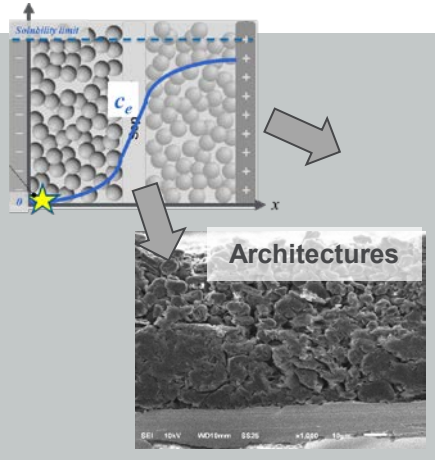
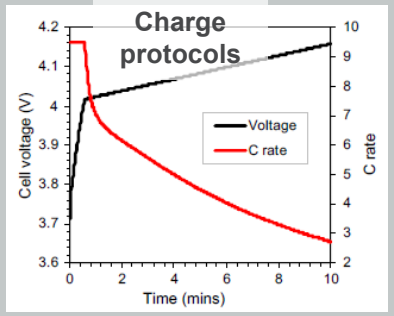
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BAT460
U.S. DEPARTMENT OF ENERGY
VEHICLE TECHNOLOGIES OFFICE ANNUAL MERIT REVIEW

XCEL
eXtreme Fast Charge Cell Evaluation of Lithium-ion Batteries

ELECTROCHEMICAL MODEL ASSESSMENT OF STRATEGIES FOR FAST CHARGE





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- **INL:** Eric Dufek, Tanvir Tanim
- **LBNL**: Marca Doeff
- **SLAC:** Mike Toney, Will Chueh, Che-Ning Yeh
- **Brigham Young University:** Dean Wheeler

U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy
VEHICLE TECHNOLOGIES OFFICE
Argonne NATIONAL LABORATORY
SLAC NATIONAL ACCELERATOR LABORATORY
NREL NATIONAL RENEWABLE ENERGY LABORATORY
BERKELEY LAB Lawrence Berkeley National Laboratory
INL Idaho National Laboratory

June 1–4, 2020
Virtual Conference

OVERVIEW

Timeline

- Start: October 1, 2017
- End: September 30, 2021
- Percent Complete: 75%

Barriers

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

Budget

- Funding for FY20 – $5.6M

Partners

- Argonne National Laboratory
- Idaho National Laboratory
- Lawrence Berkeley National Lab
- National Renewable Energy Laboratory
- SLAC National Accelerator Lab
- Oak Ridge National Lab

RELEVANCE – TRANSPORT LIMITATIONS

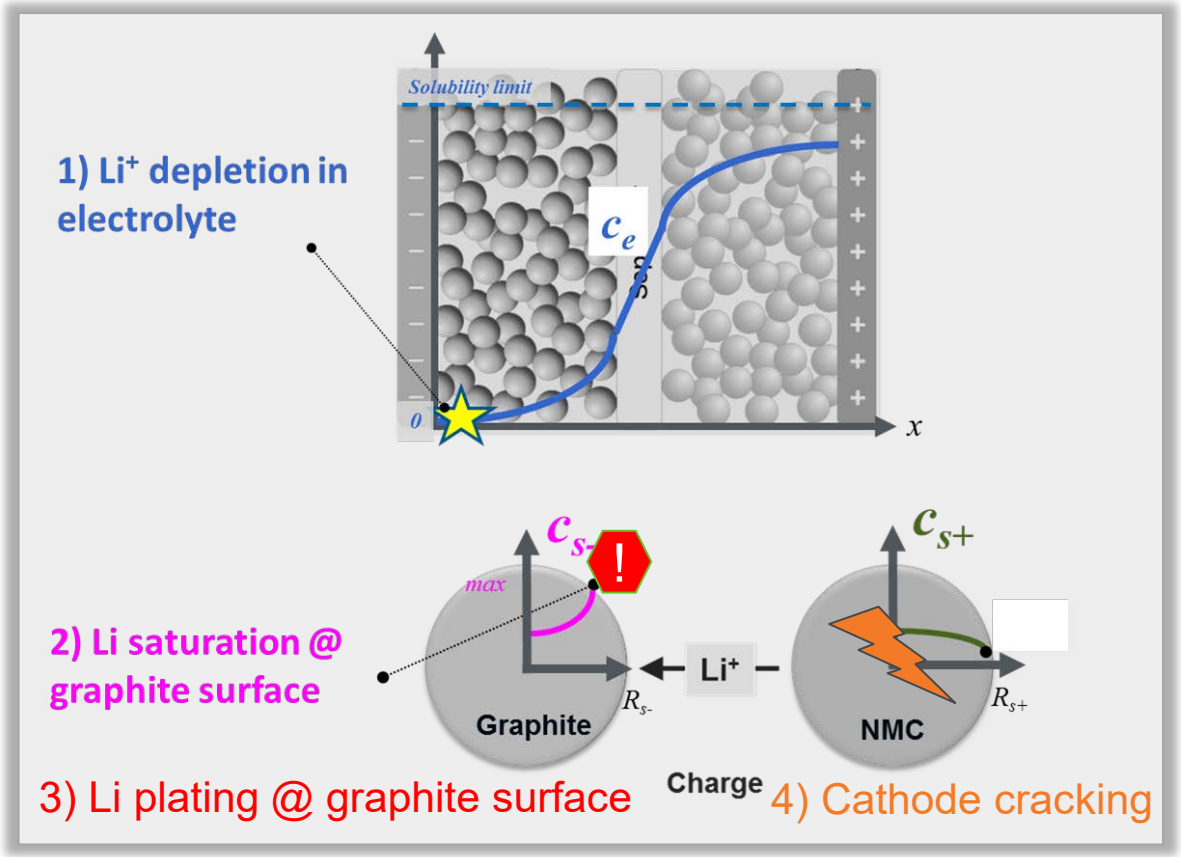
High energy density batteries have poor fast-charge acceptance

Thin
Fast charge
High cost
Low energy

Electrode thickness

Thick
Slow charge
Low cost
High energy

- Thick electrodes have low cost, high energy density, but poor fast-charge capability due to slow electrolyte lithium (Li^+) transport
- When electrolyte Li^+ ions become depleted at the back of the graphite electrode, only the front of the electrode is used
- Excessive charge rate at the front (near separator) leads to graphite saturation and Li plating.



