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(DE85001783)

DEVELOPMENT OF MOLTEN CARBONATE FUEL CELL
POWER PLANT TECHNOLOGY

Quarterly Technical Progress Report for the Period
October 1—December 31, 1980

Work Performed Under Contract No. AC02-79ET15440

United Technologies Corporation
South Windsor, Connecticut

Technical Information Center
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United States Department of Energy

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DEVELOPMENT OF MOLTEN CARBONATE FUEL CELL
POWER PLANT TECHNOLOGY

Quarterly Technical Progress Report No. 5
for Period October 1, 1980 to December 31, 1980

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United Technologies Corporation
P. O. Box 109
South Windsor, Connecticut 06074

Prepared for

DEPARTMENT OF ENERGY
UNDER CONTRACT DE-AC01-79ET15440

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FOREWORD

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ABSTRACT

This report summarizes the work performed under U.S. Department of Energy Contract No. DE-AC01-79ET15440 during the fifth quarterly reporting period from October 11, 1980 through December 31, 1980. The overall objective of this 29-month program is to develop and verify the design of a prototype molten carbonate fuel cell stack which meets the requirements of a 1990's-competitive coal-fired electrical utility central station or industrial cogeneration power plants.

During this quarter, activity continued in all four task areas: Task 1 - system studies to define the reference power plant design; Task 2 - cell and stack design, development and verification; Task 3 - preparation for fabrication and testing of the full-scale prototype stack; and Task 4 - development of the capability to operate stacks on coal-derived gas.

TASK 1

- o The first User Review Panel assessed power plant technical and economic design requirements.
- o Characterization of fuel processing subsystems was completed by Bechtel.
- o A matrix of gasifier/gas cleanup combinations was selected for overall power plant evaluations.
- o Studies were initiated to determine the effect of part-load operation on the fuel cell stack and design requirements.

TASK 2

- o A 4-in. by 4-in. subscale cell with an improved edge configuration has maintained greater than 10-psi cross-pressure through four thermal cycles.
- o Bench-scale cell tests of anodes with improved creep strength and dimensional stability show improved performance stability during thermal stress tests.
- o Furnace modifications and carrier mold plate evaluations for scaled-up electrode fabrication are continuing.
- o A second alternative nickel oxide cathode single cell is performing stably. Post test analysis of the prior alternative cathode cell suggests that there is a contact resistance problem between the cathode and housing.
- o Scaleup to one-square-foot has been demonstrated for the alternative nickel oxide cathode.
- o An evaluation of cathodes made from Inco 255 nickel was initiated.

- o Full-size separator plate samples were fabricated by laser welding.
- o A set of dielectric pipe connectors was modified to minimize adverse thermal growth stresses.
- o Design of the short stack of full-scale cells was started.
- o Thermal analysis of the power penetrator shows compliance with electrical code requirements.
- o The construction drawings for the 150-psia pressure vessel for the test of subscale stacks were completed.

TASK 3

- o Procurement of fabrication equipment for forming matrices up to 48-in. wide was initiated. Facilities for the fabrication line are under construction.

TASK 4

- o The value of the equilibrium constant for the reaction of H_2S with molten carbonate was determined for a range of temperatures and electrolyte compositions.
- o Tests to determine the effects of 0.005 ppm SO_2 and 0.05 ppm H_2S on cell performance were completed.
- o Hardware for the sulfur parametric study was fabricated and is available for cell testing.

EXECUTIVE SUMMARY

TASK 1 - ESTABLISH POWER PLANT REFERENCE DESIGN

The first User Review Panel was convened to assess power plant technical and economic design requirements derived by UTC and Bechtel. Central station power plants for both baseload and intermediate duty cycle applications were considered. The panel provided a range of utility concerns and needs, as well as an evaluation of the study findings. Their recommendation and suggestions are being factored into UTC's and Bechtel's work.

Bechtel completed characterization of fuel processing subsystems. In all 11 different gasifiers were characterized. The gasifiers were evaluated and ranked in terms of several parameters affecting their potential attractiveness for integration into molten carbonate power plant concepts. Bechtel reviewed their data on gas cleanup processes; no revisions were necessary.

A matrix of gasifier/gas cleanup matings has been selected for overall power plant evaluations. Effort by Bechtel is continuing to further screen these matings and derive the most favorable mating of a gas cleanup process with each of the selected gasifiers.

Studies were initiated to determine the effect of part-load operation on the fuel cell stack and design requirements. A mode of power plant control and operation for turndown was defined and the resulting interface operating conditions for the cell stacks and module were defined.

TASK 2 - DEVELOP AND VERIFY CELL AND STACK DESIGN

Testing of three configurations of the improved cross-pressure tape-matrix in 4-in. by 4-in. subscale cells continued. One of these configurations has maintained greater than 10-psi cross-pressure through four thermal cycles.

Anode development activity concentrated on improving dimensional stability. A variety of configurations, materials, and density variations are being tested. Initial results indicate that densification and heat treatment can improve anode dimensional stability. Evaluation of alternative nickel oxide cathodes was the primary activity in cathode development. Performance of a 4-in. by 4-in. single cell containing an alternative cathode is stable after two temperature cycles. A dry casting process was used to fabricate this cathode. This process was also used to fabricate larger area nickel oxide cathodes, but cracking has occurred during lithiation.

A second 4-in. by 4-in. single cell evaluation of an alternative nickel oxide cathode (Cell 294) is at 667 mV at 160 amps/ft² after two temperature cycles to room temperature. This component was fabricated using a dry casting process.

Measurement of the post-test conductivity of the alternative lithiated nickel oxide cathode from Cell 281 indicates that about half of the cathode side resistance loss of the cell can be attributed to contact resistance between the cathode and the cell housing. Flexural and crush strengths were lower than pre-test values, although scanning electron microscope examination showed no obvious structural degradation.

A dry casting process was used to fabricate alternative nickel oxide cathodes in areas up to one square foot. However, lithiation of one large cathode resulted in multiple fractures.

Weld trials of 3-ft. by 3-ft. separator plates showed that it is feasible to scale the present plate configuration. Tooling requirements and alternative welding techniques were identified. Local aluminum dispersion by laser deposition was verified as an acceptable aluminizing method.

Corrosion tests are being conducted in the electrolyte on various candidate materials for cathodes, separator plates, electrolyte matrix fillers, manifold seals, and stack electrical insulators. Super-FECRALLOY continues to exhibit acceptable corrosion resistance in anode and cathode environments. Analysis of 316-SS oxide films show a dual layer structure, the outer layer being Fe_2O_3 and the inner layer Cr_2O_3 .

Exposure of an alternative nickel oxide cathode structure to electrolyte at 1400°F cathode conditions for 1000 hours resulted in little change in compressive strength, but a noticeable decrease in flexural strength.

The dielectric pipe connector being prepared for subscale stack test evaluation in a parallel program successfully completed a thermal cycle. A test rig for evaluating the leakage and mechanical properties of the manifold seal was obtained. It is being installed in an existing test stand.

Specimens of nickel coated Glidcop bus bar material were prepared for evaluation in a subscale stack test being run under a parallel program. A 4-in. by 4-in. manifold seal trial survived two thermal cycles with no leakage problems.

Dielectric tests of the best potential manifold coating to date were repeated with a fully coated sample. The objective was to eliminate the possible leakage path which may have caused the failure of the previous partially coated samples. The new data showed even higher conductivities, and manufacturing differences in the samples are under investigation to explain this unexpected result.

Design of the short stack of full-scale cells was started. The full-scale short stack pressure vessel was placed on order.

TASK 3 - DEVELOP CAPABILITY FOR FULL-SCALE STACK TESTS

Subscale fabrication trials on available equipment have been sufficiently successful to warrant procurement of larger equipment for fabricating full-size matrices. An appropriate unit has been ordered, with delivery scheduled for early March. Layout of the process line and the design of ancillary equipment are underway. Completion is scheduled to coincide with the arrival of the purchased equipment.

TASK 4 - DEVELOP CAPABILITY FOR OPERATION ON COAL-DERIVED GAS

Tests designed to determine the effects of 0.005 ppm SO_2 and 0.05 ppm H_2S on cell performance were completed. Hardware for the sulfur parametric study was fabricated and is available for cell testing.

Also, the value of the equilibrium constant for the reaction of H_2S with molten carbonate was determined over a range of temperatures and electrolyte compositions.

INTRODUCTION

This report summarizes the work performed at the Power Systems Division of United Technologies under Department of Energy Contract No. DE-AC01-79ET15440 "Development of Molten Carbonate Fuel Cell Power Plant Technology" from October 11, 1980 through December 31, 1980. This work is separated into four tasks:

TASK 1 - ESTABLISH POWER PLANT REFERENCE DESIGN

This task defines a superior 1990 coal-fired electrical utility central station or industrial cogeneration power plant reference design to guide the cell design and development activities in Task 2.

A broad range of subsystem and power plant alternatives will be screened against application requirements and user evaluation criteria which will be determined as part of the task effort. Participation of Bechtel and a User Review Panel insures that a competitive design will be established in this task.

TASK 2 - DEVELOP AND VERIFY CELL AND STACK DESIGN

This task establishes the detailed design of a full-scale stack which meets the functional requirements of the reference power plant design selected in Task 1. Under this task, the cell and stack components required for this design are also developed. This effort culminates in the verification of the stack design by a test of a short stack of full-scale cells.

TASK 3 - DEVELOP CAPABILITY FOR FULL-SCALE STACK TESTS

This task provides the capability for manufacturing and definition of the facilities required for testing full-scale cells and the prototype stack. Task efforts include establishing a low cost manufacturing process for the electrolyte matrix based on a tape forming process, providing test plans for the prototype stack, and providing a plan for acquisition and construction of long lead time equipment for a test of the full-scale prototype stack.

TASK 4 - DEVELOP CAPABILITY FOR OPERATION OF STACKS ON COAL-DERIVED GAS

This task determines the mechanisms by which contaminants passing through the fuel gas cleanup system and those in the cathode air supply may affect the molten carbonate fuel cell. A mathematical model predicting the impact of contaminants over 40,000 hours will be developed. This model will permit definition of the impact of gas composition on cell performance and will be used to establish the cleanup system specification. The operability of molten carbonate fuel cells on coal-derived gas will be demonstrated using a mobile test unit for endurance tests of single cells at gasifier sites.

Section 1

TASK 1 - ESTABLISH POWER PLANT REFERENCE DESIGN

1.A SUMMARY

Work continued in Activity 101, power plant application requirements, and in Activities 111 and 113, fuel cell module and stack design requirements. Work was completed in Activity 102, power plant subsystem characterizations. Work was initiated in Activity 103, power plant conceptual design derivations.

Activity 101

During this quarter the first User Review Panel (URP) was convened. The URP reviewed the preliminary power plant technical and economic requirements derived jointly by UTC and Bechtel. Requirements for utility central station power plants in both baseload and intermediate duty cycle applications were presented. The URP agreed with the requirements in general but did make some suggestions for change. They also noted that the power plant's modular nature would alleviate utility problems in meeting load growth and securing expansion capital, and that its exhaust characteristics would aid in meeting environmental requirements. They recommended that the planned evaluation studies use a broader range of and higher coal costs in sensitivity studies and that lower capital charge factors be included in the studies.

Activity 102

All characterization of subsystem for selection of the options to be included in the feasible power plant concepts (Activity 103) has been completed. Bechtel's final submittals under this activity included revision and expansion of the list of gasifiers to include the Babcock-Wilcox entrained flow unit, bringing the total to number of 11 different gasifiers to eleven. These cover the full range of bed types, oxidant feed, and operating pressures. The gasifiers were evaluated in terms of several parameters affecting their potential attractiveness for integration

into molten carbonate power plant concepts. Bechtel reviewed their data on the six selected gas cleanup processes and no revisions were necessary. This effort completed activity 102.

Activity 103

Derivations of overall power plant concepts from the alternative subsystems, and subsequent evaluation of these concepts in Activity 103 and 104 provide the basis for selecting those candidate concepts to be refined in Subtask 1.3. Eight gasifier/gas cleanup combinations have been selected for overall power plant evaluations in this activity. The matrix includes four gasifiers and three different gas cleanup processes. Bechtel is characterizing the combinations to determine which gas cleanup process and which gasifiers have the best technical and economic characteristics.

