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Efficient Simulation and Abuse Modeling of Mechanical-Electrochemical-Thermal Phenomena in Lithium-Ion Batteries

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National Renewable Energy Laboratory

*Now On detail to Vehicle Technologies Office

Joshua Lamb
Sandia National Laboratories

Daniel Abraham, Pierre Yao, Dennis Dees
Argonne National Laboratory

Overview

This project was awarded in response to VTO FY15 Lab Call.

Timeline

- Project start date: Oct. 2015
- Project end date: Sept. 2018
- Percent complete: 60%

Budget

- Total project funding: \$ 3.15M
 - DOE share: 100%
- Funding received in FY 2016:
\$1.05 M
- Expected Funding for FY 2017:
\$1.05 M

Barriers

- Gap between modeling tools and cell design process in the industry
- Lack of simulation tools integrating mechanical failure and abuse response of batteries for practical assessment of battery safety
- Limited understanding of complex failure mechanisms resulting in expensive over-design of batteries

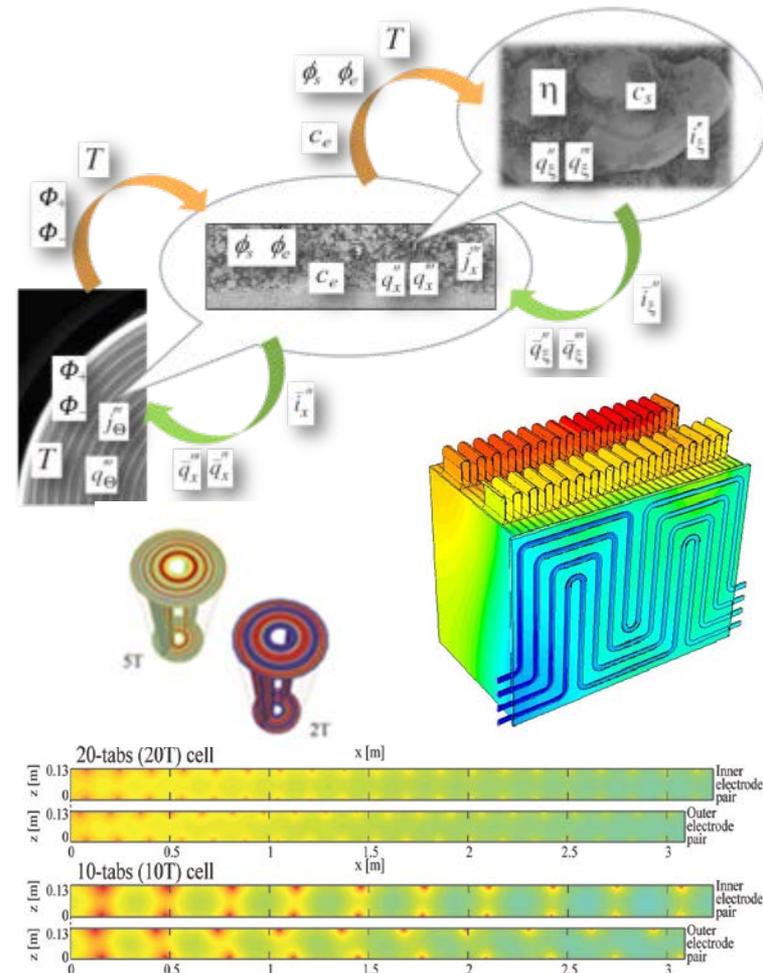
Partners

- Argonne National Laboratory (ANL)
 - Pouch Cells and data for parameter estimation
- Sandia National Laboratories (SNL)
 - Cell-level mechanical abuse testing for validation of mechanical models
- Forming Simulation Technologies, Ohio State University, George Mason University
 - Integration with ANSYS and LS-DYNA

Relevance

Background and Motivation

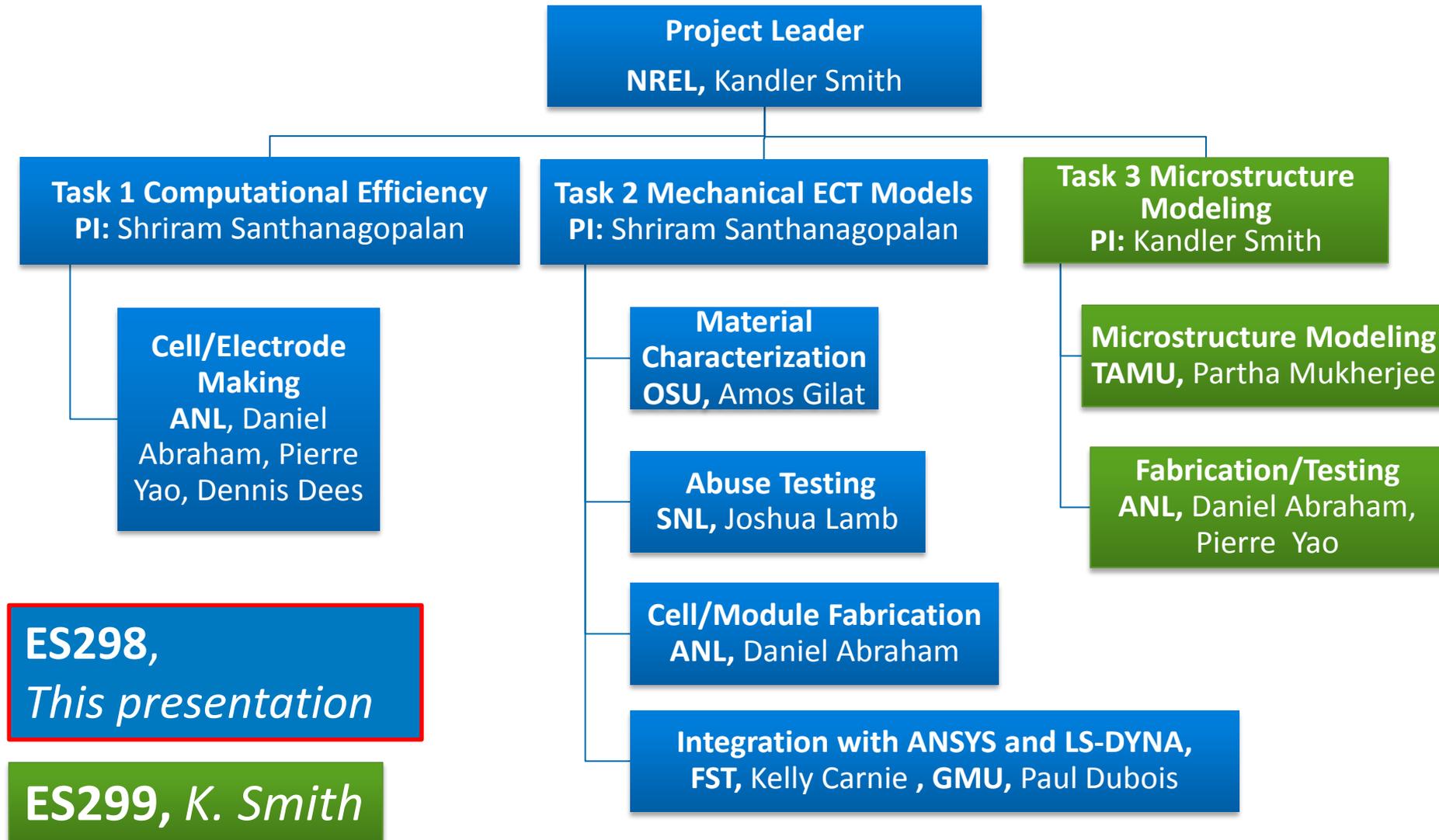
- VTO launched the Computer-Aided Engineering of Batteries (CAEBAT) project to develop validated modeling tools to accelerate development of batteries, in support of vehicle electrification R&D to reduce dependence on imported oil.
- Over 40 different end users from the community have adapted the Multi-Scale Multi-Domain (MSMD) modeling approach developed under CAEBAT.
- Feedback from the first few sets of end-users has helped us identify priorities that will enable wider use of model-based design:
 - Standardize identification of the model parameters
 - Increase computational efficiency
 - Extend the models to include mechanical failure of cells and packaging components
 - Close gaps between materials R&D and CAEBAT modeling tools



MSMD models previously developed in CAEBAT have been widely adapted in the community and helped us identify gaps.

Project Structure

Project Title: Computer-Aided Battery Engineering Consortium



Relevance

Objectives for March 2016 – March 2017

Computational Efficiency:

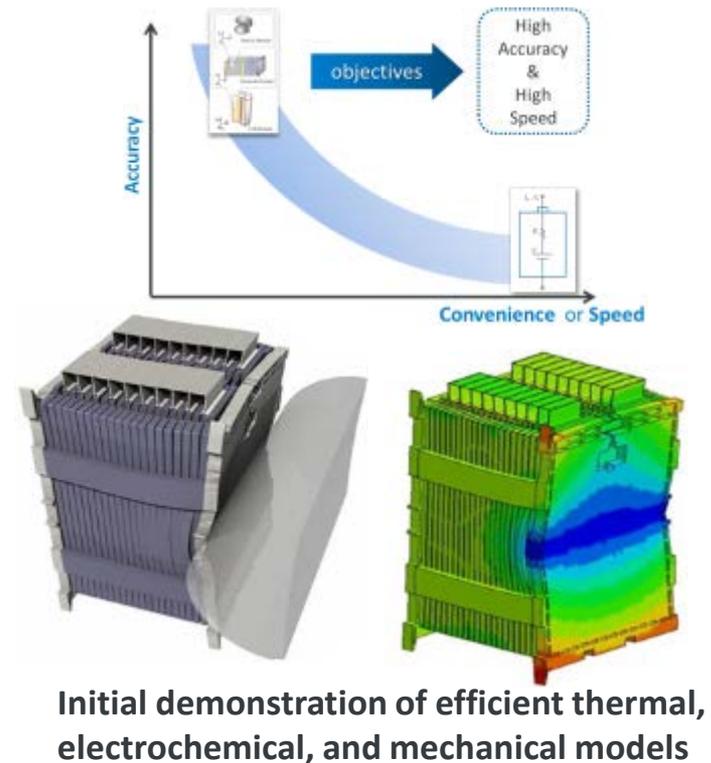
- Demonstrate 1000-fold increase in computational speed using model order reduction methods
- Document efficiency enhancement approach for deployment on to commercial software platforms

Parameter Identification:

- Develop and document the procedure to extract parameters for the MSMD models

Mechanical Models:

- Present initial demonstration of simultaneous coupling between mechanical failure and the thermal response of the cell during a crush test



- Impact:** By making disruptive CAE design tools available on desktop computers for use by the battery community, this effort supports the following goals identified by the VTO:
1. Expedite path to \$ 125/kWh electric vehicle (EV) battery costs by drastically reducing the number and duration of battery design cycles in the industry
 2. Reduce module/pack costs by maximizing insight gathered on failure modes in batteries, from a limited subset of tests currently performed

Milestones

	Milestone Name/Description	Deadline	Milestone Type	Status
Comp. Effic.	M 1.1 Draft summary documentation of GH-MSMD framework	8/31/2016	Qrt. Prog. Meas.	Done
	M 1.2 Validate GH-MSMD using half cell data from ANL	1/31/2017	Qrt. Prog. Meas.	Done
	M 1.3 Present at the DOE Annual Merit Review	6/30/2016	Qrt. Prog. Meas.	Done
	M 1.4 Perform out design evaluation and performance evolution study using newly developed multiphysics GH-constituent models	7/31/2018	Qrt. Prog. Meas.	On track
Mech. Abuse	M 2.1 Demonstrate simultaneous coupling in MECT model that shows interaction of mechanical deformation with the thermal response of the cell under different strain-rates within 10% error against data	3/31/2016	Annual SMART (Go/No-Go)	Go
	M 2.2 Detailed documentation describing the mechanical tests procedure for development and validation of constitutive models for individual battery components and battery cells with < 5% error on the mechanical response at the component level between data and models	7/31/2017	Annual SMART (Go/No-Go)	On track
	M 2.3 Interim update on mechanical models demonstrating damage propagation across multiple axes of battery cells and battery modules	12/31/2017	Qrt. Prog. Meas.	On track
	M 2.4 Report summarizing model validation for MECT simulations	4/30/2018	Qrt. Prog. Meas.	On track

