

# Impact of Additives on Cycle Life of Layered Metal Oxide Cathode in Localized High Concentration Electrolyte



Marcos Lucero, Fred Omenya, Xiaolin Li, David Reed

Pacific Northwest  
NATIONAL LABORATORY

Pacific Northwest National Laboratory (PNNL), Richland, Washington, USA

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## Abstract

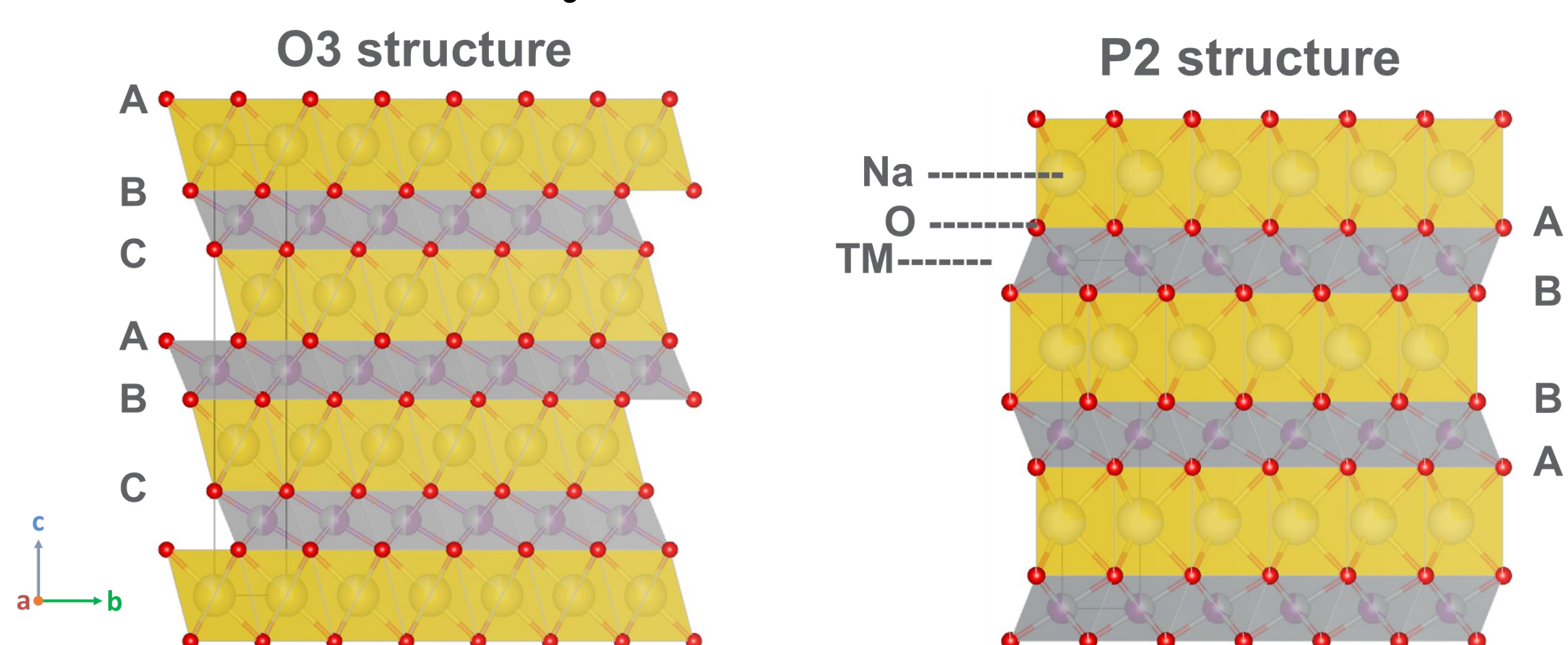
Layered metal oxides are the leading cathode materials for practical Sodium-ion batteries (SIBs). However, some drawbacks include fast capacity fade due to side reactions with the electrolyte, especially with high voltage ( $>4V$  vs  $Na^+/Na$ ). Localized high-concentration electrolytes (LHCE) have been shown to mitigate side reactions with the positive electrode due to their unique local structure, which favors the formation of a robust inorganic cathode electrolyte interphase (CEI). This study investigates the role of additives in LHCE on further extending the cycle life of layered oxide cathodes. Systematic characterization, including galvanostatic cycling, electron microscopy, X-ray diffraction, gives insights into the mechanism for extended cycle life.

