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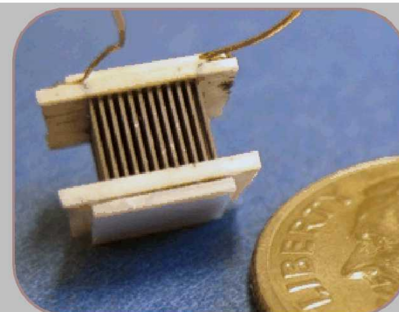

Thin Film Thermal Battery Development for High Rate Applications

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Future of Munitions Batteries Workshop

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U.S. DEPARTMENT OF ENERGY
NNSA National Nuclear Security Administration

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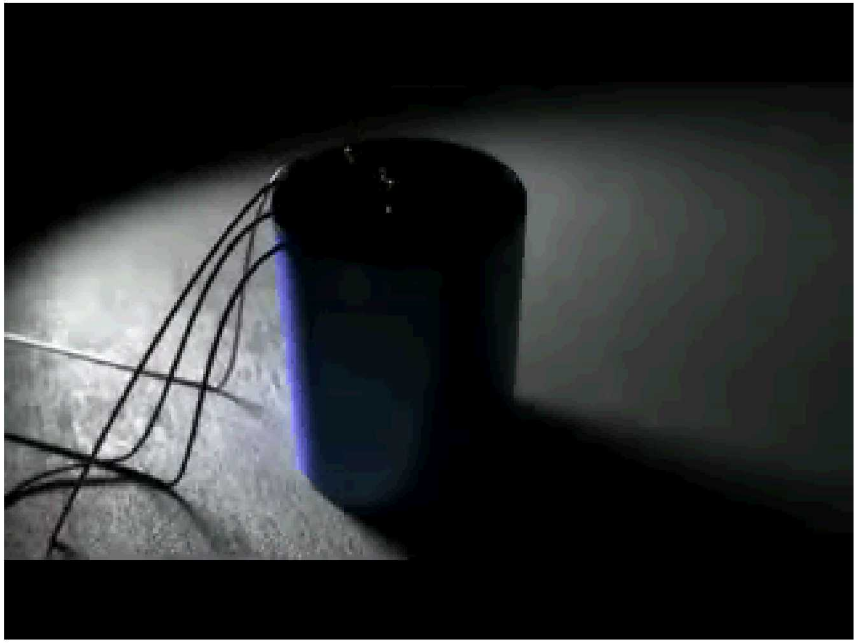
Outline

- Motivation
- Issues with current binders
- Cathode Coatings
- Cathode Test Data
- Separator Overcoating
- Separator Test Data
- Challenges in Adhesion/Cohesion
- Anode Coatings
- Next Steps

Introduction

- Thermal Batteries are excellent power sources that can survive storage for long periods of time with little change in performance
- Current methods of production are expensive, slow, and result in significant yield loss, which drives price up
- They can only be produced in cylindrical format, which forces system designers to work around this geometry
- There is a practical lower limit to size, imposed by manufacturing method
- A thermal battery that can be produced using a new method could circumvent all these limitations

