Prediction of Multi-physics Behaviors of Large Lithium-ion Batteries During Internal and External Short Circuit

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Physics of Li-Ion Battery Systems in Different Length Scales

**Atomic Scale**
- Thermodynamic properties
- Lattice stability
- Material-level kinetic barrier
- Transport properties

**Particle Scale**
- Li diffusion in solid phase
- Interface physics
- Particle deformation & fatigue
- Structural stability

**Electrode Scale**
- Charge balance and transport
- Electrical network in composite electrodes
- Li transport in electrolyte phase

**Cell Scale**
- Electronic potential & current distribution
- Heat generation and transfer
- Electrolyte wetting
- Pressure distribution

**Module Scale**
- Thermal/electrical inter-cell configuration
- Thermal management
- Safety control

**System Scale**
- System operating conditions
- Environmental conditions
- Control strategy

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**Charge Balance and Transport**
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**Pressure Distribution**
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- Pressure distribution

**Safety Control**
- Inter-cell configuration
- Thermal management
- Safety control

**Thermal Management**
- Inter-cell configuration
- Safety control

**Control Strategy**
- Inter-cell configuration
- Safety control
Thermal Runaway

Temperature ↔ Exothermic Reactions

- Temperature [K]
- Exothermic Reactions

T[K] vs t[sec]
Q[W] vs t[dcsec]

SEI
anode

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Modeling Thermal Runaway

✓ Constructed empirical reaction models using calorimetry data for component decompositions; Approach practiced by J. Dahn’s group
✓ Enhanced understanding of the interaction between heat transfer and exothermic abuse reaction propagation for a particular cell/module design, and
✓ provided insight on how thermal characteristics and conditions can impact safety events of lithium-ion batteries.

![Diagram showing internal and external temperatures, SEI decomposition, and total volumetric heat release from component reactions.](image)
Reaction Propagation

- Propagates Initially in azimuthal direction
- Forms hollow cylinder shape reaction zone
- Center axis zone starts to react
- Finally reaction goes further in outer radius cylinder zone

SEI decomposition reaction front
Runaway Propagation

In a multi-cell module

5 minutes apart between each frame
Closer Look at Reaction in an Individual Cell

2 seconds apart between each frame
Fast Heat Dissipation

✓ Small cell module: 20 x 18650
✓ Highly conductive carbon matrix wetted with phase change materials

Temperature  Heat of SEI decomposition
Developed an integrated model for multi-physics internal short circuit of lithium-ion cells by linking and integrating NREL’s unique electrochemical, electro-thermal, and abuse reaction kinetics models.

Performed 3D multi-physics internal short simulation study to characterize an internal short and its evolution over time.
Performed Case Study with A 20Ah Prismatic Cell

To investigate impacts of various short natures and cell characteristics

- Case Studies – 20Ah Stacked Prismatic Cell
  - ISC between metal (Al & Cu) current collector foils
  - ISC between electrode (cathode & anode) layers
    - Impact of short area – separator hole propagation
  - ISC between Al to anode – short bypassing cathode
  - Impact of Cell Size
    - Comments on Shutdown Separator
  - Impact of ISC location
Results Agree Well with Laboratory Observations

The simulation results have reasonably reproduced the experimental observations from other research groups/companies including SNL, Exponent, Celgard, LGchem, and Sony.
Shutdown Separator for Large Cells?

Short Between Al & Cu Metal Foils

- Cell Capacity: 20 Ah
- \( R_{\text{short}} \sim 10 \, \text{m} \Omega \)
- \( I_{\text{short}} \sim 300 \, \text{A} \) (15 C-rate)

\[
\begin{align*}
\text{Joule heat for short} & \quad \text{Temperature @10 sec after short} \\
\text{surface temperature} & \quad \text{internal temperature} \\
\text{20 sec} & \quad \text{30 sec} & \quad \text{40 sec} & \quad \text{50 sec} & \quad \text{60 sec} \\
T & \quad T & \quad T & \quad T & \quad T \\
Q & \quad Q & \quad Q & \quad Q & \quad Q \\
\end{align*}
\]

- 0.4 Ah
- \( R_{\text{short}} \sim 7 \, \text{m} \Omega \)
- \( I_{\text{short}} \sim 34 \, \text{A} \) (85 C-rate)

\[
\begin{align*}
\text{Surface temperature} & \quad \text{volume fraction} \\
\text{without shutdown} & \quad \text{shut-down functioned} \\
\end{align*}
\]
Shutdown Separator Limitation

