U.S. NAVAL RESEARCH LABORATORY

Electrode Specific Degradation Tailored by the Directionality of Thermal Gradients in Li-ion Batteries

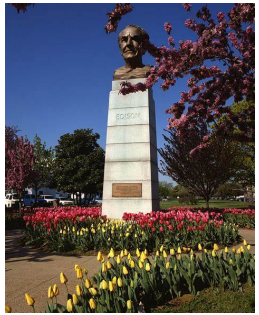
Rachel Carter[a], Robert W. Atkinson III[b], Todd A. Kingston[a,c], Emily Klein[a], Mukul Parmananda[d], Conner Fear[d], Partha Mukherjee[d], Matthieu Dubarry[e], and Corey T. Love[a]

[a]Chemistry Division, U.S. Naval Research Laboratory
[b]EXCET, Inc.
[c]Currently, Mechanical Engineering, Iowa State University
[d]School of Mechanical Engineering, Purdue University
[e]School of Ocean and Earth Science and Technology, Hawaii Natural Energy Institute

11 August 2021

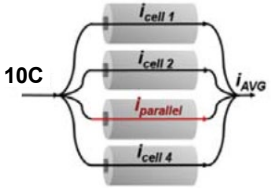
UL Battery Safety Science Symposium

Intro to NRL and our research

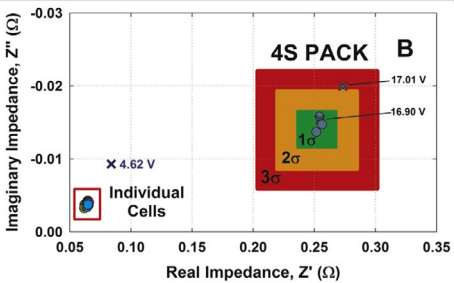


The Navy's Corporate Laboratory located in Washington, DC
Employs ~2000 PhD's spanning many topics (space, radar, optical science, chemistry, materials science, biological, and computing)

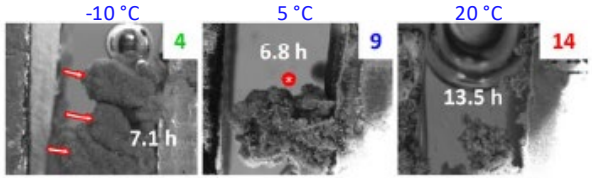
Thomas Edison,
New York Times, May 1915
"The Government," he proposed in a published interview, "should maintain a great research laboratory…. In this could be developed…all the technique of military and naval progression without any vast expense."
-$1.5M allotted by congress and construction began 1920



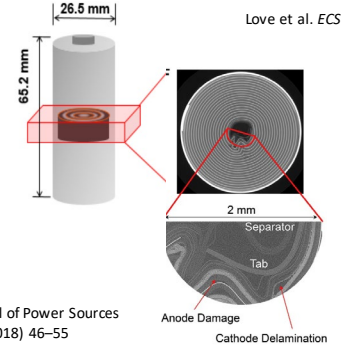
Journal of Power Sources 381 (2018) 46–5548



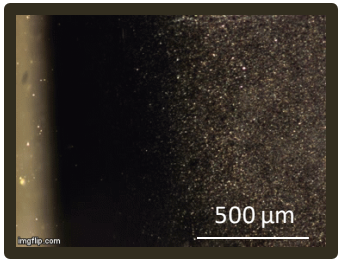
C.T. Love et al. Journal of Power Sources 266 (2014) 512e519513



Love et al. *ECS Electrochem. Lett.* **2015**, 4, A24-A27



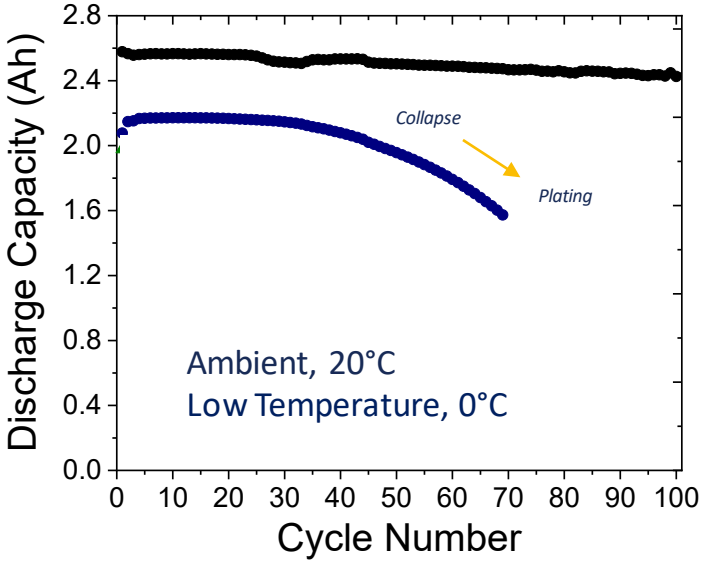
Journal of Power Sources 381 (2018) 46–55



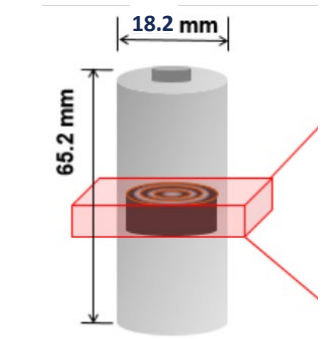
0%
25%
50%
100%

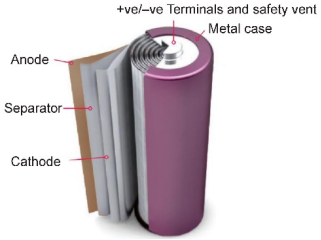
Carter et al. J Electrochemical Soc. **2020**, 167, A4

