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PHOSPHORIC ACID FUEL CELL DEVELOPMENT

FINAL TECHNICAL REPORT

SEPTEMBER 1980

Prepared by

A. Kaufman, P. Terry

Prepared for

U. S. Army Mobility Equipment Research and Development Command Fort Belvoir, Va. 22060

Contract DAAK 70-77-C-0206

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SUMMARY

The objectives of this program were to design, construct and test phosphoric acid fuel cells and fuel cell stacks capable of operating on typical product gases of a methanol or hydrocarbon reformer. Electrode performance was initially screened in a series of parametric tests in which operating temperature, fuel composition and fuel utilization were varied. Results of the parametric testing were used to establish the operating conditions to be used in the multiple cell stack testing phase of this program.

Preliminary multiple cell stack testing was performed using three-cell stacks, each electrode having an active area of approximately 0.4 ft². During initial testing on intermittent load profiles, it was observed that the three-cell stacks exhibited a marked performance decline associated with start-up/shut-down cycling. Upon observing this effect in two three-cell stacks the project effort was directed towards determination of the causes of cycling related performance losses.

Additional three-cell stacks were constructed and tested strictly for start-up/shut-down investigation. Individual cells which exhibited degradation due to start-up/shut-down cycling showed a corresponding decline in open circuit voltage. This observation led to the conclusion that the performance losses were due to reactant cross leakage caused by local breaching of the matrix, probably caused by acid loss during the transient conditions of water production and removal experienced during start-up/shut-down cycling. This conclusion was further verified by the fact that tolerance to start-up/shut-down cycling could be improved through use of a thicker, higher acid content matrix.

A parallel development program being conducted by Engelhard Industries for the Department of Energy (Contract No. DE-ACO1-78ET15366) was devoted to developing alternate matrix and electrolyte management concepts. It was, therefore, decided to postpone further start-up/shut-down tolerance testing until improved matrices were available from the matrix development program.

Upon availability of a matrix with improved acid transport properties, three additional three-cell stacks were constructed and committed to start-up/shut-down cycling. The three stacks accumulated a total of 8,496 hours of testing and 268 shut-down cycles. During the course of this testing, long-term performance declines were observed but no declines specifically related to start-up/shut-down cycling were noted.

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This work was performed by the

Research and Development Department of Engelhard Industries Division, Engelhard Minerals & Chemicals Corporation.

E. Starkovich, W. Taschek, and R. Belt of MERADCOM provided valuable assistance.

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4.0 INTRODUCTION

This program was performed in support of the U. S. Army's efforts to develop a family of lightweight, high efficiency, silent power sources suitable for military, field operation.

The objective of this program was to proceed stepwise from parametric testing of small scale, single-cell, phosphoric acid fuel cells to construction of full size, 2-KW, liquid cooled fuel cell stacks. This program was to have proceeded in the following steps.

- A. Parametric single cell testing (3 units)
- B. Fuel composition choice
- C. Three-cell stack construction and test (6 units)
- D. Ten-cell stack construction and test (2 units)
- E. Full size stack (2-KW net output at maximum rated load) construction and test (2 units)

The parametric single-cell testing was conducted utilizing a variety of fuel compositions typical of those which would be produced by a methanol or hydrocarbon reformer. As a result of the parametric testing and other inputs, a fuel composition of 65% H₂, 2% CO, 10% H₂O balance CO₂ was chosen for mixed gas testing of the multi-cell stacks.

Two of the three-cell stacks were tested using a continuous load profile. Two other stacks were tested using an intermittent load profile in which the stacks were periodically shut-down and allowed to cool to room temperature. The two stacks tested on the intermittent load profile showed substantial losses in performance which were not observed in the identical stacks operated on the continuous load profile.

As a result of this observation, the program was modified to investigate the causes of the observed performance losses associated with start-up/shut-down cycling. Four additional, identical three-cell stacks were constructed. One stack was operated on a constant load as a base-line. The other three stacks were operated on cycles which included shut-down periods of varying frequency and duration. During this testing, each individual cell's performance was monitored.

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It was found that with all individual cells that showed a performance decline associated with start-up/shut-down cycling the performance decline was accompanied by a corresponding decrease in open circuit voltage. These results suggest that the observed performance decline: could be due to breaching of the electrolyte matrix resulting from the transient operating conditions which occur during start-up/shut-down cycling.

Further testing was performed on stacks which utilized a newly developed matrix having high acid transport characteristics. During this test series of approximately 300 shut-down cycles, no matrix related performance declines were observed.

5.0 EXPERIMENTAL

5.1 Description of Test Samples - Parametric Single-Cell Testing

The parametric single-cell tests were conducted using 2 3/4 in. x 2 3/4 in. active-area cells. Carbonized graphite termination plates were used for current collection and reactant gas distribution. This was the same material that was used later in the stack tests. These 2 3/4 in. x 2 3/4 in. x 0.160 in. plates were inserted into gold-plated brass frames that had the same thickness and 2 3/4 in. x 2 7/8 in. openings. A 1/16 in. gap was left on both inlet and outlet ends for gas manifolding. 1 in. x 6 in. x 6 in. aluminum blocks were used on both sides as end-plates. These blocks were machined to allow the gases to be routed to and from the gas manifolds.

The electrodes utilized a 30% Pt/C catalyst (anode and cathode). The nominal loading for each electrode was 1.4 mg Pt/cm². Electrode substrates were Teflon-wetproofed Pfizer FD-33 carbon paper. The matrix was Engelhard's standard phosphoric acid membrane with a nominal thickness of 0.020 inches.

5.2 Description of Test Samples - Three-Cell Stack Testing

An isometric view of a three-cell fuel cell stack is shown in Figure 1. Figure 2 shows the end plate configuration of the first four stacks of this series. In an effort to eliminate the possibility of cell poisoning due to off-gasing from some non-metallic materials of construction minor changes were made in the construction of subsequent stacks. These construction changes are shown in Figure 3.