XCEL eXtreme Fast Charge Cell Evaluation of Lithium-ion Batteries

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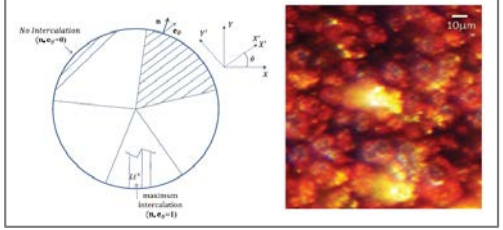
RELEVANCE – HETEROGENEITIES

Nonuniformities at all length scales cause early onset of Li plating and must be suppressed through careful design and manufacturing

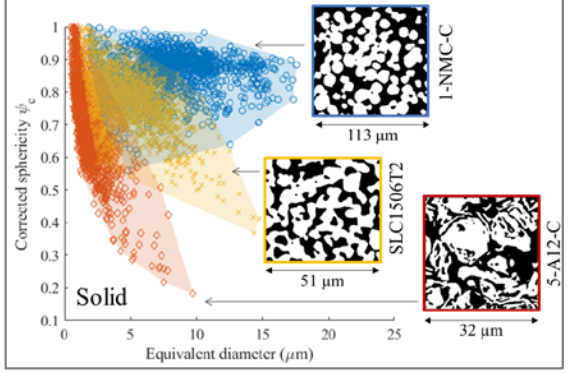
length scale		*heterogeneity*
0.1 µm	**Grain**	• Crystal anisotropy • Particle-to-particle contact
1 µm	**Particle**	• Particle size, morphology
10 µm	**Electrode thickness**	• Electrolyte transport limitation leads to plating at electrode surface
100 µm	**Electrode coating**	• Porosity/tortuosity variation
1 mm	**Edge effects**	• Anode overhang • Electrode misalignment • Electrolyte shorting
1 cm	**Cell**	• Dry out, delamination • Pressure • Temperature • Tab configuration
10 cm		

examples

ΔNucleation sites [1]



Particle morphology/size [2]



Cell-scale [3]



1. *Harris et al. JES (2012)*
2. *F. Usseglio-Viretta et al., JES, accepted*
3. *Photo: Ira Bloom, ANL*

OBJECTIVES

10- to 15-minute fast charge of high-energy, 15-year life electric vehicle battery

- Use models as feedback to experiments and provide guidance on cell-level designs/requirements/strategies to
  - Improve fast-charge acceptance
  - Suppress Li plating
- Project charge acceptance and energy density of various strategies including
  - Reduced tortuosity electrode architectures
    - Secondary pore channels, dual-layer and graded electrodes
  - Elevated temperature
  - Next generation electrolytes
  - Other: Negative/positive ratio, porosity, electrode loading

FY 2020 MILESTONES

Thrust Area	NREL Milestone	Date	Status
Anode	Identify optimal anode structures for freeze cast, laser-ablation, multi-layer coating and pore-former technologies using multi-scale models	Q2	Complete
Anode	Summary report of different anode architectures	Q4	On track
Charge protocol	Optimize charge strategies using electrochemical model	Q2	Complete
Cathode	Obtain microCT data from SLAC for crack identification	Q1	Complete
Cathode	Identify advanced algorithm framework to output grain orientations, sub-particle tortuosity, particle and grain morphologies, grain boundary distributions and crack distributions based on microCT, EBSD and other techniques.	Q3	On track
Cathode	Build models to investigate cathode particle size/distribution, orientation and coating effects on cracking. Define modeling framework, build and validate models. Investigate performance as a function of cracking	Q4	On track
Heat	Define the critical parameters that affect heat generation within a cell.	Q1	Complete
Heat	Develop 3D model capable of assessing heterogeneities, heat transport, and strategies to mitigate temperature rise under XFC conditions	Q4	On track
Heterogeneities	Select framework for lithium plating model and summarize methodology/equations	Q2	Complete
Heterogeneities	Draft journal article comparing detailed lithium plating kinetic model with experimental results from LBNL and SLAC	Q4	On track

microCT: Micro-scale X-ray computed tomography imaging
XFC: Extreme fast charging

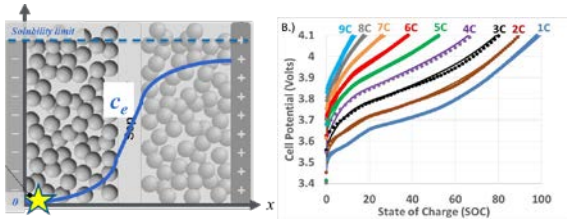
APPROACH

XCEL cell builds and experiments

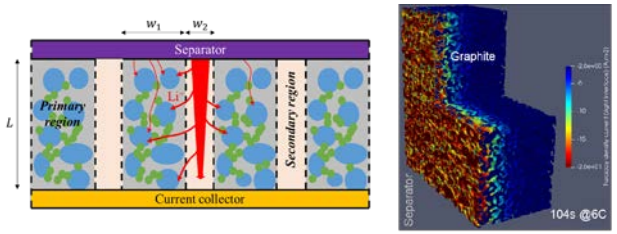
- 1.5-2.5 mAh/cm^2, 4.1V

Electrochemical models

- 1D macro-homogeneous



- 2D and 3D models



Design strategies

- 3-4 mAh/cm^2, 4.2V

Charge protocols

Heterogeneities

- Li plating avoidance

Advanced architectures

Design / experiment feedback

OUTLINE

- Electrochemical model validation
  - Round 1 and Round 2 cells (1.5 to 2.5 mAh/cm^2)
- Electrochemical model projections at higher loadings (3.0 to 4.0 mAh/cm^2)
  - What is needed to enable fast charge
- Enabling technologies
  - Electrolyte transport: Advanced electrolyte, electrode architectures, elevated temperature
  - Avoidance of Li plating: Charge protocols
- Additional design considerations
  - Suppression of heterogeneities
  - Heat management

