Performance Effects of Electrode Coating Defects and IR Thermography NDE for High-Energy Lithium-Ion Batteries

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Overview

Timeline
- Task Start: 10/1/14
- Task End: 9/30/19
- Percent Complete: 30%

Budget
- Total task funding
  - $1750k
- $350k in FY15
- $350k in FY16

Barriers
- Barriers addressed
  - By 2022, further reduce EV battery cost to $125/kWh.
  - Materials processing cost reduction and electrode quality control (QC) enhancement.
  - Achieve deep discharge cycling target of 1000 cycles for EVs (2022).

Barriers

Partners
- Interactions/Collaborations
  - Equipment Suppliers: Frontier Industrial Technology, Keyence, FLIR
  - Battery Manufacturers: XALT Energy, Navitas Systems
  - Materials Suppliers: TODA America, Superior Graphite
  - National Laboratories: ANL, NREL
- Project Lead: ORNL
Relevance & Objectives

Main Objective: To reduce the amount of scrap electrode by at least 75% and the associated amount assembled into finished cells.

- Reduce lithium ion battery system cost by implementing in-line NDE and electrode QC.
- Quantification of effects of different defect types on rate performance and cell lifetime.
- Identify manufacturing defects and their relation to cell failure.
- Implement materials characterization to investigate the cell failure mechanism(s).
- Collaborate with battery manufacturers for QC technology development.
- Use electrode thermal excitation and associated IR emissivity to determine in-line porosity (ORNL/NREL).
- Use in-line XRF to determine areal weight uniformity across and down the web (ORNL/NREL).

• Relevance to Barriers and Targets

- Implementation of critical QC methods to reduce scrap rate by creating feedback loops (by 2022, reduce EV battery cost to $125/kWh).
- Quantification of various defect effects on cycle life (to achieve 1000 deep-discharge cycles for EVs by 2022).
## Project Milestones

<table>
<thead>
<tr>
<th>Status</th>
<th>Milestones</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>6/2016</td>
<td>FY16 Milestone (Completed 5/2016)</td>
<td>Quantify long-term capacity fade (1000 1C/-1C cycles) for at least three different types of anode and cathode coating defects in full 1-Ah pouch cells and publish findings (i.e. transfer technology to domestic LIB manufacturers).</td>
</tr>
<tr>
<td>9/2016</td>
<td>FY16 Milestone On Schedule</td>
<td>Verify performance of an optimally configured active IR thermography system using ABR baseline anodes and cathodes with known thickness, porosity, and bulk density differences on the ORNL slot-die coating line.</td>
</tr>
</tbody>
</table>
Project Approach

• Problems to be addressed:
  – Excessive scrap rates of electrodes and lack of ability to detect coating defects prior to formation cycling; novel, low-cost methods of NDE and QC are required.
  – Conventional electrode QC involves inspection only with optical CCD camera, which may miss many subtle, but important, defects.
  – Effects of different types of defects on cell rate performance and cycle life must be quantified to help determine which coating defects must be identified in the first place.
  – For in-line porosity measurements, white light or thermal excitation of the electrode coatings can be utilized to generate an IR emissivity signature from electrode coatings. Subsequently, IR emissivity can be measured and correlated to a coating temperature profile for input into a mathematical model based on electrode physical properties (IR absorbance, heat capacity, thermal conductivity, bulk density, etc.).
  – Experimentally obtained calibration curves can also be used. Comparison between modeled and measured heat loss down the web is used to generate electrode porosity and/or thickness profiles. A good model and accurate set of material input parameters is needed.

• Overall technical approach and strategy:
  1. Continue identification and demonstration of efficacy of in-line QC techniques utilized in other industries (plastics, textiles, ceramic coatings, photovoltaics, etc.) on ORNL pilot coating equipment.
  2. Correlate in-line NDE and QC methods with systematic cell performance data of various defect types (i.e. identify defects and test electrodes with these defects in coin and pouch cells).
  3. Quantify defect effects on cell performance (power, energy and lifetime).
  4. Employ advanced materials characterization techniques to devise cell failure mechanism(s) associated with defects.
Technical Accomplishments – Executive Summary

• FY15 Q3-Q4:
  – Developed methods to generate different electrode coating defects such as pinholes, blisters, large agglomerates, divots, and metal particle contaminants for evaluation in full coin-cell test matrix.
  – Obtained comprehensive, statistically representative full coin-cell data on different types of electrode coating defects to determine which types of defects cause cell failures or substandard performance (rate performance and capacity fade).
  – Investigated and correlated IR thermography electrode QC data with full coin-cell statistical data.

• FY16 Q1-Q2:
  – Obtained microstructural information for understanding cell failure mechanism(s) associated with electrode defects.
  – Tested various coating defects in full 1.5 Ah pouch cells.
  – **Porosity** proof-of-concept experiments were completed at progressively more realistic conditions:
    • Transient temperature decay with different power levels.
    • Line speed = 5-10 ft/min (*over an order of magnitude improvement from FY15*).
  – Additional mathematical modeling results:
    • Effect of porosity on the temperature profile
    • Effect of thickness on the temperature profile
1. Technical Accomplishments

Systematic analysis of effect of defects on capacity retention and microstructural investigation

2. Technical Accomplishments

1Ah pouch cells were successfully built with three types defective NMC532 electrodes

Schematic regular, and customized shim set used to apply an uneven coat of electrode slurry

- NMC532 with pinholes
- NMC532 with 1X big defect
- Schematic regular, and customized shim set used to apply an uneven coat of electrode slurry

(a) Nonuniform coating (1X big defective area)

(b) Optical image

1X Big Defective Area

3X Small Defective Area

(c) Nonuniform coating (3X small defective areas)
3. Technical Accomplishments

Capacity fading comparison among three different type of defective electrodes at higher current density

- 80% capacity retention
- 3X small defective electrodes show faster capacity degradation
4. Technical Accomplishments

Microstructural investigation on “coated” and “uncoated” interface in 1Xbig/3X small defective electrodes

Poor coating thickness, electrode delamination in the interfacial lengths

![Cross SEM images of baseline and defective NMC532 electrodes](image-url)
5. Technical Accomplishments (ORNL/NREL)

New heater installed in web-line (10× more power than LED source)

Comparing to the previous LED-optical fiber-cylindrical lens system, a few differences were observed:

- Illuminated line is wider. In the case of the old LED heat source, the radiation was focused on a smaller spot (~3 mm wide). For the new heater, the diameter is ~13 mm. Consequently the focus line is approximately 13-25 mm wide.

