Altering the Degradation Mode in Li-ion Batteries Through Directional Application of an Interelectrode Thermal Gradient

Todd A. Kingston,[1] Rachel Carter,[2] Robert W. Atkinson III,[3] Mukul Parmananda,[4] Matthieu Dubarry,[5] Conner Fear,[4] Partha P. Mukherjee,[4] and Corey T. Love[2]

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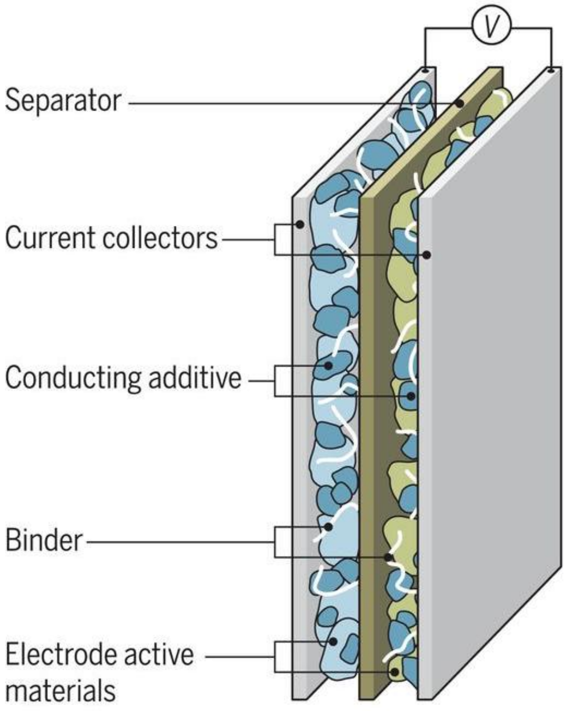
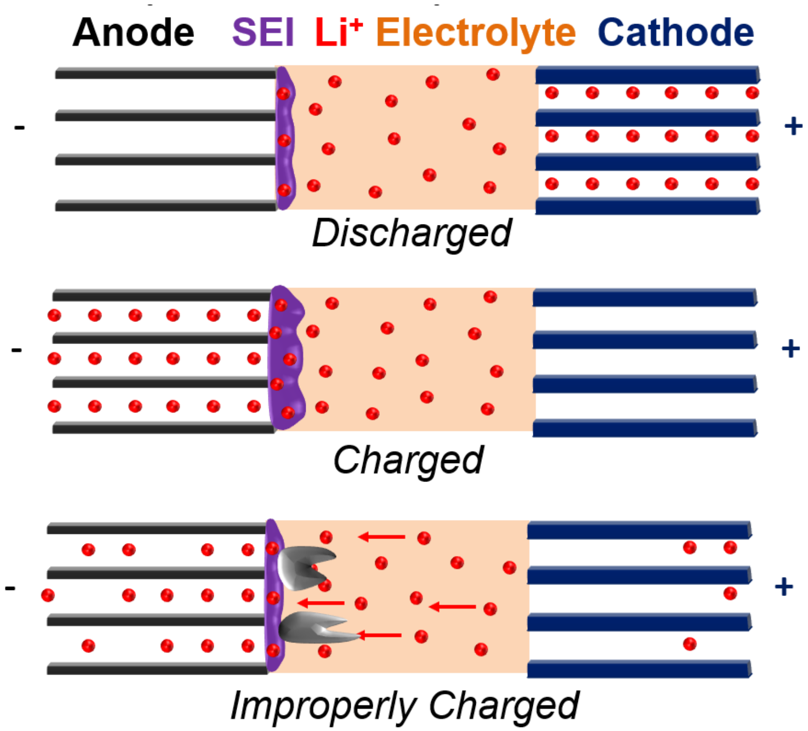
[1] Department of Mechanical Engineering, Iowa State University, Ames, IA, 50011, USA
[2] Chemistry Division, U.S. Naval Research Laboratory, Washington, D.C., 20375, USA
[3] EXCET, Inc., Springfield, VA, 22151, USA
[4] School of Mechanical Engineering, Purdue University, West Lafayette, IN, 47097, USA
[5] School of Ocean and Earth Science and Technology, Hawaii Natural Energy Institute / University of Hawaii at Manoa, Honolulu, HI, 96822, USA

