Fuel Cell Electrode Structures Containing Sulfonated Organosilane-Based Proton Conductors

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Current fuel cell technologies rely on environmentally and financially costly materials to achieve optimum performance. Two of these materials are platinum and Nafion®. Ceramic carbon electrodes (CCEs) have been proposed as a support for the platinum catalyst, as they require cheaper materials while maintaining or exceeding current fuel cell efficiency. CCEs consist of electronically conductive carbon particles bound by a ceramic binder. In this work, the ceramic binder is an organosilane material which is much cheaper than Nafion®. The organosilane component will create conductive channels through which electrons and protons can flow. Incorporating sulfonated organosilane precursors into the electrode structure would facilitate enhanced proton transport.

CCEs are prepared via the sol-gel process, forming a porous network of suspended carbon-supported platinum and liquid phase. The key advantage of ceramic electrode structures is that organosilanes will polymerize around the solid support, increasing the ability of protons to reach platinum sites. Liquid ionomer polymerizes upon addition to the catalyst layer resulting in a thoroughly mixed, evenly distributed ionomer. Silicate ionomer can also chemically bind to the catalyst via surface hydroxyl groups on the carbon support, providing the proton conductor with improved proximity to the active catalytic surface.

Previous research determined CCEs can be successfully prepared using unsulfonated organosilane precursors. Optimal performance composition was determined through an extensive electrochemical study. While Nafion®-based electrodes outperformed the unsulfonated CCEs, their conductivity and active area were impressive considering a lack of proton-conducting moieties.

In this study, ceramic carbon electrodes have been prepared using variable quantities of carbon-supported platinum and the sulfonated organosilane precursor 3-(trihydroxysilyl)-1-propane sulfonic acid (THSPS). To date, CCEs with silica loadings of ~50-75% have been studied via thermogravimetric analysis and electrochemical studies. Half-cell cyclic voltammetry (CV) has shown that at high silane concentrations, active platinum area decreases as silane loading increases. This is similar to the results for unsulfonated CCEs, as high silicate concentrations result in clogged pore structures and high electronic resistance.

Electrochemical impedance spectroscopy (EIS) data gathered for the CCEs was analyzed using Nyquist and capacitance plots. Impedance results were compared to standard Nafion®-based electrodes and the most active unsulfonated CCE from previous testing. The Nyquist plot indicated sulfonated CCEs are less resistive than for CCEs without sulfonate groups. The capacitance plot illustrates high proton conductivity in sulfonated CCEs, as they have steeper initial slopes than the unsulfonated CCE. Limiting capacitance is directly proportional to active area, and limiting capacitance data confirms CV results, indicating the CCE active area decreases at higher silicate concentrations.

Composite CCEs were created with both sulfonated (THSPS) and unsulfonated (tetraethyl orthosilicate) silanes to determine the optimal degree of sulfonation for proton transport. With a fixed silane composition of 45%, the ratio of sulfonated to unsulfonated monomer was varied. This effectively varies the ion exchange capacity (IEC). The results thus far suggest sulfonated CCEs have potential to surpass the electrochemical performance of materials previously designed and tested for fuel cell applications.

References: