INL/CON-19-53477-Revision-2

Lithium Metal Electrode Understanding Its Unique Characteristics and Functions

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Lithium Metal Electrode — Understanding Its Unique Characteristics and Functions

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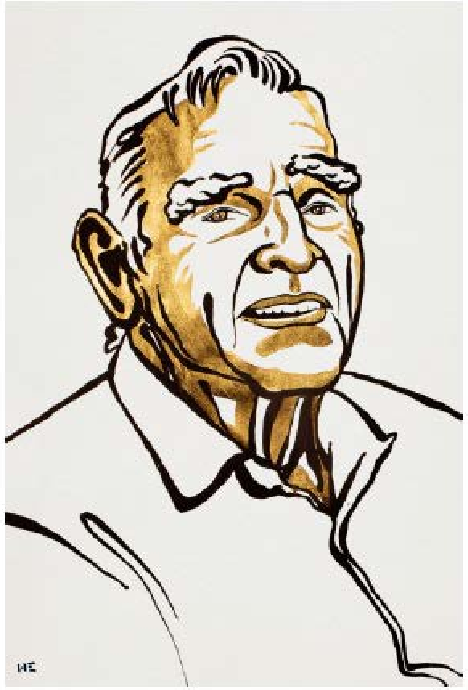
BATTERY500 CONSORTIUM

Congratulations

The Nobel Prize in Chemistry 2019



Ill. Niklas Elmehed. © Nobel Media.

John B. Goodenough

Prize share: 1/3

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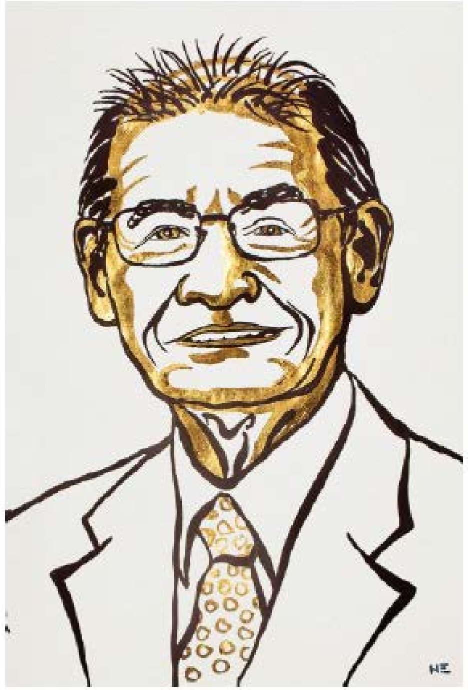

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Akira Yoshino

Prize share: 1/3

MLA style: The Nobel Prize in Chemistry 2019. NobelPrize.org. Nobel Media AB 2019. Sun. 13 Oct 2019. <https://www.nobelprize.org/prizes/chemistry/2019/summary/>

Remember the Pioneers

Carl Wagner in 1970’s

https://www.ncbi.nlm.nih.gov/pmc/articles/PMC5090311/

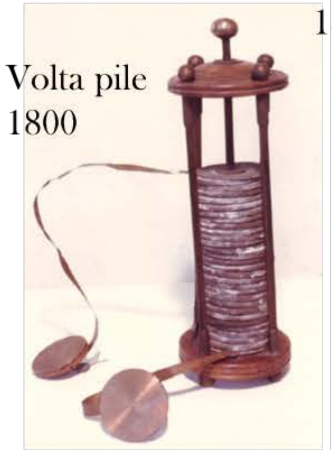
Bob in 1980’s

https://profiles.stanford.edu/robert-huggins

and, many others…

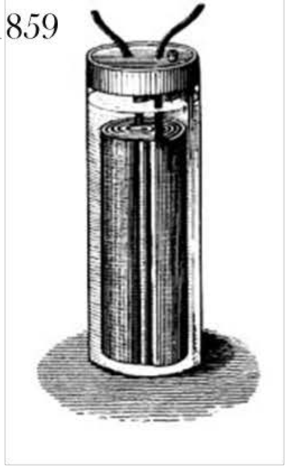
Make the Rechargeable Li Battery Great Again

Franklin battery
1749
(capacitor)



Volta pile
1800

Planté lead-acid
1859

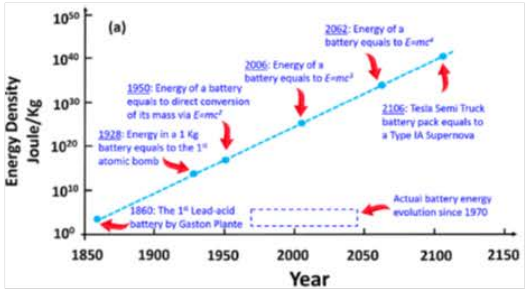


Matsushita's Li
primary cell
1972

Moser's solid
Li-I cell
1972

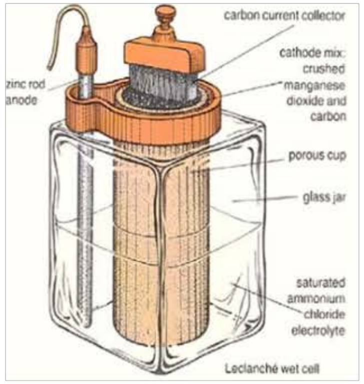
Sony's Li-ion
battery cell
1991

Pushing the limits
2019 ——



1800 1900 2000

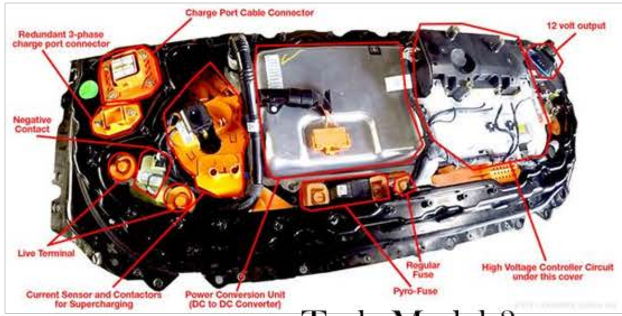
Baghdad battery
150 BC – 640 AD



Leclanché cell
1866

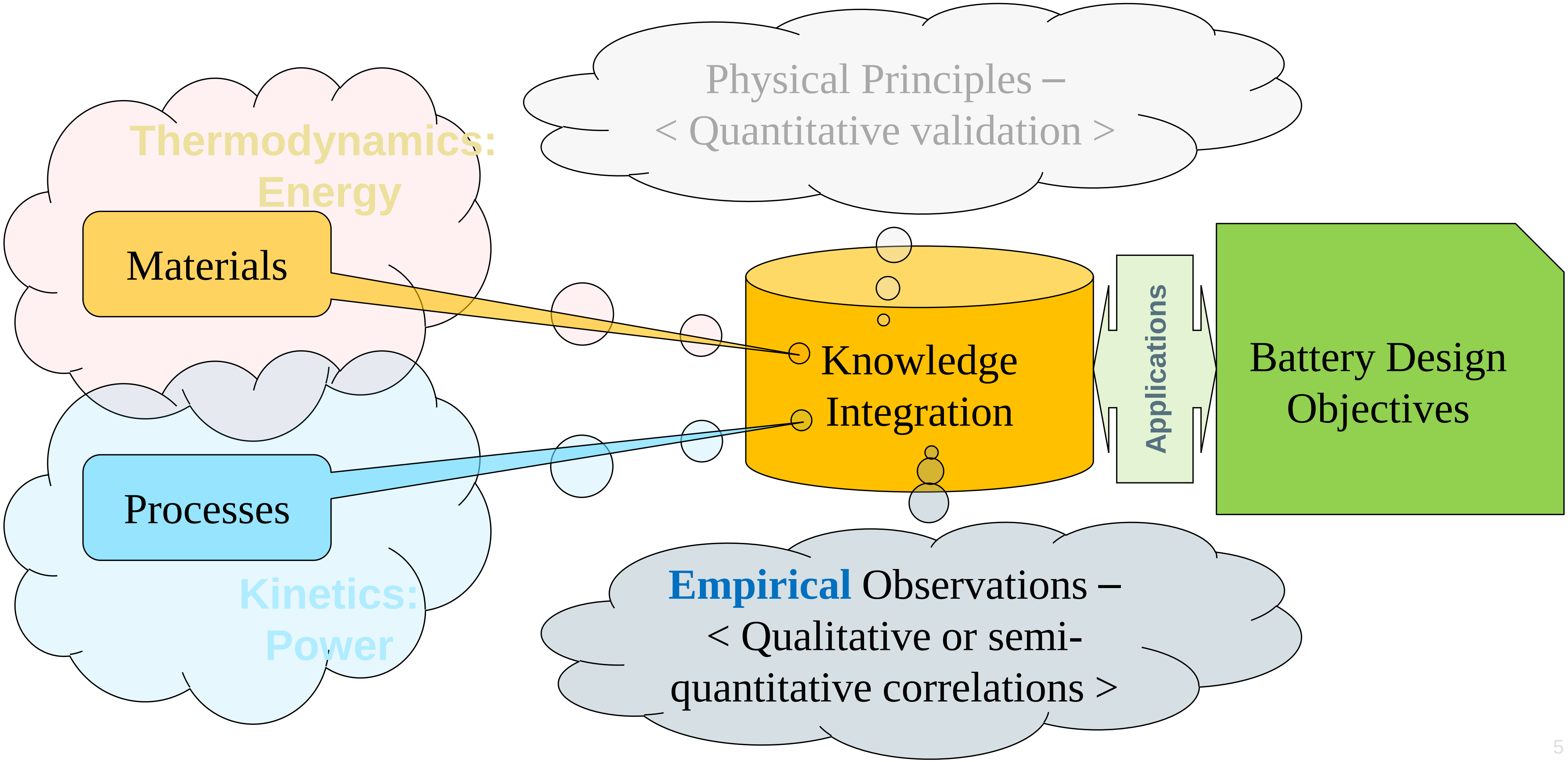
Whittingham's
organic rechargeable
Li battery (RLB)
1977