Task 1 - Computational Efficiency

Approach

Step 1: Nonlinear Multiscale Implicit Formulation

$$\phi = f(i; \mathbf{x}, \mathbf{p})$$

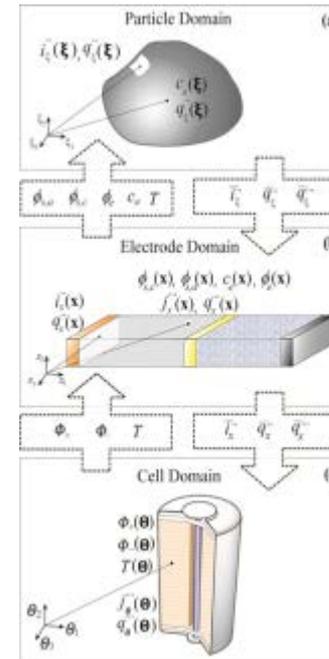
Step 2: Timescale Separation & Variable Decomposition

$$\phi = g(i; \mathbf{x}, \mathbf{p}) + h(i; \mathbf{x}, \mathbf{p})$$

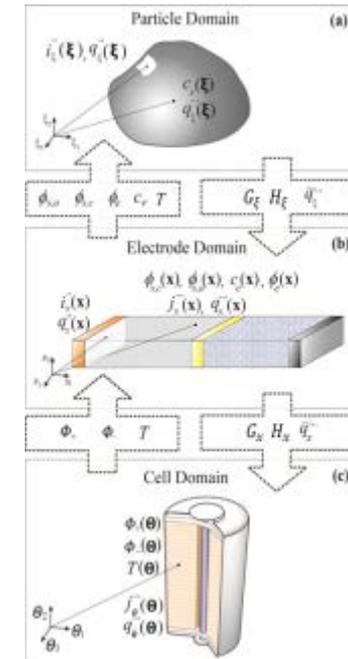
Step 3: Partial/Selective Linearization

$$G(i; \mathbf{x}, \mathbf{p}) = \frac{dg}{di}$$

$$\phi = G(i; \mathbf{x}, \mathbf{p})i + H(i; \mathbf{x}, \mathbf{p})$$



MSMD (Previous work)



GH-MSMD (New)



Efficient and Extensible Quasi-Explicit Modular Nonlinear Multiscale Battery Model: GH-MSMD

Gi-Heon Kim, Kandler Smith,^{*,z} Jake Lawrence-Simon, and Chuanbo Yang^{*}

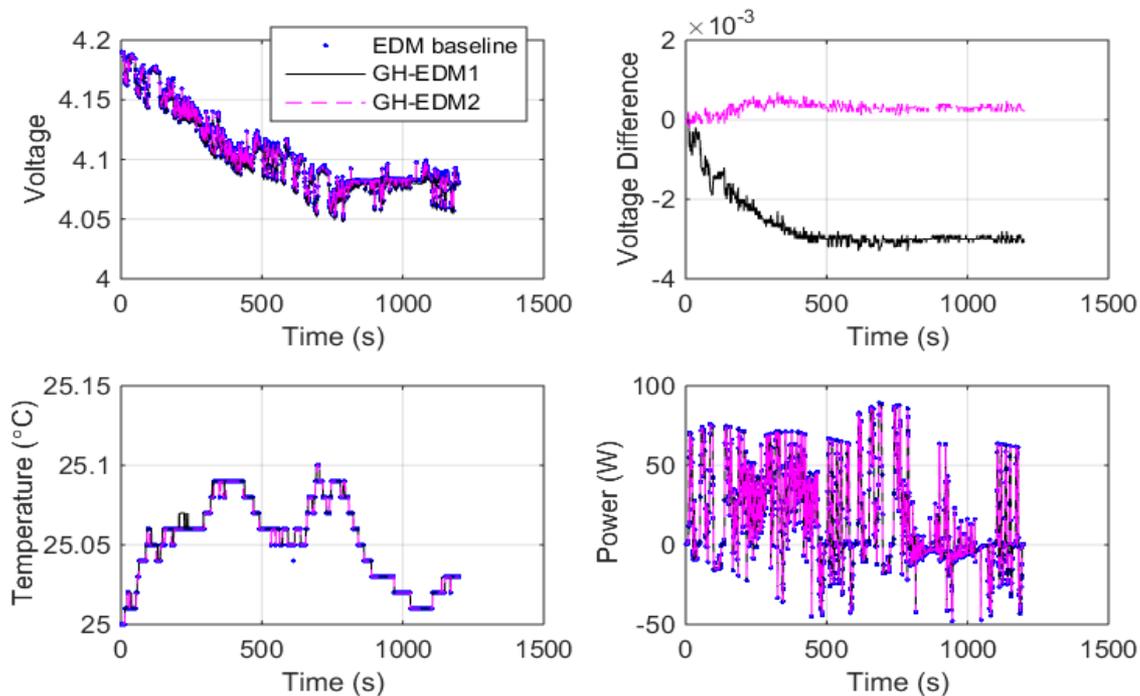
National Renewable Energy Laboratory, Golden, Colorado 80401, USA

Complex physics and long computation time hinder the adoption of computer aided engineering models in the design of large-format battery cells and systems. A modular, efficient battery simulation model—the multiscale multidomain (MSMD) model—was previously introduced to aid the scale-up of Li-ion material & electrode designs to complete cell and pack designs, capturing electrochemical interplay with 3-D electronic current pathways and thermal response. This paper enhances the computational efficiency of the MSMD model using a separation of time-scales principle to decompose model field variables. The decomposition

G.-H. Kim et al., *J. Electrochem. Soc.*, A1076-88 (2017)

Technical Accomplishments and Progress

The selective G-H linearization approach drastically reduces computational burden!



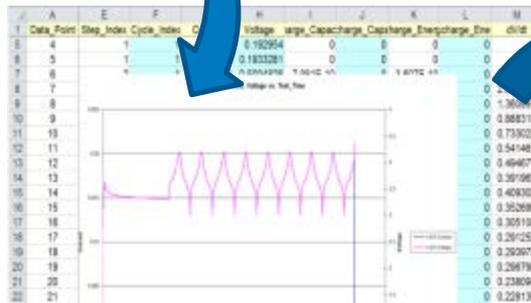
Simulation case		Computation time for Electrode Domain Models (EDM) in seconds		
Load Profile	Temperature ($^{\circ}\text{C}$)	EDM baseline	GH-EDM1	GH-EDM2
1C	25	360.13	3.03	0.44
1C	0	816.21	3.50	0.47
Drive cycle	25	1205.92	7.06	0.83
Drive cycle	0	8786.45	45.00	1.27

Model Parameter Identification Workflow



Experimental set up to cycle cells for collecting data

Pre-processing and filtering of raw data



Format data from native formats for battery cyclers

Setup baseline MSMD Inputs

$$\frac{\partial}{\partial x} \left(\frac{D_s}{L} \frac{dc_s}{dx} \right) + \frac{\partial}{\partial x} \left(\kappa_{eff} \frac{\partial}{\partial x} \ln c_s \right) + j_x''' = 0$$

$$\kappa_{eff} = \kappa \varepsilon_p^0$$

$$\kappa_{eff}^0 = \frac{2RT\kappa_{eff}}{F} (t_0^0 - 1) \left(1 + \frac{d \ln f_{\pm}}{d \ln c_s} \right)$$

$$\frac{\partial(\varepsilon_p c_p)}{\partial t} = \frac{\partial}{\partial x} \left(D_p^{eff} \frac{dc_p}{dx} \right) + \frac{1-t_0^0}{F} j_x''' - \frac{i_s^0}{F} \frac{\partial t_0^0}{\partial x}$$

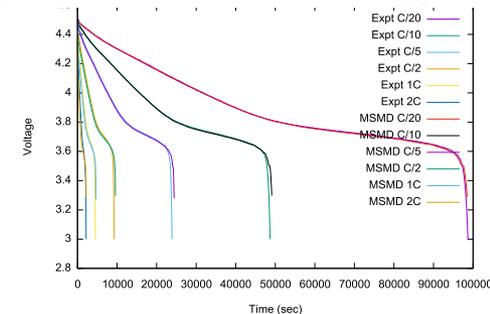
MSMD-Model

Fitting of model to data

- Python script parses data to meet model needs
- Parameter estimation based on Levenberg-Marquardt algorithm
- Workflow independent of model(s)/data set(s)
- Can use the same approach for multiple models and/or datasets – as long as the list of inputs and outputs are standardized (e.g., using the OAS)
- Process can be easily wrapped with a GUI as workflow stabilizes

$$C_{Li}^{max} = 4.905037e+04 \pm 7.067772e+01 \text{ mol/m}^3$$

$$D_s^{ref} = 4.392799e-15 \pm 2.562774e-17 \text{ m}^2/\text{s}$$



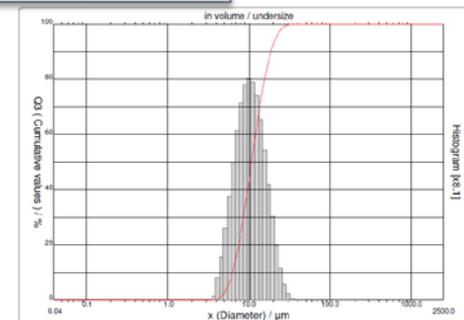
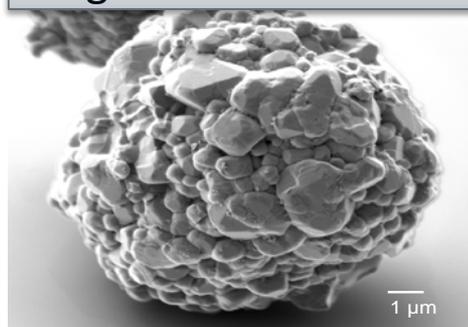
Calibrated Model and Parameters

Material Data and Cells from CAMP Facility at ANL

Material specifications for the cell components provided by ANL include:

- Electrode composition
- Thickness, porosity, loading density
- Particle size and distribution
- Current collector thicknesses

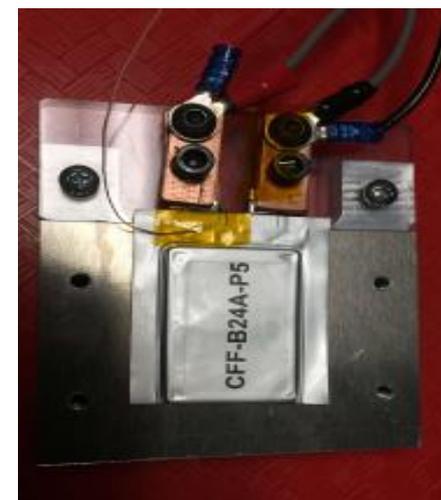
Figure Credit: Dan Abraham, ANL



~15 μm sized secondary particles contain many primary particles

A-C015(+) is matched to A-A002A(-) for 4.4 V full cell cycling

A-C015(+): made by CAMP (NCM523)	A-A002A(-): made by CAMP
Positive Electrode:	Negative Electrode:
90 wt% $\text{Li}_{1.03}(\text{Ni}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3})_{0.97}\text{O}_2$	91.8 %wt ConocoPhillips: CGP-A12 graphite
5 wt% C45 (Timcal)	2 wt% C45 (Timcal) + 0.17 %wt Oxalic Acid
5 wt% PVdF binder (Solvay 5130)	6%wt KF-9300 Kureha PVDF binder
9.17 mg/cm ² loading density - coating	5.88 mg/cm ² loading density - coating
8.25 mg/cm ² loading density - active/oxide	5.51 mg/cm ² A12 graphite loading density
33.5% electrode porosity	38.4% electrode porosity
34-μm-thick composite coating	44-μm-thick composite coating
20-μm-thick Al current collector	10-μm-thick Cu current collector