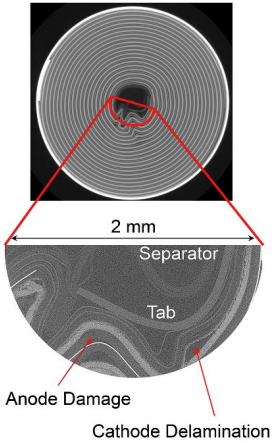
Plating Conditions Commercial cells

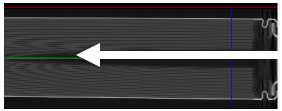


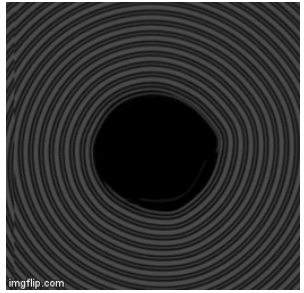
Carter et al. Journal of Power Sources 437 (2019) 226820



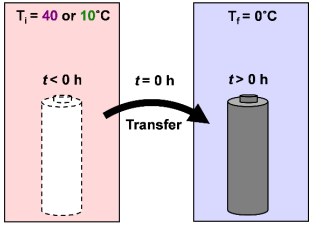


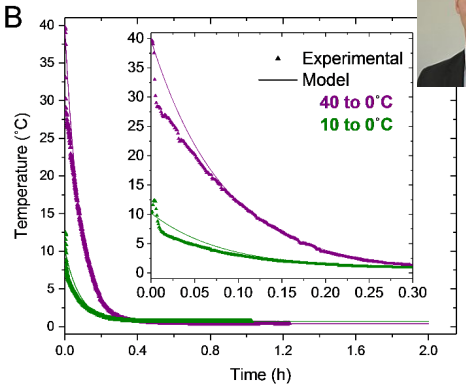




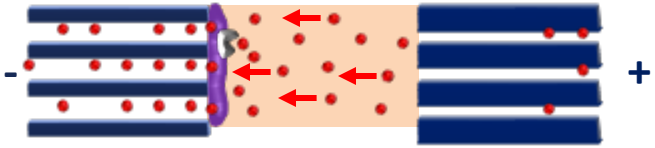


Thermal Transient Facilitates Plating in COTS 18650s

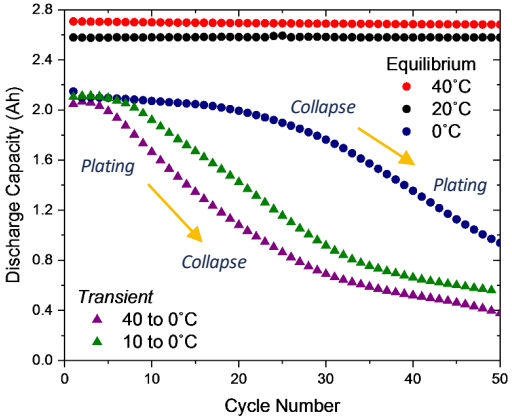




Only Distinction is the Temperature in the First Cycle

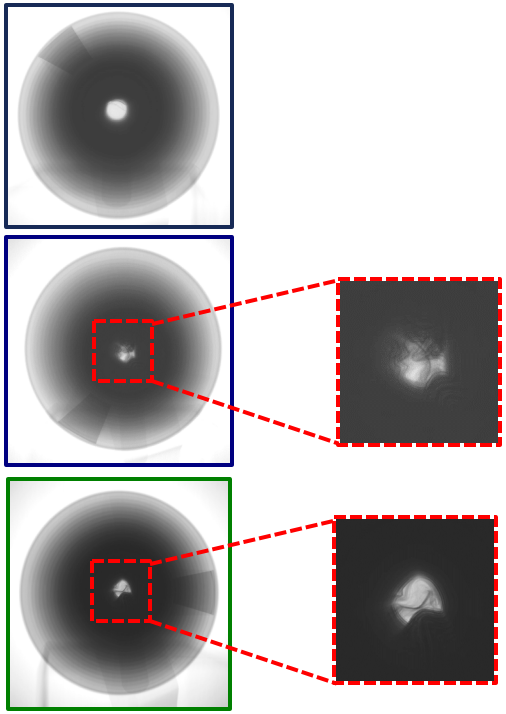


Over half the ions deposit as metal



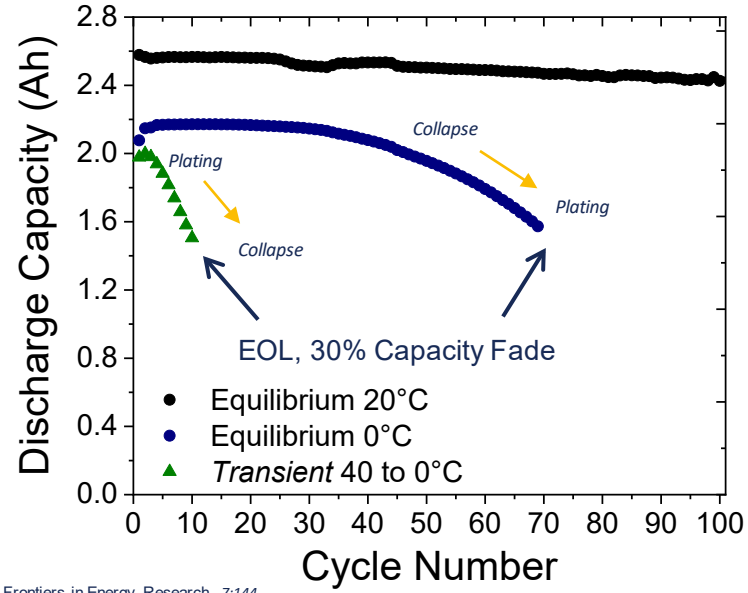
Carter et al. Frontiers in Energy Research, *7:144. doi: 10.3389/fenrg.2019.00144,* 2019.

EOL X-ray Radiographs

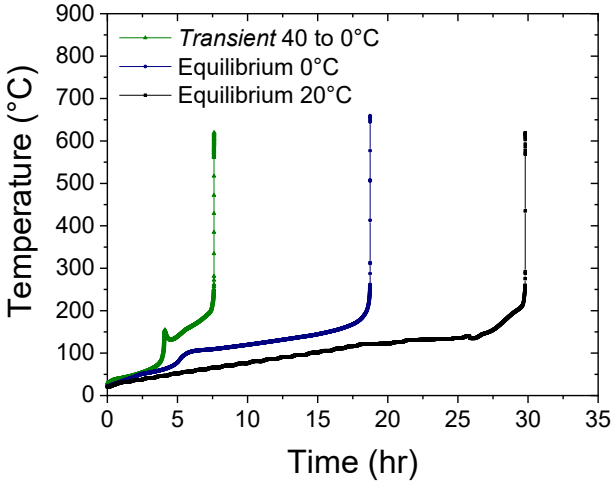


U.S. NAVAL RESEARCH LABORATORY

Safety Implications of Cell's Thermal and Electrochemical History



Carter et al. Frontiers in Energy Research, *7:144. doi: 10.3389/fenrg.2019.00144,* 2019.



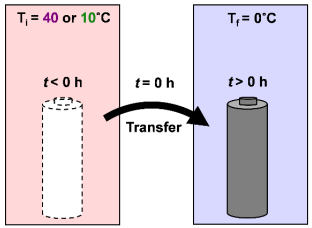
Transient 40 to 0˚C

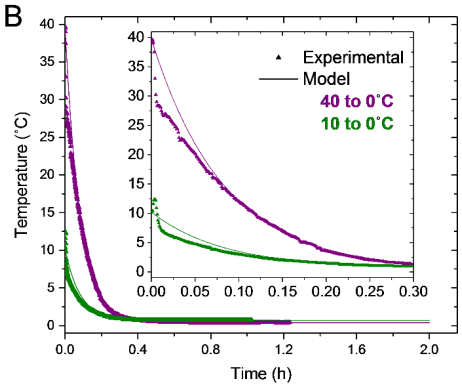
Equilibrium 0˚C

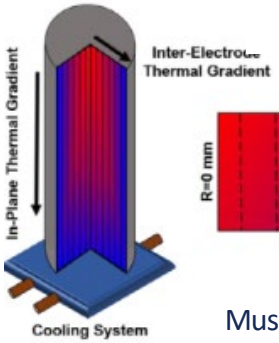
Equilibrium 20˚C

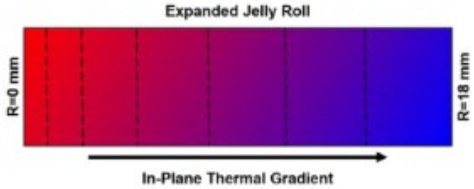
- Revealed Importance of Temporal Variation in Temperature
- Transition to coin cells for Fundamental Understanding: spatial and temporal control over temperature

Thermal Gradients in metallurgy: an analog for electrochemistry



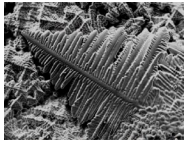


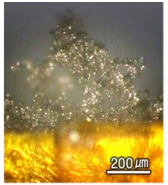




Must consider *Temporal* and *Spatial* Thermal Variations

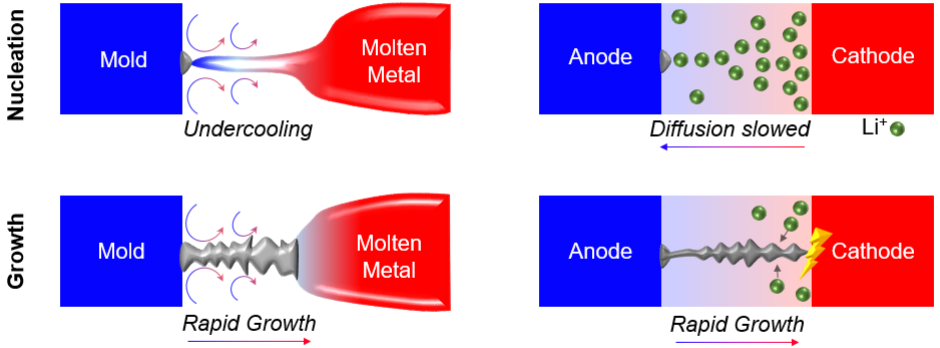
C Fear et al. Energy Storage Materials 35 (2021) 500–511





