Virtual electrode design of thick Li-Ion batteries

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Motivation

Design of Li-Ion battery electrodes with high areal capacity

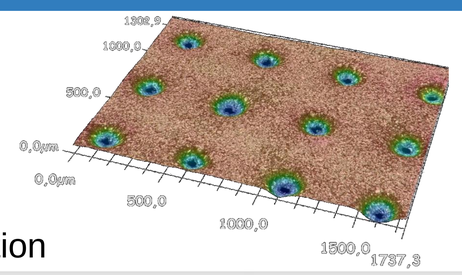
Advantages:
- Reduction of inactive materials & production time
  - ➔ Cost & theoretical capacity

Challenges:
- Long transport pathways
  - ➔ Low rate capability
- Challenging production process

Approach

Laser perforation of electrode layers
- Improved electrolyte transport
- Homogeneous cell filling
- But: Active material loss

➔ Design space for electrode optimization

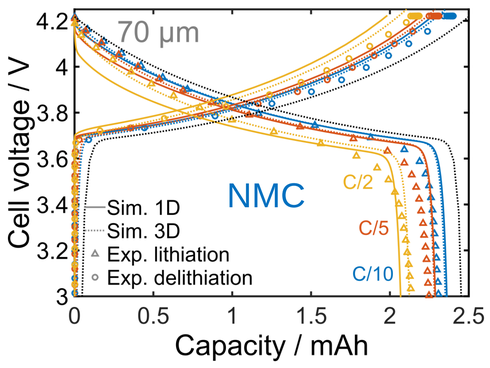


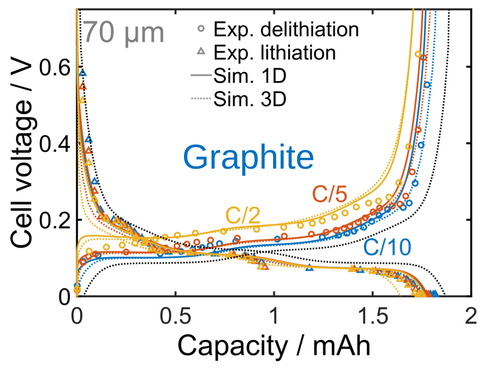
Model Parameterization & Validation

- NMC and Graphite electrodes [1,2]: 70 and 320 µm
- Half-cell (Area ~ 1cm²) & full-cell measurements (Area ~ 25 cm²)
  - ➔ Charge/discharge curves at C/10, C/5, C/2
- Micro-structure resolved simulation [3]
  - ➔ Finite-Volume Code based on CoRheoS framework of Fraunhofer ITWM

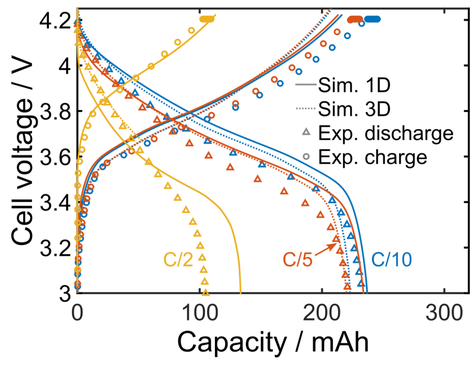
BEST
Battery and Electrochemistry Simulation Tool

- Half-cell simulations: Parameterization (thin 70 µm electrodes)





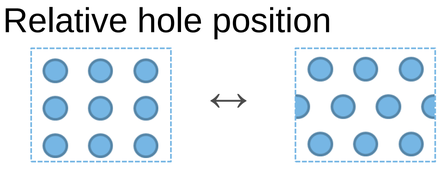
- Full-cell simulations: Validation on thick cells with 320 µm electrodes

