

High-Performance, Matching, PEM Fuel Cell Components and Integrated Pilot Manufacturing Processes

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Subcontractor: General Motors Corporation, Honeoye Falls, NY

Objectives

- Develop a set of high-performance, matched proton exchange membrane (PEM) fuel cell components and pilot manufacturing processes to facilitate high-volume, high-yield stack production.
- Demonstrate the matched component performance in a 1-kW fuel cell stack.

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 R,D&D Plan:

- O. Stack Material and Manufacturing Cost
- Q. Electrode Performance

Approach

Phase 1

- Utilize the 3M-patented, nanostructured thin film catalyst support system and catalyst deposition process to easily generate new compositions and structures of (a) binary and ternary catalysts for higher cathode performance and (b) binary anode catalysts and electrode structures for higher reformate tolerance with a non-precious metal replacement for Ru.
- Develop carbon electrode backing/gas diffusion layer (EB/GDL) media optimized for performance with the catalyst system and flow field.
- Optimize the fuel cell flow field design for optimized water management and air bleed utilization with the catalyst and EB/GDL components.
- Define pilot manufacturing scale-up of the processes for fabrication of catalysts, catalyst-coated membrane (CCM) assemblies, and EB/GDL.

Phase 2

- Optimize pilot-scale manufacturing of roll-good fabricated (made by a semi-continuous process and stored on a roll) catalyst support films and catalyst deposition.

- Specify membrane electrode assembly (MEA) component parameters and conduct final pilot-scale runs to generate process statistics.
- Fabricate, test, and provide a 1-kW stack to subcontractor for evaluation using optimized flow field and matched MEAs fabricated by pilot processes.

Accomplishments

- Completed multiple pilot-scale roll-good fabrication runs and statistical evaluation of nanostructured catalyst support films with multiple lots of cathode (PtAB) and anode (PtM) catalyst depositions on the substrates.
- Completed multiple pilot-scale fabrication runs and statistical evaluation of multiple lots of roll-good fabricated CCM assemblies, using multiple lots of PEMs and catalyst roll-goods.
- Completed fuel cell testing and statistical analyses of roll-good fabricated EB/GDL materials and CCMs having matched properties for optimum performance.
- Fabricated and tested two air-cooled and one liquid-cooled stack designs (> 1 kW) with unique compression, manifolding, and cooling features for evaluating 200-cm² MEAs.
- Completed successful liquid-cooled stack testing of MEAs made with roll-good fabricated, nanostructured, thin film catalysts and matching components at subcontractor and 3M facilities. Obtained 1.9 kWe + 2.25 kW heat at 62% air utilization under "Vehicle Test Conditions" with ~ 0.34 mg Pt/cm²/MEA total. Results meet DOE 2005 target of 0.6 g Pt/peak kW.

Future Directions

- Contract period of performance ended April 30, 2003.
- New work on high-performance, lower-cost MEAs, which utilizes the accomplishments of this contract, is being conducted under DE-FC04-02AL67621, "Advanced MEAs for Enhanced Operating Conditions."

Introduction

The MEA is the core component set of a PEM fuel cell stack. An MEA consists of five basic components: anode and cathode catalysts, ion exchange polymer membrane, and anode and cathode electrode backing/gas diffusion layers (EB/GDL). The functions of these basic components are intimately related, and their properties must be matched for optimum performance. For large-scale volume fabrication at the costs and quality targets required by transportation applications, very high yields and in-line process control of integrated processes based on cost-effective materials are required. To realize the full performance of the MEAs in a stack, the stack design must be optimized so as not to compromise the single-cell performance of the MEA. This requires understanding how to design the coolant and gas manifolds and

compression control scheme and match the flow fields to the EB/GDL to obtain uniform reactant, temperature and current density distributions. This contract was directed towards demonstrating high-performance, matching PEM fuel cell components manufacturable by integrated pilot processes in a 1-kW sized stack.

Approach

The approach to the development of high-performance MEAs involved the development of components that were individually optimized and then matched for best overall performance at a particular set of operating conditions. It included the development of new anode and cathode catalysts directed at increasing performance and stability and reducing precious metal loading, using the 3M nanostructured, thin film catalyst support system and

catalyst deposition process, which easily generates new compositions and structures via a dry, roll-goods process. The approach also included development of roll-goods EB/GDL media optimized for performance with the catalyst system and flow field and of a flow field design for optimized water management and air and fuel utilization. Evaluation of each component involved a variety of characterization techniques and single-cell fuel cell performance measurements under a broad range of controlled conditions. Pilot-scale, roll-goods manufacturing processes were optimized for fabrication of catalyst support films and catalyst depositions, CCM assemblies, and EB/GDL media. Process statistics were gathered in pilot-scale runs to assess the potential for high-volume manufacturing. Stack development was carried out to optimize uniformity of the stack reactant flows, compression, and temperature distributions over the MEA. Finally, performance of the matched components was demonstrated in short stacks (>1 kW), which were assembled at 3M and evaluated at the subcontractor's facilities and at 3M.

