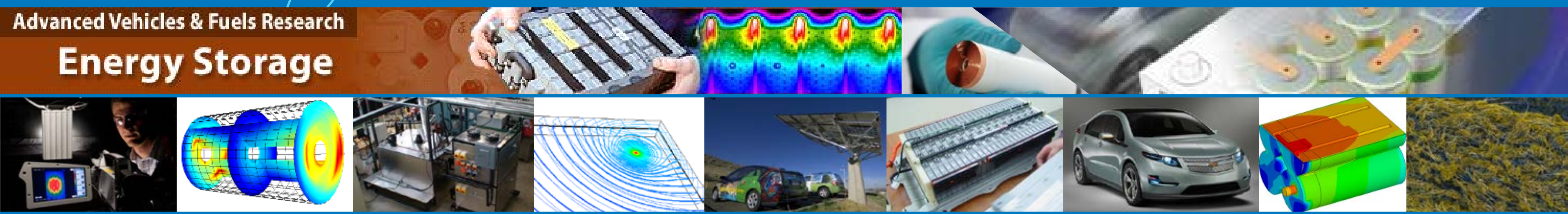
NREL
NATIONAL RENEWABLE ENERGY LABORATORY

Coupling of Mechanical Behavior of Cell Components to Electrochemical-Thermal Models for Computer-Aided Engineering of Batteries under Abuse



P.I.: Ahmad Pesaran

Team: Tomasz Wierzbicki and Elham Sahraei (MIT)
Genong Li and Lewis Collins (ANSYS)
M. Sprague, G.H. Kim and S. Santhangopalan (NREL)

June 17, 2014

NREL/PR-5400-61885

Project ID: ES199

Overview

Timeline

- **Project Start:** October 2013
- **Project End:** September 2015
- **Percent Complete:** 20%

Budget

- **Total Project Budget:** $1,003K
- **Funding Received:** $1,003K
  - **MIT Share:** $200K
  - **ANSYS Share:** $100K
  - **NREL SHARE:** $703K

Barriers Addressed

- Safety concerns of 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
- NREL Energy Storage Team
- NREL High Performance Computing Team
- Project Lead – NREL

Relevance – DOE VTO Program

- The EV Everywhere Grand Challenge aims to produce plug-in electric vehicles as affordable and convenient for the American family as gasoline-powered vehicles by 2022
  - Electric vehicles must be as safe as conventional vehicles
  - EV batteries must not lead to unsafe situations under abuse conditions
- This project was started in October 2013 based on a proposal in response to the January 2013 DOE VTO FOA.
- The goal is to develop computer-aided engineering tools to accelerate development of safer lithium-ion batteries.

Relevance – Project Objectives

This project addresses two abuse conditions: "crush" and "thermal ramp" of cells

- The main objective is to develop a model to couple the electrochemical-thermal (ECT) behavior of a lithium-ion cell to its structural behavior after rapid mechanical deformation
- A second objective is to develop a model to predict the thermal response of cells to thermal ramp
- A supporting objective is to make the models compatible with CAEBAT-1 and its Open Architecture Software (OAS) for wider proliferation of their use.

Milestones – Met (✔) or On-Track

Milestone Title		Due Date
Place subcontracts with MIT	✔	December 2013
Place Subcontract with ANSYS		December 2013
Develop user-interface software for running chemical kinetics model		August 2014
Document user-interface software and demonstration of CAEBAT-1 compatibility		November 2014
Demonstrate compatibility of chemical kinetic model with OAS		February 2015
Document validation of chemical kinetic abuse model and computational model performance		April 2015
Chemical kinetic abuse model input files available on CD and/or publicly accessible website		June 2015
Document results of mechanical deformation experiments	✔	April 2014
Document validation and computational mechanical deformation and damage model		September 2014
Document simulation results of ECT response of mechanically crushed cell		October 2014
Document validation and computational model performance		April 2015
Document user interface for importing mechanical-ECT model to CAEBAT-1 platform		June 2015
Document implementation of mechanical-ECT software with Oak Ridge National Laboratory's (ORNL's) OAS		September 2015

Approach – Thermal Ramp

Transfer NREL chemical kinetic abuse model to ANSYS CAEBAT-1 platform

- Model chemical kinetics reactions happening in lithium-ion cells at elevated temperatures, including
  - Solid electroplate interface (SEI) decomposition
  - Negative-solvent reactions
  - Positive-solvent reactions
  - Electrolyte decomposition
- Include actual internal geometry of the cells
- ANSYS to create user-friendly interface to access the above chemical reaction model in CAEBAT-1 platform
- Make the model/tool compatible with OAS

Approach – Cell Crush Modeling

Couple MIT's mechanical model with NREL's ECT model

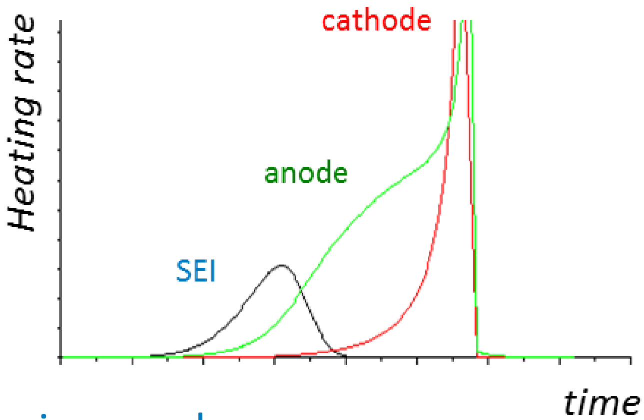
- Simulating simultaneous mechanical, electrochemical, and thermal response of a cell due to crush is very complex and requires modeling simplifications
- Our approach is to assume that crush is rapid (e.g., the cell is damaged in less than a fraction of a second)
- We also assume that electrochemical and thermal response of a cell takes longer than seconds
- This allows us to couple the mechanical aspect with the thermal aspect in a sequential, one-way fashion

Accomplishments – NREL Thermal Ramp

NREL's Abuse Reaction Kinetics (ARK) model has been documented and shared with ANSYS