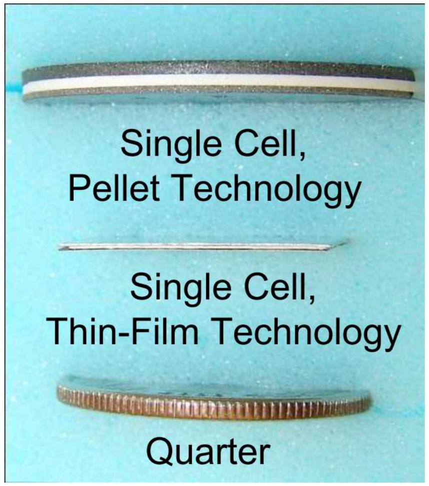


Manufacturing Batteries

Conventional Pellet Press

.VS

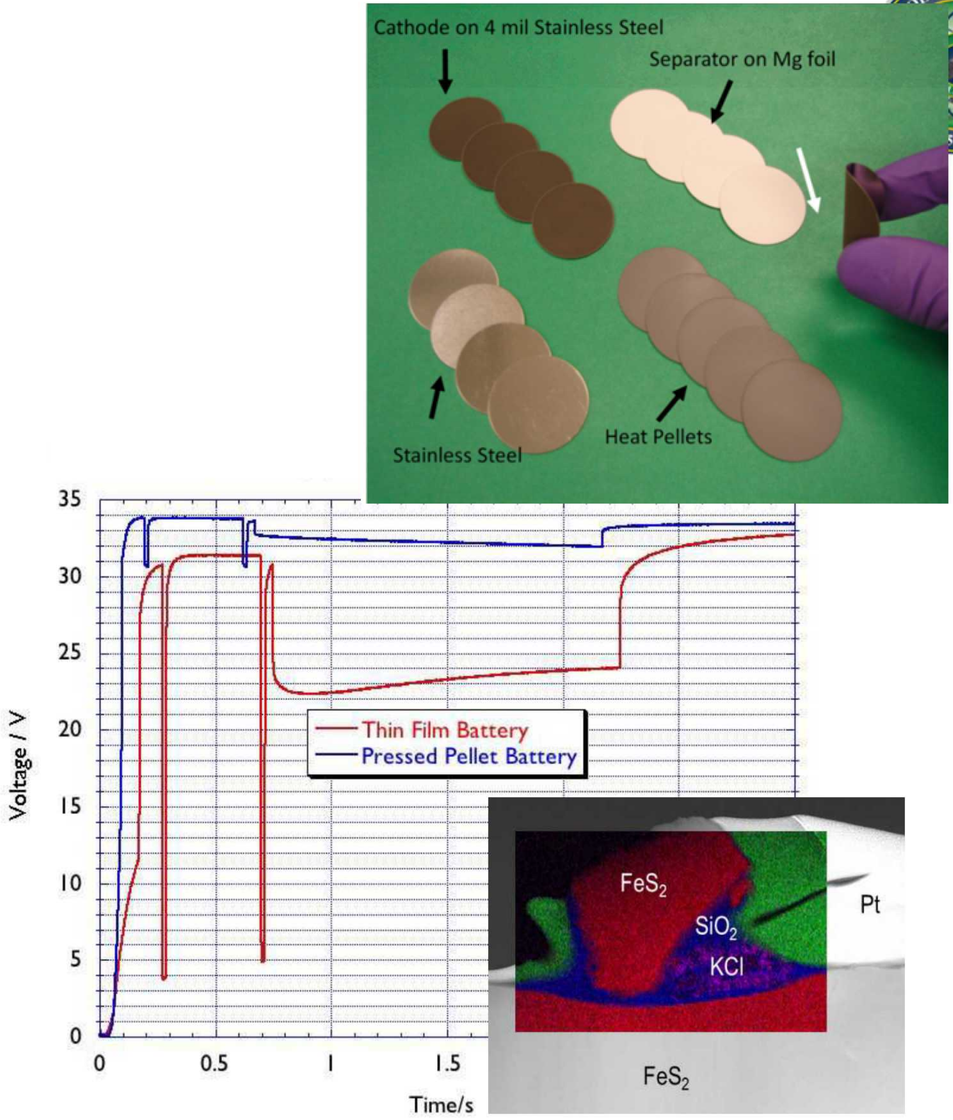
Reel to Reel Coating System



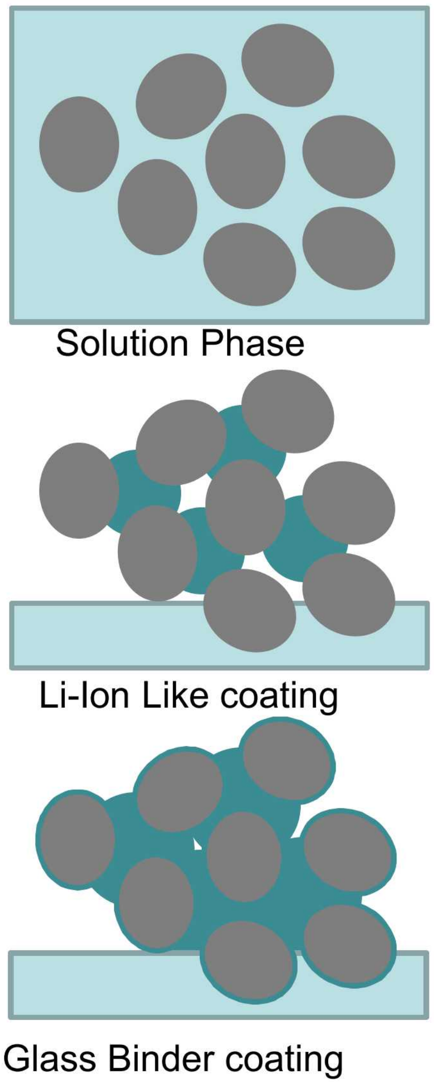
Zero Tolerance Punch

Motivation

- Cells containing a silica binder already exist, and meet most requirements
- However, high rate discharge (>300mA/cm^2) result in significant polarization
- Believed to be due to silica coating FeS_2 during processing, adding a high resistance element to the cathode

