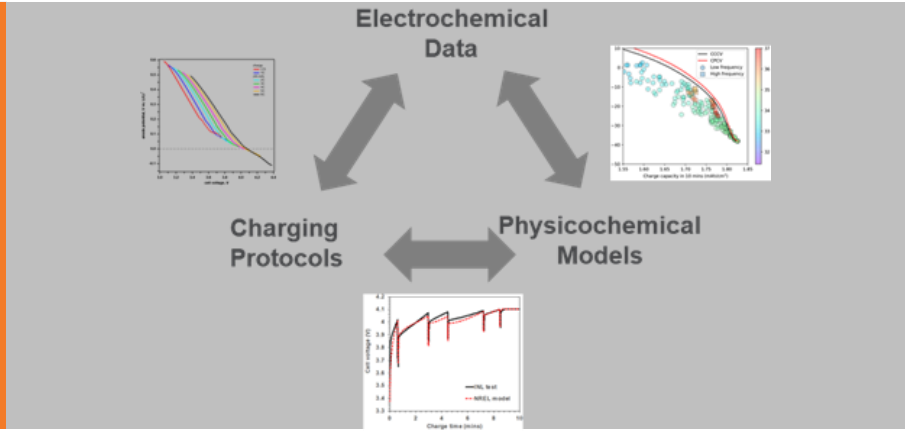
XCEL
eXtreme Fast Charge Cell Evaluation of Lithium-ion Batteries

BAT462: AGING AND THE ROLE OF FAST-CHARGE PROTOCOLS



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June 22, 2021
Web Conference

INL/CON-21-62771

OVERVIEW

Timeline

- Start: October 1, 2017
- End: September 30, 2021
- Percent Complete: 94%

Budget

- Funding for FY20 – $5.6M

Barriers

- Cell degradation during fast charge
- Low energy density and high cost of fast charge cells

Partners

- Argonne National Laboratory
- Idaho National Laboratory
- Lawrence Berkeley National Lab
- National Renewable Energy Laboratory
- SLAC National Accelerator Lab
- Oak Ridge National Lab
- Close collaboration with Behind-the-meter-storage (bat442), Machine Learning for Accelerated Life Prediction and Cell Design (bat492), and Direct XFC (elt257) and RECHARGE (elt202)

RELEVANCE

Impact:

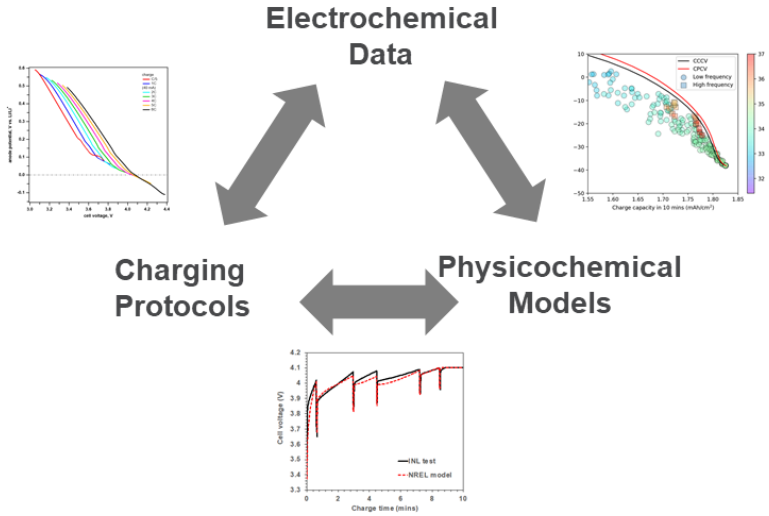
Understanding the impact of new charge protocols impacted by lack of clear methods and incomplete information

Enhanced knowledge can be gained by aligning key electrochemical data and physicochemical models to understand role of new protocols

Developed framework can be readily transitioned to other chemistries and cell designs

Objective

Aligning Models and Electrochemical Data to Enhance Understanding and Advance New Charge Protocols and Methods for Understanding Performance Fade



TASK MILESTONES

Milestone	Due	Status
Refine predictive aging impacts to identify LLI from early cycling to provide charge protocol feedback (INL) Develop model that provides thermal boundaries during charge and discharge for 2 scaled thermal management systems (NREL)	12/31/20	Complete
Refine effective cathode loss and impedance rise for fast charge conditions for 811 and modified electrode compositions wrt to refined charge protocols (INL, ANL) Develop test methods and fixture that can adjust T during cycling cells and initiate testing to provide refined understanding on aging associated with rapid temperature swings associated with XFC (ANL, NREL)	3/31/21	Complete
Develop final deliverable charge profile which reduces NMC811 aging and LLI cascade (INL, NREL)	6/30/21	In process
Initiate performance assessment of FY21 Hero Cell (INL)	9/30/21	In process
Finalize review on charge protocols, needs and impacts	9/30/21	In process

