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FUEL CELL

RESEARCH & DEVELOPMENT

ION EXCHANGE FUEL CELL
REPORT NUMBER 1

CONTRACT NUMBER DA-36-039-AMC-00095(E)
TASK NO. OST 76-11-001-36
ARPA ORDER NUMBER 80

FIRST SEMI-ANNUAL TECHNICAL REPORT
OCTOBER 1, 1962 - DECEMBER 31, 1962
DATE OF CONTRACT: 1 OCTOBER 1962
AMOUNT: \$198,880.00

U. S. ARMY ELECTRONICS RESEARCH AND DEVELOPMENT LABORATORY
FORT MONMOUTH, NEW JERSEY

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ION EXCHANGE MEMBRANE FUEL CELL

REPORT NO. 1

CONTRACT NUMBER DA-36-039-AMC-00095(E)

TECHNICAL GUIDELINES EPP NO. 58238 OF JULY 17, 1962

TASK NO. OST 76-11-001-36

ARPA ORDER NO. 80

FIRST SEMI-ANNUAL TECHNICAL REPORT

OCTOBER 1, 1962 - DECEMBER 31, 1962

OBJECT: RESEARCH ON ION-EXCHANGE MEMBRANE FUEL CELLS

REPORT PREPARED BY:

Henri J. R. Maget

Henri J. R. Maget

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E. A. Oster

E. A. Oster

The work performed under this contract was made possible by the support of the Advanced Research Project Agency under Order Number 80, through the United States Army Electronics Research and Development Laboratory.

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1.0 PURPOSE

The work undertaken under this contract and presented in this Report is related to research investigations to contribute to the development of air-breathing fuel cells based on ion exchange membranes as "solid" electrolytes.

Technical guidelines EPP No. 58238 of the contract were defined as follows:

1. These Technical Guidelines cover the requirements for the investigation of hydrogen-oxygen ion exchange membrane fuel cells. These guidelines further cover limited investigations of fuels other than hydrogen which show practical and economical feasibility with respect to military applications.
2. The objective of these investigations is to improve the power density and lifetime characteristics of ion exchange membrane fuel cell power supplies for ground power applications. Cells utilizing fuels other than hydrogen shall also be investigated. Emphasis shall be placed on the improvement of ion exchange membrane fuel cell technology and a systematic understanding of the phenomena observed. This effort should lead to an increase in the state of the art which will allow the design of improved fuel cell power sources to meet ground power requirements with a known reliability.
3. The Contractor shall conduct the necessary investigations leading to the establishment of an analytical model expressing factors determining fuel cell performances. This model shall be used to further improve the overall operating performance of the ion exchange membrane fuel cell. The Contractor shall perform the following experimental and theoretical studies:
 - a. Determine the factors causing ion exchange membrane oxygen electrode performance to decrease when operating on convected ambient air as the source of oxygen. Work will be directed toward obtaining optimum air electrode performance.
 - b. Conduct fuel cell performance and life studies, using new improved cation exchange membrane materials. Emphasis shall be placed on structures which improve ionic conductance, physical properties, and stability.

1.0 PURPOSE (Cont'd)

- c. Complete a study of factors determining the membrane's water balance.
- d. Investigate factors influencing electrode-electrolyte coupling to produce low electrode polarization. This work shall be directed toward optimization of ion exchange membrane electrodes.
- e. Investigate fuels other than hydrogen in ion exchange membrane fuel cells. These fuels shall be restricted to those found by other investigators to work such as dissolved liquid fuels in acid electrolytes. Emphasis shall be given to determining the Faradaic efficiency, main reactions, and causes of anode polarizations. Studies of the methanol ion exchange membrane cell shall be included in these investigations.

The initial experimental program started in October 1962, and approved on November 1, 1962, is considered compatible with the Technical Guidelines.

The following TASK organization was adopted:

TASK I: Interface Structure and Interfacial Properties of IEM/Electrode Systems

1. Isotropic Characteristics of an Ion Exchange Membrane.
2. Ion Exchange Membrane/Electrode Morphology.
3. Interfacial Bonding Forces.

TASK II: Water Transport Across Ion Exchange Membranes

1. Water Permeability.
2. Electro-osmotic Water Transport.
3. Water Gradients in Membranes.

TASK III: Kinetic Study of Electrochemical Oxygen Reduction at Variable Oxygen Partial and Total Pressures

1. Electrode Reproducibility.
2. Variables Affecting Reaction Rates.
3. Transient Rate Measurements.
4. Steady-State Rate Measurements.

TASK IV: Heat and Mass Transfer Investigations of the Air-Breathing IEM Cell and Battery

1. Over-all Mass Transfer Processes.
2. Detailed Mechanism.
3. Temperature Distribution and Profile.
4. Property Measurements.
5. Analytical Model.
6. Life Testing.

TASK V: Design Parameters Affecting Air-Operated IEM Fuel Cells

1. Performance Data as Related to Water Transport.
2. Systems Evaluation for Product Water Conservation.
3. Air Electrode for Constant Water Removal.
4. Air Electrode Configuration and Geometry.
5. Cell-Stack Operation.

TASK VI: Ion Exchange Membranes

1. New Ion Exchange Membrane Materials.
2. Materials Properties.

Postponed TASK VII: IEM Methanol Fuel Cell

1. Methanol Transport through IEM.
2. Rate of Methanol Oxidation in Solution.
3. Reduction of Cathodic Electrode-Poisoning.
4. Reaction Rates of Intermediate Products.

This Report presents experimental work regarding TASKS I, II, III and IV. Work on Design Parameters (TASK V) will start in January 1963 and will be based on experimental evidence obtained during the period covering this report as well as on experience acquired during the previous contract period, Oct. 1961 - Oct. 1962.

2.0 ABSTRACT

During the contract period from October 1, 1962 to December 31, 1962, experimental work has been conducted on membrane and interface membrane/electrolyte properties. Reaction rates for the electrochemical oxygen reduction as well as heat and mass transfer studies have yielded interesting results regarding air-breathing fuel cell electrodes. The reported results suggest a continued effort of the proposed program throughout the remaining contract period.

Preparation techniques to obtain reliable IEM/electrode interfaces for microscopic examination have been investigated. Some micrographs are available and reported. Attempts to measure interfacial properties by capacitance determination have yielded results suggesting applicability of a Ratio Transformer Bridge. A modified Heaven's apparatus has been built for the determination of relative adhesion between the membrane and electrode.

Experimental results obtained for water transport across an ion exchange membrane for pressure gradients up to 35 cm Hg have yielded permeability constants which respond to an Arrhenius type equation and activation energies of the order of 7000 cal/mole. An apparatus for electro-osmotic water transport measurements has been completed.

Limiting current densities have been measured for IEM-electrode systems as a function of oxygen partial pressure at 1 atm. total pressure. Nearly linear dependence on oxygen partial pressure has been shown and electrode polarization at current densities up to 50 mA/cm² is not appreciably dependent on this partial pressure.

A modified air electrode current collector has been developed for IEM Fuel Cell Operation. In absence of wicking systems, continuous operation for above 150 hours at 50 Amps/ft² and 0.50 volt terminal output has been demonstrated, with voltage fluctuations less than 10 mv. Experimental results from unmonitored operation conditions as well as observed cell resistance variations suggest a control device for optimized cell operation with proper product water management.

3.0 PUBLICATIONS, LECTURES, REPORTS AND CONFERENCES

3.1 Work Planning Conference with Contracting Officer, Fort Monmouth, N. J.

A meeting was held at Fort Monmouth, New Jersey, on November 1, 1962, between Messrs. A. Daniel, H. Hunger, J. Murphy, J. Perry and J. E. Wynn of the USAELRDL and Messrs. T. K. Johnson, H. J. Maget and E. A. Oster of the General Electric Company, Direct Energy Conversion Operation, Lynn, Mass. A detailed work program was presented and discussed. The program was approved in toto, and agreement was reached on postponing temporarily work on TASK VII regarding IEM Methanol Fuel Cells.

4.0 FACTUAL DATA

4.1 TASK I - Interface Structure and Interfacial Properties of IEM/Electrode Systems

4.1.1 Introduction

Fabrication of an ion-exchange membrane (IEM) fuel cell results in the formation of a new interfacial zone, comprising the boundary region between the electrode and the IEM electrolyte. The importance of this interface is known from the dependence of the cell's operational characteristics on the method of combining the two phases, as well as by the great effort that has been expended in developing optimum manufacturing procedures.

Intuitively, one would expect this dependence to be somehow related to ionic conductance and gas and water transport across the interface (not to exclude other factors); and to the variation of these parameters with the nature, homogeneity, and time-variance of the latter. Any more fundamental treatment of the problem, however, will require investigation of the basic nature of this outwardly solid-solid interface. Only then will it be possible to relate the cell's performance to its operating conditions and fabrication methods in any but a superficial manner.

4.1.2 Scope

The overall objectives of this phase can readily be divided into several interrelated and interdependent goals as listed below:

1. To determine the general nature of the IEM/electrode interfacial region.
2. To investigate properties of this "interphase" and to compare and separate them from those due to interfacial regions of adjoining phases (e. g., IEM/wetproofing agent and catalyst metal/aqueous solution interfaces).
3. To specify proper conditions for insuring isotropy at the interfacial surface, based upon a more fundamental knowledge of its morphology and properties.

4.0 FACTUAL DATA (Cont'd)

The amount of work accomplished in the general realm of solid-solid interfaces has been quite meager when compared to the considerable published literature available on the other facets of surface chemistry. This observation is particularly pertinent to interfaces composed of a (hard) metal and a soft non-metal, such as are involved in IEM fuel cell components. This is due, no doubt, to their relative experimental inaccessibility when contrasted to partially fluid interfacial systems.

The first task, therefore, was to investigate and develop possible experimental techniques for attacking the problem-- with three representative methods being chosen for the initial evaluation. These are:

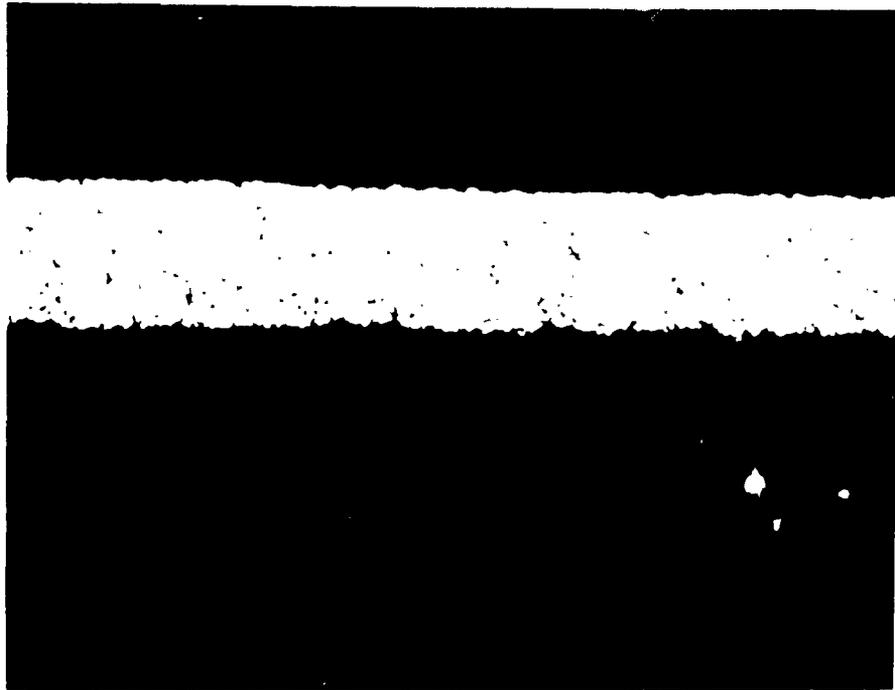
1. Direct visual observation by optical microscopy.
2. Determination of relative surface areas by electrical impedance.
3. Measurements of adhesion.

4.1.3 Work in Progress

4.1.3.1 Microscopy

Visual observation of the interface by (optical) microscopic examination of cell cross-sections appears to be a simple and direct way of determining their morphological characteristics. The method is capable of yielding information concerning particle segregation and "pore" properties, as well as the uniformity, roughness and relative interpenetration of the various phases. It can also be used to determine, at least superficially, the homogeneity and "tightness" of the electrode/IEM interfacial boundary.

Plates I and II are good illustrations of the method's potentialities. Although both show, at 1000:1 magnification, cross-sectional regions of the same type of cell ("grafted" polystyrene sulfonic acid - GPS), the results of different catalyst loadings (weight/unit area) and different fabrication conditions are much in evidence. One can contrast the uniform catalyst structure of Plate I with the fissures and holes (here filled with the plastic setting compound) of Plate II. Likewise, the poor adhesion of the latter interface may be deduced from the large amount of setting plastic that has penetrated up to the membrane boundary.