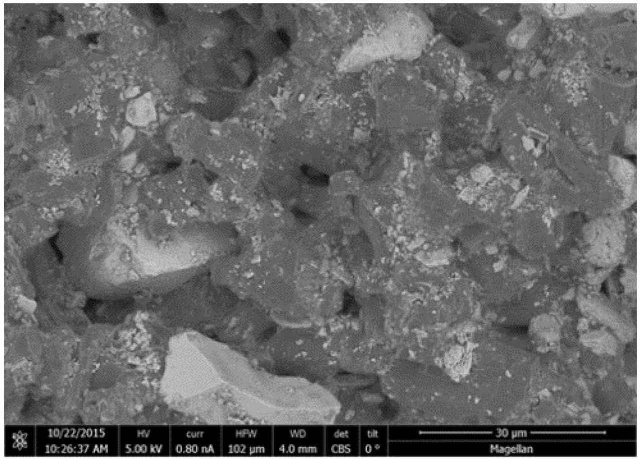


Alternative to Silica Binder

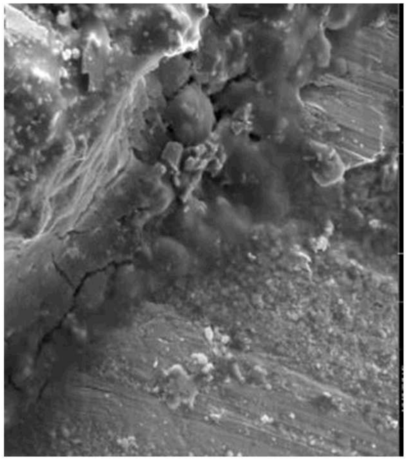


- In a suspension, the particles are separated and surrounded by solvent and binder
- In a Li-ion battery, this binder content is low (~3%), and is well below the solubility limit
- In the silica binder system, the solubility of the silicates in xylene is moderate, and therefore it starts to precipitate while there is still significant solvent left (also why it sticks to current collectors so well)
- With our improvments, we hope to get back to a system more like the Li-ion system

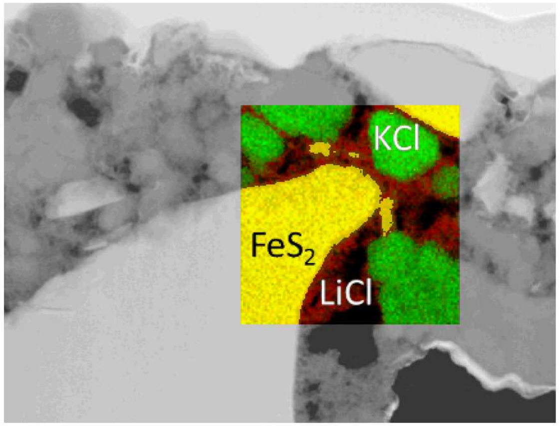
Structure of New Binder System



binder yields coherent film with well incorporated FeS_2 active material



binder yields film well adhered to current collector

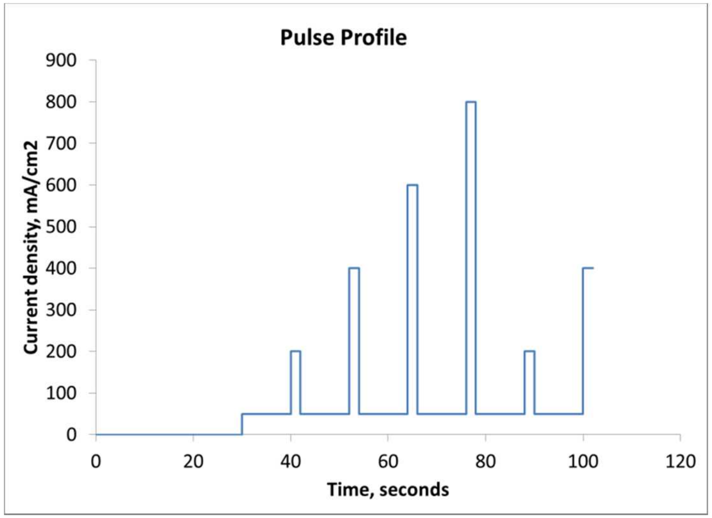


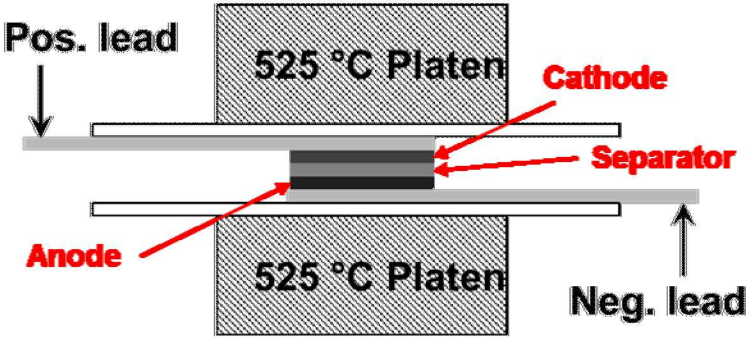
salt is in good contact with active FeS_2 surfaces