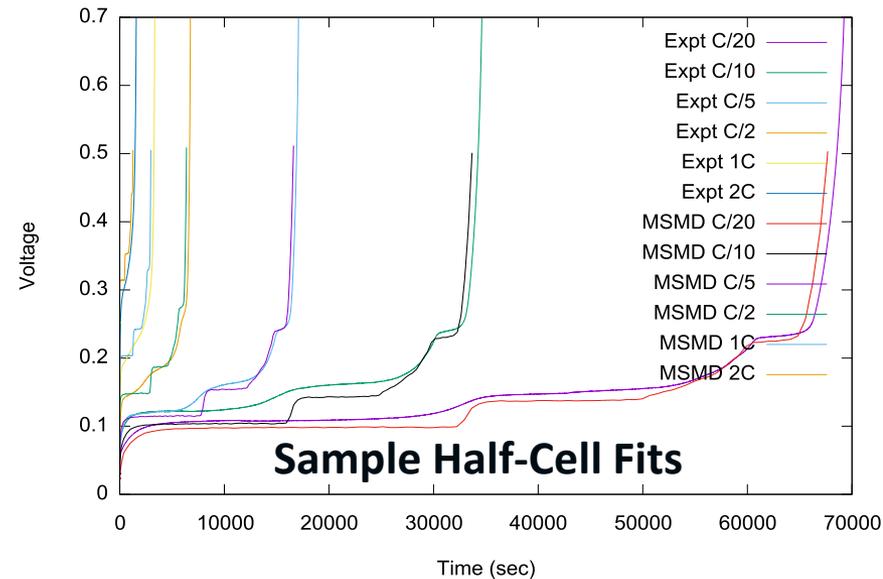
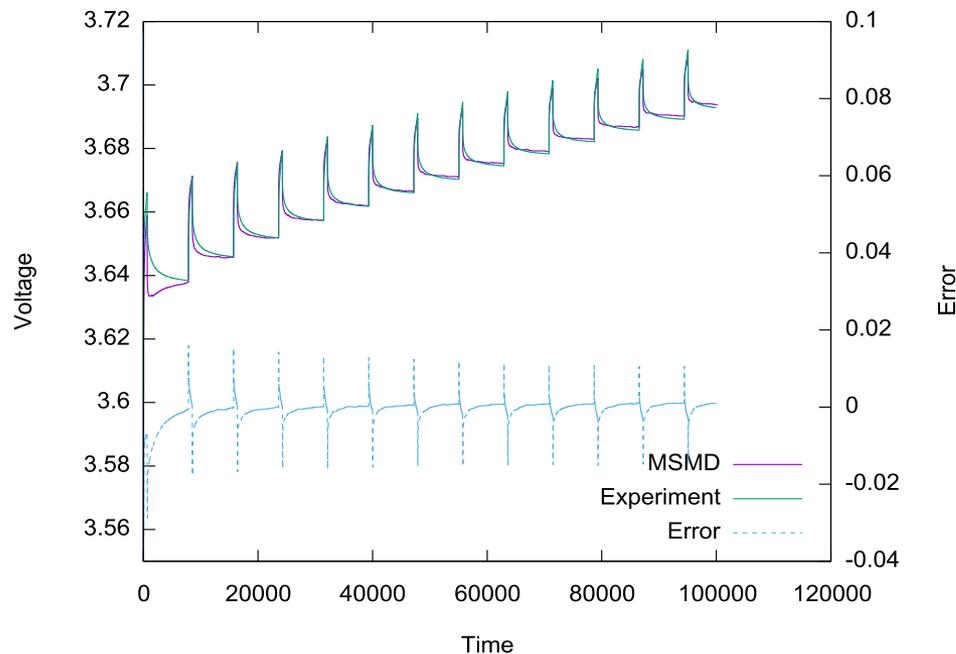
Pouch cells with 300 mAh nameplate capacity

Baseline electrolyte: 1.2M LiPF_6 in EC:EMC (3:7, w/w)
 Baseline separator: Celgard 2325 (trilayer, PP/PE/PP)

CAMP: Cell Analysis, Modeling, and Prototyping

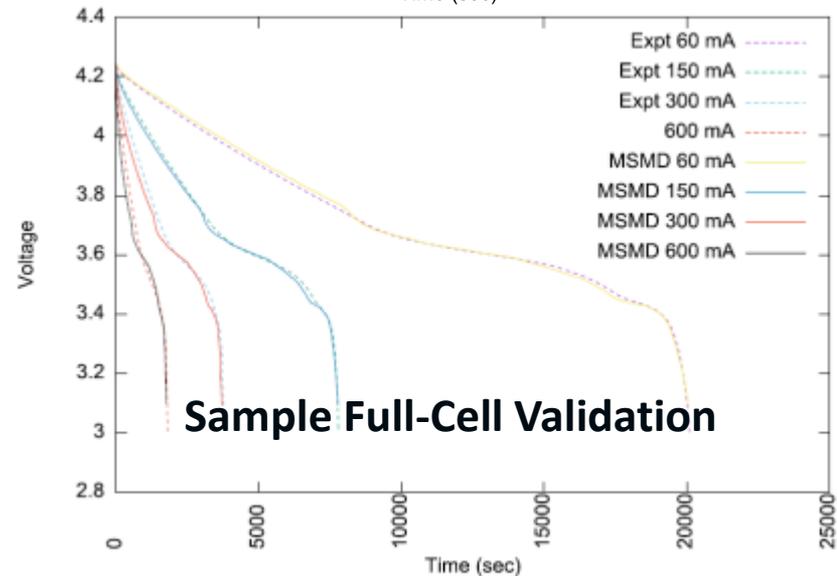
Parameter Identification Results

GITT: Model vs. Data



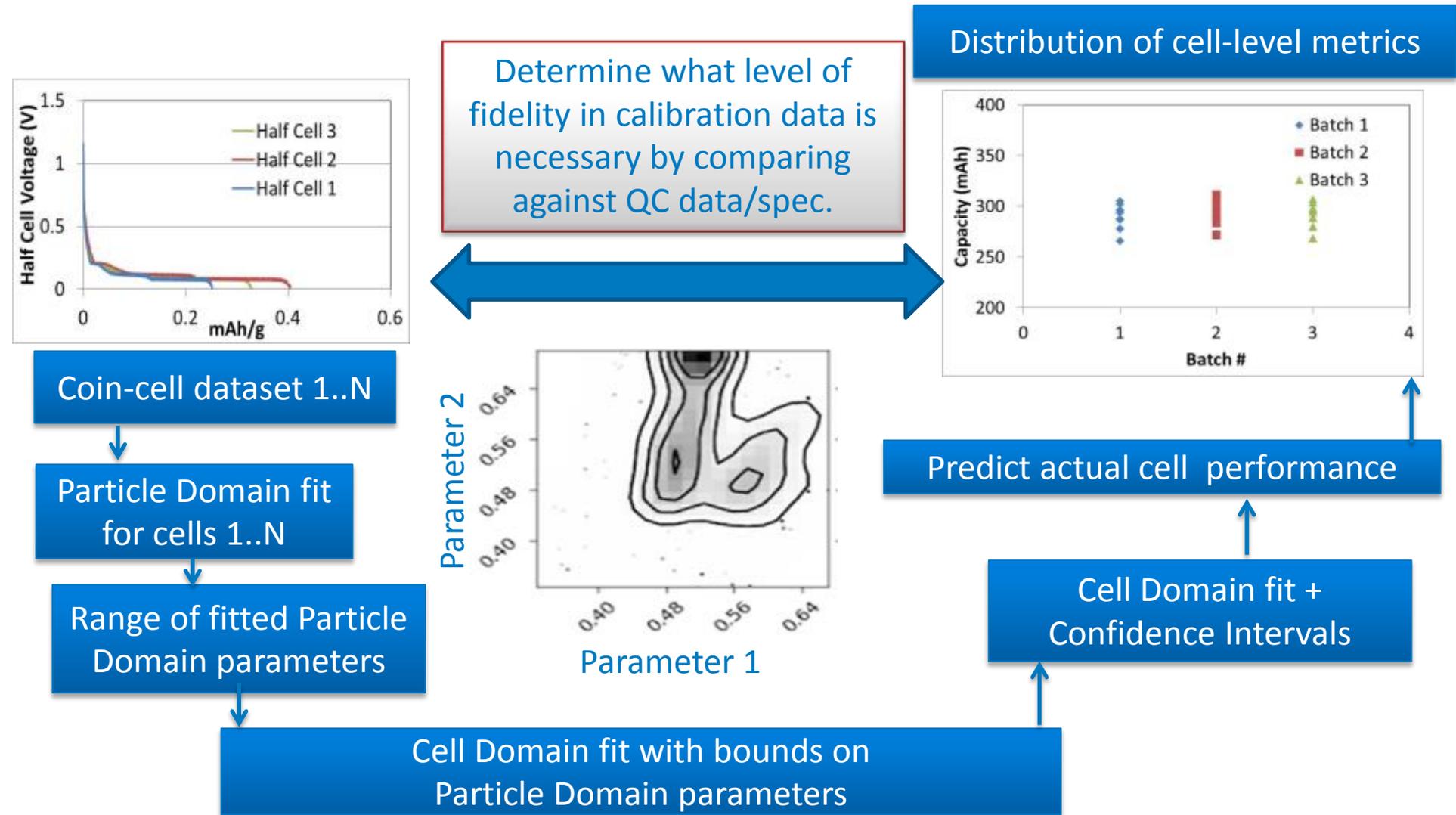
Examples of Parameters and Confidence Intervals

Parameter	Anode	Cathode
C_{Li}^{max} (mol/m ³)	2.9511e+04 ± 2.5377e+02	4.9050e+04 ± 7.0677e+01
D_s^{ref} (m ² /s)	3.015e-15 ± 2.469e-15	4.393e-15 ± 2.5634e-17



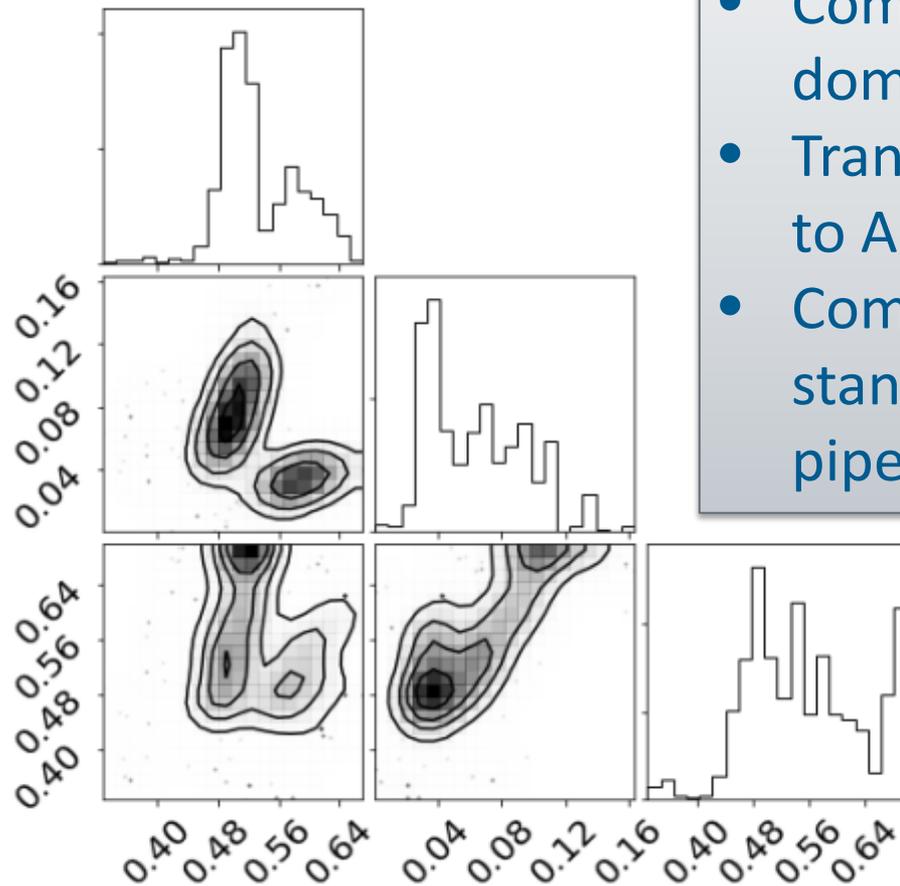
Automated procedure calibrates models with data from cyclers to a max. relative error < 5%

Application: Analysis of Material/Data Quality



Closing the loop between lab-scale calibration data and production cell specs. will reduce development costs by directing improvements to processes that impact on cell quality the most.