Tabular data on the fuel cell stacks are given below:

Cell active area $6" \times 9-3/4" (0.406 \text{ ft}^2)$ Electrodes, anode & cathode Stacks la, lb, 2a, 2b, 3a, 3b, 3c & 3d Pfizer FD-33, 0.015" thick Substrate 30% Platinum on carbon Catalyst type 1.4 mg/cm² each Platinum loading Stacks le, 2e & 3e Stackpole PC-206, 0.015" thick Substrate Catalyst type 10% Platinum on carbon 0.46 mg/cm^2 each Platinum loading

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Matrix type

Stacks 1a, 1b, 2a, 2b, 3a, 3b, 3c & 3d

> Standard Engelhard phosphoric acid membrane 0.020" thick (Stacks 3b, 3c & 3d employed 2 matrices per cell for extra acid inventory)

Stacks le, 2e & 3e

Laminated, SiC based, with provision for acid addition, 0.010" thick

Bipolar Plates

The bipolar plates were $7" \times 10.688"$ overall and 0.140" thick. They were machined from Stackpole Grade 2020 graphite. Air and fuel gas flow grooves were machined in the corresponding surfaces of the plates.

The porosity of the plates was sealed off by coating the surfaces with Tylan Corporation's Vitrigraf process.

Fuel Cell Edge Seal

The edges of the individual fuel cell electrodes were sealed from exposure to the opposite reactant manifolds by sealing strips placed along the edges of the electrodes. These sealing strips were of a free-standing matrix material as described in U. S. patent 3,453,149.

5.3 Description of Test Facilities - Parametric Single-Cell Testing

All parametric single-cell tests utilized the same test apparatus. The gas flow diagram for this apparatus is shown in Figure 4. Figure 5 shows the wiring diagram for this test station.

5.4 Description of Test Facilities - Three-Cell Stack Testing

Three separate test stands were used in the course of fuel cell stack testing. Each of the three test stands was of identical flow configuration. Figure 6 shows the flow schematic of the test stands. Figure 7 shows the electrical schematic.

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PARAMETRIC SINGLE-CELL TEST STATION

GAS FLOW DIAGRAM



LEGEND

- 1. Fuel Cell 2. Gas Flow Meter
- 3. Saturator
- Heating Tape
 Gas Washing Bottle
- Pressure Gauge & Regulator 6. 7. 8. " 9. 3 Way I 10. Beaker . 3 Way Ball Valve



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PARAMETRIC SINGLE-CELL TEST STATION

WIRING DIAGRAM



LEGEND

.

- 1. Fuel Cell
- Heating Pads
 Ammeter Shunt
- 4. Load Resistance
- 5. Millivoltmeter
- 6. Millivoltmeter
- 7. Binding Posts

- Temperature Controller 8.
- 9. Transformer
- 10.
- Saturator Heating Tape Variable Transformer 11.
- 12. Temperature Indicator

Figure 5

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GAS FLOW DIAGRAM

FUEL CELL STACK TEST STAND



15. Fuel Cell Stack

Figure 6

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WIRING DIAGRAM





LEGEND

- Fuel Cell Stack 1.
- Saturator 2.
- Carbon Pile Resistor 3.
- Load Switch 4.
- Ammeter Shunt 5.
- Millivoltmeter 6.
- 7. Voltmeter

- 8. Over Temperature Protector 9. Stack Temperature Controller
- 10. Saturator Temperature Controller

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- 11. Hydrogen Solenoid Valve
- 12. Temperature Indicator
- 13. AC Bus

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5.5 Test Procedure - Parametric Single-Cell Testing

5.5.1 Cell No. 1

Data on pure hydrogen were acquired at 300°F in the current-density range 0-150 ASF in 25 ASF increments. At each stable current-density, voltage was recorded for a hydrogen utilization of 90%. At a current-density of 150 ASF, voltage was recorded for hydrogen utilizations of 70%, 80%, and 90%. Testing then continued using an anode feed gas of the following composition:

65%	H ₂
10%	H ₂ 0
0.5%	C0
24.5%	C02

The composition of 10% H₂O was obtained by saturating the appropriate dry gas mixture with water at a temperature of 115° F. Voltages were recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilizations of 70\%, 80\%, and 90% at each current-density. Pure hydrogen was then rerun as described above. The following anode feed gas was then utilized:

65%	^H 2
10%	H ₂ 0
1%	C0
24%	C02

Voltages were again recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilization of 70%, 80%, and 90% at each current-density. Baseline hydrogn was then run again as described initially. This was followed by a third 'reformate' gas:

65%	H ₂
10%	H ₂ 0
2%	C0
23%	C02

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Voltages were recorded as before at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilizations of 70%, 80%, and 90% at each current-density. Baseline hydrogen was again repeated as described initially. The following anode feed gas was then run:

65% H₂ 10% H₂0 3% C0 22% C0₂

Voltages were recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilization of 70%, 80%, and 90% at each current-density. Finally, testing was concluded by rerunning pure hydrogen as described initially.

The temperature was then raised to 325°F, and the entire sequence of events described above was repeated for this temperature.

5.5.2 Cell No. 1A

Testing for this cell was identical to that for Cell No. 1 except that each time pure hydrogen was run voltages were recorded for hydrogen utilizations of 70%, 80%, and 90% at each current-density (not just at 150 ASF).

5.5.3 Cell No. 2

Data on pure hydrogen were acquired at 300°F in the current-density range 0-150 ASF in 25 ASF increments. At each stable current-density, voltage was recorded for hydrogen utilizations of 70%, 80%, and 90%. The following anode feed gas composition was then utilized:

65% H₂ 8% H₂0 1.5% C0 25.5% C0₂

Voltages were recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilizations of 70%, 80%, and 90% at

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each current-density. Pure hydrogen was then rerun as described above. Testing then continued with an anode feed gas of the following composition:

> 65% H₂ 10% H₂0 1.5% C0 23.5% C0₂

Voltages were again recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilizations of 70%, 80%, and 90% at each current-density. Pure hydrogen was then run again as described above. The following anode feed gas was then run:

65% H₂ 12% H₂0 1.5% C0 21.5% C0₂

Voltages were recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilizations of 70%, 80%, and 90% at each current-density. Finally, pure hydrogen was run again as described above.

The temperature was then raised to 325°F, and the entire sequence of events described above was repeated for this temperature.