High quality electrode utilizing binder

Test Protocol

- Cathode films tested at 525° C vs. pellet anode of Li-Si
- Trying to get higher performance rates as well as data on polarization and interfacial losses
- Use "pulse-like" test profile to deliver both standby and load currents
- 50mA/cm^2 standby, 10 seconds
- 200, 400, 600, 800mA/cm^2 rates for 3 seconds
- Minimum voltage 0.1V

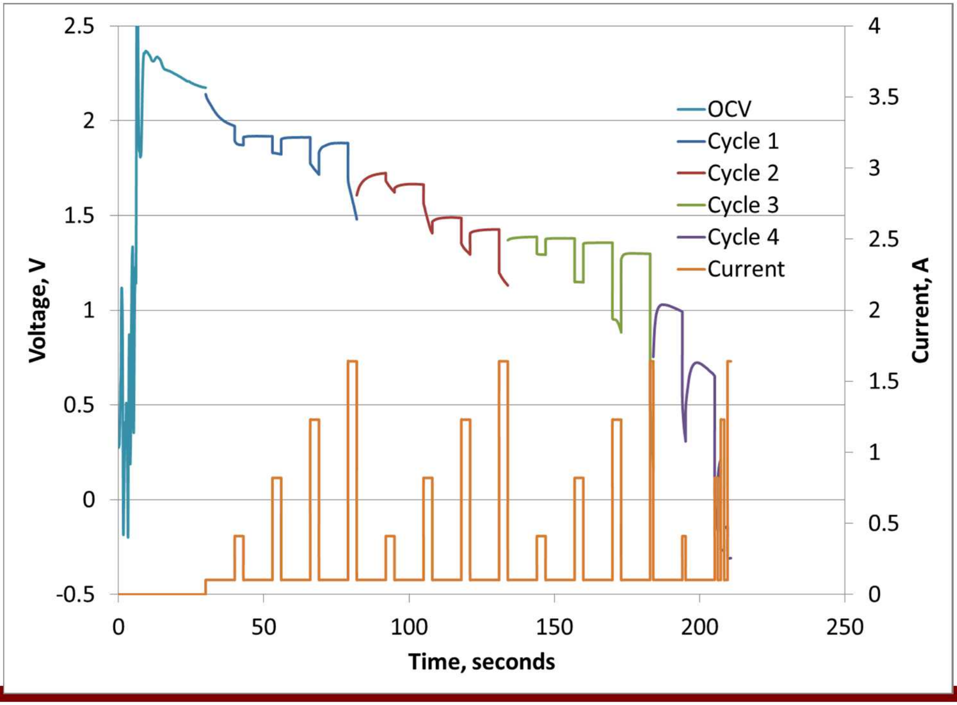




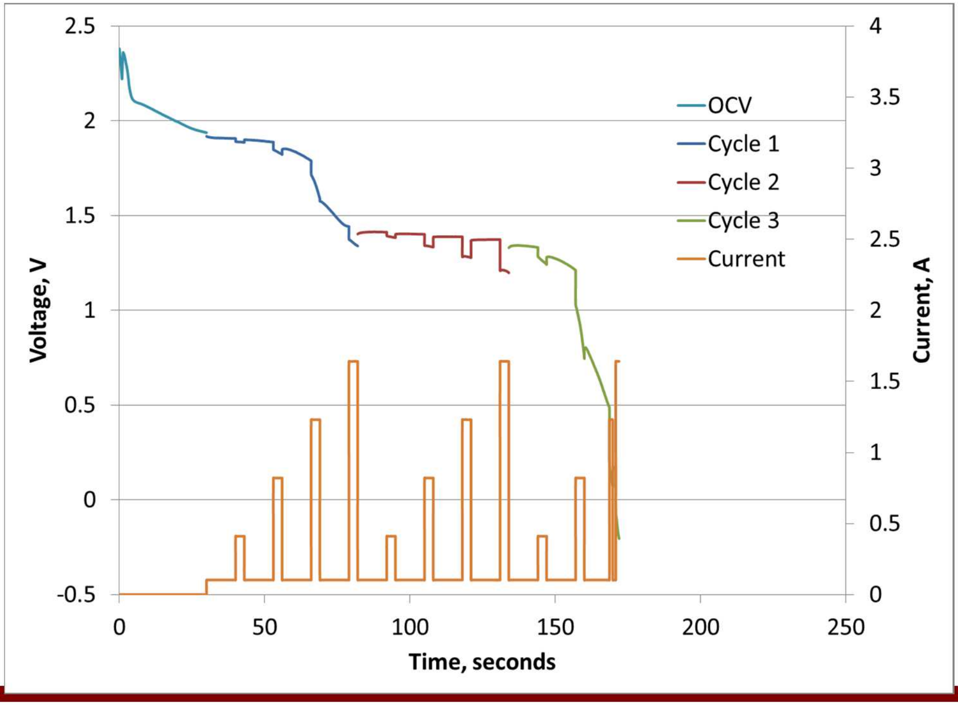
Test Results

