NREL
NATIONAL RENEWABLE ENERGY LABORATORY

Coupling Mechanical with Electrochemical-Thermal Models for Batteries under Abuse

P.I. and Presenter: Ahmad Pesaran
National Renewable Energy Laboratory (NREL)
Team: Tomasz Wierzbicki and Elham Sahraei (MIT)
Stephen Dajka and Genong Li (ANSYS)
S. Santhanagopalan, Chao Zhang, G.H. Kim, and M. Sprague (NREL)

June 9, 2015

Project ID # ES199

This presentation does not contain any proprietary, confidential, or otherwise restricted information.

NREL/PR-5400-64148

Overview

Timeline

- **Project Start:** October 2013
- **Original Project End:** Sep 2015
- **New Project End:** February 2016
- **Percent Complete:** 60%

Budget

- **For Two Years**
- **Total Project Budget:** $1,003K
  - **DOE Share:** 100%
- **Funding Received:** $1,003K
  - **MIT Share:** $200K
  - **ANSYS Share:** $100K
  - **NREL Share:** $703K

Barriers Addressed

- Safety concerns of lithium (Li) batteries
- Thermal runaway of Li batteries due to heating
- Understanding the thermal response of batteries after crash-induced crush

Partners

- Massachusetts Institute of Technology (MIT)
- ANSYS Inc.
- NREL Energy Storage Team
- NREL High Performance Computing Team
- Project Lead – NREL

Relevance – DOE VTO Program

- The DOE's EV Everywhere Grand Challenge aims to produce plug-in electric vehicles (PEVs) as affordable, safe, and convenient for the American family as gasoline-powered vehicles by 2022.
  - PEVs must be as safe as conventional vehicles.
  - PEV batteries must not lead to unsafe situations under abuse conditions.
- The 2011–2014 DOE VTO Computer Aided Engineering for Electric Drive Vehicle (CAEBAT) activity was very successful in releasing electrochemical-thermal models in commercial software tools.
- The DOE VTO FOA released in January 2013 called for proposals to link mechanical models to electrochemical-thermal tools (as a follow-on phase to CAEBAT) to study crash-induced crush.
- This project, based on our proposal in response to the FOA, started in October 2013.
- The goal is to develop CAEBAT tools to reduce the development cycle of safer designing in-vehicle Li-ion battery systems, thus reducing costs of batteries and making PEVs affordable.

Relevance – Project Objectives

This project addresses two abuse conditions that could be experienced in PEVs: “overheating/thermal ramp” and “crash-induced crush” of cells

The objectives are to:

- Develop a model to predict the thermal response of cells to thermal ramp
- Develop a model to couple the electrochemical-thermal (ECT) behavior of a Li-Ion cell to its structural behavior after rapid mechanical deformation such as crush
- Make the models compatible with CAEBAT-1 and its Open Architecture Software (OAS) for wider proliferation of their use.

Milestones (Revised Dates) – 1

Revised Date*	Milestone Go/No-Go	Description	Status
Dec 2013	Milestone	Place subcontracts with MIT	Complete
Oct 2014	Milestone	Place Subcontract with ANSYS (10 month delay)	Complete
Feb 2015	Milestone	Develop user-interface software for running chemical kinetics model	Complete
May 2015	Milestone	Document user-interface software and demonstration of CAEBAT-1 compatibility	On-track
May 2015	Milestone	Demonstrate compatibility of chemical kinetic model with OAS	On-track
Jun 2015	Go/No-Go	Document validation of chemical kinetic abuse model and computational model performance	On-track
Jul 2015	Milestone	Chemical kinetic abuse model input files available on CD and/or publicly accessible website	On-track

* End of month

Milestones (Revised Dates) – 2

Revised Date*	Milestone Go/No-Go	Description	Status
Apr 2014	Milestone	Document results of mechanical deformation experiments	Complete
Sep 2014	Go/No-Go	Document validation and computational mechanical deformation and damage model	Complete
Oct 2014	Milestone	Document simulation results of ECT response of mechanically crushed cell	Complete
Oct 2015	Milestone	Document validation of computational mechanical-ECT (MECT) model performance	On-track
Dec 2015	Milestone	Document user interface for importing MECT model to CAEBAT-1 platform	On-track
Feb 2016	Milestone	Document implementation of MECT software with Open Architecture Software	On-track

* End of month

Approach – Thermal Ramp Modeling

Transfer NREL chemical kinetic abuse model(s) to ANSYS CAEBAT-1 platform.

- Model chemical kinetics reactions happening in Li-ion cells at elevated temperatures including:
  - Solid electroplate interface (SEI) decomposition
  - Negative-solvent reactions
  - Positive-solvent reactions
  - Electrolyte decomposition
- ANSYS to create user-friendly interface to access the above chemical reaction model in CAEBAT-1 platform
- Make the model/tool compatible with OAS through ANSYS Battery Design Tool developed in CAEBAT-1

Technical Accomplishments and Progress

Implemented NREL Thermal Abuse Model Implementation In Fluent

- Total heat generation due to thermal abuse modeled as four exothermic reactions

$$S_{\text{abuse\_chem}} = S_{\text{sei}} + S_{\text{ne}} + S_{\text{pe}} + S_{\text{ele}}$$

1. SEI decomposition reaction

$$R_{\text{sei}}(T, c_{\text{sei}}) = A_{\text{sei}} \exp\left[-\frac{E_{\text{a,sei}}}{RT}\right] c_{\text{sei}}^{m_{\text{sei}}}$$

$$\frac{dc_{\text{sei}}}{dt} = -R_{\text{sei}}$$

$$S_{\text{sei}} = H_{\text{sei}} W_{\text{c}} R_{\text{sei}}$$

Constant: A_{sei}, m_{sei} , $E_{a,\,sei}$, H_{sei}, W_c

2. Negative-solvent reaction

$$R_{\text{ne}}(T, c_{\text{e}}, c_{\text{neg}}, t_{\text{sei}}) = A_{\text{ne}} \left(\frac{t_{\text{sei,ref}}}{t_{\text{sei}}}\right) c_{\text{neg}}^{m_{\text{ne,n}}} \exp\left[-\frac{E_{\text{a,ne}}}{RT}\right],$$

$$\frac{dc_{\text{neg}}}{dt} = -R_{\text{ne}}$$

$$S_{\text{ne}} = H_{\text{ne}} W_{\text{c}} R_{\text{ne}}$$

Constant: A_{ne}, $m_{ne,n}$, $E_{a,\,ne}$, H_{ne}, W_c, $t_{sei,ref}$

Technical Accomplishments and Progress

Implemented NREL Thermal Abuse Model Implementation In Fluent (cont'd)