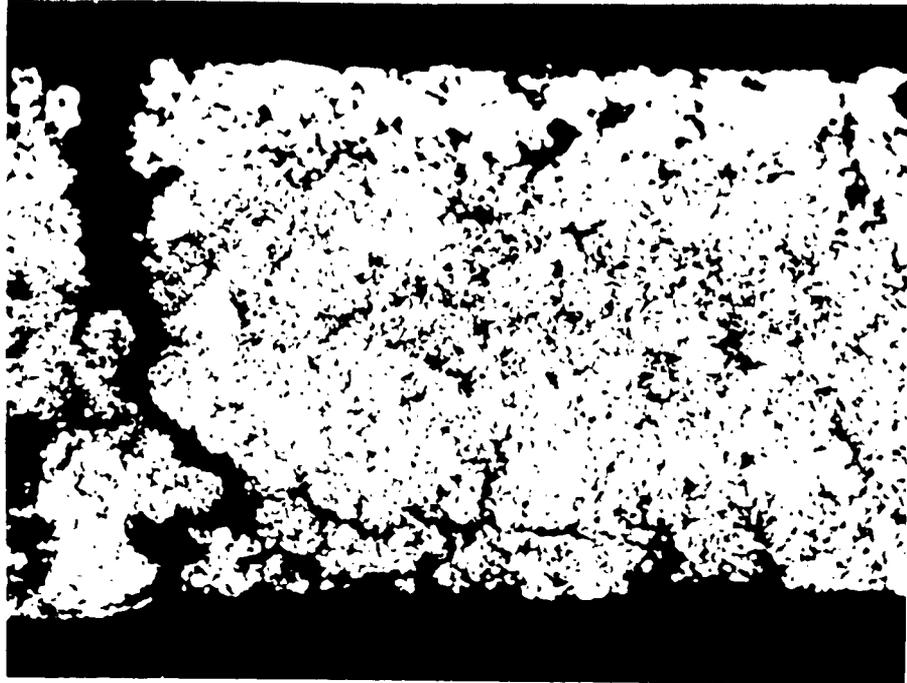
CROSS-SECTION OF ELECTRODE MEMBRANE (GPS)
INTERFACE, AT 1,000-1 MAGNIFICATION, WITH
MEMBRANE AT BOTTOM

(Obtained with collaboration of Mr. R. Russel
of G. E. Research Laboratory)

Fuel Cell Lab.
DECO - G. E. Co.
Lynn, Mass.

PLATE I

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S. Krumbain 12/31, 62



CROSS-SECTION OF ELECTRODE MEMBRANE (GPS)
INTERFACE, AT 100X MAGNIFICATION, WITH
MEMBRANE AT BOTTOM

Note boundary between epoxy setting
resin and the IEM membrane.

(Obtained with collaboration of Mr. R. Russel
of G.E. Research Laboratory)

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PLATE II

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4.0 FACTUAL DATA (Cont'd)

In contrast, a third cell cross-section is pictured, at 100:1 magnification, in Plate III. Evidence of insufficient catalyst mixing for this particular cell is seen by the "black" inclusions of the wetproofing agent in the electrode phase. In addition, the looseness of both interfaces on the right shows that the two phases are beginning to separate, while those on the left are still comparatively tightly bound. At the same time, the non-crosslinked sulfonated polystyrene membrane (PSS) used in this cell (in contrast to the GPS membrane) shows a macro-porous structure. The pores or cracks have assumed, for the most part, a flattened lamellar structure, parallel to the electrodes and close to them, while those in the center remain, for the most part, randomly oriented.

These micrographs were obtained with the cooperation of the General Electric Research Laboratory from samples prepared by two slightly different methods. They are presented only as illustrations of what can be expected from this type of experimental procedure. Any extensive program, however (viz. comparing Plate III with that of a cell fabricated at higher temperature and pressure and perhaps relating them to their respective performance characteristics), is dependent upon the development of reproducible sample-preparation techniques for these unusual materials and configurations that will faithfully conserve the interface and eliminate artifacts. This will also be a necessary first step for the further development of sample-preparation for electron microscopy.

The necessary equipment has been ordered and experimental facilities are being installed. This next phase of the program is expected to begin early in 1963.

4.1.3.2 Impedance Bridge Method

As discussed in a previous report (1), the measured capacitance of an IEM fuel cell component at high frequencies is assumed to be related to the resultant of a complex parallel circuit of different RC ratios, but free from practically all faradaic effects. Although the magnitude of these limiting capacitance values will be far below the "true" ones (2), they should, nevertheless, serve as a comparative measure of the relative double layer capacities of different electrode systems. Hence, if all other parameters of these systems (catalyst loading, superficial electrode area, fabrication conditions, etc.) are maintained constant, one should obtain a reasonable approximation of the relative surface area and roughness of the respective interfacial regions.



CROSS-SECTION OF ION EXCHANGE MEMBRANE (PSS)
FUEL CELL AT 100:1 MAGNIFICATION.

The large oblong white spots are cross-sections of the screen wires.

(Obtained with collaboration of Mr. R. Russel
of G. E. Research Laboratory)

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Lynn, Mass.

PLATE III

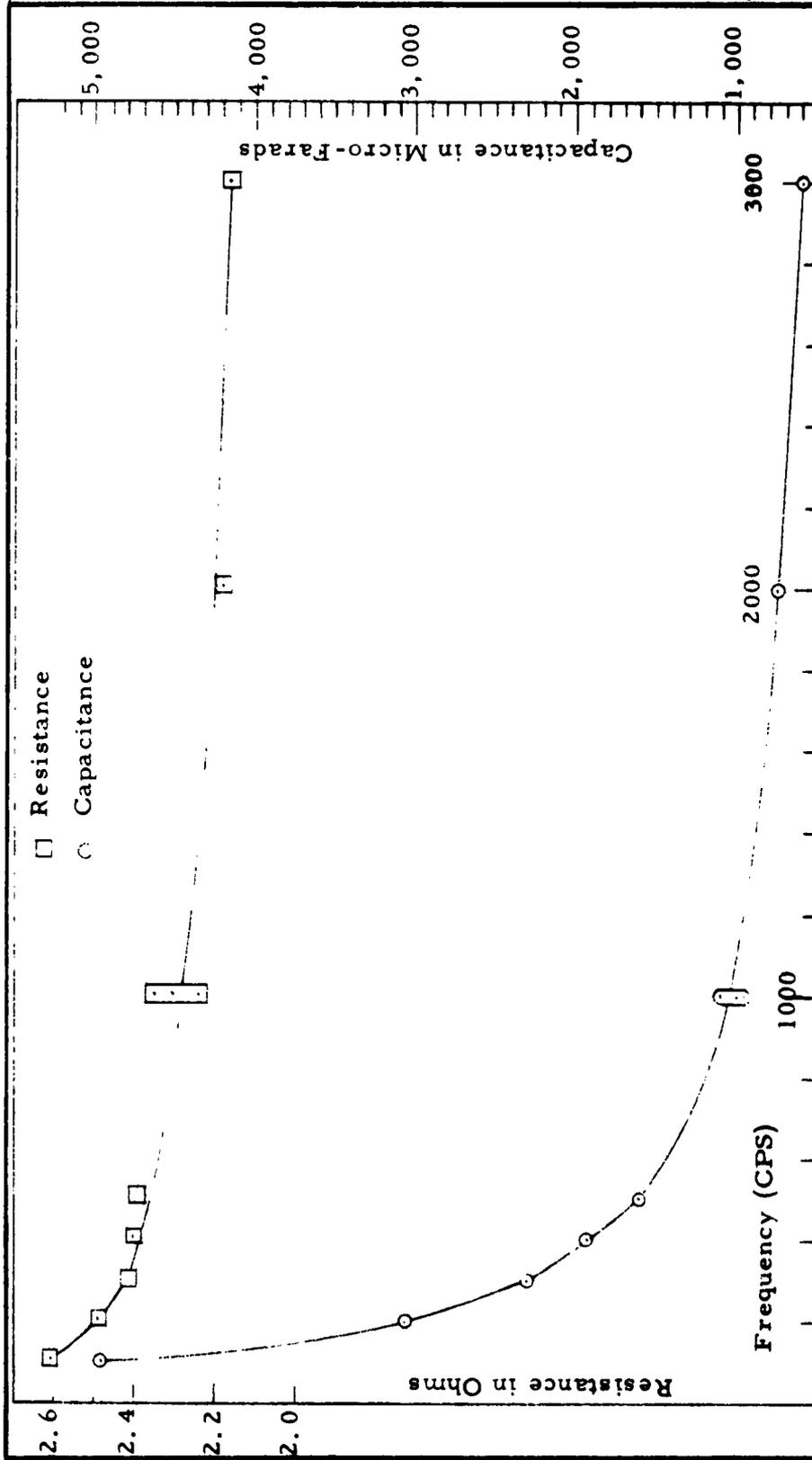
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In order to avoid the ambiguities of asymmetrical geometries (1, 3) when using the bridge technique, it was decided to confine the work to "back-to-back" parallel configurations, where the measured interfacial areas would be symmetrically disposed and of approximately identical character. Preliminary results were obtained for a rather thick (Type PSS) fuel cell configuration, in air, -- using the ratio-transformer bridge (4) of the Electrical and Physical Standards operation of General Electric's General Engineering Laboratory. These results are plotted in Figures 1 and 2, wherein the various functional relationships are seen to be similar to those obtained previously in this laboratory. The absolute magnitude (at 3,000 cps), however, is now of the order of $10^2 \mu\text{f}$ per cm^2 of superficial area* -- due to the improved geometric orientation of the test cell.

Experimental work is now in progress to attempt to establish the feasibility of obtaining meaningful data on the solid/solid "interphase" from capacitance measurements by using this very accurate bridge equipment. More specifically, it would be desirable to acquire certain insights into the nature of the measurement, such as might be described by the following questions:

- a. What interfaces are actually being measured? There are at least two "conducting" interfaces in the IEM fuel cell (viz. the metal/aqueous solution interface in the porous electrode and the metal/IEM interface), and the measured capacitance would be affected by both.
- b. Is it experimentally possible to resolve the measured values into the individual interfacial components specified in (a)?
- c. Can identical experimental impedance data be obtained from supposedly identically fabricated cells? The answer to this question should be a test of the veracity of the original assumption.
- d. Can the variations (if any) of the measured values with applied D. C. potential be related to those obtained from "classical" (i. e. metal/aqueous) systems?

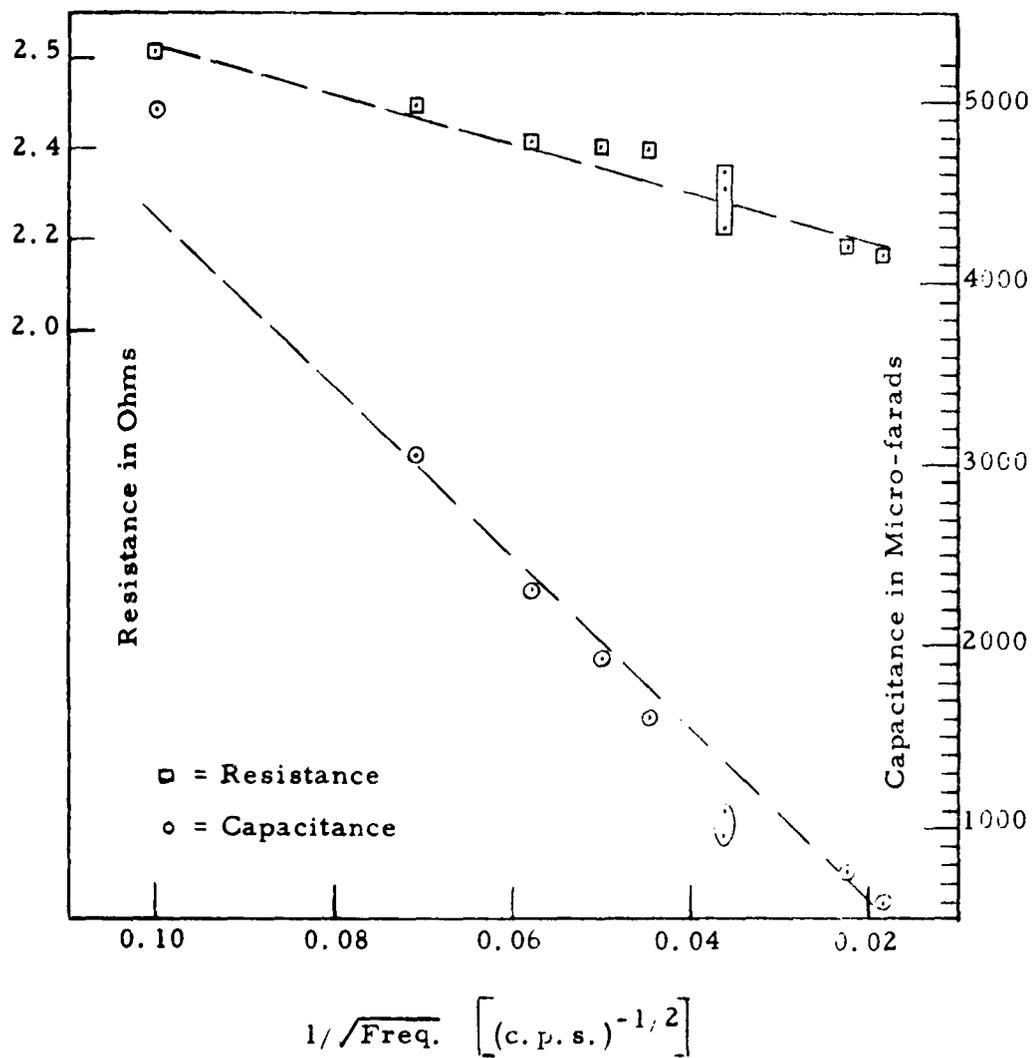
* Based upon areas of ca. 8 cm^2 each for two electrodes connected in series (i. e. transversely across the membrane) with the membrane assumed to be an ideal electrolyte.



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FIGURE 1
CAPACITANCE AND RESISTANCE OF AN
IEM FUEL CELL CONFIGURATION IN AIR:
FREQUENCY DEPENDENCE

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CAPACITANCE AND RESISTANCE OF AN IEM FUEL CELL CONFIGURATION IN AIR AS FUNCTIONS OF (FREQUENCY)^{-1/2}

Fuel Cell Lab.
DECO - G. E. Co.
Lynn, Mass.