Future Work for Task 1



Multi-level Bayesian calibration

Computational Efficiency:

- Complete validation for the cell-domain in FY17
- Transfer GH-MSMD capabilities as UDF to ANSYS Models
- Complete generalization and standardization of the automation pipeline for model identification

Parameter Identification:

- Multiple data sets: what quality of data is needed to induce a given confidence level in the parameters?

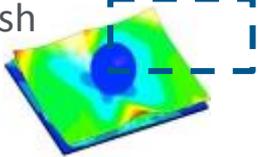
Any proposed future work is subject to change based on funding levels.

Task 2 – Mechanical-Electrochemical- Thermal Modeling of Abuse Phenomena

Mechanical Modeling Approach

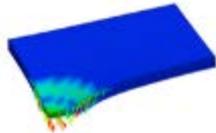
Objective: Predict battery behavior during a crash event to optimize safety and weight reduction

Displacement under
Crush



Step 2: Explicit Simulations
Parameterize Material Response

Predicts cell
temperatures to $\pm 10^\circ\text{C}$

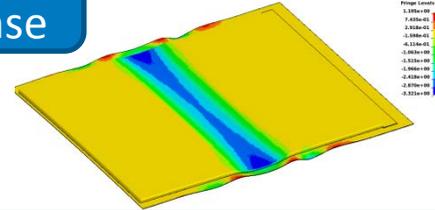


Current density
under short-circuit

Photo Credits: Jim
Marcecki, Ford

Step 1: Start with Component and
Cell-Level Test Results as Input

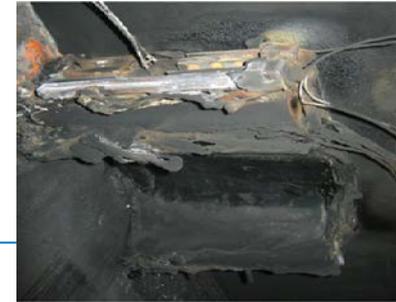
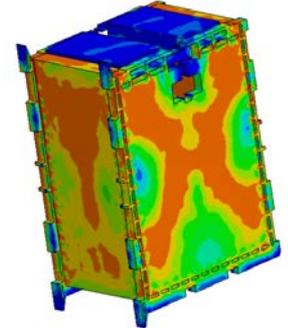
Step 3: Simulate Cell-Level
Response for Multiple Cases



Step 5: Validate against
Experimental Data

Goal: Identify localized failure modes
and onset loads to within 30 MPa

Step 4: Scale to Module-Level



Sample Input:

- Stress-strain curves for cell components (separator, current collector, etc.)
- Failure strengths for particles
- Mechanical data for cell packaging
- Temperature vs. C-rate for cell
- Abuse reaction data from calorimetry for specific chemistries

Sample Output:

- Current distribution among the different cells within the module
- Localized heat generation rates far away from damage zone
- Stress distribution across multiple parts of the battery module

Constitutive Model Development

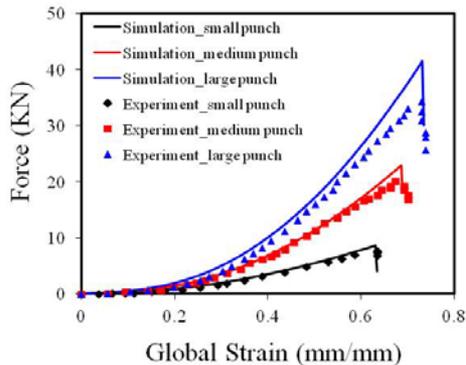
Step 1. Develop physics-based component models

$$\sigma_{ij,j} + \rho f_i = \rho u_{i,tt}$$

$$\sigma_{ij} = C_{ijkl} \gamma_{kl}$$

$$E = \begin{cases} E_{max} e^{\beta(\epsilon - \epsilon_p)} & \epsilon < \epsilon_p \\ E_{max} & \epsilon \geq \epsilon_p \end{cases}$$

Step 3. Validate against independent dataset

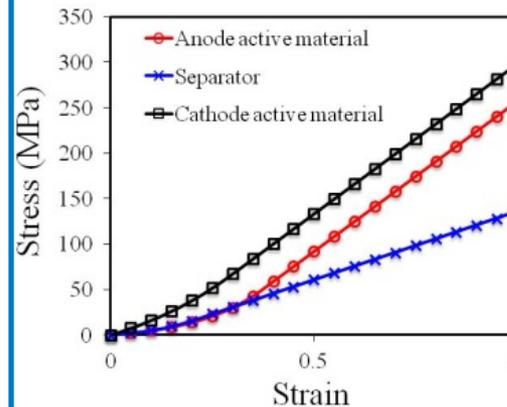


Cell-level data vs. Model

Step 2. Obtain model parameters

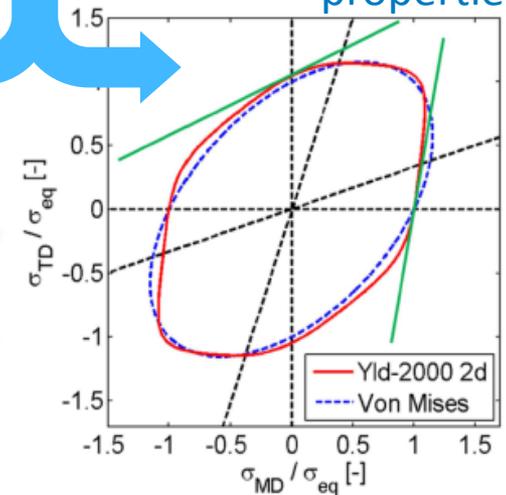
Approach a:

Calibrates parameters out of component-level stress-strain data



Approach b:

Phenomenological models for material properties



$$\bar{\sigma}(\sigma) = \bar{\sigma}(\sigma_{xx}, \sigma_{yy}, \sigma_{xy}) = \frac{1}{2^{1/a}} (|S'_I - S'_{II}|^a + |2 \cdot S''_I + S''_{II}|^a + |S''_I + 2 \cdot S''_{II}|^a)^{1/a}$$

Multiscale Simultaneously Coupled Modeling Framework

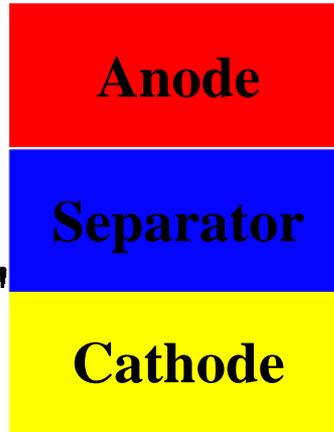
Macro-scale 3D homogenized mechanical-thermal model

Meso-scale quasi-3D mechanical-thermal model

Pseudo 2D electrochemical-thermal model

Element of the macro-scale model

$$\begin{aligned} & \xrightarrow{d\bar{\varepsilon}, dt, t, T} \\ & \xleftarrow{\bar{\sigma}, \sigma_i, \bar{\varepsilon}, \varepsilon_i, T'} \end{aligned}$$



$$\begin{aligned} & \xrightarrow{R_{short}, Ks_i, dt', t', T} \\ & \xleftarrow{T'} \end{aligned}$$

$$\sigma_{ij} = C_{ijkl} \varepsilon_{kl}$$

Approach for Coupling Methodology:

- Retain fidelity of damage models at the component level (e.g., separate failure criteria for separator, current collector, etc.)
- Solve for potential and temperature as additional degrees of freedom at the component scale
- Simulate multi-cell effects using a micro-mechanical homogenization scheme

$$\begin{Bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{12} \\ \sigma_{33} \\ \sigma_{23} \\ \sigma_{13} \end{Bmatrix} = \begin{bmatrix} C_{11} & C_{12} & C_{16} & C_{13} & C_{14} & C_{15} \\ C_{12} & C_{22} & C_{26} & C_{23} & C_{24} & C_{25} \\ C_{16} & C_{26} & C_{66} & C_{36} & C_{46} & C_{56} \\ C_{13} & C_{23} & C_{36} & C_{33} & C_{34} & C_{35} \\ C_{14} & C_{24} & C_{46} & C_{34} & C_{44} & C_{45} \\ C_{15} & C_{25} & C_{56} & C_{35} & C_{45} & C_{55} \end{bmatrix} \begin{Bmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \varepsilon_{12} \\ \varepsilon_{33} \\ \varepsilon_{23} \\ \varepsilon_{13} \end{Bmatrix}$$

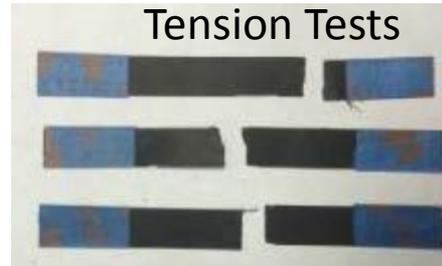
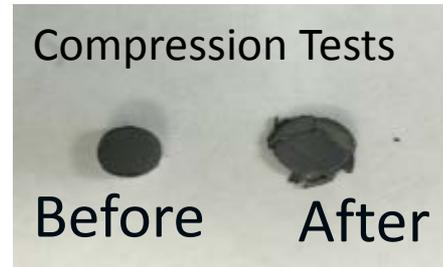
$$\begin{Bmatrix} \sigma_{=} \\ \sigma_{\perp} \end{Bmatrix} = \begin{bmatrix} C_{=} & C_{\times}^T \\ C_{\times} & C_{\perp} \end{bmatrix} \begin{Bmatrix} \varepsilon_{=} \\ \varepsilon_{\perp} \end{Bmatrix}$$

Zhang, Chao, et al. *IJES* 2016

Accomplishments: Component-Level Parameter Identification

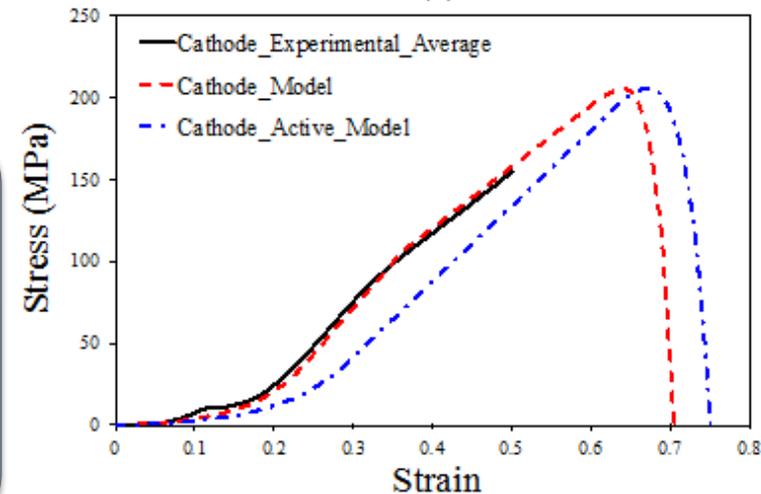
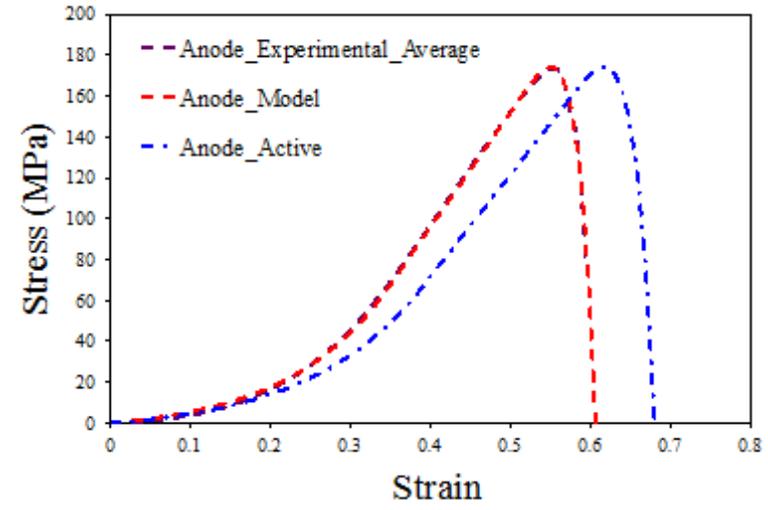


