Understanding Electrolyte Filling of Lithium-Ion Battery Electrodes on the Pore Scale

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Motivation and Aim

- Optimization of battery production in general and filling process in particular
- Reduction of process time and costs
- Improvement of battery performance

Aim:

- Structurally resolving the electrolyte distribution in realistic battery microstructures
- Understanding wetting phenomena at the pore-scale
- Enhancing simulation efficiency by developing a multi-scale approach using lattice Boltzmann method (LBM) and pore network models (PNM)

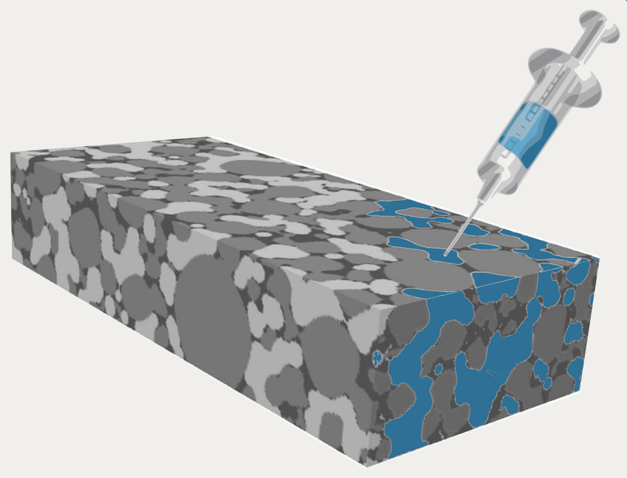

Fig. 1: Electrolyte filling of realistic battery microstructure

Fundamentals of LBM

- Fluid flow solver based on the Boltzmann equation

$$\frac{\mathrm{d}f}{\mathrm{d}t} = \underbrace{\frac{\partial f}{\partial t} + \xi\frac{\partial f}{\partial x}}_{(1)} + \underbrace{\frac{F}{\rho}\frac{\partial f}{\partial \xi}}_{(2)} = \underbrace{\Omega(f)}_{(3)}$$

(1) Advection
(2) Force field
(3) Collision

- *Advantages*:
  - Mesoscopic method for reliable insight into pore-scale phenomena
  - Simulation of multi-physics issues in complex geometries

Fundamentals of PNM

- Reconstruction of realistic porous media into pores and throats with idealized geometrical shapes
- Solver for transport processes and simple equations in reconstructed pore space on a pore-to-pore basis

$$P_c = C_{geo} \cdot \frac{2\sigma\cos(\theta)}{r}$$

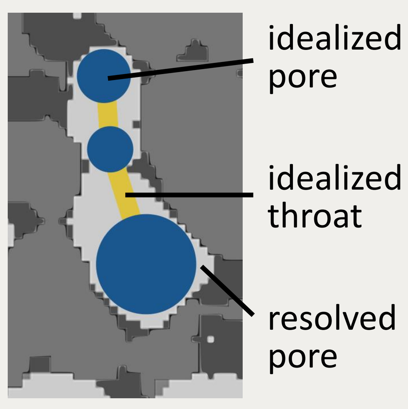

Fig. 2: Reconstruction of resolved pore space

- *Advantages*:
  - Efficient simulations by complexity reduction, large degree of locality, and computational parallelization
  - Upscaling to large-scale structures

Simulation Overview

- Realistic stochastically generated 3D NMC microstructures with and without binder in µm resolution
- Modelling of electrolyte-air flow and wetting behavior
- Modelling of sub-resolution pore effects
- Pressure-driven flow
- *Influencing factors*: wettability, structural effects, and binder content
- *Analysis*: pressure-saturation behavior, air entrapment, and battery performance

