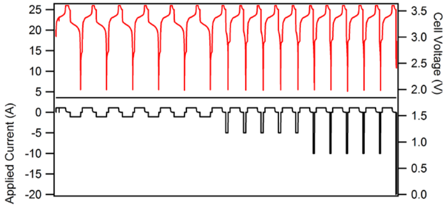
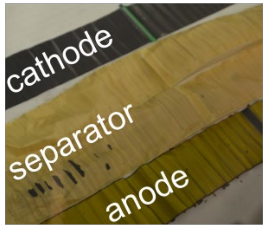
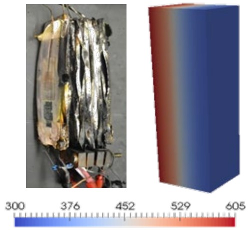
Impact of Aging on the Safety of Lithium-ion Batteries







PRESENTED BY

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Project objective and OE mission alignment

OBJECTIVE: Assess the impact of chemistry, aging level, and abuse technique on the abuse response of unused vs. aged cells

SIGNIFICANCE

- Testing for battery safety standards + publications is typically done with unused cells, but cells are 'unused' for only a small fraction of their lifetime
- Understanding how the abuse response of aged cells differs from unused cells will enable the design of more effective energy storage failure mitigation systems, especially for second-life systems

ALIGNMENT WITH CORE MISSION OF DOE OE

- Energy storage systems contribute to resilience, reliability, and flexibility of energy infrastructure
- Safety concerns are a major barrier to system deployment and collecting safety data for more relevant configurations will lead to improved system designs

Gap: Standards only call for safety tests on unused cells

Example Testing Report

Test	Unused Cell
Continuous charging at constant voltage	✓
External short circuit	✓
Free fall	✓
Thermal abuse	✓
Crush	✓
Forced discharge	✓
Forced internal short circuit	✓

Source: LG Chem 18650HG2, IEC Test Report issued under UL

Gap: Standards only call for safety tests on unused cells

Example Testing Report

Test	Unused Cell	Aged Cell
Continuous charging at constant voltage	✓	x
External short circuit	✓	x
Free fall	✓	x
Thermal abuse	✓	x
Crush	✓	x
Forced discharge	✓	x
Forced internal short circuit	✓	x

Source: LG Chem 18650HG2, IEC Test Report issued under UL

Failure mitigation mechanisms are developed based on the outcomes of these tests

Influence of battery aging on safety is unclear

- More safe (due to capacity loss)?
- Less safe (due to materials/component instability)?
- Little difference?

Previous work on safety of aged batteries

In 2022, we published the first comprehensive review of all public data on the safety of aged Li-ion batteries in collaboration with UL Electrochemical Safety Research Institute

Key takeaways

- "Safety" depends on the metric and aging conditions
- Li plating lowers thermal runaway onset temperatures

Identified areas with limited data

- Electrical and mechanical abuse response
- LFP cathodes
- Off-gassing*
- Modules

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Perspective—On the Safety of Aged Lithium-Ion Batteries

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Concerns about the safety of lithium-ion batteries have motivated numerous studies on the response of fresh cells to abusive, off-nominal conditions, but studies on aged cells are relatively rare. This perspective considers all open literature on the thermal, electrical, and mechanical abuse response of aged lithium-ion cells and modules to identify critical changes in their behavior relative to fresh cells. We outline data gaps in aged cell safety, including electrical and mechanical testing, and module-level experiments. Understanding how the abuse response of aged cells differs from fresh cells will enable the design of more effective energy storage failure mitigation systems.

Manuscript submitted December 10, 2021; revised manuscript received January 19, 2022. Published March 4, 2022. *This paper is part of the JES Focus Issue on Women in Electrochemistry.*

High voltage and high energy density lithium-ion (Li-ion) batteries are increasingly used in many applications, including consumer electronics, electric vehicles, and grid-tied energy storage systems. However, with increasing energy density and installation size come heightened concerns about safety, including the risk of thermal runaway and severe fires.[1–4] Thermal runaway is traditionally defined as an accelerating release of heat inside a cell, due to a

summarize critical parameters by which the safety of aged cells relative to fresh cells may be evaluated. We then identify aspects of aged battery safety that merit further exploration.

