MASTER

PHOSPHORIC-ACID FUEL-CELL STACK

AND SYSTEM DEVELOPMENT

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ABSTRACT

This Technical Progress Report covers the period January 1 - March 31, 1979. Primary emphasis during the period was on stack component development - Task I. However, effort is accelerating in the fuel processing sub-system area - Task II. Integrated system analysis - Task III - has begun, but the stack component scale-up effort - Task IV - will not start until later in 1979.

Two three-cell stacks utilizing nominal 0.46 mg Pt/cm^2 electrodes were started on-test during the quarter. Routine electrolyte-additions were employed for one stack which exhibited good stability at 150 amp/ft^2 (161 ma/cm^2) over the $325\text{-}375^\circ\text{F}$ ($163\text{-}191^\circ\text{C}$) temperature range. Stack temperature was recently increased to 400°F (204°C), and this led to a noticeable decline in voltage.

The second stack was subjected to shutdown/start-up cycles without electrolyte addition. Two cells exhibited good tolerance over the 11 cycles that were performed. The other cell lost considerable performance because of electrolyte flooding of the cathode.

CVD-upgraded reticulated vitreous carbon elements have shown good mechanical and electrical properties. Air permeability is sufficient to permit service as a non-grooved bipolar plate element. The RVC was found not to have good phosphoric acid tolerance, but CVD coating appears to offer protection.

Laminated matrix layers have served to lower the cell IR-losses substantially - to about 40 mv @ 150 ma/cm^2 . Porous carbon-fiber acid-transport layers are being used between two matrix layers to improve in-plane electrolyte-migration. Configurations for on-board electrolyte-storage are in the evaluation stage. One approach appears promising over the first 1000 hours in a single-cell test.

Base-line nominal 10% Pt on carbon catalysts have shown rapid Pt surface-area loss during the first 50 hours of aging in phosphoric acid at 0.7 volts (RHE). Subsequent area-loss up to 500 hours is very small. There is a wide disparity in surface-area between samples aged at 325°F (163°C) and those aged at 400°F (204°C).

Methanol has been selected as the fuel to be used in the fuel processing and system development portions of this program. A new sub-scale test station for parametric steam-reforming studies is near completion. A test facility for integrated burner/reformer design studies is being prepared.

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INTRODUCTION

This program is directed toward the upgrading of phosphoric acid stack component technology in association with fuel cell system development effort. Work began in October of 1978 under the joint sponsorship of DOE and Engelhard; some activity was carried out earlier in 1978 under a preliminary spending agreement. This report covers work conducted during the second quarterly period, from January 1 through March 31, 1979.

The primary emphasis to-date has been placed on the stack component development effort - Task I. In the bipolar plate area, initial single-cell and stack testing has utilized a carbon-sealed graphite material that had been evaluated under a previous contract. Although results have been very satisfactory, the development effort has focused on different approaches because of the high cost of this material. Presently, resin-sealed graphites and multiple-element (ABA) plates are of primary interest.

The matrix effort is being directed toward laminate forms since these have shown better stability and lower IR-losses than the membrane-type matrix. Emphasis is being placed on the means for acid-replenishment of the matrix.

Electrodes having a precious metal loading of 0.5 mg Pt/cm² (each electrode) are presently being evaluated in the program. The temperature range is 163-204°C, although most testing to-date has been conducted at 163°C. Catalysts are being supplied to the program through a complementary effort - Task V - that is wholly sponsored by Engelhard. Base-line nominal 10% Pt on carbon catalysts are being utilized for both anodes and cathodes. Advanced catalyst preparations are under development in Task V, and these will be utilized in Task I as appropriate.

Single-cell testing in subscale (3 in. x 3 in.) and nominal one-half square foot cells is proceeding as the main diagnostic evaluation of components under Task I. Also, small stack testing (three-cell) has begun using the nominal one-half square foot cells. Six sub-stacks (10 or 12 cells) will be run under the program as well as two 5-KW stacks.

A 5-KW fuel processor will be constructed and tested under Task II. Initial activity consisted of thermodynamic equilibria determinations and preliminary system analyses for natural gas, methanol, and naphtha systems. These efforts have been completed. Methanol has been chosen as the fuel for the 5-KW processor. Sub-scale reformer testing will begin at the end of April.

System analysis for an integrated methanol-air powerplant has begun - Task III. The system design will be directed toward an on-site integrated energy system application.

Later in 1979, scale-up activity for phosphoric acid stack components will begin - Task IV. Technology developments of Task I will be directed toward application in large-area cells (four to six square feet).

TASK I - PHOSPHORIC ACID STACK COMPONENT DEVELOPMENTSTACKS

The first three-cell stack (G08-03-01) was started up during the quarter and continued in operation through March. It has accumulated about 1600 hours on load. Details of the components and the stack configuration are given in Table I. The endurance history to-date is presented in Figure 1.

Stability in the range 163°C to 191°C was reasonably good throughout the test-period. Recent performance decline at 204°C is attributable primarily to the middle cell as shown in Figure 2. Decay in this cell appears to result from corrosion of niobium shims that were used to structurally protect the electrode edges, causing reactant gas groove-blockage. Subsequent stack testing has shown that the electrode edge protection is not required, and the shims will be eliminated from future builds.

The open-circuit voltage stability is shown in Figure 3. Although the initial open-circuit voltages were low, these have not declined severely with time. Routine acid-addition procedures have been employed. Total running time at 191°C and above has been about 900 hours. The results from the testing of this stack tend to confirm the viability of the present stack components for application in larger stacks. (However, the high initial open-circuit voltages typically obtained in sub-scale single-cell tests have not been duplicated in stacks constructed to-date; and matrix fabrication techniques are being optimized in order to obtain high-quality elements in stack-size cells.)

The second three-cell stack (G08-03-02) that was started up during the quarter had identical construction to that of the first stack. This stack has been utilized to examine the tolerance of this matrix type to upsets in the form of shutdown/start-up cycling. Acid-additions have been withheld during this testing in order to observe the basic durability of this configuration.

The endurance plot is shown in Figure 4. The 11 shutdown cycles employed thus far are indicated on the graph. Losses have been predominantly attributable to the top cell; these appear to be caused primarily by electrolyte flooding of the cathode. Loss of wetproofing in the cathode substrate is thought to be responsible.

Performance losses in the middle and bottom cells have not been excessive, despite the shutdowns. This indicates a substantially improved matrix durability in comparison to the membrane-type matrix. As in the case of Stack G08-03-01, initial open-circuit voltages were low; but, here again, these have not decreased severely with time - see Figure 5.

BIPOLAR PLATES

Goals for the bipolar plate program are as follows:

- (i) Electrical resistivity of 0.05 ohm-cm (about 3 mv IR-loss at 150 ma/cm²) or less

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- (ii) Hydrogen permeability equivalent to 0.5 ma/cm² or less at 204°C
- (iii) Flexural strength of 4000 psi or greater (ref. ASTM-D790-71)
- (iv) Weight loss of 0.1% or less in phosphoric acid at 204°C and 1.0 volts (RHE)
- (v) Projected high-volume cost of \$4/ft²

Apparatus used to evaluate categories (i), (iii), and (iv), above, are shown in Figures 6, 7, and 8, respectively.