5.5.4 Cell No. 3

Data on pure hydrogen were acquired at 325° F in the current-density range O-150 ASF in 25 ASF increments. At each stable current-density, voltage was recorded for hydrogen utilizations of 70%, 80%, and 90%. This was followed by an 85% H₂-15% CH₄ gas mixture. Voltages were recorded at current-densities of 0, 25, 50, 75, and 100 ASF with hydrogen utilizations of 70%, 80%, and 90% at each current-density.

This procedure was then repeated for an $85/15 H_2/CH_4$ mixture containing 100 ppm H₂S. This was followed by $85/15 H_2/CH_4$ mixtures with 200 ppm H₂S, 300 ppm H₂S, and 500 ppm H₂S, respectively.

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In each case, voltages were recorded at current-densities of 0, \cdot 25, 50, 75, and 100 ASF with hydrogen utilizations of 70%, 80%, and 90% at each current-density.

5.6 Test Procedure

5.6.1 Stacks la & 2a

5.6.1.1 Fuel

Stack la was operated on a simulated reformer product gas of 65% hydrogen, 2% CO, 10% H₂O, balance CO_2 . Stack 2a was operated on 90% H₂, 10% H₂O.

5.6.1.2 Load Profile

Both stacks were operated on the continuous load profile as shown in Figure 8. The stack operating temperature was varied a minimum of three times during the test. Each temperature change was maintained for 50 hours. At all hours marked X on Figure 8, a current density/voltage curve was obtained from 0-200 amperes per square foot in 25 ASF increments along with hydrogen utilizations of 70, 80 and 90% at each increment. All stack temperatures were recorded for each current density/voltage curve. Minor modifications were made to the load change and data collection sequence in order to accommodate the normal work day.

5.6.2 Stacks 1b & 2b

5.6.2.1 Fuel

Stack 1b was operated on the simulated reformer product gas as used in Stack 1a. Stack 2b was operated on 90% H₂, 10% H₂0.

5.6.2.2 Load Profile

Both stacks were operated on an intermittent load profile as shown in Figure 9. The stack operating temperature was varied in the continuous load profile test except when the stack was shut-down (see below). At all hours marked X on Figure 9, a current density/voltage curve and hydrogen utilizations were obtained as in the continuous load profile test.

5.6.2.3 Start-Up/Shut-Down Procedure

The following procedures were used when starting and stopping the fuel cell stacks. These procedures were designed to simulate the



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CONTINUOUS LOAD PROFILE TEST 3 CELL STACK

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ENCELNARD Ī ł g ş 8 200 (SUUCH) Figure 9 1 804 300 200 ŧ 1 1 4 1 8 ŀ 1 ī 1 Ĩ 1 ł 100 75 (אבאכבאד סד דערב אאדבס פטיינא) 8 . 25



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start and stop event sequence which would occur in a typical field model fuel cell system with an integrated reformer.

Start-Up Procedure:

- A Apply external electrical load (in a field model this load could be internal supplementary starting heaters).
- B Heat fuel cell stack from ambient temperature to approximately 240°F.
- C Open fuel and air exits.
- D Start air flow.
- E Start fuel flow.
- F Allow stack to heat to operating temperature.
- G Adjust load.
- H Control at operating temperature.

Shut-Down Procedure:

- A Leave external electrical load on.
- B Shut off air flow.
- C Turn off heating pads.
- D 1-1/2 minutes after shutting off air flow, shut off fuel flow.
- E Close fuel and air exits.
- F Allow stack to cool to ambient temperature.
- 5.6.3 Stacks 3a, b & c

5.6.3.1 Fuel

Stacks 3a, b & c were operated on 90% H_2 , 10% H_20 .



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5.6.3.2 Load Profile

To evaluate the effects of start/stop cycling only, Stacks 3a, b & c were operated on constant load interrupted by shut-down periods. The start-up/shut-down procedure of Section 5.6.2.3 was followed. Shut-downs were performed at random and were of varying duration.

5.6.4 Stack 3d

5.6.4.1 Fuel

Stack 3d was operated on 90% H₂, 10% H₂O.

5.6.4.2 Load Profile

In order to provide baseline data, fuel cell Stack 3d was operated on constant load with no intentional shut-down or temperature cycling.

5.6.5 <u>Stacks le, 2e & 3e</u>

5.6.5.1 Fuel

During the first 1400 hours of testing, each fuel cell stack was to be operated on a fuel mixture of 90% hydrogen, 10% water. At the completion of 1400 hours of testing, one fuel cell stack was to continue operating on the hydrogen/water mixture, one fuel cell stack was to be operated on 65% H₂, 23% CO₂, 10% H₂O, 2% CO and the third stack was to be operated on 88% H₂, 10% H₂O, 2% CO.

5.6.5.2 Load Profile

The stacks were operated at a continuous load of 150 Amps per square foot. The continuous load was interrupted with complete shut-down/start-up cycles. Cycling frequency was selected to yield a minimum of fifty shut-down cycles during the first 1400 hours of operation (approximately 6 shut-downs per week). The shut-down/start-up procedure of section 5.6.2.3 was followed.

ENGELHARD

6.0 RESULTS AND DISCUSSION

6.1 Parametric Single-Cell Testing

6.1.1 Cell No. 1

The parametric single-cell test results for Cell No.] are shown in Tables 1 and 2 for temperatures of 300° F and 325° F, respectively. These are also shown in Figures 10 through 17.

It should be noted that the performance on "reformate" gas at 300°F was only slightly inferior to that at 325°F. In two cases at 325°F and hydrogen utilizations of 90% substantial penalties were incurred on reformate gas. These occurred at a current-density of 100 Amp/ft² using the 1% and 3% CO gases.

In a few instances running on hydrogen at 90% utilization, stable performance could not be obtained. This occurred only at low current-densities, and the cause is thought to be hydrogen maldistribution as a result of extremely low exit flows under these conditions.

6.1.2 Cell No. 1A

The results for Cell No. 1A are shown in Tables 3 and 4 for temperatures of 300°F and 325°F, respectively. This cell was a repeat of Cell No. 1 except that additional data were acquired during running on baseline hydrogen.

The performance of Cell No. 1A was similar to that of Cell No. 1, but the tolerance of high hydrogen utilizations was more consistent.