3. Positive-solvent reaction

$$\frac{d\alpha}{dt} = R_{pe}$$

$$R_{pe}(T, \alpha, c_e) = A_{pe}\alpha^{m_{pe,p1}}(1-\alpha)^{m_{pe,p2}} \exp\left[-\frac{E_{a,pe}}{RT}\right]$$

$$S_{pe} = H_{pe} W_p R_{pe}$$

Constant: A_{pe}, $m_{pe,p1}$ $m_{pe,p2}$, $E_{a,pe}$ H_{pe}, W_{pv}

4. Electrolyte decomposition reaction

$$S_{ele} = H_e W_e R_e$$

$$\frac{dc_e}{dt} = -R_e$$

$$R_e(T, c_e) = A_e \exp\left[-\frac{E_{a,e}}{RT}\right] c_e^{m_e}$$

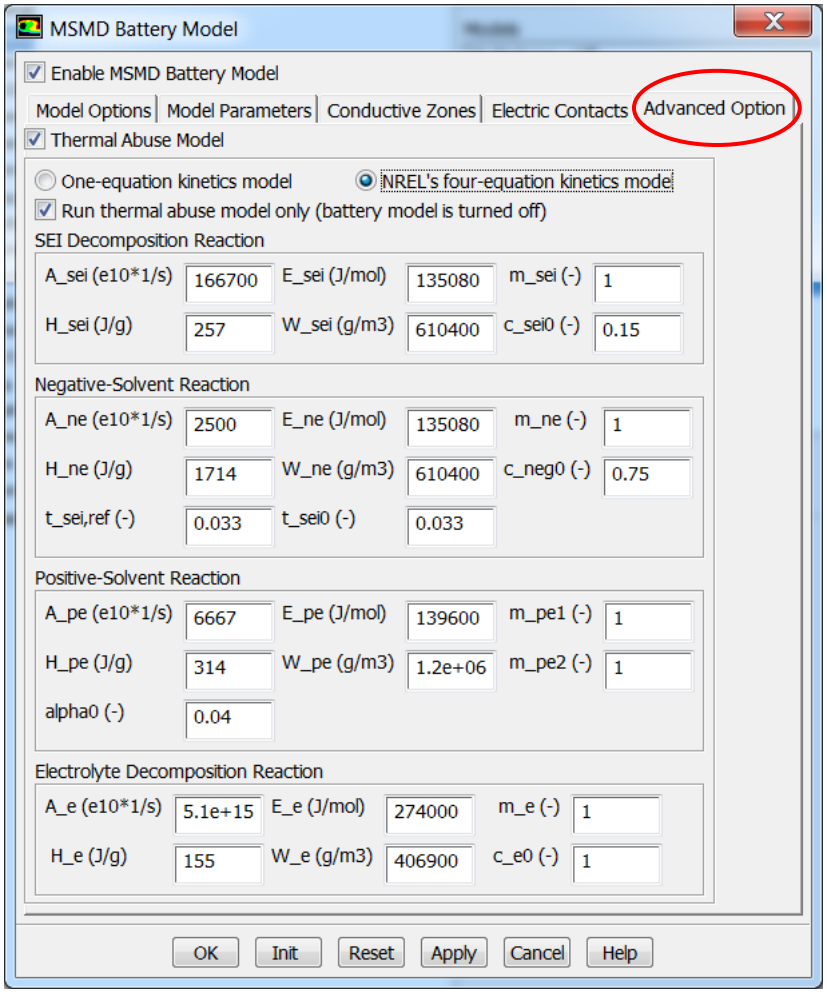
Constant: A_e, $E_{a,c}$,m_e, H_e, W_e

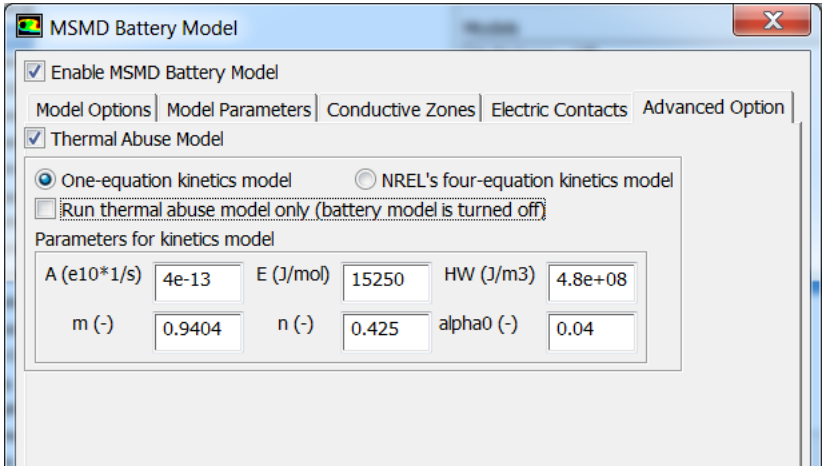
Technical Accomplishments and Progress

ANSYS Parameter Estimation Tool Developed

- Thermal abuse model kinetics data are battery-specific.
- Without a general tool to estimate the related model parameters, the models have very limited practical use.
- Reaction kinetics parameters can be determined by fitting to experimental test data (e.g., oven test, Accelerated Rate Calorimeter).
- An estimation tool for the one-equation model parameters has been made available in Fluent.

Thermal Abuse Model GUI Developed in ANSYS





- Thermal abuse model can be run alone (without running E-chem model).
- The kinetics parameters in the one-equation model can be obtained by running Fluent-provided parameter estimation tool.

Status: Thermal Abuse Model

- Delivered models based on ANSYS/Fluent R15 release
  - R16 version also has been transferred to NREL for evaluation.
  - Merged into development (R17) version for future release.
  - ANSYS provided documentation/presentation and demonstration of model to satisfy ANSYS deliverable.
- ANSYS has reproduced results from Kim, Pesaran, Spotnitz paper (original Abuse Kinetics Model) after some initial challenges such as:
  - Electrolyte reaction rate appeared different
  - Figuring out why kinetics parameters in paper were different from what is used in simulation.
- ANSYS and NREL continued interactions
  - NREL tested general model usage.
  - NREL provided feedback on setup/parameters from ANSYS tests mentioned above in order to satisfy deliverable.

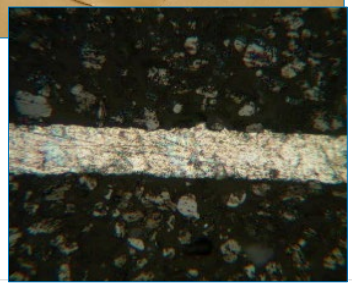
Approach – Cell Crush Modeling

Coupling mechanical models to the ECT models

- Simulating simultaneous mechanical, electrochemical, and thermal response of a cell due to crush is complex and requires modeling simplifications
- Assumed that crush is rapid and electrochemical and thermal response takes longer
- Measured the mechanical properties of cell components such as electrodes and separators
- Related the mechanical aspect with the electrical and thermal aspects in a sequential, one-way fashion
- Simplified the structural modeling of many thin layers of a cell into a smaller number of thicker representative layers

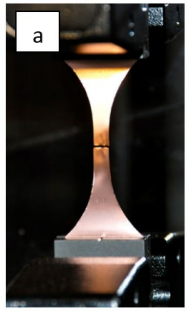
Development and Calibration of a Homogenized Anisotropic Jellyroll Model through Micro-Mechanical Tests at MIT Impact and Crashworthiness Lab

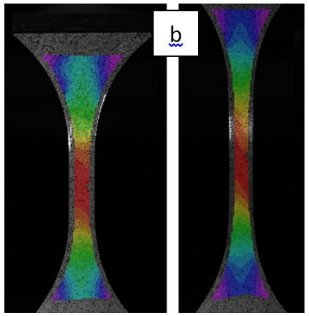
Tensile Testing of Components at MIT

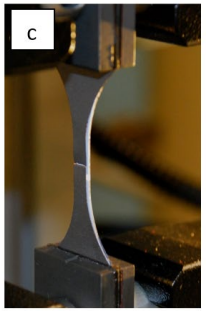


Components tested:

- Copper and aluminum current collectors
- Anode and cathode
- Separator



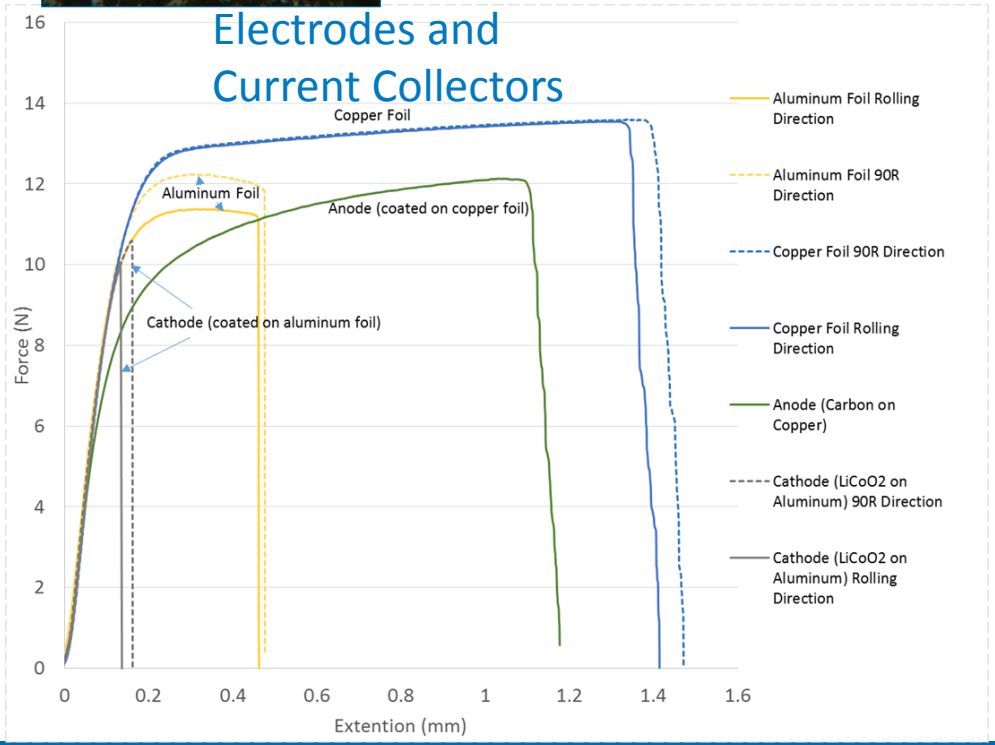




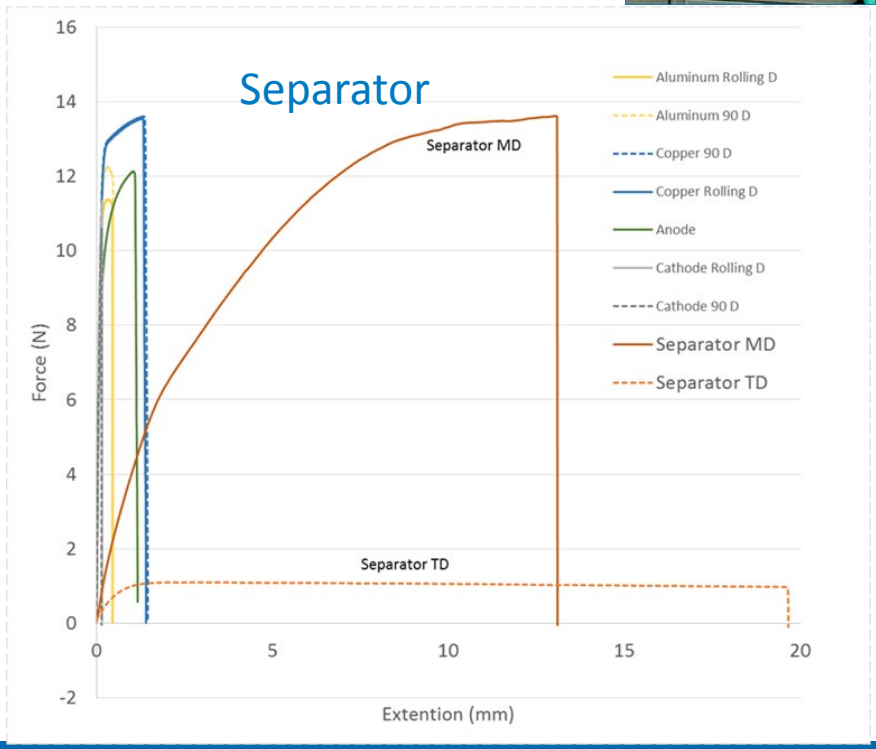
Digital Image Correlation (DIC) to estimate strains

Photo Credits: Elham Sahraei (MIT)

Electrodes and Current Collectors



Separator



Anisotropic Model with Element Removal

Mechanical Model

- **Six load curves, different properties in tension and compression**

Yield surface
$f = 0$

$$f_{ij} = |\sigma_{ij}| - Y_{ij} = 0$$

Hardening formulation

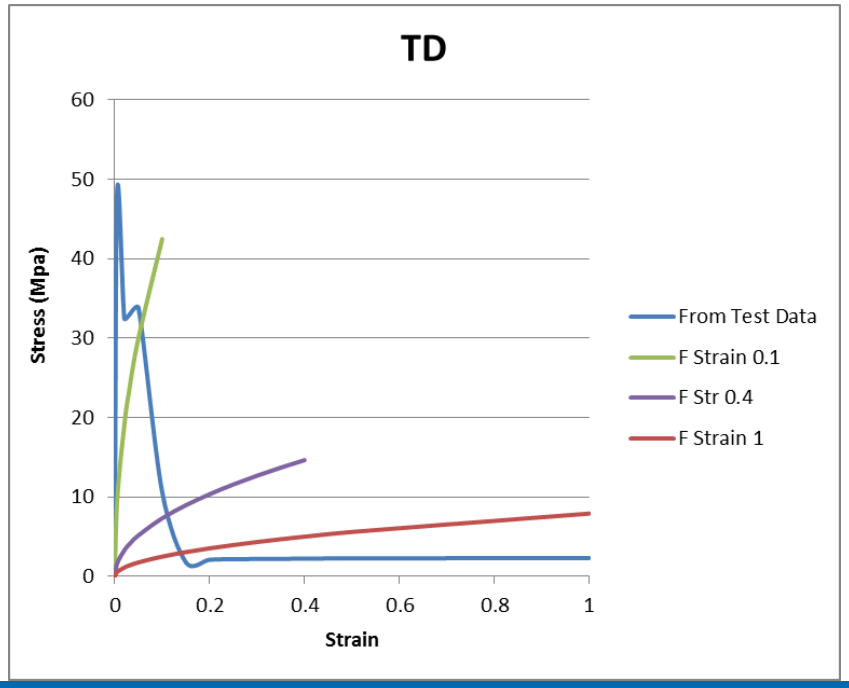
$$Y_{ij} = Y_{ij}^{0} + H_{ij}(e_{ij})$$

Maximum Strain Failure Criteria

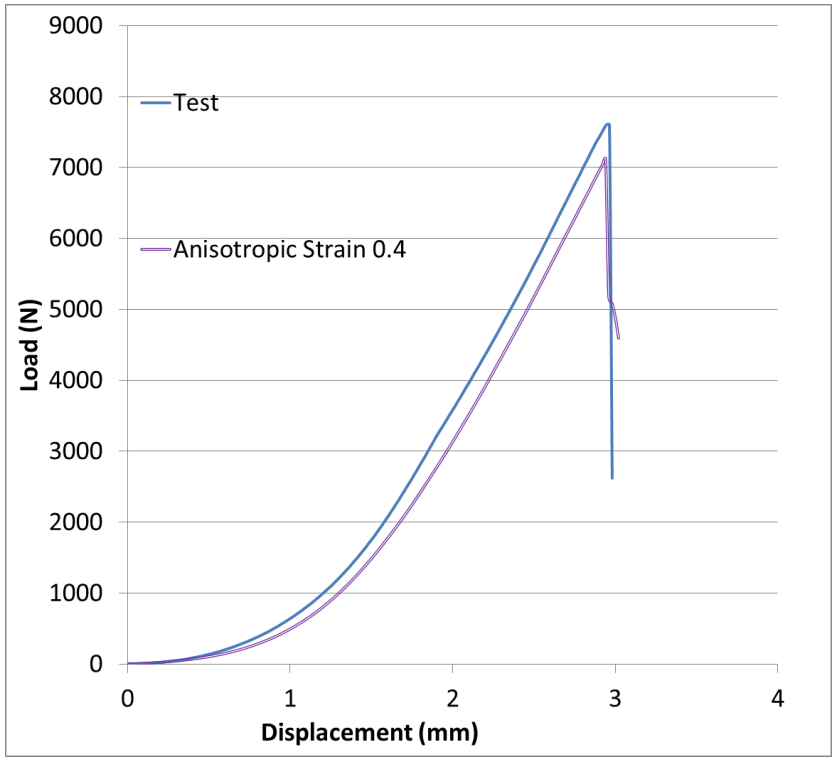
A hardening curve in tension with equal amount of energy absorption before failure