The bipolar plate development effort in this program falls into three general categories:

- (i) All-carbon plates
- (ii) Resin-bonded and resin-sealed plates
- (iii) Multiple-element plates (ABA-type)

The discussion which follows is broken down accordingly.

All-Carbon Plates

The all-carbon plates investigated and/or utilized in this program to-date include carbon-sealed graphite (Tylan*), vitreous carbon, and vitreous carbon impregnated graphite (Vitrecarb). The disposition of these materials in terms of the properties of interest is presented in Table II. Phosphoric acid tolerance is discussed separately.

As indicated in Table II, all of the candidate materials are attractive in terms of electrical resistivity and flexural strength. The carbon-sealed graphite and the vitreous carbon materials have also shown hydrogen impermeability; and Table V indicates that their phosphoric acid tolerance is good. However, the projected cost for the carbon-sealed graphite is unacceptably high (about \$30/ft²), and this material will continue to be used only to support the experimental program. Likewise, the vitreous carbon material is too expensive (over \$50/ft²), and recent estimates of possible cost-reduction procedures by the processor do not indicate a change in this picture.

In the case of the vitreous carbon impregnated graphite, four levels of impregnation were examined. As shown in Table II, none of these had sufficient hydrogen impermeability; therefore, phosphoric acid tolerance testing was not conducted. Furthermore, cost estimates from the processor - based on preparation of these samples - were not attractive, and effort on this approach has been discontinued.

* U.S. Patent 3,854,979

Resin-Bonded and Resin-Sealed Plates

The physical properties of the resin-bonded and resin-sealed materials are shown in Table III. The phenolic-bonded graphite was molded by Sparta Manufacturing Company at a resin content of 25% by weight. Electrical resistivity was acceptably low; hydrogen impermeability and flexural strength have not yet been determined. Phosphoric acid weight-loss was high, as seen in Table VI. Further evaluations of phosphoric acid tolerance will be conducted under less severe conditions.

The polyphenylene sulfide-bonded graphite material was also molded by Sparta Manufacturing Company at a resin content of 25%. It was found to be attractive in terms of electrical resistivity, hydrogen impermeability, and flexural strength (see Table III). However, Table VI indicates that phosphoric acid tolerance is poor under the severe potentiostatic conditions. This material will also be examined under less severe conditions.

Graphite impregnated with Kearney Industries' CO-40 proprietary resin showed good electrical, impermeability, and strength properties (Table III). It can be noted from Table VI that its degree of tolerance to phosphoric acid is as yet uncertain. A sample that had non-impregnated edges lost 0.7% of its weight after 1000 hours at 204°C and 1.0 volts; however, in the next 1000 hours this sample lost an additional 9.5% of its weight. There was evidence of physical disengagement of carbonaceous particles in the acid-containing vessel, and the non-impregnated edges could have been a complicating factor.

A sample of the same material but with impregnated edges was subsequently started on-test. No weight-loss measurements have been made thus far, but the corrosion-current history through the first 500 hours is shown in Figure 9. The corrosion current at the 500-hour point is about 0.085 mA/g (Table VI). Based on the conversion of carbon to carbon dioxide, this current would result in a weight-loss of about 1% in a 1000-hour period.

Also shown in Figure 9 is the corrosion-current history of the Great Lakes graphite precursor (i.e., without resin impregnation). The lower corrosion current of this material indicates that the corrosion current of the resin-impregnated graphite - although not severe - is mainly attributable to the resin.

A Tafel plot of the corrosion current was taken at the 328-hour point, and the results are shown in Figure 10. The Tafel slope was very high - about 400 mv/decade - indicating that improvement in corrosion resistance as a result of operating at a lower potential is likely to be relatively minor. Testing is continuing.

In order to accelerate the overall evaluation of the CO-40/graphite material, a single-cell test using these plates was placed on-test. The results were encouraging, as shown in Figure 11. The cell ran for about a week at 325°F (163°C) with virtually no loss in performance. As a result of this, further single-cell testing of this material will be conducted. The tests will be of longer duration, and the effects of higher temperature will be examined.

Table VI shows the phosphoric acid tolerance data for polyether sulfone to-date. The immersion-test data indicate low, but significant weight-loss. Longer-term testing is required in order to see trends. In the potentiostatic testing, weight-loss results are not available yet; but the corrosion-current history is shown in Figure 12. The corrosion current of 0.085 mA/g after 840 hours suggests weight-loss over the 1000-hour period will be on the order of 1%. This would be well above the goal of 0.1%, but the relatively low Tafel slope for this process shown in Figure 13 - about 120 mv/decade - is very encouraging. This indicates that substantial reductions in corrosion rate are likely as the potential is reduced from 1.0 volts. Testing will continue.

A polyparabanic acid film of 0.010-inch thickness has also tested - see Table III. Processing temperatures have not been optimized yet, but it appears that these should be slightly higher than that which is most suitable for polyether sulfone. Unfortunately, initial phosphoric acid weight-loss results from the 1000-hour immersion test are quite discouraging. As seen in Table VI, the weight-loss at 204°C was almost 10% over the period. No additional effort involving the use of this polymer is planned.

Multi-Element Plates

Effort to-date in the area of multi-element plates has focused on the porous, non-grooved "A-element" approach in an "ABA" configuration - see Figure 14. Goals for the A-element and B-element in such a configuration are shown in Table IV. Reticulated vitreous carbon (Chemotronics) is an attractive precursor for the A-element since its large pore, open structure is amenable to low pressure-drop.

Although untreated RVC has inadequate mechanical and electrical properties, CVD-upgraded versions of this material appear promising. Table VII shows the properties of two RVC candidates that had been upgraded to 6-8 times their original weight with chemically-vapor-deposited carbon. Each incurs a small penalty because of excess IR-loss. Beyond that, both candidates are acceptable; but the 45 pore/inch material has superior margin in P, while (although not yet measured) the 60 pore/inch material is expected to have superior margin in crush strength. Both types have been utilized in single-cells; typical cell-voltage and IR-loss values were obtained at normal reactant gas flow-rates and cell compressive-loadings.

The phosphoric acid tolerance of RVC A-elements is shown in Table VIII. The weight-loss of the first sample of CVD-upgraded RVC was found to be a surprisingly high 9% after 1000 hours at 204°C and 1.0 volts. Subsequent observations indicated that attack of the RVC skeleton was responsible for the weight-loss. Microscopic examination of the aged material showed a loss of "crispness" in the RVC phase at the cut edges. Microporosity was found to be 0.08 cm³/g in the aged sample as opposed to zero in a fresh sample. Finally, a potentiostatic aging test of an untreated RVC sample resulted in disintegration of the sample (Table VIII).

The corrosion-current history of a second CVD-upgraded RVC sample is shown in Figure 15. Weight-loss has not yet been determined, but the high corrosion current suggests that material is being lost at a substantial rate. Figure 16 shows Tafel plots taken at 24 and at 96 hours. In both cases two different corrosion mechanisms appear to be taking place - one with a Tafel slope of about 700 mv/decade that predominates below 0.9 volts, and the other with a slope of about 90 mv/decade that predominates above 0.9 volts.

