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Towards bottom-up design of porous electrode microstructures – coupling evolutionary algorithms and pore network modelling

Maxime van der Heijden[1]*, Rik van Gorp[1], Gabor Szendrei[1], Mohammad Amin Sadeghi[2], Jeffrey Gostick[2], and Antoni Forner-Cuenca[1]

[1]*Eindhoven University of Technology, (Chemical Engineering and Chemistry), (Mem-brane Materials and Processes), 5600 MB Eindhoven, Netherlands*

[2]*University of Waterloo, (Chemical Engineering), Waterloo, ON, Canada*

**m.v.d.heijden@tue.nl*

Porous electrodes are performance- and cost-defining components in electrochemical systems that must facilitate mass transport, provide surfaces for electrochemical reactions, and conduct electrons and heat. Thus, understanding and optimizing the electrode microstructure offers a promising pathway to cost reduction. To accelerate progress, microstructure-informed multiphysics simulations can be leveraged to aid the theoretical understanding and design of advanced electrode architectures. In this work, we explore the following question: *Can we deploy three-dimensional simulations combined with evolutionary algorithms to enable bottom-up artificial generation of porous electrodes?*

In the first part of this talk, I will discuss the modeling framework and experimental validation. Using a pore network modeling open-access platform (OpenPNM), we built a computationally inexpensive, microstructure-informed, and electrolyte-agnostic simulation framework with the focus on redox flow batteries. The model utilizes a network-in-series approach to account for species depletion over the entire length of the electrode and was validated with symmetric flow cell experiments for two distinct electrolytes (aqueous and non-aqueous) and two types of porous electrodes (carbon paper and cloth). The dry electrode microstructures were obtained with x-ray computed tomography and converted into a network of spherical pores and cylindrical throats. The electrochemical performance of the non-aqueous electrolyte was well captured by the model. For the aqueous electrolyte, we find that incomplete wetting of the electrode results in overprediction of the electrochemical performance, which was accounted for by employing a fitting parameter to account for the near-surface mass transfer coefficient.

In the second part of the talk, I will describe a genetic algorithm that optimizes porous electrode microstructures from the bottom-up by coupling the pore network model with an evolutionary algorithm. The microstructure evolves driven by a fitness function that minimizes pumping power requirements and maximizes electrochemical power output. The analyzed systems show significant improvement of the networks' fitness, which increased by 30%. The presented framework offers great potential for predictive design of electrode microstructures tailored for specific redox chemistries and reactor architectures, which will accelerate and broaden the design and fabrication of advanced electrode structures.

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