NREL's ARK Model

- Empirical reaction model
  - Built from calorimetric test data
- Evaluates the rate of heat from exothermic decomposition reactions



Bottom-up approach

- Abuse reactions among cell components superimposed
- Incorporated exothermic component reactions commonly addressed; readily accommodates other abuse reactions

Model Equation – Box Model

$$\begin{cases} S = HW\dfrac{d\alpha}{dt} \\ \dfrac{d\alpha}{dt} = k(T)f(\alpha) \\ k(T) = Ae^{-\frac{Ea}{RT}} \\ f(\alpha) = \alpha^m(1-\alpha^n)(-\ln(1-\alpha))^p \end{cases}$$

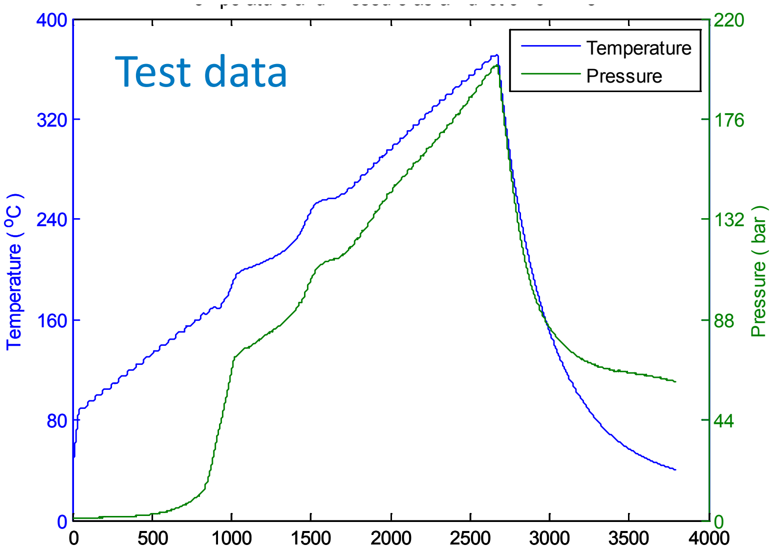
S : volumetric reaction heat
H : heat of reaction
$d\alpha/dt$: reaction rate
$k(T)$: temperature-dependent rate constant

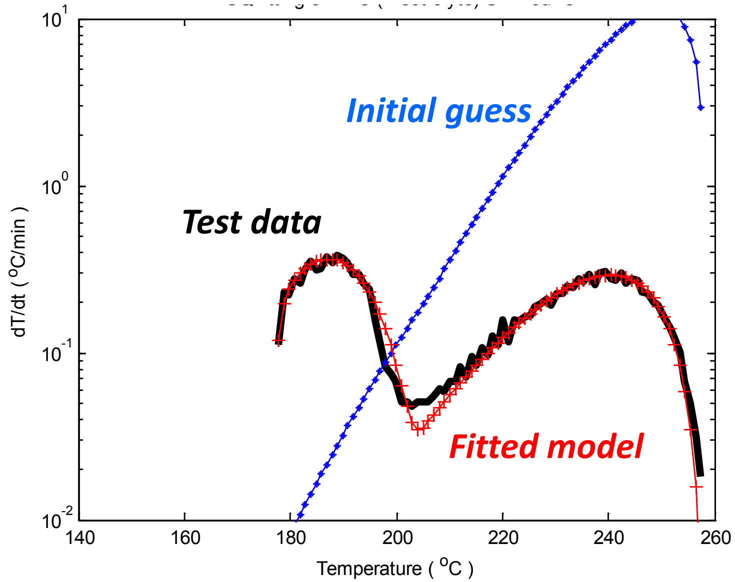
NREL | MIT Massachusetts Institute of Technology | ANSYS

Accomplishments – NREL Thermal Ramp

Developed a user friendly tool for parameter identification for the ARK model

Example: *Identifying electrolyte decomposition reaction parameter from ARC data*





ARC: Accelerating Rate Calorimeter

- In-house ARC tests carried out for a type of lithium-ion battery electrolyte
- Superposition theorem is used to isolate the peaks; fit these two peaks individually

	aE	rK	m	n	p
Initial	2.74E5	5.14E25	1	0	0
R1	2.53E5	5.14E25	3	0.3762	0.2430
R2	2.80E5	5.14E25	0	0.0991	2.1145

Note: The curve shown above is not a unique solution

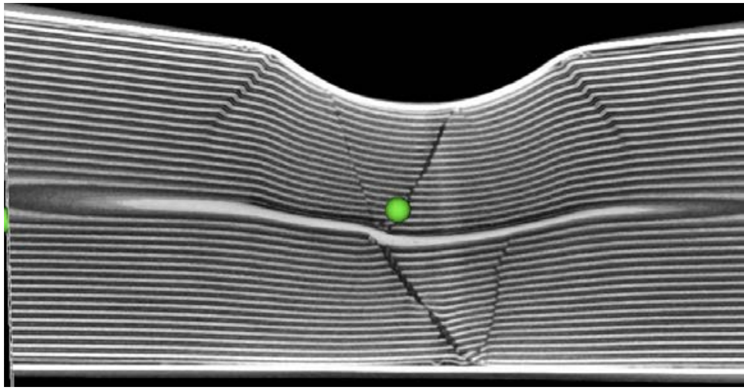
Accomplishments – ANSYS

- At the time this presentation was prepared, the ANSYS subcontract was still being negotiated
- We expect the subcontract to be finalized and signed by mid May 2014
- Although ANSYS has not started work at the time this presentation was prepared, we do not think this will adversely affect the future milestones/deliverable for this project

Accomplishments – MIT Crush

In order to couple the existing ECT model with the mechanical model of a cell, it is necessary to determine:

- Location of mechanical failure within the jellyroll
- Crack orientation
- Size of the fractured region

