Investigating the Ability of Plastic Current Collectors to Isolate Internal Shorts in High Energy Cells

With the European Synchrotron Radiation Facility

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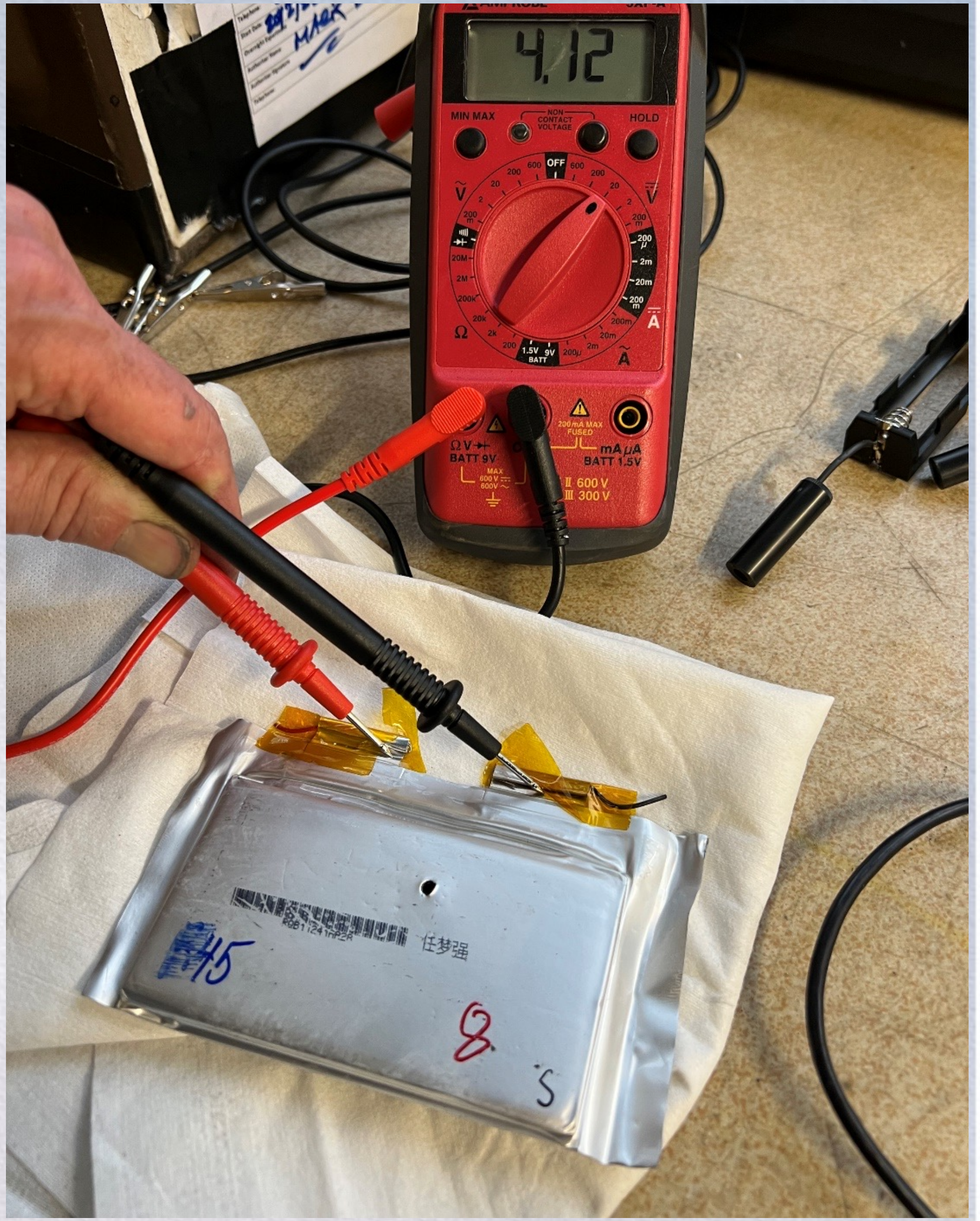
NASA Aerospace Battery Workshop
Huntsville, AL
14-16 Nov 2023

Agenda

- Motivation
  - What limits the effectiveness of Plastic Current Collectors (PCCs) in isolating shorts?
  - Obviate the design burden of achieving passive propagation resistance (PPR)
- Team Effort
- Cell Designs
  - 18650
  - 21700
  - 10Ah pouch
- Test and Examination Results
- Investigating Cell Design Drivers for Success



5 Design Driving Factors for Reducing Hazard Severity from a Single Cell TR

- Reduce risk of cell can side wall ruptures
  - Without structural support most high energy density (>660 Wh/L) designs are very likely to experience side wall ruptures during TR
  - Battery should minimize constrictions on cell TR pressure relief
- Provide adequate cell spacing and heat rejection
  - Direct contact between cells nearly assures propagation
  - Spacing required is inversely proportional to effectiveness of heat dissipation path
- Individually fuse parallel cells and strings
  - TR cell becomes an external short to adjacent parallel cells and heats them up
  - TR cell in a string in parallel with other strings needs fusing
- Protect the adjacent cells from the hot TR cell ejecta (solids, liquids, and gases)
  - TR ejecta is electrically conductive and can cause circulating currents
- Prevent flames and sparks from exiting the battery enclosure
  - Provide tortuous path for the TR ejecta before hitting battery vent ports equipped flame arresting screens

Reference: Darcy, E. C., Jacob, D., Walker, W., Finegan, D. P. & Shearing, P. Driving Design Factors for Safe, High-Power Batteries for Space Applications. in Advanced Automotive Battery Conference (2018).

M3 Findings and Battery Design Metrics

- All ISCD Trigger Cells activated without TR propagation. Blast plates protected axially stacked virtual cells well.
- Virtual cell degradation due to thermal abuse:
  - Top virtual cell: 2% degradation, no blown fuses
  - Middle virtual cell: 4% degradation, 3 adjacent cell fuses blown
  - Bottom virtual cell: 3% degradation, 2 adjacent cell fuses blown
- PPR battery energy: **4.8 kWh** with 134P-3S electrical topology (12 Molicel M35A trigger cells)
- PPR Battery overall mass: 28.84 kg [63.59 lbs]
- Gravimetric specific energy: **173.6 Wh/kg**
- PPR Battery calculated mass factors:
  - Percent cell mass versus total battery mass: **64.7%**
  - Parasitic mass factor: **1.544**
- PPR Battery miscellaneous metrics:
  - Mass percentage of heat sinks: **24.3%**
  - Mass percentage of blast plates: **3.9%**

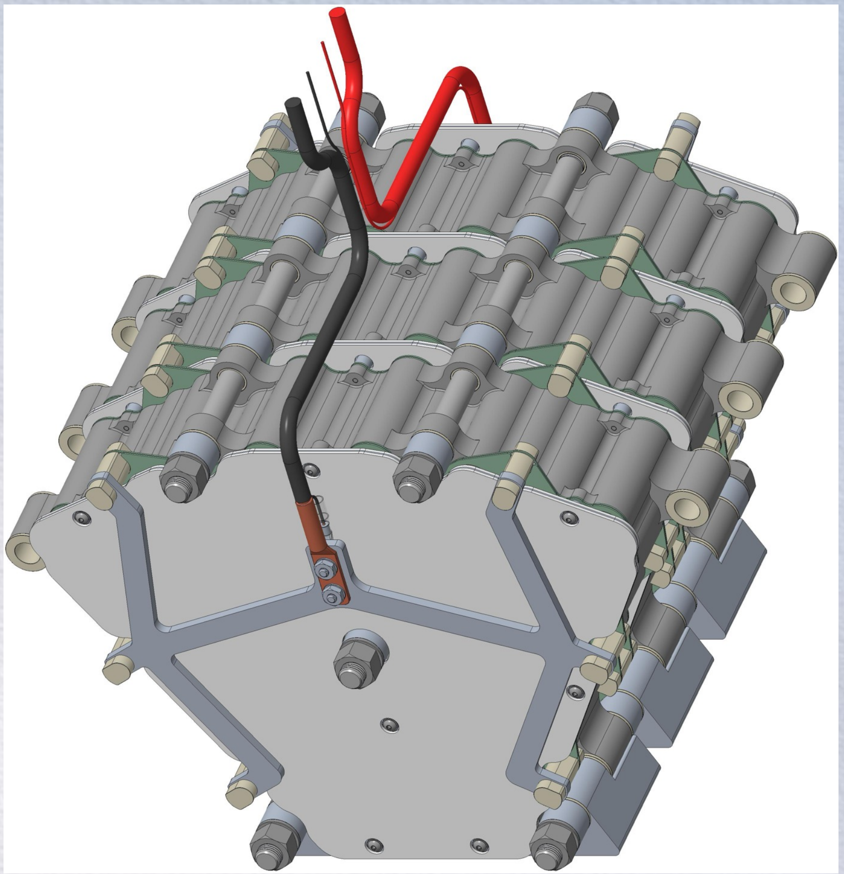
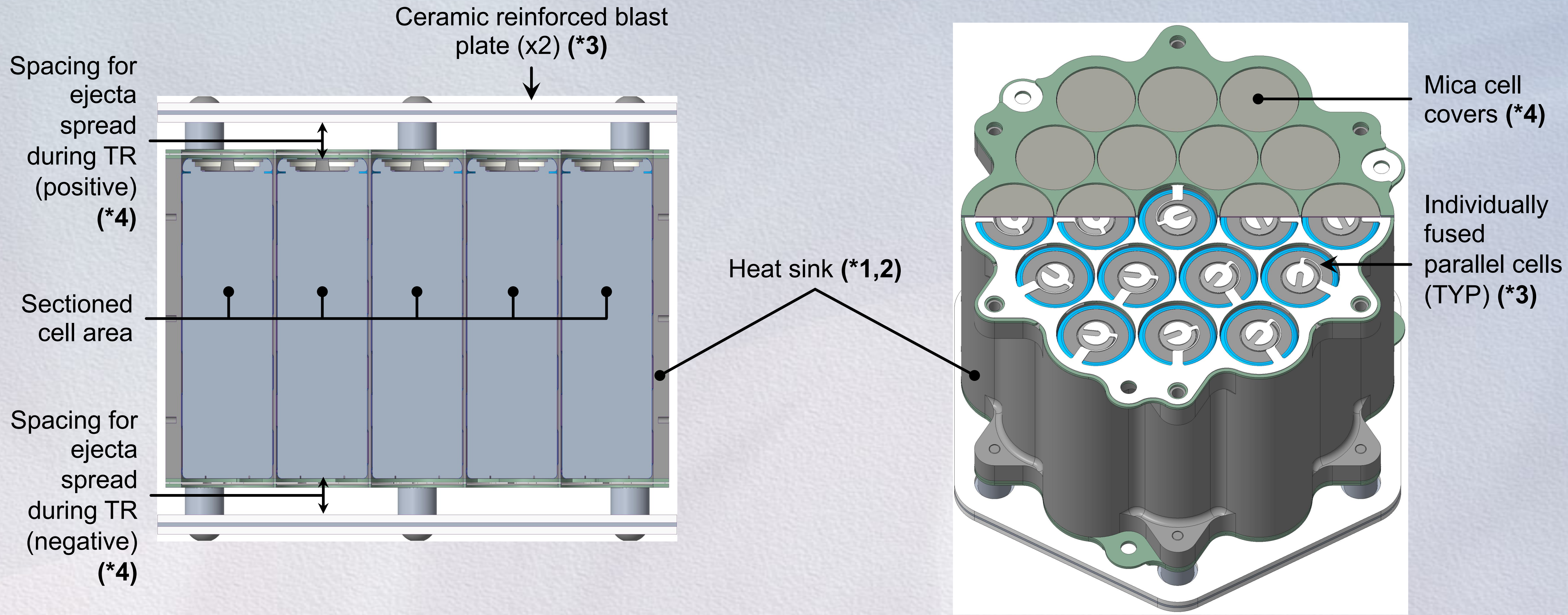


Figure: CAD rendered image of assembled M3 PPR Battery.

Petrushenko, D, "***Scale-Up Challenges for Passively Propagation Resistant Batteries: 18650 to 21700,"*** 49th Power Sources Conf, Fort Madison, Jun 2023

M5 Subscale Battery PPR Design Features



Note: an asterisk followed by a number (e.g. ***1**) indicates the PPR Battery Guideline the feature correlates to. Guideline 5 (battery enclosure) example was not represented in this battery design per the application requirements.

Petrushenko, D, "***Scale-Up Challenges for Passively Propagation Resistant Batteries: 18650 to 21700,***" 49th Power Sources Conf, Fort Madison, Jun 2023

M3, M5 Subscale Battery Energy Densities

	M3 0.020" M35A	**M5** 0.020" 50S	**M5** 0.020" M52V
Form Factor:	18650	21700	21700
Cell Type:	Molicel M35A	Samsung 50S	LG M52V
Battery Capacity:	231.1 Wh	340.3 Wh	351.5 Wh
Total Battery Mass:	1.311 kg	1.915 kg	1.863 kg
Gravimetric Energy Density:	**173.6 Wh/kg**	**177.7 Wh/kg**	**188.7 Wh/kg**
Parasitic Mass Factor:	**1.544**	**1.439**	**1.457**
Total Cell Mass Percentage:	64.7%	69.5%	68.6%
Heat Sink Mass Percentage:	24.3%	21.3%	21.9%
Blast Plate Mass Percentage:	3.9%	3.2%	3.3%

21700 cell format enables ~9% improvement in PPR battery specific energy vs 18650

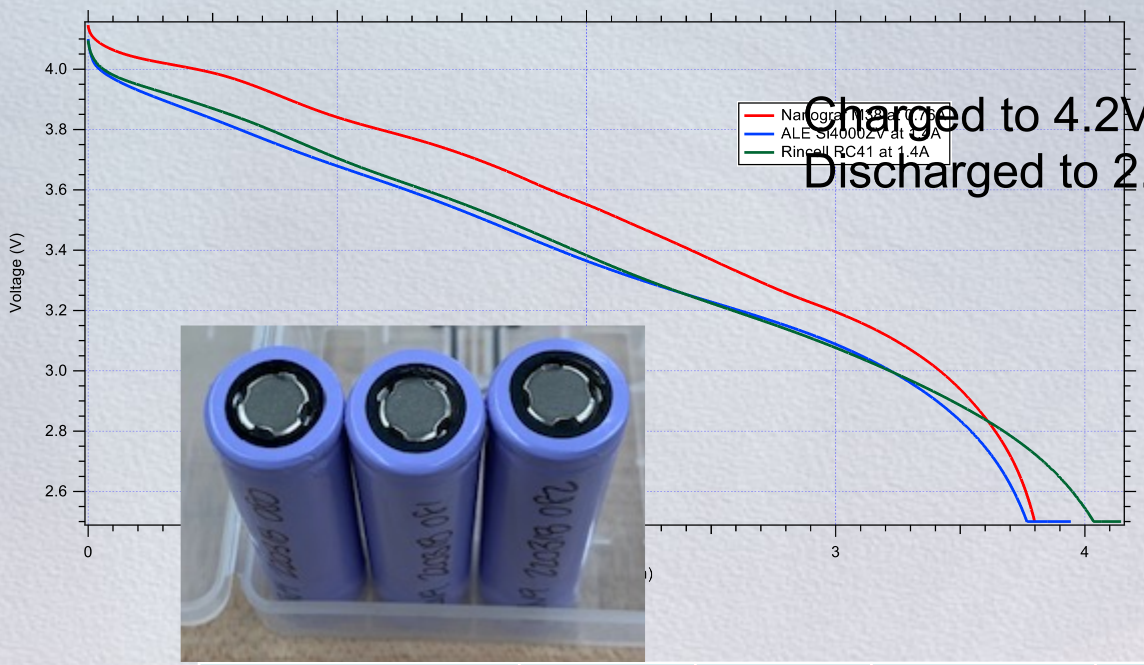
	Wh	Mass (g)	Wh/kg	TR kJ at 4.2V
M35A	12.4627	45.3692	274.7	59
M52V	18.9183	67.3159	281.0	94.6
Delta	6.456	21.947	6.3	35.6
Delta%	51.8%	48.4%	2.3%	60.3%

