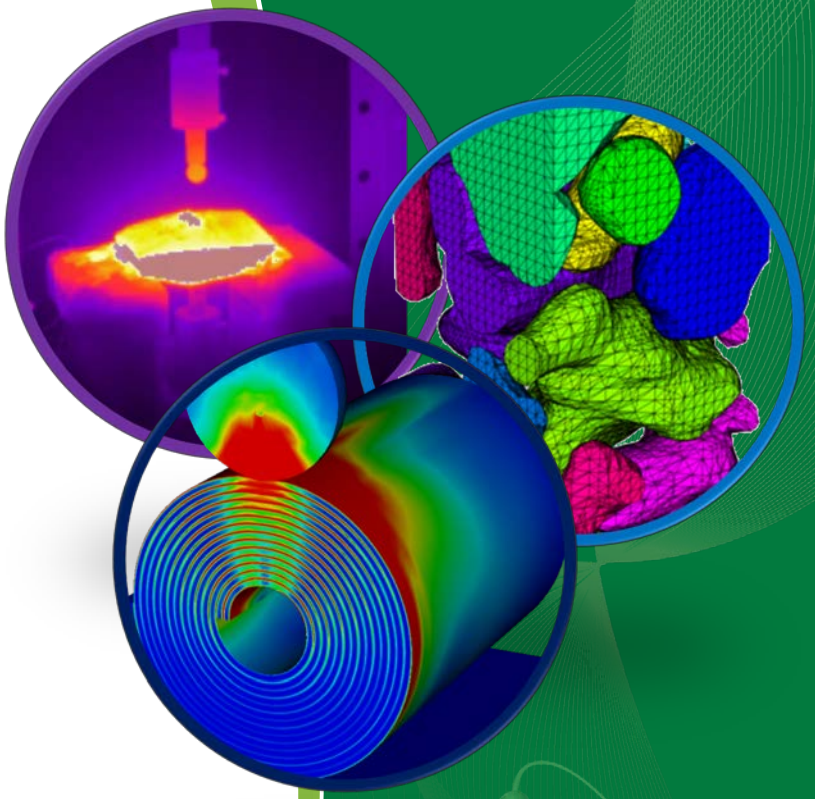
Experiments and Models for the Mechanical Behavior of Battery Materials

ORNL is managed by UT-Battelle
for the US Department of Energy



PI: John Turner

Team: S. Allu, W. Elwasif, Z. Feng, M. Ferraro, S. Gorti, K. Higa, S. Kalnaus, A. Kumar, J. Li, S.A. Roberts, Y. Sheng, S. Simunovic, V. Srinivasan, B.Trembacki, B. Turcksin, H. Wang, Y. Wang

Oak Ridge National Laboratory

2017 U.S. DOE Vehicle Technologies Office Annual Merit Review

June 6, 2017 **Project ID:** ES301

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

CABS Consortium for Advanced Battery Simulation

Overview

Timeline

- Start
  - October 2015
- Finish
  - September 2018
- Percent complete: 55%

Barriers Addressed

- Performance
- Abuse Tolerance, Reliability
- Life

Budget

- FY16
  - Total CABS: $2,265K
  - This effort: $482K
- FY17
  - Total CABS: $2,225K
  - This effort: $525K

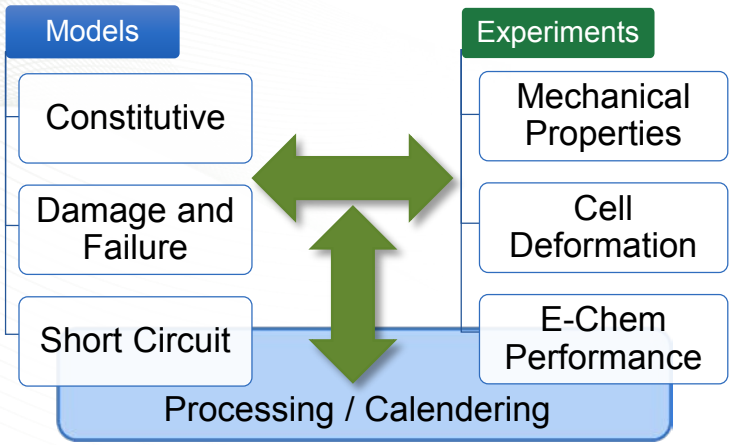
Partners

- LBNL
- SNL
- ANL
- as well as the NREL-led CAEBAT project team

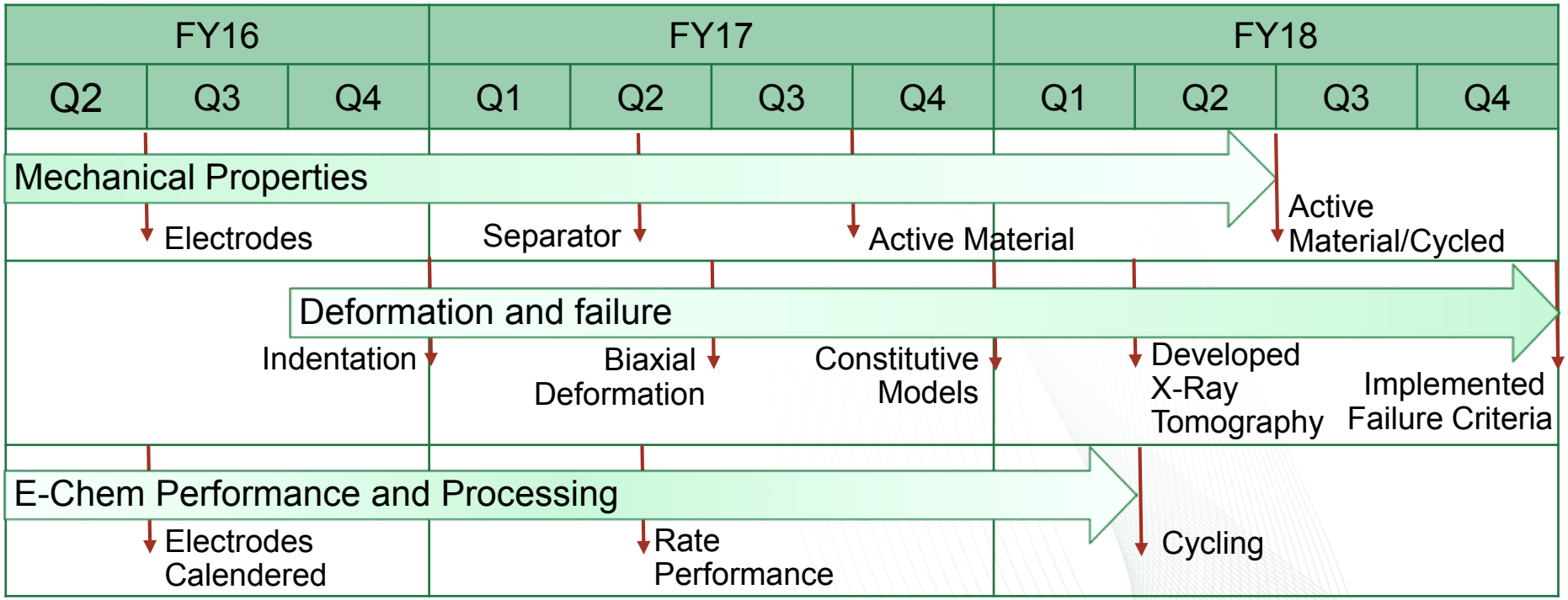
Relevance

- Major barriers for increasing battery energy density and power, increasing safety and reducing cost include
  1. ***insufficient understanding of the underlying physical phenomena that limit battery performance and safety, particularly the role of microstructure***, and
  2. lack of validated predictive simulation tools.
- CABS is addressing (1) ***by developing new experiments and developing new validated models that allow researchers to explore battery response under both normal and abusive conditions***, and is addressing (2) by deploying increasingly capable and computationally efficient releases of the Open Architecture Software (OAS) and components of the Virtual Integrated Battery Environment (VIBE), developed as part of CAEBAT 1.

