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6. AUTHOR(S) Dr. A. John Appleby Dr. S. Srinivasan			
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Center for Electrochemical Systems and Hydrogen Research Texas Engineering Experiment Station 238 WERC Building Texas A&M University College Station, Texas 77843-3577		8. PERFORMING ORGANIZATION REPORT NUMBER 6035	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Department of the Navy Office of the Chief of Naval Research 800 North Quincy Street Arlington, Virginia 22217-5000 Code 1513:LHO		10. SPONSORING/MONITORING AGENCY REPORT NUMBER	
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13. ABSTRACT (Maximum 200 words) The objectives of this project are to (1) investigate the physico-chemical properties of some solid polymer electrolytes and of electrode kinetic parameters of oxygen reduction at the Pt/solid polymer electrolyte interface; (2) optimize electrodes and membrane/electrode assemblies to attain the highest possible power density in fuel cells; and (3) model the performance of an electrochemical cell stack taking into consideration the electrochemical, mass transfer and heat transfer characteristics to demonstrate the feasibility of attaining the stated goals. Highlights of the accomplishments on these tasks are as follows: (i) using a microelectrode technique, the electrode kinetic parameters for oxygen reduction at the platinum-solid polymer electrolyte interface and the oxygen solubility and diffusion coefficient in this electrolyte were determined; (ii) with optimized electrode structures and membrane/electrode assemblies, high power densities ($>1\text{W}/\text{cm}^2$) were attained in solid polymer electrolyte fuel cells (single cells; and (iii) the modeling studies reveal that (a) thermal and water management can be effective with recirculation of gases with external condensation, and (b) with thin metallic bipolar plates (weight $0.5\text{ g}/\text{cm}^2$) and assuming a power density of $4\text{ W}/\text{cm}^2$ (which is possible with a thinner electrolyte membrane), a stack power density of $7\text{ kW}/\text{kg}$ can be achieved.			
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END-OF-THE-YEAR REPORT
1990

PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS/STUDENTS
REPORT

for

Grant N000-14-88-K-0389
R&T Code: B 41C002DAR 01
B 41C002DAR 02

A LIGHTWEIGHT SOLID POLYMER ELECTROLYTE
FUEL CELL WITH STACK POWER DENSITY OF
3kW/lb (7 kW/kg)



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PART 1

a. Papers Submitted to Refereed Journals (and not yet published):

A. Parthasarathy, C. R. Martin, S. Srinivasan, "Electrochemical Investigation of the Oxygen Reduction Reaction at the Pt/Solid Polymer Electrolyte Interface Using a Microelectrode"; submitted to the Journal of the Electrochemical Society.

b. Papers Published in Refereed Journals:

S. Srinivasan, D. J. Manko, H. Koch, M. A. Enayetullah and A. J. Appleby, "Recent Advances in Solid Polymer Electrolyte Fuel Cell Technology With Low Platinum Loading Electrodes", Journal of Power Sources, 29, 367 (1990).

c. Books (and sections thereof) Submitted for Publication: None

d. Books (and sections thereof) Published:

(i) A. J. Appleby and F.R. Foulkes, Fuel Cell Handbook, Van Nostrand Reinhold, New York (1989).

(ii) S. Srinivasan, S. Somasundaram, D. H. Swan, H. Koch, D.J. Manko, M. A. Enayetullah, H. Koch, and A.J. Appleby, "A Design Study of High Power Density Solid Polymer Electrolyte Fuel Cells", Proceedings of the Symposium on Fuel Cells, Annual Meeting of the American Institute of Chemical Engineers, Nov. 6 and 7, 1989, San Francisco, CA Proceedings Volume, 89-14, The Electrochemical Society, Pennington, NJ (1989).

e. Technical Reports Published and Papers Published in Non Refereed Journals:

S. Srinivasan, M.A. Enayetullah, S. Somasundaram, D. H. Swan, H. Koch, D.J. Manko, M.A. Enayetullah, H. Koch, and A.J. Appleby, "Recent Advances in Solid Polymer Electrolyte Fuel Cell Technology With Low Platinum Loading Electrode," IECEC Proceedings, 24, 1623 (1989).

f. Patents Filed: None

g. Patents Granted: None

h. Invited Presentations at Topical or Scientific/Technical Society Conference:

(i) S. Srinivasan, S. Somasundaram, D. H. Swan, H. Koch, D.J. Manko, H. Koch, and A.J. Appleby, "Recent Advances in Solid Polymer Electrolyte Fuel Cell

Technology With Low Platinum Loading Electrode," 24th Intersociety Energy Conversion Engineering Conference, August 6-11, 1989.