APPROACH

Understand the impact of different charge protocols

- Understand how protocols scale from cell to full pack for infrastructure needs
- Refine physicochemical models to evaluate new protocols
- Transition protocols from model to electrochemical validation

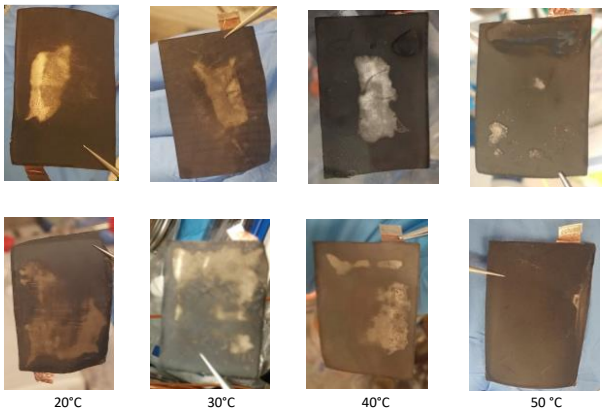
Identify key barriers as different charge and cell design conditions are used

- Use single layer pouch cells and 3-electrode cells to develop new protocols which limit aging and adjust for cell design

Reduce the time needed to determine failure mode

EFFECT OF ELECTRODE POROSITY AND TEMPERATURE ON LITHIUM PLATING

- If the local potential at graphite surface is less than 0 V (vs. Li/Li^+), lithium can plate. High current densities may exacerbate the likelihood of plating. An engineering solution to decrease local current density is to increase the surface area of the graphite electrode by decreasing porosity
- Cycle two types of electrodes, high (47%) and low (26%) porosities, in the temperature range of 20 to 50°C using 6-C charge and C/2 discharge currents



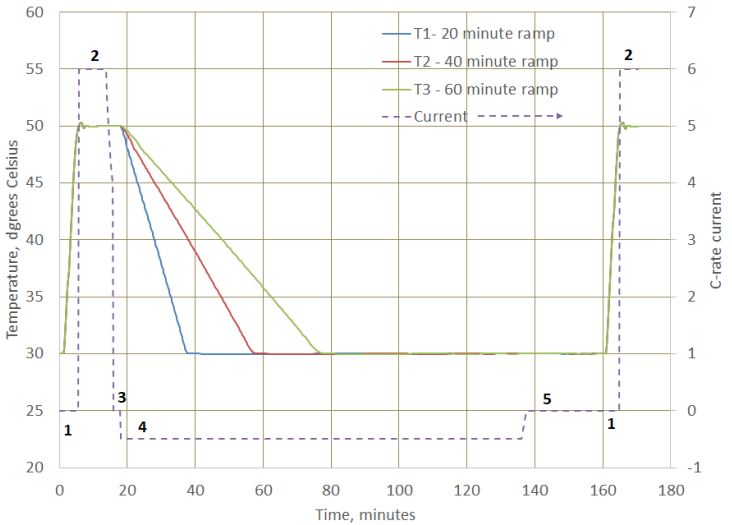
- Temperature had a larger effect on lithium plating than porosity

David C. Robertson, et al, Energy Technology, (2020) 2000666

THERMAL RAMP XCEL TESTING

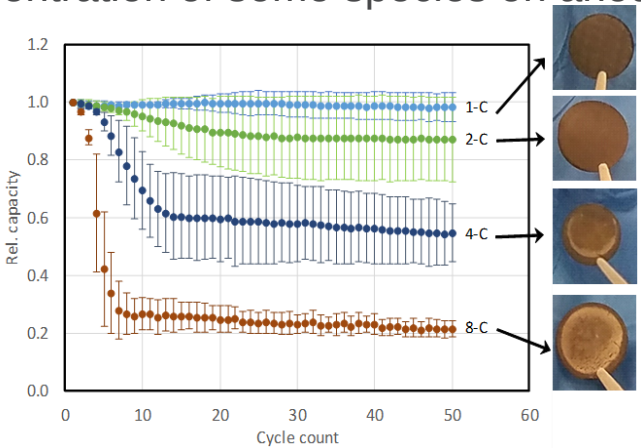
Investigating 6C CCCV charging at 50°C and various cooling rates back to 30°C during discharge