40-Ah PHEV cells (NMC-LMO/Gr) were cut open to characterize components



- Failure strains for each cell component under compression and tension were measured
- Properties for the active material were regressed based on composite structure response
- Constitutive model equations represent composite response reasonably well (errors in ultimate stress values < 10%)

Wu, et al. *JES* 2017 (Under Review)

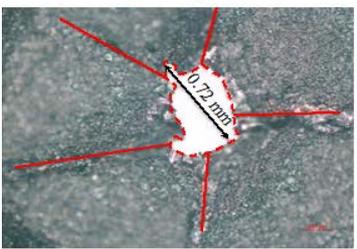


Constitutive response of electrode composites: Model vs. Data

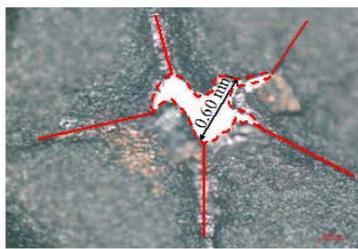
Mechanism of Failure Initiation following a Crush

Side facing the indenter

1st layer



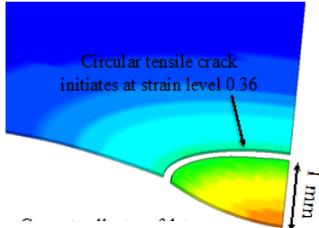
4th layer



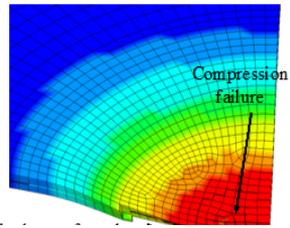
7th layer



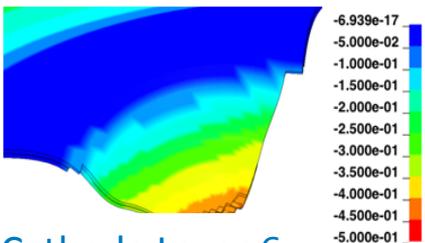
Sahraei et al. *Journal of Power Sources*, 2014



Copper foil Layer 1
Failure of Copper Foil



Anode Layer 4



Cathode Layer 6
Cathode-Anode Short

Cell-level crush tests used to have a “pass” or “fail”



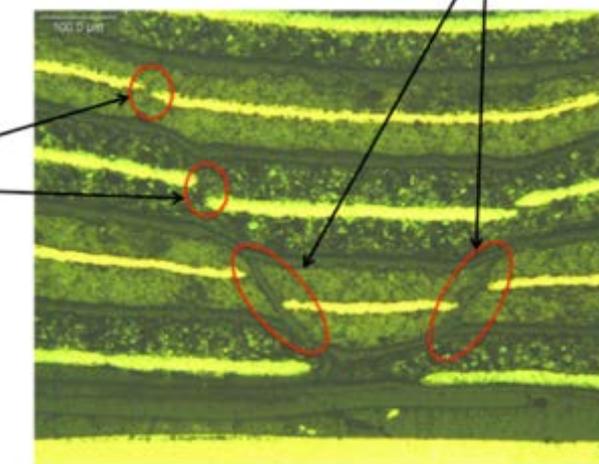
Shear failure of active material layers within a battery

C. Zhang, et al., *J. Power Sources*, Accepted (Mar. 2017)

Outcome:

- Comprehensive understanding of failure thresholds and propagation mechanism for each component within the cell
- Better explanation of test data results and recommendations for test-methods
- Light-weighting/right-sizing of cells without compromising safety

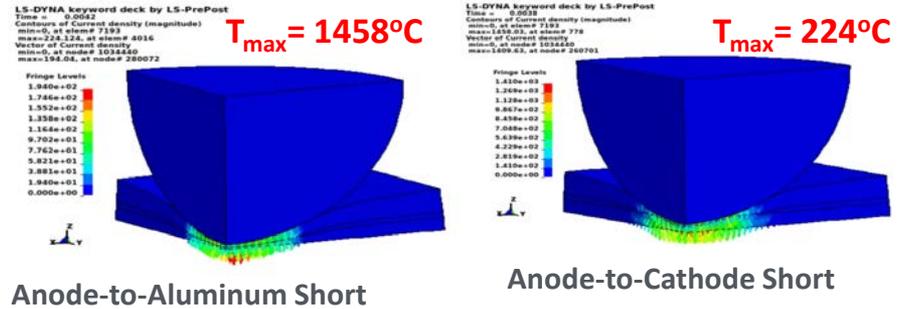
Copper foil fails before separator ruptures



Wang, Shin et al., *Journal of Power Sources* 306 (2016): 424-430.

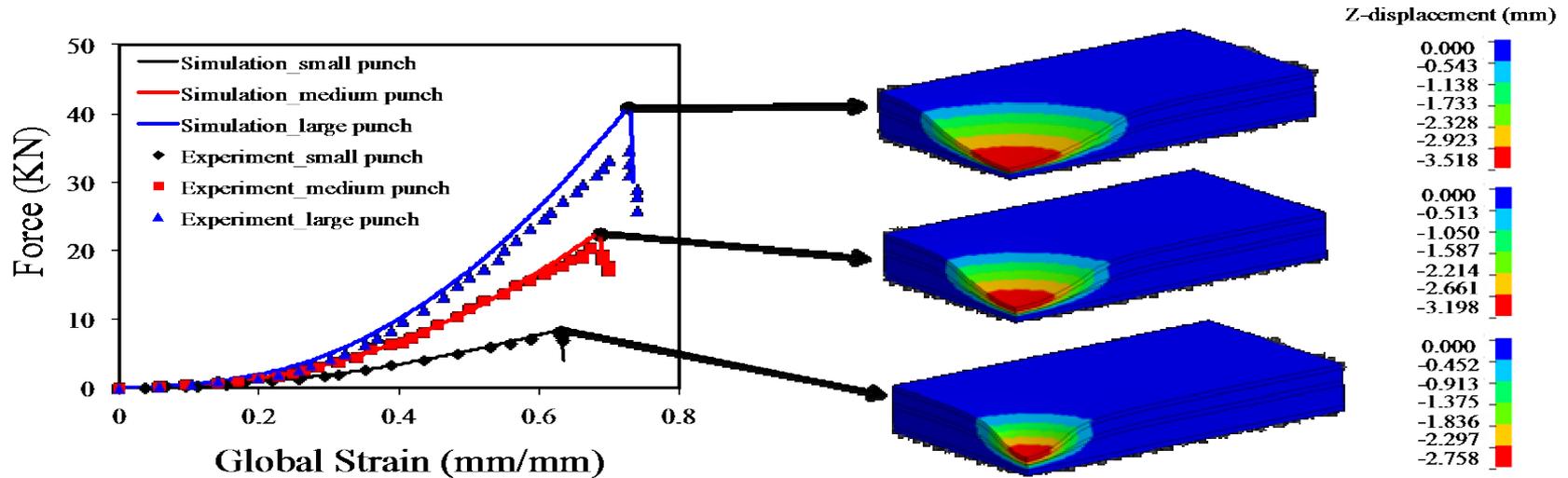
Cell-Level Results

Sahraei et al. Journal of Power Sources, 2014



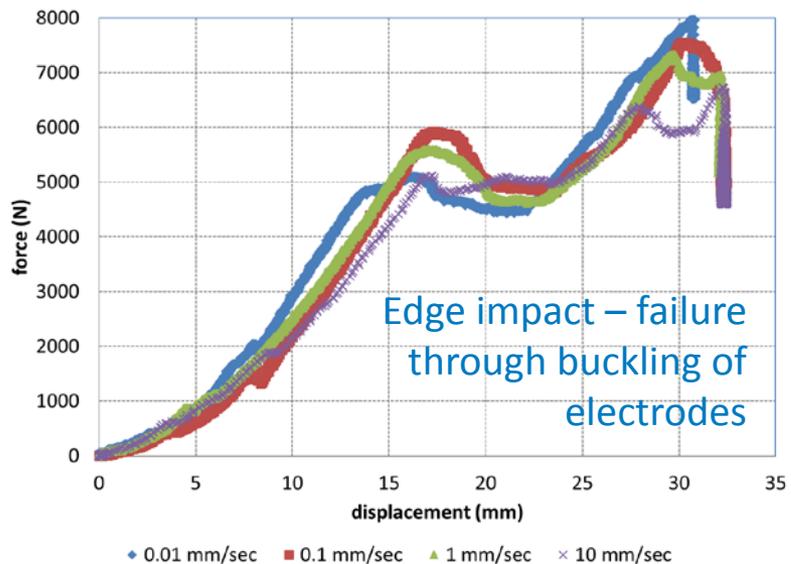
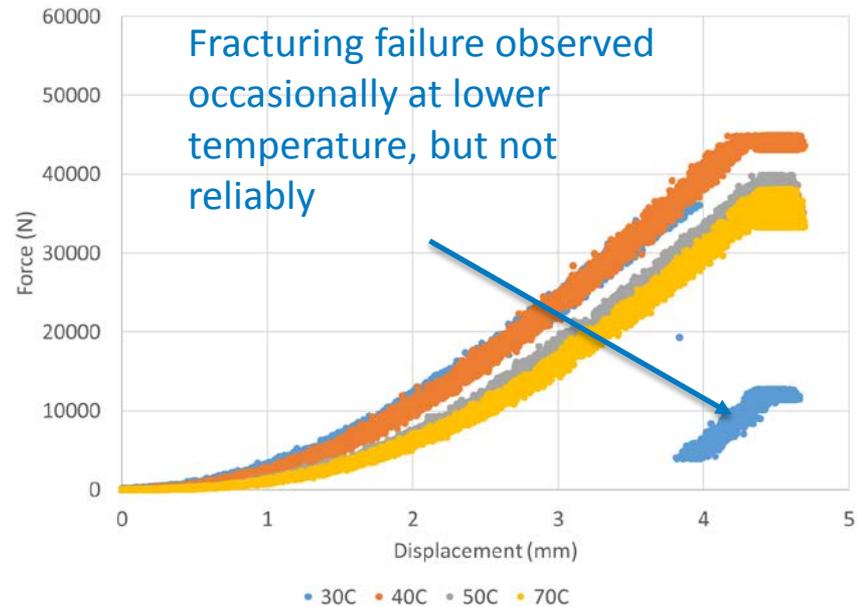
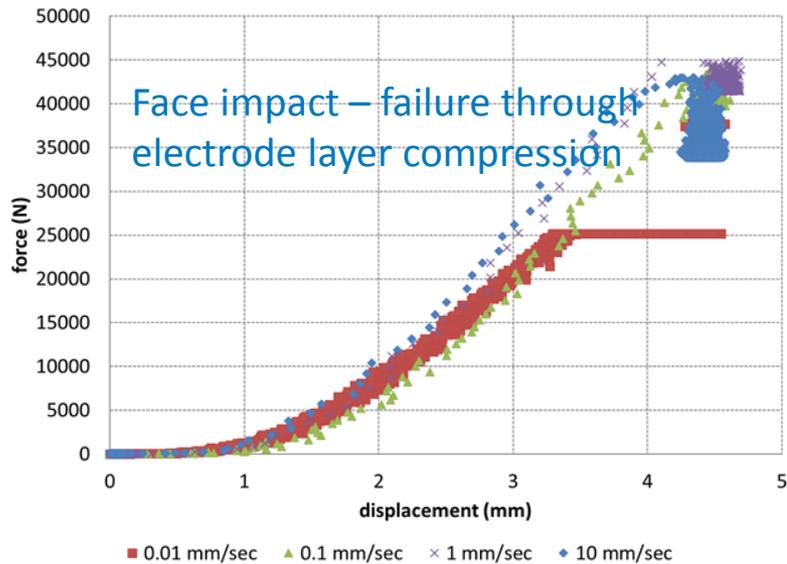
Cell Thermal Response under various types of short-circuit

S. Santhanagopalan, Presented at the International Battery Seminar & Exhibit, 2017.



Models adequately capture mechanical and thermal response under different test conditions.