Armand's polymer
electrolyte RLB
1978



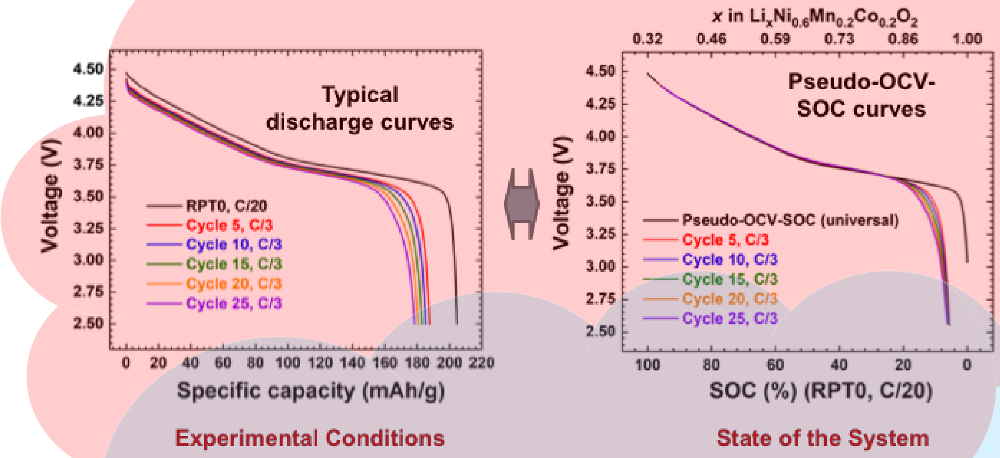
Tesla Model 3
2018

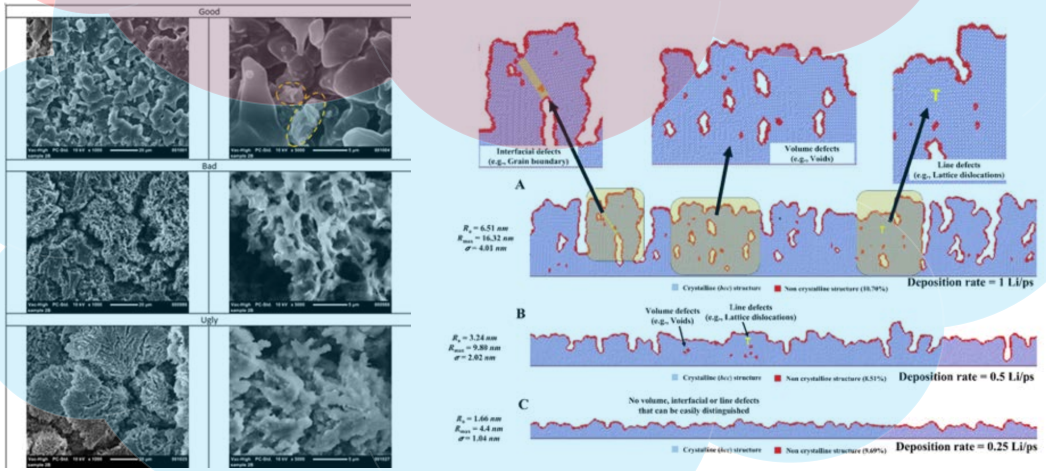
Conventional Approach



Battery Failure Analysis to Life Prediction

Thermodynamic Attributes:





Kinetic Attributes:

Physical Principles –
< Quantitative validation >

Knowledge Integration

Applications

Battery Design Objectives

Empirical Observations –
< Qualitative or semi-quantitative correlations >

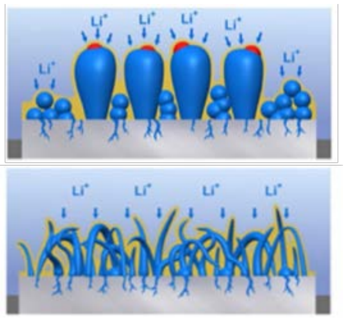
Li Stripping & Deposition – A Perspective

- **Challenging issues for rechargeable Li batteries with high-energy density and higher cycle life**

A dense Li deposition is needed

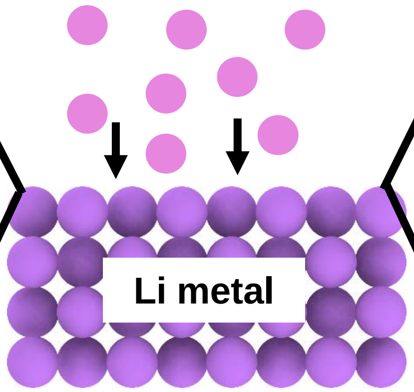
Dendrite

- Fundamental mechanisms unclear
- Boundary condition is difficult to define

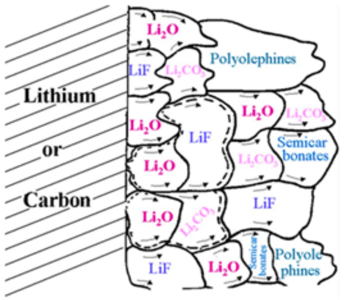


Layla, et al. *Scientific reports* 6 (2016)

