Fabrication and Electrochemical Performance of High Surface Area Catalyzed Carbon Fiber Cathodes

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A new approach has been developed for preparing high surface area arrays of carbon microfibers. These catalyst supports are prepared using a novel method called electrostatic flocking. The carbon microfibers (nominally 10 µm in diameter and 500 µm long) are oriented normal to a flexible graphite current collector. This high surface area architecture is being used to project a H₂O₂ reduction catalyst into the cross section of a flowing catholyte in a Mg – H₂O₂ semi-fuel cell (SFC). The objectives are to reduce H2O2 diffusion distances, the effects of concentration polarization, and increase cell power density. Because of the higher surface area and penetration of catalyst into the flowing electrolyte, the carbon microfiber array based SFC shows better rate capability and higher H₂O₂ utilization than a SFC which uses a planar carbon paper cathode.

In electrostatic flocking, the carbon fibers are evenly spread on a stainless steel plate which is connected to a high voltage source. A flexible graphite foil which has been coated with a carbon-based electrically conductive adhesive is at ground potential. The SS plate and graphite foil are aligned in parallel and separated by 10 cm. Upon application of the high voltage, the charged fibers travel across the electric field (typically 5 kV/cm) and embed in the wet adhesive. After flocking, the fiber/wet adhesive/foil composite is heated to cure the conductive adhesive. Scanning electron microscopy shows that the fibers protrude from the adhesive and graphite current collector like bristles of a brush (Figure 1).

To form High Electrolyte Penetration (HEIP) cathodes, an alloy of Pd and Ir [1] is electrochemically deposited onto the carbon microfiber array (CMA). Figure 2 shows a cross section of a typical HEIP electrode, a Pd:Ir catalyzed CMA. Cauliflower-in appearance Pd:Ir nodules grow at the tips of the fibers while a smoother coating appears over the length of the fibers.

SFCs are novel electrochemical couples being developed as low power, high specific energy sources for unmanned undersea applications by the United States Navy [2]. More specifically, a Mg-H₂O₂ SFC currently under development uses a flowing catholyte of H₂O₂, 0.2 M H₂SO₄ and 40 g/L NaCl.[3] The polarization and constant current performance of Mg-H₂O₂ SFCs have been characterized using high surface area HEIP cathodes and planar (control) carbon paper cathodes.

The rate capabilities of SFCs with HEIP and planar carbon paper cathodes are shown in Figure 3. The higher surface area of the HEIP electrode results in higher cell voltages at high current densities.

Constant current profiles for the SFC with a HEIP cathode and a carbon paper control cathode further show the advantage of increased surface area. At 25 mA/cm² and 60 mM H_2O_2 , the HEIP cathode SFC

maintained a cell voltage which was 0.40V to 0.45V (30%) higher than the carbon paper SFC.

Tests currently underway show that the HEIP electrode based SFC has a higher specific energy and shows better H₂O₂ utilization than the carbon paper SFC.



Figure 1. Electron micrograph of carbon microfiber array showing graphite current collector, conductive carbon adhesive, and pitch-based carbon fibers.



Figure 2. Electron micrograph of HEIP electrode showing Pd:Ir catalyzed carbon microfiber array.



Figure 3. Polarization of $Mg-H_2O_2$ SFCs with HElP and catalyzed carbon paper cathodes. Because of higher electrode surface area, the HElP based SFC has better rate capability.

References:

1. "Investigation of a Palladium-Iridium Catalytic Surface Formed by Cyclic Voltammetry", R.R. Bessette, M.G. Medeiros, C. M. Deschenes, and L.G. Carreiro, 197th Meeting of the Electrochemical Society, 14-18 May, 2000, Toronto, Canada

 "Magnesium-Solution Phase Catholyte Seawater Electrochemical System", M. G. Medeiros and E. G. Dow, *Journal of Power Sources*, 80, May 1999, 78-82
"Magnesium-Seawater/Acid Solution Phase Catholyte Semi-Fuel Cell System", M.G. Medeiros, R.R. Bessette, 39th Power Sources Conference, Cherry Hill, NJ, June 2000, pp 453 of Proceedings