Approach



- Develop experimental protocols
- Formulate and validate mechanics models
- Coupling to other models (E-Chem, Thermal)
- Formulate and validate failure criteria



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CABS Milestones

IDs indicate whether milestones are primarily experimental (E), computational (C), or integrated (I).							
ID	FY16	Lead	Q1	Q2	Q3	Q4	Status
C.1	Baseline performance profile of VIBE/OAS/AMPERES	ORNL	P				Complete
I.1	*Report on experimental techniques*	*ORNL*		*P*			*Complete*
E.1	Produce segmented tomographic reconstructions of electrodes	LBNL			P		Complete
E.2	*Demonstration of single side indentation test with incremental deformation*	*ORNL*				*P*	*Complete*
C1.1	Collect constitutive models for NMC materials	SNL				P	Complete
I.2	Deployment of VIBE/OAS with enhanced extensibility and hybrid models	ORNL				S	Complete
ID	FY17	Lead	Q1	Q2	Q3	Q4	Status
I.3	Demonstration of ability to construct 3D meshes using reconstructions from micro-tomography	SNL	P				Complete
E.3	Potential-dependent solid diffusivities for Li-ion and EIS	LBNL		P			Complete
I.4	Demonstrated ability of VIBE/OAS to simulate onset of short-circuit	ORNL		D			Complete
E.4	*Data from mechanical deformation tests*	*ORNL*			*P*		*Ongoing*
C.2	*Validated constitutive models and failure criteria*	*ORNL*				*P*	*Ongoing*
I.5	Deployment of VIBE/OAS with integrated multiscale capability	ORNL				S	Ongoing

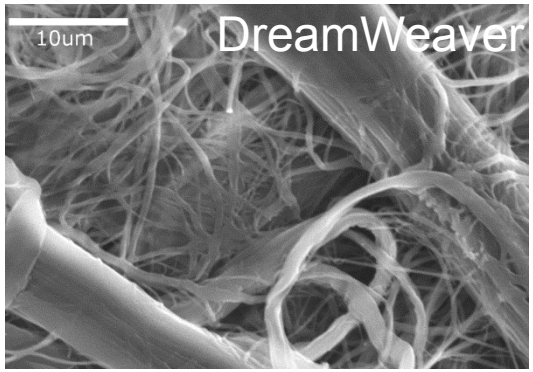
Separators: comparison study

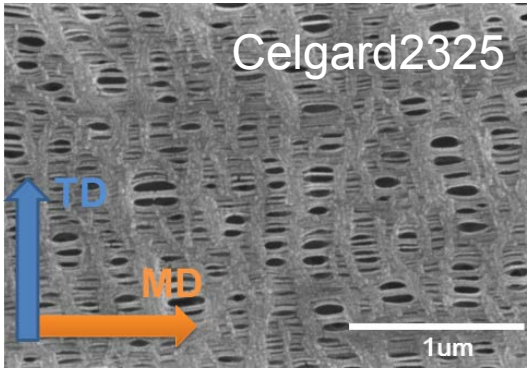
Technical Accomplishments

Requirements for mechanical properties of Li-ion battery separator:

- Puncture strength: 300 g/mil [1,2]
- Mix penetration strength: 100 kgf/mil [1]
- Tensile strength in MD: 2% offset 1000 psi [1,2]

Is this enough? Battery separator – critical safety component!





Detailed study of mechanical behavior of three commercial separators

Separator	Composition	Thickness µm	Porosity %
Celgard 2325	PP/PE/PP	25	39
Celgard PP2075	PP	20	48
Dream Weaver Gold	Non-woven para-aramid fibers	40	68

Strain rates:

1e-04 (~ 1C charge)

1e-01 (~ 4 mm/s, fast indentation)

[1] P. Arora, Z. Zhang, Chem Rev 2004, 104 4419-4462.
[2] NASA/TM – 2010 – 216099

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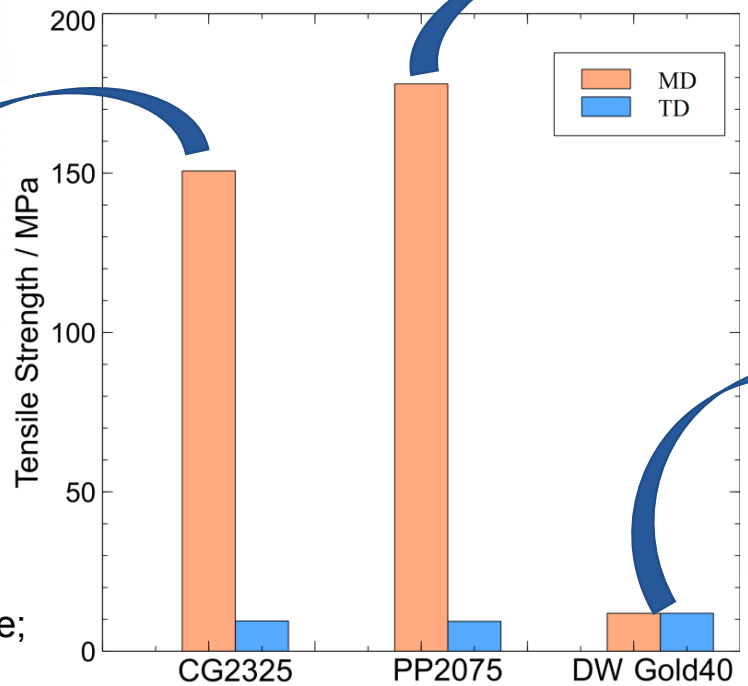
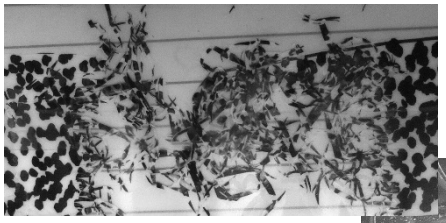
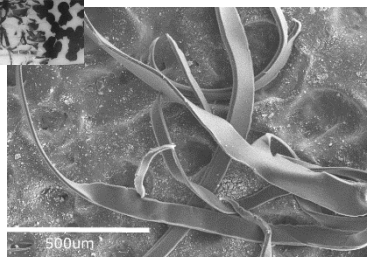
Separators: comparison study

Technical Accomplishments

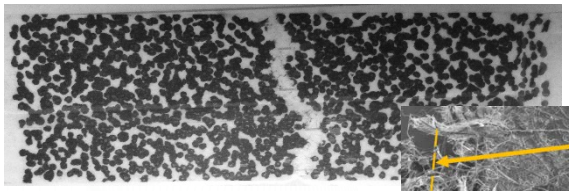


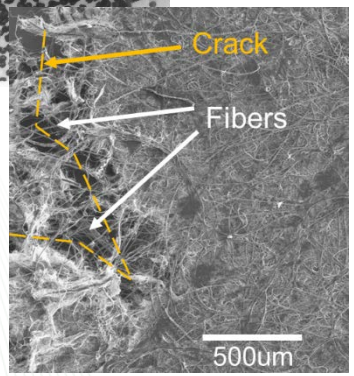




Celgard 2075:
- Separation into pieces;
- Anisotropy.

Celgard 2325:
- Straight-line failure;
- Anisotropy.





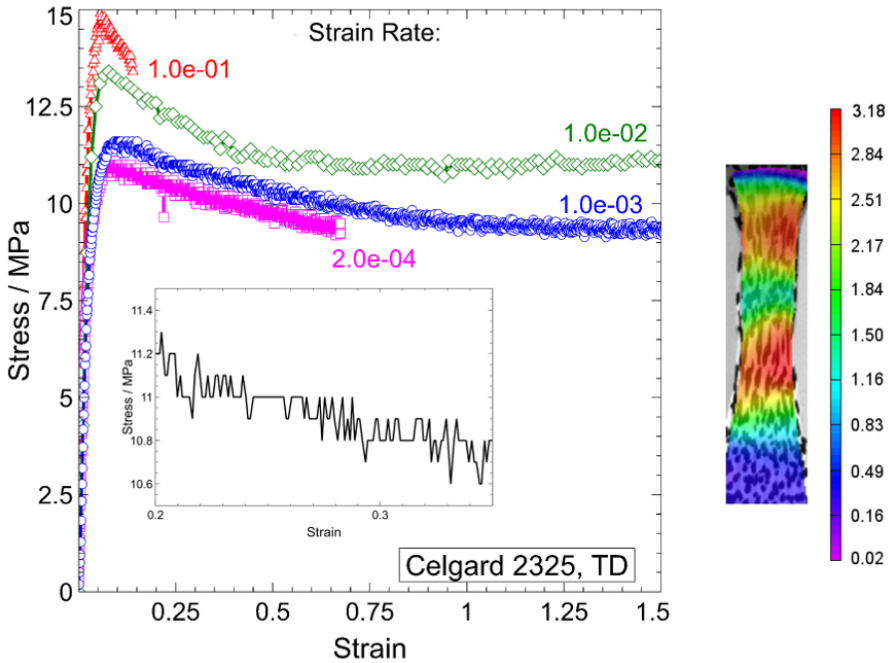
DreamWeaver Gold40:
- Gradual failure by fiber pull-out;
- Isotropic.

