



Progress on new methods, materials, and insights for safety Li-ion battery systems

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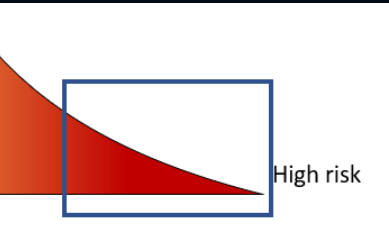
NASA: Eric Darcy, Will Walker, John Darst

UCL: Paul Shearing, Martin Pham, Tom Heenan

Contents

1. Testing and insights for safer battery systems
 - I. Understand what causes the spectrum of risks
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2. Acoustic diagnostics for detecting failure
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18650 cells

High-risk failure mechanisms



Bursting: Top



Bursting: Bottom



Breach: Top



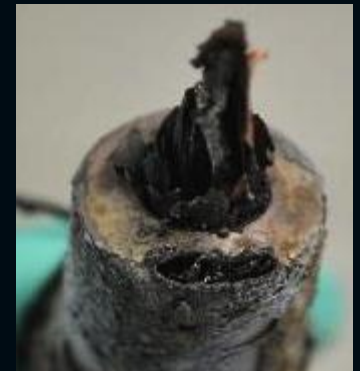
Breach: Top



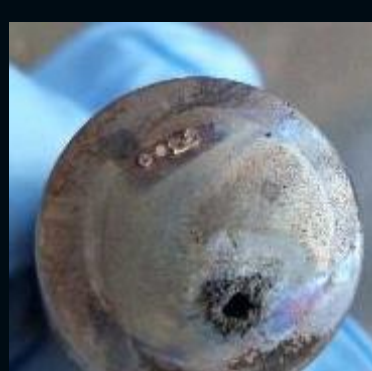
Breach: Side



Breach: Side



Breach: Bot



Breach: Bot

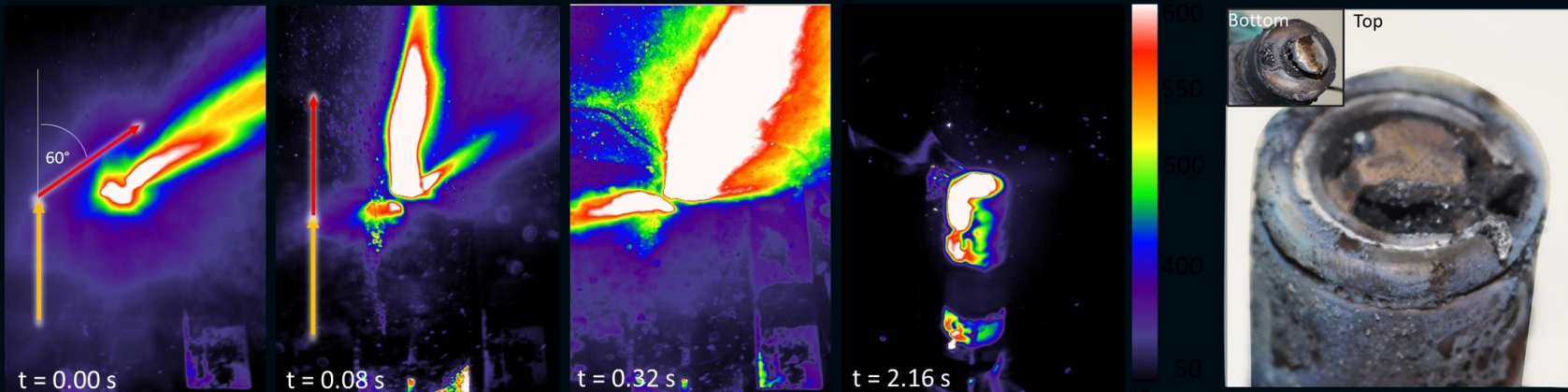
Hazardous flare stemming from breach



Image courtesy of E. Darcy (NASA)

Most challenging failure mechanism to handle

Characterizing breaching mechanism



Cell type: Li-ion 18650
Capacity: 3.4 Ah
State of Charge: 100 % (4.2 V)
Bottom vent: Yes
Wall thickness: 220 μm
Orientation of cell: Positive end up
Location of ISCD radially: 6 winds in
Location of ISCD longitudinally: Top
Side of ISCD in image: Right

Location of FOV longitudinally: Top
Frame rate: 2000 Hz
Frame dimension (Hor x Ver): 1822 x 1140 pixels
Pixel size: 10 μm

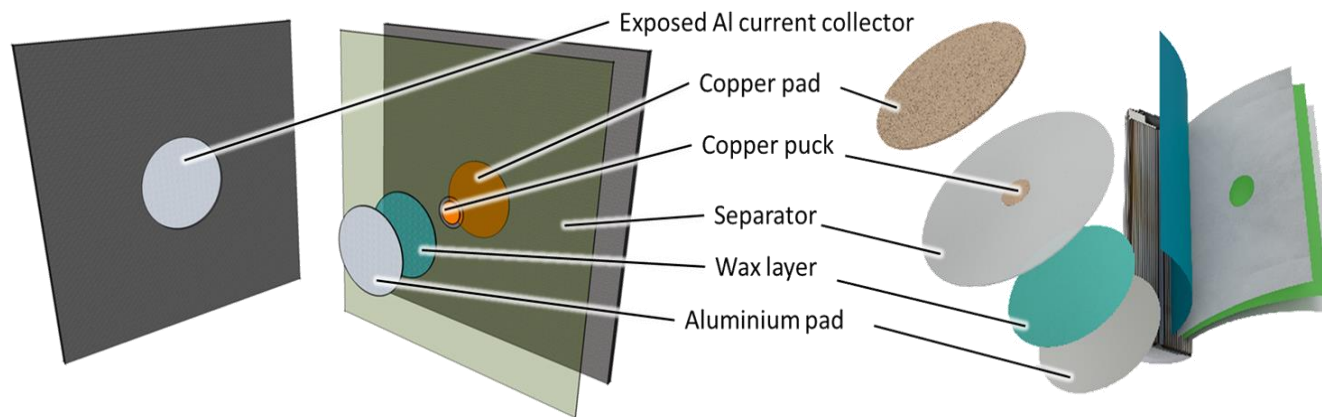
Cause of breach

- Reacting material fluidizes and flows towards the top vent
- Material deflects off the spin-groove, causing thermal stress
- The spin-groove melts leading to a breach and escape of hot material

Selective Positioning of the ISC Device

Internal short-circuiting device

- 18650 cells were manufactured with the ISC device placed at 3 different longitudinal locations



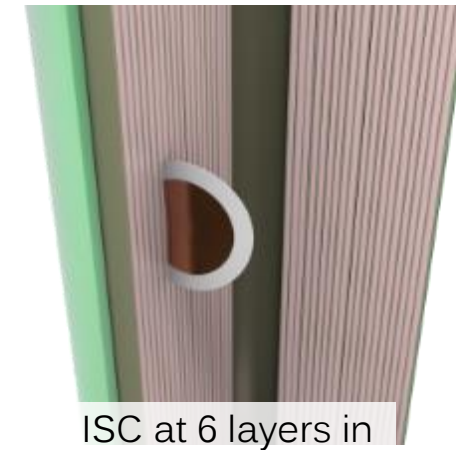
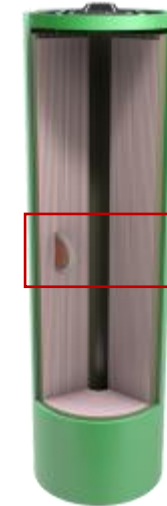
ISC at bottom



ISC at top

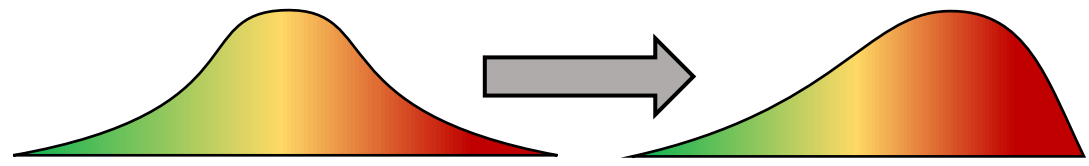


ISC at midway

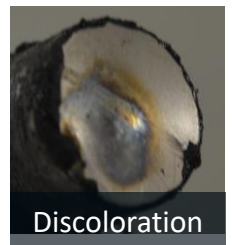
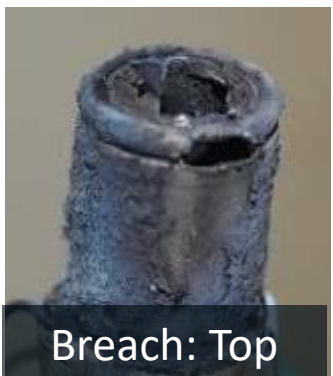
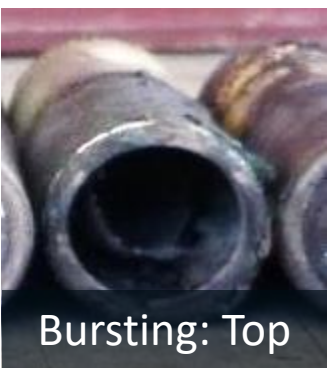


Risk map

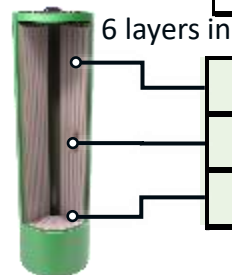
From a study of 200 cells, the propensity of cell to undergo certain failure mechanisms, under certain conditions, was mapped.



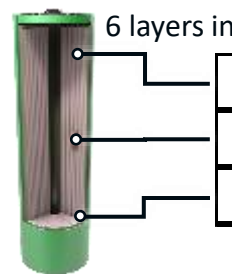
The number in each box represents the fraction of cells of that particular design, to undergo a particular failure mechanism



ISC Position	Design	Total	Bursting		Breach			Contained
			Top	Bot	Top	Side	Bot	
None	220 μm, BV	45	0.00	1.00	0.00	0.00	0.44	0.00
None	220 μm, NBV	46	0.02	0.00	0.07	0.02	0.16	0.98
None	250 μm, NBV	43	0.00	0.00	0.07	0.00	0.00	1.00



Top	220 μm, BV	11	0.09	0.27	0.64	0.00	0.27	0.64
Mid	220 μm, BV	13	0.08	0.54	0.08	0.00	0.62	0.38
Bot	220 μm, BV	12	0.00	1.00	0.00	0.17	1.00	0.00



Top	250 μm, NBV	9	0.00	0.00	0.56	0.00	0.00	1.00
Mid	250 μm, NBV	7	0.00	0.00	0.43	0.14	0.43	1.00
Bot	250 μm, NBV	8	0.00	0.00	0.38	0.13	0.25	1.00

3 layers in



Finegan et al., Modelling and experiments to identify high-risk failure scenarios for testing the safety of lithium-ion cells, *J. of Power Sources*, 2019

