V.L.5 PEMFC Using Aligned Carbon Nanotubes as Electrodes in MEA

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Objectives

- To develop a method of preparing aligned carbon nanotube layers as electrode catalysts for proton exchange membrane fuel cells (PEMFCs).
- To develop a method of fabricating membrane electrode assemblies (MEAs) and PEMFCs using aligned carbon nanotube based electrodes.
- To evaluate the performance of aligned carbon nanotube-based proton exchange membrane (PEM) fuel cells.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(A) Durability
(B) Cost
(C) Performance

Technical Targets

This project aims at developing an MEA with an innovative architecture using aligned carbon nanotubes (ACNT) as the electrode catalyst layer. It targets simplified cell design with reduced platinum loading, enhanced power density, and stability for PEMFCs by placing catalytically functionalized ACNT layers with three-dimensional patterns between the ion-conductive membrane and current collector. The successful outcome of the project would help develop PEMFCs that meet DOE 2010 stack targets:

- Power density: 650 W/L
- Cost: $45/kWe

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Approach

Our approach is to transform the conventional MEA to an ACNT-based electrode assembly, as illustrated in Figure 1. The concept is made possible based on recent progress at Argonne in developing a three-dimensional patterned ACNT electrode catalyst layer using a template-free, chemical vapor deposition (CVD) process [1,2]. The conventional catalyst layer in a PEMFC is prepared with an ink-based preparation method, with which it can be difficult to achieve the maximum "triple phase boundary", where ion-electron conduction and gas phase-surface reaction occur simultaneously. In an ACNT-based MEA, vertically oriented carbon nanotube layers are synthesized and functionalized with the electro-catalytic active sites before being transferred to the surface of the electrolyte membrane. The proton and electron transfers take place at the surface of nanotubes, which is also exposed to reactant gases. Such an electrode structure has the potential to optimally utilize the catalyst material, such as Pt, with unobscured exposure. The direct contact between the electrolyte and the current collector through the nanotube layers could also improve both electronic and thermal conductivities. In addition, the ACNT "carpet" can be fabricated with a pre-designed gas flow field through a 3-D patterning process, and thus can further improve

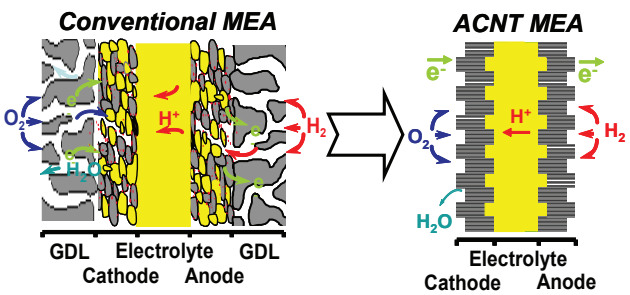

FIGURE 1. Schematic Comparison of the Electrode Construction between Conventional and ACNT-Based MEAs

mass transfer and water management with a thinner stack configuration.

Accomplishments

Since the inception of the project, we have accomplished the following tasks as outlined in our initial work plan:

- Optimized ACNT synthesis process: we have conducted a variety of experiments aimed at improving ACNT morphology and stability. Nanotubes with different geometries (diameter, length, etc.) and crystallinities were prepared with various CVD reaction conditions and precursors. Shown in Figure 2 are the scanning electron microscopy (SEM) images of some ACNT samples prepared in our laboratory. Thermal oxidative stability of ACNTs prepared under different conditions was also investigated by temperature-programmed oxidation.