?

$$E = \int_0^{\epsilon_f} \sigma_{average}(\epsilon)\mathrm{d}\epsilon = \int_0^{\epsilon_f} A\,\epsilon^n\,d\epsilon$$



Failure Strain Calculated to be 0.4



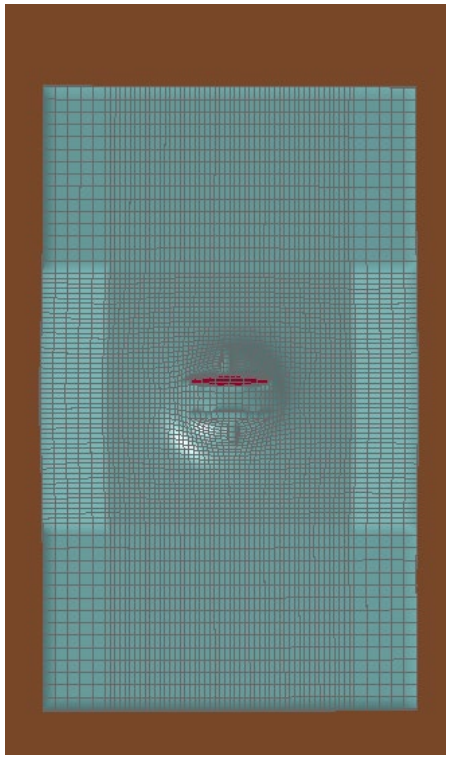
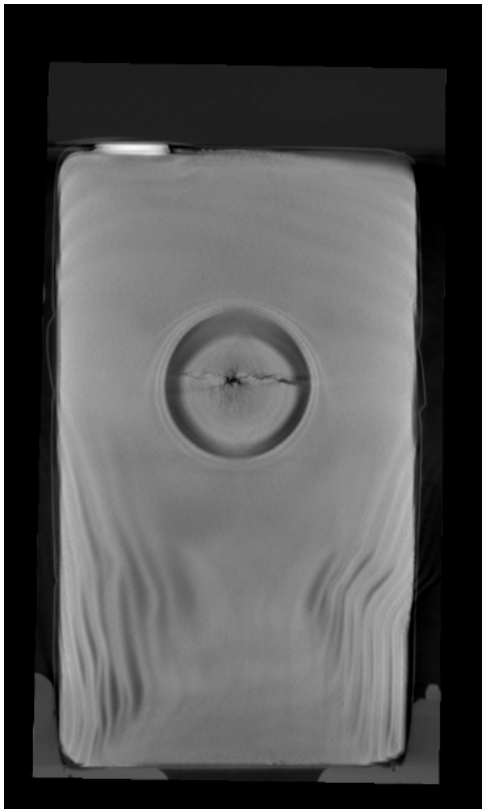


Illustration Credit: Elham Sahraei (MIT)



CT Scan
Courtesy of Exponent

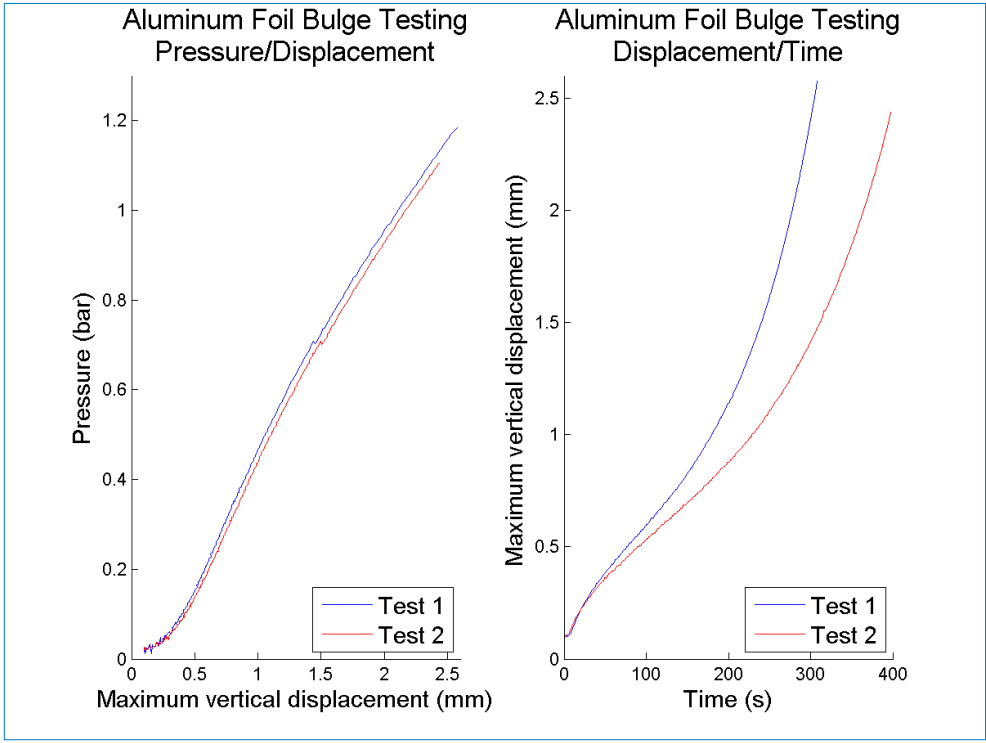
Bulge Testing – Benefit: No Edge Failure

Motivation

- Material parameters for aluminum
- No edges on the specimens
  - Simple preparation of specimen
  - No premature failure of specimen
- That same approach was used recently (Kana et al., 2014) for this same reason

Room for improvements

- Plane strain testing via elongated pressure chambers, used by Vlassak and Nix (1992) on thin films



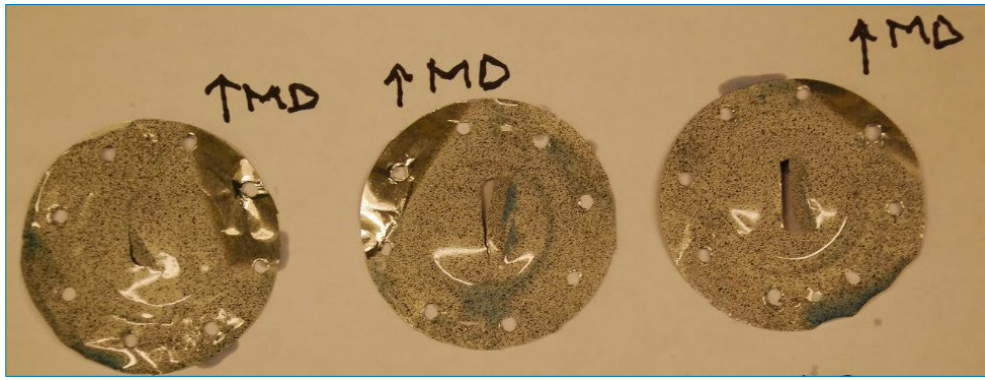


Photo Credits: Elham Sahraei (MIT)