The Cell No. 1 and Cell No. 1A test results on "reformate" fuel indicate that the combined effects of CO (to concentrations up to 3%) and hydrogen dilution could be tolerated for hydrogen utilization rates to at least 80%. In fact, it appears that a 90% hydrogen utilization rate can be accommodated - with moderate performance penalties - at least for temporary periods.

It is seen that the effect of CO concentration in the range of 1-3% was not severe. Also, reasonable tolerance of CO was maintained to a temperature as low as $300^{\circ}F$.

6.1.3 Cell No. 2

The results for Cell No. 2 are shown in Tables 5 and 6 for temperatures of 300° F and 325° F, respectively. These are also shown in Figures 18 through 23.

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PARAMETRIC SINGLE-CELL TESTING

<u>CELL NO. 1</u>

T=300°F

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ANODE GAS	<u> </u>	CELL VOLTAGE (MV)						
	0	25	_50_	75	100	125	150 AMP/FT2	
H ₂							607	70
H ₂							606	80
H ₂	959	761	715	680	652	6 26	603	90
0.5% CO	ſ	750	698	657	631			70
65% H ₂ (s < 964 −	750	697	664	635			80
24.5% CO2	970	629	691	651	623			90
H ₂							616	70
H ₂							616	80
H ₂	961	754	711	679	657	635	614	90
1.0% CO]	971	763	708	670	640			70
10% H ₂ 0	٤ 971	749	698	658	630			80
24% CO ₂	971	740	683	642	620			90
H ₂							607	70
н ₂							606	80
H ₂	960	757	712	679	653	625	604	90
2.0% CO	970	752	701	661	631			70
10% H ₂ 0	{	750	695	656	628			80
23% CO ₂	l	7 46	69 0	640	616			90
н ₂							607	70
H ₂							606	80
H ₂	952	761	710	679	651	625	604	90
3.0% CO]	971	755	702	662	629			70
10% H ₂ 0 >	{	749	687	656	624			80
65% H ₂ 22% CO ₂	L	742	685	634	613			90
⁻ γ	•						6 06	70
H ₂							605	80
H ₂	978	735	681	678	651	628	604	90

Table 1

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			C	ELL NO. 1				
<u>T = 325°F</u>								
ANODE GAS	CELL VOLTAGE (MV)							2 H2 UTILIZATION
	0	_25	50	75	100	125	150 AMP/FT ²	
н ₂		774	728	693	665	638	620	70
H2	976	772	726	692	664	637	618	80
H ₂		770	725	689	663	637	614	90
0.5% CO	910	760	710	673	643			70
10% H ₂ 0 L	\sim	747	707	669	642			8 0
24.5% CO2	933	743	681	653	633			90 🖑
H ₂	•						618	70
H ₂							616	80 _
H ₂	925	768	721	686	662	637	614	90
1.0% CO	894	756	708	672	643			70
10% H ₂ 0 (ł	752	704	669	641			80
24% 002	934	734	69 0	643	586			90
H ₂	•						615	70
H ₂							613	80
H ₂	908	-	-	668	659	638	609	90
2.0% CO	904	751	702	664	634			70
65% H2	ζ	747	697	660	628		\$	80
23% CO ₂	L	730	681	636	621		-	90
H ₂	•						616.	70
H ₂							613	80
H ₂	884	-	700	660	653	630	609 ·	90
3.0% CO	941	754	707	666	633			70
10% H ₂ 0	ξ	723	697	656	625		5	80
22% co2	934	703	672	630	576			90
H ₂		773	724	689	660	634	611	70
н ₂		772	723	688	659	633	609	80
H ₂	915	-	693	679	657	635	611	90

PARAMETRIC SINGLE-CELL TESTING

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Table 2

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PARAMETRIC SINGLE -CELL TESTING - CELL No. 1

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Figure 10

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PARAMETRIC SINGLE-CELL TESTING - CELL No. 1

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 Figure 12
PARAMETRIC SINGLE-CELL TESTING - CELL No. 1

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PARAMETRIC SINGLE-CELL TESTING - CELL No. 1

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PARAMETRIC SINGLE · CELL TESTING - CELL No.

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PARAMETRIC SINGLE-CELL TESTING

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CELL NO. 1A

T = 300°F

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ANODE GAS			c	ELL VOLTA	GE (MV)			% H2 UTILIZATION
	0	25	50	<u>75</u>	<u>100</u>	<u>125</u>	150 AMP/FT2	
H ₂	921	763	718	685	655	629	604	70
H ₂	927	763	717	684	655	628	604	80
н2	928	762	717	684	655	629	605	90
0.5% CO	930	756	705	670	642			70
10% H ₂ 0 (65% H ₂ (> < 928	750	699	668	641			80
24.5% CO ₂	926	743	6 91	659	632			90
н ₂	926	760	715	683	655	631	605	70
н ₂	926	765	718	686	657	632	607	80
^н 2	926	763	718	685	658	632	607	9 0
1.0% CO	924	748	699	663	635			70
10% H ₂ 0 (923	744	695	664	63)			80
24% CO2	924	749	687	654	626			90
н ₂	932	761	716	683	656	631	608	70
H ₂	932	764	717	685	657	633	609	80
н ₂	932	762	715	680	654	630	607	90
2.0% CO	928	749	699	663	634			70
10% H ₂ 0 65% H ₂ }	\$ 928	749	695	659	630			80
23% CO2	928	744	694	649	622			90
H ₂	931	760	715	682	654	631	608	70
н2	931	765	718	6 86	657	633	610	80
H ₂	931	763	718	683	655	632	608	90
3.0% CO	926	747	696	660	631			70
65% H ₂ 0	\$ 925	743	693	656	628			80
22% co ₂	928	735	687	647	615			90
H ₂	935	762	715	683	655	629	607	70
H ₂	935	762	715	682	655	630	607	80
н2	937	762	716	683	656	630	608	90



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<u>T = 325°</u>F

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PARAMETRIC SINGLE-CELL TESTING