Metal Solidification
Liquid Undercooling Induced Dendrite

Electrochemical Deposition
Thermal Gradient Induced Dendrite



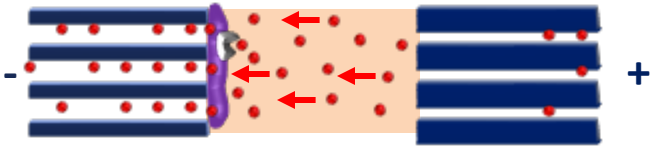
$T_{Mold} < T_{Melt}$

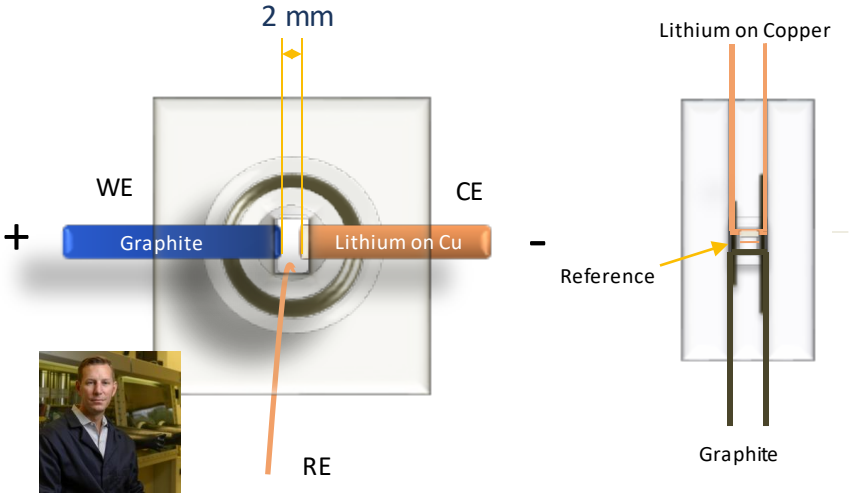
$T_{Anode} < T_{Cathode}$

R. Carter, C.T. Love, *ACS Appl. Mater. Interfaces*, 2018, 10 (31), pp 26328–26334

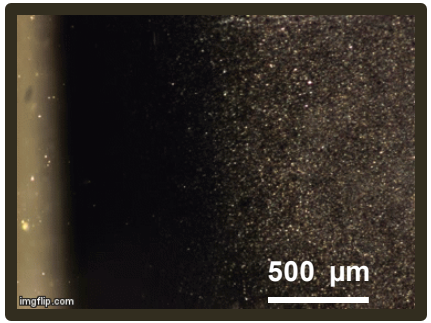
Proof of Concept in Optical Cell

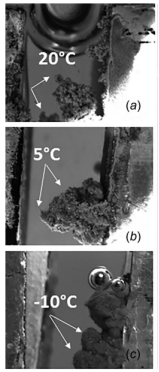
Opportunity for in-situ observation lithiation under abusive conditions





Ambient (~22˚C)

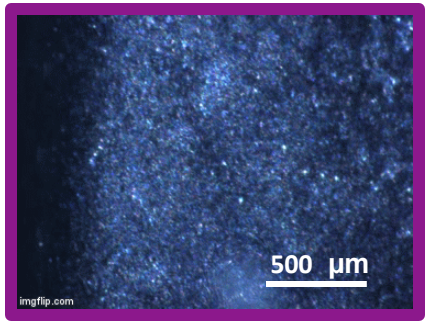


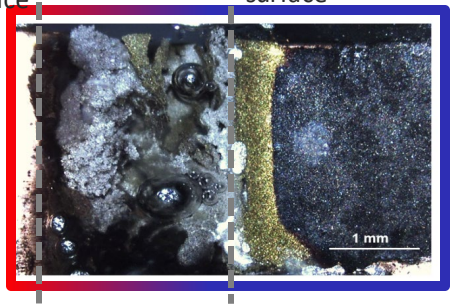


Love et al.
ECS Electrochem. Lett.
2015, 4, A24-A27

Thermal Gradient

(surface 0˚C, opposite electrode 40˚C)

