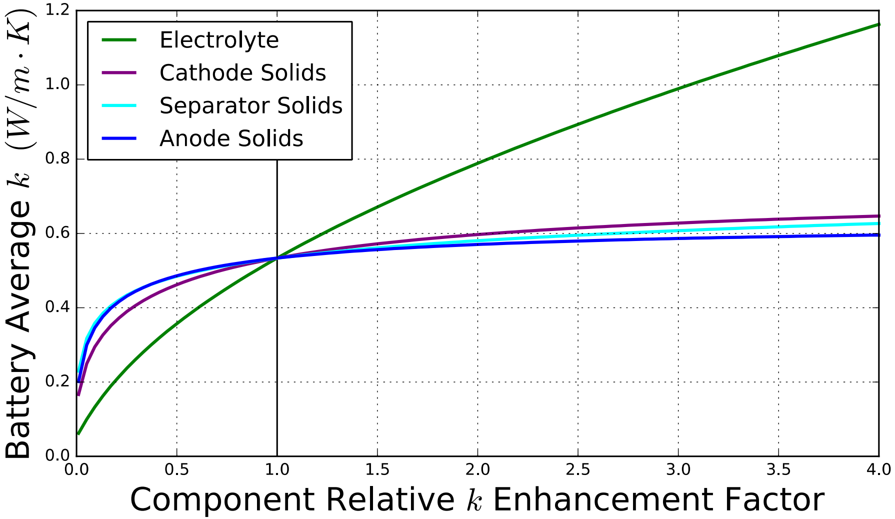


- For a Chevy Bolt size pack, it will require about 12 kW to cool the pack in 10 minutes when the cooling power is applied directly to the cells. The pack cooling capacity will need to double, 24 kW, if the heat goes through a bottom cold plate.
- With a bottom cold plate removing heat from cells, a large temperature difference will result with higher cooling capacities. This can be improved by optimizing heat dissipation paths.
- Cooling performance is affected by various design factors including cell thermal conductivity, thermal resistance along cooling pathway, and arrangement for surface cooling.

FY20 ANALYSIS SHOWED DOMINANT HEAT LOSS FROM ELECTROLYTE TRANSPORT

Two dominant losses are from electrolyte transport and then charge transfer reactions.



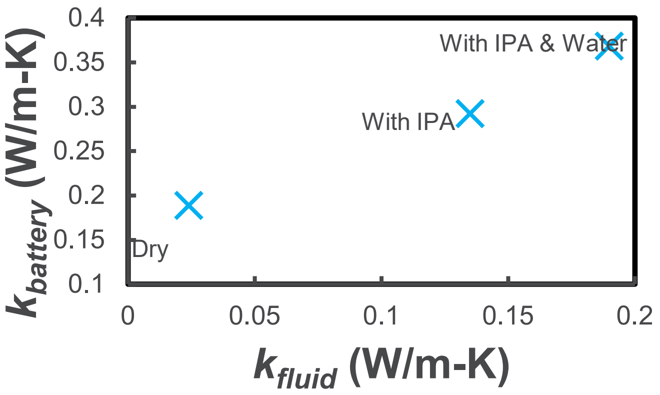


Lowering tortuosity (τ) and increasing temperature decreases heat generation.