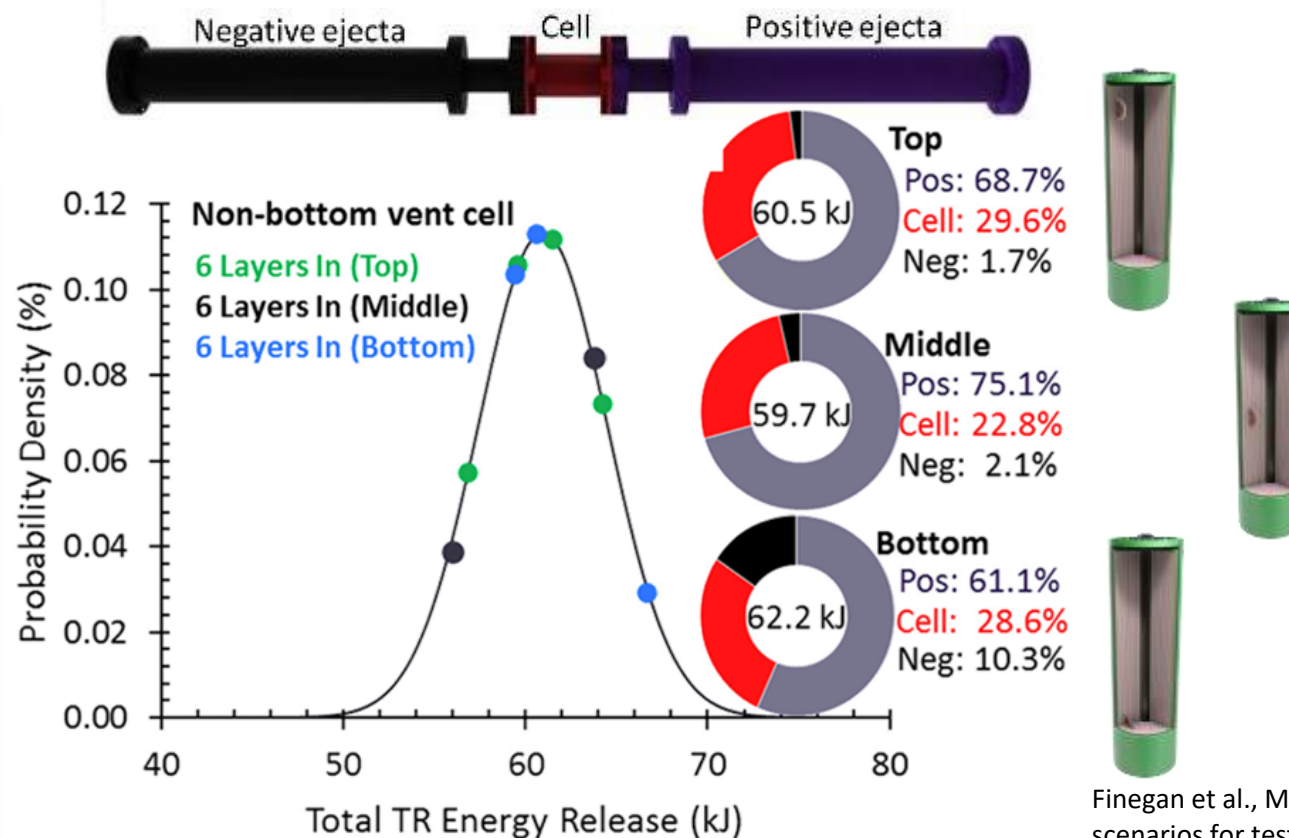
Key findings:

1. Proximity of the ISC device to either end increases the risk of breach/ bursting at that end.
2. Thicker casings reduce the risk of bursting but have a similar risk of breaching.
3. Bottom vents reduce the risk of breaching overall, but increase the risk of bottom breaching.

Fractional thermal runaway calorimeter (FTRC)

Ejected and non-ejected heat output

- 3.6 Ah 18650 cells
- Location of thermal runaway initiation **does not have significant impact on total heat output**, but does **influence the fraction of heat ejected**
- Around **70% of heat is ejected**, mostly through the positive vent
- Initiation near the bottom increases risk of bottom breach and heat from the bottom

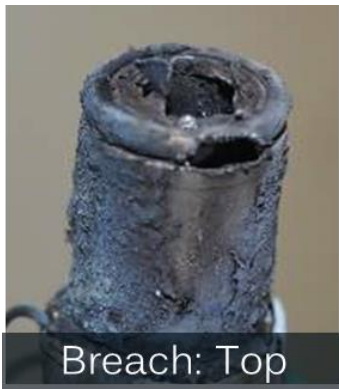


Finegan et al., Modelling and experiments to identify high-risk failure scenarios for testing the safety of lithium-ion cells, *J. of Power Sources*, 2019

Thermal stress and bursting pressure

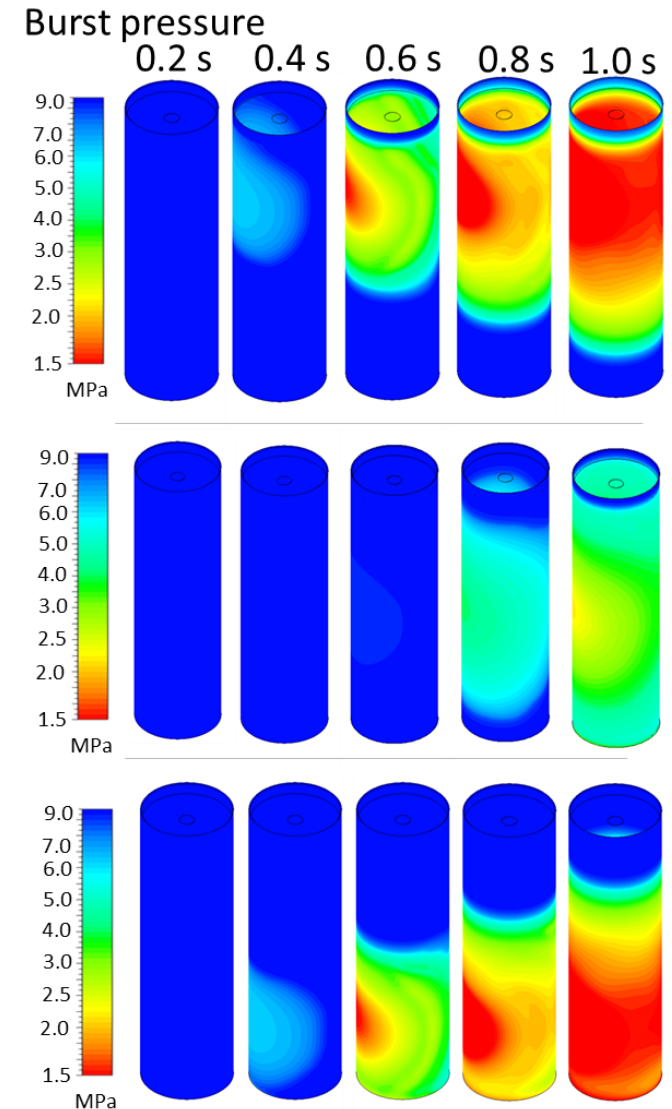
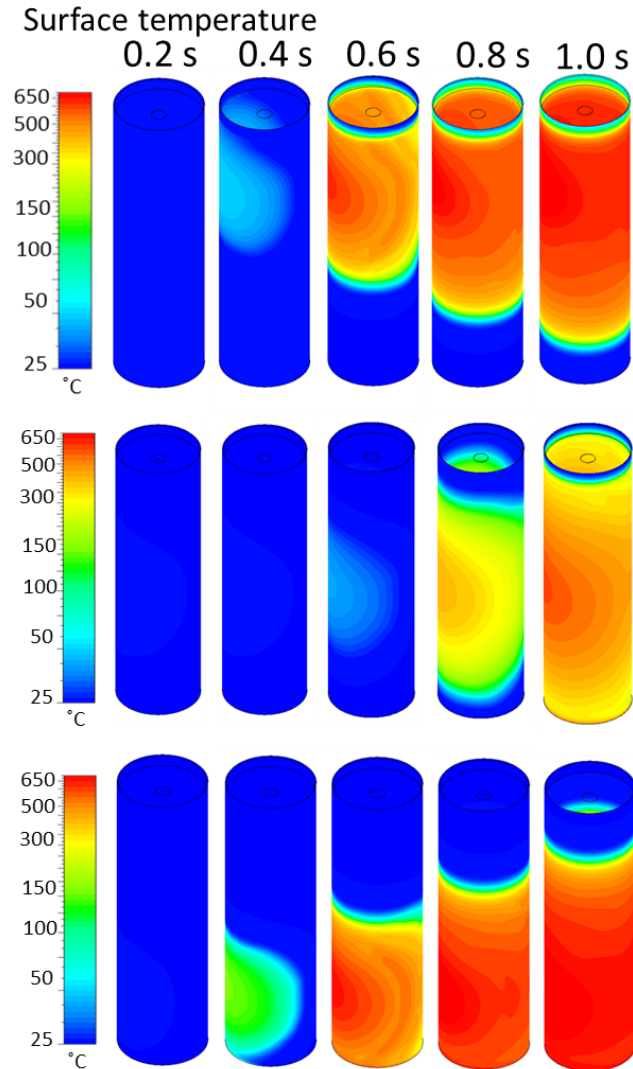
Surface temperature and burst pressure

- The **highest risk** scenarios for pressure-induced breaches are when initiation of **thermal runaway** occurs near **either end of the 18650 cell**.
- Burst pressures can reach < 1.5 MPa for temperatures > 650 °C.
- If a cell produces 6 L of gas, and is clogged, the internal pressure could reach 30 Mpa..



Breach: Top

Explains increased risk of breaching occurring, but not the consistent location at spin groove