M52V delivers 52% more Wh and yields 60% for TR heat

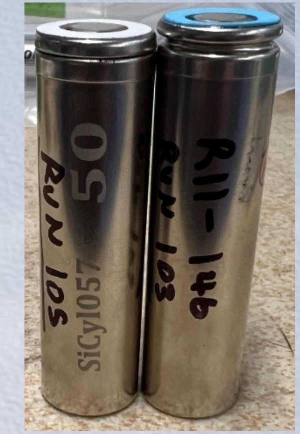
Battery design without any PPR features can achieve a parasitic mass factor of 1.2

Petrushenko, D, "***Scale-Up Challenges for Passively Propagation Resistant Batteries: 18650 to 21700,***" 49th Power Sources Conf, Fort Madison, Jun 2023

New High Capacity 18650 Cell Designs



Charged to 4.2V with 100mA taper
Discharged to 2.5V

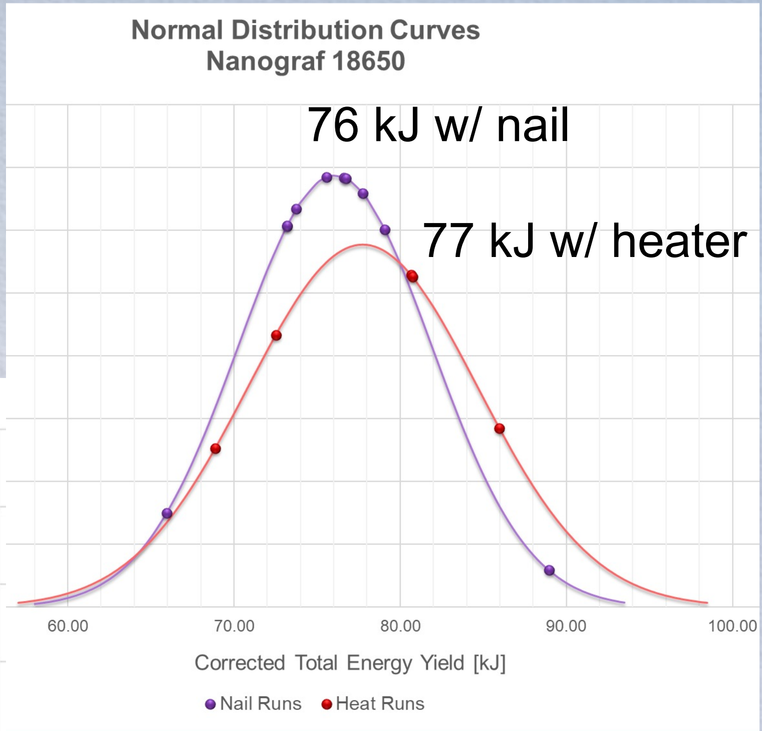


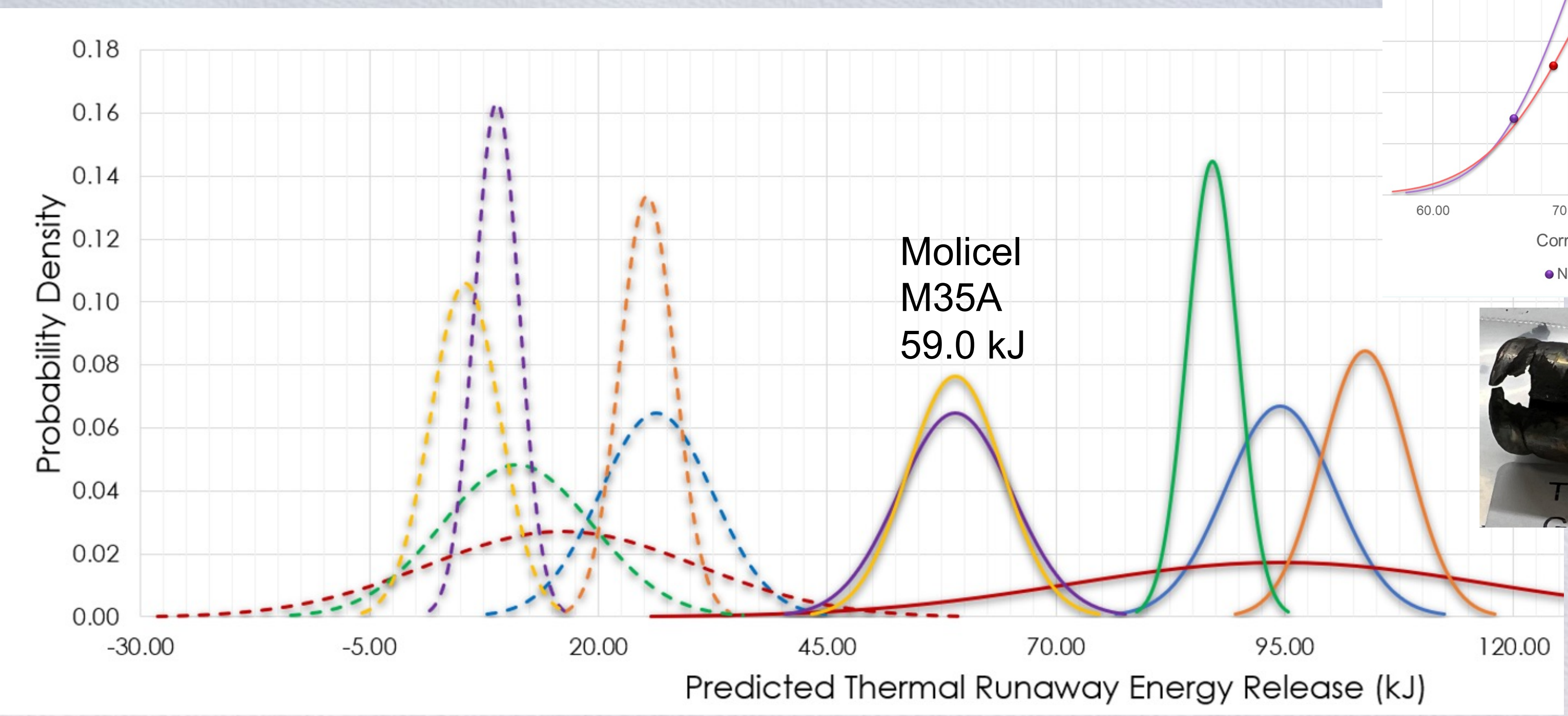
Cell Design	Ah	Wh	Mass (g)	Wh/kg
Nanograf M38	3.799	13.62	46.367	293.7
ALE Si4000ZV	3.768	12.80	45.627	280.5
Rincell RC41	4.035	13.60	46.876	290.1

Impact on TR

	Wh	Mass (g)	Wh/kg	TR kJ at 4.2V
M35A	12.4627	45.3692	274.7	59
M38	13.6	46.876	290.1	76
Delta	1.137	1.507	15.4	17.0
Delta%	9.1%	3.3%	5.6%	28.8%

Nanograf M38 delivers 9% more on discharge energy but generates 29% more TR heat than Molicel M35A

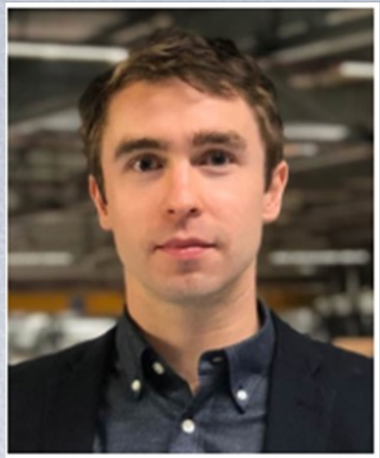




Team Effort

Dr. Eric Darcy John Darst David Petrushenko Jesus Trillo

NASA-JSC



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- Donal Finegan/NREL, Golden, CO, USA
- Charlie Kirchner-Burles, Mark Buckwell, Hamish Reid, Matilda Fransson, and Rhodri Jervis/UCL, London, UK
- Inez Kesuma and Paul Shearing, Oxford University, Oxford, UK
- Ludovic Broche and Alexander Rack/ESRF, Grenoble, France
- Jesse Mutter, Josh Gaskin, and Ed Buiel/Coulometrics, Chattanooga, TN, USA
- Brian Morin & Carl Hu/SoteriaBIG, Greenville, SC, USA

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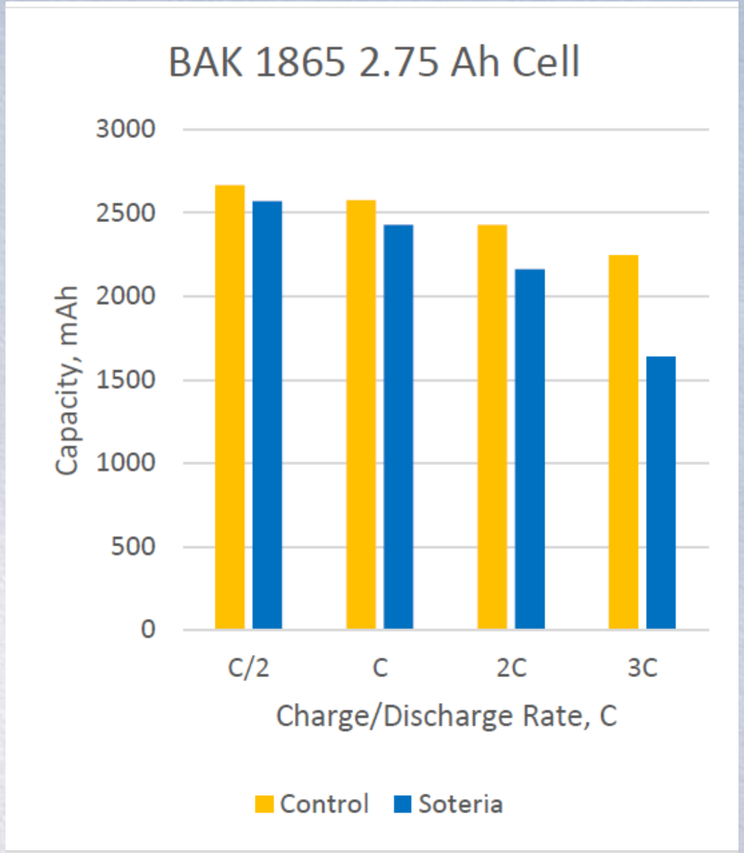
- Orion, CCP, HLS, and NAVSEA

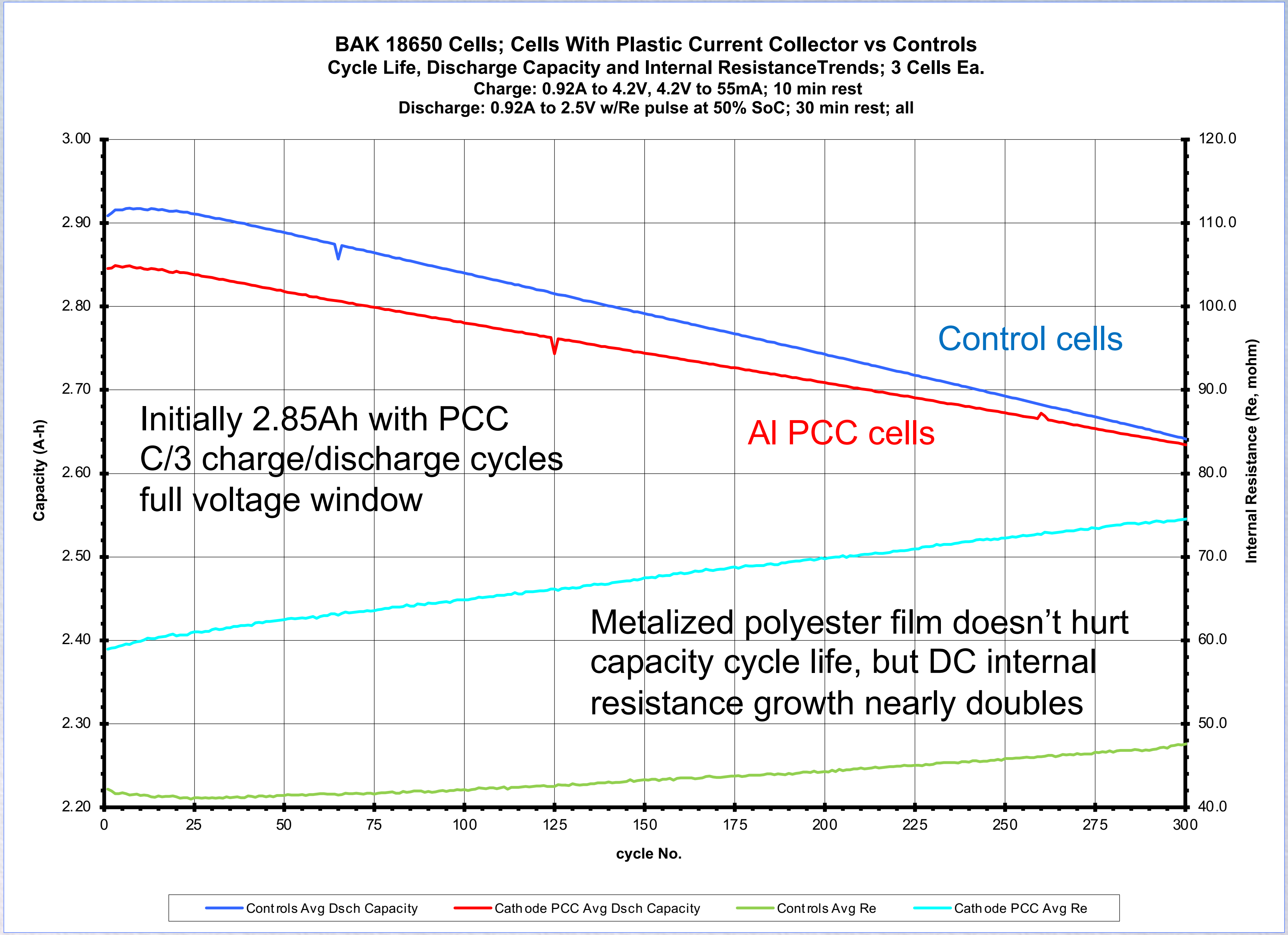
BAK 18650 Cell Designs

Soteria Cell Preliminary Specifications	
Manufacturer	BAK Power Battery
Separator	Polyolefin Film
Current Collector	Soteria Al, Standard Cu
Nominal Voltage	3.6V
Capacity	2.75Ah
AC impedance	51 mΩ
Weight	44 g
Energy Density	200 Wh/kg
Voltage Range	2.5V-4.2V

Control Cell Preliminary Specifications	
Manufacturer	BAK Power Battery
Separator	Polyolefin Film
Current Collector	Standard Foils
Nominal Voltage	3.6V
Capacity	2.75Ah
AC impedance	34 mΩ
Weight	45 g
Energy Density	209 Wh/kg
Voltage Range	2.5V-4.2V