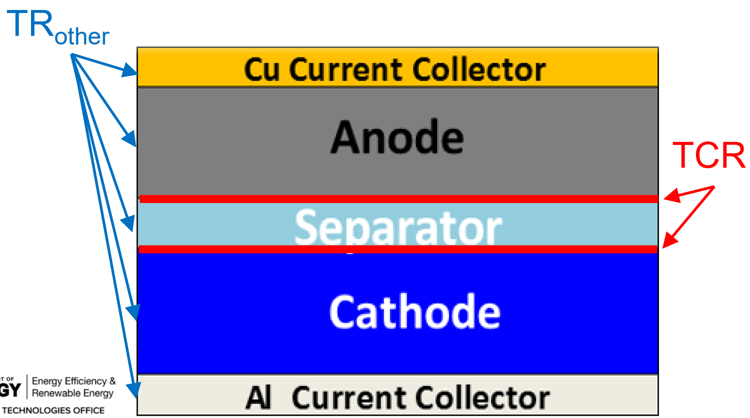
LBNL – Sean Lubner

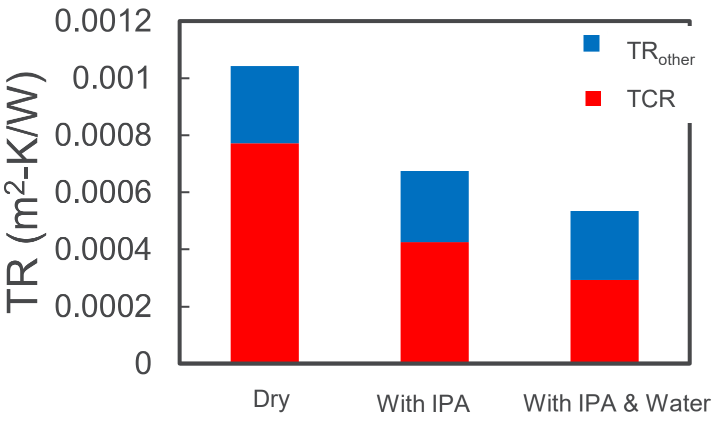
AFFECTING HEAT GENERATED IN ELECTROLYTE

k_{fluid} strongly affects the average battery thermal conductivity ($k_{battery}$)



Increasing k_{fluid} reduces both the thermal contact resistance (TCR) between layers, and the combined thermal resistance (TR_{other}) of battery layers themselves.





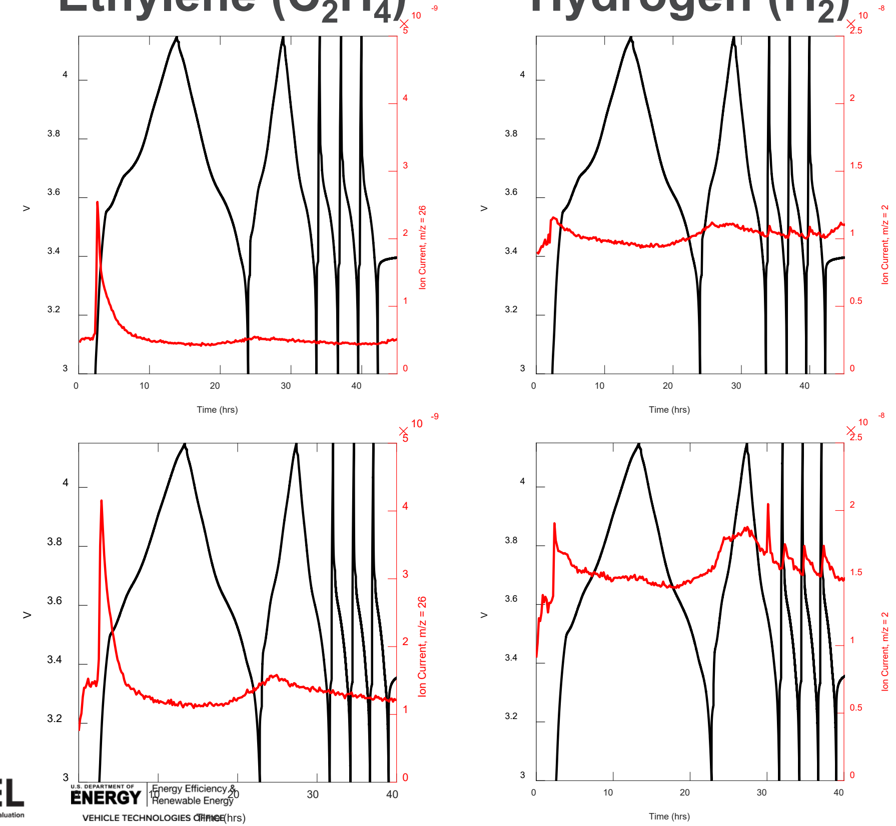
OPERANDO GAS MEASUREMENTS IN POUCH CELLS

Differential Electrochemical Mass Spectrometry (DEMS)

Ethylene (C_2H_4) Hydrogen (H_2)