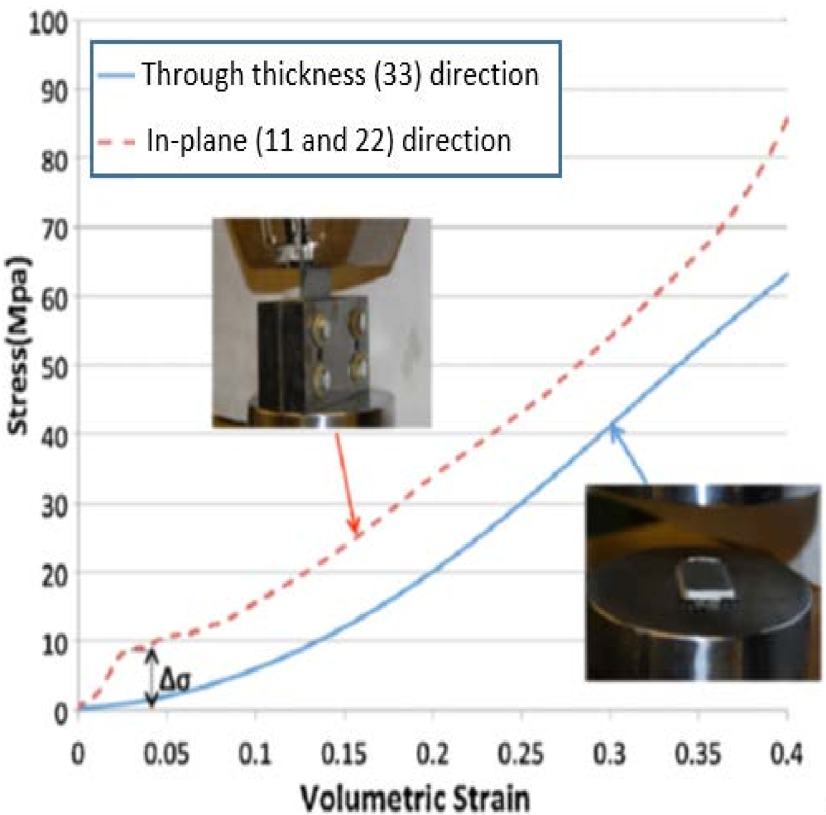


This information is provided by the new enhanced anisotropic model of the jellyroll

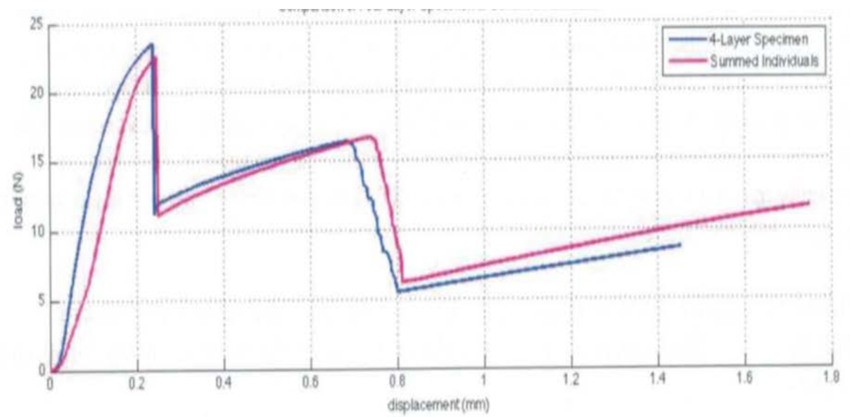
Accomplishment – MIT Crush

Calibration of a Single Cell

Compression tests in in-plane and through thickness directions



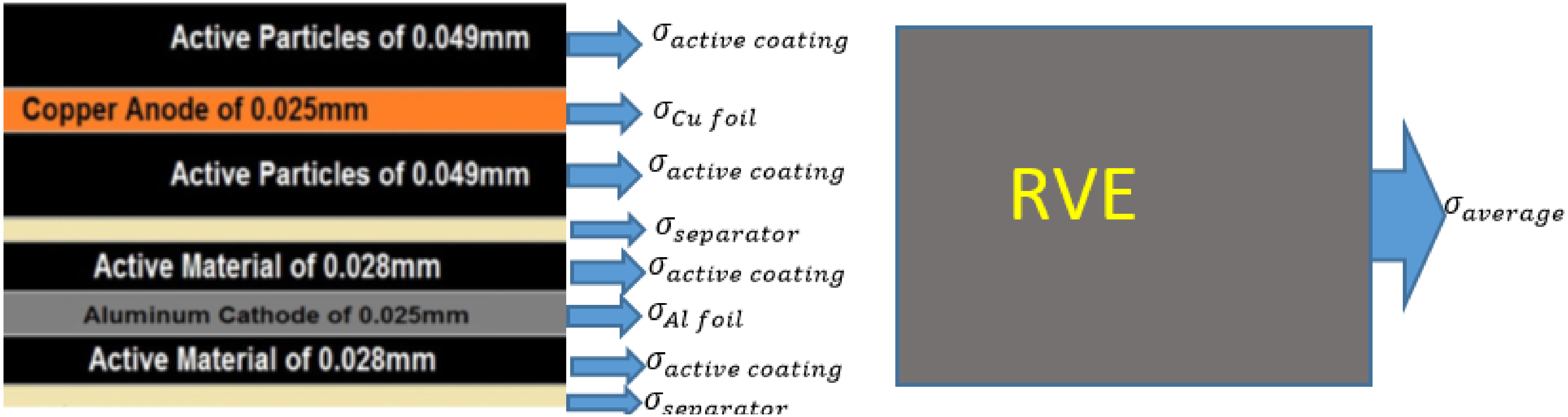
Tensile tests on components of the electrode/separator assembly