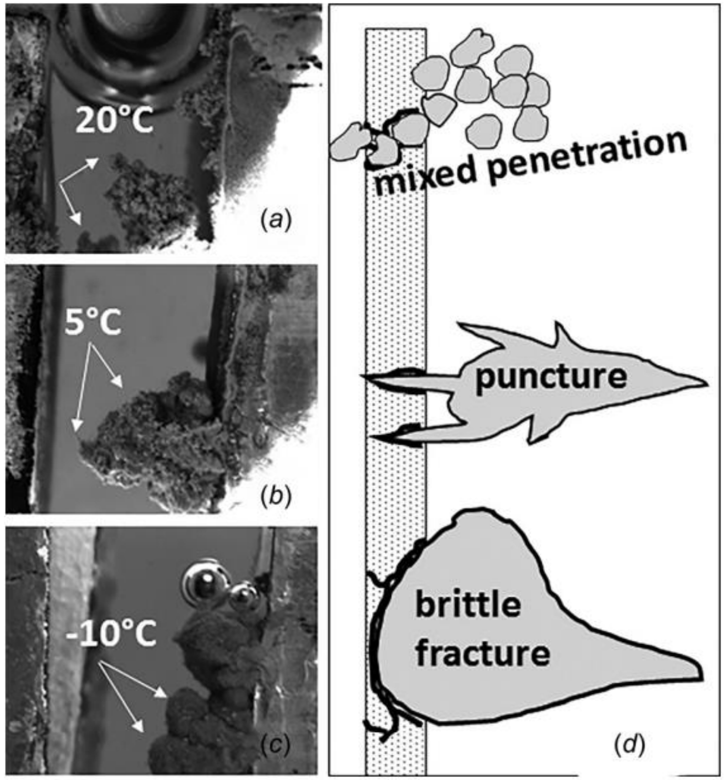
Temperature-dependent Electrochemistry in Li-ion Batteries

- Modern Li-ion batteries consist of a graphite anode, metal oxide cathode, and an electrically insulating separator soaked with an organic solvent-based electrolyte
- Li-ion electrochemistry and lithium morphology are heavily dependent on local thermal conditions