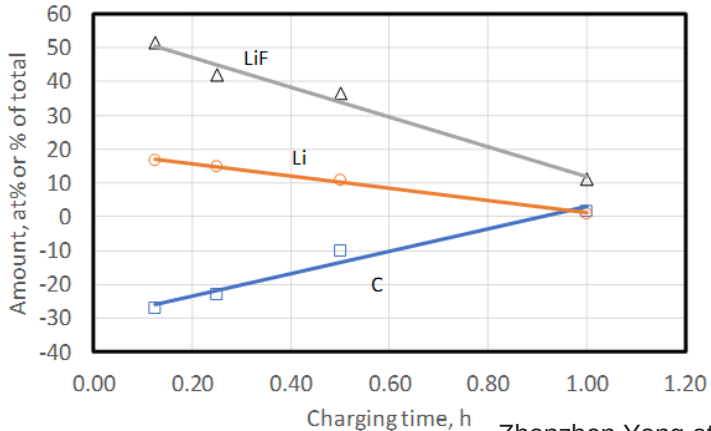
- 9 NMC811/1506T cells with Gen 2 electrolyte and 9 with B26 electrolyte under test at 3 thermal ramp rates during C/2 discharge (20, 40 and 60 minutes) (3 each)
- Different ramp rates intended to simulate different size thermal management systems
  - 1. 3-4 minute heat
  - 2. 10 minute 6C CC-CV
  - 3. 2 minute rest
  - 4. C/2 Discharge to Vmin
  - 5. 15 minute rest



EFFECT OF EXTREME FAST CHARGING ON ELECTROLYTE COMPOSITION AND ANODE SURFACE

- The changes in the electrolyte and anode surface can impact the behavior of the electrode and cell. In principle, i^2R heating at high charge rates and/or presence of large amount of plated lithium metal could be cause of these changes – Gen2
- In coin cells, we found no changes in the electrolyte which correlated with charge rate
- The concentration of some species on anode surface were sensitive to rate

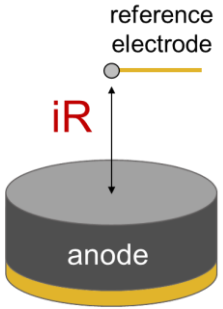




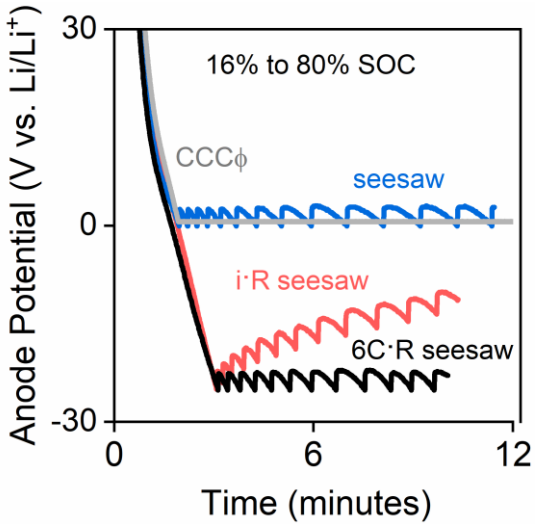
Zhenzhen Yang et al Energy Technology, (2020) 2000696

FAST CHARGING PROTOCOLS INFORMED BY A REFERENCE ELECTRODE (RE)

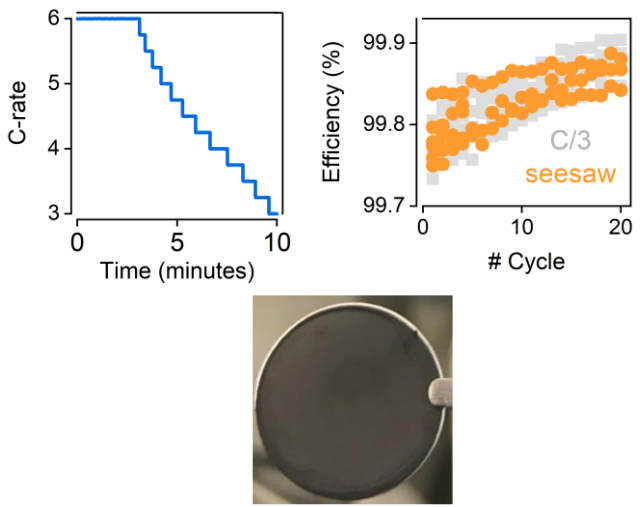
Correcting RE errors helps estimate the practical limits of charging



The finite distance between the RE and the anode leads to *measured* potentials differing from *true* values by ~iR