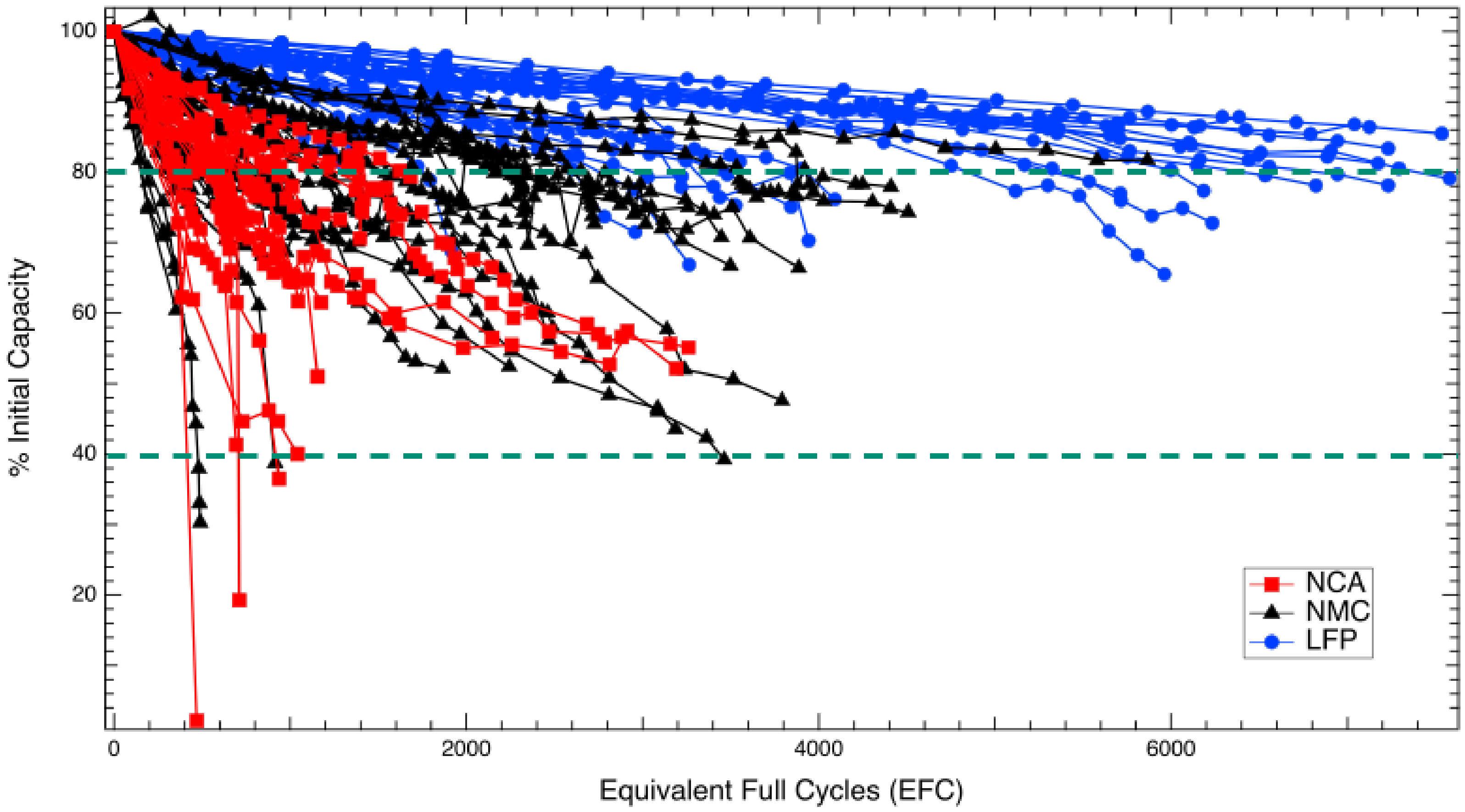
Current Status

Thermal abuse of aged cells.—Batteries in fielded systems can

*current Sandia focus (Nathan Johnson)

Project methodology – Use cells from previous OE work on battery aging

Cells cycled for 7+ years at various conditions
(full cycling history and materials characterization are available)



Preger et al. *J. Electrochem. Soc*. **2020**, *167*, 120532; Wittman et al. *J. Electrochem. Soc*. **2023**, *170*, 120538. See Sahana Upadhya's poster for more details on cycling study.