Results

In the FY 2002 Annual Progress Report for this contract (Ref. 1), results were presented for Pt binary and ternary cathode catalyst constructions that outperformed pure Pt. In particular, MEAs made with a ternary cathode designated as PtAB-2 and a Pt anode were highlighted. The single cell performance with lab-scale fabricated roll-good MEAs corresponded to the DOE 2005 target of 0.6 g/kW at 0.8 V and 0.3 g/peak kW of precious metal. During Phase 2, the nanostructured thin film catalyst support system and the PtAB catalyst deposition process were successfully scaled up on pilot line equipment, and the same fundamental performance enhancements of the ternary were maintained. A new binary anode catalyst, "PtM," having a non-precious metal replacement for Ru had shown promise in Phase 1, and its scale-up was started in Phase 2 along with testing of MEAs made with PtM/PtAB. However, it did not perform as well with the PtAB cathode catalyst as PtRu. Also, changing from the lab to the pilot-scale process affected its performance. Because optimizing the PtM construction and process would have required more time than was available on the contract, the PtRu

anode was selected for scale-up. Both the PtRu and the PtAB nanostructured catalysts were successfully scaled up in a pilot production facility. Multiple lots were fabricated by roll-good processes on pilot production equipment and used in pilot scale-up experiments for the CCM assembly process. Characterization of the down-web and cross-web catalyst composition and loading verified a high level of uniformity.

The CCM process experiments, in which the anode and cathode catalyst layers were transferred from the customized nanostructured substrate to the PEM, were run on pilot-scale, roll-goods equipment using multiple lots and types of catalyst and PEM (3M fabricated) roll-goods. This included catalyst lots that had been coated on reused substrate as a way to reduce costs. Extensive characterization of the roll-good fabricated CCMs to validate properties such as precious metal loading and to investigate variability indicated that the processes had been successfully scaled up and also that the reuse of the substrate was successful. The CCMs used in the stack tests discussed below were fabricated in the final pilot-scale runs.

The EB/GDL component was developed and the processes scaled up in Phase 1, and preliminary results were reported in Ref. 1. During Phase 2, statistical analyses of the process variability for key response variables were completed, and a nested design of experiments was designed and completed for fuel cell response variables. The fuel cell response was less sensitive than the EB/GDL off-line physical property responses, which indicated that good process control for those key responses is sufficient for low variability in the EB/GDL fuel cell performance. Overall, the assessment indicated good potential for meeting anticipated process capability requirements in high-volume production.

Cost projections were made using a detailed proprietary cost model for the MEA components and roll-goods processes. Different production scenarios were evaluated beginning with the processes and facilities currently used and then calculating improvements in areas identified with significant cost reduction potential. The final scenario utilized hypothetical values of process, labor, and material costs that might be realized when a high-volume

market size is reached. Under the final scenario using the detailed cost model, the MEA cost is not inconsistent with currently projected high-volume, long-term automotive pricing requirements.

The performance of the matched components was successfully demonstrated in stacks developed at 3M with consultation from the subcontractor. The stack design goal was to optimize the uniformity of the compression and the reactant and coolant flows for the MEA well enough to realize the same performance as obtained in single-cell testing. This was achieved using a 3M proprietary flow field design (discussed in Ref. 1), a unique modular compression control and gasket scheme, and computational fluid dynamic and thermal modeling designed gas and coolant flow manifolds. Bipolar plate development was not part of the scope, so gold-plated Al was used based on fabrication ease for small quantities, electrical conductivity, and corrosion resistance. Three 1.4-kW (nominal) designs (two air-cooled and one liquid-cooled) were fabricated and tested at 3M. One air-cooled design was targeted at reformat/air operation, with the other air-cooled design and the liquid-cooled design targeted for H₂/air. The liquid-cooled stack, "Beta-1," was then fully evaluated by the subcontractor at the General Motors Fuel Cell Activities facility under various protocols designed to demonstrate different aspects of the stack and MEA characteristics, including automotive test conditions. The stack contained fourteen 200-cm² MEAs made with the pilot-scale roll-good processes. The nanostructured Pt/PtAB CCMs had ~0.34 mg Pt/cm²/MEA total. Under automotive (vehicle simulation and compressor load following) test conditions, the stack achieved a mean cell voltage within 11 mV of the subcontractor's target of 680 mV at 0.8 A/cm² under pressurization conditions at 80°C and stoichiometric flow ratios of 3.0 anode and 1.6 cathode with sub-saturated anode and dry cathode reactant streams. The required anode and cathode pressure drops across the stack were demonstrated. Cell-to-cell uniformity was excellent (23 mV spread, 6 mV standard deviation @ 1 A/cm²). In other tests, fuel and air utilization was also shown to be very good: the stability at very low stoichiometric flow ratios greatly exceeded expectations, achieving 95% hydrogen utilization and 77% air utilization at ambient pressure. The vehicle polarization test

