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

U.S. Department of Energy Contract No.
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PROGRESS REPORT FOR QUARTER 4 OF THE PHASE II EFFORT

20 OCTOBER 1995

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EXECUTIVE SUMMARY

This 4th quarter report summarizes activity from July 1, 1995 through October 1, 1995; 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 quarter focused on the continued design/development of the full scale fuel processing hardware. The design has been completed and model shop hardware is expected during the next quarter. Development work will be initiated as soon as this hardware is delivered. The combustor test stand has been completed allowing more detailed testing of the various parts of the combustor sub-system; this subsystem is currently being evaluated using the dual fuel (methanol/hydrogen) option to gain a better understanding of the control issues. Control of the proper air/fuel ratio is especially critical when the combustor fuel is switched from 100% methanol to a combination of the two fuels. Additional effort was placed on evaluating an isothermal PROX supplied from Harrison; data collection is not complete at the time of this writing.

The **Fuel Cell Stack** (WBS 2.0) team activity focused on material analysis and testing to determine the appropriate approach for the first GM stack. Five hundred hours of durability was achieved on a single cell fixture using coated titanium plates (anode and cathode) with no appreciable voltage degradation of the SEL (Stack Engineering Lab) produced MEA. Additionally, the voltage level drop across each of the plates remained low (<5mv) over the full test period. The first 340 cm² active area MEA was constructed using the modified PHI press, a major achievement in scale-up from the previous 50 cm² hardware. The MEA appears to be a very solid design and a full scale test program will be initiated with JDC made MEAs. A hydrogen adsorption/desorption test apparatus was also designed and constructed and is now operational. This piece of test equipment allows measurements of active platinum surface area, yielding the effective platinum catalyst utilization of MEAs. Additionally, computational fluid dynamic modeling of the flow in the anode and cathode has been initiated. This modeling technique should help shorten MEA/flowfield development time.

The **System Integration and Controls** (WBS 3.0) team focused on the initial layout and configuration of the 30-kW system. The purpose was to insure that the system can be packaged in a manner that will allow easy access to all of the critical components. The system test stand is nearing completion and will be ready for debug early in 1996. Appropriate power absorption has been procured from AeroVironment. Prototype componentry continues to arrive for the first system build and those parts are being integrated into the 10-kW mule for initial testing. A remaining critical system issue is the procurement of appropriate compressor/expander hardware; two vendors have now been selected for this prototype hardware, VAIREX and ADL. Contract proposals have been submitted by both of them to Harrison.

The **Reference Powertrain Design and Commercialization Studies** (WBS 4.0) have not been started and are currently under review

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 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, 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 30-kW GM/LANL 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 the JDC and on-site at the various industrial subcontractors are described in some detail in this fourth quarter report.

WBS TASK 1. FUEL PROCESSOR DEVELOPMENT

SUMMARY

During the fourth quarter of Phase II, activity in the Fuel Processor area focused on: designing the reformer for the 30-kW system, building the combustor test stand, testing the combustor for the 10-kW system, and building the combustor for the 30-kW system. An isothermal version of the Preferential Oxidation reactor (PrOx) was fabricated and tested in the 10-kW system.

The reformer design for the 30-kW system is a combination stirred and isothermal plug flow reactor. This design allows the inclusion of benefits of a stirred reactor as well as the benefits of an isothermal reactor. To date, the estimated mass of the reformer is 60 kg. The estimated volume of the reformer is 60 l. The expected delivery date of the reformer will occur during the next quarter.

Efforts in the combustor area included integration of control and testing of the atmospheric, dual fuel combustor to be used in the 10-kW reformer. In addition, the development combustor for the 30-kW system was designed and built.

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

Experiments on the 10-kW combustor were initiated. The primary focus of these experiments has been the development of the control algorithms for the combustor. The first steps in the control algorithm development were calibration of the methanol injector and nozzle and the stepper motor control valves for the combustion and dilution air flows. The primary EPICS control functions were then implemented for the methanol and air flows and the ignition system. Once these control functions were implemented the first tests of the combustor were conducted. The ignition sequence and start-up conditions were identified and the flow limits for flame stability were measured. The power range of the combustor was measured to be from 2.6 kW to 15.5 kW, a 6 to 1 turndown ratio. Following these experiments, the combustor was coupled to the heat exchanger of the Harrison-modified 10-kW reformer. Since the heat exchanger changes the back pressure on the combustor, and could possibly change its performance, the ignition conditions and stable power range of the combustor were reconfirmed.

Continued experimentation with the combustor/heat exchanger focused on the development of the data acquisition and control system for transient control and measurement of the combustor operating conditions. A higher level of flow control was implemented to allow a single power level input to control both the methanol and combustor air flows at stable conditions. Tests of flame detectors were conducted to determine their optimal configuration. Control of the diluted exhaust temperature was implemented with an open-loop control based on the methanol and combustor air inlet flows. Closed-loop control of the exhaust temperature proved difficult because the exhaust is not completely mixed at the outlet of the combustor. Control system development is being

continued to further automate the combustor functions to permit the inclusion of the combustor in the 10-kW system in the next stage of combustor/system experiments.

Work continued on the fabrication of the gas flow control systems to be shared between the combustor test stand and the 30-kW system stand. The primary components of the flow control systems have been received and the fabrication of the flow control system(s) is nearly complete.

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

Hardware for the 30-kW ECE combustor has been fabricated and assembled to assure proper fit. All preliminary cold flow analyses of the combustion hardware have been performed. Testing included analysis of air and gas flow paths with discharge into atmospheric pressure using air. The data was used to estimate the actual pressure of various flow streams at operating pressure and composition. The fuel system is in the process of being calibrated at various back pressures to assure proper fuel metering throughout the turndown ratio. The fuel system is setup with the proper pressure biasing to theoretically provide predictable fuel metering control. The key issues being addressed are the fuel pressure regulator gain and hysteresis characteristics at various absolute pressures and total fuel flows. The dual-stage multi-flow path air-blast atomizer has been characterized over the expected turndown ratio while discharging into atmospheric pressure. Necessary fuel flow calibration for the start nozzle (pressurized atomizer) was performed to assure acceptable performance for startup.

A 30-kW system combustion air preheater has been added to the system and the design/build specifications have been sent to Harrison. With this addition, primary emphasis is concentrating on the integration of the test stand combustion concept into the "reformer" (conceptualization stage); Delco Electronics has been supplied the necessary information on the combustion controls to begin the controller evolution.

Task 1.1.4 Alternate Combustor Concepts

No work was performed on alternate combustor concepts in this quarter.

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 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.

Task 1.2.1 Reformer Development for 10-kW System

The 10-kW system is being used for the testing and development of components for the 30-kW system fuel processor. The 10-kW reformer is a stirred reactor; a new combustion heated version of this reformer has been delivered. The new reformer will also allow the methanol and water injectors to be mounted directly onto the reformer vessel, thus eliminating the pool boiler.

Autothermal Reforming of Methanol. Autothermal reforming of methanol combines partial oxidation and steam reforming to produce a hydrogen-rich fuel for fuel cell use. This approach could eliminate the necessity for system heat transfer that pure steam reforming requires. The previous quarterly report discussed the theoretical possibility of autothermal reforming. Thermodynamic calculations indicated that autothermal reforming has significant potential for producing a hydrogen-rich fuel. This quarter, an intensive experimental study of this approach was conducted. A list of the experiments performed is presented in Table 1. The catalysts used are labeled Catalysts A and D due to proprietary concerns.

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TABLE 1

METHANOL AUTOTHERMAL REFORMING EXPERIMENTS

<u>Date</u>	<u>Catalyst</u>	<u>Purpose of Experiment</u>
7/10-7/12	A, 25 g	Feasibility of autothermal reforming.
7/18-7/20	A, 100 g	Effect of different air/methanol ratios on product composition and bed entrance temperature.
7/20-7/28	A, 100 g	Life test of catalyst using high air/methanol ratio. Catalyst was unchanged from previous experiment. 69-hr run.
8/8-8/18	A, 150 g	Test of different reactor configuration to reduce catalyst decay. 110-hr run.
8/22-8/25	D, 123 g	Test of Catalyst D using second reactor configuration.
8/29-9/12	D, 137 g.	Further test of Catalyst D; effects of shutdowns and startups.
9/14-9/19	D, 126 g	Test of third reactor configuration to reduce catalyst decay.
9/19-9/22	D, 121 g	Tests of different operating conditions to reduce catalyst decay.

The three-month series of experiments yielded the following conclusions:

- The autothermal reforming of methanol is a feasible approach to create a hydrogen-rich fuel without the necessity of heat input or cooling for the process.
- At operating temperatures within the 225-275°C region, an approximate thermodynamic equilibrium product composition can be reached with reasonable amounts of catalyst.
- Dry-basis hydrogen composition in this particular autothermal reforming product (~64%) is less than the dry-basis hydrogen composition of the pure methanol steam reforming product (~74%). System analyses will have to be used to determine whether elimination of the heat-input requirement justifies this loss in product hydrogen content.
- A "hot spot" tends to form at the entrance to the catalyst bed, where the heat generated by oxidation is greater than the heat absorbed by reforming. This "hot spot" causes the catalyst to decay. As time progresses, the "hot spot" moves down the catalyst bed, eventually killing the entire bed.
- Proper choice of operating conditions can minimize the "hot spot" and slow its progress to a tolerable level. Catalyst life of over a thousand hours of operation is predicted for some of the tested operating conditions and catalyst loadings.
- As a result of the previous two conclusions, catalyst life then becomes a function of the amount of catalyst in the reactor. Any desired life can be attained if sufficient catalyst is used.
- In the system using Catalyst A during the 110-hr life test, it is estimated that 5.6 kg of catalyst would be needed for a 10-kW (net) system to produce a catalyst lifetime of ~1500 hours.
- Tracking product composition does not indicate catalyst deterioration during early stages of operation. Measurement of bed temperatures is necessary to detect catalyst deterioration.
- The system can be subjected to sudden shut-downs and start-ups without noticeably affecting the catalyst or process operation.
- Catalyst D is significantly more active than Catalyst A, reflecting similar observations during steam reforming of methanol.

- There are some indications that Catalyst D may not be as stable as Catalyst A under autothermal or steam reforming conditions.
- Since less water is required in the autothermal reforming process than in steam reforming of methanol, similar or even increased system efficiency may be possible if autothermal reforming is used in preference to steam reforming. However, in these tests a significant amount of water vapor was added with the methanol vapor; consequently, the catalyst was operated in a reduced state which may have contributed to its deterioration during autothermal operation.
- Optimum operating conditions have not yet been ascertained. Additional experiments will almost certainly yield significant improvements in this process.

The dry-basis product composition from a 110-hour autothermal reforming test using Catalyst A is presented in Figure 1.1. For the entering feed that was used in the test, the thermodynamic equilibrium values for a 500°K exit temperature are 64.1% H₂, 22.4% CO₂, and 1.4% CO. Temperatures were varied at different times in the experiment, so these values only approximate the true equilibrium value at the reactor exit at different times. Nevertheless, the plot indicates that essential thermodynamic equilibrium was maintained in the product throughout

the test. During this test, the measured temperatures indicated some catalyst deterioration in the vicinity of the bed entrance.

Search for More Active Reforming Catalysts. Recent technical papers have described new catalysts for the steam reforming of methanol; these catalysts are apparently superior to any that have been previously tested. Toward the end of this quarter preparations were underway to formulate these catalysts at the JDC laboratory. The necessary chemicals have been ordered and equipment has been collected for this purpose. Consultation with the catalysts' developers yielded insight into the formulation process, complementing the literature recipes. Preparation and testing of the catalysts is expected during the next quarter.

Reformer Design.

- **Isothermal Plug Flow Reformer Testing:** Testing of the isothermal plug flow reformer is complete; the results were previously reported in the third quarterly report. As this concept utilized a recirculating high temperature heat transfer fluid, adaptation of such a device for automotive use was believed to be impractical; pursuit of this concept has been suspended. However, one item should be noted here; prior to shutting down the experiment the same apparatus was used to test autothermal reforming for several hours. It was believed that the recirculating heat transfer fluid, in combination with the close heat exchanger proximity, could be used as a thermal moderator for the exothermic oxidation reaction. This would prevent overheating and, consequently, deactivation of the catalyst. When the reformer was

opened, after the autothermal experiments were completed, there was no evidence of catalyst deactivation as was observed in the other autothermal reforming experiments.

- **Combustion Heated 10-kW System Reformer:** The combustion heated version of the 10-kW reformer has been built and delivered to the JDC. In addition to accommodating combustion heat, this reformer allows the water and methanol to be injected directly into the vessel. Currently, the combustion heated reformer for the 10-kW system is being prepared for installation into the system. Initially, the reformer will use the original pool boiler. Once stable operation is obtained using the pool boiler the injectors will then be mounted onto the reformer itself to test direct vaporization.
- **Combustion Heated 30-kW System Reformer:** The 30-kW system reformer design has been completed and is now in the fabrication phase. The design takes advantage of the benefits of both a recycle and plug flow reactor. Projected mass of this reformer is 60 kg. Projected volume is 60 l. The expected delivery date of this reformer is during the next quarter.

TASK 1.3 SHIFT REACTOR DEVELOPMENT

Efforts on the development of the shift reactor during the past quarter have been minimal. The size of the shift reactor for the 30-kW system has been determined. More work is now taking place to better understand the water gas shift reaction while more shift catalysts are being evaluated using the Caldwell Reactor. Any improvements in the CO output of the reformer will reduce the size of the shift reactor. Functionally, the shift reactor is quite simple, therefore the final design will be straight forward.

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. The isothermal PrOx for the 10-kW system was built and delivered to the JDC at the beginning of July, 1995. Test results from this device are discussed in the System Management section of this report.

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

Samples of the sensors have arrived at the JDC and are being tested in the 10-kW system by the ECE System Team.

TASK 1.7 REFORMER OVER-PRESSURE SWITCH DEVELOPMENT

An over-pressure switch to detect an over-pressure condition in the reformer has been specified and delivered to the JDC. Testing of the switch will be performed by the ECE System Team on the 10-kW system.

TASK 1.8 DEVELOPMENT OF CO SENSING

CO Sensing is described under the ECE System Integration and Controls section of this report.

TASK 1.9 FUEL PROCESSOR MODELING

EASY5 (transient simulation) model components for the principal sub-assemblies of the reformer/heat exchanger (hex) have been developed and coded; these include the: burner/hex, plug flow reactor/hex, and the recycle reactor. In addition, the methanol/water evaporator component model has been coded.

Comparison of the JDC developed burner/hex model performance to Delphi Harrison's heat exchanger performance code indicates good agreement in hex performance. The two models are complementary in that the Harrison model uses more rigorous heat-exchanger design information but constant and averaged thermodynamic/transport properties. The EASY5 model incorporates the resultant "design" data into a flexible model allowing computational investigation of a component's performance with transients, turn-down, and varying fluid properties.

With reasonable reaction rates assumed for methanol reforming the recycle reactor model also exhibits good transient agreement with design expectations.

With the addition of "minor" model components (heat exchange to the reactor shell, mass storage, and fan sub-assembly) expected to be completed this week, full "integration and testing" of the transient reformer simulation code will be initiated.

Previously, transient models for the other reactor components, the PrOx and shifters, have been coded; combining these models with the reformer simulation code will complete the bulk of the fuel processor subassembly transient simulation model. As the fuel cell and humidifier components have already been modeled, the overall fuel processor/stack assembly transient simulation model is approaching completion. The air flow and heat rejection subassemblies, part of which have been modeled, still remain to be completed. These codes consist primarily of library modules (with some modification) so their transient simulation coding should proceed more rapidly than transient code describing more novel reactor components.

For interim design information, FCSYS, the steady-state code, has been enhanced. Major changes include the capability to set desired anode and cathode humidification levels and the expander pressure. Overall performance now includes the effects of stack humidification and condensation.

WBS TASK 2. FUEL CELL STACK DEVELOPMENT

SUMMARY

- A hydrogen adsorption/desorption test apparatus was designed and constructed, and is now operational. Measurements from this device enable evaluation of the active platinum surface area, yielding insights on the effective platinum catalyst utilization of MEAs
- 500 hours of single cell operation was achieved with coated titanium plates (anode and cathode) with no appreciable voltage degradation of the SEL (Stack Engineering Lab) MEA due to the metal plates. The voltage drop across the metal plates remained low (< 5 mV) over the entire testing period
- The active area for the full size stack will be increased to 500 cm^2 to improve power density, eliminate over 1300 pieces per stack, and ultimately reduce production cost
- Computational fluid dynamic modeling of the flow in the anode and cathode plates was initiated. The modeling included provisions to simulate the flow of gas across neighboring channels through the backing paper
- The first 340 cm^2 active area membrane and electrode assembly was produced using the modified PHI press. This appears to be a very good design with excellent electrode lamination; consequently, the GM full scale stack can now be built with SEL products.

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

Task 2.1.1 Dow Membrane Development Contract

Due to a Dow management decision to eliminate all of their fuel cell related efforts, Dow has terminated their relationship with the program. During this quarter Dow did supply 9 small MEAs and 7 large MEAs. The following paragraph reflects Dow's decision on their choice to exit the program.

"Dow has elected to discontinue its plans to aggressively pursue development of fuel cell systems and components based upon proton exchange membrane (PEM) technology. This is a business decision based upon Dow's ability to structure an acceptable ongoing business relationship beyond Dow's current collaboration program with Ballard Power Systems of Vancouver, BC, Canada. A number of factors contributed to this business conclusion including:

- The substantial research and development investment still required
- The relatively long projected development time to commercialization
- Uncertainties about future markets for dispersed and mobile electric power generation
- Costs of competing energy technologies
- Government funding support.

As a result, Dow was unable to appropriately satisfy the interests of the shareholders of both companies.

Dow remains confident in the versatility and capability of PEM fuel cell technology to produce electricity in an efficient and environmentally benign manner. Dow believes PEM [technology] has the potential to eventually compete favorably in satisfying an evolving set of market and societal demands regarding energy generation sources. While Dow's internal financial requirements and business priorities do not at this time support its participation, Dow believes others will undoubtedly proceed with completing the development of this promising technology."

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Dow is currently ramping down its collaborative efforts with Ballard Power Systems, internal research and development efforts, contract research, and membrane production.

Plans for Terminating Dow Participation. The primary issues from Dow's perspective are to minimize any negative impact on the program and to define and facilitate appropriate technology transfer. Meetings in early July with GM program and technical management resulted in general agreement between the two companies. Dow has agreed to the following deliverables to provide an ordered project transition with completion expected by the end of October.

1. Deliver MEAs to Support Stack Development
 - Lab Scale ($1/20 \text{ ft}^2$)
 - Stack Scale ($1/3 \text{ ft}^2$)
2. Evaluate Fabricated Membranes using an Abbreviated Experimental Design
 - Provide "start-up" performance data
 - Measure "in-situ" permeation rates
3. Write a "Final" Report.

The first two items are largely complete and the final report is in progress. Supply of additional Pt-black membrane-electrode assemblies was needed to support a smooth transition to the supported catalyst electrode components. These enable effective comparison of stack and lab cell data without introducing issues that could pertain to a change in MEA structure. Completion of a smaller than planned set of experiments at Dow will bring their membrane development efforts to an appropriate stopping point. Dow has active efforts to license their membrane technology and secure a supplier of Dow polymers and membranes.

Progress Summary. The following list of progress toward Dow's deliverables is presented as a brief summary. Results and experimental detail will be provided in the final report which will be completed and submitted to the program technical director by the end of November, 1995.

Membrane/MEA Supply to Support Stack Development

- MEA process modified to provide $1/3 \text{ ft}^2$ MEAs
- Supplied requested MEAs - Lab and Stack Sizes

Membrane Cost Reduction

- Epoxidation - Process change unsuccessful (design, installation, initial tests completed)
- Thinner Membranes - Lower polymer requirements (part of membrane improvement below)

Membrane Performance Improvement

- Experimental Design Developed
- Membrane Fabrication Completed
- Polymers Synthesized (4, 50 lbs each)
- Membranes Fabricated (20, 40 ft^2 each)
- "Standard" Properties Measured
- Fuel Cell Testing Initiated
- Performance Decay Measurements Initiated
- Electrode/System Determined
- Decay Rates: No Impact of "Extractables"
- Pt-black Sintering Observed

Methods/Hardware Development

In-Situ Measurements of Gas Permeation Rate
Cell Effluent Fluoride
Modified Flow Field Plates/Compression.

Dow's cost reduction efforts focused on the most significant long-term cost factor under high volume production conditions, i.e., the polymer cost in the membrane. The quantity of polymer used is important, but by far the largest contributor to polymer cost is the monomer; improvements of yield and production rate in the epoxidation step can provide the largest cost reduction and were the first targets addressed in Dow's program.

Dow's program focused on optimization of polymer properties, primarily through equivalent weight variation. In this program phase detailed performance comparisons were planned, focusing on both stability and optimum transport properties. Polymers were made in quantities which would have enabled testing through short stack and full stack demonstrations. Basically, polymers and membranes were produced and testing was initiated.

Two primary performance factors, membrane stability and gas permeability were to be studied in depth. These required development of additional experimental capability. In-situ measurement of gas permeability was needed to help define optimum performance and, hence, selection of the best compromise between membrane conductivity and permeability utilizing the best choice of equivalent weight and thickness. Stable hardware is critical to provide the capability to study the performance stability of both membrane and electrode structures.

Task 2.1.2 DuPont Membrane Development Contract

Development of Manufacturing Process for Advanced Membranes. Process modifications designed to increase the fuel cell membrane production capabilities of existing commercial facilities were implemented. These modifications were instrumental in the successful production of 42 square meters of NF-112 membrane (200 pieces, 18" x 18") for the DoE/GM program. This membrane was shipped to the JDC during this quarter as a contract deliverable.

Development of Catalyst Coated Membranes. The work focused on modifications to the fuel cell test laboratory and standardization of the procedures both for preparation of catalyst coated membranes and for fuel cell performance evaluation. Significant improvements were achieved in fuel cell performance as a result of a number of modifications to the procedures for catalyst ink preparation and treatment of the coated membrane.

In addition to experiments on coated membranes prepared in-house, testing was also performed on catalyst coated membranes prepared at the Joint Development Center (JDC) at Los Alamos National Laboratory (LANL). Preliminary results indicate that DuPont MEAs with a catalyst loading of ~ 0.4 mg Pt/cm² on NF-112 (N112) membrane performed equal to or marginally better than the GM/JDC MEAs (with ~ 0.3 mg Pt/cm² on N112 membrane). The tests were conducted at DuPont. With hydrogen and air as the reactant gases at 30 psig and a cell temperature of 85°C, a current density of 1.1 A/cm² could be obtained at 0.7V over a total active area of 50 cm². During these experiments it was generally realized that these MEAs were much more sensitive to changes in water content in the incoming gases than the MEAs prepared by conventional methods (i.e., by coating the catalyst on carbon paper/cloth and hot pressing the carbon cloth/paper onto the membrane). Efforts are under way to improve the water management characteristics of the MEAs. Performance measurements on two different test stations showed very good reproducibility. Also reasonably good reproducibility was achieved with several identical MEAs prepared by the same procedure.

Task 2.1.3 Electrocatalyst Development

Hydrogen Adsorption/Desorption (HAD) Catalyst/MEA Evaluation. A stand alone hydrogen adsorption/desorption test apparatus was designed and constructed, and is now operational. Measurements from

this device enable evaluation of the active platinum surface area, yielding the effective platinum catalyst utilization of the JDC's MEAs, as well as other MEAs. Hydrogen adsorption/desorption and CO oxidation are both used to evaluate the active amount of Pt surface area in the catalyst layer of a MEA.

MEAs with different catalyst loadings of 0.07, 0.18 and 0.29 mg Pt/cm² and different Nafion loadings (25, 30, 35, 41, 45, and 50 % Nafion) have been examined since the test apparatus has been finished. The results are still being analyzed and will be presented at a later date. Early HAD data, indicating the utility of this technique for

rapid MEA screening, was presented at the October Electrochemical Society Meeting in Chicago.

Single Cell Performance Testing. The effect of CO₂ dilution on single cell performance was measured. The results are presented in Figure 2.1. The test configuration included an anode catalyst loading of 0.29 mg Pt/cm² and a cathode catalyst loading also of 0.29 mg Pt/cm²; no anode air injection was utilized. The results indicate that CO₂ dilution alone does not result in as much performance loss in SEL MEAs as was observed in the much higher Pt-black loaded Ballard stacks. This result is not surprising as others have reported significant CO₂ caused intermediate species loss in

Figure 2.1 Effect of Anode Gas Composition on SEL MEA Performance.
Anode catalyst: 0.29 mg Pt/cm², Cathode catalyst: 0.29 mg Pt/cm²; no anode air injection.

high Pt loaded cells.

The effect of synthetic reformat on the performance of SEL MEAs was measured and is reported in Figure 2.2. The configuration for this test was an anode catalyst loading of 0.29 mg Pt/cm² and a cathode catalyst loading also of 0.29 mg Pt/cm²; again no anode air injection was utilized. As expected, performance loss was much greater with this fuel mix than with CO₂ only, indicating that catalyst poisoning is occurring from CO. Air injection and/or anode alloy catalysts have been proposed to address this problem.

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The effect of different catalyst loadings on performance was also measured and is shown in Figure 2.3. This test is a designed factorial experiment where the separate effects of anode and cathode catalyst loadings can be investigated. The analysis is currently underway.

A performance comparison was made between NF-112 (N112) and NF-115 (N115) membranes, as shown in Figure 2.4. MEAs made with N115 result in the expected performance loss at high current densities as compared to N112 due to higher membrane resistance. However, at very low current densities the performance on the N115 membrane is higher due to lower H₂ crossover.

At the GM R&D Center, optimization of the membrane/electrode assembly preparation by the lamination approach continued with detailed studies varying the cathode Nafion content in the 20-40% range. H₂/air fuel cell performances were measured with a Dow membrane at 80°C, 30 psig hydrogen (1.5 stoichiometry), 55 psig air (3.0 stoichiometry) and a Pt loading of 0.45 mg/cm²/cell. The actual electrochemical active area was measured using hydrogen electrochemisorption. Both methods showed that the optimum Nafion content in the catalyst layer is 30 w/o. Additional evaluations involved a Nafion 105 membrane, which demonstrated lower performance compared to the Dow membrane, and a 0.8 mil thick Gore membrane that demonstrated reasonable cell performance, but a lower open-circuit voltage.

Task 2.1.4 Develop Phase II MEA

MEA Quality Control and Diagnostics. An important part of the fabrication process is microscopic evaluation of the production samples. SEM (Scanning Electron Microscopy) and EDS (Energy Dispersive Spectroscopy) have been used to characterize various materials currently being used in the project. These include bipolar plate materials, graphite papers, MEAs, and related project components.

Membrane electrode assemblies (MEAs) are continually analyzed as they are produced from the stack

Figure 2.5 SEM Micrograph (backscattered electron image) of a SEL prepared MEA using Membrane A material. The 16-Series catalyst layers are uniform and have consistent thickness without large catalyst particles.

engineering laboratory (SEL). These analyses have documented dramatic improvements during this quarter regarding the uniformity of catalyst layers in the MEAs. Selected data are presented to demonstrate that progress.

A backscattered image of a 16-Series ink formulation on Membrane A material, which was obtained from a source outside the program, is presented in Figure 2.5. The catalyst layer is very uniform, however, the catalyst layer thickness does vary. Higher resolution pictures show that small particles with locally high Pt content exist. (Most likely such small heterogeneity will have no effect on the device performance.) However, the processing

does provide MEAs with uniform catalyst particles and consistent catalyst layers.

Figure 2.6 SEM Micrograph (secondary electron image) of SEL prepared MEA using Membrane A. The small cracks are areas where catalyst is not usefully present.

Figure 2.6 is a secondary SEM image of a different ink formulation on Membrane A material and was taken following tests with disappointing results. In this figure the two thick layers are the electrode layers, both of which are thicker than the middle membrane layer. The electrode layers are identical in thickness to those deposited on Nafion™ materials, therefore, these photographs appear different because of the very thin Membrane A material. In this MEA small cracks can be observed where catalyst voids form along the membrane surface. These cracks are not observed in backscattered electron microscopy,

indicating that the catalyst crack is very shallow. Due to the locations and sizes of the cracks they were most likely formed during the catalyst decal drying. Proper ink viscosity is a key concern in preventing the formation of cracks such as these. This has not generally been a problem during MEA production in the SEL (with Nafion™ materials), but ink viscosity effects are being thoroughly evaluated.

Figure 2.7 is a secondary SEM image of an experimental ink formulation on Membrane A material. The catalyst layer of this MEA shows catalyst particles large enough to form holes in the membrane. In fact, this membrane was not tested in the single cell apparatus because of high crossover rates denoted by a low open circuit potential.

Figure 2.7 SEM Micrograph (backscattered image) of a SEL prepared MEA using Membrane A material. The catalyst layer shows large catalyst particles with sufficient size to penetrate the membrane during the hot pressing step.

Gas Transport. A test protocol has been developed to evaluate the hydrogen gas transport through membranes and MEAs. The results of these tests are presented in Figure 2.8. The curves allow differentiation between gas diffusion effects driven by partial pressure differences and leakage effects driven by total pressure differences. The diagnostic value of this test is the ability to detect holes in materials with high permeability. The overall systems impact of the test results is a better understanding of the trade-offs when high performance, high permeability membrane materials are utilized. The hydrogen cross-over rate of 2.5 sccm H_2 in Fig. 2.8 represents 2% of the total H_2 consumed electrochemically at 0.25 A/cm^2 .

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Task 2.1.5 Ballard Stack Development Contract

MK5E Stack Support. Stack 108 was returned to the Joint Development Center. There has not been any additional requirement for MK5E hardware activities at Ballard during this period. Support availability for these stacks continued during this period as well as support for other Phase Two stack related activities. These activities include advice on air and air compressor specifications and cell voltage monitoring methods.

Gen I Ballard Stacks. The Gen I Ballard stacks are rated at 30-kW each. They are comprised of three columns of cells manifolded together with internal humidifiers in each column. Detailed performance, operating, and interface specification have been provided to the program.

Gen II Ballard Stacks. Gen II Ballard stack concepts are based on a power requirement of 50-kW at around 100 volts. Based on these requirements, several design concepts have evolved, with two particularly favored choices. Both concepts have active areas of about 0.5 ft² and have power densities in the range of 500 to 700 watts/liter. The main objectives of the Ballard Gen II design are to increase the power density, match the system voltage and other target specifications as closely as possible, scale up the stack power output, and separate the humidifiers from the stack.

Stack design concepts are being evaluated through computer modeling and testing of prototype cells. Novel designs are being evaluated to optimize active area utilization, heat transfer, fluid flow, stack scale-up, and other

parameters. Good progress is being made in minimizing performance sensitivity to reactant pressure, stack temperature, and anode air bleed quantities.

Various catalysts are being continuously evaluated. These catalysts employ different platinum loadings, composition, and production processes. A state of the art production catalyst with the best combined characteristics of performance level and stability, anode air bleed, temperature, and reactant relative humidity sensitivities will be used in the Ballard Gen II stacks.

TASK 2.2 FUEL CELL TEST HARDWARE DEVELOPMENT

Task 2.2.1 Single Cell Test Stands

Most of the work to develop improved single cell test stands is complete and these stands are now in routine use. A tank farm has been installed to supply synthetic reformat (with 15 ppm CO) to the single cell test stands. A data analysis software program was written to quickly plot and extract constant stoichiometry data from the test protocol. The remaining activity in this task involves bringing an earlier version of a "PETE" test stand up to current specifications. (During the past months, it was decided to upgrade the wattage rating of this fixture design.) When completed, one of two identical PETE test stands will be transferred to General Motor's laboratories.

The physical construction of a Long Term Single Cell Test Stand is now finished, with full LabVIEW based computer data acquisition. The test stand is now in operation being used mainly for long term evaluation of both bipolar plate materials and MEAs. Both short term testing (100 - 200 hours) and longer term testing (500 hours) have been completed with excellent test stand performance.

Task 2.2.2 10-kW Fuel Cell Stack Test Stand

The newly-improved 10-kW stand has been completed, with the exception of the steam generator that is required for testing stacks that do not include humidification sections. Importantly, the LabVIEW based computer control and data acquisition is fully operational. This stand has been programmed to be capable of conducting a series of stack test protocols.

Task 2.2.4 60-kW Fuel Cell Stack Test Stand

The large scale fuel cell stack test stand is being fabricated at a vendor location for delivery to Rochester during November, 1995. Protocols written for the 10-kW test stand will also support the computer control system on this fixture.

TASK 2.3 BIPOLAR PLATE DEVELOPMENT

The objective of this task is to identify and evaluate candidate materials to replace the graphite bipolar plates which now serve in early stack designs. The first candidates have been formed from titanium, both from commercial grade metal and that coated with various surface treatments. The surface treatment is designed to slow formation of a poorly-conductive surface layer.

Test results have demonstrated 500 hours of operation with coated titanium plates (anode and cathode) with no appreciable voltage degradation of the MEA due to the metal plates. The voltage drop across both metal plates remained low (< 5 mV) over the entire 500 hour testing period. The SEL MEA also has demonstrated 500 hour durability under these conditions. (This is the longest test duration of any SEL manufactured MEA to date.)

There was some physical evidence of corrosion on the coated Ti plates. However, weight loss of the metal plate was minimal (0.01 to 0.05 g), and the corrosion did not appear to effect the MEA performance or the

conductivity of the surface layer. The anode plate showed very stable resistance, while the cathode plate resistance did slowly increase due (probably) to the result of slow formation of a TiO_2 layer.

The conclusion of these PEM tests is that coated Ti plates appear to be an acceptable candidate for the GM Gen 2 stack design and do permit incorporation of metal plates in the next generation GM stack design. Titanium appears to meet the 500-hour performance target. Such titanium plates, however, are probably not a long term, low cost solution for PEM automotive stacks. However, these titanium pieces will be less costly than the present machined graphite plates.

Other candidate materials which have been tested in shorter duration (150 hours) include coated Al, uncoated Ti, and untreated stainless steel. These results are preliminary, however, and need replication. The uncoated Ti and stainless steel (316L) do not appear useful, especially for the cathode material.

At the GM R&D Center, a record of invention was filed regarding the development of a corrosion resistant and conductive bilayer coating for aluminum bipolar plates. Also, the effect of surface chemical treatment prior to the coating process is under investigation to resolve the problem of insufficient coating coverage on surface defects. Chemical and mechanical polishing procedures and deoxidizing treatments are also under study.

TASK 2.4 GENERAL MOTORS STACK DEVELOPMENT

Task 2.4.1 Develop Stack Design

The development of the Gen 1 General Motors Stack encountered a major testing delay due to corrosion problems with the test fixture end plates. Corrosion of the end plates was introducing large quantities of metal ions into the gas flow passages. Degradation of membrane performance from metal ion contamination is a widely recognized mechanism. The relatively low performance during the initial testing was at least partly attributed to this corrosion. All of the fixture modifications to address the corrosion problem are now completed.

During this quarter new data on vehicle drive electronic capabilities was received. The drive electronics design group now feels that converter ratios as high as 4 to 1 can be provided with a minimal efficiency penalty. By dropping the stack voltage from 150V at full load to 100V and increasing the active area proportionally, the same power level can be achieved with only two thirds of the cells. Over 1300 pieces per stack could be eliminated with this change producing a significant reduction in cost. The reduced number of cells also reduces the number of mating part interfaces which typically improves reliability. With the larger active area the fixed width required for the header and seal areas becomes less significant resulting in improved volumetric and gravimetric power densities. Based on all of these reasons the group decided to pursue the change immediately. The new design has 500 cm^2 of active area. Graphite test hardware for this size is called Gen 2. The metal plate design for this large active area is called Gen 3.

The concept for the exterior and non-repeating components has been modeled and is presented in Figure 2.9. The overall dimensions are expected to be 300 mm by 450 mm by 770 mm.

Figure 2.9 Gen 3 Stack Concept

TASK 2.5 STACK EVALUATION

Task 2.5.1 Test Protocols

During this period the 10-kW Test Stand became fully operational with the new DOS data acquisition and control system. This system utilizes a Pentium Computer operating LabVIEW Software and National Instrument data acquisition hardware. All electrical and data wiring is complete. The LabVIEW software program to control this equipment was

developed by Viewpoint (Rochester, New York). Full experimental control includes operation of the valves, mass flow controllers, pressure regulators, the current pulse width modulator, and temperature. Data acquisition/control time for this system is 1 second, about 10 times faster than the older Hewlett Packard software.

LabVIEW scripts have been written which specify operating conditions, set points, parameter shutdown limits, and test procedures. Seven Test Protocols are now in place, written in these LabVIEW scripts. A list of these Protocols is presented below:

- Protocol #1. PRESSURIZATION AND TRANSFER TEST
- Protocol #2. ACCEPTANCE TEST
- Protocol #3. BASELINE POLARIZATION CURVE
- Protocol #4. CATHODE PERFORMANCE
- Protocol #5. PRESSURE EFFECTS ON PERFORMANCE
- Protocol #6. SYNTHETIC REFORMATE EFFECTS ON PERFORMANCE
- Protocol #7. ENERGY, MASS, AND PRESSURE DROP TEST.

Task 2.5.2 Fuel Cell Stack Testing

A series of experiments were completed using Ballard Stack (Mark VE) Serial Number 109. This stack has been utilized to investigate the new testing protocols. Protocol #7, a long duration, steady state test generates reliable water balance and temperature data. A plot of the current density and voltage from this protocol is presented in Figure 2.10. Water balances for this same data set are presented in Figure 2.11.

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Figure 2.10 Current Voltage Results from Ballard SN 109. Hydrogen/air test results, showing average single cell data over an 11,000 s (3 hr) test period at 500 and 1,000 ma/cm².

TASK 2.6 FUEL CELL MODEL DEVELOPMENT

Task 2.6.1 Develop Electrode Model

Work continues on improving the basic understanding of PEM fuel cell performance. Recent advances at the University of Kansas (UoK) involve the addition of convective transport terms driven by pressure differences across the membrane separator. The formulation assumes that pressure drop occurs totally across the membrane and not within the porous catalyst layers. An additional energy balance equation has now been incorporated to describe temperature distributions along the solid body, resulting from electrochemical heat terms.

At the JDC, the through-the-membrane model has been integrated with this 2-D model, originally written by a LANL staff member and now UoK professor, Dr. Trung Nguyen, to obtain a comprehensive mechanistic model for predictive purposes. Calculation yields net water transport through the MEA structure and cell current as the flow moves down the channel. Cases considering both constant inlet flow rates and constant stoichiometry are computed. Extensive effort yielded a model that is numerically stable and converges under almost all of the test cases. Example results are described below.

With both reactants humidified to 100% RH membrane water content is maintained for adequate performance. However, adding such large quantities of water to the cathode stream is an energetic operation. Another mode of stack operation under consideration is feed the anode at 100% relative humidity (RH) while the cathode is fed at 50% RH. This specific case is illustrated in Figure 2.12 in which data is plotted as a relative distance along the channel. The anode relative humidity, initially 100%, is lowered by water loss, the result of water "drag" from

the anode to the cathode. The inlet cathode water humidity steadily increases from the inlet value of 50% to 100%. However, this relative humidity increase progresses slowly, and 100% RH is not reached until 0.6 of the channel length is traversed. This result occurs even with the thin Nafion™ 112 which has rapid water transport. The current, depicted as the upper trace, indicates an initial decline and then, as the anode humidity increases,

increases at this constant voltage level.

At 50% of the channel length, the cathode relative humidity becomes greater than the anode. Competition between electroosmotic transport and back diffusion of water toward the anode yields an anode RH that remains slightly less than unity in this model. After the 40% point local current density slowly decreases as oxygen is consumed.

This model is also useful to predict results from various operational PEM stack procedures. For example, one procedure is to promote anode feed water reduction by transferring part of the water flow from the cathode compartment to the anode compartment. This can be accomplished by feeding the fuel stream at less than 100% relative humidity (RH). In this second example, the anode and cathode relative humidities are 80 and 100%, respectively. The measured and predicted net transport of water as a function of current density is presented in

Figure 2.13. At low current densities water is transferred from the cathode to the anode just by the differences in relative humidity. At higher current densities, however, electroosmotic water transfer (drag) becomes more

dominant.

While these predicted trends of water transport are apparent, higher net water transport rates are generally predicted than are observed. The model at present continues to remove anode water, condensing steam into the membrane. Observed gas temperatures appear to rise from the heat of condensation. This additional heat tends to reduce the net water transfer. Heat transfer rates between the gas stream and channel are being included in the improved model.

The 2-D model predicts lower cell performance than either the 1-D model or experimental observations. More accurate modeling of the reactant consumption tends to predict decreased performance. It is suspected that kinetic data predicted from micro-electrodes under-predicts the exchange current density for fuel cells. As indicated by the authors who published the micro-electrode data which are used in this model, fuel cell kinetics may be a factor of 10 higher than those measured by micro-electrode experiments. A higher ratio of actual

platinum surface area to contaminants is suspected to cause the difference. Recent experimental data will be used to adjust the kinetic parameters.

Task 2.6.2 Develop Gas Composition Model

Computational fluid dynamic modeling methods for macroscopic representation of the reaction of gases at the catalyst layer were discussed. User defined subroutines can be utilized to define the mass flux at the catalyst layer boundary as a function of several variables including local gas temperature and composition. A macroscopic gas composition model could then be developed by combining the catalyst boundary condition with models of the gas flow both along the channels and across the lands through the backing paper. The behavior of the water droplets and films is still beyond the capability of available CFD software; however, models which do not explicitly represent the liquid behavior are still expected to provide significant insight for flow field optimization. A stepwise approach has been taken to allow various levels of model complexity to be validated in sequence. The first step is to model single component gases flowing along the channels and through the backing paper as described in the following section.

Task 2.6.3 Develop Gas Flow Model

A spreadsheet for calculating pressure drops in flow fields has been used extensively in design and evaluation work. This calculation evaluates the flow of reactant gases across neighboring flow channels in a given flow field through the porous backing paper. Pressure drop measurements were made in the flow visualization cell both with and without a paper backing layer. A pressure drop was calculated for the flow field without bypassing flow through the paper backing layer. These measurements should help validate predictions made by computational fluid dynamic (CFD) models.

Computational fluid dynamic modeling of the flow in the anode and cathode plates was initiated to further understand the flow characteristics of various plate designs. The modeling included provisions to simulate the flow of gas across neighboring channels through the backing paper. The permeability of the backing paper was derived from empirical data and then converted to porosity coefficients for use in the modeled flow field. Currently, the means of quantifying the relationship between porosity coefficients and permeability data is being optimized. Therefore, the mesh developed for the model produced a means to qualitatively compare various flow geometries.

In each of the models created, the solution was confined to a steady state incompressible analysis of pure air for the cathode and hydrogen for the anode. The analysis was simplified by neglecting the effects of relative humidity as well as heat transfer. Overall pressure drop, channel flow distribution, and flow across the channel lands were three variables tracked in the initial analysis. In addition, the effect of channel blockage by a water drop was simulated to determine which flow field design could best deal with such a phenomenon. The model indicated that the design level "03" channel configuration yielded the highest pressure build-up behind a simulated blockage, thus maximizing the force to blow the drop downstream. Comparisons of flow across the channel lands without simulated blockage were also made.

Current modeling efforts are centered on determining the mesh refinement required to more accurately predict pressure drop along the length of the flow path. Data has been obtained on current flow field designs without backing paper effects to provide a baseline to work toward thorough CFD modeling. Once the necessary mesh refinement is attained and proper porosity coefficients determined, a more quantitative analysis can be performed on various flow field designs.

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- Quarterly production resulted in excess of 80 complete MEAs, including samples with ultra-low Pt loading, Pt-Ru alloy catalysts, and unique catalyst layer ionomer properties.

WBS TASK 3. ECE SYSTEM INTEGRATION AND CONTROLS

SUMMARY

Prototype componentry is being designed and fabricated for integration into the scaled up ECE system. The Ballard 30-kW Stack and Fuel Processor Subsystem hardware will become available at approximately the same time next quarter. The fuel processor, including the combustion burner, will be assembled and tested as a subsystem. The Ballard Stack will be characterized off line on the Stack Test Stand in Rochester, NY prior to being integrated with the Fuel Processor Subsystem. The respective Thermal Management hardware will also be integrated into the system at that time. Shop air will provide the required system air flow until the air compression and expansion hardware become available for system integration. The GM Fuel Cell Stack will replace the Ballard Stack in the system when hardware becomes available in the summer of 1996.

A load absorbing power supply is on order from AeroVironment Inc. This device will be able to supply and absorb the necessary power to conduct testing of the integrated system.

TASK 3.1 AIR MANAGEMENT

After a thorough examination of the air compressor industry it was concluded that an air compressor did not exist that would meet the requirements of the ECE system. It was decided to procure existing technology units with limited modifications to expedite system integration and gain development experience within the existing program timing goals. Therefore Eaton Corporation, a major supplier of automotive superchargers to General Motors Corporation, had a tentative agreement in process to develop a (2) stage compressor under Delphi Harrison design direction. The first stage incorporated modifications to an existing Eaton Supercharger. The second stage was based on a high efficiency "Roots" design that was conceptualized and prepared for fabrication under Delphi Harrison direction.

Recent team discussions concerning altitude effects on compressor and ECE performance revealed concerns regarding high compressor operating temperatures that would exceed the practical limits of lubrication. (At altitude, to maintain stack performance at full load on reformat, the required compression ratio increases significantly from sea level conditions). Possible solutions were identified, but it was decided to halt further activity on the (2) stage compressor project so that existing resources could be better utilized in pursuit of a more optimized design for the ECE system.

The following strategy was developed:

- Vairex Corporation, a small R&D firm, is developing a variable displacement compressor. This unit is expected to provide the best overall efficiency, being able to respond to the wide range of required operating conditions, including the ultimate capability to operate as a combination compressor/expander. A preliminary contract proposal was received and is being forwarded to GM Legal Staff for review. More details, with costs, are due approximately 10/9/95.
- A.D. Little Inc. (ADL) has been developing various scroll compressor technologies and has presented sufficient technical detail to demonstrate that they can fabricate a compressor that meets the program requirements. ADL is currently preparing a project proposal to Delphi Harrison.
- Delphi Harrison is also investigating other design alternatives that could meet the ECE requirements. These also fall into the category of small business developers. However, each have some specific design features that could provide some performance advantages.
- In the mean time, "shop air" will be used in the initial development of the 30-kW ECE system.

TASK 3.2 THERMAL MANAGEMENT

Up to this point emphasis on heat exchanger development has been focused on the requirements of the Fuel Processor Subsystem. Delphi Harrison Thermal Systems is currently prototyping a compact heat exchanger unit for the 30-kW system reformer. This unit combines and integrates the heat exchange requirements of the stirred and plug flow portions of the reformer as well as the heat energy for methanol/water vaporization and superheat.

In early August a meeting took place at the JDC to discuss and schedule the heat exchanger needs for the ECE system. The following list outlines the thermal hardware requirements and related support that Delphi Harrison will provide over the next (6-9) months.

- CSTR/PFR reformer heat exchanger assembly
- Burner combustion air preheater
- Radiator w/electric fan assembly
- Adiabatic PrOx #2 pre-cooler
- Adiabatic PrOx #3 pre-cooler
- Coolant flow control valves for PrOx pre-coolers
- Heat rejection coolant flow control valve
- External cathode humidifier (10-kW stack)
- External anode humidifier (50-kW stack)
- External cathode humidifier (50-kW stack)
- Cathode & burner exhaust condenser.

The latest HEX units for the CSTR/PFR reformer, combustion air preheater, and 10-kW cathode humidifier are on schedule to be designed and fabricated by the year's end.

TASK 3.3 CONTROLS, DATA ACQUISITION, DIAGNOSTICS, AND SAFETY

Task 3.3.1 Laboratory Controls

10-kW Power System Development. Fuzzy logic control was implemented for the heaters in the 10-kW system mule with considerable success. The various reactors quickly achieve their requested setpoints across the operational range, with both response and accuracy improved over the previously implemented proportional-integral-differential (PID) feedback control scheme. The pool boiler, historically the most troublesome component in the system, is now well controlled. Fuzzy control is readily implemented using a subroutine record specifically developed for compatibility with the Experimental Physics and Industrial Control System (EPICS) software being used for control of complex systems in this project. The fuzzy rule sets will be further refined with actual system data from tests such as the fuel processor Designed Experiment described below.

The prototype gas mass air meters, developed by Delphi Energy and Engine Management Systems - Flint and described in last quarter's report, have been installed in the 10-kW system mule and are functional through the EPICS control system. Characterization of these meters, through testing, was initiated this quarter. Initial checkout across the desired operational range, with an in-series Hastings flow meter, demonstrated very good agreement. When testing is complete a report will be generated for Flint.

A preferential oxidation reactor designed to operate with relatively small temperature gradients was installed in the 10-kW system mule for testing. This PrOx reactor concept uses a stainless-steel heat-exchanger with a catalyzed heat-transfer surface developed jointly by Delphi-Harrison Thermal Systems and Delphi-Energy and Engine Management Systems (Flint). Testing of this PrOx concept will confirm whether improved temperature control can reduce the reverse water-gas shift that converts desirable product hydrogen and carbon dioxide back into water and carbon monoxide. The initial modular reactor design, depicted in Figure 3.1, uses a single-pass cross-flow heat exchanger core. This design will not achieve actual isothermal operation, but should be useful to demonstrate the concept. PrOx reactors used to date have been uncooled over the catalyst-bearing length (hence the description as adiabatic), leading to high temperatures and the potential for rapid reverse shifting.

Figure 3.1 Coated monolith used in adiabatic PrOx reactors (left) and experimental PrOx reactor based on a catalyzed heat exchanger (right). The finned surface (reformate side) is coated with a proprietary Delphi-Flint preferential oxidation catalyst.

A computer-controlled, expander-inlet throttle valve was installed to enable cathode pressure control over the operational range of the 10-kW power system. The smaller Powerex scroll expander (an air compressor being run backwards on a common shaft with the air compressor, with the stream to be expanded introduced into what is normally the compressor discharge port) provides adequate backpressure at full-flow conditions to achieve 375 kPa (about 40 psig at the JDC) cathode inlet pressure. As the system power is turned down and the air flow is reduced an additional flow resistance across the throttle valve is required to maintain adequate cathode pressure for Ballard stack operation on reformat. This throttle-valve pressure drop represents an unrecoverable loss in expander power and adversely affects system efficiency. In the current configuration this loss affects part-load efficiency. An alternate configuration would be to size the expander for pressure control at the part-load "sweet spot," with a bypass valve or waste gate diverting flow around the expander at higher flows to maintain pressure control. Both configurations would require a clutch (like that used on an automotive air conditioning compressor) to disengage the expander when the net recovery falls to zero. (Neither configuration enables highly efficient operation across the power range, making the variable displacement expander proposed by Vairex Corporation very attractive).

The combustion-heated reformer was prepared for installation in the 10-kW system mule. A trial parts assembly was performed and the necessary modifications to the test stand were designed. Fabrication of the reformer fan drive was completed; the stand and fan-motor mount are now in fabrication. The new reformer will be installed and testing will begin as soon as the combustor, including its necessary controls, is ready.

10-kW Fuel Processor Designed Experiment. A test matrix on the 10-kW fuel processor, that was developed using the formalized "Design of Experiments" (DOE) approach, to determine parameter combinations has been completed. Design of Experiments is a statistical method for minimizing the number of data points that must be taken while maximizing the information that can be extracted from the raw data. With proper design, a relatively few data points (fractional factorial) can characterize a complex system's response. This test matrix involved 5 blocks of test points (5 test days) with approximately 19 statistically determined parameter sets each day. Each test block came back three times in random order to "center points" to determine system repeatability. This feature can be used to prevent unintended shifts in system behavior from confounding the results. The seven

control factors varied in the experiment were overall fuel processor power (kW electric equivalent based on Ballard Mark 5 reformate polarization curves), temperature in the middle of the reformer catalyst bed, power of the reformer fan, molar ratio of Shift2 water injection to vaporizer methanol injection, temperature of the Shift2 inlet gas, temperature of the PrOx1 inlet gas, and PrOx1 air injection as a volume (molar) percentage of the total flow entering the PrOx. The central-composite experimental design required five levels for each control factor. The median level for each factor was chosen as the "nominal setpoint", i.e., that value currently considered as optimal and used for ordinary system operation. As this experiment was not intended to push the limits of the fuel processor a reasonable range was determined for each factor and the remaining four levels were centered around the nominal level within this range. The set with all control factors at their median levels was denoted as the "center point." To evaluate the system's performance seven gas concentration measurements were taken along the process stream: CO concentration at the reformer outlet, methanol concentration at the reformer outlet, CO concentration at the Shift2 outlet, the proportion of CO converted in the shift reactors, CO concentration at the PrOx1 outlet, and the CO, methanol, and hydrogen concentration at the fuel processor outlet.

A problem arose during testing resulting in a marked and sustained drop in reformer methanol conversion occurring between Test Day 3 and Test Day 4. The "blocked" design of the experiment allowed this "additive" shift in performance to be statistically accommodated. Subsequent model analysis, as described below, indicates this problem was successfully explained by reformer data analysis (downstream components were affected by methanol breakthrough). Subsequent system disassembly also showed that the reformer catalyst ("Catalyst D") had significantly deteriorated. There was evidence of densification (lower volume per pellet), mass transport (lower mass per pellet), and physical damage (metallic inclusions, cracking, and color changes). There was also evidence of a copper-bearing film plated out in parts of the reformer and on some down stream tubing. This behavior had not been observed with the baseline United Catalysts C18HC catalyst and the problem is under investigation.

The resulting data were analyzed, including graphical presentations that clarify system tendencies as a function of parameter variation. A response surface methodology was used to fit low-order polynomial functions to local test regions. Such models involve linear and quadratic terms in each factor and additional interaction terms based on the product of each possible pair of factors. Each model was judged by analysis of residuals, the relative variation explained by the model compared to unexplained residual variance, analysis of the absolute values of the coefficients compared to the error in their estimation, and physical sensibility. These models were then used to develop polynomials that determine setpoints for control factors based on the measured values of the remaining factors. Results are expected to lead to not only optimization of setpoints, but also to improved operating strategies and to high-payoff areas for system design refinement.

A formal report (GM Confidential) was generated¹ and a review of the results from the formal experimental test matrix on the 10-kW system fuel processor was held at the JDC, with Delphi Energy and Engine Management Systems and Delco Electronics engineers in attendance. Although it now appears the catalyst degradation midway through the test did confound some results, a great deal of information was extracted and understood using these statistical techniques. Future applications of this approach will enable the Systems Team to "work smarter, not harder."

System-Level Preferential Oxidation Tests. Testing of the "isothermal" PrOx reactor concept described earlier was initiated in the 10-kW system test mule. This heat exchanger, with a catalyzed heat transfer surface on the reformate side, was air cooled for initial testing. In order to provide stable PrOx inlet conditions all upstream

¹ Laura Iwan and Tom Ruscetti, "A Designed Experiment to Analyze the Performance of the 10-kW Fuel Processor in Response to Variations in Controlled Parameters," Delphi Energy and Engine Management Systems Advanced Product Engineering Report 0785, GM Confidential, August 31, 1995.

catalysts were replaced. Results to date are encouraging, with spot reductions in CO content of approximately 96% measured across a single stage of this device. However, this experimental reactor, built on a readily available heat exchanger core, does not actually achieve isothermal conditions. Cross-flow of the coolant results in a significant temperature variation in outlet gas temperature in the cross-flow direction (determined by a thermocouple array or "temperature rake"). It is difficult to achieve conditions across the device where the entire reformat stream sees both high-enough temperatures for oxidation (lightoff) and low-enough temperatures for water-gas-shift suppression. Analysis of the data was confounded by the use of a single tap for the gas chromatography sample as it is suspected that the local temperature variations lead directly to local PrOx performance variations. Modifications to the experimental setup are underway to enable gas sampling at each point where temperatures are measured, which will greatly improve understanding of the device operating conditions and thereby enable design of a more appropriate (more isothermal) heat exchanger.

In an attempt to even out the reformat temperatures across the PrOx "section," an experiment was run with two catalyzed heat exchangers in series, with coolant cross flow in opposite directions in the two units to achieve more uniform outlet gas temperatures. Because of hardware design limitations a single point of air injection upstream of the first unit was used. A heater in the air coolant line allowed the various flows available in the system to be simulated to determine if a simpler cooling scheme (such as diverting cathode exhaust through the coolers) could be implemented. Results of this testing were mixed. As before, very high spot reductions in CO content were achieved (the reduction factor approached 100x). These tests were run prior to the modification that added additional gas chromatography sample points, so overall "average" behavior was not obtained. Single point air injection for multi-stage PrOx reactors, a hoped for simplification, was not effective. At first-stage temperatures, where CO oxidation was effective, essentially all of the oxygen injected was consumed, with the result that the second stage was "inert." When temperatures in the first stage were low enough to allow oxygen to reach the second stage, CO oxidation in the first stage was poor. Also, those conditions leading to a high CO reduction featured air injection rates that would consume significant amounts of product hydrogen. This effect may result from PrOx reactor geometry (the flow path is very different in a single stage of this design than in the 400-channel-per-square-inch monoliths used in the adiabatic PrOx reactors). Additional testing will focus on a very well instrumented single stage in order to obtain the data required for an improved reactor design.

System Configuration Studies. The effect on net output and overall system efficiency of various electrical component efficiencies and power bus configurations were examined in a set of system computational studies. Considerations included drive motor efficiencies for the compressor, the cooling fan, the reformer recirculation fan, the fluid pumps, the efficiencies of the DC/DC upconverter between the fuel cell and the power bus (at battery voltage), and the DC/DC converter required to power the 12V loads (such as the system controller and injector fluid pumps) from the power bus. Alternate configurations powered the major drive motors from either side of the DC/DC upconverter. Net output and system efficiency were calculated using the FCSYS model at conditions from idle to maximum continuous power. Implications for further system development are being considered.

Task 3.3.2 Sensors

Gas Temperature Sensors. Delphi Packard Electric Systems has shipped the first set of Gas Temperature Sensors (GTS) for characterization and test in the 10-kW system. All sensors will be mounted using "Swagelock" type fittings in order to determine the optimum length for a sensing element. After initial test Delphi-P will build a set of GTS with defined length and welded 1/8" NPT fittings for installation in the 30-kW net system.

Combustor Flame Detection/Spark Control Module.

- **Direct Spark Ignition Module:** A direct spark ignition module that is designed to control spark sequence based on flame presence was purchased, modified, and tested. This device uses a flame ionization principle for detection with subsequent spark and time-out control. The module has been characterized on the bench with follow-up tests in the 10-kW combustion burner at the JDC. The tests yielded the following results.
 - The module could control flame-out sequence and spark initiation with response time less than 100 msec.
 - The flame ionization principle to detect the presence or absence of the flame is a reliable and effective way to perform this task compared to other methods such as UV or flame temperature.
 - The direct spark ignition module is, however, designed specifically for industrial boilers and cannot be easily modified to accommodate system requirements.
- **Delphi-E Flame Detection/Spark Control Module:** A Flame Detector/Spark Control Module was designed based on a flame ionization induced AC current rectification phenomena. The design utilizes a production ignition module pulse generator and has the flexibility to control spark duration, frequency, and duty cycle. The final device will be able to detect the presence or absence of flame and react to a failure mode such as a short in the sensing electrode. This module is undergoing prototype build with bench testing to follow.

Carbon Monoxide/Hydrocarbon Sensor.

- The basic components of the CO/HC sensors have been designed and are presently in fabrication. A key accomplishment has been the layout and validation of the electronics board which contains the following major components:
 - Low noise amplifiers - 4 channels for CO, HC, CO₂, and reference
 - Multiplexer
 - A/D Converter
 - Microprocessor
 - Feedback to analog chain
 - D/A and output interface
- Assembly code for acquisition and processing of digitized data has been written.
- The sampling cell design has been modified to utilize a single straight tube. Flanges with inserts for optical windows have been machined. Assembly of the cell with associated optics is now in progress; all IR filters have been designed and fabricated. These are presently being integrated with the thermopile detectors. The IR light source has been selected and will soon be integrated into the subassembly.

Hydrogen Sensor. The concept of using a Pd-based alloy as the sensing element in a resistive hydrogen sensor has been demonstrated. This thin film sensor has been integrated with a temperature controller and the integrated device has been tested at various temperatures and hydrogen concentrations. A linear response to hydrogen in the concentration range of 10-100% has been demonstrated.

Linear Oxygen Sensor. A Linear Oxygen Sensor has been purchased, installed, and tested in the MEA curing furnace at the JDC. The sensor will be used to detect the minimum oxygen content in the furnace during the MEA manufacturing process.

Water Conductivity Meter. A deionized water conductivity meter, Figure 3.2, has been received. The production device has been modified to meet the requirements of a 0-5V output signal and a 12V power supply. This sensor will measure 0-5.0 $\mu\Omega/\text{cm}^2$ with cell constants switchable at 0.5, 5.0, and 10.0.

Gas Relative Humidity Sensor. Three Gas Relative Humidity Sensors, Figure 3.3, have been built and delivered. A standard humidity sensor with temperature sensing element has been modified, repackaged, and tested based on requirements for 0.5-4.5Vdc output that is proportional to 0-100% RH. The RH sensor has better than 1%RH total accuracy and can operate in 0 - 85°C ambient temperatures.

Figure 3.2 Water Conductivity Meter

Figure 3.3 Gas Relative Humidity Sensor

Liquid Level Sensors. Optical type minimum tank level sensors were built, tested, and delivered. The standard Liquid Level Sensors were redesigned to accommodate Fuel Cell System requirements for accuracy, response time, operating environment, and interface.

These optical devices can only operate in non-freezing liquids (under normal ambient temperatures of -40 to +135°C) such as methanol and glycol-based coolant. The liquid level sensors using ultrasound for detection have been selected to measure the

deionized water level. This device can withstand liquid freezing without degradation in performance. Both liquid level sensor types are depicted in Figure 3.4.

Figure 3.4 Liquid Level Sensors

Air Compressor Input/Output Air Metering Modules.

- An automotive throttle body, with integral mass air flow meter, has been selected as a means to measure compressor inlet flow up to 120 g/s. Electronic Throttle Control can be added to control compressor outlet flow if the compressor is mechanized to run at constant speed and is a fixed displacement machine. In addition, a standard 10 micron air filter can be fitted to the throttle body inlet.
- An air compressor output manifold has been designed to distribute pressurized air to the fuel cell cathode and the combustion burner. Burner air flow is measured with a modified production mass flow meter and controlled by an automotive Idle Air Control Valve. The manifold assembly is depicted in Figure 3.5.

"USE OR DISCLOSURE OF REPORT DATA IS SUBJECT TO THE RESTRICTION ON THE TITLE PAGE OF THIS REPORT
AND IS IN ACCORDANCE WITH PUBLIC LAW 100-679"

Figure 3.5 Air Compressor Manifold Assembly

Task 3.3.3 Develop 30-kW Vehicle Control System

System Mechanization. The following modifications to the 30-kW system mechanization have occurred over the last quarter. The EECM hardware I/O requirements have not been affected.

- Cathode exhaust flow path revisions were made, resulting in significantly reduced flow requirements, in order to use an automotive actuator for control of combustor exhaust gas temperature
- The flow path of a valve diverting combustor exhaust flow around the reformer was revised to reduce the size requirements of the valve, thereby allowing use of existing automotive technology for this valve
- A cathode exhaust backpressure regulator was eliminated
- A mass air flow meter in the cathode line was moved to measure compressor inlet flow
- A combustion air preheater was added to facilitate methanol vaporization in the burner.

Hardware.

Based on the previously defined ECE System I/O for Mechanization Concept 14E, the Electrochemical Engine Control Module (EECM), Figure 3.6, is in the prototype fabrication phase at Delco Electronics. Hardware is on schedule for December 1, 1995 delivery. Current hardware status is as follows:

- The EECM case has been fabricated and is in the process of being plated
- The (4) layer fuel board has been fabricated and is currently being populated
- The custom (10) layer I/O board is in final layout
- A second Class II communication Bus has been designed and can be added to facilitate communication between the EECM and a vehicle control module. The existing Class II interface will be used for communication between the EECM and the (2) BPMs which will monitor fuel cell stack parameters.

A custom test simulator has been designed for verification testing of the EECM. The mechanical and electrical designs have been completed. Component parts have been procured and fabrication is in process.

Figure 3.6 EECM Case

Software. The base operating system for the EECM has been run on the advanced controller processor board. It is currently being expanded for use with the ECE hardware I/O.

The control algorithm development task is in progress. Two team meetings were held in August, one at the JDC and one at Delphi-E Rochester, to develop Control Block Diagrams for the ECE system. The general structure consists of a variety of software functions that are called as necessary. Some function examples include: startup, various running conditions, diagnostic, and shutdown routines. Significant progress has been made on a startup routine and portions of the general run control. Considering everything necessary for a fully functioning ECE system it is estimated that 5-10% of the total algorithms have been defined in Control Block Diagram form.

Task 3.3.4 Develop 30-kW Power Electronics

No activity occurred in this task for this quarter.

WBS TASK 4. REFERENCE POWERTRAIN DESIGN AND COMMERCIALIZATION

Activity on this task has not commenced since the subcontracts are still under review.

WBS TASK 5. PROGRAM MANAGEMENT AND REPORTING

In addition to the regular biweekly reports and conference calls a program review was held in Washington, DC on September 11, 1995. Proceedings of the meeting have been distributed to all program participants and the DoE. Per instructions from the DoE, a restructuring of the program is being planned in close consultation with DoE program managers. The GM program manager presented talks on the Fuel Cell Program at the DoE Fuel Cell Contractors Review meeting held at Morgantown, WV on August 9 and at the Joint US/Russian Workshop on Fuel Cell Technology held at Albuquerque, NM on September 27. A literature survey of processes for producing hydrogen fuel from different sources for automotive-propulsion fuel cells has been published as a Los Alamos National Laboratory unclassified report. A poster presentation on fuel cell bipolar plate material requirements for transportation applications was presented at the US/Russian Workshop on fuel cells.

FIG. 5-1 Total Program Cost Plan Graph

Cost Plan Revised May 25, 1995
DE-AC02-90CH10435

Electrochemical Engine Project

General Motors, Los Alamos National Laboratory, US Department of Energy

LEGEND

Task

Progress

Summary

Electrochemical Engine Project
General Motors, Los Alamos National Laboratory, US Department of Energy

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Electrochemical Engine Project
General Motors, Los Alamos National Laboratory, US Department of Energy

ID	WBS #	Task Name	1995															1996															
			1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31
47	2.6.1	Develop electrode model	■	■	■																												
48	2.6.2	Develop gas composition model	■	■	■																												
49	2.6.3	Develop gas plenum model	■	■	■																												
50	2.6.4	Develop stack thermal management strategy and hardware				■	■	■																									
51	2.6.5	Develop stack stress models (seals)	■	■	■																												
52	2.7	JDC Facilities Modifications	■	■	■	■	■	■	■	■	■	■	■	■	■	■																	
53	2.7.1	Proprietary Controls	■	■	■																												
54	2.7.2	Stack engineering laboratory at LANL	■	■	■	■	■	■	■	■	■	■	■	■	■	■																	
55	2.7.3	Modify current test stands for new tests	■	■	■	■	■	■	■	■	■	■	■	■	■	■																	
56	2.7.4	Office Improvements	■	■	■	■	■	■	■	■	■	■	■	■	■	■																	
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			1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32
58	3	ECE System Management																																
59	3.1	Air Management																																
60	3.2	Thermal Management																																
61	3.3	Controls, Data Acquisition, Diagnostics and Safety																																
62	3.3.1	Laboratory Controls																																
63	3.3.2	Sensors																																
64	3.3.3	Develop 30 kW vehical control system																																
65	3.3.4	Develop 30 kW Power Electronics																																
66	3.4	Miscellaneous Systems / Control Activities																																
67	3.5	30kW System Testing																																

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4	Reference Powertrain Design																																		
4.1	System Model Development and Application																																		
4.1.1	Power Source Model																																		
4.1.2	Integration of Power Source Model with Hybrid Vehicle Model																																		
4.2	Mission Definition																																		
4.3	Trade-off Analysis																																		
4.3.1	Powertrain Components Requirements																																		
4.3.2	Reference Powertrain Design Reports																																		
4.4	Commercialization Study																																		
4.4.1	Environmental Issues & Regulations																																		
4.4.2	Fuel Cell Transportation Economics																																		
4.4.3	Fuel Issues																																		

GM / DOE Electrochemical Engine Project

	Task Name	1995	1996
		1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31	
5	Program Management and Reporting		
5.1	Program Management and Administration		
5.2	Government / Industry Technical Meetings		
5.3	Reporting		
	Quarterly Reports	◆ 1/13 ◆ 4/14 ◆ 7/14 ◆ 10/13	◇ 1/10 ◇ 10/11 ◇ 7/12 ◇ 4/12 ◇ 1/12
	Reporting 1	◆ 1/13	
	Reporting 2	◆ 4/14	
	Reporting 3	◆ 7/14	
	Reporting 4	◆ 10/13	
	Reporting 5		◇ 1/12

Task

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Milestone

Rolling Up Task

Proprietary ECE Project Information

Project Plan as of October 1, 1995

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		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31
	Reporting 6																			◇	4/12											
	Reporting 7																					◇	7/12									
	Reporting 8																								◇	10/11						
	Reporting 9																												◇	1/10		
	Program Plan		◆										◆																			
			12/2										10/2																			
	Final Report																														◇	
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