Figure 17 shows the corrosion-current history of a CVD-upgraded RVC sample that had been protected at all edges by CVD-coating. It is seen that the corrosion current is greatly diminished as a result of the CVD coating at the edges. Figure 18 is a Tafel plot for the same sample after 24 hours. The Tafel slope resembles that of the low potential segment of Figure 16, but corrosion current is lower - even in this region. It appears CVD-coating throughout greatly enhances the phosphoric acid tolerance of these candidate A-elements.

Potentiostatic testing of the CVD-upgraded RVC material will continue, and phosphoric acid tolerance at the hydrogen potential will be examined. This will help determine the suitability of this material for acid-storage function (see "Matrix", below). The options for establishing viability at cathode potentials at this time are complete CVD protection, wetproofing, and alternate precursors.

A number of 7 inch x 10.7 inch RVC A-elements upgraded throughout with CVD will be available in late April (45 pores/inch). Plans call for evaluating the functional suitability of this material in a three-cell stack.

Development effort on the B-element (separator) for the ABA-type bipolar plate has focused on the woven fabric precursor material. Attempts to seal off the porosity through the use of CVD alone were unsuccessful. Bubble test observations indicated random pin-holes.

Subsequent sealing approaches utilized resin impregnation prior to CVD treatment. Testing for hydrogen impermeability yielded encouraging results. Figure 19 shows that three of four samples exhibited no leakage of hydrogen at 204°C; even the fourth sample showed a minor leakage rate. Optimization of the processing techniques is presently in progress.

Electrical resistivity measurements on finished B-elements yielded an average IR-loss of about 1 mv @ 150 ma/cm² (four samples), meeting the pre-established goal for this element. Handleability is excellent with a substantial amount of flexibility being present in these pieces. Also, phosphoric acid tolerance is excellent, as shown in Table VIII.

Bonding of the B-elements to the A-elements during final processing has not yet been successful. Efforts are continuing, but the elimination of warping must be worked out. The initial 7 inch x 10.7 inch B-elements will be available in early May, but they will not be joined to the A-elements. The initial three-cell ABA stack will address the problem of joining these elements during stack assembly.

MATRIX

Goals for the matrix development program are as follows:

- (i) 30 mv IR-loss at 150 ma/cm²
- (ii) Operability in phosphoric acid at 177-204°C
- (iii) Projected high-volume cost of \$3/ft²

Effort under this program has involved two approaches: (i) formulations based on the existing Engelhard membrane-type matrix (U.S. Patent 3,453,149) and (ii) formulations based on lamination of matrix layers onto other cell components. Emphasis during this quarter has been directed toward the latter approach. The incentives for pursuing this approach include: (i) substantially lower cell IR-losses, (ii) superior structural stability at 163°C and beyond, (iii) better acid-wicking properties, and (iv) lower projected high-volume cost.

Various inorganic materials - mostly carbides and nitrides - have been obtained for examination as candidate filler materials. Silicon carbide is being used in the first phase of the experimental program. Pastes were made up using TFE-30 Teflon as a binding agent. Most matrix preparation methods utilized to-date have involved the application of matrix layers onto both cell electrodes by various techniques.

Initially, matrix pastes were simply brushed onto the surface of the electrode catalyst layer, but these were found to be too prone toward pin-hole imperfections. Electrodes of single-cell size (3 inch x 3 inch active area) were then laminated with matrix material using contact screen-printing techniques. This approach provided a degree of reliability such that optimization procedures were pursued on this basis. Within a short time, all samples prepared passed the 10-inch H₂O bubble-pressure screening test (see Figure 20), and evaluation proceeded to the single-cell stage where open-circuit voltage is the key indicator of matrix integrity.

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Initial open-circuit voltages were somewhat low - about 860 mv @ 163°C - but these were quite steady over the short-term. This performance was subsequently improved by utilizing finer particle-size filler material. Table IX shows the average open-circuit voltage and the maximum deviation as a function of filler particle-size. These data - acquired from about 50 single-cell tests - illustrate the greater magnitude and dependability of open-circuit voltage as filler particle-size is reduced.

The typical thickness of these matrix layers is about 0.005 inches, and the total matrix thickness is therefore about 0.010 inches. This compares to a typical thickness of about 0.020 inches for the membrane-type matrix. Consequently, cell IR-losses have been greatly diminished - to about 40 mv @ 150 ma/cm², as compared to about 70 mv.

Since these laminated matrix layers are quite thin, their acid-inventory capacity is low. It is therefore necessary to implement the means for acid-replenishment in this type of cell. Figure 21 shows three candidate configurations for providing additional acid to the matrix.

Single-cell test results for Configuration B were quite satisfactory over the short-term running on hydrogen and air at 163°C. These have not yet been evaluated on reformat-type fuels at reasonably high hydrogen utilization rates. Testing of Configurations A and C has been discontinued because excess IR-losses have been obtained.

Two 1800-hour single-cell tests have been run using an innovative approach.* The results of the first test (Cell No. 444) are shown in terms of the open-circuit voltage stability in Figure 22. The relatively coarse filler particles were used in this matrix, and initial open-circuit voltage was somewhat low. Nevertheless, open-circuit voltage remained quite steady through the first 46 days. Cell voltage, on the other hand, began to decline after the first 32 days, as shown in Figure 23. This resulted primarily from acid-loss, which caused a substantial increase in the IR-losses of the cell.

Cell No. 444 was disassembled after 46 days and acid was added. Performance on load immediately improved (Figure 23), but open-circuit voltage was apparently adversely affected by the cell disassembly and restart (Figure 22). The results of this test illustrate the necessity of providing acid-storage or acid-replenishment (ref. A-element storage tests discussed below).

* The details are not disclosed because of possible patent action.

The other 1800-hour single-cell test (Cell No. 461) showed higher initial open-circuit voltage as shown in Figure 24. This is attributable to the finer particle size of the matrix filler material. Also, open-circuit voltage remained quite steady until a retorquing of the cell after 64 days - despite the fact that no acid-additions had been performed. However, Figure 25 indicates that - as in the case of Cell No. 444, above - cell voltage on load had shown previous declines. These, again, are attributable to increasing cell-IR from acid-loss.

Subsequently, plans for extended single-cell tests incorporated provisions for either on-board acid-storage or routine acid-addition without cell-disassembly. The latter was practiced successfully in Stack G08-03-01 (above) at both 163°C and 191°C.

Acid-storage concepts making use of the 60 pores/inch CVD-upgraded RVC A-element are now being evaluated. Excess acid has been added to the anode-side A-element upon cell assembly in the configuration shown in Figure 26. This configuration is presently in-test and has shown encouraging results.

The results for an innovative configuration* are shown in Figure 27. Although the cell voltage is relatively low - mostly because of high oxygen diffusion losses - performance has been quite steady throughout the test. Likewise, the open-circuit voltage stability has been excellent as shown in Figure 28. Testing is continuing.