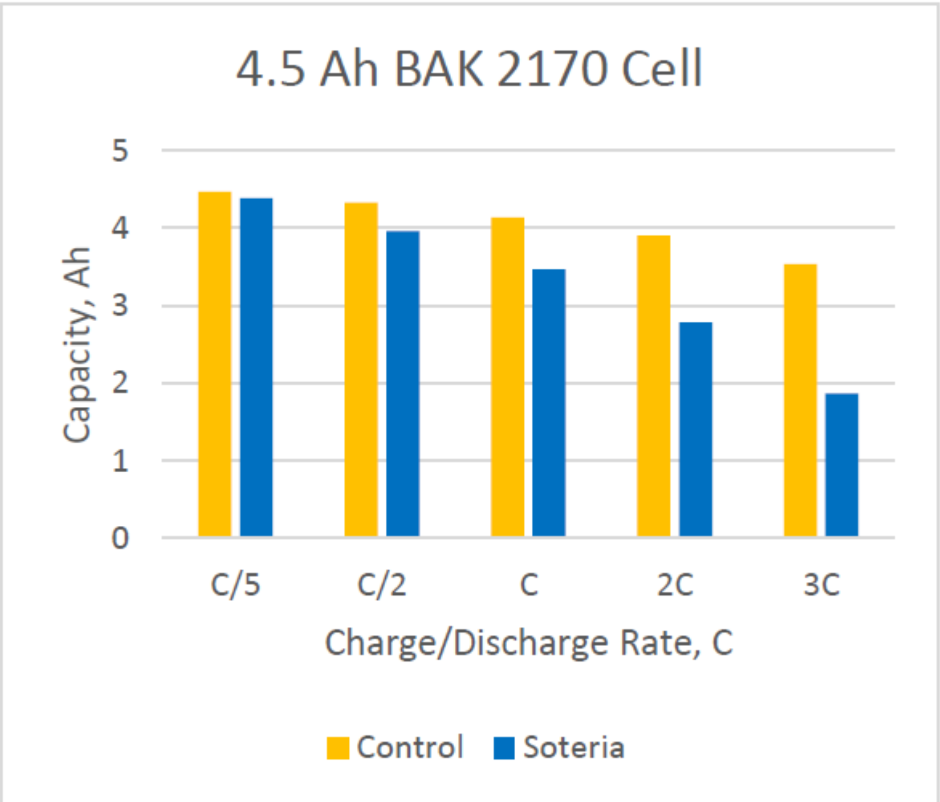
- Soteria polyester PCC only applied to cathode
- Impressive power capability (2C) with Soteria's PCC, not too far behind control cells
- They have found a decent tab to PCC welding schedule
- Measured 233 Wh/kg and 622 Wh/L on initial cycle with the Al PCC achieving 2.85Ah


BAK 18650 Cells; Cells With Plastic Current Collector vs Controls
Cycle Life, Discharge Capacity and Internal ResistanceTrends; 3 Cells Ea.
Charge: 0.92A to 4.2V, 4.2V to 55mA; 10 min rest
Discharge: 0.92A to 2.5V w/Re pulse at 50% SoC; 30 min rest; all
Capacity (A-h)
Internal Resistance (Re, mohm)
cycle No.
Control cells
Al PCC cells
Initially 2.85Ah with PCC
C/3 charge/discharge cycles
full voltage window
Metalized polyester film doesn't hurt
capacity cycle life, but DC internal
resistance growth nearly doubles
Controls Avg Dsch Capacity
Cathode PCC Avg Dsch Capacity
Controls Avg Re
Cathode PCC Avg Re

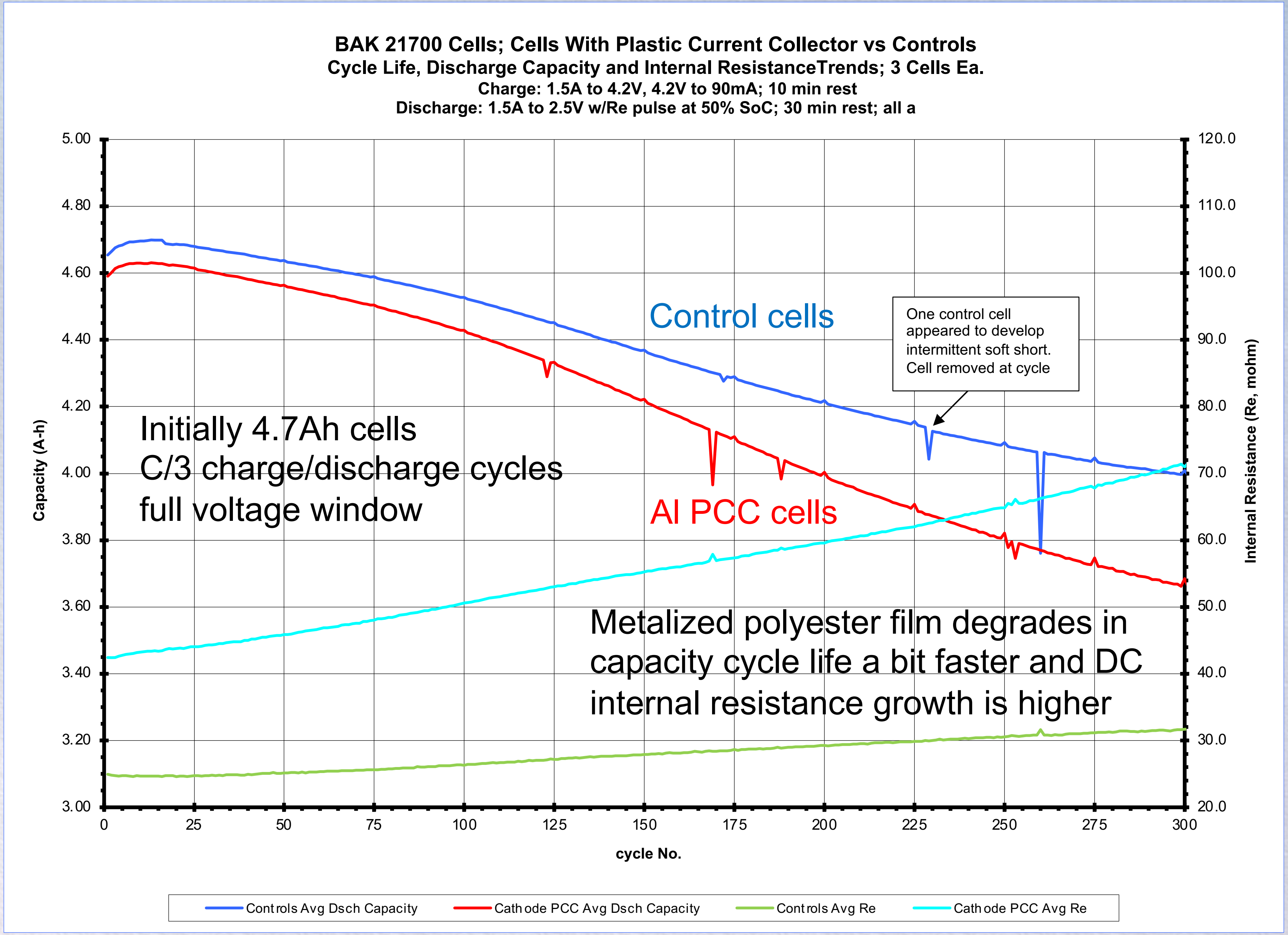
BAK 21700 Cell Designs

Soteria Cell Preliminary Specifications	
Manufacturer	BAK Power Battery
Separator	Polyolefin Film
Current Collector	Soteria Al, Standard Cu
Nominal Voltage	3.6V
Capacity	4.5Ah
AC impedance	42 mΩ
Weight	66 g
Energy Density	205 Wh/kg
Voltage Range	2.5V-4.2V

Control Cell Preliminary Specifications	
Manufacturer	BAK Power Battery
Separator	Polyolefin Film
Current Collector	Standard Foils
Nominal Voltage	3.6V
Capacity	4.5Ah
AC impedance	20 mΩ
Weight	67 g
Energy Density	227 Wh/kg
Voltage Range	2.5V-4.2V

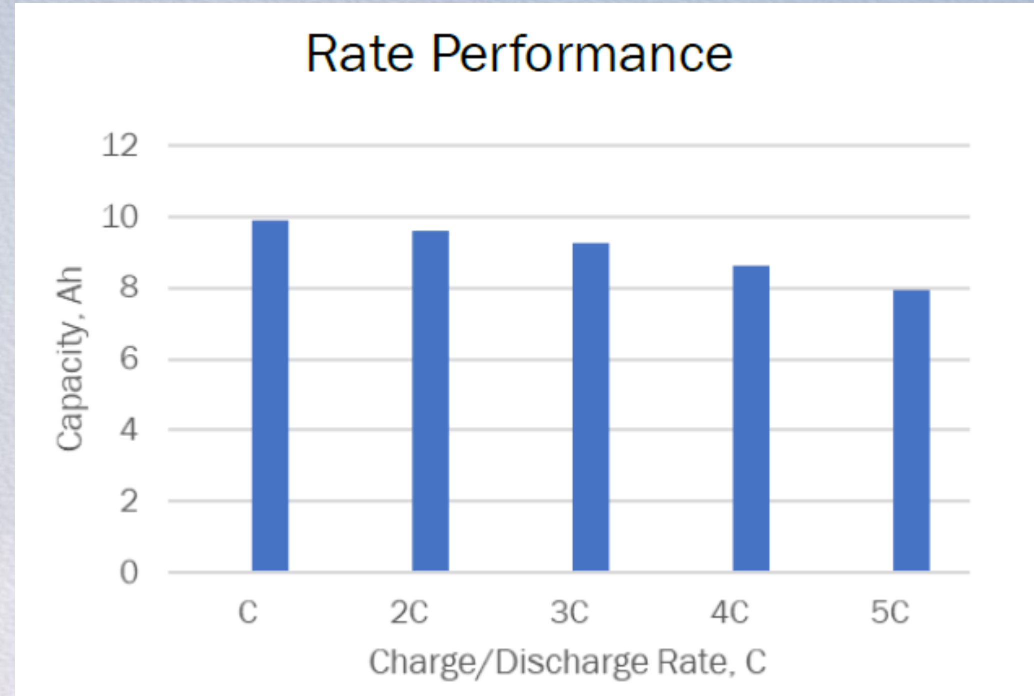


- Soteria polyester PCC only applied to cathode
- Decent power capability (up to 1C) with Soteria's PCC, not too far behind control cells
- They have found workable tab-to-PCC welding schedule
- Measured 251 Wh/kg and 684 Wh/L on initial cycle with the Al PCC


BAK 21700 Cells; Cells With Plastic Current Collector vs Controls
Cycle Life, Discharge Capacity and Internal ResistanceTrends; 3 Cells Ea.
Charge: 1.5A to 4.2V, 4.2V to 90mA; 10 min rest
Discharge: 1.5A to 2.5V w/Re pulse at 50% SoC; 30 min rest; all a
Capacity (A-h)
Internal Resistance (Re, mohm)
cycle No.
5.00
4.80
4.60
4.40
4.20
4.00
3.80
3.60
3.40
3.20
3.00
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
0
25
50
75
100
125
150
175
200
225
250
275
300
Control cells
Al PCC cells
One control cell appeared to develop intermittent soft short. Cell removed at cycle
Initially 4.7Ah cells
C/3 charge/discharge cycles
full voltage window
Metalized polyester film degrades in capacity cycle life a bit faster and DC internal resistance growth is higher
Controls Avg Dsch Capacity
Cathode PCC Avg Dsch Capacity
Controls Avg Re
Cathode PCC Avg Re

SVolt 10Ah Pouch Cell Design

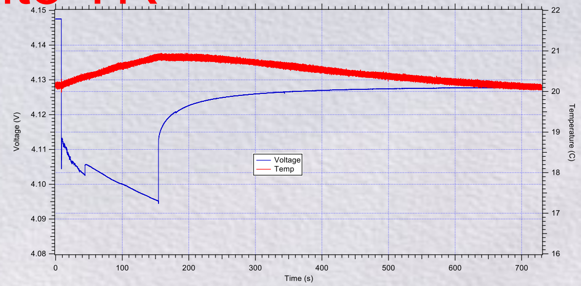
Cell Specifications	
Manufacturer	SVolt Energy
Separator	Polyethylene w/ ceramic coating
Current Collector	Soteria Al Standard Cu
Cathode Material	NCM 811
Anode Material	Synthetic Graphite
Capacity	10Ah
Voltage Range	3.0-4.2V
Energy Density	240 Wh/kg



- Soteria polyester PCC only applied to cathode
- Very impressive power capability (up to 5C) with Soteria's PCC
- They have found a great tab-to-PCC welding schedule
- 243 Wh/kg with NCM 811 and Al PCC

BAK 18650 2.9Ah Test Matrix and Results (Nail)

- Soteria metalized polyester (9)
  - PCC only on cathode (cell achieves 233 Wh/kg)
  - Cu foil on anode like all other features in control version
  - All 9 cells tolerated nail penetration
    - No fire, sparks, venting, or TR
- Control cells (6)
  - Al and Cu foil CCs
  - All 6 cells went into TR



$\Delta t_{max} < 1°C$
$\Delta V_{max} < 55mV$

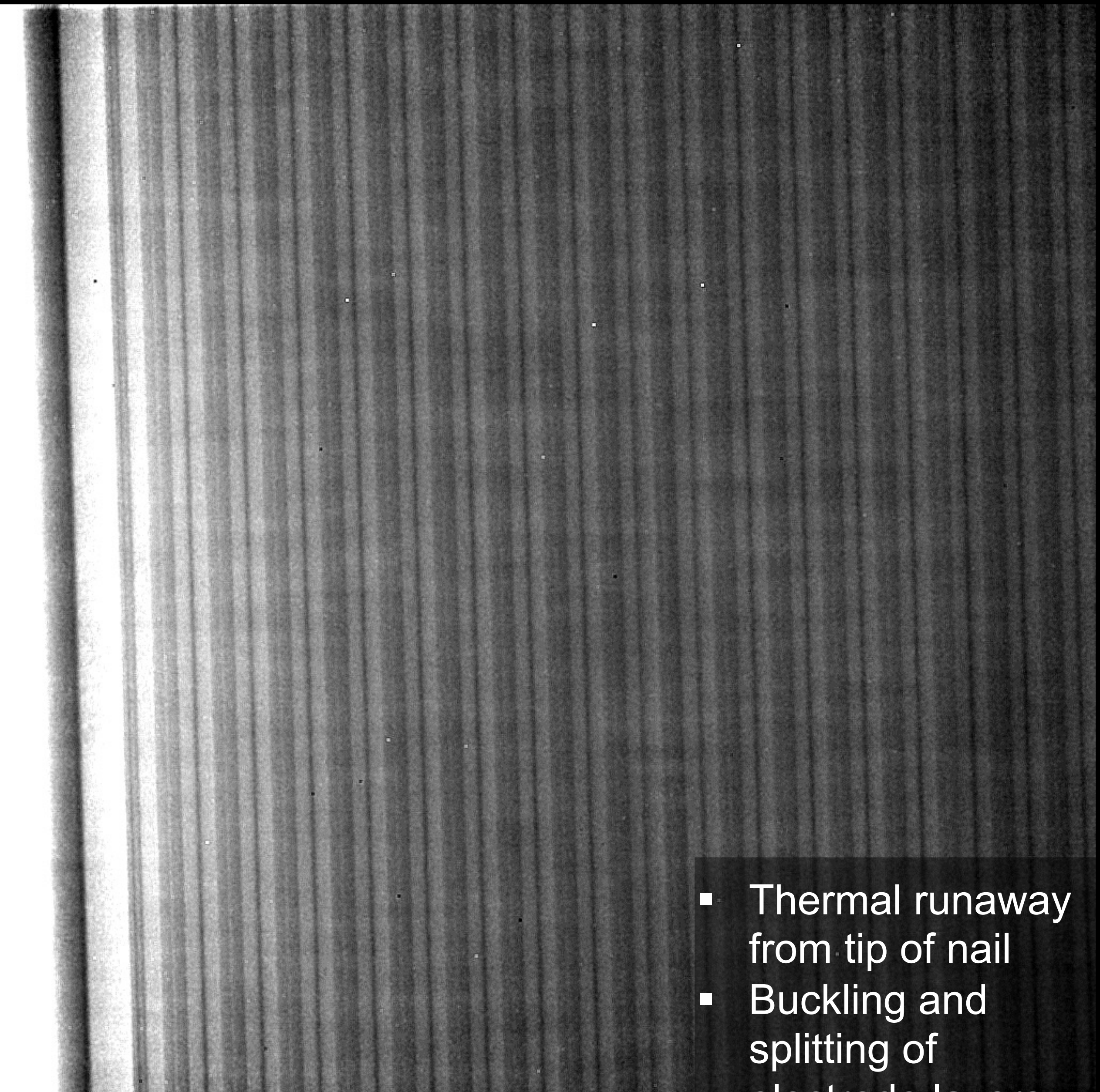
Tolerance demonstrated with near zero degradation of OCV!!!