Palacín & de Guibert, *Science*, 2016

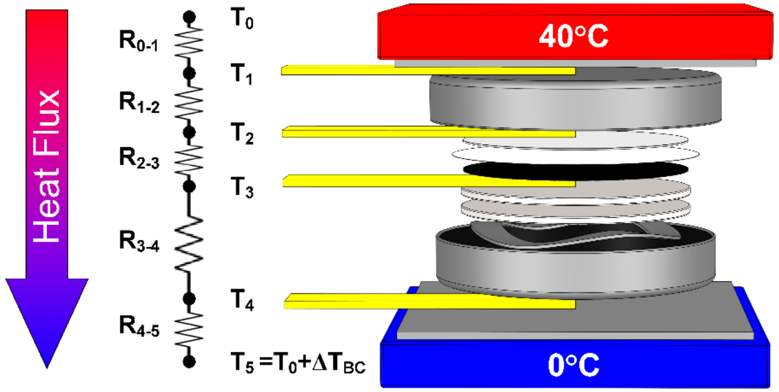


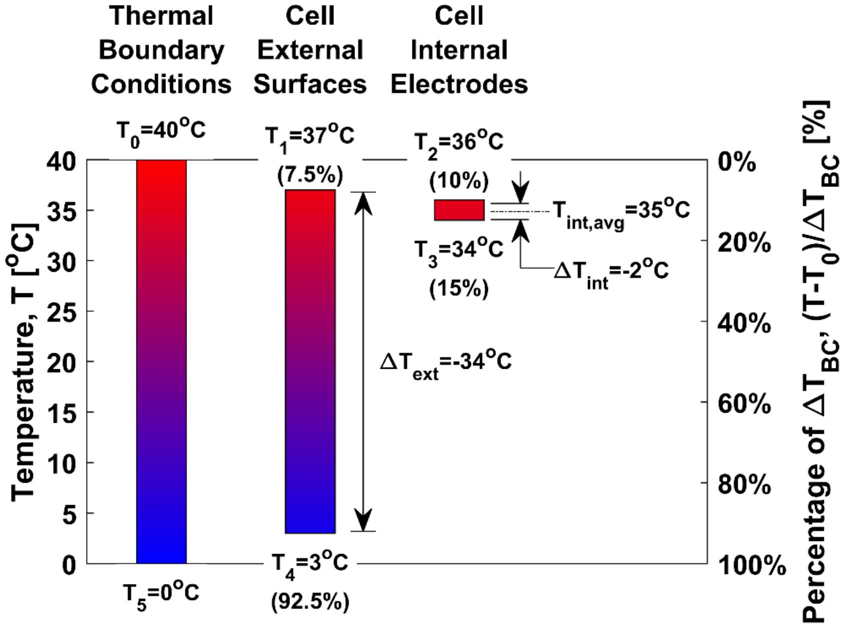


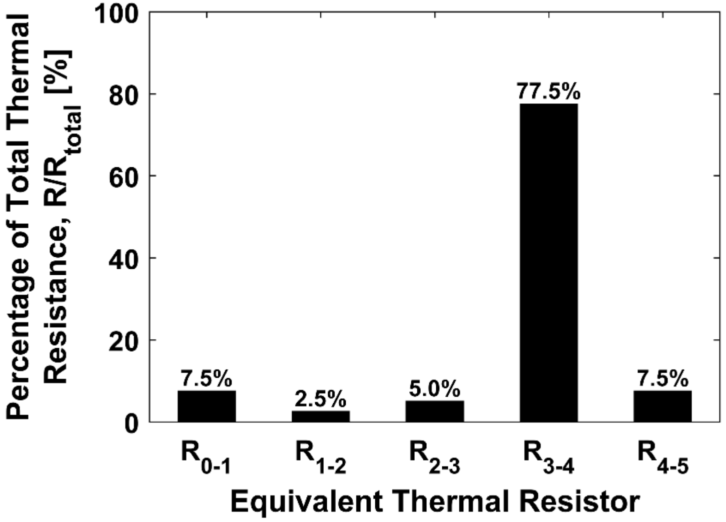
Love, C.T., *JEECS*, 2016

Application and Measurement of Interelectrode Thermal Gradient (ITG)

- Instrumented coin cells to enable local temperature measurement at each electrode
- Applied a thermal gradient using two heat exchanger plates