results exceeded design expectations, producing 1.9 kW peak actual with 62% air utilization versus the 1.4 kW with 45% air utilization expected. The stack was subsequently operated for 400 hours with no substantial change in performance. The values of 0.34 mg Pt/cm² and 1.9 kW represent 0.50 g Pt/peak kW, which meets the DOE 2005 target of 0.6 g Pt/peak kW. Results from a subset of the tests conducted at the subcontractor's facilities are shown in Figures 1-4.

Figure 1 compares the "Beta-1" stack output under ambient and pressurized constant flow conditions. The stack results match well with single-cell test results (not shown) using a 3M protocol. The up and down scans are both included, so 28 curves are superimposed for each pressure condition, illustrating the tight uniformity of the response, including hysteresis, of the 14 MEAs, which were

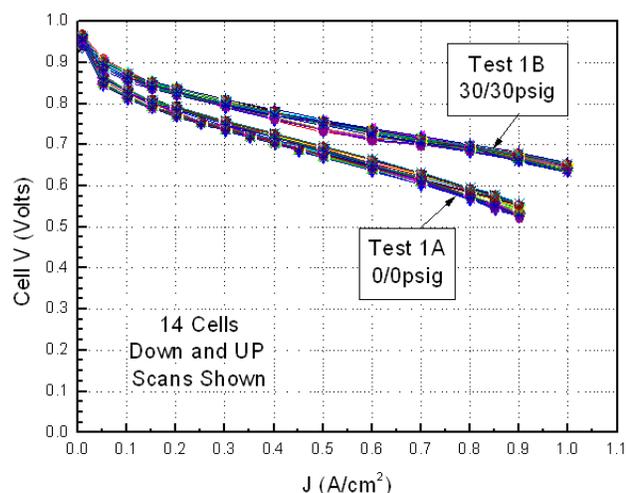


Figure 1. Comparison of ambient and pressurized operation of the 3M "Beta-1" stack at the subcontractor's facilities under a 3M test protocol for comparison with single-cell tests. The roll-good fabricated MEAs have 0.34 mg precious metal/cm²/MEA. Test 1A: 75°C stack, 70/70°C dew points, 25/72 standard liters per minute (SLM) H₂/air, galvanodynamic scanning, 1 min. per point. Test 1B: 85°C stack, 70/70°C dew points, 30/90 SLM H₂/air, galvanodynamic scanning, 1 min./point. Both up-scan and down-scan curves are shown for the 14 MEAs in both the ambient and pressurized conditions.

taken at 1-foot intervals along the CCM and MEA roll-goods.

Figure 2 shows the “Baseline High-Pressure Polarization Curve” under the subcontractor’s protocol. At 1 A/cm², the standard deviation from the average cell voltage among the 14 MEAs is only 6 mV.

In Figure 3, the stack response under the “Vehicle Polarization Test Conditions” is shown. The stack inlet pressures and dew points (proprietary) are varied with current density. The stack and MEAs yielded good cell-to-cell uniformity over a majority of the current density range. At 1.2 A/cm², the spread in voltage among the 14 cells was only 42 mV for a relatively low cathode stoichiometry of 1.6. The voltage spread appears to be largest, 49 mV, at 0.4 A/cm², but, excluding cells 1 and 2, it reduces to only 22 mV. At the maximum tested current density, a peak power of 1.9 kW is produced with 2.25 kW of heat.

Figure 4 compares the stack/MEA response under Test 7 (see Figure 3) with Test 3B, the subcontractor’s compressor-load following protocol.