Activities 111 & 113

During this quarter studies to determine the effect of part-load operation on the fuel cell stack and design requirements were initiated. The first objective was to determine the minimum reactant flow conditions imposed on the cell stack for this mode of operation. A mode of power plant control and operation for part-load was selected. It was chosen to provide high part-power and to result in the most stringent part-load conditions for the stacks and module were defined. These results have been included in design requirements being used in Task 2.

1.B DEFINITION OF USER REQUIREMENTS AND EVALUATION CRITERIA (ACTIVITY 101)

The objective of this activity is to define user requirements and evaluation criteria for molten carbonate power plant applications. Both central station and cogeneration applications are to be considered.

Summary

The first meeting of the User Review Panel was held at UTC on November 13, 1980. Representatives from eight electric utilities participated. The technical, operating, and economic requirements for a utility central station molten carbonate power plant as derived by UTC and Bechtel were reviewed and critiqued.

Discussion

The definition of power plant technical and economics requirements and evaluation criteria are being defined in this program by joint UTC, Bechtel, and End-users effort. The approach is for UTC and Bechtel to establish a reference set of requirements for both central station and cogeneration applications, which are then reviewed by and modified to reflect comment by end-users. This approach permits participation of a large group of users, and enables the users to focus their resources power plant and application group of users that the requirements derived evaluations. This reflect ensures a broad spectrum of user needs and applications.

The utilities represented at the User Review Panel meeting were:

- o Boston Edison
- o Commonwealth Edison
- o Consumers Power
- o Duke Power
- o Niagara Mohawk
- o Northeast Utilities
- o Public Service Electric and Gas
- o Tennessee Valley Authority

Two other utilities - City Public Service (San Antonio) and Los Angeles Department of Water and Power - were unable to attend.

The meeting format was as follows:

- 1) Review of the DOE program.

- 2) Review of the molten carbonate power plant concept and features.
- 3) Review of projected utility requirements, and resulting technical and operational requirements for a coal fueled power plant.
- 4) Review of the assumptions and methodology, used in defining and the resulting economic requirements for a coal fueled molten carbonate power plant.

The molten carbonate power plant concept and features that UTC presented were based on studies in this program and previous studies including the ECAS and EPRI RP239-4 studies. They are summarized in Table 1-1 with the benefit they provide to the user.

TABLE 1-1. MOLTEN CARBONATE POWER PLANT DESIGN FEATURES

<u>FEATURE</u>	<u>APPLICATION IMPACT</u>
o Low Heat Rate	o Reduced Fuel Costs
o Rating Flexibility at Low Heat Rates	o Increased Sitability o Matched Load Growth o Reduced Financial Burdens
o Environmental <ul style="list-style-type: none"> • Low Emissions • Low Water Requirements • Low Waste 	o Increased Sitability
o Modularity	o Good Availability

Figure 1-1 shows the fuel cell's heat rate characteristic relative to competing options.

Figure 1-1. Molten Carbonate Power Plant Rating Features

Figures 1-2, 1-3, and 1-4 compare the molten carbonate technology with other coal technologies for SO_x emissions, waste disposal requirements, and water resource requirements, respectively. The low emissions result from the fuel cell's requirement for a clean fuel gas; low waste disposal and water resource requirements are due to the low heat rate of the power plant. In the case of water requirements, the ratio of fuel cell/steam turbine power output is high, thereby reducing specific cooling water make-up requirements relative to other coal technologies. Figure 1-5 indicates that these features could increase the capacity of a selected site by almost a factor of 3 relative to conventional coal/ steam plants with flue gas desulfurization, without deterioration of the site in terms of SO_x or water availability.

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Figure 1-2. Molten Carbonate Power Plant Emissions

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801211

Figure 1-3. Molten Carbonate Power Plant Waste Disposal Requirements

Figure 1-4. Molten Carbonate Power Plant Water Requirements

Figure 1-5. Low Emissions and Water Requirements Improve Sitability

Bechtel presented projected requirements of the utility industry and the technical and operating design requirements for a coal fueled power plant designed to meet those requirements. Bechtel identified both baseload and intermediate duty cycles as applications for a coal fueled power plant. The design requirements for these two applications are listed in Tables 1-2 and 1-3. The baseload requirements are consistent with those previously defined earlier in the program. The intermediate application is based upon a smaller power plant rating, and a different duty cycle (reduced capacity factor and load factor) than the baseload; anticipated coal types and environmental concerns are identical to the baseload. Review of these requirements by the panel resulted in the following recommendations:

- o A rating range of 200-400 MW is more desirable than 400 MW's for future intermediate applications.
- o Cycling capabilities are desirable in a baseload power plant since duty cycle requirements of a power plant change with time. Cycling coal plants are being planned for utility applications.
- o Base load power plant design goals should include at least one startup/shutdown cycle per month.
- o Repowering of the sites near industrial areas may represent a large market. Typical installations would be in the range of 50-100 MW. Power plants in this capacity range should be considered.

TABLE 1-2. PRELIMINARY DESIGN REQUIREMENTS
BASE LOAD PLANT

Unit Size, Mwe	400,800
Fuel	High Btu Coal, Low Btu Coal
Emission Requirements	NSPS, >NSPS
Cooling System	Wet, Wet/Dry
Capacity Factor	70.0
Plant Load Factor	85-95
Availability	70-80
Planned Outage Rate	8-13
Starts/Stops (per year)	TBD

TABLE 1-3. PRELIMINARY DESIGN REQUIREMENTS
INTERMEDIATE PLANT

Unit Size, Mwe	400
Fuel	High Btu Coal, Low Btu Coal
Emission Requirements	NSPS, >NSPS
Cooling System	Wet, Wet/Dry
Capacity Factor	35.0
Plant Load Factor	70-90
Availability	70-80
Planned Outage Rate	8-13
Starts/Stops (per year)	TBD

The panel also reviewed the assumptions and methodology UTC used to establish study results "allowable" capital costs for the molten carbonate fuel cell power plant. These costs make the fuel cell competitive with other equipment options for several utility application scenarios. These costs are summarized in Figure 1-6.

Figure 1-6. Influence of Alternate Duty Cycle

The user's comments were as follows:

- o The coal costs assumed in the study (\$2.00/106 Btu) appear low; currently at least one utility is paying 2.50/106 Btu for coal. Also coal transportation costs may be a limiting factor for small unit applications.
- o The study should consider the sensitivity of the result to variations in fixed charge rate. Investment tax credits may be available for retrofit and new plant sulfur cleanup systems, which would tend to lower fixed charge rates.
- o The high cost of coal gasification equipment may make it difficult to achieve competitive economics for intermediate applications. An alternative would be to run the gasifier(s) at full capacity all of the time, with on-site capability for conversion of the fuel gas to a storable liquid product during periods of low electrical load.

B. Planned Work for Next Reporting Period

The comments presented above are a summary of the immediate response of the User Review Panel to UTC's and Bechtel's presentation. UTC will determine the effect of the user's comments on power plant design requirements and will solicit additional comment from the Users Review Panel prior to establishing a set of finalized power plant design requirements.

1.C ESTABLISH ALTERNATIVE SUBSYSTEM CHARACTERIZATION (ACTIVITY 102)

The objective of this activity is to define pertinent characteristics of alternative types of equipment which may be used in the subsystem comprising a power plant design. For purposes of study and reporting, the power plant is divided into two sections: fuel processing, including gasifier and gas clean up and all associated equipment, and the power section, incorporating all equipment downstream of fuel gas cleanup. The latter includes the fuel cell, gas expansion and compression equipment, and bottoming cycle and/or heat recovery steam generation equipment, depending on the end application.

Summary

Subsystem characterizations for alternative gasifiers and gas cleanup processes in the fuel processing section were completed by Bechtel during this quarter. This work completed the effort in this activity.

The final characterizations provided by Bechtel include some revisions to the list of selected alternative gasifiers plus data describing the Babcock-Wilcox entrained flow gasifiers, bringing the total number of gasifiers included in the study to 11. Each was evaluated relative to three operational parameters that provide a measure of the anticipated performance of molten carbonate power plant sections integrated with these gasifiers. Bechtel also reviewed previous characterization data supplied on the six alternative gas cleanup processes selected in this activity; no revisions were necessary.

The alternative subsystems selected, and their characterizations derived in this activity, are the basis for defining the overall power plant concepts in Activity 103.

Discussion

Additional fuel processing equipment characterizations have been provided by Bechtel for the Babcock-Wilcox O₂-blown entrained flow gasifiers, the O₂-blown version of the Westinghouse gasifier, and revised performance data for Shell-Koppers gasifier. The Babcock-Wilcox gasifier is similar to the Shell-Koppers gasifiers; both are dry coal feed, O₂-blown, entrained flow gasifiers. The study has been expanded to include the Babcock-Wilcox gasifier to ensure that design information will be available for this type of gasifier in later stages of the study. Bechtel has found that the level of characterization detail necessary for the next stage of this work may not be available for the Shell-Koppers gasifier. Characterization of the Westinghouse gasifier was completed by the addition of the O₂-blown case. Earlier work characterized only the air-blown case. The technical and operating data for these gasifiers is listed in Table 1-4. All the data presented are available in the public domain.

TABLE 1-4. GASIFIER CHARACTERISTICS

Technology	Westinghouse	Shell-Koppers	Babcock-Wilcox
Gasifier Type	Fluidized Bed, pressurized, slagging	Entrained flow, pressurized, slagging	Entrained flow, pressurized, slagging
Capacity Single Unit	Up to 3150 TPD	Up to 2650 TPD	Up to 5000 TPD
Oxidants Accepted	Oxygen, air	Oxygen	Air, Oxygen
Coal Types Accepted	All, crushed, dried	All, crushed, dried	All, pulverized
Fraction of Design Capacity at Maximum Turndown	0.33	0.67	0.50
Resources Required	Coal, Air, Water	Coal, Air, Water	Coal, Air, Water
Solids, Liquids Emitted	Char	Slag	Slag
Commercial Status	3rd generation	2nd generation	2nd generation
Range, Outlet Temps, °F	1600 to 2000	2700	1800
Range, Pressures, psig	200 to 400	250 to 450	0 to 350
Contaminants to Remove from Gas	S compounds, trace metals	S compounds, trace metals	S compounds, trace metals
<u>Typical Operating Parameters</u>			
<u>Reference</u>	Communication Westinghouse/Bechtel	1979 AIChE Annual S.F. Meeting-Foreign Synfuels Tech.	B&W Report SP110 Rev. 7/78, adjusted by Bechtel

TABLE 1-4. GASIFIER CHARACTERISTICS (Continued)

Technology	Westinghouse	Shell-Koppers	Babcock-Wilcox
<u>Oxidant</u>			
Purity	98.4	99.9	98.4
Temp., °F	650	N/A	77
Press., psig	370	450	65
<u>Coal</u>			
Type	Illinois No. 6	Illinois No. 6	Pittsburgh No. 8
HHV Btu/lb	12,092	12,143	12,279
Wt. % C	65.83	66.72	69.66
H	4.94	4.70	4.69
O	7.95	9.31	5.63
N	1.34	1.03	1.38
S	3.68	3.67	1.58
Ash	10.27	12.57	7.98
Moisture	5.99	2.00	9.00
<u>Steam Feed</u>			
Temp., °F	900	N/A	None used
Press., psig	370	450	None used
<u>Gasifier Feed</u>			
Temp., °F	1850	2730	1800
Press., psig	350	435	50
<u>Vol %</u>			
H ₂	25.16	31.6	30.47
CO	37.76	64.0	67.27
CO ₂	10.85	0.8	0.23
H ₂ O	16.70	1.5	0.37
N ₂ + AR	0.31	0.7	1.06
H ₂ S	1.20	1.3	0.57
COS	---	0.1	---
CH ₄	7.02	---	0.03
C ₂	---	---	---
NH ₃	1.0	---	---

TABLE 1-4. GASIFIER CHARACTERISTICS (Continued)

Technology	Westinghouse	Shell-Koppers	Babcock-Wilcox
Oxidant	O ₂	O ₂	O ₂
Material & Heat Flows on a Basis of 10 ⁶ Btu H ₂ + CO:			
1. <u>Material Flows</u>			
Coal, lb	137.37	100.32	98.01
Coal, lb Atom C	7.54	5.57	5.68
Oxidant Stream, lb	80.25	73.04	81.61
Oxygen, lb-Atom O	4.95	4.59	5.00
Steam Feed, lb	68.69	6.60	None
Steam Feed, lb-moles	3.81	0.37	None
Gas, lb-moles	13.00	8.56	8.38
Gas, scf	4,933	3,250	3,179
2. <u>Heat Flows, 10⁶ Btu</u>			
Coal, HHV	1.611	1.218	1.203
H ₂ + CO, HHV	1.000	1.000	1.000
CH ₄ + C ₂ , HHV	0.349	---	0.001
Char C, HC			
Liquids, HHV	0.036	0.020	0.005
Waste Heat Boiler	0.157	0.180	0.180

Two parameters were previously selected as primary criteria by which to judge gasifier compatibility with a fuel cell and to provide high overall power plant performance. They are:

- o Coal to Syngas Energy Ratio. The ratio of the energy content of coal feed to the energy content of the H₂ and CO produced. Gasifiers with low ratios maximize the chemical energy (H₂ and CO) that can be converted electrochemically at the high fuel cell efficiency relative to the coal energy input to the system. This tends to increase power plant efficiency.