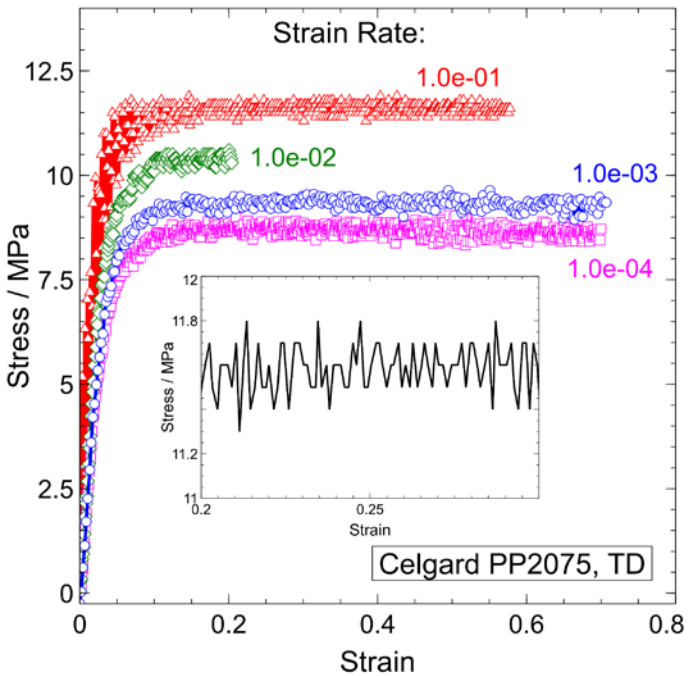
All satisfy standard requirements, but differ in:
- Yield and tensile strength
- Anisotropic behavior
- Failure mode

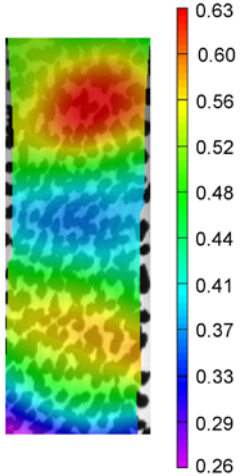
Note: Speckled pattern was applied to the samples for Digital Image Correlation (DIC) technique to observe the strain distribution.

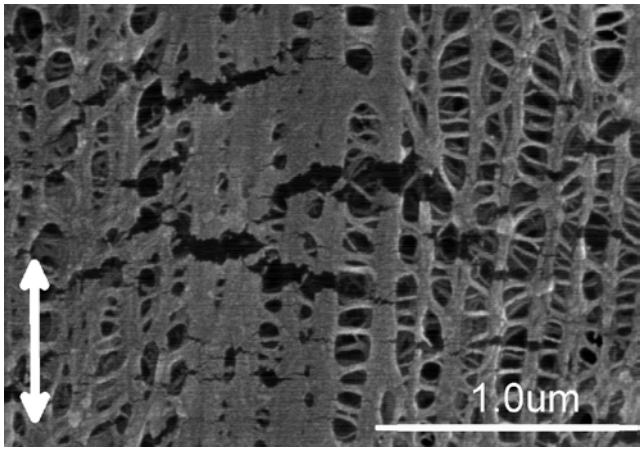
Strain distribution and failure in TD

Technical Accomplishments









- Significant accumulation of strain in bands
- Unstable neck propagation
- Clear yield point and cold drawing region
- Failure of lamellae observed
- Strain rate sensitivity

S. Kalnaus et al., *J. Power Sources* 348 (2017) 255-263.

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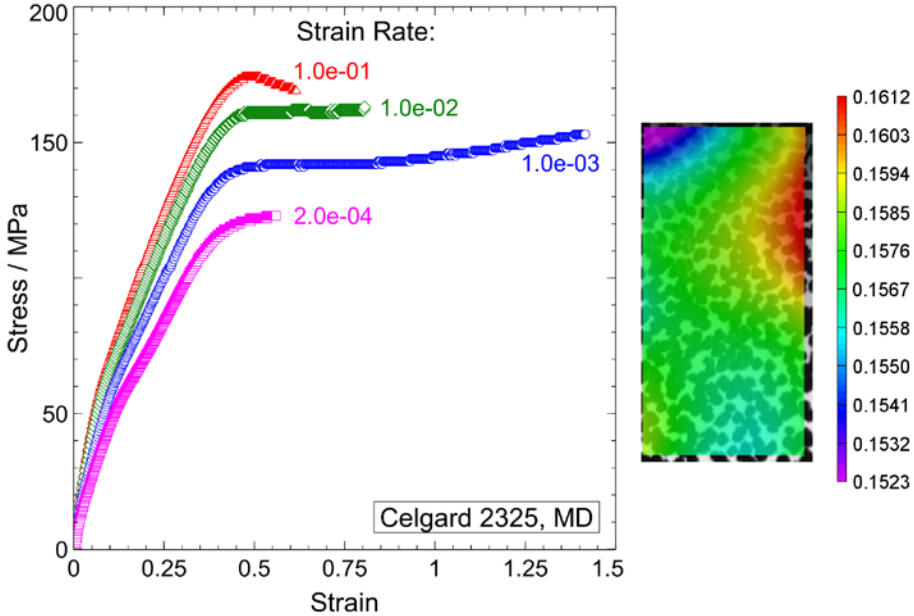
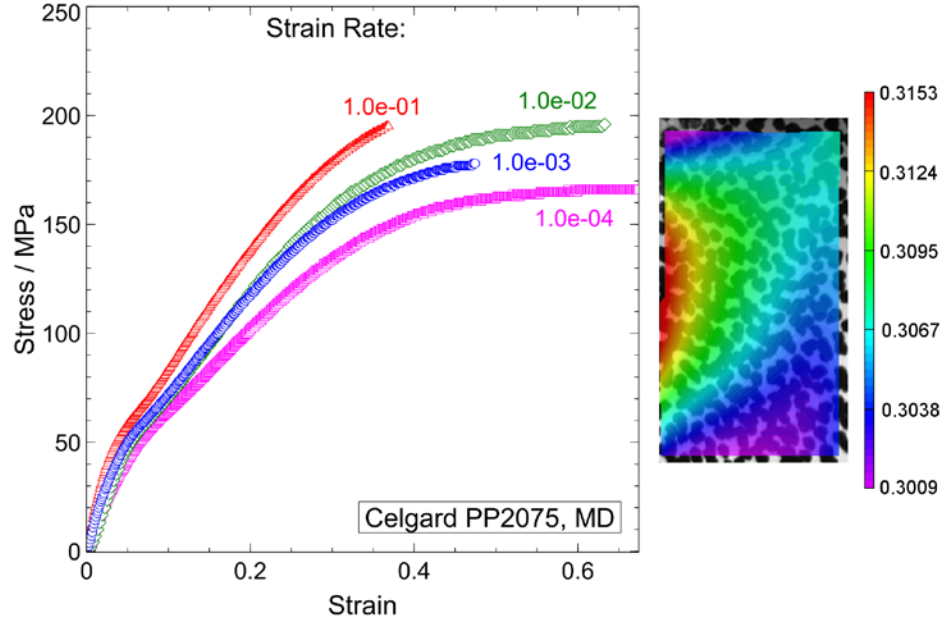
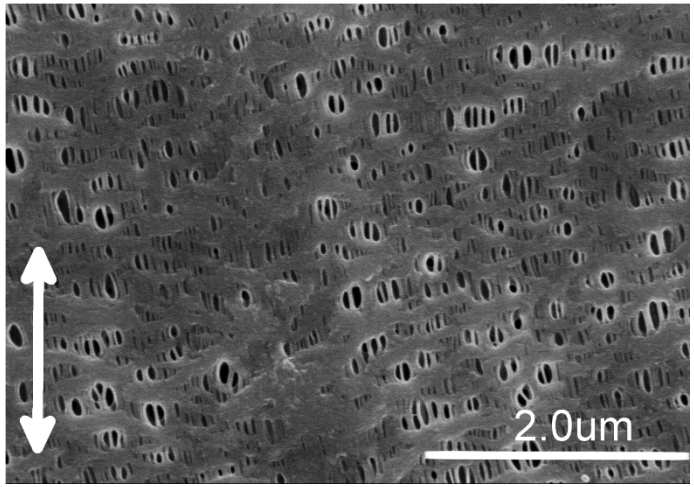
Strain distribution and failure in MD

