

Towards multiscale modeling of porous electrodes: Connecting the meso- to the macroscopic scale

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Redox flow batteries are an emerging technology for grid energy storage applications thanks to their promising properties, such as long cycle life and safety. Porous electrodes are a core component of flow batteries that facilitate the electron transfer between the liquid electrolyte and solid electrode by providing high specific surface areas.

We are interested in macroscopic homogenized descriptions of the coupled processes of mass transport and heterogeneous reactions in porous electrodes, allowing for efficient simulations over macroscopic domains. The effective macroscopic properties, such as the dispersion tensor or the effective reaction rate depend on the pore-scale properties of the porous electrode, such as the morphology and surface properties of the electrode.

Here we consider periodic porous media with simplified geometries, allowing the pore-scale transport problem to be formulated over periodic unit cells. The electrolyte is modelled as a dilute, multicomponent mixture occupying the pore-space Ω_β . Assuming an incompressible Newtonian fluid, the steady-state pore-scale flow can be described by

$$(1) \quad \text{Re}((\mathbf{v} \cdot \nabla)\mathbf{v}) = \Delta \mathbf{v} - \nabla p, \quad \mathbf{x} \in \Omega_\beta$$

and the dimensionless mass transport problem of species α is given by

$$(2) \quad \begin{aligned} \text{Pe}(\mathbf{v} \cdot \nabla c_\alpha) &= \Delta c_\alpha, \quad \mathbf{x} \in \Omega_\beta, \\ -\mathbf{n} \cdot \nabla c_\alpha &= \text{Ki} \cdot r_\alpha, \quad \mathbf{x} \in A_{\beta\sigma}, \end{aligned}$$

where $A_{\beta\sigma}$ is the interfacial area, Pe is the Peclet number and Ki denotes a kinetic number. The source term r_α accounts for interfacial mass fluxes resulting from electrochemical reactions or adsorption of species at the electrode surface.

We use the method of volume averaging [5] for upscaling the pore-scale problem to obtain effective macroscopic descriptions and study their dependence on the pore-scale properties.

In future work we intend to consider more complex electrochemical interface descriptions based on the framework of non-equilibrium thermodynamics [2, 3, 1], which allow incorporating additional interface properties that could be provided by lower-scale descriptions, such as kinetic Monte Carlo simulations.

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REFERENCES

- [1] W. Dreyer, C. Gohlke, and R. Müller. Bulk-Surface Electrothermodynamics and Applications to Electrochemistry. 20(12):939.
- [2] W. Dreyer, C. Gohlke, and R. Müller. Modeling of electrochemical double layers in thermodynamic non-equilibrium. 17(40):27176–27194.
- [3] M. Landstorfer, C. Gohlke, and W. Dreyer. Theory and structure of the metal-electrolyte interface incorporating adsorption and solvation effects. 201:187–219.
- [4] F.J. Valdés-Parada, C.G. Aguilar-Madera, and J. Álvarez Ramírez. On diffusion, dispersion and reaction in porous media. 66(10):2177–2190.
- [5] St. Whitaker. *The Method of Volume Averaging*, volume 13 of *Theory and Applications of Transport in Porous Media*. Springer Netherlands.