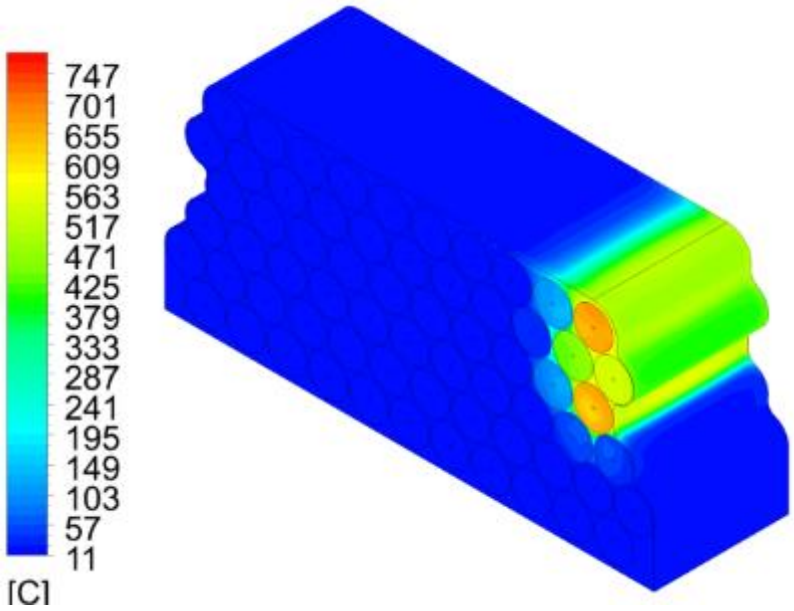


Based on tensile strength properties for S350GD mild steel

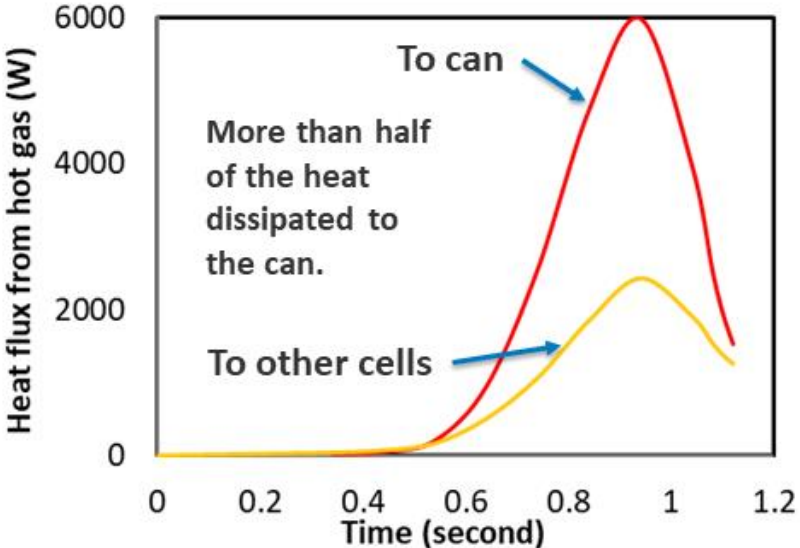
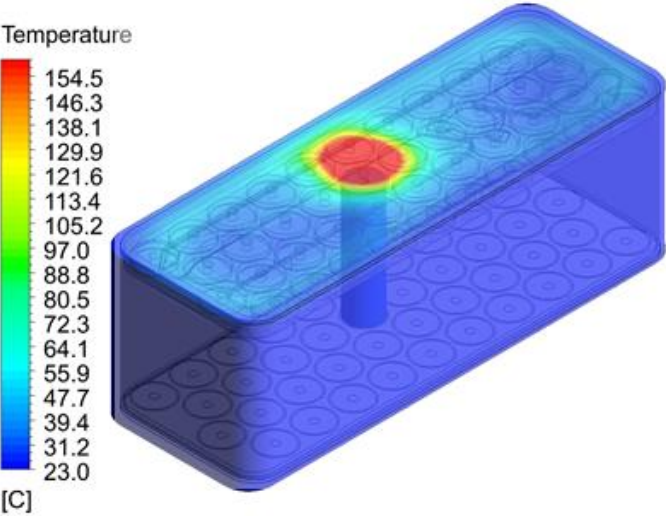
Results guiding safe battery designs

- Single cell data applied to battery pack simulations
 - Modelling sizing of heat sinks to avoid propagation
 - Estimating temperatures of pack enclosures when subject to ejected heat
 - Spatially quantifying the distribution of heat within an enclosure following cell failure

Heat sink sizing



Enclosure (can) subject to ejected heat



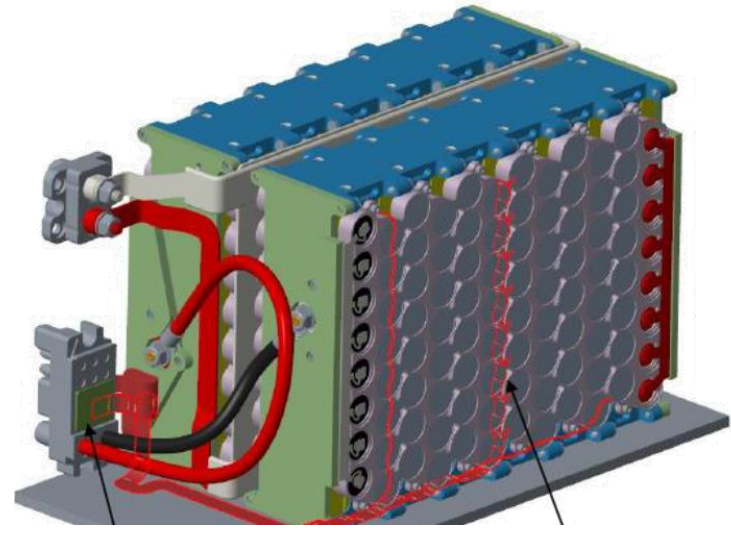
Work by Chuanbo Yang (NREL)

Results guiding safe battery designs

NASA X57 electric aircraft

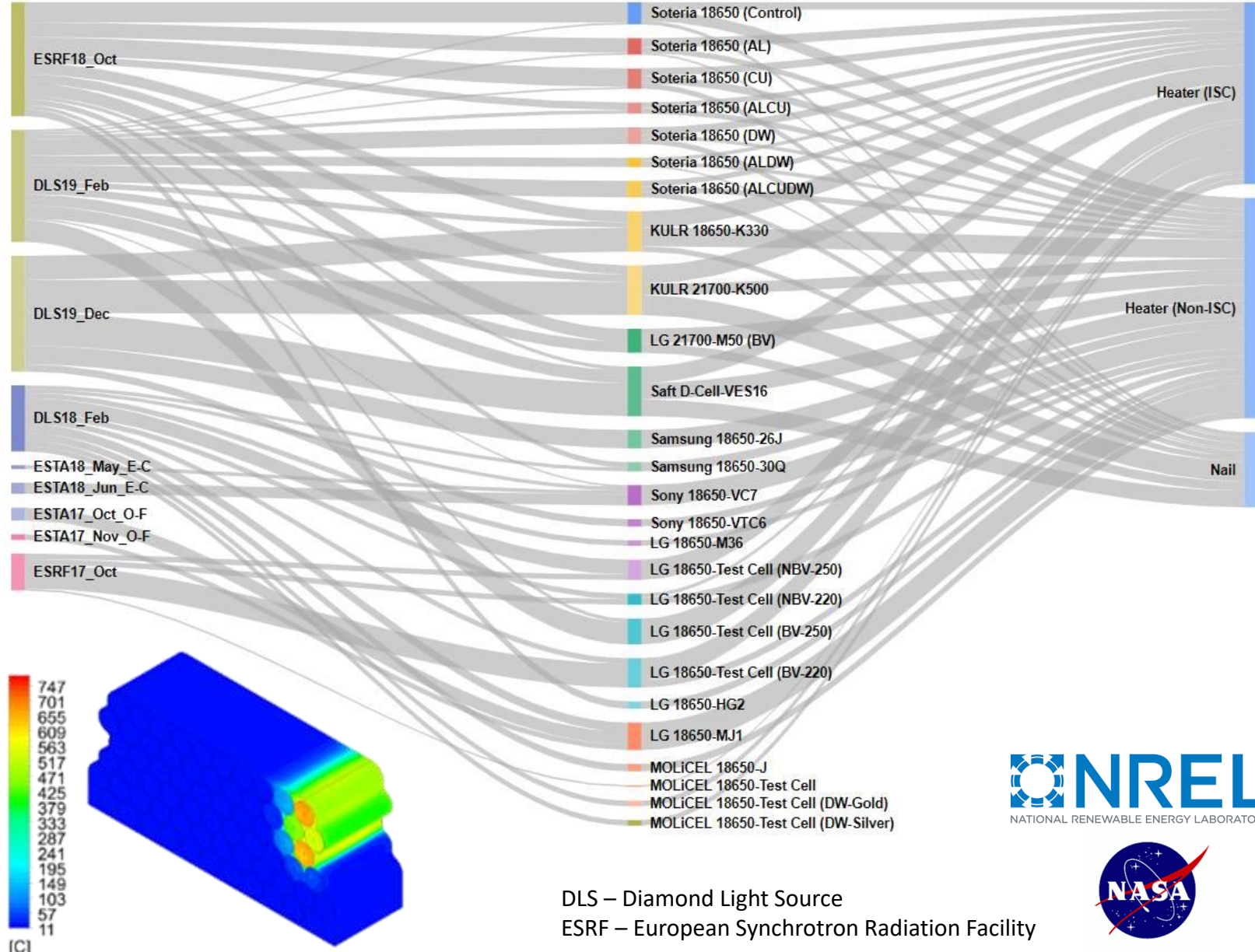


Eric Darcy and team at Johnson Space Center



Battery failure databank

- Radiography and thermal data from over 300 tests of commercial cells
 - Providing engineers and researchers with data to inform models
- Link internal phenomena with external risks
- Compare heat output and mass ejection from different abuse mechanicals
 - Nail penetration
 - Thermal abuse
 - Internal short circuiting
- Compare different models of cells
 - Power cells
 - Energy cells



DLS – Diamond Light Source
 ESRF – European Synchrotron Radiation Facility

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Acoustic Time of Flight - Introduction

- Rapid in-line, non-destructive cell diagnostic technique
- High-frequency pulse-echo mechanism to evaluate electrode level changes

Attenuation

$$\Delta Z = \sqrt{|\Delta\rho|K}$$

Velocity

$$v = \sqrt{\frac{K}{\rho}}$$

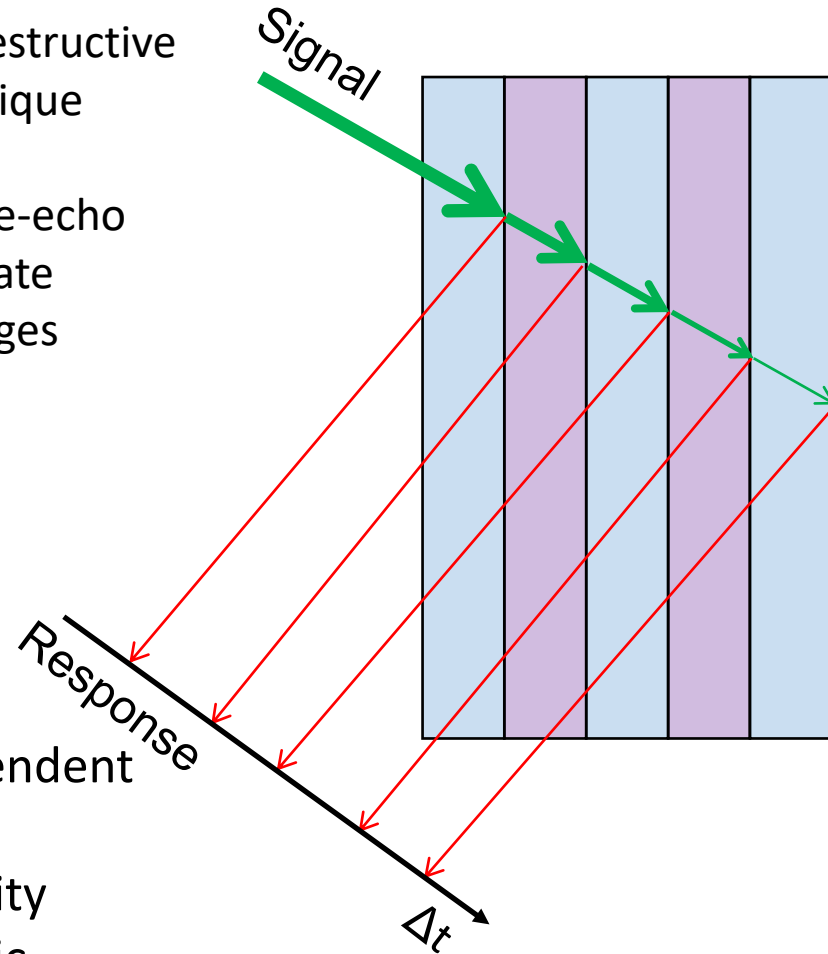


Figure 1: Illustrating the fundamental principles of acoustic time of flight

- Time of flight dependent on:
 - Material density
 - Material elastic modulus
 - Thickness
 - Structural changes