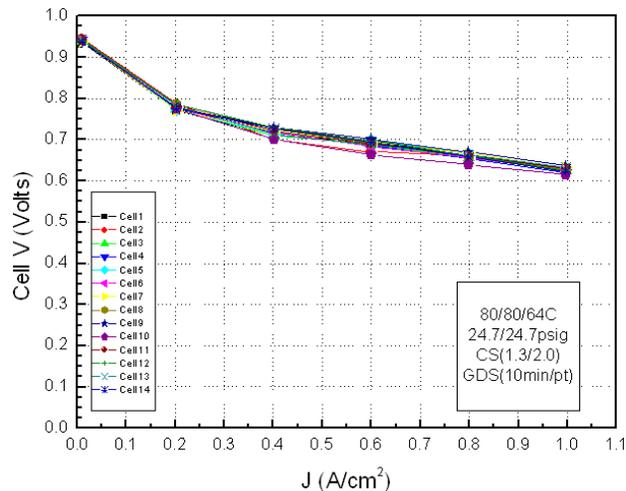


Figure 2. Test 4: "Baseline High-Pressure Polarization Curve," 80°C stack, 80/64°C anode/cathode dew points, 1.3/2.0 H₂/air constant stoichiometry (CS), 24.7/24.7 psig inlet pressure, galvanodynamic scanning (GDS), 10 min./point.

The pressure-load profile was the same, but the stack temperature, reactant humidification, and flow stoichiometries differed. Even though Test 3B had a higher cathode flow stoichiometry (2.0) than Test 7 (1.6), the humidification was somewhat greater than under Test 7, indicating that the MEAs and stack perform better under hotter and drier conditions.

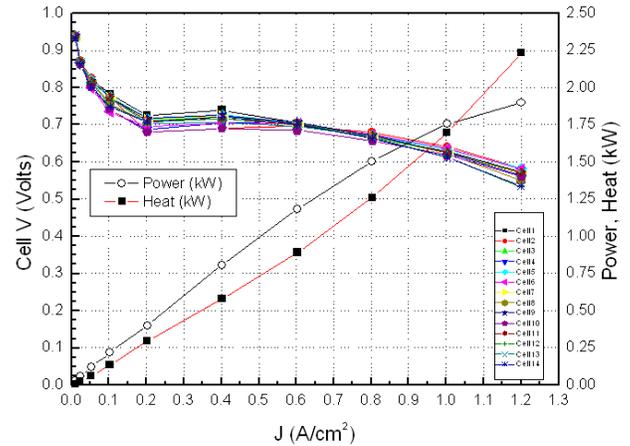


Figure 3. Test 7: "Vehicle Polarization Test Conditions," 61-84°C stack, variable (proprietary) dew points, 3.0/1.6 H₂/air stoichiometry, 2.9-24.7 psig inlet pressure, galvanodynamic scans, 10 min./point.

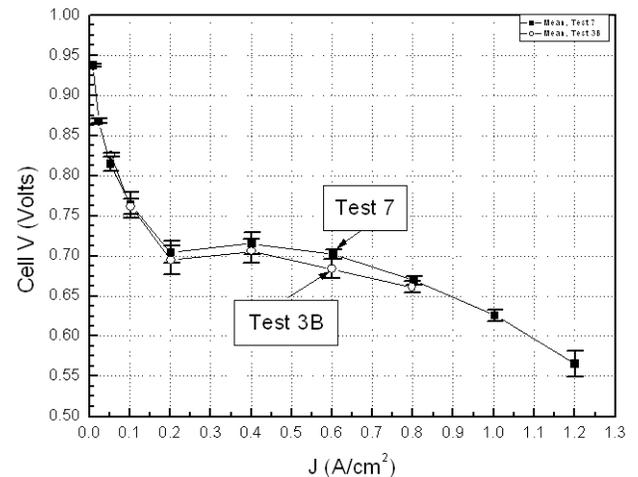


Figure 4. Comparison of the stack/MEA response under Test 7 (see Figure 3) and Test 3B, the subcontractor’s "Compressor-Load Following" protocol. Even though the cathode stoichiometry for Test 3B is higher than that of Test 7, the conditions are hotter and drier for Test 7, and this gives better performance in the mid-current range.

Conclusions

The development and roll-goods fabrication of a set of high-performance, matched PEM fuel cell components was completed and successfully demonstrated in stack testing at the subcontractor's facilities. The results under H₂/air correspond to 0.50 g Pt/peak kWe, which meets the DOE 2005 target of 0.6 g Pt/peak kW. The MEA components used in the stack testing were produced by roll-good, pilot-scale processes that have the potential for meeting projected high-volume automotive requirements.

References

1. Hydrogen, Fuel Cells, and Infrastructure Technologies FY 2002 Progress Report, page 379.