Accomplishment – MIT Crush

Anisotropic Homogenized Material Model

Separate tests should be done in machine, transverse, through thickness directions



$$\sigma_{average} = \frac{\sum \sigma_l t_l}{\sum t_l}$$

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

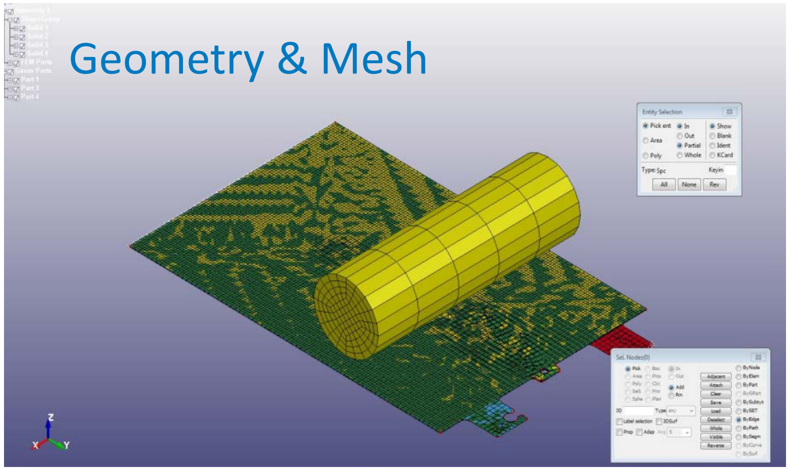
$$\sigma_l = [\ \sigma_{Al\ foil}, \sigma_{Cu\ foil}, \sigma_{active\ coating}, \sigma_{separator}\ ]$$

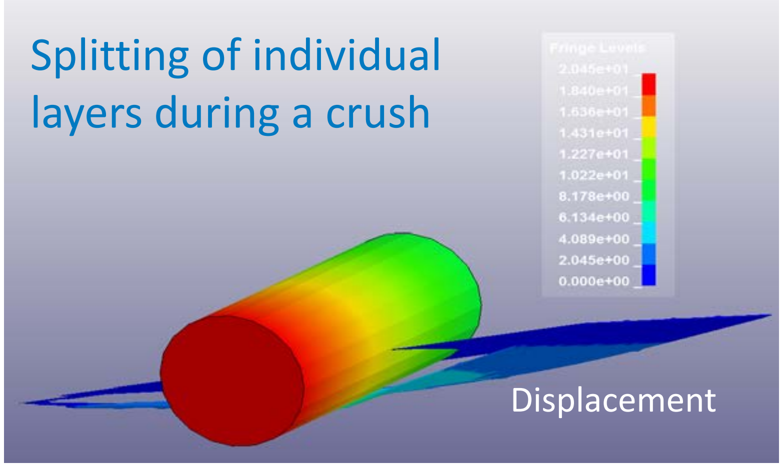
$$t_l = [\ t_{Al\ foil}, t_{Cu\ foil}, t_{active\ coating}, t_{separator}\ ]$$

NREL MIT Massachusetts Institute of Technology ANSYS

Accomplishments – NREL Crush

- **Software installed on NREL cluster and first round of models set up**
- **To facilitate integration of the different models, a "“test-case” crush simulation is performed at NREL**
- **This set of simulations will help:**
  - Establish what parameters must be exchanged among the models
  - Compute local heat generation rates in an arbitrary deformed cell-geometry
- **When crush models from MIT become available, NREL will integrate the ECT models to the deformed geometry**



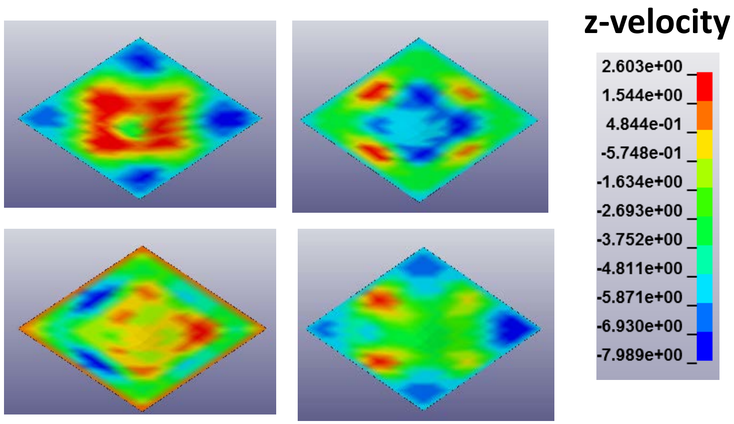


Crush-simulation on an electrode pair: for different crush bar orientations, the contact between individual layers is simulated

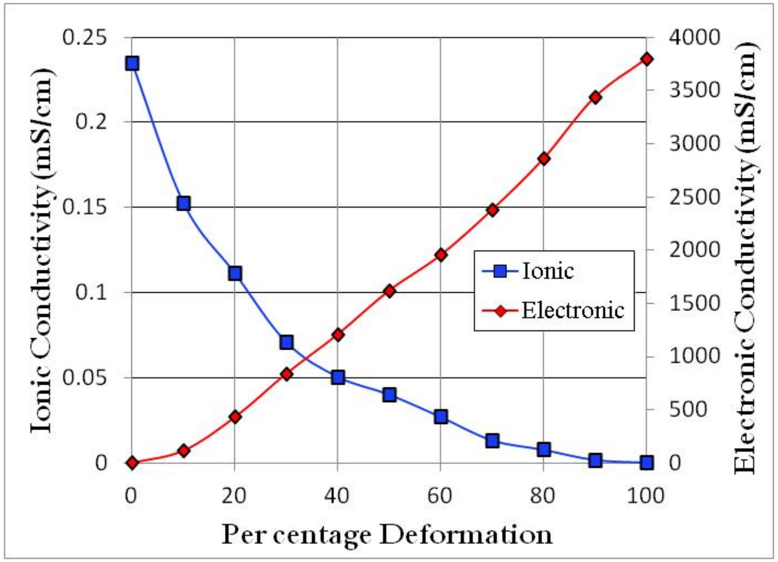
Accomplishments – NREL Crush (Contd.)