Micro-testing

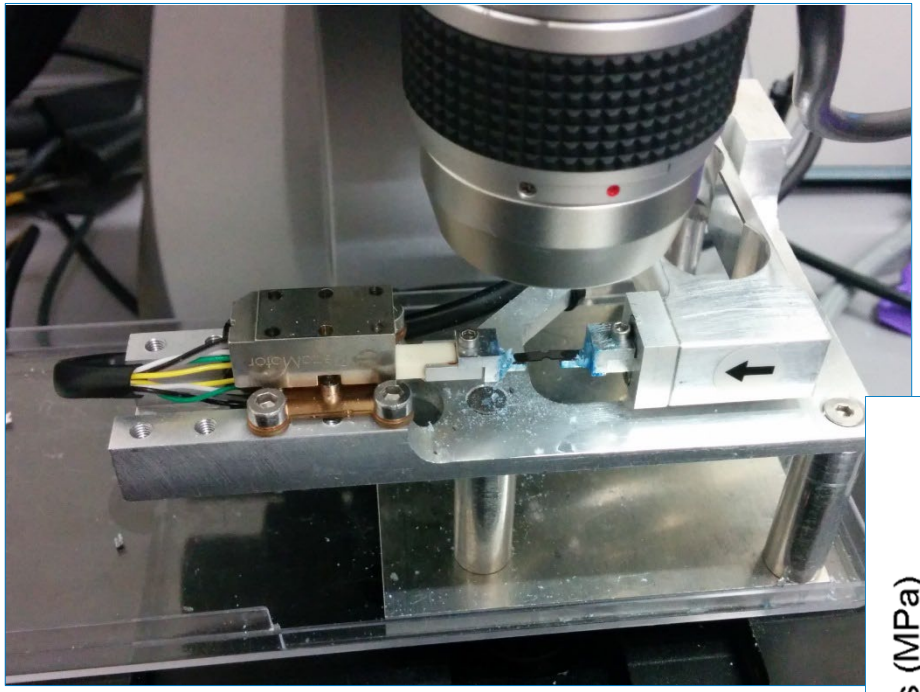
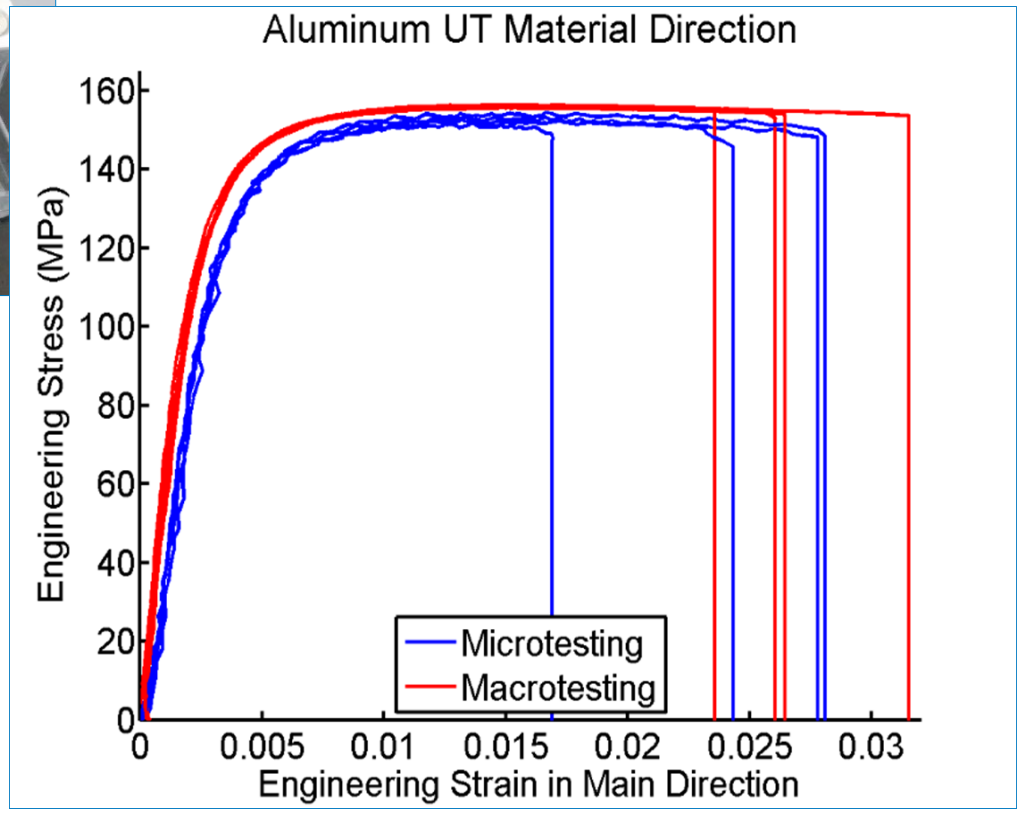


Photo Credits: Elham Sahraei (MIT)



Conclusions – MIT Anisotropic Model

- An anisotropic homogenized material model was successfully adopted to predict deformation of a battery cell during mechanical loading.
- The model closely predicts load-displacement, onset of failure, and size and location of crack.
- Previous experimental protocol was modified.
  - Allows for better reproducibility of testing on various geometries.
- Phenomenological modeling of the bare foils is under way.
- A bulge test apparatus was developed.
  - Avoids any influence of the specimen edges.
- Previous experimental protocol was adapted to a lower scale.
  - Allows direct observation of the material (no paint).
  - Allows proper observation of coatings.
- Separator properties in three directions—the differences were investigated.

Approach – Coupled Modeling of Cell Crush

Mechanical-electrical-thermal (MECT) coupled model

- Develop a representative sandwich (RS) model for coupled modeling of cell crush, which explicitly represents each individual component.
- Our approach assumes an anode-cathode short occurred followed by the failure of the separator due to mechanical crush.
- This allows us to predict mechanical abuse-induced short circuit using a sequential, faster one-way method.

Model validation

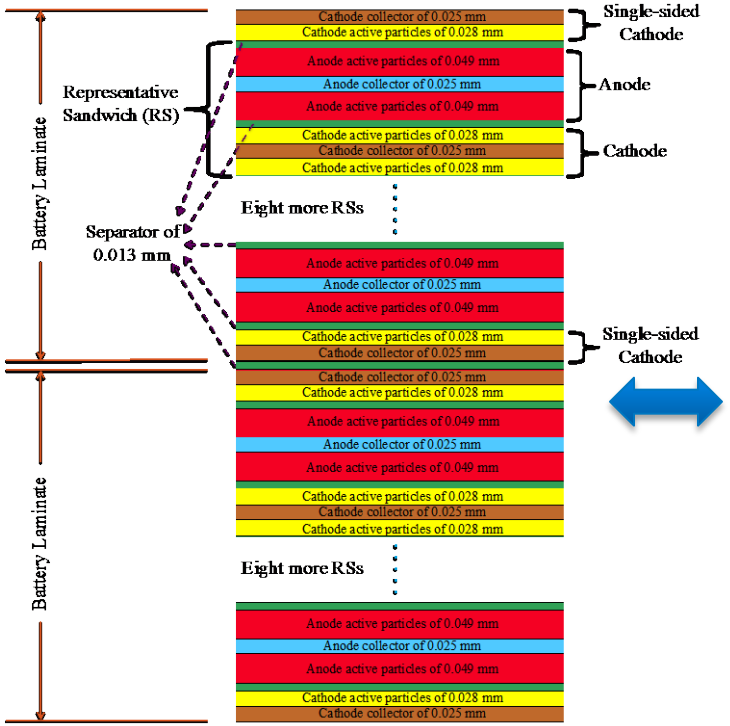
- NREL in collaboration with MIT is developing an efficient way to extract material properties from experimental measurements and implemented into the coupled model.
- We are correlating the coupled model with experiments on mechanical failure, voltage drop, and thermal ramp.

Couple mechanical model with NREL's ECT model

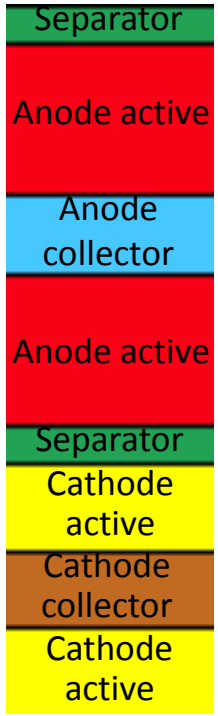
- We modeled electrochemical responses followed by mechanical abuse-induced short circuit using NREL's ECT model on the mechanical model predicted deformed geometry.

Accomplishments – NREL Cell Crush Model

- A single RS finite element model is developed to simulate the coupled response to crush a pouch battery cell under mechanical crush, which includes an explicit representation of each individual component (copyrighted)



Full pouch cell layout

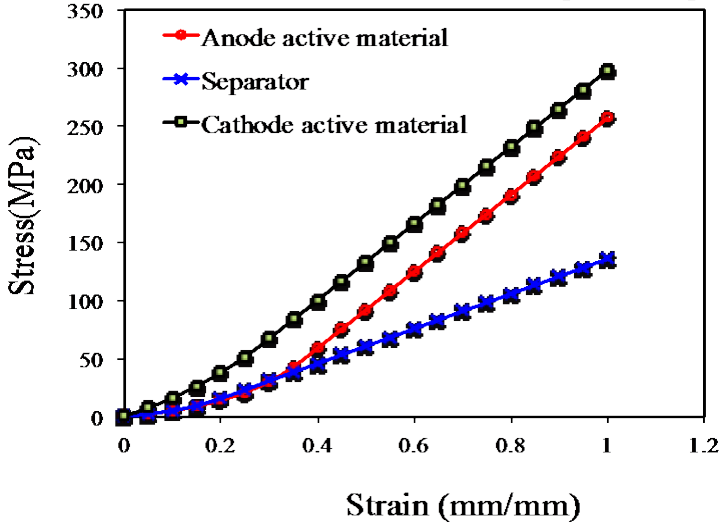


Single RS

- An analytical procedure is developed to derive through-thickness stress-strain responses for active materials and separator from compressive experimental data of a full pouch cell

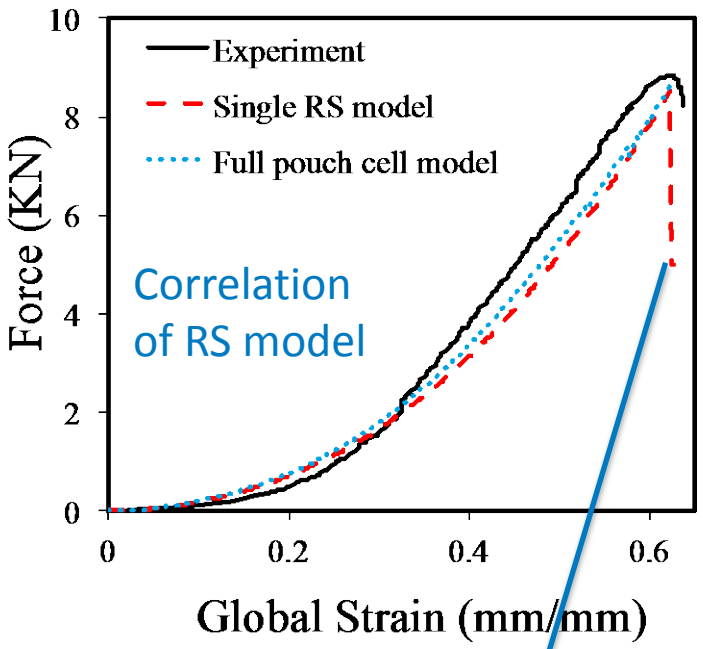
$$\sigma = \begin{cases} \dfrac{E_{\max}(e^{\beta\varepsilon}-1)}{\beta e^{\beta\varepsilon_p}} & \varepsilon < \varepsilon_p \\ \dfrac{E_{\max}(1-e^{-\beta\varepsilon_p})}{\beta} + E_{\max}(\varepsilon-\varepsilon_p) & \varepsilon \geq \varepsilon_p \end{cases}$$

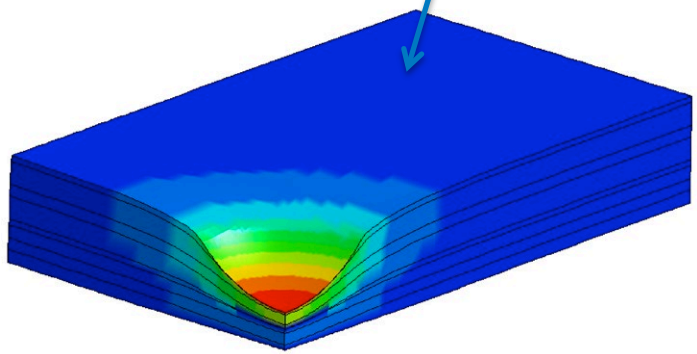
$$\varepsilon_{eff} = v_{actives}\varepsilon_{actives} + v_{collectors}\varepsilon_{collectors} + v_{separator}\varepsilon_{separator}$$



Accomplishments – NREL MECT Simulation

- **The single RS model correlated well with experiments in predicting the global effective responses and fracturing of the battery structure.**
- **The RS model can be used to:**
  - Conduct electrical-thermal simulation on geometries with different extents of deformation.
  - Predict the onset of a short circuit during the mechanical crush.
  - Investigate the effect material properties of the individual component have on the safety behavior of the battery.
  - Integrate with the ANSYS ECT model to study the electrochemical responses.