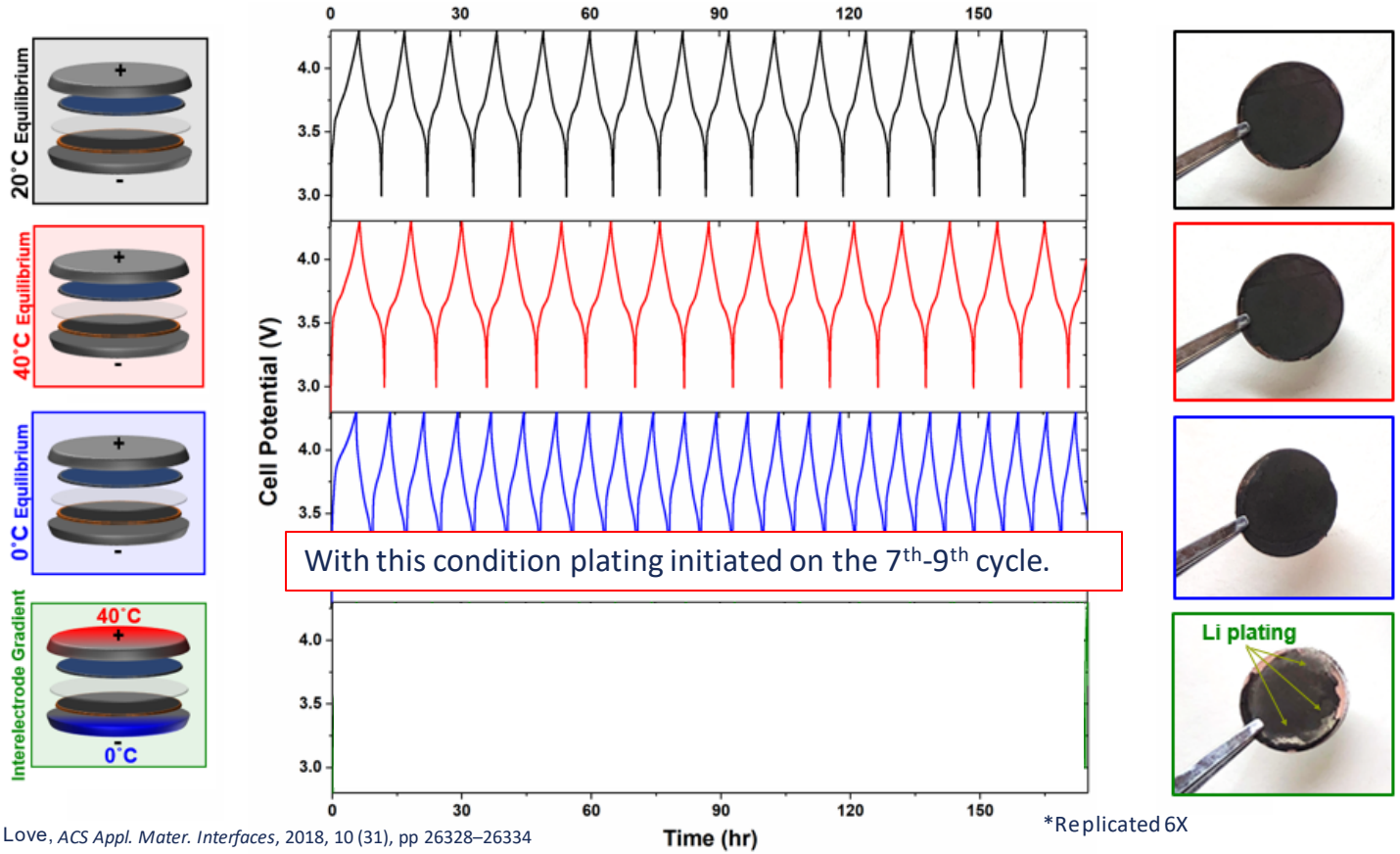




Carter et al. Journal of The Electrochemical Society, 2020 167 060510

U.S. NAVAL RESEARCH LABORATORY

Thermal Gradient Reliably Induces Plating at Graphite Anode



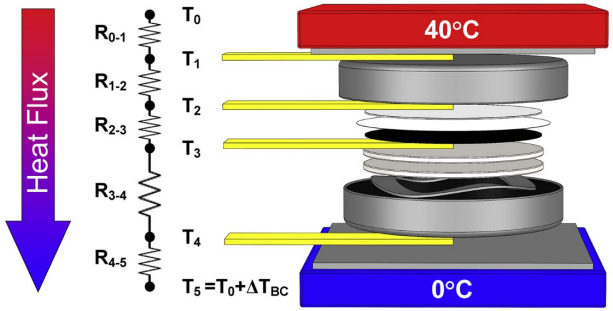
With this condition plating initiated on the 7th-9th cycle.

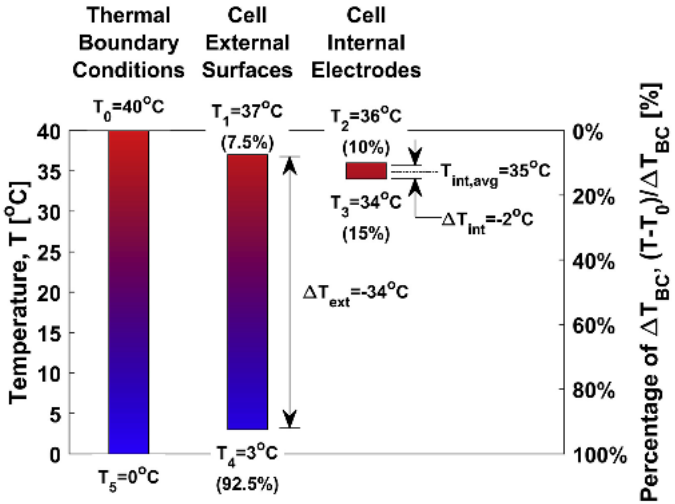
*Replicated 6X

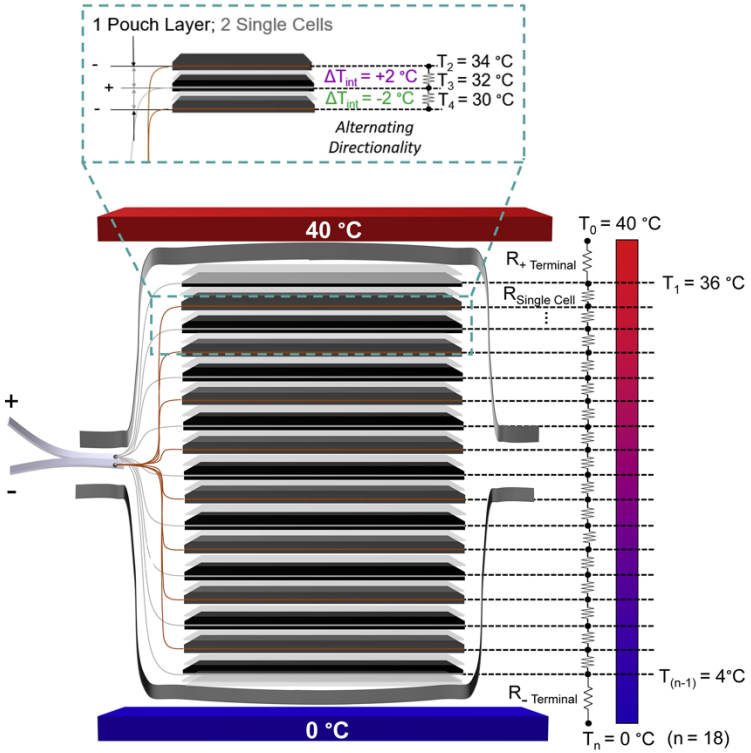
R. Carter, C.T. Love, *ACS Appl. Mater. Interfaces*, 2018, 10 (31), pp 26328–26334