(ii) S. Srinivasan, M. A. Enayetullah, S. Somasundaram, D. H. Swan, D. J. Manko, H. Koch, and A. J. Appleby, "International Fuel Cell Workshop on Fuel Cell Technology Research and Development, Tokyo, Japan, September 16, 1989.

(iii) S. Srinivasan, M. A. Enayetullah and A. J. Appleby, "Global Perspectives of Fuel Cell Technology and Their Applications," S. Srinivasan, 40th International Society of Electrochemistry Meeting, Kyoto, Japan, September 17-22, 1989.

(iv) S. Srinivasan, S. Somasundaram, D. H. Swan, H. Koch, D. J. Manko, H. Koch, and A.J. Appleby, "A Design Study of High Power Density Solid Polymer Electrolyte Fuel Cells", Proceedings of the Symposium on Fuel Cells, Annual Meeting of the American Institute of Chemical Engineers, Nov. 6 and 7, 1989, San Francisco, CA Proceedings Volume, 89-14, The Electrochemical Society, Pennington, NJ (1989).

i. Contributed Presentations at Topic or Scientific/Technical Society Conferences:

(i) "Electrode Kinetics of Oxygen Reduction at the Platinum/Solid Polymer Electrolyte Interface: Studies Using Microelectrodes and Supported Electrocatalysts: A. Parthasarathy, C. R. Martin, M. A. Enayetullah, S. Srinivasan and A. J. Appleby. Paper presented at the American Chemical Society Meeting in Boston, MA, April 1990.

(ii) M. A. Enayetullah, D. J. Manko, H. Koch, I. Kakwan, D. Swan, S. Srinivasan, and A. J. Appleby, "Development of Electrodes for High Energy Efficiency and High Power Density Solid Polymer Electrolyte Fuel Cells and Their Performance Evaluation," 177th Meeting of the Electrochemical Society, Montreal, Canada, May 6-11, 1990.

j. Honors/Awards/Prizes: None

k. Number of Graduate Students Receiving Full or Partial Support on ONR Grant or Contract: One

l. Number of Postdoctoral Fellows Receiving Full or Partial Support on ONR Grant or Contract: One

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1990
(continuation)

PART II

- a. **Principal Investigators:** Dr. A. John Appleby (PI), Drs. S. Srinivasan, C.R. Martin and R. E. White (CoPIs)
Telephone (409) 845-8281
- b. **Cognizant ONR Scientific Officer:** Dr. Robert Nowak
- c. **Current telephone number:** (202) 696-4409
- d. **Brief description of the Project:**

The aim of the project was to attain high power densities in solid polymer electric fuel cells (single cells) with the goals of developing an electrochemical cell stack with a power density of 7kW/kg. In order to achieve this goal, the objectives of the research were:

- (i) Investigation of the physiochemical properties of various solid polymer electrolytes and of electrode kinetics at Pt/solid polymer electrolyte interfaces to attain maximum fuel cell performance;
- (ii) Optimization of low and high Pt loading gas diffusion electrode structures, membrane thickness, and of electrode-membrane bonding to obtain the most effective catalyst-membrane interface;
- (iii) Mathematical modeling of electrochemical, mass transfer and heat transfer characteristics during cell operation to optimize methods and operating conditions to attain the stated power density.

The work carried out during the two-year period (June 1988 to May 31, 1990) consisted of, of three tasks:

- (i) Identifying the most suitable solid polymer electrolyte and optimizing the membrane thickness so as to minimize cell resistance and hence maximize power density,
- (ii) Optimizing solid polymer-catalyst bonding to obtain the most effective catalyst-electrolyte interface and hence maximizing catalyst utilization, and
- (iii) Mathematical modeling of heat and mass transfer so as to optimize methods and operating conditions to attain the goal of 7kW/kg for the power density of the electrochemical cell stack.

e. Significant Results During Last Year:

Attainment of high power densities has been demonstrated in single cells (5 cm^2) with low (0.40 mg/cm^2) platinum loading electrodes. These electrodes (Prototech, 20%Pt/C) were modified by (a) impregnation of proton conductor, Nafion into the electrode and (b) localization of a thin layer of Pt (0.05 mg/cm^2) on the active surface of the electrode. Chemical and electrochemical deposition have been found to be viable alternate methods to sputtering for platinum localization. The role of type as well as of thickness of the polymer membrane on fuel cell performance has been established; the thinner the membrane, the better is the performance. The best performance was obtained in single cells with Pt sputtered Prototech electrodes and Dow polymer membrane (thickness: $125 \text{ }\mu\text{m}$) for the electrolyte layer, i.e., 1.4 W/cm^2 (2 amps at 0.6 volts). In a cell with a high platinum loading (10 mg/cm^2) from Electrochem, Inc., and with the Dow membrane, the cell potential was 0.7 V at 2 A/cm^2 . Further, this cell showed a high energy efficiency at low current densities (0.92 V at 200 mA/cm^2). The peak power density was approximately 2 W/cm^2 in this cell. A preliminary thermal and mass analysis indicates that cell cooling can be accomplished by high pressure recirculation of reactant gases with external water condensing and gas cooling.

f. Brief Summary of Plans for Next Year's Work:

This project terminated on May 31, 1990. The Final Report will be provided prior to June 30, 1990.

g. List of Names of Graduate Students and Postdoctoral(s) Currently Working on the Project:

- 1) Mr. Arvind Parthasarathy (Ph.D. Graduate student in Chemistry)
- 2) Mr. Herman Koch (Exchange Student from Ruhr - Universität Bochum, Master's Degree in Mechanical Engineering)
- 3) Dr. Mohammad A. Enayetullah (Postdoctoral Research Associate)
- 4) Dr. David H. Swan (Research Scientist)

h. Technical Reports Submitted to ONR During the Past Year:

Three (3) Quarterly Reports submitted to DARPA/ONR
One(1) End of Year Report

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END-OF-THE-YEAR REPORT

(continuation)

PART III

- a. An introductory "viewgraph": (See viewgraph 1)
- b. A figure: Potential and Power Density vs. Current Density in High Performance Single Cells (See viewgraph 2). The figure represents the best results obtained to date.
- c. A concluding "viewgraph": (See viewgraph 3)
- d. A paragraph of explanatory text:
A Lightweight Solid Polymer Electrolyte Fuel Cell With Stack Power Density at 7kW/kg.
End of Year Report 1990

The mass transport parameters diffusion coefficient and solubility of oxygen in the solid polymer electrolyte (Nafion) were obtained using a potentiostatic transient technique at a microelectrode. Electrode kinetic parameters such as the Tafel slope and the exchange current density were obtained by analyzing the activation controlled region of the slow-sweep data (inverses E) for the electrochemical reduction of oxygen at the Pt micro-electrode/Nafion interface. The conditions under which the experiments were carried out emulate those in the solid polymer electrolyte fuel cell. With the methodology established for Nafion, similar investigations are underway for the Dow polymers. The role of ambient humidity, pressured temperate on the electrodes kinetics are also in progress.

A vacuum technique is being explored to substitute for the current brushing technique used to impregnate Nafion solution into the electrode prior to hot-pressing. High power densities (1-2 to 1.4 W/cm²) have been obtained in single cells with low and high platinum loading electrodes. The conditions for hot-pressing of the electrodes to the membrane were varied to determine the optimal time, pressure. The catalyst utilization was checked by measuring the electrochemically active area using cyclic voltammetry.

The performance goal of 7 kW per kg of fuel cell stack weight provides extraordinary electrochemical and engineering goals. The present estimate based on extrapolation of experimental data is that a high performance electrode-membrane assembly, provided with adequate fuel and ideal thermal and water conditions, will be capable of achieving a specific cell power of $4\text{W}/\text{cm}^2$. Thus, the goal of $7\text{kW}/\text{kg}$ requires a specific cell weight of $0.6\text{ g}/\text{cm}^2$. The components of a fuel cell stack are the membrane-electrode assembly (ME), and the bipolar plate. The present estimate for the specific weight of a high performance ME is $0.10\text{ g}/\text{cm}^2$. This estimate is based on a $75\text{ }\mu\text{m}$ thick membrane and a platinum loading of $10\text{ mg}/\text{cm}^2$ in the electrode. The bipolar plate assembly is a lightweight design ($0.5\text{ g}/\text{cm}^2$) that uses impingement cooling and re-circulated oxygen for thermal and water management. A promising material for the bipolar plate is titanium with a gold coating.