Radiography at 3000 fps of 18650 cells

Dense material is dark (nail, can, NMC)

Control cell

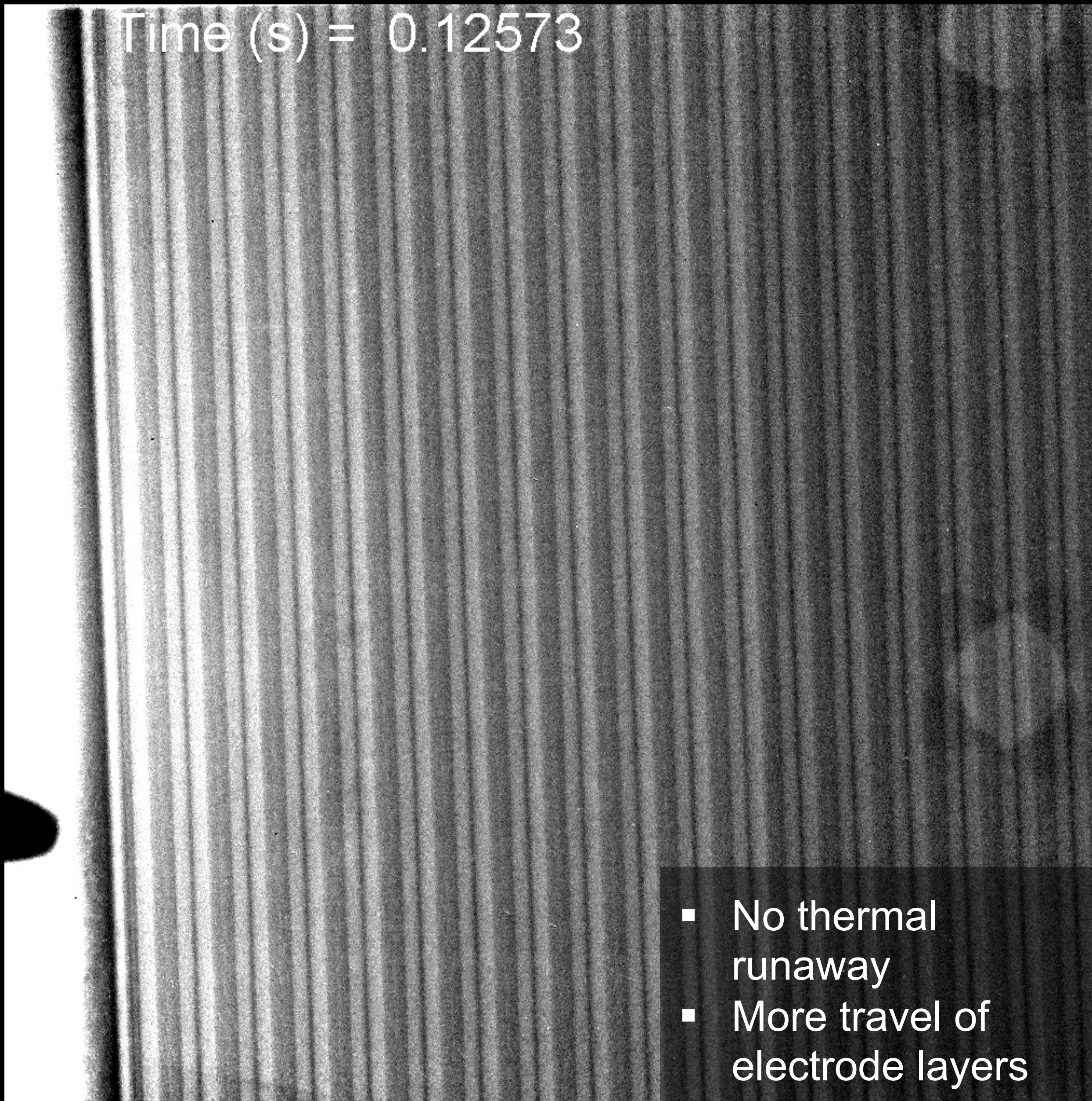
Run 034



- Thermal runaway from tip of nail
- Buckling and splitting of

Cell with PCC

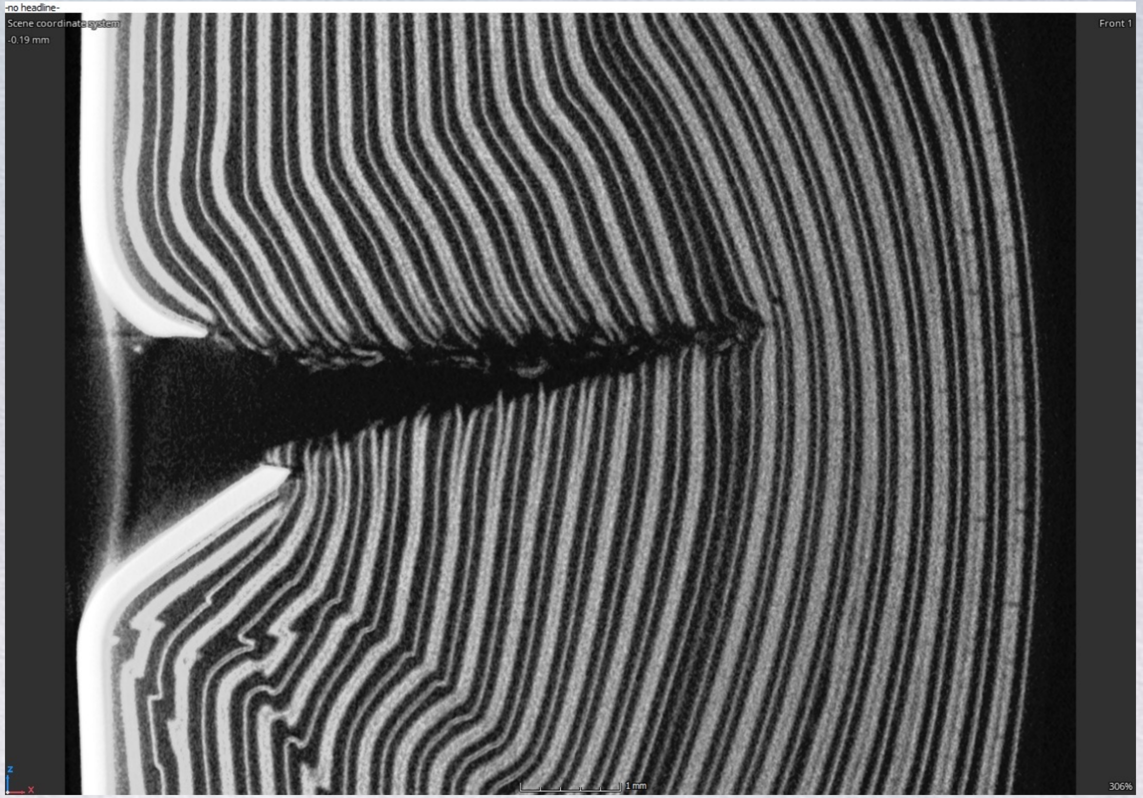
Run 031



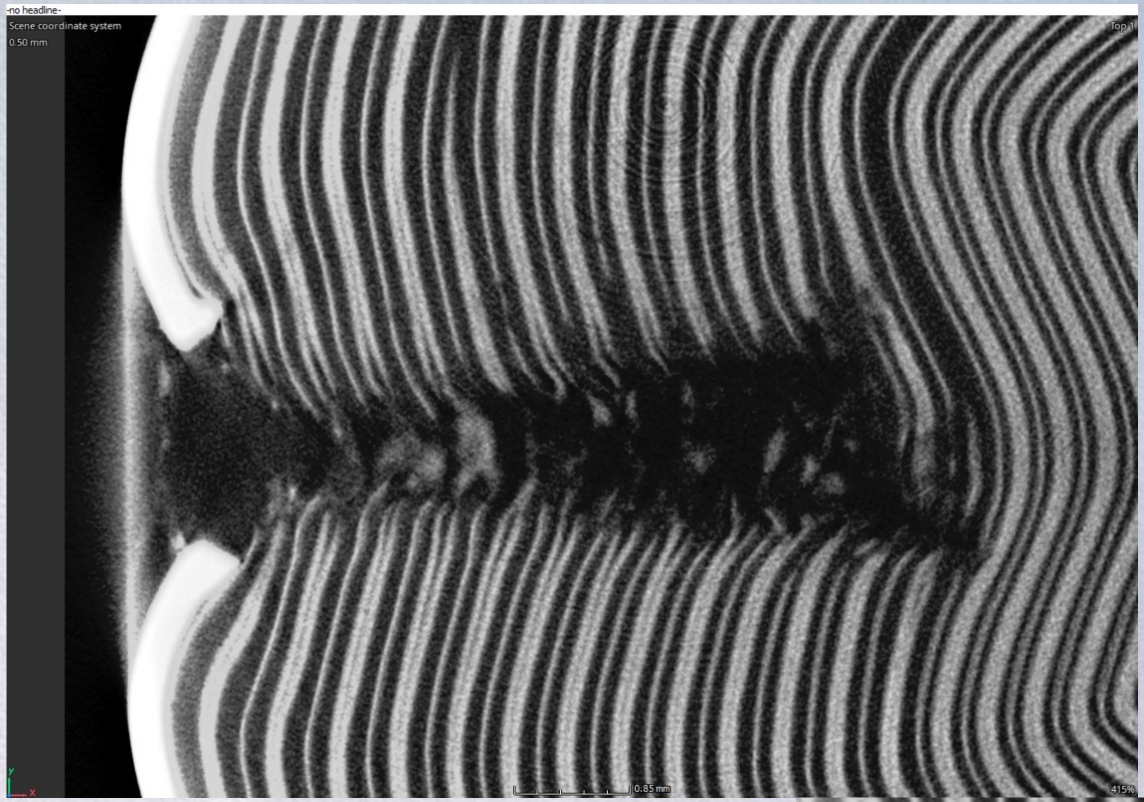
- No thermal runaway
- More travel of electrode layers

CT Images of BAK 18650 with Cathode PCC

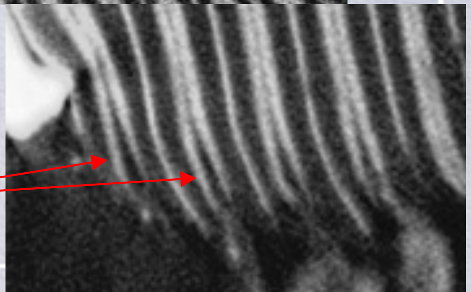
Axial view of nail penetration zone



Radial view of nail penetration zone

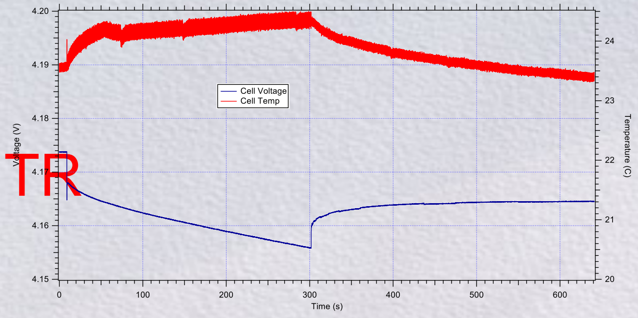


Reversing the image brightness from the video: Bright is most dense material, cell can, NMC.
Al coated PCC for cathode is thin gray layer between NMC active material coatings.
Axial and radial view show cathode PCC is clearly missing at nail interface (split ends).



BAK 21700 4.6Ah Test Matrix and Results (Nail)

- Soteria metalized polyester (15)
  - PCC only on cathode (cell achieves 251 Wh/kg)
  - Cu foil on anode like all other features in control version
  - 14 of 15 cells tolerated nail penetration
    - No fire, sparks, venting, or TR in those 13
    - Muted TR in 1 cell, generating ~50% kJ of control average
- Control cells (8)
  - Al and Cu foil CCs
  - All 8 cells went into TR



$\Delta t_{max} < 1°C$
$\Delta v_{max} < 19mV$

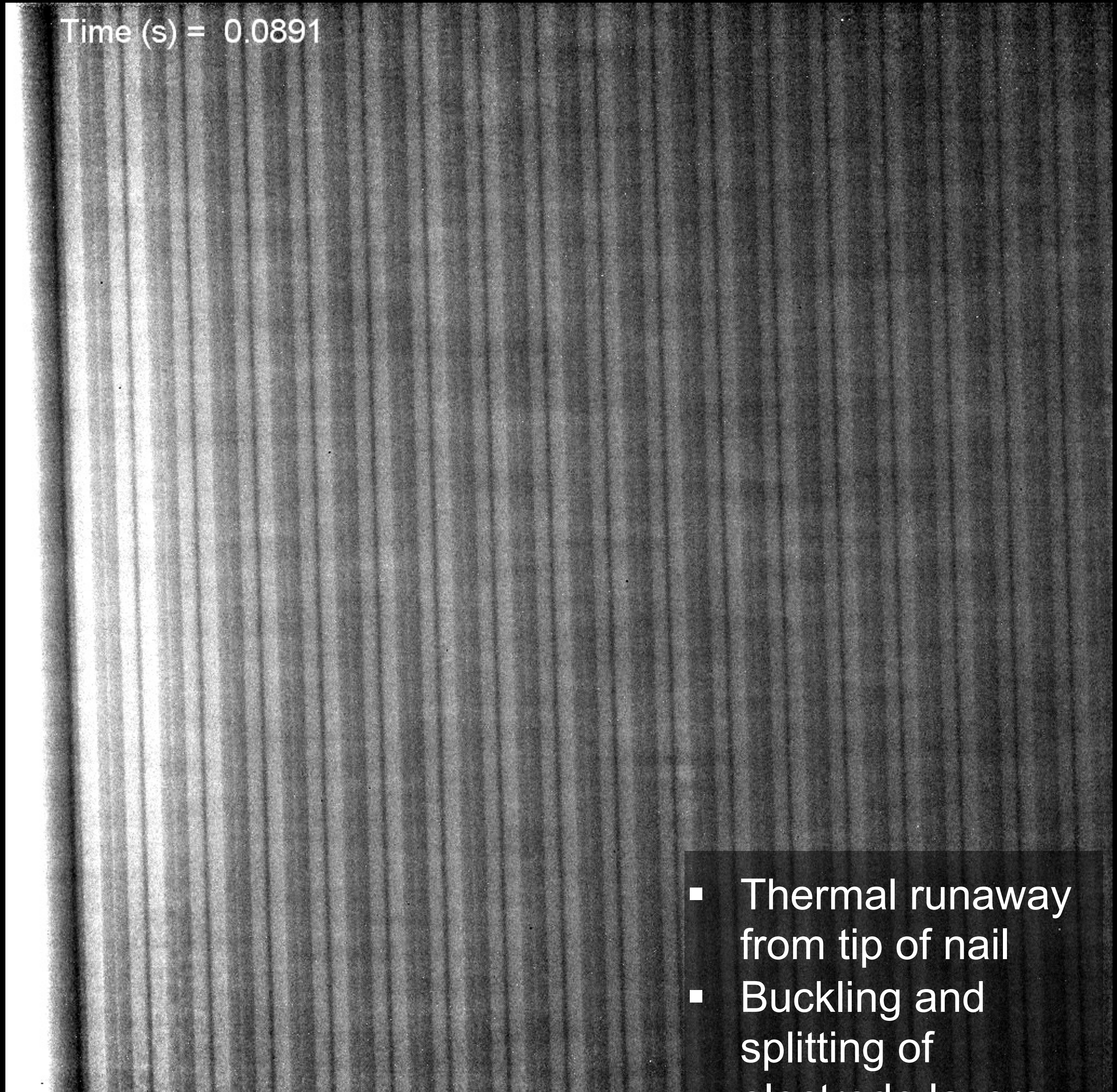
Tolerance demonstrated with near zero degradation of OCV!!!