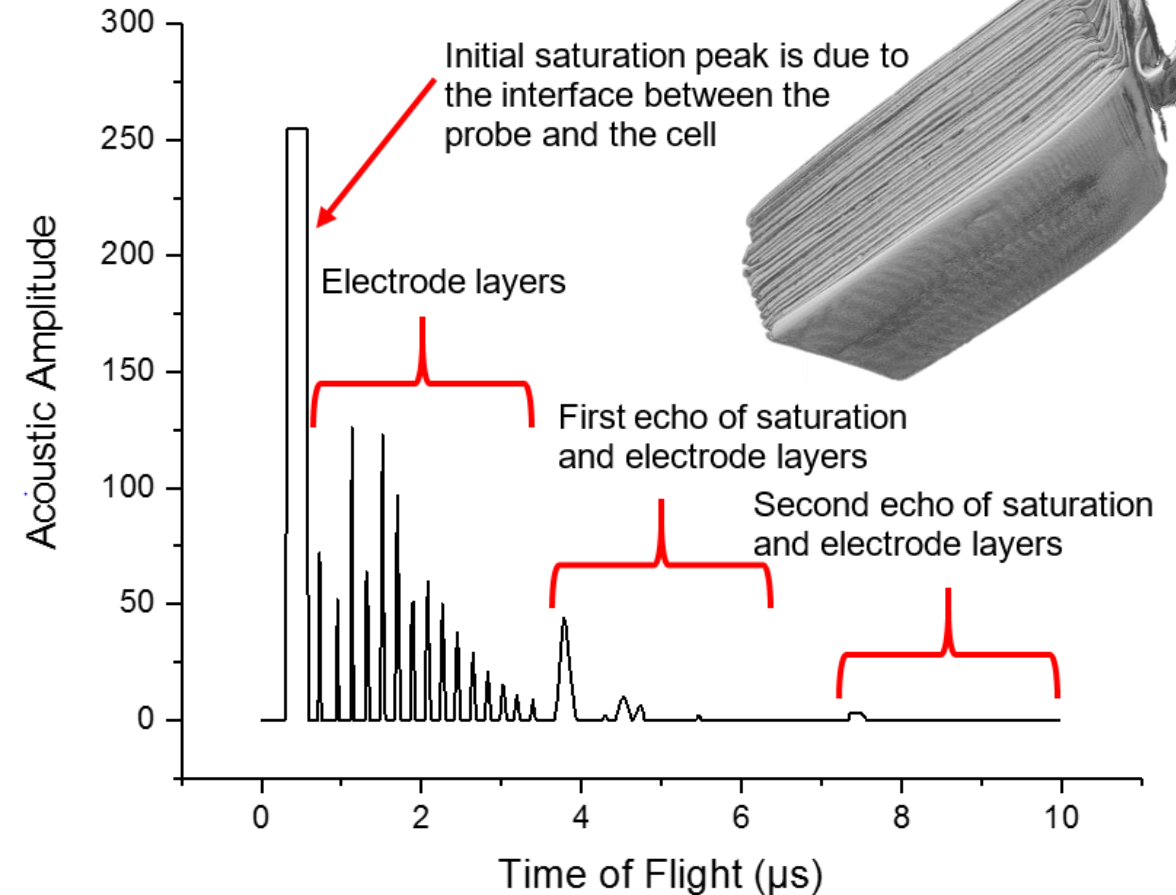


Figure 2: Representation of a characteristic acoustic time of flight waveform for a pouch cell, CT of corresponding pouch cell shown inset

Acoustic Time of Flight - Introduction

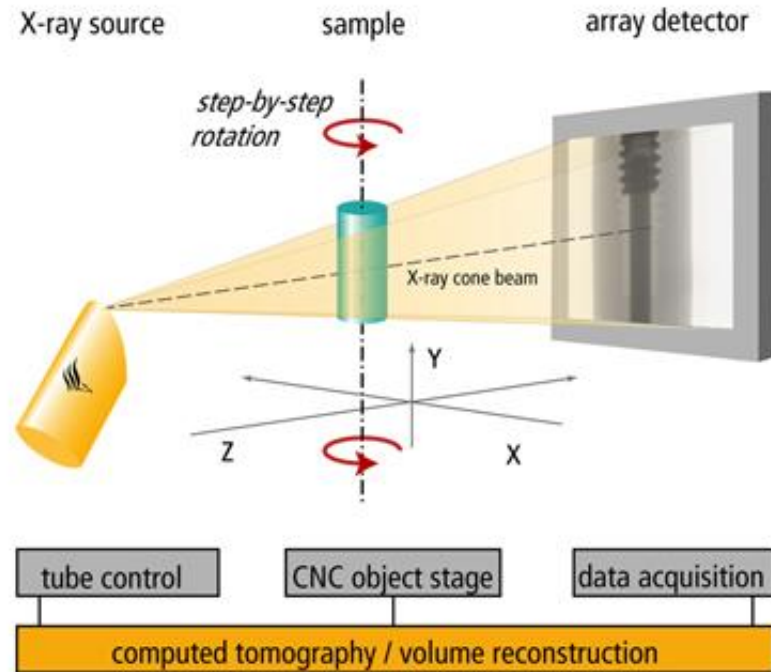


Figure 3: A simple schematic of the experimental set-up used in the Nikon XT H225 to produce simultaneous acoustic spectroscopy and X-ray imaging.

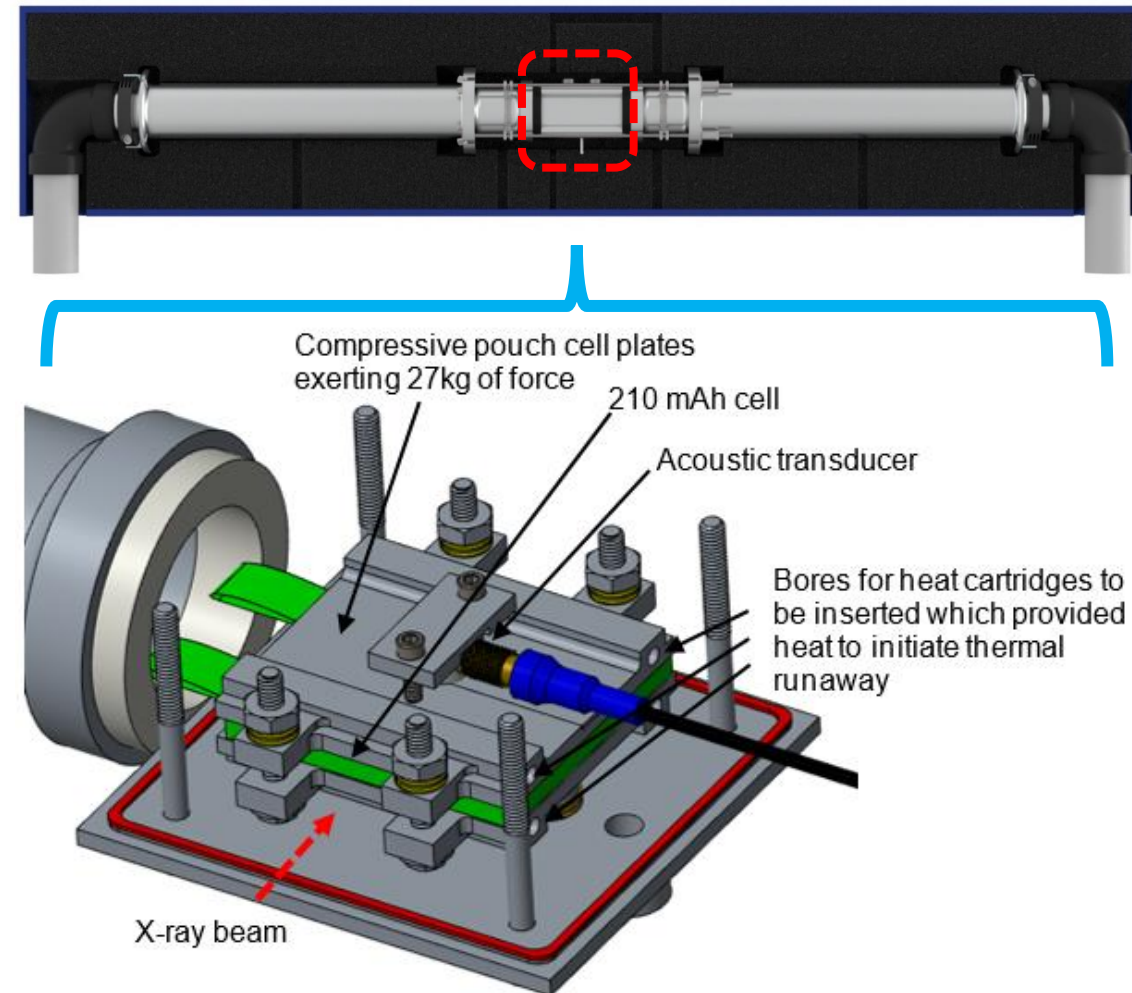


Figure 4: CAD rendering of the ultrasonic transducer in the pouch cell calorimeter cell chamber stack under compression