A LIGHTWEIGHT SOLID POLYMER ELECTROLYTE FUEL CELL WITH

STACK POWER DENSITY OF 3 kW/lb (7 kW/kg)

TEXAS A&M UNIVERSITY SYSTEM

A. J. APPELBY, S. SRINIVASAN, E. A. TICIANELLI, M.A. ENAYETULLAH, D.J. MANKO,
H. KOCH, D.H. SWAN, A. PARTHASARATHY, C. R. MARTIN, AND R. E. WHITE

OBJECTIVES

- INVESTIGATIONS OF PHYSICO-CHEMICAL PROPERTIES OF VARIOUS SOLID POLYMER ELECTROLYTES AND ELECTRODE KINETICS AT PT/SOLID POLYMER ELECTROLYTE INTERFACES.

- OPTIMIZATION OF LOW AND HIGH PT-LOADING GAS DIFFUSION ELECTRODE CONDITIONS, STRUCTURES, MEMBRANE BONDING TO OBTAIN MOST EFFECTIVE ELECTROCATALYST-MEMBRANE INTERFACE AND HENCE ATTAIN MAXIMUM FUEL CELL PERFORMANCE.

- MATHEMATICAL MODELING OF ELECTROCHEMICAL, MASS TRANSFER AND HEAT GENERATION CHARACTERISTICS ASSOCIATED WITH CELL OPERATION TO OPTIMIZE FUEL CELL THERMAL AND WATER MANAGEMENT.

APPROACHES

- DEVELOP MICRO-ELECTRODE TECHNIQUE TO INVESTIGATE KINETICS AT PT/SOLID POLYMER ELECTROLYTE INTERFACES AND DETERMINE MASS TRANSPORT PARAMETERS OF REACTANTS.

- DEVELOP METHODS, ALTERNATIVE TO SPUTTER DEPOSITION, TO LOCALIZE PLATINUM NEAR FRONT SURFACE OF ELECTRODE.

- OPTIMIZE (1) AMOUNT OF PROTON CONDUCTOR IMPREGNATION INTO POROUS GAS DIFFUSION ELECTRODES AND (2) HOT PRESSING OF ELECTRODES TO MEMBRANES.

- REDUCE OHMIC OVERPOTENTIAL IN CELL BY USING THINNER NAFION® DOW MEMBRANES: THE LATTER HAS BETTER WATER RETENTION CHARACTERISTICS.

- MODELING TO OPTIMIZE METHODS AND OPERATING CONDITIONS FOR ATTAINING HIGH POWER DENSITY, WITH AIM OF PRODUCING STACK POWER DENSITY OF 7 KW/KG.

A LIGHTWEIGHT SOLID POLYMER ELECTROLYTE FUEL CELL WITH

STACK POWER DENSITY OF 3 kW/lb. (7 kW/kg)

TEXAS A&M UNIVERSITY SYSTEM

A. J. APPLEBY, S. SRINIVASAN, E. A. TICIANELLI, M. A. ENAYETULLAH, D. J. MANKO,
H. KOCH, D. H. SWAN, A. PARTHASARATHY, C. R. MARTIN, AND R. E. WHITE

ACCOMPLISHMENTS TO DATE

- NOVEL MICROELECTRODE TECHNIQUE DEVELOPED AND USED TO DETERMINE TAFEL PARAMETERS AT Pt/ SOLID POLYMER ELECTROLYTE INTERFACE AND MASS TRANSPORT PARAMETERS OF O₂ IN MEMBRANE.
- HIGH POWER DENSITY OF 1.4 W/CM² (0.7 V AT 2A/CM²) ATTAINED WITH HIGH PLATINUM LOADING ELECTRODES (10 MG/CM²) AND DOW MEMBRANE. RESULTS INDICATE THAT BY USE OF MEMBRANE 75 μM THICK, POWER DENSITY OF 4 W/CM² CAN BE ACHIEVED.
- MODELING STUDIES OF (I) THERMAL AND WATER MANAGEMENT REVEAL THAT RECIRCULATION OF GASES WITH EXTERNAL CONDENSATION PROVIDES BEST BIPOLAR DESIGN AND (II) ASSUMING A POWER DENSITY OF 4 W/CM², WEIGHT OF THIN METALLIC BIPOLAR PLATE WILL HAVE TO BE 0.5 G/CM² IN ORDER TO ACHIEVE STACK POWER DENSITY OF 7 KW/KG.