Technical Accomplishments







- 10x stresses compared to TD
- Homogeneous strain distribution
- Some cold drawing and strain hardening
- Extension of fibrils
- Strain rate sensitivity

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Rate and temperature dependent behavior (Celgard 2325)

Technical Accomplishments

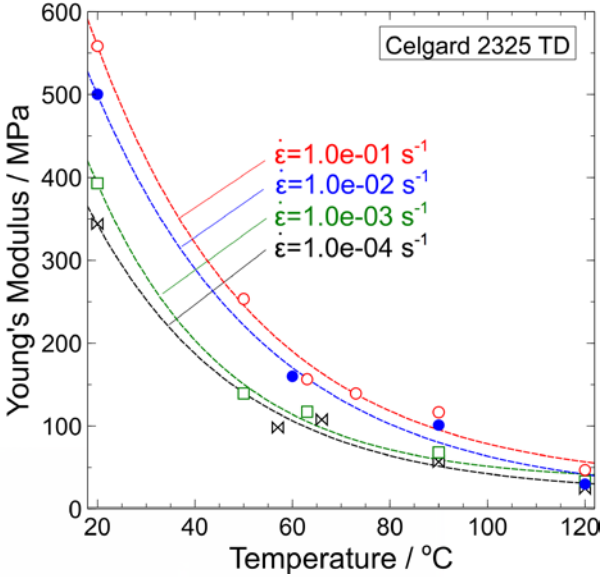
Motivation

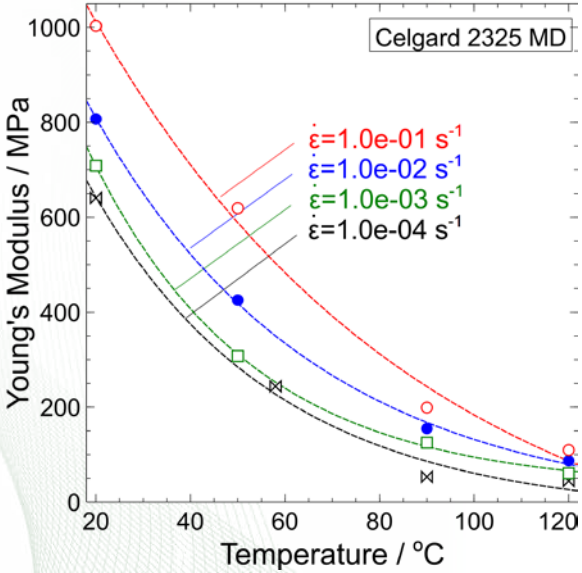
- Knowledge of thermomechanical behavior is necessary for two-way coupling with thermal transport.
- Complete map of strain rate and temperature dependent behavior is now available within 20 °C – 120 °C.
- The upper temperature was picked close to the PE melting point (130 °C) simulating conditions close to those triggering safety shutdown of the separator.

Observations

- Order of magnitude decrease in stiffness and strength
- At high temperature material properties are nearly rate independent

Effects of temperature cannot be ignored: about factor of 2 decrease in modulus when temperature is increased to 50 °C.





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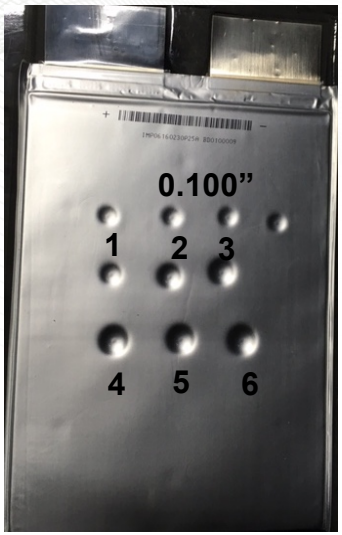
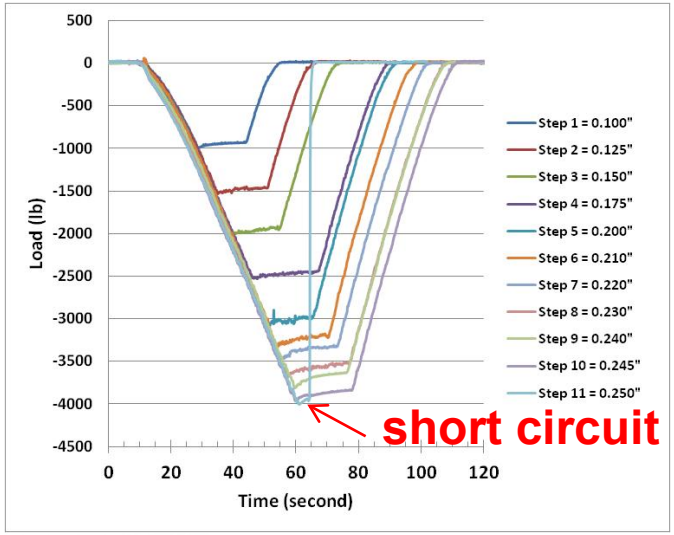
Determination of critical conditions for short circuit

Technical Accomplishments





Cell#1:

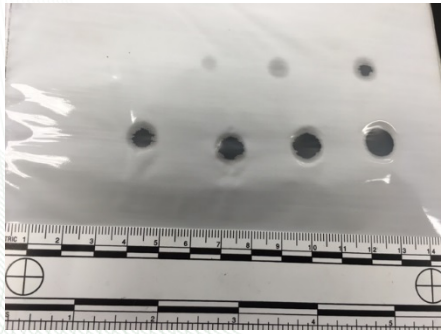
Test Steps	Displacement(in)	mm	Short
1	0.1	2.54	No
2	0.125	3.175	No
3	0.15	3.81	No
4	0.175	4.445	No
5	0.2	5.08	No
6	**0.21**	**5.334**	**No**

Cell#2:

Test Steps	Displacement(in)	mm	Short
6	**0.21**	**5.334**	**No**
7	0.22	5.588	No
8	0.23	5.842	No
9	0.24	6.096	No
10	**0.245**	**6.223**	**No**

Cell#3:

Test Steps	Displacement(in)	mm	Short
10	**0.245**	**6.223**	**No**
11	0.250	6.350	**Yes**



- After 10 steps the cell was disassembled in glovebox
- Plastic deformation in separator is visible as transparent areas
- Areas were measured and compared to those predicted by simulations

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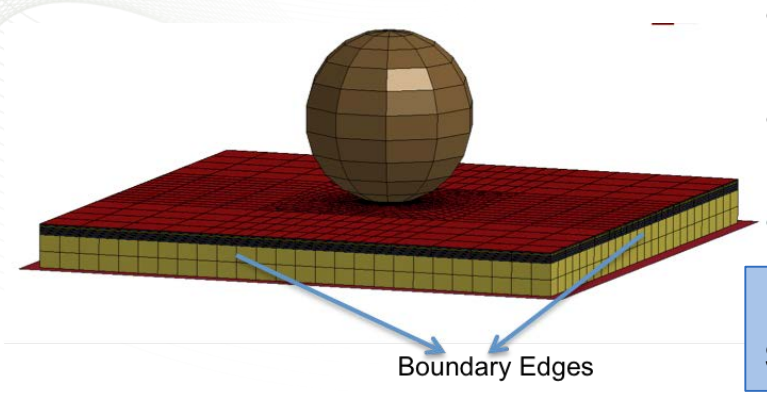
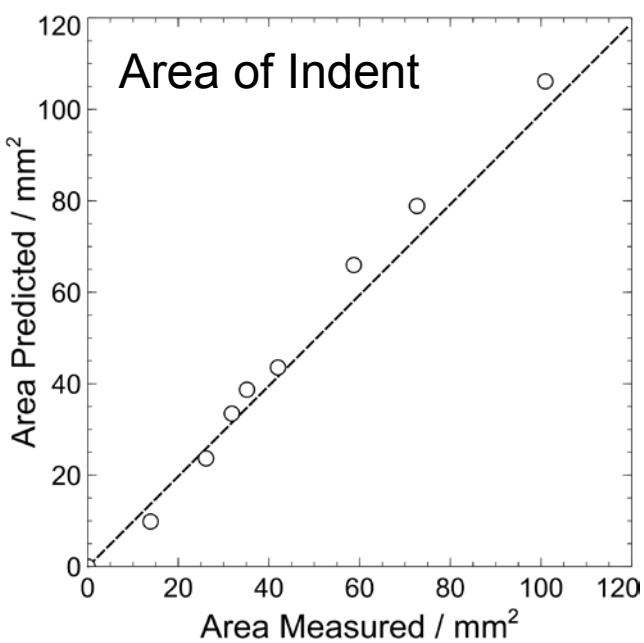
Anisotropy of separator included in model