## Objectives and Methodology

- Examine effect of additive on cycle life of sodium-ion batteries
- Reveal role of additive on cathode-electrolyte interphase (CEI).
- Understand the evolution of the cathode structure and effect on cycle life.

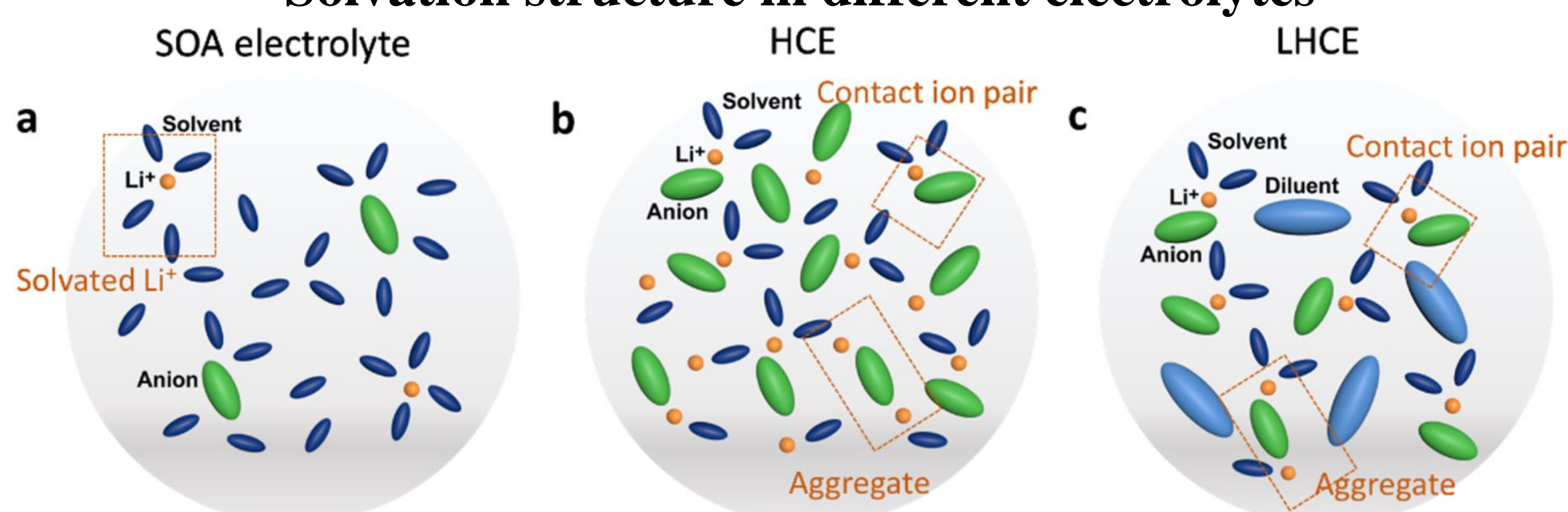