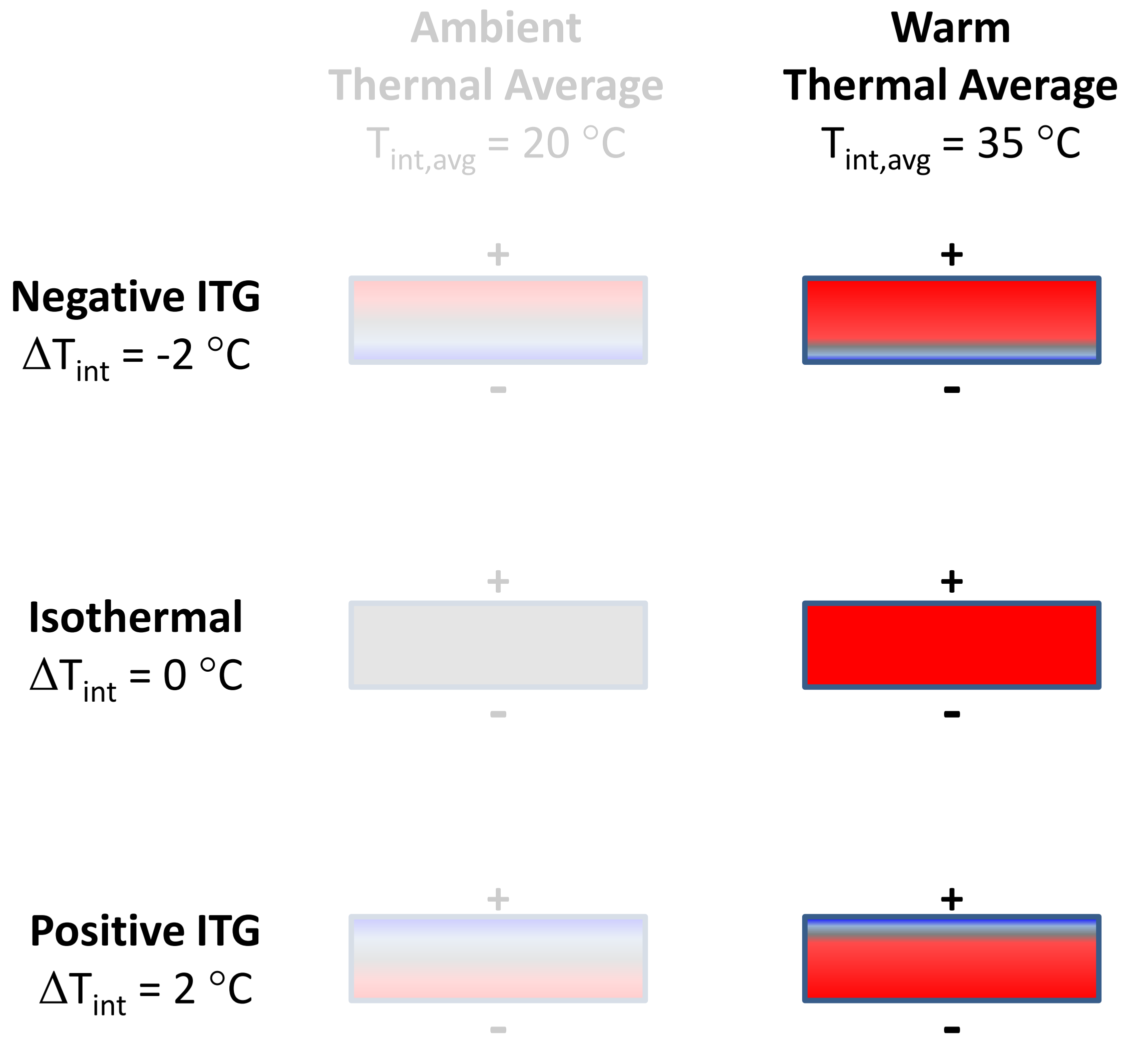




Carter, Kingston, Atkinson, Parmananda, Dubarry, Fear, Mukherjee, and Love, Directionality of thermal gradients in lithium-ion batteries dictates diverging degradation modes, *Cell Reports Physical Science*, 2021, 2(3) 100351.

ITG Design of Experiments

- NMC/graphite coin cells
- Investigate the effect of:
  - Thermal average: 20, and 35 °C
  - ITG: -2, 0, 2 °C
- Perform galvanostatic cycling at C/5 for 20 cycles
  - Voltage limits: 3.0 and 4.3 V
  - Current: 0.74 mA