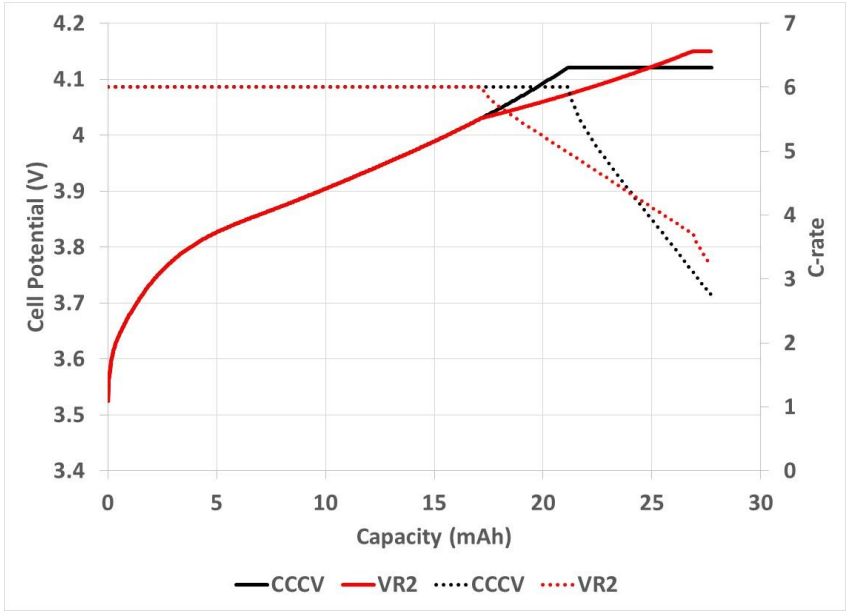


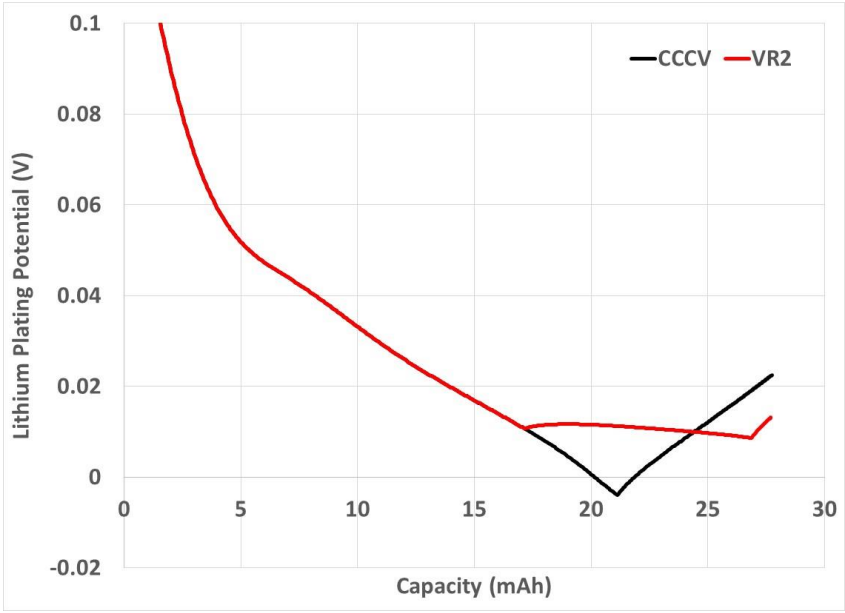
In our 3 electrode cells, **iR = 4.4 mV/C-rate**. Hence, cells can be charged at 6C until the measured potential is -25 mV vs. Li/Li+ with minimal risk of Li plating



A fast charging protocol built using a RE was able to repeatedly charge cells from 16 to 80% SOC in **under 10 minutes with no Li plating**. Charging faster than that would likely expose the anode to plating conditions

RAMP PROTOCOL MODIFIED TO ACHIEVE 90% 10-MINUTE ACCEPTANCE





- Both protocols obtain 27.7 mAh (89.5%) in 10 minutes
- Ramp protocol raises lithium plating potential by roughly 15 mV to provide buffer
- Model predicts charge acceptance with same plating buffer using CCCV is limited to 80%

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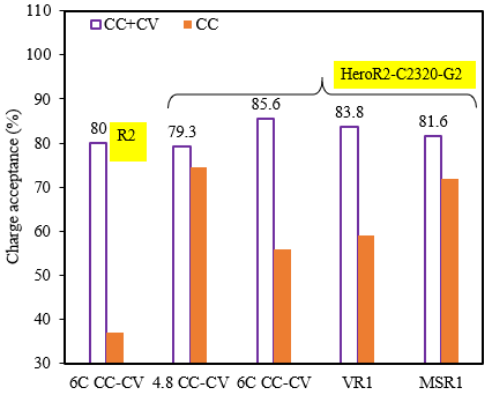
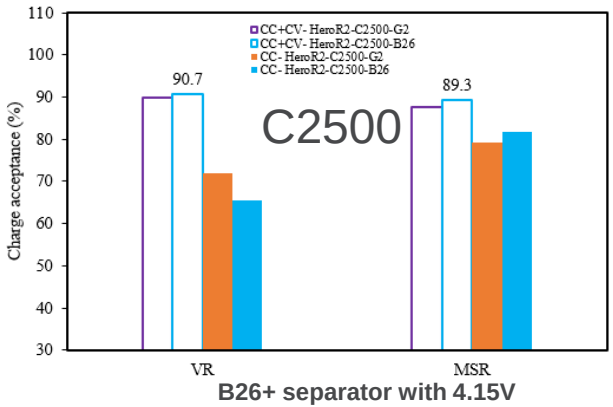
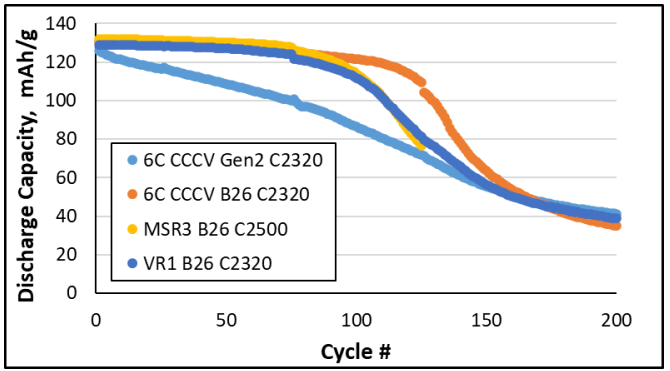
Mai, Colclasure, Smith. *J. Electrochem. Soc.* **167**:080517 (2020)