FIGURE 2

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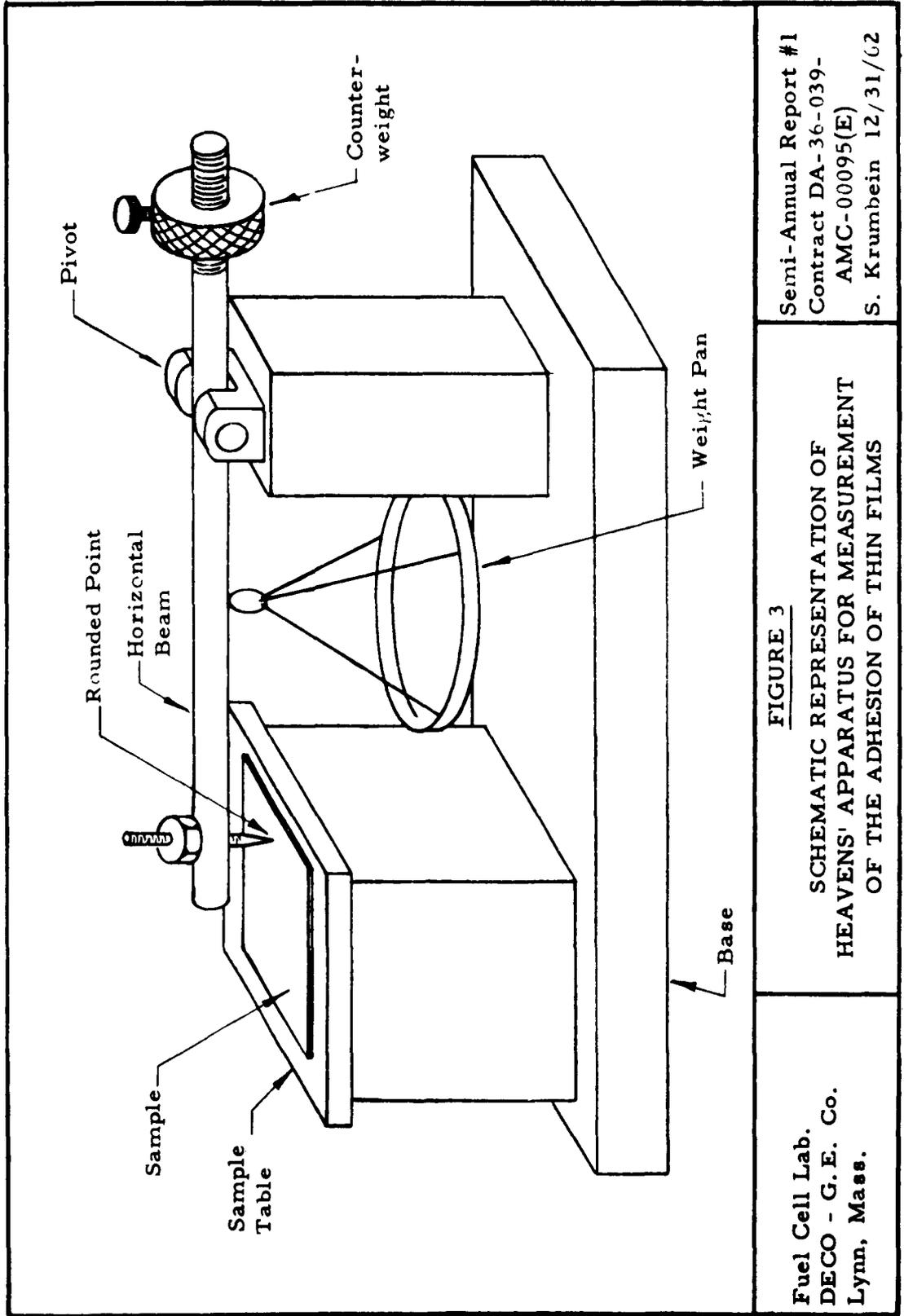
4.0 FACTUAL DATA (Cont'd)

4.1.3.3 Adhesion Measurement

A program is now under study for determining the relative adhesion existing between the catalyst and membrane phases of IEM fuel cell configurations. The method with the most immediate promise appears to be that developed by Heavens (5) and, later, put on a semi-theoretical basis by Benjamin and Weaver (6). Heavens' apparatus is depicted schematically in Figure 3 and consists, essentially, of a horizontal beam with a weight pan at its center and a smoothly rounded point at its end. Samples are drawn across a flat table, directly under the point, so that the latter travels across the metal (i. e. electrode) surface. Weights are added to the pan after each run, until -- at some critical weight -- the film is cleanly stripped off the substrate.

Previous experimental work (6) has shown that the critical vertical forces are determined primarily by the nature of the film/substrate interface. The hard point is assumed to slide over the metal film for increasing loads up to the point where deformation of the substrate causes a loosening of the film. For identical substrate materials (ion-exchange membranes, in our case) comparative measurements of adhesion can be obtained simply from the required vertical load, according to Benjamin and Weaver -- although this parameter will also be a function of the fraction of interfacial area on which actual IEM-metal bonding has taken place. In order to calculate the shearing forces (and, hence, the adhesion energies), however, it will also be necessary to consider the hardness of the substrate.

It is still too early to assess the applicability of a method developed primarily for relatively coherent, very-thin films on "clean" hard substrates to the porous electrode/soft plastic makeup of typical IEM fuel cells. Although there appears to be no theoretical difference in the treatment for the two types of systems (6), certain experimental modifications will undoubtedly have to be developed as work progresses.



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FIGURE 3
 SCHEMATIC REPRESENTATION OF
 HEAVENS' APPARATUS FOR MEASUREMENT
 OF THE ADHESION OF THIN FILMS

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 Lynn, Mass.

4.0 FACTUAL DATA (Cont'd)

4.2 TASK II - Water Transport Across Ion Exchange Membranes

4.2.1 Introduction

This study is divided into three main parts: (1) determination of diffusional water transport by means of permeation measurements; (2) determination of electro-osmotic water transport, and (3) direct measurement of the water gradient established in a membrane by passage of electric current. Almost all the necessary apparatus for all three phases of the program is now available, and some data have been obtained under phase (1) as described below.

4.2.2 Diffusional Water Transport Experiment

A Stabin osmometer was used to determine the hydraulic permeability of a polystyrene sulfonic acid membrane of I. E. C. = 1.65 meg/g 0.065 cm thick. Discs of the membrane were fitted to the osmometer cell, which contains a volume of about 1 cm³ and gives an exposed membrane area of 15.92 cm². Connected to the cell are a capillary tube with a bore cross-section of $1.716 \pm 0.004 \times 10^{-3}$ cm² and a filling tube. A push rod coated with vacuum grease seals the filling tube and makes it possible to adjust the level of the meniscus in the capillary. The apparatus was tested for leaks by immersing in water and applying a pressure of almost 1 atm. by means of a mercury bulb connected to the capillary tube. No bubbles were evident over a period of two hours. The hydraulic permeability was measured by determining the rate of flow in the capillary with a cathetometer and stop watch with distilled water both inside the cell and surrounding it. Measurements were made at 15°, 25°, 35°, and 45°C and with applied pressures up to 35 cm Hg. The membrane was allowed to equilibrate with the surrounding water overnight at each change of temperature. Although the osmometer is made of stainless steel, slight corrosion was observed. In order to minimize any resulting osmotic pressure difference, the water in the cell was replaced by water from the surrounding bath just before each run. At the three lower temperatures, the steady state rate of permeation was measured 20-30 minutes after applying the pressure. At 45°C the movement of the meniscus was observed as a function of time so that the transient period was also included.

4.0 FACTUAL DATA (Cont'd)

4.2.3 Experimental Results

The permeability, P , is given by:

$$P = \frac{sh}{A} \quad (1)$$

where s is the slope of a plot of the molar rate of flow of water through the membrane, h is the membrane thickness, and A is the area available for transport. s was determined by using the method of least squares, requiring the line to pass through the origin. The results are summarized in Table I.

Table I: Hydraulic Permeability of I. E. M. to Water

T (°C)	15	25	35	45
P (mole/cm-atm.-sec) 10^{10}	4.3 ± 0.2	7 ± 2	9.6 ± 0.3	13.5 ± 0.7

Between four and seven points were used in determining each slope.

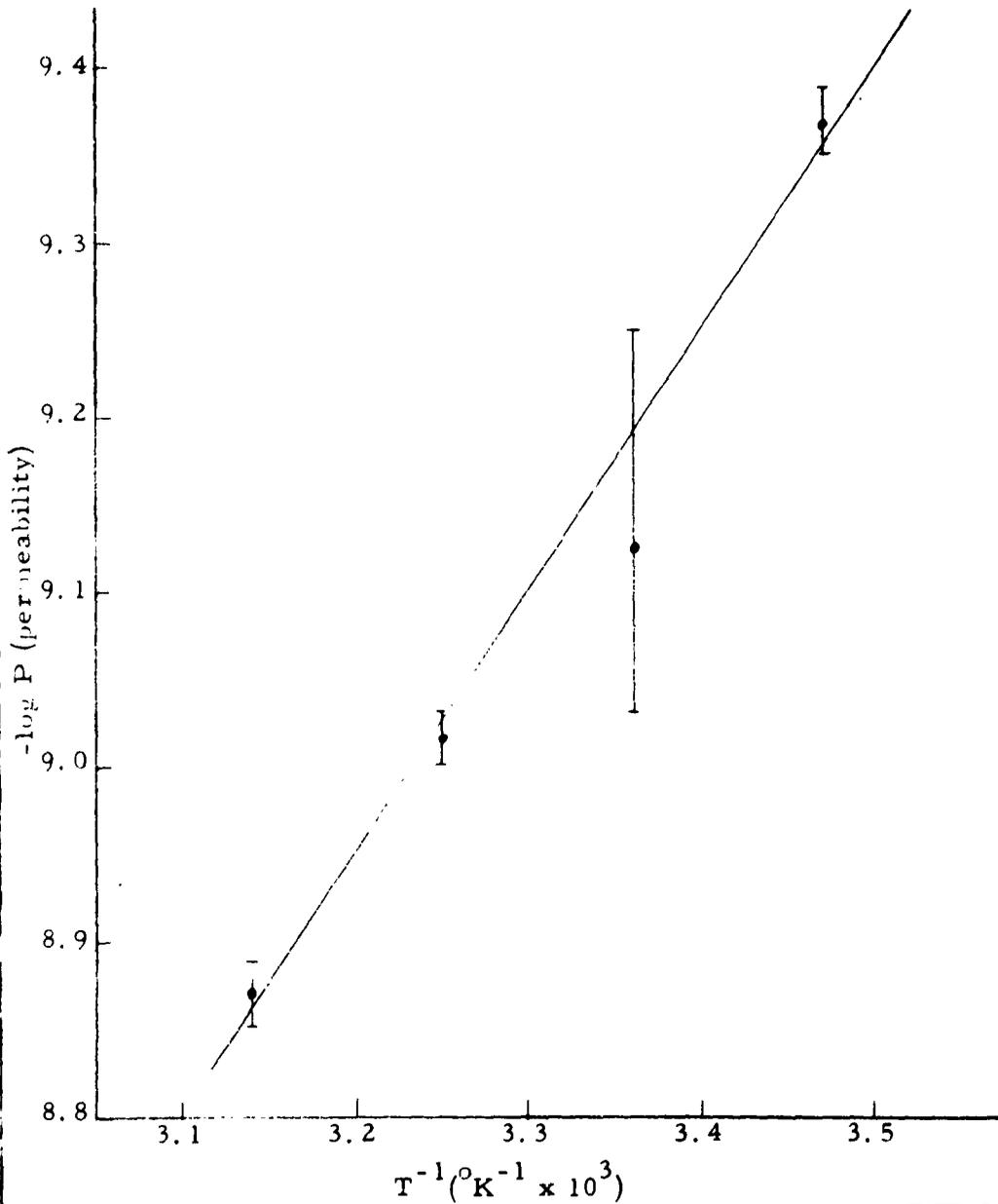
The slope of the plot of $-\log P$ versus $1/T$ is related to the energy of activation in the water transport process. This plot appears in Figure 4. Since the point at 25°C is unreliable, it was not considered in calculating the least squares slope. The resulting activation energy is 6900 cal/mole.

The full permeation-time curve was obtained at 45°C. Figure 5 shows a typical result. Here, the height of the meniscus measured from some arbitrary reference point is plotted as a function of time. There is first a sharp drop due to the displacement of the membrane followed by a slow approach to the steady state. The time required to reach the steady state is imprecise, but it is about the same at every pressure and has the value 28 ± 3 min. If it is assumed that this is a sort of lag time (τ) necessary for the diffusion of water across the membrane, an order of magnitude value for the diffusion coefficient (D) may be obtained from Equation 2

$$D \propto \frac{h^2}{\tau} \quad (2)$$

Proportionality relationships between D and $1/\tau$ can be obtained from various sources (7, 8) and are sufficient to determine diffusion activation energies. In the case discussed in this Report, $D \approx 10^{-6}$ cm²/sec.