The results for the configuration in Figure 26 are shown in Figure 29. The initial high performance eroded sharply after 10 days. The cell was disassembled, and additional acid was charged to the A-element. It is seen that this served to stabilize performance through the present time.

Figure 30 shows the open-circuit voltage history of this cell (No. 490). This plot indicates the need for acid-addition earlier and more pointedly than the cell-voltage plot. Also, the open-circuit voltage plot shows signs of another acid-deficiency in the matrix after 40 days. (This has not yet affected the cell voltage on load - Figure 29.)

Although testing of Cell No. 490 is continuing, it appears that improvement in the acid communication between the A-element and the matrix would be required in order for this configuration to be suitable. It must be determined whether the deficiency is in the A-element, the non-wetproofed periphery of the anode substrate, or in the matrix. The cell package will be examined upon disassembly. At the present time, impregnation experiments (e.g., using matrix material) are being conducted in order to increase dry-out resistance at the anode substrate periphery.

* The details are not shown because of possible patent action.

The fabrication of matrix elements in the 7 inch x 10.7 inch size - in preparation for 10-cell sub-stack testing - has indicated that contact screen-printing results are not easily reproducible with stack-size pieces. Alternate fabrication techniques are being explored, including off-contact screen-printing and screen-less blade-coating. These are being evaluated in terms of their suitability for automated processing. With regard to the preparation of matrix elements for near-term stacks, screen-less blade-coating appears to be yielding good reproducibility under manual processing. (Large-size single-cell testing of these components is underway, and initial results under open-circuit and load conditions are attractive.)

ELECTRODES

Goals for the electrode development program are as follows:

- (i) 0.68 volts IR-free hydrogen-air performance @ 150 amp/ft² (161 ma/cm²) and 350-400°F
- (ii) Precious metal loading of 1.0 mg Pt/cm², total cell
- (iii) Non-P.M. projected high-volume cost of \$5/ft², total cell

Nominal 10% Pt on carbon catalysts are being utilized in the electrode program. These are supplied by Engelhard under a complementary program that is wholly sponsored by Engelhard - Task V. Catalysts are co-deposited with Teflon onto porous, wetproofed carbon-fiber substrates according to Engelhard-developed techniques. Nominal precious metal loading on each electrode is 0.46 mg Pt/cm². Both woven (Pfizer) and non-woven (Stackpole) substrates are being used.

Considerable study of catalyst preparation variables was undertaken before arriving at a type of preparation that yielded what became base-line 10% Pt on carbon catalysts. Table X presents the results of a group of single-cell tests that was used to aid in screening of electrocatalyst types.

Electrode performance benefited substantially from the lower IR-losses obtained with the laminate-type matrix. Figure 31 shows the cell voltage versus current-density curve for a cell with base-line 10% Pt on carbon, 0.46 mg Pt/cm² (nominal) anode and cathode. Initial cell voltage on H₂-air is seen to be about 0.65 volts at 150 amp/ft² (161 ma/cm²) and 325°F (163°C). IR-loss was 44 mv at this current-density.

Figure 32 shows the IR-free cell voltage versus current-density for the same cell. This plot indicates a very high catalyst activity for this cathode (about 17 ma/mg at 0.9 volts on H₂-O₂, compared to the typical 10 ma/mg at 325°F). On the other hand, oxygen diffusion losses are seen to be unusually high for this cathode. This apparently resulted from inadequate cathode wet-proofing. Accordingly, the high initial H₂-air cell performance began to retreat somewhat as shown in Figure 33. The increasing deviation between H₂-O₂ and H₂-air performance is indicative of hydrophobicity loss.

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Figure 34 shows the cell voltage as a function of current-density for a cell using another base-line catalyst. This cell is also seen to have high initial performance (about 10 mv higher than recent average). However, as illustrated in Figure 35, the performance characteristics are quite different. This cathode catalyst has a more typical activity of 11 ma/mg at 0.9 volts on H₂-O₂, but oxygen diffusion losses are quite low. The net result is essentially the same H₂-air performance at 150 amp/ft².

The different degrees of wetproofing effectiveness of these two cathodes are believed to result from wetproofing variability in the non-woven substrate (Stackpole). This substrate generally offers better catalyst utilization than the non-woven type because there is minimal penetration of catalyst into the interior. However, the wetproofing effectiveness is less dependable, and improvements are being sought.

The use of FEP-120 is being explored as a wetproofing agent instead of TFE-30. Acid-uptake tests had shown FEP-120 to wetproof non-woven substrate material more effectively than TFE-30 at moderate loadings (about 5 mg fluoropolymer/cm²). Initial single-cell test results have been very encouraging.

Figure 36 shows an initial performance plot for a cell with FEP-wetproofed non-woven cathode and anode substrates. It is seen that the diffusion polarization for this cell is extremely low, indicating good wetproofing. Other cells of this type recently started on-test have shown similar characteristics. However, the longest-running cell in the series has shown an O₂-gain increase - indicative of diminished hydrophobicity - of 15 mv after about 30 days on load. Considerably more test data will have to be accumulated before firm conclusions can be drawn.

CATALYST STUDIES

Cyclic-voltammetric studies have been performed to screen new electro-catalysts and to monitor the effects of electrocatalyst aging experiments. Figure 37 shows a diagram of the Teflon test cell in which the cyclic-voltammetric evaluations are performed. The electrode-holder for this apparatus is shown in Figure 38. The electrical schematic is shown in Figure 39. Figure 40 is a cyclic-voltammetric plot illustrating the various characteristic regions, including the two hydrogen adsorption peaks which are analyzed quantitatively in order to convert the associated charge to the corresponding platinum surface-area. Figure 41 indicates the type of variations in the shape of the voltammogram that result from changing the platinum loading of the test sample while keeping the sweep-rate constant.

The aging characteristics of base-line 10% Pt on carbon catalysts are being studied by exposure to phosphoric acid in potentiostatic stations of the type shown in Figure 8. Samples are withdrawn periodically, and the platinum surface-area loss is monitored by cyclic-voltammetric examination. Figure 42 shows the platinum surface-area of samples of a base-line catalyst as a function of phosphoric acid exposure-time at a potential of 0.7 volts (RHE).

The results of Figure 42 indicate that platinum sintering in phosphoric acid proceeds at a high rate initially but then slows down drastically within the first 100 hours. Also, a substantial temperature effect on the degree of sintering - at least within the first 500 hours - is evident over the range 325-400°F (163-204°C). Longer-term testing is in progress.

Figure 43 shows the effect of temperature on the platinum sintering of another base-line catalyst after 50 hours at 0.7 volts. This plot illustrates the wide range of initial sintering rates in this temperature band.

The cathode of Cell No. 444, which operated for 1800 hours at 163°C (above), was analyzed for platinum surface-area by cyclic-voltammetry. It was found to have a respectable platinum specific-surface-area of 85 m²/g, as compared to 144 m²/g for the fresh catalyst (the same as that of Figure 43). This represents an area-loss of about 41% over this period.