23°C

40°C



Conditions

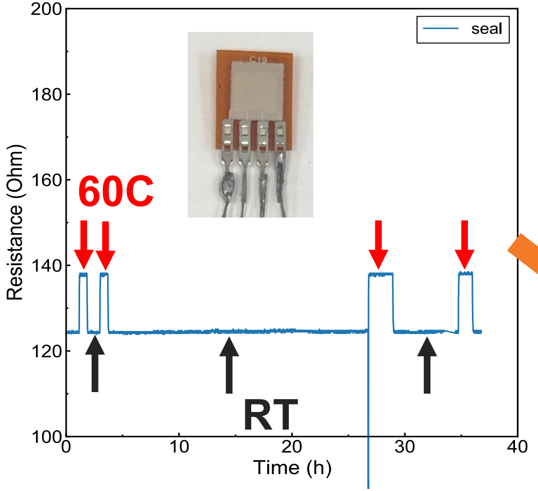
- Determine the effect of temperature on cell gassing behavior
- Inform the design of electrolytes tailored to elevated temperature operation.

Gen 2 Results

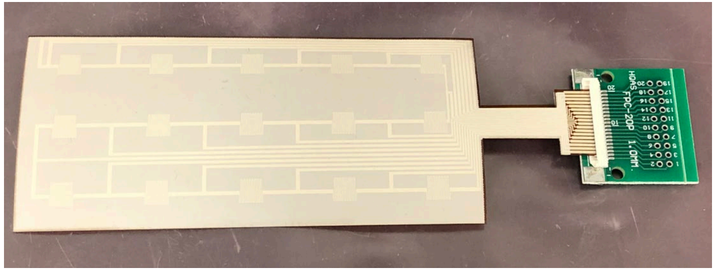
- Increased C_2H_4 evolution (from EC reduction) at 40°C
- Increased H_2 evolution (from thermal degradation of PF_6^-) at 40°C.