- o Oxygen to Syngas Energy Ratio. The ratio of the oxygen required to the energy content of H_2 and CO produced. Oxidant supply represents a large parasitic power consumption in the gasification system. Low ratios minimize those parasitic requirements, tending to increase power plant efficiency.

A third gasifier parameter has been identified which permits selection between two gasifiers with similar coal and oxygen to syngas energy ratios. It is

- o Sensible Heat to Syngas Energy Ratio. It is the ratio of the sensible heat recoverable from the raw fuel gas to the energy content of the H_2 and CO produced. This is a measure of the energy that can be used to raise steam for a bottoming cycle or cogeneration application. High ratios tend to increase power plant efficiency.

Table 1-5 compares all 11 gasifier candidates now included in the study in terms of these parameters. Data are based on a normalized coal feed. Several conclusions can be drawn from these data which permit selection of the gasifiers to be included in the feasible power plant design concepts to be carried out in Activity 103. The conclusions are:

- o The O_2 -blown entrained flow gasifiers yield the lowest Coal to Syngas Energy Ratios. These gasifiers also exhibit relatively low Oxygen to Syngas Energy Ratios, and high Sensible Heat to Syngas Energy Ratios. These gasifiers are the most attractive candidates for integration into the feasible power plant concepts.
- o The O_2 -blown fluidized bed gasifiers have attractive Oxygen to Syngas Energy and Sensible Heat to Syngas Energy Ratios, although their Coal to Syngas Energy Ratios are high.
- o Except for the BGC/Lurgi (Slagging) gasifier, the moving/fixed bed gasifiers do not appear as attractive for integration into power plant concepts.
- o The molten salt gasifier has a low Coal to Syngas Energy Ratio, but a relatively high Oxygen to Syngas Energy Ratio, and an unknown Sensible Heat to Syngas Energy Ratio.

These characterizations are the basis for selection of gasifiers to be incorporated in the next phase of Task I (Activity 103).

TABLE 1-5. COMPARISON OF GASIFICATION SYSTEMS ACCORDING TO COAL AND OXYGEN DEMANDS, AND RECOVERABLE SENSIBLE HEAT

Gasification System	Oxidant	Coal to Syngas Energy Ratio, $\frac{10^6 \text{ Btu Coal}}{10^6 \text{ Btu H}_2 + \text{CO}}$	Oxygen to Syngas Energy Ratio, $\frac{1\text{b} - \text{Atoms O}}{10^6 \text{ Btu H}_2 + \text{CO}}$	Sensible Heat to Syngas Energy Ratio, $\frac{10^6 \text{ Btu}}{10^6 \text{ Btu H}_2 + \text{CO}}$
<u>Bearing/Fixed Bed</u>				
o Lurgi	Air	1.685	4.42	0.06
	O ₂	1.937	4.62	0.06
o BGC/Lurgi	O ₂	1.412	3.32	0.02
o Riley-Morgan	O ₂	1.521	4.40	Negligible
o Kilngas	Air	1.551	5.74	No data
<u>Fluidized Bed</u>				
o Winkler	Air	1.544	6.36	0.30
	O ₂	1.418	4.37	0.20
o Westinghouse	Air	1.620	6.53	No data
<u>Entrained Flow</u>	O ₂	1.661	4.95	0.157
o Koppers-Totzek	O ₂	1.365	5.52	0.20
o Texaco	O ₂	1.294	5.43	0.18
o Shell-Koppers	O ₂	1.218	4.59	0.18
o Babcock & Wilcox	O ₂	1.203	5.00	0.18
<u>Molten Salt</u>				
o Rockwell	O ₂	1.280	5.45	No data

*British Gas Council

Gas Cleanup Characterizations. Six acid gas cleanup processes had previously been selected as alternatives for power plant integration. The six selected processes are:

- o Selexol
- o Rectisol
- o Benfield Hi Pure
- o Alkazid DIK
- o Stretford
- o Hot Iron Oxide

Two of the criteria used in selecting these six were that they required low to moderate steam and power consumption, and were in the lower range of projected costs relative to a list of 14 processes previously judged suitable for application with coal-derived gases (Ref. 1). Additional data has been provided by Bechtel in an effort to further compare the capital costs and operating utility requirements of these processes.

Table 1-6 provides a tabulation of capital costs for the six gas cleanup systems. The costs are expressed in mid-1978 dollars per standard cubic feet (scf) per hour of gas treatment capacity. References cited in the table are given at the end of this report. The costs fall into two groups reflecting the severity of the sulfur removal requirement:

- o Shallow removal, reducing H_2S to 200 to 1000 parts per million by volume (ppmv).
- o Deep removal, reducing H_2S and COS to 0.1 to 20 ppmv.

Shallow removal is usually adequate for meeting moderate fuel environmental standards. It is not a removal level adequate for molten carbonate fuel cells. Deep removal cases in Table 1-6 involve cleanup to protect methane or methanol synthesis catalysts which are poisoned by H_2S and organic sulfur compounds. The level of removal corresponds closely to that required for molten carbonate fuel cells.

TABLE 1-6. CAPITAL COST DATA ESCALATED TO MID 1978
FOR GAS CLEANUP SYSTEMS

CASE	PLANT CONFIGURATION PARAMETERS						ACID GAS REMOVAL PARAMETERS						CAPITAL REQUIREMENT, \$ PER SCF/H FEED GAS			COST REFERENCE, METHOD
	Acid Gas Removal Process	Sulfur Recovery Process	Gasifier	Oxidant	Coal Type	Plant Coal Intake 10 ⁶ Btu/hr	Treatment Pressure psig	H ₂ S Removal Req'd.	COS Removal Req'd.	CO ₂ Removal Req'd.	CO ₂ Inlet Vol. %	Acid Gas Removed lb-moles 10 ⁶ SCF	Acid Gas Removal	Sulfur Recovery	Total	
1	Iron Oxide	Allied/ Claus	UGas	Air	Eastern	4,000	140	Shallow	Shallow	None	6	17	.50	1.3	1.80	UTC/Burns & Roe 76
2	Selexol 3 trains	Claus	Texaco	O ₂	Eastern	5,000	600	Shallow	Partial	None	11	59	-	-	1.50	EPRI AF-642 Fluor
3	Selexol 4 trains	Claus	Texaco	Air	Eastern	5,000	600	Shallow	Partial	None	8	55	-	-	1.20	EPRI AF-753 Fluor
4	Selexol 3 trains	Claus	BGC	O ₂	Eastern	5,000	300	Shallow	Partial	None	2	59	-	-	1.50	EPRI AF-642 Fluor
5	Stretford 4 trains	-	CE	Air	Eastern	5,000	1	Shallow	None	None	5	12	-	-	.70	EPRI AF-244 Fluor
6	Stretford 3 trains	-	CE	O ₂	Eastern	5,000	1	Shallow	None	None	2	31	-	-	1.50	EPRI AF-244 Fluor
7	Stretford	-	Lurgi	Air	Western	3,000	270	Shallow	None	None	15	6	-	-	1.10	Stearns-Rogers 73, Factored with Case 11
8	Alkasid	Claus	GEGAS	Air	Eastern	5,000	310	Shallow	None	None	6	28	.50	.90	1.40	Bechtel 1976
9	Alkasid & Hydrolysis	Claus	Winkler	O ₂	Eastern	800	110	Deep	Deep	None	17	110	-	-	5.90	Davy 1974
10	Rectisol	Stretford	Lurgi	O ₂	Western	16,000	300	Deep	Deep	Shallow	33	880	1.00	.60	3.60	Stearns-Rogers 73; Factored with Case 11
11	Selexol	IFP Stackpol	Lurgi	O ₂	Western	16,000	300	Deep	Deep	Shallow	33	880	2.30	2.50	8.00	Braun 1976
12	Selexol	?	Lurgi	O ₂	Eastern	16,000	300	Deep	Deep	Shallow	33	880	4.70	3.90	8.60	Braun 1978 Inferred
13	H ₂ Pure	IFP Stackpol	Steam-Iron Hygas	O ₂	Eastern	16,000	1100	Deep	Deep	Shallow	7	200	2.50	3.00	5.50	Braun 1976, 1978 Inferred
14	H ₂ Pure	?	BGC	O ₂	Eastern	25,000	300	Deep	Deep	Shallow	2	110	.60	.60	1.20	EPRI AF-523 Parsons
15	H ₂ Pure	?	K-T	O ₂	Eastern	25,000	600	Deep	Deep	Shallow	8	240	2.70	.60	2.30	EPRI AF-523 Parsons
16	H ₂ Pure	?	Texaco	O ₂	Eastern	25,000	600	Deep	Deep	Shallow	10	300	3.20	.60	2.80	EPRI AF-523 Parsons
17	H ₂ Pure	?	Foster Wheeler	O ₂	Eastern	25,000	300	Deep	Deep	Shallow	14	430	3.00	.60	3.60	EPRI AF-523 Parsons
18	Selexol	?	?	O ₂	Eastern	16,000	300	Deep	Deep	None	?	?	2.35	-	-	Bechtel Factor times Case 12

The costs for deep removal in Table 1-6 depend on the following parameters:

- o Chemical versus physical solvent.
- o The number of moles of acid gas removed per 10^6 scf of feed gas.
- o Whether CO_2 must be removed along with H_2S .

The costs in Table 1-6 are in many cases factored or inferred from rather fragmentary information from diverse sources. Consequently they are not suitable for comparisons of or screening of the processes. The costs in the table which appear soundest and most applicable are those for Hi Pure and Selexol in Cases 13 and 18.

Table 1-7 provides data on typical utility demands of the acid gas removal processes under conditions applicable for molten carbonate fuel cell application. However, utility requirements can vary substantially depending on design conditions, including the composition and pressure of the gas to be treated, and also with the strategy for design of the units. Some ways in which the utility requirements can vary for various processes are as follows:

- o The use of refrigeration reduces capital costs, but increases compression power demands.
- o The pressure of the acid gas leaving the liquid regenerator influences stripping steam requirements.
- o The temperature approach in rich/lean liquid heat exchanger influences stripping steam requirements.
- o The volume of acid gas to be removed, and for some, CO_2 levels influence utility requirements.
- o For some processes, stripping steam can be reduced if nitrogen from an air separation plant can be used for removing residual H_2S from the solvent.

Review of the data in Tables 1-6 and 1-7 indicates further screening of the gas cleanup processes will be required, and all six will be carried to Activity 103.

TABLE 1-7. UTILITY REQUIREMENTS FOR ACID GAS REMOVAL
PROCESSES PER 10^6 SCF OF GAS TREATED

Process	Treatment Pressure psig	Acid Gas Removed, moles	Steam Required, lb	Power Required, MW-hr	Reference
Selexol	600	50	20,000	0.8	Bechtel data
Hi Pure	600	200	7,000	0.06	Benfield 1974
Alkazid	100	100	22,000	---	Davy 1974

This completes the planned work in this activity.

1.D DEFINITION OF FEASIBLE POWER PLANT CONCEPTS (ACTIVITY 103)

The objective of this activity is to derive several power plant concepts for both central station and cogeneration applications. The concepts are derived from the subsystem alternatives previously defined (Activity 102). Feasibility is determined by power plant performance relative to technical and operating requirements for the application as defined in this program (Activity 101).