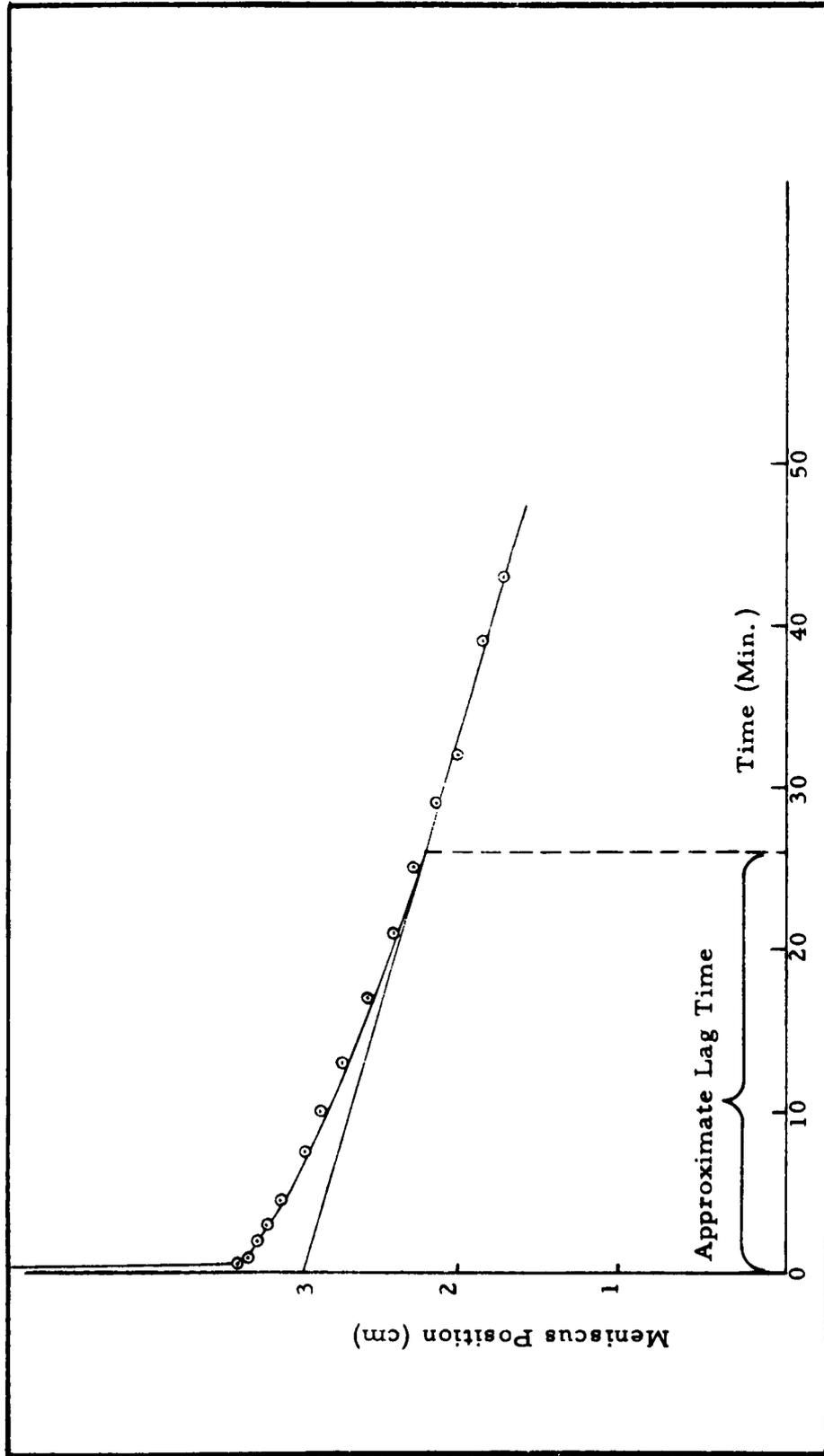
DETERMINATION OF THE ACTIVATION ENERGY
 FOR WATER TRANSPORT THROUGH A
 POLYSTYRENE SULFONIC ACID MEMBRANE



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FIGURE 4

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FIGURE 5
 PERMEATION-TIME CURVE AT 45°C AND
 11.84 CM HG PRESSURE. WATER TRANSPORT
 THRU A POLYSTYRENE SULFONIC ACID MEMBRANE

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4.0 FACTUAL DATA (Cont'd)

Experimental work regarding the determination of diffusional water transport will be conducted on other types of ion exchange membranes in parallel with phases 2 and 3 described above.

4.3 TASK III - Kinetic Study of the Electrochemical Oxygen Reduction at Variable Oxygen Partial and Total Pressure

4.3.1 Introduction

Rates of cathodic oxygen reduction, that is, cell currents, are dependent on partial pressures of the oxidant if rate-controlling steps involve the concentration or partial pressure of oxygen. This is likely to be observed since reaction rates will be either liquid film or gas diffusion controlled. However, cases can arise where removal of the reaction product may be hindered by slow transport processes, thus resulting in, possibly, appreciable lower rates. This could be the case of water removal from the catalyst surface of an oxygen electrode. If local current densities are either dependent on partial pressures of oxygen or water, it will become necessary to establish relationship predicting such local current densities and to design electrode geometries favorable to uniform current distribution, and as a result uniform distribution of the main influential variables affecting oxygen electrode performance.

4.3.2 Objectives

The ultimate goal, that is, quantitative description of current-voltage relationships for an IEM/electrode structure as a function of main variables, can be attained by involved studies, including at least:

- the determination of limiting current densities
- an attempt to establish the rate-controlling process for known electrode structures in the current density range corresponding to 0.85 - 0.5 volts
- the derivation of relationships describing the current-voltage behavior over practical operational ranges

Experimental results, as well as the equations derived to predict limiting local current densities for purely gas-diffusion controlled processes, obtained during this period will be discussed in more detail in the succeeding Semi-Annual Report when more experimental evidence will be available.

4.0 FACTUAL DATA (Cont'd)

4.3.3 Experimental Equipment and Procedures

Partial as well as limiting currents were obtained by applying manually set voltages by means of a 300 ma potentiostat. A Luggin capillary/SCE is used as the reference electrode, the capillary tip being tightly held against the membrane. Such reference electrodes have been described and discussed elsewhere (1). Due to the limited current output of the potentiostat and to expected high limiting currents at high partial pressures of oxygen, the electrode surfaces were either 0.33 or 0.70 cm² (Figure 6). These electrodes could be operated for forced flow conditions (Figure 6 B-1) or as a self-breathing device (Figure 6 B-2). Currents were recorded for short periods of time on a Leeds and Northrup Co. Speedomax G Recorder at chart speeds of about 30 cm/min. It was hoped that surface conditions regarding water partial pressure and accumulation would be unchanged during such short time intervals. Conditions surrounding the electrode were kept constant by placing the device into a constant temperature and humidity chamber (Aminco Climate-Lab). Electrode surface temperatures were measured by a fine Pt-Pt/Rh thermocouple, inserted into the electrode catalyst layer.

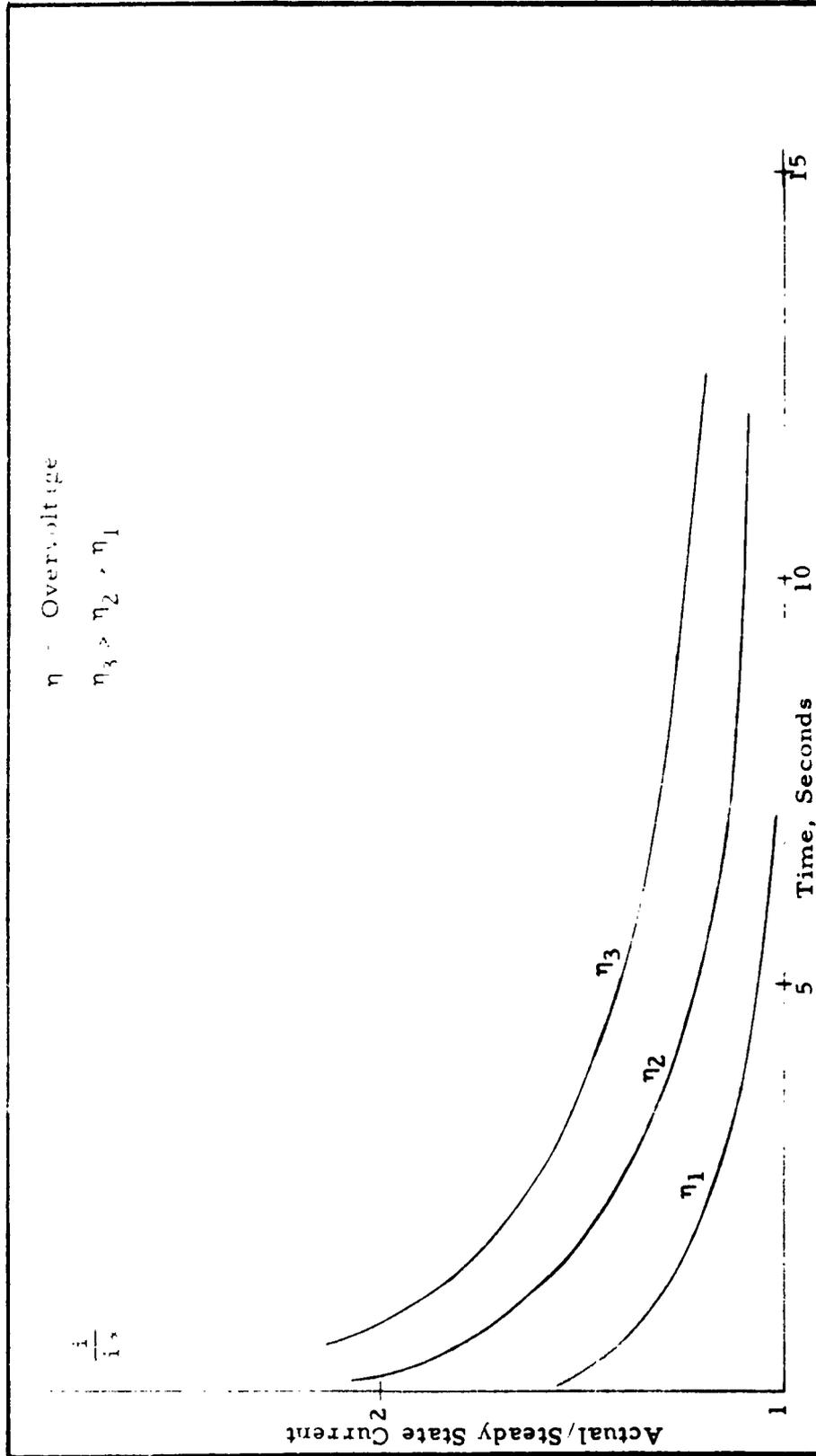
4.3.4 Experimental Results

Current Decay

All experimental results obtained by applying fixed potentials display initially large "surge" currents which are not directly relevant to the present report. However, currents would decay rather rapidly, as represented in Figure 7, to steady state or quasi-steady state values. Steady state values reported in the following figures represent extrapolated values to $t = 0$ of the linear current-time function of Figure 7. As expected, currents would increase with increased applied potentials up to certain limiting currents.

Limiting Currents

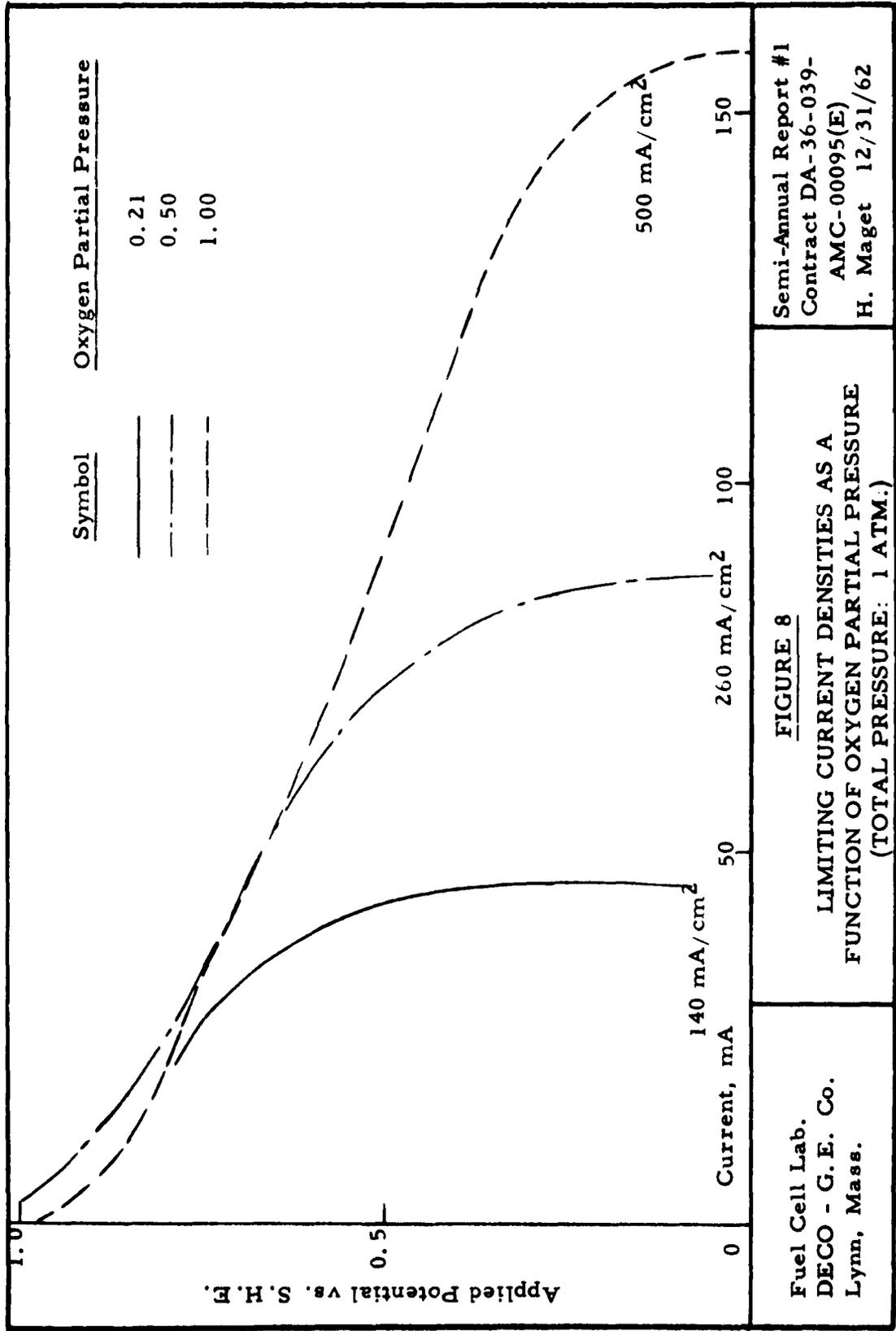
Such limiting currents are represented in Figure 8 for various oxygen partial pressures (air, 50% O₂/50 N₂, pure oxygen) at 22°C and 1 atm. total pressure. Two important results are noticeable: 1. Up to about 70 mA/cm² the polarization curves do behave almost identically, independently of partial pressure, and 2. Limiting current densities are nearly proportional to oxygen partial pressure. These results obtained for large flow rates indicate that air-operated electrodes may be practical for current densities up to about 100 mA/cm².