Single-Cell Test Results from SNL



- Detailed characterization for different orientations, loading rate, temperature
- Some reduced resistance to compression was observed, particularly above 50°C
- Minimal changes with strain rates for range of conditions (0.1–10 mm/s) studied
- Numerical validation of models initiated

Multi Cell Test Results

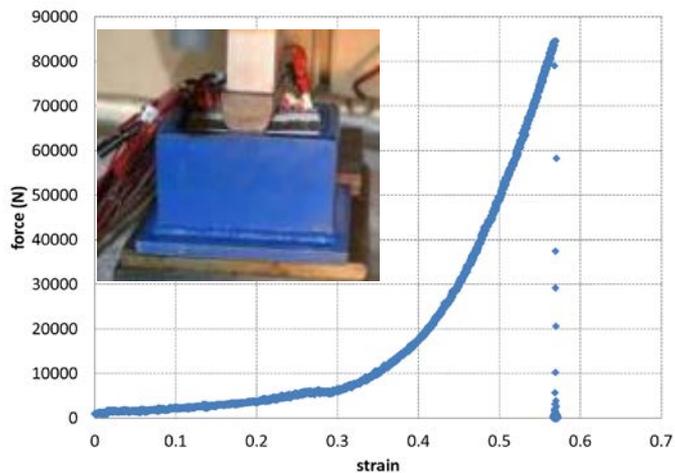
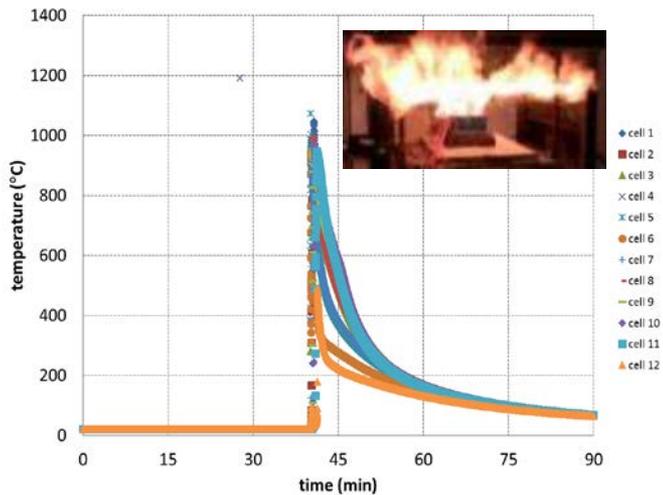
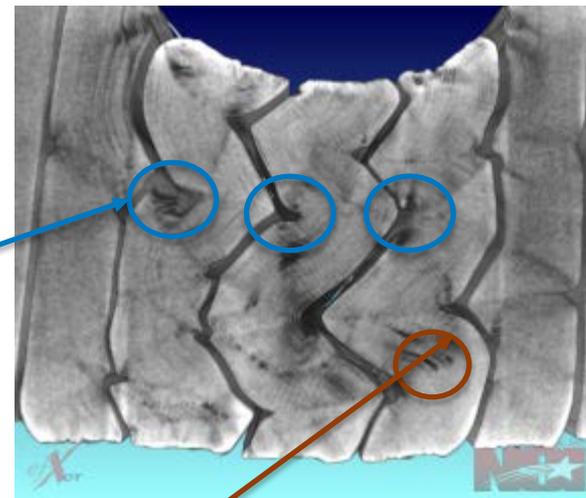


Photo Credits: Joshua Lamb, SNL

Bar crush of a 12-cell string



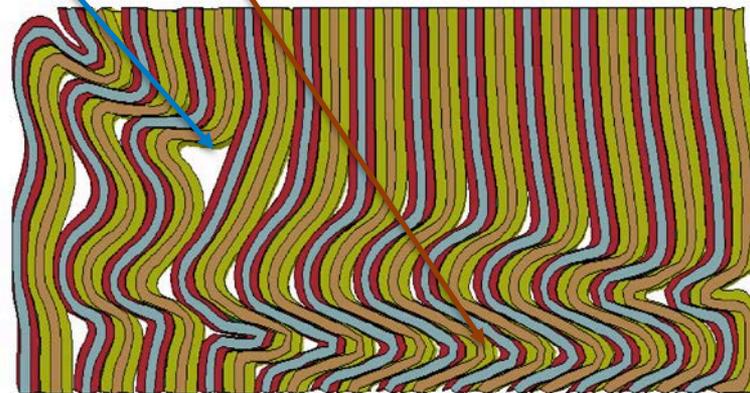
Data



Compression from buckling

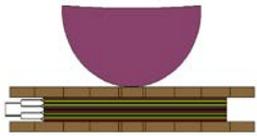
Separation of electrode layers

Model



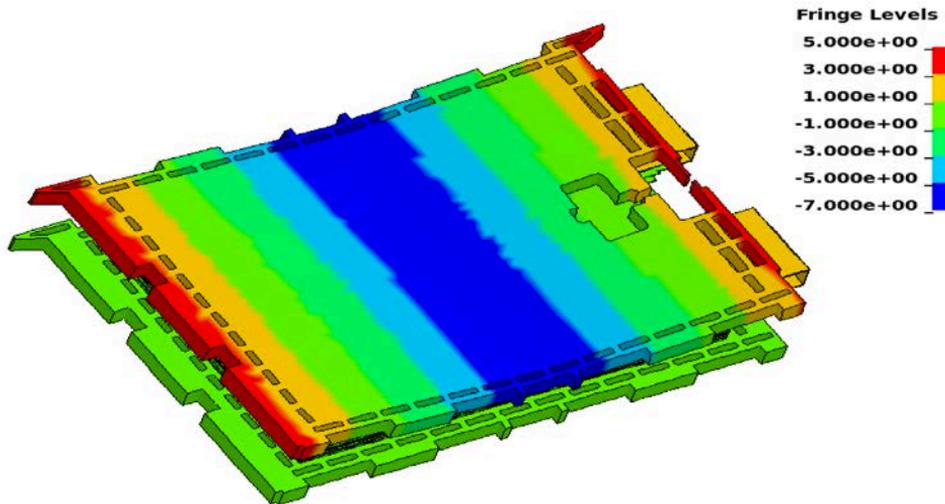
Models capture qualitative features; numerical comparison of failure strains underway.

Multi-Cell Simulations: Sample Results

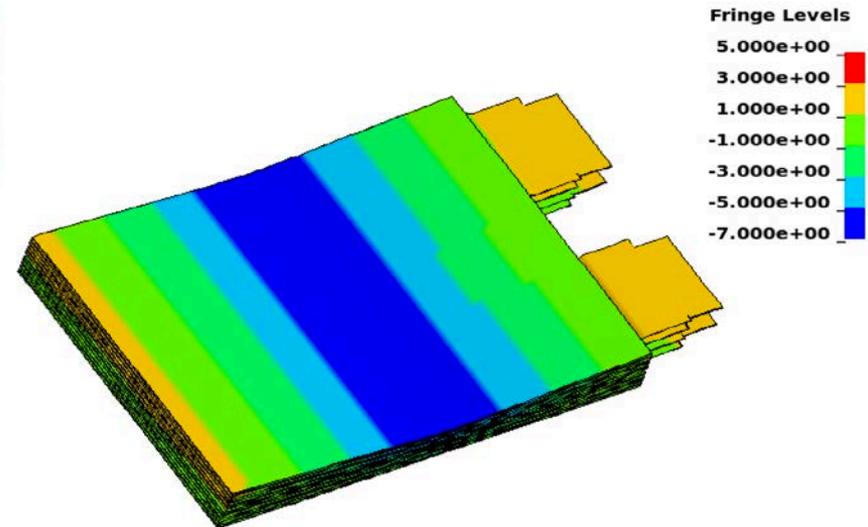


S. Santhanagopalan, Presented at the International Battery Seminar & Exhibit, 2017.

Deformation of packaging material



Deformation of the cells

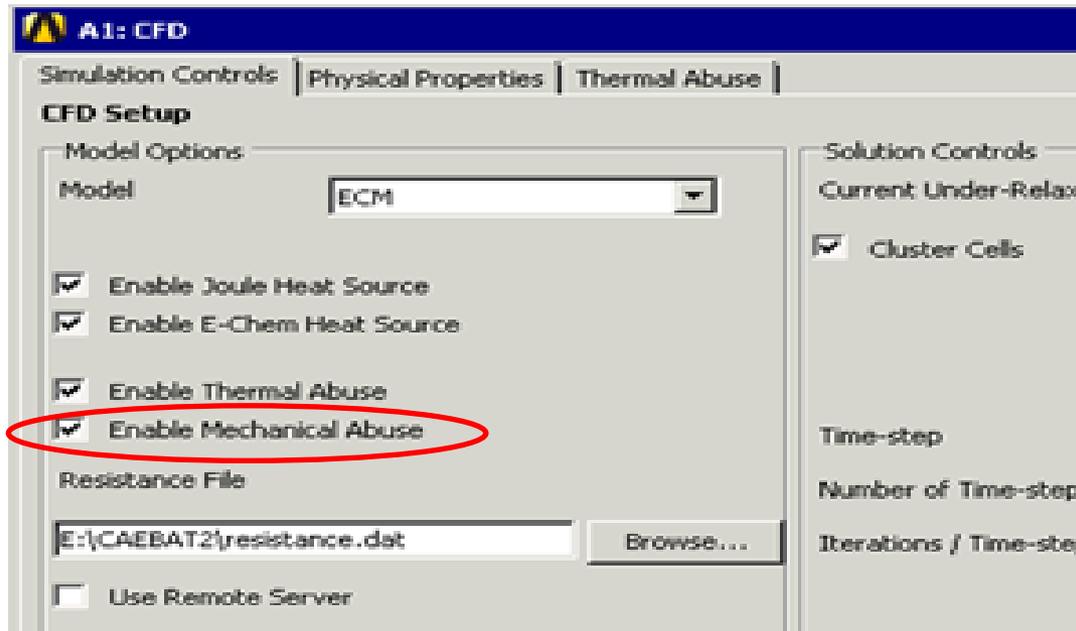


Models show that:

- The packaging can prevent deformation of the cells by as much as 50% under these crush test conditions.
- There is a significant scope to lightweight the pack, even after the safety threshold is met.

Future Work: Task 2

- Publish procedure for building constitutive relationships for the mechanical models for battery electrodes (2017)
- Complete validation against cell-level and multi-cell data from SNL (2018)
- Full cell numerical studies comparing sequential and simultaneous coupling approaches (2018)
- Develop plug-ins to link with other CAEBAT models in ANSYS/LS-DYNA user-defined models (2018)



Mechanical Plug-in for ANSYS

Any proposed future work is subject to change based on funding levels.

Response to Previous Year Reviewers' Comments

- Comment: The modeling approach involves building a front-end to commercial solvers (e.g., Fluent), which is very useful for the industry. However, will the tools be accessible to academic researchers without access to these large commercial codes?

Response: In response to this reviewer's comment, we put together a standalone executable version of the models, which is available to academia and industry alike, for use without requiring licenses to commercial CFD packages. This version is arguably limited in capabilities, but allows end-users to perform quality 3D battery simulations with reasonable computational resources.