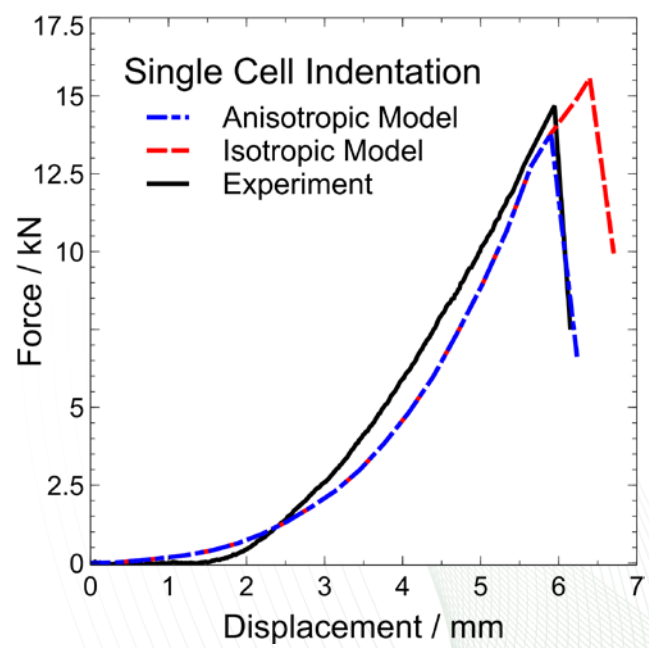
Technical Accomplishments



- Anisotropic model for separator based on Barlat model (MAT 36 in LS DYNA)
- All of the material parameters for the simulation were experimentally determined in this effort
- Failure based on critical strain

Better prediction when anisotropy of separator included.





H. Wang, *et al.*, *J. Power Sources*, 341 (2017) 156-164.

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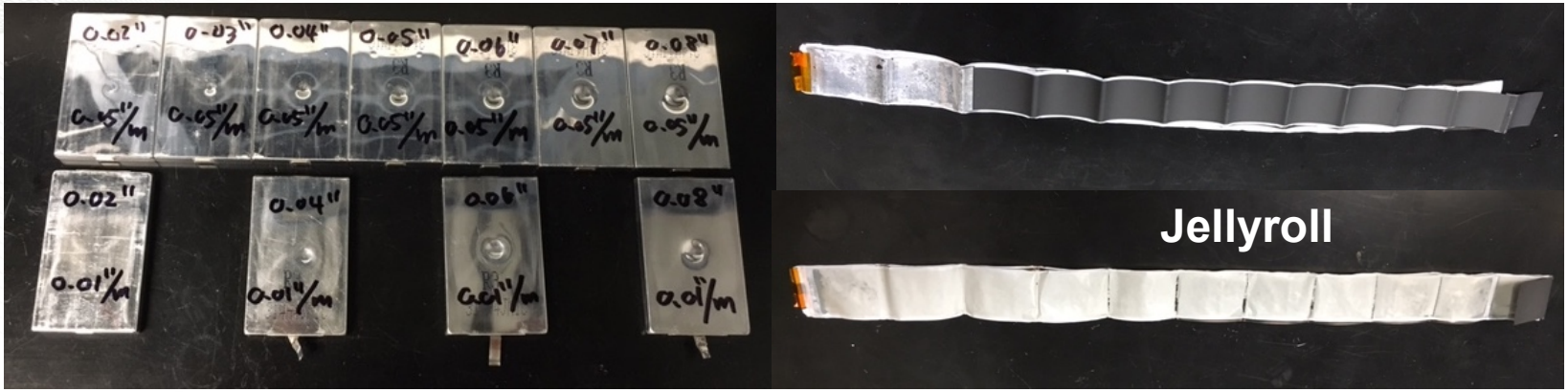
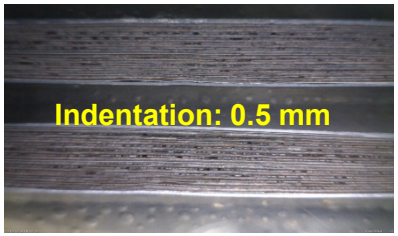
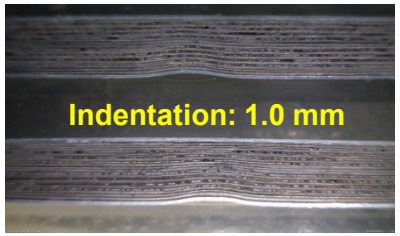
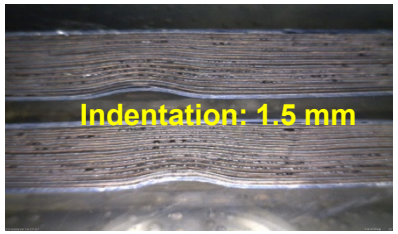
Observation of internal strain distribution and damage in cells

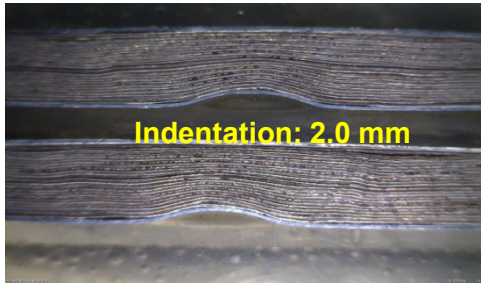
Technical Accomplishments

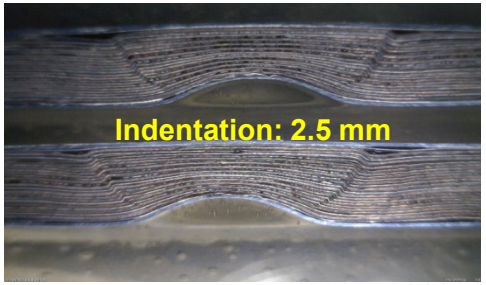




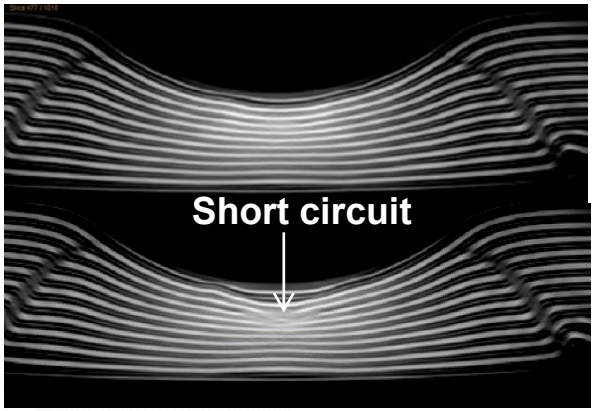








Optical Images Cross-Sections



3D X-ray Tomography Scan

We use multiple tools to investigate deformation and failure inside the cells.