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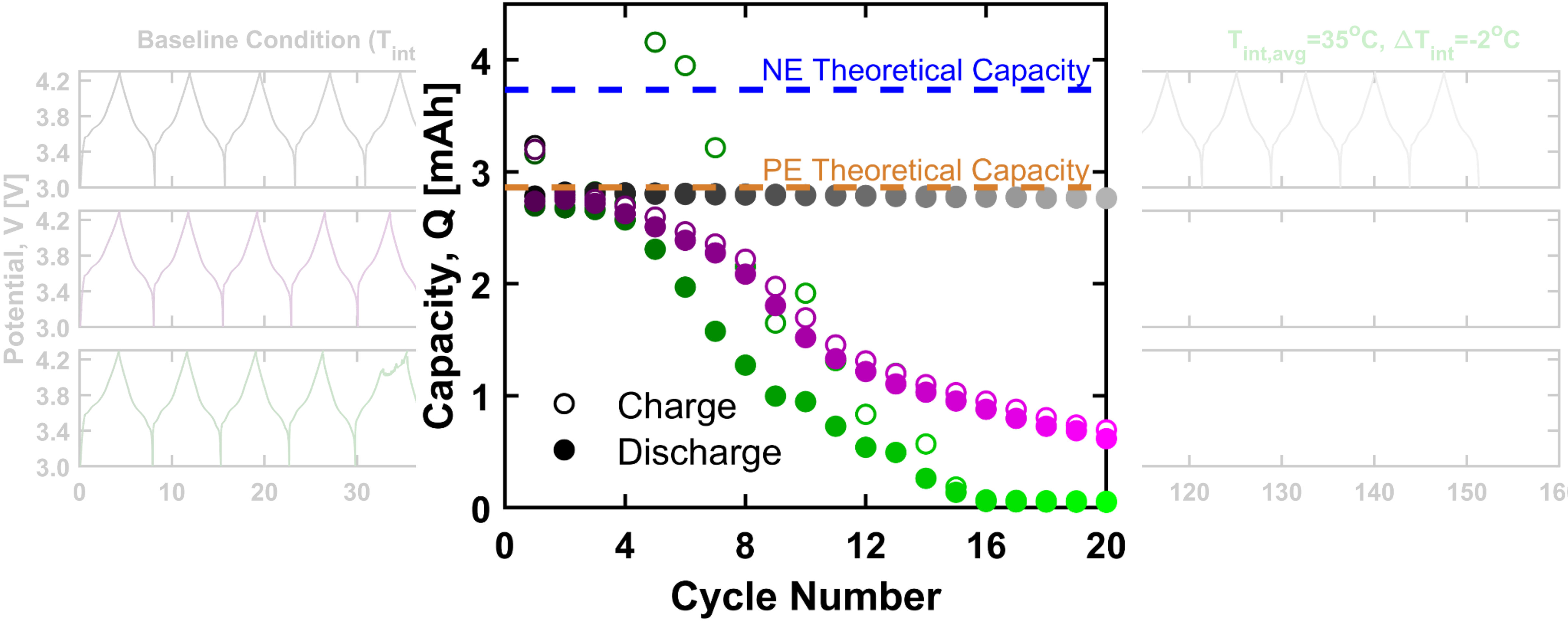
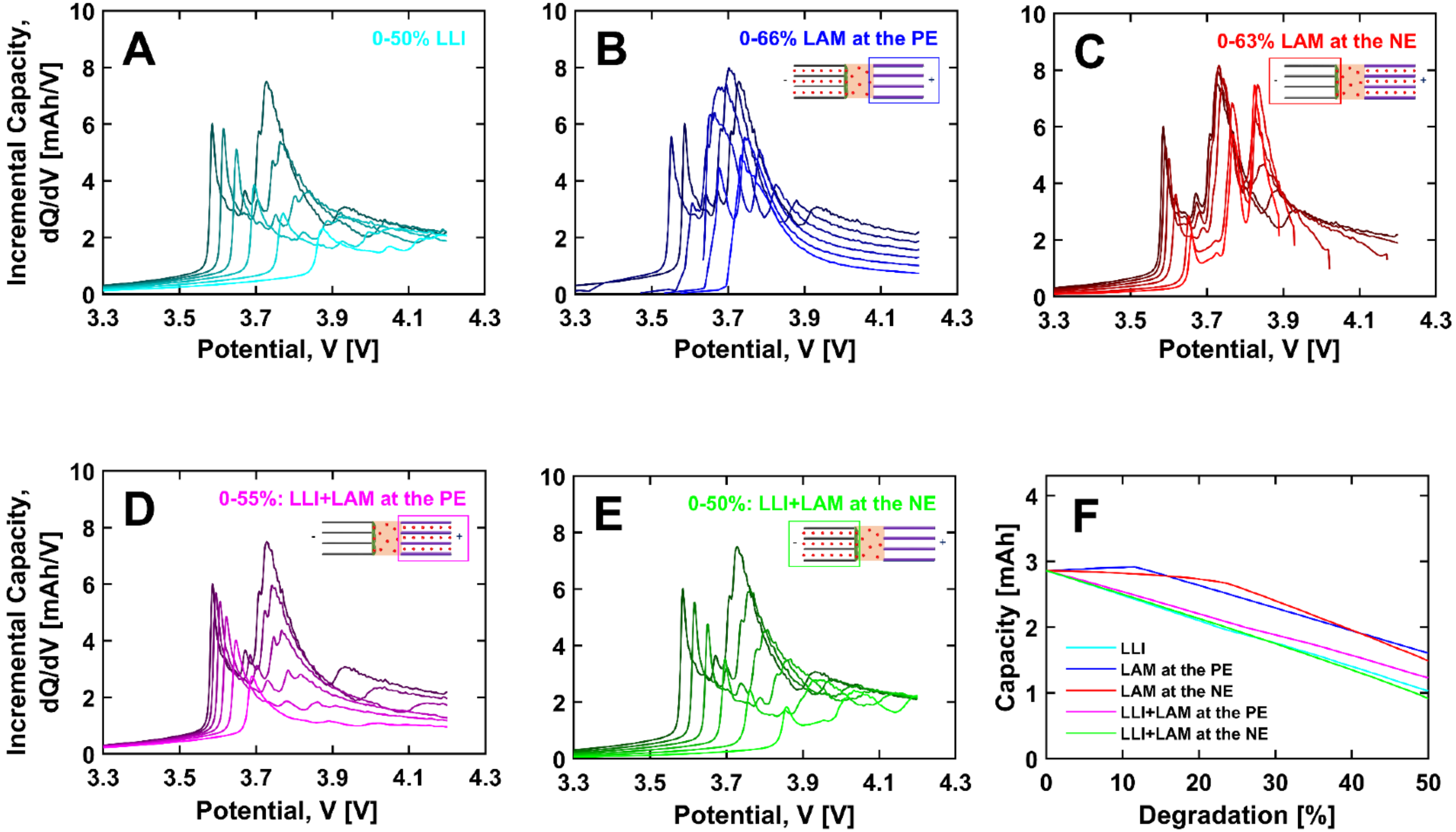
Electrochemical Cycling Behavior