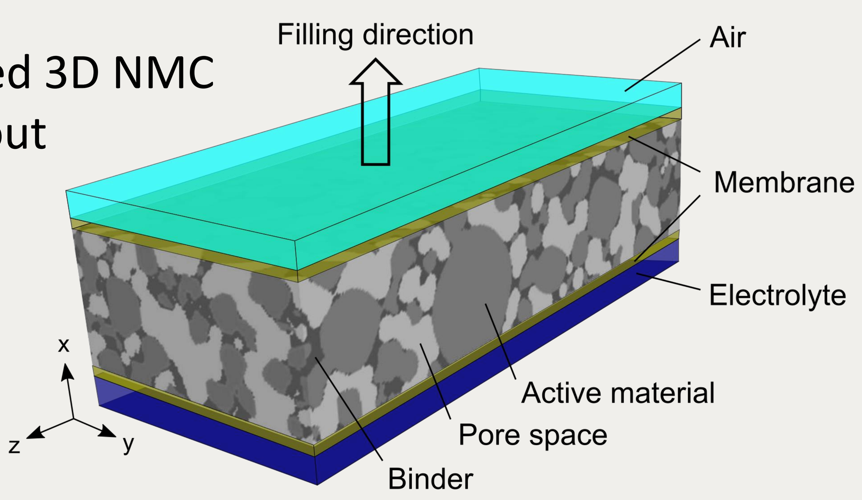

Fig. 3: Schematic setup for electrolyte filling of porous electrodes under a pressure gradient.

Results

Pressure-Saturation Behavior

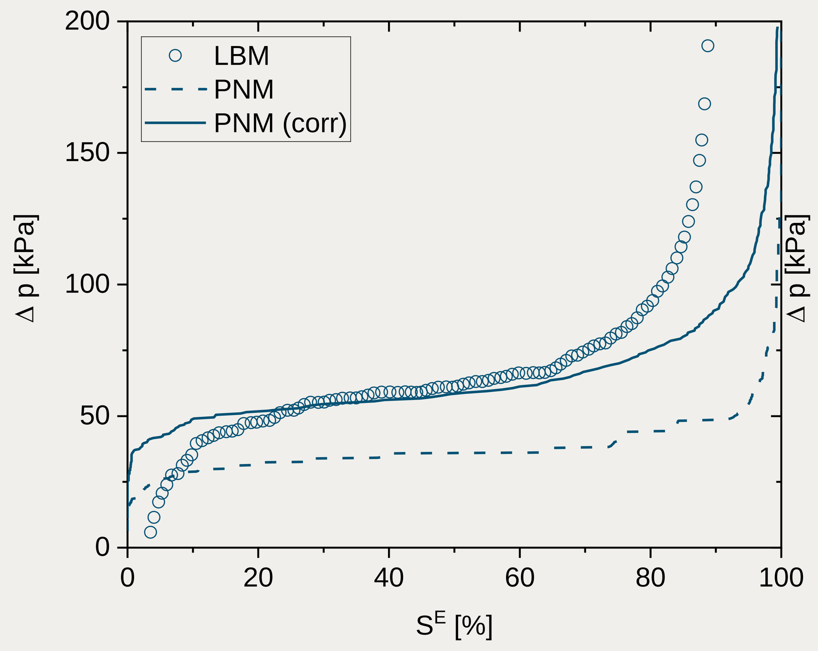

Fig. 4: Comparison of LBM (circles), basic PNM (dashed line), and PNM with geometrical correction (solid line).

