RAPID ELECTROCHEMICAL DIAGNOSIS OF BATTERY HEALTH AND SAFETY FROM CELLS TO MODULES

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INTRODUCTION

- Rapid electrochemical battery state diagnostics enable…
  - Monitoring of battery state in active systems
  - Additional data points to complement digital twins
  - Rapid screening of batteries at end of ‘first life’
- DC measurements are easier than AC measurements (for now)
- For real-world implementation, the limitations rapid measurements needs to be better understood, and the impact of pulse magnitude/duration/design has not been clearly studied

CHALLENGE OF RAPID ELECTROCHEMICAL DIAGNOSTICS

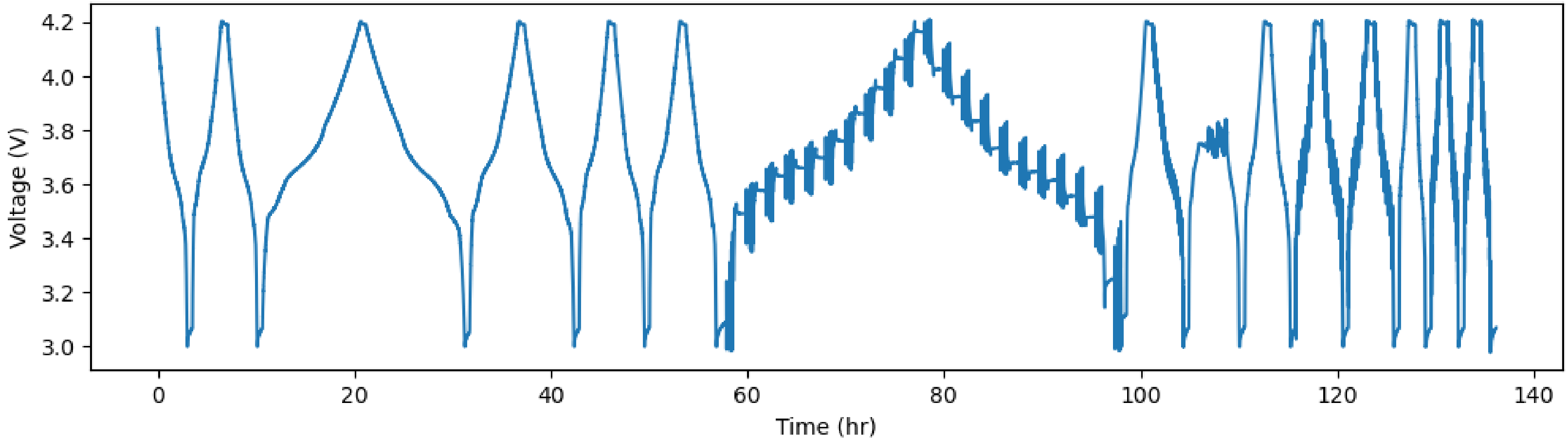
- Rapid measurements are primarily sensitive to resistance
  - For temperature prediction, this is good
    - ionic charge transfer and diffusion resistances are highly sensitive to temperature
  - For SOC and SOH prediction, resistance is not necessarily a good measure
- The evolution of resistance and capacity throughout cell lifetime is not always monotonic, leading to non-linear relationships between the two
- Resistance and capacity measures have different sensitivity to temperature and hysteresis
- Temperature, SOC, SOH, and hysteresis effects on resistance **are not independent**

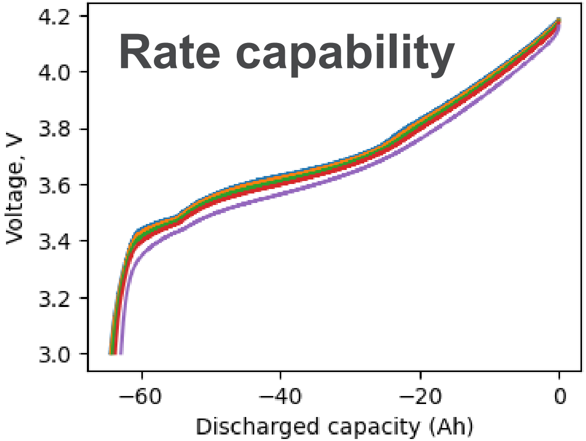
Machine-learning is an ideal tool to study this challenge.

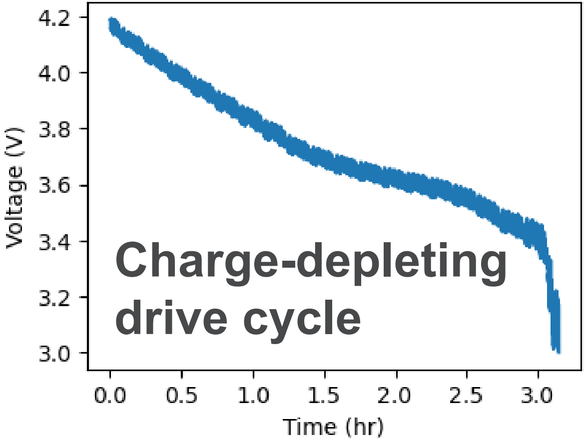
THE DATA SET

- 6-day characterization test with…
  - Charge/discharge rate capability test (C/10, C/5, C/3, P/3, C/2, 1C)
  - 3-hour charge-depleting and charge-sustaining application cycles
  - Pulses at rest and dynamic (overlaid on charge/discharge)
- 68 total measurements:
  - 24 batteries (16x 64 Ah NMC|Gr, 8x Nissan Leaf modules)
  - 3 ambient temperatures per battery
- Pulses across SOC range during charge and discharge after rests (HPPC) and ‘dynamic’ pulses overlaid on C/2 and 1C
  - ~13,000 distinct pulse measurements