BOL CHARACTERISTIC- CHARGE ACCEPTANCE AND EARLY CYCLING







- VR1 & MSR1 Designed for ~83% charge acceptance
- VR2, 3 and MSR 2,3 designed for ~90% charge acceptance
- During early cycling no distinct indication of Li plating for top performing cells (methods described in Bat492)
- **Compared to 2018 R1 electrodes:**
  - **3x more energy accepted prior to CV portions of charge**
  - **Over 25% total increase in energy**

*Charge acceptance is scaled by BOL C/1 discharge capacity.

Chen et al., *Cell Reports Physical Science*, ***2021***, *2(3)*, 100352

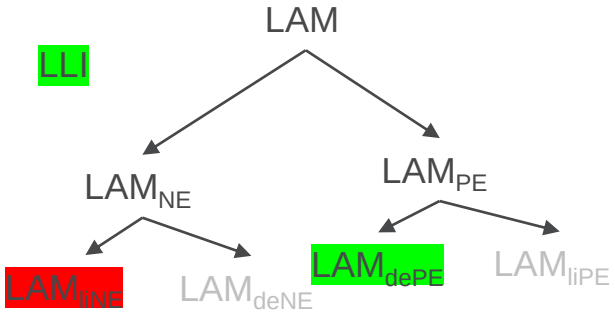
UNDERSTANDING RAPID CAPACITY FADE

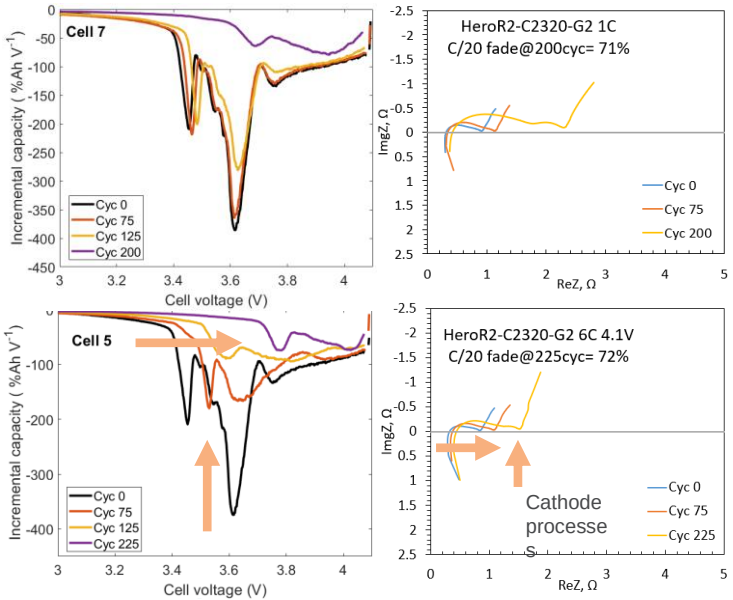
- Early cycling shows no signs of Li plating
- Using decision tree framework were able to directly analyze results
- Electrochemical analysis suggested loss of negative electrode material preceded Li plating – confirmed as delamination
- Modified future electrode preparation

Distinct LAM in NE drove N:P<1 causing Li plating-

A convoluted aging scenario mainly triggered by NE electrode (most likely delamination)