- films were cast thinner and heated under vacuum to fully remove solvent
- Longer cell life and better high current performance

5 psi compression

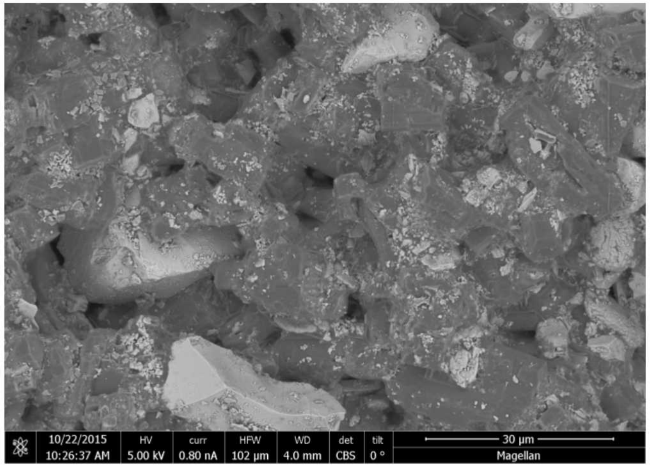


20 psi compression



Better dispersion with higher loading

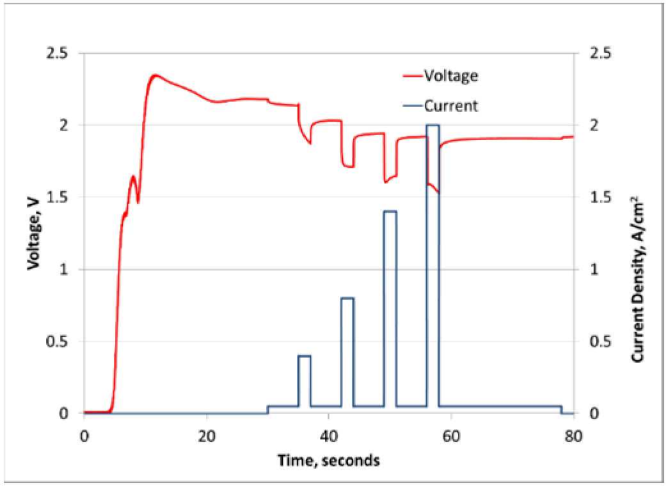
- mixed with milling media for 4 hours and then cast
- Allows for thinner electrode casting and higher solids content
  - 15 mil WFT
  - 38 vol % solids
- Significantly improved coating quality