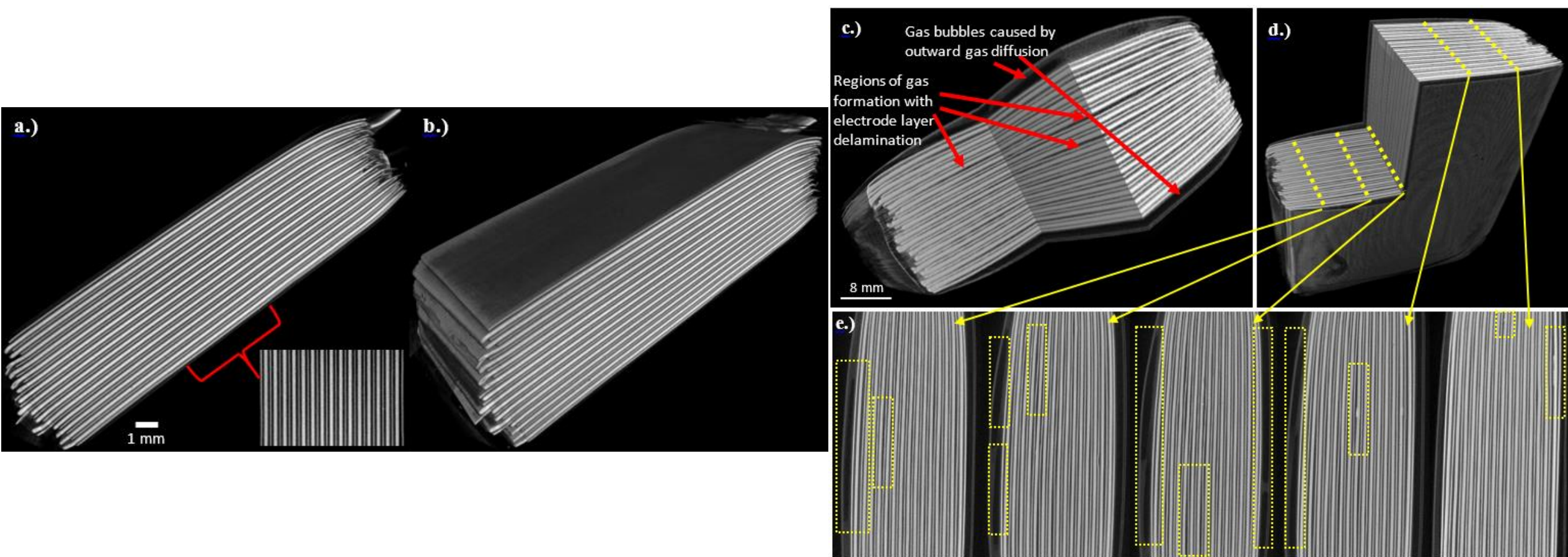
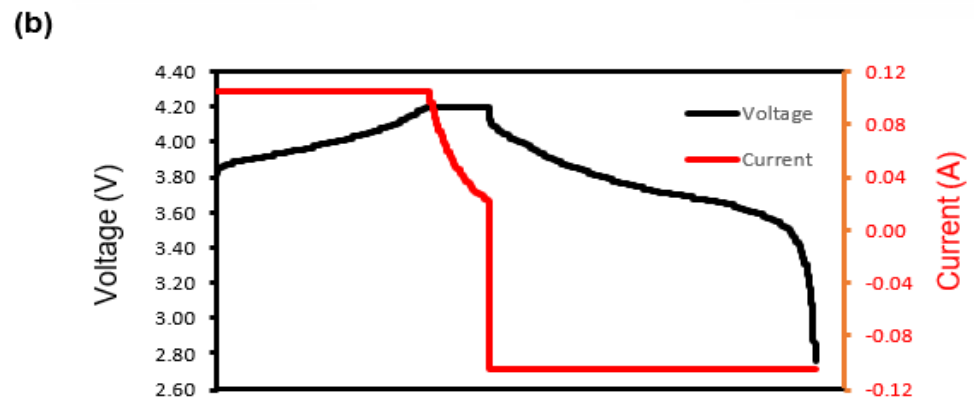
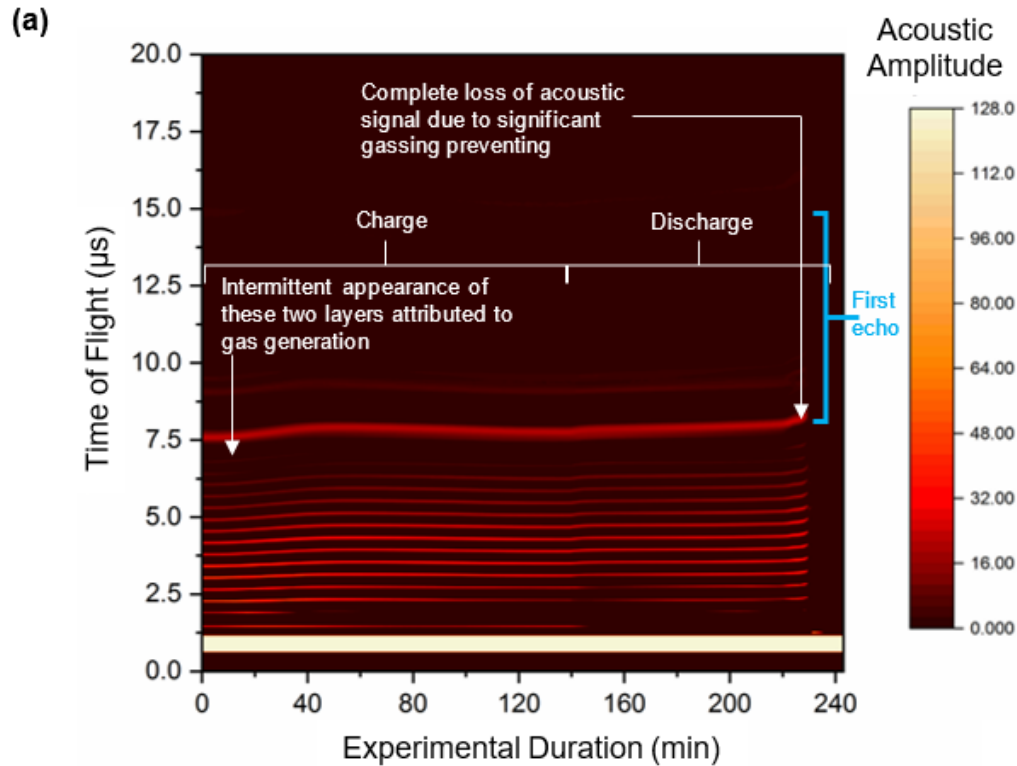


Figure 7: (a,b) Pre- and (c, d, e,) post- mortem XCT of defect driven delaminations in a commercial Li-ion cell

Acoustic Time of Flight – Defect Detection



Instantaneous acoustic pulse-echo 'snapshot' acquisition, at Experimental Duration = 0 min

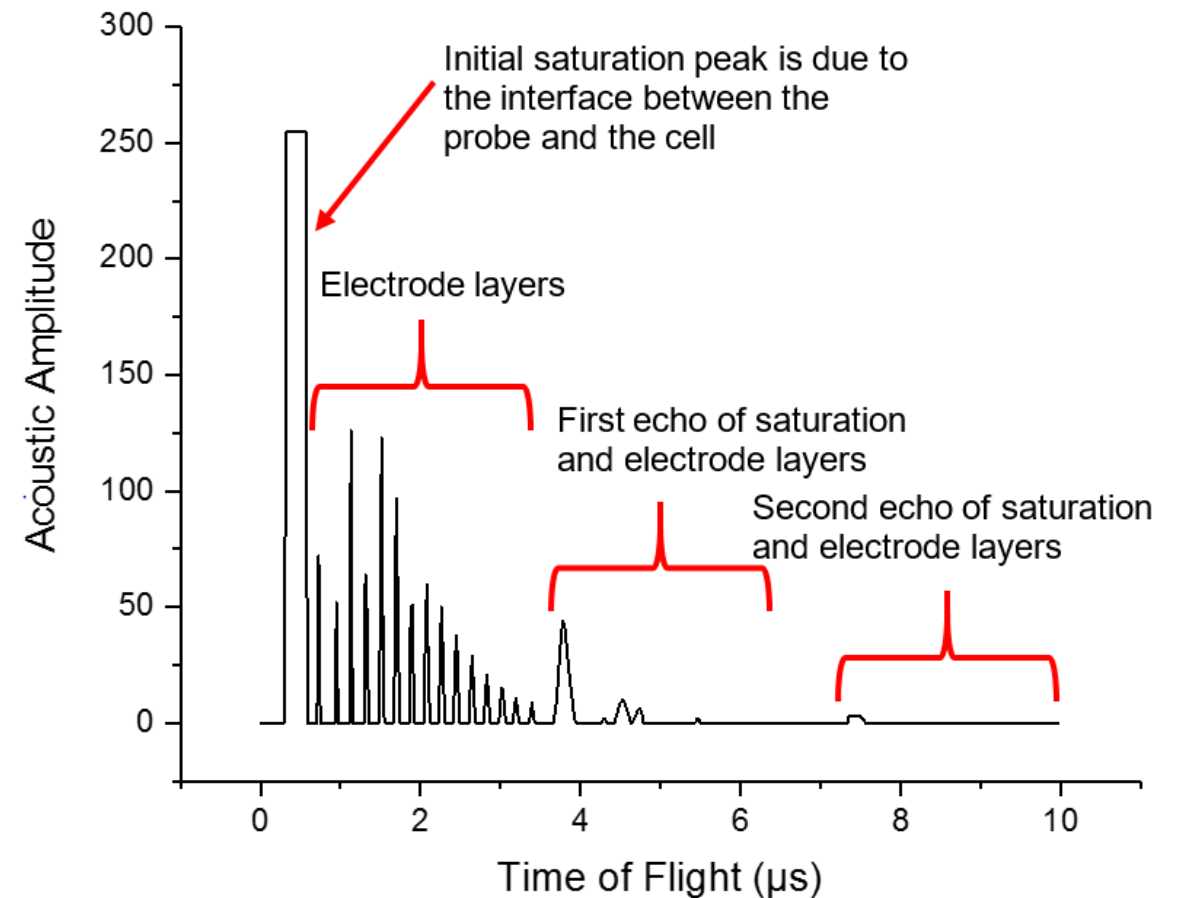


Figure 5: Acoustic time of flight during defect induced delamination during operation of a 210 mAh pouch cell.

Acoustic Time of Flight – Thermal Runaway

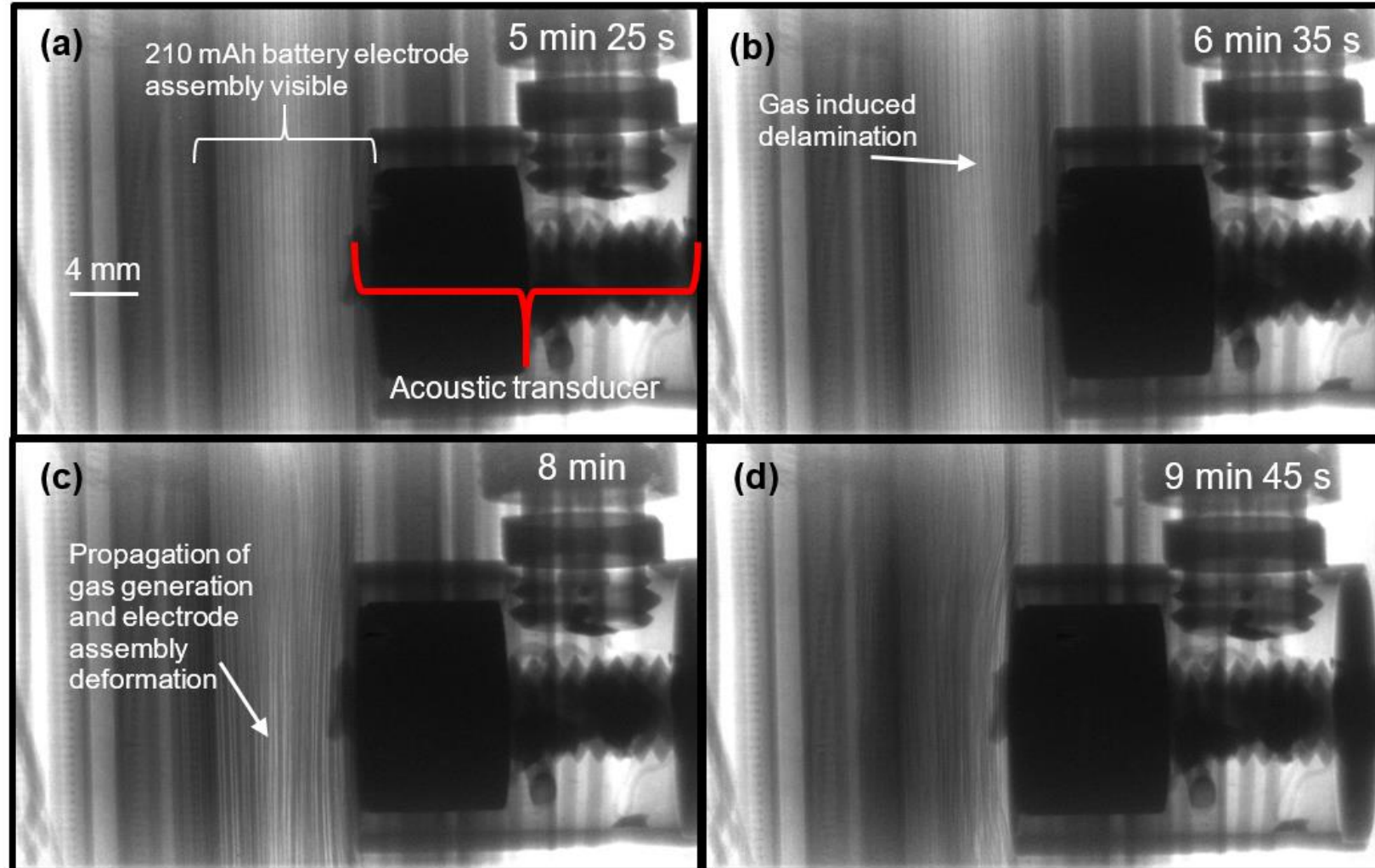


Figure 8: Selected radiography frames from Cell 1 highlighting significant structural changes during thermal runaway in the first failure test. Delamination and gas generation predicated the initiation of widespread thermal runaway.