Project testing matrix

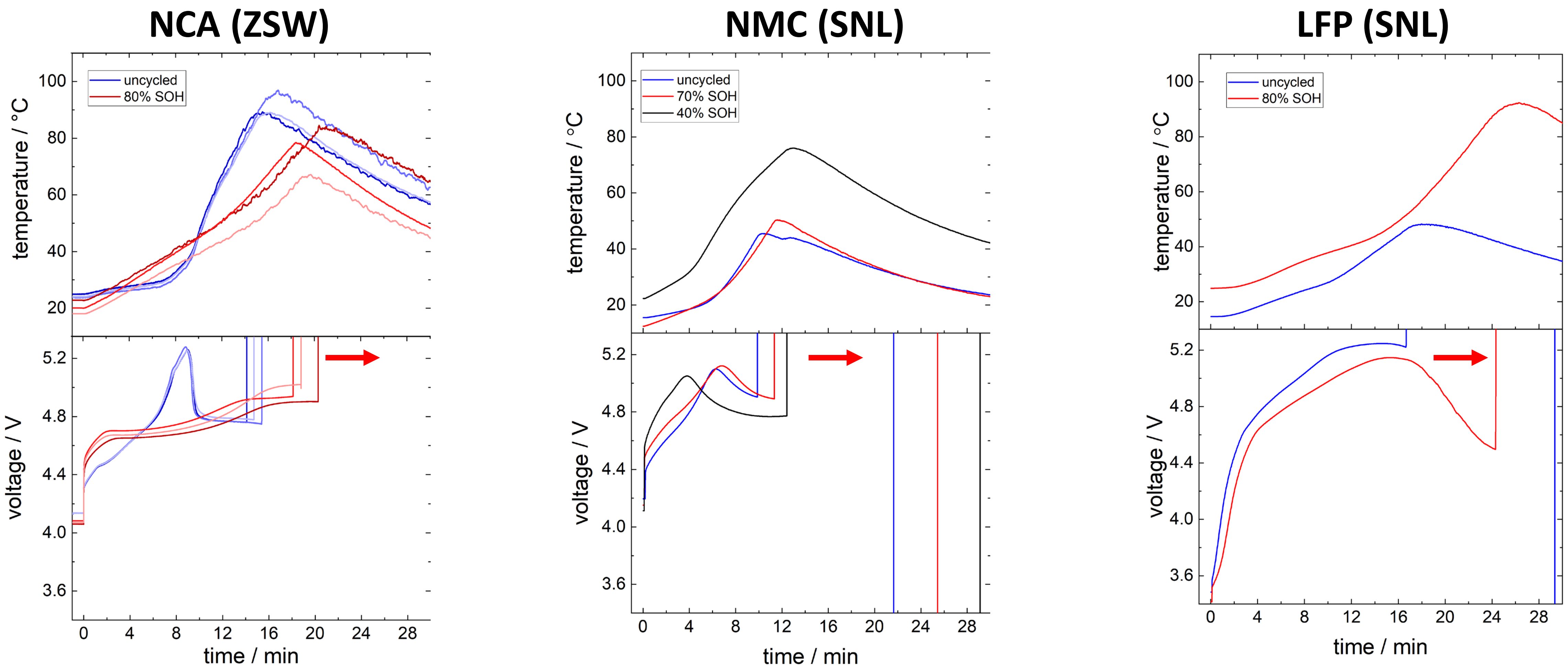
Collaboration with Center for Solar Energy and Hydrogen Research in Germany (ZSW) allows us to broaden the matrix