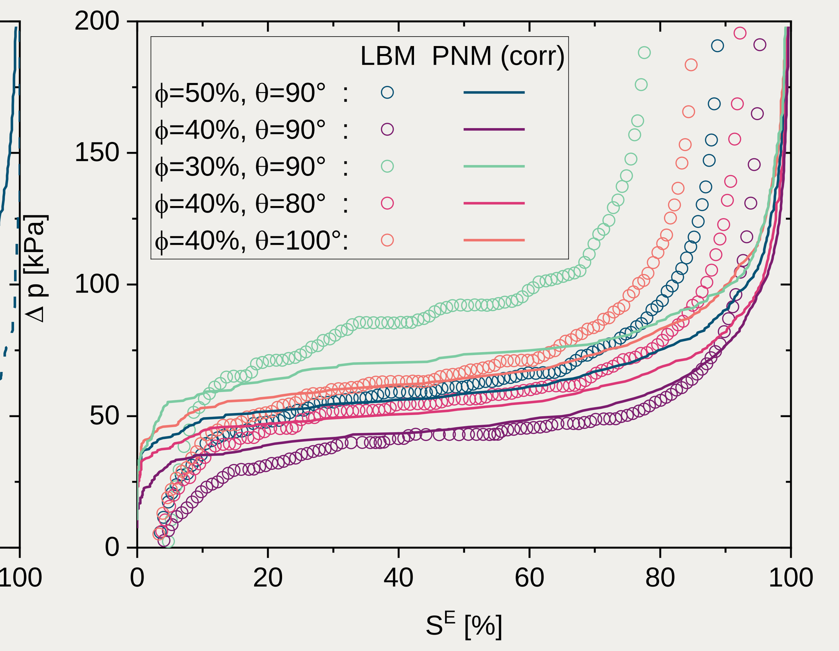

Fig. 5: Influence of porosity ϕ and wettability θ using LBM (circles) and PNM with geometrical correction (solid lines).

→ PNM with geometrical correction is able to predict characteristic pressures. However, it deviates in predicting the final saturation.

Air Entrapment

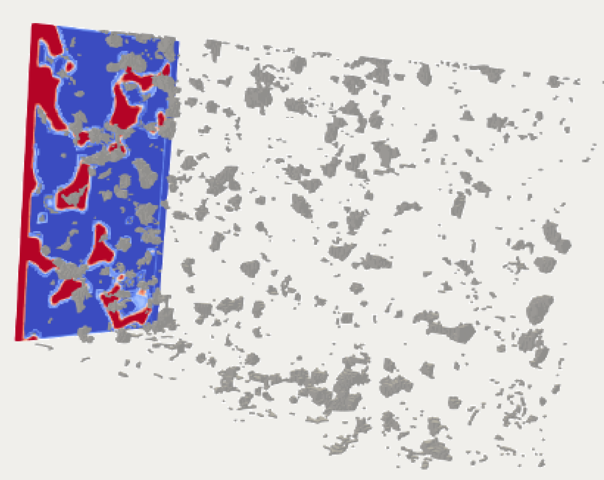

Fig. 6: LBM simulation of electrolyte distribution in porous electrodes. Entrapped air bubbles are depicted in grey.

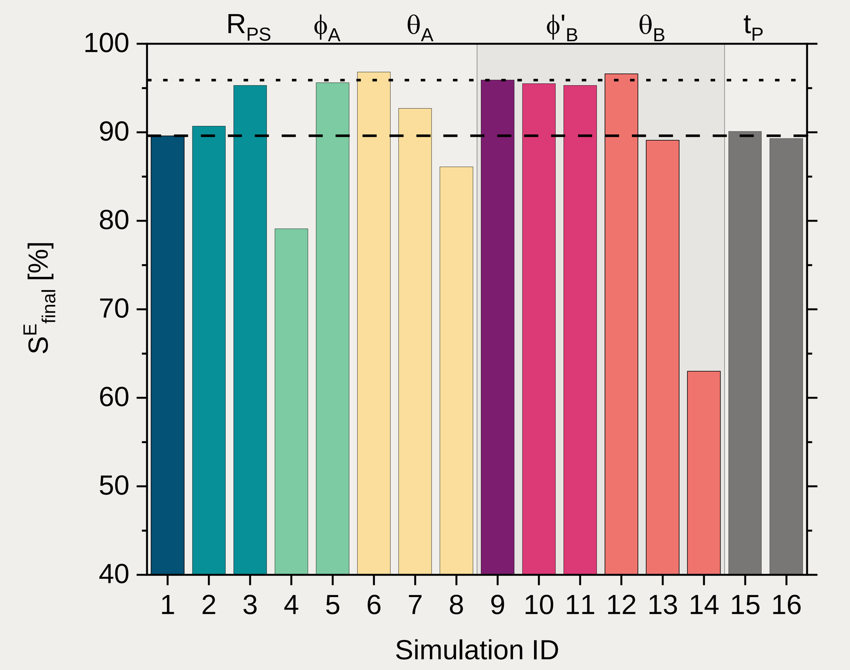

Fig. 7: Influence of particle size R, porosity ϕ, wettability θ, and process time t_p on the final electrolyte saturation. Deviations from $S^E_{final} = 1$ are due to air entrapment.

→ LBM predicts the location and distribution of entrapped air, which is influenced by the wettability and pore size mainly.

