Title: Starting from the bottom: Coupling a genetic algorithm and a pore network model for porous electrode optimization

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Porous electrodes are performance- and cost-defining components in modern electrochemical systems as they determine the hydraulic resistance, facilitate mass transport, conduct electrons and heat, and provide surfaces for electrochemical reactions [1]. Thus, electrode engineering is an effective approach to improve cost competitiveness by increasing power density. In convection-enhanced technologies, currently used porous electrodes are fibrous substrates developed for low-temperature fuel cells, but their microstructure and surface chemistry limit the performance of emerging electrochemical systems. Microstructure-informed multiphysics simulations can be leveraged to aid the theoretical design of advanced electrode architectures [2]. However, they have only recently been deployed for the bottom-up design of electrode microstructures [3]. The combination of microstructure-informed multiphysics with evolutionary algorithms could accelerate progress in the optimization of porous electrodes for a given application. In this work, we combine three-dimensional simulations with a genetic algorithm for the bottom-up design of porous electrodes for redox flow batteries.

In the first part of the talk, I will describe a methodology to couple an experimentally validated microstructure-informed, electrolyte-agnostic pore network modeling framework [4] with an evolutionary algorithm [5]. This genetic algorithm is used to optimize electrode microstructures by evolving the structure driven by a fitness function that minimizes pumping power requirements and maximizes electrochemical power output, where the optimization only relies on the electrolyte chemistry and initial electrode and flow field geometries as inputs. The analyzed proof-of-concept employs a flow-through cubic lattice structure with fixed pore positions and shows significant improvement of the fitness function over 1000 generations. The fitness improved by 75% driven by a reduction in the pumping requirements by 73% and an enhanced electrochemical performance of 42%. The evolutionary design resulted in a bimodal pore size distribution containing longitudinal electrolyte flow pathways of large pores and an increased surface area at the membrane-electrode interface.

In the second part, I will discuss our latest progress on the genetic algorithm by implementing integrated flow field geometries, commercial fibrous electrodes as offspring networks, and extended evolutionary freedom during the optimization. Coupling the genetic optimization to the flow field geometry affects the fitness evolution, shifting the balance between electrochemical and hydraulic performance, emphasizing the interaction between flow fields and electrodes. By including additional evolutionary freedom (i.e., by allowing merging and splitting of pores outside fixed coordinates), commercial electrodes can be enhanced by reducing their pumping losses. The presented genetic algorithm offers potential for the predictive design of electrode microstructures tailored for specific electrochemical systems. While applied to flow batteries in this study, this methodology can be leveraged to advance electrode microstructures in other electrochemical systems by adapting the relevant physics.

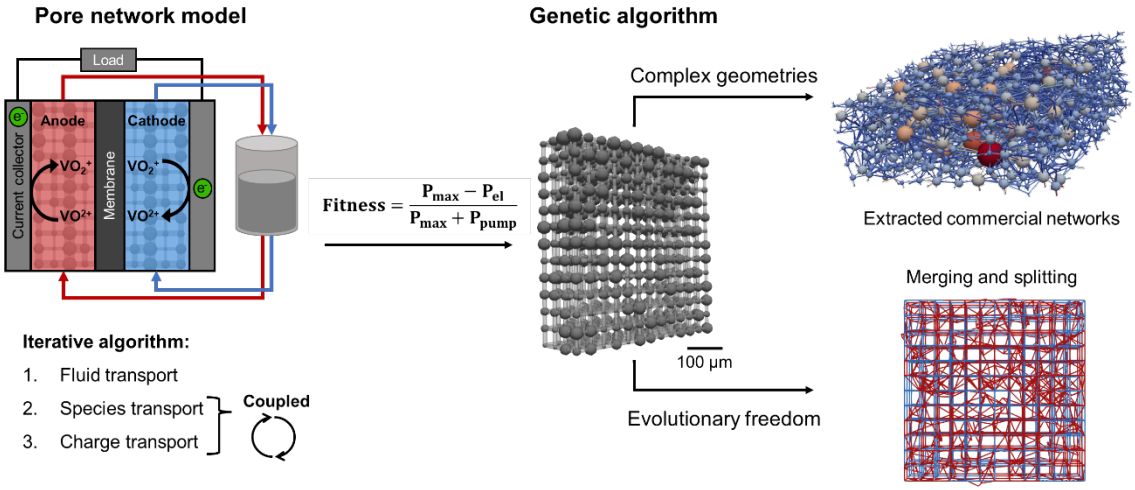

Figure 1: Schematic representation of the pore network model and genetic algorithm framework. The pore network model is solved for the electrolyte fluid transport and couples both half-cells by iteratively solving the species and charge transfer. This model is coupled to a genetic algorithm that evolves the porous electrode microstructure driven by a fitness function that minimizes pumping power requirements and maximizes electrochemical power output. By implementing complex geometries and evolutionary freedom, the genetic algorithm offers great potential for the predictive design of electrode microstructures tailored for specific reactor architectures and redox chemistries

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[3] V. Beck *et al., J. Power Sources,* **512**, 230453 (2021)
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[5] R. van Gorp & M. van der Heijden *et al.*, *Chem. Eng. J.*, 139947 (2022)