Summary

Starting with a broad matrix of alternative gasifiers and gas cleanup processes selected in Activity 102, a smaller matrix has been derived for further study representing feasible fuel processing combinations for integration into overall power plant concepts. Selection of the processes included in the smaller matrix was based on screening the alternative gasifiers and gas cleanup processes using the characterization data derived in Activity 102, and was a joint effort by UTC and Bechtel.

Effort is continuing by Bechtel to furnish conceptual configurations and thermodynamic data for the of selected feasible fuel processing sections. This will permit derivation of a range of feasible power plant concepts for evaluation in later program effort.

Discussion

Activity 102 identified 11 gasifiers and 6 cleanup processes as alternative subsystems for use in molten carbonate power plants. The matrix of these processes is shown in Table 1-8, indicating potentially 66 different cases; inclusion of both air and oxygen-blown gasifier versions expands this to 90 possible cases.

TABLE 1-8. MATRIX OF GASIFIER/GAS CLEANUP ALTERNATIVE SUBSYSTEMS DERIVED FROM ACTIVITY 102

Gasifier	Oxidant	Gas Cleanup Process					
		Alkazid DIK	Benfield Hi Pure	Rectisol	Selexol	Stretford	Hot Iron Oxide
Lurgi	O ₂						
	Air						
BGC Slagger	O ₂						
Riley Morgan	O ₂						
Kilngas	Air						
Winkler	O ₂						
	Air						
Westinghouse	O ₂						
	Air						
Koppers-Totzek	O ₂						
Texaco	O ₂						
	Air						
Shell-Koppers	O ₂						
Babcock & Wilcox	O ₂						
Rockwell Molten Salt	Air						

(15 x 6 Matrix = 90 possible cases)

Discussions with Bechtel together with review of the gasifier characterization data indicated in Table 1-5 above resulted in selection of 4 gasifiers as most promising for integration into power plant concepts. These gasifiers are:

- o Shell-Koppers entrained flow bed - O₂-blown
- o Babcock & Wilcox entrained flow - O₂-blown
- o Westinghouse fluidized bed - O₂-blown
- o Texaco entrained flow bed - O₂-blown

These gasifiers exhibit favorable characteristics in terms of relatively low coal to syngas and oxygen to syngas ratios, and were in the high range of the sensible heat to syngas ratios, as previously indicated above. Although three entrained flow gasifiers are included, the Texaco is slurry feed, whereas the other two are dry feed. The reasons for including the Babcock-Wilcox were indicated above.

Additional factors were used to confirm these choices. They are that:

- o Although the Koppers-Totzek entrained flow and the Winkler O₂-blown fluidized bed gasifiers exhibit good performance parameters, they both are ambient pressure gasifiers. This would require pressurization of the coal gas for integration with the molten carbonate power section. Raw gas cleanup with ambient processes limits gas cleanup choices; alternatively pressurization of the raw gas prior to cleanup is questionable due to the severity of the environment for the compression equipment. Trends in the gasifier industry have been toward pressurized operation to facilitate cleanup and reduce piping and component costs.
- o Except for the BGC gasifier, the moving bed gasifiers did not appear attractive. The BGC gasifier however: (1) lacks coal flexibility, specifically inability to operate on Eastern coking coals; and, (2) it produces residual by-products (tars and phenols) that can not be recycled to extinction.
- o The Rockwell molten salt gasifier is considered far term technology, beyond the time frame of this program.
- o In general, the air-blown gasifiers exhibited poorer performance parameters than the O₂-blown versions; additionally, cleanup of sulfur to low levels is more difficult with the higher mass flows associated with air-blown gasifiers (Reference 1).

Of the 24 resulting study cases (4 gasifiers and 6 cleanup processes), the matrix was further screened to eliminate less promising fuel processing combinations by review of the gas cleanup data provided in Activity 102. The resulting matrix of 8 study cases is indicated in Table 1-9. Reasons for selection of these cases include:

- o Hot iron oxide has not been demonstrated commercially, as compared to the commercial status of all the other processes. Furthermore, the process does not have the capability for deep sulfur removal thermodynamic equilibrium, necessitating a second stage of regenerable sulfur scrubbing prior to passing through a ZnO guard bed.
- o Stretford has primarily been used for scrubbing H₂S from near ambient pressure gases. A concern for high pressure applications is that high CO₂ pressures reduce the efficiency of the reagent for H₂S absorption, and the process does not appear as economically competitive as the others under consideration.
- o Rectisol was similar to Selexol, except for: (1) Volatility of the solvent, requiring solvent makeup, and (2) Lower temperatures of operation, which result in greater loss of sensible heat from the coal gas.
- o Selection of Selexol for the Texaco gasifier is based on a recent study by C. F. Braun (Reference 2).
- o The Babcock-Wilcox gasifier has similar characteristics to the Shell-Koppers and therefore is shown with the same cleanup process.

TABLE 1-9. MATRIX OF GASIFIER/GAS CLEANUP PROCESSES SELECTED FOR STUDY IN ACTIVITY 103

Gasifier	Gas Cleanup		
	Alkazid DIK	Benfield Hi Pure	Selexol
Westinghouse	X	X	X
Texaco			X
Shell-Koppers	X	X	X
Babcock-Wilcox	Select one based on results from Shell-Koppers studies.		

Work Planned for Next Reporting Period

Bechtel will define the most promising gas cleanup process for both the Westinghouse and Shell-Koppers gasifiers, based on the economics of supplying clean fuel gas. Data from other studies will be normalized where required. Process flow

sheets will be prepared for each of four preferred fuel processing subsystem combinations.

1.E DEFINE FUEL CELL MODULE AND STACK REQUIREMENTS (ACTIVITIES 111 AND 113)

The objective of this activity is development of a complete set of requirements for the fuel cell module and stack. These include physical dimensions, interactions with other subsystems, and performance and other operating conditions. Development of these requirements was initiated early in the program, with the selection and definition of a preliminary reference design. These requirements will be updated and revised to be consistent with the reference power plant design selected later in this program.

Summary

Work to determine the impact of power plant turndown on the fuel cell module and stack design requirements was initiated. The basis for the study was the preliminary reference design concept previously defined. A control mode was selected for part power operation which results in good part power performance. This control mode maintains the cell stacks at constant pressures and temperature gradients. The principal change in operating conditions is reduced fuel and oxidant flows as the power plant is turned down. These operating conditions have been included in the design requirements of Task 2.

A second effort investigated assembly alternatives for the fuel cell modules. Factory assembly, as opposed to field assembly, was selected to minimize costs and increase reliability.

Discussion

Operational Requirements Definition. A preliminary reference utility molten carbonate power plant design was defined early in the program as a basis for establishing preliminary design requirements for the cell stack and module. The concept

integrated a molten carbonate/steam cycle with a Texaco O_2 -blown gasifier and Selexol acid gas cleanup. Based on this system concept preliminary design requirements were established for rated power operation. Additional work was required to determine the effect of part-load operation and startup and shutdown on cell stack and module design requirements.

The principal focus of this work was on operating pressure, effects, temperature gradients, and the minimum reactant flow rates determining resulting from part power operation. Minimum flows for cell stack operation must be considered in the design effort to insure that maldistribution of reactants does not occur. Minimum flow rates represent the most stringent operating requirements for cell and cell stack design.

To start this work fuel processing subsystem characteristics at part-load were required. Bechtel provided typical turndown performance estimates for the fuel processing section of the preliminary reference design. These estimates are shown in Table 1-10.

Several fuel cell subsystem control modes for turndown were considered. The options included both constant and variable speed turbocompressor machinery, and the location of bypass control valves in the cathode loop. Using an existing steady state proprietary computer model, a control mode was selected based on good part power performance. The control mode selected for the power section maintains the fuel cells at constant pressure and reduces the coolant flow with load. The system pressure was held constant to provide the capability to follow load changes quickly. The turbocompressor is operated at constant speed to maintain constant system pressure. The part power performance is highest by flowing all of the air through the fuel cell stacks. Thus, the overall oxygen utilization decreases and the CO_2 utilization is constant as power output decreases. The coolant flow (cathode recycle) is reduced at part power to maintain a constant temperature rise and thus maintain constant cell temperature gradients.

TABLE 1-10. ESTIMATED TURNDOWN PERFORMANCE OF FUEL PROCESSING MODULES SERVING MOLTEN CARBONATE FUEL CELL POWER PLANTS

Plant Module	Coal Handling	Oxygen Supply	Texaco Gasification	Selexol S Removal	Claus/SCOT Recovery
Maximum Turndown Possible, One Train	25%	50%	50%	50%	33%
Component Turndown Features	<ul style="list-style-type: none"> • Unloading facility would operate fewer hours • Reclaim facilities consume power at slightly reduced rate 	<ul style="list-style-type: none"> • Cold box can operate at 50% capacity • Air compressors operate at 70% and vent 20% • Oxygen compressors operate at 70% with recycle O₂ 	<ul style="list-style-type: none"> • Slurry, oxygen flows turndown to 50% • Assume particulate scrub water flow stays at 100% 	<ul style="list-style-type: none"> • Absorber gas rate can be reduced to 50% • Liquid circulation rate would be reduced to 70% although Allied Chemical Co. believes it can be reduced to as low as 50% • With increased ratio of liquor to gas flow rates, CO₂ pickup will increase and reduce acid gas H₂S concentration 	<ul style="list-style-type: none"> • Lower feed H₂S concentration leads to lower percent sulfur conversion in Claus unit • SCOT unit should be able to recover extra sulfur in Claus tailgas, operating near 100% capacity
Estimated Fraction of Design Utility flows at 50% Turndown					
Steam Generated	---	---	50%	---	50%
Steam Consumed	---	---	50%	70%	100%
Cooling Water Consumed	---	75%	60%	70%	100%
Electricity Consumed	60%	75% (Assuming all drives electric)	60%	75%	95%
Maximum Turndown Rate, Percent of Design Per Minute	10	1	4	4	4

The estimated performance for this control mode from rated power down to the gasifier turndown limit established from Table 1-10 is shown in Figure 1-7. Turn-down of the fuel processor to a minimum of 50% capacity results in a power plant output that is 42% of rated. The efficiency at this point is 39.4%, as compared to 48% at rated output. The figure also shows power plant efficiency as a function of power output for the selected fuel cell system control mode assuming that both fuel cell trains and gasifiers were turned off during turndown. This option maintains high power plant efficiencies down to 40-50% of rated power.

Figure 1-7. Molten Carbonate Power Plant Part Power Performance

A comparison of the fuel cell stack operating conditions for rated and part-power operation are shown in Table 1-11. This is the minimum part-power point assuming that all gasifiers and fuel cell modules are operating, and it represents the lowest reactant flow rates and therefore most stringent part power design conditions. No significant gas composition changes are anticipated; however, volume flows are reduced to 50% at the anode, and to 40% at the cathode.

TABLE 1-11. COAL GAS MOLTEN CARBONATE CELL FLOWS

	Rated Power (B.O.L.)*				Minimum Power (B.O.L.)* (42% of Rated Power)			
	Anode		Cathode		Anode		Cathode	
	Inlet	Exit	Inlet	Exit	Inlet	Exit	Inlet	Exit
<u>Total Flow (Wet)</u>								
Moles (% of rated power conditions)	100.	100.	100.	100.	51.2	48.3	40.4	40.0
<u>Composition (Vol % Dry)</u>								
H ₂	38.3	5.57	---	---	38.3	7.5	---	---
CO	56.0	3.84	---	---	56.0	5.1	---	---
CO ₂	4.8	80.1	18.0	15.7	4.8	86.9	15.8	8.1
CH ₄	0.1	0.07	---	---	0.1	0.1	---	---
N ₂	0.7	0.4	57.9	76.3	0.7	0.4	67.2	77.4
O ₂	---	---	14.1	8.0	---	---	17.0	14.9
H ₂ O/Dry Gas (Vol/Vol)	0.755	0.23	0.27	0.29	0.755	0.692	0.13	0.151
Temperature (°F)	1100	1220	1140	1300	1189	1220	1050	1313
Pressure (psia)	~150	---	~150	---	~150	---	~150	---
Fuel Utilization (overall)	85.0%		---		80.2%		---	
CO ₂ Utilization (overall)	---		24.6%		---		40.8%	
O ₂ Utilization (overall)	---		25.0%		---		16.1%	

* Beginning of Life

These conditions have become part of the Task 2, design requirements. They will be revised, as necessary, to be consistent with the evolution of the reference power plant design.

Physical Design Requirements. A second effort in this activity was investigation of factory assembly and field assembly of the fuel cell modules. Figure 1-8 indicates the range of options under consideration.

Factory assembly of fuel cell modules was selected on the basis of maximizing reliability of the assembled fuel cell modules, and minimizing power plant costs. This selection was based on discussions with Bechtel, and is consistent with their most recent design and construction trends for minimum plant costs. Bechtel indicated that optimization studies in this area would be site specific, and therefore, predicated on specific site selection(s). The two major site specific parameters include labor and productivity rates, and transportation costs. The latter must consider relative locations of power plant site, the fuel cell stack facility, and the pressure vessel facility, as well as options for transportation from the facilities to the power plant site.

Work Planned to the Next Reporting Period

Startup and shutdown procedures will be studied to determine their impact on cell requirements.

Figure 1-8. Factory and Field Assembly Options

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Section 2

TASK 2 - DEVELOP AND VERIFY STACK AND CELL DESIGN

2.A SUMMARY

This task provides a detailed design of a full-scale cell stack which meets the functional requirements of the power plant design selected in Task 1. It also includes the development of cell and stack components used in this design. Design verification of the cell and critical stack components is accomplished single cell, subscale stack, full-scale cell short-stack, and out-of-cell component tests.

Electrolyte structure development activity this quarter has included testing of three alternative tape-matrix edge configurations 4 in. by 4 in. subscale cells. One of these configurations has maintained a cross-pressure capability that is greater than 10 psi through four thermal cycles.

Anode development activity concentrated on upgrading dimensional stability. A variety of configurations, materials, and density variations are being tested. Initial results indicate that densification and heat treatment can improve anode dimensional stability.

Evaluation of alternative nickel oxide cathodes was the primary activity in cathode development during this period. Performance of a 4 in. by 4 in. single cell containing an alternative cathode is stable after two temperature swings. A dry-casting process was used to fabricate this cathode. This process was also used to fabricate larger area nickel oxide cathodes, but cracking has occurred during lithiation.

Welding trials using 3-ft. by 3-ft. separator plates showed that it is feasible to increase the size of the present plate configuration to full-scale. Tooling requirements and alternative welding techniques were identified. Local aluminum dispersion by laser deposition was verified as an acceptable aluminizing method.

Design of the short stack of full-scale cells was started. The full-scale short stack pressure vessel was ordered.

2.B DEVELOP ELECTROLYTE STRUCTURES (ACTIVITY 150)

Summary

Three alternative tape-matrix edge configurations are being evaluated in 4 in. by 4 in. cells. All three demonstrated acceptable initial performance. Those which provided uniform support of the matrix throughout the seal area have shown the best retention of cross-pressure after multiple thermal cycles of any tape matrix cells yet run. From these results we conclude that edge cracks following multiple thermal cycles have been the primary source of cross leakage in the cell.

Discussion

Effort this quarter focused on developing an improved edge configuration for the multi-layer tape-matrix. Because the layers of the matrix are of different widths a step occurs at the edge. This step is suspected to be the cause of edge cracks which occur after repeated thermal cycles. The cracks in turn lead to reactant cross leakage. The configurations being evaluated are alternatives which eliminate the step at the edge of the matrix.

Early versions of multi-layer matrix had no step at the edge. They were made by a process including a compression molding procedure in which no step occurred. This process, however, did not scale to layer sizes well because of the compression molding step.

In the first alternative, the step is eliminated by solvent bonding strips of tape-matrix into the void region. This approach is the primary candidate for eliminating the step in the matrices to be fabricated for the next stack to be built under EPRI program RP1273. Although this is a tedious fabrication process, this approach should produce an edge equivalent to those in the successful molded

version of the tape-matrix. Cell 289 was built with a matrix of this configuration. It has run for 2300 hours and is performing normally at 680 millivolts at 160 amps/ft². Oxidant leakage was initially high, but has dropped to 0.8 ccm, and reactant crossover is zero. The solvent bonded strips along the cell edges have had no discernible effect on cell performance or operation.

The other two options are more attractive from the manufacturing standpoint, but are a greater departure from present experience. The second option is to make all the layers of equal width. The first cell having this configuration, Cell 260, was built and tested but the evaluation of the matrix configuration was inconclusive. Cell 292 was built during this quarter to retest this configuration, and is presently operating at 660 mV at 160 amps/ft² after 1800 hours. It has been thermally cycled four times from room to operating temperature and its cross-pressure remains above 10 psi. This is the first tape-matrix cell to survive this number of thermal cycles at a cross-pressure that exceeds 10 psi. This provides strong evidence that the steps in the edges of the earlier tape matrices stimulated edge cracking. The cracks became cross-leakage sites that developed after multiple thermal cycles.

The third option uses a metal foil to fill the step in the matrix in the edge area. This configuration is currently being tested in Cell 293, which was thermal cycled three times in 1200 hours, and which also maintained a greater than 10-psi cross-pressure. However, a test equipment problem caused a major performance loss and the cell has been shut down. Post test inspection is in progress.

These results favor development of one of these two options.

Effort in the next quarter will include post-test analysis of Cell 293, continued thermal cycle evaluation of Cell 292.

2.C ANODE DEVELOPMENT (ACTIVITY 155)

Summary

During this period, anode development was directed at correcting the structural deficiency identified during operation of the Niagara-Mohawk tape-matrix stack. In that test a significant performance loss occurred in one cell, immediately after a rapid temperature change. Loss of load in the active area due to anode dimensional changes has been postulated as the most probable cause of the observed performance loss. The objective of recent anode development has therefore been to improve anode structural stability.

Four approaches were previously identified to improve anode creep strength: two material modifications, "A" and "B", one configurational modification, and densification (reduction in anode porosity to approximately 50%). Emphasis during this report period has been on identifying means to improve anode dimensional stability under an applied mechanical load. Testing was conducted to determine dimensional stability of the improved creep strength anode formulations, and to determine the effect of a 1500°F preheat treatment in achieving subsequent anode stability. Based on these tests, conducted at 1300°F, material "A", densification, and the 1500°F preheat treatment were all found to improve anode dimensional stability. Room temperature flexural testing indicates that alternative anode material "A" has lower strength and modulus than standard anodes, while densification and heat treating improve strength and modulus.

Anodes incorporating these improvements are being tested in 4 in. by 4 in. cells. Their performance and sensitivity to rapid changes in temperature are being evaluated. Cell 286 is the most promising. It contains a densified material "A" type anode.

Scale up activities are continuing. Design of the belt furnace modification was completed, and material procurement and subassembly fabrication are nearing completion. Graphite was selected as the material for full-scale carrier/mold plates.

Discussion

Dimensional Stability Testing. Out-of-cell dimensional stability tests at 1300°F were continued to 475 hours using 1-in.² anode coupons. The objectives of these tests are: (1) to determine the effect of a 1500°F heat treatment on subsequent anode dimensional stability at 1300°F, and (2) to evaluate the selected approaches for improving compressive creep resistance on dimensional stability. Nine duplicate specimens were heat treated for 1 hour at 1500°F prior to the addition of electrolyte and testing at 1300°F. Three other duplicate specimens were prefilled and tested without the 1500°F heat treatment.

Volume and thickness changes as a function of time are shown on Figures 2-1 and 2-2, respectively, for 1500°F heat treated anodes. Based on the overall volume changes, Figure 2-1, both the material "A" and densification approaches reduce volume shrinkage relative to that of nickel stabilized with 2% chromium. Higher chromium percentages result in significant volume increase, as in the case of the PSD 9.1% chrome or the vendor supplied 10% chrome anodes. Material "B" shows no improvement in volume stability, and a shrinkage in thickness, see Figure 2-2. The anodes with the best dimensional stability contain 2% chromium and combine material "A" and densification. These anodes, densified/type "A"-10 and "A"-11) exhibit volume changes within $\pm 1\%$, and 0 to 0.5% thickness change at 475 hours. The test will be continued to 800 hours.

The effect of the additional 1500°F heat treatment on subsequent stability at 1300°F is shown in Figure 2-3, volume changes, and Figure 2-4, thickness changes. Three specimens were selected for comparison: unstabilized nickel, vendor 10% chrome and PSD 2% chromium/type "A"-10. The 2% chromium "A"-10 specimens consistently had smaller dimensional changes at 475 hours as a result of the 1500°F heat treatment. The unstabilized nickel anode showed no benefit from the 1500°F pre-treatment. The Gould 10% chromium sample showed a slight increase in volume.

Figure 2-1. Anode Dimensional Stability Test

Figure 2-2. Anode Dimensional Stability Test

Figure 2-3. Effect of 1500°F Heat Treatment on Anode Volume Change at 1300°F

Figure 2-4. Effect of 1500°F Heat Treatment on Anode Thickness Change at 1300°F

Mechanical Property Tests. The effect of heat treatment (1 hour at 1500°F, 10% hydrogen/balance nitrogen) on room temperature flexural strength and modulus was determined using 2% chromium-stabilized material "A" anodes. Samples with 60% porosity and densified to 54% porosity were tested. The results are shown in Table 2-1, together with comparison data for standard 2% chromium anodes. The

results can be summarized as follows:

- o Material "A" anodes have about two thirds the strength and modulus of standard 2% chrome anodes.
- o Densifying material "A" anodes improves strength and modulus by a factor of approximately 1.5.
- o Heat treating material "A" anodes more than doubles flexural strength and more than triples flexural modulus.

TABLE 2-1. ANODE FLEXURAL PROPERTIES AT ROOM TEMPERATURE

Anode Composition	Porosity	Strength (10^3 psi)		Modulus (10^6 psi)	
		Standard	Heat-Treated	Standard	Heat-Treated
2% Chrome	65%	1.66 (4)	--	0.13 (4)	--
2% Chrome/A-10	60%	1.03 (3)	2.13 (2)	0.085 (3)	0.30 (2)
2% Chrome/A-11 Densified	54%	1.52 (4)	4.21 (3)	0.14 (4)	0.53 (3)

NOTES:

- o Four point flexure at 0.05-in./min. crosshead speed.
- o Specimens are unfilled, 0.5-in. wide by $2\frac{1}{2}$ -in. long. Thickness is 0.060-in. for 60-65% porosity, and 0.044-in. for 54% porosity.
- o Heat treatment for 1 hour at 1500°F in 10% H_2/N_2 .
- o Number of test specimens in parenthesis.

4 in. by 4 in. Cell Testing. Seven 4-in. by 4-in. cells were tested during this report period to evaluate experimental anodes: one to determine the performance of anodes using lower chromium stabilizer content; one containing a formed lower chromium content anode; four to evaluate improved creep resistance and dimensional stability; and one to evaluate improved creep resistance and dimensional stability at higher temperature (1300°F).

Cell 271, which was tested to evaluate the performance of an anode fabricated of material "C" with 1.8% chromium, was shut down at 2012 hours after exhibiting continuing decay for the last 1150 hours. Final performance was 677 mV at 100 amps/ft². This cell has been disassembled for determination of post-test anode dimensions, surface area, and porosity. These data will be compared with the post-test characteristics of the 2% chromium-stabilized anode of Cell 263.

Cell 266 was tested to determine the performance and stability of a formed 2% chromium-stabilized anode. It completed 2150 hours of testing at 1200°F. The cell underwent an additional thermal stress test with no permanent performance loss, although a temporary loss of 64 mV at 160 amp/ft² was observed. This cell has now operated for 96 hours at 1300°F. It will be directly compared with the characteristics of Cell 295. Cells 266's performance is summarized in Table 2-2.

TABLE 2-2. PERFORMANCE OF 4-IN. BY 4-IN. CELLS USED TO EVALUATE THERMAL STRESS TEST STABILITY

Cell No.	Anode Description	Hours @ 1200°F	Thermal Stress Tests	Performance		Initial Performance	
				mV @ 160 amp/ft ²	Cell IR (mV) @ 100 amo/ft ²	mV @ 160 amp/ft ²	Cell iR (mV) @ 100 amp/ft ²
266	Formed, 2% Chromium/Ni	2150	5	673	40	710	29
		96 @ 1300°F	0	741	30	738	30
286	Formed 2% Chromium/A-11 Densified	1147	5	696	51	754	28
287	Formed, 2% Chromium/A-11 Heat-Treated	833	3	678	42	721	32
290	Formed, 2% Chromium/A-11 B-2	582	2	744	38	749	32
291	9.1% Chromium, Folded Metal Current Collector	510	1	705	41	731	35

The performance history of the four cells being tested to evaluate the effect of improved anode creep strength and dimensional stability on performance is also summarized in Table 2-2. All cells in this table contained formed nickel anodes stabilized with 2% chromium, except Cell 291 which has an anode of 9.1% chromium content. Additional modifications are as noted.