- Thermally triggered
- Block the ion current in circuit

Difficult to apply in
- Large capacity system
- High voltage system
Ceramic Reinforced Separator

**Short Between Cathode and Anode Electrodes**

- Shorted area: 1 mm x 1 mm
  - \( R_{\text{short}} \approx 20 \, \Omega \)
  - \( I_{\text{short}} \approx 0.16 \, \text{A} \) (< 0.01 C-rate)

- Thermal signature of the short is hard to detect from the surface
- The short for simple separator puncture is not likely to lead to an immediate thermal runaway

- Maintaining structural integrity of separator seems critical to delay short evolution

- 3 cm x 3 cm separator hole propagation
  - \( R_{\text{short}} \approx 30 \, \text{m}\Omega \)
  - \( I_{\text{short}} \approx 100 \, \text{A} \) (5 C)

- Temperature at 1 min after short
External Short of Multi-Cell Battery

**Background**

- Cell **PTC** device proven effective control for over-current hazards at Li-Ion cell and small battery level
- Known as ineffective in high-voltage or large capacity battery designs
- Need to verify if NASA’s spacesuit battery design (16P-5S) array could depend on cell PTC devices to tolerate an external short
Commercial lithium ion 18650 cells typically have a current-limiting PTC (positive temperature coefficient) device installed in the cell cap to limit external currents in the event of an external short to the cell.

The PTC device consists of a matrix of crystalline polyethylene containing dispersed conductive particles, usually carbon black. The resistance of the PTC device increases sharply with temperature.

Once triggered, PTC behaves as a thermal regulator.

PTC device often fails to function in high voltage / high capacity systems.

PTC Resistance vs. Temp

Data: SRI
Model Development Approach

**Integrated Thermal and Electrical Network Model** of a Multicell Battery for Safety Evaluation of Module Design with PTC Devices during External Short

**Unit Cell Model**

**Multicell T&E Network Model**

**Electrical Model**

**5-Node Thermal Model**

**Electrical/thermal interaction**

**Thermal Network Model**

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Unit Cell Model – Electrical

ECM including PTC device

Jellyroll

PTC

PTC Resistance vs. Temp.

Data: SRI

National Renewable Energy Laboratory

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Unit Cell Model – 5-node Thermal

Detailed Cell Thermal Model
- Large computational requirement
- Not suitable for multicell modeling

5-Node Cell Thermal Model
- Low order dynamic model
- Suitable for multicell modeling

Comparison of Detailed and 5-Node Models for different heat generation conditions

Steady Form
\[ Q_i = \sum_j K_{ij} \left( T_i - T_j \right) \]

Unsteady Form
\[ Q_i = \sum_j K_{ij} \left( T_i - T_j \right) + MCp_i \frac{dT_i}{dt} \]
Multicell Network Model – Thermal

Thermal Network Model

**Thermal Mass**: Identifying thermal mass at each node

**Heat Generation**: PTC heat, discharge/charge heat (, abuse reaction heat)

**Heat Transfer**: Quantifying heat exchange among the nodes

\[
Q_{\text{transport}} = \sum_{j=1, j \neq i} Q_{ij}, \quad Q_{ij} = Q_{ij, \text{radiation}} + Q_{ij, \text{connector_conduction}} + Q_{ij, \text{convection}} \ldots
\]

**Cell-to-Cell Irradiative Heat Transfer**

**Transverse Heat Transfer Through Plates**

**Heat Conduction Through Air Gap**

**Heat Rejection Through Wires**

**Heat Transfer to Ambient**
16P model validated against a bank short test

- 10 mΩ external short
- Peak inrush current
- PTC device trip time
- Steady-state behavior
- Temperature rise profiles for all 16 cells
Simulation Results at Various Values of $R_{\text{short}}$

- $R_{\text{short}} \leq 40$ mΩ: PTC-limited
- $R_{\text{short}} \geq 50$ mΩ: SOC-limited

- Tripped PTC device serves as thermal regulator

$[dR_{\text{PTC}}/dT]_{130^\circ\text{C}} = 3 \; \Omega / ^\circ\text{C}$

(5 orders of magnitude > than at 25°C)

- Large pre-trip heat rates are safe provided that they have
  - Short duration
  - Sufficient thermal mass
  - Sufficient heat dissipation
11 nodes are vertically placed at 80 cell locations
Node thermal connections are defined considering various heat transfer modes
Aluminum enclosure box is considered thermally lumped
$11 \times 80 + 1 = 881$ node system
Model Validation for Pack External Short