The same cathode was analyzed by transmission electron microscopy to observe the degree of platinum crystallite-size growth. Sections of fresh and aged catalyst are shown in Figures 44 and 45, respectively. Sintering of the platinum crystallites is clearly evident from these micrographs, but the degree appears to be less severe than that obtained earlier with 30% Pt on carbon catalysts. The results are presently being analyzed.

The examination of a representative cross-section of base-line catalysts is helping to define sintering trends. Testing will be extended to follow these trends for longer time-periods. The rigorous determination of sintering trends in base-line catalysts will provide an important frame of reference for monitoring progress in the development of advanced catalysts that will be tested later.

TASK II - FUEL PROCESSING SUB-SYSTEM DESIGN AND DEVELOPMENT

A major task of this program is the development of a complete 5-KW fuel processing sub-system. The analysis and design for an overall fuel cell system are being addressed in Task III.

The fuel processor development effort includes (i) thermodynamic analysis of the steam-reforming process for three candidate fuels, (ii) preliminary system and economic analysis for the purpose of fuel selection, (iii) component development activity (steam-reformer, burner, etc.), (iv) sub-system design, and (v) construction and testing.

STEAM-REFORMING THERMODYNAMIC ANALYSIS

The thermodynamics of the steam-reforming process were analyzed for three candidate fuels - natural gas, naphtha, and methanol. The results generated included the steam-reformer effluent composition and the heat requirements for the steam-reforming process. Results were obtained over a wide range of operating temperatures and H₂O/C ratios.

An APL computer program was developed and utilized for this purpose. Random checks indicate virtual agreement with published thermodynamic equilibrium data. An example of the computer output data is shown in Table XI.

PRELIMINARY SYSTEM ANALYSIS

Preliminary system and economic analysis was conducted in order to provide background for the selection of a fuel for this program. This effort was directed toward estimation of factors such as fuel cost (ϕ /KW-hr) and system capital cost; the impact of price availability and supply-logistics for the various fuels was also taken into account. Table XII shows the procedural outline that was used to generate the desired economic data.

A computer program was written in APL language to facilitate system heat and material balance calculations. This was of great utility in establishing thermal-management concepts for the respective systems. A sample data sheet from this program is shown in Table XIII.

Simplified on-site integrated-energy system schematics using natural gas (methane) and methanol fuels are shown in Figures 46 and 47, respectively. Table XIV shows the economic estimates for these fuels that resulted from the analysis and the assumptions listed therein. Although the methane system has an economic advantage, both systems appear to be viable for the on-site application (especially in light of the fact that conservative fuel price and capital cost estimates were made). Other factors were also considered (Table XV) in recommending methanol for use in this program. The selection of methanol was approved by DOE, and the Task II and Task III efforts are now proceeding on that basis.

COMPONENT DEVELOPMENTReformer

A sub-scale steam-reformer test facility will be utilized to study important parameters affecting the reformer design. These include:

- (i) temperature profile
- (ii) H_2O/C ratio
- (iii) space velocity
- (iv) catalyst type
- (v) catalyst pre-conditioning
- (vi) decay modes
- (vii) effect of shutdown/start-up cycles

A suitable facility was designed and constructed at Menlo Park, and this is now near completion. Testing will begin during April, and initial results will be reported next month.

Burner

A full-size 5-KW reformer-burner test facility is also being prepared at Menlo Park. This will be used in conjunction with preliminary reformer designs to confirm burner design concepts and develop hardware. Such activity will be highly complementary to the sub-scale test effort.

TASK III - INTEGRATED SYSTEM DESIGN AND DEVELOPMENT

In light of the recent selection of methanol as the fuel for this program, analysis of the methanol-air fuel cell system is proceeding. This activity will lead toward optimization of a selected on-site integrated-energy-system.

TASK IV - LARGE STACK DESIGN AND DEVELOPMENT

The stack component scale-up activity of Task IV will begin later in 1979.

TASK V - ELECTROCATALYST DEVELOPMENT

Catalyst preparations are being supplied to the program as the result of a parallel effort that is wholly sponsored by Engelhard. These catalysts are evaluated under Task I by cyclic-voltammetry and, where warranted, by single-cell testing.

A base-line catalyst type having a nominal composition of 10% Pt on carbon has been selected as a standard for utilization in the Task I program. Comprehensive aging studies are being performed on base-line catalysts. Advanced catalysts presently under development will be similarly evaluated - as appropriate - and results will be compared with those of the base-line catalysts.

TABLE ICONFIGURATION OF THREE-CELL PRELIMINARY STACK #1

No. of Cells:	3
Cell Area:	75 in ² (7 in x 10.7 in)
Cell Active Area:	58 in ²
Bi-Polar Plates	Carbon-Sealed Graphite (Tylan)
Matrix:	Screen-applied to each electrode; silicon carbide filler
Acid Management:	Acid added to cells on routine basis
Catalyst:	10% Pt on carbon
Electrodes:	0.5 mg Pt/cm ² nominal loading, each electrode; non-woven precursor Stackpole substrate (anode and cathode)

TABLE II
PROPERTIES OF ALL-CARBON BIPOLAR PLATE MATERIALS

	<u>Electrical Resistivity</u>	<u>Hydrogen Impermeability</u>	<u>Flexural Strength</u>
Carbon-sealed graphite	negligible ⁽¹⁾	negligible ⁽²⁾	acceptable ⁽³⁾
Vitreous carbon	negligible ⁽¹⁾	negligible ⁽²⁾	acceptable ⁽³⁾
Vitreous carbon impregnated graphite	negligible ⁽¹⁾	unacceptable ⁽⁴⁾	acceptable ⁽³⁾

(1) less than 1 mv @ 150 ma/cm²

(2) below detection level ($\ll 0.5$ ma/cm² equivalent, 204°C)

(3) greater than 4000 psi

(4) $\gg 0.5$ ma/cm² equivalent; includes four samples with varying levels of impregnation

TABLE III
PROPERTIES OF RESIN-BONDED AND RESIN-SEALED BIPOLAR PLATE MATERIALS

	<u>Electrical Resistivity</u>	<u>Hydrogen Impermeability</u>	<u>Flexural Strength</u>	<u>Remarks</u>
Phenolic/graphite (25/75)*	acceptable ⁽⁴⁾			(1)
Polyphenylene sulfide/graphite (25/75)	negligible ⁽⁵⁾	negligible ⁽⁶⁾	acceptable ⁽⁸⁾	(1)
C0-40**/graphite	negligible ⁽⁵⁾	negligible ⁽⁶⁾	acceptable ⁽⁸⁾	(2)
Polyether sulfone/graphite	negligible ⁽⁵⁾	negligible ⁽⁷⁾		(3)
Polyether sulfone	negligible ⁽⁵⁾	negligible ⁽⁷⁾		(3)
Polyparabanic acid/graphite	negligible ⁽⁵⁾	negligible ⁽⁷⁾		(3)

(1) molded composition

(2) resin impregnation

(3) film-bonded

(4) less than 3 mv @ 150 ma/cm²

(5) less than 1 mv @ 150 ma/cm²

(6) below detection level ($\ll 0.5$ ma/cm² equivalent, 204°C)