Acoustic Time of Flight – Thermal Runaway

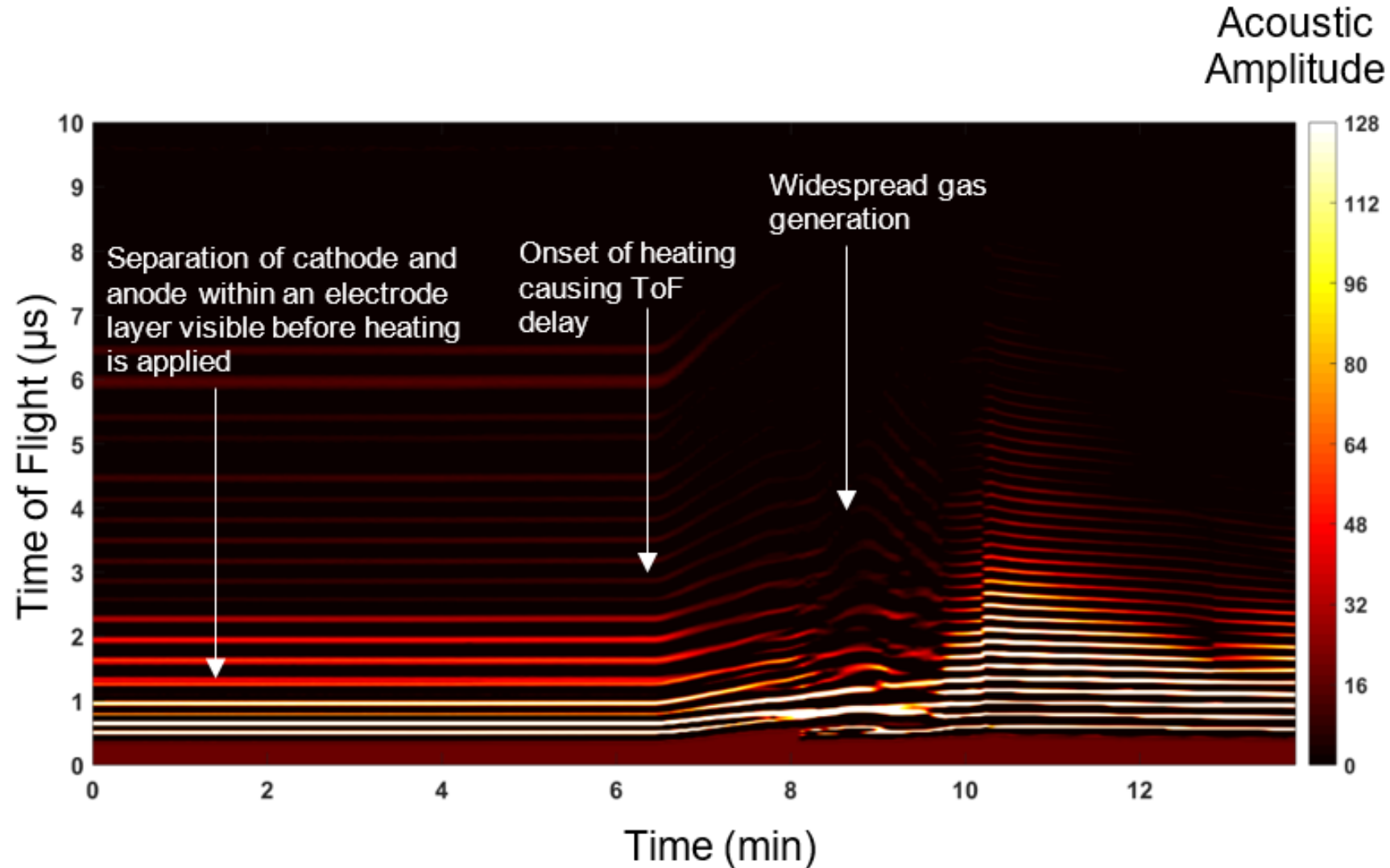


Figure 9: Acoustic time of flight during thermal runaway of Cell 1

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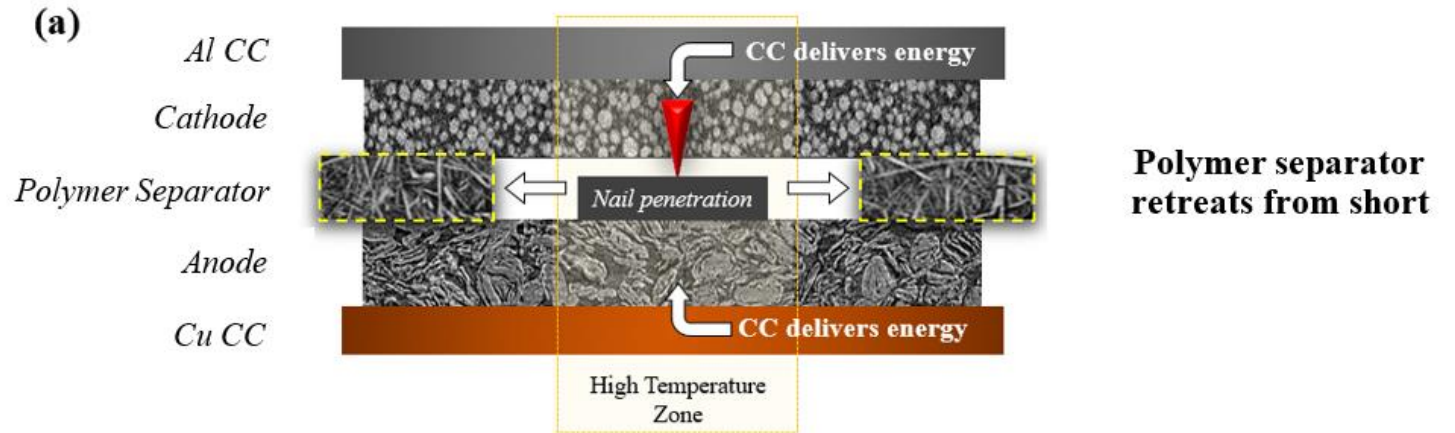
3. Materials for safer Li-ion batteries



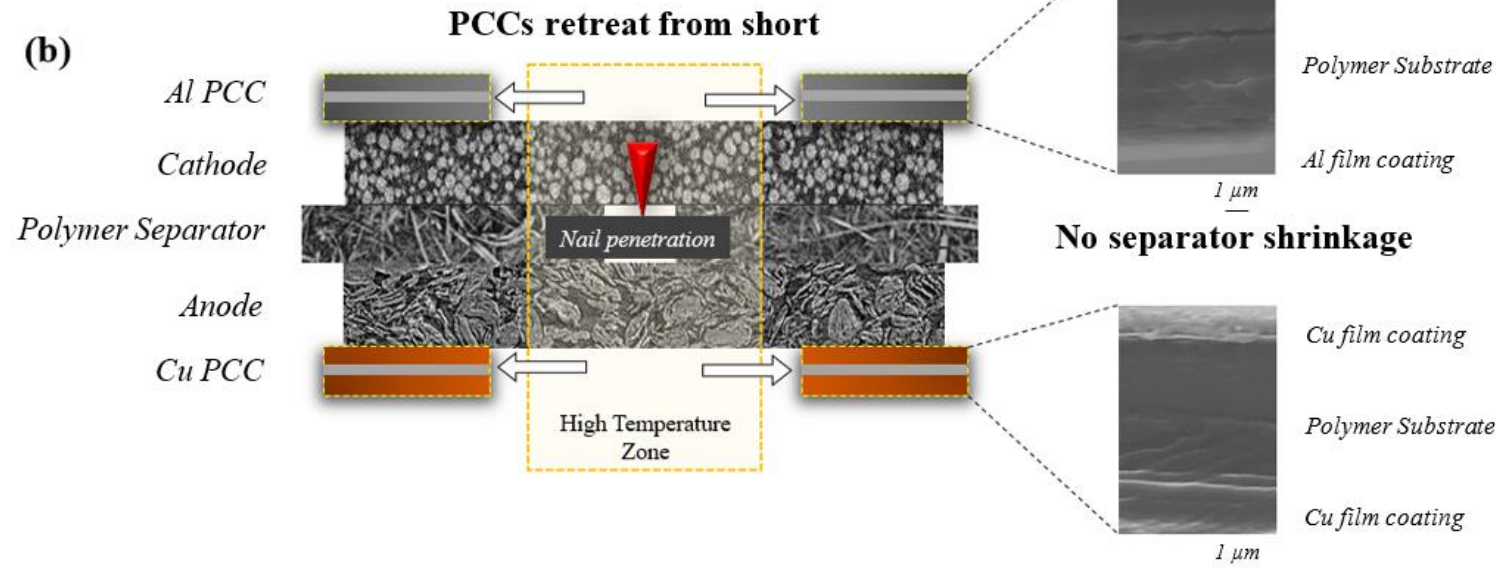
PCCs: Thermal Runaway Mechanisms

- Anode and cathode contact via the nail causes an unmitigated short-circuit
- Elevated temperatures due to Joule heating causes thermal decomposition of electrode components, initially with the polyolefin separator and electrolyte
- In cells with the PCC, the PCC would react to the elevated temperatures before the separator could fail, this would prevent cathode-anode contact as well as electrically isolating the cathode from the nail and the anode.
- Whether the PCC's safety mechanism acted before the failure of the 10 μm polypropylene separator would dictate the efficacy of the PCC.