## Introduction

### Layered Oxide Cathodes



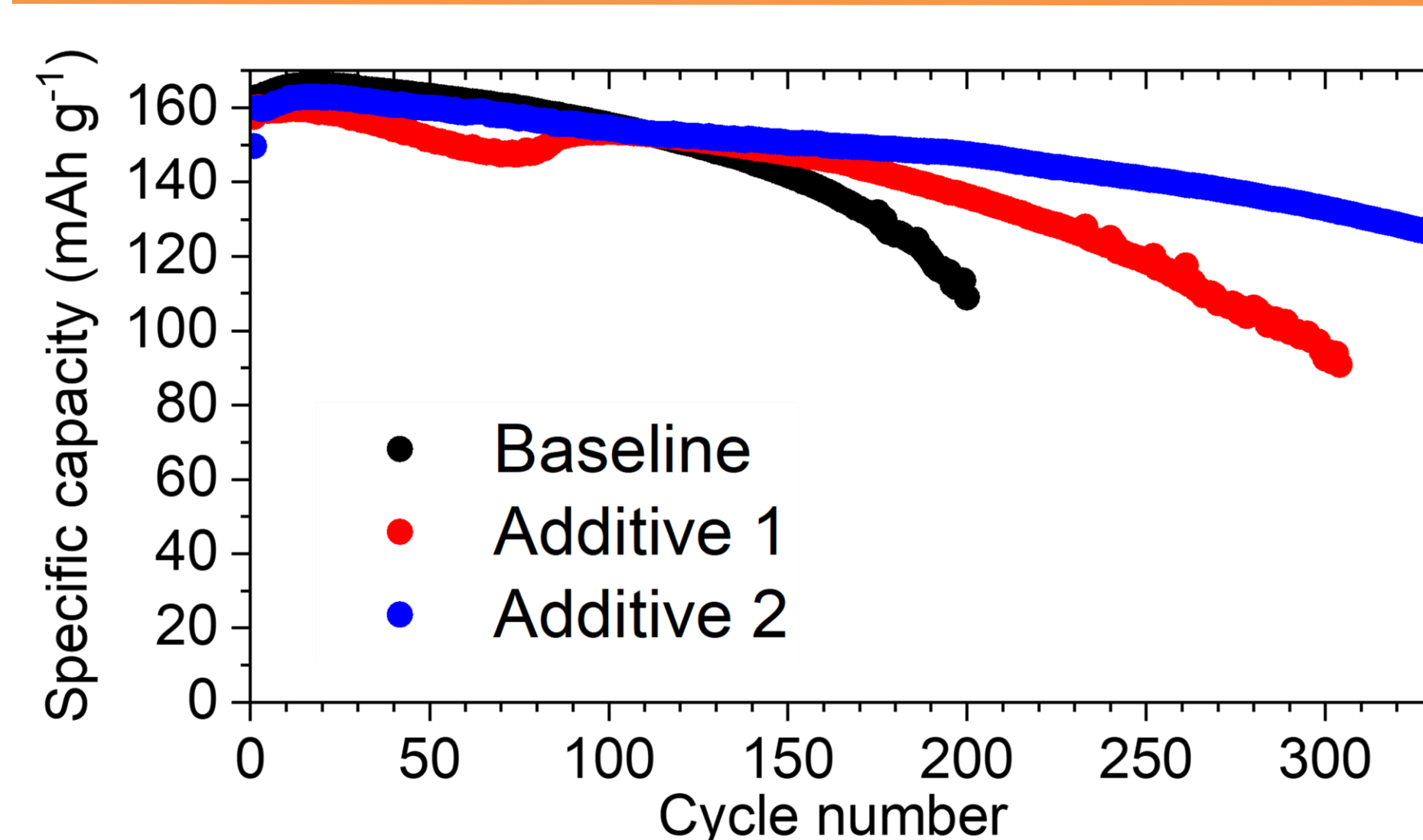
- Promising class of materials that has potential for low cost, long life and high energy dense batteries.<sup>1,2</sup>
- Key challenges include irreversible phase transformation and electrolyte degradation during cycling at high voltages resulting in poor cell performance.