Resistivity vs. Deformation:

- Select deformation threshold for electrical contact to be formed
- Generate deformed geometry
- Compute effective resistance as function of percentage deformation
- Develop simplified relationships to scale up results from individual layers across the cell
- Include simplified expressions in cell-level ECT simulations



Electrical contact is predicted using the local z-velocity and the deformation threshold



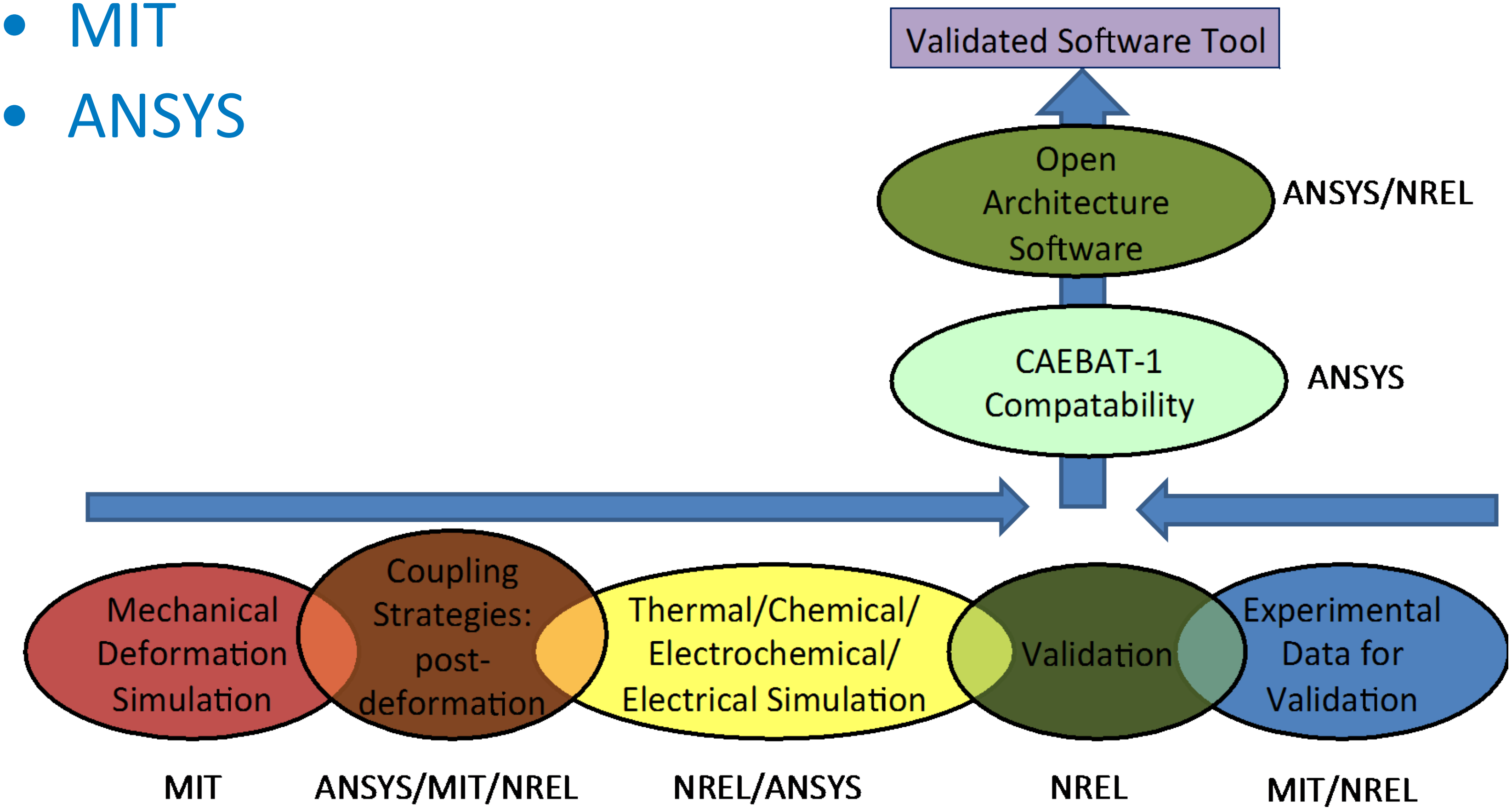
Change in resistivity during crush

Responses to Previous Year Reviewers' Comments

This project is new and was not presented at AMR in FY2013.

Collaborations and Coordination

- NREL Energy Storage Team (lead)
- NREL High Performance Computing Team
- MIT
- ANSYS



Remaining Challenges and Barriers

- "Multi-layer puncture" approach for loose coupling of mechanical deformation to ECT behavior may not be automatable to capture damage zones created by different crush loads and orientations
- Model validation with experimental data may take longer than expected; simulation may not represent a particular experiment, resulting in repeated simulation, and insufficient time and budget for further validation
- Integration of new code with emphasis on mechanical/structural/chemical physics into ORNL's OAS