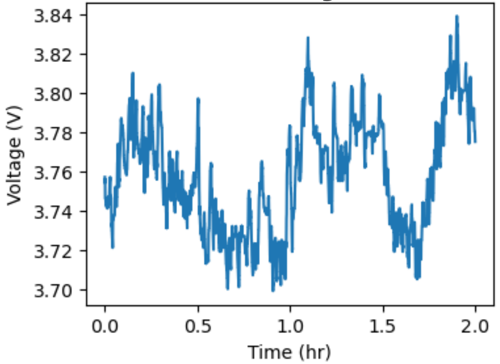
THE DATA SET





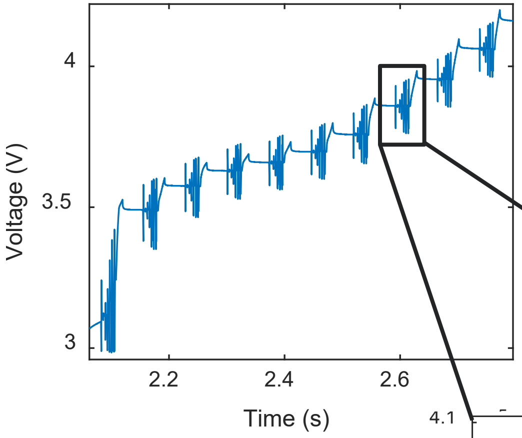


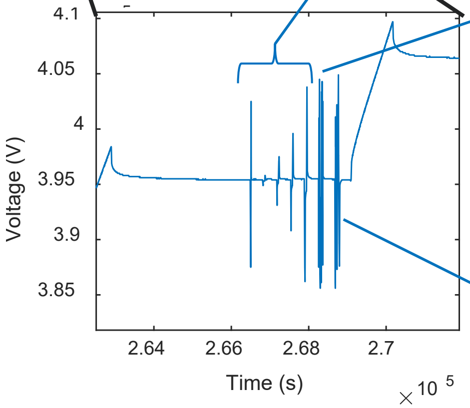
Charge-sustaining drive cycle



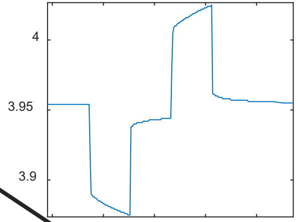
THE DATA SET

HPPC

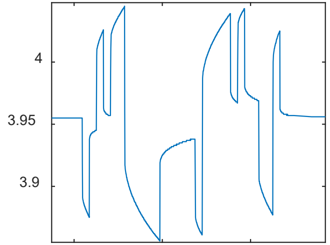




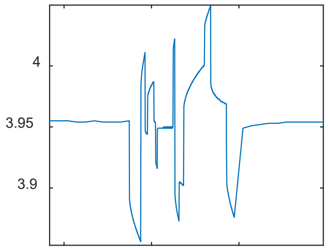
Single pulses
10s-40s-10s (USABC) and 4s-4s-4s-4s (rapid)
USABC pulses @ C/10, C/2, 1C, 2C



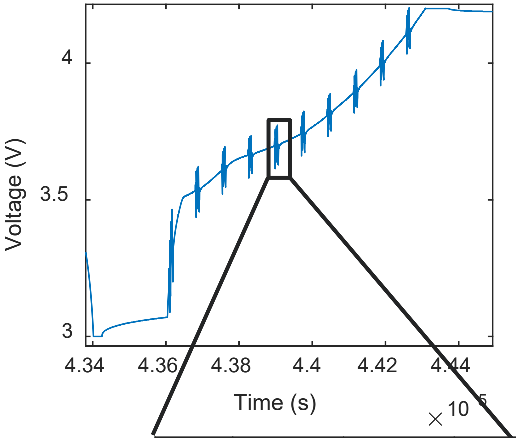
Pseudo-**R**andom **P**ulses (PsRP)
Simple pulse sequence (PsRP 1)
4s steps, [-2, 0, 2] C current, 120s