U.S. NAVAL RESEARCH LABORATORY

Actual Interelectrode Gradient and Context for Pouch Cells





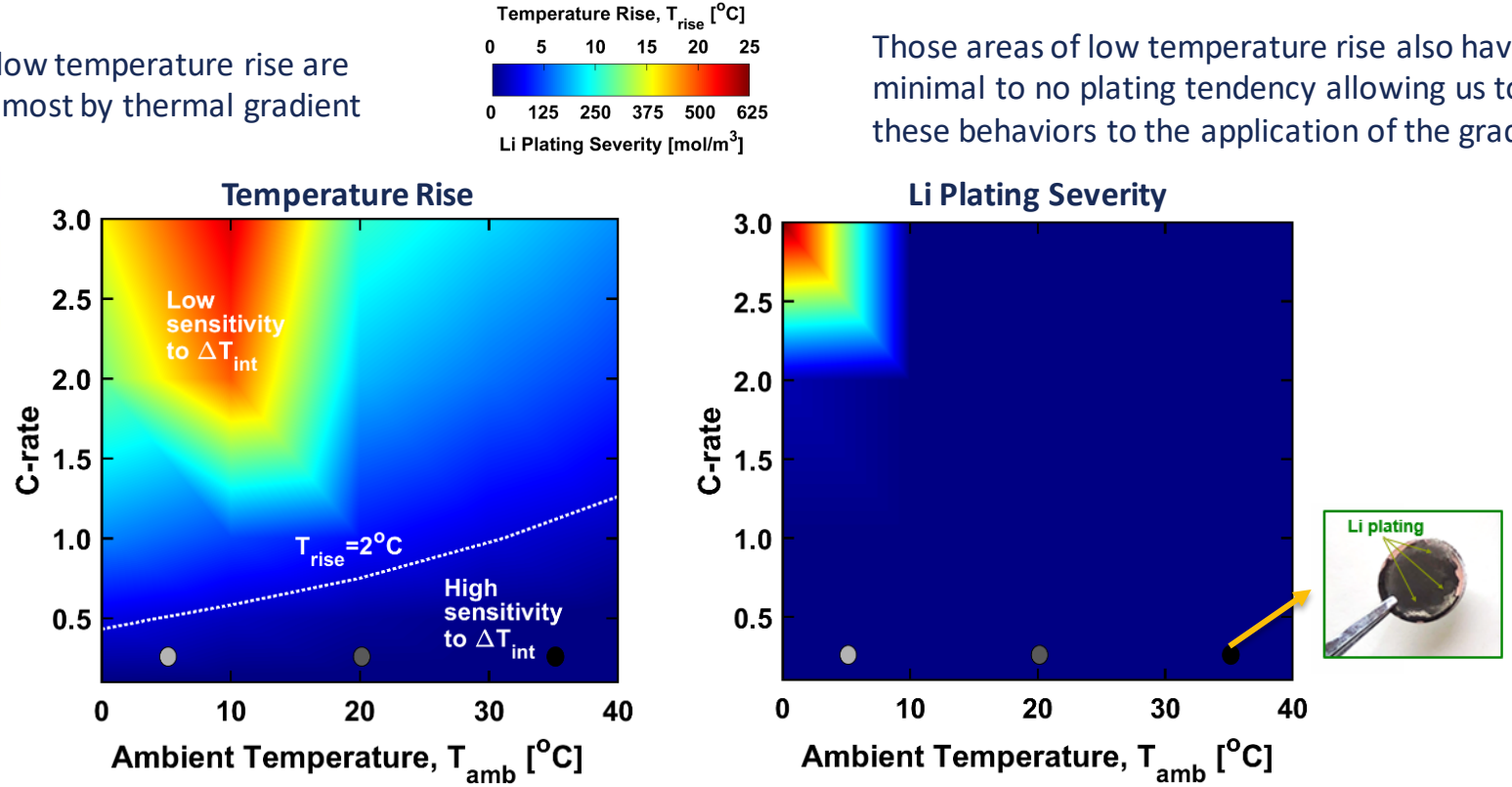


Carter, Kingston et al. Cell Reports Physical Science 2, 100351, March 24, 2021

Considering Operating Condition to Assess Risk

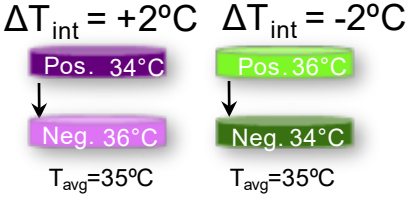
Areas with low temperature rise are altered the most by thermal gradient

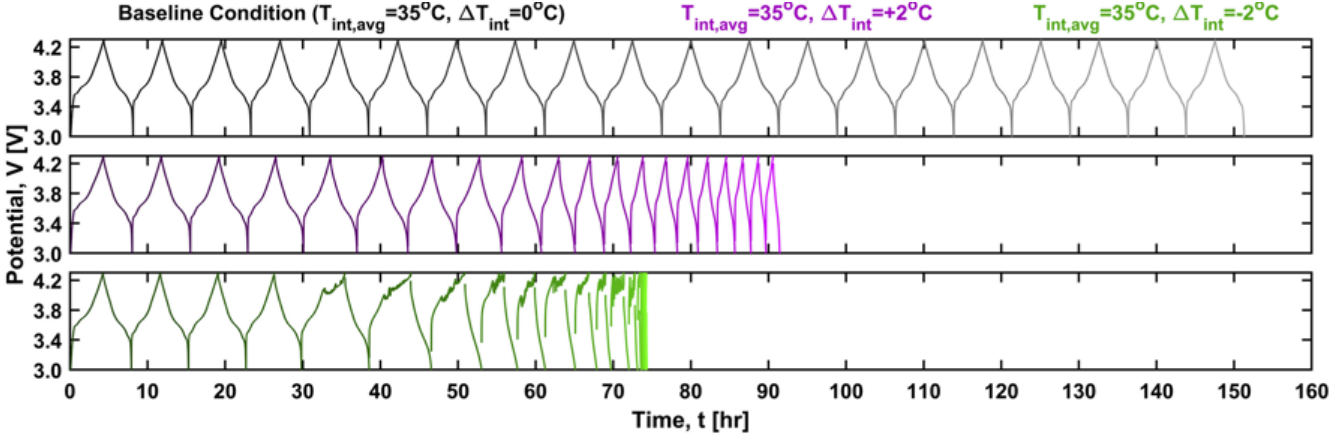
Those areas of low temperature rise also have minimal to no plating tendency allowing us to isolate these behaviors to the application of the gradient

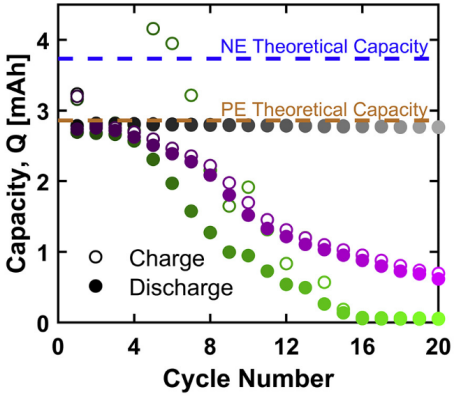


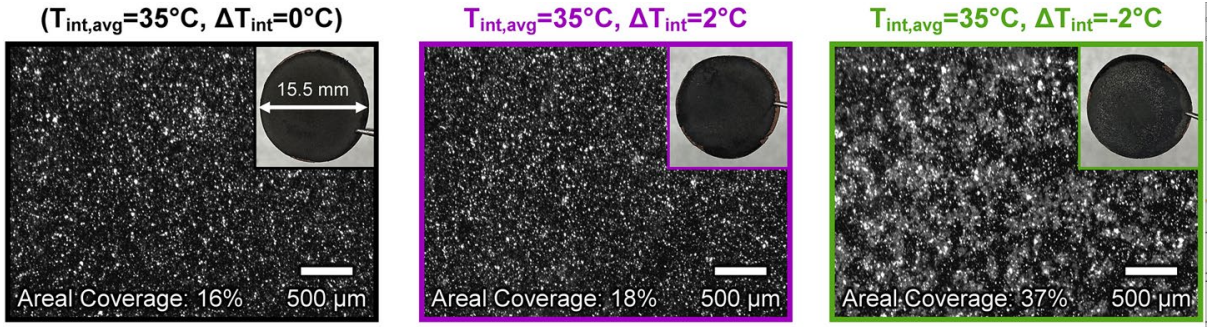
Carter, Kingston et al. Cell Reports Physical Science 2, 100351, March 24, 2021