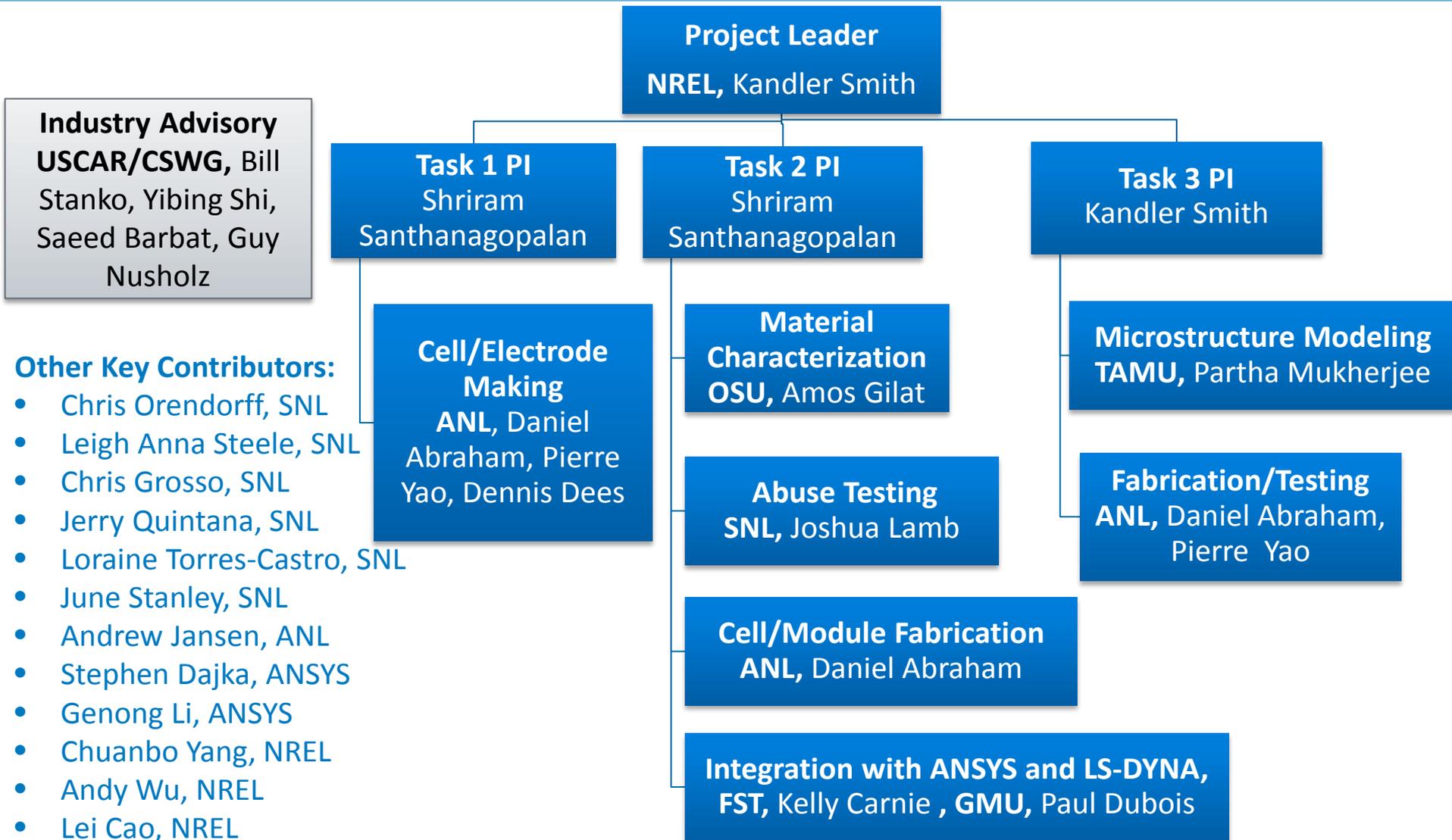
- Comment: Can this effort leverage the project lead by the GM/ANSYS team, co-funded by the Army and the DOE to increase the pack level combined mechanical/electrochemical/ thermal modeling efficiency?

Response: Yes. The team is working separately with ANSYS to implement the GH-MSMD method into Fluent to achieve similar speed up of models in commercial software.

Response to Previous Year Reviewers' Comments (Contd.)

- Comment: The reviewer cautioned that the community may start making incompatible predictions using different tools put forward by the ORNL, NREL and Ford teams, which could lead to confusion and slow progress.
Response: The three teams have similar, but complimentary set of goals. We hold quarterly review meetings and host two joint workgroups between the two lab-teams to eliminate minimize overlap, similar to those raised by this reviewer. We also hold monthly updates with the Crash Safety Work Group that includes participants from Ford, GM and FCA where we open the floor for feedback and review.
- Comment: Mechanical failure is a statistical, not deterministic process in which the presence and intensity of local inhomogeneities may control failure rate.
Response: We account for this artifact by building safety maps to assess room for error in the event of a mechanical failure. The deterministic models are used to build the safety maps by conducting a constraint function sweep across the parameter space to identify robustness of a given cell design.

Collaborators and Partners



Summary

- **Task 1.** The GH-MSMD model that provides 100 – 1000x computational speed-up than the MSMD models for battery electrochemical/thermal simulations, is now available for licensing and can run pack simulations on a laptop
 - Publication: G-H. Kim et al., *J. Electrochem. Soc.*, A1076-1088 (2017).
 - Speed enables direct full model use in parameter identification: an automated pipeline to calibrate model from battery-cycler data is under development.
- **Task 2.** Simultaneously coupled mechanical-electrochemical-thermal model for mechanical abuse simulation
 - Multi-scale model can include multiple failure criteria for each component in a module- or pack-level simulation.
 - Initial set of comparisons against test data at the component level shows good promise for homogenization approach.
 - Comprehensive model validation is ongoing in partnership with SNL.
 - Effort to streamline interfacing with off-the-shelf software tools (ANSYS/LS-DYNA) is underway.

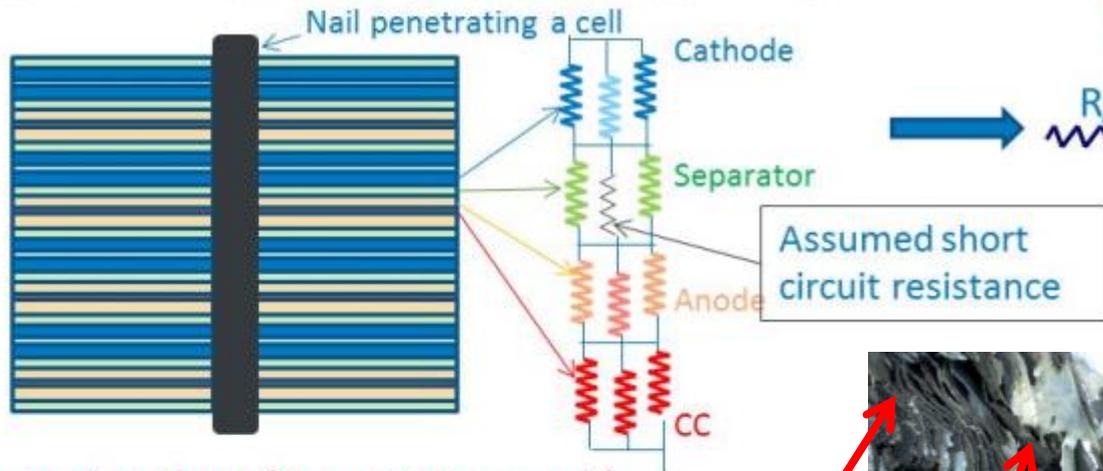
Acknowledgements

- We appreciate support and funding provided by Vehicle Technologies Office at the U.S. Department of Energy
 - Brian Cunningham
 - David Howell
 - Samuel Gillard

Technical Back-Up Slides

Estimating Short-Circuit Resistance

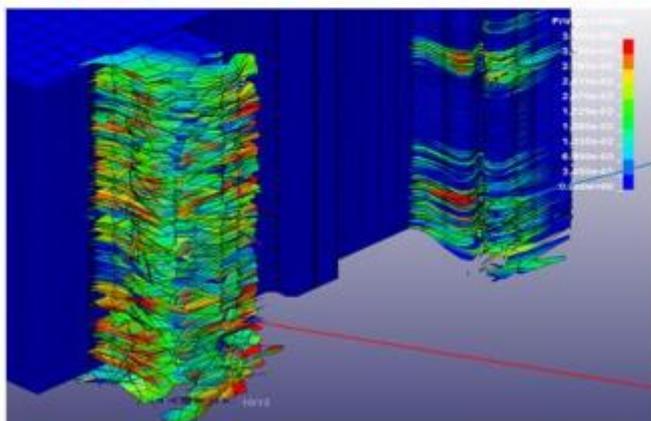
Regular Short (Previously Reported Approach):



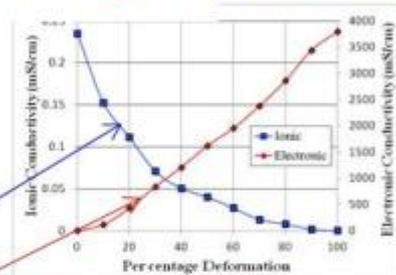
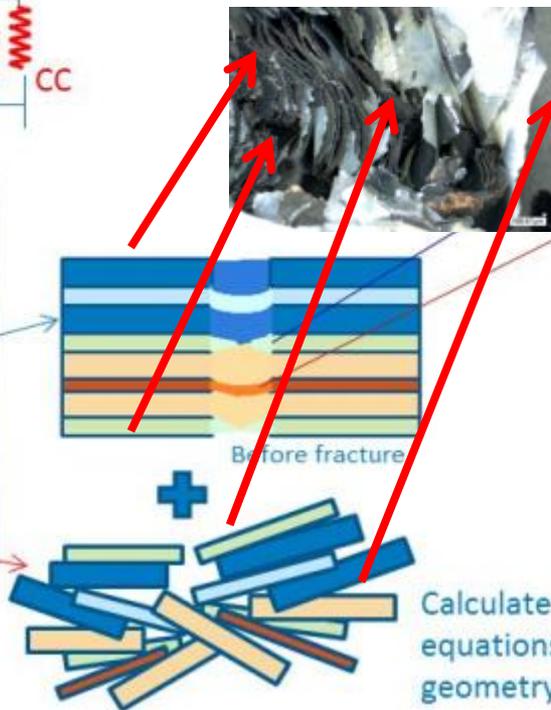
Gi-Heon Kim, 6th Lithium Mobile Power Conference, Boston MA, 2010.

Kalupson et al., ECS Spring Meeting, Orlando FL, April 2014.

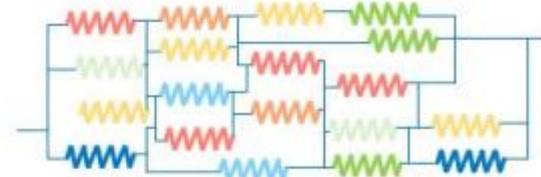
Irregular Short (Current Approach):



Deformed geometry obtained using CAD geometry of cells subjected to crush in LS-DYNA simulations