Cell	SOH* %	Overcharge	Overdischarge	ARC	Nail penetration	Crush after overcharge	Experiments conducted
NCA-uncycled	100	3	3	2	3		ZSW
NCA-aged-80%	74-85	3	3	3	3		ZSW
NMC-uncycled	100	1	1	1		1	SNL
NMC-aged-70%	70-74	1	1	1		1	SNL
NMC-aged-40%	43	1	TBD	TBD		1	SNL
LFP-uncycled	100	1	1	1		1	SNL
LFP-aged-80%	80-83	1	1	1		1	SNL
LFP-aged-40%		TBD	TBD	TBD		TBD	SNL

1. Does the impact of aging vary for **different abuse protocols**?
2. Does the safety of aged cells vary with **chemistry**?
3. Does the safety of aged cells vary for **different aging levels**?
4. How **reproducible** are the results?

*avoid conditions with Li plating

Impact of aging and chemistry on overcharge tolerance



Aged cells were **overcharged more** prior to CID activation (voltage spike), in contrast to previous literature. This highlights the sensitivity of abuse response to aging conditions.

Overcharge visuals reveal another dimension of safety

Uncycled LFP

EUCAR Rating: 2

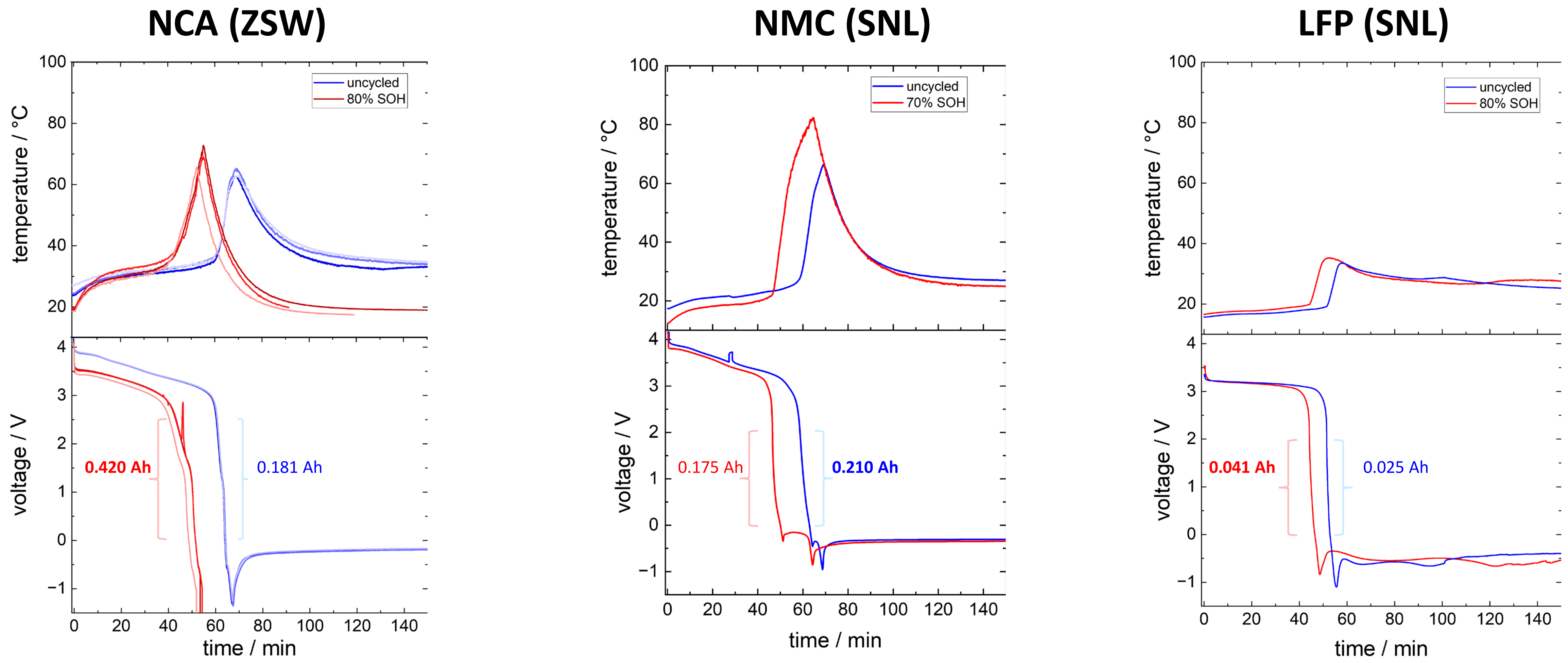
- CID activation
- Loss of functionality