Simulated Degradation Modes

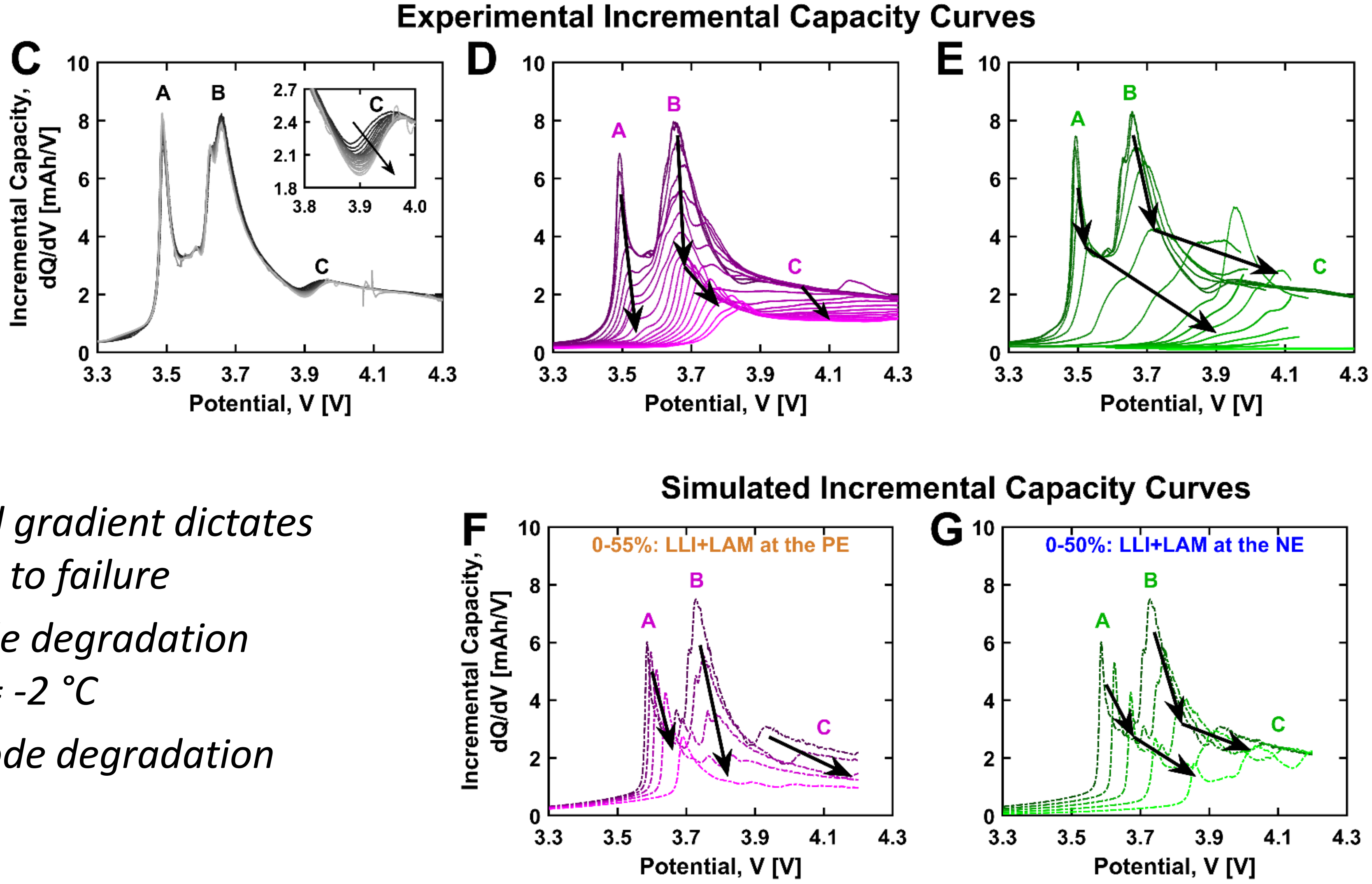
- Simulated modes of degradation are used to identify the experimentally observed degradation modes using IC analysis
- Primary degradation modes:
  - **A**: Loss of lithium inventory (LLI)
  - **B**: Loss of active material at positive electrode (LAM at PE)
  - **C**: Loss of active material at negative electrode (LAM at NE)
- Mixed degradation modes:
  - **D**: LLI + LAM at PE
  - **E**: LLI + LAM at NE



See Dubarry, Truchot, and Liaw (2012) *JPS* for additional details regarding IC simulation

