Increasing PEM Fuel Cell Catalyst Effectiveness Through Sputter Deposition

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In a recent solicitation, the U.S. Department of Energy set long term goals for Proton-Exchange Membrane fuel cells (PEMFCs) performance in a 50 kW stack that included operation with cathode loadings of 0.05 mg/cm^2 or less of precious metals (1).

The goal of this work was to examine the sputter-deposition technique as a means to improve performance and/or reduce the catalyst loading of proton-exchange membrane (PEM) fuel cells. First Pt was sputtered on the different substrates (Nafion® 117 membranes, uncatalyzed GDLs (Toray) and membrane-electrode assemblies (MEAs)) that comprise individual fuel cells. These were built into MEAs and compared to MEAs made through ink based methods and ink-based MEAs augmented through sputter-deposition. Nafion® membranes were then subjected to the layered technique first developed by Cha and Lee (2) with the goal of reducing the amount of Pt catalyst used and increasing fuel cell performance. Based upon the findings, improvements were made on Cha and Lee's technique allowing for the manufacture of thinner, more effective electrodes for MEAs.

All MEAs generated and/or modified through sputter-deposition were compared to baseline MEAs made through traditional catalyst ink-based preparation techniques (3). Anode and cathode target loadings for the baseline ink-based MEA were both 0.15 mg Pt/cm^2 for a combined MEA loading of 0.30 mg Pt/cm^2.

Of the three substrates studied (membrane, GDL and MEA), sputter-depositing Pt on the GDL showed the best performance, equaling that of the baseline MEA for an equivalent amount of Pt. However, sputter depositing multiple layers of Pt on the GDL showed no improvement in performance over an equivalent amount of sputtered-deposited Pt as a single layer.

The multi-layer electrode technique (2) was used to increase the regions of active Pt area by increasing the number of layers of sputter-deposited Pt. MEAs were prepared from PEMs with multiple layers of sputter-deposited Pt and spray deposited Nafion-Carbon Ink (NCI - containing Nafion, carbon and no catalyst) as shown in Figure 1.

To determine the effect of spraying multiple catalyst layers on the anode and cathode sides of the PEM, three different layered catalyst structures were fabricated: (1) An anode and cathode consisting of Nafion-Carbon Ink NCI + (1 minute of sputter-deposited Pt + NCI) x 3, (2) an anode and cathode consisting of NCI + (0.5 minutes of sputter-deposited Pt + NCI) x 6, (3) an anode and cathode consisting of NCI + (0.5 minutes of sputter-deposited Pt + NCI diluted 1:5 with isopropanol) x 6.

MEA #2 contains the same amount of Pt as MEA #1, but MEA #2 contains twice the amount of Pt-NCI interfaces as a result of doubling the number of Pt layers from three to six. The active area and hence cell performance was found to be a function of the number of sputtered layers and not the amount of Pt. Therefore, MEA #2 has roughly double the active area of MEA #1. MEA #3 contains the same number of Pt layers as #2, but the NCI used in #3 was diluted in order to reduce the thickness of the NCI region between each Pt layer.

Figure 2 shows that as the number of electrode layers is increased from three (MEA #1) to six (MEA #2), the cell performance increases (0.105 to 0.132 A/cm^2 at 0.6 V). However, there is not a doubling of performance that accompanies the doubling of the active area. This is because the spray deposited NCI layer was ~12 μm thick. Therefore for MEA #2, the anode and cathode stacks are roughly ~70 μm thick (the thickness of the Pt is negligible). This was much thicker than the anode and cathode of the baseline MEU; which were typically 5-10 μm thick. The NCI was found to comprise at least 99.9% of the thickness of the multi-layer electrodes. The increased diffusional, ionic and electronic resistances caused by the increased thickness of the electrodes of MEA #2 versus MEA #1 are likely causes for the limited performance increase seen in Figure 2. Versus the 3-layer electrode, gases, protons and electrons must travel further in the 6-layer electrode to get to and from all available Pt sites. Like the layers of sputter-deposited Pt, it is not necessary for the NCI layers to be so thick. To achieve a multi-layered electrode of less than 10 microns (MEA #3), a dilution of 1:5 NCI to isopropanol was used.

As shown in Figure 2, MEA #3 outperforms MEA #2 0.170 to 0.132 A/cm^2 at 0.6 V. This value is closer to double that of MEA #1, containing 3-layer electrodes. The loading for each electrode for the MEA#3 was ~0.0324 mg Pt/cm^2 anode/cathode (0.0648 mg Pt/cm^2 in the entire MEA) resulting in an activity of 2650 A/g Pt at 0.6 V, the highest value of all MEAs tested and nearly three times greater than the baseline (905 A/g).

References

1. Solicitation For Financial Assistance Applications (SFAA) NO. DE-RP04-01AL67057 Research And Development And Analysis For Energy Efficient Technologies In Transportation And Buildings Applications. November 21, 2000.
2. S. Y. Cha, W.M. Lee. *J. Electrochem. Soc.*, **146**, 4055 (1999).
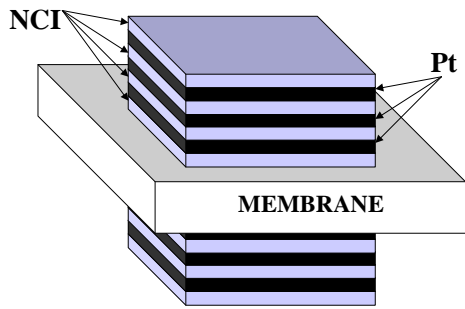3. M. S. Wilson. United States Patent #5211984. May 18, 1993.

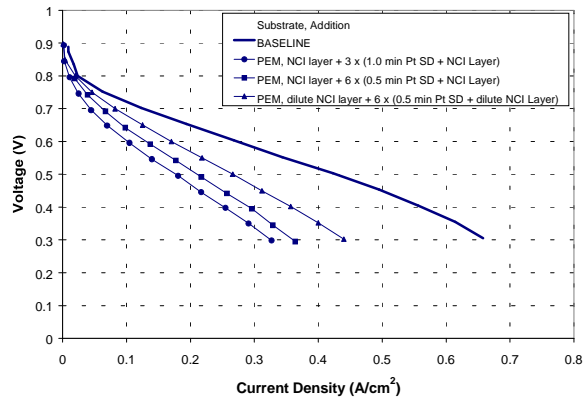Figure 1: Representation of a three layer Pt-NCI MEA.

Figure 2: Performance comparison of MEAs comprised of 3 and 6 layers (anode and cathode) using pure NCI. P = 1 atm, T = 70°C.