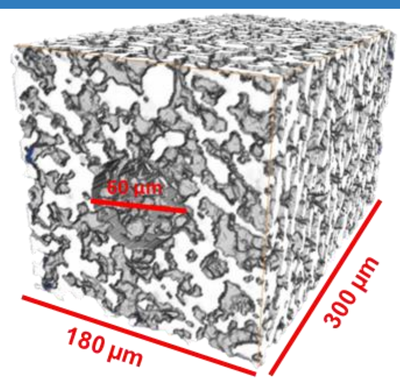


Good agreement with experiments
- Deviation Exp. ↔ Sim. at C/2
  - ➔ Carbon black or Li plating?
- Transport limitations in electrolyte during C/2 discharge
  - ➔Depletion of Li ions in cathode

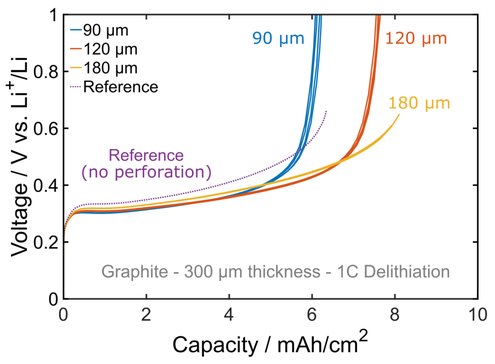
Electrode design studies

- Design space for laser perforation:
  - Electrode thickness (300 µm)
  - Hole diameter (60 µm)
  - Hole distance (90 / 120 / 180 µm)
  - Relative hole position





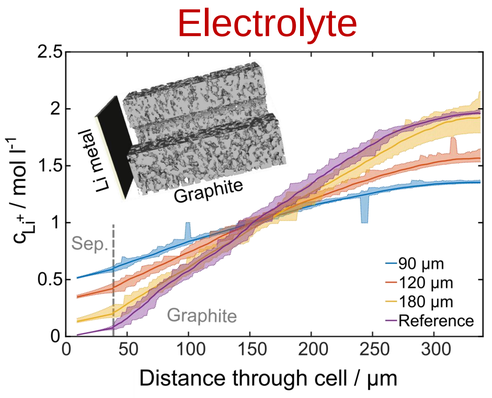
➔ Electrode geometry via stochastic 3D structure generator [4]



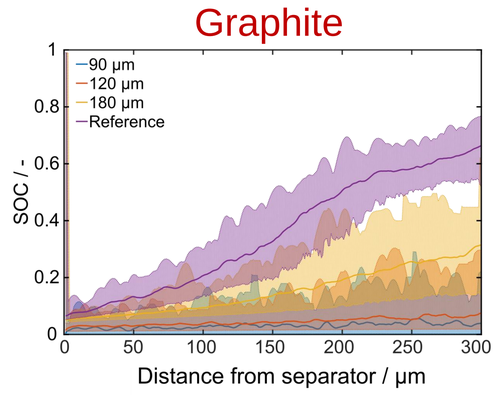
- Electrochemical simulations:
  - Effect of hole distance on half-cell performance:
  - ➔ Small: Active material loss
  - ➔ Medium: Capacity gain
  - ➔ Large: Transport limitations

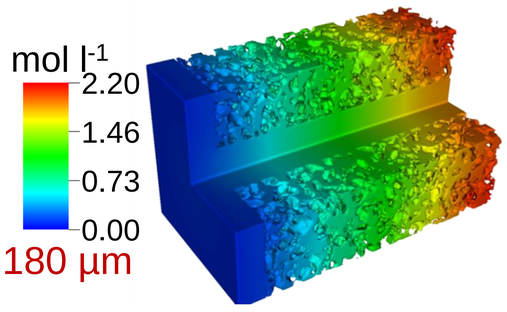
- Concentration distributions:
  - Improved Li^+ transport in electrolyte ➔ reduced gradient in c_{Li}
  - Delithiation of graphite close to CC ➔ higher graphite utilization
  - Larger variation in Li concentration ➔ inhomogeneity at edges