RTD (ARRAY) FOR XFC TEMPERATURE MEASUREMENT

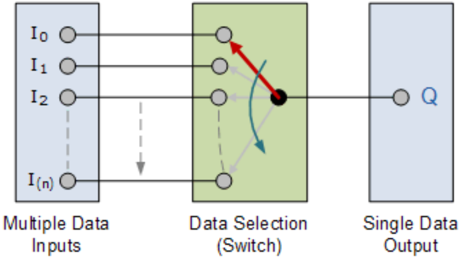
Single RTD - Stable in electrolyte



RTD Array

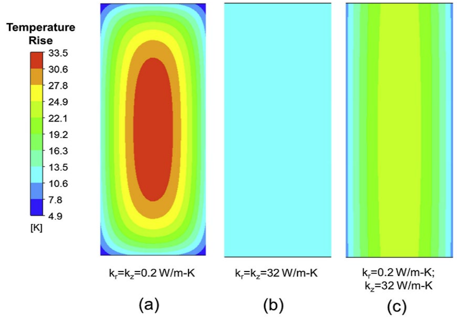


Circuit design



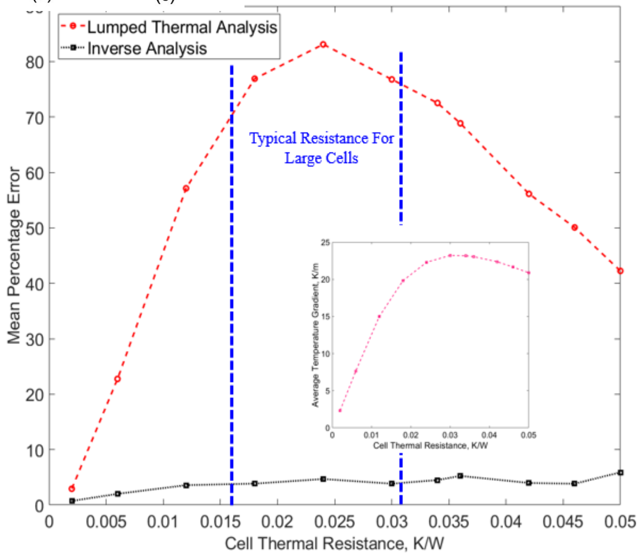
Incorporate RTD Array In Cell

INVERSE ALGORITHM MORE ACCURATE FOR TIME-RESOLVED CALORIMETRY OF THICK CELLS

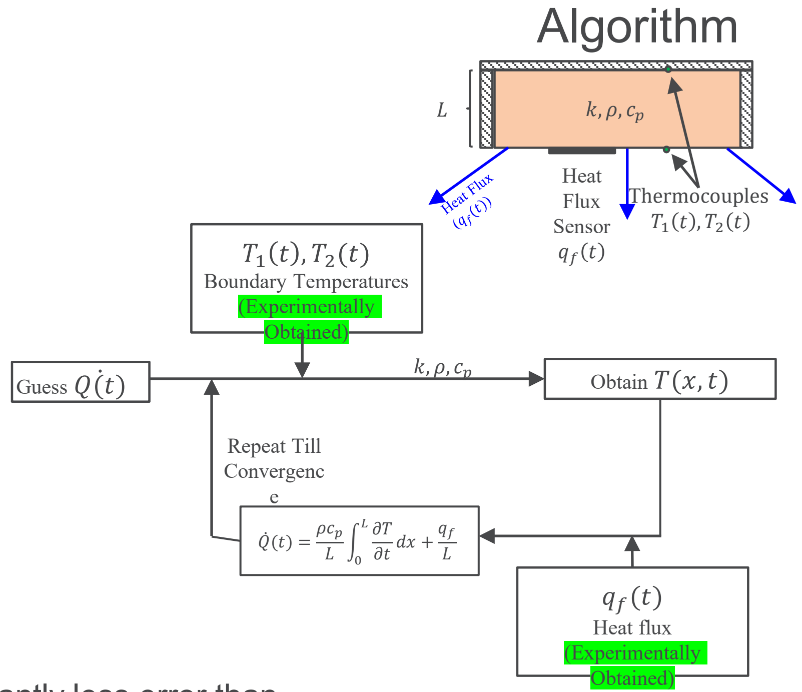


"Lumped" thermal analysis not valid for thick cells (because they are not isothermal)

S.J. Drake et al. *Journal of Power Sources* 252 (2014)



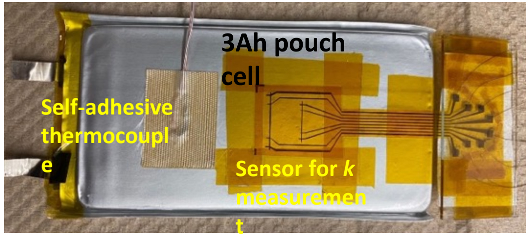
Significantly less error than standard lumped thermal analysis



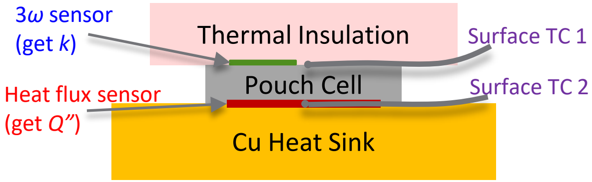
COMBINE COMPLEMENTARY SURFACE AND INTERNAL MEASUREMENTS FOR COMPLETE THERMAL PICTURE