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CELL NO 1A

AHODE GAS	<u> </u>	CELL VOLTAGE (MV)							
	0	_25	50		<u>100</u>	125	150 AMP/FT2		
H ₂	934	771	725	693	6 66	640	618	70	
н ₂	9 10	761	718	687	66 0	635	614	80	
H ₂	929	766	722	69 0	665	640	617	90	
0.55 CO 105 H20 655 H2 24.55 CO2	930 923 921	766 751 745	712 703 698	677 664 ช64	650 643 642			70 80 90	
н2	9 20	761	717	686	660	641	619	70	
H ₂	932	768	723	691	666	641	62 0	80	
H2	933	769	724	692	665	642	62 3	90	
1.0% CO 10% H ₂ O 65% H ₂ 24% CO ₂	> { 931 928 928 928	755 748 753	708 704 701	676 669 661	649 643 637			70 80 90	
Hi2	924	769	722	691	665	641	618	70	
н2	930	765	722	689	665	64 0	620	80	
H2	934	768	722	691	6 66	642	620	90	
2.0% CO 10% H2O 65% H2 23% CO2	> { 928 928 927	752 749 737	706 702 686	672 670 656	645 641 628			70 80 90	
H ₂	9 26	768	723	6 92	667	642	620	70	
H ₂	936	767	724	692	666	642	620	80	
Н ₂	935	768	722	692	665	642	620	90	
$\begin{array}{c} 3.0\% \text{ CO} \\ 10\% \text{ H}_20 \\ 65\% \text{ H}_2 \\ 22\% \text{ CO}_2 \end{array}$	≥ { 928 928 924	753 748 732	705 697 690	672 664 654	642 638 621			70 80 90	
H ₂	925	770	723	691	664	640	618	70	
Н ₂	933	771	724	692	6 65	641	618	80	
H2	933	76 8	722	69 0	664	640	618	90	

Table 4

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$T = 300^{\circ}F$								
ANODE GAS			<u></u>	CELL VOLT	AGE (MV)			# H2 UTILIZATION
	0	_25	50	75	100	125	150 AMP/FT2	
H2	938	753	709	676	651	628	607	70
н ₂	943	759	714	682	656	633	612	80
H ₂	942	758	714	683	657	634	612	90
1.5% CO	935	748	700	666	639			70
8% H ₂ 0	935	743	697	660	62 9			80
66.5% H2	935	736	688	642	6 16			90
24% CO2								
H ₂	943	760	716	684	658	636	614	70
H ₂	941	763	717	685	659	636	614	80
H ₂	943	760	715	683	658	636	614	90
1.5% CO	942	748	699	664	634			70
105 H ₂ 0	×< ⁹³⁶	742	692	6 60	6 28			80
65≾ Н ₂	936	741	685	651	617			90
23.5% CO2								
H2	943	762	716	684	656	636	614	70
H ₂	942	763	718	6 86	660	636	614	80
H ₂	942	760	716	684	658	634	614	90
1.5% CO	940	747	696	66 0	630			70
12% H20	937	739	696	660	633			80
63.5% H ₂	937	735	680	649	616			90
23% CO ₂								
Н ₂	942	760	715	683	656	633	612	70
н ₂	94 0	759	714	682	657	632	612	80
н ₂	940	758	713	682	655	632	612	90

PARAMETRIC SINGLE-CELL TESTING CELL NO. 2

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Table 5

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				CELL NO.	2			
<u>T = 325°F</u>								
ANODE GAS				CELL VOLT	AGE (MV)			1 H2 UTILIZATION
	0	25	_50_	75	<u>100</u>	125	150 AMP/FT2	
H2	937	769	725	696	670	648	626	70
H ₂	937	771	728	698	671	649	627	80
H ₂	937	767	726	695	67 0	64 8	627	90
1.5% C0 8% H ₂ O 66.5% H ₂ 24% CO ₂	934 934 934 934	754 749 736	709 704 694	674 668 657	646 641 627			70 80 90
H2	942	768	724	693	668	647	625	70
н ₂	939	767	725	694	668	647	627	80
H ₂	940	766	724	693	667	646	626	90
1.5% C0 10% H20 65% H2 23.5% C02	<pre> 935 935 935 935 </pre>	752 750 742	708 705 694	676 672 662	647 643 630			70 80 90
H ₂	942	768	725	695	672	647	625	70
н ₂	944	768	725	694	669	646	6 26	80
н ₂	945	767	724	695	670	648	628	90
1.5% C0 12% H20 53.5% H2 23% C02	930 935 935 935	748 747 746	702 704 700	672 672 658	648 640 629			70 80 90
H2	940	762	719	688	665	643	624	70
H2	942	765	722	690	667	643	622	80
H ₂	943	766	721	692	665	641	622	90

PARAMETRIC SINGLE-CELL TESTING

Table 6

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Performance was not strongly influenced by H₂O content in

the range of 8-12%. The performance at 325°F was generally 10-15 mV higher than that at 300°F.

6.1.4 Cell No. 3

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The results for Cell No. 3 are shown in Table 7. The data indicate no clear effect of H₂S in the 0-500 ppm range. At high hydrogen-utilizations the dilution effect of CH₄ in 85/15 H₂/CH₄ mixtures far outweighs any possible effect from added H₂S.

6.2 Three-Cell Stack Testing

6.2.1 Stack la

Figure 24 (see note below) shows the performance history of Stack la. The initial performance decline of Stack la was primarily attributable to the decline of the top and bottom cells.

At approximately 42 days into the test, the top cell was observed to decline more rapdily than the lower two cells. Open circuit readings taken at the 50 day point showed a depression in the top cell's open circuit, 0.788 volts versus 0.838 and 0.825 for the middle and bottom cells respectively.

6.2.2 Stack 2a

Figure 25 shows the performance history of Stack 2a.

Testing of Stack 2a was terminated after 696 hours of load profile testing (744 hours total load time) due to an over-temperature failure. The over temperature condition was caused by mixing of fuel and air within the inlet manifolds. Causes for the mixing were creep of the hydrogen manifold gasket and improper original positioning of this gasket.

Note: Termination plate/current collector plate interface

To provide electrical contact between the carbon termination plates and the aluminum current collector plates an expanded copper mesh was employed in the preliminary stacks. Dissimilar metal corrosion between the copper and aluminum caused excessively high resistive losses at these interfaces. Examples of these losses are shown in Figures 24 and 25. The high resistive losses were reduced by copper plating the current collector plates and using a sheet of Grafoil between the collector and termination plate. Stacks la and 2a were rebuilt in this manner. All subsequent stacks employed this construction.

