3D Micro-Structure Resolved Simulations of Thick Li-Ion Batteries

Timo Danner[1,2], Tianyi Wen[1], Arnulf Latz[1,2,3], Madhav Singh[4], Jörg Kaiser[5], Horst Hahn[2,4]

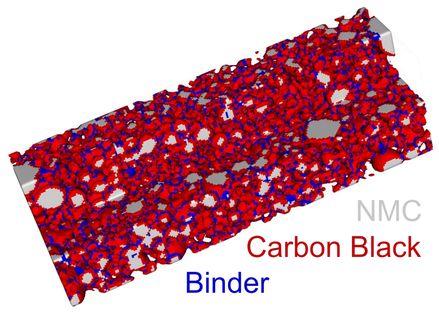
Helmholtz Institute Ulm
Electrochemical Energy Storage

[1] Institute of Engineering Thermodynamics, German Aerospace Center (DLR), Stuttgart, Germany, [2] Helmholtz Institute Ulm for Electrochemical Energy Storage (HIU) and [3] Institute of Electrochemistry, University of Ulm, Ulm, Germany, [4] Institute of Nanotechnology and [5] Project Competence-E, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

Motivation

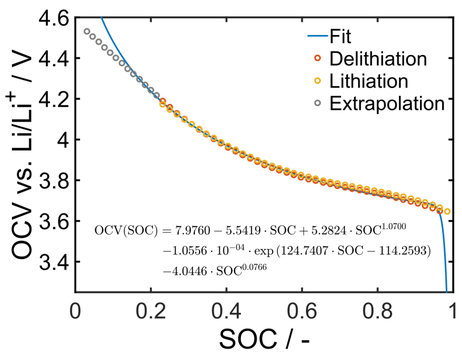
Advantages:
- Reduction of inactive materials & production time
  - ➔ Lower cost & improved energy density

Challenges:
- Long transport pathways
  - ➔ Low rate capability and degradation



Experimental

- NMC and Graphite electrodes
  - ➔ Thickness of 70 and 320 µm
- Half-cell measurements (Area ~ 1cm²)
  - ➔ De-/lithiation at C/10, C/5, C/2
  - ➔ OCV data (current pulses)
- Battery cells (Area ~ 25 cm²)
  - ➔ Charge/discharge curves
- ➔ Details can be found in: [1]

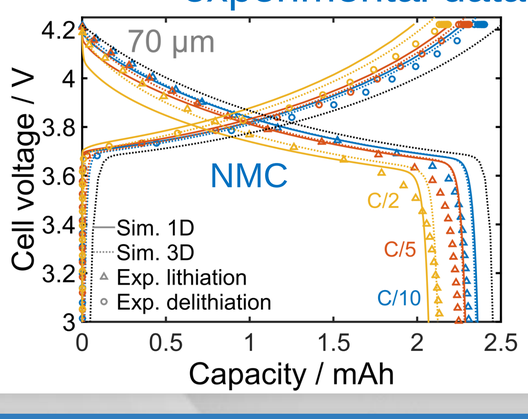


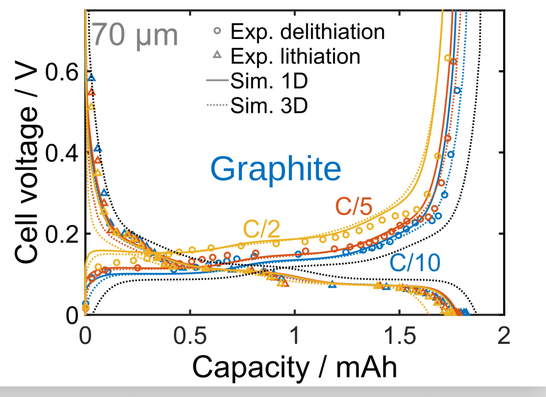
Model Parameterization & Validation

- Micro-structure resolved simulation [2]
  - ➔ Finite-Volume Code based on CoRheoS framework of Fraunhofer ITWM

BEST
Battery and Electrochemistry Simulation Tool

- Electrode structure based on tomography data [3] and SEM images
- Parameter estimation via 1+1D model [4]
  - ➔ Fit of exchange current density to half-cell data (70 µm electrodes)
    - ➔ Excellent agreement of 3D simulations with experimental data