Cell 286, with a densified material "A" anode, has undergone a total of five thermal stress tests in 960 hours. No loss in performance has occurred as a result of these thermal tests. The cell lost 38 mV at 160 amps/ft² during a planned shutdown at 1068 hours to room temperature. The balance of the loss is typical cell decay. This test is continuing; a total of 1147 hours have been accumulated to date.

Cell 287, with a heat-treated material "A" anode has undergone three thermal stress tests with no permanent performance losses due to these tests. This test is continuing and additional thermal stress cycles are planned.

Cell 290 was tested to determine the performance of a combined material "A"-11 "B"-2 anode. This cell has undergone two thermal stress tests in 582 hours. No permanent or temporary performance losses have occurred. It is currently operating at 744 mV at 160 amps/ft². Additional thermal stress tests will be conducted.

Cell 291 was built with PSD 9.1% chromium anode and a sheet metal current collector. The objective of this test was to confirm that maintaining load in the cell active area is required for stable cell performance. This objective was to be accomplished by demonstrating performance stability through multiple thermal stress tests, using an unimproved anode in a cell which provided load follow-up. A continuous positive load on the active area of the cell was to be provided by starting the test at a low ram pressure of 10 psi, with gradual adjustments to higher pressures. This required inserting a layer of tile around the wet seal area, to allow follow-up to occur. The cell lost 34 mV at 160 amps/ft² during its first thermal stress test at 366 hours. The ram pressure on the cell at that time was 20 psi. This is a much lower loss than the 223 mV exhibited by Cell 289 after

a thermal stress test at 365 hours. Cell 289 contained an unimproved anode stabilized with 2% chromium and a sheet metal current collector. It had no provision for load follow-up. The test of Cell 291 is being continued.

Cell 295 was built to determine the performance stability of a material "A"-11 anode at higher temperature (1300°F). This cell contains a 2% chromium-stabilized formed material "A"-11 anode which has been heat treated at 1500°F. It is the same type of anode used in Cell 287.

Scaleup. Fabrication of the assemblies required for modifying the belt furnace to accept full-scale parts is nearly complete. These will be pre-assembled prior to installation on the belt furnace to save time. The modified facility is expected to be ready for full-scale part trials by March.

Based upon recent evaluation of ceramic board and an update price quotation for graphite plate, graphite plate is a more satisfactory and cost effective material than ceramic board for carrier plates and molds. An order for 18 full-size graphite plates has been placed.

The latest evaluations of the lightweight rigidized ceramic board material indicates that extensive in-house processing is required to upgrade the material strength and surface quality to levels required for use as a carrier plate or mold plate material. A similar material with higher density and strength was found to be dimensionally stable up to 1200°F but bowed after exposure to temperatures over 1600°F. Further process development is therefore required to make the ceramic board into a suitable carrier/mold plate material for use in the belt furnace. Evaluation of the ceramic board will be continued, at a reduced level, as a candidate support plate for full-scale tape process components to be processed in an oxidative atmosphere.

2.D CATHODE DEVELOPMENT (ACTIVITY 160)

Summary

A second 4 in. by 4 in. cell evaluating an alternative nickel oxide cathode placed on test. The cell, 294, has a performance of 667 mV at 160 amps/ft². It has completed two temperature cycles to room temperature. The cathode was fabricated using a dry-casting process.

The dry-casting process was used to fabricate the alternative nickel oxide cathode in areas up to one square foot. Lithiation of one 10-1/2 inch square component resulted in multiple fractures however.

Measurement of the post-test conductivity of the alternative lithiated nickel oxide cathode from Cell 281 indicates that about half of the cathode side resistance loss of the cell can be attributed to contact resistance. The resistance is between the cathode and the cell housing. Post-test flexural and crush strength of this cathode were lower than pre-test values. Scanning electron microscope examination of the cathodes showed no obvious structural degradation.

A cathode was fabricated of Inco 255 nickel powder with surface area equivalent to that of cathodes fabricated of Inco 287 powder cathodes. The Inco 255 however, exhibited lower cell performance than those fabricated of Inco 287 powder.

Discussion

Cell 294 is being run to determine the effect of endurance on the alternative lithiated nickel oxide cathode. Both performance and structural characteristics of the cathode are being evaluated. The cell had an initial conductivity of $5 (\Omega\text{-cm})^{-1}$. The measured cell resistance loss of this cell was initially 43 mV at 100 amps/ft²; after 1600 hours of operation and two temperature cycles, it has risen to 50 mV at 100 amp/ft². Cell performance peaked at .710 mV at 160 amps/ft² at 191 hours. It is presently 667 mV at 160 amp/ft² 1800 hours of test.

The alternative cathode in Cell 281 had an initial measured conductivity of $1.5 (\Omega\text{-cm})^{-1}$, but a significantly higher value, $5.5 (\Omega\text{-cm})^{-1}$, at shutdown after 1100 hours of test. A calculation of cathode side resistance loss at 100 amps/ft², based on post-test cathode conductivity, is approximately 30 mV. Measured cathode side resistive loss at shutdown was 62 mV. It is concluded that the balance of the iR is contact resistance at the cathode/housing interface. Examination of the cathode under the scanning electron microscope showed no visual degradation. Mechanical testing, however, showed that crush strength had decreased from a pre-test value of 1600 psi to 860 psi. Flexural tests were not run because samples cracked during preparation. Further data on post-test cathode mechanical properties will be obtained when the endurance test of Cell 294 is complete.

A dry casting process was used to fabricate the alternative cathode in sizes up to 1-ft². The fabrication of larger components by this method appears feasible. Further scaleup will be deferred, however, to resolve the cracking problem encountered during the lithiation step.

Cell 289 was built with a cathode fabricated of Inco 255 powder especially formulated to have a high mean pore size: 9.6μ versus the more typical 7.5μ of standard cathodes fabricated of Inco 287 nickel. Initial cell performance was 716 mV at 160 amps/ft². Cell resistance was determined to be 37 mV at 100 amps/ft², and diagnostics indicated that the cathode performance was on the low side of the normal range. Surface area measurements showed the surface areas a bench in the cathodes of Inco 287 and Inco 255 nickel are equivalent, even though the Inco 255 cathodes were sintered at higher temperatures, see Table 2-3. Additional 4-in. by 4-in. cell tests are planned to verify the performance level of Inco 255 cathodes.

Development activity during the next report period will be directed at reducing the contact resistance between the alternative nickel oxide cathode and the cathode housing.

Additional work under this activity includes development of items for which patent protection is being considered. To protect their patentability, discussion of that work is not included in this report.

TABLE 2-3. BET SURFACE AREA MEASUREMENTS OF NICKEL CATHODE

Part No.	Ni Powder	Maximum Oven Temp(°F)	% Rolled	Mean Pore Size(μ)	BET Surface Area (M ² /g)
7330-22	Inco-287	1735	10	6.4	0.20
1432-17	Inco-255	1880	19	9.6	0.21
1432-16	Inco-255	1880	22	9.1	0.19

2.E SEPARATOR PLATE DEVELOPMENT (ACTIVITY 165)

Summary

Two sample 3-ft by 3-ft plates were laser welded to identify whether any limitations exist in scaling up the subscale plate configuration. These plates are not of the quality of the subscale components, but techniques to improve the welds and plate flatness have been identified. A partial evaluation of the improved techniques using sections of large plates has demonstrated improvement in quality.

A procedure for locally aluminizing separator sections for corrosion protection by a laser deposition process was shown to be feasible. The conclusion is based upon several development samples which had a uniformly distributed aluminum layer within the heat zone.

Discussion

Two complete 3-ft by 3-ft separator plates were manufactured. The fillet welds between the rail and plate were made with power settings and speeds identical to those used for 12-in. by 12-in. plates. The rail was observed to pull away from the sheet in the region about to be welded as the fillet progressed forward. This was attributed to cumulative differential thermal expansion between the sheet and rail due to the difference in their masses. Successful welds were ultimately

obtained by stitch welding, at 0.015 in. intervals, prior to making the full fillet weld. All edge welds were satisfactory. The result of this initial trial is a plate with local crimping at some fillets, a relatively sharp sheet bend at the inside of the rails, and some evidence of a burned weld, see Figure 2-5. These trials were also effecting excessive bow in the rails. To compensate for the bow, a greater degree of beam travel was required across the weld path than is desirable. Based on these elementary tests we have concluded that satisfactory full-scale plates can be welded.

The first phase of a test program to evaluate methods for repairing regions of the plate in which the aluminum protective layer has been damaged by welding was completed. Quantitative determinations show that adequate aluminum was uniformly distributed within the weld melt zone to reprotect the plate. Electron beam microprobe analysis showed 25 wt% aluminum, uniformly dispersed up to a depth of 0.012 in. in the repaired regions. The result was the same for repairs made to both plasma coated and aluminum powder coated samples.

Additional work under this activity includes development of items for which patent protection is being considered. To protect their patentability, discussion of that work is not included in this report.

Figure 2-5. 3-ft. by 3-ft. Trial Separator Plate

2.F SINGLE CELL TEST SUMMARY (ACTIVITY 114)

Discussion

Fourteen 4-in. by 4-in. cells were tested this quarter in support of component development activities of Task 2 and sulfur tolerance evaluations of Task 4. Cells 301 and 302 were tested to evaluate low cost current collector materials. Cells 291, 292, and 293 were tested as part of the matrix development effort. Cells 279, 281, and 294 were tested in the cathode development effort. Cells 266, 271, 286, 287, and 290 were tested in the anode development effort. Cells 274 and 275 were tested as part of the sulfur tolerance determination under Task 4. A summary of these cells is given in Table 2-4.

TABLE 2-4 SUMMARY OF CELL CONFIGURATION, TEST OBJECTIVES, AND PERFORMANCE

Cell	Operating Time, Hours	Description	Cell Performance mV @ 160 amp/ft ²	Cell iR mV @ 100 amp/ft ²	Peak Performance mV @ 160 amp/ft ²	Test Purpose
266	2534 (terminated)	Anode - reference design Cathode - Std Matrix - Std C/C - reference design on anode - perforated plate on cathode	607	49	713	Evaluation of reference design anode.
271	2012 (terminated)	Anode - Low Chromium Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	674 @ 100 amp/ft ²	71	746	Evaluation of low chromium content anode.
274	3680 (terminated)	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	612 (.05 ppm H ₂ S in Texaco O ₂ -blown fuel 85% fuel util.)	34	743	Sulfur tolerance test.
275	3140	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	678 (Texaco O ₂ -blown fuel 85% fuel util. .005 ppm 50 in oxidant)	33	722	Sulfur tolerance test.
278	1346 (terminated)	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	737 (Texaco O ₂ -blown fuel 57% fuel util.)	54	687	Evaluation of cell at reference design conditions.
279	1512 (terminated)	Anode - Std Cathode - reference design Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	756 @ 100 amp/ft ²	52	758 @ 100 amp/ft ²	Evaluation of reference design cathode.
281	1102 (terminated)	Anode - Std Cathode - reference design Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	670	64	681	Evaluation of reference design cathode.
286	2120 (terminated)	Anode - reference design Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate and folded sheetmetal on cathode	651	61	754	Evaluation of reference design anode.

