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RESEARCH AND DEVELOPMENT OF A PROTON- EXCHANGE-MEMBRANE (PEM) FUEL CELL SYSTEM FOR TRANSPORTATION APPLICATIONS

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PROGRESS REPORT FOR QUARTER 8 OF THE PHASE II EFFORT

8 NOVEMBER 1996

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EXECUTIVE SUMMARY

This eighth (8th) quarterly report summarizes activity from 1 July 1996 through 30 September 1996; the report is organized as usual into sections describing background information and work performed under the main WBS categories.

The **Fuel Processor** (WBS 1.0) team activity during this eighth quarter of Phase II focused on the move into the new facilities at Metro Park and preparation of the fuel processor, catalytic combustor, and related equipment for testing. The move into the new laboratory space was accomplished by the end of August. The small scale experiments bench and the reformer test stand have now been fabricated, the combustor stand and its supporting gas flow carts have been installed, and gas analysis equipment set-up in the new facility. By the end of this reporting period the test stands were about ninety percent completed. Testing of the fuel processor components (reformer and catalytic combustor) at the new facility is planned to be initiated by the end of October.

Initial testing of the new and innovative catalytic combustor has already been accomplished. A preliminary control strategy to operate the combustor under EPICS has also been completed.

In the meantime, individual components for the fuel processor have received considerable attention. The reformer was reassembled with a new fan motor housing; subsequently, the motor was tested to determine if it could achieve maximum speed. Maximum attainable speed is now believed to be limited only by the air gap between the motor's stator and rotor. This problem is currently being addressed. Additionally, new reformer methanol and water injectors have been designed, delivered, and tested. The test stand and alternative, autothermal reformer reactor, currently under construction at the GM R&D Center, are being modified to produce enough hydrogen to power a typical 30-kW fuel cell stack.

A rate expression was derived for the C18HC copper oxide/zinc oxide catalyst based on Caldwell Reactor test results from LANL. Using these results, a shift reactor volume of 11.7 lit. was calculated to be required if a 63% reduction in CO is desired.

After examining the test results from LANL it was believed that, under moderately high reactor throughputs, the PrOx reactor was not operating in an isothermal mode. The distance between the cooling tubes allowed some areas of the catalyzed fin material to get hotter than that considered to be optimum. The intent of the next bench-scale and full scale PrOx builds is to minimize the distance between the cooling medium and the reacting surface. Also different design concepts are being considered for air injection into the PrOx units.

The **Fuel Cell Stack** (WBS 2.0) team activity focused both on hardware analysis and testing to determine the appropriate approach for the GM and Ballard stacks and MEA preparation. DuPont continued to improve their MEA fuel cell performance in order to satisfy GM test conditions. While various software operating changes, adopting GM protocols, were incorporated into the DuPont test stand, and different carbon backing paper diffusion layers were used to make new DuPont large active area MEAs, the performance of these MEAs was still not adequate when compared to Delphi made MEAs using the same DuPont membrane. This problem now appears, however, to be resolved. The DuPont MEA has a very long (>9 hr.) hysteresis during start-up. A modification of Delphi's test procedures has resolved the majority of the performance disparity. DuPont is also investigating high performance novel catalyst coated composite membranes which appear to be very promising.

In the meantime, Delphi is investigating various low cost, high electrical conductivity carbon fiber mats for use as diffusion layer (backing paper) materials. Delphi is also investigating the average Pt particle size produced on alternative carbon supports in an effort to find the best carbon support by determining which of them produces the largest available Pt surface area. GM R&D is continuing to study cathode carbon supports for dispersing Pt catalyst by investigating the effects of (heat treatment, or lack thereof, on) average carbon support pore radius and slurry pH which control the thickness of the water film on the pore walls. This water thickness can impede

oxygen diffusion to the reaction sites. Delphi is also continuing to improve its large active area MEA production capability.

Delphi's test stands use automated test protocols and both large active area Gen 1 single and 3 cell short stacks have been tested over a range of operating conditions on both hydrogen/air and reformate/air. The performance of the 3 cell short stack was about 30 mV below that of the single cell performance; both tests were about 100 mV below the desired system target.

At Ballard, fabrication of components for the full scale 220 cell stack (50-kW) is underway. A Ballard test station is being modified and set-up in preparation for testing the full scale stack. Other Ballard activities include the fabrication of components for the air-membrane humidifier for Delphi's Gen 3 stack and close coordination with Delphi to identify various stack humidification issues and their impact on the overall system.

Two Delphi single cell test stands are now fully operational in the new Metro Park facility; these test stands are now fully automated. A third test stand for flow visualization is currently under construction. The 10-kW test stand has now been moved into the new facility and the flow systems are currently being connected to the test stand. A Delphi Gen 3 short stack will be tested on this stand in mid December. The 60-kW test stand was prepared for testing the Ballard Gen I and Gen II stacks. The Ballard Gen I stack was successfully tested during this reporting period and met or exceeded specified performance targets. Several problems with the test stand control were, however, discovered and are currently being resolved. Bipolar plate development is continuing with the emphasis on titanium plates and methods to produce a uniform TiN coating. Additionally, the plans and hardware implementation for the Gen 3 metal plate stack have been finalized with parts now beginning to be fabricated/delivered for initial build and test. Fuel cell model (electrode, gas composition, gas flow, and stack stress) development is continuing.

The **System Integration and Controls** (WBS 3.0) team focused on revising the ECE system specification and hardware build for operation at the high pressure, low current density conditions discussed last quarter. The revised hardware build includes a methanol/hydrogen catalytic burner and a make-up water condenser located upstream of the expander. The steady-state system model, FCSYS, was updated to reflect these changes and then used to both determine component requirements and predict system operating efficiencies.

Hardware was fabricated for the Aurora compressor and the Fisher Electric drive motor during this quarter. Most of the air management valves required for the system pallet were procured or ordered. Technical discussions with Allied Signal, in conjunction with system modeling, were used to investigate the feasibility of incorporating the Allied Signal turbocompressor into the ECE system and its subsequent impact on overall system efficiency. The Allied Signal turbocompressor appears viable if a controlled stack bypass flow modification is adapted.

The thermal management subsystem was further defined and component specifications were determined. The majority of the coolant system hardware required for the system pallet has been ordered or received.

The Delco Electronics controller (EECM) and modified Battery Pack Monitors (BPMs) were also delivered during this quarter. Integrated operation of the EECM, BPMs, sensors, and control actuators was verified on the bench in preparation for control of the system pallet. Testing and development activity continued on temperature, hydrogen, and CO concentration sensors.

Equipment was moved into the new systems laboratory facility at Metro Park. Installation of the Milli-Pore DI water system and the bi-polar power supply/load bank has been completed. Preparations are underway for the installation of H₂/CO₂/N₂ and air mass flow controllers to support operation of the Ballard Gen I stack on the system pallet.

The **Reference Powertrain Design and Commercialization Studies** (WBS 4.0) have not been initiated.

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Program Scheduling and Relative Progress for all of the tasks described are presented in Appendix A at the end of this report.

BACKGROUND

The overall objective for Phase II is the development of a stand-alone 30-kW (net power) Fuel Cell System running on methanol reformat. In addition, the successful operation of this fuel cell system will require it to be built within the constraints of automotive system needs and operational conditions. The program is structured to focus on those areas in most need of technological innovation.

SCOPE

The scope of the project is as follows:

- 1. Fuel Processor:** Includes burner development; reformer catalyst and heat exchanger development; temperature and pressure sensor development; shift reactor catalyst development; CO, CO₂, HC, H₂, and humidity sensor development; fuel injection hardware; PrOx catalyst and heat exchanger development; reformat cooling and humidification; fuel specification and tolerance issues; and model development.
- 2. Fuel Cell Stack:** Includes membrane development; electrode development; backing paper development; fuel cell test hardware; bipolar plate development; stack evaluation; model development; evaluation of vendor stack performance; and an internal design, build, and test program for at least a 50-kW (gross power), 30-kW (net power) GM stack.
- 3. ECE System Management:** Includes compressor/expander subsystem development; thermal management for the entire system; laboratory test fixture development and build; laboratory electronics and controls development; sensor development; system modeling; and system integration and performance testing.
- 4. Reference Powertrain Design and Commercialization Study:** Includes powertrain component and vehicle trade-off analysis and commercialization studies that consider environmental and regulatory issues, fuel cell transportation economics, and fuel cell assessment considerations.
- 5. Program Management and Reporting:** Includes the program management schedule and milestones, attendance at Government/Industry Technical meetings, and reporting as required by the DoE.

The results of the last three (3) months of activity that have been conducted at GM and on-site at the various industrial subcontractors are described in some detail in this eighth quarterly report.



WBS TASK 1. FUEL PROCESSOR DEVELOPMENT

SUMMARY

During the eighth quarter of Phase II activity in the Fuel Processor area focused on the move into the new facilities at Metro Park and preparation of the fuel processor, catalytic combustor, and related equipment for testing. The move into the new laboratory space was accomplished by the end of August. The small scale experiments bench and the reformer test stand have now been fabricated, the combustor stand and its supporting gas flow carts have been installed, and gas analysis equipment set-up in the new facility. By the end of this reporting period the test stands were about ninety percent completed. Testing of the fuel processor components (reformer and catalytic combustor) at the new facility is planned to be initiated by the end of October.

Initial testing of the new and innovative catalytic combustor has already been accomplished. A preliminary control strategy to operate the combustor under EPICS has also been completed.

In the meantime, individual components for the fuel processor have received considerable attention. The reformer was reassembled with a new fan motor housing; subsequently, the motor was tested to determine if it could achieve maximum speed. Maximum attainable speed is now believed to be limited only by the air gap between the motor's stator and rotor. This problem is currently being addressed. Additionally, new reformer methanol and water injectors have been designed, delivered, and tested. The test stand and alternative, autothermal reformer reactor, currently under construction at the GM R&D Center, are being modified to produce enough hydrogen to power a typical 30-kW fuel cell stack.

A rate expression was derived for the C18HC copper oxide/zinc oxide catalyst based on Caldwell Reactor test results from LANL. Using these results, a shift reactor volume of 11.7 lit. was calculated to be required if a 63% reduction in CO is desired.

After examining the test results from LANL it was believed that, under moderately high reactor throughputs, the PrOx reactor was not operating in an isothermal mode. The distance between the cooling tubes allowed some areas of the catalyzed fin material to get hotter than that considered to be optimum. The intent of the next bench-scale and full scale PrOx builds is to minimize the distance between the cooling medium and the reacting surface. Also different design concepts are being considered for air injection into the PrOx units.

TASK 1.1 COMBUSTOR DEVELOPMENT

The combustor's primary role in the electrochemical engine system is to provide heat for vaporization of water and methanol and the steam/methanol reforming endotherm. The final combustor will be capable of using methanol and/or hydrogen for fuel.

Task 1.1.1 Atmospheric Methanol Combustor Development

The tasks under this item are considered complete. See the progress report for Quarter 2, 1 April 1995.