Directionality of Thermal Gradient causes Diverging Modes









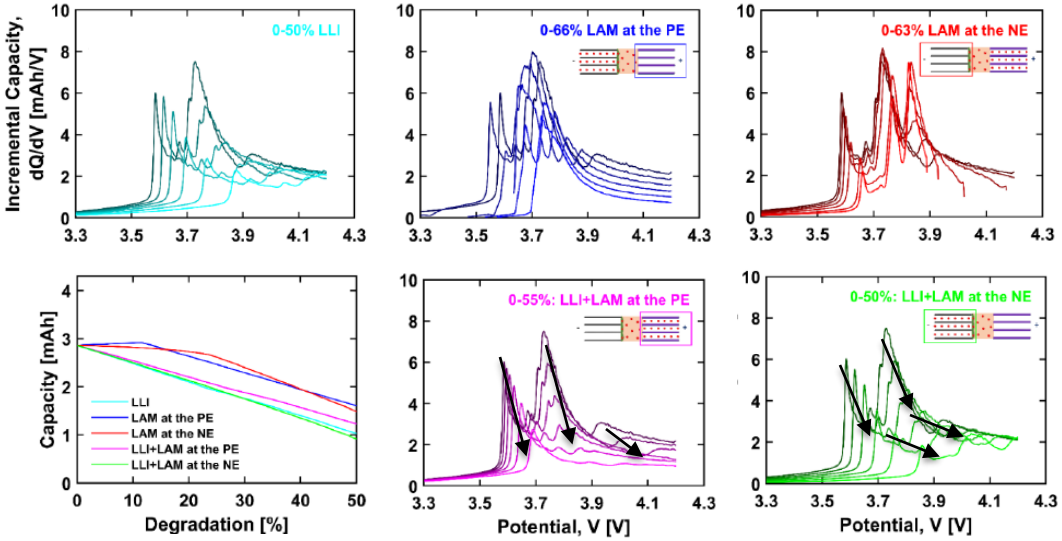
Carter, Kingston et al. Cell Reports Physical Science 2, 100351, March 24, 2021

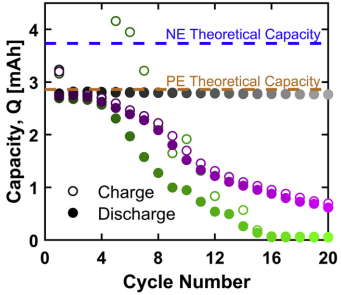
Determining Degradation Modes

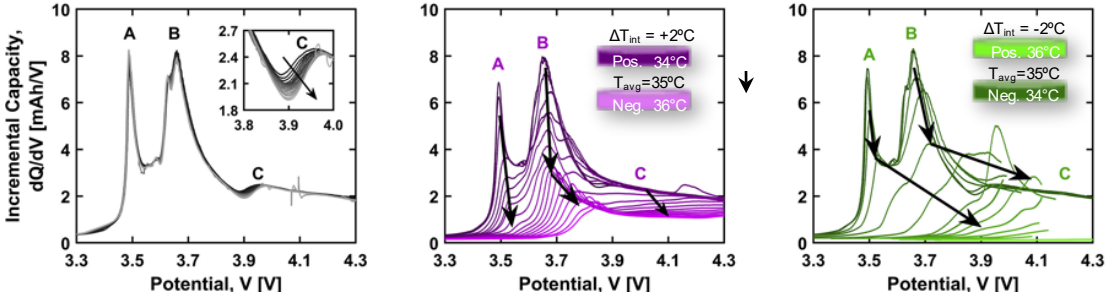
Incremental capacity analysis for SOH
Grid and EV

M. Dubarry et al. Journal of Power Sources 196 (2011) 3420–3425

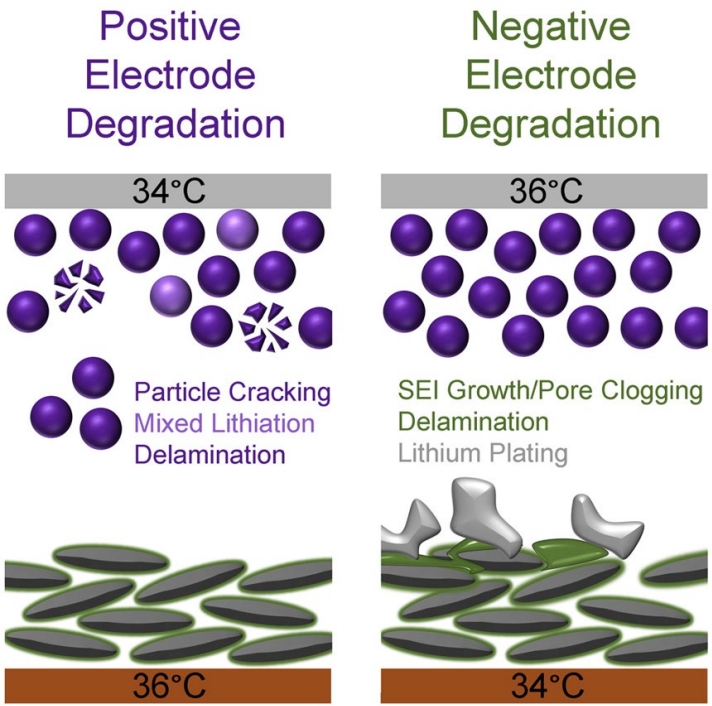
Carter, Kingston et al. Cell Reports Physical Science 2, 100351, March 24, 2021