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FIGURE 7
CURRENT-TIME BEHAVIOR AT
VARIOUS APPLIED POTENTIALS

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FIGURE 8
 LIMITING CURRENT DENSITIES AS A
 FUNCTION OF OXYGEN PARTIAL PRESSURE
 (TOTAL PRESSURE: 1 ATM.)

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4.0 FACTUAL DATA (Cont'd)

Operating Conditions

Since gas flow conditions are doubtlessly very influential on limiting current densities, preliminary measurements have been reported in Figure 9 to indicate such influence. Limiting current densities of 140, 130 and 110 mA/cm² were obtained for forced air flow (Figure 6 B-1), exposed electrodes (Figure 6 B-2) and open-end tube set-up (Figure 6 B-1) respectively. Data reproducibility for two electrode sizes is represented in Figure 9B.

4.3.5 Specific Conclusion

Definite conclusions would be premature, although preliminary results indicate that in an air-channel operating under conditions such that large gradients of oxygen partial pressure are present, current density distributions may be very unfavorable for proper electrode operation.

4.0 FACTUAL DATA (Cont'd)

4.4 TASK IV: Heat and Mass Transfer Investigations of the
Air-Breathing IEM Cell

4.4.1 Introduction

The stable operation of the General Electric Ion Exchange Membrane Fuel Cell on hydrogen and force convected air at 72°F and 0% R. H. at high current densities, i. e. about 50 amps/ft², was virtually impossible with wicking and possible without wicking for relatively short periods of time, only 1 hour maximum, using the standard air electrode-current collector. This restriction on the level of performance and the period of steady operation was due to the uneven evaporation of product water from the catalyst surface and its removal from the air passages caused by poor air distribution. Since good mass and heat transfer studies depend on the capability of maintaining steady operation for long periods of time, an air electrode-current collector design which had this characteristic was imperative.

The objective of the experimental work presented in this report was to determine the feasibility and the operating characteristics of a recently modified air-electrode current collector as compared to those of the standard collector design.

The procedures used and the preliminary results obtained are presented in this section of the report.

4.0 FACTUAL DATA (Cont'd)

4.4.2 Experimental Procedure

The ion exchange membrane fuel cells used in this investigation consisted of GPS type membranes and the standard wet-proofed platinum black electrode.

The single fuel cell fixture had the same amount of insulation and nearly the same dimensions as the one used in the five cell stack studies reported elsewhere (1). The fuel cells were bonded to standard hydrogen collector frames. The air electrode-current collectors were various configurations of a newly designed type. Some runs were made with the standard collector for comparison. Compressed bottled air flowed at a reduced pressure of a few inches of water across the surface of the catalyst at right angles to the direction of hydrogen flow. The electrode-current collectors contacted the catalyst in crossed-ribs fashion.

The geometrical catalyst area was marked off with insulating tape to 0.07-0.08 ft².

The air flowrate was regulated by a calibrated flowmeter.

The temperature gradients along the catalyst surface in directions perpendicular and parallel to the direction of air flow and across the air gap were determined by six platinum/platinum - 10% rhodium thermocouples placed in a pattern across the catalyst surface and another six directly opposite these on the collector surface.

The fuel cell resistance under load was determined by dividing the difference between the AC voltage (voltage uncorrected for $i R_C$ drop using a Kordesch-Marko Bridge) and the DC voltage by the current.

4.4.3 Results and Discussion of Results

4.4.3.1 A Comparison of Maximum Hydrogen-Air and Dead-Ended Hydrogen-Oxygen Performance

The maximum performance obtained on recently prepared ion exchange membrane fuel cells of the GPS type without wicking in an insulated fixture on hydrogen and force convected air at 72°F and 0% R.H. with 100% product water removal is shown in Figure 10.

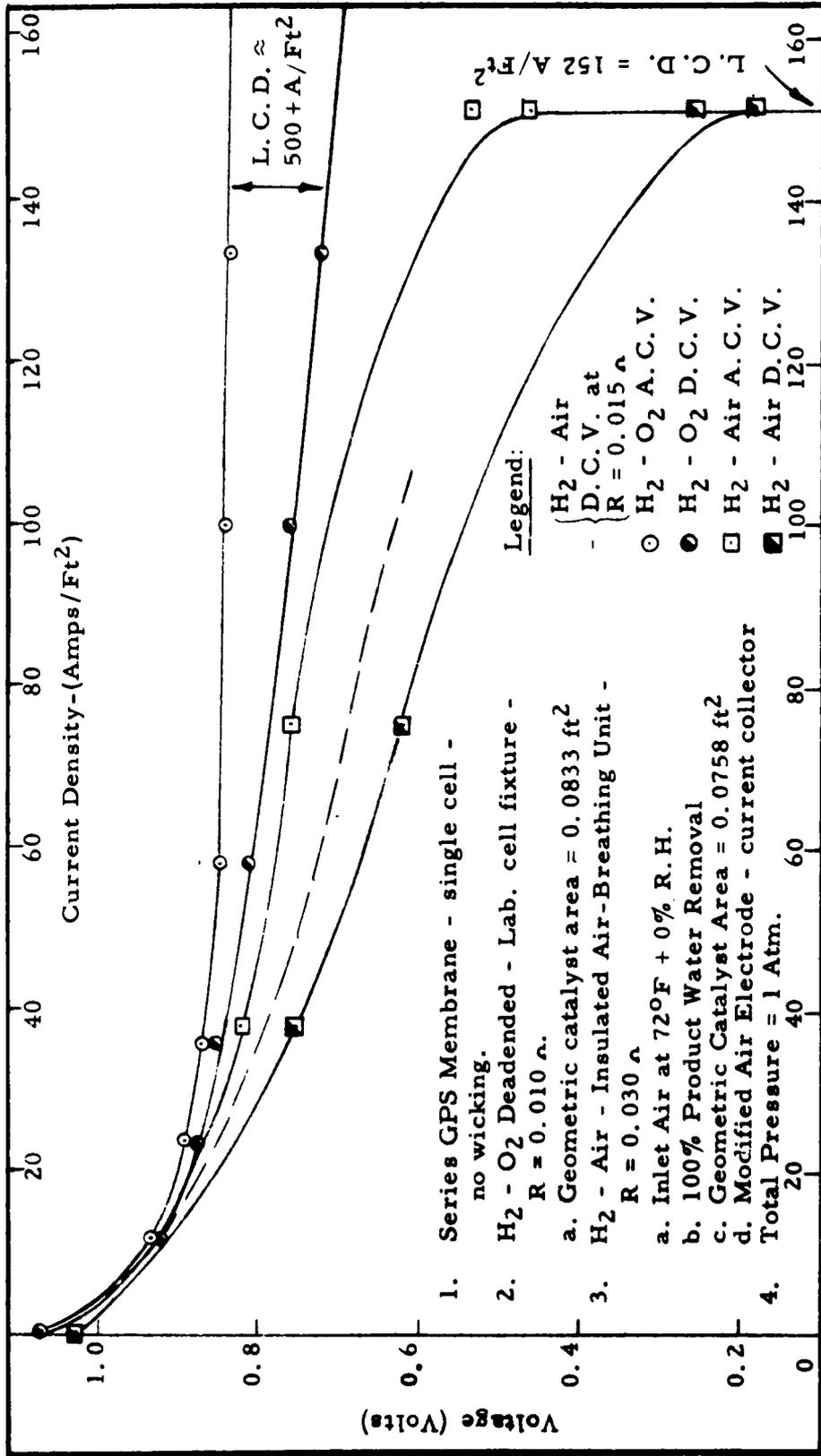


FIGURE 10

A COMPARISON OF MAXIMUM HYDROGEN-AIR PERFORMANCE WITH DEADENDED HYDROGEN-OXYGEN PERFORMANCE

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4.0 FACTUAL DATA (Cont'd)

The data points were maintained for periods of from one to seven hours except at the limiting current density of 152 amps/ft² where the voltage decreased within fifteen minutes to very low values while the current remained essentially constant. At current densities of 50 and 100 amps/ft², the AC (uncorrected for iR_c loss) and DC voltages were 0.790 and 0.700 volt for the former and 0.710 and 0.540 volt for the latter. By comparison, the AC voltages for dead-ended hydrogen and oxygen at 50 and 100 amps/ft² were 0.855 and 0.840 volt. The limiting current density obtained by extrapolation was over 500 amps/ft².

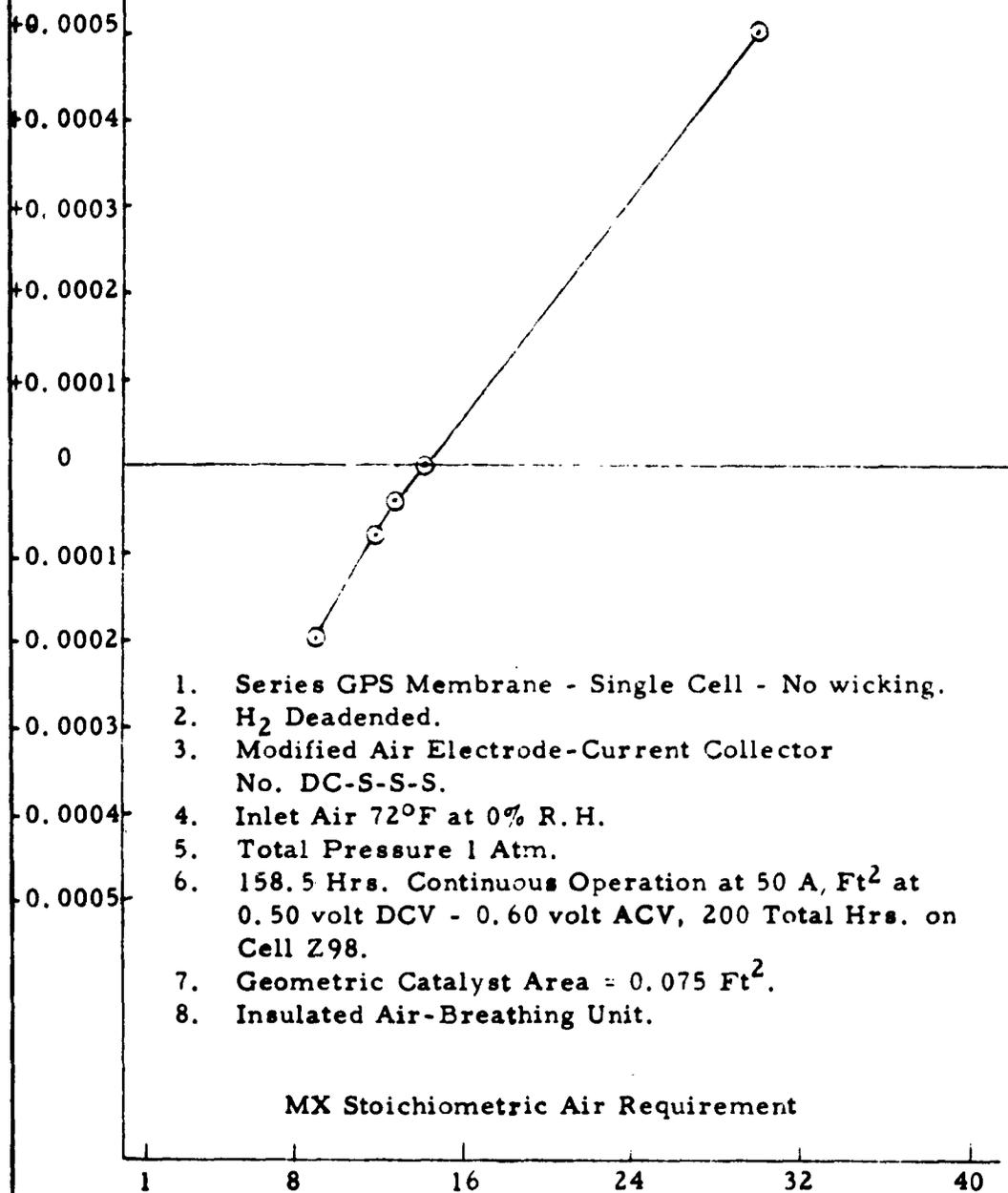
Cells which had aged in either the wet or the dry state 6 months before use gave voltages which were 0.15 and 0.30 volt less than the maximums at 50 and 100 amps/ft², respectively. (Ref. Table II) Higher DC voltages on hydrogen and air, 0.75 and 0.63 volt at current densities of 50 and 100 amps/ft², should be possible with the use of lower resistance cells (Ref. Figure 10). The changes in cell design needed to lower cell resistance are known.

The reasons for the differences between the limiting current densities for oxygen and air seem to be dependent solely on oxygen partial pressure. The detailed explanation is presented in another section of this Progress Report.

4.4.3.2. The Effect of Air Flowrate on the Rate of Change of Cell Resistance

The effect of air flowrate on the rate of change of cell resistance is shown in Figure 11 for the GPS type membrane fuel cell (Z98) with a modified collector type DC-S-S-S (Ref. Table II). A plot of cell resistance versus time for a given air flowrate indicated that after a few minutes the time rate of change of cell resistance was essentially linear for cell resistances lower than the value at the maximum cell performance point and slightly higher than this value. The air flowrate at which the time rate of change of cell resistance was zero represented the 100% product water removal point. Below this reference level, the cell was wetting, while above it the cell was drying. Similar curves were plotted for each of the other modified air electrode-current collectors as well as the standard type to yield the air flowrate at which 100% of the product water was removed from the air electrode. These data are reported in Table II under nXST, the number of times the stoichiometric air requirement at the specified current densities in the range 31 to 76 amps/ft².

THE EFFECT OF AIR FLOWRATE ON THE RATE
OF CHANGE OF CELL RESISTANCE FOR THE CELL
RUN CONTINUOUSLY FOR 158.5 HRS.
AT 50 AMPS/FT²



1. Series GPS Membrane - Single Cell - No wicking.
2. H₂ Deadended.
3. Modified Air Electrode-Current Collector
No. DC-S-S-S.
4. Inlet Air 72°F at 0% R.H.
5. Total Pressure 1 Atm.
6. 158.5 Hrs. Continuous Operation at 50 A, Ft² at
0.50 volt DCV - 0.60 volt ACV, 200 Total Hrs. on
Cell Z98.
7. Geometric Catalyst Area = 0.075 Ft².
8. Insulated Air-Breathing Unit.

MX Stoichiometric Air Requirement

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FIGURE 11

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FACTUAL DATA (Cont'd)

This technique will be useful in experimental work for determining whether individual cells in a membrane stack are wetting or drying or at the 100% product water removal point. (The equipment for doing this on small and large cell stacks is on hand.) The change in the rate of drying under a drying air flowrate for a given current density as the mode of evaporation changes from a flooded catalyst surface, to the pores of the catalyst and, finally, to the membrane, might also be studied by this method. The wetting condition could be studied by lowering the air flowrate so that less than 100% of product water is removed. Correlation of these resistance changes with independent measurements of the change in resistance of a dry catalyst electrode to one saturated with product water would have to be made as a function of temperature as well as with resistance change of the membrane as a function of water content (this data is available.) This data can be checked with drying experiments conducted without the occurrence of the electrochemical process.

The extrapolation of single cell data consisting of the rate of change of cell resistance versus air flowrate to 1.0 times the stoichiometric air requirement might yield the rate of evaporation due to cell heat rejection when correlated with the catalyst electrode-product water resistance data.

In conclusion, there is a possibility of determining electrically the separate contributions of cell heat rejection and air flowrate on the evaporation rate of product water while the cell is in operation.

4.0 FACTUAL DATA (Cont'd)

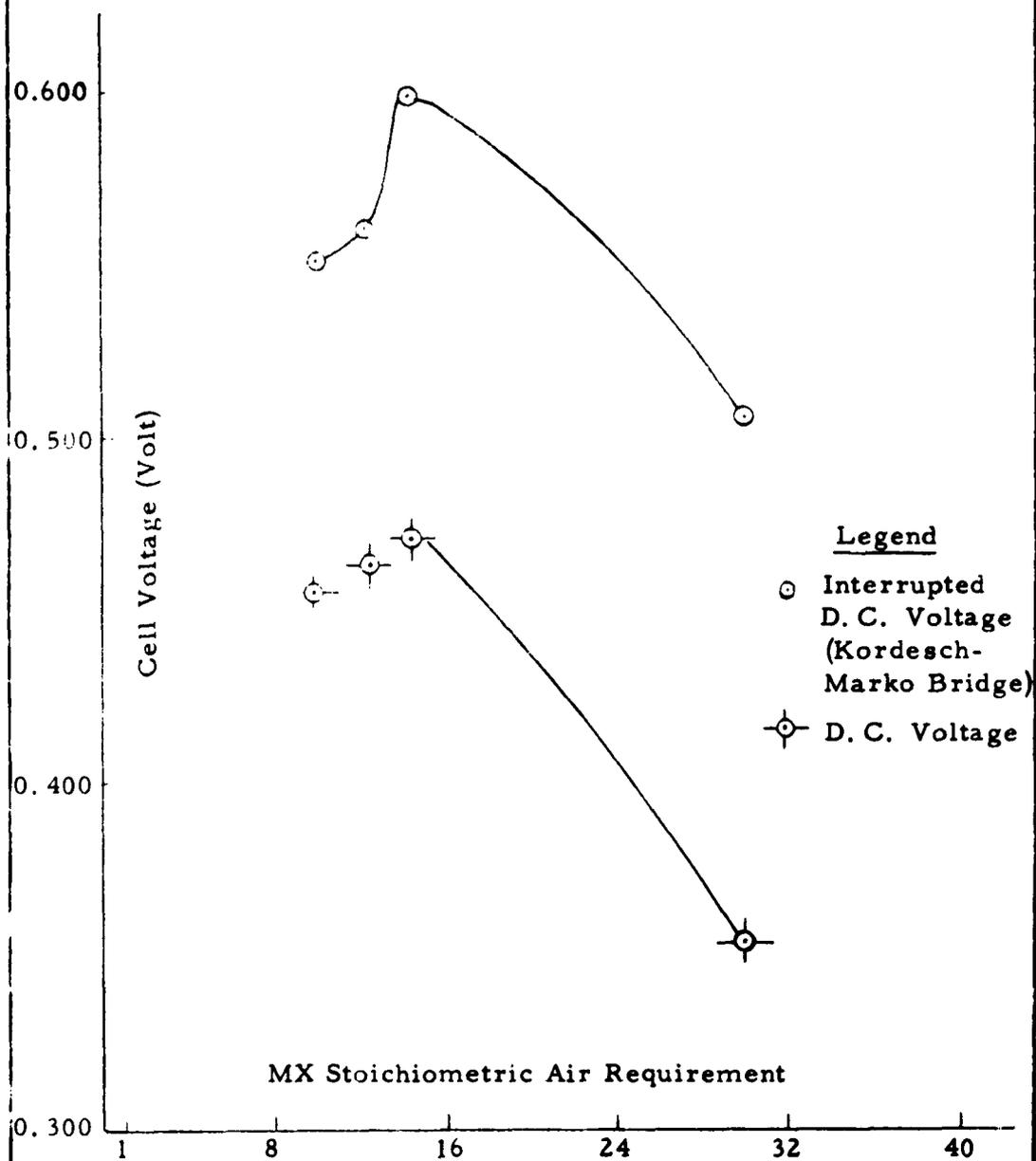
4.4.3.3. The Effect of Air Flowrate on Cell Voltage and Resistance

The effect of air flowrate on the AC and DC cell voltages and resistance are shown in Figure 12 for the fuel cell described in section 4.4.3.2. The data represent the cell with about 200 hours of accumulated operating time. The peak voltages occurred when the cell resistance was slightly higher than the minimum value. As the air flowrate was increased above the 100% product water removal point, the voltages decreased as the cell resistance increased due to drying. When the air flowrate was decreased below the 100% point, the voltages and cell resistance decreased to a minimum value for the sixteen hour data point.

The DC voltage as a function of the rate of evaporation of product water can be used in a practical system to locate and maintain the cell performance at the maximum point. For example, if the DC voltage of a cell is decreasing, the cell can be either wetting or drying. If an increase in air flowrate causes the voltage to decrease at a faster rate, the cell is in the dry region. If an increase in air flowrate causes the voltage to increase, the cell is in the wet region. Thus, whether by manual or automatic adjustment of air flowrate, the cell performance in a stack may be brought to the maximum performance point. Complications to this scheme are the same as those discussed under Section 4.4.3.2.

The magnitude of the deviation of the AC voltage from its maximum value at any time for a given flowrate may be useful in estimating the effective water film thickness on the air electrode. Resistance versus temperature data for product water must be available. This value may then be compared with the effective film thickness calculated from the diffusion equation.

THE EFFECT OF AIR FLOWRATE ON CELL
 VOLTAGE AND RESISTANCE FOR THE CELL
 RUN CONTINUOUSLY FOR 158.5 HRS.
 AT 50 A/FT²



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FIGURE 12

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4.0 FACTUAL DATA (Cont'd)

4.4.3.4 Continuous Run on Hydrogen-Air With Modified Air Electrode-Current Collector Without Wicking

An ion exchange membrane fuel cell of the GPS Type (Z-98-4 months old) in an insulated single cell fixture with a modified air electrode-current collector (DB-S-S-S* Ref. Table II) was operated continuously for 158.5 hours without wicking on air at 72°F and 0% R.H. at voltages of about 0.600 A.C. and 0.500 volt D.C. at a current density of 50 amps/ft². The cell which was set for low voltage automatic hydrogen shutdown and an upward hydrogen purge about every 90 minutes was left unmonitored for five periods of sixteen hours overnight with the air flowrate set for a slightly wetting condition (10-12XST). During these periods the voltage decreased about 50 mv. When manually monitored the cell was kept at peak performance (100% product water removal at the optimum cell resistance) for periods up to seven hours with only a 10 mv change in cell voltage. The cell was not left at the 100% product water removal point during the unmonitored period at night because any change in air flowrate or load might cause an extreme drying condition. Over a 30 hour unmonitored period during a week-end, the cell voltage decreased 0.150 volt at an air flowrate which was about 6 times the stoichiometric air requirement instead of the 14 required to remove 100% of the product water.

The average cell temperature (t_c in Table II) was 108 ± 6°F with a range of 100 to 118°F. The temperature gradients perpendicular (Δt in Table II) and parallel (Δt in Table II) to the direction of air flow were about equal at 5.0°F/in. of catalyst surface. The gradient from the catalyst surface across the air gap to the current collector was 0.6°F (Δt gap in Table II). Compared to the standard air electrode-current collector (S in Table II) which had a ratio of a Δt gap of 1.1°F at 2/3 the current density and 1/4 of the air flowrate, these temperature characteristics are very favorable.

During the 158.5 hours of operation about 236 g of product water was formed, all of which was removed both by the air and the auxiliary water removal system. The air electrode-current collector on inspection after the run showed no water accumulation. The gas volume of the air collector was about 15 cc. About 5.5 g of water which accumulated on the hydrogen side were part of the 9.5 g of water lost from the hydrogen water bubbler during the run. Since the hydrogen purge was upward, none of the water collected on that side could be removed during the run.

4.0 FACTUAL DATA (Cont'd)

- 4.4.3.4 This means that no product water was transferred from the air side to the hydrogen side, but 57% of the water which entered the hydrogen gas compartment was transferred to the air electrode side. This result is in exact agreement with all other previous overall water balances performed on both single and multi-cell I. E. M. units without wicking. Ref. "Ion Exchange Membrane Fuel Cell Report No. 3; ARPA Order No. 80-61, U. S. Army Signal Corps. Res. and Dev. Lab., Ft. Monmouth, N. J., Oct. 1961-1962, Final Technical Summary Report, Task A-Phase 5.

The accumulation of water in the hydrogen electrode gas compartment from the hydrogen water bubbler may be eliminated by passing the hydrogen through a small bed of silica gel after the water bubbler or water adsorbent material may be placed on the hydrogen collector to spread the water evenly so that possibly 100% of it gets transferred to the air electrode.

The decrease of 0.100 volt in A. C. voltage during the run was due to contamination of the catalyst and membrane with iron from the 304 S. S. collector since appreciable amounts of rust were found where the ribs contacted the catalyst.

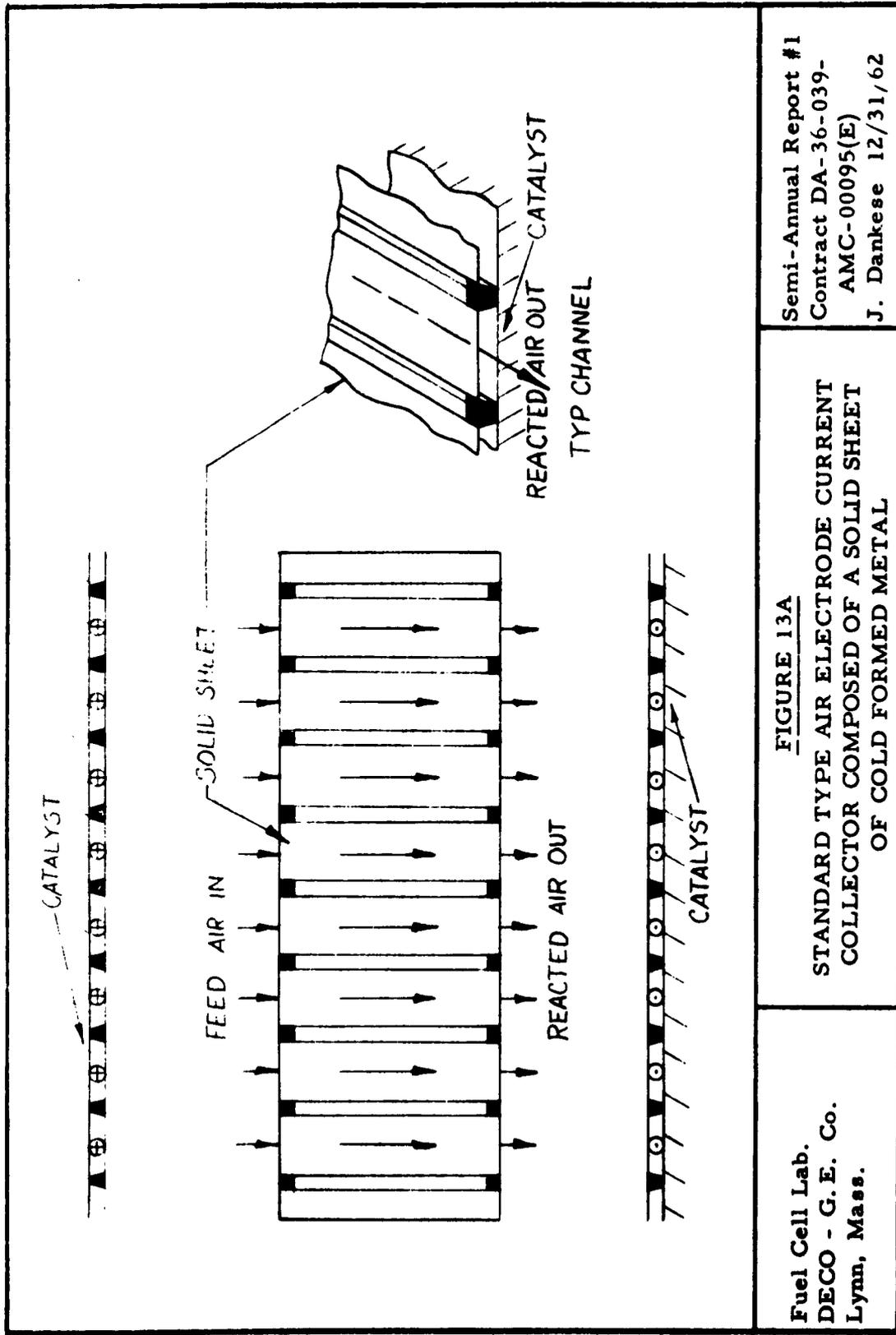
4.4.3.5 Comparison of the Modified Air Current-Collectors With The Standard Type

In Table II - Summary of Data, a basic comparison between the operating characteristics of the various configurations of the modified air electrode-current collector and the standard type is presented. (Ref. Figure No. 13) The symbol nXS refers to the number of times smaller or larger a given measured variable is than the value observed with the standard air collector at 38 amps/ft².

The current densities used during the experimental work consisted of two ranges, 31 - 38 amps/ft² and 63 to 76 amps/ft².

1. Product Water Management

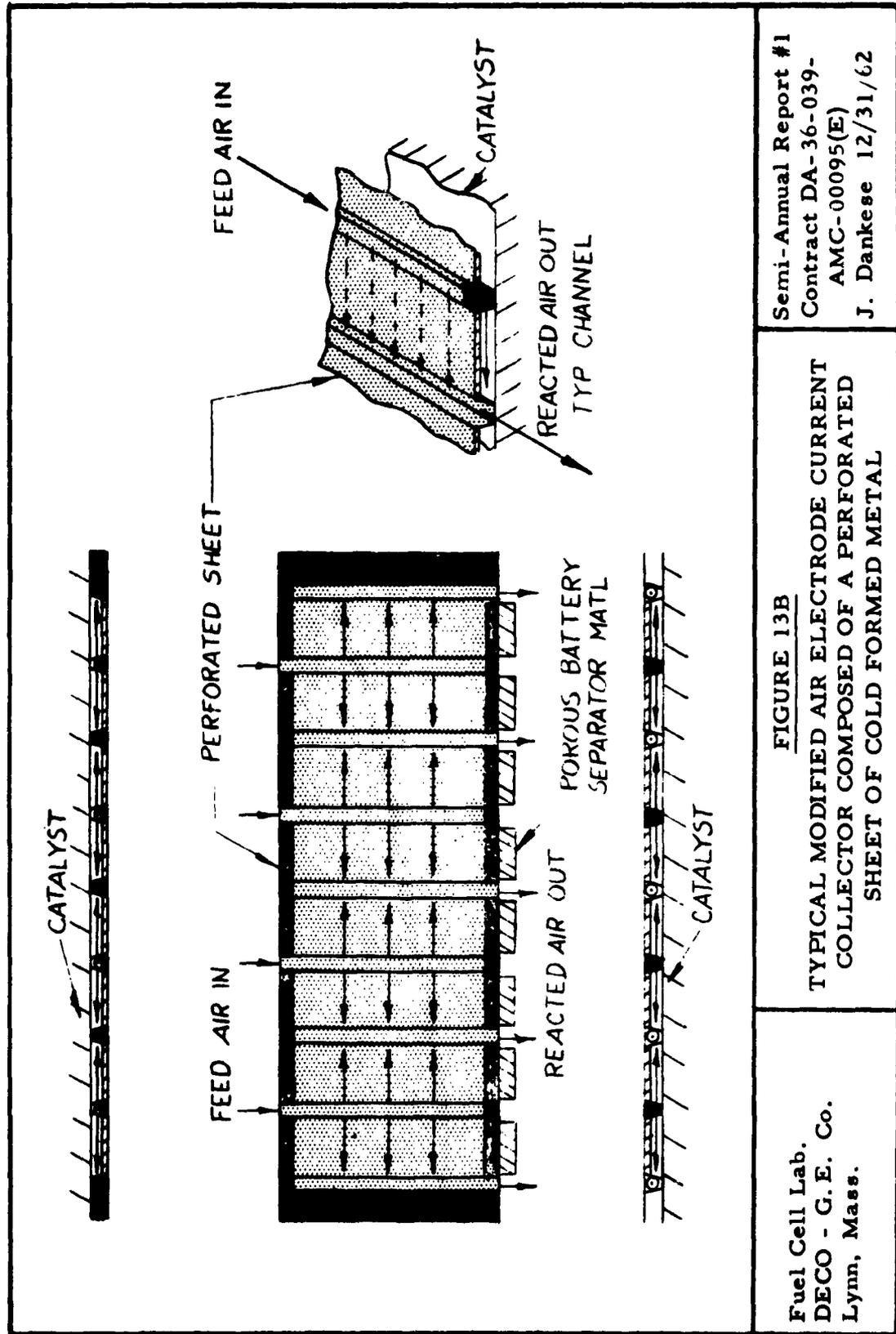
After only 2.2 hours of operation at a current density of 36 amps/ft², 33% of the air channels of the standard air electrode-current collector became clogged with product water and after 3.3 hours of operation the performance decreased markedly. Removal of the water from the channels was not possible by increasing the flowrate which instead caused severe drying gradients.



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FIGURE 13A
 STANDARD TYPE AIR ELECTRODE CURRENT
 COLLECTOR COMPOSED OF A SOLID SHEET
 OF COLD FORMED METAL

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FIGURE 13B
 TYPICAL MODIFIED AIR ELECTRODE CURRENT
 COLLECTOR COMPOSED OF A PERFORATED
 SHEET OF COLD FORMED METAL

Fuel Cell Lab.
 DECO - G. E. Co.
 Lynn, Mass.

4.0 FACTUAL DATA (Cont'd)

Although the direction of flow was downward, gravity action alone was insufficient to remove product water. Operation at 72 amps/ft² for more than minutes was impossible. In comparison, even the first configurations of the modified air electrode-current collector permitted operation at 31 amps/ft² for periods up to 9 hours and 2.5 hours at 63 amps/ft² with only about 50% of the collector covered with product water and 25% of the flow area plugged. Later versions were run intermittently for periods up to 22.5 hours which consisted of up to 8 hours at 66 amps/ft² and 14.5 hours at 33 amps/ft² with no product water accumulation. Finally, a continuous run for 158.5 hours at 50 amps/ft² was run with no product water accumulation.

2. Collector Corrosion

Since the only materials which were readily available for the fabrication and evaluation of the various versions of the modified air electrode-current collector were 304, 316, and 347 stainless steel, corrosion of the collectors with time was expected. Of course, 304 S.S. was the worse material but was chosen for the extended performance test because that collector configuration was the most promising one. Future collectors will be fabricated from Carpenter, 20Cb stainless steel since this material has shown essentially no tendency to corrode or affect fuel cell performance when in contact with the catalyst for periods up to 5000 hours. The standard air electrodes were made of this stainless steel and therefore did not corrode.

3. Temperature Gradients

a. Maximum Cell Temperature Gradient (Δt_c Max.)

Maximum temperature difference across the catalyst surface were reduced to as little as 0.3 (8°F) times the gradient observed with the standard collector (23°F) at a current density of about 38 amps/ft² and to about the same value (23°F) even when the current density with the modified collector was double that of the standard.

FACTUAL DATA (Cont'd)

Although the modified collectors do reduce the maximum cell temperature differences markedly, they will not eliminate temperature gradients caused by variations in catalyst activity, membrane resistance, and control between the electrode and membrane across the I. E. M. fuel cell surface. There is evidence that this condition exists. (Ref. Random Temperature Variation reported in "Ion Exchange Fuel Cell Report Number 2.", ARPA Order No. 80-61, U. S. Army Signal Corps. Res. and Dev. Lab., Ft. Monmouth, N. J., Jan. 1, 1962-June 30, 1962, Second Semi-annual Technical Summary Report, Task A-Phase 5.)

b. Cell Temperature Gradients Perpendicular and Parallel to the Direction of Air Flow

Cell temperature gradients in directions perpendicular, Δt_x , and parallel, Δt_z , to the direction of air flow were varied from the same gradients as for the standard collector, $9.7^\circ\text{F}/\text{in}$ in the z direction and $2.5^\circ\text{F}/\text{in}$ in the x direction, to as little as 0.02 times the z value and 0.7 times the x value at the same current density. At double the current density, these gradients were reduced to as low as 0.2 to 0.7 times the z and x values for the standard collector at one half the current density. The important point is that these gradients can be balanced equally at a minimum value by the proper configuration of the modified air-electrode current collector whereas with the standard collector the ratio of the z value to x value is fixed at 3.9. For example, the cells run continuously for 158.5 hours at $50 \text{ amps}/\text{ft}^2$ had a ratio of 1.2 while the gradients themselves were about $5.0^\circ\text{F}/\text{in}$.

c. Temperature Gradients Across the Air Gap

Temperature gradients across the air gap from the catalyst to the collector surfaces were varied from 0.6 to 4 times the gradient observed with the standard collector at the same current density. At a current density of $50 \text{ amps}/\text{ft}^2$, the extended operation cell (158.5 hours continuous) had a Δt gap of 0.6°F . This indicates that the vapor suppression part of the collector was effective.

4.0 FACTUAL DATA (Cont'd)

4. Air Flowrate

With the modified air electrode-current collector, air flowrates up to 4 times that required by the standard electrode to remove 100% of the product water at 36 amps/ft² were possible. The same levels of air flowrate were also permissible at double the current density in spite of the increased cell temperature level. This result shows that the vapor suppression device was functioning.

4.4.3.6 Effect of Catalyst Masking on Cell Performance

The effect of masking the catalyst area from the air stream with a 0.027" thick layer of porous inert material is shown in Table II under Collector DC-D-L-L-PC. At a current density of 38 amps/ft² which was maintained for six hours, a 0.150 volt loss in voltage was observed. The temperature drop across the layer was 13°F. Some evidence of the suppression of water rate of evaporation is indicated by the fact that the air flowrate was 3 times that required to remove 100% of the product water from the bare catalyst surface.

Much thinner layers of material may be useful for suppressing evaporation of water with higher air temperatures (120°F).

4.4.4. Specific Conclusions

1. A modified air electrode-current collector has proved feasible for use with ion exchange membrane fuel cells since stable operation of a single GPS type ion exchange membrane fuel cell (catalyst area 0.075 ft²) without wicking was maintained continuously for 158.5 hours on air at 72°F and % R.H. at a current density of 50 amps/ft² with A.C. and D.C. voltages of 0.600 and 0.500 volt, respectively. Removal of product water from the cell was 100% complete. Product water transfer to the hydrogen electrode was found to be non-existent. 110 of the total hours of operation were unmonitored with the longest and shortest periods being 30 and 16 hours, respectively. In comparison, the standard air electrode-current collector could only be run for about one hour maximum under the same conditions before plugging of the air channels with water caused performance to decrease rapidly.

4.0

FACTUAL DATA (Cont'd)

1. Corrosion of a 304 S.S. collector caused a loss of about 0.100 volt A.C. due to catalyst contamination.
2. A total of 260 hours of testing have shown that the modified air-current collector evaporates product water from the air electrode more evenly because of much better air distribution than is possible with the standard type. Higher air flowrates for a given current density are possible because of more even air distribution and a vapor suppression component incorporated into the collector. Auxiliary product water removal is another function of this part. Product water plugging of critical air flow areas is prevented by local high air velocities compared to those in the standard collector. The modified collector shows good potential for large cell sizes, i. e. 8" x 8". There is also the possibility of reducing the temperatures of middle cells in a stack by a suitable configuration of this collector.
3. An electrical technique for determining the air flowrate at which 100% of the product is removed from the air electrode surface has been successfully used. This method can be used to obtain product water removal data for individual cells in stacks. The use of the technique for controlling the performance of the 158.5 hour continuous cell operation was successful. Variations may be used to estimate cell drying rates and diffusion layer effective film thickness
4. The limiting current density for hydrogen air with GPS type I. E. M. fuel cell was found to be 152 amps/ft². This is considerably lower than the +500 amps/ft² for dead-end hydrogen and oxygen. The difference seems to be solely oxygen partial pressure dependent. The maximum performance on air with high performance cells and low resistance fixtures appears to be 0.75 and 0.63 volt D.C. at current densities of 50 and 100 amps/ft², respectively.
5. Preliminary data on the suppression of product water evaporation by masking the catalyst with porous inert material for high inlet air temperatures seems promising.

