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Elucidating Structure-Property Relationships in Highly Permeable Perfluorinated Sulfonic Acid Ionomers

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Abstract

Commercially available perfluorinated sulfonic acid ionomers (PFSAs), utilized as polymer electrolytes in membrane electrode assemblies (MEAs), have driven rapid improvements in fuel cell performance. These materials have a polytetrafluoroethylene (PTFE) backbone and semicrystalline matrix which imparts mechanical integrity and low gas permeability, making them attractive membrane materials. However, their low gas permeability introduces significant mass-transport limitations in catalyst layers, especially severe in oxygen reduction at the cathode. In this study, we present the synthesis of an amorphous PFSA incorporating perfluoro(2-methylene 4-methyl-1,3-dioxolane) (PFMMD) in the backbone. This impacted the material nanostructure at multiple length scales, simultaneously increasing gas permeability (>3x oxygen permeability of Nafion) via reduced crystallinity and increased fractional free volume while reducing proton conductivity via changes in matrix mechanical properties which inhibited phase separation of ionomer domains. When integrated in a fuel cell MEA, this trade-off yielded significant improvements; specifically, current density per cm² platinum increased up to 22% upon substituting the PFMMD based ionomer for Nafion in the cathode binder. In this presentation, I will discuss the facile synthesis of PFMMD based ionomers with tunable PFSA content. Furthermore, I will discuss structure-property relationships resolved from transport measurements and morphological characterization. Finally, I will discuss the implementation of these materials in fuel cells and the potential for meaningful performance improvements. These results demonstrate the value of rational ionomer design toward better performing electrochemical devices.