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Pore-scale Resolved 3D Simulations of Aqueous Organic Flow Batteries

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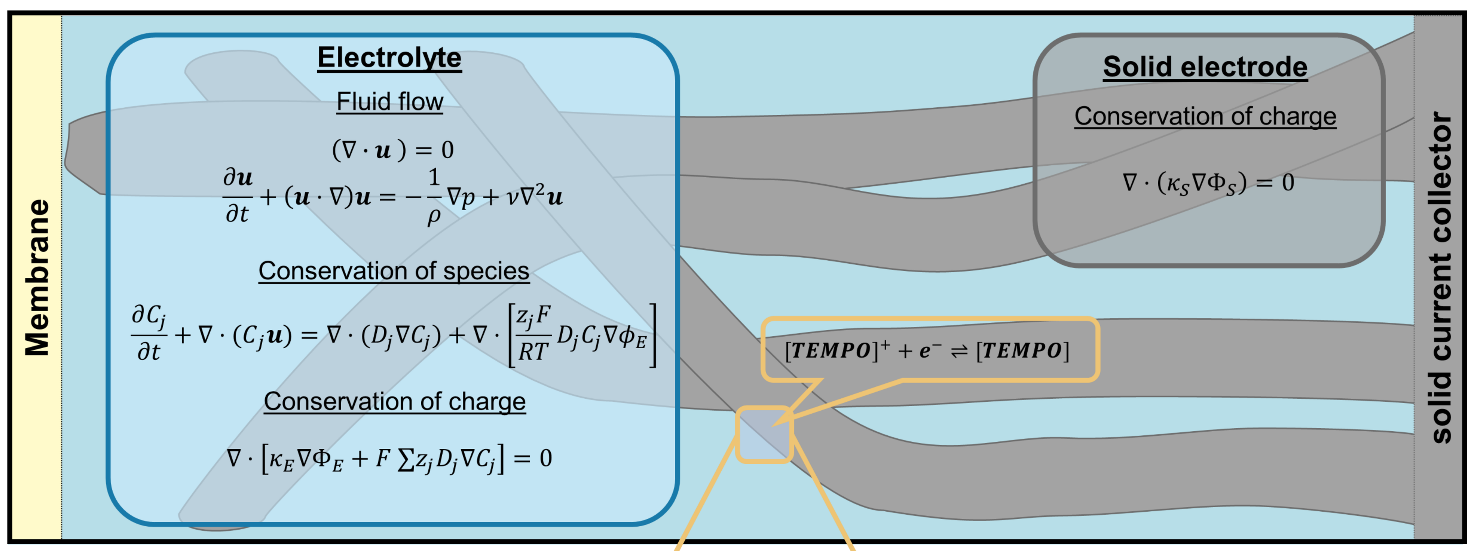
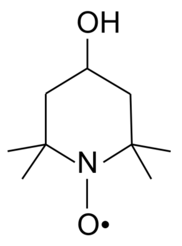
Pore-scale resolved half-cell Model [1] parameterized with 4-OH-TEMPO





Kinetic boundary condition: Butler-Volmer-Type

$$-\boldsymbol{N}_{T^+} = +\boldsymbol{N}_T = k \cdot (C_T^s)^{\alpha} \cdot (C_{T^+}^s)^{(1-\alpha)}\left[exp\left(\frac{(1-\alpha)F\eta}{RT}\right) - exp\left(-\frac{\alpha F\eta}{RT}\right)\right]$$

$$\eta = \Phi_S - \Phi_E - E^0 - \frac{RT}{F}\ln\left(\frac{C_{T^+}^s}{C_T^s}\right)$$

$$\boldsymbol{N} = \frac{\boldsymbol{J}}{F}$$

Motivation

- Examination of promising, non-rare and non-toxic organic material candidates on the pore-scale for flow battery performance
- Identification and visualization of cell limitations such as structure-dependent transport limitations

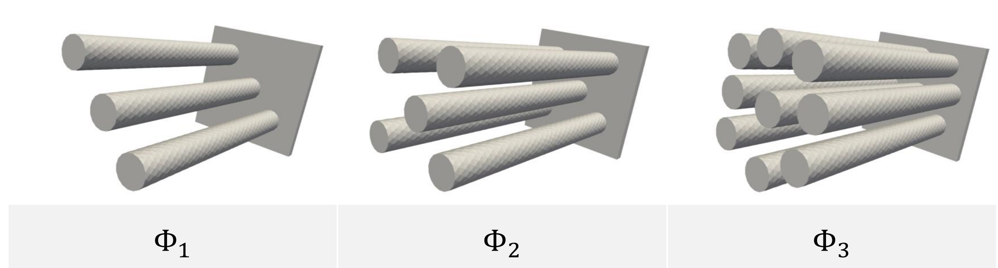
Research Objective

- 3D numerical simulation of coupled electrolyte flow and electrochemical processes
- Deduction of qualitative and quantitative recommendations for the optimization of flow batteries using novel active materials