Future Work

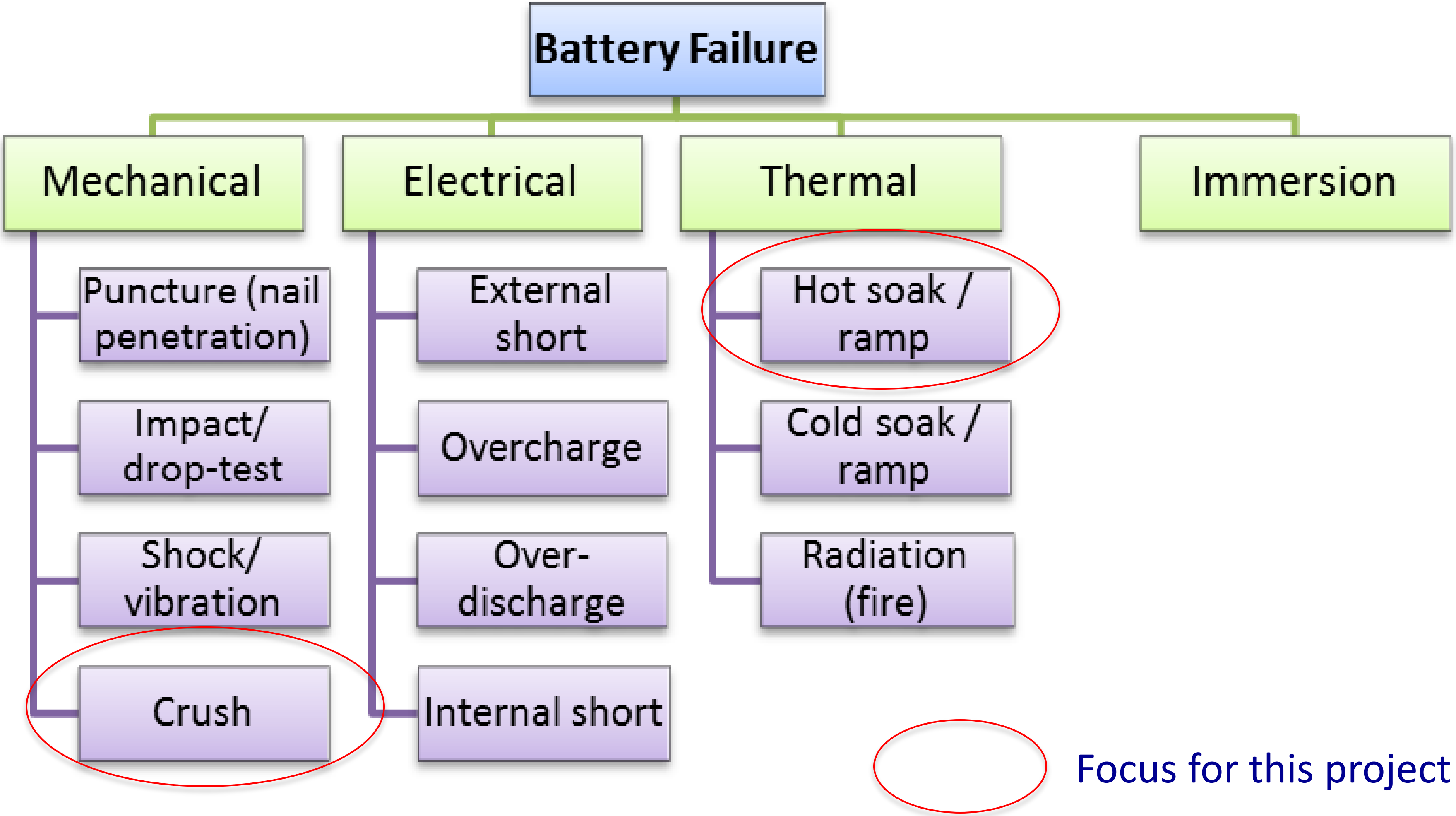
- Transfer Existing Chemical Kinetic Abuse Model to Current CAEBAT-1 Platform, Maintaining Compatibility with OAS Framework
- Validate Software from Task 2 in Thermal Ramp Abuse Conditions against Published Experimental Data
- Create Validated Mechanical Deformation (Crush) Computational Models
- Couple 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

- This project was funded based on the team's proposal to the 2013 DOE VTO FOA
- This project is focused on coupling models of crush and thermal runaway of cells
- Eventual goal is to be able to predict the onset of thermal runaway after crash-induced crush or thermal ramp
- MIT is under subcontract; ANSYS subcontract is pending
- We have documented our chemical kinetics reaction model for incorporation into the ANSYS CAEBAT-1 platform (FLUENT-15)
- A procedure is being developed for an anisotropic model for the jellyroll with testing and calibration
- An approach to link mechanical deformation to short resistivity was created