Typical cell failure from nail penetration

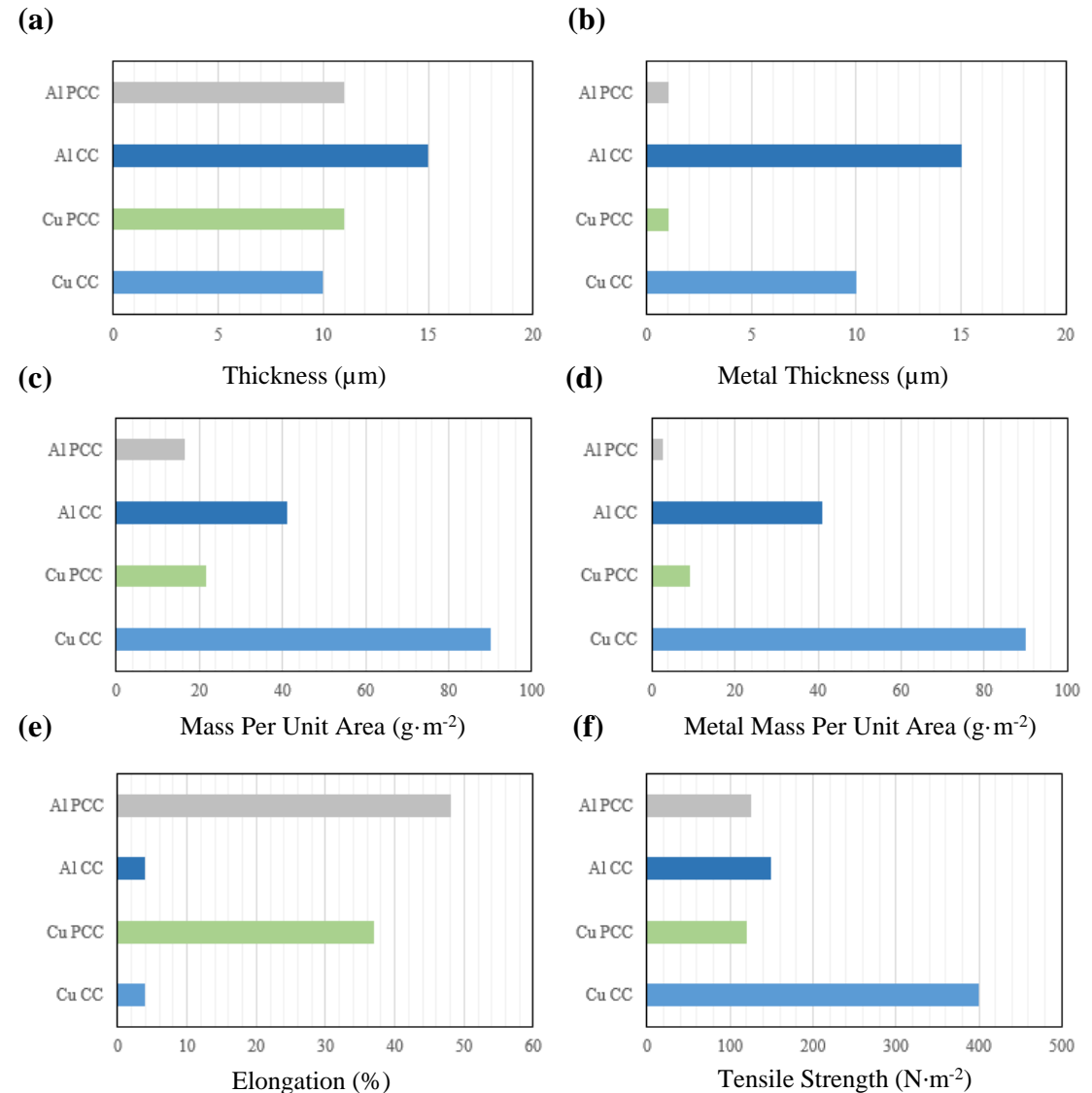


PCC thermal runaway prevention mechanism



PCCs: Gravimetric Energy Density

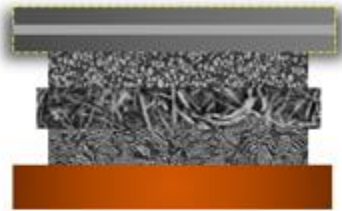
- Similar thicknesses to commercial metal foil current collectors (CCs):
 - Al CC: 15 μm , Al PCC: 11 μm
 - Cu CC: 10 μm , Cu PCC: 11 μm
- PCCs have a polymer substrate (ca. 8 μm thickness) with ca. 0.5 μm metal film coating of Al or Cu
- Therefore, a significant reduction in the amount of metal required by the PCC compared to commercial metal foils used as current collectors
- This reduction in metal is noticeable on the cell level as the average mass reduction was 2.2 grams, ca. 5% of total mass of a commercial metal foil CC control cell
- Mechanical properties of PCCs allow for assimilation with current cylindrical cell roll-to-roll manufacturing technology



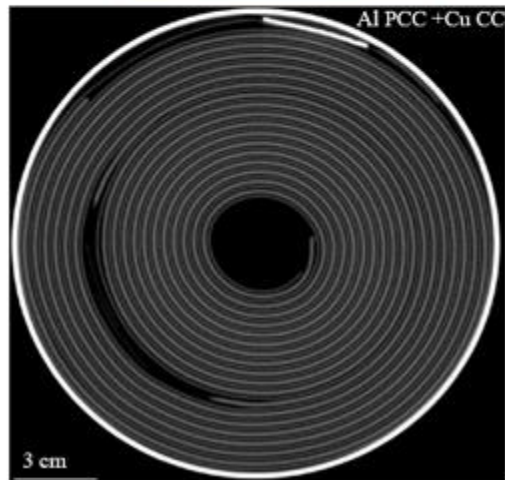
PCCs: Cell Groups and CT

Group 1, G1

Al PCC

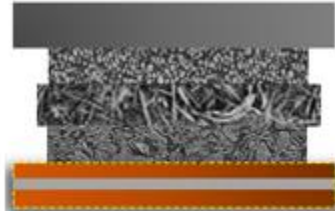


Cu CC

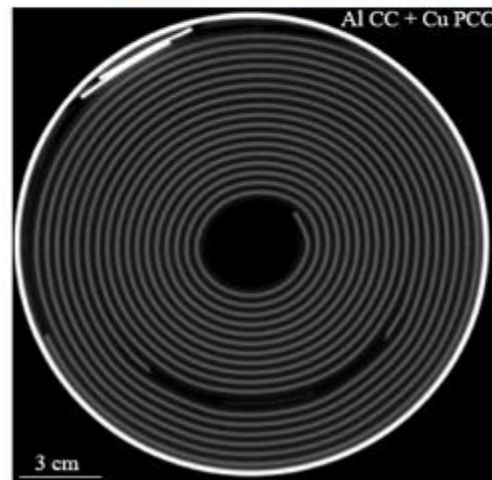


Group 2, G2

Al CC

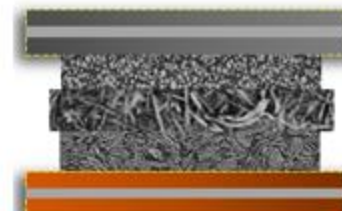


Cu PCC

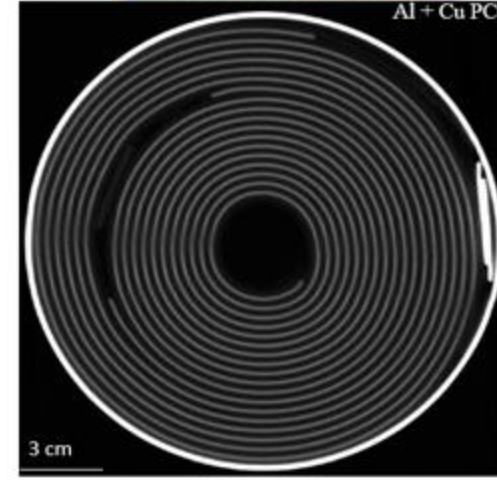


Group 3, G3

Al PCC

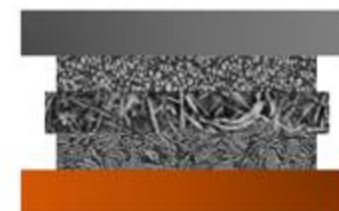


Cu PCC

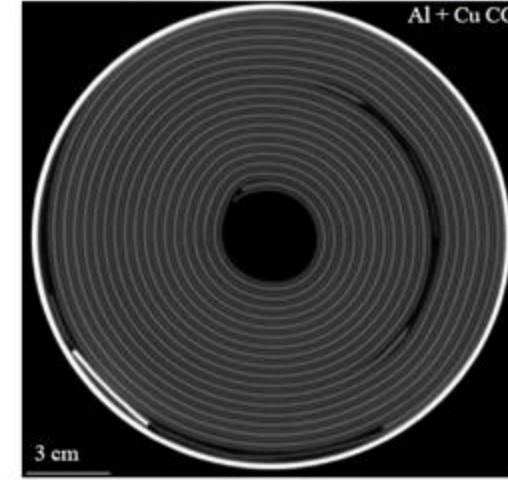


Group 4, G4

Al CC

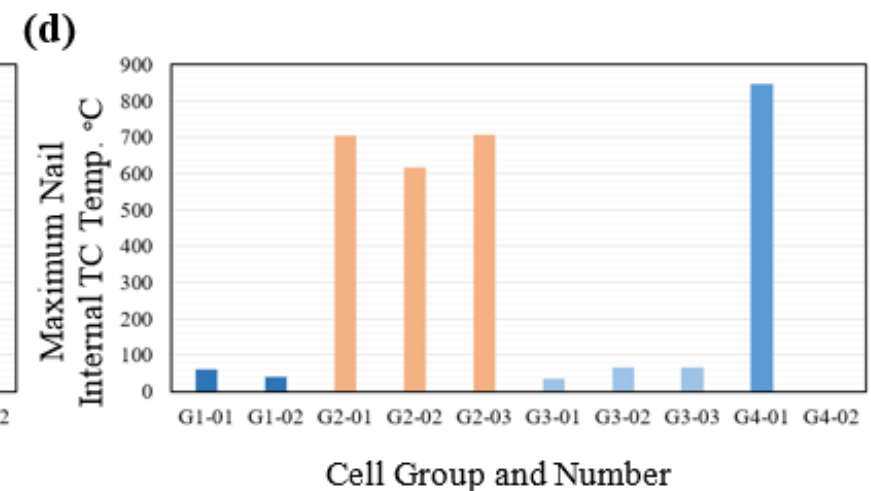
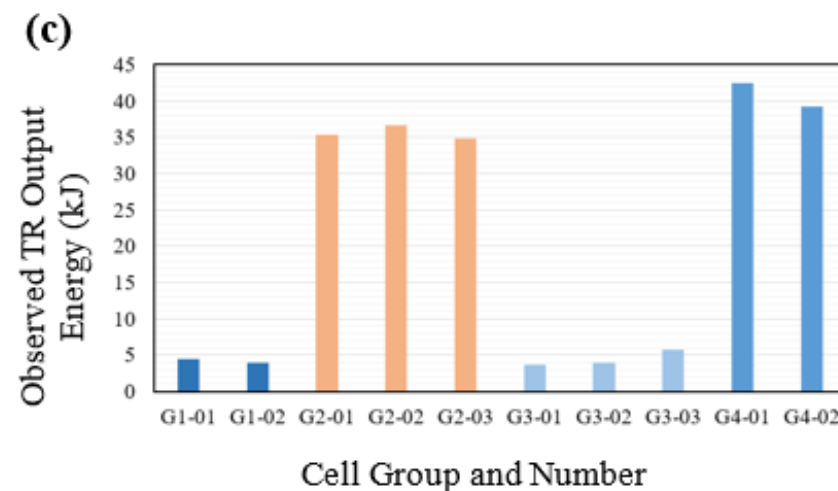
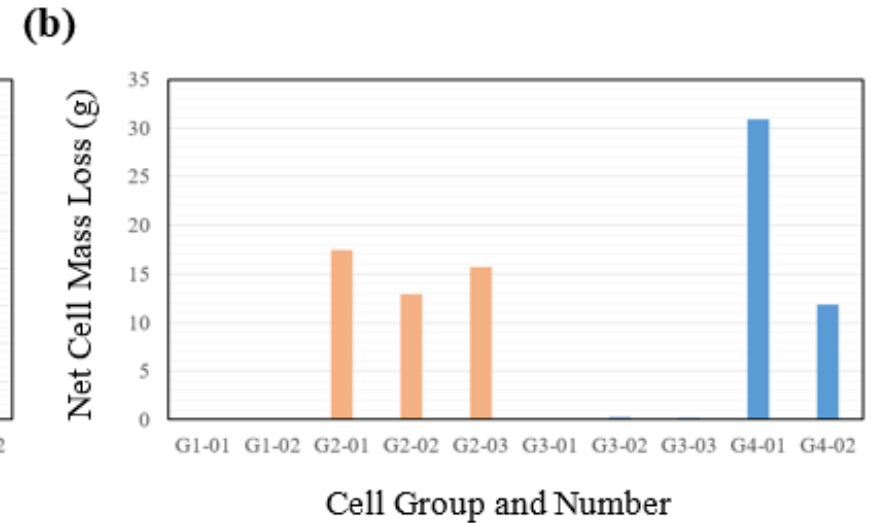
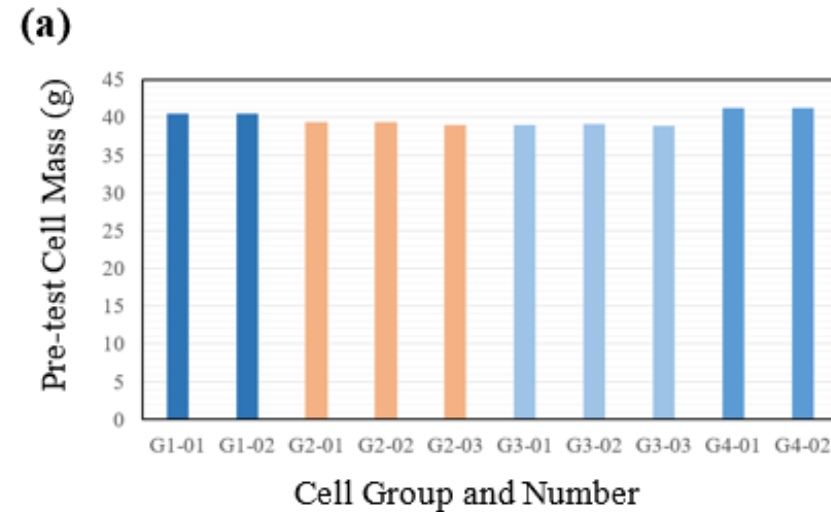