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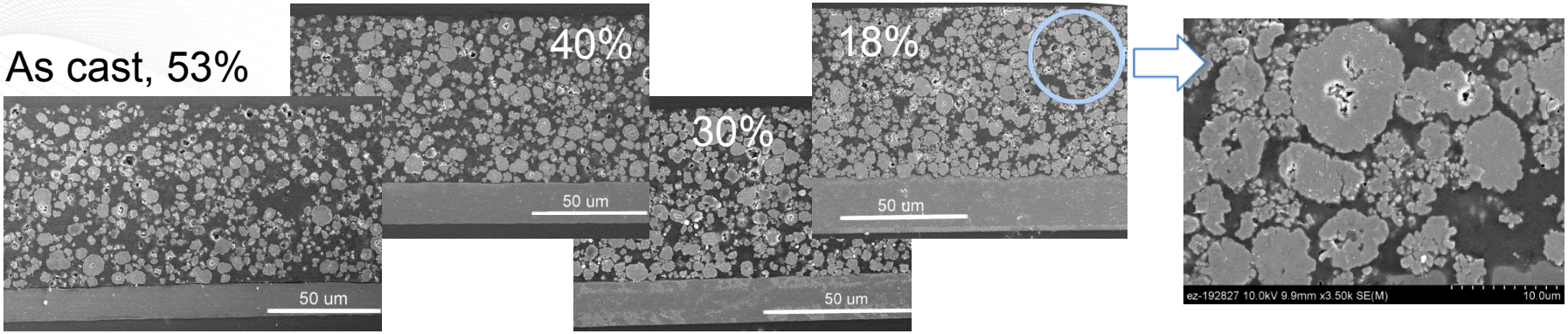
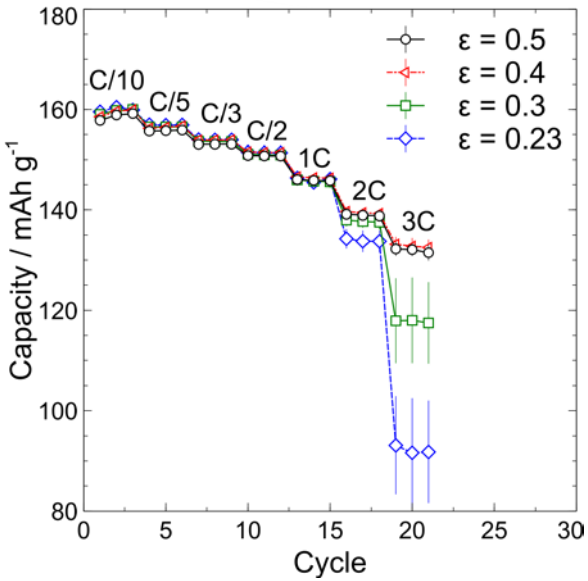
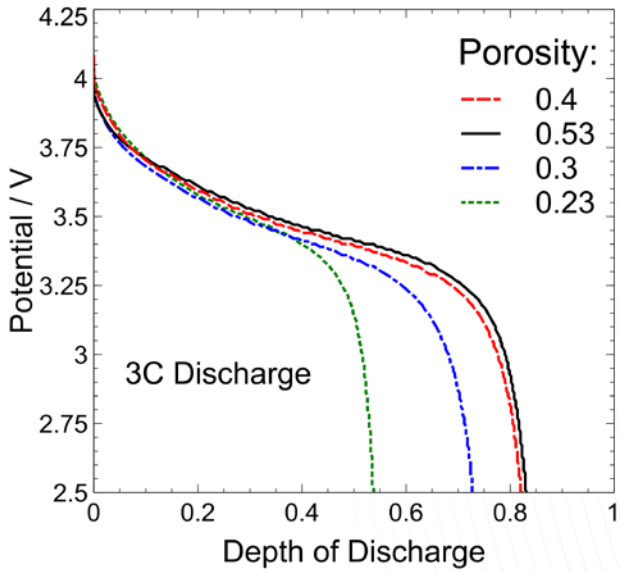
Calendering – electrochemical behavior

Technical Accomplishments







- NMC532 (Toda America); wt% NMC/PVdF/carbon black 90/5/5
- Electrodes with 6 different porosities created by calendering (Porosities: 0.53, 0.5, 0.4, 0.3, 0.23, 0.18)
- Half-cell cycling started at ORNL's Battery Manufacturing Facility

- Significant drop in capacity for $\varepsilon \leq 30\%$ and high currents.

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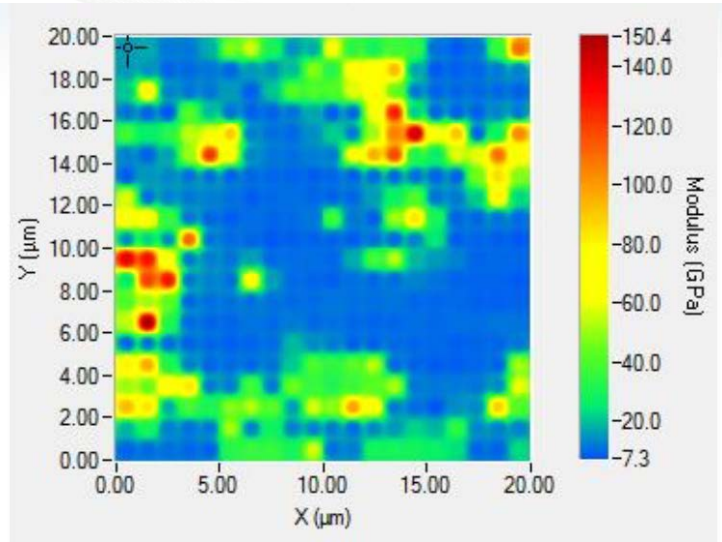
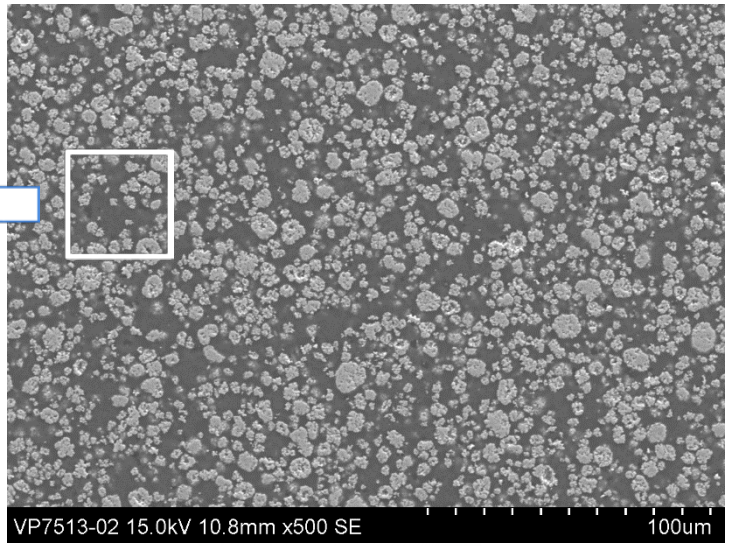
Mechanical properties of active material

Technical Accomplishments





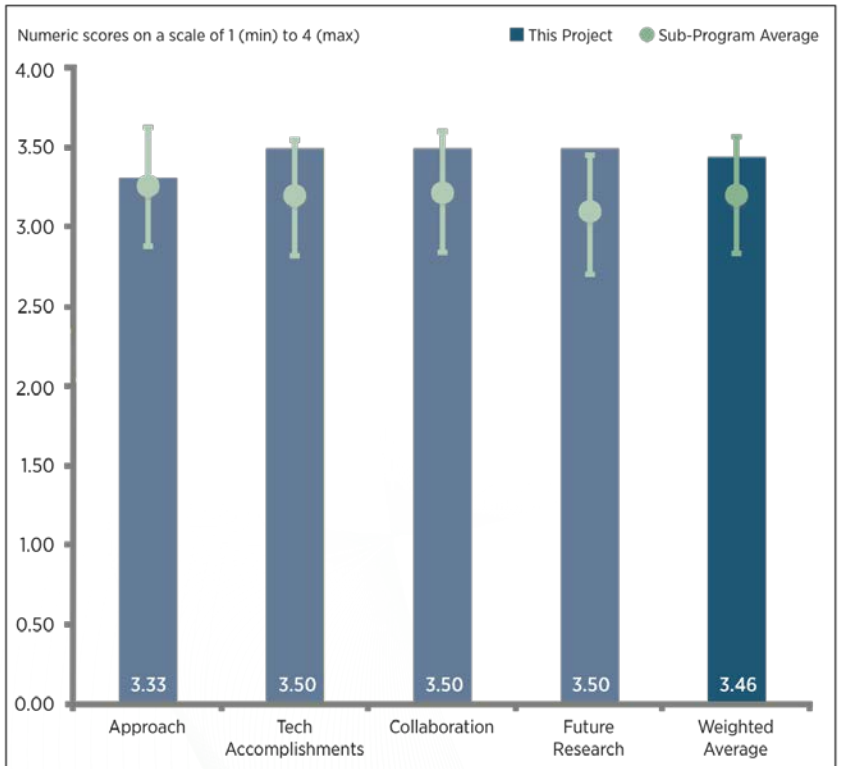
Nano-indentation of individual particles is challenging.

- Accelerated property mapping (XPM)
- Grid of nano-indentations 20x20 with 1 µm step
- Electrode filled with metallographic epoxy and polished
- 120 µN load with step duration of 0.2s

Gather data on change in mechanical properties in cycled electrodes

Responses to Previous Year Reviewers' Comments

Overall AMR16 coments were positive and encouraging. The following are the comments related to this effort:



Comment 1: More robust methodology to validate the completed models.