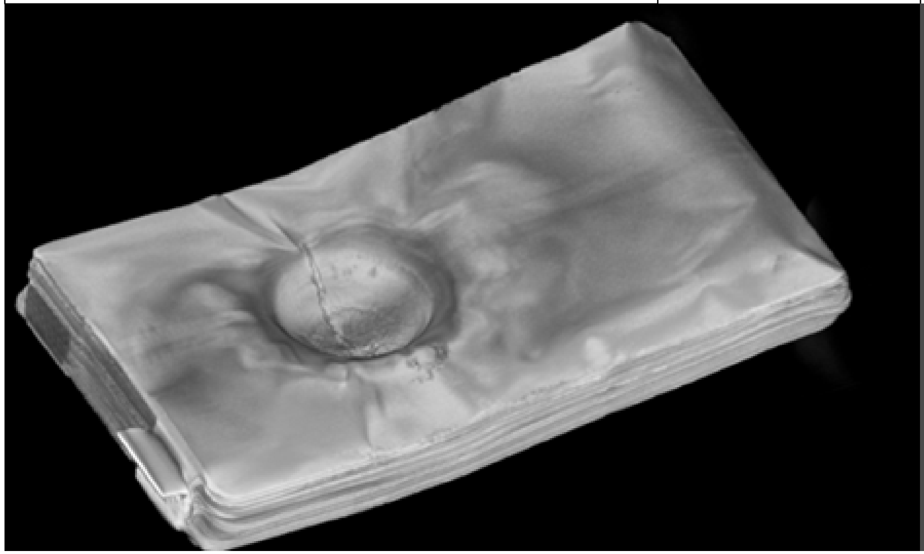
Back Up Slides

Causes of Battery Failure

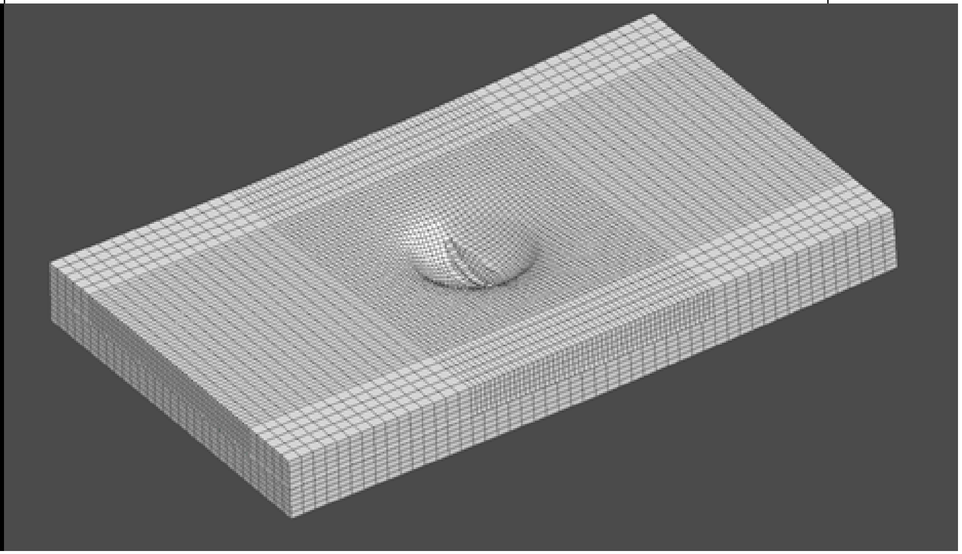


Crash Formation under Hemispherical Punch

Crack as seen in CT Scan

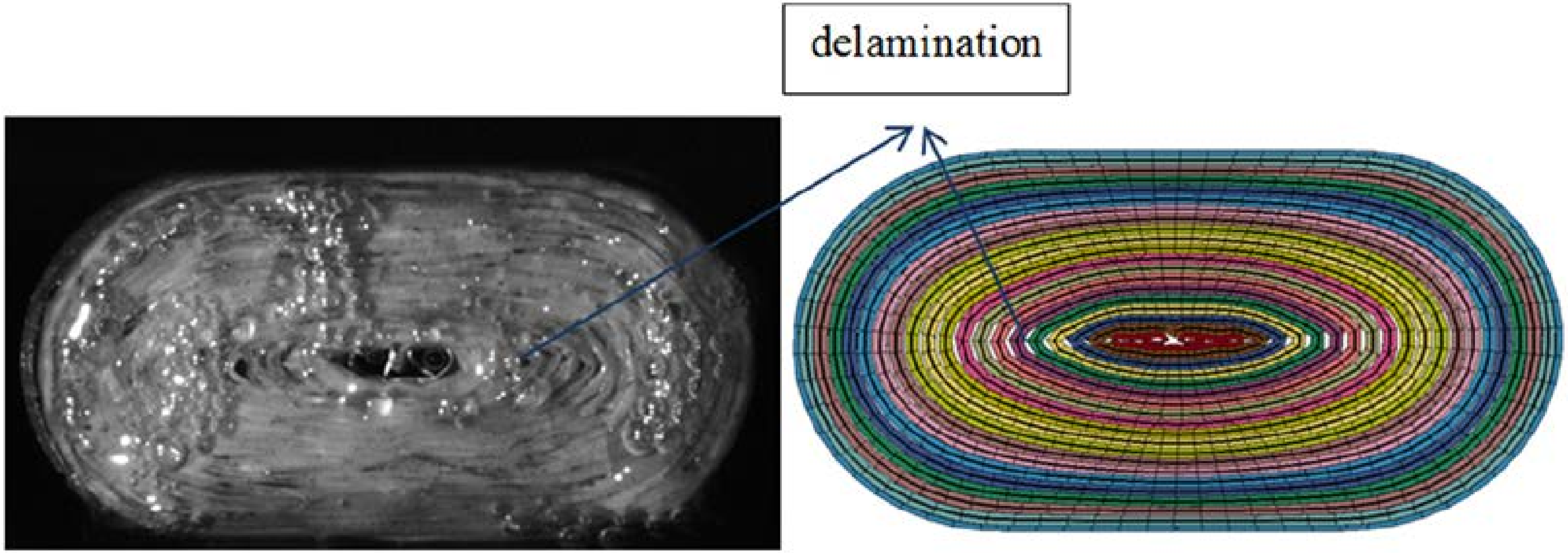


Crack as seen in FE simulation



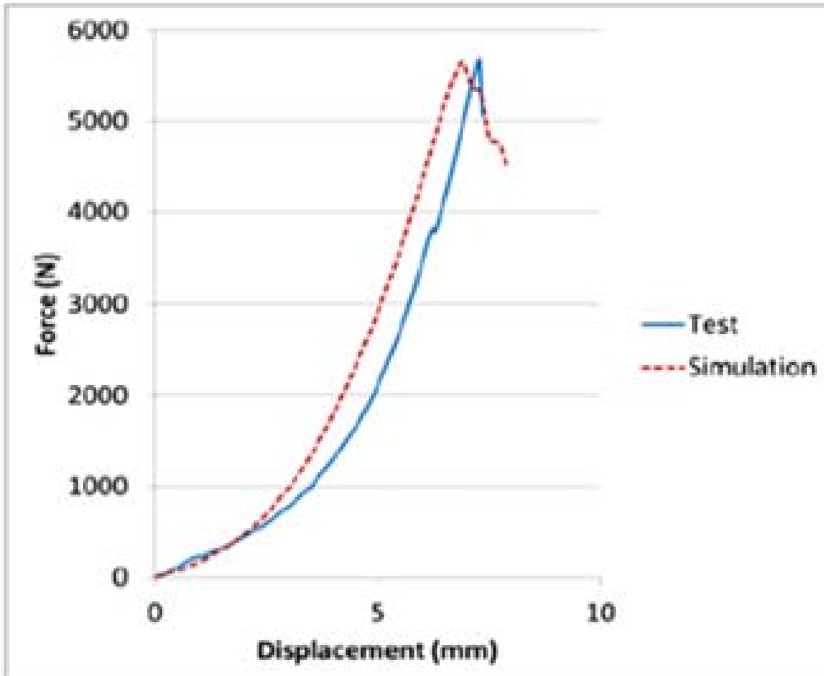
Demonstrating MIT's capabilities of the element-deletion technique:
The figures above compare the finite element (FE) prediction with an actual crack observed by CT scan of a cell under load; good results were obtained by choosing a proper value of fracture strain (maximum principal tensile strain)