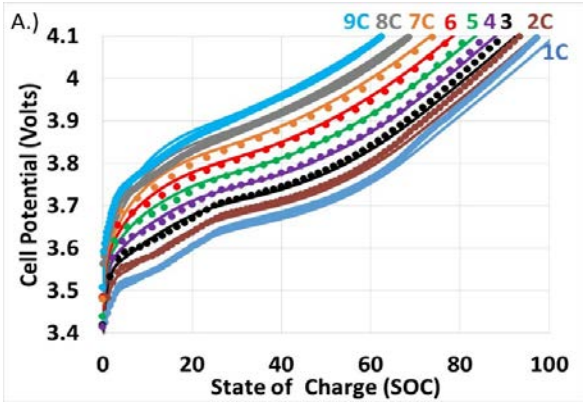
ECHEM MODEL VALIDATION

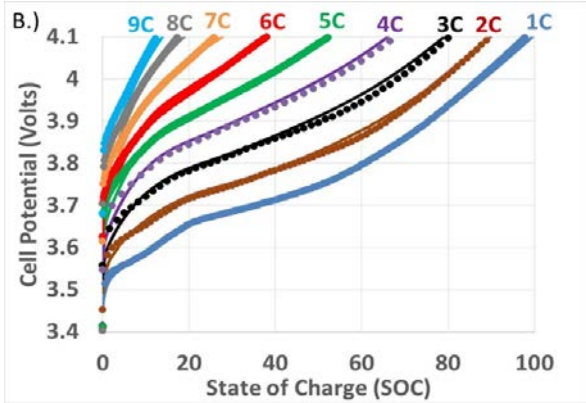
Symbols: Data
Lines: Model

1.5 mAh/cm^2 loading (40 µm) XFC capable

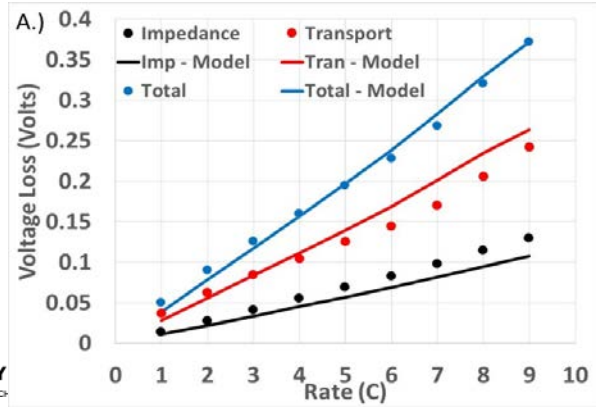
2.5 mAh/cm^2 loading (70 µm) Barely XFC capable

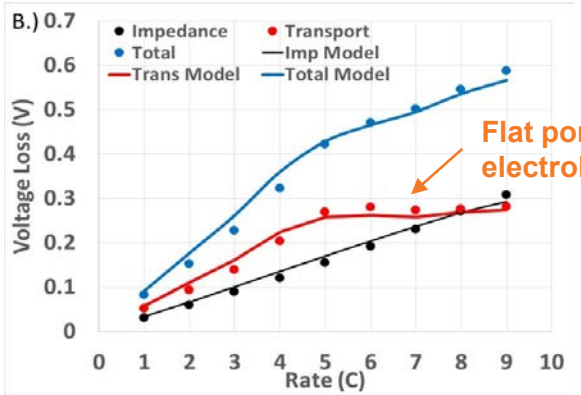
Voltage





End-of-charge relaxation





Additional validation

- In situ beamline X-ray diffraction of 100-µm electrodes resolving Li_xC_6 concentrations and Li plating across thickness
- Ex situ and teardown studies resolving plated Li
- Titration, quantifying Li plating/stripping amounts

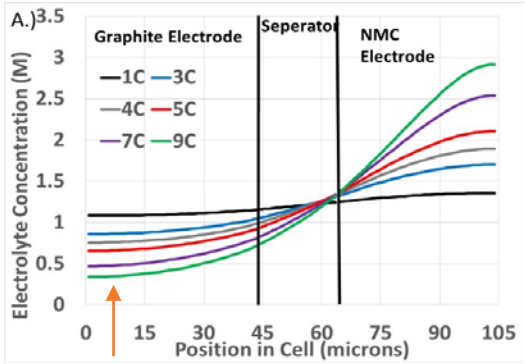
Colclasure et al., *Electrochimica Acta* (2020)

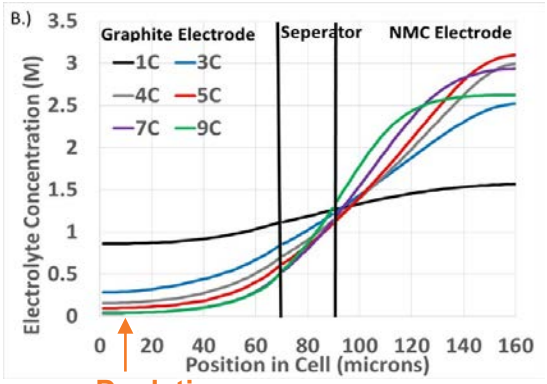
ECHEM MODEL RESULTS

1.5 mAh/cm² loading (40 µm)
XFC capable

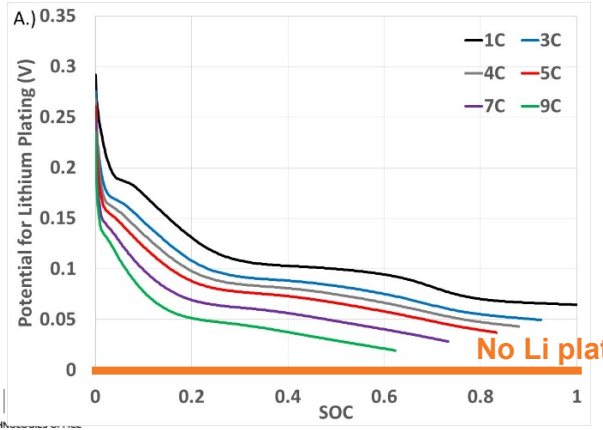
2.5 mAh/cm² loading (70 µm)
Barely XFC capable