ABSL experiment: Bank 3 short through external resistor

80 cell battery in test enclosure

10 mΩ resistor

Photo: ABSL

Photo: NASA
ABSL Instrumentation

Cell Temperature Sensor Locations

Brick Temperature Sensor Locations

Center-most cell

LLB-wall side

Circuit-board side

Bank 3

LLB wall

Photos: ABSL
Model Validation – First 6000 seconds

Symbols: ABSL test data
Lines: NREL model output

Temperature (°C) vs Time (s) graph showing:
- Center cell
- Edge cell
- GRP
- Al plate-c
- Al plate-p
- Corner cell
- Box

Temperature at Measured Spots:
- Center cell (blue)
- Edge cell (green)
- GRP (red)
- Al plate-c (purple)
- Al plate-p (pink)
- Corner cell (black)
- Box (yellow)

Symbols:
- ABSL test data: *
- NREL model output: +

Temperature range: 20°C to 120°C
Time range: 0s to 6000s
Cell Temperature Distribution at 6000 seconds

Height of cylinder depicts peak jellyroll temperature ($^\circ$C) of each cell as predicted by model.
E.g., bank 3 short is caused by foreign object between banks 3 and 4*

* Requires more than two faults: Introduction of foreign object debris & penetration of Kapton/Nomex/Kapton divider between banks
• Short runs through can of cell from adjacent bank 4
• Bare walls of cells are negatively biased
• Note that 3-layer (Kapton-Nomex-Kapton) bank-to-bank insulator is omitted for clarity
Bank 3 Short from 100% SOC

- Cell 42 (bank 3) participates in electrical discharge
- Cell 56 (bank 4) does not electrically discharge; its external can wall serves as a path for short current
- Model assumes ohmic heat of short shared equally by cells 42 and 56
- Internal-to-pack short more thermally severe than external-to-pack
- Thermal mass dominates – negligible dependence on Earth vs. space boundary conditions
- Runaway possibly prevented at 10 mΩ
- Runaway predicted at 20,30 mΩ with collateral damage

<table>
<thead>
<tr>
<th>$R_{\text{short}}$</th>
<th>Short Condition (SOC$_0$ = 100%)</th>
<th>Cell 42 $T_{\text{max}}$ (Bank 3)</th>
<th>Cell 56 $T_{\text{max}}$ (Bank 4)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 mΩ</td>
<td>External-to-pack, earth</td>
<td>97°C @ 6000-s</td>
<td>75°C @ 6000-s</td>
</tr>
<tr>
<td></td>
<td>Internal-to-pack, earth</td>
<td>150°C @ 16-s</td>
<td>146°C @ 16-s</td>
</tr>
<tr>
<td></td>
<td>Internal-to-pack, space</td>
<td>153°C @ 16-s</td>
<td>147°C @ 16-s</td>
</tr>
<tr>
<td>20 mΩ</td>
<td>Internal-to-pack, space</td>
<td>525°C @ 110-s</td>
<td>522°C @ 110-s</td>
</tr>
<tr>
<td>30 mΩ</td>
<td>Internal-to-pack, space</td>
<td>595°C @ 240-s</td>
<td>591°C @ 240-s</td>
</tr>
</tbody>
</table>
Bank 3 Short from 100% SOC: 10 mΩ vs. 20 mΩ

10 mΩ:
Bank 3 PTC devices trip quickly and uniformly because high inrush current causes PTC self-heating

Cell 42 PTC trips at 8 s

Remaining bank 3 PTC devices trip at 16 s

20 mΩ:
Bank 3 PTC devices trip slowly at different times, depending upon bank 3 temperature distribution

Cell 42 PTC trips at 10 s

Remaining bank 3 PTC devices trip between 60 s and 110 s
**Bank 3 short from 100% SOC: 10 mΩ vs. 20 mΩ**

**10 mΩ:**
Bank 3 PTC devices trip quickly and uniformly due to high in-rush current causing PTC self-heating

**20 mΩ:**
Bank 3 PTC devices trip slowly, at different times dependent upon bank 3 temperature distribution

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**Graph 1:**
- **Temperature (°C) vs. Time (s)**
- **All bank 3 PTC devices trip by 16 s**
- **Cell 42**
- **Rest of bank 3**