Identifying Degradation Modes Using Incremental Capacity Analysis

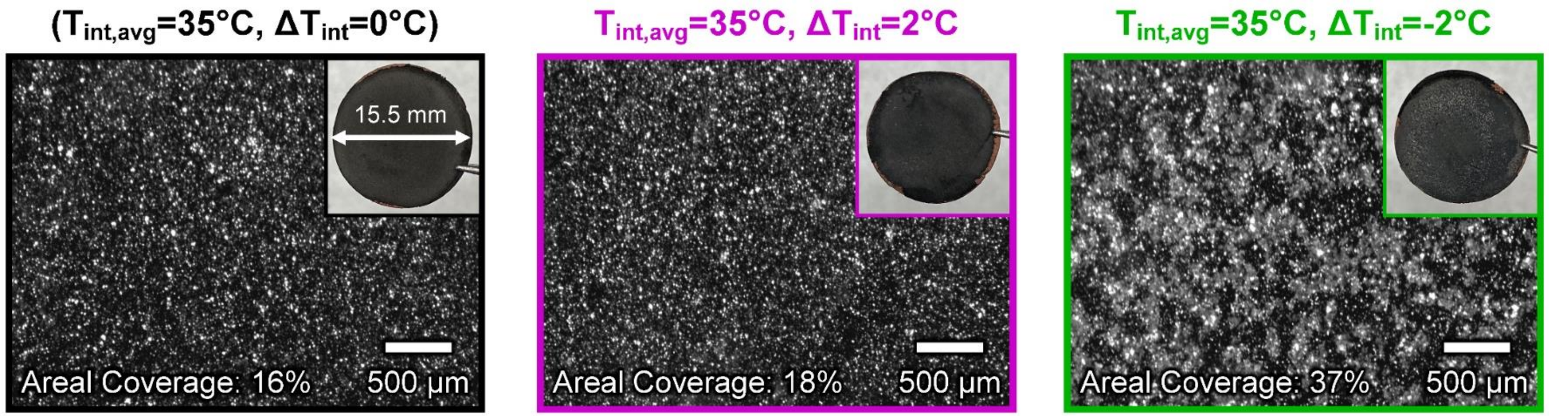
- Compare experimental IC curves to simulated ones with explicit degradation modes

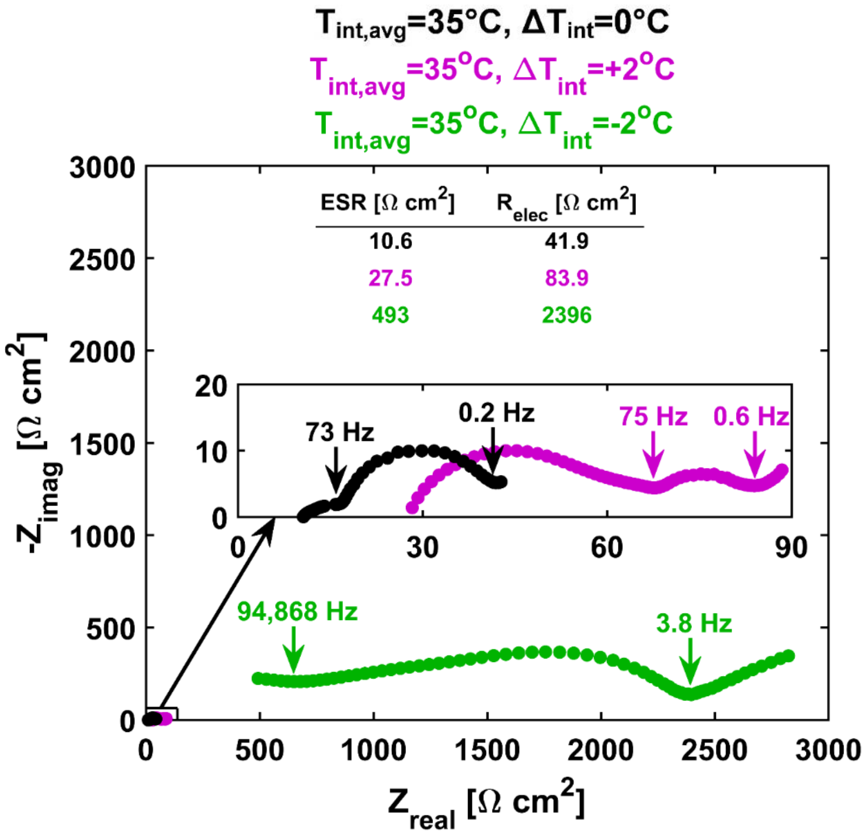


- Conclusion:
  - *Directionality of thermal gradient dictates which electrode is prone to failure*
  - *Rapid, negative electrode degradation with Li plating for ΔT_{int} = -2 °C*
  - *Gradual, positive electrode degradation for ΔT_{int} =+2 °C*

Post-mortem Optical Analysis and EIS

- Plated Li identifiable in optical images
- Large shift in EIS response for cells cycled at ΔT_{int} = -2 °C