LFP – 80% SOH

EUCAR Rating: 3

- CID activation
- Loss of functionality
- **Electrolyte leakage**

Impact of aging and chemistry on overdischarge tolerance



Aged NCA and LFP cells (but not NMC) were **overdischarged more** prior to dropping to 0 V

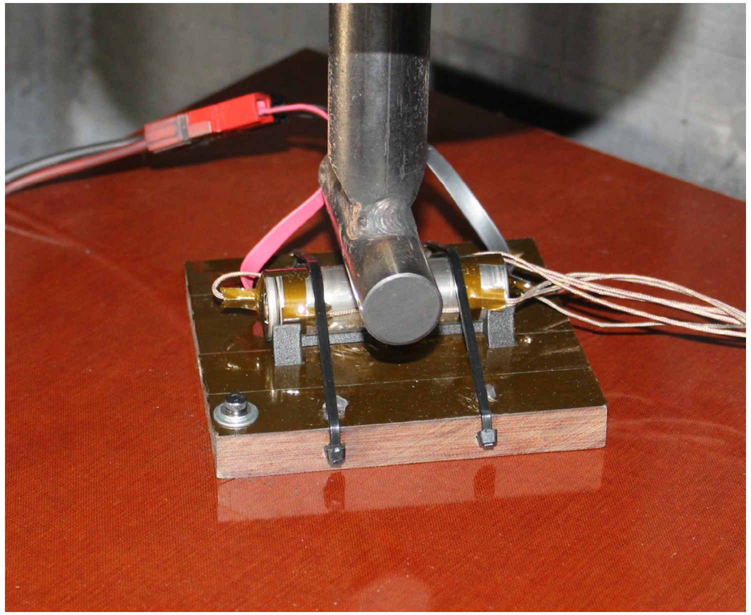
Overdischarge visuals don't capture the differences

Uncycled NMC

EUCAR Rating: 2

- Loss of functionality
- No venting

NMC – 70% SOH



EUCAR Rating: 2

- Loss of functionality
- No venting

Multiple metrics needed to fully assess safety profile of aged cells

Key takeaways

1. Does the impact of aging vary for **different abuse protocols**?
   Yes

2. Does the safety of aged cells vary with **chemistry**?
   Yes

3. Does the safety of aged cells vary for **different aging levels**?
   Yes

4. How **reproducible** are the results?
   Depends on the technique

Future work

- Publications based on: (1) current testing matrix, (2) cycling beyond 80% SOH, (3) materials characterization at 80% and beyond
- More abuse tests as additional cells reach end-of-life

Innovations and impacts

We know batteries can last a long time and degrade in different ways, but there has been limited testing to assess how their safety profiles change over time

- Cycling study represents the broadest public work on post 80% Li-ion aging
- Previous review with UL represents the first comprehensive assessment of aged battery safety
- Current dataset with ZSW represents the broadest single dataset on the abuse of aged cells

Results are relevant for anyone dealing with battery control or emergency response, e.g.,:

- Battery management system tolerance for overcharge/discharge
- Thermal management in case of failure
- Second life battery policy questions

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Sandia team
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Cell aging: Reed Wittman, Chaz Rich

Perspective on safety of aged cells – UL Electrochemical Safety Research Institute
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Electrical abuse of aged cells – ZSW
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UL Research Institutes

ZSW

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