Conclusion

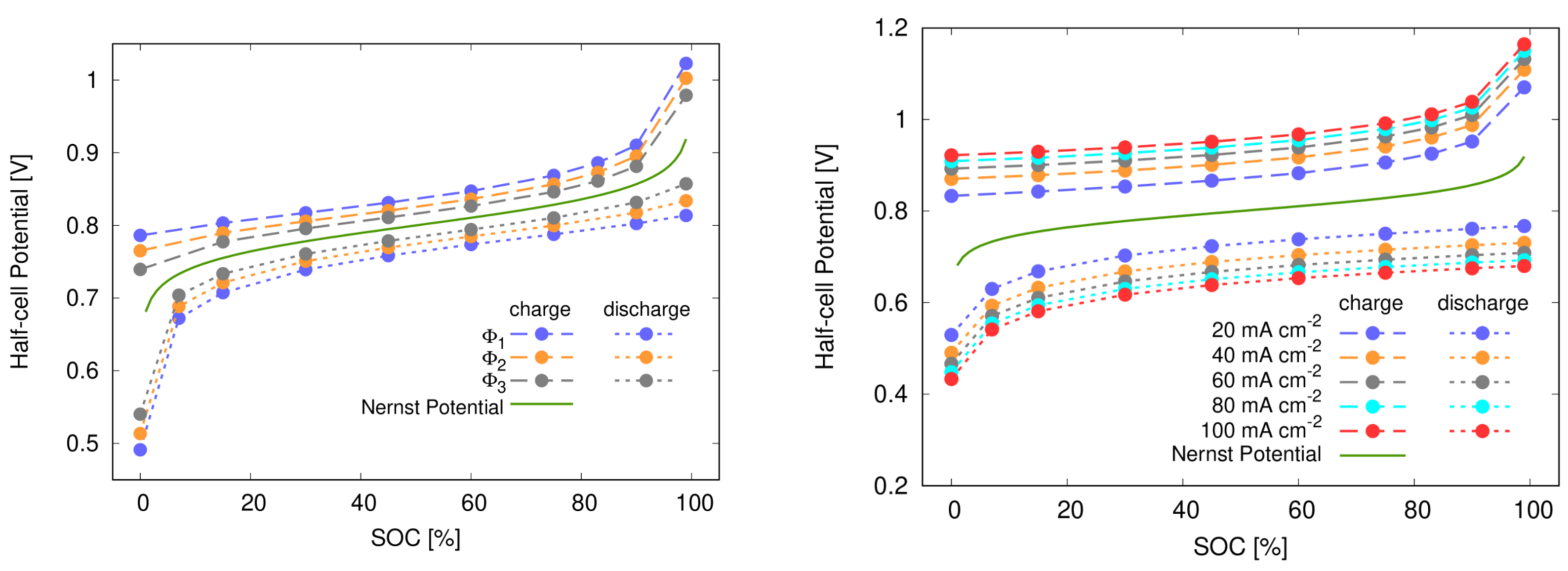
- Reproduction of concentration and active surface dependent flow battery characteristics
- SOC dependent optimal Reynolds Number around ~10^{-2} regarding mass transport and half-cell potential

Results – Structured electrodes [1]



	Φ_1	Φ_2	Φ_3
Domain size	100 µm x 50 µm x 50 µm		
Fiber diameter	10 µm		
Porosity Φ	0.92	0.86	0.75
Specific active surface area	43 761 m² m⁻³	66 269 m² m⁻³	111 283 m² m⁻³
Number of cells	114 077	138 898	168 889

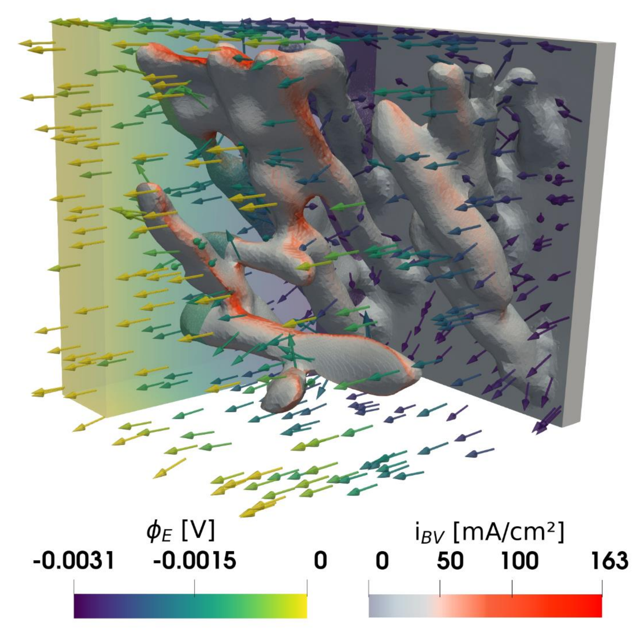
- Increasing active surface area leads to reduced potential differences due to reduced activation overpotential
- Increasing current density leads to higher potential differences due to higher ohmic losses