Li deposition



Solid Electrolyte Interphase

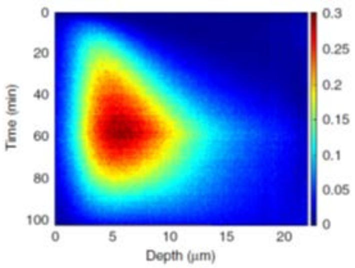


- Complex phenomena
- Difficult to characterize

Peled, *Journal of The Electrochemical Society* 164.7 (2017): A1703-A1719

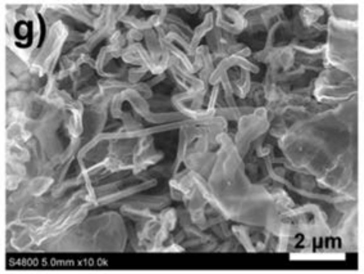
Localized Effects

- Uncontrollable experimental parameters



Shasha, et al. *Nature communications*9.1 (2018): 2152

Morphology & Porosity

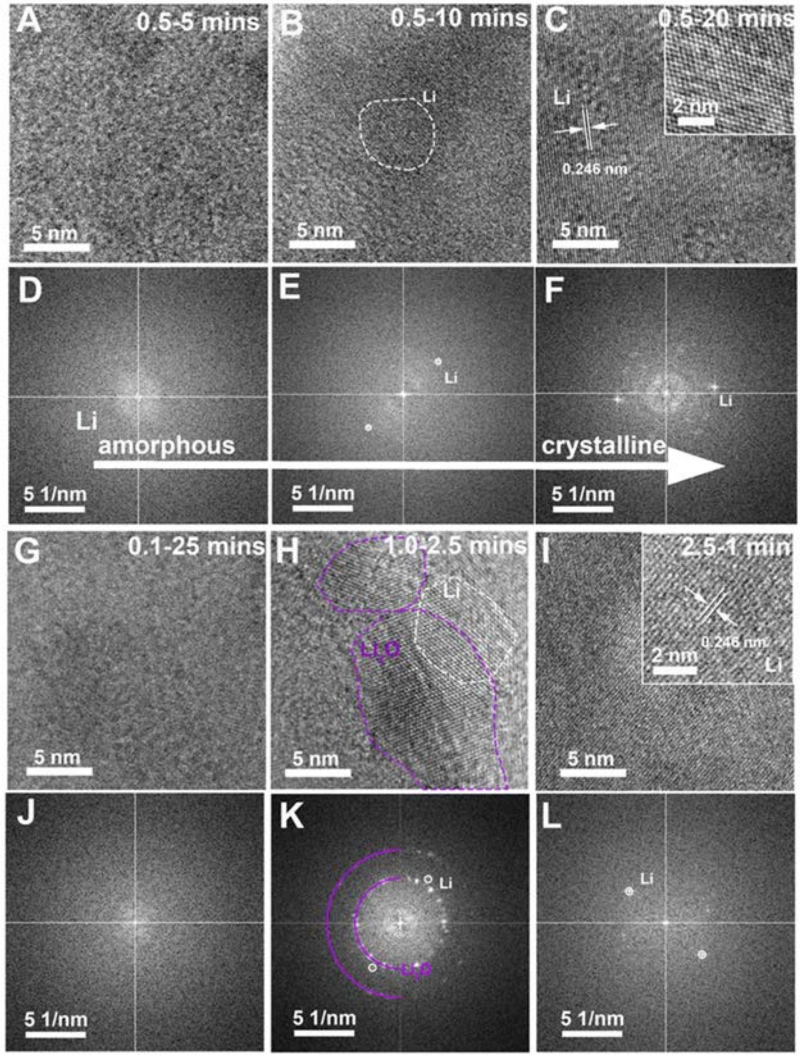


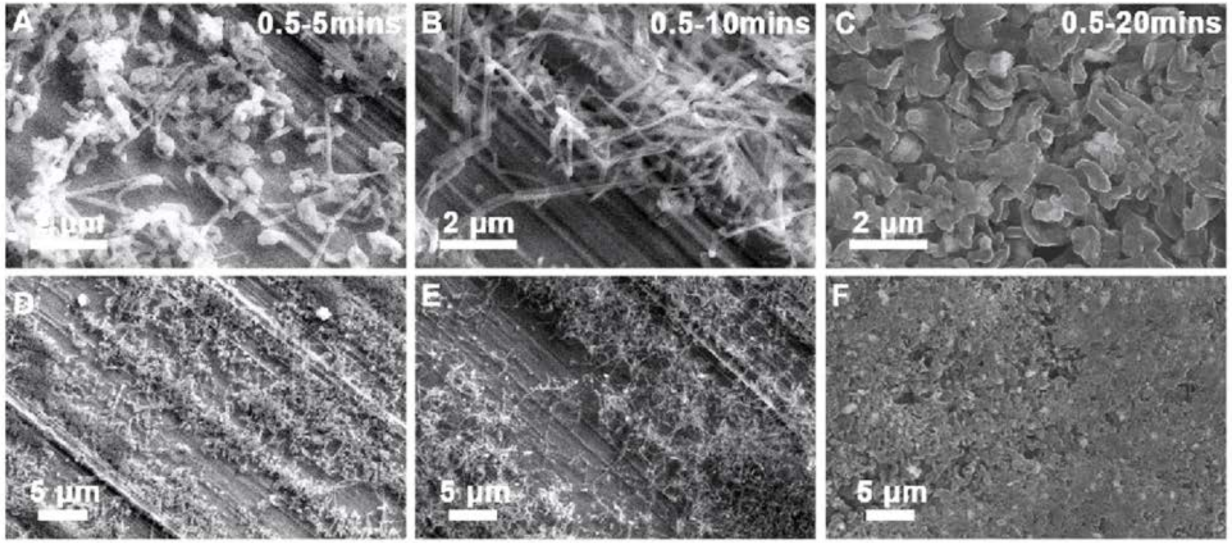
- Variability in morphology
- Undesirable effects

- Li, *Nano Energy* 32 (2017): 241-246.

How different is the fundamental process involved in these phenomena?

Cryo-TEM Observations on Li Deposition

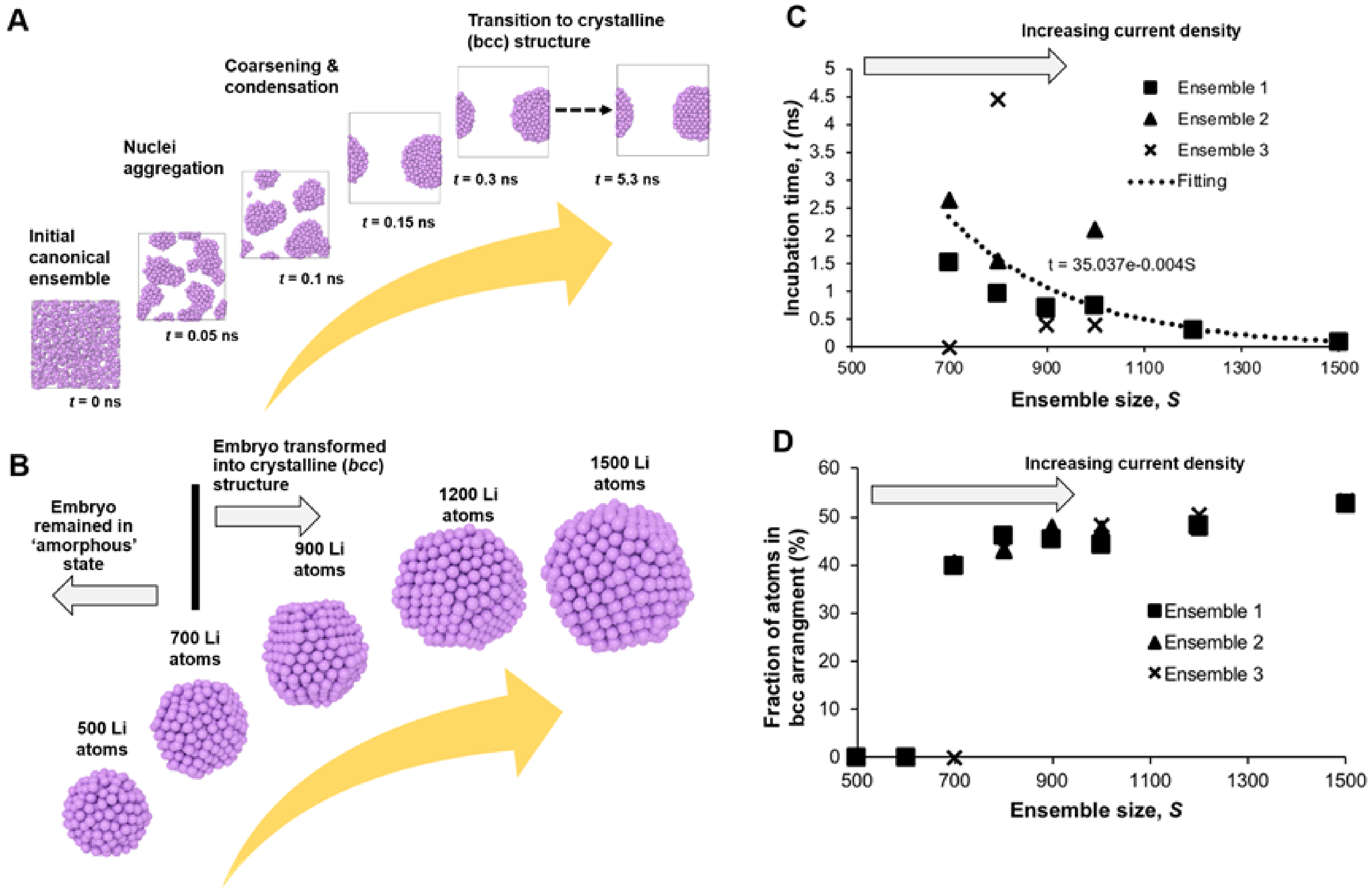




- **Morphology of LME depends on current density and duration**
- **Amorphous Li (a-Li) was found !**
- **Lower the current density more a-Li found !**