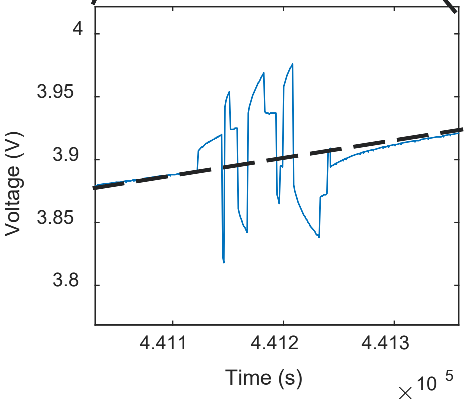


Complex pulses sequence (PsRP 2)
[1:1:15]s steps, [-2:1:2] C current, 120s



Dis/charge + PsRP





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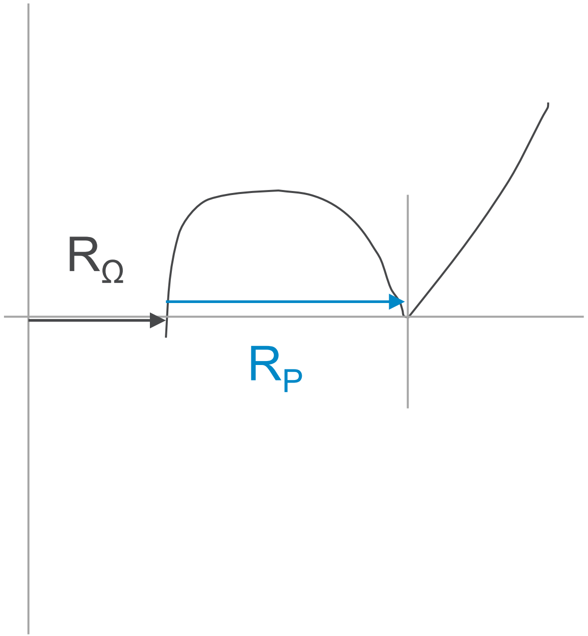
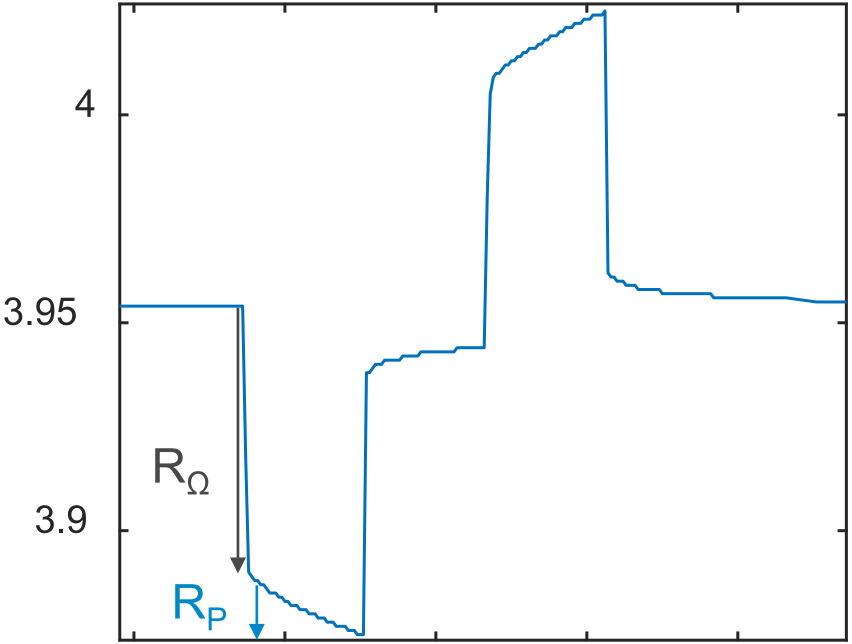
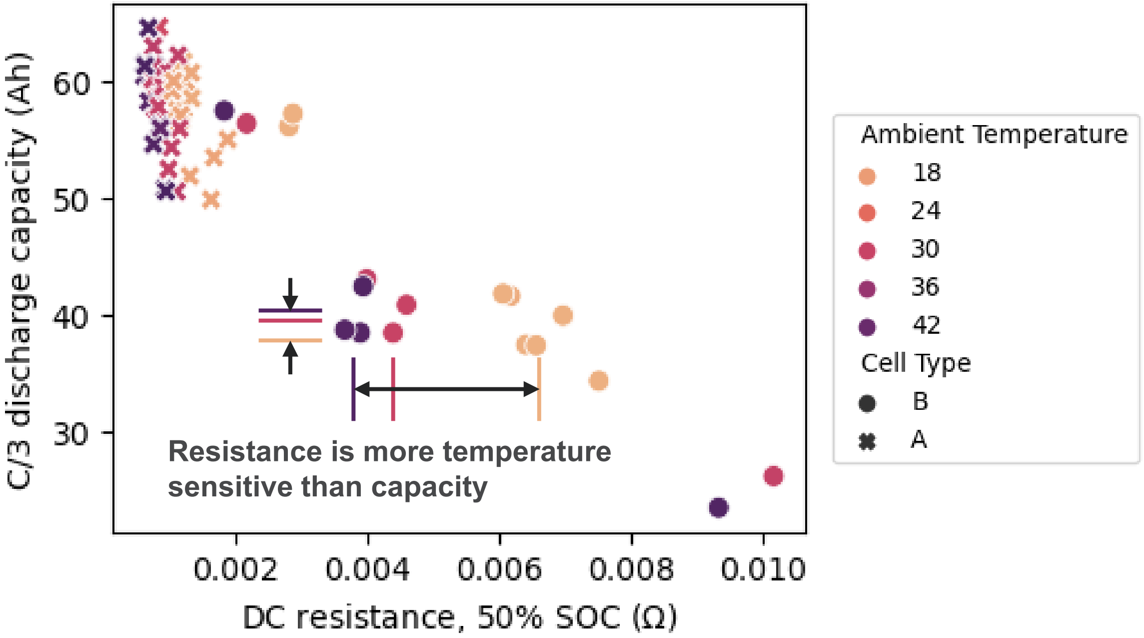
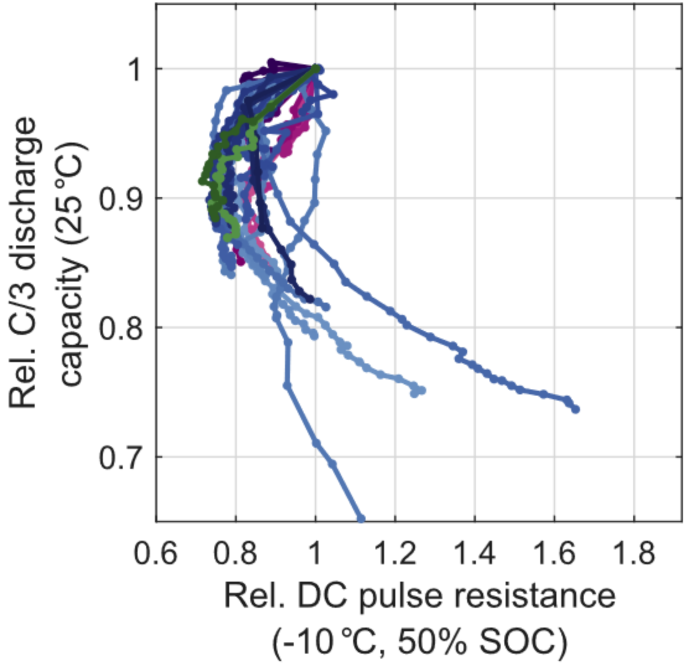




CAPACITY AND RESISTANCE ARE CORRELATED…

But the correlation is not strong, and both capacity and resistance may evolve non-monotonically throughout a cell's lifetime.





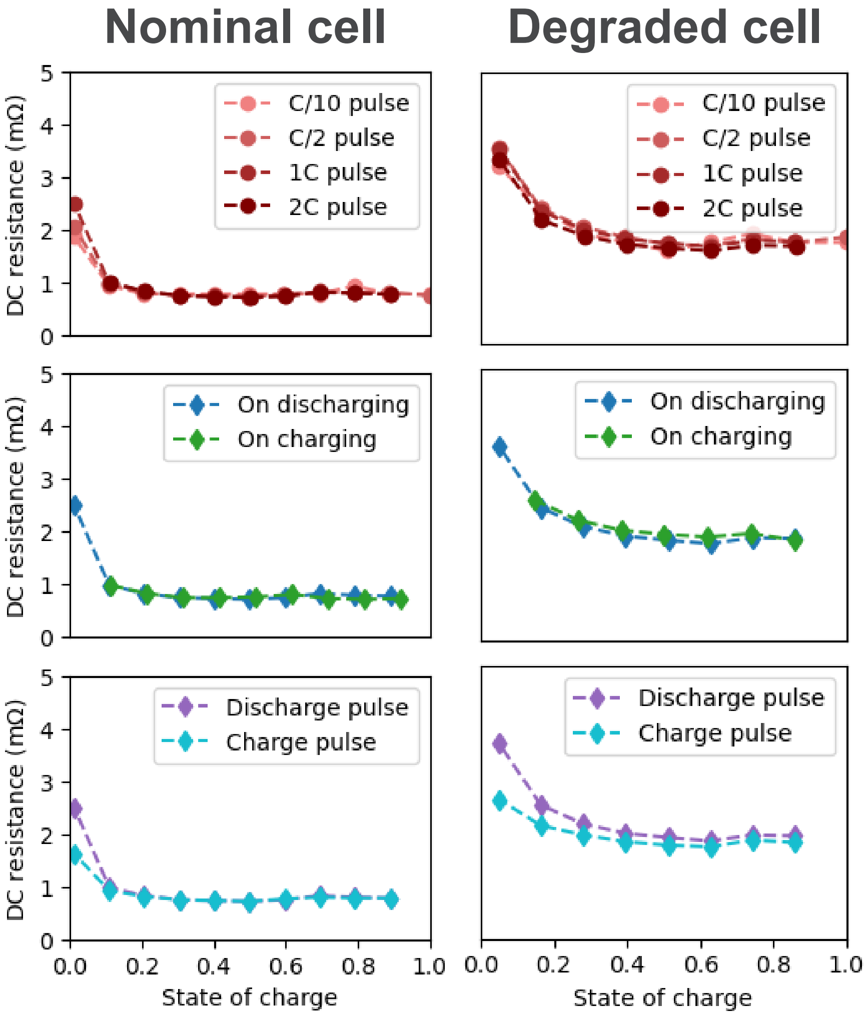
Gasper et al, *Cell Reports Phys. Sci.* 2022 3 12