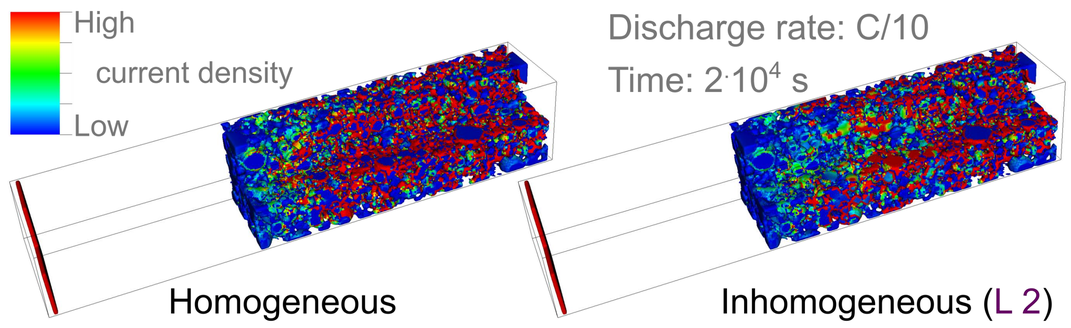
Simulation of Li-Ion Batteries

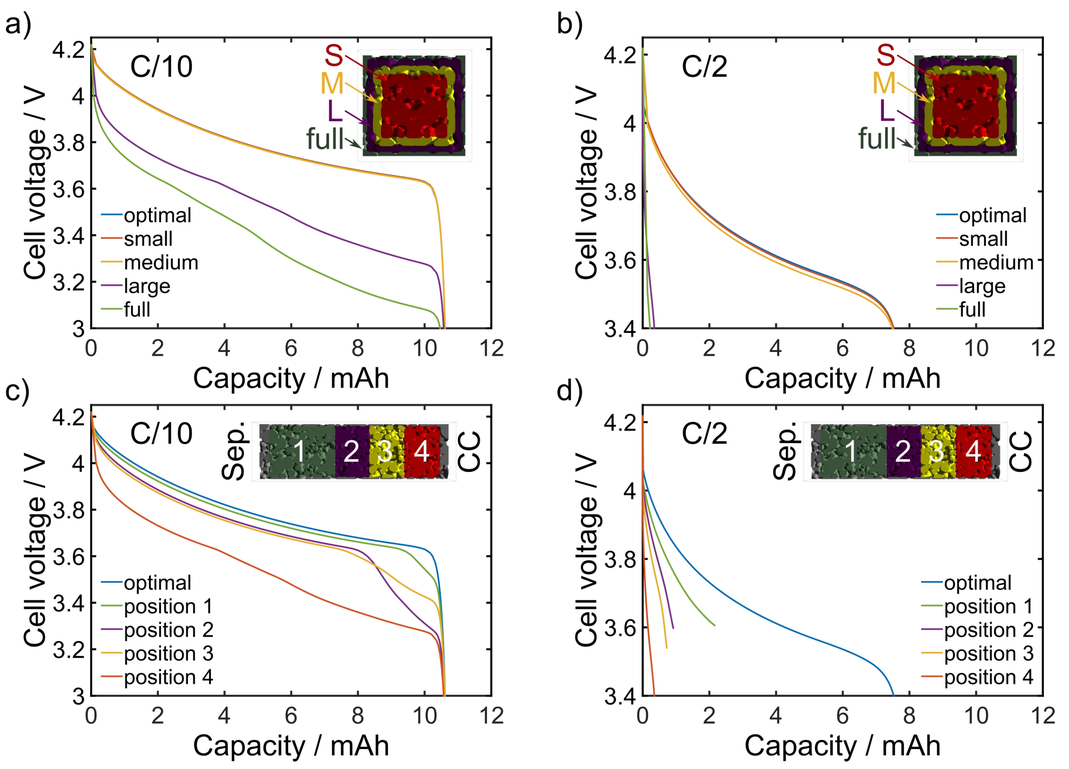
70 µm electrodes:
- Agreement between 1D and 3D
- Capacity: Experiment < 3D < 1D
  - 3D: disconnected particles
  - Experiment: Fluctuations in electrode loading?