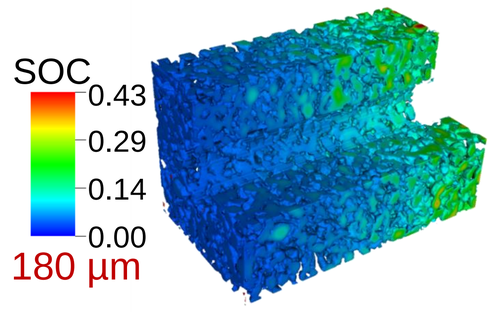
Electrolyte



Graphite

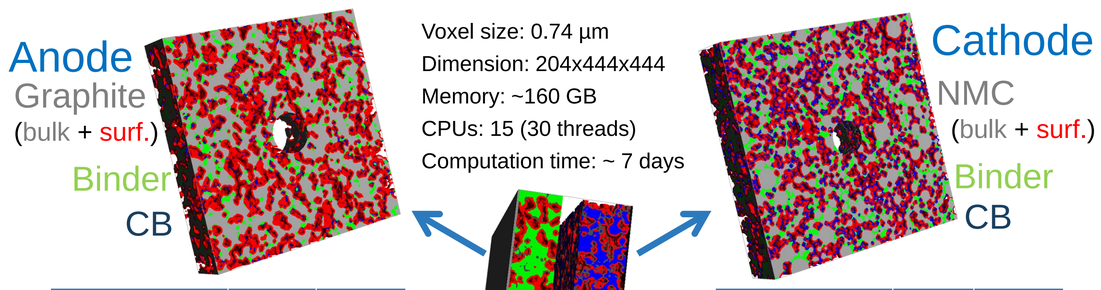






➔ Significant optimization potential by laser perforation

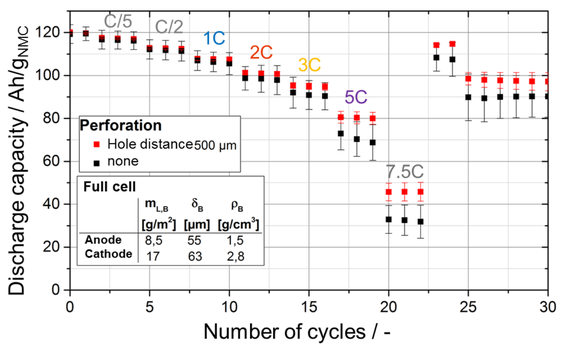
Proof of concept: 'Thin' electrodes



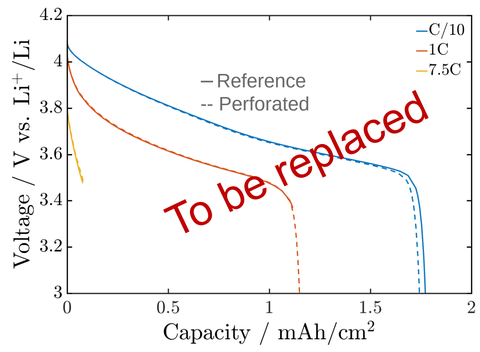
Anode	wt.-%	vol.-%
Binder	4	3.4
Carbon black	3	2.3
Graphite	93	61.7
Pore space	0	32.5

Cathode	wt.-%	vol.-%
Binder	4	6.3
Carbon black	6	8.9
NMC 111	90	53.1
Pore space	0	31.8

- Experiments:



- Simulations:



➔ Improved performance at high C-rates

Summary

- Micro-structure resolved simulations of thick Li-Ion batteries [3]
- Parameters from literature [5] and dedicated experiments [2]
  - ➔ Good agreement with half- and full-cell experiments [1]
- Transport limitations of Li in electrolyte at high C-rate
  - ➔ Depletion zones of Li and strong concentration gradients
- Positive effect of laser perforation on Li transport in electrolyte
  - ➔ Improved rate performance & higher capacity

[1] M. Singh *et al.*, *J. Electrochem. Soc.*, 162(7): A1196–A1201, 2015.
[2] T. Danner *et al. J Power Sources*, *334*, 191–201, 2016.
[3] A. Latz *et al.*, *Beilstein J. Nanotechnol.*, 6:987–1007, 2015.
[4] D. Westhoff *et al. Comput Mater Sci*, 126, 453–467, 2017.
[5] M. Ebner *et al.*, *Adv. Energy Mater.*, 3(7):845–850, 2013.

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