Safety of Li-ion Batteries: A Thermal Engineering Perspective

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Email: jaina@uta.edu, Web: www.uta.edu/mtl/
• **Mission:** To Understand and Optimize Heat Transfer in Engineering and Biological Materials, Processes and Systems.

• **Research directions:** Thermal Measurements and Modeling, Electrochemical Energy Conversion & Storage, Microscale Heat Transfer.

• **Sponsors:**

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Li-ion Cell: A Multiphysics Multiscale System

- Li-Ion Materials
- Electrode Layers
- Electrode Assembly
- Cells and Battery Packs
- Electrochemical Energy Systems

Length scales: nm, μm, mm, m

Electrical
- Ion Transport
- Potential Generation
- Temperature-Dependent Mobility
- Joule Heating

Electrochemical
- Exothermic Heat Generation
- Temperature-Dependent Reaction Kinetics

Thermal
Thermal Concerns in Li-ion Batteries

- Fundamental Heat Transfer Questions of Much Practical Relevance:
  - How much heat is generated in a Li-ion cell and how?
  - What is the nature of thermal conduction through the Li-ion cell?
  - How does one optimize heat generation and conduction processes?
  - How does one thermally interrogate the cell?
  - How does one reconcile thermal and electrochemical trade-offs?
Outline

1. Non-invasive core temperature measurement *(Thermal x-ray)*

2. Prediction of thermal runaway *(When is a battery at risk of exploding?)*

3. Material-level thermal transport measurements *(Why does a cell get so hot?)*

4. Heat pipe based cooling *(How to passively cool a cell)*
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Measurement of Internal Temperature

- Surface temperature measurement is a poor indicator of core temperature.

- A non-invasive method for core temperature measurement is very desirable.

- Such ‘x-ray’ capability exists for other physical properties such as stresses, but not for temperature.

Forgez, et al., *J Power Sources*, 2010


Non-Invasive Core Temperature Measurement

• We have shown that the core temperature of a heat-generating cylinder in steady state is given by

\[ T_{\text{core}} = \frac{1}{2\pi} \int_0^{2\pi} T_0(\theta) d\theta + \frac{QR^2}{4kr} \]

where \( T_0(\theta) \) is the temperature around the outside surface of the cylinder.

• Similarly, in transient conditions, we have shown that

\[ T_{\text{core}}(t) = T_{1,\text{core}}(t) + T_{2,\text{core}}(t) = \left[ \frac{QR^2}{4kr} + \sum_{n=1}^{\infty} A_n \exp\left( -\alpha_r \lambda_{0n}^2 \cdot t \right) \right] + \sum_{n=1}^{\infty} B_{0n}(t) \]

where

\[ A_n = -\frac{Q}{4kr} \int_0^r \left( R^2 - r^2 \right) \cdot r J_0(\lambda_{0n}r) dr \]

\[ B_{0n}(t) = \frac{\alpha_r \lambda_{0n} R J_1(\lambda_{0n} R)}{N_{r,n}} \int_0^t w_{0f}(\tau) \exp\left[ -\alpha_r \lambda_{0n}^2 (t - \tau) \right] d\tau \]

\[ w_{0f}(\tau) = \frac{1}{2\pi} \int_0^{2\pi} T_0(\theta, \tau) d\theta \]

• These equations show that the core temperature can be determined using appropriate integrals of measured surface temperature, either in steady state or in transient.

Non-Invasive Core Temperature Measurement

- A thermal test cell similar in construction and thermal properties as a 26650 Li-ion cell.
- IR camera for surface temperature measurement.
- Theoretical models from last slide used to determine $T_{\text{core}}$ and $T_{\text{core}}(t)$ in various operating conditions.
- Embedded core thermocouple for measurement validation.

Non-Invasive Core Temperature Measurement

- Very good agreement between this measurement method and measurement from embedded thermocouple.

- Able to capture temperature at steady-state and during transients.

- Good agreement for ON-OFF type of heating typical for cyclic charge/discharge.

Core Temperature Measurement in a Li-ion Cell

- 26650 Li-ion cells with embedded thermocouples were assembled.
- Internal temperature was predicted during realistic discharge processes and validated against embedded thermocouple measurements.

**Fabrication Steps**

**Final Cell**

Measurements on 26650 Li-ion Cell

- Core temperature is measured on a 26650 cell instrumented with an internal thermocouple, up to 10C discharge rate.

- Good agreement between predicted temperature and internal thermocouple measurement over entire discharge period.

- We are working towards core temperature prediction during thermal runaway.