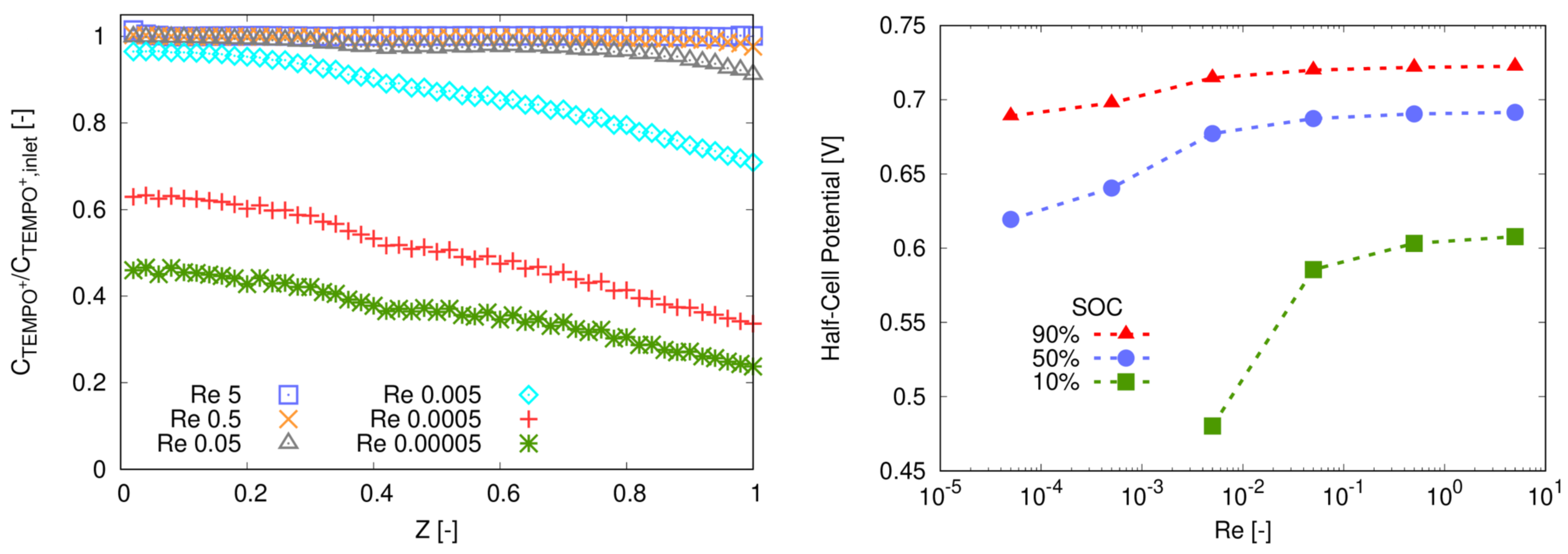
Half-cell potential over state of charge (SOC) for different porosities at constant galvanostatic current density of 40 mA cm⁻² (left) and for different charge/discharge rates (right). Concentration is 0.5M TEMPO in 1.5 NaCl. Lines are used for orientation. The simulations assume a constant supply of electrolyte at the inlet.

Results – Reconstructed microstructure [1]

- Increasing depletion of active material with reducing flow rate due to insufficient mass transfer
- Decreasing half-cell potential with decreasing SOC and flow rate
- Mass transfer limitations occur for flow rates with Reynolds numbers of around 10^{-2}