TABLE 2-4 SUMMARY OF CELL CONFIGURATION, TEST OBJECTIVES, AND PERFORMANCE
(Continued)

Cell	Operating Time, Hours	Description	Cell Performance mV @ 160 amp/ft ²	Cell iR mV @ 100 amp/ft ²	Peak Performance mV @ 160 amp/ft ²	Test Purpose
287	1101 (terminated)	Anode - reference design Cathode - Std Matrix - Std C/C - reference design on anode - perforated plate and folded sheetmetal on cathode	595	55	721	Evaluation of reference design anode.
288	502 (terminated)	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	--	34	694	Evaluation of cell at reference design conditions.
289	2170	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	600	58	728	Evaluation of cell at power plant conditions.
290	1970	Anode - reference design Cathode - Std Matrix - Std C/C - reference design on anode - perforated plate and folded sheetmetal on cathode	653	62	749	Evaluation of reference design anode.
291	1940	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	664	58	731	Evaluation of cell at power plant conditions.
292	1690	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	663	65	743	Bubble barrier extended to outside of edge seal.
293	1360 (terminated)	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	574	105	711	Evaluation of bubble barrier edge reinforcement.
294	1570	Anode - Std Cathode - reference design Matrix - Std C/C - folded sheetmetal on anode - reference design on cathode	666	72	698	Evaluation of reference design cathode.
301	460	Anode - Std Cathode - Std Matrix - Std C/C - perforated plate on anode - perforated plate on cathode	746	30	751	Evaluation of alternative anode side material.
302	290 (terminated)	Anode - Std Cathode - Std Matrix - Std C/C - folded sheetmetal on anode - perforated plate on cathode	--	230	630	Evaluation of alternative cathode side material.

C/C - Current Collector

Std Anode - 0.060-in. thick, stabilized Ni

Std Cathode - 0.015-in. thick

Std Matrix - .018- .022-in.

Cell performance at H₂ utilization of 96%, CO₂ utilization of 30%, O₂ utilization of 21%

	Fuel	Oxidant
Dry Vol %	H ₂ - 71.5	CO ₂ - 0.18
	CO - 13.8	O ₂ - 0.13
	CO ₂ - 14.7	N ₂ - 0.69

Cells 278 and 288 were tested to define performance characteristics at reference design conditions. Both are of identical configuration. The performance of Cell 278 increased 69 mV at 160 amps/ft² after pressurization to 150 psia on simulated oxygen-blown Texaco gasifier fuel. The increase predicted was 61 mV. It operated at ambient pressure on steam reformed naphtha fuel for 950 hours. The cell voltage at the reference design conditions was 749 mV at 160 amps/ft². After 1170 hours of operation on load the cell was inadvertently subjected to a reactant overpressure of 50 psi. This was caused by rupture of a fuel inlet line dielectric isolator. The rupture was the result of overheating the isolator in turn due to a faulty heater control. The heater control has been modified to prevent recurrence of the failure. The cell retained its seal integrity and showed no crossover as a result of this overpressure. The test was terminated at this point because of severe performance loss following the reactant overpressure.

Cell 288 operated at ambient pressure for 300 hours prior to pressurization to 150 psia. The change in voltage between the ambient and pressurized condition was negligible, whereas a 50 mV improvement was predicted. After 200 hours at pressure the stand pressure regulators were upset by an electrical power interruption and the cell was subjected to a severe reactant overpressure. This event resulted in significant performance loss and reactant leakage. The test was terminated. The stand pressure regulator controls have been modified to tolerate electrical power interruption.

Discussions of the remaining cells in Table 2-4 are presented in the corresponding sections of this report.

2.G STACK AND CELL MATERIALS DEVELOPMENT (ACTIVITY 175)

Summary

Corrosion tests are being conducted in the electrolyte on various candidate materials for cathodes, separator plates, electrolyte matrix fillers, manifold seals, and stack electrical insulators. Super-FECRALLOY continues to exhibit acceptable corrosion resistance in anode and cathode environments. Analysis of 316 stainless steel oxide films show a dual layer structure, the outer layer being Fe_2O_3 and the inner larger Cr_2O_3 .

Exposure of an alternative nickel oxide cathode structure to electrolyte at 1400°F cathode conditions for 1000 hours results in little change in compressive strength, but a noticeable decrease in flexural strength.

Discussion

Separator Plate Material. Tests to determine the corrosion resistance of Super-FECRALLOY (21 Cr, 5 Al, 0.4 Y, Balance Fe) are in progress at both anode and cathode conditions in electrolyte at 1400°F. Weight and electrical resistance changes after 3,000 hours of testing are shown in Table 2-5. Super-FECRALLOY exhibits relatively low specific contact resistance at both anode and cathode conditions. Similar tests on AISI type 316 stainless steel were started to determine the specific contact resistance of the material in these test environments. The results are also shown in Table 2-5 and indicate that the Super-FECRALLOY exhibits lower specific contact resistance than 316 stainless steel. The tests at cathode potential, -150 mV, will be continued through 5,000 hours.

Corrosion of the 316 stainless steel results in the formation of a dual layer oxide shown in Figure 2-6. X-ray diffraction analyses show that the outer layer is Fe_2O_3 and the inner layer is Cr_2O_3 . EDAX analyses also show the presence of a Cr depleted area in the 316 stainless steel next to the Cr_2O_3 layer. The Cr_2O_3 layer is microcracked and the Cr_2O_3 and Cr depleted layers are porous, permitting corrosion of the base metal to continue.

Future work includes characterization of the oxide films protecting the Super-FECRALLOY materials, and a retest of thicker FECRALLOY A.

TABLE 2-5. CORROSION TEST RESULTS OF CANDIDATE SEPARATOR PLATE MATERIALS

Time Hrs.	-950 mV		-150 mV	
	Weight Change (Percent)	Specific Contact Resistance (ohm-cm ²)	Weight Change (Percent)	Specific Contact Resistance (ohm-cm ²)
<u>Super-FECRALLOY</u>				
0	--	0.02	---	0.02
250	-0.05	0.11	+0.04	----
500	-0.09	----	+0.01	0.05
1000	-0.06	0.08	+0.04	0.14
3000	+0.52	0.17	+0.02	0.07
<u>AISI Type 316 Stainless Steel</u>				
0	--	0.05	---	0.05
250	+0.60	82.8	+2.20	26,400
500	+0.60	516	+3.40	68,400
1000	+0.70	8,200	+3.90	---
3000	---	---	+4.30	7,400,000

Figure 2-6. Oxide Films on 316 Stainless Steel Due to Corrosion at Cathode Conditions (3000 hrs, 1400°F, 160 mV)

Electrolyte Fillers. Tests to determine the stability SrZrO_3 , Li_2SnO_3 and TiO_2 were completed through 3000 hours. Tests of PrO_2 , SrTiO_3 and SnO_2 are under-way in electrolyte at 1400°F at both anode and cathode conditions. SrZrO_3 exhibited significant grain growth at both anode and cathode conditions; Li_2SnO_3 , showed minimal change through 3000 hours at cathode and 1000 hours at anode conditions, and TiO_2 converted to Li_2TiO_3 at both conditions. PrO_2 showed only grain growth at cathode conditions but reacted to form a Pr perovskite at anode conditions in tests conducted for 1000 hours.

Tests of SrTiO_3 and SrO_2 have also been initiated. Samples of Li_2TiO_3 , high purity alpha Al_2O_3 and Al_2O_3 (Cr_2O_3) have been obtained and are currently being analyzed prior to corrosion testing. The test results to date are shown in Table 2-6.

Testing of the LiSnO_3 , PrO_2 , SrTiO_3 , and SnO_2 will be continued to 3000 hours.

TABLE 2-6. CORROSION TEST RESULTS OF ELECTROLYTE FILLER MATERIALS

Material	Time (Hrs)	Recrystallization *		Crystalline Structure	
		Anode	Cathode	Anode	Cathode
SrZrO_3	0	--	--	Cubic SrZrO_3	
	1000	Increasing	Increasing	95% Orthorhombic	80% Orthorhombic
	3000	Increasing	Increasing	90% Orthorhombic	80% Orthorhombic
Li_2SnO_3	0	--	--	Monoclinic Li_2SnO_3	
	1000	Increasing	Increasing	Monoclinic	Monoclinic
	3000	--	No Further Change	--	Monoclinic
TiO_3	0	--	--	Tetragonal TiO_2	
	1000	Increasing	Increasing	90% Li_2TiO_3	95% Li_2TiO_3
	3000	Increasing	Increasing	100% Li_2TiO_3	100% Li_2TiO_3
PrO_3	0	--	--	Face Centered Cubic $\text{PrO}_{1.8}$	
	1000	Increasing	Increasing	Orthorhombic Pr Perovskite	Face Centered Cubic
SrTiO_3	0	--	--	Simple Cubic SrTiO_3	
SnO_2	0	--	--	Tetragonal SnO_2	

* Actually a measurement of grain growth.

Electrode Materials. A test was also performed to determine the effect of electrolyte exposure on strength of a lithiated nickel oxide cathode. The test was conducted by placing cathode specimens in contact with an electrolyte-filled matrix, applying a dead load, to approximate the compressive stresses encountered in cell operation, and heating the specimens to and holding them for various times at 1400°F all at cathode potential. Table 2-7 is a summary of the results.

TABLE 2-7. FLEXURAL AND COMPRESSIVE STRENGTH OF NiO CATHODE

<u>Strength (psi)</u>	<u>Exposure Time (Hours) at Cathode Conditions</u>			
	<u>0</u>	<u>250</u>	<u>500</u>	<u>1000</u>
Compressive	940	860	870	860
Flexural	525	430	340	---

These data show that no significant changes are occurring in compressive strength there is, however, a decrease in flexural strength. The test will be continued to determine if the flexural and compressive strengths have stabilized.

Additional work under this activity includes materials for which patent protection is being considered. To protect their patentability discussion of these materials is not included in this report.

2.H STACK HARDWARE DEVELOPMENT (ACTIVITY 180)

Summary

Stack hardware requiring development includes the manifold gas seal, dielectric isolators, pressure vessel power penetrators and dielectric pipe connectors. The dielectric pipe connectors being prepared for subscale stack test evaluation in EPRI program RP 1273 developed cracks in several ceramic parts. Subsequently they leaked during a thermal cycle. A modified connector successfully completed a thermal cycle.

A test rig for evaluating the leakage and mechanical properties of the manifold seal was obtained. It is being installed and readied for testing.

Specimens of nickel coated Glidcop bus bar material were prepared for evaluation in a subscale stack test under EPRI program RP1273. A 4-in. by 4-in. manifold seal sample underwent two thermal cycles with no leakage.

Dielectric tests of the best potential manifold coating to date were repeated with a fully coated sample. The retest was conducted to eliminate a possible leakage path which may have caused the failure of the previous partially coated samples. The new data showed even higher conductivities. Manufacturing differences in the samples are being studied to determine if they can explain this unexpected result.

Discussion

A dielectric piping connector was thermally cycled to 1300°F. Leakage was not observed during heat up, but as the part was cooling leakage began and increased with additional temperature drop. Upon disassembly, one ceramic component was found to be cracked. The design was reviewed for stresses due to differential thermal growth. No design discrepancies were found, however the sensitivity of the design to the thermal expansion characteristics of the component materials became more obvious. Several modifications were identified to minimize the vulnerability of the design to thermal stresses and were incorporated into the hardware. A subsequent thermal cycle resulted in no leakage or part failure. Four connectors have been modified and prepared for the next stack test under EPRI program RP1273.

The present manifold seal material has performed satisfactorily during several subscale stack tests and remains the primary seal material candidate. Current development efforts are aimed at evaluating the characteristics of the seals. The effects of thermal cycling, surface roughness and variable compressive loading on seal effectiveness are being determined. The first of a series of 4-in. by 4-in. gasket tests demonstrated the capability of filled gaskets to maintain a seal after

two room temperature to 1300°F thermal cycles. The initial leakage was 18 sccm per inch of perimeter. This dropped to less than 0.1 sccm at 1200°F by the second cycle. The seal was compressed to 20% of its free height. Additional tests of this type are planned.

Dielectric tests of a 20% magnesium-bal. zirconia flame sprayed coating for the manifold seal were repeated using a fully coated sample. Prior tests on a sample coated only on one side gave inconclusive results. An electrolyte path may have been established to the uncoated surface. A dry test initiated at 735°F and completed at 1240°F showed that resistance decreases with increasing temperature. At 1240°F a current of 100 mA/cm² was measured of a potential of 20 volts. With electrolyte present the current measured was twenty times that obtained earlier with the partially coated sample at the same 20 volt test condition. Microphotos of this coating were compared with those of the earlier test samples, and significant topographical differences were noted. These photos are being reviewed to determine if the second coating samples were properly manufactured.

Additional work under this activity includes development of items for which patent protection is being considered. To protect their patentability, discussion of this work is not included in this report.

2.1 ANALYTICAL DESIGN OF CELL AND STACK COMPONENTS (ACTIVITY 116)

Summary

Design of the full-scale cell and short stack of full-scale cells was started during the quarter. The full-scale short stack pressure vessel was placed on order.

