Effect of Porosity on the Oxygen Reduction Reaction in PEM Fuel Cell Cathode Electrodes

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ABSTRACT

In order to maximize the utilization of the electrocatalyst in proton exchange membrane (PEM) fuel cell cathode electrodes, it is necessary to understand the impact of electrode micro- and nano-structure on cell efficiency. In an ideal electrode architecture, the catalyst (typically Pt) is structured in such a way that maximizes utilization. For current commercial Pt/C nanoparticle electrodes, Pt utilization is limited to 10%-20% due to simultaneous percolation requirements of the ionic, electronic and gas phases at a given catalyst site. Sputtering has recently emerged as a path for applying low loadings of Pt in a consistent, scalable process, and it is possible to utilize 100% of the exposed surface area of the resulting ultrathin electrode. However, the absolute power density of sputtered electrodes is low, because such layers have low porosity. The goal of this research is to create high-utilization sputtered electrodes with controllable porosity, and to quantify the impact of that porosity on the power conversion efficiency of PEM fuel cell cathode electrodes.

First, it is shown that the porosity of sputtered layers can be controlled by varying the deposition angle. Glancing-angle deposition (GLAD) of Pt onto the gas diffusion layer results in Pt nanorods; by comparing the efficiency of nanorods with continuous sputtered layers deposited from normal incidence, the impact of porosity on catalyst utilization is quantified. Next, this approach is extended by building core/shell supported catalysts comprising CrN nanorods coated with Pt. These supported catalysts have higher Pt utilization than solid Pt nanorods, since only Pt on the outer surfaces is exposed to oxygen. However, the hydrophobic Pt/CrN materials system exhibits a reversible de-wetting of electrode pores at low water production rates, which results in

suppressed output current at high potential due to poor proton conduction within the electrode. This de-wetting is the first observation of incomplete wetting in ultrathin sputtered electrodes. Third, a novel electrochemical etching process is described, in which pores are controllably created within a running fuel cell by applying an electrolysis current to the cell. It is shown that etching carbon nanorods is an effective means of controlled pore generation, in which mass transport can be tailored by the amount of etching charge applied. This etching process could eventually be used to open pores of precise position and morphology in arbitrarily complex structures fabricated by a 3-D lithography process. Finally, the morphology of the pores between the nanorods is controlled using GLAD on a patterned substrate. The minimum pore size is determined by the substrate pattern, resulting in pores with a more uniform width, which in turn results in more effective mass transport of oxygen within the electrode. Sputtered electrodes with controllable porosity may lead to future fuel cells with higher power densities and lower cost.