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Thermal Runaway Prediction

- Heat accumulation in a Li-ion cell results in thermal runaway.

\[
\left( \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} \right) + \frac{Q(T)}{k_r} = \frac{1}{\alpha_r} \frac{\partial T}{\partial t}
\]

Increasing temperature due to heat generation

Increasing heat generation due to temperature rise

\[
T(r,t) = \sum_{n=1}^{\infty} C_n J_0 \left( \frac{\mu_n r}{R} \right) \cdot \exp \left[ \alpha \left( \frac{\beta}{k} - \frac{\mu_n^2}{R^2} \right) \cdot t \right]
\]

Governing Energy Equation

General Solution

- This shows that in order for the cell temperature to stay bounded,

\[
TRN \equiv \frac{\beta \cdot R^2}{k_r \mu_1^2} < 1
\]

\[
\beta = \frac{dQ}{dT}
\]

- TRN is a fundamental, non-dimensional number that governs whether thermal runaway occurs or not.

Experimental Validation

• Experimental data confirm this theoretical prediction for a variety of cases.

• Thermal runaway is seen to occur only when $TRN > 1$. 

Battery Behavior in Realistic Conditions

- Electrochemical reactions in a battery follow Arrhenius kinetics.
- Experimental data show that temperature remains bounded while $TRN<1$, but thermal runaway occurs once $TRN$ exceeds 1.
- Data indicates the possibility of pro-active prediction of onset of runaway.

$$Q(T) = Q_0 \exp\left(-\frac{E_a}{R_u T}\right)$$

Prediction of Safe/Unsafe Thermal Design Space

- The experimentally-validated model can predict safe and unsafe regions in the thermal design space.

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Measurement of Material-level Thermal Transport

• Measurements show that thermal conductivity of even the least conducting material in a Li-ion cell is greater than the measured cell-level thermal conductivity.

• This indicates that interfacial thermal resistances in a Li-ion cell may be important.

Vishwakarma, et al., *J Power Sources*, 2014
Measurement of Material-level Thermal Transport

- Separator-cathode thermal contact resistance contributes ~88% of total thermal resistance in a Li-ion cell.

- Measurements in good agreement with acoustic mismatch theory for interfacial phonon transport.

<table>
<thead>
<tr>
<th>Baseline (Experiment A)</th>
<th>Thermal Resistance</th>
</tr>
</thead>
<tbody>
<tr>
<td>μKm²/W</td>
<td></td>
</tr>
<tr>
<td>&lt;1</td>
<td>Current Collector</td>
</tr>
<tr>
<td>11</td>
<td>Cathode</td>
</tr>
<tr>
<td>100</td>
<td>Separator</td>
</tr>
<tr>
<td>840</td>
<td>Separator-Cathode Interface</td>
</tr>
<tr>
<td>951</td>
<td>Total Thermal Resistance</td>
</tr>
<tr>
<td>0.24</td>
<td>$k_{	ext{eff}}$ (W/mK)</td>
</tr>
</tbody>
</table>

Vishwakarma, et al., J Power Sources, 2015
Enhanced Thermal Transport Through Interface Engineering

• Surface modification of separator/cathode resulted in significant reduction in separator-cathode thermal contact resistance.

Vishwakarma, et al., J Power Sources, 2015
Thermal Enhancement without Electrochemical Deterioration

• This approach significantly improves thermal performance while preserving electrochemical performance.

• Large-scale implementation presents additional challenges.

Vishwakarma, et al., J Power Sources, 2015
Summary

• Thermal engineering plays a key role in ensuring safety of Li-ion based electrochemical energy storage devices and systems.

• Optimization of material-level thermal transport is important for device-level and system-level improvements in safety and performance.

• Synergistic partnerships between Universities, National Labs and companies is critical for transformative progress.
Facilities

• Thermal property measurement: Netzsch LFA467, Custom-built 1D Heat Flux System, CTherm TPS2200, TA Instruments Fox50.

• Thermal imaging: FLIR A6703sc mid-wave IR Camera, Meiji MT6000 fluorescence microscope, Fastec IL5 high speed camera.

• Electrochemical Measurements: Kikusui PFX2512 C/D system, VersaSTAT4 Potentiostat/Galvanostat.

• Electrical measurements and data acquisition: Multiple lock-in amplifiers, function generators, power sources, picoammeters, etc.

• Custom-built thermal characterization setup for cells/packs.

• Chemical handling: LC101 glovebox and Thermo Scientific 1300 chemical fume hood.

• Vacuum chambers: KL XTEMP−BX, Instec HCS662V

• Simulation & modeling: Advanced multiphysics simulation software, 256-core Xi computer workstation.

• Detonation facilities and high-rate cell cycling facilities (through collaborators).
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