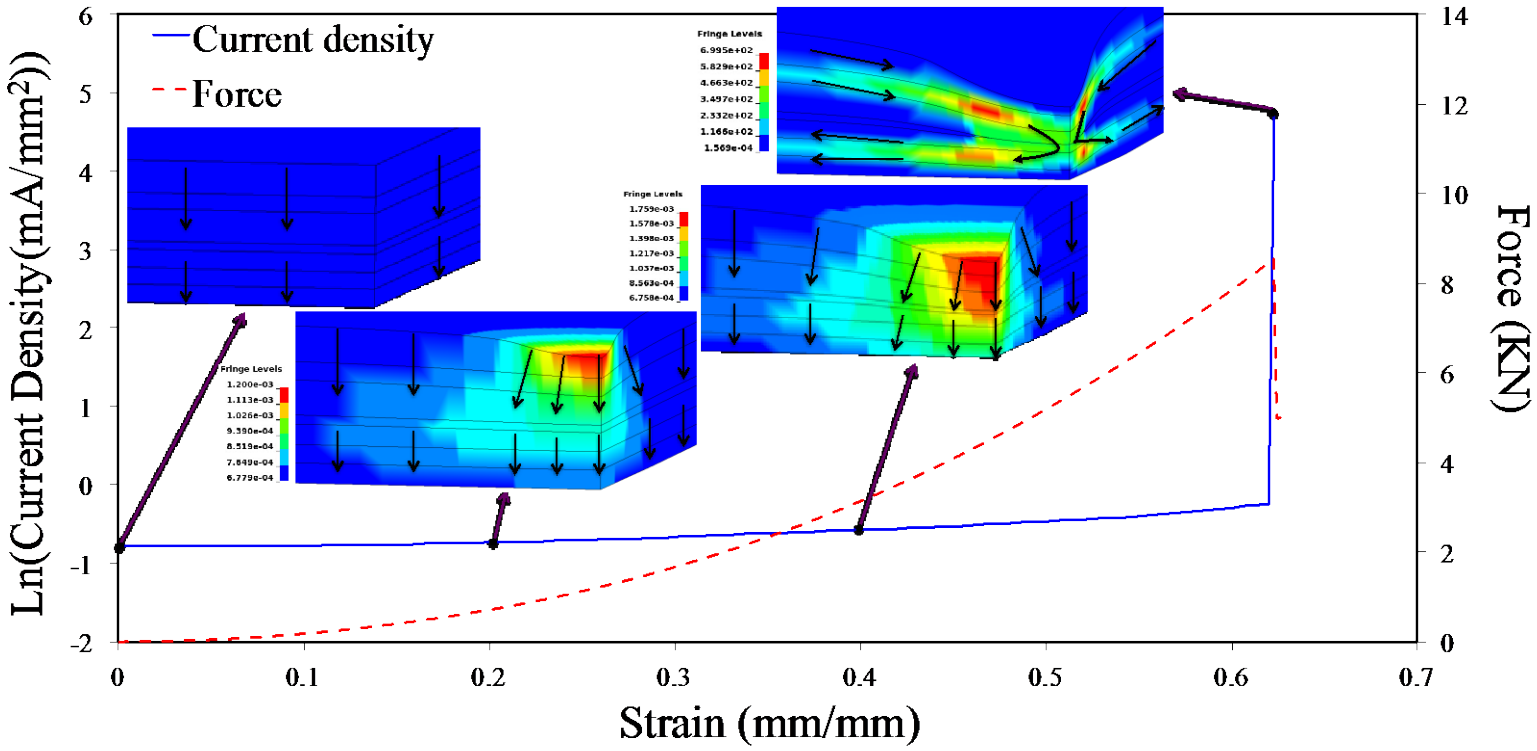




Numerically predicted deformed geometry at the moment of mechanical failure

Accomplishments – NREL Coupled Model

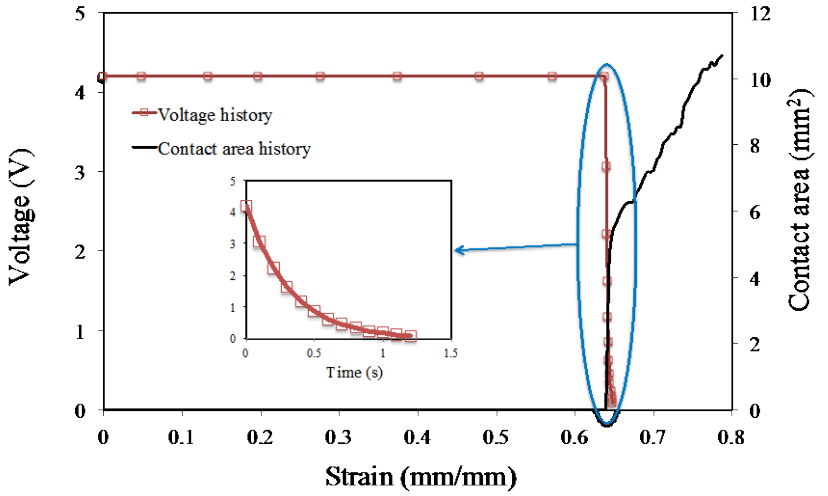
- A sequential mechanical-electrical-thermal coupled modeling approach is established to predict mechanical crush-induced short circuit.
- The onset of short circuit is predicted, which occurs simultaneously with the mechanical fracture of the battery structure.



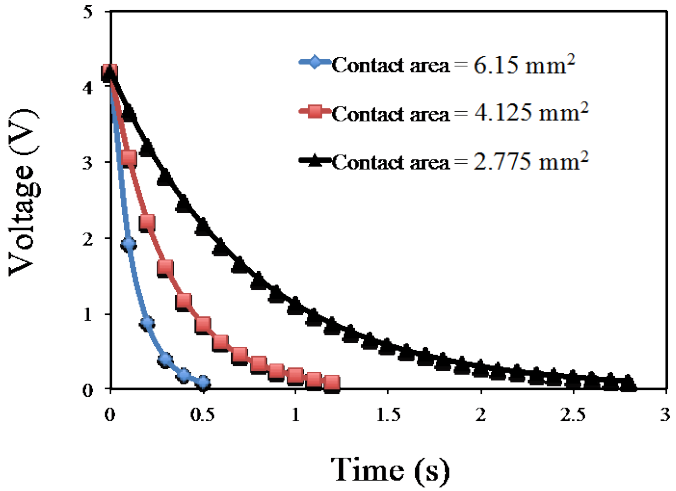
Evolution of current density across the short-circuit area during a mechanical indentation loading condition

Accomplishments – NREL Coupled Model

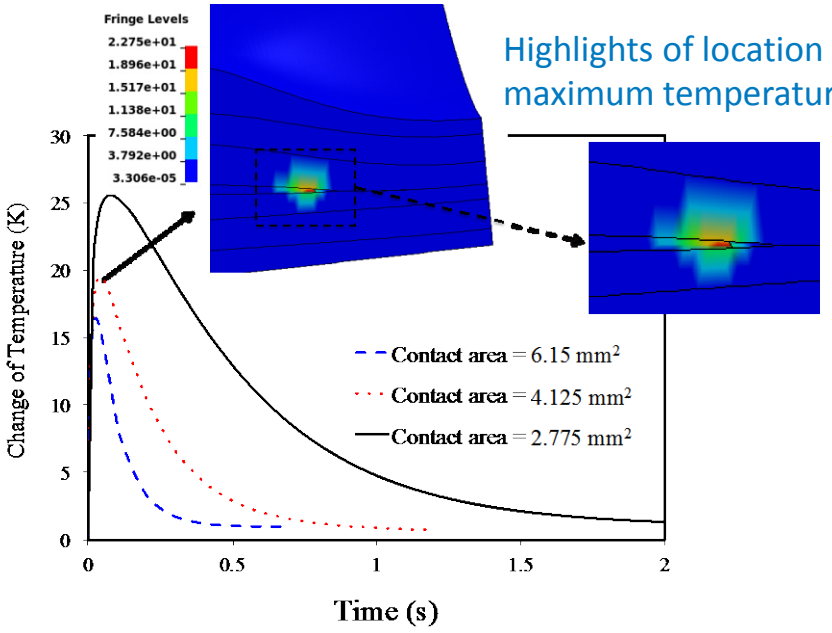
- The voltage evolution and thermal responses before and after short can be predicted using the coupled modeling approach.
- The coupled model shows the potential to study different short-circuit conditions, for example, different electrical contact areas.



Effect of electrical contact area on voltage drop and thermal responses



Highlights of location of maximum temperature



Response to Previous Year's 3 Main Reviewers' Comments

Q1. What are the current limitations of the MIT mechanical and NREL chemical kinetic abuse models? Would these limitations hamper the extension of these models to abuse tests and how you are addressing them?

A. The current MIT model is capable of predicting compression and tension responses in possible abuse scenarios. However, the MIT model is not able to predict delamination and therefore is only suitable for one-way coupling, where the geometry of deformation is used as an input for further chemical and thermal modeling. This limitation was known in advance and is not impeding the current progress.

B. The current NREL chemical kinetics abuse model is based on reactions occurring between major components in a cell. In principle, there is no limitations to extension of the models. These reactions are mostly identified by other researchers in the field, the challenge is measuring the reactions rates with through sufficient testing with accelerating rate calorimeters.