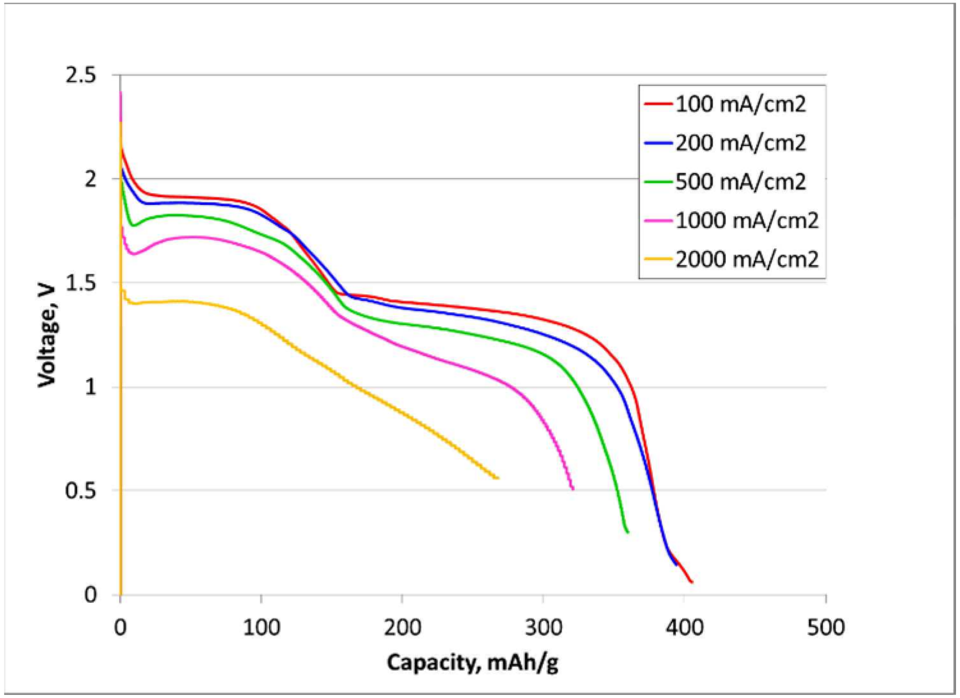
Cathode Performance

- Demonstrated effective performance at up to 2 A/cm^2 current draw

OCV = 2.20 V

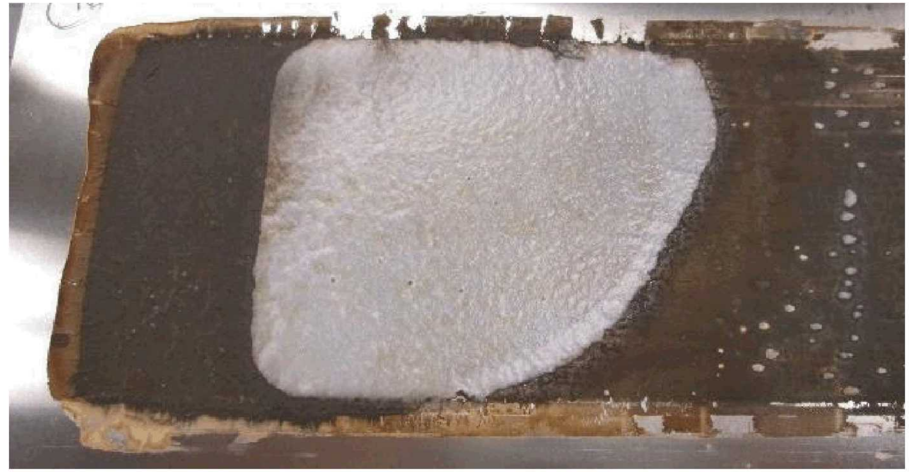


Applied Current (mA/cm^2)	Polarization (mV)
400	200
800	300
1400	320
2000	350

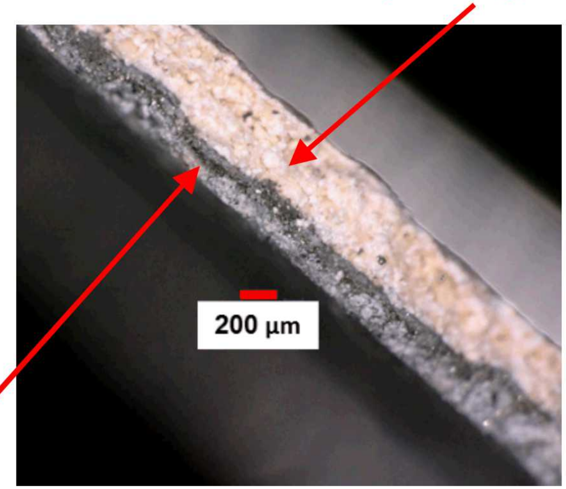


Separator Overcoat

- **Coat and dry cathode coating on stainless steel**
- **Switch to separator coating**
- **Overcoat and dry**
- **Punch and test**