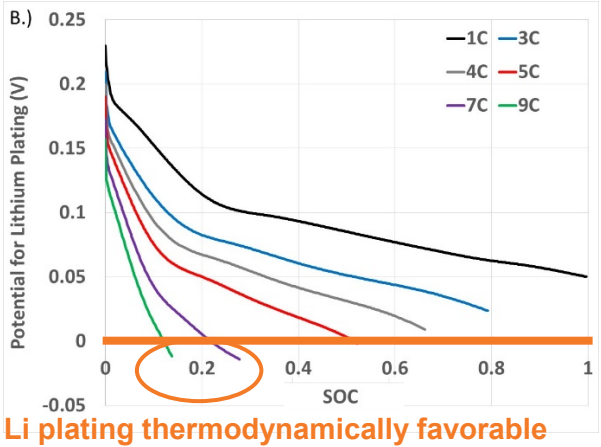
Electrolyte concentration





Li plating onset

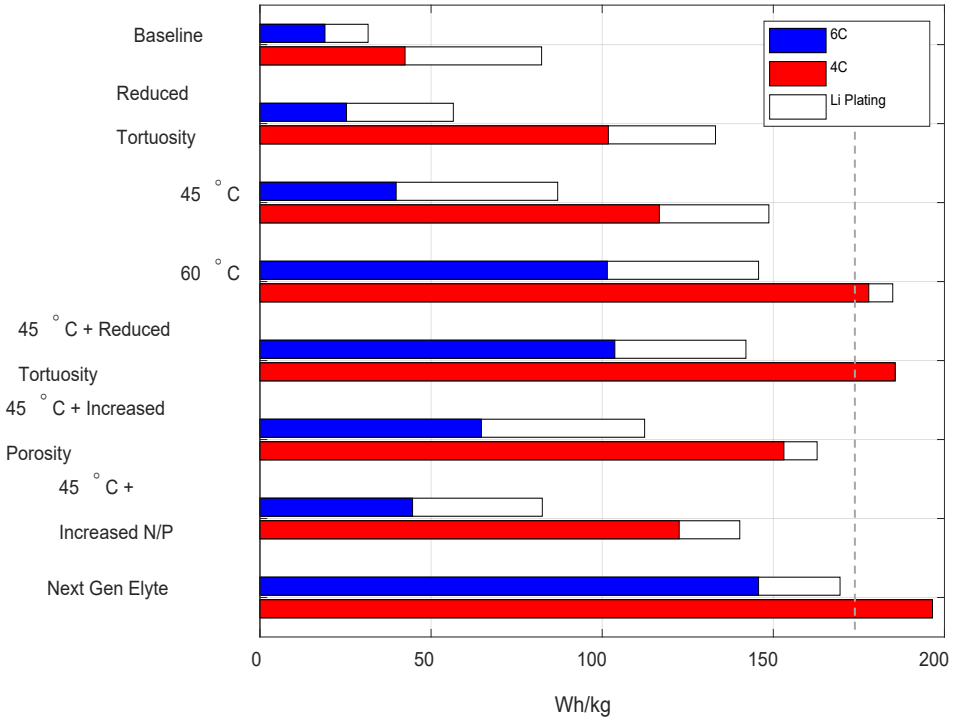




Colclasure et al., *Electrochimica Acta* (2020)

ENABLING XFC AT LOADING OF 3 mAh/cm^2

81-µm electrodes, 220-Wh/kg cell



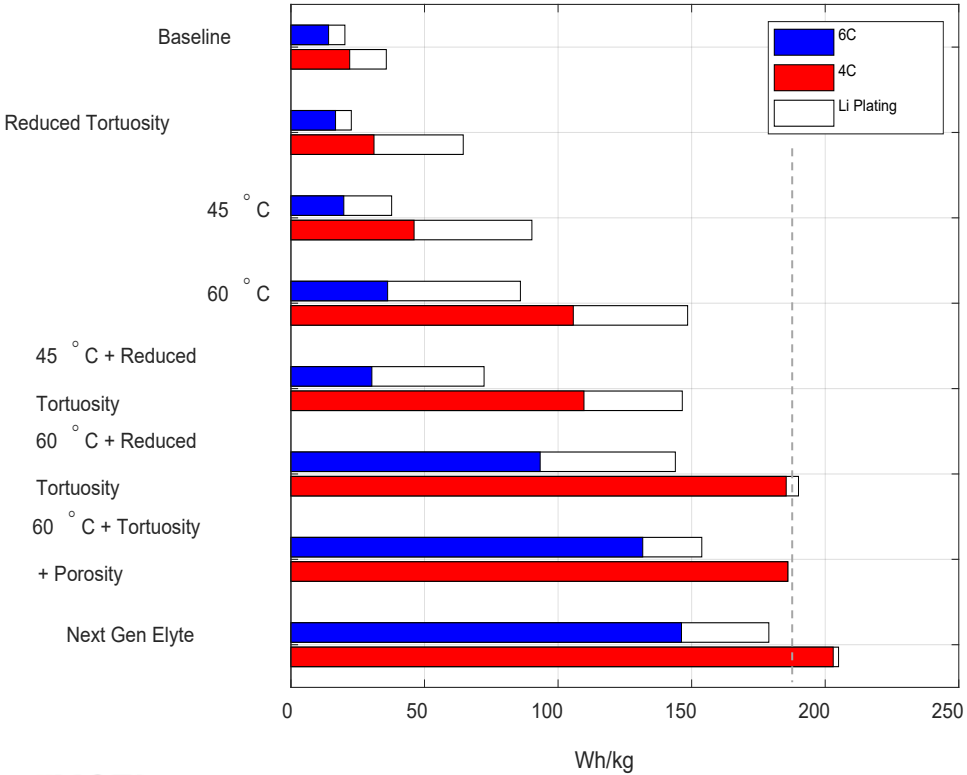
Constant current to 4.2-V cutoff (no constant voltage)

Colclasure et al., *Electrochimica Acta* (2020)

- Elevated temperature and reduced tortuosity are viable near-term technologies
- Porosity and Negative/Positive (N/P) ratio provide less value and reduce 1C density
- Important to continue research on electrolyte with enhanced transport
  - Next Gen Elyte = 1.8x ionic conductivity, 3x diffusivity, and transference number increased by 0.05