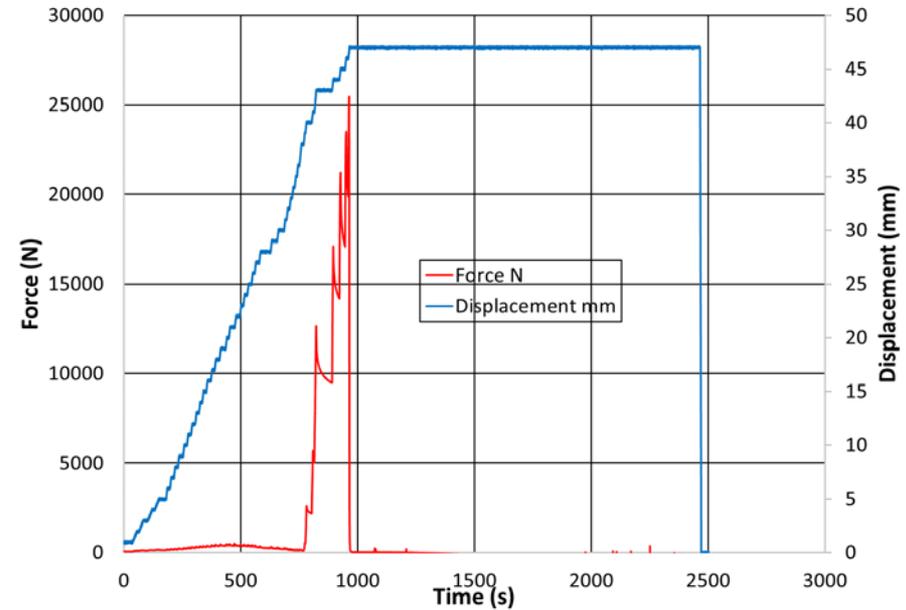
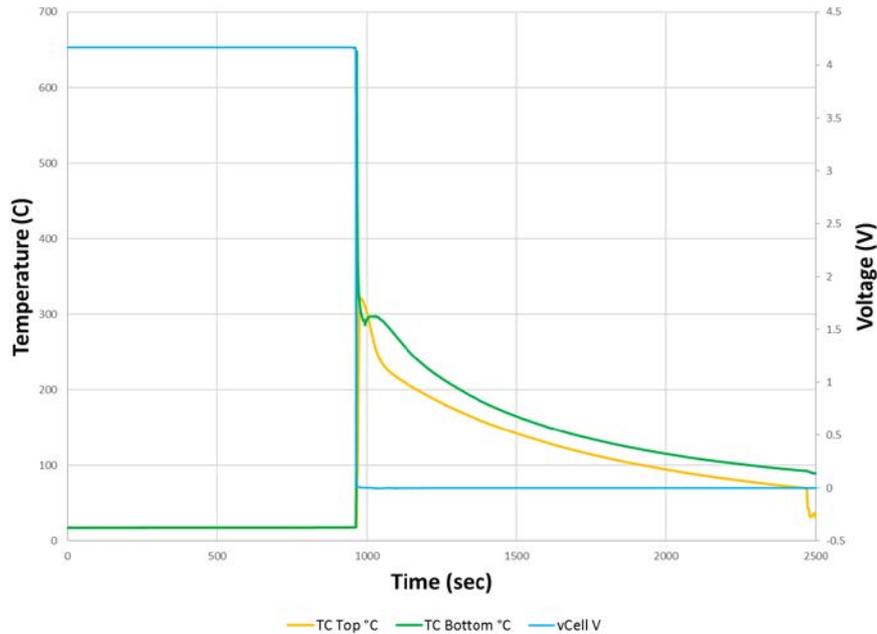


Use ionic and electronic resistivity of the deformed layers change as a function of thickness



Calculate short resistance by solving Maxwell's equations in Ansys or DYNA using deformed geometry and properties of individual layers

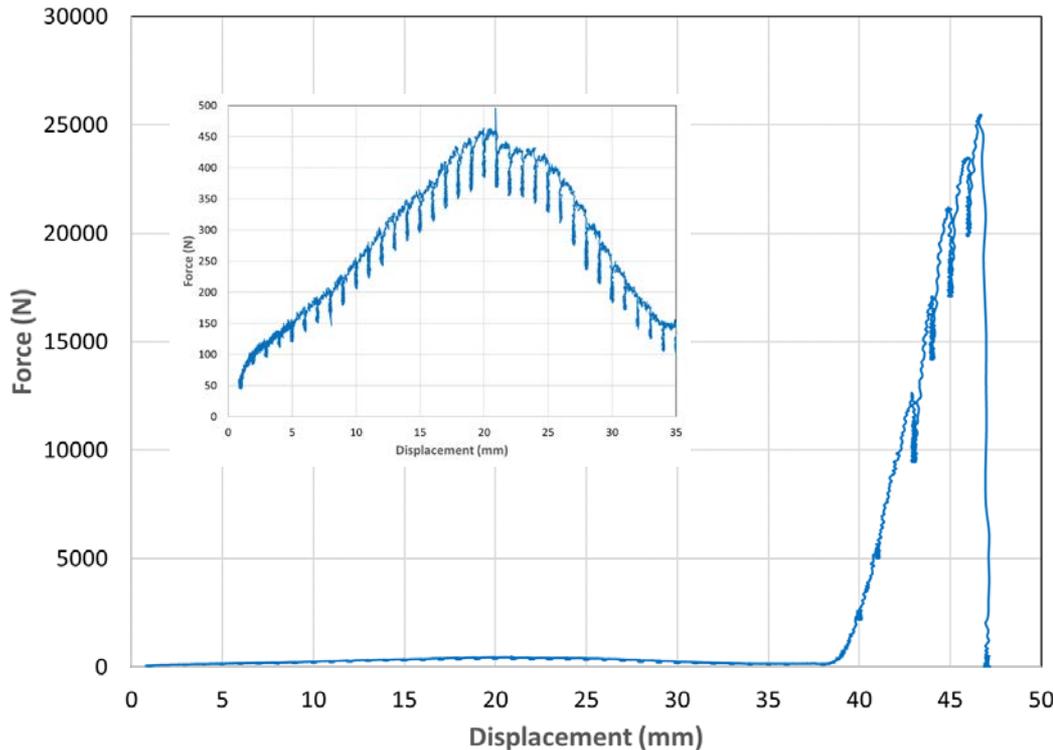
Three-Point Bend Test – Fully Charged Cell



Full charge test

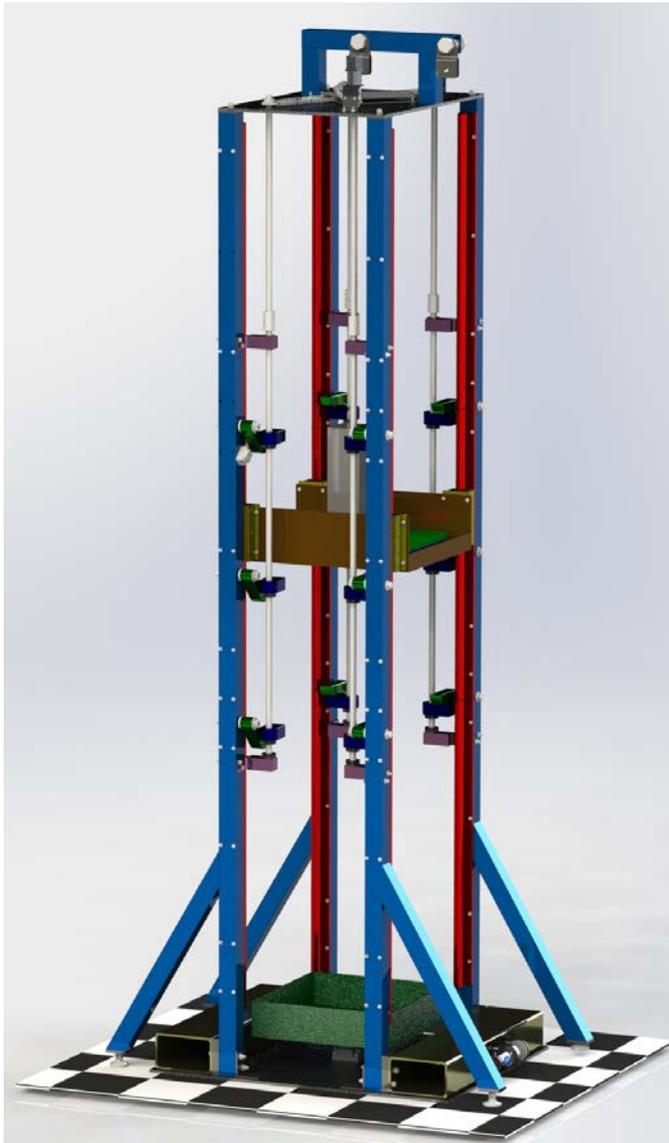
- Initial test conducted in 1-mm steps to determine point of failure
- No failure observed during bend of cell
- Failure achieved through cell compression at end of test

Three-Point Bend Test – Fully Charged Cell



- Bend portion of test shows a yield of ~ 450 N.
- Cell failure required a compressive force of ~ 25 kN.
- “Pre-load” portion of bend observed where initial compression is applied to cell before bending occurs.
- After yield of cells to bend the cell is put into compression.

Drop Tower – Impact Tester



Specifications:

- Overall height: 14 feet (4.3 m)
- Drop height: up to 10 feet (3.1 m)
- Drop weight: 50 to 500+ pounds (22.7 – 226.8 kg)
- Max impact velocity ~ 25.4 ft/s (7.74 m/s)
- Impact force (assuming a 6" stopping distance): 10,000 lbs-f (44,482 N)
- Remote operation
- Data collection:
 - Displacement
 - Impactor velocity
 - Force at impact
 - Temperature
 - Voltage

Figure Credit: Joshua Lamb, SNL

Drop Tower – Impact Tester

Current Status:

- ✓ CAD model – complete
- ✓ Drawing package – complete
- ✓ Hardware bill of materials (BOM) – complete
- ✓ Controls box design – complete
- ✓ Controls BOM – complete
- ✓ BATLab personnel to order all controls hardware – near complete
- Build request, including drawing package – submitted to contractor
- BATLab personnel to order all hardware for build – waiting on contractor readiness
- BATLab personnel to complete final assembly of drop tower – waiting on completion of contractor build

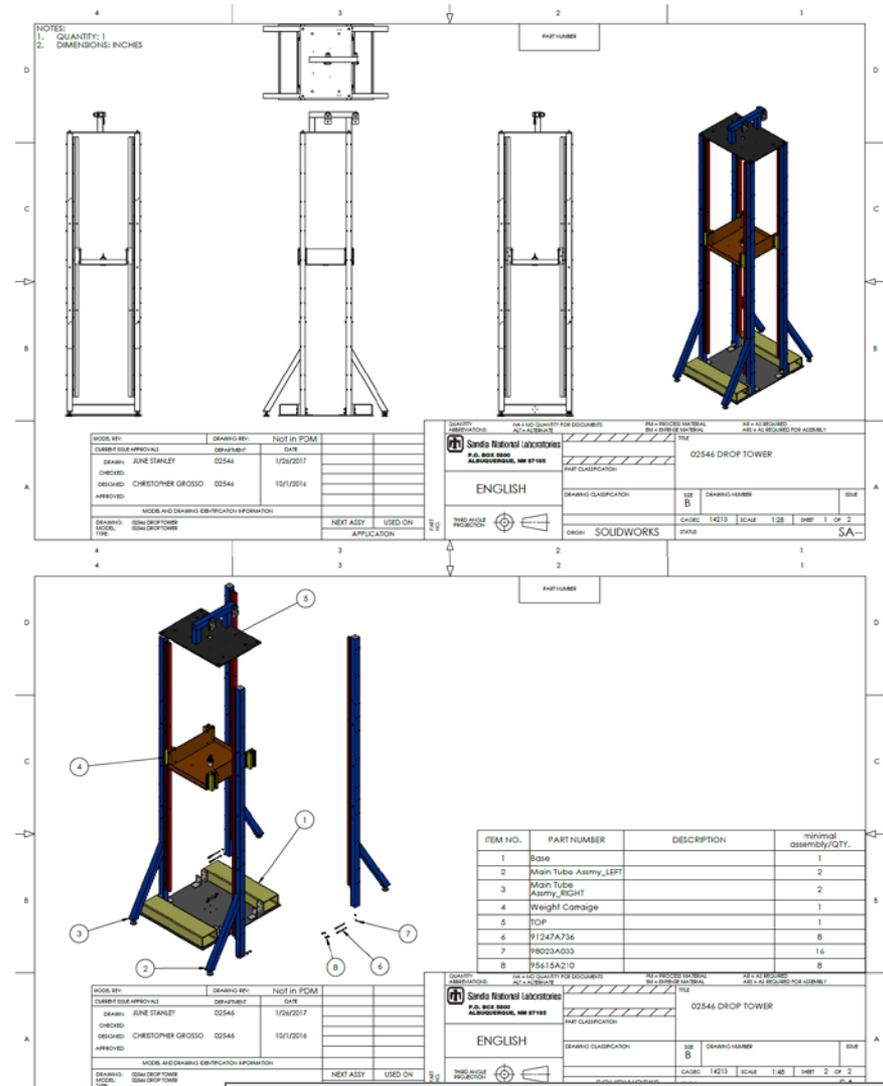


Figure Credit: Joshua Lamb, SNL