Radiography at 3000 fps of 21700 cells

Dense material is dark (nail, can, NMC)

Control cell

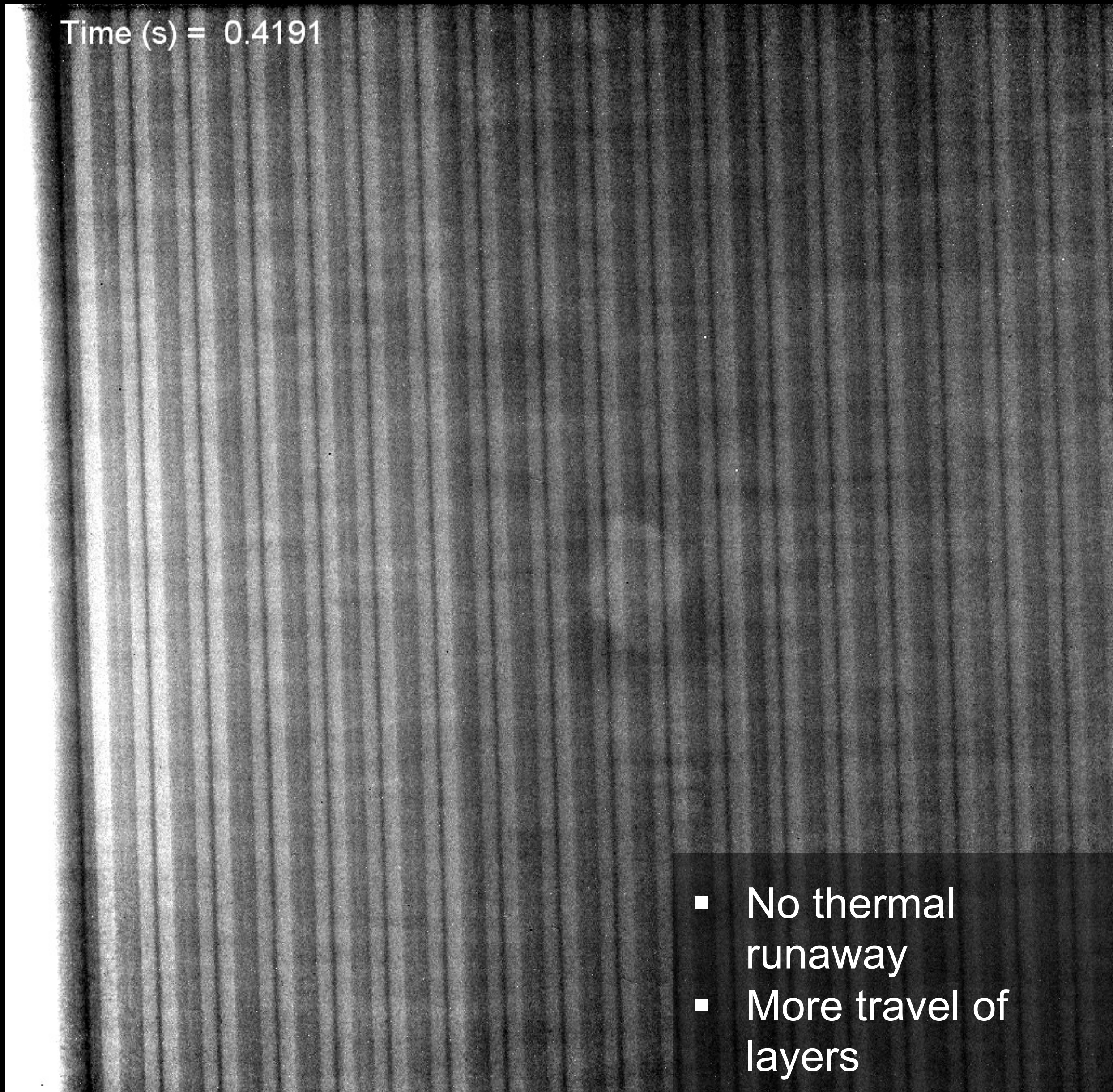
Run 025



- Thermal runaway from tip of nail
- Buckling and splitting of

Cell with PCC

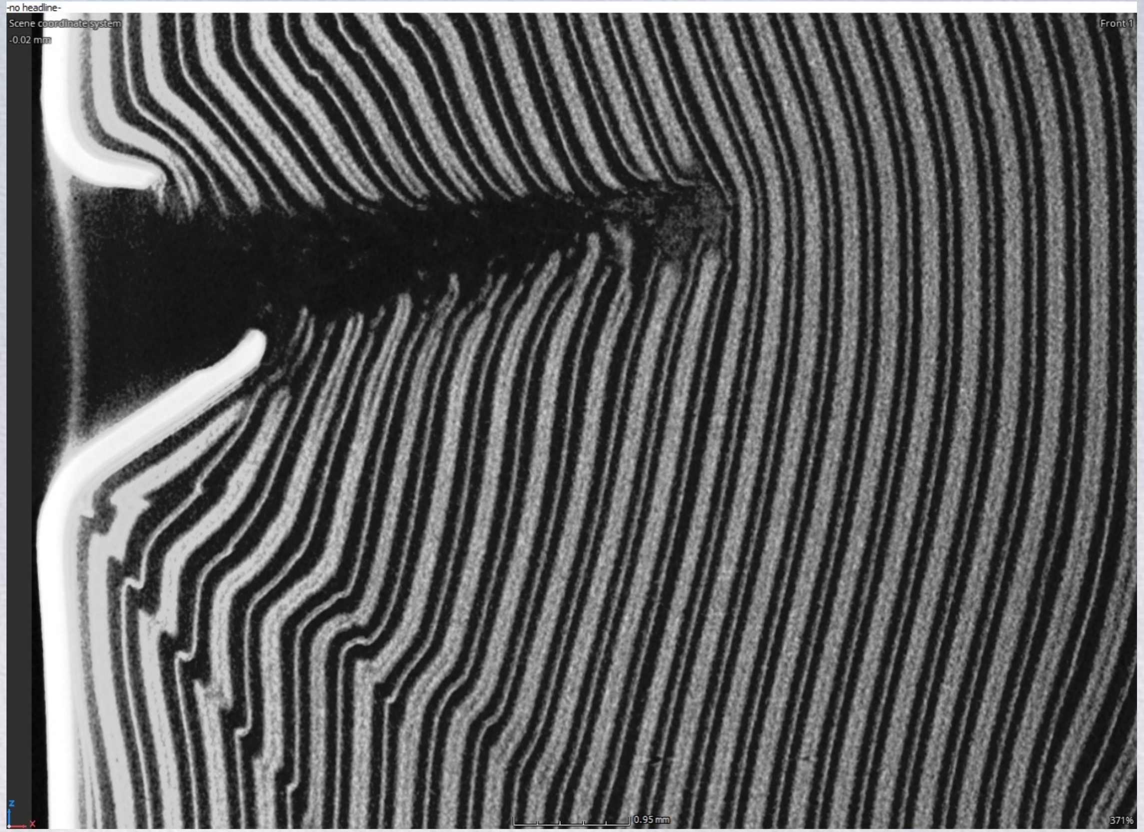
Run 020



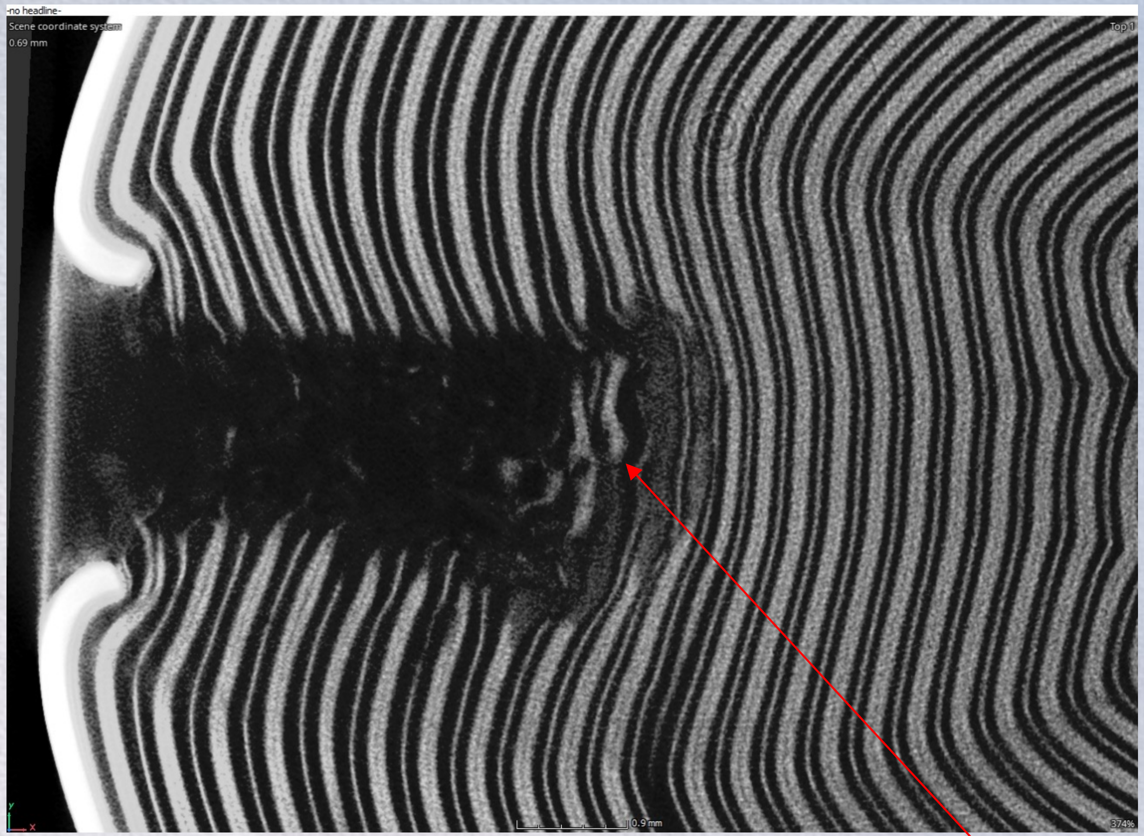
- No thermal runaway
- More travel of layers

CT Images of BAK 21700 with Cathode PCC

Axial view of nail penetration zone



Radial view of nail penetration zone

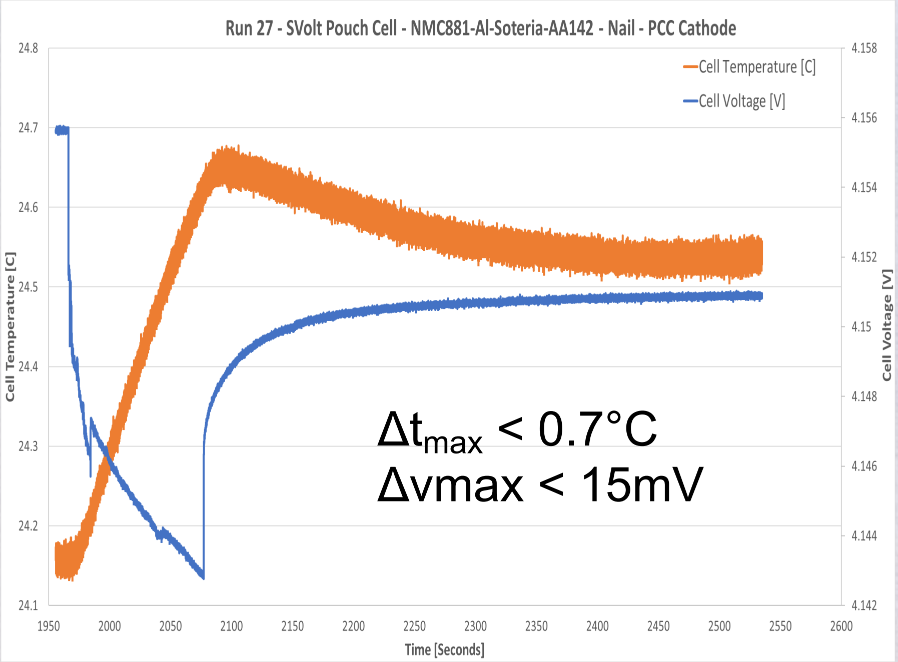


Stranded NMC

Reversing the image brightness from the video: Bright is most dense material, cell can, NMC
Al coated PCC for cathode is thin gray layer between NMC active material coatings.
Axial view shows cathode PCC is clearly missing at nail interface (split ends).

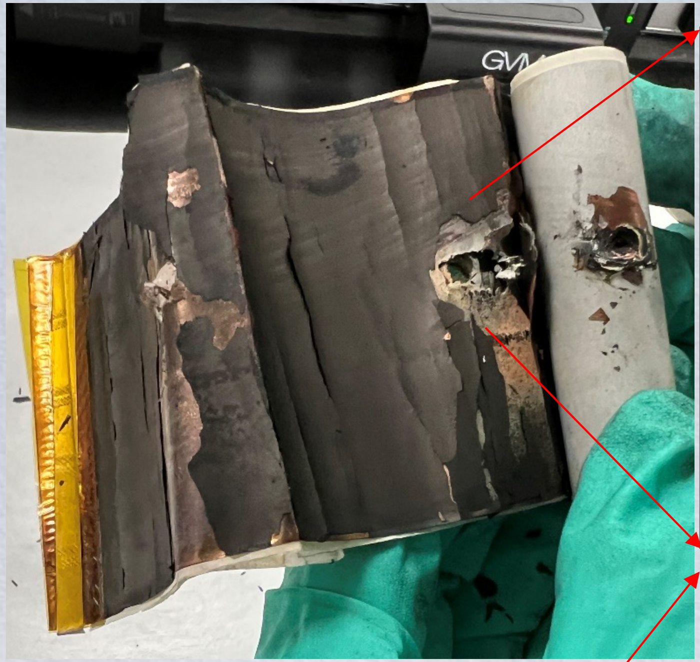
Svolt 10Ah Test Matrix and Results (Nail)

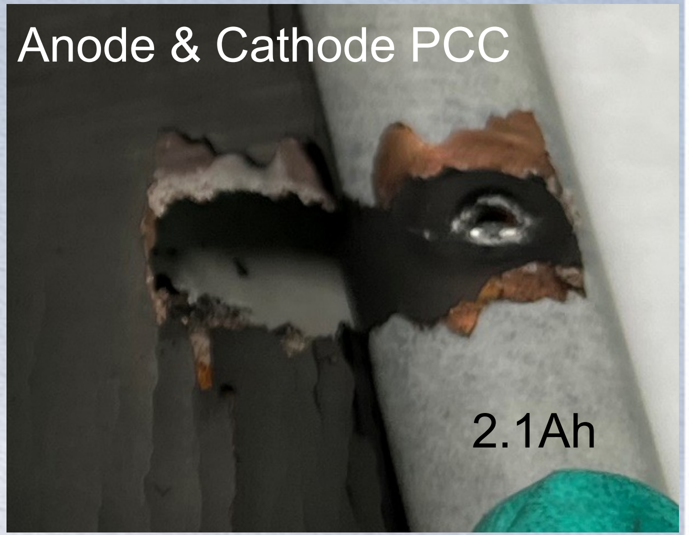
- Soteria metalized polyester (9)
  - PCC only on cathode (Cell achieves 243 Wh/kg)
  - Cu foil on anode like all other features in control version
  - All 9 cells tolerated nail penetration
    - No fire, sparks, venting, or TR
- Control cells (7)
  - Al and Cu foil CCs
  - All 7 cells went into TR