(7) no leakage on basis of room-temperature bubble test, 10 psi

(8) greater than 4000 psi

* weight ratio

** Kearney Industries proprietary resin

TABLE IV

ELEMENT GOALS FOR NON-GROOVED ABA BIPOLAR PLATE CONFIGURATION

	<u>A-Element</u>	<u>B-Element</u>
IR-Loss @ 150 ma/cm ²	<1 mv	<1 mv
Air Pressure-Drop	<2 in. H ₂ O*	**
Hydrogen Impermeability	**	<0.5 ma/cm ² equivalent

* @ 2x stoichiometric flow, 150 ma/cm²; 6-inch length

** Not applicable

TABLE V
PHOSPHORIC ACID TOLERANCE OF ALL-CARBON BIPOLAR PLATE MATERIALS

	<u>Immersion Tests*</u>	<u>Potentiostatic Tests*</u> <u>(204°C, 1.0 volts)</u>	<u>Remarks</u>
Carbon-sealed graphite		No measurable weight-loss after 1000 hours	
Vitreous carbon	0.07% weight-loss after 1000 hours at 175°C**		
Vitreous carbon impregnated graphite			(1)

(1) Testing not conducted because of failure to pass hydrogen impermeability tests.

* Fresh samples washed and oven-dried before weighing; aged samples hot-soaked, washed and oven-dried before weighing.

** Data obtained in prior contract.

TABLE VI

PHOSPHORIC ACID TOLERANCE OF RESIN-BONDED AND RESIN-SEALED BIPOLAR PLATE MATERIALS

	<u>Immersion Tests*</u>	<u>Potentiostatic Tests*</u> (204°C, 1.0 volts)	<u>Remarks</u>
Phenolic/graphite (25/75)*		4.6% weight-loss after 1000 hours	(1)
Polyphenylene sulfide/graphite (25/75)	0.19% weight-loss after 1000 hours at 175°C**	Disintegrated	(1)
CC-40***/graphite		0.7% weight-loss after 1000 hours; 10.2% weight- loss after 2000 hours	(2)
		0.085 mA/g corrosion current after 500 hours	(3); see Fig.9,10
Polyether sulfone	0.56% weight-loss after 1000 hours at 204°C		(4)
Polyether sulfone	0.18% weight-loss after 1000 hours	0.085 mA/g corrosion current after 840 hours	(5); see Fig.12,13
Polyparabanic acid	9.7% weight-loss after 1000 hours at 204°C		(4)

(1) molded composition

(2) resin impregnation, exposed edges

(3) resin impregnation, sealed edges

(4) resin only

(5) film-bonded

* fresh samples washed and oven-dried before weighing; aged samples hot-soaked, washed, and oven-dried before weighing

** data obtained in prior contract

*** Kearney Industries proprietary resin

TABLE VII
PROPERTIES OF CVD-UPGRADED* RETICULATED VITREOUS CARBON A-ELEMENTS

	<u>Original Porosity</u>	<u>IR-Loss @ 150 ma/cm²</u>	<u>Air Pressure-Drop**</u>	<u>Crush Strength</u>
RVC @ 45 pores/inch	98%	2 mv	acceptable ⁽²⁾	150 psi
RVC @ 60 pores/inch	98%	2 mv	acceptable ⁽¹⁾	

* to 6-8 times the original weight; 3 samples of each type

** @ flow equivalent to 2x stoichiometric, 150 ma/cm²; corrected to 6-inch length, 350°F

(1) less than 2 in. H₂O

(2) less than 1 in. H₂O

TABLE VIII

PHOSPHORIC ACID TOLERANCE OF ABA BIPOLAR PLATE MATERIALS

	<u>Potentiostatic Tests*</u> <u>(204°C, 1.0 volts)</u>	<u>Remarks</u>
Reticulated vitreous carbon	Disintegrated	
CVD-upgraded RVC	9.0% weight-loss after 1000 hours	(1)
	2.8 mA/g corrosion current after 387 hours	(2); see Fig.15,16
	0.07 mA/g corrosion current after 113 hours	(3); see Fig.17,18
Woven fabric precursor B-element	No measurable weight-loss	

- (1) exposed at cut edges
- (2) exposed at cut edges - second sample
- (3) coated by CVD at edges

* fresh samples washed and oven-dried before weighing; aged samples hot-soaked, washed and oven-dried before weighing

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TABLE IX
LAMINATED MATRIX-FILLER MATERIALS

CORRELATION BETWEEN CRYSTALLITE SIZE AND OPEN-CIRCUIT VOLTAGE

<u>Crystallite Size (microns)</u>			<u>Open-Circuit Voltage* (mv)</u>
<u>Average</u>	<u>Maximum</u>	<u>Minimum</u>	
17	28	8	860 $\pm$ 45
10	16	5	862 $\pm$ 15
6	12	2	880 $\pm$ 27
4	9	1.5	896 $\pm$ 25

* Measured at 325°F after 24 hours with H₂-Air reactants.

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TABLE X

Results on Electrocatalyst Performance
Evaluation by Single Cells

Electro- catalyst	Electrode*	Run	Time (Hours)	Open Circuit (mV)	O ₂ ** Gain (mV)	IR** Drop (mV)	Voltage** (IR-Free) (mV)	Perfor- mance
10914-12	510	456	24	914	73	58	695	Very Good
10914-15	369	385	24	818	93	94	671	Very Good
10914-21	455	436	24	862	71	70	681	Very Good
10914-35	463	451	24	751	137	92	600	Poor
10914-37	437	423	40	772	86	96	658	Fair
10914-39	516	457	24	912	92	42	687	Very Good
10914-40	451	427	24	872	127	120	440	Poor
10914-41	453	431	24	692	112	120	594	Poor
10914-43	465	450	24	760	112	116	624	Poor
10914-45	469	462	24	892	100	44	641	Fair
10914-49	483	453	24	838	81	34	662	Good
10957-3-1	489	455	24	850	90	36	665	Good
10974-04	521	464	24	881	98	46	683	Very Good
10974-05	534	469	24	867	108	52	641	Fair
10929-7	416	424	24	910	92	88	663	Good

* Anode and cathode; the nominal Pt loading is 0.46 mg/cm² (10% Pt on C)

** At 150 amp/ft²

Temperature: 325°F

TABLE XI
NAPHTHA REFORMING - EQUILIBRIUM CALCULATIONS

TEMPERATURE=1400.00DEGREES F. PRESSURE= 1 ATM.
H2O/CARBON RATIO= 2

	<u>FINAL MOLES FROM REACTOR</u>	<u>MOLE PER CENTS</u>	<u>MOLE PER CENT WATER FREE</u>
CNHH	.00	.00	.00
H2O	5.92	18.60	.00
CH4	.09	.27	.33
CO2	2.17	6.80	8.36
CO	5.75	18.06	22.19
H2	17.91	56.26	69.12
<u>WEIGHT BALANCES, PER CENT LOSS</u>			
CARBON	.00		
HYDROGEN	.00		
OXYGEN	.00		