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PARAMETRIC SINGLE-CELL TESTING

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CELL NO. 3

<u>1 = 325°F</u>								
ANODE GAS				CELL VOLTA	GE (MV)			1 H2 UTILIZATION
	0	25	50	_75_	100	125	150 AMP/FT2	
H ₂	926	763	715	6 85	658	634	610	70
H ₂	926	754	711	684	658	634	612	80
H ₂	928	744	704	6 81	651	632	6 10	90
85°. H2	927	753	707	677	647			70
15% CH4 5	\$ 927 }	746	702	760	651			80
)	928	732	671	658	628			90
85% H2	927	752	701	676	64 8			70
-100 PPM H25	328	739	697	668	642			80
BAL. CH4	928	737	693	654	632			90
85: H ₂	928	756	707	674	648			70
200 PPM H25	1929	733	698	670	643			80
BAL. CH4	930	722	689	665	629			90
851 H	(124	753	705	674	648			70
- 300 PPM H25	۹31 -	715	696	669	644			80
BAL. CH4	430	693	667	64 3	639			90
85 H2	929	746	706	673	645			70
400 PPM 135	ζ <u>931</u>	719	694	667	641			80
BAL, CH4	$\sum_{i=1}^{31}$	714	647	652	617			90
851 H2	(929	752	704	674	649			70
500 PPM 4-55	1933	748	647	6 69	643			80
BAL CH4		729	640	645	633			90

Table 7

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100 ASF 125 ASF 83 ASF 57 ASF 26 ASF 56 ASF 2.2 . 25% D.P. 2.0 49% D.P. 1.8 50% D.P. 68% D.P. 74% 99% D.P. DESIGN POWER (D.P.): 91 WATTS 1.6 83% D.P. TERMINATION PLATE VOLTAGE 814CK VOLTAGE CURRENT COLLECTING PLATE VOLTAGE FUEL: 90% H₂, 10% H₂0 80% H₂ UTILIZATION OXIDANT: AIR TEMPERATURE: 300°F * OXYGEN UTILIZATION DECREASED 0.8 ** OXYGEN UTILIZATION RETURNED TO 33% MERADCOM 3-CELL STACK 2a 0.6 PERFORMANCE-TIME 0.4 0.2 0 8 10 12 14 16 18 2 4 6 0 TIME (DAYS) -45-Figure 25

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Prior to failure Stack 2a operated nominally. One day before failure, stack open circuit voltage was 2.672 volts. Individual cell voltages at 125 ASF, 300°F were 0.591, 0.590 and 0.600 for the top, middle and bottom cells respectively.

6.2.3 Stack 2b

Figure 26 shows the performance history of Stack 2b. Figure 27 shows the open circuit voltage history of Stack 2b.

The performance decline in Stack 2b was characterized by an accompanying decline in open circuit voltage, see Figure 27 Stack 2b - Open Circuit vs Time.

The extreme decline observed during the final two days of testing was primarily caused by the bottom cell. On day thirty, the bottom cell had declined to 0.263 volts at 125 ASF, 350°F as opposed to 0.601 and 0.565 respectively for the top and middle cells. Open circuit voltages of all cells were unacceptably low at the thirty day point with the bottom cell having the lowest voltage. Open circuit voltages at this point were: top cell 0.768, middle cell 0.726, bottom cell 0.583.

6.2.4 Stack 1b

Figure 28 shows the performance history of Stack lb. Figure 29 shows the open circuit voltage history of Stack lb.

As in Stack 2b, performance declines were accompanied by declines in open circuit voltage. See Figure 29, Stack 1b. Open Circuit Voltage vs. Time. Decline was uniform across the three cells. Final open circuit voltages (24 days) were: top cell 0.729 volts, middle cell 0.722 volts, bottom cell 0.738 volts.

At 24 days into the test, the temperature controller for Stack lb failed in the on position causing gross overheating of the stack. As a result, no further testing was possible.

6.2.5 Stack 3a

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Figure 30 shows the performance history of Stack 2a.

Stack 3a was the first stack specifically devoted to investigation of start-up/shut-down effects. It can be seen in Figure 30 that overall stack performance declines were accompanied by

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MERADCOM 3-CELL STACK 2b

PERFORMANCE-TIME



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losses in individual cell open circuit voltages. Two shut-downs (Nos. 9 and 10) were conducted by keeping the stack at operating temperature under an inert gas blanket. No change in the stack decline trends was observed as a result of this shut-down procedure.

Because it was felt that the loss of open circuit voltage might be related to reactant cross leakage caused by breaching of the electrolyte matrix due to acid loss, samples of the electrodes and matrix were titrated for acid content after termination of the run. Results of the titrations are given in Table 8.

Each of the three cells shows a total acid inventory approximately equal to that of a fresh matrix, suggesting that acid loss from the overall cell laminate package was not severe although a nominal amount of acid was transferred from the matrix to the electrodes. Acid loss from localized areas of the matrices which would not be detected by these titrations might have caused the open circuit voltage losses.

6.2.6 Stack 3b

Figure 31 shows the performance history of the individual cells of Stack 3b.

Because the previous stack testing indicated that poor shut-down tolerance was caused by breaching of the matrix through acid loss, Stack 3b was constructed to incorporate a higher acid inventory. The top cell of Stack 3b employed a standard matrix; however, the electrodes were prefilled with acid to a level typical of that observed in cells which had operated for one thousand hours. The lower two cells each employed matrices of almost twice the normal thickness containing twice the acid volume of a standard matrix.

Initially, the performance of all three cells declined at a rate similar to that observed in previous stacks which were subjected to shut-down cycling. At approximately the 225 hour point, the decline in performance of the center cell stopped and this cell ran stably for the final 400 hours of the test. The fact that the open circuit voltage of the middle cell remained high throughout the test indicates that no substantial breaching of the matrix occurred.

Testing of Stack 3b was terminated at 625 hours due to the inability of the top and bottom cells to support a load.

6.2.7 Stack 3c

Figure 32 shows the performance history of the individual cells of Stack 3c.