NREL Calorimetry

LBNL - 3ω sensor

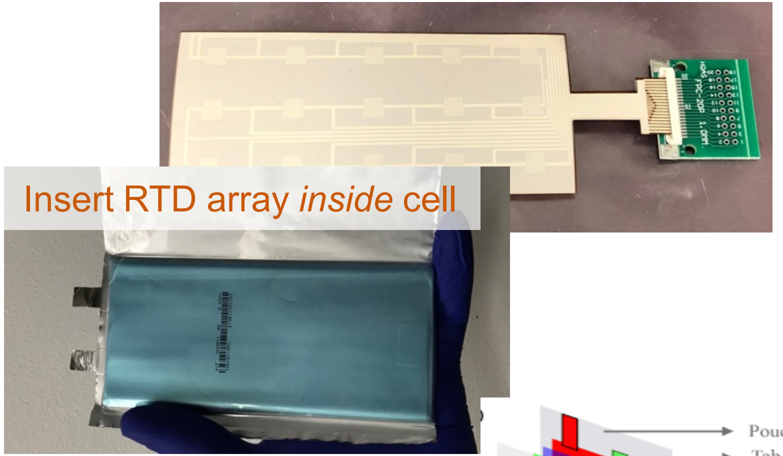


$$Q'' = k \frac{\Delta T}{L}$$

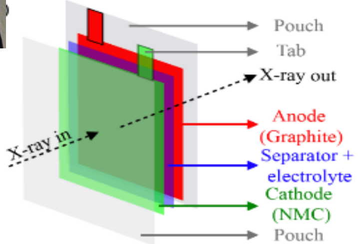


Measure: $k(x)$, $Q''(t)$, $T_{surface}(t)$
Calculate: $Q''(x, t)$, $T(x, t)$

Stanford



Measure: $T(x, t)$



XRD Temperature CU/NREL

Compare & Validate

RESPONSES TO PREVIOUS YEAR'S COMMENTS

Not reviewed during the previous AMR.

REMAINING CHALLENGES AND BARRIERS

- **Heat Generation**
  - Determine methods to reduce the heat produced from electrolyte transport and charge transfer reactions.
  - Fast charging at elevated temperatures limits Li plating and allows for the cell to be charged at higher efficiencies, however, life/degradation, gassing, and delamination concerns will have to be addressed.
  - Develop a high-fidelity, computational efficient, integrated 3D model capable of predicting Li plating and aging heterogeneity at cell level.
- **RTD**
  - Decrease RTD size
  - Determine reliable method to pass electrical feedthroughs into cells.
- **DEMS Experiment**
  - Novel electrolyte compositions may be too unstable at elevated temperature to acquire reliable DEMS data.
  - Possible that major interphase forming reactions for novel electrolyte compositions do not evolve gas.
- **Thermal Transport Experiments**
  - Validate inverse calorimetry algorithm on actual high-capacity cells.
  - Measure long term effects of XFC cycling on thermal transport properties.
  - Investigate ways to improve thermal transport within the electrolyte [related to but different from research into reducing heat generation in electrolyte.

PROPOSED FUTURE WORK

- **Adaptive Protocols**
  - Charge acceptance and aging as a function of temperature
  - Control to temperature difference—cell, module, pack
  - **Sensor input—3-omega, RTD**
  - **X-ray diagnostics** to test efficacy of the active/adaptive protocols
  - **Empirical and/or physics-based**
    - Use high fidelity model to adapt for various scenarios—**machine learning** for real-time adaptative protocols
- **Optimize the thermal properties of the cells**—Thermal conductivity, electrolyte thermal conductivity, etc.
- **Evaluate the thermal management problem in its entirety—charging station, pack, module, cell**
- **3D architectures addressing thermal transport in cell.**

SUMMARY

- Developed a 3D model electrochemical model to understand how to mitigate the temperature effects of fast charging
- Temperature can exacerbate voltage and DOD heterogeneities within a battery during extreme fast charging
- Investigated multiple cooling strategies for fast charge applications; bottom cooling, typically used in EV applications, can cause large variations in SOC and temperature
- XFC at 60°C extends the life by 150% as compared to charging at 35°C
- Rapid cooling of cells after XFC at high temperatures is beneficial to battery life, however, large on-board cooling systems will be required to rapid cool cells
- Modifying the thermal conductivity of electrolyte can positively affect heat removal in batteries
- A method has been developed to understand gas evolution from electrolytes at higher temperatures
- RTD array is being developed to monitor the internal temperatures of a cell
- Inverse calorimetry reduces error when predicting the internal temperature of a cell.

CONTRIBUTORS AND ACKNOWLEDGEMENTS

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COLLABORATION ACROSS LABS & UNIVERSITIES

Argonne NATIONAL LABORATORY — Cell and electrode design and building, performance characterization, post-test, cell and atomistic modeling, cost modeling

INL Idaho National Laboratory — Performance characterization, failure analysis, electrolyte modeling and characterization, Li detection, charging protocols

BERKELEY LAB — Li detection, electrode architecture, diagnostics

NREL NATIONAL RENEWABLE ENERGY LABORATORY — Thermal characterization, life modeling, micro and macroscale modeling, electrolyte modeling and characterization

OAK RIDGE National Laboratory — Detailed Li plating kinetic models, SEI modeling

SLAC NATIONAL ACCELERATOR LABORATORY — Li detection, novel separators, diagnostics

XCEL eXtreme Fast Charge Cell Evaluation of Lithium-ion Batteries

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