CO₂/CO=.38

TEMPERATURE=1400.00DEGREES F. PRESSURE= 1 ATM.
H2O/CARBON RATIO= 3

	<u>FINAL MOLES FROM REACTOR</u>	<u>MOLE PER CENTS</u>	<u>MOLE PER CENT WATER FREE</u>
CNHH	.00	.00	.00
H2O	12.62	31.60	.00
CH4	.03	.06	.09
CO2	3.40	8.52	12.45
CO	4.57	11.44	16.73
H2	19.33	48.38	70.72
<u>WEIGHT BALANCES, PER CENT LOSS</u>			
CARBON	.00		
HYDROGEN	.00		
OXYGEN	.00		

CO₂/CO=.74

TEMPERATURE=1400.00DEGREES F. PRESSURE= 1 ATM.
H2O/CARBON RATIO= 4

	<u>FINAL MOLES FROM REACTOR</u>	<u>MOLE PER CENTS</u>	<u>MOLE PER CENT WATER FREE</u>
CNHH	.00	.00	.00
H2O	19.79	41.26	.00
CH4	.01	.02	.04
CO2	4.22	8.79	14.96
CO	3.77	7.86	13.39
H2	20.18	42.07	71.62
<u>WEIGHT BALANCES, PER CENT LOSS</u>			
CARBON	.00		
HYDROGEN	.00		
OXYGEN	.00		

CO₂/CO=1.12

TEMPERATURE=1400.00DEGREES F. PRESSURE= 1 ATM.
H2O/CARBON RATIO= 5

	<u>FINAL MOLES FROM REACTOR</u>	<u>MOLE PER CENTS</u>	<u>MOLE PER CENT WATER FREE</u>
CNHH	.00	.00	.00
H2O	27.22	48.61	.00
CH4	.01	.01	.02
CO2	4.79	8.55	16.64
CO	3.21	5.73	11.15
H2	20.77	37.10	72.19
<u>WEIGHT BALANCES, PER CENT LOSS</u>			
CARBON	.00		
HYDROGEN	.00		
OXYGEN	.00		

CO₂/CO=1.49

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TABLE XII

FUEL CELL FUEL COMPARISONS

PROCEDURE

1. Thermodynamic model to determine concentrations and reaction heats for reformer and shift converters.
2. Design schematics for heat and material balances.
3. Size equipment.
4. Cost equipment and fuel requirements.
5. Economic calculations.

TW
3/1/79

[:
.1290 .0178 .2044 .6438
ENTER REFORMER OPERATING TEMPERATURE, °F.

[:
525
ENTER REFORMER HEAT LOAD IN BTU/HR.

[:
4322.515
ENTER PER CENT EXCESS AIR TO FUEL CELL

[:
100
ENTER PER CENT H2 BLEED

[:
16
FUEL CELL CONCENTRATIONS IN LBS/HR.
AIR IN:

NITROGEN 49.085644 OXYGEN 14.912
REFORMER GAS IN:
WATER 1.657774353 CO 0.3579753391 CO2 6.459644883 H2 0.932

AIR OUT:
WATER 7.809880245 NITROGEN 49.085644 OXYGEN 8.64896
BLEED OUT:
WATER 0.903814108 CO 0.3579753391 CO2 6.459644883 H2 0.14912

ENTER AIR FEED TO BURNER IN LBS/HR AT 325 F

[:
15
ENTER BURNER EFFICIENCY (FRACTION)

[:
.95
ENTER FRACTION OF AIR THROUGH BURNER AIR PREHEATER

[:
1
ENTER REFORMER SPACE VELOCITY, SCC H2/HR/CC CATALYST

[:
500
H2 BLEED FROM FUEL CELL= 16 PER CENT
EXCESS AIR TO FUEL CELL= 100 PER CENT

BURNER SPECIFICATIONS:

HEAT OF PRODUCTS=10872.69686 BTU/HR.
HEAT OF PRODUCTS AT FLAME TEMPERATURE=10872.1271 BTU/HR.
BURNER FLAME TEMPERATURE=1655.30779 °F.
EXCESS OXYGEN= 248.7169195 PER CENT

REFORMER SPECIFICATIONS:

OPERATING TEMPERATURE= 525 °F.
TRUE ENTHALPY FROM REFORMER=6550.181859 BTU/HR.
HEAT OF COMBUSTION GAS FROM REFORMER=6546.525965 BTU/HR.
COMBUSTION GAS TEMPERATURE FROM REFORMER=990.8663445 °F
 $\Delta T_{LM}=749.640759$ °F

SURFACE AREA FOR HEAT TRANSFER=2.883057617 FT. SQ.
LENGTH OF 1 INCH TUBING FOR REACTION=70.05010539 FT.
LENGTH OF 1 INCH TUBING FOR HEAT TRANSFER=11.76758211 FT.
CATALYST VOLUME=9475.0672 CC.
SPACE VELOCITY=500 SCC H2/HR/CC. CATALYST

PREHEATER SPECIFICATIONS:

TRUE ENTHALPY FROM PREHEATER=1790.15535 BTU/HR.
HEAT OF COMBUSTION GAS FROM PREHEATER=1707.580967 BTU/HR.
COMBUSTION GAS TEMPERATURE FROM PREHEATER=179.0737177 °F
 $\Delta T_{LM}= 300.6314648$ °F.

TABLE XIII

METHANOL FUEL CELL SYSTEM

COMPUTER INPUT AND OUTPUT -

THERMAL MANAGEMENT CALCULATIONS

TABLE XIV
COST FACTORS FOR ON-SITE SYSTEM

A. BASIS	<u>METHANE</u>	<u>METHANOL</u>
1. AUXILIARY POWER (% OF NET)	12	8
2. INVERTER EFFICIENCY (%)	95	95
3. H ₂ UTILIZATION IN FUEL CELL (%)	80	85
4. VOLTAGE EFFICIENCY (%HHV)	43.9	43.9
5. FUEL PRICE	40¢/CCF	52¢/GAL
B. FUEL COST (¢/KWH) *	4.4	7.1
C. SYSTEM CAPITAL COST (\$/KW) *	646	550
D. LIFE-CYCLE COST (¢/KWH) *	6.8	8.9

* COSTS ARE PER UNIT ELECTRICAL OUTPUT

TABLE XV

METHANOL FUEL IN ON-SITE APPLICATION -
OTHER FACTORS AFFECTING CHOICE

- DEVELOP NEW SYSTEM TECHNOLOGY
- FUEL ALTERNATIVE
- AREAS NOT HAVING ACCESS TO NATURAL GAS
- ATTRACTIVE CANDIDATE AS FIRST-GENERATION COAL-DERIVED FUEL
- LONG-TERM PRICE SCENARIO (VIS-A-VIS COAL) POSSIBLY ATTRACTIVE

FIGURE 1

PERFORMANCE STABILITY

IN THREE-CELL STACK NO. G08-03-01

TEMP. = 325°F (163°C)

CURRENT DENSITY = 150 A/ft²

Pt LOADING: 0.46 mg/cm², EACH ELECTRODE

VOLTAGE

2.0

1.8

1.6

1.4

1.2

1.0

0

6

12

18

24

30

36

42

48

54

60

TIME (DAYS)

TEMP.
TO
350°F (177°C)

TEMP.
TO
375°F (191°C)

TEMP.
TO
400°F (204°C)

OK
4/20/79

FIGURE 4

PERFORMANCE STABILITY

IN THREE-CELL STACK NO. G08-03-02

TEMP. = 325°F (163°C)

CURRENT DENSITY = 150 A/ft²

Pt LOADING: 0.46 mg/cm², EACH ELECTRODE

+ TOP CELL

x MIDDLE CELL

o BOTTOM CELL

SHUT-DOWN
PERIODS

TEMP.
TO
375°F (191°C)
↓

VOLTAGE

0.7

0.6

0.5

FIGURE 6

V_{A-D} = VOLTAGE DROP ACROSS SAMPLE

TEST CHECK :

$$V_{B-C} = (V_{A-C}) + (V_{B-D}) - (V_{A-D})$$

FIGURE 7 - HYDROGEN PERMEABILITY TEST APPARATUS

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RESEARCH & DEVELOPMENT DEPT.