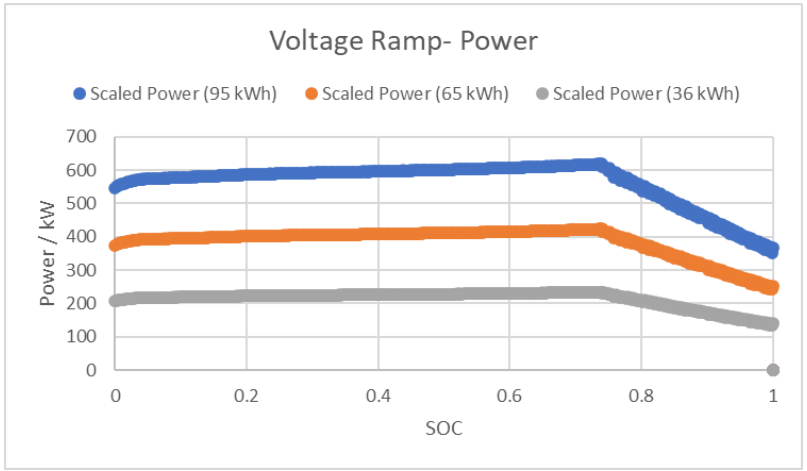
Failure modes

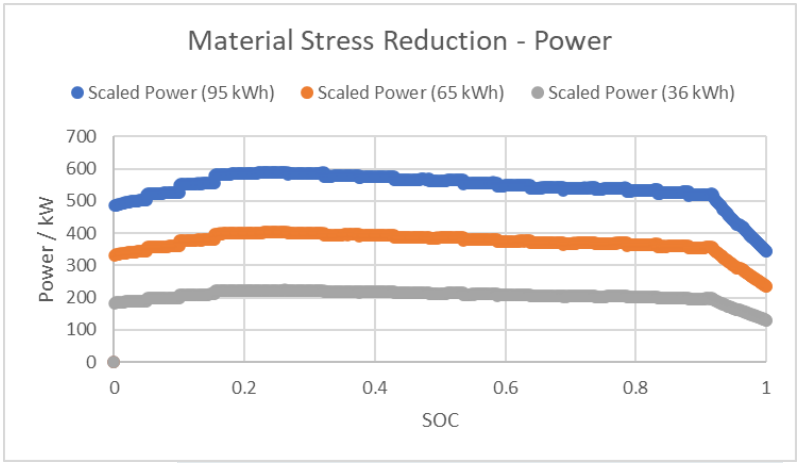




Tanvir R. Tanim et al *J. Electrochem Soc* **2019**. 166 A1926
Chen et al., *Cell Reports Physical Science*, ***2021***, *2(3)*, 100352

SCALED PROFILES – IMPACTS ON EV INFRASTRUCTURE NEEDS AND PLANNING





- Scaled XCEL profiles using vehicle scenarios developed as part of the DirectXFC Project (elt257)
- As protocol and cell advances have progressed max power has reduced from ~ 1 MW to 600 kW for a Gen3 SUV

	Usable battery Capacity (kWh)	kWh Charge (200 mi/10 min)
SUV Gen 3 (10 Min)	142.5	95
XFC Car Gen 3 (10 Min)	97.5	65
XFC Car Gen 3 (short range) - 10 min	45	36
XFC SUV Gen 2 (350 kW)	142.5	95
XFC Car Gen 2 (300 kW)	97.5	65
BEV Car Gen 2 (150 kW)	45	36

RESPONSE TO PREVIOUS YEARS REVIEWS

- Not reviewed in Fiscal Year 2020

COLLABORATION ACROSS LABS AND UNIVERSITIES

Argonne NATIONAL LABORATORY

Cell and electrode design and building, performance characterization, post-test, cell and atomistic modeling, cost modeling

INL Idaho National Laboratory

Performance characterization, failure analysis, electrolyte modeling and characterization, Li detection, charging protocols

BERKELEY LAB

Li detection, electrode architecture, diagnostics

NREL NATIONAL RENEWABLE ENERGY LABORATORY

Thermal characterization, life modeling, micro and macro scale modeling, electrolyte modeling and characterization

OAK RIDGE National Laboratory

Detailed Li plating kinetic models, SEI modeling

SLAC NATIONAL ACCELERATOR LABORATORY

Li detection, novel separators, diagnostics

XCEL eXtreme Fast Charge Cell Evaluation of Lithium-Ion Batteries

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BYU ESRF UCL

REMAINING CHALLENGES AND BARRIERS

- Transport is limited by cell design and materials – shifts in both can impact ultimate optimized protocol
  - Focus on tool development which can be broadly applied
  - Continue to cascade to higher energy designs and multi-cell architectures
- Protocols developed at the beginning of life may not be best suited for near end of life use and for charging not starting at 0% SOC.
  - Expand efforts on adaptive protocols
  - Modify protocols based on starting at mid-SOCs.
- Continued expansion of early failure mode characterization
- Understanding aging and implications of fast charge when not starting from 0% SOC

PROPOSED FUTURE RESEARCH

- Continue to expand fundamental understanding of charge protocols and how adaptation can impact optimization of protocol, life and performance
  - Adaptive protocols that minimize aging including both negative and positive electrodes
  - Mid-range SOC charging
  - Revision of protocol with continued advances in electrode and electrolyte design
- Continued coordination with Grid & Infrastructure and Behind-the-Meter Storage Projects