- Broadband IR radiation is used. Some portion of the radiation is reflected and affects the temperature measured by an IR camera \( \dot{q}_{\text{measured}} = \dot{q}_{\text{emitted}} + \dot{q}_{\text{reflected}} \). For example, when a cathode with an apparent \( T \) of 50°C is detected, then the actual \( T \) of the specimen is 13°C lower.
6. Technical Accomplishments

Effect of wider beam (modeling)

<table>
<thead>
<tr>
<th>beam type</th>
<th>powerDensity</th>
<th>SD</th>
<th>powerDensityIntegral</th>
<th>power for 10in wide web</th>
</tr>
</thead>
<tbody>
<tr>
<td>low_pr_narrow_beam</td>
<td>1.56 W/cm²</td>
<td>1.0 mm</td>
<td>0.156 W/cm</td>
<td>4.0 W</td>
</tr>
<tr>
<td>low_pr_wide_beam</td>
<td>0.1014 W/cm²</td>
<td>20 mm</td>
<td>0.2028 W/cm</td>
<td>5.2 W</td>
</tr>
</tbody>
</table>

20× wider beam required only 30% more energy to reach the same T peak height.
Having narrow beam radiation beam is not critical for operation of the porosity scanner.
7. Technical Accomplishments

Web-line experiment WEB4 ($T_{heater} = 235^\circ C$, cathode only)

Web-line was run at two speeds. For each speed there were 3 repetitions. Total of 6 runs were carried out.
8. Technical Accomplishments

Web-line experiment WEB3 ($T_{heater} = 250^\circ C$, cathode only)

- These charts were generating by averaging $T$ over time within the flat section of $T$ vs. frame plot.
- Specimen effect $\Delta T$ is a difference in response of NMC/NMP98 and NMC/NMO2764 specimens.
- Error bars show standard random uncertainty (U95R).
9. Technical Accomplishments

Web-line experiment WEB4 ($T_{\text{heater}} = 235^\circ\text{C}$, cathode only)

New heater

- Specimen effect $\Delta T$ is inversely proportional to speed. This is consistent with what we modeled and measured previously.
- The new heater enabled going from 0.5 to 5 ft/min keeping the specimen effect roughly the same.
- Even higher speeds can be tested by increasing $T$ of heating rod.
- Clearly run-to-run variations can be larger than the random uncertainty.
Collaborations

• Partners
  – Equipment Suppliers: Frontier Industrial Technology, Keyence, FLIR Systems
  – Battery Manufacturers: XALT Energy, Navitas Systems
  – Raw Materials Suppliers: TODA America, Superior Graphite
  – National Labs: ANL, NREL

• Collaborative Activities
  – Significant leveraging of EERE FCTO and VTO funding to advance this research.
  – Ongoing discussion with industry partners XALT Energy, Navitas Systems, and A123 Systems on implementation of IR thermography, laser thickness measurement, optical reflectance, and thermal diffusivity techniques on industrial electrode production lines.
Future Work

• Remainder of FY16
  – Complete long-term capacity fade (1000 1C/1C cycles) tests for at least three different types of anode and cathode coating defects in full 1-Ah pouch cells.
  – Employ modeling to understand the lithium transport with respect to different coating defects.
  – Complete installation of active IR thermography systems on ORNL slot-die coating line and NREL R2R diagnostic line.
  – Publish findings (i.e. transfer technology to domestic LIB manufacturers).
  – Identify industrial partner to scale in-line QC methods.

• Into FY17
  – Increase line speed of in-line porosity measurement system to 10-20 ft/min.
  – Revisit in-line XRF for measuring coating areal weight with NREL equipment.
  – Develop software package for laser caliper thickness input and XRF areal weight input for in-line porosity model.
  – Develop feedback loops with NREL and key battery manufacturer.
  – Analysis of cost saving by implementing NDE.
Summary

• **Objective:** This project facilitates lowering unit energy cost of EVs and PHEVs by addressing the electrode scrap rate, QC enhancement, and long-term performance.

• **Approach:** Distinguishes between defects critical to performance and those not as important, and implements QC measures utilized effectively in other industries.
  - Ease of implementation of measurement technology with low equipment cost.
  - Quantify effect of electrode coating defects such as divots, blisters, pinholes, agglomerates, and metal-particle contaminants on cell rate performance and cycle life.

• **Technical:** Successful design of new higher-speed active IR thermography system at NREL; identification of defects critical to cell capacity fade (coin and full pouch cells); characterization of defective electrodes and investigation of cell failure mechanisms.

• **Collaborators:** Active discussions with industry partners XALT Energy and Navitas Systems on scaling measurement techniques; publishing QC enhancement methodology for lithium-ion cell industrial implementation.

• **Commercialization:** High likelihood of technology transfer because of strong industrial collaboration, significant electrode production cost reduction, and lower-cost QC measurement equipment.
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Thank you for your attention!
Reviewer Comment Slides (Task Not Reviewed in FY15)