A detailed thermal analysis of the power takeoff system indicates that the power penetrator selected for the containment vessel meets applicable design and electrical codes.

Construction drawings were completed for the pressure vessel for testing twenty cell subscale stacks at 150 psia.

Discussion

Full-Scale Cell and Stack Design. Design of the twenty-cell short stack of full-scale cells was started during the quarter. The purchase order for the vessel, which is the longest lead-time item for the test, was placed. Work has concentrated on the analytical design of the cell stack hardware such as, end plates, manifolds, insulation, power penetrators and the endplate heater requirements. The design of some of these components, such as the endplates and the tie rod system, cannot be finalized until the cell package is better defined.

Stack Hardware. A thermal analysis of the design of the power takeoff system for the full-scale prototype stack was completed. The "power penetrator", which conducts power through the pressure vessel walls to the exterior bus, must not operate at a temperature in excess of 600°F in order to meet the National Electric Codes in the presence of combustible fuels. The penetrator selected for this application is the same penetrator used in the 4.8-MW phosphoric acid fuel cell power plants pressure vessels. The analysis, which calculates the temperature distribution of the penetrator and vessel, shows that the housing designed for the power penetrator provides sufficient cooling to satisfy the requirements of the National Electric Code. The analysis accounts for conduction, convection, and radiation heat transfer. The temperature profile calculated for the vessel and penetrator assuming a 150°F ambient temperature is shown in Figure 2-7.

Subscale Stack Design. Construction drawings were completed for an upper dome designed to fit the lower half of an existing pressure vessel. This unit will be used for testing subscale stacks of ten or twenty cells at up to 150 psia operating conditions.

Additional work under this activity includes development of items for which patent protection is being considered. To protect their patentability, discussion of the work is not included in this report.

Future effort in this activity will continue to address the design of alternative configurations for the full-scale cell, and will complete the construction drawings for subscale and full-scale stack hardware. Analysis of structural test data for cell components will continue, in support of the cell configuration design effort.

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Figure 2-7. Molten Carbonate Fuel Cell Penetrator Thermal Analysis

Section 3

TASK 3 - DEVELOP CAPABILITY FOR FULL-SCALE STACK TEST

3.A DEVELOP LOW COST MANUFACTURING METHOD FOR PRODUCING ELECTROLYTE MATRICES

Summary

Studies conducted in Task 1 of this program established a reference design cell area of approximately 9-ft². Fabrication of this size cell requires a matrix at least 36 inches wide. Subscale fabrication trials on available equipment have been sufficiently successful to warrant procurement of larger equipment for development of full-size matrices. An appropriate unit has been ordered, with delivery scheduled for early March. Layout of the process line and design of ancillary equipment is underway. Its completion is scheduled to coincide with the arrival of the purchased equipment.

Discussion

Equipment limitations preclude manufacture of one piece electrodes for large area cells. Previous experience, however, indicates electrodes may be pieced to form composite electrodes of large size. It is desirable that the matrix be a single unseamed component to provide a continuous barrier to reactant cross-leakage. A one-piece matrix also eliminates alignment problems on assembly. The major effort during this quarter was devoted to evaluating, acquiring and designing equipment to make tape-formed matrices in widths up to 48 inches.

A major item of ancillary equipment for the matrix process development line is the accumulator shown in Figure 3-1. It stores the matrices in a controlled environment. Accumulator design has been completed, and the unit is currently under construction.

Work during the next quarter will be devoted primarily to installing the process equipment and verifying that it is ready for full-scale matrix development.

A portion of the work under this activity includes development items for which patent protection is being considered. To protect their patentability discussion of that work is not included in this report.

Figure 3-1. Accumulator for Molten Carbonate Tape-Matrices

Section 4

TASK 4 - DEVELOP CAPABILITY FOR OPERATION OF STACKS ON COAL-DERIVED GAS

4.A SUMMARY

The molten carbonate fuel cell power plant will operate using gasified coal. In this task, reactant gas purity specifications are developed which allow the operation of the molten carbonate fuel cell without adverse effects of impurities in the coal gas.

During this quarter, tests designed to determine the effects of 0.005 ppm SO₂ and 0.05 ppm H₂S on cell performance were completed. Hardware for the sulfur parametric study has been fabricated and is available for cell testing.

Also, the value of the equilibrium constant for the reaction of H₂S with molten carbonate was determined over a range of temperatures and electrolyte compositions.

4.B SULFUR STUDIES (ACTIVITIES 124 & 129)

Summary

Cell 275 has been operated for 2500 hours on oxidant containing about 0.005 ppm SO₂ without decay. In contrast, Cell 274 experienced a loss of about 100 mV within 1700 hours of operation under simulated O₂-blown Texaco fuel gas and 0.05 ppm H₂S. Bulk exit gas analysis indicated that the fuel was in water-gas-shift equilibrium during the decay. This result is contrary to previous cell tests at higher H₂S contents which showed that a major part of the performance loss was due to the inhibition of the shift reaction with consequent partial starvation of the anode. Subsequent testing of Cell 274 on sulfur-free, hydrogen-rich fuel resulted in complete iR-free performance recovery.

Discussion

Figure 4-1 shows the performance plot of Cell 275. Voltage readings at 160 amp/ft² fluctuated between 675 mV and 690 mV during its 2500-hour test on sulfur. The 0.005 ppm SO₂ value is a calculated number based on the permeation tube manufacturer's specifications and the cells operating conditions. No attempt was made to measure the SO₂ content in the oxidant gas because of the very low anticipated concentration.

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Figure 4-1. Cell 275 Performance on 0.005 ppm SO₂ (Nominal) 1200°F, 1 atm

A similar performance history of Cell 274 is illustrated in Figure 4-2. This cell lost approximately 100 mV at 160 amp/ft² in a minimum of 2200 hours of operation on simulated O₂-blown Texaco fuel with a nominal 0.05 ppm H₂S. The length of time is in question due to the fact that the permeation tube emptied prematurely.

After 2000 hours on test, the N_2 carrier gas passing through the permeation tube was analyzed for H_2S . The quantity found was very close to the value calculated to give 0.05 ppm after dilution with the wet fuel stream. Performance at that time was 685 mV at 160 amp/ft² so that the performance decayed 22 mV while on sulfur. Approximately 700 hours later the permeation tube was examined and found to contain no liquid H_2S . The exact time the permeation tube emptied is not known. However, during this time period the cell decayed an additional 6 mV; therefore, the loss of H_2S had little or no effect on performance. Calculations based on measured permeation rate and on manufacturer's information indicated that a new tube should have lasted at least 1000 hours longer.

Figure 4-2. Cell 274 Performance on 0.05 ppm H_2S (Nominal) 1200°F, 1 atm

A new permeation device was placed on test. The cell continued to decay with an additional 70 mV loss in the next 700 hours. Gas analyses conducted during this period indicated that the water-gas-shift reaction was near equilibrium, see

Table 4-1. This is in contrast to previous cell tests at higher H₂S contents which showed that a major part of the performance loss was due to inhibition of the shift reaction with consequent partial starvation of the anode. A second H₂S analysis of the N₂ stream near the end of the test resulted in a lower value which translated to a 0.03 ppm level after dilution. Equipment and sampling problems prevented measurements of H₂S at the fuel cell exit.

TABLE 4-1. BULK EXIT GAS ANALYSIS OF CELL 274

Temperature 1200°F, 1 atm simulated oxygen-blown Texaco fuel saturated at 185°F dewpoint 85% utilization at 160 amps/ft ²		
Gas	Analysis	Calculated Assuming Shift Equilibrium
H ₂	6.5	5.4
CO	1.4	2.3
CO ₂	72.1	75.5
N ₂	20.0	16.8

At this time the cell was placed on sulfur-free, hydrogen-rich fuel resulting in complete iR-free performance recovery. This strongly suggests that much of the decay was due to the presence of H₂S, but by a mechanism other than that of inhibition of the water-gas-shift reaction as previously found.

A summary of test results for cells operating under simulated oxygen-blown Texaco fuel with various levels of inlet H₂S is shown in Figure 4-3. In this Figure, the time before rapid cell decay can be seen to vary with between the -3/4 and -1 power of the H₂S content in the fuel.

Figure 4-3. Effect of H₂S on Cell Performance

Cell hardware for the sulfur parametric study has been fabricated, and is available for cell testing.

Planned activity for the next quarter includes:

- o Start of sulfur parametric study
- o Improvement of analytical techniques to measure very low levels of H₂S, SO₂, CS₂, and COS.
- o Improvement of station design to provide better control of gas flows and permeation tube temperatures and to facilitate gas analyses.

Equilibrium Studies. One of the mechanisms by which H₂S affects the molten carbonate cell is the conversion of the carbonate to sulfate by the reaction:

thereby changing the carbonate ion activity and thus the thermodynamic potential of both electrodes. The value of the equilibrium constant for this reaction has been determined to permit these losses to be calculated for various cell operating conditions.

The measurement of the equilibrium constant, K_{EQ} , for the reaction of H_2S with molten carbonate electrolyte was completed for temperatures of 1100, 1200, and 1300°F, and CO_3^{2-}/SO_4^{2-} ratios of 4, 1, and 0.67. The results are presented in the Table 4-2.

TABLE 4-2. MEASURED EQUILIBRIUM CONSTANTS

T (°F)	CO_3^{2-}/SO_4^{2-}	K_{EQ} (atm)
1100	4	0.50
1200	4	0.04%
1300	4	0.0024
1200	0.67	0.092
1200	1	0.012

These values for K_{EQ} are much smaller than those reported to EPRI in a previous study* where values from 0.05 to 19.2 were obtained. The effect on electrolyte sulfation is shown below:

	K_{EQ}	% Conversion to Sulfate	
		Inlet	Exit
EPRI	max 19.2	6×10^{-2}	87
Study	min .05	1.6×10^{-4}	1.7
This study	.045	1.4×10^{-4}	1.5

*Proceedings of DOE/EPRI Workshop on Molten Carbonate Fuel Cell, No. 1979
EPRI WS 78135

The reasons for the high values of K_{EQ} obtained previously and the large variation with experimental conditions is not known with certainty, but slow removal of the sulfate contained in the starting salts may have been the problem. The deep bucket design of the sample container used in that study could have contributed to the slow conversion of sulfate to gaseous sulfide by reducing the amount of convection in the sample.

Two major conclusions can be reached from these data concerning the effect of H_2S on the fuel cell:

- o Because the value of K_{EQ} is small and it decreases at high temperatures (corresponding to cell exit conditions where P_{H_2} is small) the amount of $SO_4^{=}$ formed will be relatively small, even at the exit. About 0.5% $SO_4^{=}$ will be formed for each ppm of H_2S for 2% H_2 . Since the H_2S concentration must be limited for other reasons, sulfate formation will not result in significant thermodynamic loss.
- o The low sulfur capacity of the melt means that it will not act as a sump for sulfur. Therefore, in systems operating without venting of the anode exit or without anode exit H_2S scrubbers, the H_2S pressure will build more rapidly and require more frequent clearance cycles to remove the sulfur if performance degradation due to sulfur is experienced.

The apparatus used in these experiments is shown schematically in Figure 4-4. A small quantity (ca. 5) of the appropriate mixture of carbonate and sulfate was placed in a crucible furnace. A gas atmosphere was mixed of H_2 , to which the H_2S was added, and CO_2 which contained the necessary water vapor to closely correspond to the expected equilibrium composition. Equilibrium was reached by adjusting the H_2 flow rate until $P_{H_2S} (in) = P_{H_2S} (out)$. These partial pressures were small such that a negligible change in melt composition occurred during the test. The inner nickel tube allowed gas samples to be removed for analysis to determine the extent of shift equilibrium at a point just above the melt.

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Figure 4-4. Apparatus for Measuring Equilibrium Constant

The preliminary values reported for K_{EQ} in the previous quarterly report were based on the assumption that essentially no shift would occur in the incoming gas because of the short residence time in the hot zone of the inlet tube. Samples taken through the nickel tube demonstrated that this assumption was essentially correct: the degree of shift was only a few percent.

The time required to attain equilibrium by altering the gas composition was much more rapid than by modifying the melt composition. In that method the time to reach steady state after each addition was long (over 24 hours in some cases), probably due to concentration gradients caused by differences in density of the added salts.

During the next quarter, measurements of the effect of H_2S on anode kinetics will be resumed.