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

SUMMARY

- Developed charge protocols which increase charge acceptance to 80-90% in 10 min for a moderate loading cell.
  - Protocols minimize impact from Li plating by relaxing current as negative electrode approaches 0V.
  - Evaluated and refined in both single layer pouch cells and three-electrode cells.
- Developed methods to more directly understand impact from elevated temperature and thermal cycling during charging.
- Advanced understanding of cell-level aging including isolating LLI from SEI growth vs Li plating from early cycling and how electrolyte composition changes with aging
- Scaled profiles to understand how modifications made at the cell level will transfer to infrastructure and vehicle needs.

CONTRIBUTORS AND ACKNOWLEDGEMENTS

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eXtreme Fast Charge Cell Evaluation of Lithium-ion Batteries

RELEVANT PUBLICATIONS

1. M.T.F. Rodrigues, S.-B. Son, A.M. Colclasure, I.A. Shkrob, S.E. Trask, I.D. Bloom, D.P. Abraham, "How Fast Can a Li-Ion Battery Be Charged? Determination of Limiting Fast Charging Conditions", ACS Appl. Energy Mater. 4, 1063 (2021). DOI:0.1021/acsaem.0c03114
2. I.A. Shkrob, M.T.F. Rodrigues, D.P. Abraham, "Fast Charging of Li-Ion Cells: Part V. Design and Demonstration of Protocols to Avoid Li-Plating", J. Electrochem. Soc. 168 010512 (2021). DOI: 10.1149/1945-7111/abd609
3. M.T.F. Rodrigues, I.A. Shkrob, A.M. Colclasure, D.P. Abraham, "Fast Charging of Li-Ion Cells: Part IV. Temperature Effects and "Safe Lines" to Avoid Lithium Plating", J. Electrochem. Soc. 167 130508 (2020). DOI: 10.1149/ 1945-7111/abb70d
4. A. Raj, M.-T.F. Rodrigues, D.P. Abraham, "Rate-Dependent Aging Resulting from Fast Charging of Li-Ion Cells", J. Electrochem. Soc. 167, 120517 (2020). DOI: 10.1149/ 1945-7111/abace9
5. D.W Dees, M.-T.F. Rodrigues, K. Kalaga, S.E Trask, I. Shkrob, D.P Abraham, A. Jansen, "Apparent Increasing Lithium Diffusion Coefficient with Applied Current in Graphite", J. Electrochem. Soc. 167, 120528 (2020). DOI: 10.1149/1945-7111/ abaf9f
6. P.R. Chinnam, A.M. Colclasure, B.R. Chen, T.R. Tanim, E.J. Dufek, K. Smith, M.C. Evans, A.R. Dunlop, S.E. Trask, B.J. Polzin, A.N. Jansen, "Fast charging aging considerations: Incorporation and alignment of cell design and material degradation pathways" *submitted*
7. B.R. Chen, M.R. Kunz, T.R. Tanim, E.J. Dufek "A machine learning framework for early detection of lithium plating combining multiple physics-based electrochemical signatures" Cell Reports Physical Science 2(3), 100352 (2021), DOI:10.1016/j.xcrp.2021.100352
8. Z. Yang, J. W. Morrissette, Q. Meisner, S.B. Son, S.E. Trask, Y. Tsai, S. Lopykinski, S. Naik, I. Bloom, "Extreme fast-charging of lithium-ion cells: Effect on Anode and Electrolyte" Energy Technology, 2000696 (2020), DOI: 10.1002/ente.202000696
9. D.C. Robertson, L. Flores, A.R. Dunlop, S.E. Trask, F.L.E. Usseglio-Viretta, A.M. Colclasure, Z. Yang, I. Bloom, "Effect of anode porosity and temperature on the performance and lithium plating during fast-charging of lithium-ion cells" Energy Technology, 2000666 (2020), DOI:10.1002/ente.20200066

CONSTRUCTING AN ML ALGORITHM THAT SEPARATES LI-PLATING AND SEI

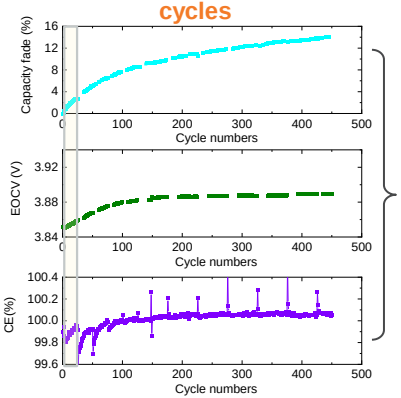
Physically meaningful electrochemical signatures