ENABLING XFC AT LOADING OF 4 mAh/cm^2

110-µm electrodes, 230-Wh/kg cell



Constant current to 4.2-V cutoff (no constant voltage)

Colclasure et al., *Electrochimoca Acta* (2020)

- 60°C would require development of surface-stabilizing coatings/electrolyte additives
- Important to continue research on electrolyte with enhanced transport
  - Next Gen Elyte = 2x ionic conductivity, 4x diffusivity, and transference number increased by 0.15
  - More important to improve diffusivity and transference number than conductivity

ENHANCED ELECTROLYTE TRANSPORT (1/2)

Goal: Improve diffusivity 3–4x, conductivity 2x, transference +0.05–0.15

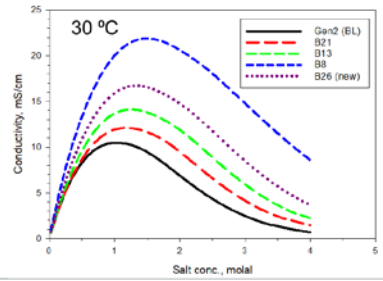
1) Advanced electrolytes[1]

- Models[a,b] show as much as 7x improved diffusivity; 1.5x improved conductivity vs. Gen2[c]
- Experiments underway
- Challenge: Cost and stability (life)

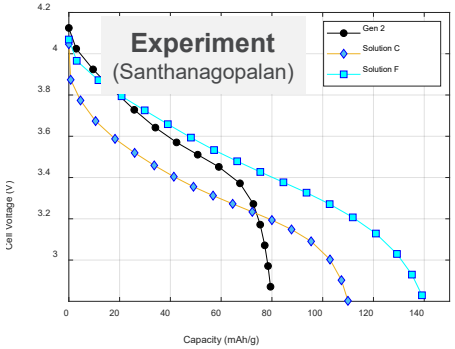
2) Elevated temperature

- Compared to next-gen. electrolyte, it achieves
  - @ 45°C, 20%–50%
  - @ 60°C, 60%–100%
- Cell-internal heater demo[2] (EC Power)
  - Preheated cell to 50°C followed by 6C charge
  - Achieved 2,500 cycles lifetime
  - Challenges: Calendar life. Cooling BEV300 battery 20°C in 15 min. requires >7.5-kW cooling

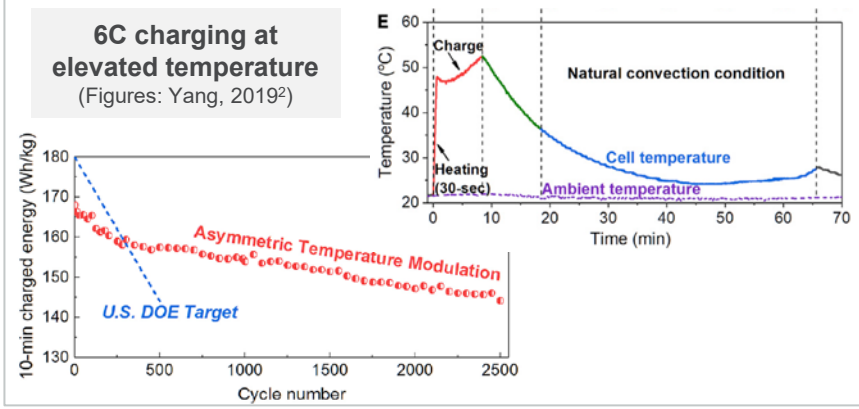
Advanced Electrolyte Model
(Gering)



Experiment
(Santhanagopalan)



6C charging at elevated temperature
(Figures: Yang, 2019[2])



1. See XCEL poster presentation on electrolyte
2. Xiao-Guang Yang et al. 2019. "Asymmetric temperature modulation for extreme fast charging of lithium-ion batteries." *Joule* (3): 1-18.

a. NREL molecular dynamics calculation of "Solution F": 30% 3-Oxabicyclo[3.1.0]hexane-2,4-dione, 70% Gen2
b. INL Advanced Electrolyte Model calculation of "B8": EC:DMC:EA:PN:TMP (15:30:20:20:15) w/ (4%VC, 3%FEC) + $LiPF_6$
c. Gen2: EC:EMC (30:70) + $LiPF_6$

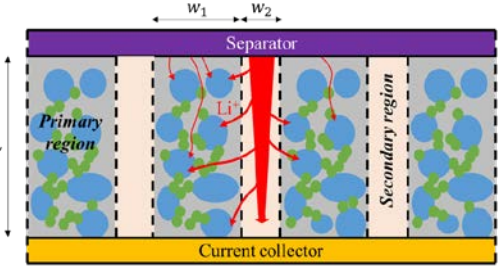
ENHANCED ELECTROLYTE TRANSPORT (2/2)

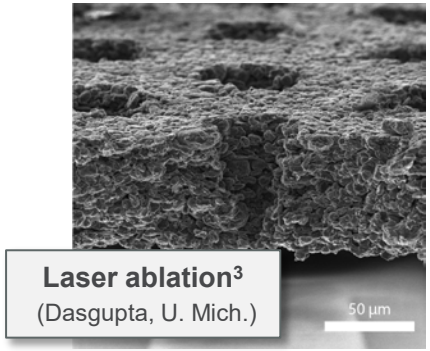
Goal: Improve diffusivity 3–4x, conductivity 2x, transference +0.05–0.15

3) Lower electrode tortuosity…

a) …globally: secondary pore networks

- Implemented at both electrodes, models predict
  - 35% benefit at 3 mAh/cm^2
- Flake-like particles work well w/ secondary pores [a,1]
- Challenge: Cost. Channel spacing and width must be small (<10 μm, <5 μm, respectively) evenly repeated and occupy ~25% of total volume [2]



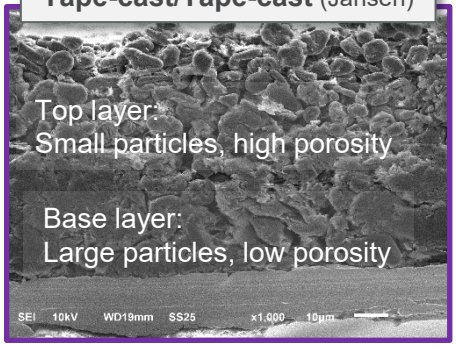


Laser ablation[3] (Dasgupta, U. Mich.)

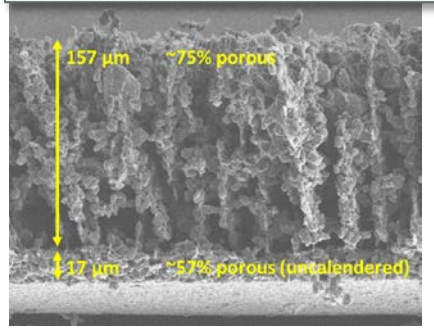
b) …locally: graded or dual-layer electrodes

- Model predicts 20%[b] to 68%[c] more 6C charge acceptance without plating
  - Base: 25% porosity, 8.0-μm particles
  - Top: 45% porosity, 2.4-μm particles
- Challenge: Cost, process optimization

Tape-cast/Tape-cast (Jansen)



Tape-cast/Freeze-cast (Doeff)

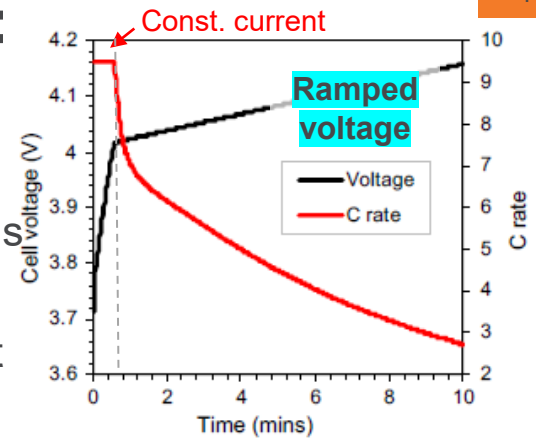


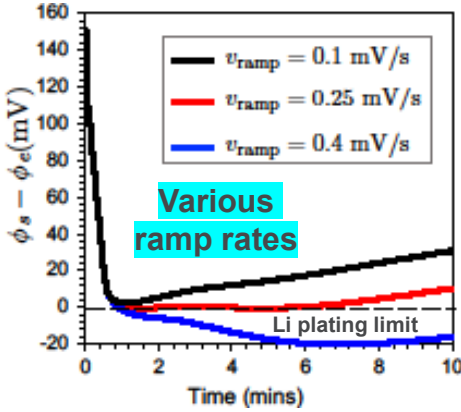
1. F.L.E. Usseglio-Viretta et al. 2020. "Enabling fast charging of lithium-ion batteries through secondary-/dual- pore network: Part I – Analytical diffusion model." *Electrochimica Acta* 342: 136034.
2. W. Mai et al. 2020. "Enabling fast charging of lithium-ion batteries through secondary-/dual- pore network: Part II – Numerical diffusion model." *Electrochimica Acta* 341: 136013.
3. K.-H. Chen, M.J. Namkoong, V. Goel, C. Yang, J. Mazumder, K. Thornton, J. Sakamoto, and N.P. Dasgupta. "Efficient Fast-charging of Lithium-ion Batteries Enabled by Three-dimensional Graphite Anode Architectures" (submitted

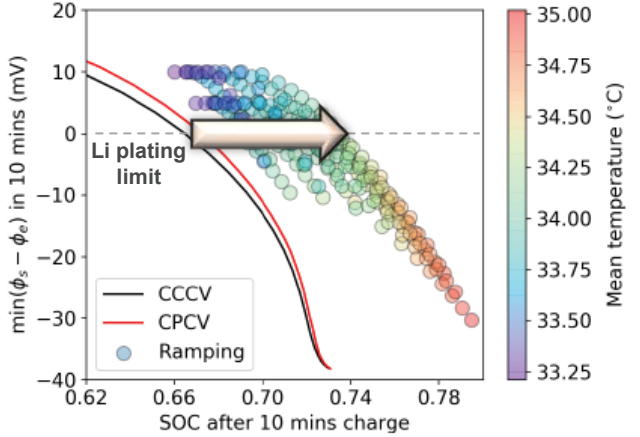
a. Without secondary pore network, spherical particles are preferred for low electrode tortuosity. Spherizing graphite adds cost.
b. Anode bi-layer porosity (2.5 mAh/cm^2 loading)
c. Anode bi-layer particle size and porosity + cathode bi-layer porosity (2.5 mAh/cm^2 loading)

SEMI-OPTIMAL CHARGE ALGORITHM[1]

- Used echem model to screen hundreds of CCCV, CPCV, and multi-step charge protocols
- Found optimal[2] multi-step/smooth profile would exactly ride the Li plating potential limit
  - Key is to avoid high current and high voltage at the same time
- Model found a **near-optimal profile: constant-current, ramped-voltage**
  - Experimental validation begun at INL







1. W. Mai, A.M. Colclasure, and K. Smith, "Model-instructed design of novel charging protocols for the extreme fast charging of lithium-ion batteries without lithium plating," *J. Echem. Soc.*, accepted.*
2. Considering Li plating limit only (not considering cathode cracking, plating/stripping current reversal, elevated temperature, etc.)

Achievement with Voltage Ramping

11% XFC improvement @ 2.5 mAh/cm^2

19% XFC improvement @ 4.0 mAh/cm^2

CCCV = constant-current, constant-voltage charge protocol
CPCV = constant-power, constant-voltage charge protocol

ADDITIONAL DESIGN CONSIDERATIONS (1/2)

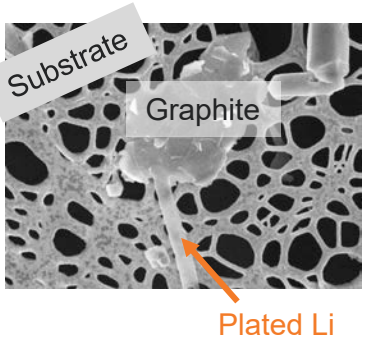
Building understanding of and how to suppress heterogeneities that cause early Li plating

- In real systems as well as 2D/3D models, Li plating occurs earlier than what the 1D macro-homogeneous electrochemical model predicts. Experiments and modeling studies are underway to understand each and develop strategies.