Response: The electrochemical and thermal models will be validated by cycling and thermal data for a Nissan Leaf module (performed in collaboration with NREL). The same data will be used to evaluate the software computational speed-up. Coupled mechanics-electrochemical models will be validated by data from cycling of cells in deformed configuration. Mechanics models will be validated against the data from mechanical testing of cells.

Comment 2: Mechanical failure is a statistical, not deterministic, process.

Response: We can address the statistical aspect of failure through simulation that includes the variation in microstructure and estimate the effect.

Collaboration and Coordination with Other Institutions

Organisation	Type of collaboration
Argonne NATIONAL LABORATORY	Tomography, diffusivity measurements. Sub in CABS project.
BERKELEY LAB	Tomography, microstructure models, diffusivity measurements. Sub in CABS project.
Sandia National Laboratories	Microstructure modeling of electrodes; computation of effective properties. Sub in CABS project.
NREL NATIONAL RENEWABLE ENERGY LABORATORY	General collaboration within CAEBAT project, experimental data exchange. NREL provides data from battery module cycling for model validation.
DOT/NHTSA	Provided Nissan Leaf modules for testing. Development of crashworthiness models for automotive batteries.
FORD	New Finite Element formulation for layered structure of batteries.

Remaining Challenges and Barriers

- Measurement of residual stress in electrode particles resulting from calendering. Nano-indentation and/or Raman microscopy will be used.
- Strength of secondary particles (NMC) measurements.
- Formulation of failure criteria for layered battery structure.
- The major chalenge remains development of techniques for validation of models, especially at the microstructure level.

Proposed future work is planned to address the above challenges

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

Proposed Future Research

- Experiments involving other modes of deformation of cell components.
- Electrochemical testing of calendered electrodes at different temperatures.
- Electrochemical testing of cells in deformed configuration.
- Include temperature and strain rate sensitivity in separator models for finite element analysis.
- Investigate residual stress in electrode particles due to calendering.
    - Evaluation of residual stress adds additional component which may help separating changes in performance due to reduction in porosity and thickness from those due to coupling with stress.
- Models upscaling stress-strain relationships to the electrode level.
    - Started in 2nd quarter of FY 2017
    - Developed methods for derivation of microstructure tensors from X-ray tomography and 2D data
    - Next steps:
        1. *Formulation of modified stress and stiffness tensors based on microstructure models and experiments*
        2. *Yield function definition*
        3. *Correlation with experiments*

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

Summary

Relevance

- Better understanding of mechanics of batery materials and their failure enables development of improved predictive models for battery safety.

Approach

- Development of experimental protocols to measure mechanical properties of electrode materials and cell components.
- Development of experimental protocols to study failure leading to internal short circuit.
- Formulation of new models for mechanical behavior of battery components.
- Formulation of microstructure-based continuum model.

Accomplishments and progress

- Complete study of strain rate and temperature effects on mechanical behavior of battery polymer separators.
- Implemented anisotropy of separator into finite element model for battery.
- Dedeloped method to study details of internal failure in a battery under mechanical loading.
- Studied effect of electrode calendering on electrochemical performance of positive electrodes.
- Developed methods for derivation of microstructure tensors from X-ray tomography and 2D data

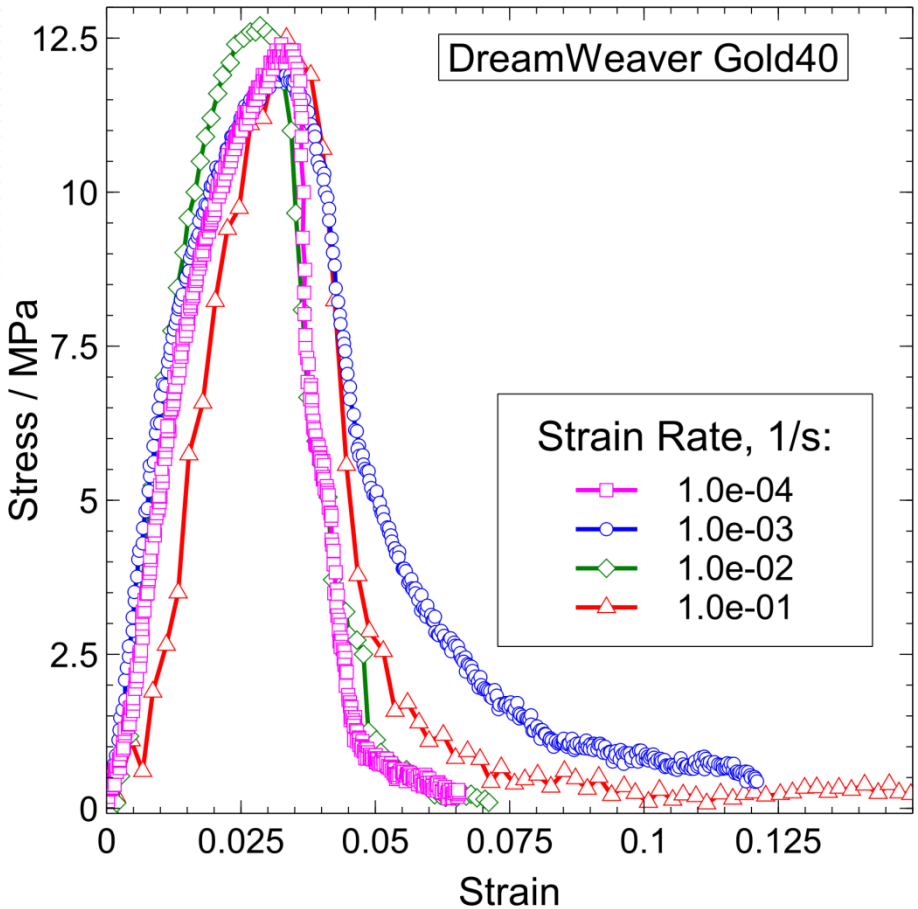
Future work

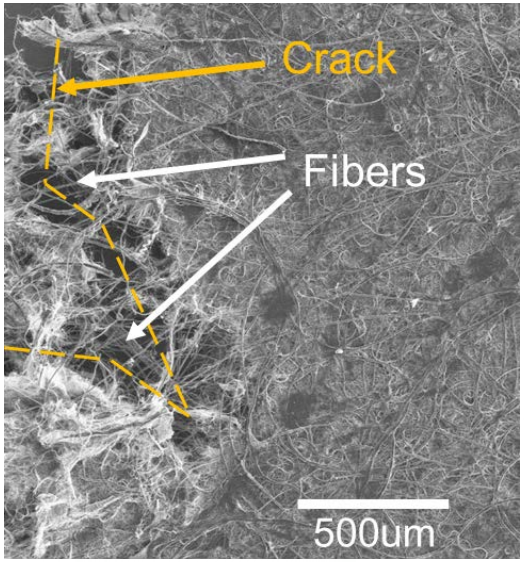
- Development of microstructure-based continuum model
- Integration of cell component models into finite element computations
- Investigating other modes of deformation of battery components