— submitted to *Science*

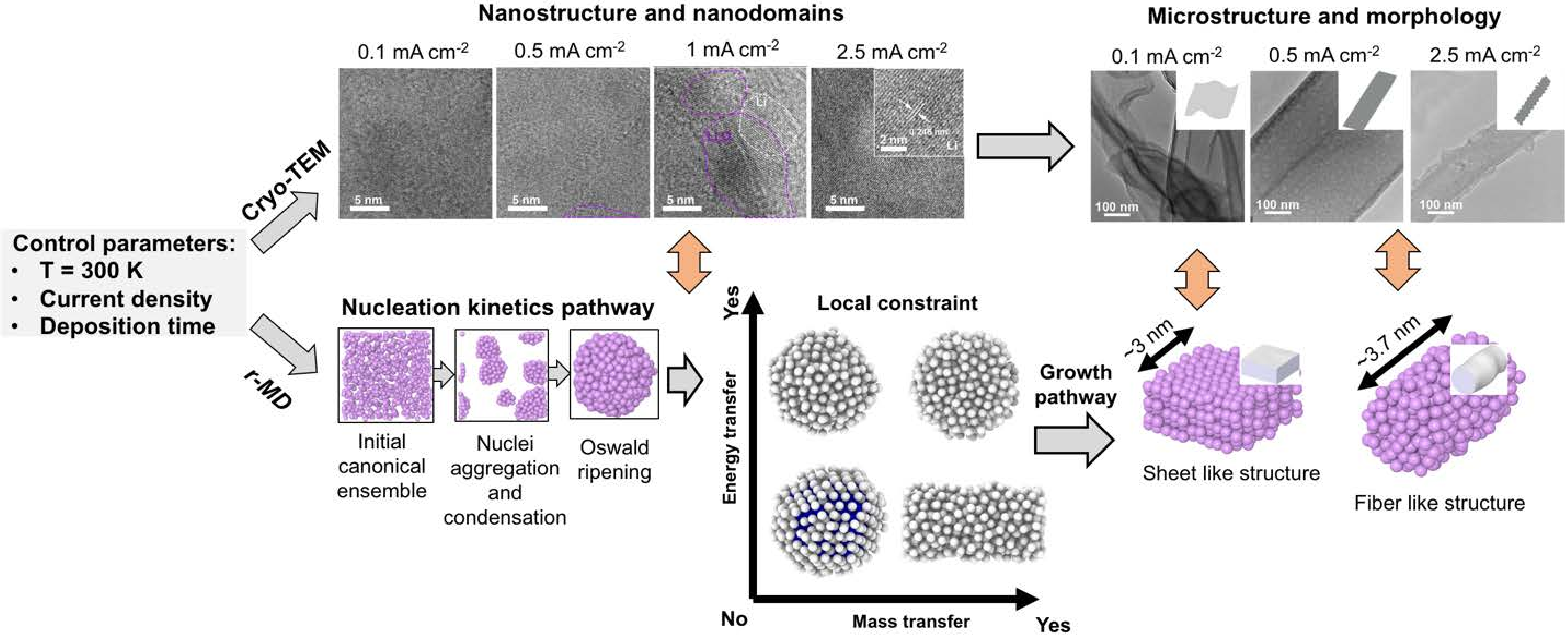
Understanding of Reaction Kinetics via Reactive MD Simulation — Nucleation



The r-MD simulation suggests:

- **Reaction kinetics of nucleation is sensitive to Li embryo size, which is related to current density and duration**
- **Below a threshold size the Li deposit is an aggregate of Li in a disordered state — amorphous phase or a-Li**
- **There is an amorphous-crystalline (a-Li to c-Li) transition and incubation**
- **The threshold size is 2-3 nm**

Temporal Evolution of Bulk Microstructure



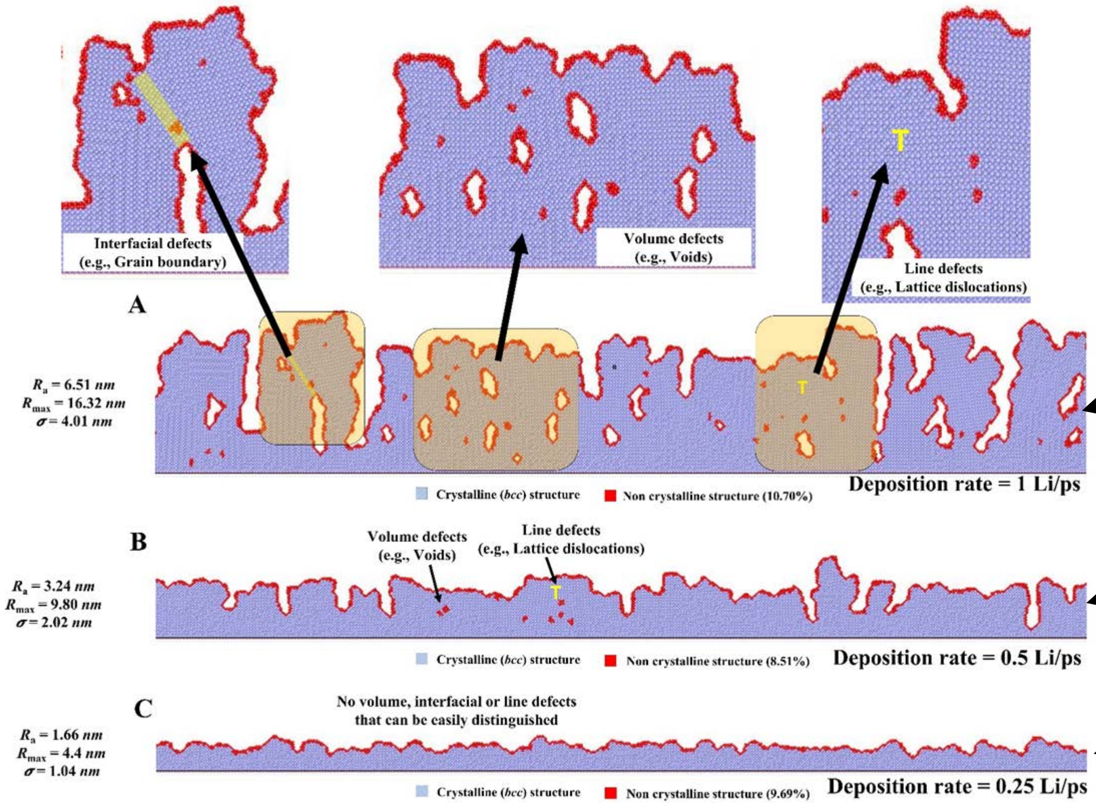
A systematic understanding can enable the identification of kinetic pathways that affects cycle life

Understanding of Reaction Kinetics via Reactive MD Simulation — Growth

- **Surface morphology and bulk properties**

Li deposition rate significantly affects the surface and bulk lithium characteristics

Rate Dependent Surface Morphology and Bulk Properties



Evolution of interfacial, volume and line defects

High deposition rate

Intermediate deposition rate

Lower deposition rate

Higher deposition rates profoundly impact lithium metal surface and bulk lithium metal characteristics