Task 1.1.2 Methanol/Anode Exhaust Atmospheric Combustor Component Development

The tasks under this item are considered complete unless additional relevant knowledge is required. Refer to the progress report for Quarter 4, 10 November 1995. The information gathered from the device was used to develop the necessary understanding of both the component sensitivity and the EPICS control subsystem to properly operate the combustor within the overall electrochemical engine system.

Task 1.1.3 30-kW System Methanol/Anode Exhaust Combustor Development

All combustor development effort has been shifted from the baseline premixed, prevaporized combustor to a catalytic combustor.

Task 1.1.4 Alternate Combustor Concepts

This quarter's focus has involved understanding the data collected from the first catalytic combustion hardware as well as integrating revised system specifications, proposed 24 July 1996, in order to properly design a new catalytic combustor for the revised ECE system. Without having an accurate database for reaction rates of the input reactants, a versatile catalytic combustor configuration was designed. In order to meet the tight program timing some design preferences were superseded with available materials, e.g.: tubing, clamps, substrates, mixing media, electrically heated catalyst (EHC), tube vaporizer, etc. The initial catalytic combustor hardware utilizes a multi-flanged assembly held together using V-clamps. This assembly permits rapid change of the flame arrestor, mixing media, electrically heated catalyst section, catalyst substrate, catalyst volume, and liquid fuel vaporizer. The design, fabrication, and calibration of this innovative catalytic combustor and fuel system are now complete. The initial testing of this catalytic combustor, using an automotive TBI controller test rig, is complete. All testing was performed using only air and methanol. The unit operated over its designed turndown ratio with radial temperature profiles throughout the substrate varying by 30 degrees Celsius at minimum power (outer edge to core, end to end) to 300 degrees Celsius at maximum power. After its preliminary evaluation the catalytic combustor was considered applicable for integration into the EPICS controlled test stand; the catalytic combustor preliminary control strategy has been completed. As soon as the EPICS controlled catalytic combustor test program, using methanol and air, has been completed the catalytic combustor will be incorporated into the reformer. Stage two EPICS catalytic combustor/reformer development will also implement control of both hydrogen and carbon dioxide flow, with hydrogen combustion, to closely simulate actual tailgas system conditions. Catalytic combustor hardware that require further development before the combustor subsystem can be incorporated into the electrochemical engine system are the mixing media and the vaporizer. The goal of the mixing media is to mix the incoming gases so as to achieve a homogeneous mixture flowing into the catalyst bed. With a homogeneous mixture the catalyst reaction temperatures should be uniform, resulting in a very durable unit. The vaporizer, with assistance from Delphi Harrison, needs to be optimized to vaporize methanol fuel throughout the turndown ratio, but not allow the inner wall temperature to rise above 465 degrees Celsius during periods when no methanol is flowing through the vaporizer section and a hydrogen tailgas reaction only, outside of the vaporizer, is occurring.

In parallel to the combustor development, efforts were also concentrated on making the combustion lab operational. The combustion lab required some hardware and software development and builds on the air cart, H₂/CO₂ cart, and the combustor test stand. All lab to test stand electrical and plumbing connections are now near completion.

TASK 1.2 REFORMER DEVELOPMENT

The purpose of the reformer is to convert methanol and water into a hydrogen-rich gas stream that contains some small amount (<1%) of carbon monoxide. The water and methanol are fed into the reformer by modified fuel injectors. The heat for water/methanol vaporization and the reforming endotherm are provided by the combustor via a gas-to-gas heat exchanger.

The small scale experiments bench is complete; the components that were brought back from LANL were reinstalled at the new facility. The first set of experiments are planned for early November. These experiments will be used to verify some of the data previously collected on PrOx catalyst samples. Another set of experiments are planned to accurately quantify the performance of a small-scale isothermal PrOx.

A reformer test bench was also constructed during this quarter; this bench will permit the reformer and combustor to operate independently of the rest of the system.

Task 1.2.1 Reformer Development for the 10-kW System

As the 10-kW system remained at Los Alamos National Laboratory to provide LANL with system capabilities, no work was performed on this system during this quarter.

Search for More Active Reforming Catalysts. No activity occurred in this area during the eighth quarter.

Combustion Heated 10-kW System Reformer. Although the combustion heated, stirred 10-kW reformer has been relocated to the new laboratory facility, no work was performed on this device during this quarter.

Task 1.2.2 Steam Reformer Development for the 30-kW System

Combustion Heated 30-kW System Reformer. The 30-kW system reformer design has been reassembled with a new fan motor housing and the motor was tested to its maximum speed. Two pods that hold the automotive water and methanol injectors have been fabricated and installed and automotive pressure regulators have been affixed to these pods.

Injector design during this quarter focused on pencil stream spray development and improvements to withstand the corrosive effects of water, methanol, and temperature. Pencil stream spray optimization included a new single orifice director plate design utilizing a rounded, smoothed entry into the orifice. The pencil stream spray is desired because the fuel and water are to be injected onto the backside of the reformer fan for even distribution into the recirculating stream. Previous testing has documented that when a laminar fuel stream enters the spray orifice a uniform pencil spray, with minimal droplet "fliers" after the fuel exits the orifice, can be produced. Visual evaluation of the reformer injector spray was performed from 70 kPa to 200 kPa operating pressure. At low pressure, a uniform pencil stream was produced with no droplet fliers. As pressure was increased a pencil stream spray continued to be formed, but the number of droplet fliers around the stream began to increase. Further evaluation of the injectors will be performed, with the fan running to determine system sensitivity to spray, before they are installed in the reformer. With respect to corrosion, efforts were directed at areas of the injector most susceptible to corrosion and wear due to the decreased lubricity of deionized water. Focus was directed toward the guiding and bearing surfaces. This injector design was previously optimized for 100% methanol fuel exposure; further prevention to corrosion was obtained by substituting hastelloy directors and seats. An amorphous coating is currently being evaluated to reduce wear at the guiding and bearing surfaces. Injector durability testing is planned during the next quarter to evaluate the coating and finalize the design.

The reformer fan motor has been tested up to 6,500 RPM. It is believed that the extremely large (3 mm) air gap between the motor's stator and rotor is now the limiting factor. This motor, combined with a more conventional (0.5 mm) air gap, should permit the maximum 10,000 RPM design point to be obtained.

In order to better incorporate redesigns, modifications, and new hardware, a small, dedicated test bench for testing the fuel processor and developing control strategies is being fabricated. Having two test benches, one for system testing and a simpler one for fuel processor testing, permits parallel test efforts that better enable major milestone schedules to be met. The majority of the elements of the fuel processor test bench have now been received and the reformer test bench is ninety percent completed. All that remains to be done is to tie the I/O into the EPICS controller.

Some manual testing of the reformer, prior to the automated testing, can be accomplished. An injector driver box has been obtained from the Henrietta Engineering Center; this box provides precise, manual control of the water and methanol injectors. A heat transfer fluid heater/pump has been modified to provide up to 9-kW of heat transfer fluid at 280°C to the combustor heat exchanger. This will allow efforts to be focused on the reformer operation while providing a relatively safe source of heat until the new catalytic combustor is ready to be integrated into the fuel processor.

Task 1.2.3 Autothermal Reformer Development for a 30-kW System

As an alternative to steam reforming, an autothermal reformer which has the advantage of being able to generate hydrogen exothermically can be useful to promote rapid start-up time and fast transient response. Such a reformer could also result in a more compact and simple overall system. Various ECE system modeling studies indicate that, even though the autothermal reformer produces a lower H_2 concentration than a steam reformer (56% to 70+%, respectively), the excess H_2 not used by the stack to produce electricity can be utilized by a burner to supply energy to an expander, off-setting much of the compression work. In steam reforming, the excess H_2 from the fuel cell stack is burned to provide heat for the endothermic steam reforming process. As a consequence, the overall system efficiencies of the two reforming systems are quite similar. The initial GM autothermal reactor design, described in the 6th Quarterly Report, has been validated through tests conducted, with GM personnel, at Argonne National Laboratory; no "hot-spot" catalyst degradation was observed. The test stand and autothermal reactor, currently under construction at the GM R&D Center, are being modified to produce enough hydrogen to power a typical 30-kW fuel cell stack.

TASK 1.3 SHIFT REACTOR DEVELOPMENT

A rate expression was derived for the C18HC copper oxide/zinc oxide catalyst based on Caldwell Reactor test results from LANL. Using these results, a shift reactor volume of 11.7 lit. was calculated to be required if a 63% reduction in CO is desired. Finalization of the shift reactor design will include evaluation of catalysts C18HC and MDK20. Injector design issues are identical to those discussed in Task 1.2.2. Injector calibration and integration into the system will be finalized pending reformat analysis.

TASK 1.4 PROX DEVELOPMENT

The ProX is used to oxidize carbon monoxide left in the reformat after passing through the reformer and shift reactors. The ProX reactor must be able to oxidize CO preferentially to hydrogen and minimize the tendency to produce CO via the reverse shift reaction. The reformat exiting the ProX must contain less than 20 ppm CO.

ProX Design. Work was initiated during this eighth quarter to design a bench scale isothermal ProX based on knowledge gained from previous investigations. The previous ProX design utilized a tube-and-fin arrangement with the fins covered with catalyst and cooling fluid passing through the tubes. After examining the test results from LANL it was believed that, under moderately high reactor throughputs, the reactor was not operating in an isothermal mode. The distance between the end of the fin and the cooling tube allowed some areas of the catalyzed fin material to get hotter than that considered to be optimum. The intent of the next bench-scale and full scale ProX builds is to minimize the distance between the cooling medium and the reacting surface.

ProX Air Injector Design. Two design concepts are being considered for air injection to the ProX: a flat disk metering design, and a pintle design. Calibration development is underway and the design that best meets system requirements will be selected during the next quarter.

TASK 1.5 REFORMAT COOLING/HUMIDIFICATION

The thermal management portion of this report is described in the ECE System Integration and Controls Section.

TASK 1.6 INPUT/OUTPUT TEMPERATURE SENSOR DEVELOPMENT

Temperature sensor development is described in the ECE System Integration and Controls Section of this report.



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TASK 1.7 REFORMER OVER-PRESSURE SWITCH DEVELOPMENT

Over-pressure switch development is described in the ECE System Integration and Controls Section of this report.

TASK 1.8 DEVELOPMENT OF CO SENSING

CO Sensing is described in the ECE System Integration and Controls Section of this report.

TASK 1.9 FUEL PROCESSOR MODELING

Fuel processor modeling is included as part of the System Integration and Test Modeling effort and is described in the ECE System Integration and Controls portion of this report.

WBS TASK 2. FUEL CELL STACK DEVELOPMENT

SUMMARY

The focus of the work at DuPont continued to involve improvement of their MEA fuel cell performance in order to satisfy GM test conditions. While various software operating changes, adopting GM protocols, were incorporated into the DuPont test stand, and different carbon backing paper diffusion layers were used to make new DuPont large active area MEAs, the performance of these MEAs was still not adequate when compared to Delphi made MEAs using the same DuPont membrane. This problem now appears, however, to be resolved. The DuPont MEA has a very long (>9 hr.) hysteresis during start-up. A modification of Delphi's test procedures has resolved the majority of the performance disparity. DuPont is also investigating high performance novel catalyst coated composite membranes which appear to be very promising.

