Electrochemical Investigation of Ceramic Carbon Electrodes for Low Humidity Fuel Cell Applications

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State-of-the-art electrodes for proton exchange membrane fuel cells are composed of platinum catalyst and a Nafion® proton-conducting binder. These are expensive materials, and Nafion® suffers from a narrow range of optimal operation conditions. Ceramic carbon electrodes (CCEs) are an alternative electrode structure which may provide improved access to platinum reaction sites. We have replaced Nafion® with organosilane materials as the binder, due to their low cost and ability to tolerate a wider range of temperature and humidity conditions. The catalyst material is added to organosilane precursors, which grow a polymer network *in situ* via the sol gel process to form a porous structure that contains an evenly distributed ionomer.

We have previously demonstrated addition of small amounts of sulfonated organosilane precursor enhanced proton conductivity and structural characteristics of ceramic carbon electrodes¹. Fuel cell testing of CCE cathode catalyst layers suggested high Pt electrochemical surface area (ECSA) and similar performance under standard operating conditions (80°C, 100% RH) compared to a commercially available cathode catalyst layer containing Nafion®.² To better evaluate applicability as cathode materials, a range of operating conditions must be examined.

Here we will present an overview of structural and electrochemical characteristics of commercial Nafion®-containing ELAT cathodes to CCE cathodes prepared with 6% sulfonate content. Membrane electrode assemblies (MEAs) were monitored over several start-stop cycles and multiple relative humidities (RH). Cyclic voltammetry showed ECSA of Pt for both cathodes decreased over several days of operation. Impedance spectroscopy data was analyzed via Nyquist and capacitance plots. Nyquist plots suggested that catalyst layer resistance increases over time, while capacitance plots illustrate proton conductivity decreases over multiple start-stop cycles. Fuel cell polarization curves suggested that while initial performance of ELAT and CCE cathodes are similar, CCE cathodes are prone to

flooding at 80°C and 100% RH which limits performance at high current densities.

Cathode RH was varied between 20-100% to study performance under dry conditions. Impedance results show higher resistance and lower proton conductivity for both cathodes as %RH decreases, however limiting capacitance was higher for CCE cathodes at each % RH. Figure 1 suggests as % RH decreases, flooding of CCE cathodes is alleviated and performance is maintained as low as 20% RH. Performance of ELAT electrodes decayed with reduced cathode RH and could not be operated below 43% RH on our system.

Figure 2 displays the membrane resistance for MEAs containing identical membranes but CCE or ELAT cathodes. The plot suggests that use of CCE cathode materials results in lower membrane resistance. This may explain retention of performance at low relative humidity, as water from the cathode can undergo back diffusion which aids in membrane hydration under dry conditions.



Figure 1. Comparison of H_2/O_2 fuel cell polarization curves obtained for (a) ELAT and (b) SS-CCE cathode catalyst layers at multiple relative humidities.



Figure 2. Membrane resistance as a function of relative humidity for SS-CCE cathode and ELAT cathode at 80° C.

References:

- 1. J.I. Eastcott, J.A. Powell, A.J. Vreugdenhil, and E.B. Easton, *ECS Trans.* **41**(1) (2011) 853 864.
- 2. J.I. Eastcott, K.M. Yarrow, A.W. Pedersen, and E.B. Easton, *J. Power Sources.* **197**, (2012) 102 106.