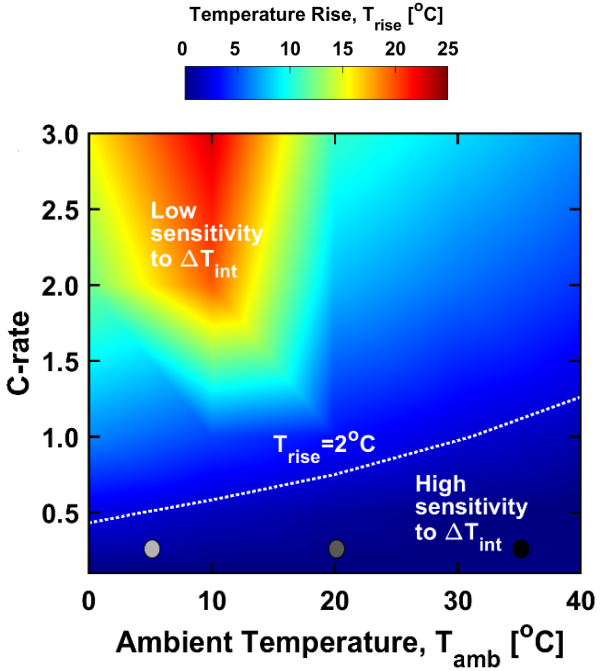






Electrode Specific Degradation and Informed Sensitivities

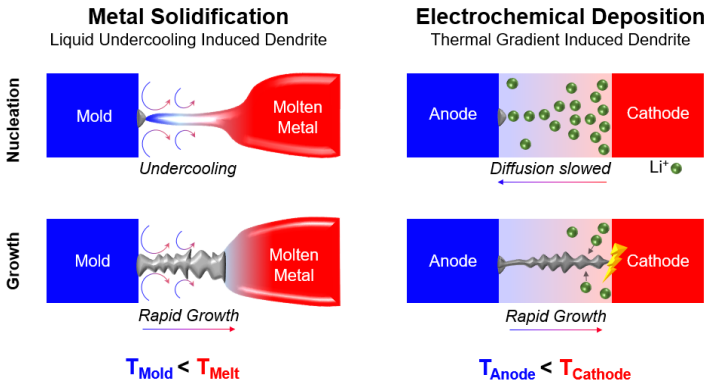


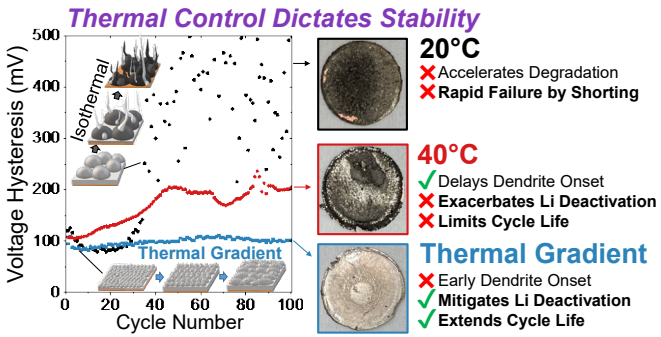


- Tailored Accelerated Aging Technique
- Understand behavior for Thermal Management Sensitivity

Carter, Kingston et al. Cell Reports Physical Science 2, 100351, March 24, 2021

Impact and Application of Thermal Gradient Findings





1. Understand its thermal-electrochemical coupling

- Intercalation electrodes (graphite, NMC)
- Reproduce realistic battery failure modes
- Understand and improve battery safety

Carter, C.T. Love, *ACS Appl. Mater. Interfaces*, 2018, 10 (31), pp 26328–26334

Fear et al. Energy Storage Materials 35 (2021) 500–511

Carter et al. Cell Reports Physical Science 2, 100351, March 24, 2021

2. Enable high-energy, next-generation batteries

- Li metal anode
- *Promote reversible lithium plating and stripping*
- *Enable high-rate charging*

Mistry, et al. *ACS Energy Lett*, (2019) 4 (1), 156-162

Atkinson et al. Energy Storage Materials 22 (2019) 18–2819

Atkinson et al. Materials Today Energy 18, 100538, 2020

Acknowledgments

Rachel Carter[a], Robert W. Atkinson III[b], Todd A. Kingston[a,c], Emily Klein[a], Mukul Parmananda[d], Conner Fear[d], Partha Mukherjee[d], Matthieu Dubarry[e], and Corey T. Love[a]

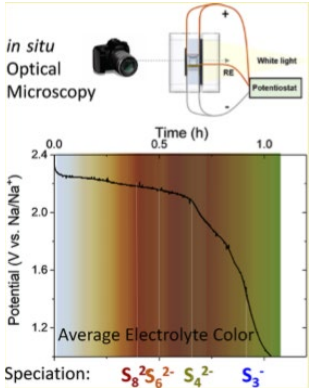
[a]Chemistry Division, U.S. Naval Research Laboratory; [b]EXCET, Inc,; [c]Currently, Mechanical Engineering, Iowa State University; [d]School of Mechanical Engineering, Purdue University; [e]School of Ocean and Earth Science and Technology, Hawaii Natural Energy Institute

NRL
Dr. Abhi Raj
Dr. Matthew (Jake) Lefler
Mr. Dillion Hayman
Dr. Karen S. Lyons (now Plug Power)
Dr. Andrew Geltmacher

ORNL
Dr. Mengya Li
Dr. Jianlin Li

University Collaborators
Prof. Iryna Zenyuk- UC Irvine
Prof. Matthew McDowell- GA Tech
Prof. Partha Mukherjee- Purdue University
Venkatesh Kabra
Susmita Sarkar
Aashutosh Mistry (now Argonne NL)

B-Team top golf 2019



Carter et al. ACS Sustainable Chem. Eng. 2021, 9, 92−100