### Solvation structure in different electrolytes

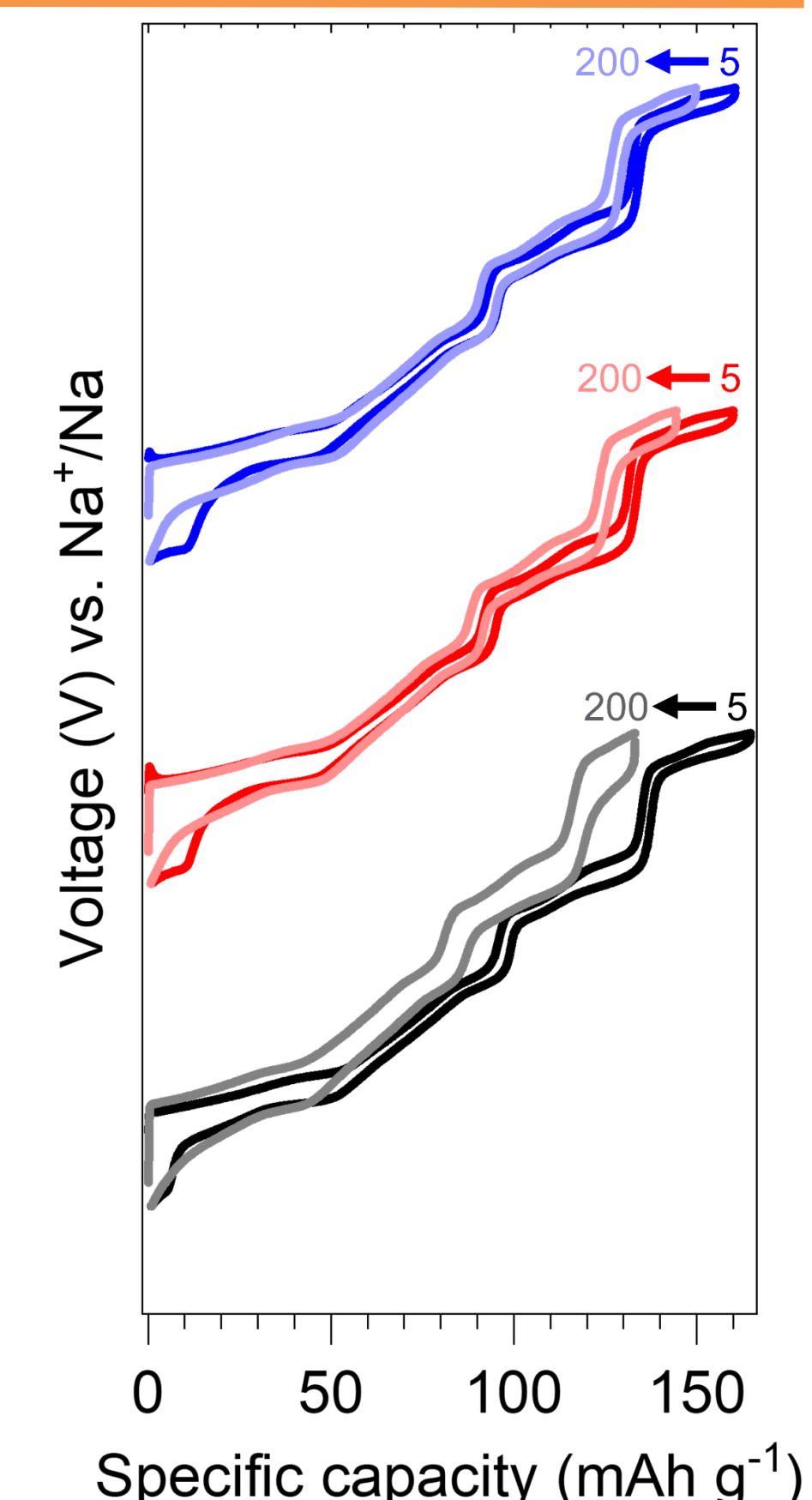


- Solvation structure dictates interphase chemistry and plays major role in cell performance
- SOA electrolyte – Solvent primarily occupies coordination shell. Solution contains sufficient free-solvent molecules.
- High concentration (HCE) – Lack of free-solvent leads sharing and merging of coordination shells (e.g., contact-ion pairs).<sup>3</sup>
- Localized high concentration (HCE) – High solubility salt, solvent and non-solvating solvent. Localized contact-ion pairs surrounded by non-solvating solvent.<sup>3</sup>