**Graph 2:**
- **Temperature (°C) vs. Time (s)**
- **All bank 3 PTC devices trip by 110 s**
- **Cell 42**
- **Rest of bank 3**
**Bank 3 Short from 100% SOC: Cell-to-Cell Radiation**

**Design question:**
Would a high-emissivity coating applied to bare cell walls help limit thermal excursion?

<table>
<thead>
<tr>
<th>$R_{\text{short}}$</th>
<th>Short Condition (SOC$_{0} = 100%$)</th>
<th>Cell wall emissivity</th>
<th>Cell 42 $T_{\text{max}}$ (Bank 3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 mΩ</td>
<td>Internal-to-pack, earth</td>
<td>$\varepsilon = 0.3$ (Nominal)</td>
<td>525°C @ 110 s</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\varepsilon = 0.9$ (Coating)</td>
<td>410°C @ 102 s</td>
</tr>
</tbody>
</table>

(Insufficient impact)
Is battery design tolerant to pack-internal shorts when stored at low SOCs?

<table>
<thead>
<tr>
<th>$R_{\text{short}}$</th>
<th>Short Condition</th>
<th>Initial SOC</th>
<th>Initial OCV</th>
<th>$\text{Cell 42 } T_{\text{max}}$ (Bank 3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 mΩ</td>
<td>Internal-to-pack, earth</td>
<td>1.5%</td>
<td>3.428 V</td>
<td>117°C @ 85 s</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.5%</td>
<td>3.346 V</td>
<td>83°C @ 80 s</td>
</tr>
</tbody>
</table>

No thermal runaway when stored at 0% SOC (3.25 OCV).
Summary

- NREL performed an internal short model simulation study to characterize an internal short and its evolution over time by linking and integrating NREL’s electrochemical cell, electro-thermal, and abuse reaction kinetics models.

- Initial heating pattern at short events depends on nature of short, cell characteristics, and system configuration.

- Temperature rise for short is localized in large capacity cells.

- Short current is carried mostly by metal collectors.

- A simple puncture in the separator is not likely to lead to an immediate thermal runaway of a cell.

- Maintaining the integrity of the separator seems critical to delay short evolution.

- PTC device is an effective thermal regulator. Maximum cell temperature (final state) is very similar for a variety of initial and boundary conditions.
Summary

• 80-cell spacesuit battery electrical/thermal model
  • Captures relevant physics for cell-external shorting events, including PTC behavior
  • Agrees well with pack-external bank 3 short experiment run by ABSL
  • Predicts that design will tolerate all pack-external short resistance conditions

• Relocating short from pack-external (experimental validation) to pack-internal (modeling study) causes substantial additional heating of cells that can lead to cell thermal runaway
  • Negligible sensitivity to earth/space boundary conditions (thermal mass dominates)
  • Large sensitivity to $R_{\text{short}}$
    • $R_{\text{short}} < 10$ mΩ: 16P bank PTC devices trip quickly, most likely preventing runaway
    • $10$ mΩ < $R_{\text{short}}$ < 60 mΩ: Thermal runaway appears likely
  • Nevertheless, this finding re-emphasizes the general imperative of battery pack assembly cleanliness

• Design is tolerant to pack-internal short when stored at 0% SOC
Acknowledgments

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• Brad Strangways

ABSL
• Craig Flora

Thank you for your attention!
Multi-physics design and analysis paves the road for future automotive batteries

Designing Li-Ion cells and modules using computer aided design and engineering tools to

- Reduce the process of product design, build, and test cycle.
- Accelerate product development cycle to reduce battery cost.

The goal is to use state of the art battery modeling tools and codes developed by NREL, universities, National Labs, battery companies and others in an integrated system for universal use.

The requirements for lithium-ion batteries for next generation electrified vehicles must be addressed over various length and time scales in which physical and chemical processes are occurring—from atomic variations to vehicle interface controls.

Integrated multi-scale models need to provide a pathway toward expanding knowledge on the interplay of different scales and times in battery physics and chemistry to expedite the process of advanced battery system development enabling green mobility technologies.

Authors: Ahmad Pesaran, Gi-Heon Kim, Kandler Smith
DOE’s New CAEBAT Program

- Will integrate the accomplishments of battery modeling activities in national lab programs and make them accessible as design tools for industry
- Will shorten time and cost for design and development of EDV battery systems

![Diagram of CAEBAT Program]

- **Task 1:** Open Architecture Development
- **Task 2:** Pack Model Development
- **Task 3:** Cell Model Development
- **Task 4:** Electrode/Material Models

**Industry/University Participation (RFP)**