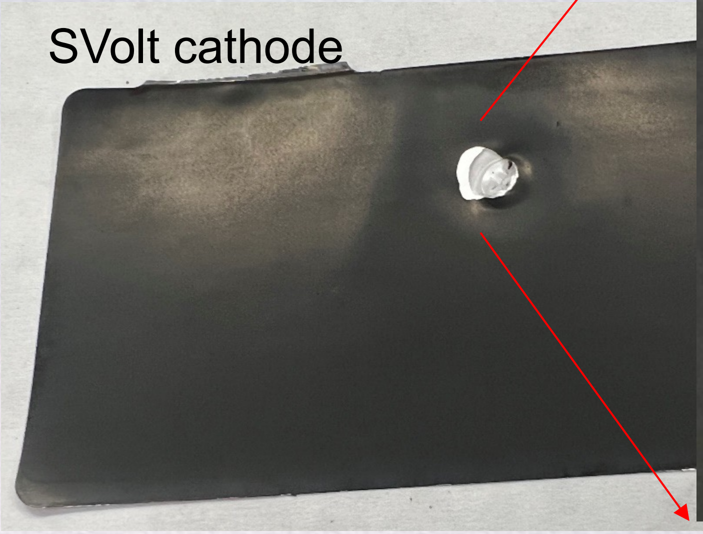


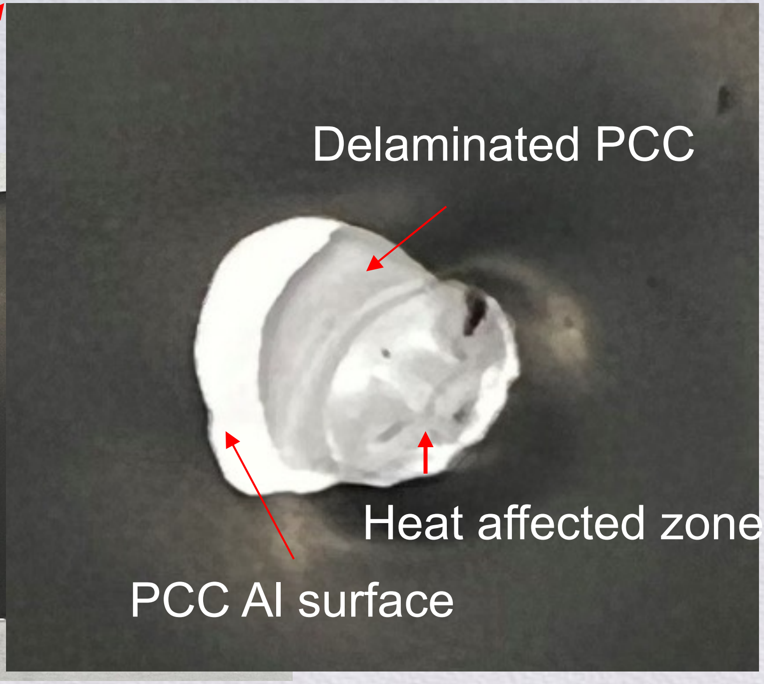
Only 14mV dip and 0.7°C rise during 2min nail penetration!!!

DPA Reveals Thermal Effect in PCC Response



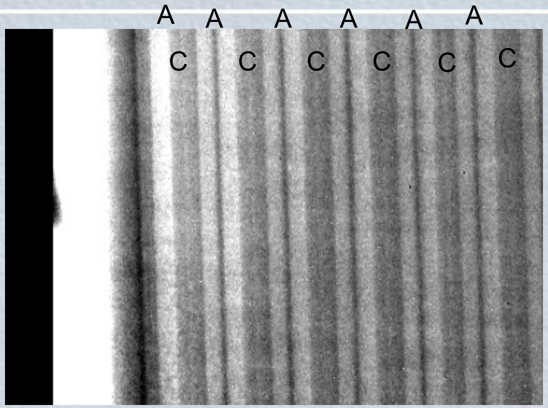






- Difficult unwinding due to melting of polyester CC and polyolefin separator ending glued together at nail interface
- Nail hole reveals thermally stressed PCC

High density material is dark in this negative X-ray



A Anode with dark Cu foil

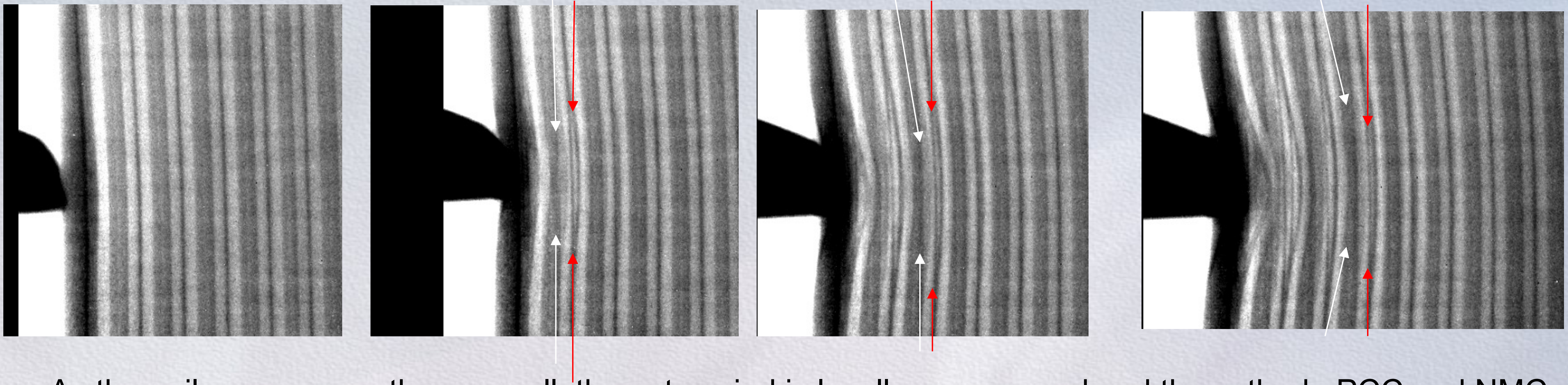
C Cathode with very light Al stripe from PCC film

Each snapshot is successive (~300 fps or 3.3 ms increments)

1st PCC local impact
Adj Cu foil bulges

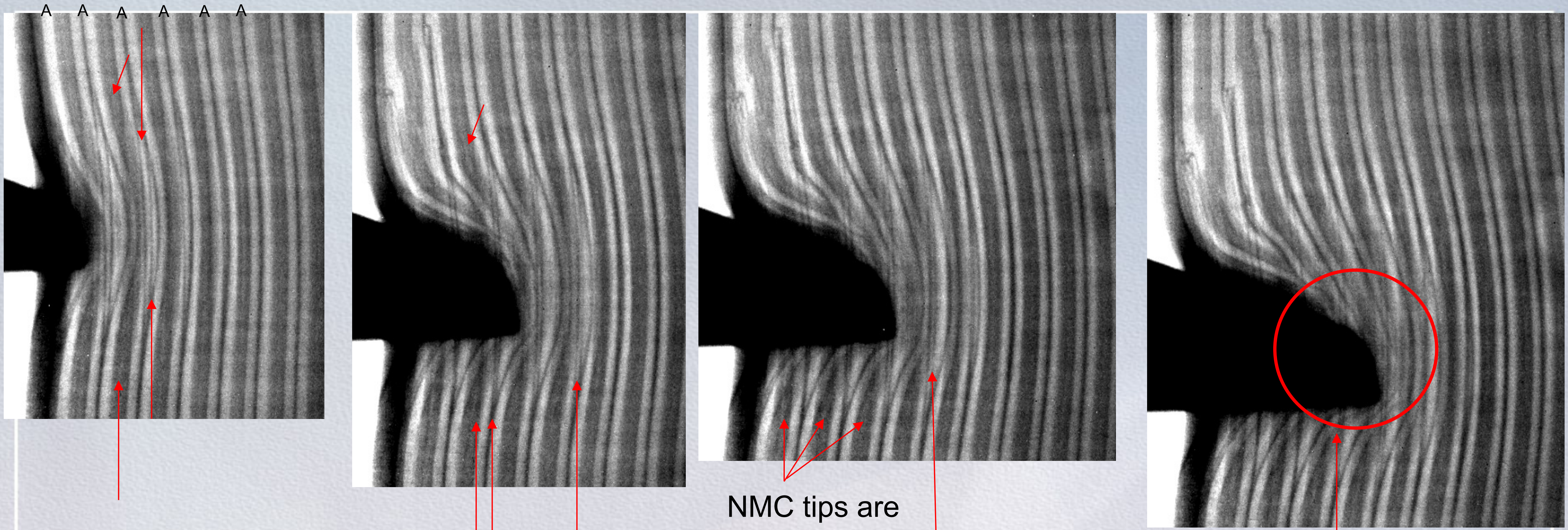
2nd PCC darkens
Next Cu foil bulges

3rd PCC darkens
Next Cu foil bulges



As the nail compresses the can wall, the outer wind is locally compressed and the cathode PCC and NMC darkens (densifies) with light Al stripe absent while gap forms “inside” adjacent Cu foil. In the next frames, that 1st PCC area loses density. The pattern repeats for the 2nd and successive winds.

We’re seeing the nail push NMC through the separator and into the graphite causing localized internal short circuit (ISC) hot spot. This causes Al PCC to vaporize as the electrode densifies and then looses density along with adjacent graphite.



As the 2nd PCC wind is crushed, gaps open at adjacent anode

Nail pushes wedge of graphite down into anode gap

Graphite gains density due to crushing

NMC tips are isolated from shorting

Graphite in front of nail turns gray as layers are compressed

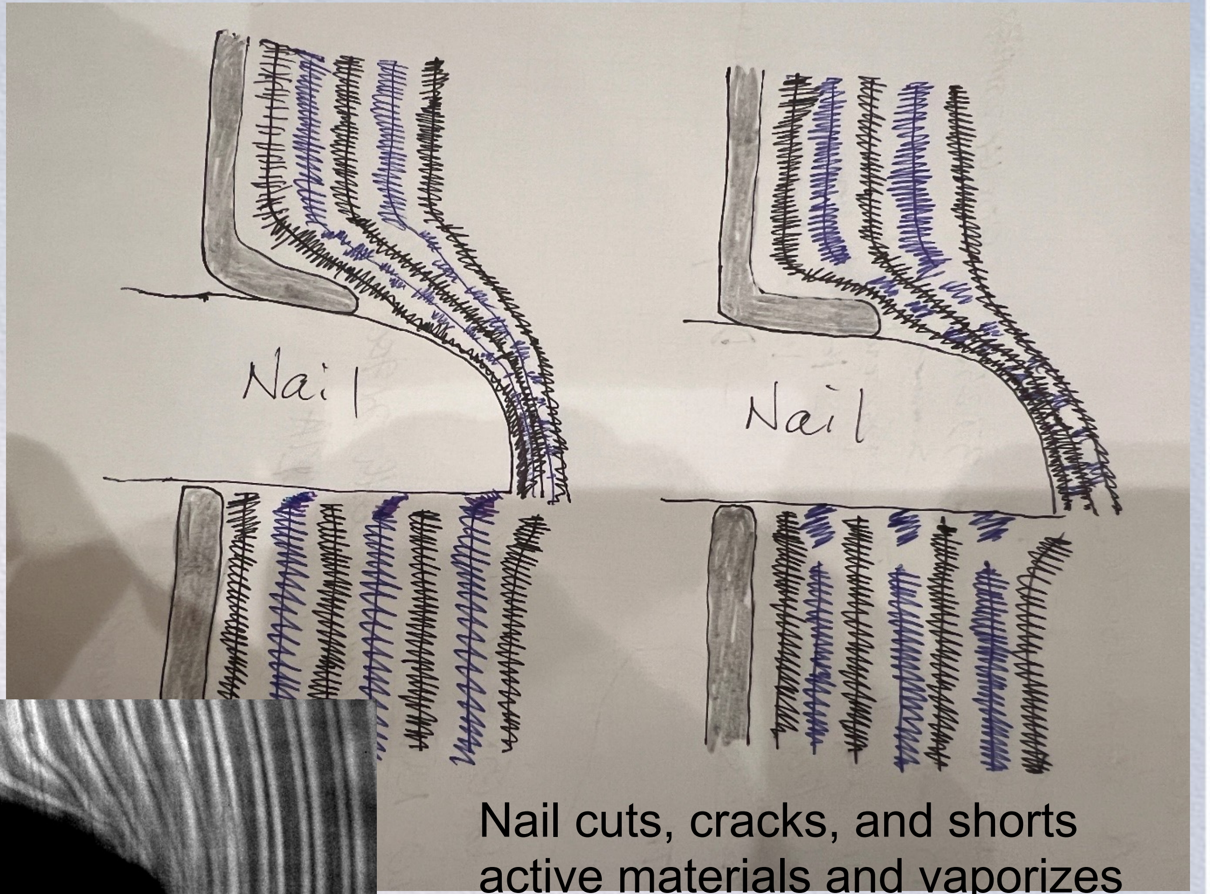
NMC/graphite at nail interface appear fluidized, possibly detaching from JR

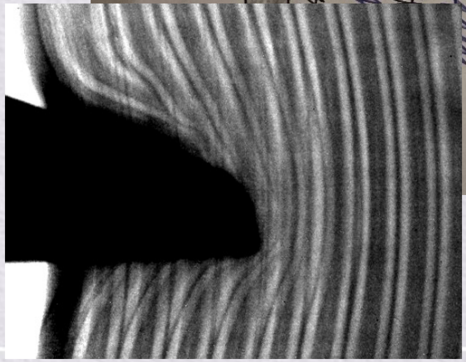
Probable PCC Mechanisms for Isolating Shorts

- Nail front crushes outer winds into inner winds
  - Causes anode/cathode contact
  - Active materials at nail interface discharge, get hot, fluidize, and generate a bit of gases
  - Vaporizing Al PCC near shorted materials
  - Active materials involved in short are electrically stranded from rest of JR
- Nail edge stretches and cuts the polymer film
  - NMC delaminates from stretched and vaporized PCC at nail interface
  - NMC material at nail interface is sheared from JR NMC and stranded

Stretched PCC cracks NMC coating

Heat of crushed/shorted active materials vaporizes the PCC





Nail cuts, cracks, and shorts active materials and vaporizes PCC which strands NMC