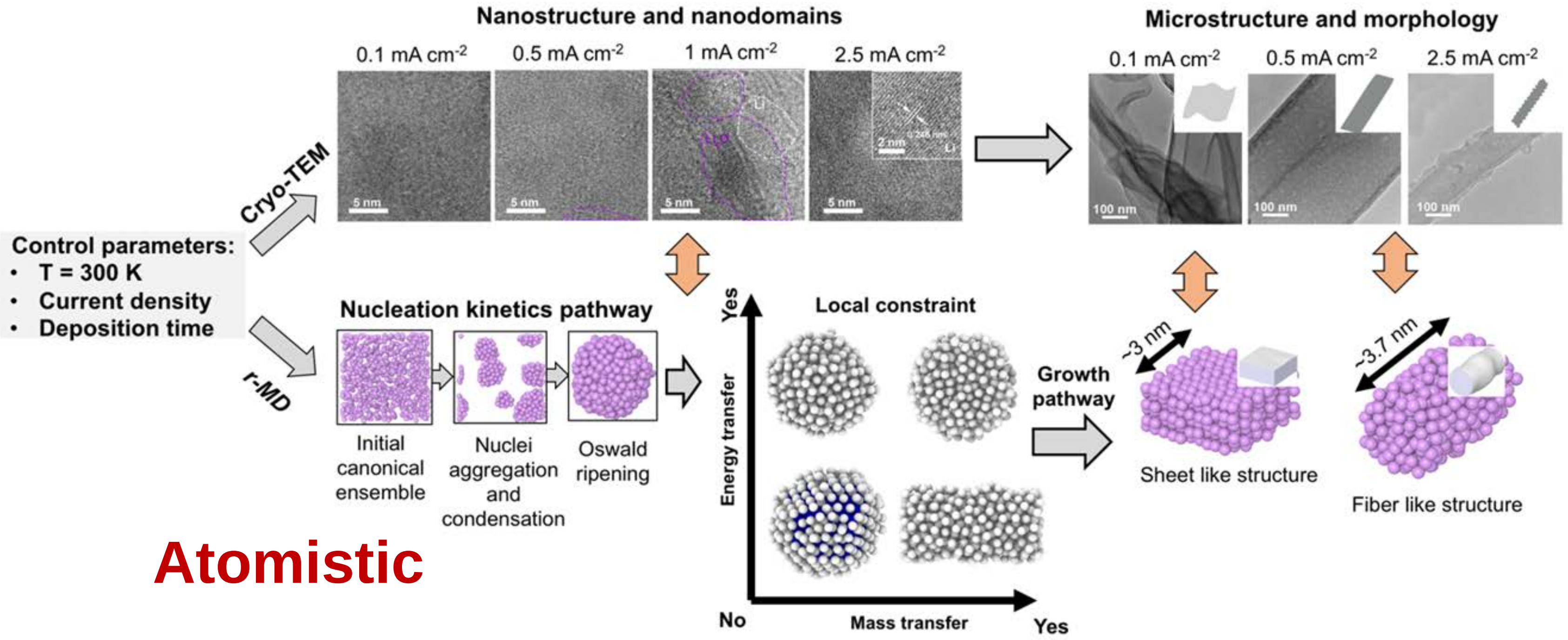
Multi-Scale Fidelity and Reality

- **What governs this multi-scale phenomena is kinetics and the associated energy-transfer process—not thermodynamics.**

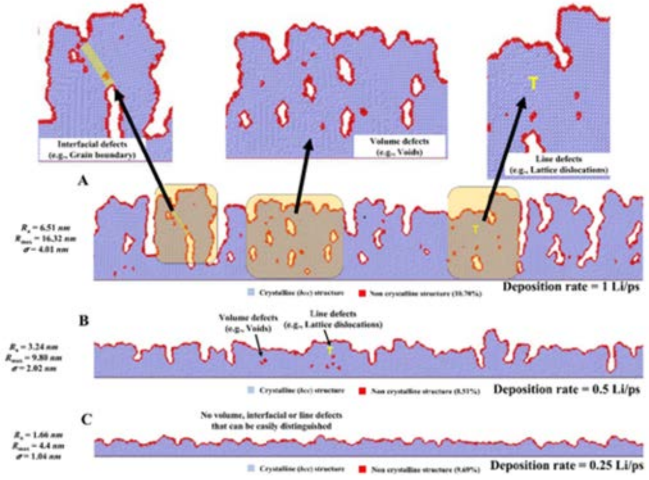
Using Li metal electrode (LME) as a case study