FIGURE 8

POTENTIOSTATIC AGING TEST CELL

FIGURE 11

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FIGURE 14

A - B - A

CONSTRUCTION

PT, ,

CORROSION CURRENT (mA/g)

5

4

3

2

1

0

1

10

TIME (HOURS)

100

1000

FIGURE 15

CORROSION-CURRENT HISTORY OF

CVD-UPGRADED RVC A-ELEMENT

Temperature = 204°C

Potential = 1.0 Volts (RHE)

Conc. Phosphoric Acid
(RVC exposed at edges)

1

2

3

4

5

6

7

8

9

10

- 45 -

AC
4/10/79

2

3

4

5

6

7

8

9

1

2

3

4

5

6

7

8

9

10

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46 5810

CORROSION CURRENT (mA/g)

0.10

0.08

0.06

0.04

0.02

0

TIME (HOURS)

10

100

1000

FIGURE 17

CORROSION-CURRENT OF
CVD-UPGRADED RVC A-ELEMENT

Temperature = 204°C

Potential = 1.0 Volts (RHE)

Conc. Phosphoric Acid

(RVC not exposed at edges)

- 47 -

Ac
4/10/1

POTENTIAL (mV vs. RHE)

1000

800

600

400

200

0

0.001

0.01

CORROSION CURRENT (mA/g)

0.10

1.0

FIGURE 18

TAFEL PLOT FOR CORROSION OF

CVD-UPGRADED RVC A-ELEMENT

Temperature = 204°C

Time = 24 Hours

Conc. Phosphoric Acid

(RVC not exposed at edges)

10
9
8
7
6
5
4
3
2
1
0

3

4

5

6

7

8

9

1

2

3

4

5

6

7

8

9

FIGURE 19

"B" Element Hydrogen Leakage Test

SAMPLE	AVG. TEMP. (°C)
+ (A-1)	215
X (A-3)	220
⊙ (B-2)	208
Δ (B-4)	190

Chamber Pressure - PSIG

Time - Hours

R.A. SMITH 12/6/78

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FIGURE 20

BUBBLE-PRESSURE TESTING APPARATUS

64-22
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FIGURE 21

ACID-MANAGEMENT CONFIGURATIONS

RK
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FIGURE 22

H.F.
 5/18/70

FIGURE 23

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FIGURE 24

FIGURE 25

H.F.
 4/8/73

FIGURE 26

METHOD FOR UTILIZING A-ELEMENT
FOR INTERNAL ACID STORAGE

FIGURE 27

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FIGURE 28

FIGURE 29

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FIGURE 30

FIGURE 31

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FIGURE 32

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SINGLE-CELL PERFORMANCE OF 10% Pt ON CARBON ELECTRODES

FIGURE 33

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SINGLE CELL PERFORMANCE STABILITY

CELL
VOLTAGE

CELL NO.: #457

CATHODE: 0.46 mg Pt/cm²(10914-39)

ANODE: 0.46 mg Pt/cm²(10914-39)

MATRIX: Laminated

CURRENT DENSITY: 150 Amp/ft²

TEMP: 325°F

H₂-O₂

H₂-Air

0 4 8 12 16 20 24 28 32 36 40
TIME (DAYS)

FIGURE 34

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FIGURE 35

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SINGLE-CELL PERFORMANCE OF 10% Pt ON CARBON ELECTRODES

FIGURE 36

SINGLE-CELL PERFORMANCE OF 10% Pt

ON CARBON ELECTRODES

WITH FEP-WETPROOFED SUBSTRATES

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IR-FREE
CELL VOLTAGE

0.9

0.8

0.7

0.6

CELL NO.: 512
CATHODE: Wet-proofed with FEP(593)
ANODE: Wet-proofed with FEP(592)
MATRIX: Laminated

TEMP. 325°F
TIME: 24 Hours

H₂-O₂

H₂-Air

DIFFUSION
POLARIZATION

1

2

10

20

50

100

200

CURRENT DENSITY (Amp/Ft²)

H.F.
5/8/79

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FIGURE 37

CYCLIC-VOLTAMMETRIC TEST CELL (TEFLON)

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FIGURE 38

ELECTRODE HOLDER SHOWING THE ASSEMBLY OF THE TEST ELECTRODE

AC

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FIGURE 39

ELECTRICAL ARRANGEMENT FOR CYCLIC VOLTAMMETRY

AC
4/10/79

FIGURE 40

CYCLIC VOLTAMMOGRAM OF Pt BLACK CATALYST

IN 25% H₃PO₄, T = 25°C

10 mV/S

FIGURE 41

Cyclic Voltammogram of Electrocatalysts with Various Pt Loading

.25% H_3PO_4

Sweep Rate = 20 mv/s

FIGURE 42

EFFECT OF TIME ON Pt SINTERING

10% Pt ON CARBON BASE-LINE ELECTROCATALYSTS

Electrocatalyst: 10914-39

Sintering Conditions: 700 mv(vs.RHE)

100% H_3PO_4

○ 400°F ± 5

□ 325°F ± 5

$S_0 = 140 \text{ m}^2/\text{g}$

FIGURE 43

EFFECT OF TEMP. ON Pt SINTERING

10% ON CARBON BASE-LINE ELECTROCATALYSTS

Electrocatalyst: 10914-21

Sintering Conditions: 700 mv(vs.RHE)

50 $\pm$ 2 hrs,

$S_0 = 144 \text{ m}^2/\text{g}$

100% H_3PO_4

FIGURE 44
TRANSMISSION ELECTRON MICROGRAPH OF
FRESH 10% Pt ON CARBON CATALYST
(X246,000)

-74-

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FIGURE 45
TRANSMISSION ELECTRON MICROGRAPH OF
AGED 10 % Pt ON CARBON CATALYST
(X 246,000)

-75-

ENGELHARD

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FIGURE 46

BASIC SCHEMATIC -
METHANE FUEL CELL SYSTEM
FOR ON-SITE APPLICATION

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FIGURE 47

BASIC SCHEMATIC -
METHANOL FUEL CELL SYSTEM
FOR ON-SITE APPLICATION