PULSES ARE SENSITIVE TO EVERYTHING…

…with varying magnitude. Effects of cell state of pulse response are **not independent** (SOH impacts SOC dependence, …)

For NMC and LMO positive electrode cells, pulses show the most sensitivity to cell state at very low SOC.

Slightly more sensitivity in aged cell than nominal cell to all variables.



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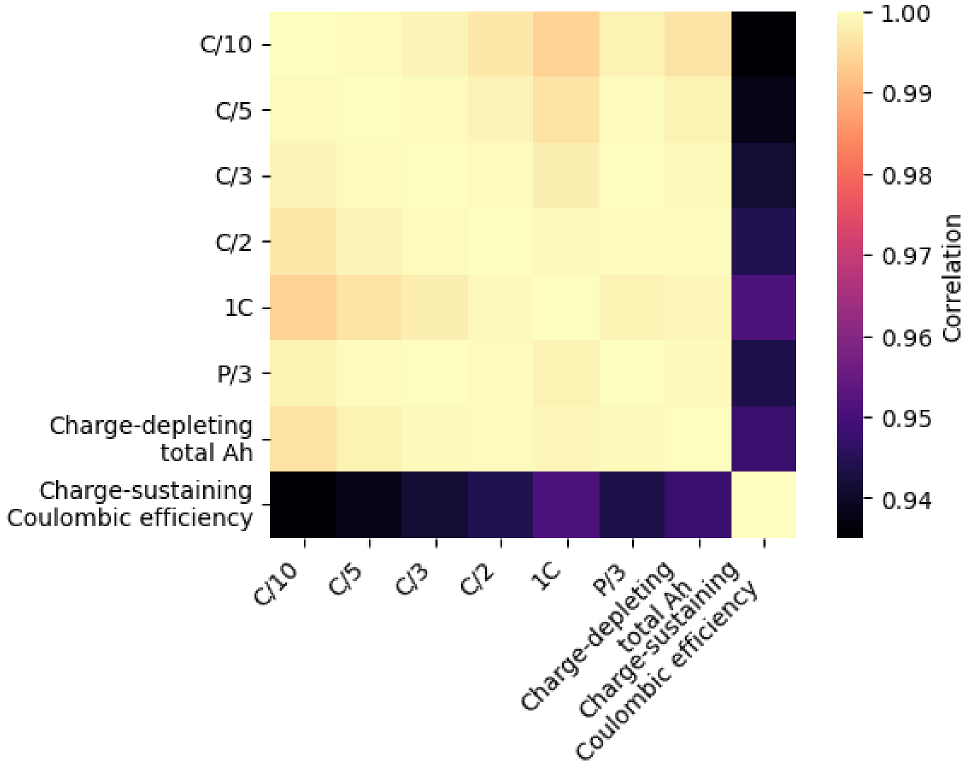
HEALTH IS MORE THAN JUST C/3 CAPACITY

Discharge capacities become less correlated as rate increases, i.e., rate-capability curves across a group of cells becomes more variable as they age.

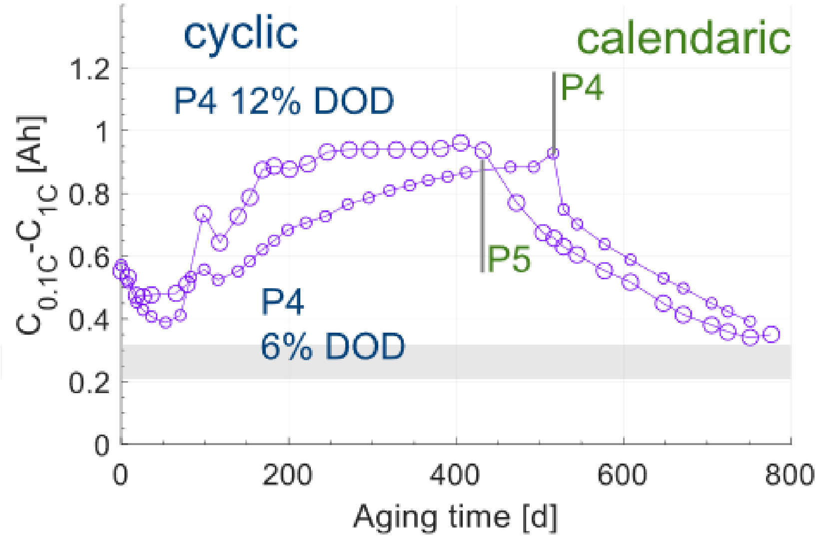
Drive cycle performance not necessarily perfectly correlated to capacity measurements.

How to quantify 'safety' for a model to predict?



HEALTH IS MORE THAN JUST C/3 CAPACITY

Prediction at ‘high’ rates is probably artificially easy as cells had rested for several months following cycling.