Response to Previous Year's 3 Main Reviewers' Comments

Q2. MIT has long term expertise with small cylindrical cell characterization and crush modeling. Do the results from this cell size and form factor could be particularly transferred with validation toward large format and/or large module configurations?

A. MIT has used the same principles and modeling approaches to develop models for small, medium, and large pouch cells. The large pouch cell is identical in terms of size and form factor to the cells used in some EV battery packs on the market, and the models have successfully predicted the mechanical deformation and failure of those cells. Please see "Characterizing and modeling mechanical properties and onset of short circuit for three types of lithium-ion pouch cells," Elham Sahraei, Joseph Meier, and Tomasz Wierzbicki, *Journal of Power Sources,* 247 (2014) pp. 503–516 for further details.

B. In addition to this, NREL has developed lumped representations of the mechanical model that incorporate the constitutive properties of the individual components (anode, separator, current collector, etc.). The representative sandwich model was developed specifically to address scale-up to large format cells and complicated multi-cell geometries.

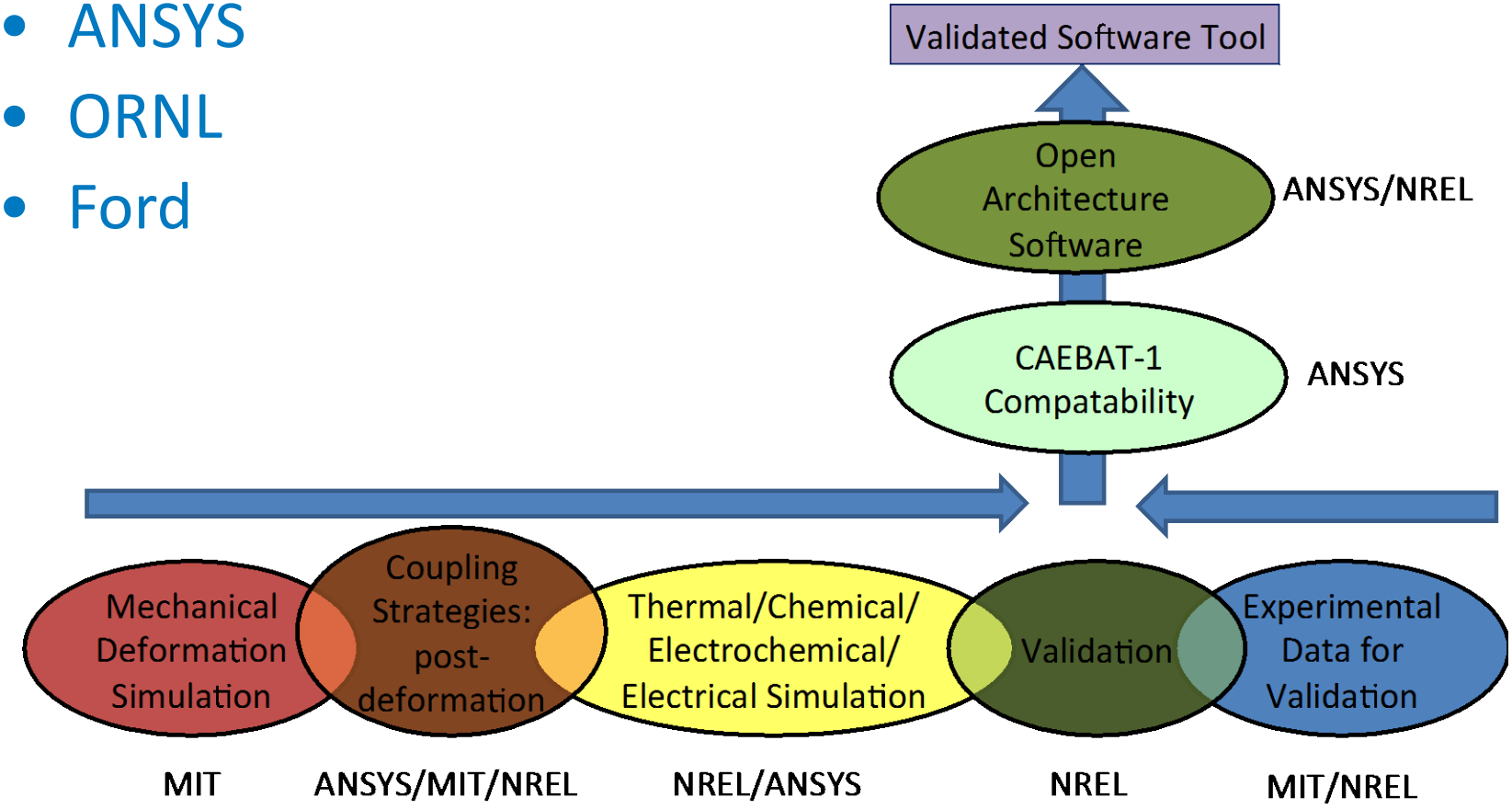
Response to Previous Year's 3 Main Reviewers' Comments

Q3. In (coupled) simulations, what parameters must be exchanged between the models, as there is difficulty of using the multi-layer puncture approach to capture damage zones created by different crush loads and orientations and the fact that model validation with experimental data may be complicated by the difficulty of matching.

A. At present, the work flow is as follows: i) perform mechanical simulations to generate deformed mesh/geometry, ii) calculate resistance of the short-circuit by applying an arbitrary current across the different layers and measuring the voltage drop, and iii) porting the short-circuit resistance as the parameter/field variable to exchange with the ANSYS/Fluent ECT model. This approach is simple, albeit with limitations, but given that there are no prior examples to couple mechanical deformation with battery abuse, we consider this a reasonable starting point.

Collaborations and Coordination

- NREL Energy Storage Team (lead)
- NREL High Performance Computing Team
- Massachusetts Institute of Technology (MIT)
- ANSYS
- ORNL
- Ford



Remaining Challenges and Barriers

- Model validation with experimental data may take longer than expected as simulation may not represent a particular experiment, resulting in repeated simulation or experiments, thus insufficient time and budget for further validation.
- Integration of new code with emphasis on mechanical/structural/chemical physics into ORNL's OAS.

Future Work

- Transfer existing chemical kinetic abuse model to current CAEBAT-1 platform, maintaining compatibility with OAS framework.
- Validate software from Task 2 in thermal ramp abuse conditions against published experimental data.
- Continue validating mechanical deformation (crush) computational models.
- Improve coupling MIT's mechanically deformed models with NREL's ECT models.
- Implement coupled mechanical-electrical-chemical-thermal model in ANSYS CAEBAT-1 platform and make it compatible with OAS.

Summary

- Eventual goal is to be able to predict the onset of thermal runaway after crash-induced crush or thermal ramp.
- Requested a 5-month extension for project competition due to 10-month delay of ANSYS subcontract.
- Coded the ARK model and provided documentation to ANSYS.
- ANSYS's subcontract executed and work began on thermal ramp.
- MIT has conducted many tests on cell components – more are underway.
- Developed a new anisotropic model with RS for the jellyroll, which is calibrated with test data from MIT.
- ANSYS has initiated making software compatible with ABDT.
- NREL developed a model to estimate resistivity of shorts created by mechanical deformations.

NREL
NATIONAL RENEWABLE ENERGY LABORATORY

Technical Back-Up Slides

(Note: please include this “separator” slide if you are including back-up technical slides (maximum of five). These back-up technical slides will be available for your presentation and will be included in the DVD and Web PDF files released to the public.)

Coupling with ECT Simulations

Initial Geometry

Crush + EM Simulations in DYNA

Feed Mesh and Resistance to ANSYS

Perform ECT Simulations