PHOSPHORIC ACID CONTENT OF FUEL CELL STACK

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COMPONENTS AFTER RUN NO. 3a

		mg. Bhos. Acid/ cm ² of Matrix*	% Volume **
	Matrix & Electrodes	35.5	37.4%
TOP CELL	Matrix	23.2	24.5%
	Electrodes	12.3	-
	Matrix & Electrodes	37.1	39.1%
MIDDLE CELL	Matrix	28.1	29.5%
	Electrodes	9.0	-
	Matrix & Electrodes	40.2	42.1%
BOTTOM CELL	Matríx	23.4	24.6%
	Electrodes	16.8	-

* Std. Matrix, 0.05 cm thick

** Assuming Phos. Acid density of 1.9 mg/1 ; based on matrix volume.

Table 8

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In Stack 3c, all three cells were duplicates of the middle cell of Stack 3b (see Results and Discussion, Stack 3b).

It can be noted from Figure 32 that the performance of the top cell initially declined at a greater rate than did the performance of the lower two cells. As in previous tests, the performance decline of the top cell was accompanied with a corresponding decline in open circuit voltage.

There was a more gradual depression of open circuit voltages of the lower two cells, and their performance was not significantly affected by the shut-down cycling.

Upon disassembly, a small burn mark indicative of breaching of the matrix was observed on the top cell.

6.2.8 Stack 3d

Stack 3d was constructed to be identical with Stack 3c. Stack 3d was operated under constant load to provide a comparison with stacks undergoing start-up/shut-down cycling.

Stack 3d experienced three thermal upsets during the test. During the first day of operation a temperature control failure caused the stack temperature to decline to approximately 200°F.

At 210 hours, the temperature controller cycled the stack from operating temperature to lower temperatures for a period of about 8 hours. The lowest temperature observed by automatic recording equipment during this period was 172°F.

At 830 hours, the stack temperature controller failed in the off position permitting the stack to cool for a period of approximately 6 hours. When this fault was discovered, the stack operating temperature was observed to be 118°F (approximately the temperature of the hydrogen saturator).

The results of testing of Stack 3d are shown in Figure 33. All cells performed with minor degradation until the temperature controller malfunction occurred at the 830 hour point. After this malfunction, the middle cell continued to perform stably; however, the top and bottom cells suffered permanent performance losses. These performance losses were probably due to acid loss from the matrix caused by the build-up of product water within the cells at the low operating temperature. Upon disassembly, a burn mark, indicating breaching of the matrix, was found on the lower cell.




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6.2.9 Stack le

The performance history of Stack le is shown in Figure 34. The individual cell performance history, starting with the twenty-fifth day of operation is shown in Figure 35. Individual cell volt-ampere curves taken at 10 days, 50 days, and 100 days of operation are shown in Figure 36.

On approximately the twenty-fifth day of operation, the voltage under load of Stack le was observed to decline. This performance decline was caused solely by a decrease in voltage of the upper cell (see Figure 35).

It was observed that the phosphoric acid on the acid addition "shelf" of the top cell had turned blue, indicating that this acid was in communication with the copper plating of the termination plate/current collector plate interface. To reduce the possibility of similar contamination of the lower cells, acid addition to the upper cell was stopped.

On the 118th day of operation, the fuel of Stack le was changed from 90% H₂, 10% H₂O to 88% H₂, 10% H₂O, 2% CO. Operation was not continued on this fuel mixture due to the low performance of the upper cell.

The temperature control thermocouple failed open on the 130th day of operation permitting the stack to cool to approximately 110°F with the reactants flowing. Upon restoring the stack to operating temperature, a definite loss of performance of all three cells was observed (see Figure 35).

6.2.10 Stack 2e

The performance history of Stack 2e is shown in Figure 37. Individual cell volt-ampere curves taken at 10 days, 50 days, and 100 days of operation are shown in Figure 38.

Stack 2e was operated on the 90% H₂, 10% H₂O fuel mix. Between the 45th and 65th days of operation, the stack exhibited erratic performance accompanied by an increased sensitivity to air flow. Because air sensitivity can be caused by manifold leakage, the air inlet manifold was replaced on the 66th day. After replacement of the manifold, performance of the stack stabilized; however, some air sensitivity remained.

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Figure 34

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0.7 CELL VOLTAGE @ 150 asf 177 7171 0.6 Toṗ Cell 0.5 Middle Cell Bottom Cell 0.4 0.3 25 40 30 35 45 50 60 55 65 70 75 80 85 DAYS ON TEST MERADCOM 3 CELL STACK 1e H₂-AIR, 350°F

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Figure 35



Figure 35



STACK VOLTAGE - VOLTS

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Figure 37



STACK VOLTAGE - VOLTS

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6.2.11 Stack 3e

The performance history of Stack 3e is shown in Figure 39. Individual cell voltages under load beginning with 60th day of operation are shown in Figure 40. Voltage-current relationships taken on the 10th, 50th, C3rd, and 100th days of operation are shown in Figure 41.

On the 61st day of operation, Stack 3e was switched to operation on the 65% H₂, 23% CO₂, 10% H₂O, 2% CO fuel mixture. During the 12 days of operation on this fuel (see Figure 40), erratic performance was observed in the lower two cells. The upper cell, however, remained stable in performance. The stack was returned to operation on 90% H₂, 10% H₂O and diagnostic tests were performed to determine whether the decline in performance of the lower cells was due to poor flow distribution or CO effects. The results of this testing are shown in Table 9.

The two fuel streams that did not contain CO did not show any definite trends in decline of individual cell performance. Operation on the two CO containing streams, however, showed declines similar to those observed earlier. In both cases, the bottom cell showed the greatest performance loss, followed by the center cell. The upper cell showed negligible or zero loss. These results indicate that the decline in performance resulted primarily from decreased tolerance to CO effects rather than from unequal flow distribution or hydrogen partial-pressure sensitivity.

Because CO tolerance can be a function of cell operating temperature, temperature profiles of the three cells were taken. Figures 42 through 44 in the appendix show in-plane temperature profiles of the cells when operating on pure hydrogen. Figures 45 through 47 in the appendix show the in-plane temperature profiles of the same cells operating on the 65% H₂, 23% CO₂, 10% H₂O, 2% CO mix. It should be noted that these measurements are not true "center-of-the-cell" measurements but measurements taken between the cell cathode and the adjacent, lower bipolar plate. Figure 48 in the appendix shows diagrammatically the location of the thermocouple traverses.