Highest Energy 21700 Cell Build

Metallized plastic current collectors

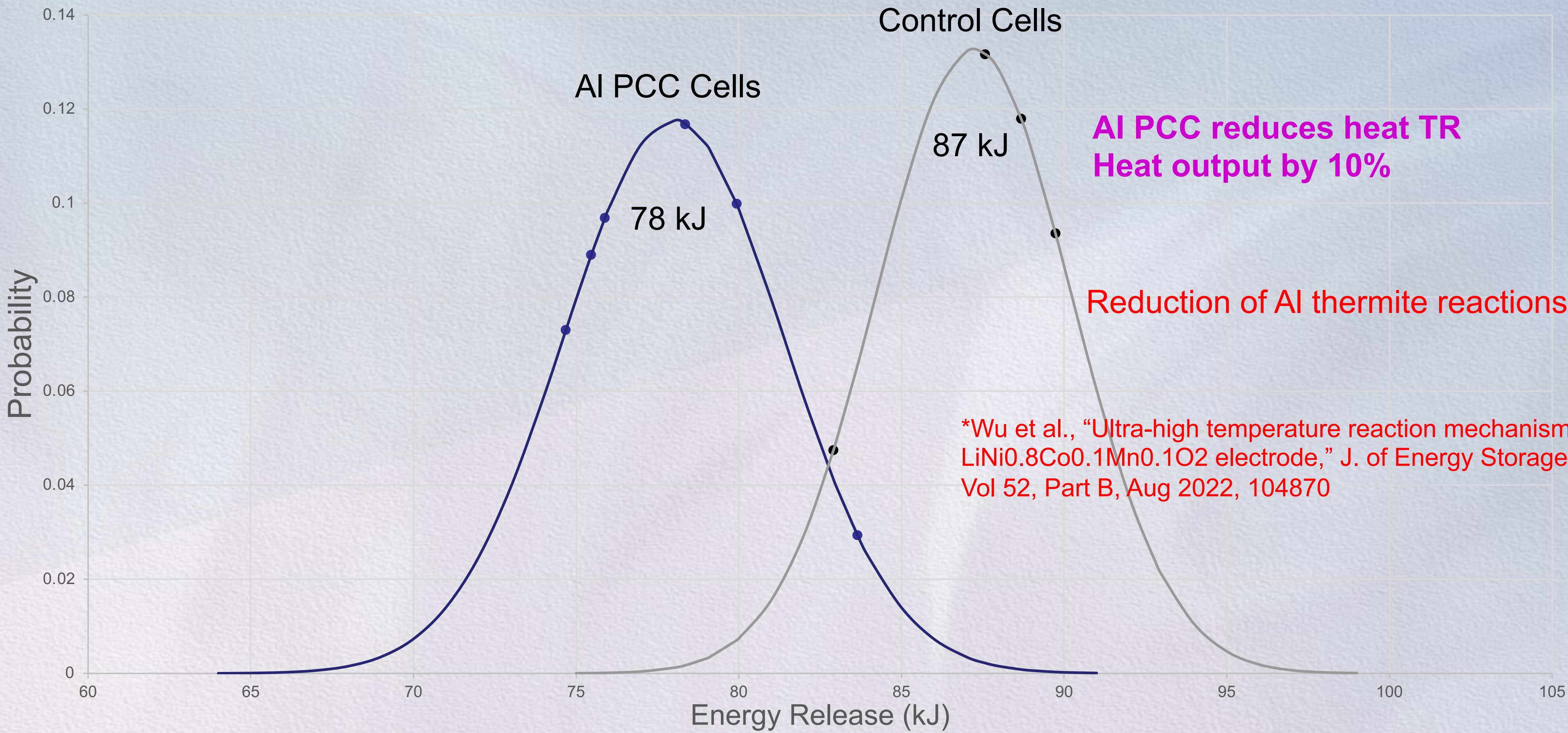
- 10 μm polyester films coated with Al (0.5 to 1.0 micron) from Soteria
  - Graphite/NMC 811
  - Polypropylene separator (plain simple)
  - ACR (~50 mΩ) with Al PCC
  - ACR of control (~24 mΩ) with Al foil
  - Built in 2023
- Nail penetration Test Results
  - 24 of 27 Al PCC cells driven into TR
    - 15 runs performed at 100% SoC
    - 3 runs each at 90 and 80% SoC
    - 6 runs at 70% SoC (3 tolerated nail)
      - Corresponding to 246, 219, and 191 Wh/kg
  - All 4 control cells driven into TR at 100%

Parameter	2302-N41
Cap, Ah	5.192 ± 0.033
Nominal V	3.6
Mass, g	68.35
Wh/kg	273.5
Dia, mm	21.5
Length, mm	71.1
Vol, (L)	0.0257
Wh/L	723.7

21.5mm is fatter than 21.2 mm “norm” for COTS 21700 cells

5.1Ah Coulometrics 21700 TR Heat Output

Nail Penetration Fractional TR Calorimetry at 100% SoC



Al PCC reduces heat TR Heat output by 10%

Reduction of Al thermite reactions?*

*Wu et al., "Ultra-high temperature reaction mechanism of LiNi0.8Co0.1Mn0.1O2 electrode," J. of Energy Storage, Vol 52, Part B, Aug 2022, 104870

Other Design Features We Can Add

- Thermally Stable Nonwoven Separator
  - Plain polypropylene separator could be shrinking near nail and inducing electrode shorts
  - Cellulose nanofibers and aramid fibers (Kevlar or Twaron) that are stable up to 500°C and don’t shrink due to heat exposure
  - Commercialized as Dreamweaver by Soteria BIG
- Cu Metallized PET Current Collector
  - Very similar to the Al PET PCC

More High Energy Cell Builds with Al PET PCC

Design Parameter	2302-N41 21700 Coulometrics	2306-N43 21700 Coulometrics	2306-N45 21700 Coulometrics	INR21700 BAK	Pouch Cell SVolt
+ Collector	Al PET (Soteria)	Al PET (Soteria)	Al PET (Soteria)	Al PET (Soteria)	Al PET (Soteria)
+ Active Material	NMC811	NMC811	NMC811	NMC811 (TBV)	NMC811
Separator	Polyolefin	DW Cellulose	DW Cellulose	Polyolefin	Polyolefin
- Active Material	Graphite	Graphite	Graphite	Graphite, Si	Graphite, Si
- Collector	Cu foil	Cu foil	Cu PET (Soteria)	Cu foil	Cu foil
Capacity, Ah	5.192 ± 0.033	4.97 ± 0.10	4.94 ± 0.02	4.6	10
ACR, mΩ	52.4 ± 7.1	41.3 ± 2.1	93.1 ± 4.8	42	
Mass, g	68.584 ± 0.222	67.830 ± 0.099	63.606 ± 0.253	66	149.46
Wh/kg	272.6	263.8	279.6	251	242.8
Diameter, mm	21.50	21.50	21.50	21.20	N/A
Nail Penetration	TR ≥ 70% SoC 50% success @70% (6 cells)	TR ≥ 70% SoC (15 cells)	TR ≥ 70% SoC (15 cells)	Tolerance at 100% SoC (14 out of 15)	Tolerance at 100% SoC (9 of 9)

Adding cellulose separator and Cu PCC for anode didn't improve safety of 5Ah Coulometric 21700 design!

Potential Root Causes for Nail TR

- Wh/kg, Wh/L limit reached for PCCs?
- No: Lower SOC tests reach TR with equivalent of 191 Wh/kg

- Need more thermally stable separator?
- No: Replacing sep with 500°C cellulose (Dreamweaver) results in TR at 100, 90, 80, and 70% SoC

- Need to add (-) Cu coated PCC?
- No: Replacing Cu foil with Cu coated PCC results in TR at 100, 90, 80, & 70% SoC

- Cathode active material adhesion needs to be poor?
- Possible: BAK cells have poor cathode adhesion

- Winding tension is too high?
- Possible: Since 20Ah prismatic pouch cells tolerate nail with same electrode design as in 5Ah 21700

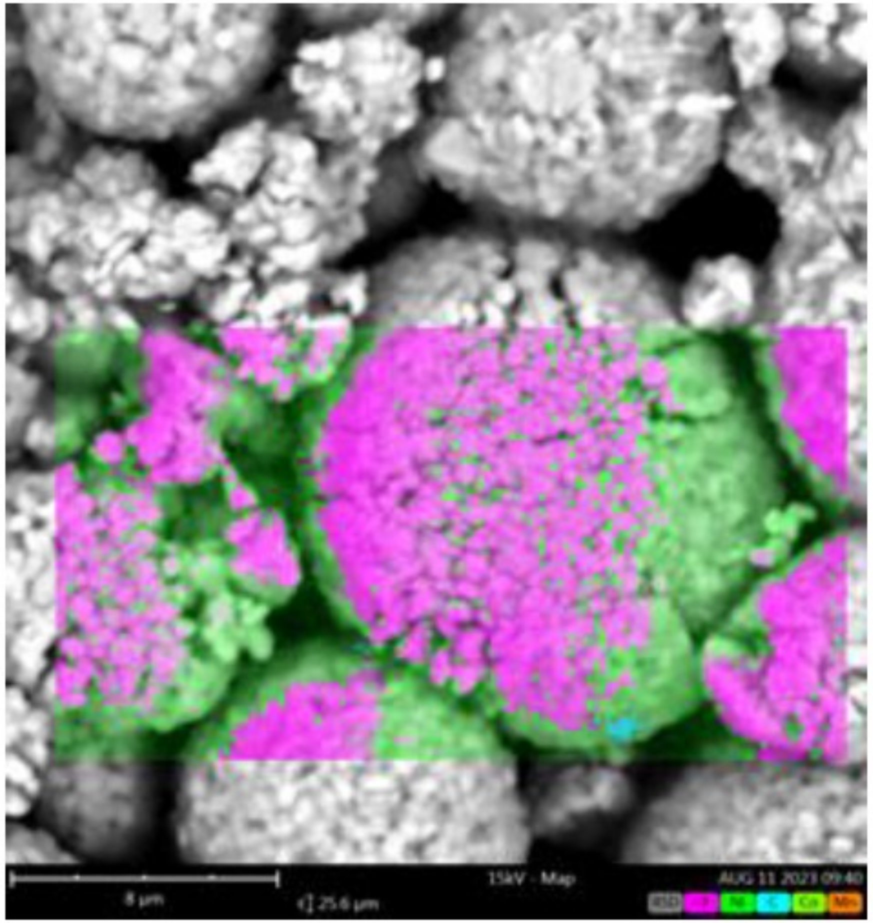
Cathode Active Material Adhesion to Collector

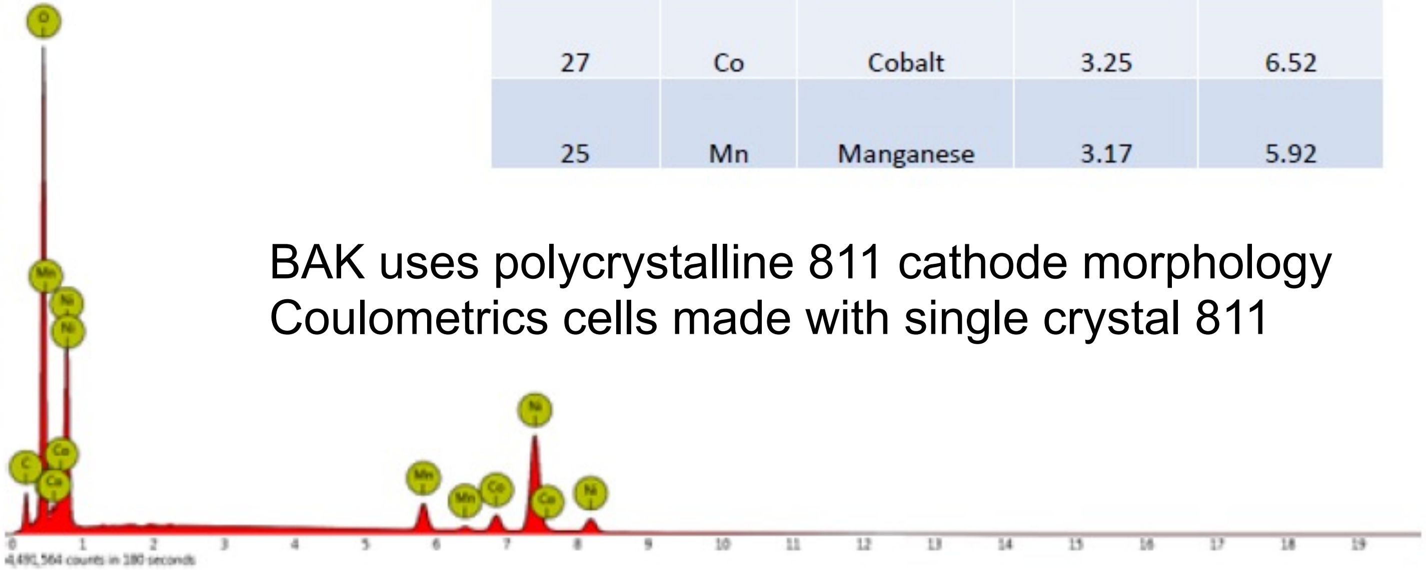
- DPA of BAK 21700 revealed poor adhesion of cathode active material to Al coated PCC
- Slight wrinkling of the cathode cause lots of delamination

BAK Cell Teardown
EDS – Cathode Sample 1

Coulometrics

Element Number	Element Symbol	Element Name	Atomic Conc.	Weight Conc.
8	O	Oxygen	51.06	27.77
28	Ni	Nickel	26.74	53.35
6	C	Carbon	15.78	6.44
27	Co	Cobalt	3.25	6.52
25	Mn	Manganese	3.17	5.92





BAK uses polycrystalline 811 cathode morphology
Coulometrics cells made with single crystal 811

Summary of our 2022-2023 Effort

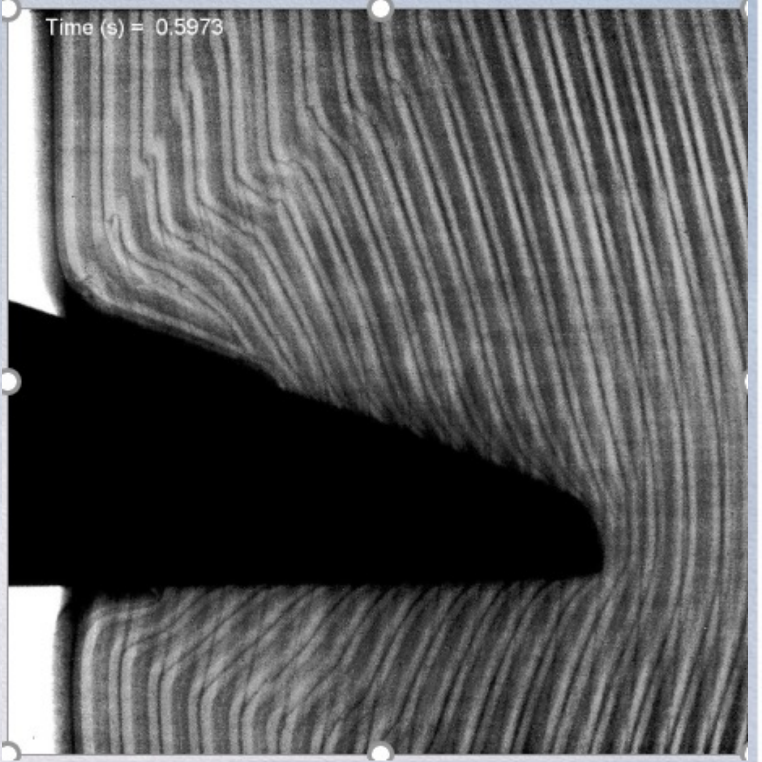
Soteria polyester PCC is reliable in tolerating nail penetration in 64 out of 65 Li-ion cells with <251 Wh/kg

- 2.1Ah Coulometric 18650s (33 for 33) up to 193 Wh/kg
- 2.9Ah BAK 18650s (9 for 9) achieving 233 Wh/kg
- 4.6Ah BAK 21700s (13 for 14) achieving 250 Wh/kg
- 10Ah SVolt Pouch Cells (9 for 9) achieving 243 Wh/kg

Higher energy (>260 Wh/kg) 21700 cell designs fail nail penetration every time so far

- 5.165Ah Coulometrics (3 for 24, even at 70% SoC, 191 Wh/kg)
  - 3 for 6 at nail tolerance at 70% SoC, 0 for 18 at ≥ 80% SoC
  - Al PCC reduces TR heat output vs Al foil cells by ~10%
- 4.97Ah Coulometrics with cellulose (DW) separator
  - 0 for 12, even at 70% SoC
- 4.94Ah Coulometrics with DW separator & Cu PCC
  - 0 for 12, even at 70% SoC