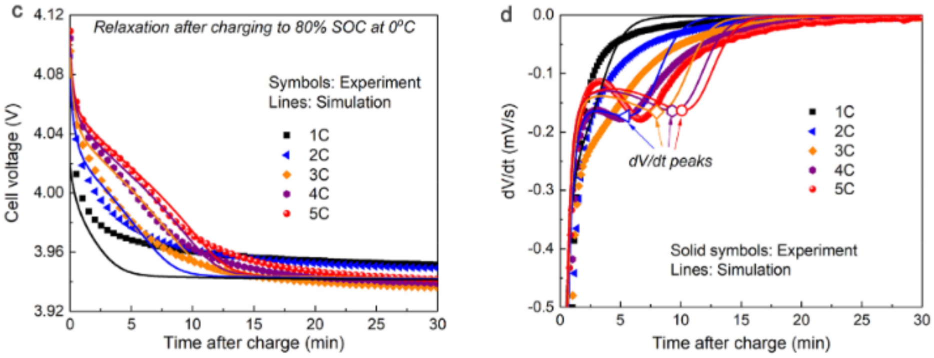


Lewerenz et al, J. Energy Storage 21 (2019) 680-690

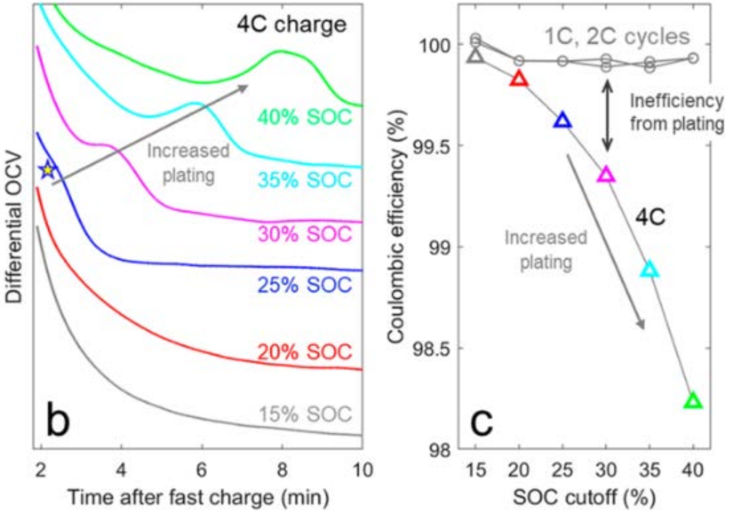
QUANTIFYING SAFETY FOR ML

Lithium plating has been shown experimentally and analytically to affect the curvature of the post-charge voltage relaxation.

https://doi.org/10.1016/j.jpowsour.2018.05.073



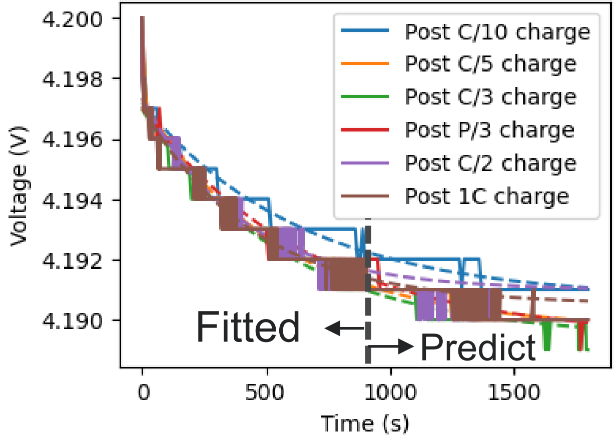
https://doi.org/10.1021/acsenergylett.0c00831



QUANTIFYING SAFETY FOR ML

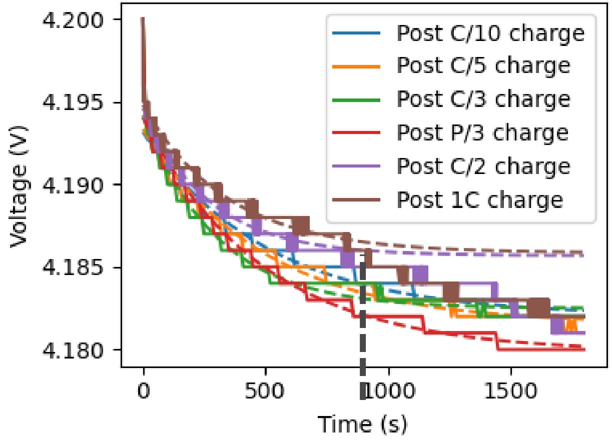
First half (15 minutes) of the voltage relaxation after charge is fit with an exponential relaxation curve; deviation from this curve from 15-30 minutes of rest is used as a simple method to quantify deviation from 'ideal' battery behavior.

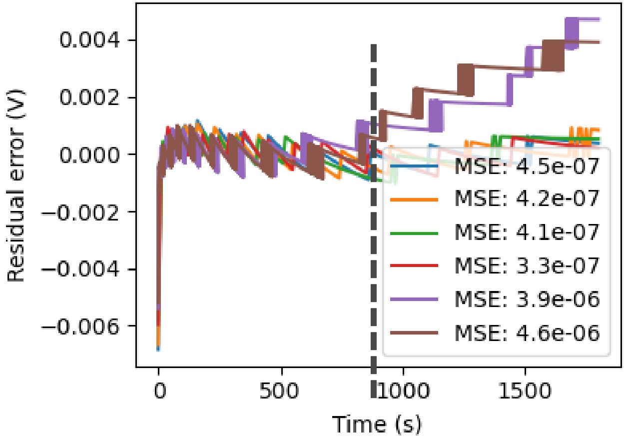
Nominal cell



Degraded cell w/ suspected Li plating

(~15% capacity loss, ~35% DCIR growth)