Cu CC



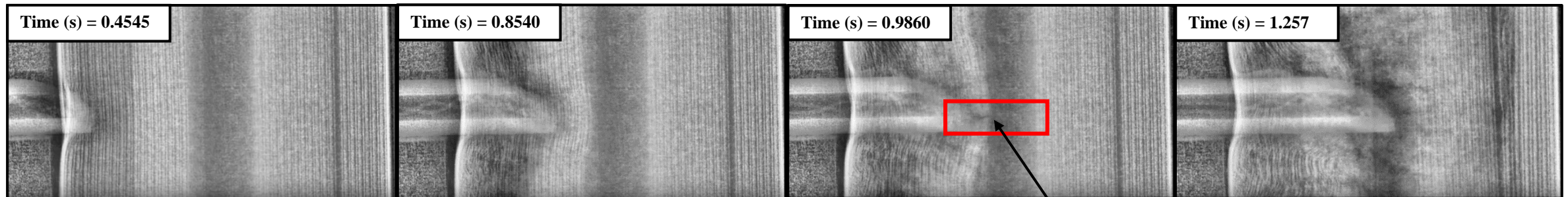
Results: Failure Prevention and Calorimetry

- Mass reduction in cells with one or both of the PCCs, mass reduction of 2.2 g with both
- Significant difference in observed TR output energy, maximum nail TC temperature and net cell mass loss with Al PCC cells
- Cu PCC alone did not protect the cells from undergoing thermal runaway but mitigation of the output energy was observed. This was due to the nail in contact with the cell during penetration, thus the Cu PCC despite protecting the anode, the graphite was connected to the cell can and shorted with the unprotected cathode.



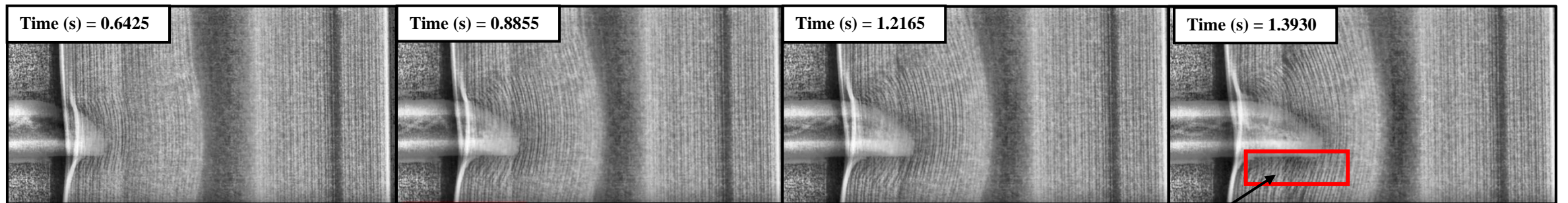
Results: Synchrotron X-ray Radiography

(a) G4-01 (Al CC + Cu CC) Radiography



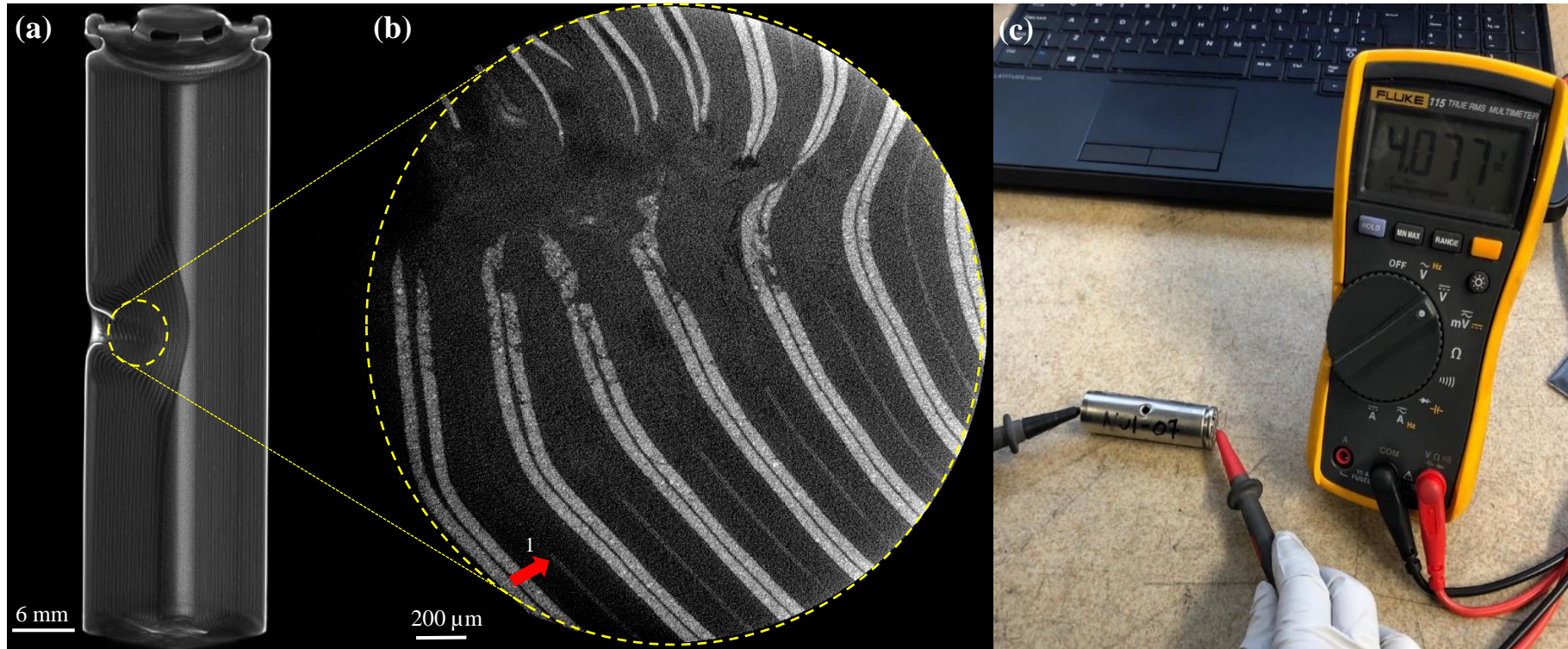
Characteristic cracking when Al PCC is absent

(b) G1-01 (Al PCC + Cu CC) Radiography



Electrode layers adjacent to nail splitting

Results: Post-mortem CT and 4.077 V



Sub-micron X-ray CT reveal Al PCC had shrunk away from the elevated temperature of the nail, which caused the microscopic short-circuit. This prevented further short-circuiting, subsequent OCV measurement showed 4.077 V. Cells retained voltage for over 10 months.

- High-speed X-ray imaging useful for **guiding and validating thermal runaway models** for identifying internal and external hotspots.
- Highest surface temperatures and **lowest burst pressures were achieved when initiation occurred near either ends of the cell**, due to relatively poor heat dissipation.
- The likelihood of high-risk failure scenarios can be increased by **selectively locating** the point of thermal runaway initiation within a cell.
- Thermal data from the fractional thermal runaway calorimeter (FTRC) is useful for accurately modelling efficacy of heat sinks and enclosures for withstanding thermal runaway.
- An **open-source database** of radiography and thermal data to be released soon.
- Acoustic technique demonstrated to enable detection of deformed electrode layers leading up to thermal runaway.
- Soteria's polymer substrate current collectors demonstrated to **withstand nail penetration without thermal runaway** where otherwise thermal runaway consistently occurs. Light-weight, cost effective, and mechanically robust for scaling up.

Thank you for listening

Donal Finegan

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List of relevant publications:

1. Finegan et al., Characterising thermal runaway within lithium-ion cells by inducing and monitoring internal short circuits. *Energy & Environmental Science* **2017**, 10 (6), 1377-1388.
2. Finegan et al., Identifying the Cause of Rupture of Li-Ion Batteries during Thermal Runaway. *Advanced Science*, 1700369, **2017**.
3. Finegan et al., In-operando high-speed tomography of lithium-ion batteries during thermal runaway. *Nature Communications* **2015**, 6.
4. Finegan et al., Tracking Internal Temperature and Structural Dynamics during Nail Penetration of Lithium-Ion Cells. *Journal of The Electrochemical Society* **2017**, 164 (13), A3285-A3291.
5. Finegan et al., Modelling and experiments to identify high-risk failure scenarios for testing the safety of lithium-ion cells, *J. of Power Sources*, **2019**
6. Walker et al., Decoupling of heat generated from ejected and non-ejected contents of 18650-format lithium-ion cells using statistical methods, *J. Power Sources*, **2019**
7. Robinson et al., Spatially Resolved Ultrasound Acoustic Investigations of Li-Ion Batteries, *Phys. Chem. Chem. Phys.*, **2019**
8. Pham et al., Correlative acoustic time-of-flight spectroscopy and X-ray imaging to investigate gas-induced delamination in lithium-ion pouch cells during thermal runaway, *J. Power Sources*, **2020**

www.nrel.gov