TABLE II - SUMMARY OF DATA

Type of Collector	Cell No.	A.C.V. Volt.	D.C.V. Volt.	I A/T ₂	I MS	V ₀ OF	Range OF	Δt _c MAX OF	Δt _c MS	Δt _c % OF/Min.	Δt _c MS	Δt _c % OF/Min.	Δt _{gap} OF	Δt _{gap} MS	Δt _{gap} % OF/Min.	MS	Hrs. of Operation	Condition of Collector			
S	2336	0.747	0.669	36	5.0	1.0	95	84	1.0	9.7	1.0	2.5	1.0	3.9	1.0	1.1	17.2	1.0	2.2	H ₂ O chemical plugging occurs. 35% of channels plugged. 50% of collector covered with H ₂ O.	
SS	2336	0.770	0.701	36	17.5	3.5	100	89	1.0	4.4	0.4	3.6	1.4	1.2	0.3	-	-	-	2.2	No H ₂ O	
SS	2336	0.754	0.650	72	6.0	2.4	111	113	1.0	5.8	0.6	5.8	2.3	1.0	0.3	-	-	-	2.2	No H ₂ O accumulation.	
DA-U-L-L	235	0.760	0.660	31	8.5	1.5	92	86	1.0	8.5	0.9	4.0	1.4	2.1	0.5	0.5	10.0	0.6	3.0	50% of collector covered with H ₂ O - 25% of flow area covered - No H ₂ O on cell.	
DA-D-L-L	235	0.683	0.593	31	10.0	1.7	91	84	0.8	4.8	0.5	4.4	1.8	1.1	0.3	1.0	19.2	0.9	6.0	No water accumulation	
DA-D-L-L	235	0.590	0.400	63	10.0	3.5	108	91	38	1.6	1.7	10.9	4.4	0.2	0.2	1.1	99.7	3.5	0.5	No H ₂ O accumulation.	
DA-D-L-L-P	235	0.753	0.653	31	15.0	2.6	93	85	16	0.7	3.4	0.4	3.3	1.0	0.3	0.9	34.6	2.0	2.8	No H ₂ O accumulation.	
DA-D-L-L-P	235	0.710	0.550	63	9.8	3.4	115	101	40	1.7	8.4	0.9	10.7	4.3	0.2	2.2	84.7	4.9	2.5	No H ₂ O accumulation.	
DB-S-S-S	298	0.744	0.672	33	17.0	3.1	95	88	12	0.5	1.7	0.2	2.6	1.0	0.7	0.2	1.7	50.0	2.9	14.5	Some H ₂ O on collector. 1 or 2% of flow area affected. No H ₂ O on cell.
DB-S-S-S	298	0.640	0.500	66	11.5	4.2	120	111	81	0.9	6.5	0.7	6.4	2.6	1.0	0.3	2.7	80.0	4.7	22.5	No H ₂ O accumulation - continuous.
DB-S-S-S	298	0.720	0.640	33	11.0	2.0	97	94	9	0.4	1.1	0.1	3.0	1.7	0.4	0.1	1.0	38.0	2.2	12.5	1-2% of collector covered with H ₂ O. No H ₂ O on cell.
DB-S-S-S	298	0.590	0.444	66	7.5	2.8	120	111	19	0.8	0.2	0.0	7.0	2.8	0.0	0.0	3.0	115.0	6.7	17.0	No H ₂ O accumulation.
DC-S-S-S-P	298	0.610	0.507	50	14.3	4.0	108	100	18	0.8	5.6	0.6	4.8	2.0	1.2	0.3	0.6	17.7	1.0	198.5	
DC-D-L-SL	2108	0.773	0.703	38	6.5	1.4	84	81	8	0.3	1.5	0.2	2.6	1.0	0.6	0.2	0.7	11.5	0.7	5.0	
DC-D-L-SL	2108	0.728	0.568	76	5.5	2.2	107	97	25	1.0	9.0	0.9	3.0	1.2	3.0	0.8	2.6	43.7	2.5	3.8	
DC-B-SL-L	2108	0.723	0.653	38	4.5	1.0	90	84	14	0.6	6.9	0.7	2.7	1.1	2.6	0.7	1.2	19.7	1.1	7.0	
DC-B-SL-L	2108	0.650	0.500	76	6.5	2.6	122	110	22	1.0	13.7	1.4	1.8	0.7	7.6	2.0	1.9	31.1	1.8	17.8	

CONCLUSIONS

Although in some instances conclusions may be premature, since results are only preliminary, general conclusions and trends regarding the reported work may be formulated.

1. The membrane interfacial region seems to be limited to a contact between membrane and electrode rather than appreciable intermixing of the two phases. Thus, bonding is restricted to interaction between electrode and membrane surface.
2. Rates of water transport by hydraulic pressure gradients are rather small, suggesting that during actual fuel cell operation other gradients will have to justify high observed transport rates.
3. Gas-gas diffusional processes, i. e. oxygen through nitrogen and water vapor, may justify observed current densities for and air-breathing electrode, providing air channel geometry is favoring such processes. Current densities up to 500 ma/cm² are observable for pure oxygen, whereas 140 ma/cm² is representative of an air-breathing electrode.
4. New air-breathing current collector design has been allowed steady-state operation of a single cell working on hydrogen and air at current densities of 50 amps/ft² and 0.50 volt terminal voltage.
5. Characteristic variable changes, i. e. cell resistance, during fuel cell operation are suggested as control variables for optimized cell operation and water transport management.

6.0

Program for Next Interval

Planned work, up to the Second Semi-Annual Technical Report (covering the period from January 1, 1963 to June 30, 1963), will remain in agreement with the Technical Guidelines, EPP No. 58-238 of July 17, 1962, and include:

- Task I:** Interfacial membrane/electrode properties-optical examination, capacitance measurements, electrode/IE Membrane bonding forces. Experimental work will emphasize 1. the determination of the relative contribution of electrode/IEM and electrode/liquid electrolyte capacitance to total fuel cell electrode capacitance, 2. the preparation of reproducible samples for optical (microscope) examination and 3. measurements of bonding forces by means of Heaven's apparatus.
- Task II:** Water transport through membranes - permeability coefficients, electro-osmotic transport, conductivity - water content relationships. Experimental work will emphasize 1. permeability and diffusion constants measurements on membranes with various IEC, 2. rates of water transport under conditions of applied potential for membranes with various IEC and 3. calculations of membrane water gradients and conductivity gradients.
- Task III:** Electrochemical oxygen reduction - influence of channel geometry on air-electrode polarization, relationship between current and voltage. Experimental work will emphasize 1. current density distribution for self-breathing electrodes of various channel geometries, 2. current density distribution for forced air flow conditions and 3. analytical interpretation of the results.
- Task IV:** Heat and mass transfer investigation - analysis of an analytical model for the air-breathing electrode - parameters and properties measurements - testing of novel designs. Emphasis will be given to investigations regarding 1. the characteristics of the modified air current-collector at air inlet temperatures up to 120°F, 2. the means of vapor suppression to reduce product water evaporation at inlet temperatures up to 120°F and

6.0

Program for Next Interval (Cont'd)

Task IV: 3. the means of product water conservation for use in (Cont'd) humidifying the inlet air. An automatic cell voltage control device will be tested (based on air flowrate modulation), and applied to single cells. A large fraction of the overall Task will be related to the development of a heat and mass transfer model.

Task V: Design parameters affecting air-operated fuel cell-program to be initiated - general heat and mass transfer analysis - systems control - study of steady state and transient operation - air flow distribution in channels - influence of channel geometry - air electrode testing.

IDENTIFICATION OF KEY PERSONNEL

During the period covering October 1, 1962 to December 31, 1962, the following principal investigators have been associated with the program tasks:

Dr. J. R. Boyack - Water Transport in Ion Exchange Membranes

Mr. J. Dankese - Heat and Mass Transfer - Air Operated IEM Cell

Dr. S. J. Krumbein - Interface Structure and Interfacial Properties

Dr. H. J. Maget - Kinetics of the Electrochemical Oxygen Reduction

Detailed Time DistributionA. Principal Investigators

Dr. J. R. Boyack, Physical Chemist & Electrolytes	392 hours
Mr. J. Dankese, Laboratory Chemical Engineer	438 hours
Dr. S. J. Krumbein, Physical Chemist - Electrochemistry	448 hours
Dr. H. J. Maget, Senior Chemical Engineer	<u>379 hours</u>
	1657 hours

B. Supporting Laboratory Technicians

Mr. J. Amore, Technician - Fuel Cell Electrode	453 hrs.
Mr. R. Donovan, Technician - Laboratory	160 hrs.
Mr. J. Huss, Fuel Cell Evaluation Lab. Technician	472.5 hrs.
Others with individual contribution less than 100 hours	<u>59 hrs.</u>
	1144.5 hrs.

The technical background of principal and main additional contributors is presented in the following resumes:

IDENTIFICATION OF KEY PERSONNEL (Cont'd)James R. Boyack, Ph.D. - Electrolyte Physical Chemist

Dr. Boyack completed all his work for a Ph. D. from the University of Utah in November 1961, (degree June 1962) and joined the Fuel Cell Laboratory of General Electric's Aircraft Accessory Turbine Department in December 1961. He received his B.S. in Chemistry in 1958 at the University of Utah. His doctoral thesis was concerned with zone diffusion and mobility in paper electrophoresis. Included in his background is a summer as research assistant at Eastman Kodak Company.

Since joining General Electric, Dr. Boyack has been working with electrolyte transport mechanisms in various systems such as ion-exchange polymers, gels, etc.

Joseph P. Dankese - Laboratory Chemical Engineer

Mr. Dankese received his B.S. in Chemical Engineering from the Massachusetts Institute of Technology in 1954, and has done graduate work at Northeastern University.

Before joining General Electric in early 1961, he accumulated a wide background in chemical engineering work at Microwave Associates, Ionics, Inc., Dewey and Almy Chemical Co., and A. D. Little, Inc. This included studies of conductive coatings, glass to metal seals, and high temperature sintering of metals; design of the electrical and hydraulic systems for ion-exchange membrane electro dialysis equipment and field testing of the final product; process design and development for the vinyl chloride and butadiene type copolymer plants; developed a battery separator for electric storage batteries (U. S. Patent No. 3,026,366-March 20, 1962); analysis and process design of the liquid oxygen and hydrocarbon fueling system for the Titan missile - this work involved heat transfer, mass transfer and thermodynamics; conducted experimental studies on the effect of radiation on the reactivity of gaseous hydrocarbon in fuel cells; surveyed various power sources including batteries, fuel cells, thermoelectric and thermionic devices.

IDENTIFICATION OF KEY PERSONNEL (Cont'd)

In General Electric's Direct Energy Conversion Laboratory, he has conducted mass and heat transfer studies and authored a G. E. Technical Report No. 62DE-2 on Mass Transfer and Voltage Control in Fuel Cells, investigated new fuels and submitted two joint patent disclosures on borohydride and methanol liquid fuels, improved electrode structures, advanced ion exchange membranes for fuel cells (GPS Membranes), and developed a modified air-electrode current collector for use with I. E. M. fuel cells for which a patent disclosure will be submitted.

Mr. Dankese is a registered professional engineer in Massachusetts and a member of the Boston Chapter of the American Institute of Chemical Engineering.

Simeon J. Krumbein, Ph. D. - Physical Chemist - Electrochemistry

Dr. Krumbein received his Ph. D. in Physical Chemistry from New York University in October, 1961 and joined the Fuel Cell Laboratory of General Electric's Direct Energy Conversion Operation immediately thereafter. In addition to being a research assistant at New York University from 1957 to 1959, he had been working for the past two years under a National Science Foundation Fellowship. His doctoral thesis dealt with the effects of proteins on the electrometric behavior of simple inorganic ions.

Prior to this, he worked in the laboratory of the Technical Research Group (TRG), Inc., where he conducted analysis of the properties of materials.

Since coming to the Direct Energy Conversion Operation, Dr. Krumbein has been primarily concerned with fundamental electrochemical mechanisms as they exist in fuel cell applications.

Henri J.R. Maget, D.Sc. - Senior Chemical Engineer

Dr. Maget has a background combining organic chemistry and chemical engineering. He holds degrees: B.S. and Licence es Sciences from University of Strasbourg, France, 1950 where he majored in Chemistry; M.S. in Petroleum Engineering from Petroleum School, Paris, 1953; M.S. in Chemical Engineering from University of Michigan, 1956; and D.Sc. from University of Toulouse, France, 1959. He joined the Aircraft Accessory Turbine Department in August 1960.

7.0

IDENTIFICATION OF KEY PERSONNEL (Cont'd)

His background includes two years of research at the University of Michigan as an exchange scientist. Here he worked on reactions of epoxides with alcohols, solubility of CO₂ in organic solvents and thermodynamic properties of Freon 22. This was followed by two years of research work at Institut du Genie Chimique in France and more recently as research scientist at Yale University. Dr. Maget has been working in the field of thermodynamic properties of gases under pressures up to 10,000 atmospheres. His specialization leading to his doctorate was chemical kinetics and polymerization of epoxides.

Since he joined the General Electric Company, Dr. Maget has been working on physico-chemical properties of electrochemically active catalysts, and is investigating the relationship between these properties and the performance of fuel cells.

8.0

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