Interplay of Electrostatic Interactions, Nanoparticle Dispersion, and Ion Transport in Ionomer Nanocomposites

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Ionomer nanocomposites have emerged as a promising replacement to traditional polymer electrolyte membranes for technologies like vanadium redox flow batteries as they curtail undesired vanadium ion crossover while maintaining high proton conductivity. However, studies have shown that the nanoparticles exist in both the hydrophobic and hydrophilic domains of Nafion, indicating that simple steric hindrance, as previously hypothesized, may not adequately describe the mechanism for reduced vanadium ion crossover. In this study, nanoparticle surface chemistry, along with nanoparticle loading and diameter, were systematically altered to tune nanoparticle dispersion within the ionomer (also known as the nanoparticle 'dispersion state'). The impact of the nanoparticle dispersion state on vanadium ion crossover was measured via ultraviolet-visible. Additionally, electron imaging was used to determine the impact of these interactions on the nanoparticle dispersion state. Results indicate that the silica nanoparticle surface chemistry plays a vital role in controlling both nanoparticle dispersion and ion transport through the nanocomposite membranes. Specifically, silica nanoparticle aggregation reduced vanadium ion crossover as compared to well-dispersed particles with the same end functionality. Additionally, the overall nanoparticle surface charge is a key factor in controlling vanadium ion permeation through these composite membranes.