Failure mode classification using 25 cycles

Capacity fade

End of charge voltage

Coulombic efficiency



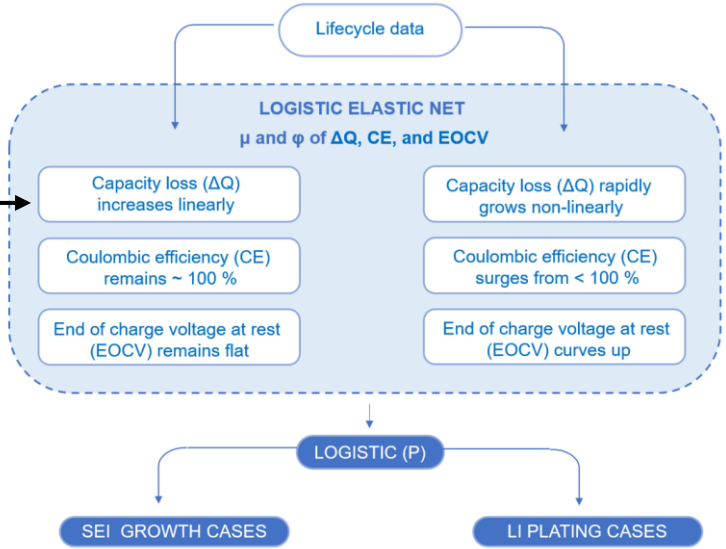
ML classification
Logistic elastic net

Mean (μ) → initial state

Autoregressive (φ) → time dependence

Correlation between coherent EC signatures

Decision making procedure (classification of cells)

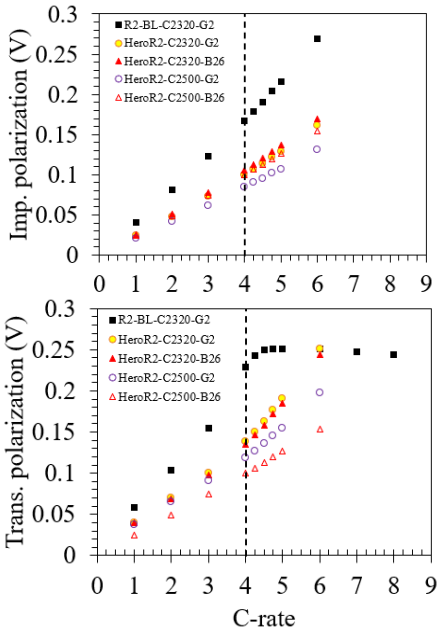


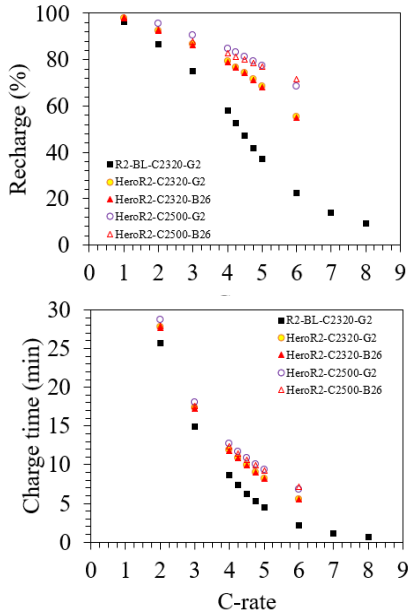
- Elastic net framework reduces time to classify SEI growth vs LLI by at least 4x
- Provides ability to track multiple signals for future adaptive work
- Coordinated analysis in collaboration with Bat492

Chen et al., *Cell Reports Physical Science*, ***2021***, *2(3)*, 100352

BOL CHARACTERISTIC- RCT

- Increased active material content in Hero Cells improved performance/lowered polarization. **No transport plateau up to 6C.**
  - Celgard2500 separator performed better w/ both electrolytes
    - Imp. Polarization is lowest in Celgard2500 + Gen2 build
    - Trans. Polarization is lowest in Celgard2500 + B26 build
- Significant improvement in CC charge acceptance. **Celgard2500 + B26 recharges 71.6%** and Celgard2500 + Gen2 recharges 68.5%.





INL #	Group	Cell design
1		
2		
3		
4	A	FF-B46A: Celgard 2320 Separator (20µm thick PP:PE:PP) and Gen2 Electrolyte
5		
6		
7		
8		
9		
10		
11		
12	B	CFF-B46B: Celgard 2320 Separator (20µm thick PP:PE:PP) and B26 Electrolyte
13		
14		
15		
16		
17		
18	C	CFF-B46C: Celgard 2500 Separator (25µm thick PP) and Gen2 Electrolyte
19		
20		
21		
22	D	CFF-B46D: Celgard 2500 Separator (25µm thick PP) and B26 Electrolyte
23		
24		

Note: All cells have the same electrode materials with R2 loading (Anode 95.83% and Cathode 96% active mat.)