Partially Crushed 18650 Cell between Rigid Plates



Crush experiments and FE simulation performed at MIT

LS-Dyna Simulation of 18650 Cell Crush



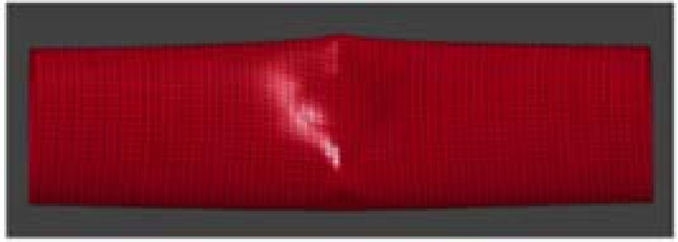
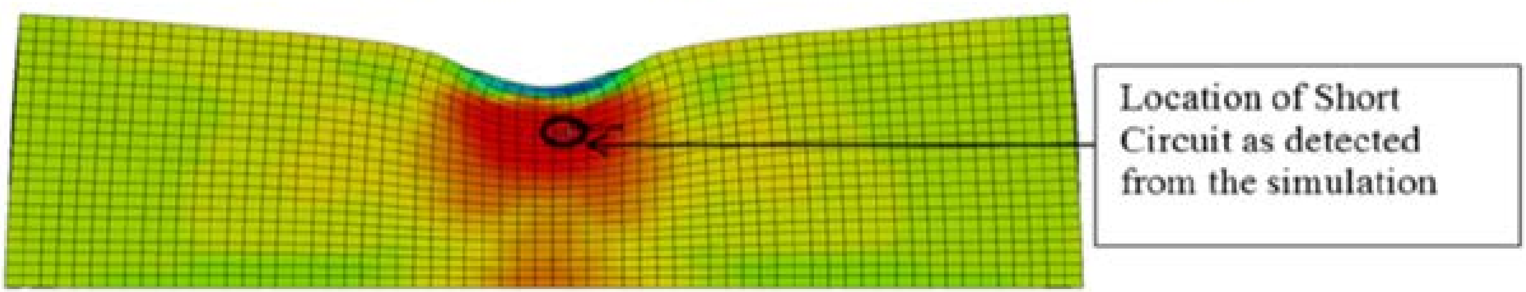
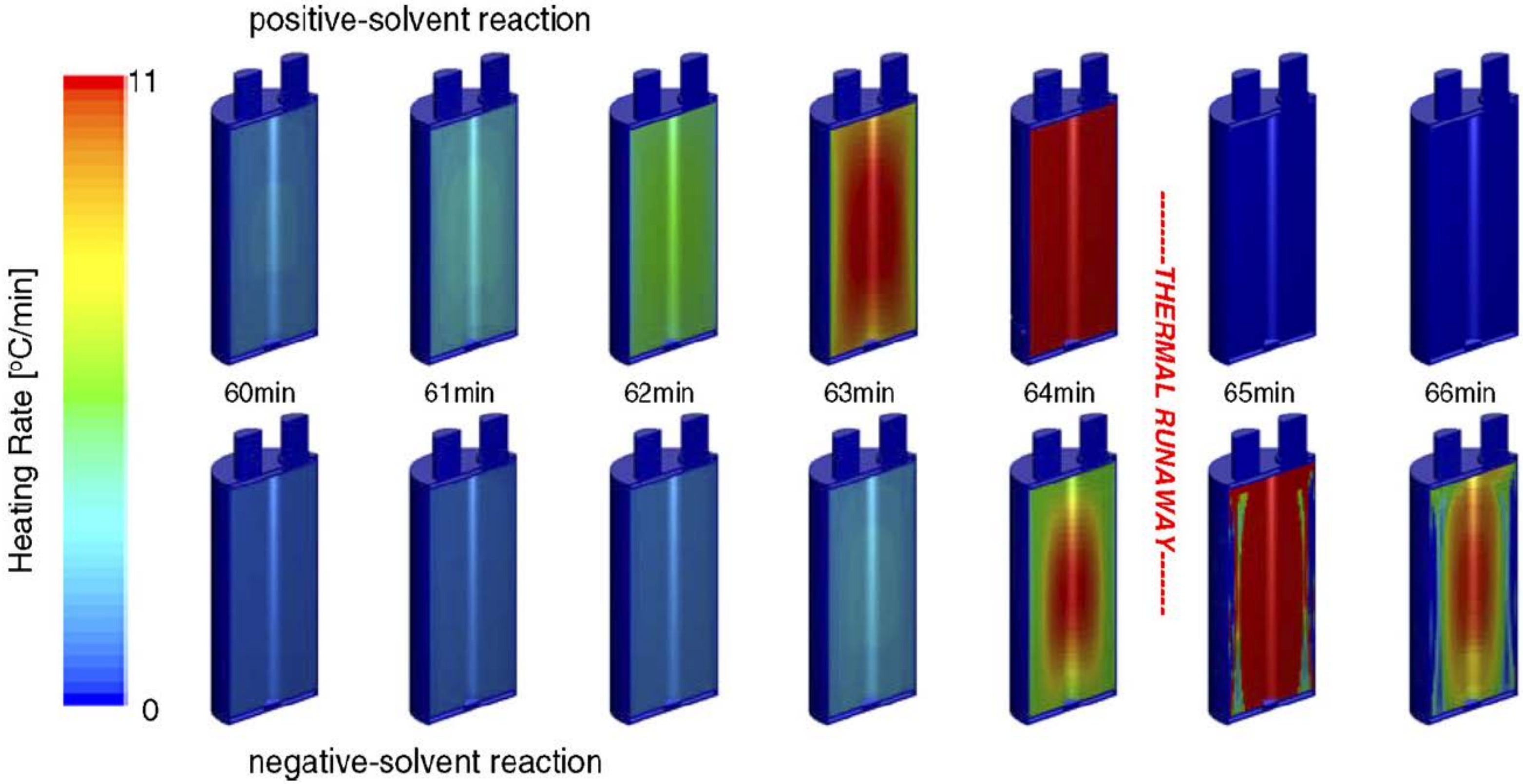


Fig. 16. Comparison of tests and simulation for the hemispherical punch crush loading.



Typical Results of NREL Chemical Kinetics Reaction Model

Electrode active material/solvent reactions trigger thermal runaway in a $LiCoO_2$/graphite cell



NREL MIT Massachusetts Institute of Technology ANSYS