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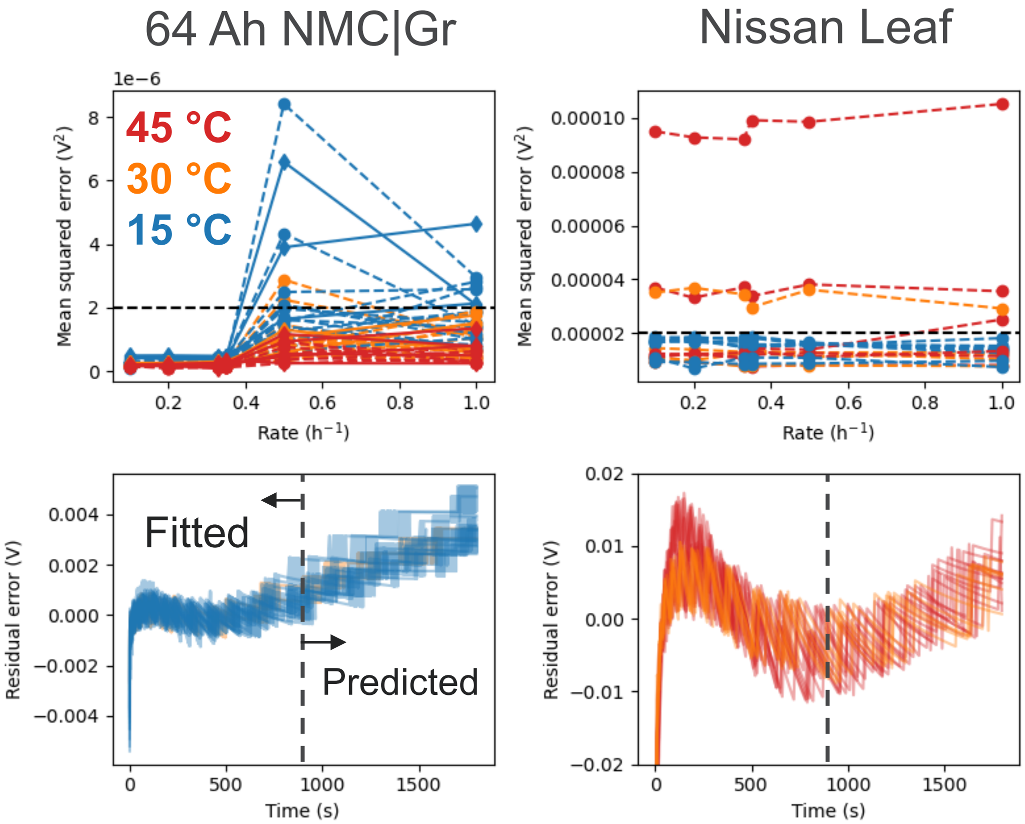
QUANTIFYING SAFETY FOR ML

NMC|Gr cells exhibit error consistent with lithium plating:

- Error increases with decreasing temperature, increasing rate
- Positive error after fit region

Nissan Leaf cells show deviation at high temperature, independent of rate, suggesting another physical root cause. Gas generation suspected.



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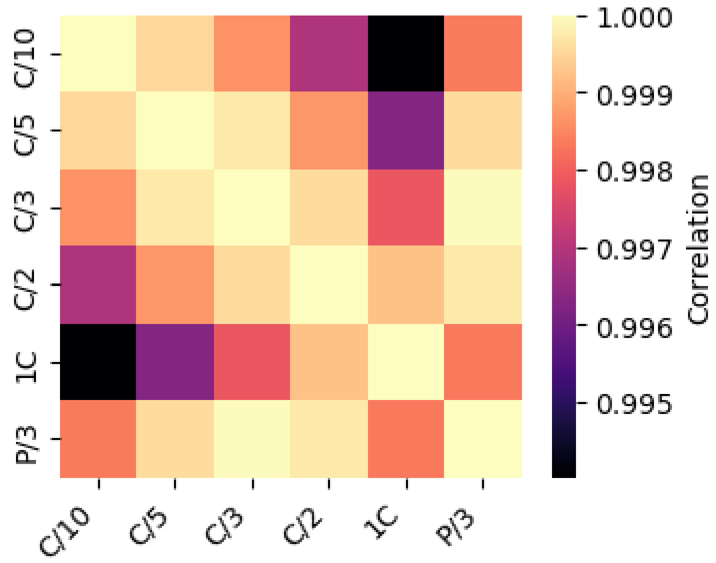
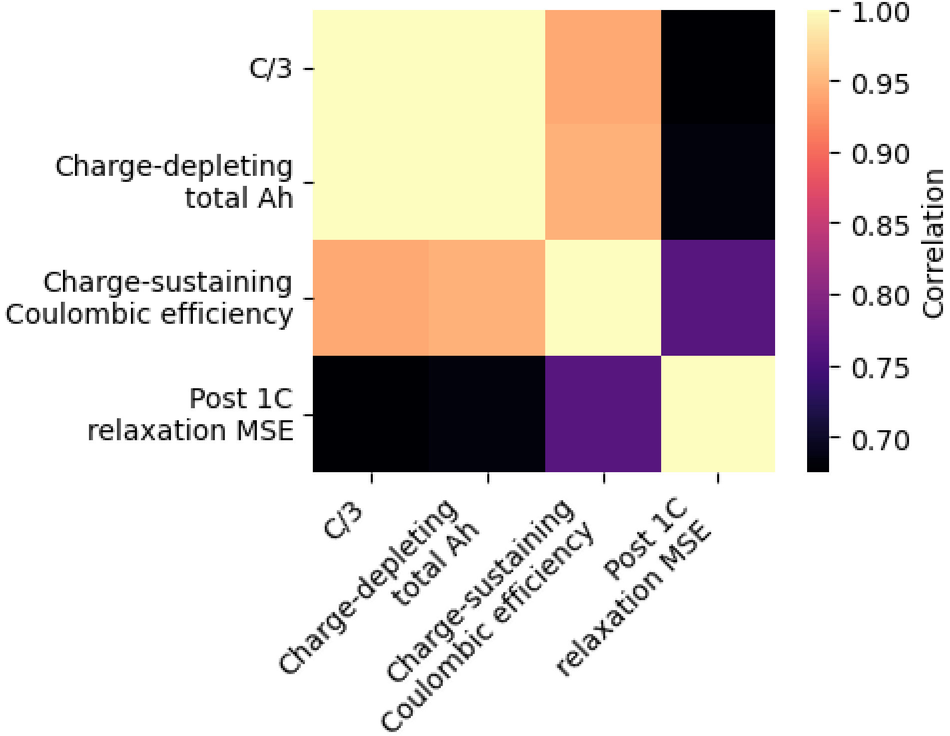