Battery Performance

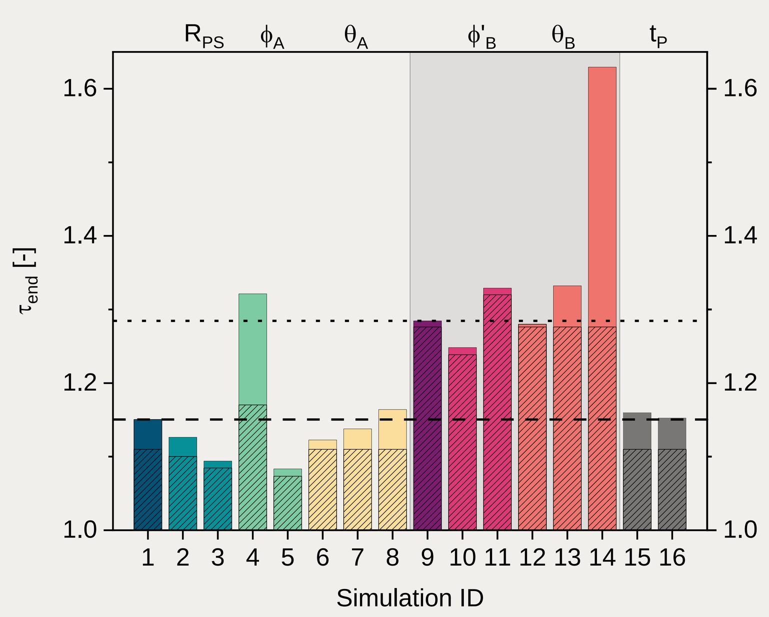

Fig. 8: Minimum tortuosity (shaded bars) vs. actual tortuosity (colored bars). Deviations are due to air bubbles blocking the pores.

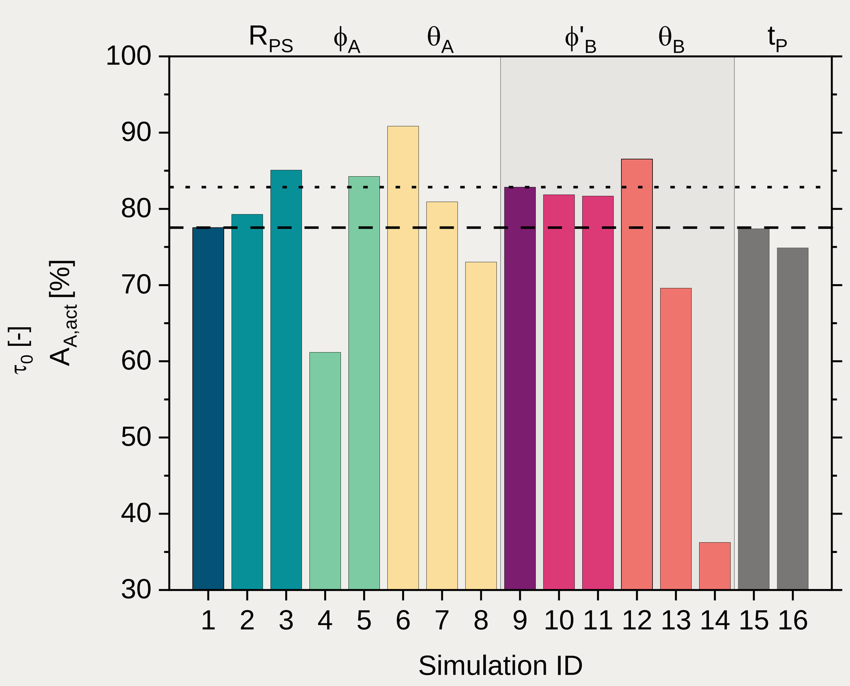

Fig. 9: Electrochemically active electrode area after filling. Deviations from $A_{A,act} = 1$ are due air bubbles passivating the surface.

Summary

- Application of LBM and PNM to successfully study filling processes within complex microstructures at high structural resolution
- PNM with geometrical correction is an efficient alternative to LBM for the prediction of pressure behavior
- LBM allows detailed insight into pore-scale phenomena and is useful for the optimization of structural, physical and process parameters

DEFACTO

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