50 nm ◄---------- 10 cm

Crystal anisotropy

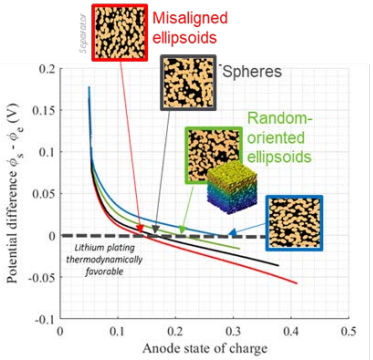
Single-particle SEM



- Lithium tends to grow at the tip or on the edge planes of graphite (Chueh, Yeh)

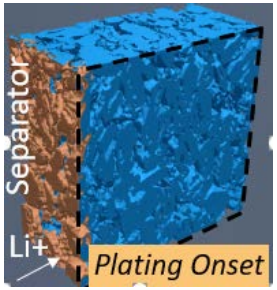
Particle surface, morphology, orientation

3D microstructure model



- For the same surface area, through-plane-aligned ellipsoids are best, then spherical particles

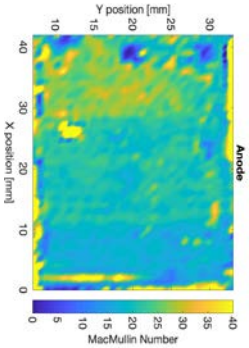
Real 3D microstructure vs. spheres (macro model assump.)



- Particle and pore heterogeneity of real electrode causes it to plate 2.5% earlier

Tortuosity variation

Ionic resistance of aged cell



- Mapping electronic and ionic resistance of fresh/aged cells to determine correlation with Li plating (Wheeler)

Electrolyte wetting/trapped gas

Electrolyte wetting model



- Model matches different Li plating patterns in pouch and coin cells (Dees)
- Pouch cell tear down image (Tanim)

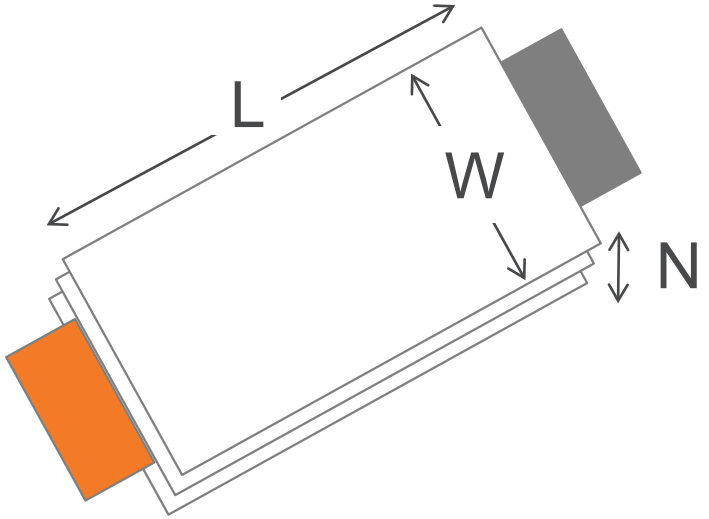
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VEHICLE TECHNOLOGIES OFFICE

- 50% surface area increase (via smaller particle size) delays plating 15% ΔSOC but reduces calendar life

ADDITIONAL DESIGN CONSIDERATIONS (2/2)

Thermal management for 6C charge



- **Voltage drop** in current collectors inside cell
  - Proportional to L^2
  - Must limit L < 10–15 cm* to suppress Li plating heterogeneity (V_{drop} < 10 mV)
- **Efficiency**
  - Dominant heat source is electrolyte (55%), followed by reaction (25%)
  - Losses drive heat load and temperature rise. For 100-kWh battery,
    - Heat removal to stay isothermal ~40–55 kW**
    - Adiabatic temperature rise ~50°C–70°C**
  - Today's cells have 88%–92% efficiency at 6C, 30°C. Desire >95%.
- **Heat removal** (calendar life, safety)
  - Desire <3°C ΔT within cell and pack
  - Cathode and cathode/separator interface are bottlenecks for through-plane thermal conductivity
  - Temperature difference proportional to
    - H^2 if cooled from bottom cold plate (H = cell or cooling-fin height)
    - N^2 if cooled from cell faces (N = number of layers). Limits cell size to ~30 Ah if cooling single face.

*In-plane thermal conductivity and voltage drop can both improved by doubling foil thickness. Enables cells with length L of 20–30 cm but reduces energy density from 230 to 210 Wh/kg.
**With realistic system size closer to 5–10 kW, battery temperature will fluctuate. (System is neither isothermal nor adiabatic).

RESPONSES TO PREVIOUS YEAR'S COMMENTS (1/2)

- "Findings regarding **dual-coated electrode** and **onset of plating** as well as **edge-effect prediction** seem like **particularly valuable** accomplishments. The reviewer **would like to better understand any related outcomes with actual cells**."
  - Modeling results prompted ANL-CAMP to **manufacture several dual-coated electrode combinations** based on available graphite particle sizes which **did show expected Li plating order**…
    - —8-µm particles at front, 17-µm particles at back → less plating
    - —17-µm particles at front, 8-µm particles at back → more plating
  - …but did not yet achieve target particle size (2 µm at front, requiring new graphite supplier) or porosity (requiring process optimization). The **ANL-CAMP team is presently working to optimize the particle size, porosity and fabrication methodology**.
  - Based on model recommendations for secondary pore networks, **LBNL is focusing on methods to densify their freeze-cast electrodes to ~35% overall porosity**. Their recent dual-layer tape-/freeze-cast electrodes are coming closer.
  - Recent **modeling of electrolyte wetting predicts gas entrapment at the different preferential Li plating locations in pouch cells** (center) **versus coin cells** (ring near outside). The team is **designing experiments to test whether this is indeed the cause** (e.g., by electrolyte-filling the cells in different manners and cycling) and validate the model.
- "Utilizing **computerized tomography (CT) of graphite** used in the program **enables best particle-design strategy** for fast charging, and it could be **useful input to graphite manufacture**. It might be useful to **apply to positive electrodes**."
  - We have characterized both anode and cathode using CT. We find most **cathode particles are already nearly spherical** and have **less tortuosity** than anodes. (Nonetheless, secondary pores are also beneficial to cathode).
  - Described in the Cathode Thrust Poster, **detailed cathode sub-particle modeling is underway** to understand **polycrystalline architecture** impact on cathode solid tortuosity, mechanical stress, and capacity fade. We will make architecture recommendations.