MACHINE-LEARNING APPROACH

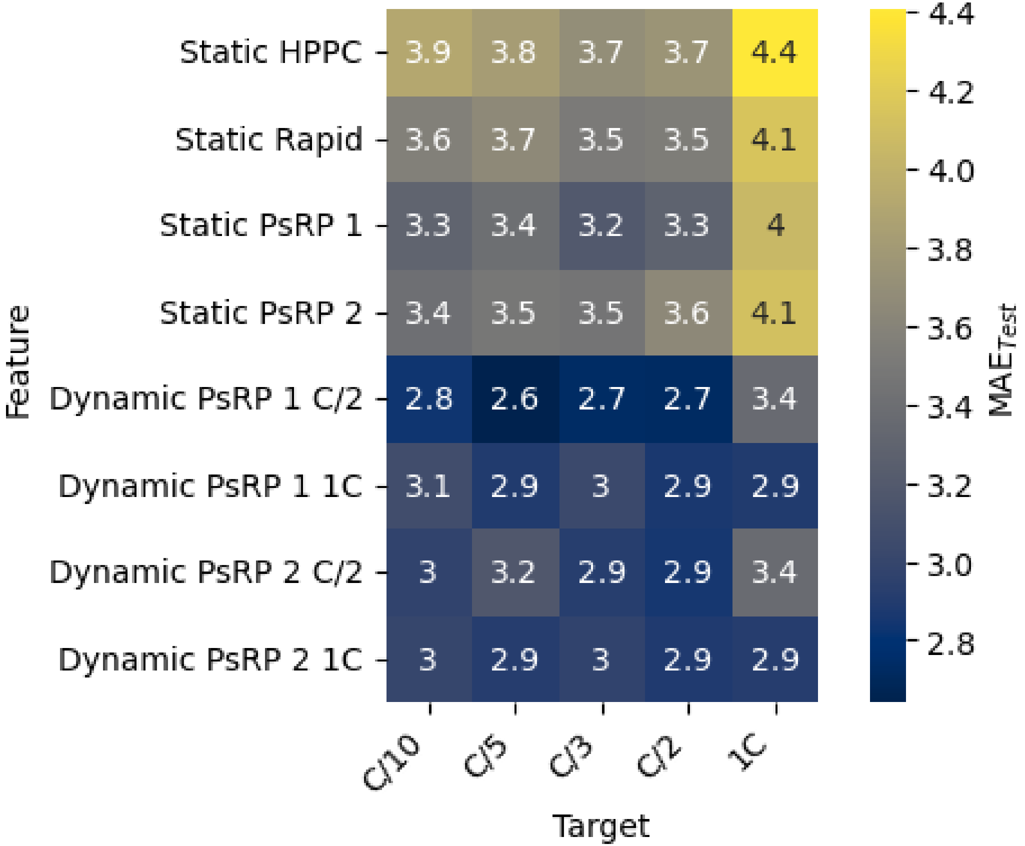
- Features: Raw voltage and current measurements from pulse sequences
  - Includes rests in between pulses
  - Feature dimensionality reduced using PCA (slightly better performance than raw data)
- Targets: SOH variables
- Model architecture: XGBoost
- Hyperparameter optimization via 50-iteration randomized grid search for XGBoost parameters and PCA components
- Data splitting: 80:20 train:test, grouped by measurement to avoid data leakage
  - Hyperparameter optimization via 5-fold grouped cross-validation
- Average results over 50 train:test splits

RESULTS - SOH

Average prediction error of ~4-6% (70-80% correlation) for all capacity values.

Pulse ‘design’ seems not to matter, but static (after rest) or dynamic (during dis/charge) does; dynamic pulses better on all targets.

1C rate predicted most accurately by 1C dynamic pulses, perhaps due to the incorporation of the charge segment into the pulse.

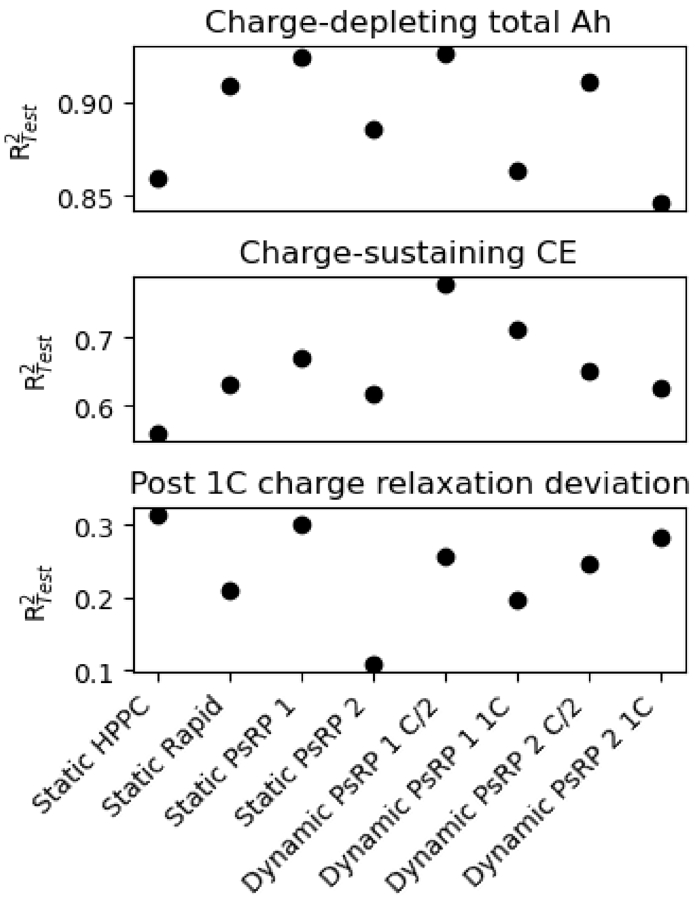


RESULTS - SOH

Charge-depleting drive cycle is ‘easy’, with the best result of any target.

Charge-sustaining coulombic efficiency target predicted with near the same accuracy as the capacity metrics; dynamic pulses perform slightly better, on average.

‘Safety’ prediction is relatively poor, despite reasonable correlation with capacity.



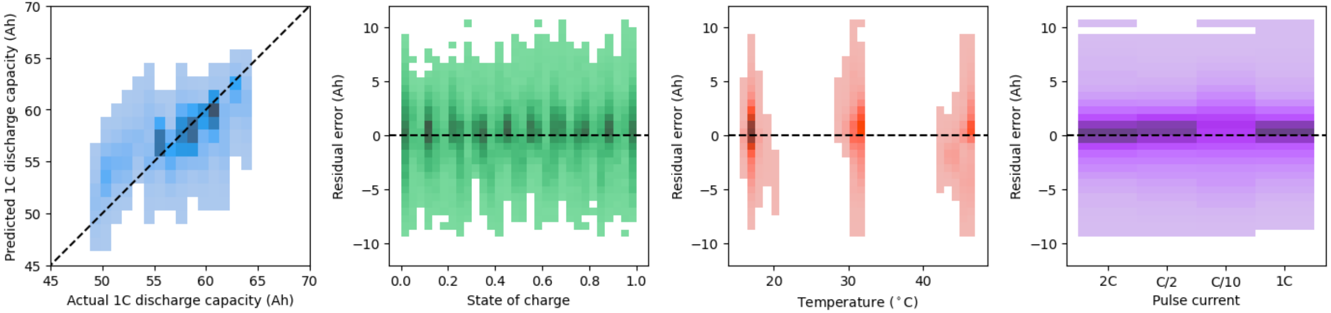
RESULTS - SOH

Models are biased to overpredict capacity at low SOH, underpredict at high SOH.