Technical Back-Up Slides

High-porosity DreamWeaver Gold 40 Battery Separator

Technical Back-Up Slides

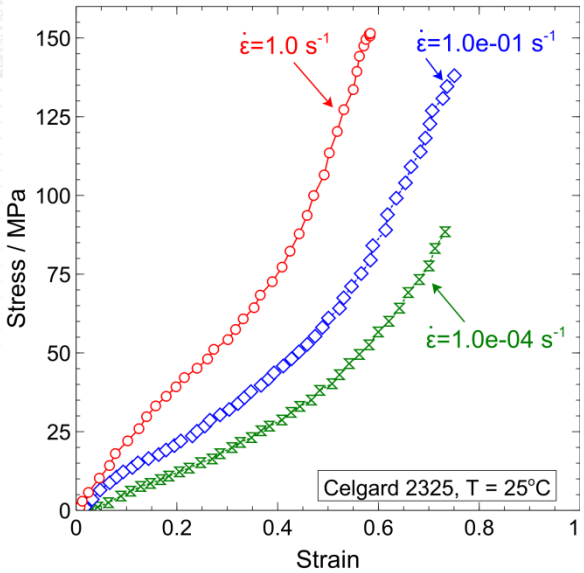


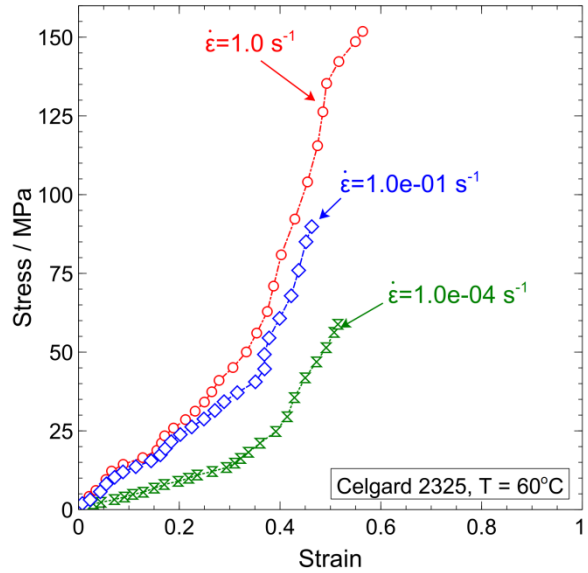


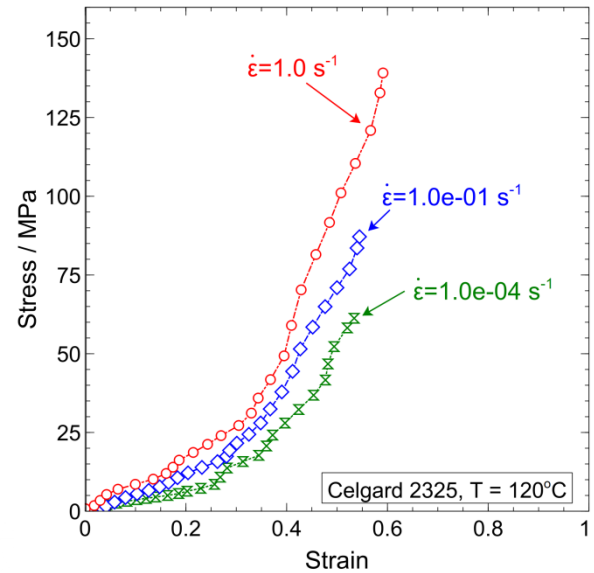
- Isotropic
- No strain-rate sensitivity
- Capable of supporting some load via fibers after fracture initiation

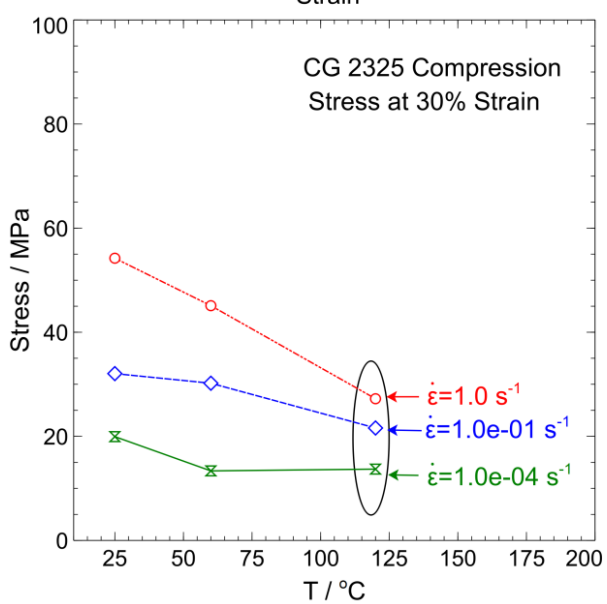
Effect of temperature – compression

Technical Back-Up Slides









For comparison purpose – stress at mid-strain (30%)

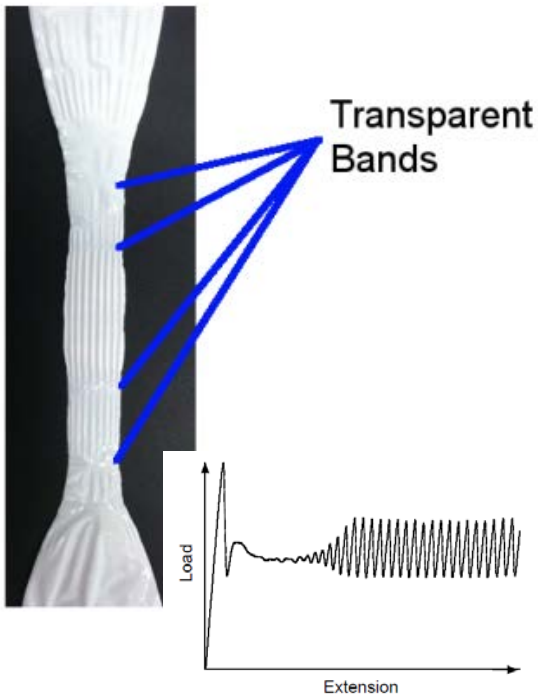
- Rate effects become less pronounced with temperature
- Trends are similar to tension but not as significant
- Temperature has little effect on slow strain rate behavior

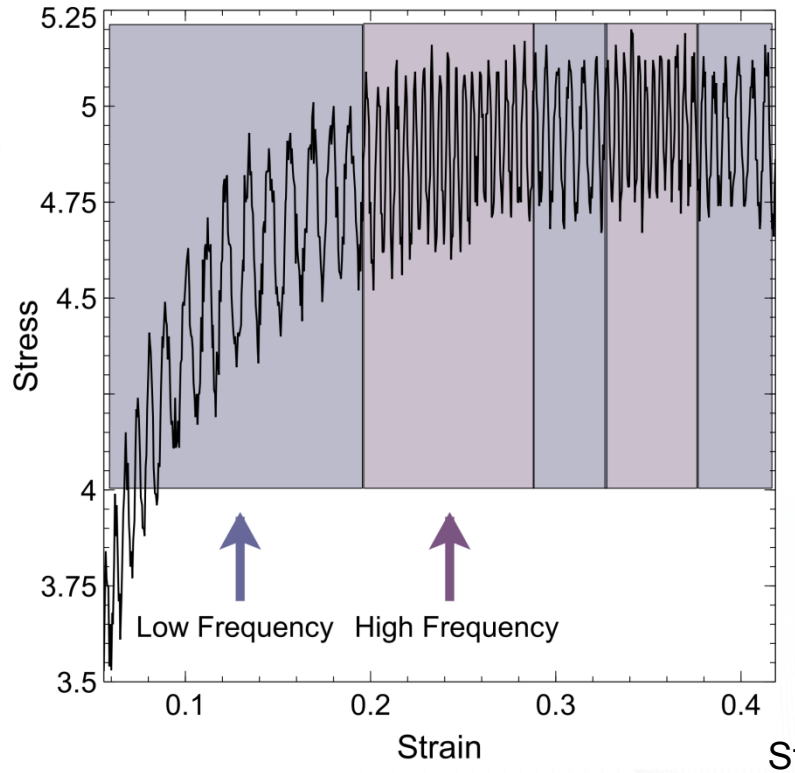
CABS Consortium for Advanced Battery Simulation

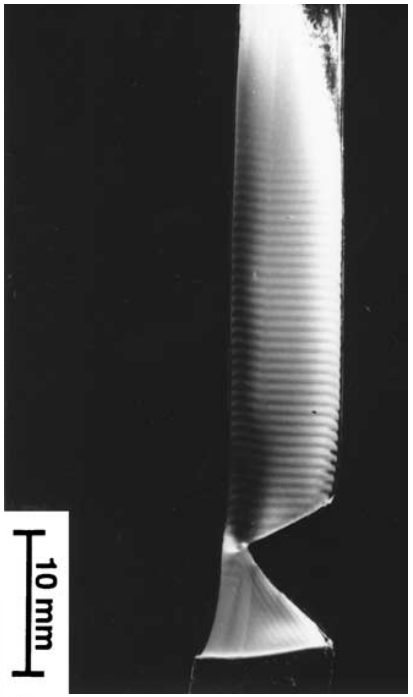
Oak Ridge National Laboratory

Strain distribution and stick-slip mechanism

Technical Back-Up Slides







Striations associated with neck propagation in poly(ethylene terephthalate) (PPET)

J. Karger-Kocsis, *et al*, J Mater Science 36 (2001)
A. Toda *et al*., Polymer 42 (2002).

- Stress oscillations more pronounced in HT testing
- Indicative of stick-slip mechanism and neck propagation
- Frequency of oscillations has been reported as a function of drawing rate; observed mix of frequencies