400µm separator film

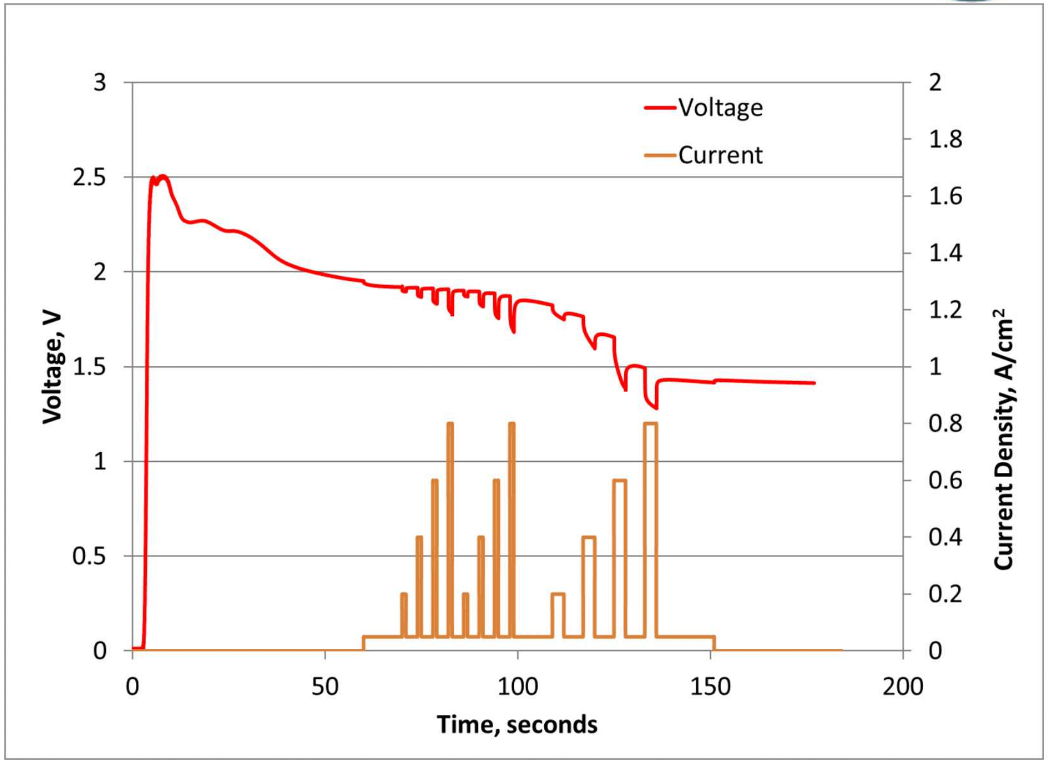


200µm cathode film

Separator Performance

- DMSO coated separator showed stable voltage and low polarization on pulses
- Low rise time retained from coated separator

Applied Current (mA/cm^2)	Polarization (mV)
200	20
400	45
600	75
800	115

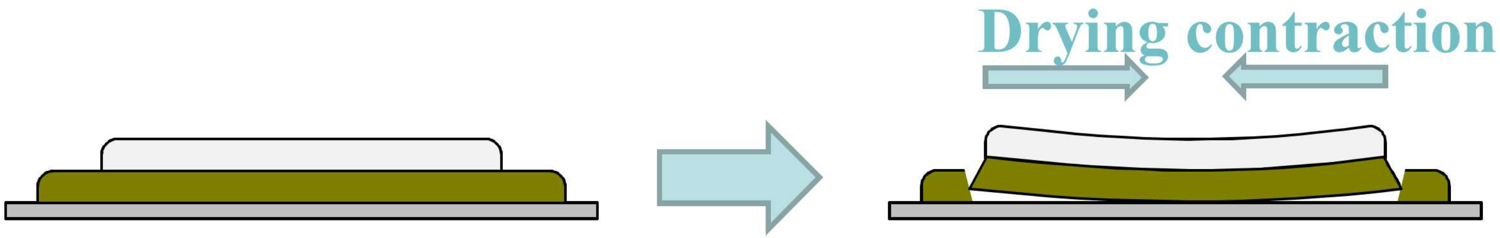


Pellet stack impedance: **0.44** Ω-cm^2

Cast cathode stack impedance: **0.21** Ω-cm^2

Cast cathode/separator stack impedance: **0.16** Ω-cm^2

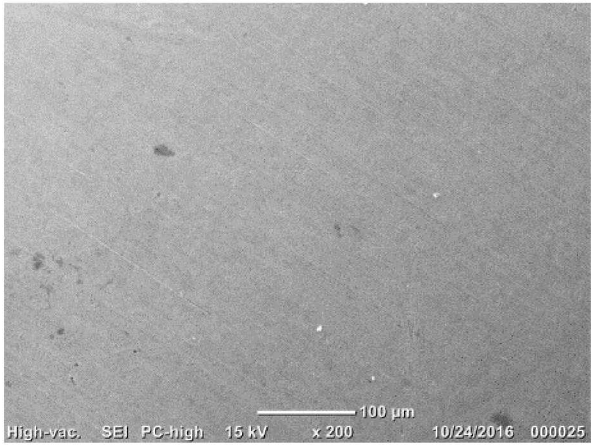
Delamination Issues with Overcoat



- Cathode films generally show good adhesion to current collectors but upon separator overcoat they delaminate along the separator borders
  - Contraction of separator upon drying delaminates cathode
    - Greater adhesion between separator/cathode than cathode/current collector
  - Explore more compliant and/or rougher surface current collectors
  - Explore alternative coating configurations