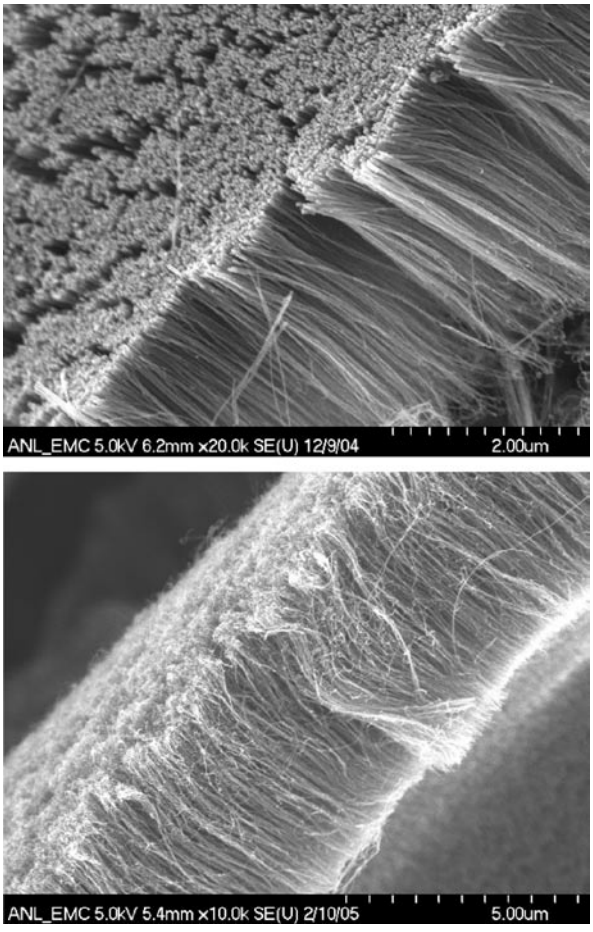

FIGURE 2. SEM Images of ACNTs Prepared by Different CVD Processes

- Developed a catalyzing method tailored for ACNT: due to its high graphitic nature and the need for intact alignment, new catalyzing methods have to be developed for ACNT-based electrode material. Two parallel techniques are currently under study in our laboratory to functionalize aligned nanotubes. They are gas phase co-CVD and wet chemistry approaches, respectively. Some preliminary progress has been made, through which the ACNTs were decorated with highly dispersed Pt crystallites with dimensions in the range of 2 nm to 10 nm.

Special Recognitions & Awards/Patents Issued

1. "Method of fabricating electrode catalyst layers with directionally oriented carbon support for proton exchange membrane fuel cell," Di-Jia Liu and Junbing Yang, *US patent application 20060269827.*

2. "Aligned carbon nanotube with electro-catalytic activity for oxygen reduction reaction," Di-Jia Liu and Junbing Yang, *US patent application filed 2006.*

FY 2007 Publications/Presentations

1. "Functionalized Aligned Carbon Nanotubes as a Novel Catalytic Electrode for PEM Fuel Cells," Junbing Yang, Di-Jia Liu, and David J. Gosztola, poster presentation at *Fuel Cell Seminar*, Nov. 11–13, 2006, Honolulu, HI.

2. "Investigation of Aligned Carbon Nanotubes as a Novel Catalytic Electrodes for PEM Fuel Cells," Di-Jia Liu, Junbing Yang, and David J. Gosztola, ECS Transactions, 5 (1) 147–154 (2007).

3. "Effect of nitrogen dopant type on the structure and electrocatalytic activity of aligned carbon nanotubes," Junbing Yang and Di-Jia Liu, oral presentation at *211th Electrochemical Society Meeting*, May 6–11, 2007, Chicago, IL.

4. "Novel PEMFC Stack Using Patterned Aligned Carbon Nanotubes as Electrodes in MEA," Di-Jia Liu, Magali Ferrandon, Nancy Kariuki, Jennifer Mawdsley, Suhas Niyogi, and Junbing Yang, poster presentation at *2007 DOE Hydrogen Program Annual Merit Review Meeting*, May 15–18, 2007, Washington, D.C.

5. "Functionalized Aligned Carbon Nanotubes as Pt-free Electrocatalyst with Novel Nanoarchitecture for PEMFC," Junbing Yang and Di-Jia Liu, oral presentation at *20th North American Catalysis Conference*, June 17–22, 2007, Houston, TX.

References

1. Vajtai R., Wei B.Q., Ajayan P.M., *Phil. Trans. R. Soc. Lond.* A 2004; 362: 2143-2160.

2. Yang J., Dai L., Vaia R.A., *J. Phys. Chem. B.*; 2003; 107(45): 12387-12390.