The six thermal profiles show that for both fuels, the temperature profiles of the top and center cells are very similar, differing by approximately 1°F. The lower cell temperature profiles are approximately 5°F below those of the upper two. To determine if the temperature profiles might have been influenced by unequal



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Figure 39





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3-Cell Stack 3-e

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Diagnostic Testing Summary

Hydrogen/≈150 ASF

Utilization -	70%	80%	<u>90%</u>	△mv
Top cell Middle cell Bottom cell	610 mv 590 596	606 590 595	566 563 578	44 27 18
65% H ₂ , 23% CO ₂ , 10% H ₂ O, 2	% CO/≈125A	SF		
Utilization (approx.)	<u>50%</u>	55%	60%	<u>s mv</u>
Top cell Middle cell Bottom cell	598 mv 513 423	598 508 415	598 501 406	0 12 17
65% H ₂ , 25% N ₂ , 10% H ₂ 0/ \approx 1	25 ASF			
Utilization (approx.)	<u>70%</u>	80%	90%	<u>∧ mv</u>
Top cell Middle cell Bottom cell	584 mv 577 570	582 564 571	570 551 552	14 26 18
88% H ₂ , 10% H ₂ 0, 2% CO/≈12	25 ASF			
Utilization (approx.)	<u>55%</u>	<u>70%</u>	<u>75%</u>	<u> <u> </u> <u></u></u>
Top cell Middle cell Bottom cell	602 mv 560 557	596 546 521	591 536 506	11 24 51

Table 9

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operation of the end plate heating pads, the power consumption of each pad was checked. The upper and lower heating pads were found to be operating equally.

Temperature differences can contribute to CO tolerance effects. It is doubtful, however, that the small temperature differences observed were responsible for the reduced performance of the center and bottom cell.

6.3 2-KW Fuel Cell Stack Design

A design for a 2-KW, liquid cooled fuel cell stack was developed as part of this effort. The fuel cell stack design had the following characteristics:

No. of cell	62
Cell active area	6" x 9 3/4" (0.406 ft ²)
Overall cell dimension	7" x 10.688"
No. of cooling plates	8
Cooling plate frequency	every 8 cells
Bare stack dimension (excluding manifolds and bolting)	7" x 10.688 x 15.73 h
Overall stack dimensions	10" x 12 3/4" x 18 3/4" h
Bare stack weight	55.5 lbs.
Overall stack weight	78.1 lbs.

The test stacks operated during this program employed a heavy, laboratory type bolting system. The overall weights and volumes given above are based on a conceptual design utilizing honeycomb stiffened, aluminum end plates.

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7.0 CONCLUSIONS

The initial 3-cell stack testing demonstrated conclusively that the primary performance losses associated with start-up/shut-down cycling were due to breaching of the electrolyte matrix. The testing of 3-cell Stacks 1, 2 & 3e showed that start-up/shut-down cycling losses would be eliminated or greatly reduced by the use of a matrix having improved acid transport characteristics combined with regular acid addition. Although the original cause of rapid cell decay associated with shut-down cycling appears to be eliminated, some long term decay in all three stacks was observed. This decay was greater than that observed in single-cells and stacks operated for similar periods of time but without shut-down cycling.

Figures 36, 38 and 41 show the voltage-current relationships of the three stacks taken after 10, 50 and 100 days of operation. Individual cell voltages at 150 ASF taken at these times are given in the figures. In all three stacks, over half the decay seen over the ninety day period was due to decay of one of the three cells. The center cell of Stack le was exceptionally stable showing only a 2 mV loss after 2400 hours of testing. In general, the changes in offset and slope of the V-A curves with time indicate that the decline in stack performance was due to both decreased catalyst performance and electrode flooding. It can be seen that between the 50 and 100 day lines of Stack le and between the 10 and 50 day lines of Stack 3e only a change in offset is present. This indicates that the performance declines observed during these periods were due primarily to catalyst activity losses. All other V-A curve "pairs" show change in offset and slope.

The inability of Stacks le and 3e to operate stably on CO-containing fuel streams after approximately 1400 hours of operation on pure hydrogen was traced to diminished anode catalyst activity, with poisons other the CO suspected of playing a role. The center cell of Stack 3e showed a nominal performance loss associated with operation on CO-containing fuel streams, indicating that the factors contributing to decreased CO tolerance did not affect all cells equally.

Initial cell stacks had high voltage losses associated with corrosion of the current collector/termination plate interface. These losses were reduced by copper plating the current collector plates and interposing a sheet of graphite foil between the current collector plates and termination plates.

A 2 kilowatt, liquid cooled, fuel cell stack was designed based on the components utilized in the three cell stack testing program. A conceptual design of a lightweight bolting system was prepared to permit an overall stack weight consistent with the requirements α f portable field equipment.

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8.0 RECOMMENDATIONS

- 1. Additional investigations should be performanced to determine the effects of carbon monoxide containing fuel streams on fuel cell stacks subjected to multiple start-up/shut-down cycling.
- 2. Methods of improving CO-tolerance of fuel cell anodes should be investigated. This would include higher-temperature operation and anode composition modifications.
- 3. If carbon monoxide effects are shown to be deleterious to long term fuel cell performance, methods of elimination (or partial elimination) of carbon monoxide from the fuel stream should be investigated.
- 4. The start-up/shut-down testing of Stacks le, 2e and 3e utilized electrodes of similar hydrophobicity. The effects of high wet-proofing levels on start-up/shut-down tolerance should be investigated.
- 5. In Stacks le, 2e and 3e acid inventory was maintained by external, manual acid addition. Similar testing should be conducted on cell stacks incorporating internal acid storage.
- 6. The bipolar plates used in the three-cell stack testing phase of this program showed excellent corrosion resistance and dimensional stability under cell operating conditions. The fabrication costs, however, are presently too high for commercial use. Alternative bipolar plate construction and fabrication methods are presently being investigated. Such efforts should continue in order to attain cost-effective bipolar plate structures.



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APPENDIX

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THERMAL PROFILE TESTING



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