Elucidating Multiscale, Multiphysics Coupled Transport Phenomena in Polymer-Electrolyte Membranes

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ABSTRACT

The performance of polymer-electrolyte-fuel-cells (PEFCs) is inherently linked to the mass-transport and thermodynamic properties of the polymer membrane. The prototypical class of PEFC-membrane material is perfluorosulfonic-acid (PFSA) polymers, which phase separate into nanoscale hydrophilic water-filled domains in which ion and water transport occurs and a hydrophobic matrix that provides structural integrity and durability. Mass transport across the macroscopic membrane is inherently a multiscale problem with species moving through nanoscale domains that are connected to form a mesoscale transport network that results in macroscopically observable properties. The reasons behind a given mass-transport phenomena in the membrane is therefore often attributed to a given lengthscale without respect for the others. Therefore, to optimize PEFC operation and membrane material design effectively there is a need to develop a framework to decouple and understand the impacts of each lengthscale on each other in terms of mass transport. Such a framework for understanding conductivity and solvent sorption is presented in this work using a multiscale modeling approach.

The molecular-scale interactions between species in hydrophilic domains of PFSA membranes is modeled using a mean-field local-density approach with an experimentally consistent 3D domain geometry and accounts for solvation, electrostatics, solvent dielectric saturation, and finite size and confinement effects. The nanoscale model is validated using atomistic simulations and used to calculate multicomponent transport and thermodynamic properties. The nanoscale simulation results for conductivity, ion migration, and water transport are upscaled to macroscale properties by accounting for the interconnectedness of the hydrophilic domains, which allows for comparison to experimental data. The possibility of using a phase-field, Cahn-Hillard approach to account for the dynamic evolution of the mesoscale network will also be discussed. Using the combined experimental and modeling results, the impact of each lengthscale on transport and solvent uptake are elucidated.

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