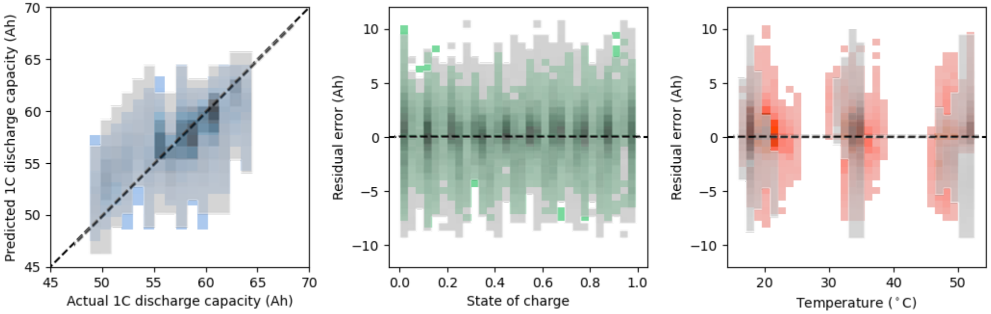
C/10 rate pulses have more error than higher rate pulses, but C/2 no different than 2C. Max error not a function of pulse current.

Dynamic PsRP model has lower max errors.

HPPC pulses → 1C capacity



PsRP 2 1C pulses → 1C capacity



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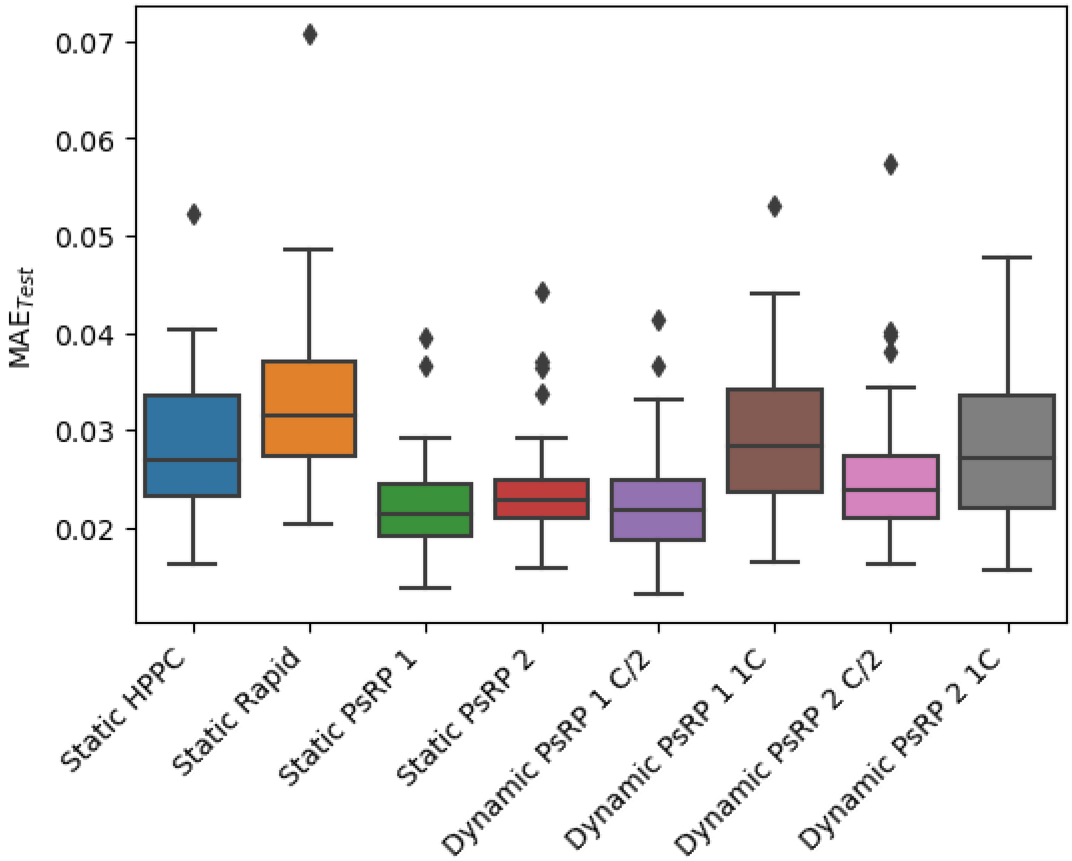
RESULTS - SOC

State-of-charge is predicted with good accuracy despite variation due to temperature, health, and hysteresis.

Unlike health prediction, dynamic pulses perform no better than static pulses.

‘Complex’ PsRP pulses perform better than simple pulses for SOC prediction.

SOC prediction is ‘easier’ than SOH prediction (higher R^2).



CONCLUDING REMARKS

- Battery state can be predicted with useful accuracy from short pulse sequences despite competing influences of temperature, SOC, SOH, and hysteresis on pulse response
  - Prediction of the ‘safety’ metric devised here is poor, more modeling effort and better ‘safety’ target variables may improve results. Quantifying safety is a critical challenge for battery system operation and recertification of cells for second-life applications
- Pulse design and implementation have small but clear impacts on prediction accuracy, suggesting more complex pulses contain more useful data for monitoring battery state than simple pulses

FY 24 PLANNED WORK

- Ford Fusion NMC111-Gr cells (BOL and field aged) characterization
  - BOL characterization complete, cells cycling for more data
- Cycling of healthy Leaf cells for more data
- Automotive grade LFP-Gr cells – extraction of CATL 160 Ah prismatic cells from Model 3 module
  - BOL characterization and then cycling
- Collaborating with CMU to receive field aged 21700 LFP-Gr A123 cells from hybrid bus pack
- Improvements to ML approach and fitting of physics-based models to data

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This work was authored by the National Renewable Energy Laboratory, operated by Alliance for Sustainable Energy, LLC, for the U.S. Department of Energy (DOE) under Contract No. DE-AC36-08GO28308. Funding provided by the U.S. Department of Energy Office of Energy Efficiency and Renewable Energy Vehicle Technologies Office. The views expressed in the article do not necessarily represent the views of the DOE or the U.S. Government. The U.S. Government retains and the publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for U.S. Government purposes.

NREL/PR-5700-87741