Micro-scale **Meso-scale** **Macro-scale**

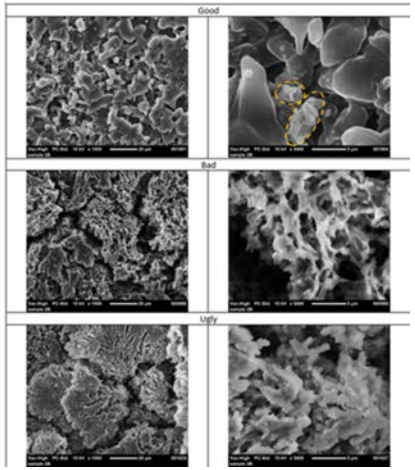
Nano-scale



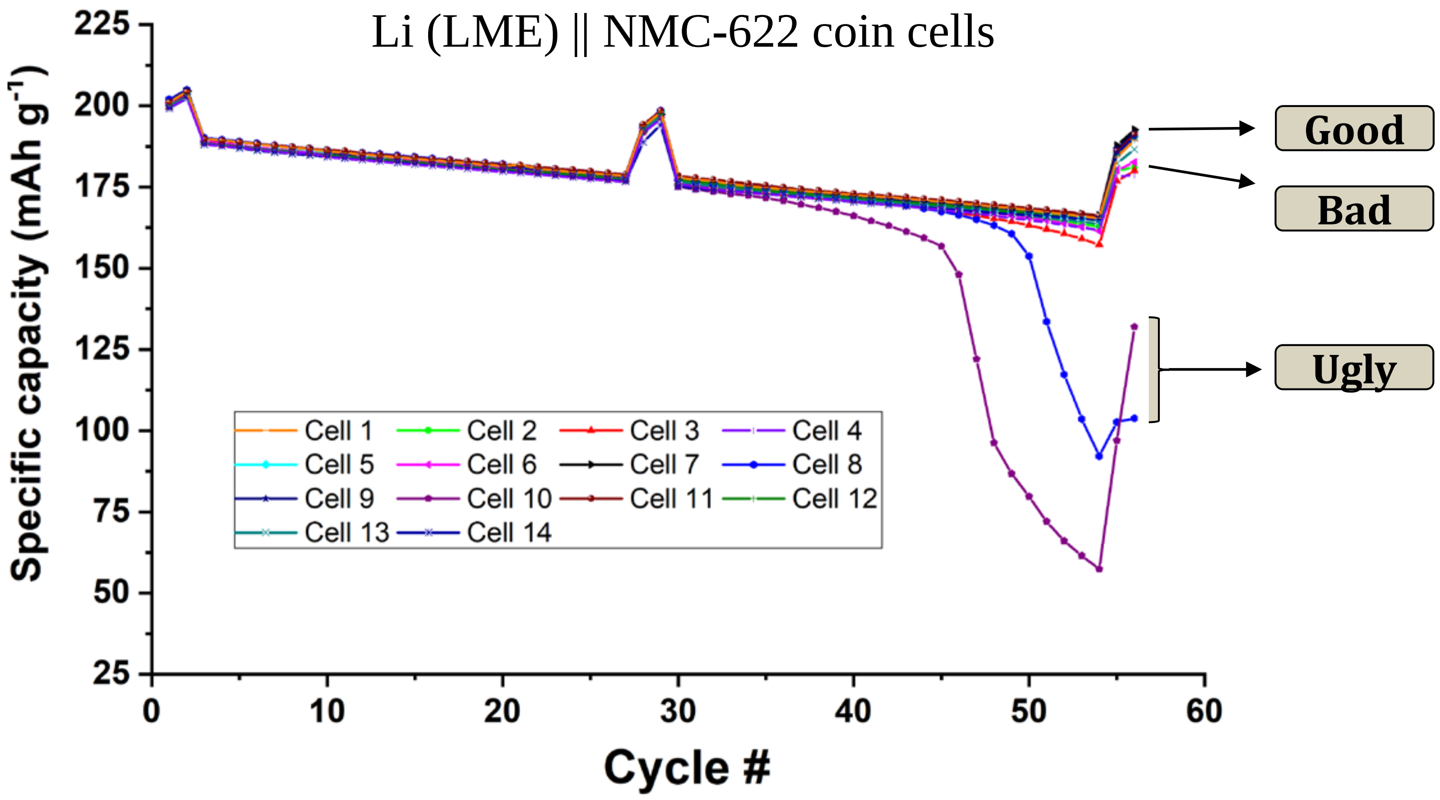
Atomistic



Real-life-scale

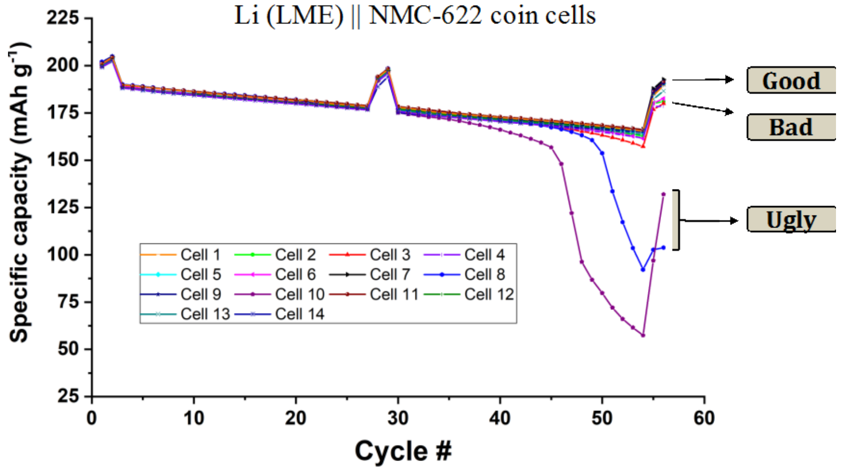


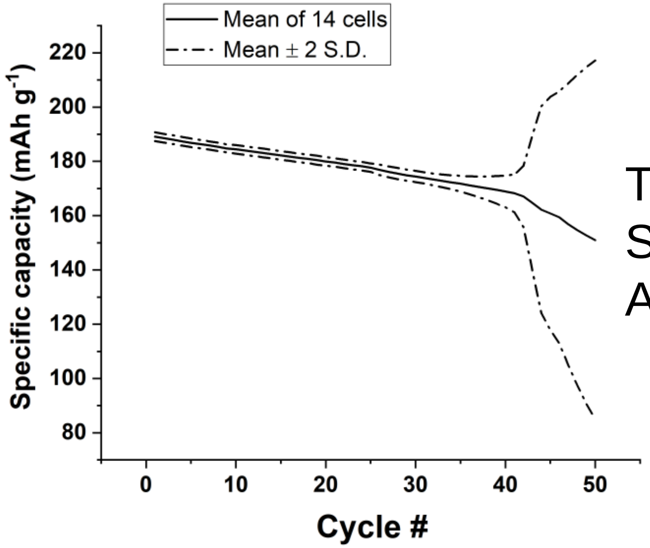
Cycle Life in Li || NMC Cells



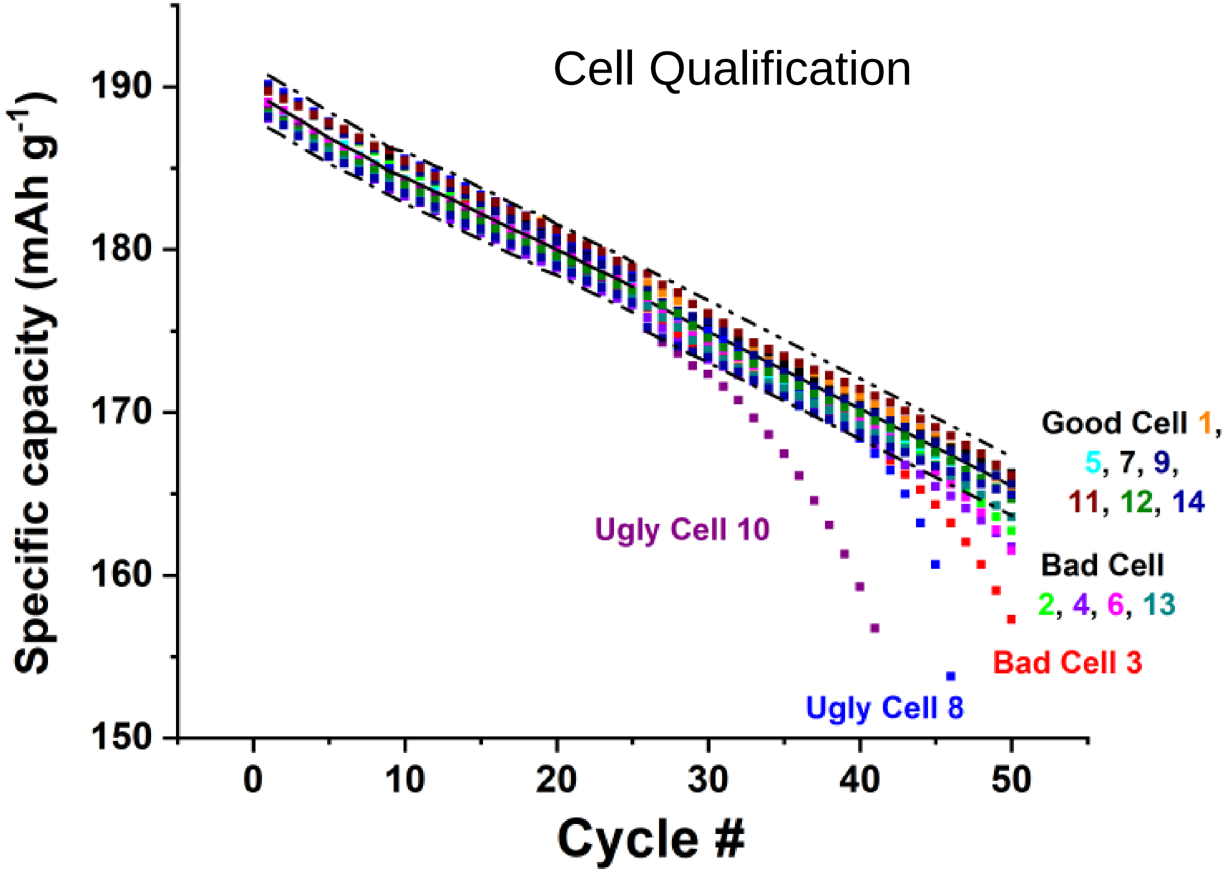
- **Consistency in 14 cells offers an excellent basis for eCAD**
- **Complete life cycle revealed from Good to Bad and Ugly**
- **Full analysis on failure mechanism to identify every single attribute to capacity fade**
- **Quantitative results for all capacity fade attributes**
- **Uncover and quantify Loss of Li Inventory (LLI) for charge and discharge regime, respectively, which does not appear in charge retention measurements**
- **Life prediction for individual cell**

Cell Qualification vs. Statistical Analysis



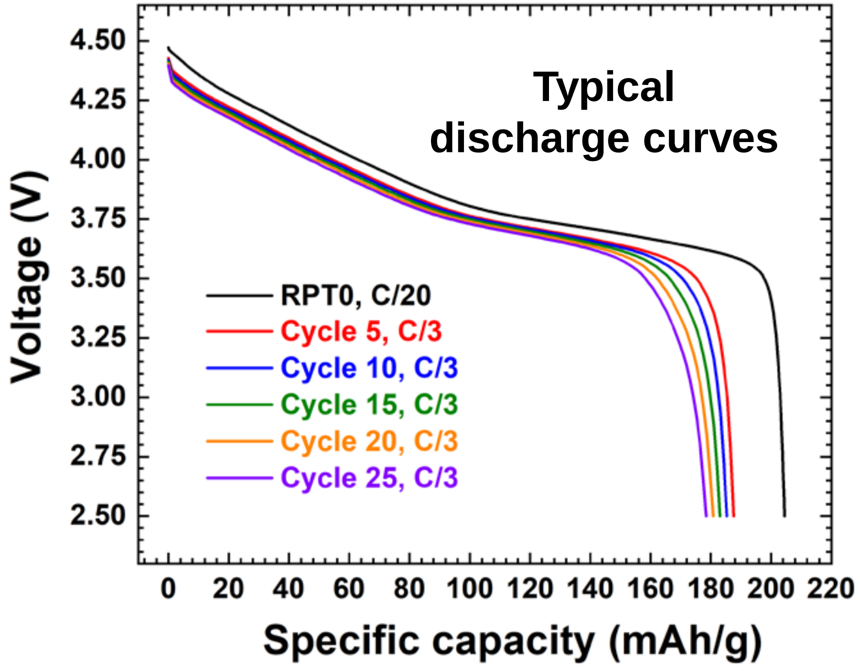


Traditional Statistical Analysis

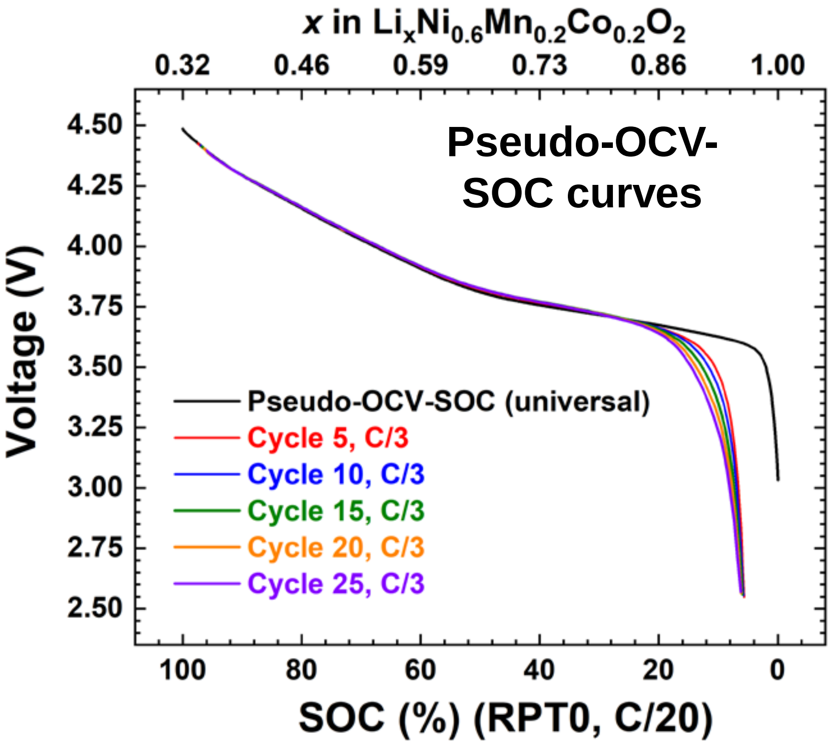


State of Charge (SOC)-based Performance Analysis

- **Remove bias from experimental conditions to reveal true SOC correspondence — Separate thermodynamic and kinetic attributes**

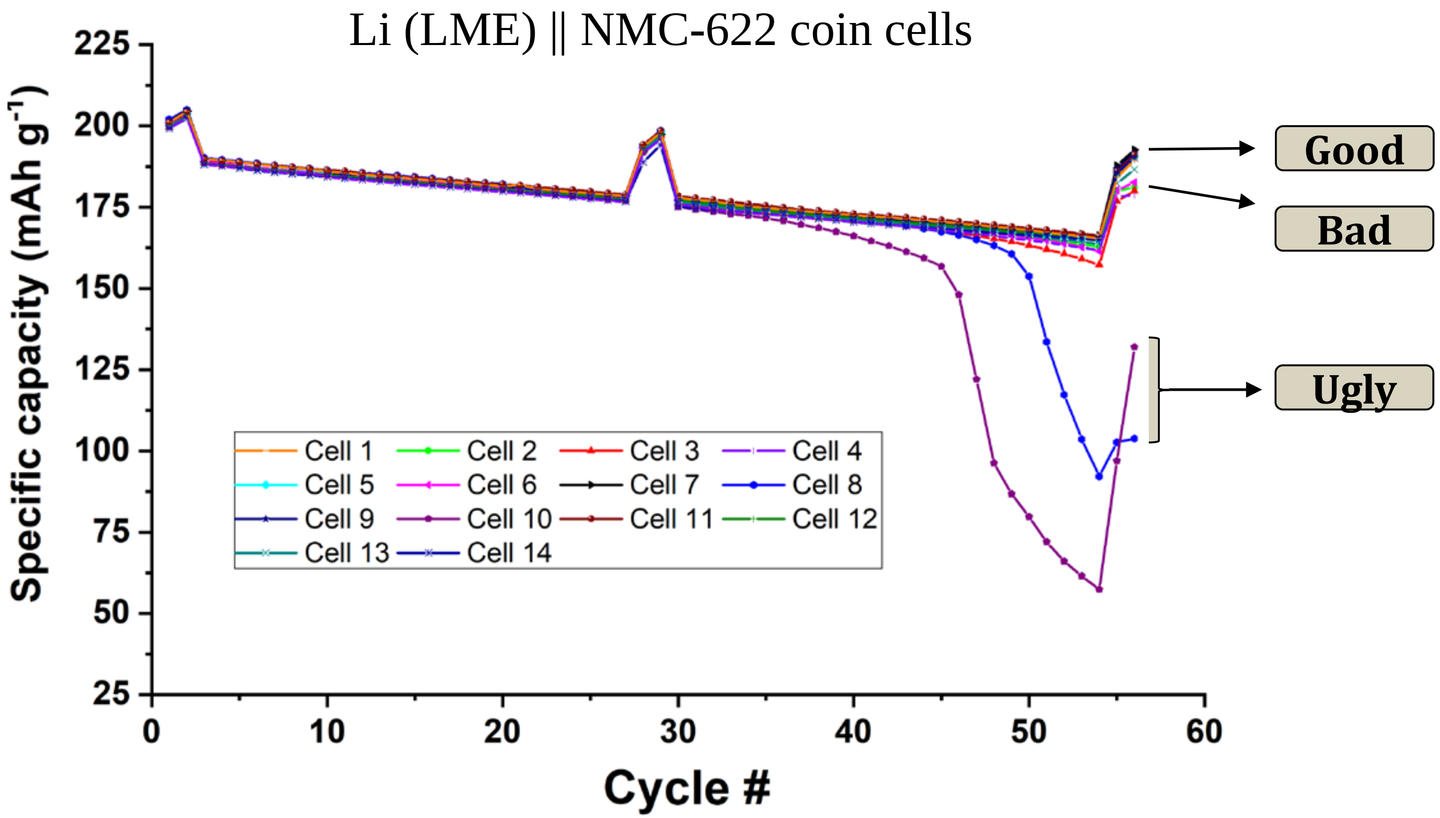


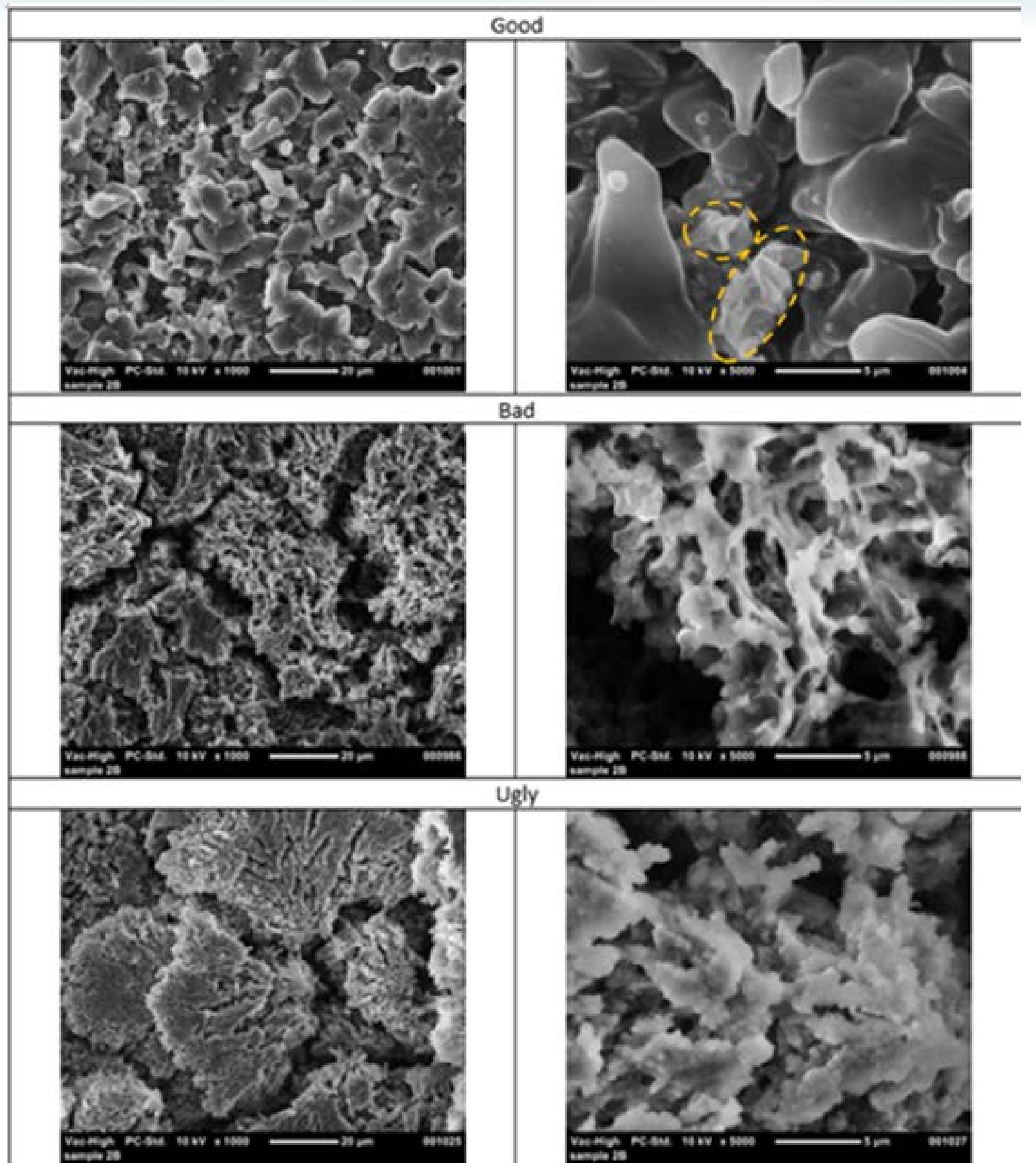
Experimental Conditions



State of the System

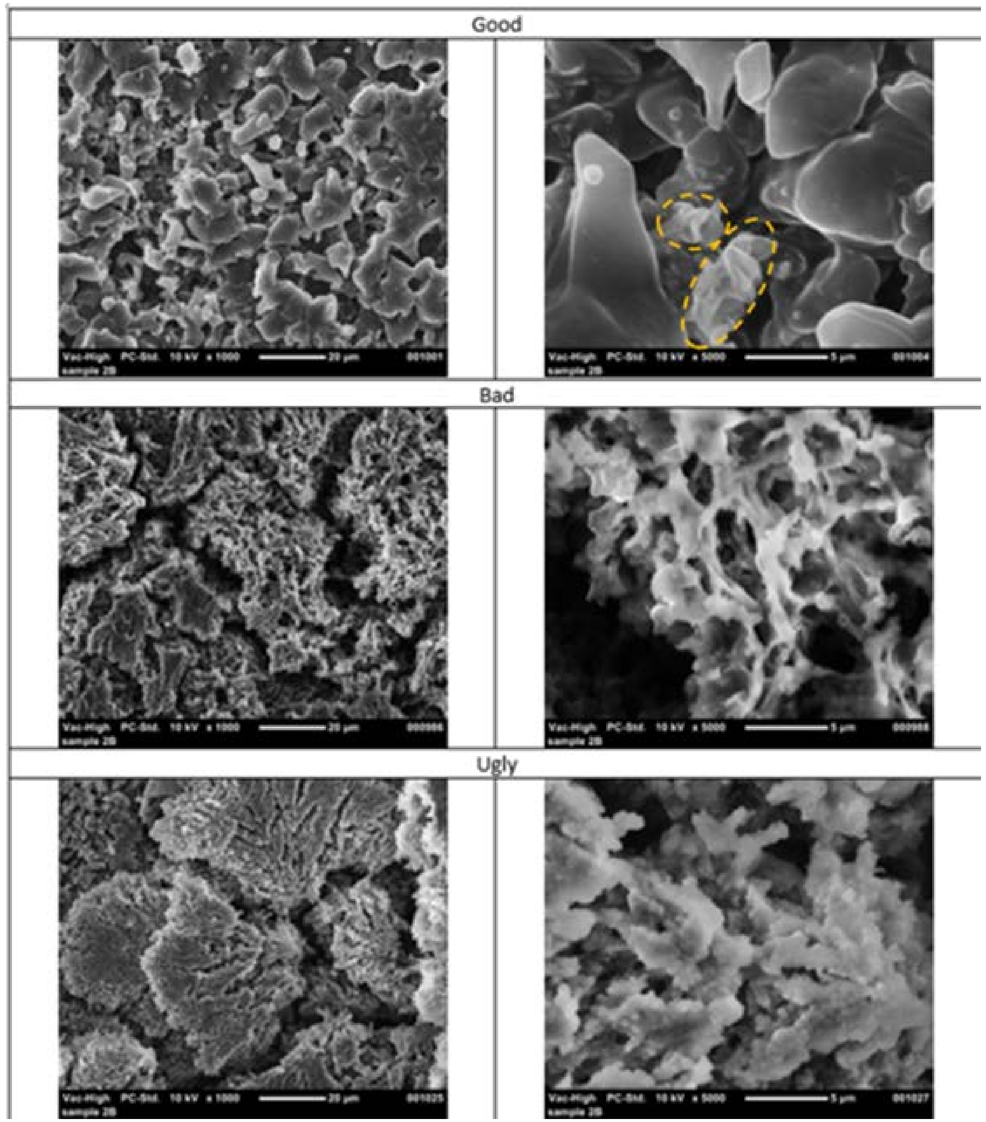
Cycle Life in Li || NMC Cells

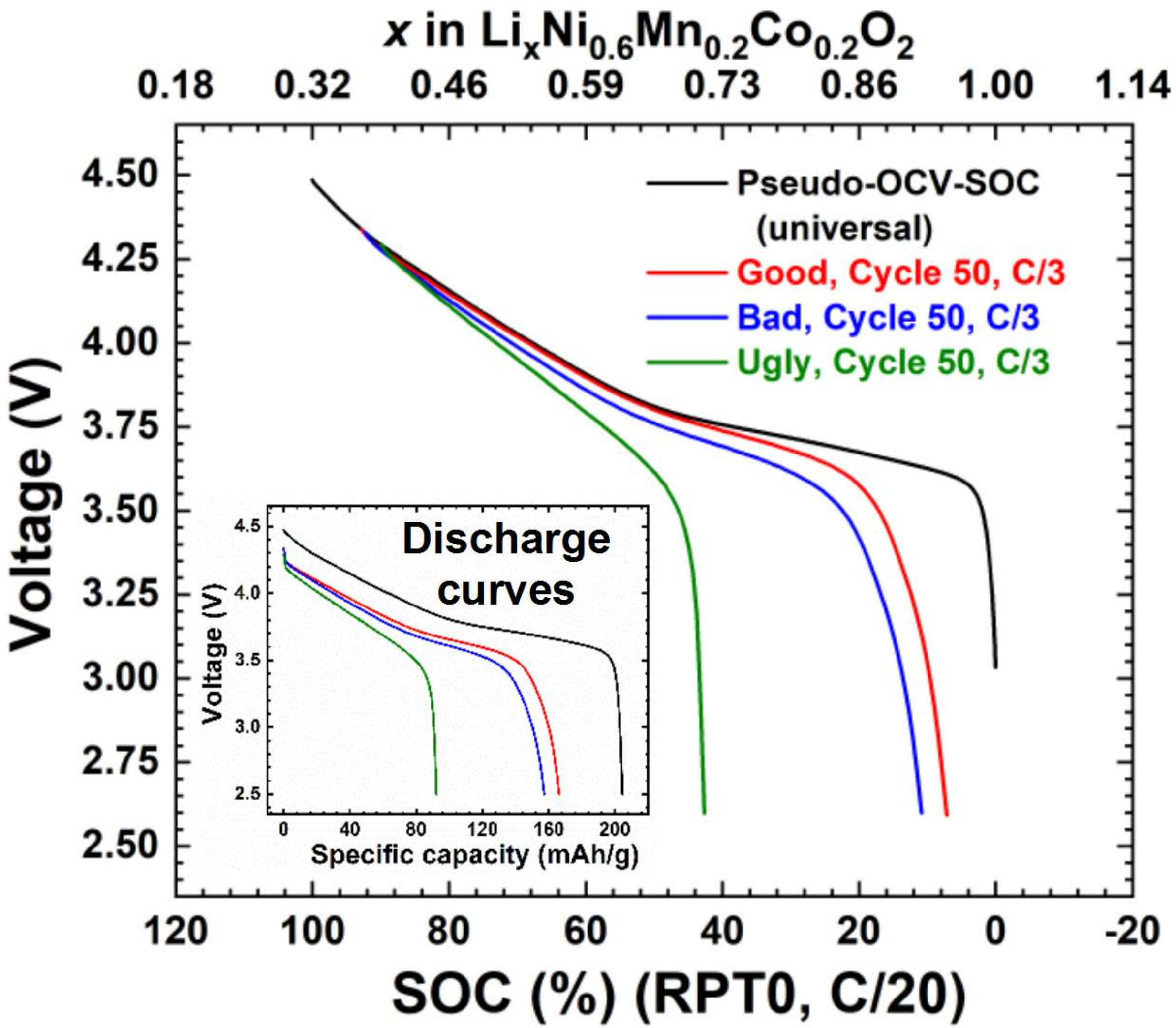




Cycle life sensitively depends on Li inventory

Relate Failure Modes to Li Anode Morphology





Conclusion

- Li metal electrode comes a long way to practical applications
- Multi-scale understanding of the kinetics by experiments and model simulations
- Unlock the mystery of Li stripping and deposition with better control of kinetics

Acknowledgements

- Office of Vehicle Technologies of the U.S. Department of Energy in the Advanced Battery Materials Research (BMR) Program (Battery500 Consortium) — David Howell, Tien Duong
- PNNL — Jun Liu, Jason Zhang, Jie Xiao, Wu Xu, Xiaodi Ren
- UC Irvine — Materials Research Institute (IMRI) for the use of Cryo-Electron Microscopy Facility and Kratos XPS

To Bob Huggins

- Happy the 90th B-day !
- ***Thank you for great mentoring and guidance !!***