RESPONSES TO PREVIOUS YEAR'S COMMENTS (2/2)

- "Proposing the best electrode structure is valuable information, but it is also **critical to check the feasibility of making such an electrode in the real world**. It is **difficult to make an ellipsoid-shaped graphite particle aligned along the electrolyte transport direction** since **orientation of the particle can be altered during the calendering process**."
  - We acknowledge that **not all architectures are scalable** (though they may still help our understanding) and thus **seek many possible solutions and provide as many practical solutions as possible to the community**.
  - This year we published a paper that reviewed electrode architectures,[1] listing lab-scale manufacturing methods able to lower tortuosity factor. **Ellipsoid-shaped graphite can be aligned with magnets**, for instance.
  - **Calendering is indeed an issue**. Some other densification method may be needed to not damage alignment. LBNL's partial solution is to tape-cast and calendar a dense layer, followed by freeze-casting of a less-dense aligned layer. Laser ablation avoids these problems.
- The reviewer added that **investigating at the sub-particle level provides more insight**, and the development of **predictive modeling helps the vast majority of the user community**. However, the reviewer remarked that the **continuum-modeling is too soft and should directly support ongoing experimental activities**.
  - We share this goal. In FY19, the **model was more extensively validated (slides 8–9) and**, late in FY19, **used to make a wide range of projections (slides 10–11) of what near- and far-term technologies best enable fast charge**. We hope the reviewer is encouraged by these results, **published in Journal of the Electrochemical Society**.[2]
  - We are building **detailed sub-particle models** and will have results later this year (**cathode polycrystalline architecture**, graphite surface **locations of Li nucleation**).

1. F.L.E. Usseglio-Viretta, W. Mai, A.M. Colclasure, M. Doeff, E. Yi, and K. Smith. 2020. "Enabling Fast Charging of Lithium-Ion Batteries through Secondary-/Dual-Pore Network: Part I - Analytical Diffusion Model." *Electrochimica Acta* 342(10): 136034.
2. A.C. Colclasure, T.R. Tanim, A.N. Jansen, S.E. Trask, A.R. Dunlop, B.J. Polzin, I. Bloom, D. Robertson, L. Flores, M. Evans, E.J. Dufek, and K. Smith. 2020. "Electrode scale and electrolyte effects on extreme fast charging of lithium-ion cells." *Electrochimica Acta* 337.

COLLABORATION ACROSS LABS AND UNIVERSITIES

ANL: Cell and electrode design and build, performance characterization, post-test, cell and atomistic modeling, cost modeling

LBNL: Li detection, electrode architecture, diagnostics

INL: Performance characterization, Li detection, failure analysis, electrolyte modeling and characterization, acoustic detection

SLAC: Li detection, novel separators, diagnostics

NREL: Thermal characterization, life modeling, micro- and macro-scale modeling, electrolyte modeling and characterization

ORNL: Detailed graphite/electrolyte interface and Li nucleation, plating/stripping kinetic modeling

Brigham Young University: Ionic and electronic resistance measurement/2D mapping

REMAINING CHALLENGES AND BARRIERS

- Li-plating heterogeneities are characterized by a large range of length scales
  - Requires separate models and experiments to resolve all length scales
  - Coupling models at different scales (e.g., sub-particle with microstructure scale) requires a significant increase in degrees of freedom and/or requires a more complex numerical scheme
  - Representativity of small length-scale heterogeneity is difficult to evaluate for the whole electrode, and requires a systematic (or a least a statistically relevant) measurement plan
- Manufacturing structured electrodes (dual layer, secondary pore network) is challenging, requires extra manufacturing steps and must be cost effective.

FUTURE WORK

- Continue to refine and apply models to guide design of XFC-capable battery
  - Sub-particle modeling to understand detailed crystalline structure, graphite surface, and surface energy effects on transport and Li plating/stripping
  - Electrode/electrolyte strategies vs. cost and life
  - Cell/pack tradeoffs in system sizing, energy density, cost, and life
- Perform experimental validation of model-guided findings
  - Voltage-ramping charge protocol
  - Dual layer, secondary pore network and reduced carbon-binder additives electrode designs
  - Electrolyte wetting/trapped gas as cause of local plating pattern
  - Advanced electrolytes
- Incorporate aging mechanisms
  - Li plating/stripping
  - Cathode cracking
  - Calendar life vs. time at temperatures.

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

SUMMARY

- Electrochemical models were **validated for conventional electrodes and electrolyte** with 1.5 to 2.5 mAh/cm^2 cells built by ANL-CAMP facility
- Models elucidate **strategies to achieve 10–15-minute charge** for 3–4 mAh/cm^2 (220–230 Wh/kg) cells
  - Near-term: Combination of reduced electrode tortuosity and charging at elevated temperature (~45°C)
  - Future: Next-generation electrolyte (3–4x diffusivity, 2x conductivity, +0.05–0.15 transference)
- Promising modeling results (listed below) are being followed up with experimental validation
  - **Advanced electrolytes** part way to next-generation electrolyte goals but with stability/life issues
  - **Secondary pore networks** increase charge acceptance 35%
  - **Dual-layer electrodes** increase charge acceptance 20%–68%
    - —Base layer: 25% porosity, 8.0-µm particles
    - —Top layer: 45% porosity, 2.4-µm particles
  - **Voltage-ramping charge protocol** increase charge acceptance 11%–19%
  - Electrolyte wetting model explains different localized Li plating patterns in Round-2 coin and pouch cells
  - Material design, quality control, and 3D cell/system design needed to suppress heterogeneities that cause early Li plating
- Thermal management challenges require higher cell efficiency (less heat generated), careful cell design, and innovative cooling strategies to remove heat.

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