In the meantime, Delphi is investigating various low cost, high electrical conductivity carbon fiber mats for use as diffusion layer (backing paper) materials. Delphi is also investigating the average Pt particle size produced on alternative carbon supports in an effort to find the best carbon support by determining which of them produces the largest available Pt surface area. GM R&D is continuing to study cathode carbon supports for dispersing Pt catalyst by investigating the effects of (heat treatment, or lack thereof, on) average carbon support pore radius and slurry pH which control the thickness of the water film on the pore walls. This water thickness can impede oxygen diffusion to the reaction sites. Delphi is also continuing to improve its large active area MEA production capability.

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Two Delphi single cell test stands are now fully operational in the new Metro Park facility; these test stands are now fully automated. A third test stand for flow visualization is currently under construction. The 10-kW test stand has now been moved into the new facility and the flow systems are currently being connected to the test stand. A Delphi Gen 3 short stack will be tested on this stand in mid December. The 60-kW test stand was prepared for testing the Ballard Gen I and Gen II stacks. The Ballard Gen I stack was successfully tested during this reporting period and met or exceeded specified performance targets. Several problems with the test stand control were, however, discovered and are currently being resolved. Bipolar plate development is continuing with the emphasis on titanium plates and methods to produce a uniform TiN coating. Additionally, the plans and hardware implementation for the Gen 3 metal plate stack have been finalized with parts now beginning to be fabricated/delivered for initial build and test. Fuel cell model (electrode, gas composition, gas flow, and stack stress) development is continuing.

TASK 2.1 ACTIVE FUEL CELL COMPONENT DEVELOPMENT (M&E, BACKING PAPERS)

A carbon fiber backing paper material has been specified and ordered; this material has high conductive fibers which do not require an expensive graphitization process. This material should also be much lower in cost than current diffusion layer (backing paper) materials; however, it does not have the compressive strength of the current graphite backing layer papers. If the conductivity of the material is adequate it will be tested in single cells to determine if its lack of compressive strength will prohibit its use.

Two different thickness' of carbon fiber mat were specified for production. One of the mats has a thickness similar to graphite backing layer paper material currently in use, the other is a thicker version to make up for the general lack of mat structural strength compared to graphite backing paper materials. These mat materials should be delivered during the next quarter and evaluation tests will be initiated upon their receipt.

Task 2.1.1 Dow Membrane Development Contract

The subcontract with Dow was definitized and completed during the last reporting period.

Task 2.1.2 DuPont Membrane Development Contract

Development of Advanced Fuel Cell Membranes. The major focus of the work at DuPont during this reporting period was on optimizing the fuel cell performance of DuPont MEAs in order to satisfy GM test conditions. The latest discussions between the Delphi and DuPont teams indicated the necessity to use anode and cathode carbon backing (diffusion layer) papers less than 8 mils thick as opposed to the 11.5 mil thick paper that was normally being used by DuPont. Additional experiments are being conducted toward this redirection. Preliminary results using the thinner carbon backing paper indicate that the performance of the MEA was not very sensitive to the thickness of the carbon backing paper. In the recent past Delphi software, containing protocols required for continuous operation of the test station, was installed. After extensive debugging the software is now operational and continuous 8 hour tests are now conducted on a routine basis. Also, the latest operating conditions, as obtained from Delphi, are now being adopted to test all MEAs used for this program. Additional experiments were also conducted using Delphi treated carbon backing papers with a DuPont CCM (see below) and the results were compared with those obtained using DuPont carbon backing (diffusion layer) papers. The performance results were still not very different. Efforts to optimize large active area standard Nafion made DuPont MEAs are continuing; however, some resolution of the problem appears to have been discovered. Hysteresis observed during start-up of the DuPont MEA can be very long (> 9 hr.). A modification of Delphi's test procedure has now produced similar performance between DuPont and Delphi made large active area MEAs, refer to Task 2.1.4.

Membrane efforts were also focused towards developing better performing CCMs/MEAs based on other structures. Additional experiments towards this goal resulted in samples with fuel cell performances very similar to those depicted in the previous quarterly report. The reproducibility in preparing the samples on a lab scale appear to be very good. Steady-state performances of 0.65 to 0.7 V were achieved at 1.0 A/cm² on a 50 cm² sample using the original GM system specification conditions.

Development of Membrane Coating Technology. Novel catalyst coated membranes (CCMs) have been prepared which possess improved toughness and increased dimensional stability. These CCMs have also performed extremely well in the 50 cm² single cell test stand over a range of temperatures, pressures, and stoichiometries. A more complete characterization of these CCMs is in progress; this study will include optimization of process and property parameters.

Task 2.1.3 Electrocatalyst/Membrane Assembly Development at Delphi and GM R&D

Delphi Catalyst Studies. Pt catalysts supported on different supports (Printex and Ketjen Black) were obtained and analyzed for Pt particle size. The average particle sizes for Pt, Pt/Sn, and Pt/Ru on XC-72 and Pt supported on alternative supports (Printex and Ketjen) are presented in Table 1.

TABLE. 1

Catalyst	Average Pt particle size (angstroms)
20% Pt / XC-72	34
20% Pt/Sn / XC-72	43
20% Pt/Ru / XC-72	26
20% Pt / Printex	293
20% Pt / Ketjen Black	290

The platinum supported on Printex and Ketjen Black demonstrate large particle sizes, nearly an order of magnitude larger than the Pt supported on XC-72. These large particles sizes may reduce the available surface area of the Pt on a weight basis compared to that supported on XC-72. Hydrogen adsorption/desorption measurements will be made on the catalysts to determine what effect the particle size has on the noble metal catalyst surface area. The particle size measurements for the first three samples were conducted by XRD at GM R&D. These XRD measurements were then verified at Delphi, using a Delphi XRD, while Delphi personnel were becoming familiar with this analytical technique; after the initial measurements and verification, the catalyst samples with the alternative supports were analyzed at Delphi.

Carbon Support Studies for the Air Electrode at GM R&D. The effect of heat treatment of eight carbon supports (for dispersing Pt catalyst) on air electrode performance was studied. The carbon supports had an average pore radius in the range 1 - 15 nm and a slurry pH in the range 2.3 - 9.3. Heat treatment increased the pH of the carbon to 10.0 ± 0.1 , irrespective of how acidic the carbon was in the as-received form. This may be attributed to the removal of all carboxylic groups on the carbon surface due to the heat treatment. It is not known if this chemical condition will remain stable in a fuel cell environment or even after exposure to ambient air. Heat treatment had only a minor effect on the BET surface area and pore distribution. In general, a small (< 10 %) decrease in the BET surface area occurred due to heat treatment.

Acetylene Black, Ketjen Black and Vulcan XC-72R carbons demonstrated the highest fuel cell performances at various operating conditions (various temperatures, pressures, and use of both air/oxygen as the cathode reactant) both in the as-received and heat-treated conditions. Heat treatment did not have a beneficial effect on the performance of carbons that were alkaline in their pre-treated conditions. Heat treatment of Raven-5000 and Black Pearls led to a large increase in fuel cell performance, though the enhanced performance is still not of practical interest. Air electrode results at lower cell voltages demonstrated that the cell performance was determined by diffusion of gaseous oxygen in the electrode pores and the rate of diffusion of dissolved oxygen in the water films covering the pore walls. As average pore radius and surface pH decrease, the thickness of the water film on the pore walls increase, thus impeding the transport of gaseous oxygen to the reaction sites.

Task 2.1.4 Develop Phase II MEA

General. MEA development efforts focused on applying applications determined during interim operation at the Henrietta facility while getting processing equipment on-line at the new Metro Park facility. New processing equipment includes: an upgraded screen printer, new press fixtures, and improvements in chemical treatment. The final production improvements, prior to beginning construction of MEAs for the first full scale stack build, were implemented. With the existing design and processing techniques MEA production for the stack build will require about 26 working days or less, depending on the final MEA design decisions being made over the next 3 months.

Testing. Many tests were conducted on the fuel cell test stands using the Gen 1 hardware. These tests include many synthetic reformat tests conducted under the new (September, 1996) "system specification" operating conditions. These tests indicate that at the high power load point (0.6 A/cm^2) Delphi-made MEAs produce about 630 mV on hydrogen and 600 mV on synthetic reformat with 18 ppm CO and 2% air injection. These tests were repeated with one cell over a one month period with no significant change in performance. Three-cell short stacks using Gen 1 hardware were also run for about one month with no significant change in performance. This indicates that the short-term life of the Gen 1 components seem to be adequate. More details of these tests are presented in Tasks 2.5.1 & 2.5.2.

Other Vendor MEA Testing. In the previous quarterly report it was stated that Gen 1 size MEAs made by DuPont and tested at Delphi were not developing performances even close to that measured at DuPont on 50 cm^2 hardware. The reason for this anomaly has now been discovered. The hysteresis observed while operating DuPont made MEAs can be very long (> 9 hours). A modification in the test procedure to eliminate initial test points was required so that the hysteresis effect is eliminated. With this modification similar fuel cell performance has now been observed on both the DuPont made Gen 1 size MEAs and their 50 cm^2 size MEAs.

Task 2.1.5 Ballard Stack Development Contract

Introduction. Ballard's statement of work for this project consists of two subtasks. Subtask 1 involves the maintenance and support of the MK5E fuel cell stacks that were previously delivered during Phase One of this project. Subtask 2 consists of supply, support, and maintenance of Advanced Power PEM stacks. Ballard delivered two 30-kW Improved Stacks (Gen I) to the program in November, 1995 and is to deliver a 50-kW Advanced Stack (Gen II) during late November to early December, 1996. The following is a summary of Ballard's stack development progress during the period from 1 July to 30 September 1996.

Phase One MK5E Stack Support. Support availability for the MK5E stacks continued during this period. The stacks were utilized for test stand commissioning at Delphi with minimal need for direct support from Ballard.

Advanced Power Ballard Gen I and Gen II PEM Stacks. During this period the Ballard Gen I stack, previously delivered to the Delphi Engineering Center, was successfully commissioned and operated in a Delphi test stand. Ballard personnel were in attendance to assist the unit start-up and to provide operational training. The stack performed well, meeting or exceeding all design goals. An extensive matrix of parametric tests was conducted by Ballard, at Delphi's request, to assist the program in defining precise interface specifications. The tests utilized Ballard Gen II hardware and explored operation at a variety of pressures and temperatures. Based on Ballard's test results, Delphi's subsequent system analysis indicates that the optimum stack operating conditions at maximum power should be:

Current density	0.6 A/cm^2
Fuel and air pressure	26 psig
Temperature	80°C
Air stoichiometry	2.0
Fuel stoichiometry	1.4.