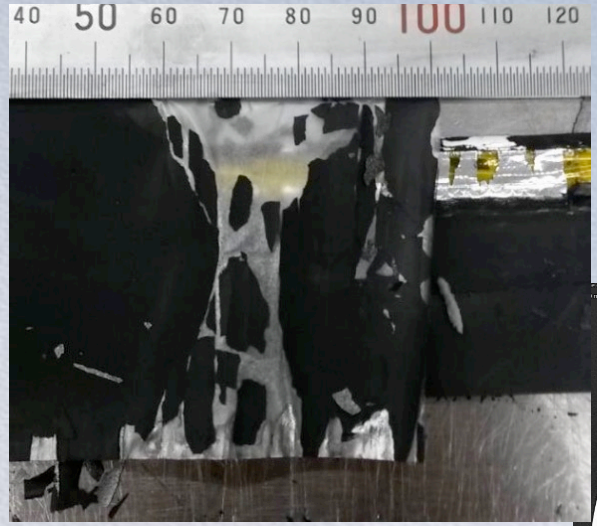
Nail Tolerance

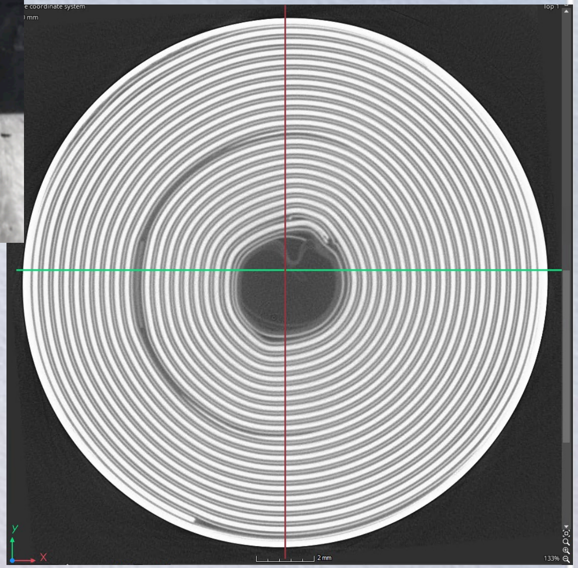


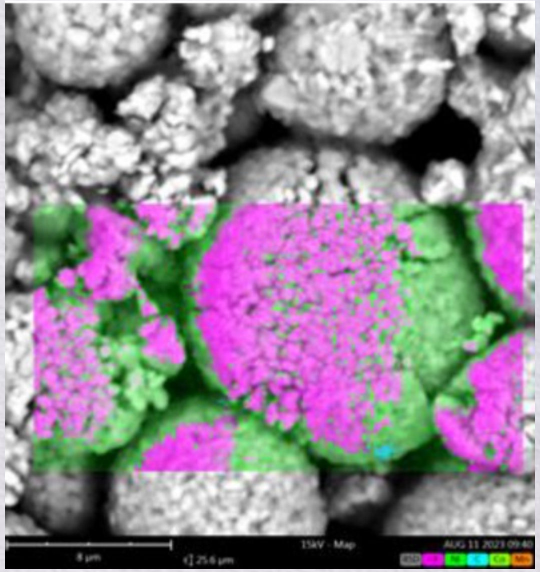
Our Investigation Continues

Planned new cells builds

- Lower adhesion of cathode active materials to collector
  - Reduce binder content in cathode mix
- Reduce winding tension by putting 4.97Ah JR in fatter can or shorter JR in same can
  - Is the tension of the cylindrical format too high?
- How important is the role of a thermal stable separator?
  - Meta Materials all-ceramic separator <1% shrinkage at 220°C and 5x higher thermal conductivity
- Single crystal vs polycrystalline 811?
- Perform nail penetration on fully discharge cells
  - Decouple the thermal from the mechanical delamination and cracking phenomena
- Any other suggestions?







BACK UP

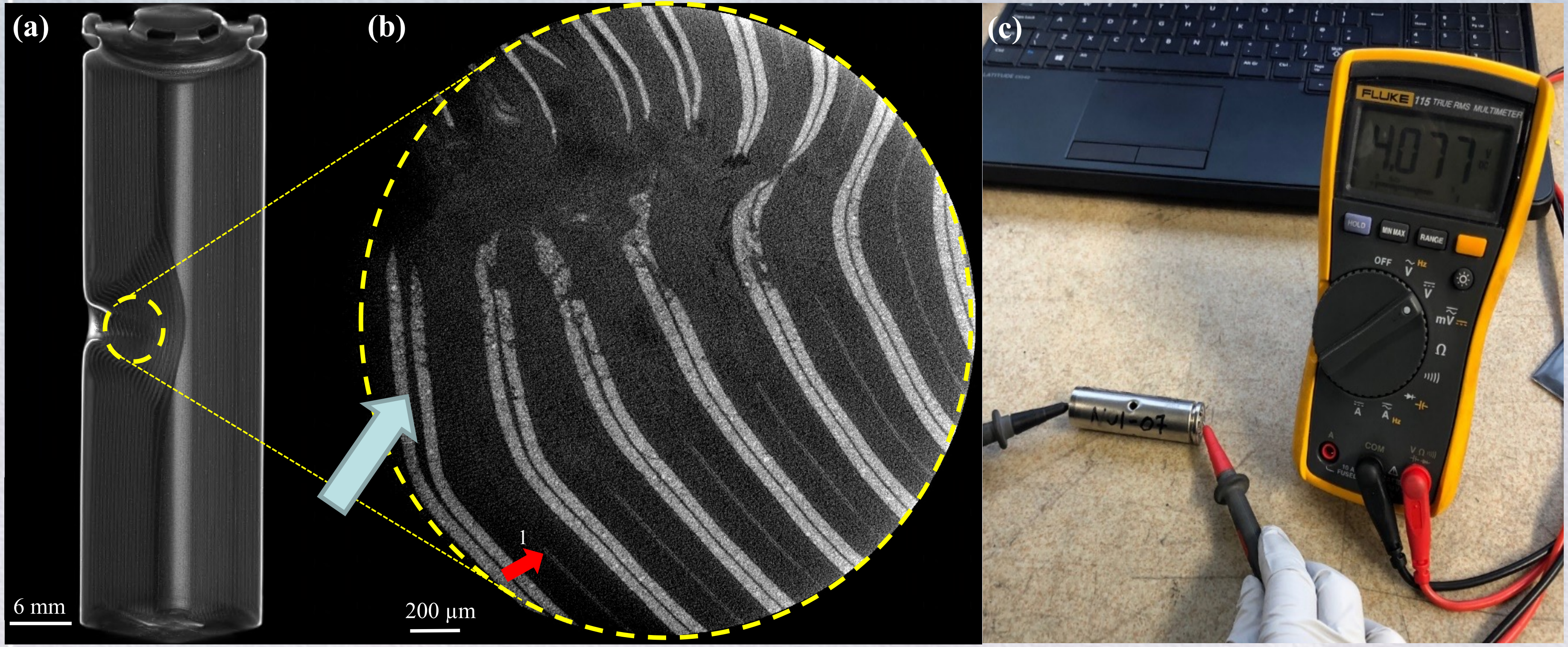
CT Scan of Nail Penetration

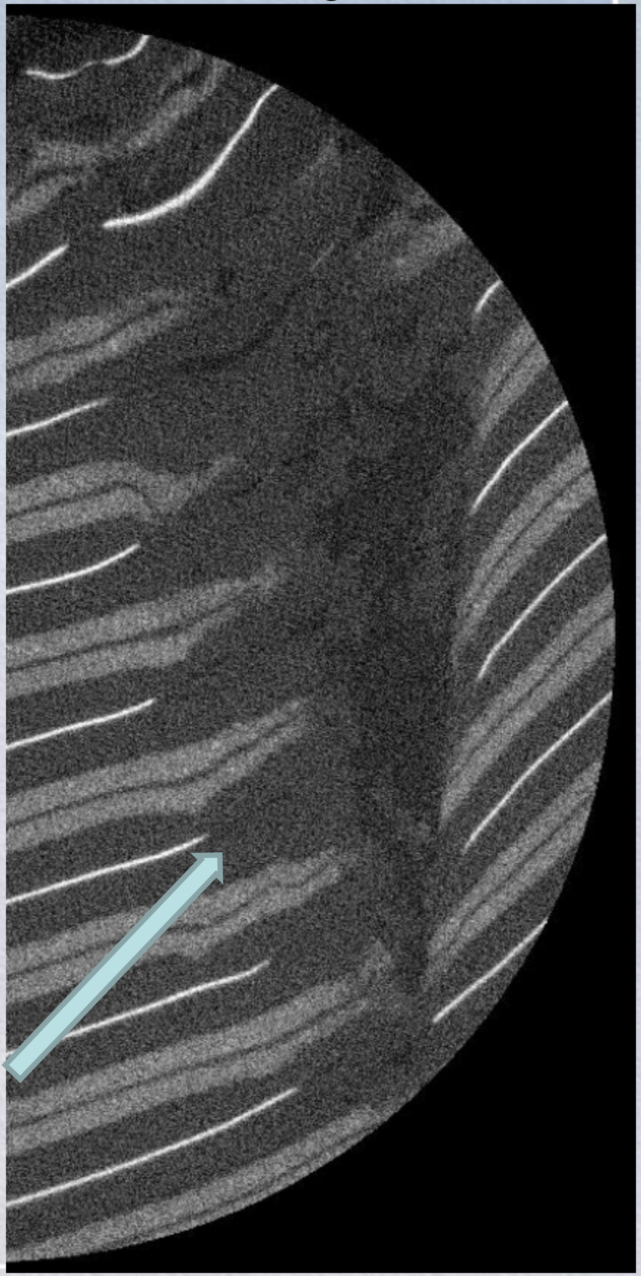
2.1Ah cells from 2020

Bright CC is Cu PCC coated with darker graphite
Cathode PCC is gray layer substrate for bright NMC

Pham et al., Cell Reports Physical Science 2, 100360, Mar 2021

PCC cell



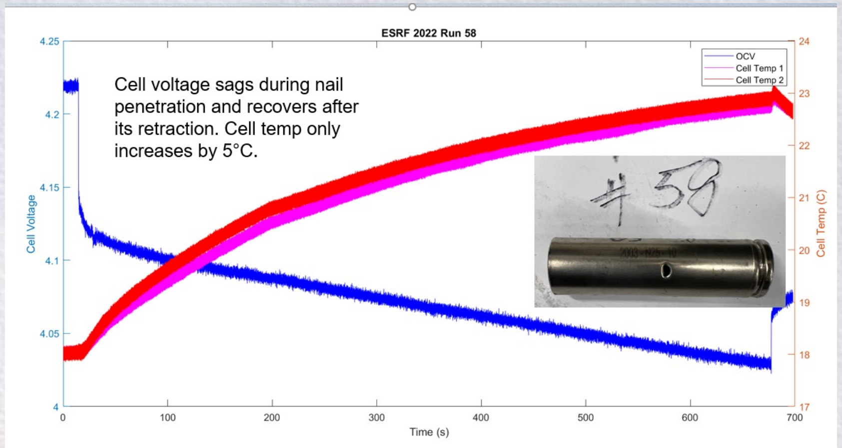


X-ray CT reveal Al and Cu PCCs withdrawn from the nail, thus reducing further short-circuiting. OCV measurement showed 4.07 V; cells retained voltage for over 10 months.

Coulometrics 2.1Ah Test Matrix (Nail)

- Soteria metalized polyester (PET)
  - 8 with cathode PCC (no TRs)
    - 189 Wh/kg
  - 8 with anode & cathode PCCs (no TRs)
    - 193 Wh/kg
  - 8 control cells with metal CCs (all TRs)
    - 184 Wh/kg

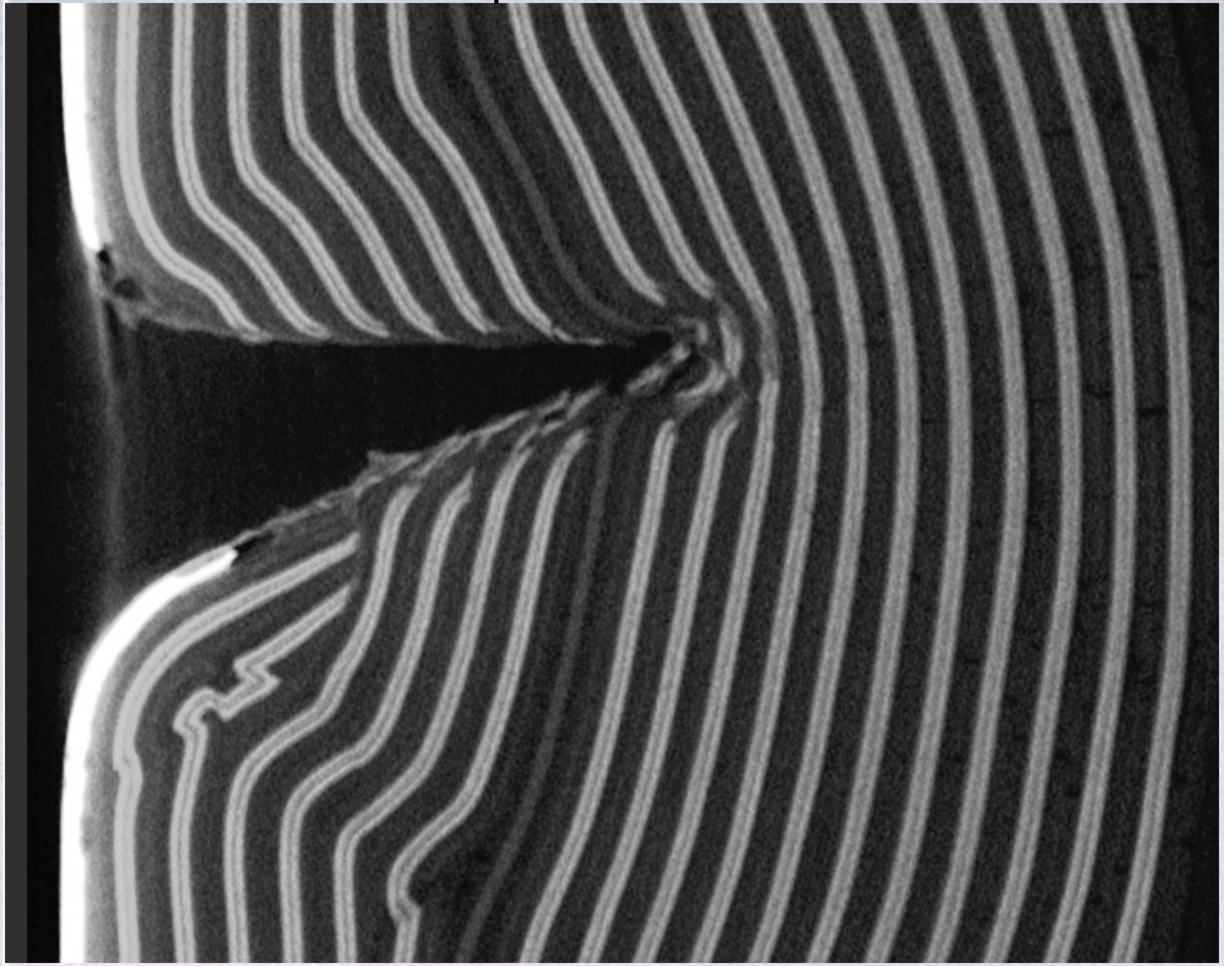
Tolerance demonstrated with small degradation of OCV



CT Images of Coulometrics 18650 with Polyester Anode & Cathode PCCs

Dense material appears bright (cell can, NMC)

Axial view of nail penetration zone



Radial view of nail penetration zone

PCC not present at nail interface, presumably vaporized