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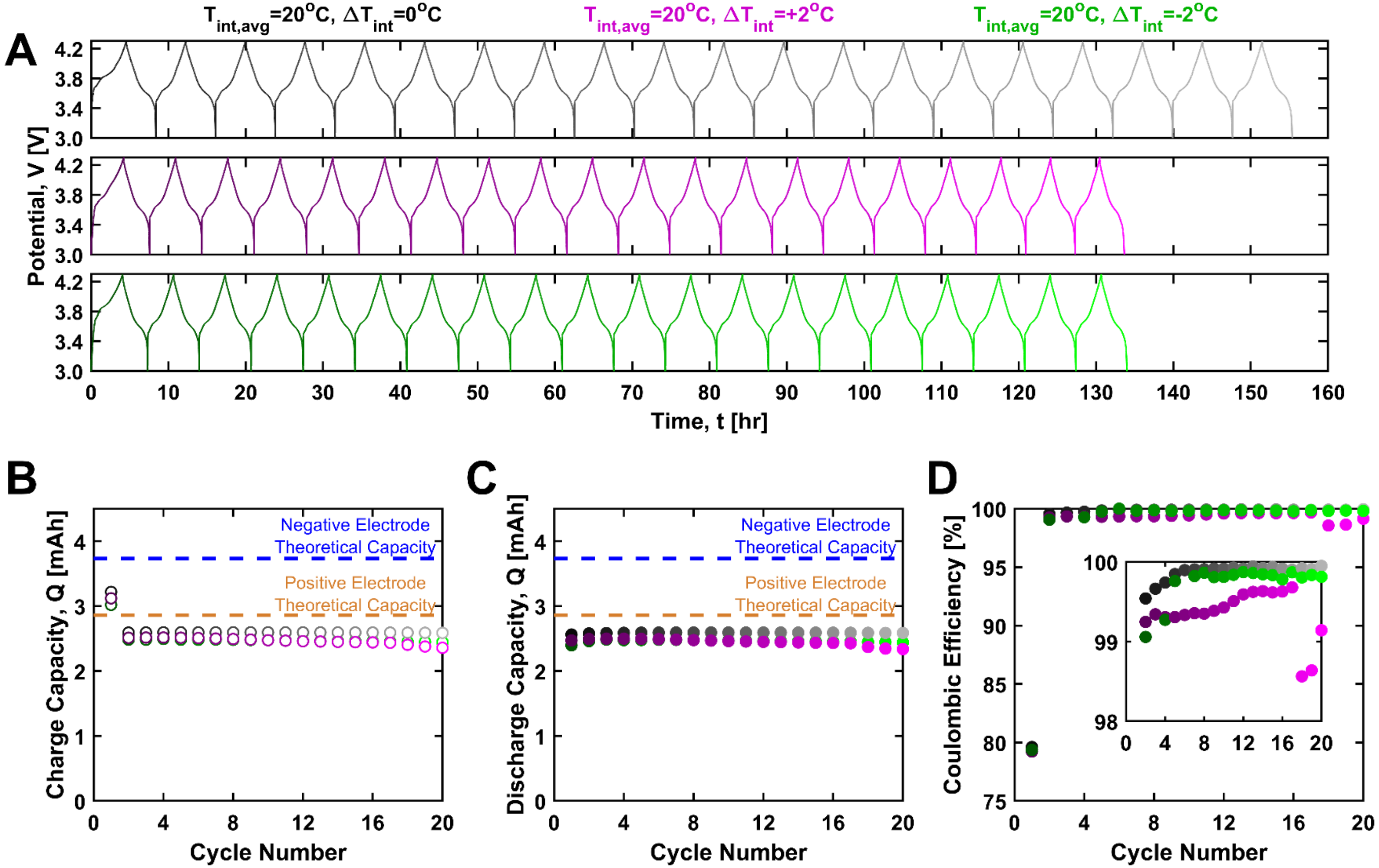
Electrochemical Behavior at Ambient Temperatures

- Cells cycled at an ambient thermal average (20 °C) had no degradation



Primary Takeaways

- Li-ion electrochemistry is dependent on ITGs
- Subjecting Li-ion cells to a warm, ITGs degrades the cell relative to isothermal conditions
- Directionality of the ITG dictates the degradation mode
- Accelerated aging of Li-ion cells can accomplished by applying an ITG
- Improved thermal management strategies are needed to ensure safe and reliable operation of Li-ion batteries

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Directionality of thermal gradients in lithium-ion batteries dictates diverging degradation modes

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