As Delphi's system analysis indicates that the optimum stack maximum current density should be 0.6 A/cm^2 , the number of cells in the full scale fuel cell stack has been increased to 220 from an original count of about 185. Fabrication of components for the full scale 220 cell stack is underway. Some of the stack subassemblies are going through final development testing. Quality control testing of fabricated components is being conducted through short stack testing prior to integration into the full scale unit.

A Ballard test station is being modified and set-up in preparation for testing of the full scale 220 cell stack. This requires building test fixtures, modifying some controls, and expanding data acquisition capability, particularly

cell voltage monitoring channels. Other Ballard activities include initiating fabrication of components for the air-membrane humidifier and working with Delphi to identify and assess the various stack humidification issues and their impact on the overall system. This is a part of an ongoing dialog to communicate system interface specifications to Delphi.

In summary, Ballard is on track to deliver the completed 220 cell Gen II stack to the program during the late November to early December time frame.

TASK 2.2 FUEL CELL TEST HARDWARE DEVELOPMENT

The Delphi test stand used for single cell durability testing of materials and MEAs is now operational and has been operated for 2 eight hour checkout periods; most of the test stand is operating as required. A few items require some additional work including: computer acquired temperature measurements, water feed to the test stand, and connection to the hydrogen vent. Durability testing of samples will be initiated as soon as the additional work is completed. In other test hardware development a corrosion cell is now being used for corrosion measurements of potential metal bipolar plate materials, and a Hydrogen Adsorption/Desorption (HAD) test facility for electrocatalyst evaluation has also become operational.

Task 2.2.1 Single Cell Test Stands

Hardware. Two fuel cell test stands are fully operational in area C-136 of the new Metro Park facility; the large active area multiple cell test stand has the ability to monitor both cell voltage and 1 kHz cell resistance of each cell in a large active area, multi-cell stack. The ability to measure the 1 kHz cell resistance of a single cell is still limited to current densities less than about 0.6 A/cm^2 . Air cooling of the recirculation water, via heat exchangers, is now in place and working on the Big Pete test stand. A 3 cell large active area stack requires active cooling of the recirculation water when the current density is above 0.5 A/cm^2 . A similar feature is also still required on the Small Pete test stand. Synthetic reformat bottles have been plumbed to both Pete test stands in area C-136. An air exhaust relative humidity sensor has been installed on the Small Pete test stand. This change permits increased accuracy in the water balance measurements. A similar feature is still required to be installed on the Big Pete test stand.

A 4-bottle hydrogen manifold has been ordered for the hydrogen cabinet in C-136. This will permit longer duration testing without bottle replacement and will also make bottle replacement much easier while allowing for complete hydrogen consumption of each bottle.

The third fuel cell test stand under construction in the Metro Park facility is continuing and will initially be used to supply humidified air to the flow visualization fixture.

Software. The HP VEETEST control software has been updated so that up to 5 cell stacks can be monitored; the software is capable of gas stoichiometric control based on the number of cells, data logging up to 5 cell's voltage and resistance, and test shutdown occurring on any one low cell voltage. Data analysis software (in EXCEL & HP VEETEST) has also been written to analyze the data from the additional cells. The test protocol software has been updated to allow constant stoichiometric control of synthetic reformat. Constant stoichiometric polarization curves have been automated to allow the control of synthetic reformat flow throughout the range of the polarization curve (0.1 to 0.9 A/cm^2). These test sequences explore the dependence of cell performance on cathode air stoichiometry, RH, and pressure based on the new (September, 1996) "system specification" operating conditions. Unattended overnight operation of these tests greatly increases test throughput. All of these software updates have also been supplied to DuPont.

Task 2.2.2 10-kW Fuel Cell Stack Test Stand

The 10-kW test stand built at LANL has been moved into its new lab at the Metro Park facility; utilities and gas supplies have been connected, and the steam humidification system is being installed. A Delphi Gen 3 short stack will be installed and tested on this test stand in mid-December.

Task 2.2.3 Develop Advanced Insitu Diagnostics

The Mathcad data analysis program used to compile data taken from the single cell test stands was improved so that averaged values of the flow characteristics could be determined at each of the protocol test operating conditions. By averaging values of: pressure drop, absolute pressure, flow rate, relative humidity, discharge coefficients, density and viscosity; the amount of water build-up, channel bypassing, and flow stability can be qualitatively analyzed. Statistical analysis of the data also permits further insight into the flow field characteristics. The bulk of the analysis concentrated on the cathode flow field due to the fact that understanding water management is vital to determining optimal cell operating conditions. A contour plot of pressure drop as a function of relative humidity and flow rate can be produced by performing a multivariate regression on these three parameters. The data consistently demonstrated that if the overall flow rate is held constant the pressure drop increases as the humidity increases. Moreover, the increase in pressure drop with relative humidity is accentuated by the production of water associated with an operating fuel cell. This phenomenon suggests that water droplets are forming, obstructing the channels, and thus increasing the pressure drop.

Two other methods also suggest that water drops are forming in the channels and increasing the flow resistance. The first method involves quantifying the standard deviation in the pressure drop measurements during a given protocol test condition operating point. At each test condition over 100 data points are measured, from these measurements the standard deviation can then be computed. When this is done, the data consistently indicated that the pressure standard deviation increases with relative humidity, again suggesting that water drop activity increases with humidity. The second method of quantifying water droplet action involves determination of the overall flow field discharge coefficient (Cd). The discharge coefficient is computed by dividing the product of the flow rate and viscosity by the pressure drop. Therefore, as defined, Cd is a measure of how geometry affects the flow characteristics. In theory, Cd should remain constant under all flow conditions, provided that the geometry does not change. However, water droplets can obstruct a channel, thus changing the geometry. The data in this area is less conclusive in terms of exactly determining the method by which water activity impacts Cd. One reason for this is that channel bypassing also changes the flow field geometry and cannot be easily separated from the data. Work continues to progress in this area as more data is accumulated on the test stands.

More controlled experiments focusing on the effects of water activity and flow field geometry on pressure drop are planned for the Gen 3 hardware on the new flow visualization stand. On this stand humidified air will be fed into a flow field and the formation and motion of water drops will be quantified. In addition, specific water activity will be tied to deviations in pressure drop on a real-time basis. Effort in this area should commence by the end of October.

In addition, a study in the effect of fuel cell flow field materials and surface treatments was begun, in part, to determine the amount of force required to move a water drop through the flow field. This experimental work involves measuring the contact angle and contact radius between a deionized water drop of known volume and the solid media on which it rests. These measurements are then repeated for drop volumes ranging from 0.1 to 1.0 μl . The slope of the cosine of the contact angle versus the inverse drop contact radius provides a relative measure of the drop's line tension. In addition, contact angle is a direct measure of the relative hydrophobicity of the solid in question, whereby, an increase in contact angle translates into an increase in hydrophobicity. The theoretical limit is a 180° contact angle which occurs when the drop is touching the surface of the solid at one point. Work in this area is still in progress; however, early conclusions suggest that grinding has a limited effect on increasing a solid's hydrophobicity. Conversely, non oil-based surface treatments to a graphite flow field did improve its relative hydrophobicity.

Task 2.2.4 60-kW Fuel Cell Stack Test Stand

The main efforts related to stack testing during the last quarter were in preparing the 60-kW test stand for the Ballard Gen I and Gen II stacks. Ballard personnel participated in two days of testing for the start-up of their Gen I 30-kW stack at Henrietta. The stack performed well, meeting or exceeding the performance recorded at Ballard's facilities on both hydrogen and simulated reformat. These first tests of a high power stack uncovered several problems with the test stand that were not evident at the 5-kW power levels used to initially debug the stand. The most serious of these problems involved reformat gas control and software induced test stand shutdowns. In late September it was decided to cease testing of the 30-kW stack in order to correct all test stand problems in time for the arrival of Ballard's Gen II stack.

TASK 2.3 BIPOLAR PLATE DEVELOPMENT

Samples of titanium plates nitrided via a gas phase nitriding process utilizing ammonia have been received at Delphi (Flint). Two samples were gas phase nitrided under varying conditions. Both samples demonstrated excellent conductivity for use as bipolar plates in a fuel cell; conductivity testing indicated an overall material resistivity of about $0.03 \Omega\text{-cm}$, which is excellent. Post analysis (via XRD) of the nitriding has shown that both the γ -phase (Ti_2N) and Δ -phase (TiN) were observed on the Ti surface. Both samples show the golden TiN color observed when a PVD process is used indicating the sample should have good conductivity. Only about a 4 mV loss would be expected for operation at 1 amp/cm^2 . Since the substrate material is titanium, corrosion of the samples should not be an issue.

Titanium plates have also been nitrided using a standard production gas phase nitriding operation, but these samples do not have a good uniform coating. Gas phase nitriding of these titanium plates resulted in a surface which does not have sufficient conductivity. XRD analysis of gas phase nitriding of these titanium plates indicates that there does not appear to be any TiN formation. New samples were ordered to be nitrided at a higher temperature (800°C) in a special fluidized bed specifically developed to produce higher operating temperatures. These new samples, however, also do not appear to have formed a surface nitride, similar to the samples produced at lower temperature. Conductivity testing indicates a surface oxide formation as the samples are still not conductive. It is suspected that water or oxygen contamination is preventing nitriding from occurring and operation under very strict conditions are required. These operating conditions are currently being identified and will be forwarded to the nitriding operation to see if the nitriding reactor can meet the specified conditions.

Gen 1 titanium cooling plates have been PVD TiN coated and are being used in the single cell fixture. Currently, one TiN coated cooling plate has over 100 hours of operation without any noticeable increase in material resistance.

Considerable time has also been spent on brazing the anode, cathode, and cooling plates; Delphi-Harrison has been involved in this procedure. Test coupons have been brazed with good results and are demonstrating good strength, flowability, and cleanliness (no heavy oxides). This has been accomplished with low cost Microbraz 51. These tests have given the current vendor confidence to commit to meet the time schedule. A fear of warpage during processing and furnace time still remains. Full scale prototypes will determine if these procedures produce warpage.

TASK 2.4 DELPHI STACK DEVELOPMENT

Task 2.4.1 Develop Stack Design

Early in this quarter the many material problems plaguing the Gen 1 hardware, described below, have finally been resolved. The nearly one year effort required to identify and solve these problems (labeled as "Gen 1 hardware debug" in the milestone charts) has lead to a restructure of the general hardware debugging effort. The restructured effort now allows for the "off-line" and parallel testing of the many components in a complex fuel

cell design. It permits the fuel cell test stands to be used primarily for testing fuel cells rather than identifying material problems. These improved procedures have permitted the identification of many problems within the Gen 2 hardware, also described below, without having to test a complete stack in a test stand. For example, these results provided the data required to conclude that material problems with Kynar graphite would not allow cell voltage measurements to be made with an accuracy better than 100 mV. As a result of this restructured effort the Gen 3 hardware, again described below, has entered into the debug phase earlier than expected. This has potentially saved months of program time.