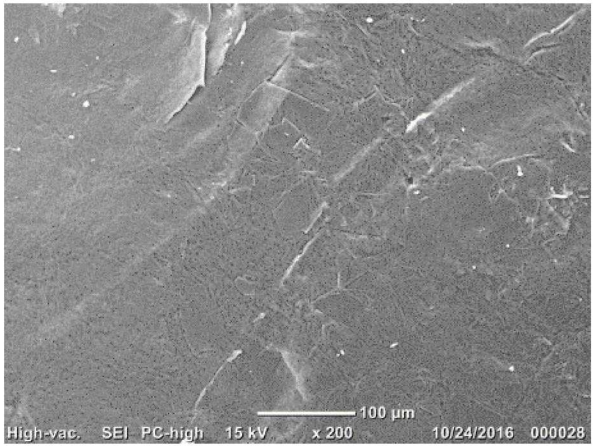
Cathode: New Current Collectors

Stainless Steel



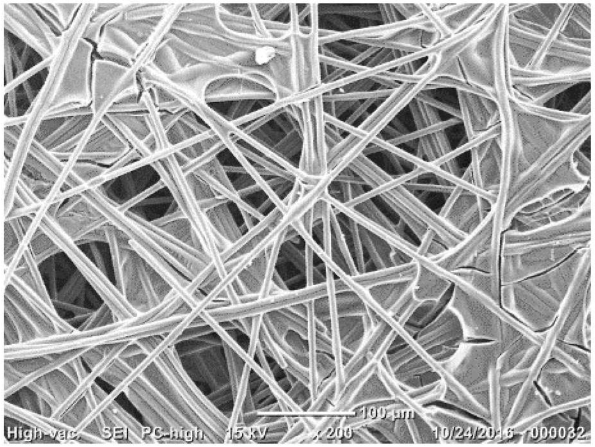
- Legacy material
- Smooth, non-porous surface
- Stiff, not-compliant

Graffoil



- Improved performance in actual batteries
- More compliant, rougher surface

Carbon Paper



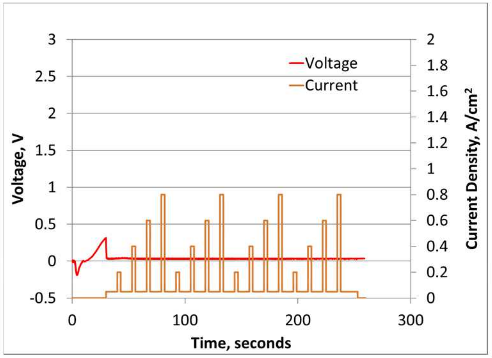
- Rough, porous surface for improved film adhesion
- Somewhat compliant

Primary motivation comes from cathode delamination during separator overcoat and lower salt content of anode compatible solvents

Current Anode Film Development

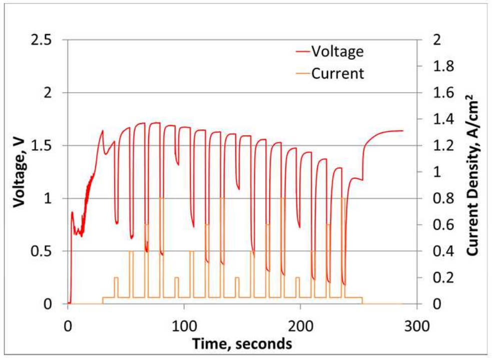
Binder 1

- Solvent was seen to visibly react with Li-Si
- **Near zero voltage** in test cell
- Unable to support any current draw



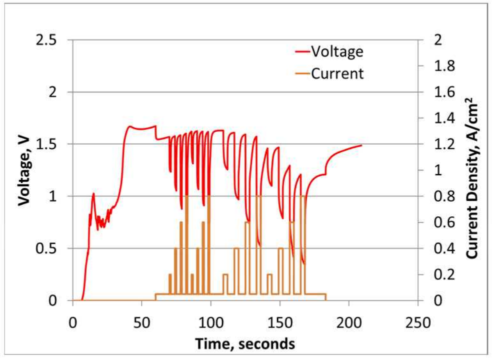
Binder 2

- likely a contributor to observed **slow rise time**
- Voltage at ~ 1.7V
- **High polarization** upon current pulses



Binder 3

- likely a contributor to observed **slow rise time**
- Voltage at ~ 1.7V
- **High polarization** upon current pulses



Future Work

- Improve film quality and solve delamination issue of overcoat separators
  - Change coating configurations, reduce coating thickness

  - Continue testing of alternative current collectors
  - Optimize drying schedule for combined films
- Continue development of anode films and improve performance
  - New solvents to improve film quality and reduce reactivity with Li-Si
  - New current collectors to improve adhesion
  - High temperature casting as alternative approach
- Larger scale, closer to real-world testing of cast cathodes

Cathodes for assembly into multi-cell TB stack

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