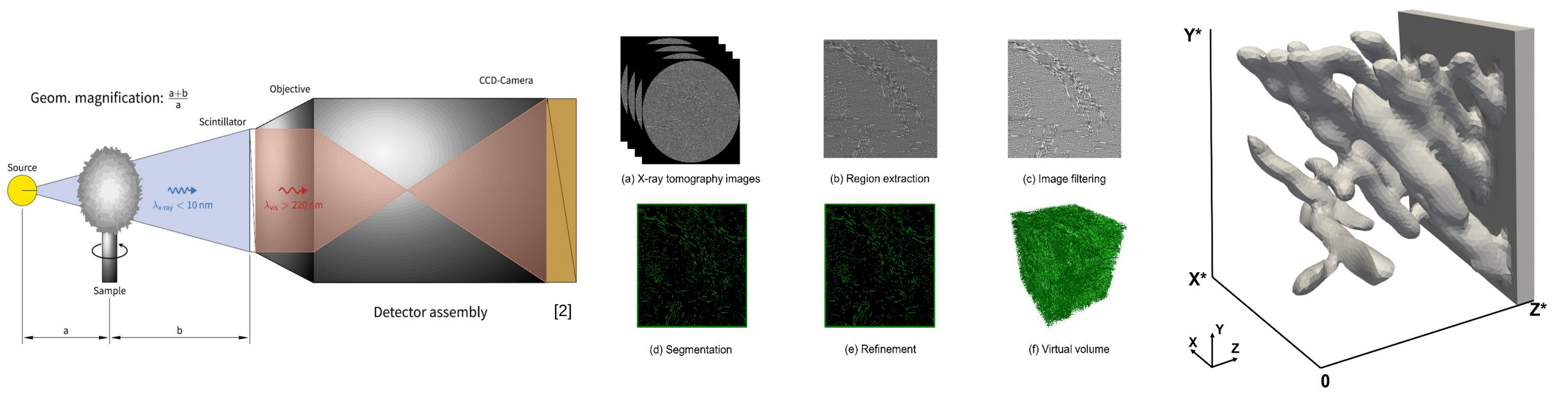
Domain size	78 µm x 78 µm x 78 µm
Specific surface area	58 471 m² m⁻³
Porosity Φ	0.914
Number of cells	2.8 million



Volume-weighted mean value of TEMPO⁺ concentration normalized to the inlet value along the dimensionless Z-direction (Membrane at Z=0 and current collector at Z=1) for different flow rates (left). Half-cell potential over Reynolds number for different SOC. Concentration is 0.1M TEMPO in 1.0M NaCl and galvanostatic discharge current density is 40 mA cm⁻². The simulations assume a constant supply of electrolyte at the inlet.

Microstructure characterization: X-ray computed micro-tomography

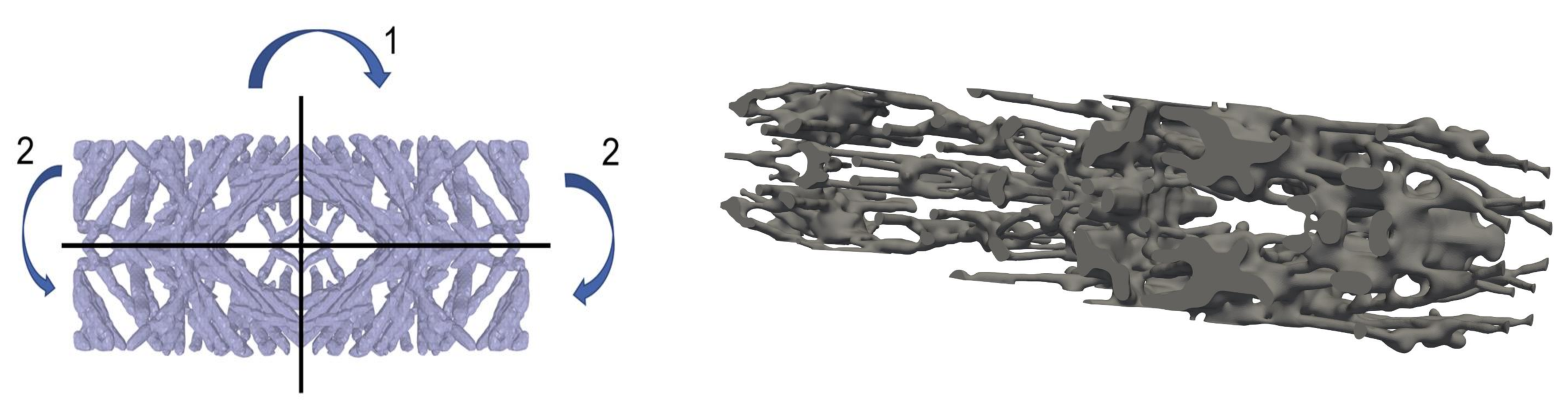
- Method for the image reconstruction of real porous electrodes



µCT Scan → Image processing → Digital twin

Outlook: Generation of representative periodic microstructures

- Development of a method for mirroring the porous electrode micro structure



References

[1] C. Roth, J. Noack, M. Skyllas-Kazacos, Redox Flow Batteries. From Fundamentals to Applications. Weinheim: Wiley-VCH, 2023.

[2] T. Dobler, B. Radel. "Quasi-Continuous Production and Separation of Lysozyme Crystals on an Integrated Laboratory Plant". In: Crystals 11.6 (2021). issn: 2073-4352. doi:10.3390/cryst11060713.

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