Gen 1 Design. Several approaches were pursued simultaneously to resolve the persistent problem with leakage at the bushing seal. The first two approaches reduced the mechanical loads on the bushing. Steel rule dies were fabricated to eliminate the intentional overlap of the seal and the edge of the backing paper layer and a spacer was designed to eliminate the shear load in the bushing flange. The third approach addressed the basic material strength. Based on data produced by the Delphi Materials Lab, which indicated that the molded bushing material was still amorphous, samples were oven treated to achieve a fully crystalline state. Tests in a running cell and in an intensified off-line fixture demonstrated that the problem has been solved.

Minor changes have been made to increase the throughput in the testing area. One example reduced the hardware warm-up delay on the Gen 1 short stack test through modifications to the insulating sheets. Many holes were required in the end plates for supply, exhaust, and diagnostic ports for the reactant gases and heat transfer. To avoid flexing the plates the structure behind them must have good parallelism and high compressive stiffness. Insulating sheets common in the injection molding industry were initially used, but allowed too much heat loss. A simple geometric pattern was developed which greatly reduced the conductive heat losses while maintaining adequate stiffness and parallelism.

Gen 2 Design. The Gen 2 hardware configuration designed for use without a separate flow field insert was assembled with a nonfunctional MEA for check-out purposes. Leak testing at temperature verified that the molded graphite material used for the plates was sufficiently impermeable. The test also verified that the test fixture could deliver adequate heat to the hardware. In order to maintain the overall project timing, Gen 3 items have been treated as a higher priority than Gen 2 development whenever the two competed for resources. Improvements in the formed plate delivery have caused the activities to overlap to an extent that initial samples of Gen 3 hardware can be pulled ahead of the Gen 2 configuration with flow field inserts. The priorities have now been adjusted to make Gen 2 hardware a follow-up activity primarily for the development of Insitu Diagnostics and Gas Flow Models (Tasks 2.2.3 and 2.6.3, respectively).

Gen 3 Design. During this reporting period, the Gen 3 stack development activities focused on procuring the special tools and initial samples of each component. Titanium sheet stock was obtained for the anode plates, cathode plates, and centers which will be brazed together to form the bipolar plates. The centers have been fabricated using CNC punching. The tooling to etch and final form the anode and cathode plates are nearly completed. A supplier was selected for the molded rubber seals. Design modifications for molding considerations were incorporated without impacting the mating parts already in process.

Task 2.4.4 Assemble Stack-Gen 3, 50-kW Bipolar Plates

Material to produce a titanium stack has been requisitioned as have the appropriate processing, forming, and coating procedures and facilities. The plate material and coatings for the bipolar plates should not inhibit production of the stack. The Gen 3 stack build requires a conductive coating on the titanium plates; the currently planned procedure utilizes a PVD (physical vapor deposition) TiN (titanium nitride) coating. Quotations for the TiN coating on the Ti bipolar plates for the entire stack have been received. It appears that turn-around for all 250 pieces for a PVD TiN coating should require 3 weeks.

Three current possible coatings to maintain surface conductivity of the bipolar plates are being considered:

PVD TiN Coating. PVD TiN coating for items of this size is expensive and will, most likely, never attain cost targets regardless of the production volume. Vendors have been specified and the coating procedure has been proven by durability testing in a single cell test stand. A 3-week delivery time has been quoted for the entire stack (250 pieces). Since the plates will become available in batches, the batches will be delivered to be coated as they become available. This should decrease the overall time between reception of the final brazed assemblies and final coating. Unless a better solution occurs this is the coating which will be used for the Gen 3 stack build.

Gas Phase Nitriding. This is the cheapest alternative coating technique and the closest to a production process. Throughput should be fast compared with the PVD process. This process has been proven on a laboratory scale in a quartz furnace. Scale-up for production of the bipolar plates for the 50-kW stack build at this point appears to be difficult. Scale-up for production of bipolar plates in the long term appears more feasible. Contact with other Delphi personnel, who are working on the same scale-up process for a different project, also indicates that short term, low volume production may be difficult. However, if a production gas phase titanium nitriding operation is developed it could be used for this phase of the project. The scale-up problem apparently arises from water and oxygen impurities which lead to a titanium oxide formation; these impurities must be purged from any nitriding system to below 1 ppm. Commercial gas phase nitriding systems typically do not have this capability as it is not required for the nitriding of steels. The laboratory type equipment has very good control of impurity levels so gas phase nitriding of titanium was easily accomplished. The gas phase nitriding in commercial equipment does not have the ability to keep oxygen and water concentrations at low levels, thus, the gas phase nitriding operation in the commercial equipment ends up oxidizing the titanium rather than forming a nitride coating. Vendors are currently being sought that can accomplish low impurity gas phase titanium nitriding; it is also possible the gas phase nitriding could be accomplished using Delphi's laboratory equipment if sufficient time and money were to be invested in the proper equipment.

Platinum Coating. This coating would only be used as a last resort if absolutely required. Two different Pt coating processes and vendors have been identified.

TASK 2.5 STACK EVALUATION

Tasks 2.5.1 & 2.5.2 Test Protocols/Automated Stack Tests

Automated test protocols have been used throughout the test program for collecting the data. This allows for a very representative evaluation of the cell performance without consuming many hours of operator time. For example, in a typical protocol the cell conditions are allowed to stabilize for 20 minutes **before** data is collected for performance evaluation at those conditions. The operator then can, at one time, generate a sequence of test points at which to evaluate the cell performance. When the automated test sequence is initiated the test stand can operate unattended for more than 24 hours.

Typical results from such an automated test protocol, run with a single cell Gen 1 size Delphi-made MEA operating on synthetic reformat, is presented in Figure 1. The system specification target performance is also presented as X's. The protocol establishes two load points (0.4 and 0.8 A/cm²) at the system specified operating conditions; furthermore, the protocol evaluates the effect of different cathode pressures, stoichiometries, and relative humidities. Automated constant stoichiometry synthetic reformat polarization curves from 0.1 to 0.9 A/cm² have also been run on this MEA. As depicted in Figure 2, the data result in nearly identical measured performance levels. A least squares fit (LSF) line is calculated for all of the data points. The 0.6 A/cm² intercept is 593 mV for the LSF polarization curve data and 601 mV for the steady-state performance protocol of Figure 1.

A 3-cell short stack was assembled and tested. Constant stoichiometry polarization curves were run on this stack from 0.1 to 0.8 A/cm². A typical result from such an experiment is presented in Figure 3. The average cell voltage is plotted and then a least squares fit line is calculated to those points. The 0.6 A/cm² intercept is about 30 mV lower than the single cell Delphi made MEA described above. Reduction in overpotentials required to drive low current density points may explain the general upturn of the curve below current levels less than 0.2

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A/cm². Differences in the deviation from one cell to the next may be a result of differences in reactant gas flow distribution from one cell to another, MEA to MEA variations, thermal non-uniformities from cell to cell, or some other explanation. In any case these types of analyses greatly contribute to understanding the effects that influence the performance of fuel cell stacks.

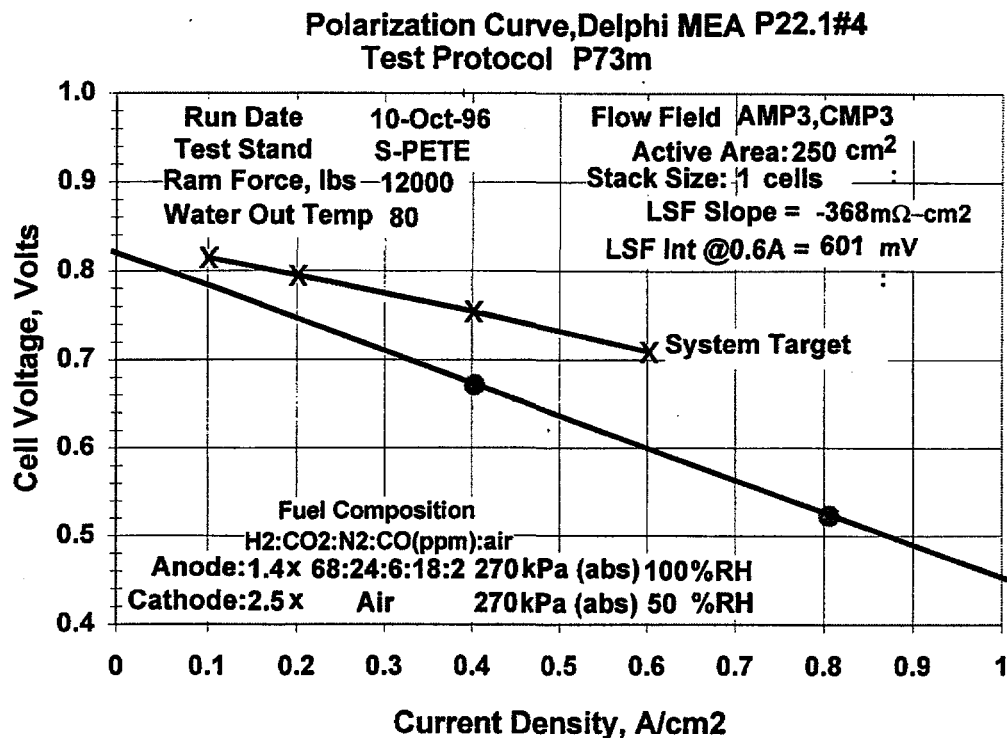


Fig. 1

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AND IS IN ACCORDANCE WITH PUBLIC LAW 100-679"

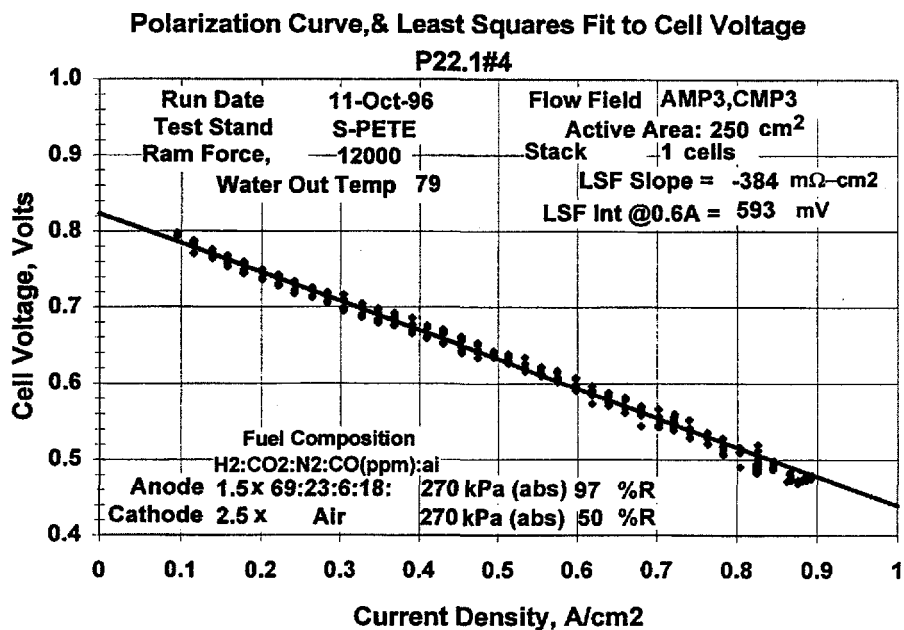


Fig. 2

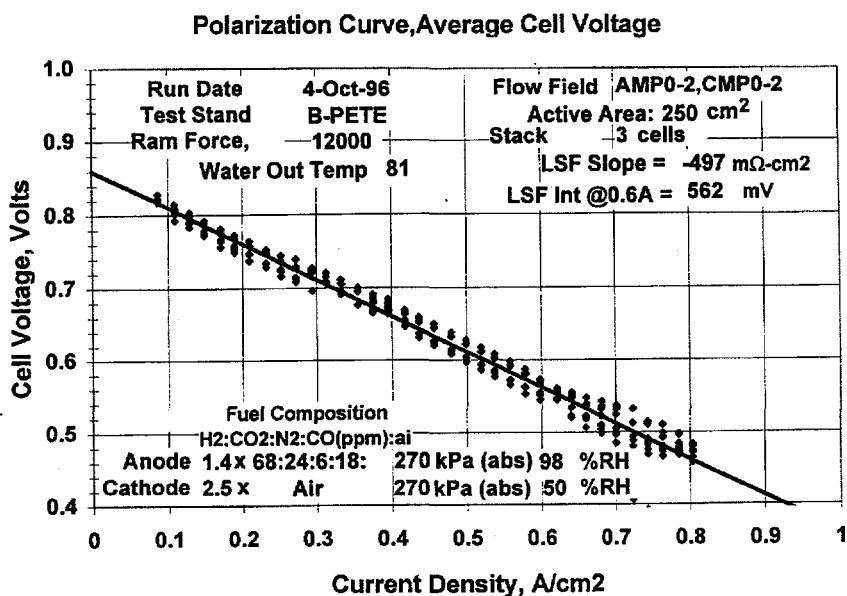


Fig. 3

TASK 2.6 FUEL CELL MODEL DEVELOPMENT

Task 2.6.1 Develop 'Down-the-Channel' Model of Mass and Energy Transport Within MEAs

Professor Trung Nguyen of the University of Kansas has delivered a 'Down-the-Channel' code for the modeling of mass transport and energy transport processes within the channels of fuel cell bipolar plates and MEAs. Professor Trung Nguyen visited Delphi-E in Rochester NY to present his final report entitled 'Development of a PEM Fuel Cell Simulator as a Design Tool'. The model incorporates heat and mass transfer in a 'Down-the-Channel' manner of operation to model the processes within the fuel cell bipolar plate channels and MEAs. He has delivered a completed code for the model which is being used by Delphi personnel. Further revisions will be conducted at the University of Kansas, and Professor Trung has indicated that the updated codes will be delivered to Delphi at no additional cost. Delivery of the final model code and report to Delphi fulfills the contract with Professor Trung Nguyen and the University of Kansas.

Task 2.6.2 Develop Gas Composition Model

Computational fluid dynamics (CFD) modeling efforts concentrated on determining the impact of flow field geometry on parameters such as gas bypassing, pressure drop, and transport of gas to and from the reaction layer. The main emphasis of the modeling was centered on the cathode flow field where pressure drop is critical to establishing proper water transport. The detail of this type of model was much more complex than previous efforts and required development of the CFD code itself. In particular, a reaction zone must be added to the porous cells representing the paper backing layer, and an algorithm which accounts for the change in porosity as a function of liquid water content had to be written. The models of the two flow field geometries studied also required a more refined mesh to adequately discretise the flow domain. At the present time work in conjunction with CFD code developers is continuing in order to implement the enhancements in the code necessary to properly model the flow field.

Anode Overpotential Calculations in the Existing 'Through-the-Electrode' Model. Overpotential calculations include separate calculations of the cathode and anode overpotentials. Cathode overpotentials to date have been calculated as a simple means to compare with model calculations and single cell data. Anode overpotentials have not been included thus far in the existing 'Through-the-Electrode' model, thus, calculations to compare CO₂ dilution effects and CO poisoning effects have not been available. To explain operational results on simulated reformat, anode overpotential calculations are required.

The anode overpotential is calculated by the Tafel equation:

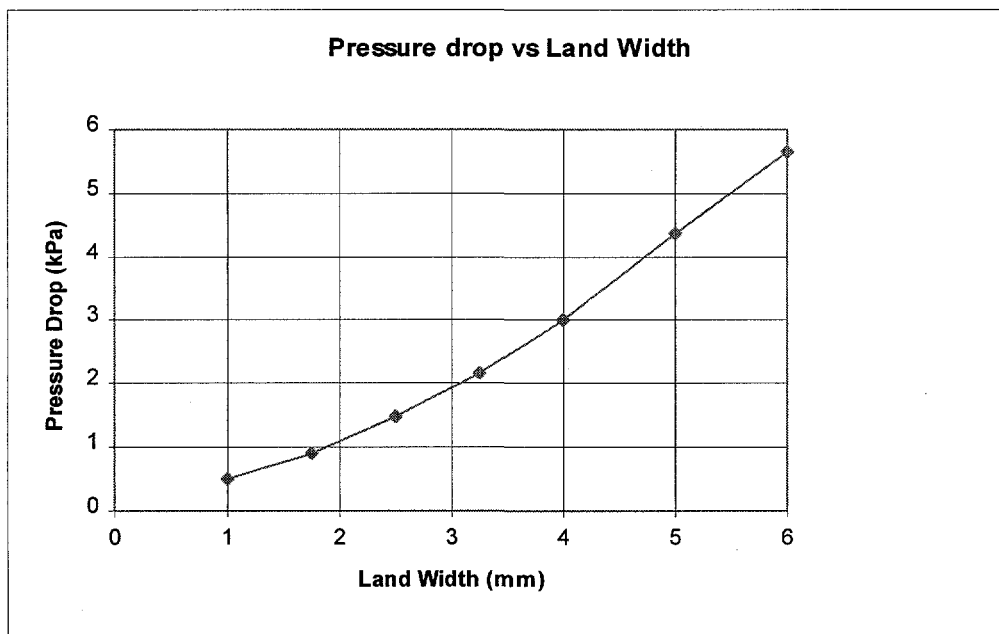
$$\eta = RT/\alpha nF * \ln[i/i_o]$$

where, i_o = the exchange current density, and α = the transfer coefficient = (0.5).

The anode overpotential changes from operation with pure H₂ to operation with 75% H₂/25% CO₂. Single cell testing indicates between a 25 and 40 mV drop in performance when operation is changed from pure H₂ to 75% H₂/25% CO₂ at 0.4 amps/cm²; however, at 0.8 amps/cm² the performance drop is between 0 and 60 mV. The calculated value at 0.4 amps/cm² is about 20 mV (depending upon the exact operating conditions) while the calculated value at 0.8 amps/cm² is about 40 mV, both of which match the data well. The single cell results and matching analysis suggest that CO₂ does not present a poisoning problem in the cell fixtures and the losses observed are due to the dilution of the hydrogen. An important caveat to this observation is the fact that 2% air is being injected into the anode whenever synthetic reformat is being used as the anode reactant. Previous work at Los Alamos indicated that the presence of this air injection is primarily responsible for eliminating CO₂ poisoning effects.

Task 2.6.3 Develop Gas Flow Model

A CFD study was also conducted on the feasibility of implementing an interdigitized flow field. An interdigitated flow field has channels which do not provide a continuous path between the inlet supply and outlet return headers. Instead, the flow of gas is forced through the backing paper layer and over the lands, which, in theory, increases the rate of gas delivery to the catalyst layer. In addition, such a flow field would be less complex and, thus, less expensive to manufacture. For the study, simple straight channels were used and the width of both the land and groove were varied to determine their impact on pressure drop. Results of the modeling indicated that, when normalized for flow, the width of the groove had no impact on the rate of flow, and, hence, the pressure drop over the land. However, the linear length of the groove did directly impact the amount of pressure drop over the land. Varying the land width also had a significant impact on the pressure drop across the flow field. The attached graph illustrates the effect of pressure drop as a function of land width. In this study the overall gas flow rate and active area were held constant as the land width varied from 1 mm to 6 mm. The nearly quadratic shape of the relationship is largely due to the fact that, as the land width increases the number of grooves decreases, and each groove is then required to carry more gas flow which increases the pressure drop.



Task 2.6.5 Develop Stack Stress Models

No activity was performed on this task during this reporting period.

WBS TASK 3. ECE SYSTEM INTEGRATION AND CONTROLS

SUMMARY

The ECE system specification and hardware build have been revised for operation at the high pressure, low current density conditions discussed last quarter. The revised hardware build includes a methanol/hydrogen catalytic burner and a make-up water condenser located upstream of the expander. The steady-state system model, FCSYS, was updated to reflect these changes and then used to both determine component requirements and predict system operating efficiencies.

Hardware was fabricated for the Aurora compressor and the Fisher Electric drive motor. Most of the air management valves required for the system pallet were procured or ordered. Technical discussions with Allied Signal, in conjunction with system modeling, were used to investigate the feasibility of incorporating the Allied Signal turbocompressor into the ECE system and its subsequent impact on overall system efficiency. The Allied Signal turbocompressor appears viable if a controlled stack bypass flow modification is adapted.

The thermal management subsystem was further defined and component specifications were also determined during this quarter. The majority of the coolant system hardware required for the system pallet has been ordered or received.

The Delco Electronics controller (EECM) and modified Battery Pack Monitors (BPMs) were also delivered during this quarter. Integrated operation of the EECM, BPMs, sensors, and control actuators was verified on the bench in preparation for control of the system pallet. Testing and development activity continued on temperature, hydrogen, and CO concentration sensors.

Equipment was moved into the new systems laboratory facility at Metro Park. Installation of the Milli-Pore DI water system and the bi-polar power supply/load bank has been completed. Preparations are underway for the installation of $H_2/CO_2/N_2$ and air mass flow controllers to support operation of the Ballard Gen I stack on the system pallet.

TASK 3.1 AIR MANAGEMENT

Tasks 3.1.1 & 3.1.2 Compressor/Expander Development

Aurora Compressor and Drive Motor. Fabrication of component parts and assembly of the Aurora compressor occurred during this quarter. Also, a small test stand fixture was fabricated for mounting and coupling the compressor to the Fisher Electric drive motor. Initial testing of the compressor is planned to start in early October at Aurora Technologies. Delivery of the Fisher drive motor and speed controller is expected by mid-October. Aurora will mount the compressor and drive motor onto the test fixture and perform the necessary alignment upon receipt of the drive motor. The assembly will then be shipped to Delphi-E for final testing, and integration into the system pallet.

Delivery of the Aurora compressor/drive motor was delayed by one month (Sept. 20th to Oct. 22nd) due to a change in system operating voltage. Reducing the maximum operating current density from 1000 mA/cm^2 to 600 mA/cm^2 raised the fuel cell output voltage by more than 50 volts; this voltage change caused a redesign of the compressor drive motor and electronics. Since the system airflow requirements were also reduced from the original high current density specification, the maximum compressor speed was reduced from 10,000 RPM to 8,000 RPM. The combination of increased operating voltage and reduced speed is expected to provide an improvement in drive motor operating efficiency.

PRDA Compressor/Expander. Delphi-E and Delphi-H personnel attended a Design Review meeting with each of the PRDA contractors in late July. Several coordinated conference calls with Allied Signal followed in

August. Discussions centered around the operational characteristics of the Allied Signal turbocompressor and its characteristic pressure drop-off during turndown. Allied Signal supplied projected data for operating efficiencies, output pressures, and flow rates. The data was input into the FCSYS model to analyze ECE performance and water management issues. Details of this work are discussed in TASK 3.5.

In summary, the Allied Signal air compressor/expander machinery is compact and highly efficient, but there are some significant water management (humidification and condensing) issues associated with low pressure operation during turndown. One identified solution is to operate this machine in a bypass mode where a greater than required airflow is compressed to maintain the desired pressure, but only the required airflow is humidified and directed to the fuel cell stack. The remaining airflow is bypassed to the expander for partial work recovery. Allied Signal will provide the bypass flow circuit and flow control valve as part of their deliverable to this program. Hardware delivery is projected for April 30, 1997.

Flow and Pressure Control Components. Many of the flow control valves required for air management in the ECE system were specified, procured, or ordered during this quarter. The approach was to find available, off-the-shelf hardware that could provide the required system functions, but would also represent first generation components. This approach supports development of the initial system pallet while, at the same time, providing experience to help specify improved requirements for second generation hardware that will be more compact and automotive oriented.

Hardware that was procured or ordered this quarter includes anode and cathode bypass and shutoff valves, air compressor inlet and outlet filters, an electronic backpressure control valve (required until an expander becomes available), a burner exhaust bypass valve for the reformer, and a burner inlet air control valve. Engineering discussions were initiated with valve manufacturers with respect to second generation hardware concepts.

TASK 3.2 THERMAL MANAGEMENT

The thermal management subsystem was defined and component specifications were nearly completed during this quarter. The majority of the coolant system hardware required for the system pallet has been ordered or received.

Tasks 3.2.1 & 3.2.2 Condensing Heat Exchanger/Heat Rejection

Delphi-H delivered a liquid-to-liquid heat exchanger that will reject heat from the stack coolant loop to the laboratory process water cooling loop. Other manual flow control valves, a coolant pressure regulator, and a process water circulation pump were procured for the cooling subsystem. The makeup water condenser will be specified and procured during the next quarter.

Coolant Flow Control Valves. Two prototype coolant flow control valves were delivered by Delphi-H this quarter. One of these valves will control the stack operating temperature through control of the coolant flow to the heat rejection radiator.

Task 3.2.3 Anode/Cathode Humidifiers/Coolers

The gases exiting the fuel processor and compressor need to be cooled prior to entering the fuel cell anode and the cathode membrane humidifier, respectively. Projected heat loads were calculated for the full power condition and cooler locations in the stack coolant loop were determined. Request for Quotations were issued by Delphi-H for both coolers.

TASK 3.3 CONTROLS, DATA ACQUISITION, DIAGNOSTICS, AND SAFETY

Tasks 3.3.1 & 3.3.2 Laboratory Controls/Sensors

Carbon Monoxide/Hydrocarbon/Carbon Dioxide Sensor. This sensor was characterized and reprogrammed using the Motorola emulation board. A calibration process that is more representative of actual reformat gas conditions (high temperature, flow, and pressure) is under development.

Low Concentration Hydrogen Sensor. Six low concentration hydrogen sensors were performance evaluated on a test fixture at GM R&D. Output voltage of the sensor was found to be both proportional to the hydrogen flow rate and nearly independent of gas temperature and pressure. Based on these results it was concluded that the sensor would be well suited for detecting hydrogen crossover or leakage in the cathode exhaust. Five sensors were shipped to Delphi-E.

An electrochemical model was developed to interpret the voltage response of the non-equilibrium, mixed potential sensor. The model successfully predicts the dependence of sensor voltage on hydrogen concentration, oxygen concentration, and gas flow rate.

High Concentration Hydrogen Sensor. Two of six prototype sensors fabricated last quarter were received from GM R&D. A previously developed electronic conditioning module will be added at Delphi-E. The sensor will then be characterized and calibrated as an assembly.

Temperature Sensor. Two high temperature sensors (20 - 900°C) for measuring burner exhaust temperature were developed and delivered by Delphi-P. The new design is lower cost, more robust, and better suited for high volume production.

Tasks 3.3.3 & 3.3.4 Develop 30-kW Vehicle Control System/Power Electronics

System Hardware Build. The hardware build for the 30-kW deliverable was revised based on the system modeling, heat exchanger design analysis, and catalytic burner development reported last quarter. The system specifications and hardware assembly are now designed for a system operating at a 600 mA/cm² maximum current density that employs a catalytic rather than a flame type burner. The system condenser has been located just upstream of the expander inlet and condenses water in the combined burner and cathode exhaust streams. The reformat gas cooler and compressor air cooler are both cooled by the stack cooling water loop.

Controls. One Electrochemical Engine Control Module (EECM) and two modified Battery Pack Monitors (BPMs) were received from Delco Electronics during this quarter. With the support of Delco Electronics personnel the control system was assembled at Rochester and tested with each major type of input sensor and output actuator. All elements of the control system, including sensor readings, actuator outputs, and communication networks (Class II and IEEE488), were verified to be properly operating.

Delco Electronics is in the process of updating the names of control variables and calibration constants that will be used in the software associated with the revised system hardware assembly. Delco Electronics has also started work on the serial instrumentation package and calibration development system that will provide a means to monitor and calibrate the control variables in the ECE system.

TASK 3.5 30-KW SYSTEM INTEGRATION AND TESTING

FCSYS (Steady State Model). FCSYS was applied to analyze numerous aspects of the system component specifications including the cathode and anode coolers, the cathode humidifier, and the air compressor. The model was also used to estimate overall system efficiencies for the recently revised system specification. A number of modifications were made to the code to make it more representative of the system deliverable. Modifications included: (1) replacement of the pre-mix flame burner with a catalytic burner utilizing cathode exhaust for its oxidant supply; (2) a polarization curve based on Ballard Gen II short stack data (with cell voltage

computed as a function of current density, cathode stoichiometry, and stack pressure and temperature); (3) condenser placement before the expander; and (4) stack and system pressure drops as a function of various flow parameters. With the exception of accurate compressor and expander efficiencies the code should adequately model the efficiency of the 30-kW deliverable.

Computed efficiency versus net electric power is presented in Figure 3.1. The "baseline" case assumed constant compressor and expander efficiencies of 65% and 60%, respectively. Overall system efficiency for this modeled case increases from 31.4% at full power (30-kW) to over 37% at mid-power (10-15-kW). As net power is further reduced to 6-kW, the efficiency decreases to 35.5% due to the increased impact of fixed parasitic losses, such as sensor, actuator, and pumping loads.

During this quarter Allied Signal also supplied estimates of their centrifugal turbocompressor performance. This compressor/expander combination appears to develop very high operating efficiencies within a compact package. However, a key characteristic of this type of dynamic air compressor is its strong supply pressure dependence upon flow rate (pressure decreases during turndown). As indicated in last quarter's modeling results, system water issues (humidification and, thus, make-up condensation requirements) are adversely effected by decreased system pressure. FCSYS was used to investigate the feasibility of incorporating the Allied Signal turbocompressor in the ECE system and its subsequent impact on overall system efficiency. Calculated system efficiencies, incorporating the Allied Signal estimated turbocompressor efficiencies (operated as described below), are compared to the baseline case in Figure 3.1.

The Allied Signal turbocompressor was designed for a high current density system and is, therefore, over-designed for the current Delphi-E design specification (76 g/s air flow vs. approximately 50 g/s at full power, respectively). This turbocompressor produces a discharge pressure of only approximately 220 kPa when operated at 50 g/s. At this operating condition FCSYS predicted a very high condenser make-up water requirement, 7.9 g/s, combined with a fairly low condenser outlet temperature of 60°C, and a condenser heat load of 26-kW. For comparison, the baseline case requires 4.1 g/s of make-up water, the condenser outlet temperature is 70°C, and the condenser heat load is 16-kW. The combination of a large heat load at a relatively small delta-T (a 38°C ambient condition design case is assumed) would result in an unacceptably large condenser design. Alternately, the compressor could be operated at the rated 76 g/s flow rate to maintain the baseline operating pressure. However, condenser requirements are still undesirably large (6.6 g/s make-up water, 64°C condenser outlet temperature, and a condenser heat load of 23-kW) compared to the baseline case and the increased air flow would still necessitate a larger cathode humidifier. One solution appears to be to use the Allied Signal turbocompressor in a "bypass" mode where a greater than required air flow is compressed to deliver the desired pressure; however, only the required amount of air flow is humidified and directed to the stack while the remainder is bypassed to the expander for partial work recovery. For the current oversized Allied Signal turbocompressor approximately 20 g/s would have to be bypassed at full power; this does, however, result in a nearly identical condenser design compared to the baseline case.

The bypass criteria for turndown was to limit the minimum condenser outlet temperature to 60°C. Based on this criteria the percentage of flow bypassed increases as net power decreases and system efficiency is compromised. Curve A/S-76, depicted in Figure 3.1, indicates that system efficiency is approximately 1% better than the baseline case at full power (due to higher air compressor/expander efficiencies) but is considerably less efficient below 20-kW net power.

A redesign of the turbocompressor to match the 50 g/s specification was considered. This case would not require bypass at full power but would require limited bypass during turndown. Such a redesign would increase system efficiency by about two percentage points at full power and up to one percentage point down to 20-kW. This curve is presented as A/S-50 in Figure 3.1. While the predicted increase in efficiency compared to the high flow, high bypass operating condition (A/S-76) is significant, it was decided to continue with the existing design due to scheduling penalties and the anticipation that future ECE systems will require greater net power and, therefore, greater airflow.

