SiO_x Impregnated Electrodes in PEM Fuel Cells for Improved High-Temperature Perfomance

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Operation of PEM fuel cells at 80°C poses problems of CO tolerance, slow kinetics of the oxygen reduction reaction (ORR) and poor thermal and water management. Many of these problems can be resolved by running PEM cells above 100°C, however, the conductivity of the Nafion membranes is the highest only when the membrane is fully hydrated, and operating the fuel cells above ~90°C dries out the membrane. Consequently, the Nafion in the active layer of the electrode also dries out at these temperatures causing increased resistance in the electrodes decelerating the ORR. Several attempts have been made to overcome high temperature dehydation issues; one reported method being to make a composite Nafion membrane that can retain water in at high temperatures[1-3]. However, most of the previous efforts have not attempted to modify the electrocatalyst layer of the electrodes to enhance catalytic activity at elevated cell temperatures.

This paper shows that the modification of the active layer of the electrodes improves the performance of the fuel cell above 100°C. In the present study, silicon oxide was incorporated into the Nafion membranes and also into the active layer of the electrodes. Figure 1 compares the polarization curves of high temperature cells based on commercial Nafion 115 membranes at 130°C and 30 psig reactanct gases with and without silica sol-gel impregnated electrodes. Figure 2 compares the polarization curves of high temperature cells based on recast Nafion silica composite membranes at 130°C and 30 psig reactanct gases with and without silica sol-gel impregnated electrodes. The electrodes impregnated with silica sol-gel clearly show an increase in performance of PEMFC above the water boiling point.

The improvement is related to a net decrease in MEA resistance. The probable mechanism involves a decrease in the interfacial charge transfer resistance at elevated temperature due to an improvement in water retention at the catalyst interface. The concomitant addition of silica to the membrane and the electrode may also enhance the mechanical strength of the membrane electrode assembly and elevate the glass transition temperature of the system making it possible to temperature-cycle the PEMFCs below and above 100°C.

References:

1. K. T. Adjemian, S. J. Lee, S. Srinivasan, J. Benziger and A. B. Bocarsly, Journal of the Electrochemical Society, In press 2001.

2. Yongchao Si et. al., Abstract No.116, 199th Meeting of Electrochemical Society at Washington, D.C.

3. C. Yang, P. Costamagna, S. Srinivasan, J. Benziger and A. B. Bocarsly, J. Power Sources, 2001, 103, 1.