320 µm electrodes:
- Deviation Exp. ↔ Sim. at C/2
  - ➔ Carbon black distribution?
  - ➔ Li plating?
  - ➔ Anode structure?

Carbon Black Distribution

- Variation of size and position of inhomogeneity in the distribution of conductive additive
  - ➔ Influence only for large unconnected areas



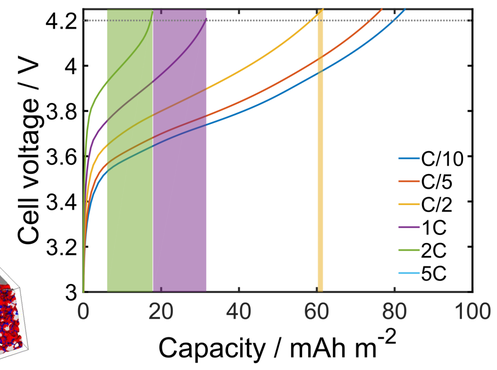


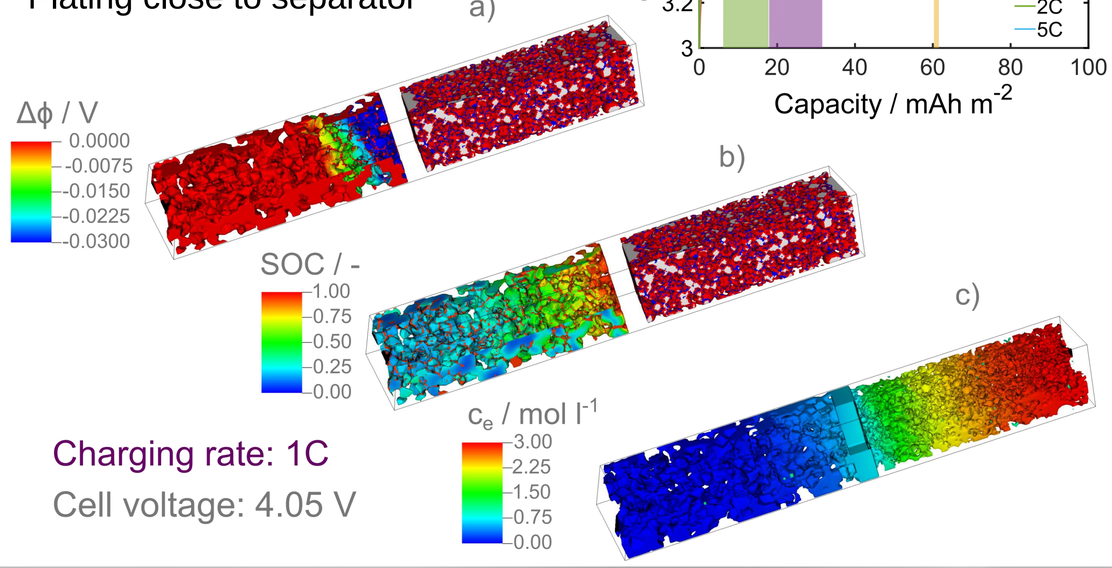
Lithium Plating

- Condition for Li plating

$$\eta = \phi_{elode} - \phi_{elyte} = \Delta\phi < 0$$

- Plating possible if charge rate > C/2
  - ➔ Lower currents: cut off voltage
- Plating close to separator





Summary

- Micro-structure resolved simulations of thick Li-Ion batteries
- Parameters from literature and dedicated experiments
  - ➔ 1+1D model for efficient parameter estimation
- Deviation between simulations and experimental data at high C-rate
  - ➔ Investigation of carbon black distribution and Li plating

[1] M. Singh *et al.*, *J. Electrochem. Soc.*, 162(7): A1196–A1201, 2015.
[2] A. Latz *et al.*, *Beilstein J. Nanotechnol.*, 6:987–1007, 2015.
[3] M. Ebner *et al.*, *Adv. Energy Mater.*, 3(7):845–850, 2013.
[4] M. Doyle *et al.*, *J. Electrochem. Soc.*, 140(6):1526–1533, 1993.

KIT
ulm university universität ulm
ZSW
DLR Deutsches Zentrum für Luft- und Raumfahrt German Aerospace Center
bw HPC – C5