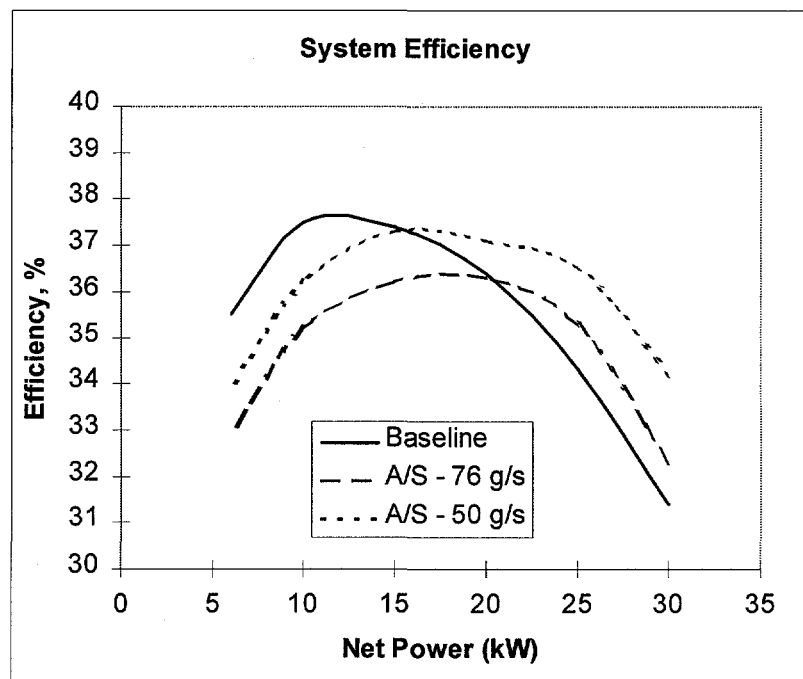


Figure 3.1 Modeled System Efficiency for the Allied Signal Compressor/Expander vs. Baseline Case

ECETRAN (Transient Model). The transient model accuracy has been determined to be very dependent on system reaction rates. Data from experiments in the Caldwell Reactor performed while at Los Alamos were re-analyzed for both the methanol reforming and water-gas shift reactions. Reaction rates as a function of temperature and pressure were determined for the two catalysts most likely to be used in the reformer and shift reactors:

$$k(\text{reforming, catalyst "A"}) = 3.3 \times 10^3 \exp(-7160/T) / P^{0.68} \text{ m}^3/\text{kg-s}$$

$$k(\text{reforming, catalyst "D"}) = 9.6 \times 10^4 \exp(-8380/T) / P^{0.82} \text{ m}^3/\text{kg-s}$$

$$k(\text{shift, catalyst "A"}) = 1.1 \times 10^6 \exp(-6830/T) / P^{2.3} \text{ m}^6/\text{kgmole-kg-s}$$

$$k(\text{shift, catalyst "D"}) = 2.4 \times 10^6 \exp(-7230/T) / P^{1.7} \text{ m}^6/\text{kgmole-kg-s}$$

The new reaction rate coefficients were applied to the transient reformer model. Initial analysis indicates that the reformer should be capable of providing a reformat stream with greater than 99% methanol conversion. Further validation of the behavior of the model, as it applies to steady-state results, will rely on empirical reformer data.

PID controls are being incorporated into the reformer model. The actual response of the system will be highly dependent on the catalytic burner transient behavior (as yet unknown and still to be modeled); however, preliminary model results are encouraging, indicating that the reformer should be robust in transient response (i.e., high methanol conversion). Further, the model also appears to exhibit a great deal of numerical stability, indicating that the reformer predictions are quite accurate. It is also expected that the model will be useful as a "test-bed" for preliminary control system design and calibration.

The transient shift reactor model is being utilized in the shift reactor design. This model can be used both for sizing and for the investigation of optimal water injection as a function of methanol feed rate. Water injection

plays a twofold role; it lowers the catalyst bed temperature and increases the water concentration, both of which enhance CO conversion to CO₂ via the water-gas shift reaction equilibrium. However, the lower catalyst bed temperature also results in reduced reaction rates, particularly that of the methanol conversion reaction. The optimal water injection rate is tied to the temperature change, reaction kinetics, and reactor residence time (itself a function of reactor volume and flow rate, i.e., turndown ratio). The model provides an easy way to consolidate these effects and identify the unique optimal water injection rate for given input conditions.

Transient performance predictions and control studies involving the PrOx are continuing. An extensive study was performed to determine a correlation between available measurements and the CO concentration exiting the PrOx. Possible candidates were the exiting oxygen concentration, the axial location of the "hot spot" along the reactor, and the conversion of H₂ across the PrOx. The latter was determined to provide the best indirect measure of outlet CO concentration. A simple PI (proportional-integral) controller has been implemented in a PrOx model which incorporates sensor delay. With a sampling period of 100 ms, and relatively low controller gains, the settling time of the system in response to disturbances or setpoint changes is predicted to be on the order of ten seconds.



WBS TASK 4. REFERENCE POWERTRAIN DESIGN AND COMMERCIALIZATION

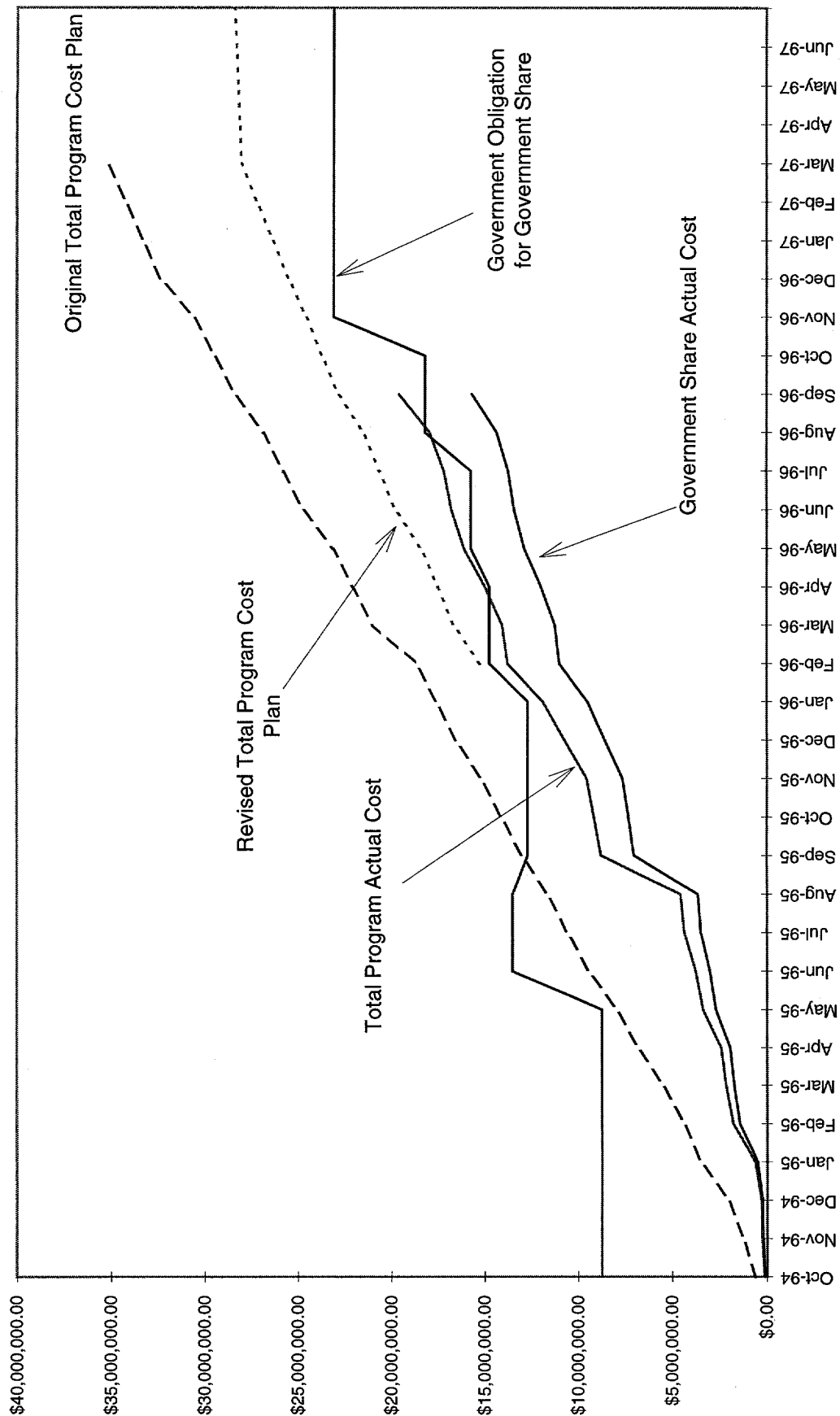
No activity on this task was performed during this quarter.

WBS TASK 5. PROGRAM MANAGEMENT AND REPORTING

The GM/DoE Quarterly Review was held at Rochester, NY on July 19th. The program will be extended by four months to the end of July, 1997 to enable Delphi Energy to complete PEM stack development and testing efforts. The Delphi Energy facility at Metro Park became fully operational during this quarter. The Program Manager attended the DoE Fuel Cell Review at Morgantown, WV on August 20th, the DoE National Lab Review at Washington, DC on September 25th, and also served as a member of the evaluation panel reviewing National Lab activities. As a member of the PNGV Fuel Cell Tech Team, the Program Manager participated in the monthly fuel cell meetings, gave a presentation on membrane/electrode manufacturing issues at the Fuel Cell Manufacturing Workshop held on July 23rd, and contributed to the planning of the PNGV peer review meeting to be held in October.

FIG. 5-1 Total Program Cost Plan Graph

Cost Plan Revised August 15, 1996
DE-AC02-90CH10435



General Motors, US Department of Energy

[illegible]

Electromechanical Engineering Project

General Motors, US Department of Energy

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Appendix A Milestone Schedule

Proprietary ECE Project Information

Project Plan as of March 15, 1996

Electrochemical Engine Project
General Motors, US Department of Energy

ID	WBS #	Task Name	1995												1996												1997																			
			S	O	N	D	J	F	M	A	M	J	J	A	S	O	N	D	J	F	M	A	M	J	J	A	S	O	N	D	J	F	M	A												
42	2.5.1	Write test protocols																																												
43	2.5.2	Automate stack tests																																												
44	2.6	Fuel Cell Model Development																																												
45	2.6.1	Develop electrode model																																												
46	2.6.2	Develop gas composition model																																												
47		intentional blank																																												
48	2.6.4	Develop stack thermal management model																																												
49	2.6.5	Develop stack stress models (seals)																																												
50	2.7	Facility Modifications																																												
51		intentional blank																																												
52		intentional blank																																												
53	2.7.3	Modification of Current Test Stands																																												
54																																														

General Motors, US Department of Energy

ID	WBS #	Task Name	S	O	N	D	J	F	M	A	M	J	J	A	S	O	N	D	J	F	M	A	M	J	J	A	S	O	N	D	J	F	M	A
55	3	ECE System Management																																
56	3.1	Air Management																																
57	3.1.1	Compressor and expander development																																
58	3.1.2	Flow and pressure control components																																
59	3.2	Thermal Management																																
60	3.2.1	Condensing heat exchanger																																
61	3.2.2	Heat rejection																																
62	3.2.3	Anode/cathode humidifiers/coolers																																
63	3.3	Controls, Data Acquisition, Diagnostics and Safety																																
64	3.3.1	Laboratory Controls																																
65	3.3.2	Sensors																																
66	3.3.3	Develop 30 kW vehical control system																																
67	3.5	Misc. Systems / Control Activities																																
68		30-kW System Integration and Testing																																
69																																		

General Motors, US Department of Energy

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