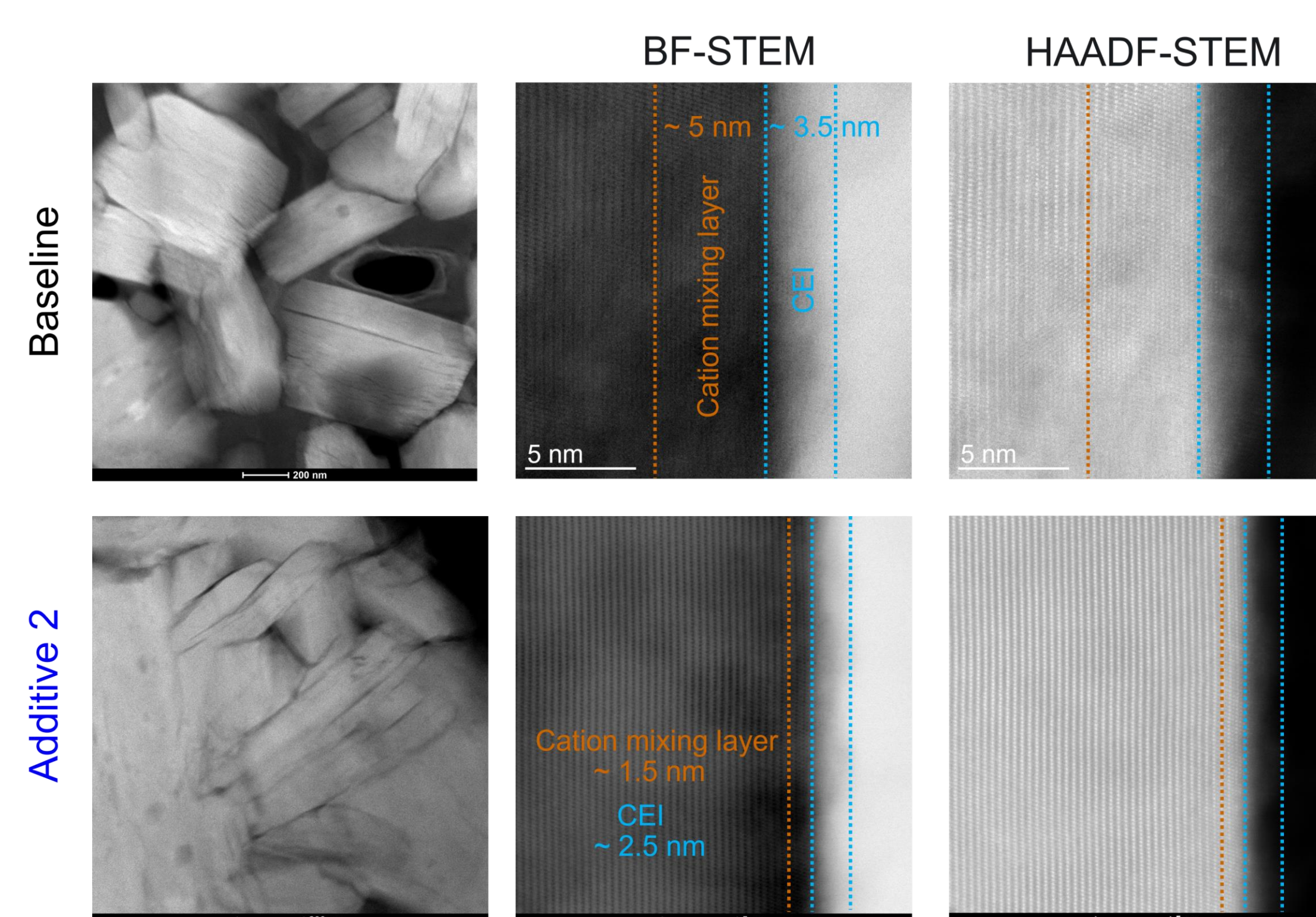
## Results and Discussion



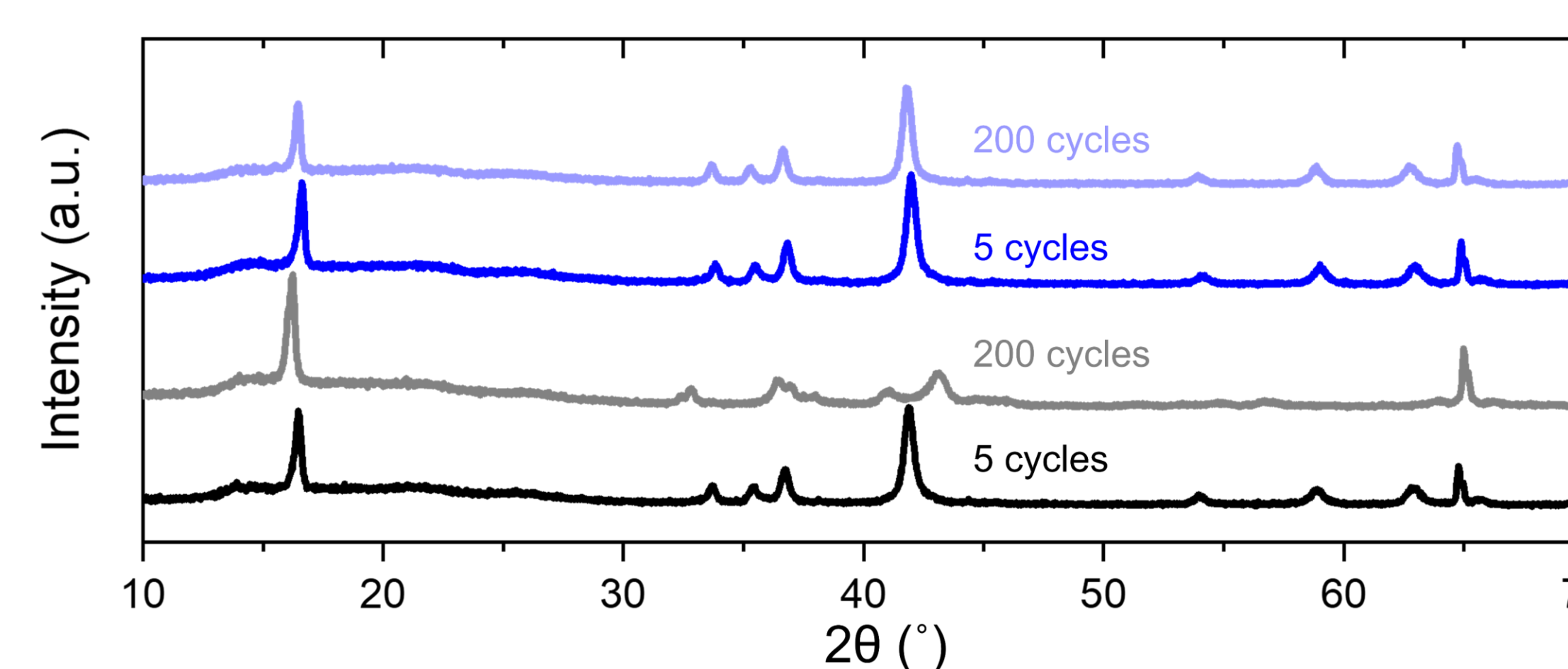
- Modified electrolyte can extend cycle life (@ 80%) from 170 to 320 cycles.
- Voltage curves at early cycles reveal similar shape. Later cycles reveal shortening of high voltage plateau and increased polarization.



## Physical Characterization



- Baseline electrolyte: Thick cation mixing layer and CEI
- Modified electrolyte: Thin cation mixing layer and CEI.
- Solvation structure in electrolyte plays major role in interphase formation and subsequently the cycle life and performance of layered oxide cathodes.



- Baseline electrolyte (black) shows irreversible structural changes @ 200 cycles
- Modified electrolyte (blue) reversible structural changes @ 200 cycles

## Summary and Future Work

- Modifying the electrolyte can significantly improve the cycle life of layered cathodes for high-performance sodium-ion batteries.
  - Appropriate additive extended cycle life by ~2x.
- Future work will focus on large cell demonstration and advancing the material sustainability and performance.

## Acknowledgements

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## References

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